

Study of mass transfer correlations for rotating packed bed columns in the context of solvent-based carbon capture

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Abstract

The application of rotating packed beds (RPBs) in solvent-based carbon capture processes, will greatly reduce the physical footprint, capital and operating cost of the process. However, in designing RPBs, correlations for predicting mass transfer parameters are generally limited in literature and their prediction accuracies have not been demonstrated independently. In this paper, an RPB absorber model was developed in gPROMS ModelBuilder® and used to test and compare different correlations for predicting the effective interfacial area, liquid and gas film mass transfer coefficients. Our results showed that the modified packed column mass transfer correlations where the “g” term (*i.e.* gravitational acceleration) is replaced with “ rw^2 ” (*i.e.* centrifugal acceleration) commonly used in literature for RPBs generally give poor predictions compared to using correlations developed specifically for RPBs. Also, the Tung and Mah correlation has better predictive accuracy for the liquid film mass transfer coefficient in RPBs than more complex correlations. Finally, a set of new data for the gas film mass transfer coefficient for RPBs were also derived from overall volumetric mass transfer coefficient ($K_G a$) experimental data from literature. This is the first report of gas film mass transfer data for RPBs. The results in this paper will guide researchers in selecting suitable correlations for predicting mass transfer parameters in RPBs.

Keywords: solvent-based CO₂ capture; rotating packed bed; effective interfacial area; liquid film mass transfer coefficient; gas film mass transfer coefficient

Nomenclature

a	Effective interfacial area of packing per unit volume (m ² /m ³)
a_t	Total 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 <i>et al.</i> (2012a) correlation ($c = 3.5$ mm, $d = 1.0$ mm)
C_p^L	Liquid specific heat capacity (J/kg K)
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)
D_G	Gas diffusivity (m ² /s)
E	Enhancement factor
G^m	Gas molar flowrate (kmol/s)
h_G	Gas phase specific molar enthalpy (J/kmol)
h_L	Liquid phase specific molar enthalpy (J/kmol)
$h_{g/l}$	Interfacial heat transfer coefficient (W/m ² K)
H	Henry constant
ΔH_r	Heat of absorption (J/kmol)
ΔH_{vap}	Heat of vaporisation of H ₂ O (J/kmol)
k_G	Gas film mass transfer coefficient (m/s)
$K_G a$	Overall volumetric mass transfer coefficient based on gas side (1/s)
k_L	Liquid film mass transfer coefficient (m/s)
$K_L a$	Overall volumetric mass transfer coefficient based on liquid side (1/s)
k_{app}	Apparent reaction rate constant
L^m	Liquid molar flowrate (kmol/s)
L_m^*	Liquid mass flowrate per unit tangential section area (kg/m ² s)
N_i	Component molar fluxes (kmol/m ² s)
Q_L	Liquid volumetric flowrate (m ³ /s)
Q_V	Gas 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)
RPM	Revolutions per minute
T_g, T_l	Gas and liquid side temperature (K)
u_L	Liquid velocity (m/s)
V_G	Parameter for Chen <i>et al.</i> (2011) gas film model = $1 - 0.9 \frac{V_o}{V_t}$
V_L	Parameter for Chen <i>et al.</i> (2006) liquid film model = $1 - 0.93 \frac{V_o}{V_t} - 1.13 \frac{V_i}{V_t}$
V_i	Volume inside the inner radius of the bed (m^3) = $\pi r_i^2 Z$
V_m^*	Gas mass flowrate per unit tangential section area ($\text{kg}/\text{m}^2 \text{ s}$)
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) = $\pi r_s^2 Z$
x_i	Component molar fraction in liquid phase
y_i	Component molar fraction in gas phase
Z	Height of the rotor (m)

Greek Letters

σ_c	Critical surface tension for packing material (= 0.075 N/m)
σ_L	Liquid surface tension (N/m)
ε	Packing porosity (m^3/m^3)
ρ_G	Gas density (kg/m^3)
ρ_L	Liquid density (kg/m^3)
λ_L	Liquid thermal conductivity (W/m K)
μ_G	Gas dynamic viscosity (Pa s)
μ_L	Liquid dynamic viscosity (Pa s)
ω	Rotating speed (rad/s)

22 1. Introduction

23 1.1 Background

24 The gas-liquid packed columns are an important unit operation in natural gas treating and solvent-based CO₂
25 capture processes where they are used for absorption and desorption. The packed columns in these processes are
26 large in size, contributing significantly to physical footprint, capital and operating costs (Lawal *et al.*, 2012;
27 IEAGHG, 2013; Oko, 2015). An engineering estimate showed that absorbers in a solvent-based CO₂ capture
28 (PCC) plant using monoethanolamine (MEA) solvent for capturing CO₂ from a 500 MWe coal-fired subcritical
29 power plant will have diameters up to 25 m and packing height over 27 m (Oko, 2015). This will significantly
30 increase the land use per MWe when coal and gas fired power plants are integrated with PCC plants (Florin and
31 Fennel, n.d.).

32 Through process intensification (PI), wherein the packed columns are replaced with rotating packed beds (RPBs),
33 the physical footprint of the process could be reduced significantly (Joel *et al.*, 2014; Thiels *et al.*, 2016).
34 Theoretical investigations by Agarwal *et al.* (2010) and Joel *et al.* (2014) showed about 10-12 times reduction in
35 the absorber size when it is replaced with an RPB. The HiGee Environment and Energy Technologies Inc. USA
36 also reported about 10 times size reduction in a commercial scale RPB installed to replace a packed column at the
37 Fujian Refining and Petrochemical Company Ltd, China (HiGee, 2014). The reported size reductions are
38 consistent with predictions about RPBs in earlier investigations by Chambers and Wall (1954) and Ramshaw and
39 Mallinson (1981).

40 1.2 Principle of RPB and problem statement

41 The RPB generally includes a cased annular packed bed (rotor), made of packing materials such as glass bead
42 (Munjal *et al.*, 1989a&b), corrugated disk (Chen *et al.*, 1997; Chen *et al.*, 1999), wire mesh (Luo *et al.*, 2012),
43 expamet (Jassim *et al.*, 2007), blade packing with static baffles (Tsai and Chen, 2015), nickel foam (Chu *et al.*,

2015) etc. and mounted on a rotating shaft (Fig. 1). The gas and liquid phases enter the RPB through the outer and inner sections respectively, each flowing radially as shown in Figure 1 Sectional view of an RPB . 1. The gas-liquid flow are usually countercurrent flow, but co-current and cross flow configurations are also possible (Kolawole *et al.*, 2018, Oko *et al.*, 2018). As the RPB rotates, the liquid and gas phases are subjected to intense centrifugal acceleration which is many times the gravitational acceleration in packed columns. As a result, the RPB generally allows:

- Higher flooding limit leading to drastic reduction in packing volume (Guo *et al.*, 1997; Chen *et al.*, 2008; Garcia *et al.*, 2017)
- Lower liquid holdup and consequently achieves steady state more quickly (Nascimento *et al.*, 2009)
- More viscous solvents e.g. 80-100 wt% MEA solvent (Chambers and Wall, 1954; Jassim *et al.*, 2007; Oko *et al.* 2018).

Consequently, similar capture levels (in CO₂ capture applications) as in packed columns can be achieved in RPBs using significantly reduced packing volume (Agarwal *et al.*, 2010; Joel *et al.*, 2014; Thiels *et al.*, 2016). However, the presence of centrifugal force field in RPBs presents new research challenge as mass transfer correlations for packed columns cannot be used to predict mass transfer in RPBs with acceptable accuracy (Joel *et al.*, 2014; Kang *et al.*, 2014).

