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

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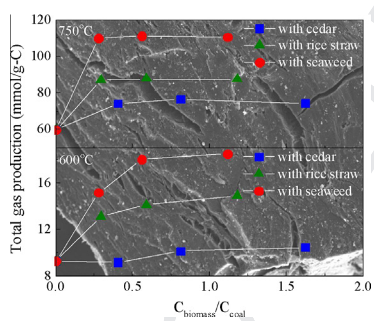
HIGHLIGHTS

- Cogasification of low rank coal with biomass was investigated.
- Synergy effect between coal and biomass in cogasification process was clarified.
- Different promoting effect from different biomass was observed.
- Promoting effect from biomass was different for different coal microstructure.

GRAPHICAL ABSTRACT



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ABSTRACT

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

1. Introduction

Coal plays an important role in the world energy supply. To date, it is the second largest of energy source after oil and the largest source for electricity. Due to its cost feasibility, most of countries use coal for power generation. In Japan, coal contributes 21% of primary energy supplies in 2007 [1] and the percentage will grow up in the future [2]. Energy from coal may be obtained either

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by direct combustion or thermochemical conversion such as pyrolysis and gasification. Thermochemical route is more favorable since it offers many advantages. Especially, it can convert solid fuel to syngas and liquid fuels which are easier to be utilized and it can also reduce the emission compared with that from direct combustion [3,4].

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

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

The present study focused on the effect of biomass type on the performance of cogasification of coal with biomass. Low rank coals (lignite) from two different sources were respectively blended with three kinds of biomass, i.e., cedar wood, rice straw, and seaweed. Low rank coal was chosen since it contains higher amount of volatile matters than high rank coal and can be gasified at lower temperatures. Each of the selected biomass had different characteristics so that the cogasification performance would be different as well. Gasification was performed at relatively low temperatures in order to investigate the possibility of coal gasification at lower temperatures in the presence of biomass. The results of present study are expected to provide a better understanding in selecting biomass to be cogasified with coal for the practical process.

2. Experimental

2.1. Materials

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

All samples were ground and sieved into particle size with a range of 1–2.8 mm at first, and then dried at 105 °C overnight and stored in desiccators prior to the testing. For copyrolysis/cogasification process, blended samples of coal and biomass were prepared by physical mixing of them with biomass-to-coal weight ratios of 1:2, 1:1, and 2:1, respectively.

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

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_2O_3	19.83	36.01	n.d	n.d.	n.d.
Fe_2O_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_2O	0.28	n.d.	n.d	n.d.	n.d.
K_2O	0.81	0.88	4.01	17.749	26.853
P_2O_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_2O	n.d.	n.d.	n.d	n.d.	8.736
MnO_2	n.d.	n.d.	n.d	n.d.	0.510
Br_2O	n.d.	n.d.	n.d	0.203	0.015
Rb_2O	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.

2.2. Cogasification of coal with biomass

Pyrolysis/gasification was performed in a fixed bed downdraft reactor. Schematic illustration of the experimental setup is presented in Fig. S1. Stainless steel tube reactor with an inner diameter of 16.5 mm and a length of 500 mm was put inside an electric furnace which provided heat for the reaction. For each run, 1 g of the sample was loaded into a sample holder in the middle section of reactor. The setup was then flushed by argon gas prior experiment in order to remove air out of the system. The reactor was heated from room temperature with a heating rate of 25 °C/min until it reached the desired gasification temperature. The total reaction time from the beginning of heating was fixed at 120 min for all tests. Gasification temperature was varied in the range of 500–750 °C. Steam as gasifying agent was generated by heating distilled water with a flow rate of 0.09 g/min in a furnace with a temperature of 250 °C. Ribbon heater was used to avoid condensation of steam in the steam–gas line between steam generator and reactor. Steam was introduced into the reactor from the beginning of heating, which was carried by 50 cm^3/min of argon gas flow.

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

2.3. Analysis

The collected gas was analyzed using a gas chromatograph (Agilent 7890A GC system) with two thermal conductivity detectors

(TCD). One TCD was connected with three packed columns (2 molecular sieves 5A and 1 HayeSep Q column) to separate CO, CH_4, and CO_2 using helium as carrier gas while the other TCD was connected with a molecular sieve 5A column to measure H_2 using argon as carrier gas. Carbon content in the char was analyzed using an elemental analyzer (Vario EL cube elemental analyzer). Specific surface area of char was determined using BET sorption isotherm method (Quantachrome NOVA 4200e) and its surface state was observed using a scanning electron microscope (SEM) equipped with and an energy dispersive X-ray spectrometer (EDX) (SEM HITACHI SU6600).

