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WOOD GASIFICATION. INFLUENCE OF PROCESS PARAMETERS ON THE TAR FORMATION AND GAS CLEANING

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ABSTRACT

The article presents an analysis of influence of biomass pre-treatment and change of gasifying agent on the performance of an oxygen-steam-air updraft gasification plant and a technological process capable of delivering high quality producer gas. The paper shows that high temperature gasification process is stable for wood pellets, torrefied pellets and dry wood chips leading to syngas with calorific value of 5-5.5 MJ/m_N³ and moderate tar content below 10g/m_N³. After collection of liquids removed from syngas during the cleaning process it has been observed that the hydrocarbon composition in the torrefied wood fuel sample differs significantly from samples derived from wood chips or wood pellets. The paper also covers the influence of oxygen-steam-air combination on the quality of producer gas showing it can produce gas with LHV greater than 8 MJ/m_N^3 . The effects of process parameters change on composition of tars collected with an absorption type gas purification unit, designed for dust and tar removal are also reported.

1. INTRODUCTION

The technology of biomass gasification for energy and fuel production is now a firmly established alternative to incineration (Farzad, 2016; Sikarwar et al. 2017; Kirsanovs, 2017) in small to large scale installations (Kurkela, 2016; Dudyński, 2018). In cases of more specialized applications such as liquid fuels production or successful energy generation with piston engines the tar present in producer gas must be removed with cheap and efficient processes if the technology were to achieve commercial viability and compete with wind and solar installation (Broer, 2015). The amount of tar and other contaminants in the gas depends on several factors such as: the pre-treatment of biomass e.g., drying and compactification for wood pellets, torrefaction or partial pyrolysis for torrefied pellets, the specifics of the gasification process and apparatus, the choice of the gasification agent, temperatures of the process and details of the gasification chamber construction (Dudyński et. al, 2015; Dudyński, 2019). We compare the composition and volume of tars and hydrocarbons collected from producer gas coolers during air gasification of wood chips, wood pellets and torrefied wood pellets. We then chose wood chips as the fuel for tests using three different gasification agent compositions - air, air and steam, and air, steam and oxygen - and compare the resulting composition of producer gas and various hydrocarbons collected in various stages of the gas purification process. The system used

for the experiment is an updraft gasifier equipped with a steam generator and oxygen enrichment unit presented on Figure 1 equipped with an absorptive gas cleaning system described in (Dudyński, 2019).

The gasification systems can be used for effective energy production in a variety of ways (Arena et al. 2010; Bang-Møller et al., 2010) The conventional solution of generating energy from locally produced biomass waste, wood pellets or wood chips by utilizing producer gas burning for steam generation process, which is then used for integrated heat or/and electricity production is now a firmly established method (Dudyński et al. 2012; Dudyński 2018). This solution has found widespread use in cogeneration installations (Kirsanovs et al., 2017), but the electric energy effectiveness is usually far below 20% for small systems. Improvements are possible with application of a gas engine with effectiveness up to 30%, but such solutions are still under development. Gas for piston engines should contain less than 50mg/m $_{\!\!N}^{\!\!3}$ of solid particles and up to 100 mg/ $m_{_{N}}^{_{3}}$ of tar. Attaining such gas parameters for updraft gasifiers, requires intensive gas cleaning making such systems expensive and difficult to operate in small plants (Koido at al., 2017). The more innovative solid oxide fuel cells (SOFC) systems with much higher effectiveness of electric energy production are also under active development (Brunashi et al., 2017). SOFC systems operating on producer gas are a promising technology improving both the electrical and heat efficiency of the systems however they also require



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comprehensive gas cleaning and operate at high temperature exceeding 900K thus requiring dry, high temperatures resistant, filter systems (Poncratz at al., 2021) which creates new problems for designs and operations (Marcantonio et al., 2020). An excellent review of biofuel production from biomass gasification including recent hydrogen production facilities can be found in (Molino et al., 2018). Supercritical water gasification for hydrogen production is described in (Correa et al., 2018). It can be an important route for bio hydrogen production from biomass. These new technologies which include steam gasification and supercritical water gasification, show a high potential in field-scale applications, but the selectivity and efficiency of hydrogen production must be improved in effective industrial applications to compare with other sources of hydrogen (Cao at al., 2020) and insight in the tar content and composition in the producer gas is of importance for all such research.

The fixed bed, updraft gasification systems are robust, reliable, and can scale up to higher capacities although they do require effective gas cleaning units and improvements of syngas guality to achieve successful coupling with piston engines. Air gasification is a notably simple and inexpensive form of the process, however due to nitrogen dilution of the producer gas it results in syngas LHV below 5.5 MJ/m_N³ for dry biomass and even lower for wet materials like sawdust or feathers (Dudyński, 2015; Chmielniak, 2019). Typical tar levels for updraft air gasification range from 10 to 150 g/m $_{N}^{3}$. The tar levels are inversely related with syngas temperature, with a notable, sharp drop for temperatures over 1050K (Bassu, 2018). Steam is a very promising choice of gasification agent as it leads to high H₂ content but requires temperatures over 1000K for the water gas and water shift reaction to be effective. In industrial installations a mixture of steam and air is applied to improve gas quality. A steam generator requires additional energy but in can be obtained from heat recuperated from the piston engines used for electricity generation which in turn leads to the improvement of overall efficiency of the system. One of the most promising methods of improving gas quality is to replace air as the gasification agent with oxygen enriched air, mixed with steam to enhance the calorific value of the gas and limit the tar levels (Liu et al., 2018; Dudyński, 2019). We developed such a system, the details of its design and the results of its operation on dry wood chips, pellets or torrefied material and various gasification gases are presented below.

The oxygen generator is a large energy consumer and in the presented system up to 20% of the electric energy which can be produced with clean gas is internally used, however this can be improved in larger systems. Therefore, the electricity and energy consumed in the fuel feed, oxygen and steam production, syngas transport and cleaning systems are not included in the efficiency calculations for the process.

