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Bio-DME production based on conventional and CO₂-enhanced gasification of biomass: A comparative study on exergy and environmental impacts

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Abstract

In this study, a novel single-step synthesis of dimethyl ether (DME) based on CO₂-enhanced biomass gasification was proposed and simulated using ASPEN PlusTM modelling. The exergetic and environmental evaluation was performed in comparison with a conventional system. It was found that the fuel energy efficiency, plant energy efficiency and plant exergetic efficiency of the CO₂-enhanced system were better than those of the conventional system. The novel process produced 0.59 kg of DME per kg of gumwood with an overall plant energy efficiency of 65%, which were 28% and 5% higher than those of conventional systems, respectively. The overall exergetic efficiency of the CO₂-enhanced system was also 7% higher. Exergetic analysis of each individual process unit in both the CO₂-enhanced system and conventional systems showed that the largest loss occurred at gasification unit. However, the use of CO₂ as gasifying agent resulted in a reduced loss at gasifier by 15%, indicating another advantage of the proposed system. In addition, the LCA analysis showed that the use of CO₂ as gasifying agent could also result in less environmental impacts compared with conventional systems, which subsequently made the CO₂-enhanced system a promising option for a more environmental friendly synthesis of bio-DME.

Keywords: Exergy analysis; Environmental analysis; Bio-DME; CO₂-enhanced gasification; conventional gasification

26 **1 Introduction**

27 Biomass derived dimethyl ether (Bio-DME) is a clean synthetic fuel that has high cetane number
28 and similar physical properties as LPG [1, 2]. The combustion of bio-DME generates small amount
29 of NO_x, almost zero SO_x and particulate matter. Thus, bio-DME is considered as a sustainable
30 alternative to diesel and LPG. Compared with commercially available double-step synthesis, the
31 single-step synthesis is a better option for DME production due to its low investment and low
32 production costs [3, 4]. Among single-step synthesis technologies developed, JFE technology, a
33 process adopting H₂: CO ratio of 1:1 for DME synthesis, was found to be more cost-effective than
34 other technologies, such as Hardlor Topsoe technology [1, 5].

35 Over the past two decades, the use of CO₂ as a gasifying agent in biomass gasification has drawn
36 increasing interests [6-8]. One of its unique features in this technology is that the H₂/CO ratio in
37 syngas can be adjusted by controlling the amount of CO₂ injected to gasifier, despite the additional
38 heat need to be supplied into gasifier due to the endothermic nature of reaction involved during
39 gasification [9-11]. Recent research demonstrated that for DME production based on CO₂-enhanced
40 gasification, the water gas shift (WGS) reactor and the energy intensive CO₂ removal process could
41 be avoided while the production of DME could be enhanced [12]. In addition, the pure CO₂ can be
42 easily recycled as CO₂ is a major by-product of DME synthesis. However, not much effort has yet
43 been made to understand the exergy efficiency and environmental impacts of this novel system.

44 Generally, exergy analysis specifies the location, type and magnitude of process irreversibility [13-
45 15]. It also helps better understand the benefits of energy utilization by providing more useful and
46 meaningful information than what energy analysis could possible provide. Therefore, exergy
47 analysis is commonly used to compare the performance of different processes, such as biomass-
48 gasification and coal-gasification based processes [16-21]. With regard to exergy analysis of bio-
49 DME production, to date, only a few studies have been reported [22, 23]. Exergy analysis of
50 double-step bio-DME production using steam as gasification agent was carried out by Zhang et al.

51 [23] to measure the exergy efficiency of the entire system as well as the exergy losses occurred in
52 each unit of the system. The reasons behind these losses were explored and suggestions to improve
53 the exergy efficiency of the system were made. Recently, Xiang et al. [22] conducted the exergetic
54 evaluation of a single-step bio-DME production from biomass where pure DME was obtained as
55 the final product. The estimation of system exergetic efficiency and the origin of losses were also
56 identified and quantified. Moreover, the causes of the inefficiencies were investigated and by
57 optimising various process parameters, the higher exergetic efficiency of the system was achieved.
58 However, in spite of the great potential of DME production based on CO₂-enhanced gasification, in
59 depth assessment of this new route has hardly been explored due to the lack of detailed process
60 design. To the best of our knowledge, there is not any published research on the exergetic
61 assessment of bio-DME production using CO₂ as the gasifying agent. In addition, although life
62 cycle assessment (LCA) is commonly used to evaluate environmental impacts of a product over its
63 life cycle [16, 24, 25], the application of LCA approach for the environmental assessment of bio-
64 DME production based on CO₂-enhanced gasification has not been reported.

65 This study was focused on the simulation of conventional DME synthesis system as well as DME
66 production based on CO₂-enhanced gasification of biomass. The goal of this study was to assess the
67 exergetic and environmental performance of bio-DME production using CO₂ as a gasifying agent.
68 The comparison of energy, exergetic and environmental analyses between the two processes were
69 also carried out. In addition, effort was made to show the location, magnitude and causes of the
70 process unit inefficiencies.

71 **2 Process description and design**

72 **2.1 Process overview**

73 Fig. 1 shows the simplified process flow diagrams of the single-step DME production based on
74 conventional and CO₂-enhanced biomass gasification systems. The configurations have been

75 optimized following the guidelines proposed elsewhere [1, 22, 23, 26, 27]. In these systems,
76 biomass feedstock is converted to syngas in a fluidised-bed gasifier, which is then conditioned prior
77 to DME synthesis. The present work was focused on the assessment of the influence of CO₂
78 addition on the overall system performance.

79 The gasifiers for the two processes are operated under relatively different conditions. As seen in
80 Fig. 1, the conventional process uses oxygen and steam as the gasifying agent while the CO₂-
81 enhanced process utilises carbon dioxide with steam. For the CO₂-enhanced biomass gasification
82 based DME production (CEBG-DME) process, since H₂/CO ratio can be tuned by selecting proper
83 CO₂/biomass ratio and steam/biomass ratio (as shown in Fig. 1b), the WGS and energy intensive
84 CO₂ separation unit are eliminated. Another beneficial feature of this process is the avoidance of
85 using of the oxygen separation unit, which could cause significant energy consumption as well as
86 high capital and operating costs. As CO₂ is one of the main products of the single-step DME
87 synthesis, in the CEBG-DME system, a portion of the emitted CO₂ is used in the gasifier as the
88 gasifying agent, which helps reduce net CO₂ emission of the system. However, due to the
89 endothermic nature of gasification reactions, additional energy is required to maintain a desired
90 temperature in the gasifier. Since fluidized bed gasifier is used in this study, it was proposed that
91 heat was introduced into the gasifier using an inert energy carrier. However, during simulation,
92 electrically-heated gasifier was considered in the current study.