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

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

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

3. Results and discussion

3.1. Pyrolysis/gasification of pure sample

Fig. 1 shows char and gas production yields from 5 kinds of samples when they were gasified at 600 °C with and without steam. It can be seen that the char yield decreased while gas yield increased for all samples in the presence of steam, indicating that steam gasification occurred even at the low temperature. Products distribution and yields from the gasification process were highly dependent on the characteristic of the feedstock as well as the gasification condition. The comparison of char and total gas yield per gram of carbon from the pyrolysis/gasification of two kinds of low rank coals and three kinds of biomass at gasification temperatures in the range of 500–750 °C are presented in Fig. 2. Here, steam was used as gasifying agent at a flow rate of 0.09 g/min. As shown in Fig. 2, the char yield decreased while the total gas yield increased with the increase of temperature in all cases. The results shown in Fig. 2(a) revealed that Adaro coal produced a little more char than Loy Yang coal. In general, the decrease of char yield represents the higher carbon conversion due to the steam gasification, hence, this result indicates that Loy Yang coal was more easily gasified by the steam than Adaro coal. Gasification conversion rate is generally proportional with the volatile matter content in the coal. Sample with higher volatile matter will be more reactive and can be gasified more easily and produced less char [4]. Proximate analysis of both coals also indicate that Loy Yang coal contained more volatile matter than Adaro coal. Therefore, in this study, more carbon in Loy Yang coal was converted. However, the gas yields shown in Fig. 2(a) indicated that the steam gasification of Adaro coal produced more gas than Loy Yang coal. Higher gas yield from Adaro coal than that from Loy yang coal may be due to the microstructure difference of the coals. SEM results of the coal chars showed that Loy Yang char structure was more porous than Adaro char (Fig. S2 in Supplementary Data). It is reported that coal with more pore structure could react faster, but tend to produce more tar at lower temperatures [4]. Therefore, in this study, Loy Yang coal produced less gas than Adaro coal. It should be noted that the differences of the gas yields at lower temperatures (<650 °C) and higher temperatures (750 °C) were minor for both low-rank coals.

Char yields resulted from the steam gasification of biomass in Fig. 2(b) shows that seaweed was almost completely gasified at relatively low temperature (650 °C). Two other biomass, i.e., rice straw and cedar wood, still produced high amount of char even at 750 °C. Ash content analysis indicated that seaweed ash contained about 77% of alkali and alkaline earth metal (AAEM) (Table 1), which is much higher than those in the ashes of other biomass. AAEM species is known to have good catalytic activity on the gasification [22–27]. The presence of AAEM species in the sample could promote char decomposition effectively since it is highly dispersed in the carbon matrix. On the other hand, as shown in Fig. 2(b), gas yield from the seaweed sample was also much higher than those from other biomass. It is reported that AAEM species, especially potassium, have higher catalytic activity on the tar reforming [28,29]. Therefore, the gas yield from the steam gasification of seaweed was very high even at low temperatures. Furthermore, it should be noted that the gas yield increased significantly from the gasification of seaweed when the gasification temperature was increased from 600 to 650 °C.

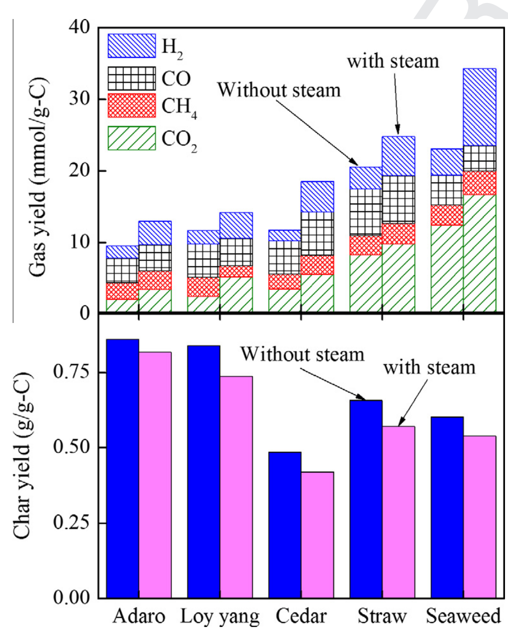

Fig. 1. Effect of steam addition on the char and gas yields from pyrolysis/gasification of 5 kinds of samples.

