CONFERENCE PROCEEDINGS

Feasibility Test for CO_2 Capture by Dry sorbents in Two Fluidized Bed Reactors

Chang K. Yi*, Korea Institute of Energy Research
S.H. Jo, Korea Institute of Energy Research
Y.W. Seo, Korea Institute of Energy Research
S.D. Park, Korea Institute of Energy Research
K.H. Moon, Doosan Heavy Industries, Inc.
J.S. Yoo, Doosan Heavy Industries, Inc.
J.B. Lee, Korea Electric Power Research Institute
C.K. Ryu, Korea Electric Power Research Institute

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ABSTRACT

One of the advanced concepts for capturing CO_2 is an absorption process with dry regenerable sorbents. The purpose of our work is to develop the low cost CO_2 capture process consisting fluidized reactors. Spray-dried sorbents were used to examine the CO_2 capture characteristics in a flue gas condition.

The reactivities of pure $NaHCO_3$ material (Dongyang Chemical Co, Korea), Sorb NHR, Sorb NHR5 and Sorb NX30 were examined in a fast fluidized-bed reactor. The CO_2 removals of Sorb NHR and NHR5 are initially maintained at 100% level for a short period of time and quickly drops to 10 to 20 % removal level. However, CO_2 removal of Sorb NX30 sorbent is maintained 100% for 12 minutes. It is the first time that a dry sorbent is able to capture all of the 10% CO_2 in the flue gas within 3 sec of residence time in a fast fluidized reactor.

The feasibility of dry sorbent CO_2 capture process was also investigated in the continuous solid circulation mode between a fast fluidized carbonator of 6 m in height and a bubbling fluidized regenerator. The performance and process parameters obtained from 22 hour continuous operation are discussed.

1. INTRODUCTION

The CO_2 concentration in earth atmosphere is increased by combusting fossil fuels to generate electricity. Capture of CO_2 in flue-gas streams is essential for the control of CO_2 emissions. Capture of CO_2 will be best carried out at large point sources of emissions, such as fossil fuel-fired power plants, oil refineries, petrochemical, fertilizer and gas processing plants, iron and steel industries and paper mills. The reduction of cost for CO_2 capture is critical step in the overall greenhouse gases mediation program. Much attention has been recently directed towards cost-effective and energy-efficient CO_2 separation techniques aimed at capturing CO_2 released from fossil fuel–fired power plants and boilers [1-4]. One of the advanced concepts for capturing CO_2 is an absorption process with dry regenerable sorbents.

The process for CO_2 capture with dry regenerable sorbents consists of carbonation reactor and regeneration reactor. The following reaction occurs in each reactor:

Carbonation: $M_2CO_3 + CO_2 + H_2O \rightarrow 2\ MHCO_3 + \text{Heat}$ (1)

Regeneration: $2MHCO_3 \rightarrow M_2CO_3 + CO_2 + H_2O$ (2)

Where M is one of alkali metal.

We have adapted two fluidized bed type reactors for a carbonator and a regenerator. The fluidized reactor can be a bubbling fluidized type or a fast fluidized type, which depends on gas velocity, reactivity of sorbents. The purpose of this work is to identify chemical characteristics of sorbents in a fast fluidized-bed reactor of 0.025 m (ID).

2. EXPERIMENTS

Figure 1 shows the schematic of the fast fluidized-bed reactor. The general feature of the system includes a fast fluidized-bed reactor, a mixing zone in the lower part of the fast fluidized-bed reactor, a cyclone, a standpipe, a bubbling fluidized reactor. The fast fluidized-bed is a 5.4 m tall pipe of 0.025 m (ID). The lower mixing zone is a 0.6 m tall section of 0.035m (ID). The bubbling fluidized-bed reactor is a 1.28m tall bed of 0.1m (ID), and has a perforated distributor.

For the fluidized-bed CO_2 capture process, sorbent should have high chemical reactivity and high attrition resistance. Also, it should be regenerable over multicycle use or continuous solid circulation mode between carbonation and regeneration. Commercial grade $NaHCO_3$ solid from Dongyang Chemical Co., spray dried Sorb series sorbents (Sorb NHR, Sorb NHR5, and Sorb NX30) were used to measure

chemical reactivities in a fast fluidized-bed. Sorb series sorbents was made by Korea Electric Power Research Institute, after processing through the comminution of raw materials, a colloidal slurry preparation, a spray drying and calcinations steps [1, 3].