93

94 **2.2 Biomass to syngas train**

95 In this study, gumwood was selected as the biomass feedstock because it is widely available in
96 China as well as in South-East Asian countries. Its properties are listed in Table 1 [28].

97 During gasification, the C, H, and O are transformed to CO, H₂, CO₂, and CH₄, while N and S are
98 converted to NH₃, H₂S and COS, respectively. Since particulates (such as fly ash) that can
99 potentially foul and/or poison the catalyst in the downstream, prior to DME synthesis, particulate

100 matter is removed using a bag filter. A heat exchanger is used in the HRSG unit to recover waste
101 heat to produce steam that is used in gasification unit as well as other process units. Part of the
102 recovered heat is used for electricity generation, which is to meet electricity demand of the entire
103 plant.

104 In conventional DME synthesis process, the gas product (syngas) is then transported to a WGS unit
105 to adjust the H₂/CO molar ratio of 1 to satisfy the requirement for DME synthesis, followed by the
106 purification prior to synthesis because H₂S is poisonous to the synthesis catalyst [3, 29] and an
107 excessive amount of CO₂ will reduce the conversion efficiency of DME synthesis [2, 12]. In the
108 purification section, a typical chemical absorption process with monoethanolamine (MEA) is
109 selected to remove H₂S and CO₂, which is detailed elsewhere [30, 31].

110 However, for CO₂-enhanced gasification based DME synthesis (Fig. 1b), the required H₂/CO molar
111 ratio and CO₂ concentration in syngas is attained by adjusting gasification operating parameters
112 such as temperature, CO₂/biomass ratio and steam/biomass ratio. Consequently, the WGS reaction
113 unit and energy intensive CO₂ purification unit, the two essential steps for conventional DME
114 production process (Fig. 1a), are avoided. It is important to note that with the addition of steam in
115 the gasifier, WGS unit in conventional system can be avoided. However, from the perspective of an
116 overall plant economics, this approach is not likely to be feasible, as many factors such as type of
117 gasifier need to be considered. In order to prevent catalyst from being poisoned, only H₂S removal
118 unit is installed.

119 **2.3 Syngas to DME train**

120 The purified syngas is then fed into the compressor and cooler, and subsequently introduced to the
121 single-step synthesis reactor. As the single-step DME reaction is exothermic, to maintain an
122 optimal reaction temperature, certain amount of heat must be removed rapidly from the reaction
123 system, which can be utilized to generate high-pressure steam. Hence, a slurry reactor was

124 considered in this study due to its easiness in temperature control and uniform temperature
125 distribution [1, 22]. The product stream from the synthesis reactor is cooled down and flashed so
126 that the unreacted syngas is separated from the DME-methanol-water mixture. Afterwards, the
127 majority of the unreacted syngas from the flash separator is recycled to DME synthesis reactor.
128 This leads to an overall high DME conversion, while the rest of the stream was purged. The DME–
129 methanol–water mixture contains some residual gas such as CO and CO₂, hence, it is sent to CO₂
130 removal unit to remove the residual gas. It is essential to note that a fraction of emitted CO₂ by the
131 synthesis reactor is fed to the gasifier as it is used as one of the gasifying agents for the CO₂-
132 enhanced biomass gasification. In the meantime, bottom product of the CO₂ removal unit is fed to
133 the DME distillation unit. Distillate from this unit is taken as DME product. The methanol-water
134 mixture, the bottom product, is sent to methanol distillation unit to separate water from methanol.
135 The recovered methanol is also recycled to the DME reactor, whereas bottom product water, which
136 still contained a very low amount of methanol, requires further purification. A base-case design of
137 this study is illustrated in Fig. 1, which is for the preliminary assessment of a process design and
138 has not yet been optimized at this stage.

139 **2.4 Key gasification parameters**

140 Apart from operating pressure, temperature and oxidising agent are the two important factors that
141 dictate the composition of syngas in any gasification processes. Whilst for syngas, H₂/CO ratio and
142 the percentage of CO₂ are the two crucial parameters that have significant impacts on its
143 purification and application. There are other factors commonly used to evaluate gasification
144 process, such as lower heating value (LHV) of gas product, cold gas efficiency (CGE), and
145 gasification system efficiency (GSE) [9, 10]. Moreover, for JFE single-step DME synthesis, the
146 H₂/CO ratio is also vital, which should be controlled to be 1.

147 In order to design a process for the highly efficient synthesis of DME, it is important to understand
148 the relationship of these parameters. In this study, the parametric analysis was therefore conducted

149 to optimize the gasification process. The parameters for the gasification process used in this work
150 are listed in Table 2, in comparison with those for a conventional (O₂-steam) process. In addition,
151 for comparison purposes, Table 2 also includes a set of gasification data that were reported by
152 others under similar conditions [1].

153 **3 Methodology**

154 **3.1 Process simulation**

155 Simulation of the two DME systems was conducted using Aspen PlusTM to establish materials
156 balance, and to estimate the energy and utility requirements as the inputs for exergy and
157 environmental analysis. Details of assumptions and modelling procedures have been discussed
158 elsewhere [9, 12].

159 In this study, a RYield module (yield reactor) integrated with a RGibbs module (chemical and
160 phase equilibrium by Gibbs free energy minimisation) was employed to simulate gasifier, which
161 was operated (0.1 MPa). Thus, heat inputs to the gasifier included heat duty of RYield and RGibbs
162 reactors. The operating conditions of the gasifier for both systems are presented in Table 2.

163 **3.2 Model validation**

164 Before thermodynamic evaluation of the studied systems, the CO₂-enhanced gasification model has
165 to be validated. In this study, the model developed was validated against data published by other
166 researchers [10, 21, 32]. As mentioned previously, biomass gasification was modelled using Gibbs
167 minimisation approach, which has been validated with experimental data [33-35]. Since there is no
168 relevant data available on CO₂-enhanced gasification using gumwood as the raw material, in order
169 to validate the model developed in this work, data of steam gasification of wood and CO₂
170 gasification of biomass were extracted from literature and compared with the simulation results
171 (under the same experimental conditions as adopted in the literature) as shown in Table 3 and Table

172 4, respectively [10, 21]. Based on these comparisons, it can be concluded that the simulation results
173 are in good agreement with data reported in literature [10, 21, 32]. The deviation was found to be in
174 a range of 4.1%-8.6%. Therefore, the CO₂-enhanced gasification model developed in this study is
175 reliable, and was subsequently used for the prediction of CO₂-enhanced gasification of gumwood.

