

The effect of CO₂ or steam partial pressure in the regeneration of solid sorbents on the CO₂ capture efficiency in the two-interconnected bubbling fluidized-beds system

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Abstract—The effect of CO₂ or steam partial pressure in the regeneration of CO₂ solid sorbents was studied in the two-interconnected bubbling fluidized-beds system. Potassium-based dry solid sorbents, which consisted of 35 wt% K₂CO₃ for CO₂ sorption and 65 wt% supporters for mechanical strength, were used. To investigate the CO₂ capture efficiency of the regenerated sorbent after the saturated sorbent was regenerated according to the CO₂ or steam partial pressure in the regeneration, the mole percentage of CO₂ in the regeneration gas was varied from 0 to 50 vol% with N₂ balance and that of steam was varied from 0 to 100 vol% with N₂ balance, respectively. The CO₂ capture efficiency for each experimental condition was investigated for one hour steady-state operation with continuous solid circulation between a carbonator and a regenerator. The CO₂ capture efficiency decreased as the partial pressure of CO₂ in the fluidization gas of the regenerator increased, while it increased as that of steam increased. When 100 vol% of steam was used as the fluidization gas of the regenerator, the CO₂ capture efficiency reached up to 97% and the recovered CO₂ concentration in the regenerator was around 95 vol%. Those results were verified during 10-hour continuous experiment.

Key words: CO₂ Capture, Solid Sorbent, CO₂ Partial Pressure, Steam Partial Pressure, Capture Efficiency, Fluidized-beds

INTRODUCTION

In today's world, industrialization makes the energy demand of the whole world increase so that the climate change has been caused by the emission of greenhouse gas. CO₂, one of the greenhouse gases, has the largest contribution from human activities for the climate change. To reduce the concentration of CO₂ in the earth's atmosphere, net CO₂ emissions into the atmosphere should be reduced. In general, fossil fuel-fired power plants contribute approximately 40% of total CO₂ emission from energy sources [1]. To sustain continuous use of fossil fuels for power generation, CO₂ capture and storage (CCS) will be an urgent and important option for mitigating global warming [2,3].

One of the improved techniques for post-combustion CO₂ capture is chemical absorption with solid sorbents such as alkali metal-based sorbents [4-18], liquid-impregnated clay solid sorbents [3,19,20], and molecular basket sorbents [21]. Liquid-impregnated clay solid sorbents and molecular basket sorbents have been reported that the sorption capacity is higher and the regeneration energy is less than the alkali metal-based sorbents, but any continuous process application data were not reported. On the other hand, several continuous operation results have been reported using dry alkali-metal sorbents by Yi et al. [5], Park et al. [13] and Nelson et al. [15].

The reaction using alkali metal-based solid sorbents for the CO₂ capture is as follows:



where M represents alkali metal, g represents gas phase, and s represents solid phase. The reaction is reversible and the forward reac-

tion is highly exothermic, so heat control will be an important factor in the real commercial process. Thus, a fluidized-bed reactor can be a good solution for this process [22]. Yi et al. [5] proposed a CO₂ capture process which consists of a fast fluidized-bed type carbonator and a bubbling fluidized-bed type regenerator. Ryu et al. [23] proposed a two-interconnected bubbling fluidized-beds system which is favorable for the slow reactions and longer contact time between gas and solid. To improve the efficiency of the CO₂ capture process, Park et al. [24] developed a CO₂ capture system using the concept of a two-interconnected bubbling fluidized-beds system, which consists of a bubbling fluidized-bed type carbonator and a bubbling fluidized-bed type regenerator. Park et al. [24] reported the optimal operating conditions to get above 80% of CO₂ removal efficiency of the two-interconnected bubbling fluidized-beds system. To utilize the recovered CO₂ for CCS or other utilizations, the high-concentrated CO₂ should be recovered in a regenerator. When the fluidized-bed reactor is used for solid sorbent regeneration, the fluidization gas should be introduced to make the beds bubbling phase. Thus, CO₂ or steam, which is a product of the regeneration reaction, should be used as a fluidization gas in a regenerator to recover high-concentrated CO₂. However, the regenerability of the solid sorbent could be reduced as the partial pressure of the product is increased. So, we investigated the CO₂ capture efficiency of the solid sorbent by increasing the partial pressure of CO₂ or steam in the regenerator by introducing CO₂ or steam as a fluidization gas itself.

