Modelling and Design of Carbon Dioxide Absorption in Rotating Packed Bed and Packed Column

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Abstract: This study compares the carbon dioxide capture efficiency of packed bed (PB) column and rotating packed bed (RPB) absorber by mono-ethanolamine (MEA) solution. Carbon dioxide absorption experiments were carried out in laboratory scale packed bed column and rotating packed bed at various lean carbon dioxide loadings of the MEA solution. They were modelled successfully by an ASPEN Plus advanced customer model (ACM). Using these models, we found that substantial savings of packing volume can be achieved using a series arrangement of RPB and PB for the CO2 capture process using 30wt% MEA solution.

Keywords: rotating packed bed, packed bed, carbon dioxide capture, mono-ethanol amine

1. INTRODUCTION

Global warming caused by greenhouse gas emissions has become a central issue of the 21st century. Carbon dioxide capture and storage (CCS) technologies need to be developed to reduce carbon dioxide concentration in Earth’s atmosphere. Chemical absorption processes are currently the preferred technology for post-combustion carbon dioxide capture (PCCC) (IPCC, 2005). However, the huge amount of exhausted gas of fossil fuel power plants entering the PCCC process for CO2 absorption is a drawback for conventional absorption apparatus such as packed bed columns due to significant mass-transfer limitations at the gas-liquid interfacial area. To improve the mass transfer between the gas and the liquid, the rotating packed bed (RPB) was proposed by Ramshaw and Mallinson (1981). In a rotating packed bed, the downward liquid flow and upward gas flow due to gravity and density difference in a packed bed is outward liquid flow and inward gas flow due to centrifugal force. The high rotating speed disperses the liquid into small droplets. Consequently the gas-liquid interfacial area is increased and therefore the mass transfer rate.

Many studies have been carried out in recent years investigating the feasibility of CO2 capture by a RPB using various amine solutions (Lin et al., 2003); (Jassim et al., 2007); (Cheng and Tan, 2009); (Yu et al., 2012). However, most of these studies employed fresh amine solutions. In an actual absorption process, the lean solvent input is not free of CO2. For a 30 wt% MEA process, the lean and rich loading should be in the range of 0.35 to 0.49 (Abu-Zahra et al., 2007a; Abu-Zahra et al., 2007b) to minimize energy expenditure during regeneration.

The purpose of this study was to obtain experimental data using 30wt% MEA solutions at various loadings. These data were then used to validate the model propose by Kang et al. (2014) implemented in the Aspen Custom Modeller. The validated model was then used to design absorber arrangement that is most suitable for the absorption and stripping process.

2. EXPERIMENT

The dimensions of the RPB and PB absorbers used in our experiments are presented in Table 1.

| Table 1: Dimensions and operating conditions of the packed bed column and the rotating packed bed |
|---------------------------------|-----------------|-----------------|
| Apparatus                      | Packed column   | RPB             |
| Diameter (m)                   | 0.025           | 0.125 (OD);     |
|                                 |                 | 0.025 (ID)      |
| Packing height (m)             | 0.6             | 0.023           |
| Packing volume (m³)            | 294.5           | 271             |
| Packing type                   | Plastic Raschig rings (6mm) | Stainless wire mesh |
| Porosity                       | 0.62            | 0.96            |
| Specific surface area (m²/m³)  | 710             | 887.6           |
| Rotator speed (rpm)            | --              | 1600            |
| Gas flow (L/min)               | 5               | 5 – 60          |
| Gas CO2 fraction (mol %)       | 10              | 10              |
| MEA concentration (wt %)       | 30              | 30              |
| Liquid MEA fraction (mol %)    | 11.2            | 11.2            |
| Liquid flow (L/min)            | 0.1             | 0.1             |
| CO₂ lean loading (molCO₂/molMEA) | 0.365 – 0.444 | 0.0; 0.157; 0.249; 0.380; 0.442 |
| Inlet temperature (°C)         | 25 (Gas); 50 (Liquid) | (Gas and Liquid) |

