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Steam Gasification of an Australian Bituminous Coal in a Fluidized Bed

Woon-Jae Lee†, Sang-Done Kim* and Byung-Ho Song**

Energy/Coal and Chemical Process Research Team, Research Institute of Industrial Science and Technology,
Pohang 790-330, Korea
*Department of Chemical Engineering and Energy & Environment Research Center,
Korea Advanced Institute of Science and Technology, Daejeon 305-701, Korea
**Department of Chemical Engineering, Kunsan National University, Kunsan 573-701, Korea
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Abstract−To produce low calorific value gas, Australian coal has been gasified with air and steam in a fluidized bed reactor (0.1 m-I.D×1.6 m-high) at atmospheric pressure. The effects of fluidizing gas velocity (2-5 U_f/U_{mf}), reaction temperature (750-900 °C), air/coal ratio (1.6-3.2), and steam/coal ratio (0.63-1.26) on gas composition, gas yield, gas calorific value of the product gas and carbon conversion have been determined. The calorific value and yield of the product gas, cold gas efficiency, and carbon conversion increase with increasing fluidization gas velocity and reaction temperature. With increasing air/coal ratio, carbon conversion, cold gas efficiency and yield of the product gas increase, but the calorific value of the product gas decreases. When steam/coal ratio is increased, cold gas efficiency, yield and calorific value of the product gas increase, but carbon conversion is little changed. Unburned carbon fraction of cyclone fine decreases with increasing fluidization gas velocity, reaction temperature and air/coal ratio, but is nearly constant with increasing steam/coal ratio. Overall carbon conversion decreases with increasing fluidization velocity and air/coal ratio, but increases with increasing reaction temperature. The particle entrainment rate increases with increasing fluidization velocity, but decreases with increasing reaction temperature.

Key words: Fluidized Bed, Steam Gasification, Coal

INTRODUCTION

Conversion of coal to gaseous fuel has been envisioned as a major contribution to the energy picture in the near future and several feasible conversion processes have been proposed. Currently, a great deal of effort is being expended to improve the performance of the existing coal gasification processes [Saffer and Ferrell, 1988; Watkinson et al., 1983; Gutierrez and Watkinson, 1982] and to develop new types of gasifiers for processing larger volume of coal [Kim et al., 2000; Riley and Judd, 1987]. Catalysts have been tested to reduce operating temperture [Lee et al., 2001; Lee and Kim, 1995; Song and Kim, 1993]. However, there are many complicated problems to solve, such as an insufficient information on fundamental knowledge of coals, their reactivity, and the reaction kinetics under the actual gasification conditions, and the technological difficulties involved in handling, preparation and feeding of coal, the collection and removal of ash. Of the many contacting devices proposed for coal gasification, fluidized beds are widely used due to their advantages such as ease of controlling bed temperature, ease of solids handling, possibility of treating wide range of coal particles of different size, and ease of turn down. However, fluidized beds also have several disadvantages: limitation of temperature range due to ash melt, carbon loss by bed drain and entrainment of fine particles, and treatment of cohesive coal [Saffer et al., 1988; Watkinson et al., 1983; Purdy et al., 1981]. The Winkler process is the oldest among the fluidized bed gasifiers and it was the originator of the development of fluidization. Although there are many types of gasifiers which are in use or under development, fluidization in the presence of water vapor and oxygen remains a basic technology for coal gasification. Further study is needed for a fluidized bed coal gasifier to overcome the problems involved and improve reactor performance.

The present study is to carry out steam gasification of coal in a fluidized bed to produce low calorific value gas. The effects of fluidization gas velocity, bed temperature, air/coal ratio, and steam/coal ratio have been determined not only on the composition, yield and calorific value of the product gas but also on the cold gas efficiency and carbon conversion.

EXPERIMENTAL

A schematic diagram of the fluidized bed reactor (0.1 m-i.d.×1.6 m height) is shown in Fig. 1. This gasifier is made of stainless steel columns and consists of the following parts: feeding part of air and steam, gas plenum, main column, freeboard, coal feeder, cyclone, product gas cleaning, and gas sampling and analysis.