EXPERIMENTAL

1. Material and Apparatus

The solid sorbents used in this study consisted of K₂CO₃ of 35% for CO₂ sorption and supporters of 65% for mechanical strength. Fig. 1 shows a schematic diagram of the CO₂ capture process com-

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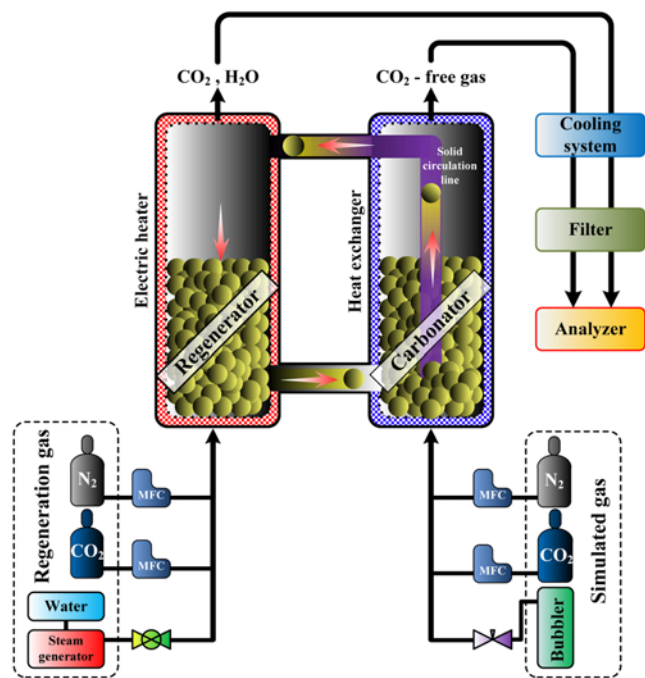


Fig. 1. Schematic diagram of the CO₂ recovery process composed of two bubbling fluidized-bed reactors.

posed of the two-interconnected bubbling fluidized-bed reactors. The dimensions of both carbonator and regenerator are the same: 0.11 m of I.D. and 1.2 m of height. The solid circulation line with holes for transporting dry solid sorbents is installed in the middle of the carbonator. The major components are a steam generator which supplies steam in a regenerator quantitatively, mass flow controllers for introducing fluidization gases in each reactor and a solid circulation line, heat exchange tubes which control the temperature of carbonation reaction, an electric heater for supply the tempera-

ture of regeneration reaction, a cooling system for condensation of moisture discharged after reaction, filters for collection of abrasion particle and a gas analyzer for measuring gas concentration.

2. Experiments and Conditions

Every experiment in this study consists of four phases described in Fig. 2 which shows the experimental procedure to obtain the steady-state results when 100 vol% of N₂ was used as a fluidization gas in the regenerator. In phase 1, the temperature of a regenerator increased by an electric heater and that of a carbonator increased by the solid sorbents circulated between two reactors. Since the solid circulation rate was constant, differential pressure of both reactors was maintained stably. It usually took one hour to reach the target temperature of both reactors. In phase 2, the saturated water vapor was supplied to a carbonator using a bubbler. In this phase, pure N₂ was introduced to a carbonator and became a saturation condition. The solid sorbents in a carbonator reacted with a water vapor and the density of the solid sorbents was changed so that the differential pressure initially fluctuated. In phase 3, after both temperature and differential pressure profiles of both reactors became stable, 10 vol% of CO₂ with N₂ balance was introduced to a carbonator after passing through a bubbler. In this phase, the simulated flue gas consisted of CO₂, water vapor and N₂. When the simulated flue gas was introduced to a carbonator, cooling water was started to circulate in the jacket of the carbonator to maintain the carbonation temperature since the carbonation reaction was exothermic. The solid sorbents, which absorbed CO₂, were circulated from the carbonator to the regenerator. It took around one hour for the solid sorbents to become chemical equilibrium state. In phase 4, a steady-state was reached so that the CO₂ concentration of both reactors was maintained stably. The steady-state experimental data were obtained in this phase. The CO₂ removal efficiency was analyzed using phase 4 data.

The CO₂ capture efficiency of the carbonation and the regeneration was investigated according to various experimental conditions using N₂, CO₂ and steam as a regeneration gas. In the continuous system, the CO₂ concentration in the simulated flue gas was about 10 vol% in dry basis. The reaction temperature in the carbonator and regenerator was maintained at about 70 °C and above 150 °C, respectively. The content of water vapor introduced in a carbonator using a bubbler was 19.7%, which was the saturated condition of 60 °C, and the solid circulation rate was 10.3 kg/m²·sec. The gas velocity in a carbonator and a regenerator was maintained 0.05 m/sec and 0.06 m/sec, respectively. The gas flow line from the bubbler to the carbonator was preheated by electric heater to prevent water vapor condensation. The mole fraction of CO₂ introduced in a regenerator varied from 0 to 50 vol% with N₂ balance. The mole fraction of steam varied from 0 to 100 vol% with N₂ balance. The steam generator was used to supply pure steam, and the volumetric pump was used to manage the mole fraction of steam in the fluidization gas by controlling the amount of water injected to the steam generator. The residence time of solid sorbents in the carbonator and the regenerator was about 12 min and 10 min. The differential pressure of the carbonator and regenerator was maintained about 400 mmH₂O and 320 mmH₂O, respectively, so that the contact time between gas and solid in the carbonator and regenerator was about 8 seconds and 5 seconds, respectively. The 10-hour continuous experiment was also performed in the phase 4 status using 100 vol% of steam as the fluidization gas of a regenerator. Table 1 shows exper-