The performance of the RPB at different lean loadings was studied experimentally. Five different CO2 lean loadings (0, 0.157, 0.249, 0.380, 0.442 molCO₂/molMEA at the liquid inlet) of the MEA solution were use. The experimental data of CO2 concentrations of the outlet gases obtained at different gas flow rates were shown in Table 2.
Table 2: Experimental CO2 Removal by Rotating Packed Bed at Various Lean Loadings and Gas Flow Rates

| Lean Loading [mol CO2/mol MEA] | 0.0 | 0.157 | 0.249 | 0.380 | 0.442 |
|---------------------------------|-----|-------|-------|-------|-------|
| Gas Flow rate (L/min) | Outlet CO2 mole fractions | 0.008 | 0.017 | 0.045 | 0.019 | 0.033 | 0.060 | 0.081 | 0.032 | 0.063 | 0.078 | 0.055 | 0.070 | 0.083 | 0.049 | 0.059 | 0.075 |
| 5 | - | 0.017 | 0.045 | - | 0.084 | - | - | - | - | - | - | - | - | - | - | - |
| 10 | - | 0.017 | 0.038 | 0.068 | - | - | - | - | - | - | - | - | - | - | - | - | - |
| 20 | 0.013 | 0.030 | 0.071 | 0.088 | - | - | - | - | - | - | - | - | - | - | - | - | - |
| 30 | 0.022 | 0.048 | 0.081 | - | - | - | - | - | - | - | - | - | - | - | - | - | - |
| 40 | 0.032 | 0.063 | 0.078 | - | - | - | - | - | - | - | - | - | - | - | - | - | - |
| 50 | 0.041 | 0.070 | 0.083 | - | - | - | - | - | - | - | - | - | - | - | - | - | - |
| 60 | 0.049 | 0.075 | 0.084 | - | - | - | - | - | - | - | - | - | - | - | - | - | - |

The experimental data of CO2 concentrations of the outlet gases obtained for the packed bed column at different lean loadings using a gas flow rate of 5 L/min and a liquid flow rate of 0.1 L/min were shown in Table 3.

Table 3: Experimental CO2 Removal by Packed Bed Column at Various Lean Loadings

| Lean Loading [mol CO2/mol MEA] | 0.365 | 0.370 | 0.386 | 0.408 | 0.438 | 0.444 |
|---------------------------------|-------|-------|-------|-------|-------|-------|
| Outlet CO2 mole fractions | 0.040 | 0.038 | 0.046 | 0.056 | 0.058 | 0.067 |

3. MODEL AND VALIDATIONS

The simulation models used for the RPB absorbers were reported by (Kang et al., 2014). The model of rotating packed bed was implemented in the simulation software ASPEN Custom Modeler (ACM). The absorption process in both apparatus is described by the simple two-film theory.

Table 4: Material and Energy Balance of the Two-film Model for RPB

| Material balances for gas phase | \( \frac{dC_{v,i}}{dt} = \frac{\partial(C_{v,i},v_{i})}{\partial r} \cdot \frac{1}{2\pi Z \cdot r_{s} - r_{i}} - \varepsilon d_{g} \cdot \frac{N_{i}}{C_{p}} \) |
| Material balance for liquid phase | \( \frac{dC_{l,i}}{dt} = -\frac{\partial(C_{l,i},v_{l})}{\partial r} \cdot \frac{1}{2\pi Z \cdot r_{s} - r_{i}} + \varepsilon d_{g} \cdot \frac{N_{i}}{C_{p}} + e_{r} \cdot n_{i} \) |
| Energy balance for gas phase | \( \frac{\partial E_{v,i}}{\partial r} \cdot C_{v,i} = \frac{\partial(C_{v,i},v_{i})}{\partial r} \cdot \frac{1}{2\pi Z \cdot r_{s} - r_{i}} + \varepsilon d_{g} \cdot \frac{h_{i}(T_{i} - T_{s})}{C_{p}} \) |
| Energy balance for liquid phase | \( \frac{\partial E_{l,i}}{\partial r} \cdot C_{l,i} = \frac{\partial(C_{l,i},v_{l})}{\partial r} \cdot \frac{1}{2\pi Z \cdot r_{s} - r_{i}} - \varepsilon d_{g} \cdot \frac{h_{i}(T_{i} - T_{s})}{C_{p}} - \Delta H_{m} N_{w} - \Delta H_{m} N_{g} \) |

The list of liquid phase reactions are given in Table 5. The equilibrium constants of these reactions were given by (Austgen, Rochelle et al. 1989).