Only a few correlations have been reported for predicting effective interfacial area, liquid and gas film mass transfer coefficients (Tung and Mah, 1985; Munjal *et al.*, 1989a; Chen *et al.*, 2006a; Chen *et al.*, 2006b; Chen *et al.*, 2006; Chen, 2011; Rajan *et al.*, 2011; Luo *et al.*, 2012). Modification of mass transfer correlations for packed columns such as Onda *et al.* (1968) and Billets and Schultes (1999) correlations by replacing the “g” term (*i.e.* gravitational acceleration) with “ rw^2 ” (*i.e.* centrifugal acceleration) have also been recommended and widely used (Joel *et al.*, 2014; Kang *et al.*, 2014; Thiels *et al.*, 2016). There has not been a clear independent demonstration of the performance of the various mass transfer correlations for RPBs against experimental data. This will highlight the strengths and weaknesses of various options and provide a basis for determining the most accurate option for predicting mass transfer parameters in RPBs.

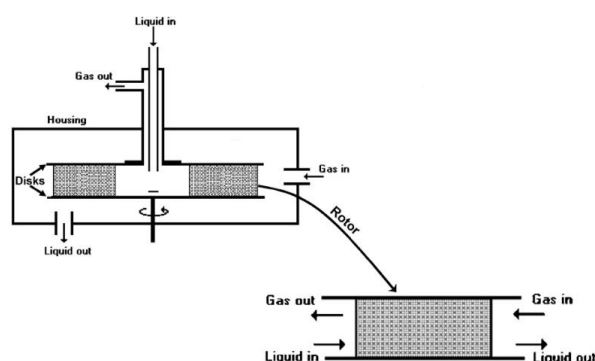


Figure 1 Sectional view of an RPB (Llerena-Chavez and Larachi 2009)

1.3 Aim of this study

As noted earlier, the predictive accuracies of mass transfer correlations for RPBs (Tung and Mah, 1985; Munjal *et al.*, 1989a; Chen *et al.*, 2006a; Chen *et al.*, 2006b; Chen *et al.*, 2006; Chen, 2011; Rajan *et al.*, 2011; Luo *et al.*, 2012), including modified mass transfer correlations for packed columns (Onda *et al.*, 1968; Billets and Schultes, 1999) ought to be independently assessed. Joel *et al.* (2014) and Kang *et al.* (2014) attempted comparing and validating some of the correlations through process simulation. In their work, the mass transfer correlations were organised in sets – each set including correlations for predicting effective interfacial area, liquid and gas film mass transfer coefficients – and used separately in their model. Their RPB models were then validated using experimental data from RPB rigs. In their approach, several correlations are changed at a time in the model, and as such the individual performance of the correlations cannot be seen. What the authors (Joel *et al.*, 2014; Kang *et al.*, 2014) showed instead was that some sets of correlations were better than others. In this study, the aim is to provide a comprehensive review of existing correlations, compare and validate the correlations individually using experimental data obtained from literature.

1.4 Novel contribution

This study provides an extensive review and comparison of all published correlations for estimating different mass transfer parameters for RPBs, namely effective interfacial area, liquid and gas film mass transfer coefficients. As noted in Section 1.3, related study had been reported by Joel *et al.* (2014) and Kang *et al.* (2014). However, neither study included comparisons for gas film mass transfer coefficient and they considered the correlations in sets (See Section 1.3) and validated overall predictions of their RPB model and not specific predictions of the mass transfer parameters. This study will therefore address the following gaps identified from existing studies:

- No information on performance of correlations for predicting gas film mass transfer coefficients for RPBs
- No specific performance comparison of different mass transfer correlations for RPBs.
- No data for the gas film mass transfer coefficient for RPBs. An assessment comparing liquid and gas film resistances to mass transfer for CO₂ absorption in different MEA concentrations in an RPB absorber (Table 1) show that the gas film resistance could be over 10% of the overall resistance and cannot be ignored. Obtaining data for the gas film mass transfer coefficient is therefore essential. The gas film mass transfer coefficient data were derived from overall volumetric mass transfer coefficient ($K_G a$) experimental data from the literature.

Table 1: Liquid and gas film resistances for an RPB absorber with MEA solvent*

MEA (wt%)	Liquid film resistance (Pa m ² s/mol)	Gas film resistance (Pa m ² s/mol)
55	240490.2	23265.93
75	172447.4	25305.32

*The liquid and gas film resistances have been obtained using conditions from Jassim *et al.* (2007) and reaction data from Ying and Eimer (2013). The liquid and gas film mass transfer coefficients were respectively obtained using Tung and Mah (1985) and Chen (2011).

2. Methodology – model development

The mass transfer parameters for the RPB derived from experimental measurements are reported in literature. In this study, selected RPB absorber rigs from literature used for deriving different mass transfer parameters are represented using models derived from first principle. The details of the selected rigs are given in Section 3 (effective interfacial area), Section 4 (liquid film mass transfer coefficient) and Section 5 (gas film mass transfer coefficient). In the RPB absorber model, different mass transfer correlations (See Sections 3, 4 & 5) are used to predict mass transfer parameters. The predicted values for different correlations are then compared to their counterpart derived from experimental measurements for the selected case in the literature. The RPB absorber model, developed using gPROMS ModelBuilder®, are represented using Equations 1-9. The thermo-physical properties are obtained using a combination of the electrolyte Non-Random Two-Liquid (elecNRTL) model in Aspen Plus® and data obtained from the literature. The elecNRTL model is accessed from gPROMS ModelBuilder® platform through the CAPE-OPEN interface. The model has been validated for CO₂ absorption in MEA cases and presented in Oko *et al.* (2018). The following assumptions have been made in developing the model:

- Steady state conditions.
- One-dimensional differential mass and energy balances for liquid and gas phases
- Heat losses are neglected
- Heat and mass transfer are described using the two-film theory
- Reactions (where applicable) are accounted for using an enhancement factor in the overall mass transfer coefficient

Material balance

$$\text{Gas phase: } 0 = \frac{1}{2\pi r Z} \frac{\partial(G^m y_i)}{\partial r} - a N_i \quad (1)$$

$$\text{Liquid phase: } 0 = -\frac{1}{2\pi r Z} \frac{\partial(L^m x_i)}{\partial r} + a N_i \quad (2)$$

Energy balance

$$\text{Gas phase: } 0 = \frac{1}{2\pi r Z} \frac{\partial(G^m h_G)}{\partial r} - a h_{g/l} (T_l - T_g) \quad (3)$$

$$\text{Liquid phase: } 0 = -\frac{1}{2\pi r Z} \frac{\partial(L^m h_L)}{\partial r} + a(h_{g/l}(T_l - T_g) - \Delta H_r N_{CO_2} - \Delta H_{vap} N_{H_2O}) \quad (4)$$

The molar fluxes for molecular components are obtained as follows based on the two-film theory:

$$N_i = K_{G,i} (P_{g,i} - P_i^{eq}) \quad (5)$$

The overall mass transfer coefficient ($K_{G,i}$) comprise of mass transfer resistances on both the gas and liquid film (Eqn. 6). $P_{g,i}$ and P_i^{eq} are respectively gas phase component partial pressure and component equilibrium partial pressure in the liquid phase.

