Research article

Co-gasification of beech-wood and polyethylene in a fluidized-bed reactor

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ABSTRACT

The co-gasification of beech-wood and polyethylene has been investigated in a lab-scale fluidised-bed reactor in the presence of four different types of bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite). ZSM-5 zeolite is very effective as a catalytic bed material in fluidized-bed reactor for wood-only gasification and co-gasification in terms of high hydrogen production and CGE. Na-Y zeolite is more effective compared with ZSM-5 zeolite in co-gasification of the beech-wood and polyethylene process. The catalytic activity in co-gasification of beech-wood and polyethylene can be ranked accordingly: Na-Y zeolite > ZSM-5 zeolite > olivine. In general, higher amounts of steam injected in the fluidized-bed reactor and more polyethylene would lead to higher hydrogen production in the co-gasification process.

1. Introduction

Depending on the means of production, hydrogen can be considered a clean energy that has the potential to reduce the world consumption of fossil fuels to meet sustainability targets [1]. Currently, the methods to produce hydrogen energy are relatively high cost. There is around $5 \times 10^{11}\,N\,m^3$ of hydrogen production in the world every year, and around 96% of this hydrogen produced from fossil fuels. The principal production routes are methane reforming (48%), oil/naphtha reforming (30%), coal gasification (18%) and water electrolysis (3.9%) [2]. The costs and sources are issues for the development of 'hydrogen economy' [3]. There is increasing effort to develop new feedstocks to produce hydrogen. The use of waste feedstock can be a potentially significant source because it can help solve waste disposal issues and maximise the value of wastes by producing hydrogen rich syngas and other fuels [4].

With the rapid increase in energy demand around the world, biomass has become one of the most popular alternative energy sources [5,6]. A major advantage being that, unlike renewable such as wind and solar energy, biomass can easily be converted into liquid (methanol, ethanol or other hydrocarbons) and gaseous fuel (hydrogen rich syngas) [7].

The large quantity of plastics consumption around the world causes enormous amounts of waste plastics production. In 2017, the global plastics production reach to 348 million tonnes and the Europe contributes to 64.4 million tonnes [8]. In 2012, 65.41 million tonnes of polyethylene (PE), 52.75 million tonnes of polypropylene (PP),

19.8 million tonnes of polyethylene terephthalate (PET) and 10.55 million tonnes of polystyrene (PS) were produced in the world [9]. There is approximately 19.9 million tonnes of waste plastics generated every year in Europe. One of the main waste plastics generated in the EU is polyethylene which includes high density polyethylene (HDPE) and low density polyethylene (LDPE) [10]. Waste plastic has a relatively high content of hydrogen. Many researchers have studied the thermo-chemical decomposition of plastics and proposed the potential of producing hydrogen [11–14]. It is suggested that the availability of large quantities of waste plastic could produce a significant amount of hydrogen. Gasification as a thermochemical conversion technique with the addition of gasifying agent such as air, oxygen and steam has been applied widely to convert biomass into high calorific value gaseous products [15]. Gasification of plastics is also more desirable for energy recovery of waste plastics to prevent the waste going to landfill [16–18].

Co-gasification has also been studied by researchers as the synergies between different feedstock would increase the heating value of the gaseous products [19,20]. Pinto et al. [19] have investigated the co-gasification of coal, biomass and waste plastics in a fluidized-bed system. They reported that the improved gasification temperature would boost the further cracking of formed hydrocarbons to release more hydrogen where the tar formation would reduce simultaneously. An increasing flow rate of steam will promote the reforming reaction during the co-gasification process that would increase the heating value of gaseous products. Pinto et al. [20] also reported that the addition of

plastics will promote more hydrogen production accompanied with less carbon monoxide production from pine gasification by using a circular cross-section gasifier. Alvarez et al. [21] found that the hydrogen production will be increased by adding plastics to biomass pyrolysis-gasification process with a two-stage fixed-bed reactor.

Co-gasification is a more effective technique that would also help to ease the problems generated during the gasification of single feedstock, such as the incomplete gasification of plastics caused by reducing the fine dust and tar formation in biomass gasification process [20,22–25]. Lopez et al. [22] reported the synergetic effect of the co-gasification of high density polyethylene with forest pine wood in a conical spouted bed reactor reduces the tar and char formation and increases carbon conversion efficiency. Mastellone et al. [23] investigated the co-gasification of coal, plastics and wood in a bubbling fluidized bed reactor. They concluded the presence of wood and coal in plastics gasification reduces the tar production. Aznar et al. [24] investigated the optimal condition for co-gasification of coal, waste plastics and biomass. They found the optimal temperature to be at 850 °C and equivalent ratio at 0.36. The addition of waste plastics in the gasification of coal and biomass could help to ease the problems generated from seasonal biomass.

