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Gasification and Co-gasification Low-rank Coal with Biomass

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Abstract. Recently, there has been significant research interest in cogasification of coal and various types of biomass blends to improve biomass gasification and syngas production. In addition, ash present in biomass catalyses the gasification of coal. This experiment was conducted on the cogasification of various types of coal and biomass using drop tube gasifier under two sets temperatures 1173 and 1273K respectively. Most of the reactions are considered as endothermic, the heat input is needed to conduct the reactions. The additions of biomass to NL coal during co-gasification give no effect on gasification, since the reaction is endothermic. The increasing temperature from 1173 to 1273K only give small increase in the yield gas and efficiency due to the short of residence time. **Keywords:** Gasification, Co-gasification, Low-rank Coal, Biomass

Introduction

Gasification has been considered for many years as an alternative to combustion of solid or liquid fuels. The gasification process can produce clean gaseous mixtures by gasification technique. The clean gas can be used in internal combustion-based power plants.

Electricity generation from renewable sources such as biomass becomes interesting due to higher conversion efficiency and low-cost. The gasification technologies are also available for the biomass to produce the gaseous fuel. For the requirement to reduce the CO_2 content of the global atmosphere, on the other hand biomass is recognized as one of the suitable fuel.

There are several experimental gasification systems available, which are mainly developed for either coal or biomass wastes gasification, but there is not much information available about co-gasification of coal mixed with biomass or wastes. The results obtained by Reinoso *et al.* in two fluidized bed plants of 1.7 and 2 MW (Reinoso *et al.*, 1995) showed that the gasification was improved by addition of wood wastes to the coals. Other authors also reported that co-gasification of biomass with coal did not lead to any serious operational problems (Mc Lendon *et al.*, 2004).

In this experiment, pure CO_2 is used as the gasification agent to elucidate fundamentals on the co-gasification characteristics. The objective of this study is to elucidate co-gasification characteristics of biomass with coal, comparing the results for only biomass or coal gasification.

Materials and Methods

The experiments of gasification and co-gasification of several types of biomass and coals are also conducted, using an electrically heated drop tube furnace (DTF) as shown in Figure 3.1 in chapter 3. Principally, the procedure for the gasification experiments by using the drop tube furnace is almost similar to the combustion tests. Coal and biomass samples are injected from the fuel feeder in the upper parts of the furnace system. Pure CO₂ gas used as the gasification agent is supplied from a CO₂ cylinder. The CO₂ flow rate is maintained constant at 1.08 l/min. In order to maintain the similar condition, the fuel feed rate is adjusted regarding to the ratio of carbon in CO₂ to in the sample of 1.2. The experiments are conducted for around 60 minutes for each set of experiment. The temperature is maintained constant for two set of 1173 and 1273K. Residue, tars, soot and condensable liquids carried by the gas are removed by using a thimble filter and two

impingers before introducing a gas analyser. The weight of residue for each experiment is determined by weighing the filter before and after the experiments. The sampling probe is water-cooled and is quenched and diluted by argon gas at the probe tip to prevent additional chemical reactions and water-vapour condensation through the sampling line. The reaction gas is diluted at three times. The produced gas such N₂, CO₂, CO, O₂, H₂, CH₄ and several light hydrocarbons gases are measured continuously by a micro GC. Rigaku (TAS 300) thermo gravimetric analyser (TGA) and Yanaco CHN coder are also used to determine the carbon conversion of each fuel.

Two coals and one sawdust are gasified as a sample. Their proximate and ultimate analyses and other relevant properties are shown in Table 1. This table shows that the biomass contains a high VM content and a small amount of ash. The NL coal has the greatest FC content and highest fuel ratio, comparing to TH coal. The experimental conditions of the co-gasification test are shown in Table 2. The furnace temperature and the ratio of carbon in CO_2 to samples are maintained at 1073 K and 1.2, respectively by adjusting the feed rate, respectively. Pure CO_2 is used as the gasification agent. The flow rate is kept at 1.08 l/min in all experiments. Pyrolysis experiments in nitrogen atmosphere are also conducted to discuss the effect of gasification.

