

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 5×10^{11} N m³ 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

35 issues and maximise the value of wastes by producing hydrogen rich syngas and other fuels
36 [4].

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

41 The large quantity of plastics consumption around the world causes enormous amounts of
42 waste plastics production. In 2017, the global plastics production reach to 348 million tonnes
43 and the Europe contributes to 64.4 million tonnes [8]. In 2012, 65.41 million tonnes of
44 polyethylene (PE), 52.75 million tonnes of polypropylene (PP), 19.8 million tonnes of
45 polyethylene terephthalate (PET) and 10.55 million tonnes of polystyrene (PS) were
46 produced in the world [9]. There is approximately 19.9 million tonnes of waste plastics
47 generated every year in Europe. One of the main waste plastics generated in the EU is
48 polyethylene which includes high density polyethylene (HDPE) and low density polyethylene
49 (LDPE) [10]. Waste plastic has a relatively high content of hydrogen. Many researchers have
50 studied the thermo-chemical decomposition of plastics and proposed the potential of
51 producing hydrogen [11-14]. It is suggested that the availability of large quantities of waste
52 plastic could produce a significant amount of hydrogen. Gasification as a thermochemical
53 conversion technique with the addition of gasifying agent such as air, oxygen and steam has
54 been applied widely to convert biomass into high calorific value gaseous products [15].
55 Gasification of plastics is also more desirable for energy recovery of waste plastics to
56 prevent the waste going to landfill [16-18].

57 Co-gasification has also been studied by researchers as the synergies between different
58 feedstock would increase the heating value of the gaseous products [19, 20]. Pinto et al. [19]
59 have investigated the co-gasification of coal, biomass and waste plastics in a fluidized-bed
60 system. They reported that the improved gasification temperature would boost the further
61 cracking of formed hydrocarbons to release more hydrogen where the tar formation would
62 reduce simultaneously. An increasing flow rate of steam will promote the reforming reaction
63 during the co-gasification process that would increase the heating value of gaseous products.
64 Pinto et al. [20] also reported that the addition of plastics will promote more hydrogen
65 production accompanied with less carbon monoxide production from pine gasification by
66 using a circular cross-section gasifier. Alvarez et al. [21] found that the hydrogen production
67 will be increased by adding plastics to biomass pyrolysis-gasification process with a two-
68 stage fixed-bed reactor.

69 Co-gasification is a more effective technique that would also help to ease the problems
70 generated during the gasification of single feedstock, such as the incomplete gasification of
71 plastics caused by reducing the fine dust and tar formation in biomass gasification process
72 [20, 22-25]. Lopez et al. [22] reported the synergetic effect of the co-gasification of high
73 density polyethylene with forest pine wood in a conical spouted bed reactor reduces the tar
74 and char formation and increases carbon conversion efficiency. Mastellone et al. [23]
75 investigated the co-gasification of coal, plastics and wood in a bubbling fluidized bed reactor.
76 They concluded the presence of wood and coal in plastics gasification reduces the tar
77 production. Aznar et al. [24] investigated the optimal condition for co-gasification of coal,
78 waste plastics and biomass. They found the optimal temperature to be at 850 °C and
79 equivalent ratio at 0.36. The addition of waste plastics in the gasification of coal and biomass
80 could help to ease the problems generated from seasonal biomass.

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



86 Reza [27] and Gao et al. [28] summarized the gasification reaction as an endothermic
87 reaction that will decompose biomass or plastics to produce hydrogen-rich syngas at a
88 temperature between 650 - 1200 °C. The involved reactions are the char gasification
89 reaction ($\Delta H = 131.5 \text{ kJ mol}^{-1}$), as shown in Equation (3); the water gas shift reaction ($\Delta H = -$
90 41 kJ mol^{-1}), as shown in Equation (4); the steam methane reforming reaction ($\Delta H = 206 \text{ kJ}$
91 mol^{-1}), as shown in Equation (5); the Boudouard reaction ($\Delta H = 172 \text{ kJ mol}^{-1}$), as shown in
92 Equation (6) and the methanation reaction ($\Delta H = - 74.8 \text{ kJ mol}^{-1}$), as shown in Equation (7).