To convert the biomass and plastics into gaseous products is a very complex process that involves many reactions. Zhang et al. [26] described the steam reforming and hydrocarbon decomposition process of hydrocarbons related to Eqs. (1) and (2).

$$C_nH_m + 2H_2O \rightarrow nCO + \left(n + \frac{m}{2}\right)H_2 \tag{1}$$

$$C_nH_mO_z \rightarrow zCO + (n - z)C + H_2 \tag{2}$$
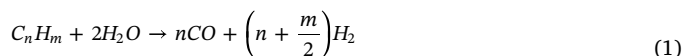
Reza [27] and Gao et al. [28] summarized the gasification reaction as an endothermic reaction that will decompose biomass or plastics to produce hydrogen-rich syngas at a temperature between 650 and 1200 °C. The involved reactions are the char gasification reaction ($\Delta H = 131.5 \, kJ \, mol^{-1}$), as shown in Eq. (3); the water gas shift reaction ($\Delta H = -41 \, kJ \, mol^{-1}$), as shown in Eq. (4); the steam methane reforming reaction ($\Delta H = 206 \, kJ \, mol^{-1}$), as shown in Eq. (5); the Boudouard reaction ($\Delta H = 172 \, kJ \, mol^{-1}$), as shown in Eq. (6) and the methanation reaction ($\Delta H = -74.8 \, kJ \, mol^{-1}$), as shown in Eq. (7).

$$C + H_2O \rightarrow CO + H_2 \tag{3}$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \tag{4}$$

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 \tag{5}$$

$$C + CO_2 \rightarrow 2CO \tag{6}$$

$$C + 2H_2 \rightarrow CH_4 \tag{7}$$

The catalyst used plays an important role in the gasification process, especially in fluidized bed gasification. Adding plastics to beech-wood increase considerably the volatility of the fuel. This translates in higher propensity to tars production and fines entrainment. When highly volatile materials such as plastics, are fed into a fluidized bed from above, the rapidly devolatilise and do not get in contact with the fluidising material. Alternatively, the fuel can be fed directly inside the fluidized bed, using a catalyst within the bed inventory. This could reduce the number of unit operations and simplify the process. Olivine, Na-Y zeolite and ZSM-5 zeolite are going to be investigated as bed materials in this research.

Olivine is one of the mineral catalysts. It mainly consist of silicate minerals with magnesium and irons cations in the silicate tetrahedral structure [29]. The catalytic activity of olivine relates to the magnesite (MgO) and iron oxide (Fe_2O_3) contents [30]. Olivine has been reported as one of the catalysts to minimize tar formation in the gasification process [22,31]. Lopez et al. [22] used olivine as bed material to eliminate tar formation in co-gasification of biomass and polyethylene reaction. Marinkovic et al. [31] studied the activity of olivine which presents the beneficial catalytic role for tar destruction in an indirect

biomass gasifier and promotes more hydrogen production. Alkali metal based catalysts have been applied as one of the most effective types of catalyst for tar reduction and good resistance for coke formation, which can either be used as it is or with a support [32]. The alkali content of the catalyst in the gasification process would promote coke/char gasification reaction as shown in Eq. (3) [7]. Lee et al. [33] found the addition of sodium carbonate promotes the gas production in gasification of rice straw reaction and that they also observed the sodium carbonate promotes the highest gas production compared with other alkali metal carbonates (K, Cs and Li). Zeolites are crystalline aluminosilicate catalysts with a porous structure made of tetrahedra of four oxygen anions surrounding a silicon or aluminium ion as the primary building block. The structure of zeolite formed by the arranged combination of silica and alumina tetrahedra with different pore sizes, which make the zeolite with an open framework structure consist of microspore channels. Y-zeolite with faujasite structure, with large channels and supercages is one of the widely applied types of zeolite in catalytic cracking of larger hydrocarbon molecules [34]. ZSM-5 zeolite has a smaller pore sizes in a pentasil - structure [35,36].

Although many researchers have worked on the co-gasification of different woody biomass and plastics, but there still unclear reaction characters as the complexity of the feedstocks. Co-gasification is even more complicated than gasification because of chain reaction and synergetic interactions between the different feedstocks. The issues occurred during the co-gasification are still need to be investigated, as it could be the reasons to hinder the future commercialisation. To find out the optimum operation condition specifically for co-gasification of beech wood and polyethylene (PE), in this research, low density PE was added in the beech-wood gasification process in a fluidized-bed reactor to explore how the gaseous products are affected. The research starts with the wood only gasification using four different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) as the reference. Then a mixture of beech-wood and PE with 1:1 weight ratio was investigated to find out how the bed materials affect the gaseous production from the co-gasification process. Different steam injection rate (0 and 400 g h^{-1}) and beech-wood-to-PE ratio (4:0, 1:1 and 3:1) were then investigated.

2. Materials and methods

2.1. Feedstock

The beech-wood sawdust sample with particle size around 3 mm was obtained from the Energy Research Centre of The Netherlands (ECN) part of Netherlands Organisation for Applied Scientific Research TNO and the low density polyethylene (PE) pellets with size around 6 mm were supplied by Vantage Polymer Ltd. The thermochemical properties of beech-wood and polyethylene are list in Table 1. The ultimate analysis and lower heating value of beech-wood and PE were provided by ECN using established method [37]. The proximate analysis of beech-wood was estimated by using a Perkin Elmer Pyris 1 TGA which is shown in Table 1. Approximately 4 mg of beech-wood sample was placed in the sample crucible and dried at 105 °C for 30 min with 20 °C min^{-1} heating rate for moisture removal. Then the sample was heated to 700 °C with a 5 °C min^{-1} ramp rate and maintained at 700 °C for 30 min to estimate the volatiles from the beech-wood. The gas flow then was switched to air for 5 min to oxidize the carbon, so that the carbon content could be quantified. This method was reported by Saldarriaga et al. [38].

The bed materials silica sand and Na-Y zeolite were also from ECN, olivine from Magnolithe GmbH, Austria and ZSM-5 zeolite was from ACS Materials LLC® with silicon oxide to alumina molar ratio of 38. All of the four bed materials were sieved with the particle size between 0.25 and 0.5 mm. The chemical structures of the polymer molecule are normally a linear or branched chain or a network with peripheral atoms or atom groups. Every polymer structure consists of a summation of structural groups, which include hydrocarbon groups, non-hydrocarbon groups and composed groups (such as –COOH and –CONH$_2$). PE with a

Table 1
Thermochemical properties of beech-wood and polyethylene.

