**Effect of biomass type on the performance of cogasification of
low rank coal with biomass at relatively low temperatures**

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19 **Abstract**

20 Cogasification of low rank coal with three kinds of biomass, i.e., Japanese cedar, rice
21 straw, and seaweed, at relatively low temperatures were performed in a fixed bed
22 downdraft reactor. The effect of different biomass addition on the coal gasification
23 performance was investigated by comparing the experimental char and gas yields with
24 the calculated prediction data. Effect of the blending ratio was also investigated. It was
25 found that the mixing of coal with biomass revealed a synergy effect. Biomass
26 promoted the coal gasification rate, but the extent of promoting effect was depended on
27 the characteristics of biomass themselves. Alkali and alkaline earth metal (AAEM)
28 content played an important role in this synergy effect. Seaweed which contained high
29 AAEM content promoted the coal gasification significantly. Structure of coal was also
30 found to have great effect on the cogasification performance. The results indicated that
31 cogasification of coal with biomass is feasible at relatively low temperatures.

32 **Keywords:** *Cogasification; Low rank coal; Biomass; Synergy effect; Alkali and alkaline*
33 *earth metal*

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36 **1. Introduction**

37 Coal plays an important role in the world energy supply. To date, it is the second
38 largest of energy source after oil and the largest source for electricity. Due to its cost
39 feasibility, most of countries use coal for power generation. In Japan, coal contributes
40 21% of primary energy supplies in 2007 [1] and the percentage will grow up in the
41 future [2]. Energy from coal may be obtained either by direct combustion or
42 thermochemical conversion such as pyrolysis and gasification. Thermochemical route is
43 more favorable since it offers many advantages. Especially, it can convert solid fuel to
44 syngas and liquid fuels which are easier to be utilized and it can also reduce the
45 emission compared with that from direct combustion [3,4].

46 Biomass is considered as one of environmentally friendly fuels since it is renewable
47 energy source and contains less sulfur. A number of studies have been conducted to
48 estimate gasification performances using various kinds of biomass such as rice straw [3,
49 5, 6], woody biomass [7-9], palm empty fruit bunches [10], as the feedstock.
50 Unfortunately, biomass utilization also has some disadvantages. For instances, energy
51 density of biomass is lower than coal. Gasification of biomass also produces high
52 amount of tar which could reduce gasification efficiency significantly [11].

53 Cogasification of coal with biomass is an alternative way to overcome these

54 problems. Recently, there have been several studies in cogasification of coal with
55 various kinds of biomass. Pan et al. [12] cogasified low-rank coal with pine chips and
56 found that at certain mixing ratios, overall thermal efficiency was enhanced and carbon
57 conversion from coal was also significantly increased. Collot et al. [13] found that
58 synergy effect occurred when cogasified coal with several kinds of biomass, resulting in
59 the volatile yield increases about 5% higher than the expected. Such a synergy effect
60 was also found in other studies [11, 14-19]. However, not all cogasification studies
61 showed the synergy effect. Kastanaki et al. [20] reported that no interaction was
62 observed in the solid phase during thermal conversion of lignite-biomass blends. Meesri
63 and Moghtaderi [21] also reported that woody biomass and coal in the blended sample
64 were gasified independently without any interactions between each other. Several
65 factors may affect cogasification performance, such as reactor type, operating
66 temperature, heating rate, and also the type of biomass and coal as well as their blending
67 ratio in the sample.

68 The present study focused on the effect of biomass type on the performance of
69 cogasification of coal with biomass. Low rank coals (lignite) from two different sources
70 were respectively blended with three kinds of biomass, i.e., cedar wood, rice straw, and
71 seaweed. Low rank coal was chosen since it contains higher amount of volatile matters

72 than high rank coal and can be gasified at lower temperatures. Each of the selected
73 biomass had different characteristics so that the cogasification performance would be
74 different as well. Gasification was performed at relatively low temperatures in order to
75 investigate the possibility of coal gasification at lower temperatures in the presence of
76 biomass. The results of present study are expected to provide a better understanding in
77 selecting biomass to be cogasified with coal for the practical process.

78

79 **2. Experimental**

80 *2.1. Materials*

81 Two kinds of low rank coals (Loy Yang coal and Adaro coal) and three kinds of
82 biomass (brown seaweed/*Sargassum horneri*, rice straw, and Japanese
83 Cedar/*Cryptomeria japonica*,) were used. Biomass were collected from local area of
84 Aomori Prefecture, Japan. Proximate, ultimate, and ash content analysis for these
85 samples are shown in Table 1. Here, proximate analysis was done based on ASTM
86 3172-75. Ultimate analysis was carried out using an elemental analyzer (Vario EL cube
87 elemental analyzer). Ash compositions were analyzed using an energy dispersive X-Ray
88 spectrometer (EDX-800HS, Shimadzu).

