VOLUME 23 / June 2023

# 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

ISSN 2611-4135 / ISBN 9788862650328
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
- DOAJ Directory of Open Access Journals
- Google Scholar

Partner University:
- Polytechnic Department of Engineering and Architecture, University of Udine, Italy

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
Front page photo credits: Roshni Kumar Alappat, India - Waste to Photo 2017 / Sardinia Symposium

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/
For any enquiries and information please contact the Editorial Office at editorialoffice@detritusjournal.com

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

**www.detritusjournal.com**

VOLUME 23 / June 2023

# 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

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, DOAJ Directory of Open Access Journals 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.

EDITOR-IN-CHIEF:

**Raffaello Cossu**, University of Padova, Italy
E-mail: raffaello.cossu@unipd.it

ASSOCIATE EDITORS:

**Damià Barcelo**, ICRA Catalan Institute for Water Research, Spain
E-mail: damia.barcelo@idaea.csic.es

**Andreas Bartl**, Vienna University of Technology, Austria
E-mail: andreas.bartl@tuwien.ac.at

**Alexandre Cabral**, Sherbrooke University, Quebec, Canada
E-mail: alexandre.cabral@usherbrooke.ca

**Jutta Gutberlet**, University of Victoria, Canada
E-mail: gutber@uvic.ca

**Pierre Hennebert**, INERIS, France
E-mail: pierre.hennebert@ineris.fr

**Ole Hjelmar**, Danish Waste Solutions ApS
E-mail: oh@danws.dk

**Jurate Kumpiene**, Lulea University of Technology, Sweden
E-mail: jurate.kumpiene@ltu.se

**Anders Lagerkvist**, Lulea University of Technology, Sweden
E-mail: anders.lagerkvist@ltu.se

**Michael Nelles**, University of Rostock, Germany
E-mail: michael.nelles@uni-rostock.de

**Abdul-Sattar Nizami**, King Abdulaziz University, Saudi Arabia
E-mail: nizami_pk@yahoo.com

**Mohamed Osmani**, Loughborough University, United Kingdom
E-mail: m.osmani@lboro.ac.uk

**Alessandra Polettini**, University of Rome “La Sapienza”, Italy
E-mail: alessandra.polettini@uniroma1.it

**Marco Ritzkowski**, TuTech Innovation GmbH, Germany
E-mail: m.ritzkowski@tuhh.de

**Howard Robinson**, Phoenix Engineering, United Kingdom
E-mail: howardrRobinson@phoenix-engineers.co.uk

**Rainer Stegmann**, TuTech Innovation GmbH, Germany
E-mail: stegmann@tuhh.de

**Masaki Takaoka**, Kyoto University, Japan
E-mail: takaoka.masaki.4w@kyoto-u.ac.jp

**Ian Williams**, University of Southampton, United Kingdom
E-mail: i.d.Williams@soton.ac.uk

**Jonathan Wong**, Hong Kong Baptist University, Hong Kong
E-mail: jwcwong@hkbu.edu.hk

**Haoran Yuan**, Guang Zhou Institute of Energy Conversion, Chinese Academy of Sciences, China
E-mail: yuanhr@ms.giec.ac.cn

**Liangtong Tony Zhan**, Zheijangu University, China
E-mail: zhanlt@zju.edu.cn

EDITORIAL OFFICE:

**Giacomo Gallinaro**, Eurowaste Srl, Italy
E-mail: editorialoffice@detritusjournal.com

MANAGING EDITORS:

**Maria Cristina Lavagnolo**, University of Padova, Italy
Info from the global world
E-mail: mariacristina.lavagnolo@unipd.it

**Jutta Gutberlet**, University of Victoria, Canada
Grassroots social innovationn
gutber@uvic.ca

**Maria Pettersson**, Luleå University of Technology, Sweden
Legislation and policies on Circular Economy
maria.pettersson@ltu.se

**Alberto Pivato**, University of Padova, Italy
Research to industry and industry to research
E-mail: alberto.pivato@unipd.it

**Alberto Pivato**, University of Padova, Italy
**Claire Gwinett**, Staffordshire University, United Kingdom
**George Varghese**, NIT Calicut, Kozhikode, India
Environmental Forensic
E-mail: alberto.pivato@unipd.it, c.gwinnett@staffs.ac.uk, gkv@nitc.ac.in

**Marco Schiavon**, University of Padova, Italy
Books Review
E-mail: marco.schiavon.2@unipd.it

**Marco Ritzkowski**, TuTech Innovation GmbH, Germany
Portraits
E-mail: m.ritzkowski@tuhh.de

**Rainer Stegmann**, TuTech Innovations GmbH, Germany
Detritus & Art
E-mail: stegmann@tuhh.de

EDITORIAL ADVISORY BOARD:

**Mohammad Alamgir**, Khulna University of Engineering & Technology, Bangladesh

**Luca Alibardi**, Cranfield University, United Kingdom

**Ala’a H. Al-Muhtaseb**, Sultan Qaboos University, Oman

**Sven Andersson**, Babcock & Wilcox Vølund AB, Sweden

**Dezhen Chen**, Tongji University, China

**Christophe Cord’Homme**, CNIM Group, France

**Hervé Corvellec**, Lund University, Sweden

**Frederic Coulon**, Cranfield University, United Kingdom

**Lisa Doeland**, Radboud University Nijmegen, The Netherlands

**Marco Frey**, Sant’Anna School of Advance Studies, Italy

**Dieter Gerten**, Potsdam Institute for Climate Impact Research and Humboldt University of Berlin, Germany

**Apostolos Giannis**, Nanyang Technological University, Singapore

**Valentina Grossule**, University of Padova, Italy

**Ketil Haarstad**, Norwegian Institute for Bioeconomy, Norway

**Uta Krogmann**, Rutgers University, USA

**Maria Cristina Lavagnolo**, University of Padova, Italy

**Jianguo Liu**, Tsinghua University, China

**Wenjing Lu**, Tsinghua University, China

**Weisheng Lu**, University of Hong Kong, Hong Kong

**Claudio Fernando Mahler**, COPPE/UFRJ, Brazil

**Sandeep Panda**, Süleyman Demirel University, Turkey

**Maria Pettersson**, Luleå University of Technology, Sweden

**Marco Ragazzi**, University of Trento, Italy

**Jörg Römbke**, ECT GmbH, Germany

**Silvia Serranti**, University Roma La Sapienza, Italy

**Natalia Sliusar**, Perm National Reseach Polytechnic University, Russia

**Masaki Takaoka**, Kyoto University, Japan

**Elisabeth Tilley**, University of Malawi, Malawi

**Giuseppe Tipaldo**, University of Turin, Italy

**Evangelos Voudrias**, Democritus University of Thrace, Greece

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

Editorial

# ADVANCING BIOENERGY FOR A GREEN FUTURE

A major pivot in achieving a successful energy transition is the transformation from fossil-based to zero net carbon emission sources. Indeed, modern bioenergy (referred to as the low-impact use of biomass, in contrast to traditional low-efficiency combustion techniques) is a pillar for decarbonization thanks to its near zero-carbon footprint. Modern bioenergy has become the most significant form of renewable energy globally - covering 55% of renewables and 6% of the global energy supply (IEA, 2021a). In addition to its renewability, modern bioenergy provides a series of advantages. Among others, it is able to use existing infrastructures, displays flexibility in assisting hard to decarbonize transportation sectors (such as aviation, marine, and trucking), and embraces a multitude of feedstocks, comprising wastes.

In the current catastrophic climate situation, governments and investors have shown increasing interest in bioenergy, and in July 2022 the European Parliament adopted targets to be achieved by sustainable aviation fuels blended with biofuels from biomass waste and residues. However, these efforts are insufficient to ensure we are on track with the zero-emission scenario, with the IEA indicating the need for a dizzying production growth of 16% per year and expansion from 8 to 45% of total biofuel demand derived from biomass wastes (IEA, 2021b).

In order to yield bioenergy, the chemical energy contained in the biomass needs to be converted to thermal energy (through combustion) or energy carriers. A range of different conversion technologies are currently available at diverse levels of technological readiness and based on different principles - operating at low or high temperatures, based on thermochemical or biological processes, and adapted to different substrates, from wood residues to municipal wastes and sewage sludge.

In general, modern biorefineries are increasingly focused on combining various technologies to produce a mixture of compounds with target properties and impacts, rather than individual substances, to reduce overall production costs and increase plant flexibility. Technologies such as combustion and anaerobic digestion are well-consolidated on the market, while other modern approaches, including gasification, pyrolysis or hydrothermal processes, encounter primary barriers in obtaining cost-competitive, high-quality, and quantity products. The fossil fuel market continues to represent the most convenient option for investors and close-minded governments who tend to avoid investing in arguably risky long-term projects. Increasingly global and synergistic efforts are sorely needed, whilst we, as scientists, are called upon to develop strategies to provide market-ready biomass technologies and bio-based products for industrial implementation.

A list of current, promising conversion processes follows.

## Direct combustion

It is largely based on traditional low-efficiency systems (particularly in developing countries) and it is the most widespread technique for biomass conversion worldwide (IEA, 2021a). Modern forms of biomass combustion, including highly efficient domestic pellet boilers and (co)firing in existing coal power stations, remain poorly exploited mainly due to the extremely low cost of coal (50 $/ton in 2020) and scarce attention to the heat sector. The technology is mature and has potential for use in buildings and industries necessitating high temperatures – iron and cement production – and cogeneration plants. Examples of this type of use include the Alholmens Kraft plant in Finland that uses forest residues in a circulating fluidized bed boiler of 500 MW to produce electricity, heat for district heating, and process steam for a pulp and paper mill (Alholmens Kraft Ab, 2023).

## Gasification

With the aim of producing syngas, gasification converts biomass through a partial oxidation under high temperatures (600-1200°C) and often without requiring an external heat supply. On a commercial level, this technology is ready for use, but operating plants are mainly present on a pilot scale operating with relatively small volumes. A series of hurdles represented by high operating costs, vague reliability, handling of by-products (tar in particular), and slight substrate flexibility have hampered widespread use on an industrial level. However, gasification offers innovation potential, particularly when integrated with CHP and district heating, and when purpose-designed for a target substrate. The latter is the case of the Alpine Italian region of South Tyrol, which had 46 small-scale gasifiers fed by wood chips in operation in 2016.

## Pyrolysis

When performed at fast heating rates (>100°C/s), pyrolysis, occurring under an inert atmosphere of 400-700°C, converts the biomass into a bio-oil, with char and gaseous compounds as by-products. Upgrading processes such as hydroprocessing or catalytic cracking should then be applied to convert the bio-oil into a drop-in fuel as the oil has a very high oxygen content (25-40%), is unstable, has

Editor in chief:
Raffaello Cossu
email: raffaello.cossu@gmail.com

Detritus / Volume 23 - 2023 / pages 1-2
https://doi.org/10.31025/2611-4135/2023.18278

a high viscosity, and is corrosive. Pilot and early commercial plants have been set up for heat and power generation, while the full integration of pyrolysis and upgrading in systems that operate in continuous mode is currently at the proof-of-concept stage. Studies aimed at optimizing bio-oil characteristics using catalysts and the scale-up of catalytic reactors are ongoing. The integration of fast pyrolysis with existing petroleum refineries to upgrade bio-oil using petroleum products represents a promising option. For example, the commercial Cote Nord plant in Canada is a 38 ML/year oil facility that converts woody biomass to oil for heating purposes and as feedstock for refinery co-processing. When performed at low heating rates, pyrolysis converts the biomass mainly into a biochar that could play an important role for carbon sequestration through soil application.

**Hydrothermal processes**

An emerging class of conversion process is represented by hydrothermal processes, which exploit the properties of water under high pressure and temperature to convert biomass to a char-like phase (hydrochar), a bio-oil, and a gas phase, the yields of which vary in line with operating conditions. Hydrothermal processes are suitable for the treatment of wet feedstocks (e.g., organic wastes, sewage sludge, algae), avoiding any drying stage. Hydrothermal carbonization is carried out under lower temperatures (180-250°C) to convert biomass into hydrochar for use as biofuel in coal co-combustion, for advanced carbon-based materials, and soil amendments - and an organic-rich liquid phase containing platform chemicals such as HMF. Thanks to the mild operating conditions and relatively moderate costs, this process has a high potential for integration in existing plants in the waste treatment sector. For example, it enhances biogas production and waste volume reduction in anaerobic digestors and wastewater treatment facilities. A commercial development of the process is underway and recently a plant in Mexico City has been set up to treat municipal biowaste to produce hydrochar for use in a coal-fired power plant (TerraNova Energy, 2023). Hydrothermal liquefaction (at 300-400°C) is the wet version of pyrolysis and produces a lower oxygen content bio-oil, which however still needs to be upgraded, e.g. through hydroprocessing. The technological level is demonstrative and significant research is ongoing, with particular focus on the aviation sector. The continuous pilot plant in Denmark (Aalborg) is a pioneering example of the stage of advancement of this process. Under harsher hydrothermal conditions (400-700°C), biomass undergoes supercritical water gasification forming $CH_4$ and $H_2$. However, the industrial implementation remains unlikely in the near future, largely due to high operating costs (Lee et al., 2021).

**Anaerobic digestion**

In the category of biological conversion processes, anaerobic digestion represents a mature technology which is widely adopted for wet biomass waste to produce biogas. After purification, biomethane is obtained, and an increasing number of policies support its injection into the methane grid and its usage in the transport sector. In this regard, in 2022, a European industrial partnership was launched to support the achievement of targets established by the REPowerEU plan for biomethane production (35 bcm in 2030).

**Conclusions**

The world has an impelling need for clean energy and a circular development model: the conversion of waste biomass into bioenergy meets this need. A large range of technologies are available for use, each featuring its own strengths and weaknesses. Different technologies suit different substrates and purposes. Combustion can convert very heterogeneous substrates, even municipal solid wastes, and is reliable and consolidated. One big limit is the final product: heat, usable as it is or requiring further conversion stages with their own complexity and efficiencies. From the perspective of bio-oil production, the choice between fast pyrolysis and hydrothermal liquefaction highly depends on the initial substrates – dry biomass or wet substrates, respectively. Hydrothermal carbonization converts wet heterogeneous biomasses into a carbonaceous phase, whose use is still limited by its quality and scarce attractiveness for the market. Finally, gasification is relatively inflexible for the range of treatable biomasses but produces syngas for several purposes. Therefore, technological response to the problems associated with climate change clearly cannot be univocal. Conversely, political response should be immediate and unambiguous with clear-cut and decisive choices supported by technical expertise (of scientists and technologists) and appropriately communicated to the population.

There is no more time to waste: bioenergy represents a decisive path for a better and sustainable future.

Giulia Ischia, Filippo Marchelli and Luca Fiori *
*Department of Civil, Environmental and Mechanical Engineering, University of Trento, Trento, Italy*
** luca.fiori@unitn.it*

## REFERENCES

Alholmens Kraft Ab, 2023. Alholmens Kraft [WWW Document]. URL https://www.alholmenskraft.com/en/home (accessed 6.15.23).

IEA, 2021a. Key World Energy Statistics 2021 – Statistics Report. IEA Publ. 1–82.

IEA, 2021b. Net Zero by 2050: A Roadmap for the Global Energy Sector. Int. Energy Agency 224.

Lee, C.S., Conradie, A. V., Lester, E., 2021. Review of supercritical water gasification with lignocellulosic real biomass as the feedstocks: Process parameters, biomass composition, catalyst development, reactor design and its challenges. Chem. Eng. J. 415, 128837. https://doi.org/10.1016/J.CEJ.2021.128837

TerraNova Energy, 2023. TerraNova Energy [WWW Document]. URL https://www.terranova-energy.com/en/applications/ (accessed 6.15.23).

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# CO-COMPOSTING OF SAWDUST WITH FOOD WASTE: EFFECTS OF PHYSICAL PROPERTIES ON COMPOSTING PROCESS AND PRODUCTS QUALITY

Hadi Bello [1],*, Jamiu Olamilekan Ajao [2] and Nusirat Aderinsola Sadiku [2]

[1] *Faculty of Engineering and Technology, Agricultural and Biosystems Engineering, University of Ilorin, Saboline Road, Ilorin, Nigeria*
[2] *University of Lagos, Akoka, Lagos, Nigeria*

**Article Info:**
Received:
21 August 2022
Revised:
21 April 2023
Accepted:
22 May 2023
Available online:
15 June 2023

**Keywords:**
Composting
Sawdust
Food waste
Physical properties
Bulk density

## ABSTRACT

Sawdust and food waste have been part of solid organic waste causing great environmental pollution. Composting is a popular utilization method of converting waste like sawdust and food waste to sanitized and stabilized soil amendment. Unfortunately, many composting systems failed due to a dearth of information as a result of little or no scientific research focused on the effects of the physical properties of composting materials on the composting process. To fill this knowledge gap, three composting experiments of food wastes mixed with sawdust at ratio 20:80, 30:70, and 40:60 and compacted to different initial bulk densities of 15, 20, and 25 kg/m$^3$ respectively was carried out to explore the effect of physical properties on composting of sawdust with food wastes. Physicochemical parameters monitored include bulk density; porosity; particle density, temperature, moisture content; pH, and electrical conductivity (EC). The highest temperature (65.3°C) was recorded by trial 3 while trial 1 recorded the lowest temperature (49.3 0C). Among trials 1, 2, and 3, the maximum pH (9.2) and EC (5.1 mS/cm) were observed in compost trial 3. Additionally, the lowest pH (5.3) and EC (1.4 mS/cm) was observed in trial 1. Trial 3 had the highest percentage finest and lowest fibrosity content. A significant increase in bulk density, porosity, and particle density was observed in the three compost trials. The compost's bulk density of (25 kg/m$^3$) in trial 3 was observed to attain maturity and stability as compared with trials 1 and 2.

## 1. INTRODUCTION

A large amount of waste generated from agriculture and food waste is one of the global serious issues (Bello et al., 2021). The rate at which solid organic waste is being generated worldwide due to the rise in population and industrialization is alarming (Wu et al., 2014). Many of these wastes are disposed of untreated and it has caused environmental and health challenges (Lukashe et al., 2019; Sukholthaman et al., 2016, Awasthi et al., 2014;). Household wastes are mixed with other wastes and are disposed of at dumpsites without any efficient material or energy recovery (Oudal et al., 2016). Recently in Nigeria, there is an increase in demand for wood for furniture and other construction purposes, as a result, a considerable amount of sawdust from timber is generated from sawmills. South-west Nigeria alone with over 10,000 sawmills is currently processing over 500,000 logs of wood per year, with about 50-55% as waste informs of sawdust (Adegoke et al., 2014). As a result, a huge landfill of sawdust is being created at sawmill and it poses a threat to humans and the environment. Getting rid of sawdust requires maximum operational cost, as a result, many were dumped to form sawdust piles and many were burnt regularly. Abdul-Halim et al. (2019) reported that improper disposal and or indiscriminate burning of biomass are responsible for depleting the quality of air, contributing to an increase in greenhouse gas emissions, and significantly contributing to global warming, climate change, a source of contaminant to drinking water, soil pollution (Hwang et al., 2020) and threat to the environment and human health. Therefore, the need for more effective waste management and a reasonable plan should be adopted to overcome this environmental concern (Moh et al., 2017). Instead of burning sawdust as normal practices in some advanced Nations and due to its high carbon content (Bello et al., 2021), energy recovery through conversion of this ligno-cellulosic materials into compost could assist in soil carbon sequestration which has been widely adopted in agricultural practices.

Concerning food waste, it forms a major threat global-

* Corresponding author:
Hadi Bello
email: hadibello7@gmail.com

Detritus / Volume 23 - 2023 / pages 3-15
https://doi.org/10.31025/2611-4135/2023.17276

ly because it increases daily due to the increase in human population and improvement in the global economy. FAO statistics report of 2020 indicated that about one-third of food produced is wasted globally (FAO 2020) and Nigeria alone contributed about 32 million tonnes of wasted food (Adebayo et al., 2020). However, food waste is becoming an environmental challenge that threatens man and environmental resources (land and water resources) if it is discharged into the environment without prior treatment. It may cause pollution and harmful effects on human and animal health (Ghosh et al., 2019). It is however important to find effective management of these wastes to promote the effort concerning the development of a sustainable society.

Research has proved that organic waste products can be recycled using different technologies such as aerobic digestion, composting, fermentation, refuse-derived-fuels, and gasification (Nizami et al., 2017; Mashat, 2014; Kelleher, 2007). Composting has advantages over other methods and it is also considered a sustainable treatment for converting organic wastes into valuable products. Converting waste to valuable products through composting is eco-friendly, in addition to its cost-effectiveness, the valuable end product among others makes composting the most widely adopted method (Al-Rhumaihi et al., 2020). It is also considered the best option for recycling waste without causing environmental hazards (Calaby-Floody et al., 2019, Chen et al., 2018). Composting is gaining more attention as many developed nations have adopted composting as a method of recycling and processing waste. In Europe, over 4 million tonnes of organic waste were recycled by more than 124 compost facilities. Also, countries like; Holland, Spain, and France composted 24, 33, and 14% of the total waste generated in the year 2005 alone (Kelleher, 2007). Composting by-products has been extensively used as organic fertilizer to replenish lost soil nutrients (Wang et al., 2019) and as bioremediation to remove soil organic contaminants (Chen et al., 2015; Purnomo et al., 2011).

Composting converts organic waste into a hummus-like organic product through microbial decomposition under aerobic conditions to produce quality organic soil conditioner (Fernandez et al., 2014; Hemidat et al., 2018; Bao et al., 2016). The composting process starts with the mixing of an organic substrate which is the prerequisite for every composting management condition (Cao et al., 2020). However, setting up good management composting system requires a good knowledge of the physical properties of the substrates and the bulking agents. Knowing this could necessitate efficient maintenance and run of a composting plant. Inadequate information about the physical properties of compost material has made many composting systems fail. It is, therefore, necessary to have vital information about the physical properties of the materials involved and understand the composting system.

Composting needs oxygen and moisture for microbial activities (Assandri et al., 2021). The distribution of moisture, oxygen, and temperature within compost is an important factor in maintaining aerobic conditions during composting. The movement of moisture and air across the compost is greatly affected by the geometry and arrangement of the bulking agent where physical properties play a major role (Orthodoxou et al., 2015; Agnew et al., 2005). Moisture and air are required in moderate proportion during composting. Too much moisture and air could lead to excessive cooling and could prevent the compost from reaching the thermophilic condition that is necessary for optimum decomposition and sanitization of the matured compost. Also, inadequate moisture and air in the compost could result in to decrease in oxygen availability to microorganisms and heat evenly distributed across the compost matrix and this could lead to anaerobic conditions. Moisture and air transfer in the composting system are greatly controlled by the physical properties of the substrate.

Most of the physical properties of composting materials that attracted great attention are bulk density, porosity, and particle density. Bulk density of compost is the ratio of the mass of composting materials to the volume of the composting materials and it determines some mechanical properties of the compost such as strength, porosity, and ease of compaction (Mayur et al., 2018). The bulk density of compost determines how fast or slows the degradation of the compost. Due to this, knowing the bulk density of compost is an important requirement for designing an efficient compost system.

## 2. THEORETICAL RELATIONSHIP OF PHYSICAL PROPERTIES IN A POROUS ORGANIC MATRIX

When forming a compost pile, the physical properties of the composting mixture must be considered for optimal performance of the moisture content, and carbon-to-nitrogen ratio, and to provide a favourable condition for microorganisms (Mayur et al., 2018). Mayur et al. (2018) have identified some of the important physical parameters that affect the optimal performance of the composting process such as bulk density, moisture content and air-filled-porosity or free air space. As soon as the compost pile is formed, difficulties are often observed because the effects of some physical properties are always ignored or information about it is not available. For example, bulk density plays a crucial role in the strength and porosity of the compost pile. If the pore space gets filled with water in the presence of high moisture content, then there would be an increase in air space resistance, which results in oxygen deficit in the pile, and anaerobes started dominating aerobes. Bulk density is the ratio of the mass of compost to its volume. Its mathematical expression is kilogram per cubic meter (Kg/m$^3$) (Equation 1).

$$Bd_w = \frac{mass\ of\ the\ material}{volume\ of\ the\ bin} \tag{1}$$

$$BD_d = Bd_w\ x\ \frac{100\ -\ \%\ moisture}{100} \tag{2}$$

Another physical parameter that determines the distribution of air and moisture across the compost matrix is porosity. Porosity or air-filled porosity as popularly called in compost literature can be expressed in terms of bulk density and moisture content as described in Equation (3).

$$Porosity\ (\eta) = \left(1 - \frac{Y_{wet}}{P_d}\right) \quad (3)$$

where:

$Y_{wet}$ is wet bulk density (Kg m$^3$)

$P_d$ is the particle density

Many studies have been carried out on composting sawdust with food wastes or with other substrates (Jae-Han et al., 2020, Zaihua et al., 2020). These studies explained the efficiencies of unconventional bulking agents in composting food waste and end product (Jae-Han et al., 2020), about biological parameters such as oxygen uptake rate, carbon dioxide ($CO_2$) evolution rate, and Physico-chemical properties such as moisture content, volatile solids, C/N ratio and heavy metals (Singh and Kalamdhad 2013a; Nayak et al., 2014). However, little scientific data exist on the best physical properties of composting sawdust with food waste. This study was designed to investigate the effects of physical properties of composting materials on composting process during the composting of sawdust with food waste as well as to evaluate the best bulk density for composting sawdust with food waste.

## 3. MATERIALS AND METHODS

### 3.1 Experimental materials

*3.1.1 The compost bin*

60 liters of non-biodegradable plastic containers were used in this study (Figure 1). The inner diameter of the container was 290 mm while the height was 380 mm. Holes of 3 mm diameter separated by 10 cm were drilled on the side and bottom of the container corresponding to 10% surface porosity, for proper aeration and the drainage of the leachate during the composting.

*3.1.2 Food waste and sawdust*

The food wastes were collected from the University of Ilorin canteens and were made up of leftover cooked rice, bread, and waste vegetable. The bigger food particles were cut to smaller sizes < 1.5 cm. The sawdust was from a soft tree (Malaina tree) and was packed from a local sawmill. It was sieved by a 5 cm aperture size sieve to obtain the size of the same material. The two were then mixed at different proportions with a spade and were then loaded into bins. Three different composting mixtures at three different initial bulk densities were formulated as shown in Table 2.

*3.1.3 Characterization of the raw materials*

The composting materials have different properties as shown in Table 1. The moisture contents were found to be approximately 10.23% for sawdust and 64.23% for food waste. In composting experiment, the total organic carbon and nitrogen content of the materials is the nutritional characteristics of the composting materials, the ratio of the two (C:N ratio) is used to assess the nutritional balance for the microorganism. However, the C:N ratio of sawdust was 49.59, while that of food waste was 34.95. the C:N ratio of sawdust was slightly higher than that of the food waste. In addition, the two composting materials were slightly acidic with pH of 5.6 and 5.3 for sawdust and food waste respectively. The electrical conductivity of the composting materials was within the range of 7.55 dS/m for sawdust and 41.63 dS/m for food. The higher electrical conductivity recorded by food waste may be because of some salts present in the food materials. Fibrosity content was determined for sawdust only and was found to be 89.0%. Bulk densities for both materials were 15.12 and 10.23 Kg/m$^3$ respectively.

**TABLE 1:** Basic characterization of raw materials used in the composting experiments.

| parameters | Sawdust | Food waste |
|---|---|---|
| TN (%) | 1.62 ± 0.16 | 1.54 ± 0.29 |
| TOC (%) | 80.34 ± 1.47 | 53.83 ± 0.20 |
| C/N | 49.59 | 34.95 |
| pH | 5.65 ± 0.11 | 5.36 ± 0.47 |
| EC (dS/m) | 7.55 ± 0.47 | 41.63 ± 0.65 |
| Moisture content (%) | 10.23 ± 0.23 | 64.23 ± 0.13 |
| Fibrosity content (%) | 89.0 ± 0.02 | * Nd |
| Bulk density (Kg/m$^3$) | 15.12 ± 0.01 | 10.23 ± 0.14 |

*Nd = not determined*

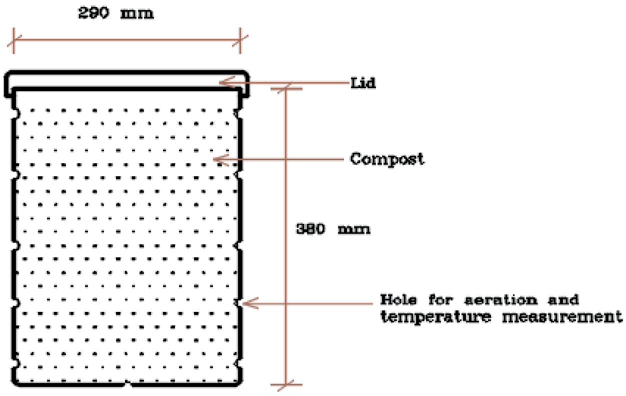


**FIGURE 1:** Schematic diagram of composting system.

**TABLE 2:** The initial experimental conditions of each of the composting mixture.

| | Composting mixture | | | |
|---|---|---|---|---|
| Mixture | Sawdust (%w/w) | Waste food (%w/w) | Initial moisture Content (%) | Initial bulk density (kg/m$^3$) |
| 1 | 80 | 20 | 60 | 15 |
| 2 | 70 | 30 | 60 | 20 |
| 3 | 60 | 40 | 60 | 25 |

## 3.2 Experimental setup and monitoring process

### *3.2.1 Bulk density*

Equation 1 was used to measure the bulk densities of the composting experiments. The bins were filled with a mixture of composting materials previously mixed at different proportions. Each bin was moderately compacted from predetermined heights of 100 mm, 150 mm and 200 mm respectively. After compaction, the procedure was repeated to fill the bin to the desired level and bulk densities. At the end of the process, each experimental mixture has the initial bulk densities of 15 kg/m$^3$, 20 kg/m$^3$, and 25 kg/m$^3$ respectively. The initial moisture contents of the experiments were set to 60%.

### *3.2.2 Physicochemical analysis during the composting period*

The three composting bins were veered weekly. The maturity indexes were also measured weekly till the compost was matured. The temperature was recorded three times a day; at 8 a.m, 3 p.m, and 8 p.m, and a Reotem analog compost thermometer was used. Also, compost samples were oven dried at 105°C to constant weight to ascertain their moisture content.

0.050 Kg of compost were randomly picked from different parts of the bin and blended to earn 0.15 Kg weekly which was divided into three. For pH and Electrical conductivities (EC) determinations, 0.050 Kg of the blended compost was used. The pH and EC were measured by mixing the known sample with 10 cm$^3$ of distilled water and then shaking. A pH meter (pH-3C, Shanghai, China) was inserted to measure pH, and Electrical Conductivity (EC) was also determined using a conductivity meter (LeiCi, Shanghai, China). The method described by Wang et al. (2021a) was used to measure the total organic carbon (TOC). Total nitrogen (TN) was measured using the Kjeldahl method (Cao et al. 2018) and the ratio of TOC to TN gives the C: N ratio. The remaining 0.05 kg compost sample was used to measure the bulk density and moisture content of the sample throughout the composting period.

The free air space or porosity of the compost was measured from the bulk density and particle density of the compost mix using Equation (3). Bulk density was measured using a plastic container whose volume is approximately 50 cm$^3$. The plastic container was filled to one-third height and gently tapped on the plain surface to eliminate voids; then, filled up to two-thirds and after that up to the top brim of the container. Bulk density was calculated by dividing the mass of the compost by the volume of the plastic container. A pycnometer was improvised using a plastic bottle having a screw cap to measure the particle density of the compost material. A hole of 2 mm diameter was drilled on the cap. Small-plastic tubing was attached to the hole with about 30 mm length of tube projecting into the bottle when covered with a cap. The rest of the tube was bent on top of the cap and conducted excess liquid out when the bottle was filled with liquid. The lower density of compost makes distilled water unsuitable to use as displaced fluid to measure the density of the compost material; therefore, kerosene was used as a reference fluid because of its lower density following the Mayur's et al. (2018) method.

## 3.3 Analysis of the final products

The physical, biological, and chemical properties of the final compost obtained were analyzed. The chemical properties of the final compost assessed include; EC, pH, Cation exchange capacity (CEC), C: N ratio, Phosphorus, and Nitrogen contents, while the physical assessment was based on the loose bulk density, and percentage finest of the product. The biological assessment of the product was based on phytotoxicity evaluation using the Germination index (Tibu et al., 2019). Compost extracts were prepared from the final compost by mixing 20 g of air-dried compost in 10 cm3 of distilled water. The mixture was then shaken for 30 min after which was filtered using Whatman No 4 filter paper to produce compost extract which was then used in germination index tests. Tomatoes seed (Solanum lycopersicum) (Viability as tested = 90%) was used for the germination index test. Whatman No 4 filter paper already moistened with an extract from compost was laid in a Petri dish and Ten (10) viable seeds of tomatoes were placed on it. A control experiment was also set up using deionized water only. The experiments were replicated thrice and were set up in the laboratory where the temperature was maintained at room temperature. After 7 days of incubation, germinated seeds were counted (Tibu et al., 2019) and the germination index (GI) was evaluated according to Equation 4.

$$GI\,(\%) = \frac{\%\ seed\ Germination\ \times Root\ Lenght\ of\ Treatment}{\%\ seed\ Germination\ \times Root\ Lenght\ of\ Control} \times 100 \quad (4)$$

where:
GI is the germination index

## 3.4 Physical properties analysis of the final compost

### *3.4.1 Percentage finest determination*

The percentage finest determination of the matured compost was done using a mechanical shaking method. From each of the compost, a known representative sample of dried compost was placed on a stack of 5 standard test sieves arranged on the shaker and shaken for 10 minutes. The mass (g) of compost retained on each sieve was measured and recorded. The procedure was repeated three times for the products. The retained compost samples on each sieve were classified into four different fraction sizes: oversize, coarse, pin, and fine. Particle size < 24 mesh (>850 µm) were oversize, 24-60 mesh (500 - 850 µm) were coarse, 60-70 mesh (400-500 µm) were pin size and 70 - 80 mesh (177-400 µm) were classified as fine particle size.

### *3.4.2 Fibrosity content determination*

The fibrosity content of the matured compost was measured by the method described by Boylan et al. (2009). A compost sample with Known volume and water content was saturated overnight in a concentration of 40g/L solution of sodium hexametaphosphate to disperse the fibers. The sample was then washed in a 150µm sieve with distilled water. The retained material on the sieve was then gently rubbed by hand and the remaining fibers with a diameter greater than 0.5 mm were removed using tweezers. This was then oven-dried at 80°C to a constant mass. Percentage fiber was then calculated using Equation (5).

$$Fibrosity\ content\ (\%) = \frac{M_{fibre\text{-}dry}}{M_{original\text{-}dry}}\ x\ 100 \tag{5}$$

where:

$M_{fibre\text{-}dry}$ is a dry mass of fibers

$M_{original\text{-}dry}$ is the original dry specimen mass

### 3.5 Statistical analysis

All the experiments were repeated three times and for each sampling; the mean and standard deviation were reported in this study. All the calculations and graphical analysis were done using Microsoft excel 2010.

## 4. RESULTS AND DISCUSSION

### 4.1 Composting temperature evolution

Composting temperature is one of the key parameters of the stability index that indicate the stability and maturity of the compost (Mayur et al., 2018). The breaking down of complex organic compounds into simpler units is enhanced by temperature (Waqsa et al., 2018). Figure 2 shows the temperature profile of mixtures 1, 2, and mixture 3 for different bulk densities respectively. As shown in the figures, temperature ranges and duration at each stage differ in each of the experimental mixtures and this could attribute to the different experimental conditions of each of the composting mixtures. Microbes' activities are responsible for an increase in temperature during an active composting period (Prashant et al., 2019). For it to perform at optimum, it should be provided with adequate nutrients, moisture, and oxygen. In this study, it was observed that the temperature of the three composting mixtures increased rapidly from day 1 of the experiment, peaked on different days, and started cooling until they were stable and reached ambient temperature. This shows that biodegradation of organic materials through the activities of microbial has started. Jakubus (2020) recorded the same in the study of comparative compost prepared from various organic wastes based on biological and chemical parameters. In this study, each of the composting mixtures recorded the three temperature phases i.e mesophilic < 45 oC, (heating period), thermophilic > 45°C (high temperature period) and cooling phase < 45°C (Mayur et al., 2018). The optimum temperature range to kill pathogen is 40-65°C (Wang et al. 2021, Bao et al. 2016) and it must last for three to four days to sanitize the compost. In this study, the observed temperature ranged between 42.3°C and 65.3°C throughout the experimental period and this is enough to kill pathogens in the compost.

Mixture 3 recorded the highest temperature (65.3°C) and it lasted for more than three days. In mixture 3, the initial bulk density was 25 Kg/m$^3$; therefore, the physical structure of this mixture allows even distribution of oxygen and moisture for microorganisms. The increase in the activities of microorganisms in this mixture leads to a rapid increase in temperature. Compost mixture 1 recorded the lowest temperature during the high-temperature period (49.3°C). The lowest temperatures recorded in this mixture indicate lower activities of microorganisms. In mixture 1, the compost materials are closely packed together as a result of compaction, a high rate of biodegradation occurred and this led to high-temperature evolution within the compost. This shows that a bulk density of 25 K/gm$^3$ is favourable for temperature rise as free air space is reduced therefore more oxygen and moisture is available for microorganisms. Therefore, it may be assumed that the high bulk density corresponds to less free air space. Microorganism decomposes organic matter and heat is released; the temperature of the composting mixture increases at the beginning of the process. With the decrease in organic matter content of the materials and through heat loss by ventilation and evaporation (Arias et al., 2021), the temperatures of the mixture gradually decreased and reach ambient temperature. However, the best bulk density from the perspective of temperature in this study was that of experiment mixture 3.

### 4.2 Moisture content

Estimation of moisture is important for optimum productivity of composting process and one of the major fac-

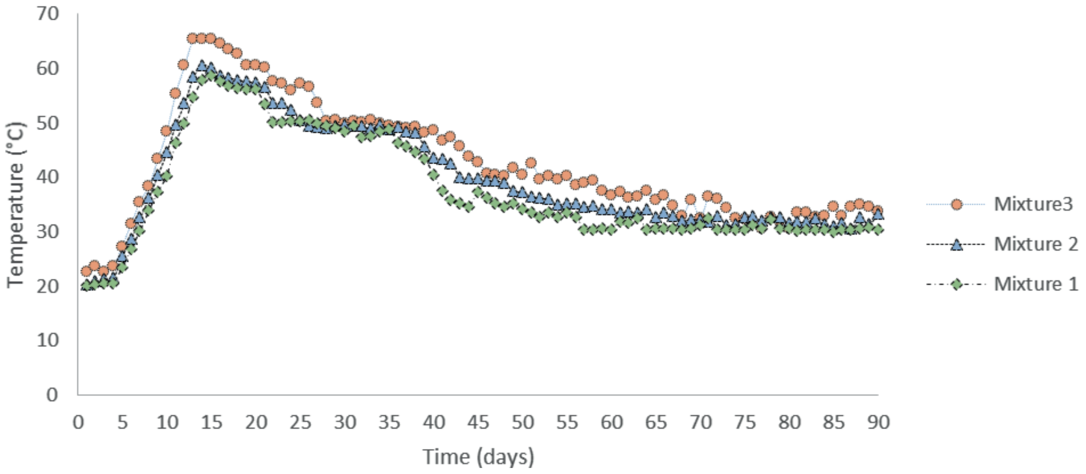


**FIGURE 2:** Variation of the temperature of the three mixtures during composting.

tors that need to be considered in the composting system design (Hemidat et al., 2018). Moisture must flow for an adequate supply of oxygen for proper microbial activities. Literature reported different moisture content, for example, Liang et al. (2003) recommended moisture within the range of 70% while Wang et al. (2021) recommended moisture within the range of 50-60%. Moisture content greater than 60% is not recommended as it prevents oxygen from the tiny pore of the compost pile and lowers its aerobic activities (Nahm, 2005). This is supported by Looper (2002) who found that moisture content above 60% produces odour and stops temperature to rise to thermophilic during composting. Bulk density directly influenced the moisture needed for effective composting. The theoretical volume of water needed is related to the initial bulk density and air-filled porosity of the compost (Equations 1 and 2). Concerning mixture 1, 2, and 3, the initial moisture contents were adjusted to the required range of 60% (Wang et al., 2021). Consistent with other reported data, this moisture range was the best to support microbial activities. Generally, the moisture content of the three compost mixtures decreased gradually during the composting process in the first four weeks of the composting period. This can be attributed to active microbial activities and the turning frequency of the compost (Cao et al., 2020). The change in moisture content was more pronounced in mixture 3, followed by mixture 2, and least in mixture 1. This result is in line with the findings of Pezzola et al. (2021) in the study of the use of new parameters to optimize the composting process of different organic waste.

The moisture content of mixtures 2 and 3 was significantly decreased and the highest temperatures were also recorded in these mixtures. The decrease in moisture content in mixtures 2 and 3 may be due to the activities of microorganisms that consume moisture (Jae-Han et al., 2020). In mixture 3 which had the highest bulk density, a maximum moisture content reduction of more than 50% was observed. This could be a result of the highest bulk density of the compost (Mayur et al., 2018). During active composting, bulk density increases and eliminates or reduces the air-filled-porosity which is believed to be inaccessible to microorganisms (Mayur et al., 2018) and this leads to a decrease in porosity; as a result, less moisture would be available for microorganisms and this would reduce their activities (Makan et al., 2013). As the compost approached stability, the moisture content in all the mixtures gradually decreased to around 23.33%, 21.54%, and 18.43% in mixtures 1, 2, and 3 respectively. There was no significant relation between mixing ratio and moisture content, but mixture 3 with the highest bulk density recorded the lowest reduction of moisture content.

## 4.3 Evaluation of pH and electrical conductivity (EC)

### *4.3.1 pH*

Figure 4 shows the evaluation of the pH of the composts. The pH range during the composting period is used to assess the progress of composting as it influences the microorganism growth and gaseous loss of ammonia (Hemidat et al., 2018). Changes in pH were observed in the three mixtures throughout the composting period in this study. Many researchers have reported that the initial pH value of the mixture should range from about 6.0 to 7.0 (Chang et al., 2019; Varelas, 2019). However, the initial pH value of the mixtures after mixing in this study was slightly alkaline (6.3-6.9) which was optimum for microbial activities. This was in agreement with the finding of Abdul-Halim et al. (2019) who recorded the same range of initial pH values. As the composting was progressing, the pH value varies across the experiment. However, after the third week, i.e during the thermophilic stage, pH values in the mixtures significantly increased. The significant increase in pH value was more pronounced in mixture 3. The pH increase in all the mixtures may be a result of volatilization of organic acid under high temperatures (Manu et al., 2019), consumption of organic acids by microorganisms, the production and accumulation of NH4+ and humic substances (Elkinci et al., 2019, Manu et al., 2019), and mineralization of acidic compounds such as carboxylic and phenolic group (Madejon et al., 2021) and due to the breaks down of complex amino acids and peptides with the release of $NH^{+4}$ (Sundberg et al., 2013). The pH changes in mixtures 1 and 2 followed a similar pattern. After then, pH values in all the mixtures were then decreased. According to Wang et al. (2021), the production of NH3 gas from the decomposition of nitrogen tends to increase the pH value in the early weeks of composting but decreases later due to the decomposition of organic acid to organic matter. At the end of the experiment, the pH of the three mixtures of this study decreased and was observed to be lower than the initial values and almost alkaline. Mixture 1 with an initial bulk density of 15 Kg/m$^3$ showed the lowest final pH range (7.4), while mixture 2 and 3 was a little bit higher than mixture 1 (Figure 3).

### *4.3.2 Electrical conductivity of the composting mixtures*

Electrical conductivity (EC) determination is crucial during composting as it indicates the salinity and the usability of final compost products. An increase in EC would lead to phytoinhibitory effects (Zhou et al., 2019). The electrical conductivity greater than 4 mS/cm is considered injurious to plants (Manu et al., 2018) because the soluble salts can negatively affect seed germination. The electrical conductivity of each of the mixtures displays an irregular pattern throughout the composting period. It first increased then decreased and later increased at the end of the experiment. When the experiment was started, compost mixture 3 recorded the highest EC (Figure 5). This observation might be a result of the highest proportion of food waste in mixture 3 which leads to the buildup of soluble salt which is assumed to be a result of food salinity or the presence of mineral salts like phosphates and $NH_4^+$ through the breakdown of compost materials (He et al., 2020). The initial EC of mixture 3 is significantly higher than compost mixtures 1 (1.8 mS/cm) and 2 (1.8 mS/cm). This may be due to the highest proportion of food waste in compost mixture 1. Mixture 1 had the smallest proportion of food waste so it had the smallest initial EC value. Early in the third week, compost mixtures 1 and 2 showed a similar value of EC, except for mixture 3 which still maintained a higher EC value

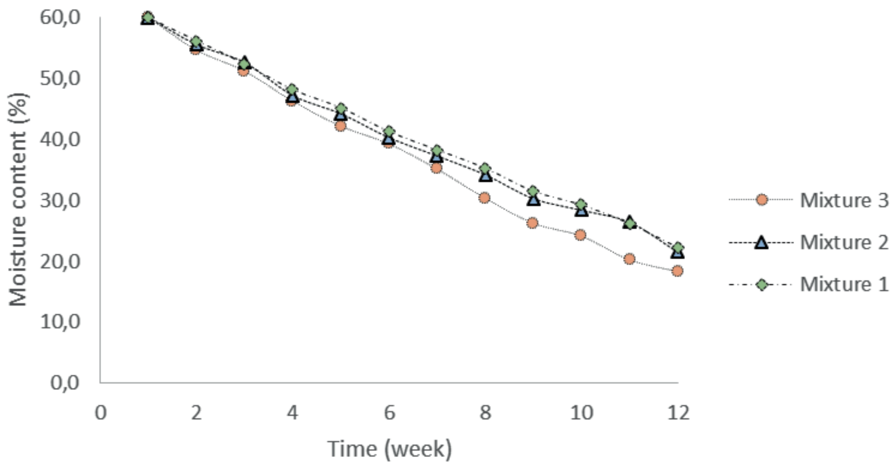


**FIGURE 3:** Variation of moisture content of composting mixture.

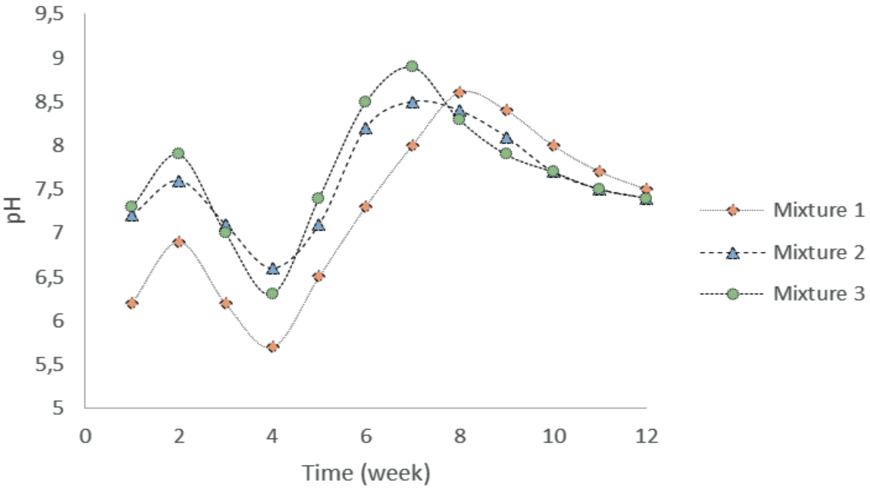


**FIGURE 4:** pH of the composting mixtures.

than mixtures 1 and 2. After the 8th week of experiment, no significance reduction was observed in EC of compost mixture 3. It still maintained higher EC than mixtures 1 and 2. However, in the early 10th week, all the compost mixtures had similar EC values. The final EC was below 4 mS/cm which is good for plant production (He et al., 2020).

## 4.4 Total Organic Carbon, Total Nitrogen, and C/N ratio

### *4.4.1 Total organic carbon*

Organic carbon from the bulking agent is consumed by microorganisms as food for metabolic activities, this leads to the degradation of organic matter by microbial activities in the presence of oxygen with the release of $CO_2$ gas leading to the production of organic matter, therefore, total organic carbon decreases generally during the composting process (Bello et al., 2021). As shown in Figure 6, TOC showed a downward trend in all the mixtures with mixture 3 exhibiting the greatest decrease in TOC, while mixture 1 recorded the lowest decrease in TOC. The greatest reduction in total organic carbon was observed at the thermophilic stage, and the main reason for the decrease in TOC content in mixture 3 was because high temperature and vigorous microbial activities recorded in this mixture, so at the end of the process, TOC was lowest in this mixture. In mixture 2, organic carbon degradation was gradual while it was slowest in mixture 1. The reason for the slowest organic matter degradation in compost mixture 1 was because of the lowest temperature recorded in the mixture. In mixture 1, the composting materials are less compacted leaving more space for air and moisture to penetrate, therefore microbial activities were slowest in this mixture and as a result, the temperature was lowest in this mixture. At the end of the composting process, total organic carbon reduction in compost mixture 1 was the least.

### *4.4.2 Total Nitrogen*

Figure 7 shows the nitrogen variation of each of the mixtures. Total nitrogen was first decreased in all the mixtures during the earlier stage and later increased continuously till the end of the composting period. Several scholars reported similar observations in their study. For example, Yu et al. (2019) observed that nitrogen content first decreased and then increased during the study of changes in carbon,

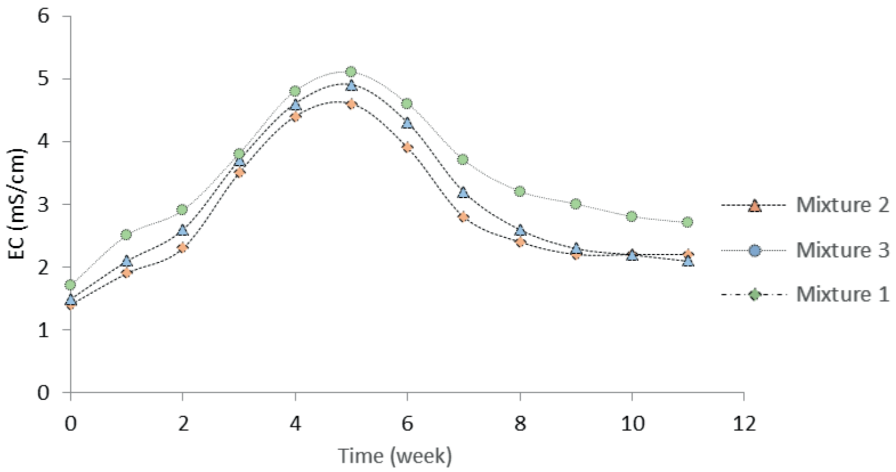


**FIGURE 5:** Electrical conductivity of the composting mixtures.

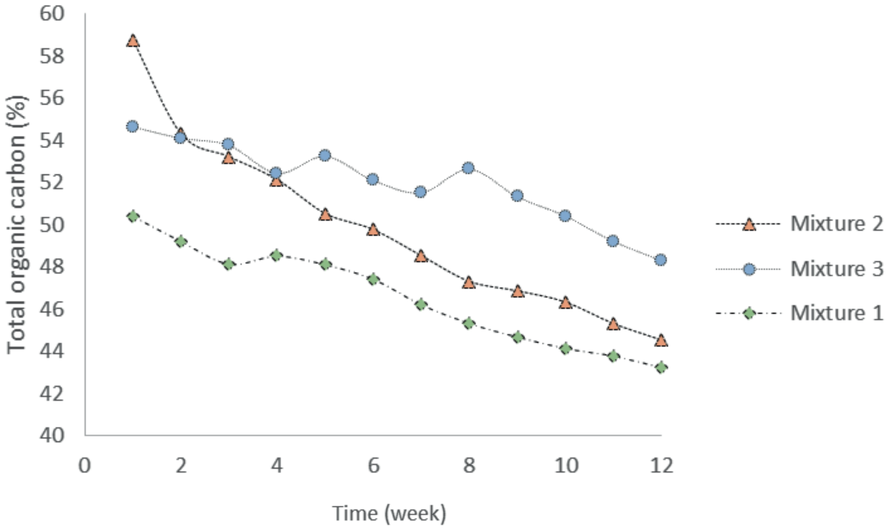


**FIGURE 6:** Total organic carbon of the composting mixtures.

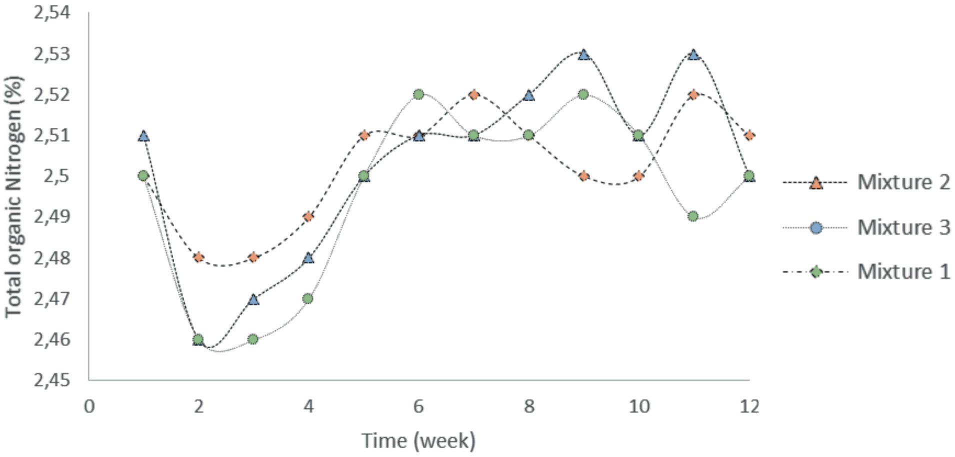


**FIGURE 7:** Total organic nitrogen of the composting mixtures.

nitrogen components, and humic substances in organic-inorganic aerobic co-composting. The authors claimed that the loss in nitrogen at the early stage of composting could be attributed to volatilization (Yu et al., 2019) and due to some loss in the form of $NH_3$-N (Cao et al., 2020; Sun et al., 2017; Lu et al., 2016). The loss in nitrogen content at the

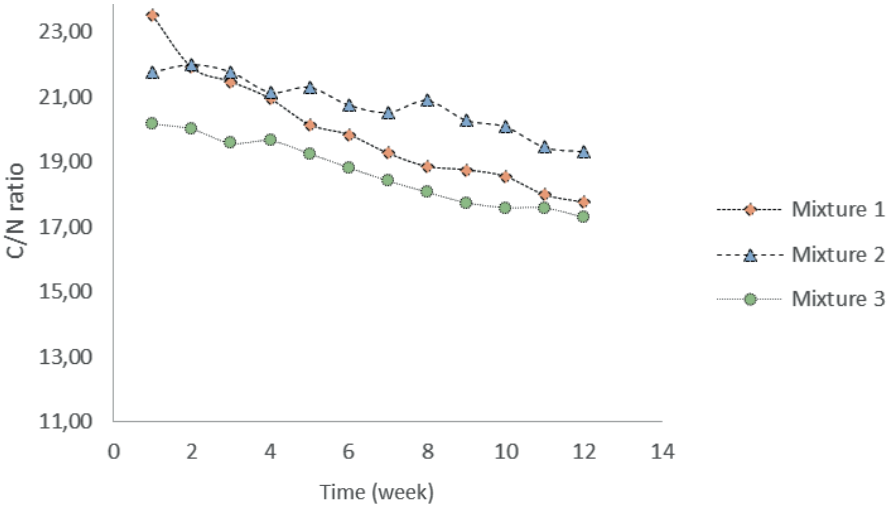


**FIGURE 8:** N ratio of the composting mixtures.

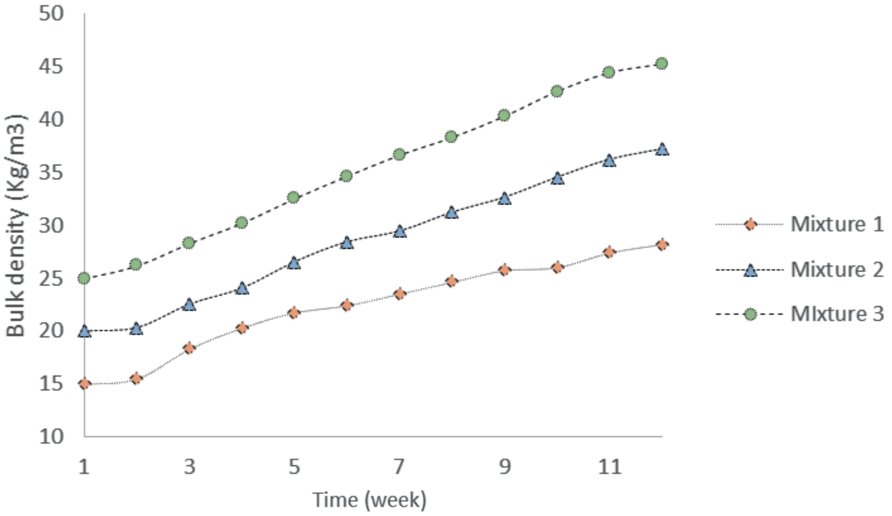


**FIGURE 9:** Variations of bulk densities during the composting period.

early stage of composting could be due to the consumption of nitrogen content by microorganisms for growth and reproduction (Ren et al., 2016) and due to leaching from the compost. However, as the composting is progressing, a significant increase in nitrogen content was recorded in all the mixtures during the subsequent sampling. An increase in nitrogen may be because of the degradation of organic carbon compounds (He et al., 2017).

### *4.4.3 C/N Ratio*

Another important parameter for the determination of composting time, quality of the final compost, and evaluation of compost maturity is the C: N ratio. It also plays an important role in formulating the nutritional balance of a composting mixture. Carbon and nitrogen are needed by microorganisms as a source of energy for metabolic activities. A proper C: N ratio is favourable to microbial growth and production and also good for soil and plant growth (Kebibeche et al., 2019). As the composting progressed, the C/N ratio decreased throughout the period in all the mixtures as shown in Figure 8. A decrease in C/N ratio was highest in mixture 3, moderate in mixture 2, and lowest in mixture 1. This may be due to variation in temperature as a result of compaction.  Mixture 1 had the lowest bulk density (15 Kg/m$^3$) so, the temperature was lowest in this mixture and it recorded the lowest reduction in C/N ratio value. In compost mixture 3, the changes in total nitrogen and organic carbon were high; therefore, the reduction of the C/N ratio was highest in this mixture. At the end of the process, the highest reduction in C/N ratio was in mixture 3 and the lowest reduction was observed in mixture 1. This result was in line with the result of Getahum et al. (2012) in the composting of municipal solid waste.

## 4.5 Physical properties variation during composting

### *4.5.1 Bulk density, particle density, and porosity*

Figure 9 shows the variation of bulk density of the mixtures throughout the composting period. The initial bulk density of the compost mixtures was 15 kg/m$^3$, 20 kg/m$^3$, and 25 kg/m$^3$ for mixtures 1, 2, and 3 respectively. During the process, an increase in bulk density was observed in all the mixtures. This may be a result of the settlement of the compost as soon as it was compacted. A similar result was reported by Zhao et al. (2011) during composting of municipal solid waste of different particle sizes. Zhao et al. (2011) reported that an increase in bulk density was a result of the decrease in particle size of the waste. During the compaction of the compost, the volume of the compost is reduced while its mass remains unchanged. The highest bulk density in this study was observed in compost mixture 3, while the lowest bulk density was observed in compost mixture 1. Therefore, an increase in bulk density in mixture 3 may be a result of the compaction effect.

For an effective composting process, air (oxygen) must flow to achieve maximum performance. Bulk density, porosity, and free air space are interconnected and play a critical role in air movement in the compost mix (Iqbal et al., 2010). Free-air-space or air- filled-porosity as cited in literature with a minimum value of 30% and 60% maximum is required to ensure aerobic condition (Mayur et al., 2018). Different studies reported different free air space values. For example, Ahn et al. (2008) and Ruggieri et al. (2009) reported a maximum value of 85-90% without any negative effects on compost. But in this study, a decreasing trend in porosity value was observed with maximum and minimum porosity values of 46.34% and 37.41% were observed. The minimum and maximum porosity value of 37.41% and 39.23% was recorded in compost mixture 1. For mixture 2, 33.64% minimum and 40.36% maximum were recorded. While 46.34% maximum and 30.67% minimum were recorded in mixture 3.

## 4.6 Evaluation of Final Compost

### *4.6.1 Maturity and stability analysis of the matured compost*

Compost maturity simply refers to the level of decomposition of the poisonous substances formed during the composting phase (Wu et al., 2000). The maturity and stability of the final compost are important for its use in agriculture. If compost is not stable, microbial activities in it can cause adverse effects and can affect plant growth since the final product will be of agricultural use. The properties of the final products are shown in Table 3. The final pH of all the mixtures was within the recommended value (6-8), with the value being close to alkaline (7.4-8.3). Total nitrogen in mixture 3 (1.78%) was slightly higher than in mixtures 1 (1.14%) and 2 (1.45%). The electrical conductivity ranged between 3.15 and 2.13 dS/m, the C/N ratio ranged between 10.56 and 19.59, the CEC value between 13.66 and 26.27, a phosphorous value between 8.29 and 11.08 Cmol/Kg, and final moisture content values between 20.20 and 23.20%.

### *4.6.2 Phytotoxicity of the matured compost*

The matured compost must be free of any poisonous substance before it can be used as soil organic fertilizer. Determination of the level of toxicity is important as it gives insight into the agricultural value of the final product. The most common and economical method used to evaluate the agricultural value of the final compost is the germination index method (Tibu et al., 2019). A germination index test was carried out to evaluate the level of phytotoxicity of the final compost of this study using tomato seed recorded 100% in mixtures 1 and 2 while mixtures 3 recorded 90% germination. Similar observations were reported by Tibu et al. (2019). In his study of phytotoxicity, a germination index between 80 and 100% was reported. The high germination index recorded in this study might be due to the presence of nutrients in adequate proportion in the final compost of each of the mixtures. Tibu et al. (2019) reported that if the germination index values are greater than 80%, then the compost is phytotoxin-free and it is safe and good to use. So, in this study, all the compost mixtures showed germination index values greater than the limit value, and therefore considered phytotoxin-free and safe to use.

## 4.7 Physical properties of the final compost

### *4.7.1 Finest and fibrosity content of the matured compost*

The physical characteristics of the final compost were also evaluated based on the finest ratio and fibrosity content. As shown in Table 4, mixture 3 had the highest percentage of finest (97%), followed by mixture 2 with 94% finest, and experiment mixture 1 had the lowest percentage finest 89% finest. Fibrosity contents vary significantly among the experimental mixtures, experiment mixtures 3 and 2 recorded zero fibrosity while experiment mixtures 1 recorded 5% fibrosity content. The zero fibrosity recorded in mixtures 2 and 3 shows that all the sawdust in these mixtures were decompose totally while mixture 3 contains some fiber content as a result of incomplete decomposition of the sawdust by microorganisms and heat.

## 4.8 Regression analysis

The interactions between the physical parameters (bulk density, porosity, particle density, and moisture content) considered in this study were analyzed using correlation analysis (Wu et al., 2019). A strong positive correlation was found between porosity and moisture content and it exceed 0.9. The correlation between bulk density and porosity was negatively correlated (-0.942), and the highest negative correlation was between moisture content and bulk density (-0.978). The reason for the negative correlation between bulk density and porosity is that bulk density increases throughout the composting process the porosi-

**TABLE 3:** Chemical properties of the final compost.

| Composting Mixtures | C: N | pH | EC | CEC (Cmol/Kg) | Phosphorus (Cmol/Kg) | Nitrogen (%) | Moisture content (%) |
|---|---|---|---|---|---|---|---|
| Mixture 1 | 19.59 ± 0.25 | 7.8 ± 2.11 | 2.13 ± 0.24 | 13.66 ± 0.08 | 8.29 ± 1.52 | 1.14 ± 0.02 | 23.20 |
| Mixture 2 | 14.85 ± 0.10 | 8.3 ± 2.01 | 2.91 ± 0.22 | 22.20 ± 0.28 | 11.08 ± 0.81 | 1.45 ± 0.01 | 22.40 |
| Mixture 3 | 10.56 ± 0.20 | 7.4 ± 1.07 | 3.15 ± 0.16 | 26.27 ± 0.12 | 8.25 ± 1.44 | 1.78 ± 0.01 | 20.50 |

**TABLE 4:** Physical properties of the final compost.

| Mixtures | Finest (%) | Fibrosity (%) | Bulk density (K/gm$^3$) | |
|---|---|---|---|---|
| | | | Initial | Final |
| Mixture 1 | 89.0 ±0.01 | 0.5 ± 0.012 | 15.0 | 28.21 ± 0.2 |
| Mixture 2 | 95.0 ± 0.12 | 0.0 ±0.00 | 20.0 | 37.21 ± 0.5 |
| Mixture 3 | 97.0 ± 0.11 | 0.0 ± 0.00 | 25.0 | 45.23 ± 0.3 |

**TABLE 5:** Effects of treatment on the physicochemical parameters measured during the experiment.

| Parameters | Mixture group | | |
|---|---|---|---|
| | 1 | 2 | 3 |
| pH | 7.25a | 7.6b | 7.6b |
| MC (%) | 37.38a | 39.85b | 40.53b |
| TOC (%) | 49.80a | 51.99a | 46.59b |
| TON (%) | 2.50a | 2.5a | 2.19b |
| C/N ratio | 19.91a | 20.78a | 18.68b |

*Note: Values are the averages (n = 3). Different letters are related to significant differences tested by Duncan's multiple ranges (p < 0.05)*

ty shows decreasing trend. The same thing happened between moisture contents and bulk density, as composting progressed moisture was lost and bulk density increases.

## 5. CONCLUSIONS

The study was carried out to investigate the influence of the physical properties of composting materials on composting of sawdust with food waste. For this reason, sawdust and food waste was composted in a bin at the initial bulk densities of 15, 20, and 25 kg/m$^3$. According to the results obtained and presented above, the three composting mixtures at different initial bulk densities had reached an acceptable degree of maturation and stability at the end of the composting process. The highest temperature of 65.3 o C was recorded in mixture 3 with the highest bulk density and lasted for more than four days. Loss in moisture content was more pronounced in mixtures 3 than in mixtures 1 and 2. The lowest EC was observed in mixture 1 and the highest in mixture 3. The bulk density increases throughout the process, porosity, and moisture content decrease during the composting process. Mixtures 3 and 2 had the highest finest and lowest fibrosity content and mixture 1 had the lowest finest and highest fibrosity content. Based on the result presented above, the best compost was produced at 25 kg/m3 bulk density and 46.34% porosity.

## ACKNOWLEDGMENT

The authors are grateful to the Department of Agricultural and Biosystems Engineering, University of Ilorin, Nigeria, without which this study would not have been undertaken.

## REFERENCES

Abdul-Halim, A., Ben, K. B., Banful, L. A., 2019. Standardizing the Quality of Composts Using Stability and Maturity Indices: The Use of Sawdust and Rice Husks as Compost Feed Stocks. American Journal of Plant Sciences, 10, 2134 -2150

Adedayo, V., Adeaga, O., Akomolafe, D., 2002. Volume and Content Estimate of Household Food Waste: A Strategy for Reducing Food Insecurity in Lagos State, Nigeria. Direct Reserc jor of Agric and food Sci. 8 (6) 208 - 216

Adegoke, O.A., Fuwape, J. A., Fabiyi, J. S., 2014. "Combustion properties of some tropical wood species and their pyrolytic products characterization".Energy and Power. 4(3):54-57

Agnew, J.M., Leonard, J.J., Feddes, J., Feng, Y. A., 2003 Modifed air pycnometer for compost air volume and density determination. Can. Biosyst. Eng. 45, 6–27

Ahn, K.K., Richard, T.L., Glanville, T.D., 2008. Laboratory determination of compost physical parameters for modeling of airflow characteristics. Waste Manage. 28, 660–670

Al-Rumaihi, A.; McKay, G.; Mackey, H.R.; Al-Ansari, T., 2020. Environmental impact assessment of food waste management using two composting techniques. Sustainability, 12, 1595

Arias, O., Vina, S., Soto, M., 2021. Co-composting of forest and indusmixture wastes watered with pig manure. Environ. Technol. 42 (5), 705–716. https://doi.org/10.1080/ 09593330.2019.1643411.

Assandri, D., Pampuro, N., Zara, G., Bianco, A., Cavallo, E., Budroni, M., 2021. Co-Composting of Brewers' Spent Grain with Animal Manures and Wheat Straw: Influence of Two Composting Strategies on Compost Quality. Agronomy, 11, 1349. https://doi.org/10.3390/agronomy1107134

Awasthi, M.K., Pandey, A.K., Khan, J., Bundela, P.S., Wong, J.W.C., Selvam, A., 2014. Evaluation of thermophilic fungal consortium for organic municipal solid waste composting. Bioresour. Technol. 168, 214-221.

Bao, Y., Chen, J., Yan, L., Guan, L., 2016. Fundamental conditions of composting. Chin J. Soil Sci 37(1):164–169. https://doi.org/10.19336/j.cnki.trtb.2006.01.037

Barrena, R., Turet, J., Busquets, A., Farrés, M., Font, X., Sánchez, A., 2011. Respirometric screening of several types of manure and mixtures intended for composting. Bioresour. Technol. 102, 1367–77.