98 The catalyst used plays an important role in the gasification process, especially in fluidized
99 bed gasification. Adding plastics to beech-wood increase considerably the volatility of the

100 fuel. This translates in higher propensity to tars production and fines entrainment. When
101 highly volatile materials such as plastics, are fed into a fluidized bed from above, the rapidly
102 devolatilise and do not get in contact with the fluidising material. Alternatively, the fuel can
103 be fed directly inside the fluidized bed, using a catalyst within the bed inventory. This could
104 reduce the number of unit operations and simplify the process. Olivine, Na-Y zeolite and
105 ZSM-5 zeolite are going to be investigated as bed materials in this research.

106 Olivine is one of the mineral catalysts. It mainly consist of silicate minerals with magnesium
107 and irons cations in the silicate tetrahedral structure [29]. The catalytic activity of olivine
108 relates to the magnesite (MgO) and iron oxide (Fe_2O_3) contents [30]. Olivine has been
109 reported as one of the catalysts to minimize tar formation in the gasification process [22, 31].
110 Lopez et al. [22] used olivine as bed material to eliminate tar formation in co-gasification of
111 biomass and polyethylene reaction. Marinkovic et al. [31] studied the activity of olivine which
112 presents the beneficial catalytic role for tar destruction in an indirect biomass gasifier and
113 promotes more hydrogen production. Alkali metal based catalysts have been applied as one
114 of the most effective types of catalyst for tar reduction and good resistance for coke
115 formation, which can either be used as it is or with a support [32]. The alkali content of the
116 catalyst in the gasification process would promote coke/char gasification reaction as shown
117 in Equation (3) [7]. Lee et al. [33] found the addition of sodium carbonate promotes the gas
118 production in gasification of rice straw reaction and that they also observed the sodium
119 carbonate promotes the highest gas production compared with other alkali metal carbonates
120 (K, Cs and Li). Zeolites are crystalline aluminosilicate catalysts with a porous structure made
121 of tetrahedra of four oxygen anions surrounding a silicon or aluminium ion as the primary
122 building block. The structure of zeolite formed by the arranged combination of silica and
123 alumina tetrahedra with different pore sizes, which make the zeolite with an open framework
124 structure consist of microspore channels. Y-zeolite with faujasite structure, with large
125 channels and supercages is one of the widely applied types of zeolite in catalytic cracking of
126 larger hydrocarbon molecules [34]. ZSM-5 zeolite has a smaller pore sizes in a pentasil -
127 structure [35, 36].

128 Although many researchers have worked on the co-gasification of different woody biomass
129 and plastics, but there still unclear reaction characters as the complexity of the feedstocks.
130 Co-gasification is even more complicated than gasification because of chain reaction and
131 synergetic interactions between the different feedstocks. The issues occurred during the co-
132 gasification are still need to be investigated, as it could be the reasons to hinder the future
133 commercialisation. To find out the optimum operation condition specifically for co-gasification
134 of beech wood and polyethylene (PE), in this research, low density PE was added in the
135 beech-wood gasification process in a fluidized-bed reactor to explore how the gaseous

136 products are affected. The research starts with the wood only gasification using four different
 137 bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) as the reference. Then a
 138 mixture of beech-wood and PE with 1:1 weight ratio was investigated to find out how the bed
 139 materials affect the gaseous production from the co-gasification process. Different steam
 140 injection rate (0 and 400 g h⁻¹) and beech-wood-to-PE ratio (4:0, 1:1 and 3:1) were then
 141 investigated.

142

143 2 Materials and methods

144 2.1 Feedstock

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

158 *Table 1 Thermochemical properties of beech-wood and polyethylene.*

Ultimate analysis (% dry basis)	Beech-wood	PE
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

159 a. Data from ECN-TNO

160

161 The bed materials silica sand and Na-Y zeolite were also from ECN, olivine from Magnolithe
162 GmbH, Austria and ZSM-5 zeolite was from ACS Materials LLC[®] with silicon oxide to
163 alumina molar ratio of 38. All of the four bed materials were sieved with the particle size
164 between 0.25-0.5 mm. The chemical structures of the polymer molecule are normally a
165 linear or branched chain or a network with peripheral atoms or atom groups. Every polymer
166 structure consists of a summation of structural groups, which include hydrocarbon groups,
167 non-hydrocarbon groups and composed groups (such as –COOH and –CONH₂). PE with a
168 relatively simple structure, only contains –CH. The polymer structures terminated with end-
169 groups play an important role in their chemical prosperities but not on the physical properties
170 [39].

