*Refereed Proceedings*

*The 13th International Conference on Fluidization - New Paradigm in Fluidization Engineering*

Engineering Conferences International *Year* 2010

# A MODEL ON AN ENTRAINED BED-BUBBLING BED PROCESS FOR CO2 CAPTURE FROM FLUE GAS

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# A MODEL ON AN ENTRAINED BED-BUBBLING BED PROCESS FOR $CO_2$ CAPTURE FROM FLUE GAS

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## ABSTRACT

A simplified model has been developed to investigate effects of important operating parameters on performance of an entrained-bed absorber and bubbling-bed regenerator system collecting $CO_2$ from flue gas. The particle population balance was considered together with chemical reaction to determine the extent of conversion in both absorber and regenerator. Effects of several absorber parameters was tested in a laboratory scale process. The $CO_2$ capture efficiency decreased as temperature or gas velocity increased. However, it increased with static bed height or moisture concentration. The $CO_2$ capture efficiency was exponentially proportional to each parameter. Based on the absolute value of exponent of the parameter, the effect of gas velocity, static bed height, and moisture content was a half, one third, and one fourth as strong as that of temperature, respectively.

## INTRODUCTION

Accumulation of $CO_2$ in the atmosphere is being recognized firmly as one of important causes accelerating the global warming. Many studies have been carried out finding ways to remove $CO_2$ from flue gas massively with regenerable sorbents which are based on Na, K, and Ca (1-11). The fluidized-bed $CO_2$ capture system consisting of an entrained-bed absorber and a bubbling-bed regenerator is used due to several potential advantages. The entrained bed can decrease the absorber diameter because of its high gas velocity. It is better to maintain uniform temperature distribution and therefore is easier to operate than the bubbling bed. A mathematical model can provide a tool for investigating the effects of various operating parameters on the reactor performance in advance. It is also good to save money and efforts in systematic understanding of experimental result, design and operation. However, there have been no available reports on steady state analysis of the present process yet.

The purpose of this study was to develop a simplified model to investigate effects of important operating parameters on the efficiency of an entrained bed – bubbling bed $CO_2$ capture process using a Na-based regenerable sorbent. A particle population balance was considered in both beds assuming negligible effect of chemical reaction. Reaction rates for absorption and regeneration were determined by kinetic rates measured with a thermo-gravimetric analyzer (TGA) and an experimental result obtained from the KIER's lab-scale continuous process. Effects of several absorber parameters was investigated in a laboratory scale process.

## MODEL

Figure 1 shows the schematic diagram of the process considered in this study, which uses an entrained-bed absorber and a bubbling-bed regenerator. Flue gas containing $CO_2$ is introduced to the absorber and fluidizes the bed of sorbent particles. Sodium-based sorbent particles were used to absorb $CO_2$ according to the following reaction:

$$Na_2CO_3(s) + CO_2(g) + H_2O(g) = 2NaHCO_3(s) \quad (1)$$

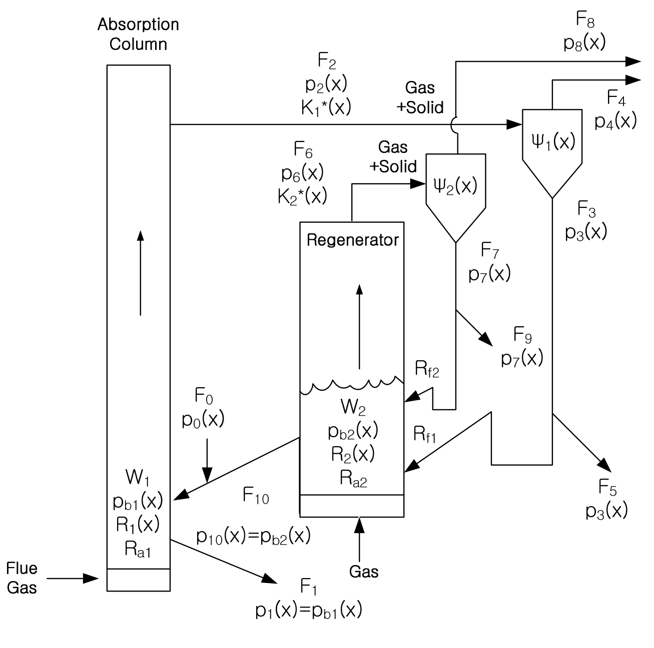


Figure 1. Process flow diagram.