2. THE GASIFICATION UNIT

The paper presents a new, oxygen-steam-air driven biomass gasification system and process capable of producing syngas with calorific value up to 8 MJ/m_N^{-3} on a tar

free basis. This device is an improvement on the biomass gasification units successfully used in many industrial localisations in Poland, intended for energy production using waste from technological processes as fuel (Kwiatkowski et al., 2013; Dudyński, 2018). The operating scheme of the system is presented on Figure 1. In respect to the installation presented in (Dudyński, 2019) the feed system was changed by adding a hydraulic press for wet or bulky materials and technical details of equipment mixing oxygen rich air with steam and hot air making the system more flexible. Moreover, all coolers have been grouped into a single system and scrubber order in the gas purification system have been rearranged leading to improved performance.

The thermocouple T1 is located at the bottom of the gasification chamber, T2 above the cones delivering the air+steam mixture, T3 at the center of the gasification zone, two meters from the bottom and T4 at the top of the chamber close to the syngas outlets. The gasification chamber is 6000 mm high with an inside diameter of 2000 mm. This installation allows for gasification of different biomass materials including bulky and wet ones as the residence time can be as long as 8 hrs. Various tests for different materials and conditions can thus be performed.

The presented system is a gasification fixed bed 2MW unit which can use hot air, hot air – steam mixture and up to 40% oxygen air – steam – hot air combination as gasification agents. It uses heat from producer gas cooling to heat up the primary and secondary air to temperatures above 520K. The primary air can be mixed with 450K steam to form controllable gasifying gases which are injected into the chamber at the bottom of the installation. Combination of these two streams allows the system to operate in two distinct modes suitable for different feeds.

Low bed height – low steam content in primary air which can be applied for wet low calorific fuels. The producer gas has a temperature above 1000K at the outlet, LHV bellow 3.5MJ/m³ and less than 10 g/m³ of tars.

High bed height - high steam content in primary air, effective for dry, high calorific fuels. The gas temperature in the gas chamber is 650K, the LHV above $5MJ/m^3$ and tar content can be well above 10 g/m³.

We presented the extended discussion of the gasifying process in the low bed mode for wet feather fuel in (Dudyński et al., 2012; Kwiatkowski et al., 2013) and in the present work the high bed mode is analysed.

The oxygen rich air, containing up to 40% of oxygen and flow speed standing up to at 100 m³h⁻¹ is produced in a separate unit utilising molecular sieves. This air stream is mixed with steam prior to its application in the gasification process. The role of steam is twofold. First it performs as an oxygen dispersive medium preventing occurrences of high temperature spots at the bottom of the gasifier, where the char and gas burning processes are the most intense. Application of oxygen rich air can cause the temperatures to locally exceed 2000K posing a serious threat to the gasifier and equipment. Steam and additional air lower the oxygen content to a maximum of 25% causing the temperatures in the bottom part of the gasifying chamber to stay well below 1500K thus ensuring the operations are smooth and safe. These temperatures are still very high and in or-



FIGURE 1: Operating scheme of the biomass gasifying unit.

der to prevent ash sintering we must take care to eliminate sand from biomass fuels and keep the temperature in the lower part of the gasifier where ash is predominantly present below 900K to prevent ash and silica agglomeration and melting. The hot gases move up in the chamber and in the upper part of the gasification unit the overheated steam and CO₂ can react with fixed carbon and tars to produce CO and H₂ in a water-gas and water shift reaction, efficient in high temperatures thus significantly improving the producer gas quality and generally lowering temperatures in the carbon burning zone. We constructed a unit allowing for various combinations of gasification gases delivered via a multilayer and multipoint injection system. At the bottom of the gasifier four rotating cones equipped with multiple inlets are located. The oxygen rich air mixed with steam is injected into the bottom part of the unit through these cones. Approximately 50% of the oxygen necessary for the gasification process is provided through these four injection ports and most of the hot CO₂ and H₂O, necessary for the process, is produced in the bottom area of the reactor. The remaining air necessary for the process is injected with 32 nozzles located above the cones, close to the bottom of the pyrolysis zone. The delivered air reacts with the hot pyrolytic carbon producing a CO rich gas and extending the high temperature zone in the gasifier thus intensifying the process of wood drying and carbonising occasionally, in cases of very dry materials, even extending the drying process with flash pyrolysis of smallest particles. Such a solution guarantees a more uniform distribution of the gases and solid material in the chamber, and therefore improves the mixing of the carbonised material with gasifying agents. This enhances the effectiveness of the gas production and unification of the temperature's distribution in the gasification process. The control unit allows us to continuously change the parameters and the composition of the gasifying gases leading to better control of the producer gas parameters.

The system maintains 0.2 kPa of negative pressure below the atmospheric pressure (1013.25 hPa) in the upper part of the gasification chamber. The air flow at the bottom of the gasifier can be adjusted to maintain control and conditions of the process as well as the stability of syngas generation. The test begins with wood chips being fed to the gasifier. After achieving stable conditions for bed height and temperature distribution inside the gasification chamber the fuel feed is adjusted to the required capacity and the test runs commence. It means that all test start with gasifier containing approximately 2000kg of charcoal from wood chips with a layer of ash at the bottom. We commence all tests with the same initial conditions keeping a constant level of material in the gasifier.

Dry fuels are fed with two screw conveyors, one on each side of the gasifier, to maintain the level of the bed. Liquid fuels can be introduced with a pump system. Both dry and liquid fuels are fed in a continuous fashion. Wet bulky materials on the other hand are introduced with a hydraulic loading drawer in a discontinuous fashion.

Gas leaving the gasification unit still contains high amounts of carbon dust and heavy hydrocarbons which must be removed before gas is fed into the engine. With high levels of tars and carbon dust the produced gas requires a very efficient purification system. There are various methods of gas cleaning and virtually every wood gasification unit developed their own unique technology (Boerrgter et al., 2004; Bocci et al., 2010; Dudyński, 2019). Our system consists of a high temperature ceramic cyclone, integrated with the gasifier for particle removal, one air cooler, two water coolers capable of lowering the gas temperature below 60°C, water scrubber, oil scrubber for elimination of tars and hydrocarbons and an active carbon filter for final gas conditioning. The operating scheme for this unit presents on Figure 2 indicating the material flow and temperatures during the process.

