Performance of a coal gasification pilot plant with hot fuel gas desulfurization

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Abstract−A coal gasification pilot plant operation with hot fuel gas desulfurization (HGD) was performed taking two coals (Indonesian ABK and MSJ) that differ in their carbon and sulfur contents. A dry-feeding entrained-bed type gasifier was used for gasification with oxygen and capable of operating at 30 bar pressure and 1,550 °C. The HGD unit consisted of a transport desulfurizer, a bubbling regenerator and a multi-cyclone. Attention was focused on attaining high carbon conversion and cold gas efficiency in the entrained bed reactor and the sulfur removal efficiency of the hot fuel gas desulfurization unit. The optimum conditions for achieving high performance of the operation are reported.

Key words: Entrained Bed Coal Gasification, Hot Fuel Gas Desulfurization, Bituminous Coals of Indonesia

INTRODUCTION

Gasification is the process of converting various carbon-based feedstocks such as coal, heavy refinery residues, petroleum coke, biomass and municipal wastes to clean syngas. During gasification, the feedstocks are partially combusted with oxidants such as oxygen and steam at high temperature and pressure. The syngas thus generated through an incomplete combustion is primarily a mixture of hydrogen and carbon monoxide. The syngas produced can be used as a fuel for boilers or gas turbines to generate electricity, or to make synthetic natural gas, hydrogen gas, synthetic fuels, methanol & DME (dimethyl ether) or other chemical products [1-3].

In general, the integrated coal gasification combined cycle (IGCC) operation proceeds by gasification, syngas cooling, filtration, acid gas removal (AGR) for separation of sulfur compounds and CO_2 and the production/purification of the syngas in gas (and/or steam) turbine for power generation [4,5]. The entrained bed gasifier used for coal gasification has several advantages like the ability to utilize nearly all types of coals with high throughputs per reactor volume, simpler mechanical design and the ability to reach nearly 100% carbon conversion. Koppers-Totzek, Texaco, Shell, Prenflo and GSP (GAS Schwarze Pumpe) are the well-known entrained-flow coal gasification processes in the world [6-8]. Currently, the hot fuel gas desulfurization (HGD) system, one of the elemental technologies in the IGCC [9,10], is receiving considerable attention for having both high thermal efficiency and very low emissions. The existing conventional wet-type desulfurization processes require cooling and reheating of the gas stream, resulting in significant reduction in thermal efficiency of the system as well as necessitating costly wastewater treatment. The HGD system, on the other hand, is a new method to efficiently remove H_2S and COS in fuel gas with regenerable sorbent at high temperature/pressure condition [11,12].

In the present work, we report the performance of a coal gasifi-

cation pilot plant operation which is a combination of entrained bed gasification (3 ton/day), filtration by metal filter and hot gas desulfurization (100 Nm^3/h). In gasification, we used bituminous coals of different sulfur contents and estimated both carbon conversion and cold gas efficiency (CGE). In the HGD system, sulfur concentrations in fuel gas and removal efficiency for the sulfur compounds are examined. The objective of this work is to operate both gasifier and HGD system continuously and generate data for the scale-up of IGCC processes.

EXPERIMENTS

1. Coal Samples

Coals for the experiments were chosen from the coals of sub-

Table 1. Properties of coals

Items	Coal	Indonesian ABK (dry basis)	MSJ coal (dry basis)
Proximate analysis[a] (wt%)	Moisture	11.0/-	4.9/-
	Volatile matter	38.3/43.0	40.3/42.4
	Fixed carbon	44.6/50.1	45.9/48.3
	Ash	6.1/6.9	8.9/9.3
Ultimate analysis[b] (wt%)	C	74.5	66.1
	H	4.3	5.6
	N	1.0	1.5
	S	0.34	0.84
Ash fusion temperature (°C)	IDT	1,230	1,210
	ST	1,250	1,336
	HT	1,270	1,342
	FT	1,290	1,415
Gross heating value[b] (kcal/kg)		6,682	5,963

[a]As received

[b]Moisture free basis

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Table 2. Inorganic analysis for two coals

