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Devolatilization characteristics of high volatile coal in a wire mesh reactor

Ho Won Ra*,, Myung Won Seo*, Sang Jun Yoon*, Sung Min Yoon*, Jae Kwon Kim*,
Jae Goo Lee*,†, and Seung Bin Park****

*Clean Fuel Department, Korea Institute of Energy Research (KIER), 152, Gajeong-ro, Yuseong-gu, Daejeon 305-343, Korea
**Department of Chemical and Biomolecular Engineering, Korea Advanced Institute of Science and Technology (KAIST),
291, Daehak-ro, Yuseong-gu, Daejeon 305-701, Korea

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Abstract−A wire mesh reactor was used to investigate the devolatilization process of coal particle during entrained flow gasification. Coal from Indonesia East Kalimantan mine, which has high moisture and high volatile matter, was chosen as a sample. Experiments were carried out at the heating rate of 1,000 °C/s and isothermal condition was kept at peak temperature under atmospheric pressure. The char, tar and gas formation characteristics of the coal as well as the composition of the gas components at peak temperatures were determined. The experimental results showed that devolatilization process terminated when temperature reached above 1,100 °C. Most of tar was formed at about 800 °C, while the rate of tar formation decreased gradually as the temperature increased. CH_4 was observed at temperatures above 600 °C, whereas H_2 was detected above 1,000 °C. The amount of formed gases such as H_2, CO, CH_4 and C_nH_m increased as the temperature increased. From the characteristics of devolatilization with residence time, it was concluded that devolatilization terminated within about 0.7 second when the temperature reached 1,000 °C. As the operating temperature in an entrained flow gasifier is higher than ash melting temperature, it is expected that the devolatilization time of high volatile coal should be less than one second in an entrained flow gasifier.

Keywords: Devolatilization, Wire-mesh Reactor, Coal, Char, Gaseous Species

INTRODUCTION

Coal is a reliable and abundant source for primary energy now and it will continue to serve as an important energy resource in the future. Coal gasification is in the spotlight as an environmentally friendly technology for heat, power generation, gaseous fuels and chemical products. To increase gasifier efficiency and improve the accuracy of plant simulation, it is necessary to obtain the gas-solids reaction data from laboratory scale unit similar to the actual gasifying conditions. These experimental results will provide a better understanding of the devolatilization and gasification process and be instrumental for optimizing the operating condition and developing an advanced type of gasifier [1-4]. Devolatilization is the first step of coal gasification process, and numerous substances such as char, tar, gas and water are formed. Because the chemical properties of coal are different with the source and rank of coal, many previous researchers reported studies related with coal devolatilization since the 1960's [5,6]. The wire mesh reactor (WMR) was first developed by Loison and Cahuvin in France in 1964 [7]. Because it can be operated at a high heating rate (up to 5,000 °C/s), at high temperatures (up to 2,000 °C), and at high pressure (up to 150 bar), it has been widely used for research on devolatilization and gasification. Research groups at many institutions around the world, such as MIT [8,9] and Princeton in the US [10,11], Munchen University in Germany [1,2], the London Imperial College [12,13,19] in the UK, GE Global Research-Shanghai in China [3,17], and Monash University in Australia [5,14,15], have all conducted studies using WMR reactors. So it is not surprising that tremendous advances have been made for the past 40 years not only in heating method and temperature control systems, but also in reactor design. In particular, the Kandiyoti Research Group of London Imperial College has made brilliant contributions to the furtherance of WMR technology over the past 20 years [12,13,16,19]. However, while most of researchers mentioned about char or volatile yields, there were few studies on the product distribution and gas composition in WMR. For these reasons, our objective was to obtain the basic data of the devolatilization process in detail product distribution and gas compositions. To investigate the effects of temperature and residence time, the weight of char, tar and gas were all quantitatively measured and the low concentrations of each gas were analyzed. X-ray maps, FT-IR and SEM analysis were performed to investigate the structural changes during coal devolatilization with temperature.