Distilled water is fed into the steam generator (1 kW) through a 'Masterflex' pump. Steam generated is mixed with compressed air and introduced to the gas plenum. The inside of the gas plenum is filled with inert material to improve the mixing of air and steam, and an electric heater (2 kW) is installed on the plenum wall to preheat the feed gas mixture. A bubble cap gas distributor having seven caps was situated between the main column and gas plenum for the feed gas to be evenly distributed through the bed. Main reactor column is 0.1 m-i.d.×0.85 m high and is expanded into larger column (0.2 m-i.d.×0.6 m high) to reduce the entrainment of bed par-

†To whom correspondence should be addressed.
E-mail: wjlee@risnet.rist.re.kr
‡This paper is dedicated to Professor Dong Sup Doh on the occasion of his retirement from Korea University.

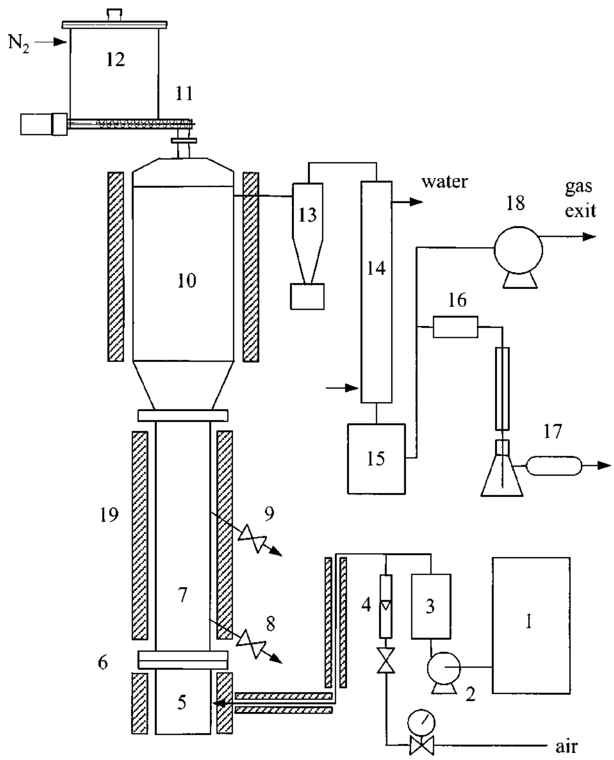

Fig. 1. Schematic diagram of the fluidized bed gasifier.

1. Water reservoir
2. Masterflex pump
3. Steam generator
4. Air flow meter
5. Gas plenum
6. Gas distributor
7. Main bed
8. Bed drain
9. Overflow
10. Freeboard
11. Screw feeder
12. Coal hopper
13. Cyclone
14. Condenser
15. Collector
16. Dust filter
17. Gas sampling bottle
18. I.D fan
19. Heater

ticle. An electric heater of 6 kW was installed at the reactor wall to heat the bed to the ignition temperature of coal (around 500 °C). After that, the reactor was insulated by 'Kao-wool'. One sight glass was mounted at 0.5 m above the gas distributor and an ash drain port at the bottom, and an overflow drain port at 0.45 m above the distributor. To measure pressure drop along the bed, eight pressure taps were mounted flush on the reactor wall at 0.1 m height intervals above the distributor. Also six K-type thermocouples were mounted along the bed to measure the axial temperature profile. The coal was fed into the top of the reactor through a screw feeder, where the feed rate of coal was controlled by a variable-speed DC motor. The product gas is cooled down passing cyclone, condenser and a dust collector. A small amount of product gas is sampled at the outlet of dust collector and its composition is determined by a gas chromatograph (HP 5890 II) with a thermal conductivity detector with 'Molecular sieve 5A' and 'Porapak Q' columns.

At the beginning of the experimental run, the electric heater is turned on to heat up the bed of sand particles (d_p=0.27 mm) with feeding only air. When the bed temperature reaches 450-500 °C (ignition temperature of coal), the coal is fed into the reactor and then the bed temperature will rise rapidly due to coal combustion. When the temperature reachs a desired level, steam is introduced into the air stream. It usually takes about 40 min for the reactor to reach its steady state. At the steady condition, some amount of the product gas samples is taken and the amount of collected particles in the cyclone collector is measured. The reactor operating variables could be changed by adjusting air/coal and steam/coal feed ratio. The Australian bituminous coal used in this study is 0.25-1 mm in diameter and its properties is given in Table 1.