Composition (wt%)	SiO$_2$	Al$_2$O$_3$	TiO$_2$	P$_2$O$_5$	Fe$_2$O$_3$	CaO	MgO	Na$_2$O	K$_2$O	SO$_3$
ABK	39.46	32.73	0.41	-	1.22	5.90	6.09	2.51	0.57	10.90
MSJ	47.47	24.17	0.61	0.61	8.45	4.39	2.45	1.35	1.77	8.10

Composition (wt%)	MnO	SrO	BaO	ZrO$_2$	V$_2$O$_5$	Cr$_2$O$_3$	CuO	ZnO	CoO	NiO
ABK	0.02	0.02	0.078	0.04	0.007	-	-	-	-	-
MSJ	0.04	0.07	0.11	0.01	0.03	0.01	0.01	0.02	0.01	0.01

bituminous ranks. Table 1 illustrates detailed analytical data for the tested coals. The ash contents (dry basis) of two Indonesian coals were 6.9 (ABK) and 9.3% (MSJ), respectively. Sulfur content of MSJ coal was 0.84% much more than that of ABK coal (0.34%). Basic compounds in the ash like CaO, Fe$_2$O$_3$ as given in Table 2, are found to be the ingredients in lowering ash fluid temperature. MSJ coal contains high Fe$_2$O$_3$ content that resulted in high ash fluid temperature of 1,415 °C.

2. Configuration of the Pilot Plant

A schematic diagram of dry-feeding entrained-bed type gasifier along with the filtration and the hot fuel gas desulfurization units is shown in Fig. 1. The gasifier treated 3 t/d of coal at a maximum pressure of 30 bar and 1,550 °C, and the main target feeds were sub-bituminous and bituminous coals. The slag was collected at the bottom of the gasifier. The syngas produced was cooled in indirect water-cooled heat exchanger, and then the unreacted coal and fine dust were removed in filtration section with metal filter.

Coal size was identical with that conventional power plants using pulverized coal, as 80-90% passing −200 mesh. During the pulverizing step it was ensured that the dried coal powder contained

Table 3. Operation conditions of a coal gasifier

Items	ABK	MSJ
Gasifier temperature, °C	1,250-1,300	1,200-1,450
Gasifier pressure, bar	19-20	19.8-20.2
Coal feed rate, kg/hr	60-65	75.6
Oxygen, Nm^3/hr	28-40	45-55
Nitrogen, Nm^3/hr[a]	65-80	70-75

[a]Nitrogen is used for coal transportation and are included the amount of vent (about 20 Nm^3/hr)

less than 3% moisture content. Pulverized coal was pneumatically conveyed with nitrogen gas as dense-phase into the feeding nozzle system, where 99% pure oxygen was mixed with the coal powder. Major operational variables of the gasification were the oxygen/coal weight ratio, temperature inside the gasifier and the gasification pressure. Operation conditions are in Table 3. Normal operation consisted of preheating, pressurization, transient operation, normal gasification operation and the shutdown steps. The LPG burner at
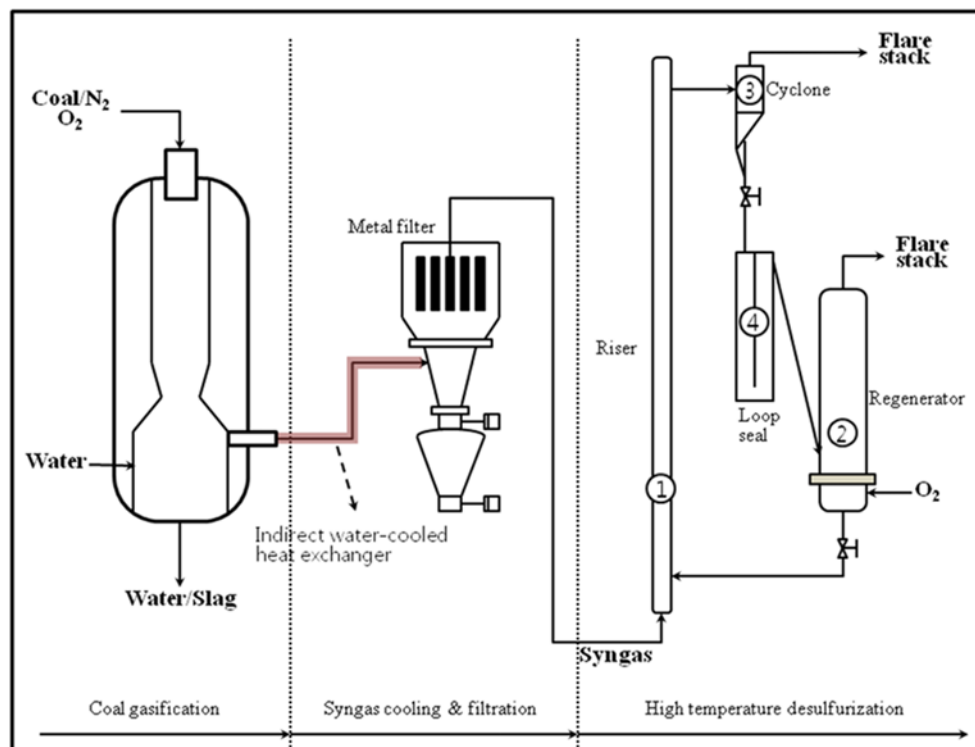