89 All samples were ground and sieved into particle size with a range of 1 – 2.8 mm at

90 first, and then dried at 105 °C overnight and stored in desiccators prior to the testing.
91 For copyrolysis/cogasification process, blended samples of coal and biomass were
92 prepared by physical mixing of them with biomass-to-coal weight ratios of 1:2, 1:1, and
93 2:1, respectively.

94

95 2.2 *Cogasification of coal with biomass*

96 Pyrolysis/gasification was performed in a fixed bed downdraft reactor. Schematic
97 illustration of the experimental setup is presented in Fig.S1. Stainless steel tube reactor
98 with an inner diameter of 16.5 mm and a length of 500 mm was put inside an electric
99 furnace which provided heat for the reaction. For each run, 1 g of the sample was loaded
100 into a sample holder in the middle section of reactor. The setup was then flushed by
101 argon gas prior experiment in order to remove air out of the system. The reactor was
102 heated from room temperature with a heating rate of 25 °C/min until it reached the
103 desired gasification temperature. The total reaction time from the beginning of heating
104 was fixed at 120 min for all tests. Gasification temperature was varied in the range of
105 500 – 750 °C. Steam as gasifying agent was generated by heating distilled water with a
106 flow rate of 0.09 g/min in a furnace with a temperature of 250 °C. Ribbon heater was
107 used to avoid condensation of steam in the steam-gas line between steam generator and

108 reactor. Steam was introduced into the reactor from the beginning of heating, which was
109 carried by 50 cm³/min of argon gas flow.

110 Volatile product and steam left the reactor from the bottom side were condensed in
111 a cold trap. Cold trap consists of two bottles placed in the ice bath. Non-condensable
112 gases passed through a gas purifier contained calcium chloride to remove out the
113 remained moisture. Dry gas was then collected in a gas bag from the beginning of
114 heating. After the reaction, the reactor was cooled down and the remained solid in the
115 sample holder was weighted and defined as char and ash.

116

117 2.3 Analysis

118 The collected gas was analyzed using a gas chromatograph (Agilent 7890A GC
119 system) with two thermal conductivity detectors (TCD). One TCD was connected with
120 three packed columns (2 molecular sieves 5A and 1 HayeSep Q column) to separate CO,
121 CH₄, and CO₂ using helium as carrier gas while the other TCD was connected with a
122 molecular sieve 5A column to measure H₂ using argon as carrier gas. Carbon content in
123 the char was analyzed using an elemental analyzer (Vario EL cube elemental analyzer).
124 Specific surface area of char was determined using BET sorption isotherm method
125 (Quantachrome NOVA 4200e) and its surface state was observed using a scanning

126 electron microscope (SEM) equipped with and an energy dispersive X-ray spectrometer
127 (EDX) (SEM HITACHI SU6600).

128 Experimental char or gas production yield was compared with the predicted data.
129 Predicted production yield (Y_{pred}) is the cogasification result assuming that there are no
130 any interactions occurred between coal and biomass during the
131 copyrolysis/cogasification process. Y_{pred} is calculated as follow:

$$132 \quad Y_{pred} = X_{bio} \times Y_{bio} + (1 - X_{bio}) \times Y_{coal} \quad (1)$$

133 where X_{bio} is mass fraction of biomass in the mixed sample while Y_{bio} and Y_{coal} are gas
134 and char production yields from biomass and coal when they are separately gasified,
135 respectively.

136

137 **3. Results and discussion**

138 *3.1. Pyrolysis/Gasification of pure sample*

139 Fig.1 shows char and gas production yields from 5 kinds of samples when they
140 were gasified at 600 °C with and without steam. It can be seen that the char yield
141 decreased while gas yield increased for all samples in the presence of steam, indicating
142 that steam gasification occurred even at the low temperature. Products distribution and
143 yields from the gasification process were highly dependent on the characteristic of the

