IMPACT OF UNCERTAINTY OF PHYSICAL PROPERTIES ON CO_2 ABSORPTION DESIGN

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Abstract

The mass transfer coefficients, interfacial area and pressure drop of a packed bed are essential properties that need to be evaluated prior to the design of a CO_2 absorption column. Various mathematical models have been proposed to predict these properties under different process conditions. This work has compared several mathematical models for pressure drop, mass transfer coefficients and interfacial area and discussed how the uncertainty of physical properties and process conditions affect the evaluation of packed bed height in a CO_2 absorption column. A case study has been performed to study the propagation of uncertainty in input variables through the packed bed height design equations. Here, it was found as 12% from uncertainty in physical properties and 60% from uncertainty in choice of mathematical model of the calculated packed bed height. A recommended safety factor for the absorption packing height is 60 % for a generic packing, but this safety factor can be reduced considerably if experimental data for pressure drop and mass transfer coefficients are available for the specific packing.

Keywords: *CO_2 capture, mass transfer coefficients, pressure drop, column design, uncertainty*

1. Introduction

Physicochemical properties like density, viscosity and surface tension are vital in the design of process equipment such as absorption and desorption columns, heat exchangers, reboilers, condensers and pumps in post combustion amine-based CO_2 capture. In the design of a CO_2 absorption column, the gas side and liquid side mass transfer coefficients and interfacial area can be calculated by proposed mathematical models under different liquid and gas flow rates. The proposed models for mass transfer coefficients are based on physical theories of wetted wall theory and penetration theory [1]. In addition to the traditional methods, the applicability of ANN (artificial neural networks) correlations for mass transfer coefficients and interfacial area have been discussed by Piche and co-workers [2].

In our previous work, the propagation of uncertainty of physical properties through mass transfer models was discussed [3]. It was observed that the uncertainty of viscosity has a high influence on mass transfer coefficients. These uncertainties further propagate through design equations for sizing of the packed bed in absorber columns. Therefore, it is important to decide a safety factor for packing height to acquire the desired CO_2 removal efficiencies. This study discusses the mass transfer coefficients, interfacial area and pressure drop calculations from available mathematical models in the literature. Further, the study discusses the effect of uncertainty of physical properties and other process parameters on the evaluation of packed bed height. The work performed by Nookuea *et al*. [4] discussed the impact of physical properties of gas and liquid on design of an absorption column. Kvamsdal and Hillestad [5], and Razi and Svendsen [6] investigated mass transfer and physical property models considering CO_2 absorption into aqueous MEA. Mathias *et al*. [7] performed a quantitative analysis of the effects of uncertainty in property models on the simulation of CO_2 capture. And Nookuea *et al*. [8] indicates that the density and diffusivity show opposite effect to viscosity in the estimation of packing height. A review of property impact on carbon capture and storage processes has been performed by Tan *et al*. [9].

2. Mass transfer coefficients, interfacial area and pressure drop calculations

Liquid hold-up and pressure drops have been calculated using Excel spreadsheets, by the methods in Rocha *et al*. [10], Billet and Schultes [11] and Stichlmair *et al*. [12]. All these correlations are based on the dimensionless numbers defined below (2 to 5):

$$d_E = 4 \cdot \frac{\varepsilon}{a_N} \tag{1}$$

$$Re_L = \frac{v_L \cdot d_E \cdot \rho_L}{\mu_L} \tag{2}$$

$$We = \frac{{v_L}^2 \cdot d_E \cdot \rho_L}{\sigma} \tag{3}$$

$$Fr = \frac{v_L{}^2}{g \cdot d_E} \qquad (4)$$

$$Sc_G = \frac{\mu_G}{\rho_G \cdot D_G} \qquad (5)$$

$$v_{REL} = \frac{v_G}{(1-h_L)\cdot \varepsilon \cdot 0.7071} \qquad (6)$$

The h_L was calculated using a correlation from Billet and Schultes [11] which is valid up to the loading point:

$$h_L = \left[\frac{12 \cdot \mu_L \cdot v_L \cdot a_N{}^2}{g \cdot \rho_L}\right]^{0.333} \qquad (7)$$

The liquid hold-up was calculated to 0.087 and 0.089 at the specified top and bottom conditions. A constant value of 0.09 was then used in later calculations of the other parameters in all the correlations.