The reaction temperature was maintained in the range from 40 to 70 ^{o}C. The gas velocity was changed from 1.5 to 3.5 m/s. The solid circulation rate was changed from 15 to 25kg/m^2•s. The initial charge in the reactor was 7kg in the whole experimental apparatus. The simulated gas composition was 10% CO_2, 10-20% H_2O and balanced N_2.

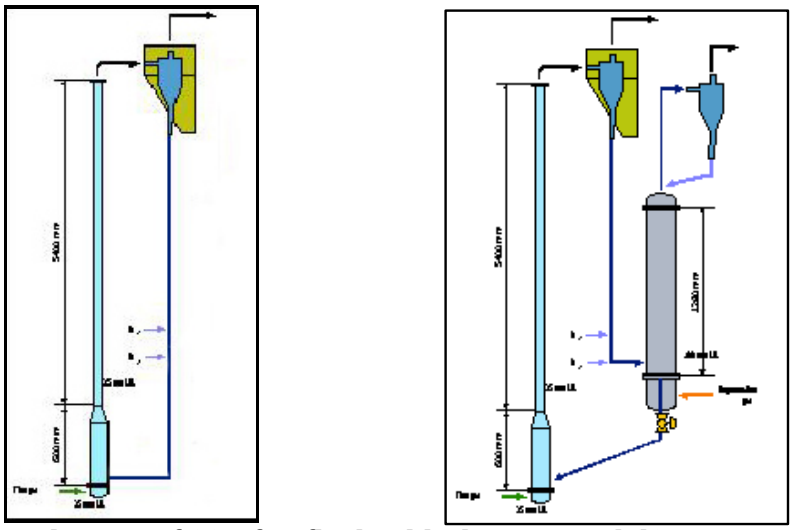

Figure 1. Schematic of (a) a fast fluidized-bed reactor and (b) two-reactor system.

3. RESULTS AND DISCUSSIONS

3.1 Reactivity of $NaHCO_3$ in a fast fluidized-bed reactor

Figure 2 shows CO_2 removal and reactor temperature profiles during carbonation reaction in a fast fluidized-bed reactor with $NaHCO_3$. It shows CO_2 removal of about 9 % at the conditions of temperature 45°C, gas velocity 2 m/s, and solid slide valve opening 25%. The residence time in a fast fluidized carbonator is 2-3 s.

Figure 3 shows the effects of reactor temperature on CO_2 removal at the conditions of 2 m/s gas velocity, 10% steam, and 30% slide valve opening. As carbonation temperature is increased, the CO_2 removal decreases as we expect in thermodynamics. The CO_2 removal is 16.5% at the temperature of 40°C and it is 3% at the temperature of 70°C.

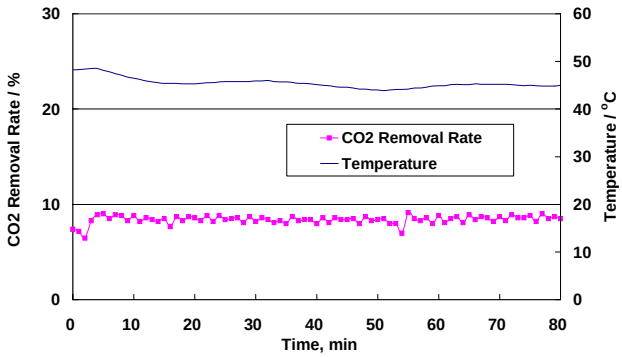

Figure 2. CO_2 removal and temperature profiles in a fast fluidized-bed reactor.