176 The product gas from the gasifier was sent to the SSplit (SSEPARAT) module to remove ash, prior
177 to cooling down to 220 °C. HeatX (HRSG) module was used to recover heat (via steam at 0.5 MPa,
178 225 °C) from hot syngas. Afterwards, syngas was sent to WGS reactor to adjust its H₂/CO ratio.
179 The WGS reactor (0.1 MPa, 220 °C) was modelled with a REquil module. After WGS reactor, the
180 syngas was cooled down to 60 °C before being directed to syngas purification unit. The removal of
181 CO₂ and H₂S by MEA absorption process in syngas purification unit was modelled as a simple
182 component separator SEP2 module due to the complexity of the system. The energy required for
183 this system was assumed to be 3 MJ/kg of CO₂ captured by MEA absorption process [10]. The
184 single-step DME synthesis was operated at 6 MPa and 250 °C. The purified syngas was then
185 pressurised, cooled down, and introduced into the DME reactor. A REquil module was employed
186 as the DME reactor to simulate the synthesis process, which is reliable in DME synthesis
187 simulation [36, 37]. The other operating parameters and DME synthesis reactions simulated in the
188 present work were adopted from literatures [36-39].

189 Product of the DME reactor was cooled down to -30 °C in MHeatX (X) module and consequently,
190 non-condensable gases were removed using Flash2 (FLASH) separator module. In order to recycle
191 the non-condensable gaseous, most of the gas was sent back to DME reactor and the remainder was
192 purged. After the flash separator, the liquid stream was then sent to the product purification
193 process. This section is consisted of three units, i.e., CO₂ removal unit (2 MPa), DME distillation
194 unit (1.52 MPa) and MeOH unit (1.52 MPa) as illustrated in Fig 1. Distillation column was
195 employed for each of the unit, which was simulated using RadFrac module. The distillation
196 parameters, such as the number of stages and reflux ratio, were set to achieve high purity (99.9

197 mol%) of DME product. The reaction heat from synthesis reactor was recovered using Heat Stream
198 option in Aspen PlusTM to generate high-pressure steam at 3.5 MPa and 244 °C.

199 3.3 Exergy analysis

200 From a thermodynamic point of view, exergy is defined as the maximum amount of work that can
201 be generated by an energy system as it comes to equilibrium with the reference of environment [15,
202 17]. It measures both the quality and quantity of energy involved in the conversion within a system.
203 Therefore, it enables the detection of losses and identifies the opportunities for the improvement of
204 energy consumption. The objective of exergy analysis is to identify process units with relatively
205 high inefficiency. In addition, exergy analysis can also be used to compare different process
206 configurations to find better options.

207 Generally, the exergy balance of individual process unit within energy transformation system can
208 be expressed as:

$$209 \quad \sum \dot{E}ex_{in} = \sum \dot{E}ex_{out} + \dot{E}ex_{loss} + \dot{E}ex_{des} \quad (1)$$

210 where, $\sum \dot{E}ex_{in}$ and $\sum \dot{E}ex_{out}$ denote the sums of exergy rates for inlet flows and outlet flows,
211 respectively, including the materials streams, thermal stream and work. The $\dot{E}ex_{loss}$ stands for the
212 exergy loss rate of the streams that exit the system without further use. Meanwhile, $\dot{E}ex_{des}$ depicts
213 the exergy destruction rate, which represents the inefficiencies existed in the system. For simplicity,
214 these two parts were merged in a term called exergy losses and destructions, $\dot{E}ex_{L\&D}$, which is
215 evaluated as follows:

$$216 \quad \dot{E}ex_{L\&D} = \sum \dot{E}ex_{in} - \sum \dot{E}ex_{out} \quad (2)$$

217 In the present work, exergy of streams, such as biomass, gases, liquid and heat, was calculated
218 using the concept, which has been explained elsewhere [9, 17, 40, 41]. The considered systems
219 were decomposed into different functional process units. For each unit, exergy balance was
220 performed and $\dot{E}x_{L\&D}$ was calculated.

221 **3.4 Energy and exergy efficiencies**

222 The fuel energy efficiency is the fraction of energy stored in the biomass that is converted into
223 energy of the fuel product, as given in Eq. (3) [27, 42].

$$224 \quad \eta_{Fuel} = \frac{E_{fuel,out}}{E_{biomass,in}} \quad (3)$$

225 where, $E_{fuel, out}$ is the energy in the fuel produced and $E_{biomass,in}$ is the energy in the feedstock.

226 As Eq. (3) does not include all inputs and useful outputs of the system, the plant energy efficiency
227 was also used to evaluate performance of the entire system as shown in Eq. (4).

$$228 \quad \eta_{Plant} = \frac{E_{fuel,out} + Q_{net,out}}{E_{biomass,in} + E_{agent,in} + E_{Q,in} + E_{W,in}} \quad (4)$$

229 where, $E_{agent,in}$, $E_{Q,in}$, and $E_{W,in}$ are the total energy input as gasification agent, thermal energy and the
230 net work in plant, respectively, whereas $Q_{net,out}$ represents the total heat output as district heat level.

231 Gasification process efficiency was evaluated using cold gas efficiency (CGE) and gasification
232 system efficiency (GSE), which are described elsewhere [9].

233 The system exergy efficiency (η_{ex}) was calculated using Eq. (5) (derived from Eqs. 1 & 4) as the
234 useful exergy content in outlet flows divided by the exergy content in inlet flows.

235
$$\eta_{ex} = \frac{Eex_{out}}{Eex_{in}} \quad (5)$$

236 **3.5 Environmental analysis**

237 The goal of environmental analysis was to assess the environmental impacts of CO₂ utilisation in
238 bio-DME production. Thus, investigation was conducted to compare the two scenarios:
239 conventional bio-DME production (scenario 1) and CO₂-enhanced bio-DME production (scenario
240 2). Energy and exergetic assessment of these two scenarios indicated that using CO₂ as gasification
241 agent can significantly improve the process performance. Moreover, CO₂ was consumed in CO₂-
242 enhanced process. This meant that, the net CO₂ emission in CO₂-enhanced system was lower than
243 that of conventional. The environmental impact assessment was performed using the ReCiPe 2008
244 v.3.14 method from SimaPro 8.0 software package. ReCiPe 2008 encompasses two sets of impact
245 categories (mid-point level and end-point level) associated with two sets of categorization factors.
246 Detail explanation of impact categories and qualitative indicator can be found elsewhere [9, 43-45].
247 In the present work, approximately 1 kg of DME produced from conventional and CO₂-enhanced
248 biomass gasification was used as the functional unit for environmental analysis. LCA of individual
249 input and output streams of the overall system, including full life cycle of components, was
250 conducted. The input data (*i.e.*, consumption of feedstock, agents and energy as well as emissions)
251 of this analysis are obtained from Aspen PlusTM simulation results. The system boundaries set for
252 this study are illustrated in Fig. 1. The scope of this study includes the following aspects: (1) supply
253 of biomass, gasification agents and other utilities to the DME production system; (2) production of
254 DME via biomass gasification process; (3) heat recovery from the system; and (4) utilisation of
255 emitted CO₂. Both CO₂ and CH₄ were considered as the main GHG (greenhouse gases) for the
256 assessment of environmental impact.