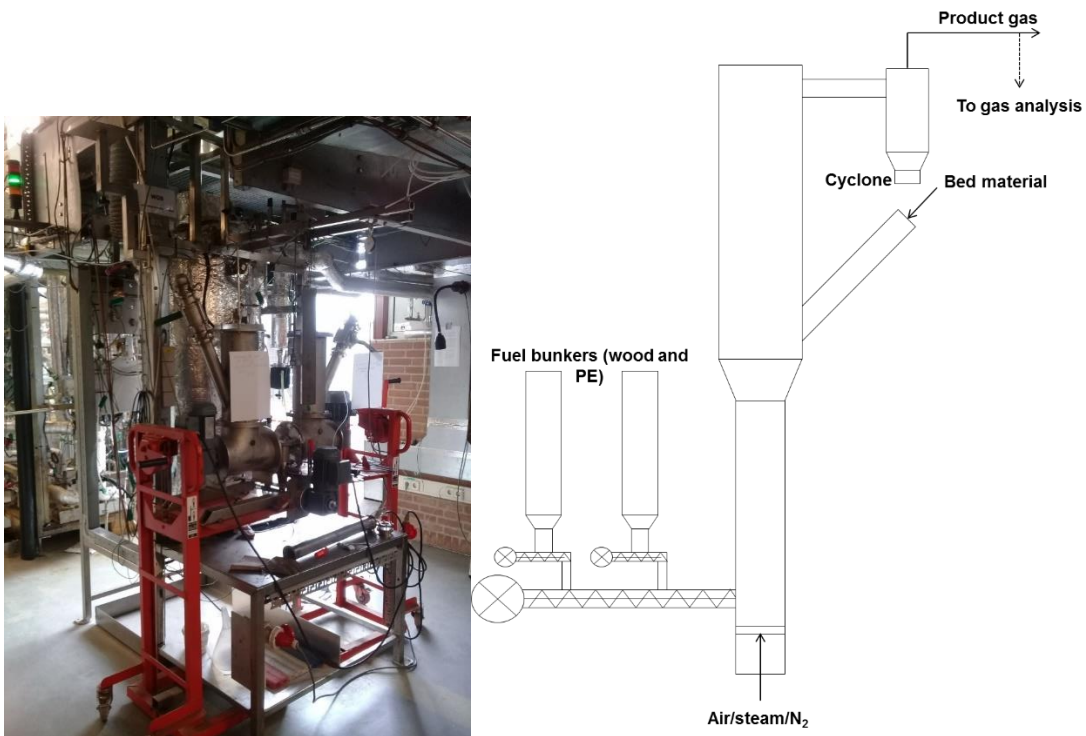
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172 2.2 Experimental setup

173 The gasification of beech-wood and co-gasification of beech-wood and polyethylene
174 experiments were carried out at ECN with a lab-scale fluidized-bed reactor as shown in
175 Figure 1. The fluidized-bed reactor has a 78 mm inner diameter, freeboard with a diameter of
176 102 mm and height of 900 mm, the total height of the reactor is 1630 mm. The steam was
177 fed at 150 °C with targeting injection rate. Typically, 1 kg of bed material was placed in the
178 gasifier. Beech-wood and PE pellets were co-fed from the fuel bankers with total feeding rate
179 at 400 g h⁻¹. Neon was constantly kept at 10 mL_N·min⁻¹ as balance gas in the co-gasification
180 process to calculate the gaseous products. The air and steam were fed to the bottom of the
181 gasifier and the equivalent ratio (ER) was kept constant for all cases by changing the flow
182 rate of air. The ER is defined as the ratio of the air or oxygen to the system divided by that
183 required for complete combustion [40]. The produced gaseous products from the
184 devolatilization of wood and PE were detected and quantified by different micro-gas
185 chromatography analysers, all of the detected gaseous products are H₂, Ar/O₂, CO, CO₂,
186 CH₂, C₂H₂, C₂H₄, C₂H₆, H₂S, C₆H₆ and C₇H₈. Tars were not analysed due to the difficulty of
187 collection and sampling. Carbon conversion was calculated by dividing the carbon in
188 produced gas to the total carbon inlet from feedstock.