Fig. 1. Schematic diagram of coal gasification pilot plant.

Table 4. The physical properties of sorbent

Sorbent	Shape	Calcination temperature (°C)	Average particle size (μm)	Density (g/cm³)	Surface area (m²/g)	Porosity (%)	Abrasion index (%)
PS107SU	Dimple	650	118	0.82	59.5	26.9	7.6

the bottom of the gasifier did the preheating of the gasifier at least for 20 h. Then, nitrogen was introduced to pressurize the gasifier till the pre-set gasifier pressure was achieved before oxygen and coal powder were injected. The current study did not use any steam; only oxygen was employed to control the temperature and the degree of conversion [13].

Coal supply was first started at the low feeding range so that no sudden pressure buildup would take place in the gasifier, thus causing no back pressurization into the coal feeding lines. This step took normally less than 1 h. The normal hot test operation step for obtaining gasification data was maintained at the steady state for at least 4 h to provide enough gas, slag, and other process data.

On-line gas analyzers connected to the gasification system provided instantaneous gas composition of H_2, CO, CO_2, CH_4, and an additional on-line gas chromatograph (MTI Analytical Instrument, P200H) gave more precise quantitative gas composition with a 3-minute interval. The carbon conversion and the conversion to CH_4 are calculated using the following equations based on mol%:

Carbon conversion (%)

$$= \frac{\begin{array}{c}\text{Produced gas (Nm}^3\text{/h)} \times (CO+CO_2+CH_4) \\ \text{concentration in products (mol\%)}\end{array}}{\begin{array}{c}\text{Feedstock (kg/h)} \times \text{C content (wt\%)} \\ \times 0.024 \text{ (Nm}^3\text{/mol)}\end{array}} \times 100 \quad (1)$$

Cold gas efficiency (%)

$$= \frac{\text{Produced gas (mol)} \times \text{HHV (MJ/mol)}}{\text{Feedstock (kg}_{d.b.}) \times \text{HHV (MJ/kg}_{d.b.})} \times 100 \quad (2)$$

The HGD system of 100 kW scale consisted of transport desulfurizer, bubbling regenerator, multi-cyclone and a loop-seal, as shown in Fig. 1 (designed by Korea Institute of Energy Research (KIER)). The lower part of the transport desulfurizer was of 0.059 m I.D. and the upper part 0.049 m I.D. The desulfurizer was designed to operate at 2.0 m/s and the regenerator at 0.05 m/s, considering the particle size and density of the sorbents.

In the HGD system (Fig. 1), the temperature was set to the desired temperature using N_2 as the fluidization gas in the desired pressure conditions. The sorbent from the bubbling regenerator (②) at 650 °C passed through underflow standpipe and was sent to the transport desulfurizer (①) at 550 °C via a horizontal pipe. The sorbent from the desulfurizer was passed through a horizontal pipe and a riser, and subsequently to the multi-cyclone (③) [10]. The collected sorbent in the cyclone was sent to the regenerator through a loop-seal (④) at 500 °C to prevent the gas and sorbents from flowing backward. The solid circulation rate was controlled by opening the slide valve located at the bottom of the regenerator. The HGD system pressure was controlled by two control valves installed for each reactor because they should exactly control a pressure balance between two reactors. The spray-dried sorbent for desulfurization was supplied by Korea Electric Power Research Institute (KEPRI). The prepared desulfurization sorbent had physical properties suitable

for fluidized-bed applications. Most of the sorbent had dimple shape, which should be improved to spherical shape to reduce the amount of attrition loss. The dimple type of sorbent had an average particle size of 118 μm for PS107SU and was calcined at 650 °C. The properties of sorbent are shown in Table 4. The capacity of two sorbents was more than 10 wt% $g_{sulfur}/g_{sorbent}$.

