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Operational characteristics of the pilot-scale coal gasification with filtration and hot fuel gas desulfurization

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Abstract−Experimental research on coal gasification with a filtration and desulfurization system for the development of an integrated gasification combined cycle (IGCC) was performed with Indonesian LG, KPU and Canadian Arch coals. A dry-feeding entrained-bed type gasifier was operated below the fusion temperature of the coal and at 20 bar of pressure. The filtration system was designed for continuous capture and subsequent removal of the fly ash and the unreacted coal residue via a specialty metal filter. The hot fuel gas desulfurization unit (HGD) consisted of a transport desulfurizer, a bubbling regenerator and a multi-cyclone. The research objective was to investigate the feasibility of applying a partial slagging coal gasifier to attain high carbon conversion and cold gas efficiencies as well as to attain an operational capability for combining with filtration and HGD. A Pilot-scale test demonstrated that the coal fines were effectively removed and the overall sulfur removal efficiency of the hot fuel gas desulfurization unit was higher than 95.3%.

Keywords: Entrained-bed, Coal Gasification, Filtration, Hot Fuel Gas Desulfurization, Metal Filter

INTRODUCTION

The gasification process converts various carbon-based feedstocks such as coal, heavy refinery residues, petroleum coke, biomass and municipal wastes to syngas. During gasification, the feedstocks react with oxygen and steam at high temperature and pressure. The syngas thus produced via incomplete combustion is primarily a mixture of hydrogen and carbon monoxide and can be used as fuel for boilers or gas turbines to generate electricity or make synthetic natural gas, hydrogen gas, synthetic fuels, methanol and dimethylether (DME) or other chemical products [1,2]. In general, the integrated coal gasification combined cycle (IGCC) process for power generation proceeds by successive unit processes of gasification, syngas cooling, filtration, acid gas removal (for separation of sulfur compounds and CO_2) and gas (and/or steam) turbine operation [3].

The entrained-bed coal gasifier has several advantages, including the versatile use of coals with high throughputs per reactor volume, a simpler mechanical design, near-100% carbon conversion rate and higher cold gas efficiency. Shell, GE Energy, Uhde and CBI are well known worldwide for their commercially available technology in the field of entrained-flow coal gasification processes [4,5]. Post-gasification fly ash and unreacted coal residue were captured by a metal or ceramic filter system [6,7]. The thermal stability of ceramic filter media depends on the materials used, so temperatures up to 1,000 °C and higher are possible [8,9]. The U.S. Department of Energy's Morgantown Energy Technology Center (DOE METC) has a hot-gas cleanup (HGC) program intended to develop and demonstrate gas stream cleanup options for use in combustion- or gasification-based advanced power systems. A hot-gas filter vessel (HGFV) developed under the HGC program is based on ceramic candle filters. The average incoming particulate loading capacity of the HGFV was 6,700 ppm, whereas the outgoing loading capacity was 300 ppm [10]. However, the main disadvantage of this filter element design is the risk of the upstream cells becoming plugged, which cannot be removed by back pulsing [11]. Therefore, a metal filter system was adopted in the present filtration system. In the IGCC system, the removal of H_2S is needed mainly to protect the gas turbine from high-temperature corrosion, which induces the formation of alkali sulfates, and the H_2S concentration must be limited to <20 ppm [12]. For the removal of sulfur compounds of H_2S and COS, a hot fuel gas desulfurization (HGD) system has been intensively investigated for its high thermal efficiency and very low emissions [13,14]. Conventional wet-type desulfurization processes utilize cooling and reheating of the gas stream, resulting in significant reduction in thermal efficiency of the system and costly wastewater treatment. However, an IGCC process with an HGD system is a new and untested method that has been claimed to remove sulfur compounds efficiently from syngas via regenerable sorbent at high temperature/pressure [15]. RTI's warm syngas desulfurization process demonstration unit has accumulated more than 1,000 h of operation using commercially available coal- and petcoke-based syngas feed, achieving greater than 99.7-99.9% total sulfur removal (for both H_2S and COS) at tempera-

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tures up to 600 °C [16].

In the present study, the operational performance of a coal gasification pilot plant with entrained-bed gasification and subsequent filtration by a metal filter and HGD system is comprehensibly reported. Three subbituminous coals of varying sulfur contents were used to obtain the data on the carbon conversion and cold gas efficiency (CGE) of gasification in addition to the particulate removal and sulfur removal efficiencies. The post-gasification fly ash and unreacted coal were mostly removed by a dust collector. The removal efficiency of the sulfur compounds in the HGD system was quantitatively examined via measurement of residual sulfur concentrations in the syngas. The present study is aimed to generate comprehensive data for continuous operation of a gasifier attached with filtration and HGD system for future application in the scaled-up gasification process.

EXPERIMENT

1. Coal Preparation

Table 1 lists detailed analytical data for the sub-bituminous coals tested. The ash content of coals was 8.62% (Indonesian LG), 11.79% (Indonesian Komisi Pemilihan Umum (KPU)) and 5.47% (Canadian Arch). The sulfur content of Arch coal was 0.64%, greater than that of LG (0.3%) and KPU (0.47%) coals. The elementary oxides of CaO and Fe_2O_3 in the coal ash tend to lower the ash fluid temperature. Arch coal contained a higher CaO content, resulting in a reduced ash fluid temperature of 1,251 °C. The coal was pulverized to -200 mesh size; 80-90% of the coal was able to pass through the mesh, which was quite similar to the size of the pulverized coal powders utilized at conventional pulverized coal power plants. During the pulverization step, the coals were dried to the desired moisture content. The final moisture content of the pulverized and subsequently dried LG, KPU and Arch coals was measured to be 6.93, 4.29, and 19.15 wt%, respectively. The typical dryness of coal powder in commercial dry-feeding coal gasifiers is controlled to be less than 3 wt%, which demands a significant amount of energy during drying. Considering the inherent moisture content of 15-30% in subbituminous coals, drying to the level of 3%, which is necessary for smooth feeding through narrow nozzle tips to the gasifier, remains a problem to be resolved. In this study, we were also interested in the gasification characteristics when applying coals of higher moisture content. In principle, coal feeding through the narrow nozzle with high speed would be influenced only by the surface moisture in dried coal powder, not the inherent moisture that resides inside the coal structure. For Arch coal, we attempted containment of the coal even at the 19% level. The pulverized coal was pneumatically conveyed by compressed nitrogen gas into the gasifier via a feeding nozzle system.

2. Configuration of the Pilot Plant

2-1. Coal Gasification System

A schematic diagram and photos of the dry-feeding entrained-bed type gasifier with the filtration and hot fuel gas desulfurization units are shown in Fig. 1. The width and height of the applied pilot-scale coal gasifier were 1,158 and 6,320 mm, respectively. The gasifier capacity was 3 t/day of coal at a maximum pressure of 30 bar and 1,550 °C. The main object feeds were of subbituminous and bituminous coal-types. A partial slagging entrained-bed type gasifier was employed in the present study. When a small amount of slag was generated when the gasifier temperature rose above the ash fusion temperature of coal in a partial gasification mode, the produced slag would be removed through the lower quencher of the gasifier. Considering the small quantity of the slag that was generated, the gasifier operation was not interrupted for any maintenance-related activity by slags. The syngas produced was cooled in an indirect water-cooled heat exchanger, and the unreacted coal residue and fine dust were captured by the metal filter in the filtration section [17].

