

VOLUME 28 / September 2024

detritus

Multidisciplinary Journal for Circular Economy and
Sustainable Management of Residues

Editor in Chief:
RAFFAELLO COSSU

detritusjournal.com

an official journal of:

iwwg
international waste working group

CISA



ISSN 2611-4135 / ISBN 9788862650441

DETRITUS - Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

Detritus is indexed in:

- Emerging Sources Citation Index (ESCI), Clarivate Analytics, Web of Science
- Scopus, Elsevier
- Google Scholar

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The journal contents are available on the official website: www.detritusjournal.com

Legal head office: Cisa Publisher - Eurowaste Srl, Via Beato Pellegrino 23, 35137 Padova - Italy / www.cisapublisher.com

Graphics and layout: Elena Cossu, Anna Artuso - Studio Arcoplan, Padova / studio@arcoplan.it

Printed by Cleup, Padova, Italy

For subscription to printed version, advertising or other commercial opportunities please contact the Administration Office at administration@eurowaste.it

Papers should be submitted online at <https://mc04.manuscriptcentral.com/detritusjournal>

Instructions to authors may be found at <https://detritusjournal.com/guide-for-authors/>

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Registered at the Court of Padova on March 13, 2018 with No. 2457

www.detritusjournal.com

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Detritus - Multidisciplinary Journal for Waste Resources and Residues - is aimed at extending the “waste” concept by opening up the field to other waste-related disciplines (e.g. earth science, applied microbiology, environmental science, architecture, art, law, etc.) welcoming strategic, review and opinion papers. **Detritus is indexed in Emerging Sources Citation Index (ESCI) Web of Science, Scopus, Elsevier and Google Scholar.** Detritus is an official journal of IWWG (International Waste Working Group), a non-profit organisation established in 2002 to serve as a forum for the scientific and professional community and to respond to a need for the international promotion and dissemination of new developments in the waste management industry.

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Editorial

THE NEED FOR SINKS DURING THE TRANSITION TO A GLOBAL CIRCULAR ECONOMY

In the management of solid waste the deposit on soil has historically played a fundamental role.

This role originates directly from the life cycle of (degradable) products that obey the natural principle of mass balance (in a closed system the mass at any point will be the same) and Lavoisier's Law of Conservation of Mass: (material cannot be lost - it can only be transformed). A general representation of the life cycle is given in Figure 1.

Material resources (renewable or non-renewable, e.g. metals, stones, timber) are withdrawn from a natural pool above the soil or in the subsoil to feed production of goods. Elements and compounds in the material resources are naturally present in a non-mobile form (x) that can be mobilized after the extraction and processing steps necessary to obtain the primary materials for Production. At the end of their Use-life, products will become waste and will enter a Management step (deposit, collection, sorting, transport, preparation for reuse and recycling, etc. – not necessarily in this order).

Non-reused and unrecycled wastes represent residues that need to be disposed of, through Treatment and sometimes, via a Back to Earth deposit.

Along all these steps elements and compounds after extraction are generally in a mobile form(s).

During the different steps, the mobile elements can be emitted (dS/dt) either legally (emissions respecting limits set by national regulations) or illegally and might give rise to diffused or concentrated unsustainable contamination (pollution).

The general scheme in Figure 1 can represent all the different approaches and strategies developed in different countries over the years. Some steps might be excluded, technological levels might differ significantly, different tools for contamination control might be adopted, some steps (e.g. extraction, waste management, final waste disposal) might be performed in countries different from the ones where goods are produced or used, etc.

The management of all the above influences the sustainability of the system in terms of control of depletion of non-renewable resources, quality of the environment and optimization of the protection of public and ecological health.

We can analyze the development of the scheme under different scenarios:

- Low and middle income countries
- Industrialized countries with a linear approach to municipal waste management
- Actual Circular Economy approach
- Sustainable Circular Economy strategy

Developing countries

In many low and middle income countries, extraction of material resources might be performed without observing the highest technological and environmental protection standards. The most important materials are exported for production in industrialized countries. Waste management is frequently seen as a low priority, largely due to lack of

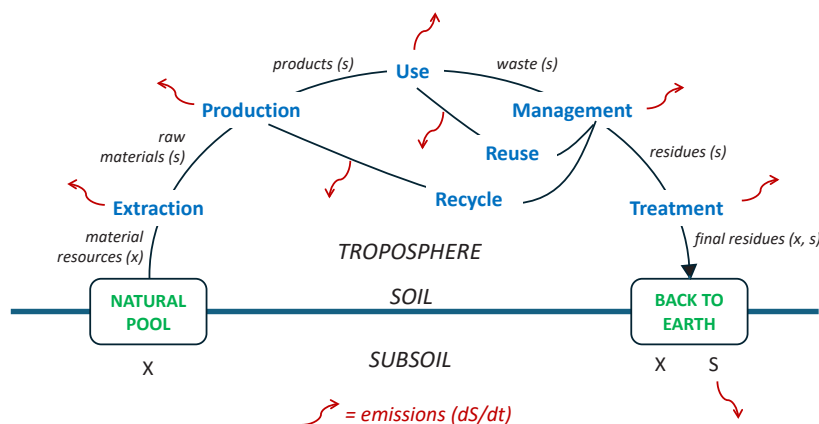


FIGURE 1: Graphical representation of the Life cycle of products and waste. X= non-mobile form of contaminants, S = mobile form of contaminants, dS/dt = emissions of contaminants.

financial resource, but it can also be “invisible” (Mukhtar et al, 2018) and/or there can be poor environmental awareness/unwillingness to adopt new technologies amongst politicians and the public (Cossu and Williams, 2015). Waste collection is not implemented consistently or might be carried out in an inappropriate manner. Collected waste is often disposed of in open dumps or poor landfills with no or low emission control. Generally, waste is not adequately compacted, so littering occurs attracting insects, birds, and mammals, increasing risk for disease transmission.

The collection of reusable and recyclable materials is often practiced by the informal sector in communities and on dumpsites. In many cases, these informal systems can be quite effective and economically feasible. However, scavengers working on dumps might subject themselves to injury from falls, fires, toxic fumes, and dangerous waste components.

Diffusion of contaminants might originate severe environmental impacts and risks for health.

Industrialized countries with MW linear approach

The linear approach (“Take-Make-Waste”) prevailed in industrialized countries during the intense industrial development between the 1960s to the 1990s (Cossu and Stegmann, 2018). Material resources were extracted, products generated and used, and wastes were directly disposed of in landfills, without too much care in controlling emissions. In some countries, particularly in megacities, incineration was used, no source segregation and separate collection was generally adopted. Examples of material recycling by manual or mechanical sorting were rarely documented.

Since the 1960s, the use of uncontrolled dumping was gradually replaced by controlled tipping, encoded by the World Health Organization (WHO) in a series of guidelines that represented, over a considerable period, the main technical references in the field (Cossu, 2010). Landfill dominated waste management; for example, although the European Union’s Waste Framework Directive (Council Directive 75/442/EEC) introduced the waste hierarchy into European waste policy in 1975, it had little impact until the European Parliament announced a new version to its legislation (Directive 2008/98/EC), which Member States had to introduce into national laws (Williams, 2015).

Landfills were sited in areas of low permeability where wastes were deposited in thin, uncompacted layers aimed at enhancing the establishment of aerobic conditions. The wastes were subsequently covered with inert materials (preferably clay) to avoid contact between wastes and animals (dogs, birds, rodents, insects, etc.).

Collection of leachate and biogas was not provided for; leachate was allowed to infiltrate into the ground (principle of “dilute and disperse”), whilst biogas production was not contemplated.

The repeated occurrence of noxious situations (odours, leachate ponding around the landfill sites, etc.) and an increased public awareness of environmental issues, jointly with technical and scientific progress in the field, has led since the 1980s to the increasing use of “Contained landfill”, based on adoption of physical barriers for controlling emissions. Combined artificial lining systems (clay + syn-

thetic membrane), leachate drainage and collection, heavy waste compaction, anaerobic processing, biogas collection and final capping were the main technical features. The collected leachate was treated with increasing process complexity (from lagoons to sophisticated biological technologies, reverse osmosis, activated carbon absorption, chemical precipitation and oxidation, evaporation, phyto-reduction, etc.)

Biogas was collected and converted to energy with different technologies (production of thermal and electric power, upgrading for domestic use, vehicle fuel, etc.).

Landfilling was driven by technical regulations (Butti et al., 2018) in the different industrialized areas of the world: in US the “Resource Conservation and Recovery Act” (RCRA) of 1976 which demanded to the Environmental Protection Agency (EPA) the power to enact specific implementing regulations, in Europe Union the framework Union Directive, 1999/31/EC which was implemented in the different member countries, in Japan the Ordinance No. 1/1977 of the Prime Minister’s Office and Ministry of Health and Welfare.

Throughout the years, the “Contained landfill” displayed its numerous limitations. In particular, the potential efficiency of new lining materials has led to an excessive use of them resulting in siting landfill even in vulnerable areas without considering the limited life span of the physical barriers; life spans are largely inferior to the long-term emissions potential of contaminants.

Additionally, the high management costs associated with leachate treatment promoted the tendency to minimize leachate production, adopting low permeable top cover. The consequent reduction of water availability for the attenuation processes (biological stabilization, contaminants flushing, etc.), jointly with several other technical factors (i.e. clogging of drainage systems) resulted in prolonging the long-term negative impacts of landfills (Grosule and Stegmann, 2020).

These kind of landfills are inherently unsustainable for several scientific reasons further to the short- and long-term contamination of water and soil: climate change contribution due to the generation of greenhouse gases during the anaerobic degradation process (methane, CO₂); land space consumption; air quality impact; loss of biodiversity; health risks; waste of material resources; landscape degradation; need for costly long-term monitoring and maintenance; wildlife disruption in term of habitats; displacement of, and behaviour alteration by, animals; etc.

Actual Circular Economy approach

The global economic growth and the increasing need for primary raw materials, jointly with the awareness for environmental protection and resources depletion, have led modern societies to shift attention from the limited and fixed stocks of raw materials to the anthropogenic stocks of materials. This created the base for the development of circular economy as a strategy for recovering of resources from waste (Curran and Williams, 2012). Since the late 1990s and beginning of the 2000s, policies worldwide started to limit the massive landfilling by prioritizing waste management schemes based on waste minimization, reuse, recycle and energy production (“Pollution prevention

act” of 1990 in US; “The basic act for establishing a sound material-cycle society” -Act n. 110 of 2000 in Japan; “Waste Directive” 2008/98/EC in the European Union).

Further to the traditional fractions of MSW normally considered in source segregation programs (plastics, paper, cardboard, glass containers, cans, putrescibles , etc.), other waste streams have been increasingly considered for recycling, such as waste from electrical and electronic equipment, end-of-life vehicles, scrapped tires, construction and demolition waste, combustion residues, food waste, batteries, road sweeping waste, water treatment sludges, exhausted oils, old landfilled waste, residues from food industries, slags and other industrial waste.

These materials can alternatively be used to obtain different kinds of products, such as secondary raw materials, building materials, fuel and biofuel, composites, fertilizers, etc. and regulations are continuously implemented for qualifying the passage from waste to product (“End of Waste”).

So far, the Circular Economy concept is expanding successfully in several other areas of our society (i.e. sustainable architecture and recovery of buildings), but in routine waste management some issues are becoming increasingly critical and problematic:

- Modern products are continuously introduced in the market without caring about the environmental impacts (related to quality and quantity) when they will become waste (e.g. smart phones, composite plastics);
- Minimization is detrimentally influenced by a continuous production of goods with planned obsolescence (electronics, appliances, fast fashion garments, children's toys, etc.);
- Products that are introduced daily into the market contain a wide range of substances and additives (particularly non-degradable organics, nanoparticles, metals) which could present problems either in terms of impairing the recycling process (e.g. elastane and electronic components in textiles) via accumulation of contaminants in the recycled products and related release during use (e.g. microplastics, various xenobiotics) and by concentration in the residues to be disposed of;
- Even after recycling a considerable amount of waste (unrecycled, residues from different recycling loops, recycling process, etc.) remains, and are disposed by landfill or incineration;
- Landfilling of untreated waste, particularly in Europe, has been banned without a clear communication towards the citizen and visions about the need of closing the material balance in waste management where deposit on soil might still be necessary;
- Some phases of the circular economy are carried out in countries where environmentally sustainable standards in extraction of resources, manufacturing processes, recycling and waste management/disposal are not observed.

Landfilling for residual waste still maintains the main technical characteristics of the “contained landfill”, despite regulations that have occasionally been updated to reduce the volumes of waste conferred, to prevent waste of re-

sources, to minimize the production of greenhouse gases and, in general, to lower environmental impacts and risks. Moreover, most industrialized countries have introduced criteria for the monitoring of landfills during the post-closure phase, proposing the principle of financial provision and responsibility for landfill operators if the landfill continues to constitute a risk for the environment.

However, the evolution of the contained landfill into a residual landfill did not develop at the same pace as the need to respect environmental sustainability, aimed to avoid leaving the future generations to manage unacceptable burdens.

Although present measures applied to control long-term impact (post-closure care, environmental long-term operator responsibility) are adopted by regulations, the appropriate tools for their implementation are still lacking. Indeed, termination of post-closure care is defined according to time rather than environmental performance and no criteria have been established to define acceptable conditions on which to assess operator responsibility. Furthermore, as mentioned previously, residual landfills are largely based on the technology applied for contained landfills, thus featuring the same negative characteristics ranging from the deterioration of lining and leachate drainage systems to the adoption of leachate minimization measures and related negative consequences.

Lastly, a landfill for residual waste might require greater technical and environmental care than traditional landfills due to different analytical composition and mechanical properties of the materials to be buried: contaminants mobility could result enhanced and mechanical stability could be impaired.

Global world with a sustainable approach

The critical issues present in Circular Economy implementation can be substantially summarized in the increasing production of fast-moving goods and in the poor control of the mass balance which drives material flows (i.e. quality of goods, circulation and diffusion of contaminants, sustainable management of residues).

Possible solutions could be the following:

- Manufacturer responsibility for the products should be extended and more severely enforced to control quality and quantity of waste.
- Symbiosis between producers and recyclers should be stronger and more systematic to increase recycling capability and to control the removal of detrimental substances and contaminants along the recycling processes (e.g. ban use of non-recyclable plastics in short-lifetime, consumable products);
- Residual wastes should be managed with the view to provide a safe and permanent sink for contaminants, closing material loops.

The sinks should be designed to convert mobile contaminants into stable substances that do not pose harm for the environment and the health of people and animals. Sinks can be represented by treatment technologies, based on thermal, biological, chemical, or physical processes

(e.g. incineration, composting, inertness, washing, etc.) and by natural or anthropogenic “back to earth” recipients, where substances should be in a non-mobile form, in an equilibrium with the environment.

In this regard, any sort of the landfill model which has been experienced so far (uncontrolled, contained, residual, etc.) cannot effectively act as a sink.

While landfills might accumulate some organic carbon, they cannot be regarded as an anthropogenic sink due to incomplete decomposition and the natural carbon storage capacity of ecosystems like peatlands or forests are much more efficient. Biological stabilization such as Anaerobic digestion and Composting would represent a more efficient way to sequester organic carbon in the agricultural soil.

However, landfills may act sinks for inert and inorganic carbon which includes materials like non-recyclable plastics, glass, treated wood, and minerals/aggregates. Unlike organic carbon, which decomposes into gases, inert carbon remains relatively stable over time. Some inorganic materials, such as glass and ceramics, have a long lifespan and do not readily break down. When these materials are disposed of in landfills, they effectively store carbon for extended periods.

Inert plastics, although technically organic, degrade very slowly in landfills due to their chemical composition. Plastic items, such as bottles and packaging, can remain largely intact for decades or even centuries. During this time, they effectively trap carbon.

Inorganic contaminants in landfills particularly under anaerobic conditions (e.g. ammonia, heavy metals) can leach for centuries. This is the same for persistent organics, such as halogenated solvents.

Something like a Back to Earth Sink (BES) has been proposed in the past with the term of “Sustainable Landfill” (i.e. Grossule, 2020), which, particularly for the population might be considered as an oxymoron due the negative experiences associated with landfilling thus far. It is similar to designing a modern house respecting all the environmental requirements, and to present it with the term of “sustainable hovel”. To move toward the implementation of BES, it would be necessary to ban not only landfilling, but also the term itself and the related existing regulations. New regulations should be urgently agreed and issued. In addition, an extensive communication campaign would be of paramount relevance as people everywhere are against landfilling and widely convinced that Circular Economy is equivalent to “Zero Waste”.

Conclusions

The use of an acceptable “Back to Earth” (BES) sink is crucial during the global transition to a circular economy. Firstly, a BES would minimize environmental impacts by reducing greenhouse gas emissions and leachate production, which can contaminate soil and water. Secondly, a BES can provide a safe disposal method for non-recyclable and hazardous waste, ensuring public health and safety. Thirdly, the use of BES would give countries time to plan and resource their move towards circular economy practices by promoting waste avoidance, reuse, segregation, recycling, and composting whilst still recognizing the practical realities involved in this process. We suggest that Back to Earth Sinks are going to be a vital component in the development a circular economy, and that their implementation is urgent.

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COLLABORATIVE PLATFORMS SUPPORTING CIRCULARITY IN THE BUILT ENVIRONMENT. IDENTIFYING STRATEGIC ACTIONS AND REPLICABLE GOOD PRACTICES

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Article Info:

Received:

31 May 2024

Revised:

30 July 2024

Accepted:

6 August 2024

Available online:

2 September 2024

Keywords:

Circularity

Construction sector

Building materials

Collaborative platforms

Stakeholder engagement

ABSTRACT

This contribution highlights the role that collaborative platforms can offer for the promotion of circularity in the construction sector, through the interaction between different stakeholders and, in particular, through the collection and promotion of good practices. In fact, the work reports the outcomes of stakeholder consultation experiences, developed within the Italian Circular Economy Stakeholder Platform, aimed at identifying the strategic areas and priority actions for the scale-up circular solutions to a large scale, and activities of collection, selection and analysis of good practices at different scales (products, buildings, tools) that exemplify the possibility of concretely implementing - in a highly replicable manner - the circular solutions, until now often considered experimental cases that are difficult to be repeated.

1. INTRODUCTION

The construction industry involves the production, maintenance and end-of-life management of a wide variety of infrastructure and buildings. From an economic point of view, the gross domestic product (GDP) of the construction industry enhances the growth of nations. Specifically, Suwal et al. (2019) stated that the construction industry contributes 6% to GDP globally. Although the construction industry positively impacts GDP, its activities have negative consequences for the environment, society, and the economy. Recent estimates reveal that the building and construction sector consumes about 40% of materials and produces roughly 35% of waste (Agyekum et al., 2024). Moreover, construction is responsible for 36% of energy consumption and 37% of energy-related CO₂ emissions (UNEP, 2021).

Therefore, the construction industry is highly materials-intensive and significantly contributes to global solid waste generation. Furthermore, although policies have been developed at different levels to improve the sustainability of the sector, they do not focus entirely on a circular economy (Adams et al., 2017), but rather prioritize strategies to reduce operational energy consumption. Recently, much of the thinking on the circular economy has focused on short- and medium-term consumer products (Adams et al., 2017).

In fact, research and large-scale practical applications of circular economy in the built environment are still limited, both at the component/material level and at the building level (Adams et al., 2017). Moreover, a holistic perspective is necessary to assess the construction sector more broadly, ensuring genuine positive impacts in terms of reduced resource extraction and related climate-changing emissions (Baiani and Altamura, 2021).

The aforementioned data have prompted the construction industry to pursue changes in its practices and redefine concepts such as Circular Construction to facilitate the transition to a circular economy. Circular Construction involves applying circular economy principles to the built environment with a systemic vision that extends beyond 'green buildings,' which focus on technology and environmental concerns, or 'sustainable buildings,' which also address economic and social dimensions. Circular buildings prioritize the support and behavior of stakeholders - e.g. government, customers, suppliers, builders (Pomponi and Moncaster, 2017). These buildings are designed to embrace circular principles and close resource cycles at different spatial and temporal levels. This reflects the complexity and dynamic nature of the circular economy in construction, requiring consideration and integration of technical, social, economic, environmental, behavioral,



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Detritus / Volume 28 - 2024 / pages 5-18

<https://doi.org/10.31025/2611-4135/2024.19404>

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spatial, and temporal aspects (Papastamoulis et al., 2021; Yu et al., 2022).

Thus, circular design and construction processes in the built environment necessitate a significant change in the approach of all actors involved and their supply chains. This process involves not only designers but also principals, contractors, and manufacturers. Additionally, there is an increasing need to train new professionals capable of implementing innovative circular business models, such as specialists in deconstruction and the sale of salvaged components (e.g., Restado, Rotor DC, Cyrkl).

In this process of change, multistakeholder platforms can play a key role by fostering the involvement and collaboration of different actors who, based on shared values, are willing to work together to enhance their knowledge and skills systems for collective progression. Consequently, the traditional triad known as the triple helix model of innovation – formed by university, industry, and government – has evolved. Over time, this model has been reinforced with new models of knowledge generation, incorporating media, shared culture, civil society, and the environment, thus evolving into the quintuple helix model of innovation (Carayannis & Campbell, 2010).

By adopting the theoretical framework proposed by the quintuple helix model, this paper demonstrates how collaborative platforms can foster cultural exchanges to fully contribute to resource efficiency and climate neutrality goals. These platforms can also serve as repositories of best practices that promote the scalability and replicability of innovations in the market.

2. INTERNATIONAL COLLABORATIVE PLATFORMS FOR CIRCULARITY IN THE CONSTRUCTION SECTOR

This section describes a selection of existing online collaborative platforms that can be traced back to the quintuple helix model by having a multiplicity of stakeholders belonging to the different domains as well as being ‘archives’ of good practices related to the built environment. Among the different existing platforms, the following is a non-exhaustive list involving European, national as well as cross-border platforms: the European Circular Economy Stakeholder Platform (ECESP) and the corresponding Italian national platform (ICESP); the Holland Circular Hotspot; the Circular Taiwan Network. These platforms have been selected in the research because they showcase good practices which are considered virtuous examples of innovation (at the product, process, but also organizational level) that can favour the transition towards a circular construction sector. The interdisciplinary approach of the selected platforms promotes and facilitates the mapping and exchange of good practices.

2.1 European Circular Economy Stakeholder Platform (ECESP)

The European Circular Economy Stakeholder Platform (ECESP) is a joint initiative of the European Commission and the European Economic and Social Committee that supports civil society organisations and public authorities

accelerating the transition to a circular economy in Europe by promoting dialogue, knowledge sharing and exchange of best practices. As stated on its website (<https://circulareconomy.europa.eu/platform/en>) as a ‘network of networks’, ECESP provides a ‘place’ for stakeholders to come together to share and scale effective solutions and address specific challenges. The platform bridges existing local, regional and national initiatives and supports the implementation of the circular economy in multiple sectors. Stakeholder engagement is encouraged through thematic working groups (so-called Leadership Groups). In particular, the Leadership Group on the built environment has identified two main priorities: to support the integration of circularity aspects into existing (or upcoming) legislation; and to foster the standardisation of materials by promoting the introduction of digital passports for materials across the EU. The work of the leadership group and the ECESP platform database resulted in a first compilation of good practices related to the built environment (ECESP, 2021a).

2.2 Italian Circular Economy Stakeholder Platform (ICESP)

The Italian Circular Economy Stakeholder Platform (ICESP) is the Italian national mirror platform of ECESP. ICESP was created as a forum that brings together the initiatives, experiences, critical issues and prospects that Italy represents and has the potential to represent in Europe in a circular economy, promoting circularity also with specific actions. As declared in its website (<https://www.icesp.it/>), ICESP aims at: promoting the dissemination of knowledge; fostering dialogue and synergic relations among Italian actors; mapping Italian good practices; overcoming the fragmentation of initiatives at the national level; creating a permanent operational tool that can stimulate and facilitate cross-sectoral and inter-sectoral dialogue and interactions; promoting Italian excellence and ‘the Italian way of doing circular economy’, starting from traditional characteristics and related cultural, social and entrepreneurial models. The platform is open to all players in the circular economy sector (public administrations, firms, research organisations and civil society) and adopts an inclusive approach. Among the different working groups, the one on ‘Sustainable and Circular Value Chains’ includes a specific sub-group on ‘Construction and Demolition’, with the aim of developing position papers, collecting good practices in the market and organising webinars/seminars on specific topics related to the built environment. The work of the subgroup resulted in a first position paper that includes priorities to foster cycle closure in the built environment and an initial list of good practices related to the sector (Altamura et al., 2020).

2.3 Holland Circular Hotspot

The Holland Circular Hotspot is a private organization dedicated to expediting the global shift toward a circular economy. It achieves this by fostering connections among companies, knowledge institutes, and local authorities, facilitating international collaboration, and promoting knowledge exchange on the Dutch circular economy. It collabo-

rates with different partners on topics as diverse as textiles, plastics, circular cities, water and construction. In particular, the construction group explores how circular economy concepts can help address challenges in the sector, supporting the transition to a more sustainable construction sector. The work of the Holland Circular Hotspot resulted in a compilation of good practices related to the built environment in the Netherlands (Holland Circular Hotspot, 2022).

2.4 Circular Taiwan Network

The Circular Taiwan Network (CTN) is a non-profit organization dedicated to advancing the circular economy in Taiwan. Since its establishment in 2015, CTN has been actively communicating, supporting, and facilitating coordination among all sectors of society—including government, industries, universities, research institutes, civil society organizations, and media—to promote the transformation of Taiwanese industries and society. The platform's overarching vision is to integrate the principles of the circular economy into Taiwan's daily life and position Taiwan as a global leader in the circular economy movement. To achieve this, CTN engages institutions, industries, universities, research institutes, civil society organizations, media, and international entities through its “CoPartners Circular Partner Platform”. The CTN also collects good practices that are organised by theme: Agribusiness and biomass, plastics, textiles, construction, transport, electronics and chemicals. More in detail, six areas of improvement have been identified to foster the transition to the circular economy of the construction sector: Extension of the life cycle of existing buildings together with energy efficiency; Planning for the recovery and reuse of new buildings; Establishment of an appropriate evaluation system for circularity in construction; Promotion of shared building materials through

rental services; Improvement of materials through collaborative partnerships; Quantification and definition of targets for the management of the circularity of total resources in the construction industry. The work of the CTN resulted in a collection of local good practices related to the built environment (Circular Taiwan Network, 2024).

3. RESEARCH AIMS AND METHODOLOGY

The research originated within the activities of the ICESP Construction and Demolition sub Working Group (see Section 2.2) and was subsequently developed through the methodology described below. The research started from the Priorities for a circular recovery after the COVID Pandemic identified by the whole ICESP platform in 2020 in a dedicated document (ICESP, 2020). These priority areas of action were connected by the stakeholders of the different Working Groups to the three reference systems: Context (with relative priorities identified as Governance; Education, information and culture; Infrastructural system), Tools (with relative priorities identified as Economic tools; Regulatory tools; Assessment tools) and Actions (with relative priorities identified as Eco-design and circular consumption models; Market for by-products, recycled and recovered materials; Integrated planning and circular urban and territorial management).

Based on this framework, the research aimed to identify the primary strategic actions for advancing circularity within the national construction sector by consulting stakeholders participating in the ICESP Construction and Demolition sub Working Group. Figure 1 shows the results of this activity: the stakeholders in the Group were asked to reflect on the general priorities listed in the above mentioned document, shared by all the stakeholders of the ICESP plat-

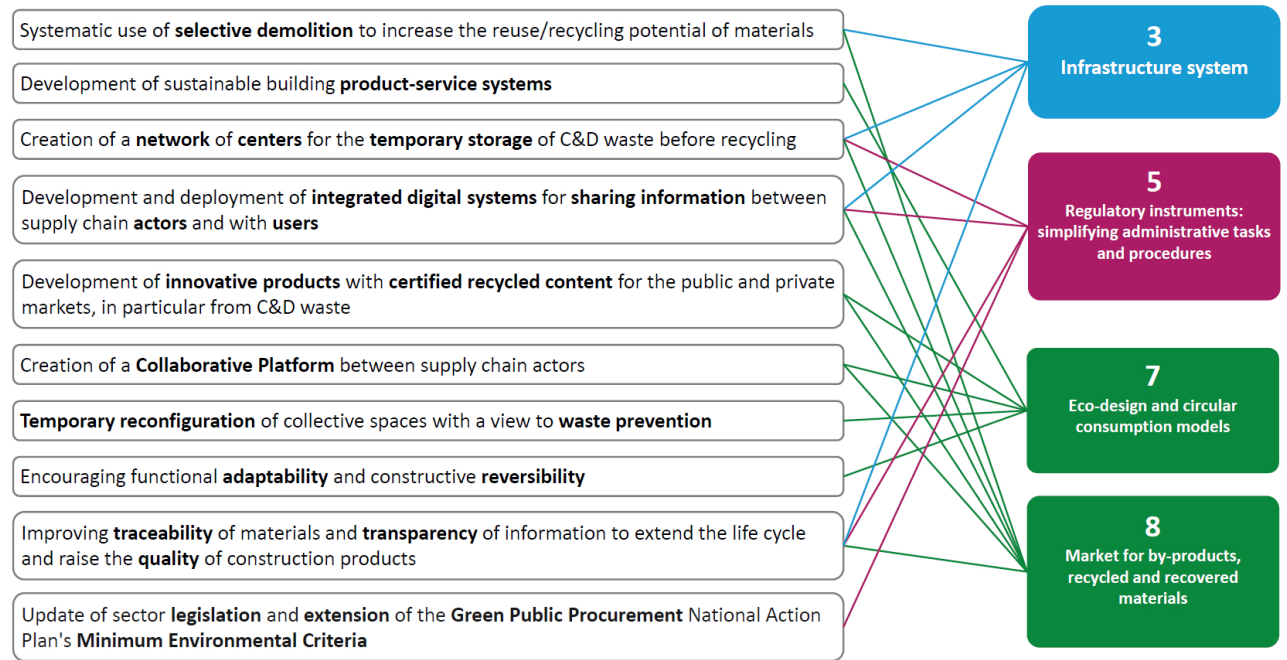


FIGURE 1: Links between the ICESP Priorities for a circular recovery after the COVID Pandemic, defined by the whole platform, and the ones identified by the C&D working group.

form, and to propose more specific priority actions – deriving and connected to the general ones - to be implemented in construction sector.

Subsequently, the research activity was further developed with the objective to uncover both national and international replicable good practices of various types and scales (including material, product, process, and building levels), able to exemplify the specific priorities identified by the Construction and Demolition sub Working Group. These examples were sourced from the database of good practices within the aforementioned platforms, as well as from the outcomes of stakeholders' engagement activities such as ECESP's EU Circular Talks (ECESP, 2021b).

The selected good practices serve to showcase the practical implementation of solutions that were previously perceived as experimental and challenging to be replicated. Adopting an exploratory qualitative approach grounded in the methodology of multiple case studies and participatory action research, the study analyzed several key Italian and international examples. These examples were then correlated with the strategic actions identified through the ICESP subgroup consultation, with the aim to select six exemplary good practices of different typologies, each embodying more than one of the priority strategic actions for the construction sector identified through stakeholders' consultation.

3.1 Outcomes of the stakeholder consultation on priorities for a circular construction sector

Among the priorities indicated by the stakeholders of the ICESP sub Working Group Construction and Demolition

(Table 1), the first is the creation of a collaborative platform, which is to be understood as a cooperative or system for collaboration between companies operating with the same type of goods, such as the Italian CPR System for the circular management of packaging in the agro-food sector, to be applied within the recycled aggregates supply chain.

A key aspect for the sector involves enhancing material traceability and information transparency to extend the life cycle and enhance the quality of construction products. Faced with recent functional re-layouts aimed at adapting spaces to new requirements (e.g., distancing, separation), it becomes imperative to prioritize not only the flexibility and reversibility of construction systems but also the traceability of materials and increased transparency of information. Only in this way will it be possible to aim at extending the life cycle of construction materials/products and avoid the generation of waste.

Specifically, concerning materials within existing built heritage, it is crucial to establish connections among various stakeholders in the supply chain (such as professionals, demolition firms, and recycling ones) to ensure traceable data flow - from pre-demolition audits to recycled products certification - passing through the characterization of the waste. Additionally, it is vital to institute feedback mechanisms from recyclers to identify prevalent contaminating factors in specific areas and types of buildings (based on construction period and/or geographical location).

A strategic front for the construction sector also includes encouraging functional adaptability and constructive reversibility. The COVID emergency has forced the widespread re-organization of spaces, from hospitals to

TABLE 1: Grouping of priorities into strategic actions for circularity practices' scale-up.

Priorities indicated by ICESP C&D working group's stakeholders	Strategic actions for circularity practices' scale-up
Collaborative platform (innovation)	Creation of a collaborative platform
Improving and certifying the quality of C&D waste products	Improved traceability of materials and transparency of information to extend the life cycle and raise the quality of construction products
Material traceability and information transparency with a view to life cycle extension	
Temporary reconfiguration of collective spaces (offices, schools, health) and waste prevention.	Temporary reconfiguration of collective spaces (offices, schools, health-care) and waste prevention
Functional adaptability and constructive reversibility	Encouraging functional adaptability and constructive reversibility
Promoting the adaptability of construction for the circular economy	
Development of Sustainable Product-Service Systems	Development of Sustainable Product-Service Systems
Creation of a network of widespread collection points for C&D waste	Creation of a network of centers for the temporary storage of C&D waste before recycling
Development of integrated systems for information sharing between actors in the supply chain.	Development and dissemination of integrated digital systems for sharing information between actors in the chain and with users
Development and dissemination of software aimed at supply chain actors and potential users	
Innovative materials produced with a high percentage of C&D waste	Development of innovative products with certified recycled content for the public and private markets, in particular from C&D waste
Development of products for the public and private markets with recycled content and certificates	
Use of recycled aggregates from C&D waste in cement production	Systematic introduction of selective dismantling to increase the recycling potential of the existing materials
Selective demolition	
Introducing selective demolition in a systematic way	
Selective demolition of buildings for improved recycling capacity of all materials present	
Systematic introduction of selective demolition	

houses, and has made it necessary to quickly set up temporary spaces. In order to avoid the increase in resource consumption and waste production linked to the adaptation, construction and dismantling processes of buildings, it is necessary to develop systems, products and process models that guarantee high reversibility within life cycle transformations, which are difficult to be predicted. For example, in interventions on the existing built heritage, it is necessary to favor design choices and intervention methods that favor the future receptivity of innovative building components. This aspect is linked to the temporary reconfiguration of collective spaces (offices, schools, healthcare facilities, etc.) through the design and creation of temporary configurations with a view to reversibility, reusability and transformability of products, building components and of buildings as a whole. In addition, it would be desirable to conduct a census by categories of all materials used in temporary situations in order to identify organisational models and supply chains for the selection/recovery/reuse/recycling of materials able to ensure an efficient use of resources.

The COVID emergency has also highlighted the need for alternative organisational and business models in the construction sector. Among these, the development of Sustainable Product-Service Systems is a promising scenario to provide customers with a sustainable result in terms of environmental load and resource utilisation, without transferring ownership.

Further priorities come from government policies. The introduction of significant incentives in the Italian 'Rilancio' Ministerial Decree (the so called 'Superbonus 110%' for buildings' energy retrofits) lead to an increase in the volume of C&D waste. Therefore, stakeholder proposed the creation of a widespread network of preliminary collection centres (e.g. at building material dealers' showrooms) to receive and temporarily store waste for subsequent recycling at duly authorised facilities. In addition, to ensure quality management of C&D waste from an environmental and technical point of view, the need for integrated digital systems for sharing information between the various stakeholders in the chain was highlighted, considering all stages of the life cycle, from demolition to waste treatment and reuse of materials. Digital tools are also needed to support operators in matching supply and demand. Hence, the development and dissemination of integrated digital systems for sharing information between supply chain actors and with users is seen as a priority.

Again, linked to government policies, given the extension of the obligation of the Italian Green Public Procurement Minimum Environmental Criteria for public buildings (Ministerial Decree) to private interventions under the Superbonus, it became a priority to develop new products with adequate recycled content and appropriate environmental certification, particularly for thermal insulation. In fact, to allow the use of products compliant to the mandatory criteria, which foresee minimum recycled content thresholds, the development of innovative certified products, providing users with the necessary traceability and performance information, plays a key role. For example, in the cement supply chain, it becomes a priority to exper-

iment the use of recycled aggregates from inert waste, currently used in cold cycles (concrete mixes), to partially replace the natural raw materials (limestone, clay, etc.) needed to make the so-called raw meal which, once fired in cement kilns, is turned into clinker (the main component of cement).

Finally, the systematic introduction of selective demolition would increase the recycling potential of materials in the built environment by ensuring quality circular processes, of which recycled aggregates for concrete are an example. Although a progressive increase in the attention of construction companies towards C&D waste sorting processes can be observed today in Italy, bureaucratic, economic and organisational barriers remain to a significant extension of selective demolition at national level. In order to overcome these barriers, an in-depth analysis of selective demolition techniques and related costs is needed, with reference to the reuse/recycling potential of the different types of materials (aggregates, plasterboard, iron, wood, plastics, etc.) and components, with a view to strengthening the associated recovery value chains. In parallel, it is indispensable to promote an architectural/technological design that, from a life cycle perspective, envisages the separability of components at the end of their useful life. Only through proper design, to be carried out prior to construction/demolition activities, can materials of a quality suitable for recovery be obtained.

4. RESULTS AND DISCUSSION

The six good practices presented here have been drawn and selected from the pool of those collected in the ICESP, ECESP and Taiwan Circular Network platforms. They were therefore gathered with a bottom-up approach by the stakeholders involved in the collaborative platforms but subjected to the scrutiny of the respective good practice evaluation committees, with reference to criteria such as levels of innovativeness and replicability. As anticipated, the six good practices exemplifying multiple priority actions identified by the ICESP's stakeholders are referred to different levels/scales: two at the building level (ZIN Project and Taitsugar Village), two at the product level (rice waste-based building products and concrete with certified recycled aggregates), two at the digital tool level (Madaster and Concular platform). The selection criteria considered in fact at the multiscale required by circularity in such a complex sector as construction and aimed at showing innovative products, building and neighborhood design as well as digital tools supporting the development of both products and buildings. Moreover, the selected good practices are promoted by different actors involved in the abovementioned Platforms and representing various Countries (Belgium, Taiwan, Netherlands, Germany, Italy), in order to highlight some international excellence experiences in circular construction.

The following table (Table 2) shows the multiple links between selected the good practices and the strategic actions identified in the ICESP's stakeholders' consultation, the results of which were described in the previous section.

TABLE 2: Comparative chart of the selected six good practices in relation to the strategic actions collected in the ICESP C&D working group stakeholders' consultation.

Good practice	1	2	3	4	5	6
Scale	Building/neighborhood		Product/material		Digital tool	
Strategic reference actions	ZIN Project	Taitsugar Village	Rice waste-based building products	Concrete with certified recycled aggregates	Madaster platform	Concular platform
Creation of a collaborative platform	X				X	X
Improved traceability of materials and transparency of information to extend the life cycle and raise the quality of construction products	X	X	X	X	X	X
Temporary reconfiguration of collective spaces (offices, schools, healthcare) and waste prevention	X	X				
Encouraging functional adaptability and construction reversibility	X	X				
Development of Sustainable Product-Service Systems		X				
Creation of a network of centers for the temporary storage of C&D waste before recycling	X			X		
Development and dissemination of integrated digital systems for sharing information between supply chain actors and with users					X	X
Development of innovative products with certified recycled content for the public and private markets, in particular from C&D waste	X	X	X	X		
Systematic introduction of selective dismantling to increase the recycling potential of the materials present	X	X				

4.1 ZIN Project (Brussels, BE)

In 2018, a brand-new application of circularity principles was initiated in the renovation of two of the four towers of the World Trade Centre in Brussels in the ZIN project, currently under construction. The good practice was promoted in a stakeholder engagement activity organised by ENEA as part of ECESP (Circular Talk, 2021), and was illustrated by Drees and Sommer, a consultancy firm that focuses on the implementation of the Cradle to Cradle design approaches and certification systems (EPEA).

The development of a materials inventory on both buildings enabled the use of different strategies to prevent C&D waste and diverted it from landfill through reuse, recycling and upcycling, and the certification of materials in cooperation with different stakeholders, adopting the 'Building as Material Bank' approach (Figure 2).

This ensured that 95 per cent of existing materials were kept in place, reused or recycled, while 97 per cent of new materials are Cradle to Cradle certified or equivalent, including concrete with onsite produced recycled aggregates (Figure 3).

The whole process was supported by EPEA and Drees and Sommer from the selective demolition of concrete, its on-site recycling, and the production of recycled aggregates to make Cradle to Cradle certified concrete to be reused in the deep tower retrofit project, which saw the introduction of a previously unforeseen functional mix.

Overall, 70.000 tons of demolition materials were up-cycled (concrete, flatglass, aluminium) for reuse on site after re-manufacturing, 1.646 tons of materials and components (0,6% of total weight) were reclaimed to be reused on site and on other sites; only 710 tons have been evacuated (Figure 4).

The towers' refunctionalization and adaptation project was, then, structured according to the principles of modularity and Design for Deconstruction, to maximise materials and components' future recovery.

4.2 Taitsugar Circular Village (Taiwan)

In the Shalun Smart Green Energy Science City in Taiwan, one of the first 'circular villages' in the world has been realised, the TaiSugar Circular Village (Bio-architecture Formosana, 2021), consisting of three housing blocks and a courtyard with shared spaces, aquaponic cultivation, phyto-purification, and solar energy production by BIPV (Building Integrated Photovoltaic) (Figure 5). The good practice is listed in the Circular Taiwan Network database.

The buildings were designed with a life-cycle and circular approach, relying on a modular and prefabricated system for both the load-bearing structure and the façade construction, using a mixed system of steel and wood (Cross Laminated Timber panels) to make assembly and disassembly more efficient. Insulating blocks made of recycled LED glass were used for the façades and interior partitions respectively.

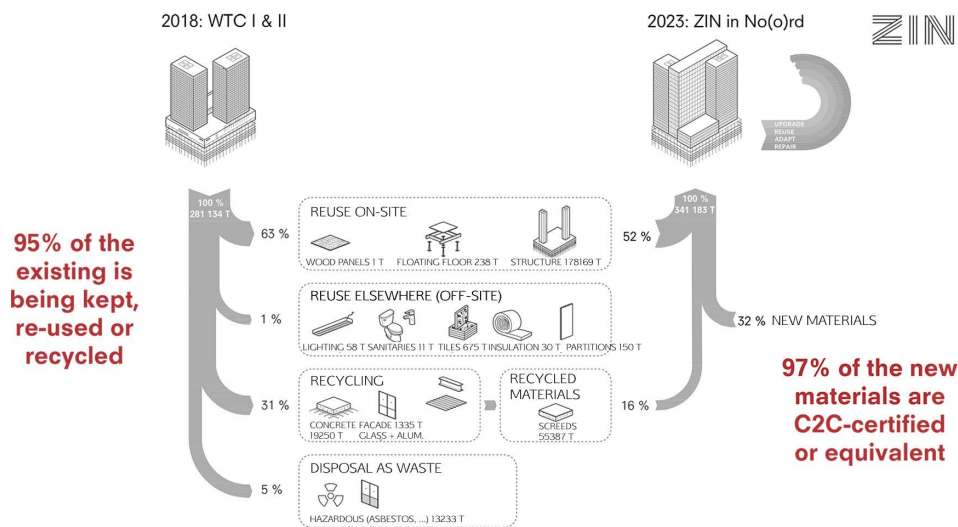


FIGURE 2: Building as Material Bank concept as implemented in the deep renovation of two of the four towers of the World Trade Centre in Brussels, within the ZIN Project (Source: M. De Moradiellos, Drees and Sommer, ECESP Circular Talk 2021).



FIGURE 3: Local closed-loop recycling of concrete in the ZIN Project in Brussels, BE (Source: M. De Moradiellos, Drees and Sommer, ECESP Circular Talk 2021).



FIGURE 4: Overview of the stocks of reclaimed building components disassembled from the World Trade Centre towers and destined to onsite/offsite reuse.

The BIM technology was also applied to integrate material and structure information to facilitate assembly and disassembly. The digital twin of the buildings will serve as a material database for future replacement or reuse, through the use of Material Passports integrated in the

BIM models (Figure 6). With a view to the building as a 'bank' of construction materials, thanks to reversible connection systems and the modularity of the components, the reclaimed materials used include wooden components obtained from the dismantling of dilapidated buildings, re-



FIGURE 5: Overall view of the Taitsugar Circular Village in Taiwan (Source: Bio-architecture Formosana).

建材銀行 / 建材護照
 material bank / material passport

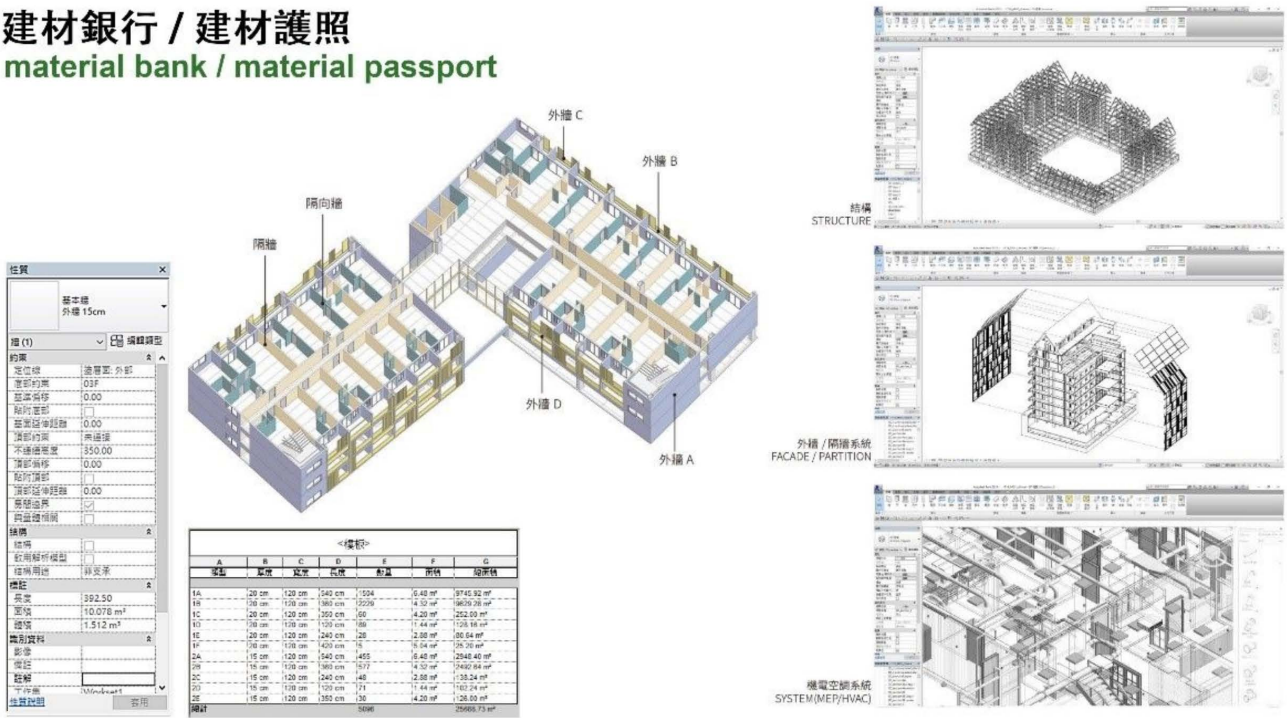


FIGURE 6: Material passports collected in the BIM models of the Taitsugar Village's buildings (Source: Bio-architecture Formosana).

used as structural components for one of the pavilions and railway tracks reused as fencing.

Moreover, a key aspect of this good practice is the servitisation of different elements and installations of the buildings. Lift blocks, lighting fixtures, furniture and sanitary components have been 'rented' rather than purchased (product as service) (Figure 7). The servitisation approach, already tried out in some pioneer projects starting with the lighting systems in large facilities such airports, is thus extended to various technical elements of the building, which can be better maintained. The flats are rented out to residents to whom the manager will provide all the necessary maintenance, from the building to furniture and appliances. In this way, the user will pay to use and not to own. In addition, the rental of cars, scooters and electric bicycles for residents has been introduced.

Overall, Tait sugar Village is a concrete example of a fully circular settlement, where the flows of building materials, energy, water and food are all managed in an efficient, closed-loop and local way.

4.3 Ricehouse: building materials made from rice husk and straw

The Italian Benefit company Ricehouse (listed in the IC-ESP and ECESP good practices' databases) has developed

a product innovation based on the valorisation of production waste from the rice value chain. Most of the secondary materials generated by primary agricultural production are treated as waste, notwithstanding their intrinsic market value, potentially exceeding the disposal costs. For each hectare of soil dedicated to rice production, 7 tons of rice is produced, while residues are on average 10 tons. The good practice regards the development of innovative construction products valorizing the by-products of rice cultivation, such as husk and straw. For example, Ricehouse has industrialised the production process of pre-stressed wood and rice straw frames, to build prefabricated passive houses. In such constructive system, rice straw insulation offers a balanced thermal comfort and requires minimum energy consumption for heating the building, which can be provided by renewable sources.

Other very innovative products are made from rice husks and lime: natural mortars are made by mixing air lime and rice husks. The most interesting ones can be used as plasters (Figure 8) and aesthetically enhance the presence of waste fibres in the mix. One of the company's most innovative clay-based mixes also guarantees the possibility of 3D printing large-scale constructions (Figure 9).

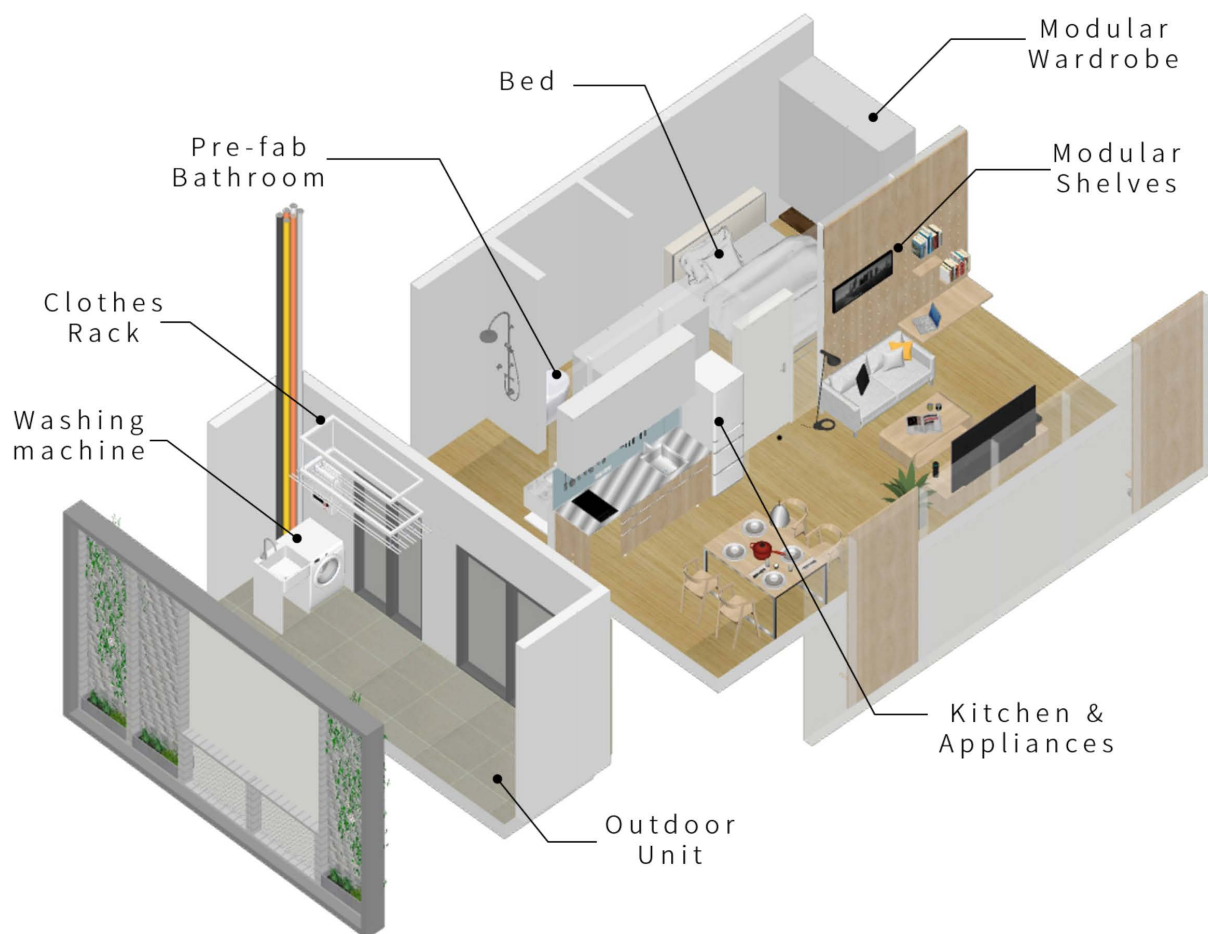


FIGURE 7: Prefabricated installations blocks rented through a servitisation procurement model, within the modular housing units (Source: Bio-architecture Formosana).



Intonachino
RH120



Intonachino naturale per regolarizzazione
RH200

FIGURE 8: Examples of Ricehouse lime and rice husk plasters (Source: <https://www.ricehouse.it/prodotti/finiture/>).



FIGURE 9: Gaia Project by Ricehouse, 3D printed house with biomass insulation built with a 3D Crane WASP printer and natural materials from the landscape nearby, in a blend of natural materials including clay, ground sand, rice straw and rice husk (Source: <https://www.ricehouse.it/storie/gaia/>).

4.4 Production of concrete with a certified content of recycled aggregates compliant to the Italian Green Public Procurement Minimum Environmental Criteria

The good practice, coming from the ICESP database, refers to the Italian company Italcementi, among the first large national companies to have activated concrete pro-

duction lines for construction, and not for infrastructures as usual, with mixes that incorporate recycled aggregates from the recovery of inert demolition waste. The circular economy practice implemented by the company consists in the production of pre-packaged concrete and filling mixes with partial replacement of natural aggregates, which represent one of the main constituents of concrete, with

aggregates recycled from demolition concrete or secondary raw materials from industrial origin (industrial aggregates) such as, for example, steel mill slag. The concretes produced reach a total content of recycled material varying between 8 and 15%. The filling mixes have a total content of recycled material larger than 70%. These average recycled contents are compliant to the Italian Green Public Procurement Minimum Environmental Criteria for buildings (in force since 2016), which require concretes with at least 5% recycled content.

In the period 2014-2019 the company used approximately 64,400 tons of aggregate recycled from C&D waste and approximately 42,450 tons of industrial aggregates. Evaluation and certification of environmental impacts and recycled percentages through LCA studies and EPD declarations (Type III Environmental Product Declarations) made it possible to use such products in works subject to Green Public Procurement.

4.5 Madaster platform aimed at sharing information about building materials

Madaster, the 'Materials Cadastre' (Figure 10), is a good practice from the ECESP database. It is the first platform that facilitates the centralized and standardized web-based generation and registration of materials passports in the construction and infrastructure sector. It is operational in 5 European countries, with projects registered all over the world.

The platform generates and records passports of materials, products, buildings and areas, providing a detailed inventory with information on quantity, quality, size and location of all materials, components and products used in a specific construction intervention (Figure 11). Furthermore, the platform calculates a financial evaluation (current and expected future residual value of the materials) and pro-

vides an overview of the environmental impact (Circularity Index, embodied carbon). The passport can be adapted and enriched over time, providing a dynamic repository of data, which can also be archived at a specific moment. The platform is interoperable with BIM models.

4.6 Concular platform for the exchange and assessment of reclaimed building components

Concular, a good practice from the ECESP database, is a digital platform (Figure 12) that allows the recirculation of building materials. The platform targets professionals in the construction industry, such as portfolio holders, project developers, building owners, manufacturers and architectural firms.

The software allows new and existing building materials to be digitized using AI technology. It also measures embodied carbon emissions and quantifies how these can be reduced by recirculating a building's materials. Architectural firms can also source low-carbon reclaimed materials and low-carbon recycled materials using the platform. The platform therefore supports the collaboration between the construction value chain's actors.

Connected to Concular is the Restado.de marketplace that brings building materials from demolition or oversupply in contact with the demand coming from new construction projects. Restado's mission is to extend the life cycle of building materials, which are reused many times. Restado's target group consists mainly of craftsmen, small construction companies and private owners.

5. CONCLUSIONS

This paper demonstrates the role of collaborative platforms in scaling up effective circular practices for

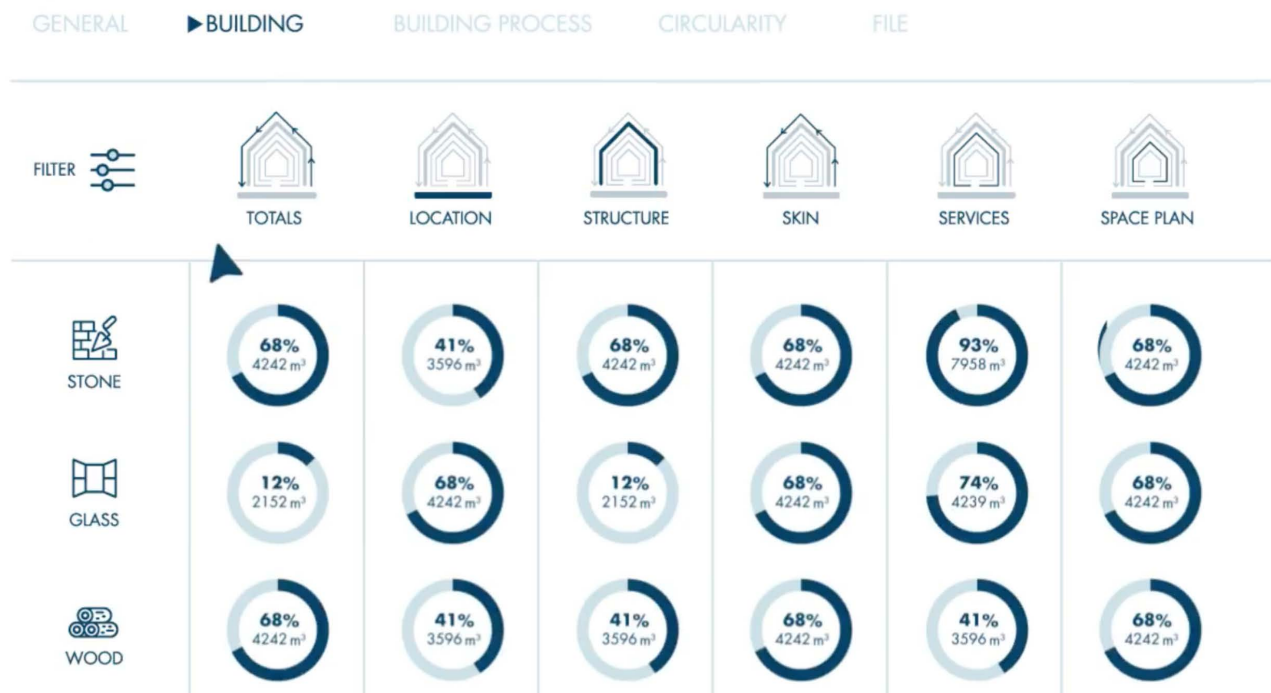


FIGURE 10: Madaster platform's interface for building materials' mapping and registering (Source: <https://madaster.com/>).

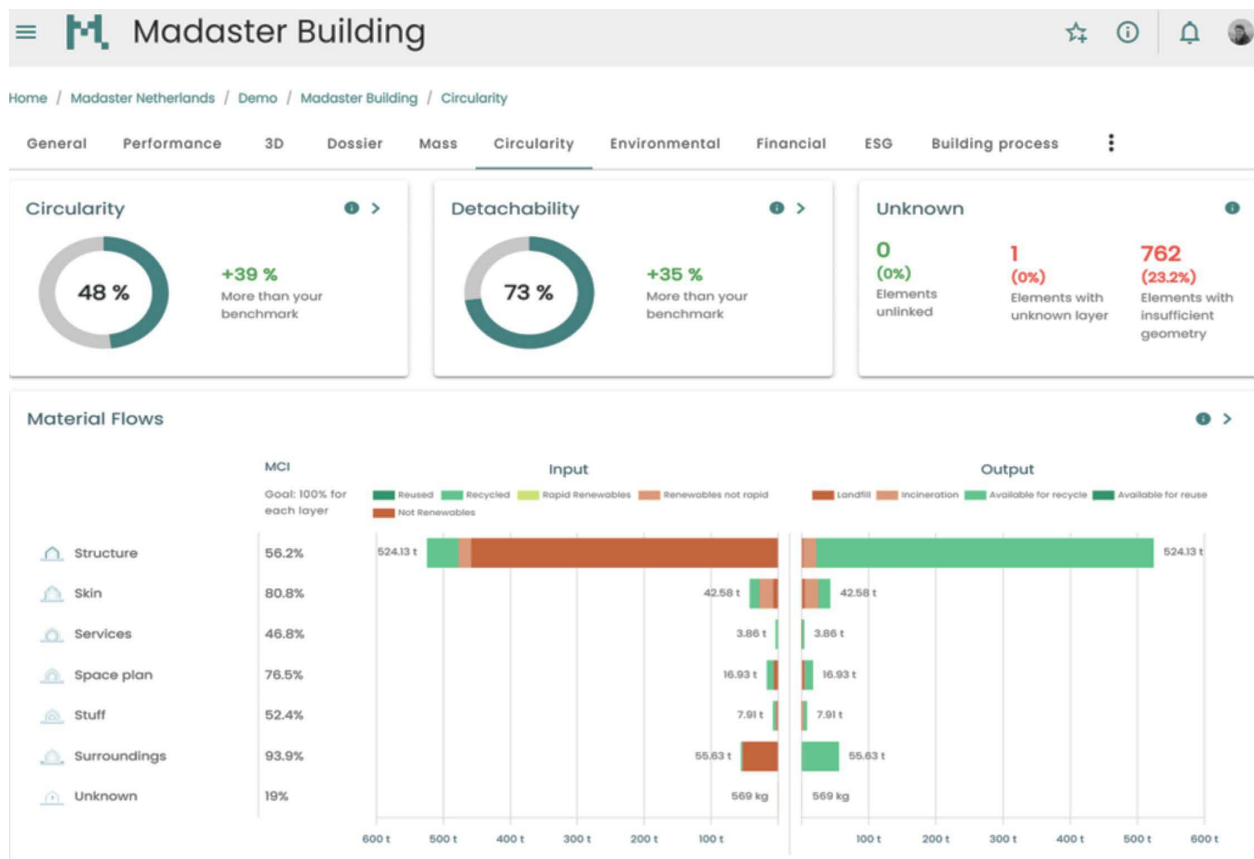


FIGURE 11: Materials mass balance and embodied carbon estimation in the Madaster platform. (Source: <https://madaster.com/>).

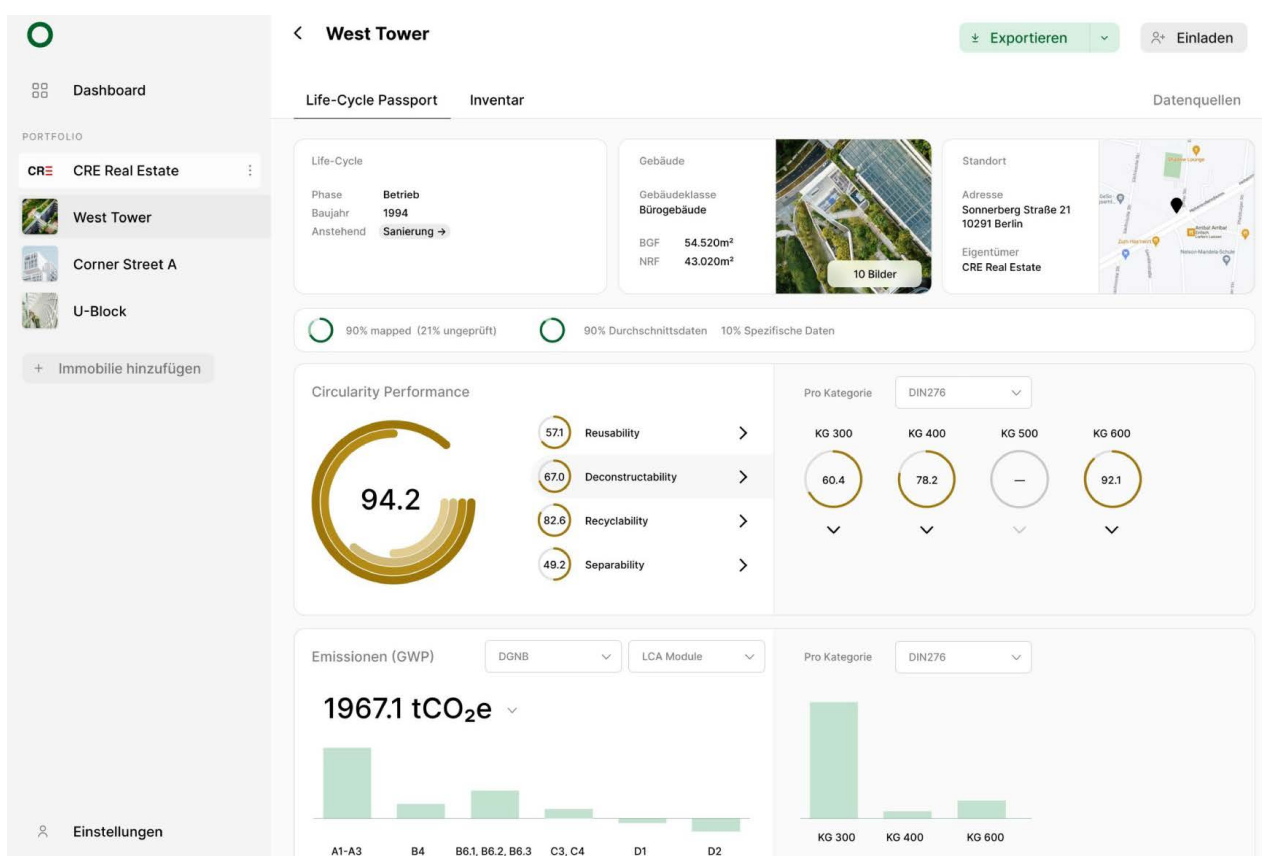


FIGURE 12: Concular platform interface (Source: <https://concular.de/>).

the built environment, serving as tools for implementing the quintuple helix model. The results reveal how national-level stakeholder discussions and exchanges have led to the identification of nine strategic actions encompassing various forms of innovation. These actions span from product and process innovations to more intricate forms related to consumption and organizational models, including supply chains. For each of these actions, we presented some exemplary practices selected from those identified in the collaborative platforms under review (ICESP, ECESP, Holland Circular Hotspot, Circular Taiwan Network). These cases showcase product, process, and organizational innovations, illustrating how circular economy principles can be applied to the built environment. The benefits include enhanced resource efficiency, reduced waste, improved environmental quality, and increased sector competitiveness. These showcased examples exemplify the identified strategic actions and underscore how collaborative platforms serve as reservoirs of innovation and knowledge for transitioning to circular construction.

The aforementioned good practices serve as examples of the strategic actions identified through stakeholder engagement activities within ICESP. Among these, the most recurring actions in the selected cases include: "Creation of a collaborative platform", "Improved material traceability and transparency of information to extend the life cycle and raise the quality of construction products", and "Development of innovative products with recycled content, certified for the public and private markets, particularly from C&D waste". These three strategies are evidently interconnected, notably demonstrated in the advanced good practice of the ZIN Project. These good practices underscore how collaborative platforms act as reservoirs of innovation and knowledge for transitioning to circular constructions and facilitating their scale-up. Moreover, the work emphasizes how the quintuple helix approach, based on cooperation between institutions, universities, industries, civil society, and the natural environment, fosters the creation, production, application, dissemination, and continuous use of knowledge and innovation, promoting eco-innovation and eco-entrepreneurship in the construction sector.

However, some limitations are present in this contribution. It is based on an exploratory qualitative methodology utilizing multiple case studies and participatory action research, which does not allow for statistical generalization of results or systematic comparison of different experiences. Furthermore, the analysis focuses only on a few selected good practices from the surveyed collaborative platforms, which are not exhaustive of all possible circularity solutions applicable to the construction sector. Nevertheless, ongoing research activities aim to expand the mapping of good practices traceable to the strategic actions identified in the stakeholder consultation ran in ICESP. This will be accomplished through a more structured census focused on specific types of good practices, particularly digital platforms enabling collaboration between different actors, traceability of components, promotion of reuse and recycling, valorization, and facilitation of meeting between demand and supply of recovered materials and components. These platforms are proving to be strategic for the scale-up of circular project good practices.

ACKNOWLEDGEMENTS

This contribution reports the results of a joint research activity between the authors and their respective institutions, developed within the activities of the ICESP Platform, Working Group 'Sustainable and Circular Value Chains' ('Catene di valore sostenibili e circolari'), sub-working group 'Construction and Demolition', coordinated by F. Ceruti and P. Altamura, and developed in collaboration with L. Cutaia, representative of the ECESP Leadership Group 'Construction and Infrastructure'.

P. Altamura participates in research activities as a 'PON Research and Innovation' Fellow Researcher (RTDA) (DM 1062/2021) on the green project 'Climate, green technologies and circular communities. Design of innovative settlement models according to the 'Green Building Approach' for the mitigation of climate change and the reduction of its impacts, through the sustainable management of technologies and resources' (2021-2024), Scientific Referent Prof. F. Tucci, PDTA Department, "Sapienza" University of Rome.

S. Baiani carried out this study within the MICS (Made in Italy – Circular and Sustainable) Extended Partnership and received funding from the European Union Next-GenerationEU (Piano Nazionale di Ripresa e Resilienza (PNRR) – Missione 4 Componente 2, Investimento 1.3 – D.D. 1551.11-10-2022, PE000000004). This manuscript reflects only the authors' views and opinions, neither the European Union nor the European Commission can be considered responsible for them.

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INNOVATIVE BUSINESS MODELS IN THE CIRCULAR ECONOMY: AN ANALYSIS OF GOOD PRACTICES FOR ELECTRICAL AND ELECTRONIC EQUIPMENT ON THE EUROPEAN CIRCULAR ECONOMY STAKEHOLDER PLATFORM

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Article Info:

Received:
3 May 2024
Revised:
5 July 2024
Accepted:
24 July 2024
Available online:
7 September 2024

Keywords:

EEE
WEEE
Circular economy platforms
Good practices
Urban mining

ABSTRACT

The pervasive integration of electronic technologies in the contemporary digital landscape underscores the significance of electrical and electronic equipment (EEE). Beyond their utility in daily life, certain components within EEE, hold classification as critical raw materials (CRMs) and transform cities into urban mines. This issue highlights the necessity of optimizing its sustainable consumption and production patterns at all stages of the life cycle and across various sectors. The optimization of the complete product life cycle serves as a foundational pillar of the circular economy (CE). While the transition from a linear to a CE has been a longstanding objective promoted by the EU, a comprehensive mapping elucidating potential shortcoming in the effective implementation of this transition in Europe for the field of EEE remains elusive. This paper seeks to investigate the state-of-the-art of CE good practices for EEEs in Europe, shedding light on potential strengths and challenges to be addressed. To obtain an empirical perspective, the European Circular Economy Stakeholder Platform (ECESP) serves as a tool to delineate the trajectory of progress throughout the transition to a CE. The analysis encompasses both life cycle phases and the production sector. The findings underscore a prevailing focus on the product's end-of-life phase and sectors associated with recycling and waste management, while the initial life cycle phase remains underexplored in terms of eco-design solutions or innovative processes. On a positive note, several instances of new business models have emerged, redirecting consumption emphasis towards services.

1. INTRODUCTION

In today's technologically driven world, our reliance on electrical and electronic equipment (EEE) is undeniable, shaping our daily routines and societal structures. At the heart of this reliance lies the supply chain of EEE, which is strongly connected to the utilization of the so-called Critical Raw Materials (CRMs). The CRMs are key components essential for various strategic sectors, including renewable energy production, electric mobility, and defense. Their economic importance is evident in their end-use applications and the value they add to EU manufacturing sectors. However, sourcing these materials is challenging, due to a risk of disruption in the EU supply chain (Carrara et al., 2023). The first list of CRMs was compiled in 2011 (European Commission, 2011) and then regularly updated

every three years (European Commission, 2014; 2017; 2020b; 2023a). All materials listed have a supply risk and economic importance higher than a threshold value set by the EU, as defined by the methodology established in 2017. These CRMs, identified by the European Union, play a pivotal role in developing the goals outlined in the 2030 Agenda for Sustainable Development, given their application in strategic sectors (Mancini et al., 2019). In order to achieve the sustainability objectives, the availability and responsible management of CRMs emerge as keystones for succeeding. However, challenges in CRMs supply chains, coupled with their indispensable role in sustainable development, implies the urgency of strategic action. The Critical Raw Materials Act, issued in March 2023 (European Commission, 2023b), aligns with this direction. It highlights



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Detritus / Volume 28 - 2024 / pages 19-27
<https://doi.org/10.31025/2611-4135/2024.19407>
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Europe's commitment to addressing these challenges and leveraging CRMs to advance towards a more sustainable future. This act emphasizes concerted efforts to ensure the availability of CRMs, promote responsible sourcing practices, and foster innovation in CRMs recycling. In this context, particular attention is drawn to the urban context, where cities brim with electronic devices. Proper management and recycling of such devices can lead to virtuous internal flows of CRMs, effectively transforming cities into real urban mines (Andooz et al., 2022). The interest in facilitating the collection of electrical devices for consumers has led to the introduction, since 2008, of extended producer responsibility (EPR) for waste electrical and electronic equipment (WEEE) (European Union, 2008). This obligates producers to manage the end-of-life phase of devices they introduce to the market, involving both assuming the financial burden of their disposal and directly supervising collection efforts. In addition, the European Directive 2012/19/EU, starting from 2019, has set a collection target of 65%, calculated as the ratio between the total weight of collected WEEE and the average weight of EEE placed on the market in the previous three years (European Union, 2012). In 2019, three European countries exceeded the target: Bulgaria, Croatia, and Poland, while Italy reached approximately 40% (European Commission, 2023c). The European effort in sustainability also holds in the action "Closing the loop - A EU Action Plan for the Circular Economy" and subsequently, since April 2018, the European package on the CE (Stahel, 2016; Stindt et al., 2014; Neligan et al., 2023; Geissdoerfer et al., 2020; Geissdoerfer et al., 2017) that contains a series of provisions aimed at promoting the transition to a new circular model. In fact, within the European Green Deal, a circular transition is considered crucial for attaining this goal (European Commission, 2019). This approach not only contributes to climate change mitigation and the promotion of sustainability but is also fundamental for advancing the Sustainable Development Goals (SDGs) from the Agenda 2030 (Lambda et al., 2024; Binsuwadan et al., 2023). In fact, in the European Green Deal, where Member States have been committed to achieving the objective of climate neutrality by 2050, the circular transition is considered crucial for achieving the goal. Therefore, the shift from a linear to a circular economy (CE) is a path advocated by Europe for many years already. However, its implementation within companies is often not widely perceived by society, resulting in a knowledge gap between industry and the broader community (Rocca et al., 2023). This gap contributes to the perception of the circular model as a distant goal rather than an achievable reality. Consequently, there is a pressing need to explore empirical cases shared by companies to gain insights into the key success factors, sustainable challenges, and the necessary steps toward achieving the paradigm shift from linear to circular models. CE platforms have emerged as a response to this need, providing a comprehensive overview of ongoing experiences (Beltrani et al., 2021; 2022). These platforms serve as invaluable tools for sharing and exchanging ideas, strategies, initiatives, knowledge, and challenges among diverse users. Through these platforms, stakeholders can collaborate and learn from each other's experiences, accel-

erating progress towards a more CE on a global scale. Furthermore, in the Action Plan updated in 2020, the European Commission identified the electronic components sector among the priority productive sectors (European Commission, 2020a). This further emphasizes the importance of having information regarding the state-of-the-art in the development of circular solutions for EEE and WEEE (Cardenia et al., 2022; Cheshmeh et al., 2023).

To this end, this contribution aims to empirically investigate the spread of CE in Europe, with a specific focus on the EEE value chain. The investigative tool employed was that of good practices (GPs), which represent the primary means of communicating innovation in the CE today. This work will present a comprehensive mapping, within the European contexts, of GPs related to the management of EEE using European Circular Economy Stakeholder Platform (ECESP) repository. ECESP is an initiative of the European Commission and the European Economic and Social Committee to support the transition towards a circular economy and the implementation of the EU Circular Economy Action Plan. The Platform is a repository of a vast amount of information, knowledge, experiences, and best practices on the circular economy in Europe that can be shared with the public. This is because one of the platform's main goals is to collect and exchange relevant practices that demonstrate circularity in European countries. This mapping of GPs is essential for understanding the characteristics, strengths, and challenges encountered across all phases of the life cycle and the economic sectors involved.

The paper is organized as follows: in the subsequent section 2, an overview of the European Circular Economy Stakeholder Platform (ECESP) is provided. Section 3 describes the methodology adopted for the analysis of the GPs, starting with the criteria for platform research, followed by the identification of study variables and groupings. Section 4 presents the results of the qualitative analysis regarding the life cycle phase and sector of origin. Finally, section 5 presents the conclusions drawn.

2. GOOD PRACTICES AND EUROPEAN CIRCULAR ECONOMY STAKEHOLDER PLATFORM

The ECESP is the main repository of GPs in the field of CE in Europe, established in 2017 as a joint initiative of the European Commission and the European Economic and Social Committee (EESC). Its primary objective is to support the dissemination of CE principles at the European level and enable stakeholders from different countries and sectors to meet and interact, highlighting cross-sectoral opportunities as well as potential challenges. ECESP is composed of a Coordination Group consisting of 24 members representing business networks, workers, consumers, civil society, and local authorities. The Coordination Group is tasked with strengthening interaction among stakeholders, facilitating the exchange of GPs, and promoting a European debate for a full transition to a CE (ECESP, 2024).

In ECESP, in the "Submit a Good Practice" section, each user can submit their GP by completing four sections: the first contains an initial description with the necessary infor-

mation for completing the following sections; the second requests contact details of the company and the person completing the GP form, as well as consent for publication; the third section requires the title, type of organization, main field of activity/sector; in the fourth and final section, the title and a description of the GP, the qualitative/quantitative results obtained, the level of application, the duration, the original language, the key areas, and the type of funding received are requested (ECESP, 2023).

ECESP has defined and published on its website guidelines (ECESP, 2018) that identify the key criteria/requirements that the review group considers before inclusion on the website, making the approval and publication process faster.

The criteria considered by the ECESP review group as fundamental principles for the publication of GPs are:

- Relevance to the CE;
- Completeness and clarity of information;
- Tangibility of expected results;
- Educational and behavioral contribution/value-added;
- Compliance with European publishing rules.

In this regard, GPs must demonstrate the valorization of resources throughout the entire production cycle: production, consumption, and disposal phases. They must also propose innovative solutions to implement circularity through new business models. The description of GPs must be clear and highlight their relevance within the CE. The results and purpose must be clear and quantifiable, and the technologies used to achieve them must be specified. The feasibility of GPs must be evidenced by concrete and reproducible results, implying that projects must already be in progress or completed to proceed with publication. GPs that do not directly relate to the production cycle may still be considered if they contribute indirectly to achieving the CE, such as by increasing consumer awareness and incentivizing behavioral changes or facilitating resource sharing and reuse. Finally, GPs included in the ECESP must always comply with European rules, avoiding ethically inappropriate and promotional content without providing useful information.

It is important, finally, to underline that parallelly with the establishment of the ECESP, several European nations have instituted platforms connected to ECESP. In this regard, in Italy, in 2018, ENEA promoted the creation of a national interface for ECESP, giving life to the mirror platform Italian Circular Economy Stakeholder Platform (ICESP). One of the main objectives of this platform is the collection and mapping of GPs in the Italian circular economy to promote understanding and replicability of successful cases (Del Vecchio et al. 2021; ICESP 2024).

3. METHODOLOGY

The analysis of the GPs presented in this contribution is based on two main criteria: their positioning during the life cycle phase and their reference sector. The choice to analyze the GPs according to these two variables stems from the fact that the objective of this work is to carry out a mapping that helps understand which phases and sectors

are most prevalent and which ones, instead, require more attention. This enables an understanding of how Europe is positioning itself regarding the development of circular solutions for EEEs. In the following subsections, the selection criteria, as well as the definition and grouping of the chosen variables, will be described.

3.1 Criteria for identifying good practices

To identify the pertinent GPs, the search engine on the ECESP website was utilized with keywords such as: “EEE”, “Electronic devices”, “WEEE” and “E-waste”. This search yielded a total of 43 selected GPs concerning electronic devices.

The information regarding the collection, systematization, review, and dissemination of GPs was sourced and analyzed starting from the document that highlights the guidelines for the CE GPs on the ICESP (Beltrani et al., 2021). Many details regarding EEE and WEEE and the related GPs were obtained from the document reported by Working Group 4 of ICESP (Cardenia et al. 2022) where also a list of GPs available on ICESP and ECESP related to WEEE is reported. Other important starting points were provided by “Analysis and Mapping of Italian Circular Economy Good Practices by Sector and Product Life Cycle Phase” (Beltrani et al., 2020) and “Analysis of the Replicability of Good Practices and Quantification of Environmental, Economic, and Social Impact at the National Scale” (Beltrani et al., 2022), both published by ICESP.

3.2 Variables definition and groupings

By studying the various options that organizations can select when submitting their practices through the drop-down menus on ECESP, several observations were made: whether the fields to be filled in the collection form on the platform were mandatory or not; whether there were differences in terms of the number of options and the possibility of multi-tagging, i.e., selecting more than one option in the respective fields. Based on the conducted study and the gathered information, the life cycle phase and the sector have been selected as study variables.

In the GP collection forms, it is necessary to provide information regarding the life cycle phase and the type of organization, while it is not mandatory to provide information about the funding type, start date, and practice location. Additionally, ECESP allows unlimited multi-tagging for all entries specified by the organization during the GP submission. Since most of the projects submitted on the platform present multiple life cycle phases and sectors chosen by the organizations, a grouping of related areas was constructed. This choice was made to facilitate the analysis, and in the following subsections, both the variables and the groups of items falling under the same main variable will be defined.

3.2.1 Life cycle phases

In Figure 1 are shown the life cycle phases of a product in the CE framework that include: “Innovation and Investments”, “Production”, “Consumption”, “Waste Management” and “From waste to resources” (or “Secondary Raw Materials”). This framework is inspired by the first European

Circular Economy Action Plan of 2015, in which specific areas of intervention are identified (European Commission, 2015). ECESP adopts these five phases (or key area) to classify European GPs in the circular economy. Our approach involves collecting and clustering this data for each EEE GP. In Table 1, each phase is reported and subsequently defined by us through activities based on the conceptual framework proposed by La Monica (2018). On the other hand, the delineation of the “Innovation and Investments” phase constitutes an original contribution by this study, synthesized from the European CE Action Plan of 2015 and insights derived from the ECESP’s GPs. In addition, when more than one life cycle phase was indicated by the organization, a critical selection of the representative one was made by studying the detailed description and websites of each GP to verify their correspondence with what is reported in Table 1 for each life cycle phase.

The analysis of 43 GPs concerning EEE on the ECESP revealed that only 4 GPs are situated in the “Innovation and Investments” phase, 6 are associated with the “Production” phase, 14 pertain to the “Consumption” phase, 14 are distributed in the end-of-life phase related to “Waste Management”, while 5 are related to “Secondary Raw Materials,” representing production scraps or materials derived from

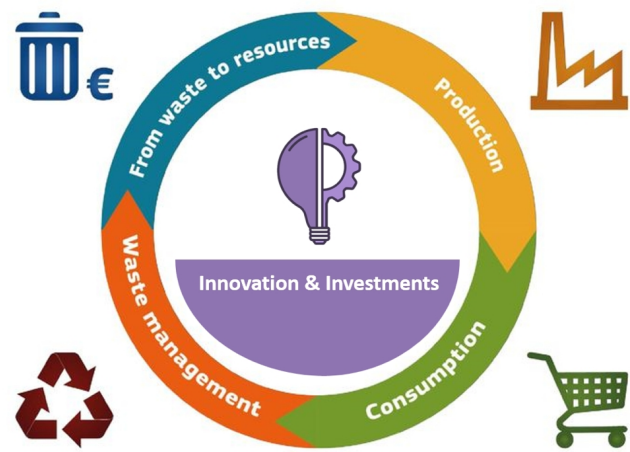


FIGURE 1: Circular Economy Framework (source: Authors’ elaboration from European Commission (2018)).

recycling processes that can be reintroduced into the economic system as new raw materials.