Table 1. Analyses of the Australian bituminous coal

	wt%		wt%
Ultimate analysis (daf base)		Ash composition	
C	72.6	SiO_2	60.2
H	5.4	Al_2O_3	26.8
N	2.6	Fe_2O_3	7.1
S	0.4	CaO	0.4
O	19.0	MgO	0.2
		TiO_2	1.4
Proximate analysis (dry base)		K_2O	1.7
Volatile	29.1	MnO	0.1
Fixed carbon	56.0		
Ash	14.9		
Heating value (kcal/kg)	6141		

RESULTS AND DISCUSSION

The reaction temperature is one of the most important operating variables affecting the performance of the gasifier since the main reaction C-H_2O is endothermic. Hence reaction temperature should

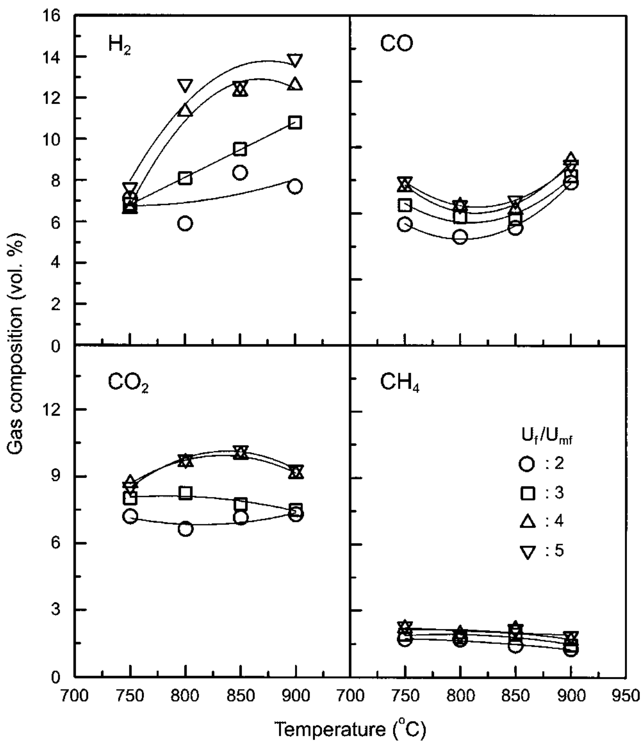

Fig. 2. Effect of temperature on product gas compositions at different fluidization gas velocity.

be at the highest tolerable level. Material of construction, ash fusion and production of undesirable gases such as NO_x will limit the reactor temperature. In the present work the material problem restricted maximum gasification temperature to 900 °C. This is relatively low temperature for carbon-steam reaction and may limit the calorific value of product gas. The effect of reaction temperature on the product gas composition at various fluidizing gas velocity is shown in Fig. 2. It can be seen from the plot that H_2 content increases with increasing reaction temperature, CO shows minimum value with temperature, and CO_2 increases with temperature at first but decreases at high temperatures. Whereas CH_4 content remains almost constant. At lower temperature, the concentration of CO and that of CO_2 in the product gas may be determined through the competition among the fast coal combustion, water gas shift reaction and the steam gasification reaction. The contents of H_2, CO, CO_2, and CH_4 are found to increase with increasing fluidization gas velocity. The apparent reaction rate of coal can be enhanced by the better solids mixing within fluidized bed from an increase of fluidization velocity.

The effect of reaction temperature on carbon conversion and gas yield (nitrogen free basis) at different fluidization gas velocity is shown in Fig. 3. Carbon conversion is defined as the ratio of carbon in CO, CO_2 and CH_4 of the product gas to carbon in feed coal. Carbon conversion increases with increasing fluidizing velocity and reaction temperature [Watkinson et al., 1987] due to the increase of reaction rate of steam-carbon and carbon-oxygen [Lee, 1987]. Also, it can be seen from the plot that gas yield increases with increasing fluidizing velocity and reaction temperature [Herguido et al., 1992; Saffer et al., 1988]. The increase of gas yield with fluidizing gas velocity is due to an enhancement in coal reactivity by more efficient solid mixing caused by the increased gas velocity.