257 **4 Results and Discussion**

258 In this study, the energetic comparison between conventional and CO₂-enhanced bio-DME
259 production, the biomass consumption per kilogram of DME production, exergy balance of the
260 entire plant and exergy losses and destructions to each unit were carried out to disclose the energy
261 saving mechanism. Finally, the environmental impacts caused by these two routes were assessed
262 using LCA method.

263 **4.1 Mass and energy balances**

264 Table 5 presents the overall mass and energy balances for the two DME production routes, i.e.,
265 conventional and CO₂-enhanced processes. The energy balances indicated that biomass feedstock
266 required most of the energy input (37-46%) in the system. It is noted that biomass input with
267 respect to its mass and energy content and the quality of product DME were equalized in both
268 cases; thus, results could be compared. It can be seen from Table 5 that CO₂-enhanced system
269 produced 0.59 kg DME per kg gumwood with a fuel conversion efficiency (detailed in Eq. 3) of
270 85.0%, which is 18% higher than the conventional process. This was because of higher amount of
271 syngas processed in DME synthesis reactor as a results of using CO₂ in the gasifier which reduced
272 the molar ratio of H₂/CO by increasing CO fraction in syngas [9]. It was also found that the
273 addition of CO₂ reduced the percentage of CH₄ in syngas. Hence, this resulted to a lower amount of
274 purge gas from the synthesis reactor in the CO₂-enhanced system as DME conversion efficiency is
275 generally limited by high CH₄ percentage in syngas [37]. The aforementioned phenomena could
276 also result in a higher DME production rate. An overview of DME system parameters of the
277 present work and published works is given in Table 6, particularly in terms of comparing the mass
278 yield and plant energy efficiency of the systems. Clearly, CO₂-enhanced system produced higher
279 amount of DME, which led to the greater plant energy efficiency. The present work is
280 fundamentally differed from the reported work as it uses CO₂ as the gasifying agent. The CO₂
281 addition could lead to the increase in CO fraction in syngas which influenced the H₂/CO ratio and

282 the yield of syngas [9]. For instance, H₂/CO ratio of 1 was achieved in the CO₂-enhanced system
283 while the value was around 0.85 in conventional systems. Moreover, the yield of syngas was also
284 increased by 20% in the proposed system. In addition to the improved CO₂-enhanced system
285 proposed in this work, the studied conventional system has also exhibited a better performance than
286 the work conducted by previous researchers, as shown in Table 6. The main difference between this
287 study and reported work [22] was in terms of DME synthesis reactor: equilibrium reactor was used
288 in this study whilst RStoic reactor was used in the reported work [22]. An important feature of
289 equilibrium reactor is that the DME synthesis reactor was assumed to have chemical equilibrium
290 property which resulted in 84% conversion of CO while the RStoic reactor was modelled by
291 assuming only 64% conversion of CO. Consequently, flow rate of the recycled stream in the
292 present work was lower which greatly increased the yield of product DME in the synthesis process.
293 Another potential reason for the high value of yield was that the treated gumwood was considered
294 as a feedstock in the current work, whereas raw sawdust was used in Ref [24]. Similar study was
295 also conducted by others [1] with a mass yield around 0.37. This value was 15% larger than that in
296 [22] but 24% lower compared to the conventional system investigated in the current work. In
297 contrast, a double-step synthesis route was selected by Zhang et al.,[23] and one-pass conversion of
298 methanol to DME was about 70-85%. Accordingly, the conventional system obtained the highest
299 DME yield. Hence, in the present work, the improvement of DME yield in CO₂-enhanced systems
300 was mainly contributed by the utilisation of CO₂ in gasifier, which was not considered in the
301 conventional system, as well as by the employment of equilibrium reactor in synthesis process. As
302 also shown in Table 6, a similar trend was noticed when plant energy efficiencies were compared
303 where the highest efficiency was obtained by the CO₂-enhanced system. Compared to that of
304 conventional system, however, the increase in plant energy efficiency in the CO₂-enhanced system
305 was not as obvious as what was the case in terms of fuel energy efficiency. The reason for such is
306 further explained in the followings.

307 As shown in Table 5, CO₂-enhanced DME system had a higher plant energy efficiency (detailed in
308 Eq. 4) of 65.97%, which was about 5% higher than that of conventional systems. This was mostly
309 caused by the CO₂-enhanced system having a larger amount of both DME production and heat
310 output. The higher heat output was attributed to two sources: more syngas was produced in the
311 gasifier and the high reaction heat in DME reactor. It can be seen that the demand of total input
312 energy for gasifying agent and net heat generation in CO₂-enhanced system were significantly
313 higher than those of the conventional system. Additionally, the amount of heat required by the
314 gasifier in CO₂-enhanced process was 901,969 MJ/h, which was around 45% larger than that of
315 conventional process. However, this trade-off was worthwhile as the proposed system offered a
316 more significant increment of the plant outputs, which in this case, were DME production and heat
317 output. Besides, CO₂-enhanced system avoided water-gas shift and acid gas cleaning processes,
318 indicating lower capital costs. Thus, the addition of CO₂ was proven to be beneficial in terms of
319 feed consumption, fuel production and economical aspects. Another important feature of the
320 proposed system was that it consumed CO₂, thus, provided considerable environmental advantages.

321 There are other parameters that might influence the system efficiencies, including the properties of
322 feedstock, system configurations and operating conditions. However, the analysis presented above
323 provides first-hand information about DME yield per kg of biomass via CO₂-enhanced system.

324 **4.2 Exergy analysis of DME production processes**

325 Exergetic efficiency of the DME production process can be calculated based on the evaluation of
326 performance of the entire plant. Moreover, by analysing exergy flows within the plant, efficiencies
327 of individual process units and their significance to the overall plant performance can be estimated.
328 Table 7 presents the exergy balance of the two bio-DME routes compared in this study. In both
329 processes, biomass contributed to the major exergy flow (57-67%) followed by exergy content of
330 heat flows to the system. With respect to plant outputs, DME represented the highest contributor

331 (83-85%) whereas the additional output such as exergy of steam generated throughout the system
332 was the second highest.