144 feedstock as well as the gasification condition. The comparison of char and total gas
145 yield per gram of carbon from the pyrolysis/gasification of two kinds of low rank coals
146 and three kinds of biomass at gasification temperatures in the range of 500 – 750 °C are
147 presented in Fig.2. Here, steam was used as gasifying agent at a flow rate of 0.09 g/min.
148 As shown in Fig.2, the char yield decreased while the total gas yield increased with the
149 increase of temperature in all cases. The results shown in Fig.2(a) revealed that Adaro
150 coal produced a little more char than Loy Yang coal. In general, the decrease of char
151 yield represents the higher carbon conversion due to the steam gasification, hence, this
152 result indicates that Loy Yang coal was more easily gasified by the steam than Adaro
153 coal. Gasification conversion rate is generally proportional with the volatile matter
154 content in the coal. Sample with higher volatile matter will be more reactive and can be
155 gasified more easily and produced less char [4]. Proximate analysis of both coals also
156 indicate that Loy Yang coal contained more volatile matter than Adaro coal. Therefore,
157 in this study, more carbon in Loy Yang coal was converted. However, the gas yields
158 shown in Fig.2(a) indicated that the steam gasification of Adaro coal produced more gas
159 than Loy Yang coal. Higher gas yield from Adaro coal than that from Loy yang coal
160 may be due to the microstructure difference of the coals. SEM results of the coal chars
161 showed that Loy Yang char structure was more porous than Adaro char (Fig.S2 in the

162 supplementary data). It is reported that coal with more pore structure could react faster,
163 but tend to produce more tar at lower temperatures [4]. Therefore, in this study, Loy
164 Yang coal produced less gas than Adaro coal. It should be noted that the differences of
165 the gas yields at lower temperatures ($<650^{\circ}\text{C}$) and higher temperatures (750°C) were
166 minor for both low-rank coals.

167 Char yields resulted from the steam gasification of biomass in Fig.2(b) shows that
168 seaweed was almost completely gasified at relatively low temperature (650°C). Two
169 other biomass, i.e., rice straw and cedar wood, still produced high amount of char even
170 at 750°C . Ash content analysis indicated that seaweed ash contained about 77% of
171 alkali and alkaline earth metal (AAEM) (Table 1), which is much higher than those in
172 the ashes of other biomass. AAEM species is known to have good catalytic activity on
173 the gasification [22-27]. The presence of AAEM species in the sample could promote
174 char decomposition effectively since it is highly dispersed in the carbon matrix. On the
175 other hand, as shown in Fig.2(b), gas yield from the seaweed sample was also much
176 higher than those from other biomass. It is reported that AAEM species, especially
177 potassium, have higher catalytic activity on the tar reforming [28, 29]. Therefore, the
178 gas yield from the steam gasification of seaweed was very high even at low
179 temperatures. Furthermore, it should be noted that the gas yield increased significantly

180 from the gasification of seaweed when the gasification temperature was increased from
181 600 to 650 °C.

182

183 3.2. *Cogasification of Adaro coal with various biomass*

184 Fig.3 compares the experimental results with the predicted data for the
185 cogasification of Adaro coal with various kinds of biomass. Here, coal and biomass was
186 blended at a weight ratio of 1:1 on dry basis. One can see that the experimental char
187 yields were a little lower than the predicted data while the experimental gas yields were
188 higher than the predicted data for almost all blended samples, except for the char yields
189 from cogasification of Adaro coal with cedar wood at the temperatures of 650 and 700
190 °C. It indicates that synergy effect between coal and biomass was happened as the
191 addition of biomass could promote coal gasification. However, the extents of promoting
192 effect from different biomass were different. Cedar wood showed the lowest promoting
193 effect on the gasification of coal while seaweed showed the highest. This difference
194 should be strongly caused by the difference contents of AAEM species in the different
195 biomass. AAEM species in the biomass could undergo volatilization due to the breakage
196 of the bond between metal and char matrix [30] and then attach on the coal surface.
197 Thus, the reactivity of coal would be enhanced to some extent and easier to be gasified

198 by the steam, producing more gas. Higher content of AAEM species in the biomass
199 might be volatilized more easily and attached more on the coal surface. It is verified by
200 the EDX analysis of the coal char after the steam gasification (Fig.S3 in the
201 supplementary data). No alkali was detected on the coal char if the coal was blended
202 with cedar wood for the cogasification. However, alkali metal was detected on the
203 surface of coal char when it was cogasified with seaweed. Since alkali content in Adaro
204 coal was very low, no alkali was detected on the surface of char in its original state.
205 Therefore, alkali presence in the seaweed-blended coal char should come from the
206 seaweed.

207 The addition of cedar was also observed to have the promoting effect even though
208 very low amount of AAEM was detected on the cedar sample. The promoting effect
209 cedar wood was possibly caused by the radicals and/or H donors from the biomass
210 pyrolysis. He et al. [31] reported that a large amount of radicals were contained in the
211 biomass tar, and these radicals could enhance the gasification of coal when they contact
212 with the coal. Other researchers considered that the relatively high hydrogen in biomass
213 could play a synergistic role as H donors to coal during co-liquefaction [32] and
214 copyrolysis [33,34]. It is also possible that such a role will be played in coal gasification
215 with the biomass. The differences between the experimental and the predicated gas

216 yields became a little more significant at higher temperature since more radicals and/or
217 H donors could be released at higher temperature [31-34].

