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Presented during the French Chemical Engineering Congress SFGP 2019, Nantes 15–17 October 2019**

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Energetic study of beech wood gasification in fluidized bed reactor under different gasification conditions.

Luis Reyes^a, Lokmane Abdelouahed^a, Jean-Christophe Buvat^a, Ballydia Campusano^a, Christine

Devouge Boyer^b, Bechara Taouk^{a*}

^a Normandie Univ, INSA Rouen Normandie, UNIROUEN, Laboratoire de Sécurité des Procédés Chimiques, LSPC EA-4704, 76000 Rouen, France

^b Normandie Univ, INSA Rouen Normandie, UNIROUEN, COBRA UMR-6014, 76000 Rouen, France

* bechara.taouk@insa-rouen.fr

Abstract.

The thermodynamic study of beech wood gasification was performed in a pilot scaled fluidized bed reactor at different operation conditions, including temperature range from 600°C to 900°C, using CO₂ and steam as gasification agents and sand and biochar as bed materials. The particularity of this study was the evaluation of energy consumption of biomass gasifier based on energy balance comparison in the same experimental set-up for different experimental conditions. The comparison between pyrolysis and gasification with CO₂ showed that pyrolysis required less heat input (4.4 and 5.0 MJ/kg_{biomass}) than gasification (6.7 and 7.8 MJ/kg_{biomass}) at 800 and 900°C respectively. Meanwhile the syngas CGE were higher for gasification (0.78) as the LHV values for syngas (12.2 MJ/kg). Temperature favoured CGE and syngas LHV increase, as it also increased process endothermicity. The presence of biochar as bed material increased the syngas LHV from 3.3 to 15.4 MJ/kg for CO₂ gasification and from 3.3 to 15.1 MJ/kg for steam gasification. Both gasification agents provided similar values of CGE (>0.95) at 900°C. From 600 to 900°C the required heat input for steam gasification it was from 1.5 to 6.9 MJ/kg_{biomass}, meanwhile for CO₂ gasification was from 4.1 to 7.1 MJ/kg_{biomass}. The results offer useful details that can help for future design of gasification experiments in fluidized bed reactors.

1. Introduction

As biomass has for many years been the focus of attention to battle our dependence on fossil fuels, numerous technologies of biomass transformation are currently used (Hamelinck and Faaij, 2006). Gasification in particular, consists of the thermal decomposition of biomass into a variety of products, such as biochar, tars and syngas, among which syngas is the most desirable. Syngas from biomass gasification is constituted principally of CO, H₂, CH₄ and small quantities of light hydrocarbons, such as C₂H₂, C₂H₄, C₂H₆, C₃H₄, C₃H₆ and C₃H₈ (henceforth referred to as C₂+ and C₃+ in this work). The yield of products might vary depending of the nature of biomass, operating conditions, gasification agents and all variables interacting in the process. The type of reactor and its dynamics are also important parameters influencing gasification results (Roddy and Manson-Whitton, 2012). Amid the existing reactors used for gasification, fluidized bed reactors demonstrate interesting performances for heat and mass transfers between gaseous and solid phases (Chen et al., 2005; Wang et al., 2019).

Energy comparisons were found in literature concerning gasification technologies (van der Meijden et al., 2010). The Authors compared energy balance and process efficiency of three type of gasifiers, such as entrained flow, fluidized bed and allothermal reactor. It was concluded that allothermal and fluidized bed reactor were more energetically efficient (67% and 59%, respectively) than entrained flow gasifier (54%). The authors attributed this difference to the advantage in heat transfer and reactor gasification routines. The gasification agents used were a mixture of CO₂/Steam/O₂ as the interest of the authors was to produce synthetized natural gas. Biomass origin is an important parameter take into account in energy comparison, since the composition of biomass varies widely from one type to another. Rao and colleagues (Rao et al., 2004) compared wood chips with municipal and sun-dried soybean straw residues in a fixed-bed reactor. They concluded that residues showed similar CGE values of 73%, meanwhile wood chips CGE was 65%. Beside this fact, the global energy content of the produced syngas from residues and wood chips was very close, approximately 12.2 MJ/kg. The authors proposed empirical stoichiometric equations of partial oxidation of biomasses, it was also argued that the use of other gasification agent could vary the observed trends in the study.

Gasification is generally performed with controlled amounts of air, pure oxygen, steam or carbon dioxide. The latter agent has been less studied (Renganathan et al., 2012; Sadhwani et al., 2016) in

comparison to the former ones. Some of the reasons why CO₂ is less studied include need of external heat source that has to be added to the system, because no partial combustion of the biomass is reached like it is for air and oxygen gasification. The calculations of external heat source depend of the mass and energy balance, which involves thermodynamics notions and analysis. Thermo-chemical conversions as gasification and pyrolysis are highly endothermic (Dufour, 2016; Hosseini et al., 2012; Park et al., 2018). The use of the term endothermic did not mean that only endothermic reactions took place in gasification. As gasification is known as an intermediate step between pyrolysis reaction and combustion, exothermic reactions are also present in the process. Methane formation, water-gas shift and methanation reactions are some of the exothermic reactions taking place in the entire process (Pohořelý et al., 2014). These reactions energetically help to sustain gasification process, despite this the required energy amount to perform gasification remains elevated. Consequently, the global process is considered endothermic.

Thermodynamic analysis is usually performed in gasification systems, in order to provide detailed information of the energy of gasification products, as well as to provide information about the design, optimization and performance prediction of gasification systems (Rupesh et al., 2016). Parvez and colleagues (Parvez et al., 2016) compared thermodynamic values obtained from CO₂ and steam gasification; the latter was referred to in his work as conventional gasification. The author used computational software to simulate the gasification installation and perform energy calculations, showing that CO₂ gasification provided higher energetic values than steam gasification. It is noteworthy than in his work, only syngas was evaluated from the output streams of the system. As the gasification system in his study was a combination of several operation units, including a decomposer and a solids separator, only syngas was the final product. Several thermodynamic analyses are usually performed for the whole gasification system (Gu et al., 2019), including operation units (e.g. dryers, separators). Simulation softwares only considers physical energy of compounds, it does not consider chemical values. For this reason, the use of simulations softwares to discuss process required energy could be disputable. To overcome this issue, the chemical value of energy for all gasification compounds must be added by programming or using additional tools.

As the main aim of gasification processes is to produce syngas for the purpose of power generation, the direct focus of thermodynamic analysis is syngas. Tar represents an important issue, due to the complexity of its removal from syngas. The use of biochar as a catalyst has been employed to reduce tar and boost syngas production (Abu El-Rub et al., 2004). Other authors (Buentello-Montoya and Zhang, 2019) evaluated the steam gasification of biomass integrated with a biochar catalytic bed in a simulated two-stage gasifier. Energy analyses were performed at different temperatures and equivalence ratios. The author demonstrated the relevance of using biochar as a catalytic treatment for tar, showing the increases in thermodynamic efficiency of the syngas produced.

In literature, several researches have been conducted comparing the energy balances from different gasification systems (Hosseini et al., 2012; McCaffrey et al., 2019; Sreejith et al., 2013), even though in some case the experimental set-up and operating conditions were not exactly the same. This fact renders a proper comparison difficult.

The aim of the present study was to present a detailed comparison of the thermodynamic influence of different gasification conditions used for syngas production: use of CO₂; use of N₂; and use of steam (H₂O). The use of sand and biochar as bed material was also evaluated. This type of comparison was difficult to find in literature as the experimental set-up and conditions were not strictly the same. In this study was presented a strict normalized comparison in the same experimental set-up (fluidized bed reactor), accounting all the streams entering and exiting the gasifier. To the extent of our knowledge, only limited information can be found in the literature concerning gasification energy comparison in fluidized bed reactors. Thus, the goal of this work is to provide some details concerning these factors. Furthermore, the energetic efficiency of syngas was also evaluated.

2. Experimental Section

2.1. Materials used

The biomass selected for this study was beech wood, obtained from Ooni Corporation UK. The biomass was dried in an oven at 100°C and kept for one hour before utilization. It was also grinded and sieved to obtain an average particle size of 6 mm. **Table 1** shows the elemental and proximate

analyses of the feedstock. The bed material – washed sand with a particle size of approximately 150 μm and a density of 1.60 g/cm^3 at 20°C – was obtained from Alfa Aesar.

Table 1. Proximate and elemental analysis of biomass

	Elemental analysis (wt. %)			
Material	Carbon	Hydrogen	Nitrogen	Oxygen ^a
Beech wood	46.70	5.57	<0.01	47.72
Biochar	85.76	2.59	<0.00	11.65
	Proximate Analysis (wt. %) ^b			
Material	Humidity	Volatile matter	Fixed Carbon	Ash
Beech wood	7.44	74.19	17.52	0.85

^a Obtained by difference.