189 This piece of work includes four investigations. Firstly, the effect of types of bed materials on
190 gaseous products from steam gasification of beech-wood only has been investigated
191 keeping the wood feed rate constant at 400 g h⁻¹, steam injection rate at 250 g h⁻¹, bed
192 temperature at ~850 °C. Then, the effect of bed materials on co-gasification of wood and PE
193 with the ratio at 1:1 has been investigated with a constant total fuel flow rate at 400 g h⁻¹,
194 bed temperature of 850 °C and there was no steam addition. To investigate the effect of
195 steam injection rate on the gaseous products from co-gasification of wood and PE, four bed

196 materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) have been investigated with
197 steam injection rate at 0, 400 and 800 g h⁻¹. The bed temperature was ~ 850 °C, total fuel
198 flow feeding rate was 400 g h⁻¹, the wood-to-PE ratio at 1:1. Finally, to investigate the effect
199 of wood-to-PE ratio three different compositions were investigated, namely 1:1, 3:1 and 4:0,
200 with all other variables unchanged, with steam injection rate at 400 g h⁻¹ and bed
201 temperature at ~850 °C.



202
203

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

205

206 3 Results and discussion

207 3.1 Investigation the effect of bed materials on beech-wood steam gasification

208 Table 2 shows the gaseous production from beech-wood gasification by using a fluidized-
209 bed reactor in the presence of silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite. The
210 gasification temperature was at ~850 °C with 250 g h⁻¹ steam injection rate. The data was
211 only collected when stable operation has been achieved. The cold gas efficiency (CGE) was
212 calculated as the ratio of energy production (net calorific value) of the gaseous products
213 divided by the total lower heating value of feedstock, biomass and PE at the corresponding
214 compositions. For comparison, theoretical predictions at thermodynamic equilibrium (1 bar,
215 850 °C) are also added in Table 2.

216

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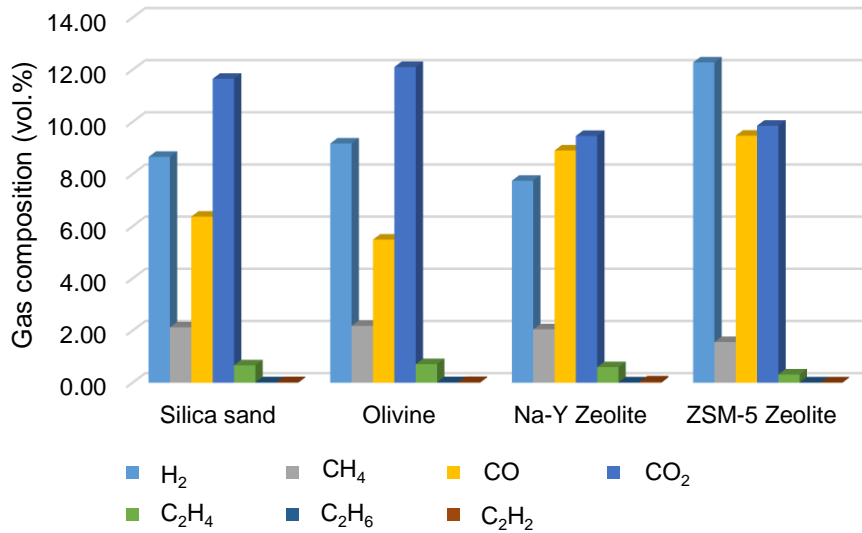
218 *Table 2 Gaseous production of beech-wood gasification in a fluidized-bed reactor with*
 219 *different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) at 850 °C and*
 220 *steam injection rate 250 g h⁻¹.*

Bed materials	Theoretical	Silica sand	Olivine	Na-Y Zeolite	ZSM-5 Zeolite
Feedstock 400gh⁻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
H₂	9.85	5.24	5.39	4.65	7.92
CH₄	0.31	1.25	1.30	1.23	1.00
CO	8.90	3.75	3.26	5.33	6.09
CO₂	7.07	6.87	7.19	5.67	6.36
C₂H₄	0.00	0.39	0.42	0.36	0.21
C₂H₆	0.00	0.01	0.015	0.009	0.008
C₂H₂	0.00	0.015	0.018	0.029	0.008
H₂S	0.008	0.0014	0.0008	0.0007	0.0000
C₆H₆	0.00	0.11	0.092	0.080	0.043
C₇H₈	0.00	0.007	0.028	0.014	0.0048
Total (N₂-H₂O 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

