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GASIFICATION AND
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THE HOT GAS DESULFURIZATION IN A COMPACT TWO BEDS SYSTEM INTEGRATED WITH COAL GASIFICATION AND FISHER-TROPSCH SYSTEM

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ABSTRACT

The hot gas desulfurization (HGD) technique is one of the elemental technologies of syngas purification having both thermal efficiency and very low emissions. The HGD is a novel method to efficiently remove H₂S and COS in the syngas with regenerable sorbents at high temperature and high pressure condition. We propose a compact hot gas desulfurization system by which its operability is improved in stabilizing pressure balances among units. The proposed compact two beds system has two bubbling beds, solid injection nozzle, solid conveying line, and riser. The compact desulfurization system was located between coal gasifier and Fisher-Tropsch (F-T) reactor to desulfurize syngas in order not to deactivate F-T catalyst. To check feasibility of the compact desulfurization system at high pressure condition, both cold mode and hot mode tests have been performed. In the integrated system, the compact desulfurization system has removed H₂S and COS in the syngas and supplied the cleaned syngas to the F-T reactor during the continuous operation at high pressure condition.

INTRODUCTION

The hot gas desulfurization (HGD) technique is one of the elemental technologies having both high thermal efficiency and very low emissions (1). HGD is a new method to efficiently remove H₂S and COS in the syngas with regenerable sorbents at high temperature and high pressure condition (2). Generally metal oxide sorbents have been used as regenerable sorbents. A large number of single or mixed metal oxides have been evaluated as regenerative sorbents (3,4). Zinc oxide based sorbents have a high efficiency and a lower cost, as they can effectively reduce the H₂S content in gases at ppm levels (5,6). Research Triangle Institute (RTI) international and Eastman Chemical Company have reported successful demonstration of a transport reactor-based desulfurization technology with zinc oxide based sorbents (7).

In this study, we used the compact HGD system proposed by Jo et al (8) to handle more than 20 Nm³/h of real coal-gasified syngas at around 30 bar gauge in the integrated system. Ryu et al (9) reported that in the compact two beds system the solid circulation rate increased as the hole diameter in the injection nozzle, the diameter of the injection nozzle, the solid height above the holes, and the number of holes on the injection nozzle increased. Jo et al (8) reported hydrodynamic test results using the compact HGD system. Using the proposed system, we tested the performance of zinc oxide based dry sorbents which was supplied by Korea Electric

Power Research Institute (KEPRI) to check feasibility of stable operation using real syngas at 30 bar gauge in the integrated system with gasification, syngas clean-up and F-T reactor.

EXPERIMENTAL

Apparatus

Figure 1 shows the schematic diagram of the compact two beds system (8). The major components consist of plenums, bubbling beds, solid injection nozzle, riser, cyclones. The regenerator is 1.2 m high with an internal diameter of 3 in and the desulfurizer is 2.0 m high with an internal diameter of 5 in. The inside diameter of solid injection nozzle is 1/4 in. The solid sorbent from a desulfurizer is passed through a solid injection nozzle and a riser and is collected in the cyclone. The collected sorbent goes into a regenerator passing through a standpipe to prevent the gas and sorbents from flowing backward. The syngas (Q_1) and the regeneration gas (Q_3) are added in the bottom of the beds through perforated plates, respectively and the gases for solid injection (Q_2) are added in the bottom of the bed through the solid injection nozzles.

We used ZnO based solid sorbent which was provided by the Korea Electric Power Research Institute (KEPRI). Mean particle size is 84 μm , bulk density is 0.91 g/cm^3 and BET surface area is 16.3 m^2/g . The particle shape is spherical, which is adequate for fluidized bed reactor.