333 Table 7 also shows a comparison of exergetic efficiency of the bio-DME process between the two
334 routes. Exergetic efficiency of the conventional plant was found to be 50.8% which was higher than
335 those reported by others [22, 23]. Meanwhile, 85% of the total exergy output was contributed by
336 DME. An important aspect of the current conventional system is that it generates more DME per
337 unit mass of biomass than those of the previous work [22, 23], which increased the net production
338 rate. This basically contributed to the higher exergetic efficiency of the current system.
339 Furthermore, various variables including process operating conditions and biomass properties, such
340 as moisture content, usually affect plant exergetic efficiency. Previous research stated that chemical
341 exergy of biomass decreased with the increase of its moisture content [46]. The biomass used in
342 this work contained 2.1 wt% moisture, lower than the values used in reported works which were
343 13.4 wt% [22] and 7.5 wt% [23]. The aforementioned characteristic also contributed to a higher
344 exergetic efficiency of the present work. The exergetic efficiency of the calculated system was 10%
345 lower compared with its energetic efficiency. This deviation was normal as biomass underwent
346 gasification process at a high temperature, which typically had a higher inefficiency associated with
347 the related chemical reactions [17]. The difference arisen was caused by: (i) energetic performance
348 which only considers energy loss due to emissions to the environment; (ii) exergetic assessment,
349 considers both the external exergy losses (caused by system emissions) and the destruction of
350 internal exergy based on the second law of thermodynamics. Hence, unlike energetic analysis,
351 exergetic analysis is useful in identifying the causes, locations and magnitudes of process
352 inefficiencies in order to improve the performance of the entire system [17, 21, 47]. On the other
353 hand, exergetic efficiency of the CO₂-enhanced system was 57.3%, almost 7% higher than that of
354 conventional one. Out of the 100% useful outputs, DME contributed as much as 83.3% while the
355 rest was the recovered heat from the system. It is worth noting that plant exergetic efficiency and
356 energetic efficiency of the CO₂-enhanced system were higher compared with those of conventional

357 systems, which was previously discussed in Section 4.1. It is found that process efficiency was
358 highly influenced by DME yield, which was also in relation with the exergy content of 30.85 MJ/kg
359 and the heating value (LHV) of 28.40 MJ/kg for the respective exergetic and energetic evaluations.
360 In this case, exergy content and heating value had less influence on the efficiencies since the
361 difference between them was relatively small.

362 Fig. 2 presents the relative exergy losses and destructions (L&D) for each process unit, which was
363 previously illustrated in Fig. 1.

364 In both routes, the major exergy losses and destructions were associated with gasifier, methanol
365 tower, CO₂ removal and DME reactor units (38-45%, 7-30%, 3-8% and 4-5%, respectively). These
366 units are therefore endowed with potential for system improvement. On the other hand, the exergy
367 L&Ds of other units such as in HRSG, WGS, Cleaner and DME Tower were much smaller. It is
368 clear from Fig. 2 that gasifier was the critical unit of the system because of it is of the largest value
369 of exergy losses and destructions. Hence, further analysis of this unit is worthwhile in improving
370 the overall exergetic performance of the system. Generally, the exergy losses in the biomass
371 gasification are highly dependent on the heat duty required to achieve chemical equilibrium at the
372 given gasification temperature [21]. It is interesting to note that gasifier in conventional process
373 consumed around 55% (619,056 MJ/h) of the total electricity requirement while the value for
374 gasifier used in CO₂-enhanced process was around 61% (901,969 MJ/h). In the gasifier, the
375 decomposition of large molecules into smaller ones at higher temperatures causes large damage of
376 chemical exergy. Previous studies showed that lower temperatures and higher pressures were
377 beneficial for exergetic performance of the gasification process [19, 21].

378 Despite the adjustment of operating parameters and/or the use of different types of gasifier could
379 increase the performance of gasifier, it cannot significantly reduce the total losses due to the
380 existence of intrinsic energy and material degradation within gasification process. In addition,
381 adjusting the composition of syngas to improve the process economy also limits the range of

382 operating parameters of the fluidized bed gasifier. It is important to highlight that the main
383 characteristic of CO₂-enhanced system is the use of CO₂ recycled from DME distillation unit as
384 gasification agent. Therefore, the amount of CO₂ or the CO₂/biomass ratio is crucial to plant
385 performance, which needs to be optimized. Accordingly, CO₂/biomass ratio was adjusted in the
386 present work. However, as the amount of CO₂ addition directly affects CO₂ percentage in syngas
387 and there is a limitation of CO₂% in syngas (around 3% maximum), the potential of adjusting
388 CO₂/biomass ratio for exergy efficiency improvement is small, which could only lead to a small
389 increment (1.2%) of exergy efficiency. Since the CO₂/biomass ratio in gasifier (Table 2) has
390 already been optimized, no significant impact was observed.

391 Similarly, marginal improvement of process efficiency might be achieved by tuning the
392 temperature and pressure of gasifier, and also the temperature of the reacting streams. Thus, the
393 increment of gasification efficiency via controlling the operating parameters is very limited.
394 Upgrading the biomass feedstock via torrefaction could improve the performance but there is a
395 compromise for extra energy required in the process that contributes to further exergy, energy and
396 environmental losses. Other types of irreversibilities, such as fluid dynamic losses and heat losses,
397 might also lead to a further degradation of the energy.

398 It can be seen from Fig. 2 that the exergy L&D in gasifier of the CO₂-enhanced system was lower
399 (by about 15%) compared to that of the conventional system although additional heat exergy and
400 gasification agent exergy were required. This phenomenon can be explained by the properties of
401 gas generated in the gasifier. The amount of H₂ and CO in the product gas in CO₂-enhanced route
402 was higher (44% and 21%, respectively) while the amount of CO₂ and H₂O was lower (47% and
403 28%, respectively) than those of the conventional one. The chemical exergy values of H₂ and CO
404 are extremely higher (236100 kJ/kmol and 275100 kJ/kmol, respectively) than those of CO₂ and
405 H₂O (19870 kJ/kmol and 9500 kJ/kmol, respectively). Accordingly, the rise of total exergy was
406 noticed to be directly proportional to the increments of H₂ and CO in the product gas. At the same

407 time, the reduction of CO₂ and H₂O in the product gas led to the decrease of total exergy. Moreover,
408 as extra heat and gasification agent were needed in CO₂-enhanced process, the input exergy to the
409 gasifier was increased, which also resulted in the increase in inefficiencies. Other than H₂ and CO,
410 the total exergy losses due to extra input exergy and the reduction of exergy values contributed by
411 CO₂ and H₂O had less impact to the total exergy. Hence, exergetic efficiency of the CO₂-enhanced
412 route was greater than that of the conventional system. Besides, CO₂-enhanced biomass
413 gasification-based DME production experienced better performance within HRSG, Syn Cleaner,
414 CO₂ removal units. However, exergy L&Ds from the Comp and Cool (compression and cooling)
415 unit in CO₂-enhanced route were higher than those in the conventional route, as more syngas needs
416 to be processed. These units are usually standard products of manufacturing industries, therefore,
417 the potential for further reduction in their exergy losses (*i.e.*, by more efficient equipment) is
418 associated with high costs [48].