Table 5: Liquid reactions considered

| Name | Reaction |
|------|----------|
| Protonation of MEA | MEAH⁺ + H₂O ↔ MEA + H₂O⁻ |
| Water dissociation | 2H₂O ↔ H⁺ + OH⁻ |
| Carbonate formation | HCO₃⁻ + H₂O ↔ CO₃²⁻ + H₃O⁺ |
| Carbamate formation | CO₂ + MEA + H₂O ↔ MEACOO⁺ + H₂O⁻ |
| Bicarbonate formation | CO₂ + OH⁻ ↔ HCO₃⁻ |

The mass transfer calculations include the correlation from Onda et al. (1968) correlation for the gas mass-transfer coefficient, Tung and Mah (1985) correlation for the liquid mass-transfer coefficient, the Puranik and Vogelpohl correlation (1974) for the gas-liquid interfacial area and Burns et al. (2000) correlation for the liquid holdup. The heat transfer coefficients are calculated using the Chilton and Colburn Method (Chilton and Colburn, 1935).

Table 6: Mass Transfer Correlations Used in the RPB Model

| Mass transfer (Onda, Takeuchi et al. 1968, Tung and Mah 1985) | \( k_{g,i} = 2(\frac{Re_{g}}{Sc_{g,i}})^{0.7} \left( \frac{D_{g}}{R_{g}} \right)^{0.7} \) |
| Gas-liquid interfacial area (Puranik and Vogelpohl 1974) | \( a_{gl} = 1.05 \frac{a_{p}}{Re_{g}} \cdot (We_{g})^{0.135} \cdot \frac{a_{l}}{a_{p}}^{-0.206} \) |
| Gas-liquid hold up (Burns, Jamil et al. 2000) | \( e_{l} = 0.039 \cdot \frac{a_{p}}{a_{l}}^{-0.5} \cdot \frac{u_{l}}{u_{p}}^{0.6} \cdot \frac{\nu_{p}}{\nu_{l}}^{0.22} \) |
| Interfacial heat transfer coefficient (Chilton and Colburn 1935) | \( h_{gl} = k_{g}RT_{g} \left( \frac{C_{p}p_{g}}{D_{g,avg}} \right)^{1/3} \left( \frac{\lambda_{g}}{D_{g,avg}} \right)^{2/3} \) |

The enhancement factors due to film reaction is calculated using apparent kinetic constant provided by Aboudheir et al. (2003).

\[ E_{CO2} = \sqrt{k_{app}DCO2} \]
\[ k_{app} = k_{MEA}(C_{MEA})^{2} + k_{H2O}C_{MEA}C_{H2O} \]
\[ k_{MEA} = 4.61 \times 10^{9} \cdot \frac{m^{6}}{k_{mol}^{2}} \cdot \exp(-4412K) \]
\[ k_{H2O} = 4.55 \times 10^{9} \cdot \frac{m^{6}}{k_{mol}^{2}} \cdot \exp(-3287K) \]
4. VALIDATIONS

Figure 1 illustrates the predicted and experimental CO₂ removal, defined as \( \Delta \text{CO}_2 = 1 - \frac{y_{\text{CO}_2,\text{out}}}{y_{\text{CO}_2,\text{in}}} \). There are good agreements between experimental and modelling results under all investigated process conditions. Hence we can conclude that the developed ACM model of the rotating packed bed implemented in is able to predict the carbon dioxide capture performance of a rotating packed bed at various CO₂ loadings of the MEA solution.