Reacted sorbent particles are carried out of the absorber, collected by a cyclone, and fed to the regenerator ($R_{f1}$=1, $F_1$=$F_5$=0). Steam is introduced to the regenerator and fluidizes the bed of sorbent particles. Sorbent particles containing $CO_2$ are regenerated according to the reverse reaction. Regenerated sorbent particles are returned to the absorber. The fractional collection efficiency of the cyclone ($\psi_i(x)$) is assumed as that of Lapple(12). We assumed a well-mixed state of bed particles in both reactors ($p_1(x) = p_2(x) = p_{b1}(x)$, $p_{10}(x) = p_{b2}(x)$). The steady state particle population balance in the absorber ($i$ =1) and regenerator ($i$ =2) gives:

$$\frac{dp_{bi}(x)}{dx} + \sum_{k=1}^{2} \alpha_{ik}(x) p_{bk}(x) - \alpha_{i3}(x) = 0 \quad (i = 1,2) \tag{2}$$

$$\alpha_{i1}(x) = \delta_{1i} \left[ \frac{F_1 + K_1^*(x)}{W_1 R_1(x)} + \frac{1}{R_1(x)} \frac{dR_1(x)}{dx} - \frac{3}{x} \right] - \frac{\delta_{2i} R_{f1} \psi_1(x) K_1^*(x)}{W_2 R_2(x)} \tag{3a}$$

$$\alpha_{i2}(x) = -\frac{\delta_{1i} F_{10}}{W_1 R_1(x)} + \delta_{2i} \left[ \frac{F_{10} + K_2^*(x)(1 - R_{f2}\psi_2(x))}{W_2 R_2(x)} + \frac{1}{R_2(x)} \frac{dR_2(x)}{dx} - \frac{3}{x} \right] \tag{3b}$$

$$\alpha_{i3}(x) = -\frac{\delta_{1i} F_0 p_0(x) + R_{ai} p_{ai}(x)}{W_i R_i(x)} \tag{3c}$$

B. C.: $p_{bi}(x) = 0$ for $x = x_{\max}$ , constraint: $\int_0^{x_{\max}} p_{bi}(x)dx = 1$ (3d,e)

The $\delta_{ij}$ is Kronecker delta. We used correlations of Choi et al. (13) for the particle entrainment rate $K_i^*(x)$ and the model of Merrick and Highley(14) for attrition rates $R_{ai}$ and $R_i(x)$:

$$R_{ai} = K_a(u_i - u_{mfi})W_i \,, \quad R_i(x) = dx/dt = -K_a(u_i - u_{mfi})x/3 \tag{4a,b}$$

We assumed that fine particles formed by abrasion had diameter < 5 μm, a uniform size distribution, and negligible attrition. The solid flow rate in the absorber is defined as

$$F_{10} = \beta(u_1 - u_t)\rho_p \varepsilon_{s1} A_1 \,, \tag{5}$$

The $u_t$ is an average terminal velocity of particles, the $\rho_p$ particle density, the $\varepsilon_{s1}$ solid holdup and the $A_1$ bed area. The constant $\beta$ was determined as 0.211 from the experimental data (15).

Reaction time of particles of a size was considered as their mean residence time in each reactor

$$\tau_i(d_p) = \frac{W_{bi}\omega_{bi}(d_p)}{\sum_{j(out-flows)} S_{ij}\omega_{ij}(d_p)} \tag{6}$$

The average concentration of gas in each reactor was simply considered as an arithmetic mean value between inlet and outlet concentration.

$$\overline{C_{ij}} = \frac{C_{ij,0} + C_{ij,f}}{2} \tag{7}$$

Based on experimental data from TGA tests(1) and a continuous process test of Korea Institute of Energy and Research (KIER; $CO_2$ capture efficiency: 0.0831; absorber condition: static bed height 0.5 m, gas velocity 3.0 m/s, temperature 50°C), reaction rates for absorption and regeneration were determined:

$$\frac{dX}{dt} = \frac{0.337 \times 10^{-8}(1 - X)}{d_p} e^{-E_1/RT_1} \overline{C_{CO_2}}\,\overline{C_{H_2O}} \,, \quad E_1 = -42.3 \text{ kJ/g mol} \tag{8}$$

B. C.: $X = X_{1i}(d_p)$ at $t = 0$, $X = X_{1f}(d_p)$ at $t = \tau_1(d_p)$

$$\frac{d\lambda}{dt} = \frac{0.728 \times 10^{-2}(1 - \lambda)}{d_p} e^{-E_2/RT_2} \frac{1}{\overline{\overline{C_{CO_2}}}\,\overline{\overline{C_{H_2O}}}} \,, \quad E_2 = 50.4 \text{ kJ/g mol} \tag{9}$$