3.2.2 Sectors

In ECESP, all sectors indicated in the drop-down menu can be selected. Generally, the sector submitted by the or-

TABLE 1: List of groupings for the life cycle phases (source: Authors’ elaboration from La Monica, 2018).

Life cycle phases	Activities
Innovation and Investments	- Eco-friendly design or eco-design - Incremental innovation and improvement - Development of new technologies for treatment, separation, and recovery - New platforms and software licenses - Development of new business models - Programs for the implementation of innovative investments
Production	- Life Cycle Thinking - Ecologically Equipped Production Areas - Industrial symbiosis - Remanufacturing - Reverse logistics - Dematerialization and digitalization - Eco-Management and Audit Scheme - Maintenance and repair - Identification and replacement of hazardous chemicals - Replacement of non-renewable materials with renewable and recycled - Dissemination of GPs and good techniques in various industrial sectors
Consumption	- Green Public Procurement – GPP - Green labels - Sharing economy or collaborative economy - Consumption of services instead of products - Repair and reuse centers - Circular city - Awareness, education, and communication campaigns - Practices to counter planned obsolescence
Waste Management	- Integrated waste management - Simplification, certainty, and clarity of waste legislation - Improvement of control and inspection regimes - Resource flow mapping - Traceability of resources, products, services, and supply chains - High-quality separate collection and recycling infrastructure - Extended producer and consumer responsibility schemes - Biological treatment of waste (e.g., bio-refining, composting...)
From waste to resources	- Creation of secondary raw material markets and recycled materials - Platforms to match supply and demand of secondary raw materials - Increased demand for secondary raw materials and recycled materials - Clarity about the sources, composition, and quality of secondary materials - Urban and landfill mining - Promotion of non-toxic material cycles - Availability of data on secondary raw materials and information exchange

ganization has been retained, but in cases where more than one sector was selected, an analysis of individual GPs was conducted to identify the most relevant one. This resulted in a single, primary sector of reference for each GP. Table 2 shows the five groups representing the sectors involved in the 43 ECESP BPs related to EEE.

Sector groupings were made for our analysis. The first grouping relates to sectors closely connected with the “End-of-Use” phase of the product; hence, the categories related to separate collection, repair and reuse, recycling, waste management, and secondary raw materials. The second group pertains to “Education and Awareness”; thus, education, consumer awareness and accountability activities, and social engagement. Following that, the third grouping involves “Innovative Solutions,” encompassing eco-design, research, and digital technologies. Additionally, “Services” include sectors related to energy and public services, product-as-a-service models, B2B services, and services for individuals and households. Finally, for the analysis, the sectors of, electronics, packaging, and chemicals were grouped under “Industry” . The sectors’ groupings are shown in Table 2.

Considering the foregoing, from a general overview of the 43 collected good GPs: 22 falls within the “End-of-Use” sector; 7 pertain to “Education and Awareness” activities; 2 relate to the “Services” sector; 7 represent innovative solutions, and 5 fall under the category of “Industry.”

4. RESULTS AND DISCUSSION

The objective of this section is to analyze the selected GPs related to EEE in the ECESP, focusing on their place-

TABLE 2: List of sectors’ groupings (source: Authors’ elaboration).

Groupings	Sectors
End-of-Use	Waste Management and secondary raw materials Separate collection Recycling Repair and reuse
Education and Awareness	Social enterprise Education Consumer empowerment
Innovative solutions	Eco-design Research Technology
Services	Energy and public services Product-as-a-service Business-to-Business (B2B) services Personal and households’ services
Industry	Electronics Packaging Chemicals

ment along the life cycle phase and the sector of the organizations that submitted them. This allows us to understand how European countries are progressing towards a circular transition where all phases of a product’s life cycle must be considered, both by producers and consumers. In order to have an idea of the geographical distribution of GPs in Europe, in Figure 2 is depicted a map that shows the number of GPs about EEE for country founded in ECESP.

In Europe, most of the GPs refer to the Central Europe area while it can be observed that Eastern European countries have fewer GPs (with the exception of Romania). In particular, the higher number of projects for country regarding the CE for EEE sector is 6 and it is found to be in Spain,

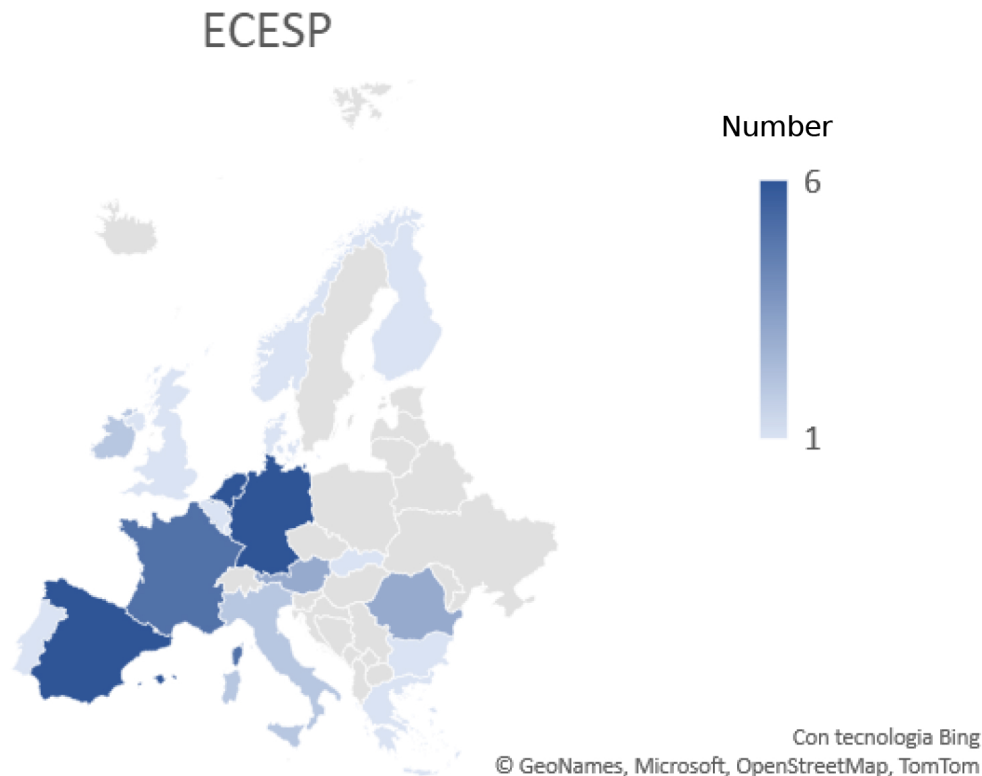


FIGURE 2: Distribution of good practices for country (source: Authors’ elaboration from data).

Germany and Netherlands. Then, there is France with 5 GPs followed by Romania and Austria with 3. Italy and Ireland present 2 GPs on the ECESP platform. Finally, it is found that Bulgaria, Denmark, Finland, Greece, Luxembourg, Norway, Portugal, Belgium and Slovakia have just 1 GP regarding EEE on ECESP.

From the life cycle phase point of view the most populated phase are Waste Management and Consumption with 14 out of 43 GPs, followed by Production with 6 GPs, Secondary Raw Materials with 5 GPs and finally Innovation and Investments with 4 GPs. This result highlights that from an empirical perspective ECESP-based and focused on the EEE value-chain the majority of the CE GP are still engaged in the final stage of the product life cycle, mostly regarding its recycling or disposal. Similarly, but in a more pronounced manner, the sector with the highest number of GPs is End-of-Use, with 22 out of 43 GPs. This indicates that half of the organizations that submitted GPs operate in recycling or repair and reuse activities. Following the End-of-Use sector, Education and Awareness, as well as Innovative Solutions, each account for 7 GPs, while Services have 5 GPs, and Industry has 2 GPs.

The lower representation of GPs in the Industry sector could be attributed to the focus of the paper on EEEs and WEEEs, which are not typically associated with sectors such as Packaging or Chemicals (see the definition in paragraph 3.2). The dominance of Waste Management and Consumption as the most common life cycle phase, coupled with the fact that half of the projects belong to the End-of-Use sector, suggests that the current system remains still rooted in a traditional business model. Of course, this is a partial picture concerning the projects found on the ECESP related to EEE and their waste. The fact that this is the specific outcome for this value chain could actually be a consequence of the introduction of EPR and Extended Consumer

Responsibility, as well as the increasing pressure from the EU regarding the collection of WEEE.

To cross-reference the information by life cycle phase and sector, or in other words, to represent how the sectors are distributed across the various life cycle phases and vice versa, the 43 selected projects in ECESP can be represented in a matrix form, using a bubble chart. Specifically, the classification according to the life cycle phase is represented on the ordinate axis, while the sector of the submitting organization is on the abscissa axis (Figure 3). The sizes of the circles in the graph are directly proportional to the number of GPs associated with the specific combination of phase and sector, with the corresponding value indicated within the respective circle. The most populated areas in the Cartesian space (sector, phase) have been highlighted in yellow.

By looking at the most populated area in the plot depicted in Figure 3, it is possible to observe that the majority of GPs found are situated in the Waste Management phase of the life cycle and in the End-of-Use sectors populated by 8 out of 43 GPs. The projects identified in this area of the plot aim to achieve efficient processes to valorize resources. The goal is to increase the collection of WEEE through technologies for sorting, separation, treatment, recovery, and recycling of materials. Proper recycling permits to valorize materials that can be reintroduced into the production cycle. Examples of this projects include the “Kujala waste symbiosis” in the Finnish region of Lahti and the “Transforming waste into resources” by the Romanian Green Group Holding. The first wants to increase recycling and reduce the amount of waste used for energy production. The mechanical sorting plant constructed for this project can process up to 65,000 tons of mixed waste, waste for energy production, and construction waste per year. From these types of waste, the plant separates fibers,

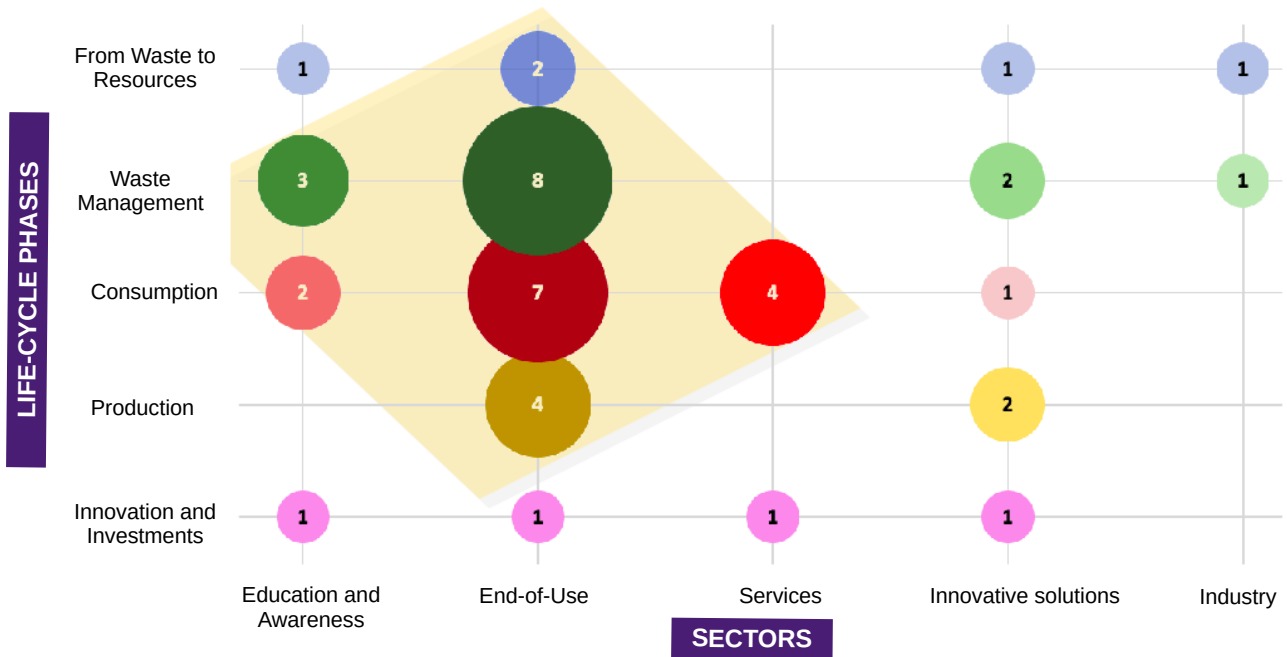


FIGURE 3: Distribution of good practices across Life cycle Phases and Sectors (source: Authors’ elaboration from data).

plastics, and metals for recycling. In this case, WEEE are just one kind of materials treated. This is not the case for the second project cited since GREENWEEE International is an integrated treatment plant for WEEE that operates under a WEEELABEX license for large domestic appliances, small appliances, cathode ray tubes, and CFA workflows.

Second in the list of the most populated area is the one that interest the Consumption phase versus still the End-of-Use sector populated by 7 out of 43 total GPs. The projects regarding the phase Consumption and falling within the End-of-Use sector mainly involve the distribution of surplus non-food products or the purchase of defective products, which are then refurbished and sold at a lower price. For example, Fairmittlerei (Fair Mediation, in German) manages a network of donor companies and manages their surplus products (including EEEs), mediates them, and delivers them to NGOs throughout Austria, creating a logistic and financial advantageous situation for all parties involved. Another project in this area is RECOSI based in Ireland, which sells high-quality refurbished laptops, desktops, and IT equipment sourced from global companies and government departments across Europe.

It is interesting to observe that while Consumption is the second most populated phase, the same is not the case for the Services sector, yet it still accounts for 4 out of its 5 GPs being applied in the Consumption phase and the other in Innovation and Investments. This highlights that nearly all organizations (80%) within the Services sector that submit a GP about WEEE and EEE are involved in projects centered around the Consumption phase. This distribution sets apart the subset of GPs within the Services phase from the complete sample and even from the other subsets studied, as Waste Management, which is one of the primary aspects in other cases, is notably absent here. This data shows the emergence of a new consumption paradigm not based on product ownership but on services, thus decoupling possession from ownership. In this area, some GPs involve the rental service of technological items, while others are projects aimed at increasing the adoption of a circular model. An example is the leasing system Commown, where subscribers pay a fixed monthly fee to use the phone but not to own it, providing users with mobile phones without having to worry about repairability, durability, or end-of-life issues. In case of hardware problems, Commown provides its subscribers with quick replacements and independently repairs individual modules or disposes of broken parts using Fairphone's retrieval system. Or another is the case of AIMPLAS that coordinates the C-SERVEES project with the aim of promoting a more efficient CE in the use of resources in the EEE supply chain, by developing new circular business models.

The Production phase has the majority of GPs involved in a single sector: End-of-Use (4 out of 6) while the other 2 belong to the sector Innovative solutions. The 2 GPs in the Innovative Solutions sector refer to the innovative use of recycled plastics in the EEE components or to effort in designing objects that are easier to dismantle, coupled with the development of new commercial models for their introduction to the market, as in the paradigmatic case of Fairphone. In fact, Fairphone aims to create a positive

social and environmental impact throughout the life cycle of a phone. In addition to using fair materials and offering good working conditions, this includes long-lasting design; reuse and recycling (by making phones easier to dismantle); product-as-a-service. Regarding the End-of-Use sector in the Production phase, there is a greater focus on the regeneration and repair of products such as printer cartridges, computers, and audio devices. Across all these circular practices, emphasis is placed on the materials used and the fight against planned obsolescence, implemented through repair services, sometimes even with perpetual duration.

In ECESP, for the Innovation and investments phase the GPs are distributed across different sectors. Specifically, the GPs involve the development of technologies for recovering plastic from End-of-Use WEEE; innovative solutions for building CE for the "Data Centre" industry; management and recovery services of CRMs from electronic devices and photovoltaic waste; and education and awareness projects such as "the WEEE caravan" to engage schools and local communities.

Regarding the GPs in the Secondary Raw Materials phase, they mostly consist of sharing platforms for the reuse, repair, or collection of WEEE. The other GPs involve the reuse of cardboard packaging for EEE, such as the practice carried out by the Elak Electronics webshop, or the paradigmatic case of Miele washing machines, whose components are disassembled and sent to foundries to be reused due to the well-known composition of metals. This allows us to observe that, at least from what has been shared on the analyzed platform in Europe, there is a focus on End-of-Use management, albeit aimed at the development of innovative digital solutions and citizen education and awareness.

Finally, in the Innovative Solutions sector, there are GPs that refer to the application and use of "Big Data", the creation of "smart bins", and innovative products in processes or materials that ultimately move towards eco-design. The products of the last two mentioned GPs are a watch and a mobile phone. The watch, designed and produced by "Circular Clockworks", is based on the efficient use of secondary raw materials mainly from WEEE, while also addressing End-of-Use management. The mobile phone is the aforementioned Fairphone. The distribution across the different phases of the life cycle is quite homogeneous; in fact, all phases have at least one GP. Specifically, the two phases of the life cycle with the highest representativeness are Waste Management and Production (both representing 29%). The other phases of the life cycle (Consumption, Innovation and Investments, Secondary Raw Materials) all have the same share, which is 14%. This latest finding is noteworthy as it suggests a growing consideration for all phases of a product's life cycle in innovative solutions. Although the attention remains relatively minimal compared to the End-of-Use sector, it signals a step forward towards a circular transition of the European economic model.

5. CONCLUSIONS

The national and international promotion of the development and the spreading of GPs has become a necessary

policy instrument for the transition toward circular models.

ECESP contributes to the collection, mapping and sharing of CE European circular economy GPs. This paper identifies virtuous examples related to the real application of circularity and gives an overview of the state of the art and level of maturity of the models applied to GPs for EEEs in Europe, highlighting the limits and barriers on which future activities should focus.

The analyzed data reveals a notable trend: the majority of GPs identified focus on the Waste Management and Consumption phases of a product's life cycle. Additionally, within the sectors of reference, the End-of-Use sector stands out with the highest number of GPs, addressing activities such as recycling, separate collection, and reconditioning. This observation highlights a critical finding in the analysis of CE practices within the EEE sector: a predominant emphasis on the latter stages of a product's life cycle. The fact that this outcome pertains to EEEs and WEEEs likely indicates how European policies related to EPR and targets for WEEE collection are driving actions aimed at enhancing efficiency in this sector and key area. Moreover, it depicts a scenario still oriented towards waste management or recycling rather than waste prevention. Specifically, initiatives related to the Innovation and Investments phase, which encompasses eco-design and the initial stages of the production process, are notably scarce.

Meanwhile, the sector of Innovative solutions is quiet homogeneously distributed among all the life cycle phases. This at least suggests a growing consideration for developing new solutions in all the value chain of a product.

On the other hand, from the analysis of the Services sector, it was found that the phase it mostly focuses on is Consumption. This highlights the emergence of new business models no longer based on ownership but on services, thus decoupling possession from ownership. This is evident in various iterations of the CE model, such as the sharing economy or eco-leasing.

Finally, based on the findings, a key point that emerges is that the dissemination of GPs related to the EEE sector at the European level is still relatively limited. This is evidenced by the low number of GPs found on the ECESP platform.

In conclusion, this study provides a preliminary analysis that contextualizes the current state-of-the-art regarding the implementation of GPs in the CE for EEEs across various sectors and life cycle phases. Further investigation should prioritize examining the organizations implementing these GPs, the types of funding utilized, the levels of applicability, and the geographical distribution across Europe. By delving deeper into these aspects, a more comprehensive understanding of the challenges and opportunities in advancing circular economy principles within the EEE sector can be developed, which will serve as the focus of future research works.

Moreover, several potential future developments warrant consideration. One avenue for exploration involves comparing the findings for EEE sector with those of other sectors of interest. Another avenue entails comparing them with other equivalent national platforms in European countries, such as the ICESP. These comparative analyses

could yield valuable insights into the effectiveness and adaptability of circular economy initiatives across different sectors and regions.

ACKNOWLEDGEMENTS

The authors thank the Erion WEEE organization for the grant provided for the "Training for Circularity" project. This work is indeed the result of that journey, which, through education and research conducted during a training period at the Enea Casaccia Research Center, aims to promote circularity in the WEEE sector.

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TOWARDS AN INFORMATIONAL UPCYCLING: LEVERAGING COMPUTATION TO (RE)DESIGN BIOMATERIALS' LIFE CYCLES

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Article Info:

Received:
4 May 2024
Revised:
23 July 2024
Accepted:
2 August 2024
Available online:
7 September 2024

Keywords:

Informational Upcycling
Technological Design
Computational design
Additive Manufacturing
Wood waste

ABSTRACT

As the imperative to mitigate climate-altering emissions and curb natural resources exploitation intensifies, the fields of architecture and industrial design are compelled to explore alternative material practices. This imperative requires a transition from the traditional cradle-to-grave model to a cradle-to-cradle design paradigm, emphasizing upcycling, the process of repurposing waste into higher-value products. However, the definition of “value” in this context remains vague. In this paper, we posit value as an informational shift, denoting the “ecological intelligence” embedded within recycled materials compared to their prior life cycles. Amidst the concurrent ecological and digital transitions, the concept of “Informational Upcycling” (IU) emerges, bridging Circular Economy principles with the creative potential offered by digital technologies and Artificial Intelligence (AI) in architectural and technological design. IU leverages computational methods to address the complexity, non-linearity, and unpredictability inherent in reclaimed materials, which pose challenges to their reuse and upcycling, thereby questioning established design methodologies and tools. This conceptual framework is informed by a case study analysis of international critical research practices and is articulated through a comprehensive digital workflow, pointing out the enabling technologies for IU, with a specific focus on wood waste. Finally, the paper illustrates the application of this theoretical framework through the FoRWARD (Furniture Waste for Circular Design) research project, funded under MICS – NextGenEU, PE11, Spoke 4. Led by the Department of Architecture at the University of Naples “Federico II”, this initiative aims to test this approach within the Italian furniture industry, showcasing practical implementations of IU principles.

1. INTRODUCTION

In many ways we can adjectivize the Age of Transition we are living – ecologic, digital, economic, cultural, social, etc. Despite the specificity of each single topic, the term “entangled” (Oxman, 2016) is probably the one the better expresses the complex nature of the significative environmental, technological and social transformations that are imprinting the ways we reflect about problems and, therefore, define the tool with and through which we take action.

In the field of design, and especially of Technological Design, “transition” has cognitive and technical aspects that represent an identity trait of the Discipline, that has always addressed the integration of the “new industrial means” (Crespi, 1988; Nardi, 2000) towards a sensibility for the intertwined network of links and consequences that the human action establishes with the environment, intended as an eco-socio-technical milieu (Grabowski et al., 2017). Indeed, such transformations are stressing out the established relations between design-process-product,

matter-form-tools and, eventually, humans-nature-technology (Perriccioli et al. 2020). Computational Design, Robotic Fabrication or Artificial Intelligence (AI) aren't just a new set of instruments that update, in a digital fashion, the previous (Negropone, 1969); neither they stand “neutral” towards the ways their use has an influence on the design processes and their outcomes (Russo Ermolli, 2020).

In a similar way, the principles of Circular Economy (CE) are questioning the conventional methods of design, thus assuming the aspect of technological drivers more than just indicators and checklists. «A Circular Economy» – the Ellen MacArthur Foundation states (2013) – «is a systemic approach to economic development designed to benefit businesses, society, and the environment. In contrast to the ‘take-make-waste’ linear model, a circular economy is regenerative by design and aims to gradually decouple growth from the consumption of finite resources».

This study makes an attempt to define a theoretical framework that links CE and Computational Technologies, here assumed to be intertwined, according to a “blue and

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green” paradigm for the Age of Transition (Floridi, 2020; De Wolf et al., 2024). The “cooperation” between circularity and computation is understood as a crucial step towards an “Informational Upcycling” (IU), here defined as the process of transforming or redesign waste into higher value products with an increased information content. IU is a design-driven recycling strategy that leverages computation and digital manufacturing to convey an informational shift in terms of material features, environmental performance and socio-economic sustainability, between a recycled material and its previous life cycle(s).

The conceptual outline of IU is informed by a case study analysis of international critical research practices and is developed in the form of a comprehensive digital workflow considering reuse and recycling strategies. In particular, the study is dedicated to wood waste, being timber one of the most widespread material in construction, furniture, packaging etc. and, despite its fame of “sustainable resource”, is craving for more rational and effective ways to be recovered (Yu & Fingrut, 2022).

Finally, the paper describes the upcoming application of this theoretical framework introducing the ForWARD – Furniture Waste for Circular Design research project (MICS – NextGenEU, PE11, Spoke 4), coordinated by the Department of Architecture, University of Naples “Federico II”, specifically addressed to test this Technological Design approach within the Made in Italy furniture industry. The ForWARD research aims at experimenting a sustainability relationship between technological innovation and communities, with the goal of reducing the gap between research and practice experimenting collaborative laboratories in weak territories as a driver for social and economic resilience. Specifically, in this paper the methodology for the design of new materials’ life cycles from recovered wood waste through digital enabling technologies is presented, and the future implementations are illustrated.

2. A COMPUTATIONAL FRAMEWORK FOR CIRCULAR ECONOMY

2.1 Circular Economy and Digital Enabling Technologies: policy, insights, challenges

CE and digital tools represent the backbone of the recent EU policy for the goal of a “sustainable built environment” (European Commission, 2015; 2019; 2020). This guidance advocates the adoption of the Key Enabling Technologies (KETs) – BIM, GIS, Virtual and Augmented Reality, Digital Fabrication, Blockchain, Internet of Things, Digital Twins, Artificial Intelligence (Pimponi & Porcari, 2020) – throughout the entire Architecture, Engineering, Construction and Operation (AECO) supply-chain, not only as a tool for improving the efficiency of the management of buildings, but especially in the early stage of the design process (Cays, 2017), whereas the impacts on decision and the expected benefit in terms of life-cycle are higher (Bernstein, 2018).

Nonetheless, among researchers, practitioners, firms and companies the use of KETs for CE is still underconsidered: in a previous study (Rigillo et al. 2023), it has been outlined how the AECO industry is struggling to implement digital processes within its workflows, and the topic of the

relationship between circularity and digitalization is still underinvestigated, notwithstanding the efforts the EU put in terms of protocols and legislation.

These results are explained by the different complexities of the KETs, as well as by the rooted distrust of the AECO industry towards technological innovation (Agarwal et al., 2016). This condition seems to be related specifically to the EU context, precisely because of the strict legislations emendated by the Union, while in other areas (USA, Australia, UK, China, etc.), the less constraining nature of the regulatory system appears to have favored technological and design experimentation.

One other impeding factor may lie in the very structure of the EU supply chain, where SMEs are the norm. This negatively influences the industrial development, especially in the AECO. Finally, the analysis underpins three main consequences: low capacity in natural resources and waste management; low ability to create and support supply chains; low education rate in terms of digital culture and awareness (Goran et al., 2017).

2.2 Beyond Technology: a “New Kind of Science” for Circular Economy

Among the abovementioned criticalities, the latter seems to be the most urgent, considering how fast technologies are evolving and changing in time. In this context, McKinsey already pointed out how the cultural preparedness of the AECO operators is essential, beyond the development of technological skills (McKinsey, 2017).

The cultural pervasiveness of digitalization, in fact, has touched every aspect of existence at technical, economic, social levels (Picon, 2020). In architecture and design, the digital tools form an ecosystem of innovative enabling technologies that involve Computational Design, Digital Fabrication, 3D Scan, Artificial Intelligence, etc., changing the way designers communicate and collaborate, analyze and simulate, fabricate and assemble (Tamke, 2019).

Digital technologies are already drawing a “new kind of science” (Wolfram, 2002) while they are enacting “the end of theory” as we know it (Anderson, 2008; Carpo, 2023): in front of growing sets of Big Data, the traditional scientific approaches are completely inadequate if compared to the new, non-linear, abductive and statistical methods of computational technologies. For design and architectural research, this involves a co-evolutionary approach (Attaianesi & Rigillo, 2021), both in processes and outcomes, within which the possibility of enucleating design questions in a radically different way and of reducing the informational distances between design and production are redefining the traditional relationships among the different actors involved in the process, as well as between the designer and his/her work.

This assumption is not immune to contradiction: not only for the total absence of semantics within the logics of computational intelligence (Hayles, 1999; Carpo, 2019) that poses serious problems for the un-falsifiability and un-recoverability of the “a-scientific” computational processes; but, indeed, for the only apparently “immaterial” substance of information, thus qualifying as energy inten-

sive and extractive, both in terms of data and resource mining (D'Abbraccio & Facchetti, 2021).

Beyond hopes and fears, towards this “inevitable” change (Kelly, 2017) current research is called to respond to the rising need for theories, more than tools (Russo Ermolli, 2020), asking whether digital technologies can make more sustainable and circular the transformation processes of the built environment.

2.3 Reclaimed resources as a computational material

As the acceleration in every social, economic, environmental and technological dynamic is unstoppably growing (McNeill & Engelke, 2016), the scarcity of resources and the need for reducing energy consumption and material exploitation are pushing to privilege reuse and upcycling over recycling (EMF, 2015) and new challenges are facing Technological Design. Unpredictability, uncertainty and non-linearity describe reclaimed materials' behaviors, performance and availability, hindering or impeding their use in the industries. Designing with recovered resources and their life cycles, indeed, means to be hinged with information and optimization depending on the capacity of available supplies; it also implies to deal with difference and non-standardization, where every piece is unique in size, number and features. As the “production” of reclaimed materials is not informed by the market demand, but happens depending on un-foreseeable circumstances, their stock is limited and heterogenous, thus requiring to be optimized. Being waste an unpredictable and unstable resource, digital technologies can help designers to grasp complex behaviors and anticipate problems, providing for a deeper understanding about materials and design.

In these terms, reclaimed resources can be considered as a computational material, and then requiring for computational, “post-scientific”, non-standard and non-linear tools. This approach is not entirely new. Mario Carpo already pointed out how computational practice is very similar to the artisanal one, both sustained by a “trial & error”, abductive logic (2017; 2019), not oriented to the general modeling of problems (i.e. like science does), but to “formulate of the specific” (Ahlquist & Menges, 2011). Nonetheless, the growing distance between “stand-alone”, state-of-the-art research and widespread design and craft practices poses serious questions and can be identified as an urgent field of reflection (Ryamarczyk, 2020).

3. TOWARDS AN “INFORMATIONAL UPCYCLING”: ESTABLISHING A COOPERATION BETWEEN CIRCULARITY AND COMPUTATION

3.1 Questioning the definition of “value” in upcycling

Most recycling involves converting or extracting useful materials from a product and creating a different product or material. Depending on the outcomes, recycling can be distinguished into upcycling and downcycling. Upcycling, also known as creative reuse, is the process of transform-

ing by-products, waste materials, useless, or unwanted products into new materials or products perceived to be of greater quality, such as artistic, use, economic or environmental value. Upcycling is the opposite of downcycling. Downcycling involves converting materials and products into new materials, sometimes of lesser quality (Braungart & McDonough, 2003; 2013).

In both cases, however, the notion of “value” is vague. Being the implementation of upcycling based on the difference between a recycled product and its precedent condition and considering information “a difference which makes a difference” (Bateson, 1973), in this study we address the concept of “value” as an informational shift, i.e. the difference in ecological intelligence – material features, environmental performance and socio-economic sustainability – that a recycled material or product holds compared to its previous life cycle.

We define “Informational Upcycling” (IU) as a circular design strategy aimed at the increase in information content (informatization), to which must correspond the decrease in environmental weight (dematerialization). IU leverages computation and file-to-factory techniques to actualize this informational shift, re-evolving industrial processes and re-organizing waste material properties into more complex informational structures provided with novel designed features and a reduced impact, e.g. in terms of carbon emission expressed in kg of CO₂ eq.

3.2 Informational Upcycling as an “extropic” design strategy

IU is an “extropic” design strategy, i.e. it is oriented at retaining the entropy of materials, processes and products originated by reuse, remanufacturing and recycling activities.

Entropy is a thermodynamic principle that describes how all things inexorably tend to dispel energy into randomness and chaos. Nevertheless, biology contravenes to this law through energetic processes that assemble simple elements into complex structures at multiple scales (Dade-Robertson et al., 2024). According to Kevin Kelly, technology acts in an analogue fashion. Technology (or “Technium”) autonomously evolves through sophistication, self-training and data compression from materiality to immateriality, augmenting information and hierarchy to result in complexity and organization. Borrowing from the philosopher Max More, Kelly calls this phenomenon “extropy” (Kelly, 2010).

This definition of “extropy” suits the concept of IU for both organic- and inorganic-derived waste materials. Nevertheless, in this paper we focus on organic materials, as wood, which pose a more problematic questions to be tackled: how can we upcycle waste deriving from informationally complex living entities?

Dade-Robertson et al. pose this question and make a distinction between organic and inorganic manufacturing. Modern fabrication methods involving biologically derived materials are, according to the Authors, unwittingly engaging in an “entropic manufacture”: «we spend energy to convert highly structured complex materials into simpler,

more disordered materials before using more energy to assemble them into composites across often geographically distributed manufacturing chains. Consider the journey of manufacturing paper. A tree, which is a complex multi-material object - a sophisticated organization of cellulose, hemicellulose, and lignin finely tuned and organized at the microscopic scale to yield functionally graded materials from the trunk to individual leaves - obtains nutrients and energy from its environment, self-assembles, is responsive, and can produce and distribute information for new trees through its seeds. To make paper, we first undergo a series of disassembly steps [...]. While paper is functionally more useful for writing a letter than a living tree, this approach, involving building through a process of disassembly, seems counterintuitive» (Dade-Robertson et al., *ibidem*).

In the case of timber waste, indeed, the most common recycling strategies contemplate entropic manufacturing by downcycling, i.e. recycling to produce low-quality products (such as particle boards) or incineration to generate energy. Particle boards, a common product from wood waste downcycling, hold significant impacts in the transportation, manufacturing and end-of-life stages. Their production involves the use of chemical or bioderived additives. Once disposed, they are further downcycled, landfilled or incinerated. In any case, the “value” of wood gets scattered throughout energy-consuming processes and its life cycle decreased, at a growing environmental cost (Campbell-Johnson et al., 2020). In this way: «[With] our manufacturing methods [we] impose disorder and randomness on our materials. We accelerate entropy to give the illusion of order in our constructed environment» (Dade-Robertson et al., *ibidem*) (Figure 1).

3.3 Informational Upcycling actualization. Using computation to (re)design material intelligence

To be effectively “extropic”, the actualization of IU requires to regeneratively design new ecological intelligence in reuse or recycle processes and products. The result is an informational shift between the recycled product and its previous condition or life cycle, realized by:

1. increasing the information content (i.e. the informational difference between life cycles);

2. reducing the environmental weight (i.e. the environmental difference between life cycles).

Defining actualization as “the process of bringing something into existence”, computation is the elective means for the actualization of IU. In the words of Achim Menges, computation is: «The processing of information and interactions between elements which constitute a specific environment. Computation produces outcomes realized from the processing of internal and external properties. It provides a framework for negotiating and influencing the interrelation of datasets of information, with the capacity to generate complex order, form and structures. Computation as a design methodology is to formulate the specific. Computational processes start with elemental properties and generative rules to end with information which derives form as a dynamic system» (Menges & Ahlquist, 2011).

Computation rediscusses the relationship between design, matter and production by introducing information as a “value” in the materialization process. Actualized by computation, IU produces an informational shift by “evolving” already crafted techniques, objects or materials.

As for biological organisms, Kelly claims that entropic evolution-through-information is an inherent trait of technology itself (Kelly, *ibidem*). Describing the shift in construction techniques from an artisanal to an industrial economy, we can compare a simple clay brick and a complex ceramic façade component by the difference in the information structure and content (Campioli, 1992). In the first case, we have the lowest level of information: a brick only contains basic “data” about material and dimensions. We have (almost) unlimited range of possibilities – we can build walls, cladding, vaults – exactly because we have no direct instructions from the product or the material itself. As Louis Kahn, we need to ask the brick “what does it want to be”, that is, any possibility can only be actualized through design, but design itself only happens after the brick. Shifting into an industrial paradigm, the same material is employed to design, fabricate and assembly a ceramic façade component. Here, we have a higher level of information. Material is shaped by design and so the product contains all the instructions the builder needs to assembly it, provid-

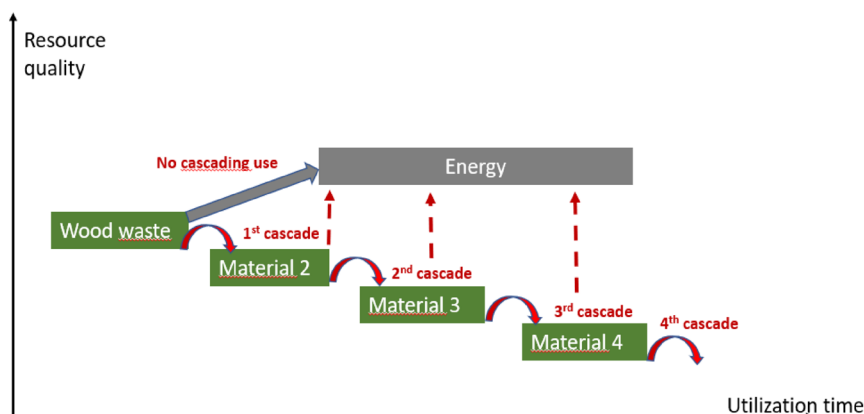


FIGURE 1: Cascading effects of wood waste. Source: <https://www.mdpi.com/2073-4360/13/11/1752>.

ed by design. Even if standardized, such a component can be specialized to specific uses (for instance, incorporating sun-shading lamellas with a specific orientation depending on the average site conditions) and positioning (e.g. base or top level).

In our post-industrial, file-to-factory manufacturing context, the balance between information and matters tends to a state of poise. Taking a 3D printed clay component, there is no loss of information between matter and design, the shape of the objects is unique and totally derived from the design. The content of information is at its maximum, the shape is optimized and through the process from artisan to industry to digital we get more immaterial content and less material weigh. Indeed, the object is directly shaped to the information of design, and the material can be additivated or modified, i.e. “genetically” modified in shape and very structure for specific, unique design intent.

It is worth noting how information (conveyed as a sum of signals and components) increases from the brick to the façade component. In the 3D printed clay object, however, the number of components decreases again. Dematerialization, just as information, is a consequence of extropy determined by computation.

4. DIGITAL OPPORTUNITIES FOR AN INFORMATIONAL UPCYCLING. AN IN-DEPTH IN THE WOOD INDUSTRY

4.1 Methodology and limitations

In this section, the concept of IU is actualized through a case study analysis of international critical research prac-

tices, collected and reviewed to illustrate opportunities and challenges of the theoretical assumptions abovementioned.

The in-depth analysis is dedicated to wood waste: timber is not only one of the most widespread materials used in construction, furniture and packaging, but it is a renewable and carbon storing resource, the use of which is strongly promoted by the EU Green Deal. Nonetheless, there is a scenario of rising demand for wood products (FAO, 2018; Gresham, 2020), that also implies a growing amount of waste will be generated during the construction, refurbishment and demolition phases. Nevertheless, at current state wood waste is usually downcycled or incinerated and used as fossil fuel, thus requiring more efficient ways to be recycled and reused (Beyer et al., 2011; Cazzaniga, 2022) (Figure 2).

The case study collection takes into exam major research centers working with digital enabling technologies for wood waste reclaiming and upcycling. The examples are selected for their representativeness regarding the topics of this dissertation. Further studies and future implementations of this will broaden both the number of case studies and the typologies of debris and materials to be considered.

In the first sub-section, this analysis is developed in the form of a comprehensive digital workflow for the reuse of reclaimed wood. The steps here included go from the use of laser scanning technologies for the creation of a digital library of wood debris component, to the implementation of automated AI e robotic methods for wood pieces performance analysis and assembly, depending

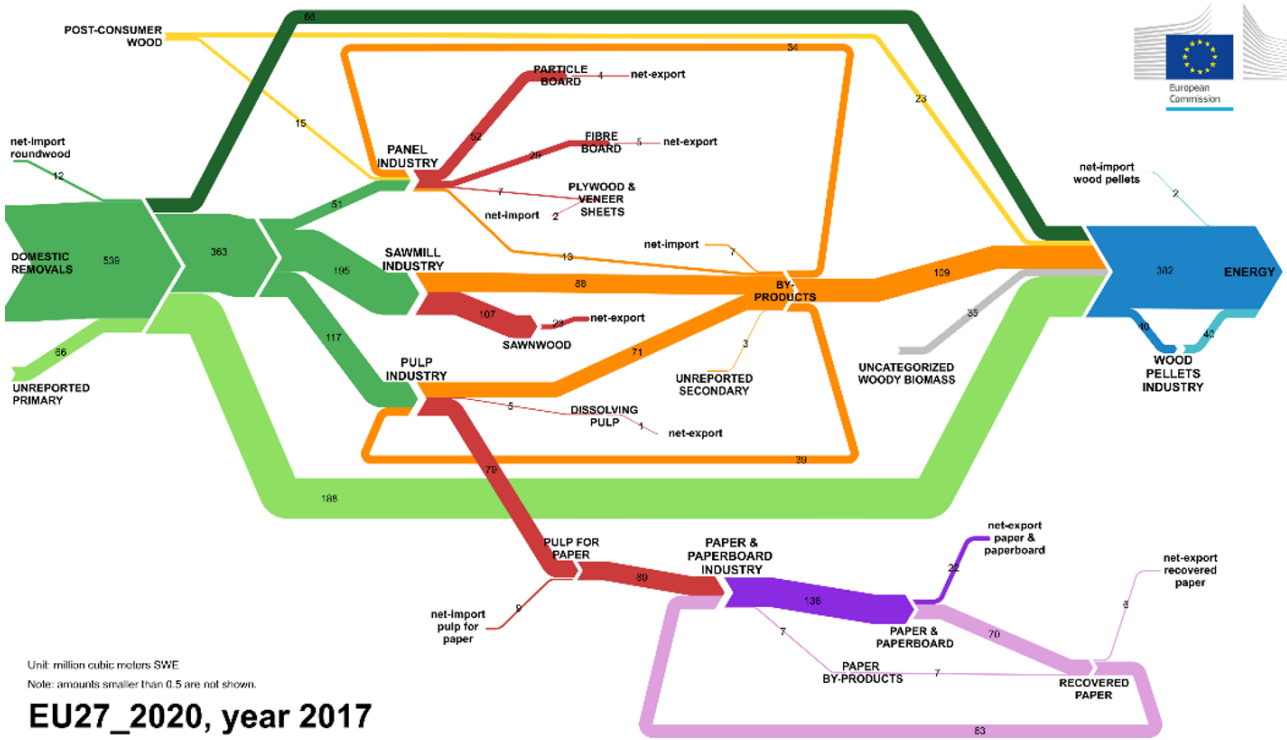


FIGURE 2: Interactive Sankey diagrams of woody biomass flows in the EU and Member States (source: https://knowledge4policy.ec.europa.eu/visualisation/interactive-sankey-diagrams-woody-biomass-flows-eu-member-states_en).

on the specific quality and quantity of each single component.

In the second sub-section, the paper examines the up-cycling of wood waste through digital and robotic fabrication. These applications are aimed at defining new materials from recycled wood that can demonstrate additional or completely different performance than wood itself.

4.2 A conceptual workflow for the reuse of reclaimed wood

4.2.1 Laser scanning and creation of a digital debris library

Working with wood waste often involves dealing with a highly diverse range of materials. In such cases, having a structured dataset that catalogues all the components, organized by size, quality, and conservation, can be instrumental in guiding designers to optimize conceptualization and anticipate issues that may arise during the assembly phase.

The work conducted by Yu and Fingrut at the UCL Bartlett School (Yu & Fingrut, 2022) offers valuable insights into the role of digital scanning technologies for controlling and classifying waste materials. In their research, the authors explored the development of a wood library, encompassing both regular and irregular forms, through the 3D scanning of waste, underused, and off-cut wood. This database was enriched with the acquisition of a digital 3D model, capturing relevant parameters and physical characteristics such as material texture. Additionally, a materials database was established, cross-referencing information with the corresponding models (Figures 3, 4).

4.2.2 Materials inspection and selection via AI and Machine Learning

Research has substantiated that 3D scanning can complete material recovery and establish a material library.

However, 3D scanning has no means to detect defects within the material, and some materials still require having their mechanical properties tested in addition to the basic inspection.

In this context, the application developed by The Living with the Princeton Embodied Computation Lab delved into leveraging Artificial Intelligence for instructing computers in identifying knots in wood waste components, sourced from disused provisional structures and scaffolding (Benjamin, 2018; Gerfen, 2018). The researchers established a user-friendly website where individuals could discern and annotate whether a given image of wood exhibited knots.

Upon inputting this human-labeled data into a computer system, the Machine Learning algorithm demonstrated its capability, even in the preliminary phase, to discern the

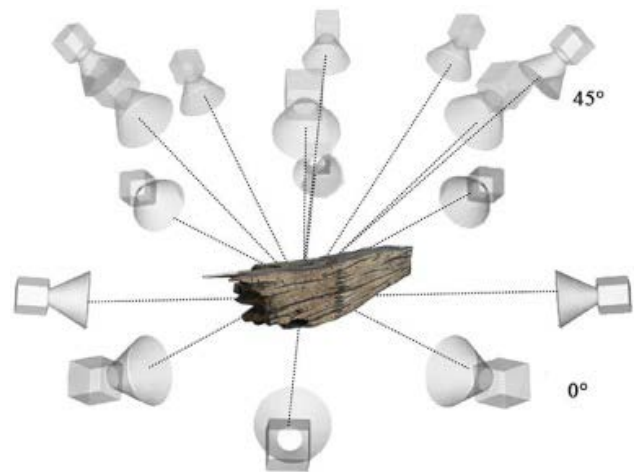


FIGURE 4a: Wood debris scanning (source: <https://www.sciencedirect.com/science/article/pii/S0921344922004025>).

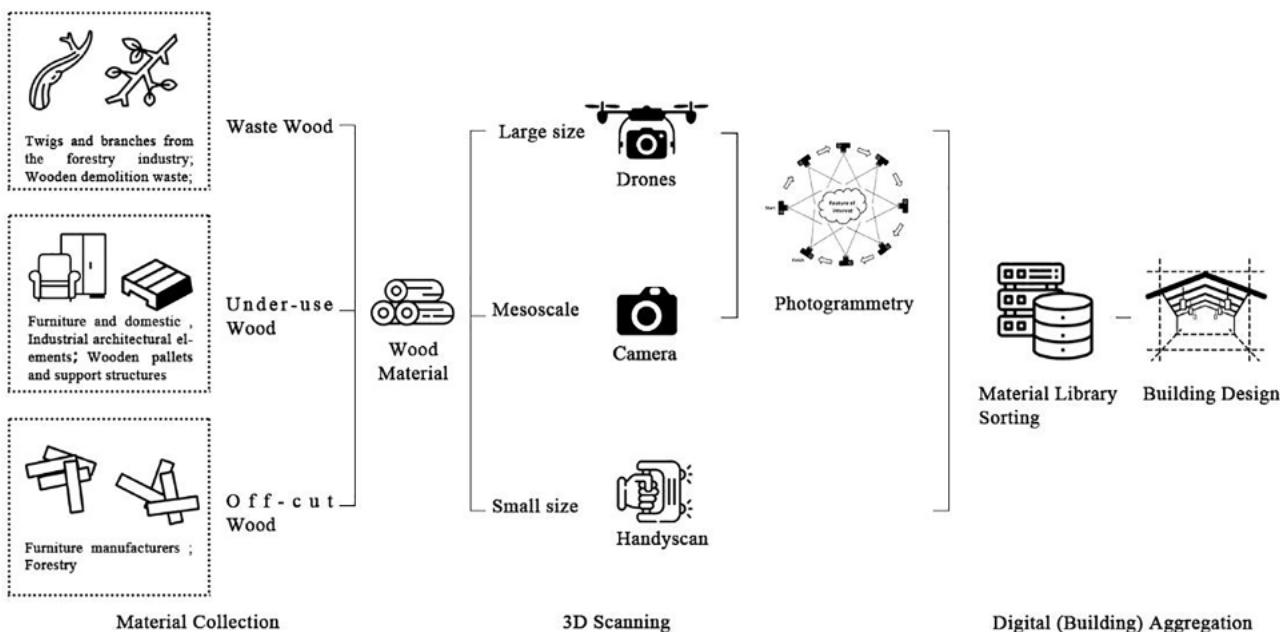


FIGURE 3: Method and workflow adopted by Yu and Fingrut (source: <https://www.sciencedirect.com/science/article/pii/S0921344922004025>).

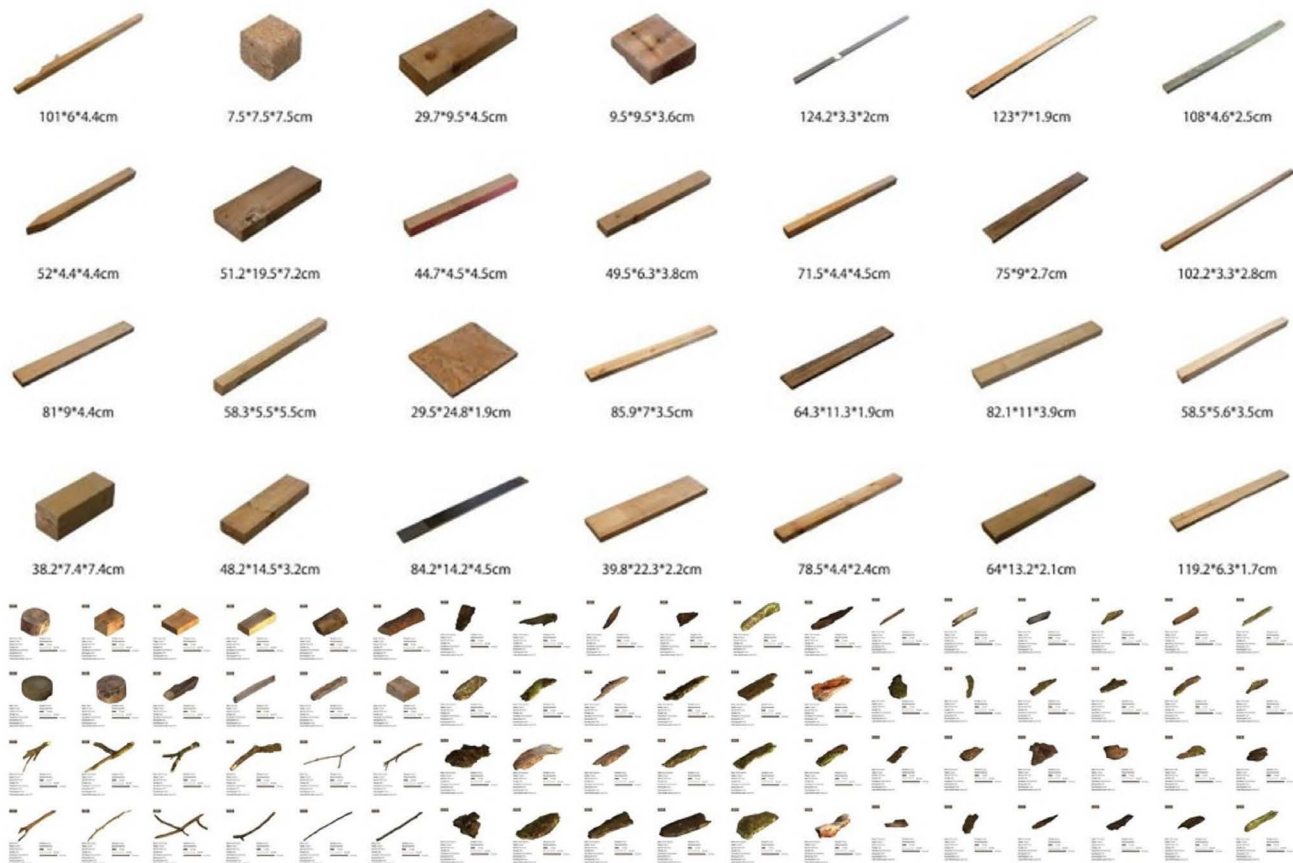


FIGURE 4b: Regular and irregular material salvaged components digital library (source: <https://www.sciencedirect.com/science/article/pii/S0921344922004025>).

presence and location of knots. Subsequently, the identified knot locations from the computer were translated into files intended for a CNC machine tailored specifically for the fabrication of such components (Figure 5).

4.2.3 Robotic crafting and assembly

Robotic fabrication is a long-studied research track at the IAAC – Institute for Advanced Architecture of Catalonia and is essential to build and craft structures digitally designed, where tolerances are lowered to the minimum and human error can put into risk the project.

In the Sourced Wood (IAAC, 2018) project, all the steps previously described (3D scan, digital library, material inspection and selection) are finalized with the fabrication and assembly with the use of robotic arms. The project is aimed to utilize waste wood in constructions and reshape them as falsework structures (Figures 6,7).

4.3 Expanding wood waste upcycling possibilities through Additive Manufacturing

4.3.1 Redesigning wasted objects with recycled wood fibers-infilled biomaterials

The “Planetary Boundaries Exhibition: Bio-Local Advanced Manufacturing,” is the result of the research carried by the Center for Information Technology in Architecture (CITA) at the Royal Danish Academy. The project aims to implement a small-scale manufacturing process utilizing

locally sourced biomaterials in Denmark (Thomsen et al., 2022). An innovative robotic spraying technique uses a biopolymer derived from bone glue collagen (an animal industry waste itself) and wood waste fibers locally sourced. The application is both addressed to produce small architectural components and furniture, but its use could be broadened to the re-design and re-fabrication of damaged pieces (Figure 8).

4.3.2 Wood waste-filled biopolymers printing

CITA employs an analog material in the robotic printing process to produce large-scale 3D printed biopolymer elements with diverse material properties, allowing adaptation to distinct performance criteria associated with design, environmental factors, and boundary conditions (Nicholas et al., 2023). The design and fabrication of these elements necessitate a close integration of material specifications with local geometry descriptions and robotic control code. This interdependence precludes the application of conventional 3D printing methods such as slicing and boundary representation-based modeling. Instead, there is a demand for approaches facilitating the local and volumetric storage, querying, and manipulation of design, material, and contextual information through generative, parametric, and fabrication workflows.

To achieve this, a compositional pattern-producing network (CPPN) is employed to generate geometries. These

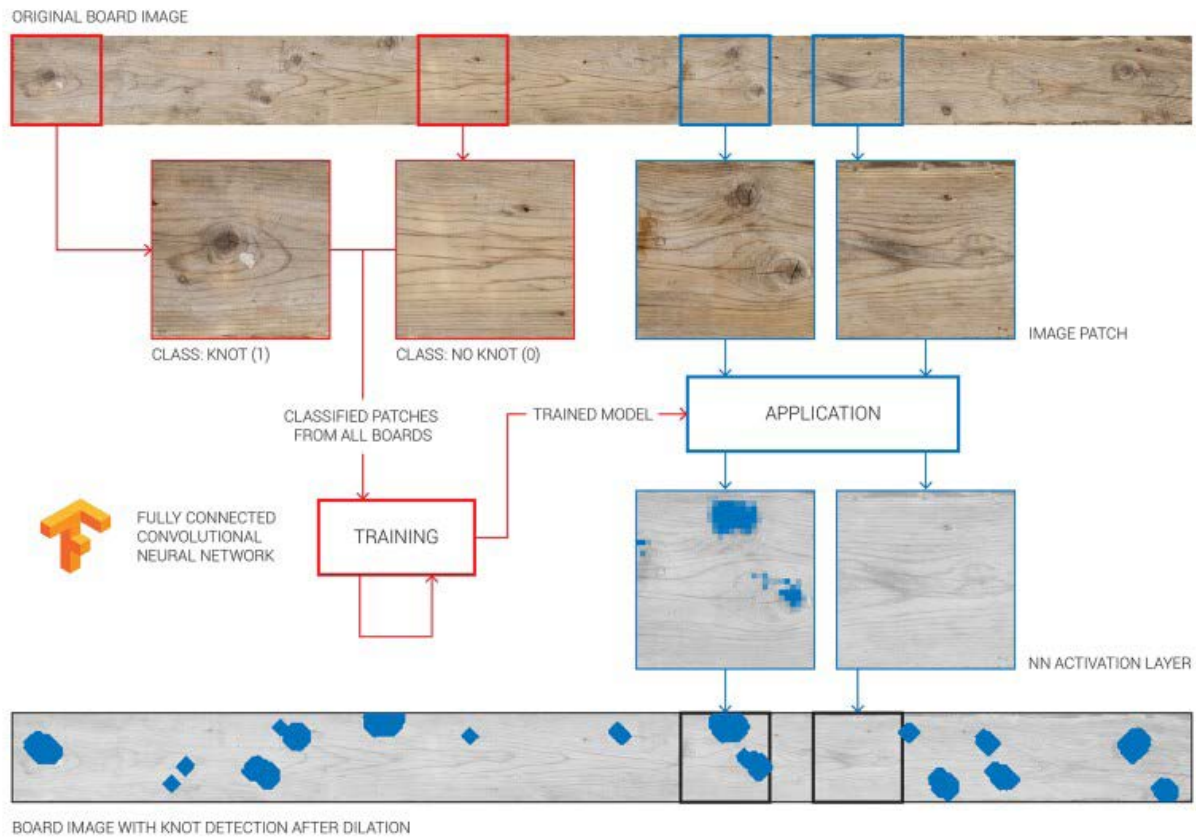


FIGURE 5: Machine Learning application for reclaimed wood knot automated detection (source: https://www.architectmagazine.com/project-gallery/embodyed-computation-lab_o).

Robotic fabrication design proposal

To develop scrap wood lattice structures that can support most construction surfaces



Upcycling :
extending the lifecycle of wood on construction site

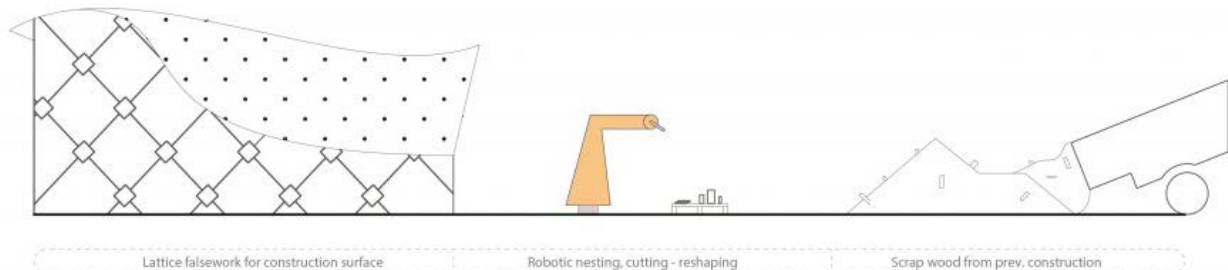


FIGURE 6: Sourced Wood robotic fabrication design proposal (source: <https://www.iaacblog.com/programs/sourced-wood/>).

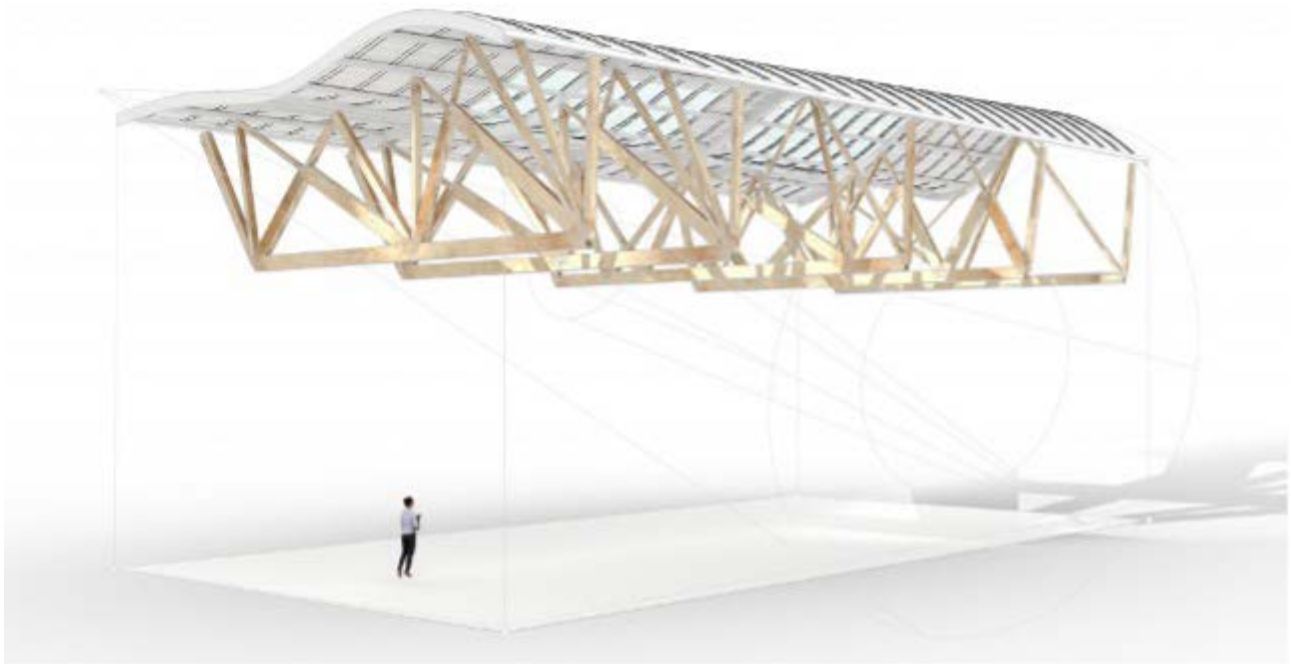


FIGURE 7: Sourced Wood robotic structural design vision (source: <https://www.iaacblog.com/programs/sourced-wood/>).

generated geometries undergo performance testing, and the outcomes inform subsequent iterations of the design process.

The efficacy of this workflow has been validated through the development of a customizable 3D-printed biopolymer-composite paneling system known as “Rad-icant”. This system, a built demonstrator measuring 7 × 5 m, was exhibited at the Aedes Gallery in Berlin during 2022/23. The “Rad-icant” collagen glue wall serves as a bespoke wall-paneling system, constructed from a biopolymer composite reinforced with various cellulose types sourced from waste products (Figure 9).

5. RESULTS DISCUSSION: ENABLING TECHNOLOGIES AND DESIGN CONSEQUENCES FOR AN IU APPROACH

The analyzed case studies allow to identify the enabling technologies for an IU approach to wood waste upcycling. Although separately reporting reuse and recycling, these two workflows can be intended as inherently linked as the components that can't be no longer reused (e.g. due to their loss of performance throughout the life cycles) can undergo recycling, thus minimizing the need for virgin resources.

Both in case of reuse or recycling, it emerges how the

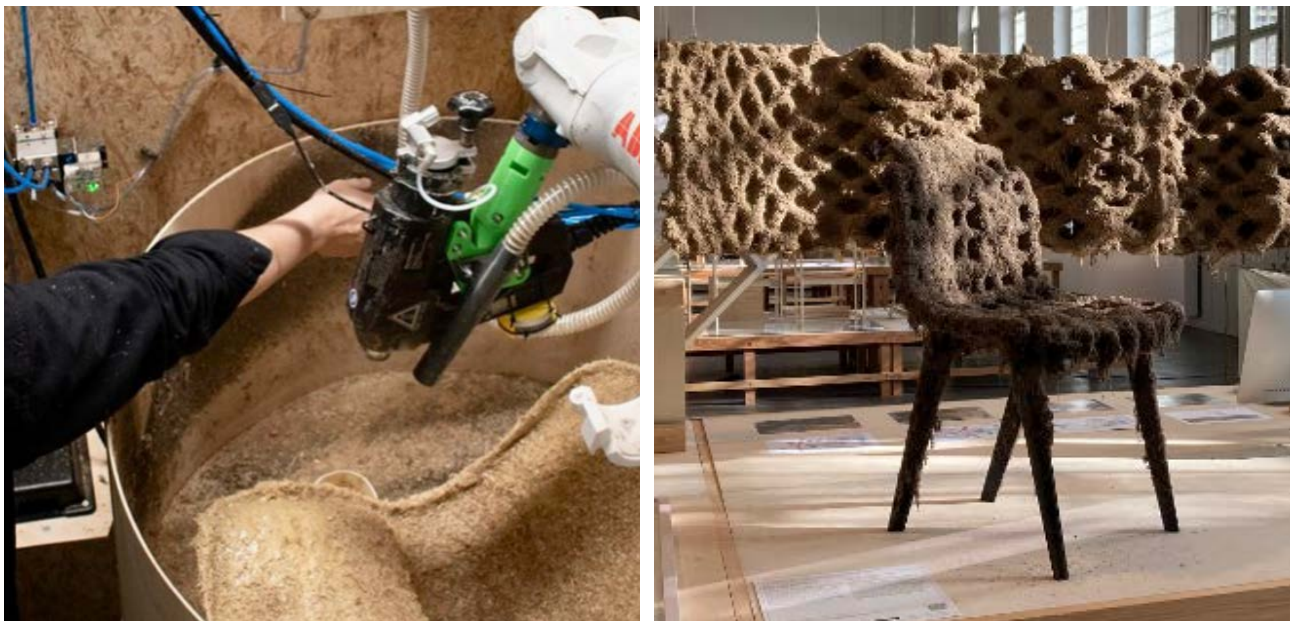


FIGURE 8: Process and prototyping of a “3D sprayed” chair, developed at Center for IT in Architecture, Royal Danish Academy, Copenhagen (DK).

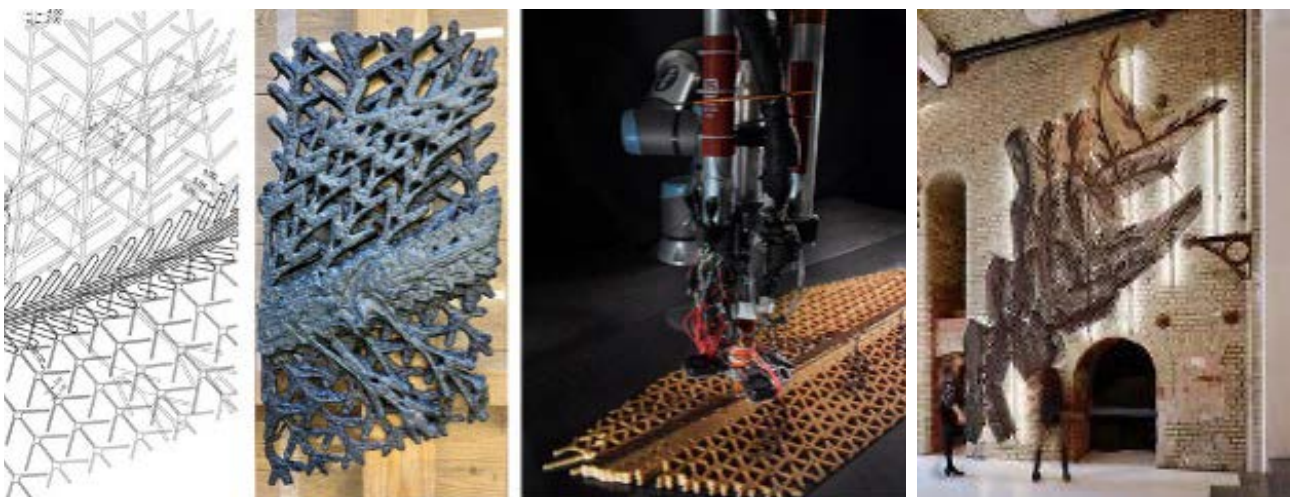


FIGURE 9: Printing and outcome of the “Radicant”, developed at Center for IT in Architecture, Royal Danish Academy, Copenhagen (DK).

use of file-to-factory technologies is essential to transmit, amplify and keep the informational content of waste through upcycling within the recycled objects. 3D scanning technologies allow to record dimensional and physical features of the waste components; AI and ML integrated inspection tool are helpful in the automation of material verification and in the supporting the definition of the reuse of recycling strategies; advanced design and fabrication technologies open opportunities for novel material organization, that couldn't be otherwise pursued.

Nonetheless, it must be underlined how the presented applications are still far from becoming replicable best practices. The reasons behind this gap between the laboratory scale and the industrial readiness can be traced in the lack of advanced expertise among the operators and the high time and cost rates of these experimentations. In

the first case it is likely to imagine the gap filled by the next generation of practitioners, that are currently gaining these new competences. In the second, it emerges how this kind of experimentation lies on a radically different conception of building and design production supply chain, entirely based on digital workflows and integrated information exchange protocols, throughout all the buildings or components life cycle, between architects, engineers, clients, supplier, producers and maintenance operators. All these enabling technologies can work in one digital ecosystem, thus reducing the information loss due to data exchange and allowing for a greater quality control.

Compared to the existing options for wood waste recycling, such as panel boards production, the presented application demonstrate how an IU approach guarantees much more design freedom and production possibilities.

Indeed, it emerges how pursuing IU means to rediscuss traditional design paradigms towards a new design concept, in which material features, formal investigation and fabrication techniques are now sequential phases of the design process, but rather different dimensions of the same design moment.

The unpredictability of waste materials can leave place to a more controlled material definition, in which matter itself is programmed upon specific design intentions; or otherwise, design can be lead in a generative way by the material features and inner “tendency”. At the same time, fabrication can be customized and adapted according to different material processes or can provide pivotal feedback regarding the design possibilities offered by the material. In the case of IAAC or The Living, the availability of components and its features is a critical driver for the whole design process, but at the same time the use of robotic fabrication allows to customize every single piece to be adapted to a specific design intent.

In the case of CITA’s bioprinting, the balance between recycled wood and binder is defined by the printing possibility provided by the 3D printer, which is in turn the result of the customization of an universal collaborative robot. These linked parameters (material formulation and machine features) are both design driving, informing the design process according to the production capacity, and yet design driven, as they can be still modified according to specific design need.

From both cases, it emerges an unprecedented complexity that can be only successfully handled by using computational technologies, and that can lead to an unexperienced design freedom.

6. TESTING INFORMATIONAL UPCYCLING IN THE MADE IN ITALY INDUSTRY: THE FORWARD RESEARCH PROJECT

The theoretical framework and the conceptual workflows developed in this paper is currently under testing within the FoRWARD – Furniture Waste for Circular Design research projects. The research is aimed at implementing circular supply chains within the wood furniture SMEs, combining digitally-enabled technological advancement, knowledge transfer and clustering activities and educational initiatives.

Wood waste management represents a fundamental issue in the furniture industry in the wake of the transition to a CE model. The European Federation of Furniture Manufacturers (UEA) reports that 80% to 90% of the EU furniture waste in Municipal Solid Waste is incinerated or sent to landfill, with about 10% recycled (FURN360, 2017). Reuse activity in the sector is considered low and where it does occur, it is mostly through commercial second-hand shops, social enterprise companies or charities. European Environmental Bureau has identified the main challenges faced by the sector to move from a linear to a Circular Economy, mainly ascribed to materials and design, poor demand and consumer information, high costs of repair and refurbishment, reuse policy failure to be prioritized over recycling (EEB, 2017).

According to the DIGIT-FUR project (CFPIMM, 2019) forecasted vision for the furniture sector in 2025, digital tools will be crucial for the furniture industry and will be massively used, on the one hand for mass-customization for consumers (online tools, on demand manufacturing models, etc.) and on the other hand for digital manufacturing, from design (virtual modelling, artificial vision, etc.), to efficient manufacturing (Industry 4.0, robotics, additive manufacturing, etc.) and distribution (smart logistics, etc.).

Accordingly, the research methodology involves both digital technologies and wood waste-based biomaterials, implementing end of waste and metrics for circularity to assess the recycling potential of wooden scraps and wastes, designing new materials lifecycles and circular technologies for sustainable Made in Italy and design and depicting design methodologies for improving the wooden furniture sector according to the Circular Economy principles.

In particular, the experimentation will cover three main aspects:

1. the implementation of a Life Cycle Assessment methodology for bio-based waste-derived materials (wood scrap, natural fibers, cellulose, etc.) and digital design and fabrication processes (Artificial Intelligence and Machine Learning, Computational Design, Robotic Fabrication);
2. the assessment of the fabrication potentialities with digital technologies of the new materials, defining strategies for machines and/or materials customization;
3. the definition of possible design configurations, in compliance both with the machines specifications and the materials features.

The main goal of the FoRWARD project, nonetheless, is to fund an investigation to the new relationships designers and researcher are establishing with Technology in the Digital Age, and how this can be addressed to principles of necessity, i.e. CE and sustainability. In these terms, environmental sustainability only represents one aspect of the problem. The growing distance between advanced research and widespread design and craft practices, indeed, is urging for reflections about the social and economic impacts of digital technologies (Piscicelli, 2023).

For this goal, along with material and technical investigation, the FoRWARD research project intends to activate a correspondence between technological innovation and territories through the organization of collaborative actions, i.e. a Living Lab, that will take place in 2024 and will involve professionals, scholars, companies, artisans and students. In a CE perspective, upcycling relies on short-supply chain and local resources and waste. In this sense, an effective IU is actualized by computation, but is operationalized in an eco-socio-technical milieu. That is, the ecological intelligence of the upcycled material is the result of the original “redesign” of digital processes within the material culture of a Design Terroir. i.e.: «A delimited geographic space defined by a human community that builds throughout its history an ensemble of distinctive cultural traits, of knowledge and practices, based on an interaction system between

the natural milieu and human factors. The know-how at play reveals an originality, confers a typicity and allows a recognition for products and services that originate from this space and for humans that inhabit it. Terroirs are living and innovative spaces that cannot only be assimilated to tradition. Design terroir represents an emergent eco-social movement in design and architecture. [...] design terroir practitioners aim at increasing rich, direct environmental experience, and fair social development by promoting local and circular economy» (Pierce & Hermansen, 2023). In other terms, actualization provides for an answer; operationalization provides for a question.

Since February 2024, the activity is undergoing within a collaboration between the Department of Architecture, Federico II and the Center for Information Technology in Architecture (CITA), made possible also thanks to the ERASMUS PLUS KA1_ "University for Innovation" - Project Ref. n. 2021-1-IT02-KA131-HED- 000011202 mobility project promoted by the SEND Mobility Consortium, Italy.

In particular, the "recipe" for the bio-polymer 3D printing is developed within the CITA's Eco-metabolistic Architecture project (ERC H2020 grant N. 101019693) and will be implemented with the use of Italian forest (e.g. fir tree) wood flour and be tested in different possible application to furniture and interior component design (Krapez Tomec & Kariz, 2022).

7. CONCLUSIONS

In this paper, a conceptual framework is outlined to link CE to Computational Technologies and methods under the novel notion of "Informational Upcycling". Reclaimed resources, and wood waste in particular, is here defined and investigated as a computational material. These challenges that reclaimed wood poses to established design methodologies – uncertainty, non-linearity, unpredictability – require a brand-new theoretical approach that might be inherently related to Computational Thinking and Technological Design and can be particularly adapted to test the boundaries in the use of digital tools. This outline is informed by a case study analysis collected and reviewed for their innovation in terms of reuse and upcycling. Finally, the paper presents the FoRWARD – Furniture Waste for Circular Design research project, that aims to test the above-mentioned approach to the Made in Italy furniture industry.

Therefore, the FoRWARD agenda for the operationalization of IU requires the definition of a set of questions, e.g.:

1. Who is the agent who will operate/benefit from our upcycle process, material or product? Where does this operation act?
2. What is the production scale we are addressing our upcycle process, material or product?
3. What are the desired/possible technical features we can get through computation?

Considering the definition of IU and the questions for its operationalization, a Living Lab (LL) approach appears to be a suitable solution to actualize Laboratory's computation within a Design Terroir. The LL approach allows for a hybrid environment in which different stakeholders can

interact, tapping into the creative potential of each, within a systemic and structured framework of innovation (Eriksson et al., 2006).

This approach would interweave Laboratory's research methods and "affirmative" design (Oxman, 2016), adopting Living Labs to confront technological innovation with material culture and communities' requirements and collocating societal contribution as a vital step in research development, aiming at overcoming limitations of computation and traditional Open Innovation.

Within a Living Laboratory's research, IU can serve to re-design computation to be rooted in a material culture. The effects of this design operation would be the (re)activation of production patterns of a Design Terroir, leveraging digital methods to intercept and encourage emerging social and technical networks across the agents, lining up circular supply chains, formalizing tacit material knowledge, developing new competences and promoting inter-generational knowledge exchange.

ACKNOWLEDGEMENTS

The present study is carried out in the framework of the FoRWARD - Furniture Waste for circular design (MICS – NextGenEU, PE11, Spoke 4) research project, coordinated by Prof. Massimo Perriccioli and Prof. Marina Rigillo (Department of Architecture, University of Naples "Federico II"), and promoted by the Department of Architecture, the Department of Social Sciences and the Department of Law, University of Naples "Federico II" (IT), the Department of Engineering of the University of Palermo (IT) and the Department of Agricultural, Food, Environmental and Forestry Science and Technology of the University of Florence.

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A CASE STUDY OF IMPLEMENTATION OF CIRCULAR ECONOMY PRINCIPLES TO WASTE MANAGEMENT: INTEGRATED TREATMENT OF CHEESE WHEY AND HI-TECH WASTE

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Article Info:

Received:
11 April 2024
Revised:
15 June 2024
Accepted:
12 July 2024
Available online:
6 September 2024

Keywords:

Processes integration
Fermentation
Green leaching
Hydrothermal carbonization
Waste
Biorefinery

ABSTRACT

In a global context characterized by severe environmental problems and increasing resource scarcity, waste represents both a challenge and an opportunity. This study aims to demonstrate with a real case the potential for optimizing the waste valorization action attainable through the synergic application of different treatments to residues of equally different nature and origin. In particular, bio-chemical (dark fermentation), chemical-physical (selective leaching) and thermo-chemical (hydrothermal carbonization) treatments were applied for the integrated valorization of whey from sheep cheese production and Hi-Tech waste (discarded electrical and electronic equipment). The treatments were applied at a laboratory scale on real samples of these residues. The organic acids used for selective leaching of valuable metals from Hi-Tech waste were obtained by dark fermentation of the cheese whey, while hydrothermal carbonization was used to convert the waste from previous stages into hydrochar feasible as solid fuel or soil improver. The dark fermentation tests have highlighted the possibility of recovering ≈ 100 g of organic acids from 1 L of whey; furthermore, it is also possible to recover bio-hydrogen depending on the operating conditions applied and the type of targeted organic acids. The leaching tests have demonstrated how the organic acids from whey fermentation have selective and quantitative mobilization capacities comparable to those of the same acids available on the market. The carbonization tests produced carbon-enriched hydrochar with promising fuel properties, as well as process waters suitable for anaerobic digestion with methane production. The results of the project led to the filing of an international patent.

1. INTRODUCTION

The term sustainability demands meeting and conciliating various needs (productive, economic, political, social, environmental) that are strongly interconnected. In fact, humans have generated imbalances of such magnitude within each of the aforementioned sectors that the effects go beyond their own boundaries. Consequently, we live in an era characterized by complex problems to which it is not possible to give simple answers ("For every complex problem there is an answer that is clear, simple, and wrong." – H.L. Mencken). The possibility of overcoming current difficulties and guaranteeing a future for life on Earth lies in thinking in a complex way.

Waste management is no exception to this. In a global context characterized by severe environmental issues and

resources which are progressively scarcer in relation to the increasing demand, waste represents an environmental burden to be properly managed, but also a potential resource.

It has come a long way for waste management. Initially, and for a long time, the approach was extremely basic: discard, collect, sink it into a faraway hole or burn it. We then moved on to the integrated management system in which valorization assumed an important role, even if meant, almost, more as landfill avoidance than a real recovery of marketable resources. In particular, the valorization of organic waste was, and still currently is, pursued mostly in the form of a few products, e.g., biogas and compost, characterized by a relatively low value (Alibardi et al., 2020; Clarke, 2018).

Today, with the application of the principles of the circular economy, waste management must acquire char-



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acteristics of complexity to fully enter the economic-productive system where it contributes to the reduction of the consumption of raw materials/resources by delaying the becoming waste of objects (reuse/maintenance) or recovering resources (recycling) (Alibardi et al., 2020; Sarc et al., 2019; Vrancken et al., 2017; Walmsley et al., 2019). Although the implementation of the circular economy favors reuse, recycling still plays a main role for degradable goods and materials, for which the possibilities of reuse are understandably very limited. In this respect, organic residues may represent a widely available and renewable source for the recovery of products and energy to be used in other activities (Alibardi et al., 2020; Ma et al., 2018; Papież et al., 2018).

To be consistent with the principles of the circular economy, recycling must aim to minimize waste (approximation of the zero-waste objective) and to recover materials and compounds of interest for the production system.

Pursuing these objectives requires overcoming the logic based on the application of a single process for the recovery of a single product, in favor of the ability to combine processes synergistically in such a way that the outflow of one is the input of another and the different recovered products meet different market needs, in other words: the ability to think in a complex way applied to waste management (Asunis et al., 2020; Flórez-Fernández et al., 2021; Longati et al., 2020).

In the case of biodegradable waste management, the synergistic combination of processes is often referred to as waste biorefinery, a concept that is considered pivotal for the implementation of the evolution of the circular economy known as circular bioeconomy. Indeed, the waste biorefinery is an evolution of the biorefinery concept to include waste as an alternative to dedicated biomass and to enhance the recovery of organic waste (Alibardi et al., 2020). Since, as it was already said, in the circular economy the economic aspect (marketing) must be as important as the environmental one (waste treatment), the ideal waste biorefinery must be able to recover from waste biomass products characterized by value on the market and, not insignificantly, usable also in production processes that have nothing in common with those that originated the waste biomass, in order to provide flexibility and resilience on the market.

In this framework, the project "Integrated treatment of cheese whey and Hi-Tech waste: a demonstration of synergistic waste management in a circular economy perspective" represents a case study of the application of the principles of the circular economy/bioeconomy to two completely different production sectors such as the dairy sector and the electrical and electronic goods one.

The paper presents the preliminary results of the project that, although they require further investigation, highlight the potential of the concept of integrated valorisation.

The project was conceived taking into consideration some principles consistent with what was stated above:

- The valorization of residues characterized by different origin and nature;
- The integration of different processes (bio-chemical, physico-chemical, thermo-chemical);

- The recovery of products characterized by high added value and flexibility of use on the market;
- The integration between the solution of potential environmental problems and the opportunity to strengthen and expand the economic assets of specific production sectors;
- Waste valorisation as element of connection between different productive sectors.

In particular, bio-chemical (dark fermentation-DF), chemical-physical (selective leaching) and thermo-chemical (hydrothermal carbonization-HTC) treatments were applied for the integrated valorization of whey from sheep cheese (CW) production and Hi-Tech waste (generic term used to describe all types of old, end-of-life or discarded electrical and electronic equipment). In particular, the treatments were applied at a laboratory scale on real samples of the residues under concern. The organic acids used for selective leaching of valuable metals from printed circuits boards-PCB were obtained by DF of the CW, while HTC was used to convert the waste from previous stages into hydro-char feasible as solid fuel or soil improver.

The processes selected to be jointly applied were chosen according to the characteristics of the residue under concern:

- Compatibility with high organic load substrates (DF, HTC);
- Capability to manage streams with high water content (DF, leaching, HTC);
- Capability to simplify complex organic molecules by preparing them for subsequent phases of recovery (DF);
- Capability to recover compounds characterized by high added value (DF, leaching of valuable metals from Hi-Tech waste).

Cheese whey is the most important residue of the dairy industry (0.8-0.9 L produced per L of milk processed) and is considered a potential environmental risk due to its quantity, high organic load, presence of salts and tendency to acidify. Improper management leads to anoxia, toxicity, contamination of groundwater and soil. Discharge of CW into sewer systems can affect the efficiency of water treatment plants and the high content of lactose and minerals causes problems if used as animal feed (Akhlaghi et al., 2017). So far, chemical-physical and biological treatments have been applied mainly aimed at removing the organic load. Some characteristics of CW have also proven to be problematic for the application of valorization options such as methanogenic anaerobic digestion (Asunis et al., 2020).