Syngas cleaning takes place by precipitation and removal of tars and heavy hydrocarbons contained therein by adequate cooling of the gas in several stages and absorption of other impurities in the syngas by gas flow through two absorption devices (scrubbers). The first scrubber uses water and the other fuel oil as absorbent. Both scrubbers are equipped with demisters placed immediately before the outlet, which keeps the scrubbing liquids inside the apparatus.

The gas from the gasification of wood is transported to the first exchanger (cooler I) in a counter-current system, the gas will be cooled to temperatures in the range of 100-110°C then the gas moves to the fan and is pumped to a two-stage heat exchanger system. On the second and third levels of cooling, the temperature of the gas drops below the precipitation point of tar and the hydrocarbons flow down the walls of the exchangers into the lower parts, where a discharge spigot enables liquefied contaminants to be collected. An important factor determining the possibility of precipitation of tars and preventing the formation of carbon deposits and clogging of exchangers is to maintain appropriate temperatures at the inlet and outlet of each cooler.

The gas then flows to the water scrubber, where it is cleaned of remaining impurities: dust, tars, and water-soluble hydrocarbons. After passing through the water scrubber, the gas will then be fed to the absorption column (oil scrubber), where the residual hydrocarbons will be washed away by the oil. After cleaning, the gas will be transported





to the engine or returned to the combustion chamber.

The oil flowing through the scrubber is pumped to the clarifier and then to a candle filter unit. The purified and cooled oil is then returned to the absorption column (oil scrubber). The water flowing through the scrubber is pumped to the clarifier and then recycled to the installation. In the future it will be used as a source for steam for the gasification process.

Fuel inflow is up to 500 kg/h, air flow in the gasifier is 600 $m_{\rm N}{}^3/h$ the raw syngas flow up to 800-900 $m_{\rm N}{}^3/h$. In the first heat exchanger up to 1000 $m_{\rm N}{}^3/h$ of air is used. The oil flow is 3.6m³/h and in the water scrubber utilises 3.6m³/h of water. The oil/syngas ratio is 2.5/1 and water/syngas is 3.6/1.

3. MATERIALS AND METHODS

The industrial tests have been divided into two stages. First, the analysis of influence of the biomass pre-treatments such as drying, pelletisation and torrefaction on the composition of the tars and liquids collected in the coolers of our cleaning system with methodology like that used in our previous tests (Dudyński et al., 2015). Three types of fuel have been used - dry wood chips, wood pellets obtained with flash drying of sawdust at 550K, and pellets from sawdust torrefied at 560-580K. The liquid condensate was cooled and then separated into water containing liquid carbohydrates and solid heavy tar. The tars were dissolved in solvents and like liquid parts analysed with gas chromatography where components were measured. The heavy tars were additionally analysed with a simulated distillation method to define boiling points of components.

3.1 Gas chromatography

All water and tar samples were analysed using gas chromatography (GC) techniques: Flame Ionisation Detector (GC-FID) (for quantification) and a Mass Spectrometer (MS) (for peak identification) fitted with a High Polarity HP-FFAP column. The HP-FFAP column (50 m x 0.2 mm x 0.33 μ m) is a high polarity column suited for analyses of organic acids, free fatty acids, and phenols. Approximately one μ L of sample was injected into the FC column with a split of 200 (if samples were too diluted a split of a 100 was used). The GC oven was programmed as follows: Initial temperature of 60°C, ramp up of 6°C per minute to 240°C and then hold the temperature for 30 minutes (until all compounds have eluted). Gas low through the column was 1.2 per minute (helium in GC-MS and hydrogen in GC-FID).

3.2 Simulated distillation

Simulated distillations (SimDis) were conducted for prepared tar samples to determine the boiling point distribution of extracts from biomass gasification tars. Simulated distillation was conducted on a high temperature GC-FD fitted with an ARX 2887 Restek column (10 m x 0.53 mm x 0.53 μ m). Approximately 0,2 μ L sample was injected into the GC column per analysis. The GC oven program was as follows: initial temperature of 40°C, ramp up of 15°C per minute to 540°C and then hold the temperature for 10 minutes.

The second stage of our experiment consists of gasifying the dry wood chips (max 25% of water) as an example of the most popular biomass feedstock with three different gasifying agents. During the test gas samples have been collected at the inlet and outlet of the purification systems, and we measured the effect of application of the complete purification scheme on the resulting producer gas quality. Additionally, water samples were collected at the outlet of each cooler separately and each sample was analysed with gas chromatography to determine the hydrocarbon composition. Table 1 presents the proximate and elementary analysis of the feedstock used for tests in our industrial experiment.

The gradual carbonisation and decomposition of hemicellulose begins in 450K which can be easily reached during drying and pelletisation of sawdust, this process is exacerbated at higher temperatures (Partridge et al., 2020) leading to a gradual rise of carbon content in biomass feed from wood chips to pellets to torrefied pellets.

4. RESULTS

4.1 Collection of hydrocarbons

A test has been conducted for three fuel types - wood chips, wood pellets and torrefied pellets. The test for each material took six hours and involved the usage of 1800-2400 kg of fuel. The process was conducted as air gasification with the gas leaving the gasifier in ranges 900-1100 K. The equivalence rate ER in the experimental runs was ER=0.38-0.4 in case of woodchip, ER-0.34 for torrefied pellets and 0,36 for wood pellets. Ash analysis shows that the carbon conversion rate was greater than 99% and the dry syngas/dry fuel ratio was 2.45 $m_{_{\!N}}^{_3}\!/kg.$ Figure 3 presents the temperatures measured in various spots inside the gasifier - at the bottom T1, at the lower part of the gasification chamber T2, at the center of the gasification zone T3, as well as the temperature of gas leaving the gasifier. It can be observed that for dry fuel the air gasification becomes a high temperature process leading to moderate tar loads in producer gases.

It can be seen from the temperatures distribution that the process is remarkably stable in all three cases. For dry fuels, the syngas and gasification zone temperatures are very close indicating that the drying zone is very narrow.