EXPERIMENTAL

1. Coal Sample

The coal used in the experiment was imported from a mine in East Kalimantan, Indonesia that is used by power plants for generating power. The coal can be classified as high volatile B bituminous coal according to ASTM D388. The proximate and ultimate analyses of the coal used in the experiment are shown in Table 1. The coal drying was conducted at 105 °C with N_2 conditions. After drying, sample coal was pulverized with pin-mill and screened with sieve shaker. The particle size of sample coal was kept at the range

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[†] To whom correspondence should be addressed.
E-mail: jaegoo@kier.re.kr

Table 1. Proximate, ultimate analysis and caloric values of the samples

Samples	Indonesian coal
Proximate analysis (wt%, as received)	
Moisture	13.66
Volatile matter	44.28
Fixed Carbon	36.97
Ash	5.1
Ultimate analysis (wt%, daf)	
C	71.13
H	5.04
O*	14.85
N	1.68
S	1.38
Calorific value (kcal/kg)	6,812

*Calculated by difference

of 125-150 µm to avoid the loss from entrainment with carrier gas [11,16,19]. Fig. 1 shows SEM photograph of wire mesh and coal particles used in the experiment.

2. Devolatilization Experiment

Fig. 2 illustrates the configuration of the WMR used in this experiment. The WMR consists of a power supply for the wire mesh reactor, a mass flow controller for supplying reaction gas, a main reactor, and a device for collecting reaction gases and tar. The main reactor was made of copper to improve electrical conductivity. There is a 30 mm hole in the middle of the reactor for gas sweeping. The sweep gas is supplied from the bottom of the reactor toward the mesh. To ensure that the power can be supplied only to the coal loaded on the mesh, each power supply wire is insulated with a mica plate. The power is supplied to the main reactor from each power supply that can control temperatures by varying the voltage, current and duration time. Voltage can be controlled at intervals of 0.1 V, and duration time can be controlled at intervals of 0.1 s. Maximum volt-

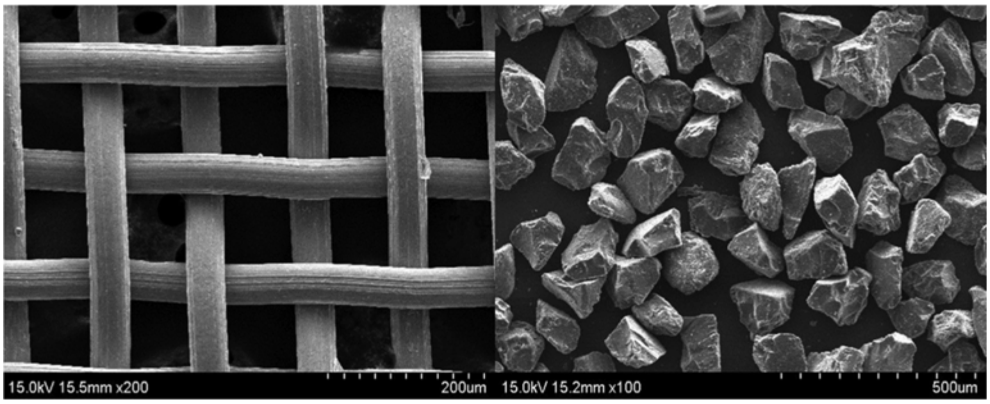


Fig. 1. Photograph of the wire mesh and coal sample.