Dark fermentation represents a mainstay towards full-scale implementation of the waste biorefinery concept. In waste management, DF has always been seen as an intermediate stage of anaerobic digestion to be overcome as quickly as possible in order not to affect the recovery of biomethane. In the new context of the circular bioeconomy, the ability of DF to mildly decompose the organic matter is instead a strong point. In fact, DF is a versatile process that can be addressed towards the desired products through

the proper setting of the main operating parameters (operating pH and temperature, organic loading rate, hydraulic retention time, etc.) to recover gaseous and soluble substances/compounds that can be used for energy purposes or as building blocks or substrates for further processes. Moreover, DF not only allows the recovery of marketable products, but it is also ideal as preparation of the original organic substrate for other processes aimed at recovering further energy and materials. Finally, DF can be preceded by treatments aimed at the extraction of intact compounds with high added value (e.g., for the medical, pharmaceutical, nutraceutical sector). In this respect, DF may represent a promising option for CW valorization (Asunis et al., 2020; Carvalho et al., 2013). The issue has been already addressed by several studies with emphasis on H₂ production, but much less attention has been paid to the possibility of recovering also other valuable products. The authors have been performing studies on the application of DF to CW for years, though with the aim of recovering bio-hydrogen, a type of recovery that is also compatible with the recovery of most of the organic acids targeted in the present study, with the exception of lactic acid (Akhlaghi et al., 2017; Asunis et al., 2020).

Printed circuit boards are of great interest due to their high content of valuable and critical elements, on the basis of which they can be considered secondary sources of raw materials. However, current industrial recovery exploits energy-intensive thermal treatments and/or chemical processes based on the use of aggressive leaching agents such as HNO₃ solutions through a non-selective approach (Burkovic, 2007; Carvalho et al., 2013; Iannicelli-Zubiani et al., 2017). Several alternatives have been proposed, highlighting the potential of weak organic acids as a more sustainable and selective solution (Jadhav et al., 2016).

Hydrothermal carbonization is a wet thermochemical process where heat and pressure are applied to treat organic matter in the presence of water and under subcritical conditions (Libra et al., 2011). Heat and pressure are applied to process organic matter in presence of water and under subcritical conditions (Kambo & Dutta, 2015). The resulting material is a carbonaceous solid, characterized by high chemical stability and energetic value and a moderately high surface area, suitable as a fuel, adsorbent or soil improver, as well as to ensure stable carbon storage. HTC is currently considered a promising approach, in particular for the valorization of organic wastes characterized by a high water content such as agro-industrial residues and fermented/digested substrates (Faru et al., 2022a; Kim et al., 2014; Li et al., 2020). The obtained material is a coal-like hydrochar characterized by high chemical stability and energetic value, and a moderately high surface area, which is suited as fuel or adsorbent or soil amender, as well as to ensure stable carbon storage. HTC is currently considered a promising approach, in particular for the valorization of organic waste characterized by high-water content, such as agro-industrial residues and fermented/digested substrates.

The novelty of the study is not only in the application of the individual treatments on the individual waste under concern, but above all in the integrated application of

processes for the joint valorization of CW and PCBs. To the best of our knowledge, this approach has never been studied, and the results obtained have led to the filing of an international patent. Given the limited number of studies reported in the literature, or full-scale applications, which refer to synergistic and combined management of residues of different nature and origin, this project is characterized by a level of innovation and interest that involves not only the single processes and the residue under concern, but more in general, the approach to the management of any waste from production activities. In this respect, the proposed study is believed to contribute to further research aimed at exploring innovative waste management and valorization strategies.

2. OBJECTIVE

The general target of the project is the study of an innovative and integrated system for the valorisation of sheep cheese whey and Hi-Tech waste. The system combines, through an appropriate sequence of processes, the control of the negative impact deriving from inappropriate management with the recovery of resources, according to the principles of sustainability and circular economy.

The general target was divided into specific goals:

- Production of organic acids through CW dark fermentation performed under operating conditions suitable to guarantee process stability and maximize production yields;
- Recovery of metals from Hi-Tech waste through selective leaching performed by using the CW dark fermentation products as waste-derived and gentle green chemicals, as alternative to conventional approaches where aggressive unselective leaching agents are used and subsequent separation and purification phases are required;
- Complete the valorisation process through HTC of the residues deriving from the clarification of the fermented CW and leaching of metals from Hi-Tech waste.

The general description of the experimental activities as well as the preliminary results obtained, are reported below.

3. MATERIALS AND METHODS

3.1 Cheese whey and printed circuit boards

Samples of ovine CW were collected at a dairy plant in Sardinia (Italy). Since it is a biodegradable material, the samples were stored in a refrigerator, taking the quantities necessary to carry out the activities from time to time. The CW was then subjected to chemical-physical characterization analyses functional to the application of the valorization treatment by means of DF.

RAM type PCBs were sampled at a recycling plant for waste from electrical and electronic equipment, pre-treated through mechanical comminution to increase the specific surface area and characterised by ICP-AES/MS, XRD, SEM-EDS.

3.2 Treatment tests

3.2.1 CW dark fermentation tests

Mesophilic batch DF tests were carried out, without adding any external inoculum, using automatically controlled bioreactors, adopting three different pH setpoints (6, 6.5 and 7). Liquid and gaseous products were characterized; in particular, lactose and HLa concentration were monitored over time in DF step 2 and VFAs concentration in DF step 3. Finally, a predictive model of the evolution of the process based on the pH value, initial lactose concentration, HLa/VFAs ratio was derived.

The CW organic load was exploited through a 3 steps DF process, without pre-treatments or external inoculum in order to reduce the complexity and cost of the process: 1) lactose hydrolysis to glucose and galactose, 2) monomeric sugars conversion to lactic acid (HLa), 3) HLa conversion to soluble (volatile fatty acids-VFAs) and gaseous (H_2 and CO_2) products (Asunis et al., 2020). In view of the subsequent steps of the integrated process, both the HLa produced during DF step 2 and the mixture of VFAs produced during DF step 3 are of interest. In this respect, the DF process was properly managed and monitored. More specifically, when HLa was the product of interest, the optimal operating pH and duration of DF step 2 were optimized in order to maximize the production of HLa and avoid its conversion into VFAs. Conversely, when the product of interest was a mixture of VFAs, the operating pH and the overall duration of DF (steps 1, 2 and 3) were optimized to obtain the maximum conversion of HLa to VFAs. Furthermore, DF step 3 can occur according to different metabolic pathways with consequent effects in terms of presence of acetic, propionic and butyric acid in the fermentation broth with different reciprocal mass ratios. Therefore, the effect of the pH on the composition of the DF broth was evaluated.

3.2.2 Tests aimed at recovering high-value metals from Hi-Tech waste

Appropriate conditions were, hence, applied to dissolve and recover selectively specific classes of metals from Hi-Tech wastes. Specifically, the fermented CW, rich in HLa or mixtures of VFAs, was used after a simple clarification step (addition of ethanol 5:3 in vol. (Morr & Lin, 1970)), as an agent for leaching base metals (metals characterized by low reduction potential, $E^\circ < 0$, like lead, tin, iron, cobalt, etc.) under gentle experimental conditions (low concentration, temperatures spanning from room to 100°C, open environment at atmospheric pressure) from comminute waste Printed Circuit Boards (PCBs), as a potentially greener and safer alternative to the conventional use of strong oxidising mineral acids. The use of fermented CW was integrated in a previously established innovative process where base metals were leached by using commercial citric acid solutions as a preliminary step to the low environmental impact and selective recovery of noble metals (metals characterized by highly positive reduction potential, $E^\circ > 0$, like copper, silver and gold) (Rigoldi et al., 2019). Powdered samples of RAM PCBs were reacted with fermented aqueous CW mixtures on varying the tempera-

ture (25-100°C), the primary leaching agent (HLa or VFAs) and the lixiviant concentration (0.5-3M) in order to set up the best conditions for a selective and efficient base metals leaching. Experiments were also performed with pure commercial lactic, acetic, propionic and butyric acids, as a reference. The leaching results were investigated through an accurate chemical-physical characterization of the materials, mainly by ICP-AES/MS, XRD, SEM-EDS, before and after chemical treatment. Activities are, hence, progressing in two different directions: i) base metals are selectively recovered from the leachate by conventional fractional precipitation procedures, specifically separating toxic lead in form of low solubility halogenide, and providing HLa-rich waste solutions useful as feedstocks for HTC valorization; ii) the residual solid fraction is treated for noble metals recovery as for the recovery scheme already established.

3.2.3 HTC tests

The residues deriving from the clarification of the fermented CW and the metal leaching process were valorized through hydrothermal carbonization. For the sake of comparison, HTC was applied also on the raw CW. The influence of the main operating parameters (temperature and residence time) was tested and optimized in order to improve the product properties. Three different temperature setpoints and three different residence times were investigated. The process temperature was set between 180 and 220°C, over a duration of 0.5, 1 and 2 hours, allowing hydrolysis, condensation, decarboxylation and dehydration reactions whose effect is to reduce the hydrogen and oxygen content of the residue, while carbon and energy content are densified in the solid material (Faru et al., 2022b). Batch HTC runs were performed on the CW and the two process residues in a 1.5 L pressurized and stirred reactor equipped with an electric jacket for temperature control. The produced hydrochar was characterized in terms of proximate and elemental analyses and calorific value measurement. Properties, such as pH, electric conductivity, nutrient availability, thermal stability, type of surface functional groups and germination ability, useful to evaluate several hydrochar applications, were assessed. The HTC process water was also characterized in order to explore possible applications on soil and for energy production (through germination test and BMP-Biomethane potential test). A schematic representation of the studied process is provided in Figure 1.

4. RESULTS

The obtained results can be summarized as follows:

- Assessment of the proper operating conditions to obtain a stable and high conversion of the CW organic load into products to be used for subsequent process phases;
- Assessment of the maximum yields obtainable for the products of interest: lactic, acetic, propionic, and butyric acid;
- Obtaining DF broths suitable as a leaching agent for the recovery of metals from Hi-Tech waste;

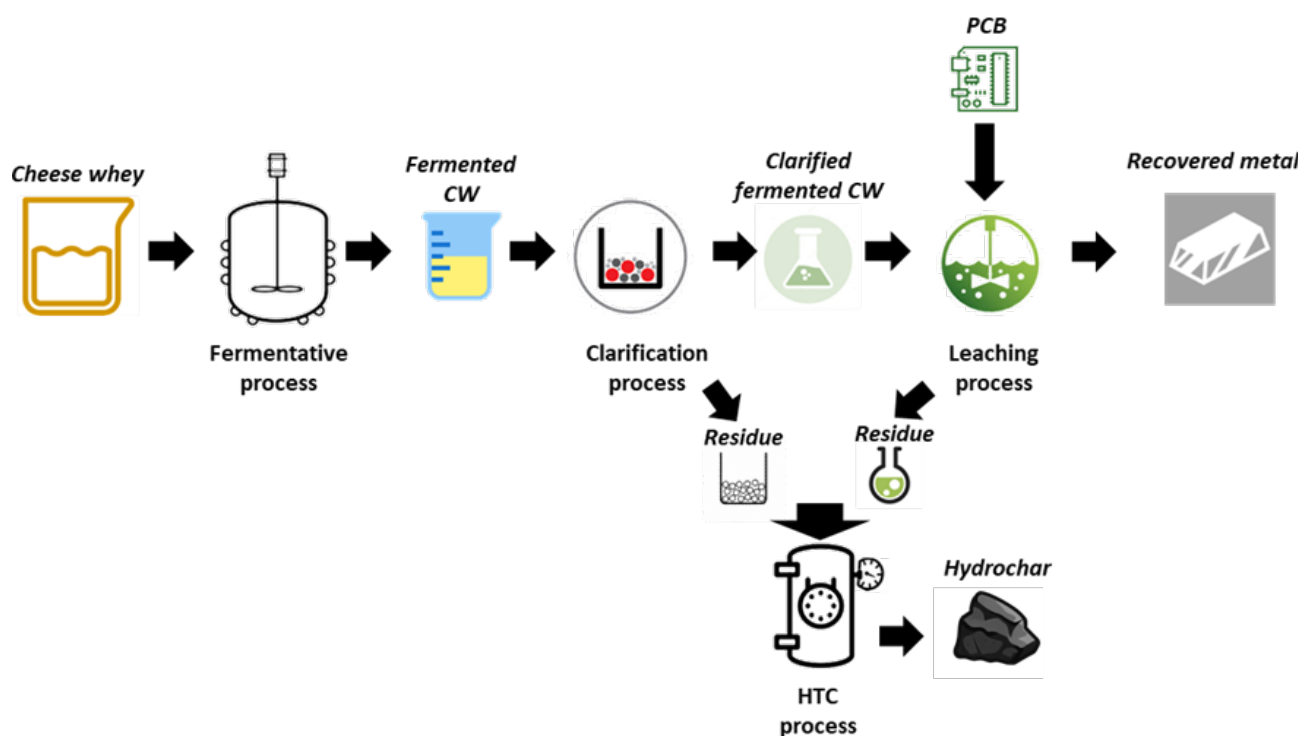


FIGURE 1: Schematic representation of the studied integrated treatment of sheep cheese whey and Hi-Tech waste.

- Identification of the more selective leaching solution able to enhance recovery of single metals or classes of metals contained in powdered PCBs;
- Addressing, in an interactive way, DF of CW towards the production of the best leaching solution;
- Finding the best conditions for recovery of metals from real Hi-Tech waste, when necessary also combining the use of fermented CW with other safe and recycled reagents for high reduction potential metals (e.g., I3⁻ for gold) in order to enhance the metal values from PCBs as much as possible;
- Assessing the operating conditions suitable to optimize the final quality of the HTC-produced hydrochar and process water;
- Obtaining HTC hydrochar suitable as solid fuel or soil improver, as well as process water suitable for methane production.

In details, the DF tests highlighted that the process develops according to two stages characterized by different products, both of interest and that can be recovered separately thanks to the adoption of suitable operating parameters: lactose is converted to lactic acid (1st step), and thereafter lactic acid is converted to VFAs (2nd step). In order to optimize the conversion yields and kinetics of both stages and allow for their separate management, the operating parameters that play the fundamental role are the operating pH, which must be controlled continuously, and the hydraulic retention time. In light of the results obtained, it is possible to hypothesize the following production yields: $\approx 60\text{--}70\text{ g HLa L}^{-1}$, $\approx 10\text{--}15\text{ g HBu L}^{-1}$, $\approx 7\text{--}10\text{ g HPr L}^{-1}$. Moreover, the production of VFAs in the second stage may lead also to the recovery of $5\text{--}7\text{ L H}_2\text{ L}^{-1}$.

Preliminary results on comminuted RAM PCBs have shown how HLa performs better as leaching agent than other VFAs contained in fermented CW, probably due to its lower pK_a in water. For this reason, further systematic experiments were focused on HLa-based solutions application. Indeed, 1:1 HLa/La⁻ buffer solutions ($pK_a = pH = 3.8$) demonstrated the highest efficacy and selectivity towards base metals even at very low concentration level (0.5M) and room temperature, obtaining an almost quantitative leaching yield for lead and tin within 24 h against desired zero or negligible rates of copper, silver and gold dissolution. Experiments confirmed the support of dissolved oxygen in the efficacy of the leaching, as already observed in previous studies on cobalt recovery (Oumarou Amadou et al., 2023). Furthermore, fermented CW mixtures demonstrated to achieve higher leaching rates towards a selection of metals, i.e. iron (73%) and nickel (72%), which are leached at a lower extent (10 and 13%, respectively) by the HLa commercial agent under the same experimental conditions (Serpe et al., 2023). In that sense, the oxidizing and complexing properties of the different soluble products of CW controlled DF showed to play a relevant role, still under investigation, in the efficiency and selectivity of metal leaching. Leaching under refluxing temperature (100°C) shows that a significant improvement occurs just on the effectiveness of Fe and Ni dissolution. The presence of significant concentrations of salts, e.g., chlorides, in the mixtures also affects the occurrence of soluble/insoluble species in the post-leaching aqueous and solid fractions and should be carefully considered in designing the steps of the recovery process. Selective precipitation recovery of metals by adding sequentially halogenides, hydroxides and

sulfides, allowed to isolate valued and, also, toxic metals and to provide a solution suitable for the following HTC treatment (suitable organic loading; metals concentration: Ni = 10.0 ppm; Cu = 11.4 ppm; Sn = 2.94 ppm; Fe = 2.18 ppm; Al = 0.21 ppm; Cd = 0.12 ppm; Pb, Cr, Co < 0.1 ppm).

So far, a set of HTC tests were performed at 220°C for 1 hour on CW, fermented CW, and a mixture of fermented CW and the residues after leaching. Under these conditions, the produced hydrochar exhibited a carbon densification (up to +53%) and a decrease in ash content (up to -63%). The resulting process water have shown significant potential for biogas production during BMP test. Based on these preliminary results, the HTC products, including hydrochars and process water, show a potential for energy recovery. The hydrochars, with their increased higher heating value (HHV, up to +82%), can be used for heat production or converted into electrical energy. The process water can be valorized within anaerobic digestion systems aimed at biogas production. Germination tests on cress seeds conducted using hydrochar, process water as well as CW indicated a high level of inhibition across all tested materials. The primary source of inhibition observed in the solid materials (CW and hydrochar) can be attributed to their elevated fat content, while the phytotoxicity observed in the process water can be attributed to the presence of dissolved organic compounds (total organic carbon up to 12 g/L).

5. DISCUSSION

The integrated treatment of cheese whey and Hi-Tech waste through DF, green leaching, and HTC demonstrates significant potential for the valorization of the residues under concern. However, the economic feasibility of the processes is crucial for their adoption in a circular economy. While this study is a preliminary report and does not include a detailed economic analysis, several factors indicate the potential for profitability. The reduction in disposal costs by converting waste into valuable products is a significant advantage. Additionally, the production of organic acids through DF, the recovery of metals from PCBs, and the production of hydrochar offer multiple revenue options. The application of green leaching agents and HTC reduces energy consumption compared to more conventional methods, decreasing the operating costs. Furthermore, the recovered metals and hydrochar have medium-high market value, thus support the economic viability. The same organic acids obtained by DF of CW could be placed on the market with good value regardless of the use as green leaching reagents. Furthermore, it is worth mentioning that the organic acids could also be used for other processes capable of recovering high added value products such as the production of biopolymers, and that also bio-H₂ is attainable during CW fermentation depending on the process operating conditions and the type of organic acid that is targeted (Asunis et al., 2020, 2022).

At a time when waste valorization seems to want to go beyond simple process schemes, characterized by the recovery of a single product of medium-low market value, to embrace multi-product approaches and oriented towards

the highest added value, it is legitimate to ask whether this vision is actually applicable. In light of this, the development of studies such as the one illustrated cannot be limited to additional, mere experimental tests to further define and optimize the operating conditions, the achievable yields, etc. The in-depth study must also include technical-economic feasibility studies, performed in cooperation with the possible stakeholders and market players, and taking into consideration the differences that exist between the treatment of materials classified as waste, and the traditional treatment of raw materials or virgin biomass. The availability of materials and the characteristics of the supply basins, logistics (storage, transport), legislation, the very certainty of current legislation, are very different, and are reflected in fundamental aspects such as the minimum economically sustainable size of the plants.

Performing comprehensive life cycle assessments (LCA) to assess the environmental foot print of the integrated processes as compared to conventional waste management methods is crucial to better understand the attainable benefits and potential trade-offs.

To fully evaluate the scalability of the processes, the feasibility and LCA studies require the availability of reliable data. In this respect, it is important to start pilot scale experiments, possibly extended to other types of waste characterized by wide availability such as municipal solid waste organic fractions or wastewater treatment sludge.

6. CONCLUSIONS

The study and the project have produced experimental evidences that support the feasibility of an integrated approach to the valorization of different wastes through the application of equally different processes. The proposed approach is consistent with the principles of sustainability, circular economy, circular bioeconomy.

In particular:

- DF converts the organic load of CW into mixtures of organic acids useful for several applications; pH confirms to be the key operating parameter for properly addressing the DF towards the desired products. The result is of particular importance since it is obtained by the application of DF to a real substrate; the control of the DF process performed on real residual substrates and using indigenous microorganisms is not straightforward, as witnessed by the relatively low number of pilot plants and, to the best of our knowledge, the absence of full-scale application cases;
- the mixtures of organic acids obtained from CW fermentation are suitable for the selective and gentle mobilization of the metals present in PCBs, thus making possible a process that is less aggressive than those usually applied;
- the final residues are converted via HTC into a product that can be used as a solid fuel or soil improver, which makes it possible to close the valorization cycle of the residues under concern; additionally, a process water is also produced that is suitable for methane production through anaerobic digestion.

More generally, the study demonstrates how integrated waste management can systematize different production sectors, limiting their environmental problems and converting them into opportunities of potential economic interest. This is of particular importance for production sectors such as the dairy one, which are affected by periodic market fluctuations of their traditional products. Paradoxically, an environmental problem can increase the flexibility on the market and the economic resilience of a production activity.

Future research should focus on scaling up the studied processes, performing detailed economic analyses, and exploring the feasibility also for other types of waste.

ACKNOWLEDGEMENTS

The research project “Integrated treatment of cheese whey and Hi-Tech waste: a demonstration of synergistic waste management in a circular economy perspective” was funded by the Fondazione di Sardegna and Regional Sardinian Government (Grant CUP CUP F75F21001350007).

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APPLICATIONS OF WOOD ASH BY MATCHING ITS CHARACTERISTICS WITH END-USER QUALITY REQUIREMENTS: A CASE STUDY

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Article Info:

Received:

19 May 2024

Revised:

2 July 2024

Accepted:

12 July 2024

Available online:

2 September 2024

Keywords:

Wood ash application

Waste valorization

Construction

Soil amendment

Regulatory requirement

Pulp paper mill

ABSTRACT

Despite being a bio-based material, wood ash generated by pulp and paper mills is mainly landfilled in Canada. This is because it is perceived as waste material and the certification requirements and regulations controlling its use are complex. To promote wood ash utilization, ash samples from mills in British Columbia (BC), Canada were characterized, and the properties were compared to quality specifications for potential applications. Three types of ash samples were collected: bottom ash, multi-clone (MC) ash, and electrostatic precipitator (ESP) ash. Each type of ash was characterized, and their suitability for various applications was determined. The study found that ESP ash had a higher calcium carbonate equivalent (CCE) value than MC ash, making it more useful as a liming material in agricultural land. The study identified quality criteria for industries where wood ash can be used, such as construction, agriculture, composting, stabilization/solidification, liming, mining, and fire-retardant. Each type of ash was evaluated for its use in these industries, and the environmental regulations for each application were considered. It was observed that the quality criteria for one application could differ dramatically from those for another. Intuitively, an ash producer would cross-check the characteristics of their ash types against the quality requirements for potential uses near the ash source because different applications have different quality requirements. This article is believed to help identify promising applications of ash thereby removing ash from landfilling and promoting the circular economy.

1. INTRODUCTION

Canadian pulp and paper mills mainly use woody residues, also known as hog fuel, to generate green energy by biomass boilers. Although most of the fuel turns into water and carbon dioxide after combustion, a portion remains and is called power boiler ash. This ash derived from wood is generally believed to contain fewer metals than coal ash. However, if the wood is chemically treated, the presence of toxic metals in the ash cannot be ignored (NWR, 2020). Despite its relatively cleaner nature, most of the produced wood ash in Canada ends up in landfills (Elliott and Mahmood, 2015; Gravesande, 2018; Stantec, 2011). According to the IEAB (2020) report, Canada produces more than 1,000 kilotons of wood ash, with over 60% of it being sent to landfills. This makes Canada the second-largest contributor to wood ash landfills, after Germany, among the selected European Union (EU) countries (Figure 1).

Landfilling wood ash conflicts with the concept of circular economy. As well, landfilling of ash consumes precious landfill space, and the deposited ash becomes an

environmental concern for years to come. Since the ash is treated as waste when landfilled, no economic value is created from this bio-based material resulting in poor economics. As such, there is a strong need to promote beneficial uses of ash rather than landfilling it for sustainable ash management with favourable economics.

To achieve this goal, four factors must be considered simultaneously: properties of the ash to be utilized, requirements of a specific ash end-use, regulatory environment, and process economics. Although there is limited information regarding ash end-use requirements and regulations controlling ash use in Canada, there are promising large-scale applications in the construction industry, agricultural sector, and forest/rural road construction, among others (Brand et al., 2021; Naylor and Schmidt, 1986; Vilane et al., 2021). Additional end uses of ash residues have been identified, albeit at relatively smaller scales. Included among these applications are ash use for mine tailings and mine-affected waters remediation, acidic waste neutralization, waste stabilization and solidification, adsorption, compost production, and structural fill to name a few



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Detritus / Volume 28 - 2024 / pages 48-59

<https://doi.org/10.31025/2611-4135/2024.19402>

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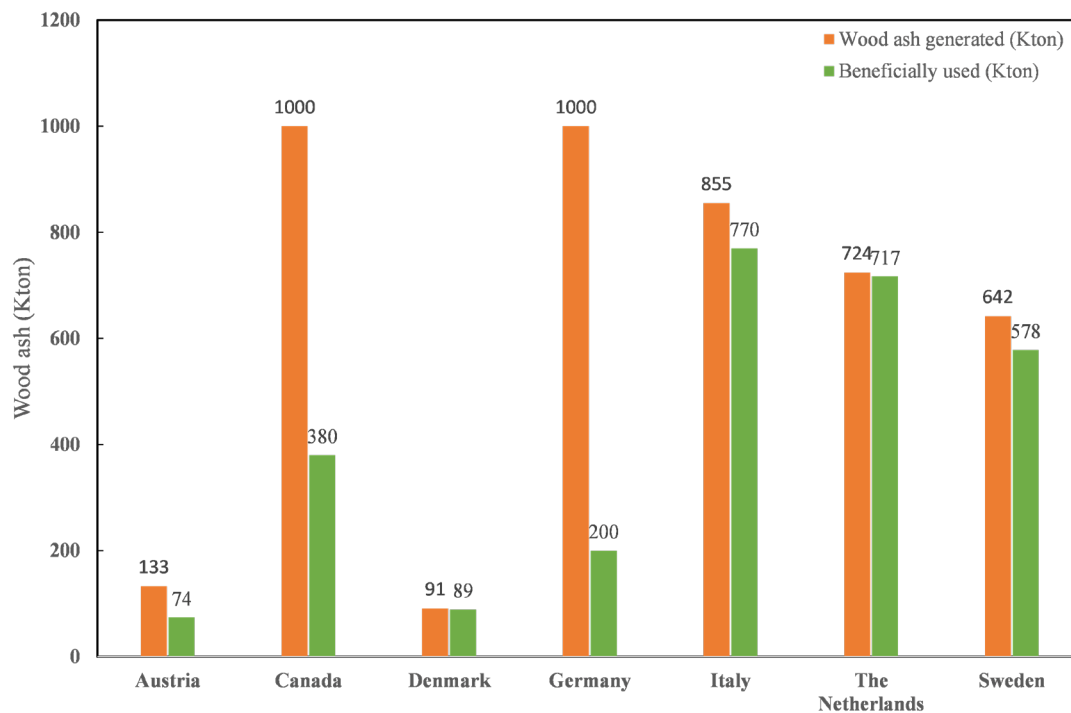


FIGURE 1: A comparison of wood ash generation and beneficial use for Canada and selected EU countries (Adapted from Bioreg, 2020; IEAB, 2018; Lazick et al., 2020).

(Elliott et al., 2022; Elliott and Mahmood, 2015; Mandpe et al., 2021; Wilk, 2020).

Characterizing wood ash is the first step toward identifying its best applications. Numerous studies have investigated the potential applications of wood ash, focusing on its physical, chemical, and mineralogical characteristics. Etiegni and Campbell (1992) have shown that wood ash yield decreases by 45% at higher combustion temperatures, with significant reduction in potassium, sodium, zinc, and carbonate content. Moreover, wood ash application has been found to have positive effects on soil properties and plant growth, making it an ecologically sound alternative for managing this industrial by-product (Demeyer et al., 2001).

Studies have also demonstrated that how wood ash varies from different materials and their impacts on the amelioration effects on acidic soils (Shi et al., 2017). Furthermore, wood ash has shown potential as a soil conditioner and an alternative nutrient source for agricultural soils (Baloch et al. 2024). While it has been found to enhance trace elements and alter microbial processes in soil, its overall positive impact on soil and plant growth makes it a promising option for sustainable soil management.

Mahmood and Kamal (2022) characterized wood ash collected from ten pulp paper mills in British Columbia, Canada. They demonstrated how the quality of wood ash varied from mill to mill and within a single mill while also exploring the relationship between different ash parameters. However, the wood ash quality was not used to identify its beneficial applications in that study. The main goals of the study are to thoroughly analyze the wood ash produced by

a typical pulp paper (PPM) mill in BC, Canada. This includes examining all three ash streams—bottom ash, multi-clone ash, and ESP ash. The objective is to match the properties of the wood ash with specific application requirements and provide recommendations for its optimal use. This innovative approach, as described in the article, will help ash producers make well-informed decisions about using their wood ash. Failure to utilize wood ash effectively results in disposal in landfills. Implementing this method would enable ash producers to divert wood ash from landfills, thereby extending the service life of landfills and fostering a circular economy. By promoting the beneficial uses of wood ash, there is potential to create economic value from this bio-based material, reduce waste, and minimize the environmental footprint.

2. MATERIALS AND METHODS

2.1 Production of wood ash in pulp paper mills

During the burning of hog fuels in boilers, the flue gases pass through multi-clone (MC) separators and are finally captured using scrubbers, baghouses, or electrostatic precipitators (ESP). Thus, fly ash (MC and ESP ash) is generated. Another type of residue is the relatively dense ash that accumulates at the bottom of the boiler and is called bottom ash or grate ash. Most ash in PPM in BC is managed after the two ash types are combined. In some mills, the fly ash and bottom ash are separated. According to the recent trend of PPM, three types of wood ash are generated: bottom ash, MC ash, and ESP ash. Ashes were named after the location where they were produced in the boiler.

2.2 The case study area

Ten pulp paper mills that were located in both inland and coastal areas of British Columbia, Canada were investigated. Some of these mills produce combined wood ash (Figure 2a), while others produce bottom ash and fly ash (Figure 2b). Most mills generate bottom ash, MC ash, and ESP ash (Figure 2c). The study focused on one of the mills that generate three streams of ash. Since the mill was located inland, the logs used in the mill were not transported by marine water. Ash produced from logs transported by marine water may contain dioxins and furans (GoC, 2024).

2.3 Sampling and analysis

We collected representative samples of bottom ash, MC ash, and ESP ash of the studied mill. Each sample was collected at the sampling point in a 20L full bucket and was not exposed to rain. Each sample was analyzed in triplicate for accuracy (n=3x3). Samples were analyzed for their physical, chemical, and engineering properties. A comprehensive list of parameters, along with the corresponding instruments used for their determination, are presented in Table 1. The details of sample preparation, methods, and tools used to determine each parameter

were discussed in the first phase of the study (Mahmood and Kamal, 2022).

3. RESULTS AND DISCUSSION

3.1 Wood ash characteristics

The parameters of wood ash needed for ash applications were determined. The analytical results are listed in Table 2.

3.1.1 pH

pH is a key parameter for wood ash applications, such as soil amendment, waste stabilization, and composting. Generally, wood ash is highly alkaline (pH>10.5). The pH of MC and ESP was highly alkaline (at about 12.5) while bottom ash had a pH of 10.7 (Table 2).

3.1.2 Moisture

It has been observed that the quality of ash deteriorates and the transportation costs increase as the moisture content increases. As expected, no sample of wood ash contained any moisture, meaning all of them had 100% total solids (Table 2). A wood ash application may require adding water to ash to achieve the desired moisture level.

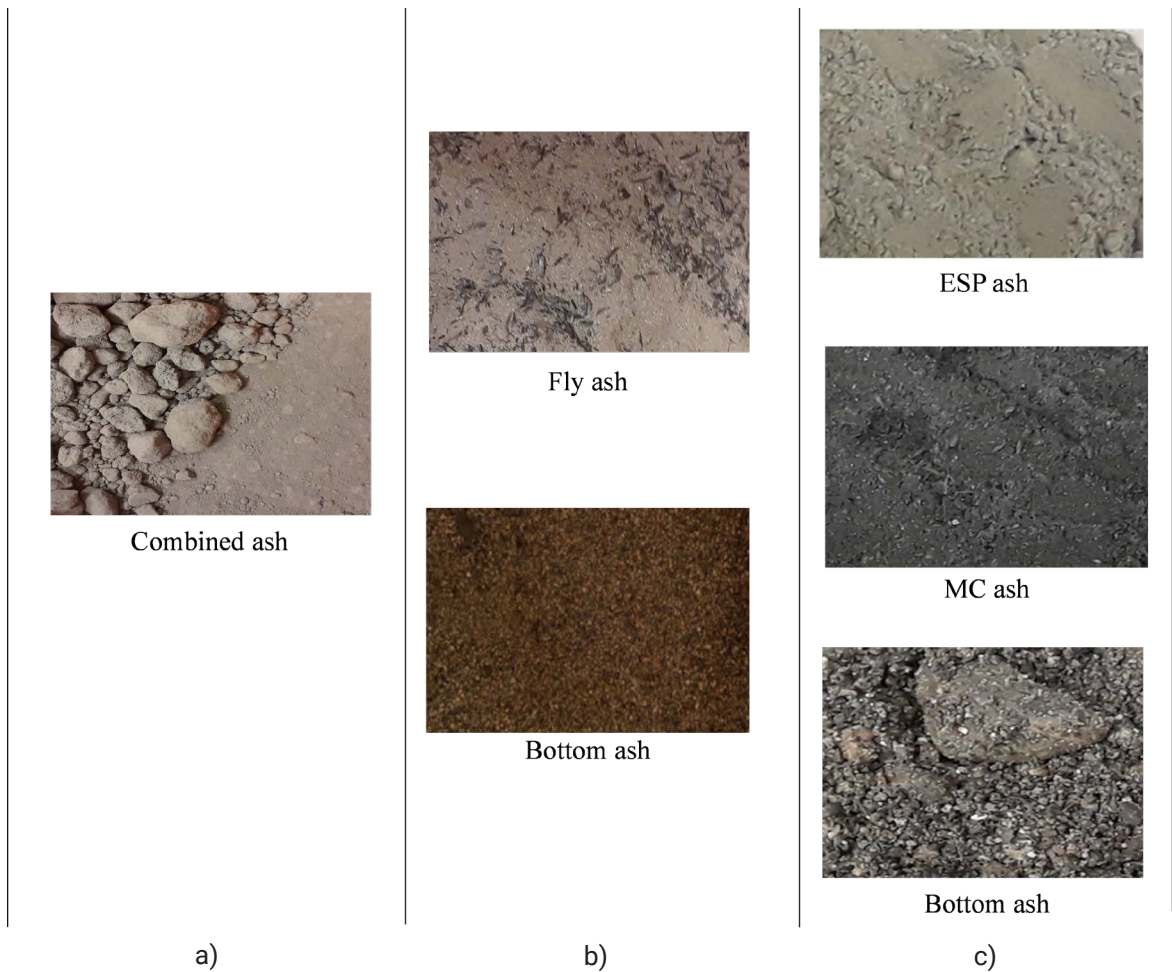


FIGURE 2: Pulp paper mills generally produce one of the three types of ash: (a) combined ash, (b) fly ash and bottom ash, (c) Electrostatic Precipitator (ESP) ash, Multi-clone (MC) ash, and bottom ash.

TABLE 1: Analytical tools/instruments used to determine the parameters of wood ash.

Parameters	Tools/Instruments	Model/Make/Place
pH	pH meter	Accumet 925 pH meter, Thermo Fisher Scientific, Burnaby, BC, Canada
Biochar/carbon	Gas chromatography	7890A, Agilent Technologies, Santa Clara, California, USA
Density	Graduated cylinder	213I11 Karter Scientific 50ml Glass Graduated Cylinder, Karter Scientific, Los Angeles, USA
Heating value	Oxygen combustion vessel	Parr 1108, Parr Instrument Company, Moline, Illinois, USA
Metals/elements	Optical Emission Spectroscopy-Inductively Coupled Plasma, OES-ICP	Thermo Scientific, Santa Clara, California, USA
Metal oxides	X-ray Fluorescence, XRF	A18-05132, Malvern Panalytical Axios, Malvern, UK
Minerals	X-ray Diffraction, XRD	D8 Discover, Bruker, Billerica, Massachusetts, USA
Chloride, Cl ⁻	Optical Emission Spectroscopy- Inductively Coupled Plasma, OES-ICP	Thermo Scientific, Santa Clara, California, USA

3.1.3 Biochar

Biochar is a solid carbon-rich material available in wood ash. It is beneficial for soil amendment but not for the concrete. There was a significant amount of biochar (i.e., total carbon) in both types of fly ash, but bottom ash had the least amount (Table 2).

3.1.4 Loss-on-Ignition (LOI)

LOI indicates the amount of organic matter present in ash, with the organic matter being the unburned carbon in ash (unburned coal). Thus, a higher LOI indicates the presence of more biochar. A concrete mix with a higher LOI of wood ash (i.e., a higher amount of biochar) may produce poor-quality concrete by impeding air entrainment, reducing freeze-thaw resistance and increasing permeability. The MC ash contained a higher amount of LOI than ESP ash (Table 2). For concrete, this makes ESP ash a better choice than MC ash.

3.1.5 Density

The higher the density, the lower the volume occupied. As expected, bottom ash had the highest density of 1640 kg m⁻³, whereas the MC ash had the lowest density of 430 kg m⁻³ (Table 2). As a result, when wood ash is disposed of in landfills without segregation, MC ash occupies the most space.

3.1.6 Heating value

The heating value of wood ash helps decide if any wood ash stream needs to be recycled with fuel feedstocks. The heating value of MC ash (8,200 kJ kg⁻¹) was higher than those of the other two ash streams (Table 2). If the heating value of any wood ash stream is comparable to that of wood (19,073 kJ kg⁻¹), then the wood ash stream may be recycled.

3.1.7 Size classification (Fineness)

The fineness of ash is the particle size. Compared to the fly ash, the bottom ash was found to be coarse (Table 2). Fly ashes used in concrete must meet the fineness requirements of ASTM C618 or CSA 3000 standards.

3.1.8 Trace elements

Coal ash contains contaminants, such as Hg, Cd, and As (USEPA, 2024). These elements can affect plant growth

and the environment in both positive and negative ways. When used for any agricultural applications, wood ash concentration in wood ash is controlled by the Canadian Food Inspection Agency (CFIA) regulations (CFIA, 1997). Generally, wood fly ash contains fewer toxic metals than coal fly ash for As, Pb, Hg, and Se. All these trace elements were below the detection limit (<0.1 ppm) of wood fly ash (Table 2).

3.1.9 Metal oxides

Wood ash contains several types of metal oxides determined to compare the chemical specifications of wood ash with those of cementitious materials. Among all metal oxides, calcium oxide (CaO) is the main ingredient that contributes to the strength of concrete. However, other oxides such as silicon dioxide (SiO₂), ferric oxide (Fe₂O₃), and alumina (Al₂O₃) also contribute to concrete strength by forming the C-S-H gel. Due to these oxides, wood ash exhibits both pozzolanic and hydraulic properties in concrete. Considering these metal oxides, fly ashes of the representative mill had a high potential to use in concrete than bottom ash (Table 2).

3.1.10 Chloride

Chloride affects the durability of concrete. Chloride may be available with the ingredients of concrete mix or may ingress into the concrete from the environment. In either case, chloride can attack the reinforcing bars of concrete. None of the ash streams of the representative mill exceeded the ACI (318) standard for chloride (Cl<1%) (Table 2).

3.1.11 Sulfur trioxide (SO₃)

SO₃ is responsible for sulphate (SO₄⁻²) attacks in concrete. Like chloride, sulphate can penetrate concrete structures. Sulfate attack forms Ettringite in concrete that expands, creates cracks and weakens the concrete. The higher the amount of SO₃, the greater the loss of concrete strength. Each stream of wood ash contained less SO₃ than the ASTM C618 standard (5% by weight) (Table 2).

3.1.12 Calcium carbonate equivalent (CCE)

The CCE value of wood ash indicates its ability to neutralize acidity. A higher CCE indicates better ash quality as a liming material/agricultural application. The CCE of ESP ash was higher than MC ash (Table 3).

TABLE 2: Characteristics of wood ash streams (n=3x3) collected at different boiler locations from a representative pulp paper mill in BC. Units are in ppm otherwise mentioned.

Parameters/Elements	Bottom ash	MC ash	ESP ash
pH	11±0.3	13±0.1	12±0.2
TS, %	100±1.5	100±1.2	100±0.5
Total carbon, %	0.7±0.0	20.1±0.3	10±0.0
Ash content, %	100±1.3	75±1.0	87±0.9
LOI, %, 550°C	0.4±0.0	25±1.2	13±0.6
Density, kg/m ³	1600±0.0	400±0.0	800±0.0
Heating value, kj/kg	135±3.0	8200±35	4143±20
Fineness / particle size, %, < 0.04 mm	5.2±0.0	42±1.5	39±1.2
Aluminum, Al	50510±597	33940±302	20690±230
Arsenic, As	<0.1±0.0	<0.1±0.0	<0.1±0.0
Boron, B	<0.1±0.0	<0.1±0.0	285±8.9
Barium, Ba	697±15	1389±25	1971±38
Calcium, Ca	53360±498	103100±842	211300±2012
Cadmium, Cd	BDL	BDL	10±0.2
Cobalt, Co	BDL	BDL	11±1.5
Chromium, Cr	38±0.5	45±0.7	669±1.7
Copper, Cu	38±0.6	42±0.7	139±1.5
Iron, Fe	21370±312	17670±245	17925±265
Potassium, K	18190±297	27610±542	75965±998
Magnesium, Mg	8151±97	11150±160	18645±312
Manganese, Mn	735±22	3893±105	7445±237
Mercury, Hg	BDL	BDL	BDL
Molybdenum, Mo	BDL	BDL	8.8±0.1
Sodium, Na	15610±292	8532±119	5580±91
Nickel, Ni	19±0.2	25±0.3	40±0.5
Phosphorous, P	807±1.1	5049±8.9	9525±22.5
Lead, Pb	<0.1±0.0	<0.1±0.0	<0.1±0.0
Selenium, Se	<0.1±0.0	<0.1±0.0	<0.1±0.0
Sulfur, S	BDL	BDL	13830±149
Silicon, Si	632899±1459	553746±1609	48770±1005
Strontium, Sr	452±5.0	605.9±6.3	934±10.4
Titanium, Ti	2032±29	1643±18	1117±22
Vanadium, V	68±0.9	38±0.5	38±0.6
Zinc, Zn	94±1.5	221±3.6	1679±10.2
Cobalt tetroxide, Co ₃ O ₄	BDL	BDL	BDL
Copper oxide, CuO	BDL	BDL	BDL
Nickel dioxide, NiO ₂	BDL	BDL	0.1±0.0
Silicon dioxide, SiO ₂	67.5±3.4	36.2±1.5	16±0.9
Aluminium trioxide, Al ₂ O ₃	12±0.2	7±0.1	4.6±0.1
Ferric oxide, Fe ₂ O ₃	4.7±0.0	3.2±0.2	3.0±0.2
Manganese oxide, MnO	BDL	0.6±0.0	1.1±0.1
Magnesium oxide, MgO	2.1±0.0	2.2±0.1	3.4±0.2
Calcium oxide, CaO	5.9±0.1	16±0.2	32±0.8
Sodium oxide, Na ₂ O	2.6±0.0	1.3±0.0	1.3±0.0
Potassium oxide, K ₂ O	2.6±0.0	3.1±0.0	5.8±0.0
Titanium oxide, TiO ₂	0.5±0.0	0.4±0.0	BDL
Phosphorus pentoxide, P ₂ O ₅	0.3±0.0	1.3±0.0	2.3±0.0
Chromium oxide, Cr ₂ O ₃	BDL	BDL	BDL
Vanadium oxide, V ₂ O ₅	BDL	BDL	BDL
Loss-on-Ignition (LOI), %, 1000°C	2.5±0.0	29±0.0	26±0.0
Chloride, Cl	BDL	BDL	BDL
Sulfur trioxide, SO ₃	<0.3±0.0	<0.3±0.0	<0.3±0.0

MC: Multi-Clone, ESP: Electrostatic Precipitator, BDL: Below Detection Limit (<0.005)

3.2 Possibility of using wood ash

Wood ash has various uses, such as in the production of concrete, as fertilizer in forests, construction material on roads, embankments and in agriculture. However, these applications are subject to specific standards, regulations, and best practices. In the next sub-sections, we will compare the quality requirements of each application with the wood ash parameters studied.

3.2.1 Ash use in the construction industry

Concrete is a regulated product that must adhere to strict specifications. Standards have been established to govern the use of coal ash in concrete. The Canadian Standard (CSA, A3000) identifies three types of ash for use in concrete production: F, CI, and CH. Similarly, the American Standard (ASTM, C618) classifies ash into three categories: N, F, and C. Each type or class has specific requirements that must be met, as outlined in Table A.1 (in the appendix).

Table A.1 (in the appendix) compares the chemical specifications of cementitious materials required in concrete by ASTM and CSA standards with those of Bottom ash, MC ash, and ESP ash. The results show that Bottom ash is unsuitable for concrete manufacturing due to its low CaO content. However, MC ash has a good amount of metal oxides and CaO, which meet the requirements of ASTM Class C, Class F, and CSA-type CH fly ash. Not only that, but it also has lower amounts of SO_3 , moisture, and chloride than the maximum value. ESP ash has the highest amount of CaO among the three ash streams, which satisfies the requirements of each class of ash for both ASTM and CSA standards. It also has a lower LOI than MC ash and SO_3 , moisture, and chloride content than the maximum values of both standards, making it an ideal choice. Therefore, if these ashes are allowed in terms of their use in concrete, ESP ash should be followed by MC ash and bottom ash.

Numerous investigations have been conducted to determine the degree of ash substitution for cement, with some recommending much higher ash substitution rates than others (Amran et al., 2021; Tosti et al., 2019; Rajamma et al., 2009). For instance, paper mill fly ash can replace up to 10% of Portland cement, resulting in mortars with positive effects on their mechanical properties (Fava et al., 2011). However, due to fly ash fineness and water absorption properties, it requires a greater amount of water. Other studies suggest much higher ash substitution rates, indicating that ash can be a suitable alternative to cement in concrete manufacturing rates (Barathan and Gobinath, 2013; Hemalatha and Ramaswamy, 2017; Rajamma et al., 2009). Therefore, it is evident that ash can have a strong impact on the mechanical properties of concrete.

3.2.2 Ash use in agriculture (soil amendment)

Canada uses most of its ash for fertilizing agricultural lands, unlike Europe, where it mainly goes to forest fertilizing (Herbert and Breton, 2009; IEAB, 2018). Wood ash contains essential nutrients like calcium, potassium, and magnesium that are crucial for plant growth. However, wood

ash samples may also contain harmful heavy metals that can harm soil quality. That's why the Canadian federal and provincial governments have set maximum concentration limits for eleven metals/elements of concern for ash application on land, including cadmium and mercury, which can be particularly concerning (Pöykiö et al., 2004).

To ensure that ash is used as a soil amendment safely, users need to follow guidelines and regulations set by the Canadian Food Inspection Agency (CFIA) and the Canadian Council of the Ministers of the Environment (CCME) at the federal level, as well as by provincial governments. As per CFIA guidelines, the maximum acceptable product metal concentrations are determined based on the maximum acceptable cumulative metal additions to the soil over a 45-year period. Each province may also have their own requirements in addition to the federal ash applications as a soil amendment.

For instance, British Columbia's "Code of Practice for Soil Amendments" includes fly ash from wood combustion (other than wood that has been exposed to marine water) in the definition of a soil amendment. This code sets strict requirements for soil amendments, such as no substances exceeding the values shown in Table A.2 (in the appendix), no sharp foreign matter, such as glass or metal, and no more than 1% (dry weight basis) foreign matter (GoBC, 2017).

Table A.2 (in the appendix) provides a comparison of the limits of toxic elements in standards with those of wood ash for materials used as soil amendments. The bottom ash, primarily composed of rocks, sands, stones, and debris, is unsuitable as a soil amendment. However, MC ash and ESP ash are suitable for use as soil amendments as they contain no foreign materials or sharp objects. Moreover, the elemental/metal concentrations in MC ash and ESP ash were much lower than those established by the BC guideline for soil amendment. Therefore, ESP ash and MC ash of the representative mill were of great interest for use as soil amendments.

The use of fly ash from PPMs as a soil amendment has also been studied by Maheswaran et al. (2019) in Newfoundland, Canada. Common uses of wood ash in Canadian provinces/territories are shown in Hannam et al. (2016). It is essential to follow guidelines and regulations to use ash safely and sustainably and maintain soil quality for future generations.

3.2.3 Ash use as a composting recipe

The use of fly ash from PPMs as a composting recipe has increased over time due to its nutrient content, low costs, and chemical nature. In general, the C/N ratio in the composting recipe mix is 30 (Tchobanoglous et al., 1993). The content of nitrogen in wood ash is minimal as nitrogen is released with flue gases during hog combustion in boilers. Predominant essential elements present in ash are Ca and K. Other desirable elements include P, S, and Mg. They are smaller in concentrations in wood ash but are essential for plant growth.

Maintaining a moisture content of 50-70% in the composting mix is crucial to ensure optimal conditions for microorganisms during composting (CWMI, 1996). Luckily,

both wet and dry ash can be used for composting, depending on the moisture content of other available ingredients.

Fly ash's high pH level is perfect for increasing soil pH and neutralizing acidic soils. However, it is important to maintain an optimal pH level of 5.5-8.5 during the composting process. To ensure the best results, it is recommended to use up to 16% of wood ash at the beginning of the composting process (Kuba et al., 2008). Additionally, the final application of wood ash should be regulated based on the selection of a high amount of biochar, and toxic elements that are lower than the standard set by the CCME composting guidelines and OMRR for BC (OMRR, 2002). While the nutrient content and CCE of ESP ash are higher than MC ash, wood ash as a composting recipe needs to be regulated by the selection of plant types, application rate, and potentially toxic elements standards.

The MC and ESP ash from PPMs have high pH levels, high nutrient content, and greater CCE values compared to literature values of wood ash and limestones. Table A3 (in the appendix) shows that the nutrient content of Ca, Mg, S, K, and P, as well as the CCE of ESP ash were higher compared to MC ash. However, MC ash had a greater amount of biochar than ESP ash. When it comes to composting, MC ash is more desirable due to its dark colour, which gives the resulting compost a darker hue.

In a survey regarding the management of solid residues from the pulp and paper industry, it was found that 14% of the total ash residues generated by 67 responding mills were blended with wastewater treatment (WWT) sludges before the mixture was composted for land application (Elliott and Mahmood, 2015). The MC ash of a PPM has been used in biosolids composting in a municipality of BC for its high nutrient values and low toxic elements.

In summary, using fly ash from PPMs as a composting recipe is an excellent choice for those looking for a cost-effective and nutrient-rich option. With its high pH level, low toxicity, and high nutrient content, this ash is an excellent choice for creating optimal composting conditions.

3.2.4 Ash as a liming material

In general, the quality factors that are used to consider wood ash as a liming material are moisture, purity as determined by CCE, particle size, and plant species selection (Lickacz, 2002). Wood ash could be an excellent liming material for your soil. Wood ash is rich in essential nutrients such as P, S, and K, making it a great option for improving soil fertility. However, not all wood ash is created equal. The quality factors that determine wood ash as a liming material are moisture, purity, particle size, and plant species selection (Lundström et al., 2003; Manan and Abdullah, 2020). For best results, the wood ash should have less than 2% moisture content and a CCE value ranging between 25-60% (Risse, 2013).

Table A3 (in the appendix) compares the characteristics of mill ash, literature ash, and limestone. While bottom ash is not suitable for liming, MC and ESP ash are. Among the two, ESP ash is preferred due to its higher CCE value. CCE values for the MC ash and ESP ash were 32% and 53%, respectively.

In BC, the Code of Practice for Soil Amendments (CoPSA) governs the use of wood ash as a soil amendment/liming material. The CoPSA allows the use of 5 m³ wood ash (equivalent to 2,150 kg of wood ash with a specific gravity of 430 kg m⁻³) without going through the approval process, but larger-scale applications must go through a stringent approval process (GoBC, 2016). It is essential to maintain the levels of toxic metals in ash to ensure that the soil is safe for plant growth.

Approximately 6.3 million acres of land in western Canada have a pH of 6.0 or less (Lickacz, 2002). Maheswaran et al. (2019) showed that the agricultural land in Western Newfoundland was acidic and can be neutralized using fly ash generated from Corner Brook Pulp and Paper Ltd. As there is a high demand for liming materials in Canadian agricultural land, the ESP ash of the representative mill is a good candidate for this purpose. With high demand for liming materials in Canadian agricultural land, using wood ash can be an affordable and sustainable solution for soil improvement.

3.2.5 Ash use as an adsorbent

Sorption has been identified as the potential end use of biomass fly ash (Anderson et al., 2012; Gomez et al., 2020; Karim et al., 2013). The unburned carbon (biochar) contained in ash is the primary sorbent. Other factors to be considered are specific surface area and toxic metals.

Interestingly, MC ash had a higher LOI than ESP ash, which falls within the range of the FPIInnovations database (3-59%), as shown in Figure 3. Mills that have a high LOI (unburned carbon) can opt to separate this biochar fraction through dry or wet classification processes (Mahmood and Elliott, 2017) and sell this fraction (biochar) as low-grade activated carbon. MC ash, with its fineness and higher char content, is a better option than ESP ash as an adsorbing material, as indicated in Table A3 (in the appendix).

Furthermore, recent research has shown that fly ash can effectively remove contaminants from in-mill streams in pulp and paper facilities (Gao and Fatehi, 2018; Yonghui et al., 2018). The study targeted the removal of COD and lignocellulosic compounds, along with their associated colour, and found that unburned carbon adsorbs these compounds. Additionally, the inorganic fraction of ash plays a significant role in the removal efficiency, as the aluminum and calcium compounds cause coagulation, leading to the removal of contaminants (Yonghui et al., 2018).

It is an excellent way to maximize the benefits of biomass fly ash by using sorption. The biochar fraction can be used for various purposes, and the inorganic fraction of ash can significantly contribute to removing contaminants from in-mill streams, making it an effective and persuasive solution.

3.2.6 Ash use in forest/rural roads

Wood ash is a valuable resource that can be utilized for rural or forest road construction, but there aren't any specific standards for its use. However, standards for coal ash use as a cementitious material can be applied to wood ash, specifically for ESP fly ash, as indicated in Table A.1 (in the appendix). The use of ash in this area has been document-

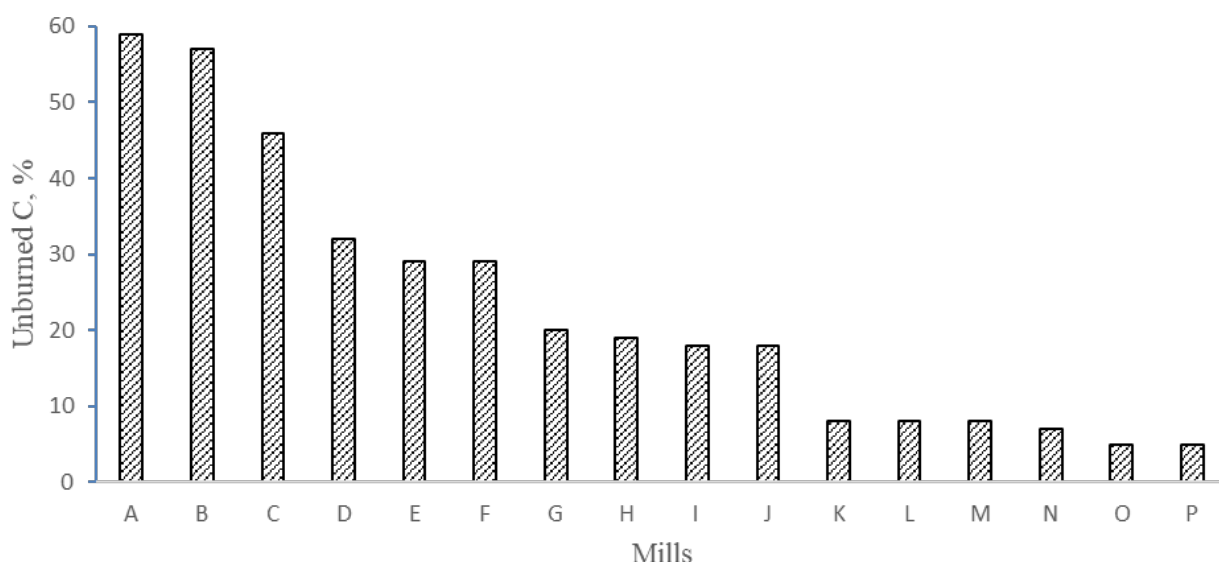


FIGURE 3: Unburned carbon content of fly ashes from 18 pulp and paper mills (FPInnovations database).

ed in various literature sources, making it a viable option (Minergy, 2021; Dalton, 2018; Fava et al., 2011).

One of the major parameters to be considered for wood ash for road construction is the leaching test of toxic elements. Cherian and Siddiqua (2021) conducted a leaching test of pulp paper mill fly (PFA) ash from the studied mill. Their study on leachate studies of PFA and PFA-stabilized soils confirmed that the cementation process reduced the bio-availability of toxic metals in the treated soil by encapsulating them in newly formed water-stable compounds. This suggests that PFA can be a safe raw material for environmentally friendly applications.

Particle size is a critical parameter when using ash for road stabilization, as it significantly affects the free lime content. The analysis required to determine its suitability as a road stabilization material includes free CaO content, total solids and LOI, inorganic elemental composition, carbon species, acid neutralization capacity, and leaching properties determined through field evaluations on test sites.

Although the representative mill did not have all the necessary parameters to consider wood ash for rural or forest roads, based on the two main criteria of CaO and LOI, it was observed that ESP ash was the most suitable, followed by MC ash, and finally bottom ash (Table A.1 in the appendix).

Geotechnical studies (Hottenroth et al., 2003) have shown that ash from the combustion of lignocellulosic materials and pulp mill residues improves soil-bearing capacity when the free CaO content of the ash is higher. The higher CaO content also reduces soil moisture and gravel requirements (Supanci and Obernberger, 2011). The CaO/SiO₂ ratio, which is the basicity of ash, is another essential parameter to predict ash performance as a soil or road stabilizer.

In conclusion, using wood ash for rural or forest road construction is a cost-effective and sustainable solution. With the right analysis and selection, wood ash can be used as a reliable and effective road stabilization material.

3.2.7 Ash use in waste stabilization/solidification

When it comes to stabilizing and solidifying waste, it's crucial to use the right binding agent. The process involves two steps: stabilization to reduce toxicity, and solidification to immobilize toxic elements. Cement, fly ash, or kiln dust can all work as binding agents, but when using fly ash, it must meet ASTM (C 618) standards for either Class C or Class F (Table A.1 in the appendix).

Choosing wood ash for waste stabilization/solidification (S/S) requires careful consideration of several factors, such as waste characterization, compatibility between cementing and waste materials, durability of treated waste or soil, and cost-effectiveness (Shi and Spence 2004, Trussell and Spence 1994). BC Hazardous Waste Guide recognizes S/S as an approved method for treating hazardous wastes containing asbestos, chromium, cyanides, fluorides, and heavy metals (GoBC, 2016).

Table A.1 (in the appendix) indicates that using bottom ash is not recommended for this purpose. Instead, ESP ash is the preferred option, but if the waste odour is an issue, MC ash is the better choice.

Waste solidification/stabilization has been used successfully for years to clean up contaminated sludge and soil in Arkansas. Limestone, fly ash, and Portland cement were used to treat 40,000 cubic yards of sludge and soil in the pit (USEPA, 2012).

In 2013, the Kruger mill in Brompton Ville, Quebec, Canada became the first pulp and paper mill in Canada to have its fly ash certified as a cement substitute, with substitutions as high as 20% (Minergy, 2021). This end-use application is the principal one currently being practiced by the mill to manage its fly ash by solidifying hazardous waste.

Therefore, to ensure the safe and effective stabilization/solidification of waste, binding agents need to be carefully chosen, considering all the factors mentioned above. The use of fly ash and cement has already been proven to work to keep the environment safe.

3.2.8 Ash use for mine remediation

Mining industries are in dire need of an effective solution to neutralize mine tailings, control acid mine drainage (AMD), and remediate abandoned mining sites. Fortunately, recent research has shown that ash may hold the key to solving these problems (Somerset, 2005; Wang, 2009). Ash contains alkaline minerals such as CaO, MgO, K₂O, CaCO₃, and Ca(OH)₂, which can be an effective substitute for purchased alkalinity (Somerset 2005). This means that fly ash generated from the coal industry and woody residues in pulp and paper power boilers can significantly reduce the acidity of waste sites (Hu et al., 2021; Yehevis et. al., 2009). By using ash for mine remediation, we can avoid the use of calcium oxide or limestone, which are commonly used but come at a high cost. Additionally, wood ash contains cementitious and pozzolanic materials that make it an ideal candidate for the mine remediation.

However, it is important to note that not all ash is generated equal. To ensure that ash is effective for mine remediation, it must be characterized by its alkaline content. By prioritizing ESP ash first, followed by MC ash and bottom ash.

3.2.9 Other end uses for ash residues

Ash has several potential applications based on its crystalline content. It can be used as a fire retardant for low-density polyethylene materials, making it an ideal material for construction and manufacturing (Dong et al., 2015; Porabka et al., 2015). Additionally, it can be used for vitrification in applications requiring glass aggregates, such as roofing shingles and ceramic processing (Minergy, 2021; Cheeseman, 2003). The key minerals - SiO₂, Al₂O₃, and CaO - present in ash are critical for these end uses. The inert fraction of ash is low-volume, making it cost-effective and efficient. Fly ash could also be considered for these applications if the LOI is low. However, wood ash has a higher LOI than the common natural fire retardants, such as concrete and brick making it unsuitable for use as a fire retardant. Upon analyzing the results provided in Table A.1 of the appendix, it is evident that the bottom ash from the representative mill, with the lowest LOI among the three ash streams studied and falling below both the ASTM C618 and CSA 4000 standards, stands out as the optimal option for use as a fire retardant. By utilizing ash effectively, we can reduce waste and create sustainable solutions for a better future.

3.3 Prioritizing the wood ash applications for the representative mill

The quality of wood ash determines its potential applications. To produce high-quality ash, segregation of wood ash is necessary. The representative mill produces three types of wood ash, each with unique characteristics and qualities. As illustrated in Table 2, the bottom ash has distinct applications compared to the other types, and it's preferred for use as a gravel substitute in concrete structures and as a fire retardant. MC ash has a high char content, and therefore, it can be used for adsorbing materials, stabilizing waste, and composting. On the other hand, ESP ash

has better pozzolanic and cementitious properties, making it the best choice for concrete-related applications. It's also an excellent mining material. When it comes to agricultural liming, ESP ash is a better choice than MC ash because of its higher CCE values. Choosing the right type of wood ash can make a significant difference in the outcome of any project.

3.4 Why inland mills are preferred to coastal mills

The pulp paper mills in British Columbia can be found either in the coastal areas or inland. The coastal mills rely on logs that are transported and stored in marine water, which increases the risk of saltwater contamination. This contamination can lead to the production of toxic chlorinated organics, such as dioxins and furans, which are hazardous to the environment. Therefore, it is essential to implement strict environmental regulations, especially for coastal mills. On the other hand, inland mills are unaffected by this type of contamination.

In both inland and coastal mills, the wood ash use must comply with BC regulations. Fortunately, the wood ash generated from inland mills can be used as a soil amendment if it complies with the Fertilizer Act (1997) of the Canadian Food Inspection Agency (CFIA). Wood ash has a pH of 10-12.5, making it a non-hazardous material suitable for use as a soil amendment. However, wood ash from coastal mills must comply with regulations that prohibit agricultural use, as stated in the code of practice for soil amendment in BC (GoBC, 2016). Nonetheless, such ashes can be used in the concrete industry or waste solidification.

Table 3 highlights the potential use of biomass ash generated in inland mills as a cementitious material, agricultural-related applications, and others. Meanwhile, Table A.4 (in the appendix) outlines the regulations concerning the land application of ash. It is crucial to adhere to these regulations to protect both the environment and public health.

TABLE 3: Potential applications of wood ash: bottom ash, multi-clone (MC) ash, and ESP ash of the studied mill.

Ash uses	Bottom ash	MC ash	ESP ash
Cementitious related applications			
concrete material	●	●●	●●●
road/forest ¹	●	●●	●●●
waste stabilization	●	●●●	●●●
Agriculture related applications			
oil amendment	●	●●	●●
composting recipe	●	●●●	●●
liming materials	●	●●	●●●
Other applications			
adsorbent	●	●●●	●●
mining	●	●●	●●●
fire retardant (other use)	●●●	●	●●

●: minimal use, ●●: moderate use, and ●●●: best potential use of an ash stream of the studied mill.

¹ In road construction, bottom ash is a better substitute for gravel than MC and ESP ashes.

4. CONCLUSIONS

The effective and optimal use of ash is crucial, and it depends on careful consideration of specific factors such as end-user ash quality requirements and environmental regulations. An in-depth exploration of these factors in a representative pulp and paper mill in BC, Canada has uncovered valuable insights that can revolutionize ash application methods in other mills. This analysis has revealed that each wood ash stream, encompassing bottom ash, MC ash, and ESP ash, possesses unique characteristics that render them ideal for specific applications. It is imperative to consider that environmental regulations and standards also wield significant influence over ash utilization, with different regulations governing various ash applications. The initial evaluation of wood ash application, based on the wood ash characterization of the studied pulp paper mill, indicated that the ESP ash is the most suitable for cementitious-related applications and as a liming agent for agricultural land application, while the MC ash is best suited for waste stabilization, composting recipes, and adsorbents. On the other hand, the bottom ash demonstrates optimal efficacy as a fire retardant. These findings emphasize the critical importance of aligning end-user ash requirements with the ash quality of a mill to pinpoint promising applications for comprehensive follow-up. By taking all these factors into account, ash utilization can be optimized, leading to the implementation of sustainable and environmentally friendly practices.

ACKNOWLEDGEMENTS

The authors acknowledge the financial support of Mitacs Accelerate Fellowship, UBC – Okanagan Campus and FPIInnovations' Member Companies. The authors would like to thank the technical staff of participating mills and FPIInnovations (Sylvain Fauteux, Paul Camolinos and Allan Elliott) who were involved in sample collection and analyses.

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A SUSTAINABLE APPROACH TO RECYCLING WASTE: EXAMPLES OF THE APPLICATION OF 3D PRINTING TO EXTEND THE LIFE CYCLE OF MATERIALS

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Article Info:

Received:
28 May 2024
Revised:
24 July 2024
Accepted:
26 July 2024
Available online:
7 September 2024

Keywords:

Waste
3D printing
Fused Filament Fabrication
Circular Economy
Recycle

ABSTRACT

The environmental questions emerged in recent decades have caused a transition from a linear economy, which involves the extraction of raw materials, production, distribution, consumption and final disposal of a good, to a circular economy (CE). CE is based on the reuse of materials in subsequent production cycles by extending their useful life, with the aim of reducing and if possible, eliminating any waste. Fused Filament Fabrication (FFF), a 3D printing technique, has emerged in recent years as a promising methodology for green and sustainable purposes. The use of natural and biocompostable materials for the production of filaments for FFF, such as poly(lactic acid) (PLA), has made this 3D printing technique one of the most promising in several fields. Furthermore, scientific research is focusing on improving the sustainability of primary materials used in 3D printing, through the development of composite filaments from green or recycled materials. This paper presents some applications concerning the development of innovative and sustainable materials focused on reducing the environmental impact of conventional FFF materials, carried out in recent years in our laboratories. In particular, various composite filaments incorporating poly(lactic acid) (PLA) and different handicraft or agro-industrial waste materials, including olive wood waste (OW), cocoa bean shell waste (CBSW), ceramic waste (CW) and Lecce stone waste (LSW), have been produced by extrusion. A comprehensive analysis was conducted on the innovative biocomposites developed, including structural, morphological, thermal and mechanical evaluations, and the comparison of the results obtained with the different subproducts is presented in the paper. Finally, some simple real objects first reproduced manually, were later 3D printed using the developed biocomposite filaments, demonstrating the effectiveness of FFF in successfully recycling artisanal and agro-industrial waste within the same production company, reducing costs and supporting the CE.

1. INTRODUCTION

Excessive use of synthetic materials (such as metallic, ceramic, stone materials) and plastic, that are generally difficult to discard, has caused increasing environmental problems. Stringent European waste management legislation emphasizes the importance of creating new material recycling strategies, highlighting a necessity for a complete transition from a linear economy to a circular economy (CE) (Ghisellini et al., 2016; Kirchherr et al., 2017). Fused Filament Fabrication (FFF), an Additive Manufacturing (AM) technique, has recently emerged in several industries as a versatile, low-cost and sustainable methodology (Fico

et al., 2022). FFF allows to reduce the disproportionate use of raw materials compared to traditional manufacturing processes, explore innovative methods of waste management and valorization, and at the same time extend the life cycle (Fico et al., 2022). Usually, the heating element of commercially available FFF machines achieves a maximum working temperature of about 300°C, so thermoplastic polymers have until now remained the most widely used materials in 3D printing, due to their processability, the multiplicity of types and colors available on the market, versatility and sufficient mechanical properties conferred to printed objects (Vyavahare et al., 2020). Among them,



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polylactic acid (PLA) is perhaps the most widely used thermoplastic polymer in FFF printing, since it is of natural origin, biodegradable and biocompatible (Mikula et al., 2021). However, despite its versatility and environmental benefits, PLA shows great sensitivity to high temperatures and moisture (Rizzo et al., 2023). In addition, the final properties of PLA-printed objects may not be suitable for structural applications (Algarni & Ghazali, 2021).

Over time, rapid advances in materials engineering have facilitated the development of innovative polymer-based composite for use in FFF printing. In particular, innovative filaments for FFF have been developed, using natural or recycled materials as fillers for the polymer matrix (Rajendran Royan et al., 2021; Rett et al., 2021); in the first case, green, natural, biodegradable and low-density composite filaments for FFF were obtained (Carichino et al., 2023; Kariz, Sernek, Obućina, et al., 2018); in the second case, scientific research has led to the development of composite filaments with better mechanical performance compared to pure polymer, with economic and ecological benefits (Corcione et al., 2017; Esposito Corcione, Scalera, et al., 2018; Fico et al., 2023).

For example, several scientific studies have developed and tested innovative filaments for FFF containing wood from different plant species as a filler added to the polymer matrix, either in the form of particles or fibers (Das et al., 2021; Fico et al., 2022; Vásquez-Garay et al., 2021; Xu et al., 2018). The FFF filaments that have been produced are non-toxic, completely biodegradable, have good elasticity but in some cases have a high stiffness and hygroscopicity compared to traditional polymer; this is the reason that scientific research continues to propose several studies with the aim of increasing the final properties of wood-based filaments. Even less used in the scientific research are cocoa bean shell waste (CBSW), whose production is growing due to the ever-increasing consumer demand for chocolate, currently disposed of as organic fertilizer, animal feed or fuel, even if with negative environmental effects. However, a few research conducted in recent years have also seen its use as a filler to the polymer matrix (poly(lactic acid) or polycaprolactone) in 3D printing (Fico et al., 2024; Puglia et al., 2016; Sanyang et al., 2017; Tran et al., 2017). The condition is different for non-biodegradable inorganic materials, such as ceramic or stone, whose growing use in the artistic, engineering, construction, biomedical, aerospace and automotive fields cause a considerable accumulation in the environment and greater difficulties and costs associated to their disposal. These requirements have given rise to many works in the scientific literature on the recycling of these materials for the production of composite filaments used in FFF, with excellent thermal and mechanical properties, and used specifically for biomedical or tissue engineering applications (Esposito Corcione et al., 2019; Esposito Corcione et al., 2018; Fico et al., 2022).

Our research group on "Additive Manufacturing" at the University of Salento (Lecce, Italy) has also been actively involved for years in the recycling of several types of organic and inorganic waste mainly supplied by local artisans and manufacturers, through FFF technology (Corcione et al., 2020; Ferrari et al., 2020, 2022; Lionetto & Esposito Corcione,

2021). The main goal of our research is not only to reduce the environmental impact, to promote the principles of the CE by giving new life to raw materials, but to produce new 3D objects starting from agro-industrial and artisanal waste, which as a novelty can help producers to reduce waste disposal practices and costs and at the same time obtain new objects that preserve the unique value of their creations.

This paper presents some of these applications regarding the development of innovative and sustainable materials for FFF printing. In particular, different composite filaments based on poly(lactic acid) (PLA) and several artisanal or agro-industrial waste materials have been produced by extrusion process. A complete structural-morphological, thermal and mechanical characterization was carried out and the results were compared with each other. Finally, some simple real objects were faithfully reproduced by hand and then three-dimensionally to be 3D printed, showing the adaptability of the FFF methodology as a low-cost, easy and sustainable solution to recycling materials in the same waste production company (Figure 1).

2. EXPERIMENTAL

2.1 Materials and methods

In this work, the applicability of artisanal and agro-industrial waste recycling of FFF printing and its possibility to extend the life cycle of materials were tested through the development of biocomposite filaments for FFF, from some scraps provided by local companies. Waste of organic and inorganic origin was added in powder form as filler to the polymer matrix selected for its green properties, the poly(lactic acid) (PLA), and extruded to form the biocomposite filaments later used for 3D printing.

Details of the materials and methodologies used are provided in the following paragraphs.

2.1.1 Materials

For the production of the new biofilaments potentially used for the 3D printing technology known as FFF, the following scraps were used: poly(lactic acid) (PLA), cocoa bean shell (CBSW), Olive wood (OW), Lecce stone (LSW) and ceramic waste (CW).

Ingeo 4043D poly(lactic acid) (PLA) was purchased in pellet form (diameter of 5 mm) from the company NatureWorks LLC (Blair, NE, USA). PLA is characterized by a melt flow index (MFI) of 6 g/10 min at a temperature of 210°C and a density of 1.24 g cm⁻³. The pellet was stored in an oven at 60°C for 24 h, ground using the Retsch ZM 100 ultracentrifugal mill (Retsch GmbH, Haan, Germany), obtaining a particle diameter of about 0.75 mm.

The organic waste used in the production of new filaments are cocoa bean shell waste (CBSW) and Olive wood (OW). The CBSW derive from the chocolate processing residues of the company "Maglio Arte Dolciaria S.R.L." (Maglie, Lecce, Italy). CBSW was supplied in the form of a coarse powder, with a brown color and a chocolate-like odor. The Olive wood (OW) waste was instead supplied by "I Lumi del Faso di Sante Musa", member of the "Fatto in Bottega" association (Brindisi, Italy). CBSW and OW were oven dried at 60°C and subsequently ground to a diameter

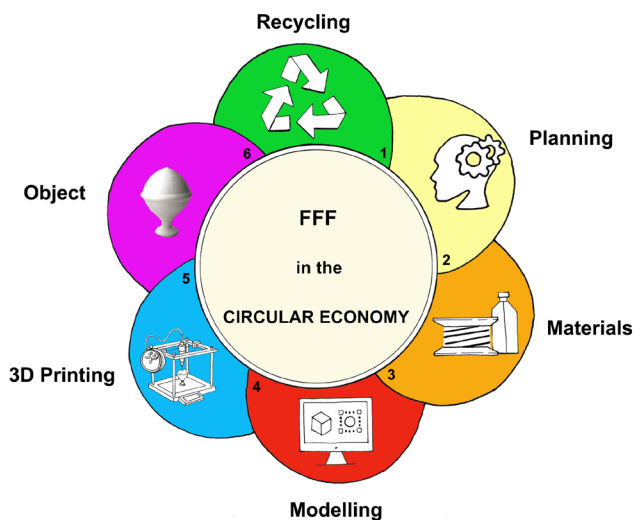


FIGURE 1: Applicability of FFF printing to Sustainability, Recycling and Circular Economy.

of about 0.25 mm, using a Retsch ZM 100 ultracentrifugal mill (Retsch GmbH, Haan, Germany).

The inorganic waste used in the production of new filaments are Lecce stone (LSW) and ceramic (CW). The Lecce stone (LSW), a local calcarenite, was supplied by the company Décor srl (Monteroni, Lecce, Italy), in the form of a monolithic block having a water content of 88% by weight. The stone was then first dried in an oven at 60°C (reducing the water content to 20% by weight) and then crushed with a mortar and sieved through a fine mesh sieve (0.25 mm diameter). Instead, the CW waste was supplied by the ceramic laboratory of the “Fatto in bottega” association (Brindisi, Italy). The waste was subjected to an initial washing under

tap water, followed by drying in an oven at 100°C. Finally, the waste was reduced to powder with a diameter of approximately 0.02-0.03 mm using first manually the hammer and then the planetary ball mill of the Ceramic Instruments company (Sassuolo, Italy).

2.1.2 Experimental procedure

The experimental activity carried out mainly consisted of following steps (Figure 2):

- Recycling of artisanal and agro-industrial waste materials through their use as an additive to the PLA polymer matrix and development of new biofilaments for FFF printing;
- 3D printing of simple prototypes (bars) starting from composite filaments used for printability evaluation tests;
- Identification of real models to be returned to the companies producing the waste and their manual reproduction;
- Three-dimensional reproduction and 3D printing.

The following composite filaments for FFF were developed starting from the waste materials indicated previously: the first consisting of 100 wt. % of PLA, the other filaments were developed starting from 90 wt. % of PLA and 10 wt. % of the several wastes CBSW, OW, LSW, CW. The denomination of the developed filaments is shown in Table 1. To obtain the new filaments, the PLA and waste powders were first mixed manually at room temperature and then inserted into the 3Devo Composer 450 Filament Maker single-screw extruder (Utrecht, The Netherlands), using the parameters reported in Table 2.

Simple prototypes (bars) were printed using the Creality CP-01 printer (Creality, London, United Kingdom) starting

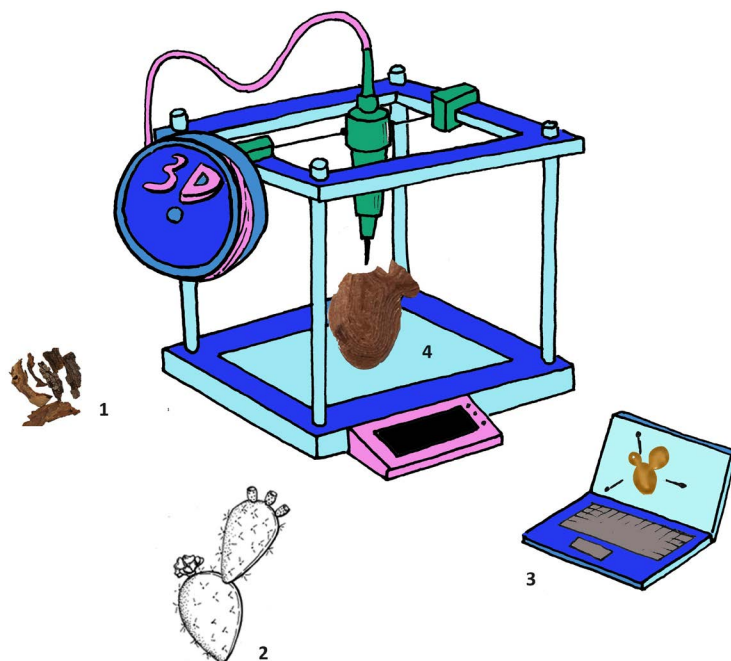


FIGURE 2: Chart of the experimental procedure: recycling of artisanal and agro-industrial waste materials (1), real object models and their manual reproduction (2), 2D and 3D reproduction (3), 3D printing (4).

TABLE 1: Biofilament and bars labels and weight composition (wt. %).

Form	Label	Composition (wt. %)
Filament	100PLA	100 PLA Ingeo 4043D
Filament	90PLA/10CBSW	90 PLA Ingeo 4043D and 10 cocoa bean shell waste
Filament	90PLA/10OW	90 PLA Ingeo 4043D and 10 Olive wood powder waste
Filament	90PLA/10LSW	90 PLA Ingeo 4043D and 10 Lecce stone powder waste
Filament	90PLA/10CW	90 PLA Ingeo 4043D and 10 ceramic waste powder
Bar	100PLA_3D	100 PLA Ingeo 4043D
Bar	90PLA/10CBSW_3D	90 PLA Ingeo 4043D and 10 cocoa bean shell waste
Bar	90PLA/10OW_3D	90 PLA Ingeo 4043D and 10 Olive wood powder waste
Bar	90PLA/10LSW_3D	90 PLA Ingeo 4043D and 10 Lecce stone powder waste
Bar	90PLA/10CW_3D	90 PLA Ingeo 4043D and 10 ceramic waste powder

TABLE 2: Extrusion process parameters for biofilaments.

Extrusion Process Parameters	100PLA	100PLA/10CBSW	100PLA/10OW	100PLA/10CW	100PLA/10LSW
Screw speed (rpm)	3.5	4	4	3.5	3.5
Fan cooling speed (%)	0	30	0	0	0
Feed zone temperature (°C)	170	165	185	200	220
Compression zone temperature (°C)	185	180	190	190	210
Metering zone temperature (°C)	190	185	185	210	190
Die temperature (°C)	200	165	187	220	170

from the developed filaments, with the aim of carrying out evaluation tests of the final properties and in accordance with the European standard ISO178:2014 (Table 1). Fusion 360 software (Autodesk, San Rafael, CA, USA) was used to create the CAD model and Cura software (Ultimaker B.V., Utrecht, the Netherlands) was used to obtain the printer-readable G-Code file. The following printing parameters were used: extrusion temperature at 200°C, platen temperature at 50°C, print speed at 50 mm/s, infill at 100%, and cooling fan speed at 100%.

An evaluation of the final properties of the biofilaments developed in the work was carried out by observing the surfaces and sections under a scanning electron microscope (SEM), model Zeiss E Evo 40 (Oberkochen, Germany) and measuring the thermal features with the DSC instrument (Mettler Toledo DSC1 StareSystem), temperature range 25°C to 200°C, heating rate of 10°C/min. For each filament, 5 measurements were carried out. An evaluation of the final properties of the biocomposites was carried out by bending testing on bars using a Lloyd LR5K dynamometer (Lloyd Instruments Ltd., Bognor Regis, UK) at a test speed of 2 mm/min and a support spacing of the 64 mm specimen. For each sample 5 measurements were carried out. The printability of the new biofilaments was also evaluated with magnify images of bars sections after mechanical tests obtained with the Dino-Lite digital microscope (AnMo Electronics Corporation, New Taipei City, Taiwan).

Finally, some simple objects to be displayed in the companies that supplied the waste materials were identified as an application example of the total experiment. These were initially reproduced manually in two dimensions; the same two-dimensional models were transformed into CAD format using Fusion 360 software (Autodesk, San Rafael,

CA, USA) or Rhinoceros software (Robert McNeel & Associates, USA) and converted to G-Code files using Cura software (Ultimaker B.V., Utrecht, The Netherlands). Specifically, 3D objects printed from CBSW and OW waste-based green filaments were made using the Creality CP-01 printer (Creality, London, UK), an extrusion temperature of 190°C, plate temperature of 50°C, printing speed of 60 mm/s and infill of 20%. Objects obtained from LSW and CW filaments were printed using 3DPRN LAB printer (3DPRN company, Castiglione M.R., Italy), extrusion temperature of 200°C, plate temperature of 50°C, printing speed of 40 mm /s and infill of 100%.