Bello, H., Akeem A. J., 2021. Valorization of sawdust by co-composting with food waste using a small-scale composter. Int J Recycl Org Waste Agricult. 2103 – 1213.10.30486/IJROWA.2021.1924988.1213

Boylan, N., Long, M., 2009. An investigation of two peat failures in the wicklow mountains. Biology and environment: proceedings of royal irish academy, 110B (3) : 173 - 184. https://doi.org/10.3318/BIOE.2010.110.3.173

Brinton, W.F., 2000. Compost Quality Standards and Guidelines Final Report by Woods End Research Laboratories for the New York State Association of Recyclers

Calabi-Floody, M., Medina, J., Suazo, J., Ordiqueo, M., Aponte, H., de La Luz, M.M., Rumpel, C., 2019. Optimization of wheat straw co-composting for carrier material development. Waste Manag 98:37–49. https:// doi.org/10.1016/j.wasman.2019.07.041

Cao, Y., Bai, M., Han, B., Impraim, R., Butterly, C., Hu, H., He, J., Chen, D., 2020 Enhanced nitrogen retention by lignite during poultry litter composting. J Clean Prod 277:122422. https://doi.org/10.1016/j.jclepro.2020.122422

Cao, Y., Huang, H., Sun, J., Wu, H., Duan, H., Xu, Y., Jin, H., Chang, Z., 2018. Effect of hyperthermerphilic pretreatment on transformation and losses of C and N during pig manure composting. China. Environ Sci 38(5):1792–1800. https://doi.org/10.19674/j.cnki.issn1000- 6923.20180223.003

Cao, Y., Bai, M., Han, B., Impraim, R., Butterly, C., Hu, H., He, J., Chen, D., 2020. Enhanced nitrogen retention by lignite during poultry litter composting. J Clean Prod 277:122422. https://doi.org/10.1016/j.jclepro.2020.122422

Chang, H.C., Li, R.D., 2019. Agricultural waste. Water Environ. Res. 91, 1150–1167.

Chang, R., Li, Y., Li, J., Chen, Q., Zhao, H., Qi, Z., 2019. Influences of the thermophilic period on biodegradation and nitrogen loss in stimulated vegetable waste composting. Glob. Ecol. Conserv. 18, e00623

Chen, H., Awasthi, M.K., Liu, T., Zhao, J., Xiuna, R., Wang, M., Duan, Y., Awasthi, S.K, Zhang, Z., 2018. Infuence of clay as additive on greenhouse gases emission and maturity evaluation during chicken manure composting. Bioresour Res 266:82–88

Chen, M., Xu, P., Zeng, G., 2015. Bioremediation of soils contaminated with polycyclic aromatic hydrocarbons, petroleum, pesticides, chlorophenols and heavy metals by composting: applications, microbes and future research needs. Biotechnol Adv. 33 (6):745–755

Elcik, H., Zoungrana, A., Bekaraki, N., 2016. Laboratory-scale investigation of aerobic compost stability of municipal solid waste in Istanbul. Sigma J Eng Nat Sci 34(2): 211–220

Ekinci, K., Tosun, İ., Bıtrak, B., Kumbul, B.S., Şevik, F., Sülük, K., 2019. Effects of initial C/N ratio on organic matter degradation of composting of rose oil processing solid wastes. Int J Environ Sci Technol 16: 5131–5140. https://doi.org/10.1007/s13762-018-1947-9

FAO, 2020. The State of Food Security and Nutrition in the World.: http://www.fao.org/3/ca9692en/online/ca9692en.html#. Assessed on 10 oct., 2021

Fernández, J.M., López-de-Sá, E.G., Polo, A., Plaza, C., 2014. Short term stabilization of organic matter in physically, chemically, and biochemically protected pools in soils amended with municipal wastes, Clean-Soil Air Water, 42, 487–493. DOI: 10.1002/ clen.201300127

Getahun, T., Nigusie, A., Entele, T., Van Gerven, T., Van Der Bruggen, B., 2012. Effect of turning frequencies on composting biodegradable municipal solid waste quality. Resour. Conserv. Recycl., 65, 79–84

Ghosh, Ray., S., Ghangrekar, M.M., 2019. Comprehensive review on treatment of high-strength distillery wastewater in advanced physicochemical and biological degradation pathways. Int. J. Environ. Sci. Technol., 16, 527–546.

GómezBrandón, M., Lazcano, C., Domínguez, J., 2008 .The evaluation of stability and maturity during the composting of cattle manure. Chemosphere 70(3):436–444. https://doi.org/10.1016/j.chemosphere.2007.06.065

He, Z., Qiang, L, Xiaoyi, Z., Kai, T., Xiangshi, K., Xingjun, T., 2020. Impacts of peat on nitrogen conservation and fungal community composition dynamics during food waste composting. Appl Biol Chem 63:72. https://doi.org/10.1186/s13765-020-00552-8

Hemidat, S., Jaar, M., Nassour, A., Nelles, M., 2018. Monitoring of Composting Process Parameters: A Case Study in Jordan. Waste and BiomassValorization. https://doi.org/10.1007/s12649018-0197-x

Hwang, H.Y., Seong, H. K., Myung, S. K., Seong, J. P., Chang, H. L., 2020 Co-composting of chicken manure with oranic waste: characterization of gases emissions and compost quality. Appl Biol Chem (2020) 63:3. https://doi.org/10.1186/s13765-019-0483-8

Iqbal, M.K., Shafiq, T., Ahmed, K., 2010. Characterization of bulking agents and its effects on physical properties of compost. Bioresour. Technol. 101 (6), 1913–1919.

JaeHan L, Deogratius L., ChangHoon L., SeongJin P., TaekKeun, O., 2020. Efficiencies of unconventional bulking agents in composting food waste in Korea. Appl Biol Chem (2020) 63:68. https://doi.org/10.1186/s13765-020-00554-6

Jakubus, M., 2020. A Comparative Study of Composts Prepared from Various Organic Wastes Based on Biological and Chemical Parameters. Agronomy , 10, 869. https://doi.org/10.3390/agronomy10060869

Kebibeche, H., Khelil, O., Kacem, M., Harche, M.K., 2019. Addition of wood sawdust during the co-composting of sewage sludge and wheat straw influences seeds germination. Ecotox Environ Safe 168: 423–430. https://doi.org/10.1016/j.ecoenv.2018.10.075

Kelleher, M., 2007. Anaerobic digestion outlook for MSW streams. BioCycle 48 (8), 51e55

Kumar, M., Ou Y. L., Lin J.G., 2010. Co-composting of green waste and food waste at low C/N ratio. Waste Manag. 30:602–609

Liang, C., Das, K.C., McClendon, R.W., 2003. The influence of temperature and moisture contents regimes on the
aerobic microbial activity of a biosolids composting blend. Bioresour. Technol., 86, 131–137

Lim, S.L., Lee, L.H., Wu, T.Y., 2016. Sustainability of using composting and vermicomposting technologies for organic solid waste biotransformation: Recent overview, greenhouse gases emissions and economic analysis. J. Clean. Prod. , 111, 262–278.

Looper, M., 2002. Whole animal composting of dairy cattle. Dairy Business Communications;108:1-4

Lukashe, N.S., Mupambwa, H.A., Mnkeni, P.N.S., 2019 Changes in nutrients and bioavailability of potentially toxic metals in mine waste contaminated soils amended with fly ash enriched vermicompost. Water Air Soil Pollution. 230:306. https://doi.org/10. 1007/s11270-019-4343-2

Lu, Y., Cui, X., Long, S., Fan, H., Peng, F., 2016. Effect of organic materials with different proportions on composting process. Res Agr Modernization 37(3):587–593. https://doi.org/10.13872/j.1000-0275.2016.0002

Madejón, E., Marco P., Paula, M., Alfredo, P.d.M., 2021. Composting as Sustainable Managing Option for Seaweed Blooms on Recreational Beaches. Waste and Biomass Valorization https://doi.org/10.1007/s12649-021-01548-1

Makan, A., Assobhei, O., Mountadar, M., 2014. Initial air pressure influence on invessel composting for the biodegradable fraction of municipal solid waste in Morocco. Int. J. Environ. Sci. Technol. 11, 53e58.

Mashat, B.H., 2014. Effective microorganisms (EM) technology as a pathway to improve municipal solid waste of Makkah city (Saudi Arabia) and as foul odor eliminator. In: The Clute Institute International Academic Conference Munich (Germany)

Manu, M.K., Rakesh, K., Anurag, G., 2019. Decentralized composting of household wet biodegradable waste in plastic drums: Effect of waste turning, microbial inoculum and bulking agent on product quality. Journal of Cleaner Production 226:233e241

Manu, M.K., Kumar, R., Garg, A., 2018. Performance assessment of improved composting system for food waste with varying aeration and use of microbial inoculum. Bioresour. Technol., 234, 167–177.

Mayur, S. J., Mohit, D., Ajay S. K., 2018. Composting physics: A degradation process-determining tool for indusmixture sludge. Ecological Engineering 116 : 14–20

Mayur, S. J., Mohit D., Ajay, S. K., 2018. Physical parameters evaluation during production of soil conditioner from aquatic waste: Hydrilla verticillata (L.f.) Royle. / Environmental Technology & Innovation 11: 64–73

Moh, Y.C., Manaf, L.A., 2017. Solid waste management transformation and future challenges of source separation and recycling practice in Malaysia. Resour. Conserv. Recycl.. 116, 1-14

Nahm, K.H., 2005. Factors Influencing nitrogen mineralization during poultry litter composting and calculations for available nitrogen. Journal of Poultry Science; 61(2):238-255.

Nayak, A.K., Kalamdhad, A.S., 2014. Feasibility of composting combinations of sewage sludge, cattle manure and sawdust in a rotary drum reactor. Environ. Eng. Res. 19 (1), 1–11

Nizami, A.S., Rehan, M., Waqas, M., Naqvi, M., Ouda, O.K.M., Shahzad, K., Miandad, R., Khan, M.Z., Syamsiro, M., Ismail, I.M.I., Pant, D., 2017 Waste biorefiner: Enabling circular economies in developing countries. Bioresour. Technol. 241: 1101-1117. https://doi.org/10.1016/j.biortech.2017.05.097

Orthodoxou, D., Pettitt, T.R., Fuller, M., Newton, M., Knight, N., Smith, S.R., 2015. An investigation of some critical physico-chemical parameters influencing the operational rotary in-vessel composting of food waste by a small-to-medium sized enterprise. Waste Biomass Valoriz. 6(3), 293–302

Ouda, O.K.M., Raza, S.A., Nizami, A.S., Rehan, M., Al-Waked, R., Korres, N.E., 2016. Waste to energy potential: a case study of Saudi Arabia. Renew. Sustain. Energy Rev. 61, 328e340

Pezzolla, D., Mirko, C., Primo, P., Roberto, C., Luca, R., Giovanni G., 2021. The Use of New Parameters to Optimize the Composting Process of Different Organic Wastes. Agronomy , 11, 2090. https://doi.org/10.3390/agronomy11102090

Prashant, P. B., Bhargavi, N. K., 2019. Effect of active and passive aeration on composting of household biodegradable wastes: a decentralized approach. International Journal of Recycling of Organic Waste in Agriculture. https://doi.org/10.1007/s40093-019-00306-7

Purnomo, A.S., Mori, T., Kamei, I., 2011. Basic studies and applications on bioremediation of DDT: a review. Int Biodeterior Biodegradation. 65 (7):921–930.

Ren, G., Xu, X., Qu, J., Zhu, L., Wang, T., 2016. Evaluation of microbial population dynamics in the co-composting of cow manure and rice straw using high throughput sequencing analysis. World J Microbiol Biotechnol 32(6): 1–11. https://doi.org/10.1007/s11274-016-2059-7

Ruggieri, L., Gea, T., Artola, A., Sánchez, A., 2009. Air filled porosity measurements by air pycnometry in the composting process: a review and a correlation analysis. Bioresour. Technol. 100, 2655–66

Singh, J., Kalamdhad, A.S., 2013. Assessment of bioavailability and leachability of heavy metals during rotary drum composting of green waste (water hyacinth). Ecol. Eng. 52, 59–69.

Sukholthaman, P., Sharp, A., 2016. A system dynamics model to evaluate effects of source separation of municipal solid waste management: a case of Bangkok, Thailand. Waste Manage 52, 50-61

Sundberg, C., Jonsson, H., 2013. Higher pH and faster decomposition in biowaste composting by increased aeration. Waste Manag. 28, 518–526

Sun, X., Xu, X., Meng, Q., Cheng, L., Zhang, W., Men, M., Xu, B., Sun, Y., 2017. Research of ammonia oxidizing bacterial community structure and its correlation with environmental factors in cow manure composting. J Agro-Environ Sci 36(1): 189–197. https://doi.org/10. 11654/jaes.2016-0960

Tibu, C., Ted, Y. A., Negasi, S., Dzidzo, Y.T., 2019. Efect of the composting process on physicochemical properties and concentration of heavy metals in market waste with additive materials in the Ga West Municipality, Ghana. International Journal of Recycling of Organic Waste in Agriculture 8:393–403. https://doi.org/10.1007/s40093-019-0266-6

Varelas, V., 2019. Food wastes as a potential new source for edible insect mass production for food and feed: a review. Fermentation-Basel 5

Wang, H., Difu, Y., Jianling, X., Xuejun, L., Lianxi, S., 2021. Investigation of technology for composting mixed deer manure and straw. Enviro Sci and Pollu Resear. https://doi.org/10.1007/s11356-021-13886-7

Wang, L., Li, Y., Prasher, S.O., Yan, B., Ou, Y., Cui, H., Cui, Y., 2019. Organic matter, a critical factor to immobilize phosphorus, copper, and zinc during composting under various initial C/N ratios. Bioresour Technol 289:121745. https://doi.org/10.1016/j.biortech.2019.121745

Waqas, Q., Min, H.L., ·Byeong, E. M., Frank G. O., Fawad, K., Mohammad N., Hyeon T.K., 2018. Composting of chicken manure with a mixture of sawdust and wood shavings under forced aeration in a closed reactor system. Intern J Recycl Organ Waste Agricult https://doi.org/10.1007/s40093-018-0212-z

Wu, T., Wang, X.M., Li, D.J., Yi, Z.G., 2010. Emission of volatile organic sulfur compounds (VOSCs) during aerobic decomposition of food wastes. Atmos. Environ. 44 (39), 5065 - 5071.

Wu, Y., Chen, Y., Shaaban, M., Zhu, D., Hu, C., Chen, Z., Wang, Y., 2019. Evaluation of microbial inoculants pretreatment in straw and manure co-composting process enhancement. J Clean Prod 239:118078. https://doi.org/10.1016/j.jclepro.2019.118078

Wu, Y., Chen, Y., Shaaban, M., Zhu, D., Hu, C., Chen, Z., Wang, Y., 2019. Evaluation of microbial inoculants pretreatment in straw and manure co-composting process enhancement. J Clean Prod 239:118078. https://doi.org/10.1016/j.jclepro.2019.118078

Yu, K.M., Daniel, C.W.. T., Alex, C.K.Y., Season, S.C., Yong S.O.k , Chi S.P., 2019. Valorization of food waste into hydroxymethylfurfural: Dual role of metal ions in successive conversion steps. Bioresource Technology 219, 338–347

Zaihua, H., Qiang, L., Xiaoyi, Z., Kai, T., Xiangshi, K., Xingjun, T., 2020. Impacts of peat on nitrogen conservation and fungal community composition dynamics during food waste composting . Appl Biol Chem 63:72. https://doi.org/10.1186/s13765-020-00552-8

Zhao, S., Lian, F., Duo, L., 2011. EDTA-assisted phytoextraction of heavy metals by turfgrass from municipal solid waste compost using permeable barriers and associated potential leaching risk. Bioresour. Technol. 102, 621–626.

Zhou, G., Xu, X., Qiu, X., Zhang, J., 2019. Biochar influences the succession of microbial communities and the metabolic functions during rice straw composting with pig manure. Bioresour Technol 272:10–18. https://doi.org/10.1016/j.biortech.2018.09.135

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues
CISA

# ANAEROBIC DIGESTION OF BREWER'S SPENT GRAIN WITH BIOCHAR – BIOGAS PRODUCTION KINETICS AND PROCESS EFFICIENCY

Kacper Świechowski [1,]*, Ewa Syguła [1], Waheed Rasaq [1], Alan Gasiński [2] and Jacek Łyczko [3]

*[1] Department of Applied Bioeconomy, Wrocław University of Environmental and Life Sciences, Chełmońskiego St. 37a, Wroclaw, Poland*
*[2] Department of Fermentation and Cereals Technology, Wrocław University of Environmental and Life Sciences, Chełmońskiego St. 37, Wroclaw, Poland*
*[3] Department of Chemistry, Wrocław University of Environmental and Life Sciences, Chełmońskiego St. 37, Wroclaw, Poland*

**Article Info:**
Received:
21 January 2023
Revised:
3 April 2023
Accepted:
22 May 2023
Available online:
15 June 2023

**Keywords:**
Spent grain
Anaerobic digestion
Methane fermentation
Biogas kinetics
Pyrolysis, biochar

## ABSTRACT

Biochars made from brewer's spent grain were added to the anaerobic digestion of brewer's spent grain to enhance the methane fermentation process and improve biogas production. In research, the effect of biochars made at 300, 450, and 600°C and doses of 1-8% added to anaerobic digestion was tested. The biochemical biogas potential tests in mesophilic conditions were performed. The tests took 28 days, the biogas yield for each reactor varied from 500-650 $ml \times g_{VS}^{-1}$, and around 60% substrate degradation was obtained. For each test, the kinetics parameters using the first-order model were determined. The constant biogas production rate (k), and the biogas production rate (r) varied from 0.05-0.08 $d^{-1}$, and 42-60 $ml \times (g_{VS} \times d)^{-1}$ respectively. Though the differences in biogas production turned out to be statistically insignificant ($p<0.05$) due to the high disappearance in obtained data and conflicting effects, the response surface area analysis showed that biochar made at 450°C at the share of 1-4% could be used to maximize biogas production. Nevertheless, supplementation with biochar needs to be done carefully since in many cases, a reduction in biogas production was observed.

## 1. INTRODUCTION

Beer is one of the most globally consumed alcoholic beverages and it is one of the most popular drinks after water, tea, milk, and coffee. Over the two decades, beer production and consumption have steadily increased from 1.3 billion hL to almost 2 billion hL. In 2020, beer consumption was around 1.9 billion hL (Conway, 2022b), and the overall bear market was worth around 743.8 billion USD (Beer market report, 2022). Beer production is evenly spread between three regions of the world. Asia, Americas, and Europe are responsible for 0.55 billion hL, 0.61 billion hL, and 0.50 billion hL of beer production, respectively (Conway, 2021). Although the biggest beer manufacturers are China (0.34 billion hL), the United States (0.21 billion hL), Brazil (0.15 billion hL) (Conway, 2022c), the European Union (EU) countries produce 0.32 billion hL of beer. The biggest manufacturer in the EU is Germany and Poland with 0.087 and 0.039 billion hL produced in 2020, respectively (Conway, 2022a).

Though beer is a popular beverage, its production has a negative effect on the environment. The brewing industry is considered one of the largest industrial users of water. In the beer production process, water is used for technological processes like washing, cleaning sterilizing, and beer production itself. It is estimated that modern breweries consume from 4 to 7 L of water per 1 L of produced beer (Olajire, 2020). Besides water, the brewing process required a lot of energy. According to the Brewers Association, to produce 1 L of beer, electrical energy from 0.10 to 0.19 $kWh_{el}$ and thermal energy from 0.32 to 0.37 $kWh_{t}$ are needed (Cheri et al., 2014). The specific values of consumed water and energy depend on the used technology and the size of the brewery. The larger the size, the lower the specific resource consumption. Besides resource consumption, beer production leads to waste production and greenhouse gas emissions (Olajire, 2020). It is estimated that beer production has a global warming potential (GPW) of 0.40-1.47 $kg_{CO2eq.} \times L_{beer}^{-1}$ (Amienyo & Azapagic, 2016) and together with other alcoholic beverages accounts for 0.7% of global greenhouse gas (GHG) emissions when the complete product lifecycle is considered (R. Shin & Searcy, 2018). The beer production process consists of several steps during which various waste and by-products are

* Corresponding author:
Kacper Świechowski
email: kacper.swiechowski@upwr.edu.pl

Detritus / Volume 23 - 2023 / pages 16-27
https://doi.org/10.31025/2611-4135/2023.17277

generated. With each liter of beer, around 7 liters of wastewater is created. Also by-products like malt barley rootlets (MBR ~ 0.03-0.05 $kg\times kg_{malt}^{-1}$), brewer's spent grain (BSG ~ 0.14-0.19 $kg\times L_{beer}^{-1}$), spent hops/hot trub (HT ~ 0.002-0.004 $kg\times L_{beer}^{-1}$), and brewer's spent yeast (BSY ~ 0.02-0.04 $kg\times L_{beer}^{-1}$) are produced (Cimini & Moresi, 2021). Spent grains, hops, and yeast are high-energy materials that have the potential for vast applications in biotechnology for microalgae production, biofuel production, extraction of proteins, polyphenolic and antioxidative substances, and the food industry (Karlović et al., 2020). Nevertheless, most of the potential applications are at the beginning of the research and it is unknown if they are economically feasible for all brewery wastes. Mainly, the high moisture content and perishable nature of by-products prevent their safe usage in the human food chain and other applications (Cimini & Moresi, 2021). As a result, in most cases, brewing by-products are used as animal feedstock, are spread on the field, or are incinerated (Karlović et al., 2020).

Even though some applications are not economically feasible yet, they may turn out to be feasible in the future. Due to the huge amount of beer produced annually worldwide, by-products are available in large quantities throughout the year and their proper and smart utilization may reduce the negative effects of beer production. Taking into account that worldwide around 1.9 billion hL of beer is produced annually (Conway, 2022b), and with each litter of beer around 0.14-0.19 kg of wet spent grain is produced (Cimini & Moresi, 2021), the world spent grain potential is around 26.6-36.1 million Mg of which 7.0-9.5 million Mg in the EU.

One of the potential applications and economically feasible processes that can be applied to BSG is methane fermentation (a.k.a anaerobic digestion). Anaerobic digestion (AD) is a process that allows converting a huge quantity of wet and biodegradable biomass into biogas and digested in a relatively short time. Biogas is a flammable gas that can provide heat and electricity to the brewing process as a replacement for natural gas or coal while digestate can be used as a fertilizer. Using digestate reduces the need for fuel consumption related to synthetic fertilizers production. As a result of AD, beer production can become more environmentally friendly and provide additional income to the owner of the brewery plant (Li et al., 2011; Miller et al., 2021). The biochemical methane potential tests (BMP) show that spent grains are characterized by a methane yield (MY) of 305 $m^3_{CH4}\times Mg_{VS}^{-1}$. Assuming that BSG's total solids and volatile solids are 15% and 95% respectively, the MY of fresh BSG is 43.4 $m^3_{CH4}\times Mg_{wetBSG}^{-1}$ (Oliveira et al., 2018). The methane yield of BSG is comparable to other biomasses and wastes that are applied to AD worldwide. Most manures have an MY of 157-438 $m^3_{CH4}\times Mg_{VS}^{-1}$. The MY of lignocellulosic biomass varies from 160 to 212 $m^3_{CH4}\times Mg_{VS}^{-1}$, and the MY of organic municipal solid waste varies from 143 to 516 $m^3_{CH4}\times Mg_{VS}^{-1}$. The MY depends on biomass compositions (the content of carbohydrates, proteins, and lipids) and AD process conditions (process time, temperature, and used technology) (Nwokolo et al., 2020).

BSG is characterized by high protein and fiber content (hemicellulose, cellulose, and lignin). By dry weight, protein constitutes 15.3-24.7%, hemicellulose 19.2-29.6%, cellulose 16.8-25.3%, lignin 11.9-27.8%, and ashes 0.12-0.46% (Ikram et al., 2017). Though BSG is high-energy content material (high heating value of 21 $MJ\times kg^{-1}$) (Arranz et al., 2021), its lignocellulosic nature hinders the anaerobic digestion and does not allow for fully utilized energy potential. It is mainly due to the slow rate of lignocellulose degradation under AD conditions and the fact that biogas production can be inhibited by phenolic intermediates (such as p-cresol) produced during lignocellulose degradation (Bougrier et al., 2018). The AD of BSG as a mono-substrate is troublesome, even at a low organic loading rate (OLR ~1–2 $g_{VS}\times dm^{-3}\times d^{-1}$). Usually, mono-fermentation of BSG collapses after ~2-4 months (Bougrier et al., 2018; Sežun et al., 2011). Nevertheless, proper supplementation with trace elements solves this problem and the process can be performed efficiently and stably (Bougrier et al., 2018).

A lot of methods for pretreatment of lignocellulose biomass before AD was proposed and tested, i.e., mechanical, thermal-pressure, chemical, and biological treatments. These methods are used to change the physical properties and chemical composition of the biomass making it more available for the AD microorganism. For physical properties change counts decrease in particle size, increase in pore volume, and specific surface area available for microorganisms (Stachowiak-Wencek et al., 2021). Pretreatment which affects the chemical structures of biomass promotes the effective enzymatic conversion of carbohydrate polymers into monomeric sugars. For example, chemical treatment with base results in breaks in lignin structure and breaking bonds between lignin and other carbohydrates in biomass. Also, alkali treatment reduces the degree of cellulose polymerization and crystallinity, making them more available for microorganisms (Zborowska et al., 2022).

Another method recently studied extensively to enhance AD is biochar supplementation. Biochar (BC) is a carbonaceous material made during the pyrolysis of biomass. Biochar due to its specific properties can promote the AD process and improve its stability. BC supplementation works at many levels and stages of AD. It is stated that BC has adsorption and immobilization ability of ammonia, heavy metals, and toxins. Besides, AD microorganisms can attach to the highly porous surface of BC which promotes an increase in microorganism populations. In the case of organic overloading, BC can absorb generated metabolites improving process stability. The stability of the process is also improved by BC buffering ability, which comes from the presence of functional groups (–OH, –COOH, $–NH_2$), alkali metals ions ($Na^+$, $K^+$), and alkaline-earth metals ions ($Ca2^+$, $Mg^{2+}$) (W. Zhao et al., 2021). BC is also considered a conductive material promoting direct interspecies electron transfer (DIET) between syntrophic bacteria and methanogens. As a result enhances the syntrophic conversion of organic substances to methane, increasing process stability and decreasing the lag phase (Chen et al., 2022). Nevertheless, the effect of biochar supplementation depends on biochar properties, amounts of added BC, and AD characteristics. Due to the abundance of possible AD substrates and BCs properties, there is a need for more research on biochar addition to the AD process to find the right biochar properties and their amounts (Syguła et al., 2022). Biochar properties significantly depend on the substrate used in the pyrolysis process and pyrolysis conditions (temperature, residence time, heating rate, etc.). In general, the higher temperature of the pyrolysis the better quality of biochar is obtained. Nevertheless, in some cases, with increasing temperature, important properties deteriorate. Moreover, the higher the pyrolysis temperature and time the higher the cost of BC production (Morales et al., 2015).

Taking into account that BSG is abundant material that can be processed in AD, and BC can improve the AD process, in this study, the effect of the addition of different biochars made from substrates of AD was tested. According to our knowledge, there is no other research except Dudek et al., 2019, where BC made at 300°C from BSG was added to the AD process. The idea of using the substrate for BC production came from the fact that such BC can be produced in a biogas plant using residual heat from the CHP unit. The temperature of exhaust gases from a gas turbine differs from 400 to 600°C (OGL, 2021), while residual heat consists of around 70% of all heat produced in a biogas plant (Sobol et al., 2021). In the case of significant improvement of the AD process efficiency by BC such combined processes could be beneficial to the environment and economy.

## 2. MATERIALS AND METHODS

Firstly, materials used in the study were collected and prepared for analysis. After preparation, materials were subjected to analyses to reveal their characteristics. Then, the biochemical biogas potential test (BBPT) was performed with different biochars added to the process. Next, AD data were used to determine process kinetics. Finally, statistical analyses were performed to find the effect of BC supplementation on biogas production, kinetics, and process efficiency.

### 2.1 Materials

In the study, liquid digestate, the brewer's spent grain, and biochars were used (Figure S1 a-c). The digestate (D) used as inoculum in batch tests was collected from a 1 $MW_{el}$ commercial biogas plant (Bio-Wat Sp. Z o. o., Świdnica, Poland). The biogas plant was fed mainly with maize silage and other unspecified seasonal agricultural substrates. The digestate was collected from a post-fermentation chamber and placed in a plastic canister with a total volume of ~100 $dm^3$. The same day, digestate was taken to the laboratory where it was strained through a tetra cloth diaper to remove large solids particles and other solid contaminations. As a result, two digestate fractions were obtained, solid and liquid respectively. The solid digestate was ejected, while liquid digestate was stored in plastic containers in a laboratory incubator (POL-EKO-APARATURA, model ST 3 COMF, Wodzisław Śląski, Poland) at 4°C.

The main substrate used in the batch tests was brewer's spent grains (BSG). BSG was obtained from a laboratory-scale beer production installation (Wroclaw University of Environmental and Life Sciences, Wrocław, Poland) as a residual after the beer production process. The beer was made from a mashed pilsner malt Viking Malt (Strzegom), produced from malting barley. After the beer production process, the BSG was dried at 80°C to dry mass in a laboratory dryer (WAMED, model KBC-65W, Warsaw, Poland). A standard drying temperature for biomass (105°C) was not used to prevent a possible occurrence of Maillard's reactions. The dry BSG was stored at -31°C in a laboratory freezer (Electrolux, model EC5231AOW, Jászberény, Hungary).

Biochars (BC) were made from brewery-spent grains. According to the previous methodology, biochars were produced at temperatures 300, 450, and 600°C respectively (Świechowski et al., 2020). In short, biochars were made using a laboratory muffle furnace (SNOL, model 8.1/1100, Utena, Lithuania). Around 300 g of dry BSG was placed in the glass tray and placed into the furnace chamber (Figure S1 d). Afterward, the chamber was filled with $CO_2$ inert gas, and the furnace was turned on. The $CO_2$ was supplied into the chamber during the whole pyrolysis process to keep an inert atmosphere. The heating rate of 50°C·$min^{-1}$ was used to heat the reactor from room temperature (~20°C) to the setpoint temperature. The material was pyrolyzed at setpoint temperature for 60 minutes. After carbonization, the furnace was turned off and left to cool. Thus, produced biochars were stored in plastic bags at room temperature.

### 2.2 Methods

#### 2.2.1 Materials analyses

Basic and elemental analyses were performed on the study's materials, including the liquid digestate, the spent grain from the brewery, the biochar, and the process residues from the biochemical biogas potential test (BBPT). The basic analysis included total solids (TS) and volatile solids (VS), while the elemental analysis include carbon (C), hydrogen, (H), nitrogen (N), sulfur (S), and oxygen (O). Also, pH and electrical conductivity were measured (EC).

BSG and biochars were additionally subjected to proximate analysis, specific surface area (SSA) determination analysis, FTIR analysis, cation exchange capacity (CEC) determination analysis, and volatile organic compounds (VOC) analysis. The proximate analysis consists of moisture content (MC), volatile matter (VM), fixed carbon (FC), ash content (AC), and high heating value (HHV). Alongside SSA, total pore volume <50 nm ($V_t$), and average pore size <50 nm (L) were analyzed. Used equipment and methods were summarized in the Supplementary content, Table S1.

#### 2.2.2 Biochemical biogas potential test

Biochemical biogas potential tests (BBPT) were performed using the OxiTop® Control AN measuring system (Oxitop Control AN6, Weilheim, Germany) and laboratory incubator (POL-EKO-APARATURA, ST 3 COMF, Wodzisław Śląski, Poland), Figure S1 e,f. The OxiTop system consists of glass bottles (reactor chamber), head adapters, pressure measuring heads, and a reading pilot. The reactor has a total volume of 1 $dm^3$ and is ended with three stubs. The side stubs are for biogas collection/pressure release while the middle stub is for pressure measuring head. The head is connected to a reactor by an adapter. The principle of using BPPT is to measure the pressure increase caused by produced biogas and its recalculation to the volume of produced biogas in standard conditions.

BBPT was performed in 3 setups that were duplicated. One setup analyzed the effect of one biochar (BC300, BC450, BC600). During each setup, 10 reactors were used. Always 1 reactor contained inoculum, 1 reactor contained inoculum and BSG and 8 reactors contained inoculum, BSG, and biochars in shares ranging from 1 to 8% by TS of the BSG. The substrate-to-inoculum ratio (SIR) was kept around 0.80-0.86 by VS (0.48-0.52 by TS, ~0.10 by wet mass). Each reactor was filled with 160 g of liquid digestate (inoculum) and around 3.4 g of dry BSG mixed with BC. The mass of specific materials placed into the reactors and the reactors' main parameters were summarized in Table S2.

### *2.2.3 Biogas production, kinetics, and process efficiency determination*

The results of the BBPT were subjected to kinetics parameters determination by estimation to the first-order kinetic model, Equations (1) and (2). The model provides information about the constant reaction rate (k), the estimated maximum biogas production potential (emBBP), methane production rate (r), and cumulative biogas production (BBP) after a given time (t). The kinetics determination was performed using Statistica 13.0 software (TIBCO Software Inc., Palo Alto, CA, USA).

$$BBP = emBBP \times \left(1 - e^{(-k \times t)}\right) \quad (1)$$

$$r = k \times emBBP \quad (2)$$

where:

- BBP is the cumulative biogas production after a given time t, ml×$g_{VS}^{-1}$;
- emBBP is the estimated value of experimental maximum biogas production, ml×$g_{VS}^{-1}$;
- e is the mathematical constant (a.k.a. Euler's number) equal to ~2.718, -;
- k is constant biogas production rate, $d^{-1}$;
- t is process time, d;
- r is biogas production rate, ml×$(g_{VS} \times d)^{-1}$.

To determine process efficiency (degree of substrate conversion into biogas), theoretical biochemical biogas potential (TBBP) production was calculated according to Equation (3) which is Boyle's modification of the Buswell and Mueller stoichiometric formula.

$$C_aH_bO_cN_dS_e + \left(a - \frac{b}{4} - \frac{c}{2} + \frac{3d}{4} + \frac{e}{2}\right) H_2O \rightarrow \left(\frac{a}{2} + \frac{b}{8} - \frac{c}{4} - \frac{3d}{8} - \frac{e}{4}\right) CH_4 + \left(\frac{a}{2} - \frac{b}{8} + \frac{c}{4} + \frac{3d}{8} + \frac{e}{4}\right) CO_2 + dNH_3 + eH_2S \quad (3)$$

where:

- $C_aH_bO_cN_dS_e$ is the elemental composition of the substrate, C – carbon, H – hydrogen, O – oxygen, N –nitrogen, S – sulphury, and a, b, c, d, e stands for molar % share of specific elements contained in the volatile solids of the substrate.
- $H_2O$ is the water needed for substrate decomposition, mol;
- $CH_4$ is the methane, mol;
- $CO_2$ is the carbon dioxide, mol;
- $NH_3$ is the ammonia, mol;
- $H_2S$ is the hydrogen sulfide, mol.

The description of how to calculate TBBP using Equation (3) is presented elsewhere (Świechowski et al., 2022). Afterward, substrate conversion into biogas (BD) was calculated using data from the BBPT experiment and TBBP according to Equation (4).

$$BD = \frac{EBMP}{TBMP}\ x\ 100 \quad (4)$$

where:

- BD is the substrate biodegradation (degree of substrate converted into biogas), %;
- EBMP is the experimental biochemical biogas potential, ml×$g_{VS}^{-1}$;
- TBMP is the theoretical biochemical biogas potential, ml×$g_{VS}^{-1}$.

Next, to determine quantitatively the effect of biochar added on process efficiency, the biogas production effect (BPe) was calculated according to Equation (5). BPe provides information on how much percent biogas production increased/decreased after biochar was added in comparison to control without biochar added.

$$BPe = \frac{Biogas_{with\ BC} - Biogas_{without\ B}}{Biogas_{without\ B}}\ x\ 100 \quad (5)$$

where:

- BPe is the biogas production effect, %;
- $Biogas_{with\ BC}$ is the biogas produced from a substrate without biochar added, ml;
- $Biogas_{without\ BC}$ is the biogas produced from a substrate with biochar added, ml.

### *2.2.4 Statistical analyses of the BC effect on the AD*

Due to a large number of BBMP measurements, its typical presentation in the form of a line diagram with standard deviations is unreadable. For the better visualize obtained data and the effect of biochar added on process kinetics and efficiency, the regressions using the response surface area model were performed. To study the effect of biochar dose, and temperature of its production on process kinetics and efficiency it was assumed that the independent variables are biochar share, and temperature of biochar production while the dependent variables are emBBP, r, k, BD, and BPe. The regression analysis was performed using Statistica 13.0 software (TIBCO Software Inc., Palo Alto, CA, USA).

To check if between obtained results are statistically significant differences, ANOVA with post hoc Tukey test at the level of α = 0.05 was performed using Statistica 13.0 software (TIBCO Software Inc., Palo Alto, CA, USA).

## 3. RESULTS AND DISCUSSION

### 3.1 Materials analyses

In Table 1 and Table 2, the properties of the materials used in the study are presented. The moisture content of fresh BSG was 79.6% and dry mass consist of 20.4%. BSG was characterized by high organic matter content since the volatile solids were 96.2%, and ash content was only 3.8%. The main elements in BSG were carbon 48.6%, and oxygen 35.0%. The overall energy potential of BSG was 20.76 MJ×kg$^{-1}$ (Table 1).

The obtained biochars made from BSG had also a high amount of organic matter, the VS was in the range of 94.4-85.8%, and its value decreased with the increasing temperature of the pyrolysis. Though a similar amount of VS in comparison to BSG was observed, most of the organic matter was in the form of fixed carbon. The FC in biochar was between 42.8-70%, while BSG had only 14.5% of FC (Table 1). More so, this is also visible in the carbon content (C) which relative amount increased in each biochar from 48.6% in unprocessed BSG to 77.7% in the BC600. With increasing pyrolysis temperature, also a decrease in H, N, S, and O was observed. The change is very significant, especially in the case of oxygen which decreased from 35% for BSG to 1% for BC600 (Table 1). The basic properties of studied BSG and produced BC (Table 1) are similar to those from the literature. The BSG is characterized by the VM of 77-80.3%, the FC of 16.1-19.3%, the AC of 2.1-6.2%, the C of 48.8-49.2%, the H of 6.5-6.8%, the N of 3.9-4.4%, the O of 36-36.8% and HHV of 18.6-21.7 MJ×kg$^{-1}$. (Balogun et al., 2017; Sanna et al., 2011; Sieradzka et al., 2022). Also, the properties of BCs and trends in changes in their properties are similar to other studies. Only oxygen content in BC450 and BC600 is much lower than in the work of (Sanna et al., 2011) where biochars produced at 460-540°C were characterized by oxygen content of 31.7-24.5%. Nevertheless, such differences may be due to different methods of pyrolysis.

For BSG, determination of specific surface area (SSA), total pore volume <50 nm ($V_t$), average pore size <50 nm (L), and cation exchange capacity failed. It was due to the physical characteristic of the BSG. The pore size and its amount were too small to be measured and the procedure for CEC determination in biochars turned out to be not suitable for BSG. BSG's pH was 6.4 and its electrical conductivity was 718 µS×cm$^{-1}$. For comparison, biochar used in the study had similar pH (5.92-7.85), but it had much lower electrical conductivity (214-332 µS×cm$^{-1}$) (Table 2) which is surprising since biochar is considered the material that supposes to increase conductivity and electron transfer in the anaerobic digestion process and enhance DIET mechanism (Z. Zhao et al., 2020). With increasing pyrolysis temperature, the SSA of biochars increased significantly from 0.5 m$^2$×g$^{-1}$ to 292 m$^2$×g$^{-1}$ for BS300 and BS600 respectively. A similar trend was observed for total pore volume which increased from 0.001 to 0.137 cm$^3$×g$^{-1}$ for the same biochars. Though SSA and $V_t$ increased with pyrolysis temperature, the average pore size slightly decrease from 4.6 nm to 1.9 nm. At the same time, cation exchange capacity increased from 8.7 cmol(+)×kg$^{-1}$ to 31.8 cmol(+)×kg$^{-1}$ (Table 2). Produced BCs properties slightly differ from the work of (Xi et al., 2014) which produced BC from BSG at temperatures from 300°C to 700°C and a processing time of 2-4 h. Biochars made at 300, 400, 500, and 600°C were characterized by pH of 10.3-11.5, CEC of 18.5-22.3 cmol(+)×kg$^{-1,}$ and SSA of 5.86-10.6 m$^2$×g$^{-1}$ (Xi et al., 2014). This shows that the initial substrate used for pyrolysis and pyrolysis procedures affects significantly biochar properties. The most important parameter that affects BC properties is quality and type of substrate, process temperature, and pyrolysis type (Morales et al., 2015). Nevertheless, the most commonly used biochar for AD is BC made of wood or agricultural residues at the temperature of 300-800°C, and as a result, their properties differ significantly (W. Zhao et al., 2021). In general, the most desirable BC for AD is the one that has the potential to adsorption of ammonia, heavy metals, and excess VFAs (volatile fatty acids), and immobilize toxic substances (e.g. antibiotics) (Ngo et al., 2022). The adsorption and immobilization ability come from the specific surface area, porosity, and functional groups placed on the biochar surface. It is worth noting that too strong an absorption ability or a too large dose of BC may inhibit AD microorganisms as well (Ambaye et al., 2021). Another important BC feature is alkaline pH and the ability to increase reactor stability due to buffer capacity enhancement. It is possible due to the presence of alkaline functional groups and metal ions (Fidel et al., 2017).

**TABLE 1:** The basic characteristic of BSG and BCs.

| Material | TS** (%) | VS (%) | MC** (%) | VM* (%) | FC* (%) | AC* (%) | C* (%) | H* (%) | N* (%) | S* (%) | O* (%) | HHV (MJ×kg$^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| BSG | 20.4±0.3 | 96.2±0.0 | 79.6±0.3 | 82.4±0.3 | 14.5±0.4 | 3.1±0.2 | 48.6±0.1 | 7.0±0.0 | 4.4±0.2 | 2.0±0.2 | 35.0±0.3 | 20.76 |
| BC 300 | 97.1±0.7 | 94.4±0.1 | 2.9±0.7 | 52.8±0.7 | 42.8±0.9 | 4.5±0.4 | 60.0±0.3 | 5.3±0.0 | 4.8±0.2 | 1.6±0.2 | 23.9±0.3 | 26.00 |
| BC 450 | 99±1.2 | 89.3±3.0 | 1.0±1.2 | 21.9±1.2 | 68.9±1.2 | 9.2±0.3 | 69.6±4.1 | 3.8±0.2 | 5.0±1.0 | 1.1±0.0 | 11.3±3.7 | 26.18 |
| BC 600 | 96±0.4 | 85.8±0.2 | 4.0±0.4 | 17.3±0.4 | 70.0±0.2 | 12.8±0.3 | 77.7±0.5 | 2.8±0.0 | 4.7±0.3 | 1.0±0.1 | 1.0±0.9 | 24.75 |

**as dry base, **as received base*

**TABLE 2:** Additional characteristics of BSG and BCs.

| Material | SSA (m$^2$×g$^{-1}$) | $V_t$ (cm$^3$×g$^{-1}$) | L (nm) | CEC (cmol(+)×kg$^{-1}$) | pH | EC (µS×cm$^{-1}$) |
|---|---|---|---|---|---|---|
| BSG | 0 | 0 | 0 | - | 6.4 | 718 |
| BC300 | 0.5 | 0.001 | 4.6 | 8.7 | 5.92 | 214 |
| BC450 | 3.3 | 0.004 | 4.4 | 14.2 | 6.03 | 223 |
| BC600 | 292 | 0.137 | 1.9 | 31.8 | 7.85 | 332 |

To check, if volatile organic compounds contained in biochar may affect methane fermentation, VOCs determination in BSG and BCs was performed. VOCs are a very vast group of organic compounds and some of them are toxic. The work in the field of biochar shows that during the thermal conversion of organic materials (torrefaction, pyrolysis, etc.) numerous (VOCs) are formed. Due to the porous structure of biochar and the condensation of residual vapors that take place at the last step of pyrolysis (cooling), VOCs stay on the biochar surface, and biochar itself may become a source of VOCs pollutants (Łyczko et al., 2021). Therefore in this study, we wanted to check if VOCs contained in biochars that were applied to the AD process could affect biogas production.

The shortlist of most abundant VOCs (compound share >5%) found in studied materials is presented in Table 3, while a full list of all detected VOCs is presented in the Supplementary content, (Table S3). It turns out that there were no VOCs in the BC450 and BC600.

This is probably due to high pyrolysis temperature and long residence time which result in total organic compounds decomposition (Białowiec et al., 2018). The pyrolysis of BSG at 300°C resulted in a change in the chemical composition of volatile organic compounds and their number. The unprocessed BSG had 37 VOCs compounds, while BC300 had 44 (Table S3). The main VOCs in BSG were 1-Pentanol, 4-methyl- 20.89%, Ethyl amylketone 18.86% and Cyclobutane, 1,2-bis(1-methylethenyl)-, trans- 8.79%, while main VOCs contained in BC300 were Cyclobutane, 1,2-bis(1-methylethenyl)-, trans- 24.95%, Pentanal, 3-methyl- 14.74% and Thuja-2,4(10)-diene 10.35% (Table 3). It is worth noting that the relative amount of Cyclobutane, 1,2-bis(1-methylethenyl)-, trans- increased from 8.79% in BSG to 24.95% in BC300.

Due to the lack of VOCs in BC450 and BC600, it may be concluded that VOCs in biochars produced at higher temperatures than 450°C do not affect anaerobic digestion, and other mechanisms need to be investigated.

The FTIR spectroscopy was performed to determine functional groups present on BSG and BCs surfaces. The spectra with the largest peaks are shown in Figure 1.

The BSG spectra show the largest peaks at 3298, 2924, 2856, 1743, 1632, 1536, 1150, 1075, and 1123 cm$^{-1}$. Due to BSG being lignocellulose materials, most of the peaks are considered to come from the main polymers which are hemicellulose, cellulose, and lignin (Nasir et al., 2021). A signal at 3298 cm$^{-1}$ is attributed to O-H and N-H bonds that come from hydroxyl, amine, and amide groups. While hydroxyl groups are common in lignocellulosic biomass, the presence of amine and amide groups results from the high content of protein contained in BSG (Hejna et al., 2021). The peaks at the band region of 3000-2800 cm$^{-1}$ show C–H stretching related to the presence of hemicellulose and cellulose, while peaks at the band region of 1700-1600 cm$^{-1}$ shows the presence of amide I and amide II or the aromatic hydrocarbons of lignin (Naibaho et al., 2021). The peak at the band region of 1550-1500 is probably related to N-O stretching resulting from the presence of some nitro compound (IR Spectrum Table&Chart, 2022). The peaks at the band range of 1100-1000 cm$^{-1}$ indicate stretching of C–O–C that comes from the presence of functional groups of aliphatic ethers (Naibaho et al., 2021). Most of the peaks found on the BSG were not observed on the BCs or were less intense. For example, no peaks in the range of 1600-3300 cm$^{-1}$ were found for BC450 and BC600, though smaller peaks for BC300 in the range of 2800-3300 cm$^{-1}$ can be observed. Also in the range of 1000-1530 cm$^{-1}$, almost all peaks visible on BSG are not present at BCs (Figure 1). Nevertheless, in the case of BC450 and BC600, some peaks that were not present on BSG can be found at 877 and 742 cm$^{-1}$. These changes in spectra indicate structural changes in BCs composition occurred during the pyrolysis. Flattening of absorbance at the range of 1000-1530 cm$^{-1}$ shows a decrease in the C-H stretching bands in the biochars which are related to aliphatic compounds (Borel et al., 2020).

## 3.2 Biogas production, kinetics, and process efficiency

In Figures S1-S3 (Supplementary content), mean values from the biochemical biogas potential test (BBPT) are presented. It can be seen that biogas production from BSG, after 28 days varied from 500 ml×$g_{VS}^{-1}$ to 650 ml×$g_{VS}^{-1}$, while theoretical biochemical biogas potential (TBBP) production calculated according to Equation (3) is 1020 ml×$g_{VS}^{-1}$. It means that on average, BSG was converted 56% into biogas while 44% of BSG was not utilized. Due to numerous results presented in Figures S1-S3, the response surface model was used to show the main effects of biochar addition. The results of the regression analysis are presented in Figure 2.

Results show the effect of biochar dose (from 1% to 8%) and biochar production temperature (from 300°C to 600°C) on the biogas production effect (Figure 2a) and substrate biodegradation (Figure 2b). Due to the complex

**TABLE 3:** Shortlist of most abundant volatile organic compounds contained in materials.

| BSG | | BC300 | |
|---|---|---|---|
| **Compound name** | **%** | **Compound name** | **%** |
| 1-Pentanol, 4-methyl- | 20.89 | Cyclobutane, 1,2-bis(1-methylethenyl)-, trans- | 24.95 |
| Ethyl amylketone | 18.86 | Pentanal, 3-methyl- | 14.74 |
| Cyclobutane, 1,2-bis(1-methylethenyl)-, trans- | 8.79 | Thuja-2,4(10)-diene | 10.35 |
| 2,3-Butanediol | 8.78 | Naphthalene, 1,2,3,4,4a,8a-hexahydro- | 6.86 |
| Butanoic acid | 6.32 | Benzoyl isothiocyanate | 5.32 |
| Hepten-3-ol | 5.77 | Furan <2-butyl-> | 5.03 |

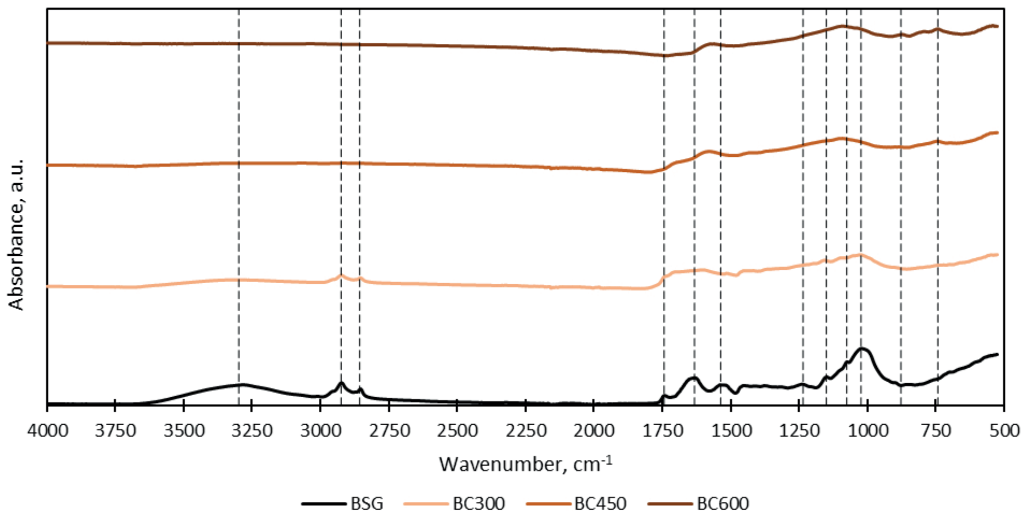


**FIGURE 1:** FTIR spectra of BSG and its biochars.

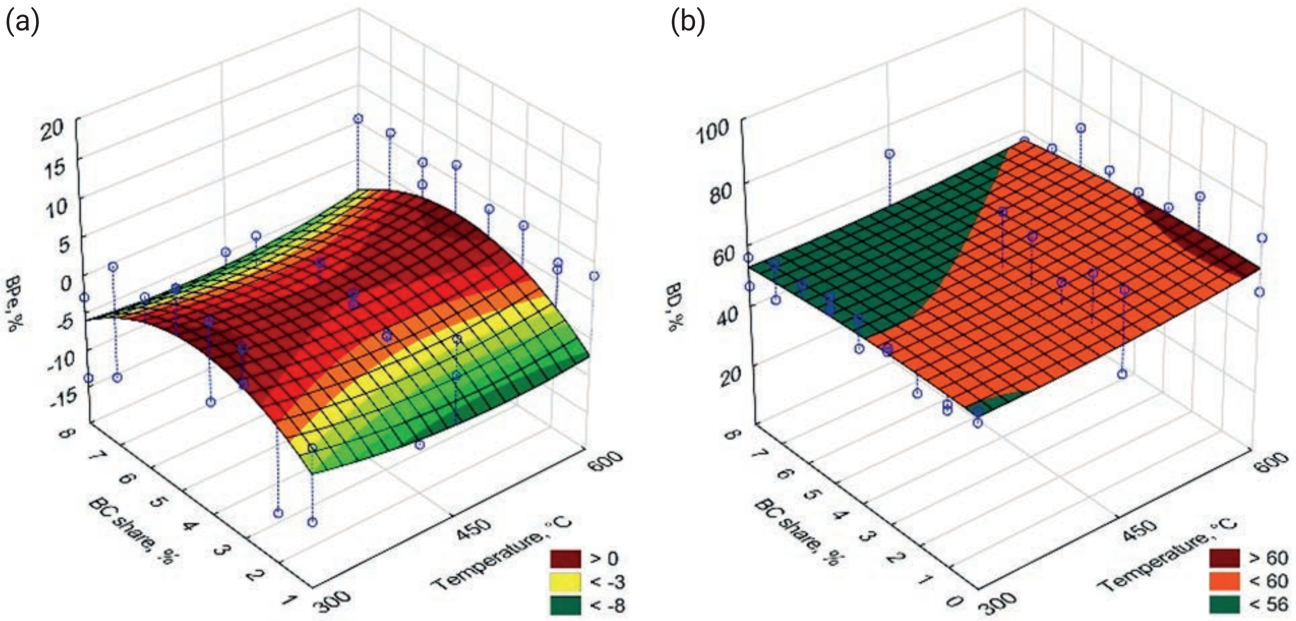


**FIGURE 2:** a) biogas production effect (BPe), b) substrate biodegradation (BD).

nature of biochar and the biochar mechanisms affecting anaerobic digestion, a large discrepancy in measurements can be observed (blue circles). It can be observed that biochar share, regardless of biochar production temperature (and thus its properties) affects BPe. For BC shares above 6% and lower than 3%, the response surface takes negative values of the BPe though some measurements (blue circles) are far from negative values (Figure 2a). For BD values, the response surface area shows the highest values (>60%) for biochar made at 600°C and BC dose up to 3% though some measurements for 450°C (BC share 1-4%) are much over >60%. (Figure 2b).

The effects of biochar share and biochar production temperature on biogas production kinetics are presented in Figure 3. The estimated value of experimental maximum biogas production (emBBP), the constant biogas production rate (k), and the biogas production rate (r) varied from ~620-820 ml×$g_{VS}^{-1}$, ~0.05-0.08 $d^{-1}$, and ~42-60 ml×$(g_{VS}×d)^{-1}$, respectively (Figure 3).

Data shows that the emBBP reaches the highest value for BC produced at 450°C and its share of 2-6%, while lower and higher BC doses and other temperatures result in an emBBP decrease (Figure 3a). At the same time, the k shows a different trend, and the highest k value is obtained at a BC share of 0-3% and a temperature of 600°C (Figure 3b). It seems that k has the opposite trend in comparison to the emBBP, the higher emBBP, the lower the k is. On the other hand, the biogas production rate that results from the multiplication of emBBP and k shows that r has the highest value at a BC dose of 0-3% (Figure 3c).

The effect of biochar addition to the AD process is visible. BC supplementation, on one hand, increases maxi-

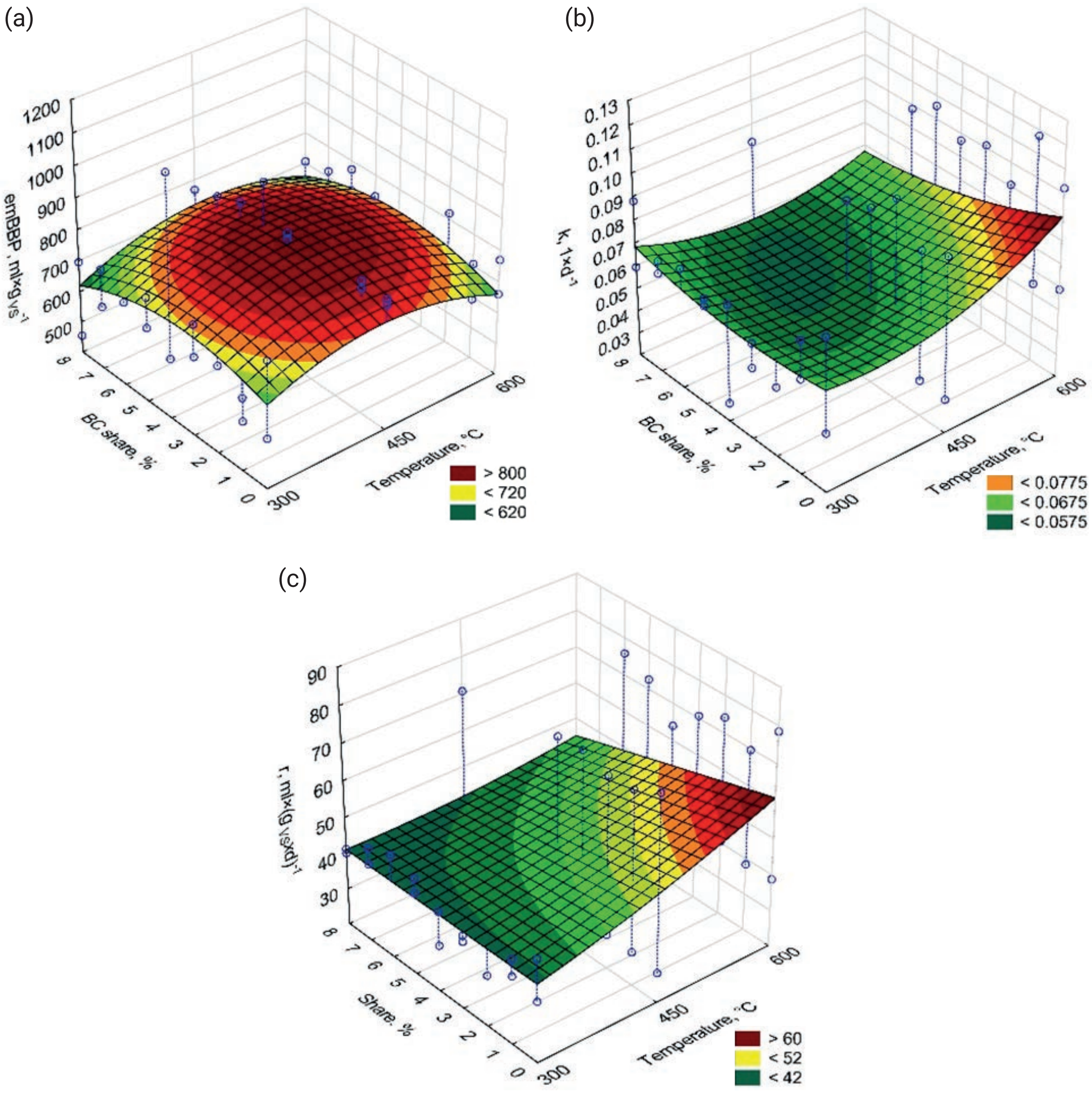


**FIGURE 3:** Biogas production kinetics, a) the estimated value of experimental maximum biogas production (emBBP), b) constant biogas production rate, c) biogas production rate.

mum biogas production (emBBP) and on the second-hand results in a constant biogas production rate decrease (k). It means that somehow BC increases the amounts of biogas that can be produced from the substrate, and at the same time decrease the speed of its production. Moreover, the biogas production rate (r) that combines emBBP and k is shown to increase with increasing BC production temperature and to decrease with increasing BC share. Though the response surface area shows that the greatest positive effect of BC addition can be obtained for BC600 at the share of 1-3%, all differences in the results turned out to be statistically insignificant ($p<0.05$). The lack of statistical significance may question obtained results and the sense of adding BC made from a substrate to methane fermentation of BSG. Nevertheless, the lack of statistically significant differences comes from the large discrepancy in the measurements and the fact that there are probably other factors that should be taken into account in future research.

To sum up, the theoretical biogas production potential of studied BSG was 1020 ml×$g_{VS}^{-1}$ and during 4 weeks of BSG methane fermentation, around 60% (500-650 ml×$g_{VS}^{-1}$) of this value was obtained in batch reactors. The addition of BC made from the substrate affects the amount of obtained biogas (BPe), substrate conversion (BD), and kinetics parameters of biogas production (emBBP, k, r) sometimes leading to a decrease and sometimes to an increase in its value. Though biochar addition does not make a statistically significant difference (p<0.05), to maximize biogas production from BSG, BC450 at the share of 1-4% can be used (Figures 2a and 3a), and to maximize biogas production rate BC600 at the share of 1-3% (Figure 3b-c). Unfortunately, due to the high dispersion of the obtained BBPT results and its complexity, it was not possible to assign specific biochar features that lead to an increase/decrease in biogas production during the anaerobic digestion of BSG.

The average methane concentration in biogas produced during AD of BSG is around 60% (Čater et al., 2015; Poulsen et al., 2017). Since the biogas production in this research varied from 500 to 650 $ml \times g_{VS}^{-1}$ (Figure S2-S4), it can be assumed that around 300-390 $ml_{CH4} \times g_{VS}^{-1}$ were produced. These results are similar to other studies. According to Oliveira et al., 2018, raw spent grain has a biomethane potential of 271-387 $ml_{CH4} \times g_{VS}^{-1}$. Gomes et al., 2021 studied the effect of BSG loading (8.3-19.7 $g \times L^{-1}$) and the AD temperature (31-59°C) on biomethane production. In a batch test at SIR of 0.5, after 21 days obtained methane yield of 81-290 $ml_{CH4} \times g_{VS}^{-1}$, Interestingly for the AD at 35°C and BSG concentration of 10 $g \times L^{-1}$ obtained the highest biomethane yield. This show that biogas and biomethane yields are sensitive to initial AD conditions, substrate quality, and hydraulic retention time (HRT). In theory, maximal biomethane yield is obtained when the organic loading rate (ORL) is low and hydraulic retention time (HRT) is long. (Gunes et al., 2019). In the case of batch reactors, such conditions are mainly obtained by the proper substrate-to-inoculum ratio (SIR ~ 0.5 by VS), and process time of around 30 days. Though a shorter time can be applied if biogas/biomethane production over three following days is lower than 1% of the already cumulated biogas/methane (Filer et al., 2019). Due to the biogas yield being similar to data obtained in other studies, it can be assumed that BBPT tests were performed correctly.

Though there is a lot of research about the supplementation of carbonaceous materials to the AD process, according to our knowledge only two studies analyzed the effect of the BC addition to the AD of BSG. Dudek et al., 2019 studied the effect of biochar addition made from BSG at 300°C and doses ranged from 1% to 50% share by TS of BSG, while Mainardis et al., 2019 studied the effect of biochar made from red spruce woodchips at a temperature of 650°C at a dose of 0.2 $g_{BC} \times g_{VS}^{-1}$ of BSG. In the case of Dudek et al., 2019, the AD process took 21 days, and the biogas yield differed from 61.3 to 122.0 $ml \times g_{VS}^{-1}$. The highest increase in biogas production was observed at 5% BC share where the highest value of 122 $ml \times g_{VS}^{-1}$ was obtained. In comparison to the control (92.3 $ml \times g_{VS}^{-1}$) it was an increase of 32%. On the other hand, BC doses over 20% by TS of BSG lead to a decrease in biogas yield and suggest that an overdose of BC may inhibit the AD process (Dudek et al., 2019). In the case of Mainardis et al., 2019 two types of BSG were tested, BSG1 and BSG2 respectively. The process took around 19 days and methane yield in control samples differed from 300 $ml_{CH4} \times g_{VS}^{-1}$ for the BSG2 that acidified the process after 7 days, up to the 360 $ml_{CH4} \times g_{VS}^{-1}$ for the BSG1 that worked normally by 19 days. Interestingly biochar addition help to overcome acidification for BSG2 increasing methane yield by +26.6% up to 388 $ml_{CH4} \times g_{VS}^{-1}$, while for BSG1 a reduction to 342 $ml_{CH4} \times g_{VS}^{-1}$ was noted (Mainardis et al., 2019).

Contradictory results can be also found for other substrates and BC types. In the work of (D. C. Shin et al., 2022), used five food-waste to produced biochars with the following quantities (0.1, 0.5, 1.0, 3.0, 5.0% by volume, (v/v%)) were added to the AD of sewage sludge processed at 40°C. The BC was made at 500°C and a retention time of 10 min and showed biogas and biomethane production increase with increasing dose. For the 5% variant, the biogas production during the 60-day process was improved by almost 20%, while methane concentration increased from 50.6% to 55.8% (D. C. Shin et al., 2022). On the other hand (Wambugu et al., 2019) performed batch AD of food waste with BC made of brewery residues and waste wood at doses of 0.7, 1.3, 2.0, 2.7, 3.3, 5.0, and 8.0 $g_{BC} \times L^{-1}$. The AD was performed at 30°C and lasted 6 days after the process stopped due to acidification. Moreover, the addition of biochars lowered the amount of biogas produced by the control with only food waste. Interestingly wood waste biochar at a dose of 8 $g_{BC} \times L^{-1}$ used in a continuous up-flow anaerobic sludge blanket reactor (UASB) results in significant biogas production enhancement and increased chemical oxygen demand removal efficiency (Wambugu et al., 2019). The biochar addition does not only show complex effects on biogas production but also on the characteristics of the microorganisms. In the work of (Zhang et al., 2019), nine biochars made from three different materials at 400, 500, and 600°C were added to the AD of sewage sludge at a dose of 8 $g_{BC} \times L^{-1}$ and chosen biochars were tested at doses 6.2, 15.9, 26.1, and 34.2 $g_{BC} \times L^{-1}$. In most cases, the BC supplementation enhanced AD process stability by increasing buffering capacity, releasing volatile fatty acid accumulation, and alleviating ammonia inhibition. Still, excessive BC supplementation turned out to be inhibitory. Interestingly BC addition increased the abundance of acetoclastic methanogens that convert acetate ($CH_3COOH$) to methane and carbon dioxide, while reducing the abundance of hydrogenotrophic methanogens that produce methane from hydrogen and $CO_2$ (Zhang et al., 2019). That is opposite to the findings of (S. Wang et al., 2022), that added straw biochar (600°C for 20 min) at 7.1 $g_{BC} \times L^{-1}$ to the AD of cow dung. (S. Wang et al., 2022) also obtained methane production enhancement after biochar addition but this effect was due to an increase of hydrogenotrophic methanogens abundance, while acetoclastic methanogens such as Methanosaeta decreased massively (S. Wang et al., 2022). On the contrary results of (Masebinu et al., 2021) showed that for a well-working AD system without any severe disturbances, biochar does not improve methane production nor decrease it and microbial community composition is not altered.

Such conflicting effects also applied to the data obtained in this study show that in some specific cases, biochar may help to improve the process while in others can inhibit it. The reason for that may be the initial properties of the used inoculum, substrate, biochar, and AD conditions. Proper initial conditions in the AD process allow avoid of volatile fatty acid accumulation and provide enough time for microorganisms to convert almost all available organic matter (Filer et al., 2019). Too high of organic input cause a drop in the pH that may affect the activity of some AD microorganisms. AD consists of 4 stages, hydrolysis, acidogenesis, acetogenesis, and methanogenesis. In each stage, other groups of microorganisms play a vital role. These groups are respectively hydrolytic, fermentative (acidogenic), syntrophic (acetogenic), and methanogenic microorganisms. In terms of optimal pH, there are two groups. Acid-producing bacteria (acidogenic) with optimal pH of 5.5-6.5, and methane-producing bacteria (methanogens) with optimal pH of 6.6-7.5. The acid-producing bacteria are less sensitive to lower pH values. Therefore the high pH drop related to the depletion of buffer capacity caused by the overloading of the reactor can inhibit methanogens activity and result in process failure (Khanal, 2008). There are pieces of evidence that biochar can enhance the buffer capacity (Shi et al., 2017) and allows for the mitigation of acidification in the AD process (D. Wang et al., 2017). In the research of Wang et al., 2017, the addition of 5% of BC by weight of the total loading to an overloaded AD reactor (50 $g_{TS} \times kg^{-1}$) was studied. The results showed that without biochar, the process was not able to start while BC addition mitigated acidification and kept biogas production going.

It seems that in the case of the performed research, the lack of significant biogas production improvement by supplementation of BC300, BC450, and BC600 could be related to the fact that, the AD process was performed in optimal conditions. And therefore, biochar buffering properties weren't used. Moreover, the observed decrease in some cases after biochar supplementation may come from the fact that biochar has strong adsorption and immobilization ability. Probably as a result of a lack of inhibitors like a too-high concentration of ammonia or VFAs, biochar adsorbs other microelements that could be necessary for microorganisms.

## 4. CONCLUSIONS

In this research, anaerobic digestion of BSG with BCs addition was performed. The theoretical biogas potential of tested BSG was 1020 $ml \times g_{VS}^{-1}$. The biogas yield after 28 days varied from 500-650 $ml \times g_{VS}^{-1}$ and around 60% substrate degradation was obtained. The kinetics parameters (emBBP, k, r) varied from ~620-820 $ml \times g_{VS}^{-1}$, ~0.05-0.08 $d^{-1}$, and ~42-60 $ml \times (g_{VS} \times d)^{-1}$, respectively. The addition of biochars showed complex and sometimes conflicting effects. As a result, no specific dependencies between the properties of biochar related to the temperature of its production and its dose were found. Though the differences in biogas production turned out to be statistically insignificant ($p<0.05$) due to the high disappearance in obtained data and conflicting effects, the response surface area analysis showed that to maximize biogas production, biochar made at 450°C at the share of 1-4% can be utilized, and to maximize the biogas production rate, the biochar made at 600°C at the share of 1-3% can be used. The comparison results with other works resulted in a hypothesis that the lack of biogas production improvement could be related to the fact that the AD process was performed in optimal conditions. As a result, biochar could not optimize process performance. In addition, a slight decrease in biogas production was probably due to biochar's strong adsorption and immobilization ability that in higher doses immobilized substances required by the AD's microorganisms.

Therefore, more research is required, where various initial conditions (with higher organic loading) and various BC concentrations would be tested. This would make it possible to determine the appropriate quantity of BC for the particular stress level of the reactor. Future research also should focus on the economic aspects of biochar supplementation and different strategies for BC acquisition. In the current study, BC was made from the substrate which due to the fact of being converted into BC could not be used during AD decreasing total methane production. For that reason, BC made from the AD digestate should be considered as a potential source for pyrolysis feedstock. Also, high-energy demand pyrolysis that requires dry feedstock could be replaced with the hydrothermal carbonization process (HTC) performed at lower temperatures (180-300°C). The HTC can produce biochar-like products (hydrochar) from both raw or anaerobically digested material with slightly difference in its properties (Catenacci et al., 2022). Also, HTC does not require a drying step and can be supplied with waste heat from biogas incineration in combined heat and power units.