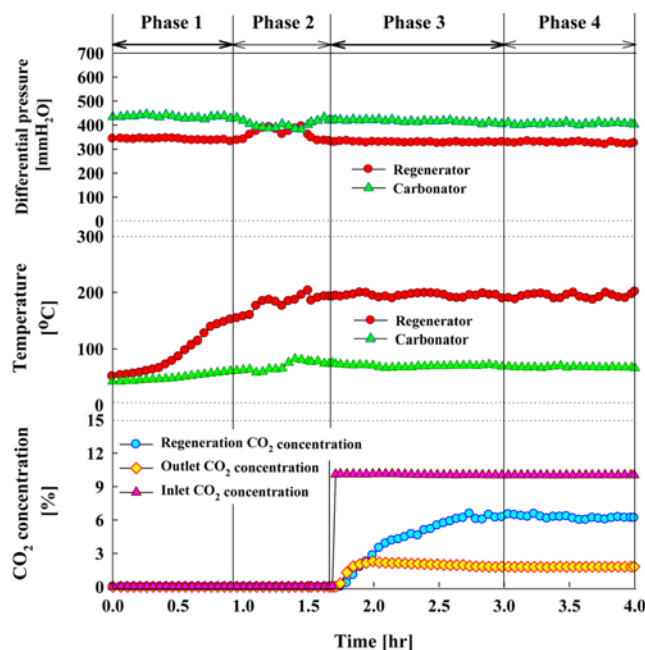
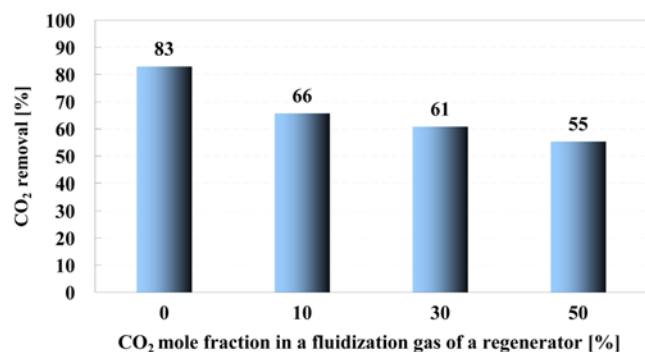
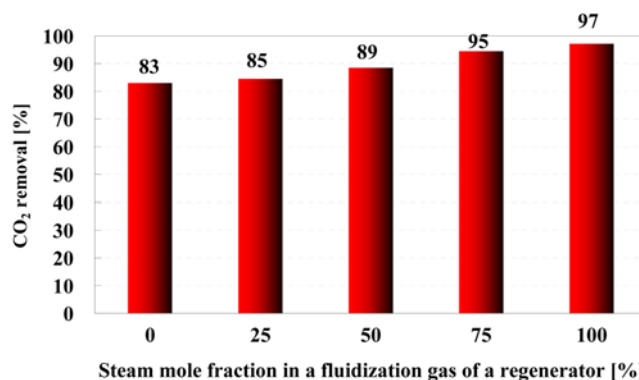


Fig. 2. Operating procedures.

Table 1. Experimental conditions of the CO₂ recovery process composed of two bubbling fluidized-bed reactors

Item	Unit	Carbonator			Regenerator				
		Simulated flue gas			Fluidization gas				
Inlet gas	vol%	CO ₂	10 (Dry basis)	CO ₂	0	10	30	50	
		H ₂ O	19.7	H ₂ O	0	25	50	75	100
		N ₂	Balanced	N ₂					
Temperature	°C	70			>150				
Gas velocity	m/s	0.05			0.06				
Pressure	atm				1				
Solid inventory	kg				10				
Solid circulation rate	kg/m ² ·s				10.3				

**Fig. 3. Effect of CO₂ mole fraction in a fluidization gas of a regenerator on CO₂ removal in the process.****Fig. 4. Effect of steam mole fraction in a fluidization gas of a regenerator on CO₂ removal in the process.**

imental conditions.