The major operational variables of the gasification were the oxy-

Table 1. Properties of the coal types

Items	Coal types	Indonesian LG coal	Indonesian KPU	Canadian Arch
Proximate analysis[a] (wt%)	Moisture	6.93	4.29	19.15
	Volatile matter	36.01	36.39	34.02
	Fixed carbon	48.44	47.53	41.37
	Ash	8.62	11.79	5.47
Ultimate analysis[b] (wt%)	C	65.25	70.5	72.47
	H	5.2	4.5	5.21
	N	1.26	1.36	1
	S	0.3	0.47	0.64
Ash fusion temperature (°C)	IDT	1,114	1,193	1,173
	ST	1,378	1,305	1,222
	HT	1,390	1,321	1,225
	FT	1,416	1,365	1,251
Gross heating value[b] (kcal/kg)		6,221.1	6,195.8	6,314.0

[a] Analyzed for the feed coal after drying

[b] Moisture-free basis

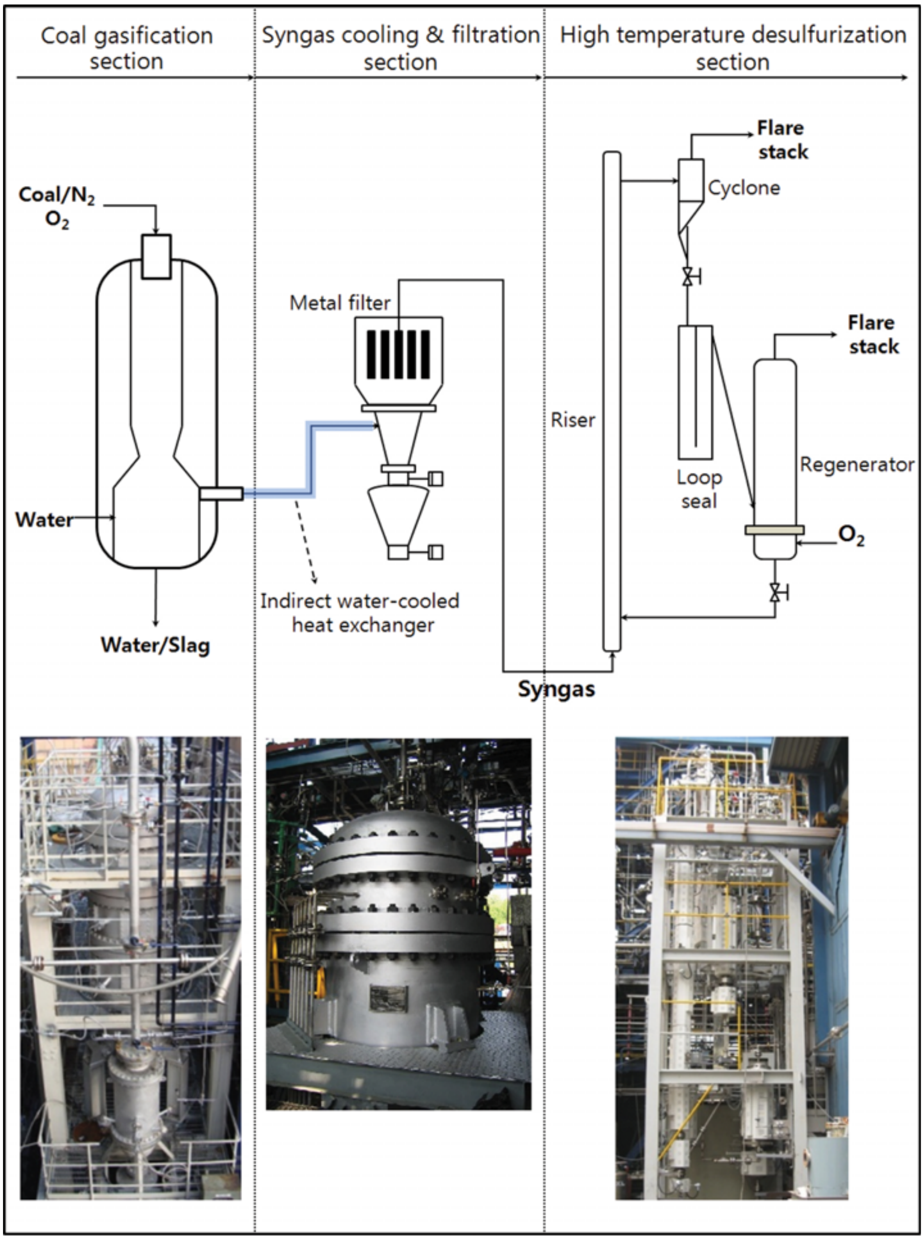

Fig. 1. Configuration of the coal gasification, filtration and HGD system.

gen/coal weight ratio, reaction temperature of the gasifier, and gasification pressure. The overall unit operations consisted of pre-heating, pressurization, transient and normal gasification and the final shutdown steps. The liquefied petroleum gas (LPG) burner at the top of the gasifier preheated it for a minimum of 20 h until the gasifier temperature read above 900 °C. The LPG burners were then turned off, and two oppositely-positioned compressed natural gas (CNG) burners located just below the top coal burner replaced the gasifier heating until the gasifier pressure reached 16 bar. After reaching 16 bar, coal feeding started with pure oxygen flow. Within 1 h, gasifier pressure would reach 20 bar for continuous coal feeding. Oxygen was the sole oxidizer in the present study for controlling the temperature and conversion rate. Coal was initially supplied at a low feed rate so that no sudden pressure buildup would take place in the gasifier, thus ensuring no back pressurization into the coal feeding lines. This step normally took less than 1 h. The usual hot test operation step for gasification data acquisition was maintained at the steady state for a minimum of 4 h to ensure the supply of sufficient gas to obtain various process data. In this study, the operation time was 50 to 90 h.

The syngas amount was measured after the HGD section by the gas flow-meter (ABB Swirl Flowmeter, FS4000-ST4), which is based on the principle of measuring the swirl difference by the gas flow. On-line gas analyzers connected to the gasification system monitored the gas composition of H_2, CO, CO_2, CH_4 in real time. More precise gas composition was also quantitatively monitored at 3-min intervals by an additional on-line gas chromatograph (MTI Analytical Instrument, P200H). The carbon conversion and cold gas efficiency were calculated using the following equations:

Carbon conversion (%)

$$= \frac{\begin{array}{l}\text{Produced gas }(\text{Nm}^3/\text{h})\\ \quad \times(\text{CO}+\text{CO}_2+\text{CH}_4)\text{ in produced gas (mol\%)}\\ \quad \times 12.011(\text{kg/kmol})\end{array}}{\text{Feedstock (kg/h)}\times\text{C content (wt\%)}\times 22.1(\text{Nm}^3/\text{kmol})}\times 100 \qquad (1)$$

Cold Gas Efficiency (%)

$$= \frac{\text{Produced gas (mol/hr)}\times\text{HHV (MJ/mol)}}{\text{Feedstock (kg/hr)}\times\text{HHV (MJ/kg)}}\times 100 \qquad (2)$$

In the present study, the cold gas efficiency has been regarded as a more important index of gasification performance from the context of the overall gasification condition.

2-2. Filtration System

The filtration system captured the fly ash and unreacted coal residue generated from the gasification system and also continuously discharged them to the outside. It is composed of a main body with numerous metal filters, a lower hopper to capture and discharge the dust, and a receiver tank for transferring and storing the dust accumulated on the lower hopper. The collected dust accumulated on the outer surface of the filter was regularly removed by a pressurized pulse of nitrogen per the differential pressure at the fore and aft sections of the dust collector. The filtration system contained 12 metal filters (of 1,500 mm length and 60 mm diameter).

