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Study of mass transfer correlations for intensified absorbers in post-combustion CO_2 capture based on chemical absorption

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Abstract

Process intensification (PI) technologies such as rotating packed beds (RPBs) could reduce the size of absorber used in post-combustion CO_2 capture (PCC) based on chemical absorption processes by about 12 times compared to absorber with standard packed beds. However, mass transfer correlations for predicting effective interfacial area and liquid film mass transfer coefficient in RPBs are limited in literature and their prediction accuracy against experimental data is yet to be compared. This need is addressed in this study by evaluating the performances of different correlations through comparison with experimental data. Of all the correlations assessed, it is found that Lou *et al*. [1] and Tung and Mah [2] correlations give reliable estimate of the effective interfacial area and liquid film mass transfer coefficients respectively.

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Keywords: Post-combustion CO_2 capture; chemical absorption; packed bed; rotating packed bed; mass transfer correlations

1. Introduction

1.1. Background

The unfavourable role of CO_2 in stimulating climate change has generated concerns as CO_2 level in the atmosphere

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continues to increase. These concerns have paved way for carbon capture and storage (CCS) from large stationary sources such as coal-fired power plants. With CCS, electricity will continue to be generated from secure and cheap energy sources such as coal and natural gas with minimized impact on the environment. Post-combustion CO_2 capture (PCC) based on chemical absorption is a near-term option for implementing CCS commercially. However, absorbers/strippers with packed bed used in PCC processes are huge in size contributing significantly to plant footprint, capital and operating costs. For example, engineering estimates showed that absorbers for a PCC plant using MEA solvent for capturing CO_2 from a 500 MWe coal-fired subcritical power plant will have diameters up to 25 m and packing height over 27 m. Through PI, the sizes of the absorber/stripper could be reduced significantly [3-4]. Agarwal *et al*. [3] and Joel *et al*. [4] reported 7 and 12 times absorber column size reduction respectively for separate cases involving replacement of packed bed with RPB for PCC applications. RPB have been successfully demonstrated in industry for different applications such as natural gas desulphurization.

Nomenclature

a	Effective interfacial area of packing per unit volume (m^2/m^3)
a_t	Actual area of packing per unit volume (m^2/m^3)
A	Tangential section area (m^2) = $2\pi rZ$
c, d	Packing parameters for Luo et al. (2012) correlation
d_h	Hydraulic diameter (m) = $4\epsilon/a_t$
d_p	Effective diameter of packing (m) = $6(1- \epsilon) /a_t$
D_L	Liquid diffusivity (m^2/s)
k_L	Liquid film mass transfer coefficient (m/s)
L_m^*	Liquid mass flowrate per unit tangential section area (kg/m^2 s)
Q_L	Liquid volumetric flowrate (m^3/s)
r	Radius (m)
r_i	Inner radius of the packed bed (m)
r_o	Outer radius of the packed bed (m)
r_s	Radius of the stationary housing (m)
u_L	Liquid velocity (m/s)
V_i	Volume inside the inner radius of the bed (m^3) = $\pi r_i^2 Z$
V_O	Volume between the outer radius of the bed and the stationary housing (m^3) =$\pi(r_s^2 - r_o^2)Z$
V_t	Total volume of the RPB (m^3) = $\pi r_s^2 Z$
V	Parameter for Chen *at al*. [5] model = $1 - 0.93\frac{V_o}{V_t} - 1.13\frac{V_i}{V_t}$
Z	Height of the rotor (m)
σ_c	Critical surface tension for metallic packing material (N/m)
σ_L	Liquid surface tension (N/m)
ϵ	Packing porosity (m^3/m^3)
ρ_L	Liquid density (kg/m^3)
μ_L	Liquid dynamic viscosity (Pa s)
ω	Rotating speed (rad/s)

1.2. Principle of RPB absorber and problem statement

RPB absorber comprises of an annular packed bed (rotor) mounted on a rotating shaft. The gas and liquid enters the rotor through the outer and inner sections respectively so that they flow counter-currently across the bed (Fig.1). The liquid and gas are subject to intense centrifugal acceleration which is many times the gravitational acceleration in conventional packed beds [6]. As a result,

- RPB allows high flooding rate leading to drastic reduction in packing volume
- RPB permits viscous solvents such as concentrated solutions of monoethanolamine (MEA). Concentrated solutions will result in more rapid kinetics and therefore higher CO_2 absorption rate.

