

# A Novel Approach to the Modelling of Transport Phenomena in Random Packings

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The separation characteristics of absorption columns are commonly predicted by simplified stage models (equilibrium or rate-based). Such models employ several parameters which have to be determined by experiments. This represents a major weakness of stage models, as, generally, these parameters are not directly transferable to different chemical systems or geometries. An alternative approach to the modelling of transport phenomena in fluid separation processes is based on so-called hydrodynamic analogies. Hereby, the actual flow in a packing is replaced by a combination of different, simplified flow patterns depending on the specific geometry of column internals. The transport of heat and mass in such flows can be described by rigorous conservation equations. As a consequence, the use of most of the above mentioned experimental parameters can be avoided. In this work, the hydrodynamic analogy approach is applied to random packings. To validate the model, the separation efficiency of Pall rings for the reactive absorption of CO<sub>2</sub> into aqueous sodium hydroxide solution was investigated experimentally in an industrial-scale column. In this paper, the modelling method is described in detail and the validation of the simulation results is presented.

## 1. Introduction

The removal of carbon dioxide (CO<sub>2</sub>) from industrial flue gases is an important operation, which is mostly performed by the reactive absorption of CO<sub>2</sub> into aqueous solutions of sodium hydroxide or other bases. Technical absorption processes are commonly realised in columns equipped with packed beds containing either structured or random packings.

Mass and heat transfer in absorption columns are usually described by using simplified stage models (equilibrium or rate-based). Equilibrium models consider the column internals as a sequence of stages where the incoming and leaving streams are at equilibrium. Since this consideration neglects all packing specific properties, these models use lump parameters like the "height equivalent to a theoretical plate" (HETP) for the column design. The HETP values must be found experimentally. Rate-based models follow another approach in which the actual mass and heat transfer rates are considered. Depending on which theory is applied within the rate-based modelling (film model, penetration model, etc.) the corresponding parameter (film thickness, residence time, etc.) must be estimated by correlations based on experiments. The rate-based approach is state of the art for modelling CO<sub>2</sub> absorption processes (Gabrielsen et al., 2007; Harbou et al., 2014; Hüser and Kenig, 2015). The strong dependence on parameter correlations appears to be the major weakness of stage models, since these correlations are often fraught with uncertainties and require extensive experimental effort to be transferable to different chemical systems or geometries.

In order to reduce the dependence on correlations, the concept of hydrodynamic analogies was proposed. The main idea is to represent the actual process fluid dynamics by a simplified flow pattern or a combination of different simplified flow patterns like film, jet or droplet flow. One of the first models using this approach was developed for distillation processes in structured packings by Shilkin and Kenig (2005). Later it was used by Brinkmann et al. (2013) to describe reactive CO<sub>2</sub> absorption processes, also in structured packings. In the present work, the hydrodynamic analogy approach is applied to the modelling of reactive absorption in columns equipped with 50 mm Pall rings.

## 2. Hydrodynamic analogy approach for random packings

Observation of the fluid flow inside the investigated column revealed basically three different types of flow pattern. These are film flow over the packing surface as well as jet and droplet flow in the free space between the packing elements. Therefore, the liquid flow patterns that are modelled are film, jet and droplet flow. The column is represented as a combination of channels whose diameter equals the hydraulic diameter of the packing given by Equation 1:

$$d_H = 4 \frac{\varepsilon}{a} \quad (1)$$

The film is considered to flow downwards along the inside channel wall. Furthermore, jets and drops are assumed to fall through the channel centre. The gas is modelled to flow counter-currently to the liquid through the channels. The total number of channels is determined using Equation (2), so that the cross-sectional area of all channels equals the cross-sectional area of the actual packed column that is free of packing elements.

$$N_C = \varepsilon \frac{d_{pac}^2}{d_H^2} \quad (2)$$

Since the packing is oriented randomly inside the column, the angle between the longitudinal axis and the horizontal is assumed to be  $45^\circ$ , which is the average of all possible angles. The fluid dynamics in the gas phase and in the film and jet flow can be described rigorously by the momentum equation in radial coordinates (Levič, 1962):

$$\frac{1}{r} \frac{\partial}{\partial r} \left( r \mu \frac{\partial u}{\partial r} \right) - \frac{\partial P}{\partial x} + \rho g \sin(\alpha) = 0 \quad (3)$$