$$K_{G,i} = \frac{1}{\left(\frac{RT_g}{k_{G,i}}\right) + \left(\frac{H}{k_{L,i}E}\right)} \quad (6)$$

The enhancement factor (E) is used to account for the reactions in reactive cases. The enhancement factor (E) is obtained on the basis of a pseudo first-order reaction regime as given in Eqn 7.

$$E = \frac{\sqrt{k_{app} D_{L,CO_2}}}{k_{L,CO_2}} \quad (7)$$

Finally, the interfacial heat transfer coefficient ($h_{g/l}$) is obtained based on the Chilton-Colburn analogy:

$$h_{g/l} = k_L \rho_L C_p^L \left(\frac{\lambda_L}{\rho_L C_p^L D_L} \right)^{\frac{2}{3}} \quad (8)$$

3. Case 1: Effective interfacial area

3.1 Experimental data and correlations

In the literature, effective interfacial area data for RPB have been derived from measurements of CO₂ absorption in NaOH solutions (Munjali *et al.*, 1989b; Chen *et al.*, 1997; Chen *et al.*, 1999; Rajan *et al.*, 2011; Yang *et al.*, 2011; Luo *et al.*, 2012a; Guo *et al.*, 2014; Chu *et al.*, 2015; Liu *et al.*, 2015; Tsai and Chen, 2015; Luo *et al.*, 2017) based on the approach proposed by Sharma and Danckwerts (1970). The reported data are mainly packing effective interfacial area; a few studies (Yang *et al.*, 2011; Guo *et al.*, 2014; Luo *et al.*, 2017) reported the effective interfacial area for the different mass transfer zones, namely the packing, cavity and the end zones. It was found that the packing effective interfacial area makeup more than half of the total effective interfacial area (Yang *et al.*, 2011). Recent studies have also investigated the effective interfacial area for novel packing designs, namely blade packing RPB with static baffles (Tsai and Chen, 2015), nickel foam packing (Chu *et al.*, 2015) and structured wire mesh packing (Luo *et al.*, 2017). Changes in the packing design was shown to have significant impact on the effective interfacial area (Tsai and Chen, 2015). The data from Luo *et al.* (2012a) was selected for this work. The Luo *et al.* (2012a) experiments comprised of a 1M NaOH solution as the liquid phase and a mixed CO₂ and N₂ gas with approximately 10 mol% of CO₂ as the gas phase. The data is preferred to the data from other sources for the following reasons:

- The RPB used for obtaining the measurements (Table 2 Specification of RPB from is equipped with wire mesh packing. Wire mesh packings are proven to be very suitable for RPBs due to their better mass transfer performance and rigidity (Chen *et al.* 2006). Munjal *et al.* (1989b) data was obtained from an RPB with glass bead packing. Chen *et al.* (1997) and Chen *et al.* (1999) data were obtained from an RPB with corrugated disk packings.
- The packing is a traditional RPB design, wherein the packings are loaded uniformly across the radial depth of the RPB without gaps in-between packing rings, so called unsplit packing configuration (Figure 2). The packing is held between two disks and rotated by a single motor. Rajan *et al.* (2011) and Liu *et al.* (2015) on the other hand are based on split packing configuration, a relatively new packing design for

RPBs. The split packing configuration comprise of alternate annular packing rings attached to two separate disks with a small radial gap between adjacent rings when the two disks are brought together (Figure 3) with the disks rotated by two separate motors counter-currently or co-currently.

- Finally, the experimental data include several data points and relevant parameters making it more convenient for the data to be reproduced through modelling.

Five correlations for predicting effective interfacial area in RPB have been evaluated in this study (Table 3 Correlations for calculating effective interfacial area in RPB). These include popular correlations for packed columns, namely Onda *et al.* (1968), Billets and Schultes (1999) and Puranik and Vogelpohl (1974), which have been used commonly for RPB design and modelling (Jassim *et al.*, 2007; Joel *et al.*, 2014; Kang *et al.*, 2014). Others include Rajan *et al.* (2011) and Luo *et al.* (2012a) which were developed specifically for RPBs. Luo *et al.* (2017) proposed a new correlation for structured wire mesh packings as Luo *et al.* (2012a), developed for unstructured wire mesh packing, was not good enough for structured wire mesh packings (Luo *et al.*, 2017). The new correlation (Luo *et al.*, 2017) was not included in this study as we are focused on unstructured wired mesh packings. In addition, Lin *et al.* (2000) proposed a correlation for predicting the packing wetting area in RPBs. The correlation (Lin *et al.*, 2000) was found to be obviously inaccurate for predicting the effective interfacial area and as a result was excluded from our evaluations.

Table 2 Specification of RPB from Luo *et al.* (2012a)

Dimensions (mm)				Packing		
r_i	r_o	r_s	Z	Type	a_t	ϵ
78	158	248	50	Wire mesh	400	0.90

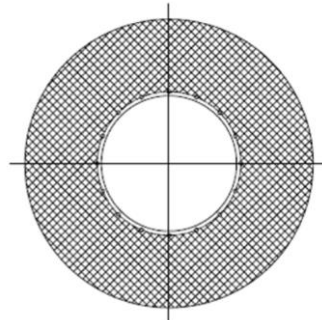


Figure 2 Unsplit packing configuration for RPB (Luo *et al.*, 2012b)

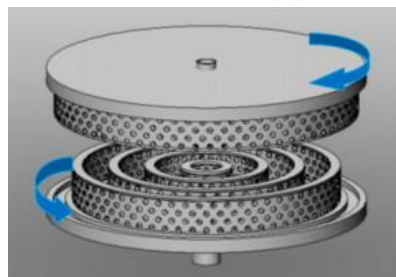


Figure 3 Split packing configuration for RPB (Liu *et al.*, 2015)

3.2 Results and discussion

For each of the cases (involving different correlations for predicting the effective interfacial area), the Tung and Mah (1985) and Chen (2011) were used for predicting the liquid and gas film mass transfer coefficients. The

reported results are also mean values of the effective interfacial area across the flow direction in the RPB. The results in Figures 4 and 5 show that the predictions with Luo *et al.* (2012a) correlation provide the best agreement with experimental data. Modified Onda *et al.* (1968) correlation with “g” term replaced by “ $r\omega^2$ ” term which is widely used in literature for RPB design and modelling (Jassim *et al.*, 2007; Joel *et al.*, 2014; Kang *et al.*, 2014) underpredicts the effective interfacial area by about 50% and this increased with flowrate (Figure 5). The predictions of Onda *et al.* (1968) correlation with “g” term replaced by “ $r\omega^2$ ” term do not quite show impact of rotational speed on the effective interfacial area (Figure 4). More accurate prediction is obtained with modified Billets and Schultes (1999) correlation (*i.e.* with “g” term replaced by “ $r\omega^2$ ” term) although the deviation becomes increasingly large at high rotating speed. The predictions of Puranik and Vogelpohl (1974) correlation show nearly 50% deviation. Comparing the predictions of Puranik and Vogelpohl (1974) with others at different rotating speed also highlight the impact of centrifugal acceleration. Although, Puranik and Vogelpohl (1974) correlation has been used successfully for packed columns, they clearly show poor prediction accuracy for RPBs. This partly because the correlation that do not have an acceleration term. Finally, the performance of Rajan *et al.* (2011) correlation which is developed for RPB was a bit surprising. The predictions deviated by nearly 50%. The Rajan *et al.* (2011) correlation is based on split packing configuration and this is viewed as a major reason for the large error when the correlation is used for predicting the effective interfacial area for unsplit packing configuration in this study. It is recommended that the Rajan *et al.* (2011) correlation be used for split packing configuration cases only as it clearly underperforms for unsplit packing configuration case as shown in this study.