5. PROCESS DESIGN

Despite the favourable comparison presented in the above sections, the comparison may not be very relevant in actual process applications. First of all, we note that structural packings are usually used for large scale absorbers. The above experiments are performed using random packings. Secondly the PB experiments were performed are very close to flooding while the RPB experiments performed are quite far away from flooding. Most importantly, the key parameter for an efficient process is the required energy in the stripping process for the regeneration of the solvent. In the absorption-stripping process an optimal flow rate for a given removal can be identified. If solvent rate is too small, there is not enough capacity unless the near rich solution is regenerated to a very low lean loading of CO₂. If the solvent rate is too high, a lot of sensible heat is required to overcome the temperature difference between the absorber and the stripper. (Abu-Zahra et al., 2007a,b) identified the best process condition for CO₂ absorption using 30 wt.% MEA solution is to use a lean loading of 0.32 mol-CO₂/mol-MEA and a liquid rate that achieved a rich loading of 0.49 mol-CO₂/mol-MEA after the absorption process. None of the experiments above cover the full range. However, given the two relatively reliable models for PB and RPB, we are able to compare the design results of the two types of absorber.

Table 7 listed the flue gas rate and inlet conditions which has been used in this study to design an absorber. The design objective of the absorption apparatus is to achieve four different CO₂ removal rates (80%; 90%, 95%, 99%).

| Mass flow [kg/hr] | 240 |
|-------------------|-----|
| Pressure [atm]    | 1.24|
| Temperature [°C]  | 50  |
| Composition [vol. %] | |
| N2                | 73.1|
| CO2               | 14.8|
| H2O               | 6.8 |
| O2                | 5.3 |
For the packed bed column flooding limits are easily calculated in Aspen Plus packing sizing module. However, the flooding limit has to be determined experimentally. To determine the flooding point of the RPB, the liquid flow rate and rotating speed were fixed at first, and then the gas flow rate was increased gradually until there was a significant amount of water trapped out from the gas outlet pipeline. The detail procedure for visual determination of the flooding point could be found in Jassim (2002). The results are obtained using rotating speeds of 700, 1000, 1600 rpm were plotted using the Sherwood correlation (Figure 3). Figure 3: Sherwood correlation for flooding limits of the RPB absorber. To design absorbers, a flooding factor of 60% was chosen so that the volumes of the packing in both the PB and RPB can be determined. Given the flooding data, the height of the RPB can be chosen so that the velocity of gas flow is approximately 60% of the flooding limit shown in Figure 3. Given the height of the RPB, and an inner diameter approximately equals to the height, the outer diameter can be determined by our model simulation to achieve 90% removal.

Figure 4 compares the required packing volumes in a packed bed column and a RPB applying different rotor speed to achieve different carbon dioxide removal rates given that a 30 wt.% MEA solution with a lean loading of 0.32 mol-CO2/mol-MEA and a liquid rate that achieved a rich loading of 0.493 mol-CO2/mol-MEA after the absorption process were used. Surprisingly, the comparison showed that an RPB with a low rotor speed of 700 rpm requires the smallest packing volume. More importantly, the comparison between PB and RPB points out that only at a removal rate of 95% or more, substantial savings in packing volume can be achieved in using the RPB.

Figure 5 compared increase in packing volume required in a RPB with rotor speed of 700 rpm to the increase in packing volume required in a PB column to achieve reach different CO2 loadings of the rich MEA solution at the outlet. It was found that the RPB absorbs carbon dioxide very efficiently until the MEA solution reaches a value of 0.45 mol-CO2/mol-MEA. However a lot of packing volume is required to achieve a rich loading beyond 0.48. For further CO2 absorption more packing volume is required which makes the process very inefficient. On the other hand, the increase in packing volume approaches a linear trend as the solution become saturated in CO2 in a PB absorber. There are two factors that results in such a phenomenon. Firstly, in a RPB, the volume increases as the square of the outer radius, but the packed bed column increases linearly as the height of the absorber. Therefore the RPB is less effective in volume intensification as the gas-liquid contact length increases. Furthermore, the RPB reduces mass transfer resistance. At low CO2 loading, the advantages of fast chemical reaction will be manifested when mass transfer resistance is removed. However, at high CO2 loading the reaction of CO2 to form bicarbonate and carbamate is almost near equilibrium will slow reaction kinetics. Rapid physical mass transfer into solution results in build-up of unreacted CO2 concentration. The RPB is even less efficient than a PB. This rapid increase in volume of packing required is also observed at rotating speed of 1000 rpm and 1600 rpm but at slightly lower saturations. This lead to the increase in volume required when rotating speed is increased.
In summary, the RPB shows a very efficient CO2 absorption at moderate CO2 loadings of the MEA solution but a poor performance at high loadings whereas the packed bed column is worse than the RPB at moderate loadings but more efficient in terms of reaching the desired rich loading. Therefore, the efficiency of the absorption process can be improved by combining the RPB and the packed column in a series process as shown in Figure 6. In this arrangement the lean solvent enter the RPB first, and then the PB, and the saturated rich solution comes out at the bottom of the PB. The flue gas enters to the bottom of the PB column first and then the RPB absorber. The CO2 ridded flue gas come out at the inner exit of the RPB. The point of transition from RPB to PB is optimized with respect to the total volume of packing used.