3. RESULTS AND DISCUSSION

New and sustainable composite filaments were produced by the extruder with a diameter of 1.75 mm. Following the extrusion of the new filaments from the waste materials, some initial evaluations under a scanning electron microscope (SEM) were made. While the 100PLA filament preserves the smooth surface (Figure 3-A), the addition of fillers based on waste materials causes a different surface property. Among the different additives used for the PLA polymer matrix, OW scraps show the greatest increase in surface roughness and aggregation of particles into larger lumps (Figure 3-B). Tao et al. 2017 in the paper regarding the incorporation of wood flour into the PLA matrix to produce 3D printing filaments, highlight an increase in roughness compared to neat polymer filament (Tao et al., 2017). In addition, their paper shows poor interfacial adhesion between the hydrophilic wood particles and hydrophobic polymer particles, caused by the difference in polarity at the interface of the two materials, which causes problems

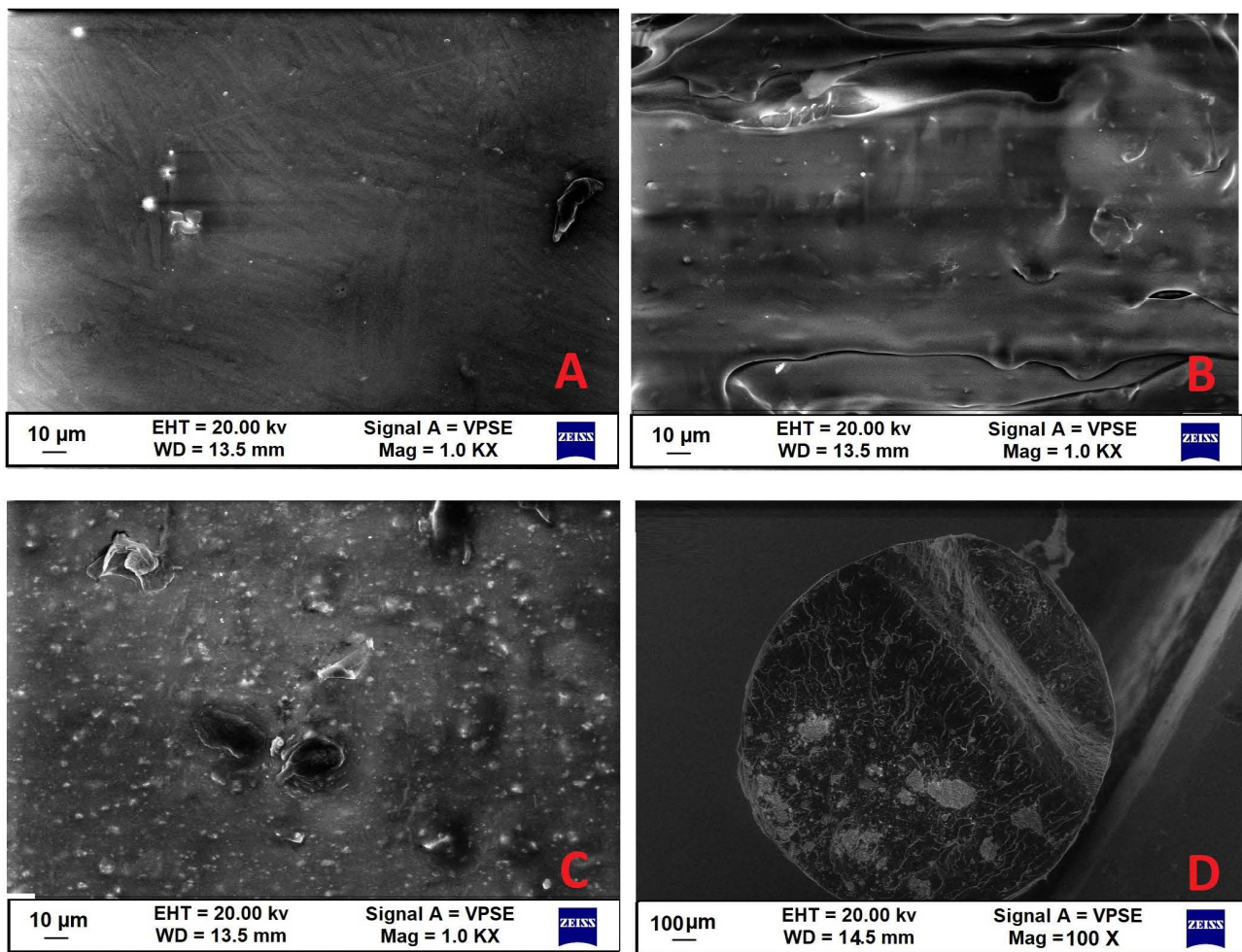


FIGURE 3: SEM images of filaments: 100PLA external filament surface (A), 90PLA/100W external filament surface (B), 90PLA/100CW external filament surface (C) and 90PLA/100CW filament section (D).

of biocomposite inhomogeneity and preferential fracture planes. The inhomogeneity of the developed biocomposite may also be due to the different diameter of the particles in the blend used, and according to the scientific literature can sometimes be reduced through the use of compatibilizing agents. For example, Xie et al. 2017 studied the behavior of two plasticizers, glycerol and tributyl citrate, to improve the interaction between poplar wood flour and PLA, demonstrating better compatibility following the addition of 4% of tributyl citrate (Xie et al., 2017).

Otherwise, the new biofilaments containing CBSW or inorganic wastes (LSW and CW) fillers have a fairly homogeneous surface, although it is roughened in 90PLA/10LSW and 90PLA/10CW filaments (Figure 3-C). However, all biocomposites show particle aggregates in section, especially the 90PLA/10CW filament (Figure 3-D). Inhomogeneity may also be in these cases due to different particle diameters between the polymer matrix and the fillers.

The results of the DSC analysis of PLA-based filament and waste biofilaments are shown in Table 3, in which the glass transition temperatures T_g were measured using the inflection point. The 100PLA filament has a T_g equal to 59.19°C, values similar to those reported in the literature

(Algarni & Ghazali, 2021; Fico et al., 2022). The addition of organic waste fillers to the polymer matrix causes a reduction of the glass transition temperatures compared to pure polymer in the 90PLA/10CBSW and 90PLA/100W samples (Table 3). This indicates that the CBSW and OW particles interacted with the matrix PLA on a molecular scale, causing some changes in the mobility of the polymer chains. The similar thermal behavior of biocomposites based on organic waste is also observed in the crystallization and melting processes. The crystallization temperature T_c of the 90PLA/10CBSW and of the 90PLA/100W filaments decreases compared to the 100PLA at 106.99°C

TABLE 3: Thermal properties obtained from DSC analysis of polymeric neat materials: glass transition temperature (T_g), temperature of crystallization (T_c) and melting temperature (T_m , T_{m1}).

Label	T_g (°C)	T_c (°C)	T_m (°C)	T_{m1} (°C)
100PLA_3D	59.19	124.39	154.68	/
90PLA/10CBSW_3D	56.66	106.99	148.12	155.40
90PLA/100W_3D	53.43	105.67	148.67	154.50
90PLA/10LSW_3D	61.30	125.43	156.54	/
90PLA/10CW_3D	59.40	116.11	152.24	/

and 105.67°C, respectively. Moreover, the thermograms of both biofilaments manifest a double melt peak (T_m and T_{m1}) unlike pure PLA and filaments containing inorganic fillers (LSW and CW). The presence of the double melt peak has also been observed by several authors in the literature and has been attributed to the presence of biocomposites to different behavior between the polymer and the organic filler, as well as to different degradation temperatures of cellulose and hemicellulose, the main constituents of CBSW and OW particles (Bryś et al., 2016; Leonard & Berrio, 2021; Tsujiyama & Miyamori, 2000).

The result of the DSC analysis of the 90PLA/10LSW sample show a negligible increase in the temperatures T_g , T_c and T_m , compared to the 100PLA sample (Table 3). While the 90PLA/10CW sample retains thermal properties similar to PLA (Table 3). The double melting peak is absent in both thermograms.

After 3D printing of bars from the developed composite filaments, mechanical tests were performed and the overall results are shown in Table 4.

The Young's modulus (E) of 2.74 ± 0.03 GPa, the flexural stress (σ_R) of 102.5 ± 4.12 MPa and the flexural strain (ϵ_R) of $4.1 \pm 0.3\%$ relative to the virgin PLA 3D bars, fall into the ranges reported in the literature (Fico et al., 2022). The flexural test results of the CBSW-based bars indicate a value of σ_R (MPa) about half as low as that of the pure poly(lactic acid) bars. ϵ_R (%) also decreases to about $3.7 \pm 0.2\%$. These values, in accordance with the scientific literature (Puglia et al., 2016; Tran et al., 2017), indicate an increase in fragility and decrease in ductility of CBSW-based composites compared to PLA bar. However, an increase in Young's modulus (E) of 3.14 ± 0.08 GPa was measured for the CBSW-based biocomposite. As demonstrated in the literature (Rothon, 2003), CBSW particles create polymer phase modifications by interacting with PLA to form an adsorbed polymer interphase. The value of Young's modulus therefore depends on the thickness of the interphase formed and the CBSW-PLA bond, and thus on the shape, size, percentage amount of filler particles and surface area of interaction. The CBSW particles seem to have inhibited the mobility of the polymer chain of the PLA matrix, establishing a strong bond and thus increasing the stiffness of the 3D sample.

A deterioration of the mechanical properties is observed in wood-based biocomposites. Specifically, the flexural modulus decreases 2.1 ± 0.05 GPa, but the most significant changes are observed for σ_R (MPa) and ϵ_R (%) (Table 4).

Das et al., 2021 in their review of the use of wood for 3D printing technology, report several studies showing a

reduction in mechanical properties, at least for substantial percentages of wood added to the polymer matrix (Das et al., 2021). Le Guen et al. 2019, on the other hand while showing a decrease in flexural strength and deformation with the addition of wood powder, indicate an increase in flexural modulus (Le Guen et al., 2019). The phenomenon described in the paper is attributed to the poor interfacial bonding between polymer and wood particles, which does not promote encapsulation of the particles in the polymer binders, resulting in low bonding and the presence of voids in the biocomposites causing a decrease in mechanical strength. According to Kariz et al. 2018 the low affinity between the polymer and the wood particles also results in some difficulties in the melting process of the composite biofilament, resulting in nozzle clogging during the 3D printing process (Kariz et al. 2018; Kariz et al., 2018).

Overall, the results of bending tests obtained from the addition of LSW and CW waste powders are different from the OW-based composites. Specifically, while the σ_R (MPa) and ϵ_R (%) values decrease, the Young's modulus (E) of LSW- and CW-based composites increases, compared to those of virgin PLA (Table 4). As a result, the mechanical performance of the developed biocomposites is sufficient when the addition of fillers reaches 10 wt. %, even if successful mixing of the particles in the polymer matrix has not occurred.

The study of the printability of the biocomposite filaments developed with the waste materials was also accompanied by a Dino digital optical microscope observation of the breaking surfaces of the 3D bars, subjected to the bending tests (Figure 4).

As expected, the 100PLA_3D sample has perfect adhesion between layers (Figure 4-A, F). Good printing properties were obtained in the 90PLA/10CBSW_3D bar, whose magnified image shows good adhesion between layers (Figure 4-B, G) and the 90PLA/10CBSW filament manifests excellent printability. In addition, the organoleptic features of cocoa are preserved in the printed bars, despite the low percentage of filler relative to the polymer matrix and the high temperature melting processes subjected (extrusion and printing). Difficulties due to the printing process occur for the 90PLA/10OW and 90PLA/10LSW filaments, while the breaking surfaces of the composite bars (Figure 4-C, H and Figure 4-D, I, respectively) show poor adhesion between the layers; deformation and voids are also evident in some areas. In addition, the presence of increased porosity of the samples is observed, which probably inhibits the mechanical performance of the composite material, especial-

TABLE 4: Mechanical flexural properties: E flexural modulus of elasticity (GPa), σ_R flexural stress (MPa), ϵ_R flexural strain (%).

Label	E (GPa)		σ_R (MPa)		ϵ_R (%)	
100PLA_3D	2.74	0.03	102.5	4.12	4.1	0.3
90PLA/10CBSW_3D	3.14	0.08	56.39	1.53	3.7	0.2
90PLA/10OW_3D	2.10	0.05	40.90	2.61	3.3	0.2
90PLA/10LSW_3D	3.10	0.03	70.02	2.45	3.9	0.3
90PLA/10CW_3D	3.16	0.02	76.07	5.42	3.7	0.2



FIGURE 4: Magnified images (50x) of the 3D bars after bending test: printing surface of 100PLA (A), 90PLA/10CBSW (B), 90PLA/10OW (C), 90PLA/10LSW (D) and 90PLA/10CW (E); breaking surface after bending test of 100PLA (F), 90PLA/10CBSW (G), 90PLA/10OW (H), 90PLA/10LSW (I) and 90PLA/10CW (L).

ly in the wood sample. Finally, 90PLA/10CW_3D, although showing some air voids between the layers, shows good printability (Figure 4-E, L).

The study of the morphological and thermal features of the new biofilaments and the printability and mechanical properties of the bars printed using FFF were finally accompanied by some application examples that the authors want to show below to attest to the success of the overall experiment.

In collaboration with the companies producing the waste, the authors selected some real objects to reproduce starting from the recycling of raw materials. Specifically, in most cases these are objects widespread by hand or naturally in Puglia (Italy), such as the so-called pumo, the

monument in Piazza Sant' Oronzio in Lecce or the Opuntia Ficus Indica plant, or objects that retain the shapes of the original raw materials, for example chocolates. The manual and faithful reproduction of the real objects (Figure 5) was followed by the simplified reproduction of the respective CAD models (Figure 6). The CAD files were converted into GCode and the prototypes were printed using FFF, both in the case of 90PLA/10CBSW, 90PLA/10CW biofilaments with good printability, and in the case of 90PLA/OW and 90PLA/10LSW with sufficient printability.

The experimental activity demonstrates the effectiveness of the FFF methodology in recycling artisanal and agro-industrial waste within the same production company, in favor of important issues such as the circular econo-

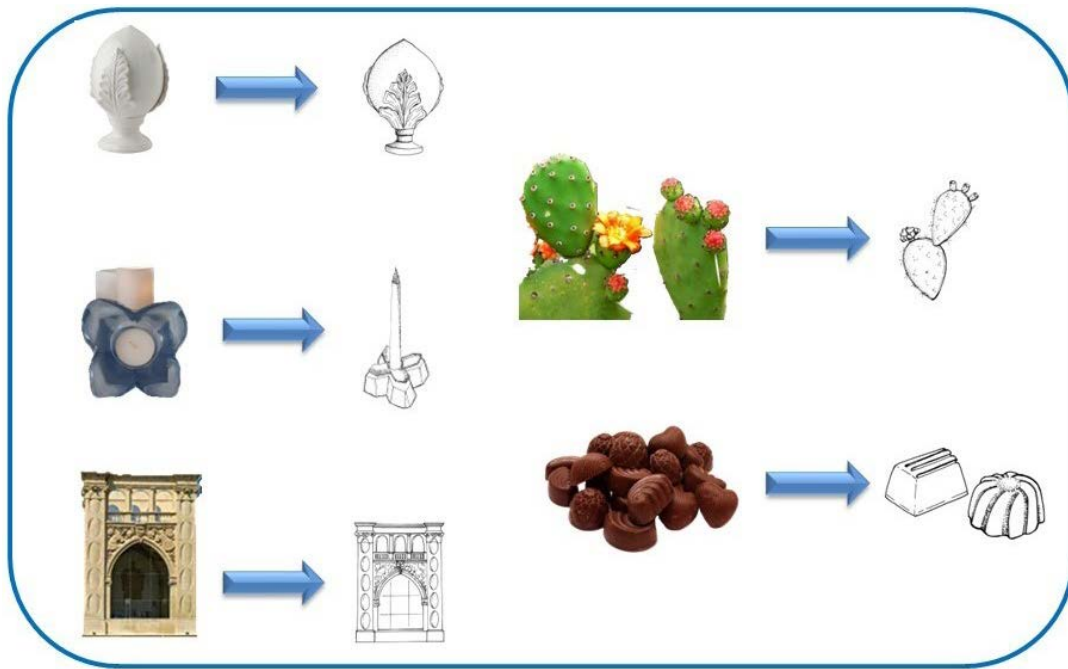


FIGURE 5: Real objects and manual and faithful reproduction.

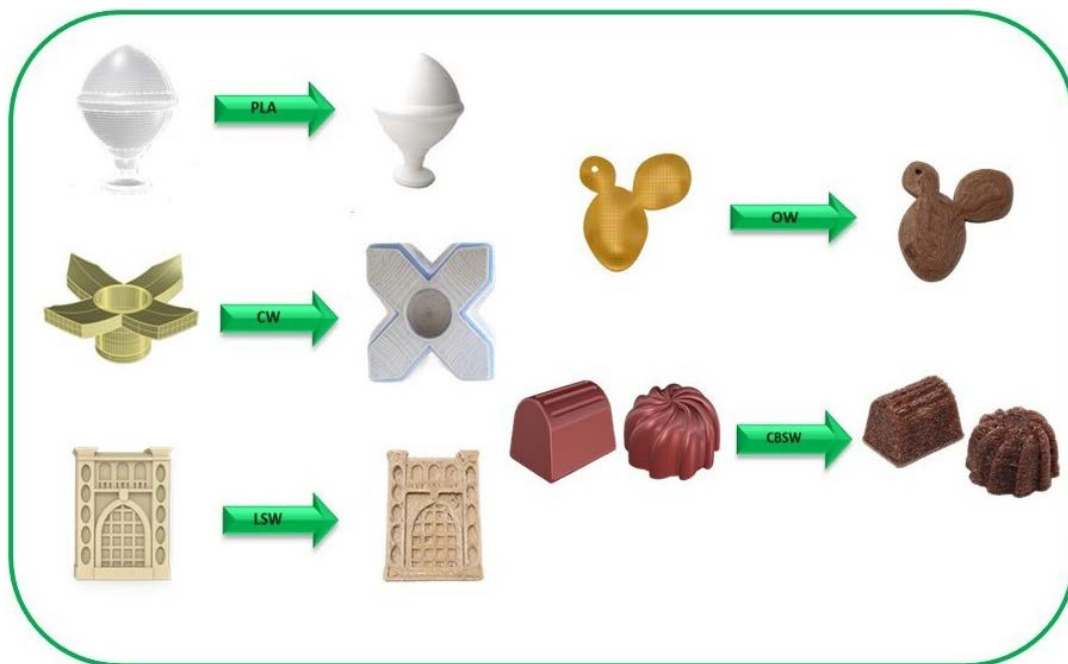


FIGURE 6: CAD models and related 3D printed objects.

my and sustainability. At the same time, with FFF printing, green objects with a unique design are produced in an easy and low-cost way.

4. CONCLUSIONS

In this paper, the authors demonstrated some examples of the application of Fused Filament Fabrication (FFF) printing to current and pressing issues of sustainability and circular economy. Specifically, the feasibility of pro-

ducing green biocomposite filaments for FFF from waste derived from artisanal and agro-industrial processes and using polylactic acid (PLA) as the matrix of the biocomposite, due to its biodegradability and biocompostability properties, was studied. For this purpose, local inorganic or organic wastes, such as cocoa bean shell (CBSW), olive wood (OW), leccese stone (LSW), and ceramic (CW) were oven-dried, pulverized, and used in a mixture with PLA, at a concentration of 10 wt. %. Biocomposite filaments with a diameter of 1.75 mm and suitable for the use in FFF were

obtained by extrusion and the properties of these and 3D printed bars were evaluated by microscopic-structural observations and bending tests. The results highlight the printability of all biocomposite filaments obtained from waste materials. Among them, CBSW and CW-based composites show similar or even better mechanical properties than neat PLA. The biocomposites OW and LSW-based show a lower adhesion at the interface between the filler and the polymer and the greater presence of particle aggregates in the filaments, which also leads to greater porosity of the 3D composites and a worsening of the flexural properties. Following the study of the printability of the developed filaments, real models were selected to be returned to the waste producing compagnie. These models were reproduced manually and transformed into a digital format suitable for 3D printing. The 3D printed objects produced from the developed biofilaments conserve in some cases the material properties of the original waste, such as the roughness of Lecce stone or the organoleptic characteristics of chocolate. Furthermore, with FFF printing it is also possible to create objects with complex shapes and quickly, which are difficult for artisans to create by hand.

Overall, the study demonstrates the potential of FFF printing technology to recycle waste by extending the life cycle of materials, with numerous environmental, economic and ecological benefits.

ACKNOWLEDGEMENTS

The authors thank Valentina De Carolis for the support provided in the 3D modeling design and printing.

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PHYSICAL AND CHEMICAL CHARACTERIZATION OF LABORATORY-GENERATED TYRE WEAR PARTICLES

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Article Info:

Received:
7 May 2024
Revised:
24 July 2024
Accepted:
28 July 2024
Available online:
7 September 2024

Keywords:

Microplastics
Tyre wear
Physical characterisation
Chemical characterisation
Microscopy
Gas chromatography-mass spec-
trometry

ABSTRACT

Tyre wear particles are generated by the frictional forces between a tyre and the road during driving. Tyre wear represents one of the biggest sources of synthetic polymer-based material released into the environment, significantly contributing to microplastic pollution and the associated ecological consequences. However, the extent of tyre particle pollution is not fully understood, and research and understanding are hindered by a lack of described chemical compounds that meet all the criteria for an effective marker of tyre particles. The aims of this study were to develop a methodology for generating tyre particles using a pin-on-disc tribometer; and to distinguish the chemical components in the particles – using two-dimensional gas chromatography-mass spectrometry – to propose a suite of potential markers. The results show that the morphology of the tyre particles reflected that of particles generated in previous literature and during driving. Additionally, the suggested markers were common across the three tyre brands studied here, thus meeting one of the criteria for a successful marker. Future research into measuring the concentration of tyre particles in environmental samples is necessary to understand further their distribution, and assessing the contribution of tyre particles to non-exhaust emissions can inform on future engineering strategies to minimise their release.

1. INTRODUCTION

Plastics are synthetic polymers that are not naturally occurring (Geyer, 2020). An early example of plastic was Parkesine, a thermoplastic nitrocellulose created by Alexander Parkes in the 1850s. However, Parkesine was not fully synthetic (Rasmussen, 2021). The first truly synthetic plastic was “Bakelite,” which was introduced over a century ago and was used in a variety of household items such as radios (Napper & Thompson, 2020). Plastics were not produced on an industrial scale until after the Second World War, reaching a global annual production of two million tonnes by 1950 (Geyer, 2020). Since then the plastic industry has increased exponentially, with the global output now exceeding 350 million tons per annum (Geyer, 2020), 50% of which is disposed of after a single use (Xanthos & Walker, 2020). The growth of the plastics industry was fuelled by their desirable properties, including affordability, durability, malleability, and low weight (Wang et al., 2021), resulting in their rapid incorporation into every aspect of life, including packaging, clothing, electronics, furniture, vehicles, buildings, sports equipment, medical supplies, and agriculture (Geyer, 2020). Supplementary Table S1 shows

the uses, properties, and greatest environmental sinks of common plastics.

Plastic debris is commonly classified based on particle size, as this eases comparison between different studies (Ramkumar et al., 2022). Microplastics are the size fraction that has received the most attention since evidence of their presence in oceans and marine sediments was first reported by Thompson et al. in 2004. The most agreed-upon definition for microplastics is “any synthetic solid particle or polymeric matrix of any shape and with size ranging from 1 µm to 5 mm, of either primary or secondary derivative/degraded material from the primary origin, which is insoluble in water”, which was proposed by Frias & Nash in 2019. Plastic particles that are greater than 5 mm are referred to as “macroplastics”, whereas those less than 1 µm are termed “nanoplastics” (Ramkumar et al., 2022). The term “primary microplastics” describes plastics that were manufactured to be between 1 µm and 5 mm in size for use in cosmetics, industrial cleaners, or as virgin plastic pellets, whereas secondary microplastics are formed from the physical abrasion and chemical breakdown of larger plastic fragments (Wang et al., 2021).

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Plastic's resistance to degradation means that it is present in every habitat on Earth (MacLeo et al., 2021), with soils and oceans being the first destination for 32% of discarded plastics (Machado et al., 2018). The ocean is currently considered to be the largest environmental sink for plastic waste (Wang et al., 2021), with land-originated plastics comprising 40-80% of marine litter (Barnes et al., 2009). Awareness of marine plastic waste began over 50 years ago when there were reports of plastic fragments being ingested by seabirds, and now plastic debris is distributed in oceans across the planet, along coastlines, in sea ice, at the surface and on the sea floor (Barnes et al., 2009). The latter is a major sink of ocean plastic, where it accumulates after breaking down into smaller particles, losing buoyancy and sinking (Abel et al., 2023). The remote nature of the deep-sea means that little is known about the extent of plastic pollution here, but a recent study discovered microplastics at depths of over 5000 m in the Kuril-Kamchatka Trench of the Pacific Ocean (Fischer et al., 2015). Plastic pollution differs from other pollutants in that it can induce three types of toxic effects: physical, chemical, and biological (Leistenschneider et al., 2023), which are detailed in Table 1.

Although the oceans are considered to be the world's biggest plastic pollution sink, terrestrial soils – particularly agricultural soils – could harbour more microplastics (Machado et al., 2018). It is estimated that one third of all plastic waste reaches soils, breaking down into microplastics and nanoplastics (Tudor et al., 2019). Wastewater treat-

ment plants (WWTPs) are one of the largest contributors to soil microplastics, as they release plastics from textile fibres, vehicle tyres, industrial processes and microplastic beads into the environment (Tudor et al., 2019). According to an estimate by Rochman et al. in 2015, 99% of microbeads in WWTPs settle into sludge, half of which is spread on agricultural land as fertiliser, adding 44,000-300,000 tons of microplastics to North American soils annually (Hurley & Nizzetto, 2018). The eight trillion microbeads remaining in the effluent are released into aquatic environments daily from WWTPs in the United States (US) (Rochman et al., 2015). Microbeads in cosmetic products have since been banned in many countries, including many European countries, the US, China, South Korea, Brazil, and New Zealand. In the UK, the Department for Environment, Food and Rural Affairs' (DEFRA) microbead ban covered "any water-insoluble solid plastic particle of less than or equal to 5 mm in dimension" and has been in place since 2018 (Anagnosti et al., 2021).

However, microbeads are not the only source of microplastics in sewage. The wear of vehicle tyres is one of the biggest sources of solid, water-insoluble, synthetic polymer-based material released into the environment (Knight et al., 2020; Sommer et al., 2018). Tyre wear particles are generated by the friction and shear forces that occur because of contact between a tyre and the road surface during driving (Baensch-Baltruschat et al., 2020). Many factors impact the properties of these particles, including the tyre material; vehicle characteristics (e.g. weight); driving style

TABLE 1: The physical, chemical, and biological toxic effects induced in marine organisms by exposure to microplastics.

Type of effects	Mechanism of interaction	Effects
Physical effects	- Macroplastic debris can trap and entangle marine mammals - Microplastics and nanoplastics can be ingested by marine organisms at all trophic levels via a variety of feeding techniques. - Plastic debris can sometimes be ingested due to being mistaken for a specific prey (e.g. green turtles mistaking plastic bags for jellyfish).	- Entrapment can cause death by starvation, blood-loss, or oxygen deprivation (Guzzetti et al., 2018). - Ingestion of microplastics results in digestive tract obstructions resulting in a pseudo-satiety sensation, dehydration, and poor nutrition (Guzzetti et al., 2018). - Ingestion also causes internal abrasions leading to physiological stress (Guzzetti et al., 2018). - Microplastics can translocate between body tissues (Leistenschneider et al., 2023). - Nanoplastics can pass up the food chain from low trophic levels (Leistenschneider et al., 2023).
Chemical effects	- Plastic additives (add useful properties to plastic) can leach out of plastic debris due to not being chemically bound. - Other environmental pollutants such as polychlorinated biphenyls (BCPs) and polycyclic aromatic hydrocarbons (PAHs) can adsorb onto the surface of microplastics causing additional toxic effects.	- Tanaka et al. found flame retardants in the tissues of <i>Puffinus tenuirostris</i> that had leached from ingested microplastics. Flame retardants are associated with a range of toxic effects, including reproductive toxicity, carcinogenicity, neurotoxicity and genotoxicity (Deng et al., 2018). - Causes endocrine disruption and cell cycle interruption, often leading to death (Hermabessiere et al., 2017).
Biological effects	- Surface of microplastic particles acts as a vector for pathogens, forming a biofilm. - Biofilms can decrease the buoyancy of plastic debris, increasing the availability of plastic toxicity throughout the water column.	- Pathogens can enter marine organisms upon ingestion of microplastic vectors (Leistenschneider et al., 2023). - Some species (e.g. copepods) have been observed being preferentially attracted to biofilm microplastics as opposed to pristine ones (Dahms et al., 2007). - Plastic debris may be accidentally ingested when a marine organism wants to ingest the biofilm (Dahms et al., 2007). - Reduction in fertility (Guzzetti et al., 2018)

(e.g. speed, acceleration, cornering); road material; and environmental conditions (Wagner et al., 2018).

Tyre particles in the environment are vastly different compared to the original tyre material. Firstly, shearing forces generate coarser particles which roll across the road surface in a “snowball” manner, collecting other road material to form a tyre and road wear particle (TRWP) complex. Moreover, heat from friction between the tyre and road surface changes the chemical composition of the particles compared to the original tyre material (Kreider et al., 2010). Harsher driving conditions, such as tyre slip, result in increased friction, generating a significantly higher proportion of ultrafine particles (≤ 100 nm) through evaporation and condensation of volatile organic compounds (VOCs) from the tyre tread (Liu et al., 2022).

It is estimated that almost six million tonnes of tyre wear particles are generated annually, around 12% of which end up in the atmosphere (Ding et al., 2023). Here, they can cause many effects upon inhalation, such as the release of inflammatory markers in human alveolar cells (Gualtieri et al., 2008). Another 6% of tyre wear particles end up in surface waters (Ding et al., 2023), the effects of which have been researched in depth (Kreider et al., 2010) despite this being a small sink of tyre particles. 15% of tyre wear particles end up in sewers, and the remaining 67% end up in soils, making soils the largest environmental sink of tyre-related microplastics. However, the consequences of tyre particles in soils have been researched far less compared to in aquatic environments (Ding et al., 2023). The ecological and health implications of tyre wear particles in various environmental compartments are shown in Table 2.

In the literature, tyre particles are described as being black in colour and elongated in shape, ranging from “sausage rolls” to rough spheres (Dannis, 1974), gathering road

material as they roll across the road (Adachi & Tainosho, 2004). In previous studies tyre wear particles have been generated in the laboratory to study their physical and chemical characteristics, but there is a lack of a standardised methodology. For example, Kreider et al. (2010) generated tyre wear particles using a road stimulator laboratory at the German Federal Highway Research Institute, which mimics a variety of driving conditions. Nevertheless, whilst this method produces realistic tyre wear particles, it uses specialist, expensive equipment that is inaccessible for most researchers. In the study by Son & Choi (2022), tyre wear particles were mixed with microplastics to replicate real-world TRWP. However, precise replication of TRWP is time consuming and is unlikely to be representative of the particles in question. Conversely, Gualtieri et al. (2005) generated tyre wear particles by rotating a new vehicle tyre against a steel brush, resulting in particles with a similar morphology to those released during real driving conditions. However, using a new tyre means that it has not been exposed to weathering and friction with the road surface, meaning the chemical characteristics of the particles are unlikely to be comparable to those in the environment (Wagner et al., 2022).

A reliable chemical marker of tyre wear particles should be specific to the rubber polymer; comparable across different tyre brands; water-insoluble so it cannot leach out; stable under environmental conditions; and easily and precisely detectable (Baensch-Baltrusch et al., 2020). Previous studies have assessed the accuracy of different markers such as benzothiazole (Liao et al., 2018) and zinc (Zn) (Chae et al., 2021). However, the concentration of benzothiazole varies significantly across different tyre brands (Chae et al., 2021), and it is also highly polar and hence water-soluble (Wagner et al., 2018). Additionally, Zn

TABLE 2: The ecological and health impacts of tyre particles and their leachates including zinc (Zn), other metals and organic compounds (e.g. benzothiazoles and resin acids).

Impacts of airborne tyre particles on human health	Impacts of leachate on aquatic organisms	Impacts of tyre particles and leachate on soil organisms	Impacts of tyre particles on sediments and energy cycle
- Inhalation can cause alveolar irritation and inflammation (Gottipolu et al., 2008). - Smaller metal particles may migrate directly into the systemic circulation, directing cardiac changes (Gottipolu et al., 2008). - Oxidative stress inhibits mitochondrial enzymes (Gottipolu et al., 2008). - Cell mortality and DNA damage (Wik & Dave, 2009). - Modification of cell morphology (Wik & Dave, 2009). - Can induce reactive oxygen species (ROS), causing an inflammatory response (Wik & Dave, 2009).	- Lethal in concentrations ranging from 50-100,000 mg/l to organisms such as algae, water fleas, zebrafish, and frogs (Wik & Dave, 2009). - Reduced growth or growth inhibition (Ding et al., 2023). - Reduced number of offspring (Ding et al., 2023). - Delayed development (Camponelli et al., 2009) - Deformity (Camponelli et al., 2009). - Teratogenic (interferes with normal fetal development) (Wik & Dave, 2009). - Mutagenic (interferes with an organism's DNA, causing gene mutations) (Wik & Dave, 2009). - Endocrine disruption. - Induction of cytochrome P450 (involved in detoxification) (Camponelli et al., 2009).	- Zn increases soil pH, increasing rate of nitrification, leading to nitrate leaching from soils (Gualtieri et al., 2005). - Leachates such as bisphenol A and di-n-butyl phthalate affect hydrogen and methane production of waste active sludge (Ding et al., 2023). - Heavy metals, antibiotics and pathogens can adsorb to tyre wear particles, selecting for antibiotic resistance genes amongst soil microbiota and transferring pathogens (Ding et al., 2023). - Ingestion of tyre wear particles can damage earthworms' guts, and toxic compounds can leach out in the gut decreasing their reproductive rate and survival (Ding et al., 2023). - Smaller tyre wear particles can be up taken by plant roots, having a toxic effect (Ding et al., 2023). - Some leachates, e.g. PAHs, can inhibit plant growth (Ding et al., 2023).	- Tyre wear particles slowly settle in sediments, following their entry into water bodies (Ni et al., 2024) - Burrowing activity of sediment dwelling organisms might be affected (Ni et al., 2024). - Microbial community involved in ecological cycling of nitrogen and carbon can get affected, disturbing the ecosystem (Ni et al., 2024).

is released from other traffic-related sources such as brake wear (Baensch-Baltrusch et al., 2020), and hence neither of these markers is useful when identified alone. Oleamide is added to tyre materials to increase abrasion resistance and improve the compounding process, and it is not commonly found in other components of road dust (Chae et al., 2021). Furthermore, a previous study successfully identified oleamide in organic extracts of tyre wear particles using gas chromatography-mass spectrometry (GC-MS) (Chae et al., 2021). Additionally, liquid chromatography-MS (LC-MS) has been used to determine the concentrations of known tyre additives such as 1,4-benzenediamine, N-(1,3-dimethylbutyl)-N'-phenyl- (commonly known as 6-PPD) and benzothiazoles (Rauert et al., 2022). However, GC is the more commonly employed in the analysis of VOCs due to its greater selectivity, resolution and sensitivity compared to LC-MS (Pandey et al., 2011). When combined with detection techniques such as MS, GC provides a powerful tool for the identification of separated compounds from complex mixtures, with lower limits of detection (LOD) than GC alone (Braithwaite & Smith, 1999; Pandey et al., 2011). However, some chemical mixtures contain too many target analytes which can co-elute as part of an unresolved complex mixture (UCM), complicating identification and quantification of individual components. Two-dimensional (2D) GC mitigates this issue by utilising two separation columns with different stationary phases connected in series, which separate the components by volatility and polarity, resulting in a higher chromatographic resolution and greater selectivity of individual components. 2D GC is a powerful technique for separating large numbers of organic constituents, and hence it has often been applied to the analysis of complex environmental samples (Worton et al., 2012).

The first aim of this study was to develop and characterise a method for the generation of realistic tyre particles so the study can be repeated by other researchers. The objectives taken to achieve this aim were: (1) generating realistic tyre wear particles from samples of Michelin, Bridgestone, and Radial tyres in a tribometry laboratory; and (2) giving a comprehensive overview of the steps taken throughout this project, using accessible materials and equipment. The second aim was to assess the physical and chemical characteristics of tyre wear particles to propose a suite of characteristics that can be used to identify them in the environment. This was achieved by: (1) physically characterising the particles using an optical microscope and a scanning electron microscope (SEM); (2) employing 2D GC-MS to separate and identify the chemical components present in the tyre wear particles; and (3) evaluating the chemical composition of the different tyre brands in order to select the compounds most likely to be effective markers for tyre wear particles in environmental samples.

2. METHODS

2.1 Generation of the tyre wear particles in the laboratory

Used tyres of distinct brands (Michelin, Bridgestone and Radial) were supplied by a local vehicle repair garage (Southampton, UK). These tyre brands were readily acces-

sible as used tyres from a local automotive workshop. Old tyres were selected because they have already been subjected to environmental factors such as wind, sunlight, and rain, making them environmentally relevant (Wagner et al., 2022).

Three samples were cut from each tyre using a band-saw (Axminster Tools, Devon, UK) (Supplementary Figure S1). The samples were cut 120° apart to assess intra-tyre variability and to provide repeated measurements for each tyre brand. In a specialist tribometry laboratory each tyre sample was glued to a separate aluminium pin using a strong adhesive, allowing each sample to be attached to the pin-on-disc (PoD) tribometer (RJH Finishing Systems Ltd, West Yorkshire, UK).

Tyre wear particles from each tyre sample were generated using the PoD tribometer which consisted of the sample glued to an aluminium pin and a rough-textured stainless-steel disc (Supplementary Figure S2). The roughness of the disc, as measured by an Alicona G4 Infinite-Focus (Alicona, Raaba/Graz, Austria), was 160 µm. The tribometer was run under a vertical force of 38 N. It was observed that increasing the vertical force beyond 38 N elevated the contact pressure between the pin and the disc, potentially leading to wear of the disc due to rough texture of the disc. Therefore, a 38 N force was suitable to have a balance between effective particle generation and minimizing damage to the disc. The pin of the tribometer was positioned close to the central point of rotating disc. Each sample was run on the PoD tribometer for 20 mins (30 rpm, clockwise run direction) or until sufficient mass of tyre particles for physical and chemical characterisation was generated (200-300 mg). The PoD tribometer was run at 30 rpm as this speed generated sufficient particles without generating ultra-fine stainless-steel particulates that could contaminate the samples. The particles from each tyre sample were collected into separate petri dishes (Supplementary Figure S3) that had been pre-weighed using an analytical balance (Mettler-Toledo Ltd, Leicester, UK). The coarse and ultra-coarse particle fractions were collected from the disc using stainless-steel tweezers and the finer fraction was removed from the disc and into the petri dish using a plastic toothbrush in a sweeping motion. A magnet was swept over each particle sample to remove any potential contaminating stainless-steel particulates. The disc was cleaned between being run for different tyre samples using the toothbrush, and any remaining particulates on the brush were removed by nitrile-gloved hands. Generating tyre particles using the PoD was loud and hence 3M Peltor Optime III ear defenders (RS Components Ltd, Northamptonshire, UK) were worn as an item of personal protective equipment (PPE) in accordance with the risk assessment.

2.2 Physical characterisation of the tyre wear particles

Once the particles had been generated using the PoD tribometer, they were stored in their petri dishes in a standard laboratory refrigerator (4°C) until they were physically characterised. The particles were spread evenly in the petri

dishes and their sizes and morphologies were observed by placing each petri dish under the lens of a Leica M205C optical microscope (Leica Microsystems Ltd, Milton Keynes, UK). The Leica camera was used to capture a range of photographs, and the lengths of different particles were measured to assess the size range of the particles in each sample.

A JEOL JCM-6000 SEM (JEOL Ltd, Welwyn Garden City, UK) was used to conduct further analysis of the morphology of the tyre particles, including their microstructure. Particles were selected at random from each sample using aluminium tweezers and were stuck onto aluminium stubs using carbon tape. The particles on the stub were then coated with a thin layer of gold using a JEOL JFC-1300 sputter coater (JEOL Ltd, Welwyn Garden City, UK). Gold was used to sputter coat the samples due to its incredibly high conductivity, meaning it increases the signal-to-noise ratio during SEM imaging, producing better quality images. Tyre wear particles themselves are not conductive and their surface would function as an electron trap, charging the particles and creating bright-white regions on their surface on the produced image. Gold has a high secondary electron yield, and hence it is often used when examining surface morphology of samples (Heu et al., 2019). Magnification ranging from x20 to x1500 was used to analyse and measure the particles, their microstructure, and any irregularities in the tyre material.

2.3 2D GC-MS separation and identification of VOCs from the tyre wear particles

To analyse the chemical composition of the tyre particles, a single particle from each tyre brand underwent thermal desorption at 240°C and 500°C to extract the VOCs. The VOCs released at each thermal desorption temperature were then concentrated and injected into a Pegasus BT 4D GC x GC TOF MS (LECO Corp., St. Joseph, Michigan, US) in turn. For each run, the GC column flow rate was 1 ml/min with a helium carrier gas, constant GC inlet and a transfer line temperature of 250°C. Split 200:1 1 µl injections were made onto an Rxi-5SilMS primary column (30 m length x 250 µm inlet diameter x 0.25 µm film thickness) coupled to an Rxi17SilMS secondary column (1 m length x 250 µm inlet diameter x 0.25 µm film thickness). The primary oven temperature was held at 40°C for 2 min, ramped 5°C/min to 300°C and held for 4 mins. The secondary oven temperature offset was +5°C relative to the primary oven. A modulation period of 8 secs with hot pulse duration of 2.4 secs and cooling time between stages of 1.6 secs was used for the data acquisition. The detector was on Maximum Sensitivity Mode. The modulator temperature was maintained +15°C relative to the secondary oven. The MS ion source temperature was 250°C and the MS data was collected at a spectral acquisition rate of 200 spectra/sec. Data acquisition with an m/z range of 35-550 started after a solvent delay of 250 secs.

2.4 Data handling and analysis

The data output from the 2D GC-MS was managed on Microsoft Excel. The total number of identified organic

compounds in each tyre particle sample was recorded before filtering the data so that only organic compounds with a chromatogram peak similarity to the MS database of ≥ 900 were included in the analysis. Conditional formatting was used to highlight any compounds that were found in all three tyre brands at each thermal desorption temperature, and these compounds were then sorted into a table based on their chemical categories (aliphatic; aromatic; polycyclic aromatic; hydrocarbons; and sulphur-, nitrogen- and silicon-containing) (Supplementary Tables S2 and S3). A literature search of each compound along with “tyres,” “rubber,” “industrial uses” and “sources in environmental samples” was performed using Google Scholar and Web of Science in order to identify the compounds with distinct links to tyre manufacturing or the rubber industry.

3. RESULTS

3.1 Comparison of the morphology of tyre wear particles from the different tyre brands

Observation of the tyre particles from the three tyre brands using an optical microscope revealed that they were all black in colour and ranged from sausage-shaped to spherical. Many of the Michelin tyre particles were larger than the Bridgestone and Radial particles, with some reaching over 11.0 mm in length (Figure 1A). However, most tyre particles from the Michelin tyre ranged from approximately 1.0 mm to 5.0 mm in length (Figure 1B), and the larger tyre particles tended to be more sausage-shaped than the smaller ones, which were more spherical (Figure 1B). The optical microscope analysis of the Michelin tyre particles also revealed the presence of a colourless fibrous material interwoven with the tyre rubber polymer (Figure 1B). This material was much less abundant in the Bridgestone particles (Figures 1C and 1D) and in the Radial particles (Figures 1E and 1F).

Initial observation of the tyre particles from the Bridgestone tyre revealed that, on average, they were smaller than the Michelin particles (Supplementary Figure S4), with many measuring under 2.5 mm in length (Figure 2D). Although the smaller Bridgestone particles were more spherical as opposed to sausage-shaped, the larger Bridgestone particles displayed the characteristic sausage shape, and ranged from approximately 1.5 mm to 6.0 mm (Figure 2C).

The Radial tyre particles were a similar shape and size to the Bridgestone particles, and most were between 3.0 mm and 6.0 mm (Figure 1E), with the largest reaching approximately 9.0 mm in length (Figure 4F). Similarly to the Michelin and Bridgestone particles, the Radial particles were mostly sausage-shaped, although the smaller particles (≤ 2.0 mm) tended to be more spherical (Figures 1E and 1F).

Analysis of the microstructure of the tyre wear particles from the different tyre brands using the SEM revealed that there were few stark differences between them. However, the Michelin tyre particles appeared less dense than the other two brands, and the microstructure seemed porous and stretchy (Figure 2A). The colourless fibrous material seen with the optical microscope was also visible in the

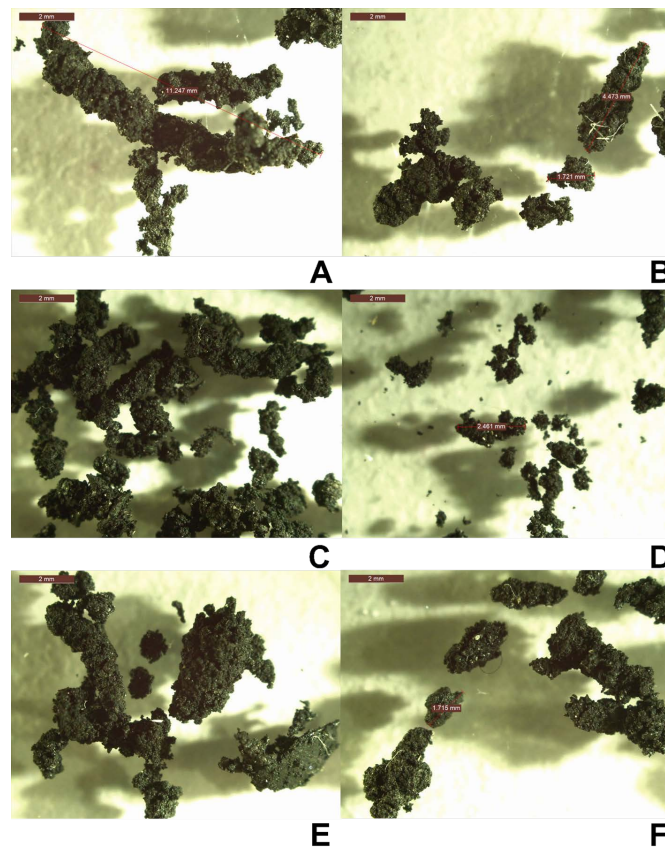


FIGURE 1: The optical microscope images of tyre wear particles from the three tyre brands (Michelin, Bridgestone and Radial).

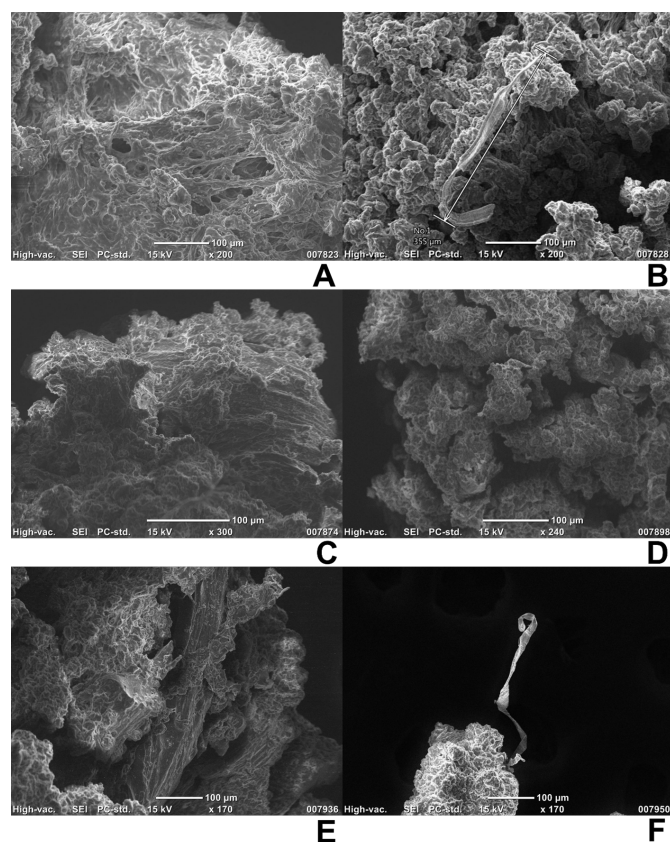


FIGURE 2: SEM images of tyre wear particles' microstructure.

SEM imaging, appearing as long, ribbon-shaped artefacts protruding from the rubber (Figure 2B).

In comparison, the Bridgestone tyre particles had a much denser microstructure with less visible porosity (Figure 2C) compared to the Michelin tyre particles. Furthermore, the surface of the particles appeared cracked (Figure 2D), indicating a potentially harder tyre composition.

Similarly to the Bridgestone tyre particles, the Radial tyre particles had a dense microstructure (Figure 2E). The ribbon-shaped artefacts were more sparsely distributed in the Radial particles compared to in the Michelin particles, but some were clearly visible extending from the rubber (Figure 2F).

3.2 Compounds detected in the different tyre brands after thermal desorption at 240°C

The chromatograms for the 2D GC-MS at a thermal desorption temperature of 240°C (Figure 3) showed that more compounds were released from the Michelin and Bridgestone tyre particles (Figures 3A and 3B) than from the Radial tyre particle (Figure 3C). Analysis of the 2D GC-

MS output prior to filtering the data revealed that, in total, there were 146 identified compounds released from the Michelin tyre particle at 240°C; 159 from the Bridgestone tyre particle and only 116 from the Radial tyre particle.

After filtering data, there were most similarities between the chemical composition of the tyre particles from the Michelin and Radial tyres, whereby there were 37 compounds common to the Michelin and Radial tyre particles. Furthermore, there were 33 compounds common to the Michelin and Bridgestone particles, and only 23 compounds present in both the Bridgestone and Radial particles.

Nineteen compounds were present in all three tyre brands after thermal desorption at 240°C (Supplementary Table S2), 21% of which were straight chain alkanes (ranging from C₁₈-C₂₅) and 32% were other hydrocarbons, mainly fatty acids which are commonly used in the synthetic rubber industry (Hirata et al., 2014). Compounds linked to the tyre industry were p-xylene, a thermal desorption product of natural rubber (Han et al., 2023); and cyclohexanethiol and benzothiazole (and its derivative 2-mercaptobenzothiazole) which are a processing aid (Molanorouzi & Mo-

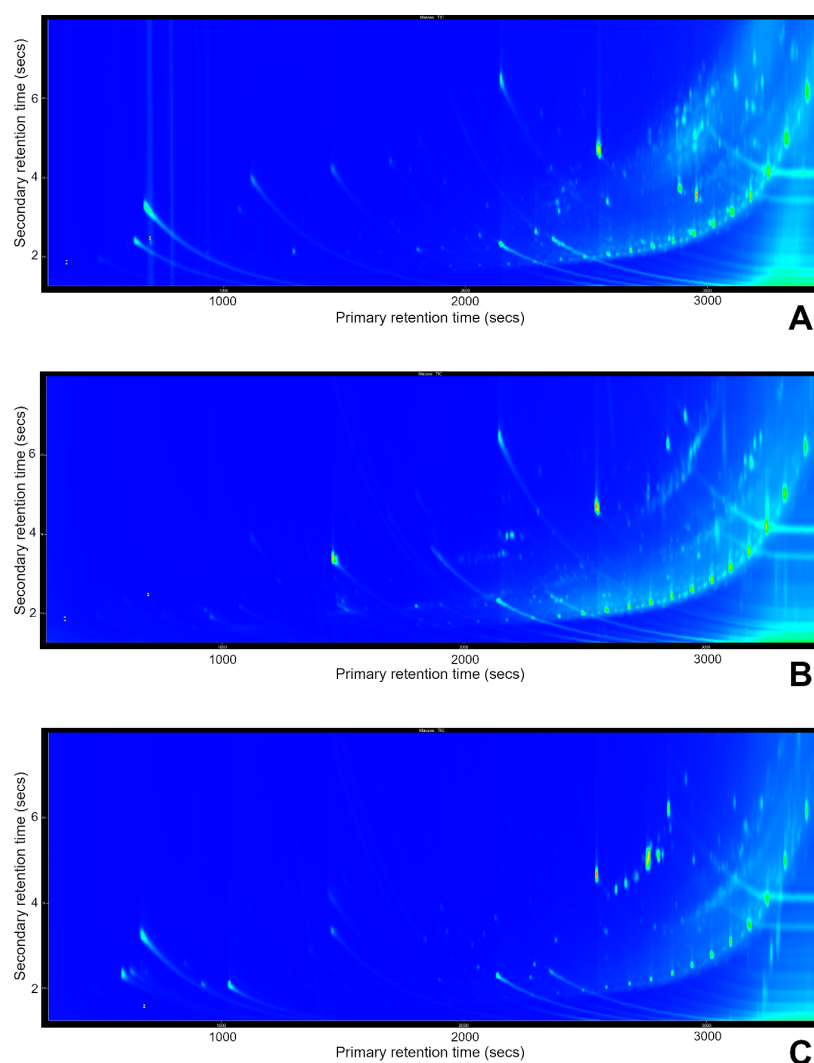


FIGURE 3: The chromatograms from the 2D GC-MS after thermal desorption at 240°C for the (A) Michelin; (B) Bridgestone and (C) Radial tyre particles.

haved, 2016) and a vulcanisation accelerator (Valdes et al., 2016) in synthetic rubber processing, respectively.

Styrene is used in the production of styrene-butadiene rubber (SBR) – the most common synthetic tyre rubber (Grieco et al., 2008) – yet at 240°C it was only present in the Michelin and Bridgestone tyre particles, but not in the Radial tyre particle until 500°C. α -methylstyrene is blended with styrene in synthetic rubber manufacturing to modify the viscoelastic properties of the tyre (Wolf et al., 2021). At 240°C it was present in the Radial tyre particle, but it was not present in the Michelin and Bridgestone tyre particles until 500°C.

6-PPD is an antioxidant and antiozonant widely used in rubber tyres to increase their resistance to weathering and thus extend their useful life (Chen et al., 2023). There was a peak on the chromatograms for the Michelin and Bridgestone tyre particles that was a close match to the MS database for 6-PPD. 6-PPD was also present in the Radial tyre after thermal desorption at 240°C, but with a peak similarity of 887. Nonetheless, it was included in these results because of its proven link to tyre particles in previous literature (Rauert et al., 2022).

Figure 4 is a hierarchy presenting the compounds with the strongest links to the tyre manufacturing industry. Although none of these compounds fulfil all the criteria for a suitable marker of tyre wear, they provide a strong indication of tyre wear particles in environmental samples when identified together. These compounds were common to all the tyre brands at 240°C (except styrene and α -methylstyrene), which is a key consideration when identifying a successful tyre wear marker (Baensch-Baltruschat et al., 2020).

3.3 Compounds detected in the different tyres after thermal desorption at 500°C

The chromatograms after thermal desorption at 500°C (Figure 5) showed that many more VOCs had vaporised

from the tyre particles compared to after thermal desorption at 240°C. As with thermal desorption at 240°C, more compounds were released from the Michelin and Bridgestone particles (Figures 5A and 5B) than from the Radial particle (Figure 5C). There were 710 identified compounds in the Michelin particle; 790 in the Bridgestone particle; and only 504 in the Radial particle. However, once the data was filtered so that only compounds with a similarity to the MS database of ≥ 900 were included, the number of compounds in each tyre brand were within a range of 10: 148 in the Michelin particle; 158 in the Bridgestone particle and 150 in the Radial particle. Of these compounds, ninety were present in both the Michelin and Bridgestone tyre particles; 89 in both the Bridgestone and Radial tyre particles; and 92 in both the Michelin and Radial tyre particles.

There were seventy compounds present in all three tyre brands after thermal desorption at 500°C, a 268% increase from the number of compounds present in all three brands at 240°C. The majority of these compounds were aromatic, with 31% being single ring aromatics and 29% being polycyclic aromatic hydrocarbons (PAHs) with two to four aromatic rings. The remaining compounds were aliphatic compounds (straight chain alkanes and alkenes ranging from C_{10} - C_{24}); hydrocarbons (mainly non-aromatic cyclic compounds); sulphur compounds; nitrogen compounds (including benzothiazole); and CMS compounds (Supplementary Table S3).

The only compounds identified after thermal desorption at 240°C that were still present at 500°C were benzothiazole, tetracosane, styrene/ α -methylstyrene and two CMSs. Benzothiazole is used as an antibacterial in hospitals and for the management of viral diseases in the agriculture sector (Zou et al., 2023). Similarly, styrene/ α -methylstyrene is used as a raw material in the formulation of road paints (Hoppe et al., 2005), which may lead to overrepresentation of tyre wear particles on roads. Tetracosane is an alkane, and its presence is not indicative of tyre particles. CMSs

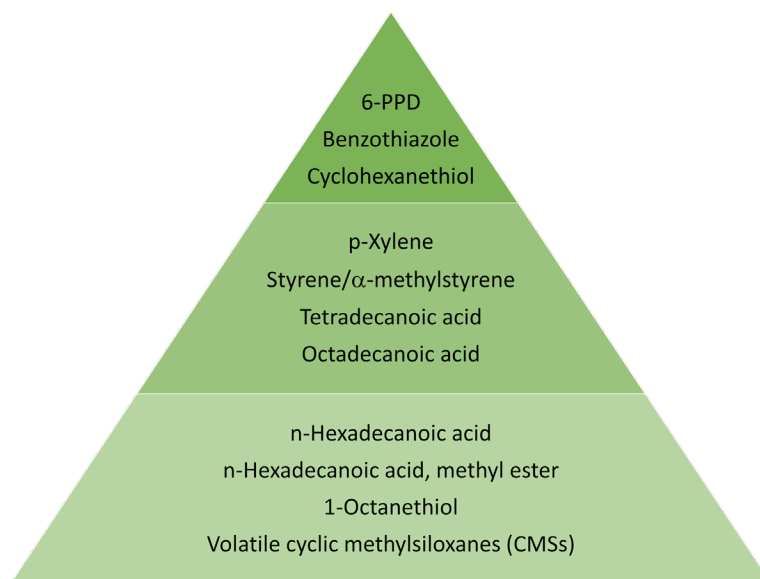


FIGURE 4: A hierarchy of the most useful compounds present in the tyre particles from the three tyre brands after thermal desorption at 240°C.

are widespread in the environment because they are used in personal care products and industrial processes related to silicon and rubber (Genualdi et al., 2011).

Other compounds present after thermal desorption at 500°C that have links to the rubber and tyre industry were aniline; cyclopentanone; and various PAHs, which were included in a hierarchy of potential markers (Figure 6). However, the increase in the proportion of PAHs to 29% at 500°C compared to just 5% at 240°C suggests that PAHs at 500°C are either by-products of degradation of larger compounds or are formed through reactions with other composites. For example, Richter & Howard (2000) found that PAHs react with single ring aromatic compounds, leading to successive growth of PAHs. Moreover, PAHs, as constituents of petrochemical fuels, are produced through combustion of fuel in engines (Dandajeh et al., 2019). Since PAHs are associated with burning of fuel and rank lower in hierarchy of potential markers (Figure 6), they might not be suitable indicators of tyre wear particles in the environment.

As with the hierarchy of potential markers at 240°C, these compounds were present in all three tyre brands

(except styrene/ α -methylstyrene), which is a key consideration when proposing a suitable marker compound (Baensch-Baltruschat et al., 2020). Compounds related to tyre manufacturing that were identified at 240°C, such as 6-PPD, cyclohexanethiol and p-xylene, were absent at 500°C.

4. DISCUSSION

Our study's first aim was to develop and characterise a method of generating tyre wear particles in the laboratory. Initial examination of the tyre particles' morphology using an optical microscope revealed that the particles from each tyre brand were black in colour and ranged from sausage-shaped to rough spheres, supporting observations made in previous literature (Adachi & Tainosho, 2004; Danis, 1974; Gualtieri et al., 2005; Padovan et al., 1999). Tyre particles generated here ranged from approximately 200.0 μm to 11.0 mm in length, and were significantly larger than the tyre particles generated in previous research at the University of Southampton. Preceding studies similarly used

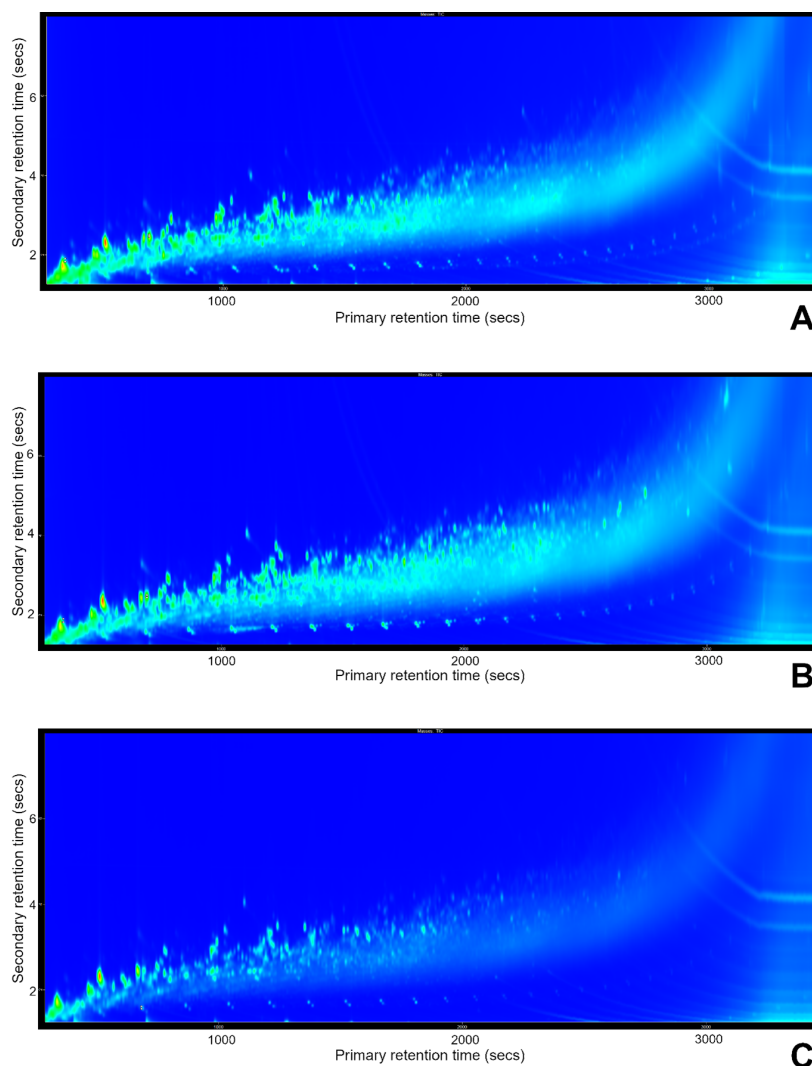


FIGURE 5: The chromatograms from the 2D GC-MS after thermal desorption at 500°C for the (A) Michelin; (B) Bridgestone and (C) Radial tyre particles.

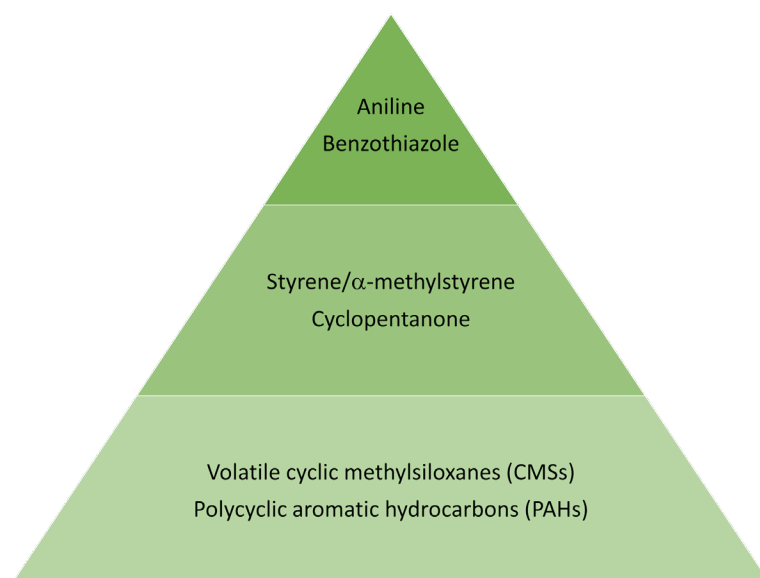


FIGURE 6: A hierarchy of the most useful compounds present in the tyre particles from the three tyre brands after thermal desorption at 500°C.

a PoD tribometer, but with a finer textured disc, producing tyre particles ranging from 120.0 μm to 3.0 mm in size. The texture of the disc used in this study was coarser, and consequently the tyre particles were larger in size. Subsequent analysis using an SEM revealed that the microstructure of the tyre particles was consistent in particles from the three brands. However, the Michelin particles were larger on average and their microstructure was less dense compared to the Bridgestone and Radial tyre particles. One potential explanation for the unique characteristics of the Michelin tyre particles is that the rubber polymer is softer and has a higher viscoelasticity, resulting in larger particles being formed during wear conditions (Cao et al., 2022). The higher viscoelasticity would also account for the porous appearance of the Michelin tyre particles' microstructure. Nonetheless, these differences were not pronounced enough to distinguish the Michelin tyre wear particles from other brands.

Our study's second aim was to propose a suite of chemical compounds that can be useful indicators for the presence of tyre wear particles in the environment. To address this aim, a 2D GC-MS was used to analyse the chemical composition of tyre particles from each tyre brand. No previous research was found evidencing the application of this technique to the chemical analysis of tyre particles, and hence its usage here is a novel approach to the study of tyre wear. Previous studies have often focused on a small number of potential marker compounds (Chae et al., 2021; Rauert et al., 2022), whereas this study provided a comprehensive overview of all the compounds in the tyre particles that have links to the tyre and rubber industries. Benzothiazole (Chae et al., 2021) and 6-PPD (Rauert et al., 2022) have been suggested as markers for tyre wear in previous literature and they were present here after thermal desorption at 240°C. There were nineteen compounds common to all three tyres after thermal desorption at 240°C, the majority of which were aliphatic compounds

and other hydrocarbons. The compounds deemed to be the most indicative of tyre particles in environmental samples were those that are used as additives or processing aids in tyre manufacturing, or those that are thermal desorption products of natural and synthetic rubber. Styrene was not released from the Radial tyres until 500°C, whereas it was released from the Michelin and Bridgestone tyres at 240°C. α -methylstyrene on the other hand, was released from the Radial tyres at 240°C, but not from the other two brands until 500°C. Oleamide, used as a marker of tyre wear in a previous study quantifying tyre particles in road dust (Chae et al., 2021), was notably absent from all the tyres after thermal desorption at both 240°C and 500°C. A possible explanation for the absence of oleamide is that it is not used in the manufacturing processes for Michelin, Bridgestone or Radial tyres. Alternatively, it may be present in concentrations below the lower limit of detection for the 2D GC-MS. Future research would benefit from repeating this study with a wider range of tyre brands and assessing whether oleamide is a suitable marker compound for tyre particles.

The proportion of aromatic compounds released from the tyre particles increased after thermal desorption at 500°C, whereby 60% of the compounds present in all three tyre brands were either single ring aromatics or PAHs. This observation is explained by the fact that more aromatic rings contain higher numbers of delocalised electrons, resulting in increased intermolecular Van der Waals interactions (Achten & Andersson, 2015). To overcome these forces a higher energy input is required, meaning the PAHs were vaporised at 500°C and not at 240°C (Alonso et al., 2014). This accounts for the increased number of compounds present in the three tyre brands after thermal desorption at 500°C compared to at 240°C.

However, although there were more compounds common to all three tyre brands at 500°C, fewer were suggested as potential markers because many were not unique to

tyres. Benzothiazole and styrene/ α -methylstyrene were the only marker compounds present at 240°C that remained at 500°C. Toluene and benzene (and its derivatives) are solvents used in tyre manufacturing (Nazarpour-Noshadi et al., 2022), but they have many other industrial sources (Yu et al., 2022) and their presence in environmental samples does not conclusively indicate the presence of tyre wear. Additionally, petrogenic PAHs are found in crude oil and products such as plastics, rubber, and asphalt, whilst pyrogenic PAHs arise from incomplete combustion of organic compounds such as crude oil and wood (Saha et al., 2012). Consequently, PAHs are not a reliable indicator of tyre wear when identified alone, but they can provide extra evidence for the presence of tyre wear particles if other indicative compounds are also present, and hence they were included in the marker hierarchy (Figure 6). However, there are large variations in the PAH content of different tyres depending on when they were manufactured. The dominant source of PAHs in tyres produced before 2010 were the highly aromatic extender oils used as processing aids during tyre manufacturing. In 2010, the European Union (EU) banned the use of extender oils with a regulated PAH content of over 10 mg/kg, and thus tyres manufactured after this date have a lower PAH content (Alves et al., 2020). Furthermore, the internal temperature reached by a typical passenger tyre during normal driving conditions is between 20 and 60°C (Genovese & Timpone, 2022), and the tread temperature rarely exceeds 160°C (Mavros, 2019). For these reasons, future research would ideally focus on the compounds released at 240°C, which more accurately represents tyre temperatures experienced during driving.

The optical microscope analysis revealed an unexpected colourless material protruding from some of the tyre particles, particularly those from the Michelin tyre. Closer analysis using an SEM showed that this material was ribbon-shaped with jagged edges (Supplementary Figure S4F). Daniela Taverner – an undergraduate researcher at the University of Southampton – ground chunks of tyre tread to generate particles instead of using the PoD tribometer, resulting in colourless flakes separating from the Michelin tyre tread. Observation of these flakes using an optical microscope and an SEM (Supplementary Figures S4B-S4E) revealed similarities with the material in the Michelin tyre particles generated here. One hypothesis was that the material was silica, commonly used as a tyre filler to improve the wet tyre grip and decrease rolling resistance. Silica tyre filler was developed by Michelin in the 1990s as an alternative to carbon black (Veiga et al., 2017), a probable reason it was observed in higher quantities in the Michelin tyre particles compared to the other two brands.

A strength of this study was the specifications for the tyres used for particle generation compared to previous literature. For example, Gualtieri et al. (2005) used laboratory-generated tyre particles to assess the toxicity of tyre leachates. However, they generated particles using a single, unused vehicle tyre, whereas this study used three brands of tyres – Michelin, Bridgestone and Radial – allowing comparison of the physical and chemical characteristics of the particles from each brand. This was vital in ensuring

that suggested markers were present in all the tyre brands, satisfying one of the criteria for a successful marker of tyre wear. Furthermore, there is evidence that the quantities of certain organic compounds in tyre tread change over the course of a tyre's life. For instance, the compounds 1-octanethiol, phenanthrene and anthracene have been found in higher proportions in new tyres compared to old tyres of the same brand and model (Wagner et al., 2022), reducing the reliability of the toxicity results from Gualtieri et al.'s study in 2005 which used only new tyres. The tyres used here were used/old and had been exposed to weathering and friction with the road during their use, meaning the chemical composition of the tyre particles was comparable to those present in the environment.

Although used tyres were selected here for particle generation, the exact history of each tyre was unknown, introducing an uncontrolled variable. Each tyre could have experienced significantly different driving conditions, leading to different alterations in their chemical compositions. Future work should attempt to minimise the variation in the tyres' history as much as possible, whilst acknowledging that this is a challenging aspect to control.

Several previous researchers have cryogenically milled tyre tread (Miller et al., 2022; Shin et al., 2023; Son & Choi, 2022) to generate large volumes of particles, for studying the chemical composition of tyres (Wagner et al., 2022). However, cryogenic milling generates larger tyre particles than those from road simulators and real-world driving, which are not the characteristic sausage-shape (Wagner et al., 2022). Contrastingly, the tyre particles were generated here by friction between samples of tyre tread and the rotating disc of a PoD tribometer, closely replicating the driving process and producing tyre particles with a similar morphology to those identified in environmental samples in previous literature (Klockner et al., 2021).

5. CONCLUSIONS

This study has established a reproducible methodology for generating tyre wear particles in the laboratory. Several factors should be considered for effective generation of wear particles. For instance, the disc of tribometer should be abrasive enough to generate particles. Future studies could evaluate particle generation with a disc of roughness greater than 160 μm . In this study, a vertical force of 38 N was most suitable for production of wear particles; horizontal forces could be reviewed in subsequent studies. Reducing the distance between the pin and the centre of rotating disc is essential to reduce the torque, as higher torque could reduce the functioning of tribometer.

The study has provided a comprehensive suite of potential marker compounds and physical characteristics that can be used to identify tyre wear particles in environmental samples. For example, 6PPD and benzothiazole – at the top of the hierarchy (Figure 4) and present in the tyres studied when analysed at 240°C – could be indicators for tyre wear particles in the environment, recognising that other potential sources of these compounds should be taken into consideration. Future research should focus on assessing the concentration of tyre wear particles in samples from

different environmental compartments to evaluate the relative significance of tyre particle pollution when compared to other, more researched pollutants, such as microplastics and airborne particulate matter (PM). The physical characteristics for tyre wear particles are important in this type of study. For example, if a study aims to evaluate the number of tyre wear particles in environmental samples, sausage-shaped assemblies of particles should be used, as this shape is unique to tyre wear particles. Since section 3.1 of this paper outlines that the generated tyre wear particles ranged from sausage-shaped to spherical, spiking experiments should be carried out to assess the under-representation of particles other than sausage-shaped. For the determination of tyre wear particle mass, 6PPD along with benzothiazole could be used.

More work is necessary to understand the factors that impact the emission of tyre wear particles, and how these factors may change. For instance, the average electric vehicle weighs ~20% more than a similarly sized internal combustion engine vehicle (Liang et al., 2021), with their growing usage increasing the emissions of tyre wear particles. It is important to assess the contribution of tyre wear particles to non-exhaust emissions, which can inform future engineering strategies to minimise the release of tyre particles into the environment.

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MSW-TO-RDF CONVERSION FOR CEMENT PLANT IN INDONESIA THROUGH PILOT PROJECT AND MODELING

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Article Info:

Received:
16 April 2024
Revised:
3 July 2024
Accepted:
26 July 2024
Available online:
26 August 2024

Keywords:

Municipal solid waste
Refused-derived fuel
Cement plant
MSW characterization
RDF production pilot project
Modelling
Fuel substitution
CO₂ reduction

ABSTRACT

This study investigates MSW management and its application in the cement industry through the conversion of MSW into RDF in four Indonesian regions: Jakarta, Bogor Regency, Depok, and Bekasi. Collectively, these areas produce 14,500 Mg/day of MSW. Samples totaling 1,265 Mg of this MSW were analyzed to determine their physical and chemical properties, particle size distribution, and fraction. Results indicated LHV of 5.4 MJ/kg and a high moisture content of 55%, underscoring the need for improved MSW treatment processes. The most common particle size was 50-80 mm, accounting for 31.6% of the MSW. The organic fraction was predominant at 43%, followed by plastics at 24% and paper at 13%. In a pilot project involving 10 trials with 1,000 Mg of MSW, processing through a series of pre-shredder, trommel, magnetic separator, wind-shifter, and fine shredder produced RDF with an LHV of 8.7 MJ/kg and 47% moisture, along with a lower-grade RDF with an LHV of 3.0 MJ/kg and 61% moisture. This effort resulted in 441 Mg of low-grade RDF and 423 Mg of higher-grade RDF, resulting in an estimated reduction of 217 Mg of CO₂ emissions. Further modeling suggested that using hot gases at the cement plant could produce drier RDF. To meet the cement plant's demands, thirteen RDF production units processing 6,500 Mg/day of MSW would be required. This would generate 3,100 Mg/day of RDF, achieving a 51% fuel substitution rate at the cement plant, reducing CO₂ emissions by 24%, and cutting MSW by 45% across the regions

1. INTRODUCTION

Cement is an essential building material known for its durability and versatility. It serves as a fundamental component widely used in the construction and development of infrastructure, playing a significant role in advancing human life. According to Cembureau (2023), the global cement production capacity in 2022 stood at 4.3 billion Mg and is expected to continue increasing. However, this upward trend in cement production is likely to intensify the challenges faced by the cement industry in reducing CO₂ emissions. The global cement industry currently faces at least three global issues that require continuous solutions. The first challenge is reducing global CO₂ emissions from the cement industry. The second challenge involves production efficiency, encompassing cost efficiency, natural resource efficiency, and fuel usage. The third challenge pertains to the implementation of economic circularity. Previous research on CO₂ emissions reduction in the cement industry has led to savings in the purchasing of materials required during cement production (Rootzén & Johnsson, 2017). Therefore, the cement industry bears a significant responsibility for climate protection, particular-

ly in addressing CO₂ emissions reduction and preserving natural resources (Salamanova et al., 2020; Schneider, 2019).

Similar to global challenges, the cement industry in Indonesia also faces similar issues, including production efficiency, CO₂ emissions reduction, and concerns about sustainable business practices. According to the Ministry of Industry of the Republic of Indonesia (Kemenperin) (2022), cement production in Indonesia contributes to 28% of the country's CO₂ emissions. One of the leading cement companies in Indonesia, Indocement, is actively seeking to address these environmental challenges with the goal of becoming a more sustainable cement producer. In its 2022 Sustainability Report, Indocement stated that sustainable practices, such as greenhouse emission reduction, waste management through circular economy, and water and energy efficiency, were carried out (Indocement, 2022). Indocement also puts attention on the use of alternative fuel (AF) sources with a target to achieve a 50% coal substitution rate by 2030. This is estimated to equate to around 6,000 Mg per day of alternative fuel.

MSW management in Indonesia is a pressing issue, as the country generates nearly 69 million Mg of MSW

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annually. This poses challenges for landfills and raises health concerns, especially in the densely populated cities of Jakarta, Bogor, Depok, and Bekasi, known as Jabodebek, which contribute approximately 14,500 Mg of MSW daily (BPS Jakarta, 2023; KLHK, 2023). Despite having regulations in place for waste reduction and comprehensive waste management, the execution of these measures remains inefficient (Fatimah et al., 2020). To address this problem, the conversion of MSW to refuse-derived fuel (RDF) emerges as a potential solution that aligns with the government's waste policies. RDF, derived from MSW through specific processes, offers a cross-sectoral remedy for environmental performance and waste management issues. It presents a cost-effective and ecologically friendly fuel replacement, supporting the cement industry's transition towards sustainable practices (Mateus et al., 2023).

RDF shares similar characteristics with conventional fuel as it predominantly consists of the combustible fractions of MSW (Paszowski et al., 2020). Ideally, RDF composition has a high content of paper, plastic, wood, and textiles, contributing to higher heating value. It has a high calorific value (around 16-18 MJ/kg), low production costs, and a homogeneous particle size. Despite its benefit as an AF and its positive environmental impact, RDF has several disadvantages related to heterogeneity, moisture, and sulphur, chlorine, and ash contents (Brás, 2017; Paszowski et al., 2020). To circumvent these disadvantages, optimizing RDF production from MSW through pilot project and modeling is required. In European countries, a legal framework has been established to promote waste valorization as a way to reduce the amount of waste by RDF production from the rejected streams of multi-municipal waste management systems (Brás, 2017). For instance, in Europe and globally, cement manufacturers currently use RDF as a common AF to replace coal, aiming to optimize fuel costs (Paszowski et al., 2020).

The utilization of RDF presents a convergence of solutions between cement production challenges, including production efficiency, CO₂ emission reduction, and the implementation of sustainable business practices, with MSW management issues. This signifies a promising cross-sectoral solution that has the potential to address two distinct yet interconnected challenges. Processing MSW into RDF for AF can also be a collaborative solution between cement producers and the government, particularly at the city level. Thus, this research aimed to examine RDF's feasibility as an AF in Indocement, located in the Jabodebek region cement industry, proposing a model that harmonizes resource optimization with industrial sustainability.

2. MATERIALS AND METHOD

2.1 Study area description and MSW collection

The study was conducted on four regions located near Indocement, a cement plant in Bogor, Indonesia. These regions were Jakarta, Bogor Regency, Depok, and Bekasi (Jabodebek). MSW was collected from local intermediate MSW storage facilities located in these four regions, and these storage facilities were located within 50 km of the cement plant. The MSW sample collection for each region is summarized in Table 1.

The MSW samples were collected from December 2022 to June 2023, which also comprised the rainy and summer seasons. The samples were transported to the cement plant using trucks with a capacity of 6-8 m³, with an average carrying capacity of 2-3 Mg per truck. The MSW analysis and RDF production and analysis were conducted at the cement plant.

2.2 Experimental design

The research consisted of three phases (Figure 1). First, the collected MSW was analyzed. Next, the MSW was processed into RDF on a pilot project scale, followed by RDF production modeling.

TABLE 1: MSW Sample Collection.

Item	Unit	Urban Area				Total
		Jakarta City	Bogor Regency	Depok City	Bekasi City	
Sample collection duration	days	8	8	8	8	32
Sampling in summer						
Sampling schedule	N/A	8-15 May 2023	22-29 May 2023	5-12 Jun 2023	19-26 Jun 2023	N/A
Trucks sample count	unit	56	56	56	56	224
Rainfall	mm/month	50-100				N/A
Sampling in rainy						
Sampling schedule	N/A	12-19 Dec 2022	9-16 Jan 2023	28 Nov - 5 Dec 2022	23-30 Jan 2023	N/A
Trucks sample count	unit	56	56	56	56	224
Rainfall	mm/month	250-400				N/A
Total trucks sample count	unit	112	112	112	112	448
Samples taken per truck	Mg	2.84	2.79	2.78	2.88	2.82
Total samples taken	Mg	318	313	312	323	1,265
Daily waste generation	Mg/day	8527	2688	1400	1830	14,445

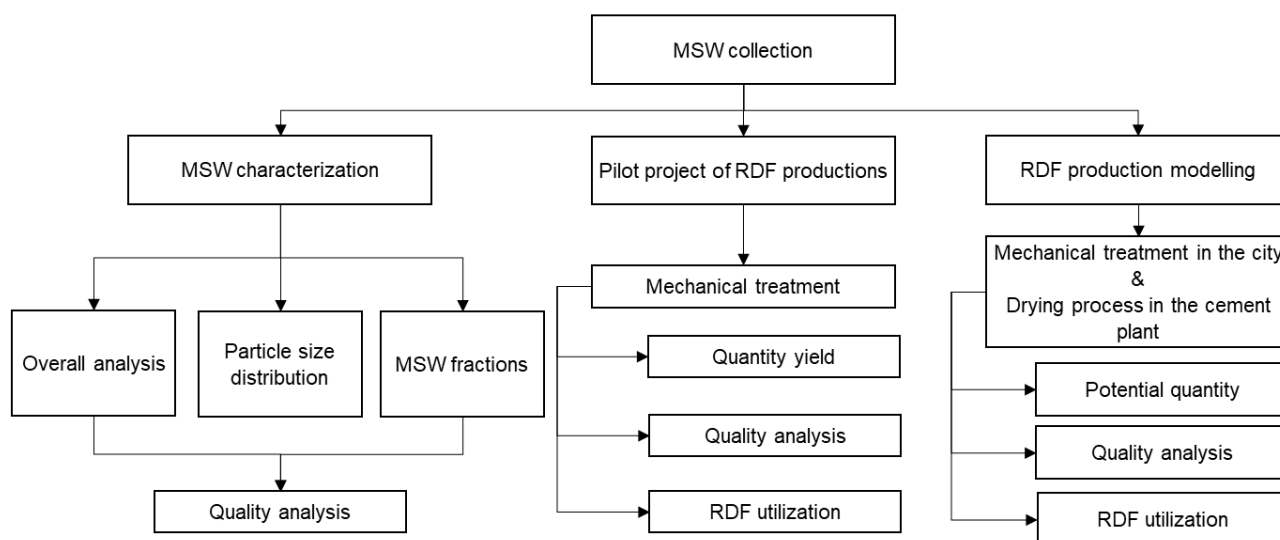


FIGURE 1: Schematic flow of the study.

2.2.1 MSW characterization

The MSW collected from different areas was properly mixed, as advised by Wirosoedarmo et al. (2021), to ensure sample homogeneity and consistent results (Figure 2). Then, the samples were subjected to the following analysis:

- Chemical and physical properties.

Moisture content (MC), calorific value, and ash content were determined according to EN 15414-3: 2011, EN 15400: 2011, and EN 15403: 2011, respectively. Total carbon (C), hydrogen (H), and nitrogen (N) analysis was done following EN 15407: 2011, while oxygen content was deduced stoichiometrically by subtracting carbon, hydrogen, nitrogen, and ash percentages from 100%. Total contents of sulfur (S) and chlorine (Cl) were assessed using XRF Epsilon 5 analyzer.

- Particle size distribution.

To devise effective strategies for waste processing and management, a particle size distribution analysis was conducted (Zhang et al., 2019). The particle size range employed in this study was >200 mm, 100-200 mm, 80-100 mm, 50-80 mm, 10-50 mm, and <10 mm. Combustible, non-combustible, biomass, and non-biomass fractions were determined. Combustible fraction was the sum of the organics, paper, wood, textile, plastic, and rubber components while the sum of the other components was non-combustible fraction (Dharmendra, 2022). The sum of organics, paper, and wood components was considered as biomass fraction and the sum of the rest of the components was considered as non-biomass fraction (Perea-Moreno et al., 2019).

- MSW fraction.