Table 3 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 <i>et al.</i> (1968)	These correlations have been modified for RPB by replacing the “g” term with “ $r\omega^2$ ” term.
$\frac{a}{a_t} = 1.5 (a_t d_h)^{-0.5} \left(\frac{\rho_L u_L d_h}{\mu_L} \right)^{-0.2} \left(\frac{\rho_L u_L^2 d_h}{\sigma_L} \right)^{0.75} \left(\frac{u_L^2}{r \omega^2 d_h} \right)^{-0.45}$	Billets and Schultes (1999)	
$\frac{a}{a_t} = 1.045 \left(\frac{L_m^*}{a_t \mu_L} \right)^{0.041} \left(\frac{L_m^{2*}}{\sigma_L \rho_L a_t} \right)^{0.133} \left(\frac{\sigma_c}{\sigma_L} \right)^{0.182}$	Puranik and Vogelpohl (1974)	This do not have a “g” term. It was 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 <i>et al.</i> (2011)	These correlations are developed for RPB. Rajan <i>et al.</i> (2011) is based on split packing type RPB rotated by two separate 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 <i>et al.</i> (2012a)	

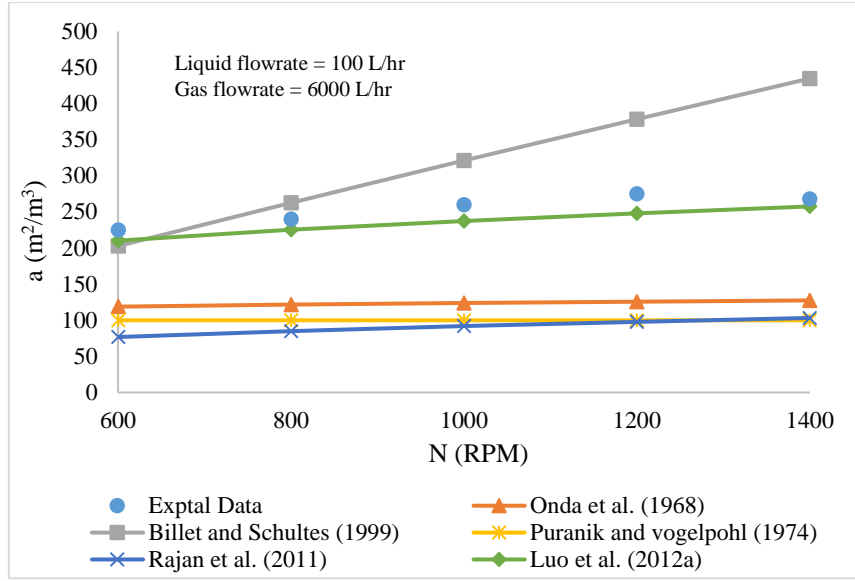


Figure 4 Predictions of different correlations for effective interfacial area at different RPM

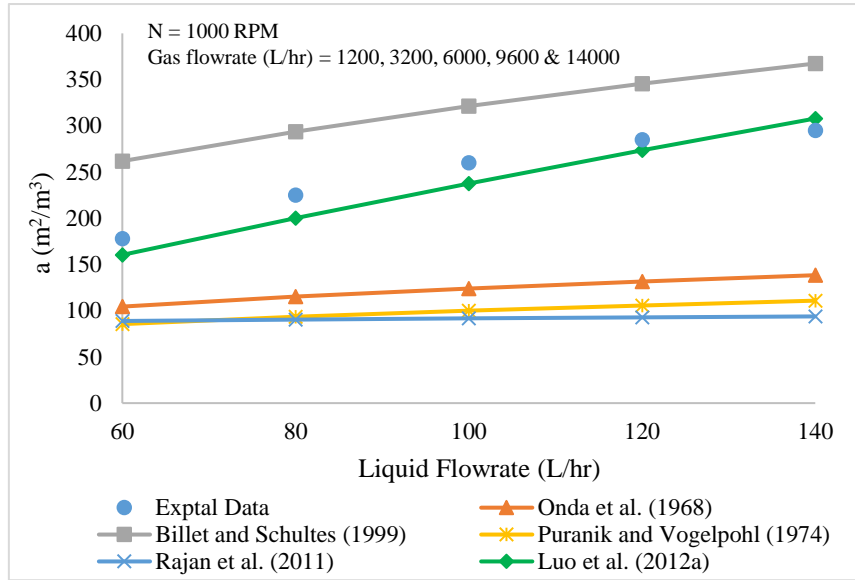


Figure 5 Predictions of different correlations for effective interfacial area at different liquid flowrate

4. Case 2: Liquid film mass transfer coefficient (k_L)

4.1 Experimental data and correlation

The study of liquid side mass transfer in RPBs is reported widely in literature, although it is the overall volumetric mass transfer coefficients ($K_L a$) and the volumetric liquid side mass transfer coefficients ($k_L a$) rather than the liquid film mass transfer coefficient (*i.e.* k_L) that are generally determined from experiments due to the difficulties in estimating the effective interfacial area in RPBs (Chen *et al.*, 2005a; Chen *et al.*, 2005b; Chen *et al.*, 2006; Lin and Liu, 2007). The $K_L a$ is the overall driving force for the liquid phase while the $k_L a$ is the driving force for the liquid film only. The only existing k_L data is reported by Luo *et al.* (2012b) and the experimental data has been selected for independently verifying different correlations for predicting liquid film mass transfer coefficients in this study. The data were derived from measurements of CO₂ absorption in NaOH solutions based on the approach proposed by Sharma and Danckwerts (1970). The authors (Luo *et al.*, 2012b) assumed a pseudo-first order reaction kinetics regime with mass transfer controlled by the liquid phase resistance. The liquid and gas phases were respectively 0.05 M NaOH solution and a mixed gas of CO₂ and N₂ with about 2 mol % of CO₂. A summary of the RPB parameters from Luo *et al.* (2012b) is given in Table 4.

Table 4 Specification of RPB from Luo *et al.* (2012b)

Dimensions (mm)				Packing		
r_i	r_o	r_s	Z	Type	a_t	ε
78	153	248	50	Wire mesh	500	0.96

Presently, only five correlations (Table 5) for predicting the liquid film mass transfer coefficient have been published in literature (Tung and Mah, 1985; Munjal *et al.*, 1989a; Chen *et al.*, 2005a; Chen *et al.*, 2005b; Chen *et al.*, 2006); an elaborate model based on surface renewal theory has also been proposed by Ding *et al.* (2000). The reported correlations were developed from theoretical principle based on penetration theory (Tung and Mah, 1985; Munjal *et al.*, 1989a) and the film theory (Chen *et al.*, 2005a; Chen *et al.*, 2005b; Chen *et al.*, 2006). In this study, the performance of these five correlations, the summary of which are given in Table 5 Correlations for calculating liquid film mass transfer coefficient in RPB5, are evaluated. The modified Onda *et al.* (1968) was selected to demonstrate their performance for predicting k_L for RPBs. The Tung and Mah (1985) correlation is simpler and requires fewer parameters compared to others. The Chen *et al.* (2006) correlation on the other hand is very elaborate, accounting for both the packing geometry and mass transfer in the end zones otherwise called the end effect phenomenon.

