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# General mass-transfer model for gas phase in structured packings



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#### 1. Introduction

Counter-current gas-liquid contactors – absorption and distillation columns – are the most widely used separation devices in the chemical industry. Humphrey (1995) estimated that only in US there are more than 40 000 of distillation towers which handle almost 90% of all separation processes. The columns are equipped either with trays or with random or structured packings. The latter are widely used in these contactors because of the low ratio of pressure drop to separation efficiency. Their mass-transfer efficiency is usually described by means of HETP or, in the case of the rate-based modeling, by the mass-transfer coefficients in the liquid and gas/vapor phase –  $k_{\rm L}$  and  $k_{\rm G}$  – and the effective interfacial area *a*.

Numerous models of the mass transfer in structured packings can be found in literature; their comprehensive overview and comments are given in Wang et al. (2005). Some of them are already incorporated into Aspen Plus or CHEMCAD engineering packages and they are routinely used in a column design. In our previous papers (Rejl et al., 2015; Valenz et al., 2011) we discussed the reliability of  $k_G$  and  $k_Ga$  values predicted by these models for absorption and distillation conditions and Mellapak 250.Y structured packing. Significant differences in predicted  $k_G$  and  $k_Ga$  values have been revealed; the predictions differ not only in the values of these parameters but also in their dependencies on gas/vapor flow rate. The power-law dependence of  $k_G \propto u_{Gs}^{\alpha}$  with  $\alpha$ 

ABSTRACT

Volumetric mass-transfer coefficient in the gas phase,  $k_Ga$ , for Mellapak 250.X and 250.Y structured packings has been measured by standard method of absorption of SO<sub>2</sub> from carrier gas to aqueous solution of NaOH in the i.d. 150.6 mm absorption column. Three carrier gases – He, N<sub>2</sub> and SF<sub>6</sub> – have been utilized providing wide range of Sc<sub>G</sub> from 0.49 to 2.32. The gas-phase mass-transfer coefficient,  $k_G$ , has been calculated from the experimental  $k_Ga$  data as the ratio of  $k_Ga$  and of the effective interfacial area, *a*, measured utilizing CO<sub>2</sub>/NaOH system. The experimental and literature  $k_G$  data for Mellapak 250.X, 250.Y, 350.Y and 500.Y have been correlated by dimensionless correlation  $Sh_G = 2.7 + 0.038 \cdot Re_{LG}^{0.88} \cdot Sc_G^{\beta}$  with  $\beta = 0.33 + 0.9 \cdot \exp(-0.6 \cdot Sc_G)$ . The correlation fits the data in the wide range of  $Re_{LG}$  and  $Sc_G$  covering the process conditions of common absorption and distillation columns.

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ranging from 0.6 to 1 is predicted according to these models. Almost all models (except of the correlation of Wang et al. (2016)) involve the dependence of  $k_G \propto D_{12,G}^{2/3}$  taken from the Chilton-Colburn analogy (Chilton and Colburn, 1934). The differences found in existing models led us to re-opening of the question about modeling of the gas-phase mass transfer in structured packings.

For the purpose of the gas-phase mass-transfer modeling the structured packing is usually described as a bundle of inclined channels of hydraulic diameter  $d_h$ . The modeling of the mass transfer in the packing channel is then based on the correlations for  $k_G$  in a wetted wall column.

The turbulent character of the gas flow in the packing channel is tacitly assumed due to frequent direction changes in the gas flow.

The gas-phase mass-transfer characteristics of the wetted wall column are usually published in a dimensionless form as a dependence of gas phase Sherwood number,  $Sh_G$ , on system properties characterized by Schmidt number,  $Sc_G$ , and the hydrodynamic conditions characterized by Reynolds number,  $Re_G$ .<sup>1</sup>

The simplest and often utilized relation for  $\mathsf{Sh}_\mathsf{G}$  is a simple power-law form

$$Sh_{\rm G} = A \cdot Re^{\alpha}_{\rm LG}Sc^{\beta}_{\rm G} \tag{1}$$

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<sup>&</sup>lt;sup>1</sup> Similarly the heat-transfer characteristics are correlated as a dependence of Nusselt number on Prandtl and Reynolds numbers.