B. C.: $\lambda = 0 \;\; at \;\; t = 0$, $\lambda = \lambda_f(d_p) \;\; at \;\; t = \tau_2(d_p)$; $X = X_{2i}(d_p) \;\; at \;\; t = 0$,

$X = X_{2f}(d_p) = X_{1f}(d_p)(1 - \lambda_f(d_p)) \;\; at \;\; t = \tau_2(d_p)$

The following relations between moles of $CO_2$ and moles of $NaHCO_3$ (formed and disappeared) must be satisfied in each reactor:

$$\Delta N_{1,NaHCO_3} = -2\Delta n_{1,CO_2}, \;\; \Delta N_{2,NaHCO_3} = -2\Delta n_{2,CO_2} \qquad (10)$$

By combining Equations (2) to (10), the present model can calculate the particle flow rate, particle size distribution, concentration of $CO_2$ in gas phase and $NaHCO_3$ in particles. The overall capture efficiency of $CO_2$ can be determined from the mole balance on $CO_2$.

## CALCULATION CONDITIONS

The present model was applied to the KIER's process (absorber: 0.025 m i.d., 6 m height; regenerator: 0.1 m i.d., 1.2 m height). Table 1 summarizes size distribution of fresh sorbent. The apparent particle density was 808 kg/m$^3$. The following settings were held constant: pressure, 101.3 kPa; attrition coefficient of particle=3×10$^{-9}$ 1/mm; $F_1$=1, $R_{f2}$=0, $R_{f1}$=1 and $F_7$=$F_9$; temperature of the regenerator, 128$^o$C; gas velocity and static bed height of the regenerator, 1.14 m and 0.02 m/s, respectively; and fluidizing gas of the regenerator, pure $N_2$. Four absorber variables were tested in each range: static bed height in the absorber=0.3, **0.5**, 0.7 m; gas velocity in the absorber=**3.0**, 3.5, 4.0 m/s; temperature of the absorber=40, **50**, 60, 80$^o$C; composition of feed gas to the absorber= $N_2$: 78.6%, $CO_2$: 11.4%, $H_2O$: 5, **10**, 20, 30%. Other variables had the bold underlined values when a variable was tested.

Table 1 Size distribution of fresh sorbent particles

| Sieve size [μm] | -335+212 | -212+150 | -150+106 | -106+75 | -75+63 | -63+53 |
|---|---|---|---|---|---|---|
| Weight fraction | 0.0122 | 0.1188 | 0.7036 | 0.0853 | 0.0772 | 0.0031 |

## RESULTS AND DISCUSSIONS

Figure 2 shows the solid flux and mean particle diameter in the absorber, and feed rate of fresh sorbent particles which were calculated by the present model with variation of absorber gas velocity and static bed height. Particle size distributions in absorber and regenerator were nearly the same. As gas velocity or static bed height in the absorber increased, the solid flux in the absorber and feed rate of fresh sorbent increased, however, mean particle diameter decreased in the absorber. In order to maintain the same static bed height in the absorber, the solid flux should be increased because particle velocity increases as the gas velocity increases. In other words, the static bed height increases with an increase of solid flux in the absorber at a given gas velocity as can be seen in Equation (5). As the gas velocity increases, particle attrition in the absorber and loss of formed fine particles at the cyclone increase. This results in a decrease of mean particle diameter in the absorber and an increase of required feed rate of fresh sorbent. As the static bed height increases at a given gas velocity, particle attrition rate increases in the absorber as can be seen in Equation (4a) and loss of formed fine particles also increases at the cyclone. Thus, the mean particle diameter in the absorber decreases and the higher feed rate of fresh sorbent is needed.

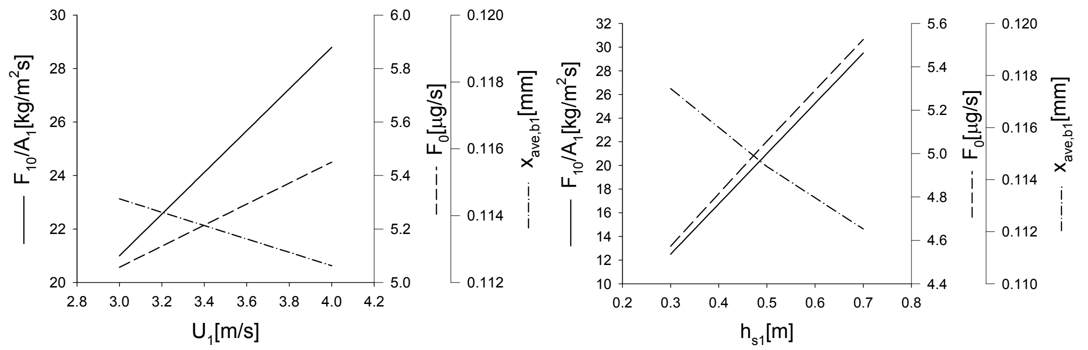


Figure 2. Effects of gas velocity and static bed height of absorber on solid flow rates.

