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Gasification of an Indonesian subbituminous coal in a pilot-scale coal gasification system

Yongseung Yun† and Seok Woo Chung

Plant Engineering Center, Institute for Advanced Engineering, Yongin 448-863, Korea
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Abstract−Indonesian Roto Middle subbituminous coal was gasified in a pilot-scale dry-feeding gasification system and the produced syngas was purified with hot gas filtering and by low temperature desulfurization to the quality that can be utilized as a feedstock for chemical conversion. Roto middle coal produced syngas that has a typical composition of 36-38% CO, 14-16% H_2, and 5-8% CO_2. Particulates in syngas were 99.8% removed by metal filters at the operating temperature condition of 200-250 °C. Sulfur containing compounds of H_2S and COS in syngas were also desulfurized in the Fe chelate system to yield less than 0.5 ppm level. The full stream gasification and syngas purifying system has been successfully operated and thus can provide clean syngas for the research on the conversion of syngas to chemicals like DME and on the future IGFC using fuel cells.

Key words: Gasification, Syngas, Subbituminous Coal, Desulfurization, Particulates Removal

INTRODUCTION

Interest in coal gasification has revived recently because of current high oil prices and the high probability of an oil production peak in 20 to 50 years. Gasification exhibits better performance than combustion in terms of the amount of pollutants generated and CO_2 as well as the nature of the energy carrier. While combustion converts S and N-containing compounds into SO_x and NO_x and carbon compounds mainly into CO_2, gasification instead produces H_2S and NH_3 that are more cleanly removable and the carbon compounds convert mainly as the syngas of H_2 and CO. The syngas can be utilized as basic materials for the C1 chemistry. Moreover, syngas is an energy carrier that contains heating value as a chemical energy, which means that energy content is maintained even if the flue gas temperature is lowered because energy would be liberated when combusted in the final appliances. Syngas also can be an energy source to a gas turbine and thus more efficient combined cycle systems are applicable. In contrast, CO_2 from a combustion system only contains energy as a hot gas and thus has a limitation to use mainly in steam turbines.

As widely known, coal can support the world energy system for the next 150-200 years at the current energy consumption rate. Furthermore, environmental concerns and recent issues on climate change mainly by CO_2 prompt the development of clean coal technology, especially since the year 2000. Among the many clean coal technologies, IGCC (Integrated Gasification Combined Cycle) is the most promising one. Advantages of IGCC are high environmental performance in that SO_x and NO_x can be reduced more than 80-90% compared to the conventional pulverized coal power plants, as well as the tendency of producing a high CO_2 concentration in the flue gas for carbon capture and sequestration.

Korea possesses low-grade anthracite only as a domestic energy source and imports 97% of its energy. With a diversification strategy of energy sources that is inevitable in Korea's situation, coal should remain as a key player in the energy sector through the 21st century although there are many technological and economical barriers to future coal usage. The main barrier resides in environmental concerns when fossil fuels including coal are utilized. Among technologies to cope with the ever-stringent environmental regulations, IGCC and USC (Ultra Supercritical Combustion) technologies are suggested by the IEA (International Energy Agency) Clean Coal Center as the most prominent choices [1].

The essence of gasification technology is that dirty feeds containing high levels of sulfur and ash such as coal, petroleum residues, and even municipal wastes can be utilized as a clean energy source with higher efficiency than conventional methods. Examples of final syngas application are fuel cells, ammonia production, and fertilizers.

If current concerns for the environment and energy get more severe, a technology shift from the combustion-based to gasification-based processes appears to be a natural trend in the long run. However, since the gasification-based process deals with explosive and toxic syngas, the complexity of equipment cannot be avoided. Thus, the most important deciding factor in commercialization would be how the process could be designed and constructed safely and reliably, which is the key factor to be solved if IGCC is to be widely accepted.