2-3. Hot Fuel Gas Desulfurization (HGD) System

The HGD system of a 0.1 MW scale consisted of a transport desulfurizer, bubbling regenerator, multi-cyclone and loop-seal, as shown in Fig. 1 (designed per contemporary Korean technology). The lower and upper sections of the transport desulfurizer were of 0.059 and 0.049 m I.D., respectively. The desulfurizer and regenerator were designed to operate at 2.0 m/s and 0.05 m/s, respectively, considering the particle size and density of the sorbents. In the HGD system, the sorbent from the bubbling regenerator passed through the underflow stand-pipe and to the transport desulfurizer via a horizontal pipe. The sorbent from the desulfurizer was then passed through a horizontal pipe and a riser to the multicyclone [18]. The collected sorbent in the cyclone was again transported to the regenerator through a loop-seal to prevent the gas and sorbents from flowing backward. The circulation rate of the sorbent was controlled by opening the slide valve located at the bottom of the regenerator. The HGD system pressure was controlled by two valves installed at each reactor to render a pressure balance between two reactors. The spray-dried sorbents for desulfurization were also developed per contemporary Korean technology at KEPRI (Korea Electric Power Research Institute).

The spherical-type sorbents were calcined at 650-750 °C with the capacity of sorbents specified to be more than 10 wt% $g_{sulfur}/g_{sorbent}$. The sorbents used in HGD basically consisted of 50 wt% zinc oxide (ZnO) as an active material, 50 wt% matrices of multibinders, and a Ni-based promoter added at 7.5 wt% [13]. A detailed description of the sorbent preparation process is available elsewhere [13]. Table 4 lists the physical properties of the sorbents used for desulfurization of syngas produced by different coal samples. PS107 was used for Indonesian LG coal, and PS-129TP and SC-229 were

Table 2. Inorganic analysis result for the three coal types

Composition (wt%)	SiO_2	Al_2O_3	TiO_2	P_2O_5	Fe_2O_3	Cr_2O_3
LG	49.53	25.19	0.84	0.57	7.08	0.01
KPU	45.08	22.73	0.95	0.4	5.54	-
Arch	21.98	16.16	1.24	1.22	5.96	-
Composition (wt%)	CaO	MgO	Na_2O	K_2O	SO_3	ZnO
LG	4.22	2.33	1.02	1.87	6.94	0.02
KPU	6.47	2.34	2.27	1.67	12.14	0.02
Arch	22.82	4.94	1.29	0.39	23.36	-
Composition (wt%)	MnO	SrO	BaO	Z_rO_2	V_2O_5	-
LG	0.004	0.007	0.11	0.02	0.06	-
KPU	0.03	0.04	0.16	0.01	0.03	-
Arch	0.04	-	-	-	-	-

Table 3. Operating conditions of the coal gasifier

Items	LG	KPU	Arch
Gasifier temperature (°C)	1,200-1,300	1,150-1,250	1,050-1,250
Gasifier pressure (bar)	18.8-19.2	19.8-20.2	19.8-20.3
Coal feed rate (kg/h)	74.39	76.8	72.7
Oxygen (Nm^3/h)	40-45	39-43	37-38.5
Nitrogen (Nm^3/h)	3-40	39-43	41-43
Oxygen/Coal	0.77-0.86	0.73-0.80	0.73-0.76
Oxygen/Carbon	0.50-0.56	0.51-0.56	0.53-0.55

Table 4. The physical properties of the sorbents

Sorbent	Shape	Calcination temp. (°C)	Average particle size (μm)	Density (g/cm^3)	Surface area (m^2/g)	Porosity (%)	Attrition index (%)
PS107	Spherical	650	118	0.82	61.1	26.9	7.6
PS-129TD	Spherical	700	98	0.89	56.6	33.6	10.4
SC-229	Spherical	750	89	1.03	45.7	32.2	12.1

used for Indonesian KPU and Canadian Arch coals, respectively. The particulate density was based on tap density, and the abrasion loss of the individual particles was measured per the ASTM D-5757-95 standard. The gas analysis system of HGD consisted of an online gas chromatograph (HP 5890 series II with a flame photometric detector) for H_2S, COS, and SO_2 gases; two continuous ultraviolet (UV) H_2S analyzers (RADAS2, Hartmann & Braun Co.); a paramagnetic oxygen analyzer (775R, Rosemount Analytical Inc.); a 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. Gasification Operations

Operational parameters, such as operating temperature and pressure, coal feed rate and oxygen flowrate, are listed in Table 3. More specifically, the temperature of the gasifier was maintained below the ash fluid temperature (FT) of the tested coal samples to operate in partial slagging mode. Even in partial slagging mode, earlier test results obtained from the same gasifier show carbon conversion rates greater than 90% with a single pass of coal feed through the gasifier [19]. The recycling of the un-reacted carbon fraction resulted in 98-99% conversion. In comparison, most commercial entrained-bed coal gasifiers are operated under full slagging mode, which necessitates operating temperatures higher than the ash fluid temperature to accommodate several thousand tons of coal in a single gasifier, and the unreacted coal fines are recycled into the gasifier to obtain a 98-99% carbon conversion rate. The partial slagging mode gasification yielded lower carbon conversion in the first pass through the gasifier, but it guaranteed lower construction cost and much more efficient gasification by fluid slags at the slag-tap and other parts of the system. Operational problems such as the slag-tap blocking caused by viscous slags were frequently encountered in a small pilot-scale coal gasifier with an inside reactor diameter of 20-30 cm.

Typical gasification pressure and temperature profiles for the three coal types are shown in Fig. 2 and Fig. 3, respectively. The gasifier pressure was maintained at 20 bar by manipulating the flow control valve located at the end of the desulfurization section, located just before the flare stack. When the gasification reached a steady state, the gasifier pressure did not change appreciably. Commercial coal gasifiers typically operate at pressure levels that accommodate the operational pressure drops expected in the subsequent syngas utilization facilities. For electricity generation in the IGCC (integrated gasification combined cycle), the gasifier should generate sufficiently high syngas pressure for the subsequent gas turbine operation. Commercial IGCC gasifiers before 2000 were operated at the 22-28 bar range depending on the gas turbine type. Recent advancements in gas turbine technology require a higher coal gasifier pressure, 42 bar, such as in the Taean IGCC power plant in Korea and Edwardsport IGCC plant in the USA. Regarding chemi-