^b Based on TGA experiments according to the method established by (García et al., 2013).

2.2 Biochar Preparation

The used biomass to prepare biochar was beech wood. The biomass was introduced in the fluidized bed reactor and was heated at temperature of 900°C at 3°C/min and kept for 1 h (in order to ensure that no volatile matter was present after devolatilization). A nitrogen flowrate of 0.5 l/min as gas carrier was used. The biochar was recovered from the reactor and then sieved to a particle size of approximately 450 μm , with a bulk density of 0.33 g/cm^3 at 20°C.

2.3 Experimental set up

The gasification runs were done in a semi-continuous fluidized bed reactor (**Figure. 1**). The reactor and its oven were obtained from MTI corporation (Ref. OTF-1200X-S-FB). The reactor material was stainless steel, with an inner diameter of 22 mm and an external diameter of 25 mm. Gasification agent CO_2 and carrier gas N_2 were then fed from the bottom of the reactor. Steam was fed into the reactor through an automatic syringe driver (Ref. AP14 ASCOR). The gasification process was isothermal. The bed materials were introduced in the reactor and then heated at the desired operation temperature. A constant flow of N_2 was used to keep an inert atmosphere, when the desired temperature was reached the gasification agent was added. The Biomass was fed from the top of the reactor to the centre of the bed through a stainless-steel tube. Gaseous products exited from the top of the reactor

through a separate tube. Two condensers and a flask were finally placed in a cold bath at -10°C to collect all liquid products. Non-condensable gases passed through a cotton filter, in order to retain possible solid particles. Gaseous species were analysed continuously using micro-gas chromatograph, from Chemlys corporation (Ref. PN 074-594-P1E).

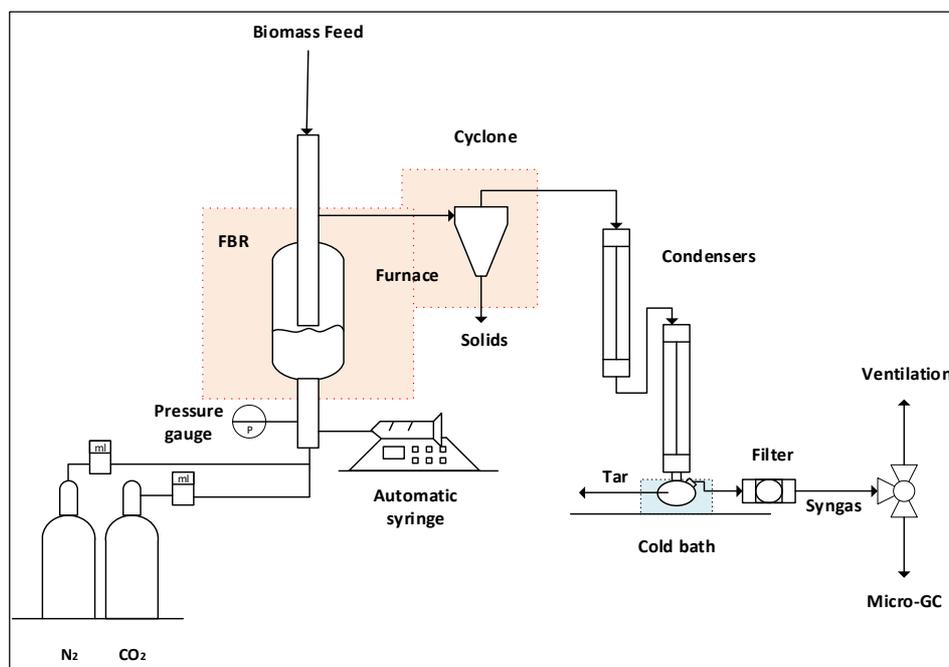


Figure 1. Fluidized bed gasifier setup.

The gasification experiments were conducted at temperatures ranging from 600 to 900 $^{\circ}\text{C}$. The biomass feed rate was 1 g/min. The washed sand or biochar as bed materials were placed inside the reactor before each run, with a 40 mm height. The partial pressure of gases was $P_{\text{N}_2} = 0.05$ atm and 0.95 atm for the gasification agent (CO_2 or H_2O). N_2 was used as an internal standard for gas flowrate calculations. The total flowrate of gasification agent entering the gasifier was 1.15 l/min. For product collection, solid particles were obtained after each experiment from the reactor and the cyclone, while liquid products, such as tar and water, were collected from condensers and the flask using an organic solvent (acetone, purity 99.98 %) and then analysed using gas chromatography. For tar molecules identification a chromatograph-mass spectrometer (GC-MS, Varian 3900 -Saturn 2100T with column VF-1701 ms (Agilent) (60 m \times 0.25 mm \times 0.25 μm film thickness) was used. For tar molecules quantification it was used a GC-Flame Ionization detector (GC-FID Scion 456-GC Bruker

instrument). Water content was obtained using Karl Fisher titration, Tar was classified in substances groups (**Table A.1 Annexe**).

2.4 Energy Balance

The energy balance of the system was done by isolating the gasifier from other units (such as cyclone, condensers, etc) and considering only its energy input and output streams (**Figure. 2**). Following first law of thermodynamics, energy is conserved. Applying an energy balance to the gasification system, as shown in **Figure 2**, it turns into:

$$\sum \dot{E}n_{in} = \sum \dot{E}n_{out} \quad Eqn. (1)$$

$$\dot{E}n_{biomass} + \dot{E}n_{agent} + \dot{Q}_{Heat} = \dot{E}n_{syngas} + \dot{E}n_{Fluegas} + \dot{E}n_{Tar} + \dot{E}n_{biochar} \quad Eqn. (2)$$

For this study, the heat loss through the walls of reactor was neglected, then $\dot{Q}_{loss} = 0$.

Where $\dot{E}n_{biomass}$, $\dot{E}n_{agent}$, $\dot{E}n_{syngas}$, $\dot{E}n_{Fluegas}$, $\dot{E}n_{Tar}$ and $\dot{E}n_{biochar}$ were the energy rates of biomass, gasification agent, syngas, flue gas, tar and biochar, respectively, and \dot{Q}_{Heat} was the specific additional heat input introduced to the system to perform gasification at specified temperature. Since no heat loss was taken into account, then $\dot{Q}_{Heat} = \dot{Q}_{gasification}$

The energy rate of a stream can be calculated as follows:

$$\dot{E}n = \dot{E}n_{ph} + \dot{E}n_{ch} + \dot{E}n_{po} + \dot{E}n_{ki} \quad Eqn. (3)$$

Where the subscripts *ph*, *ch*, *po* and *ki* were the physical, chemical, potential and kinetic energy rates, respectively. Potential and kinetic energies of streams were considered to be very small when compared to physical and chemical (Zhang et al., 2015), and were hence neglected. Therefore, *Eqn. (3)* is expressed as:

$$\dot{E}n = \dot{E}n_{ph} + \dot{E}n_{ch} \quad Eqn. (4)$$

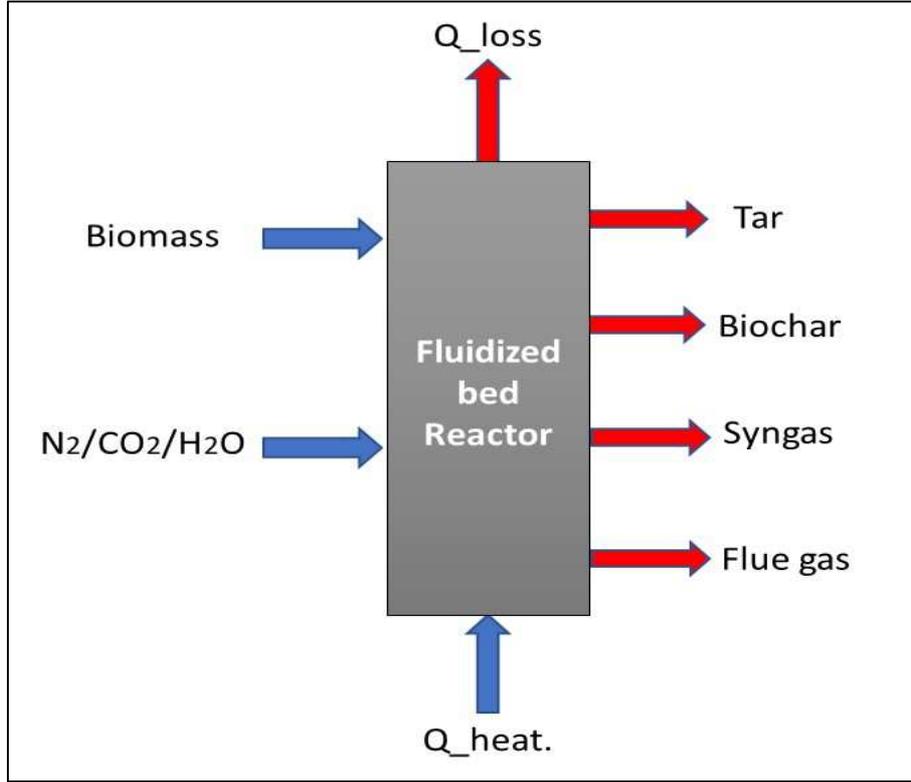


Figure 2. Energy input and output streams for gasifier.