Consequently, similar capture levels as in conventional packed beds can be achieved in RPBs using significantly reduced packing volume [3-4]. Due to the presence of centrifugal force field in RPBs, mass transfer correlations for conventional packed columns cannot be used to predict mass transfer in RPBs with acceptable accuracy [4,7]. For RPBs, only a few correlations have been reported for effective interfacial area and liquid film mass transfer coefficients [1-2, 5,8]. Modification of the correlations for conventional packed beds by replacing the "g" term (*i.e.* gravitational acceleration) with "rw^2" (*i.e.* centrifugal acceleration) have also been suggested [7,9-10]. Predictions from these correlations need to be compared against experimental data to determine their prediction uncertainty and thereafter identify the most accurate options for predicting mass transfer coefficients in RPBs.

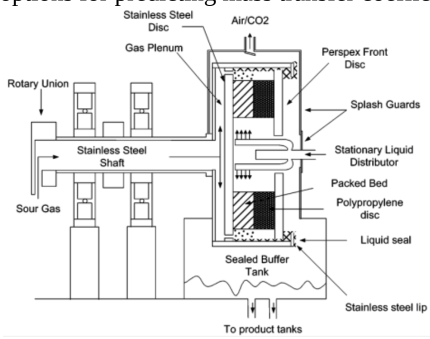

Fig. 1. Tangential sectional view of an RPB [9]

1.3. Aim and objective of this study

As discussed in Section 1.2, only a few options are reported in literature for estimating interfacial area and liquid film mass transfer coefficients in RPBs. It is a question mark to judge better option among them. Kang *et al*. [7] and Joel *et al*. [10] attempted comparing and validating some of them through process simulation. In their work, the mass transfer correlations were organised in sets and each set was used separately in their model. The problem with this approach is that several correlations are changed in the model, so individual performance of the correlations cannot be seen. What they showed instead was that some set of correlations were better than others. In this study, the aim is to compare and validate the correlations individually using experimental data obtained from literature.

2. Effective interfacial area

2.1 Experimental data and correlations

In literature, effective interfacial area data was derived from measurements of CO_2 absorption in NaOH solutions [1,11-12] based on the approach proposed by Sharma and Danckwerts [13]. Data from Lou *et al*. [1] was selected for this work. Data from Munjal *et al*. [12] is for glass bead packings; wire mesh packings are preferred as they are proven to have better mass transfer performance and rigidity for RPB [5,9]. On the other hand, data from Luo *et al*. [1] included several points and necessary parameters are given making it possible for the data to be used for validation purposes. Five correlations for interfacial area in RPB have been evaluated in this study (Table 1). These includes popular correlations for conventional packed bed, namely Onda *et al*. [14], Billet and Schultes [15] and Puranik and Vogelpohl [16], which have been used commonly for RPB design and modelling [4,7,9]. Others include Luo *et al*. [1] and Rajan *et al*. [8] which are developed for RPBs. The correlations have been simulated using gPROMS ModelBuilder® with physical properties obtained from Aspen Plus® through CAPE OPEN interface.

2.2 Results

The results (Fig.2) show that the predictions with Luo *et al*. [1] correlation provide the best agreement with experimental data. Modified Onda *et al*. [14] correlation with "g" term replaced by "$r\omega^2$" term which is widely used in literature [4,7,9] underpredicts the effective interfacial area by nearly 50%. More accurate prediction is obtained with modified Billet and Schultes [15] correlation (*i.e.* with "g" term replaced by "$r\omega^2$" term) although the deviation becomes increasing large at high rotating speed. Predictions of Puranik and Vogelpohl [16] correlation shows nearly 50% deviation. Comparing them with others at different rotating speed also highlights the impact of centrifugal acceleration. An important finding is that it is impossible to use correlations that do not explicitly account for centrifugal acceleration to estimate interfacial area in RPBs. In contrast, Puranik and Vogelpohl [16] correlation has been used successfully in conventional packed beds. Finally, performance of Rajan *et al*. [8] correlation which is developed for RPB was a bit surprising. The predictions deviated by nearly 50%. The large error of Rajan *et al*. [8] correlation is attributed to the split packing configuration used in the RPB for their experiments as opposed to single packing configuration used as basis in this study.