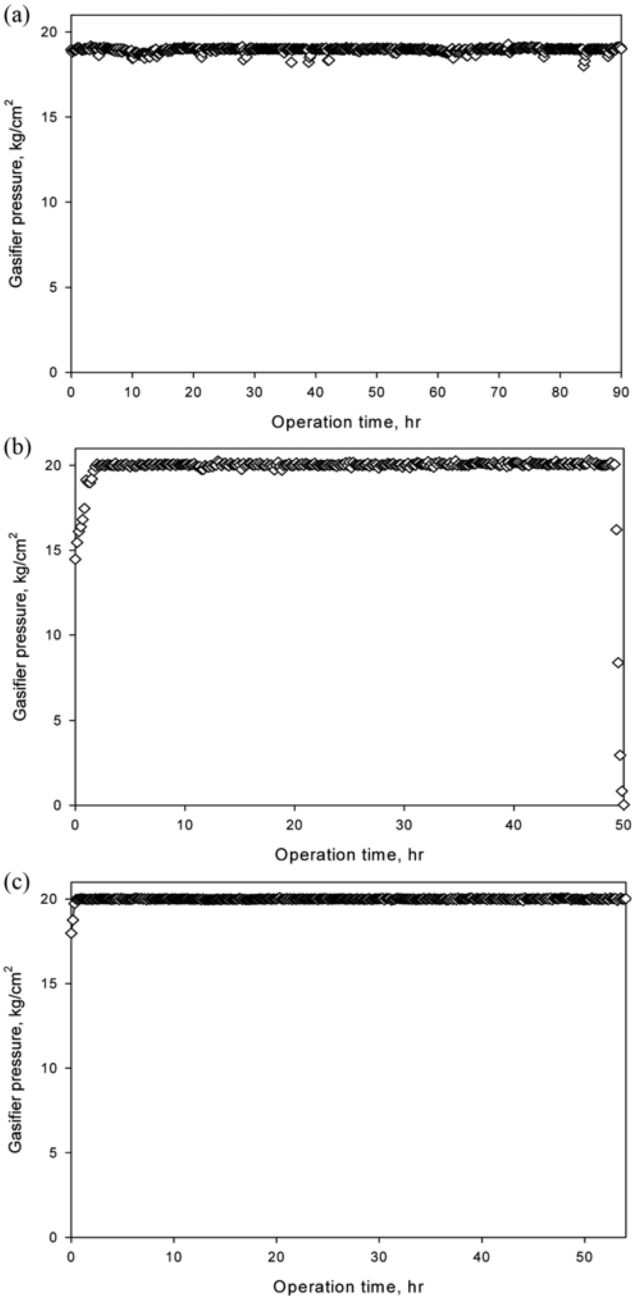

Fig. 2. Operation pressure of the gasifier for three different coal types.

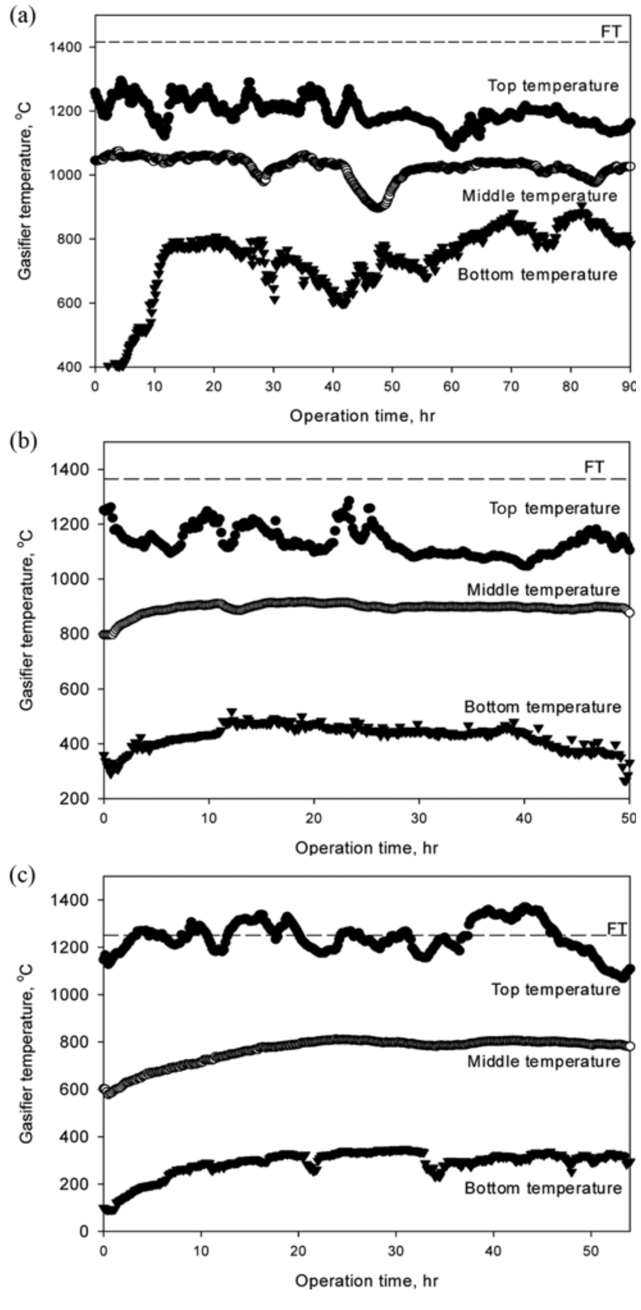

Fig. 3. Operation temperatures of three different gasifier sections.

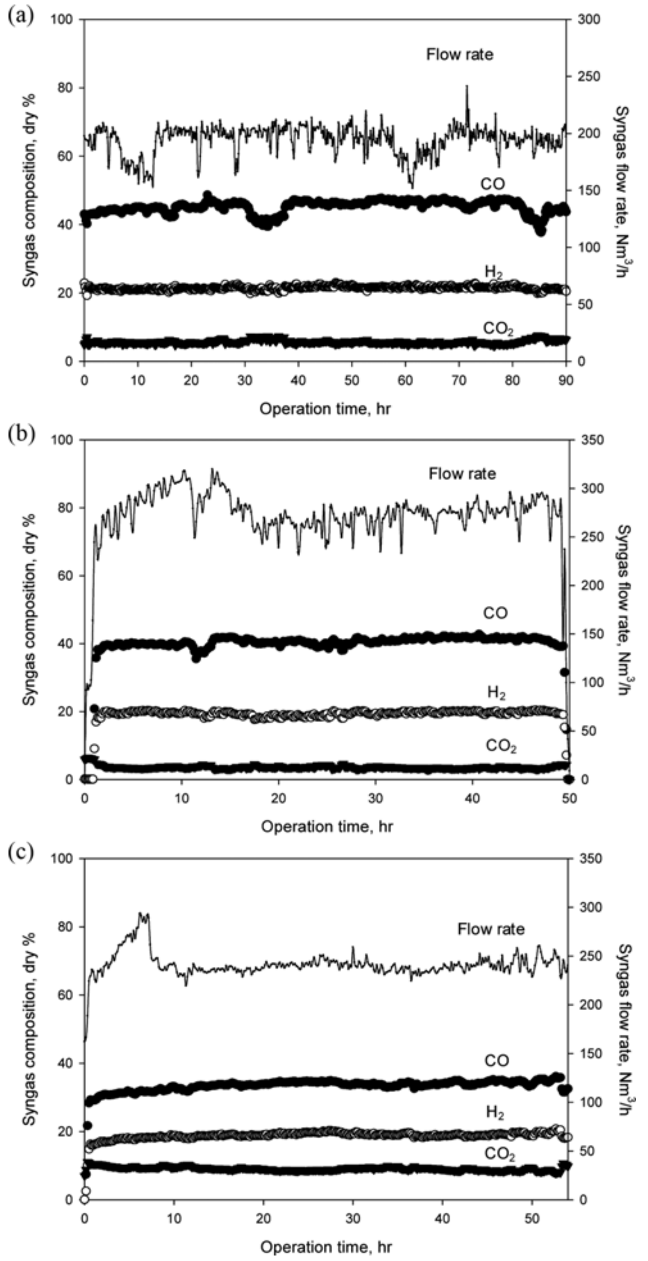

Fig. 4. Flowrate and syngas composition profiles for the three tested coal types.

cal plants using syngas as a raw material, the coal gasifier pressure should be higher than that of the chemical conversion reactor. This route is the most economical compared with lower-pressure coal gasification with post-gasification syngas pressurization, considering the additional cost incurred by the additional mechanical compressor and related maintenance.