Substituting each energy term by its definition, it is found that physical energy (sensible heat) was defined as follows,

$$\dot{E}_{n_{ph}} = n_i \int C_{p_i} dT \quad \text{Eqn. (5)}$$

Meanwhile the chemical energy (enthalpy of formation) was defined as follows,

$$\dot{E}_n = \sum_i n_i h_{f,i}^\circ \quad \text{Eqn. (6)}$$

Substituting Eqn. (5) and Eqn. (6) in Eqn. (4), it turned into

$$\dot{E}_n = n_i \left(\int C_{p_i} dT + \sum_i h_{f,i}^\circ \right) \quad \text{Eqn. (7)}$$

For solids

For non-conventional fuel (e.g. biochar and biomass), the enthalpy of formation was calculated based on their combustion reaction (Atsonios et al., 2015):

$$h_{f, fuel}^\circ = \alpha h_{f, CO_2}^\circ + \beta h_{f, H_2O}^\circ + LHV_{fuel} \quad \text{Eqn. (8)}$$

Where α , β , h_{f,CO_2}° , and h_{f,H_2O}° were the stoichiometric coefficient and enthalpies of formation of CO_2 and H_2O , respectively. LHV was the lower heating value of a compound at $15^\circ C$.

The above *Eqn. (8)* was believed to provide the chemical energy for non-conventional fuels. The physical energy of biomass was now difficult to calculate because of the occurrence of the devolatilization reaction. Due to this fact, researchers only used the chemical energy; otherwise, they calculate the physical energy at a temperature just before devolatilization reaction (Hosseini et al., 2012; Richard and Thunman, 2003). In this study, the energy equation used for biomass and biochar was:

$$\dot{E}_{fuel} = \dot{n}_{fuel} * (LHV_{fuel} + \Delta h_{sensible}) = \dot{n}_{fuel} * (LHV_{fuel} + \int Cp_{fuel} dT) \quad Eqn. (9)$$

In the case of biomass, T was the devolatilization temperature, while for biochar, T was the operating temperature. The LHV of the fuels was calculated using the Dulong formula.

$$LHV_{fuel} = (33.80 x_C + 144.20 x_H - 18.03 x_O) \quad Eqn. (10)$$

Where, x_C , x_H and x_O were the carbon, hydrogen and oxygen composition (wt. %), obtained for each fuel from the elemental analysis.

For gases

For gaseous streams (e.g. syngas, flue gas and gasification agents), the energy rate was described as follows:

$$\dot{E}_{gaseous\ streams} = \sum_i n_i (\int Cp_i dT + LHV_i) \quad Eqn. (11)$$

In *Eqn. (11)*, n_i , Cp_i , and LHV_i were the molar flow rate, heat capacity at constant pressure and LHV of gases, respectively.

For tar

In order to calculate the tar energy rate, only the major compounds of each substance group were taken into consideration. Due to the fact that number of tar molecules was very elevated (>50 compounds), it

was necessary to choose the major compound in terms of quantity for each substance group. The latest was done to simplify calculation. It was also taken into consideration the enthalpy for phase change of compounds. The energy equation for tar was described as follows:

$$\dot{E}n_{Tar} = \sum_i n_i (\int C_{p_i} dT + \Delta h_{phase_change} + LHV_i) \quad Eqn. (12)$$

Coefficients of heat capacity along with other thermodynamic properties for tar and gaseous species were shown in **Table A2-A3 Annexe**.

2.5 Energetic efficiency

To evaluate the efficiency of the gasification system, cold gas efficiency (CGE) was frequently used as an important parameter. It was defined as:

$$CGE = (m_{syngas} LHV_{syngas}) / (m_{biomass} LHV_{biomass}) \quad Eqn. (13)$$

The values of mass and LHV for syngas and biomass were in dry basis.

3. Results and discussion

3.1 Products distribution.

The results obtained from the gasification of beech wood using different gasification agents and bed materials were reported in the **Table A4 Annexe**. For the first part studied, where CO₂ was used as gasification agent and sand as bed material, results were similar to those obtained by (Sadhvani et al., 2016) at similar conditions. The residence time of gases was calculated from the flow rate and kept constant for each experiment, (15.20 s). The fluidization regime was identical to Sadhwani study (Sadhvani et al., 2016). The observed products trend was the same, but the values varied due to the fact that the biomass feed rate and CO₂ partial pressure were different from this study.

For the case of liquids, tar amounts corresponded with values found elsewhere in the literature for high temperatures, in the range of 1-15 g/Nm³ of syngas produced (Milne et al., 1998) for fluidized bed reactors. In the cases where biochar was used as bed material, upgrading of syngas quality was observed. This was attributed to the catalytic effect of biochar (Abu El-Rub et al., 2004) over tar compounds due to its mineral composition (e.g. K, Na, Mg).

3.2 Energy balances

In the following section, the results obtained for energy consumption and production for the different performed gasification tests were detailed. The results presented a total error of 1.70%, due to experimental uncertainty. The errors surrounding the experimental test were those found after deviation calculations after experiment repetitions, mass weighting, values rounding and equipment tolerance. The uncertainty of values was added with error bars over the presented results. The errors for each parameter was described as follows: The majority of the variation of results came from experiments repetitions ($\pm 1.47\%$) and $\pm 0.23\%$ came from values rounding (two significant numbers) and equipment tolerance such as: balance tolerance (0.01 grams), furnace tolerance (1°C) and Gas-Chromatography tolerance (0.001%). More information about the uncertainty analysis can be found in the **Annexes**.

In addition, the energy values were presented in dry basis.

3.2.1 Effect of gasification temperature.

The energy distribution values of various reaction temperatures are shown in **Figure 3**. Sand was used as a bed material in this case. Only the output products of energy distribution were shown. For inputs, including the biomass and the gasification agent, the sum of energy values varied from 16.7 to 17.2 MJ/kg_{biomass} at 600 to 900 °C when temperature increased from 600 to 900 °C. As biomass quantity and CO₂ flow rate were kept constant, these variations were attributed to the change of sensible heat from the gasification agent. The total energy of the products increased as temperatures increased because of biochar conversion and tar cracking reactions.

Biochar

For unconverted biochar, the energy trend decreased as temperatures increased. Due to the presence of gasification agent and the incrementation of temperature, boudouard reaction and biochar conversion were clearly favoured. Thus, biochar energy was in other words transferred to syngas. At 600 °C and 700 °C, biochar still represented 48 % and 44 % (9.55 and 8.25 MJ/kg_{biomass}) of the output energy of

the system, respectively. This observation showed that less biochar was converted at low temperatures. At 800 °C and 900 °C, temperatures that in gasification terms were considered high.

Table 2. Results of Energy balance and product composition of gasification tests^a.