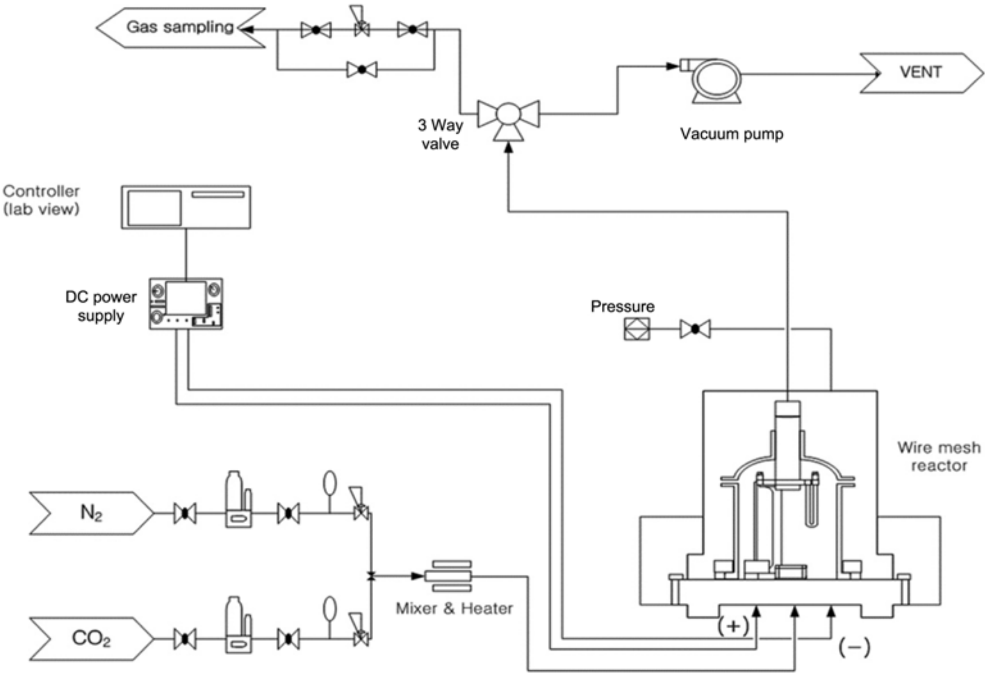


Fig. 2. Schematic diagram of the wire mesh reactor.

age is 20 V with maximum current being 1,000 A. Temperatures of coal particles loaded on the mesh are measured by a K type (Chromel/Alumel) thermocouple with high response time. The width of thermocouple is 0.05 mm. To get the heating rate and peak temperature under the same conditions, the power settings (voltage, current and duration time) are varied and temperatures are monitored with the thermocouple. In this manner, the power settings for each temperature are determined to find the most suitable combinations. If the power settings are the same, the temperature is confirmed to be the same through repeated measurements [16].

In general, small quantities of coal samples (10 mg or less) are sandwiched as a single layer between two layers of mesh. The mesh is placed between two electrodes with power supplied to them to bring the entire assembly to a high temperature very quickly. At this time, the mesh itself serves as a resistance heater. During the heating, the products of devolatilization are transported from the reaction zone using carrier gases (inert gases like helium and nitrogen) with flow rate of 3.0 L/min. The sweep gas velocity was calculated as 0.08 m/s at this condition. Power was halted when the maximum required temperature and target residence time at maximum temperature were reached. The samples were then cooled by the carrier gas flow. The quantity of coal that remained was then weighed and then that weight was compared to the weight of the sample prior to the test. For accurate measurement, a high-precision scale with a resolution of 0.01 mg was used (Mettler Toledo, Model XP205). To evaluate the influence of temperature at the heating rate of 1,000 °C/s from room temperature, the experiment was conducted between 400 °C and 1,100 °C. Also, to evaluate the influence of the holding time, the holding time was changed from 1 s to 10 s. Fig. 3 shows a photograph of the wire mesh reactor used in the experiment operating at 1,000 °C.

3. Analysis of the Tar and Gas

The experiment was conducted so that the tar and gas formed during devolatilization would leave the mesh surface as the devolatilization due to the inert gas supplied from the bottom of the reactor took place. The tar was collected with a quartz collector. The inside of the collector was cooled by injecting liquid nitrogen during the devolatilization. The outside diameter of the collector was 32 mm; the inside diameter was 28 mm. An 8 mm diameter tube was installed inside the collector. Quartz wool was put into the 8 mm diameter tube; immediately before the experiment, liquid nitrogen was used to quickly cool the collected tar from the reactor. After the experiment was over, the tar collector was separated and stored in a desiccator at room temperature with both ends sealed. The difference between the weight of the tar collector measured before the experiment and that measured after the experiment was used to measure the weight of the tar formed.

Before the reaction started, a 3-way valve was used to purge the gas; when power was supplied to the mesh, the direction of the valve was automatically switched by the control program to supply gas to the sample bag. A gas chromatographer (HP 7890) with two detectors, a flame ionization detector (FID), and a thermal conductivity detector (TCD) were then used to analyze the components of the gas collected. As the coal loaded into the wire mesh reactor weighed considerably less, the quantity of formed gas was small. Also, to reduce the secondary reaction of the tar and gas formed during the devolatilization process, the concentration of the gas formed due to the supply of the carrier gas was kept low. For these reasons, conventional FID and TCD led to many errors in measurement. To reduce errors in the measurement of the gas, a gas chromatograph (HP 7890) with a methanizer was used. About 100 μl of gas formed during the devolatilization was collected in a sample bag using a syringe; gas was then analyzed. The unionized substances (CO and CO_2) in the gas were methanized busing the methanation catalyst; these substances were then measured with the FID.