Gasification technology that has been developed as a core part of IGCC technology can be applied not only to coal but also wastes, petroleum coke and petroleum asphalt that were not considered as an energy source until recently. The Korean government has set a goal of replacing 5% of total electricity with new and renewable energy till 2011. With many technologies of solar power and wind power, etc. that are inherently not suitable for big scale power generation and also produce electricity with very high price, it's not enough to cope with the ever-increasing pressure to increase a portion of new and renewable energy with only these technologies.

In Korea, among clean coal technologies, IGCC has been selected as the next generation technology for coal power plants. Most likely, a commercial-scale 300 MW IGCC plant would start to oper-

†To whom correspondence should be addressed.
E-mail: ysyun@iae.re.kr
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ate after 2012. USC and PFBC (Pressurized Fluidized Bed Combustion) technologies would be the next option that can be tried in Korea when the commercial-scale plant is fully operational.

As a study to prepare the introduction of IGCC plants in Korea, a pilot-scale coal gasification plant has been operating since 1994. From the former studies using the pilot-scale dry-feeding coal gasification system, subbituminous coals have been determined as the best suitable coal due to their high reactivity and high content of volatile matter [2]. Among the tested subbituminous coals, Indonesian subbituminous coals were chosen as the best candidate coal for IGCC. Volatile matter contents for all tested Indonesian subbituminous coals were in the range of 43-45% and fixed carbon contents are 47-50 wt%. Gasification results using the same gasification system for other Indonesian subbituminous coals can be found elsewhere [3,4].

Another subbituminous coal from Alaska, USA, yielded poor performance in the pilot gasifier, mainly by clogging in the syngas cooler inlet. Low ash fusion temperature in the Alaskan Usibelli coal induced fly-slag clogging in the pipe just after the gasifier.

In this study, Indonesian Roto Middle subbituminous coal, which has a little lower quality than Indonesian Adaro coal, was gasified and sent to the hot gas filtering and the low temperature desulfurization system that was designed by in-house technologies in order to test the feasibility of providing cleaned syngas of particle-free and less than 1 ppm sulfur-containing trace gases.

EXPERIMENTAL

A dry-feeding type gasifier facility that is located at Ajou University in Suwon, Korea and can treat 3 ton/day at maximum 28 bar, 1,550 °C was built in April 1995. The facility (8 m×17 m×20 m) is located in a 30 m×50 m space. Main target feeds are subbituminous and bituminous coals. Coal feed is of identical size with that of conventional power plants using pulverized coal, as 80-90% passing −200 mesh. Pulverized coal is pneumatically conveyed with nitrogen gas in dense-phase into the feeding nozzle system, where 99%-purity oxygen and steam are mixed with the coal powder. Nitrogen that is employed for injecting coal powder into the gasifier was used at ambient temperature without auxiliary heating. Oxygen was heated just before being introduced into the gasifier with electric heaters to 150-200 °C. Steam is injected separately from the oxygen and coal powder, but the current study does not use any steam but only oxygen to control the temperature and the degree of conversion.

Normal operation consists of the preheating, pressurization, transient operation, normal gasification operation, and the shutdown steps. An LPG burner at the bottom of the gasifier did preheating of the gasifier for at least 20 hours. Then, nitrogen was introduced to pressurize the gasifier till the operating pressure range in less than 30 min; after this step, oxygen as well as coal powder was being fed into the gasifier. Coal supply was first started at the low feeding range in order not to cause any sudden pressure buildup in the gasifier and thus cause any back pressurization into the coal feeding lines. This step takes normally less than one hour. Normal hot test operation step for obtaining gasification data is maintained at steady state for at least 4 hours to provide enough gas, slag, and other process data.