Proximate and ultimate analysis of sample							
Sample				H. Sawdust	NL coal+H. Sawdust	NL coal	TH coal
Proximate analysis	Moisture		air dry	11	6.4	1.8	5.2
	VM	wt%		86.6	56.8	27	45.4
	FC	WC /0		1.2	27.7	54.2	37.6
	Ash			1.2	9.1	17	11.8
Fuel ratio				0.01	2.051	0.498	1.207
Ultimate analysis	С		air dry	55.87	66.07	76.26	66.9
	Н			7.06	3.81	0.56	6.6
	Ν	wt%		0.24	1.25	2.26	0.68
	S			0	0.125	0.25	0.04
	0			36.83	28.75	20.67	25.82
H/C mol ratio			1.52	0.058	0.007	1.52	
Combustible			87.8	84.5	81.2	83	
Carbon in residue			49.05	55.82	61.92	55.53	

Table 2 Experimental conditions during Gasification and Co-gasification

Sample	Hinoki sawdust	H. sawd	lust + NL coal	NL coal	TH coal
Temperature [K]			1173, 1273		
Atmosphere			CO ₂		
C(CO ₂)/C(sample) ratio			1.2		
Sample feed rate [g/min]	0.96	0.73		0.69	0.84
CO ₂ flow rate (total)			1.08		
-Primary			0.23		
-Secondary			0.85		
Dilution with Ar (Sampling)			3 times		
Flow in sampling probe	0.49	0.43		0.46	0.47
Sampling distance [mm] Sample gas			1200 CO ₂ , CO, H ₂ , CH ₄ , C	кНу	

Results and Discussion Gasification and co-gasification characteristics

In gasification where the gasifying agent is CO_2 and the products of gasification are combustible gases like Carbon monoxide (CO), Hydrogen (H₂) and traces of Methane and non-useful products like tar and dust, the production of these gases is by reaction of water vapour from fuel itself and carbon dioxide through a glowing layer of charcoal. Thus the key to gasifier design is to create conditions such that fuels (biomass and coal) are reduced to charcoal and then charcoal is converted at suitable temperature to produce CO, H₂, CH₄ and other light hydrocarbons. Three distinct processes take place in gasification by using CO_2 as gasifying agent. They are drying of fuel, pyrolysis which a process in which tar and other volatiles are driven off, and reduction.

It is desirable to use fuel with low moisture content because heat loss due to its evaporation before gasification is considerable and the heat budget of the gasification reaction is impaired (Davidson, 1997). Thus in order to reduce the moisture content of fuel some pretreatment of fuel is required.

Gasification begins with pyrolysis giving a solid residue; char, consisting mainly of carbon and ash, a gas phase and condensable phase consisting of tar and water. The products of gasification and gasifying agent (water, uncombusted partially cracked pyrolysis products and carbon dioxide) react with charcoal where the following reduction reactions take place (Mizutani, 2002).

Boudouard		
$C + CO_2 = 2CO$	(+ 164.9 MJ/kg mole)	(1)
Water - gas		
$C + H_2 O = CO + H_2$	(+ 122.6 MJ/kg mole)	(2)
Water – gas shift		
$CO + H_2O = CO + H_2$	(+ 42.3 MJ/kg mole)	(3)
Methanation		
$C + 2H_2 = CH_4$	(+ 75 MJ/kg mole)	(4)
Steam reforming		
$CH_4 + H_2O = CO + 3H_2$	(+226.6 MJ/kg mole)(5)	
Dry reforming		
$CH_4 + CO_2 = 2CO + 2H_2$		(6)

Reactions (1) and (2) are main reduction reactions (main gasification reaction) and being endothermic has the capability of reducing gas temperature. Reaction 4 only occurs for high hydrogen concentration. Consequently the temperatures in the reduction zone are normally 800-1000°C. Temperature in the gasification was supplied by 3 (three) sets electric heaters and maintain isothermal for 2 (two) sets temperature at 1173 and 1273K. The heat input was used for the reaction above to convert fuel carbon to combustible gas.

In order to elucidate the gasification process, gasification experiments for all cases were also conducted at 1173 and 1273K as shown in Figure 1 (a) and (b). The figures show CO_2 and another combustible gas is produced such as CO, H_2 , CH_4 and C_2H_4 . If we compare Figure 1 (a) and 1 (b) we can observe the summary effect of increasing temperature on combustible gas produced. Reaction temperature is one of the most important variables in the gasification process, since the main gasification reactions are endothermic. When temperature rises it also increases the rate of several reactions as mention earlier. Corté et al., (1987) studied the influence of temperature on fast pyrolysis of beech wood in an electrically heated furnace. These authors found that the optimum temperature range for producing fuel gas is about 800 to 900°C; above this temperature, both the hydrocarbon yield and the heating value decrease. The higher temperature range of 900 to 1000°C, however, is an optimum for the production of synthesis gas that is mainly composed of a mixture of H_2 and CO. Figure 5.9 and 5.11 represent the summary effect of temperature on gasification and co-gasification for all cases. From the figures it was also detected a decrease in CO_2 concentration with the rise of temperature, which may be explained as a result of CO_2 consumption by boudouard reaction (1) and dry reforming reaction of CH_4 (6). The water – shift gas reaction (3) would produce CO_2 , but results obtained by Gil *et al.*, 1999 also seem to indicate that when temperature increase the CO₂ consuming reactions

would be more important than the shift reaction. On the higher temperature the CO produced for co-gasification of NL coal and Hinoki sawdust biomass increases significantly. The highest conversion of combustible gas produced is also observed on TH coal gasification.