RESULTS AND DISCUSSION

1. Effects of Mesh Material and Mesh Area

The mesh served as a resistor in the wire mesh reactor. During the reaction, the temperature characteristics were determined according to the material, area of the mesh, heating rate, and the maxi-

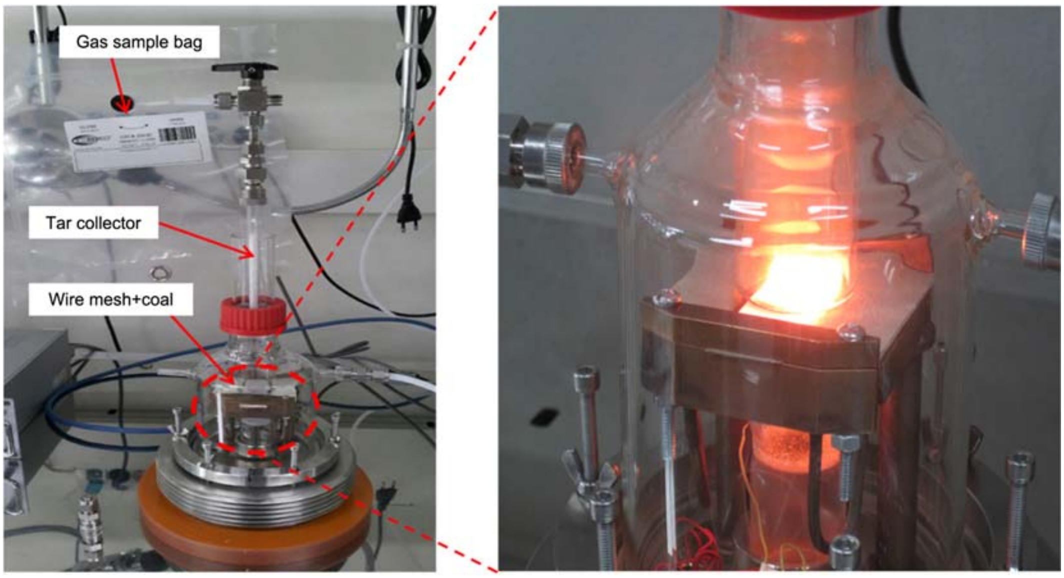


Fig. 3. Photograph of the WMR operating at 1,000 °C.

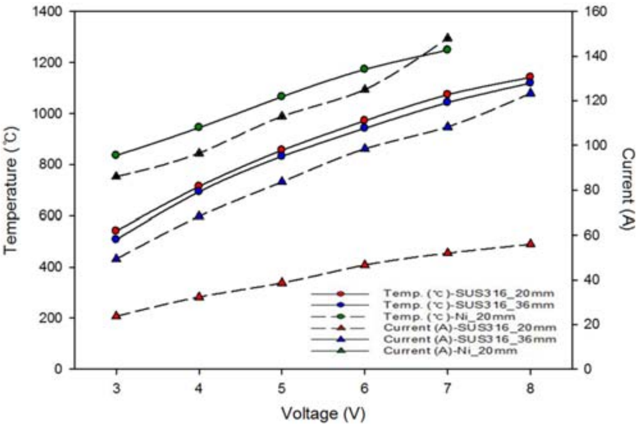


Fig. 4. Results of the temperature condition experiment according to the material and area of the mesh.

