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Torrefaction and gasification of biomass for polygeneration: Production of biochar and producer gas at low load conditions

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Abstract

In this paper, a matter of biomass torrefaction and gasification is closely looked at from different points of view during low load and low equivalence ratio regime, defined as $\lambda = 0.08$. Considering gas production, the hot gas efficiency of conversion (30%) and its energy content (4.14 MJ m^{-3}) were not quite satisfying, however, this matter of fact was compensated by an interesting yield of biochar. This material was generated in 0.387 and 0.314 rates for torrefaction and torrefaction + gasification processes, respectively, which, in both cases, represents an attractive, alternative approach to the functional energy storage. It was determined that a CO_2 offset of 721 kg and 660 kg could be achieved per 1 tonne of woodchips for gasification of raw woodchips and a 2-stage process with torrefaction and gasification, respectively. The measured data from both technological complexes were compared with the computational model, applying equilibrium reactions for gas components determination. In addition, the question of tar compounds contained within the producer gas, is investigated through GC–MS analysis on both qualitative and quantitative basis.

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Keywords: Gasification; Torrefaction; Biochar; Syngas; Tar compounds; Computational model

1. Introduction

One of the challenges, for mankind, in the upcoming decades will be the joint effort to decrease emissions of GHG in order to comply with the Paris Agreement, signed in multilaterally by the majority of the world countries (United Nations, 2016). Achieving such an ambitious goal requires a difficult transition towards decarbonisation and

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Nomenclature

BECCS	bioenergy carbon capture and storage
GHG	greenhouse gases
λ	equivalence ratio
ERC	Energy Research Centre
LHV	lower heating value
HHV	higher heating value
SRF	solid recovered fuel
HGE	hot gas efficiency
GC–MS	gas chromatography with mass spectrometer analysis
TC	tar compounds
MTR	multistage tape reactor
NDIR	non-dispersive infra-red
TCD	thermal conductivity detector

an increased share of renewables in the energy mix [1]. One of the established routes towards decarbonisation is the use of biomass, which is an important renewable energy source with significant potential in Europe [2,3].

BECCS is a way that could potentially offer a significant decrease in CO₂ emissions, reaching negative emissions and carbon offset [4,5]. Some BECCS solutions using gasification technologies have been proposed [6]. Some authors also indicated the possibility of integrating CO₂ capture prior to combustion [7]. However, such solutions could only work on a big scale. Some works proposed smaller co-generation plants, using 1.5 MW of biomass and 1.4 MW of biogas, which could achieve installed power of 750 kW_e and 600 kW_t of heat and capture 1620 tonnes of CO₂ per year at the same time [8]. Nonetheless, the issue of logistics between capture point and storage for such small-scale installations remains unsolved. On the other hand, biochar can be considered an effective form of long-term carbon storage in soil [9–11]. Moreover, it could offer essential benefits in terms of soil properties, such as water holding capacity, wettability and organic matter content [12–14] thus resulting in improved yields [15].

Regarding biochar production and application, one of the crucial aspects is the value chain and all the constraints on its economic feasibility due to logistics aspects, such as feasible transportation distance and storage requirements [16]. Moreover, the scale of the installation, cost of biomass, price of electricity, as well as quantity and quality of produced gas are critical for achieving profitability in the gasification of biomass [17]. Torrefaction of biomass is considered one of the feasible ways of improving fuel logistics [18,19] and could also help with respect to biomass cost, as it is a viable way for the valorisation of low-quality biomass [20–23]. Some also consider torrefaction a viable way to produce biochar for subsequent use as a soil amendment [24,25].

A limited amount of research has been published so far on gasification at low load conditions with low λ [26]. However, flexibility is an important issue from the point of view of energy systems [27,28], and gasification installations could play significant roles in flexible systems by helping to balance supply from intermittent energy sources, such as solar or wind [29,30].