## FUNDING

The research was funded by the National Science Centre, Poland, grant number 2019/35/N/ST8/02498, titled "Study of the influence of selected properties of biochar made from the substrate on the process of methane fermentation of brewer's spent grain".

## REFERENCES

Al-Wabel, M. I., Al-Omran, A., El-Naggar, A. H., Nadeem, M., & Usman, A. R. A. (2013). Pyrolysis temperature induced changes in characteristics and chemical composition of biochar produced from conocarpus wastes. Bioresource Technology, 131, 374–379. https://doi.org/10.1016/j.biortech.2012.12.165

Ambaye, T. G., Rene, E. R., Nizami, A. S., Dupont, C., Vaccari, M., & van Hullebusch, E. D. (2021). Beneficial role of biochar addition on the anaerobic digestion of food waste: A systematic and critical review of the operational parameters and mechanisms. Journal of Environmental Management, 290. https://doi.org/10.1016/j.jenvman.2021.112537

Amienyo, D., & Azapagic, A. (2016). Life cycle environmental impacts and costs of beer production and consumption in the UK. The International Journal of Life Cycle Assessment, 21(4), 492–509. https://doi.org/10.1007/s11367-016-1028-6

Arranz, J. I., Sepúlveda, F. J., Montero, I., Romero, P., & Miranda, M. T. (2021). Feasibility analysis of brewers' spent grain for energy use: Waste and experimental pellets. Applied Sciences, 11(6). https://doi.org/10.3390/app11062740

Balogun, A. O., Sotoudehniakarani, F., & McDonald, A. G. (2017). Thermo-kinetic, spectroscopic study of brewer's spent grains and characterisation of their pyrolysis products. Journal of Analytical and Applied Pyrolysis, 127, 8–16. https://doi.org/10.1016/j.jaap.2017.09.009

Beer market report. (2022). Beer Market Size, Share & COVID-19 Impact Analysis, By Type (Lager,Ale, Stouts, and Others), Packaging (Glass Bottle and Metal Can), Distribution Channel (On-trade and Off-trade), and Regional Forecast, 2021-2028. https://www.fortunebusinessinsights.com/beer-market-102489

Białowiec, A., Micuda, M., Szumny, A., Łyczko, J., & Koziel, J. A. (2018). Quantification of VOC emissions from carbonized refuse-derived fuel using solid-phase microextraction and gas chromatography-mass spectrometry. Molecules, 23(12). https://doi.org/10.3390/molecules23123208

Borel, L. D. M. S., Reis Filho, A. M., Xavier, T. P., Lira, T. S., & Barrozo, M. A. S. (2020). An investigation on the pyrolysis of the main residue of the brewing industry. Biomass and Bioenergy, 140. https://doi.org/10.1016/j.biombioe.2020.105698

Bougrier, C., Dognin, D., Laroche, C., & Cacho Rivero, J. A. (2018). Use of trace elements addition for anaerobic digestion of brewer's spent grains. Journal of Environmental Management, 223, 101–107. https://doi.org/10.1016/j.jenvman.2018.06.014

Catenacci, A., Boniardi, G., Mainardis, M., Gievers, F., Farru, G., Asunis, F., Malpei, F., Goi, D., Cappai, G., & Canziani, R. (2022). Processes, applications and legislative framework for carbonized anaerobic digestate: Opportunities and bottlenecks. A critical review. Energy Conversion and Management, 263, 115691. https://doi.org/10.1016/j.enconman.2022.115691

Čater, M., Fanedl, L., Malovrh, Š., & Marinšek Logar, R. (2015). Biogas production from brewery spent grain enhanced by bioaugmentation with hydrolytic anaerobic bacteria. Bioresource Technology, 186, 261–269. https://doi.org/10.1016/j.biortech.2015.03.029

Chen, L., Fang, W., Chang, J., Liang, J., Zhang, P., & Zhang, G. (2022). Improvement of direct interspecies electron transfer via adding conductive materials in anaerobic digestion: mechanisms, performances, and challenges. In Frontiers in Microbiology (Vol. 13). Frontiers Media S.A. https://doi.org/10.3389/fmicb.2022.860749

Cheri, C., Crandall, S., del Grande, D., Flores, T., Gilliland, M., Horwitz, L., Opela, C., Swersey, C., & Utz, M. (2014). Energy Usage, GHG Reduction, Effi ciency and Load Management Manual. https://www.brewersassociation.org/educational-publications/energy-sustainability-manual/

Cimini, A., & Moresi, M. (2021). Circular economy in the brewing chain. Italian Journal of Food Science, 33(3), 47–69. https://doi.org/10.15586/ijfs.v33i3.2123

Conway, J. (2021, October 15). Global beer production by region 2008-2020. Statista. https://www.statista.com/statistics/270270/worldwide-beer-production-by-region/

Conway, J. (2022a, March 16). European beer production volume by country 2020. Statista.

Conway, J. (2022b, June 27). Global beer production 1998-2020. Statista. https://www.statista.com/statistics/270275/worldwide-beer-production

Conway, J. (2022c, October 15). Global leading countries in beer production 2020. Statista. https://www.statista.com/statistics/270269/leading-10-countries-in-worldwide-beer-production/Source:https://www.statista.com/statistics/270269/leading-10-countries-in-worldwide-beer-production/

Dudek, M., Świechowski, K., Manczarski, P., Koziel, J. A., & Białowiec, A. (2019). The effect of biochar addition on the biogas production kinetics from the anaerobic digestion of brewers' spent grain. Energies, 12(1518), 1–22. https://doi.org/doi:10.3390/en12081518

Dyjakon, A., Sobol, Ł., Noszczyk, T., & Mitręga, J. (2022). The Impact of Torrefaction Temperature on the Physical-Chemical Properties of Residual Exotic Fruit (Avocado, Mango, Lychee) Seeds. Energies, 15(2), 612. https://doi.org/10.3390/en15020612

Fidel, R. B., Laird, D. A., Thompson, M. L., & Lawrinenko, M. (2017). Characterization and quantification of biochar alkalinity. Chemosphere, 167, 367–373. https://doi.org/10.1016/j.chemosphere.2016.09.151

Filer, J., Ding, H. H., & Chang, S. (2019). Biochemical Methane Potential (BMP) Assay Method for Anaerobic Digestion Research. Water, 11(5), 921. https://doi.org/10.3390/w11050921

Gomes, M. M., Sakamoto, I. K., Silva Rabelo, C. A. B., Silva, E. L., & Varesche, M. B. A. (2021). Statistical optimization of methane production from brewery spent grain: Interaction effects of temperature and substrate concentration. Journal of Environmental Management, 288, 112363. https://doi.org/10.1016/j.jenvman.2021.112363

Gunes, B., Stokes, J., Davis, P., Connolly, C., & Lawler, J. (2019). Pre-treatments to enhance biogas yield and quality from anaerobic digestion of whiskey distillery and brewery wastes: A review. Renewable and Sustainable Energy Reviews, 113, 109281. https://doi.org/10.1016/j.rser.2019.109281

Hejna, A., Marć, M., Kowalkowska-Zedler, D., Pladzyk, A., & Barczewski, M. (2021). Insights into the thermo-mechanical treatment of brewers' spent grain as a potential filler for polymer composites. Polymers, 13(6), 879. https://doi.org/10.3390/polym13060879

Ikram, S., Huang, L., Zhang, H., Wang, J., & Yin, M. (2017). Composition and nutrient value proposition of brewers spent grain. Journal of Food Science, 82(10), 2232–2242. https://doi.org/10.1111/1750-3841.13794

IR Spectrum Table&Chart. (2022). Merck. https://www.sigmaaldrich.com/PL/pl/technical-documents/technical-article/analytical-chemistry/photometry-and-reflectometry/ir-spectrum-table?fbclid=IwAR18GnIQ5wowNCU-oNUwwmbGNYMXzFk9rAu32e-jQkVlSz3HjJsoDQIQ5rpg

Karlović, A., Jurić, A., Ćorić, N., Habschied, K., Krstanović, V., & Mastanjević, K. (2020). By-products in the malting and brewing industries—re-usage possibilities. Fermentation, 6(3), 82. https://doi.org/10.3390/fermentation6030082

Khanal, S. Kumar. (2008). Anaerobic biotechnology for bioenergy production principles and applications (1st ed.). Wiley-Blackwell.

Li, Y., Qi, Q., & Li, J. (2011). Energy use project and conversion efficiency analysis on biogas produced in breweries. Industrial Energy Efficiency, 1489–1496. https://citeseerx.ist.psu.edu/viewdoc/download?doi=10.1.1.452.56&rep=rep1&type=pdf

Łyczko, J., Koziel, J. A., Banik, C., & Białowiec, A. (2021). The proof-of-concept: The transformation of naphthalene and its derivatives into decalin and its derivatives during thermochemical processing of sewage sludge. Energies, 14(20). https://doi.org/10.3390/en14206479

Mainardis, M., Flaibani, S., Mazzolini, F., Peressotti, A., & Goi, D. (2019). Techno-economic analysis of anaerobic digestion implementation in small Italian breweries and evaluation of biochar and granular activated carbon addition effect on methane yield. Journal of Environmental Chemical Engineering, 7(3), 103184. https://doi.org/10.1016/j.jece.2019.103184

Masebinu, S. O., Fanoro, O. T., Insam, H., Mbohwa, C., Wagner, A. O., Markt, R., & Hupfauf, S. (2021). Can the addition of biochar improve the performance of biogas digesters operated at 45°C? Environmental Engineering Research, 27(2), 200648–0. https://doi.org/10.4491/eer.2020.648

Miller, K. E., Herman, T., Philipinanto, D. A., & Davis, S. C. (2021). Anaerobic digestion of food waste, brewery waste, and agricultural residues in an off-grid continuous reactor. Sustainability, 13(12). https://doi.org/10.3390/su13126509

Morales, V. L., Pérez-Reche, F. J., Hapca, S. M., Hanley, K. L., Lehmann, J., & Zhang, W. (2015). Reverse engineering of biochar. Bioresource Technology, 183, 163–174. https://doi.org/10.1016/j.biortech.2015.02.043

Naibaho, J., Korzeniowska, M., Wojdyło, A., Figiel, A., Yang, B., Laaksonen, O., Foste, M., Vilu, R., & Viiard, E. (2021). Fiber modification of brewers' spent grain by autoclave treatment to improve its properties as a functional food ingredient. LWT, 149, 111877. https://doi.org/10.1016/j.lwt.2021.111877

Nasir, N. A., Yakub, I., Razali, N. A., & Rosid, S. J. M. (2021). Hydrothermal liquefaction of an industrial biomass waste: Brewer's Spent Grain (BSG). Nano Hybrids and Composites, 31, 65–72. https://doi.org/10.4028/www.scientific.net/nhc.31.65

Ngo, T., Khudur, L. S., Hakeem, I. G., Shah, K., Surapaneni, A., & Ball, A. S. (2022). Wood biochar enhances the valorisation of the anaerobic digestion of chicken manure. Clean Technologies, 4(2), 420–439. https://doi.org/10.3390/cleantechnol4020026

Nwokolo, N., Mukumba, P., Obileke, K., & Enebe, M. (2020). Waste to energy: A focus on the impact of substrate type in biogas production. Processes, 8(10), 1224. https://doi.org/10.3390/pr8101224

OGL. (2021). Combined heat and power-technologies A detailed guide for CHP developers-Part 2. https://www.gov.uk/government/collections/combined-heat-and-power-chp-developers-guides#chp-development-guides

Olajire, A. A. (2020). The brewing industry and environmental challenges. Journal of Cleaner Production, 256, 102817. https://doi.org/10.1016/j.jclepro.2012.03.003

Oliveira, J. v., Alves, M. M., & Costa, J. C. (2018). Biochemical methane potential of brewery by-products. Clean Technologies and Environmental Policy, 20(2), 435–440. https://doi.org/10.1007/s10098-017-1482-2

PKN ISO/TS 12902 : 2007 Solid mineral fuel - determination of total carbon, hydrogen and nitorgen - instrumental methods. (n.d.).

PN-EN 14346:2011 Standard. Waste characteristics. Calculation of dry mass on the basis of dry residue or water content. (n.d.).

PN-EN 15169:2011 Standard. Waste characteristics. Determination of organic matter content for waste, slurry and sludge. (n.d.).

Poulsen, T. G., Adelard, L., & Wells, M. (2017). Improvement in CH4 / CO2 ratio and CH4 yield as related to biomass mix composition during anaerobic co-digestion. Waste Management, 61, 179–187. https://doi.org/10.1016/j.wasman.2016.11.009

Sanna, A., Li, S., Linforth, R., Smart, K. A., & Andrésen, J. M. (2011). Bio-oil and bio-char from low temperature pyrolysis of spent grains using activated alumina. Bioresource Technology, 102(22), 10695–10703. https://doi.org/10.1016/j.biortech.2011.08.092

Sežun, M., Grilc, V., Zupančič, G. D., & Logar, R. M. (2011). Anaerobic digestion of brewery spent grain in a semi-continuous bioreactor: inhibition by phenolic degradation products. Acta Chim Slov, 58(1), 158–166. https://pubmed.ncbi.nlm.nih.gov/24061957/

Shi, R., Hong, Z., Li, J., Jiang, J., Baquy, M. A.-A., Xu, R., & Qian, W. (2017). Mechanisms for Increasing the pH Buffering Capacity of an Acidic Ultisol by Crop Residue-Derived Biochars. Journal of Agricultural and Food Chemistry, 65(37), 8111–8119. https://doi.org/10.1021/acs.jafc.7b02266

Shin, D. C., Kim, I.-T., Jung, J., Jeong, Y., Lee, Y.-E., & Ahn, K.-H. (2022). Increasing Anaerobic Digestion Efficiency Using Food-Waste-Based Biochar. Fermentation, 8(6), 282. https://doi.org/10.3390/fermentation8060282

Shin, R., & Searcy, C. (2018). Evaluating the greenhouse gas emissions in the craft beer industry: An assessment of challenges and benefits of greenhouse gas accounting. Sustainability, 10(11), 4191. https://doi.org/10.3390/su10114191

Sieradzka, M., Kirczuk, C., Kalemba-Rec, I., Mlonka-Mędrala, A., & Magdziarz, A. (2022). Pyrolysis of biomass wastes into carbon materials. Energies, 15(5), 1941. https://doi.org/10.3390/en15051941

Sobol, Ł., Dyjakon, A., Suardi, A., & Preißmann, R. (2021). Analysis of the possibility of energetic utilization of biomass obtained from grass mowing of a large-area golf course—a case study of tuscany. Energies, 14(17). https://doi.org/10.3390/en14175520

Stachowiak-Wencek, A., Bocianowski, J., Waliszewska, H., Borysiak, S., Waliszewska, B., & Zborowska, M. (2021). Statistical prediction of biogas and methane yields during anaerobic digestion based on the composition of lignocellulosic biomass. BioResources, 16(4), 7086–7100.

Świechowski, K., Hnat, M., Stępień, P., Stegenta-Dąbrowska, S., Kugler, S., Koziel, J. A., & Białowiec, A. (2020). Waste to Energy: Solid Fuel Production from Biogas Plant Digestate and Sewage Sludge by Torrefaction-Process Kinetics, Fuel Properties, and Energy Balance. Energies, 13(12), 3161. https://doi.org/10.3390/en13123161

Świechowski, K., Matyjewicz, B., Telega, P., & Białowiec, A. (2022). The influence o low-temperature food waste biochars on anaerobic digestion of food waste. Materials, 15(3). https://doi.org/10.3390/ma15030945

Syguła, E., Gałęzowska, M., & Białowiec, A. (2022). Enhanced production of biogas using biochar–sulfur composite in the methane fermentation process. Materials, 15(13), 4517. https://doi.org/10.3390/ma15134517

Torquato, L. D. M., Crnkovic, P. M., Ribeiro, C. A., & Crespi, M. S. (2017). New approach for proximate analysis by thermogravimetry using CO2 atmosphere: Validation and application to different biomasses. Journal of Thermal Analysis and Calorimetry, 128(1). https://doi.org/10.1007/s10973-016-5882-z

Wambugu, C. W., Rene, E. R., van de Vossenberg, J., Dupont, C., & van Hullebusch, E. D. (2019). Role of Biochar in Anaerobic Digestion Based Biorefinery for Food Waste. Frontiers in Energy Research, 7. https://doi.org/10.3389/fenrg.2019.00014

Wang, D., Ai, J., Shen, F., Yang, G., Zhang, Y., Deng, S., Zhang, J., Zeng, Y., & Song, C. (2017). Improving anaerobic digestion of easy-acidification substrates by promoting buffering capacity using biochar derived from vermicompost. Bioresource Technology, 227, 286–296. https://doi.org/10.1016/j.biortech.2016.12.060

Wang, S., Shi, F., Li, P., Yang, F., Pei, Z., Yu, Q., Zuo, X., & Liu, J. (2022). Effects of rice straw biochar on methanogenic bacteria and metabolic function in anaerobic digestion. Scientific Reports, 12(1), 6971. https://doi.org/10.1038/s41598-022-10682-2

Xi, X., Yan, J., Quan, G., & Cui, L. (2014). Removal of the pesticide pymetrozine from aqueous solution by biochar produced from Brewer's spent grain at different pyrolytic temperatures. BioResources, 9(4), 7696–7709. https://doi.org/10.15376/biores.9.4.7696-7709

Zborowska, M., Waliszewska, H., Waliszewska, B., Borysiak, S., Brozdowski, J., & Stachowiak-Wencek, A. (2022). Conversion of carbohydrates in lignocellulosic biomass after chemical pretreatment. Energies, 15(1). https://doi.org/10.3390/en15010254

Zhang, M., Li, J., Wang, Y., & Yang, C. (2019). Impacts of different biochar types on the anaerobic digestion of sewage sludge. RSC Advances, 9(72), 42375–42386. https://doi.org/10.1039/C9RA08700A

Zhao, W., Yang, H., He, S., Zhao, Q., & Wei, L. (2021). A review of biochar in anaerobic digestion to improve biogas production: Performances, mechanisms and economic assessments. Bioresource Technology, 341, 125797. https://doi.org/10.1016/j.biortech.2021.125797

Zhao, Z., Li, Y., Zhang, Y., & Lovley, D. R. (2020). Sparking anaerobic digestion: promoting direct interspecies electron transfer to enhance methane production. IScience, 23(12), 101794. https://doi.org/10.1016/j.isci.2020.101794

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# BEYOND WASTE-TO-ENERGY: BIOENERGY CAN DRIVE SUSTAINABLE AUSTRALIAN AGRICULTURE BY INTEGRATING CIRCULAR ECONOMY WITH NET ZERO AMBITIONS

Maja Arsic [1], Cathryn A. O'Sullivan [1,*], Anton P. Wasson [2], Diogenes L. Antille [2] and William P. Clarke [3]

*[1] CSIRO Agriculture and Food, Queensland Biosciences Precinct, St Lucia, QLD 4067, Australia*
*[2] CSIRO Agriculture and Food, Black Mountain Science and Innovation Precinct, Canberra, ACT 2601, Australia*
*[3] School of Civil Engineering, University of Queensland, St Lucia, QLD 4067, Australia*

**Article Info:**
Received:
13 February 2023
Revised:
29 April 2023
Accepted:
23 May 2023
Available online:
15 June 2023

**Keywords:**
Circular bioeconomy
Renewable energy
Waste management

**ABSTRACT**

The race to meet net zero targets by 2050, while rapidly transitioning to a circular economy (CE) within the next decade, is shaping strategic Australian sustainability policy. While the success of integrating CE concepts relies on coordinating system-wide change, policies and strategies are still evolving under the traditional silos of waste and energy management. This presents multiple barriers to critical sectors, such as agriculture, which aims to become an $AUD100 billion industry by 2030. Agri-food systems face the challenge to meet growing global food demand, expected to increase by 70% by 2050, while decreasing emissions, resource use and waste production. Agriculture plays essential push and pull roles in meeting net zero targets and in developing a truly CE. Bioenergy, a critical part of the renewable circular bioeconomy, sits at the intersection of net zero and CE by producing renewable energy and recovering bioresources from waste biomass. By integrating agricultural end-users as key stakeholders, bioenergy can shift from a waste-to-energy process to a multi-resource generating process. These policy areas could be integrated via a similar approach to the Australian National Agricultural Innovation Policy Statement, with the goal of supporting agricultural production, while reducing emissions and maximising renewable resource use efficiency.

## 1. INTRODUCTION

### 1.1 The challenge of sustaining food production while reducing emissions and resource use

The calls to reach "net zero" greenhouse gas (GHG) emissions by 2050 has triggered one of the largest global transitions since the industrial revolution (IPCC, 2018). This transition is occurring in concert with moving from linear systems of production, consumption, and waste management towards a "circular economy" for material and biological resources (the bio-based or bioeconomy) (Arsic et al., 2022; Carus and Dammer, 2018; Stahel, 2016). However, with an estimated 8.5 billion people by 2030, and food demand rising by 70% by 2050, it will be challenging to reduce emissions and resource use while meeting the 2030 Sustainable Development Goals (SDG), notably SDG 2 "End hunger, achieve food security and improve nutrition and promote sustainable agriculture" (UNICEF, 2021). It is therefore critical to apply all three circular economy principles ("design out waste and pollution, keep products and materials in use, and regenerate natural systems") to ensure that valuable resources within organic biomass "wastes" are utilised to their full potential (Ellen MacArthur Foundation, 2015). From an Australian perspective, agriculture has an important role in international and domestic food security, supplying over 90 per cent of domestic food while exporting over 70 per cent of produce (ABARES, 2022). The sector aims to reach $AUD100 billion production value by 2030 to meet global food demands (Delivering Ag2030) (DAFF, 2022). However, growth must be aligned with strategic shifts for energy and resources.

### 1.2 Beyond waste-to-energy?

The bioenergy sector has valorised organic wastes for several decades through "waste-to-energy" technologies, which can be defined as "…any waste treatment process that creates energy in the form of electricity, heat or transport fuels (e.g., diesel) from a waste source" (World Energy Council, 2013). In addition, some waste-to-energy technologies such as anaerobic digestion (AD) and pyro-

---

* Corresponding author:
Cathryn O'Sullivan
email: cathryn.osullivan@csiro.au

Detritus / Volume 23 - 2023 / pages 28-34
https://doi.org/10.31025/2611-4135/2023.17278

genic carbon capture and storage (PyCCS) can generate by-products that have reported beneficial properties as soil amendments and bio-based fertilisers (e.g., digestate, biochar) while reducing GHG emissions (carbon and methane) (Pivato et al., 2023). However, the development of these by-products into safe and effective agricultural inputs has been limited by a range of barriers, including technoeconomic (cost of transport, specialised machinery for land spreading, variability in volumes of production, elemental composition, presence of contaminants, and nutrient availability) (e.g., Antille et al., 2013), regulatory (variability in contaminant concentrations, emerging contaminants, variability in landfill levies, regional bans on thermal treatment, lack of waste reuse regulations), and policy (lack of national and state bioeconomy policies, policy "silos" between waste and energy). These barriers must be addressed to facilitate the development of viable novel circular value-chains and biomass value webs or networks, which are "…complex systems of interlinked value chains in which biomass products and by-products are produced, processed, traded, and consumed" (Callo-Concha et al., 2020). Waste-to-energy technologies should be utilised for their potential beyond waste valorisation for energy, as they sit at the energy-waste-food nexus with the potential to reduce emissions and return essential plant nutrients to agricultural soils, while keeping valuable resources out of landfill. This approach will also address some of the barriers currently limiting the expansion of the bioenergy sector in Australia, as it faces issues between energy and waste management regulatory and policy silos (Arsic et al., 2022).

This discussion paper aims to highlight the joint opportunities to sustainably utilise organic waste resources in Australia by exploring the synergies between bioenergy production and providing safe and effective inputs for agricultural systems. Key barriers preventing waste valorisation are outlined and high-level Australian and European Union (EU) governance structures across climate, waste management and agricultural policies compared. Based on this analysis, three key actions that could be readily implemented to address these barriers are suggested.

## 2. VALORISATION OF ORGANIC WASTES AND BIOMASS: AN AUSTRALIAN PERSPECTIVE

### 2.1 Bioenergy in Australia

This discussion piece is focused on the use of organic wastes and agricultural residues for bioenergy and circular fertiliser production, rather than purpose-grown bioenergy crops. The total energy supply in Australia is dominated by fossil fuels (5390 PJ), with oil (33 per cent), coal (32 per cent) and natural gas (27 per cent) contributing over 90 per cent in 2019 (CEC 2020). Bioenergy supplied 5 per cent of total clean energy production (394 PJ) and 1.4 per cent of total Australian electricity generation in 2020 (CEC, 2020). Technologies such as anaerobic digestion (AD) and thermal treatment (e.g., pyrolysis, gasification, incineration) have been used to process municipal, industrial, and agricultural wastes. In 2021, the Australian Renewable Energy Agency's (ARENA) Bioenergy Roadmap stated bioenergy could "reduce emissions by about 9 per cent, [and] divert an extra 6 per cent of waste from landfill by 2030" (ARENA, 2021). The report also notes, "Organic wastes and residues are the largest resource opportunity, for developing the industry in the short term, representing 37 per cent of Australia's current [bioenergy] potential." (ARENA, 2021).

### 2.2 Barriers to bioenergy sector development

There are two key barriers to sector development. Firstly, operational silos between energy and waste at policy, regulatory, and technological adoption levels must be addressed. The current focus in Australia is on identifying the mix of technologies that can most rapidly transition away from fossil fuels towards renewable energy sources, such as hydrogen, solar and wind power (CEC, 2020). Bioenergy systems have been primarily installed for waste management purposes to either replace or reduce the flow of organic materials to landfill, reduce odors, or to mitigate GHG emissions at landfill. While this diversion of materials is important and represents both cost savings for industry and reducing the volume of materials sent to landfill, bioenergy risks being viewed as a waste management solution, leading to a lack of recognition and investment from within the energy sector. Secondly, the full potential of bioenergy must be utilised beyond waste-to-energy towards "waste-energy-bioresources". There is recognition that the residues from bioenergy systems can have value as agricultural inputs, but there is a need for better engagement with fertiliser companies and end-users to create viable products (GHD, 2022). Similar technical and economic barriers faced by the development of organic waste fertilisers (e.g., sustainable feedstock sourcing, reliability, transport logistics, developing supply chains) are currently limiting the investment in and adoption of bioenergy as a renewable energy technology. Value chains need to be assessed from end-to-end to ensure that there is suitable feedstock, viable energy balance, value added fertiliser products and market demand. A lack of appropriate coordination across industry sectors risks fueling competition for feedstocks and organic waste resources, which may not consider their current value on-farm in terms of natural capital or productivity or may create new barriers that prevent the full valorisation of waste streams at their highest order use.

### 2.3 Beyond barriers: Valorising bioresources from organic wastes by integrating the agricultural sector

These barriers may be addressed by acknowledging the essential roles that the agricultural sector plays in the development of a viable bioenergy sector in Australia. Agriculture contributes both "pull" (energy and fuel demand, need for GHG offsets and waste management solutions) and "push" (historical acceptance of industrial organic waste by-products as a landfill diversion strategy, an increasing demand for organic or bio-based fertilisers) drivers for bioenergy technology adoption. Integrating the waste management service provision by bioenergy systems with their capacity to produce a range of bioresources is important, as the cost savings from improved waste management can be reinvested to explore circular resource recovery from other system by-products. New circular value chains and webs can be created by applying a biorefinery model to fully capture bioresources from organic wastes via "the sustainable processing of biomass into a spectrum of marketable products and energy" (Cherubini et al., 2009). For example, the application of organic carbon-rich products such as digestate and biochar to agricultural soils has the potential to increase soil carbon sequestration (Breunig et al., 2019). The possibility to generate income through carbon trading will help to offset the cost of utilising the organic residues from bioenergy systems at scale and create viable business models, while valuing the full range of products generated by bioenergy will require the development of new circular value chains (Verra, 2022). Developing an environmental monitoring and auditing system to measure, trace and ensure compliance of sustainability, contaminant limits and product agronomic effectiveness will be critical to producing safe and effective bioresources from waste-to-energy technologies.

## 3. OPPORTUNITIES AND BARRIERS FOR UTILISING WASTE BIOMASS IN AGRICULTURE

### 3.1 Opportunities for bioresources within agricultural and food systems

Applying organic wastes as soil amendments has long been practiced on-farm for improving plant nutrition, building soil carbon and fertility, and therefore enhancing soil physico-chemical properties and soil function (e.g., Quilty and Cattle, 2011). Farmers have accepted organic wastes for free with delivery costs (and sometimes field application costs) met by the waste producer, as this has historically been a cost reduction strategy to divert waste from landfill or incineration. Soil amendments include "by-products" from processes such as anaerobic digestion (digestate) and pyrolysis/gasification (biochar), which have been applied for sewage sludge treatment as well as bioenergy generation (Abbott et al., 2018; Nkoa, 2014). While current organic fertiliser production is small (16 Mt produced in 2020 compared to approximately 200 Mt of mineral fertilisers), market analyses project demand for nutrients will almost double by 2030 (Richardson, 2022; Technavio, 2020). Interest in bio- or organic-based fertiliser materials has been growing rapidly, partly in response to raising energy costs and partly due to increasing demand for carbon-rich products to build soil carbon and fertility (Richardson, 2022). Farmers are beginning to engage with financial incentives such as carbon farming methods, which are a subset of carbon trading initiatives and include increasing soil carbon sequestration by applying carbon-rich material to soils (CER, 2023). Similarly, biodiversity credits aim to stimulate private investment in agricultural stewardship, supported by the recent "Agriculture Biodiversity Stewardship Package" (DAFF, 2022). The Emissions Reduction Fund (ERF), Australia's national mechanism to stimulate investment in activities that store carbon and mitigate climate change, includes biochar as one of the approved inputs for the "Estimating soil organic carbon sequestration using measurement and models method" (CER, 2023).

### 3.2 Technoeconomic, social, regulatory and policy barriers

There are several types of barriers to uptake for farmers across geographical scales. Technical barriers include the availability and consistency of waste feedstocks, the location of feedstocks compared to the location of end-users, transportation costs and logistics, and the type of technology used to process organic wastes and the resulting "by-products". The variability in nutrient composition and nutrient availability compared with mineral fertilisers, makes it difficult to predict nutrient supply from organic materials (Quilty and Cattle, 2011). This has consequences for accurately estimating field application rates and optimising the timing of soil application to minimise environmental losses, and maximise agronomic efficacy and economic return (Antille et al., 2017). Similar barriers exist in terms of determining the presence, concentrations, and fate of contaminants such as trace metals, persistent organic pollutants, and microplastics. Further, the feasibility of spreading bulky organic products on-farm can pose additional costs compared to manufactured fertilisers. This includes increased energy use through fuel consumption, which was estimated to be up to three times higher for spreading organic materials such as cattle paunch, or through engaging external contractors to spread materials using specialised field equipment (Antille et al., 2018). Research is needed to substantiate the claims made about various products for farm productivity or carbon and natural capital sustainability metrics. For example, although many commercial products and services note the potential for biochar applications for soil carbon sequestration and improving soil fertility and physical properties (e.g., water holding capacity) or biochemically (e.g., improving nutrient cycling), positive results are highly dependent on a range of complex factors such as feedstock, pyrolysis conditions, biochar formulation, application rate, soil and crop type, the interaction with biological and environmental stress, and monitoring time (performance over years compared to decades and beyond) (Joseph et al., 2021).

Soft barriers such as social, policy and regulatory issues are also challenging. Social barriers include attitudes to waste products, willingness to pay versus accepting free residues, odor, concerns over soil contamination and risk of transfer of such contaminants to the food chain. Skepticism around product benefits, capacity to accept risks associated with changing management practices on-farm, and a lack of co-design practices between engineers, waste treatment processors and agricultural end-users to work towards generating enhanced-quality products for farmers have also been reported (e.g., Marchuk et al., 2023; McCabe et al., 2020). Policy and regulatory barriers include waste management regulations and the lack of clarity in emerging "end of waste codes" to allow the application of waste products in soils. There has also been stakeholder disagreement on acceptable contaminant levels for blended products such as composts (Australian Standard AS 4454-2012, Composts, soil conditioners and mulches), and reports of some industrial waste producers using blending processes to dilute contaminated waste streams into "clean" streams to avoid disposing of regulated wastes. While recent efforts have been made at the local, state, and federal government levels to facilitate the shift to CE models, developing CE frameworks and decision-making tools based on inorganic materials such as metal, glass and plastics risks missing opportunities for organic resources (Circular Australia, 2022). Additional barriers also include the administrative cost burden on farmers to access emerging carbon or natural capital markets, which otherwise is a significant opportunity as farmers manage over 55% of Australia's total land area (ABARES, 2022).

## 4. TOWARDS A NATIONAL CIRCULAR BIOECONOMY FOR ENERGY AND AGRI-FOOD SYSTEMS

Developing a holistic approach towards a circular bioeconomy could address the range of barriers identified earlier by moving towards full valorisation of biological resources, from virgin feedstocks to organic wastes. Effective implementation of circular strategies will likely deliver positive economic, social, cultural, and environmental outcomes (Burggraaf et al., 2020; 2022). Bioeconomy can be defined as "the production, utilisation, conservation, and regeneration of biological resources, including related knowledge, science, technology, and innovation, to provide sustainable solutions (information, products, processes and services) within and across all economic sectors and enable a transformation to a sustainable economy" (IACGB, 2020). Circular bioeconomy integrates CE principles into this definition, by "aim[ing] to provide sustainable wellbeing through the provision of ecosystem services and the sustainable management of biological resources (plants, animals, micro-organisms, and derived biomass, including organic waste). These [resources] are transformed in a circular manner into food, feed, energy, and biomaterials – within the ecological boundaries of the ecosystems that it relies on." (Palahi et al., 2020).

To identify potential actions that could be taken, relevant climate, waste management and agricultural policies were compared between Australia and the EU. The European Commission has pioneered both the strategic development of the bioeconomy (2012 Strategy "Innovating for Sustainable Growth: A Bioeconomy for Europe, Action Plan in 2018) and the CE (2015 Action Plan for the Circular Economy), which are now both key elements in the 2019 EU Green Deal. From an Australian perspective, policies and legislative drivers include the recent Climate Change Act 2022, which confirms Australia's commitment to reduce GHG emissions by 45% by 2030 and to reach net zero by 2050. The National Waste Policy 2018 (NWP) and National Waste Policy Action Plan 2019 (NWPAP) aim to "...embod[y] a circular economy... [to] maintain the value of resources for as long as possible..." (DEE, 2018; 2019). The NWP and NWPAP include strategies for municipal organic waste but not agricultural wastes, likely due to their large volumes and dispersed nature. A National Waste Roadmap for the agricultural, forestry and fisheries sectors in Australia has been drafted and is currently under review (AgriFutures, 2022). However, proposed strategies for agricultural waste reuse must consider feasibility (e.g., large farm sizes and distances from processing centers or customers) and must weigh the competition between new proposed processes and products from collecting wastes (e.g., stubble) with their current provision of natural capital (e.g., providing habitat for biodiversity) and productivity values (e.g., bedding, soil cover, returning carbon and nutrients to soils) (Brady et al., 2015). While CE policies have recently been adopted federally and by most states, there is currently no national bioeconomy policy. While three states have specific bioeconomy policies, other states have developed policy documents for aspects of the bioeconomy such as waste-to-energy, biotechnology, or "organics" more broadly (Arsic et al., 2022).

By comparing the relevant high-level governance structures between Australian and the EU, there are three key gaps from an Australian perspective (Iriarte et al., 2021) (Figure 1). Firstly, while Australia is participating in international bioeconomy forums (as an observer), the lack of a national policy means there is a gap in addressing complex institutional co-ordination and actor cooperation across the waste-energy-bioresources nexus. Secondly, the EU system of governance includes two critical instruments that are missing from the Australian context: the system underpinning the creation of new circular value chains, the Eco-Management and Audit (EMA) Scheme (EC, 2022), and the Circular Economy Package Fertiliser Regulation (Regulation (EU) No 2019/1009) for production and tracing of "circular" fertilisers (EU, 2019). Finally, cross-sectoral key research, development and extension priorities need to be identified that would allow for the expansion of these emerging industries and products supported by sustainability metrics. These actions could be incorporated into a framework such as the National Agricultural Innovation Policy Statement, which outlines a vision, the priorities needed to achieve the vision, current and future reforms, the key participants in the system and their strategic roles and relationships, and outlines how to monitor and evaluate success (DAWE, 2021). There is a resurgence of interest in industrial policy, beyond innovation policy, particularly as a means of addressing environmental challenges; an approach that is appropriate to issues that involve inter-industry linkages and clusters of technological innovations (Aiginger and Rodrik, 2020). In addition, by coordinating this policy through the Federal Department of Agriculture, Fisheries and Forestry, the importance of returning nutrient or carbon-rich inputs to support growing productivity in agricultural systems would support the sector's ambitious $AUD100 billion production value goal by 2030 (Ag2030), as well as supporting natural capital in agroecosystems. The development of these governance tools would leverage Australian agriculture's emerging potential for supporting global food security, as well as investing in bioenergy technologies and international sustainability markets (Figure 2).

## 5. CONCLUSIONS

The sustainable utilisation of biomass derived from organic waste in sectors such as bioenergy and agriculture

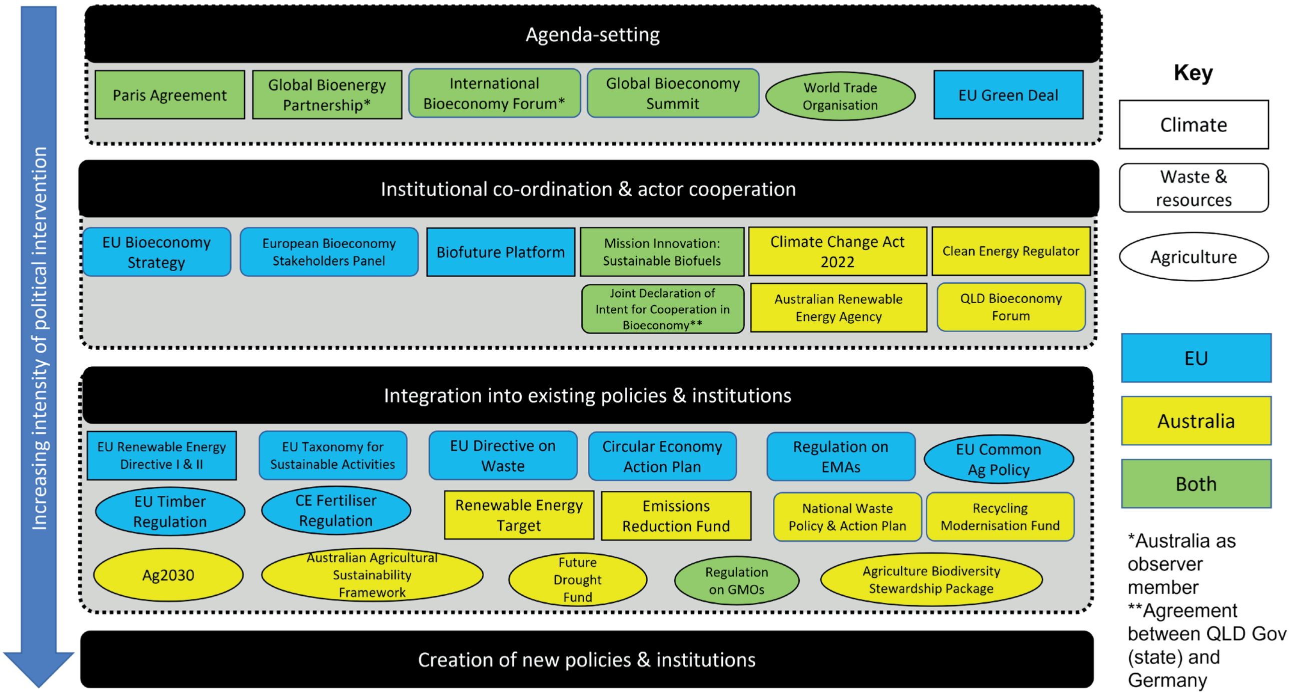


**FIGURE 1:** Governing a circular bioeconomy: A comparison between the European Union and Australia (modified from Iriarte et al. 2021).

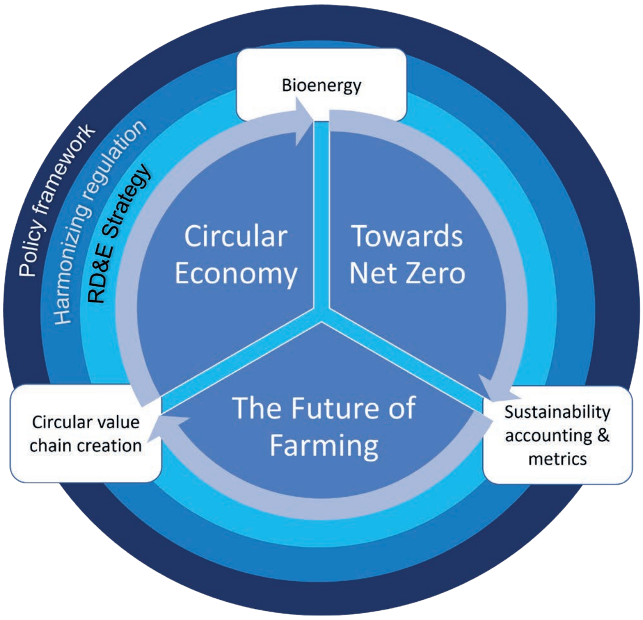


**FIGURE 2:** A conceptual model outlining the potential roles of bioenergy, emerging circular value chains, and the development of transparent and traceable sustainability accounting platforms and metrics to coordinate actions towards CE models, net zero pathways and sustainable agricultural systems.

rely on the successful integration of circular bioeconomy concepts across multiple systems and governance frameworks. The techno-economic, social and policy barriers limiting the use of these resources could be addressed by recognising the key role of agricultural stakeholders in emerging bioenergy technologies and supply chains, where bioenergy has the potential to generate multiple valuable bioresources to support sustainable agricultural production. Future actions to realise this potential include the development of a national circular bioeconomy framework and governance structure, integrating systems such as auditing mechanisms within new sustainability tracing and accounting platforms, and identifying critical research and technology, development, and extension priorities. Through this approach, bioresources in organic wastes could be used to sustainably establish energy and agri-food production, while reducing emissions and improving resource use efficiency.

## FUNDING AND ACKNOWLEDGEMENTS

This work was funded by CSIRO through the Agriculture & Food Winanga-y Early Research Career (CERC) Fellowship scheme. The authors would like to acknowledge the CSIRO circular economy community-of-practice, particularly Pablo Juliano, Colleen MacMillan and Rebecca Haling, for contributing to the thinking that led to this paper. We would also like to thank the CSIRO Agriculture and Food leadership team for supporting the development of the circular agriculture research team.

## REFERENCES

ABARES (Australian Bureau of Agricultural and Resource Economics and Sciences) (2022). Snapshot of Australian Agriculture 2022 [Online]. Available: https://www.agriculture.gov.au/abares/products/insights/snapshot-of-australian-agriculture-2022#agricultural-production-is-growing [Accessed 26 September 2022].

Abbott, L. K., Macdonald, L. M., Wong, M. T. F., Webb, M. J., Jenkins, S. N., & Farrell, M. (2018). Potential roles of biological amendments for profitable grain production–A review. Agriculture, Ecosystems & Environment, 256, 34-50. https://doi.org/10.1016/j.agee.2017.12.021

Aiginger, K., & Rodrik, D. (2020). Rebirth of industrial policy and an agenda for the twenty-first century. Journal of industry, competition and trade, 20, 189-207. https://doi.org/10.1007/s10842-019-00322-3

Antille, D.L., Godwin, R.J., Sakrabani, R., Seneweera, S., Tyrrel, S.F., Johnston, A.E. (2017). Field-scale evaluation of biosolids-derived organomineral fertilizers applied to winter wheat in England. Agronomy Journal, 109(2): 654-674. https://doi.org/10.2134/agronj2016.09.0495.

Antille, D. L., McCabe, B. K., Van Der Spek, W., & Baillie, C. P. (2018). An investigation into the fertilizer potential of slaughterhouse cattle paunch. Transactions of the ASABE, 61(1), 87-101. https://doi.org/10.13031/trans.12342.

Arsic, M., O'Sullivan, C.A., Wasson, A.P., Juliano, P., MacMillan, C.P., Antille, D.L., Haling, R.E., & Clarke, W.P. (2022). Australia needs a national policy approach to successfully implement circular bioeconomy in agriculture and food systems. Farm Policy Journal, 19, 46-60. ISSN: 1449-2210.

Brady, M. V., Hedlund, K., Cong, R. G., Hemerik, L., Hotes, S., Machado, S., Mattsson, L., Schulz, E. & Thomsen, I. K. (2015). Valuing supporting soil ecosystem services in agriculture: a natural capital approach. Agronomy Journal, 107(5), 1809-1821. https://doi.org/10.2134/agronj14.0597

Breunig, H. M., Amirebrahimi, J., Smith, S., & Scown, C. D. (2019). Role of digestate and biochar in carbon-negative bioenergy. Environmental science & technology, 53(22), 12989-12998. https://doi.org/10.1021/acs.est.9b03763

Burggraaf, V.T., Lucci, G.D., Ledgard, S., Antille, D.L., Snow, V.O., de Klein, C.A.M. (2020). Application of circular economy principles to New Zealand pastoral farming systems. Journal of New Zealand Grasslands, 82: 53-59. https://doi.org/10.33584/jnzg.2020.82.426.

Burggraaf, V.T., Mazzetto, A.M., Romera, A.J., Mercer, G.J.K., Ledgard, S.F. (2022). Application of circular economy principles to pastoral farming: Development of an assessment framework. Animal Production Science, 62(8): 721-725. https://doi.org/10.1071/AN21167.

Callo-Concha, D., Jaenicke, H., Schmitt, C. B., & Denich, M. (2020). Food and non-food biomass production, processing and use in sub-Saharan Africa: Towards a regional bioeconomy. Sustainability, 12(5). https://doi.org/10.3390/su12052013

Carus, M., & Dammer, L. (2018). The circular bioeconomy—concepts, opportunities, and limitations. Industrial biotechnology, 14(2), 83-91. https://doi.org/10.1089/ind.2018.29121.mca

CEC (Clean Energy Council) (2020). Bioenergy [Online]. Available: https://www.cleanenergycouncil.org.au/resources/technologies/bioenergy#:~:text=and%20solar%20farms.-,Bioenergy%20generation,of%20total%20clean%20energy%20generation [Accessed 28 September 2022].

CER (Clean Energy Regulator) (2023). Agricultural methods (Emissions Reduction Fund [Online]. Available: https://www.cleanenergyregulator.gov.au/ERF/Choosing-a-project-type/Opportunities-for-the-land-sector/Agricultural-methods [Accessed 26 September 2022].

Cherubini, F., Jungmeier, G., Wellisch, M., Willke, T., Skiadas, I., Van Ree, R., & de Jong, E. (2009). Toward a common classification approach for biorefinery systems. Biofuels, Bioproducts and Biorefining, 3(5), 534-546.

Circular Australia (2022). Circular Economy Metrics – A Review [Online]. Available: https://www.uts.edu.au/isf/news/circular-economy-metrics-australia [Accessed 13 February 2023].

DAFF (Department of Agriculture, Fisheries and Forestry) (2022). Agriculture Biodiversity Stewardship Package [Online]. Available: https://www.agriculture.gov.au/agriculture-land/farm-food-drought/natural-resources/landcare/sustaining-future-australian-farming [Accessed 28 September 2022].

DAFF (Department of Agriculture, Fisheries and Forestry) (2022). Delivering Ag2030 [Online]. Available: https://www.agriculture.gov.au/agriculture-land/farm-food-drought/ag2030 [Accessed 26 September 2022].

DAWE (Department of Agriculture, Water and the Environment) (2021). National Agricultural Innovation Policy Statement [Online]. Available: https://www.agriculture.gov.au/sites/default/files/documents/dawe-innovation-policy-statement.pdf [Accessed 6 February 2023].

DCCEEW (Department of Climate Change, Energy, the Environment and Water) (2022). Recovering organic waste [Online]. Available: https://www.dcceew.gov.au/environment/protection/waste/food-waste/recovering-organic-waste [Accessed 27 September 2022].

DEE (Department of the Environment and Energy) (2018). National Waste Policy: Less Waste More Resources [Online]. Available: https://www.dcceew.gov.au/sites/default/files/documents/national-waste-policy-2018.pdf [Accessed 15 November 2022].

DEE (Department of the Environment and Energy) (2019). National Waste Policy Action Plan [Online]. Available: https://www.dcceew.gov.au/sites/default/files/documents/national-waste-policy-action-plan-2019.pdf [Accessed 15 November 2022].

Ellen MacArthur Foundation. (2015). Delivering the circular economy: A toolkit for policymakers. Ellen MacArthur Foundation. [Accessed 20 April 2022].

Enea and Deloitte for Australian Renewable Energy Agency (ARENA) (2021). Australia's Bioenergy Roadmap Report [Online]. Available: https://arena.gov.au/knowledge-bank/australias-bioenergy-roadmap-report/ [Accessed 15 November 2022].

European Commission (2022). Eco-Management and Audit Scheme [Online]. Available: https://ec.europa.eu/environment/emas/index_en.htm [Accessed 28 September 2022].

GHD for Bioenergy Australia (2022). Turning Circle – How bioenergy can supercharge Australia's circular economy [Online]. Available: https://www.ieabioenergy.com/wp-content/uploads/2022/06/Turning-Circle-report.pdf [Accessed 15 November 2022].

IACGB (International Advisory Council on Global Bioeconomy) 2020. Global Bioeconomy Summit. "Expanding the Sustainable Bioeconomy–Vision and Way Forward". Communiqué of the Global Bioeconomy Summit 2020 [Online]. Available: https://gbs2020.net/wp-content/uploads/2020/11/GBS2020_IACGB-Communique.pdf [Accessed 20 April 2023].

IPCC (Intergovernmental Panel on Climate Change) (2018). Summary for Policymakers. In: Global Warming of 1.5°C. An IPCC Special Report on the impacts of global warming of 1.5°C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the global response to the threat of climate change, sustainable development, and efforts to eradicate poverty [Masson-Delmotte, V., P. Zhai, H.-O. Pörtner, D. Roberts, J. Skea, P.R. Shukla, A. Pirani, W. Moufouma-Okia, C. Péan, R. Pidcock, S. Connors, J.B.R. Matthews, Y. Chen, X. Zhou, M.I. Gomis, E. Lonnoy, T. Maycock, M. Tignor, and T. Waterfield (eds.)]. Cambridge University Press, Cambridge, UK and New York, NY, USA, pp. 3-24. https://doi.org/10.1017/9781009157940.001.

Iriarte, L., Fritsche, U., & van Dam, J. (2021). Sustainability governance of bioenergy and the broader bioeconomy. Technical Paper prepared for IEA Bioenergy Task, 45.

Joseph, S., Cowie, A. L., Van Zwieten, L., Bolan, N., Budai, A., Buss, W., Luz Cayuela, M., Graber, E.R., Ippolito, J.A., Kuzyakov, Y., Luo, Y., Sik Ok, Y., Palansoorya, K.N., Shepherd, J., Stephens, S., Weng, Z.H. & Lehmann, J. (2021). How biochar works, and when it doesn't: A review of mechanisms controlling soil and plant responses to biochar. GCB Bioenergy, 13(11), 1731-1764.

Marchuk, S., Tait, S., Harris, P., Antille, D.L., McCabe, B.K. (2023). Biosolids-derived fertilisers: A review of challenges and opportunities. Science of the Total Environment, 875, 162555, https://doi.org/10.1016/j.scitotenv.2023.162555

McCabe, B. K., Harris, P., Antille, D. L., Schmidt, T., Lee, S., Hill, A., & Baillie, C. (2020). Toward profitable and sustainable bioresource management in the Australian red meat processing industry: A critical review and illustrative case study. Critical Reviews in Environmental Science and Technology, 50(22), 2415-2439. https://doi.org/10.1080/10643389.2020.1712310. https://doi.org/10.1080/10643389.2020.1712310

Nkoa, R. (2014). Agricultural benefits and environmental risks of soil fertilization with anaerobic digestates: a review. Agronomy for Sustainable Development, 34, 473-492. https://doi.org/10.1007/s13593-013-0196-z

Palahí, M., Pantsar, M., Costanza, R., Kubiszewski, I., Potočnik, J., Stuchtey, M., Nasi, R., Lovins, H., Giovannini, E., Fioramonti, L., Dixson-Declève, S., McGlade, J., Pickett, K., Wilkinson, R., Holmgren, J., Trebeck, K., Wallis, S., Ramage, M., Berndes, G., Akinnifesi, F.K., Ragnarsdóttir, K.V., Muys, B., Safonov, G., Nobre, A.D., Nobre, C., Ibañez, D., Wijkman, A., Snape, J., Bas, L. (2020). Investing in Nature as the true engine of our economy: A 10-point Action Plan for a Circular Bioeconomy of Wellbeing. https://doi.org/10.36333/k2a02

Pivato, A., Lavagnolo, M.C., Mensi, C., Beggio, G., Ricci, S.B., Peng, H., Arsic, M., O'Sullivan, C.A., Antille, D.L., Wasson, A.P., Haling, R.E., Lombardelli, G., Di Bona, D., Viganò, F., Consonni, S., Schievano, A. (2023). Pyrolysis of organic industrial sewage sludges: opportunities and challenges. Detritus, 22(4-7). https://doi.org/10.31025/2611-4135/2023.17249.

Quilty, J. R., & Cattle, S. R. (2011). Use and understanding of organic amendments in Australian agriculture: a review. Soil Research, 49(1), 1-26. https://doi.org/10.1071/SR10059

Richardson, A. (2022). Fertiliser manufacturing in Australia [Online]. IbisWorld. Available: https://my.ibisworld.com/au/en/industry/c1831/products-and-markets [Accessed 28 September 2022].

Stahel, W. R. (2016). The circular economy. Nature, 531(7595), 435-438.

Technavio (2020). Global Organic Fertilizers Market: 2021-2025. Chicago, IL, USA.

UNICEF (United Nations International Children's Emergency Fund) (2021). The state of food security and nutrition in the world 2021 [Online]. Available: https://www.fao.org/3/cb4474en/online/cb4474en.html [Accessed 15 November 2022].

Verra, 2022. Verra Publishes VCS Biochar Methodology [Online]. Available: https://verra.org/verra-publishes-vcs-biochar-methodology/ [Accessed on 26 September 2022].

Wilkinson, K., Price, J., Biala, J., and McDonald, D. (2021). Review of Regulations and Standards for Recycled Organics in Australia – Final Report for the Department of Agriculture, Water and the Environment. Frontier Ag & Environment, Commonwealth of Australia.

World Energy Council (2013). World Energy Resources: Waste to Energy. London, UK.

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# VALORISATION OF EYEWEAR BIOPLASTICS THROUGH HTC AND ANAEROBIC DIGESTION: PRELIMINARY RESULTS

Filippo Marchelli [1], Roberta Ferrentino [1], Giulia Ischia [1], Marco Calvi [2], Gianni Andreottola [1] and Luca Fiori [1,*]

*[1] Department of Civil, Environmental and Mechanical Engineering (DICAM), University of Trento, Via Mesiano 77, 38123 Trento (TN), Italy*
*[2] Certottica S.c.r.l., Italian Institute of Certification of Optical Products, Villanova Industrial Area, 32013 Longarone, Italy*

**Article Info:**
Received:
14 March 2023
Revised:
30 May 2023
Accepted:
15 June 2023
Available online:
30 June 2023

**Keywords:**
Bio-based plastics
Hydrothermal carbonization
Anaerobic digestion
Biomethane
Cellulose acetate

**ABSTRACT**

Bioplastics are increasingly replacing traditional plastics in many sectors, but the legislative and operative frameworks for their disposal remain unclear: they should be collected and treated together with the organic fraction of municipal solid waste (OFMSW), but often do not biodegrade satisfactorily in the plants that treat OFMSW. This work focuses on a type of cellulose diacetate employed in the eyewear industry to analyse hydrothermal carbonization (HTC) as a pre-treatment before anaerobic digestion (AD). The results show that HTC can hydrolyse this bioplastic even at moderate temperatures, reaching an almost total dissolution in the liquid phase at 210°C and, at higher temperatures, producing hydrochar. When the HTC slurry obtained at 210°C is fed to mesophilic or thermophilic AD, both the amount and the production rate of biogas are enhanced compared to the raw bioplastic. In particular, in thermophilic conditions, the amount of produced biogas undergoes at least a threefold increase compared to the untreated cellulose diacetate. Thus, this work confirms that a prior HTC step may be a suitable approach to enhance the disposal and energy recovery of bioplastics through AD.

## 1. INTRODUCTION

Bio-based plastics (or simply "bioplastics") is an umbrella term that identifies materials that are similar to traditional plastics in their mechanical properties, but are either biodegradable or obtained from biomass (or, in most cases, both) (Karan et al., 2019; Nandakumar et al., 2021; RameshKumar et al., 2020; Thakur et al., 2018). The depletion of fossil sources and the environmental impact of traditional plastics have fostered the development of bioplastics, which are slowly but steadily growing in many applications. Nonetheless, for bioplastics to become completely established, more research is needed in order to clarify some still controversial aspects.

Most commercial bioplastics are labelled as biodegradable or compostable. "Biodegradable" merely describes a material that can be broken down due to the action of microorganisms; conversely, "compostable" specifies that a biodegradable material can disintegrate into non-toxic substances in composting conditions and within a specific time frame. In principle, compostable bioplastics should be collected with the organic fraction of municipal solid waste (OFMSW) and disposed of through the same methodologies. However, the tests to assess whether a material can be defined as compostable (mainly based on UNI EN 13432 and UNI EN 14995) employ harsher conditions and longer residence times than those of most plants that treat the OFMSW – often through a sequence of anaerobic digestion and composting. As a result, in real plants bioplastics often do not biodegrade satisfactorily (Folino et al., 2023), slowing down or clogging the whole process or reducing the quality of the produced compost (Gadaleta et al., 2023). Several news outlets have reported on cases from Italian OFMSW treatment plants in which bioplastics are discarded at the entrance of plants and sent to landfilling (Athanassiou, 2021; il Dolomiti, 2019; Il Tirreno Empoli, 2019), which is a deprecated and inefficient way to dispose of organic waste. The scientific community is also increasingly aware of the problem. Battista et al. (2021) experimentally proved that most bioplastics have a low biogas production and long biodegradation time, which are not enhanced by chemical pretreatments via acids or bases at room temperature. Cazaudehore et al. (2023) showed that polyhydroxybutyrate (PHB) can be biodegraded together with biowaste, while for polylactic acid (PLA) a previous thermo-alkaline pre-treatment was necessary. The recent review by Vardar et al. (2022) further emphasises that the biodegradability of bioplastics in anaerobic digestion sys-

---

* Corresponding author:
Luca Fiori
email: luca.fiori@unitn.it

Detritus / Volume 23 - 2023 / pages 35-42
https://doi.org/10.31025/2611-4135/2023.18275

tems is often unsatisfactory, and that pre- and post-treatment methodologies must be further investigated to make these materials treatable by OFMSW plants.

The hydrothermal carbonization (HTC) process may represent a solution to this problem. HTC is an induced coalification process, performed in a temperature range of 180–250°C and in liquid water. It is able to treat a wide variety of carbonaceous feedstocks, including MSW, breaking their heavy constituent molecules into simpler ones. The products are the hydrochar (solid fraction), the HTC liquor (liquid fraction) and gases (mainly $CO_2$). When the hydrochar and/or the HTC liquor are fed to anaerobic digestion or composting facilities, they enhance the performance of the plants, increasing the product yield and quality (Ischia & Fiori, 2021), as assessed by a few recent studies (Bona et al., 2022; Ferrentino et al., 2020; González et al., 2021; Scrinzi et al., 2022). HTC has been applied to various types of biomass and waste, and hydrothermal processes were also proved suitable to decompose traditional plastics, despite sometimes requiring harsh operative conditions or the use of a catalyst (Mumtaz et al., 2023). However, researchers have neither reported whether the HTC of bioplastics is feasible nor if it can facilitate anaerobic digestion. This study aims at investigating this research gap, assessing whether a prior HTC step can enhance the disposal of residual eyewear bioplastics by promoting their conversion into valuable products, such as biogas and biomethane.

This work is part of the Occhio al Bio! project (literally “watch out for the bio!”), funded by Fondazione Cariverona and involving the University of Trento and Certottica. The project is aimed at tackling the uncertainty in the labelling and policy framework, and the correct and most fruitful approach to dispose of bioplastics, focusing on those employed in eyewear production (frames, lenses, etc.). Eyewear is a very large industry: it is estimated that 1.2 billion people use glasses, which are almost always made of plastic materials. The share of bioplastics in eyewear is increasing, but there is a complete lack of a policy framework to certify that a specific product is truly bio-based and to describe how the residual material should be disposed of. Although at present polylactic acid (PLA) is the most abundantly produced bioplastic, in the eyewear industry cellulose acetates constitute by far the main class of employed bioplastics. One of the typical applications of cellulose acetate (in particular cellulose diacetate, CDA) is in the eyewear sector, especially for the manufacture of spectacle frames (Gilbert, 2017; Hansraj et al., 2021), due to its sensorial and hypoallergenic characteristics (Carollo & Grospietro, 2004; Hansraj et al., 2021) and eco-friendliness (Kabasci, 2020). By contacting eyewear companies, we were able to confirm that CDA is by far the most employed bioplastic in the eyewear industry, although no official data on its market share are available. Contextually, in the last years, companies are moving towards formulations with biobased plasticisers, avoiding the use of diethyl phthalate.

In this framework, the present work employed CDA as a case study with the aim of verifying whether a combination of HTC and anaerobic digestion can be a valid route to enhance the disposal phase.

## 2. MATERIALS AND METHODS

### 2.1 Employed bioplastic

The chosen bioplastic feedstock (in the following referred to as CDA) is commercially known as M49. It is based on cellulose diacetate, and was provided in the form of 2x2x1 cm blocks, with an average mass of about 3.5 g per block. Despite not being as commercially widespread as other types of bioplastics, cellulose acetates are widely employed for various applications, such as membranes, cigarette filters, photographic films and packaging. Puls et al. (2011) estimated its global production to be about 800 kton/y in 2008, and unverifiable web searches suggest that this amount may have undergone a tenfold increase in recent years. The literature on the biodegradation of cellulose acetate materials is often contradictory (Puls et al., 2011; Yadav & Hakkarainen, 2021), and the biodegradability of the material depends on several factors, such as the degree of acetylation, the presence of additives, and the biodegradation environment. Kosheleva et al. (2023) reported that cellulose acetate can be biodegraded via anaerobic digestion, but at significantly lower rates than food waste, which further decrease when the test is performed in a semi-continuous apparatus rather than in a batch one. To the best of the authors’ knowledge, the literature does not feature any article reporting on the HTC of cellulose acetate.

CDA was characterised in terms of elemental composition through a LECO 628 Elemental Analyser in accordance with ASTM D-5375 for carbon, hydrogen and nitrogen. The ash content was determined by keeping a sample of CDA at 550°C overnight. The oxygen content was determined by difference with the percentages of C, H, N, and ash. Table 1 reports its composition.

**TABLE 1:** Elemental composition of CDA (mass fractions, with O calculated by difference).

| C (%) | H (%) | N (%) | O (%) | Ash (%) |
|---|---|---|---|---|
| 48.9 ± 0.1 | 6.2 ± 0.3 | 0.0 | 44.9 ± 0.3 | 0.0 |

### 2.2 HTC tests

The HTC runs were performed in a 50 mL batch reactor built in-house at the University of Trento, described in detail in previous publications (Fiori et al., 2014). Each test was performed at least twice. At the beginning of each HTC run, a CDA block was placed inside the reactor together with tap water, at a fixed bioplastic to water mass ratio of 0.125. Then, the reactor was closed, flushed with nitrogen, and heated through an electric resistance up to the selected temperature (180,190, 200, 210, 220 and 250°C). The residence or reaction time was counted after the reactor had reached the set point temperature; the heating time from room temperature to set point temperature was approximately 15-20 minutes. After a residence time of 1 h, the reactor was quickly cooled down dipping it in liquid water. The produced gas volume was measured by letting it flow in a closed water column and measuring the displacement of the water head. Then, the reactor was opened, and its content was filtered with a 0.45 μm filter, separating the

HTC liquor from the hydrochar, which was dried at 105°C overnight – also the HTC reactor was placed in the oven at 105°C overnight. After the drying phase, the reactor and the filter were weighted to assess the mass of the produced hydrochar.

The solid yield (SY) was calculated as the ratio between the mass of the hydrochar produced and that of the initial bioplastic block. The mass of the gas produced was calculated through the ideal gas law, assuming the gas entirely consisted of $CO_2$; from this, the gas yield (GY) was calculated by dividing the mass of the gas produced by the initial mass of the bioplastic. Finally, the liquid yield (LY) was calculated as the one's complement of SY and GY. The yields are reported in percentage terms in what follows.

### 2.3 Anaerobic digestion tests

The anaerobic digestion tests were performed on the untreated bioplastic and on the whole slurry resulting from the HTC process (i.e. the mixture of liquid and solid products), selecting the temperature for which the highest liquid yield was achieved. The slurry was characterised by chemical analyses performed in triplicate. Volatile solids (VS), total solids (TS), total chemical oxygen demand (TCOD), soluble COD (sCOD) were quantified according to Standard Methods (APHA et al., 2012). sCOD was measured after sample filtration on a 0.45 µm paper filter. Total organic carbon (TOC) was measured using a TOC/TN analyzer (Formacs$^{HT-I}$, Skalar).

Biochemical methane potential (BMP) tests were carried out to assess the anaerobic biodegradability of the untreated bioplastic and the whole HTC slurry produced from CDA. Tests were conducted in both mesophilic and thermophilic conditions using serum bottles of 135 mL volume inoculated with anaerobically digested sludge. For mesophilic conditions, digestate samples from the Trento (Trentino, Italy) wastewater treatment plant (M-WW) and Lana (South Tyrol, Italy) OFMSW treatment plant (M-OF) were used as inoculum. Moreover, a digestate sample from the Cadino (Trentino, Italy) OFMSW treatment plant (T-OF) was used as the inoculum for thermophilic tests after its dilution with tap water to obtain a TS content approximately equal to 3%. The use of three different inoculums was chosen to operate with three different bacterial communities and evaluate their influence on the variation in specific biogas production. The main characteristics of the three inoculums used are reported in Table 2.

The inoculums were pre-incubated for 14 days at 37 ± 0.1°C and 55 ± 0.1°C, respectively, for the mesophilic and thermophilic conditions to completely biodegrade the organic substances eventually present in the digestate used as inoculum. The anaerobic digestion tests were carried out in duplicate considering a feeding/inoculum ratio (F/I) equal to 0.5 g $VS_{substrate}$/g $VS_{inoculum}$, with a serum bottle volume fixed at 80 mL. Biogas and biomethane productions were measured according to the procedure reported by Ferrentino et al. (2019). The BMP tests were continuously monitored and stopped when no further changes in the biogas production were observed. Data collected allowed the determination of the daily biogas production (DBP), expressed as $mL_{biogas}$/d, while cumulative data allowed the evaluation of the specific biogas production (SBP), reported as $mL_{biogas}$/g $VS_{added}$. Moreover, the percentage of biodegraded organic matter converted into methane was evaluated according to the method proposed by Beniche et al. (2021).

## 3. RESULTS AND DISCUSSION

### 3.1 HTC

The results of the tests show that HTC is effective for decomposing CDA; Figure 1 shows the trends of the product yields against temperature. In all cases, the solid yield is lower than 100%, proving that even at the lowest temperatures the material undergoes a sort of solubilisation. There is a sharp change between 190 and 200°C, as proved by the drastic decrease of the solid yield: such a small temperature variation is evidently sufficient to activate decomposition reactions, causing nearly all the bioplastic constituents to dissolve into the liquid phase. The solid yield reaches a minimum at 210°C (7.2%) and increases again for higher temperatures, achieving a maximum value of 20.4% at 250°C. This is probably due to the formation of secondary char through the back polymerisation and condensation of organics dissolved into the liquid phase. Meanwhile, the liquid yield follows a trend opposite to that of the solid, while the gas yield increases with the temperature due to a higher occurrence of carboxylation and decarbonylation reactions.

Figure 2 shows the appearance of CDA and of the solid products of HTC, obtained at different temperatures. From

**TABLE 2:** Properties of the inoculums.

| | M-WW | M-OF | T-OF |
|---|---|---|---|
| TS [%] | 2.9 ± 0.1 | 3.8 ± 0.1 | 3.0 ± 0.2 |
| VS [%] | 72.5 ± 1.1 | 56.7 ± 1.3 | 60.8 ± 0.6 |
| TKN [mg TKN/L] | 2300 ± 100 | 4400 ± 250 | 1800 ± 90 |
| $NH_4$-N [mg NH4-N/L] | 1110 ± 30 | 3030 ± 110 | 1200 ± 20 |
| $PO_4$-P [mg PO4-P/L] | 121 ± 5 | 129 ± 8 | 230 ± 10 |
| TP [mg TP/L] | 860 ± 30 | 570 ± 40 | 260 ± 5 |
| sCOD [mg sCOD/L] | 1500 ± 400 | 6050 ± 450 | 4800 ± 230 |
| COD [mg COD/L] | 22100 ± 3500 | 27100 ± 2250 | 23400 ± 3600 |
| pH | 7.6 ± 0.5 | 7.9 ± 0.4 | 7.3 ± 0.2 |

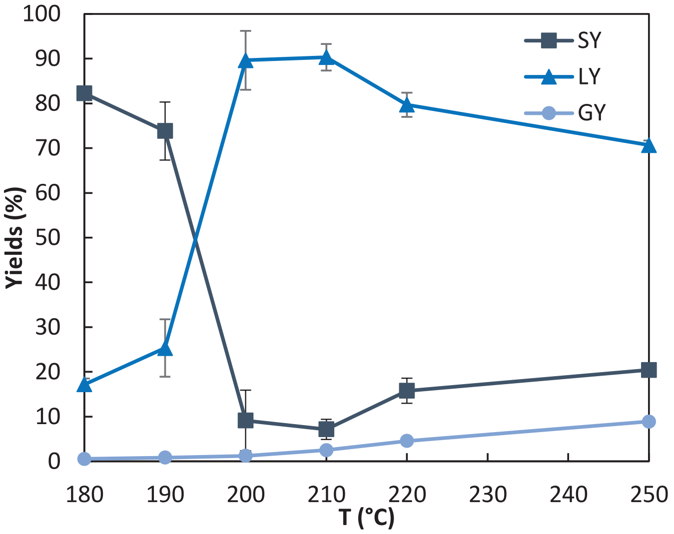


**FIGURE 1:** Trend of the solid (SY), liquid (LY) and gas (GY) yields for the HTC of CDA at various temperatures. The indicators represent the experimental data, the curves connecting the indicators are intended to help the reader in the comprehension of the figure.