Test	Temperature °C	Bed material	Syngas Components: MJ/kg _{biomass}					^a Tar: MJ/kg _{biomass}								Energy streams: MJ/kg _{biomass}					
			H ₂	CO	CH ₄	C ₂₊	C ₃₊	(a)	(b)	(c)	(d)	(e)	(f)	(g)	(h)	Syngas	biochar	Tar	Fluegas	^b Agent	
Gasification with CO ₂	600	Sand	0.11	1.44	1.01	0.12	0.02	2.73	0.59	0.39	0.80	0.53	0.48	0.31	0.03	2.70	9.55	5.85	1.59	1.16	
	700		0.45	1.70	2.56	0.17	0.19	1.09	0.56	0.23	0.57	0.29	0.21	0.20	0.02	5.07	8.25	3.18	2.07	1.33	
	800		1.07	5.12	3.93	0.27	0.38	0.08	0.88	0.09	0.54	0.20	0.39	0.40	0.08	10.77	6.37	2.67	2.40	1.51	
	900		2.29	6.13	4.55	0.51	0.01	0.04	0.30	0.09	0.34	0.12	0.29	0.49	0.11	13.50	5.69	1.78	2.38	1.69	
Pyrolysi s	800		0.59	2.44	1.89	1.28	0.81	0.16	1.55	0.35	1.01	0.32	0.70	0.64	0.13	7.01	6.54	4.86	1.51	1.06	
	900		1.35	2.82	2.18	1.72	0.63	0.06	0.70	0.29	0.99	0.17	0.53	0.98	0.18	8.71	6.23	3.91	1.67	1.17	
Gasification with CO ₂	600		Biochar	0.07	1.80	0.74	0.06	0.87	2.52	0.36	0.26	0.59	0.71	0.39	0.19	0.02	3.53	9.08	5.05	1.98	1.67
	700			0.38	6.58	1.83	0.23	0.95	0.83	0.32	0.10	0.26	0.17	0.11	0.10	0.01	9.97	7.12	1.90	2.14	1.91
	800	0.76		8.30	2.50	0.31	0.67	0.14	0.65	0.11	0.32	0.11	0.32	0.26	0.00	12.53	4.85	1.90	2.45	2.17	
	900	1.98		11.27	3.46	0.22	0.25	0.04	0.14	0.04	0.17	0.03	0.09	0.15	0.03	17.18	1.80	0.69	2.93	2.42	
Steam gasification	600	0.49		1.64	1.40	0.02	0.06	2.25	0.66	0.18	0.88	0.59	0.39	0.22	0.03	3.60	7.47	5.20	0.74	0.45	
	700	1.33		3.61	3.10	0.08	0.13	0.39	0.92	0.27	0.83	0.58	0.55	0.40	0.17	8.25	5.40	4.11	0.86	0.58	
	800	2.66		4.07	4.41	2.15	0.61	0.11	0.82	0.08	0.34	0.10	0.37	0.27	0.02	13.90	4.34	2.11	0.99	0.59	
	900	3.00		4.78	5.33	1.90	1.54	0.13	0.28	0.05	0.37	0.05	0.43	0.53	0.00	16.55	2.79	1.82	1.26	0.65	

^aValues included uncertainty of $\pm 1.7\%$.

^ba) Others aliphatic compounds, b) Phenols, c) Furans, d) Heterocyclic aromatic compounds, e) Aromatic compounds, f) Light poly-aromatic hydrocarbons, g) Naphthalenes, h) Heavy poly-aromatic hydrocarbons.

^cGasification agent.

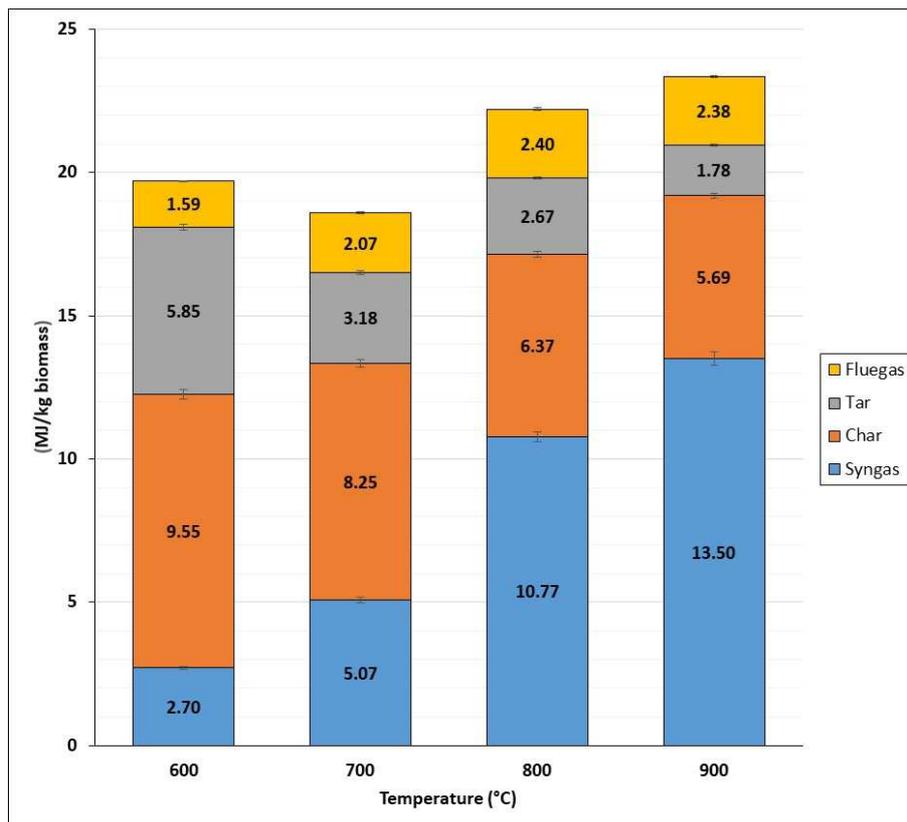


Figure 3. Effect of temperature on energy products distribution; Agent: CO₂, Bed material: Sand.

The biochar energy percentage changed to 29 % and 24 % (6.37 and 5.69 MJ/kg_{biomass}), respectively, as syngas represented the highest percentages: 48 % and 58 % (10.77 and 13.50 MJ/kg_{biomass}) of the total produced energy, respectively. For the highest temperature 900°C, biochar energy value was 5.7 MJ/kg_{biomass}. This reduction in biochar energy can also be explained with the fact that biochar conversion and reactivity increased with temperature. These biochar energy values still represented a high amount of energy to be conceded to the syngas, if better biochar conversion was achieved.

Tar

As aforementioned for biochar energy, the tar energy was reduced as temperatures increased. At 600 °C, the energy available from tar represented 30 % (5.85 MJ/kg_{biomass}) of the total energy of the products, while at 900 °C, it represented only 8 % of the total distribution (1.78 MJ/kg_{biomass}). This reduction in tar energy can be explained by the fact of thermal cracking reactions. Tar energy was transferred to the syngas stream as new gas molecules were formed.

Gas

As can be seen in the previous graphic (**Figure 3**), syngas energy increased with temperature: 13.5 MJ/kg_{biomass} at 900 °C were obtained, proving the high energetic value of syngas produced from gasification. As temperature favoured biochar conversion and tar cracking, the syngas energy increased directly. Due to the fact that biochar conversion led to syngas formation and tar cracking led to smaller molecules formation, which were included syngas. A similar tendency was observed for the flue gas exiting the system, noting that in this study, the water produced, N₂ and CO₂ were considered as “flue gas”. For each experiment at different temperatures, CO₂ was also produced through the devolatilization process. Hence, flue gas molar flow rates increased lightly as temperatures went up.

Figure 4a shows the energy distribution for the principal compounds in the syngas, as a function of temperature. As can be seen, CO and CH₄ were predominant in energetic terms. H₂ increased significantly as temperature increased, but lower values than the ones for CH₄ were obtained due to the higher energy density values of CH₄. At high temperatures, CO represented the highest values because of high molar flow rates in the syngas. Other gas species, such as C₂₊ and C₃₊ were increased as temperatures were higher, except at 900 °C, where C₃₊ amount decreased because of its decomposition to smaller molecules.

Heat input

The specific heat input needed to perform gasification was calculated without taking into consideration the energy of the gasification agent stream. The latest was done; in order to normalize the obtained results and compare them with the literature (Ephraim, 2016; Renganathan et al., 2012; Wan et al., 2013). As the CO₂/C molar ratio significantly influences the specific heat of gasification (Sadhvani et al., 2016), it was recommended to normalize the energy input, before comparing with other results. Now, the higher the molar flow rate of the gasification agent, the higher its sensible heat – and vice versa. This occurrence was frequently taken into consideration for these calculations. **Figure 4b** shows the input heat of gasification as a function of the reaction temperature, compared with the amount of energy obtained from the syngas. Globally, from 600 to 900 °C, the heat of gasification increased from

3.7 to 7.3 MJ/kg_{biomass}. Between 600 and 700 °C, there was a decrease of the input heat for gasification. This phenomenon was also observed by Renganathan, (Renganathan et al., 2012) which argued that a minimum in the curve of heat input vs. temperature using pure CO₂ as a gasification agent for various carbonaceous feedstock occurred. For different CO₂/C ratios vs. temperature, a minimum of heat input can be found at temperatures between 600 and 800°C. The author explained that for a given condition of feedstock and gasifying agent, the heat input required tendency could vary because of the used flowrate of CO₂ and the temperature region. In the low temperature region, with the increase in temperature the quantity of CO₂ required decreases radically reducing the heat input needed steeply. An incrementation in exothermic reactions could have been attended, providing sustainably to the gasification reaction and reducing the endothermicity as less heat input was required.