Low load operation with low λ , performed during the gasification process, could be considered a chance for a flexible polygeneration installation that would co-produce energetic gas and biochar when supply from intermittent sources is high. Such installation would be capable of achieving negative CO₂ emissions and potential benefits related to CO₂ offset since such biochar could be applied to soil and considered a carbon sink and carbon storage. However, the equivalence ratio significantly influences the production of tars during gasification [31], which is problematic for the final use of producer gas [32]. Therefore, the aim of this paper is an assessment of two different configurations of such installation, using experimental data from commercial-scale units:

- Straightforward gasification of wood biomass, without pre-treatment
- Gasification of torrefied wood biomass

Such assessment has been done within the scope of this work, considering biochar productivity for each configuration, as well as the quality of producer gas, focusing especially on the production of heavy tars. These are

problematic due to high melting temperatures that cause problems concerning tar deposition [26]. Furthermore, modelling of gasification and its validation using producer gas composition from the commercial-scale installation has also been included within the scope of this work, in order to provide viable prediction capabilities for gasification process under low λ operation.

2. Materials and methods

2.1. Biomass and its torrefaction

The fuel material used for the torrefaction and gasification experiments was beechwood chips, with a top size of 20 mm. Torrefaction was performed in a patented MTR. The reactor is described in more detail in other works [33,34]. Woodchips were roasted continuously, utilising the heat from the combustion of torgas. The heat was delivered to the woodchips by the top surfaces of heated hollow shelves. The heat was delivered to the heated surfaces by hot flue gases. Woodchips were moved successively through the reactor using a set of scraper arms attached to chains. Torrefaction was performed under an oxygen-depleted atmosphere, only with negligible amounts of air going through airlocks with the feedstock. The average temperature, measured inside of the hollow shelves at the top, was approx. 300 °C while the average residence period of the wood chips was approx. 45 min. For the start-up of the technology and to ensure suitable combustion, a simple oil burner was used. The composition of permanent gases in torgas was measured using Gas 3100R analyser (Pollutek Gas Analysis, Belgium), which uses NDIR sensors for measurements of CO₂, CO, and CH₄, a TCD sensor to measure H₂, and an electrochemical sensor to measure O₂ content.

Both raw wood chips, remaining after torrefaction, as well as torrefied woodchips, were stored in big bags in the semi-open shed. Compositions of raw and torrefied woodchips are shown in Table 1.

Table 1. Composition of raw and torrefied woodchips.

	HHV ^d MJ kg ⁻¹	FC ^d % wt.	VM ^d % wt.	W ^r % wt.	A ^d % wt.	C ^d % wt.	H ^d % wt.	N ^d % wt.	S ^d % wt.	O ^d % wt.
Raw wood chips	20.1	14.8	84.0	22.0	1.2	49.7	6.1	0.2	0.1	42.7
Torrefied wood chips	23.5	32.9	64.0	10.4	3.1	59.5	5.6	0.2	0.1	31.5

2.2. Gasification – experimental investigation

The process of gasification is significantly dependent on input material type and quality, gasification parameters and technology design [35]. The ERC gasification technology is equipped with a 200 kW energy input reactor with a sliding bed over the circular grate. It was constructed for operation with air as oxidising media and fuel of 1–5 cm grain size. Its thermal output is around 50 kW, and the generated heat is utilised for thermochemical reactions – an autothermal regime with partial combustion. Feeding of the fuel is realised by two synchronised screw conveyors which deliver it above the grate area. The oxidiser is brought by four tangential nozzles (Fig. 1) and the producer gas is carried away in the upper part of the reactor. Thus, the design of the reactor is best described as cross/updraft. The flow of the media is realised by a suction fan situated at the tail of the technology. The spent fuel (char) falls spontaneously into the collection vessel. The producer gas is subsequently purified using a cyclone barrier and filtration tissues of a hot filter for particulate matter separation. TC (organic compounds with a molecular weight greater than benzene) and gas moisture are then separated in water condensers of tube-in-tube design with 6–8 °C operating temperature. The monitoring of the course of the gasification and its control are realised by a virtual interface and programmable platform.

The technology is capable of producing up to 40 m³ h⁻¹ (273 K, 101325 Pa) of clean and moisture-free producer gas while the inner reactor temperature is around 1120 ± 100 K. The value of λ ranges from 0.05 to 0.35 and *LHV* of the producer gas from 3.5 to 5.5 MJ m⁻³, both values dependent on operation conditions and source fuel. The *HGE* parameter (sensible heat of the gas included) reaches up to 90%. Several experimental measurements were performed on this technology in the past, using softwood pellets [36] or SRF [37].