The MSW fraction analysis was carried out to identify the proportion of combustible and non-combustible

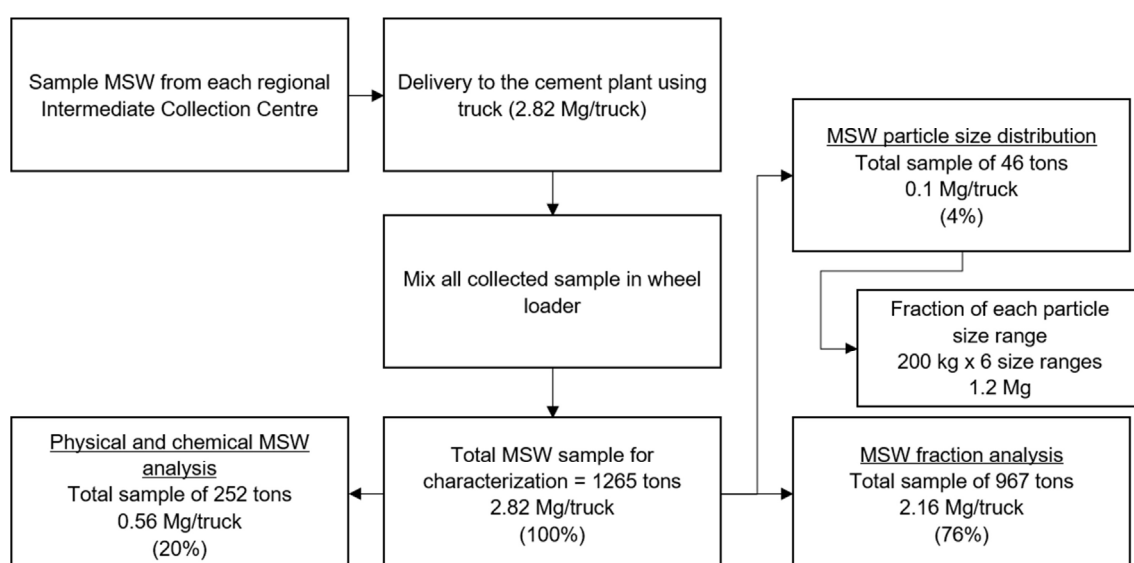


FIGURE 2: MSW sample collection, processing, and distribution for characterization.

went processing through a fine shredder (<50 mm), and the other was low-grade RDF with a size of <80 mm (Figure 3).

2.2.3 RDF production modeling

The pilot project served as the basis for RDF production modeling. Modeling occurred in two locations: the pre-drying process in the city and the drying process at the cement plant using a box dryer with hot gas (Figure 4). This modeling arrangement reduced land usage, a significant factor in urban settings, as the cement plant features a spacious location with 10 kilns. Two units of box dryer were designated to dry the fine and low-grade RDF separately and op-

In the second phase, a pilot project was conducted to process MSW into RDF. The pilot project consisted of 10 trial runs, initiated in December 2023, with around 100 Mg of MSW utilized and transported to the cement plant's pilot facility for each trial. The MSW went through the pre-shredder to produce materials sized between 200-300 mm. This equipment effectively reduces MSW to a smaller, more consistent size, improving later treatment processes (Oladejo et al., 2020). Next, the MSW was processed using a trommel (80 mm mesh) to ensure the RDF is within the optimal size range of 50-80 mm, capturing combustible fractions and accommodating the lower calorific value of finer materials (Malafák et al., 2018). A magnetic separator installed above the conveyor belt removed ferrous metals from waste, and then a wind-shifter was utilized to divide waste into heavier and lighter fractions. Lastly, a fine shredder was used to produce a consistent and homogeneous particle size of 50x50 mm, ensuring the output reliably meets standard fuel quality specifications (Nanda & Berruti, 2021). Two RDFs were produced in this study: one under-



TABLE 2: The Chemical and Physical Properties of MSW Samples.

Parameters	Unit	Summer				Rain				Mean±Std dev.	Rep. Value
		Jakarta	Bogor	Depok	Bekasi	Jakarta	Bogor	Depok	Bekasi		
HHV	MJ/kg	8.8	8.6	8.3	8.3	8.5	8.2	8.2	8.3	8.4±0.1	4-10 ¹
LHV		5.9	5.4	5.1	5.3	5.9	5.4	5.1	5.3	5.4±0.1	5-10 ²
MC	% wt. (ar)	52	54	54	56	55	56	55	57	55±3	20-67 ¹
Ash	% wt. (db)	11	11	11	12	11	11	12	11	11±1	1-30 ³
S		0.22	0.23	0.23	0.23	0.23	0.23	0.24	0.23	0.2±0.0	0.1-0.7 ⁴
Cl		0.40	0.38	0.39	0.42	0.41	0.38	0.39	0.43	0.4±0.0	0.24-1.9 ^{5,6}
C		49	52	54	52	50	56	53	49	52±4	11-54 ^{1,7}
H		7	8	6	7	8	9	7	8	7±2	6-10 ¹
N		1	1	1	1	1	1	1	2	1±0	0.6-1.0 ¹
O		32	27	28	28	29	23	27	30	28 ±1	30-55 ¹

Rep. Value: reported value; ¹ Amen et al., 2021; ² Drudi et al., 2019; ³ Siddique, 2010; ⁴ Cheng et al., 2023; ⁵ Ma et al., 2010; ⁶ Shao et al., 2010; ⁷ Tursunov et al., 2015

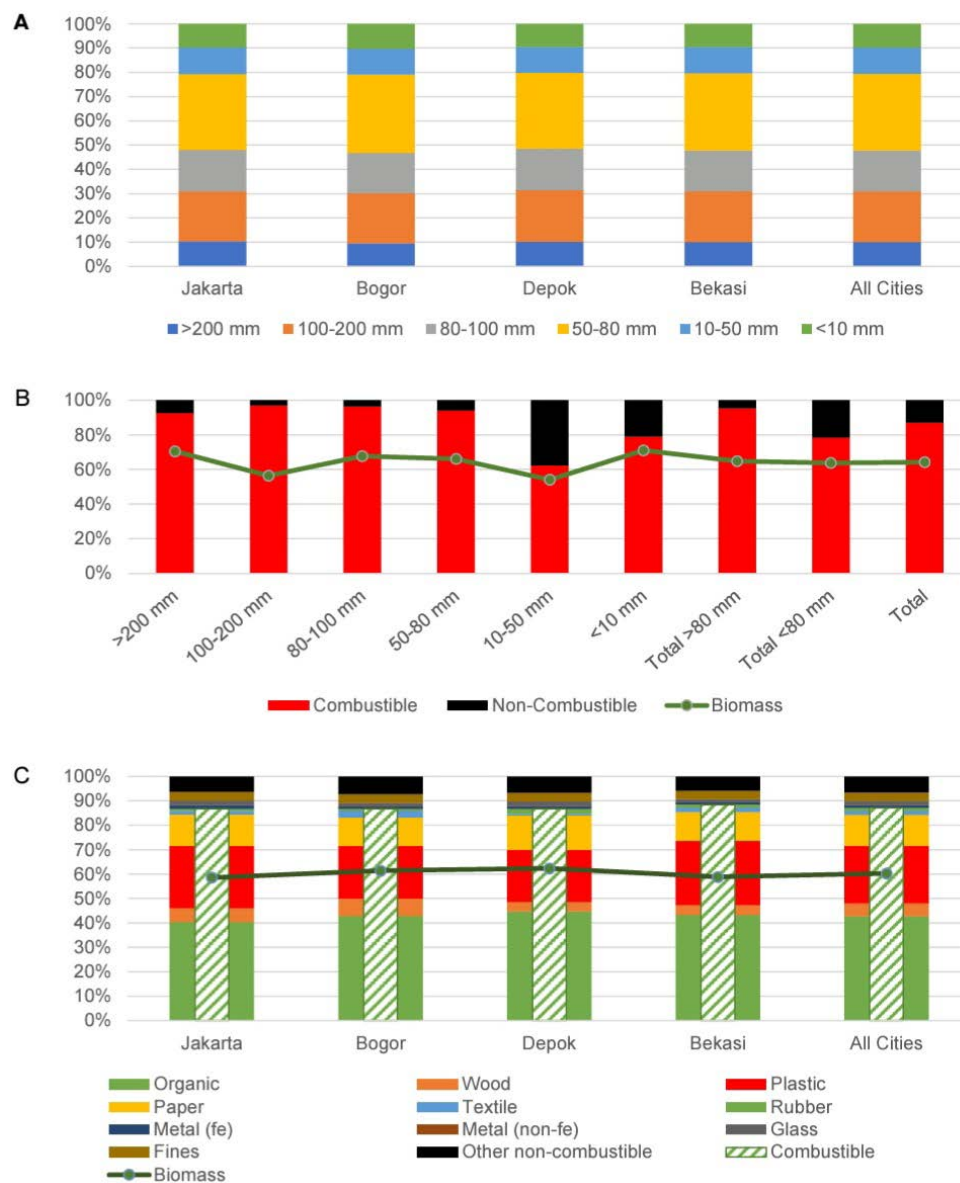


FIGURE 5: Particle size distribution and MSW fraction analysis results. (A) Particle size distribution in mm; (B) Combustible, non-combustible and biomass fractions of each particle size range; (C) MSW fraction analysis.

non-combustible rate of 38%, followed by particle sized 10-50 mm with a combustible rate of 21% (Figure 5B). The higher non-combustible rate may be attributed to inert content. Overall, 87% of MSW was combustible (Figure 5B). This finding is aligned with a study in Jordan that reported high combustibility across all sizes (Al-Hajaya et al., 2021). Our combustible fraction was similar to those reported in Indonesia and India, with values of 88% and 87%, respectively (KLHK, 2023; Singhal et al., 2022). This similarity suggests comparable waste habits and less source sorting in Indonesia and India (World Bank, 2020). Germany had a lower combustible fraction of 81%, indicating better waste management and segregation, further implying simplified processing of MSW into RDF (BMUV, 2023).

The biomass content within particle size distributions was also determined by estimating the waste material types. Organic waste, including wood and paper, was classified as biomass fraction (Perea-Moreno et al., 2019). A larger proportion of the MSW particle sizes was biomass, with an average value of 64% (Figure 5B). The biomass fraction of MSW, industrial waste, and sewage sludge in incineration facilities were shown to be 57%, 41%, and 78%, indicating biomass fraction variability between sources (Kang et al., 2017). Biomass is considered a carbon-neutral source of energy because the CO₂ released during its combustion is roughly equivalent to the CO₂ absorbed by the plants during photosynthesis, making it a net zero carbon option on the path to carbon reduction (Zagaria et al., 2023). Therefore, in the calculation of CO₂ emissions, the biomass content can reduce the emission factor when it is combusted.

3.1.3 The MSW fraction analysis result

The collected MSW fraction samples constituted a total mass of 967 Mg (Figure 2). This substantial volume, due to the scale of sample collection, is crucial to ensure representativeness of the study. This extensive data set enables an in-depth understanding of the MSW stream's variety, essential for creating targeted waste management plans and models that inform strategic decisions. Organic materials constituted the largest fraction, averaging 43%, followed by plastics and paper, which accounted for 24% and 13% of the total fraction, respectively (Figure 5C). The plastic data is consistent with other data in Jakarta, which was reported as 23% (BPS Jakarta, 2023). Wood, textile, rubber, metals (Fe and non-Fe), glass, and fines were found to be in lesser quantities (Figure 5C). Other non-combustible materials, such as stones and ceramics, had an average value of 6%. Valuable items were detected in minimal amounts because the samples were taken from intermediate MSW storage facilities where items with commercial value, such as 3D plastic, glass, and metal, were often collected by waste pickers for resale (Mastufatul et al., 2023). In general, the data indicated diversity in the solid waste profile, consistent with previous studies (Brás et al., 2017; Świechowski et al., 2020).

The biomass fraction of each area was assessed. Similar to the biomass content of particle size distributions result, biomass content had a larger percentage in all areas, making up a total average of 60% (Figure 5C).

A table detailing the chemical and physical properties of each MSW fraction is presented in Table 3, where only the fines fraction was analyzed for non-combustible content. Organics exhibited an HHV of 4.5 MJ/kg with a high moisture level at 78%, possibly lowering its energy potential. Plastic had a high HHV at 17.8 MJ/kg and LHV of 13.8 MJ/kg with a moisture level of 48%, indicating a promising energy source. Paper and textiles also had notable calorific values with reasonable moisture content. The content of plastics and paper emerges as a crucial factor for producing high-calorific-value RDF, suggesting that higher fractions of these materials will lead to a correspondingly higher caloric value of the RDF (Tihin et al., 2023b). Rubber stood out with the highest calorific values (HHV of 28.5 MJ/kg and LHV of 25.7 MJ/kg) and low MC, enhancing its fuel quality. Overall, the HHV and LHV values of the fractions were found to be lower than in previous studies (Komilis et al., 2012; Zhou et al., 2014; Boumanchar et al., 2016; Zhen et al., 2019). The lower calorific values are likely due to high MC of the major fractions, which have been shown to decrease HHV and LLV (Komilis et al., 2012). In terms of carbon content, plastics have the highest percentage (64%), contributing to their combustion quality. The combined average of all MSW fractions had a moisture level of 55% and a carbon content of 47%, influencing MSW's viability as a fuel. Carbon is a crucial factor in assessing energy potential and CO₂ emissions, which affects environmental and carbon footprint evaluations (Wienchol et al., 2020). The majority of elemental analysis results was similar to the results of investigations in Morocco, China, and Greece (Komilis et al., 2012; Zhou et al., 2014; Boumanchar et al., 2016). Meanwhile, there is no earlier report on the the properties of fine fraction of MSW (Table 3).

To corroborate the findings with the overall MSW analysis, the weighted average of each parameter from each fraction was calculated by weighing it with % fraction wt, as presented in Figure 5C. The weighted average for each parameter was similar to the overall MSW analysis result, except for carbon content. The carbon content in the analysis was 52% (Table 2), while based on MSW fraction, the carbon content was 47% (Table 3 section C). This difference occurs due to differences in the total amount of sample used and the sample collection approach.

3.2 Pilot project of RFD production

The completed pilot project solely utilized mechanical treatment methods and did not incorporate drying processes. Each batch processed 100 Mg of MSW, and the pilot project was evaluated over 10 trials. The trial outcomes showed that the trommel screen's material processing performance dropped by approximately 50% from its initial capacity, processing only 3-7 Mg due to the 55% MC causing substantial spillage and separation challenges. The fine shredder's efficacy also fell to 1-4 Mg/hour, about 20-30% of its designed throughput, as moisture caused material clumping and machinery blockages. These challenges highlight the significance of moisture control for maintaining optimal mechanical treatment operations (Ismail et al., 2019).

TABLE 3: The chemical and physical properties of MSW fractions.

No.	MSW Fractions	HHV	LHV	Moisture	Ash	S	Cl	C	H	N	O
		MJ/kg		% wt. (ar)							% wt. (db)
A	Combustible										
1	Organic	4.5±0.4	1.2±0.4	78±7	17±2	0.4±0.04	0.7±0.07	50±6.5	6±1	1.8±0.2	19±2
2	Wood	7.5±0.8	4.9±0.8	47±5	5±1	0.1±0.09	0.1±0.09	47±3.9	7±1	0.3±1	40±1
3	Plastic	17.8±1.6	13.8±1.6	48±5	6±1	0.1±0.01	0.3±0.02	64±6.6	13±1	0.1±0.0	15±1
4	Paper	11.2±1.2	8.3±1.2	51±5	3±1	0.2±0.02	0.1±0.09	43±3.8	7±1	0.4±0.0	44±1
5	Textile	11.6±1.1	8.7±1.1	53±6	13±1	0.1±0.01	0.1±0.01	49±5.7	7±1	3.2±0.4	22±1
6	Rubber	28.5±2.9	25.7±2.9	4±1	3±1	0.4 ±0.04	0.1±0.01	53±5.7	12±1	0.9±0.1	29±1
B	Non-combustible										
1	Fines	1.2±0.1	0±0	16±1.7	60 ± 5	0.4±0.04	0.1±0.01	26±3	3±0.3	0.2±0.0	11±1.4
C	Weighted average by % fraction	8.5±0.8	5.5±0.8	55±5	11±1	0.2±0.03	0.4±0.05	47±5	7±1	0.9±0.1	20±1

The pilot project produced two types of RDF, i.e., RDF and low-grade RDF (Figure 1). The analysis results of the RDF are shown in Figure 6. RDF had the highest HHV and LHV compared to MSW and low-grade RDF, with values of

12.1 and 8.7 MJ/kg, respectively (Figure 6A). Meanwhile, LHV values of the MSW, low grade, and RDF were lower than the respective HHV values, possibly due to water and hydrogen correction of MC. RDF also had a better characteristic since it had the lowest MC of 47% (Figure 6B). In contrast, low-grade RDF had the highest MC of 61%. This high moisture level happened because the refinement process did not include a sufficient drying phase, leaving the low-grade RDF with excessive dampness. Following a comparison with previous studies (Hemidat et al., 2019; World Bank, 2020; Sarquah et al., 2023), both RDFs produced had lower HHV and LHV values and higher MC, indicating a lower RDF quality.

Overall, the elemental measurements of low-grade RDF and RDF were all within the previously reported values (Gallardo et al., 2014; Alfè et al., 2022; Sarquah et al., 2023). However, an increase in ash content was observed during the conversion, hinting at an accumulation of non-combustible materials in the RDFs. Additionally, a modest rise in sulfur and chlorine was recorded post-conversion, especially in the low-grade RDF. The MSW-to-RDF conversion increased the carbon content, suggesting greater energy potential in RDF, attributable to the increased presence of plastic and paper in the RDF. The hydrogen content of RDF was higher than the low-grade RDF, while the oxygen content of low-grade RDF was higher than RDF (Figure 6C).

The HHV values of both products were still in the range of the cement plant's specifications, although both products had higher MC values than the specification (<45%). Both RDF products did not meet Indonesian standards while according to European standards ISO 21640:2021, they fell under class 5 (Kemenperin 2017; CEN, 2021). Since the low-grade RDF and RDF met the cement plant's specifications in terms of HHV, sulfur, and chlorine content, they were utilized in the cement plant, resulting in the output presented in Figure 7. The total amount of low-grade RDF and RDF utilized were 441 Mg and 423 Mg, respectively. RDF produced higher LHV and total energy generated than low-grade RDF, indicating RDF as a better source of fuel. The CO₂ emissions factor values in this project were 50 Kg CO₂/GJ and 65 Kg CO₂/GJ for RDF and low-grade

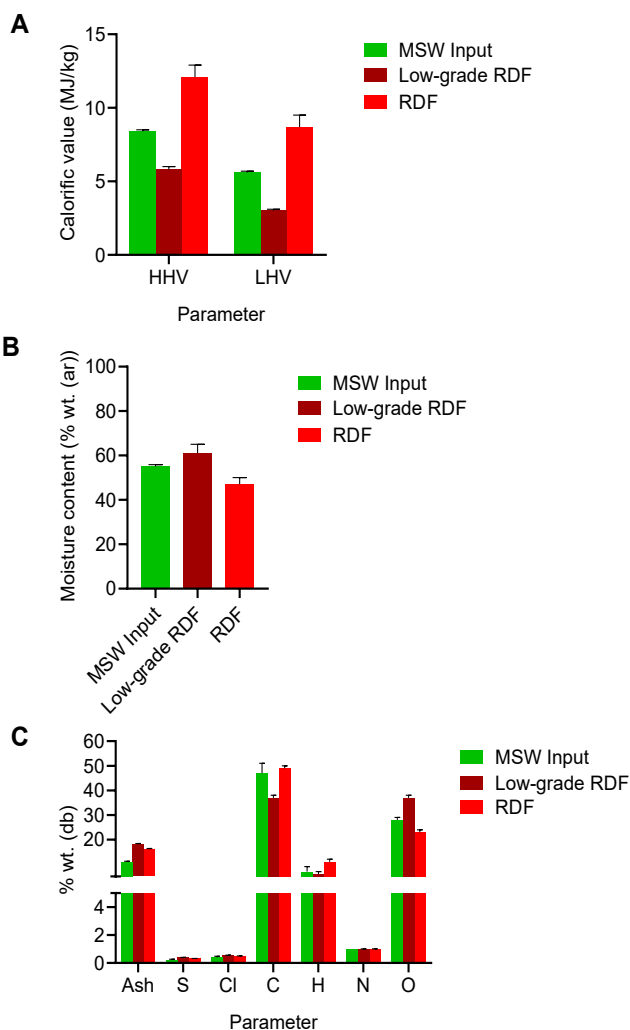


FIGURE 6: Pilot project output quality comparison with MSW input. (A) Comparison of calorific value; (B) Comparison of moisture content; (C) Comparison of ash and elemental.

RDF, respectively. In another study, the reported emissions factor was estimated to be around 27 Kg CO₂/GJ for RDF (MVW Lechtenberg & Partner, 2008). As low-grade RDF had a higher emission factor, the CO₂ emission reduction was lower than the RDF. In total, the RDF used led to an estimated total of 217 Mg CO₂ savings.

3.3 RDF production modeling

The pilot project indicates the need to incorporate a drying process in the production of higher quality RDF. Hence, two steps of drying processes were used in this modeling. The first involved a pre-drying process in the city, and the second involved a drying process at the cement plant using a box dryer with hot gas. This modelling arrangement reduced land usage, a significant factor in urban settings, as the cement plant features a spacious location with 10 kilns.

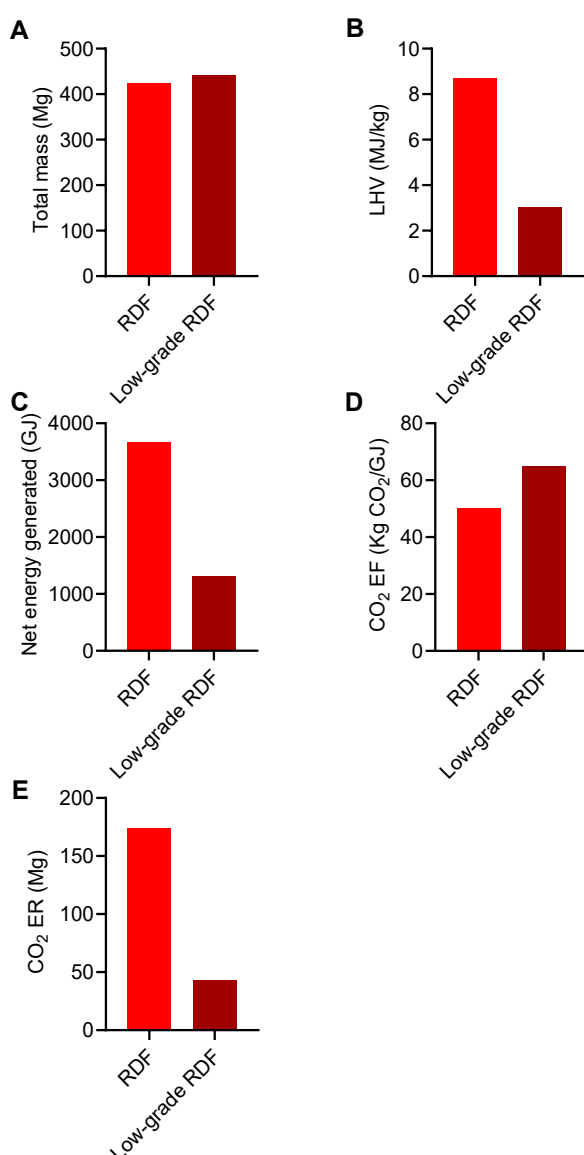


FIGURE 7: Utilization of pilot project output. (A) Total mass produced; (B) LHV value; (C) Net energy generated; (D) CO₂ emission factor (EF); CO₂ emission reduction (ER).

The design and operational characteristics of the drying system are crucial for preparing the fuel for use in the calciner. Tests for the box dryer project have been carried out at the Heidelberg cement plant in China (Indocement, 2024), showcasing important performance metrics. The box dryer has a capacity of 270 Mg each. There were two units designated for processing material sizes greater than 80 mm and another two units for sizes less than 80 mm. The dryers were operated at 130-175°C, allowing the drying process to be completed in one day. The type of the box drier was concrete with a grab crane with an air-blowing capacity of 24 m³/min. The hot gas exits from the bag filter, and the drier has a moisture reduction input of 55-60% wt. (ar) and output of 30-35% wt. (ar). This output is inline with previous studies that reported a thermal moisture reduction of 10-60% (Tun & Juchelková, 2019; Zamrudy et al., 2019). The drying system is pivotal for boosting RDF quality by increasing its calorific value and reducing moisture content. Its proximity to the cement kiln allows efficient use of the plant's thermal energy, streamlining operations and potentially cutting heat generation costs (Conceição & Rolim, 2019).

Figure 8 shows that the modeling results in better RDF output than that of produced in the pilot project. The yield of the total RDF product received was 48%, which aligns with a review by Gadaleta et al. (2022) focusing on the mass flow analysis of mechanical biological treatment (MBT) facilities, indicating that approximately 15-50% of the processed material becomes RDF. The RDF produced met the cement plant's RDF specifications, with fine RDF catering to medium-quality specifications (class 3 of ISO 21640:2021) and low-grade RDF as low-quality specifications (class 5 of ISO 21640:2021).

The RDF modeling and pilot project mainly composed of plastic and paper, reaching up to approximately 60% (Figure 9). In contrast, the organic materials in the MSW and low-grade RDF modeling made up the majority of the bulk. The differences emphasize the changes in material composition during the RDF process, with a decrease in organics and an increase in plastics and paper, thereby enhancing energy recovery potential (Tihin et al., 2023). Unfortunately, the analysis of the low-grade RDF fraction in the pilot project could not be conducted due to challenges in identifying smaller-sized stabilized materials. In low-grade RDF modeling, the non-combustible fraction was 30%, consisting of fine materials. For cement plants, this is not a significant issue as the ash content contains raw cement materials like silica, which can be utilized effectively in the calciner (Wang, 2021).

Lastly, a feasibility study to examine the potential of RDF to replace 50% of the cement plant's fuel, based on an annual heat requirement of around 12,650 TJ out of 24,800 TJ. The modeling processed 500 Mg/day of MSW, serves as the reference unit for an RDF production plant, and CO₂ emission reductions were calculated against coal, with its emission factor of 97.6 kg CO₂/GJ (Juhrich, 2022) and an LHV of 20.8 MJ/kg (Indocement, 2024). Two scenarios were set to represent a condition where coal consumption is 100% (scenario 1) and where 50% of fuel requirement is fulfilled by RDF modeling (scenario 2) (Figure 10).

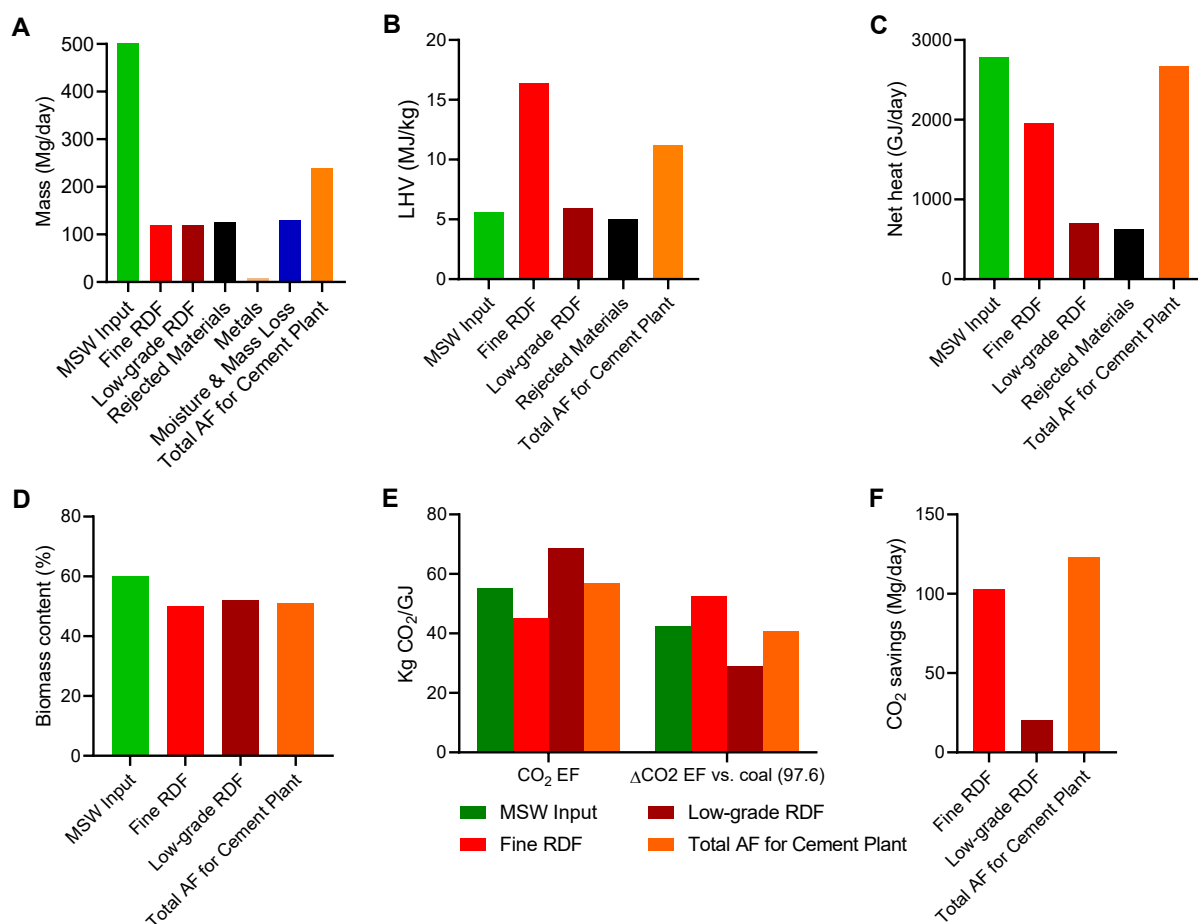


FIGURE 8: RDF Modeling analysis and output. (A) Total mass; (B) LHV; (C) Net heat; (D) Biomass content; (E) CO₂ emission factor (EF) and CO₂ RF difference vs. coal (97.6); (F) CO₂ saving.

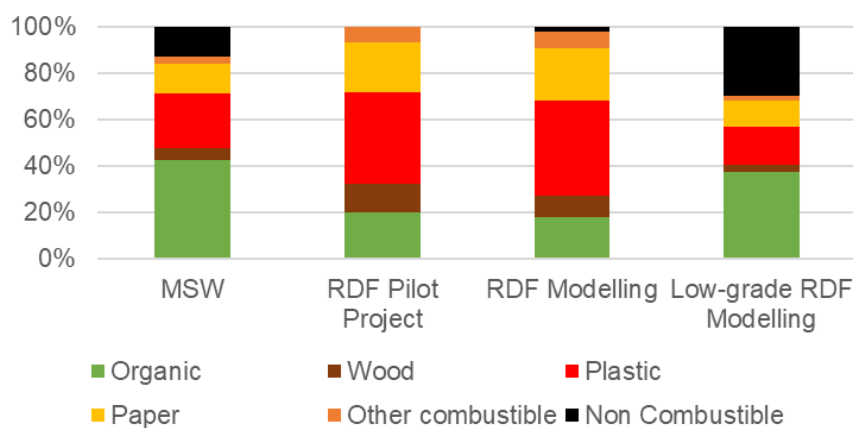


FIGURE 9: Comparison of MSW and RDF fraction analysis.

To meet the 24,800 TJ energy requirement, scenario 1 would require approximately 1.2 million Mg/year of coal. To achieve a minimum 50% AF substitution rate in scenario 2, 13 RDF plant units would be needed to produce a total RDF of 3,100 Mg/day. This setting would result in a 51% AF substitution rate, reducing coal consumption by 610,000 Mg/year and decreasing CO₂ emissions by 24% (585,000 Mg/year; Figure 10). Furthermore, implementing RDF at the cement plant could significantly reduce MSW

output from the four cities by 500 Mg/day for each of the 13 available units, resulting in a daily MSW reduction of 6,500 Mg. This reduction could account for up to 45% of the daily MSW from the Jabodebek area, which produces approximately 14,500 Mg/day. The results demonstrate that utilizing RDF in the cement plant as an AF presents economic and environmental benefits by reducing the fossil fuel use, decreasing CO₂, and utilizing MSW from the nearby areas.

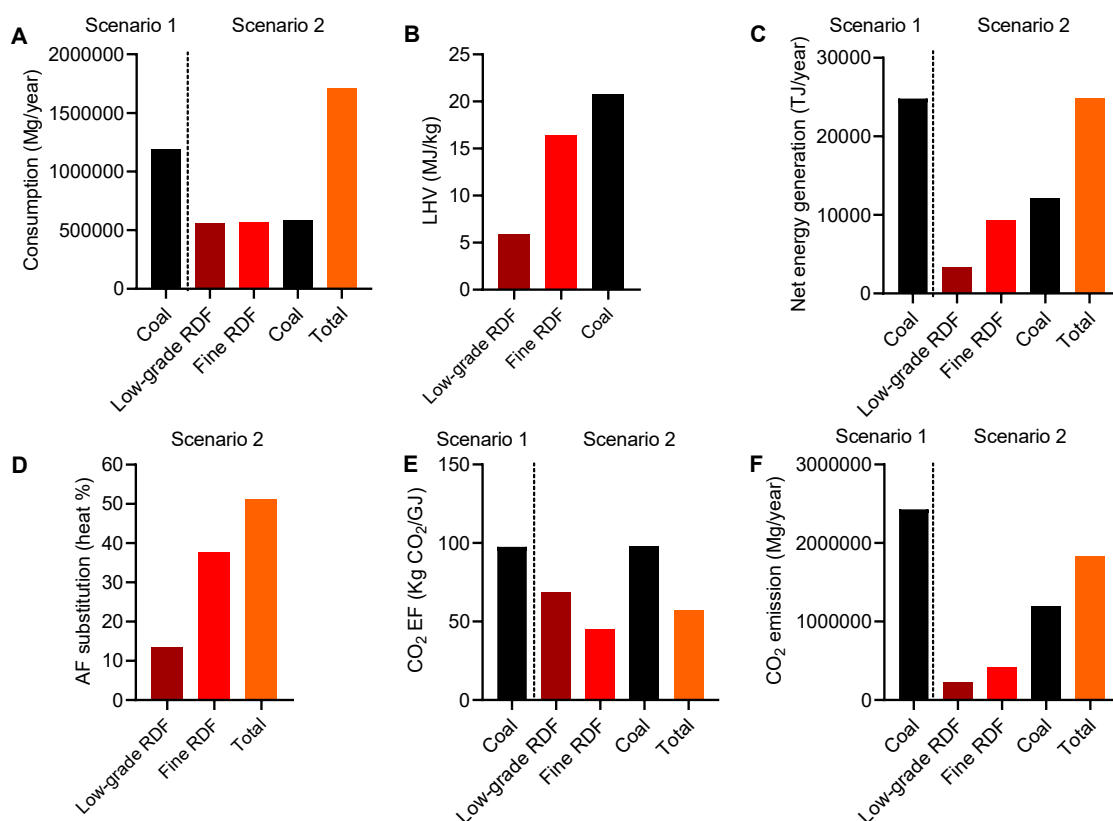


FIGURE 10: Energy and CO₂ emission of 100 % coal consumption (scenario 1) and 50% alternative fuel (AF) substitution using RDF (scenario 1). (A) Fuel consumption; (B) LHV of fuel; (C) Net energy generated by the fuel; (D) AF substitution of scenario 2; (E) CO₂ emission factor (EF); (F) CO₂ emission.

4. CONCLUSIONS

Low LHV and high MC of the MSW collected from Jabodebek underscore the need to further process MSW before RDF conversion, where the combustible material composition corresponds to RDF quality. The high MC also presented challenges in the mechanical treatment operations during the pilot project. Without the drying process in the pilot project, 1,000 Mg of MSW resulted in RDF with an LHV of 8.7 MJ/kg and MC of 47%, and low-grade RDF with an LHV of 3.0 MJ/kg and MC of 61%. Albeit the RDF output comply with the cement plant's specification, the output did not meet quality standards in Indonesia and other countries, indicating the need for additional intervention. Consequently, a drying process was implemented in the modeling stage.

The RDF plant production modeling involved pre-drying in the city and drying at the cement plant, utilizing excess hot gas from a plant producing 10,000 Tg of cement annually from 10 kilns. The drying process resulted in better RDF quality output than RDF produced in the pilot project. Processing 500 Mg/day of MSW yielded 119 Mg of fine RDF with 25% MC and an LHV of 16.4 MJ/kg, along with an equivalent amount of low-grade RDF with 42% MC and an LHV of 5.9 MJ/kg. CO₂ emission factors of RDF were lower than coal, which could contribute to reducing CO₂ emission. After a feasibility was conducted, achieving a minimum 50% substitution of AF at the cement plant would require 13 RDF plant units derived from 6,500 Mg/

day of MSW. This setup would yield a 51% substitution rate, cut coal consumption by 610,000 Mg/year, and lower CO₂ emissions by 24%, totaling a reduction of 585,000 Mg/year. The adoption of RDF could significantly cut the daily MSW volume from the four regions by 6,500 Mg, potentially reducing the Jabodebek area's daily waste generation by up to 45%. Overall, the results highlight the potential of MSW-to-RDF conversion as a cross-sectoral solution for environmental performance and waste management issues.

ACKNOWLEDGEMENTS

This study was conducted with the assistance of the local governments of Jakarta, Bogor Regency, Depok, and Bekasi, which provided coordination for MSW sampling. This study was supported by PT Indocement Tunggul Prakarsa Tbk, which provided the laboratory and equipment for the pilot project.

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AGRI-FISH: SUSTAINABLE AND INNOVATIVE FEEDS FROM AGRICULTURAL WASTES FOR A RESILIENT AND HIGH-QUALITY AQUACULTURE

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Article Info:

Received:

18 April 2024

Revised:

18 June 2024

Accepted:

1 July 2024

Available online:

18 August 2024

Keywords:

Sustainable aquaculture

Plant-based feed

Circular economy

One Health

ABSTRACT

The Agri-fiSh project has been funded under Horizon 2020 - PRIMA, with the research partnership of three Mediterranean countries: Algeria, Italy and Spain. The project will contribute to the transition to a more resilient agro-livelihood system through an innovative and circular farming approach, which promotes an efficient use of biological and natural resources with a valuable increase in productivity. New potential for farms, agri-food systems and local markets will arise from the valorization of agricultural wastes to be used for the creation of high-value plant-based feeds for a sustainable inland aquaculture. Nutraceutical potential of specific agricultural wastes (i.e. grape seeds, legumes and cereals), in combination with other components, as Lactic Acid Bacteria (LABs) fermented whey and medicinal herbs) to create plant-based feeds for aquaculture, will foster animal and human health with consequent benefits for the environment, local economies and consumers.

1. INTRODUCTION

1.1 Project's aim

Agri-fiSh project aims at increasing the environmental and socio-economic performances of small-scale farming systems through the development of an innovative model, which will help in improving organizational aspects of the agri-systems, while creating a new value chain governance model, especially at the local levels. For the first time agriculture and aquaculture are linked together to achieve a common goal: the application of a circular economy model, defined as “from the field to the net”, for the creation of sustainable and innovative feeds from agricultural wastes for a resilient and sustainable aquaculture. The overall objective of the project is to reinforce the resilience of local agri-food systems face to climate change, through capacity building and cooperation among the actors involved in the agriculture and aquaculture sectors, while adopting a circular model, which will effectively contribute to increase ecosystem services, biodiversity diversification and farmer's incomes. The scope is also to provide communities with evidence-based and promote a sustainable inland aquaculture in rural lands as an effective and cost-saving activity, thus promoting farming diversification in regions characterized by degraded soils, like Northern Africa and

Southern Europe. The project is built on interconnected research activities, to support the sector's transition to sustainable agriculture and aquaculture and to the circular economy model proposed, which could be further on up-scaled for aquaculture at sea. The circular model is applied to conceive innovative feeds, with high-nutraceutical values and low environmental impact (from cradle-to-grave), for aquaculture productions starting from the reuse of agricultural wastes and with the combination of natural antimicrobial substances deriving from sustainable agriculture, such organic and climate-smart agriculture (CSA), LABs and some medicinal plant species, such as *Opuntia ficus-indica*, *Pistacia lentiscus*, *Eucalyptus globulus*, *Pegnum harmala*, *Ziziphus lotus*, *Phoenix dactylifera*, *Thymus algeriensis* and *Allium spp.* among others.

1.2 Socio-economic impacts

The project itself contributes to achieve socio-economic impacts by scaling-up research activities from the laboratory dimension to the adoption of a circular model, adaptable and replicable according to inner territorial needs and characteristics, while generating new socio-economic opportunities. Agriculture and aquaculture are directly linked within this project, with the aim of enhancing their mutual potentials in a circular process, reinforcing the sustaina-



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bility of local communities, promoting knowledge transfer and creating an innovative sector of employment. Moreover, such combination constitutes an effective approach for the rehabilitation, development and the maintaining of local populations in abandoned and marginalized arid inlands, such as in Algeria. Agri-fiSh intends to create plant-based feeds to limit the animal competition on biodiversity resources, and the principal sectors of research and investigations are grape, cereal and legume productions, as well as dairy products and medicinal herbs. Attention is also given to mitigation and adaptation to climate change, since the system is conceived to reduce greenhouse gas emissions from the sector, limit or eliminate the use of antibiotics, and promote the limitation of mycotoxins and fungal contamination in feeds used for aquaculture, through the use of natural ingredients from waste valorization.

2. BACKGROUND OF THE PROJECT

2.1 Aquaculture and its environmental impacts

The United Nations General Assembly has declared 2022 the International Year of Artisanal Fisheries and Aquaculture (IYAFA 2022) (FAO, 2022), and growing attention is given to aquaculture worldwide due to the increase in its demand and productions.

According to FAO, aquaculture is among fastest growing food production sectors worldwide and has become the main source of fish proteins for human consumption since 2016, as a consequence of the marine overfishing and depletion of wild fish stocks (FAO, 2022).

Unfortunately, intensive aquaculture represents a threat for climate change and biodiversity, especially due to the release of pollutants in the environment.

According to latest publications, The State of World Fisheries and Aquaculture from FAO and World Fish (FAO, 2022), the number of people employed in the primary fisheries and aquaculture sectors is 59.5 million, of which 14% are women. In the world, 20 million people are directly engaged in aquaculture and its production has increased by 527% since 1990. About 80% of world aquaculture production comes from developing countries. Globally, aquaculture produced 82.1 million tons of aquatic animals in 2018. Europe is the continent with the highest consumption of fish products (24.5 kg/capita), higher than the world average of 18 kg per year. It currently participates in 18% of world aquaculture production and the sector accounts for almost 20% of fish production, employing about 85,000 people. Aquaculture production is projected to increase by a third by 2030, reaching 109 million tons, and by 2050 it will provide most of the "aquatic" protein in people's diets. One of the most difficult challenges aquaculture faces is the climate crisis, since the heating of the water influences the metabolism of fish and therefore the production of the supply chain.

Thus, the combination of a higher demand and the increase of climatic threats that impoverish biological resources, result in the need of providing communities with resilient and sustainable aquaculture systems to guarantee their food security. The Agri-fiSh project responds to such challenges.

2.2 Agriculture and environment

As known, the relationship between agriculture and climate change is two-sided, since agriculture is the economic sector most affected by the ongoing processes. It is a large emitter of greenhouse gases (GHGs), but it could also represent a solution in limiting GHG emissions through the development of sustainable agricultural approaches (FAO, 2013). Hence, to use agricultural discards as starting material for preparing fish feed is aimed at reducing environmental impact of both aquaculture and agriculture.

This sector emits substantial amounts of methane from enteric fermentation in ruminant animals and rice paddies, nitrous oxide from fertilized soils, and carbon dioxide from deforestation and fossil fuel use in farm operations. These emissions exacerbate the greenhouse effect, leading to global warming and associated climatic disruptions. Conversely, agriculture is also highly vulnerable to the impacts of climate change. Alterations in temperature, precipitation patterns, and the increased frequency of extreme weather events can severely affect crop yields, pest and disease dynamics, and water availability. These changes pose significant threats to food security and the livelihoods of farmers, making the agricultural sector one of the most affected by ongoing climate change processes. However, agriculture holds the potential to be part of the solution to climate change through the adoption of sustainable practices. Sustainable agriculture aims to reduce environmental impact while maintaining productivity. Organic farming, for instance, eschews synthetic fertilizers and pesticides, promotes soil health, and enhances biodiversity. CSA is another approach that focuses on increasing agricultural productivity sustainably, improving resilience to climate impacts, and reducing greenhouse gas emissions.

Thus, one innovative solution to reducing the environmental impact of both agriculture and aquaculture is the use of agricultural discards as a starting material for preparing fish feed. Traditionally, fish feed relies heavily on wild-caught fish or soy, both of which have significant environmental footprints. By repurposing agricultural by-products and discards, this approach not only minimizes waste that would otherwise contribute to methane emissions in landfills but also provides a sustainable alternative to conventional fish feed. This practice supports a more circular economy, where waste products are repurposed rather than discarded.

2.3 Agri-fiSh model

The model proposed will improve the Mediterranean agroecosystems' resilience with focus on the concept of productivity and quality valorization, and the maintaining of the well-being of the environment and the population. In fact, not only wastes, especially issued from agroecological activities, will be used as quality materials due to their nutraceutical potential, but also the testing of the final product (plant-based feeds), will enhance the nutritional quality of the fish. Moreover, by integrating agricultural discards into fish feed, both agriculture and aquaculture will reduce their environmental footprints, enhance sustainability, and contribute to broader climate change mitigation

efforts. This dual approach underscores the importance of innovative solutions and sustainable practices in addressing the complex interplay between agriculture and climate change.

Further on, consumers will find in the market a product of higher-quality level not only for their well-being but also for the well-being of the environment and the local economy. Consequently, producers (farmers and fishermen) will benefit from this virtuous system in terms of revenues and decrease in their vulnerability to climate change, since this model will guarantee yield prosperity and biodiversity abundance, while reducing the environmental impacts, by promoting sustainable agriculture and aquaculture and limiting the costs of adoption of the system.

Moreover, to respond to current environmental challenges in the Mediterranean area, the proposed model aims at identifying and undertaking innovative practices, suitable for aquaculture production and in line with Sustainable Development Goals (SDGs) and circular economy objectives, through adaptation and mitigation strategies.

3. AGRI-FISH ACTIVITIES

3.1 Partnership and support

The project's partnership is composed in Italy by the University of Camerino, being the coordinator, and the National Research Council – Institute of Marine Biological Resources and Biotechnologies (CNR-IRBIM), in Spain by the University of Valencia and in Algeria by the University Ibn Khaldoun of Tiaret.

The project is funded under Horizon 2020-PRIMA and started its implementations in June 2022, while the experimental phase of the pilot project in Italy in Visso (Marche Region) has started in October 2023 on 900 trouts (*Salmo trutta fario*) with trials on two diets and on 400 trouts (*Oncorhynchus mykiss*) with trials on one diet.

3.2 Activities

The main activities (Figure 1), relate to the definition of innovative feeds with a low environmental impact through the partial/total replacement of animal protein and lipid components with organic agricultural wastes (at least 70% of the composition of the feed), contributing to lessen CO₂ emissions from the sector; an in-depth knowledge of the bioactive compounds content of bioproducts, mainly of grape seeds from various local grapes (Italy, Spain and Algeria), legumes and medicinal plants, in order to better assign the biological properties of each compound to the desired objectives of the project in enhancing fish growth and resistance to microbial diseases, improving the taste, etc.; the valorization of organic agricultural wastes and reduction of the costs of disposal through the creation of sustainable and quality feeds, with at least 20% lower costs; the improvement of animal growth and welfare by strengthening the immune system of fish with the bioactive compounds contained in the new feeds and consequent reduction in the use of antibiotics; the creation of a virtuous and circular local system that strengthens collaborations in the agri-food sector; the definition of a sustainable and economically favourable and competitive system that

supports small-farmers in implementing organic productions, especially of legumes, which are a vegetable source of proteins and are soil nitrogen-fixing; the empowerment of local communities, with attention to gender and youth dimensions, strengthening the local dimension and the training system through direct collaboration at multiple levels (schools, universities and young entrepreneurs); the development of cooperation among Mediterranean countries through capacity building and knowledge sharing enhancement at different levels; the verification of the main organic and inorganic pollutants on the agricultural waste reused to develop the innovative feeds; and the development of a sampling protocol for sampling of water and fish in order to evaluate the state of the water quality of tanks and the well-being of the fish.

3.2.1 Testing of feed

Concerning the testing phase on fish on trout and tilapia, analysis will be conducted to evaluate the zootechnical performance and welfare of tested fish to assess the experimental diets and the setup of water quality and fish sampling protocol.

The feeding tests of the fish will be carried out to test the validity of the experimental diets containing the by-products of the sustainable supply chain. Each diet will be distributed to fish with an initial average weight of 30-40 gr for trout, and 100-150 gr for tilapia, will be reared at the same density and will be fed until a level close to satiety is reached. This will avoid subjecting the fish to forced feeding. After 90 days from the start of the test on Trout, the main growth performances will be measured (average weight, average length, average weight gain, specific growth rate, survival rate). The tests will continue by carrying out, at periodic intervals, the weight and length measurements on a representative sample of fish belonging to the different experimental and control groups. At the end of the tests, the zootechnical performance and the food conversion index for each test group will be determined. Such method will be replicated for targeted species in Spain and Algeria.

3.2.2 Water quality

Analysis will be carried out on the temperature and dissolved oxygen of the water and samples of 500 cc of water will be taken at the exit from the basins where the feeding tests of the trout with the experimental diets will be carried out, in comparison with the Control diet. To verify the functioning of the treatment path, the monitoring of the chemical-physical parameters (temperature and dissolved oxygen) and the analysis of the different nitrogenous forms (nitrites, nitrates and ammonium) present in the water from the tanks using a spectrophotometer, applying internationally recognized analytical methods. The concentration of dissolved oxygen directly affects both the self-purifying capacity of the water and the possibility of survival of the fish species. On a bi-weekly basis, as required by the protocol, the colorimetric analyzes (Strickland and Parson, 1972) are carried out after filtration (0.7 micron filter, Whatman GF/F) to quantify the presence of: NH₃, NO₃⁻, NO₂⁻.

4. PROJECT IMPLEMENTATIONS

The Project intends to solve the following problems related to productions: i) waste volumes and disposal, by valorizing them; ii) unsustainable aquaculture, by creating innovative feeds which are plant-based; iii) use of pesticides and antibiotics in agri-food productions, by promoting sustainable agriculture and using LABs and medicinal herbs in the feeds; iv) post-harvest issues, by developing a new solution to prevent post-harvest issues based on non-toxic, eco-friendly biocide intended to substitute synthetic fungicides with non-toxic eco-friendly substances (GRAS, bio-stimulants, resistance inducers and natural substances) and biological control agents (antagonistic bacteria, yeasts and fungi) to prevent the storage fungal contaminants of aquaculture feeds; v) impoverishment of agricultural system, by developing a sustainable model, which is profitable for smallholders and creates competitiveness and new employment, especially in rural areas. During these first three months of project implementation, the main research activities focused on the characterization of agricultural wastes. In fact, the valorization of wastes has a strong adaptation potential, since an abundant agriculture production results in a high quantity of processing wastes. These wastes represent an important disposal cost to be borne by farmers and agriculture producing and processing companies. Hence, it is necessary to increase value of these wastes, both reducing their disposal and related costs, and valorizing their potential and intrinsic value. The production of vegetable by-products and discards leads to the formation of waste and effluents that are naturally rich in carbohydrates, proteins, fibers and lipids, as well as vitamins and other bioactive compounds, such as resveratrol, present in wine wastes. Studies have shown the health potential for human consumption of nutrients present in grape skin and seeds, such as resveratrol and other polyphenols, which can be transferred into meat (e.g., livestock or fish) and remain still available after consumption (Frombaum et al., 2012; Ragni et al, 2014). Polyphenols, among all, have the propriety to lower cholesterol levels in blood and limit the oxidative stress in the cells for human health (Frombaum et al., 2012). Analyzed wastes are: a) grape pomace, b) grape seeds, c) discard of pasta making of: -fava beans, -peas, -chickpeas, -lentils,

-pasta made with Norberto wheat flour (wholemeal pasta), - pasta made with Senatore Cappelli wheat flour (both wholemeal and not), for a total of 9 discards/ingredients. The analysis on wastes have been directed, from one side, to the determination of nutrients in the wastes (proteins, lipids, carbohydrates, fibers), and from the other to bioactive and health promoting compounds, with special attention to macro- and micro-elements, fatty acid profile, including mono- and poly-unsaturated ones, aminoacidic profile, mineral profile with macro- and micro- elements, to end with Total Phenolic Content (TPC) and Total AntiOxidant Activity (AOA). Aminoacidic and fatty acid profiles were determined using High Performance Liquid Chromatography, coupled to a Mass Spectrometry and/or a Diode Array Detector. Mineral profile, related to 16 elements, was obtained using an ICP-MS apparatus. Finally, TPC and AOA were determined spectrophotometrically, with results given as Gallic Acid Equivalent and Trolox Equivalent, respectively.

5. RESULTS AND DISCUSSION

Table I shows some analytical results made prior to the definition of the main agricultural wastes. As expected, pomace and grape seeds showed a very high TPC (from 15 to 60 times higher than the other ingredients), while for Antioxidant activity the most effective ingredients showed to be Fava beans.

About elements, all discards showed to be from good to excellent source of them, especially for Ca, P, Mg, K, Zn, Cu, Mn.

More, pomace and, particularly, grape seed were excellent source of n-6 PUFA. First results on the testing of the two diets to trout in Itlay showed a fish growth rate of around 20% when compared to control diet but further assessments are needed since the tests will last until September 2024.

6. CONCLUSIONS

This study gives an overview of the main ongoing activities and the major expected impacts and outcomes, but it could not be exhaustive due to the recent inception of the project.



FIGURE 1: Agrifish model.

TABLE 1: Some analytical data of analyzed discards; all concentrations are expressed as mg/kg, except for fatty acid (n-6 PUFA), which is in g/100g.

	Peas	Fava beans	Lentils	Chick peas	Cappelli normal pasta	Cappelli wholemeal pasta	Norberto wholemeal pasta	Pomace	Grape seeds
Total phenolic content (mg GAE/kg)	844,55	1926,63	866,37	549,43	872,81	1293,95	1387,77	29828,9	30880,9
Antioxidant activity (mg TE/kg)	498,89	1277,82	691,31	533,35	619,2	628,26	697,77	1189,38	932,31
Ca	1101	865	303	714	243	569	804	6.299	7.543
P	3986	7998	4255	5523	1.933	4.702	5.149	3.209	3.582
Mg	1298	1788	941	1853	574	1.683	42,8	1.592	1.980
K	12994	19534	10693	15707	2.121	4.441	5.106	27.991	7.088
Zn	37,3	38,7	37,00	48,9	13,2	53,9	60,6	10,4	13,3
Cu	8,4	15,2	7,50	11,9	3,4	8,2	8,6	26,1	11,9
Mn	12,3	12,3	9,60	31,6	6,6	35,2	42,8	9,4	11,5
n-6 PUFA:									
18:2 n6 C	0,26	0,90	0,46	2,98	0,37	0,96	0,69	3,54	10,85

Beyond the state of the art, Agri-fiSh will develop an innovative model, easily replicable, and will be crucial for pursuing sustainability and creating new business initiatives in the agri-food sectors.

Due to its high-degree of novelty it will represent an asset for national economies and for further investments in the sectors (1° innovation). The standardization in the use of medicinal plants and whey (fermented by LABs) as components of fish feeds for food quality and security issues will become a protocol (2° innovation). The creation of plant-based feeds from sustainable agricultural wastes will become a protocol of innovation to be released in the market (3° innovation). This sustainable model will provide fish food enriched with bioactive constituents useful for the consumers' health, with the scope to reinforce the Mediterranean lifestyle, develop food traceability and provide products with high added value (nutritionally and economically) (4° innovation).

Particular attention is given to the use of medicinal plants, which in aquaculture has exploded recently, and have been used since ages by rural fish farmers, since they produce various beneficial effects to aquaculture such as antistress, appetite stimulation, growth promotion, immunostimulation, and have antipathogens properties due to their active compounds (Reverter et al., 2017).

Antiviral and antifungal activities are also able to prevent high mortality rates in aquaculture and the addition of LABs to the innovative feed constitutes an asset (Dadras et al., 2023).

In fact, the widespread application of many probiotic LAB species and *S. cerevisiae* in foods and their status as Generally Recognized as Safe (GRAS) organisms, makes them suitable candidates for strategies devised to diminish oral exposure to chemical contaminants and reducing mycotoxins (Chiocchetti et al, 2019).

The combination of such technologies will result in the application of an effective model, which will contribute to reduce the vulnerability of the agri-food systems face to climate change.

ACKNOWLEDGEMENTS

This paper is supported by the PRIMA Programme, which is supported by the European Union.

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SUSTAINABLE INTERVENTIONS TO MANAGE RETAIL FOOD WASTE: A SCOPING REVIEW

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Article Info:

Received:

11 April 2024

Revised:

11 June 2024

Accepted:

22 July 2024

Available online:

26 August 2024

Keywords:

Food waste

Retail

Sustainability

Waste management

Food waste prevention

ABSTRACT

Food waste (FW) affects the economy, sustainability, food security, and climate but effective practices to reduce waste in the food retail sector have received limited attention in existing studies. To address this gap, a scoping review was conducted to systematically identify and categorise research about interventions targeting retail FW. After searching four electronic literature databases, study selection allowed for the inclusion of 43 studies in the review. The narrative synthesis illustrates possible outcomes of such responses to retail FW and aggregates similar interventions and actors involved. Findings comprised 132 interventions from 41 countries, mostly European (73.2%). While supermarkets were responsible for implementing 86.4% of the initiatives, the interventions targeted both consumers (50.8%) and their own businesses (46.2%). Utilising a FW hierarchy framework, interventions were classified into seven categories, predominantly focusing on preventative measures such as awareness campaigns, price reduction, operational optimization, legal regulation, and FW repurposing for human consumption through donations. These prevention and reuse strategies hold promise across diverse contexts and warrant further exploration. About half of the interventions quantified their effects, and we address the most relevant environmental, social, and economic impacts. The findings underscore the need for research investment in the global South.

1. INTRODUCTION

In recent years, food waste has emerged as a pressing global issue. Growing concerns about food security and the socio-environmental impacts associated with food waste have attracted the attention of national and international policymakers, non-governmental organisations (NGOs), and academics. The Food and Agriculture Organization of the United Nations (FAO) underscores the urgency, noting that food loss and waste contribute significantly to global greenhouse gas emissions, making it a critical factor in addressing climate issues and global sustainability (FAO, 2022). Food waste's impacts on different fields such as food security, human health, and the environment make it a pressing area of study (Xue et al., 2017).

The literature lacks a universally agreed-upon definition of food waste, often adapting the term to suit the focus of investigations (Hanson et al., 2016). The FAO distinguishes

between food loss - occurring between agricultural production and arrival at retail - and food waste, occurring at the retail and consumption stages (FAO, 2019). However, within this framework, both food loss and food waste pertain solely to the edible portions of commodities intended for human consumption. Determining what is considered edible or inedible is complex, context-dependent, influenced by culture, evolving over time, and variable within and between populations (Nicholes et al., 2019).

This study follows the FAO definition, considering the stage of the chain where the food waste was generated while also considering the inedible parts of commodities, as in Rolker et al. (2022). Following the suggestion of Hanson et al. (2016), this broader definition was selected based on the research goal of this study: to identify food waste intervention strategies in retail. On the one hand, including the inedible part provides a more complete pic-



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ture of possible interventions; on the other, the focus on different stages of the food system helps to tackle specific interventions in the retail sector.

The global problem of food loss and waste requires more attention in food and sustainability research as well as food politics. In 2007, around a third of food produced for human consumption was lost or wasted, roughly 1.3 billion tons per year (FAO, 2011). Approximately 14% is lost between harvest and retail (FAO, 2019), while an estimated 17% is wasted in households, retail and food services (United Nations Environment Programme - UNEP, 2021). Various factors contribute to food waste, including mould (Lanfranchi et al., 2016), pests (Beausang et al., 2017), weather conditions (Göbel et al., 2015), cooking losses, and inadequate storage systems (Parfitt et al., 2010), with fundamental effects on global food insecurity (McCarthy et al., 2018; Munesue et al., 2015). Today, 822 million individuals face insufficient access to food required for an active and healthy life (Skaf et al., 2021).

Food waste contributes to global warming, acidification and eutrophication (Scherhauser et al., 2018; Tang, 2020). Approximately 8-10% of global greenhouse gas emissions are associated with edible food that goes to waste (UNEP, 2021). Food waste contributes negatively to climate change, fossil and water depletion and nitrogen pollution (Skaf et al., 2021; Grizzetti et al., 2013).

The Food Waste Index Report (UNEP, 2021) and Food Loss Index (FAO, n.d.) indicate that this challenge continues to threaten progress towards Sustainable Development Goal 12.3: to halve per capita global food waste at retail and consumer levels and reduce food losses along production and supply chains, including post-harvest losses, by 2030. In 2019, food waste from households (61%), the food service industry (26%) and retail outlets (13%) amounted to 931 million tonnes per year (UNEP, 2021). To commit to the SDG, many governments such as Australia (Commonwealth of Australia, 2017), Brazil (CAISAN, 2018), and Germany (BMEL, 2017) developed specific strategies to tackle these various and interconnected problems related to food waste on the national or local scale. A prominent example is the Food Waste Reduction Roadmap which includes a sector specific action list also addressing food waste in the retail sector as part of the UK's Waste and Resources Action Plan (WRAP, 2023).

Despite contributing minimally to overall food waste, retail remains a critical node of the food chain to explore since retailers have the capacity to influence policies, decision-making and behaviours of both consumers and producers. Presently, however, more focus is paid to other stages of the food chain, and recent literature reviews identifying viable and applicable solutions for the retail context, considering both preventive and mitigative actions, are lacking. To identify food waste intervention strategies in retail, this article proposes a scoping review to analyse and systematise existing research in this field and extract relevant gaps for this pressing global issue.

1.1 Food waste research

Food waste is an established field of research, but many studies use a supply chain perspective (Aramyan et

al., 2021; De Boni et al., 2022; Lemaire and Limbourg, 2019; Magalhães et al., 2021; Mmereki et al., 2023; Nicastro and Carillo, 2021) rather than focusing on a specific stage. This offers a broader systemic perspective but makes it difficult to identify concrete possibilities of intervention at each stage of food waste management.

Some studies were identified that focus on a specific stage of the chain, such as Onwude et al. (2020), which studied tools to reduce the quality loss of fruits and vegetables during packaging, storage, and transportation cold chain operations. Wunderlich and Martinez (2018) studied the causes of food loss and waste at the production and consumption stages, their environmental implications and the actions needed to deal with them. Lebersorger and Schneider (2011) discussed how to calculate the food waste in the household level. Meanwhile, Yetkin Özbük and Coşkun (2020) focused on downstream entities of the supply chain, showing that most existing studies focus on food services due to their relatively high contribution to food waste generation. The authors identify a research gap concerning wholesale food waste. This last study suggests that research about food waste management in the retail sector remains neglected. The absence of systematic studies focused on retail in the food chain is evident from the wider literature.

Many studies focus on specific interventions to manage food waste, such as anaerobic digestion (An et al., 2024; Assis and Gonçalves, 2022; Pilarska et al., 2023; Zhang et al., 2023), composting (Awasthi et al., 2020; Sánchez, 2022), hydrothermal carbonization (Wu et al., 2023), and animal feed production (Boumans et al., 2022; Shurson et al., 2023; Torok et al., 2021). Such research studies waste recovery and valorisation options in greater depth, but broader studies are needed that include more than one intervention and allow for their comparison while considering specific stages of the food chain.

Furthermore, studies focused on quantification of retail food waste, such as the research of Eiçaitė et al. (2022) in Central and Eastern Europe, show the large quantity generated and the reasons, which include expired shelf-life, spoilage, damage, and overstocking, and discuss the need for interventions to solve this issue. These authors even conclude the article by recommending that future research identifies actions that are being adopted in retail to reduce food waste.

We identified only three reviews specifically focused on retailers (supermarkets, markets, and wholesale), two focused on specific interventions - promotional pricing (Tsalis et al., 2021) and valorisation of animal by-products (Pinto et al., 2022) - and the final on causes of food waste and reduction practices (Moraes et al., 2020). This paper's strong contribution was limited by inclusion of studies published only between 2008 and 2017. Later reviews show an increase in relevant studies after 2015 (Rolker et al., 2022; Tamasiga et al., 2022; Yetkin Özbük and Coşkun, 2020), reinforcing the need for updated literature reviews on this topic. Furthermore, Moraes et al. (2020) only address reduction interventions, neglecting the significance of mitigation interventions, which are prevalent in low and middle-income countries' retail industries (Rolker et al., 2022).

This scoping review aims to address the identified

gaps through answering the question: what interventions by civil society, state and market to manage food waste in supermarkets and markets (retail) can be identified in the literature? By doing so, we provide an overview of existing research on interventions targeting food waste in the retail sector, including both preventive and mitigative actions, and covering more years, languages and knowledge areas than previous reviews.

The article is structured as follows. Section 2 outlines the scoping review process. In Section 3, we present our results and discussions, and some suggestions for future research. Finally, section 4 presents our conclusions.

2. METHODS

This scoping review was designed and executed in accordance with the JBI Methodology for Scoping Reviews (Peters et al., 2020) and is reported following the PRISMA Extension for Scoping Reviews guidelines (Tricco et al., 2018). Prior to commencing the review, a protocol was published on OSF Registries (Castro et al., 2022). All relevant data and procedure was made available on the review OSF page, available at: doi.org/10.17605/OSF.IO/5ES2T

2.1 Eligibility Criteria

The PCC (Population-Concept-Context) framework was used to structure the research question and determine specific selection criteria.

2.1.1 Population

This review included studies from any country introduced by three organisational types: civil society, the state and the market. Initiatives were categorised under 'civil society' where introduced by social movements, community/local actors, and non-governmental organisations. 'State' actors included political parties, national governments and transnational political coalitions. 'Market' interventions involved companies, cooperatives and business associations.

2.1.2 Concept

In this review, research concerning strategies intended to reduce food waste was considered for inclusion. This includes strategies to prevent, reduce, mitigate, repurpose or treat food waste. Studies that did not discuss actioned strategies or solutions were excluded.

2.1.3 Context

Included studies concerned food waste interventions in the retail sector: markets, supermarkets, wholesale, grocery stores, local shops and other similar food retail settings. Results from the personal/household level or concerning retail for immediate consumption (i.e. restaurants, cafes, catering etc) were excluded.

2.1.4 Types of sources

Qualitative, quantitative, and mixed-methods studies were considered. These include experimental and quasi-experimental designs, analytical and descriptive observational studies, and qualitative designs such as phenomenology, grounded theory, ethnography, qualitative

description, action research and feminist research. Journal articles and other peer-reviewed published research pieces were included, but non-academic reports and reviews of any type were excluded.

2.2 Search

The search strategy performed in October 2022 aimed to locate published studies in four electronic databases: Scopus, Web of Science Core Collection (WOS), Embase, and GreenFILE (Supplementary material - Table S.1). An initial exploratory search of Scopus, WOS, and Embase was undertaken to identify articles on the topic. Words contained in the titles and abstracts of relevant articles were used to develop the search strategy, adapted for each specific database. Search terms included synonym variations and controlled vocabulary related to "food waste", "prevention/reduction", "initiatives", and "retail" (Supplementary material - Table S.2). Studies published in Portuguese, Spanish, and English were included without time constraints.

2.3 Selection

After the search, all identified references were collated and uploaded into the Rayyan QCRI web application (Ouzzani et al., 2016), and duplicates were removed. After a pilot test, titles and abstracts were screened in Rayyan by two or three authors assessing against the inclusion criteria for the review. The full text of included documents was assessed in greater detail against the inclusion criteria by two authors. All authors (AMRCC, LLZ, CDLJ, RM, DJSC) were involved in this process, interchangeably, in pairs when necessary. After full-text evaluation, reasons for exclusion of rejected sources were recorded and reported in this scoping review. Any conflicts that arose between authors at each stage of the selection process were resolved by a third author.

2.4 Data collection

The data for studies included in this analysis were gathered using a standardised extraction form that was tested by all authors on a subset of studies (Supplementary material - Table S.3). In addition to details about the article's publication (author, year), data was recorded about the study's aims, location, scope, the actors responsible for carrying it out, the intended audience, the specifics of the intervention (including delivery methods, materials used, and duration), and other findings related to food waste.

2.5 Data analysis

Following data extraction, certain elements were compiled, such as information pertaining to the countries involved and the extent of the intervention's scope. Our primary focus was on categorising these interventions into themes. This categorisation process unfolded through a four-step iterative procedure: (1) two authors described interventions in one sentence (LLZ, CDLJ); (2) a third author compared descriptions and suggested adjustments to summarise them into robust categories (DJSC); (3) all five authors read the resulting categories and commented based on the whole process to determine the final category

(AMRCC, LLZ, CDLJ, RM, DJSC); (4) a final author (CDLJ) checked the suggestions and approved the final categories according to the seven steps of waste hierarchy (Joint Research Centre, 2020). The whole categorization was reviewed at the end by the two expert authors in the area who have the most experience with the topic, one on waste (AMRCC) and the other on food (LLZ).

In order to systematise food waste interventions, this article follows the waste hierarchy framework, which indicates an order of prioritisation of actions in waste management (Williams, 2015). The waste hierarchy aims at reducing waste generation and opting for more circular management options such as reuse, recycling and recovery, in line with the circular economy model (Ellen MacArthur Foundation, 2013).

Variations of the WH have emerged (Redlingshöfer et al., 2020; Papargypoulou et al., 2014) and, since 73.2% of the articles retrieved in this review highlight European cases, this review uses the “Hierarchy for prioritisation of food surplus, by-products and food waste prevention strategies” (Joint Research Centre, 2020). This WH has 7 stages, in

the following preference order: prevention, redistribution of food for human consumption, animal feed, reuse of by-products and recycling of food waste, recycling with nutrient recovery, incineration with energy recovery, and disposal.

3. RESULTS AND DISCUSSION

The search yielded a total of 1,316 references (see Figure 1). After eliminating duplicates, 850 references remained to be screened. These were reviewed independently by two researchers, who determined 720 of them irrelevant based upon their titles and abstracts. The remaining 130 texts underwent a thorough examination, with at least one author reading each text in full. In cases where authors chose to exclude a study, a second author checked the complete text for confirmation. Any conflicts in decision-making were resolved by a third author. Excluded full-texts are presented in supplementary material (Table S.4), with justifications. After selection, this scoping review included a total of 43 studies. All included publications are listed in Table A.1.

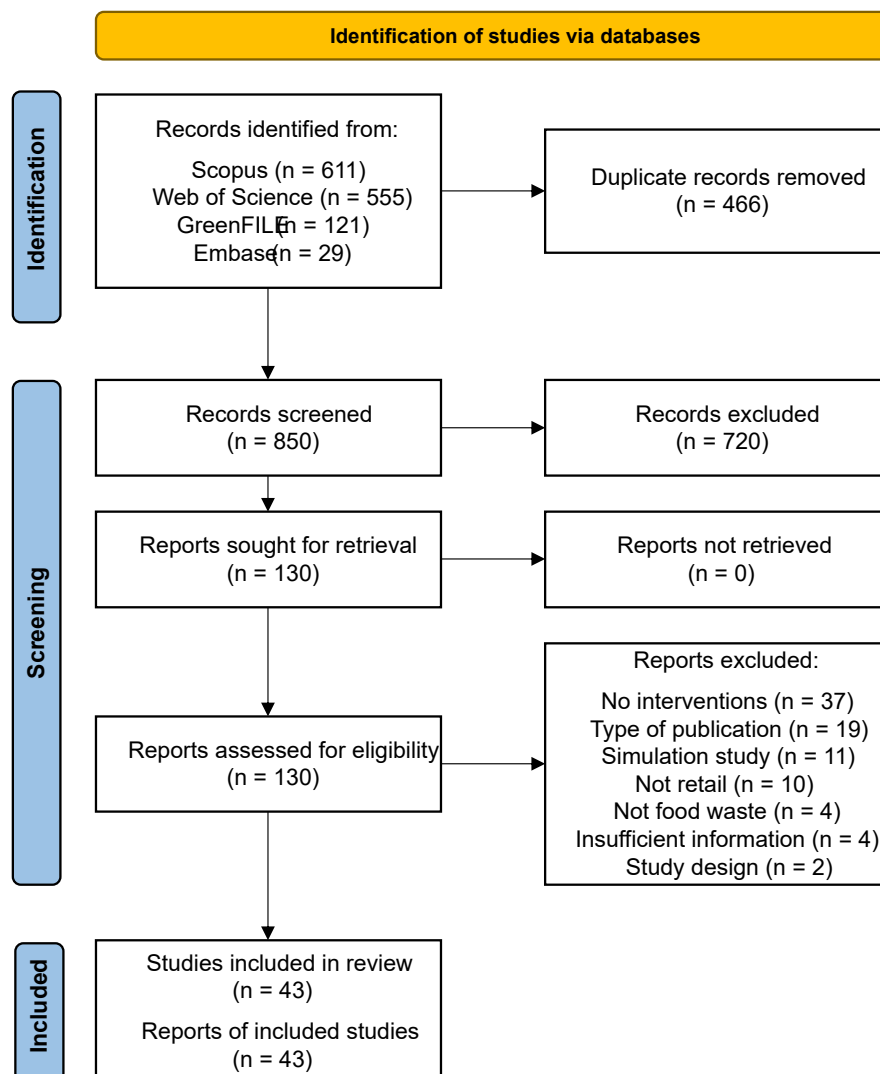


FIGURE 1: Study selection flow diagram. Adapted from Page et al. (2021).

3.1 Study characteristics

We identified studies from 2001 to 2022, a third of which were published from 2020 onwards. Most were published after 2014 (95.3%). With the exception of three cases (Holweg et al., 2016; Mullick et al., 2021; Schinkel, 2019), all studies provided details of the case's geographical location. The included studies explored initiatives spanning 41 countries, most being in Europe (73.2%). While most studies focused on cases within individual countries, two presented extensive international analyses (Chalak et al., 2018; Pulker et al., 2018).

Italy, Germany, and the UK feature most frequently in the included studies (Figure 2). The preponderance of results from Europe may be attributed to our review's language criteria, which included studies published in English, Spanish, and Portuguese. However, this imbalance raises questions about food waste management and its study elsewhere. The limited studies emerging from Latin America, despite inclusion of Spanish and Portuguese language research, might suggest little academic focus on food waste in this region. Yet Rolker et al.'s (2022) review, which extracted research in multiple languages, highlighted cases in Africa and Asia. This demonstrates the limitations of our language selection in globalising our research. Given that most literature identified in both our and Rolker et al.'s (2022) study were in English, it may instead be true that most relevant research is published in English, regardless of the researcher or case study's origins. More likely, our requirement for included publications to peer-reviewed academic works has generated a bias towards works in English, given that the language dominates academic publishing globally.

3.2 Intervention categories

We identified 132 interventions in the literature, and separated them into the 7 FW hierarchy categories to organise and discuss the results (Figure 3 and Appendix - Table A.2), using the "Hierarchy for prioritisation of food surplus,

by-products and food waste prevention strategies" (Joint Research Centre, 2020), as described in section 2.5. The majority of interventions relate to the hierarchy's most favoured approaches, prevention (50%) and re-use for human consumption (30.3%), and can be classified as preventive measures (Rolker et al., 2022). These results contrast with Rolker et al.'s (2022) findings in low- and middle-income countries, which showed that retail has more mitigative interventions (e.g., other types of reuse, recycling, energy recovery) than preventive ones.

The fourth stage of the food waste hierarchy, "re-use of by-products and recycle food waste", which includes "revalorise i) by-products from food processing and ii) food waste into added-value products by processes that keep the high value of the molecule bonds of the material" (Joint Research Centre, 2020) was not found amongst our included research. The lack of results in this category leads us to raise some hypotheses: 1) this category is still little studied since more academic attention is being given to previous stages of the hierarchy; 2) the authors may be using other words to describe these processes, other than reuse and recycling, and that is why these articles did not appear in our searches; 3) this technique may be more widely used for food waste from other stages of the chain, such as manufacturing (Mirabella et al., 2014), and is still little explored in the retail sector; 4) This category is still being further explored at a theoretical or laboratory level, as Lee and Tongaralak (2017), and as we only included concrete cases already applied by retail in our review, we did not have results.

In any case, we chose to maintain the 7 levels of the hierarchy in this article, to highlight that the lack of data found is also a result and indicates a research gap. The results found in other categories of the hierarchy are detailed below.

3.2.1 Prevention

Preventive interventions are those aimed at directly or indirectly preventing the generation of food waste. These

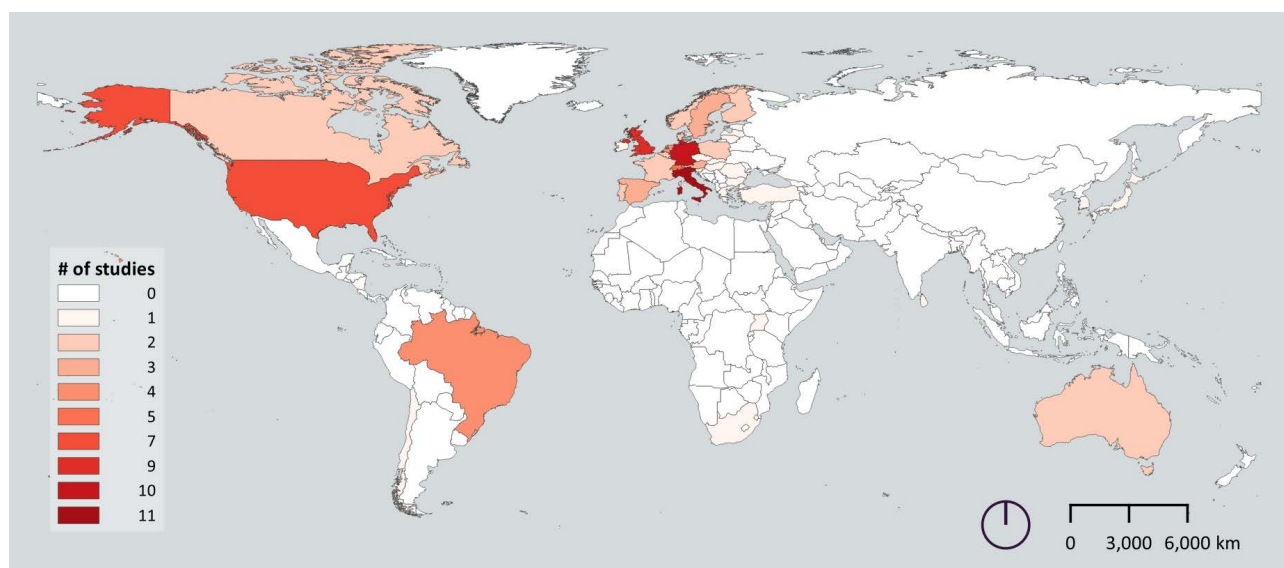


FIGURE 2: Map of studies included per country. Source: Created by the authors using QGIS (v. 3.22 Białowieża).

INTERVENTIONS ALONG THE WASTE HIERARCHY

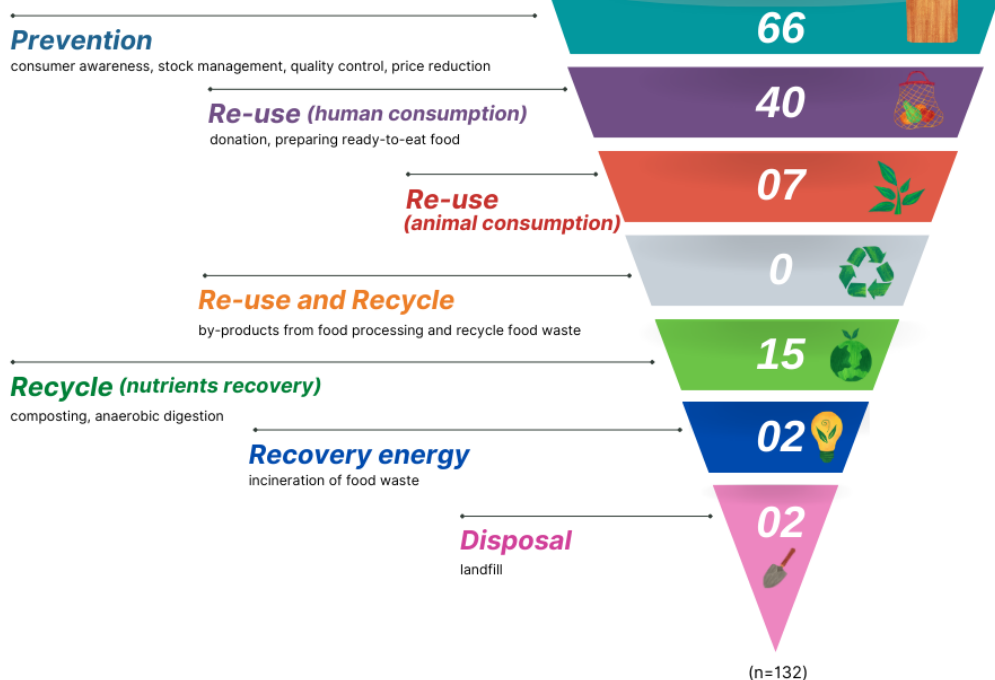


FIGURE 3: Adaptation of the European Food Waste Hierarchy according to findings (by the authors).

interventions were identified most frequently in the literature (n = 66) and were separated into 4 subcategories: awareness, corporate social responsibility practice, operations and regulations, as shown in Table 1. The four subcategories were created based on a qualitative content

analysis (Mayring and Fenzl, 2014) and a thematic coding approach (Saldaña, 2021).

Regarding the first subcategory on awareness, we found interventions focused on the actions of both employees and consumers. As pointed out by Mattsson and

TABLE 1: Categories and subcategories based on studies included in the scoping review in the waste hierarchy stage of prevention (n = 66). Source: Authors.

Category	Subcategory	n	References
Awareness	Consumer awareness	7	Busetti, 2019; Chalak et al., 2018; Fassio and Minotti, 2019; Filimonau and Gherbin, 2017; Mura et al., 2019; Schinkel, 2019; Trewern et al., 2022
	Employee awareness	5	Marrucci et al., 2020; Mattsson and Williams, 2022; Reitemeier et al., 2021; Watanabe et al., 2022; Wenderits Silva et al., 2021
Corporate social responsibility practice	-	2	Chalak et al., 2018; Moggi et al., 2018
Operations	Digital operation	1	Strotmann et al., 2017
	Packaging strategy	6	Beitzen-Heineke et al., 2017; Blanke, 2015; Filimonau and Gherbin, 2017; Mattsson and Williams, 2022; Shrivastava et al., 2022; Wu and Honhon, 2023
	Price reduction	17	Bech-Larsen et al., 2019; Blanke, 2015; Filimonau and Gherbin, 2017; Gustavo et al., 2021; Hermsdorf et al., 2017; Holweg et al., 2016; Horoš and Ruppenthal, 2021; Mattsson and Williams, 2022; Mullick et al., 2021; Reitemeier et al., 2021; Ribeiro et al., 2019; Souza et al., 2021; Tröger et al., 2020; Watanabe et al., 2022; Wenderits Silva et al., 2021; Wu and Honhon, 2023
	Purchasing management	3	Horoš and Ruppenthal, 2021; Pulker et al., 2018; Wenderits Silva et al., 2021
	Quality control	6	Horoš and Ruppenthal, 2021; Mattsson and Williams, 2022; Ribeiro et al., 2019; Souza et al., 2021; Watanabe et al., 2022; Wenderits Silva et al., 2021
	Source reduction	2	Chen and Chen, 2018; Evans and Nagele, 2018
	Stock management	12	Beitzen-Heineke et al., 2017; Holweg et al., 2016; Horoš and Ruppenthal, 2021; Marques et al., 2022; Marrucci et al., 2020; Mattsson and Williams, 2022; Reitemeier et al., 2021; Ribeiro et al., 2019; Souza et al., 2021; Watanabe et al., 2022; Wenderits Silva et al., 2021
Regulation	Legal regulation	4	Busetti, 2019; Chalak et al., 2018; Evans and Nagele, 2018; Schinkel, 2019
	Government incentive	1	Busetti, 2019

Williams (2022), although employees are usually identified as a cause of food waste in retail, they are also essential actors in its prevention when well-trained. These authors demonstrated this through a case in which employees received training and participated in regular meetings where they discussed waste quantities and goals.