	Beech-wood	PE
Ultimate analysis (% dry basis)		
C	48.1	85.8
H	5.9	14.2
O	45.4	0.0
N	0.2	0.0
Proximate analysis (wt% wet basis)		
Volatiles	74.8	–
Fixed carbon	15.7	–
Ash 750 °C	0.7	–
Moisture	8.8	–
Lower heating value (MJ kg^{-1} dry basis)	15.0[a]	44.2[a]

[a] Data from ECN-TNO.

relatively simple structure, only contains –CH. The polymer structures terminated with end-groups play an important role in their chemical prosperities but not on the physical properties [39].

2.2. Experimental setup

The gasification of beech-wood and co-gasification of beech-wood and polyethylene experiments were carried out at ECN with a lab-scale fluidized-bed reactor as shown in Fig. 1. The fluidized-bed reactor has a 78 mm inner diameter, freeboard with a diameter of 102 mm and height of 900 mm, the total height of the reactor is 1630 mm. The steam was fed at 150 °C with targeting injection rate. Typically, 1 kg of bed material was placed in the gasifier. Beech-wood and PE pellets were co-fed from the fuel bankers with total feeding rate at 400 g h^{-1}. Neon was constantly kept at 10 mL$_N$·min^{-1} as balance gas in the co-gasification process to calculate the gaseous products. The air and steam were fed to the bottom of the gasifier and the equivalent ratio (ER) was kept constant for all cases by changing the flow rate of air. The ER is defined as

the ratio of the air or oxygen to the system divided by that required for complete combustion [40]. The produced gaseous products from the devolatilization of wood and PE were detected and quantified by different micro-gas chromatography analysers, all of the detected gaseous products are H_2, Ar/O_2, CO, CO_2, CH_2, C_2H_2, C_2H_4, C_2H_6, H_2S, C_6H_6 and C_7H_8. Tars were not analysed due to the difficulty of collection and sampling. Carbon conversion was calculated by dividing the carbon in produced gas to the total carbon inlet from feedstock.

This piece of work includes four investigations. Firstly, the effect of types of bed materials on gaseous products from steam gasification of beech-wood only has been investigated keeping the wood feed rate constant at 400 g h^{-1}, steam injection rate at 250 g h^{-1}, bed temperature at ~850 °C. Then, the effect of bed materials on co-gasification of wood and PE with the ratio at 1:1 has been investigated with a constant total fuel flow rate at 400 g h^{-1}, bed temperature of 850 °C and there was no steam addition. To investigate the effect of steam injection rate on the gaseous products from co-gasification of wood and PE, four bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) have been investigated with steam injection rate at 0, 400 and 800 g h^{-1}. The bed temperature was ~850 °C, total fuel flow feeding rate was 400 g h^{-1}, the wood-to-PE ratio at 1:1. Finally, to investigate the effect of wood-to-PE ratio three different compositions were investigated, namely 1:1, 3:1 and 4:0, with all other variables unchanged, with steam injection rate at 400 g h^{-1} and bed temperature at ~850 °C.

3. Results and discussion

3.1. Investigation the effect of bed materials on beech-wood steam gasification

Table 2 shows the gaseous production from beech-wood gasification by using a fluidized-bed reactor in the presence of silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite. The gasification temperature was at
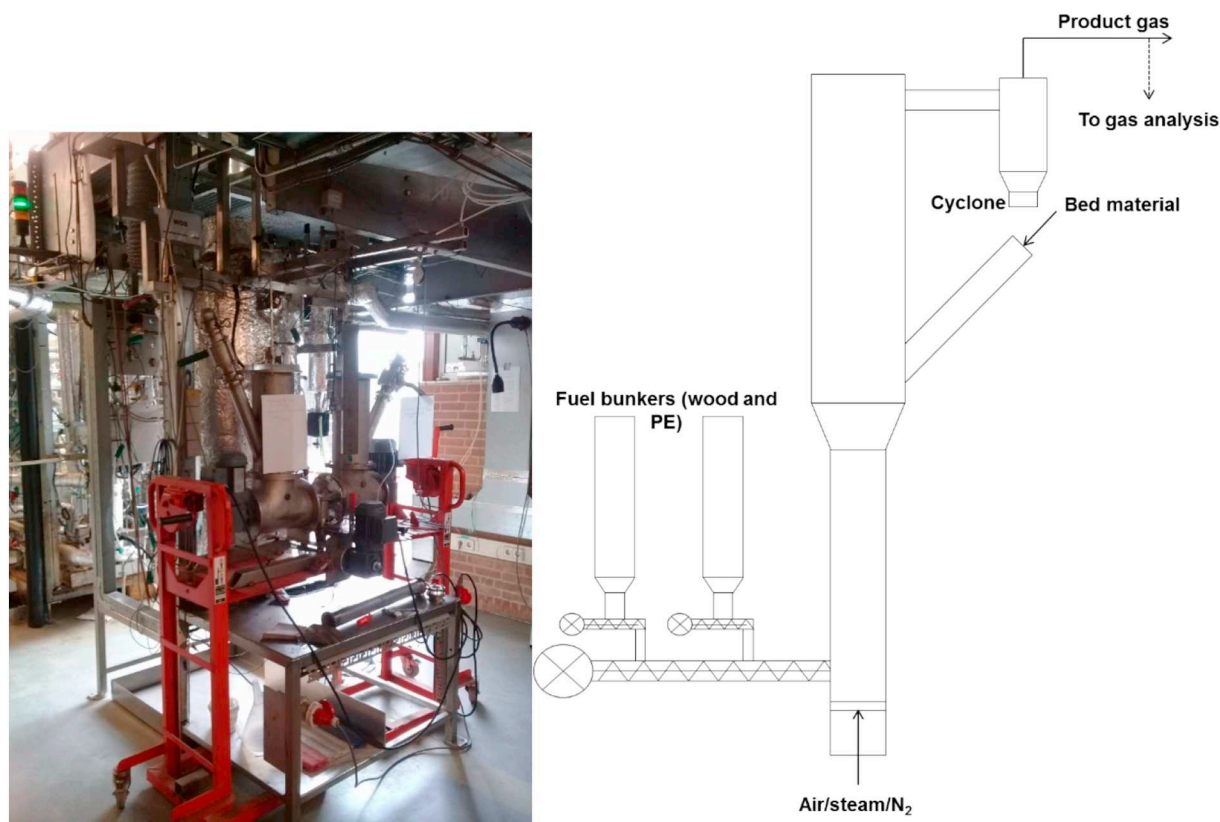