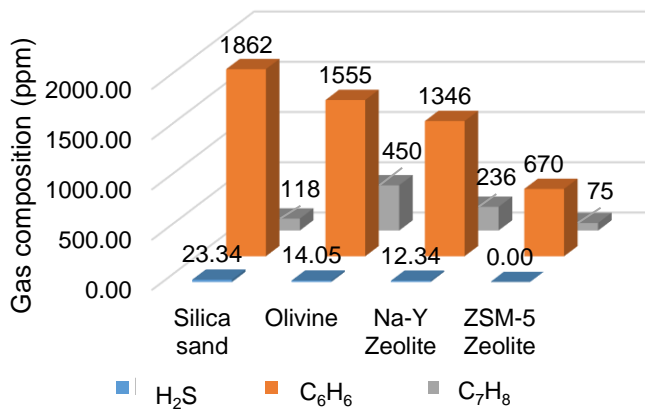
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222 The results shown in Table 2 indicate that the zeolite plays an important role in beech-wood
 223 gasification for hydrogen rich syngas production, with ZSM-5 zeolite given the highest
 224 hydrogen production and CGE which are 7.92 mol h⁻¹ and 83.57 %, respectively. These
 225 values are very close to those predicted by thermodynamic equilibrium, suggesting that tar
 226 production is indeed reduced when zeolites are used as bed materials. The carbon
 227 monoxide production reaches the highest amount of 6.09 mol h⁻¹ in correspondence with the
 228 highest carbon conversion. Figure 2 graphically summarised the results shown in Table 2
 229 and shows that ZSM-5 zeolite gave the highest production of hydrogen compared to the
 230 other bed material compositions. Notably, the relatively high CGE values in all cases are

231 indicative or high calorific value syngas production. This is also due to the presence of
 232 external heaters in the fluidised bed that allow lower ER to be used, while still maintaining
 233 temperature at above 800 °C.



234 (a)



235 (b)

236 *Figure 2 Gas compositions of beech-wood gasification in a fluidized-bed reactor with*
 237 *different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-5 zeolite) at 850 °C, steam*
 238 *injection rate 250 g h⁻¹ and ER at 0.3. (a)H₂, CH₄, CO, CO₂, C₂H₂, C₂H₆ and C₂H₄; (b) H₂S,*
 239 *C₆H₆ and C₇H₈.*

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

246 Na-Y zeolite promotes the CGE to 75.79 %, which is higher than silica sand and olivine. This
247 relates to the relatively high production of carbon monoxide at 5.33 mol h^{-1} . However, the
248 hydrogen production is only 4.65 mol h^{-1} , and carbon dioxide generation is also the lowest of
249 the various catalysts. The result could relate to the Na-Y zeolite promoting the carbon
250 gasification reaction in Equation (3), the reverse water gas shift reaction in Equation (4), the
251 forward direction of methane reforming reaction in Equation (5) and the Boudouard reaction
252 in Equation (6). This could be caused by the large pore structure of Y-zeolite compared to
253 ZSM-5 zeolite, such that the produced hydrocarbon gases pass through the supercages
254 without cracking. Gayubo et al. [42] showed that the ZSM-5 zeolite is more suitable for
255 cracking of hydrocarbons shorter than C_{12} which have higher thermal stability. It has been
256 reported that the products of biomass gasification at $>800 \text{ }^\circ\text{C}$ result in gaseous products with
257 lower molecular weight [43]. Olivine does not appear to play a relevant catalytic role in
258 beech-wood gasification as there is little difference in the hydrogen production and CGE
259 compared with silica sand. This result is consistent with previous research [44] , which has
260 shown the pore structure of olivine is damaged by high temperature sintering.

261

262 3.2 Effect of bed materials on co-gasification of beech-wood and polyethylene (PE)