Consumer awareness is also important in combating food waste. This was found in Fassio and Minotti (2019)'s study about the Re-design Project of Organic waste in Porta Palazzo market, Turin. A communication campaign focused on consumers, traders, and volunteers was amongst the approaches used to manage food waste, promoting the recovery and redistribution of food and improving the source separation of organic waste.

The second subcategory, focusing on the implementation of corporate social responsibility, was explored in a study by Chalak et al. (2018). They provided various examples of such interventions, including the 2010 commitment made by members of the Food and Drink Federation in the UK to achieve individual targets for reducing food waste, monitoring waste generation, and diverting waste away from landfills. The ultimate objective was to completely eliminate the disposal of food waste in landfills within five years. Notably, two of the country's largest food retailers embraced these initiatives. One of these, Marks and Spencer, implemented measures like price discounts on products with a short shelf life.

Prevention in retail operations can include processes to increase operational efficiency through new digital operational platforms (Strotmann et al., 2017) and management innovations in purchasing (Horoš and Ruppenthal, 2021; Pulker et al., 2018; Wenderits Silva et al., 2021), stock management (Beitzen-Heineke et al., 2017; Holweg et al., 2016; Horoš and Ruppenthal, 2021; Marques et al., 2022; Marrucci et al., 2020) and product quality control (Souza et al., 2021; Watanabe et al., 2022; Wenderits Silva et al., 2021) implemented through staff training and protocols.

Reitemeier et al. (2021) examined food waste management in Colombo amidst growing public and policy interest, highlighting stock management as a method to predict and monitor demand to minimise surplus and waste. Retailers also adopt strategies like discounting near-expiry items and staff training. Bech-Larsen et al. (2019) also focus on expiration date-based pricing. Danish retailers lower prices of items nearing expiration, without affecting sales of regular-priced goods. Beitzen-Heineke et al. (2017) examine the role of zero-packaging stores in diminishing food and packaging waste. Utilising interviews with store owners and experts, the study evaluates various operational and logistical aspects of these stores.

The last subcategory of preventive strategies comprise regulatory interventions which enforce legislation and promote fiscal and financial incentives (Chalak et al., 2018; Evans and Nagele, 2018; Schinkel, 2019). Chalak et al.'s (2018) study on food waste in the hospitality and food retail sectors highlights the effectiveness of legal regulation. The paper underscores a multi-faceted approach, combining legal, fiscal, and educational strategies, as the most effective way to address food waste in these sectors.

3.2.2 Re-use for human consumption

The second main intervention strategy identified in the literature was the reuse of food for human consumption ($n = 40$), which primarily involves interventions to repurpose food that would otherwise be wasted (see Table 2). Studies often explored donating food to food banks (Alexander and Smaje, 2008), staff (Marrucci et al., 2020), or consumers (Buseti, 2019; Chalak et al., 2018).

Alexander and Smaje (2008) evaluate the impact of large retailers' food donation programs through the British charity FareShare, tracing back to understand the donation flows. What stood out in FareShare's donation system were the different food waste destinations. A considerable amount was donated to projects working against food pov-

TABLE 2: Categories based on studies included in the scoping review in the waste hierarchy stage of re-use for human consumption and recycling. Source: Authors.

Stage	Category	n	References
Re-use for human consumption	Donation (non specified)	24	Alexander and Smaje, 2008; Arnold, 2022; Beitzen-Heineke et al., 2017; Cakar, 2022; Cannavò et al., 2019; Chen and Chen, 2018; de Hooge et al., 2018; Evans and Nagele, 2018; Fassio and Minotti, 2019; Filimonau and Gherbin, 2017; Garrone et al., 2014; Hermsdorf et al., 2017; Holweg et al., 2016; Horoš and Ruppenthal, 2021; Mattsson and Williams, 2022; Mura et al., 2019; Ng et al., 2019; Pulker et al., 2018; Reitemeier et al., 2021; Ribeiro et al., 2019; Sisson, 2016; Warshawsky, 2016; Watanabe et al., 2022; Werderits Silva et al., 2021
	Donation to food bank	6	Alexander, 2021; Bech-Larsen et al., 2019; Blanke, 2015; de Hooge et al., 2018; Garrone et al., 2014; Mura et al., 2019
	Donation via digital platform	3	Filimonau et al., 2017; Gollnhofer, 2017; Sewald et al., 2018
	Preparing ready-to-eat food	6	Alexander, 2021; Arnold, 2022; Beitzen-Heineke et al., 2017; Holweg et al., 2016; Reitemeier et al., 2021; Watanabe et al., 2022
	Social supermarket	1	Bech-Larsen et al., 2019
Recycling	Anaerobic digestion	3	Mattsson and Williams, 2022; Ng et al., 2019; Warshawsky, 2016
	Composting	7	Alexander, 2021; Chen and Chen, 2018; Evans and Nagele, 2018; Fassio and Minotti, 2019; Holweg et al., 2016; Reitemeier et al., 2021; Ribeiro et al., 2019
	Biodiesel production	1	Chen and Chen, 2018
	Industrial use	1	Evans and Nagele, 2018
	Recycling (unspecified)	3	Filimonau and Gherbin, 2017; Marrucci et al., 2020; Werderits Silva et al., 2021

erty; FareShare delivered fresh fruit, vegetables, and ready-to-eat meals donated by retailers. Another flow comprised direct donations to elder care centres, hostels and homeless shelters. Some donations were also used in animal feed.

The study by Sewald et al. (2018) described the potentials and limitations of food donation via digital platforms. They assessed the 'Food Rescue Robot', a digital tool used by Boulder Food Rescue for managing fresh food donations and distribution. This web-based application was replicated in over 40 cities and focuses on facilitating community participation in the donation process.

Fassio and Minotti (2019) present their case study, RePoPP (Re-design Project of Organic Waste in Porta Palazzo market), in Turin, Italy as a key example for successful food waste donation. Here, the redistribution of food recovered occurs through distributing fruit and vegetable boxes aiming to feed a family for two days. Further, the initiative has a strong social and integrative facet, since 43 of the programme's volunteers are asylum seekers.

Alexander's study (2021) explores donation interventions by Cultureghem, a Brussels-based NGO. The paper focuses on programs like "Barratoir," a zero-waste pop-up restaurant, and "Collectmet," which distribute surplus market produce to people in need. The initiative repurposes unsold food for communal meals and distribution, effectively reducing food waste while promoting community involvement and social inclusion.

3.2.3 Re-use for animal consumption

Re-use for animal consumption was only mentioned in 7 studies (Chen and Chen, 2018; Evans and Nagele, 2018; Holweg et al., 2016; Horoś and Ruppenthal, 2021; Ng et al., 2019; Reitemeier et al., 2021; Watanabe et al., 2022), suggesting that limited research focuses on this strategy. Ng et al. (2019), for example, explore valorisation of food waste into animal feed as a facet of resource recovery. The study assesses the approach's potential in contributing to a circular economy and lessening environmental impacts. It underscores the need for cohesive and collaborative waste management strategies that encompass both commercial and domestic sectors, demonstrating the viability of transforming food waste into animal feed.

3.2.4 Recycling

When reuse is not possible, recycling provides an opportunity to recover the value of food waste, promoting recovery of nutrients through composting, and energy through anaerobic digestion. The works that addressed recycling initiatives in retail are listed in Table 2, including biodiesel production, unspecified industrial use and recycling.

Fassio and Minotti (2019), for example, analysed composting as a measure to manage food waste at the Porta Palazzo market in Turin, Italy. The composting initiative was a part of the RePoPP project since the very beginning, aiming to reduce waste by transforming it into compost for agricultural use to close the circle of food waste to production through recycling.

Three studies explored energy generation through anaerobic digestion. One example, Warshawsky (2016),

presents the Kroger Recovery System (KRS) in California, where anaerobic digestion is used to transform 150 tons of unused food daily into renewable energy.

3.2.5 Energy recovery

Alongside Warshawsky (2016), two studies focused on energy recovery from FW, including incineration as well as anaerobic digestion. However, it is worth noting that these two options are separated within the waste hierarchy. Anaerobic digestion should be prioritised and has more desirable by-products (fertiliser and energy) than incineration (ash and gas).

Mattsson and Williams (2022), showed that waste generated in the stores studied was collected by a waste company and used for biogas production or incinerated for energy recovery. Ng et al. (2019) also highlight these two approaches in their case study of Tesco supermarkets, where 54.3% of surplus food considered unsafe for human consumption was sent to anaerobic digestion and 11.3% to incineration.

3.2.6 Disposal

Disposal, which is the least preferred strategy for managing food waste according to the hierarchy, was mentioned in only two included publications. Garrone et al. (2014) showed that 95% of surplus food waste generated in a large Italian retailer were sent to disposal by the municipal waste management system. They attribute this to the standard practice of keeping products on shelves until their expiration date, making redistribution impossible. The authors also highlighted the lack of economic incentives to avoid landfills, since waste disposal tariffs are mainly fixed fees.

In another example, Holweg et al. (2016) discusses a differentiated collection system, with separation between organics (fruits, vegetables, bread and dairy products without packaging) and waste (everything else not donated or sent to recycling). Both are collected by companies however no data showed the percentage of total waste sent for final disposal. Furthermore, the low number of studies found and their year of publication corroborates the argument that disposal is the least desirable strategy for food waste management and researchers are likely dedicating more time investigating alternative options.

3.3 Stakeholders: implementation actors and target population

The 132 different interventions identified were implemented by a range of different actors (see Figure 4), with supermarkets involved in 86.4% of them. Notably, the only interventions where supermarkets typically did not take on the implementation role were regulatory initiatives. Following supermarkets, government (14.4%), NGO (14.4%), community (8.3%) and academic (7.6%) actors were responsible for implementing food waste interventions.

The included studies described interventions targeted mostly consumers (50.8%) and supermarkets (46.2%). Charities (16.7%), food banks (13.6%), farmers (12.1%) and markets (8.3%) were also key beneficiaries. Reflecting our findings, Moraes et al.'s (2020) review showed a myriad of

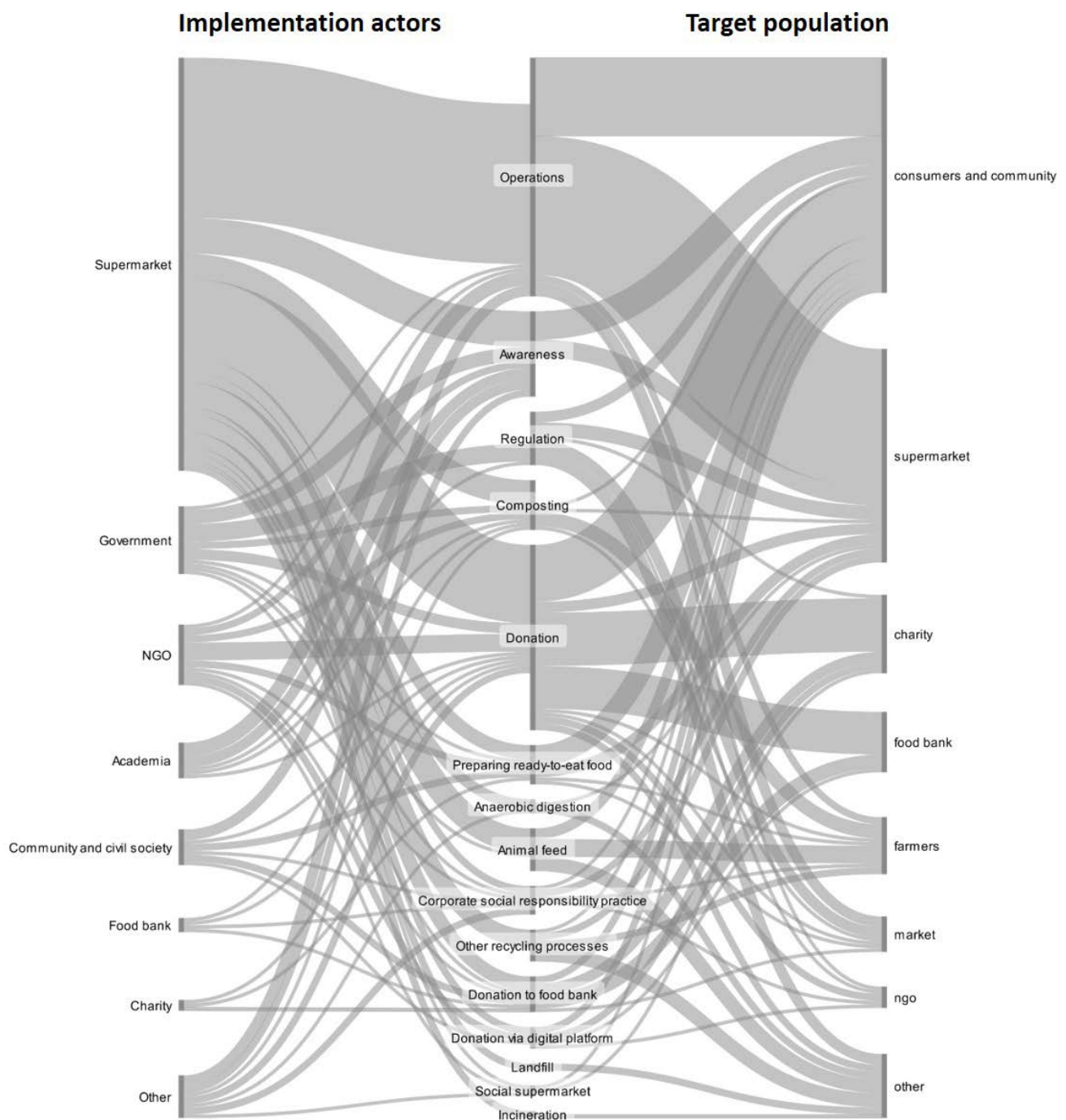


FIGURE 4: Types of actors involved in the implementation and target population of interventions described according to category. Source: Created by the authors using SankeyMATIC.

stakeholders involved in the problem of retail food waste. Consequently, the complexity of solutions to retail food waste persists.

3.4 Intervention outcome

To evaluate the impact of the interventions identified in the literature, we mapped outcomes of each study and added them to our database (see dataset on OSF). However, identifying and assessing the outcomes of interventions posed a challenge since few articles offered identifiable direct and measurable outcomes. Only 22 of the 43 analysed

articles identified quantifiable outcomes, with a total of 91 concrete measurable results (see supplementary material - Table S.5). Most articles instead presented not clearly measurable, superficially described or no intervention outcomes at all. Consequently, systematised outcomes here do not represent all interventions identified and categorised in Section 3.2. Below, we instead present a collection of outcomes mentioned in the articles.

After data extraction, in the initial systematisation step, we organised outcomes along the interventions and stages in the waste hierarchy. Appendix Table A.3 summarises

the main outcomes of prevention interventions identified in our literature review. Of the 66 preventive interventions, outcomes were defined in only 19 cases (28.8%). Results were quantifiable in only eight cases (marked in green). Furthermore, no information about outcomes was found for five of the preventive intervention subcategories identified: employee awareness, purchasing management, quality control, source reduction, and government incentive.

Table A.3 shows the broad range of outcomes found through the review, demonstrating the challenge of systematically presenting these findings. Consequently, we present here a selection of exemplary interventions that have a demonstrable, quantifiable and direct outcome on food waste in the retail sector, focusing on interconnected environmental, social, and economic impacts of these interventions.

Exemplifying quantifiable direct environmental outcomes, Warshawsky's (2016) research about supermarket chain Kroger's food waste recycling initiative highlighted their conversion of 150 tons of food waste into biogas daily, powering 20% of their distribution centre with 13 million kWh of electricity. Despite this effectiveness, Warshawsky notes the project's high cost, and lack of data on environmental impact provided by Kroger management. Consequently, he argues that promoting this initiative aims to project a positive image of the company, rather than having tangible environmental benefits.

Another example of positive environmental impact is Reitemeier et al.'s (2021) study of the food waste landscape in Colombo, Sri Lanka. There, many hotels, restaurants, markets and canteens redistributed food waste unsuitable for commercial use for re-use through animal consumption. 23 of 24 piggeries surveyed employed this practice, collecting a daily average of 1480 kg of food waste. Farmers independently collected waste, visiting up to 20 sites each and collecting around 80% of the food businesses would otherwise waste. In minimising required resources for animal feed production, farmers have a positive environmental impact here.

An example of quantifiable social outcomes is Fasio and Minotti's (2019) study of the effects of consumer awareness campaigns in Turin, Italy. They noted an 11.3% decrease in total waste from 2018 to 2019, primarily due to a 9.5% reduction in general waste. This reduction indicates that increased consumer awareness led to a decrease in food waste. Approximately 150,000 kilograms of food was rescued and distributed to individuals in need. Consequently, food waste was reduced while simultaneously making a social contribution.

Social impact is also demonstrated through the donation via digital platform described in Sewald et al. (2018). Its implementation by Boulder Food Rescue significantly reduced food waste through redistributing for human consumption. In 2016, the organisation saw a 423% increase in redistributed food compared to the previous year, equaling 4.1 million pounds annually. Their Food Rescue Robot facilitated 443,000 pick-ups, handling 44 million pounds of food in 40 cities, with help from 42,000 volunteers. This approach mitigated food waste while also enhancing food security in communities.

Direct economic outcomes are illustrated in Alexander's (2021) ethnography of the Cultureghem NGO and its work in a Brussels market. The organisation established a zero-waste pop-up restaurant on the site, serving regular meals produced using vendors' unsold produce. This reduced vendors' waste disposal costs, ensuring 40% of their edible food waste was recirculated instead of incinerated, while feeding up to 200 people weekly. Cultureghem's weekly food bank also distributes fresh goods to anyone attending, and their composting initiative allowed cattle farmers to collect waste for fertilising when bringing their animals to the market. Contributing to a circular economy, these initiatives also had a positive social impact on the city's poorest communities residing around the Abattoir, who could receive fresh produce and cooked meals at minimal cost.

The studies discussed above showed direct or indirect linkages between economic, environmental and social impacts of food waste interventions in retail. However, the lack of clear indicators to measure and evidence positive outcomes for the environment, economy or society is a limitation of this body of research, presenting a major challenge for comparative work on food waste, and implementation of interventions. To increase the practical utility of these studies, more systematised research on quantitative and qualitative outcomes of food waste interventions is needed.

3.5 Strengths and limitations

This scoping review was systematically conducted in accordance with established methodologies and reported following recognized guidelines after a protocol was published. The comprehensive search strategy, two-step selection involving all authors and a pilot test, adds rigour to study selection. Data collection and analysis employed a standardised extraction form and a systematic categorization approach aligned with a well established waste hierarchy framework. However, some methodological choices may limit our findings. There is potential language and geographical bias due to the exclusion of grey literature and language restrictions. Additionally, selection bias may have occurred during screening and selection processes, even with efforts to resolve conflicts among multiple authors.

4. CONCLUSIONS

This scoping review identified 132 interventions related to food waste management in retail across the 43 articles analysed. Studies focused on initiatives in the northern hemisphere, especially Europe, and published after 2020, when the 'Hierarchy for prioritisation of food surplus, by-products and food waste prevention strategies' was established in this region, dominated our findings. This policy may have increased the number of interventions in the region, causing greater research interest and, consequently, more publications. We argue that this regional discrepancy is not a problem and that interventions identified in countries where legislation is more advanced in combating food waste can serve as inspiration for other locations, providing they are adapted to the region's unique characteristics.

The FW hierarchy category with the most results was

prevention (50%) while incineration (1.5%) and final disposal (1.5%) had the least. Research therefore appears to prioritise interventions favoured by the hierarchy. This focus may also reflect pressing political discourses in the global North on innovation, circular economy and sustainable transformation, leading research to focus on novel alternatives.

Within the prevention category, price reduction was the most investigated subcategory (25.8%), which can reduce food waste within retail environments while encouraging increased purchasing and later wastage in households. However, recent research has challenged this norm, suggesting that such purchasing can reduce wastage across the food chain as a whole (Wu and Honhon, 2023). These contradictions suggest that retail interventions should be analysed with caution, and support the benefit of whole-systems approaches when dealing with the challenge of food waste.

Donation initiatives were also prevalent in this review (25%), including interventions related to food banks, through digital platforms and other unspecified types. The high quantity of such interventions may be associated with preferences to offer food to those experiencing food insecurity, an intensifying challenge following the pandemic. This reflects the political urgency of addressing food security which involves deep humanitarian and ethical aspects on top of policy challenges.

Identifying and analysing the results of identified interventions was highly challenging since few studies provided quantitative data to contextualise FW reductions, donations or designations. In qualitative studies, identifying the tangible outcomes of each intervention was difficult. The absence of clear and comparable data on outcomes hindered our ability to effectively evaluate the interventions identified. Therefore, we recommend more researchers seek to quantify or qualitatively analyse the impact of each intervention applied. A universal standardised indicator to measure food waste in the retail sector would be an important step in facilitating comparative studies in the future.

This research originated from collaboration between researchers with different disciplinary backgrounds, based in countries across the northern and southern hemispheres. The consequent multiscalar and interdisciplinary perspective this brought to the study led us to recognise the benefits of initiatives which require fewer technology and resource investments, particularly those in prevention and reuse (e.g. price reduction, employee training, donation), which might have greater potential for application in diverse retail contexts.

Thus, future studies must focus on food waste management in retail in the regions of the global South, about which we found fewer publications. Furthermore, we recommend further research on policies relating to food waste, their application and regulation, such as the waste hierarchy in Europe, since such frameworks appear to influence retailers' approach to food waste management.

ACKNOWLEDGEMENTS

The authors would like to thank Robert Cowley, Sergio Costa, and Fabio Kon for their initial supervision during the

Global Research Academy: Sustainable Cities (GRA), where this review was designed. We would also like to thank Universidade de São Paulo, Freie Universität Berlin, and King's College London for their work in designing and facilitating the GRA programme. This work was made possible in part by doctoral grants #2021/02334-0 and #2022/12729-4, São Paulo Research Foundation (FAPESP), respectively to CDLJ and AMRCC. LLZ is hired as a doctoral researcher in the project Food for Justice: Power, Politics and Food Inequalities in a Bioeconomy financed by the German Federal Ministry for Education and Research (BMBF). RM received doctoral funding from the London Interdisciplinary Social Science Doctoral Training Partnership (LISS-DTP/ESRC).

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WASTE VALORISATION FROM PHOTOVOLTAIC PANELS AND ANALYSIS OF THE BUSINESS POTENTIAL FROM RECYCLING IN SPAIN AND ITALY

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Article Info:

Received:
30 April 2024
Accepted:
10 June 2024
Available online:
30 September 2024

Keywords:

Photovoltaic panels
Circular Economy
PV recycling

ABSTRACT

Spain and Italy stand out as frontrunners in renewable energy adoption, particularly in photovoltaic (PV) technology, as part of the strategy for the Energetic Transition to clean sources. However, as Europe progresses towards clean energies, managing the end-of-life waste from solar panels emerges as a pressing challenge. In the coming years, the PV waste will grow exponentially, reaching 23 and 39 kt in 2030 in Italy and Spain, respectively. European regulations require that these wastes be properly managed using Circular Economy techniques to maximise the recovery of its components. Among these components, Si or Cu (CRM) are particularly important because of the supply risk and strategic importance to the EU. This study aims to forecast the magnitude of this waste stream in Spain and Italy from 2024 to 2030, both significant players in solar energy production, while also exploring the potential economic opportunities associated with its recycling.

1. INTRODUCTION

The general growing concern about global warming led to the holding of the “United Nations Climate Change Conference (COP21),” in 2018 in Paris, where the Paris Agreements were established “to substantially reduce greenhouse gas emissions to limit the global temperature increase this century to 2°C and to strive to limit this increase to even more than just 1.5°C” (Balthasar et al., 2019). These agreements forced an Energetic Transition to cleaner energy sources, such as renewable energies, to reduce greenhouse gas emissions (GHG) and the use of fossil fuels, both are key factors against global warming (Wolniak et al., 2022; Kumar, 2014). Since then, renewable energies suffered a boost in the development and implementation (Balthasar et al., 2019; Hinrichs-Rahlwes, 2012).

The European Union (EU) is a strong and active proponent of setting global ambitions and targets for climate protection (Młynarski, 2019). It has placed itself as one of the world leaders in energy transformation, reducing GHG emissions and substituting fossil fuel power generation with renewable energy (Wolniak, 2022). Among all renewables, solar energy is one of the most abundant and potential sources, contributing to 26% of global energy production (Kumar, 2014). Solar photovoltaic (PV), in particular, has great potential in terms of both technical and sustainable solutions for energy needs (Kumar, 2014). The PV pow-

er capacity in the EU is growing every year (Wolniak, 2022). It peaked in 2011, with 70% capacity of the worldwide installations. Now, the EU market accounts for only 50% of the worldwide PV capacity (Wolniak et al., 2022; Madsen et al., 2019).

The role of Spain and Italy in the global energy market should be highlighted because both countries are at the head of the PV capacity in the EU (EurObservER, 2023). These countries started with PV installations in the 90s, but it was not until the early 2000s that they began to build installations consistently. The two countries have developed this industry at different rates due to their circumstances. Both had a subsidy system to encourage installation (Ministerio de Industria, Turismo y Comercio, 2008; Ministerio de Industria, Turismo y Comercio, 2007).

In Spain, due to the financial crisis in 2008, the government removed the funds. This marked the end of the first Spanish “boom”, which lasted from 2006 to 2008 and represented 3.384 MW installed. The second boom began in 2018 and continues to this day. By the end of 2022, the total number of installations increased to 67.886 with a total capacity of 22.256 MW.

The Italian growth period is only one and took place from 2005 to 2015. It overlaps with the European growth peak in 2011, when Italy had already installed 13.216 MW. In 2022, there were 1.225.431 PV installations, with a total



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capacity of 25.064 MW. These figures highlight the difference in capacity of the Italian and Spanish PV installations, as Italy is more focused on self-consumption and Spain on industrial generation, which usually has bigger plants with greater capacity.

When PV modules were first installed in the EU in the 2000s, they had a lifespan of 25 years; as the technology progressed, the lifespan increased to 35 years in 2023 (Sultan et al., 2022). However, the capacity to transform light into energy is not constant over the years. Up to 10 years, the module's transformation yield remains constant at more than 90% of conversion. Then, it drops to 80% in 25 years. Some modules need to be replaced in these years, generating waste. The expected end-of-life of the modules combined with the number of installations in both countries raises concerns about the volume of waste that is going to be generated by PV modules (Marcuzzo et al., 2022; Chowdhury et al., 2020) and the difficulties related to its management.

This study analyses the forecast of waste generation in Spain and Italy in the following years and assesses the potential value of this specific waste to identify the business opportunity of PV recycling, in addition to the needs regarding waste management and the Circular Economy policies.

2. PHOTOVOLTAIC PANEL WASTE PRODUCTION

The data of the historical installations, together with the end-of-life of the solar panels, provide a forecast of the waste generation in the coming years. It is presented in Figure 1.

The waste growth forecast for the coming years is exponential. Spain expects to reach 5 kt in 2025, while Italy will not reach 5 kt until 2027. By 2030, Spain will be close to 40 kt and Italy 25 kt.

The increase in waste is mainly due to the ageing of the PV panels. As the first installations in Spain were built between 2006 and 2008, the systems are now around 20 years old, and some will soon need to be replaced. The boom in Italy came years later, and the situation will be the same as in Spain, with a delay of a few years (around 2

years). As the years go by, the installations will become obsolete and produce more waste.

Although the specific number of facilities, their capacity and their age are available parameters, there is no clear consensus on the rate of waste generation. Based on the available literature, the amount of accumulated waste expected by 2030 in Spain is estimated to be between 71 and 180 kt (Santos et al., 2018), which is in line with the data shown in the graph. On the contrary, the Italian prediction is quite far from the data presented. In 2015, a forecast predicted a waste accumulation in 2030 of 757 t compared to the 23.000 t displayed in Figure 1 (Paiano, 2014). This estimation only considers those installations before 2013, while the model used in this research considers all installations until 2023. What can be said, for sure, is that PV waste generation will increase exponentially in the coming years and have to be managed.

3. PHOTOVOLTAIC PANEL WASTE VALUE

The waste generated by the PV industry is a potential source of business, although PV waste processing still presents some challenges in terms of economic feasibility (Masoumian et al., 2015). The economic profit of PV recycling is around 0,58 \$/MW, which means around 0,0076 €/t (Corcelli et al., 2018; Peeters et al., 2017). This small profit potential, coupled with the challenges of proper waste management and fluctuating commodity prices, makes PV waste recycling a real challenge (Peeters et al., 2017).

In EEUU, which had a PV energy cumulative capacity of 40.500 MW in 2020, 450 M\$ value creation is predicted (Majewski et al., 2021) between 2030-2060 (Domínguez et al., 2019). Comparing the PV capacity of EEUU against Spain and Italy's capacities (11.700 MW and 21.700 MW, respectively), it is possible to expect an interesting business opportunity in both countries.

The average composition of PV panels based on the bibliography (Paiano, 2014; Corcelli et al., 2018; Choi et al., 2014; Latunussa et al., 2016), and the stock prices of different components sourced from the Mineral Commodity Summaries report of the United States Geological Survey (US Geological Survey, 2023; Mineral Commodity Summaries, 2024; Di Giuseppe, 2020; Recycling, 2024) and differ-

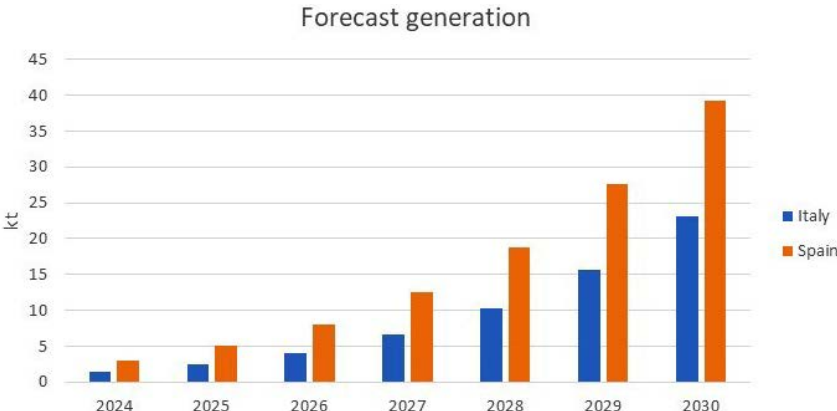


FIGURE 1: Forecast generation of Spanish and Italian PV waste from 2024 to 2030.

ent bibliographies (EVA Prices, Monitor, 2024) have been analyzed is displayed in Table 1.

Not all the Secondary Raw Materials (SRM) listed in the previous table present potential for valorization. The ethylene vinyl acetate (EVA) and the polyvinyl fluoride (PVF) are not always recoverable from the thermal processes applied for the PV waste recovery (Majewski et al., 2021; Latunussa et al., 2016). The recycling yield of each material indicated in the previous table is based on the bibliography (Domínguez et al., 2019; Choi et al., 2014). However, it is strongly dependent on the recycling technique. Moreover, Silicon and Silver show very high recycling rate, which can not be reached with standard mechanical techniques but with a reduction technology expected in 2026 (Domínguez et al., 2019).

The combination of the recycling yield with the current price of basic materials and metals (US Geological Survey, 2023; Mineral Commodity Summaries, 2024; Di Giuseppe, 2020; Recycling, 2024) provides an initial estimation of the PV waste value of 748,07 €/t.

Based on the waste production data per year, the expected business opportunity from PV recycling for each country is shown in Table 2.

Considering only the potential revalorization of metals and glass for the estimated PV waste generation, the potential business opportunity for PV waste recycling in Italy, together with Spain, is around 133 M€.

However, another study established a maximum OPEX of 596 €/t to produce silicon of metallurgy grade (Spath et al., 2022), which would reduce the PV waste recycling

business in Spain and Italy to around 27 M€ in the next six years.

All this underlines the importance and fundamental role of recycling technologies. It also shows that they will be a key factor in solving the waste management problem. However, there is room for improvement in terms of economic viability, the key factor being the development of improved technologies for the recovery of the various components.

Several promising techniques for the recovery of the main metals are currently available in Europe (Aravelli et al., 2021; Dias et al., 2019; Dias et al., 2018; Dias et al., 2016). One of these opportunities is developed in the PARSIVAL project, whose main objective is to recover and recycle silicon from solar cells and reuse it for different applications such as Li-ion batteries, ferroalloys, and after a purification process, again in PV cells.

4. CONCLUSIONS

This research shows that with the development of solar PV energy in Europe and the installations made during the 2000s, especially in Spain and Italy, they will face the challenge of the imminent generation of waste from PV panels. By 2030, these two countries will account for around 65 kt of PV waste.

Proper management of this waste is essential to raise a sustainable business model along the entire value chain and also to achieve the actual transition to clean energies. Although this management can be a challenge, it creates an opportunity for a source of Secondary Raw Materials.

TABLE 1: General composition of PV waste and unit value of the components in the market.

Secondary Raw Materials (SRM)	Composition (kg/t)	Recycling yield (%)	PV waste value (€/ PV t)	Stock Price (€/ton)
Glass	739,42	96,96%	46,60	65,00
Al	166,30	100,00%	402,20	2.418,55
Cu	20,96	88,89%	122,26	6.562,02
Ethylene vinyl acetate (EVA)	81,76			3.166,67
Polyvinyl fluoride (PVF)	35,27			8,26
Si metal	39,96	92,31%	144,11	3.907,08
Ag	0,06	95,00%	32,90	588.119,66
Others	11,60			
Total	1000		748,07	

TABLE 2: Estimation of the PV waste value per year in the Spanish and Italian market.

Secondary Raw Materials (SRM)	Italy (€)	Spain (€)	Total (€)
2024	1.054.380	2.299.634	3.354.015
2025	1.822.489	3.782.871	5.605.360
2026	3.040.429	6.036.272	9.076.702
2027	4.909.806	9.355.719	14.265.525
2028	7.691.330	14.092.824	21.784.154
2029	11.705.538	20.628.461	32.334.000
2030	17.321.172	29.316.754	46.637.926
Total	47.545.145	85.512.538	133.057.684

This business opportunity should be deeply analysed, considering not only the income but also the operating costs.

From the PV waste components with recycling potential, aluminium is the most valuable element because of its high content, and silver due to the high-market value of the metal. Silicon and copper are important, not only due to their economic value but also due to their supply risk in the EU, which makes them Critical Raw Materials. Currently, the PV waste value is estimated to be around 748 €/t.

Currently, there are several PV waste recycling techniques in development, and the main challenge is to develop a recycling process with low operative costs to make it economically feasible for SRM recovery. The PARSIVAL project is an example of this and could improve the sustainable supply of raw materials to the EU.

In any case, the business potential in Italy and Spain over the next five to six years is promising. Approximately 85 M€ (gross revenues) can be obtained for PV waste treatment.

There are several issues that are out of the scope of this publication but should be analysed in order to better understand the future of this waste. Firstly, a deep study and validation of the model used for waste forecasting is needed. This model could be applied to other countries of the EU to analyse the business potential as a whole. In addition, an in-depth analysis of the recycling techniques is needed to identify the value of the products (metals) obtained. In particular, the grade of silicon metal obtained from the PV waste recycling is a key parameter. Finally, it is necessary to know the costs of the recycling operations in order to analyse the overall economic viability.

ACKNOWLEDGEMENTS

The authors thank the Gomez Pardo Foundation for its support of the research and development of this scientific publication.

FUNDING

This research was partially funded by the PARSIVAL project PN 22001. Supported by EIT RawMaterials co-funded by the European Union.

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RESEARCH ACTIVITY ON PV WASTE RECYCLING IN ENEA

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Article Info:

Received:

5 May 2024

Accepted:

28 June 2024

Available online:

7 September 2024

Keywords:

EoL PVs

Recycling

Silicon

ABSTRACT

ENEA is dedicating significant efforts to the research activity concerning the treatment of End-of-Life (EoL) photovoltaic panels (PVs), which is a worldwide emergent issue to be urgently solved. With PVs typically having a lifespan of 25 years, the volume of PV waste is projected to increase rapidly by 2030. This issue is compounded by the composition of photovoltaics, which includes hazardous materials, necessitating the development of technological solutions to mitigate the environmental impact of commonly used recycling methods. Additionally, strategic materials present in PVs make the valorization of recovered materials highly advantageous. ENEA is actively involved in several research projects focused on EoL PVs recycling. This article will outline the main objectives of these projects and present the key findings achieved by ENEA's research team.

1. INTRODUCTION

The installed photovoltaic (PV) power worldwide has experienced exponential growth since 1990, reaching 1047 GW by the end of 2022 (Irena, 2023). Under IRENA's 1.5°C Scenario, this growth would continue, exceeding 5400 GW by 2030 and 18200 GW by 2050 (Irena, 2023). Considering the typical lifespan of a photovoltaic panel, which ranges from 25 to 30 years, it is estimated that approximately 60 to 78 million tons of panels will require disposal globally by 2050 (Irena and IEA-PVPS, 2016).

The performance of PV modules can degrade due to various factors, including UV irradiation, high temperatures, humidity (Oreski, 2005), mechanical shock, local pollutants, and toxic fumes. Aging effects can lead to electrical shock, moisture and pollutant ingress, and overall failures of PV modules. It's worth noting that the improper disposal of EoL PVs not only results in the loss of resources such as silicon, aluminum, and glass but also poses significant environmental risks due to the leaching of metals like Al, Cu, Ni, and Pb from crystalline silicon (c-Si) or amorphous silicon (a-Si) module pieces, depending on pH conditions.

Dismissed PVs constitute a resource of useful and precious materials (silicon, silver, copper, aluminum). Some of them, such as silicon, are classified by the European Union as strategic materials, i.e. materials that can be used for strategic areas such as renewable energy, digital, aerospace and defense technologies, but they are at risk of potential future supply shortages; that is, global imbalances

between supply and demand are expected for these materials. Additionally, strategic raw materials possess unique properties that make them challenging to substitute in various technologies. For instance, silicon is utilized in approximately 95% of PVs currently available on the market for converting solar energy into electrical energy (Fraunhofer, 2023). China stands as the primary global supplier of silicon, accounting for 76% of the worldwide production (CRM, 2023). Silicon holds critical importance not only in the photovoltaic and electronics industries but also in metallurgy and numerous other supply chains (SCREEN, 2023; Carrara, 2023). Despite silicon being the second most abundant element in the Earth's crust after oxygen (RSC, Daviddarling), it primarily exists in nature as silica (SiO₂). Its production from silicon oxide entails high environmental impact due to its energy-intensive nature. Therefore, the ability to recover silicon from EoL PVs becomes immensely beneficial for reintegrating it into various supply chains. Nonetheless, it's imperative for recovery technologies to have minimal environmental impact to ensure a sustainable approach to supplying this valuable material.

Directive 2012/19/EU (2012), transposed into Italian law by Legislative Decree 49/2014 and subsequent amendments, introduced EoL PVs into the category of waste electrical and electronic equipment (WEEE) for the first time. It mandates the recovery of materials composing these modules up to 80% by weight. This recovery target can be met for c-Si PVs by recovering glass and aluminum frames ex-

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clusively. In fact, silicon PVs are typically formed by the silicon cell embedded between the upper glass (on the front side) and a polymeric multilayer foil called backsheets (on the backside). Both the glass and the backsheets perform the function of protecting the cell from environmental factors such as rain, wind, hail and dust. They are glued to the cell by means of the encapsulant material which is generally Ethylene Vinyl Acetate (EVA). The cell is then “encapsulated” between the two layers of EVA, which form a protective and insulating sheet around it and acts as a binder between the cell itself and the upper (glass) and lower (polymeric) layers of the PV panel. The whole stack is embedded in the aluminum frame. Therefore, a photovoltaic module is typically made of glass for approximately 70% of its weight, aluminum for 15%, silicon for 3% and other metals (<1%), including silver and copper, used to make electrical contacts and in pastes soldering.

One of the most adopted EoL PVs pre-treatment process, before recycling, consists in the hammer milling of PV panels, once the aluminum frame and the cables/junction box have been removed. The outputs of this first step consist of three different materials: a fraction of vitreous fragments, EVA agglomerates, and splinters containing the active material (silicon) embedded within polymeric layers, besides silicon powder, aluminum, and copper bits. This fraction contains precious resources of potentially recyclable materials such as polyethylene terephthalate (PET), silicon, and metals. Therefore, it would be desirable to develop recycling processes useful to recover them.

The recycling methods currently adopted can be divided into two categories: thermal and mechanical. In both cases they can be followed by chemical (etching) and/or mechanical treatments for the possible recovery of other materials (silicon, copper, silver, etc.). For the recovery of metals and silicon with high purity, a subsequent hydrometallurgical treatment is required.

In conventional heat treatments, heating is carried out between 500 and 600°C, usually inside furnaces with a controlled atmosphere, for complete removal of the layers of EVA and the backsheets. It is worth mentioning that there are different types of backsheets on the market, some of which are based on fluorinated polymers which, due to their particularly toxic fluorine content, impose constraints on the methods of treatment. The plants that should burn the fluorinated backsheets, to obtain the silicon cells as residue, must be equipped with systems for controlling and reducing fluorine in the fumes. In detail, by burning fluorinated backsheets, hydrogen fluoride is produced (Ardenne, 2019, Danz et al. 2019, Fraunhofer, 2017), which can be subsequently removed from the gas phase via wet or dry systems where alkaline reagents are added to retain the halogens in solid form (e.g., CaF_2) (Aryan et al., 2018, Biganzoli et al., 2015, Vehlou, 2015). As an alternative to thermal treatment, PV panels could be delaminated to separate the silicon cell from the backsheets; then, recyclable PET could be recovered from the backsheets, while the silicon cell could be thermally treated to recover silicon powder by eliminating the encapsulant polymeric material.

Mechanical processes involve crushing, initially in a coarse manner and then finely, with the aim of reducing the

contact surface between the glass, EVA, and backsheets layers. This reduction facilitates detachment, which is followed by subsequent chemical and/or physical treatments such as sieving and gravimetric separation. The crushing processes demand significant energy expenditure, particularly as they yield satisfactory results (with a recovery rate of approximately 95%) only after undergoing at least two grinding stages—first coarse and then fine—to achieve the finest possible mesh size. However, these processes also come with a drawback: the hammers and knives utilized for crushing experience high levels of wear due to the inherent hardness of the glass and silicon composing the panels. Silicon dioxide (SiO_2), for instance, ranks at level 7 on the Mohs Scale, where diamond, the hardest material, is classified at level 10.

ENEA's research activities, concerning EoL PVs treatment and processing, are primarily aimed at providing technical solutions to reduce the environmental impact of commonly used recycling methods. Additionally, ENEA's focus extends to the valorization of materials recovered from EoL PVs, particularly silicon extracted from PV cells, which can be utilized as anode material in lithium-ion batteries. The main ongoing research projects involving ENEA, concerning EoL PVs recycle, will be hereby described and the key results obtained will be presented.

2. RECENT RESEARCH PROJECTS INVOLVING ENEA

2.1 Mission Innovation IEMAP

Italy is a member country of Mission Innovation (MI), a global multilateral cooperation initiative born in Paris in 2015 whose primary aim is to accelerate the innovation processes of clean technologies, both in the public and private sectors, through the commitment of the participating countries to double the public share of investments dedicated to research, development and innovation of decarbonisation technologies in order to make clean energy accessible to consumers and create green jobs and commercial opportunities. Within Mission Innovation, the IEMAP (Italian Energy Materials Acceleration Platform) project, financed by MASE for the years 2022-2024, intends to create an experimental and computational infrastructure for the accelerated design and selection of advanced materials for energy. As part of the MI IEMAP Project, ENEA is implementing new methodologies with low environmental impact for the treatment of EoL PVs and hydrometallurgical processes for the recovery of valuable materials (WP4, LA 4.4, 4.5 and 4.6). Moreover, a mechanical process for the recovery of silicon powder and the reuse of the recovered silicon in lithium-ion batteries has been optimized (WP2, LA 2.5, 2.7 and 2.9).

2.1.1 The thermomechanical delamination of the PV panel

The methodology developed by ENEA, that is proposed as an alternative to thermal and mechanical processes, is based on the ENEA-BetaTech patent N.102017000033488 (Italian Patent and Trademark Office, UIBM). The process consists of a method for recovering the main components (glass, EVA, backsheets, cells, electrical contacts) of the c-Si

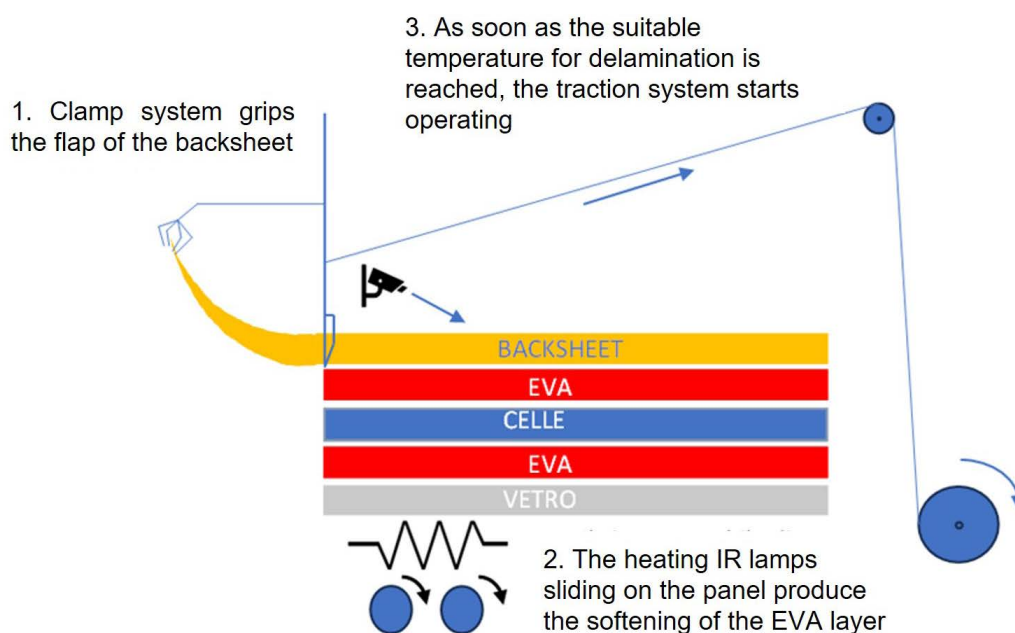


FIGURE 1: Principles of operation of the PROTEO Prototypal System at the ENEA Casaccia Research Center, Rome.

PVs through a heat treatment inducing the partial thermal degradation of the EVA layer, allowing separation of the different components. Thanks to the avoided degradation of the entire EVA and backsheet layers, as in other methods based on thermal treatments, gaseous emissions into the atmosphere are much lower. Compared to the other methods currently used, it has the advantage of being cheaper, as it is less energy-consuming, it has a low environmental impact and finally it allows recovering backsheet and EVA.

Within MI-IEMAP the Technology Readiness Level (TRL) of the process has increased to 4-5 since a prototypal plant, based on the patented technology, has been realized (Figure 1). Therefore, the process parameters (heat source, exposure time, distance between the heat source and the surface of the panel) have been tested on two types of panels on the market. To this aim it has been necessary to recognize the structure and chemical composition of the panels. It is worth mentioning that EoL PVs do not present any label indicating their structural composition. Therefore, the EoL panels have been characterized before the treatment

through several characterization techniques (FTIR, Raman, XRD and XRF spectroscopy).

2.1.2 The recovery of metals through hydrometallurgical processes

Metals are found in various components of the PVs, including the aluminum frame, screws, pins, junction box, cells, and copper ribbons. For the aluminum frame, screws, pins, and junction box, once these materials are selectively separated, they can be sent for recycling through established recovery chains. However, for the cells and ribbons, the metals within the PV panel must be extracted and purified. To ensure the representativeness of the sample and the scalability of the process across the entire category, two distinct types of PVs were selected, differing significantly in size, weight, production year, and technological characteristics. The first panel (PV type A) belongs to an older generation, with a production history of over 30 years, while the second panel (PV type B) is from a newer generation, manufactured in 2016. Table 1 presents the percent-

TABLE 1: Percentage composition of the examined panels.

	PV type A	PV type A	PV type B	PV type B
	(g)	(% p/p)	(g)	(% p/p)
Aluminum frame	1400	31	3500	17,5
Glass	1730	38,4	12250	61,3
Backsheet	80	1,8	640	3,2
PV cells	290	6,4	800	4
Ribbons	45	0,01	80	0,4
Screws and pins	10	<0,01	10	<0,01
Junction box	230	5,1	400	2
Silicone	15	<0,01	40	0,01
EVA	700 calculated by difference	16	2280 calculated by difference	11,3

age composition of these two panels. Notably, the composition of the two PV types varies significantly for each component fraction. When comparing the older generation device (PV type A) to the more recent one (PV type B), the weight percentage of the fraction containing materials of higher economic value (PV cells) decreases, while the percentage of glass increases. The glass fraction, once separated, can be recycled, whereas the fractions containing metals and plastics require treatment for valorization.

Chemical characterization of the cells and ribbons conducted using the Agilent MP-AES 4100 spectrometer (Table 2) has revealed that silver (Ag) is the most abundant and valuable material within the cells, reaching up to 8% by weight, while copper (Cu) accounts for approximately 80% of the ribbons, with tin (Sn) comprising 28%.

The separation of these elements has involved the following steps:

- Leaching;
- Fractional precipitation;
- Separation and purification of products.

In Figure 2, the diagrams depicting the processes for recovering Ag from PV cells (a) and Cu from ribbons (b) through hydrometallurgical methods are presented.

Subsequently, the recovered materials have been characterized using various analytical techniques. Cu and Ag have been obtained as products with defined chemical specifications and purity levels. Specifically, the two products obtained are:

1. CuC_2O_4 (Copper Oxalate): Purity grade: >96%. Usage: This high-purity copper compound finds applications in various fields. It is primarily utilized in laboratory settings for analytical and synthetic purposes. CuC_2O_4 serves as a reagent in qualitative and quantitative analysis, particularly in identifying the presence of certain ions in solution, such as calcium, barium, and strontium, by forming insoluble precipitates. Additionally, it acts as a precursor for synthesizing other copper compounds. For instance, it can be decomposed to yield copper oxide (CuO) or reduced to produce metallic

copper. These compounds have several applications including catalysts, pigments, and electrical conductors.

2. AgBr (Silver Bromide): Purity grade: >96%. Usage: Silver bromide is a photosensitive material primarily employed in traditional photography and certain specialized printing techniques. Its light-sensitive nature makes it suitable for creating photographic images on photosensitive films and papers. While its use has declined with the rise of digital photography, it still holds significance in specific segments of analog photography and specialized printing processes.

Data concerning the characterization of primary materials present in EoL PVs, as well as information regarding the extracted secondary materials, have been submitted to the IEMAP platform. This facilitates the creation of a database containing the chemical and microstructural properties of the secondary extracted raw materials related to the initial PV type and treatment parameters.

2.1.3 The reuse of silicon for lithium-ion batteries

Within MI-IEMAP, ENEA is also working on the reuse of silicon-based powder recovered from EoL PVs as anode material for lithium-ion batteries (LIBs). The recovered silicon will replace graphite, currently used to make the anode of commercial LIBs. It is worth mentioning that natural graphite is a strategic material imported for the 40% from China (CRM, 2023). The objective of the research activity is to obtain high-capacity anodes for LIBs with higher energy densities than commercial ones. Silicon offers a theoretical specific capacity of 4200 mAh g^{-1} (Huggins, 1999), considerably higher than the theoretical specific capacity of graphite (372 mAh g^{-1}). The limiting factor of silicon, used as anodic material, is related to its volumetric expansion which can reach values up to 300% during battery charge-discharge cycles (Rahman et al, 2021). Such a high volumetric variation involves progressive fragmentation and loss of active material (which is electrically isolated from the rest of the electrode) with the progress of charge-discharge cycles, resulting in a rapid decrease of the accumulated capacity.

TABLE 2: Chemical characterization of ribbons and cells for the two analyzed PVs (Type A and B).

Element	% (w/w) Ribbons in PV type A	% (w/w) Ribbons in PV type B	% (w/w) Cells in PV type A	% (w/w) Cells in PV type B
Na	<0,03	<0,03	0,03	0,03
Al	<0,03	<0,03	0,13	8,85
Cu	57,5	77	0,12	1,88
Ca	<0,03	<0,03	<0,01	0,08
Ag	0,7	1,4	7,9	1,8
Sn	27,7	13	0,08	0,08
Pb	13,2	7	0,4	0,33
Ni	<0,03	<0,03	0,06	<0,01
Fe	<0,03	<0,03	<0,01	0,02
Si, Ti, V, Cr, Mn, Co, Pd, Zn, Cd, Ba, Au	<0,03	<0,03	<0,01	<0,03
Residue	no	no	90	86

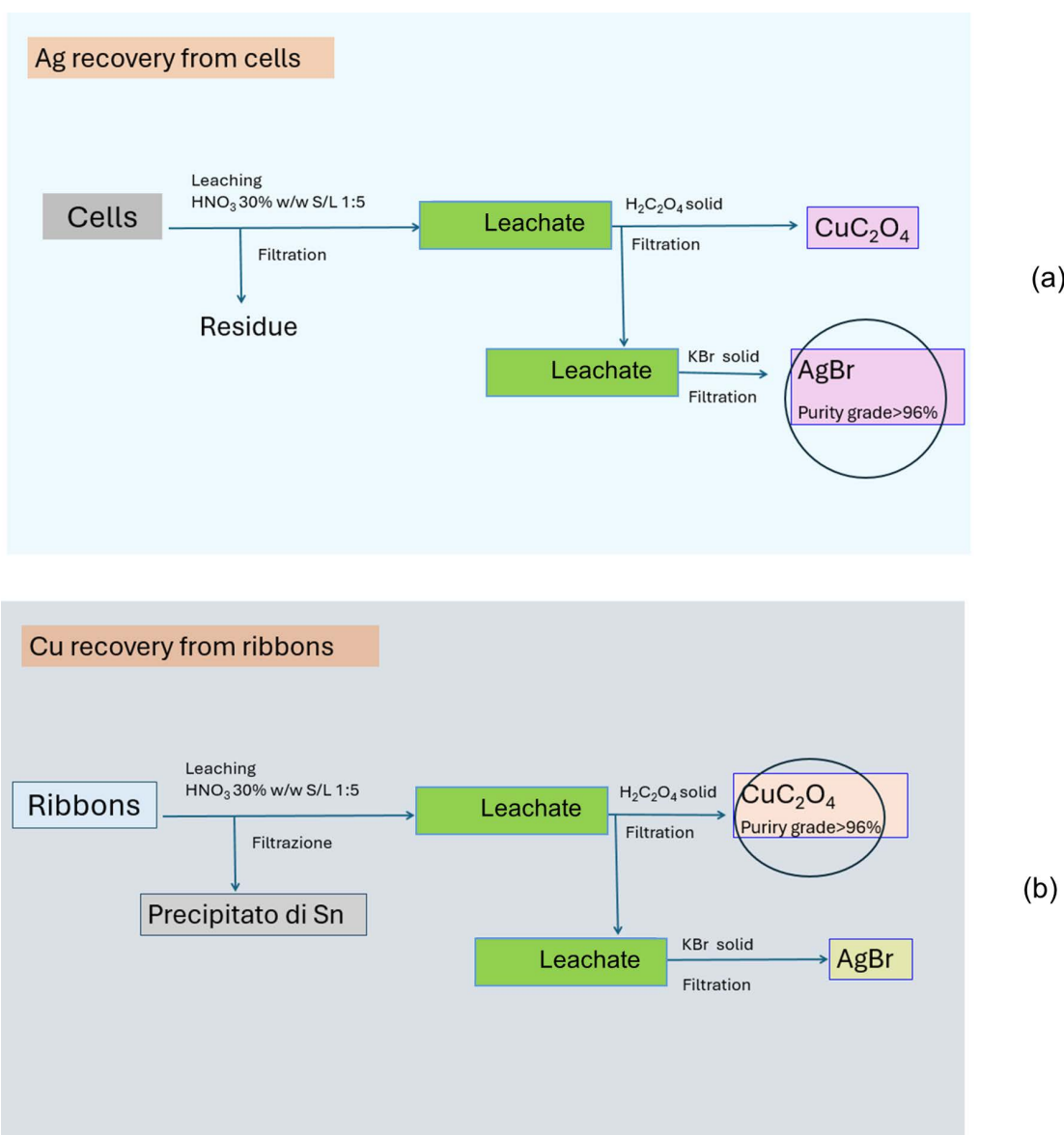


FIGURE 2: Processes for the recovery of Ag from PV cells (a) and Cu from ribbons (b) by hydrometallurgical methods.

The process illustrated in Figure 3, based on the ENEA patent N.102023000018996 deposited on September 15, 2023 (Italian Patent and Trademark Office, UIBM), has been employed in order to recover silicon-based powder suitable to be reused as anode material for LIBs.

The innovative process, which consists of an appropriately studied sequence of grinding and sieving steps, aims to eliminate the oxidized components with low electrical conductivity and to reduce the silicon powder to sub-micrometric dimensions to obtain, in the electrode manufacturing phase, a high surface area and low volumetric expansion of the anode material during cell operation. The powder was then characterized with different diagnostic techniques and was mixed with an electronic conductor (nano-carbon) and a polymeric binder, with the aim of guaranteeing electronic conduction and mechanical stability to the electrode, respectively. This mixture was dispersed in water and deposited on a copper sheet (having the func-

tion of a current carrier). After removal of the solvent and drying, the electrode was electrochemically characterized (voltammetry, impedance, and charge/discharge cycles in electrochemical cells) to validate its performance. The electrochemical tests, carried out preliminarily, have shown that silicon, obtained through recycling processes of PVs without resorting to chemical purification methods, can form alloys with lithium and can therefore potentially be used to create high-voltage anodes.

At present the invention is at TRL 4 (technology validated in the laboratory). Further studies are necessary to improve the electrochemical performance of the battery obtained, especially in terms of life cycles and capacity decay.

2.2 The PARSIVAL Project

The PARSIVAL (PV Panels Recycling to create Silicon VALue chain) project, funded by the KIC EIT RawMaterials

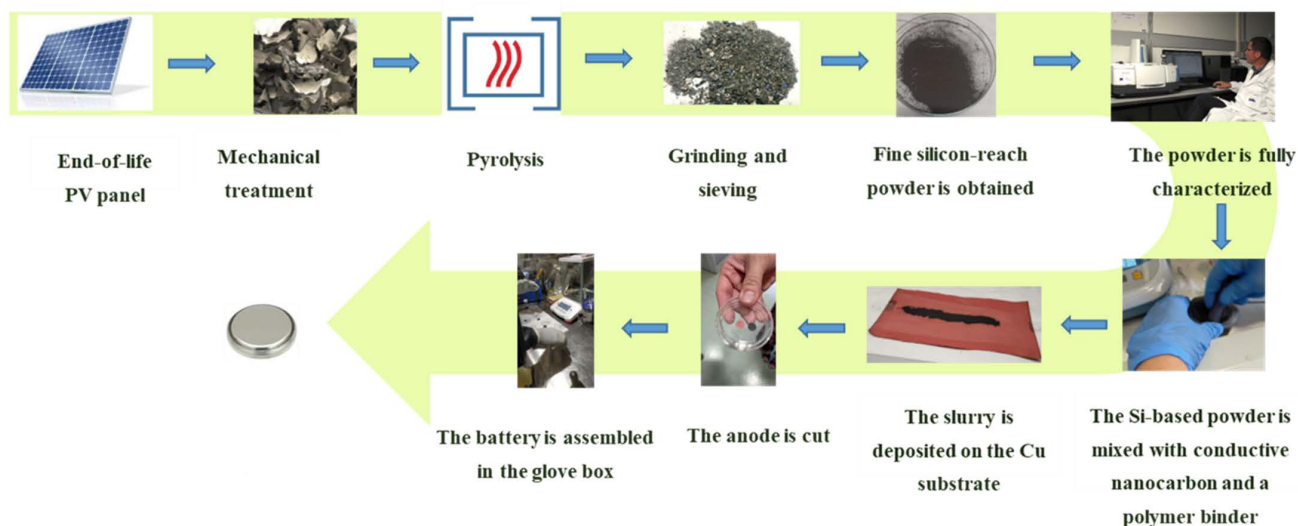


FIGURE 3: The ENEA process, based on the ENEA patent N.102023000018996 (Italian Patent and Trademark Office, UIBM), employed to recover a silicon-based powder suitable to be reused as anode material for LIBs.

for the years 2023-2024, aims to leverage the knowledge triangle and establish a European network focused on the challenge of recovering and reusing materials from EoL PVs, particularly silicon. The goal is to lay the groundwork for the development of facilities dedicated to refurbishing and recycling decommissioned PVs. This initiative seeks to extend the lifespan of repairable PVs and repurpose materials, recovered from PVs that are no longer viable for use, in various supply chains.

Specifically, the project will explore three different applications for silicon:

- Silicon-based ferroalloys,
- Material for battery anodes,
- Production of new photovoltaic cells.

PARSIVAL operates as a Regional Innovation Scheme (RIS) capacity building project, targeting territorial challenges associated with raw material extraction, recycling, and reuse within RIS areas. These areas typically comprise countries with moderate or modest innovation scores as defined by the European Innovation Scoreboard.

The project's activities are concentrated in two European regions situated within RIS areas that are experiencing emerging issues with EoL PVs disposal: Apulia, in Italy, and Extremadura, in Spain. Apulia ranks as the second largest Italian region in terms of installed photovoltaic capacity (3.05 GW). Given that PVs typically have a lifespan of around 25 years and the surge in photovoltaic installations occurred predominantly in the 2000s, a significant volume of decommissioned solar panels is anticipated to require management in the coming years. Approximately 300,000 tons of decommissioned solar panels are projected to accumulate in Apulia within the next 15 years.

Presently, there are no specialized treatment facilities for handling this type of waste in either Apulia or Extremadura.

Nine partners from four European countries (Italy, France, Germany, and Spain) are involved in the Project, each with their own designated tasks:

- 9Tech: coordination, feasibility study to implement its own recycling process in the Apulia Region, supply of recovered silicon/aluminum cells to study different applications.
- ENEA: development of a treatment process to make the recovered silicon suitable for application in lithium-ion battery anodes, communication and dissemination activities.
- ERION: analysis of PV installations in Apulia and forecasting of PV waste streams, contribution to the feasibility study for the implementation of a refurbishment/recycling plant.
- University of Padova: development of a treatment process to produce silicon-based ferroalloys from recovered photovoltaic cells, scientific education and dissemination activities.
- CEA: support for the feasibility study for the implementation of a photovoltaic panel treatment plant in Apulia region to repair and reuse EoL modules.
- JPM Silicon: validation of purification processes to make the recovered silicon suitable for different applications, in particular photovoltaics.
- University of Salento: high education and dissemination initiatives on recycling in Apulia.
- Gomez Pardo Foundation: Study for the implementation of project results in Extremadura (Spain).
- NORCSI: realization and validation of anodes and batteries using silicon recovered from EoL PVs.

2.3 The ReSiELP Project

The purpose of the ReSiELP (Recovery of Silicon and other materials from EoL PVs) project, funded with 2.5 million euros under the Knowledge Innovation Community on Raw Materials (KIC Raw Materials), was to optimize the recycling of EoL c-Si PVs, for efficient resources utili-

zation and waste reduction, in accordance with European legislation 2012/19/EU (also known as the WEEE Directive) and the principles of the circular economy. ReSiELP was a three-year project, implemented between 2017 and 2020.

Specifically, ReSiELP aimed to develop a recycling method for recovering not only the major components of c-Si PVs, like glass and aluminum (from the frame), but also the minor components that are precious and critical elements, such as silicon, silver, and copper (Audoin et al., 2019; Cerchier et al., 2022). Indeed, silicon is currently classified as a critical raw material, and silver is very close to reaching critical status. The first phase of the ReSiELP recycling method concerns the separation and recovery of glass, aluminum (frame), copper, and PV cells, by combining mechanical and thermal treatments. In detail, PV panels, after removal of aluminum frame and junction box by manual disassembling, undergo a thermal treatment, in order to eliminate the polymeric fraction (EVA encapsulant and backsheet). Afterwards, copper electrical ribbons are manually collected, while a separating machine (equipped with a vibrating table, a cooling fan for mechanical air separation, and a 3 mm mesh net) is used to separate glass and silicon cells. Copper is sent to recyclers whereas the recovered glass is reused as inert material in prefabricated concrete elements, to promote sustainable construction and reduce the depletion of non-renewable natural resources. As for the silicon cells, after removing glass contamination, they undergo a hydrometallurgical treatment, consisting of two phases. In the first phase, sodium hydroxide is used to remove aluminum from the backside of the silicon cells, while the second phase utilizes nitric acid to detach silver electrical contacts from the front side of the cells. Silver is hence recovered through precipitation as silver chloride which is reduced to metallic silver (99.9% purity), through a heat treatment, in a muffle, containing sodium carbonate. As for the silicon from hydrometallurgical treatment, it goes through further basic and acid chemical treatments, mainly based on the use of sodium hydroxide and tartaric acid, to reach a purity of 6N (99.9999% silicon) and become suitable for reuse in the PV sector.

Within the project, ENEA contributed to the development of the management system for gaseous effluents resulting from the PV thermal treatment and the design of the process for the neutralization of the wastewater from PV cells hydrometallurgical treatment (Cerchier et al. 2022). Additionally, ENEA assessed the sustainability of every process phase through the Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) methodologies (Ansanelli et al., 2021; Fiorentino et al., 2022). The LCA analyses indicated that, overall, the ReSiELP recycling process brings environmental advantages, thanks to the retrieval of secondary materials from EoL c-Si PVs. Similarly, the LCC analyses showed that, due to the recovery of secondary materials, the ReSiELP recycling process achieves a net reduction in environmental damage costs (externalities).

2.4 The EVERPV Project

The EVERPV project (Highly efficient delamination technologies to recover and reuse metals, glass, polymers from EoL PVs), funded under the Horizon Europe program

(HORIZON-CL5-2022-D3-03-09), is dedicated to the development of recycling solutions for EoL c-Si PVs, in a circular economy perspective and in compliance with the European WEEE Directive. It is a 36-months project, started in 2023, which aims to develop a technology that is both efficient and environmentally, economically, and socially sustainable.

Specifically, within EVERPV project, two delamination processes will be developed up to TRL 7 (pilot plant), one utilizing grinding and the other based on the ENEA-BetaTech patent N.102017000033488 (Italian Patent and Trademark Office, UIBM), exploiting infrared (IR) radiation technology to weaken the encapsulant. A third hybrid process will also be investigated, involving the thermal treatment of c-Si PVs, after removing backsheet through grinding or by exploiting the IR radiation.

In the EVERPV project, ENEA will define the optimal conditions of the hydrometallurgical treatment for Ag, Cu and Sn recovery. Moreover, ENEA researchers will develop the technical specifications for implementing the "IR delamination methodology" at TRL7.

Finally, ENEA, in collaboration with other project Partners, will assess the economic and environmental performance of the delamination and purification processes, studied by EVERPV, through Life Cycle Costing (LCC) and Life Cycle Assessment (LCA) methodologies, in compliance with UNI EN ISO standards 14040-14044 (2006). The objective is to identify the economically and environmentally critical points enabling targeted improvement interventions and supporting the development of sustainable recovery technologies. Furthermore, the environmental and economic impacts of the two investigated delamination processes (grinding and IR), scaled up TRL7, will be compared with those of an established and innovative technology, involving the use of hot knives. This comparison aims to promote technologies that are better, from an economic and environmental point of view, than those currently available. Additionally, the environmental impacts and the economic costs associated with the processes, set up within EVERPV, for recovering and purifying materials from EoL c-Si PVs, will be assessed and compared with those related to the production of corresponding virgin materials.

Lastly, in line with guidelines developed by the Life Cycle Initiative and the United Nations Environment Programme, the social impacts of the recycling processes, developed through the EVERPV project, on various stakeholder groups, will be assessed, considering factors such as working conditions, job creation or loss, and the need for specialized skills.

Any positive results will be very valuable for fostering material recovery from EoL c-Si PVs, at both entrepreneurial and political levels.

3. CONCLUSIONS

ENEA research activity, dedicated to the challenging issue of EoL PVs recycle, is mainly focused to provide solutions to some critical issues limiting the sustainable recovery and valorization of materials from EoL PVs. Within RESIELP project (2017-2020), ENEA contributed to the

development of a management system for the gaseous effluents resulting from the thermal treatment and design of the process for the neutralization of the wastewater from PV cells hydrometallurgical treatment. Additionally, ENEA conducted an environmental assessment of every process phase using the LCA methodology. Within MI-EMAP (2022-2024), ENEA is now developing a process, based on the ENEA-BetaTech patent N.102017000033488 (Italian Patent and Trademark Office, UIBM) allowing the recovery of the main components of PVs (glass, EVA, backsheet, cells, electrical contacts) through an IR heat treatment allowing the partial thermal degradation of the EVA layer. Avoiding the degradation of the entire EVA and backsheet layers, as other methods based on thermal treatments do, gaseous emissions into the atmosphere are reduced. The methodology will be brought to TRL 7 within EVERPV Project and will be compared (and eventually integrated) with grinding technology. Moreover, within MI-EMAP, ENEA is investigating the possibility to employ silicon recovered from EoL PVs in lithium-ion batteries through a mechanical process based on the ENEA patent N.102023000018996 (Italian Patent and Trademark Office, UIBM). Within PARSIVAL capacity building project, ENEA is finding out technical solutions for the treatment of silicon cells recovered by 9-Tech plant, in order to obtain a suitable silicon-based anode material for lithium-ion batteries.

ACKNOWLEDGEMENTS

ReSIELP activities (2017-2020) have received funding from the European Institute of Innovation and Technology (EIT) under the Horizon 2020, the EU Framework Programme for Research and Innovation. In particular, the project was supported by the Knowledge and Innovation Communities Raw Materials (KIC RM) in the category: Recycling and material chain optimization for EoL products. Project website: www.resielp.eu.

Project MI-EMAP is funded by MASE for the years 2022-2024, while PARSIVAL is funded by EIT RawMaterials for the years 2023-2024. EVERPV project is funded by the European Union under the Horizon Europe program (HORIZON-CL5-2022-D3-03-09).

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LI-ION BATTERY RECYCLING AT PILOT SCALE: DEMONSTRATING THE RESYNTHESIS ROUTE ALSO FOR NMC-LFP MIXED BLACK MASS

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Article Info:

Received:
23 April 2024
Revised:
20 June 2024
Accepted:
12 July 2024
Available online:
17 September 2024

Keywords:

Li-ion batteries
Recycling
Pilot plant
Re-synthesis
NMC precursors
LFP

ABSTRACT

The hydrometallurgical recycling of end-of-life Li-ion batteries constitutes a sustainable path for the integral recovery of battery components. However, the heterogeneity of electrode materials constitutes a limitation for process optimization that must provide robust and flexible strategies. In this work, the preliminary results obtained as part of the demonstration activities of the LIFE DRONE project are reported with particular attention to the possibility of integrating the recycling process based on the resynthesis of NMC (Nickel Manganese Cobalt) cathodes with the treatment of mixed black mass also containing LFP (Lithium Ferrous Phosphate) electrodic powders. The experimental results obtained highlight the possibility of leaching NMC black mass in the absence of hydrogen peroxide when NMC-LFP mixtures are used with almost quantitative extraction yields of metals. In particular, working without H₂O₂ at 60°C for 3 h the following yields of extraction were obtained: 89% for Ni, 69% for Mn, 89% for Co, 91% for Li, 90% for Fe, 100% for P, 56% for Al and 32% for Cu. Furthermore, the experimental data showed that by varying the pH of the leach liquor, it is possible to separate selectively iron phosphate by quantitative precipitation (100% for Fe, 100% for P and 60% for Al) before proceeding with NMC resynthesis.

1. INTRODUCTION

The recycling of Li-ion batteries constitutes a fundamental step in the green transition process, both for the recovery of material resources (secondary raw materials) and for preventing the release of toxic metals into the environment. Recent updates to European regulations include Li-ion batteries as a new target type for which specific recycling rate requirements must be ensured (EU Regulation 2023/1542). The new Regulation opens the way to hydrometallurgical processes with significantly lower environmental impacts than conventional pyrometallurgical ones (Domingues et al., 2023). Nevertheless, hydrometallurgical processes still present various challenges in terms of safety and economic sustainability, leaving the door open for technical innovations that can simplify the recycling process. However, this need clashes with the structural and chemical complexity of Li-ion batteries

The automotive and electronics markets are predominantly characterized by NMC (Nickel Manganese Cobalt) batteries, which feature cathodes in the form of lithium

transition metal oxides, such as nickel, manganese, and cobalt with various stoichiometries, while heavy-duty vehicles and stationary storage systems are characterized by Li-ion batteries with lithium iron phosphate (LFP) cathode active materials. However, this classification is vague as materials such as LFP can also be mixed within NMC batteries to increase thermal stability and therefore battery safety. There are several technical challenges in recycling Li-ion batteries.

The safe opening of Li-ion batteries requires measures to prevent explosion due to the presence of flammable solvents in the electrolytes. Furthermore, the evolution of cathodic material compositions, such as transition metal oxides, has led to materials entering the market (and thus into waste streams to be fed into recycling plants) with highly variable contents of nickel, manganese, and cobalt. The original recycling approach relied on downstream procedures typical of hydrometallurgy (chemical precipitation and solvent extraction) to remove metallic impurities (aluminium, iron, copper) and to separate manganese and



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nickel from cobalt, which was the target metal of an initial generation of Li-ion batteries predominantly based on Li-CoO₂ (Pagnanelli et al., 2016; Abo Atia et al., 2019). The limited availability and high price of cobalt have driven the evolution of oxide chemistry towards stoichiometries with lower cobalt content, and therefore recycling processes cannot avoid recovering all the different types of metals present in batteries (nickel, manganese, cobalt, and lithium). The approach of recovering metal salts separately (via solvent extraction) incurs significant investment and operating costs that compromise the economic sustainability of the recycling process, whose potential is limited by end-of-life battery collection. An alternative and simpler approach from the downstream operations perspective is re-synthesis, in which the metals present in the purified leachate, free from metallic impurities, are directly recovered by producing mixed precursors (generally hydroxides) with metal stoichiometry that can be adjusted and corrected by adding salts (Schiavi et al., 2021).

Further complications arise from the lack of automated sorting systems for Li-ion batteries based on their chemistry (e.g., NMC and LFP) and the mixing of different cathode materials within the same batteries or in electric vehicle battery packs (such as various stoichiometries of NMC or NMC-LFP). This requires additional flexibility in hydrometallurgical processes to handle mixed cathodic materials. In the literature, studies on LFP batteries are mainly focused on direct recycling processes (without cathode dissolution) due to the low intrinsic value of these materials (Wang et al., 2022; Xu et al., 2024). However, for mixtures of NMC-LFP materials, the direct recycling strategy would necessitate a preliminary separation of the different cathodic materials, which has not yet been achieved. This implies that, to treat mixed NMC-LFP black masses, it will be necessary to dissolve both materials for their recovery.

Some hydrometallurgical recycling works of LFP batteries have been reported in the literature, typically utilizing an acid (sulfuric acid) to obtain phosphates, iron, and lithium in solution (Zheng et al., 2016). To keep iron in solution, it's necessary to prevent its oxidation, as it would otherwise precipitate as iron phosphate (Gerold et al., 2022). Alternatively, it is possible to work in the presence of an oxidizing agent (in the case of iron, hydrogen peroxide can be used), promoting the selective dissolution of lithium and recovering FePO₄ in the solid phase along with graphite (Li et al., 2017).

Recent works are addressing the problem of treating this mixture of black mass proposing a selective leaching of other metals except Fe, and then performing an addi-

tional leaching of the solid residue to recover Fe (Song et al., 2023).

In this general context, this work reports the first results at pilot scale of cathodic resynthesis from NMC black mass and also the results of simultaneous leaching of NMC-LFP black mass under the transferability activity foreseen in the LIFE DRONE project (LIFE DRONE project) for the extension to other types of wastes of the developed process route of NMC re-synthesis. The process in comparison with previously reported consider the simultaneous extraction of all metals and the following selective recovery of graphite, NMC precursors, FePO₄ and Li₂CO₃.

2. MATERIALS AND METHODS

2.1 Integrated process for recycling of Li-ion batteries and production of NMC cathodes

The process under validation at pilot scale within the activities of the LIFE DRONE project, includes the shredding of end of life (EoL) Li-ion batteries and the separation of the black mass from other recyclable fractions (steel, aluminium and copper), then the hydrometallurgical section for black mass dissolution and precursors resynthesis (as NMC hydroxides), and finally the production of lithiated mixed oxides as cathodic materials for new batteries. In Figure 1 the overall diagram of the integrated process is reported.

2.1.1 Main process operations at pilot scale

The integrated process for cathodic material production from black mass through resynthesis route was performed at pilot scale in different operative sections (Figure 2).

Dismantling of Li-ion batteries have been performed using the pilot plant designed and constructed during the LIBAT project (LIFE LIBAT project) dedicated to the recycling of Li(0) primary batteries. The process includes a cryo-mechanical crushing, dry separation (sieving and magnetic) and final bath in water (Pagnanelli et al., 2023). The cryo-mechanical section (400 kg/day potentiality) included a cryogenic pretreatment performed on the batteries in order to prevent explosions and control flame formation during crushing. Thermally stabilized batteries (−80°C for 45 min using a N₂ liquid shower in a cabinet) were then crushed in a hammer mill with 10 mm under-sieve. Crushed material was sieved (1 mm) and the over-sieve was separated by magnetic separation using a magnetic over-belt placed above the sieving belt. After this treatment, three

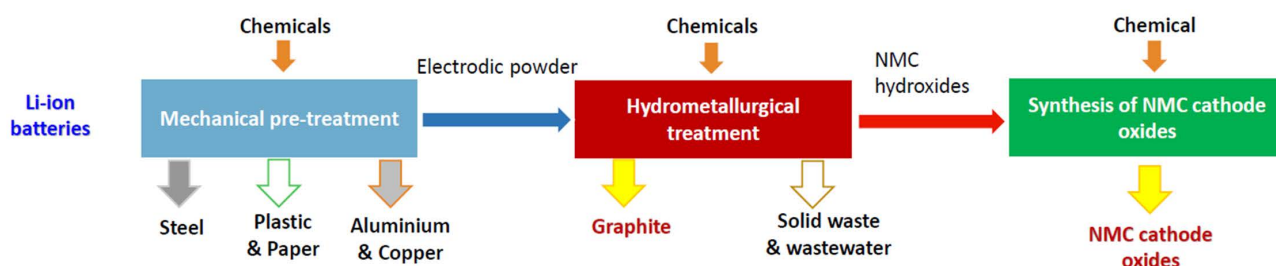


FIGURE 1: Schematic representation of the different sections of the recycling process.



FIGURE 2: Sections of the pilot plants: the cryo-mechanical section (a), the hydrometallurgical section for resynthesis (b), and the cathodic material production section (c).

fractions were collected: fine powder made up of electrode materials, a coarse magnetic fraction made up of external steel cases, and a non-magnetic coarse fraction made of large fragments of collectors and separators of plastic and papers. Copper and aluminium fragments from undersieve fraction and non magnetic oversieve fraction are separated by eddy currents.

The sequence of operations in the hydrometallurgical section was reported in Figure 3.

Black mass (30 kg per batch) was leached by using H_2SO_4 (18%ww, provided by Soave & Fontana) and adding 30%v H_2O_2 solution (provided by Soave & Fontana), with 1:10 kg/L solid to liquid ratio for 3 h under mechanical stirring at 60°C. Leach liquor was separated by filtration, purified with pH increase by NaOH (30%ww provided by Soave & Fontana) and then recovered by filtration. Pre-purified leach liquor was then corrected by metal (Ni, Mn, Co) sulphate (provided by Soave & Fontana) addition for obtaining the target stoichiometry (NMC111) and then used for mixed oxide precursor precipitation adapting a previous recipe (Schiavi et al., 2021): solution was added with NH_4OH (25%ww provided by Soave & Fontana) and NaOH (30%ww provided by Soave & Fontana) for pH correction (11), then kept under mechanical stirring at 60°C for 8 hours, periodically injecting N_2 in the reactor. Precursors were filtered and washed by water, dried in an oven at 100°C overnight, mixed with 10% stoichiometric excess of Li_2CO_3 (recovered from LIBAT process (Pagnanelli et al., 2023)) and thermally treated in a muffle with a two-stage ramp: 5 hours at 450°C and then 10 hours at 950°C.

2.2 Characterization of black mass and cathodic materials from pilot scale

NMC black mass was provided by S.E.Val. by performing cryo-mechanical treatment of EoL Li-ion batteries of NMC type according to the optimized process route developed in the LIBAT project (Pagnanelli et al., 2023), while LFP black mass is made up of cathode scraps furnished by FAAM. Cathodic materials are those obtained by pilot plant according to previously described route (Section 2.1.1).

The characterization of the incoming black mass and produced cathodic materials was performed by mineralization. The mineralizations were carried out by weighing 0.100 g of solid; these were placed in a 50 mL flask into which 4 mL of hydrochloric acid (VWR Chemicals, 37%ww), 4 mL of nitric acid (VWR Chemicals, 65%ww) were added, and 2 mL of hydrogen peroxide solution (Sigma Aldrich, 30%ww). The sample was subjected to magnetic stirring at a temperature of approximately 100°C for 3 hours. The mineralization operation just described was carried out 3 times in order to have 3 replicates for the chemical characterization of the sample. After 3 hours, the flasks are allowed to cool and brought to volume, to carry out the various dilutions starting from 1:10, 1:100, 1:1000; 1:5000. Finally, these dilutions are analyzed with the Inductively Coupled Plasma – Optical Emission Spectrophotometer, ICP-OES (Perkin Elmer, optical emission spectrometer Avio 220 Max) from which the different concentrations of the metals of interest are obtained.

Cathodic materials are characterised by Scanning Electron Microscopy (Zeiss Auriga FE-SEM).

2.3 Hydrometallurgical process for mixed NMC-LFP black mass at lab scale

The hydrometallurgical route for resynthesis of NMC cathodic materials is reported in Figure 3 putting in evidence the additional step for FePO_4 recovery after leaching. NMC-LFP mixed black mass (1:1 ratio in weight) were leached in thermostated jacketed vessels under magnetic stirring with 1.5 M sulfuric acid (VWR Chemicals, 95%ww) and 1/10 solid liquid ratio in different operating conditions of H_2O_2 dosage (0 and 15% as % volume added to the suspension of 30%ww H_2O_2 solution, Sigma Aldrich), temperature (60 and 85°C), and time (3 and 5 h). Iron phosphate recovery was performed adding H_2O_2 (up to 600 mV) and increasing pH by 1 M NaOH solution (prepared from Sigma Aldrich pellets).

3. RESULTS AND DISCUSSION

The pilot-scale demonstration activities for the resynthesis of cathode materials from end-of-life Li-ion batteries

are still in progress within the activities of the LIFE DRONE project, which is scheduled to conclude in June 2024.

The following results pertain to the initial campaigns for the resynthesis of precursors at pilot scale and the transferability activities, at laboratory-scale, evaluating the application of the LIFE DRONE process using mixed NMC-LFP black mass.

3.1 Resynthesis at pilot scale of cathodic materials from NMC black mass

The starting electrode powder fed to the leaching section (black mass from S.E.Val) has the composition reported in Table 1. LFP scraps were composed by LiFePO_4 particles with V coating according to the following composition: 39 ± 3 mg/g of Li, 330 ± 30 mg/g of Fe, 180 ± 20 mg/g of P, and 3.0 ± 0.5 of mg/g V. Considering the stoichiometric ratios among nickel, manganese and cobalt this powder is equivalent to an NMC325 cathode material (not existing on the market and probably due to contamination effects as the mechanical plant is used also for the treatment of other types of batteries such as Li primary batteries and Ni metal hydride batteries. Fe, Al, and Cu contents in terms of atomic percentage (%at) are 1.5%, 4.3%, and 2.3%, respectively.

TABLE 1: Chemical composition of NMC black mass input material and NMC cathodes obtained by resynthesis route.