Fig. 1. Photography (left) and schematic diagram (right) of fluidized-bed reactor (ECN).

Table 2

Gaseous production of beech-wood gasification in a fluidized-bed reactor with different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) at 850 °C and steam injection rate 250 g h^{-1}.

Bed materials	Theoretical	Silica sand	Olivine	Na-Y zeolite	ZSM-5 zeolite
Feedstock 400 gh^{-1} (wood only)		–	–	–	–
Steam injection rate (gh^{-1})	250	250	250	250	250
Bed temperature (°C)	850	856	854	855	854
Experimental stable period		18:00–18:30	16:25–17:05	17:15–17:55	12:00–13:30
Experimental results					
Gas production (mol h^{-1})		Silica sand	Olivine	Na-Y zeolite	ZSM-5 zeolite
H_2	9.85	5.24	5.39	4.65	7.92
CH_4	0.31	1.25	1.30	1.23	1.00
CO	8.90	3.75	3.26	5.33	6.09
CO_2	7.07	6.87	7.19	5.67	6.36
C_2H_4	0.00	0.39	0.42	0.36	0.21
C_2H_6	0.00	0.01	0.015	0.009	0.008
C_2H_2	0.00	0.015	0.018	0.029	0.008
H_2S	0.008	0.0014	0.0008	0.0007	0.0000
C_6H_6	0.00	0.11	0.092	0.080	0.043
C_7H_8	0.00	0.007	0.028	0.014	0.0048
Total (N_2-H_2O $_{free}$)	25.85	17.64	17.71	17.37	21.64
Energy production (MJ h^{-1})	5.18	4.26	4.25	4.44	4.89
CGE (%)	86.32	72.78	72.56	75.79	83.57
Carbon conversion (%)	100.00	89.59	92.08	90.65	98.20

~850 °C with 250 g h^{-1} steam injection rate. The data was only collected when stable operation has been achieved. The cold gas efficiency (CGE) was calculated as the ratio of energy production (net calorific value) of the gaseous products divided by the total lower heating value of feedstock, biomass and PE at the corresponding compositions. For comparison, theoretical predictions at thermodynamic equilibrium (1 bar, 850 °C) are also added in Table 2.

The results shown in Table 2 indicate that the zeolite plays an important role in beech-wood gasification for hydrogen rich syngas production, with ZSM-5 zeolite given the highest hydrogen production and CGE which are 7.92 mol h^{-1} and 83.57%, respectively. These values are very close to those predicted by thermodynamic equilibrium, suggesting that tar production is indeed reduced when zeolites are used as bed materials. The carbon monoxide production reaches the highest amount of 6.09 mol h^{-1} in correspondence with the highest carbon conversion. Fig. 2 graphically summarized the results shown in Table 2 and shows that ZSM-5 zeolite gave the highest production of hydrogen compared to the other bed material compositions. Notably, the relatively high CGE values in all cases are indicative or high calorific value syngas production. This is also due to the presence of external heaters in the fluidised bed that allow lower ER to be used, while still maintaining temperature at above 800 °C.

These results indicate that the ZSM-5 zeolite promotes the carbon gasification reaction and forward direction in Eq. (3) of methane reforming reaction in Eq. (5). This is also supported by other studies [41]. Samolada et al. [41] reported that ZSM-5 zeolite addition in the biomass gasification process promotes tar reduction simultaneously with more gaseous production. It plays the catalyst role to crack heavy molecular weight products to lighter molecular weight products.

Na-Y zeolite promotes the CGE to 75.79%, which is higher than silica sand and olivine. This relates to the relatively high production of carbon monoxide at 5.33 mol h^{-1}. However, the hydrogen production is only 4.65 mol h^{-1}, and carbon dioxide generation is also the lowest of the various catalysts. The result could relate to the Na-Y zeolite promoting the carbon gasification reaction in Eq. (3), the reverse water gas shift reaction in Eq. (4), the forward direction of methane reforming reaction in Eq. (5) and the Boudouard reaction in Eq. (6). This could be caused by the large pore structure of Y-zeolite compared to ZSM-5 zeolite, such that the produced hydrocarbon gases pass through the

supercages without cracking. Gayubo et al. [42] showed that the ZSM-5 zeolite is more suitable for cracking of hydrocarbons shorter than C_{12} which have higher thermal stability. It has been reported that the products of biomass gasification at > 800 °C result in gaseous products with lower molecular weight [43]. Olivine does not appear to play a relevant catalytic role in beech-wood gasification as there is little difference in the hydrogen production and CGE compared with silica sand. This result is consistent with previous research [44], which has shown the pore structure of olivine is damaged by high temperature sintering.