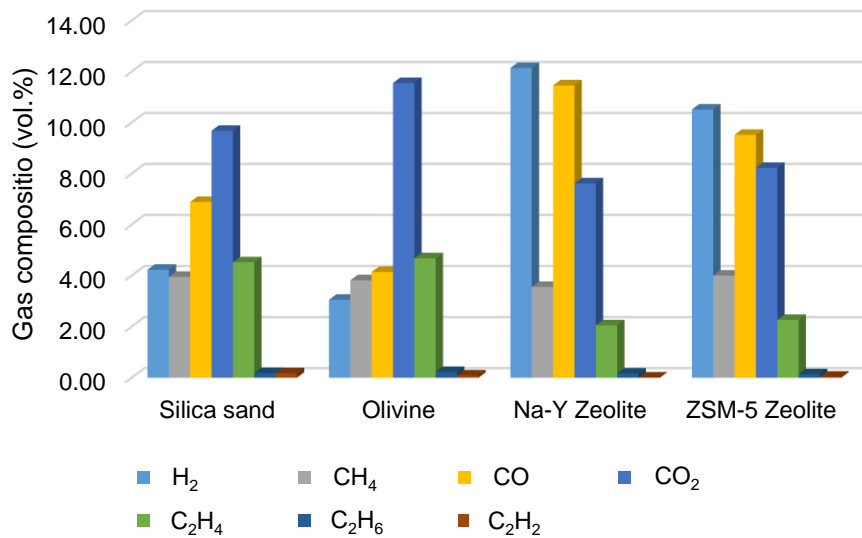
263 Table 3 shows the gaseous products from co-gasification of beech-wood and PE at $\sim 850 \text{ }^\circ\text{C}$
264 with no steam injection. The first observation with comparison with theoretical results at
265 equilibrium is that in all cases, much lower cold gas efficiencies were observed. This might
266 be due to the addition of plastics material, which is more prone to production of organic
267 species (including tars), which are not measured in these tests. This is also confirmed by the
268 lower carbon conversion values, which indicate that other carbon species were produced.
269 However, some generic conclusions with regards to catalysts activity could be drawn. For
270 example, the results show that Na-Y zeolite this time gave the highest hydrogen production
271 of 7.08 mol h^{-1} while ZSM-5 zeolite gave a relatively high amount of hydrogen production at
272 6.26 mol h^{-1} . This could be due to the effect of alkaline metal that crack volatile products
273 from biomass [45] and plastics [46, 47], in line with the results from other studies. In a review
274 of catalysts for biomass gasification, Bulushev et al. [45] showed that zeolite has been
275 widely applied for catalytic biomass or bio-oil upgrading due to its porous structure. Yumiko
276 et al. [46] reported that the isobutene (C_4) and isopentane (C_5) are selectively produced from
277 polyethylene with Na-Y zeolite catalyst in catalytic decomposition process. In the review of
278 Kunwar et al. [48], different types of zeolite for plastics cracking process were considered. In
279 agreement with the findings, Na-Y-zeolite was found to give the highest CGE at 69.08%
280 associated with the highest energy production is 8.13 MJ h^{-1} .

281 Figure 3 shows the gas compositions of beech-wood and PE co-gasification in a fluidized-
 282 bed reactor. There is a significant difference between the hydrogen production from zeolite
 283 and olivine, but no obvious difference between Na-Y zeolite and ZSM-5 zeolite. There is no
 284 catalytic effect of olivine in co-gasification of beech-wood and PE, the hydrogen production
 285 with olivine is even less than that for silica sand as shown in Table 3. Also, the carbon
 286 conversion has no significant difference. The results are consistent with those in Section 3.2
 287 confirming that the olivine has no catalytic activity in both of wood gasification and co-
 288 gasification process. Furthermore, both types of zeolite (Na-Y zeolite and ZSM-5 zeolite)
 289 show catalytic activity in co-gasification, while Na-Y zeolite is not active for the wood
 290 gasification process.

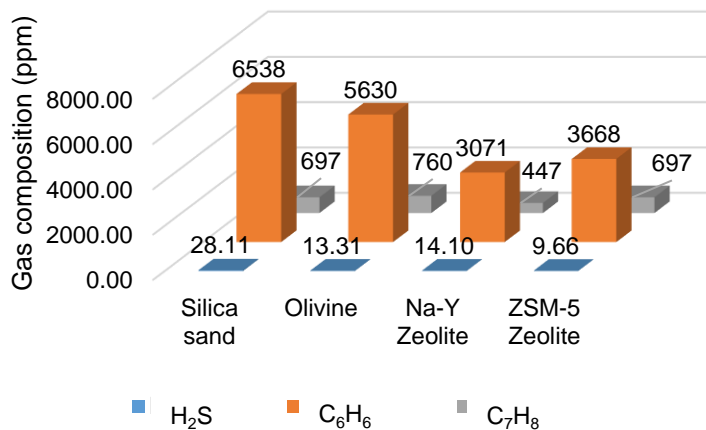
291 *Table 3 Gaseous production of beech-wood and polyethylene (PE) co-gasification in a*
 292 *fluidized-bed reactor with different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-*
 293 *5 zeolite) at 850 °C, no steam injection rate.*