TABLE 1: Properties of gasified biomass feeds.

| | Unit | Wood pellets | Torrefied pellets | Wood Chips |
|------------------|----------------|-----------------|-------------------|---------------|
| Water content | [%] | 10 | 4.5 | 20-25 |
| Volatiles | [%] | 79.1 | 70.6 | 81.36 |
| Fixed-Carbon | [%] | 19.2 | 39.5 | 18.3 |
| Ash | [%] | 0.5 | 0.9 | 0.7 |
| Ultimate analysi | is (dry basis) |) | • | |
| С | [%] | 50.6 | 56.0 | 48.8 |
| Н | [%] | 5.9 | 5.0 | 5.7 |
| 0 | [%] | 43.3 | 38.6 | 45.5 |
| Ν | [%] | 0.2 | 0.4 | 0.1 |
| LHV | [MJ/kg] | 17.7 | 20 | 14.4-15.5 |



FIGURE 3: Operating scheme of the biomass gasifying unit.

The summary of the gasification process can be found in Table 2.

The syngas leaving entering and leaving the purification system was analysed with a gas chromatograph (Varian CP-4900) calibrated to determine the levels of following compounds H_2 , CO, CO₂, CH_4 , C_2H_2 , C_2H_6 , C_3H8 , N_2 and O_2 . The levels of CO, CO₂, CH_4 and O_2 were also independently measured by and online gas analyser Ultramat 23 while Colomet 6 was used for secondary hydrogen measurements. The gravimetrical determination of liquid hydrocarbons was conducted with samples taken every two hours. The averaged results were presented in Table 3.

Tar samples from all three coolers were collected into a single barrel per fuel type - thus producing three barrels in total which were then analysed in a laboratory. The bulk results are presented in Table 4.

4.2 The heavy tar analysis

The collected water was black in colour and at the bottom contained solid (gummy like) substance being a composition of heavy tar. These substances constituted less than 1% of mass of collected liquids. The elementary analysis of these materials is presented in Table 5, these samples were collected separately for each of three woody biomass materials. As the test requires more time to collect the liquid condensates in amounts allowing for solid tars to be easily separated the process was conducted as air gasification with the gas leaving the gasifier in ranges of 900-1000K.

We dissolved the tar in tree solvents (ethanol, tetrahydrofuran (THF), and 1-methyl-2-pyrrolidone (NMP)) and the chromatographic results averaged (as different solvents present different molecular composition of the same tar).

| | ······································ | | | | | | | | |
|-------------------|--|--|-----|--|-------------------------------|--|------|--|--|
| Fuel | Syngas temperature [K] | Fuel streamAir streamDry gas stre[kg/h][m³]ma/mauntaria[ma/mauntaria[ma][ma]fuelfuel | | Dry gas stream [m _N ³/kg of dry fuel] | LHV [MJ/m _N ³] | Process efficiency% [Egas/Efuel] | | | |
| Wood chips | 1020 | 400 | 500 | 2.05 | 3.06 | 5.0-5.1 | 65.4 | | |
| Torrefied pellets | 1120 | 300 | 500 | 2.15 | 3.06 | 5.5 | 68.6 | | |
| Wood pellets | 1080 | 310 | 500 | 2.12 | 3.0 | 5.2 | 65.8 | | |

TABLE 2: Process parameters of air gasification during tar collection.

TABLE 3: Average parameters of syngas during tar collection.

| Syngas components % of vol (Dry gas) | | | | | | | | | |
|--------------------------------------|----------------|----------------|------|-----------------|-----|-------------------------------|--------------------------|---|--------------------------------|
| Fuel | H ₂ | N ₂ | со | CO ₂ | CH₄ | C ₂ H ₄ | Others hy- drocarbons | Calorific value of cold syngas [MJ/m _N ³] | Tars [mg/ m _N ³] |
| Wood chips | 6.2 | 60.9 | 16.7 | 12 | 2.8 | 1 | 0.4 | 5.06 | 6500 |
| Torrefied pellets | 7.4 | 59.3 | 18 | 8.8 | 4.4 | 1.1 | 0.3 | 5.5 | 3200 |
| Wood pellets | 6.8 | 59.1 | 17.3 | 11.8 | 3.7 | 0.9 | 0.4 | 5.2 | 5100 |

| TABLE 4: Summar | y of tar | collection | in | cooler | system. |
|-----------------|----------|------------|----|--------|---------|
|-----------------|----------|------------|----|--------|---------|

| | Unit | | Torrefied pellets | Wood pellets | |
|---------------------------|---------|--------|-------------------|-----------------|--|
| Liquid collected | [kg] | 115 | 41 | 60 | |
| Hydrocarbons collected | [kg] | 10.8 | 2.5 | 8.5 | |
| Hydrocarbons to fuel | [kg/kg] | 0.0045 | 0.0016 | 0.0047 | |

TABLE 5: Elementary analysis of the biomass raw tars.

| | | Wood pellets | Torrefied pellets | Wood Chips |
|-------------------|---------|-----------------|-------------------|---------------|
| Dry mass | | | | |
| С | [%] | 62.2 | 38.6 | 55.1 |
| Н | [%] | 7.4 | 8.1 | 7.2 |
| N | [%] | 4.5 | 0.8 | 1.7 |
| S | [%] | 0.0 | 0.0 | 0.0 |
| O (by difference) | [%] | 26.1 | 52.6 | 36.1 |
| Calorific values | [MJ/Kg] | 26.4 | 17.7 | 22.2 |
| H/C (atomic) | | 1.4 | 2.5 | 1.6 |
| H/O (atomic) | | 4.5 | 2.5 | 3.2 |

The results of analysis are presented in Table 6.