3.2. Effect of bed materials on co-gasification of beech-wood and polyethylene (PE)

Table 3 shows the gaseous products from co-gasification of beech-wood and PE at ~850 °C with no steam injection. The first observation with comparison with theoretical results at equilibrium is that in all cases, much lower cold gas efficiencies were observed. This might be due to the addition of plastics material, which is more prone to production of organic species (including tars), which are not measured in these tests. This is also confirmed by the lower carbon conversion values, which indicate that other carbon species were produced. However, some generic conclusions with regards to catalysts activity could be drawn. For example, the results show that Na-Y zeolite this time gave the highest hydrogen production of 7.08 mol h^{-1} while ZSM-5 zeolite gave a relatively high amount of hydrogen production at 6.26 mol h^{-1}. This could be due to the effect of alkaline metal that crack volatile products from biomass [45] and plastics [46,47], in line with the results from other studies. In a review of catalysts for biomass gasification, Bulushev et al. [45] showed that zeolite has been widely applied for catalytic biomass or bio-oil upgrading due to its porous structure. Yumiko et al. [46] reported that the isobutene (C_4) and isopentane (C_5) are selectively produced from polyethylene with Na-Y zeolite catalyst in catalytic decomposition process. In the review of Kunwar et al. [48], different types of zeolite for plastics cracking process were considered. In agreement with the findings, Na-Y-zeolite was found to give the highest CGE at 69.08% associated with the highest energy production is 8.13 MJ h^{-1}.

Fig. 3 shows the gas compositions of beech-wood and PE co-gasification in a fluidized-bed reactor. There is a significant difference
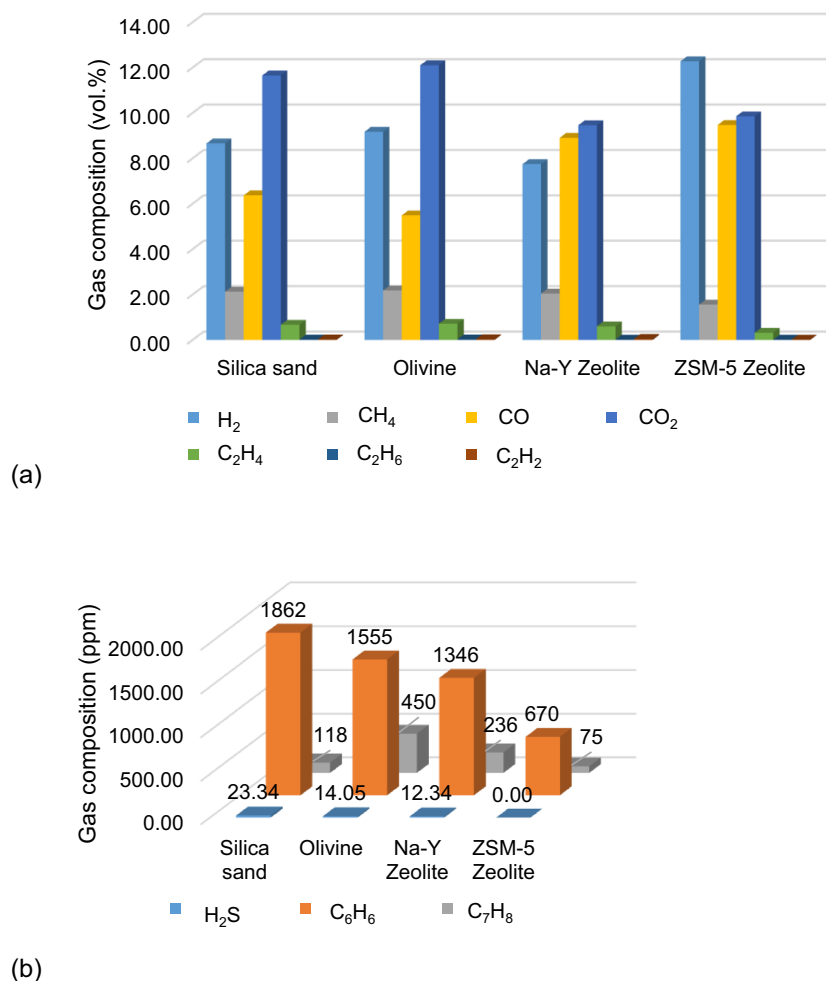
Fig. 2. Gas compositions of beech-wood gasification in a fluidized-bed reactor with different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) at 850 °C, steam injection rate 250 g h^{-1} and ER at 0.3. (a) H_2, CH_4, CO, CO_2, C_2H_2, C_2H_6 and C_2H_2; (b) H_2S, C_6H_6 and C_7H_8.

Table 3
Gaseous production of beech-wood and polyethylene (PE) co-gasification in a fluidized-bed reactor with different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) at 850 °C, no steam injection rate.