218 Fig.4 shows the total gas yield from Adaro coal when it was cogasified with
219 biomass at various blending ratio. Here, the blending ratio was represented by the ratio
220 of carbon in biomass to carbon in coal ($C_{\text{biomass}}/C_{\text{coal}}$). The total gas yield from the coal
221 was calculated from the experimental total gas yield by assuming that the gas yield from
222 biomass was proportional with the amount of biomass in the blended sample.
223 Investigation was done at two different temperatures, i.e., 600°C and 750°C. It is found
224 that the total gas yield from seaweed-blended coal was the highest; followed by rice
225 straw-blended coal and cedar-blended coal. Herein, it should be noted that different
226 trends were observed at different temperatures. At the lower temperature (600 °C), the
227 total gas yield from coal increased along with the increase in the amount of biomass in
228 the blended sample. AAEM species in the biomass are usually more easily released,
229 moved to the surface of coal and assist the steam gasification of coal at higher
230 temperature. As such, at low temperature, less AAEM from per gram of biomass will be
231 released for the coal gasification promoting. In this case, one way is to increase the
232 biomass amount in the blended sample and as such, the total amount of AAEM released
233 from the biomass will be increased so that the coal gasification rate is also increased.

234 Also, as stated above, radicals and/or H donor from biomass pyrolysis could improve
235 the gasification rate of coal [31-34]. However, at low gasification temperature, less
236 volatile matter could be released from biomass. In this case, more biomass addition in
237 the blended sample is expected to produce radicals and/or H donors for the coal
238 gasification. Therefore, in this study, the total gas yield from coal increased with the
239 increase in the amount of biomass in the blended sample at low temperature (600 °C). In
240 contrast, at the higher temperature (750°C), the blending ratio seemed to have no effect
241 on the total gas yield from coal. At high temperature, biomass char amount is generally
242 low and in this case, more volatile matter and AAEM species could be generated from
243 biomass so that effective contact can happen even with a small amount of biomass in
244 the blended sample. Increasing biomass amount could not further increase gas
245 production since the coal gasification could be well catalyzed by the enough AAEM
246 species as well as the radicals and/or H donors from the biomass pyrolysis. On the other
247 hand, it is reported that the presence of hydrogen gas in a high concentration resulted in
248 an inhibition effect on the gasification process [35]. In this study, the gasification of coal
249 char could be also hindered by the high amount of hydrogen gas produced at high
250 temperature. Besides, the addition of cedar did not promote the gasification rate of coal
251 obviously. As shown in Table 1, only 0.6 wt% of ash was contained in the cedar,

252 indicating that only a little amount of AAEM species can be released from the cedar.
253 Although more volatile matter could be released from it, the main catalytic activity for
254 the coal gasification should be from the AAEM species. For rice straw, although it
255 contained high content of ash, the main composition in its ash is silica, which could
256 hinder the release of AAEM species from it [28]. Hence, a lower promoting effect was
257 observed when comparing with the seaweed.

258

259 3.3 *Copypyrolysis/cogasification of Loy Yang coal with seaweed*

260 From the previous results, seaweed had the highest promoting effect on the
261 gasification of Adaro coal when compared with other selected biomass. To confirm this
262 promoting effect, seaweed was also blended with Loy Yang coal for the cogasification.
263 As shown in Fig.5, the similar results as those for Adaro coal were obtained, indicating
264 that seaweed also has excellent promoting effect on other low rank coal gasification.
265 Fig.6 compares the total gas yield from Adaro and Loy Yang coals when they were
266 cogasified with seaweed at various blending ratios. One can see that the total gas yield
267 from Loy Yang coal was much higher than that from Adaro coal at any blending ratio. It
268 indicates that the promoting effect of seaweed on the steam gasification of Loy Yang
269 coal was higher than that of Adaro coal. As stated above, Loy Yang coal produced more