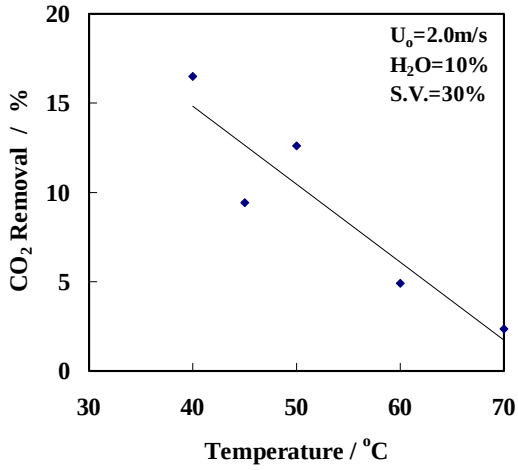

Figure 3. Effect of carbonation temperature on CO_2 removal in a fast fluidized-bed reactor

3.2 Reactivity of spray-dried sorbents in a fast fluidized bed

The reactivities of spray-dried sorbents were examined in a fast fluidized-bed reactor. The reactor was operated at the carbonation temperature of 50°C and gas velocity of 2 m/s with the initial sorbent inventory of 7kg to compare CO_2 concentration profiles in effluent gas for spray-dried Sorb NH series and NX30 sorbent form KEPRI. Figure 4 shows the comparison of CO_2 concentration profiles in effluent gas of Sorb NHR, NHR5, and NX30 in a fast fluidized-bed reactor. The CO_2 removals of Sorb NHR and NHR5 are initially maintained at 100% level for a short period of time and quickly drops to 10 to 20 % removal level. However, CO_2 removal of Sorb NX30 sorbent was maintained 100% for 12 minutes. It is the first time that a dry sorbent is able to capture all of the 10% CO_2 in the flue gas within 3 sec of residence time in a fast fluidized reactor. The sorption capacity of Sorb NX30 is also much greater than the two other sorbents and $NaHCO_3$. Sorb NX30 showed fast kinetics in a fast fluidized reactor so that it captured 10% CO_2 within 3 seconds of fast fluidized reactor condition.

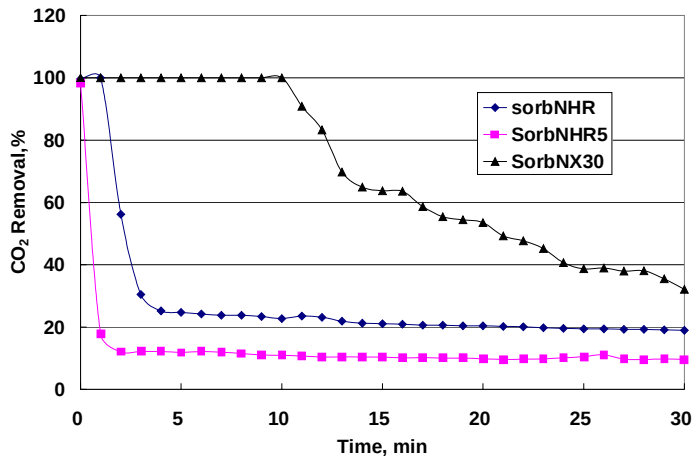

Figure 4. Comparison of sorbent kinetics and sorption capacities of spray dried sorbents, Sorb NHR, NHR5, and NX30.

3.3 Continuous operation in two-reactor system

The feasibility of dry sorbent CO_2 capture process was also investigated in the continuous solid circulation mode between a fast fluidized-bed carbonator of 6 m in height and a bubbling fluidized-bed regenerator. Flue gas from a boiler or a combustor enters a fast fluidized-bed carbonator, CO_2 in the flue gas is captured by recycled sorbent selectively in a carbonator where sorbent and flue gas are well mixed and cocurrently flow. Cleaned flue gas and solid sorbent particles move to a cyclone in which solid sorbent is separated and goes to a regenerator. Saturated sorbent with CO_2 and H_2O is regenerated in a bubbling fluidized-bed regenerator by external heat or steam. Regenerated sorbent sends to the fast fluidized-bed carbonator again and is ready to capture CO_2 in the flue gas.

Figure 5 shows carbonator temperature profile for 22 hours during continuous operation. The carbonator temperature was maintained at 40°C??50 ^{o}C???. Since carbonation of sorbent with steam and CO_2 is exothermic reaction, it is really difficult to maintain carbonation optimum temperature in a fixed bed or a moving bed, however, the system with a fast fluidized-bed carbonator is well maintained at its temperature with heat transfer unit of water jacket type. The operating conditions were changed twice during test. The solid circulation rate was increased after 9 h run and the carbonator temperature was increased to 50°C after 15 h run. When the solid circulation was increased, the carbonator temperature was little changed as shown in Figure 5.