Bed materials	Theoretical	Silica sand	Olivine	Na-Y zeolite	ZSM-5 zeolite
Feedstock 400 g h^{-1} (wood-to-PE ratio)	1:1	1:1	1:1	1:1	1:1
Steam injection rate (g h^{-1})	0	0	0	0	0
Bed temperature (°C)	850	858	853	848	848
Experimental stable period		11:30–12:20	11:15–12:05	08:00–11:50	13:00–14:00

| Experimental results					
Gas production (mol h^{-1})		Silica sand	Olivine	Na-Y zeolite	ZSM-5 zeolite
H_2	18.45	2.14	1.61	7.08	6.26
CH_4	0.02	2.10	2.02	2.20	2.38
CO	19.73	3.63	2.19	6.90	5.67
CO_2	1.84	5.09	6.10	4.68	4.90
C_2H_4	0.00	2.38	2.47	1.37	1.35
C_2H_6	0.00	0.096	0.12	0.10	0.081
C_2H_2	0.00	0.095	0.067	0.0077	0.026
H_2S	0.004	0.0016	0.0007	0.0006	0.0006
C_6H_6	0.00	0.35	0.30	0.20	0.22
C_7H_8	0.00	0.036	0.040	0.030	0.042
Total (N$_2$-H$_2$O $_{free}$)	40.04	15.92	14.92	22.57	20.93
Energy production (MJ h^{-1})	10.10	7.85	7.19	8.13	7.78
CGE (%)	85.35	66.74	61.12	69.08	66.15
Carbon conversion (%)	100.00	85.8	84.2	85.1	82.9
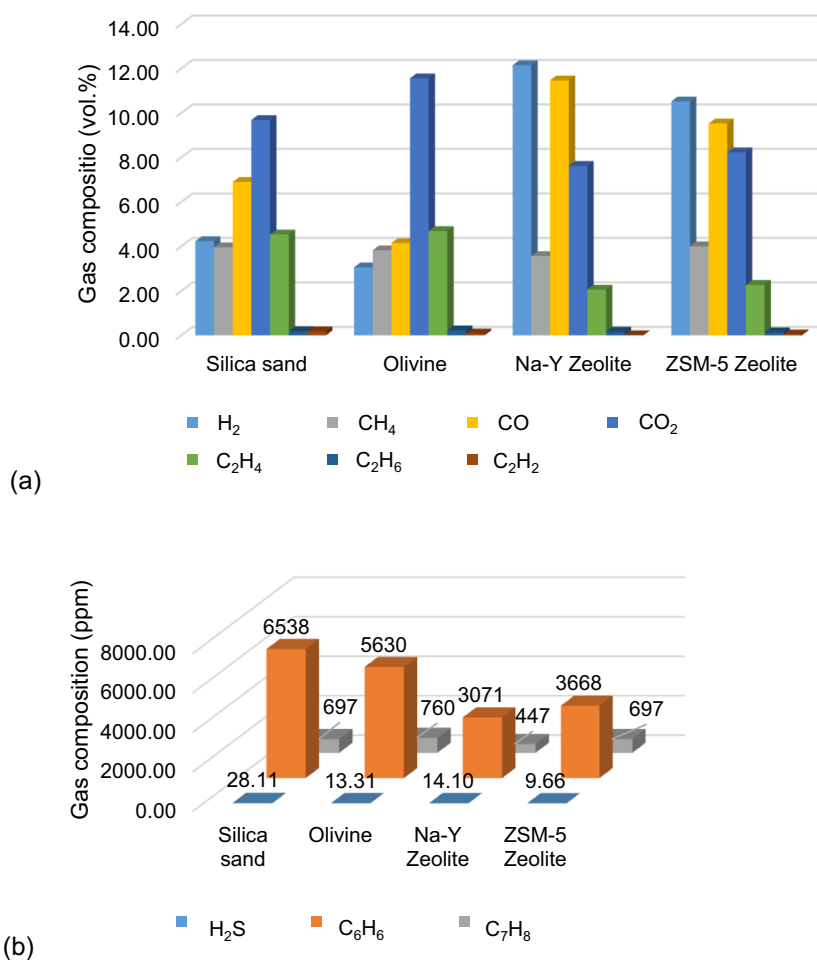
Fig. 3. Gas composition of beech-wood and polyethylene (PE) co-gasification in a fluidized-bed reactor with different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) at 850 °C, no steam injection rate and ER at 0.27. (a) H_2, CH_4, CO, CO_2, C_2H_2, C_2H_6 and C_2H_2; (b) H_2S, C_6H_6 and C_7H_8.

between the hydrogen production from zeolite and olivine, but no obvious difference between Na-Y zeolite and ZSM-5 zeolite. There is no catalytic effect of olivine in co-gasification of beech-wood and PE, the hydrogen production with olivine is even less than that for silica sand as shown in Table 3. Also, the carbon conversion has no significant difference. The results are consistent with those in Section 3.2 confirming that the olivine has no catalytic activity in both of wood gasification and co-gasification process. Furthermore, both types of zeolite (Na-Y zeolite and ZSM-5 zeolite) show catalytic activity in co-gasification, while Na-Y zeolite is not active for the wood gasification process.

3.3. Effect of steam injection rate on co-gasification of beech-wood and polyethylene (PE)

Table 4 lists the gaseous products of co-gasification with beech-wood-to-PE ratios of 1:1, gasification temperature at ~850 °C. The only variable in this set of experiments is the steam injection rate which increased from 0 to $400 \, g \, h^{-1}$.