3.2. Cogasification of Adaro coal with various biomass

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

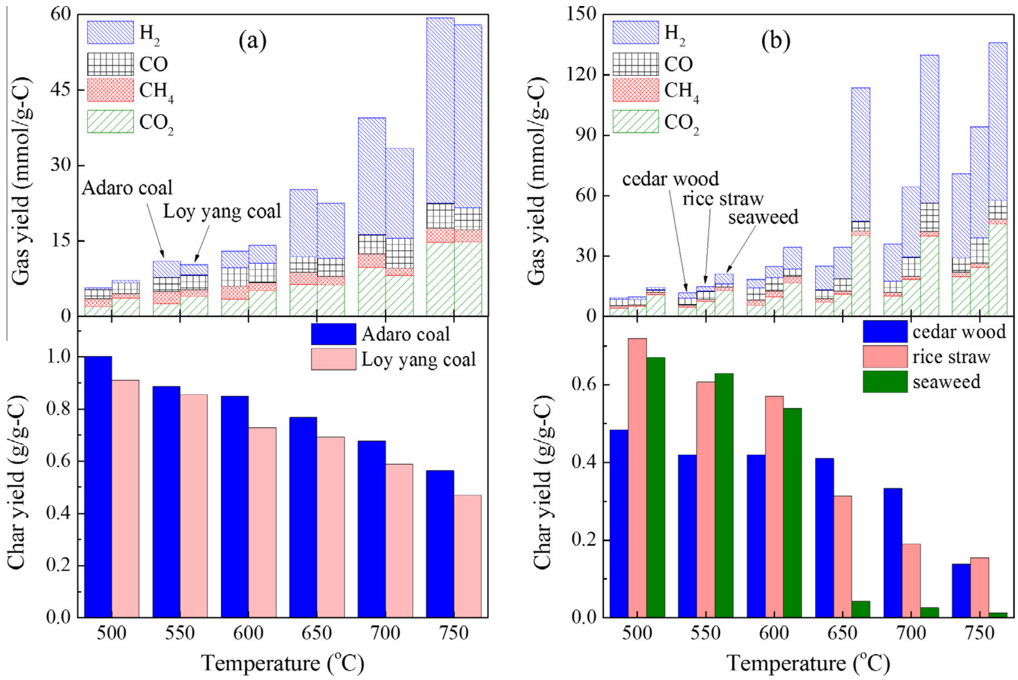

Fig. 2. Comparison of char and gas yields from different samples; (a) coals and (b) biomass.

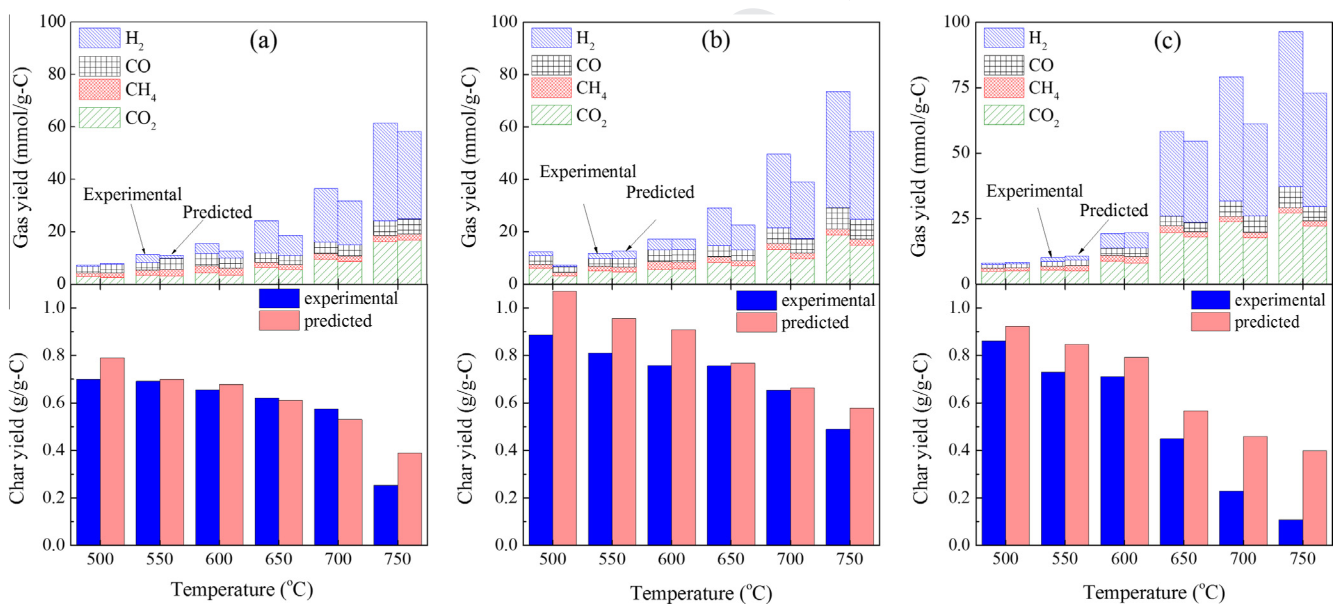

Fig. 3. Char and gas yields from copyrolysis/cogasification of Adaro coal with various biomass at a biomass-to-coal weight ratio of 1:1; (a) Adaro coal with cedar; (b) Adaro coal with rice straw; and (c) Adaro coal with seaweed.