Figure 6: Flowsheet of a RPB and packed bed column in series.

Figure 7: Comparison of volume required using a PB, RPB and PB+RPB in series, a rotating speed of 700 rpm was used in RPB

The advantages of the series arrangement can be illustrated using the design calculations shown in Figure 7. On the left are the packing volumes of a single PB required to achieve the specified removal using a leaning loading of 0.32 and a flow rate that achieved a rich loading of 0.49. In the middle column are corresponding the packing volumes of a single RPB required to achieve the specified removal. On the right are the optimized packing volumes of the RPB and PB in the series arrangement required to achieve the specified removal. We can see that the total volumes of packing and the packing volumes of both RPB and PB were substantially reduced to 1/2 to 1/3 the size of the original PB.

Figure 8: Power savings of the RPB+PB arrangement

The advantages of the series arrangement can be illustrated when we examine the additional power to rotate the the RPB. The required power is derived from three main sources (Keyvani and Gardner, 1988): (1) power required to accelerate the liquid in the rotor; (2) power require to overcome the frictional windage drag of the rotor; and (3) power consumed by the bearing friction. In this work the power required for the bearing friction is neglected. It is proportional to the rotation speed and has to be determined experimentally. The power required to accelerate the liquid is the product of the moment of inertia and angular velocity. Hence it increases with the square of the outer radius. The power require to overcome the frictional windage drag of the rotor is proportional to square of the angular velocity and fifth power of the outer radius. In this study, we showed that the model by (Kang et al., 2014) of the carbon dioxide absorption process by mono-ethanolamine solution in a RPB absorber is able to predict the CO2 capture performance at various lean loadings. In addition, simulations of PB column using Aspen Plus rate-based model also agree well with experimental results at various lean loadings. In these experiments, the RPB shows a more efficient CO2 capture performance compared to PB column since the required power is only one tenth of the power requirement when a single RPB was used.

6. CONCLUSIONS

In this study, we showed that the model by (Kang et al., 2014) of the carbon dioxide absorption process by mono-ethanolamine solution in a RPB absorber is able to predict the CO2 capture performance at various lean loadings. In addition, simulations of PB column using Aspen Plus rate-based model also agree well with experimental results at various lean loadings. In these experiments, the RPB shows a more efficient CO2 capture performance compared to PB column since the

In order to integrate the RPB technology into the CO2 capture process, it is necessary that the entering lean solution and rich solution coming out of the absorber have the lean CO2 loading and rich CO2 loading that are most favourable for stripping. For 30 wt% MEA solutions, this range is
approximately 0.32 to 0.49 mol-CO2/mol-MEA. Moreover to prevent flooding and use of large PB diameter or RPB height, a flooding factor of 60% is used. Using these criteria, the sizes of PB and RPB absorbers were designed for a given flue gas feed. It was found that the savings in packing volume by using a RPB is limited. Further detailed analysis showed that the advantages of RPB is only manifested when the MEA solution has loadings less 0.48. As the loadings of the MEA solution approaches saturation of 0.5, a lot of additional packing volume is required in the RPB.

A series arrangement consisting of a RPB and a PB is proposed. By connecting the RPB and the PB column in series so that the lean MEA solution enters the RPB first and then the packed bed column and the flue gas first the packed column and then the RPB, substantially less packing volumes are required. The analysis of the energy requirement for operating the RPB confirms that a combination of RPB and packed bed column is preferential. Since the volume and therefore the outer radius of the RPB in combination with a PB column can be reduced, the required energy is decreased significantly.

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