As it can be appreciated from thermodynamic model predictions, the addition of the steam boosts the hydrogen and carbon dioxide productions. The same behaviour is observed in experimental results, regardless of the types of bed materials. This is mostly due to the forward direction of waste-gas-shift reaction in Eq. (4). However, the addition of steam does not have a significant effect on the energy production and CGE except in the co-gasification with ZSM-5 zeolite. The energy production from co-gasification of beech-wood and PE with ZSM-5 zeolite increased from 7.78 to 8.74 MJ h^{-1}, when the steam feed is increased from 0 to $400 \, g \, h^{-1}$. This occurs simultaneously with the

increase in CGE from 66.15 to 74.28%. Na-Y zeolite has a negligible influence compared with ZSM-5 zeolite in terms of hydrogen production and CGE. Na-Y zeolite has no obvious effect on gasification of beech-wood or co-gasification, regardless of the steam injection rate and composition of feedstock that is consistent with the results shown in Table 2 in Section 3.1.

Table 4 shows that the olivine plays an important catalytic role in co-gasification of beech-wood and PE with steam, but not in the co-gasification process without steam injection. The olivine is more active with steam in presence in the co-gasification process, which promotes more tar cracking, corresponding with higher hydrogen production. For example, the hydrogen production increases dramatically from 1.61 to 5.80 mol h^{-1} when the $400 \, g \, h^{-1}$ steam feed is added to the process. Simultaneously, the CGE and energy production increase from 7.19 to 8.06 MJ h^{-1} and 61.12 to 68.52%, respectively. The carbon conversion is in the similar trend as CGE, the highest CGM normally comes with the highest carbon conversion.

In Table 4, we can also observe that H_2S production was generally increased with the addition of steam in the co-gasification process. One reason could be the sulphur was retained in solid residue, if any, within the bed. At contact with steam, this is released from the solid state and measured as H_2S. Another reason could be explained by the involved reaction between COS (or any other organic sulphur component) with steam and hydrogen toward H_2S production. Although our experimental tests did not provide enough evidence of this effect, similar results have been reported in other studies, with steam-to-carbon ratio being one of the most important element to affect the H_2S production in steam gasification of biomass [49,50]. The increase of the steam-

Table 4
Gaseous production from beech-wood and polyethylene (PE) co-gasification in a fluidized-bed reactor with different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) and different steam injection rate (0, 400 and 800 g h^{-1}) at 850 °C.

Bed materials	Theoretical		Silica sand		Olivine		Na-Y Zeolite		ZSM-5 Zeolite	
Feedstock 400 gh^{-1} (Wood:PE ratio)	1:1	1:1	1:1	1:1	1:1	1:1	1:1	1:1	1:1	1:1
Steam injection rate (gh^{-1})	0	400	0	400	0	400	0	400	0	400
Bed temperature (°C)	850	850	858	861	853	852	848	850	848	851
Experimental stable period			11:30–12:20	15:30–16:15	11:15–12:05	14:00–15:15	08:00–11:50	12:05–13:25	13:00–14:00	14:45–16:00

Experimental results	Theoretical		Silica sand		Olivine		Na-Y zeolite		ZSM-5 zeolite	
Steam injection rate (g h^{-1})	0	400	0	400	0	400	0	400	0	400
Gas production (mol h^{-1})										
H_2	18.45	25.06	2.14	7.45	1.61	5.80	7.08	7.43	6.26	9.92
CH_4	0.02	0.00	2.10	1.99	2.02	2.18	2.20	2.28	2.38	2.48
CO	19.73	13.18	3.63	3.72	2.19	2.28	6.90	5.93	5.67	6.31
CO_2	1.84	8.39	5.09	6.71	6.10	7.47	4.68	5.70	4.90	6.43
C_2H_4	0.00	0.00	2.38	1.55	2.47	2.25	1.37	1.63	1.35	1.30
C_2H_6	0.00	0.00	0.096	0.050	0.12	0.077	0.10	0.067	0.081	0.069
C_2H_2	0.00	0.00	0.095	0.073	0.067	0.067	0.0077	0.040	0.026	0.034
H_2S	0.004	0.004	0.0016	0.0035	0.0007	0.0017	0.0006	0.0024	0.0006	0.0010
C_6H_6	0.00	0.00	0.35	0.27	0.30	0.30	0.20	0.20	0.22	0.19
C_7H_8	0.00	0.00	0.036	0.012	0.04	0.049	0.030	0.030	0.042	0.034
Total (N$_2$-H$_2$O $_{free}$)	40.04	46.63	15.92	21.83	14.92	20.47	22.57	23.31	20.93	26.77
Energy production (MJ h^{-1})	10.10	9.85	7.85	7.55	7.19	8.06	8.13	8.33	7.78	8.74
CGE (%)	85.35	83,20	66.74	64.15	61.12	68.52	69.08	70.81	66.15	74.28
Carbon conversion (%)	100.00	100.00	85.8	81.8	84.2	90.0	85.1	88.1	82.9	92.1

to-carbon ratio causes a decrease of COS, and an increase of H_2S production, according to:

$$COS + H_2O \rightarrow H_2S + CO_2 \tag{8}$$

$$COS + H_2 \rightarrow H_2S + CO \tag{9}$$

3.4. Effect of beech-wood to polyethylene (PE) ratio on co-gasification

Carbon conversion efficiency, however, is affected by the high volatile content of plastics, which translates into higher tars content. Despite the catalytic effect of some bed materials, it appears that the fraction of devolatilising feedstock which escapes the bed at high temperature is still significant. Future work will highlight the effect of lowering the location of the feed into the fluidised bed reactor to maximise contact time with catalytic materials.