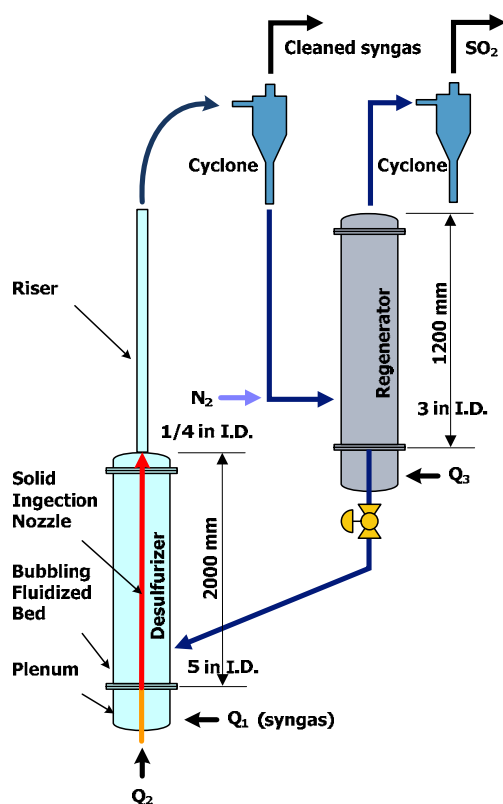


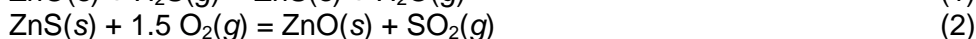
Figure 1. Schematic diagram of compact two beds system.

Procedure

The integrated system pressurized using coal-gasified syngas up to 30 bar gauge. We first pressurized HGD system up to 20 bar gauge first 2 hours using 20 Nm²/h of coal-gasified syngas of which H₂S inlet content was around 300 ~ 400 ppm. The F-T system tested catalysts below 20 bar gauge for 3 hours and then, HGD system was re-pressurized up to 30 bar gauge for 1 hour and operated at this pressure during 4 hours. During whole operation, desulfurized syngas was supplied to F-T system and H₂S content in the syngas before/after desulfurizer was detected using gas detector tube every 20 minutes.

RESULTS AND DISCUSSION

The sulfidation and regeneration reactions are as follows:



where *g* represents gas phase and *s* solid phase. The proposed system consists of desulfurizer and regenerator in order to use the solid sorbents continuously. Jo et al (8) reported hydrodynamic tests results such that the minimum fluidization velocity was 0.007 m/s and the solid circulation rate was around 2 g/s above 2 m/s of the gases for solid injection (*Q*₂). They also reported that stable operation with solid circulation could be made at the pressure of 5 bar gauge.