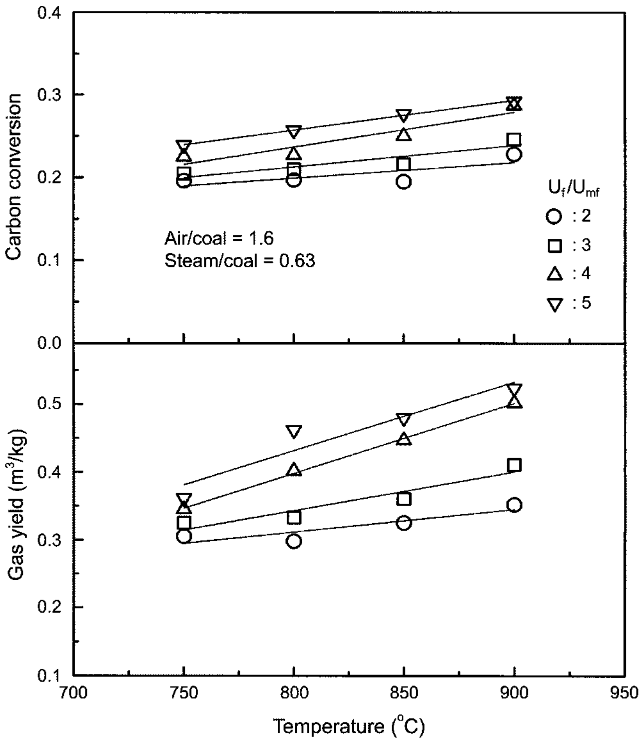

Fig. 3. Effect of temperature on carbon conversion and yield of the product gas at different fluidization gas velocity.

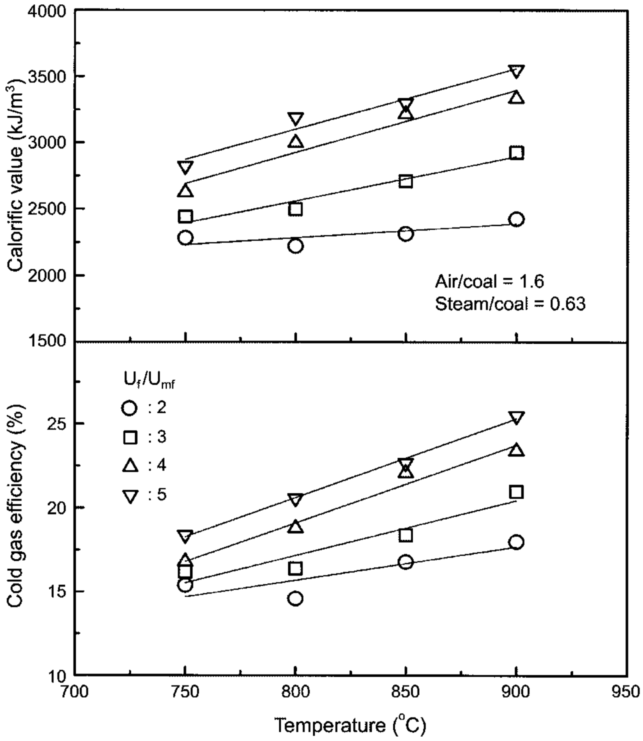

Fig. 4. Effect of temperature on calorific value of the product gas and cold gas efficiency at different fluidization gas velocity.

On the other hand, the increase in both the coal feeding and reactant gas can cause larger gas yield. Also, the gas yield increases with reaction temperature due to the increase of gas production from initial pyrolysis by rapid heating, char-steam reaction, and coal combustion reaction.

The effect of reaction temperature on the gas calorific value and cold gas efficiency is shown in Fig. 4. As can be seen in this plot, the calorific value of the product gas increases with increasing reaction temperature and fluidization gas velocity due to the increase of H_2 and CO productions caused by the increase of gasification reaction and the increase of CH_4 production by rapid pyrolysis [Foong et al., 1980, 1981]. In this plot, cold gas efficiency, defined as the ratio of product of the gas heating value and the gas production rate to the heating value of the coal fed, increases with increasing reaction temperature and fluidization gas velocity.

The effect of air/coal ratio in the feed on product gas compositions at 850 °C and steam/coal ratio of 0.63 is shown in Fig. 5. As can be seen in this plot, the volume percentage of H_2, CO and CH_4 decrease, but that of CO_2 increases with increasing air/coal ratio. When air/coal ratio increases, the corresponding increase of oxygen in the feed gas gives high rate of C-O_2 and the product of CO_2 increases. In this case, the decrease of H_2 and CH_4 concentrations may be caused by the increase of combustion of H_2 and CH_4 by oxygen with the increase in air feed rate. The increase of CO_2 content comes from the proportional loss of H_2 and CH_4 as well as from combustion of carbon and CO to produce CO_2 [Sue-A-Quan et al., 1991; Foong et al., 1981].