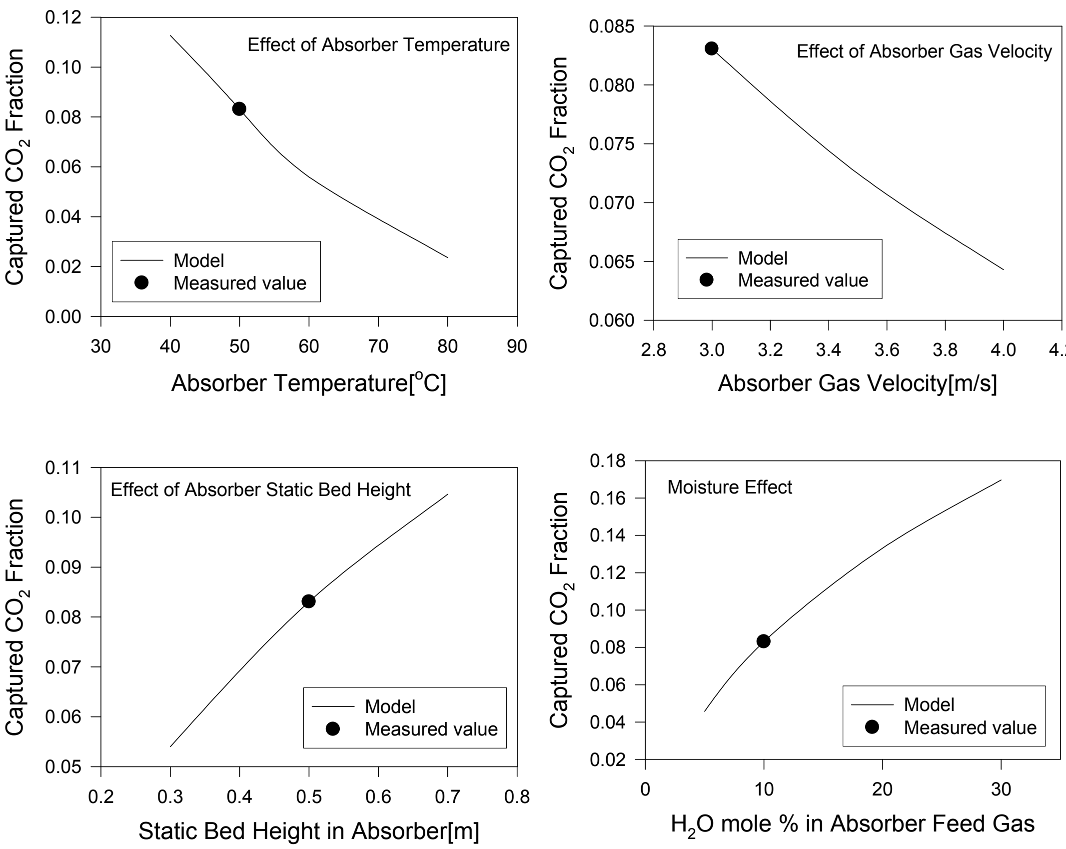


Figure 3. Effects of operating parameters on $CO_2$ capture efficiency.

Figure 3 show effects of operating parameters on $CO_2$ capture efficiency predicted by the present model. The $CO_2$ capture efficiency decreased as temperature in the absorber increased. Because the absorption reaction is endothermic, the equilibrium moves reverse direction as temperature increases. Therefore, the extent of $CO_2$ capture decreases with an increase of temperature. The $CO_2$ capture efficiency is proportional to the absorber temperature with an exponent -2.28. The $CO_2$ capture efficiency decreased as gas velocity in the absorber increased. Retention time of $CO_2$ in the absorber decreases with an increase of gas velocity. Therefore it caused the decrease of $CO_2$ capture. The $CO_2$ capture efficiency is proportional to the absorber gas velocity with an exponent -0.980. The $CO_2$ capture efficiency increased with an increase of static bed height in the absorber. It is because the absorption capacity increases with an increase of solid holdup in the absorber. The $CO_2$ capture efficiency is proportional to the absorber's static bed height with an exponent 0.786. The $CO_2$ capture efficiency increased as the moisture content of flue gas increased. It is because the absorption reaction rate increases with moisture concentration. Ratio of $N_2$ to $CO_2$ in the flue gas was maintained 0.786/0.114 constantly on calculations. The $CO_2$ capture efficiency is proportional to the moisture content of flue gas with an exponent 0.591.