Figure 6 shows pressure drops at four positions of bottom section, middle I section, middle II section and upper section in a fast fluidized-bed reactor. The differential pressure at bottom section is about 100 mmH_2O, that of middle I section is 300mmH_2O, that of middle II section is 150 to 450 mmH_2O, and that of upper section is about 50mmH_2O, that give us a hint to estimate design value of fan power.

Figure 7 shows CO_2 concentration profile from effluent gas of carbonator and CO_2 removal for 22 hour continuous operation. The CO_2 concentration in inlet simulated gas is maintained at 11.2% at wet basis. Initially, CO_2 removal drops quickly and maintains about 40%. After solid circulation rate is

increased at 9 hr, regenerator temperature is decreased from 210°C to 170°C resulting in the decrease of CO_2 removal. When the carbonator temperature is increased from 40°C to 50°C, the CO_2 removal is decreased to 30%. For 22 hours the CO_2 capture process is stably maintained in the continuous solid circulation mode. Dry sorbent CO_2 capture process consisting of a fast fluidized-bed carbonator of 6 m in height and a bubbling fluidized-bed regenerator is feasible as shown in Figure 5, 6 and 7.

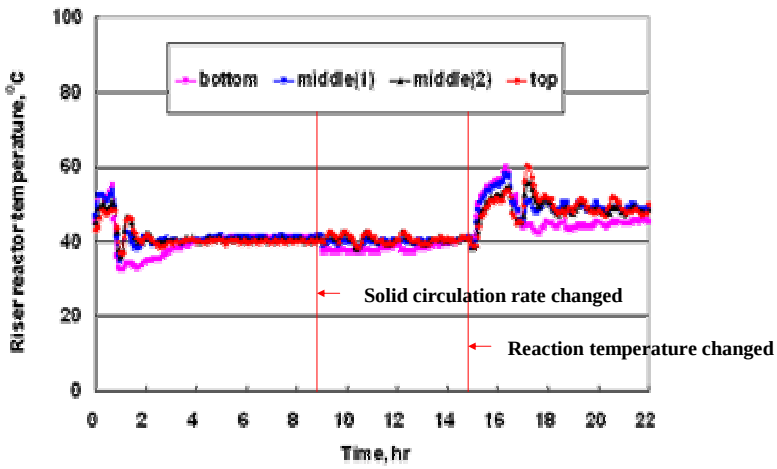

Figure 5. Carbonator temperature profile during continuous operation.

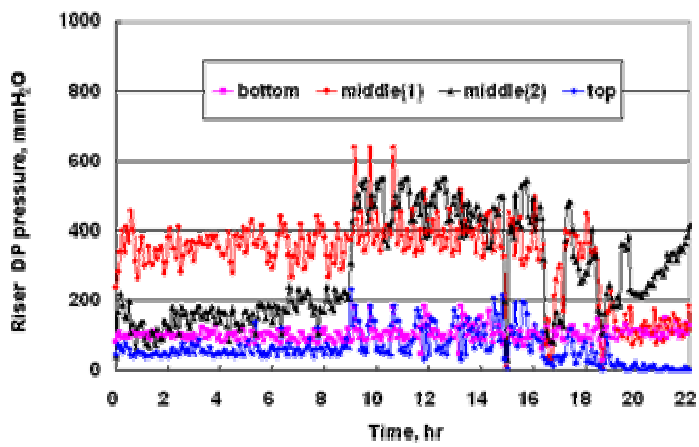

Figure 6. Axial differential pressure profile in carbonator.

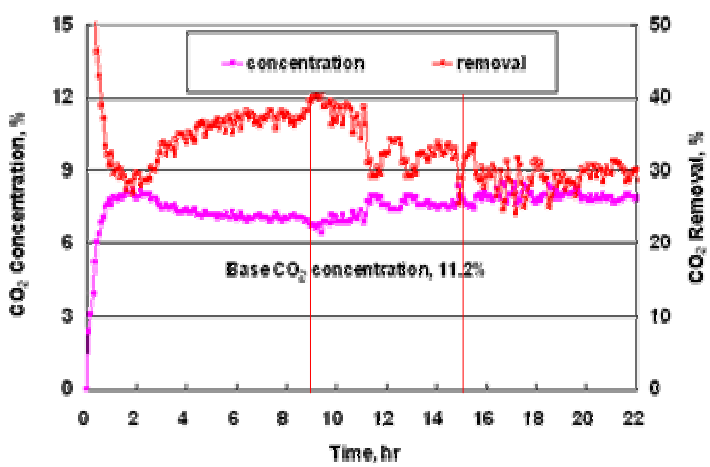

Figure 7. CO_2 concentration profile in effluent gas of carbonator and CO_2 removal.