Table 5 Correlations for calculating liquid film mass transfer coefficient in RPB

Correlations	Source	Comment
$k_L \left(\frac{\rho_L}{\mu_L r \omega^2} \right)^{\frac{1}{3}} = 0.0051 \left(\frac{L_m^*}{a \mu_L} \right)^{\frac{2}{3}} \left(\frac{\mu_L}{\rho_L D_L} \right)^{\frac{1}{2}} (a_t d_p)^{0.4}$	Onda <i>et al.</i> (1968)	Modified for RPB by replacing the “g” term with “ $r\omega^2$ ” term.
$\frac{k_L d_p}{D_L} = 0.918 \left(\frac{\mu_L}{D_L \rho_L} \right)^{\frac{1}{2}} \left(\frac{L_m^*}{\mu_L a_t} \right)^{\frac{1}{3}} \left(\frac{a_t}{a} \right)^{\frac{1}{3}} \left(\frac{d_p^3 \rho_L^2 r \omega^2}{\mu_L^2} \right)^{\frac{1}{6}}$	Tung and Mah (1985)	The correlations are developed for predicting k_L in RPBs. They do not account for end effect and packing type
$k_L = 2.6 \frac{\pi L_m^*}{2 a \rho_L X} \left(\frac{\mu_L}{D_L \rho_L} \right)^{\frac{1}{2}} \left(\frac{2 \pi L_m^*}{\mu_L a_t} \right)^{\frac{2}{3}} \left(\frac{X^3 \rho_L^2 r \omega^2}{\mu_L^2} \right)^{\frac{1}{6}}$	Munjal <i>et al.</i> (1989a)	
$\frac{k_L a d_p}{D_L a_t} = 0.9 \left(\frac{\mu_L}{D_L \rho_L} \right)^{0.5} \left(\frac{L_m^*}{\mu_L a_t} \right)^{0.24} \left(\frac{d_p^3 \rho_L^2 r \omega^2}{\mu_L^2} \right)^{0.29} \left(\frac{L_m^{2*}}{\sigma_L \rho_L a_t} \right)^{0.29}$	Chen <i>et al.</i> (2005a)	The correlations are developed for predicting $k_L a$ in RPBs. Chen <i>et al.</i> (2006) accounts for both end effect and packing type
$\frac{k_L a d_p}{D_L a_t} V_L = 0.65 \left(\frac{\mu_L}{D_L \rho_L} \right)^{0.5} \left(\frac{L_m^*}{\mu_L a_t} \right)^{0.17} \left(\frac{d_p^3 \rho_L^2 r \omega^2}{\mu_L^2} \right)^{0.3} \left(\frac{L_m^{2*}}{\sigma_L \rho_L a_t} \right)^{0.3}$	Chen <i>et al.</i> (2005b)	
$\frac{k_L a d_p}{D_L a_t} V_L = 0.35 \left(\frac{\mu_L}{D_L \rho_L} \right)^{0.5} \left(\frac{L_m^*}{\mu_L a_t} \right)^{0.17} \left(\frac{d_p^3 \rho_L^2 r \omega^2}{\mu_L^2} \right)^{0.3} \left(\frac{L_m^{2*}}{\sigma_L \rho_L a_t} \right)^{0.3} \left(\frac{a_t}{a_p} \right)^{-0.5} \left(\frac{\sigma_c}{\sigma_L} \right)^{0.14}$	Chen <i>et al.</i> (2006)	

4.2 Results and discussion

The Luo *et al.* (2012a) correlations for effective interfacial area, demonstrated in Case 1 to give good predictions for unsplit and unstructured wire mesh packing, was used to predict the effective interfacial area for all the cases. Although, this potentially increases the prediction uncertainty, the results (Figure 6 and 7) show a reasonably good agreement for Tung and Mah (1985) and Chen *et al.* (2006). The gas film mass transfer coefficient was predicted

for all cases using Chen (2011). The reported results are mean values of the liquid film mass transfer coefficient area across the flow direction in the RPB. The results further showed that the predictions of Tung and Mah (1985), Chen *et al.* (2005a), Chen *et al.* (2005b) and Chen *et al.* (2006) correlations for liquid phase mass transfer coefficient were in the order of 10^{-4} . This is a typical range for liquid film mass transfer coefficients for RPBs which have been generally reported in the literature (Rao *et al.*, 2004). The correlations of Onda *et al.* (1968) and Munjal *et al.* (1989a) respectively showed under-prediction and over-prediction in the orders of 10^{-5} and 10^{-3} at different rotating speed and liquid flowrate (Figures 8 and 9). The predictions of Onda *et al.* (1968) were in the typical range for the packed columns. It is concluded that modifying Onda *et al.* (1968) correlation by replacing the “g” term with “ $r\omega^2$ ” term do not result in good estimation of the liquid film mass transfer coefficient in RPBs.