## CONCLUSIONS

This study has developed a simplified model to investigate the system behavior and effects of important operating parameters for an entrained-bed absorption and bubbling-bed regeneration process which collects $CO_2$ from flue gas. Effects of four principal absorber parameters were tested in a laboratory scale process. The $CO_2$ capture efficiency decreased as temperature or gas velocity increased. However, it increased with an increase of solid holdup or moisture concentration. The $CO_2$ capture efficiency was exponentially proportional to each parameter. Based on the absolute value of exponent of the parameter, the temperature appeared to have the strongest effect on the $CO_2$ capture efficiency. The effect of gas velocity, static bed height, and moisture content was about a half, one third, and one fourth as strong as that of temperature, respectively.

## ACKNOWLEDGEMENT

The authors are grateful for the financial support of the CDRS R&D Center at Korea Institute of Energy Research and the Ministry of Education, Science and Technology of Korea Government.

## NOTATION

$A_1$: bed area of absorber [$m^2$]

$C_{ij,o}$ , $C_{ij,f}$ , $\overline{C_{ij}}$ : inlet, outlet, and average concentration of gaseous reactant j in reactor i [kg mol/$m^3$]

$d_p$ : particle diameter [m]

$E_i$: activation energy of reaction i [kJ/g mol]

$F_j$ : solid flow rate of stream $j$ [kg/s]

$h_{s1}$ : static bed height of absorber [m]

$K_a$ : particle attrition rate constant [1/m]

$K_i^*(x)$ : particle elutriation rate from bed $i$ [kg/s]

$\Delta N_{i,NaHCO_3}$ : total moles of $NaHCO_3$ in inflow of solids minus outflow of solids [kg mol/s]

$\Delta n_{i,CO_2}$ : total moles of $CO_2$ in inflow of gas minus outflow of gas [kg mol/s]

$p_{ai}(x)$ : probability density function of particles formed by attrition in bed $i$ [1/m]
$p_{bi}(x)$ : probability density function of particles in bed $i$ [1/m]
$p_j(x)$ : probability density function of particles in stream $j$ [1/m]
$p_0(x)$ : probability density function of fresh feed particles [1/m]
$R$ : gas constant, 8.314 [kPa $m^3$/kg mol K]
$R_{ai}$ : overall formation rate of fine particles by attrition in bed $i$ [kg/s]
$R_{fi}$ : recycle fraction of solid collected by cyclone [-]
$R_i(x)$ : particle attrition rate in bed $i$ [m/s]
$S_{ij}$ : mass flow rate of total particle in outflow stream $j$ from bed $i$ [kg/s]
$t$ : time [s]
$T$ : temperature [K]
$u_i$ , $u_{mfi}$ : fluidizing and minimum fluidizing velocity in reactor $i$ , respectively [m/s]
$u_t$: average terminal velocity of particles
$W_i$ : weight of bed $i$ [kg]
$x$ , $x_c$ , $x_{max}$ : spherical particle diameter, cut diameter of cyclone and maximum $x$ [m]
$X_{ii}$ , $X_{if}$ : conversion of $Na_2CO_3$ to $NaHCO_3$ for in- and out- flow of reactor $i$ [-]

## Greeks

$\alpha_{ik}$ , $\alpha_{i3}$ : functions defined as Eqs. 2(a, b, c) [1/m, 1/$m^2$]
$\beta$ : coefficient [-]
$\delta_{ij}$: Kronecker delta [-]
$\varepsilon_{s1}$: solid holdup [-]
$\lambda$ : conversion of regeneration reaction [-]
$\rho_p$: particle density [kg/$m^3$]
$\tau_i$ : mean particle residence time in reactor $i$ [s]
$\psi_i(x)$ : cyclone collection efficiency [-]
$\varpi_{bi}(x)$ : mass fraction of particle in bed $i$ [-]
$\varpi_{ij}(x)$ : mass fraction of particle in outflow stream j from bed $i$ [-]

## Subscripts

$ave$ , $b$ : average and bed, respectively
$i$ : free index, 1 for absorber and 2 for regenerator
$j, k$ : indices

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Key Words: Model, Fluidized Bed, Entrained Bed, Sodium Carbonate, Carbon Dioxide, $CO_2$, Removal, Capture, Absorption, Regeneration