The effect of air/coal ratio in the feed on carbon conversion and yield of the product gas at 850 °C and steam/coal ratio of 0.63 is shown in Fig. 6. It can be seen that carbon conversion and yield of

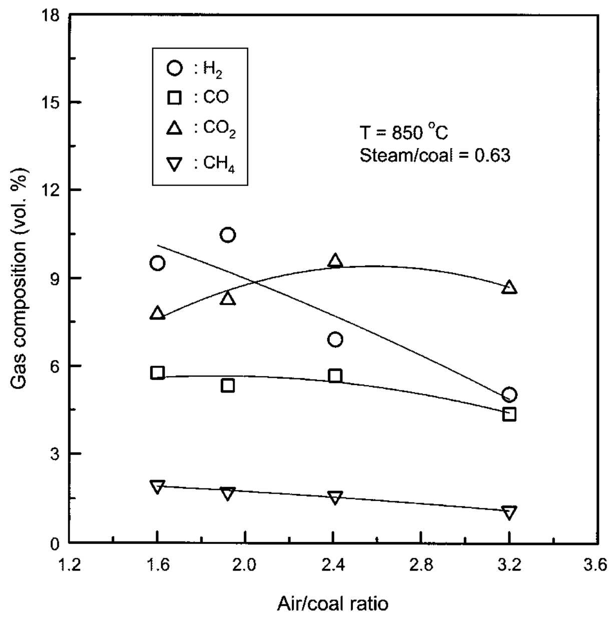

Fig. 5. Effect of air/coal ratio on the product gas compositions.

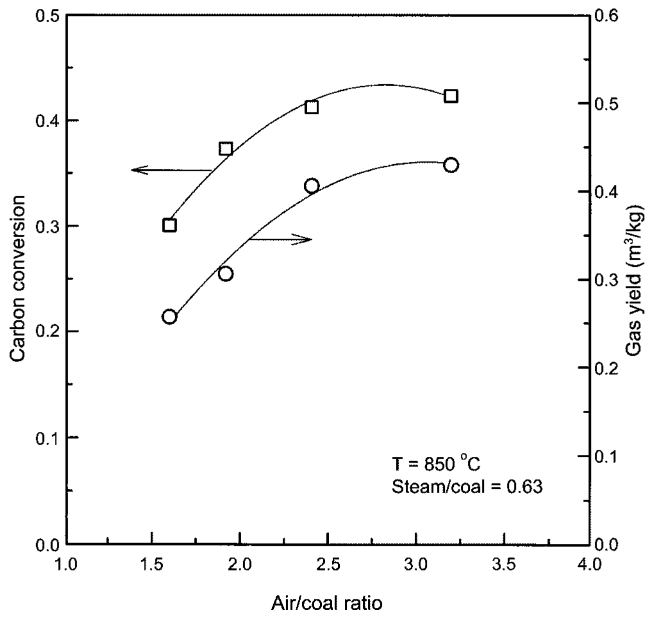

Fig. 6. Effect of air/coal ratio on carbon conversion and yield of the product gas.

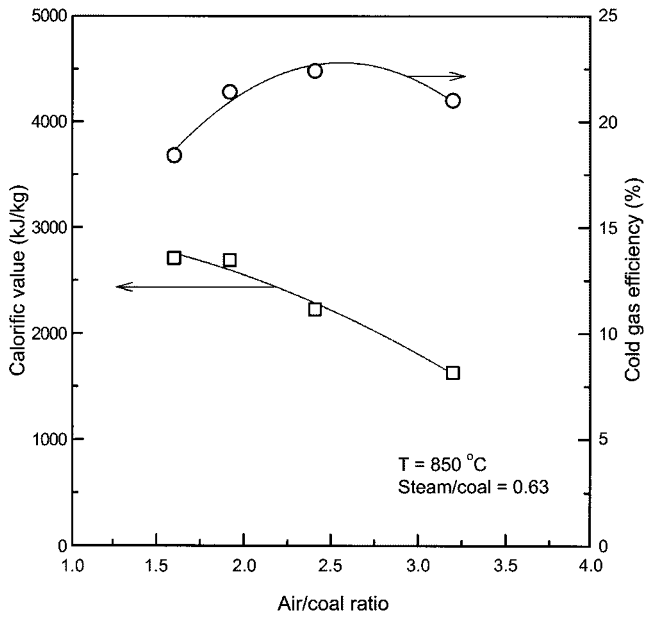

Fig. 7. Effect of air/coal ratio on calorific value of the product and cold gas efficiency.