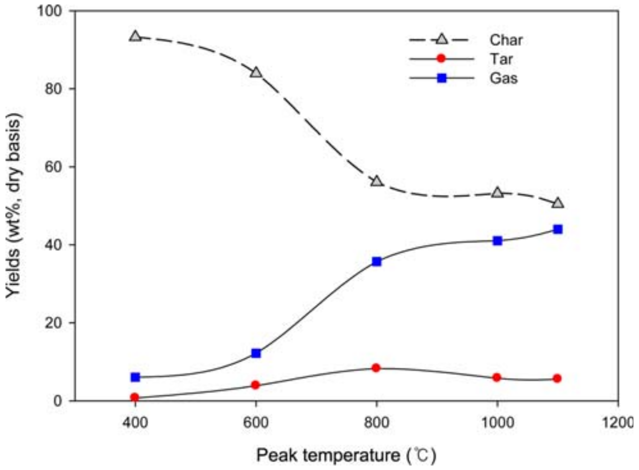


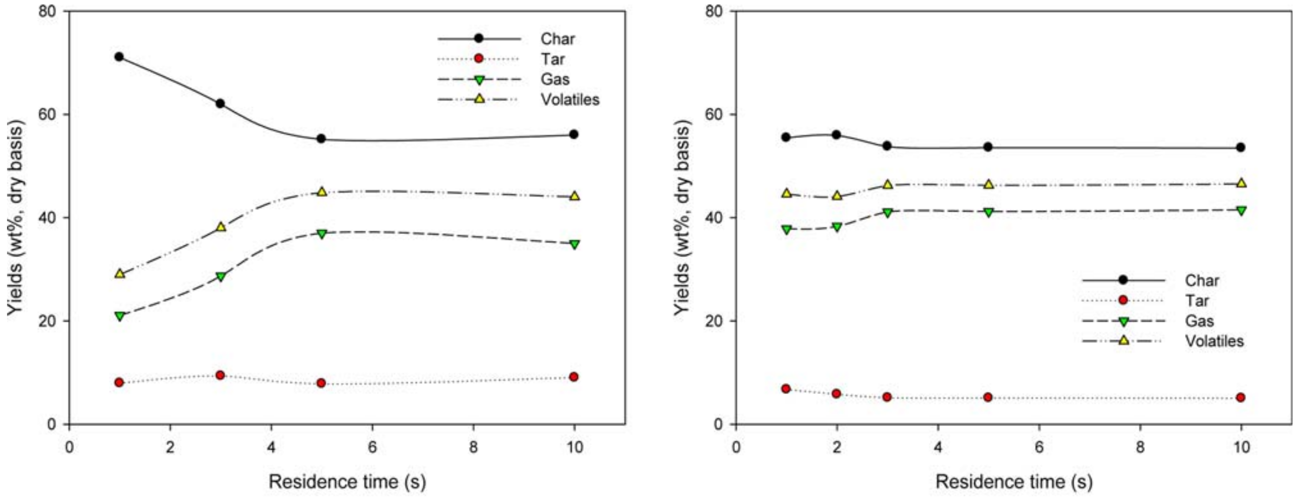
Fig. 5. Devolatilization characteristics according to peak temperatures.

mum operating temperature. The most widely used material for a WMR is SS 316L [8,10,14,16], while molybdenum wire mesh [17, 19] and Ni mesh [18] are also used. To understand the changes in temperature depending on the type and area of the mesh, both SS316L and Ni mesh were prepared for this experiment. Two sizes of mesh were used in this experiment, 20 mm×76 mm and 36 mm×76 mm. The measured temperature according to the material (SS316L, Ni mesh) and the area of the mesh are shown in Fig. 4. About 4.5 V of power was supplied to an Ni mesh with a 20 mm width; it was confirmed that the mesh reached 1,000 °C with the current at about 105 A. As for an SS316L mesh with a 20 mm width, when about 6.2 V of power was supplied, the mesh reached 1,000 °C with the current measuring about 48 A. When the area of an SS316L mesh made of the same material was changed to 36 mm width and the temperature measured at 1,000 °C, the voltage was 6.7 V, with the current being about 104 A. It was determined that the maximum allowable temperature for an Ni mesh was about 1,250 °C, while for the SS316L mesh it was somewhat less, at about 1,150 °C. Thus, for the SS316L mesh, the difference in resistance was more than double when the area was increased by less than double. It can be concluded that to have a relatively low current is advantageous for experimental purposes.

If the current capacity increases, the thickness of the electric wire and the cost of the noise filter, circuit breakers of power supply would also increase. Furthermore, the size of the reactor and auxiliary equipment would also increase. If a device with a low current capacity is used, the cost for equipment configuration and the size of the equipment around the reactor could be down. Therefore, SS316L with a 20 mm width was used in this study. That is, if materials with a relatively small area and high resistance are used, the desired temperature will be reached at a lower current and voltage.