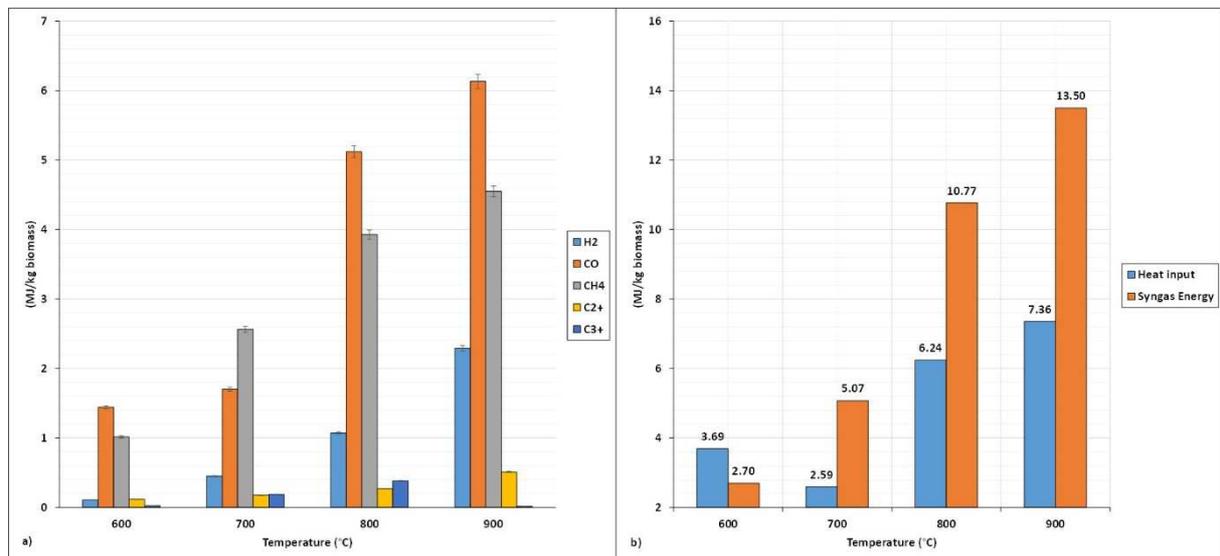


Figure 4. a) Gas energy distribution b) Heat input for gasification at different temperatures; Agent: CO₂, Bed Material: Sand.

3.2.2 Effect of varying gasification agent.

In this section, high-temperature pyrolysis was compared to gasification under CO₂, both with sand as the bed material at 800 and 900 °C. The products energy distribution can be observed in **Figure 5**. For pyrolysis, the total energy of the products increased very slightly from 800°C to 900 °C: 19.9 to 20.5 MJ/kg_{biomass}. For gasification case from 800 °C to 900 °C the total energy varied from 22.2 to 23.3

MJ/kg_{biomass}. It was evidenced that gasification provided higher energy values of products than pyrolysis. The impact on energy balance of gasification agent was clearly observed when CO₂ is used in gasification. Despite having been carried out under the same operating conditions, the pyrolysis could not achieve as efficient conversion of biochar and tar energy into syngas as gasification.

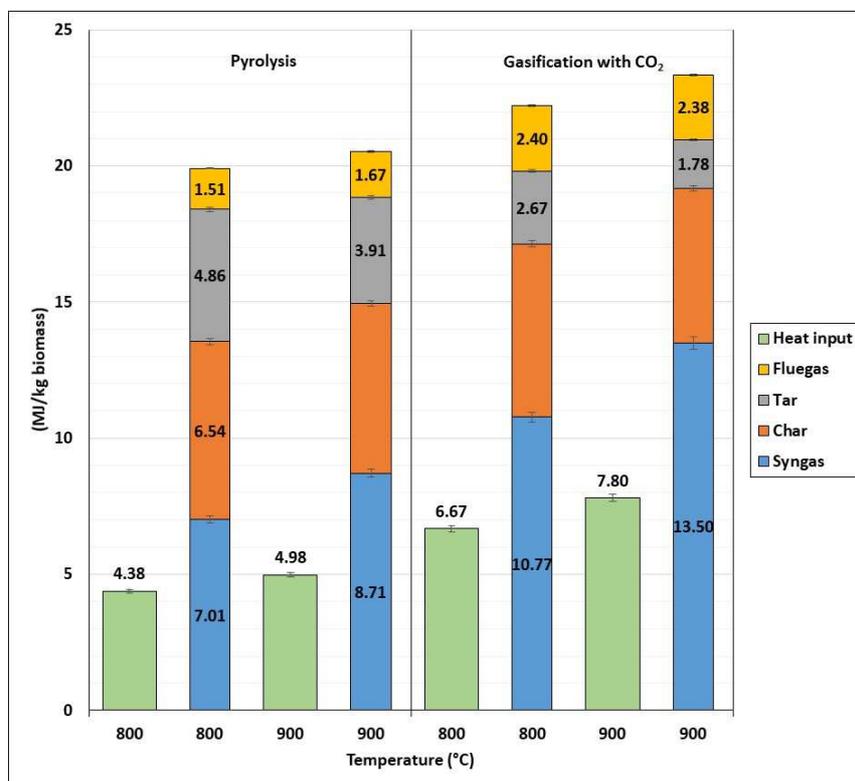


Figure. 5 Energy products distribution and heat input for pyrolysis and gasification; Agent CO₂/N₂, Bed material: Sand.

Biochar

As the conversion of biochar was influenced by the presence of the gasification agent, higher energy values of biochar for pyrolysis were observed. This was due to its inferior conversion in pyrolysis than gasification. At 800°C and 900°C, values were 6.54 and 6.22 MJ/kg_{biomass}, for the pyrolysis case and 6.37 and 5.69 MJ/kg_{biomass} for gasification, for the respective temperatures. Generally, as temperature increased, the energy value of the output biochar stream decreased. In pyrolysis for a reduction in the mass balance of biochar yield of 1.1% from 800°C to 900°C, its energy was reduced by 0.32 MJ/kg_{biomass}. Comparing the latest value with gasification which yield reduction was 2.2% for the

respective temperatures, biochar energy was reduced by 0.68 MJ/kg_{biomass}. This evidence the high energetic value that represented non-conventional solids fuels.

Tar

As it was observed for biochar, tar energy values decreased with temperature increase. This statement was clearly influenced by tar thermal cracking reactions. For pyrolysis, tar energy decreased from 4.9 to 3.9 MJ/kg_{biomass}, and from 2.7 to 1.8 MJ/kg_{biomass} for gasification, for the respective temperatures of 800°C and 900°C. for the respective temperatures of 800°C and 900°C. The values of tar energy for pyrolysis represented between 19.4 to 24.4% of the total energy distribution of the products. The latest values were considered a negative point for thermal conversion process, which syngas was yearned as the principal energy stream. On the other hand, in gasification tar only represented 12.0 to 7.6% of the total energy of products. Globally less tar energy was obtained from CO₂ gasification than pyrolysis, due to the fact that gasification agent was present, favouring dry-reforming of tar molecules (Caprariis et al., 2014; Kaisalo, 2017).

Gas

For the syngas, the results were more significant. In pyrolysis, it was observed at 800 °C that 7.0 MJ/kg_{biomass} were contributed by the syngas produced, while at 900 °C, 8.7 MJ/kg_{biomass} were contributed. In the case of gasification with CO₂, 10.7 to 13.5 MJ/kg_{biomass} were noted for 800 and 900°C respectively. For power generation purposes, the syngas obtained from gasification with CO₂ gives higher energetic values than that obtained from pyrolysis. Globally, both temperatures for pyrolysis showed lower energetic values when compared with gasification which can be explained by the fact that the use of CO₂ as agent, favoured tar and biochar conversion which consequently increased the syngas energy value.

The energy distribution of the gas obtained from pyrolysis was represented in **Figure 6a**. As it was the case for gasification, CO and CH₄ represented the higher energetic values for the gaseous components. Both energetic values increased as temperature increased. The energetic values of the C₂+ and C₃+ gases were very remarkable for the case of pyrolysis, where the values were all higher than the H₂

values, except for C₃₊ at 900 °C. In comparison with gasification, only C₂₊ and C₃₊ values are higher for pyrolysis. All the other gases displayed higher values for gasification. The fact that C₂₊ and C₃₊ values were higher for pyrolysis showed that gasification was able to crack heavier molecules into lighter compounds.