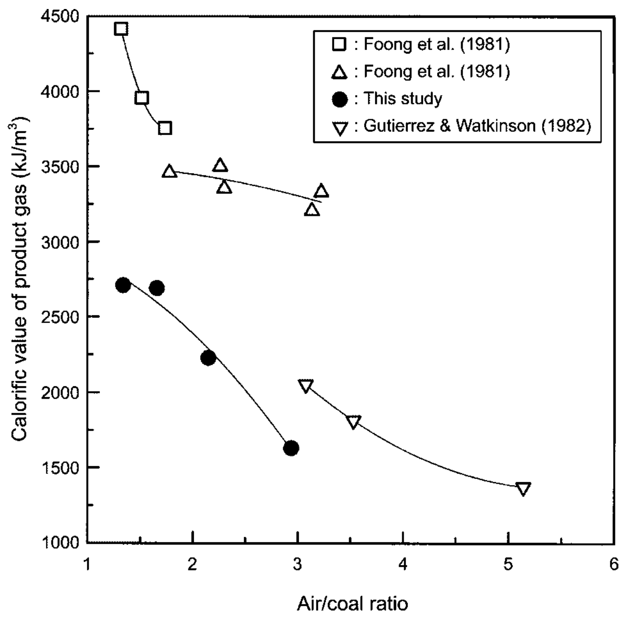

Fig. 8. Calorific value of the product gas from the several studies of steam gasification of coal in fluidized beds.

the product gas increase with increasing air/coal ratio. This may due to the enhancement of coal combustion. The effect of air/coal ratio on calorific value of the product gas and cold gas efficiency is presented in Fig. 7. In this plot, calorific value of the product gas decreases with increasing air/coal ratio due to the re-combustion of H_2, CH_4, and CO by excess oxygen and the dilution of the product gas by nitrogen in air feed. However, cold gas efficiency is slightly increases with increasing air/coal ratio.

The changes of calorific value of the product gas from the present and previous study [Gutierrez and Watkinson, 1982] with variation of air/coal ratio are shown in Fig. 8. The calorific value of product gas decreases with increasing air/coal ratio because of reduction of H_2, CO and CH_4 and enhancement of coal combustion. The trend of calorific value in this study seems to be similar to that in the experiment of Gutierrez and Watkinson [1982] with variation of air/coal ratio. They used somewhat large air/coal ratio compared to others.

The effect of steam/coal ratio on the product gas composition at 850 °C and air/coal ratio of 1.6 is shown in Fig. 9. As can be seen in this plot, H_2 and CO_2 contents of product gas increase with increasing steam/coal ratio. On the other hand, CO slightly decreases and CH_4 remains approximately constant [Gutierrez and Watkinson, 1982] with increasing steam/coal ratio. This result seems to

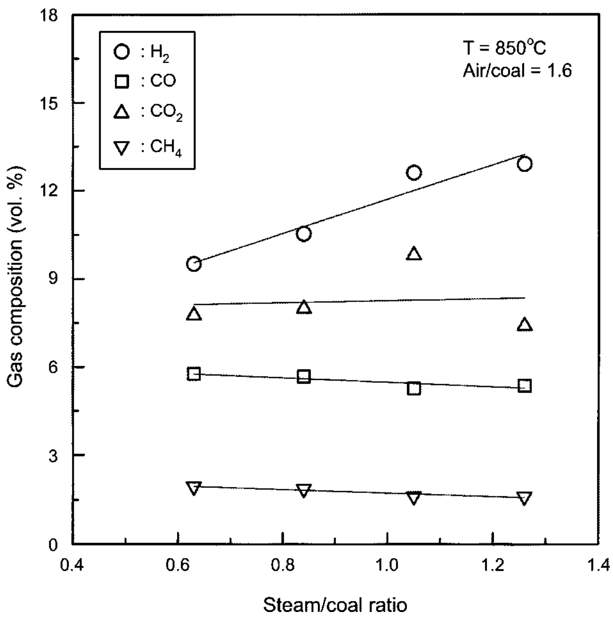

Fig. 9. Effect of steam/coal ratio on the product gas composition.