We changed variables such as carbonator temperature, gas velocity in a fast fluidized-bed carbonator, steam content in the feed gas, solid circulation in continuous operation states to see the CO_2 removal capacity of sorbent. Base operating conditions are 40°C carbonator temperature, 2 m/s gas velocity in a fast fluidized-bed carbonator, 10% steam content in the inlet gas, 22 kg/m^2•sec solid circulation rate.

Figure 8 shows the effects of carbonator temperature on CO_2 removal at the conditions of gas velocity 2m/s, steam 10%, and slide valve opening 30%. As carbonation temperature is increased, the CO_2 removal decreases slightly from 30% to 29% as we expect in thermodynamics.

Figure 9 shows the effects of gas velocity on CO_2 removal at the conditions of carbonation temperature 40°C, steam 10%, and a slide valve opening 30%. The CO_2 removal is 37% at the gas velocity of 1.7m/s and 23% at the gas velocity of 3m/s. The CO_2 removal is decreasing as gas velocity is increased because of the decrease of the contact time between sorbent and gas.

Figure 10 shows the effects of steam content in the inlet gas on CO_2 removal at the conditions of carbonator temperature 40°C, gas velocity 2m/s, and a slide valve opening 30%. It is not easy to inject needed steam quantity because steam content at a temperature depends on reaction temperature. We increased the bubbler temperature to increase the steam content. As the carbonator temperature is increased, steam content at saturated condition increases. We used a bubbler for the steam injection. All of the simulated flue gas passes through a water bubbler in which water temperature is controlled by insulated heaters. When we inject 30% of steam at 40°C carbonator temperature, real steam content in feed gas is smaller since condensation occurred. However even water drops can affect sorbent activation at the surface of the sorbent. The CO_2 removal is 22% at the steam content of 7% and it is increased to 43% at the steam content of 30%. Carbonation reaction should have equivalent mole of H_2O and CO_2 as shown in equation (1). According to the result, it is expected that the 3 seconds of reaction time is not enough for the mass transfer of steam into the pore or surface of the sorbent.

Figure 11 shows the effects of solid circulation rate on CO_2 removal. As the solid circulation rate depending on slide valve opening is increased, the CO_2 removal increases. It is considered that solid holdup is increasing in the riser reactor and the contact frequency between active sorbent and CO_2 gas is increasing as solid circulation rate is increased. The figure shows that the CO_2 removal is increased to 33% at the solid circulation rate 42 kg/m^2/s.

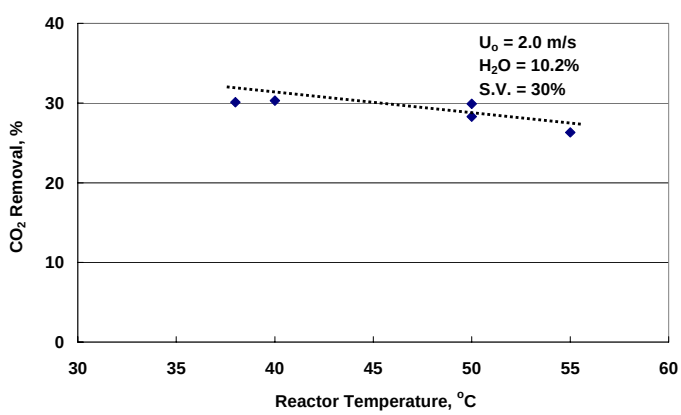

Figure 8. Effects of carbonator temperature on CO_2 removal during continuous operation..