The pressure gradient in radial direction is neglected. When applied to the liquid phase, the pressure gradient in axial direction is neglected too. In the gas phase the gravitation term is neglected. The drops are considered as discrete objects, and their sedimentation velocity is determined as proposed by Maćkowiak (2010), based on the force equilibrium of a falling drop.

Heat and mass transfer are described using the respective conservation equations for the species mass and heat conservation (Bird et al., 2017):

$$u(r) \frac{\partial C_k}{\partial x} = \frac{1}{r} \frac{\partial}{\partial r} \left( r D_k \frac{\partial C_k}{\partial r} \right) \quad k = 1, \dots, N_k \quad (4)$$

$$u(r) \frac{\partial T}{\partial x} = \frac{1}{r} \frac{\partial}{\partial r} \left( r \kappa \frac{\partial T}{\partial r} \right) \quad (5)$$

In both equations, radial convection and axial diffusion / conduction are neglected.

The velocity profiles for the film, jet and gas flow are analytically solvable, whereas the mass and heat transfer equations are solved numerically. Since it is assumed that there is no influence of mass and heat transfer on the flow, the momentum transfer equations are solved only once in the beginning of the calculations. Figure 1 shows an example of velocity profiles for the case of jet flow (1a) and of concentration profiles for the film flow (1b).

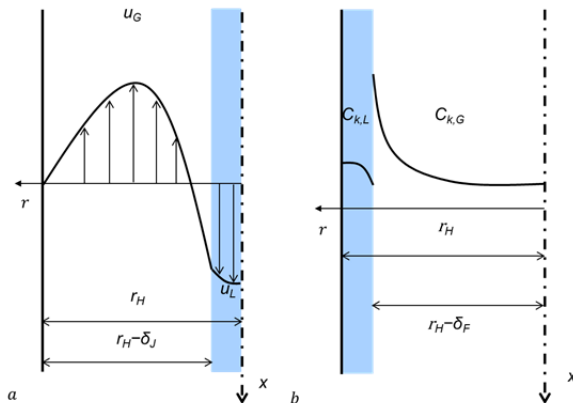


Figure 1: Schematic profiles of velocity profiles for jet flow (a) and concentration profiles for film flow (b)

The interaction between the gas and liquid phases occurs at the gas-liquid interface inside the channels. To account for the interaction of the different flow patterns and flow disturbances, e.g. through redirection of the

flow or transformations of films into jets or drops and vice versa, the three flow patterns are treated simultaneously in parallel. The phases are assumed to interact at mixing points, after passing a certain distance. These distances depend on specific geometrical parameters of the packing.

## 2.1 Mixing points in gas flow

The gas flow is assumed to pass a Pall ring through the inside along the longitudinal axis. When the gas exits the Pall ring, it is redirected interacting with gas streams coming from the surrounding packing elements (Figure 2a). At this point, the gas phase is assumed to be mixed. In the model, mixing results in some constant values obtained by averaging the radial concentration/temperature profiles (integral mean values). This is realised for the three gas streams which correspond to all three liquid flow patterns. To account for the mass and heat exchange between the individual gas streams, the integral mean concentrations/temperatures are weighted according to the volumetric ratios of the three patterns.

## 2.2 Mixing points in liquid flow

The whole liquid flow is assumed to be spread into three patterns with a respective length of undisturbed flow. For the film flow, the length of undisturbed flow is determined by the distance between the entrance of the liquid to a packing element and a detachment point. The film is assumed to flow along the circumference of a packing element, and it detaches either at the half of the circumference or when it reaches the edge of the circumferential part of the packing element. The second case is shown in Figure 2b. Which of these cases occurs, depends on the angle of the packing element to the horizontal.