is verified by the EDX analysis of the coal char after the steam gasification (Fig. S3 in Supplementary data). No alkali was detected on the coal char if the coal was blended with cedar wood for the cogasification. However, alkali metal was detected on the surface of coal char when it was cogasified with seaweed. Since alkali content in Adaro coal was very low, no alkali was detected on the surface of char in its original state. Therefore, alkali presence in the seaweed-blended coal char should come from the seaweed.

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

Fig. 4 shows the total gas yield from Adaro coal when it was cogasified with biomass at various blending ratio. Here, the blending ratio was represented by the ratio of carbon in biomass to carbon in coal ($C_{biomass}/C_{coal}$). The total gas yield from the coal was calculated from the experimental total gas yield by assuming that the gas yield from biomass was proportional with the amount of biomass in the blended sample. Investigation was done at two different temperatures, i.e., 600 °C and 750 °C. It is found that the total gas yield from seaweed-blended coal was the highest; followed by rice straw-blended coal and cedar-blended coal. Herein, it should be noted that different trends were observed at different temperatures. At the lower temperature (600 °C), the total gas yield from coal increased along with the increase in the amount of biomass in the blended sample. AAEM species in the biomass are usually more easily released, moved to the surface of coal and assist the steam gasification of coal at higher temperature. As such, at low temperature, less AAEM from per gram of biomass will be released for the coal gasification promoting. In this case, one way is to increase the biomass amount in the blended sample and as such, the total amount of AAEM released from the biomass will be increased so that the coal gasification rate is also increased. Also, as stated above, radicals and/or H donor from biomass pyrolysis could improve the gasification rate of coal [31–34]. However, at low gasification temperature, less volatile matter could be released from biomass. In this case, more biomass addition in the blended sample is expected to produce radicals and/or H donors for the coal gasification. Therefore, in this study, the total gas yield from coal increased with the increase in the amount of biomass in the blended sample at low temperature (600 °C). In contrast, at the higher temperature (750 °C), the blending ratio seemed to have no effect on the total gas yield from coal. At high temperature, biomass char amount is generally low and in this case, more volatile matter and AAEM species could be generated from biomass so that effective contact can happen even with a small amount of biomass in the blended sample. Increasing biomass amount could not further increase gas production since the coal gasification could be well catalyzed by the enough AAEM species as well as the radicals and/or H donors from the biomass pyrolysis. On the other hand, it is reported that the presence of hydrogen gas in a high concentration resulted in an inhibition effect on the gasification process [35]. In this study, the gasification of coal char could be also hindered by the high amount of hydrogen gas produced at high temperature. Besides, the addition of cedar did not promote the gasification rate of coal obviously. As shown in Table 1, only 0.6 wt% of ash was contained in the cedar, indicating that only a little amount of AAEM species can be released from the cedar. Although more volatile matter could be released from it, the main catalytic activity for the coal gasification should be from the AAEM species. For rice straw, although it contained high content of ash, the main composition in its ash is silica, which could hinder the release of AAEM species from it [28]. Hence, a lower promoting effect was observed when comparing with the seaweed.

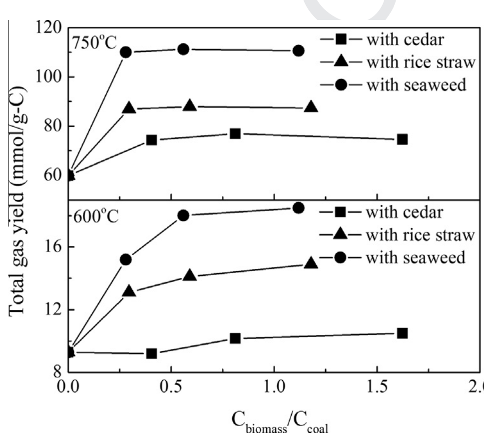

Fig. 4. Effect of blending ratio on the gas yield from coal when it was cogasified with various biomass at two different temperatures, 600 and 750 °C.