Metal	Black mass (mg/g)	Cathodes (mg/g)
Li	40+/-3	50+/-20
Ni	96+/-6	200+/-30
Mn	77+/-2	150+/-20
Co	163+/-9	170+/-20
Fe	5.2+/-0.6	0.237+/-0.005
Al	7+/-2	5+/-3
Cu	9+/-2	9+/-2

NMC111 cathodes were obtained after correction of leach liquor composition by hydroxide precipitation and annealing in presence of Li salt. The stoichiometric coefficients of the NMC cathodes evaluated by chemical composition reported in Table 1 are 0.38, 0.30, and 0.32 for Ni, Mn, and Co, respectively, in line with the desired target stoichiometry (NMC111), while Li is under-stoichiometric (-17%). The atomic percentages of Fe, Al, and Cu are 0.04%, 2.1%, and 1.5%, respectively. The effect of such levels of metal impurities on electrochemical perfor-

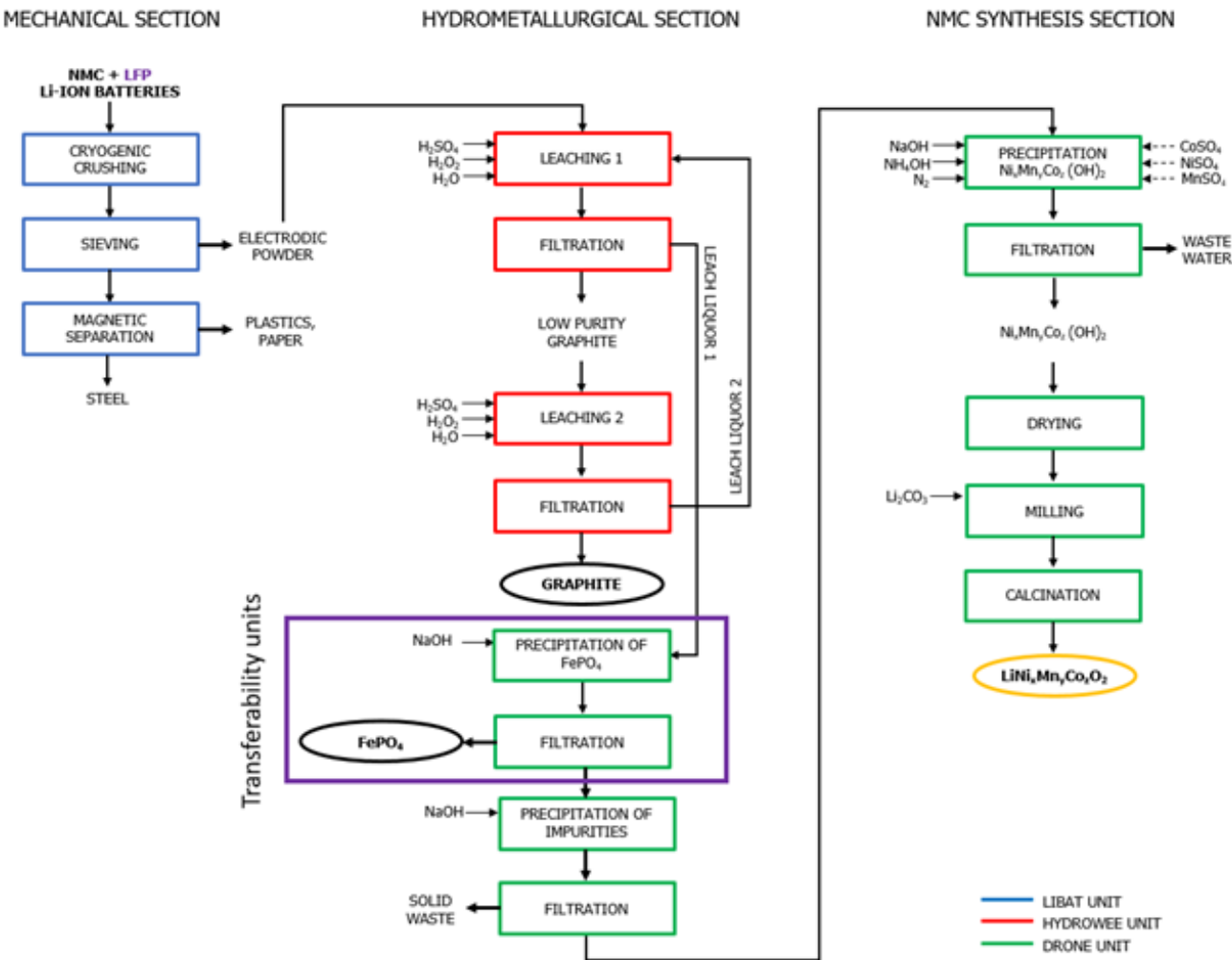


FIGURE 3: Process flow diagram for the integrated process of recycling of NMC-LFP black mass.

mances are still under evaluation. These metallic impurities present in the purified leach liquors are incorporated into the crystal lattice of the cathode material during coprecipitation. The presence of such metals can exhibit dopant effects below certain concentrations (Zhao et al., 2020; Sa et al., 2015), particularly for limit concentrations of 0.2% for aluminum (Zhang et al., 2020), 0.25% for iron (Park et al., 2018), and 1% for copper (Zhang et al., 2020). The simultaneous presence of different metal impurities requires further dedicated investigation as never reported in the literature.

SEM characterization of re-synthesized cathodic materials evidenced primary crystallites with size ranging from 200 to 300 nm, aggregating in larger structures (50-150 micron) as reported in Figure 4.

3.2 Transferability at lab-scale: leaching of mixed NMC-LFP black mass and FePO_4 recovery

Leaching of mixed NMC-LFP black mass by using the mildest conditions (60°C and 3h leaching) and without

H_2O_2 , enables achieving a metal extraction yield larger than to 90%, except for Mn and metal impurities (Al, Cu) as reported in Figure 5.

In particular, working without H_2O_2 at 60°C for 3 h the following yields of extraction were obtained: 89% for Ni, 69% for Mn, 89% for Co, 91% for Li, 90% for Fe, 100% for P, 56% for Al and 32% for Cu.

Average chemical composition of leach liquor is: 4400 ± 800 mg/L for Li, 7000 ± 500 mg/L for Ni, 7600 ± 800 mg/L for Mn, 3000 ± 300 mg/L for Co, 600 ± 200 mg/L for Cu, 16000 ± 1000 mg/L for Fe, 210 ± 50 mg/L for Al, and 7700 ± 900 mg/L for P.

FePO_4 recovery was performed by raising the pH of leach liquor to 2 provided the optimal option in terms of solid recovery (100% precipitation for Fe and P) and purity, as all metals except Al (60% precipitation) remain in solution at this pH. The presence of this impurity could be avoided by improving physical pretreatment (thus reducing initial amount of Al in black mass) or performing an initial basic leaching of the black mass.

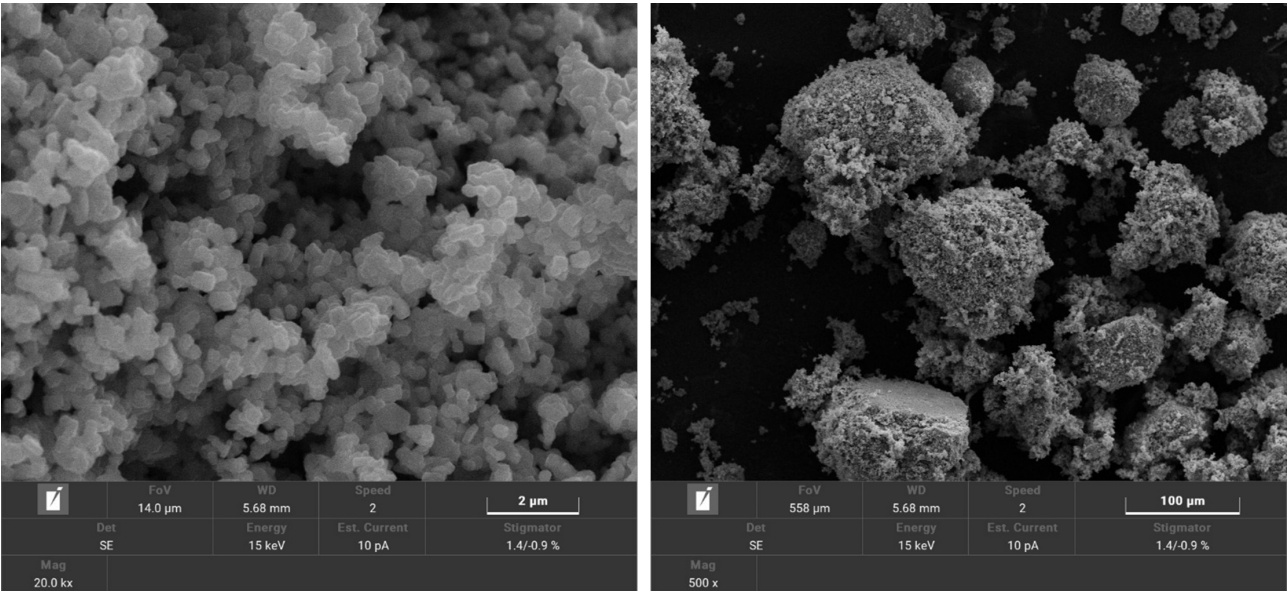


FIGURE 4: SEM characterization of re-synthesized cathodic materials at different magnifications.

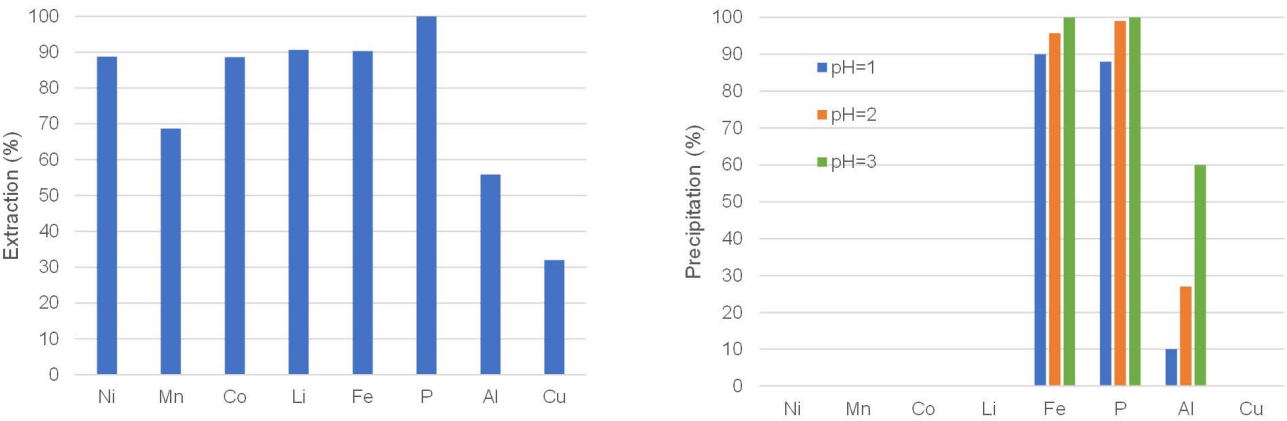


FIGURE 5: Metal extraction in NMC-LFP leaching (left); FePO_4 precipitation by pH increase (right).

4. CONCLUSIONS

The pilot-scale production activities of mixed cathodes through the resynthesis process are still ongoing, and the electrochemical performance of the obtained materials is also under evaluation, considering the presence of multiple metallic impurities resulting from the automated mechanical treatment process. In fact, currently, few literature information are available about the effect of metal impurities in the resynthesis route from EoL black mass. Electrochemical validation of produced materials could give new insight about the effect of simultaneous presence of different metallic impurities.

Even considering the necessary phases of process optimization, preliminary results have highlighted the effectiveness of the treatment regarding the ability to produce mixed cathodes with the desired stoichiometry starting from end-of-life Li-ion battery black mass with heterogeneous composition.

The lab-scale leaching tests of mixed NMC-LFP electrode powders have shown the possibility of extracting all solid metals quantitatively. Additionally, laboratory-scale tests have also highlighted the possibility of recovering iron before the precursor production process, allowing the treatment of mixed NMC-LFP black mass in the re-synthesis-based process.

ACKNOWLEDGEMENTS

This work was co-funded by the EU LIFE+ program within the project "LIFE DRONE project: Direct pROduction of New Electrode materials from battery recycling (ENV/IT/000520) www.lifedrone.eu.

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GEL POLYMER ELECTROLYTES ELABORATED BY UV-IRRADIATION FOR THE ECO-DESIGN OF LI-ION BATTERIES

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Article Info:

Received:
29 May 2024
Accepted:
13 July 2024
Available online:
7 September 2024

Keywords:

Gel
Electrolyte
Polymers
Li-ion batteries
UV irradiation
Poly(ethylene glycol) diacrylate

ABSTRACT

This study examines the chemical, thermal, and dielectric properties of gel polymer electrolytes (GPEs) based on poly(ethylene glycol) diacrylate (PEGDA, average $M_n=700$ g/mol). GPEs were synthesized via a single-step UV-irradiation process, whereby PEGDA was mixed with varying concentrations of a commercial liquid electrolyte (1 M LiPF₆ in EC/DEC 50/50 v/v) ranging from 0 to 60 wt%. The results of the infrared analysis indicated that the liquid electrolyte was successfully immobilized within the polymeric matrix. The thermally stable nature of the synthesized GPEs was confirmed across a wide temperature range (-40 to 90°C) through thermal analysis. Furthermore, dielectric measurements were conducted in a frequency range of 0.1 to 10⁶ Hz, with a sinusoidal AC amplitude of 10 mV, from 20 to 100°C. The results demonstrated that the ionic conductivity of GPEs increased with the addition of liquid electrolyte. The GPEs prepared with 60 wt% liquid electrolyte exhibited a conductivity value of approximately 6.6x10⁻³ S/m at 30°C, which is consistent with the conductivity range reported in the literature.

1. INTRODUCTION

The transition to renewable energies presents a significant challenge in the 21st century, particularly in the area of electrochemical energy storage (Arshad et al., 2020). Lithium-ion batteries (LIBs) have become a crucial component in various applications, including portable electronic devices, industrial equipment, medical devices, and electric vehicles (Grewal et al., 2022; Wang et al., 2021). These accumulators exhibit several advantages over conventional technologies, including lead acid, Ni-Cd, and Ni-metal hybrid batteries. They lack the memory effect, exhibit a low self-discharge rate, possess a high energy density, and can be charged rapidly. Moreover, they exhibit a relatively long lifetime (Raj et al., 2022). Nevertheless, energy storage systems with liquid electrolytes are associated with several disadvantages. These include the risk of explosion or fire, electrolyte leakage, and internal short circuits caused by the formation of lithium dendrites within the anode during charging and discharging (Yao et al., 2019). In response to these concerns, researchers have developed less hazardous alternatives over recent decades. These alternatives seek to replace or adapt the liquid electrolyte with solid materials, including polymers, ceramics, and composites (Maia et al., 2022).

Research on solid polymer electrolytes (SPEs) has primarily focused on polyethylene oxide (PEO) due to its low glass transition temperature ($T_g = \sim -60^\circ\text{C}$) and its ability to effectively complex with Li⁺ ions, despite its low dielectric constant (~ 5) (Mindemark et al., 2018; Q. Zhang et al., 2017). Other polymer families, including acrylates/methacrylates (Kurapati et al., 2019), polynitriles (Arof et al., 2014), fluoropolymers (J. Zhang et al., 2023), polycarbonates (Xu, 2004), and polyesters (Mindemark et al., 2018), have also been the subject of research. Nevertheless, their industrial application remains challenging due to the costly, energy-intensive, and highly sophisticated synthesis techniques required for SPEs. Polymer electrolytes may present certain drawbacks, including low ionic conductivity at room temperature, poor mechanical properties, and limited biodegradability (Zhou et al., 2019). Consequently, it is of paramount importance to develop novel polymer electrolytes that are capable of meeting the demands of LIB applications. The ideal polymer electrolytes would possess excellent mechanical and thermal properties, high ionic conductivity, and enhanced safety.

In recent years, gel polymer electrolytes (GPEs) have emerged as a promising alternative to liquid electrolytes, offering enhanced design versatility and preventing elec-

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trolyte leakage (Ren et al., 2021). GPEs trap conventional liquid electrolytes, which are composed of lithium salts dissolved in carbonated organic solvents, within a polymer matrix, thereby reducing the risk of leakage. It is notable that these GPEs serve the dual function of electrolyte and separator, insulating the positive and negative electrodes. This feature significantly reduces the risk of short-circuiting between the electrodes due to the formation of lithium dendrites.

The objective of this study is to examine GPEs, particularly those comprising poly(ethylene glycol) diacrylate (PEGDA), through a single-step synthesis process. The PEGDA will be subjected to UV irradiation in the presence of a photoinitiator. It is generally accepted that UV-light induced polymerization/crosslinking processes enhance the mechanical properties of a GPE. The incorporation of a conventional liquid electrolyte can markedly enhance the conductivity of the polymer electrolyte. The selection of PEGDA was based on its two reactive C=C end bonds and ether functions, which act as electron donors and enhance the ionic conductivity of the synthesized GPEs. Furthermore, the polymer utilized in the synthesis of these GPEs is non-toxic, biodegradable, and exhibits high mechanical strength with a low T_g (Liu et al., 2020). It is also noteworthy that the use of this non-toxic polymer could facilitate the recycling of LIBs.

2. MATERIALS AND METHODS

2.1 Materials

The materials utilized in this study include PEGDA (average $M_n=700$ g/mol) as a prepolymer, 2-hydroxy-2-methylpropiophenone (Darocur 1173) as a photoinitiator, and a liquid electrolyte (1M LiPF_6 in ethyl carbonate/diethyl carbonate (EC/DEC) 50/50 v/v). All of the aforementioned products were procured from Sigma-Aldrich and utilized in their original form.

2.2 Synthesis of gel polymer electrolytes

To elaborate the GPEs, PEGDA and Darocur 1173 (2 wt% related to PEGDA) were combined with varying concentrations of liquid electrolyte, ranging from 0 to 60 wt%. The mixing process was conducted within a glove box, which was maintained under an argon atmosphere, in order to prevent the electrolyte from being exposed to air, which can cause instability. The glove box was maintained at oxygen (O_2) and water (H_2O) levels of less than 0.1 parts per million (ppm). The mixtures were vigorously stirred for four hours, after which polymerization/

crosslinking was performed using the ultraviolet irradiation technique.

The liquid mixtures were placed on a Teflon mold in an argon atmosphere and exposed to 365 nm UV light for 30 min, as illustrated in Figure 1a. Following the UV curing process, the samples were placed in an oven maintained at 40°C and 10^{-2} mbar.

The resulting materials are designated as P-X% S, where X represents the weight fraction of liquid electrolyte. Consequently, GPEs manufactured with 20%, 40%, and 60% liquid electrolyte are designated as P-20% S, P-40% S, and P-60% S, respectively. Following the synthesis of the GPEs into disks of approximately 2 cm in diameter and 1 mm in thickness (see Figure 1b), their chemical, thermal, and dielectric properties were characterized. For purposes of comparison, a polymerized/crosslinked PEGDA sample was prepared under similar conditions.

2.3 Chemical, thermal, and dielectric characterizations of GPEs

Fourier transform infrared spectroscopy (FTIR) was employed to monitor the rate of PEGDA polymerization in gel polymer electrolytes (GPEs), the structure of the polymer network, and the impact of lithium salt addition on crosslinking. The measurements were conducted using the attenuated total reflectance (ATR) mode with a diamond prism at wavenumbers ranging from 4000 to 500 cm^{-1} on a Perkin Elmer Frontier spectrometer.

The thermal properties of the GPEs were analyzed using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). Initially, TGA was employed to ascertain the thermal stability of the samples. The measurements were conducted using a TA Instruments TA Q50 thermogravimetric analyzer under the following experimental conditions: a temperature range of 25 to 800°C, a heating rate of 10°C/min, and a nitrogen gas flow rate of 50 mL/min. The Perkin-Elmer DSC8000 model was employed to ascertain the glass transition temperature (T_g) of GPEs. Samples with masses ranging from 6 to 12 mg were analyzed using a temperature ramp of 10°C/min between -70 and 100°C. To ensure the reliability of the results, three heating and cooling cycles were conducted.

Dielectric spectroscopy was employed to quantify the ionic conductivity of GPE. The measurements were conducted using a ModuLab-MTS impedancemeter (Solartron Analytical) with a sinusoidal AC voltage amplitude of 10 mV in the frequency range of 0.1 to 10^6 Hz at temperatures ranging from 20 to 100°C. Temperature studies were conducted using a Prolabo oven, which is a standard labora-

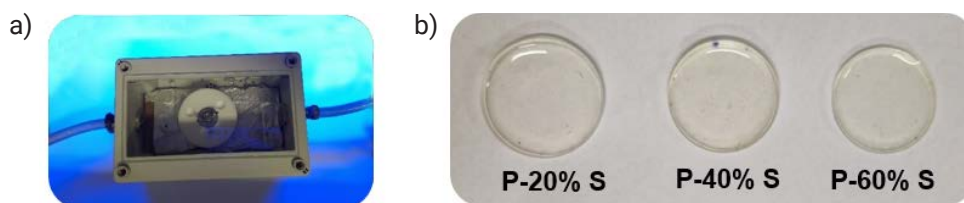


FIGURE 1: a) Teflon mold placed in an argon atmosphere for the UV-irradiation process, b) synthesized GPEs as disks measuring approximately 2 cm in diameter and 1 mm in thickness (P-20 %S, P-40% S and P-60% S).

tory apparatus. The experiments were conducted using a custom-built Swagelok apparatus. The device comprises two stainless-steel electrodes in contact with the sample on both sides and supported by a spring system. Conductivity values were determined from the Nyquist plot, which is obtained by plotting the imaginary ($-Z''$) and real (Z') parts of the complex impedance (Z^*). A fit was performed on the resulting semicircle to obtain the resistance (R), which is related to Z' , and equation (1) was used to calculate the conductivity of the GPEs.

$$\sigma' = d/RS \quad (1)$$

where σ' is the electrical conductivity (S/m), d stands for the sample thickness (m), R represents the apparent resistance (Ω), and S corresponds to the sample surface area (m^2).

3. RESULTS AND DISCUSSION

3.1 Chemical properties

Figure 2 depicts the Fourier transform infrared (FTIR) spectra of PEGDA before and after ultraviolet (UV) irradiation, with the addition of 20, 40, and 60 weight percent (wt%) of the liquid electrolyte, which forms the gel polymer electrolytes (GPEs; P-20% S, P-40% S, and P-60% S), as well as the pure liquid electrolyte. The PEGDA prepolymer spectrum exhibits characteristic bands at 1410 and 810 cm^{-1} , which are attributed to the acrylic double bonds $-CH=CH-$. The disappearance of these bands after exposure to UV light indicates that the acrylic functions have undergone complete conversion, resulting in the formation of a crosslinked polymer matrix.

In the spectra of the GPE samples (P-20% S, P-40% S, and P-60% S), new bands appear around 1780 and 1830 cm^{-1} , corresponding to the carbonate groups ($C=O$) present in the organic solvents ethyl carbonate (EC) and diethyl carbonate (DEC). This evidence corroborates the presence of a liquid electrolyte within the matrix.

Furthermore, peaks at 840 and 550 cm^{-1} are observed, which are attributed to the vibrational mode (t_{1u}) of $[PF_6]^-$ present in the lithium salt, thereby confirming its presence in GPEs (Xuan et al., 2005). It is notable that the PEGDA polymer matrix was selected for the synthesis of these GPEs due to the presence of polar carbonyl and ether groups, which act as electron donors (Liu et al., 2020). The coordination of Li^+ ions with the oxygen atoms present in these groups has the potential to enhance the ionic conductivities of GPEs.

3.2 Thermal characterization

Figure 3 depicts the thermograms obtained from the TGA analysis of polymerized/crosslinked PEGDA, GPEs with varying liquid electrolyte contents, and liquid electrolyte. The PEGDA polymer exhibited a single degradation step with an onset degradation temperature of approximately 350°C. In contrast, the pure liquid electrolyte is comprised of three distinct phenomena: (i) thermal degradation of $LiPF_6$ in electrolyte solution below 100°C, (ii) evaporation of DEC near its boiling point (130°C), and (iii) evaporation of EC at 250°C. Additionally, several degradation processes were identified for the GPE samples, including thermal decomposition of $LiPF_6$ and solvent evaporation below 250°C. The weight loss percentages indicate that the rate of solvent evaporation is slower in the GPEs than in the electrolyte. This effect becomes more pronounced as the percentage of electrolyte in the mixture decreases. Furthermore, the lithium salt in the GPEs tends to dissociate at low temperatures. The final degradation step occurred at approximately 300°C and was attributed to the degradation of crosslinked PEGDA. This indicated a loss of thermal stability in comparison to the pure PEGDA polymer. At the conclusion of the analysis at 800°C, approximately 8% of the mass of the GPE sample remained. This residue may be indicative of the formation of carbon residues during analysis under a nitrogen (N_2) atmosphere. The technical specifications for LIBs require

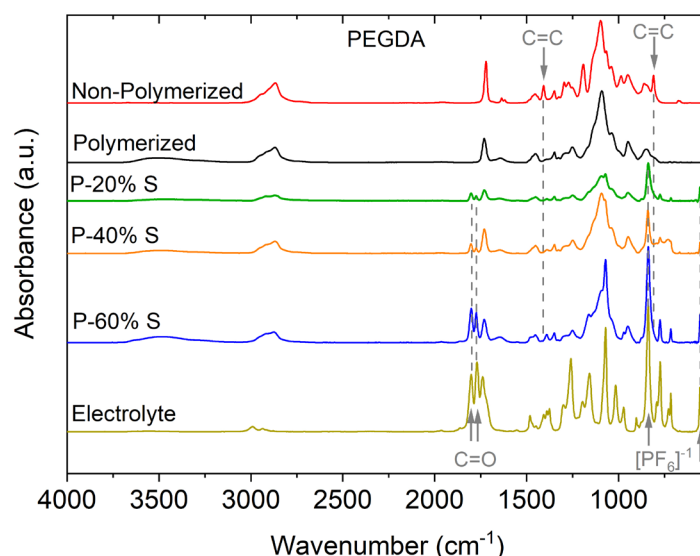


FIGURE 2: FTIR spectra of pristine PEGDA before and after polymerization/crosslinking, P-20 %S, P-40% S, P-60% S, and the pure liquid electrolyte.

a thermal stability range of -20 to 60°C. Our GPEs meet this requirement, as they are thermally stable up to at least 90°C.

Figure 4 displays the DSC curves for the PEGDA polymer and GPEs with varying liquid electrolyte concentrations.

Figure 4 illustrates that each sample exhibits a single T_g . This T_g represents the minimum temperature at which the GPEs can be effectively utilized for an application in LIBs. The T_g of PEGDA polymer is approximately -42°C, and the addition of a liquid electrolyte results in a slight shift in T_g values toward lower temperatures. As previously stated, the optimal operational temperatures for a LIB are between -20 and 60°C, in accordance with its specifications. Given that all the GPEs synthesized in this study have T_g values above -40°C, they are suitable for use in LIBs.

3.3 Dielectric characterization

The ionic conductivity of GPEs is a crucial property for their utilization in LIBs. Consequently, as previously stated, dielectric studies are of paramount importance in order to ascertain the conductivity values of GPEs. Figure 5a and b display Nyquist plots for PEGDA polymer and GPE synthesized with 20 wt% of liquid electrolyte (P-20% S), respectively, over a temperature range from 20 to 100°C.

The conductivity was calculated using Equation (1) based on the resistance measured on the circular part of the curves, as previously mentioned. Figure 5 indicates that the polymerized/crosslinked PEGDA sample exhibits higher resistance values than the GPE sample with 20 wt% liquid electrolyte, resulting in lower conductivities. Specifically, at 20°C, the ionic conductivity of the PEGDA polymer

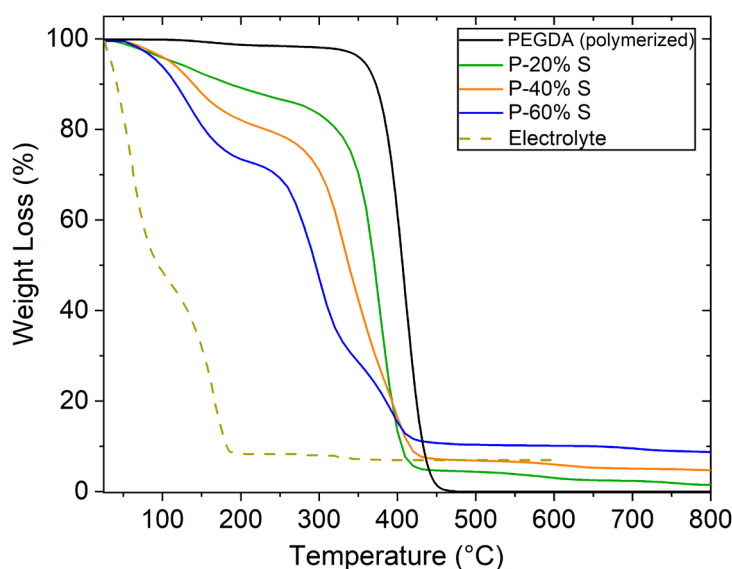


FIGURE 3: TGA curves of polymerized/crosslinked PEGDA, P-20 %S, P-40% S, P-60% S, and liquid electrolyte. Data for the liquid electrolyte were extracted from (Netzsch, 2019).

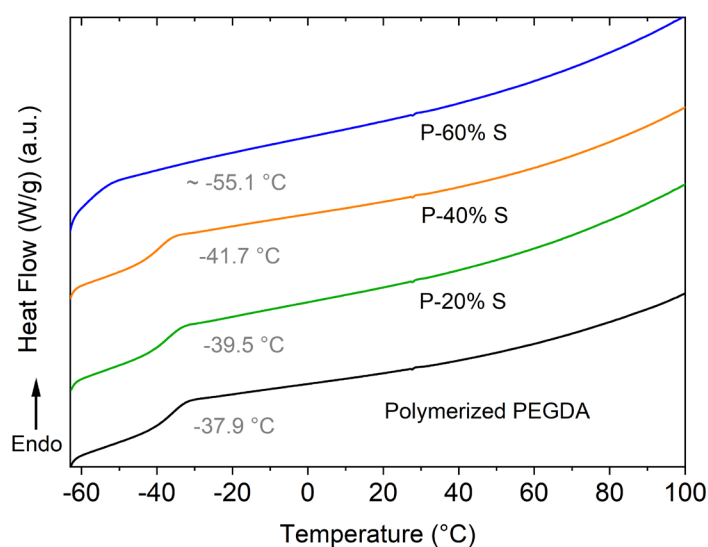


FIGURE 4: DSC thermograms of polymerized/crosslinked PEGDA, P-20% S, P-40% S, and P-60% S in the temperature range of -70°C to 100°C at a rate of 10°C/min. The temperatures indicated in the figure correspond to the glass transition.

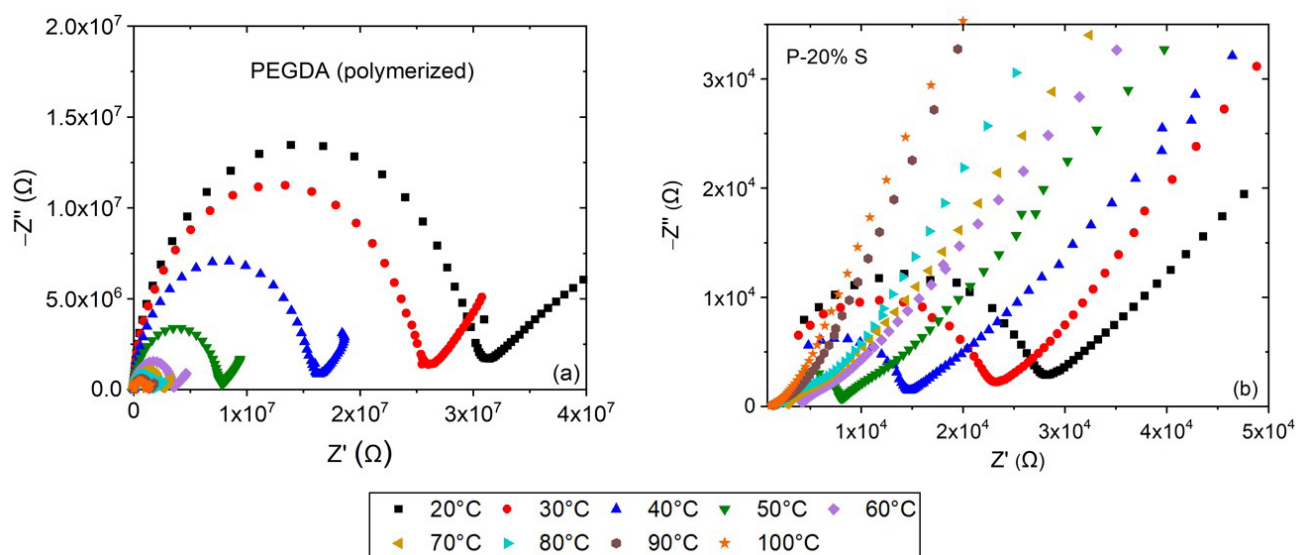


FIGURE 5: Nyquist plots of (a) polymerized/crosslinked PEGDA and (b) GPE synthesized with 20 wt% of liquid electrolyte (P-20% S), respectively. The measurements were conducted over a temperature range of 20 to 100°C.

is 3.6×10^{-6} S/m, while that of the GPE (P-20% S) is 3.4×10^{-4} S/m, representing a two-decade increase. Moreover, it has been observed that an increase in temperature results in a reduction in the radius of the circular portion of the Nyquist plots, which in turn leads to an increase in the conductivity values of the samples. At a temperature of 100°C, the conductivity value of GPE is 1.1×10^{-2} S/m, which is two orders of magnitude higher than the conductivity value at 20°C. This is attributed to the enhanced mobility of charge carriers and the facilitated movement of polymer chains within the network.

Figure 6 presents a summary of the evolution of ionic conductivity with temperature and liquid electrolyte concentration. The conductivity values increase with rising temperature and liquid electrolyte concentration in the synthesized GPEs. The sample with a 60 wt% liquid electrolyte

concentration (P-60% S) exhibited the highest conductivity values among the other GPEs, with a value of 6.6×10^{-3} S/m at 30°C. The reported conductivity is comparable to that of traditional liquid electrolytes, which range from approximately 10^{-3} to 1 S/m (Liu et al., 2020; Mindemark et al., 2018; Zhou et al., 2019).

The temperature dependence of the ionic conductivities of polymerized/crosslinked PEGDA and synthesized GPEs was analyzed using the Arrhenius law. Figure 6 illustrates that all samples exhibited Arrhenius-type behavior within the temperature range of 20 to 100°C. The activation energies (E_a) were determined to be 0.44, 0.42, and 0.38 eV for the P-20% S, P-40% S, and P-60% S GPEs, respectively, by fitting the experimental data. These activation energies exhibit a slight increase with increasing polymer content and fall within the typical range of those reported for GPEs

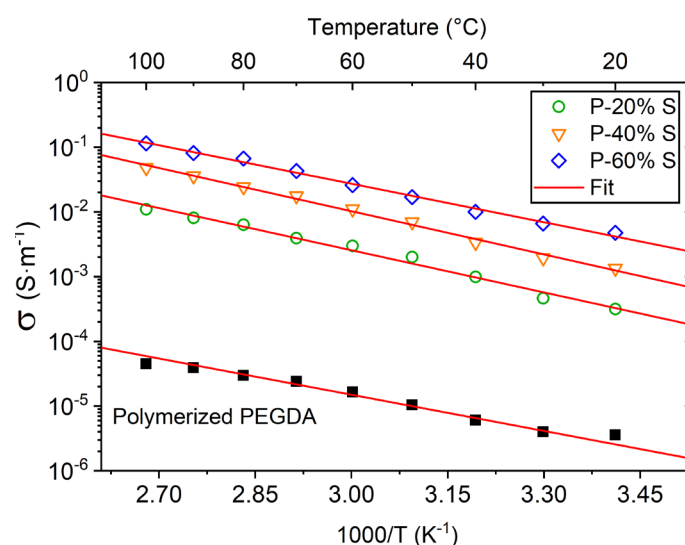


FIGURE 6: Conductivity measurements of polymerized/crosslinked PEGDA, P-20% S, P-40% S, and P-60% S at varying temperatures. The solid red lines represent data analysis conducted in accordance with the Arrhenius law.

synthesized elsewhere (D'Angelo & Panzer, 2017; Shalu et al., 2013).

4. CONCLUSIONS

In this study, PEGDA-based GPEs with varying concentrations of LiPF_6 dissolved in carbonated organic solvents were successfully synthesized via ultraviolet (UV) irradiation. Infrared spectroscopy demonstrated that the liquid electrolyte was confined within the polymer matrix. Thermal analysis demonstrated that all synthesized GPEs exhibited thermal stability within a temperature range of -40°C to 90°C , which is higher than the conventional temperature range of -20°C to 60°C typically observed for LIBs. Dielectric measurements demonstrated that the ionic conductivity of the GPEs increased with increasing temperature and liquid electrolyte concentration. The GPE with 60 wt% liquid electrolyte (P-60% S) exhibited the highest ionic conductivity among the samples, with a value of $6.6 \times 10^{-3} \text{ (S/m)}$ at 30°C . The ionic conductivity is comparable to that reported in the literature for electrolyte polymers. The aforementioned properties indicate that the GPEs prepared via the one-step method may be suitable as electrolytes in LIBs.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the financial support provided by the University of Lille and the Région Hauts-de-France.

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SIMULATION AND ECONOMICAL ANALYSIS OF HYDRO-ND PROCESS FOR THE RECOVERY OF RARE EARTH FROM END-OF-LIFE PERMANENT MAGNETS: NEW-RE AND INSPIREE PROJECTS

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Article Info:

Received:
18 April 2024
Revised:
3 June 2024
Accepted:
12 July 2024
Available online:
30 September 2024

Keywords:

Rare earth elements
Recycling
Economic Analysis
Permanent Magnets
Hydrometallurgy
Circular Economy

ABSTRACT

The growing demand for rare earth elements (REEs) and sustainable development issues have made the REEs recovery from permanent magnets (PMs) attract the attention of many researchers in the last decade. The NEW-RE and INSPIREE projects have been introduced to evaluate the recovery of REE elements from permanent magnets on a pilot and industrial scale. In this research, the economic aspect of the mentioned projects was performed using SuperPro software. The main aim of the work is to highlight the critical aspects of the process for a targeted optimization. The results showed that the CAPEX and OPEX for treating 3,600 tons/year of permanent magnets are 9 and 110 million euros, respectively, and an EBITDA of 4.8 million euros can be achieved (payback period less than two years). In the economic model, the cost of spent PMs was considered equal to 50% of its REEs value (14,251 €/ton). Also, it was found that the main OPEX costs are raw materials (65%) and energy (16%), respectively. It needs to be pointed out that the maximum price of permanent magnets can be 15,230 €/ton (BEP).

1. INTRODUCTION

The increase in sensitivity towards issues such as safeguarding resources and the environment is increasingly pushing towards adopting circular economy models. Thanks to this phenomenon and the strong profitability of recovering critical raw materials from end-of-life devices, numerous companies have been created to recycle industrial waste and electrical and electronic equipment.

The application of permanent magnets containing rare earth elements (REEs) has increased dramatically in various electrical and electronic devices due to their special and unique properties. The supply chain (China, Australia, and Russia are the primary world producers (Kapustka et al., 2020)) and their increasing consumption (the consumption is expected to be more than 350,000 tons per year in 2035 (Yang et al., 2017) have led to the classification of several REEs such as Nd, Pr, Dy, and Y as the most critical raw materials by the European Union (Eyl-Mazzega et al., 2020). In addition, depositing end-of-life electronic equipment causes loss of mineral resources and conse-

quently increases mining activities. Thus, recovery of rare earth elements (REEs) from end-of-life permanent magnets (PMs) is essential from sustainable development, economic, and environmental points of view. To date, numerous devices containing REPMs are starting to return as waste from their respective markets: wind turbine generators, HDDs, electric motors, and acoustic devices. As a result, there is a need to have more and more operational plants to manage these waste flows correctly, providing adequate solutions to avoid the loss of critical raw materials.

Although hydrogen decrepitation (HD) is the most widely used method of REEs recovering from PMs, this method has some drawbacks, such as huge waste production, and is not applicable for treating all PMs types. In light of these considerations, as well as the advantages of hydrometallurgical compared to pyrometallurgical processes (less energy required, removal of gas pollution, and environmentally friendly) (Romano et al., 2023 (a)), hydrometallurgical methods can be considered one of the most promising technologies in this field.

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Various research works on a laboratory scale were carried out to investigate the effect of chemical and operational parameters on dissolving and separation efficiencies. It was found that the demagnetization of PMs is an essential step to increase the selectivity and efficiency of the leaching stage as well as to facilitate the subsequent steps of the recycling process (Kumari et al., 2018; Klemettinen et al., 2021). Previous studies showed that organic (Gergoric et al., 2018; Romano et al., 2023 (b)) and inorganic (Lee et al., 2013; Uysal et al., 2023) acids can effectively dissolve valuable metals of PMs. Also, it was reported that DEHPA is more efficient in separating both light and heavy REEs than Cyanex 272 and Cyanex 572 (Yang et al., 2017; Ni'am et al., 2020).

In 2019, a novel method for the recovery of REEs from end-of-life permanent magnets, based on hydrometallurgical methods, was introduced by Birloaga and Veglio. This method leaches demagnetized and ground spent PMs by the citric acid solution to dissolve REEs, iron, and boron. The REEs are extracted at the solvent extraction, while iron and boron remain in the raffinate solution and are recovered as the byproduct of this process. Also, the loaded organic phase is stripped by phosphoric acid to produce REEs- phosphate and convert to REE-oxalate in the next step. In the end, the REE oxide is achieved by introducing the REE-oxalate to a furnace. According to the appropriate laboratory results of this method, the NEW-RE and IN-SPIREE projects were defined to evaluate the REEs recovery on pilot and industrial scales, respectively.

The main objective of this work is to highlight the critical aspects of the Hydro-Nd process to optimize it in a targeted way. This result is achieved through a detailed simulation that allows material and energy balance realization.

The simulation allows us to analyze the system's material flows and energy consumption, identifying inefficiencies and improvement points. This activity is essential for the scale-up of the process, moving from the laboratory to the industrial scale through the pilot phase. This way, it is possible to intervene precisely in the critical process phases, ensuring a more effective and sustainable optimization and facilitating the transition to large-scale production.

2. MATERIALS AND METHODS

This work aims to create a model to describe the hydrometallurgical process using the SupePro Designer v9.5 process simulator. This simulator is widely used in the hydrometallurgical sector due to the numerous features it offers. The scenario analyzed concerns the continuous treatment of approximately 3600 tons/year of end-of-life PM.

2.1 PM sample

In order to proceed with the process simulation, it is necessary to identify all the input streams. Among the different streams, the one most influenced by the market is that of permanent magnets. Results obtained from laboratory tests on samples of NdFeB permanent magnets are used as a reference composition for this current. Figure 1 shows the composition of the supply stream.

2.2 Process Description

The hydrometallurgical process studied is composed of different phases. The permanent magnets are sent to an oven for the thermal demagnetization phase at approximately 400°C. The oven is powered by the hydrogen

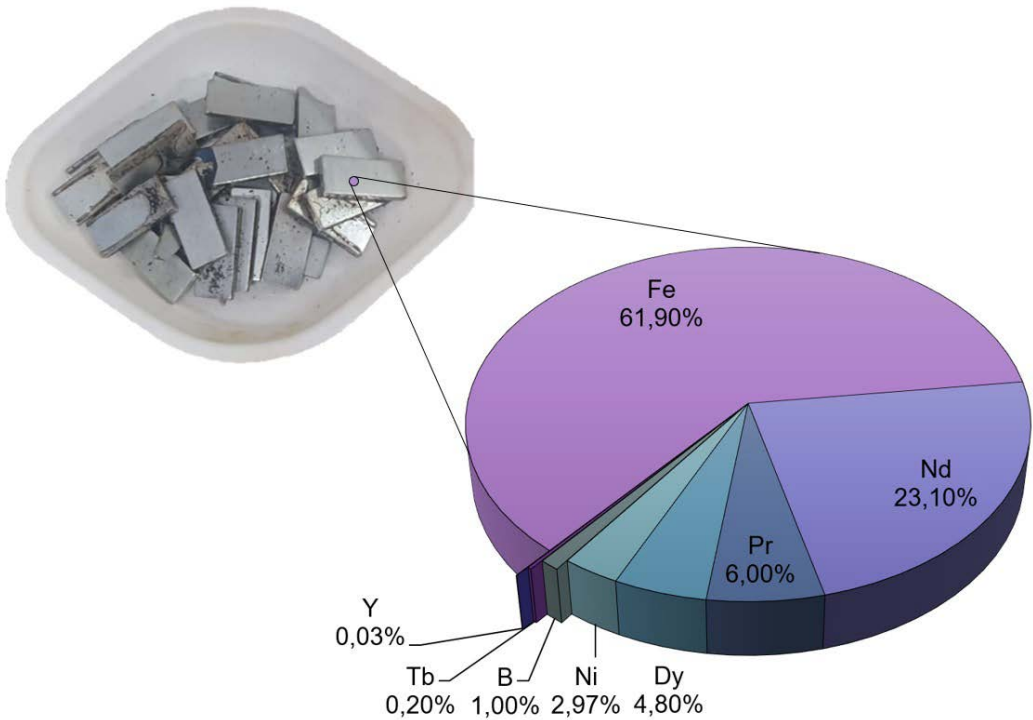
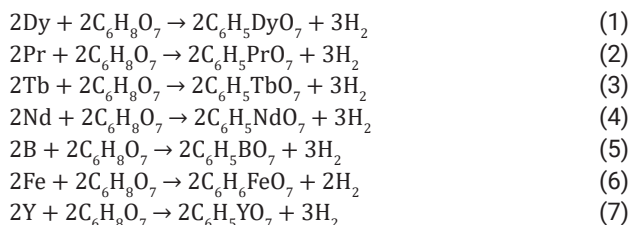


FIGURE 1: Element composition of PM sample considered in the simulation.

produced in the extraction phase, described below. Once demagnetized, the magnets are removed from the rotors and subjected to a grinding process. The granulate is then introduced into the reactor, where the REEs, iron, and boron contained in the demagnetized powders are leached with a citric acid solution. The following reactions are involved in the process:



In the overall chemical reaction, in addition to the solubilization of REEs (Nd et al.), boron, and iron, hydrogen is developed, extracted from the reactor, and sent to the demagnetization oven. Figure 2 shows the simplified block diagram of the process.

Once the reaction is over, the resulting solution is filtered, obtaining a solid residue rich in nickel and a liquid solution rich in rare earth to be recovered.

The solution is sent to a mixer, where an extraction with solvent (D2EHPA and n-Heptane) takes place. Subsequently, thanks to a settler, the two liquid phases are separated. Aqueous solution rich in iron, boron, and citric acid is first treated with oxalic acid. The reaction between the acid and

the iron ions precipitates iron oxalate, which is separated from the liquid waste and filtered again. The filtered water is recirculated in the system while the waste treated with calcium hydroxide and calcium citrate goes to a boron recovery section. Boron hydroxide is recovered, and the remaining water is sent for reuse.

The other solution that comes out of the settler, rich in rare earth ions, is sent to a mixer for the second extraction with solvent: phosphoric acid is inserted into the processing unit, which causes the rare earth ions to return to the aqueous solution purifying the solvent that is recirculated. The aqueous solution is treated with oxalic acid, obtaining rare earth oxalates, which, once precipitated, are recovered.

2.3 Simulation setup

A model was created on the SuperPro Designer v9.5 software to characterize each flow rate and equipment of the process, starting from the above data. Figure 3 shows the complete simulation flowsheet.

The diagram highlights the process inputs (for example, PMs Powder) in light blue and all the outputs (for example, Hydrogen) in orange. The scheme also includes the process for the recovery of rare earth oxalates and the recovery of iron. The simulation was set directly with the possibility of recycling part of the solutions used (purple dashed lines). All recycles were analyzed in order to minimize reagent consumption.

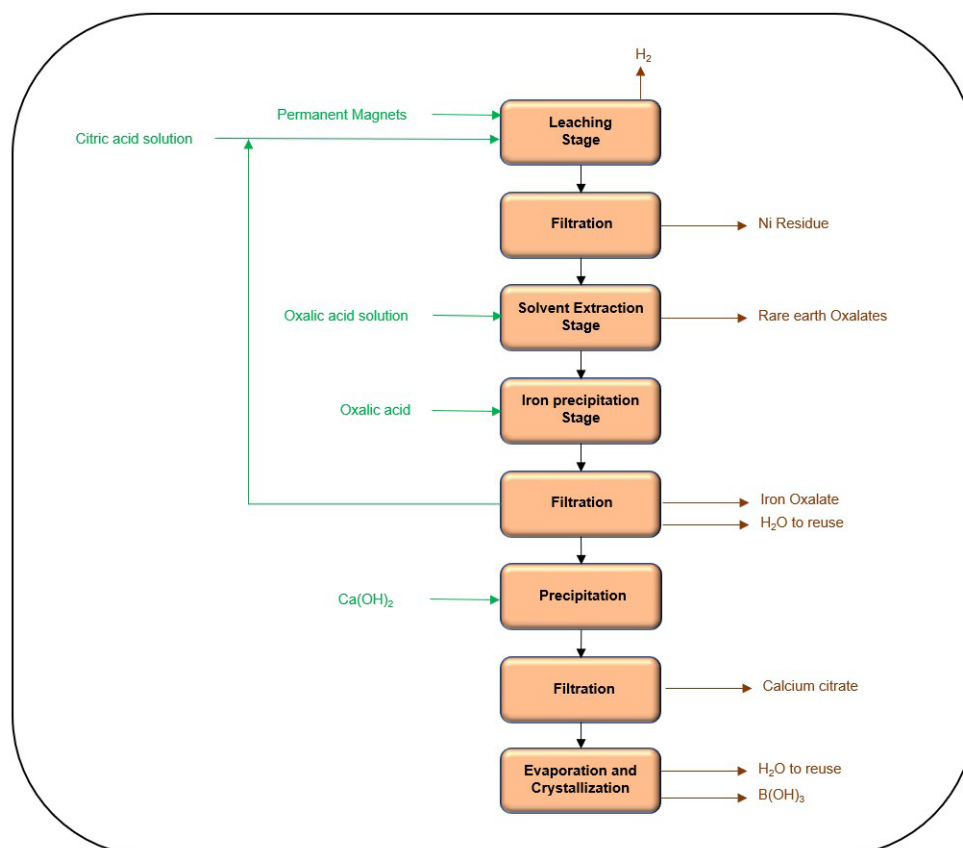


FIGURE 2: Simplified block diagram of the Hydro-Nd process.

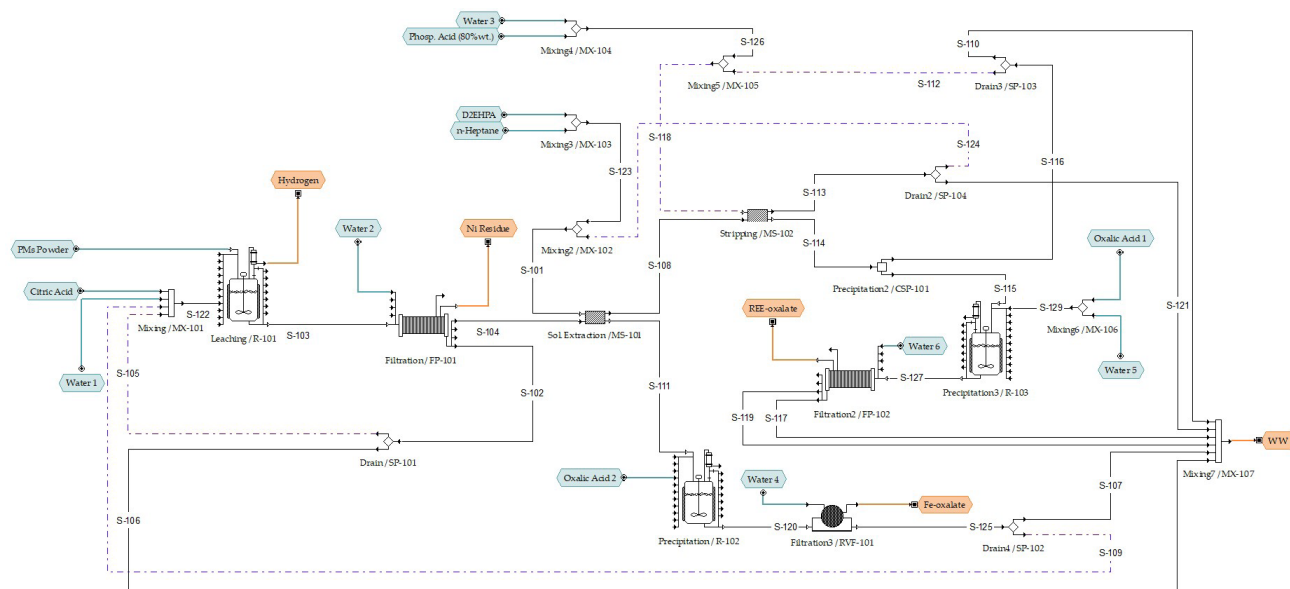


FIGURE 3: Flowsheet of SuperPro Designer simulation.

2.3.1 Equipment setup

The different equipment was configured based on the results obtained from laboratory-scale experimental tests. Table 1 briefly summarizes the procedures and various operations.

2.3.2 Input streams setup

The variables of the different flow rates entering the plant were calculated based on the optimizations carried out on a laboratory scale for each of the envisaged unit operations. As regards the leaching section, a citric acid concentration of 1.45 mol/L was considered, with a pulp density of approximately 10% wt./vol. The solvent extraction section used a volume ratio between the two phases

(organic and aqueous) equal to 1. The exact ratio was used to prepare the organic phase: the exact volume quantities of D2EHPA and n-Heptane. Regarding the stripping phase, the volumetric ratio of the organic phase/aqueous phase is 2:3. The quantity of phosphoric acid entering this section was chosen to obtain the optimal value of 1.8 mol/L during the operation. In the conversion phase of rare earth phosphates into rare earth oxalates, a solid-liquid ratio of 7% was considered, with an incoming oxalic acid concentration of 1.6 mol/L. Finally, for the precipitation phase of the iron oxalate to recover this element, a quantity of oxalic acid over 80% compared to the stoichiometric quantity obtained from the following reaction was used:

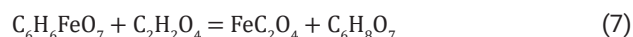


TABLE 1: Summary of Equipment Setup.

Equipment	Operation	Procedure	Setting
R-101	Leaching	REACTION	Reaction Progress: 95-99%
FP-101	Filtration	FILTER	Solid fraction removed: 100%
		CAKE-WASH	Wash stream amount: 30%vol. leaching solution
MS-101	Solvent Extraction	REACTION	REEs Reaction Progress: 95-99%
MS-102	Stripping	REACTION	Fe Reaction Progress: 30%
R-102	Precipitation	REACTION	Reaction Progress: 100%
R-103	Precipitation	REACTION	Fe Reaction Progress: 100%
FP-102	Filtration	REACTION	REEs Reaction Progress: 100%
		FILTER	Solid fraction removed: 100%
RVF-101	Filtration	FILTER	LOD Cake Dryness: 30%
		CAKE-WASH	Wash stream amount: 30%vol. leaching solution
RVF-101	Filtration	FILTER	Solid fraction removed: 100%
		CAKE-WASH	Cake Porosity - Cake Dryness: 0.4 vol/vol
			Available in Wash-In Stream

To estimate the volumes of the different equipment it was necessary to enter the different residence times. The residence times used were:

- Leaching Time: 180 min;
- Solvent Extraction: 20 min;
- Stripping: 90 min;
- Oxalates production: 60 min;
- Iron Production: 60 min.

All operations are conducted at room temperature. This aspect allows for a process with low energy consumption. The energy consumption of the process includes the mixing operations in the different reactors and the pumps for moving the different streams (obviously, if the thermal sections of the demagnetization oven and the oven for converting the oxalates into oxides are excluded). Figure 4 shows the summary of INPUT streams.

2.3.3 Economic Analysis

Table 2 briefly summarizes the calculations and assumptions made for the economic analysis.

3. RESULTS

3.1 Material balance

3.1.1 Drain Optimization

Before analyzing the results of the material and energy balances in detail, an optimization of Drains 1 and 2 was carried out. In this regard, the influence of the split ratio between the two currents coming out of each Drain was evaluated to obtain the lowest possible reagent consumption in the leaching reactor. Figure 5 shows the results of this optimization.

Thanks to purges, significantly reducing the demand for raw materials is possible. In particular, the following is

achieved: a reduction of approximately 50% in the requirement for citric acid and the total removal of the need for dilution; a 90% reduction in the demand for D2EHPA and n-Heptane and a 90% reduction in the demand for sulfuric acid and the water needed to dilute it. Due to the demand for oxalic acid, it is impossible to optimize everything through recirculation; thanks to water in the solution in which the iron citrate is present, dilution is optional.

3.1.2 Output

The material and energy balances of the proposed method were investigated for comprehensive characterization of all process streams. Figure 6 shows the results of the outputs of the process streams. It needs to be pointed out that the required reactions of this method take place under ambient temperature and pressure conditions, which is important from economic, technical, and safety standpoints.

The final product, REE-oxalate, consists of more than 86% Nd and Pr oxalate. The high concentration of REEs in the REE-oxalate confirms the subsequent industrial applications of this production. On the other hand, due to the high concentration of Ni in leaching residues (78.5%), recovery of nickel from these wastes can be important from economic and environmental aspects.

Although ~ 25% of iron was extracted by DEHPA in the process, the low iron concentration in the REE-oxalate powder (~0.1%) presents a notable advantage. Also, the iron oxalate achieved using this method showed an impressive purity level of 98%, which facilitates its conversion into iron oxide for subsequent industries as a raw material for Nd-FeB permanent magnet production.

Another point of consideration is the low concentration of boron in loaded DEHPA and was remained in the waste solution of the iron recovery stage. To address this,

Stream Name		PMs Powder	Citric Acid	Water 2	D2EHPA	n-Heptane	Phosp. Acid	Water 3
Temperature	°C	25	25	25	25	25	25	25
Pressure	bar	1	1	1	1	1	1	1
Mass Flow	kg/h	500,00	743,00	1615,87	277,12	183,24	278,00	730,00
Mass Composition	%wt.							
Liquid phase				100,00%	100,00%	100,00%	100,00%	100,00%
Water				100,00%				100,00%
D2EHPA					100,00%			
n-Heptane						100,00%		
Phosphoric Acid							100,00%	
Solid Phase		100,00%	100,00%					
Boron		1,00%						
Dysprosium		4,77%						
Iron		62,00%						
Neodymium		23,00%						
Nickel		3,00%						
Praseodymium		6,00%						
Terbium		0,20%						
Yttrium		0,03%						
Citric acid			100,00%					
Oxalic acid								

FIGURE 4: Summary of INPUT streams.

TABLE 2: Assumptions and calculations of economic analysis.

Parameter	Value / Comment	
Base year	2025 (January)	
	EUR/USD = 1.0837	
Currency	EUR	
Plant lifespan	20 years	
Plant location	Ceccano (Italy)	
Plant construction material	-	
	Material Factor (f_m) = 1.5	
ISBL	$ISBL = C_E + C_{er} + C_p + C_i + C_{el} + C_c + C_s + C_l$	
Equipment Purchase Cost (C_E)	Sum of individual purchased equipment	
Equipment Erection	$f_{er} = 0.5$	$C_{er} = C_E (f_{er} / f_m)$
Piping	$f_p = 0.6$	$C_p = C_E f_p$
Instrumentation and control	$f_i = 0.3$	$C_i = C_E (f_i / f_m)$
Electrical works	$f_{el} = 0.2$	$C_{el} = C_E (f_{el} / f_m)$
Civil works	$f_c = 0.3$	$C_c = C_E (f_c / f_m)$
Structures and buildings	$f_s = 0.2$	$C_s = C_E (f_s / f_m)$
Lagging and paint	$f_l = 0.1$	$C_l = C_E (f_l / f_m)$
OSBL	$f_{OSBL} = 0.4$	$OSBL = ISBL f_{OSBL}$
Design and Engineering (D&E)	$f_{D\&E} = 0.25$	$CD\&E = ISBL f_{D\&E}$
Contingency (X)	$f_x = 0.1$	$C_x = ISBL f_x$
Total Investment Cost (TIC)	$TIC = ISBL + OSBL + D\&E + X$	
Fixed production costs estimation		
Operating labor (OL)		
Supervision	25% of OL	
Direct salary overhead	60% of (OL + Supervision)	
Maintenance	5% of ISBL	
Taxes & insurance	2% of ISBL	
Rent of land & buildings	2% of (ISBL + OSBL)	
General plant overhead	65% of (OL + Supervision)	
Allocated environmental charges	1% of (ISBL + OSBL)	
Fees & royalties	1% of TIC	
Capital charges	1% of TIC	
Iron Oxalate	1.500,00	€ · t ⁻¹
Nichel	25.000,00	€ · t ⁻¹
Dysprosium Oxide	247.000,00	€ · t ⁻¹
Neodymium Oxide	52.560,00	€ · t ⁻¹
Praseodymium Oxide	52.880,00	€ · t ⁻¹
Terbium Oxide	724.860,00	€ · t ⁻¹
Yttrium Oxide	55.460,00	€ · t ⁻¹
Wastewater	100.000,00	€ · t ⁻¹

the application of other methods to recover B needs to be evaluated. It should be noted that recovering B not only increases overall resource efficiency but also minimizes waste generation.

3.2 Economic Analysis

Through the economic analysis, it was possible to identify some of the indices to evaluate the feasibility of

the investment necessary for the plant's construction. For the case study of 3600 ton/y of permanent magnets (500 kg/h) with a cost equal to 50% of the intrinsic economic value (IEV) associated with the rare earth content, the following values were calculated: a CAPEX of approximately 9 M€, an OPEX of approximately 110 M€ and revenues worth approximately 115 M€. Based on these values, an EBITDA of approximately 4.8 M€ and a PBP of less than two years

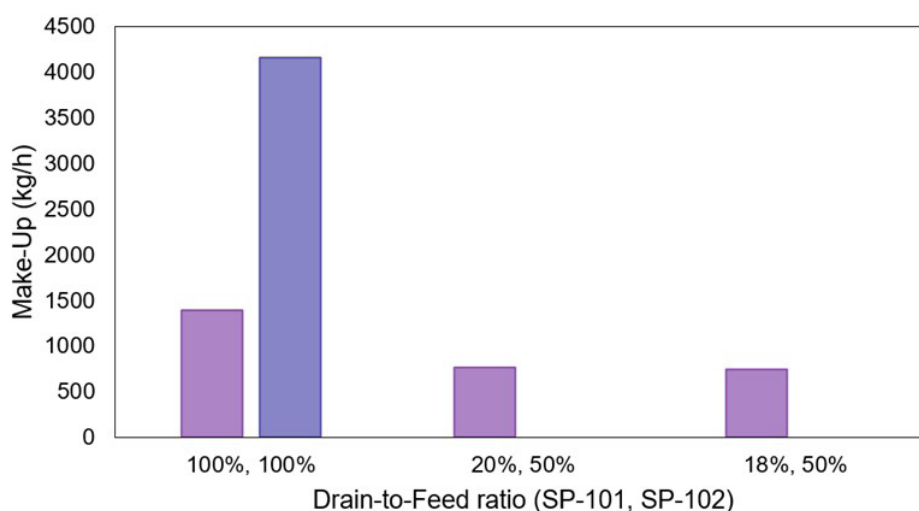


FIGURE 5: Optimization of the Drain-to-Feed ratio for the two splits SP-101 and SP-102.

Stream Name	Hydrogen	Ni Residue	REE-oxalate	Fe-oxalate	WW
Temperature °C	25	25	25	25	25
Pressure bar	1	1	1	1	1
Mass Flow kg/h	15,99	26,27	436,45	1.344,22	10071,85
Mass Composition %wt.					
Gas phase	100,00%				
Hydrogen	100,00%				
Liquid phase		27,24%	28,84%	43,60%	100,00%
Water		100,00%	100,00%	100,00%	77,53%
Oxalic Acid					7,53%
Citric Acid					6,48%
D2EHPA					2,81%
Phosphoric Acid					2,45%
n-Heptane					1,82%
Boron Citrate					0,90%
Iron Phosphate					0,48%
Solid Phase		72,76%	71,16%	56,40%	
Boron		0,52%			
Dysprosium		2,50%			
Iron		3,24%			
Neodymium		12,03%			
Nickel		78,47%			
Praseodymium		3,14%			
Terbium		0,10%			
Yttrium		0,02%			
Dysprosium Oxalate			13,23%	0,17%	
Neodymium Oxalate			68,30%	0,85%	
Praseodymium Oxalate			17,78%	0,23%	
Terbium Oxalate			0,57%		
Yttrium Oxalate			0,12%		
Iron Oxalate				98,75%	

FIGURE 6: Results of E&M balance (OUTPUT streams).

were calculated. Table 3 shows the results of the Business Plan created per ton of PMs. It should be noted that the required amount of OPEX is significantly higher than CAPEX for this process.

4. DISCUSSIONS

Figure 7 shows a breakdown of the OPEX involved in the process of recovering REEs from spent rare earth

permanent magnets. From the economic analysis, it was observed that the costs related to the procurement of raw materials, which constitute 65% of the total OPEX, and the energy required for this process, which constitutes 13% of the OPEX, constitute the main parts of the OPEX portion. It is worth noting that the major share of the cost of raw materials is considered for the use of permanent magnets, the price of which is determined based on the market demand.

TABLE 3: Estimated CAPEX, OPEX, and hydrometallurgical process cost for studied scenarios. Values referred per ton of treated PMs.

	€ · t ⁻¹
CAPEX (capital intensity)	1.956,49 €
ISBL	1.436,31 €
Purchased equipment	582,29 €
OSBL	574,52 €
Design&Engineering	359,08 €
Contingency	143,63 €
OPEX (annual)	31.239,59 €
Wastewater	2.014,40 €
Utilities	4.069,68 €
Raw materials	20.348,38 €
Contingencies	4.539,09 €
Operating labor	268,06 €
Supervision	67,01 €
Direct salary overhead	201,04 €
Maintenance	71,82 €
Taxes & insurance	28,73 €
Rent of land & buildings	40,22 €
General plant overhead	217,80 €
Allocated environmental charges	20,11 €
Fees & royalties	25,14 €
Capital charges	25,14 €
Revenues (Sales)	32.099,22 €

Furthermore, the findings show that waste storage costs account for 10% of total OPEX. It should be noted that this cost can be reduced by recovering nickel. Thus, the waste volumes and associated storage costs can be reduced.

Based on the current flowsheet, the key to improving the economic efficiency of this process is identifying solutions to procure spent permanent magnets at a lower cost. Addressing this aspect holds promise as the primary avenue for improving the overall cost-effectiveness of the process.

Based on the previous observations, thanks to the model created, a sensitivity analysis was performed to understand the trend of the PBP as the price of the lead magnets varied. The results of this sensitivity analysis are shown in Figure 8.

The PBP varies very little for permanent magnet costs below 14,500 €/ton. However, if this value is exceeded, the PBP grows significantly asymptotically at the Break Event Point (BEP) by approximately 15,230 €/ton. Magnet costs higher than this value led to an unprofitable activity. Consequently, the increase in the price of spent permanent magnets beyond 15,230 euros/ton, without a corresponding adjustment in the selling price of the final product (REE-oxalate) leads to the investigation of alternative methods for the recovery of REEs through hydrometallurgical approaches. Notably, the industrial plant of the INSPIREE project is designed with a flexible circuit that allows for modification of the flow of materials, solutions, and chemicals. Also, to reduce the economic risk of these projects, the recycling of lithium battery cathodes (LFP) and photovoltaic panels was also considered in the design phase. This strategic decision not only expands the scope of the project but also increases its flexibility by diversifying potential revenue streams and reducing dependence on a single source of raw materials.

5. CONCLUSIONS

The recovery of material and energy from industrial waste and end-of-life products is increasingly a fundamental element for the sustainable development of countries. In particular, recovering rare earths from permanent magnets at the end of their life is of great strategic interest.

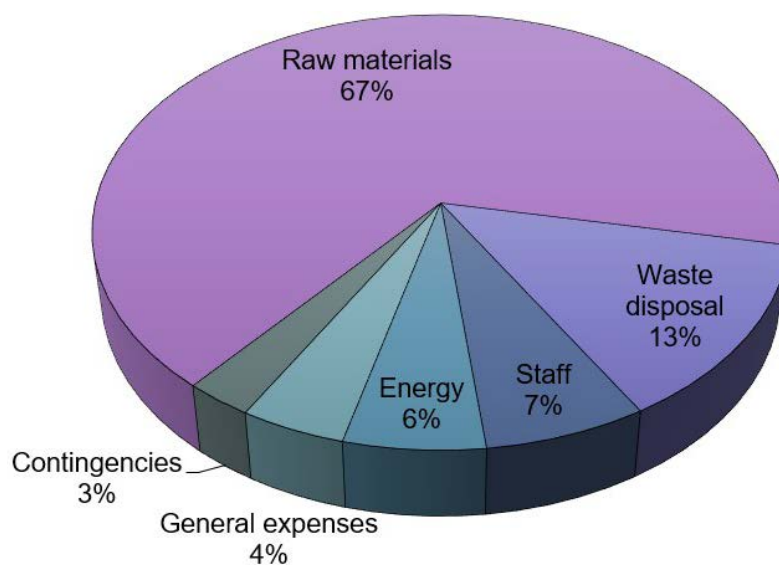


FIGURE 7: Influence on OPEX of some factors.

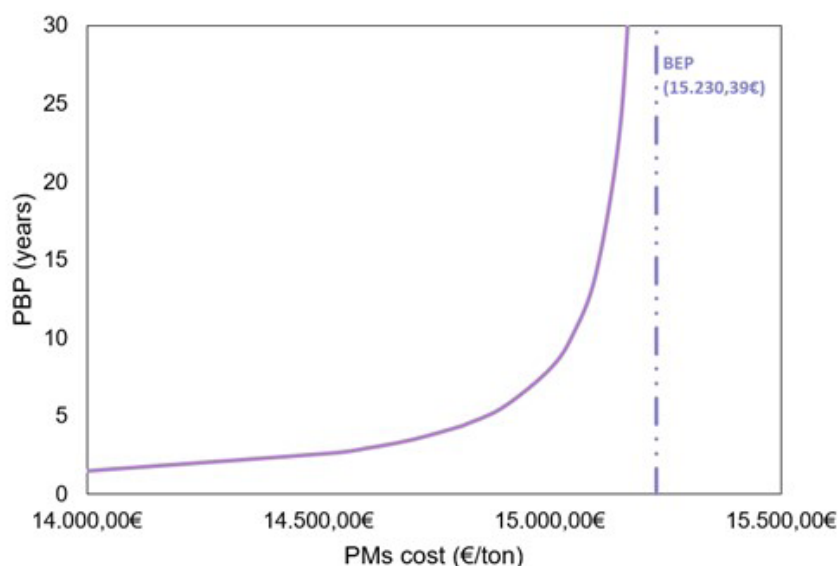


FIGURE 8: Trend of the PBP with the price of permanent magnets.

In this work, the Hydro-Nd hydrometallurgical process was analyzed from an economic point of view.

By creating a simulation using SuperPro Designer v9.5 software, it was possible to characterize all the process currents and create a model that allows sensitivity analyses to be carried out for different scenarios.

The scenario with an inlet flow rate of 3600 tons/h (500 kg/h) was studied in detail. Thanks to the model created, the process was optimized in terms of reagent consumption thanks to the inclusion of various recirculations. An economic analysis was set up starting from the optimized process.

For the scenario considered, with a magnet price of around 14,251 €/ton, there is a CAPEX of around 9 M€ and an OPEX of around 110 M€. This high OPEX value is due to the high cost of the magnets. However, an equally high revenue of around 115 M€ allows for an EBITDA of around 4.8 M€ and a PBP of less than two years. By evaluating the influence of the price of magnets on the PBP, the BEP was determined to be approximately 15,230 €/ton.

The economic analysis has allowed us to identify the critical points of the Hydro-Nd process for subsequent targeted optimization. It has been seen that the most critical aspect is linked to the cost of raw materials. In this regard, work is being done on a laboratory and pilot scale to optimize this aspect, facilitating the scale-up of the process towards large-scale production.

FUNDING

This study is financially supported by the LIFE22-ENV-IT-INSPIREE project (number: 101113882).

ACKNOWLEDGEMENTS

The authors thank all the NEW-RE and INSPIREE Consortium partners for collaborating in this research activity. The authors would like to thank the University of L'Aquila for their precious support.

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DESIGN OF LANDFILLS AND THE UTILIZATION OF WASTES AS SUSTAINABLE LINER MATERIALS: A MINI-REVIEW

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Article Info:

Received:
10 December 2023
Revised:
7 April 2024
Accepted:
19 April 2024
Available online:
2 September 2024

Keywords:

Landfill
Leachate
Landfill liner
Wastes
Environment

ABSTRACT

Modern landfills, designed to minimize environmental and health risks, play a crucial role in solid waste disposal. Poorly managed municipal solid waste facilities have severe environmental consequences, prompting intervention and remediation. Landfill construction is not enough; regular maintenance is essential to prevent harm. Landfills' historical evolution reflects societal needs, technological advancements, and environmental awareness. The 20th century saw engineered landfills with practices like compaction, daily covering, and leachate collection. Despite progress in recycling and incineration, landfills remain dominant, posing challenges like methane emissions, leachate contamination, and land use. Research emphasizes integrating landfills with recycling, composting, and waste-to-energy technologies for a sustainable future. Engineered landfills are advantageous over dumpsites, in terms of groundwater and air protection, odor control, energy generation, and job creation. Seismic analysis of landfills addresses deformation and seismic reactivity, emphasizing proper liner construction to reduce seismic impact. Leachate, a potential environmental hazard, requires careful management with effective liners to prevent groundwater and soil pollution. Various materials, including fly ash, paper mill sludge, waste foundry sand, recycled concrete aggregate, rice husk ash, and plastic waste, are reviewed for landfill liners. Research shows these materials can offer effective barriers, reducing environmental impact and promoting sustainability. Future studies should continue exploring alternative materials, considering factors like strength, hydraulic conductivity, and environmental protection.

1. INTRODUCTION

Modern landfills are sophisticated containment systems meant to reduce health and environmental hazards (Hughes et al., 2005). Li et al. (2017) stated that due to their ease of use and low construction costs, landfills are significant systems for discarding solid wastes. Employed in the context of municipal waste management, this system facilitates its ultimate disposal, and it was designed and built with minimal environmental impact in mind. In the early 1990s, the landfill design concept favoured absolute containment and isolation. Subsequent aftercare and renewal of barriers is necessary where waste is non-degradable and hazardous. The fundamental tenet of all contemporary landfills is that waste products which would not eventually become stable or inactive will be considered as "Stored," not "Disposed" (Rani & Chandra, 2017).

The effect of poorly managed municipal solid waste (MSW) facilities on the environment cannot be over-emphasized. A case was studied by Tamunobereton-ari et al. (2012) in Port-Harcourt, Nigeria. Canvassing techniques

were utilized to gather the necessary data, and the results revealed that the operators of these facilities did not adhere to operational norms. The overall environment around the facilities had deteriorated in appearance and was severely polluted. Since groundwater near the landfills was potentially contaminated, government agencies took swift action. They closely monitored the landfill operators' efforts to follow regulations. This included assessing safe operation limits, closure procedures, and long-term environmental impacts. The focus was on ensuring the safety of the surrounding area for as long as the landfill remains in use and even after closure. Landfills are significant contributors to global methane emissions, a potent greenhouse gas with 28 times the warming potential of CO₂ over a 100-year period (IPCC, 2021). Also, leachate leakage from unlined landfills can contaminate groundwater sources, posing health risks to nearby communities (Renou et al., 2008). These factors can become a severe issue overtime. Hence, the provisioning of landfills should not only stop at its construction but should also encompass proper design and regular maintenance so that it does not

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cause more harm than good. To mitigate these environmental concerns, the focus of landfill design has shifted towards sustainability, aiming to minimize environmental impact throughout the landfill's lifecycle (Parameswari et al., 2021).

Landfills, though often associated with modern life, have a historical background. Their evolution reflects changing societal needs, technological advancements, and growing awareness of environmental impacts.

Some of the earliest evidence of landfills dates back to 3000 BC in Knossos, Crete, where large pits were dug to dispose of refuse. Similar practices were observed in ancient Athens around 500 BC, with the first known waste regulations requiring residents to dump refuse outside city limits (Commercial Zone Products, 2023). During this period, waste management primarily consisted of dumping refuse in pits, ditches, or open areas, often leading to environmental and public health concerns (Rihn, 2021).

The 20th century witnessed a shift towards engineered landfills designed to minimize environmental impact. Compaction, daily covering with soil, and leachate collection became standard practices. With increasing waste generation and limited land availability, landfills face challenges in terms of capacity and environmental impact. Concerns regarding methane emissions, leachate contamination, and land use are driving research into alternative waste management solutions (Rihn, 2021). Significant advancements have been seen in the previous three decades. Separation, re-use, and recycling operations have been effectively adopted in several nations, with the goal of extending the life of landfills. Despite progress toward recycling and incineration in numerous nations, landfilling is a dominating waste disposal technique and will continue to be so in the near and long term (Galvão et al., 2008).

However, the future of landfills likely lies in their integration with other waste management strategies like recycling, composting, and waste-to-energy technologies. Mini-

mizing waste generation and promoting resource recovery are crucial steps towards a more sustainable future (Wikipedia Contributors, 2023).

Solomon and Poulouse (2018) proposed three major landfill types:

- Municipal solid waste landfill (MSWLF); able to hold residential waste. A MSWLF might take additional non-hazardous wastes, like incinerators with integrated reprocessing mechanisms. To reclaim the material, filters are utilised.
- Construction and Demolition Landfills (C&D); collects construction and demolition waste, which consists customarily of excavated debris, roadwork resources, demolition waste, construction/renovation waste, and site clearing rubbish. Industrial solid wastes and hazardous materials are not dumped in C&D landfills unless they are approved to receive such waste and meet certain conditions.
- Industrial Landfills; accept certain industrial effluents. Industrial waste can be summed up as solid waste collected through production or industrial procedures. It also contains byproducts of production. Mining and oil and gas industry waste are not considered to be industrial waste. Landfills for industrial wastes are frequently mono-fills connected to a particular site or company.

In Figure 1, its standard features are showcased.

With respect to a study carried out by Ireaja et al. (2018), the precedence engineered landfills have over dumpsites are outlined.

- It does not contaminate the groundwater and air;
- It keeps odours and unsightliness at bay;
- It is a helpful energy source, similar to methane gas;
- It monitors fly breeding, rat and vermin infestation, and bird clustering for airplanes;
- It provides jobs for low-skilled workers, who are widespread in developing countries;

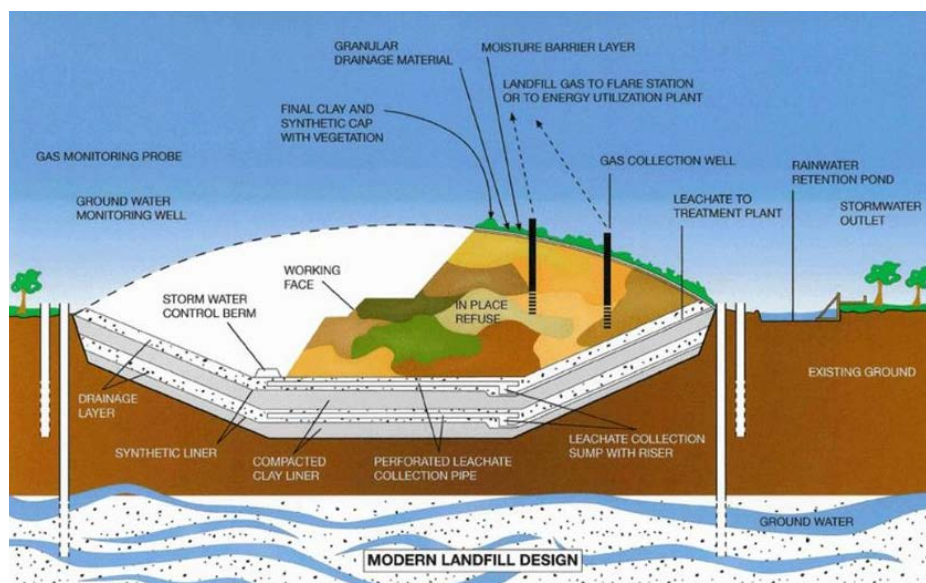


FIGURE 1: Common elements of contemporary landfill architecture (CSIR, 2000).

- It is a large business for investors;
- It is advantageous in waste management.

Furthermore, during the rainy season, public waste dumps encourages breeding for rats, rodents and other disease-carrying organisms, while polyethylene terephthalate (PET) bottles and other items that are not biodegradable from these garbage dumps choke waterways. This can lead to epidemics like lassa fever, which is caused by rodents living in an unhealthy environment, as is the case in Nigeria right now (Ukwaba et al., 2018). Hence, the need for engineered landfills with effective liner systems that are locally suitable and sustainable. To this end, different materials, especially wastes, have been employed within the protective layers of liner systems. This is with the purpose of reducing the skyrocketing degree of environmental wastes and also develop satisfactory landfill technologies. The utilization of certain waste materials in the construction of liners is a promising avenue, which potentially offers a dual benefit of waste management and improved environmental performance (Rubinos & Spagnoli, 2018). Therefore, this mini-review explores the potential of utilizing waste materials in landfill liner construction, contributing to the advancement of sustainable waste management practices. Utilizing wastes as landfill liner materials can contribute to Sustainable Development Goal (SDG) 12: Responsible Consumption and Production (United Nations, 2015), in some ways. Traditional landfill liners often rely on mined resources like clay and specific types of gravel. By finding ways to use treated waste products as replacements, we can conserve natural resources. Furthermore, landfills are taking up valuable space, and finding alternative uses for some waste streams helps reduce the overall amount needing disposal. This translates to less land being used for landfills and potentially even diverting some waste from landfills altogether.

2. SEISMIC ANALYSIS OF LANDFILLS AND LANDFILL LEACHATES

A key factor in the complex dynamic soil-structure interaction challenges including seismic response and earthquake-induced landfill movement is the geosynthetic interface. Feng et al. (2018) analyzed landfill deformation and seismic reactivity while rationally taking into account the liner contact influence. The work conducted adopted a softening-displacement variational elastic plastic constituent structure was created and tested using empirical values from nonlinear shear tests and shaking table validation to illustrate the dynamic friction behaviour of the geosynthetic clay liner interaction under a wide range of functional stress conditions. The shifting displacement and dynamical reactions along the geosynthetic clay liner link are addressed using a two-dimensional in nature time-domain dynamical finite element calculations of a prevalent above-the-ground landfill. These studies include the newly proposed dynamic interfacial framework, as shown in Figure 2. There are frequently large discrepancies when the nonlinear elasto-plasticity and deformations characteristics of the geosynthetic interface are ignored. The study's

findings indicated that the characteristics of the geosynthetic interface have a significant impact on the seismic response and long-term displacement of above-ground landfill. Therefore, careful consideration should be given to the liner layer's design in order to limit the response and deformation brought on by earthquakes.. The dynamic behaviour of the geosynthetic interface should be carefully taken into account when assessing the seismic stability of MSW landfills with potential slippage along the liner interface. Furthermore, when employing simplified dynamic analytic methods for landfill assessment or seismic design, great caution is needed.

When water passes through the landfill, it gathers in the refuse and other water-soluble materials, creating leachate. Water from runoff or even waste itself could be the source of this substance. Risks to the health of people and the surroundings are presented by the landfill's leachate, which has the potential to pollute both groundwater and soil (Onyelowe et al., 2021). Depending on a range of factors, leachate can have distinct constituents. The leachate typically contains humic acid and organic compounds, whereas inorganic components include sodium, cadmium, ammonium, iron, and calcium. For the sake of not deteriorating the environment, especially degrading the soil and groundwater, the leachate from the landfill ought not to spill out. A liner that acts as a leachate barrier must be provisioned in the landfill with the goal of safeguarding against harm to the soil and groundwater supplies (Budiardjo et al., 2019).