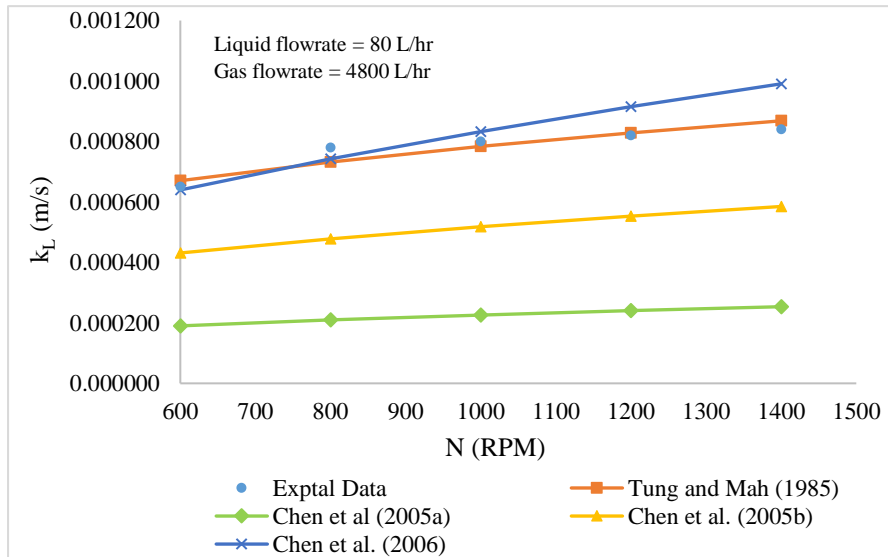


Figure 6 Liquid film mass transfer coefficient at different RPM

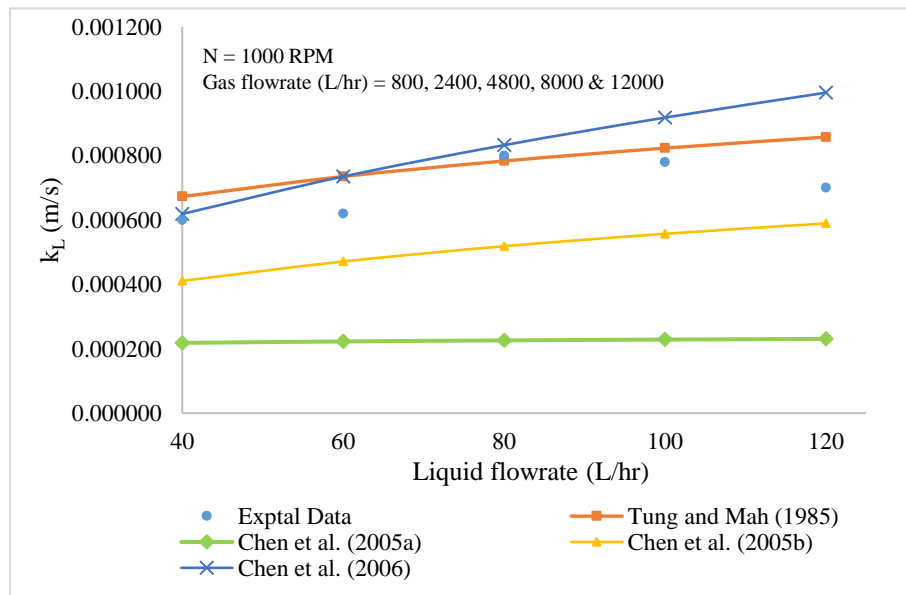


Figure 7 Liquid film mass transfer coefficient at different liquid flowrate

Comparing the Chen correlations (Chen *et al.*, 2005a; Chen *et al.*, 2005b; Chen *et al.*, 2006), Chen *et al.* (2006) gave the best prediction compared to others. This is because apparently more extensive data set that cover different packing types, fluid types (Newtonian and non-Newtonian) and radial depth were used to develop the correlation. The Chen *et al.* (2006) and Tung and Mah (1985) correlations gave the best prediction for all conditions. The performance of Tung and Mah (1985) is particularly interesting as it is simpler, requiring fewer parameters and most of all does not account for the end effect and the packing type. The predictions of Chen *et al.* (2006)

correlation, a supposedly more robust correlation that accounts for both end effect and packing type, tend to deviate as rotating speed and liquid flowrate increased. This deviation could be as a result of a combination of uncertainties from interfacial area and physical property predictions. Regardless, the maximum deviation was about 11% which is acceptable for most applications.

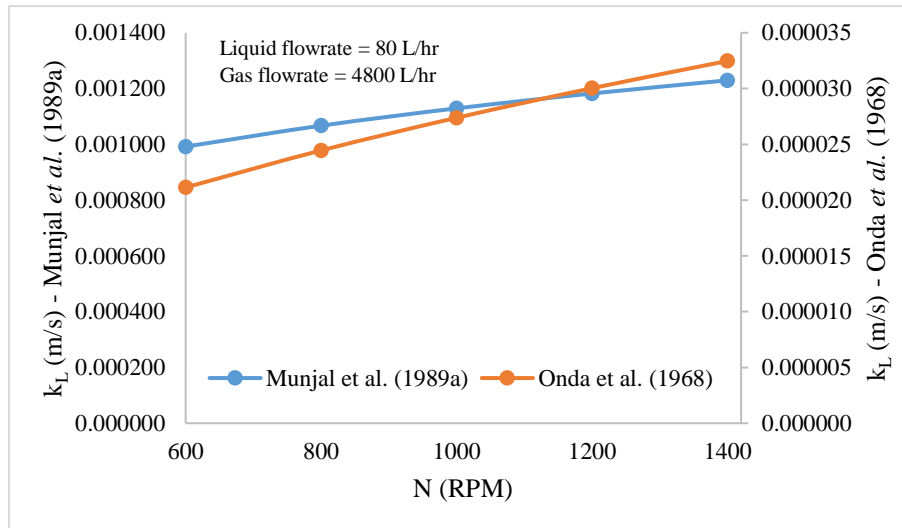


Figure 8 Liquid film mass transfer coefficient at different RPM

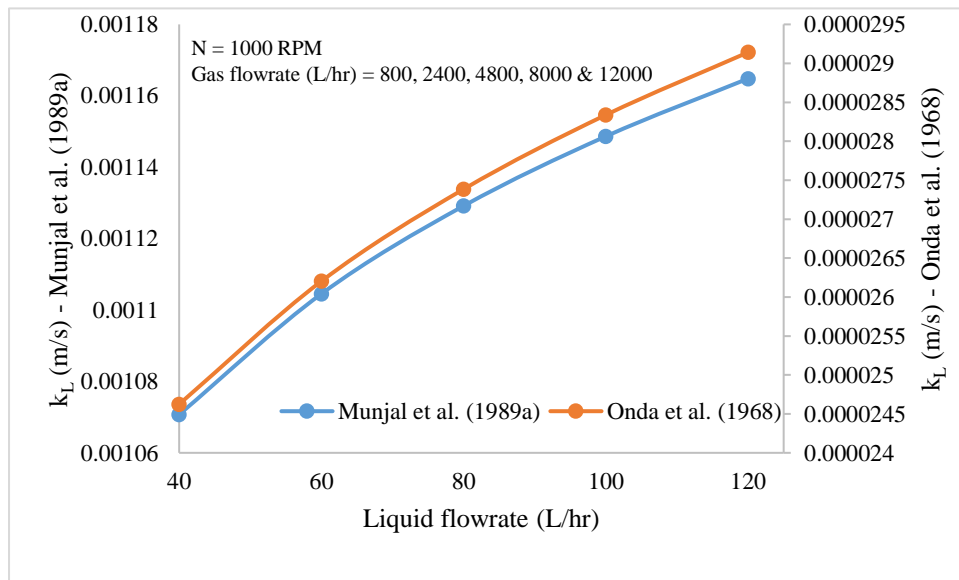


Figure 9 Liquid film mass transfer coefficient at different liquid flowrate

5. Case 3: Gas film mass transfer coefficient (k_G)

5.1 Experimental data and correlations

Experimental data for the gas film mass transfer coefficient (k_G) calculation for RPBs are not available in the literature. What have generally been reported are overall volumetric mass transfer coefficient (*i.e.* K_{Ga}). The K_{Ga} data are obtained using mass balance and the transfer unit concept (Liu *et al.*, 1996; Chen and Liu, 2002; Lin *et al.*, 2003; Lin *et al.*, 2004; Chiang *et al.*, 2009; Lin and Chu, 2015). The gas film volumetric mass transfer coefficient (*i.e.* $k_G a$) data have also been reported by Chen (2011). The $k_G a$ data from Chen (2011) was obtained based on the two-film theory (Eqn. 6) using a combination of published K_{Ga} data and $k_L a$ data predicted using the Chen *et al.* (2006) correlation. In this study, a similar approach as Chen (2011) has been adopted to obtain gas

film mass transfer coefficient data (k_G) from published K_{Ga} data alongside effective interfacial area and k_L data predicted using Luo *et al.* (2012a) and Tung and Mah (1985) correlations respectively. Two independent sources, namely Lin *et al.* (2004) and Chiang *et al.* (2009), for K_{Ga} data were selected. The Lin *et al.* (2004) and Chiang *et al.* (2009) data involved isopropyl alcohol absorption in water and ethanol absorption in water respectively. Parameters of the RPB rigs used in both cases are summarized in Table 6.

Table 6 Specification of RPB from Lin *et al.* (2004) and Chiang *et al.* (2009)

	Dimensions (mm)				Packing		
	r_i	r_o	r_s	Z	Type	a_t	ϵ
Lin <i>et al.</i> (2004)	35	80	150	35	Wire mesh	791	0.96
Chiang <i>et al.</i> (2009)	20	40	60	20	Wire mesh	1024	0.944

Existing correlations for the *gas-side mass transfer coefficients* are presented in Table 7. The correlations of Lin *et al.* (2004), Liu *et al.* (1996) and Chen and Liu (2002) are formulated for predicting overall volumetric mass transfer coefficient (K_{Ga}). The gas film mass transfer coefficient (k_G) can be calculated from these correlations using Eqn 6. This will involve predicting several parameters namely the Henry's constant, enhancement factor (where applicable), liquid film mass transfer coefficient, effective interfacial area and physical properties such as density, viscosity and surface tension. The uncertainties in predicting these parameters could result in significant error in the gas film mass transfer coefficient. Therefore it was concluded that the correlations proposed in Lin *et al.* (2004), Liu *et al.* (1996) and Chen and Liu (2002) are not good options for predicting the gas film mass transfer coefficient and was therefore not considered for validation in this study.