In view of this result, the pilot-scale gasifier pressure was set at 20 bar for the partial slagging gasification mode with the filtration/HGD units. The coal gasifier performance was not significantly affected by the gasification pressure, except for some morphological variation of the fly fines. Therefore, the gasifier pressure was set for the final syngas utilization, without considering any operational improvement or optimization. Higher pressures increase construction costs and safety-related issues, although higher coal gasification pressure ensures a more compact reactor suitable for commercial gasifiers with a 1,000-5,000 t/day capacity.

Gasification temperature is one of the more important operating parameters affecting the performance of gasification and should be well optimized for the various endothermic and exothermic reactions in the gasifier [20]. The gasifier was pre-heated to 1,000-1,100 °C by LPG combustion gas, and then pressurized to 16 bar via compressed natural gas (CNG) combustion. Pulverized coal and oxygen were then supplied at slightly higher O_2/coal ratio until the desired gasifier temperature was reached. The O_2/coal ratio was then subsequently controlled to maintain the final temperature and ensure stable syngas composition, as shown in Fig. 4.

A partial slagging entrained-bed type gasifier was used in this gasification tests. Although a partial slagging gasifier transforms the inorganics mostly into ash form, thus rarely causing a heavy metal leaching problem [20], partial slagging operation could guarantee long-term continuous operation without damaging the refractory. Therefore, it was necessary to maintain the gasifier temperature below the melting point of the coal types used.

An earlier result [18] mentioned only the near flame temperature inside the gasifier, which is the top temperature in Fig. 3. In the present study, the inside gasifier temperatures at three different locations were averaged. The top temperature of the coal gasifier burner region was caused by the autogenous reactions involving active carbon, oxygen, and hydrogen reactants. The measured temperature at the middle section of the gasifier reflected any probable fouling caused by ash components along the inside wall of the gasifier. This middle section temperature gave some hint of proper gasifier operation without causing any wall fouling by molten ash, which could be used as an empirical guide or index to compare gasifier performance results obtained from different coal types. The bottom temperature measuring point was located near the syngas entering port of the heat recovery system above the slag tap. The bottom temperature also gave an important guideline for gasification with different coal types without significant ash melting/fouling at the lower temperature section of the gasifier. Arch coal contained 19.15 wt% moisture and resulted in a lower bottom temperature than that of the other two coal types. Arch coal could have been dried to the desired level, but the operational performance as such was investigated in the present study. Surface drying was sufficient for feeding and the subsequent gasifier operation. However, the lower bottom temperature was observed, which might cause ash deposition during long-term operation. There should be an optimal point between the allowable moisture level for coal feeding and the temperature drop at the lower part of the gasifier. Because coal drying is costly and the gasification reaction involves a water gas shift reaction, a certain level of moisture in the feeding coal is desirable to attain an H_2O/coal ratio of 0.1. Although the 19 wt% moisture level of the Arch coal is deemed unsuitable for gasification, coal feeding was not hindered.

In summary, the top gasifier temperature above 1,100 °C should ensure fast chemical reaction within the fixed gasifier volume with partial slagging. Gasifiers are operated to attain this minimum top gasifier temperature by optimizing the O_2/coal ratio. However, the precise measurement of the coal feeding amount in a small-scale pilot plant is technically challenging in view of the small size of the feeding pipe and the consequent difficulty of using a commercial measuring device. Therefore, the coal feeding amount was measured by the on-line weighing scale in this study.

2. Syngas Compositions & Carbon Conversion

At gasification temperatures above 1,200 °C, chemical reaction rates are sufficiently high so that chemical equilibrium calculation can almost precisely estimate the syngas composition. When a sufficient gasifier volume to satisfy the residence time for the coal powder of 4-8 s (typically in the full slagging coal gasifiers) remains inside the gasifier, along with a sufficiently gasifying temperature (typically in the full slagging coal gasifiers, above 1,400 °C), syngas composition by calculation of thermodynamic equilibrium is sufficient to provide an accurate value.

Fig. 4 shows the post-gasification syngas composition and flowrates of the syngas measured by the gas flow meter. With the coal feed rates into the gasifier range of 72.7-76.8 kg/h and the corresponding oxygen flowrates of 37-45 Nm^3/h, the average syngas flowrates were 203, 276 and 241 Nm^3/h for LG, KPU and Arch coals, respectively. The syngas results show that the syngas generation amounts per ton/per day (TPD) were 113, 149, and 139 Nm^3/hr/TPD, respectively. Typical commercial coal gasifiers produce syngas in the range of 80-100 Nm^3/hr/TPD. The pilot gasification test in this study had to employ less than 0.5 in of tube for coal powder feeding and thus required more transport nitrogen in the feeding process to compensate for the wall friction. In Fig. 4, the remaining gas above 100%, after subtracting the added CO, H_2, and CO_2 compositions, is primarily the nitrogen that was used for coal feeding, so the N_2 composition in syngas was 28%, 38%, and 39% in the three coal tests. When the coal feeding capacity increases, the N_2 fraction in syngas drops dramatically. A 20 TPD scale coal gasifier system with identical partial slagging mode demonstrated an N_2 content less than 5% in syngas composition (results will be reported elsewhere). In a commercial 3,000 TPD coal gasifier, dry-feeding full-slagging operation produces a syngas amount of less than 6% N_2 [22]. Therefore, N_2 transport gas amount can be reduced significantly even after the scale of 20 TPD.

Whereas, the H_2 compositions of the three tested coals exhibit similar values of 20.5%, 19%, and 19%, the CO composition varies from 46% in LG coal to 41% in KPU coal and 35% in Arch coal. The CO_2 concentration was 4.5%, 3.5%, and 9% in the three coal types. The H_2 in syngas basically comes from three sources: devolatilization of H components in volatile matter, gasification of the C component in coal with an H_2O component such as steam ($C+H_2O\rightarrow CO+H_2$), and the water-gas shift reaction ($CO+H_2O\rightarrow CO_2+H_2$). The CO in syngas is generated through the devolatilization of volatile matter (ether bonds, etc.) and the reaction of carbon in coal with O_2 that was introduced as an oxidant ($C+1/2O_2\rightarrow CO$) and with CO_2 ($C+CO_2\rightarrow 2CO$), as well as with H_2O ($C+H_2O\rightarrow CO+H_2$). Detailed reaction formulas of these chemical reactions for CO and H_2 production from char and water gas shift reaction are as follows.

$$C(s)+1/2O_2\rightarrow CO;\ \Delta H_R^0=-110.5\ \text{MJ/kmol (exothermic)} \tag{3}$$

$$C(s)+H_2O(g)\rightarrow CO+H_2;\ \Delta H_R^0=+131.4\ \text{MJ/kmol (endothermic)} \tag{4}$$

$$C(s)+CO_2\rightarrow 2CO;\ \Delta H_R^0=+172.0\ \text{MJ/kmol (endothermic)} \tag{5}$$

$$CO+H_2O(g)\rightarrow CO_2+H_2;\ \Delta H_R^0=-41.0\ \text{MJ/kmol (exothermic)} \tag{6}$$

where all ΔH_R^0 are at 298.15 K and atmospheric pressure. Because oxygen is the most reactive gas component inside the gasifier environment, among the above reaction routes, the $C+1/2O_2$ reaction is the first and fastest reaction whenever oxygen exists, whereas the reaction involving H_2O is slower than oxygen-related reactions. From the syngas composition results, it is clear that the CO concentration is more sensitive to the variation of coal properties and the operating conditions in gasification.