Table 1 Correlations for calculating effective interfacial area in RPB

Correlations	**Source**	**Comment**
$\frac{a}{a_t} = 1 - exp\left[-1.45\left(\frac{\sigma_c}{\sigma_L}\right)^{0.75}\left(\frac{L_m^*}{a_t\mu_L}\right)^{0.1}\left(\frac{a_t L_m^{2*}}{r\omega^2\rho_L^2}\right)^{-0.05}\left(\frac{L_m^{2*}}{\sigma_L\rho_L a_t}\right)^{0.2}\right]$	Onda *et al*. [14]	These correlations have been modified for RPB by replacing the "g" term with "$r\omega^2$" term.
$\frac{a}{a_t} = 1.5(a_t d_h)^{-0.5}\left(\frac{\rho_L u_L d_h}{\mu_L}\right)^{-0.2}\left(\frac{\rho_L u_L^2 d_h}{\sigma_L}\right)^{0.75}\left(\frac{u_L^2}{r\omega^2 d_h}\right)^{-0.45}$	Billet and Schultes [15]	
$\frac{a}{a_t} = 1.045\left(\frac{L_m^*}{a_t\mu_L}\right)^{0.041}\left(\frac{L_m^{2*}}{\sigma_L\rho_L a_t}\right)^{0.133}\left(\frac{\sigma_c}{\sigma_L}\right)^{0.182}$	Puranik and Vogelpohl [16]	This do not have a "g" term. They have been selected to know if good predictions are possible in RPB without explicitly accounting for acceleration.
$\frac{a}{a_t} = 54999\left(\frac{\rho_L d_p u_L}{\mu_L}\right)^{-2.2186}\left(\frac{u_L^2}{r\omega^2 d_p}\right)^{-0.1748}\left(\frac{\rho_L d_p u_L^2}{\sigma_L}\right)^{1.3160}$	Rajan *et al*. [8]	These correlations are developed for RPB. Rajan *et al*. [8] used split packing rotated by separate co-rotated motors.
$\frac{a}{a_t} = 66510\left(\frac{\rho_L d_p u_L}{\mu_L}\right)^{-1.41}\left(\frac{u_L^2}{r\omega^2 d_p}\right)^{-0.12}\left(\frac{\rho_L d_p u_L^2}{\sigma_L}\right)^{1.21}\left(\frac{c^2}{(c+d)^2}\right)^{-0.74}$	Luo *et al*. [1]	

3. Liquid mass transfer coefficient

3.1 Experimental data and correlations

Mass transfer in RPBs has been studied widely although the $k_L a$ instead of k_L are generally determined due to the difficulties in estimating the interfacial area, a [17]. Measurement of k_L have been reported by Luo *et al*. [18] and the experimental data has been selected for independently verifying different correlations for liquid film mass transfer coefficients in this study. The data were derived from measurements of CO_2 absorption in NaOH solutions based on the approach proposed by Sharma and Danckwerts [13]. The reaction kinetics were assumed to be pseudo-first order kinetics and mass transfer controlled by the liquid phase resistance. Two correlations, namely Tung and Mah [2] and Chen *et al*. [5] were selected for comparison (Table 2). Both correlations are developed for RPB. Tung and Mah [2] is simpler and requires less parameters than the Chen *et al*. [5] correlation. Similarly, the correlations have been simulated using gPROMS ModelBuilder® with physical properties obtained from Aspen Plus® through CAPE OPEN interface.

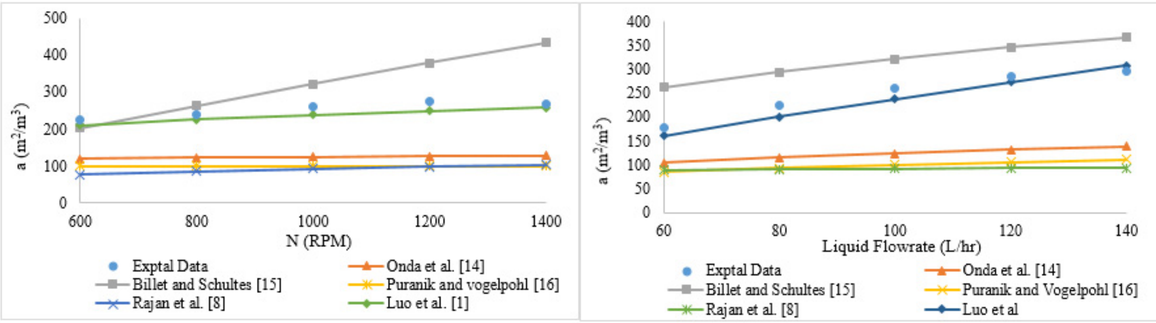

Fig. 2. Validation of different correlations for effective interfacial area predictions