Dry packing pressure drop and total pressure drop have been calculated by the correlations for pressure drop from Stichlmair *et al.* [12], from Billet and Schultes [11] and from Rocha *et al.* [10]. The equations used may differ slightly from the original correlations and are given in earlier work [13].

Pressure drops in dry packing (with only gas) and total pressure drop (with gas and liquid) for the conditions in Table 1 are calculated in Excel and the results for total drop are shown in Figure 1:

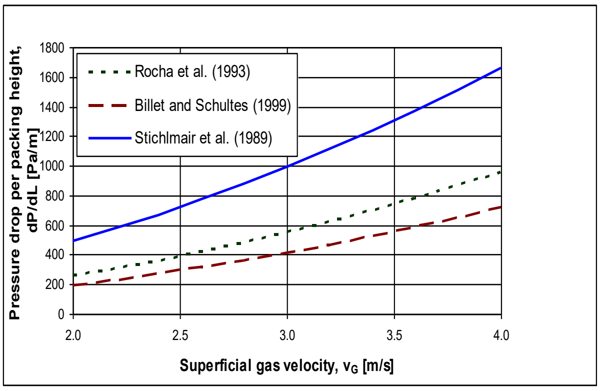

Figure 1: Calculated pressure drop from correlations using packing type Mellapak 250Y at typical CO_2 absorption top column conditions as a function of gas velocity. From [13].

As shown in Figure 1, the pressure drop increases with the increase of superficial gas velocities. The pressure drop from the correlation given by Stichlmair *et al.* [12] shows relatively large deviations from Billet and Schultes [11] and Rocha *et al.* [10] especially at high superficial gas velocities.

The effective relative interfacial areas for the conditions in Table 1 were determined based on estimation methods proposed in Rocha *et al.* [10], Billet and Schultes [11] and deBrito *et al.* [14]. The equations used are (8-10).

$$a_{EFF} = 0.465 \cdot \left(\frac{v_L \cdot \rho_L}{\mu_L \cdot a_N}\right)^{0.3} \qquad (8)$$

$$a_{EFF} = 1.5 \cdot (a_N \cdot d_E)^{-0.5} \cdot Re_L^{-0.2} \cdot We^{0.75} \cdot Fr^{-0.45} \qquad (9)$$

$$a_{EFF} = 0.35 \cdot 29.12 \cdot (We \cdot Fr)^{0.15} \cdot \frac{d_E^{0.359}}{Re_L^{0.2} \cdot \varepsilon^{0.6} \cdot (1-0.93\cdot 0.9)\cdot 0.7071^{0.3}} \qquad (10)$$

The calculated a_{EFF} from Rocha *et al.* [15], Billet and Schultes [11] and deBrito *et al.* [14] are presented in Figure 2.

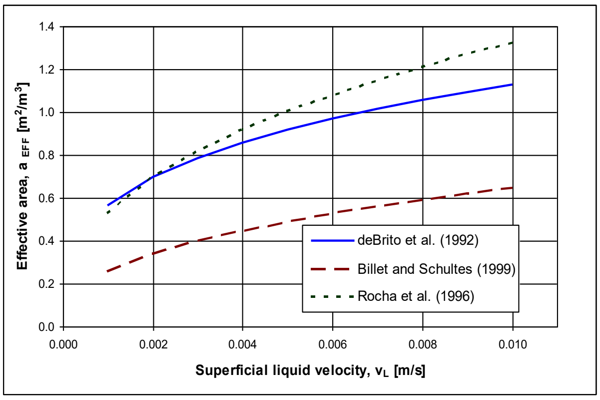

Figure 2: Calculated effective relative interfacial area from correlations at typical CO_2 absorption column top conditions as a function of superficial liquid velocity. From [13].