The CO_2 concentration in Arch coal, which contains more moisture inside the coal structure–as high as 19.15 wt% through insufficient drying on purpose–is approximately 9%, whereas the other two coals exhibit 3.5-4.5%. More moisture requires a higher energy input to the gasifier to maintain the gasifier temperature required for the carbon in coal to be consumed through particle combustion, resulting in a higher CO_2 concentration.

The total amount of produced syngas and its composition varied for the three coal types as shown in Fig. 4. The H_2/CO ratio of the individual syngas was 0.48 for LG and KPU coal and 0.57 for Arch coal. Minor components included methane- and sulfur-containing gases of less than 1%, and the balance was nitrogen. Methane formation is strongly affected by the gasification temperature.

The transient carbon conversion and the cold gas efficiency are shown in Fig. 5. The carbon conversion, calculated by Eq. (1), ranged between 80.0 and 122%, with average values of 97% for LG and KPU and 96% for Arch coal. The carbon conversion exceeding 100% in Fig. 5 could be attributed to the fluctuation of syngas flowrates, uncertainties in the coal feed rate and accumulated measurement errors. In the study, the coal feed rate was calibrated with the rotating speed of a coal injection screw, and the syngas flow was measured at ambient pressure by the flow-measuring device of a v-cone type meter located after the pressure control valve [18].

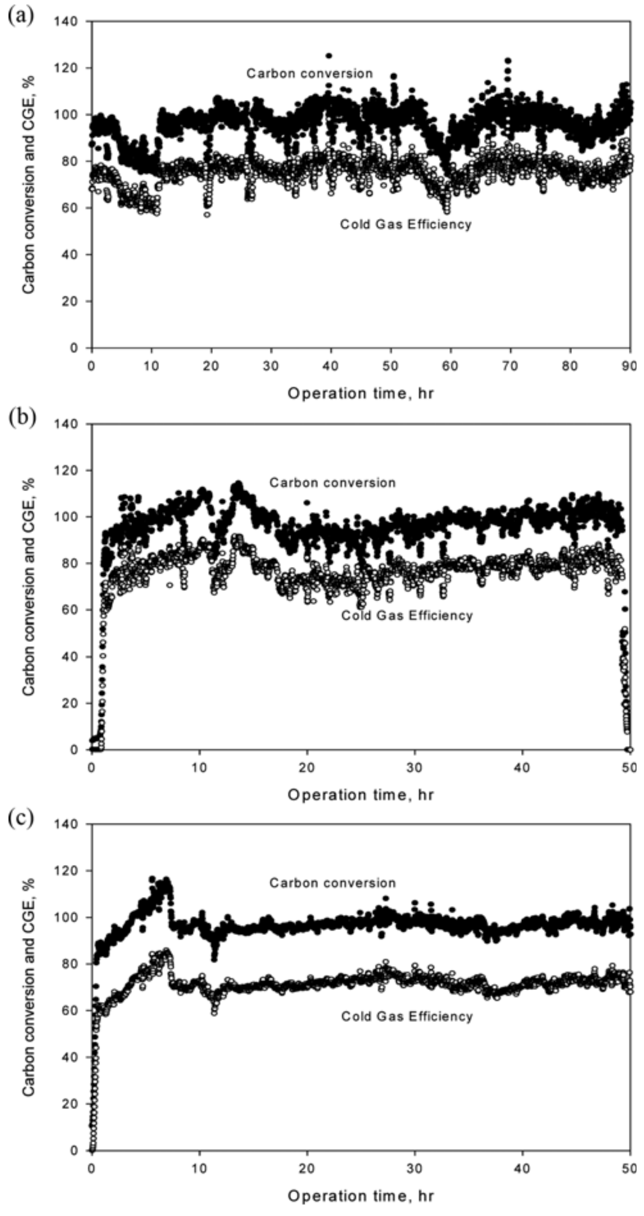

Fig. 5. Carbon conversion and cold gas efficiency results for the three tested coal types.

Cold gas efficiency is defined as the energy recovered by syngas compared with the input energy of the feed coal, which is calculated by Eq. (2). For the LG and KPU coal, average cold gas efficiency of 73.2 and 72.1% was obtained, respectively. In contrast, the average cold gas efficiency of Arch coal was 69.7%. The lower cold gas efficiency and the higher CO_2 concentration of Arch-derived syngas could be attributed to the different moisture content of the tested coals.

3. Filtration System Operations

The inlet temperature of the filtration system, the differential pressure and a backwash pressure of nitrogen are shown in Fig. 6. A backwash for LG was activated when the differential pressure

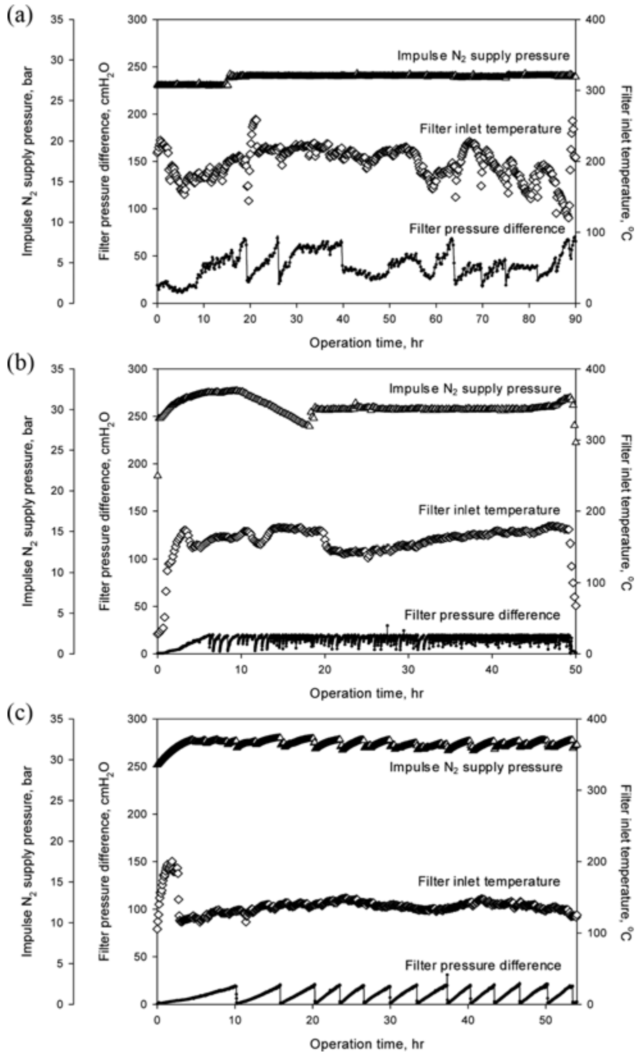

Fig. 6. Operation profiles of the filtration system for the three tested coal types.