The tar from wood chips contains mainly alkylphenols, organic acid and small amounts of polyfunctional aromatic oxygenates. Wood pellets tar contained linear and cyclic aliphatic oxygenates, polyfunctional aromatic oxygenates and alkylphenols. Torrefied pallets tar contained acids, aliphatic alcohols, alkylphenols, aliphatic oxygenates and alcohols. The tars were strongly different with shorter molecules length often observed in more pre-treated material. The biomass tar contained a range of molecular sized chains including large polymeric molecules which are larger and spatially more complicated than these observed in coals or charcoal feedstocks (Vrenghenhil et al., 2009). The process of torrefaction changes lengths and structures of

TABLE 6: GC-MS composition of the tars from three feedstocks.

hydrocarbons in collected tars as can be clearly seen from simulated distillation analysis presented below.

4.3 Simulated distillation

The simulated distillation was conducted for all samples to determine the boiling point distribution of extracts from three feedstocks. The results clearly indicate that intensive drying and torrefaction lower the complicated structure of wood thus the tar derived from more pre-treated material have simpler structures (lower boiling point). We present on Figure 4. the result of distillation of tars.

We can see clearly from Figure 4 that the process of wood torrefaction lower significantly the maximal boiling point of heaviest tars derived from the material confirming our observation that the structure and lengths of hydrocarbons obtained in the gasification process simplify in comparison with tar extracted from fresh wood or wood pellets. I future work more careful analysis with more carbonized material like charcoal and annealed charcoal should be conducted for more conclusive results.

4.4 The analysis of water condensates

The water condensates derived from each of three feedstocks were analysed with help of gas chromatography and are presented in Table 7.

The main organic species determined by GC-MS in water condensates were acids, aliphatic alcohols, alkylphenols, and linear and cyclic oxygenates but in slightly different proportions then in appropriate tars from these materials. As the hydrocarbons in tars were oxygenated and highly reactive further work should consider characterisation on site during test or rapid quenching to stop fast degradation or polymerisation with other molecules which change the results of measurements.

These results are not conclusive. The variations of the hydrocarbon composition in differently prepared feedstocks can be partially attributed for the effects of the intensive drying as in case of wood pellets or torrefaction at

| | | Wood pellets | Torrefied pellets | Wood Chips |
|--|-----|--------------|-------------------|------------|
| Molecular component | | | | |
| Aliphatic | [%] | 0.1 | 0.8 | 0.4 |
| Acids | [%] | 11.1 | 50.15 | 22.1 |
| Aliphatic esters | [%] | 0.2 | 0.05 | 0.1 |
| Aliphatic aldehydes and ketones | [%] | 1.6 | 0.8 | 5.7 |
| Aliphatic alcohol | [%] | 1.8 | 7.7 | 0.7 |
| Alkylbenzenes | [%] | 0.8 | 0.4 | 5.2 |
| Alkylphenols | [%] | 21.9 | 11.8 | 34.1 |
| Furan | [%] | 0.1 | 1.2 | 7.9 |
| Furan (polyfunctional oxygen) | [%] | 5.2 | 0.0 | 1.1 |
| Linear and cyclic aliphatic oxygenates (polyfunctional oxygen) | [%] | 27.0 | 10.7 | 6.2 |
| Aromatic oxygenates | [%] | 25.0 | 12.2 | 15.2 |
| Nitrogen and sulfur heteroatoms | [%] | 4.2 | 4.0 | 1.2 |
| Total | [%] | 100 | 100 | 100 |



elevated temperatures which simplified the original wood chemical structure and lowered the maximal lengths of molecular chains of derived tars. On the other hand, gasification and pyrolysis as processes in an industrial scale gasifier depend on the humidity, compactification, and oxygen content of the material as these determine speed, layer profiles in the bed and temperatures of the gasification process resulting in different distribution of hydrocarbons lengths in tars collected in the process and the temperatures and speed can seriously change the molecular distribution of derived hydrocarbons. These relations shall be

investigated more closely in future but clearly suggest the advantage of using torrefied material as preferred feed for gasification to reduce tar content in gas. Such approach was used in entrained flow gasification process for liquid fuel production developed in (Eberhard et al., 2020).

4.5 The gasification runs with different gasification gases

The system can operate on wood chips or pellets, with both air-steam mixtures and oxygen enriched gasification gases. We tested the process with dry wood chips at 20-

| | | Wood pellets | Torrefied pellets | Wood Chips |
|--|-----|--------------|-------------------|------------|
| Molecular component | | | | |
| Aliphatic | [%] | 0.1 | 0.3 | 0.9 |
| Acids | [%] | 66.9 | 59.3 | 63.9 |
| Aliphatic esters | [%] | 0.7 | 2.0 | 1.6 |
| Aliphatic aldehydes and ketones | [%] | 4.9 | 3.7 | 5.2 |
| Aliphatic alcohol | [%] | 9.3 | 7.3 | 3.9 |
| Alkylbenzenes | [%] | 0.1 | 0.3 | 1.2 |
| Alkylphenols | [%] | 6.7 | 13.5 | 10.6 |
| Furan (polyfunctional oxygen) | [%] | 3.4 | 1.8 | 1.8 |
| Linear and cyclic aliphatic oxygenates (polyfunctional oxygen) | [%] | 7.4 | 11.4 | 10.45 |
| Aromatic oxygenates (polyfunctional oxygen) | [%] | 0.1 | 0.1 | 0.05 |
| Nitrogen and sulfur heteroatoms | [%] | 0.4 | 0.2 | 0.2 |
| Total | [%] | 100 | 100 | 100 |
| Total organic content in water | [%] | 14.1 | 6.1 | 9.4 |

TABLE 7: GC-SM composition of water condenses.

25% humidity and average diameter of 5-20 mm. We have compared the producer gas quality during simple air gasification (Run 1) with mixture of air and steam (Run 2) and finally with steam-oxygen-air composition (Run 3); these are presented below in Table 9. These runs were conducted under high bed condition with temperatures of the gas leaving gasifier kept at 800-850K. Temperature distribution in the gasifier is presented on Figure 5 for air and air-steam gasification and on Figure 6 for oxygen-steam-air gasification runs.

We observe that in contrast to air and stem-air gasification the introduction of oxygen into the gasification process changes the dynamics of the process leading to elevation of temperatures in gasification and pyrolysis zones and as a result the process requires more effort to stabilize.

The gas parameters measured during these runs are presented in Table 8.