The interfacial area increases with superficial liquid velocity as shown in Figure 2 for all correlations. The model proposed by Billet and Schultes [11] underestimates the property compared to the other two models.

Gas side mass transfer coefficients have been calculated in a spreadsheet using the estimation methods from Rocha *et al.* [15], Billet and Schultes [11] and deBrito *et al.* [14]. The equation forms are the versions in Brunazzi *et al.* [16]. The equations used are defined in [13].

The packing type assumed in the calculations are Mellapak 250Y from Sulzer. The packing specific parameter (0.41) is specified to the average of the values from Billet and Schultes [11] for the Montz packings B1-200 and B1-300 which are similar packings with nominal specific areas of 200 and 300 m^2/m^3.

The physical properties liquid viscosity, gas viscosity and diffusion coefficients are calculated from the equations described in [13]. The calculated k_G from Rocha *et al.* [15], Billet and Schultes [11] and deBrito *et al.* [14] are presented in Figure 3.

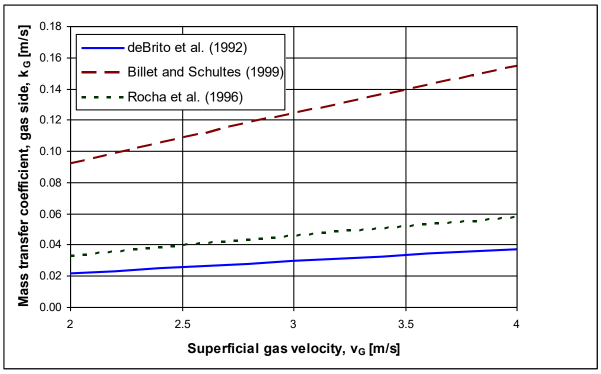

Figure 3: Calculated gas side mass transfer coefficients at typical CO_2 absorption column top conditions as a function of gas velocity. From [13].

The gas side mass transfer coefficient increases with the increase of superficial gas velocity. The considered models behave similarly with the variation of superficial gas velocity. A relatively high deviation is reported by Billet and Schultes [11] at both lower and higher superficial gas velocities compared to Rocha *et al.* [10] and deBrito *et al.* [14] as illustrated in Figure 3.

The liquid side mass transfer coefficients have been calculated in an Excel spreadsheet using the estimation methods from Rocha *et al.* [15], Billet and Schultes [11] and deBrito *et al.* [14]. The equation forms are the versions in Brunazzi *et al.* [16] and the equations are defined in [13]. The calculated k_L from Rocha *et al.* [15], Billet and Schultes [11] and deBrito *et al.* [14] are presented in Figure 4.

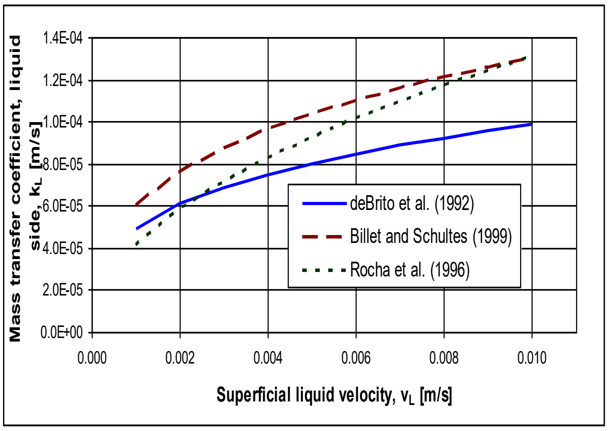

Figure 4: Calculated liquid side mass transfer coefficients as a function of liquid velocity. From [13].

