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Integrated mathematical modeling for prediction of rich CO_2 absorption in structured packed column at elevated pressure conditions

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ABSTRACT

Modeling and simulation on lean carbon dioxide (CO_2) absorption has been widely studied for low pressure conditions. However, modeling on rich CO_2 absorption in packed column requires consideration on significant decrease of total gas flow rate along the column. In this work, the absorption of CO_2 in countercurrent structured packed column at 50% concentration level in inlet gas was modeled to predict the mass transfer of CO_2 from gas phase into liquid phase. The integrated mathematical model, which is a combination of hydraulic and mass transfer correlation from SRP II model as well as mass conservation equation, was developed to elucidate the CO_2 capture profile along the absorption column for pressure conditions of 0.1, 1, 3 and 5 MPa. Based on the validation with experimental data, the mathematical model developed in this study had satisfactorily represented the mass transfer behavior of high CO_2 concentration gas removal along the absorption column at elevated pressure conditions.

1. Introduction

Natural gas is known to be the cleanest and most hydrogen-rich of all the hydrocarbon energy sources with high energy conversion efficiencies for power generation (Economides and Wood, 2009). However, the existing lean carbon dioxide (CO_2) natural gas reserves are depleting fast. In order to support the continuous increasing demand for energy worldwide (IEA, 2008), there is an urgent need to explore the vastly undeveloped natural gas reserves with rich CO_2 content up to 80% (Tan et al., 2012).

Removal of CO_2 via absorption process for natural gas purification has been in the industry for decades (Kohl and Nielsen, 1997). Research on CO_2 capture from flue gas is also being conducted rigorously worldwide in effort to reduce the catastrophic global warming effect (Tan et al., 2012). Nevertheless, the existing system/research is mainly focused on low CO_2 concentration up to 20% (Yang et al., 2008). Hence, in order to move forward with the implementation of purification process for rich CO_2 content natural gas, it is of utmost importance to have an understanding of the mass transfer performance under continuous flow process at high CO_2 content and elevated pressure condition.

The absorption performance of CO_2 at high concentrations (50%) in CO_2-natural gas mixtures has been investigated before (Tan et al., 2015). The experiment was conducted from 0.1 MPa to 5 MPa for 20% (by weight) monoethanolamine (MEA) aqueous solution in a countercurrent packed column. The elevated pressure in this study refers to conditions above ambient pressure. However, pressure condition of 0.1 MPa was still included in this research work as reference point. The results indicated that there was a higher CO_2 removal efficiency at elevated pressures.

Model for rich CO_2 content absorption at high pressure conditions are not readily available in open literature. Previous modeling and simulation studies by other researchers were mostly conducted for lean CO_2 absorption process and at low pressure conditions (Aroonwilas et al., 2003; deMontigny et al., 2006; Gabrielsen et al., 2007; Sema et al., 2012; Khan et al., 2011; Liu et al., 2006; Moioli and Pellegrini, 2015; Aroonwilas et al., 2001; Gabrielsen et al., 2006; Moioli et al., 2013; Naami et al., 2012). Some of the models applied inert gas flow rate which was constant along the column or constant gas flow rate assumption and the reduction of total gas velocity along the column was not taken into consideration. This assumption was acceptable in those studies since the inlet gas flow

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contained low concentration of CO_2 (up to 20%). Recently, Rahimpour et al. (Rahimpour et al., 2013) investigated natural gas sweetening process at approximately 6.7 MPa pressure condition using computational fluid dynamics (CFD) model but the inlet gas consisted of lean CO_2 content at approximately 4% level only. Thus, there is a knowledge gap on the behavior of rich CO_2 absorption process at elevated pressure which needs to be further explored. Bridging of this knowledge gap is very crucial, especially since low CO_2 content natural gas reserves are depleting and there is a possible need to start exploring high CO_2 content natural gas in order to continue fulfilling the energy demand worldwide. Thus, in this study, an integrated mathematical model for capturing high concentrations of CO_2 capture along the absorption column at elevated pressure conditions is developed.

In our absorption process, the inlet gas contained rich CO_2 concentration at 50% level and the pressure condition investigated was up to 5 MPa. When the solute absorbed is present at moderate or high concentrations in the gas, the decrease of total gas flow rate along the column would be significant, hence, must be included in the model (McCabe et al., 2005; Abdul Halim et al., 2015). Hence, in this work, a combination of hydraulic, mass transfer and mass conservation equation model was applied to elucidate the transport of the conserved mass with the significant reduction of total gas flow rate along the column from high concentration of CO_2 at inlet point to low concentration of CO_2 at gas outlet. The predicted trend of absorption performance using the developed model would then be compared with the actual experimental results from our previous study (Tan et al., 2015) for model validation.

2. Methodology

2.1. Mathematical modeling

The high concentration CO_2 absorption process for the countercurrent packed column was modeled based on the mass conservation of liquid and gas phases. The CO_2 concentration in inlet gas stream at the bottom of the column was initially high. The total gas flow rate along the column reduces as the gas stream travels from bottom to the top of the column and CO_2 in gas phase transfers across a gas–liquid interface into the liquid phase.