**FIGURE 2:** Appearance of CDA and of the solid products obtained at different HTC temperatures.

a visual point of view, when treated at 180 and 190°C the material becomes white and still appears plastic-like. Given the high values of the solid yield, it is likely that only some additives are released, but the CDA core structure is not significantly altered. As temperature increases, the solid products are more visibly altered and, at the highest value of 250°C, they show the typical appearance of hydrochar, being black and granular.

Figure 3 displays the trend of the pH of the HTC liquor against temperature. The pH markedly decreases in the 180-210°C temperature range, whereas it seems to be mostly constant in the 210-250°C range. The pH decrease indicates the increasing concentration of acids in the liquid phase; these may be both compounds that are released from the polymer (such as acetic acid) and other acids that form due to the HTC reactions (Ischia et al., 2022). The relatively low value of the pH at 180°C further proves that the material undergoes some sort of solubilisation even at the lowest tested temperature, despite its change in appearance is not as drastic as for the other temperatures.

From a chemical point of view, the behaviour observed for CDA is rather unusual and not comparable to that of other feedstocks under HTC conditions. For example, the solid yield of plain cellulose diminishes monotonically with increasing values of the HTC temperature (Volpe et al., 2020). While it is difficult to explain this behaviour without detailed chemical analyses, it is known that for cellulose acetate the action of heat causes the release of acetyl groups from the cellulose backbone, which represents the first step in its decomposition (Yadav & Hakkarainen, 2021). This causes the formation of acetic acid, known to catalyse the HTC reactions (Lynam et al., 2011). Thus, the cellulose backbone undergoes chain scissions, leading to the formation of sugar derivatives (like furfural and 5-HMF) and other decomposition compounds dissolved into the liquid phase. At higher temperatures, these compounds undergo repolymerisation and condensation reactions, forming the abovementioned secondary char (Ischia et al., 2022). The trend of the pH seems to support these observations.

On the one hand, the HTC operative conditions that originate the highest liquid yield may be the most suitable for a subsequent anaerobic digestion step, due to the improved accessibility to microorganisms of the chemical species dissolved in the liquid phase. On the other hand, hydrochar is well known for having several interesting applications (Masoumi et al., 2021) – for example, it could simply be burned or gasified, providing thermal energy in a possible biorefinery, or be used as a base for advanced carbons, like adsorbents (Purnomo et al., 2018). While these applications are interesting and may be suitable to valorise a relatively pure stream of cellulose acetates, this

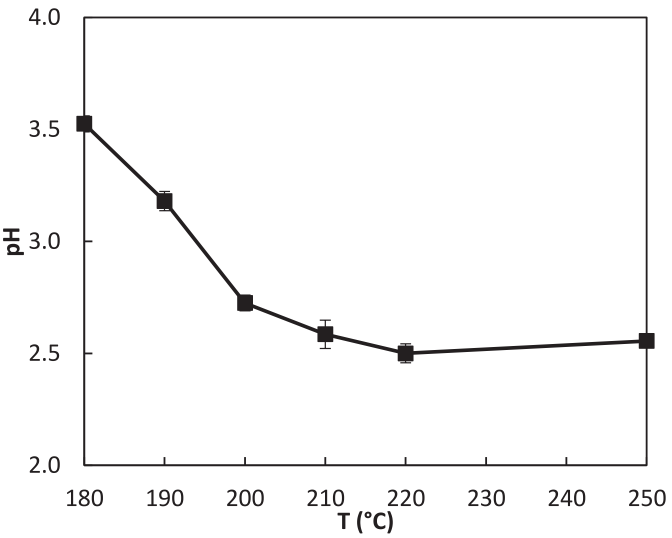


**FIGURE 3:** Trend of the pH of the HTC liquor at various temperatures. The indicators represent the experimental data, the curve connecting the indicators is intended to help the reader in the comprehension of the figure.

work strictly focuses on applying HTC as a pre-treatment to anaerobic digestion, with the aim of enhancing the already-existing disposal chain for OFMSW. Thus, extensive characterisations of the hydrochar and liquor as a function of the HTC operative conditions are left for future studies.

### 3.2 Anaerobic digestion

The anaerobic digestion tests were run on the untreated bioplastic and on the slurry (i.e. hydrochar plus HTC liquor, without any filtration) obtained from HTC at 210°C, which showed the highest liquid yield. The HTC slurry had the following characteristics: 5.3±0.1% TS, 99.5±0.1% VS, 120±5 g COD/L, 50±5 g TOC/L and pH equal to 2.3±0.5. At the beginning of the test, each sample of CDA or HTC slurry was mixed with the inoculum. The pH of the mixture of all the BMP tests ranged between 6.7 and 7.6.

Figure 4a depicts the cumulative SBP. Starting from the raw CDA, it is clear that anaerobic digestion is unable to biodegrade this material satisfactorily (or at all): the final production of biogas is null for the M-OF inoculum and low for the T-OF inoculum. This result agrees with some previous literature, in which thermophilic conditions were reported to be more efficient in biodegrading bioplastics (Yu et al., 2023). Since the inoculums used for the BMP tests were collected in plants where different criteria for OFMSW collection are used, it is likely that the microbial communities are different due to the adaptation to different feedstocks. In the municipalities served by the OFMSW plant in Lana, the disposal of bioplastics, even in the form of collection bags, into OFMSW is strictly prohibited. On the contrary, it has been estimated that the OFMSW sent to the Cadino plant has a mass fraction of bioplastics of about 4%. Since the addition of bioplastics to the OFMSW influences the microbial communities (Bandini et al., 2022), it is possible that with longer residence times and adaption the biodegradation of CDA might increase. However, this aspect needs to be deeply investigated in future works. Moreover, it should be pointed out that reducing the size of the CDA block (for instance by milling) may increase its biodegradability, despite being energy consuming. However, since this is unlikely to happen in a real OFMSW treatment plant and since the whole block was also fed to the HTC process, we decided to feed the whole block to anaerobic digestion as well.

Compared to raw CDA, HTC markedly increases the biogas production. Since HTC converts almost all the bioplastic to liquid compounds, the feedstock becomes much more accessible to microorganisms, as shown by the fact that the biogas production peak is reached after 1-2 days from the beginning of the BMP test (Figure 4b). For the T-OF inoculum, two separate peaks can be observed, which are likely ascribable to the biodegradation of different compounds. The trends of the biomethane production are not reported, as they are identical to those of biogas that are shown in Figure 4, with the methane/biogas volume ratio always ranging between 56% and 59%.

By employing the methodology by Beniche et al. (2021), we were able to assess that a complete biodegradation of the COD could not be achieved in any test. However, the biodegradability of the untreated CDA was 0% in mesophilic conditions and 4% in thermophilic conditions; a pre-treatment with HTC causes at least a threefold increase of this value, which respectively becomes 12%, 13% and 15% for M-OF, T-OF and M-WW. Although there are no studies in the literature on pre-treating CDA via HTC before anaerobic digestion, the results of this study can be compared with previous studies on anaerobic biodegradation of cellulose-based bioplastics. Yagi et al. (2009) achieved a total of 80% biodegradation of cellulose under mesophilic and thermophilic conditions in 15 and 13 days, respectively. Both Calabro' et al. (2020) and Shin et al. (1997) found significant methane production from cellulose-based bioplastics, namely 310 L $CH_4$/kg VS and a methane conversion efficiency of 85% in 44 and 20 days, respectively. On the contrary, Gomez and Michel Jr (2013) observed that after 50 days of anaerobic digestion, only 20-25% of the test-

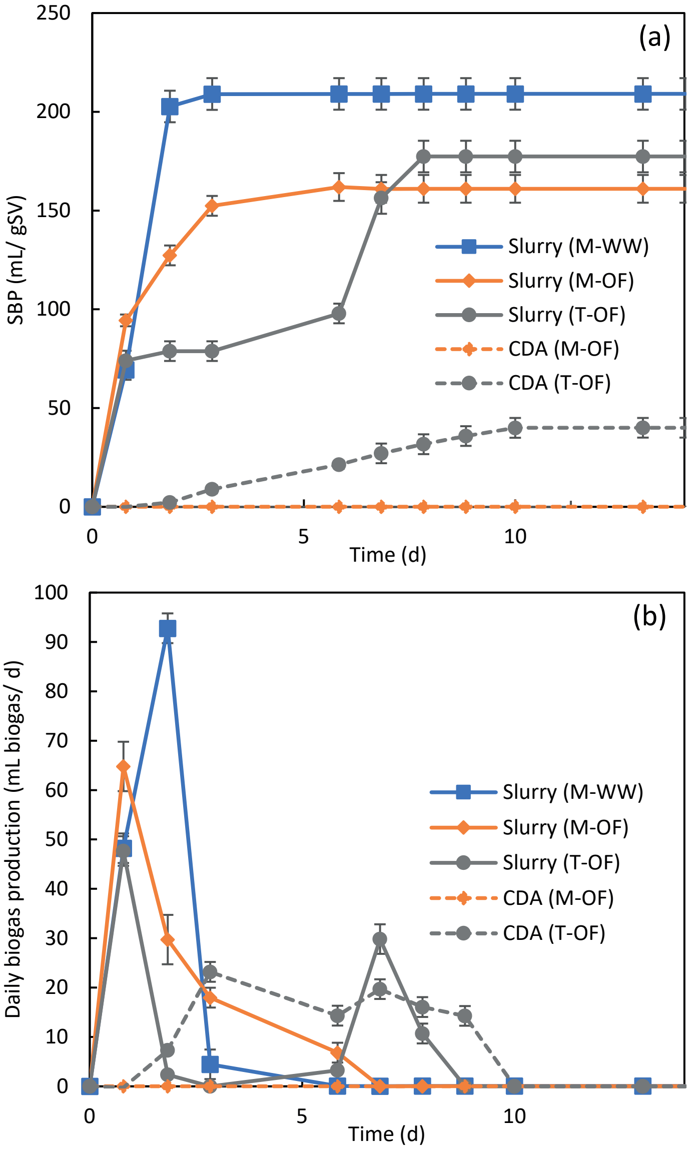


**FIGURE 4:** Cumulative (a) and daily (b) specific biogas production obtained in the BMP tests. The indicators represent the experimental data, the curves connecting the indicators are intended to help the reader in the comprehension of the figure.

ed bioplastics were converted into biogas. Furthermore, Puechner et al. (1995) observed a low degree of biodegradation for cellulose acetate, which was 22% in 60 days. It can thus be noted that the results of previous studies on anaerobic biodegradation of cellulose-based bioplastic are very different.

In any case, these results confirm that HTC is a valid pre-treatment for bioplastics that are destined to anaerobic digestion, both increasing the biogas production and shortening the biodegradation time.

## 4. CONCLUSIONS

A commercial bioplastic based on cellulose diacetate and employed in the eyewear industry was subjected to HTC at temperatures ranging from 180 to 250°C. The material reaches its highest solubilisation level at 210°C, with a residual solid yield of 7.2% and most of the bioplastic dissolved in the liquid phase. For the higher tested temperatures, the solid yield notably increases, likely due to the formation of secondary char due to liquid-to-solid reactions (e.g., polycondensation). The behaviour of cellulose diacetate under HTC conditions is interesting and will be analysed in greater detail in future works, in order to assess the decomposition pathway and products.

The anaerobic digestion (BMP) tests showed that this bioplastic as it is is poorly biodegraded by microbial activity, with low or null biogas yields depending on the employed inoculum. If the HTC slurry is fed to anaerobic digestion instead, both the biogas yield and the biodegradation rate are markedly enhanced, with the peak in the biogas production being reached within a couple of days from the beginning of the BMP test. Also in the case of anaerobic digestion, there are specific aspects that merit to be studied in better detail, such as the role and adaptability of the microbial communities.

In summary, coupling HTC and anaerobic digestion appears as a valuable and effective strategy to enhance biomethane production from waste bioplastics, at the same time facilitating their disposal. While this study focused on a selected bioplastic and batch anaerobic digestion tests, future works may explore whether the present conclusions are still valid when processing a mixture of bioplastics (or bioplastics and traditional plastics) and other organic residues, and how the microbial communities may adapt to these changes in the feedstock.

## AKNOWLEDGEMENTS

The authors are grateful to Fondazione Cariverona for the economic support to the Occhio al Bio! project, funded within the Ricerca&Sviluppo 2020 call. The publication was created with the co-financing of the European Union – FSE-REACT-EU, PON Research and Innovation 2014-2020 DM1062/2021.

The authors thank eyewear companies for providing bioplastic samples.

## REFERENCES

APHA, AWWA, & WEF. (2012). Standard methods for the examination of water and wastewater. In American Public Health Association (21st ed.).

Athanassiou, A. (2021). Si fa presto a dire bioplastiche non illudiamoci: dovremo investire nel loro smaltimento. https://www.corriere.it/pianeta2020/21_aprile_21/si-fa-presto-dire-bioplastiche-non-illudiamoci-dovremo-investire-loro-smaltimento-d52d2b7a-a213-11eb-b3ed-ee5b64f415b7.shtml

Bandini, F., Vaccari, F., Soldano, M., Piccinini, S., Misci, C., Bellotti, G., Taskin, E., Cocconcelli, P. S., & Puglisi, E. (2022). Rigid bioplastics shape the microbial communities involved in the treatment of the organic fraction of municipal solid waste. Frontiers in Microbiology, 13, 4410. https://doi.org/10.3389/fmicb.2022.1035561

Battista, F., Frison, N., & Bolzonella, D. (2021). Can bioplastics be treated in conventional anaerobic digesters for food waste treatment? Environmental Technology & Innovation, 22, 101393. https://doi.org/10.1016/J.ETI.2021.101393

Beniche, I., El Bari, H., Siles, J. A., Chica, A. F., & Martín, M. Á. (2021). Methane production by anaerobic co-digestion of mixed agricultural waste: cabbage and cauliflower. Environmental Technology, 42(28), 4550–4558. https://doi.org/10.1080/09593330.2020.1770341

Bona, D., Scrinzi, D., Tonon, G., Ventura, M., Nardin, T., Zottele, F., Andreis, D., Andreottola, G., Fiori, L., & Silvestri, S. (2022). Hydrochar and hydrochar co-compost from OFMSW digestate for soil application: 2. agro-environmental properties. Journal of Environmental Management, 312, 114894. https://doi.org/10.1016/j.jenvman.2022.114894

Calabro', P. S., Folino, A., Fazzino, F., & Komilis, D. (2020). Preliminary evaluation of the anaerobic biodegradability of three biobased materials used for the production of disposable plastics. Journal of Hazardous Materials, 390, 121653. https://doi.org/10.1016/j.jhazmat.2019.121653

Carollo, P., & Grospietro, B. (2004). 5.5 Plastic materials. Macromolecular Symposia, 208(1), 335–352. https://doi.org/10.1002/masy.200450414

Cazaudehore, G., Guyoneaud, R., Lallement, A., Souquet, P., Gassie, C., Sambusiti, C., Grassl, B., Jiménez-Lamana, J., Cauzzi, P., & Monlau, F. (2023). Simulation of biowastes and biodegradable plastics co-digestion in semi-continuous reactors: Performances and agronomic evaluation. Bioresource Technology, 369, 128313. https://doi.org/10.1016/J.BIORTECH.2022.128313

Ferrentino, R., Merzari, F., Fiori, L., & Andreottola, G. (2020). Coupling Hydrothermal Carbonization with Anaerobic Digestion for Sewage Sludge Treatment: Influence of HTC Liquor and Hydrochar on Biomethane Production. Energies, 13(23), 6262. https://doi.org/10.3390/en13236262

Fiori, L., Basso, D., Castello, D., & Baratieri, M. (2014). Hydrothermal carbonization of biomass: Design of a batch reactor and preliminary experimental results. Chemical Engineering Transactions, 37, 55–60. https://doi.org/10.3303/CET1437010

Folino, A., Pangallo, D., & Calabrò, P. S. (2023). Assessing bioplastics biodegradability by standard and research methods: Current trends and open issues. Journal of Environmental Chemical Engineering, 11(2), 109424. https://doi.org/10.1016/J.JECE.2023.109424

Gadaleta, G., Ferrara, C., De Gisi, S., Notarnicola, M., & De Feo, G. (2023). Life cycle assessment of end-of-life options for cellulose-based bioplastics when introduced into a municipal solid waste management system. Science of The Total Environment, 871, 161958. https://doi.org/10.1016/J.SCITOTENV.2023.161958

Gilbert, M. (2017). Cellulose Plastics. In Brydson's Plastics Materials (pp. 617–630). Elsevier. https://doi.org/10.1016/B978-0-323-35824-8.00022-0

Gómez, E. F., & Michel, F. C. (2013). Biodegradability of conventional and bio-based plastics and natural fiber composites during composting, anaerobic digestion and long-term soil incubation. Polymer Degradation and Stability, 98(12), 2583–2591. https://doi.org/10.1016/j.polymdegradstab.2013.09.018

González, R., Ellacuriaga, M., Aguilar-Pesantes, A., Carrillo-Peña, D., García-Cascallana, J., Smith, R., & Gómez, X. (2021). Feasibility of Coupling Anaerobic Digestion and Hydrothermal Carbonization: Analyzing Thermal Demand. Applied Sciences, 11(24), 11660. https://doi.org/10.3390/app112411660

Hansraj, R., Govender, B., Joosab, M., Magubane, S., Rawat, Z., & Bissessur, A. (2021). Spectacle frames: Disposal practices, biodegradability and biocompatibility – A pilot study. African Vision and Eye Health, 80(1). https://doi.org/10.4102/aveh.v80i1.621

il Dolomiti. (2019). Bioplastica e "'Plastic tax'", tanti si oppongono (anche i gestori dei biodigestori) ma c'è altra via? L'Ue va in quella direzione e l'Italia è tra i maggiori produttori di plastica. https://www.ildolomiti.it/societa/2019/bioplastica-e-plastic-tax-tanti-si-oppongono-anche-i-gestori-dei-biodigestori-ma-ce-altra-via-lue-va-in-quella-direzione-e-litalia-e-tra-i-maggiori-produttori-di-plastica

Il Tirreno Empoli. (2019). Alia conferma: bioplastica non riciclabile Presto un tavolo per risolvere il problema. https://iltirreno.gelocal.it/empoli/cronaca/2019/08/30/news/alia-conferma-bioplastica-non-riciclabile-presto-un-tavolo-per-risolvere-il-problema-1.37398545

Ischia, G., Cutillo, M., Guella, G., Bazzanella, N., Cazzanelli, M., Orlandi, M., Miotello, A., & Fiori, L. (2022). Hydrothermal carbonization of glucose : Secondary char properties , reaction pathways , and kinetics. Chemical Engineering Journal, 449(June), 137827. https://doi.org/10.1016/j.cej.2022.137827

Ischia, G., & Fiori, L. (2021). Hydrothermal Carbonization of Organic Waste and Biomass: A Review on Process, Reactor, and Plant Modeling. Waste and Biomass Valorization, 12(6), 2797–2824. https://doi.org/10.1007/s12649-020-01255-3

Kabasci, S. (2020). Biobased plastics. In Plastic Waste and Recycling (pp. 67–96). Elsevier. https://doi.org/10.1016/B978-0-12-817880-5.00004-9

Karan, H., Funk, C., Grabert, M., Oey, M., & Hankamer, B. (2019). Green Bioplastics as Part of a Circular Bioeconomy. Trends in Plant Science, 24(3), 237–249. https://doi.org/10.1016/j.tplants.2018.11.010

Kosheleva, A., Gadaleta, G., De Gisi, S., Heerenklage, J., Picuno, C., Notarnicola, M., Kuchta, K., & Sorrentino, A. (2023). Co-digestion of food waste and cellulose-based bioplastic: From batch to semi-continuous scale investigation. Waste Management, 156, 272–281. https://doi.org/10.1016/J.WASMAN.2022.11.031

Lynam, J. G., Coronella, C. J., Yan, W., Reza, M. T., & Vasquez, V. R. (2011). Acetic acid and lithium chloride effects on hydrothermal carbonization of lignocellulosic biomass. Bioresource Technology, 102(10), 6192–6199. https://doi.org/10.1016/j.biortech.2011.02.035

Masoumi, S., Borugadda, V. B., Nanda, S., & Dalai, A. K. (2021). Hydrochar: A Review on Its Production Technologies and Applications. Catalysts, 11(8), 939. https://doi.org/10.3390/catal11080939

Mumtaz, H., Sobek, S., Werle, S., Sajdak, M., & Muzyka, R. (2023). Hydrothermal treatment of plastic waste within a circular economy perspective. Sustainable Chemistry and Pharmacy, 32, 100991. https://doi.org/10.1016/J.SCP.2023.100991

Nandakumar, A., Chuah, J.-A., & Sudesh, K. (2021). Bioplastics: A boon or bane? Renewable and Sustainable Energy Reviews, 147, 111237. https://doi.org/10.1016/j.rser.2021.111237

Puechner, P., Mueller, W. R., & Bardtke, D. (1995). Assessing the biodegradation potential of polymers in screening- and long-term test systems. Journal of Environmental Polymer Degradation, 3(3), 133–143. https://doi.org/10.1007/BF02068464/METRICS

Puls, J., Wilson, S. A., & Hölter, D. (2011). Degradation of Cellulose Acetate-Based Materials: A Review. Journal of Polymers and the Environment, 19(1), 152–165. https://doi.org/10.1007/S10924-010-0258-0/FIGURES/11

Purnomo, C., Castello, D., & Fiori, L. (2018). Granular Activated Carbon from Grape Seeds Hydrothermal Char. Applied Sciences, 8(3), 331. https://doi.org/10.3390/app8030331

RameshKumar, S., Shaiju, P., O'Connor, K. E., & P, R. B. (2020). Bio-based and biodegradable polymers - State-of-the-art, challenges and emerging trends. Current Opinion in Green and Sustainable Chemistry, 21, 75–81. https://doi.org/10.1016/j.cogsc.2019.12.005

Scrinzi, D., Bona, D., Denaro, A., Silvestri, S., Andreottola, G., & Fiori, L. (2022). Hydrochar and hydrochar co-compost from OFMSW digestate for soil application: 1. production and chemical characterization. Journal of Environmental Management, 309, 114688. https://doi.org/10.1016/j.jenvman.2022.114688

Shin, P. K., Kim, M. H., & Kim, J. M. (1997). Biodegradability of degradable plastics exposed to anaerobic digested sludge and simulated landfill conditions. Journal of Environmental Polymer Degradation 1997 5:1, 5(1), 33–39. https://doi.org/10.1007/BF02763566

Thakur, S., Chaudhary, J., Sharma, B., Verma, A., Tamulevicius, S., & Thakur, V. K. (2018). Sustainability of bioplastics: Opportunities and challenges. Current Opinion in Green and Sustainable Chemistry, 13, 68–75. https://doi.org/10.1016/j.cogsc.2018.04.013

Vardar, S., Demirel, B., & Onay, T. T. (2022). Degradability of bioplastics in anaerobic digestion systems and their effects on biogas production: a review. Reviews in Environmental Science and Biotechnology, 21(1), 205–223. https://doi.org/10.1007/S11157-021-09610-Z/FIGURES/2

Volpe, M., Messineo, A., Mäkelä, M., Barr, M. R., Volpe, R., Corrado, C., & Fiori, L. (2020). Reactivity of cellulose during hydrothermal carbonization of lignocellulosic biomass. Fuel Processing Technology, 206, 106456. https://doi.org/10.1016/j.fuproc.2020.106456

Yadav, N., & Hakkarainen, M. (2021). Degradable or not? Cellulose acetate as a model for complicated interplay between structure, environment and degradation. Chemosphere, 265, 128731. https://doi.org/10.1016/j.chemosphere.2020.128731

Yagi, H., Ninomiya, F., Funabashi, M., & Kunioka, M. (2009). Anaerobic Biodegradation Tests of Poly(lactic acid) under Mesophilic and Thermophilic Conditions Using a New Evaluation System for Methane Fermentation in Anaerobic Sludge. International Journal of Molecular Sciences, 10(9), 3824–3835. https://doi.org/10.3390/ijms10093824

Yu, C., Dongsu, B., Tao, Z., Xintong, J., Ming, C., Siqi, W., Zheng, S., & Yalei, Z. (2023). Anaerobic co-digestion of three commercial bio-plastic bags with food waste: Effects on methane production and microbial community structure. Science of The Total Environment, 859, 159967. https://doi.org/10.1016/J.SCITOTENV.2022.159967

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# RECOVERY AND UTILISATION OF MUNICIPAL SOLID WASTE INCINERATION BOTTOM ASH: IMPLICATIONS FOR EUROPEAN WASTE MANAGEMENT STRATEGY

Carly Adele Fletcher * and Rachel Dunk

*Department of Natural Sciences, Manchester Metropolitan University, John Dalton Building, Chester Street Manchester, Manchester, Greater Manchester M15 6BH, UK*

**Article Info:**
Received:
8 January 2023
Revised:
14 March 2023
Accepted:
26 April 2023
Available online:
15 June 2023

**Keywords:**
Circular economy
EU waste management
Incineration
End of waste criteria
Lock-in
Data analysis

## ABSTRACT

Over the last two decades, the stated intent of European waste management strategy has evolved from a specific focus on landfill diversion to enabling the transition to a circular economy. Widespread introduction of source-segregation alongside deployment of material recovery technologies have improved MSW management practices across Europe. However, with diminishing returns it has become more difficult to achieve further landfill diversion through increased recycling alone, and incineration rates (across the EU-27 as a whole) have continued to increase. The advantages of incineration include the ability to harness the energy content of the waste alongside a sizeable reduction in mass and volume. However, the remaining solid residues, the most substantial being incinerator bottom ash, present a management issue. Exploring the role of incineration and the utilisation of incineration bottom ash, this paper highlights the potential risks of lock-in in the context of evolving waste policy. A simple thought experiment suggests that while increased use of incineration may help member states achieve 2035 landfill diversion targets, it would also carry a substantive risk of placing the 2035 recycling target out of reach. To address this, a long-term vision concerning the future of incineration is required, where it is recommended that policy which focuses on landfill diversion and the recycling of residual wastes should be strengthened through mechanisms that gradually phase out incineration and distinguish between open and closed-loop recycling.

## 1. INTRODUCTION

As global population and affluence have increased, so has the consumption of goods and services. Although this has improved quality of life for current generations, it is unsustainable; contributing to environmental degradation and associated complex challenges such as resource depletion, climate change, and geopolitical tension (Clark, 2007; Moreno et al., 2016). This has been recognised within the United Nations Sustainable Development Goals, where Goal 12 is to ensure sustainable consumption and production (UN General Assembly, 2015). To address unsustainable consumption, replacing the linear 'take-make-dispose' economic model with a Circular Economy (CE) has been encouraged. In standardising environmental policy across Member States (MS), the European Union (EU) acts as a driving force to improve international standards (Wysokinska, 2016) and can be viewed as being at the forefront of the transition to a CE, having published the Circular Economy Package (CEP) in 2015 (EC, 2015a) and the Circular Economy Action Plan (CEAP) in 2020 (EC, 2020).

To achieve a CE, resource efficiency is promoted through optimisation of production systems, maintenance of resource utility, and promotion of reuse, recycling, and recovery, thereby minimising (and ultimately eliminating) landfilling of waste (Kirchherr et al., 2017). Progressive waste management thus has an integral role to play in the CE transition (Johansson et al., 2020), where one of the most complex to manage waste streams is Municipal Solid Waste (MSW). While MSW constitutes only 10% of total EU waste arisings, it has a high political profile due to its link to consumption patterns and resulting complex composition, where its management is considered an excellent indicator of the quality and efficiency of a MS's waste management strategy (EC, 2015b; Eurostat, 2021).

To date, the northern high-income MSs have been most successful in improving MSW management practices, where the last two decades have seen the accomplishment of "easy gains" (Mihai and Apostol, 2012). For

* Corresponding author:
Carly Adele Fletcher
email: carly.fletcher@mmu.ac.uk

Detritus / Volume 23 - 2023 / pages 43-57
https://doi.org/10.31025/2611-4135/2023.17274

example, the widespread introduction of source-segregation alongside deployment of technological approaches (e.g., material recovery facilities and mechanical biological treatment plants) has delivered a substantial increase in recycling and composting (Cook et al., 2015; Eurostat, 2023; Vountatsos et al., 2016). However, with diminishing returns it becomes more difficult to achieve further landfill diversion through increased recycling, where incineration has increasingly been employed to achieve landfill diversion targets (Eurostat, 2021, 2023). The advantages of incineration include the ability to harness the energy content of the waste alongside a sizeable reduction in mass and volume. However, while the mass of waste is typically reduced by ca. 80%, there remain a number of solid residues, the most substantial being incinerator bottom ash (MSW-IBA).

MSW-IBA is a granular, agglomerated material, that typically comprises a heterogeneous mix of brick, concrete, silicate-phase glass, unburnt organics, clinker and metal fragments (Bourtsalas et al., 2015; Chiang et al., 2012). The presence and concentration of elements reflects waste inputs and is dependent on combustion unit type, where the most common elements are calcium, silicon, aluminium, iron, sodium and manganese, and heavy metals such as antimony, arsenic, barium and beryllium may be present (Margallo et al., 2015). For a detailed physico-chemical analysis of MSW-IBA, see Dou et al. (2017).

Historically, MSW-IBA was landfilled; a sub-optimal solution in terms of resource conservation and environmental safety, and one subject to increasing economic costs and limited by capacity constraints (Chen and Lo, 2015). Current management strategies therefore look to realise the recovery potential for resources such as metals and aggregates (Allegrini et al., 2015; Costa et al, 2020). It is now common practice for ferrous and non-ferrous metals to be recovered through magnetic and eddy-current separation (Allegrini et al., 2014, 2015; Costa et al, 2020). Typically accounting for up to 20% by weight (ferrous 5-15%, non-ferrous 1-5%; Šyc et al, 2020), metal fragments can differ in size and quality, which in turn affects recycling efficiencies (Allegrini et al., 2014, 2015). While recovery of ferrous metals is generally around 80%, for non-ferrous metals recovery can be as low as 30% (Allegrini et al., 2014; Boesch et al., 2014), although advanced separation techniques can increase this to 70% (Biganzoli et al., 2013; Grosso et al., 2011).

The removal of metal fragments increases the quality of MSW-IBA for utilisation as an aggregate, where sieving is used to produce size separated materials with good geotechnical characteristics (Karagiannidis et al., 2013; Šyc et al, 2020). However, chemical and mineralogical characteristics, particularly alkalinity, can result in instability and leaching, where further processing is then required (Dou et al., 2017; Lancellotti et al., 2013). Stabilisation is often achieved through weathering or natural aging; the exposure of an open stockpile to the atmosphere to promote carbonation, resulting in the precipitation of minerals such as calcite and a reduction in pH (Chimenos et al., 2000; Yao et al., 2010). Although it can take up to three months to complete carbonation (such that the leaching potential is minimal), the use of a carbon dioxide enriched atmosphere has the potential to reduce this to two weeks (Margallo et al., 2015).

The most common use of MSW-IBA is as an aggregate, where similar physico-chemical characteristics to natural aggregates allow treated MSW-IBA to be employed as a partial substitute in construction applications (Ahmed and Khalid, 2011). For example, MSW-IBA can replace without detrimental effect; up to 20% of natural aggregate as a sub-base in road construction (Birgisdóttir et al., 2006), up to 25% of clinker used in cement production (Margallo et al., 2014), and up to 15% of cement in low-strength concrete production (Jurič et al., 2006).

In addition to generating income from product sales, using MSW-IBA as a secondary aggregate has two further advantages; reduction of waste landfilled and substitution of natural resources (Margallo et al., 2015; Blasenbauer et al. 2020). Diverting significant volumes of MSW-IBA from landfill reduces the economic and environmental costs of disposal (Birgisdóttir et al., 2006; Olsson et al., 2006). Likewise, substituting raw materials with MSW-IBA avoids the energy use and other environmental costs associated with extraction and processing, and also contributes to mineral stock protection and conservation (Olsson et al., 2006). Indeed, the use of MSW-IBA as an aggregate may be particularly attractive given increasing demand for construction materials and declining availability of natural aggregates (Abbà et al., 2014). However, a recent estimate indicates that full utilisation of MSW-IBA would displace <1% of primary aggregate demand in the EU, suggesting the main benefit is reduction in required landfill capacity (Blasenbauer et al., 2020).

A number of alternative processing and application options explored in the literature are summarised in Table 1. These include the potential for recovering critical raw materials, and potential use as a growth substrate, in construction related products, in hydrogen gas production, and as a purification agent. However, as these do not yet represent substantial utilisation pathways they are not discussed further here.

While the generation, treatment, and management of MSW-IBA has been extensively discussed (see Margallo et al., 2015 and references therein), the production and utilisation of MSW-IBA as a secondary material in the context of evolving EU policy and practice warrants further exploration.

This policy position paper explores the implications and potential consequences of evolving policy for future waste management within the EU, focusing on the use of incineration and utilisation of MSW-IBA in the context of increasingly stringent targets. To provide context, a review of policy documents and academic literature has been used to understand the evolving situation regarding EU waste management strategy, with a specific focus on landfill diversion and material recycling targets. We then examine the different routes to utilisation of MSW-IBA in the EU, before exploring the possible consequences of a continued reliance on incineration for achieving waste management targets under different MSW-IBA utilisation scenarios. Based on this analysis, we then make policy recommendations for achieving targets and avoiding lock-in in the transition to a CE.

**TABLE 1:** Alternative uses for MSW-IBA reported within the literature.

| References | Use | Study details | Conclusion/Limitations |
|---|---|---|---|
| **Material Recovery (Urban Mining)** | | | |
| | **Recovery of REE and CRM** | | |
| Allegrini et al. 2014 | | Detailed MFA (incl. resource recovery potential) of IBA taken from Danish recovery facility. | Conc. of REEs detected in IBA significantly lower than ore. Lack of enrichment options limits recovery of REE's from IBA. |
| Funari et al. 2016 | | Used ICP-MS to determine REE concentration in IBA following digestion (novel method). | REE conc. indicate prospective low streams. Several methods identified to facilitate urban mining (from IBA) |
| Funari et al. 2015 | | Used XRF/ICP-MS to determine elemental composition and CRM conc. of untreated IBA | Considered a low concentration stream for precious/high-tech metals. Concentration of Mg, Cu, Sb & Zn similar to low-grade ore. |
| **Growth Substrate** | | | |
| | **Green / Brown roofs** | | |
| Bates et al. 2015 | | Six-year experiment, testing effects of recycled aggregate type (including IBA) on the development of vegetation on brown roofs. | IBA is not recommended as a brown (biodiversity) roof growth substrate due to limited capacity to hold moisture but could be used in Sedum green roofs. |
| **Pyroxene ceramics** | | | |
| | **Porcelainized stoneware** | | |
| Barbieri et al. 2002 | | Glassy frits obtained from MSW-IBA compared against glass cullet as sintering promoters in production. | Glassy frits improved water absorption and spot resistance but did not significantly change bending strength. |
| Schabbach et al. 2012 | | Replaced feldspar &quartz with IBA (post treatment), characteristics and leaching potential determined. | Mechanical characteristics comparable to commercial products, ISO classification achievable and additional benefits noted. |
| Verbinnen et al. 2017 | | Discusses the use of IBA to produce ceramic materials such as tiles and stoneware. | Amorphous matrix reduces leaching. Ceramics using 5-10% IBA, technical properties not influenced, lower firing temp. |
| **Alkali Activated Cements** | | | |
| | **Hybrid cements** | | |
| Garcia-Lodeiro et al. 2016 | | Compared cement mixes (hybrid, Portland, commercial) with respect to leaching potential, mechanical strength, and reactivity. | Alkali activation of hybrid cement lowered leaching potential. Raised concentration of chloride ions in hybrid cement not suitable in manufacture of structural concrete. |
| Verbinnen et al. 2017 | | Reports on several studies which replace varying proportions of Portland cement with IBA for use in structured materials. | Advises that replacing between 5-10% of Portland cement has no influence on the structural characteristics. Mixtures made with 40+% IBA, detrimental to concrete strength. |
| Chen et al. 2020 | | Assessed the use of MSW IBA as an alkali-activated material as a promising alternative to Portland cement. | Thermal treatment of the IBA (up to 1000°C), eliminated the detrimental effects of metallic AL/Zn and increased crystallinity. Suitable for use as a fine aggregate. |
| Matsumoto & Takaoka, 2022 | | Compared five advanced chloride removal methods; addition of $Na_2CO_3$, addition of $Na_2SO_4$, accelerated carbonation, aging and acid washing against washing only. | Found presence of Friedel's salt can limit success of washing. Aging and acid washing found to improve utilisation in cement. Concludes that optimal recycling should consider environmental impacts and costs. |
| | **Geopolymers** | | |
| Lancellotti et al. 2013 | | Partial substitution of metakaolin within geopolymers, with chemical, elemental and LOI analysis. | IBA has been demonstrated as suitable source materials for producing metakaolin-blended geopolymers. |
| Lancellotti et al. 2015 | | IBA is used as sole source material for geopolymers cured for different lengths of time. | Geo-polymeric networks produced without need for metakaolin. Metallic content may lead to a porous morphology. |
| Ji & Pei, 2019 | | Investigates the use of IBA as a raw input for geopolymers, particularly the generation of hydraulic binders with water. | When mixed with DWTR, samples exhibited higher compressive strength than IBA only samples. A ratio of 80% IBA: 20% DWTR was recommended. |
| **Aeration agent** | | | |
| | **Aerated concrete** | | |
| Song et al. 2015 | | Aluminium and silica from IBA used as aerating agent in production of AAC. | Synthesized IBA-AACs had a higher density, compressive strength and shrinkage when compared against standard. |
| Li et al. 2018 | | Assessed the feasibility of using IBA as a substitute for quartz sand in the preparation of AAC. | Demonstrated that IBA-ACC had reduced gas-foaming time, compressive strength, density, and thermal conductivity. |
| **Hydrogen gas production** | | | |
| | **Use of Aluminium species to generate Hydrogen** | | |
| Saffarzadeh et al. 2016 | | Identification and characterisation of metallic AL / AL-alloys found in IBA and assessed potential to aid the generation of H gas. | Production of H gas ranged between 8.4 and 38.3 l/kg of dry ash, aided by presence of metallic-AL. Inherent alkalinity noted as key parameter in H gas generating reactions. |
| Biganzoli et al. 2013 | | Evaluated the recovery and utilisation of metallic AL, through metal recovery and to generate H gas as a clean fuel. | Successful H gas production, performing better, in terms of overall energy balance, than metal recovery. Economic investment requirements were found to be unjustifiable. |
| **Purification agent** | | | |
| | **Landfill gas purification before energetic valorisation** | | |
| Ducom et al. 2009 | | Pilot plant study assessed qualities of IBA to remove $H_2S$, $CH_4S$ and $C_2H_6S$ from landfill gas. | IBA successful in sequestering $H_2S$ and $CH_4S$ through acid-base reactions, $C_2H_6S$ retained by physical adsorption. |
| | **Mesoporous silica materials** | | |
| Liu et al. 2014 | | Mesoporous silica materials, synthesised from IBA, evaluated in the removal of heavy metals from aqueous solutions. | Mesoporous silica materials were successfully synthesized and shown to have potential as adsorbents for the removal of heavy metals from aqueous solutions. |

*Abbreviations: Rare Earth Elements (REE); Critical Raw Materials (CRM); Material Flow Analysis (MFA); Inductively Coupled Plasma - Mass Spectrometry (ICP-MS); X-Ray Fluorescence (XRF); International Standards Organisation (ISO); Loss On Ignition (LOI); Drinking Water Treatment Residue (DWTR), autoclaved aerated concrete (AAC).*

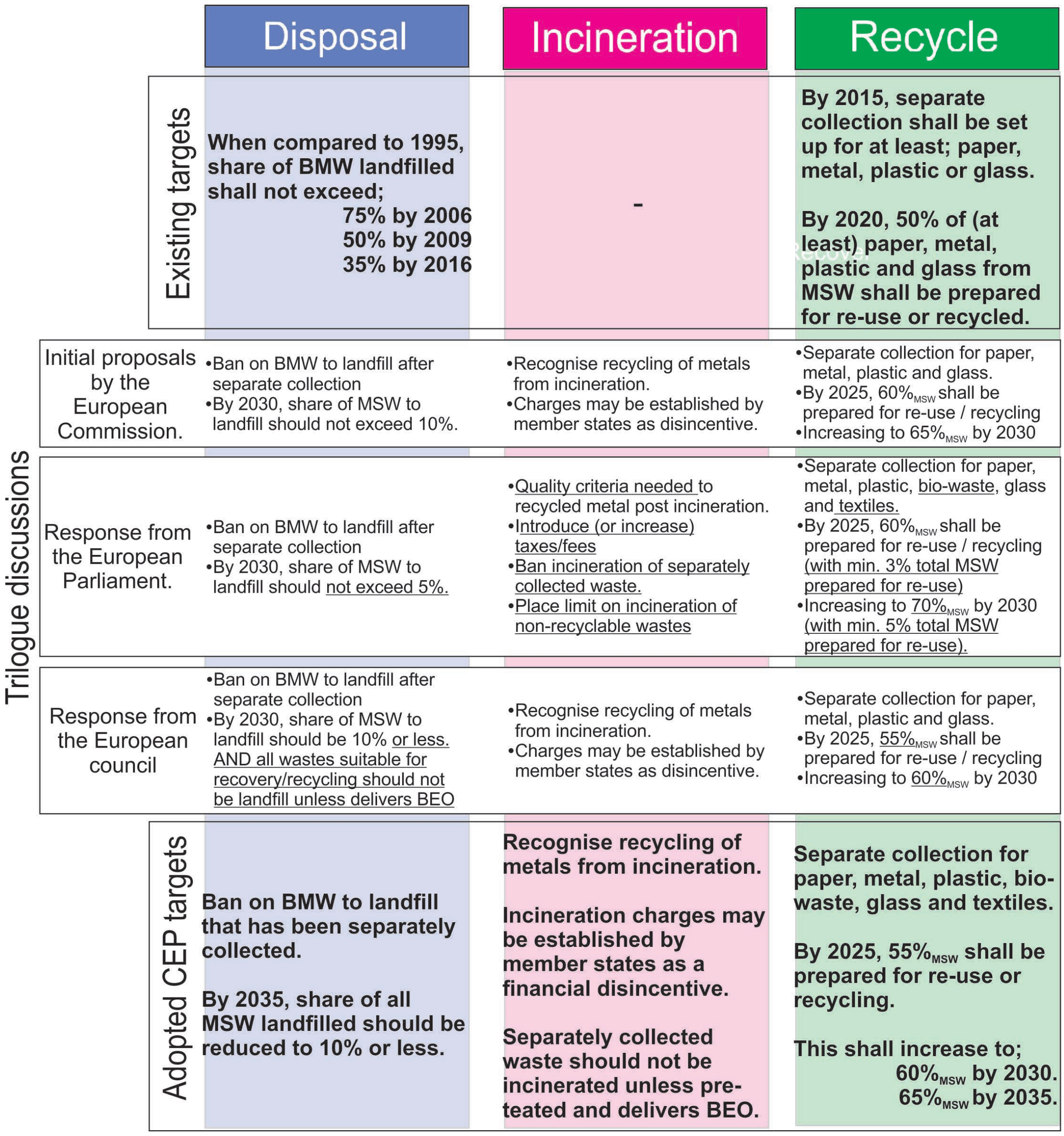
## 2. EVOLUTION OF EUROPEAN WASTE STRATEGY

Two EU directives that have driven significant changes in MSW management (by setting legally binding performance targets) are the Landfill Directive (LD; 1999/31/EC; EC, 1999) and the Waste Framework Directive (WFD; 2008/98/EC; EC, 2008). Both directives were amended by the CEP, with further targeted revision of the WFD (in line with the CEAP) expected in 2023 (EC, 2022a).

During development of the CEP, trilogue discussions between the European Commission (EC), Parliament and Council considered a number of proposed amendments (Figure 1). The final version introduced a ban on the landfilling of separately collected wastes, a maximum MSW landfill target of 10% and a recycling target of 65% by 2035 (EC, 2015a). However, with the compromises reached during trilogue, two key areas of missed opportunity can be identified, neither of which have been addressed in the CEAP.

First, the waste hierarchy itself has not been revised, where the lack of nuance could have implications in the CE transition (Gharfalkar et al., 2015). Specifically, no distinction is made between open-loop recycling (where often the value of the resource decreases i.e. down-cycling and only one extra lifecycle is achieved) and closed-loop recycling (where value is maintained i.e. re-cycling, or increased i.e. up-cycling, and several lifecycles can be achieved). As such, strategies contributing to targets do not need to consider value maintenance or the number of lifecycles achieved (Bartl, 2014; Gharfalkar et al., 2015).

Second, despite the EC's recognition that increased incineration capacity may jeopardise recycling, no limits (absolute or relative) were introduced. While incineration has a valid role to play in the treatment of other waste streams,



| | | Disposal | Incineration | Recycle |
|---|---|---|---|---|
| Existing targets | | **When compared to 1995, share of BMW landfilled shall not exceed;**<br>**75% by 2006**<br>**50% by 2009**<br>**35% by 2016** | - | **By 2015, separate collection shall be set up for at least; paper, metal, plastic or glass.**<br>**By 2020, 50% of (at least) paper, metal, plastic and glass from MSW shall be prepared for re-use or recycled.** |
| Trilogue discussions | Initial proposals by the European Commission. | •Ban on BMW to landfill after separate collection<br>•By 2030, share of MSW to landfill should not exceed 10%. | •Recognise recycling of metals from incineration.<br>•Charges may be established by member states as disincentive. | •Separate collection for paper, metal, plastic and glass.<br>•By 2025, $60\%_{MSW}$ shall be prepared for re-use / recycling<br>•Increasing to $65\%_{MSW}$ by 2030 |
| | Response from the European Parliament. | •Ban on BMW to landfill after separate collection<br>•By 2030, share of MSW to landfill should not exceed 5%. | •Quality criteria needed to recycled metal post incineration.<br>•Introduce (or increase) taxes/fees<br>•Ban incineration of separately collected waste.<br>•Place limit on incineration of non-recyclable wastes | •Separate collection for paper, metal, plastic, bio-waste, glass and textiles.<br>•By 2025, $60\%_{MSW}$ shall be prepared for re-use / recycling (with min. 3% total MSW prepared for re-use)<br>•Increasing to $70\%_{MSW}$ by 2030 (with min. 5% total MSW prepared for re-use). |
| | Response from the European council | •Ban on BMW to landfill after separate collection<br>•By 2030, share of MSW to landfill should be 10% or less. AND all wastes suitable for recovery/recycling should not be landfill unless delivers BEO | •Recognise recycling of metals from incineration.<br>•Charges may be established by member states as disincentive. | •Separate collection for paper, metal, plastic and glass.<br>•By 2025, $55\%_{MSW}$ shall be prepared for re-use / recycling<br>•Increasing to $60\%_{MSW}$ by 2030 |
| Adopted CEP targets | | **Ban on BMW to landfill that has been separately collected.**<br>**By 2035, share of all MSW landfilled should be reduced to 10% or less.** | **Recognise recycling of metals from incineration.**<br>**Incineration charges may be established by member states as a financial disincentive.**<br>**Separately collected waste should not be incinerated unless pre-teated and delivers BEO.** | **Separate collection for paper, metal, plastic, bio-waste, glass and textiles.**<br>**By 2025, $55\%_{MSW}$ shall be prepared for re-use or recycling.**<br>**This shall increase to;**<br>**$60\%_{MSW}$ by 2030.**<br>**$65\%_{MSW}$ by 2035.** |

**FIGURE 1:** Evolution of Circular Economy Package targets during trilogue discussions, with significant differences underlined (EC, 2008, 2015b-c; CEU, 2017a-b).

such as the decontamination of hospital waste (Gielar and Helios-Rybicka, 2013) and the extraction of phosphorus from sewage sludge (Kleemann et al., 2017), the incineration of MSW has negative implications for the CE. Where the CE seeks to maintain and recirculate materials and resources, incineration destroys them (albeit with energy recovery), diverting materials with high calorific value away from recycling pathways. In particular, the management of plastic wastes is largely realised through energy recovery, where reasons for this include complex material composition, inadequate source separation, a lack of automated sorting equipment, and the low cost of waste plastics relative to fossil fuels (Schneider and Ragossnig, 2015). The failure to place limits on incineration thus undermines implementation of the waste hierarchy (Malinauskaite et al., 2017) and incurs the risk of lock-in, potentially stifling the emergence of more sustainable alternatives (Corvellec et al. 2013; Svingstedt & Corvellec, 2018). Indeed, given capital costs up to €180 million and operating contracts exceeding 25 years (Nixon et al., 2013), expansion of incineration infrastructure risks both technological and contractual lock-in, where municipal authorities may be tied-in to supply contracted quantities of waste over decades, irrespective of changes in waste composition, volumes, and policy (Schneider and Ragossnig, 2015; Svingstedt & Corvellec, 2018). Despite these risks being highlighted during CEP trilogue, they are only somewhat obliquely addressed within the final text, with advice to consider the risk of “stranded assets” in investment decisions (highlighting the need to consider feedstock availability over the lifespan of new installations without neglecting separate collection and recycling obligations), while MSs with higher ambition may elect to introduce incineration charges and limits at a national level (EC, 2017). Likewise, the approach taken in the CEAP is to reduce residual waste generation (non-recycled, i.e., landfill + incineration with or without energy recovery) through promoting waste prevention and separation for recycling (rather than to place explicit limits on incineration), encouraging the wider introduction of economic instruments such as landfill and incineration taxes as a mechanism to achieve this (EC, 2020).

While the headline target within the CEAP is to reduce residual waste by half by 2030, this is an EU-wide non-binding commitment. Furthermore, in addition to a lack of incineration targets, as yet there are no specific waste prevention targets. Rather, the EC has placed an obligation on MSs to establish Waste Prevention Plans (WPP). However, where WPPs have been established by progressive MSs, they tend to be reliant on qualitative initiatives, and thus may be less effective (Johansson & Corvellec, 2018). In practice, this means the only well-defined and legally binding target-based drivers are the landfill diversion and recycling targets.

In addition to revising targets, the CEP, and to a greater extent the CEAP, did address broader aspects of consumption and production. Of particular relevance to waste management was the acknowledgement that continued uncertainty regarding secondary materials had restricted their use, thereby limiting resource recovery and landfill diversion (EC, 2015b, 2016). For example, the use of secondary aggregates in road construction has been hindered by perceived performance concerns and additional costs (Huang et al., 2007). In light of this, the CEP and CEAP addressed the further development of secondary materials markets and the strengthening of quality standards such as End of Waste (EoW) criteria (Bartl, 2015, 2020; EC, 2015b, 2020).

## 3. GENERATION AND UTILIZATION OF MSW-IBA IN LIGHT OF THE EU POLICY

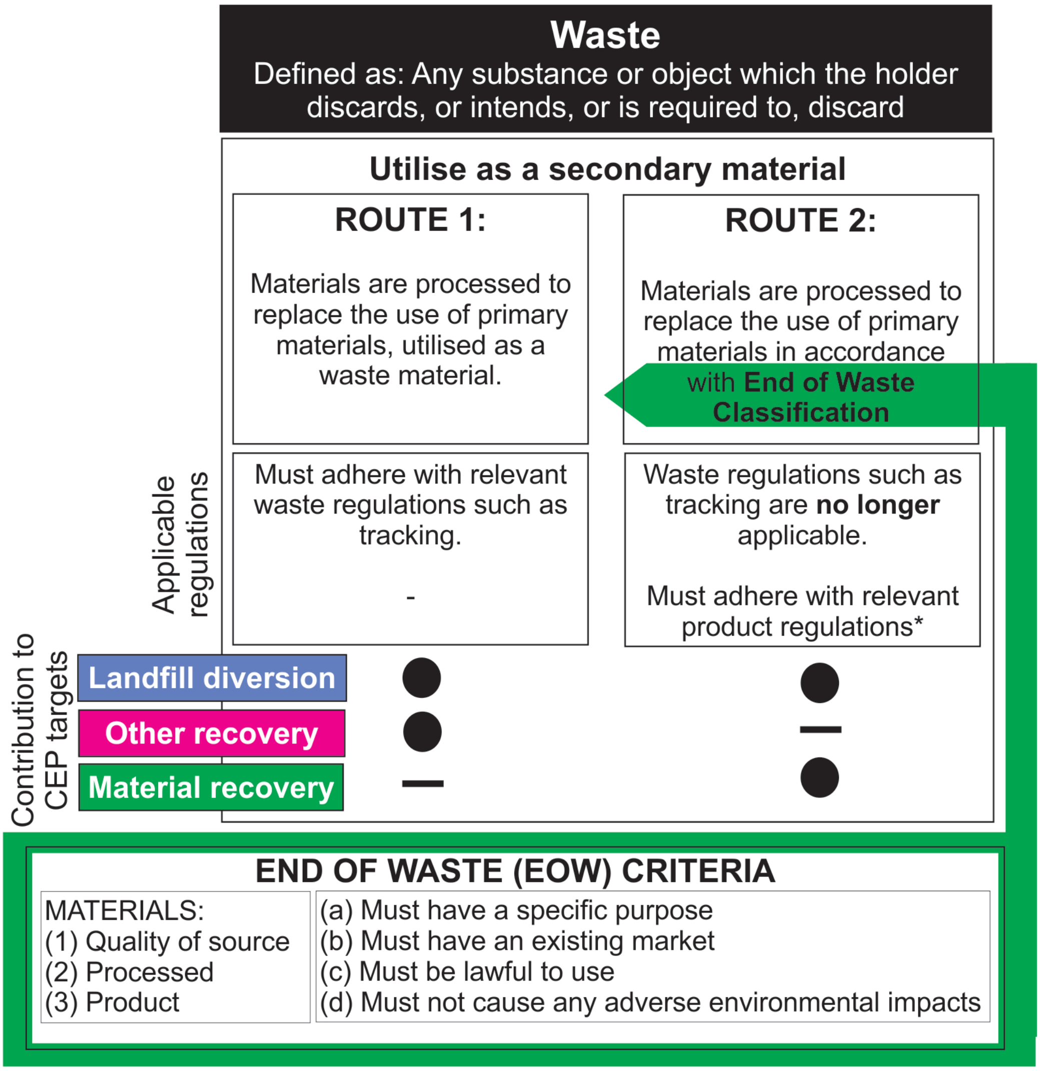
### 3.1 Recovery and utilisation of MSW-IBA as a secondary material in the EU

Within the EU, MSW-IBA may be utilised via two routes (as a waste or non-waste) with differing implications for landfill diversion and recycling targets (Figure 2).

Under Route 1, secondary materials maintain the status of a waste. As such, the transport, utilisation, and continued monitoring of MSW-IBA must comply with relevant waste legislation, be shown to have no adverse environmental effects, and adhere to restrictions and pre-treatment conditions prompted by national legislation (Kuo et al., 2013; Lancellotti et al., 2013; Van Gerven et al., 2005; van der Sloot et al., 2001). Utilisation through Route 1 contributes to landfill diversion but does not contribute to recycling, instead aligning with the definition of ‘other recovery’ (EC, 2008).

Alternatively, secondary materials can be utilised via Route 2, where EoW seeks to address known barriers to the development of secondary material markets. Specifically, the common perception that recovered materials are of lower quality than primary materials and the restricted ability to transport materials across national boundaries due to a lack of harmonisation in waste definitions between MSs (Delgado et al., 2009). Successful application of EoW criteria would classify the material as a ‘non-waste’, removing the need to apply waste regulations. Instead, the secondary material is treated in the same fashion as primary materials, being subject to product regulations, import / export regulations (with free trade within the EU internal market), and where appropriate, regulations concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) (EC, 2006). Achieving non-waste status allows the material to be counted towards both landfill diversion and recycling targets (EC, 2008).

EoW status can be defined at different stages of material recovery depending on the quality of the waste stream and the extent of processing required. Firstly, EoW can be defined for high quality waste materials that require minimal processing, where to date the EC have laid down EU-wide criteria for iron, steel, aluminium, and copper scrap, and glass cullet. However, for lower quality materials such as MSW-IBA, achieving EoW will require either processing to meet quality levels equivalent to that of primary materials, or being processed into a recognisable and marketable product. In all cases, the material / product must also adhere to the four qualifying criteria for EoW (Figure 2). A recently completed scoping assessment (carried out under the CEAP) has identified plastics and textiles as priorities



** Product regulations include those associated with consumer rules, the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), Import / Export rules and trade (European Single Market).*

**FIGURE 2:** Routes to utilisation of recovered wastes as secondary materials and implications for achievement of landfill diversion and material recovery targets (where • is positive, – is negative).

for the development of further EU-wide EoW criteria (EC, 2022a). Beyond this, Article 6 (paragraph 2) of the WFD places the onus on MSs to develop national EoW criteria, where these do not automatically apply across the EU.

Within the EU, MSW-IBA utilisation via Route 1 has become commonplace within construction, with use in cement production, as sub-base in road construction, in other civil engineering projects, and as landfill cover (Table 2). The extent of utilisation is primarily influenced by incentives which encourage use in lieu of disposal (e.g., landfill taxes) in combination with market conditions which dictate the quantities and quality of MSW-IBA required (Villanueva et al, 2006; WRAP, 2006).

Despite this widespread use, no EoW criteria have been established to date (van Zomeren and Velzeboer, 2017). While Denmark has considered developing EoW for MSW-IBA, it was concluded that it would be inappropriate in unbound applications (Villanueva et al., 2006). Specific concerns related to traceability, where removal of waste tracking and monitoring requirements has the potential to undermine environmental protection (e.g., risks to groundwater from leaching of MSW-IBA at an unrecorded site with no monitoring) (Villanueva et al., 2006). While it was acknowledged that EoW status could ease administrative and export burdens, it was also highlighted that MSW-IBA has low financial value and tends to be used locally, thus unconstrained export is not necessarily required (Villanueva et al, 2006). Indeed, Denmark uses incineration to treat a large proportion of MSW (between a half and two-thirds; Eurostat 2023a) and achieves high MSW-IBA utilisation rates (Table 2) without the use of EoW criteria.

### 3.2 EU incineration trends, MSW-IBA production and utilisation rates, and implications for targets

Examination of trends in MSW treatment within the EU clearly shows the impact of the LD (EC, 1999) and WFD (EC, 2008), where a combination of increased material recycling, composting and anaerobic digestion (which col-

**TABLE 2:** IBA production and utilisation rates in EU countries with MSW incineration plants. For IBA produced, A is calculated from Blasenbauer et al. (2020) and B from CEWEP Country Reports (CEWEP, 2021). The off-landfill utilisation rates are from Blasenbauer et al. (2020). The total (on & off landfill) utilisation rates and method of disposal or utilisation are from CEWEP Country Reports. The total values are calculated from the available data weighted according to the mass of waste incinerated or IBA produced as appropriate.

| Country | IBA produced (wt% treated waste) | | Mineral IBA utilisation rate (wt% IBA) | | Method of Mineral IBA disposal or utilisation | CEWEP report year |
|---|---|---|---|---|---|---|
| | A | B | off landfill | total | | |
| Austria | 20.4% | 21.5% | 0% | 0% | 100% landfill | 2018 |
| Belgium | 14.2% | 18.9% | 69% | - | Secondary building material | 2016 |
| Czechia | 30.8% | 23.9% | 0% | 0% | 100% landfill | 2016 |
| Denmark | 16.2% | 17.6% | 99% | 99% | Recycled (road construction, harbours etc.) | 2010 |
| Estonia | 23.2% | | 0% | | | |
| Finland | 18.8% | 16.8% | 20% | 100% | Recycled/recovered – mainly in construction but also in asphalt production and construction block | 2018 |
| France | 19.7% | 20.8% | 80% | 80% | 80% recovery (e.g. road construction); 17% landfill; 3% other | 2010 |
| Germany | 24.2% | 27.0% | 30% | - | Road construction, noise barriers & other technical applications, recovery on landfill (ways, shaping) | 2018 |
| Hungary | 28.6% | 21.8% | 0% | 0% | 100% landfill | 2018 |
| Ireland | 17.5% | 15.7% | 0% | 100% | 100% recovery on landfill (cover & engineering material) | 2018 |
| Italy | 16.9% | 17.8% | 85% | 71% | 71% recovery; 29% landfill | 2012-13 |
| Lithuania | 26.8% | | 0% | | | |
| Luxembourg | 16.5% | 16.8% | 0% | - | Road construction | 2018 |
| Netherlands | 25.0% | 22.7% | 100% | 100% | 40% road construction; 36% noise barriers; 13% landfill construction; 11% other (e.g. bound in products) | 2012-13 |
| Poland | 21.6% | 25.0% | 60% | - | Block fabrication; landfill | 2010-11 |
| Portugal | 16.9% | 19.6% | 56% | 60% | Road construction, landfill cover & backfilling | 2018 |
| Slovakia | 21.4% | | 0% | | | |
| Spain | 18.3% | 16.8% | 58% | - | Landfill use (ridge, regularization, etc), road construction, cement production | 2010-11 |
| Sweden | 18.3% | 16.3% | 0% | 100% | 100% recovery as landfill construction material | 2018 |
| **Minimum** | **14.2%** | **15.7%** | **0%** | **0%** | | |
| **Maximum** | **30.8%** | **27.0%** | **100%** | **100%** | | |
| **Total** | **20.8%** | **22.0%** | **53%** | **79%** | | |

lectively contribute to recycling targets), and incineration (with or without energy recovery), has reduced the amount of waste landfilled by 55% between 1995 and 2021 (Figure 3; Eurostat, 2023a). Focusing on incineration, while rates have been relatively stable over the last decade in some MSs (e.g. France, Italy), and declined in others (e.g. Germany, the Netherlands), they have increased in the majority of MSs, where incineration capacity in the EU-27 increased by 39% between 2010 and 2020 (from 126Mt/yr to 199 Mt/yr; Eurostat, 2023b), while the amount of incinerated waste increased by 16% over the same period (from 53 Mt/yr to 62 Mt/yr; Eurostat, 2023a). While this expansion in incineration has helped to drive landfill diversion, if it were to continue apace it could place the 2035 landfill diversion (<10% MSW) and recycling (65% MSW) targets at risk.

Here we take a closer look at the potential consequences of continued growth in incineration across the EU. While there is a high degree of variability between MSs, both in terms of landfill diversion and the extent to which incineration is utilised, examination of the data presented in Figure 4 allows four ballpark estimates of the incineration rate (INC) required to meet the 2035 landfill diversion target under current EU waste management practices.

- For all landfill rates (LF) less than 10%$_{MSW}$ (corresponding to data spanning 1999-2021 from ten MSs), the median LF was 1.5%$_{MSW}$, and median INC was 44.9%$_{MSW}$.
- Focusing on the most recent data, the eight MSs individually achieving a LF less than 10%$_{MSW}$ in 2021, have a weighted mean LF of 0.5%$_{MSW}$ and weighted mean INC of 37.6%$_{MSW}$.
- Linear regression of the 2015-2019 data and 2020-21 data indicates a LF of 10%$_{MSW}$ corresponds to an INC of 41.7-44.0%$_{MSW}$.
- Looking across the 13 best performing (lowest LF) MSs in 2019 (the most recent year for which data from all 27 MSs is available), a collective LF of 9.9%$_{MSW}$ (<10%$_{MSW}$) corresponds to an INC of 33.3%$_{MSW}$. This compares to an overall 2019 LF of 24.3%$_{MSW}$ and INC of 27.0%$_{MSW}$.

Thus, if the current trend of increasing incineration to achieve landfill diversion were to continue, it seems reasonable to assume (for a first order estimate) that achiev-

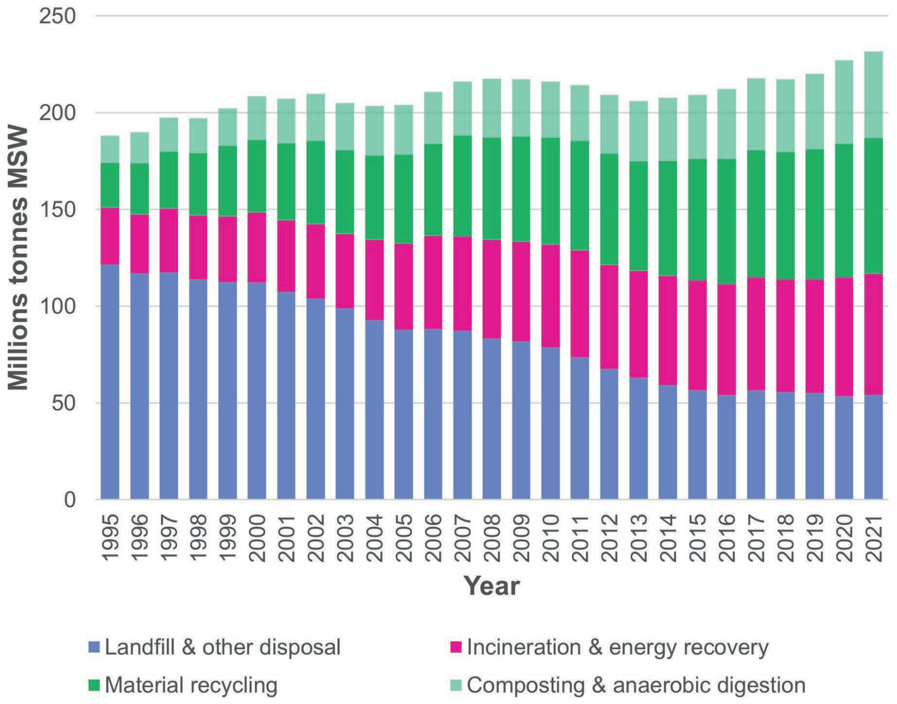


**FIGURE 3:** Treatment of MSW in the EU-27 from 1995 to 2021 (data from Eurostat, 2023a).

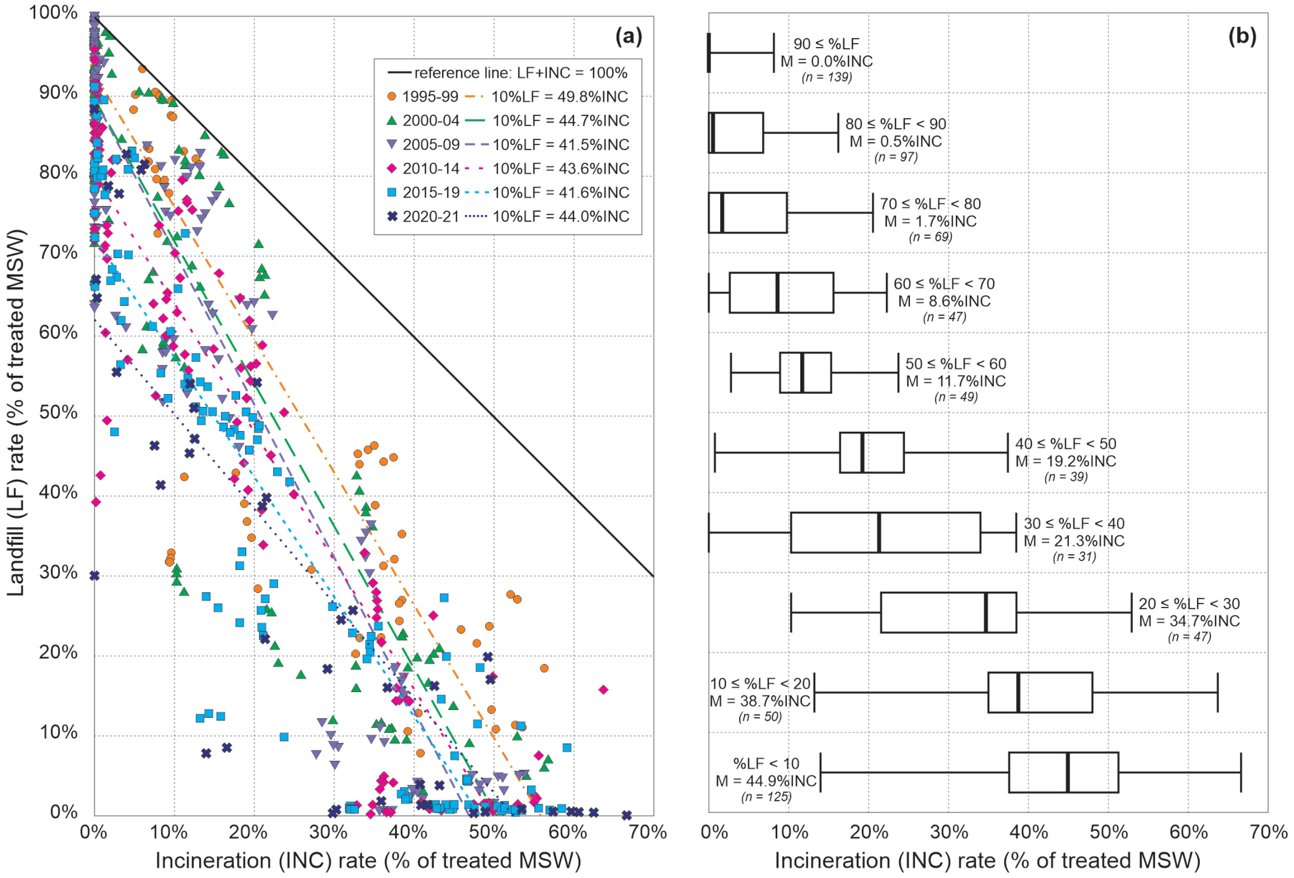


**FIGURE 4:** Incineration (INC) rate relative to landfill (LF) rate for all EU Member States 1995-2021, expressed as a percentage of treated MSW (data from Eurostat, 2023a). (a) Scatter plot with linear regression of quinquennial (R2 = 0.82 to 0.88) and 2020-21 (R2 = 0.69) data. The distance between the data and the LF+INC = 100% reference line reflects the implementation of other waste management strategies (recycling, composting, anaerobic digestion) that act to decrease LF. (b) Box and whisker plot of INC rate by LF rate decile groups, where M = median INC rate.groups, where M = median INC rate.

ing a LF of ca. 1-10%$_{MSW}$ (5.5±4.5%$_{MSW}$) would correspond to an increase in INC from ca. 27.0%$_{MSW}$ to ca. 33.3-44.9%$_{MSW}$ (39.1±5.8%$_{MSW}$). The consequences of such an increase for landfill diversion and recycling rates are illustrated in Figure 5, where the calculation of the material flows are explained briefly below.