The gas analysis system of HGD consisted of an online gas chromatograph (H.P. 5890 series II with a flame photometric detector) for H_2S, COS, SO_2 gases, two continuous UV H_2S analyzers (RADAS2, Hartmann & Braun Co.), a paramagnetic oxygen analyzer (775R, Rosemount Analytical Inc.), an SO_2 analyzer of IR type (URAS4 of Hartmann & Braun Co.), and an H_2 analyzer of TCD type (CALDOS5G, Hartmann & Braun Co.).
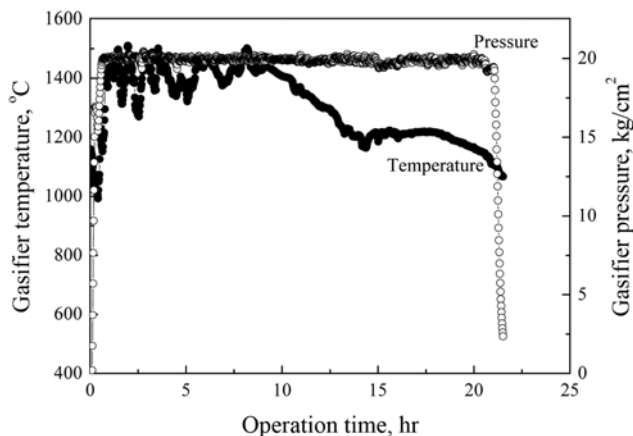
RESULTS AND DISCUSSION

1. Gasifier Operations

Gasification temperature is one of the most important operating
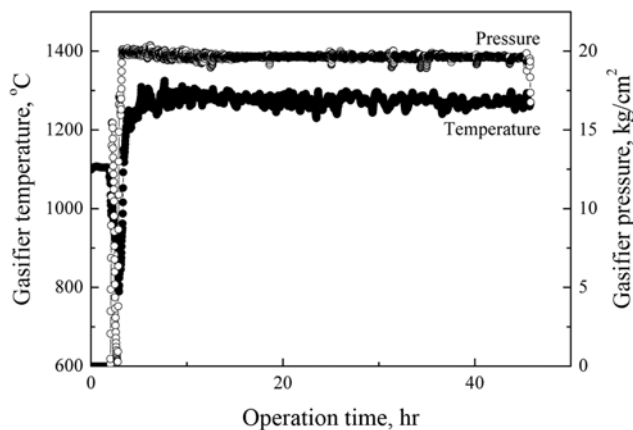
(a) Indonesian ABK

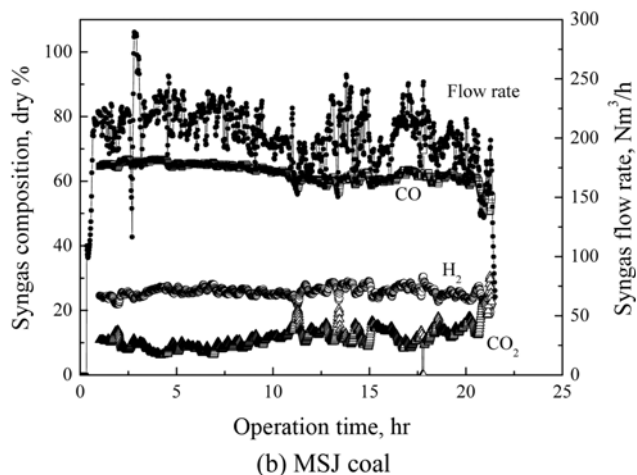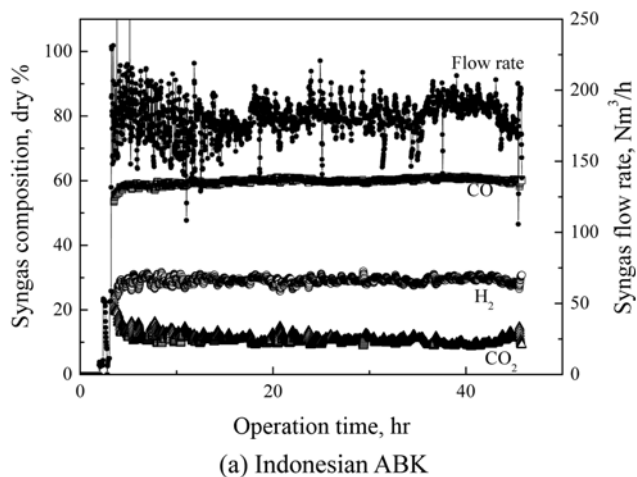(b) MSJ coal