The amount of wood used in each test was dependent on the effectiveness of the gasification process as the level of material in the gasifying chamber was kept constant. In all tests the amount of oxygen in gasification gases was also kept constant and approximately equal to 150 kg/h not counting water in the biomass feeds in different combinations of air, steam and oxygen enriched air making the results directly comparable. We observed the difference in thermal output of each run showing the dynamics of the process measured in the effective amount of wood gasified during each test was different due to the different temperatures inside the bed and different reactiveness of the gasification gases. In this experiment ER=0.4 for air gasification, ER=0.42 for air-steam and ER=0.36 for oxygen-steam-air mixture. The contamination of the producer gas leaving the gasifier after the cyclone were analysed and are presented in Table 9 below for each run.

The following table presents the energy balance of the gasification process calculated by estimating the energy added to the system with fuel and hot gasification gasses and removed due to production, transport, and cooling of syngas.

It can be observed that the LHV of the clean producer gas for each run is in good agreement with experimental values from gas composition measurements as can be seen in Table 9.

The tar contents of the gas leaving the gasifier and the composition of tars in water collected in various stages of the purification process during each run has been



FIGURE 5: Distribution of temperatures in gasifier during air and air-steam gasification.



FIGURE 6: Distribution of temperatures in gasifier during oxygen-steam-air gasification.

TABLE 8: The parameters of the process and properties of the producer gas after purification.

| | | Run 1 | Run 2 | Run 3 |
|-------------------|-----------------|---------|----------|---------|
| Parameter | | | | |
| Thermal output | [MW] | 1.35 | 1.0 | 1.8 |
| Fuel | [kgh⁻1] | 410 | 310 | 480 |
| Air flow | [m³h-1] | 600 | 500 | 400 |
| Oxygen content | [%] | 21 | 21 | 25.8 |
| Steam flow | [kgh⁻¹] | 0 | 20 | 25 |
| Syngas Parameters | After Purificat | ion | - | |
| CO | [%] | 30,10 | 26.47 | 34.01 |
| H2 | [%] | 9.0 | 8.35 | 22.40 |
| CH4 | [%] | 2.67 | 1.80 | 4.14 |
| C02 | [%] | 8.42 | 7.60 | 16.40 |
| Syngas LHV | [MJm⁻³] | 5.0-5.5 | 4.0- 4.6 | 7.2-7.8 |

measured. Table 11 presents the composition of the cyclic hydrocarbons in water condensates obtained in various stages of the purification unit for oxygen-steam-air gasification as a general characterization of cooler water condensates has been done in the first test. The levels of cyclic hydrocarbons dissolved in water has been chosen as good indicators of changes of tars characteristics in the gasification process due to different gasifying gasses combination.

From the analysis of the effluents from each cooler it has been found that the selectivity of the system is low. Mostly water was collected in each cooler with only slightly different combinations of hydrocarbons. To prevent clogging the implemented pipe system is wide enough (40-60 mm) to allow gas in the central part of the cooler to remain hot, while water could condensate on the pipe surfaces. It has been decided to treat all the effluents from the three coolers and water scrubber as if from a single unit. The problem of improving selectivity of the coolers system will be addressed in a future work. The system was intendent for lowering the syngas temperatures and only a fraction of liquids contained in the gases condensate in the coolers. It shall operate in much lower temperatures to act effectively as liquids reductors in syngas which requires specific technical solutions. Afterwards the contamination levels of the producer gas leaving the oil scrubber have been measured and as it contains less than 100 mg/Nm³ of heavier tar and less the of 600 mg/Nm3 of light hydrocarbons mainly benzene, toluene and xylene it was suitable for use in gas engine unit.

TABLE 9: The producer gas contaminations.

| | Run 1 | Run 2 | Run 3 | | | |
|---|---|--------------------------------|--------------------------------|--|--|--|
| Substance | $\left[\frac{mg}{Nm^3}\right]$ | $\left[\frac{mg}{Nm^3}\right]$ | $\left[\frac{mg}{Nm^3}\right]$ | | | |
| Dust | 673.6 | 847.3 | 817.3 | | | |
| Benzene | 6050 | 12700 | 12810 | | | |
| Toluene | 2120 | 4890 | 2980 | | | |
| Xylene | 210 | 450 | 440 | | | |
| Sum of BTX | 8380 | 18000 | 16230 | | | |
| Heavy organic compounds | 1700 | 2120 | 1860 | | | |
| Naphthalene | 1.83 | 0.20 | 0.21 | | | |
| Acenaphthylene | 0.20 | 0.86 | 0.77 | | | |
| Acenaphthene | 0.05 | 0.18 | 0.17 | | | |
| Fluorene | 0.42 | 1.26 | 1.15 | | | |
| Phenanthrene | 2.63 | 7.24 | 6.43 | | | |
| Anthracene | 0.69 | 1.93 | 1.76 | | | |
| Fluoranthene | 1.22 | 4.09 | 4.86 | | | |
| Pyrene | 1.19 | 4.27 | 5.35 | | | |
| Benzo [a] anthracene | 0.33 | 0.92 | 1.24 | | | |
| Chrysene | 0.28 | 0.83 | 1.10 | | | |
| Benzo [b] fluoranthene | <0.01 | 0.80 | 1.66 | | | |
| Benzo [j] fluoranthene | 0.31 | 0.80 | 1.61 | | | |
| Benzo [k] fluoranthene | 0.11 | 0.31 | 0.48 | | | |
| Benzo [a] pyrene | 0.26 | 0.75 | 1.21 | | | |
| Indeno [1.2.3-cd] pyrene | 0.13 | 0.32 | 0.51 | | | |
| Benzo [ghi] perylene | 0.18 | 0.50 | 0.77 | | | |
| Sum of polycyclic aromatic hydrocarbon compounds | 9.82 | 25.25 | 29.27 | | | |
| * including polycyclic aromatic | * including polycyclic promotic bydrocarbon compounds | | | | | |