Based on data presented in Table 2, the mass of IBA produced ranges from 14.2% to 30.8% of the mass of incinerated waste (22.5±8.3%$_{INC}$). While metal recovery from MSW-IBA is now relatively common practice, the extent to which it has been implemented across the EU is unclear due to incomplete reporting. Nonetheless, the available data (Table 3) indicates overall recovery rates consistent with the literature (see Section 2.1). Using a gross extraction of 8.2% of the mass of MSW-IBA and a metal fraction of 0.75±0.04 gives a metal recovery of 6.2±0.3%$_{IBA}$. Combining this with the MSW-IBA production rate and expressing it relative to the mass of incinerated waste gives a metal recovery of 1.4±0.5%$_{INC}$ and a residual (mineral) MSW-IBA production rate of 21.1±8.3%$_{INC}$. If an INC of 39.1±5.8%$_{MSW}$ is then applied, the total mass of MSW-IBA produced is 8.8±3.5%$_{MSW}$, with a mineral MSW-IBA production of 8.3±3.5%$_{MSW}$, and a metal recovery of 0.5±0.2%$_{MSW}$. If it assumed that all MSW that is not landfilled or incinerated enters recycling pathways (100-LF-INC = 55.4±7.3%$_{MSW}$), this gives a total recycling rate of 56.8±7.4%$_{MSW}$.

We now consider the implications of the above flows for meeting the landfill and recycling targets under different MSW-IBA utilisation scenarios (Figure 6).

If we consider current practice (Table 2), MSW-IBA is typically landfilled, utilised on landfill, or utilised as a waste in off-landfill construction. If all mineral MSW-IBA were landfilled, MSs would be at significant risk of exceeding their 2035 landfill allowance (Scenario 1). The risk is reduced if utilisation rates (on and off landfill) comparable to today are assumed (Scenario 2a) and is eliminated with full utilisation (Scenario 2b). However, these utilisation pathways qualify as 'other recovery' and do not contribute to recycling targets. As such, incinerating such a large proportion of MSW and maintaining current MSW-IBA management practices would put the 2030 recycling target at risk, and the 2035 target out of reach. Indeed, for both the 2035 landfill diversion and recycling targets to be achievable, a significant fraction (if not all) of the MSW-IBA would need to be utilised off-landfill via material recovery pathways (Scenarios 3a and b). Clearly, maximising metal recovery should be prioritised (given the economic value of metals and that this is a closed-loop recycling pathway), however, the potential gains are relatively small (doubling metal recovery increases the overall recycling rate by 0.5%$_{MSW}$). Conversely, if EoW were achieved for mineral MSW-IBA, then the risk of failing to meet the recycling target would

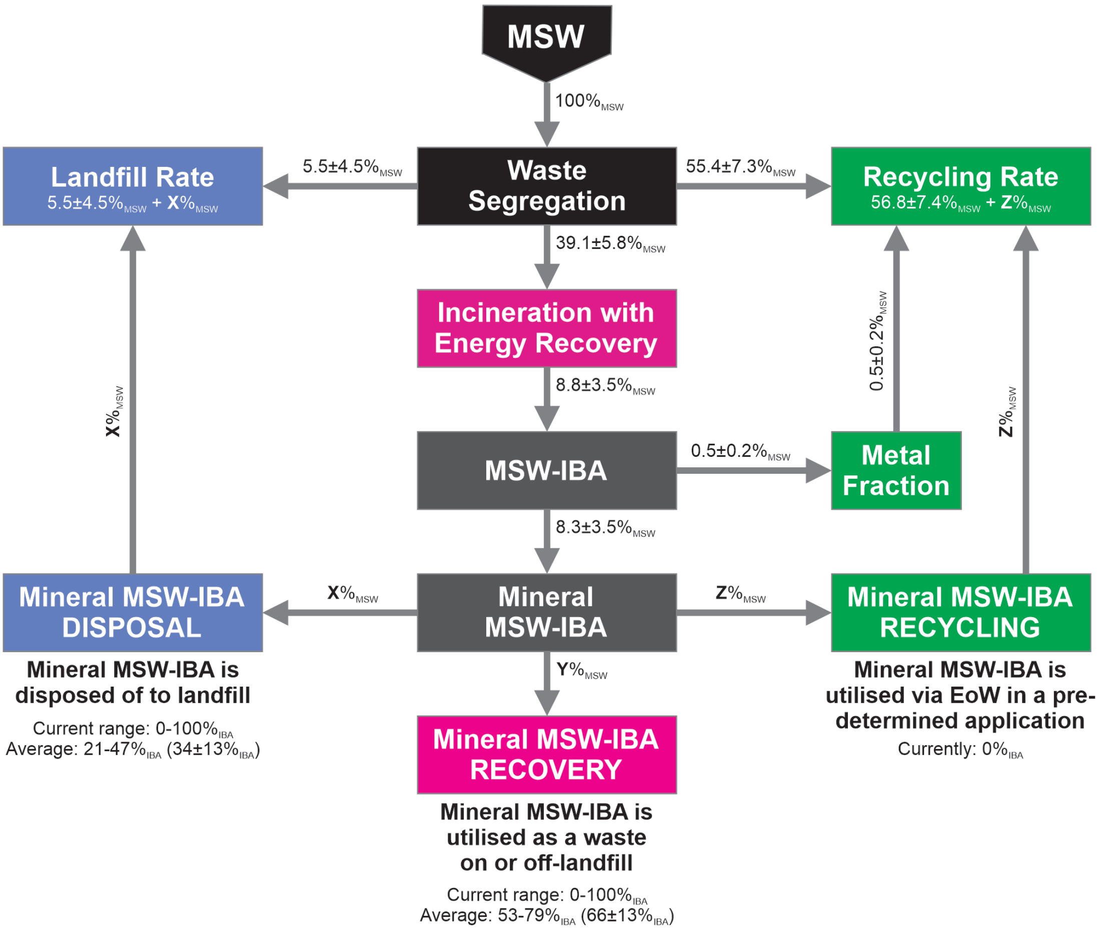


**FIGURE 5:** Calculation of materials flows based on the current rates of incineration.

**TABLE 3:** Metal recovery rates for EU countries based on available CEWEP Country Reports (CEWEP, 2021). n.d. Country Report states no data; n.r. not reported. Total gross material extracted ($\%_{IBA}$) calculated across all available data, weighted by mass of IBA produced. Total metal fraction ($f_{met}$) calculated across all available data, weighted by mass of gross material extracted. M = total metals, F = ferrous, NF = non-ferrous.

| Country | Gross material extracted ($\%_{IBA}$) | | | Metal fraction ($f_{met}$) | | | CEWEP report year |
|---|---|---|---|---|---|---|---|
| | M | F | NF | M | F | NF | |
| Austria | n.r. | 3% | n.r. | | | | 2018 |
| Belgium [1] | 8.0% | 7.1% | 0.9% | | | | 2016 |
| Czechia | 5.9% | 5.9% | 0.0% | | | | 2016 |
| Finland [2] | 11.3% | 6.5% | 4.8% | | | 0.29 | 2016 |
| Germany | 9.0% | 7.7% | 1.3% | | | | 2018 |
| Hungary | n.r. | 19.5% | n.d. | | | n.d. | 2018 |
| Ireland | 10.1% | 8.1% | 2.0% | | | 0.78 | 2018 |
| Italy | 0.0% | 0.0% | 0.0% | | | | 2012-13 |
| Luxembourg | 8.5% | 6.9% | 1.6% | | | | 2018 |
| Netherlands [3] | 11.0% | 7.9% | 3.1% | 0.76 | 0.88 | 0.46 | 2012-13 |
| Poland | 0.0% | 0.0% | 0.0% | | | | 2010-11 |
| Portugal | 6.4% | n.r. | n.r. | | | | 2018 |
| Spain [4] | 17.1% | (9.9%) | (0.3%) | 0.60 | | | 2010-11 |
| Sweden | 7.2% | 5.4% | 1.8% | | | | 2018 |
| **Minimum** | **0.0%** | **0.0%** | **0.0%** | | | | |
| **Maximum** | **17.1%** | **19.5%** | **4.8%** | | | | |
| **Total (M)** | **8.2%** | **-** | **-** | 0.71 | - | - | |
| **Total (F+NF)** | **8.2%** | **6.7%** | **1.5%** | 0.80 | 0.88 | 0.46 | |

*Notes on Country Report data: [1] States likely underestimated; [2] Company level data; [3] Ferrous and stainless steel given separately; [4] Ferrous and non-ferrous reported as net material extracted.*

be relatively low for 2025 and 2030, although a moderate to high risk remains for 2035.

In addition to allowing MSW-IBA utilisation to contribute towards recycling targets, achieving EoW may also help higher off-landfill utilisation rates to be realised, including in bound applications and higher value products, and through further exploration of alternative uses (see Table 1 for example). This is an important consideration for both the recycling and landfill diversion targets due to the expected decrease in the amount of waste landfilled, and thus the capacity to utilise MSW-IBA in landfill construction and backfilling operations. For a conservative estimate of the future reduction in landfill capacity and given the lack of quantitative targets for waste prevention, we might assume that MSW generation stays broadly constant (with waste prevention offsetting the moderate increase in waste generation observed over the past 10-15 years, Figure 3). Under these circumstances, achieving the landfill diversion target would see the amount of MSW sent to landfill decrease by around two fifths (from $23\%_{MSW}$ to $10\%_{MSW}$). Thus, if incineration is employed as a key (although non-optimal) mechanism to achieve landfill diversion, off-landfill utilisation will need to be enhanced. Verbinnen et al. (2017) argue that EoW would improve public acceptance of MSW-IBA derived materials and suggest that introduction of EU-wide criteria would boost recycling by setting unequivocal environmental standards (e.g., leaching limits). However, Blasenbauer et al. (2020) consider the feasibility of developing EU-wide EoW criteria to be low due to country specific situations (where appropriate limit values will vary according to local environmental conditions), and instead suggest a parallel approach, with defined fields of application, a risk-based assessment system for establishing limit values, and standardised test methods.

Irrespective of the approach, whether further utilisation of MSW-IBA is desirable, or would serve to facilitate further progress down a dead-end route towards lock-in of incineration, is an open question. For example, even with full utilisation of MSW-IBA via EoW (Scenario 3b), little to no headroom remains should recycling targets be strengthened in the future, a distinct possibility given that the 70% target proposed by the European Parliament during CEP trilogue (albeit not enacted) was supported by several MSs (EEB, 2017). Indeed, there would be a high risk of failing to meet a future higher ambition target without potentially costly withdrawal from incineration.

## 4. DISCUSSION AND CONCLUSIONS

While successive EU policies have driven significant improvements in waste management, prioritisation of landfill diversion has resulted in an unbalanced emphasis where mechanisms do not always align with the waste hierarchy. This is illustrated by the increasing prominence of incineration, where several (otherwise) progressive MSs have deployed incineration as a means to achieve landfill diversion targets. Driven by near-term targets, the use of incineration

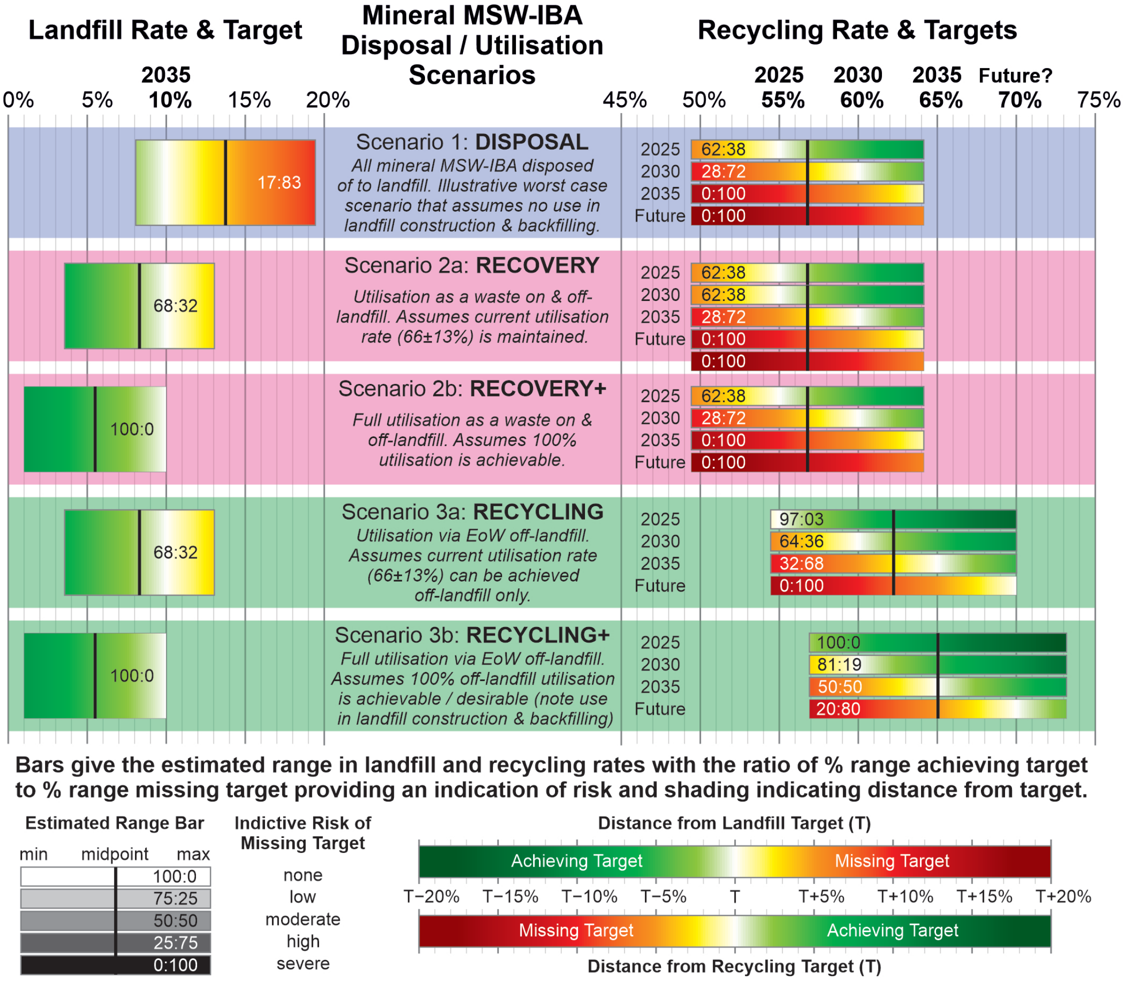


**FIGURE 6:** Implications for meeting landfill and recycling targets under different MSW-IBA utilisation scenarios.

is now at risk of lock-in, contradicting ambitions set out in the CEAP (through revision of the Industrial Emissions Directive) to "increase investment in new, cleaner technologies […] whilst avoiding lock-in to obsolete technologies" (EC, 2020). In addition, contra to CE principles, high-quality recyclates are diverted from closed-loop recycling routes to ensure that a consistent calorific value of input materials is met for efficient energy production.

That being said, it seems prudent to acknowledge that the use of incineration across the EU is likely to increase in the short to medium term. In light of the current energy crisis, the preeminent policy priority is enhancing security of supply through domestic/regional energy generation while maintaining progress towards climate neutrality (EC, 2022b), with FEAD (the European Waste Management Authority) promoting the potential contribution of energy from waste (EfW) (FEAD, 2022). This highlights the position of incineration at the intersection between waste and energy policy, where competing priorities and the application of different control mechanisms increases the risk of unintended consequences. For example, current discussions concerning the future of the EU Emissions Trading System (EU-ETS) have indicated that EfW incineration plants may no longer be exempt (ZWE, 2022). This introduces uncertainty and a likely reluctance on the part of MSs to establish or retain incineration taxes (which are promoted, but not required, by the CEAP), as inclusion within the EU-ETS would then effectively result in double taxation set against a backdrop of rising energy prices (Recycling Magazine, 2022).

With respect to the achievement of waste targets, it is acknowledged that this study has focused on analysis of aggregated data and does not consider other issues that may influence actual recycling rates, such as the success-rate of source separation initiatives. That being said, the analysis presented here clearly indicates that continuation of the current trend towards increased utilisation of incineration across the EU-27 as a whole carries a substantive risk of placing the 2035 recycling target out of reach. From a purely instrumental perspective, this could be addressed by re-defining the operations that qualify as recycling. At present, utilisation of MSW-IBA only counts towards recycling targets under EoW. However, with no EoW criteria published at either EU or MS level, and with valid questions raised regarding the desirability and operability of EoW for MSW-IBA, development of this route seems unlikely. Nonetheless, high utilisation rates in a variety of construction applications have been achieved (albeit use as a waste), thus reclassifying off-landfill utilisation as recycling would aid MSs in achieving targets. For example, as suggested by Blasenbauer et al. (2020), specific qualifying routes (for different applications) could be defined using a process based on EoW criteria, with additional allowances made for the local context.

While increased incineration would assist MSs to generate energy while achieving ambitious landfill diversion targets, and the proposal above would help achieve recycling targets, it would not help advance the transition to a CE. Additionally, while the CEAP presents a roadmap to 2035, a longer-term vision for achieving a fully CE is lacking. To address this, a clear vision for incineration is required to ensure that today's priority does not becomes tomorrow's status quo when moving beyond the current energy crisis. This vision should consider two missed policy opportunities related to the articulation and implementation of the waste hierarchy which, if unaddressed, may restrict the emergence of more sustainable solutions in the future. First, a lack of EU-wide limits or constraints on incineration (either overall or on specific material streams). Second, a lack of nuance within the waste hierarchy, where no distinction is made between open- and closed-loop recycling. As such, the open-loop utilisation of mineral MSW-IBA after EoW would currently have equal weighting to closed-loop recycling of the feedstock material. Similarly, while it is entirely conceivable that MSW-IBA could be utilised in the same application both with and without EoW, only the former would currently contribute towards recycling targets while the latter would be classed as 'other recovery'. Thus, to aid the CE transition, it is suggested that future policy development should consider the following points:

- To address the risks of technological and contractual lock in, clear policy signals on the future role of incineration within a climate-neutral CE must be formulated, and mechanisms to phase out incineration (by technology and/or of specific waste-streams) on an appropriate timeline should be developed. Given the identification of plastics and textiles as priorities for the development of new EU-wide EoW criteria (EC, 2022a), these represent excellent early candidates for introducing waste-stream specific limits on incineration. This could be similar in formulation to mechanisms within the Renewable Energy Directive, where the use of crop-based biofuels is gradually being phased out from a maximum contribution of 7% in 2020 to 0% in 2030 due to sustainability concerns (EC, 2018).
- To ensure a clear incentive for maintaining value, the definition of recycling should be expanded and the introduction of a weighting system that differentiates between closed and open-loop recycling should be considered. Such a system should reflect the relative value of each utilisation route with respect to the waste hierarchy, consider system maturity, and could also confer credit for utilisation as a waste. For example, a hard-to-treat waste stream might see a weighting <1 for post-incineration utilisation of MSW-IBA as waste material, 1 for post-incineration utilisation of MSW-IBA following application of EoW or direct open-loop recycling (i.e., no incineration), and >1 for closed-loop recycling, thereby providing an incentive for innovation. Again, such weighting mechanisms have been successfully deployed within energy policy for both renewable energy technologies and low emission vehicles (del Rio et al. 2017; EC, 2009).

In conclusion, while acknowledging that incineration will continue to take place, particularly in the near-term, this study argues that to avoid lock-in, policy focusing on promoting diversion of waste from landfill and recycling of residual wastes require bolstering by the introduction of mechanisms that gradually phase out incineration and distinguish between open and closed-loop recycling. Furthermore, to deploy the mechanisms described above, a long-term roadmap is needed, which not only provides an overarching objective for all environmental policy to realise a CE, but also indicates the relevant milestones and feedback loops required for waste management.

## REFERENCES

Abbà, A., Collivignarelli, M. C., Sorlini, S., Bruggi, M. 2014. On the reliability of reusing bottom ash from municipal solid waste incineration as aggregate in concrete. Composites Part B. 58, 502-509. https://doi.org/10.1016/j.compositesb.2013.11.008

Ahmed, A. T., Khalid, H. A. 2011. Effectiveness of novel and traditional treatments on the performance of incinerator bottom ash waste. Waste Manage. 31(12), 2431-2439. https://doi.org/10.1016/j.wasman.2011.07.015

Allegrini, E., Maresca, A., Olsson, M. E., Holtze, M. S., Boldrin, A., Astrup, T. F. 2014. Quantification of the resource recovery potential of municipal solid waste incineration bottom ashes. Waste Manage. 34(9), 1627-1636. https://doi.org/10.1016/j.wasman.2014.05.003

Allegrini, E., Vadenbo, C., Boldrin, A., Astrup, T. F. 2015. Life cycle assessment of resource recovery from municipal solid waste incineration bottom ash. J. Environ. Manage. 151, 132-143. https://doi.org/10.1016/j.jenvman.2014.11.032

Barbieri, L., Corradi, A., Lancellotti, I., Manfredini, T. 2002. Use of municipal incinerator bottom ash as sintering promoter in industrial ceramics. Waste Manage. 22(8), 859-863. https://doi.org/10.1016/S0956-053X(02)00077-6

Bartl, A. 2014. Ways and entanglements of the waste hierarchy. Waste Manage. 34(1) 1-2 https://doi.org/10.1016/j.wasman.2013.10.016

Bartl, A. 2015. Withdrawal of the circular economy package: A wasted opportunity or a new challenge? Waste Manage. 44, 1-2. https://doi.org/10.1016/j.wasman.2015.08.003

Bartl, A. 2020. The Circular Economy Package of the European Union: Are new paths being taken or is it an old story? Detritus. 12, 12-17 https://doi.org/10.31025/2611-4135/2020.13991

Bates, A.J., Sadler, J.P., Greswell, R.B., Mackay, R. 2015. Effects of recycled aggregate growth substrate on green roof vegetation development: A six-year experiment. Landscape Urban Plann. 135, 22-31. https://doi.org/10.1016/j.landurbplan.2014.11.010

Biganzoli, L., Ilyas, A., Praagh, M. v., Persson, K. M., Grosso, M. 2013. Aluminium recovery vs. hydrogen production as resource recovery options for fine MSWI bottom ash fraction. Waste Manage. 33(5), 1174-1181. https://doi.org/10.1016/j.wasman.2013.01.037

Birgisdóttir, H., Pihl, K. A., Bhander, G., Hauschild, M. Z., Christensen, T. H. 2006. Environmental assessment of roads constructed with and without bottom ash from municipal solid waste incineration. Transp. Res. Part D. 11(5), 358-368. https://doi.org/10.1016/j.trd.2006.07.001

Boesch, M. E., Vadenbo, C., Saner, D., Huter, C., Hellweg, S. 2014. An LCA model for waste incineration enhanced with new technologies for metal recovery and application to the case of Switzerland. Waste Manage. 34(2), 378-389. https://doi.org/10.1016/j.wasman.2013.10.019

Bourtsalas, A., Vandeperre, L., Grimes, S., Themelis, N., Koralewska, R., Cheeseman, C. 2015. Properties of ceramics prepared using dry discharged waste to energy bottom ash dust. Waste Manage. Res. 33(9), 794-804. https://doi.org/10.1177/0734242X15584846

Blasenbauer, D., Huber, F., Lederer, J., Quina, M. J., Blanc-Biscarat, D., Bogush, A., Bontempi, E., Blondeau, J., Chimenos, J. M., Dahlbo, H., Fagerqvist, J., Giro-Paloma, J., Hjelmar, O., Hyks, J., Keaney, J., Lupsea-Toader, M., O'Caollai, C. J., Orupõld, K., Pająk, T., Simon, F. -G., Svecova, L., Šyc, M., Ulvang, R., Vaajasaari, K., Van Caneghem, J., van Zomeren, A., Vasarevičius, S., Wégner, K., Fellner, J. 2020. Legal situation and current practice of waste incineration bottom ash utilisation in Europe. Waste Manage. 102, 868-883. https://doi.org/10.1016/j.wasman.2019.11.031

Chen, Y.-C., Lo, S.-L. 2015. Evaluation of greenhouse gas emissions for several municipal solid waste management strategies. J. Cleaner Prod. 113, 6.6-612. https://doi.org/10.1016/j.jclepro.2015.11.058

Chen, B., van Zijl, M. B., Keulen, A., Ye, G. 2020. Thermal Treatment on MSWI Bottom Ash for the Utilisation in Alkali Activated Materials. KnE Engineering, 5(4), 25–35. https://doi.org/10.18502/keg.v5i4.6792

Chiang, Y. W., Ghyselbrecht, K., Santos, R. M., Meesschaert, B., Martens, J. A. 2012. Synthesis of zeolitic-type adsorbent material from municipal solid waste incinerator bottom ash and its application in heavy metal adsorption. Catal. Today. 190(1), 23-30. https://doi.org/10.1016/j.cattod.2011.11.002

Chimenos, J. M., Fernández, A. I., Nadal, R., Espiell, F. 2000. Short-term natural weathering of MSWI bottom ash. J. Hazard. Mater. 79(3), 287-299. https://doi.org/10.1016/S0956-053X(03)00074-6

Clark, G. 2007. Evolution of the global sustainable consumption and production policy and the United Nations Environment Programme's (UNEP) supporting activities. J. Cleaner Prod. 15, 492-498. https://doi.org/10.1016/j.jclepro.2006.05.017

Confederation of European Waste-to-Energy Plants [CEWEP]. 2021. CEWEP Country Reports. Retrieved from https://www.cewep.eu/category/facts/data-and-maps/

Cook, E., Wagland, S.T., Coulon, F. 2015. Investigation into the non-biological outputs of mechanical-biological treatment facilities. Waste Manage. 46, 212-226. https://doi.org/10.1016/j.wasman.2015.09.014

Corvellec, H., Zapata Campos, M. J., Zapata, P. 2013. Infrastructures, lock-in, and sustainable urban development: the case of waste incineration in the Göteborg Metropolitan Area. J. Cleaner Prod. 50, 32-39 https://doi.org/10.1016/j.jclepro.2012.12.009

Costa, G., Polettini, A., Pomi, R., Spagnuolo, R. 2020. Enhanced Separation of Incinerator Bottom Ash: Composition and Environmental Behaviour of Separated Mineral and Weakly Magnetic Fractions. Waste Biomass Valor 11, 7079–7095. https://doi.org/10.1007/s12649-020-01106-1

Council of the European Union [CEU]. 2017a. Proposal for a Directive of the European Parliament and of the Council amending Directive 2008/98/EC on waste - Preparation for the trilogue.

Council of the European Union [CEU]. 2017b. Proposal for a Directive of the European Parliament and of the Council amending Directive 1999/31/EC on the landfill of waste - Preparation for the trilogue.

Delgado, L., Catarino, A. S., Eder, P., Litten, D., Luo, Z., Villanueva, A. 2009. End-of-Waste Criteria. Joint Research Centre, Institute for Prospective Technological Studies: European Commission https://doi.org/10.2791/28650

del Rio, P., Resch, G., Ortner, A., Liebmnn, L., Busch, S., Panzer, C. 2017. A techno-economic analysis of EU renewable electricity policy pathways in 2030. Energy Policy. 104, 484-493. https://doi.org/10.1016/j.enpol.2017.01.028

Dou, X., Ren, F., Nguyen, M. Q., Ahamed, A., Y, K., Chan, W. P., Chang, V. W. -C. 2017. Review of MSWI bottom ash utilization from perspectives of collective characterization, treatment and existing application. Renewable Sustainable Energy Rev. 79, 24-38. https://doi.org/10.1016/j.rser.2017.05.044

Ducom, G., Radu-Tirnoveanu, D., Pascual, C., Benadda, B., Germain, P. 2009. Biogas – Municipal solid waste incinerator bottom ash interactions: Sulphur compounds removal. J. Hazard. Mater. 166(2), 1102-1108.

European Commission [EC]. 1999. Council directive 1999/31/EC of 26 April 1999 on the landfill of waste. Retrieved September 1, 2022, from http://data.europa.eu/eli/dir/1999/31/2018-07-04

European Commission [EC]. 2006. Regulation (EC) No 1907/2006 of the European Parliament and of the Council of 18 December 2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH). Retrieved September 1, 2022, from http://data.europa.eu/eli/reg/2006/1907/2022-05-01

European Commission [EC]. 2008. Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain Directives. Retrieved September 1, 2022, from http://data.europa.eu/eli/dir/2008/98/oj

European Comission [EC]. 2009. Regulation (EC) No 443/2009 of the European Parliament and of the Council of 23 April 2009 setting emission performance standards for new passenger cars as part of the Community's integrated approach to reduce CO 2 emissions from light-duty vehicles. Retreived November 18, 2022 from http://data.europa.eu/eli/reg/2009/443/oj

European Commission [EC]. 2015a. Closing the loop - An EU action plan for the Circular Economy. Retrieved September 1, 2022, from https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX-%3A52015DC0614

European Commission [EC]. 2015b. Proposal for a directive of the European Parliament and of the Council amending Directive 1999/31/EC on the landfill of waste. Retrieved September 1, 2022, from https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:52015PC0594

European Commission [EC]. 2016. First circular economy action plan. Retrieved September 1, 2022, from https://environment.ec.europa.eu/topics/circular-economy/first-circular-economy-action-plan_en

European Commission [EC]. 2017. The role of waste-to-energy in the circular economy. Retrieved September 1, 2022, from https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=COM:2017:0034:FIN:EN:PDF

European Commission [EC]. 2018. Directive (EU) 2018/2001 of the European Parliament and of the Council of 11 December 2018 on the promotion of the use of energy from renewable sources. Retreived November 18, 2022 from http://data.europa.eu/eli/dir/2018/2001/oj

European Commission [EC]. 2020. A new Circular Economy Action Plan For a cleaner and more competitive Europe. Retrieved September 1, 2022, from https://environment.ec.europa.eu/strategy/circular-economy-action-plan_en

Europen Commission [EC]. 2022a. Environmental impact of waste management – revision of EU waste framework. Retrieved November 18, 2022, from https://ec.europa.eu/info/law/better-regulation/have-your-say/initiatives/13225-Environmental-impact-of-waste-management-revision-of-EU-waste-framework_en

European Commission [EC]. 2022b. REPowerEU: A plan to rapidly reduce dependence on Russian fossil fuels and fast forward the green transition. Retrieved December 31, 2022 from https://ec.europa.eu/commission/presscorner/detail/en/ip_22_3131

European Environmental Bureau [EEB]. 2017. EU environment ministers need to show more support for EU waste laws, says the EEB. Retrieved from https://eeb.org

Eurostat 2021. Municpal waste statistics. Retreived from https://ec.europa.eu/eurostat/statistics-explained/index.php?title=Municipal_waste_statistics#Municipal_waste_generation

Eurostat. 2023a. Municipal waste by waste management operations. [Data extracted 10.03.2023] Retrieved from https://ec.europa.eu/eurostat/data/database

Eurostat. 2023b Number and capacity of recovery and disposal facilities by NUTS 2 regions. [Data extracyed 10.03.2023] Retrieved from https://ec.europa.eurostat/data/database

FEAD. 2022. The waste management sector is not at its full capacity of producing energy. Retrieved November 18, 2022 from https://fead.be/position/the-waste-management-sector-is-not-at-its-full-capacity-of-producing-energy/

Funari, V., Bokhari, S.N.H., Vigliotti, L., Meisel, T., Braga, R. 2016. The rare earth elements in municipal solid waste incinerators ash and promising tools for their prospecting. J. Hazard. Mater. 301, 471-479. https://doi.org/10.1016/j.jhazmat.2015.09.015

Funari, V., Braga, R., Bokhari, S.N.H., Dinelli, E., Meisel, T. 2015. Solid residues from Italian municipal solid waste incinerators: A source for "critical" raw materials. Waste Manage. 45, 206-216. https://doi.org/10.1016/j.wasman.2014.11.005

Garcia-Lodeiro, I., Carcelen-Taboada, V., Fernández-Jiménez, A., Palomo, A. 2016. Manufacture of hybrid cements with fly ash and bottom ash from a municipal solid waste incinerator. Constr. Build. Mater. 105, 218-226. https://doi.org/10.1016/j.conbuildmat.2015.12.079

Gharfalkar, M., Court, R., Campbell, C., Ali, Z., Hillier, G. 2015. Analysis of waste hierarchy in the European waste directive 2008/98/EC. Waste Manage. 39, 305-313. https://doi.org/10.1016/j.wasman.2015.02.007

Gielar, A., Helios-Rybicka, E. 2013. Environmental impact of a hospital waste incineration plant in Krakow (Poland). Waste Manage. Res. 31(7), 722-728. https://doi.org/10.1177/0734242X13485868

Grosso, M., Biganzoli, L., Rigamonti, L. 2011. A quantitative estimate of potential aluminium recovery from incineration bottom ashes. Resour. Conserv. Recycl. 55(12), 1178-1184. https://doi.org/10.1016/j.resconrec.2011.08.001

Huang, Y., Bird, R. N., Heidrich, O. 2007. A review of the use of recycled solid waste materials in asphalt pavements. Resour. Conserv. Recycl. 52(1), 58-73. https://doi.org/10.1016/j.resconrec.2007.02.002

Ji, Z., Pei, Y. 2019. Geopolymers produced from drinking water treatment residue and bottom ash for the immobilization of heavy metals. Chemosphere, 225, 579-587. https://doi.org/10.1016/j.chemosphere.2019.03.056

Johansson, N., Corvellec, H. 2018. Waste policies gone soft: An analysis of European and Swedish waste prevention plans. Waste Manage. 77, 322-332 https://doi.org/10.1016/j.wasman.2018.04.015

Johansson, N., Velis, C., Corvellec, H. 2020. Towards clean material cycles: Is there a policy conflict between circular economy and non-toxic environment? Waste Manage. Res, 38(7), 705-707. https://doi.org/10.1177/0734242X20934251

Jurič, B., Hanžič, L., Ilić, R., Samec, N. 2006. Utilization of municipal solid waste bottom ash and recycled aggregate in concrete. Waste Manage. 26(12), 1436-1442. https://doi.org/10.1016/j.wasman.2005.10.016

Karagiannidis, A., Kontogianni, S., Logothetis, D. 2013. Classification and categorization of treatment methods for ash generated by municipal solid waste incineration: A case for the 2 greater metropolitan regions of Greece. Waste Manage. 33(2), 363-372. https://doi.org/10.1016/j.wasman.2012.10.023

Kirchherr, J., Reike, D., Hekkert, M. 2017. Conceptualizing the circular economy: An analysis of 114 definitions. Resour. Conserv. Recycl. 127, 221-232. https://doi.org/10.1016/j.resconrec.2017.09.005

Kleemann, R., Chenoweth, J., Clift, R., Morse, S., Pearce, P., Saroj, D. 2017. Comparison of phosphorus recovery from incinerated sewage sludge ash (ISSA) and pyrolysed sewage sludge char (PSSC). Waste Manage. 60, 201-210 https://doi.org/10.1016/j.wasman.2016.10.055

Kuo, W.-T., Liu, C.-C., Su, D.-S. 2013. Use of washed municipal solid waste incinerator bottom ash in pervious concrete. Cem. Concr. Compos. 37(1), 328-335. https://doi.org/10.1016/j.cemconcomp.2013.01.001

Lancellotti, I., Cannio, M., Bollino, F., Catauro, M., Barbieri, L., Leonelli, C. 2015. Geopolymers: An option for the valorization of incinerator bottom ash derived "end of waste". Ceram. Int. 41(2), 2116-2123. https://doi.org/10.1016/j.ceramint.2014.10.008

Lancellotti, I., Ponzoni, C., Barbieri, L., Leonelli, C. 2013. Alkali activation processes for incinerator residues management. Waste Manage. 33(8), 1740-1749. https://doi.org/10.1016/j.wasman.2013.04.013

Li, X., Liu, Z., Lv, Y., Cai, L., Jiang, D., Jiang, W., Jian, S. 2018. Utilization of municipal solid waste incineration bottom ash in autoclaved aerated concrete. Constr. Build. Mater.178, 175-182. https://doi.org/10.1016/j.conbuildmat.2018.05.147

Liu, Z.S., Li, W.K., Huang, C.Y. 2014. Synthesis of mesoporous silica materials from municipal solid waste incinerator bottom ash. Waste Manage. 34(5), 893-900. https://doi.org/10.1016/j.wasman.2014.02.016

Malinauskaite, J., Jouhara, H., Czajczyńska, D., Stanchev, P., Katsou, E., Rostkowski, P., Throne, R. J., Colón, J., Ponsá, S., Al-Mansour, F., Anguilano, L., Krzyżyńska, R., López, I. C., Vlasopoulos, A., Spencer, N. 2017. Municipal solid waste management and waste-to-energy in the context of a circular economy and energy recycling in Europe. Energy, 141, 2013-2044. https://doi.org/10.1016/j.energy.2017.11.128

Margallo, M., Aldaco, R., Irabien, Á. 2014. Environmental management of bottom ash from municipal solid waste incineration based on a life cycle assessment approach. Clean Technol. Environ. Policy. 16(7), 1319-1328. https://doi.org/10.1007/s10098-014-0761-4

Margallo, M., Taddei, M. B. M., Hernandez-Pellon, A., Aldaco, A., Irabien, A. 2015. Environmental sustainability assessment of the management of municipal solid waste incineration residues: a review of the current situation. Clean Technol. Environ. Policy. 17(5), 1333-1353. https://doi.org/10.1007/s10098-015-0961-6

Matsumoto, H., Takaoka, M. 2022. The application of multiple advanced chloride removal methods to synthesized Friedel's salt and municipal solid waste incineration bottom ash. Waste Manage. 141, 27-34. https://doi.org/10.1016/j.wasman.2022.01.029

Mihai, F. C., Apostol, L. 2012. Disparities in municipal waste management across EU-27. A Geographical Approach. Present Environment and Sustainable Development, 6(1), 169-180. https://ssrn.com/abstract=2548935

Moreno, M., De los Rios, C., Rowe, Z., Charnley, F. 2016. A Conceptual Framework for Circular Design. Sustainability, 8, 937-951. https://doi.org/10.3390/su8090937

Nixon, J. D., Wright, D. G., Dey, P. K., Ghosh, S. K., Davies, P. A. 2013. A comparative assessment of waste incinerators in the UK. Waste Manage. 33(11), 2234-2244. https://doi.org/10.1016/j.wasman.2013.08.001

Olsson, S., Kärrman, E., Gustafsson, J. P. 2006. Environmental systems analysis of the use of bottom ash from incineration of municipal waste for road construction. Resour. Conserv. Recycl. 48(1), 26-40. https://doi.org/10.1016/j.resconrec.2005.11.004

Recycling Magazine. 20.09.2022. Scandinavian tax relief on waste incineration will reduce landfill in Europe. Retreived from https://www.recycling-magazine.com/2022/09/20/scandinavian-tax-relief-on-waste-incineration-will-reduce-landfill-in-europe/

Saffarzadeh, A., Arumugam, N., Shimaoka, T. 2016. Aluminum and aluminum alloys in municipal solid waste incineration (MSWI) bottom ash: A potential source for the production of hydrogen gas. Int. J. Hydrogen Energy, 41(2), 820-831. https://doi.org/10.1016/j.ijhydene.2015.11.059

Schabbach, L.M., Andreola, F., Barbieri, L., Lancellotti, I., Karamanova, E., Ranguelov, B., Karamanov, A. 2012. Post-treated incinerator bottom ash as alternative raw material for ceramic manufacturing. J. Eur. Ceram. Soc. 32(11), 2843-2852. https://doi.org/10.1016/j.jeurceramsoc.2012.01.020

Schneider, D. R., Ragossnig, A. M. 2015. Recycling and incineration, contradiction or coexistence? Waste Manage. Res. 33(8), 693-695. https://doi.org/10.1177/0734242X15593421

Song, Y.M., Li, B.L., Yang, E.H., Liu, Y.Q., Ding, T. 2015. Feasibility study on utilization of municipal solid waste incineration bottom ash as aerating agent for the production of autoclaved aerated concrete. Cem. Concr. Compos. 56, 51-58. https://doi.org/10.1016/j.cemconcomp.2014.11.006

Svingstedt, A., Corvellec, H. 2018. When lock-ins impede value co-creation in service. Int. J. Qual. Serv. Sci. 10(1), 2-15. https://doi.org/10.1108/IJQSS-10-2016-0072

Šyc, M., Simon, F.G., Hykš, J. Braga, R., Biganzoli, L., Costa, G., Funari, V., Grosso, M. 2020. Metal recovery from incineration bottom ash: State-of-the-art and recent developments. J. Hazard. Mater. 393, 122433. https://doi.org/10.1016/j.jhazmat.2020.122433

UN General Assembly. 2015. Transforming our world: the 2030 Agenda for Sustainable Development, 21 October 2015, A/RES/70/1. Retrieved from: https://www.refworld.org/docid/57b6e3e44.html

Van der Sloot, H. A., Kosson, D. S., Hjelmar, O. 2001. Characteristics, treatment and utilization of residues from municipal waste incineration. Waste Manage. 21(8), 753-765. https://doi.org/10.1016/S0956-053X(01)00009-5

Van Gerven, T., Geyson, D., Stoffels, L., Jaspers, M., Wauters, G., Vandescasteele, C. 2005. Management of incinerator residues in Flanders (Belgium) and in neighbouring countries. A comparison. Waste Manage. 25, 75-87. https://doi.org/10.1016/j.wasman.2004.09.002

Van Zomeren, A., Velzeboer, I. 2017. End of Waste criteria for inert aggregates in Member States. ECN: Petten, The Netherlands, 43

Verbinnen, B., Billen, P., Van Canegham, J., Vandecasteele, C. 2017. Recycling of MSWI Bottom Ash: A Review of Chemical Barriers, Engineering Applications and Treatment Technologies. Waste Biomass Valor. 8, 1453-1466. https://doi.org/10.1007/s12649-016-9704-0

Villanueva, A., Munck-Kampmann, B., Fischer, C., Watson, D., Jacobsen, H., Bahn Kristensen, K., Vrgoc, M., Skovgaard, M., Carlsen, R. 2006. Potential economic and environmental effects in Denmark of potential changes to 'end-of-waste' definitions. Danish Environmental Protection Agency

Vountatsos, P., Atsonios, K., Itskos, G., Agraniotis, M., Grammelis, P., Kakaras, E. 2016. Classification of Refuse Derived Fuel (RDF) and Model Development of a Novel Thermal Utilization Concept through Air-Gasification. Waste Biomass Valor. 7, 1297-1308. https://doi.org/10.1007/s12649-016-9520-6

Waste and Resources Action Programme [WRAP]. 2006. The sustainable use of resources for the production of aggregates in England. The Waste and Resources Action Programme.

Wysokińska, Z. 2016. The "New" Environmental Policy of the European Union: A Path to Development of a Circular Economy and Mitigation of the Negative Effects of Climate Change. Comparative Economic Research: Central and Eastern Europe, 19(2), 57 https://doi.org/10.1515/cer-2016-0013

Yao, J., Li, W.-B., Tang, M., Fang, C.-R., Feng, H.-J., Shen, D.-S. 2010. Effect of weathering treatment on the fractionation and leaching behaviour of copper in municipal solid waste incinerator bottom ash. Chemosphere, 81(5), 571-576. https://doi.org/10.1016/j.chemosphere.2010.08.038

Zero Waste Europe [ZWE]. 2022. The EP approves the inclusion of municipal incinerators in ETS as of 2026. Retreived August 22, 2022 from https://zerowasteeurope.eu/press-release/the-ep-approves-the-inclusion-of-municipal-incinerators-in-ets-as-of-2026-2/

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues
CISA

# ANALYSIS OF SECONDARY WASTE FROM A PLASTICS RECYCLING PLANT FOR THE PRODUCTION OF CARBOXYLIC ACIDS

Kyra Atessa Vogt *, Leonie Wenzel and Iris Steinberg

*Department of Civil and Environmental Engineering, University of Applied Sciences Darmstadt, Darmstadt, Germany*

**Article Info:**
Received:
27 February 2023
Revised:
25 May 2023
Accepted:
22 June 2023
Available online:
30 June 2023

**Keywords:**
Circular bioeconomy
Chain elongation
Carboxylic acids
Secondary waste
Plastic recycling plant

## ABSTRACT

The production of bio-based platform chemicals through the chain elongation of short-chain carboxylic acids to medium-chain carboxylic acids by ethanol-acetat fermentation can be a contribution to the circular economy. To avoid further waste, secondary waste that already contains short-chain carboxylic acids can be used. The potential for the production of bio-based carboxylic acids from a secondary waste of a recycling plant for plastic waste is examined in this paper. Therefore, practical experiments with the process water of a recycling plant for plastic waste were conducted in order to assess the potential for carboxylic acids production. At the end of the experiment, the concentrations achieved by chain elongation in the secondary waste result in 496 mg/L butyric acid and 87 mg/L caproic acid and the concentration in the extraction solvent is 933 mg/L caproic acid. To conclude, chain elongation of carboxylic acids in secondary waste, in this case the process water from a treatment plant for plastic waste, is generally possible. In order to estimate the total potential for the production, the fluctuations of the quality of the process water have to be considered.

## 1. INTRODUCTION

Primary fossil raw materials are limited and their demand is steadily increasing (BMBF & BMEL, 2020). According to the Organisation for Economic Co-operation and Development, the use of primary raw materials such as biomass, fossil fuels, metals and non-metallic minerals will double by 2060 (European Union, 2020, p. 4; OECD, 2019, p. 19).

The bioeconomy is defined in the national bioeconomy strategy of the Federal Government of Germany as “the production, development and use of biological resources, processes and systems to provide products, processes and services in all economic sectors within the framework of a sustainable economic system” (BMBF & BMEL, 2020, p. 10). The national bioeconomy strategy follows the Sustainable Development Goals of the United Nations and aims to ensure for example sufficient food, health, economic growth, sustainable consumption and production as well as humane work by 2030. For this purpose, raw materials from agriculture, forestry and marine management as well as biogenic residues and waste materials are to be used (BMBF & BMEL, 2020).

A transition from a linear economy to a circular economy is also required. As part of the European Green Deal, the New Circular Economy Action Plan was published in March 2020. It introduces new initiatives that consider the entire life cycle of products, modernise and transform the European economy and protect the environment at the same time. The aim of the New Circular Economy Action Plan is to take action to prevent and reduce waste (European Union, 2020).

A combination of circular economy and bioeconomy is described as circular bioeconomy. Stegmann et al. define it as follows: “The circular bioeconomy focuses on the sustainable, resource-efficient valorization of biomass in integrated, multi-output production chains (e.g. biorefineries) while also making use of residues and wastes and optimizing the value of biomass over time via cascading.” (Stegmann et al., 2020, p. 5). Therefore, the development and investigation of existing secondary waste in relation to circular bioeconomy is important.

To contribute to circular bioeconomy, an experimental approach is utilized to produce bio-based chemicals from secondary waste. Secondary waste is waste that originates from a waste treatment process, e.g. residues from recovery and disposal processes (European Union, 2015). These include organic compounds with one or more carboxy groups (-COOH) (Federle et al., 2017) which are classified depending on the number of carbon atoms into short-chain (1-3 carbon atoms), medium-chain (4-10 carbon atoms) and long-chain (>10 carbon atoms) carboxylic acids (Hopp, 2018). These are conventionally generated by chemical synthesis from petroleum-based resources or by

* Corresponding author:
Kyra Atessa Vogt
email: kyra-atessa.vogt@h-da.de

Detritus / Volume 23 - 2023 / pages 58-64
https://doi.org/10.31025/2611-4135/2023.18277

the synthesis of natural oils (e.g. coconut or palm kernel oil) (Anneken et al., 2012).

Short-chain carboxylic acids (SCCA) in a secondary waste are converted into medium-chain carboxylic acids (MCCA) during a biotechnological treatment process with the help of specialised microorganisms and ethanol as a nutrient. This biotechnological treatment process is described as acetate-ethanol fermentation (Angenent et al., 2016). The separation of these carboxylic acids takes place during the treatment process as an in-situ extraction (liquid-liquid extraction) using a non-polar extraction solvent. These carboxylic acids can be used for the production of bio-based materials and thereby have a positive impact on the circular bio-based economy (bioeconomy). For example, precursors for lubricants and tensides can be produced (Sarria et al., 2017; Wang & Yin, 2022, p. 1). This type of waste management corresponds to material recycling according to article 4 number 1 c) of the European Waste Framework Directive (Directive 2008/98/EC).

In order to preserve primary fossil raw materials, it is essential to identify further secondary waste that can be used for the production of medium-chain carboxylic acids. A large number of different secondary waste types have already been analysed regarding the potential for the production of bio-based carboxylic acids. These include, for example, sewage sludge and municipal solid waste leachate (Wang & Yin, 2022). But also organic residues can be used, such as ethanol-containing secondary waste (yeast of beer production, wine fermentation residue (Groof et al., 2019), liqueur production (Wang & Yin, 2022), secondary waste from quark, yoghurt and cheese industry (Groof et al., 2019), swine manure or fermented sugar cane (Wang & Yin, 2022)).

In this study, the potential for a cascading utilisation of a secondary waste from a treatment plant for plastic waste is further considered, as the literature research revealed that such a secondary waste has not yet been analysed.

## 2. MATERIALS AND METHODS

### 2.1 Materials

For the experiments, secondary waste of a recycling plant for plastic films is considered. Here, the process water which is produced during the necessary washing process of the plastic films in the recycling process is used. After being treated, the process water is internally recirculated. The secondary waste already contains short-chain carboxylic acids and was used in the experiments without any addition of inoculum. A mixed bacteria culture is expected in the secondary waste, which is able to carry out ethanol-acetate fermentation. There were no detailed analyses conducted to characterise the bacteria cultures.

For the recovery of the produced medium-chain carboxylic acids a liquid-liquid extraction is applied with rapeseed methyl ester (SysKem Chemie GmbH; fatty acid methyl ester C16 - C18) as a solvent. Previous experiments at the University of Applied Sciences Darmstadt indicated that this solvent is suitable in combination with the biological process. Thus, it is added to the secondary waste in a volume ratio of 1:10.

Ethanol is also required as a nutrient for chain elongation (Carl Roth; ethanol 96% denatured) and is added into the secondary waste in a concentration of 9 g/L, since according to Sarkar et al. (2021, p. 8) the highest caproic acid production during chain elongation resulted from this concentration.

### 2.2 Experimental setup

#### *2.2.1 Reactor setup*

For the experiments, a stirred batch reactor with an integrated liquid-liquid extraction is used. The maximum capacity of the reactor is approximately 16 litres, with a final operating capacity of 14 litres. The reactor is airtight to allow an anaerobic milieu. The oxygen present in the headspace is displaced with inert gas at the beginning of the experiments. The stirrer motor (Brushless DC Motor, BPC Instruments AB) rotates the stirrer which consists of a combination of a diagonal flat blade stirrer and an anchor stirrer (Hemming & Wagner, 2017). The average rotation speed is 70°RPM.

There are two sampling devices for taking a composite sample of the secondary waste. Due to the integrated liquid-liquid extraction, the non-polar extraction solvent settles on the polar secondary waste. Since sampling takes place regularly, the filling level of the reactor is changing and thus also the sampling point of the extraction solvent. Therefore, a sampling device is required that can take a sample from the extraction solvent at any time. For this purpose, a floater is connected to a sampling device via a flexible tube. During each sampling, the volume that remains in the sampling device is to be discarded. For the regulation of the process temperature during the experiments the reactor setup is placed in a temperature-controlled room. Figure 1 illustrates the described experimental setup and the technical implementation.

#### *2.2.2 Operating parameters*

Four reactors with contents listed in Table 1 were used for the experiments. A double determination of the chain elongation with integrated liquid-liquid extraction (CE-LLE), one reactor without chain elongation but with liquid-liquid extraction (LLE) and one blind reactor with only secondary waste were used.

The experiment period is four weeks (28 days) at an operating temperature of 37°C ± 1. The pH-value of the secondary waste should range between 6.0 and 6.5 in order to inhibit the possible formation of methane. The pH-value is adjusted to 6.0 with hydrochloric acid (c = 2.87 mol/L) as required at the beginning of the experiments.

**TABLE 1:** Overview of composition in reactor (CE = chain elongation, LLE = liquid-liquid extraction).

| reactor | secondary waste | extraction solvent | ethanol |
|---|---|---|---|
| CE + LLE 1 | 12 L | 1.2 L | 9 g/L |
| CE + LLE 2 | 12 L | 1.2 L | 9 g/L |
| LLE | 12 L | 1.2 L | - |
| Blind | 12 L | - | - |

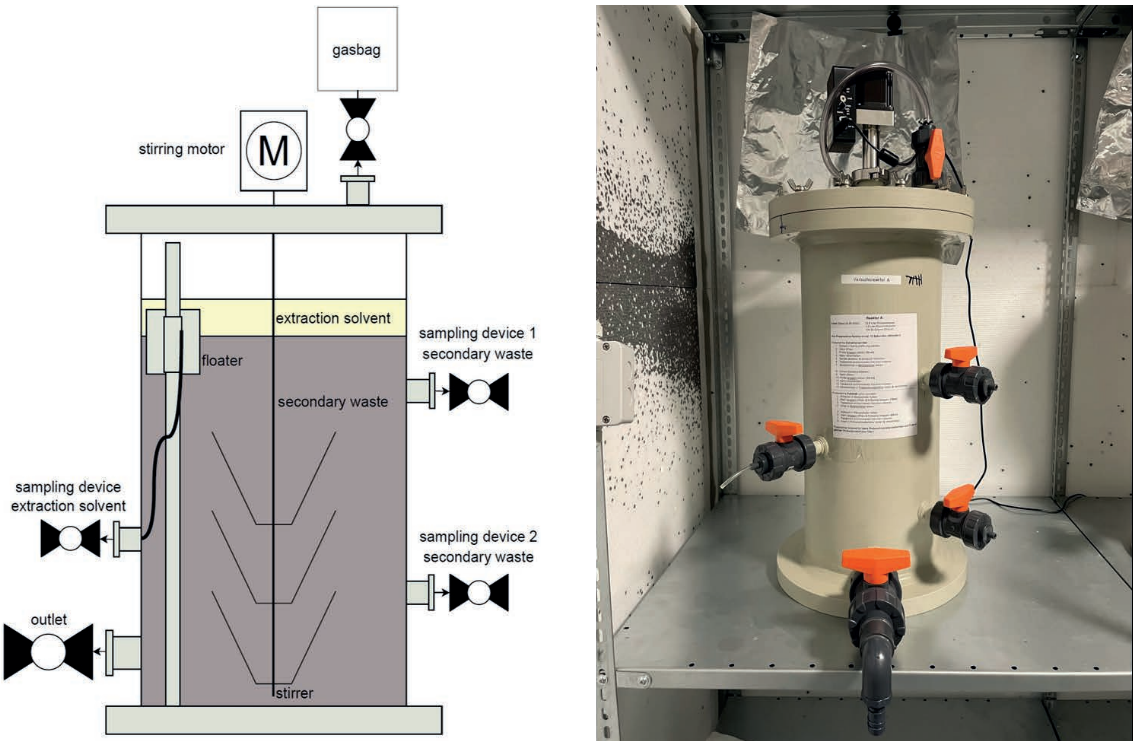


**FIGURE 1:** Schematic drawing of reactor setup (left) and technical implementation (right).

*2.2.3 Technical sampling of secondary waste and extraction solvent*

Sampling takes place twice a week. First, the stirrer is switched off one hour before sampling to ensure that the extraction solvent settles completely on the surface of the secondary waste. Firstly, 12 ml of extraction solvent is discarded, afterwards 10 ml of the extraction solvent is taken for each sample. This is followed by the sampling of the secondary waste. At first, sampling device 1 is sampled. 15 ml is discarded, and 20 ml secondary waste sample is taken. The procedure is repeated for sampling device 2. A composite sample is generated from the two secondary waste samples.

## 2.3 Analytical methods

The on-site analyses include all parameters taken immediately after sampling. These include the pH-value, the redox potential and the conductivity in the secondary waste sample.

The pH-value indicates whether the ideal conditions for the microorganisms of chain elongation are present in the secondary waste.

The redox potential indicates whether there is an aerobic or anaerobic milieu in the secondary waste. If the value of the redox potential is below minus 330 mV (Wiese & König, 2007), the milieu is to be regarded as anaerobic. This is a necessary condition for microorganisms responsible for the biological chain elongation.

After the on-site analysis, the samples of secondary waste are stored at approximately minus 20°C until further analysis. The extraction solvent samples are stored at room temperature until further analysis.

To characterise the secondary waste in more detail, the samples are defrosted. The chemical oxygen demand (COD) is determined photometrically by using cuvette tests (LCK 514, 100 - 2,000 mg/L $O_2$ and LCK 014 1,000-10,000 mg/L $O_2$, Hach Lange GmbH). In order to monitor the degradation of the additive ethanol, its concentration in the secondary waste of the CE-LLE reactors is also determined photometrically by using cuvette tests (LCK 300, 0.01-0.12 g/L, Hach Lange GmbH).

The carboxylic acids in the secondary waste as well as in the extraction solvent are analysed by gas chromatography with flame ionisation detection (GC-FID). The gas chromatograph (Shimadzu GC2025) is equipped with a Thermo Scientific TG-WAXMS A capillary column (length: 30 m; inner diameter: 0.32 mm; film thickness 0.5 µm). Helium was used as carrier gas with a flow rate at 29.1 ml/min. For the samples of secondary waste 1 µL was injected into a split injector with a split ratio of 1:10. The following column oven programme was used for the samples of the secondary waste: heating up to 80°C for 2 minutes and continuing at a rate of 20°C per minute to 235°C for 5 minutes. For the samples of the extraction solvent 0.5 µL was injected into a split injector with a split ratio of 1:10. The following column oven programme was used for the samples of the extraction solvent: heating up to 80°C for 2 minutes, continuing at a rate of 10°C per minute to 180°C and with a rate of 5°C per minute to 235°C for 10 minutes. The samples of the secondary waste as well as the samples of the extraction solvent need to be prepared for the measurements. For this purpose, the samples of the secondary waste are acidified and diluted 1:10 and 1:100 with an acetone-water mixture (ratio 1:1). It is also necessary to filter the samples with a PTFE syringe filter (pore size 0.45 µm) before the measurements. Whereas the samples of the extraction solvent need to be filtered with a PTFE syringe filter (pore size 0.45 µm) before the 1:10 dilution with n-hexane.

Furthermore, the composition of the collected gas in the gas bags are analysed by means of a biogas analyser (BM 5000, Geotech).

#### *2.3.1 Analysis*

For the analysis, only the carboxylic acids in the reactors with chain elongation and integrated liquid-liquid extraction (CE-LLE) are considered. The reactor without chain elongation but with liquid-liquid extraction (LLE) and the reactor without chain elongation and without liquid-liquid extraction (blind) are used as a monitor for the chain elongation by ethanol-acetate fermentation. The concentrations of carboxylic acids in the CE-LLE reactors are given as averaged absolute amounts. Therefore, the total amount of generated carboxylic acids is determined and a ratio of the amount of generated carboxylic acids in the secondary waste and in the extraction solvent is obtained.

## 3. RESULTS AND DISCUSSION

### 3.1 On-site analysis of secondary waste

During the experimental period, the temperature of the secondary waste in the reactors was measured continuously at 36.6°C ± 0.1.

The pH-value of the untreated secondary waste was pH 6.9. This was adjusted to pH 5.9 before the start of the experiments with hydrochloric acid. During the experiment the pH-value of the CE-LLE reactors decreased to pH 5.1. This may be caused by forming of carboxylic acids in the secondary waste. In the reference reactors, the pH-value increased to pH 6.6 for LLE and to pH 6.8 for blind. This may be responsible for the biogas formation in these reactors at the end of the experiment.

The composition of the collected gas took place at the end of the experiment period. The reference reactors produced about 3 L (LLE) and about 8 L (blind) over the entire experiment period. The LLE reactor produced 30 vol% of methane and 15 vol% of carbon dioxide. The blind reactor produced 50 vol% of methane and 20 vol% of carbon dioxide. The CE-LLE reactors produced about 1 L gas, which consisted mainly of the inert gas nitrogen at 85 vol%. It is assumed that this is due to the fact that the utilised bioreactor has a headspace volume of approximately three litres. At the beginning of the experiment, the headspace is filled with the inter gas nitrogen to ensure the necessary anaerobic conditions for the treatment process. If the biological process produces gas, at first the inert gas is led into the connected gas bag and is analysed.

There is no clear tendency in the redox potential determination. The analyses of the CE-LLE and LLE reactors do not indicate an anaerobic environment over the entire experiment period. In the blind reactor, predominantly anaerobic redox potentials can be recognised over the entire experiment period. The unsteady measurements of the redox potentials may be caused by the high salt content in the secondary waste. These are on average 22 mS/cm for the CE-LLE reactors and 23 mS/cm for the LLE and blind reactors.

### 3.2 Characterisation of secondary waste

At the start of the experiments, the chemical oxygen demand (COD) of the untreated secondary waste was 2.98 g/L $O_2$. With the addition of ethanol, the COD increased by a factor of five and reached on average 16.89 g/L $O_2$ in the CE-LLE reactors. The LLE and the blind reactor did not show higher values of COD (3.20 g/L $O_2$ and 3.01 g/L $O_2$) compared to the secondary waste at the start of the experiments. In the course of the four-week experiments, the COD remained almost constant. A slightly increased COD of the CE-LLE reactors to 17.21 g/L $O_2$ can be recognised. The COD of the LLE and the blind reactor decreased to 3.13 g/L $O_2$ and 2.29 g/L $O_2$ at the end of the experiments.

At the beginning of the experiments, 9.00 g/L ethanol was added to the CE-LLE reactors. Over the treatment period of 28 days, a degradation of the ethanol can be recognised. After the first three days of the experiment, an average of 7.00 g/L ethanol can be measured in the secondary waste. A continuous degradation of ethanol to 4.85 g/L at the end of the test period can be determined. This means that about 50 percent of the added ethanol was degraded. Due to the low production of gas in the reactors with chain elongation and liquid-liquid extraction, it can be presumed that the degraded ethanol contributed to the formation of the medium-chain carboxylic acids. The reduction of the ethanol during the experimental period is illustrated in Figure 2.

### 3.3 Potential for carboxylic acid formation

The carboxylic acids in the secondary waste and in the extraction solvent were analysed using GC-FID. In order to assess, if the secondary waste is suitable for forming carboxylic acid, the concentrations of acids were obtained and then allocated against the corresponding quantities to be able to compare the results. Therefore, the results refer to the 12 litres of secondary waste. Figure 3 presents the total amount of butyric acid (C4; in orange) and caproic acid (C6; in blue) summarized for secondary waste and extraction solvent over the experiment period in milligrams. Butyric and caproic acid were chosen as they showed the highest potential within the experiment.

The results show that there is no change in the amount of butyric and caproic acid until the tenth day of the experiment. Nevertheless, it must be mentioned that these results are only slightly above the lower limit of determination of 5 mg/L. An increase of butyric acid to 5,514 mg and caproic acid to 2,583 mg is noticeable by the tenth day of the experiment. Until the end of the experiment, these continue to rise to a maximum of 7,735 mg butyric acid and 3,786 mg caproic acid.

These results show in general, that forming butyric acid and caproic acid from secondary waste is possible. Furthermore, the potential resulting from the chain elongation of the carboxylic acids can be compared. Therefore the generated carboxylic acids in the secondary waste and in the extraction solvent (CE-LLE) were compared to those in the reference reactors (blind and LLE). An increase of

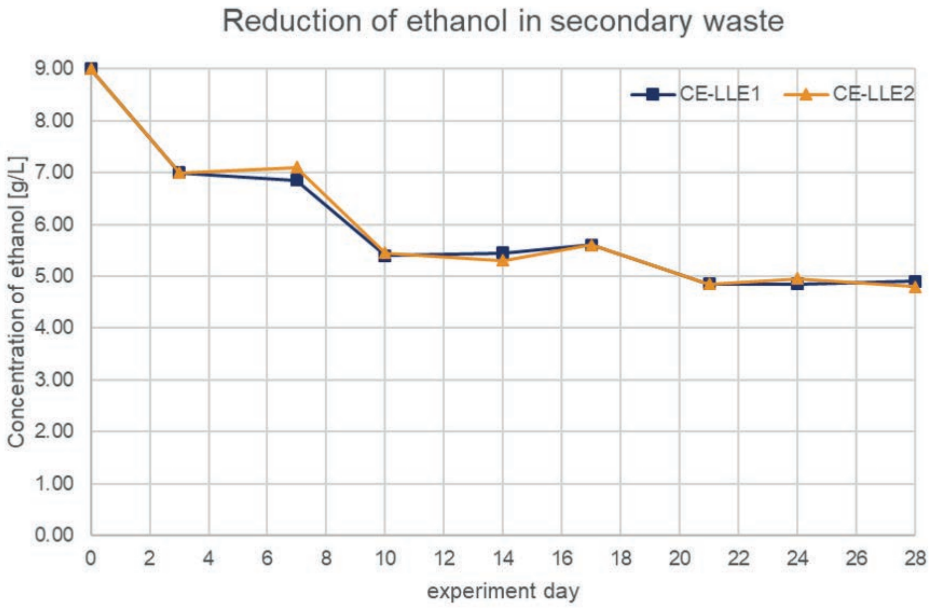


**FIGURE 2:** Reduction of ethanol in secondary waste in reactors with chain elongation and liquid-liquid extraction (CE-LLE).

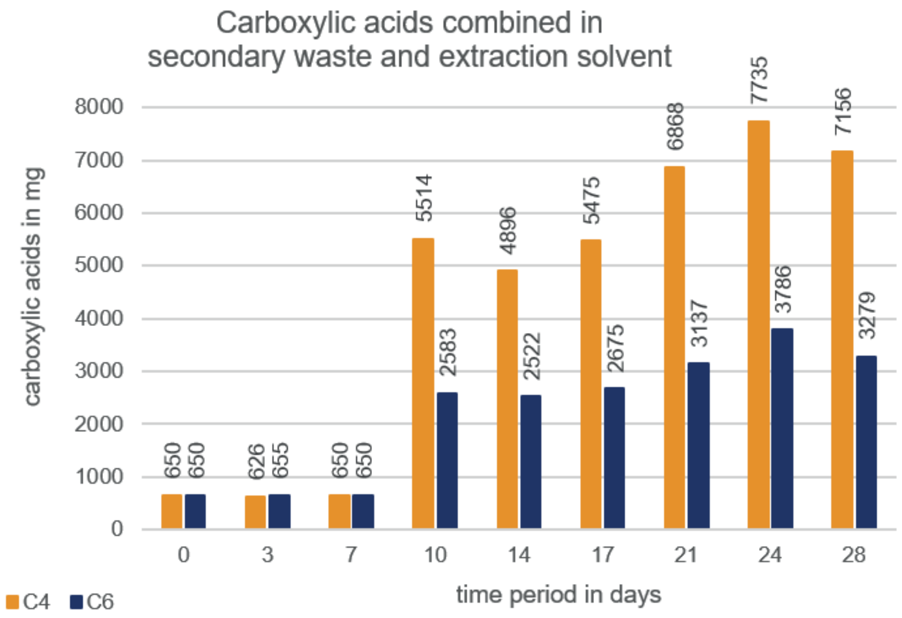


**FIGURE 3:** Total amount of butyric acid and caproic acid over the experiment period summarized for secondary waste and extraction solvent.

carboxylic acids in relation to the reference reactors is indicated. In this context, 100 percent refers to the reference reactors.

Figure 4 illustrates the percentage deviation in secondary waste with chain elongation and without. Figure 5 shows the percentage deviation in the extraction solvent with and without chain elongation. As previously butyric and caproic acid were chosen for the comparison, based on the results of the experiments.

As already shown in Figure 3, there is no increase in butyric or caproic acid until experiment day ten. Figure 5 shows that on experimental day ten the butyric acid increased by 821% due to the chain elongation, which corresponds to 4,837 mg. In comparison to the reference reactor, caproic acid increased to 221% or 1,305 mg. During the experiments the deviation decreases. Due to the chain elongation, an increase of 512% or 5,846 mg more butyric acid can be detected at the end of the experiment. The deviation of caproic acid also decreases. At the end of the experiment, 90% or 1,026 mg additional caproic acid has been formed by the chain elongation.

Considering Figure 5, the delayed start is also noticeable. From day ten of the experiment, a deviation from the extraction solvent without chain elongation of 1,116% or 633 mg caproic acid can be seen. Only a small amount of butyric acid was transferred to the extraction solvent. On experiment day 24 an increase of 206% or 108 mg butyric acid was extracted by the chain elongation. By the end of the experiment, the deviation rises to 2,069% or 1,059 mg caproic acid.

These figures illustrate that through chain elongation of short-chain carboxylic acids, medium-chain carboxylic acids can be formed in secondary waste and subsequently be extracted. At the same time, the results indicate that more butyric acid accumulates in the secondary waste and more caproic acid in the extraction solvent. This may be explained by the decreasing polarity of the carboxylic acids with increasing chain length. As a result, caproic acid is less polar and the transition into the extraction solvent is higher.

## 4. CONCLUSIONS

For the production of bio-based platform chemicals in the form of carboxylic acids, the process water from a treatment plant for plastic waste was examined as a secondary waste stream.