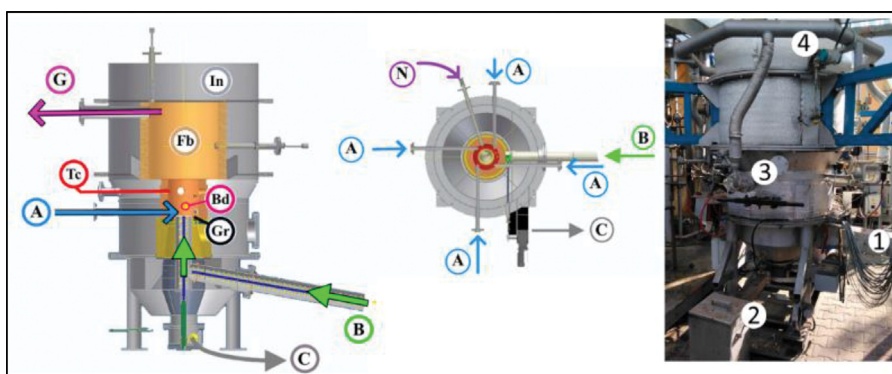


Fig. 1. ERC gasification technology — left: cross-section — view from the side; centre: cross-section — view from the top; right — photo of the unit (G — gas; A — air; N — natural gas burner for a start-up; B — biomass; C — biochar; Tc — thermocouple; Gr — grate; Bd — bed; Fb — freeboard; In — insulation; 1 — biomass auger; 2 — char removing auger; 3 — air supply; 4 — air supply manifold).

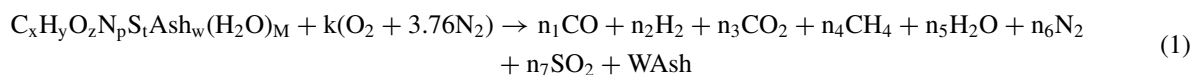
2.3. Producer gas quality investigation

In this experiment, a sample of the gas was taken from the pipeline through a sampling hole using a heated probe. The sampled gas was led through a set of three impinger bottles, each filled with 100 ml of propane-2-ol and emerged in an electric cooler SD 07R-20 (PolyScience, USA) with propylene glycol. The temperature in the cooler was held at $-10\text{ }^{\circ}\text{C}$ in order to condensate and separate TC from the producer gas. The composition of the sampled gas was investigated using Gas 3000p (Pollutek Gas Analysis, Belgium), placed behind a flowmeter and a suction pump. Each sampling procedure took exactly 20 min during a stable regime operation. A volume of the gas passed through the set of impinger bottles and was measured with a gas meter.

The tar samples were subjected to GC–MS analysis. The analytical setup consisted of a 7820-A chromatograph and a 5977B MSD mass spectrometer (both Agilent, USA). Samples of $2.0\text{ }\mu\text{l}$ were introduced into the GC injector ($250\text{ }^{\circ}\text{C}$; split = 20) via autosampler; helium was used as a carrier gas (1.0 ml min^{-1}). The GC was equipped with an HP-5MS column ($30\text{ m} \times 0.32\text{ mm} \times 0.25\text{ }\mu\text{m}$) (Agilent, USA). The oven temperature profile was as follows: holding at $50\text{ }^{\circ}\text{C}$ for 5 min, then increasing with $5\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$ to $250\text{ }^{\circ}\text{C}$, then increasing with $10\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$ to $300\text{ }^{\circ}\text{C}$ and holding the temperature for 15 min. Compound identification was performed automatically. This was done by comparing the mass spectra with the NIST-14 MS library (with the minimum match factor equal to 90%). The MS scanning range was m/z 10–450 with the frequency of 1.7 scan sec^{-1} . The quadrupole and MS source temperatures were 150 and $230\text{ }^{\circ}\text{C}$, respectively. While the GC–MS was applied mainly for qualitative analysis of the tars, for the chosen compounds, the quantitative analysis, based on calibration curves, was performed. All the GC analyses were repeated four times, with the presented results being average values.