Table 7 Correlations for calculating gas-side mass transfer coefficient in RPBs

Correlations	Source	Comment
$\frac{k_G}{a_t D_G} = K_5 \left(\frac{V_m^*}{\mu_G a_t} \right)^{0.7} \left(\frac{\mu_G}{\rho_G D_G} \right)^{\frac{1}{3}} (a_t d_p)^{-2.0}$	Onda <i>et al.</i> (1968)	Correlation for predicting k_G in packed columns
$\frac{K_{Ga} a}{D_G a_t^2} = 3.11 \times 10^{-3} \left(\frac{V_m^*}{\mu_G a_t} \right)^{1.163} \left(\frac{L_m^*}{\mu_L a_t} \right)^{0.631} \left(\frac{d_p^3 \rho_G^2 r \omega^2}{\mu_G^2} \right)^{0.25}$	Liu <i>et al.</i> (1996)	These correlations are developed for predicting K_{Ga} in RPBs. The k_G can then be derived from the predicted $K_{Ga} a$ data using Eqn 6.
$\frac{K_{Ga} a H^{0.27}}{D_G a_t^2} = 0.077 \left(\frac{V_m^*}{\mu_G a_t} \right)^{0.323} \left(\frac{L_m^*}{\mu_L a_t} \right)^{0.328} \left(\frac{d_p^3 \rho_G^2 r \omega^2}{\mu_G^2} \right)^{0.18}$	Chen and Liu (2002)	
$\frac{K_{Ga} a H^{0.315}}{D_G a_t^2} = 0.061 \left(\frac{V_m^*}{\mu_G a_t} \right)^{0.712} \left(\frac{L_m^*}{\mu_L a_t} \right)^{0.507} \left(\frac{d_p^3 \rho_G^2 r \omega^2}{\mu_G^2} \right)^{0.326}$	Lin <i>et al.</i> (2004)	
$\frac{k_G a}{D_G a_t^2} V_G = K_n \left(\frac{V_m^*}{\mu_G a_t} \right)^{1.13} \left(\frac{L_m^*}{\mu_L a_t} \right)^{0.14} \left(\frac{d_p^3 \rho_G^2 r \omega^2}{\mu_G^2} \right)^{0.31} \left(\frac{L_m^{2*}}{\sigma_L \rho_L a_t} \right)^{0.07} \left(\frac{a_t}{a_p} \right)^{1.4}$	Chen (2011)	The correlation is developed for predicting $k_G a$ in RPBs

Mukherjee *et al.* (2001) and Sandilya *et al.* (2001) had suggested that the gas phase undergoes a 'solid-body'-like rotation within the rotor of an RPB because of the drag offered by the packing and as a result suggested there was no enhancement of the gas film volumetric mass transfer resistance (k_{Ga}). Consequently, they concluded that the gas film volumetric mass transfer coefficient in RPBs was in similar range as that of packed columns. This was further demonstrated in Chen and Liu (2002) where it was shown that enhancements in k_{Ga} are mainly attributed to the interfacial area (a), while k_G remain in similar range as that of the packed columns. These conclusions have prompted the use of Onda *et al.* (1968) for predicting the gas film mass transfer coefficient in most published studies of RPBs (Joel *et al.*, 2014; Kang *et al.*, 2014). As a result, Onda *et al.* (1968) correlation for gas-film mass transfer coefficient was also evaluated in this study.

The effective interfacial area has been predicted using the Luo *et al.* (2012a) and the liquid film mass transfer coefficient predicted using Tung and Mah (1985). The reported results are mean values of the gas film mass transfer coefficient across the flow direction in the RPB. The results of comparison between predicted values and experimental data (Figures 10 and 11) show that for the two experimental data considered in this study (Lin *et al.*, 2004; Chiang *et al.*, 2009), the Onda *et al.* (1968) correlation significantly over-predicts the k_G in the RPBs for different rotating speed and gas flowrate. The predictions of Onda *et al.* (1968) are in the order of 10^{-1} in contrast to order of 10^{-2} values for the experimental data. Predictions of Chen (2011) on the other hand were in the order of 10^{-3} . With the parameter K_n updated from 0.023 to 0.23, there was good agreement between the predictions of Chen (2011) and the experimental data for the two independent data sources in this study. Onda *et al.* (1968) correlation do not have a “g” term and as such they do not show the influence of centrifugal acceleration when they are used for predicting the k_G in RPBs (Figure 10a and 11a). The experimental data as well as predictions of Chen (2011) both show that rotation enhances gas side resistance. Figures 10a and 11a both show that increasing rotating speed from 700-1600 RPM (Lin *et al.*, 2004) and 600-1800 RPM (Chiang *et al.*, 2009) respectively will enhance gas side resistance by up to 30%. While k_G appears to be in similar range as that of packed columns in agreement with the conclusions reached in Mukherjee *et al.* (2001) and Sandilya *et al.* (2001), their actual values are however affected by rotating speed. It is not possible to capture this impact when k_G in RPBs are predicted using Onda *et al.* (1968).