(a) LG coal (b) KPU coal (c) Arch coal

Fig. 7. Photographs of metal filters detached from the filtration main vessel for the three tested coal types.

reached 15 cm-H_2O. At first, the backwash of 1 cycle was automatically activated in 1 h. The set values were subsequently changed to find the optimal operating conditions. As the operation continued, the differential pressure increased to approximately 60 cm-H_2O, which was caused by the accumulation of fly ash and the unreacted coal residue accumulation in the filtration system. The fly ash and the unreacted coal residue were periodically discharged through the receiver tank connected under the filtration system, and then the differential pressure was reduced across the cleaned metal filters to initiate the next filtering cycle. For LG coal in Fig. 6(a), the fly ash and unreacted coal residue caused significant dust accumulation on the filtration system during long-term continuous operation, and the formation of a particulate cross-linked layer was observed in the upper layer up to the middle of the filtration system. Fig. 7 shows the pictures taken after the gasification tests on the metal filters detached from the main filtration vessel, and the area between metal filters above one-third of the length of the filter resulted in bridged agglomerated particles as shown in Fig. 7(a). In contrast, filters after the tests for KPU and Arch coals demonstrated a clean space between metal filters, and thus a clean pattern of particle cleaning as shown in Fig. 7(b) and Fig. 7(c). For continuous operation, the particulate removal pattern in Fig. 7(b) and Fig. 7(c) should be observed.

In contrast, in the case of KPU in Fig. 6(b), the filtration system was operated at an inlet temperature range of 140-176 °C. When the differential pressure reached 20 cm-H_2O, it was configured such that the control logic would automatically precede backwashing. After 6 h, the automatic backwash cycle of approximately 1 h was activated, and then the backwash time was used to shorten the cleaning station cycle. The differential pressure during normal operation had decreased to 3 cm-H_2O after the backwash, and it subsequently increased rapidly to 10 cm-H_2O before gradually rising to 20 cm-H_2O thereafter.

For the case of Arch coal in Fig. 6(c), the nitrogen backwashing pressure was increased to 32 bar, and the inlet temperature was maintained in the range of 130-150 °C. When the differential pressure reached 20 cm-H_2O, the filter was washed for 30 s. The backwash was then performed periodically every 3 h. As shown in Fig. 6(c), the uniformly formed dust on the surface of the dust filter cake layer could be easily removed if the dust particle was smaller than the pore size of the filter. Through this type of dust filtering experiment with variation in operating parameters, a suitable operating procedure was set for the system, which sets the impulse N_2 pressure for backwashing above 30 bar, the set point for the pressure difference in the filters as 20 cm-H_2O, and the impulse time interval of 30 s for each filter bundle when the filter pressure reaches the 20 cm-H_2O value. Typically, the standard operating rule for the particulate filtering system is to use a backwashing pressure that is double the inlet gas pressure. Because the higher-pressure N_2 necessitates higher fixed and operating costs, if there is any alternative, a lower-pressure utility gas should be used.

4. Desulfurization System Operations

The supply of coal to the gasifier and syngas to the HGD system was individually controlled while ensuring the constant composition of the syngas. The syngas was fed into the HGD system at 100 Nm^3/h via two flow control valves with back pressures exerted by the produced syngas of 203-276 Nm^3/h, as shown in Fig. 4. The pressure of the desulfurizer and regenerator was maintained at 18.8-20.3 bar. For continuous operation of the HGD system, the desulfurizer/loop seal and regenerator temperatures were maintained at 550 °C and 600 °C, respectively, via an independent electric heating system.

Reactions (7) and (8) describe the sorption of H_2S and COS by zinc oxide (ZnO)-based sorbents in the desulfurizer. Moreover, the supply of oxygen to the regenerator for regeneration of the sorbent is given in reaction (9).

$$H_2S+ZnO\rightarrow ZnS+H_2O \quad (7)$$

$$COS+ZnO\rightarrow ZnS+CO_2 \quad (8)$$

$$ZnS+\tfrac{2}{3}O_2\rightarrow ZnO+SO_2 \quad (9)$$

The H_2S and COS concentrations of the syngas before and after HGD treatment are shown in Fig. 8 and Fig. 9, respectively, for three coal samples. For LG-derived syngas, H_2S and COS in the syngas ranged from 1,650-2,100 ppm and 120-250 ppm, respectively. In contrast, H_2S of 760-830 ppm and 890-960 ppm and COS of 130-140 ppm and 130-150 ppm were detected for KPU and Arch coal-derived syngas, respectively. Note that the KPU coal-derived syn-

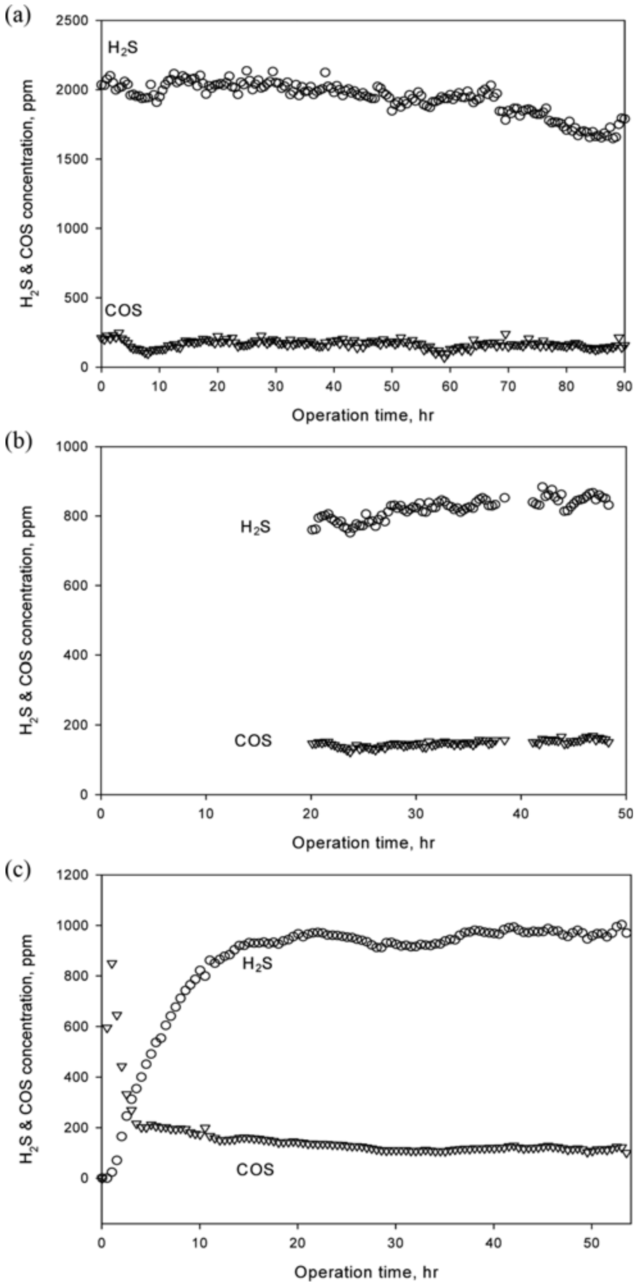

Fig. 8. Profiles of H_2S and COS concentrations in syngas before HGD treatment.