suggest that the water gas shift reaction favors higher concentration of water in the reactor; $CO+H_2O \leftrightarrow CO_2+H_2$ compared to the carbon-steam reaction; $C+H_2O \leftrightarrow H_2+CO$. Such an increase in hydrogen content enhances calorific value of the product gas from 2.5 to 2.8 MJ/m^3.

Particle entrainment plays important role in the performance of fluidized bed reactor since it causes carbon loss from the reactor. Many factors affect particle entrainment rate: gas velocity, bubble properties, particle size distribution, and bed diameter etc. [Geldart, 1986]. The effect of fluidization gas velocity on the total entrainment rate at various bed temperatures is shown in Fig. 10. It can be seen in this plot that the total entrainment rate of small particles increases with increasing fluidizing gas velocity since the bubble eruption on the bed surface and the drag force acting on the particles within the freeboard increase with increasing fluidizing velocity [Lee and Kim, 1992]. The entrainment rate is known to increase with increasing gas velocity to the 2.0 power [Geldart, 1986]. As can be seen in this plot, the total entrainment rate decreases with increasing bed temperature due to the reduction of drag force acting on the particles within the freeboard.

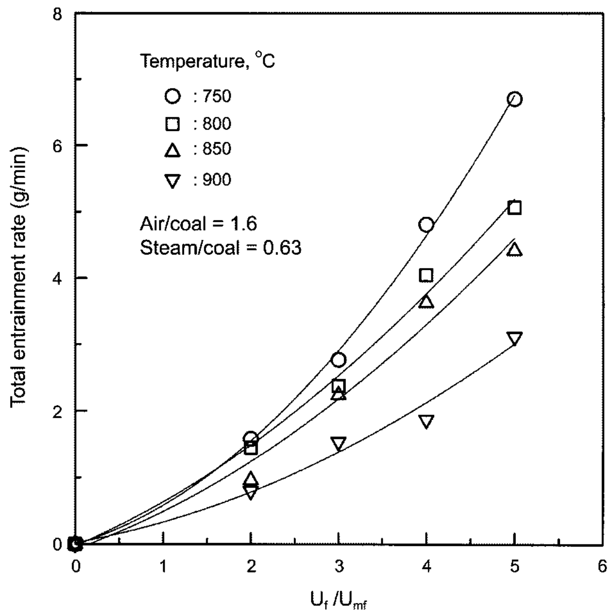

Fig. 10. Effect of fluidization gas velocity on the total entrainment rate.

Table 2. Unburned carbon fraction and overall efficiency of carbon at various operating conditions

Temp. (°C)	U_f/U_{mf}	Air/coal	Steam/coal	UCF	η_{eff}*	H_2/CO
750	2	1.6	0.63	0.817	84.6	1.29
	3	1.6	0.63	0.802	82.2	1.07
	4	1.6	0.63	0.818	76.3	0.87
	5	1.6	0.63	0.697	77.6	1.03
800	2	1.6	0.63	0.804	85.2	1.27
	3	1.6	0.63	0.809	83.2	1.39
	4	1.6	0.63	0.742	81.2	0.76
	5	1.6	0.63	0.649	87.9	1.99
850	2	1.6	0.63	0.799	89.8	1.56
	3	1.6	0.63	0.763	85.0	1.65
	3	1.92	0.63	0.797	85.9	2.04
	3	2.41	0.63	0.771	85.2	1.54
	3	3.20	0.63	0.742	83.1	1.15
	3	1.6	0.84	0.795	83.2	1.86
	3	1.6	1.05	0.691	86.3	1.96
	3	1.6	1.26	0.793	85.0	2.41
	4	1.6	0.63	0.767	81.6	2.00
	5	1.6	0.63	0.683	84.1	1.91
900	2	1.6	0.63	0.777	91.4	1.04
	3	1.6	0.63	0.777	89.0	1.40
	4	1.6	0.63	0.768	90.1	1.50
	5	1.6	0.63	0.777	86.7	1.70