2. Effects of Peak Temperature and Residence Time

To conduct an experiment on the devolatilization characteristics at different devolatilization temperatures, the heating rate was set at 1,000 °C/s, with a holding time of 10 s. The devolatilization experiment was conducted with these parameters remaining constant while the peak temperature was changed. In an entrained flow gasifier, the temperature inside the reactor is higher than 1,400 °C, but as the



(a) (b)

Fig. 6. Devolatilization characteristics according to the residence time ((a) Peak temperature: 800 °C, (b) Peak temperature: 1,000 °C).

fixed-bed gasifier and fluidized-bed gasifier are operated at relatively lower temperatures, the experiment was conducted at temperatures between 400 °C and 1,000 °C. It is reported that the devolatilization starts at around 300 °C in the cases of low-rank coal and sub-bituminous coal [3]. Fig. 5 illustrates the variation of char, tar, and gas yield produced by high volatile coal devolatilization heated at temperatures between 400 °C and 1,100 °C. When this coal was heated to 400 °C, it yielded 93.2 wt% of char, 0.7 wt% of tar, and 6.1 wt% of gas. At 1,000 °C, the yield was about 53.2 wt% of char, 5.8 wt% of tar, and 41.0 wt% of gas. As the temperature increased, it was confirmed that the quantity of the char tended to decrease, and the weight loss began to slow above 800 °C. At about 800 °C, the greatest quantity of tar was formed; as the temperature increased, the amount of produced tar declined slightly. It is thought that the tar yield decreases because part of tar decomposes into a light gas above 800 °C. Similar results were confirmed in experiments conducted by other research groups [20,21]. The amount of gas increased slowly until the temperature reached 600 °C, but near 800 °C this amount of gas sharply increased. If the devolatilization temperature is higher than 800 °C, the changes in the quantity of char, tar, and gas were insignificant. It can be concluded that the devolatilization of coal was completed at a temperature above 800 °C.

Fig. 6 shows the devolatilization of coal with variance of the residence time. The yield of char, tar, and gas was measured while changing the residence time to 1, 2, 3, 5, and 10 s with the peak temperatures fixed at 800 °C and 1,000 °C. At 800 °C, as time elapsed, the yield changed; after about 4 s, it was confirmed that there was no change in yield of char, tar, and gas. At 1,000 °C, there was no change in yield with increasing the residence time. The above results, at temperatures lower than 1,000 °C, confirmed that a sufficient residence time was necessary for the coal devolatilization process.

3. Gas Analysis with Variance of Devolatilization Temperature

The gas analysis during the devolatilization at temperatures between 400 °C and 1,100 °C (residence time: 10 s) is shown in Fig. 7. As the temperature increased, the amount of gas increased. In detail, hydrogen was observed above 1,000 °C and CO was observed at 400 °C. It is known that as the devolatilization temperature increases, the hydrocarbons CO, CH_4, CO_2, and H_2 tended to increase [3,14]. However, as the temperature increased, CO, H_2, and CH_4 increased as expected, while the quantity of CO_2 and hydrocarbon peaked at about 800 °C and then decreased above 800 °C as shown in Fig. 7. Above 800 °C, the oxygen contained in the coal reacts to the carbon during the devolatilization process to generate CO_2. Around 1,000 K, the reverse Boudouard reaction ($C+CO_2 \rightarrow 2CO$) occurs. Therefore, the liberated CO_2 reacts with char generating additional CO. As a result, the concentration of CO_2 is lowered at above 800 °C. This result is attributed to differences in the structure of the coal used in this study. Tar, aliphatic gases, and hydrogen are closely related to the primary devolatilization process and are involved in the formation of CO_2, CH_4, and H_2O. According to Chen et al. [3], the temperature at which CH_4 was formed is about 550 °C, due to the methyl group; however, we confirmed that CH_4 was formed above 600 °C. The formation of CO during devolatilization is highly related to carboxyl linkage and ether linkage; the formation of H_2 is related to the formation of aromatic hydrogen. As such, the reaction requires high activation energy; it happens at a high temperature. We observed a tendency similar to that of H_2 formation.

4. Structural Analysis According to Devolatilization Conditions

Fig. 8 provides an SEM image of the raw coal and char after the devolatilization at temperatures of 600 °C and 1,000 °C; it also shows the results of the X-ray mapping analysis [22] (C, O, Al, and Si are presented in red, blue, green, and purple, respectively). From SEM image, the surface of the raw coal is smooth, whereas the surfaces of the char during devolatilization at 600 °C and 1,000 °C are rough with developed pores. According to the X-ray maps, the components were found to be evenly distributed for the raw coal; as the temperature rose during the devolatilization process, oxygen escaped, leaving only carbon to exist on the surface. Fig. 9 shows the results of the FT-IR spectra-analysis at different temperatures [23]. The main functional group was confirmed by the analysis and was marked accordingly. Various forms of oxygen functional groups and C-H bonds were observed. As the devolatilization temperature increased, the strength of the peak tended to decrease; above 1,000 °C, it was confirmed that all peaks disappeared. As confirmed by the mass