Bed materials	Theoretical	Silica sand	Olivine	Na-Y Zeolite	ZSM-5 Zeolite
Feedstock 400 gh⁻¹ (wood-to-PE ratio)	1:1	1:1	1:1	1:1	1:1
Steam injection rate (gh⁻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 ₂ H ₄	0.00	2.38	2.47	1.37	1.35
C ₂ H ₆	0.00	0.096	0.12	0.10	0.081
C ₂ H ₂	0.00	0.095	0.067	0.0077	0.026
H ₂ S	0.004	0.0016	0.0007	0.0006	0.0006
C ₆ H ₆	0.00	0.35	0.30	0.20	0.22
C ₇ H ₈	0.00	0.036	0.040	0.030	0.042
Total (N₂-H₂O 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

294



295 (a)



296 (b)

297 *Figure 3 Gas composition of beech-wood and polyethylene (PE) co-gasification in a*
 298 *fluidized-bed reactor with different bed materials (silica sand, olivine, Na-Y zeolite and ZSM-*
 299 *5 zeolite) at 850 °C, no steam injection rate and ER at 0.27. (a)H₂, CH₄, CO, CO₂, C₂H₂,*
 300 *C₂H₆ and C₂H₂; (b) H₂S, C₆H₆ and C₇H₈.*

301

302 3.3 Effect of steam injection rate on co-gasification of beech-wood and polyethylene (PE)

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

306 *Table 4 Gaseous production from beech-wood and polyethylene (PE) co-gasification in a fluidized-bed reactor with different bed materials*
 307 *(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 and ER of 0.27.*

Bed materials	Theoretical		Silica sand		Olivine		Na-Y Zeolite		ZSM-5 Zeolite	
Feedstock 400gh⁻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)										
H₂	18.45	25.06	2.14	7.45	1.61	5.80	7.08	7.43	6.26	9.92
CH₄	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
CO₂	1.84	8.39	5.09	6.71	6.10	7.47	4.68	5.70	4.90	6.43
C₂H₄	0.00	0.00	2.38	1.55	2.47	2.25	1.37	1.63	1.35	1.30
C₂H₆	0.00	0.00	0.096	0.050	0.12	0.077	0.10	0.067	0.081	0.069
C₂H₂	0.00	0.00	0.095	0.073	0.067	0.067	0.0077	0.040	0.026	0.034
H₂S	0.004	0.004	0.0016	0.0035	0.0007	0.0017	0.0006	0.0024	0.0006	0.0010
C₆H₆	0.00	0.00	0.35	0.27	0.30	0.30	0.20	0.20	0.22	0.19
C₇H₈	0.00	0.00	0.036	0.012	0.04	0.049	0.030	0.030	0.042	0.034
Total (N₂-H₂O 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

308

309 As it can be appreciated from thermodynamic model predictions, the addition of the steam
310 boosts the hydrogen and carbon dioxide productions. The same behaviour is observed in
311 experimental results, regardless of the types of bed materials. This is mostly due to the
312 forward direction of waste-gas-shift reaction in Equation (4). However, the addition of steam
313 does not have a significant effect on the energy production and CGE except in the co-
314 gasification with ZSM-5 zeolite. The energy production from co-gasification of beech-wood
315 and PE with ZSM-5 zeolite increased from 7.78 to 8.74 MJ h⁻¹, when the steam feed is
316 increased from 0 to 400 g h⁻¹. This occurs simultaneously with the increase in CGE from
317 66.15 to 74.28 %. Na-Y zeolite has a negligible influence compared with ZSM-5 zeolite in
318 terms of hydrogen production and CGE. Na-Y zeolite has no obvious effect on gasification of
319 beech-wood or co-gasification, regardless of the steam injection rate and composition of
320 feedstock that is consistent with the results shown in Table 2 in Section 3.1.

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

330 In Table 4, we can also observe that H₂S production was generally increased with the
331 addition of steam in the co-gasification process. One reason could be the sulphur was
332 retained in solid residue, if any, within the bed. At contact with steam, this is released from
333 the solid state and measured as H₂S. Another reason could be explained by the involved
334 reaction between COS (or any other organic sulphur component) with steam and hydrogen
335 toward H₂S production. Although our experimental tests did not provide enough evidence of
336 this effect, similar results have been reported in other studies, with steam-to-carbon ratio
337 being one of the most important element to affect the H₂S production in steam gasification
338 of biomass [49, 50]. The increase of the steam-to-carbon ratio causes a decrease of COS,
339 and an increase of H₂S production, according to:



342

343 3.4 Effect of beech-wood to polyethylene (PE) ratio on co-gasification

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

349 Table 5 lists the gaseous products from co-gasification of beech-wood and PE (experimental
350 only). The results show that with the beech-wood as the dominant feedstock, energy
351 production is lower than when the feedstock composition contains more PE, while the
352 carbon conversion increases. For example, as beech-wood-to-PE ratio increases from 1:1 to
353 3:1 in presence of silica sand and Na-Y zeolite, the energy production reduces from 7.55 to
354 5.50 MJ h⁻¹ and 60.01 to 57.38 MJ h⁻¹, respectively. Carbon conversion increases from 88.1
355 to 95.7 %. The results are consistent with the significant difference in the Lower heating
356 value (LHV) of the feedstock as shown in Table 1, in that the LHV of PE is triple that of
357 beech-wood. Aznar et al. [24] reported similar results; the feedstock composition
358 significantly influences the flue gas from co-gasification of coal and plastics as they all have
359 different lower heating values. The result could also be explained by the more hydrogen
360 element involved in the co-gasification process as shown in with the
361 smaller beech-wood-to-PE ratio. All experiments were conducted with a constant feedstock
362 rate of 400 g h⁻¹. As shown in Table 1, the hydrogen composition of PE is 14.2 % and
363 hydrogen composition of beech-wood is much less than PE (5.9 %). When the total
364 feedstock rate is the same, the smaller beech-wood-to-PE ratio indicates more hydrogen
365 element input in the co-gasification process which explains the higher hydrogen content in
366 the products. Carbon conversion efficiency, however, is affected by the high volatile content
367 of plastics, which translates into higher tars content. Despite the catalytic effect of some bed
368 materials, it appears that the fraction of devolatilising feedstock which escapes the bed at
369 high temperature is still significant. Future work will highlight the effect of lowering the
370 location of the feed into the fluidised bed reactor to maximise contact time with catalytic
371 materials.

372 *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,
 373 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
 374 and ER of 0.27.

Bed materials	Silica sand		Olivine	Na-Y Zeolite		ZSM-5 Zeolite	
Feedstock 400g 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)							
H₂	7.45	6.06	5.80	7.43	4.77	9.92	8.13
CH₄	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
CO₂	6.71	7.69	7.47	5.70	6.22	6.43	6.55
C₂H₄	1.55	0.92	2.25	1.63	1.07	1.30	0.23
C₂H₆	0.05	0.023	0.077	0.067	0.036	0.069	0.0079
C₂H₂	0.073	0.037	0.067	0.040	0.050	0.034	0.013
H₂S	0.0035	0.0024	0.0017	0.0024	0.001	0.0010	0.0001
C₆H₆	0.27	0.17	0.30	0.20	0.13	0.19	0.047
C₇H₈	0.012	0.0070	0.049	0.030	0.021	0.034	0.0054
Total (N₂-H₂O_{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

375

376 4 Conclusions

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

- 380 • ZSM-5 zeolite plays a significant role in the gasification of beech-wood and gives the
381 highest hydrogen production and energy conversion, which are 7.92 mol h^{-1} and
382 83.57 %, respectively.
- 383 • Both ZSM-5 zeolite and Na-Y zeolite have a significant influence on co-gasification of
384 beech-wood and polyethylene (PE) in terms of high hydrogen production and CGE.
385 Furthermore, Na-Y zeolite has a greater influence compared to ZSM-5 zeolite. The
386 hydrogen produced in the presence of Na-Y zeolite is 0.82 mol h^{-1} , which is higher
387 than the hydrogen produced in the presence of ZSM-5 zeolite. The CGE for the co-
388 gasification of beech-wood and PE in the presence of Na-Y zeolite is ~3% higher
389 than in the presence of ZSM-5 zeolite.
- 390 • The addition of steam in the co-gasification of beech-wood and PE promotes
391 hydrogen production but has negligible influence on the CGE, except in the presence
392 of ZSM-5 zeolite.
- 393 • The composition of feedstock has a strong influence on hydrogen content from the
394 co-gasification process, with the smaller beech-wood-to-PE ratio giving higher
395 hydrogen production. The smaller beech-wood-to-PE ratio also gives higher energy
396 production because the Lower heating value (LHV) of PE is almost triple that of
397 beech-wood. However, carbon conversion efficiency is negatively affected by the
398 presence of plastics.

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