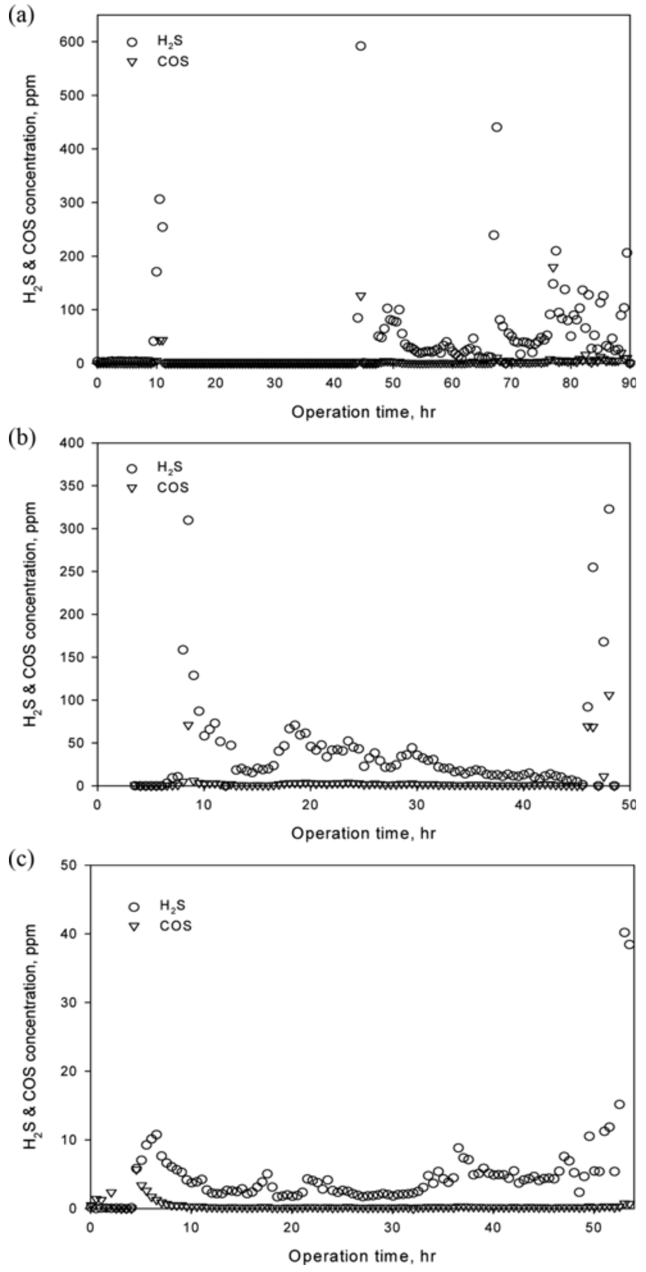

Fig. 9. Profiles of H_2S and COS concentrations in syngas after HGD treatment.

Table 5. H_2S and COS concentrations[a] in the syngas and removal efficiency

Test coal for gasification	H_2S			COS		
	Inlet (ppm)	Outlet (ppm)	Removal efficiency (%)	Inlet (ppm)	Outlet (ppm)	Removal efficiency (%)
LG	2,001.0	18.1	99.1	172.9	2.5	98.6
KPU	814.1	36.0	95.6	146.1	4.6	96.9
Arch	852.5	5.5	99.4	155.8	0.4	99.7

[a] Average values

gas was analyzed after 20 h of gasification initiation for H_2S and COS concentrations.

Table 5 lists average concentrations of H_2S and COS in the fore and aft sections of the desulfurization unit and the removal efficiencies of H_2S and COS. The removal efficiency of H_2S by the dry-sorbent desulfurization system was in the range of 95.6-99.4%,

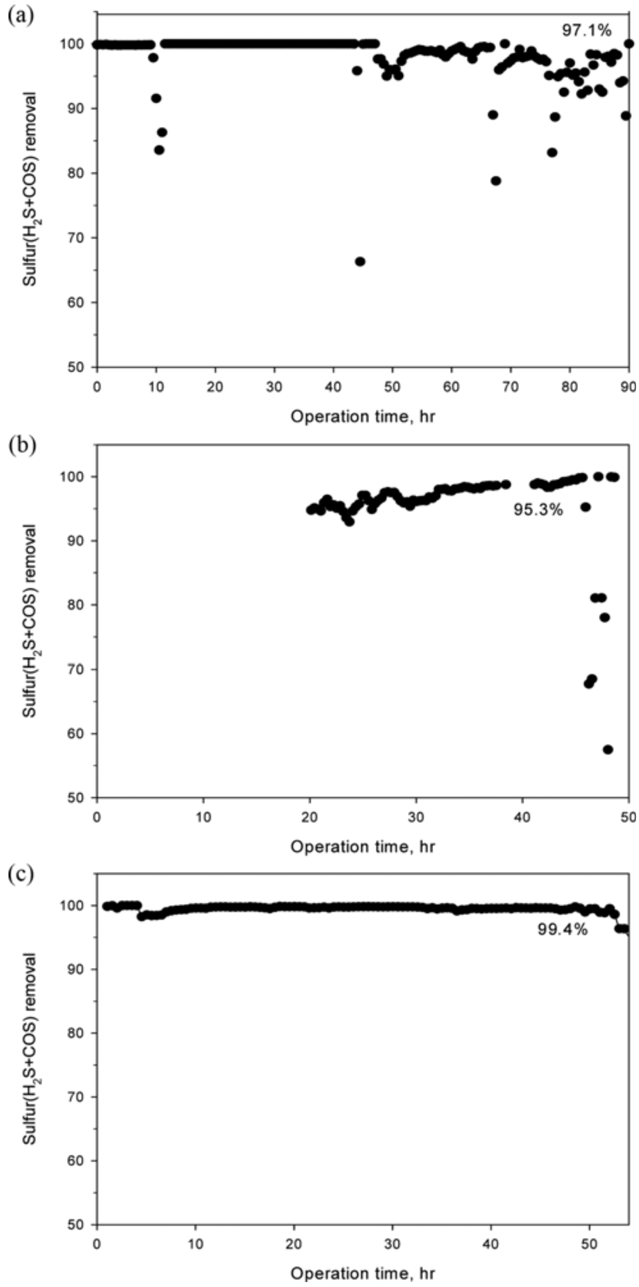

Fig. 10. Sulfur (H_2S+COS) removal efficiencies obtained by hot desulfurization for the three tested coal types.

whereas the removal efficiencies for COS ranged from 96.9-99.7%.

The removal efficiency of H_2S and COS in addition to the average sulfur (H_2S+COS) removal efficiencies of the three coal types are shown in Fig. 10. The HGD system was designed to operate at a solid circulation rate of 7-8 kg/m^2·s and handle syngas with no more than 2,000 ppm of sulfur compounds [18]. An average sulfur removal efficiency greater than 95.3% was achieved in the present study for syngas derived from the three studied coal types.

CONCLUSIONS

Syngas produced from three coal types via pilot plant gasification operation with filtration and HDG processes showed a reasonably high coal conversion rate and cold gas efficiency. The gasification temperature was selected by the melting temperature of the coal types, and the gasifier operated at the partial slagging range of 1,100-1,300 °C. The average carbon conversion for gasification using the feedstock of the three coal types was more than 96%. However, the average cold gas efficiency was 73.2% and 72.1% for the LG and KPU coals, respectively, with a lower value of 69.7% observed for the Arch coal with a moisture content of 19.15 wt%. The lower cold gas efficiency and higher CO_2 concentration observed for the Arch-derived syngas could be attributed to excessively high moisture content. The removal of the fly ash and unreacted coal residue was optimized with various operational parameters. Moreover, the sulfur removal efficiency of the coal feedstocks in the HGD system achieved 95.3-99.4% for the tested sub-bituminous coals.

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