419 In comparison with the gasifier, which involved several reactions and led to low exergetic
420 efficiency, DME synthesis reactor (DME-R) offered a significantly better performance in both
421 routes. This was due to the relatively low chemical exergy stream entered to DME-R was
422 transformed into a product stream that contained a higher chemical exergy. This product stream
423 was obtained due to the formation of compounds, such as DME, MeOH, which had a high standard
424 chemical exergy. The exergy losses in DME-R were mainly derived from the intrinsic synthesis
425 reaction, which is hard to avoid. In this work, the production of DME was analysed based on the
426 two routes. In DME purification process, the major losses were attributed to the MeOH tower unit
427 (7-15%) and CO₂ removal unit (3-7%). It was found that conventional route performed better than
428 CO₂-enhanced route in terms of irreversibilities in MeOH tower unit.

429 **4.3 Environmental analysis**

430 The DME production based on CO₂-enhanced gasification has shown clear advantages against
431 conventional gasification in terms of fuel energetic and exergetic evaluation. An assessment of

432 environmental impacts is therefore needed. Accordingly, LCA-based environmental analysis was
433 conducted to compare the two routes. The environmental impacts caused in mid-points and end-
434 points categories under the investigated operation parameters are presented in Fig. 3 and Fig. 4,
435 respectively. It is obvious that CO₂-enhanced system showed overall less environmental impacts,
436 indicating a better performance compared with the conventional system although the differences in
437 various impact categories between the two systems are not notable. This is as the environmental
438 impacts are affected by many factors which will be detailed in the following paragraphs.

439 As seen in Fig. 3, there were five significant factors being considered in mid-point category where
440 climate change human health, fossil depletion and climate change ecosystem were the most
441 significant causes whose values were lower in CO₂-enhanced system than those in conventional
442 one. It is worth mentioning that the DME production in CO₂-enhanced system was 28% higher
443 although both total energy consumption and heat recovery were higher than those of conventional
444 system. Accordingly, the consumption of biomass and water per kg DME yield in the CO₂-
445 enhanced system were significantly lower (22% and 9%, respectively), which minimized the
446 energy requirement and therefore reduced environmental impacts. Furthermore, the net CO₂
447 emission per kg DME in CO₂-enhanced system was lower (about 16%) as it was used as an agent in
448 gasifier. As CO₂ is the key issue in the evaluation of environmental impacts of any processes, hence,
449 the reduction in CO₂ emission normally has a positive effect on environmental impacts. Similar
450 result was noticed with respect to particulate matter formation where its impact was moderate. On
451 the other hand, human toxicity demonstrated an insignificant impact where the values were almost
452 similar for both systems.

453 Regarding end-points impacts, the CO₂-enhanced system showed lower environmental impacts
454 than those of the conventional system as the end-points merge the information obtained from the
455 mid-points. It can be seen from Fig. 4 that human health combined the three categories of climate
456 change human health, human toxicity and particulate matter formation from the mid-points;

457 whereas ecosystem and resources stood for climate change ecosystem and fossil depletion,
458 respectively. From the end-points graph, it is obvious that human health and resources were highly
459 influenced than the ecosystem itself, which was around 50% lower than the others. In comparison
460 with conventional system, the CO₂-enhanced system showed a better environmental performance
461 by having a high-energy output, less biomass consumption and less CO₂ emission, which
462 compensated the additional heat and gasifying agent required in the process.

463 Besides, energetic, exergetic and environmental evaluation, an economic assessment is needed to
464 check the feasibility of the industrial scale bio-DME production based on CO₂-enhanced
465 gasification to compete with the current fossil fuel and biomass-based system. In spite of the huge
466 potential of CO₂ utilization introduced in gasification process for DME system, such advantages
467 that CO₂ can offer is vital in the assessment of gasifier performance as well as the total costs of
468 DME production. Due to the increment of DME production in CO₂-enhanced system, the product
469 costs are lower compared with those of the conventional system where the operation cost and cost
470 related to CO₂ emission are essential factors that will justify the utilization of CO₂ for industrial
471 applications. Furthermore, CO₂-enhanced based bio-DME production process is expected to
472 contribute to more efficient, competitive and sustainable clean fuel in near future, in order to fulfil
473 the recent challenges in more strict environmental regulations regarding low CO₂ emission
474 combined with the demand of low-cost product for the industries.

475 **5 Conclusions**

476 This work adopted thermodynamic and environmental approaches to compare conventional and
477 CO₂-enhanced biomass gasification based bio-DME production. Based on thermodynamic
478 efficiency indexes, *i.e.*, plant energy efficiency and plant exergetic efficiency, the proposed CO₂-
479 enhanced system demonstrated better performance than the conventional system. This improved
480 performance was mainly due to higher DME production and higher heat output. Meanwhile, the
481 exergetic evaluation in both routes showed that the largest loss occurred at the gasifier unit.

482 However, the addition of CO₂ as a gasifying agent reduced the loss in gasifier unit by 15%. The
483 environmental analysis showed that the CO₂-enhanced system offered a more sustainable approach
484 for bio-DME production. This is another benefit of using CO₂ in gasification compared with the
485 conventional one. These findings could assist in the development and commercialization of CO₂-
486 enhanced bio-DME production.