The TH coal particle is smaller compared to the Hinoki sawdust and the volatile matter content is also high. This result suggests that TH coal surface area is larger than Hinoki sawdust make the conversion of TH coal is higher than Hinoki sawdust biomass. The reactivity of many types of coal and gasifying agent during gasification under atmospheric pressure has been examined by several authors [8-9].



Figure 1. Gasification and Co-gasification results at 1173K and 1273K

A Similar tendency on increasing of CO production with decreasing of CO_2 is also shows on TH coal profile for 1173 and 1273K. An increase in temperature, from 1173 to 1273K, led to a decrease in methane and other hydrocarbons concentration, while hydrogen concentration increased. The results obtained agree well to those of literature and work conducted by Kim *et al.*, 2001 that also detected an increase in H₂ and a decrease in CH₄ contents, when the temperature of coal gasification increased. Gil *et al.*, 1999 studied pine wood chips gasification in a pilot scale installation also observed an increase in H₂ and a decrease in hydrocarbons contents with the rise of temperature. Co-gasification of biomass and plastic mixtures (Pinto *et al.*, 2002) also verified the favourable influence of temperature in promoting the formation of H₂.

Cold gas efficiency

A cold-gas efficiency is used to evaluate the gasification performance. The cold-gas efficiency, η_{cg} , is defined as the percentage of the fuel heating value converted into the heating value of the product gas.

The cold gas efficiency (Li et al, 2002) is defined as

$$\eta_{cg} = \frac{[H_l]_{product}}{[H_l]_{sample}} x100\%$$
⁽⁷⁾

The lower heating value of product $[H_l]_{product}$ is determined by the concentration of CO, H_2 , CH_4 and C_2H_4 in the product gas mixture with the equation

$$\begin{split} H_l &= H_h - V_s = 10.79 [H_2] + 12.63 [CO] + 35.79 [CH_4] - 1.96 [H_2O] + 59.03 [C_2H_4] \ (8) \\ \text{Where high heating value of product gas mixture } [H_h] is determined by the equation} \\ H_h &= 12.75 [H_2] + 12.63 [CO] + 39.72 [CH_4] + 62.95 [C_2H_4] \ (9) \\ \text{The lower heating value } [H_l]_{sample} \text{ of sample is determine by equation} \\ H_l &= H_h - 2.44 (8.94h) \ (10) \\ \text{Where high heating value of input sample is determined by the equation} \end{split}$$

$$H_{h} = 33.8c + 144.3[(h-o)/7.94] + 9.425s$$
⁽¹¹⁾

Figure 2 (a) and (b) show cold gas efficiencies of gasification and co-gasification of NL and TH coals and Hinoki sawdust biomass at 1173 and 1273K respectively. Figure 2(a) shows cold gas efficiency of Hinoki sawdust is the highest among others, while NL coal is the lowest. This result because during gasification Hinoki sawdust produces high gas yields since the volatile matter in the biomass is high and easily evolves even in low temperature. The increase of hydrocarbon also increases high heating value. By adding biomass to coal in co-gasification case, it can be observed that the efficiency also increases, but the increase is average of cumulative of NL coal and Hinoki sawdust. This mean the co-gasification is not affected by biomass addition. Gasification of TH coal higher than NL coal, since the volatile matter is high and more reactive.



Figure 2. Cold gas efficiency during Gasification and Co-gasification at 1173K and 1273K. Where c, h, o and s are elemental analysis of fuels in weight percentage [wt%]. Higher or lower heating value of product gas and fuels sample was in (MJ/kg)

The effect of temperature on the increase of cold gas efficiency can be seen by comparing Figure 2 (a) and (b). The significant increase of the efficiencies when temperature increase to 1273K was observed for NL and TH coal and co-fuel cases. For Hinoki sawdust biomass gasification increasing of temperature from 1173 to 1273K give no effect on cold gas efficiency, since the volatile matter in the biomass is completely evolve at 1173K. However, for coal case, the structure in which C, H and O are bound in the fuels more aromatic molecular structures (pyridinic, pyrrolic) (Jukka *et al.*, 1995), which is difficult to decompose. Biomass consists basically of cellulose, hemi-cellulose and lignin (Vamvuka *et al.*, 2003). This leads to different behaviour during flash pyrolysis, which is together with drying the initial step in the fluidized bed gasification process (Pinto et al, 2003). This different behaviour results in a different yield of initial products. At higher temperature the conversion of carbon to gas yields for NL and TH coal is increasing significantly. However, since the residence time is very short, the efficiencies are still low. The differences in fuel type also effects on gas heating value and as a result it will also effect on cold gas efficiency (Van der Drift *et al.*, 2001).