Table 1. Analysis data of feed coal

	Item	Roto middle
Proximate analysis	Moisture	3.79
(as-received, wt%)	Volatile matter	42.95
	Fixed carbon	46.95
	Ash	6.31
Ultimate analysis	C	64.79
(moisture-free, wt%)	H	3.98
	O	23.37
	N	0.97
	S	0.33
	Ash	6.56
Ash initial deformation temperature	1,150 °C	
Higher heating value	6,000-6,200 kcal/kg	

Metal fiber filters were employed in the particulate filtering system, and a low-temperature desulfurization system based on the Fe chelate was used to remove H_2S compound. With further absorbing treatment with activated carbon or ion exchange resin, COS could be eliminated further.

The feed coal was an Indonesian subbituminous coal and the conventional analysis result is shown in Table 1. The coal was dried during the drying/pulverization step to less than 4% moisture content and this dried coal powder was used for gasification in the study. Roto Middle coal contains ash of 6.3-6.6 wt% when dried.

RESULTS AND DISCUSSION

Typical gasifier operating profiles are illustrated in Fig. 1, in that gasifier pressure is shown at the bottom line in the figure. The gasifier pressure was controlled at a constant 7.8 kg_f/cm^2 while the gasifier temperature was maintained above 1,350 °C that is required to melt the ash into molten slag. Temperature shown in the figure represents the actual temperature in the gasification zone just beside the coal feeding ports and thus exhibits some fluctuation according to the reacting coal powder with oxygen. In the commercial gasifiers, this temperature is not directly measured. Instead, the refractory temperature slightly away from the real hot gas temperature is typically measured. Syngas flow rate was maintained around 120 Nm^3/

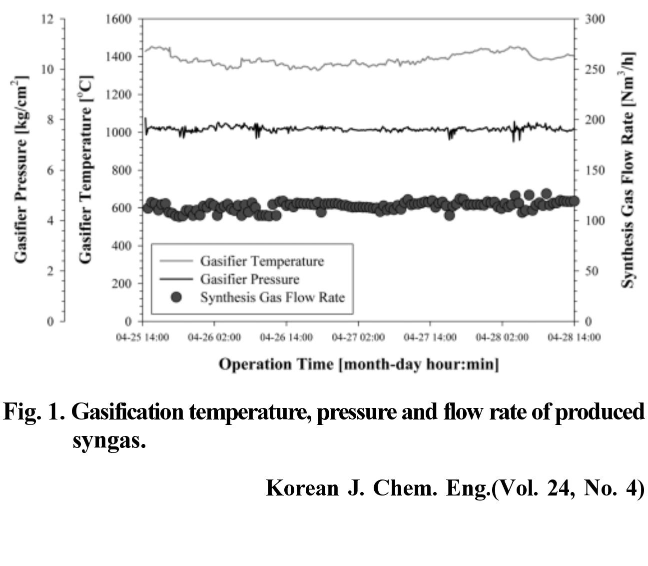

Fig. 1. Gasification temperature, pressure and flow rate of produced syngas.

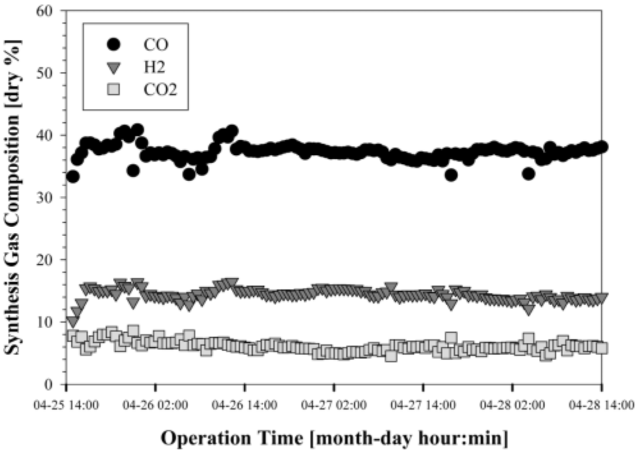

Fig. 2. Profiles of coal syngas composition for the Roto Middle coal.