3. LANDFILL LINER

Patil et al. (2009) expounded engineered landfill liners as multilayered systems consisting of a hydraulic barrier layer as an important component. This layer's intent is to lessen water percolation through the cover system by restricting water directly and indirectly facilitating drainage or water storage in the underlying layers. Because communities rely on groundwater for drinking water due to increased pollution of the available sources of surface water, effective landfill liner systems are critical in preserving groundwater from leachate contamination and controlling the release of methane and other gases to prevent buildup and potential explosions. Designed landfill's liner system works as a leachate barrier, preventing pollutants from be-

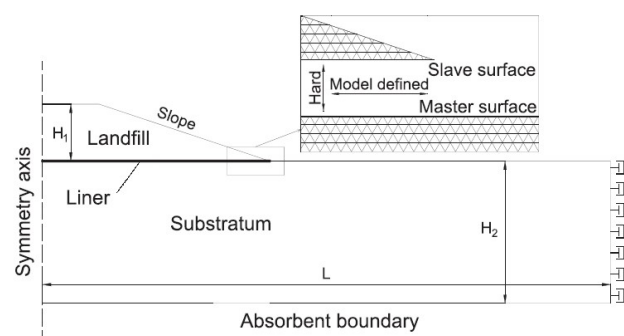


FIGURE 2: Finite element model of a typical above ground landfill (Feng et al., 2018).

ing transported to the pollution-prone surroundings. As a result, one of the most important design issues in a landfill is the liner system (Olaoye et al., 2019).

3.1 Municipal solid waste landfill liners

Du et al. (2009) examined the effectiveness of two Chinese MSW landfill bottom liner system types. The standard landfill bottom liners for municipal solid waste (MSW) were mandated by the Chinese government. Nevertheless, very little study has been done to assess the effectiveness of the typical MSW landfill bottom liners that the Chinese government recommends. The maximum leachate head, the leakage rate, the peak concentration of the intended contaminant in the aquifer beneath the assumed landfill, and the total mass per unit area of the target contaminant released into the aquifer were the criteria used in this study to assess the performance of the two kinds of Chinese MSW landfill bottom liner frameworks – Type 1 and Type 2. In Type 1, a natural clay deposit with a thickness larger than 2 m and a hydraulic conductivity less than 10^{-9} m/s were used as a containment barrier. In Type 2, a composite liner system is used. The thicknesses of the high-density polyethylene (HDPE) geomembrane liner (GM) and compacted clay liner (CCL) are greater than 1.5 mm and 1 m, respectively. Also, the hydraulic conductivity of CCL was less than 10^{-9} m/s. It is also notable that the drainage gravels' grain size surrounding the leachate collection pipes is not a requirement for the Chinese standard. After an evaluation and comparison of the Chinese and German standard MSW landfill bottom liner systems' performance was conducted, it was discovered that the German standard being more strictly prescribed has a maximum leachate head requirement of less than 0.3 m on the liners, and a grain size of the drainage gravels ranging from 16 mm-32 mm around the leachate collection pipes. It is discovered that the Chinese landfill liner systems' estimated maximum leachate head was considerably greater than the German system's. When it comes to reducing the negative effects of landfills on groundwater quality, the Chinese standard landfill liner Type 2 performed almost exactly like the German standard landfill liner in terms of leakage rate, peak concentration, and maximum total mass per unit area in the target contaminant aquifer. In contrast, the Chinese standard landfill liner Type 1 performed less well in this regard. Material differences, leachate head, overall design, etc., are factors that can influence the performance of MSW landfill bottom liners, thus they must be paid attention to.

Engineering professionals have to take into account the hazardous waste conveyance techniques of advection, diffusion, mechanical dispersion, and adsorption when defining the time necessary for breakthrough of MSW landfill liners, as suggested by Shu et al. (2019). In MSW landfills, geomembranes and compacted clay liners are occasionally adopted as bottom liners to reduce the pollution of groundwater from leachate generated by landfill migration. Leachate seepage, nevertheless, has the potential to contaminate the environment by traversing the bottom liner and carrying pollutants. In order to design a landfill, it is necessary to ascertain the bottom liner's breakout time.

This advances future design of these liner systems to be more effective when these outcomes are determined. Also, regulators and landfill developers should pay closer attention to how long each part of the landfill system will last. This will help prevent environmental issues down the line (Rowe et al., 2023).

3.2 Performance of clay liners

Experimental research was carried out by Aldaeef and Rayhani (2015) to estimate how cycles of heat on a daily basis interfere with CCLs' hydraulic efficacy in a simulation resembling a landfill. After being subjected to ambient condition and dehydration cycles, several soil samples were tested for hydraulic conductivity. 30 heat cycles increased CCL hydraulic conductivity by an order of magnitude for low plasticity-index soil materials (PI = 9.5%). Medium (PI = 25%) and high (PI = 37.2%) plasticity materials did not exhibit significant variations in its ability to transmit fluids (hydraulic conductivity) because of the ability to remediate itself. Even after 60 temperature cycles, a barrier covering kept the CCLs hydraulic conductivity constant while reducing the impact of the thermal day-to-day cycles. The low plastic hydrological characteristic of CCLs is greatly altered by wet-dry phases. On the other hand, as demonstrated in Figures 3 and Figure 4, the hydraulic performance of CCLs of medium and high plasticity remained uninterrupted during the course of the test sessions. The report highlighted the requirement for a geomembrane or temporary soil barrier to defend the CCL from atmospheric endangerment.

However, it may not always be the case that high plasticity CCLs would remain unaffected by wet-dry cycles. Melchior et al. (2010) tested the in-situ performance of landfill cover systems in Germany for a span of 18 years. Results from this study indicated that the clay barrier system can indeed be at risk of crack formations due to desiccation and ion exchange. No "self-healing" of the barriers was observed as there was a successive decline in the efficiency of the geosynthetic clay liner system during the first years, having annual leakage rates of 90-220 mm/year which was about 50% of the water that percolates through the soil cover and reaches the drainage layer above the barriers. On the other hand, the composite layer having welded geomembrane performed better during the test period, having an average leakage rate of 2mm/year. It was also noted that the cover thickness is a vital determinant of these outcomes.

If compacted clay liners, which are frequently employed as barrier frameworks for landfills, undergo exposure to the atmosphere for a long time before covering layers are placed on top of them, their hydraulic conductivity may adversely be affected. Considering the presence and absence of geotextile cover, Safari et al. (2014) investigated the fluctuation of the fracture intensity factor in three compacted clay liners while subjected to a yearly cycle of atmospheric conditions. It was found that the desiccation crack severity may be considerably diminished by the geotextile cover placed over the compacted earth. The important finding was that, in every instance, soil layers covered with geotextiles seemed to have kept moisture at the conclusion of

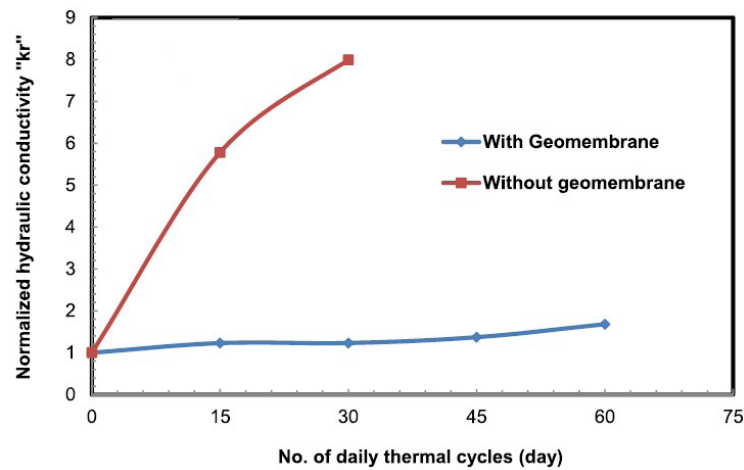


FIGURE 3: Halton clay hydraulic capabilities in the presence and absence of a geomembrane (Aldaef & Rayhani, 2015).

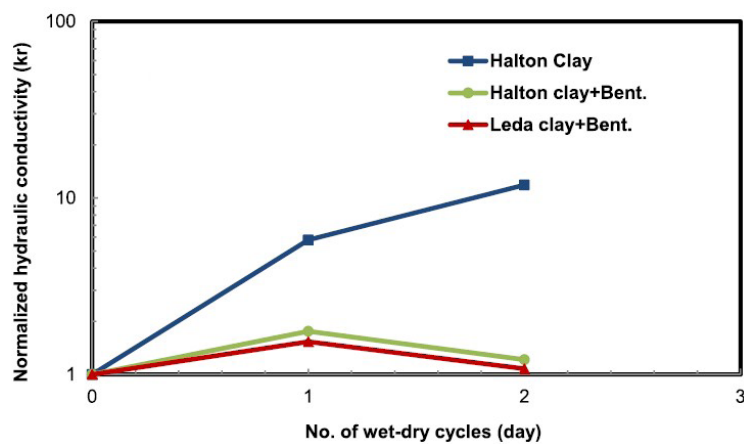


FIGURE 4: Wet-dry cycle influences CCL hydraulic effectiveness (Aldaef & Rayhani, 2015).

the trial, in contrast to significantly drier soil surfaces that were directly exposed to air conditions. This could be one of the causes of the notable impact of geotextile cover on lowering cracking magnitude, combined with the possibility that white geotextile safeguarded the soil surface colder than the surface-exposed topsoil. However, it is important to note that CCL materials tend to shrink significantly when they dry out. This shrinkage can cause cracks to form within the material leading to an increase in hydraulic conductivity. In this case, geotextiles may not be strong enough to restrain the shrinkage forces causing the desiccation cracks (Melchior et al. 2010). This further leads to the conclusion that two criteria determine the continued efficacy of clay barrier coverings; appropriate barrier layer construction to ensure that the field-scale hydraulic conductivity criterion is satisfied and maintaining the protective layer over time to keep minimal hydraulic conductivity levels (Albright et al., 2006). Due to its thickness, CCL has a higher dampening potential than geosynthetic clay liners (GCL) and is maximally inert to any permeating substances that come into touch with it. GCLs have become prevalent due to ease of fabrication and significantly less air space usage (Daniel, 1993; Vishnupriya & Rajagopalan, 2022). Research continues to advance to study and understand the perfor-

mance of clay liners, incorporated with different materials, under different environmental conditions.

4. UTILIZING WASTE FOR GOOD: EXPLORING ALTERNATIVE LANDFILL LINER MATERIALS

In landfills, waste is encapsulated by a liner system at the bottom and a cover system at the top (Solomon & Poulose, 2018). The bottom liner system, which is installed at the base of the containment structure prevents leachate from seeping into the ground and contaminating groundwater. It is characterized by low permeability, consisting of a multi-layered system, often including compacted clay, geomembranes to minimize leakage. On the other hand, the cover liner system protects the landfill from above and manages gas emissions. It is placed on top of the waste materials in a containment structure, reducing infiltration of rainwater, minimizing leachate generation and controls the release of methane and other gases to prevent buildup and potential explosions. Both systems work in tandem to minimize the environmental impact of landfills and protect surrounding ecosystems. Over the years, different materials have been explored for use within the protective layers

of these liner systems. This is in a bid to reduce the ever-growing number of environmental wastes and also develop locally suitable landfill technologies. Some of these materials are predominantly wastes, like fly ash (FA) and rice husk ash (RHA). Alone, they pose no economic value (Ikpe et al., 2020), but have protective potential when utilized within the layers of liner systems. This is given that the reduce, reuse, and recycle operation has applied recent technological developments and improved comprehension of materials, adding financial and commercial worth to discarded matter while also mitigating the negative environmental implications of uncontrolled waste (Igbinomwanhia, 2012; Ikpe et al., 2019). Research have been performed to assess the practicality of using these waste products in landfill liner systems.

4.1 Fly ash (FA)

When coal is burned in power plants, fly ash is created as a byproduct. It is frequently handled like waste and must be discarded the same way as waste from municipalities. Power plant FA make up a large amount of industrial wastes, produced in millions of tons each year (Yadav et al., 2023). Consequently, a great deal of research has been conducted, especially with regard to the viability of using FA as an engineering component (Mollamahmutoglu & Yilmaz, 2001), in an effort to reduce this alarming growth of this waste generated.

A research investigation on the hydraulic performance of FA-Bentonite mixtures in landfill isolation facilities was conducted by Budiardjo et al. (2021). The combination of FA and 25% bentonite, according to the outcomes had the least amount of permeability and satisfied the requirements for CCLs, with hydraulic conductivity (k) values up to 1×10^{-9} m/s. The combination of bentonite and FA reinforced the cohesion links between molecules and boosted stability. The conclusion that followed was that the permeability coefficient of mixed soils could be effectively decreased by combining FA, which was being utilized as a landfill material, with other materials like bentonite. In general, this combination provides substantial strength in compression, lesser hydraulic conductivity, a degree of adaptability, and reduced crack susceptibility (Amina & Ravi, 2017). FA has been applied to blends of cement as well. The 90% FA + 10% cement mixture's hydraulic conductivity was determined to be less than 1×10^{-9} m/s, meeting the requirements for a landfill lining material (Mishra & Ravindra, 2015). Furthermore, FA that had been passed through a 0.075 mm sieve (No. 200) with up to 10% rubber and bentonite included looked to be a good material for making a liner since it fulfilled the most crucial requirement—hydraulic conductivity (Cokca & Yilmaz, 2004). FA is well recognized for its pozzolanic behaviours, which, when cured, triggers it to solidify and get stronger over time (Garg et al., 2020). As a result, the curing time causes the hydraulic conductivity to decrease, thus, improving the stability of the landfill. Furthermore, FA can typically be developed in various ratios with other materials to achieve the acceptable hydraulic conductivity and strength threshold.

4.2 Paper mill sludge (PMS)

Paper mill sludge (PMS) is produced when wastewater is treated in the paper-making process. Techniques for handling and disposing of it raise significant worries. It can be rid of as waste or turned into valuable products, transforming it into a substance with extra worth (Likon et al., 2009). Using it as a material for landfill liners is one option of utilizing it so as to reduce its nuisance to the ecosystem, as approximately 400 million wet tons of PMS are generated globally every year (Faubert et al., 2016).

After evaluating the geotechnical features involved in using PMS as a bottom liner and landfill cover, Balkaya (2019) came to the conclusion that the material's geotechnical qualities make it an appealing replacement for compacted clay in landfills that house MSW. According to Kuokkanen et al. (2008), PMS has appropriate hydraulic conductivity, making it ideal for use as a barrier at the sides and base of toxic waste landfills as well as in the constructed layers of the landfill covering architecture. The combination of PMS and sepiolite has also been investigated. Their aggregation yields an effective void filler which in turn reduces void ratio and hydraulic conductivity, making them suitable for landfill liner design (Thomas & Thomas, 2019). This is also an effective way of ridding the environment of paper mill sludge.

4.3 Waste Foundry sand (WFS)

WFS is the sand that has been used repeatedly in the foundry until it is exhausted and can no longer be utilized to create moulds. It is typically a blend of fine homogeneous sand, bentonite, and other components. Millions of tons of these sands are dumped as waste from foundries each year, which poses a significant disposal issue (Ochepo, 2020, Ahmad et al., 2022). Additionally, studies have been conducted to assess WFS as a potential substitute barrier component in constructed landfills.

In a laboratory investigation conducted by Abichou et al. (2000), foundry green sands were found to meet the hydraulic conductivity demand of less than 1×10^{-9} m/s. WFS has also been incorporated with rice husk ash and fly ash to yield stronger materials with reduced hydraulic conductivity. More cementing gel is produced by this mixture, increasing the amount of bonding in the material's matrix (Ochepo, 2020; Cherian & Abraham, 2020), making it suitable as landfill liners.

4.4 Recycled concrete aggregate (RCA)

The amount of waste generated during building and demolition has increased recently, which is having a negative impact on the ecosystem. The majority of waste produced during construction and demolition (C&D) is still aggregate, accounting for 85% of all waste produced. (Tam et al., 2018). In the building sector, utilizing waste materials like recycled aggregate can have positive effects on the environment and the economy (Wagih et al., 2013). One of such applications is in the design of landfill liners. Vathani and Logeshwari (2023) carried out a study on leachate repellency in landfills using Recycled Coarse Aggregate (RCA) enhanced with paper pulp and adsorbent. The

results here also displayed promising characteristics. This caters for strength of the liner material and also effective disposal of wastes (Xu et al., 2023).

4.5 Waste wood ash (WWA)

Wood ash consists of organic and inorganic residues formed as a result of burning wood and wood products. When wood is burned, an average of 6-10% of its weight is turned into ash (Siddique, 2012). Globally, the production of woody biomass is approximately 4600 million tons per year, out of which 60% is used for energy production, 20% is used for industrial purposes, and the remaining 20% is the primary production loss that decomposes in the field (Tripathi et al., 2019). Ash from wood is a pozzolanic admixture. An alumina and silica-rich substance known as a pozzolana has barely any cementitious effect by itself, but when it splits finely and combined with moisture at room temperature, it reacts chemically with calcium hydroxide to produce compounds having cementitious attributes. In wood-fired power plants, paper mills, and other wood-burning establishments, wood ash is produced as a byproduct of burning. In the future, there will be a greater use of wood in the production of energy since it is an environmentally beneficial and renewable resource. The quantity of wood ash generated will rise consequently. This calls for the need to devise new ways to reduce this increasing amount of WWA occurring in the ecosystem (Ghorpade, 2012; Martínez-García et al., 2022). One of such ways is in the application as landfill liner materials.

Oluremi et al. (2019) looked at the use of WWA as a liner in landfill building on lateritic soil. The results of this study indicate that WWA, with a content of 4%, is an appropriate material for landfill liners since it meets the requirements for both unconfined compressive strength (UCS) of 200 kN/m² and hydraulic conductivity threshold of 1×10^{-9} m/s when coupled with laterite. This is essential for waste confinement operations and will reduce contamination and the detrimental impacts of disposing of wood waste on the ecosystem.

4.6 Rice husk ash (RHA)

Ash from burning rice husk—the outer layer that encircles the rice—in a controlled setting is known as RHA. Around 20 million tons of RHA are produced per year according to Market Data Forecast, 2023. 85-90% of it is made up of amorphous silica. It is known that RHA is a potent pozzolan. When used as a soil stabilizer/admixture in soil stabilization as well as a substitute for cement in concrete mixes, it has had favourable outcomes (Ochepo, 2020). One of its more current uses is as a material for landfill liners.

Onyelowe et al. (2021) examined RHA for its possibility for use as a liner material. It displayed good qualities as a micro-pore filler within soil matrices, which affirms it as an excellent “green” building materials which strengthen the landfill clay liner’s leachate barrier for environmentally friendly construction. According to findings, RHA with a pozzolanic chemical-based modulus of 81.47% can be used in place of cement in the design of landfills and containment facilities that are less toxic and effective. This

will protect the soil, subsurface water, and surrounding environment from carbon emissions and wastewater harm (Onyelowe et al., 2022).

4.7 Bagasse ash (BA)

Bagasse, the leftover material from sugarcane after the juice has been obtained, is often burned in sugar mill incinerators to create steam and power. It then turns into black fiber and ash. There continued to be a lot of bagasse disposals in open spaces, triggering environmental problems. When inhaled in significant quantities, BA causes respiratory diseases because it ferments and decomposes in the open atmosphere. Based on findings by Shahbandeh (2024), Brazil, India, China and Thailand produce 724.43, 439.42, 103.38, and 92.1 million metric tons of sugar cane respectively. These numbers continue to skyrocket, leaving disposal issues for wastes from sugar cane. This gives birth to the need to utilize these wastes especially in the field of engineering. Using BA is essential to lowering the quantity of waste in the natural world. Investigations have been implemented on the practical application of BA, particularly in building, as a substitute material and to address ecological concerns (Kartika et al., 2022). In the aspect of its use as a landfill liner material, BA has proved effective in its application. In waste confinement facilities (covers and liners), laterite modified by a concentration of as much as 12% BA can function as an efficient hydraulic barrier, with the best results occurring at 8% administration, according to Eberemu (2013). A hydraulic conductivity of 1×10^{-9} m/s and a UCS of more than 200 kN/m² were obtained from these BA ingredients. BA decreases the permeability of soil, which indicates that it can prevent leaching out of contaminants and can further prevent contamination of ground water (Jishnu & Mohini, 2020). These findings assist in addressing some of the environmental issues caused by bagasse that comes from the sugar industry and provide an affordable method of disposal in addition to advocating for the use of an alternate landfill lining component.

4.8 Coconut fiber (CF)

The most well-known fibrous byproduct of coconut farming is coconut fibre. At least 30 million tons of coconuts are produced worldwide yearly, and they are widely available in tropical countries’ coastal regions (Gaspar et al., 2020). According to Panyakaew and Fotios (2011), the coconut husk has a high lignin and phenolic content and is made up of 70% pith and 30% fibre. Coconut fiber’s high lignin content makes it incredibly elastic, strong, and decay-resistant. Danso (2017) found that the fibres from coconut husks were more resilient than the fibres from oil palm bark, with a mean diameter of 400 µm and a mean length of 103 mm. Given that waste materials and fibers from nature are affordable, readily available locally, and ecologically friendly, their use to improve the state of soil is advantageous (Kaushik & Singh, 2021). To this effect, the geotechnical properties of coconut fiber, otherwise known as coir, has been looked into for its use as a geotechnical material in soils and landfill liners. Puspita et al. (2023) evaluated coconut fiber and FA-composites for utilization in landfill retention layering. The results here showed that

coconut fiber additions would yield considerable and workable results in terms of strength and hydraulic conductivity. This solves the issue of coir disposal, while providing a sustainable and locally accessible liner technology.

4.9 Cassava peel ash (CPA)

Research on the effects of agricultural solid wastes from cassava on the engineering qualities of soil has been undertaken (Tiza and Iorver, 2016). This agricultural waste ash has recently been investigated as a potential barrier material, with CPA demonstrating favorable outputs. CPA-treated black cotton soil was studied by Adeyemo et al. (2022) as a potential material for hydraulic barriers in MSW containment facilities. CPA passing through 0.075mm sieve was utilized. The findings indicated that black cotton soil with a 16 % CPA concentration successfully meets the design standards needed to be applied as a liner or cover in MSW plants after it produced a UCS value more than 200 kN/m² and a hydraulic conductivity less than 1×10^{-9} m/s. Approximately 40 million metric tons of cassava peels are being generated annually in Sub-Saharan Africa (IITA, 2022). This alarming number can be catered for via utilization of the ash from this waste as a landfill liner material.

4.10 Plastic waste (PW)

Numerous types of solid wastes are generated as a consequence of population growth, fast urbanization, manufacturing operations, and evolving living expenses (Reddy et al. 2020; Kaza et al. 2018) especially plastic wastes. One of the most significant problems the world is currently experiencing is plastic waste, which has far-reaching effects. It can damage ecosystems, cause harm to wildlife, especially marine species, and impair human health. It can be found fouling everything from the highest mountains to the deepest ocean trenches. The issue of plastic waste has been more acute in recent times due to the steady increase in the consumption of this multipurpose material. Since the turn of the century, the amount of plastics produced worldwide has doubled, reaching about 400 million metric tons annually. Plastic items typically last 10 years, but depending on their composition and disposal method, they can take up to 500 years to degrade (Alves, 2024). Since plastic degrades at a slower rate, reducing plastic has become a major concern. Considering there is little recycling or repurposing of this waste in an eco-friendly manner, a significant portion of it ends up as garbage in landfills or oceans. PW-recycling is a more productive technique than landfilling and incineration. Therefore, it has been determined that using PW in geotechnical and civil engineering applications has the greatest potential for large-scale consumption, which can also aid in reducing disposal problems (Reddy et al., 2022). Abd-Aziz et al. (2019) carried out a series of shear strength tests on the application of PW as a liner material and concluded that it possesses considerable shear strength needed for engineering application. This simultaneously caters for the disposal of the increasing number of PW being generated while also proffering an alternative liner material that is sustainable.

4.11 Waste Tire Textile Fibers (WTTFs)

Globally, an estimated one billion End-of-Life tires (ELTs) are discarded yearly, and this waste flow is growing dynamically (Golawska, 2024). The increasing number of tires and automobiles on the market means that disposing of WTTFs is very harmful to the ecosystem. This is because burning them releases harmful gases into the atmosphere and takes up a lot of important landfill space. Since tires burn quickly, release dangerous fumes, and tend to contaminate groundwater and soil, burning them is not an ideal environmental practice (Gheni et al., 2019). Additionally, it is not advised to bury End-of-Life Tires (ELTs) in landfills since they take up a lot of useful space, hold water, provide a home for rodents as well as insects to reproduce, and pose a significant risk of fire (Abbaspour et al., 2019; Thomas and Gupta, 2016). Research has been done on the reuse of waste products generated during the ELT recycling process in the field of civil engineering, even in its use as landfill liner materials.

Narani et al. (2020) studied the use of WTTFs as a landfill liner material and concluded that it is feasible to employ WTTF-expansive soil combinations specifically for municipal solid waste landfills as coverings and impermeable liners. This allows for the improvement of the liner without placing an undue financial burden on the projects, while also addressing the increasing environmental hazard posed by WTTFs.

5. IMPLICATIONS FOR FUTURE RESEARCH AND DEVELOPMENT

It is not a surprise that researchers are actively scouting and testing out new approaches and technologies that can be employed for landfill liner design and construction, especially waste materials. This is because as globalization increases, so does the generation of wastes (Akinwumi et al., 2019). To this effect, there is an increasing need to devise new means to dispose of these waste substances. Their incorporation in the design of landfill liners poses to be one of such sustainable disposal methods on the long run, as research has shown that they are useful, eco-friendly, and cost effective in their application as composite liner materials.

The drive for sustainability in construction has been advancing over the years. Diverse research has been carried out and is still being carried out, with respect to finding ways in which construction can be carried out with the aim of protecting the ecosystem in the most effective ways. One of such area of construction is in the design of landfills and landfill liners, where various assessments have been carried out on potential landfill liner materials that can be incorporated in landfill design, especially the waste materials discussed thus far. However, more research and analysis can be carried out on other waste materials that are locally suitable for construction purposes, with the desire to determine their potential as effective landfill liner materials. Some of these wastes that can be looked into are textile wastes (TW), reclaimed asphalt pavement (RAP), gypsum waste (GW), groundnut shell ash (GSA), coffee

husk ash (CHA), eggshell powder (EP), etc. These materials have been employed for the stabilization of soil for construction purposes and also as concrete admixtures (Oyebisi et al., 2020; Bamigboye et al., 2021), in a bid to discover sustainable alternatives to drive environmentally sustainability (Ofuyatan et al., 2020). They infuse strength gain within the soil matrix due to their pozzolanic nature, which is as a result of the complete chemical process of hydration and cation exchange, leading to the formation of calcium silicate hydrates and calcium aluminate hydrates. These compounds then fill the soil voids, resulting in an improvement in their inter-particle cohesion and microstructural integrity (Edeh et al., 2012; Sathiparan et al., 2023; Dezfouli, 2020.). This same principle must be put in mind during future research and advances carried out in aspects of the design of innovative landfill liner materials so as to aid the discovery of alternative materials that are suitable in strength, hydraulic conductivity and also environmental protection.

6. CONCLUSIONS

The design and utilization of sustainable liner materials in landfill construction plays a crucial role in mitigating environmental and health hazards associated with improper waste disposal. Modern landfills have evolved over centuries, reflecting societal needs, technological advancements, and a growing awareness of environmental impacts. The 20th century witnessed a shift towards engineered landfills, incorporating practices such as compaction, daily covering, and leachate collection to minimize environmental impact.

Seismic analysis of landfills and the management of landfill leachates are essential aspects of landfill design, requiring careful consideration. Proper liner systems are crucial for preventing leachate contamination and controlling gas emissions, especially methane. The choice of landfill liner materials significantly influences the effectiveness of containment systems.

Research has explored various landfill liner materials, including fly ash, paper mill sludge, waste foundry sand, recycled concrete aggregate, waste wood ash, rice husk ash, bagasse ash, coconut fiber, cassava peel ash, plastic waste, and waste tire textile fibers. These materials have demonstrated potential in meeting design parameters for hydraulic conductivity and strength, making them viable options for sustainable landfill construction. The incorporation of these waste materials not only addresses the challenges of waste disposal but also contributes to environmentally friendly construction practices.

While the current research highlights the promising performance of these materials, there is a need for continued exploration of other locally suitable waste materials, such as textile wastes, reclaimed asphalt pavement, gypsum waste, groundnut shell ash, coffee husk ash, eggshell powder, among others. Future studies should focus on assessing the potential of these materials as effective landfill liner components, considering their strength, hydraulic conductivity, and environmental protection attributes. In striving for sustainability in landfill design, ongoing research ef-

forts will contribute to the development of innovative and eco-friendly solutions for waste containment and environmental conservation.

ACKNOWLEDGEMENTS

The authors express their gratitude to the Management of Covenant University and the Covenant University Centre for Research, Innovation and Discovery (CUCRID) for the support and platform provided for this study.

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LANDFILL MINING IN THE FRAMEWORK OF INTEGRATED WASTE MANAGEMENT: CASE STUDY OF AN ITALIAN LANDFILL

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Article Info:

Received:

11 April 2024

Revised:

11 June 2024

Accepted:

22 July 2024

Available online:

2 September 2024

Keywords:

Remediation

Landfill mining

Landfill bioreactor

Sustainable landfill

ABSTRACT

A remediation project involving landfill mining (LFM) was designed and is ongoing at the 40-year-old Villadose landfill in Rovigo, Italy. The original natural clay lining the bottom of the old landfill is being recovered and used as technical material. Meanwhile newly constructed landfill sectors, designed to contain more than four times more waste than previously are being filled with both excavated waste and fresh municipal solid waste. A laboratory scale experiment was conducted to simulate the expected landfill conditions and analyse the effects of different waste fractions on emissions. The fresh municipal waste although pretreated at a mechanical biological treatment plant shows a significant residual emission potential, in terms of TOC, and NH₄. A landfill housing such quality of waste would not be expected to have achieved final storage quality by the end of its aftercare life. The joint disposal of this waste fraction and the substantially degraded excavated waste may contribute to shortening this timeline. This paper reports the landfill mining operation, an assessment of the emissions from the landfill simulation reactors and the implications for the expected landfill behaviour

1. INTRODUCTION

Sustainable landfills play the fundamental role of closing the loop of the materials which cannot be further reintroduced in the circular economy, by giving them back to the Earth (Cossu, 2016) in a controlled, stable, non-contaminating form. Paradoxically, the Not-In-My-Backyard (NIMBY) syndrome is still widespread within communities as landfills are seen to occupy valuable land, making it difficult for policy and decision makers to assign spaces for construction of new landfills. Moreover, finding suitable land that meets regulatory requirements for landfill use can be a significant challenge. On a more positive perspective, the solution to this problem could come from exploiting old landfill contaminated sites in need of remediation. Of the estimated 500,000 old landfills in Europe, approximately 10% meet the EU Landfill Directive requirements and are considered sanitary landfills (López et al., 2018). The landfill mining (LFM) of old landfill sites is often seen as an opportunity both for environmental protection from ongoing or potential contamination and for resource recovery, (Fadel et al., 2013; Jones et al., 2013; Quaghebeur et al., 2013). While material recovery from waste may not always be feasible due to the unrecoverable state of the excavated waste or the cost of recovery, recovery of soil, clay and airspace may almost always be achieved in a LFM project.

Krook et al., (2012) defined LFM as a process for extracting materials or other solid natural resources from waste previously disposed of in the ground. Indeed, LFM was initially conceived for material recovery as is the case of the very first case of LFM reported in scientific literature (Savage et al., 1993). However, the motivations for LFM evolved to include environmental protection and extension of landfill life. For this reason, landfills without constructed barriers, operated before the 1982 Resource Conservation and Recovery Act (issued by the Italian Environmental Protection Agency) can be considered potential sources of contamination and therefore must be remediated (Back et al., 2000).

Many recent reports of landfill mining in the literature are dedicated to evaluating the feasibility of enhanced landfill mining (ELFM) which is landfill mining for resource recovery either by waste-to-material or waste-to-energy (Mankhair et al., 2024; Mankhair & Chandel, 2024; Phyu et al., 2024; Saravanan & Dhinakaran, 2023). The feasibility of resource and energy recovery from landfilled waste is highly dependent on the waste quality, which in turn is affected by the habits of the population, effectiveness of waste segregation, the soil cover layer, the climatic conditions (López et al., 2018). Seemingly successful reports of ELFM are often in fact feasibility studies and they are related to



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Detritus / Volume 28 - 2024 / pages 161-171

<https://doi.org/10.31025/2611-4135/2024.19403>

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landfills with high metal and combustible waste fractions. Such accounts are aimed at reporting physical characterization of the mined waste, grain size composition with relation to age of waste, determination of calorific value of combustible waste fractions, optimization of separation techniques for resource recovery, characterization of plastic waste for utilization as refuse derived fuel (Mankhair et al., 2024; Parida et al., 2024; Phyu et al., 2024; Saravanan & Dhinakaran, 2023; Tao et al., 2024). The LFM report of the Mont-Saint-Guibert landfill in Belgium for example focused on the recovery of material and energy from the fine fraction, defined as soil-like material obtained from mechanical processing that has a particle size range up to 4.5 mm (Parrodi, López, et al., 2019; Parrodi, Raulf, et al., 2019; Vollprecht et al., 2020). The Belgian landfill was used for the disposal of MSW, non-hazardous industrial waste and construction and demolition waste which explains the high exploitability of the fine fraction of waste. They however conclude like several other studies, that the implicated capital and operating expenditures continue to outweigh the revenues obtained by the valorization of recoverable fractions. In Italy, the high velocity railway line currently connecting Milan to Bologna in Italy, was designed to cross a landfill. Raga et al., (2015) report the full-scale LFM project which was funded within the context of another construction project and involving the excavation of a trench 400 m long and 80 m wide to remove approximately 200,000 tons of waste.

Landfills dedicated solely or primarily to the disposal of MSW have a low economic viability for ELM which breeds the reluctance to carry out landfill mining projects. LFM with subsequent volumetric expansion provides a synergistic approach to overcoming the financial hurdles related to the execution of LFM projects. Most old landfills are simply pits which were filled with waste until ground level and then covered with soil. LFM provides an opportunity to construct sustainable landfills with state-of-the-art technology. These new landfills constructed in the space previously occupied by old landfills exploit the pit volume and go up to and beyond 20m above ground level. This increase in landfill volume allows the supplementary disposal of residual waste whose gate fees provide funding for the LFM. Moreover, this practice is a solution to the unavailability of land for new landfills and is applicable in densely populated areas across Europe and other parts of the world.

The case study presented in this report is that of the Villadose landfill in the municipality of Rovigo, Italy where LFM in conjunction with other containment remediation strategies have been underway for the past 22 years. It is a LFM project aimed at remediation and construction of new sectors for disposal of fresh residual waste. According to the 2023 balance sheet records, the gross revenue amounts to approximately € 49.2 million and the total costs, € 49.1 million resulting in a profit of about €100,000. More specifically, the revenue from management of residual waste amounts to approximately € 7.8 million, while the waste disposal and treatment costs € 4.4 million. The state of advancement of the project in terms of remediation effectiveness, recovered material and landfill service life extension is reported. Meanwhile, with regards to the

management of the newly constructed landfill sectors, an assessment is made based on a laboratory bioreactor experiment in which the landfilled waste fractions are characterised and emissions of the different waste fractions are evaluated.

2. MATERIAL AND METHODS

2.1 Site Description and History

2.1.1 Site Description

The landfill under study is located in an open countryside, approximately 1 km from the town center of Villadose, in the municipality of Rovigo, (Veneto Region, Italy). The four landfill sectors: Taglietto 0, Taglietto 1, Taglietto 2 and Taglietto 3, (T0, T1, T2 and T3 respectively), were constructed and operated between 1980 and 1996 for the disposal of municipal solid waste (MSW) and residues from recycling activities classified as non-hazardous special waste. The T0 landfill spans for approximately 5.4 hectares. It is generally bordered by agricultural land, except on the northeast side where it shares a boundary with the T1 landfill, which spans for about 5 hectares. T1 is divided into six major cells – Cells 1,2,3,4,5 and Pipetta which is a colloquial term coined for the shape of this sector. 150 meters northwest from the northern end of T1, are the two more recent landfills, T2 and T3, as well as the storage area for construction materials and the impermeabilized storage area for excavated waste. A site map is visible in Figure 1 although T2 and T3 are not visible in the image as they are located a few hundred metres from T0 and T1. The entire area is intersected by canals and ditches for water drainage, some of which are of considerable size.

Cultivation of T0 began in the early 1980s and was soon exhausted. In 1982, the disposal area was extended by the cultivation of T1. The year of closure is uncertain, but it is likely around 1985 when the landfill was described as “practically exhausted” in a document. T1 like T0 was covered with a layer of clayey soil of about 80 cm and dedicated to agriculture. T2 was constructed as a controlled landfill in 1985. The first 2 ha of the landfill were closed, covered with agricultural soil, and sown with soybeans in 1986. The remaining 4 ha were exploited from 1988 to 1990. From 1991 to 1996, T3 was cultivated and then closed in a similar manner.

2.1.2 History and discovery of contamination

The first concerns about groundwater contamination around the Villadose landfill arose in 1997, during a routine check of the existing monitoring wells. Some anomalies were found in the concentrations of some parameters. Especially problematic were the amounts of Boron, Arsenic and Chlorides, suggesting an ongoing contamination. Further analysis and investigation led the authorities to consider the T0 and T1 landfills as possible sources of contamination and hence prescribe LFM as remedial action.

Continuous groundwater monitoring has been ongoing from 2004 when management of the landfill changed hands from DANECO to Ecogest.srl – now Ecoambiente since 2012. The occurrence of contamination corresponds with the period when the landfill was established under



FIGURE 1: Map of Villadose landfill showing Taglietto 0 (T0), Taglietto 1 (T1) and its landfill cells, the temporary waste storage area the storage area for construction materials and the locations of the pairs of piezometers. T2 and T3 are not visible as they are located a few hundred metres from T0 and T1.

less stringent legislation for environmental protection. There are 11 pairs of groundwater monitoring wells (see Figure 1) that reach into the shallow and the deep aquifer, labelled; 3s-3p, 4s-4p, 5s-5p, 6s-6p, 7s-7p, 9s-9p, 10s-10p, 11s-11p, 12s-12p, 13s-13p and 14s-14p (where s - shallow and p - deep). The stratigraphy of the area where the Villadose landfill is located is such that:

- 0 m to -2 m: Sandy loams (widely used for agricultural activities)
- 2 m to -6 m: Clay and silty clay
- 6 m to -8 m: Sand and silty sand (shallow aquifer)
- 8 m to -10.5 m: Compacted silty clay (locally known as the caranto layer)
- 10.5 m to -20 m: Sand and silty sand (deep aquifer)

The compact clay of the caranto layer was previously considered sufficient for the protection of the deep aquifer. However, geoelectrical surveys revealed the presence of a permeable discontinuity in this layer and microfractures that promote ionic exchange between the shallow aquifer (initially contaminated with leachate) and the deep aquifer.

Additionally, the aquifer has low hydraulic gradients and is subject to flow reversals at certain times, resulting in stagnation. However, the movement of groundwater is generally considered to be South/South-East. Indeed, groundwater parameters from the “upstream” piezometers 3s,3p, compared to the rest of the piezometers indicate that the landfill is the likely source of aquifer contamination.

T2 and T3 landfills, younger than the other two, were considered not dangerous, as their construction had been regulated by more restrictive legislation.

2.1.3 Preliminary remediation measures

In 2002 the Province of Rovigo approved the remediation project of the area. The remediation consisted in the LFM of the T0 and T1 areas. However, the landfill site's proximity to agricultural land necessitated a more rapid and immediate intervention. As a preliminary remediation measure, an embedded bentonite clay retention wall, 50 cm wide and 12.5 m deep, (and therefore past the caranto layer) was constructed around the perimeter of T0 and T1 to isolate them from the surrounding aquifers.

Results of geotechnical studies showed the need to contain the infiltration of leachate into the excavation area from the adjacent portions of the landfill and of groundwater from the bottom. For this reason, steel diaphragm walls were constructed within the areas of the T1 landfill. The diaphragms installed using the “vibrojet” technique to a depth of 12.5m, divide T1 landfill into nine landfill cells namely: Cell 1A, Cell 2A, Cell 1/2B, Cell 3A, Cell 3B, Cell 4A, Cell 4B, Cell 5 and “Pipetta”.

2.2 Landfill mining operation

LFM of T0 began in 2004 with the excavation of waste deposited in the 1970's. A controlled landfill was constructed in compliance with Italian Legislative Decree No. 36/2003 on waste landfilling. The new landfill was used for the disposal of excavated waste as well as of residual waste after on-site mechanical biological treatment (MBT). The supplementary disposal of residual waste provides funding for the LFM and results in thicker landfill cells.

T0 was subjected to a temporary capping intervention in 2013 after closure. The elevation levels of the landfill

were reshaped to regulate the infiltration of meteoric water. T0 is equipped with biogas wells connected to a flare which regularly extracts and burns the biogas generated. Leachate is collected and treated offsite. Geoelectric, geophysical, and geotechnical analyses were carried out to determine the necessary technical characteristics for the realisation of the final stage of remediation which is the capping which is underway at the time this paper is being written.

LFM operations on T1 commenced in 2015 and are ongoing at the time of writing of this paper. Cells 1, 2 and 3 were excavated, constructed and operated between 2016 and 2021; Cell 5 between 2021 and 2023; Cell 4 is under construction at the time of writing of this paper and; Pipetta is still to undergo remediation. Results of geotechnical investigations revealed waste buried at varying depths between -3.5 m and -4.0 m. Excavation was performed to a depth of -4.5 m in some cells and -5 m in others to ensure complete removal of waste and potentially contaminated soil while maintaining a safe distance from the aquifer. Leachate generated during excavation is pumped out and exported for treatment offsite.

As is the case for many old landfills, material recovery is not performed at this site because it is not economically feasible. However, the topsoil and bottom clay are recovered. For each cell, prior to the excavation of the waste, the temporary capping soil is removed. This soil is classified as uncontaminated due to its position above the waste and is stored for reuse as engineering material at the landfill. After removal of the waste, the bottom clay is removed until the design excavation depth is reached. This clay is considered waste until chemical analysis prove that there are no exceedances of threshold concentration values. The reclaimed soil classified as CER 170504 in the European waste catalogue) is stored in numbered heaps of approximately 1,000 m³ each. Each heap is sampled for subsequent chemical analyses, the results of which, for the most part, have shown compliance with the limits set in Table 1, Column A, of Annex 5, Part Four of Legislative Decree 152/2006 and subsequent amendments and additions, as communicated by Ecoambiente S.r.l. to the Province of Rovigo and ARPAV Rovigo. The soils are then transferred to the support area for their use as engineering materials in addition to temporary capping soil.

2.3 Waste logistics, sampling and characterisation

The logistics of the waste are organised such that excavated waste and residual waste pretreated at the MBT plant are immediately disposed of in new landfill sectors. At the MBT plant, the waste is shredded and then sieved in a trommel screen with 100 mm diameter openings, which separates the waste into an oversieve light fraction (particle size > 100 mm) and an undersieve fine fraction (particle size < 100 mm). The light fraction is sent directly to the landfill. The undersieve is biostabilised under aerobic conditions in a procedure similar to composting for approximately four weeks. The resulting material is soil-like with fragments of undecomposed waste and is used as daily cover at the landfill. Based on records of landfilled waste, new landfill sectors typically contain the light fraction of residual MSW, excavated waste, and the biostabilised fraction in the proportions 70%:20%:10% respectively. The two fractions of MBT waste are displayed in Figure 2b and 2c.

Samples of excavated waste (Figure 2a) were collected from two different landfill sectors and at varying depths using an excavator. After removing the topsoil layer, roughly 200 kg waste was placed on a geotextile onsite and homogenised for sampling using the quartering method, then about 60 kg were transported to the lab. Once at the lab it was characterised by means of fractional composition and grain size composition analysis. The waste was shredded to a size $\leq$ 6cm, then quartered further to obtain the samples for the reactors. The sample analysed is assumed to be a representative composition of the old waste. The waste was classified according to particle size (>100 mm, >50 mm, >20 mm and <20 mm) and according to the following composition: plastic, glass, metal, textile, cardboard and wood, stones, and others.

Solid waste analysis were carried out to establish the initial waste quality. Analyses of Total Solids, Volatile Solids and Moisture Content (standard IRSA-CNR 29/2003, vol 1, n. 2090a mod), Total Organic Carbon (with Shimadzu total organic carbon analyser device), and Respiration Index after 4 days (RI₄ with SaproMat respirometer) were performed on the <50 mm fraction of the excavated waste (LFM) and on the following categories of residual waste: Stabilised fine fraction; Paper and Cardboard, Plastic, Textile and Others.



FIGURE 2: a - Excavated waste; b - Light fraction of MBT waste; c - Stabilised fine fraction of MBT waste.

2.4 Lab-scale bioreactor experiment

2.4.1 Experiment setup

Four plexiglass reactors, each 106 cm high with a diameter of 24 cm (Raga & Cossu, 2013), were set up to evaluate and compare the degradation of the different waste fractions. Each reactor was equipped with the following systems: an aeration system composed of a vertical perforated PVC tube running down the centre of the reactor with an inlet on the reactor lid which connects to a Prodac Air Professional pump 360; a leachate recirculation and collection system composed of a system of pipes, taps and a peristaltic pump (Heidolph Peristaltic Pump Drive 5001); a temperature regulating system comprised of a heating jacket used to maintain the internal temperature at $35^{\circ}\text{C} \pm 2^{\circ}$ and Thermo Systems TS100 temperature probes installed inside the reactor; a biogas monitoring system comprised a gas outlet valve fitted on the lid of the reactor which then connects to an LFG 20 analyser. The LFG 20 is a portable analyser which permits simple and precise measurements of CH_4 , CO_2 and O_2 in a sample drawn by an internal pump at 200 mL/min. Figure 3 shows the conceptual diagram and an example of the reactors used.

The reactors were filled with the following waste fractions:

- Reactor 1 - OW: Old Waste excavated from old landfill sectors;
- Reactor 2 - NWL: New Waste Light fraction which is the oversieve from the MBT plant;
- Reactor 3 - NWS: New Waste Stabilised fraction which

is the aerobically stabilized undersieve from the MBT plant.

- Reactor 4 - MW: Mixed Waste which is a mixture of all the fractions.

The ratio of each waste fraction in the MW reactor was NWL 70%: OW 20%: NWS 10% which is the estimated ratio of waste landfilled in the new landfill sectors. The OW and NWL were sorted to remove stones, electronic parts, glass bottles and large metallic fractions. Then they were subjected to size reduction (4 – 6 cm) for homogeneity and to downsize real landfill conditions to laboratory scale conditions. The NWS, having a homogeneous particle size $\leq 2\text{mm}$, remained unaltered. A 10-cm thick gravel layer was placed at the bottom of each column as a drainage layer.

2.4.2 Reactor management

In March 2023, the reactors were filled with the waste samples. The reactors were filled with water by connecting a funnel to the water addition/leachate recirculation inlet until fill capacity of the waste, then with an additional 5 L of water to allow recirculation. The system was kept under anaerobic conditions and recirculation was performed once a week with the help of the Heidolph Peristaltic Pump Drive 5001 which was set to rotate at 60rpm for 20 min allowing 5l of water to be recirculated. Leachate samples were collected and analysed for pH, TOC and NH_4^+ every month. TOC was measured using the Shimadzu TOC analyser which gives results as the mean of a triplicate analysis while NH_4^+ was measured by the UV-Vis spectrometric

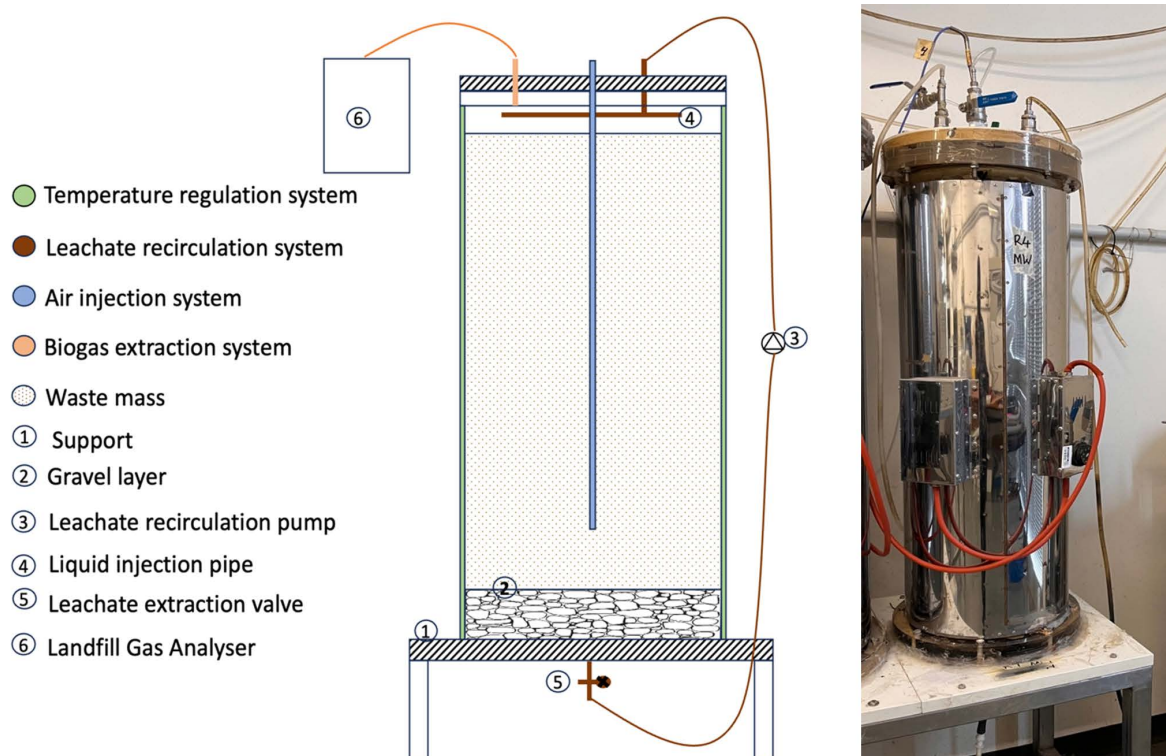


FIGURE 3: Left - Schematic representation of experiment setup; Right - Image of reactor used in experiment.*The air injection system was closed off to maintain anaerobic conditions.

method which gives results as the mean of a quadruplet analysis. The experiment lasted for 281 days.

3. RESULTS

3.1 Waste characterisation results

The material composition of the excavated waste reported as percentages by wet weight with a standard deviation of $\pm 5\%$ is outlined in Table 1. The material with particle size < 20 mm termed undersieve was mostly composed of soil-like material resulting from degradation of organic fractions as well as soil which was used as daily cover. This fraction had a percentage of 28.5%, which is much lower percentages previously reported in other studies, where soil-like fractions range from 40% to 80% (Hogland et al., 2004; López et al., 2018; Quaghebeur et al., 2013; Somani et al., 2018). The water content of the waste likely resulted in the compaction of soil-like particles into thick clayey fragments which merged with the waste to form indistinguishable fractions, classified as others. The excavated waste was not dried as done in studies dedicated to physical characterisation of waste, reason for the low percentage of fines perceived. It is closely followed by plastic with 22.1% and then others or undistinguishable fractions with 21.14%.

Results of the analysis on the excavated waste fractions show that they are in a state of advanced degradation, being over 40 years old. RI_4 value of 2.61 mgO_2/gTS , is only slightly higher than the final storage quality values suggested in the Guidelines for sustainable landfills, issued by the Lombardy region (Cossu et al., 2020), equal to 2 mgO_2/gTS . Average TOC values for excavated waste are lower than those for MBT waste while the plastic and textile fractions of MBT waste contained the highest residual organic carbon.

3.2 Laboratory results

3.2.1 Biogas quality

Biogas was only monitored qualitatively and results are expressed as percentages with a $\pm 1\%$ error margin and are reported in Figure 4. The quantification of biogas was impeded because the reactors were not perfectly airtight

allowing biogas to leak out and some measure of oxygen to seep in.

The biogas generated by OW started off at 11% in terms of CH_4 but immediately dropped to approximately 0.2% and did not exceed 0.9% for the rest of the experiment. The CO_2 was similarly low, attaining a maximum of 14.1% but generally remaining in the range 6.5%-12.5%. This low reactivity is concurrent with the assessment that old anaerobic landfills are not expected to pose a long-term problem in terms of biogas emissions as landfills are expected to have a biogas exploitation potential for a duration of approximately 30 years (Rada et al., 2015).

NWL was principally composed of slowly degrading material such as of plastics and textiles as was reflected in the CH_4 which was generally at varying degrees below 3% and with an average CO_2 14.5%. Correspondingly, the pH remained in the acidic range 5.5-6.1, which is suboptimal for methane generation - in fact, from day 114, onwards, the pH fell below 6 and remained as such.

The highest CH_4 percentage for NWS (51.3%) was recorded on day 67 with a corresponding 31.5% CO_2 . However, between day 74 and day 88, the CH_4 was in the order of 3%-12%. After this, the percentage progressively dropped below 1% until the end of the experiment. A similar decline was observed in the CO_2 . From day 185 till the end, the CO_2 recorded was between 4% and 10%, meanwhile prior to that the average CO_2 recorded was approximately 13% despite the optimal conditions for biogas production. This indicated removal of the residual emission potential by further stabilisation of the previously aerobically stabilized biodegradable waste fraction.

Only mixed waste had biogas with a significant CH_4 and CO_2 percentage, starting off at 80% CO_2 and 6.5% CH_4 on day 24, after which the composition stabilised to an average of 7% CH_4 and 17% CO_2 between day 69 and day 85. Steadily, the composition increased to the maximum emissions of 44% CH_4 and 40% CO_2 for 100 days before progressively to 20% CH_4 and 25% CO_2 around day 238. The decline in biogas composition was accompanied by a similar decline in organic carbon release in the leachate phase as the biological stability was improved over time. From the graph, the NWL and MW have a similar biogas composition

TABLE 1: Composition of excavated waste and particle size distribution.

Sieve diameter (mm)	Excavated waste composition (kg)							Weight (kg)	%w/w
	> 100	%	> 50	%	> 20	%	< 20		
Plastic	6.80	52	3.18	39.2	1.68	10.2	-	11.60	22.1
Glass	-	-	1.11	13.68	5.70	34.67	-	6.81	12.95
Metal	0.05	0.34	1.99	24.53	0.20	1.20	-	2.24	4.26
Textile	0.36	2.75	0.32	3.94	0.22	1.30	-	0.90	1.71
Cardboard & Wood	1.20	9.18	1.06	13.07	0.45	2.73	-	2.71	5.16
Stones	2.20	16.84	-	-	0.00	-	-	2.20	4.19
Others	2.46	18.79	0.45	5.54	8.19	49.81	-	11.1	21.14
Undersieve (<20mm)	-	-	-	-	-	-	14.95	14.95	28.5
Total	13.06	100	8.11	100	16.44	100	14.95	52.50	100.0

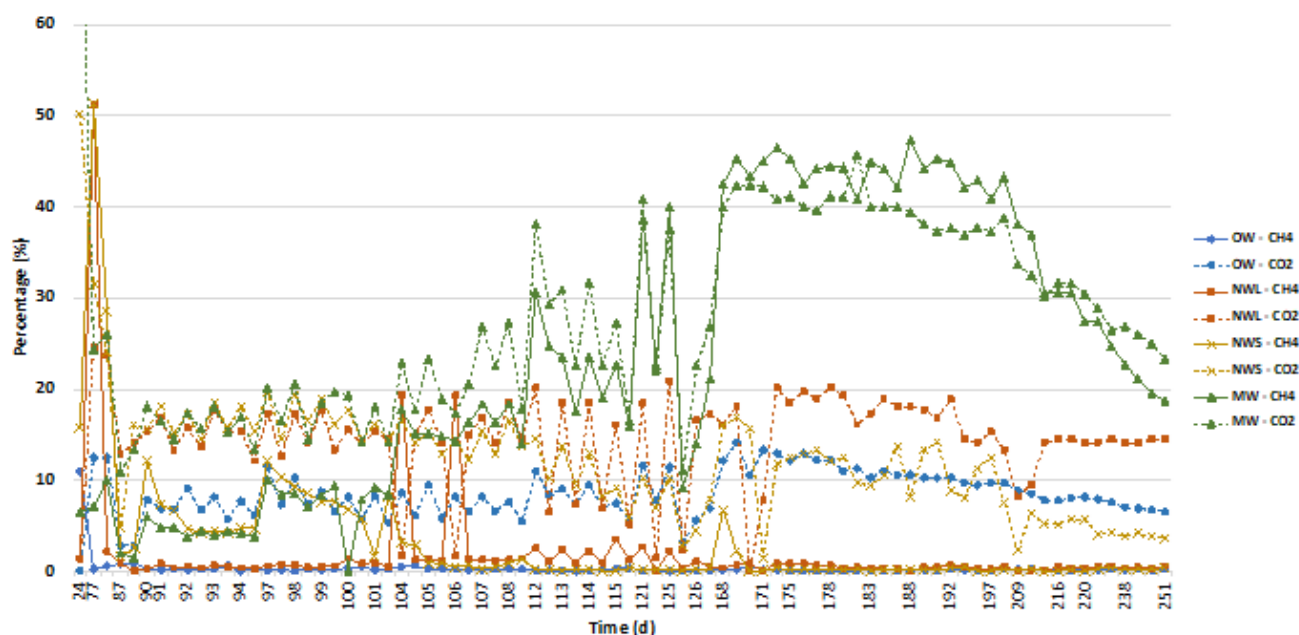


FIGURE 4: Variation in biogas composition. Where OW – Old Waste, NWL- New Waste Light, NWS- New Waste Stabilised and MW – Mixed waste.

trend up until day 110 when MW begins to release gas with more than twice the methane content while NWL proceeds on the same stable slope.

3.2.2 Leachate quality

The evolution of the leachate pH, TOC and NH_4^+ are reported in Figure 5 (a, b and c respectively). The readings have a standard deviation of ± 0.01 , ± 0.006 mg/L and ± 0.02 mg/L respectively. The OW leachate started out at a slightly acidic pH of 6.25, progressed to a range between neutral and moderately alkaline (7.05-7.3) by day 58 and remained as such for the entirety of the experiment. Consistently, the TOC values of the OW leachate were virtually stable throughout the experiment, indicating that the initial reactivity of the OW was quite low and the waste fraction was near stabilisation at the study inception as similarly reported in studies involving excavated waste (Hrad et al., 2013; Warith & Takata, 2004). The NWS fraction predictably behaved in a manner similar to the old waste in terms of pH and TOC as it had been aerobically stabilized prior to the start of the experiment. The NWS nevertheless proved to be more reactive with a TOC going from 3400 mg/L to 1400 mg/L. On the other hand, the NWL fraction proved to be the most reactive waste fraction as its leachate was the most acidic with a minimum pH of 5.51. Likewise, the TOC of the NWL leachate increased from around 2000 mg/L on day 10 to around 10000 mg/L on day 58. The TOC of the MW leachate followed the same trend but progressively dropped to approximately 900 mg/L by the end of the experiment while that of the NWL only dropped to around 6000 mg/L. The reactivity of the MW undoubtedly is a result of the of the NWL contribution (of 70% by wet weight), implying that the enhanced degradation of this waste fraction results from the action of the OW or the NWS or both.

4. DISCUSSION

From the biogas and leachate quality results we can surmise that the OW which was buried for over 40 years, had undergone the peak degradation which is expected to occur in the early stages of the landfill. The leachate quality is characteristic of old landfill leachate, showing more stabilisation in terms of pH than the leachate from a 20-year-old landfill in Ontario, Canada (Warith & Takata, 2004), but nevertheless having similarly insignificant ammonia concentrations. The carbon released via the leachate phase amounted to 0.35 g C which is 0.08 g C/kgDM. Literature values for similar experiments range from insignificant carbon release via leachate (Brandstätter et al., 2015) to 0.7 g C/kgDM (Wu et al., 2016).

The highest NWS CH_4 concentrations were detected within the first 67 days after which concentrations tailed-off to below 1% by day 153 as the MBT likely allowed for a reduction of the non-methanogenic phase and of the landfill gas generation potential (Gioannis et al., 2009). The TOC concentrations decreased from 3400 mg/L to 1400 mg/L, releasing 5.6 g of organic carbon equivalent to 0.08 g C/kgDM like the OW, reflecting the effects of the biological stabilisation. Correspondingly, the NWL fraction unexpectedly contained the highest pollution potential although it was mostly comprised of plastics and textiles which are known to have a slow degradability, not often evident within the timeframe of a laboratory experiment (Elagroudy et al., 2009; Xochitl et al., 2021). NWL and NWS come from the MBT plant, which treats waste belonging to category CER 19.12.12 of the European waste catalogue, described as “other wastes (including mixed materials) from mechanical treatment of non-hazardous waste”. Warith & Takata, (2004) also studied the anaerobic degradation of fresh MSW as discarded in residential

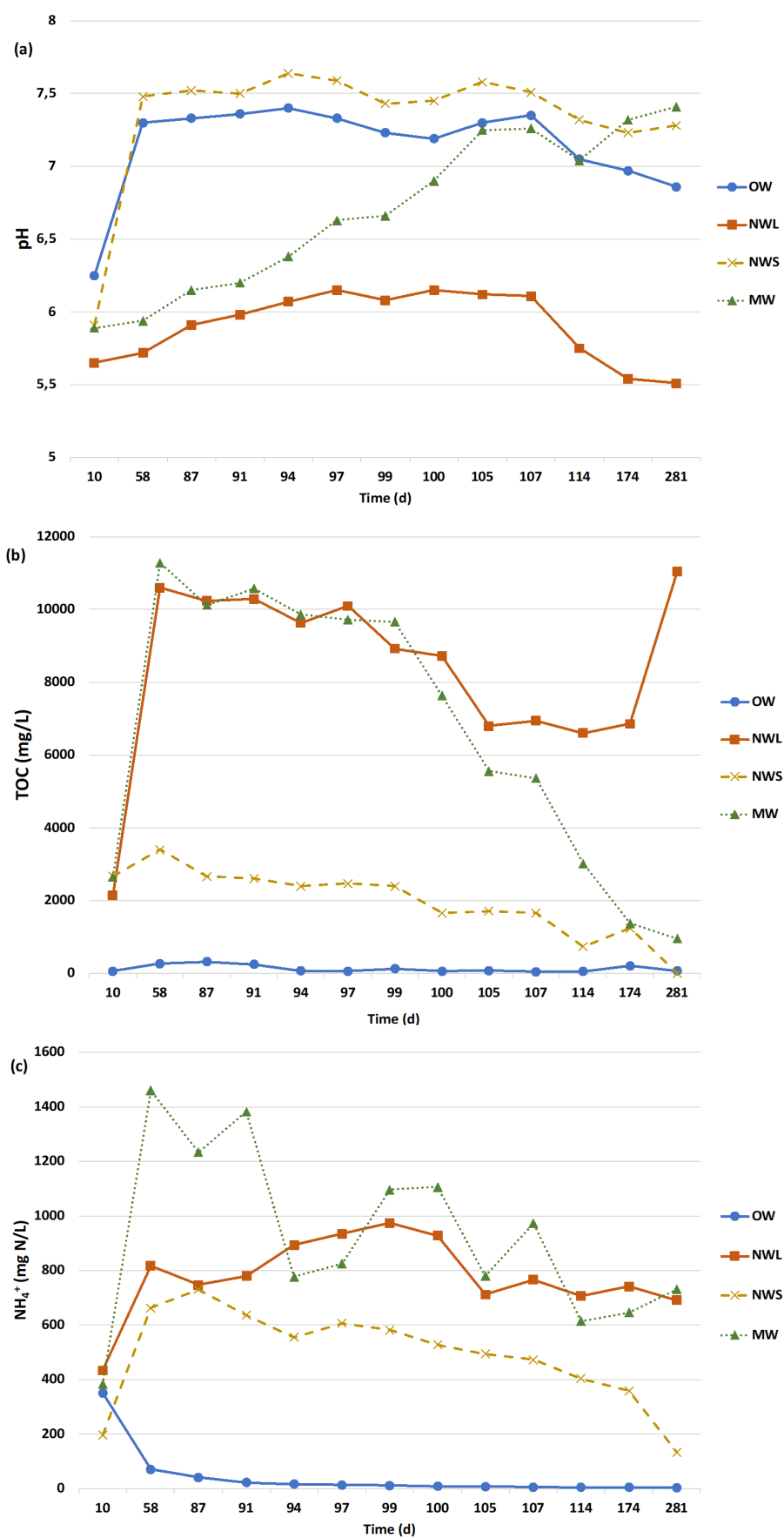


FIGURE 5: Evolution of pH (a); Total Organic Carbon (b); and Ammonia Nitrogen (c) in leachate. Where OW – Old Waste, NWL- New Waste Light, NWS- New Waste Stabilised and MW – Mixed waste.

Toronto and in contrast to the NWL in this study, they obtained a leachate with a neutral pH by the end of the experiment, as well as stable and lower ammonia concentrations within the range of 500 mg/L. In a study by Siddiqui et al. (2013), the biodegradation of the dry fraction of MBT waste with a composition similar to the NWL in this study was evaluated. The TOC concentrations of the leachate towards the end of their study, (about 650 mg/L), was more than 10 times lower than the NWL leachate TOC (> 6000 mg/L), although this could be attributed to the fact that they added anaerobically digested sewage sludge to ensure the presence of viable methanogenic bacteria and to accelerate the initiation of methanogenesis. The acidic pH and the biogas composition in the NWL reactors (av. CH₄ 1.2%, av.CO₂ 14.5%) are an indication that methanogenic conditions were probably not achieved in this reactor. A plausible reason is that plastics and textiles being the principal carbon sources did not provide enough organic matter to support methanogenic bacteria. Oxygen seepage could have also contributed to this phenomenon as regrettably, the reactors were not perfectly airtight throughout the experiment. Regardless, the TOC release from the NWL via the leachate amounted to around 20 g C (4.8 g C/kgDM).

The MW fraction which was mostly composed of NWL material showed a faster stabilisation than the NWL, probably due to the action of the microbial consortium supplied by the OW: indeed, only 15 g C were released through the leachate (2.8 g C/kgDM), 1.7times less than in NWL although the MW released more carbon in the gas phase as seen in Figure 4. One previous research exploited the wide microbial spectrum contained in old waste for the treatment of high strength leachate by passing leachate through an "aged refuse biofilter" (Youcai et al., 2002). In a similar laboratory scale research, leachate from an anaerobically digested mature landfill site was recirculated into a young landfill site to accelerate waste stabilisation (Erses & Onay, (2003). In this study on the other hand, the microorganisms contained in OW which have a strong decomposition capability for refractory organic matter (Youcai et al., 2002), are exploited for the degradation of fresh waste in the MW reactor. The practice at the Villadose landfill of executing landfill mining with the joint disposal of old waste with new waste in new landfill sectors may therefore contribute to accelerating waste stabilisation.

The newly constructed landfill sectors are managed as traditional landfills which pose a challenge in terms of removal of long-lasting recalcitrant organics and ammonia which does not have a degradation pathway in anaerobic systems (Grossule et al., 2018). This management strategy may therefore be an overlooked opportunity to improve leachate quality and shorten the aftercare phase of the landfill by managing them as aerobic or semi-aerobic bioreactor landfills. Aeration has been known to lower the leachate carbon and nitrogen values, improving the leachate quality resulting in subsequent financial savings for secondary treatment. The positive effects of aeration on waste stabilisation have been shown in several studies (Ahmadifar et al., 2016; Huo et al., 2008; Lavagnolo et al., 2018; Morello et al., 2017; Nag et al., 2016) and will be

investigated in a subsequent experiment designed by the authors, to determine the optimal and minimal aeration rate necessary for the enhanced stabilisation of mixed waste.

5. SUMMARY AND CONCLUSIONS

This paper reports the description of a landfill remediation case study in Rovigo, Italy. Essentially, the supplementary disposal of residual waste provides funding for the landfill mining and the potential for volumetric expansion provides an opportunity to construct sustainable landfills with state-of-the-art technology. A landfill simulation experiment was conducted on the disposed waste fractions to analyse the effects of different waste fractions on emissions. Three reactors were filled with distinct waste streams from landfill sectors: Old Waste (excavated from the old landfill), New Waste Light (oversieve fraction from the mechanical-biological treatment plant), and New Waste Stabilised (undersieve fraction aerobically stabilized at the same plant). A fourth reactor, simulating real landfill conditions, was filled with Mixed Waste, containing the three waste fractions in proportions similar to those at the Villadose landfill.

According to the results of the anaerobic lab-scale bioreactor tests on the disposed waste fractions, the leachate quality is characteristic of municipal solid waste landfill leachate with some disparities in the behaviour of the New Waste Light. The Old Waste shows an advanced state of degradation, the New Waste Stabilised is similarly stabilised and these two fractions appear to have an enhancing effect on the degradation of the New Waste Light. The practice of re-landfilling old waste with fresh residual waste therefore accelerates waste stabilisation.

A limitation in this study was the inability to quantify the carbon emitted via the gas phase possibly due to gas losses in the reactors or minor air infiltration preventing the establishment of perfectly anaerobic conditions. The absence of Nitrates and Nitrites was however an indication of predominantly anaerobic conditions. The findings in this research are preliminary. More experiments combining old waste from different MSW landfills and fresh residual waste need to be carried out to better understand the dynamics of the enhanced degradation and to optimize the kinetics, such as by correlating microbial composition with the synergy. This case study is relevant both for old landfills with no viable prospect of resource recovery and for those with a significant recovery potential. Recovery of materials in ELFM creates space which can be used for the re-landfilling of unrecoverable waste in conjunction with fresh residual waste, solving the problem of shortage of land for landfilling. The resulting landfill with enhanced degradation of fresh residual waste may achieve final storage quality in a shorter time than landfills dedicated solely to the disposal of fresh MSW. Aerobic degradation of Mixed Waste within the context of co-disposal of old and fresh waste also has to be investigated and the authors are currently conducting a study to evaluate the extent of this enhancement under aerobic conditions.

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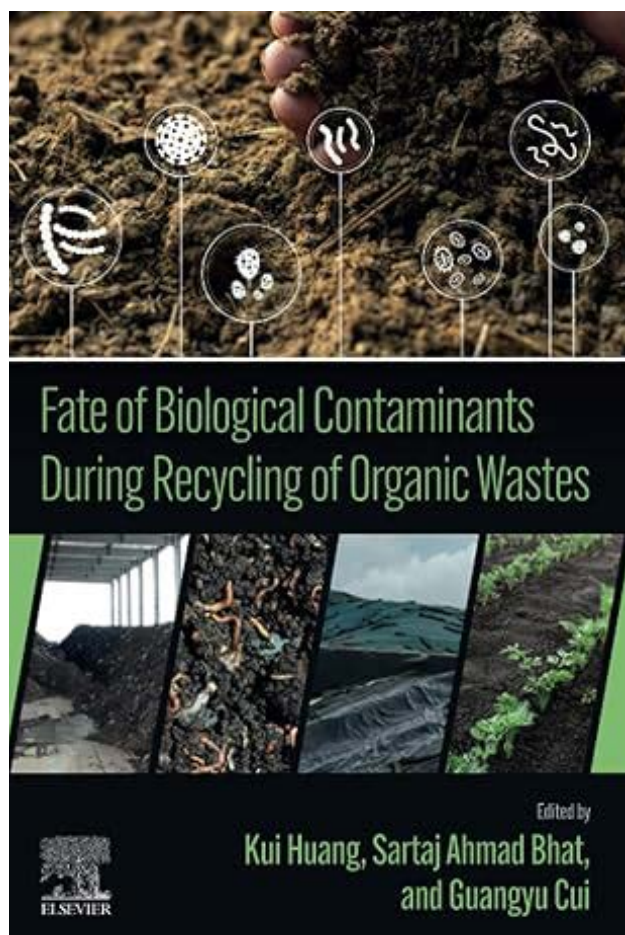
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Extra contents

COLUMNS AND SPECIAL CONTENTS

This section comprises columns and special contents not subjected to peer-review

BOOKS REVIEW



FATE OF BIOLOGICAL CONTAMINANTS DURING RECYCLING OF ORGANIC WASTES

Edited by: K. Kuang, S.A. Bhat and G. Gui

The book "Fate of Biological Contaminants During Recycling of Organic Wastes" presents the results of recent research on the presence of pathogenic microorganisms, antibiotic resistant bacteria (ARBs) and antibiotic resistance genes (ARGs) in different organic waste types and illustrates the most appropriate waste management strategies to abate those microorganisms and immobilize antibiotic-resistant genetic material.

More in detail, the book is divided into 16 chapters. Each chapter is dedicated to a specific topic, namely specific bio-contaminants, specific waste types and/or appropriate consolidated or innovative processes to abate them in the organic waste being treated. Chapters 1, 2, 6, 10, 11, 12 and 14 discuss both the presence of bio-contaminants

in different organic waste types and applicable treatments. Chapters 3, 5, 7, 8, 13, 15 and 16 focus more on innovative processes based on vermicomposting, entomocomposting and combinations with conventional processes. Chapter 9 analyzes waste management during the COVID-19 pandemic and suggests solution to minimize the risk of transmission.

Specifically, Chapter 1 provides an overview on the occurrence of ARGs in organic waste undergoing biological treatments. Food waste, activated sludge and animal manure are reservoirs of ARGs, but also ARBs and mobile genetic elements (MGEs). Conversion of organic waste into fertilizers can spread this problem to soil and then to the food chain again. Anaerobic digestion and composting, are critically reviewed. The results of literature studies – mostly carried out at a laboratory scale though – show that thermophilic conditions reduce ARGs and indicated the potential role of inocula from sewage sludge in ARG enrichment. On the other hand, vermicomposting could be beneficial, since digestion in earthworms' gut can inactivate bacteria encoding ARGs and MGEs. Addition of clay, lignite or biochar during biological treatments can alter the bacterial composition and reduce ARGs and MGEs.

Chapter 2 deals with the fate of pathogens in sewage sludge, which can contaminate soils if sludge is reused in agriculture. After an overview of the most common pathogenic microorganisms in sewage sludge, treatment options to abate their content are described. Thermophilic anaerobic digestion enables a reduction of pathogens of up to 3 log, but is not particularly effective against enteroviruses and parasites, while composting is able to reduce parasites, ARGs and MGEs. Chemical methods (at pH > 12) and physical methods (at temperature > 70°C) allow reducing pathogens but do not stabilize the sludge.

The role of entomocomposting in reducing the content of pathogens in soil is the object of Chapter 3. The chapter presents the main pathogens found in compost and move the focus on insect-mediated composting producing frass fertilizer. Black soldier fly (BSF) larvae and mealworms can accelerate the process, abate pathogens and increase the nutrient content of compost. The mechanism seems to rely on the secretion of antimicrobial peptides by insects. When frass fertilizer is applied to soil, the recognition of chitin and/or chitinase enzymes by the plants' cellular receptors could induce systemic resistance to pathogens. Recent studies have demonstrated that frass fertilizer increases soil fertility and plant productivity.

Chapter 4 focuses on fish waste valorization and the potential issues related to ARGs. In 2018, fish production reached 178.5 million tons, a 2%-4% increase over the

previous decade. However, this surge has led to substantial fishery-derived waste, equivalent to the total fish consumed, posing severe environmental challenges, including antimicrobial resistance. Fish waste includes various components such as scales, bones, and organs, with 30%-45% of fish weight wasted in processing industries. Valorization methods are here reviewed to extract bioactive compounds from fish waste for pharmaceuticals, nutraceuticals, and food technology, promoting environmental sustainability and economic benefits, and degrading ARGs and resistome complexes.

In Chapter 5, vermicomposting is discussed in detail in terms of the fate of biological contaminants. The chapter begins with an overview of the different earthworms used in vermicomposting. The increased microbial biodiversity compared to conventional compost promotes plant growth, mineral solubilization, nitrogen fixation and prevents pathogenic bacteria and fungi. In addition, different earthworms and their gut microbes are reported to effectively degrade organic contaminants (including microplastics) and immobilize heavy metals forming complexes. Gut microbes, especially *Staphylococcus* and *Pseudomonas* strains, also take part to antibiotic degradation if earthworms' exposure to antibiotics is not excessive. Finally, vermicompost is reviewed in terms of plant growth promotion and disease management, thanks to the release of plant hormones and the presence of microbes showing an antagonistic effect on pathogens.

The focus of Chapter 6 is on bio-contaminants and treatment techniques in organic waste. After an overview of the main biological contaminants (including pathogens), the chapter focuses on aerobic/anaerobic biodegradation processes to abate their concentration in waste. The parameters affecting the treatment efficiency are reviewed (biological factors, nutrient availability, oxygen concentration, toxic compounds, moisture content and temperature) and some challenges are highlighted.

The following chapter (Chapter 7) focuses on the advantages of coupling composting with vermicomposting for the removal of both organic and inorganic contaminants. In addition, a strategy based on synchrotron radiation mapping was proposed to identify the binding sites and distribution characteristics of metal and functional groups in earthworms and organic wastes at the micro-scale.

Chapter 8 goes more into details concerning vermicompost and report on the broad diversity of bacterial communities that can be present. In particular, the chapter discuss a critical issue concerning the challenge of how to eliminate harmful microorganisms with ARGs and pathogens, and simultaneously increase the concentration of beneficial microorganisms for biodegradation.

Chapter 9 reviews global research on solid waste management during the pandemic, focusing on China's experience. Since the outbreak of COVID-19 in early 2020, epidemic-related waste has surged, posing infection risks and straining waste disposal systems. This waste, rich in plastics and chlorine, differs significantly from regular municipal waste, impacting incineration and disposal processes. Proper management is crucial to prevent environmental contamination. Various countries have issued guidelines

for managing such waste, tailored to their unique conditions. The chapter highlights the increased attention on waste management in academic publications and underscores the need for effective strategies to mitigate virus transmission through waste.

Chapter 10 focuses on the effects of bio-contaminants from organic waste on soil. The increasing generation of organic wastes, such as livestock manure, food waste, and sewage sludge, is driven by economic growth and population escalation. While over 90% of organic waste products are applied directly to land, composting, anaerobic digestion, and vermicomposting offer sustainable recycling methods. However, bio-contaminants like viruses, harmful bacteria, and antibiotic resistance genes (ARGs) pose significant risks when organic waste is improperly managed. The authors conclude that effective waste management strategies, including waste collection, waste characterization, waste reduction and segregation, labeling and packing, storage, transport, decontamination, recycling, staging, treatment, disposal, tracking, and reporting are crucial to mitigate these risks and minimize soil contamination.

Chapter 11 discusses recent studies on bio-contaminants in soil and proposes future research directions and remediation strategies. Soil contamination by bio-contaminants, including viruses, human pathogenic bacteria, and ARGs, is a global concern, especially in waste disposal sites, industrial wastelands, and agricultural farmlands. Contaminants enter soil systems through wastewater irrigation, sludge application, and livestock manure use. Major contamination sources include landfill leakage, industrial waste discharge, pesticide overuse, and improper waste disposal. These contaminants pose significant risks to human, plant, and animal health. Effective remediation methods, in particular bioremediation, biochar amendment, and nanomaterial application, are essential for mitigating soil contamination.

The widespread use of antibiotics in poultry and livestock farming has led to the presence of antibiotic residues and ARGs in animal manure, which is the subject of Chapter 12. Manure enhances soil quality by adding nutrients, but also poses significant environmental and health risks. Antibiotic residues in manure exert selective pressure on soil bacteria, promoting the development and spread of ARGs. These genes can transfer to human pathogens, posing a public health threat. Effective manure management, including proper treatment and controlled application avoiding excess use, is essential to mitigate these risks and ensure soil and food safety.

In Chapter 13, the book discusses again vermicomposting, but focuses on its role in the removal of ARGs. Earthworms' digestive systems, characterized by neutral pH and low oxygen, help reduce ARGs by altering microbial populations and reducing mobile genetic elements. Vermicomposting has shown a significant decrease in ARGs, particularly from sewage sludge. However, the process's efficiency can vary based on raw materials and composting conditions. Research continues to optimize vermicomposting techniques for better ARG mitigation and environmental safety.

Chapter 14 deals with the pathogens present in municipal solid waste landfills, open dumpsites and leachate. The last two, specifically, harbor pathogens like bacteria, viruses, and fungi, posing risks to human and animal health. Pathogens such as fecal coliforms, enteroviruses, and protozoan parasites proliferate in waste, contaminating soil, water, and air. Vectors like rodents and insects spread these pathogens. Effective waste management, including sanitary landfills and advanced detection methods, is crucial to mitigate health risks and ensure environmental safety. Proper training and protective measures for waste workers are essential.

Chapter 15 presents the bioconversion of hazardous organic wastes using invertebrates, such as earthworms and insect larvae. Conventional agriculture's reliance on synthetic agrochemicals has led to significant soil contamination, impacting soil health and biodiversity. Vermicomposting and insect-based bioconversion are nature-based strategies that transform hazardous organic wastes into nutrient-rich composts. Both methods mitigate soil threats, including organic matter decline and contamination. However, the authors conclude that further research is needed to understand pollutant dynamics and ensure the safety of the resultant organic fertilizers.

The final chapter (Chapter 16) discusses the potential advantages of combining the activity of earthworms with wetlands for the biological treatment of sewage sludge. The authors investigate the impact of plant density on the reduction of ARGs in a vermi-wetland system treating excess sludge. Vermi-wetlands offer a sustainable approach to sludge treatment. Higher plant densities, particularly of *Acorus calamus*, enhance the removal rates of total solids and chemical oxygen demand through improved root interception and microbial activity. However, the effect on ARGs varies, with some genes like sulfonamide resistance genes showing higher removal rates at increased plant densities.

Overall, the book provides interesting insights into the fate of bio-contaminants in organic waste and related management activities. On the side of bio-contaminants, particular consideration is given to antibiotic resistance and

ARGs, while, on the side of management strategies, the role of vermicomposting (alone or in combination with other processes) received most attention. Other processes reviewed are conventional composting, entomocomposting, anaerobic digestion. The book also considers different organic waste types, including food waste, activated sludge, animal manure, fish waste and municipal solid waste in landfills, dumpsites and related leachates. Although the organization of book chapters may appear sometimes confusing, since they do not follow a logical sequence, each chapter provides valuable and detailed information on the specific topic it discusses, paving the way for further research on biodegradation processes and analytical methods.

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Book Info:

Editors: Kui Huang, Guangyu Cui and Sartaj Ahmad Bhat

Imprint: Elsevier

Year of publication: 2023

Page Count: 606

Paperback ISBN: 9780323959988

DETRITUS & ART / A personal point of view on Environment and Art

by Rainer Stegmann

N'Dorah as she calls herself by her artist's name was born in Cameroon and educated in France; she studied Visual Communication and Language Sciences at the Paris XIII University. In addition, she got a master's degree in design. She has worked in the museum industry as well as in fashion and opera. She can look back on a 30-year career as an artist. N'Dorah works with natural and artificial wasted hair and other fibres made usually from nylon and acrylic and uses some kind of glue.

The Ethan Cohen Gallery in New York presented N'Dorah's first solo exhibition "Braid and Weft: Twisting Codes of Beauty and Desire", in the United States in New York in 2022. (<https://www.artsy.net/artist/ndorah>). I selected from her LOST& FOUNDS Contemporary sadness series the art work "Hikkomon", The box of stone from 2017. N'Dorah describes her work as the *"The phenomenon of Hikkomori in Japan is an excuse to talk about the pressure on single women in all big cities worldwide. Not be married, no kids is considered like the worst social situation for them and their families. Some of them stay at home with no interaction, feel ashamed to become at the end grey like the color of concrete, locked in a box of stone, to become VIP,*



very invisible persons. Some of them commit suicide. Made of waste found object, like their young useless (according to them) existence, rejected".

I did not know about the intention of N'Dorah for creating this sculpture when I first looked at the original artwork that she keeps in her Venice apartment. I was very much impressed by the strong expression of grief and sadness that emanates from this curved body full of pain. The person is enclosed in this wooden cage symbolizing for me its own inner world, being left alone with its vast problems, its tragic situation. I am amazed how such a strong expression can be achieved by using these wastes. If you visit her website, you will find many other impressive examples of her work.

Waste hair production is not a real waste issue. A very rough calculation may end up for Germany with 4.000-8.000 t/a waste hair production. Long hair is also subject for reuse. Where does the residual waste hair go? I think in Germany most of it will be collected in the residual waste bin and mostly incinerated. A problem may be the diffusive distribution of short hair particles in rooms and/or the environment. Although not a significant waste problem the art work of N'Dorah makes me also think about waste hair production.

I find the art work "Hikkomon" another very impressive example of waste to art.



In the next DETRITUS Art column, I want to discuss with you to what extent is it acceptable that artwork produces large amounts of waste. I will present two examples: Rudolf Stingel using carpets with oriental design and the covering of buildings made by the artist couple Jeanne-Claude Christo.

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