2.4. Mathematical model of the gasification process

The gasification process can be represented by the global reaction defined in the following way:



where: x, y, z, p, t, W, M – molar masses of elements and components per 1 kg of feedstock in $[\text{mol kg}^{-1}]$, k – a molar mass of air in $[\text{mol kg}^{-1}]$ and $n_1, n_2, n_3, n_4, n_5, n_6,$ and n_7 – molar masses of syngas components after gasification in $[\text{mol kg}^{-1}]$. Char and tar formation are neglected. Molar masses of fuel components are calculated based on the fuel ultimate analysis, whereas syngas components are calculated based on two equilibrium reactions:



where the equilibrium constant is equal to:

$$K_1 = \frac{f_{\text{H}_2} \times f_{\text{CO}_2}}{f_{\text{CO}} \times f_{\text{H}_2\text{O}}} \quad (3)$$

f_{CO} , $f_{\text{H}_2\text{O}}$, f_{H_2} , f_{CO_2} – mole fractions of CO, H₂O, H₂, and CO₂ in [% mol]. And the second reaction:



where the equilibrium constant is equal to:

$$K_2 = \frac{f_{\text{CH}_4}}{f_{\text{H}_2}^2} \times \frac{1}{p} \quad (5)$$

f_{CH_4} , f_{H_2} – mole fractions of CH₄ and H₂ in [% mol], p – operating pressure in [atm]. Mole fraction of the particular compound f_i is defined as follows:

$$f_i = \frac{n_i}{\sum_1^7 n_i} \quad (6)$$

Equilibrium constant K is also a function of temperature and is related to the standard Gibbs free energy change of reaction in the following way:

$$\ln(K) = -\frac{\Delta G^0}{RT^2} \quad (7)$$

Therefore, for the two applied equilibrium reactions, equilibrium constants will take the following form:

$$\ln(K) = \frac{-\Delta G^0}{RT_0} + \frac{-\Delta A}{R} \left(\frac{1}{T_0} - \frac{1}{T} \right) + \frac{-\Delta B}{R} \ln\left(\frac{T}{T_0}\right) + \frac{-\Delta C}{R} (T - T_0) + \frac{-\Delta D}{2R} (T^2 - T_0^2) + \frac{-\Delta E}{3R} (T^3 - T_0^3) \quad (8)$$

where ΔA_i , ΔB_i , ΔC_i , ΔD_i and ΔE_i , are calculated for the two equilibrium reactions. For example, $\Delta A_1 = A_{\text{CO}} + A_{\text{H}_2\text{O}} - A_{\text{CO}_2} - A_{\text{H}_2}$. The A_{CO} , $A_{\text{H}_2\text{O}}$, A_{CO_2} and A_{H_2} parameters are the regression coefficients taken from [38]. Eventually, based on mass and energy balances and equilibrium constant relations, a system of equations is solved that allows obtaining the unknown syngas components after the gasification process.

2.5. Biochar production assessment by the indirect method

In commercial-scale torrefaction reactors and gasifiers, direct estimation of productivity of torrefied biomass and biochar is impractical as it is impossible to quench the reaction immediately and empty the reactor completely. Therefore, it was decided that mass balance would be performed by indirect method. The mass yield was assessed using the well-established ash tracer method, which is often used for indirect assessment of mass losses during various thermal conversion processes [39,40]:

$$Y_m = \frac{A_{\text{feedstock}}}{A_{\text{product}}} \quad (9)$$

where: Y_m – mass yield, -; A – respective ash content of feedstock and product, dry basis, % wt.

3. Results and discussion

3.1. Production of gas in gasification and torrefaction

The composition of the torgas, obtained during torrefaction of 2 cm wood chips in the MTR torrefier, is shown in the diagram in Fig. 2 below. Both calculated heating values (HHV and LHV) were very close to 5 MJ m^{-3} . It could be assumed that the real heating values were higher, as calculations were based on the content of measured permanent gases. Torgas from torrefaction of beech wood can contain significant amounts of condensable compounds, such as acetic acid or formic acid [41]. Also other gaseous compounds, not measured in this study, such as formaldehyde [41,42] or methanol [41] have been reported in the literature. This suggests that actual calorific values of torgas were actually higher. This indicates a relatively straightforward combustibility of such torgas, thus allowing to maintain the torrefaction process using the combustion of torgas as the main heat source. Oxygen was detected in torgas (Fig. 2), suggesting that small amounts of air got into the reactor during the process.