The experiments showed that the formation of carboxylic acids started with a delay of ten days. Specifi-

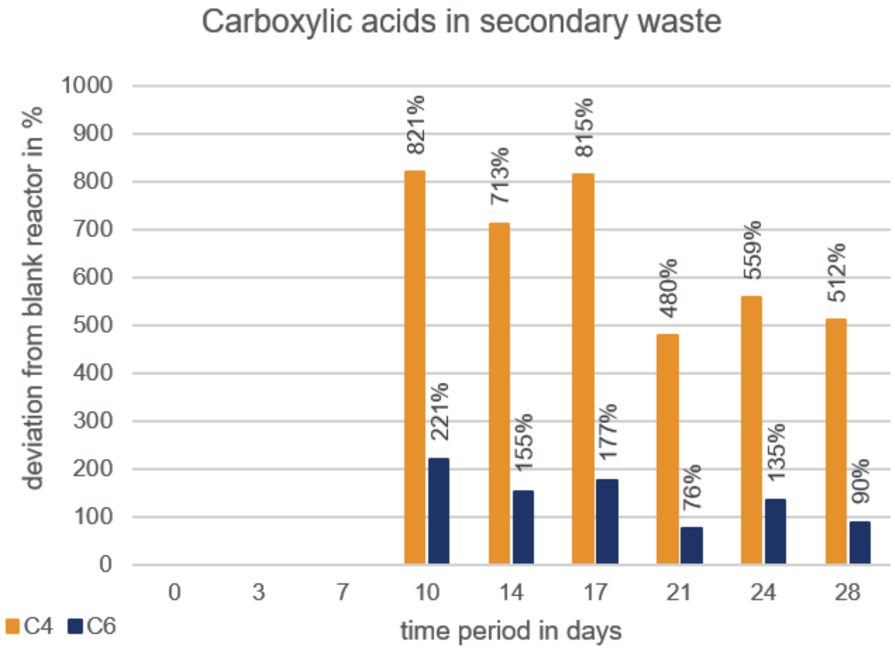


**FIGURE 4:** Percentage deviation of generated butyric acid and capronic acid in secondary waste with and without chain elongation.

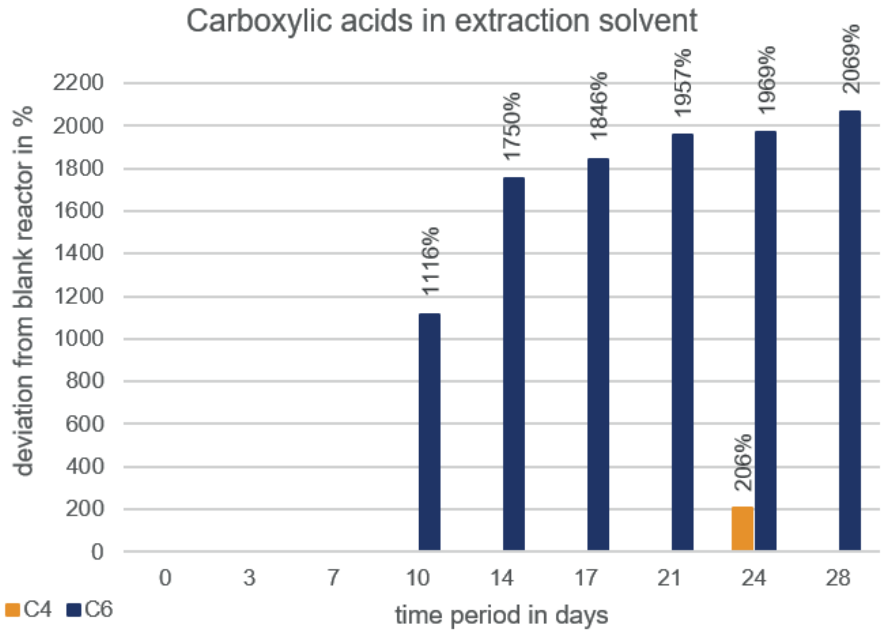


**FIGURE 5:** Percentage deviation of generated butyric acid and capronic acid in extraction solvent with and without chain elongation.

cally, the formation of butyric acid and caproic acid were considered. By the end of the experiment, 5,846 mg additional butyric acid were formed in the secondary waste compared to untreated secondary waste (reference reactor blind). On the other hand, 1,026 mg caproic acid was detected in the secondary waste at the end of the experiment. Additionally, 1,059 mg caproic acid was extracted. These amounts of carboxylic acids refer to the 12 litres of secondary waste treated in the experiment by consuming 3 kW of electrical energy and about 50 grams of ethanol. Therefore, the concentrations achieved by chain elongation in the secondary waste at the end of the experiment result in 496 mg/L butyric acid and 87 mg/L caproic acid and the concentration in the extraction solvent is 933 mg/L caproic acid.

To conclude, chain elongation of carboxylic acids in secondary waste, in this case the process water from a treatment plant for plastic waste, is generally possible. However, due to fluctuating concentrations of carboxylic acids in the original material in these experiments, the amounts of carboxylic acids detected are comparatively low and thus the potential of the examined secondary waste also is very low. Preliminary tests have shown higher concentrations. Therefore, in order to estimate the total potential for the production, the fluctuations of the quality of the process water have to be considered.

Furthermore, the experiments were realised without an inoculum. The use of already existing microorganisms in an inoculum could probably increase the potential.

## REFERENCES

Angenent, L. T., Richter, H., Buckel, W., Spirito, C. M., Steinbusch, K. J. J., Plugge, C. M., Strik, D. P. B. T. B., Grootscholten, T. I. M., Buisman, C. J. N., & Hamelers, H. V. M. (2016). Chain Elongation with Reactor Microbiomes: Open-Culture Biotechnology To Produce Biochemicals. Environmental Science & Technology, 50(6), 2796–2810. https://doi.org/10.1021/acs.est.5b04847

Anneken, D. J., Both, S., Christoph, R., Fieg, G., Steinberner, U., & Westfechtel, A. (Eds.). (2012). Fatty Acids: in Ullmann's Encyclopedia of Industrial Chemistry. Wiley-VCH Verlag GmbH & Co. KGaA. https://doi.org/10.1002/14356007.a10_245.pub2

BMBF, & BMEL. (2020). Nationale Bioökonomiestrategie. Berlin. Referat Nachhaltiges Wirtschaften, Bioökonomie (BMBF); Referat Bioökonomie, Stoffliche Biomassenutzung (BMEL).

European Union. (2015). Eurostat - Statistics Explained: Glossary "secondary waste" (ISSN: 2443-8219). https://ec.europa.eu/eurostat/statistics-explained/index.php?title=Glossary:Secondary_waste

European Union. (2020). Circular economy action plan: For a cleaner and more competitive Europe. Publications Office of the European Union. https://doi.org/10.2779/05068

Federle, S., Hergesell, S., & Schubert, S. (2017). Die Stoffklassen der organischen Chemie: Praktisch und kompakt von Studenten erklärt (1. Aufl. 2017). Springer Berlin Heidelberg.

Groof, V. de, Coma, M., Arnot, T., Leak, D. J., & Lanham, A. B. (2019). Medium Chain Carboxylic Acids from Complex Organic Feedstocks by Mixed Culture Fermentation. Molecules (Basel, Switzerland), 24(3). https://doi.org/10.3390/molecules24030398

Hemming, W., & Wagner, W. (2017). Verfahrenstechnik (12., korrigierte Auflage). Kamprath-Reihe. Vogel Media Business; Preselect.media GmbH.

Hopp, V. (2018). Chemische Kreisläufe in der Natur: Chemie - Biologie - Energetik (2. Aufl. 2018). Springer Berlin Heidelberg.

OECD. (2019). Global Material Resources Outlook to 2060: Economic Drivers and Evironmental Consequences. OECD. https://doi.org/10.1787/9789264307452-en

Sarkar, O., Rova, U., Christakopoulos, P., & Matsakas, L. (2021). Ethanol addition promotes elongation of short-chain fatty acids to medium-chain fatty acids using brewery spent grains as substrate. Journal of Environmental Chemical Engineering, 9(5), 105990. https://doi.org/10.1016/j.jece.2021.105990

Sarria, S., Kruyer, N. S., & Peralta-Yahya, P. (2017). Microbial synthesis of medium-chain chemicals from renewables. Nature Biotechnology, 35(12), 1158–1166. https://doi.org/10.1038/nbt.4022

Stegmann, P., Londo, M., & Junginger, M. (2020). The circular bioeconomy: Its elements and role in European bioeconomy clusters. Resources, Conservation & Recycling: X, 6, 100029. https://doi.org/10.1016/j.rcrx.2019.100029

Wang, J., & Yin, Y. (2022). Biological production of medium-chain carboxylates through chain elongation: An overview. Biotechnology Advances, 55, 107882. https://doi.org/10.1016/j.biotechadv.2021.107882

Wiese, J., & König, R. (2007). Praxisbericht Laboranlytik und Prozess-Messtechnik: Prozessbegleitende Fermenterüberwachung auf Biogasanlagen (DOC040.72.00447.May07). www.hach.com

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# WASTE-TO-HYDROGEN: CHALLENGES AND OPPORTUNITIES IN THE UK SCENARIO

Massimiliano Materazzi *, Suviti Chari, Alex Sebastiani and Shazeb Bajwa

*Department of Chemical Engineering, University College London, Torrington Place, London WC1E 7JE, United Kingdom*

**Article Info:**
Received:
7 March 2023
Revised:
5 June 2023
Accepted:
15 June 2023
Available online:
30 June 2023

**Keywords:**
Low carbon hydrogen
Waste gasification
BECCS
LCA

## ABSTRACT

Hydrogen is widely recognised to play a key role to decarbonise various industries, as well as transportation, heating and power sectors, for it does not generate greenhouse gas emissions at the point of use. Understanding the technologies that can generate low carbon hydrogen is essential in planning the development of future gas networks and more sustainable manufacturing processes. One promising approach is hydrogen production by gasification of waste or biomass. This paper summarises work undertaken to design a commercial Waste-to-Hydrogen (WtH$_2$) plant, which includes an assessment of current development stage of technologies, the identification of an appropriate scale for the plants, and development of specifications for process design and output streams. The overall production levels of hydrogen product is observed to be limited by the availability of sustainable feedstocks; however, the results of negative CO$_2$ emissions achieved via biohydrogen production shows that its overall potential to reduce GHG emissions is significantly better, as compared to other form or low carbon hydrogen. In particular, biohydrogen application is capable of generating negative emissions that are required to offset GHGs from other sectors in the future. In combination, low carbon hydrogen production pathways can make a very important contribution to achieving net zero commitment in UK.

## 1. INTRODUCTION

In recent years hydrogen has received increasing attention as a potential fuel that could be produced from non-fossil fossil sources, can be generated with low greenhouse-gas (GHG) emissions, and generates no emissions at the point of use. As such, hydrogen is believed to play a key role in achieving Net Zero ambitions, across Europe, and UK in particular (Gov.uk, 2019).

The UK has set a target to reach Net Zero GHG emissions by 2050. This requires the current 522 Mt CO$_2$-eq emissions per year to reduce to zero over the next three decades. Carbon emission reductions can partly be achieved through increased renewable electricity generation and electrification; however, the adoption of low or zero carbon fuels, such as hydrogen, are expected to play a significant role. Hydrogen is currently used as an industrial feedstock, mainly for ammonia production and in oil refineries (Chapman et al., 2019). It is mostly produced from fossil fuels reformation, namely steam methane reformation (SMR) and autothermal reforming (ATR) of natural gas, also known as ‘grey hydrogen’. There are three main technologies that can produce hydrogen with low carbon impact:

- Electrolysis using renewable electricity to produce ‘green hydrogen’
- ATR and SMR with carbon capture and storage (CCS) to produce ‘blue hydrogen’
- Reformation of biogas or gasification of biomass with CCS to produce ‘biohydrogen’.

Green hydrogen offers a small-scale solution that can be cost effective for some applications such as filling stations for hydrogen vehicles. However, currently the cost of hydrogen produced by electrolysis is far more expensive than SMR grey hydrogen (£6.20/kg versus £1.90/kg for transport-grade) and it does not offer significant greenhouse gas (GHG) benefits unless renewable electricity is used (Al-Qahtani et al., 2021). Power-to-gas (PtG) technologies rely on this principle. This development is particularly attractive due to the availability of renewable power generation in excess of immediate electricity demand and an expectation that this availability will increase with the share of intermittent renewable power generation (Götz et al., 2016).

Steam and autothermal methane reforming involves reacting natural gas with steam or limited amount of oxygen, at high temperatures over a catalyst to produce syngas (a

---

* Corresponding author:
Massimiliano Materazzi
email: massimiliano.materazzi.09@ucl.ac.uk

Detritus / Volume 23 - 2023 / pages 65-75
https://doi.org/10.31025/2611-4135/2023.18274

mixture of $H_2$ and CO). This is then further processed to maximise $H_2$ generation (via water gas shift reaction, WGS) and separate $H_2$ product from a $CO_2$-rich stream. Production capacities of hydrogen from a typical steam methane reforming plant range between 150 and 440 MW with an energy efficiency of typically 70% (Al-Qahtani et al., 2021). If SMR is to become a major low-carbon source of hydrogen, carbon capture and storage is essential. It is estimated that between 71% and 92% of the $CO_2$ in steam methane reforming can be captured; however higher capture rates will be needed if the process is to be used in the long term (Materazzi et al., 2019). Furthermore, CCS barriers are no exclusively technical, with CCS cost being the most significant hurdle in the short to medium term. Auto-thermal reformation (ATR) is a similar process to steam methane reformation (SMR), which is a proven technology used to produce the majority of 'grey' hydrogen in the world today. Rather than combusting natural gas to raise steam, and produce carbon dioxide, as with the SMR process, the ATR process utilises natural gas within the reactors, with carbon dioxide produced from the feedstock at high pressures and relatively high purity. This provides a single stream for carbon dioxide separation and removal rather than multiple carbon dioxide streams at variable pressures and purities from the SMR process, allowing for a higher carbon dioxide capture rate (>95%) than SMR. As well as providing a low-cost way to capture carbon dioxide, ATR also shares the same benefits of SMR, namely that the technology is based on well-proven chemical engineering technology, which can be easily scaled-up and produces hydrogen at a relatively low cost (dependent on the natural gas price). The carbon dioxide stream captured from the ATR plant can be injected into offshore carbon storage facilities, as is being planned around several industrial clusters in the UK, including the HyNet cluster in the North West of England and the Acorn project in North East of Scotland (Edwards et al., 2021).

Biohydrogen is the newest addition to the low-carbon hydrogen choices. Several techniques have been proposed by many researchers for the thermal conversion of solid organic materials to biohydrogen, via gasification or pyrolysis (Barisano et al., 2017). Biohydrogen offers the prospect of low carbon hydrogen production from low-grade – in large fraction renewable – fuels, at parity with the cost of natural gas, and with the potential of negative carbon emissions if the separated $CO_2$ is sequestered. A number of studies have been reported in the literature for biohydrogen production from first-generation biomasses, especially from starchy and sugar-rich biomasses due to easy fermentability attribute of these feedstocks by anaerobic organisms which increases $H_2$ yield compared to other organic substrates (Chong et al., 2009). Waste and second-generation biomass materials, although readily available and abundant, have limited uses in terms of chemical feedstocks, due to the need for pre-treatment and presence of many contaminants which add complexity and costs. Thermochemical treatment of waste fuels for hydrogen or chemical production, therefore, presents a number of unique issues demanding specific design and feedstock choices and technical solutions. Generally speaking, the conversion schemes use heat and various combinations of steam, oxygen and $CO_2$, to convert the feedstock to various amounts of char, hydrocarbon gases, hydrogen, and carbon oxides, with ash being a by-product of most waste feedstocks. Ash residues are usually classified as a hazardous waste on account of their high alkalinity and other pollutant species (e.g. heavy metals and soluble chloride and sulphate salts); as such, they require specific treatment before disposal. Therefore, before hydrogen from waste can be deployed commercially several barriers must be overcome. Firstly, the technical feasibility of hydrogen production from waste derived feedstock must be demonstrated to show that the concept is credible and sufficiently robust. Secondly, the process must be optimised for commercial deployment, with designs produced, environmental impact understood and costs modelled. Finally, the chosen designs must be deployed at larger scale, with hydrogen supplied to end users. Extensive work is needed to push forward commercial deployment of hydrogen production from waste by systematically working to address each barrier. Most of the work present in the literature focuses on single aspects, and in most cases on biohydrogen processes utilizing pure biomass as a feedstock (Antonini et al. 2021; Barisano et al. 2017) This work aims at addressing at least some of these barriers in a more systematic and unified form. First, main technical challenges and latest developemnts of biohydrogen plants are appraised. Different low carbon hydrogen production routes are then compared from a Life Cycle assessment (LCA) perspective, to understand deployment and integration opportunities in UK over the next 30 years.

The hydrogen production methods selected were electrolysis (on-site, large scale off-site and off-shore), methane reformation (ATR and SMR) with carbon capture and storage (CCS) and waste (MSW and waste wood) gasification with CCS. Fossil fuel, renewable feedstocks and energy sources, were flexed in the analysis, an example being replacing waste with pure biomass or plastics for the biohydrogen with CCS pathway. Upstream GHG emissions associated with electricity, natural gas and biomass production, plus transportation were also included in the study to give the most complete picture.

## 2. TECHNOLOGICAL ASPECTS

### 2.1 Feedstock quality

Thermochemical treatment of biomass feedstock, and gasification in particular, are gaining strong traction in Europe giving the numerous opportunities associated to product flexibility and low environmental impact. Recent studies have proven that Bio-$H_2$ offers the largest potential in terms of GHG removal (Rosa et al 2022; Valente et al., 2019), thanks to the biogenic origin of the carbon in the feedstock. However, Bio-$H_2$ production should ideally rely on the use of second or third generation biomass as primary feedstock to avoid land use competition with food crops and intensification of deforestation, habitat loss and loss of soil fertility. Municipal Solid Waste (MSW) and waste in general, represent an ideal source because of their large availability and low cost. From a climate change perspective, the use of waste as feedstock not only ensures large and economical availability for consistent hydrogen supply, but also avoids use of current disposal technologies, which are known to contribute enormously to GHG emissions and water and land pollution ( Materazzi & Foscolo, 2019).

Biomass to $H_2$ plant performances and environmental attributes are obviously strictly dependent on feedstock composition. Generally, the quantity of organic (biomass) content in the feedstock can vary from 40-60% in weight in household waste (see Table 1) to above 90% in waste wood. As such, hydrogen from household or commercial waste is only partially renewable due to the presence of plastics based carbon, and only the energy contribution from the biogenic portion is typically counted towards renewable energy targets (and only this element is eligible for renewable financial incentives). However, to simplify the discussion, we leave the prefix -bio in the hydrogen product from thermochemical plants regardless the percentage of biomass in the feedstock, to distinguish it from other low-carbon hydrogen routes. If the waste is pre-treated to separate out the biogenic fraction, and only this used for hydrogen production, then this can be considered wholly renewable. In fact, waste cannot be thermochemically treated in its original form when collected. The untreated municipal or commercial waste is first mechanically processed in a material recycling facility (MRF). This is done to homogenise the material and remove part of the moisture, recyclables (e.g., metals and dense plastics) and reject materials (e.g, oversize and inert). Waste treated to give greater than 90% biogenic content is considered to be on a par with biomass for many of the incentive schemes in UK, although as it is still a waste derived fuel, it remains subject to all the environmental controls relating to waste.

**TABLE 1:** Waste feedstock composition analysis.

| **Waste fractions [wt% as received]** | **MSW** | **Waste wood** |
|---|---|---|
| Paper and cardboard | 22.7 | 0.8 |
| Wood | 3.7 | 93.4 |
| Metals | 4.3 | 1.7 |
| Glass | 6.6 | - |
| Textile | 2.8 | - |
| WEEE | 2.2 | - |
| Plastics | 10 | 0.5 |
| Inert/aggregates/solid | 5.3 | 2.5 |
| Organic fines | 35.5 | 1.1 |
| Miscellaneous | 7.1 | - |
| **Proximate analysis [wt%, as received]** | **RDF** | |
| Fixed Carbon | 8.90 | 10.75 |
| Volatile matter | 64.70 | 64.24 |
| Ash | 11.80 | 0.41 |
| Moisture | 14.60 | 24.6 |
| **Ultimate analysis [wt%, dry ash free (DAF)]** | **RDF** | |
| Fossil Carbon | 20.51 | 0.80 |
| Biogenic Carbon | 36.23 | 50.13 |
| Hydrogen | 6.86 | 5.76 |
| Oxygen | 31.78 | 43.01 |
| Nitrogen | 4.1 | 0.28 |
| Sulphur | 0.18 | 0.01 |
| Chlorine | 0.34 | 0.01 |
| **Energy content [MJ/kg DAF]** | **RDF** | |
| Gross calorific value (HHV) | 28.99 | 24.08 |
| Net calorific value (LHV) | 27.02 | 22.73 |

The material is then shredded using tearing motion to achieve a rough shred of waste residues, with a homogenous, predetermined particle size between 1 and 50 mm, depending on the gasification reactor requirements. The final feedstock is in the form of floc of refuse derived fuel (RDF), which is then further dried on-site using waste heat from the process. Typically, a 100,000 tonnes MSW feed produces an output of ca. 60,000 to 80,000 tonnes of RDF with a moisture content of 10-17%, 10-20% ash content and 15-25 MJ/kg calorific value (CV) (Materazzi et al., 2019).

A good potential reference $WtH_2$ plant size could treat around 100,000 tonnes per annum of RDF, this being supplied from a reasonably sized town, accounting for residual domestic, commercial and industrial waste arisings. This is also similar in scale to small conventional energy from waste facilities. Bus fleets have been identified as the earliest likely adopters of hydrogen for transport. A typical bus will consume around 5 tonnes per annum of hydrogen. A large depot will operate around 100 buses, i.e. 500 tonnes per annum or 20 GWh. This equates to around 5% of the $WtH_2$ plant scale identified. This suggests that transport applications in the medium term are likely to be serviced by slip streams from larger plants designed to service grid or industrial customer applications.

## 2.2 Waste gasification development stage

Compared to pure biomass, waste feedstock introduces a greater concentration and diversity of contaminants, due to the high number and variability of sourcing points. This presents a major challenge, compounded by the fact that more sophisticated applications (including catalytic processes for Bio-$H_2$ production and fuel cells for transportations) have very low tolerances.

The state of technology development for biomass or waste gasification for fuels and hydrogen production is generally seen to be in the TRL (Technology Readiness Level) range 7 to 8, however, it is not a clear-cut issue. This has recently reviewed by the Department for Business, Energy & Industrial Strategy (BEIS) in UK (BEIS, 2021). Firstly, most biomass and waste-fuelled gasifiers are fundamentally unsuited to the production of syngas as an intermediate to hydrogen or gas fuel production, principally because they are air-aspirated rather than oxygen blown. Air-aspirated gasifiers entrain large volumes of nitrogen in the syngas – the removal of nitrogen from the product (hydrogen, biomethane etc.) being expensive and difficult to accomplish. It is important to note that much of industrial fatigue with biomass or waste gasification has been with gasifiers of this type. Gasifiers suited to the production of bio-hydrogen will not be air blown – they must be indirectly heated or oxygen/steam blown, and ideally they would operate significantly above ambient pressure. At least one gasification technology, the HT Winkler, having been deployed successfully in East Germany before unification, would appear to be at or near a TRL of 9. The HT Winkler is in a class of its own in this respect, but the owners of the technology, Gidara Energy, do not offer their technology for third party developers; it is reserved for their own projects. Other steam-oxygen technologies, such as the Thermoselect process, are deemed suitable for hydrogen production from waste, due to the high temperatures involved in the process which are effective towards removing tars and inertifying ashes. The process, now licensed by JFE group, has been demonstrated at sufficiently large scale for power production in gas turbines and for ancillary fuel gas for steel treatment plants (Yamada et al., 2004). However, there is no extant and diversified technology base at TRL 9 that could support the immediate and widespread implementation of thermochemical biohydrogen facilities at this time. At the current time investors in bio-hydrogen projects are faced therefore with procurement of gasification technologies that are at around TRL 7 or 8 for bio-syngas production; from which bio-hydrogen would be manufactured (Hofbauer & Materazzi, 2019). Technologies for production of hydrogen from syngas are well proven and at TRL 9, so the primary technological risk rests with the gasification technology.

Another class of suitable technologies is that of multi-stage conversion processes, which combine bulk gasification in conventional fluidized bed reactors with high temperature reforming steps to deal with tars. This has also the advantages that ashes, naturally abundant in waste feedstock, are collected in a vitrified form, which is classified as inert material.

Examples of these technologies, adopted in semi-commercial plants, are available in Canada and US, where gasification units, such as those licensed by TRI and Enerkem, are being tested at scale for production of clean syngas suitable for catalytic applications, specifically for jet fuel and methanol production. Some of these multi-stage technologies have been tested and demonstrated at pilot or demonstration scale in Europe and UK, but major challenges arise during scale-up (Materazzi & Taylor, 2019). To address this problem, the UK company ABSL embarked some years ago on a programme of developments at Swindon, beginning with a pilot-scale gasifier and plasma reforming technology, and 50kWth Bio synthetic natural gag (SNG) demonstration project. The pilot plant experience has enabled ABSL to continue development of the RDF to BioSNG/BioH$_2$ concept with a semi-commercial (1/10th scale) demonstration plant currently under commissioning in Swindon (Materazzi & Taylor, 2019). Up-scaling to a full commercial capacity (by a factor of 10) would be a reasonable stretch in capacity, subject to learning-by-doing and satisfactory performance with the semi-commercial demonstrator. Similar endeavours have been undertaken by other UK companies, such as Kew Energy, who are testing their pressurised fluidised bed gasifier (7MWt), coupled with a thermal cracker to break down the tars. However, such plants would be a first-of-a-kind facility and as such be seen by potential investors as presenting an enhanced technology risk, in comparison with technologies that had already accrued an operational track record and a TRL of 9. Hence, the demonstration of a semi-commercial facility that can be scaled-up by a reasonable scaling factor (e.g. a multiple of <10x), coupled with demonstration of satisfactory operation, will be a key step in addressing technology risk for waste to hydrogen plants, and several attempts can be observed already in UK and Europe landscapes (BEIS, 2021). Nevertheless the technology risk is enhanced compared to a "proven" technology, and pump-priming measures in the sector need to be such that investors see the enhanced risk as being acceptable; otherwise, it is likely that investment in the sector will not be forthcoming.

## 2.3 Pre-combustion $CO_2$ capture

Whilst post combustion capture from the flue gas of a biomass power station is not yet a common practice, the technologies used for both power generation and for post combustion capture are mature and each at a state of development where they could be classed as commercially proven. Hence, the technology risks associated with applying BECCS to biomass power generation are low.

Pre-combustion capture refers to removing $CO_2$ from syngas, typically post water gas shift stage in a gasification or pyrolysis plant. The same concept would apply to blue-hydrogen production plant, where syngas is generated grom natural gas, so the technology risks are shared between the two low carbon hydrogen pathways. Compared to post-combustion technology, which removes dilute $CO_2$ (~5-15% $CO_2$ concentration) from flue gas streams and is at low pressure, the shifted synthesis gas stream is rich in $CO_2$ and at ideally higher pressure, which allows for easier removal. Due to the more concentrated $CO_2$ (also due to the lack on N2 in syngas from steam-oxygen gasification), pre-combustion capture typically is more efficient, but the capital costs of the base waste gasification process and gas cleaning sections are often more expensive than traditional fossil-based power plants. Today's commercially available pre-combustion carbon capture technologies generally use physical or chemical adsorption processes, and will cost around $60/tonne to capture $CO_2$ generated by an integrated gasification combined cycle (IGCC) power plant (Balcombe et al., 2018). The goal of biohydrogen's research efforts is to reduce this cost to $30/tonne of $CO_2$. The commercial technologies for pre-combustion $CO_2$ capture available today share a similar process layout consisting of two successive phases of absorption and desorption of $CO_2$. The absorption phase uses a solvent to remove $CO_2$ from the shifted syngas, producing a $H_2$-rich stream. The following phase desorbs $CO_2$ regenerating the solvent that is recirculated to the absorption phase. These technologies differentiate according to the solvent used and the specific operating conditions that it requires. They are characterised as physical (e.g Selexol, Rectisol, etc.) or chemical (e.g. amine, Benfield, etc.) depending on whether the $CO_2$ is simply physically dissolved or is chemically bound to the solvent. A key difference is that chemical absorption requires increasing temperature for desorbing $CO_2$, whilst in physical absorption this can be achieved by solely reducing the pressure (L'Orange Seigo et al., 2014).

The $H_2$-rich stream is often purified via pressure-swing adsorption (PSA) to obtain a $H_2$ stream suitable for gas grid injection (>98% v/v) or transportation (>99.999% v/v), which is then compressed for storage. The tail gas from the PSA contains primarily $CO_2$ and $H_2$, as well as traces of other combustible (e.g. $CH_4$) and non-combustible (e.g. $N_2$) substances from syngas and of the solvent. The gas is burnt in a gas engine for generation of electricity and thermal energy. The former can be sold to the electric grid operator, whilst the latter is recovered in the WGS phase. On the other hand, the $CO_2$-rich stream from the desorption phase is compressed to 60 bar and transported via lorry, sea tankers and finally through pipelines prior to being injected in saline aquifier at 110 bar.

### 2.4 Alternative low-carbon hydrogen production routes in UK

Low carbon hydrogen in the UK is currently produced by electrolysers on-site at hydrogen refuelling stations; the emissions of which are dominated by the use of grid electricity for the electrolyser. However, by 2030, other production pathways will emerge. In addition to onsite electrolysis, low carbon hydrogen will be produced from natural gas, using either newbuild ATRs fitted with CCS, or by retrofitting old SMRs with CCS. Around 2030 or shortly after, large centralised or offshore electrolysers may emerge, with electrolysers directly connected to offshore wind turbines, and hydrogen transported to the shore by pipeline. At this time, it is likely that several biohydrogen plants with CCS will be operational in UK, and therefore, a comparison of impact of different producing technologies is critical. Figure 1 shows the comparison of different production routes, defined within specifed boundary conditions, under examination in this work.

## 3. METHODOLOGY

With the application of LCA according to ISO 14040 and ISO 14044 guidelines, a comparison of different low-carbon hydrogen production routes was undertaken (ISO, 2006a, 2006b). For the construction of this LCA model, primary inventory data for a 50 MW Bio-$H_2$ plant have been collected from the UK waste gasification company, ABSL. The production of Bio-$H_2$ from waste is considered a multifunctional process, defined as an activity that fulfils more than one function; in this case, the thermochemical process dealing with waste and energy generation. Following the relevant ISO standards, the environmental benefits from recovered resources should be accounted for by expanding the system boundaries to include the avoided burdens of conventional production (Clift et al., 2000). The environmental burdens of Bio-$H_2$ production include: the direct burdens allocated to all the operational units and elementary flows considered in the system boundaries; the indirect burdens allocated to the external supply of material and energy processes; and the avoided burdens allocated to the recovery of materials from waste during the RDF preparation stage (e.g. ferrous metals and non-ferrous metals), as well as the production of electricity and hydrogen. A conservative uncertainty analysis on the impact of Bio-$H_2$ has been carried out to account for application of different technologies and corresponding energy requirements, as well as the variation due to the waste composition which causes a large

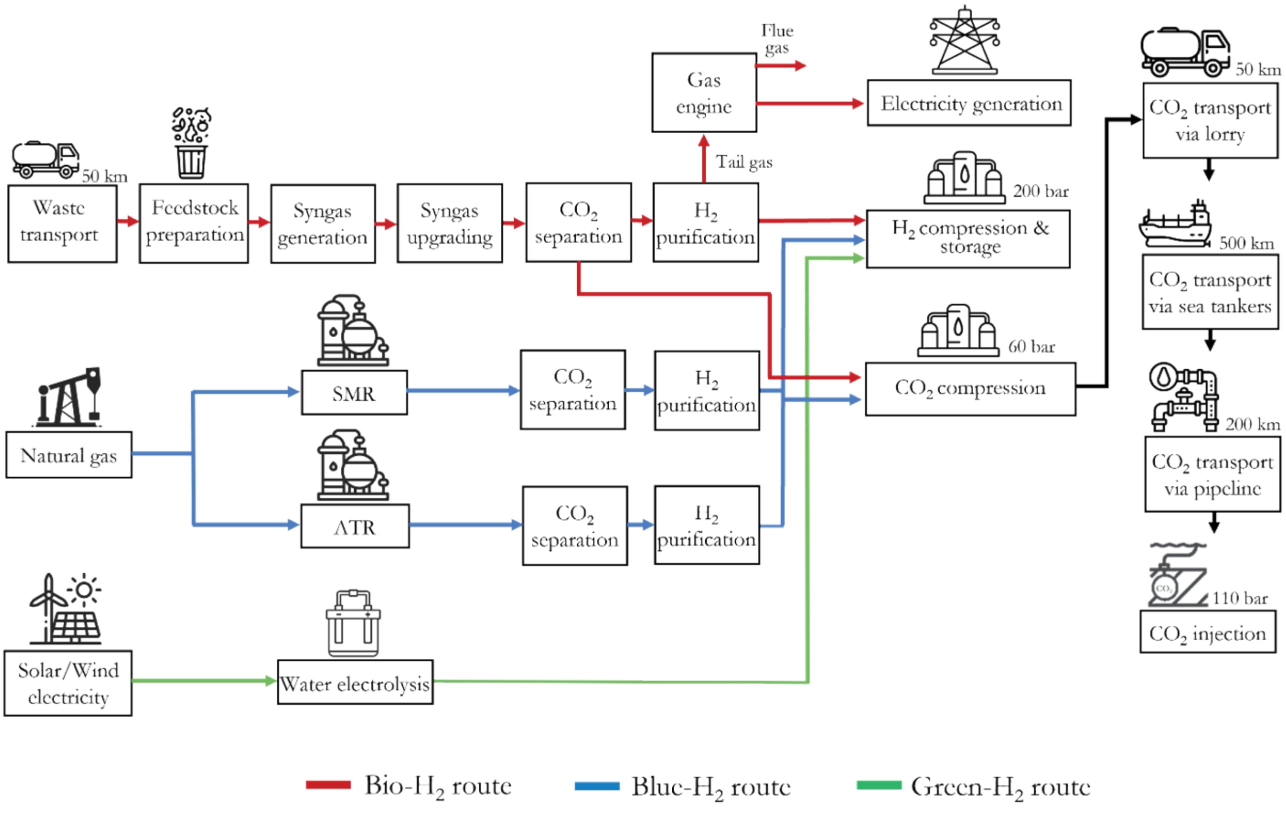


**FIGURE 1:** Schematic of low-carbon hydrogen production modelled steps.

part of the overall uncertainty (Amaya-Santos et al., 2021). A monoethanolamine (MEA) carbon capture technology was modelled with a 90% carbon capture rate for the Bio-$H_2$ models. Although MEA is not deemed the best industrial solvent for CCS, due to its low stability and high corrosivity, its choice was dictated by presence of detailed data in the literature and LCA database.

Table 2 reports the key inventory data of the three hydrogen production technologies, summarising the total input and output flows per functional unit (1 MWHHV of transport-grade $H_2$). The data from Antonini et al., (2020) for steam methane reforming and autothermal reforming coupled with 90% carbon capture using methyl diethanolamine as a solvent was used for comparative analysis. Description of the Green-$H_2$ inventory dat is presented in (Amaya-Santos et al., 2021).

The provision of external materials and energy to the process and the treatment of end-of-life wastes (Clift et al., 2000) are activities in the background system and are modelled using the cut-off system model, ecoinvent database (version 3.8) (Wernet et al., 2016). Such process include: the chemicals production and supply required as fluidising agents (e.g. oxygen), gas cleaning chemicals and CCS solvent (MEA); the net thermal energy and electricity requirements/generated; the treatment of wastewater effluents. Ecoinvent datasets were also used for $CO_2$ transportation via lorry and sea tankers. Life cycle impacts were assessed across the categories that represent the highest environmental priorities according to normalisation using the EF 3.0 global reference normalisation and weighting factors (Sala et al., 2019).

## 4. RESULTS AND DISCUSSION

### 4.1 Climate Change Impact

Different scenarios of the Bio-$H_2$ production process are presented with regards to climate change impact (CCI). These scenarios showcase the consequences of capturing point carbon emissions via carbon capture and storage (CCS) and of considering (thereby crediting) the biogenic carbon fraction of feedstock. A carbon capture rate of 90% is employed in all cases with CCS. The baseline biogenic fraction of RDF used is ~60% as per Table 1. When accounting for the difference between biogenic and fossil carbon, biogenic carbon emissions to air is considered carbon neutral and thus impacts from $CO_2$ process emissions of fossil origin are only considered. Corresponding scenarios produce a carbon negative impact when CCS is applied to the system, as carbon is effectively being removed from natural carbon cycle. This translates to the total climate change impact of the Bio-$H_2$ of -118 $CO_2$ eq./FU for scenario using RDF (from MSW) as a feedstock, and –293 kg $CO_2$ eq./FU for scenario using waste wood (WW). Bio-$H_2$ from WW outperforms MSW due to sequestration of its ~100% biogenic carbon content. The results for wood gasifcation are well corroborated with Antonini et al. (2021) with impacts ranging between -252 to -515 kg $CO_2$ eq. / MW $H_2$ albeit for different gasifier technologies modelled herein. The system boundaries of this analysis also include $CO_2$ transport and storage. Waste wood processing requires more initial feed throughput to generate the equivalent amount of hydrogen compared to MSW. Thus, resulting indirect process emissions are more positive for WW. Counterintuitively, the higher $CO_2$ production rate for waste wood also results in greater environmental impact savings.

In Figure 2, an additional analysis is presented, in which the avoided emissions associated to the MSW counterfactual are included. If not treated in advanced thermochemical facilities, current waste management practises call for disposal either through incineration or landfill. Incineration with energy recovery (WtE) represents the most common practice around the world and thus is considered as a realistic counterfactual. Similarly, to previous cases, only emissions associated to the fossil carbon fraction of feedstock have been accounted for. Although electricity and materials are recovered from the process, and thus credited on the final GHG output, the incineration option still shows a

**TABLE 2:** Key inventory data of the three hydrogen production processes. Flow quantity is referred to functional unit (1 MWHHV transport-grade $H_2$) and 1h as unit of time. *adapted from (Antonini et al., 2020).

| Key flows | Units | Biohydrogen (MSW) | Biohydrogen (Waste wood) | Blue Hydrogen | | Green Hydrogen |
|---|---|---|---|---|---|---|
| | | | | *SMR | *ATR | |
| **Input** | | | | | | |
| Feedstock type | | MSW/RDF | Waste wood | Natural gas | | Water |
| Feedstock | kg | 442.2/283.6 | 372.2 | | | 226.8 |
| | $m^3$ | | | 116.4 | 117.6 | |
| Oxygen | kg | 89.4 | 101 | | | n.a. |
| Electricity | MJ | 514 | 617 | 27.7 | 115.7 | 4974 |
| Thermal energy | MJ | 1550 | 1657 | - | - | n.a. |
| **Output** | | | | | | |
| Hydrogen [MJ] | MJ | 3600 | 3600 | 3600 | | 3600 |
| Materials recovered [kg] | Kg | 17.1 | - | - | - | - |
| $CO_2$ released [kg] | kg | 46.5 | 53.5 | 120.63 | 38.1 | 0 |
| Sequestered $CO_2$ [kg] | kg | 414.4 | 484.1 | | | n.a. |

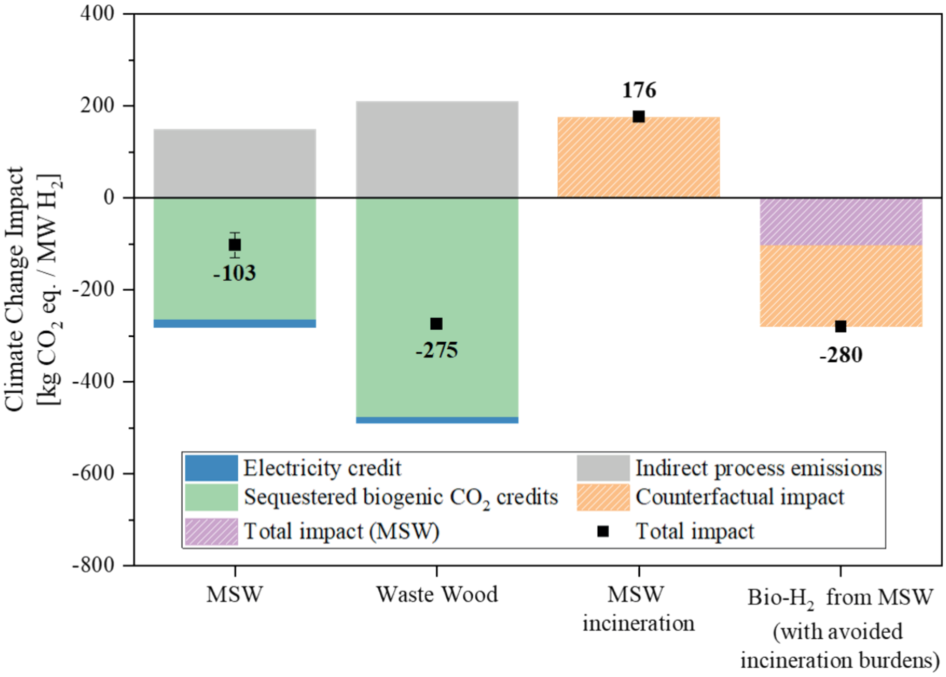


**FIGURE 2:** Climate change impact ($CO_2$ eq. per FU) regarding carbon capture and storage and considering the biogenic fraction of the $CO_2$ stream. Uncertainties calculated based on technical variations in energy usage and feedstock composition.

substantial climate change contribution of 176 kg $CO_2$ eq. for the equivalent amount of MSW to produce 1 MWHHV $H_2$. Therefore, by diverting waste from being incinerated, the benefit of Bio-$H_2$ on climate change can be further pronounced, with a negative contribution to climate change of -280 kg $CO_2$ eq. per MWHHV of $H_2$ produced. Even higher benefits could be observed if considering other waste management practises, such as landfill or incineration with no energy recovery, as a counterfactual.

## 4.2 Comparative analysis between Bio-$H_2$, Blue-$H_2$ and Green-$H_2$

### 4.2.1 Climate change impact

In the present analysis, the environmental performance of the Bio-$H_2$ technology is compared to other two competitive low-carbon technologies, Blue- and Green-$H_2$, considering the Climate Change (kg $CO_2$ eq.) impact only. The results are expressed per functional unit, 1 MWHHV of transport grade hydrogen produced from all examined processes. The comparison of the environmental performance of the three routes has been performed taking into account the environmental burdens allocated solely to the production of hydrogen i.e. excluding system expansion methodology. In this analysis, $CO_2$ transportation and storage have not been included across technologies for consistency. The contributions to climate change of the technologies are depicted in Figure 3. Bio-$H_2$ production shows the lowest contribution to climate change, equating to -293 kg $CO_2$ eq for waste wood and -118 kg $CO_2$ eq/FU for MSW. These results show that the production of hydrogen from MSW or waste wood together with the sequestration of carbon, is not only an effective solution to waste disposal, but it is also appropriate to achieve the objectives proposed by the Net Zero 2050; its implementation involves the removal of a fifth to a third ton of $CO_2$ per MWHHV of $H_2$ produced every hour.

Blue-$H_2$ produced via steam methane reforming process (SMR) with CCS process (carbon capture rate of 90%, with MDEA $CO_2$ adsorption) produces 143 kg $CO_2$ eq. per MWHHV $H_2$ as modelled by Antonini et al. (2020). The use of alkanolamines, MEA and MDEA, are widely used solvents in amine-based capture technologies. This study assumes a similar impact from MEA and MDEA for comparison, although it has been shown that for certain applications MDEA may fare better energetically. Approximately 32% of the impact derives from the embodied carbon of natural gas feedstock, rendering the process sensitive to changes in natural gas source (Antonini et al., 2020). According to the authors, 66% percent of impact derives from direct $CO_2$ emissions. The upstream emissions are associated to its processing and, for imported NG, to its liquefaction and shipping. The remaining climate change impact is ascribed to the electricity required during the steam reforming and carbon capture process, $CO_2$ liquefaction and $H_2$ compression. The difference between SMR and ATR in favour of ATR is related to the higher $CO_2$ fraction in the syngas generated by the latter, and therefore, more efficient carbon capture.A competitive Green-$H_2$ route of production is limited by the high electricity demand of the electrolyser. Amaya-Santos et al. (2021) reports a large environmental burden when electrolysis is conducted using the current electricity grid mix. By using low-carbon intensity grid supplied by renewable sources, this limitation can be overcome. In a similar vein, any processes with a high electricity input will benefit from future decarbonisation of the grid. As shown in Figure 3, the electricity demand of the electrolyser and $H_2$ compression unit met by electricity produced 100% from solar and 100% from offshore wind contribute 99 kg $CO_2$ eq. per

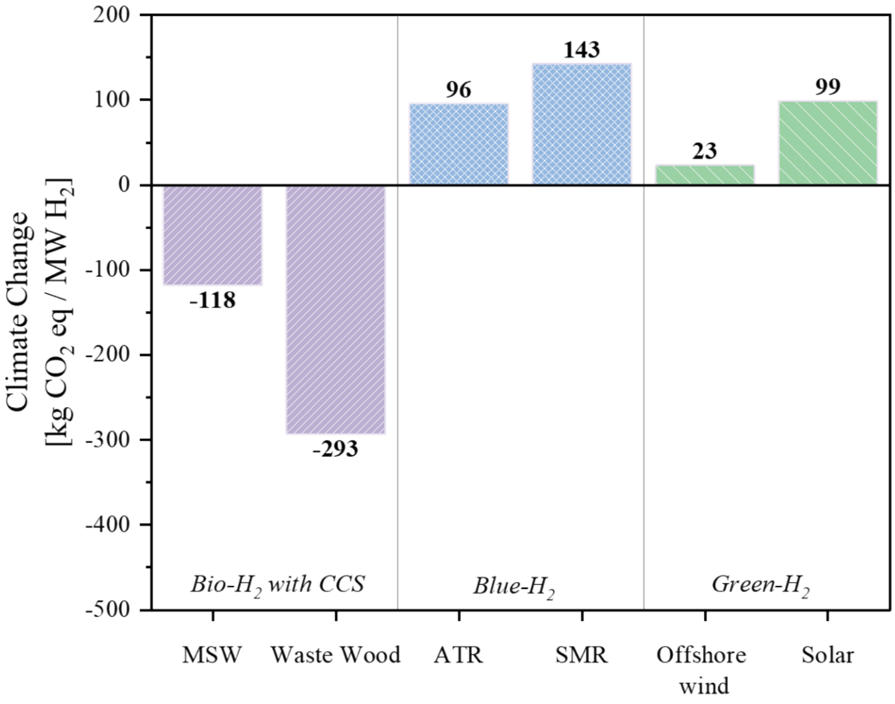


**FIGURE 3:** Climate Change contribution comparison of Bio-$H_2$, Blue-$H_2$ and Green-$H_2$ production technologies.

MWHHV $H_2$, and 23 kg $CO_2$ eq. per MWHHV $H_2$, respectively. The greater impact from solar compared to offshore wind is attributed to the manufacturing of silicon solar cells.

### *4.2.2 Environmental impact of other categories*

The impacts for all other categories are normalized to the absolute maximum value in each category thereby depicting a ranking of environmental performances between technologies, shown in Figure 4. For assessing carbon neutral or negative technologies, it becomes imperative to understand the trade-offs that come with, in the case of Blue- and Bio-$H_2$, the implementation of CCS.

Between the gasification of MSW and waste wood, waste wood tends to perform worse on all impacts categories owing primarily to differences in feedstock throughputs. MSW produces more hydrogen per input of waste feedstock than biomass owing to its higher hydrogen content and lower oxygen content than biomass (see proximate analysis, Table 1). Therefore, with overall improved energy efficiency and feedstock conversion efficiency, MSW may serve as a preferential technology that is carbon-negative while also reducing the trade-off across other categories. The higher net electricity consumption of Bio-$H_2$ (143 kWh/MW $H_2$ for MSW and 171 kWh/MW $H_2$ for WW) compared to Blue-$H_2$ reported by Antonini et al. (2020) (18 and 53 kWh/MW $H_2$ for SMR and ATR respectively) plays a part in Bio-$H_2$ faring worse in Acidification, Ecotoxicity, Eutrophication (marine & terrestrial), Ionizing radiation and Photochemical Ozone Formation. While, Resource – Use (fossil) and Ozone Depletion are led by Blue – $H_2$. Eutrophication (freshwater) is led by Green-$H_2$ from offshore wind and Human Toxicity, Land Use and Particulate Matter are led by Green-$H_2$ from solar.

## 4.3 Interaction between low carbon hydrogen production pathways

It will be extremely challenging for any one of the low carbon hydrogen technologies to meet alone the expected level of hydrogen demand set out by Net Zero ambition. It seems likely that all options will play a role in the transition to hydrogen. Green hydrogen has the potential to be produced sustainably in large volumes. However, it will take time for low carbon electricity generation to grow to the scale that meets current electricity demand, plus the additional demand required to decarbonise heat and transport with hydrogen. In addition, electrolyser technology requires several years to develop to the point that it can produce hydrogen at costs that compete with blue hydrogen. Blue hydrogen can be produced at large scale in a few years' time at relatively low cost. However, it is a less sustainable solution in the long term and cannot match the carbon savings achieved by green hydrogen and biohydrogen. Biohydrogen has the potential to generate negative carbon emissions if combined with carbon capture and storage. However, overall production of biohydrogen is limited by the availability of sustainable feedstock. It cannot be produced in sufficient volumes to meet the overall expected demand.

There are important synergies between different low carbon hydrogen production pathways. For example, blue hydrogen might establish the hydrogen market that green hydrogen will meet in future or build the carbon sequestration network required for biohydrogen to deliver negative emissions. These negative emissions can offset the residual emissions from blue and green hydrogen production. Green hydrogen might supply hydrogen to consumers that are remote from the blue hydrogen production centres. The different hydrogen production options all have different

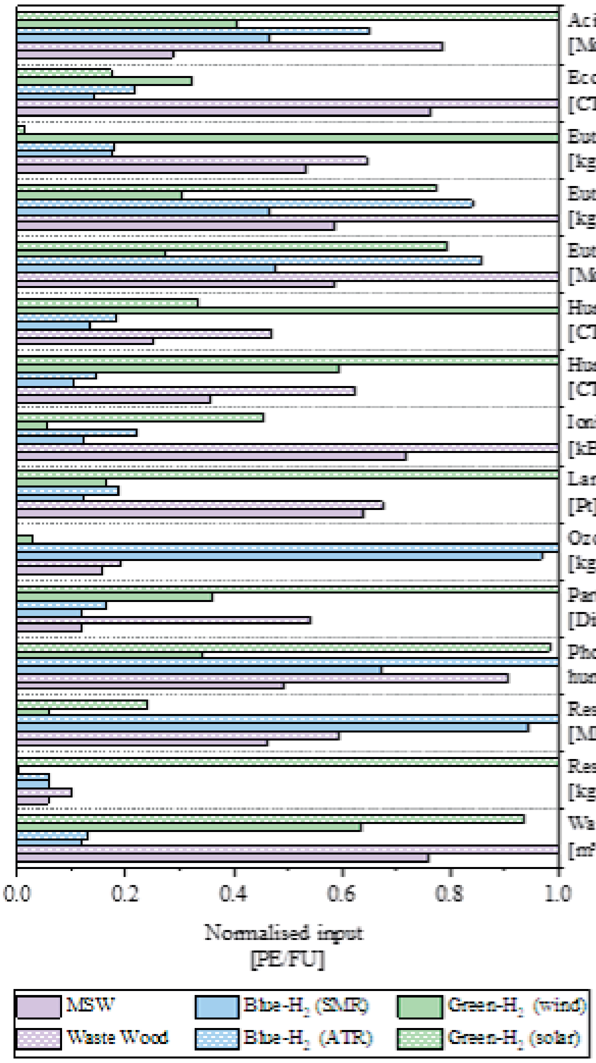


**FIGURE 4:** Life cycle performance of all other impact categories normalized. Absolute impact numbers are normalised to 1 to the technology with the highest impact in each category.

infrastructure requirements, with blue and bio hydrogen reliant on CCS infrastructure. The large-scale ATR plants required for blue hydrogen production also suits the large industrial clusters, where infrastructure, such as a supply of natural gas and potentially by-product oxygen are available. The industrial clusters across the UK would therefore suit blue hydrogen production, with the locations along the East coast and North West England developing plans for carbon dioxide pipelines for offshore carbon dioxide storage (see Figure 5). The industrial clusters in South Wales and Southampton would require shipping carbon dioxide to offshore storage sites. Green hydrogen production can be developed at smaller scale than blue hydrogen and although green hydrogen does not require CCS infrastructure, there are benefits to installing electrolysers alongside renewables or close to hydrogen demands / hydrogen infrastructure. The map of industrial clusters in Figure 3 also highlights regions where there are large energy demands from industrial processes, which could become early adopters of hydrogen. At a smaller scale, where hydrogen can be transported via road tankers to serve transport demands, green hydrogen production plants could be located where renewables are best-sited (to access lowest cost power). In the medium term, larger green hydrogen production plants will be developed either in locations close to very large renewable assets (e.g. in coastal locations where offshore wind farm electricity is landed) or in locations closer to large-scale users, such as industrial clusters, to avoid long range hydrogen transport before wider conversion of the gas network becomes available to transport 100% hydrogen. A 100% hydrogen gas network would open up more options for green hydrogen production sites, including the production of hydrogen offshore, connected to offshore wind farms. At a certain scale, the cost of transporting energy in a gaseous form (as hydrogen) can be lower than the costs of transporting energy via electricity. There would be further cost benefits for hydrogen transport if oil and gas pipelines could be repurposed for hydrogen transport. However, an environmental consideration to $H_2$ transportation means and distances in the ongoing research on the impact of fugitive hydrogen emissions on the greenhouse gas effect (BEIS, 2022). Biohydrogen would require CCS

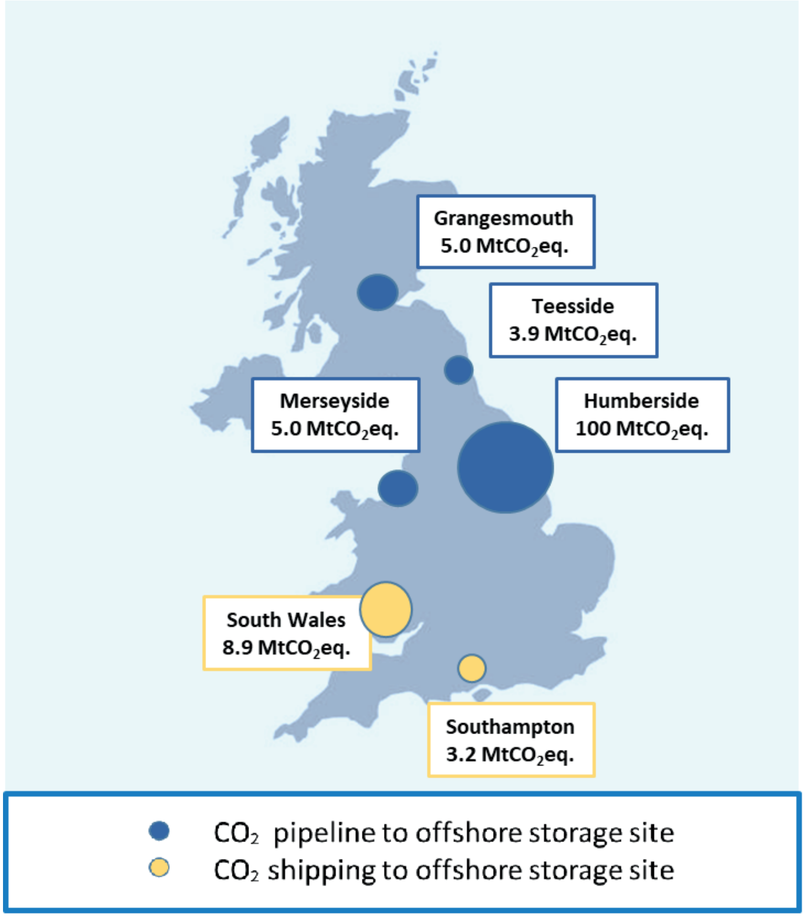


**FIGURE 5:** Location of industrial clusters in the UK and annual GHG emissions from each (Adapted from BEIS, 2021).

infrastructure to deliver very high GHG savings and therefore is suited to the industrial clusters shown in Figure 4. The use of biohydrogen without CCS can still provide GHG emission savings relative to incumbent fuels and converts waste streams into a valuable product, with hydrogen a higher value output than electricity from energy from waste plants. There could therefore be a degree of flexibility with regard to siting some of the plants at locations without CCS infrastructure across the UK, although the full benefits of the technology would require siting around the industrial clusters, or locations with carbon dioxide demand. Biohydrogen technology can also be deployed at far smaller scales than blue hydrogen, allowing it to offer a more distributed approach to hydrogen production.

## 5. CONCLUSIONS

There is a significant scale-up challenge for low-carbon hydrogen production if the UK is to meet its Net Zero target by 2050. Given the significant demands for low carbon hydrogen it is clear that all of the three low-carbon production routes are needed and that these need to be developed at pace. The build out rates for all the options presented above will be challenging to meet, and support to develop low carbon hydrogen markets will be needed to encourage investment in delivering the scale-up of the hydrogen production capacity. This work showed that Bio-$H_2$ can be a competitive technology to aid the near- and medium-term transition to hydrogen economy, as well as a long-term complement to other low carbon hydrogen alternatives. Not only it is an effective solution to waste disposal, but it is also appropriate to achieve the objectives proposed by the Net-Zero 2050 for it being a viable carbon-negative technology.

## REFERENCES

Al-Qahtani, A., Parkinson, B., Hellgardt, K., Shah, N., & Guillen-Gosalbez, G. (2021). Uncovering the true cost of hydrogen production routes using life cycle monetisation. Applied Energy. https://doi.org/10.1016/j.apenergy.2020.115958

Amaya-Santos, G., Chari, S., Sebastiani, A., Grimaldi, F., Lettieri, P., & Materazzi, M. (2021). Biohydrogen: A life cycle assessment and comparison with alternative low-carbon production routes in UK. Journal of Cleaner Production, 319, 128886. https://doi.org/10.1016/j.jclepro.2021.128886

Antonini, C., Treyer, K., Moioli, E., Bauer, C., Schildhauer, T. J., & Mazzotti, M. (2021). Hydrogen from wood gasification with CCS-a techno-environmental analysis of production and use as transport fuel. Sustainable Energy and Fuels. https://doi.org/10.1039/d0se01637c

Balcombe, P., Speirs, J., Johnson, E., Martin, J., Brandon, N., & Hawkes, A. (2018). The carbon credentials of hydrogen gas networks and supply chains. In Renewable and Sustainable Energy Reviews. https://doi.org/10.1016/j.rser.2018.04.089

Barisano, D., Bocci, E., Foscolo, P. U., Heidenreich, S., Rep, M., Courson, C., Sentis, L., Di Carlo, A., Rapagna, S., & Braccio, G. (2017). Hydrogen production from biomass via gasification process: The results of the eu UnifHY project. European Biomass Conference and Exhibition Proceedings.

BEIS. (2021). Advanced Gasification Technologies - Review and Benchmarking (Issue 038). www.gov.uk/government/publications/advanced-gasification-technologies-review-and-benchmarking

BEIS. (2022). Fugitive Hydrogen Emissions in a Future Hydrogen Economy.

Cao, L., Yu, I. K. M., Xiong, X., Tsang, D. C. W., Zhang, S., Clark, J. H., Hu, C., Ng, Y. H., Shang, J., & Ok, Y. S. (2020). Biorenewable hydrogen production through biomass gasification: A review and future prospects. Environmental Research. https://doi.org/10.1016/j.envres.2020.109547

Chapman, A., Itaoka, K., Hirose, K., Davidson, F. T., Nagasawa, K., Lloyd, A. C., Webber, M. E., Kurban, Z., Managi, S., Tamaki, T., Lewis, M. C., Hebner, R. E., & Fujii, Y. (2019). A review of four case studies assessing the potential for hydrogen penetration of the future energy system. In International Journal of Hydrogen Energy. https://doi.org/10.1016/j.ijhydene.2019.01.168

Chong, M. L., Sabaratnam, V., Shirai, Y., & Hassan, M. A. (2009). Biohydrogen production from biomass and industrial wastes by dark fermentation. In International Journal of Hydrogen Energy. https://doi.org/10.1016/j.ijhydene.2009.02.010

Clift, R., Doig, A., & Finnveden, G. (2000). The application of Life Cycle Assessment to Integrated Solid Waste Management. Part 1 - Methodology. Process Safety and Environmental Protection. https://doi.org/10.1205/095758200530790

Cristina Antonini, Karin Treyer, Anne Streb, Spek, M. van der, Christian Bauer, & Marco Mazzotti. (2020). Hydrogen production from natural gas and biomethane with carbon capture and storage – A techno-environmental analysis. Sustainable Energy & Fuels, 4(6), 2967–2986. https://doi.org/10.1039/D0SE00222D

Edwards, R. L., Font-Palma, C., & Howe, J. (2021). The status of hydrogen technologies in the UK: A multi-disciplinary review. Sustainable Energy Technologies and Assessments. https://doi.org/10.1016/j.seta.2020.100901

Götz, M., Lefebvre, J., Mörs, F., McDaniel Koch, A., Graf, F., Bajohr, S., Reimert, R., & Kolb, T. (2016). Renewable Power-to-Gas: A technological and economic review. In Renewable Energy (Vol. 85, pp. 1371–1390). https://doi.org/10.1016/j.renene.2015.07.066

Gov.uk. (2019). UK becomes first major economy to pass net zero emissions law. In Department for Business Energy & Industrial Strategy.

Hofbauer, H., & Materazzi, M. (2019). Waste gasification processes for SNG production. In Substitute Natural Gas from Waste: Technical Assessment and Industrial Applications of Biochemical and Thermochemical Processes. https://doi.org/10.1016/B978-0-12-815554-7.00007-6

ISO. (2006a). ISO 14040 Environmental Management - Life Cycle Assessment - Principles and Framework.

ISO. (2006b). ISO 14044 Environmental Management - Life Cycle Assessment - Requirements and Guidelines.

L'Orange Seigo, S., Dohle, S., & Siegrist, M. (2014). Public perception of carbon capture and storage (CCS): A review. In Renewable and Sustainable Energy Reviews. https://doi.org/10.1016/j.rser.2014.07.017

Materazzi, M., Taylor, R., & Cairns-Terry, M. (2019). Production of biohydrogen from gasification of waste fuels: Pilot plant results and deployment prospects. Waste Management, 94. https://doi.org/10.1016/j.wasman.2019.05.038

Materazzi, Massimiliano, & Foscolo, P. U. (2019). The role of waste and renewable gas to decarbonize the energy sector. In Substitute Natural Gas from Waste: Technical Assessment and Industrial Applications of Biochemical and Thermochemical Processes. https://doi.org/10.1016/B978-0-12-815554-7.00001-5

Materazzi, Massimiliano, & Taylor, R. (2019). The GoGreenGas case in the UK. In Massimiliano Materazzi & P. U. Foscolo (Eds.), Substitute Natural Gas from Waste (pp. 475–495). Academic Press (Elsevier).

Materazzi, Massimiliano, Taylor, R., & Cairns-Terry, M. (2019). Production of biohydrogen from gasification of waste fuels: Pilot plant results and deployment prospects. Waste Management. https://doi.org/10.1016/j.wasman.2019.05.038

Rosa, L., & Mazzotti, M. (2022). Potential for hydrogen production from sustainable biomass with carbon capture and storage. Renewable and Sustainable Energy Reviews. https://doi.org/10.1016/j.rser.2022.112123

Valente, A., Iribarren, D., & Dufour, J. (2019). Life cycle sustainability assessment of hydrogen from biomass gasification: A comparison with conventional hydrogen. International Journal of Hydrogen Energy. https://doi.org/10.1016/j.ijhydene.2019.01.105

Yamada, S., Shimizu, M., & Miyoshi, F. (2004). Thermoselect waste gasification and reforming process. JFE Technical Report.

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# SIMULTANEOUS REMOVAL PROCESS OF HYDROGEN SULFIDE AND SILOXANES AND FIELD APPLICATION OF IRON HYDROXIDE DESULFURIZATION AGENT FOR GREEN HYDROGEN PRODUCTION FROM BIOGAS

Jae Myung Lee [1], Jaechul Shin [1], Min Ju Lee [1], Wha Jung Kim [1], Hyoungwoon Song [2] and Shin Dong Kim [1,*]

*[1] Rearch Center of Energy & Environment, E&Chem Solution Corp., Pocheon 11154, Republic of Korea*
*[2] Plant Process Development Center, Hydrogen Energy Solution Center, Yongin 17180, Republic of Korea*

**Article Info:**
Received:
2 February 2023
Revised:
2 May 2023
Accepted:
15 June 2023
Available online:
30 June 2023

**Keywords:**
Iron hydroxide desulfurization agent
steam methane reforming
Hydrogen sulfide
Biogas pretreatment
Siloxanes

**ABSTRACT**
Biogas, one of renewable energies, is a key element necessary for a carbon-neutral policy and to build a hydrogen economy. In order to utilize biogas, impurities of biogas such as moisture, hydrogen sulfide ($H_2S$), siloxanes, and VOCs should be removed. In particular, since $H_2S$ causes corrosiveness of equipment by sulfur oxides, and is very harmful to the human body if leaked, it is a major target material to be removed. The minimum concentration of $H_2S$ obtainable from the wet method is several ppm. It is known, however that the iron hydroxide-based adsorbent in the dry method can obtain ultimately low concentration of $H_2S$ down to 0.1 ppm or less. The DeHyS was manufactured through a series of processes such as mixing iron cloride solution or iron sulfate solution, NaOH solution, and inorganic binder. During the adsorption process, $H_2S$ was removed in the form of iron sulfide through a chemical reaction, and siloxanes are known to be removed through physical adsorption. It was also applied to various biogas plant sites such as landfill gas, sewage sludge, livestock manure, and food waste. At this time, the $H_2S$ removal efficiency was known to be 99.9% or more, while simultaneous removal of 90% or more of the total siloxanes was possible. Moreover, the biogas produced at the Chungju Food Bioenergy Center was pretreated using the DeHyS and supplied to the nearby Chungju Bio Green Hydrogen Charging Station to produce hydrogen through steam methane reforming (SMR), producing 500 kg of hydrogen from 8,000 $m^3$ of biogas per day.

## 1. INTRODUCTION

According to the Intergovernmental Panel on Climate Change [IPCC] special report (IPCC, 2018), the average global temperature in 2017 has risen by about 1°C compared to the pre-industrial period (1850-1900) due to human activities based on fossil fuels, and global warming is increasing at the current rate. If it continues, it is predicted that the global temperature rise will exceed 1.5°C between 2030 and 2052, and exceed 3°C by 2100. In order to limit the increase in global temperature to less than 1.5°C by 2100, it is recommended that the reduction of $CO_2$ of at least 45% compared to 2010 by 2030 as well as the achievement of zero net $CO_2$ emission by 2050 be necessary.

In December 2015, the Paris Agreement, a common norm of the international community, was adopted at the 21st COP21 of the UN Framework Convention on Climate Change [UNFCCC] to reduce greenhouse gas emissions (UNFCCC 2015). Republic of Korea declared carbon neutrality in October 2020 to participate in the efforts of the international community. Republic of Korea in December 2020, also promoted low-carbon in all areas of the economic structure and a low-carbon industrial ecosystem, and the ‘2050 Carbon Neutral Promotion Strategy’ was announced, focusing on strengthening neutral infrastructure, etc (Ministry of Environment in the Republic of Korea, 2020). In addition, 10 core technology areas for carbon-neutral technological innovation were selected through consultation with experts and related ministries. It is therefore expected that the energy market will grow rapidly as changes in the energy ecosystem.

In particular, hydrogen can be used for large-capacity energy storage, long-distance transport and a mobility and distributed power source as needed. However, at present, about 99% of domestic hydrogen production is gray hydro-

* Corresponding author:
Shin Dong Kim
email: catalite@hanmail.net

Detritus / Volume 23 - 2023 / pages 76-82
https://doi.org/10.31025/2611-4135/2023.18276

gen produced through steam methane reforming(SMR) from natural gas. On the other hand, eco-friendly hydrogen produced by the electrolysis of water called as green hydrogen and waste resources are hardly commercialized (The Government of the Republic of Korea, 2019). Therefore, in order to realize carbon neutrality, it is important to increase the proportion of blue hydrogen that captures carbon dioxide from fossil fuel such as natural gas and green hydrogen produced from steam reforming of methane from renewable energy resource such as biogas.

Biogas is produced by anaerobic microorganisms from organic waste such as food waste, livestock manure, and sewage sludge, which is usually composed of $CH_4$ 50-65%, $CO_2$ 30-40%, $H_2S$ less than 1%, and others. Methane in biogas is then converted to hydrogen through steam reforming reaction. It is known as a carbon-neutral renewable energy source that must be actively used because it is essential through human life.

As of 2021, there were a total of 110 domestic biogas facilities with an annual biogas production of 360 million Nm3 (0.05 GW). Although only 6.9% of the current market potential is being converted into energy, it is known that the existing feed and composting facilities can be converted to biogas plants of new construction/extension and can be expanded up to 14.4 times higher than the current biogas production (Ministry of Environment of the Republic of Korea, 2022).

In order to utilize biogas, impurities of biogas such as moisture, hydrogen sulfide, siloxanes, and VOCs should be removed. Since hydrogen sulfide causes corrosiveness of equipment by sulfur oxides, and is very harmful to the human body if leaked, it is a major target material to be removed. $H_2S$ removal in biogas can be achieved either by wet or dry method. In the wet method, chemical cleaning and iron chelate cleaning are used, while in the dry method, various adsorbent systems such as iron oxide-based adsorbents, magnesium-based adsorbents, activated carbon, and iron hydroxide-based adsorbents can be used . The minimum concentration of hydrogen sulfide obtainable from the wet method is several ppm. It is known, however that the iron hydroxide-based adsorbent in the dry method can obtain ultimately low concentration of $H_2S$ down to 0.1 ppm or less (Magnone, E., Kim, S. D., & Park, J. H., 2018).