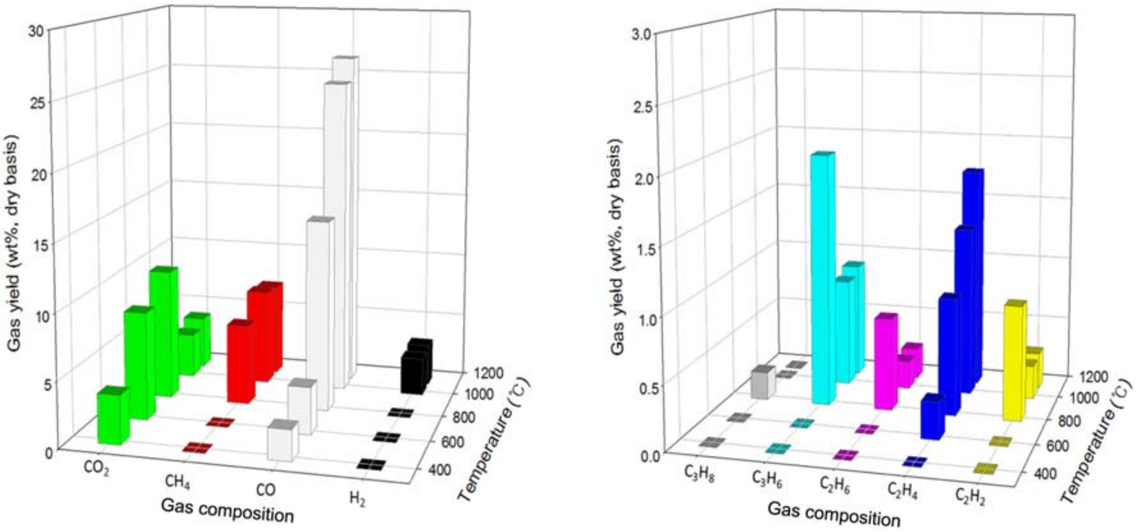


Fig. 7. Results of the gas analysis according to devolatilization conditions.