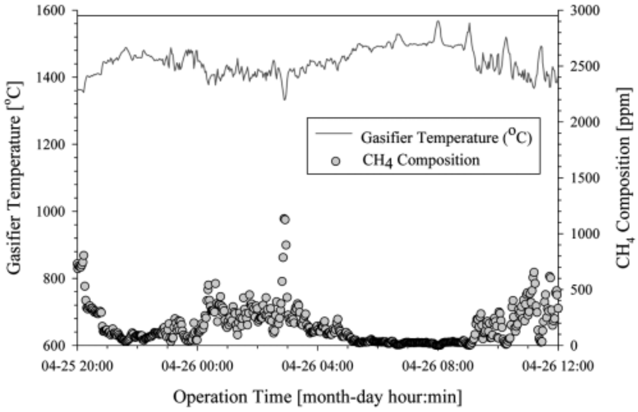

Fig. 3. Relationship between temperature inside the gasifier and methane content.

hr.

The produced coal syngas yielded about 36-38% CO, 14-16% H_2 and around 5-8% CO_2 concentrations (dry basis) as illustrated in Fig. 2. Fig. 2 shows the result of continuous operation for three days. The rest of the syngas composition is nitrogen. Normally in the pilot-scale dry-feeding gasifier, nitrogen that has been used for pneumatic coal conveying into the gasifier exists in the coal syngas. The necessary nitrogen amount for pneumatic feeding depends upon the size of the feeding nozzle, which is obviously bigger in the larger-scale gasifier. Because there's a tendency of particle blockage in the smaller-scale reactors that have a smaller feed-lance diameter, higher transport velocity for particle feeding is employed in the smaller-scale gasifier than the larger dry-feeding gasifier. Also, in a large-scale gasifier, nitrogen concentration will drop through denser phase particle feeding and also sometimes by replacing the nitrogen transport gas with the product coal-gas.

In the left-side part of the concentration profiles in Fig. 2, a difference of increasing degree in CO and H_2 concentrations is observed. While the increasing trend of H_2 remains between 14-17%, CO concentration increases from 34% to 41% and the CO_2 concentration stays at a similar level. This phenomenon has been observed through several other tests with the pilot-scale dry-feeding coal gasification system.

Temperature inside the gasifier is closely related to the methane content in the syngas. In the entrained-bed coal gasifier, measuring the temperature inside the gasifier is one of the most critical issues required for long-term operation. Typically, an R-type thermocouple is employed as the main measuring tool and the temperature inferred from syngas methane concentration is used as a backup measurement [5].

Thermodynamic analysis results reveal that methane content decreases with increasing the gasification temperature but increases by raising the gasification pressure [6]. In addition, an earlier report on the temperature sensitivity of trace gas components that had been obtained during the coal gasification test using Indonesian subbituminous coal demonstrated that the concentration changes in H_2S, COS, and NH_3 were insignificant, whereas methane concentration showed sensitive behavior upon the temperature variation [4].

Fig. 3 illustrates the relationship in that methane concentration shoots up rapidly when the temperature in the gasifier reaction zone drops below approximately 1,400 °C. When the reaction zone temperature remains above 1,500 °C, methane concentration remains at 600 ppm level. But, the methane concentration level reaches 1,000 ppm when the gasifier temperature is low as 1,350 °C. From the result in Fig. 3, it is clear that the methane concentration value can serve as an important indicator measuring the temperature of the gasifier reaction zone. As described above, a thermocouple cannot contact the hot gasifier zone directly due to melting at the thermocouple tip. Therefore, measurement of the methane concentration is an indirect tool in estimating the gasification temperature and thus can give the control data for the oxygen amount that is required to sustain the gasification reaction. If the oxygen input amount is too much at a certain time even for the limited time, a refractory or cooling screen inside the gasifier can be damaged severely in a very short time. In contrast, if the oxygen is not supplied sufficiently for the gasification reaction, ash melting into slag would not proceed well and can cause plugging of the slag tap by high viscosity during the slag flow along the inside wall of gasifier.