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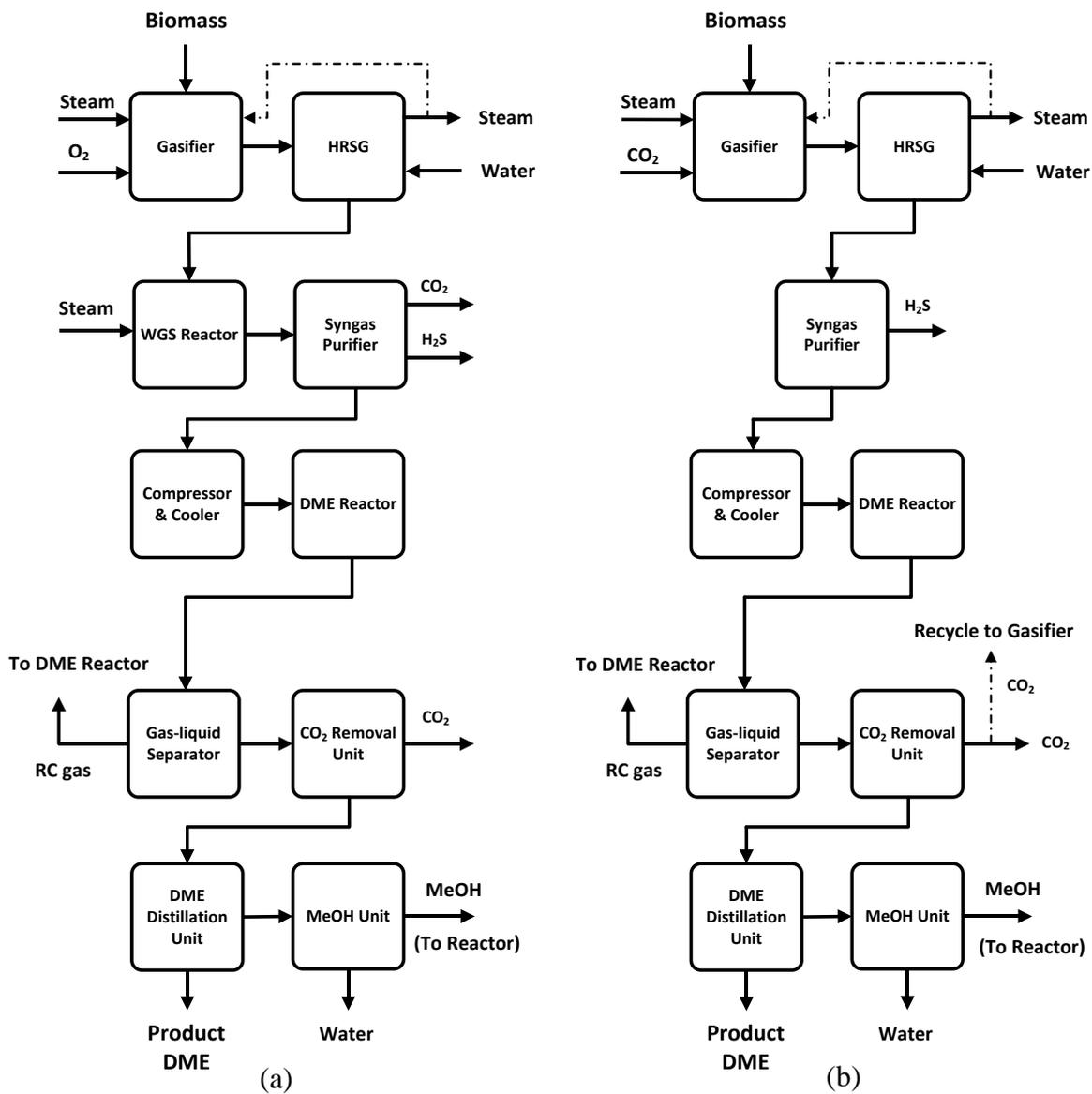
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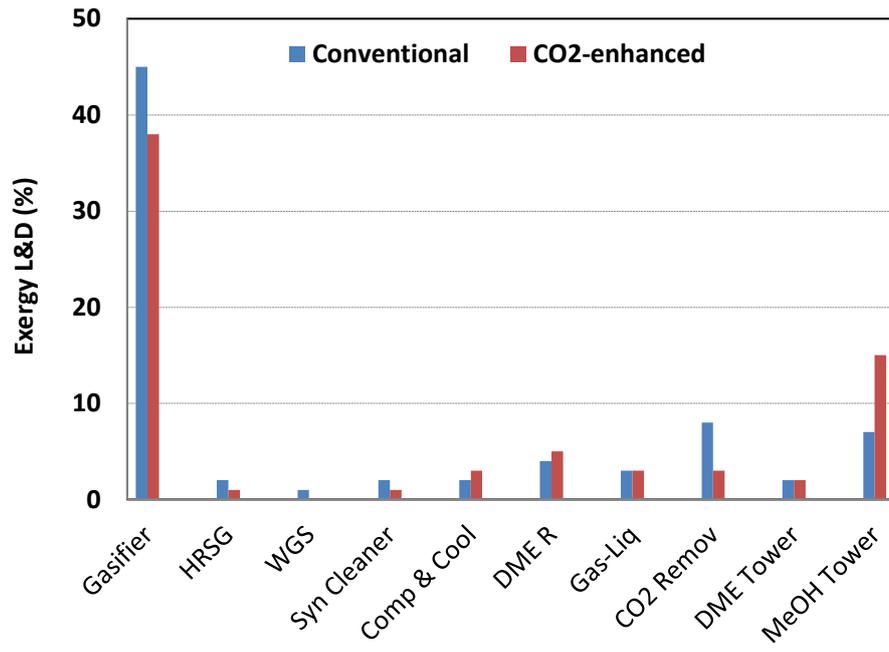


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615 **Figure 1: Process flow diagram of the single-step DME production based on (a) conventional**
 616 **and (b) CO₂-enhanced biomass gasification.**

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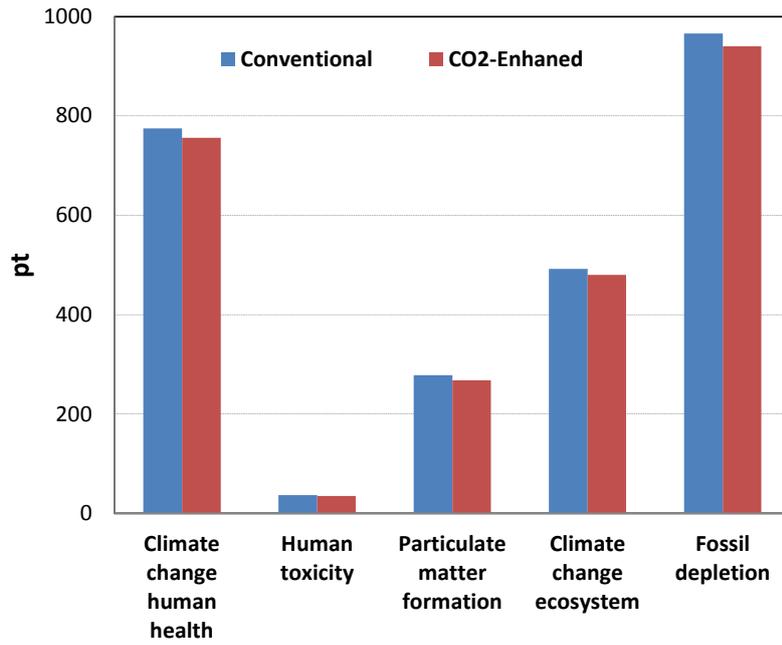
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Figure 2: Relative exergy losses and destructions of the considered bio-DME systems.