Fig. 2. Operation temperature and pressure.

(a) Indonesian ABK

(a) Indonesian ABK

(b) MSJ coal

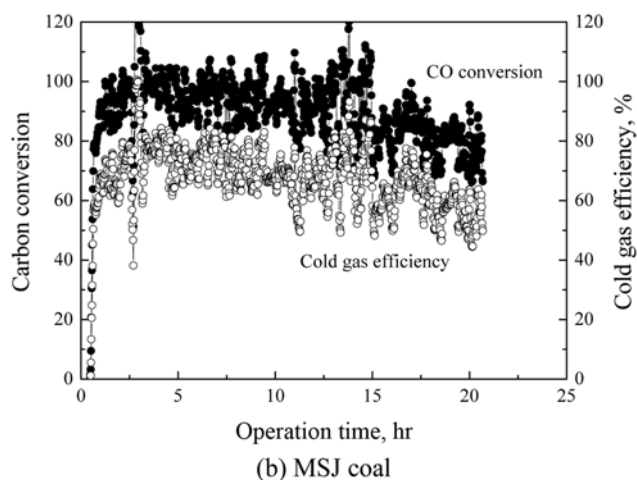**Fig. 3. Flow rate and composition in nitrogen-free of produced syngas.**

(b) MSJ coal

Fig. 4. Carbon conversion and cold gas efficiency in the gasifier.

parameters affecting the performance of gasification due to the balance between endothermic and exothermic reactions involved [14]. At a gasifier pressure of 20 bar in the 3 t/d entrained-flow gasifier, the typical temperature profile of coal gasification experiments is shown in Fig. 2. The gasifier was pre-heated to 1,000-1,100 $^{\circ}$C and pressurized to 15 bar initially, and then operating temperature and pressure were controlled by O_2/coal ratio in coal combustion step. Thus, the normal operation of the gasification step was controlled by O_2/coal ratio. This was confirmed from the syngas composition as shown in Fig. 3. As can be seen in Fig. 2, the gasifier was operated at a lower temperature than the melting point of coals to ensure non-slagging, and the temperature and pressure in gasifier were stable within 3 hours [13,15].