It is assumed that the length of undisturbed flow for the jet and droplet flow are the same and that they are formed at the detachment points of the film and fall until another wall is reached (see Figure 2c).

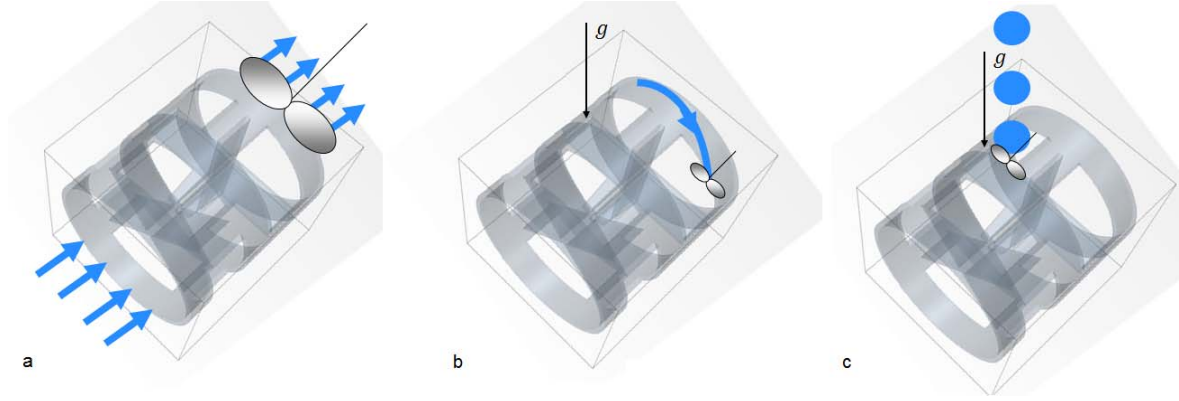


Figure 2: Visualization of the lengths of undisturbed flow for the gas (a), film (b) and jet and drop (c)

The length of the undisturbed flow of jets and drops is estimated based on the probability of a drop or jet to hit a wall. This probability is determined using the form factor  $\varphi$ , which is defined as the ratio of the perforated wall area to the total non-perforated wall area. It is assumed that this ratio just equals the probability of a drop or jet to pass one packing element. Thus, the probability that a jet or drop passes  $N_{PE}$  packing elements is:

$$E_{pass} = \varphi^{N_{PE}} \quad (5)$$

The probabilities to hit or to pass a wall sum up to one. Thus, the probability to hit a wall after  $N_{PE}$  packing elements is:

$$E_{hit} = 1 - E_{pass} = 1 - \varphi^{N_{PE}} \quad (6)$$

Rearranging and integrating Equation (6) yields the average number of passed packing elements:

$$N_{PE,ave} = \int_0^1 \frac{\ln(1-E_{hit})}{\ln(\varphi)} dE_{hit} = \frac{-1}{\ln(\varphi)} \quad (7)$$

The jets and drops do not fall through packing elements only, but also through empty spaces between packing elements. Therefore, the void fraction between neighbouring packing elements  $B$  is needed to determine the length of undisturbed flow. This void fraction is only a part of the overall void fraction  $\varepsilon$  of the packing. When  $N_{PE,ave}$  is multiplied with the length of a packing element and divided by  $B$ , it yields the length of undisturbed flow for the jet and drop modelling:

$$z_{D/J} = N_{PE,ave} \frac{l_{PE}}{B} \quad (8)$$

The length of undisturbed flow for the drops ( $z_D$ ) and jets ( $z_J$ ) is several times longer than for the film ( $z_F$ ) for the Pall rings. Thus, the film is mixed several times along the mixing length of the jet / drop. At this point, all three flow patterns are mixed together. A schematic of the model is given in Figure 3. The large mixing symbols represent the mixing of all three flow patterns, whereas the small mixing symbols represent the sole film mixing.