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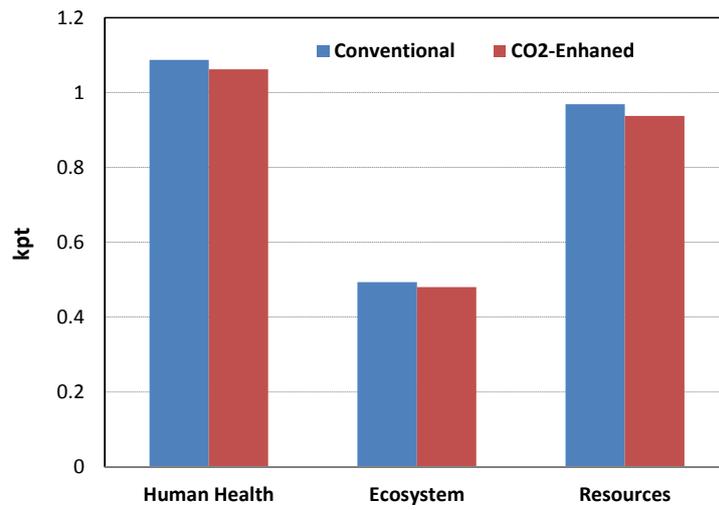
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625 **Figure 3: Environmental impacts (ReCiPe) in different categories (mid-points) – conventional**
626 **bio-DME system and CO₂-enhanced bio-DME system**

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Figure 4: Environmental impacts (ReCiPe) caused in the end-points (Human health, Ecosystem and Resources) – conventional bio-DME system and CO₂-enhanced bio-DME system

Table 1: Properties of gumwood

Net calorific value (MJ/kg)	20.0
Proximate analysis (wt %)	
Moisture	2.1
Volatile matter	86.0
Fixed carbon	11.8
Ash	0.1
Ultimate analysis ^{a,b} (wt %)	
C	47.1
H	6.3
O ^c	43.5
N	2.1
S	1.0
a: dry basis, b: ash free basis, c: by difference	

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Table 2: Gasification process parameters for the considered bio-DME systems.

Parameter	Conventional	CO₂-enhanced	Ju et al. [1]
Temperature (°C)	900	950	850
Pressure (MPa)	0.1	0.1	0.1
Biomass flow rate (kg/h)	50,000	50,000	75,600
O ₂ flow rate at 25 °C, 0.1 MPa (kg/h)	15,000	-	unknown
Steam flow rate at 150 °C, 0.5 MPa (kg/h)	2,000	11,500	unknown
CO ₂ flow rate at 25 °C, 0.1 MPa (kg/h)	-	9,500	-
Total gasification agent/Biomass	0.34	0.42	0.51
H ₂ /CO in syngas	0.85	1.0	1.0
CO ₂ mol% in syngas	6.7	2.9	17.0
LHV (MJ/Nm ³)	10.8	11.2	9.7
CGE (%)	70.5	77.8	74.6
GSE (%)	34.5	32.8	-

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641 **Table 3: Model validation against the gasification of wood (steam/biomass =0.2, P=0.1 MPa).**

Temp (°C)	H ₂ (mol. fraction)			CO (mol. fraction)			CO ₂ (mol. fraction)		
	Ref [21]	Present work	Error %	Ref [21]	Present work	Error%	Ref [21]	Present work	Error %
750	0.490	0.510	4.2	0.380	0.362	4.7	0.050	0.047	5.6
800	0.500	0.521	4.3	0.430	0.412	4.3	0.020	0.018	7.5
850	0.500	0.529	5.7	0.450	0.427	5.1	0.013	0.012	7.7
900	0.501	0.531	6.0	0.460	0.4247	7.7	0.008	0.008	6.2
950	0.508	0.543	6.9	0.465	0.446	4.1	0.001	0.0005	4.0
1000	0.510	0.552	8.3	0.470	0.449	4.4	0.0001	9.2E-05	8.0

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Table 4: Model validation against CO₂ gasification of biomass (CH_{1.56} O_{0.78}, CO₂/C= 0.5,

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P=0.1 MPa).

Temp (°C)	H ₂ (mol. fraction)			CO (mol. fraction)			CO ₂ (mol. fraction)		
	Ref [10]	Present work	Error%	Ref [10]	Present work	Error%	Ref [10]	Present work	Error%
800	0.310	0.331	7.0	0.598	0.572	4.4	0.091	0.085	6.3
1000	0.302	0.320	5.8	0.6248	0.592	5.2	0.073	0.067	8.6
1200	0.294	0.307	4.2	0.643	0.611	5.0	0.063	0.058	8.0

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Table 5: Summary of mass and energy balances for the considered bio-DME systems.

	Conventional (0.1 MPa, 900 °C)	CO₂-enhanced (0.1 MPa, 950 °C)
Plant inputs		
Biomass (kg/h)	50,000	50000
Biomass (MJ/h)	1,000,000	1,000,000
Oxygen (kg/h)	15,000	-
Oxygen (MJ/h)	-	-
Carbon dioxide (kg/h)	-	9,500
Carbon dioxide (MJ/h)	-	84,930
Steam (kg/h)	2,000	11,500
Steam (MJ/h)	26,380	151,685
Water (kg/h)	35,500	42,000
Water (MJ/h)	3,692	4,368
Electricity (MJ/h)	1,119,411	1,459,642
Plant outputs		
Dimethyl ether (kg/h)	23,312	29,941
Dimethyl ether (MJ/h)	662,061	850,324
Heat (MJ/h)	648,720	931,170
Efficiencies		
Fuel energy efficiency (%)	66.21	85.03
Plant energy efficiency (%)	60.98	65.97

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652 **Table 6: Comparison of DME yield and energetic efficiency of different bio-DME systems.**

Author	Gasification system	Biomass feedstock	Gasification agent	DME synthesis	DME yield (kg/kg biomass)	Plant energy efficiency (%)
Xiang et al. [22]	Entrained flow (1000 °C)	Raw sawdust	Steam, O ₂	Single-step	0.32	55.2
Zhang et al.[23]	Fluidized bed (880 °C)	Wood pellets	Steam	Double-step	0.34	51.3
Ju et al.[1]	Fluidized bed (850 °C)	Wood	Steam, O ₂	Single-step	0.37	52.7
Present work	Fluidized bed (900 °C)	Gum wood	Steam, O ₂	Single-step	0.46	60.98
	Fluidized bed (950 °C)	Gum wood	Steam, CO ₂	Single-step	0.59	65.97

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Table 7: Summary of exergy flow rate of the considered bio-DME systems.

	Conventional (0.1 MPa, 900 °C)	CO₂-enhanced (0.1 MPa, 950 °C)
Plant inputs		
Biomass (MJ/h)	1,056,557	1,056,557
Oxygen (MJ/h)	1,845	-
Carbon dioxide (MJ/h)	-	4,283
Steam (MJ/h)	1,259	7,240
Water (MJ/h)	1,775	1,500
Electricity (MJ/h)	516,953	753,869
Plant outputs		
Product		
Dimethyl ether (MJ/h)	719,545	923,637
Outputs		
Carbon dioxide (MJ/h)	27,778	47,791
Water (MJ/h)	637	430
Purge (MJ/h)	11,873	13,468
Heat		
Heat (MJ/h)	81,861	122,886
Efficiencies		
Exergetic efficiency (%)	50.8	57.3

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