The predicted liquid side mass transfer coefficient increases with the increase of liquid superficial velocity as shown in Figure 4. The deviations between the predictions from different models are less compared to the predictions for the gas side mass transfer coefficient. The model from deBrito *et al.* [14] underpredicts the liquid side mass transfer coefficient compared to models Billet and Schultes [11] and Rocha *et al.* [10] at higher superficial liquid velocity.

Table 1: Specifications for estimation of pressure drop, effective interfacial area and mass transfer coefficients at top condition in the absorber column.

Parameter	Top
Temperature, T [ºC]	49
Pressure, P [bar(a)]	1.01
Gas superficial velocity, v_G [m/s]	3.5
Liquid superficial velocity, v_L [m/s]	0.0041
Liquid density, ρ_L [kg/m^3]	1050
Gas density, ρ_G [kg/m^3]	1.02
Liquid viscosity, μ_L [kg/(m·s)]	0.0023
Gas viscosity, μ_G [kg/(m·s)]	0.000019
Surface tension, σ, [N/m]	0.055
Liquid CO_2 diffusivity, D_{CO2} [m^2/s]	$1.2\cdot10^{-9}$
Void fraction, ε [m^3/m^3]	0.97
Nominal surface area, a_N [m^2/m^3]	250
Side of corrugation, S [m]	0.017
Liquid hold-up, h_L [m^3/m^3]	0.09

3. Uncertainty in height calculation in absorption column

The height of the absorber packing is a function of several parameters like molar gas flow rate per unit cross sectional area G[mol/(m^2·s)], overall gas phase mass transfer coefficient K_G [mol/(m^2·Pa·s)], total pressure P [Pa], interfacial surface area a [m^2/m^3] and mole fractions y [-] of the gas inlet and outlet of the absorber. Typical design equations found in chemical engineering textbooks are (11 to 13).

$$Z = f(G, P, K_G, a, y_{CO_2}) \quad (11)$$

$$Z = \frac{G}{K_G \cdot a \cdot P} \int_{y_{CO2}\,out}^{y_{CO2}\,in} \frac{dy_{co_2}}{(y_{co_2} - y^*_{co_2})} \quad (12)$$

$$Z = \frac{G}{K_G \cdot a \cdot P} ln \left[\frac{y_{co_2}\ in}{y_{co_2}\ out}\right] \quad (13)$$

Figure 5 shows the cause-and-effect diagram to illustrate the uncertainty sources and their effect on absorber packing heigh calculation. The input variables in Equation 11 are identified as the main uncertainty sources and drawn as main branches. For the mass transfer coefficient and interfacial area, the physical properties of density, viscosity and surface tension were identified as uncertainty sources as they appear in most of the correlations. There can be other uncertainty sources in addition to the sources shown in Figure 5 and those are not discussed in here.

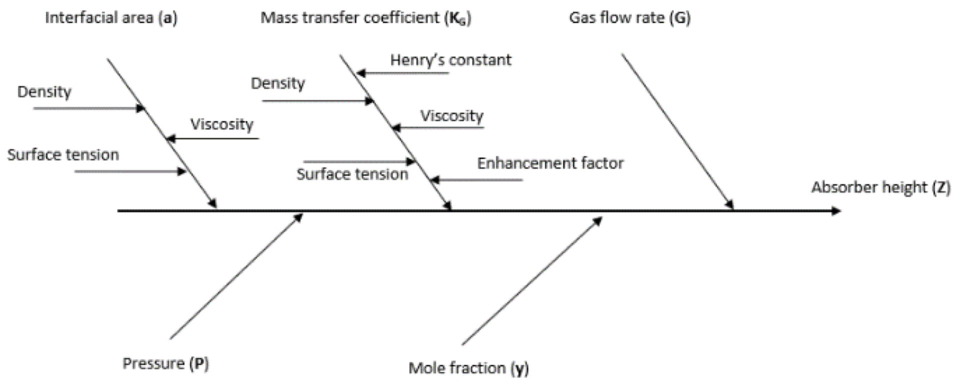

Figure 5: Cause and effect diagram

The uncertainties of the variables involved in Equation 13 are combined taking Root Sum Square (RSS). There the height of the packing is partially differentiated with respect to all the variables involved and combined as given in Equation 14.