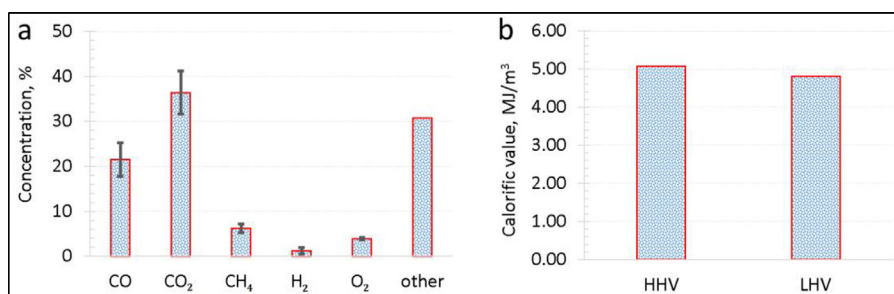


Fig. 2. Parameters of wood chips torgas. (a) chemical composition; (b) *HHV* and *LHV*.

Table 2. Process parameters during gasification experiments.

Parameter	Symbol	Unit	Value	
Fuel	[–]	[–]	Raw chips	Torrefied chips
Temperature	T	[K]	1127	1137
Relative pressure	p	[kPa]	–0.2	–0.2
Fuel flow	\dot{m}	[kg h ^{–1}]	45	33
Air flow rate	Q_a	[m ³ h ^{–1}]	13	14
Equivalence ratio	λ	[–]	0.080	0.083

The process of gasification was performed in two individual measurement rounds, one for each source fuel. The temperature of the process was held between 1100 and 1180 K, while relative pressure was around –0.2 to the atmospheric pressure. As seen in Table 2, the temperature was not very dependent on the used fuel or its flow. The λ was relatively low compared to other literature [43–46], as its value was equal to 0.080 for raw wood chips and 0.083 for torrefied wood chips. Despite such low λ in both experiments, obtained producer gas contained in both cases a relatively low share of combustible gases (CO, H₂, CH₄), as shown in Fig. 3. Moreover, it could be reasonably assumed that the compounds marked in Fig. 3 as “other” consisted mainly of N₂. This could be explained by the relatively poor efficiency of the process (see Table 3 below), which could be mainly attributed to high biochar productivity, which will be discussed in more detail in Section 2.3. Nonetheless, it should be noted that actual λ could be a bit higher in both raw and torrefied chips gasification, as the calculated value was based on measured flow rates of air and fuel (Table 2), and it is plausible to suspect that additional air is drawn into the reactor, through fuel auger and ash removing auger (Fig. 1).

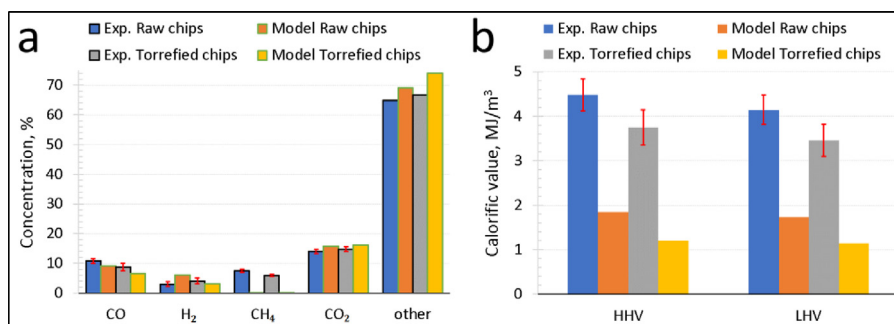


Fig. 3. Comparison of experimental results and model for (a) composition of the producer gas; (b) *HHV* and *LHV* of the producer gas.

The experimental measurements of raw and torrefied biomass have been additionally compared with the mathematical model described in Section 2.4. The achieved results are pictured in Fig. 3 for raw and torrefied biomass, respectively. For the given fuel properties and operating conditions, the model prediction of the concentration of CO, H₂, CO₂, and other compounds, such as N₂, is reasonable. However, the presented approach poorly estimates the CH₄ concentration for both raw and torrefied biomass. Based on the experimental data, the syngas from the torrefied

Table 3. Results of gasification experiments for raw and torrefied chips — efficiency of the process and gas productivity.