The mass transfer and hydraulic model applied in this work to predict the CO_2 concentrations along the absorption column was based on SRP II model which was developed by Rocha et al. (Rocha et al., 1993, 1996). According to Tsai et al. (Tsai et al., 2011), SRP II model is one of the widely used models for structured packing. A slight variation of the model was also developed by adapting an approach by Xu et al. (Xu et al., 2000). In our study, the gas and liquid phase mass transfer coefficients, k_G and k_L, as well as the liquid holdup in packed bed, h_L, from the original SRP II model were maintained. However, the effective packing area, a_e, would be changed with correlation developed by Tsai et al. (Tsai et al., 2011) and Gualito et al. (Gualito et al., 1997) in search of a model which could best fit the experimental results for MEA 20wt% aqueous solution from our previous work (Tan et al., 2015). The a_e correlation developed by Tsai et al. (Tsai et al., 2011) was selected to be incorporated in our model because Flexipac 1Y, the packing which was used in our experimental study, was one of the packings included in their mass transfer area database. Meanwhile, Gualito et al. (Gualito et al., 1997) corrected the original SRP II's a_e correlation for high pressure application up to 2.76 MPa. Hence, the a_e correlation developed by Gualito et al. (Gualito et al., 1997) was also selected to be incorporated into our model in order to test its suitability for application beyond 2.76 MPa.

Hence, a model which considers the significant reduction of the total gas flow rate was developed to predict the performance of the high CO_2 concentration absorption in the study. The model also elucidates the impact of elevated operating pressure (up to 5 MPa) to the absorption performance. The correlation of axial velocity with the CO_2 absorption rate can be obtained using mass conservation equation. The total mole conservation of the CO_2 gas phase in steady state can be expressed in Equation (1):

$$x_g d(Cu_g)/dz = x_g Cd(u_g)/dz = -N_{COa} \tag{1}$$

where, C is the total mole concentration of gas phase (kmol/m^3), N_{CO2} is the CO_2 absorption rate (kmol/m^3 s), x_g is the gas volume fraction in column and u_g is the gas phase velocity along the column height (m/s). For CO_2 rich gas stream, the correlation of gas flow rate with the CO_2 mole transfer along the column is expressed as in Equation (2):

$$x_g d(C_{co2})/dz = -N_{CO2}/u_g(1 - y_{CO2}) \tag{2}$$

where, y_{CO2} is the CO_2 mole fraction in gas phase and C_{CO2} is the CO_2 mole concentration in gas phase (kmol/m^3). Equation (2) includes the factor of $(1-y_{CO2})$ as change in molar flow rate must be taken into account.

N_{CO2} in one unit space meter cube is functioned by the overall mass transfer coefficients and partial pressure of CO_2, as expressed in Equation (3):

$$N_{CO2} = K_G a_e \left(p_{CO2} - p_{e,CO2}\right) \tag{3}$$

where, K_G is the overall mass transfer coefficient (kmol/m^2.Pa.s), a_e is the effective packing area (m^2/m^3), p_{CO2} is the CO_2 partial pressure (Pa) and $p_{e,CO2}$ is the CO_2 partial pressure in equilibrium condition (Pa). The correlation for K_G which also considers MEA enhancement is expressed in Equation (4):

$$1/K_G = (1 - y_{CO2})/k_g + H_{CO2}/k_l E \tag{4}$$

where, k_g is the gas-side mass transfer coefficient (kgmol/m^2.Pa.s), k_l is the liquid-side mass transfer coefficient (kgmol/m^2.Pa.s), E is the enhancement factor, H_{CO2} is Henry's constant (m^3 Pa/kmol). $1/(1-y_{CO2})$ was introduced in the gas mass transfer coefficient by assuming that methane (CH_4) is inert and not absorbed by the solvent. The correlations for k_g, k_l, and h_L this work were calculated using the correlation function developed by Rocha et al. (Rocha et al., 1993, 1996) as expressed in Equations (5)–(11):

$$k_g = 0.054(D_G/S)((U_{Ge} + U_{Le})\rho_G S/\mu_G)^{0.8}(\mu_G/D_G\rho_G)^{0.33} \tag{5}$$

where

$$U_{Ge} = U_{Gs}/\in(1 - h_L)\sin\theta \tag{6}$$

$$U_{Le} = U_{Ls}/\in h_L \sin\theta \tag{7}$$

$$k_l = 2\sqrt{D_L U_{Le}/\pi S} \tag{8}$$

$$h_L = (4F_t/S)^{2/3}\left(3\mu_L U_{Ls}\Big/\rho_L(\sin\theta)\varepsilon g_{eff}\right)^{1/3} \tag{9}$$

where

$$F_t = a_e/a_p \tag{10}$$

$$g_{eff} = g((\rho_L - \rho_G)/\rho_L)\left(1 - (\Delta P/\Delta z)\Big/(\Delta P/\Delta z)_{flood}\right) \tag{11}$$