$$\delta Z = \sqrt{\sum_{i=1,n} C_i^2 \cdot u(x_i)^2} \tag{14}$$

The individual contribution to the overall uncertainty was found from the term $|C_i \cdot u(x_i)|$. There, x_i and $u(x_i)$ are independent variables and associated uncertainties shown in Equation 14. The C_i are sensitivity coefficients evaluated as $C_i = \partial Z / \partial x_i$.

The following scenario with assumptions is considered in the evaluation of packed bed height. A column with a 2 m diameter was considered to calculate the gas flow rate into the column. Superficial gas and liquid flow rates were considered as given in Table 1. The expected CO_2 removal efficiency was 90%. Finally, the packed bed height was determined as 14.2 m according to the specification given in Table 1 for the absorption conditions at the column top.

Table 2: Uncertainties of the input variables.

Parameter	Uncertainty	
	Scenario 1 Karunarathne et al., 2017	Scenario 2 Øi, 2012
Gas flowrate (G)	1%	1%
Mass transfer coefficient (K_g)	5%	50%
Interfacial area (a)	4%	20%
Pressure drop (dP)	10%	30%
Mole fractions (y)	1%	1%

The considered uncertainties for this work are listed in Table 2. In scenario 2 the uncertainties for the mass transfer coefficient, interfacial area and pressure drop were decided from the experience from previous studies based on uncertainty evaluations of mass transfer coefficient and interfacial area of proposed mathematical models in literature for random packings [13]. In scenario 1 the uncertainties raised due to the propagation of uncertainties in physical properties of density, viscosity and surface tension through mass transfer and interfacial area models were considered in Karunarathne *et al.* [3]. The uncertainty in pressure drop was considered due to the pressure drop of the column and other uncertainties were due to the possible variations in the feed conditions.

The calculated error from Equation 14 is the standard uncertainty for the absorber packing height. In this case, it is ±1.7 m and it is 12% of the calculated packing height for the uncertainties based on Karunarathne *et al.* [3]. The uncertainty in packing heigh is ± 9 m and it is 60% of the calculated packing height for the uncertainties Øi [13]. The increased uncertainties in mass transfer coefficient, interfacial area and column pressure caused to increase the uncertainty in packed bed height. The uncertainty in absorber packing height from uncertainty in physical properties was calculated to 12 %. The uncertainty in absorber packing height from uncertainty due to different correlations was calculated to 60 %.

The calculated individual contributions to the overall uncertainty are shown in Figure 6 and 7 for the two scenarios given in Table 2.

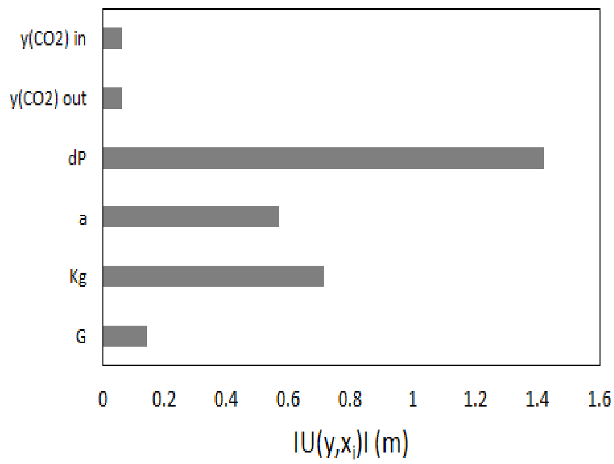

Figure 6: Uncertainty contributions from different uncertainty sources for scenario 1.