*overall efficiency of carbon

The effects of operating parameters - fluidization gas velocity, bed temperature, air/coal ratio, and steam/coal ratio on unburned carbon fraction of cyclone fine (UCF), overall efficiency, and H_2/CO ratio - are summarized in Table 2. UCF is defined as the fraction of carbon in the fine particles captured in cyclone. Overall efficiency of carbon is calculated from the ratio of carbon in the captured particles at cyclone to carbon in feed coal particles. UCF decreases with increasing fluidization velocity and reaction temperature due to the corresponding enhancement of coal reactivity. Overall efficiency increases with bed temperature, but it decreases with fluidization gas velocity due to the carbon loss by entrainment of small particles in the fluidized bed. When air/coal ratio is increased, UCF and overall efficiency decrease due to the increase of coal reactivity and particle entrainment, respectively. However, UCF and overall conversion are little changed with variation of steam/coal ratio.

On the other hand, carbon conversion is defined as the ratio of carbon in product gas to carbon in feed coal. The carbon conversion could be influenced by various operating parameters. To investigate the relation between carbon conversion and operating param-

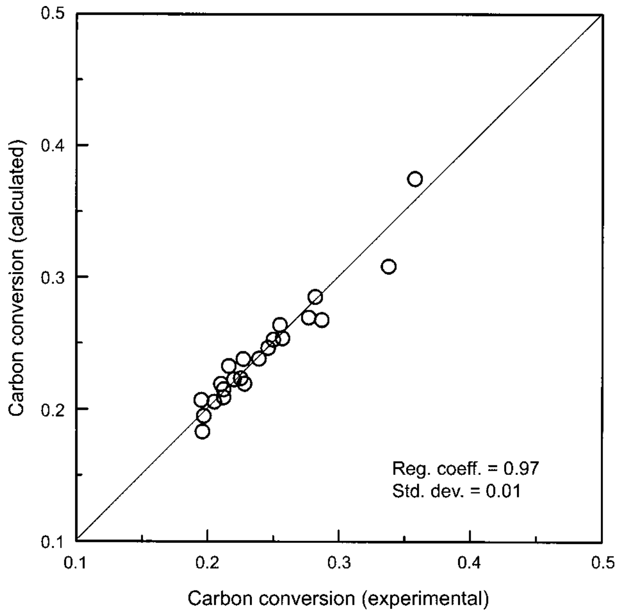

Fig. 11. Comparison of carbon conversion between the measured and calculated values.

eters used in this study, we correlated carbon conversion with dimensionless groups as

$$C.C = 0.02\left(\frac{U_f}{U_{mf}}\right)^{0.29}\left(\frac{T_{bed}}{298}\right)^{1.32}\left(\frac{F_{air}}{F_{coal}}\right)^{0.69}\left(\frac{F_{steam}}{F_{coal}}\right)^{-0.15}$$

where the obtained regression coefficient was 0.97 and standard deviation 0.01.

Comparison of carbon conversion between the calculated and experimental values is plotted in Fig. 11.

CONCLUSIONS

In the present experiment of steam gasification of a bituminous coal, carbon conversion, cold gas efficiency, gas yield and calorific value of the product gas increase with increasing reaction temperature and fluidizing velocity. With increasing air/coal ratio, the calorific value decreases, but carbon conversion, cold gas efficiency, and the gas yield increase. With increasing steam/coal ratio, the calorific value and the gas yield increase, but carbon conversion is scarcely changed. Unburned carbon fraction of cyclone fine decreases with increasing reaction temperature, fluidization gas velocity, and air/coal ratio. Overall carbon conversion increases with increasing reaction temperature, but it decreases with increasing fluidization gas velocity. Particle entrainment rate increases with increasing fluidization gas velocity, but it decreases with increasing bed temperature.

ACKNOWLEDGMENTS

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NOMENCLATURE

C.C. : carbon conversion [-]
T_{bed} : reaction temperature or bed temperature [K]
U_f : fluidization gas velocity [m/s]
U_{mf} : minimum fluidization velocity [m/s]
UCF : unburned carbon fraction [-]
F_{air} : feed rate of air [kg/s]
F_{coal} : feed rate of coal [kg/s]
F_{steam} : feed rate of steam [kg/s]
η_{eff} : overall efficiency of carbon calculated from cyclone capture [%]

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