The energy content of CO represented the major energy difference between both systems of syngas, as knowing that boudouard reaction might increase the CO formation.

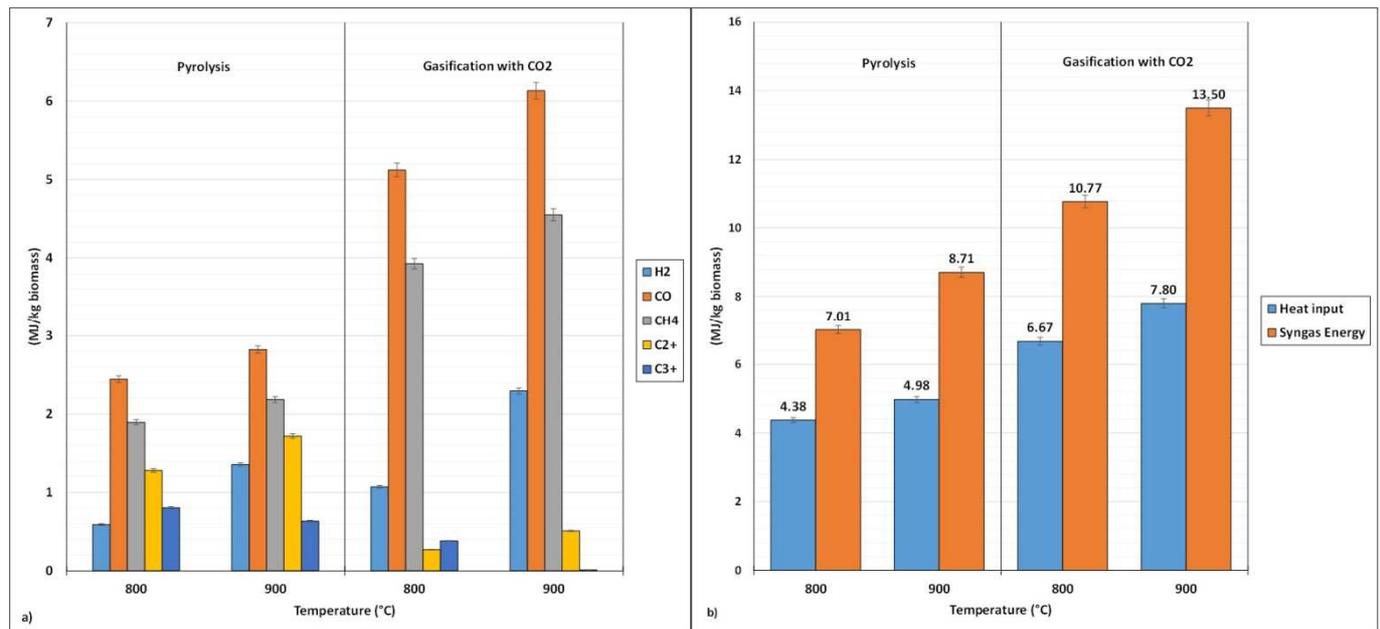


Figure. 6 Comparison between pyrolysis and gasification; Agent CO₂/N₂, bed material: sand a) energy distribution for gases and b) heat input Vs Syngas Energy.

Heat input

Figure 6b showed the values of the heat input for both set-ups compared with the amount of energy obtained from the syngas. The heat input needed for pyrolysis is lower than that needed for gasification for both temperatures. This was due to the fact that biochar conversion reactions are highly endothermic, and a higher amount of biochar was converted with using CO₂. The input heat for pyrolysis increased when temperature increased from 4.4 to 4.9 MJ/kg_{biomass}, as it did for gasification, but with higher impact, from 6.7 to 7.8 MJ/kg_{biomass}. (Atsonios et al., 2015) reported that these values could be ±15.5 % different from one process to another, due to calculation uncertainty while

calculating heating values for solid fuels. These results showed that energetically pyrolysis was more sustainable than gasification in terms of the required heat input, this statement considered that exothermic and endothermic reactions took place for both processes. Considering that gasification was an intermediate between pyrolysis and combustion as it was discussed before, this accounted energy required for pyrolysis plus energy required for biochar conversion. The latest was considered highly endothermic.

The difference in heat input between pyrolysis and gasification was about 2.3 MJ/kg_{biomass} at 800°C and 2.8 MJ/kg_{biomass} at 900°C. As for gasification at 900°C, only 2.8 MJ/kg_{biomass} of heat input was needed to obtain a difference of 4.8 MJ/kg_{biomass} in syngas energy at 900 °C against pyrolysis. It can be deduced that gasification was indeed a better option than pyrolysis in energetic terms.

3.2.3 Effect of biochar as bed material on energy balance.

Biochar was frequently used as a bed material, in order to catalytically crack the undesirable products of gasification. In this section, the energy balance of two gasification set-ups were analysed using CO₂ and steam as gasifying agents for a temperature range of 600-900 °C. The results obtained when CO₂ and steam were used were detailed in **Figure 7**. It has to be noted that for all calculations of heat input, the energy of the gasification agent was not taken into consideration, in order to normalize both results and to be able to compare them. The energy from the biomass (15.5 MJ/kg_{biomass}) was constant for all experiments. Comparing both tests, it may be observed that by using CO₂, more energy was available in the products than when using steam. At high temperatures, the energy difference was small because of similar energy values of the product streams. As the inlet energy was kept constant for both tests (only biomass), using *Eqn. (1)*, a higher energy input was required for CO₂ gasification.

Tar

The tar produced from gasification with CO₂ had lower energetic value than produced with steam gasification. As less energy of tar was obtained, it can be said that dry reforming of tar (use of CO₂) was more effective than steam reforming of tar, when biochar was used as catalytic bed in energetic terms. It had to be also added that the presence of gasification agents could eventually favour the

catalytic activity of biochar. If the latest was the case, CO₂ activation of biochar was more efficient than steam. For both set-ups, the tar energy values decreased as temperature increased. At high temperature where the lower values of tar were observed, for steam gasification most of the energy of tar came from light poly-aromatics hydrocarbons and naphthalene compounds. In the case of CO₂ gasification, the tar energy value of 0.7 MJ/kg_{biomass} was divided on naphthalene, heterocyclic and phenolic compounds representing 67% (0.47 MJ/kg_{biomass}) of the total tar energy.

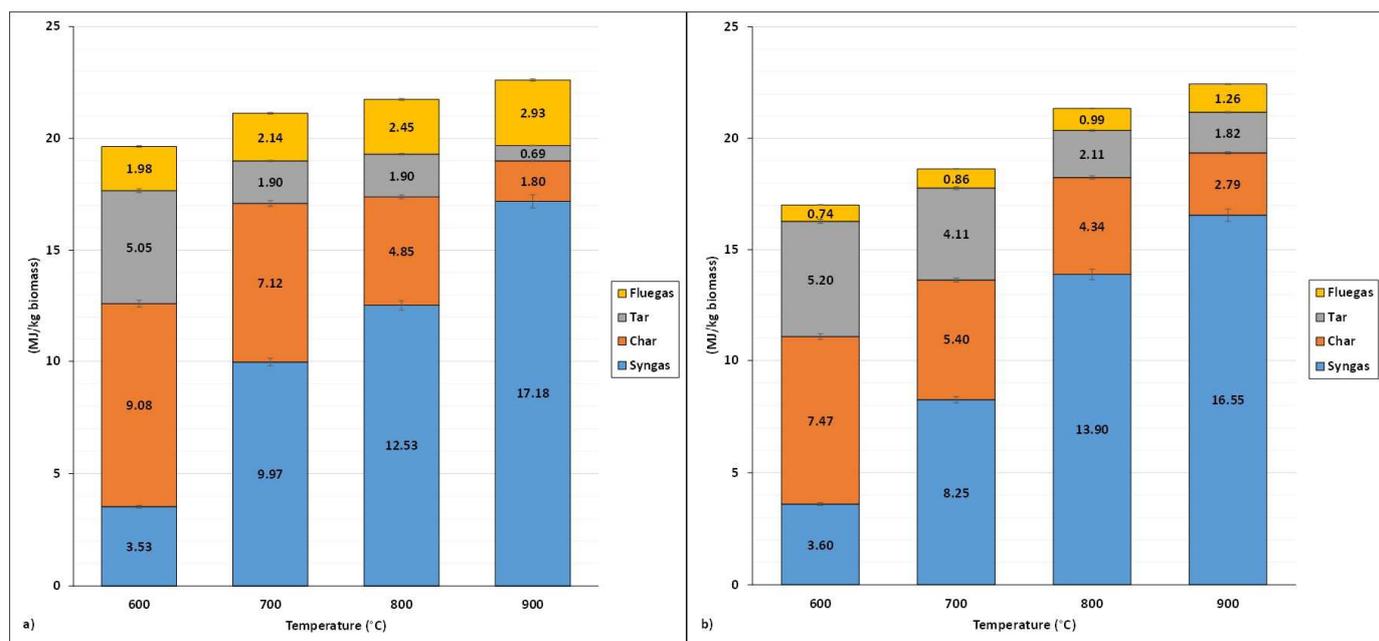


Figure. 7 Products energy distribution for gasification; Agent: CO₂/H₂O, Bed material: biochar a) CO₂ and b) steam.