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#### Nomenclature

List of symbols

- A (-) Correlation parameter independent on Sc and Re
- A<sub>Sc</sub> (-) Correlation parameter dependent on Sc<sub>G</sub>
- $a \, (m^2/m^3)$  Effective interfacial area of the packing
- $a_g$  (m<sup>2</sup>/m<sup>3</sup>) Geometrical area of the packing
- B (m/h) Liquid superficial velocity
- *b* (m) Length of the channel base
- c (mol/m<sup>3</sup>) Molar concentration
- $D_{12}$  (m<sup>2</sup>/s) Binary diffusion coefficient
- *d*<sub>eq</sub> (m) Equivalent diameter of the packing
- $d_{\rm h}$  (m) Hydraulic diameter of the packing channel
- g (m/s<sup>2</sup>) Gravitational acceleration,  $g = 9.81 \text{ m/s}^2$
- *h* (m) Height of the packing channel
- $h_{\rm L}$  (m<sup>3</sup>/m<sup>3</sup>) Liquid hold-up
- $k_{\rm G}$  (m/s) Mass-transfer coefficient in the gas phase
- $k_{Ga}$  (s<sup>-1</sup>) Volumetric mass-transfer coefficient in the gas phase
- $l_h$  (m) Characteristics length of the packing channel
- o (m) Channel perimeter
- p (Pa) Pressure
- S (m<sup>2</sup>) Channel cross-section
- s (m) Length of the channel side
- u (m/s) Velocity
- x,y,z (m) Spatial coordinates in Eq. (15)
- y<sub>i</sub> (–) Molar fraction of component i
- Greek symbols
- $\alpha$  (-) Correlation parameter, power at Re
- $\beta$  (–) Correlation parameter, power at Sc (Pr)
- $\varepsilon$  (m<sup>3</sup>/m<sup>3</sup>) Packing void fraction
- $\theta$  Channel inclination angle
- $\vartheta$  Liquid effective angle of channel
- ξ Gas-liquid interaction parameter (DELFT model)
- $\mu$  (Pas) Dynamic viscosity
- $\rho$  (kg/m<sup>3</sup>) Density

Dimensionless numbers

- Mi Mixing number
- Nu Nusselt number
- Pr Prandtl number
- Re Reynolds number
- Sc Schmidt number
- Sh Sherwood number

Subscripts, superscripts

Е	Effective	(velocity)
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- G Gas phase
- h Hydraulic
- L Liquid phase
- LG Gas phase related to liquid phase (Reynolds number) lam Laminar turb Turbulent

with parameters A,  $\alpha$  and  $\beta$  evaluated on the basis of experimental data or taken from the heat-transfer correlations utilizing the analogy of heat and mass transfer. This form of the Sh<sub>G</sub> correlation with the parameters A = 0.023,  $\alpha$  = 0.83 and  $\beta$  = 0.44 was published by Gilliland and Sherwood (1934) who evaporated several liquids into the turbulent