In particular, since the proton exchange membrane fuel cell (PEMFC) operates at a relatively low temperature compared to other fuel cells, impurities such as CO and $H_2S$ are easily adsorbed to the catalyst, which may cause poisoning and thus deactivation of catalyst. The US Department of Energy recommends that hydrogen used as fuel be included in the amount of CO and $H_2S$ well below 50 ppm and 1 ppm, respectively to reduce performance degradation caused by fuel cell poisoning (Solutions, 2000). More recently, international organization for standardization(ISO) set the CO concentration limit for the conventional Pt anode as 0.2 ppm (Li et al., 2021; St-Pierre, 2010). Consequently, hydrogen production from biogas requires more stringent pretreatment. In Republic of Korea, the acceptable limit of $H_2S$ from biogas in various application fields is classified, based on the concentration of $H_2S$ and sulfur(S) as follows (Figure 1). Gas engine power generation requires less than 150 ppm as the most tolerant application field for hydrogen sulfide concentration, followed by city gas with less than 30 ppm on sulfur content, compressed transport gas less than 10 ppm on sulfur basis, and gas conditions for hydrogen production is known to be 0.01 ppm or less based on hydrogen sulfide.

Through this case study, we would like to investigate the gas conditions for hydrogen production from biogas, and introduce the principle and field application results of $H_2S$ and siloxanes removal of iron hydroxide-based desulfurization agents. In addition, we will discuss the expansion of the scope of biogas utilization while introducing representative domestic sites that produce hydrogen from biogas and use it as a fuel for hydrogen vehicles.

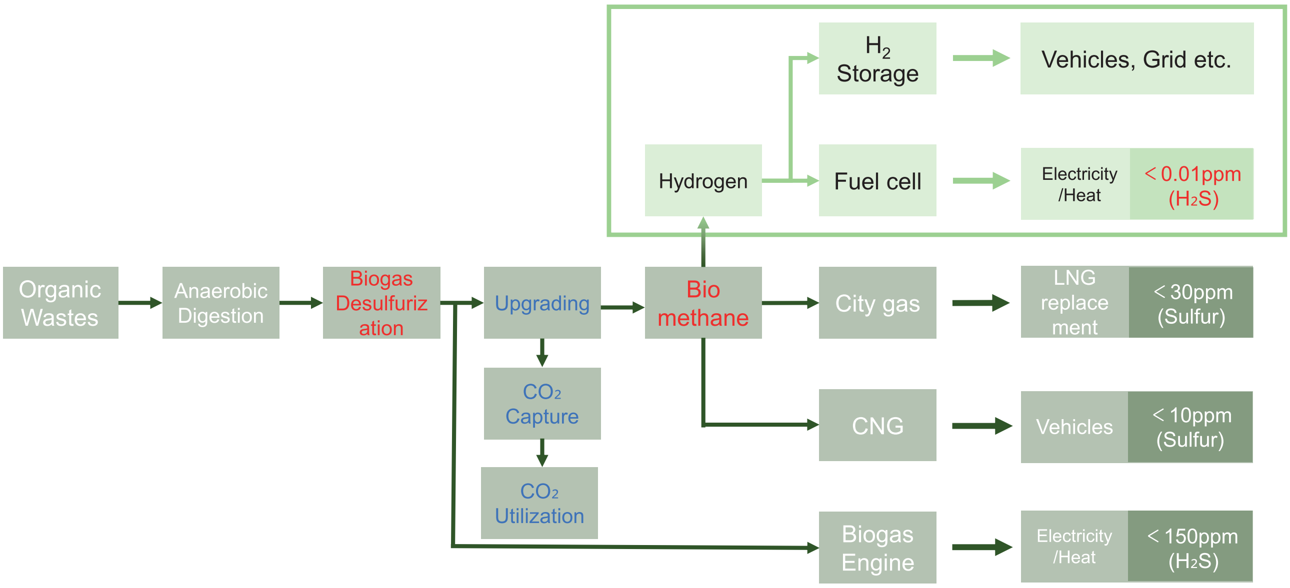


**FIGURE 1:** The acceptable limit of $H_2S$ and sulfur from biogas in various application fields.

## 2. EXPERIMENTAL

### 2.1 Materials

The adsorbent used for the removal of $H_2S$ and siloxane was porous and amorphous iron hydroxide-based commercial product, DeHyS (E&Chem Solution CO.). The DeHyS was identified using the X-ray diffraction analysis (XRD, Model D/Max 2200, Rigaku) with CuKα radiation source. 40 kV and 30 mA were adopted at 5° $min^{-1}$ and 0.08° angular resolution based on 2θ scan. DeHyS was manufactured by several steps; that is, appropriate ammounts of NaOH solution and inorganic binder (Magnone et al., 2018) were vigorously mixed under agitation to which iron chloride or iron sulfate solution was added, followed by additional 1 hr of agitation. Upon completion of agitation, the iron hydroxide-based precipitate was filtered using vacuum pump, followed by sufficient washing using distilled water. The precipitate so obtained was dried at 150°C until water content was reduced to less than 30% and cylindrical shape of pellet with diameter of 3 to 10 mm was then fabricated. The physical properties of DeHyS are summarized in Table 1 (Ryu et al. 2017).

### 2.2 Adsorption process

*2.2.1 Removal of hydrogen sulfide*

Different volume of adsorbent was loaded into the adsorption tower to which feed gas containing different concentrations of $H_2S$ and space velocities with field site as shown in Table 2 and Figure 2 was fed at ambient temperature. The packing density of adsorbent (DeHyS) in adsorption tower is 0.5~0.7 g/mL. The effluent gas passing through the adsorption tower was collected at the outlet of the tower and the residual concentration of $H_2S$ was identified by GC using pulsed flame photometric detector (GC/PFPD, Varia 450).

*2.2.2 Removal of siloxane*

Siloxane was simultaneously removed with $H_2S$ as described in previous section. In order to identify the removal efficiency of siloxane, its concentrations at the inflow and outflow of the desulfurization tower at Yeoyang Farm, Tancheon Water Treatment Center, and the Sudokwon Landfill Site were measured as follows. Total 12 L of sample was collected in a methanol absorption solution (10mL) at a flow rate of 100 mL/min for total 120 minutes, and the concentration of siloxane was then identified by GC/MS (Shimadzu, GC-2010/QP-2010).

## 3. RESULTS AND DISCUSSION

### 3.1 Results of field application for purifying biogas

The XRD pattern of DeHyS is shown in Figure 3. As can be seen in this figure, it does not show any sharp peak indicating that DeHyS is amorphous phase.

**TABLE 1:** Specification of Iron hydroxide-based desulfurization agent (DeHyS).

| Content | Range | Standard Method |
|---|---|---|
| Dimensions (diameter) | 3~10 mm | KS B ISO13385-1 |
| Fe $(OH)_3$ content | > 50 % (as $Fe_2O_3$) | KS L ISO 26845 |
| Loss on drying | < 20 % | KS M 0009 |
| Specific surface area | > 250 $m^2/g$ | KS A 0094 |
| Pore volume | 0.25 $cm^3/g$ | |
| Average pore size | 36 Å | |
| Adsorption capacity | 0.25 g/g | |
| Packing density | 0.5~0.7 g/mL | |

The DeHyS was applied to the desulfurization tower of the dry process, and the dry process comprises three steps: dehumidification process, desulfurization process, and dust removal process. It was applied to various fields such as food-derived biogas facilities, sewage sludge digester facilities, livestock manure biogas facilities, and landfill gas facilities at landfills in the Sudokwon (Table 2). The $H_2S$ concentration in biogas feed stream was varied from about 77.60 ppm to 4,450ppm depending on the site, but the that in deplete biogas stream passing through adsorption tower was measured to be 0.01ppm or less (based on KTL official test report). It showed that the required $H_2S$ concentration in the field was highly satisfied, resulting in the $H_2S$ removal efficiency up to 99.9% or more as shown in Table 2.

In Yeoyang Farm, it seems that almost no siloxanes were detected in case of biogas derived from livestock

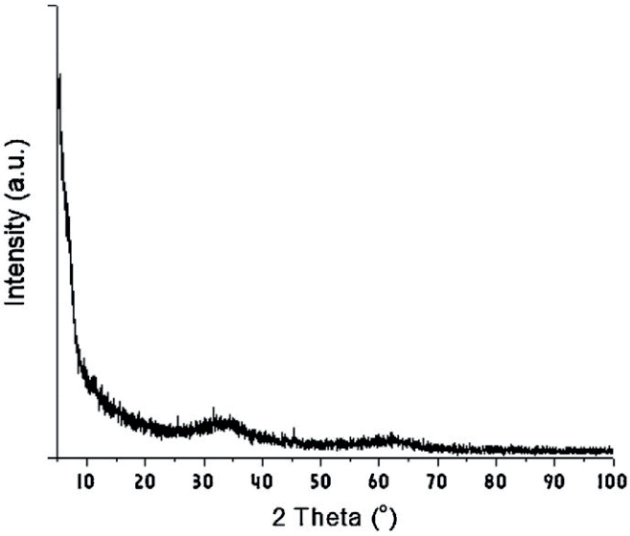


**FIGURE 3:** XRD pattern of DeHyS.

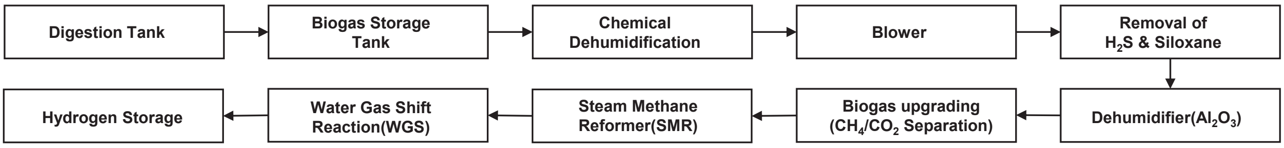


**FIGURE 2:** Schematic diagram of overall process for $H_2$ production through the removal of $H_2S$ and siloxane using DeHyS.

**TABLE 2:** $H_2S$ removal efficiency of DeHyS in various fields.

| Application site | Volume ($m^3$/d) | $H_2S$ requirement (ppm) | $H_2S$ removal Efficiency (%) | Adsorbent loaded ( $m^3$) | Space velocity ($hr^{-1}$) | Applications |
|---|---|---|---|---|---|---|
| Tancheon Water Regeneration Center | 40,000 | 10 | 99.99<br>(In: 902.50ppm,<br>Out: 0.00ppm) | 24 | 70 | Gas engine power generation,<br>Burner fuel |
| 2nd stage sludge fuel conversion facility in Sudokwon Landfill Site | 86,400 | 10 | 99.98<br>(In: 88.80ppm,<br>Out: 0.00ppm) | 25 | 144 | Burner fuel |
| 3rd stage sludge fuel conversion facility in the Sudokwon landfill site | 187,200 | 10 | 99.98<br>(In: 77.60ppm,<br>Out: 0.00ppm) | 40 | 195 | Burner fuel |
| Chilsung Energy Agricultural Corporation Cheongyang Plant | 12,000 | 10 | 99.99<br>(In: 389ppm,<br>Out: 0.00ppm) | 5 | 100 | Gas engine power generation |
| Yeoyang Farm | 3,600 | 30 | 99.96<br>(In: 4,450ppm,<br>Out: 1.4ppm) | 3 | 50 | Gas engine power generation |
| Hongcheon Eco-friendly Energy Town | 3,600 | 5 | 99.99<br>(In: 805ppm,<br>Out: 0.00ppm) | 8 | 19 | City gas production |
| Chungju Food Bio Energy Center | 8,000 | 5 | 99.99<br>(In: 950ppm,<br>Out: 0.01ppm) | 5 | 67 | Hydrogen production |
| IksanSewage Treatment Plant Food Biogasification Facility | 13,000 | 20 | 99.99<br>(In: 800ppm,<br>Out: 0.01ppm) | 5 | 108 | Transfer of drying facilities |
| Changwon Food biogas facility | 12,700 | 30 | 99.99<br>(In: 500ppm,<br>Out: 0.01ppm) | 20 | 26.5 | Gas engine power generation |

manure. On the other hand, at the Tancheon Water Treatment Center, an inflow concentration of 129.98 ppb based on total siloxanes was detected for the biogas derived from sewage sludge, and the removal efficiency was 99.45% with an outflow concentration of 0.71 ppb. In addition, the inflow concentration of 32.94 ppb based on total siloxanes was detected for the biogas derived from landfill gas in the Sudokwon landfill site, and the removal efficiency was 91.77% with an outflow concentration of 2.71 ppb (Table 3).

Based on the removal efficiency of $H_2S$ and siloxane obtained from the adsorption process using DeHyS as an adsorbent, it is concluded that during the adsorption process at ambient temperature, $H_2S$ should have been removed in the form of iron sulfide through a chemical reaction, while siloxanes are trapped in the pores through physical adsorption as shown in Figure 4 due to the well-defined pore characteristics of meso- and macro-pores (Magnone, E., Kim, S. D., Kim, G. S., Lee, K. H., & Park, J. H., 2020).

### 3.2 Results of field application for producing hydrogen

A representative site for producing hydrogen from domestic biogas is the Chungju Bio Green Hydrogen Charging Station located at 649-8 Bongbang-dong, Chungju-si, Chungcheongbuk-do. Approximately 8,000 $m^3$ of biogas per day in the anaerobic digester of the Chungju Bioenergy Center is produced from 80 tons/day of food waste generated in the Chungju area. In order to produce hydrogen from biogas, it is important to purify biogas before the reforming process. The DeHyS desulfurization agent and dry process system were applied to this facility.

At the beginning of the project, biogas was purified to upgrade to biomethane that can be supplied as fuel for city gas pipelines and vehicles. In May 2019, a hydrogen production process using biogas was introduced as the first model for the hydrogen economy by the Chungju Bio Hydrogen Charging Station Project (Figure 5).

The facility started commercial operation in March 2022, and currently produces about 500 kg of green hydrogen per day by biomethane reforming, and distributes it to nearby areas or sells it on site. This facility is equipped with fuel reforming system, hydrogen compression system, and hydrogen charging system that can accommodate all of the 700 bar hydrogen vehicle charging and 450 bar / 200 bar tube trailer charging (Figure 6).

The Chungju Bio Green Hydrogen Charging Station, which enables entire processes including waste treatment and hydrogen production through a series of processes, has demonstrated an innovative model that combines the role of both an on-site hydrogen refueling station and a mother station. Currently, It produces hydrogen directly from biogas, which is cheaper than natural gas. The cost of raw materials and distribution can be reduced, and hydrogen is being supplied at 7,700 won (about 5.7 US $),which is 9.1% lower than the national average unit price of hydrogen charging stations (as of August 30, 22) of 8,377 won (about 6.2 US $) per kg. Moreover, DeHyS as desulfuriza-

**TABLE 3:** Siloxane removal efficiency of DeHyS in various fields.

| Item | Yeoyang | | | Tancheon | | | Sudokwon | | |
|---|---|---|---|---|---|---|---|---|---|
| | In (ppb) | Out (ppb) | Remo-val. (%) | In (ppb) | Out (ppb) | Remo-val. (%) | In (ppb) | Out (ppb) | Remo-val. (%) |
| L2 | ND | ND | - | ND | ND | - | 2.87 | 2.46 | 14.28 |
| L3 | ND | ND | - | 0.08 | ND | - | ND | 0.06 | - |
| L4 | ND | ND | - | 0.40 | ND | - | 0.10 | ND | - |
| L5 | ND | ND | - | 0.10 | ND | - | ND | ND | - |
| D4 | 0.05 | 0.02 | 60 | 3.78 | 0.06 | 98.41 | 20.37 | 0.06 | 99.70 |
| D5 | 0.07 | 0.02 | 71.42 | 123.92 | 0.56 | 99.54 | 9.47 | 0.05 | 99.47 |
| D6 | 0.03 | 0.02 | 33.33 | 1.65 | 0.08 | 95.15 | 0.13 | 0.08 | 38.46 |
| Total siloxanes | 0.15 | 0.06 | 60 | 129.98 | 0.71 | 99.45 | 32.94 | 2.71 | 91.77 |

*ND: Not detected(< 0.01 ppb)*

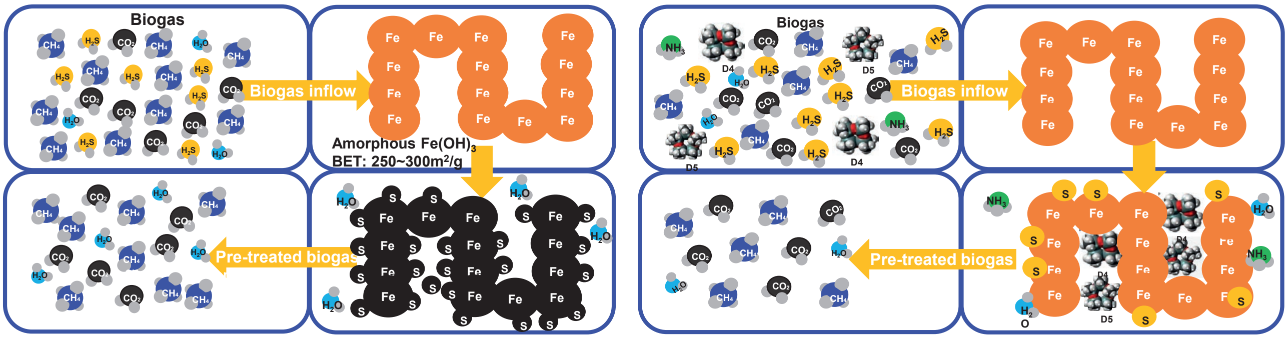


**FIGURE 4:** Schematic diagram of $H_2S$ and siloxane removal from biogas by meso/macro-porous adsorbent, DeHyS.

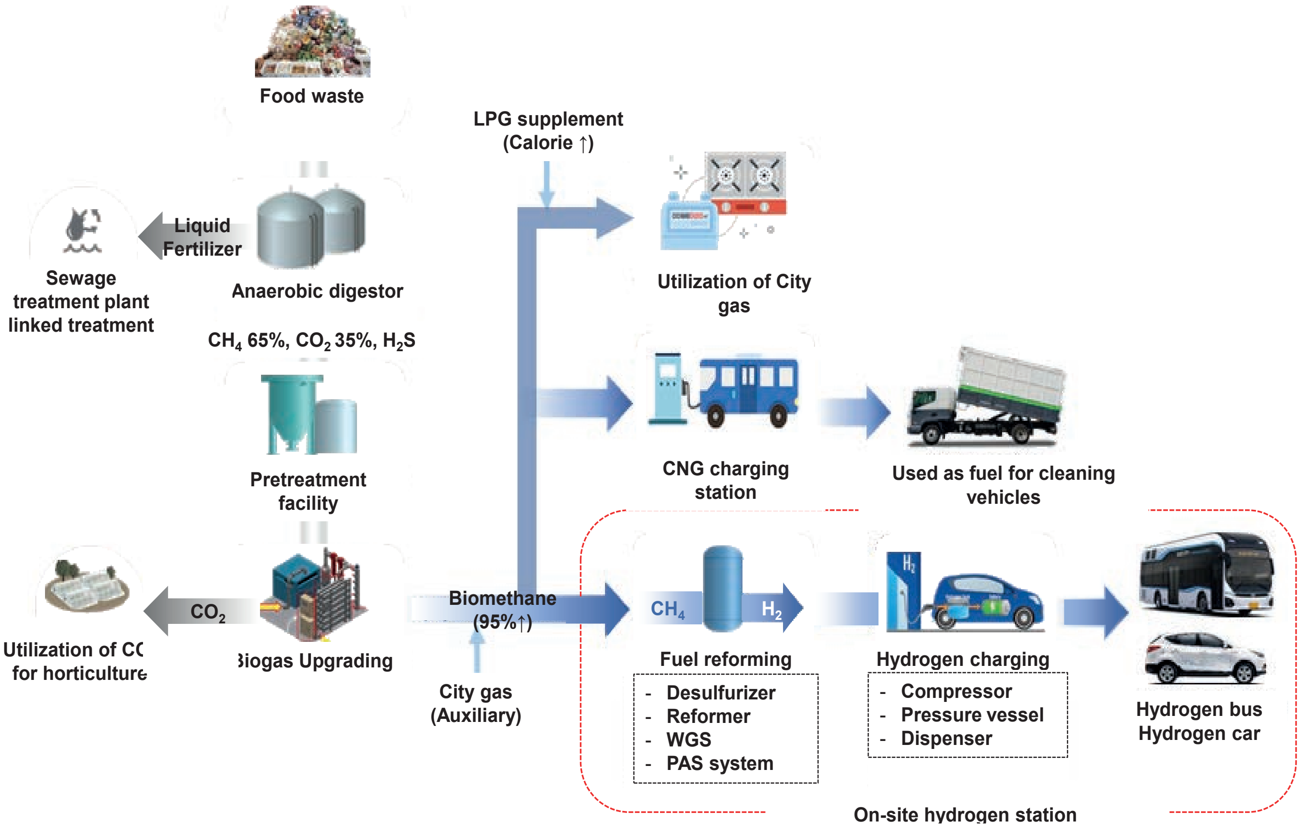


**FIGURE 5:** Schematic diagram of hydrogen production from biogas in Chungju.

Hydrogen extraction system

Tube trailer charging station

Hydrogen car charging station

**FIGURE 6:** The panorama of Chungju Bio Green Hydrogen Charging Station.

tion agent with extremely high removal efficiency for $H_2S$ and siloxanes is also highly propective in economic point of view due to its competitive price (e.g. 3.74 US $/L).

## 4. CONCLUSIONS

Biogas, one of renewable energy, is a key element necessary for a carbon-neutral policy and to build a hydrogen economy. In order to utilize biogas, impurity such as $H_2S$ must be removed firstly. The application of biogas varies depending on the concentration of $H_2S$.

In the Republic of Korea, the acceptable limit of $H_2S$ in biogas is classified, based on the concentrations of $H_2S$ and sulfur. In particular, it is known that the $H_2S$ standard is 0.01 ppm or less to prevent catalyst poisoning of the hydrogen reformer during the production of hydrogen from biogas.

The DeHyS is a desulfurization agent capable of reducing $H_2S$ from biogas to a concentration of 0.01 ppm or less, and it can also remove siloxanes simultaneously. Unlike conventional adsorbents using organic binder that require high temperature treatment to eliminate organic matter, the manufacturing process for DeHyS is very simple and highly economic due to simple drying process at mild condition. Furthermore, the $H_2S$ removal efficiency was known to be 99.9% or more, and it was known that simultaneous removal of 90% or more of the total siloxanes was possible. Therefore, it has shown to be higly effective for the stability of downstream equipment. It has already been applied to many sites for anaerobic digestion of various raw materials such as food waste, livestock manure, and sewage sludge.

In particular, it showed that the Chungju Bioenergy Center can produce 500 kg of green hydrogen per day through pretreatment and hydrogen reforming process of biogas produced from anaerobic digestion by merging about 60 tons of food waste and livestock manure per day. Currently, it is being supplied at a price more than 9.1% cheaper than hydrogen produced from natural gas.

As the expansion of the construction of hydrogen infrastructure in the future would increase the demand for green hydrogen produced from biogas, DeHyS with removal efficiency for $H_2S$ and siloxanes would be highly prospective in the economic point of view due to its competitive price.

## ACKNOWLEDGEMENTS

This study was conducted with support from the Ministry of SME (Green Venture Program for commercialization) and the Korea Environmental Industry Association (Overseas Export Business).

This work was also supported by Korea Institute of Energy Technology Evaluation and Planning(KETEP) and the Ministry of Trade, Industry & Energy (MOTIE) of the Republic of Korea (No.20193010160010).

## REFERENCES

IPCC. Special Report on Global Warming of 1.5°C. 2018. Retrieved from https://www.c40knowledgehub.org/s/article/IPCC-Special-Report-on-Global-Warming-of-1-5-C?language=en_US&gclid=EAIaIQobChMIr7uxh9L1-QIV2MFMAh1oTQJAEAAYAiAAEgKyhfD_BwE#RelatedKnowledge

Magnone, E., Kim, S. D., & Park, J. H. (2018). A systematic study of the iron hydroxide-based adsorbent for removal of hydrogen sulphide from biogas. Microporous and Mesoporous Materials, 270, 155–160. https://doi.org/10.1016/j.micromeso.2018.05.018

Magnone, E., Kim, S. D., Kim, G. S., Lee, K. H., & Park, J. H. (2020). Desiloxanation process of biogas using an amorphous iron hydroxide-based adsorbent: A comparison between laboratory and field-scale experiments. Chemical Engineering Research and Design, 163, 149–156. https://doi.org/10.1016/j.cherd.2020.08.026

Ministry of Environment of the Republic of Korea. Roadmap to expand bio-water energy using environmental facilities. 2022. Retrieved from https://eiec.kdi.re.kr/policy/ callDownload. do? num=227143 &filenum=2&dtime=20220621204205

Ryu, S. H., Seo, Y., Park, J., Kim, S. D., & Park, S. S. (2017). Adsorption Characteristics of Hydrogen Sulfide on Iron Hydroxide-based Adsorbent. Journal of Korea Society of Waste Management, 34(5), 468–473. https://doi.org/10.9786/kswm.2017.34.5.468

Solutions, E. A. E. (2000, October 31). Fuel Cell Handbook, Fifth Edition. OSTI.GOV Title Logo. https://www.osti.gov/biblio/769283

Li, Y., Wang, X., Mei, B., Wang, Y., Luo, Z., Luo, E., Yang, X., Shi, Z., Liang, L., Jin, Z. (2021) Carbon monoxide powered fuel cell towards H2-onboard purification. Sci. Bull. 66 , 1305–1311.

St-Pierre, J. (2010). PEMFC contaminant tolerance limit—CO in $H_2$. Electrochim. Acta, 55 , 4208–4211.

The Government of the Republic of Korea. Hydrogen Economy Roadmap. 2019. Retrieved from https://policy.nl.go.kr/search/searchDetail.do?rec_key=SH2_PLC20190231080&kwd=

The Government of the Republic of Korea. Carbon Neutral Promotion Strategy. 2020. Retreved form http://www.motie.go.kr/motie/ne/presse/press2/bbs/bbsView.do?bbs_seq_n=163606&bbs_cd_n=81&currentPage=1&search_key_n=&cate_n=&dept_v=&search_val_v=

UNFCCC. The Paris Agreement. 2015. Retrieved from https://unfccc.int/process-and-meetings/the-paris-agreement/the-paris-agreement

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# HAZARDOUS PROPERTIES OF MINERAL AND ORGANO-MINERAL PLASTIC ADDITIVES AND MANAGEMENT OF HAZARDOUS PLASTICS

Pierre Hennebert

*Ineris (French National Institute for Industrial Environment and Risks), BP 2, F-60550 Verneuil-en-Halatte, France*

**Article Info:**
Received:
12 December 2022
Revised:
12 April 2023
Accepted:
26 April 2023
Available online:
15 June 2023

**Keywords:**
Heavy metals
Heat stabilisers
Pigments agents
Flame retardants
Management by risk

## ABSTRACT

Many plastic additives are mineral or organo-mineral substances having functions as pigments, heat stabilizers, flame retardants, process adjuvants and the like. Are additivated plastics hazardous when they become waste? Data from the Plastic Additives Initiative, a joint industry and EU effort, was used, along with substance hazard statements from the ECHA website and hazard properties from the waste classification. 20 elements of 91 substances, namely Al, B, Ba, Bi, Cd, Co, Cr, Cu, F, I, Li, Mn, Ni, Pb, Pr, Sb, Sn, Ti, V and Zn were selected, and their additives used in 11 polymers, considered. Of the 91 substances selected, 57 are non-hazardous or are hazardous but used at too low concentration to render the plastic hazardous when it becomes waste. 34 substances (= 37% of 91) are hazardous and make plastics hazardous as waste. These are mainly heat stabilizers (for PVC), or pigments and flame retardants (for all polymers). The sorting of these plastics by the mineral concentration of their additives with online XRF is theoretically achievable. With data from previous papers, 63 additives (= 27% of 233) make plastic hazardous. The brominated flame retardants are the less documented. Only essential use should be allowed for pigments. Waste management today should focus on turning waste into non-waste, not waste leakage. With occupational safety and health regulations during processing, and with product regulations during its second life, the material should be managed as another hazardous or non-hazardous (virgin) raw material, and given end-of-waste status when it enters the loop.

## 1. INTRODUCTION

Additives are substances that improve the properties of plastics. Their use is generalised. They can be hazardous. More and more complete lists of substances used in plastic formulations are published and three of them are shortly presented in this introduction. It is underlined in these documents that numerous substances are hazardous, having as pure substance one or more hazard statement codes in the substance classification. The domain is so vast that the reader is invited to consult the available reviews and literature as in the journal Plastic Waste and Recycling.

European authorities are promoting progress towards a “toxics-free environment” (EC, 2020). Professionals fear that this "zero risk" approach is in fact a “hazard only” approach and entails significant difficulties for the circular economy, going as far as the impossibility of sorting waste fractions with very low concentration, losses of material in their loop and the costs of recycling which must devalue these fractions or incinerate or landfill these now non-recyclable fractions (EURIC, 2019). Waste could be treated by risk as products, according to REACH. It is therefore very important to develop applications in which materials containing a certain content of critical compounds can be used safely (Friege et al., 2021).

To progress towards a toxic-free environment or a risk-controlled circular economy, the substances must be prioritised. An early example is the report of Hanssen et al. (2013) of COWI consulting company, intended to be a brief handbook on plastic types and hazardous substances in plastics, their function, uses, concentration, release patterns, and alternatives, and focusing on 43 substances and families of substances. A review of POP substances, their occurrences in plastics and their potential management, including perfluorinated POPs, is presented in EC (2021).

One option is to restrict the use of hazardous additives according to the concept of “essential uses.” This concept has been applied in the context of the Montreal Protocol under which a use of a controlled substance should qualify as “essential” only if: (1) it is necessary for health and safety or

---

* Corresponding author:
Pierre Hennebert
email: pierre.hennebert@gmail.com

Detritus / Volume 23 - 2023 / pages 83-93
https://doi.org/10.31025/2611-4135/2023.17275

critical for the functioning of society (which encompasses cultural and intellectual aspects); and (2) there are no available technically and economically feasible alternatives or substitutes acceptable from the standpoint of environment and health (Weber et al., 2022). The concept of “essential uses” could also be applied to the management of other chemicals or groups of chemicals of concern, including hazardous or otherwise problematic additives that are not yet completely phased out. Only for those essential uses, where currently no substitute is available, should these additives be allowed in controlled material cycles. For uses that do not qualify as essential, hazardous additives should be substituted with safer alternatives (Weber et al., 2022).

Another option is to list all the substances that are hazardous. The three below-mentioned contributions are very large reviews of the substances that are used, and recommendations for their management to reduce (eco)toxic substances in the material flows.

(i) Aurisano et al. (2021) provide a list of more than 6 000 chemicals reported to be found in plastics and an overview of the challenges and gaps in assessing their impacts on the environment and human health along the life cycle of plastic products. They further identified 1 518 plastic-related chemicals of concern, which should be prioritized for substitution by safer alternatives. At last, the authors propose five policy recommendations for plastics:

- a transparent supply chain management
- a global and overarching regulatory framework for plastics and related chemicals, in support of a circular economy for plastics
- funds to invest in mechanisms to coordinate and support the transition of industries
- funds in research for efficient manufacturing of virgin and recycled plastics
- educate and support citizens, companies, and investor.

(ii) Wiesinger et al. (2021) investigate plastic monomers, additives, and processing aids on the global market based on a review of 63 industrial, scientific, and regulatory data sources. They identify +10 000 relevant substances and categorize them based on substance types, use patterns, and hazard classifications wherever possible. Over 2 400 substances are identified as substances of potential concern as they meet one or more of the persistence, bioaccumulation, and toxicity criteria in the European Union. Many of these substances are hardly studied (266 substances), are not adequately regulated in many parts of the world (1 327 substances) or are approved for use in food-contact plastics in some jurisdictions (901 substances). The possible ways forward are:

- establishing a centralized knowledge base
- ensuring transition to a safe and sustainable circular plastic economy (mainly information on and avoidance of hazardous substances)
- expanding and harmonizing regulatory efforts.

(iii) Weber et al. (2022) wrote an informative document for the UNEP Plastic Pollution conference (UNEP, 2022). It is a comprehensive review of all the aspects of plastics and chemicals in plastics, including capacity strengthening in particular in developing countries. With the two previous studies, they identified over 13 000 chemicals associated with plastics and plastic manufacturing across a wide range of applications, amongst which over 2 400 plastic monomers, additives and processing aids of potential concern based on their hazardous properties, with potentially significant adverse impacts. They claim access to information regarding the presence and quantity of chemicals in plastics, emissions and releases of chemicals from plastic products, as well as product use patterns. The authors rise the question of better capture of realistic exposure conditions in [hazard, exposure and risk] assessments, including exposure to chemical mixtures via multiple pathways, which would further enable science-based policy decisions that sufficiently safeguard human and environmental health.

This paper proposes, rather than lists of dangerous additives (hazard approach only), a risk approach: detecting the additives which make the plastic dangerous at their functional concentration and managing these plastics in controlled industrial loops (risk approach) so that the probability of exposure to hazards is very low, together with the phasing out substances of concern at the design stage.

Where to start identifying hazardous additivated plastics? In our opinion, reference methods like the CLP and the EU Waste classification should be used. That latter classification is derived from EU substances and mixture classification so-called CLP (Classification, Labelling and Packaging of chemicals and mixtures, EC, 2008) which is the aligned European version of the Globally Harmonised System (GHS) of the UN. The GHS is a single worldwide system for classifying and communicating the hazardous properties of industrial and consumer chemicals. The hazardous quality or not of the additivated plastic(s) depends on the properties of the additives and their concentration in the material. That later information is scarcer. Therefore, the plastics can be managed with such lists only as a first approach. It is proposed here to first assess the additives that renders plastic hazardous (according to the EU waste classification) when used at their functional concentration.

In this study, from an official list of 418 additives used in the European Union, mineral and organo-mineral additives with typically toxic or ecotoxic elements were selected, their hazard statement retrieved from the European Chemicals Agency site, and for the hazardous ones, their typical concentration in plastics (11 polymers) compared with the concentration limits making waste hazardous in the EU waste classification system.

This paper closes a series of four papers devoted to the hazardous additives in plastics: (i) brominated flame retardants (Hennebert 2021a), (ii) phosphorous, chlorinated, nitrogen and mineral flame retardants (idem 2021b), (iii) plasticisers (idem 2022a), and (iv) other mineral and organo-mineral additives (this paper). A synthesis of the hazard of the most important groups of additives is presented, and their management discussed, with a structured approach of the many possibilities of management. The objective of the waste regulation is discussed, as well as the opinion that, as soon as they enter in the loop of modern collection and industrial recycling, the additivated plastics, hazardous or not, should be managed as every raw material. The limitation of the quantities that can be present at a civic amenity site at the same time (1 tonne in France, not enough for profitable transport), the specific transport documentation, the special procedure for cross-border transport (not all countries not have all the technologies for the treatment of waste), the specific status of the recycling facilities, the undefined status after sorting and treatment are brakes and costs that limit the recycling of these materials.

### 1.1 Abbreviations

| | |
|---|---|
| ABS | Acrylonitrile butadiene styrene |
| AO | Antioxidants |
| AS | Antistatic |
| CAS no | Chemical Abstract Service number |
| CLP | Classification, Labelling and Packaging of chemical substances and mixtures |
| EC no | European Community number |
| ECHA | European Chemicals Agency |
| ELV | End-of-life vehicles |
| F | Filler |
| FC | Functional concentration (of an additive in a polymer) |
| FR | Flame retardants |
| H | Hazardous |
| Hxxx | Hazard statement code of a substance |
| HP | Hazard property of waste |
| HS | Heat stabilisers |
| HSC | Hazard statement code |
| L | Lubricant |
| NH | Non-hazardous |
| NIR | Near infra-red spectroscopy |
| NU | Nucleating agents |
| OBL | Obligations in EU regulations |
| OF | Other functions |
| OS | Other stabilisers |
| PA | Pigments agents |
| PAM | Polyamide (Nylon®) |
| PAI | Plastic Additive Initiative (joint action of the EU and industry) |
| PC | Polycarbonate |
| PET | Polyethylene terephthalate |
| PL | Plasticisers |
| PMMA | Polymethylmetacrylate |
| Polyolefin-I | Polyethylenes |
| Polyolefin-II | Polypropylenes |
| PS - (E)PS | (Expanded) polystyrene |
| PUR | Polyurethane |
| PVC (rigid) | Polyvinylchloride |
| PVC (soft) | Polyvinylchloride (softened) |
| REACH | Registration, Evaluation and Authorisation of Chemicals |
| WEEE | Waste of electrical and electronic equipment |
| XRF | X-ray fluorescence spectroscopy |

## 2. MATERIAL AND METHODS

### 2.1 Data

The Plastic Additives Initiative (PAI), a collaboration between the European Chemicals Agency - ECHA and the plastics industries delivered in 2019 a list of 418 additives currently used in products in the EU, along with their function(s), the polymer(s) they improve, and their functional concentration(s) (ECHA, 2021a). The excel file is no longer available, but the list of additives by function is available (with polymer and functional concentration) and a file can be easily reconstructed from the different screens of ECHA (2021). 20 elements were selected. For the 91 additives containing these 20 elements, the hazardous properties of human toxicity and ecotoxicity were collected from the ECHA open-access registration site of chemicals in the EU (ECHA 2021b). When their functional concentration is mentioned, it is compared with the concentration that makes a waste hazardous for the 15 hazard properties of waste (EU 2014, 2017).

### 2.2 Selection of the elements

Elements of the additives were computed from the chemical formula. This paper considers 20 elements, namely Al, B, Ba, Bi, Cd, Co, Cr, Cu, F, I, Li, Mn, Ni, Pb, Pr, Sb, Sn, Ti, V and Zn. The excluded elements are listed below. Chromium is always Cr (III) excepted in two pigment agents with Pb where it is Cr (VI) (Table 2). For sake of simplicity, these two substances have not been counted for Cr but only for Pb.

The following elements are not included in this study:

- 3 elements of the molecular organic skeleton: C, O, H;
- 5 elements of flame retardants (that are presented in in Hennebert 2021a, b): Br, Cl, N, P and Sb of $Sb_2O_3$ (a flame retardant synergist);
- 7 major (dominant in earth's crust) elements, being typically not the source of hazard if part of a hazardous substance: Ca, Fe, K, Mg, Na, S, Si.

Aluminium is as well a major element, but the aluminium hydroxide has hazard statement codes, and Al is used in organo-metallic compounds. Aluminium is noted in two cases "aluminum" in the ECHA data base (CAS 101357-30-6 Silicic acid, aluminum sodium salt, sulfurized and EC 939-582-4 Fatty acids, C16-18 (even numbered), aluminum salts). These names have not been changed here.

### 2.3 Properties of additives and classification of additivated plastic when it becomes waste

The hazard statement codes of additive substances were retrieved from the self-reported ECHA dossier (ECHA 2022). For some substances, the ECHA mentions its own "harmonised" classification, or indicates that a re-assessment is in progress. The hazard classification of the additivated plastics as waste is done according to the EU regulations (EU, 2014; EU, 2017) with maximum concentration for some properties and (weighted) summation of concentration for other properties (HP 4, HP 6, HP 8, HP 14). A synthesis of waste classification is presented in Hennebert (2019a). It has been supposed that only one additive is used in a plastic compound. The eventual other additives are not known and hence their properties and concentration have not been considered.

The functional concentrations are not always mentioned in the PAI file. When the concentration of one additive was mentioned as "n.a." (not available), the minimal and mean concentrations of the additives with the same function in the PAI file were used for that additive (Table in Hennebert, 2021b) . The function of the additive containing cobalt is not mentioned, has not been found in the literature and is assessed to "other functions".

Some special cases are the following:

- Differences in calculations of HP 14 with H400 for products and for waste: see (Hennebert 2021b). Only zinc distearate (CAS 557-05-1, Table SI 3) has the hazard statement code H400 but not H410 and has been classified by the product approach. A M-factor of 1 has been used since no data of acute ecotoxicity are available in the REACH dossier of that substance.
- $TiO_2$: it is assumed here that $TiO_2$ in plastics is not in free fine powder 10% < 10 µm and hence not H351 Carcinogenic level 2 but embedded in the plastic matrix.
- $Cr_2O_3$: 15% of notifiers have declared H360, Reprotoxic level 1. A waste is HP 10 if the concentration of H360 substance is > 0.3%. No substances of chromium (III) are classified in the harmonized classification and that approach is used here.

## 3. RESULTS AND DISCUSSION

The occurrences of the elements and their functions is presented by decreasing order in Table SI 1. If one substance contains two or more elements of the 20 elements considered here, it is counted in this table two times or more. Al, Zn, Cr (III) and Cu accounts together for half of the occurrences (56/112), and the pigments agents are the highly dominant function (73/112).

The stabilisers are the second group (heat stabilisation with 14 additives and other stabilisers with 4 additives) and are useful for the long-lasting of the products. The flame retardants (third group with 8 additives) are important for protection of humans, equipment, and infrastructures.

From this point of the paper, the additives are counted as individual substance. A multi-element additive is counted as one substance.

### 3.1 The 57 non-hazardous additives or non-hazardous plastics at the functional concentrations of hazardous additives

There are 57 non-hazardous additives or non-hazardous plastics at the functional concentrations of hazardous additives. These 57 substances are presented by substance in Table SI 2 and by elements and functions in Table 1. They are pigments agents (46), heat stabilisers (4), other stabilisers (2), lubricant (2), flame retardants (1), filler (1) and nucleating agents (1).

### 3.2 The 34 hazardous plastics at the functional concentrations of the hazardous additives

There are 34 hazardous additives that render plastics hazardous at their maximal functional concentrations, and 28 substances that render plastics hazardous at their minimal functional concentration. These 34 substances are presented by substance in Table SI 3 and by elements and functions in Table 2. There are 8 additives with Pb (including one with Cr (VI) and one with Cr (VI) and Mo), 5 with Sn, 5 with Zn (one with B), 3 with Cd (with other elements), 3 with Al, 2 with B, 2 with Sb (one with Mn and Ti), 2 with Cu (one with I) and 1 with Co, V, Li and Mn, respectively.

They are heat stabilisers (10), pigments agents (9), flame retardants (6), other functions (3), antistatic (2), antioxidants (1), other stabilisers (1), plasticisers (1) and lubricant (1).

Pigments agents is still the second group while it is the first group for non-hazardous additives or additives that don't render plastic hazardous. These 9 pigments agents contain 12 elements (Cd, Cr (VI), Cu, Mn, Mo, Pb, Sb, Se, Ti, V, Zn and Zr). With the principle of essential (and sober) use, maybe could the use of the hazardous pigments be reduced.

For these 20 elements of 91 substances, 26 substances (= 29% of 91) or 34 substances (= 37% of 91) make the plastic hazardous (Table SI 4).

#### *3.2.1 What polymers and what function(s)?*

Of the 34 additives that renders the plastics hazardous at maximal functional concentrations, the polymers in which these additives are used are available for 24 additives. Eleven different polymers are listed in the file (Table 3).

The most additivated polymers are PVC (soft and rigid), followed by polyolefin- and -II (PE and PP) and then PAM. Soft and rigid PVCs are additivated with many substances that makes them hazardous in the waste classification (20 and 18, respectively – Hennebert 2022a).

Four groups of additives can be distinguished:

- a group of 6 pigments agents and 1 flame retardant, antistatic, UV/light stabilizer (1) suitable for all the 11 polymers;
- a group of 10 heat stabilisers specific for PVC soft and rigid, including one for Polyolefin-I;
- a group of 5 substances lubricant, flame retardants, antistatic, other stabiliser and pigments agents of diverse polymers;
- 2 additives specific to PAM and PET, respectively.

#### *3.2.2 Ranking hazard properties of hazardous mineral and organo-mineral additivated plastics*

The prevalence of hazard properties at the functional concentration of these mineral and organo-mineral additives is presented in Table SI 4.

The most frequent hazard property is HP 14 'Ecotoxic': 41% (= 24/58) of the hazardous plastics are hazardous at least by HP 14. This agrees with the general finding that 50% of the hazardous waste are classified at least HP 14 when the M-factors are used, as in the product classification (Hennebert 2013, 2014). The second most frequent hazard property is HP 10 'Toxic for reproduction' (31% of the hazardous plastics = 18/58), with a maximal concentration of 0.3% for H360 substances, and 3% for H361 substances. The other properties are less frequent: HP 5

**TABLE 1:** The elements of the 57 additives that are not hazardous or that are used at concentrations lower than the concentration making the plastics hazardous (by decreasing occurrences of elements and function).

| **Number of substances** | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| **Element(s)** | **Pigments agents** | **Heat stabilisers** | **Lubricant** | **Other stabilisers** | **Nucleating agents** | **Flame retardants** | **Filler** | **Total** |
| Cu | 9 | | | | | | | 9 |
| Al | 4 | | 1 | 1 | | 1 | 1 | 8 |
| Zn | 2 | 3 | | | 1 | | | 6 |
| Cr (III) | 4 | | | | | | | 4 |
| Ba | 3 | | | | | | | 3 |
| Mn | 3 | | | | | | | 3 |
| Al P | 2 | | | | | | | 2 |
| F | 2 | | | | | | | 2 |
| Sn | 1 | 1 | | | | | | 2 |
| Ti | 2 | | | | | | | 2 |
| Bi V | 1 | | | | | | | 1 |
| Co | 1 | | | | | | | 1 |
| Co Al | 1 | | | | | | | 1 |
| Co Zn Al | 1 | | | | | | | 1 |
| Cr (III) Co | 1 | | | | | | | 1 |
| Cr (III) Cu | 1 | | | | | | | 1 |
| Cr (III) Ni | 1 | | | | | | | 1 |
| Cr (III) Sb Ti | 1 | | | | | | | 1 |
| Cr (III) W Ti | 1 | | | | | | | 1 |
| I | | | | 1 | | | | 1 |
| Ni | 1 | | | | | | | 1 |
| Sb Ni Ti | 1 | | | | | | | 1 |
| Zn Al | | | 1 | | | | | 1 |
| Zn Cr (III) | 1 | | | | | | | 1 |
| Zn Cr (III) Al | 1 | | | | | | | 1 |
| Zr Pr | 1 | | | | | | | 1 |
| **Total** | **46** | **4** | **2** | **2** | **1** | **1** | **1** | **57** |
| Total% | 81% | 7% | 4% | 4% | 2% | 2% | 2% | 100% |

'STOT', HP 4 'Irritant' for skin or eye (not relevant if embedded in a polymeric matrix), as well as HP 8 'Corrosive'. Three plastics are HP 7 'Carcinogenic' (maximal concentration 0.1% H350, 1% H351: diantimony trioxide (flame retardant), lead sulfochromate yellow and lead chromate molybdate sulfate red (pigments)), and one plastic is HP 11 'Mutagenic' (maximal concentration 0.1% H340, 1% H341; dibutyltin dilaurate (heat stabiliser)). All these substances are documented in Table SI 3.

## 3.3 Synthesis of assessment of additives that makes plastics hazardous

The results of this paper can be grouped with the assessment of brominated flame retardants (Hennebert, 2021a), other flame retardants (Hennebert, 2021b), and plasticisers (Hennebert, 2022a). The results of the assessment of these 233 additives are presented in Table 4. Brominated flame retardants are clearly less publicly documented than the other families, despite having the highest absolute and relative number of substances with on-going (re)assessment by ECHA. In total, 63 additives (= 27% of 233) make plastic hazardous at their maximum functional concentration, with the EU waste classification. Mineral and organo-mineral additives are the most numerous group that makes the plastic hazardous. The efforts towards a toxic-free environment (EC 2020) could focus first for plastic additives on these 63 substances, classified by using their functional concentrations mentioned by the producers or the importers and according to the reference methods of the EU, and hence not questionable. The progress will be probably easier than from large lists of 6 000, 10 000 or 13 000 substances (see Introduction) with unfortunately few data on actual use.

## 3.4 Sorting of the plastics with mineral and organo-mineral additives

Are the elements of these additives detectable by X-ray fluorescence? The lowest functional concentration of the

**TABLE 2:** The elements of the hazardous 34 additives that are used at concentrations higher than the concentration making the plastics hazardous.

| **Number of substances** | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| **Element(s)** | **Heat stabilisers** | **Pigments agents** | **Flame retardants** | **Other functions** | **Antistatic** | **Plasticisers** | **Lubricant** | **Antioxidants** | **Other stabilisers** | **Total** |
| Pb | 5 | | | 1 | | | | | | 6 |
| Sn | 5 | | | | | | | | | 5 |
| Zn | | | | | 1 | 1 | 1 | 1 | | 4 |
| Al | | | 3 | | | | | | | 3 |
| B | | | 1 | | 1 | | | | | 2 |
| Cd Se | | 1 | | | | | | | | 1 |
| Cd Zn | | 1 | | | | | | | | 1 |
| Cd Zr | | 1 | | | | | | | | 1 |
| Co | | | | 1 | | | | | | 1 |
| Cu | | 1 | | | | | | | | 1 |
| Cu I | | | | | | | | | 1 | 1 |
| Li | | | | 1 | | | | | | 1 |
| Mn | | 1 | | | | | | | | 1 |
| Pb Cr (VI) | | 1 | | | | | | | | 1 |
| Pb Cr (VI) Mo | | 1 | | | | | | | | 1 |
| Sb | | | 1 | | | | | | | 1 |
| Sb Mn Ti | | 1 | | | | | | | | 1 |
| V | | 1 | | | | | | | | 1 |
| Zn B | | | 1 | | | | | | | 1 |
| **Total** | **10** | **9** | **6** | **3** | **2** | **1** | **1** | **1** | **1** | **34** |
| Total% | 29% | 26% | 18% | 9% | 6% | 3% | 3% | 3% | 3% | 100% |

mineral additives is 0.1% corresponding to concentration of 0.01-0.08% of the element(s) of the additives. These concentrations are detectable with hand-held field or laboratory apparatus (about 10 mg/kg = 0.001% with some seconds of measurement). For on-line sorting instrument like Redwave (Redwave, 2022), the time of measurement is much shorter, and the limit of detection is much higher, being about 1.5% in practice. The efficiency of the sorting depends then on the distribution of the concentratiln in the plastic shreds: if it is bimodal with a non-additivated group with zero or close to zero concentration and an additivated group with a functional concentration > 1.5%, the sorting is efficient. An example for Br in cathode ray tubes shreds is given in Hennebert and Beggio (2021c).

Not considering the too light elements B and Li whom the additives are not detectable when B and Li are not associated with another heavier element, of the remaining 32 hazardous additives making plastic hazardous at maximal functional concentration (Table SI 5) , 17 have a minimal FC higher than 1.5% and 27 have a maximal FC higher than 1.5%. The corresponding elements of the 32 additives detectable by XRF are Al, Co, Cr, Cu, I, Mn, Pb, V and Zn. The sorting of these plastics by the mineral concentration of their additives is therefore theoretically achievable but the practicality (interference of dust, geometry of the shreds, presence of a coating layer) and the economic return of such operation should be checked. It is not practiced today to our knowledge.

### 3.5 Management of the POP, hazardous and non-hazardous plastics

A partial summary of the limitation of unintentional concentration of substances in products for POP substances and the four hazardous elements in WEEE is presented in (Hennebert, 2021). There are nevertheless exemptions in the POP regulation (EU, 2019). The limitations for four phthalate plasticisers (EU, 2018a) are summarised in (Hennebert, 2022a). A first step is phasing out substances of concern at design stage (EURIC 2019).

For hazard waste classification, the general method is synthetised in Hennebert (2019). Some POPs make the mixture that contain them hazardous (same reference). The hazardous waste classification has been applied to substitutive brominated flame retardants (BFR) in (Hennebert 2021a - Table 4), to Cl, P, N and some mineral flame retardants in (Hennebert 2021b – Table 3 to 6), to plasticisers in (Hennebert 2022a - Table 3) and to mineral additives in this paper (Table 3 and Table SI 3). In total 63 additives proposed in the EU are used in concentration that make the mixture hazardous (Table 4 of this paper). Waste of EEE and end-of-life vehicles (ELV) are hazardous; the parts containing POPs or that are hazardous must be separated during dismantling (EU 2018b).

The management of plastic as waste depends primarily on their concentration in regulated substances. If they con-

**TABLE 3:** The polymers and functions of 24 substances with documented functional concentration that make plastic hazardous, (no data of polymers for the other 12 substances) (FC = functional concentration; polymers: see abbreviation list; the colours illustrate the main features of the groups).

| Substance | Elt | Function | FC (%) | PVC (soft) | PVC (rigid) | Polyolefin-I | Polyolefin-II | PAM | PET | ABS | PUR | PC | IPS | PMMA | Total 11 polymers |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Group 1: Pigments (6) and Flame retardant, Antistatic, UV/light stabilizer (1) for 11 polymers** | | | | | | | | | | | | | | | |
| Cadmium zinc sulfide yellow | Cd | Pigments agents | 5 | x | x | x | x | x | x | x | x | x | x | x | 11 |
| Silicic acid, zirconium salt, cadmium pigment-encapsulated | Cd | Pigments agents | 5 | x | x | x | x | x | x | x | x | x | x | x | 11 |
| Cadmium sulfoselenide red | Cd | Pigments agents | 5 | x | x | x | x | x | x | x | x | x | x | x | 11 |
| Lead chromate molybdate sulfate red | Cr | Pigments agents | 5 | x | x | x | x | x | x | x | x | x | x | x | 11 |
| Lead sulfochromate yellow | Pb | Pigments agents | 1 | x | x | x | x | x | x | x | x | x | x | x | 11 |
| Manganese antimony titanium buff rutile | Sb | Pigments agents | 5 | x | x | x | x | x | x | x | x | x | x | x | 11 |
| Zinc oxide | Zn | Flame retardant; Antistatic; UV/light stabiliser; | 5 | x | x | x | x | x | x | x | x | x | x | x | 11 |
| **Group 2: Heat stabilisers of PVC (9) and PE (1 of the 9)** | | | | | | | | | | | | | | | |
| Pentalead tetraoxide sulphate | Pb | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| Tetralead trioxide sulphate | Pb | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| Dioxobtearateato)trilead | Pb | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| Sulfurous acid, lead salt, dibasic | Pb | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| Fatty acids, C16-18, lead salts | Pb | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| Methyl (Z,Z)-8,8-dibutyl-3,6,10-trioxo-2,7,9-trioxa-8-stannatrideca-4,11-dien-13-oate | Sn | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| 2-ethylhexyl 10-ethyl-4,4-dioctyl-7-oxo-8-oxa-3,5-dithia-4-stannatetradecanoate | Sn | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| 2-ethylhexyl 10-ethyl-4-[[2-[(2-ethylhexyl)oxy]-2-oxoethyl]thio]-4-octyl-7-oxo-8-oxa-3,5-dithia-4-stannatetradecanoate | Sn | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| Ethyl 9,9-dioctyl-4,7,11-trioxo-3,8,10-trioxa-9-stannatetradeca-5,12-dien-14-oate | Sn | Heat stabilisers | 2 | x | x | | | | | | | | | | 2 |
| Dibutyltin dilaurate | Sn | Heat stabilisers | 3 | x | x | x | | | | | | | | | 3 |
| **Group 3: Flame retardants, pigments agents, Lubricant, Antistatic, Other stabilizer, of diverse polymers (5)** | | | | | | | | | | | | | | | |
| Diantimony trioxide | Sb | Flame retardants | 8 | x | | x | x | x | | x | | | | | 5 |
| Hexaboron dizinc undecaoxide | B | Flame retardants | 0–3 - 0.4 | | | x | x | x | | | | | | | 3 |
| Aluminium hydroxide | Al | Flame retardants; Pigments agent | 0.–5 - 50.0 | | | x | | x | | | x | | | | 3 |
| Disodium tetraborate, anhydrous | B | Flame retardant; Antistatic; Other stabilisers | 5 | x | | x | x | | | | | | | | 3 |
| Zinc distearate | Zn | Lubricant | 0–5 - 1.0 | x | x | | | x | x | | | | | | 4 |
| **Group 4: Additives specific to PA (Other stabilisers) 51° and PET (function not available) (1)** | | | | | | | | | | | | | | | |
| Copper iodide | Cu | Other stabilisers | 0.5 | | | | | x | | | | | | | 1 |
| Cobalt bis(2-ethylhexanoate) | Co | (not available) | n.a. | | | | | | x | | | | | | 1 |
| **Total 24 substances** | | | | **20** | **18** | **12** | **10** | **12** | **9** | **8** | **8** | **7** | **7** | **7** | **118** |

**TABLE 4:** Synthesis of assessment of flame retardants, plasticisers and organo-mineral additives that makes the additivated plastics hazardous.

| Functions and additives (source of data) | Number of additives | Number of documented functional concentrations FC | Number of plastics hazardous at maximum FC (% of the number of additives) | Additives with on-going assessment by ECHA | Reference |
|---|---|---|---|---|---|
| Flame retardan–s - Brominated (main Producers catalogs + PAI*) | 41 | 4 | 4 (= 10%)*** | 12 | Hennebert 2021a,b |
| Flame retardan–s - Cl, P, N, Sb, B (P, Zn), Al (Na), Mg, Ca (PINFA** + PAI) | 32 | 16 | 8 (= 25%) | 5 | Hennebert 2021b |
| Plasticisers (PAI) | 69 | 47 | 17 (= 25%) | 8 | Hennebert 2022a |
| Mineral and Organo-mineral additives (PAI) | 91 | 74 | 34 (= 37%) | 2 | This paper |
| Total | 233 | 141 | 63 (= 27% of 233) | 27 | This paper |

** PAI = Plastic Additives Initiative*
*** PINFA = Phosphorus, Inorganic and Nitrogen Flame Retardants Association*
**** underestimated: 10 additives or blends without CAS number, 2 without dossier in ECHA, 10 without Hazard Statement Code in the dossier, 6 under reassessment by ECHA*

tain POP substances above Annex IV of POP regulation, the POP substances (and in practice the additivated plastics) must be "destroyed or irreversibly transformed". The PVC and PUR containing one of the four regulated phthalates with a concentration > 1000 mg/kg cannot be recycled. Hazardous plastics can be recycled.

The first step is the separate collection or the separation of plastics from other materials (eventually after shredding) of as much plastic as possible. Landfilling of plastics will be forbidden in 2025 in the EU, as any recyclable material (EC, 2014), according to the hierarchy of waste management (EU, 2008-2018). The management options for plastics with additives used in concentration making the plastic hazardous or not could be:

1. Re-use as article
2. Sorting if the wastes are mixed and recovery of the polymer or the additives of the sorted fraction
    (a) By colour: UV/visible detectors
    (b) By (additivated) polymer: X-ray transmission density, float/sink baths density, near infra-red (NIR) detectors (not effective with black plastics)
    (c) By polymer: float/sink baths density, near infra-red (NIR) detectors (not effective with black plastics)
    (d) By element of the additive(s): manual X-ray fluorescence (XRF), online XRF, and at laboratory stage by NIR (for instance Bonifazi et al., 2020)
3. Mechanical recycling: use in new product as additivated thermoplastic polymer if available in homogeneous prepared batches (from production falls, from selective demolition like window frames in PVC with lead stabiliser, pipes…, or from sorting systems) or sorted batch. This option is relevant for thermoplastics but not for thermosets (polyurethane, polyester, epoxy, silicone, rubber)
4. Chemical recycling: Purification of some polymer by selective dissolution and precipitation, or solvolysis (for instance CreaSolv® process of Fraunhofer Institute applied to brominated polystyrene and in pilot scale to plasticised PVC). Purification of the separated additive should be considered, as the hydrogenation of restricted phthalates of PVC
5. Chemical feedstock or fuel recovery from mixed plastics or sorted fractions: Pyrolysis or gasification and recovery of the liquid phase or the gas phase. Typically, the metallic elements remain in the char while the unwanted halogens are present in the liquid phase and in the gas phase
6. Element(s) recovery: Incineration with energy recovery and recovery of elements in ashes and fumes or fly ashes and air pollution control residues. For instance, Sb in Umicore copper smelter facility fed with WEEE plastics (Umicore 2022), and Br in ICL facility combined with the PolystyreneLoop facility (Polystyreneloop 2022)
7. Energy recovery: Incineration, typically prepared as solid recovered fuel for furnaces, or mixed in household waste or commercial waste in municipal solid waste incinerators
8. Landfilling of mixed plastics or sorted fractions
    (a) Plastics are not in the list of accepted waste and total organic carbon TOC must be lower than 3% in landfill for inert waste (EC, 2003) and organic material is restricted in landfill for hazardous waste (loss on ignition < 10% and total organic carbon < 6%, excluding in practice plastics)
    (b) Non-valorisable fractions in technical and economic conditions of the moment (like fluff of foam and textiles from shredding of automotives, and thermosets that do not be remelt and cannot be remoulded) are accepted in landfill for non-hazardous waste.

An additional case is the recovery of the elements of the electronic parts of the printed circuit boards (not the additive of the plastics), requiring the separation of the volatile phase of the plastics and the glass fiber part of the epoxy plates by pyrolysis, before being processed by a non-ferrous metal foundry. A discussion of waste, landfilling, sorting, mechanical recycling and chemical recycling of plastics is presented in (Hennebert, 2022a).

*Risk method for reusing and storing of non-hazardous plastics in a linear or circular economy*

The management of non-hazardous waste (frequently containing contaminants below the concentration limits which render the waste hazardous) is in practice carried out according to a risk approach and the resulting specific concentration limits must be observed for the disposal pathway or expected valorisation (Hennebert, 2022b). Concentration limits can apply to total concentration, be related to bioavailability (not defined for waste), or to leachable concentration. This approach is not specific to plastics. Research on the hazard and risk of plastic objects or (micro)particles (without or with additives) in the natural environment is active. Methods and concentration limits for polymer debris entering the terrestrial environment (mainly tire wear in traffic dust and synthetic textile fiber emitted during washing and present in sewage sludge used for land fertilisation) should be developed, based on scientific evidence and a risk approach. In the latter case, the conflict of objectives (protect the soil/recycle nutrients) must be arbitrated by the data, and technological solutions such as a filter at the exit of washing machines (compulsory in France in 2025 for new machines, RF 2020) must be promoted to resolve the conflict.

*Risk method for management of hazardous plastics in a circular economy*

The waste status should be revisited in the circular economy. 'Waste' means any substance or object which the holder discards or intends or is required to discard (EU Waste Framework Directive, 2008-2018). Waste has a legal status which aims to avoid the risks for the environment and public health if it is abandoned. The definition is based on the act of discarding, rather than the value of the material (Johansson and Forsgren, 2020). The main aim of waste management besides waste evasion should be today turning wastes to non-wastes (Pongrácz, 2002; Pongrácz and Pohjola, 2004). It is understood today that the primary objective of waste legislation is to control the fate of waste to achieve a high level of protection of human and the environment (Johannson, 2022) so that, with a toxicological and ecotoxicological formulation, the exposure of these targets to wastes contaminants is avoided and the hazard does not produce a risk.

This is meaningful in linear economy. In the linear economy, landfilling and even worse littering with the spreading of contaminants was the natural fate of waste, and an extensive regulatory system is needed to keep (mobile) contaminants tight in landfills, after eventual incineration, to avoid human and environmental exposure to contaminants of the waste. The material must be managed by its hazardous properties and the waste regulations (Hennebert, 2022b).

In circular economy, waste is disposed of in collection systems and treated in controlled modern industrial loops, so that no human and environmental exposure occurs. Only a small fraction of the material is unused and becomes waste. Accordingly, "... an object should only be considered waste, i.e. make waste legislation applicable, where necessary to protect human health and the environment. Conversely, objects that can be used safely without governance in the form of waste legislation should be considered something else" (Johannson, 2022). This author asks to "keep it simple". Other authors proposed previously a status of "certified material" (Johansson and Forsgren, 2020). In fact, the material is managed by the occupational safety and health regulations and industrial regulations during the processing, and by the products regulations during its second life. The specific demanding regulatory requirements for collection, grouping and transport of hazardous waste are shaped to minimise risk to human health and the environment, namely abandonment in the natural environment. Hazardous waste management by risk is proposed by Bodar et al. (2018). Waste could be treated by risk as products, according to REACH. Applications in which secondary materials including a certain content of critical compounds can be used safely should be developed (Friege et al. 2021). The interface between chemicals legislation and waste legislation should be as close as possible to achieve a circular economy (Friege et al., 2022; Hennebert, 2022b). In our opinion, the simplest thing is that, as soon as it enters the loop of modern collection and recycling, the material is managed like any raw material, and benefits from the status of end of waste. Another option is to create a new status for "secondary raw materials" in the European waste legislation to move away from the stark dichotomy between "waste" and "products" status for processed waste meeting industry specifications or quality standards, without prejudice to existing end-of-waste criteria, to level the playing field with primary materials, both in terms of regulatory constraints and public perception (EURIC, 2019).

## 4. CONCLUSIONS

This paper proposes, rather than lists of dangerous additives (hazard approach only), a risk approach: detecting the additives which make the plastic dangerous at their functional concentration and managing these plastics in controlled industrial loops (risk approach) so that the probability of exposure to hazards is very low, together with the phasing out substances of concern at the design stage.

Of 91 mineral and organo-mineral additives assessed in this paper, 34 additives make plastic hazardous. There are mainly heat stabilisers (10), pigments agents (9) and flame retardants (6). Two additives are under assessment by ECHA. For 24 additives, the polymers in which they are used is known. These substances should be further investigated in plastic loops. With the data of three previous papers on brominated flame retardants, other flame retardants and plasticisers, 63 additives (= 27% of 233) make plastic hazardous. The brominated flame retardants are the less documented. Only essential use should be allowed for pigments.

The sorting of the plastics with mineral and organo-mineral additives with minimal detectable concentration by online XRF could use Al, Co, Cr, Cu, I, Mn, Pb, V and Zn concentrations that are present in 32 additives making plastic hazardous at their maximal functional concentrations. The sorting of these plastics by the mineral concentration of their additives is therefore theoretically achievable.

EU waste regulation is designed to avoid human and environmental exposure to contaminants. That exposure occurs typically in linear economy by littering and secondarily by incorrect landfilling. Waste has a legal status which aims to avoid the risks for the environment and public health if it is abandoned. The definition is based on the act of discarding, rather than the value of the material. The main aim of waste management besides waste leaks should be today turning wastes to non-wastes.

In the circular economy, waste is collected in modern systems and treated in controlled industrial loops, so that there is virtually no human and environmental exposure (the probability of exposure to the hazard, i.e. the risk, is very weak). With occupational safety and health regulations and industrial regulations during processing, and with product regulations during its second life, the material must be managed as another hazardous or non-hazardous raw material (virgin), and benefit of the end-of-waste status as it enters the loop.

## ACKNOWLEDGMENTS

This work has been funded by the French Ministry of Ecological Transition and Ineris.

## REFERENCES

Aurisano, N., Weber, R., & Fantke, P., 2021. Enabling a circular economy for chemicals in plastics. Curr. Opin. Green Sustain. Chem. 31, 100513. https://doi.org/10.1016/j.cogsc.2021.100513

Bodar C, J Spijker, J Lijzen, S Waaijers-van der Loop, R Luit, E Heugens, M Janssen, P Wassenaar, T Traas. 2018. Risk management of hazardous substances in a circular economy. Journal of Environmental Management 212,108e114. https://doi.org/10.1016/j.jenvman.2018.02.014

Bonifazi G, Fiore L, Gasbarrone R, Hennebert P, Serranti S. 2021. Detection of brominated plastics from e-waste by short wave infrared spectroscopy. Recycling 2021, 6, 54. https://doi.org/10.3390/recycling6030054

Hansen Erik, Nils H. Nilsson, Delilah Lithner, Carsten Lassen 2013. COWI 2013. Hazardous substances in plastic materials. TA 3017 2013. 151 p. https://www.byggemiljo.no/wp-content/uploads/2014/10/72_ta3017.pdf

EC 2003. Decision 2003/33/EC Council decision of 19 December 2002 establishing criteria and procedures for the acceptance of waste at landfills pursuant to Article 16 of and Annex II to Directive 1999/31/EC. OJEC L 11/27. 16.01.2003

EC 2008 (CLP). Regulation EC No 1272/2008 of the European Parliament and of the Council of 16 December 2008 on classification, labelling and packaging of substances and mixtures, amending and repealing Directives 67/548/EEC and 1999/45/EC, and amending Regulation (EC) No 1907/2006. 31.12.2008 Official Journal of the European Union L 353/1. http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32008R1272&rid=1 . List of substances: EC 2018 (CLP). Annex A spreadsheet file of EU classified substances https://echa.europa.eu/information-on-chemicals/annex-vi-to-clp

EC 2014. COM(2014) 398 final. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions. Towards a circular economy: A zero waste programme for Europe. 02.07.2014. https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:52014DC0398&qid=1670408883705&from=EN

EC 2020. COM(2020) 667 final. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions. Chemicals Strategy for Sustainability Towards a Toxic-Free Environment. 14.10.2020. https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:52020DC0667&qid=1670346524865&from=EN

EC 2021. European Commission (Directorate general for Environment), Bio Innovation, INERIS, RPA 2021. Study to support the assessment of impacts associated with the review of limit values in waste for POPs listed in Annexes IV and V of Regulation (EU) 2019/1021. April 2021. KH-06-21-119-EN-N. 596 p. doi:10.2779/63162 . https://op.europa.eu/en/publication-detail/-/publication/c7c70b73-3798-11ec-8daf-01aa75ed71a1/language-en

ECHA 2021. Plastics additives initiative. Accessed 08/2021. https://echa.europa. eu/ fr/ plastic- additives-initiativehttps://echa.europa.eu/mapping-exercise-plastic-additives-initiative. For detailed list: https://echa.europa.eu/fr/mapping-exercise-plastic-additives-initiative

ECHA 2022. Information on chemicals. Accessed 2022. https://echa.europa.eu/information-on-chemicals

EU 2008-2018. Waste Framework Directive 2008/98/EC modified by EU directive 2018/851/EC. Consolidated version. Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain Directives. OJ L 312, 22.11.2008, p. 3

EU 2014. Commission Decision 20142014/955/EU of 18 December 2014 amending Decision 2000/532/EC on the list of waste pursuant to Directive 2008/98/EC of the European Parliament and of the Council. https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32014D0955&qid=1628498956448&from=EN-http ://eurlex.europa.eu/legalcontent/EN/TXT/PDF/?uri=CELEX :32014D0955&rid=1

EU 2017. Council Regulation (EU) 2017/997 of 8 June 2017 amending Annex III to Directive 2008/98/EC of the European Parliament and of the Council as regards the hazardous property HP 14 'Ecotoxic'. Official Journal of the European Union. 14.6.2017. L 150/1. https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32017R0997&rid=1

EU 2018a. Commission Regulation 2018/2005 of 17 December 2018 amending Annex XVII to Regulation (EC) No 1907/2006 of the European Parliament and of the Council concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) as regards bis(2-ethylhexyl) phthalate (DEHP), dibutyl phthalate (DBP), benzyl butyl phthalate (BBP) and diisobutyl phthalate (DIBP) (Text with EEA relevance.)