Parameter	Unit	Value	
		Raw chips	Torrefied chips
Fuel			
Producer gas flow rate	kg h ⁻¹	49.6	23.2
Hot gas efficiency	%	30	20
Cold gas efficiency	%	22	14

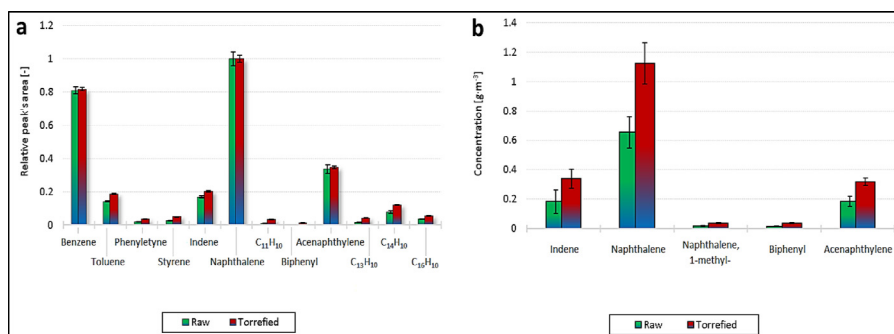
feedstock has a slightly lower calorific value than the gas from the raw biomass. It results from the lower CO and CH₄ concentrations. A similar result was obtained from the model. The main difference was caused by the model underestimating the production of methane. Due to the fact that methane has a significantly higher heating value by volume in comparison to CO or H₂, even a couple of percentage points less CH₄ in the producer gas contributed to the significant difference between experimental and model values in terms of both, HHV and LHV.

The efficiency parameters of gas production occur to be poor in both cases. Surprisingly, in the case torrefaction was applied, its value decreased (Table 3 below). The presented values are quite below standard, which may be a consequence of relatively high moisture content within feedstock. $LHV = 4.14$ and 3.45 MJ m⁻³ for raw and torrefied wood chips, respectively, also propose quite poor characteristics of the producer gas generated from these materials. In Fig. 3, the effect of torrefaction on the content of flammables in the gas is also seen. The material suffered a 21 and 19% vol. loss in the content of CH₄ and CO while H₂ was increased by 34% vol. with torrefaction. Thus, the process of torrefaction seems to be beneficial not from the overall gas quality, but from the individual compound content point of view, especially in terms of H₂ production.

3.2. Analysis of the gas quality — characterisation of tars

Results of the qualitative GC–MS analysis of tar samples obtained during gasification of raw and torrefied wood chips are presented in Fig. 4(a). The results show relative peaks' areas in relation to the most significant peak (the naphthalene peak). While these results do not represent a straightforward share of the identified compounds, they indicate which compounds dominate and, most importantly, how the torrefaction affected the TC distribution. It should be noted that the area of the share of all identified compounds is approximately 99.9% of all the peaks obtained during GC–MS analyses. This value includes heavier compounds and benzene, which is usually not even considered a TC. In Fig. 4(a), C₁₁H₁₀ stands for the sum of both naphthalene, 2-methyl- and naphthalene, 1-methyl-. C₁₃H₁₀, C₁₄H₁₀ and C₁₆H₁₀ represent the sum of possible different aromatic isomers.

Fig. 4(b) shows the quantitative analysis of some of the heavier aromatic TC present in the obtained samples. Together with the qualitative analysis, the results indicate that torrefaction increased the share and amount of 3rd class (one ring aromatics) and 4th class (two-three ring aromatics) tars [47]. The influence of torrefaction on tar production is ambiguous as it can result in both an increase or decrease of their concentration [48,49]. While the potential decrease in tar content can be explained by changes in the chemical composition of the biomass due to

**Fig. 4.** (a) qualitative analysis of tar samples, with peak areas normalised to naphthalene; (b) Quantitative analysis of selected compounds.

torrefaction, the increase can be a result of locally and temporally different conditions within the reactor fed by torrefied biomass [49].

3.3. Production of biochar

It has been clearly shown that operation under low λ is a viable way of producing significant amounts of biochar. Mass yields for the processes are shown in Table 4. Relatively high productivity of biochar in both cases is also a reasonable explanation for relatively poor efficiency of gasification, which could be attributed to high amounts of chemical energy that remained in biochar (for instance, C content up to 85% wt.). This coincides well with the low load operation of the gasifier, which could be needed in the time of high supply of energy from intermittent renewable energy sources. This also means that biochar production should be considered an important co-product of highly flexible, bio-based energy systems.