2. Gas Compositions and Carbon Conversion

Fig. 3 shows the composition and the flow rate of syngas produced after gasification on nitrogen-free basis. In these experiments, the feed rate of coal was 60-65 kg/h (Indonesian ABK) and 75 kg/h (MSJ coal), and the oxygen flow rate was 28-40 Nm^3/h. The average flow rate of syngas was 180.5 Nm^3/h for Indonesian ABK and 202.6 Nm^3/h for MSJ coal. Total amount of syngas and its composition varied according to the properties of two coals. Typically, the syngas composition on dry, nitrogen-free basis was 58.1-60.9% of CO, 25.8-29.4% of H_2 and 9.9-13.4% of CO_2 for Indonesian ABK,
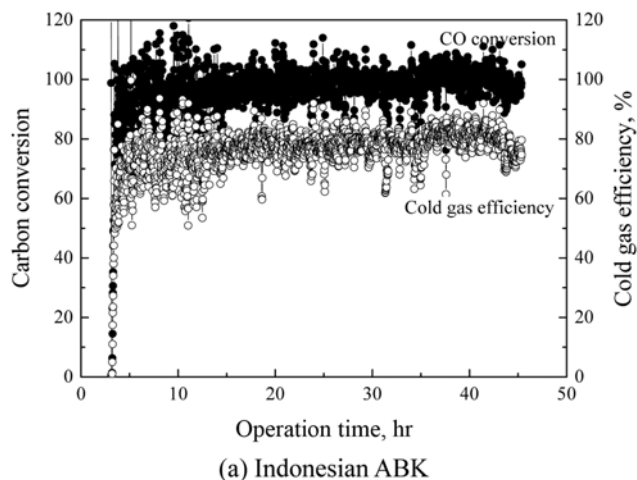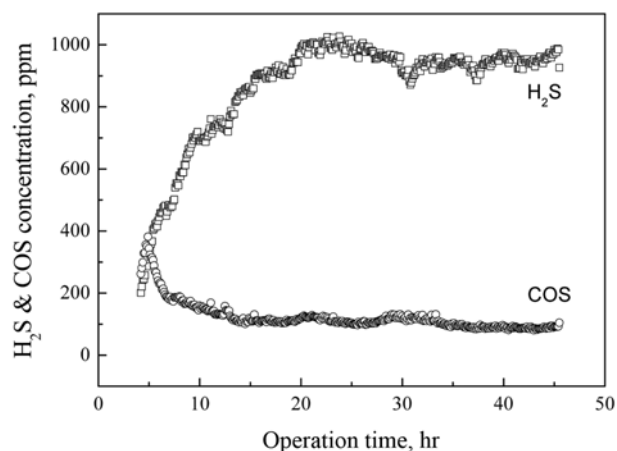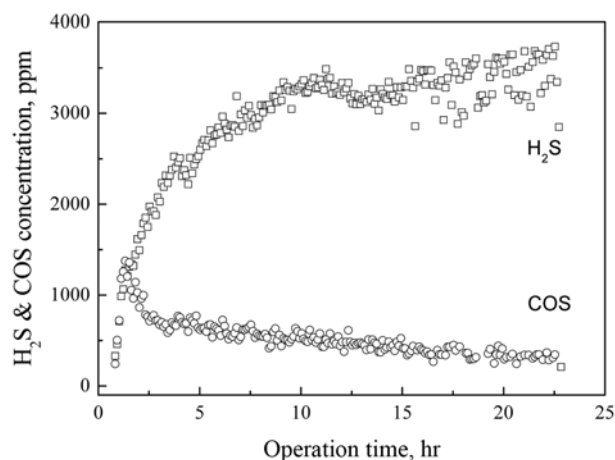
and 42.7-55.7% of CO, 5.0-15.6% of H_2 and 16.6-23.5% of CO_2 for MSJ coal. The MSJ coal required higher O_2/coal ratio for gasification operation to attain high temperatures than the Indonesian ABK. It had lower gross heating value. The O_2/coal ratio was 0.67-0.88 for Indonesian ABK and 0.75-0.94 for MSJ coal. The increase of oxygen in the feed led to higher CO_2 concentration in the products.

Fig. 4 shows the carbon conversion and cold gas efficiency. The carbon conversion (calculated by Eq. (1)) ranged between 80.4 and 118% with the average at about 100% for Indonesian ABK, and between 68.2 and 119% with the average at about 90.3% for MSJ coal. The reason for more than 100% conversion in Fig. 4 might be attributed to the accumulation of measuring uncertainties from coal feeding rate and syngas flow measurements. The coal feeding rate was calibrated with the rotating speed of coal injection screw. Syngas flow was measured at atmospheric pressure by the flow-measuring device of v-cone meter type located after the pressure control valve [13].

Cold gas efficiency is defined as the recovered energy by syngas relative to the input energy of the feed coal (calculated by Eq. (2)). For the Indonesian ABK, the cold gas efficiency ranged from 51% to 92%, which means that 51-92% of the energy input from the gasified coal was recovered as chemical energy in syngas. However, the cold gas efficiency for the MSJ coal was in the range of 47-90%. As can be seen in Figs. 3 and 4, the increase in the O_2/coal

Fig. 5. H₂S and COS concentrations in HGD system.