EU 2018b. Directive (EU) 2018/849 of the European Parliament and of the Council of 30 May 2018 amending Directives 2000/53/EC on end-of-life vehicles, 2006/66/EC on batteries and accumulators and waste batteries and accumulators, and 2012/19/EU on waste electrical and electronic equipment. OJ L 150, 14.6.2018, p. 93–99. https://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri=CELEX:02012L0019-20180704&qid=1670272941653&from=EN

EU 2019. Regulation (EU) 2019/1021 of the European parliament and of the Council of 20 June 2019 on persistent organic pollutants. Official Journal of the European Union, L 169, p. 45-77, 25.06.2019. https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32019R1021&qid=1628498569794&from=EN

EURIC 2019. The top 5 priorities of the recycling industry for the period 2019-2024. 8 p. https://euric-aisbl.eu/index.php?option=com_content&view=article&id=163:top-5-priorities-of-the-recycling-industry-for-the-period-2019-2024&catid=16:position-papers&Itemid=116

Friege H, B Zeschmar-Lahl, B Kummer, J Wagner. 2021. The new European database for chemicals of concern: How useful is SCIP for waste management? Sustainable Chemistry and Pharmacy 21, 100430. https://doi.org/10.1016/j.scp.2021.100430

Hennebert P, Papin A, Padox J-M, Hasebrouck B. 2013. The evaluation of an analytical protocol for the determination of substances in waste for hazard classification. Waste Management 33 (2013) 1577–1588. DOI 10.1016/j.wasman.2013.03.013

Hennebert P, van der Sloot, H.A., Rebischung, F., Weltens, R., Geerts, L., Hjelmar, O., 2014. Hazard property classification of waste according to the recent propositions of the EC using different methods. Waste Manag. 34, 1739–1751. DOI 10.1016/j.wasman.2014.05.021

Hennebert P. 2019a. Hazard classification of waste: review of available practical methods and tools. Detritus. 07, 13-28. https://doi.org/10.31025/2611-4135/2019.13846

Hennebert P. 2019b. Proposition of a threshold for waste contaminated with mercury (compounds) in application of the Minamata Convention on Mercury and impact assessment. Detritus, Volume 06 – 2019, pages 25-31. https://doi.org/10.31025/2611-4135/2019.13822

Hennebert P. 2020. Concentrations of brominated flame retardants in plastics of electrical and electronic equipment, vehicles, construction, textiles and non-food packaging: a review of occurrence and management. Detritus. DOI 10.31025/2611-4135/2020.13997 https://digital.detritusjournal.com/issue/volume-12--september-2020/363

Hennebert P. 2021a. The substitution of regulated brominated flame retardants in plastic products and waste and the declared properties of the substitutes in REACh. Detritus 16 – 2021, 16-25. https://doi.org/10.31025/2611-4135/2021.15122

Hennebert P. 2021b. Hazardous properties of brominated, phosphorus, chlorinated, nitrogen and mineral flame retardants in plastics which may hinder their recycling. Detritus, 7 – 2021, 49-57. https://doi.org/10.31025/2611-4135/2021.15142

Hennebert P, Beggio G. 2021c. Sampling and sub-sampling of granular waste: size of a representative sample in terms of number of particles. Detritus 17 – 2021, 1-12. https://doi.org/10.31025/2611-4135/2021.15139

Hennebert P. 2022a. Hazardous properties of plasticisers which may hinder the recycling of plastics. Submitted15 p.

Hennebert P. 2022b. Risk management of hazardous solid waste according by hazardous property including mercury contaminated wastes. Detritus, 20 – 2022, 78-89. https://doi.org/10.31025/2611-4135/2022.15212

Pongrácz E. 2002. Re-defining the concepts ofwaste andwaste management: Evolving the Theory ofWaste Management. Doctoral dissertation. Department of Process and Environmental Engineering, University of Oulu, Finland, 2002. http://www.herkules.oulu.fi/isbn9514268210/

Pongrácz E, Pohjola V J, 2004. Re-defining waste, the concept of ownership and the role of waste management. Resources, Conservation and Recycling 40 (2004) 141–153

Johannson O. 2022. Did end of waste bring the end of waste? Editorial. Detritus, 20 – 2022, 1-2. https://doi.org/10.31025/2611-4135/2022.15220

Johansson N, C. Forsgren. 2020. Is this the end of end-of-waste? Uncovering the space between waste and Products. Resources, Conservation & Recycling 155 (2020) 104656. https://doi.org/10.1016/j.resconrec.2019.104656

Polystyreneloop 2022. https://polystyreneloop.eu/

Redwave 2022. https://redwave.com/en/products/redwave-xrf

RF 2020. République Française. LOI n° 2020-105 du 10 février 2020 relative à la lutte contre le gaspillage et à l'économie circulaire (1). JORF n°0035 du 11 février 2020. https://www.legifrance.gouv.fr/eli/loi/2020/2/10/TREP1902395L/jo/texte

Umicore 2022. https://pmr.umicore.com/en/

UNEP 2022. https://www.unep.org/events/conference/inter-governmental-negotiating-committee-meeting-inc-1

Weber Roland, Narain M. Ashta, Nicolò Aurisano, Helene Wiesinger, Zhanyun Wang, Magali Outters, Martin Schlummer, Markus Blepp, Helena Andrade, Martin Scheringer, Peter Fantke. 2022. Chemicals in plastics: technical report. Draft 28 July 2022. 143 p.

Wiesinger, H., Wang, Z. & Hellweg, S. Deep Dive into Plastic Monomers, Additives, and Processing Aids. Environ. Sci. Technol 2021, 55, 9339-9351. acs.est.1c00976 (2021); https://doi.org/10.1021/acs.est.1c00976

Extra contents

# COLUMNS AND SPECIAL CONTENTS

This section comprises columns and special contents not subjected to peer-review

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

Environmental Forensic

# RISK ANALYSIS AND LIFE CYCLE ASSESSMENT IN ENVIRONMENTAL FORENSICS: PRACTICAL INSTRUCTIONS FOR THEIR USE

Alberto Pivato [1], George Varghese [2], Francesco Di Maria [3], Alessandro Manzardo [1], Massimiliano Lega [4], Maria Cristina Lavagnolo [1], Maria Pettersson [5] and Claire Gwinnett [6]

*[1] Department of Civil, Environmental and Architectural Engineering, University of Padova, via Marzolo 9, 35131, Padova, Italy*
*[2] National Institute of Technology Calicut, Kerala, India*
*[3] Department of Engineering, Università di Perugia, via G. Duranti 67, 06125 Perugia, Italy*
*[4] Massimiliano Lega, Napoli Isola C4, 80143, Italy*
*[5] Department of Business Administration, Technology, Art and Social Sciences, Luleå*
*[6] Centre for Crime, Justice and Security, Staffordshire University, United Kingdom*

## 1. INTRODUCTION

In the environmental field, we are often faced with problems that are so complex that it is necessary to find methodologies which combine the available information in a manner which allows for effective, logical, transparent and reproducible interpretation which could be qualitative or quantitative. In some cases, there is limited information present or only some information is actually provided; this is the case, for example, of the potential or ongoing environmental impacts associated with a product, process, activity, agent (such as pollutants in different media), or an event (e.g. failure of the bottom liner of a landfill). Risk analysis (RA) and life-cycle assessment (LCA) are the two dominant methods to aid in collating information from these scenarios and enabling decision to be made in the environmental field.

The need to understand the state of the environment in order to make assessments, conclusions and decisions requires elevating the concept of knowledge to that of situational awareness. Situational awareness represents one of the main objectives of any investigative process and, more generally, of environmental forensic engineering and, certainly, LCA and RA represent two valid tools for achieving such goal. In particular, among the most critical aspects of an investigative process is that of revealing the link between causes and effects, both from a logical and physical point of view; LCA and RA relate all elements through logical-conceptual models, supporting the objectification of analysis processes and ensuring the transparency and repeatability of processes by multiple subjects as required by laws and regulations.

Environmental forensic represents an area where the decisions and conclusions have significant financial, legal and social implications. Decisions in environmental forensic cases are normally conducted within the criminal justice framework or equivalent, meaning that they must be transparent and robust. RA and LCA, as previously reported, have these properties and therefore have potential benefits for use in environmental forensics.

The tools of RA and LCA have significant potential for answering key questions posed in environmental forensic scenarios (Ram, 2000), including;

- What was the source of the contamination? Environmental forensic experts may use a combination of analytical and transport modelling techniques to identify from where the chemicals responsible for pollution, came from.
- When did the release occur? With similar techniques as above, environmental forensic experts can assess when the release of uncontrolled pollutants in the environment occurred and how long the event lasted (whether prolonged, short duration or a one-time event). In this context it is important to also understand what historical industry practices and regulatory practices were in place at the time the released occurred and if an insurance coverage was available.
- How did the release occur? Answering this question involves understanding the mechanisms of transport and pathways through which the contaminants were released into the environment. At this stage, it may also be important to analyse the system's reliability, which refers to the ability of a system to consistently perform its intended function over a specified period of time under normal operating conditions (for example the bottom liner of a landfill which prevent the leakage of leachate).
- Who contributed to the problem? In many cases, multiple parties may have contributed to the problem. These may include specific individuals, companies, or government agencies that were involved in activities such as

Managing editors:
Alberto Pivato, Claire Gwinett, George Varghese
email: alberto.pivato@unipd.it, c.gwinnett@staffs.ac.uk, gkv@nitc.ac.in

Detritus / Volume 23 - 2023 / pages I-VI
https://doi.org/10.31025/2611-4135/2023.18279

industrial operations, waste disposal, or transportation of hazardous materials.
- What is the extent and magnitude of the contamination? Here it may be important to know the size of the spread of the contaminant in the environment and whether it degrades or bio-magnifies after entering the food chain. Any synergistic effects in the presence of other chemicals may also become crucial.
- What is the potential risk to human health and the environment? To be answered, this question needs detailed analysis of the release of potential contaminants in the different environmental matrices and consequently their environmental concentrations observed/modelled. Then the related possible exposure paths for the biota compartment (e.g. respiration, contact, ingestion) are analysed to assess the impact on human health and ecosystem.
- How much will the pollution cost? The total pollution cost includes two components: the damage cost and the remediation cost. The damage cost is the compensation to be paid for the death of, or injury to any person or damage to any property or environment. The remediation cost is the amount required to reinstate the environment to its pre-pollution state.
- What is the best strategy for clean-up and remediation? The strategy adopted for clean-up must be suitable for the contaminant and the polluted environmental matrix. The cost involved, disturbance caused to the soil, long-term effectiveness, time requirement, etc. are important considerations.
- How should the costs be allocated amongst the responsible parties? Environmental forensic investigations involve identifying the parties responsible for pollution and, determining the share of each party towards the total pollution cost.

Issues of risk and uncertainty are at the centre of large parts of environmental regulations. With the help of environmental regulation, we can ensure that risks are identified and assessed, and that measures to manage the risks are taken. Legal rules can also govern which measures are taken by requiring that the best possible technology be used. Risk assessments are also important as a starting point for determining safe actions and condition e.g. in obtaining a permit for environmentally hazardous activities. Risk assessments however also feed into the actual law-making process, as these types of assessments are important when it comes to deciding on what regulation to adopt, e.g. regulations on hazardous substances. From this perspective, the risk assessment that forms the basis for the environmental legislation will also contribute to direct the use of limited resources against the most significant risks (see e.g., Russel and Gruber, 1987).

Currently in environmental regulation it is important to assess the impact of a product/service/etc. not only considering their use but the whole life cycle. In addition, consideration should be given to the different environmental aspects (such as global warmings; biodiversity loss; ocean acidification; etc.) with the same assessment methodology. This is the case of a life cycle assessment.

The current column aims at investigating the applicability of RA and LCA in the environmental forensic field whilst highlighting the special characteristics of each method.

## 2. ANALYSIS OF THE TWO DECISION TOOLS

In the following paragraphs a brief description of the two tools is presented and the similarities and differences are compared. In Table 1, the detailed characteristics of RA and LCA are reported.

### 2.1 Risk Analysis (RA)

The mathematical definition of risk is mutable in different fields of applications (volcanology, seismic analysis, woodland burn, transport, ecological, nuclear, chemistry, industrial and sanitary engineering, etc.) and with proper care we need to apply the correct definition to each different field (Glickman, 1990; Asante-Duah, 1998; Salandin, 2001).

A general quantitative definition for the Probabilistic Risk Assessment defines the risk as as the product of a frequency times the magnitude of the events ‘ (Rasmussen, 1981; Asante-Duah, 1998; Glickman and Gouch, 1990; Paustenbach, 2002):

$$R = F \times M$$

where:

- R is the risk of the system (consequences/ unit time);
- F is the frequency that an adverse event can happen (event/ unit time);
- M is the magnitude of the consequences of the event (consequences/event);

Sometimes it is more useful to use the probability (P(H)) that an adverse event of a determined intensity can happen in a specific period of time (Varnes, 1984) rather than the frequency of events by year (F). In this case the previous definition becomes:

$$R = P(H) \times M$$

Therefore, the tool is rooted in two analytical approaches: probability theory and methods for identifying causal links between adverse health effects and different types of hazardous events/activities.

Environmental risk can be clearly distinguished from ecological and human health risk. According to EPA, ecological risk assessment (ERA) is the process “that evaluates the likelihood that adverse ecological effects may occur or are occurring as a result of exposure to one or more stressors” [EPA, 1996] and in general it can be considered a systematic process to organize and analyse data, information, assumptions and uncertainties for the purposes of evaluating the probability that some adverse effects to the analysed ecosystem will take place [Suter, 1993].

The term “adverse” is understood as a negative alteration to the structural and/or functional features of the ecological system examined. Conversely, the term “stressor” refers to an unwanted human action (of chemical, physical or biological type) leading to an unfavourable effect.

Health risk analysis aims at investigating the effects on humans. Risk analysis can distinguish between the effects of a dangerous event in a determined point, hypothesizing the potential presence of a human (individual risk) and the negative effects on the general population in the area of study (diffuse or collective risk).

In the last few years, human health risk analysis has developed further, often hand in hand with ecological risk analysis, as most pollutants, known to have an impact on the ecosystem, have also impacts on human health and vice versa. Nevertheless, it has been shown that a lot of contaminants (ammonium, chlorine, some pesticides, etc.) that have minimal effect on human health, can cause serious damage to aquatic organisms.

Finally, a distinction that should be highlighted is between "predictive risk assessment" and "retrospective risk assessment" (Asante-Duah, 1998; Erskine, 1997; Glickman and Gouch, 1990; Henley and Kumamoto, 1981; Paustenbach, 2002; Rasmussen, 1981; Suter 1993). In the first case, the analysis refers to the effects that could occur with the occurrence of adverse events. In the second case, the analysis is referred to environmental effects due to an event that has already occurred.

The procedure of "predictive risk assessment" involves structuring the risk assessment into two distinct phases. One phase assesses the probability of system failure, and a second phase predicts the effects on humans and/or the environment.

The procedure of "retrospective risk assessment" initially involves verifying through environmental monitoring the release of pollutants, for example from a landfill system, and assessing the level of contamination of adjacent environmental matrices.

There are some examples in the scientific literature of advanced environmental monitoring methods that also support forensic activities and, above all, allow to obtain quantitative results, useful for both methods (Persechino et al., 2013, Di Fiore et. Al, 2017).

The evaluation of damage to humans or the ecosystem resulting from the contamination is the next stage of analysis.

The full operating methodology subdivides the risk assessment into the following phases:

- **System Reliability:** it is the study of the probability of system failure (for example, a bottom liner break of a landfill causing leachate leakage; break of the air pollution control system causing the uncontrolled emissions of contaminants from the stack of a plant; etc.) by means of non-deterministic techniques. If the adverse event (failure of the system) has already happened, this phase is not explicitly required. This phase is present only in the "predictive risk assessment".
- **Hazard Identification:** it concerns the identification of the chemicals which are responsible for the potential contamination on the environment.
- **Hazard Assessment:** it is the evaluation of the hazard of the released contaminants divided in two parallel steps: the "exposure assessment" and the "dose-response assessment" (or "toxicity analysis").
  - The exposure assessment estimates the concentration of the contaminants in the environmental matrixes in correspondence to the exposition points to evaluate the level of exposure by organisms, including humans, for the given situation.
  - Dose-response assessment estimates the incremental effect of the dose of contaminants by means of ecotoxicological survey, epidemiological studies, etc.
- **Risk characterization:** it estimates the comprehensive risk, its eventual tolerability, the risk perception and the uncertainties.

There are several standards for risk assessment, but some of the most relevant ones include:

- The (U.S.) National Academy of Sciences (NAS), 1983. Risk Assessment in the Federal Government: Managing the Process.
- European Commission, 2003. Technical guidance document on risk assessment - part II, Technical Guidance Document on Risk Assessment.
- EPA, 2000. Science Policy Council Handbook. Risk characterization.
- ECHA, 2013. Guidance for human health risk assessment volume III, part B : guidance on regulation (EU) no 528/2012 concerning the making available on the market and use of biocidal products (BPR).
- EPA, 2011. Exposure Factors Handbook.
- EPA, 1998. Guidelines for Ecological Risk Assessment.

## 2.2 Life Cycle Assessment (LCA)

The Society of Environmental Toxicology and Chemistry (SETAC) was one of the first international organizations which developed the life cycle assessment (LCA). In 1991, it defined the life-cycle assessment as "*an objective process to evaluate the environmental burdens associated with a product, process, or activity by identifying and quantifying energy and materials used and wastes released to the environment, to assess the impact of those energy and materials uses and releases on the environment, and to evaluate and implement opportunities to affect environmental improvements. The assessment includes the entire life cycle of the product, process, or activity, encompassing extraction and processing of raw materials, manufacturing, transportation and distribution, use/re-use/maintenance, recycling, and final disposa*l."

Later the organization (SETAC, 1993) further developed the above statement, defining LCA as "*one of the tools used to examine the environmental cradle-to-grave consequences of making and using products or providing services.*"

Currently, the ISO 14040 (2006) defines the LCA as "*the compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product system throughout its life cycle*".

The operating methodology subdivides the LCA into the following iterative phases:

- **Goal and scope definition:** it is the definition of the objective of the study, its intended application,target audience and the specific system to be investigated. Further the functions, the functional unit (unit used as the reference in the study that represents the function of the system) and the system boundaries (processes of the system to be included in the study), are set.
- **Inventory:** it involves collection of the input/output data and related information from each process in line with the goals of the defined study. The life cycle invento-

ry modelling framework can be identified as: 1. “attributional”, depicting the system as it can be observed/measured, in which the single processes within the technosphere are linked by materials, energy and services flows; 2. “consequential” which aims to identify the consequences of a decision in the foreground system on other processes and systems of the economy, and builds the to-be-analysed system around these consequences.

- **Impact assessment:** in which input and output of the processes included in the boundaries are characterized to represent their potential consequences on the environment. Several characterization methods can be implemented to represent a comprehensive view of the potential environmental impacts of the system beinginvestigated. Results can be represented at a mid-point level, showing the potential risk of having consequences on specific impact assessment categories (e.g. climate change, freshwater eutrophication etc.) or at end-point level, showing the potential consequences for human health, ecosystem quality and depletion of resources.
- **Interpretation:** in which the results of the analysis are evaluated in terms of soundness and robustness, and overall conclusions, recommendations and decision-making are drawn in accordance with the goal and scope definitions.

The most relevant technical standards are:

- ISO 14040. Environmental Management – Life Cycle Assessment – Principles and Framework (ISO, 2006).
- ISO 14044. Environmental Management — Life cycle assessment — Requirements and guidelines (ISO, 2006).
- ISO/TS 14072. Environmental management — Life cycle assessment — Requirements and guidelines for organizational life cycle assessment

Important technical documents are also represented by:

- the ILCD Handbook – General Guidance for Life Cycle Assessment – Detailed guidance – JRC – EUR 24708 EN – 2010
- The ILCD Handbook – Recommendations for Life Cycle Impact Assessment in the European Context – JRC – EUR 24571 EN – 2011.

## 2.3 Similarities and differences

There are some key similarities and differences between the two tools, an overview of these is given in Table 1. In terms of methodology, the two approaches are both primarily technical-scientific assessment tools, concerned with quantitative modelling the potential or actual environmental impacts on the ecosystem. The results of both approaches may be presented as a single score impact index related generally to a single consequence (e.g.

**TABLE 1:** Comparison between Risk Analysis (RA) and Life Cycle Assesment (LCA).

| Characteristics | Risk Analysis (RA) | Life Cycle Assesment (LCA) |
|---|---|---|
| Definition | Although RA has a long history of use, there is no commonly accepted definition of risk and its mathematical formulation is mutable according to the different fields of applications.<br>A general, but well-used definition, describes the risk as the product of a frequency (or probability) and the magnitude of the adverse event. | Unlike RA, the definition of LCA has been agreed upon, at the international level, since the early 1990’s. .<br>LCA is the compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product system throughout its life cycle (ISO 14040:2006) |
| Aim of the methodology | RA quantifies the likelihood and severity of harm associated with a product, process, activity (e.g. smoking), agent (such as pollutants in different media), or an event (leakage of leachate from the bottom of a landfill).<br>In environmental risk assessments, both the potential exposure and the hazard associated with a chemical or chemicals, in specific release scenarios, are estimated.<br>RA focuses principally on receptors. | LCA estimates potential impacts, in diverse categories, by aggregating material and energy inputs and outputs from all the processes that take place from the start to the finish of a product/service, and reports these impacts relative to the function or service provided.<br>LCA focuses principally on emitters. |
| Object analyzed | In RA, the object may additionally include “natural” products, processes, activities. Alternatively chemicals (synthetic and naturally occurring pollutants) and events (such as floods and earthquakes) may be included. | The object analyzed in LCA can be the life cycle of a product/service, or the activities of an organization also described as a “product system”.<br>A product system is defined as a “collection of materially and dynamically connected unit processes which performs one or more defined functions” (ISO, 2006). |
| Perspective of the analysis | Prospective analysis looks forward in time<br>Retrospective analysis looks back in time | Prospective analysis looking forward in time can be implemented into LCA methodology adopting additional specific methods (system dynamics, etc.) |
| Scales of investigation | Typically RA focus on local scales with site-specific data generally being used in the models. | LCA requires generalized models and assumptions that lack the specificity typical of RA. |
| System boundaries: Spatial modelling | Spatial modelling of the related impacts (such as human health effects due to emissions) may or may not be site specific in RA. | Regionalization of impacts is related to specific impact assessment categories such as water scarcity. Generally, most of the category indicator results are not site specific. |
| System boundaries: time modelling | RA tends to focus on an endpoint (or endpoints) defined in time. | LCA results are integrated over time and hence give no information concerning the timing of impacts. |
| Outcomes | The outcome can be a numerical estimation of the likelihood of a specific harm or a comparison with criteria to define whether the risk is acceptable. | The outcomes include resource use (not present in RA), human health, and ecosystem quality . |
| Uncertainties | Uncertainties is considered in two aspects: the probability that an adverse event can happen and the ones related to the inputs values, for example, in transport and exposure modelling | Uncertainties is used to understand the variation of the parameters. Uncertainty is influenced by several factors including value choices (e.g. weighting factors). LCA Interpretation includes uncertainty analysis to investigate the robustness of results and therefore support the conclusion to be drawn. |

global warming; risk estimates related to the exposure of a non-carcinogenic compound; etc.) or by a series of impact indicators grouped per each investigated compartment (e.g. environment, resources depletion and human health). Besides, additional models (such as statistical analyses of uncertainties in data) may be used to interpret and evaluate the results.Both tools include the same general stages of process (problem identification, problem formulation, modelling, implementation, interpretation, feedback), but the operative steps are different (see above paragraphs).

LCA differs from RA in the information it provides to support decision making. LCA is mainly, but not exclusively, focussed on identifying possible improvements in a life cycle perspective rather than to compare the results against absolute standards and/or reference values. In RA, the absolute magnitude of the event under study is often a fundamental component in the analysis, and the “acceptability” of the risk, rather than identifying potential improvements , is of more interest.

There is an intrinsic difference in the scope of the two tools: LCA estimates several potential impacts of a system (products, services) from its whole life, using assumptions and information that most of the time are not site specific and considered to be marginal if compared to the pristine environmental conditions; while RA focuses on the concept of risk of a system (a plant, a contamination event, an activity, etc.) in a specific time and place.

Consequently, the outputs (aggregated or not) of RA are more site-specific and focuses on receptors, while those from LCA are generally integrated over time and space and focuses principally on emitters.

## 3. CONCLUSIONS

Life cycle assessment is motivated by gaining an understanding of the systemic environmental consequences of a product, process or service that fulfils a valuable economic or social function; LCA allows for a broader and more integrated overall view. Therefore, it is readily applied in identifying viable alternatives (e.g. a process, material, technology), evaluation of mitigation activities, its reporting and environmental management.

On the contrary, risk assessment is motivated by risk reduction and it allows for greater clarity on the risks associated with a given condition, defining the associated hazards and quantifying the consequences. RA is often applied in regulatory compliance and therefore it has a wider application in environmental forensics.

Table 2 reports a collection of common questions in the framework of environmental forensics and indicates whether the two tools can be used to provide an answer.

While recommendations to integrate the two approaches have remained a consistent challenge for the scientific community for at least 20 years; this is primarily due to the differences between the two tools. A more pragmatic approach is to apply the two methods in parallel, integrating only after obtaining separate results. This can be done by a multi-criteria decision analysis (MCDA), which can be defined as family of methods, designed to reveal the complicated tradeoffs or compromises inherent in complicated problems.

In summary, LCA and RA are two approaches with different peculiarities, each one more oriented in giving responses in specific contexts; for this reason, the first cannot substitute the second and vice versa. An integrat-

**TABLE 2:** Application of RA and LCA in environmental forensics.

| Common questions in environmental forensics | RA | LCA |
|---|---|---|
| What was the source of the contamination? | YES<br>In the retrospective risk analysis we are able to identity the sources in terms of chemical typologies | NO<br>LCA can help to better understand which of the processes involved is potentially related to the possible emission of the contaminant |
| When did the release occur? | YES<br>In the retrospective risk analysis a transport model can be used to reconstruct the history of chemical transport | NO |
| How did the release occur? | YES<br>In the retrospective risk analysis, by means of a transport model, it is possible to reconstruct the modality of transport in the different media and understand exposure during this transportation. | NO |
| Who contributed to the problem? | YES (partially)<br>Knowing the history and modality of the chemical transport we can identify the potential polluter(s) | NO<br>LCA can help to identify in a given system the processes probably related to the problem |
| What is the extent and magnitude of the contamination? | YES<br>In the prospective risk analysis, we can assess the magnitude of the potential damage when the adverse event occurs | NO |
| What is the potential risk to human health and the environment? | YES | YES<br>But unlike RA, LCA is not site specific |
| How much will the pollution cost? | Partially<br>With the risk assessment we can estimate the reduction of risk by applying some strategies and therefore the relative costs | NO |
| What is the best strategy for cleanup and remediation? | YES<br>With the risk assessment we can estimate the reduction of risk by applying different strategies for clean-up and remediation | YES<br>With LCA it is possible to compare the different systems used for the remediation |
| How should the costs be allocated amongst the responsible parties? | YES<br>If investigations reveal the presence of polluting chemicals at a site, an exposure assessment (a stage in RA), will help in assessing the amount of pollutants to which the parties claiming damage have been exposed to, through the various routes of ingestion, inhalation, and dermal contact. Once the exposure is quantified, established dose-response models can be used to evaluate if the health impacts alleged are the result of exposure to the pollutants or not. In other words, it is possible to make a comparison between calculated risks through the RA procedure and alleged risks in order to evaluate the claims from both the suspected polluters and of the affected parties. Thus, RA provides a scientific tool for the appraisal of damage claims, leading to correct decisions. | NO |

ed approach can lead to deeper information about a given phenomenon and on how some impacts can be or have materialized; this is an important part of environmental forensics and as such, it is suggested that further exploration into the use of these tools be completed.

## REFERENCES

Asante-Duah D.K. 1998. Risk Assessment in Environmental Management. John Wiley & Sons Publishers.

Baumann, H., Cowell, S.J., 1999. An evaluative framework for conceptual and analytical approaches used in environmental management. Greener Manag. Int. 26, 109–122.

Di Fiore, V., Cavuoto, G., Punzo, M., Tarallo, D., Casazza, M., Guarriello, S.M., Lega, M. 2017.Integrated hierarchical geo-environmental survey strategy applied to the detection and investigation of an illegal landfill: A case study in the Campania Region (Southern Italy). Forensic Science International, 279, 96–105. https://doi.org/10.1016/j.forsciint.2017.08.016

Environmental Protection Agency (U.S. EPA). 1996. Proposed Guidelines for Ecological Risk Assessment. EPA/630/R-95/002B.

Erskine A. 1997. What Does Risk Assessment Mean in Operating a Landfill Site. In Local Authority Waste & Environment.

Glickman T.S. and M. Gough. 1990. Readings in Risk. Resources for the Future. Washington, DC.

Henley E. J. and H. Kumamoto. 1981. Reliability engineering and risk assessment. By Prentice-Hall, Inc., Englewood Cliffs, N.J. 07632. ISBN-0-13-772251-6.

Linkov, I., Trump, B.D., Wender, B.A., Seager, T.P., Kennedy, A.J., Keisler, J.M., 2017. Integrate life-cycle assessment and risk analysis results, not methods. Nat. Nanotechnol. 12, 740–743. https://doi.org/10.1038/nnano.2017.152

Paustenbach D. 2002. Human and Ecological Risk Assessment. Published by Wiley-Interscience.

Persechino, G., Lega, M., Romano, G., Gargiulo, F., Cicala L., 2013. IDES project: An advanced tool to investigate illegal dumping, WIT Transactions on Ecology and the Environment. 173, 603–614, https://doi.org/10.2495/SDP130501.

Ram, N., 2000. The tools of environmental litigation support: How environmental litigation support teams employ a unique set of skills and tools to improve the outcome and reduce the cost of legal disputes. Environ. Forensics 1, 25–30. doi:10.1006/enfo.1999.0004

Rasmussen C.N. 1981. The Application of Probabilistic Risk Assessment Techniques to Energy Technologies". In Annual Rewiew of Energy. Annual Reviews. 6: 123-138.

Russell, M., & Gruber, M. 1986. Risk assessment in environmental policy-making. Science, 237,286-290

Salandin P. 2001. Analisi di rischio nelle opere idrauliche. In Tecniche per la difesa dall'inquinamento, Atti del 22° Corso di Aggiornamento. (In Italian).

SETAC. 1991. A Technical Framework for Life-Cycle Assessment. Workshop report from the Smugglers Notch, Vermont, USA, workshop held August 18-23, 1990.

Suter W. G. 1993. Ecological Risk Assessment. Published by Lewis Publisher.

Varnes D.J. 1984. Landslide hazard zonation: a review of principles and practice. Natural Hazards, Vol.3, Paris, France. UNESCO, 63.

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# GRASSROOTS INNOVATIONS IN SOLID WASTE MANAGEMENT

## Introduction

In this special column I would like to underscore the innovative potential that resides in the collective of waste pickers around the world. We know that particularly in the global South context, in so-called developing countries a significant contribution to waste management, to the recycling industry and consequently to the circular economy comes from these workers. In order to improve the performance of their work some of these individuals and groups have developed innovations that have allowed them to make a technological, strategic or management contribution that benefits their group of waste pickers, other groups or even the wider society. This column will provide some visibility of the developments that happen at the people's level, among waste pickers in different world regions and that make important contributions to the transition towards sustainability. This first article introduces grassroots social innovation theory and brings some reflections on the concept of innovations from below, by waste pickers, based on results of a international research collaboration through the Recycling Networks and Waste Governance project, which involves academics and waste pickers from Argentina, Brazil, Canada, Kenya, Nicaragua, Tanzania and Sweden.

## Background to grassroots innovations

Grassroots innovations are bottom-up solutions for sustainable and community-oriented developments, involving creative individuals, activists or organizations. The answers they find tend to address specific local contexts and respond to the interests and values of the communities involved. These innovations happen with minimal resources and mostly without formal support.

What characterizes many of these novel solutions are the democratic processes that give rise to grassroots innovations, actively engaging community members in the design, development or creation of alternatives. The focus of their innovations can vary from technological improvements, strategic approaches in waste management, commercialization schemes, environmental education initiatives to governance practices involving waste picker organizations.

They are called social innovations because they tend to benefit the public and bring social change, which is developed, approved and owned by the grassroots (Seyfang and Smith, 2007). Here, the innovation takes the form of community-based initiatives that emerge in a specific local context and explore whatever alternative configurations they can identify through their everyday praxis and long-term local experiences (Smith et al., 2016).

Waste pickers are increasingly being recognized both by society and by scholarship for their manifold and significant contributions to cities. The vast number of organized and autonomous waste pickers recovers a diverse array of recyclable materials and by doing so they reduce the urban carbon footprint and they help preserve the environment, besides generating an income for unemployed and vulnerable individuals. While facing innumerable challenges, waste picker organisations can be important innovation spaces for sustainability and social justice.

The waste collection, recycling, education, reusing and repairing practices enacted by waste pickers, individually and organized in groups, fit the grassroots innovation concept. Often born out of informal settlements and precarious housing and working situations, they can constitute new approaches and ways that can lead to the improvement of the quality of life of residents in informal settlements, and their own professional and human development.

Innovations as sociotechnical transitions are relevant issues studied in the economic and policy analysis fields which applies a broad multilevel perspective (MLP) (Geels, 2011). Transitions are seen as the outcomes of interactions between three levels: the landscape (macrolevel), the sociotechnical regime (mesolevel), and the niche (microlevel). This layered perspective helps understand and analyze complex and nonlinear phenomena such as historical, political, socio-economic and structural factors as well as sociotechnical transitions. The microlevel, or the niche is the spot where innovations are grounded, where innovations might evolve similar to an incubation room. Waste picker organisations are sometimes the space where new approaches and innovative ideas start, in the search for ways that can improve the working conditions or the outcomes of their work. Thus, they become innovative niches with the potential to be diffused and that thus prompt wider societal transformations and benefits. The flexible, improvised and not formalized nature of the work of waste pickers facilitates the emergence of innovation. Yet, as Morone and Cottoni (2016) posit niche spaces are also highly unstable and characterized by dispersed alternative technological niches, which usually lack the needed coordination between them to thrive and which can actually also be in competition with each other. In fact, “not every niche can survive for a long time, and only few of them will get to a point where they will really challenge the incumbent socio-technical regime” and bring desired innovations to a faulty and deficient system (Morone and Cottoni, 2016, p. 68).

For an innovation to flourish it progresses through different stages until it reaches maturity and is fully developed. For that to happen, the following three condi-

Managing editor:
Jutta Gutberlet
email: gutber@uvic.ca

Detritus / Volume 23 - 2023 / pages VII-X
https://doi.org/10.31025/2611-4135/2023.18280

tions need to coexist: (1) willingness and level of collective involvement and sharing among niche actors, (2) the presence of powerful actors with valuable assets for the development, and (3) accessibility and existence of compounding knowledge and accumulated experiences. When all three conditions are in place there is likelihood for an innovation to reach maturity. However, it is not a given that the incumbent regime might be destabilized by the niche innovation or that the benefits from it are shared equally and properly. Exploitation, corruption or manipulation can tint the social outcomes also of technological grassroots innovations (Morone and Cottoni, 2016).

## Waste pickers and their potential to innovate

How do waste pickers themselves define grassroots innovation? We asked this question during a research workshop conducted in Kisumu, Kenya in 2018, by the international research project Recycling Network and Waste Governance, involving waste pickers and academics from Brazil, Argentina, Nicaragua, Kenya and Tanzania (Azevedo et al., 2018). Waste pickers expressed the following: "Grassroots innovation processes develop human assets, they are emancipatory and promote social inclusion". In grassroots innovations "the focus is on the process" and for an innovation to be bottom-up in the context of waste pickers, "any idea, process or product, etc. must involve waste pickers as active partners in the design and development". The benefits can be various, e.g., "reducing the costs and time to make something", and they can also impact the self-esteem, inducing "a sense of excitement that makes us happy" or that "gives us pride". It is something that "fits the needs". The participants' opinions underlined the importance of process: "the way is more important than the outcome". Finally, there was a consensus that "innovation brings social change developed, approved and owned by the grassroots".

**TABLE 1:** Setbacks to social grassroots innovations among waste picker sector (source: Azevedo et al., 2018).

| | |
|---|---|
| Resources | • No starting capital, lack of trust from financial institutions<br>• Donated resources (e.g., machines) are often inappropriate<br>• Lack of facilities (e.g., for storage) machines, tools and transportation or operating in inappropriate locations (causing environmental pollution) |
| Markets | • Low material prices<br>• Market price fluctuations<br>• Competition with other recycling enterprises<br>• Difficulties in commercialization (lack of knowledge of retailers, sales of materials, supply chain, low bargaining power) |
| Legislation | • Difficult formalisation process (required certifications, permits, fees, etc.)<br>• Illegality impeding certain activities, polyethylene bag ban in Kenya, police persecution, harassment, bribes |
| Management | • Internal conflicts<br>• Lack of trust, lack of group cohesion<br>• Lack of experiences in administration, management, conflict resolution, etc.<br>• Bad leadership, bad management, absenteeism, lack of transparency<br>• Culture of working solo and lack of experience of collective management<br>• Precarious working conditions |
| Social | • Insufficient inclusion of women<br>• Social exclusion, alcoholism, conflicts, high member turnover<br>• Unequal distribution of benefits, funds<br>• Unhealthy and risk prone working conditions |
| Knowledge, identity | • Limited knowledge and capacity (e.g., to treat machines, to reach retailers)<br>• Advocacy skills<br>• Society's lack of knowledge of waste pickers and waste products, stigmatization instead of valorization |

Further extensive research conducted by the team mentioned earlier, applying a survey and key informant interviews in the 5 countries of study reveals a wealth of findings (Kain et al., 2022). Waste picker organizations often face multiple challenges, which makes it difficult for them to develop as niche for innovation (Table 1). They encounter the limitations imposed by informality and illegality, followed by persecution, exclusion and stigmatization, lack of initial capital or lack of appropriate technology to advance their work process. They experience the difficulties of formalizing their operations and the impacts of not being formalized, often punishing them by limiting their access to waste or banning waste picking in general.

Working as a collective and particularly under resource scarce conditions, as it is normal for waste picker organizations, is difficult, resulting in many additional challenges. The construction of trust between the members, with individuals who have been lifelong socially and economically excluded requires skills and patience. There are also diverse market-related challenges, due to global price fluctuations, competition with large companies and operations in deprived neighborhoods with low-income residents and low quality of recycling material that configure additional barriers that need to be overcome by these individuals and groups.

Despite the many challenges that still prevail the scenario of inclusive waste management and recycling important achievements can be highlighted as innovative, in terms of technology and product development, commercialization, creation of alliances, adapted management form, knowledge creation and formation of strong identity. The following image shows the cooperative Jovenes en Progreso in Buenos Aires, Argentina, whose members are primarily female, as they engage in partnership building with the local council, to address local waste management issues (Figure 1).

**FIGURE 1:** Members of the Youth Cooperative in Progress joining the Lomas de Zamora Deliberative Council to address the municipal resolution for inclusive recycling.

Table 2 summarizes some of the innovations captured in the research conducted by the Recycling Networks and Waste Governance project. The case studies provide examples of waste pickers innovating the commercialization or management processes, experimenting with knowledge transfer to the public, government or business community; or with innovations allowing them to add value and transform some of the materials that regularly arrive at their organizations, by creating specific machines or new processes to add value (Kain et al., 2022).

## Concluding remarks

The innovations emerging out of everyday work situations experienced by waste pickers in different parts of the world play a pivotal role in redefining the ways in which we deal with waste and recyclable materials. The research results underline the scope of the contributions to society and to the environment, coming from waste picker organizations. Despite this first systematization of grassroots social innovations presented here, there are many open questions still to be answered. Such as: What is the role of innovation in rethinking the work of waste pickers in different contexts? How can innovations re-examinate the waste market and waste policies? Does innovation improve the visibility and recognition of waste pickers? How does innovation strengthen and redefine the organization of waste pickers?

What is the role of innovation in rethinking what is considered "non-recyclable"? These reflections are relevant, particularly given the current elaboration of a global plastic treaty, the debates on sustainable and just transition or on

**TABLE 2:** Country specific grassroots innovations in waste management.

| Country | Technology | Commercialization | Management | Partnership | Social benefit | Knowledge transfer |
|---|---|---|---|---|---|---|
| Argentina | Processing new products (Reciplazas, children playground furniture). | | | Alliance with NGOs and authorities. | Prioritizing low-income neighbourhoods, children and people with disabilities in the creation of new products. | Awareness building, participating in the municipal council. |
| Brazil | Processing materials (regaining polymers, cooking oil into fuel). | Popular recycling (quality control of recycled materials), floating capital to enable collective sales, partnerships with companies (PEAD Oil, COOPERCAPS, Fundação Banco do Brasil). | Participatory decision making, self-management, transparency for all members. | Recycling contracts with local (e.g., Ourinhos), setting up conversation and support network involving other recycling networks. | Creating low barrier work opportunities. Workers' health improvement and risk reduction. | Support and capacity building (in accounting) Instituto Catasampa & Rede Cata Vida, Training program from waste picker to waste picker. |
| Kenya | Processing materials (reuse of charcoal dust into briquettes), new machines (bailing machine), new products (plastic fencing poles out of polyethylene bags, woven bags, mats and cushions), and new transportation means (hand carts). | Community clean-ups (as marketing and educational tool), implementation of clean-up and health clinics, educational tours, diversification of services (cleaning toilets, car washing, pit and septic tank emptying), engaging landlords in waste collection, training hotels (street food restaurants) for waste sorting, engaging youths for door-to-door sensitization. | Training in bookkeeping, team building, group management. | Training and capacity building in partnership with NGOs, Universities and governmental agencies, partnering with county government for transportation to the dumpsite | | Self-learning (identifying products and markets), teaching professionalism related to: materials, supply chain and markets; building partnerships with NGOs for training and capacity building. |
| Nicaragua | New products (jewelry). | | Internal management and self-organization, learning about collective interests. | Partnership with local government and private companies (transportation by boat). | Generating income for women. | |
| Tanzania | Identifying the collection of new materials (e-waste), new machines (for crushing), and transportation innovations (compressor trucks). | Selling to larger retailers, partnerships with companies (e.g., Soyana), operating in rural areas, allow for bank payment system (EFD machine), preparation of educational materials for customers. | Rotating leadership. | | Offering lunch, food, accommodation, loans for members; providing jobs for women, widows. | Training members in customer service and providing overall training. |

caron footprint reduction. In all of these issues, waste pickers are central protagonists. Looking at innovations evolving out of this sector will help frame new solutions to these very important topics.

## REFERENCES

Azevedo, A. M. M. de; Carenzo, S.; Goodluck, C.; Gutberlet, J.; Kain, J.-H.; Oloko, M. O.; P.rez Reynosa, J.; Zapata, P. & Zapata Campos, M. J. (2018) Inclusive waste governance and grassroots innovations for social, environmental and economic change: Report on first research outcomes of the project Recycling Networks & Waste Governance. In collaboration with WIEGO. ISBN 978-91-984547-3-4. Available at: https://www.wiego.org/reports/inclusive-waste-governance-and-grassroots-innovations-social-environmental-and-economic-chan

Geels, F. W. (2011). The multi-level perspective on sustainability transitions: responses to seven criticisms, Environmental Innovation and Societal Transitions 1 (1), 24–40.

Kain, J.-H., Zapata, P., de Azevedo, A. M. M., Carenzo, S., Charles, G., Gutberlet, J. Oloko, M. Reynosa, J. P. and Zapats Campos, M. J. (2022) Characteristics, challenges and innovations of waste picker organizations: a comparative perspective between Latin American and East African

countries. PlosOne, 1-27. DOI: 10.1371/journal.pone.0265889

Morone, P. & Cottoni, L. (2016). Transition to a Sustainable Agro-Food System: The Role of Innovation Policies. In Innovation Strategies in the Food Industry (pp. 61-76). Academic Press.

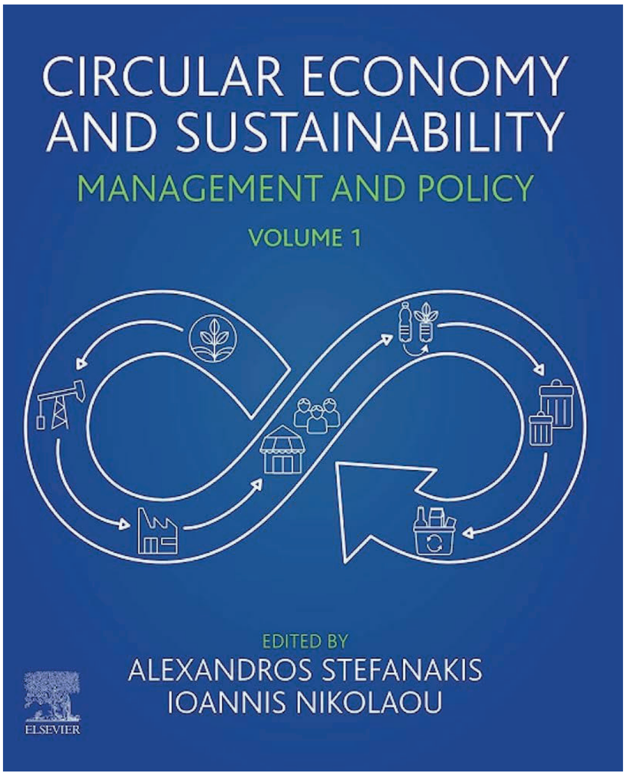
Seyfang, G. and Smith, A. (2007). Grassroots innovations for sustainable development: towards a new research and policy agenda. Environ. Polit. 16, 584-603.

Smith, A.; Fressoli, M.; Abrol, D.; Around, E. & Ely, A. (2016) Grassroots innovation movements. Routledge, New York.

detritus
Multidisciplinary Journal for
Circular Economy and Sustainable
Management of Residues

CISA

# BOOKS REVIEW 1



## CIRCULAR ECONOMY AND SUSTAINABILITY: MANAGEMENT AND POLICY (VOLUME 1)

### Edited by: Alexandros Stefanakis and Ioannis Nikolaou

The book “Circular Economy and Sustainability: Management and Practices” is an anthology consisting of 34 chapters written by a total of 82 people. The book, which comprises a total of 698 pages, is very comprehensive and thematically diversified - it covers several different aspects of circular economy (CE) and sustainability. This first volume of a two-part series focuses on the management and policy aspects of circular economy and sustainability in various fields and disciplines. Overall, the book provides a very wide-angle presentation and discussion around different solutions and concepts to implement CE. Whilst the book is not thematically divided into different parts, a certain division between themes can still be discerned. The opening contributions frame the concept of CE and the bioeconomy, followed by more business-oriented chapters on efforts and models for circularity, including various life cycle analysis perspectives. This is followed by several case studies regarding different countries as well as different types of operations, as well as some thematic studies concerning e.g., urban environments, collaborative leadership, and design practices.

Chapter 1 opens with a review of the current CE literature. The authors show which scientific areas have covered the largest part of CE literature, the development of CE studies over time, and the level of analysis that attracts the greatest emphasis regarding the CE concept.

In Chapter 2 the readers are introduced to the concept and relevance of Adam Smith’s invisible hand for CE. To reach true sustainability, the author means that the relationships between economics, society and the environment must be restored, and that it is not sufficient to merely address resource use and waste.

Chapter 3 provides an overview of system thinking and what drives systemic change, e.g., the engine of growth model. Examples are provided from both a macro level (e.g., material footprints) and micro level (e.g., linear, and circular business models).

In Chapter 4, the circular bioeconomy is defined through its key characteristics, for example the renewable nature of the inputs. While both the forest and agricultural sectors can be seen as representatives of a conceptual circular bioeconomy, the author notes that there are many challenges connected to commercial forestry and agriculture.

The relationship between the adoption of CE, focusing on strategies to optimize water use, and the financial performance among more than 10,000 European SMEs is analyzed and discussed in Chapter 5.

Chapter 6 provides an historical account of CE, from the 1960’s with the start of the environmental movement through the 1990’s and the emergence of concepts related to CE (Cradle-to-Cradle, Biomimicry, Blue economy etc.) to the 2010’s and forward, where putting CE from theory to practice is centerfold.

In Chapter 7, a framework for evaluating the involvement of firms in the CE on micro, meso and macro level is outlined, including a methodological tool in the form of a questionnaire for investigating CE strategies implemented by firms. The findings show for example that the main CE strategies adopted by firms include recycling, refurbishing, and remanufacturing.

Chapter 8 covers the topic of resource-service-systems (RSS). First, the authors identify a knowledge gap within the current RSS literature. Second, the authors propose solutions to the identified gaps by applying two analytical frameworks to explain what a circular strategy entail. Finally, this is applied to an exploratory case in the automotive industry.

The topic for Chapter 9 is CE and life cycle assessment (LCA). Where resource efficiency is supposed to increase

Managing editor:
Marco Schiavon
email: marco.schiavon.2@unipd.it

Detritus / Volume 23 - 2023 / pages XI-XIII
https://doi.org/10.31025/2611-4135/2023.18281

through closed loop technologies where waste is eradicated, is not necessarily beneficial from an environmental point of view. By combining the more linear CE and the holistic LCA, the study shows that it is possible to make more in-depth analyses of economic, social, and environmental sustainability.

Another perspective of life cycle assessment is provided in Chapter 10, where life cycle costing (LCC) is used to include economic sustainability in the CE. In the study, LCC is operationalized in a case study of the ceramic sector. By comparing the results of using a conventional LCC, which involves a strict economic evaluation, and an environmental LCC, where externalities are quantified from a life cycle perspective, the study shows that circularity does not necessarily mean a higher level of sustainability.

One of the world's largest consumers of raw materials and natural resources is the construction sector, and the sector also accounts for the largest part of waste in most countries. Chapter 11 presents a conceptual framework that shows the potential interactions of implementing three main principles of CE across different construction stages: reduced demand for buildings and/or materials, circular design of buildings and circular business models.

In Chapter 12, the author explores how eco-design is a key to success in closing the loops in CE through discussion of guidelines and the utilization of eco-design tools based on those guidelines. Some of the guidelines presented are design for assembly/design for disassembly, design for remanufacture, design for recycling or design for composting.

Chapter 13 provides a literature review of sustainable finance and begins by explaining the central topics of sustainable finance and CE, thereafter some salient parts of sustainable finance and the evolution of sustainable finance is discussed. Sustainable finance is proposed to contain three distinct elements: environmental, social and governance considerations. The chapter concludes by proposing three key sectors for future dissemination of sustainable finance and CE policies.

Chapter 14 is a case study aiming to e.g., review key features, drivers and barriers of the operational environment of CE, on different levels, including the municipal perspectives in the Kymenlaakso region in Finland, on advancing sustainable and CE-oriented public procurement. For the regional study, a questionnaire survey addressing main development factors and challenges was used. The results indicate that procurement organizations in many ways do consider sustainability and CE in planning and implementation, but that guidance and training is lacking. Public procurement is not only an important support for economic development, but also a useful tool for creating incentives for companies to move from a linear to a CE model.

The authors of Chapter 15 propose a methodological framework, a scoring system, as a practical tool for incorporating criteria for CE principles in awarding contracts and selecting tenderers.

In Chapter 16, the authors have performed an econometric analysis and a literature review to examine the connection between corporate social responsibility and corporate financial performance in the chemical industry. The analysis has taken place across several regions where conclusions about possible differences and similarities between them have been able to be drawn and discussed.

Human capability for collaborating to reach the Sustainable Development Goals (SDGs) is discussed in Chapter 17. The authors share their experiences, learnings, and results from an awareness-led social laboratory – Beyond Waste: Circular Resources Lab 2018. They share their insights on the values set and self-work needed, as the authors put it, to experience healing interconnectedness in the context of increasing complexity and diversity through "the magic of serendipity."

In Chapter 18, the authors describe how Signify (formerly Philips Lighting) has developed its sustainable design and environmental engineering, including the social dimension of sustainability. Lightning has made great technological advances that now lead to more possibilities to use light for brighter lives and a better world.

In Chapter 19 the authors analyze the role of CE for urban sustainability based on the premise that cities can play an important role in the development of a sustainable society. However, this requires a retraining of "urban thinking" and ideas for how to rethink the sustainable city are presented as the sustainable urban growth approach. The usefulness of CE is a matter of to what extent its operationalization can contribute to redefine urban growth.

The role of information and communication technology in the transition to smart and sustainable cities is explored in Chapter 20. Challenges such as privacy of citizens and possibilities such as deployment of internet-of-things or artificial intelligence are highlighted using emerging technologies in the smart sustainable city concept. A demarcation between soft smart cities and hard smart cities is presented. In addition to this, practical examples of how emerging technologies can be integrated to create a smart sustainable city are highlighted.

In Chapter 21, a framework for circular food consumption practices is proposed. Based on the "ecological perspective of consumption", consumer behavior is reconnected with CE-principles, and food consumption is seen as a stage in the biological metabolism for food. The framework is expected to make it possible to assess how likely it is that consumers will undertake CE-activities related to food.

Chapter 22 begins with a discussion of the impact of the extractive industries on the environment in general to then continue with deliberations upon the role of mineral wastes in the CE. A review of recent developments in valorization of mineral wastes is presented, especially which potential material benefits may be present and how legacy waste sites may affect other values such as eco-system services (e.g., biodiversity) culture and recreation or science.

The case study in Chapter 23 shows how waste can be used as a raw material for energy and material production. Up to 63% of the EU's raw material needs are covered by imports. EU legislation tries to solve the problems of material and energy dependence through two separate approaches. The study shows that waste recovery can solve both problems in one approach.

Chapter 24 presents an overview of solid waste collection and transport for rural communities and urban municipalities, with a more detailed insight for Poland. The authors provide examples of solutions to the optimization problems, e.g., by applying artificial intelligence algorithms to optimize the vehicle's travel route.

Through an analysis of 75 studies, Chapter 25 shows the different aspects of the new age clothing industry and its innovative sustainable practices and its contribution to a CE. Globally, the second most polluting industry is the apparel industry. Forecasts show that apparel consumption only will increase, and only about 20% of clothing is recycled and reused.

The aim of Chapter 26 is to contribute to the knowledge on transitional processes by examining existing literature on niche development and then apply the findings to the Dutch dairy sector. The results show that the dairy sector is “at the threshold of returning to a circular economy”, but that the situation is different today than it was before World War II, not least in terms of the institutional setting. A key question is at what institutional level it is appropriate to organize the CE-system: national, regional, European or even global?

Chapter 27 is an exploratory study aiming to determine conceptual categories related to CE in industry. Using content analysis technique to review the scientific literature two main factors of interest were found: waste management and production management, which is seen as confirmation of the “growing association between circular economy and development, integrating socioenvironmental management with economic and financial gain.”

In Chapter 28 the importance of the leadership factor for circular companies is discussed, and collaborative leadership is described as an appropriate leadership style. The chapter can serve as a guide for leaders who want to understand the role of, and improve, leadership skills when it comes to CE.

A trajectory analysis of eco-cement, in particular the ongoing transition from ordinary Portland cement to eco-cement, is presented in Chapter 29. The comparative analysis is based upon previous research regarding the eco-cement transition in the Netherlands which revealed six key dimensions. The authors examine the presence, and discuss the implications, of these dimensions in the ongoing transitions in China and Japan.

Chapter 30 describes the business, environmental and technical aspects of CE. Opportunities and challenges to implement CE principles at Linde Material Handling GmbH are then identified and discussed with the support of a literature study and in-person interviews.

In Chapter 31, the findings of the project VALUABLE are presented. The authors discuss the connection between electronic vehicles and drivers for a CE, through circular (vehicle) battery chains. In addition, potential barriers for such value chains are presented and which enablers would be necessary to facilitate them.

Corporate social responsibility reports from 2017 from five different companies are analyzed in Chapter 32 to assess the sustainability reporting of the cosmetic industries.

In Chapter 33, sustainable CE development in Finland is addressed from a company perspective. The authors explore what companies consider as necessary drivers for, and potential barriers against, a CE. Data is gathered through multiple-choice surveys witch then is presented and discussed.

In the final chapter of the book, the implementation of CE in two countries, Armenia in Portugal, is presented. The study is a contextual analysis in which the author accounts for how historical, geographical, and political context may affect fulfillment of a CE. Two central themes, socioeconomic relationships and emerging circular economy policies, serve as a starting point to discuss various macro, meso and micro level strategies. The chapter provides unique insights into how CE strategies are adopted in different countries.

Overall, the book comprises a comprehensive account of how circular economy can be understood from different perspectives. It provides a very wide-angle presentation and discussion around different solutions and concepts to implement circular economy. The book is designed to provide readers, including professionals, academics, engineers, researchers, government employees and industrial stakeholders, a better understanding of the concept and definition of circular economy, for it to be useful in their further work.

Maria Pettersson, Malin Johansson and Oskar Johansson
*Luleå University of Technology (SE)*
e-mail: maria.pettersson@ltu.se; malin.3.johansson@ltu.se; oskar.johansson@ltu.se

## ABOUT THE EDITORS

### Alexandros Stefanakis

He is Assistant Professor at the School of Chemical and Environmental Engineering, Technical University of Crete, Greece. His expertise lies in ecological engineering and technology, specifically in nature-based solutions for sustainable water and wastewater management. He is known as an expert and enthusiast of the green technology of constructed wetlands. He studies and investigates the role of nature-based solutions in a circular water economy.

### Ioannis Nikolaou

He is Associate Professor of Corporate Environmental Management and Performance in the Department of Environmental Engineering, Democritus University of Thrace, Greece. His current work focuses on linking knowledge-based views of firms with corporate environmental management and engineering, circular business models, as well as developing essential system dynamic models to explain strategic alliances of firms to face contemporary environmental challenges.

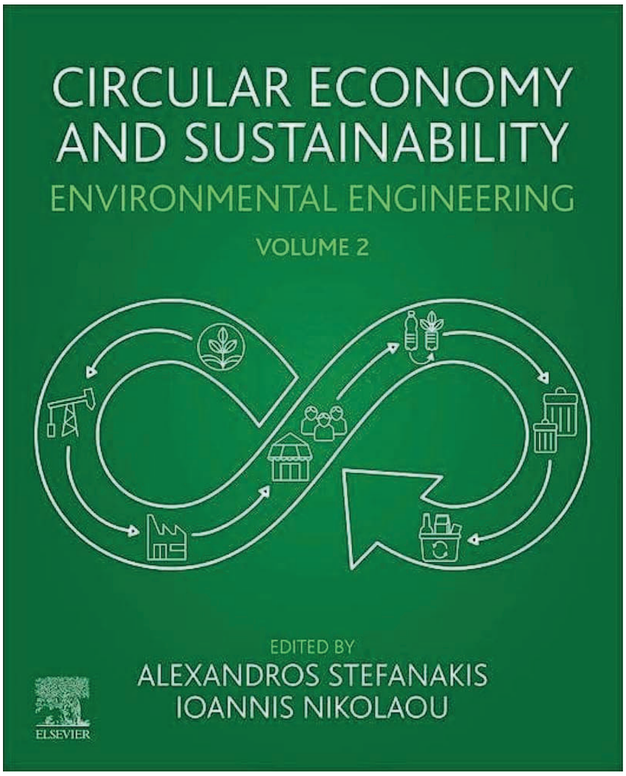
**Book Info:**

*Editors:* Alexandros Stefanakis, Ioannis Nikolaou
*Imprint:* Elsevier
*Year of publication:* 2021
*Page Count: 696*
*Paperback ISBN:* 9780128198179

detritus
Multidisciplinary Journal for
Circular Economy and Sustainable
Management of Residues

CISA

# BOOKS REVIEW 2



## CIRCULAR ECONOMY AND SUSTAINABILITY: MANAGEMENT AND POLICY (VOLUME 2)

### Edited by: Alexandros Stefanakis and Ioannis Nikolaou

This is a second volume of the two books, which are dedicated for the analysis of the concept and application of the principles of circular economy as an integral part of the sustainable development. This second volume is devoted to the engineering and technology aspects of these processes. The book presents a variety of perspectives, explores diverse solutions and concepts related to the implementation of circular economy and sustainability. The book contains information that can be useful for a broad range of professionals, scholars and decision makers by helping them to better understand the meaning of the circular economy concept.

The book comprises twenty-nine chapters, ten of which are focusing on wastewater treatment and sludge related issues, eight chapters are dedicated for nature-based solutions and constructed wetlands, two chapters are describing circular economy in the building sector, and the remaining nine chapters cover various sectors of industry, materials and waste management. Naturally, several of the chapters can be assigned to several of these areas as the studied subjects are closely related and interlinked.

Circular economy approach in the wastewater sector is highly relevant from recycling of water and nutrients perspective, as well as for ensuring food and water safety. Several circular and sustainable wastewater treatment systems and use of sludge are discussed providing examples based on cases in Greece (Chapter 4), India (Chapters 2 and 6), Brazil (Chapter 9), several Latin American countries (Chapter 2) and Iraq (Chapter 11). Although it is generally agreed that nutrient recycling is particularly important when managing wastewater treatment sludge, the risks of introducing unwanted and harmful substances to soil and food chain are also highlighted (e.g. Chapters 3, 7, 8, 10), suggesting the need for further research and technology development in this area.

A study on nature-based wastewater treatment and reuse in a compact space on one of college campuses in India (Chapter 6) shows that substantial savings of tap water can be made with relatively simple solutions, at the same time eliminating most of pollution from household. Placement of the installations close to housing did not even decrease the social acceptance of such compact, close-to-home wastewater treatment solutions.

A concept of the Wastewater Garden (WWG) is introduced in Chapter 11 as a type of nature-based wastewater treatment system resembling a constructed wetland, which not only has a function of cleaning the wastewater, but also ads an aesthetic value to the area by resembling ornamental gardens and contributing to history and culture.

Chapter 14 on the contribution of green roofs to the nature-based solutions for socially and environmentally responsible new cities provides a comprehensive review of such solutions with an example of a case study. Despite being highlighted as best management practices thanks to multiple social, environmental, and economic benefits, such solutions are still lacking support from financial incentives, regulations and public awareness.

The importance of constructed wetlands in wastewater and leachate treatment has been acknowledged for decades. Chapters 12, 13, 15, 16 and 17 highlight this technology from various angles, including circular economy, challenges that still need to be overcome in African countries, issues related with the presence of pharmaceuticals in wastewater, and recovery of electrical energy from the constructed wetlands by microbial fuel cells. Furthermore, the importance of preserving natural wetlands is discussed in the context of Philippines in Chapter 18, which also highlights the necessity of environmental education when

Managing editor:
Marco Schiavon
email: marco.schiavon.2@unipd.it

Detritus / Volume 23 - 2023 / pages XIV-XV
https://doi.org/10.31025/2611-4135/2023.18282
© 2023 Cisa Publisher

striving towards the implementation of the sustainable development goals.

Chapter 19 highlights how companies operating in the building industry design a circular business model (CBM), and how collaborative relationships across the building supply chain enforce their CBM. The study presents a step-by-step research framework, which highlights the phases and procedures that companies operating in the building sector can follow and implement for a successful CBM and analyses four Italian companies considered as circular economy (CE) champions from theoretical, management and policy-making points of view.

A study that explores how actors in Dutch construction supply chains deal with the deep uncertainty and dynamic complexity of decision-making in transitioning toward more mature closed-loop supply chains (CLSC) management is presented in Chapter 20.

Strategies for the global manufacturing of circular materials are suggested in Chapter 21. This chapter is both, a critical review and an opinion on the challenges related with circular materials. It presents the barriers and enablers of circular design, describes various current classes of material resources and highlight flaws in their designs preventing recycling, as well as discusses strategic routes to favor a circular design approach. Encouragement of recycling and repurposing, including development of advanced separation techniques, as well as redesigning materials by manufacturers are suggested as the main pathways that can enable the development of circular materials.

Chapter 22 guides the reader towards the area of Waste to energy and presents some examples of how energy issues and, in particular, the reduction of energy demand, have become the center of urban interventions in neighborhoods of Madrid periphery, as well as how it can be seen as the long-term strategy for energy refurbishment in the Spanish building sector.

Chapter 23 leads the reader to the area of circular economy models in the mining sector. This chapter describes how a circular analysis model can be applied to optimize mining operations and concludes that circular economy principles can provide an ideal framework for decoupling economic growth from environmental degradation caused by surface mining.

Chapter 24 describes the use of microorganisms in bioleaching as a tool that can be adapted to waste management sector for the recovery of various elements from MSWI ashes, by this contributing to the circular economy.

A review of circular economy initiatives implemented across Asia with emphasis on the eastern and southeastern parts is described in Chapter 25, which focuses on the circular economy applications in the forestry sector across the world, and the science-based initiatives undertaken for the benefit of the industry.

Extraction of nutrients from wastewater originating from corn-ethanol industry are described in Chapter 26. In this study, the utilization of a thermodynamic model to enhance struvite precipitation along with its operational function for P and N removal from wastewater was investigated, achieving the removal efficiency of phosphate and ammonium as high as 97% and 87%, respectively. Authors expect that this process can enhance an economic interest for nutrient recovery as struvite to be used as a high purity fertilizer.

Chapter 27 is dedicated to food processing waste as a potential source of adsorbent to be used for toxicant removal from water. Technical solutions and policy drivers have been identified as two key factors that could promote the application of food processing waste as adsorbents in wastewater treatment. Such solution is suggested as a promising green technology bringing benefits for society, businesses and the environment.

Chapter 28 complements the book with a study on sustainable circular economy design in 2050 for water and food security using renewable energy. It describes five technologies that are based on distinct circular economy processes to reach the goal of sustainability. Technological achievements in 2022 and beyond are suggested to set the path for innovations in technologies in 2050.

And finally, Chapter 29 concludes this book with a study on issues, challenges, and solutions moving towards circular economy in e-waste management sector in India. The chapter includes two case studies that highlight the benchmark practices both in the informal and formal sector. Security threats evolving from e-waste have been raised as a major issue in this field.

Overall, the book presents a versatile view of circular economy and how it is applied in various fields and disciplines to reach the goals of the sustainable development.

Jurate Kumpiene
*Luleå University of Technology (SE)*
Email: jurate.kumpiene@ltu.se

## ABOUT THE EDITORS

### Alexandros Stefanakis

He is Assistant Professor at the School of Chemical and Environmental Engineering, Technical University of Crete, Greece. His expertise lies in ecological engineering and technology, specifically in nature-based solutions for sustainable water and wastewater management. He is known as an expert and enthusiast of the green technology of constructed wetlands. He studies and investigates the role of nature-based solutions in a circular water economy.

### Ioannis Nikolaou

He is Associate Professor of Corporate Environmental Management and Performance in the Department of Environmental Engineering, Democritus University of Thrace, Greece. His current work focuses on linking knowledge-based views of firms with corporate environmental management and engineering, circular business models, as well as developing essential system dynamic models to explain strategic alliances of firms to face contemporary environmental challenges.

**Book Info:**

*Editors:* Alexandros Stefanakis, Ioannis Nikolaou
*Imprint:* Elsevier
*Year of publication:* 2021
*Page Count: 582*
*Paperback ISBN:* 9780128216644

detritus | Multidisciplinary Journal for Circular Economy and Sustainable Management of Residues

CISA

# DETRITUS & ART / A personal point of view on Environment and Art

by Rainer Stegmann

## What has Street Art to do with Waste?

Street Art is widespread, it covers house fronts, concrete walls, and many more places worldwide. Its acceptance is very divers from outright rejection to highest praise. There is a smooth transition from sprayed objects to Street Art.  Street artists often address political, social and moral themes also in an ironic and satiric way. Of course, the quality of Street Art is - as in many arts - very different.

I found this painting in a small town painted on a façade of a house in bad condition in Norway. It shows a man who falls down with an ice cream in his hand where some of it drops on his forehead. The ice-cream is the only colour in this painting. I do not see a message in this picture but it raises our interest because it is quite obscure; it lets the ugly wall vanish.

The most known Street Artist is Bansky who is highly accepted and celebrated; some of his works are sold at enormous prices.

I choose the art work from Bansky because it touches my heart. In my view it contains so many truths as the absurdity of wars, the innocence and involvement of children and their moral superiority. It switches the role of soldier and civilian and increasing this statement by letting a young girl search a soldier. I am sure that you will have your own feelings and associations.

Street Art in Stavanger, Norway

**BANKSY /** Image of a girl and a soldier on the West Bank in Bethlehem.

Coming back to my initial question about the connection of Street Art and waste. I think in contrast to many oil paintings Street Art Paintings have a relative short life time on the place where they have been created. After some years or decades, more and more pieces of plaster are coming – off, fall down to the ground and become waste. In wet facades sulfur compounds solubilize at the wall surface and the produced crystals increase in volume which makes the plaster to fall off. As a consequence, also the painting vanishes piece by piece. Moisture, sunlight, wind and frost are the drivers for corrosion making the plaster brittle and the colours pale. Waste in general shows also the transience of all kinds of materials. But different to many wastes the Street Art waste is not collected, treated often only partly safely disposed. Street Art reminds me the transience of products and their uncontrolled distribution in the environment. It is also a symbol of the insidious contamination.

But independent from this discussion, let us keep our eyes open and reflect the content of the images and enjoy well- made Street Art.

*In the next issue of Detritus I will present art that I discovered only recently: Bacterial Art. It is amazing how widespread it is in the respective circles. It is high time to present it to the waste community.*

Managing editor:
Rainer Stegmann
email: stegmann@tuhh.de

https://doi.org/10.31025/2611-4135/2023.18283

detritus
Multidisciplinary Journal
for Circular Economy and
Sustainable Management of Residues

CISA

# CONTENTS

**Editorial**

**Biological treatment**

**Recycling**

**Waste to hydrogen**

**Environmental Risks in Circular Economy**

**Columns**

**VOLUME 23 / June 2023**

**ISSN 2611-4135 / ISBN 9788862650328**