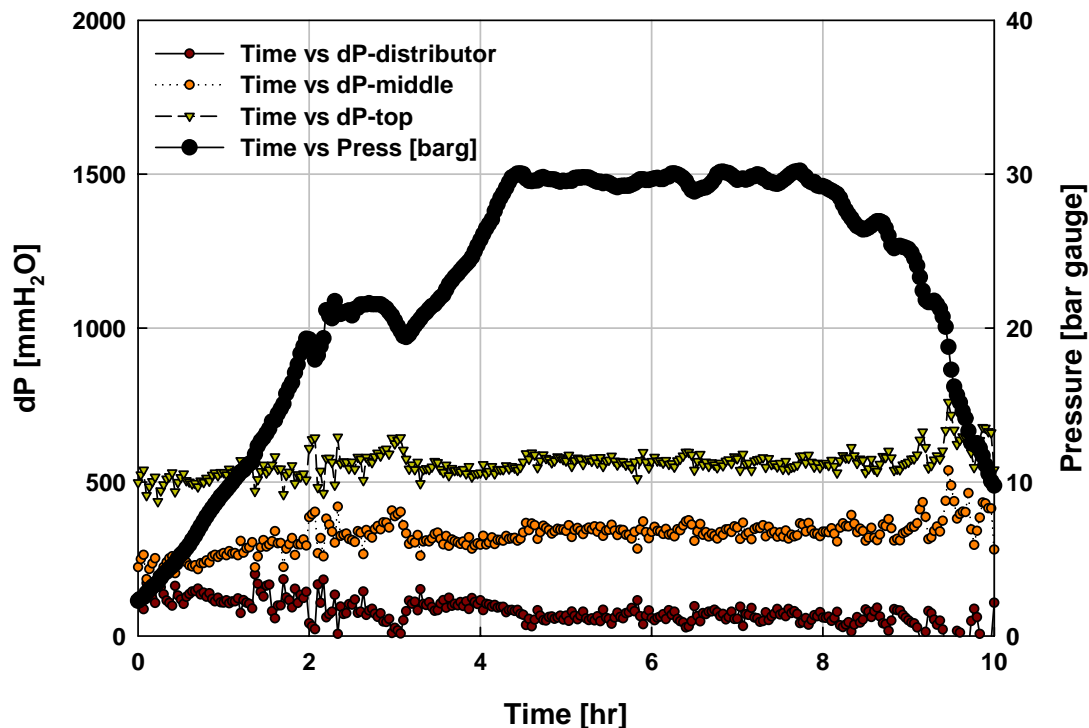


Figure 2. Desulfurizer pressure profiles in the continuous operation.

We performed the continuous operation in an integrated system at the pressure of above 30 bar. The gasification used raw materials as mixture of coal and glycerin and the continuous operations during 8 hours has been performed using coal-gasified syngas. During first 2 hours the system was pressurized up to 20 bar gauge then maintained for 1 hour. After that the system was pressurized up to 30 bar gauge and maintained during 4 hours. Figure 2 showed the system pressure and desulfurizer differential pressure profiles. The pressure in the desulfurizer was maintained in a stable manner. When the pressure has increased, the differential pressure in the reactor was a little bit fluctuated, but it was maintained stably in the constant pressure periods. The stable pressure profiles indicated that the syngas flow rate has been maintained stably in an integrated system.

Figure 3 showed desulfurizer temperature profiles. The bed temperature was maintained at 500 ~ 550°C which was a target temperature range of desulfurization. Lee et al (10) reported that the ZnO based sorbents showed about 14 to 16 wt% of sorption capacity with 70 ~ 85% of sorbent utilization in the second desulfurization cycle at 500°C using TGA. Also, Jo et al (8) reported the 99.94% of sulfur removal was achieved at 550°C of desulfurizer temperature using real coal gas. Hence we set the target temperature as below 550°C in this study. To maintain the reactor temperature, the electric heater supplied the heat of reaction since desulfurization reaction is endothermic. The temperature of the syngas was also increased by electric heater near the reactor temperature since the temperature of the syngas in the outlet of the gasifier was considerably lower than the reactor temperature.