Particulates contained in syngas are in a half-melted state or in a char shape and contain about 35-65% unburned carbon for the Rotto Middel coal. From the mass balance point of view, the amount of unburned carbon is normally less than 0.5% of the total coal that is gasified. To employ the syngas for the electricity generation or to use as a feedstock in manufacturing chemicals, syngas should be cleaned to the level that typical clean syngas should contain particulates in concentration of less than 10 mg/Nm^3.

The experimental results show that the particulate concentration before the filtering system, which is located after the gasifier and the gas cooling pipes, was in the range of 2,500-3,000 mg/Nm^3. After the hot gas filtering system, the particulate concentration drops below 4-5 mg/Nm^3 which is difficult to identify visually in the timble surface that is used for capturing the particulates.

In the gasification test with Roto Middle coal, particulate concentration at the filter inlet was measured as 2,883 mg/Nm^3 average after 5 measurements, whereas the outlet particulate concentration was 4 mg/Nm^3. From these results, particulate removal efficiency was 99.86%. Half-cut timbles used for capturing the particulates at the inlet and outlet pipes that are connected to the filtering apparatus are shown in Fig. 4.

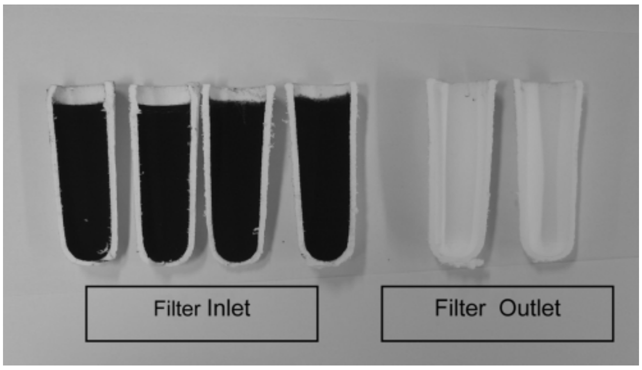

Fig. 4. Pictures of captured particulates in half-cut timble from syngas before the high temperature metal filter (left-side) and after the metal filter (right-side).

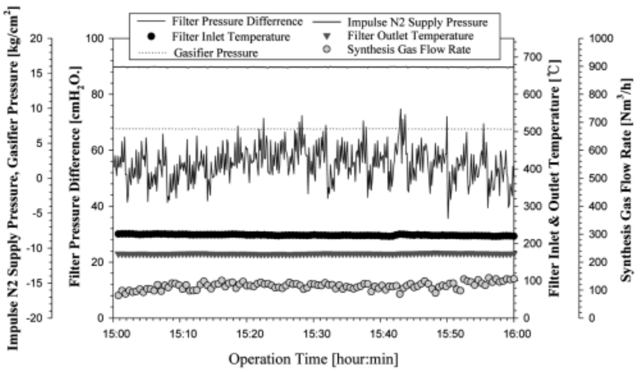

Fig. 5. Profiles of high temperature particulate removal system using metal filters.

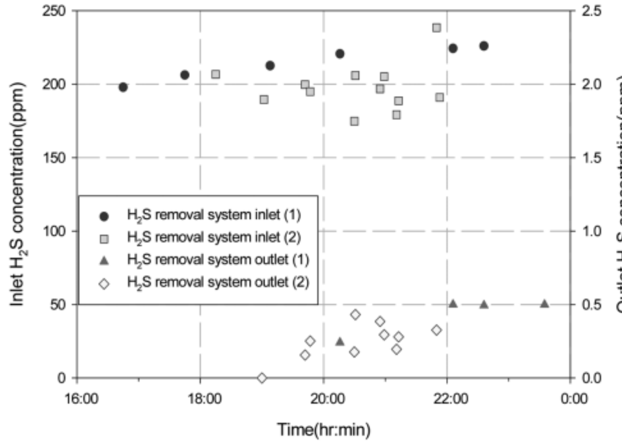

Fig. 6. Two cases of H_2S concentrations at inlet and outlet of low temperature H_2S desulfurization system for Indonesian Kideco subbituminous coal.