For the uncertainties considered in scenario 1, the highest individual contribution for the uncertainty of packing height was the pressure drop. For the scenario 2, the uncertainty of mass transfer coefficient has the highest uncertainty among the other uncertainty sources and it gives the highest individual contribution for the uncertainty of packing height.

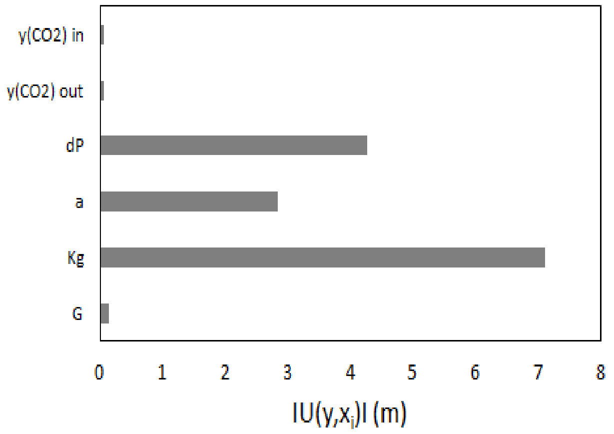

Figure 7: Uncertainty contributions from different uncertainty sources for scenario 2.

To keep a design within the uncertainty, a safety factor of 60 % in absorber packing height is calculated in this work. To reduce this large safety factor, especially the uncertainty in the correlations for the gas side mass transfer coefficient and for the pressure drop should be reduced.

For a generic packing, an uncertainty in pressure drop of 30 % as in this work is regarded as reasonable. However, for a specific packing with experimental pressure drop data, this uncertainty can be reduced.

For a generic packing, an uncertainty in gas side mass transfer coefficient of 50 % is regarded as reasonable. For specific conditions with a specified packing, a much lower uncertainty can be expected.

As a result a recommended safety factor for the absorption packing height is 60 % for a generic packing, but this safety factor can be reduced considerably with available experimental data for pressure drop and gas side mass transfer coefficients at actual conditions.

4. Conclusion

This study discusses the calculation of pressure drop, mass transfer coefficient and interfacial area of packing in an absorption column using mathematical models available in the literature. All the models show similar behaviours under the variation of gas and liquid superficial velocity.

The uncertainties in process conditions and physical properties affect the height of a packed bed in an absorber column. An uncertainty analysis as discussed leads to an evaluation of the safety margins that need to be considered in absorber design. Two scenarios were discussed considering different values for the uncertainty sources and observed how it affects the height calculation of the packed bed in an absorption column. In the first scenario, the uncertainty in pressure drop gave the largest impact and in the second scenario the uncertainty in mass transfer coefficient gave the largest impact.

A recommended safety factor for the absorption packing height is 60 % for a generic packing, but this safety factor can be reduced considerably if experimental data for pressure drop and mass transfer coefficients are available for the specific packing.

Nomenclature

Latin symbols

a	Specific area (m^2/m^3)
D	Diffusivity coefficient (m^2/s)
d	Diameter (m)
Fr	Froude's number
G	Molar gas flow rate per unit cross sectional area ($mol/(m^2{\cdot}s)$)
g	Acceleration of gravity (m/s^2)
h_L	Liquid hold-up
K_G	Overall mass transfer coefficient ($kmol/(m^2{\cdot}Pa{\cdot}s)$)
P	Pressure (Pa), (bar)
Re	Reynold's number
Sc	Schmidt's number
v	Velocity (m/s)
We	Weber's number
y	Mole fraction

Greek symbols

ε	Void fraction
ρ	Density (kg/m^3)
μ	Viscosity ($kg/(m{\cdot}s)$)
σ	Surface tension (N/m)

Subscripts

EFF	Effective
G	Gas
L	Liquid
N	Nominal
REL	Relative

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