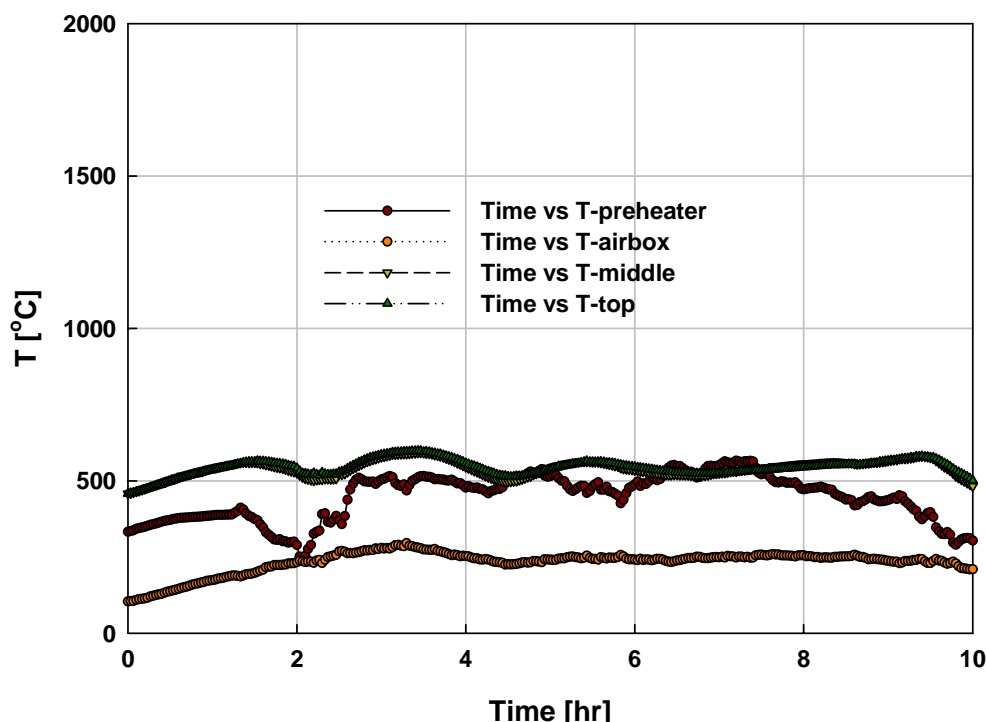


Figure 3. Desulfurizer temperature profiles in the continuous operation

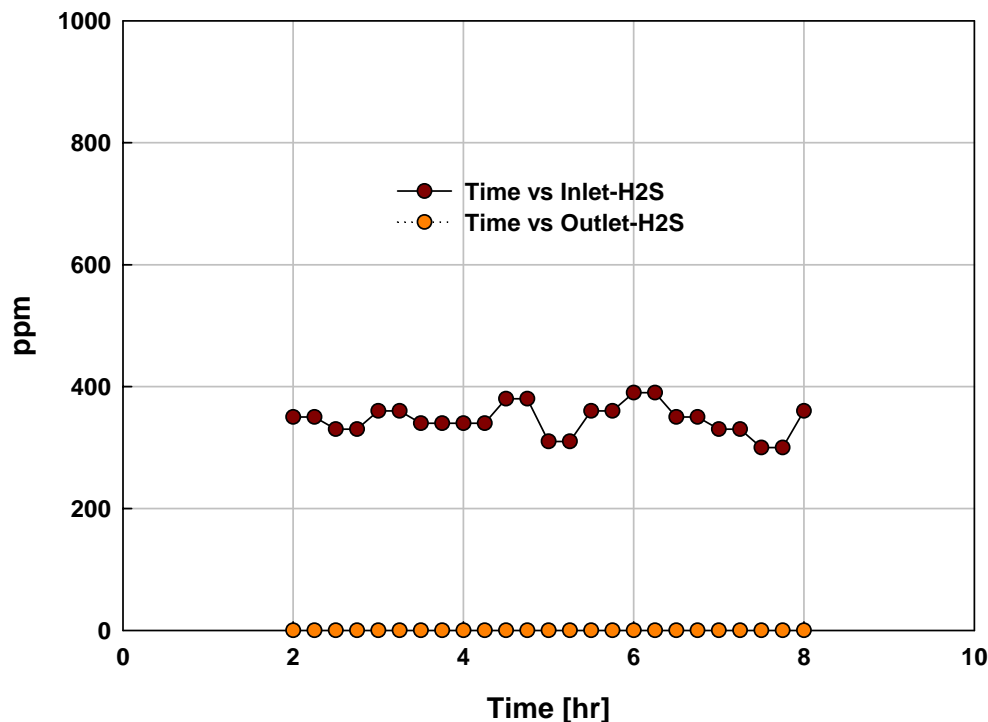


Figure 4. The H₂S content in the inlet and outlet gas streams in the continuous operation.

Figure 4 showed the H₂S content in the inlet and outlet gas stream. We measured the H₂S content using gas detector tube every 20 minutes. The H₂S content in the inlet gas stream was around 300 to 400 ppm since the feed of the gasifier was not pure coal, but the mixture of coal and glycerin. During the continuous operation, the H₂S content in the outlet gas stream was maintained below 1 ppm and the H₂S removal was above 99%. The results showed that the sulfur removal was above 99% even at the high pressure, above 30 bar gauge as well as at the middle pressure, around 5 bar gauge (8). To investigate long term stability of the proposed HGD system, we will do continuous operation in an integrated system more than 50 hours.

CONCLUSIONS

We developed HGD system which could handle above 20 Nm³/h of coal-gasified syngas treatment at around 30 bar gauge. The tests have been performed in the gasifier and F-T reactor integrated system. We experienced stable operation of pressurized HGD system with above 99% of H₂S removal and effluent sulfur was less than 1 ppm during 6 hour continuous operation. The performance of the HGD system was adequate for F-T catalyst so as not to poison catalyst since the H₂S content of the syngas flowing to the F-T reactor has been maintained below 1 ppm.

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