270 volatiles to form more tar than Adaro coal when they were gasified separately. In the
271 case of cogasification, the tar produced from coal can be also reformed by steam to
272 produce more gas in the presence of seaweed since AAEM released from the seaweed
273 also had good catalytic effect on the tar reforming [28, 29]. Furthermore, specific
274 surface area of Loy Yang char was 364.6 m²/g but that of Adaro char was only 133.2
275 m²/g. The larger surface area could make Loy Yang char to capture more volatilized
276 AAEM species than Adaro char. EDX analysis indicated that about 3 atom% potassium
277 was detected on the surface of Loy Yang char while only 0.43 atom% potassium was
278 found on the surface of Adaro char, indicating that the Loy Yang char had more
279 reactivity than Adaro char. SEM analysis also shows that Loy Yang char had more
280 porous structure than Adaro char (Fig. S2 in the supplementary data) and thus, H and
281 OH radicals derived from biomass could penetrate deeper into the porous structure, and
282 make more aromatic rings broken down. As a result, the char yield from Loy Yang coal
283 was lower while the gas yield was higher than those from Adaro coal.

284

285 **4. Conclusions**

286 Cogasification of low rank coal with 3 kinds of biomass was carried out in a fixed
287 bed downdraft reactor. It is found that the coal gasification rate was promoted by the

288 addition of biomass. However, the extent of promoting effect was different when
289 different biomass was used for the cogasification. AAEM content in the biomass played
290 an important role in this synergy effect. Biomass with high content of AAEM promoted
291 the gas production from coal significantly. The structures of coal as well as coal char
292 also had some influence on the promoting effect. At lower gasification temperature, the
293 extent of promoting effect was enhanced by the adding more biomass into the coal,
294 However, at higher temperature, a smaller amount of biomass could have great
295 promoting effect on the coal gasification.

296

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401 **Figure captions**

402 **Fig.1** Effect of steam addition on the char and gas yields from pyrolysis/gasification of
403 5 kinds of samples.

404 **Fig.2** Comparison of char and gas yields from different samples; (a) coals and (b)
405 biomass.

406 **Fig.3** Char and gas yields from copyrolysis/cogasification of Adaro coal with various
407 biomass at a biomass-to-coal weight ratio of 1:1; (a) Adaro coal with cedar; (b) Adaro
408 coal with rice straw; and (c) Adaro coal with seaweed.

409 **Fig.4** Effect of blending ratio on the gas yield from coal when it was cogasified with
410 various biomass at two different temperatures, 600 and 750 °C.

411 **Fig.5** Char and gas yields from cogasification of Loy Yang coal with seaweed at a
412 biomass-to-coal weight ratio of 1:1.

413 **Fig.6** Gas yields of two different coals when they were gasified with seaweed in various
414 blending ratios at 750 °C.

415

416 **Table caption**

417 **Table 1** Proximate, ultimate, and ash content analysis of the samples.

418

419 Table 1

sample	Adaro coal	Loy yang	cedar	seaweed	rice straw
Proximate analysis (wt.%) dry base					
Ash	2.1	1.6	0.6	13.5	14.0
VM	47.8	51.5	86.9	52.8	52.0
FC*	50.1	47	12.5	33.7	34.0
Ultimate analysis (wt.%) d.a.f					
C	70.0	67.3	48.8	39.2	35.1
H	4.9	4.9	6.6	5.0	5.3
O*	22.6	26.9	43.0	53.9	59.3
N	0.4	0.9	1.4	1.8	0.4
Ash composition (wt%)					
SiO ₂	34.97	14.79	6.83	n.d.	53.208
Al ₂ O ₃	19.83	36.01	n.d	n.d.	n.d.
Fe ₂ O ₃	21.38	7.68	0.8	0.376	2.359
CaO	11.54	6.43	50.68	34.142	2.591
MgO	2.85	13.99	n.d	25.176	n.d.
Na ₂ O	0.28	n.d.	n.d	n.d.	n.d.
K ₂ O	0.81	0.88	4.01	17.749	26.853
P ₂ O ₅	0.12	n.d.	6.45	2.453	2.421
TiO ₂	1.13	0.81	n.d	n.d.	n.d.
SO ₃	7.09	19.41	10.47	13.790	3.188
SrO	n.d.	n.d.	0.04	5.771	n.d.
Cl ₂ O	n.d.	n.d.	n.d	n.d.	8.736
MnO ₂	n.d.	n.d.	n.d	n.d.	0.510
Br ₂ O	n.d.	n.d.	n.d	0.203	0.015
Rb ₂ O	n.d.	n.d.	n.d	0.020	0.034
ZnO	n.d.	n.d.	n.d	0.227	0.040
CuO	n.d.	n.d.	n.d	n.d.	0.044

VM: volatile matter; FC: fixed carbon; d.a.f: dry ash-free; n.d: not detected

*calculated by difference.

420

421

422

423 Graphical Abstract

424

425

Figure 1

Figure 3

Figure 4

Figure 5

Figure 6