Table 2 Correlations for liquid mass transfer coefficient

Correlations	**Source**	**Comment**
$\frac{k_L d_p}{D_L} = 0.919 \left(\frac{\mu_L}{D_L \rho_L}\right)^{\frac{1}{2}} \left(\frac{L_m^*}{\mu_L a_t}\right)^{\frac{1}{3}} \left(\frac{a_t}{a}\right)^{\frac{1}{3}} \left(\frac{d_p^3 \rho_L^2 r \omega^2}{\mu_L^2}\right)^{\frac{1}{6}}$	Tang and Mah [2]	Theoretical model based on penetration theory. Do not account for end effect phenomenon
$\frac{k_L a d_p}{D_L a_t} V = 0.35 \left(\frac{\mu_L}{D_L \rho_L}\right)^{0.5} \left(\frac{L_m^*}{\mu_L a_t}\right)^{0.17} \left(\frac{d_p^3 \rho_L^2 r \omega^2}{\mu_L^2}\right)^{0.3} \left(\frac{L_m^{2*}}{\sigma_L \rho_L a_t}\right)^{0.3} \left(\frac{a_t}{a_p^i}\right)^{-0.5} \left(\frac{\sigma_c}{\sigma_L}\right)^{0.14}$	Chen *et al.* [5]	Empirical model for predicting $\boldsymbol{k_L a}$. Accounts for end effect phenomenon. $\boldsymbol{k_L}$ predicted by combining the correlation with Luo *et al.* [1] correlation for interfacial area, a.

3.2 Results

The results shown in Fig. 3 show that Tung and Mah [2] gives more accurate predictions at different conditions than Chen *et al* [5]. The deviation of Chen *et al*. [5] becomes significant at high rotating speed. This is interesting as Tung and Mah [2] is simpler, requires less parameters and most of all does not account for end effect. This is attributed to the following:

- The Chen *et al*. [5] correlation includes a fixed parameter of 3000 m^2/m^3 which is surface area of 2-mm bead packings used in their experiment. Although it is claimed that this correlation gives good predictions for different packings, validations against Luo *et al*. [18] done in this study which involve wire mesh packings suggests that this is not the case at high rotating speed.
- The original formulation of Chen *et al*. [5] is to calculate $k_L a$. It appears that combining the correlation with Luo *et al*. [1] correlation for interfacial area, a, to obtain k_L has contributed to the uncertainty leading to the higher deviations of Chen *et al*. [13] compared to Tung and Mah [2].

In summary, the maximum deviation of Chen et al. [5] observed at 1400 RPM is about 11% which is reasonable considering uncertainties in physical properties and Luo *et al*. [1] used for estimating the interfacial area.

4. Conclusions and recommendations for future research

In this study, existing correlations for effective interfacial area and liquid film mass transfer coefficient were compared against experimental data at different rotating speed and liquid flowrate. For effective interfacial area, five correlations were assessed. It was found that Onda *et al*. [20] and Puranik and Vogelpohl [22] give poor prediction. Also, Rajan *et al*. [12] which was developed for RPBs gives a poor prediction of the effective interfacial area. Luo *et al* [1] alongside Billet and Schultes [21] predictions were closest to the experiment data. For liquid phase mass transfer coefficient, two correlations were assessed. Tung and Mah [2] gave more accurate predictions at different conditions. Chen *et al* [13] deviates significantly at high rotating speed by about 11%. In conclusion, correlations used in conventional packed column updated with "rw^2" term do not give acceptable prediction of effective interfacial area, prediction error is close to 50%. On the other hand, Tung and Mah [2] gives more accurate predictions than the more complex Chen *et al*. [13] correlation. In the future, similar validations as reported here should be performed for the gas side mass transfer coefficient. This will help establish assumptions in literature that the gas side in RPB have "solid-body" like characteristics so that the gas film mass transfer coefficient lies in the same range as in their conventional packed bed counterpart.

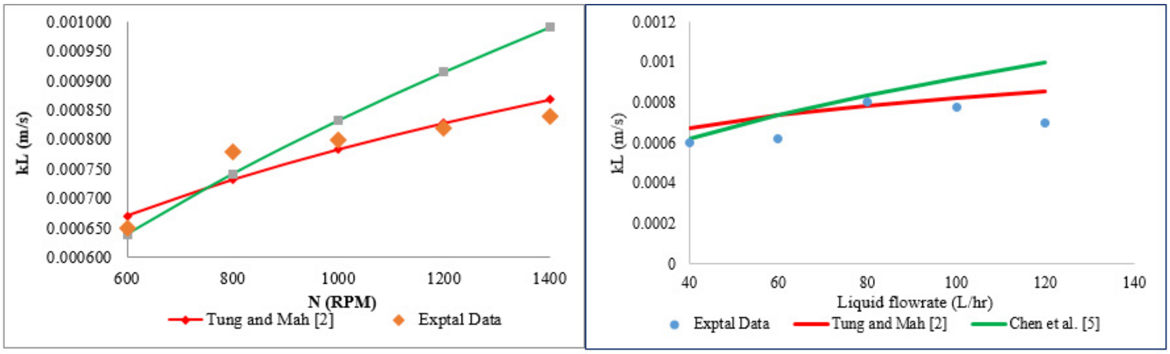

Fig. 3. Validation of different correlations for effective interfacial area predictions

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