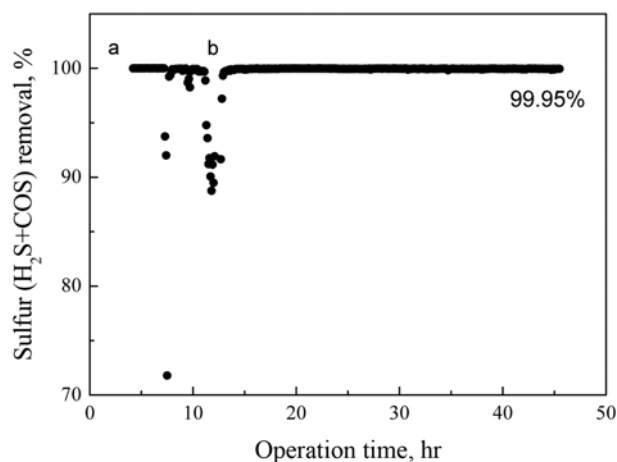Fig. 6. Removal efficiency for the sulfur compounds.

ratio caused a slight increase of CO_2 concentration in syngas, whereas H_2 concentration shows a decreasing trend with the O_2/coal ratio. In contrast, CO concentration exhibits a relatively flat response with the O_2/coal ratio variation [13].
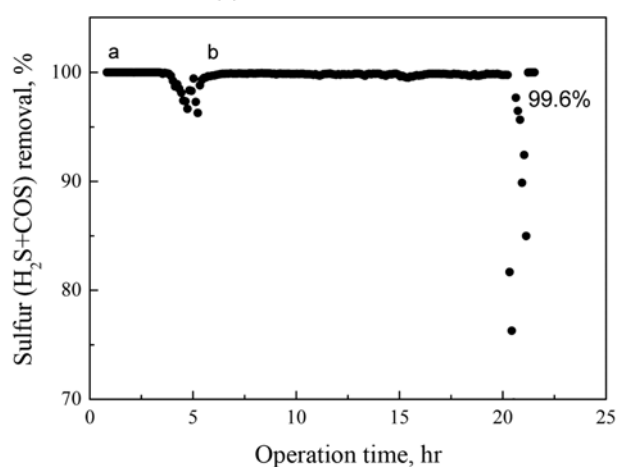
3. Desulfurization Operation

The supply of coal to the gasifier and the HGD system was operated in conjunction such that a stable composition of the syngas was attained. The syngas at 100 Nm³/h was fed to HGD system with two flow control valves. The pressure of desulfurizer and regenerator was maintained at 19 bar. For the continuous operation of HGD system, the temperature was maintained at 550 °C in desulfurizer, at 600 °C in regenerator and at 550 °C in loopseal, respectively.

Fig. 5 shows the H_2S and COS concentrations in syngas after the gasification and the filtration for the two coals. In the case of Indonesian ABK, H_2S in the syngas was in the range of 900-1,000 ppm and COS was in 100-150 ppm. On the other hand, H_2S of 2,000-3,700 ppm and COS of 300-1,000 ppm appeared in the case of MSJ coal because it contained 0.84% sulfur (Table 1).

Fig. 6 shows the removal efficiency of sulfur compounds such as H_2S and COS. The HGD system was designed to operate at a solid circulation rate of 7-8 kg/m²·s and to handle the syngas containing up to 2,000 ppm of sulfur compounds. So, MSJ coal (Fig. 6(b)) showed the sulfur removal efficiency of 99.6%, which was

lower than that in the case of Indonesian ABK. They were not completely removed as the H_2S and COS concentrations in the syngas after the desulfurization were below 17 ppm and 10 ppm, respectively. On the other hand, H_2S and COS were removed almost completely to less than 1 ppm after desulfurization in the case of ABK coal, as the removal efficiency for sulfur compounds (Fig. 5(a)) showed close to 99.95%.

CONCLUSION

The operation afforded reaching high coal conversion and cold gas efficiencies. The Indonesian ABK coal, which contains less ash content and sulfur compounds, is better in its performance in mode than the MSJ coal. The major operational variables of the gasification are the oxygen/coal weight ratio, temperature inside the gasifier, and the gasification pressure. The MSJ coal requires higher O_2/coal ratio for gasification operation to attain high temperatures than the Indonesian ABK because of its lower gross heating value. The increase of oxygen in the feed leads to higher CO_2 concentration in the products. For the Indonesian ABK the cold gas efficiency ranges from 51% to 92%, whereas that for the MSJ the range is 47-90% only. Moreover, MSJ coal in HGD system shows sulfur

removal efficiency of 99.6%, whereas ABK coal shows an efficiency close to 99.95%.

ACKNOWLEDGEMENT

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