Biochar

At 600 °C, this energy difference between both set-ups came mainly from the unconverted biochar, which represented around 9.1 MJ/kg_{biomass} for CO₂ gasification and 7.5 MJ/kg_{biomass} for steam gasification. As temperature increased, steam gasification showed lower energy values of biochar, due to higher conversion that was achieved using this agent. Only at 900°C the conversion was lower for steam than for CO₂ gasification, by the fact less biochar was converted with steam. Therefore, at temperatures between 600 and 800°C biochar energy with steam was 2.6 times higher, equivalent to 1.8 MJ/kg_{biomass}.

The reactivity of biochar with steam was known to be faster than with CO₂, this could explain the fact that lower conversion was achieved with CO₂ compared to steam gasification (Guizani et al., 2016; Morin et al., 2018). The reactivity of biochar depends on many factors such as, temperature, porosity, presence of inhibitors, heating rate and others. Temperature increased reactivity for both steam and CO₂ gasification, meanwhile as temperature increases the difference in reactivity becomes smaller for both gasification agents. At 900°C, there was a strong production of H₂ for steam gasification as hydrogen was known to be an inhibitor of biochar steam reforming gasification, it could be one of the reasons of lower conversion at 900°C compared to CO₂. Tar and hydrogen both evoke inhibition of steam gasification of char (Fushimi et al., 2011).

Gas

Significant energy values were also obtained for the flue gas (CO₂, H₂O and N₂); at 600 °C, the results were 2.1 and 0.7 MJ/kg_{biomass} for CO₂ and steam gasification, respectively. For all temperatures, the quantity of flue gas was higher when CO₂ was used as the gasification agent. Concerning syngas, steam gasification generally gave rise to higher energetic values. The exception was at 900 °C, where CO₂ produced syngas with a higher energetic value: 17.2 MJ/kg_{biomass}, opposed to steam with 16.5 MJ/kg_{biomass}.

The energy distribution of the gases in the syngas were shown in **Figure 8**. The syngas energy for gasification with CO₂ was mainly distributed in gas CO, which represented between 50 and 66 % (1.80 and 11.27 MJ/kg_{biomass}) of the total energy of the syngas. For steam gasification, it represented between 28 and 45 % (4.78 and 1.40 MJ/kg_{biomass}) and decreased as temperature increased. For steam gasification, gas product distribution was more variable; H₂ varied from 13.5 to 19.1 % (0.49 to 3.00 MJ/kg_{biomass}) and CH₄ from 32 to 39 % (5.33 to 1.40 MJ/kg_{biomass}) of the total energy value. For gasification with CO₂, the H₂ and CH₄ varied from 2.0 to 11.5 % (0.07-1.98 MJ/kg_{biomass}) and 18.3 to 20.1 % (1.83-0.74 MJ/kg_{biomass}) of the total energy of the syngas, respectively. Comparing both gases distributions, CO₂ gasification provided a mono-energetic product, in which the majority of the energy came from a single compound, CO. In the case of steam, a poly-energetic product was observed – where no one component contained the majority of the energy – was obtained.

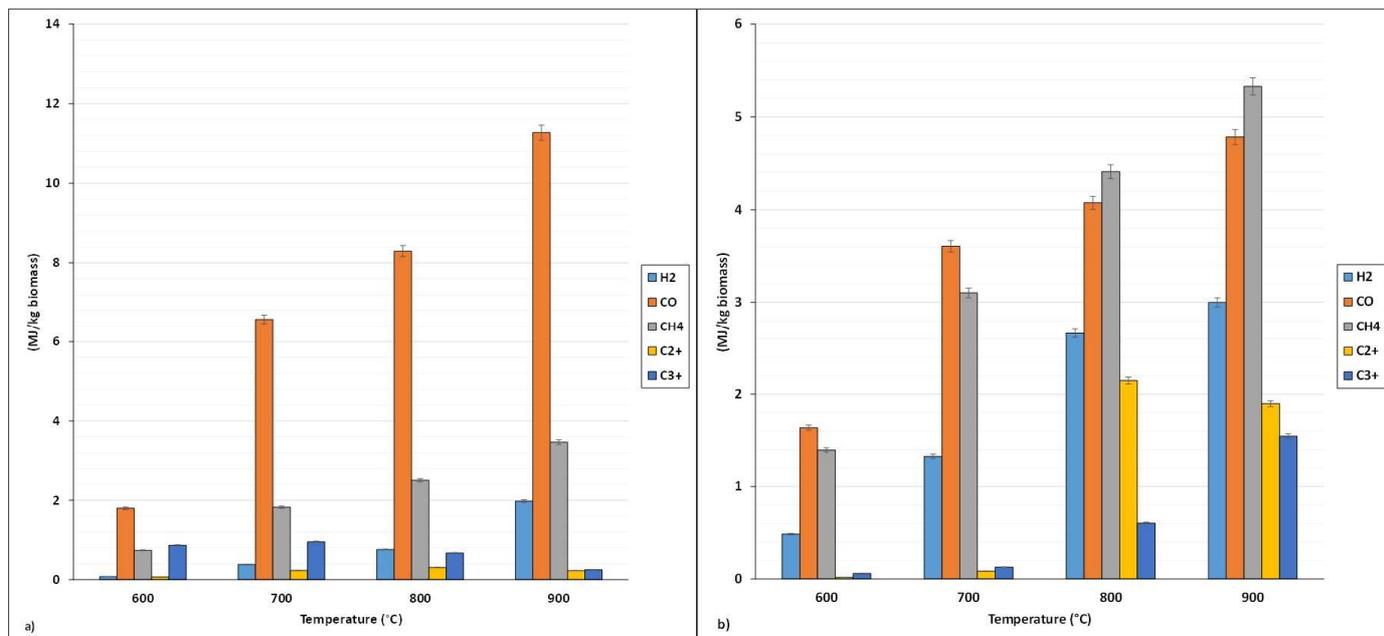


Figure. 8 Gas energy distribution; Agent: CO₂/H₂O, Bed material: biochar a) CO₂ b) steam gasification.

Heat input

Figure 9 showed the heat inputs for each setup. It can be seen that globally, the heat input needed for gasification was higher when CO₂ was used as gasification agent. For both cases, as temperature increased, the heat input also increased. At low temperatures, the difference was larger between each setup. At 600 °C, the difference was about 2.6 MJ/kg_{biomass} while at 700 °C, it was 2.5 MJ/kg_{biomass}. At 800 °C and 900 °C, the gap was closer for both setups: 0.4 and 0.17 MJ/kg_{biomass}, respectively.

These previous results demonstrate that for catalytic cracking of tars at high temperatures specially 900 °C with biochar as the bed material and CO₂ as the gasification agent, less tar would exit with the syngas. The latter would have a higher energetic value than the syngas produced with steam as the gasification agent. The additional heat input difference at these conditions would only be around 0.17 MJ/kg_{biomass}, as well. Therefore, it was only at low temperatures, especially 600-700 °C, that steam gasification is energetically more favourable.

These difference in heat input could be explained by the fact that more exothermic reactions took place when steam was used as gasification agent. This statement was verified as there was a significant increase of methane formation and hydrogen. Both reactions methane formation and water gas shift

reaction were known to be highly exothermic, consequently for this reason a compensation of energy was provided from these reactions.