air stream in the wetted wall column. Their data had been taken by Chilton and Colburn (1934) for simultaneous correlation of heat and mass-transfer coefficients resulting in famous analogy of momentum, heat and mass-transfer with values of A = 0.029,  $\alpha$  = 0.78 and  $\beta$  = 1/3. The value of exponent  $\beta = 1/3$  was later confirmed by Linton and Sherwood (1950) who measured the mass-transfer characteristics of turbulent liquids with high values of Sc. Johnstone and Pigford (1942) performed a series of distillation experiments in the wetted wall column and reported good agreement of evaluated distillation Sh<sub>G</sub> values with those calculated according to correlations of Gilliland and Sherwood and Chilton and Colburn. Years later Crause and Nieuwoudt (1999) performed a series of evaporation experiments in a short wetted wall column representing the channel of a structured packing and published the values of  $\alpha = 1$  and  $\beta = 0.5-0.55$ . Recently, we have performed absorption experiments in a wetted wall column with three carrier gases having significantly different values of Sc<sub>G</sub> (the same ones were used in this work) and found values of  $\alpha = 0.9$  and  $\beta = 0.6$  (Haidl et al., 2016). The usability of our correlation was confirmed also for distillation conditions proving the concept of absorption and distillation process analogy (Rejl et al., 2016).

In our previous work (Rejl et al., 2015) we performed series of absorption experiments on Mellapak 250.Y, 350.Y, 452.Y and 500.Y utilizing air/SO<sub>2</sub>/NaOH<sub>(aq.)</sub> system for k<sub>G</sub>*a* measurement and air/CO<sub>2</sub>/NaOH<sub>(aq.)</sub> for *a* measurement. The k<sub>G</sub> = k<sub>G</sub>*a*/*a* values for all packings were correlated in a dimensionless form

$$\left(\frac{k_{\rm G}d_{\rm eq}}{D_{12,\rm G}}\right) = 0.409 \left(\frac{d_{\rm eq}u_{\rm G,s}\rho_{\rm G}}{\mu_{\rm G}}\right)^{0.622} \left(\frac{d_{\rm eq}u_{\rm L,s}\rho_{\rm L}}{\mu_{\rm L}}\right)^{0.0592} \tag{2}$$

with  $d_{eq} = 4\varepsilon/a_g$ . The results indicate the value of  $\alpha = 0.622$  which is in agreement with findings of Wang et al. (2016) made for various structured packings using the same absorption systems. It is, however, significantly lower than the values used in the wetted wall column correlations and in other models (from 0.75 according to Billet and Schultes (1999) to 1 used in the correlation of Hanley and Chen (2012)).

This paper is focused on the experimental study of  $k_G$  dependence on physical properties of gas, i.e.  $Sc_G$ , in the range of common absorption and distillation systems. For this purpose, the absorption of  $SO_2$ from three different carrier gases – He,  $N_2$  and  $SF_6$  – into aqueous solution of NaOH has been utilized for the  $k_Ga$  measurements providing the range of  $Sc_G$  from 0.49 to 2.32. The  $k_G$  data measured in this work together with the literature data are correlated resulting in one dimensionless correlation.

### 2. Experimental

The  $k_Ga$  measurements have been performed with a proven method of absorption of diluted SO<sub>2</sub> from carrier gas (N<sub>2</sub>, He, SF<sub>6</sub>) into the aqueous solution of NaOH (Rejl et al., 2009). In this system, the mass-transfer resistance is concentrated only in the gas phase due to the instantaneous reaction of SO<sub>2</sub> with sodium hydroxide at the gas–liquid interface. Assuming plug flow of the gas phase,  $k_Ga$  can be calculated from the molar fraction of SO<sub>2</sub> in the gas sampled at the bottom and at the top of the packed bed according to Eq. (3).

$$k_{\rm G}a = \frac{u_{\rm Gs}}{h_{\rm bed}} \ln \frac{y_{\rm SO2,bottom}}{y_{\rm SO2,top}} \tag{3}$$

The experiments have been carried out in a 150.6 mm i.d. absorption column made of transparent PVC-U packed with Mellapak 250.X (M250X) or Mellapak 250.Y (M250Y) packing. The packed bed consisted of six elements of packing for measurements using N<sub>2</sub> and He as a carries gas (total bed height of 1.26 m with M250Y or 1.32 m with M250X) and of two packing elements for measurement using SF<sub>6</sub> (total bed height of 0.42 m with M250Y). The individual packing elements were rotated around vertical axes by 90° with respect to each other.

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