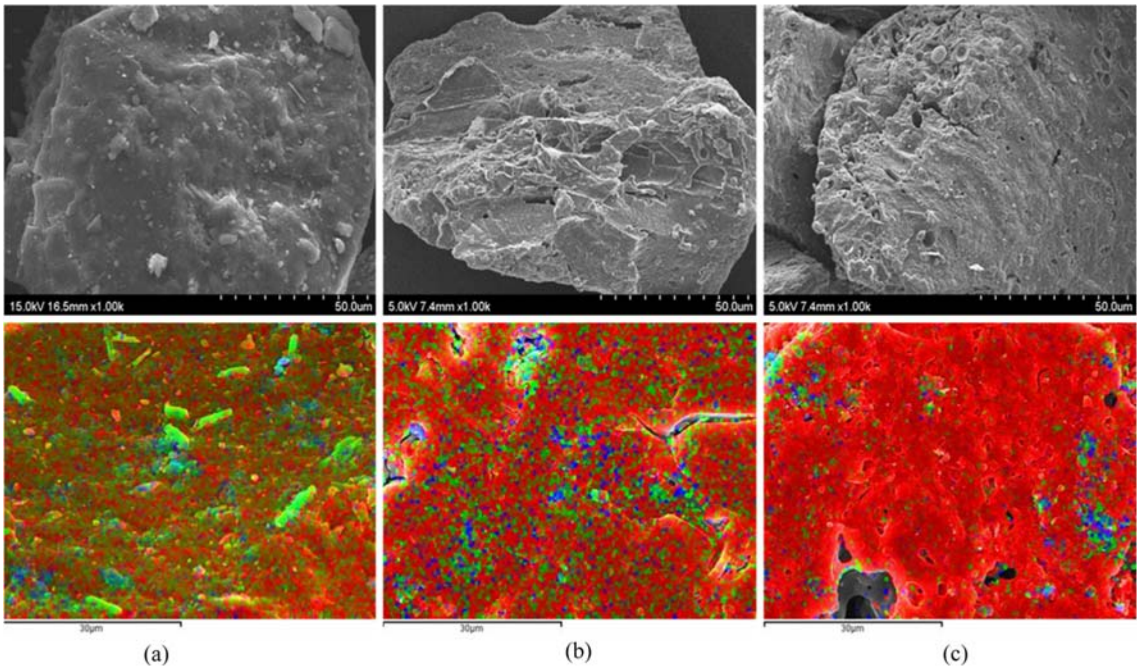


Fig. 8. SEM image and X-ray maps for major elements of the surface of Indonesian coal ((a) Raw coal, (b) Devolatilization temp: 600 °C, (c) Devolatilization temp: 1,000 °C; C, O, Al and Si are marked in red, blue, green, and purple, respectively).

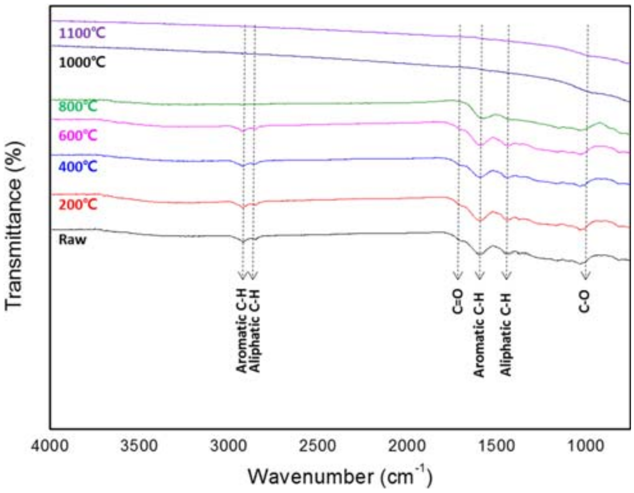


Fig. 9. Results of the FT-IR spectra analysis according to the devolatilization temperature of Indonesian coal.

balance, FTIR spectra, and X-ray maps, above 1,000 °C the devolatilization of high volatile coal was completed.

CONCLUSIONS

To understand the devolatilization characteristics of high volatile coal, which will be used at entrained flow gasification process currently under construction in Korea, an experiment was conducted using a wire mesh reactor to simulate devolatilization conditions between 400 °C and 1,100 °C. The results can be summarized as below.

1. The characteristics of the mesh, mainly the heating element in the wire mesh reactor, are such that it must have a relatively small area. When temperature is measured according to the material and area of the wire mesh, if the area of the mesh is 20 mm, a temperature of 1,000 °C could be obtained using 6.2 V and 48 A. The maximum allowable temperature of the material was about 1,250 °C for the Ni mesh, and 1,150 °C for the SS316L mesh.

2. A devolatilization experiment was conducted wherein the peak temperature and residence times remained constant while the peak temperatures were changed between 400 °C and 1,100 °C. As the peak temperature increased, the yield of the char tended to decrease. The yield of the char decreased from 93.2 wt% to 53.2 wt% with increasing temperature from 400 °C to 1,000 °C.

At temperatures lower than 1,000 °C, a sufficient residence time (4 s for 800 °C) was necessary for the coal devolatilization process. The peak temperature and residence time variables should be adjusted to change the devolatilization rate as desired.

3. The results of the analysis of the gasses generated between the reaction temperature of 400 °C and 1,100 °C are as follows: At 400 °C, a composition of CO at 2.3 wt% and CO_2 at 3.73 wt% was generated. As the temperature increased, CH_4 was generated starting from 600 °C and H_2 began to be detected after 1,000 °C. As the temperature increased, CO, CH_4 and H_2 all tended to increase. Hydrocarbon (CnHn) and CO_2 peaked at 800 °C and then decreased. The gas composition at 1,000 °C was H_2 at 2.92 wt%, CO at 24.57 wt%, CH_4 at 7.45 wt%, CO_2 at 3.44 wt%, C_2H_2 at 0.26 wt%, C_2H_4 at 1.33 wt%, C_2H_6 at 0.22 wt% and C_3H_6 at 0.85 wt%.

4. X-ray maps and FTIR Spectra analysis were conducted, and it was found that at a temperature above 1,000 °C, devolatilization was completed.

These experimental results obtained in a WMR can provide a basic understanding of the devolatilization process of high volatile coal and deliver optimum operating condition for coal entrained flow gasifier.

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