RESULTS AND DISCUSSION

Fig. 3 shows the effect of CO₂ mole fraction in the fluidization gas of a regenerator on the CO₂ removal. The mole fraction of CO₂ fed to a regenerator was varied from 0 to 50 vol% with N₂ balance. The CO₂ removal decreased from 83% to 55% as the mole fraction of CO₂ in the fluidization gas increased from 0 to 50 vol%. The CO₂ removal tended to decrease as the mole fraction of CO₂ in the regenerating fluidization gas increased so that there was no more decrease of the CO₂ capture efficiency for higher CO₂ mole fraction over 50 vol%. This result was fairly obvious since the partial pressure of CO₂, which was a product in the Eq. (1), in the regenerator increased as the mole fraction of CO₂ in the fluidization gas increased. Based on the chemical equilibrium data [25], the increase of CO₂ partial pressure in the regenerator made the system a new chemical equilibrium state so that the regenerability of the solid sorbents decreased at the given temperature. Thus, the regeneration temperature should be shifted to a high range and the additional regeneration energy for regeneration should be needed in order to increase CO₂ removal efficiency as the mole fraction of CO₂ in the fluidization gas increases.

Fig. 4 shows the effect of steam mole fraction in the fluidization gas of a regenerator on the CO₂ removal. The mole fraction of steam introduced to a regenerator was varied from 0 to 100 vol% with N₂

balance. The CO₂ removal increased from 83% to 97% as the mole fraction of steam in the fluidization gas increased from 0 to 100 vol%. The CO₂ removal increased as the mole fraction of steam in the fluidization increased, while it decreased as that of CO₂ increased. This result indicated that the increase of steam in a regenerator caused a positive effect on the CO₂ removal. Lin et al. [26] reported energy analysis of CaCO₃ calcination by steam or CO₂ stream in a calciner. It was mentioned that the CaCO₃ decomposition conversion and the content of active CaO were much higher in the steam stream than those in CO₂ stream and the calcination energy per active CaO in the steam stream was lower than that in CO₂ stream. It means that the calcination energy consumed during regeneration reaction in the steam stream is more economically advantageous than that in CO₂ stream. Seo et al. [6,7] reported that the CO₂ removal increased when the pretreatment time of water vapor for the alkali-based solid sorbents increased. It was concluded that the increase of steam in the regenerator played a role of water vapor pretreatment for the sorbents circulated between the carbonator and the regenerator. Thus, CO₂ sorption capacity of the sorbents in the carbonator was increased by pretreating the water vapor. As a result, the CO₂ capture efficiency increased by both the reduction of the regeneration energy and the effect of water vapor pretreatment.

Fig. 5 shows the experimental results of 10-hour continuous operation using 100 vol% of steam as the fluidization gas in the regenerator. During 10 hours, the CO₂ removal was maintained at the level of 95% and the recovered CO₂ concentration in the regenera-

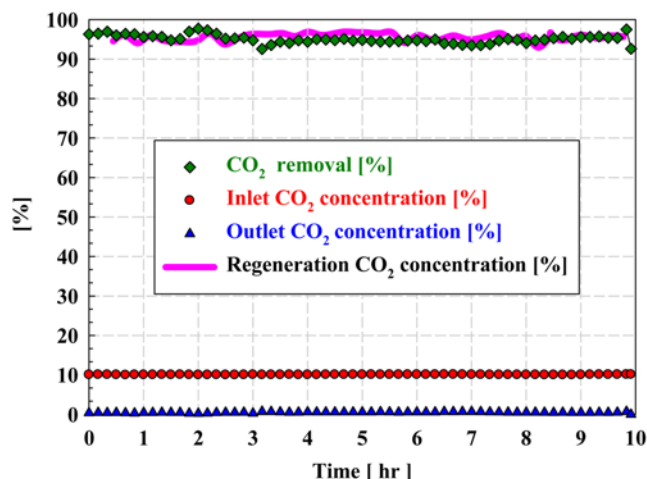


Fig. 5. Profile of CO₂ concentration by 100 vol% of steam in a regenerator for 10-hours.

tor was at the level of 95 vol%. Based on this result, high-concentrated CO₂ gas and high efficiency of CO₂ removal could be obtained by using pure steam as the fluidization gas in a regenerator.

CONCLUSIONS

The effect of the mole fraction of CO₂ or steam as a fluidization gas in a regenerator on CO₂ removal was investigated using a continuous system composed of two-interconnected bubbling fluidized-beds. The mole percentage of CO₂ introduced in a regenerator was varied from 0 to 50 vol% with N₂ balance and that of steam was varied from 0 to 100 vol% with N₂ balance. It was verified that the CO₂ capture efficiency decreased by increasing the CO₂ partial pressure, while it increased by increasing the steam partial pressure. When the 100 vol% of steam was used as the fluidization gas, the CO₂ capture efficiency increased up to 97%. The average CO₂ capture efficiency measured for 10-hour continuous experiments was the level of 95%, and the recovered CO₂ concentration in a regenerator was the level of 95 vol%. It was concluded that high-concentrated CO₂ gas and high efficiency of CO₂ removal could be obtained from a regeneration gas when pure steam as the fluidization gas in a regenerator was used.

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