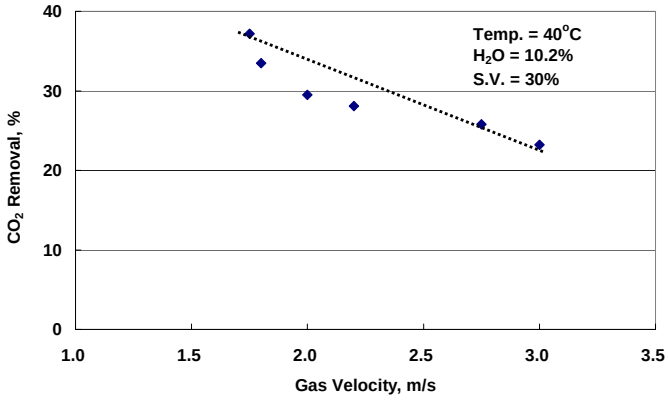

Figure 9. Effects of gas velocity on CO_2 removal during continuous operation..

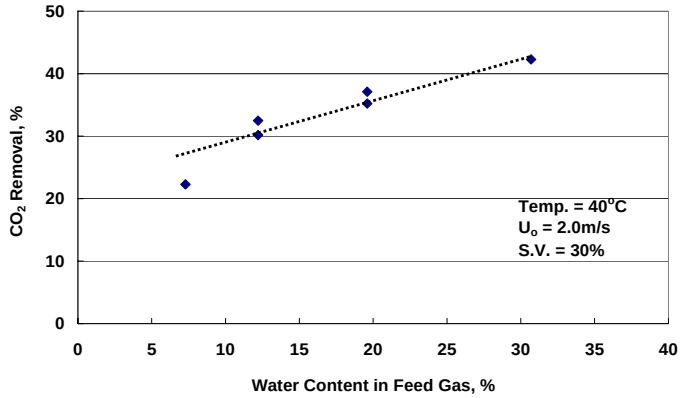

Figure 10. Effects of steam content in the inlet gas on CO_2 removal during continuous operation.

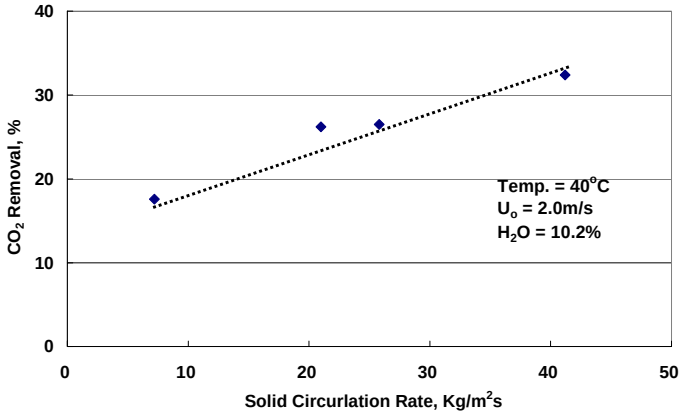

Figure 11. Effects of solid circulation rate on CO_2 removal during continuous operation..

4. CONCLUSIONS

The reactivities of pure $NaHCO_3$ solid, Sorb NHR, NHR5, and NX30 sorbents were examined in a fast fluidized-bed reactor. The CO_2 removal of pure $NaHCO_3$ solid was maintained from 9% to 15% when variables were changed. The CO_2 removals of Sorb NHR and NHR5 are initially maintained at 100% level for a short period of time and quickly drops to 10 to 20 % removal level. However, Sorb NX30 sorbent showed fast kinetics in a fast fluidized-bed reactor so that it capture 100% CO_2 in the flue gas within 3 seconds of fast fluidized reactor condition.

The feasibility of dry sorbent CO_2 capture process was also investigated in the continuous solid circulation mode between a fast fluidized-bed carbonator and a bubbling fluidized-bed regenerator. Effects of several variables [e.g., carbonation temperature (40 to 55°C), gas velocity (1.7 to 3.0 m/s), H_2O concentration (7 to 30%), and solid concentration (7 to 41 kg/m^2•s)] on sorbent activity were examined. The CO_2 removal was increased from 18% to 43% as gas velocity is decreased, as carbonation temperature is decreased, or as solid circulation rate is increased.

We are planning future work to improve CO_2 removal efficiency in continuous operation of carbonation and regeneration by optimizing sorbents and system

ACKNOWLEDGEMENT

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