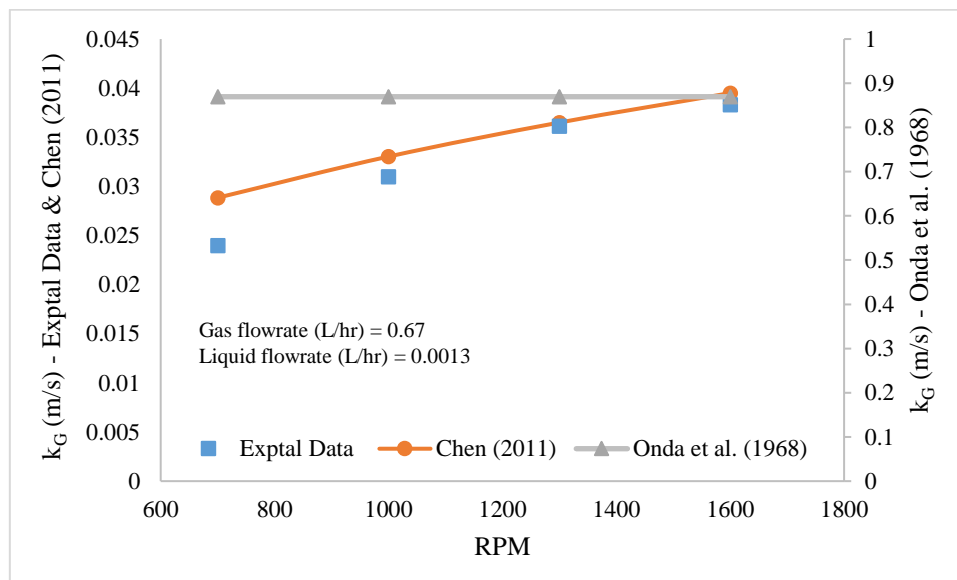


Figure 10a Comparisons to experimental data from Lin *et al.* (2004) data

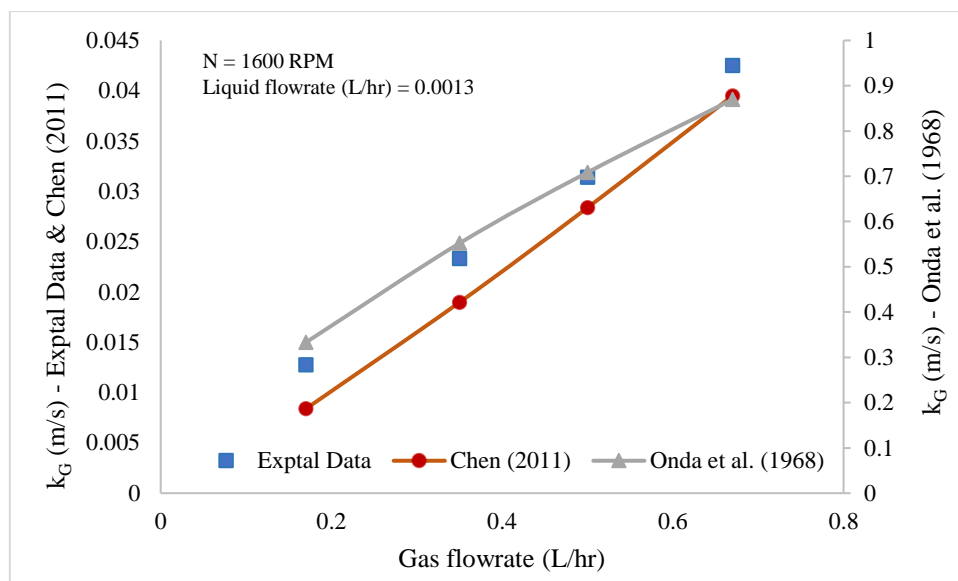


Figure 10b Comparisons to experimental data from Lin et al. (2004) data

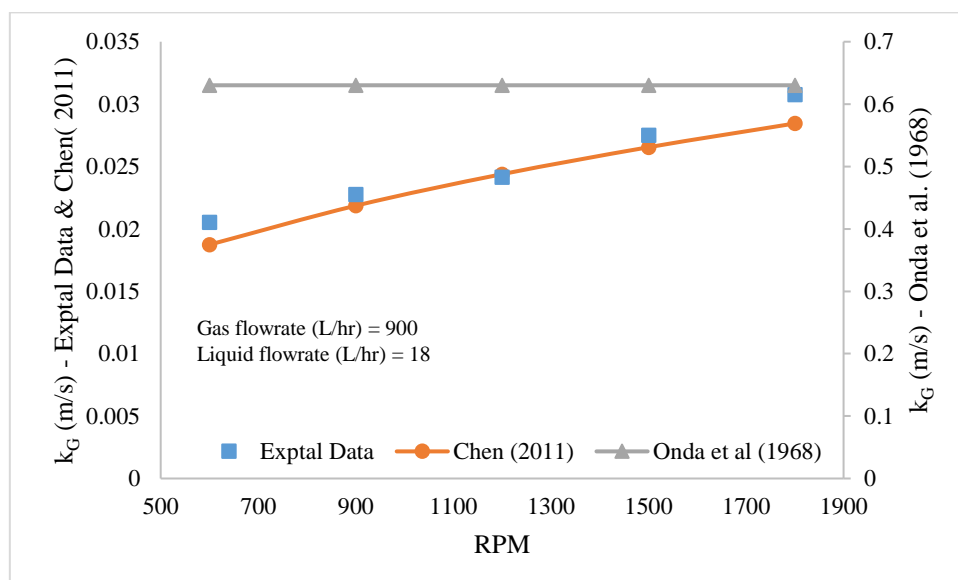


Figure 11a Comparisons to Chiang et al. (2009) data

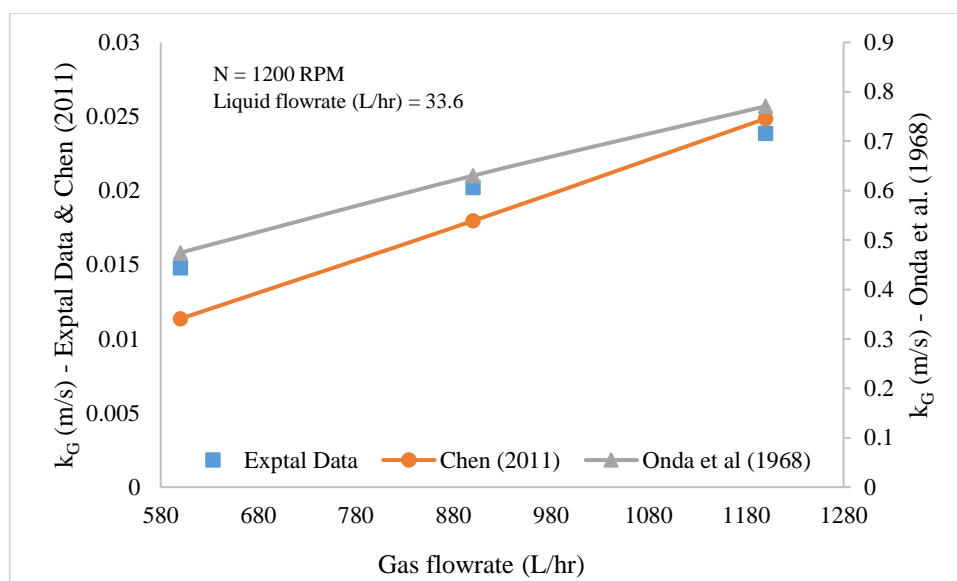


Figure 11a Comparisons to Chiang et al. (2009) data

6. Conclusions and recommendations for future study

The RPB is a promising technology that can greatly reduce the size and cost of packed columns used for absorption and desorption in solvent-based CO₂ capture and natural gas treating processes. Mass transfer prediction in RPBs is not sufficiently proven because necessary correlations for doing this are generally few in literature and the prediction accuracy for existing correlations have not been demonstrated independently. In this study, an RPB model was developed in gPROMS ModelBuilder® and used to test and compare different correlations for the effective interfacial area, the liquid and gas film mass transfer coefficients at different rotating speed and liquid/gas flowrate. The results presented in this paper show that modified packed column mass transfer correlations with the “g” term (*i.e.* gravitational acceleration) replaced with “ rw^2 ” (*i.e.* centrifugal acceleration) commonly used in literature for RPBs generally give poor predictions. The Tung and Mah (1985) correlation gave a good prediction of liquid film mass transfer coefficient in RPBs, slightly better than more complex correlations such as the Chen *et al.* (2006). The data of gas film mass transfer coefficient for RPBs were also derived from overall mass transfer coefficient ($K_G a$) experimental data from the literature. This is the first report of gas film mass transfer data for RPBs in literature. Finally, we showed that Chen (2011) fitted the experimental data of gas film mass transfer coefficient better when the parameter (K_n) is updated from 0.023 to 0.23. The validity of the analysis and conclusions in this study are based on a single type of packing (unstructured wire mesh). With other packing types, namely expamet and retimet among others, the performance of the correlations may be very different. For instance, efforts in our group to use Luo *et al.* (2012a) for predicting effective interfacial area of expamet packings showed that the predicted values were out of range, although more data is needed to confirm this finding. The performance of the correlations should be demonstrated for other types of packings for RPB such as expamet and retimet as the relevant data become available.

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