Conclusions

Gasification of different fuels with CO_2 has been conducted by drop tube gasifier under two sets temperatures 1173 and 1273K respectively. Most of the reactions are considered as endothermic, the heat input is needed to conduct the reactions. The use of CO_2 as gasifying agent in these experiments were subjected for the use of exhaust gas from coal-fired power plant which contains heat energy and has an advantages in reducing CO_2 emission in the atmosphere. Biomass gasification show higher gasification efficiency than NL coal, since the biomass contains high volatile matter and easily evolves even in low temperature. The additions of biomass to NL coal during co-gasification give no effect on gasification, since the reaction is endothermic. The result of TH coal at higher temperature higher than hinoki sawdust biomass due to the particle of TH coal is smaller than Hinoki sawdust biomass. Since the residence time of gasification and co-gasification conducted by drop tube gasifier in these experiments is 2 seconds for each sample. As a results, the cold gas efficiency, CO conversion and carbon conversion is low. The increasing temperature from 1173 to 1273K only give small increase in the yield gas and efficiency due to the short of residence time.

References

Corté P., Hérault V., Castillo S., Traverse J.P. 1987. High-temperature gasification of carbonaceous materials by flash pyrolysis: thermal aspects. Fuel, 66(8): p.1107-1114.

Davison, R. 1997. Co processing waste with coal, IEA Coal Research.

- Gil J., Miguel A., Caballero, Juan A., Martín, María-Pilar A., Corella J. 1999. Biomass Gasification with Air in a Fluidized Bed: Effect of the In-Bed Use of Dolomite under Different Operation Conditions. Industrial Engineering Chemical Resources, 38(11): p.4226-35.
- Kim Y.J., Lee S.H, Kim S.D. 2001. Coal gasification characteristics in a downer reactor. Fuel. 80(13): p.1915-1922.
- Leppälahti J., Koljonen T. 1995. Nitrogen evolution from coal, peat and wood during gasification: Literature review. Fuel Processing Technology, 43(1): p.1-45.
- Li X.T., Grace J.R., Lim C.J., Watkinson A.P., Chen H.P., Kim J.R. 2004. Biomass gasification in a circulating fluidized bed. Biomass and Bioenergy, vol. 26(2): p.171-193.
- Mc Lendon T.R., Lui A.P., Pineault R.L., Beer S.K., and Richardson S.W. 2004. High pressure co-gasification of coal and biomass in a fluidized bed. Biomass and Bioenergy, 26 (4): p.377-388.
- Miura K., Hashimoto K., Silveston P.L. 1989. Factors affecting the reactivity of coal chars during gasification and indices representing reactivity. Fuel, 68(11): p.1461-1475.
- Mizutani Y. 2002. Combustion Engineering, 3rd ed. (In Japanese). Morikita Press, Tokyo, Japan.
- Pinto F., Franco C., Andre R.N., Miranda M., Gulyurtlu I., Cabrita I. 2002. Co-gasification study of biomass mixed with plastic wastes. Fuel, 81(3): p.291-297.
- Pinto F., Franco C., Andre R.N., Tavares C., Dias M., Gulyurtlu I., Cabrita I. 2003. Effect of experimental conditions on co-gasification of coal, biomass and plastics wastes with air/steam mixtures in a fluidized bed system. Fuel, 82(15-17): p.1967–1976.
- Reinoso C., Cuevas A., Janssen K., Morris M., Lassing K., Nilsson T., et al. 1995. Clean coal technology programme. In: Bemtgen JM et al, editor. University of Atuttgarrt Paper C5, vol. 3, 1995, p.1-45.
- Takarada T., Tamai Y., Tomita A. 1985. Reactivities of 34 coals under steam gasification. Fuel, 64(10): p.1438-1442.
- Vamvuka E., Karakas E., Kastanaki P., Grammelis. 2003. Pyrolysis characteristics and kinetics of biomass residuals mixtures with lignite. Fuel, 82(15-17): p.1949-1960.
- Van der Drift A., Van Doorn J., Vermeulen J.W. 2001. Ten residual biomass fuels for circulating fluidized-bed gasification. Biomass and Bioenergy, 20(1): p.45-46.