Table 5 lists the gaseous products from co-gasification of beech-wood and PE (experimental only). The results show that with the beech-wood as the dominant feedstock, energy production is lower than when the feedstock composition contains more PE, while the carbon conversion increases. For example, as beech-wood-to-PE ratio increases from 1:1 to 3:1 in presence of silica sand and Na-Y zeolite, the energy production reduces from 7.55 to 5.50 MJ h^{-1} and 60.01 to 57.38 MJ h^{-1}, respectively. Carbon conversion increases from 88.1 to 95.7%. The results are consistent with the significant difference in the Lower heating value (LHV) of the feedstock as shown in Table 1, in that the LHV of PE is triple that of beech-wood. Aznar et al. [24] reported similar results; the feedstock composition significantly influences the flue gas from co-gasification of coal and plastics as they all have different lower heating values. The result could also be explained by the more hydrogen element involved in the co-gasification process as shown in with the smaller beech-wood-to-PE ratio. All experiments were conducted with a constant feedstock rate of 400 g h^{-1}. As shown in Table 1, the hydrogen composition of PE is 14.2% and hydrogen composition of beech-wood is much less than PE (5.9%). When the total feedstock rate is the same, the smaller beech-wood-to-PE ratio indicates more hydrogen element input in the co-gasification process which explains the higher hydrogen content in the products. Carbon conversion

efficiency, however, is affected by the high volatile content of plastics, which translates into higher tars content. Despite the catalytic effect of some bed materials, it appears that the fraction of devolatilising feedstock which escapes the bed at high temperature is still significant. Future work will highlight the effect of lowering the location of the feed into the fluidised bed reactor to maximise contact time with catalytic materials.

4. Conclusions

Beech-wood gasification and co-gasification with low density polyethylene (PE) was carried out in a fluidized-bed reactor to investigate the effect of bed materials, steam injection and feedstock compositions.

- ZSM-5 zeolite plays a significant role in the gasification of beech-wood and gives the highest hydrogen production and energy conversion, which are 7.92 mol h^{-1} and 83.57%, respectively.
- Both ZSM-5 zeolite and Na-Y zeolite have a significant influence on co-gasification of beech-wood and polyethylene (PE) in terms of high hydrogen production and CGE. Furthermore, Na-Y zeolite has a greater influence compared to ZSM-5 zeolite. The hydrogen produced in the presence of Na-Y zeolite is 0.82 mol h^{-1}, which is higher than the hydrogen produced in the presence of ZSM-5 zeolite. The CGE for the co-gasification of beech-wood and PE in the presence of Na-Y zeolite is ~3% higher than in the presence of ZSM-5 zeolite.
- The addition of steam in the co-gasification of beech-wood and PE promotes hydrogen production but has negligible influence on the CGE, except in the presence of ZSM-5 zeolite.
- The composition of feedstock has a strong influence on hydrogen content from the co-gasification process, with the smaller beech-wood-to-PE ratio giving higher hydrogen production. The smaller beech-wood-to-PE ratio also gives higher energy production because the Lower heating value (LHV) of PE is almost triple that of beech-wood. However, carbon conversion efficiency is negatively affected by the presence of plastics.

Table 5

Gaseous production of co-gasification of beech-wood and polyethylene in a fluidized-bed reactor in presence of different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) with different beech-wood-to-polyethylene (PE) ratio (1:1, 3:1 and 4:0) with 400 g h^{-1} steam injection rate at 850 °C.

Bed materials	Silica sand		Olivine	Na-Y zeolite		ZSM-5 zeolite	
Feedstock 400 g Wood:PE ratio)	1:1	3:1	1:1	1:1	3:1	1:1	4:0
Steam injection rate (gh^{-1})	400	400	400	400	400	400	400
Bed temperature (°C)	861	861	852	850	850	851	852
Experimental stable period	15:30–16:15	16:45–17:16	14:00–15:15	12:05–13:25	15:15–15:50	14:45–15:36	13:30–15:00

Experimental results	Silica sand		Olivine	Na-Y zeolite		ZSM-5 zeolite	
Wood-to-PE ratio	1:1	3:1	1:1	1:1	3:1	1:1	4:0
Gas production (mol h^{-1})							
H_2	7.45	6.06	5.80	7.43	4.77	9.92	8.13
CH_4	1.99	1.49	2.18	2.28	1.67	2.48	1.00
CO	3.72	3.53	2.28	5.93	5.94	6.31	6.01
CO_2	6.71	7.69	7.47	5.70	6.22	6.43	6.55
C_2H_4	1.55	0.92	2.25	1.63	1.07	1.30	0.23
C_2H_6	0.05	0.023	0.077	0.067	0.036	0.069	0.0079
C_2H_2	0.073	0.037	0.067	0.040	0.050	0.034	0.013
H_2S	0.0035	0.0024	0.0017	0.0024	0.001	0.0010	0.0001
C_6H_6	0.27	0.17	0.30	0.20	0.13	0.19	0.047
C_7H_8	0.012	0.0070	0.049	0.030	0.021	0.034	0.0054
Total (N_2-H_2O free)	21.83	19.93	20.47	23.31	19.91	26.77	21.99
Energy production (MJ h^{-1})	7.55	5.50	8.06	8.33	6.18	8.74	4.97
CGE (%)	64.15	62.48	68.52	70.81	70.17	74.28	84.94
Carbon conversion (%)	81.8	88.2	90.0	88.1	95.7	92.1	98.2

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