including polycyclic aromatic hydrocarbon compounds

The results clearly indicate that the application of the various combinations of oxygen, steam and air composition significantly influences the properties of the producer gas leading to significant growth of calorific value but not necessarily lowering the amount of tar and light hydrocarbons in the raw gases as is clearly seen at Table 6. We observe that the high selectivity of the tar removing system is more consistent with the application of gas engines as only the heavy hydrocarbons need to be removed but light hydrocarbons like benzene, xylene, toluene can be safely burned in the gas engine. In such cases use of dry torrefied or carbonised material as a feedstock can be a much simpler route for reducing the levels of contamination of producer gas. For a producer gas to be a source of fuel

| TABLE 10: Energy | balance | of the | process. |
|------------------|---------|--------|----------|
|------------------|---------|--------|----------|

| | Unit | Run 1 | Run 2 | Run 3 |
|--|-----------------------|-------|-------|-------|
| Enthalpy in fuel | [MJ/h] | 5945 | 4495 | 6960 |
| Net enthalpy loss in the system (including syngas cooling) | [MJ/h] | -1299 | -1035 | -1261 |
| Ethalpy loss in removed tar | [MJ/h] | -340 | -684 | -615 |
| Enthalpy in cold syngas | [MJ/m _N ³] | 5.10 | 4.08 | 7.28 |
| Process efficiency | % | 73 | 64.5 | 73 |

| TABLE 11: | The | hydrocarbon | concentration | in | coolers | condesate |
|------------|-----|-------------|---------------|----|---------|-----------|
| water (%). | | | | | | |

| Water condensate | Cooler 1 | Cooler 2 | Cooler 3 |
|-------------------------------|----------|----------|----------|
| Acetaldehyde | 0.015 | 0.027 | 0.021 |
| Acetone | 0.016 | 0.027 | 0.024 |
| Methacrolein | 0.002 | 0.003 | 0.003 |
| 2,3-butanedione | 0.010 | 0.019 | 0.018 |
| Butanal | 0.001 | 0.003 | 0.002 |
| 2-butanone | 0.005 | 0.011 | 0.009 |
| 2-butenal | - | 0.003 | - |
| 2-butanone, 3-methyl- | - | 0.002 | 0.001 |
| Benzene | 0.042 | 0.032 | 0.035 |
| 3-buten-2-one, 3-methyl- | 0.03 | 0.006 | 0.005 |
| 2-pentatone | - | 0.002 | 0.002 |
| 2,3-pentanedione | - | 0.003 | 0.002 |
| Toluene | 0.013 | 0.008 | 0.006 |
| Cyclopentanone | - | 0.002 | 0.002 |
| 1,3,5,7-cyclooctatetraene | 0.004 | 0.002 | 0.002 |
| 2-cyclopenten-1-one, 2-methyl | - | 0.001 | 0.002 |
| Phenyl, 2-methyl | 0.003 | 0.002 | 0.004 |
| Indene | 0.004 | 0.005 | 0.004 |
| Phenol, 2-methoxy- | 0.001 | 0.002 | 0.003 |
| Phenol, 2, 6-dimethyl | 0.001 | 0.001 | 0.003 |
| Creosol | 0.002 | 0.003 | 0.005 |
| Azulene | 0.003 | 0.007 | 0.006 |

for SOFC or hydrogen conversion the further work on the oxygen-air-steam gasification process and more promising catalytic tar reducing technology shall be tested and compared with the unit analysed here (Xue-Yu Ren et al., 2020).

5. CONCLUSIONS

We have conducted a series of tests for the steam-oxygen-air gasification installation and procedures with three different materials and three different gasification schemes with particular attention on the amount and characterisation of the tars and hydrocarbons produced in the process and present in the producer gas. We analysed the influence of pretreatment of biomass on the syngas quality indicating the influence of torrefaction on lowering the tar content of the gas. The oxygen enriched air combined with steam can leads to gas with LHV higher than 8 MJ/m³_N sufficient for use in gas engine or fuel cell for efficient electric energy generation. Our goal was to analyse the technical and thus economic aspects connected with the gas cleaning process required when the gas is to be applied for a more efficient and sophisticated way of application than burning and use of steam turbine cycle. There are many steam-oxygen gasification schemes recently tested (Broer et al., 2015; Kurkela et al., 2016; Dudyński, 2019) on laboratory or small-scale units with comparable results indicating the potential of this method to improve the technology of biomass gasification (Baláš et al., 2016). Application of the oxygen-steam-air combination improves significantly the LHV of the producer gas to the levels comparable with downdraft gasifiers (Kirsanovs et al., 2017) showing that such technology can be technically viable for effective heat and electricity generation. The main problem for this method of syngas improvement to be commercially applicable in small scale installations is the issue of reducing the cost of oxygen generation and disposal of wastewater and oil. Many improvements are under way: excess heat can be used for chip drying, waste oil, water and tars are to be used internally in the process for steam generation while the use of torrefied material or charcoal as a fuel is an interesting possibility for future long-time tests.