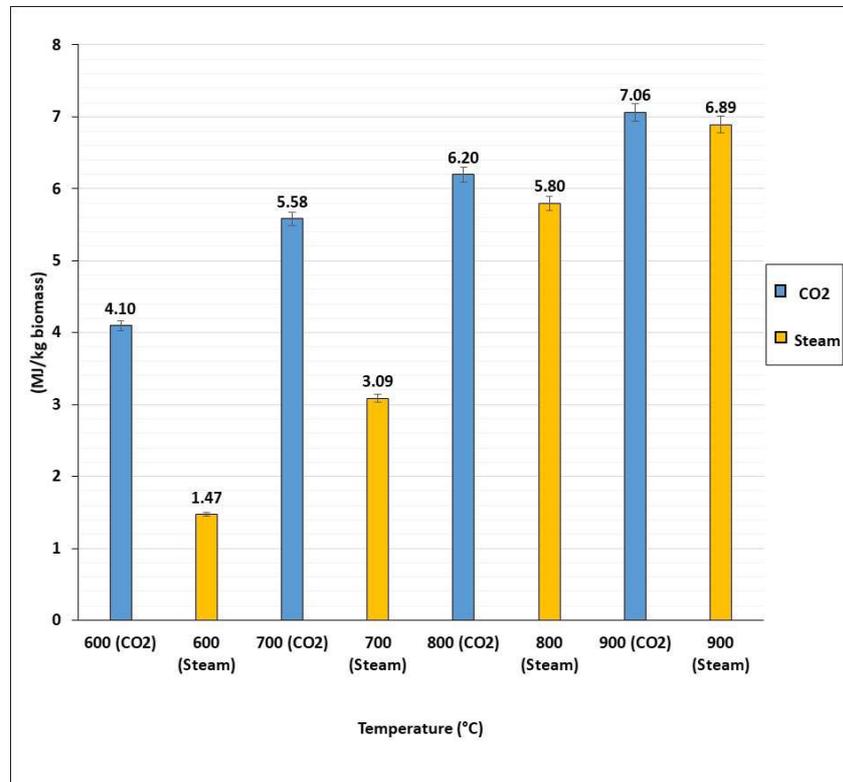


Figure. 9 Heat input for gasification: Agent: CO₂/H₂O, Bed material: biochar.

3.3 Energy efficiency

The results obtained for cold gas efficiency (CGE) Eqn. (13) for each studied configuration were shown in **Table 3**. CGE was calculated taking as energy inlet only the biomass stream (Chaiwatanodom et al., 2014). This consideration showed the efficacy of the syngas as a function of the supplied biomass energy. As it was illustrated, CGE increased as temperature increases for all experiments, and similar trends have also been obtained in literature for steam and CO₂ gasification (Buentello-Montoya and Zhang, 2019; Parvez et al., 2016; Renganathan et al., 2012; Sreejith et al., 2013). For CO₂ gasification, using sand as a bed material, the highest values of CGE, 0.63 and 0.78, corresponded to 800 and 900 °C, respectively. The CGE values obtained for pyrolysis were lower than those obtained for gasification when sand was used as the bed material. This means that when

gasification was performed, the energetic contribution of the syngas provided better energy yields than pyrolysis.

In the cases where biochar was used as the bed material, in order to catalytically crack tar, the CGE values were quite similar for both configurations (CO₂ and steam gasifications). At 600 °C, the values were superposed at 0.21. At a temperature of 700 °C, CO₂ gasification was superior, with 0.59, and at 800 °C, steam gasification was superior, with 0.82. For the highest temperature, 900 °C, both values were close, although CO₂ presented a higher result of 0.99 and steam 0.97. The CGE values closest to unity did not mean that all the energy available in biomass was transformed into syngas. These values were obtained due to the fact that heat input was not considered in the calculations. It was for this reason that it was possible to have values close to unity or even higher, as was reported by (Renganathan et al., 2012).

Table 3. LHV and cold gas efficiency of syngas

Experiment	Temperature (°C)	Bed material	<i>LHV</i>_{syngas} (MJ/kg)	CGE (ratio)
Gasification with CO ₂	600	Sand	2.5	0.16
	700		4.7	0.30
	800		9.8	0.63
	900		12.2	0.78
Pyrolysis	800	Sand	6.5	0.42
	900		7.9	0.51
Gasification with CO ₂	600	Biochar	3.3	0.21
	700		9.2	0.59
	800		11.4	0.73
	900		15.4	0.99
Gasification with steam	600	Biochar	3.3	0.22
	700		7.6	0.49
	800		12.8	0.82
	900		15.1	0.97

4. Conclusion

The energy balance of beech wood gasification in a pilot-lab scale fluidized bed reactor was analysed for different operation conditions. These variations included the change of operation temperature,

gasification agent and bed material. It was observed that when temperature increased there was a direct increase of the syngas energy amount, evidencing the positive impact of temperature to increase syngas energetic value. For CO₂ gasification, from 600 to 900°C syngas energy increased from 2.7 to 13.5 MJ/kg_{biomass}. Syngas energy incrementation was due to the conversions reactions that suffered biochar and tar. As biochar and tar yield were reduced due to gasification and cracking reactions, their energy was transferred to the syngas.

The syngas obtained from pyrolysis at 800 and 900°C, had an energetic value of 7.01 and 8.01 MJ/kg_{biomass}, compared to the syngas obtained gasification with CO₂ which values were 10.77 and 13.50 MJ/kg_{biomass}, at the same temperatures. It was concluded that syngas from gasification with CO₂ provided better energetic values than pyrolysis. The same was observed in the CGE values at these temperatures: gasification with CO₂ showed better CGE values (0.63 to 0.78) compared to pyrolysis (0.42- 0.51). Despite this, heat input required to perform the thermochemical conversion was lower for pyrolysis (4.4 to 5.0 MJ/kg_{biomass}) than for gasification (6.7 to 7.8 MJ/kg_{biomass}). This was attributed to the boudouard reaction which took place due to the use of CO₂ as gasification agent.

The change of bed material from sand to biochar boosted syngas energy content for CO₂ gasification from 3.53 to 17.18 MJ/kg_{biomass}, as for steam gasification 3.6 to 16.55 MJ/kg_{biomass}. This improvement was due to the more relevant cracking reactions of tar and biochar conversion. The presence of biochar as bed material also increased the heat input required for gasification, from 4.1 to 7.1 MJ/kg_{biomass} for CO₂ gasification and 1.5 to 6.9 MJ/kg_{biomass} for steam. As it was observed less heat for gasification was required from steam gasification than for CO₂ gasification. The latest was explained by the energy support of exothermic reactions present in steam gasification able to reduce the required energy. The CGE values range from 600 to 900°C for both gasification experience was very similar; CO₂ gasification range was from 0.21 to 0.99, meanwhile steam gasification was 0.22 to 0.97.

These findings point to the energetic efficiency of each set-up compared in the same reactor. As energy analysis was not able to quantify or qualify its degradation, an exergy balance for the presented results could be very useful for better understandings.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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Figure captions

Figure. 1. Fluidized bed gasifier setup.

Figure. 2. Energy input and output streams for gasifier.

Figure. 3. Effect of temperature on energy products distribution; Agent: CO₂, Bed material: Sand.

Figure. 4. a) Gas energy distribution b) Heat input for gasification at different temperatures; Agent: CO₂, Bed Material: Sand.

Figure. 5. Energy products distribution and heat input for pyrolysis and gasification; Agent CO₂/N₂, Bed material: Sand.

Figure. 6. Comparison between pyrolysis and gasification; Agent CO₂/N₂, bed material: sand a) energy distribution for gases and b) heat input Vs Syngas Energy.

Figure.7. Products energy distribution for gasification; Agent: CO₂/H₂O, Bed material: biochar a) CO₂ and b) steam.

Figure. 8. Gas energy distribution; Agent: CO₂/H₂O, Bed material: biochar a) CO₂ b) steam gasification.

Figure. 9. Heat input for gasification: Agent: CO₂/H₂O, Bed material: biochar.

Nomenclature:

C_p Heat Capacity [kJ/kmol K]

CGE	Cold gas efficiency [-]
$\dot{E}n$	Energy rate [MJ/kg _{biomass}]
h°_f	Enthalpy of formation [MJ/ kmol]
LHV_{fuel}	Low heating Value of fuel [kJ/kg]
LHV_{syngas}	Low heating Value of syngas [kJ/kg]
m	Mass [kg]
n	Mol flow rate [mol/kg _{biomass}]
P_i	Partial pressure [atm]
\dot{Q}_{loss}	Loss energy rate [MJ/kg _{biomass}]
T°	Reference Temperature [°C]
T	Temperature [°C]
x	Weight Fraction [%]

Subscripts

C	Carbon
Ch	Chemical.
H	Hydrogen
in	inlet
i	i th species.
Ki	Kinetic.
N	Nitrogen
O	Oxygen
out	outlet
Ph	Physical.
Po	Potential.

Abbreviations

LHV	Low Heating Value.
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Greek letters

Δh	Enthalpy change [kJ/kmol]
α	Stoichiometry coefficient of CO ₂
β	Stoichiometry coefficient of H ₂ O

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