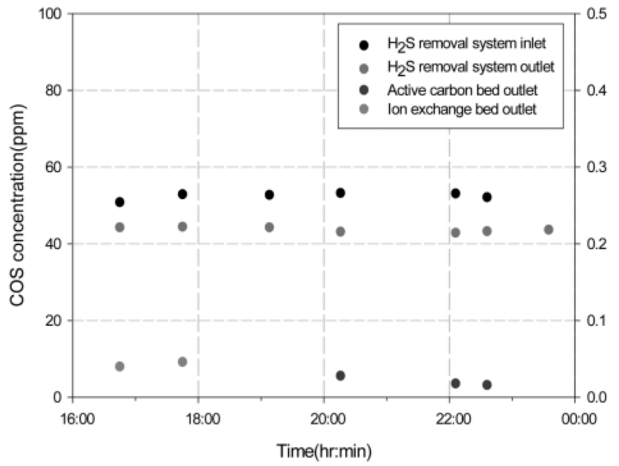

Fig. 7. COS concentrations at inlet and outlet of low temperature H_2S desulfurization system without and with COS adsorbing system.

Fig. 5 illustrates typical operating profiles during filtering. Syngas flow rate was controlled to maintain at 100 Nm^3/hr and the gasifier pressure at 7 kg/cm^2. Inlet temperature to the filtering system was 220 °C. Total 12 metal filters of 1.5 m length were employed for filtering. Pressure drop across the filters was in 40-60 cmH_2O. When the particulates accumulate in the filter surface, the pressure drop increases and the following impulse by high pressure nitrogen gas decreases the pressure drop. Impulse nitrogen pressure was 16 kg/cm^2 in this case. Typically, impulse pressure is maintained at least twice the syngas pressure. Pressure drop range could be sustained without further increasing, which means that the particulate removal system is operating steadily.

In order to use syngas for further applications in a gas turbine or DME (Di-Methyl Ether) synthesis, sulfur-containing trace compounds like H_2S and COS should be eliminated to less than 1 ppm level. For the gas turbine, these concentrations need to be removed to below a few ppm. For the chemical synthesis processes that include catalysts for the conversion step, H_2S and COS are poisons that deactivate the catalysts, and therefore should be removed as far as possible.

Figs. 6 and 7 show the low-temperature desulfurization results for two different experimental conditions with the Indonesian subbituminous coal. The H_2S has been removed to less than 0.5 ppm. The COS gas could be removed from 55 ppm to 45 ppm, even by only passing through the H_2S removal system. Further absorbing treatment with activated carbon or ion exchange resin yielded a COS level to less than 0.1 ppm. The resulting cleaned syngas of H_2S less than 0.5 ppm and COS below 0.1 ppm was satisfactory for application to chemical synthesis of DME and probably for fuel cells.

CONCLUSIONS

Indonesian coals of subbituminous rank were selected as one of the most suitable coals gasification for IGCC applications among the tested coals of bituminous and subbituminous ranks in a 3 ton/day-size pilot gasification plant. Syngas from the Roto Middle subbituminous coal was employed to test the hot gas particulate removal and the low temperature desulfurization systems. Produced syngas composition was 36-38% CO, 14-16% H_2 and around 5-8% CO_2 concentrations (dry basis). It was confirmed that the temperature inside the gasifier is closely related to the methane content in the syngas.

Particulate concentration at the filter inlet was measured as 2,883 mg/Nm^3 on average and the outlet particulate concentration was

4 mg/Nm^3. From these results, particulate removal efficiency was 99.86%. Syngas could be desulfurized to a level of H_2S less than 0.5 ppm and COS below 0.1 ppm by applying a low temperature desulfurization facility that is based upon Fe chelate and activated carbon/ion exchange resin.

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