3.3. Copyrolysis/cogasification of Loy Yang coal with seaweed

From the previous results, seaweed had the highest promoting effect on the gasification of Adaro coal when compared with other selected biomass. To confirm this promoting effect, seaweed was also blended with Loy Yang coal for the cogasification. As shown in Fig. 5, the similar results as those for Adaro coal were obtained, indicating that seaweed also has excellent promoting effect on other low rank coal gasification. Fig. 6 compares the total gas yield from Adaro and Loy Yang coals when they were cogasified with seaweed at various blending ratios. One can see that the total gas yield from Loy Yang coal was much higher than that from Adaro coal at any blending ratio. It indicates that the promoting effect of seaweed on the steam gasification of Loy Yang coal was higher than that of Adaro coal. As stated above, Loy Yang coal produced more volatiles to form more tar than Adaro coal when they were gasified separately. In the case of cogasification, the tar produced from coal can be also reformed by steam to produce more gas in the presence of seaweed since AAEM released from the seaweed also had good catalytic effect on the tar reforming [28,29]. Furthermore, specific surface area of Loy Yang char was 364.6 m^2/g but that of Adaro char was only 133.2 m^2/g. The larger surface area could make Loy Yang char to capture more volatilized AAEM species than Adaro char. EDX analysis indicated that about 3 atom% potassium was detected on the surface of Loy Yang char while only 0.43 atom% potassium was found on the surface of Adaro char, indicating that the Loy Yang char had more reactivity than Adaro char. SEM analysis also shows that Loy Yang char had more porous structure than Adaro char (Fig. S2 in supplementary data) and thus, H and OH radicals derived from biomass could penetrate deeper into the porous structure, and make more aromatic rings broken down. As a result, the char yield from Loy Yang coal was lower while the gas yield was higher than those from Adaro coal.

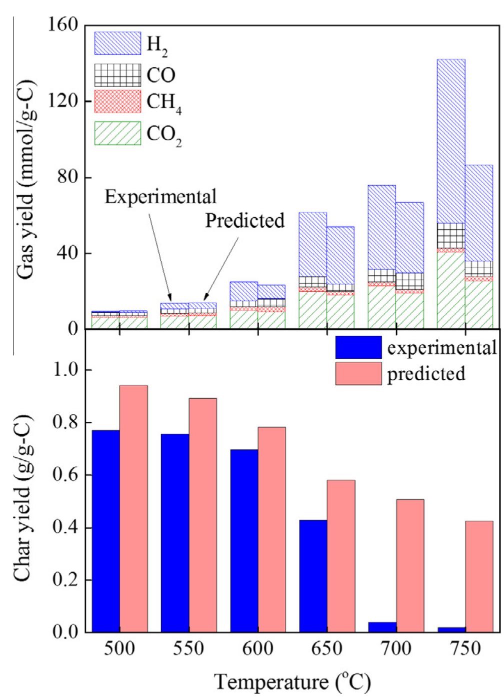

Fig. 5. Char and gas yields from cogasification of Loy Yang coal with seaweed at a biomass-to-coal weight ratio of 1:1.

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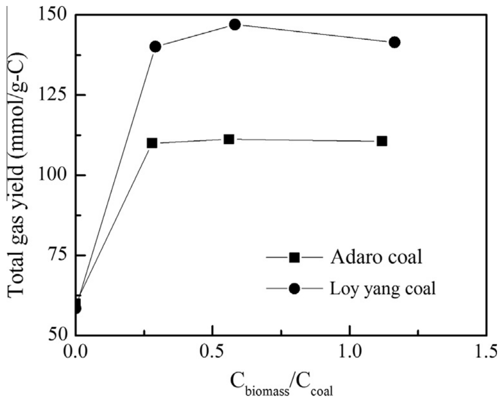

Fig. 6. Gas yields of two different coals when they were gasified with seaweed in various blending ratios at 750 °C.

4. Conclusions

Cogasification of low rank coal with 3 kinds of biomass was carried out in a fixed bed downdraft reactor. It is found that the coal gasification rate was promoted by the addition of biomass. However, the extent of promoting effect was different when different biomass was used for the cogasification. AAEM content in the biomass played an important role in this synergy effect. Biomass with high content of AAEM promoted the gas production from coal significantly. The structures of coal as well as coal char also had some influence on the promoting effect. At lower gasification temperature, the extent of promoting effect was enhanced by the adding more biomass into the coal, However, at higher temperature, a smaller amount of biomass could have great promoting effect on the coal gasification.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.fuel.2014.06.008.

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