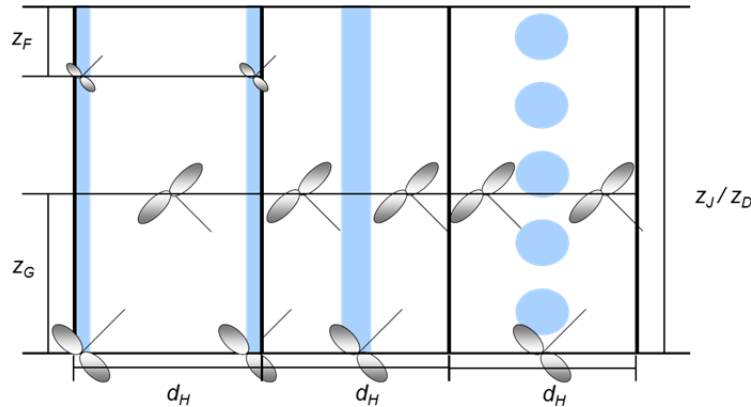


Figure 3: Schematic of the parallel film, jet and droplet flow through the channels and the corresponding lengths of undisturbed flow  $z_G$ ,  $z_F$ ,  $z_J$ ,  $z_D$ .

### 3. Experimental

The experimental investigations were carried out in a DN600 industrial scale column with a packing height of 2.9 meter. The random packing consisted of 50 mm Pall rings. The absorption of  $\text{CO}_2$  from air into aqueous sodium hydroxide (NaOH) solution was investigated, as a standard test system for the determination of mass transfer characteristics (ENVIMAC Engineering GmbH, 2012). The liquid load was varied, while the gas load was kept constant at  $F = 0.55 \text{ Pa}^{1/2}$ . The  $\text{CO}_2$  inlet and outlet concentrations in the gas phase and the inlet and outlet temperatures were measured. In Figure 4, the absorption rates are plotted against the corresponding liquid loads.

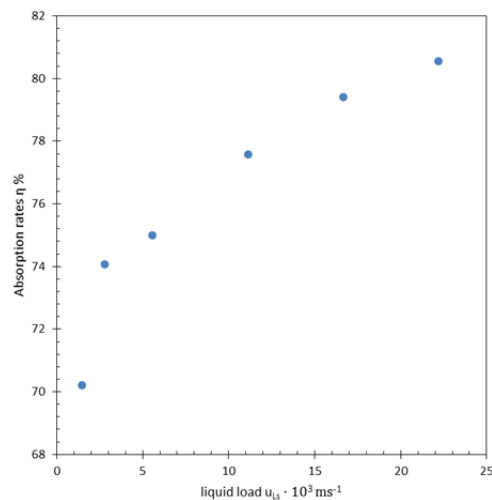


Figure 4: Measured absorption rates plotted against the liquid load at a constant gas load ( $F = 0.55 \text{ Pa}^{1/2}$ )

It can be seen that for higher liquid loads, the absorption rates increase. The general trend shows a decrease in the slope of the absorption rate with higher liquid loads. The absorption rate of the second operating point

appears to be too large because it does not fit the general trend. The last two operating points show that a further increase in the liquid load has only a slight effect on the absorption rate.

#### 4. Simulation

The experimental data were used to perform the very first validation of the new model. The simulations were carried out under isothermal conditions, because the experiments showed no significant change in temperature from the inlet to the outlet ( $< 1$  K). The reaction of  $\text{CO}_2$  with NaOH was considered by adding a reaction term to the  $\text{CO}_2$  transport equation (4) using the kinetics proposed by Kucka et al. (2002). Since NaOH was in significant excess, the transport of the hydroxyl ions was not considered to save computational effort. The liquid flow was assumed to spread equally into films jets and drops. The comparison of experimental and simulated absorption rates is given in the parity plot in Figure 5.