Table 4. Solid product yields (dry) during torrefaction and both gasification experiments.

Parameter	Unit	Value		
Fuel		Torrefaction	Gasification of raw woodchips	Gasification of torr. woodchips
Mass yield	–	0.387	0.341	0.922

A comparison of biochar production using single and two-stage production routes is shown in Fig. 5. It is clearly seen that there is only a slight difference between the amount of biochar produced in a single-stage route (gasification) in comparison to a two-stage route (gasification + torrefaction). This could be attributed to the relatively high yield of biochar during the gasification of torrefied chips. Such high yield could be explained by the influence of torrefaction on physical parameters, i.e., by shrinkage of chips during torrefaction and increased brittleness of the particles. Increased brittleness could increase the chances of broken particles in the auger. Combined with shrinkage, this could cause particles to pass the grate relatively quicker, thus providing smaller residence time in the reactor.

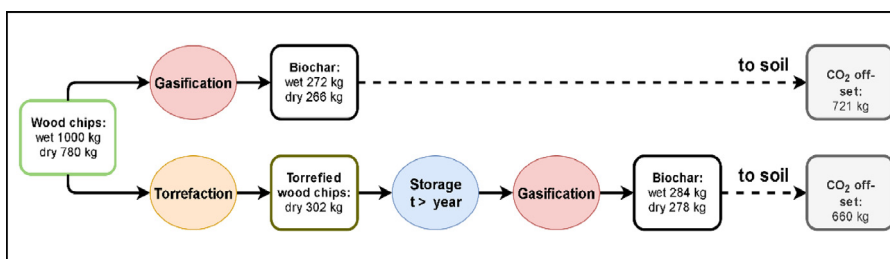


Fig. 5. Mass balance for two variants of biochar production using wood chips, including achievable CO₂ offset.

However, in terms of biochar productivity, such a situation could be considered advantageous since the productivity could be maintained. As a consequence, gasification of torrefied wood chips yielded 23.2 kg h⁻¹ of producer gas, in comparison to 49.6 kg h⁻¹ of gas for gasification of raw wood chips. This shows that for relatively similar λ significantly lower load could be achieved with gasification of the torrefied biomass, which is favourable when it comes to lowering the minimum attainable load during low grid load and high supply from intermittent renewables. Thus, it seems plausible to suspect that dual fuel operation of the gasifier and switching between raw and torrefied woodchips depending on the forecasts on the grid load and supply from wind and solar could be a viable way of operation for such installation. It should not be overlooked that the positive influence of torrefaction on sustaining long-term moisture on a relatively lower level, compared to raw woodchips, is a significant advantage from the point of view of fuel logistics, especially fuel storage. This is important since moisture has a significant impact on the LHV of a fuel [50]. Overall, for the case of gasification of raw woodchips, a CO₂ offset of 721 kg could be achieved, whereas for a two-stage system consisting of torrefaction and gasification, an offset of 660 kg of CO₂ could be achieved per 1000 kg of feedstock on an as-received basis. Future works should investigate the gasification of torrefied and subsequently pelletised wood, as it would allow obtaining much higher volumetric energy densities, thus achieving much higher power for the same operational volume of the gasification reactor.

4. Conclusions

The capability to produce a considerable amount of biochar during low λ operation coincides well with the low load operation of the gasifier, which could be needed in the time of high supply of energy from photovoltaics, wind or other intermittent renewable energy sources. This also means that biochar production should be considered an important co-product of highly flexible, bio-based energy systems, which is well-suited for small-scale gasifiers.

Considering the amount of produced biochar, torrefaction does not seem well justified, as similar productivity of biochar was achieved for both routes. However, for similar λ significantly lower load could be achieved with gasification of the torrefied biomass, which is favourable when it comes to lowering the minimum attainable load during low grid load and high supply from intermittent renewables. The quality of the gas was not much different in terms of the content of most problematic tar compounds when compared with the operation of the gasifier with higher equivalence ratios. Most certainly, such a gasification-based system would require a robust gas cleaning system. Looking at the heating values, it seems plausible to suspect that engines would require some customisation prior to operation using such gas as a fuel, which would likely require operations using lean producer gas–air mixtures. Significant CO₂ offset is possible with such an operational regime of the system.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that has been used is confidential.

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