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REFERENCES

- Arena, Umberto & Di Gregorio, Fabrizio & Santonastasi, M.. (2010). A techno-economic comparison between two design configurations for a small scale, biomass-to-energy gasification based system. Chemical Engineering Journal. 162. 580-590. https://doi. org/10.1016/j.cej.2010.05.067.
- Baláš, M., Lisý, M., Moskalĭk, J., & Skála, Z. (2016). Steam Influence on Biomass Gasification Process. The holistic approach to environment, 6(3), 127-132. orcid.org/0000-0002-4968-1535
- Bang-Møller, C., & Rokni, M. (2010). Thermodynamic performance study of biomass gasification, solid oxide fuel cell and micro gas turbine hybrid systems. Energy Conversion and Management, 51(11), 2330-2339. https://doi.org/10.1016/j.enconman.2010.04.006
- Basu P. (2018) Chapter 6 Tar Production and Destruction. Biomass Gasfication, Pyrolysis and Torrefaction (Third Edition), 189-210. Academic Press https://doi.org/10.1016/B978-0-12-812992-0.00006-6
- Bocci E., Sisinni M., Moneti M., Vecchione L.,DiCarlo A., Villarini M. (2014).State of the art of small scale biomass gasification power systems: a review of the different typologies. Energy Procedia 45,247-256. https://doi.org/10.1016/j.egypro.2014.01.027
- Boerrigter,H., Calis H.P., D.J. Slort D.J., Bodenstaff H., Kaandorp A.J., den Uil H., Rab L.M. (2004) Gas Cleaning for Integrated Biomass Gasification (BG) and Fischer-Tropsch (FT) Systems. ECN-C-04-056 https://publicaties.ecn.nl/PdfFetch.aspx?nr=ECN-C-04-056
- Broer, K. M., Woolcock, P. J., Johnston, P. A., & Brown, R. C. (2015). Steam/oxygen gasification system to produce clean syngas from switchgrass. Fuel, 140, 282-292. https://doi.org/10.1016/j. fuel.2014.09.078
- Brunaccini G., Sergi F., Aloisio D., Ferraro M., Blesznowski M., Kupecki J., Motylinski K., Antonucci V., Modeling of a SOFC-HT Battery hybrid system for optimal design of off-grid base transceiver station, International Journal of Hydrogen Energy 2017;42(46):27962-27978, https://doi.org/10.1016/j.ijhydene.2017.09.062
- Cao, L., Yu, I., Xiong, X., Tsang, D., 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, 186, 109547. doi.org/10.1016/j. envres.2020.109547
- Chmielniak T, Sobolewski A, Bigda J, Iluk T. (2019) Biomass-Based Low Capacity Gas Generator as a Fuel Source. In Szubiel M, Filipowicz M. (Eds) Biomass in Small Scale Energy Applications: Theory and Practice. (Chapter 12) CRC Press
- Correa C, Kruse A, (2018). Supercritical water gasification of biomass for hydrogen production – Review. The Journal of Supercritical Fluids, Volume 133, Part 2, 573-590. https://doi.org/10.1016/j. supflu.2017.09.019

- Dudyński M., Kwiatkowski K., Bajer K. (2012). From feathers to syngas – technologies and devices. Waste Management 32. 685-691. https://doi.org/10.1016/j.wasman.2011.11.017
- Dudyński, M. C. van Dyk J., Kwiatkowski K., Sosnowska M. (2015). Biomass gasification: Influence of torrefaction on syngas production and tar formation. Fuel Processing Technology. Volume 131, , Pages 203-212. doi.org/10.1016/j.fuproc.2014.11.018
- Dudyński, M. (2018). Gasification of Selected Biomass Waste for Energy Production and Chemical Recovery. Chemical Engineering Transactions 65, 391-396. https://doi.org/10.3303/CET1865066
- Dudyński, M. (2019). Novel oxygen-steam gasification process for high quality gas from biomass. Detritus. Vol.06. pp.68-76. https://doi. org/10.31025/2611-4135/2019.13814
- Eberhard M., Santo U .Michelfelder B., Gunther A., Weigand P., Matthes J, Waiber P., Hagenmeyer V., Kolb T., (2020). The Bioliq Entrained-Flow Gasifier – A Model for the German Energiewende. ChemBioEng Reviews 7(5). doi.org/10.1002/cben.202000006
- Kirsanovs, V., Blumberga, D., Karklina, K., Veidenbergs, I., Rochas, C., Vigants, E., & Vigants, G. (2017). Biomass gasification for district heating. Energy Procedia, 113, 217-223. https://doi.org/10.1016/j. egypro.2017.04.057
- Kurkela, E., Kurkela, M., & Hiltunen, I. (2016). Steam–oxygen gasification of forest residues and bark followed by hot gas filtration and catalytic reforming of tars: Results of an extended time test. Fuel Processing Technology, 141, 148-158. https://doi.org/10.1016/j. fuproc.2015.06.005
- Kwiatkowski, K., Krzysztoforski, J., Bajer, K., & Dudyński, M. (2013). Bioenergy from feathers gasification–Efficiency and performance analysis. Biomass and bioenergy, 59, 402-411. DOI: 10.1016/j.biombioe.2013.07.013

- Liu, L., Huang, Y., Cao, J., Liu, C., Dong, L., Xu, L., & Zha, J. (2018). Experimental study of biomass gasification with oxygen-enriched air in fluidized bed gasifier. Science of The Total Environment, 626, 423-433. https://doi.org/10.1016/j.scitotenv.2018.01.016
- Marcantonio, V.; Monarca, D.; Villarini, M.; Di Carlo, A.; Del Zotto, L.; Bocci, E (2020). Biomass Steam Gasification, High-Temperature Gas Cleaning, and SOFC Model: A Parametric Analysis. Energies , 13, 5936 doi.org/10.3390/en13225936
- Molino A, Larocca V, Chianese S, Musmarra D. Biofuels Production by Biomass Gasification: A Review. Energies. 2018; 11(4):811. https://doi.org/10.3390/en11040811
- Pongratz, G., Subotić, V., Schroettner, H. et al. (2021) Investigation of solid oxide fuel cell operation with synthetic biomass gasification product gases as a basis for enhancing its performance. Biomass Conv. Bioref. 11, 121–139 doi.org/10.1007/s13399-020-00726-w
- Sikarwar V.S., Zhao M., Paul S. Fennell P.S., Anthony E., (2017). Progress in biofuel production from gasification. Progress in Energy and Combustion Science 61, 189-248. https://doi.org/10.1016/j. pecs.2017.04.001
- Vreugdenhil B.J., Zwart R.W.R. (2009). Tar formation in pyrolysis and gasification. ECN-E-08-087 https://publicaties.ecn.nl/PdfFetch. aspx?nr=ECN-E--08-087
- Xue-Yu Ren, Xiao-Bo Feng, Jing-Pei Cao, Wen Tang, Zhi-Hao Wang, Zhen Yang, Jing-Ping Zhao, Li-Yun Zhang, Yan-Jun Wang, and Xiao-Yan Zhao. (2020) Catalytic Conversion of Coal and Biomass Volatiles: A Review. Energy & Fuels 34 (9), 10307-10363. https:// doi.org/10.1021/acs.energyfuels.0c01432