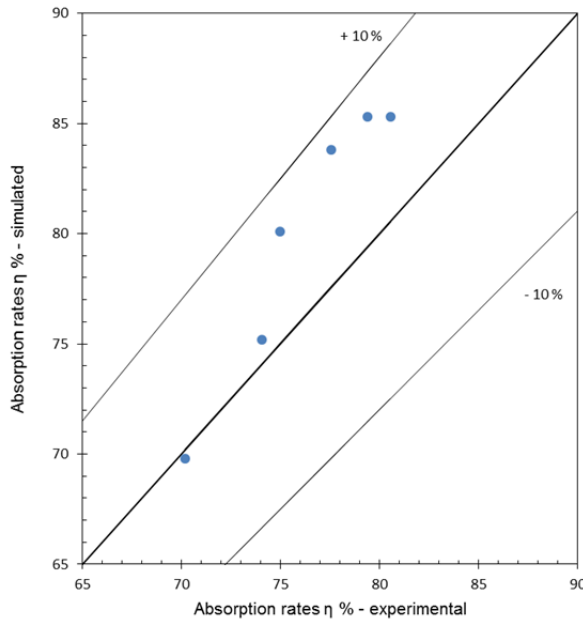


Figure 5: Parity plot of the experimental and simulated absorption rates

It can be seen that the simulated absorption rates generally fit the experimental rates fairly well, with ten percent deviation. The first two operating points are matched even with two percent deviation. For larger absorption rates, the simulation overshoots the experiments, but follows the same trend. Also the last two points show good agreement in the trend, as the experimental absorption rates differ less than two percent, while simulated absorption rates do not change.

#### 5. Conclusions

A new approach to the modelling of transport phenomena in columns equipped with random packings is proposed. This approach is based on hydrodynamic analogies between the actual fluid dynamics in the process and simplified flow pattern. This allows rigorous modelling of heat and mass transfer. The model was applied to a random packing containing 50 mm Pall rings. A multi-channel model was chosen covering the liquid flow as a combination of films, jets and droplets, and the counter-current gas flow. After a certain length, each flow is mixed either with itself or with the other patterns. The lengths of undisturbed flow depend on geometrical parameters of the packing.

The velocity profiles of film, jet and gas flow are determined using the momentum equation in radial coordinates, while the drop sedimentation velocity is estimated based on the force equilibrium of a falling drop. Heat and mass transfer are described by the convective diffusion and heat conduction equations for each flow pattern and phase. This allows concentration and temperature profiles along the column to be determined.

The first model validation was performed using experimental data from an industrial-scale absorption column equipped with 50 mm Pall rings. In the experiments, the absorption of  $\text{CO}_2$  into aqueous sodium hydroxide solution was investigated for varying liquid loads. Since the experiments showed no significant change in temperature, the simulations were performed under isothermal conditions. The comparison of the

experimental and simulated absorption rates showed a good agreement, with a maximum deviation of ten percent. In future work, the desorption behaviour of CO<sub>2</sub> will be investigated, and further types of random packings will be considered.

## Nomenclature

### Latin letters

<i>a</i>	Specific surface area (m <sup>2</sup> / m <sup>3</sup> )	$\delta$	Width of liquid flow pattern (m)
<i>B</i>	Void fraction between packing elements (m <sup>3</sup> / m <sup>3</sup> )	$\varepsilon$	Void fraction (m <sup>3</sup> / m <sup>3</sup> )
<i>C</i>	Concentration (mol/m <sup>3</sup> )	$\eta$	Absorption rate (%)
<i>D</i>	Diffusion coefficient (m <sup>2</sup> /s)	$\kappa$	Thermal diffusivity (m <sup>2</sup> /s)
<i>d</i>	Diameter (m)	$\mu$	Viscosity (Pa s)
<i>E<sub>hit</sub></i>	Probability of jets/drops hitting a packing element	$\rho$	Density (kg/m <sup>3</sup> )
<i>E<sub>pass</sub></i>	Probability of jets/drops passing a packing element	$\varphi$	Form factor (m <sup>2</sup> /m <sup>2</sup> )
<i>F</i>	Gas capacity factor (Pa <sup>1/2</sup> )		
<i>g</i>	Gravity (m/s <sup>2</sup> )		
<i>l</i>	Characteristic length (m)		
<i>N</i>	Number of elements		
<i>P</i>	Pressure (Pa)		
<i>r</i>	Radius (m)		
<i>T</i>	Temperature (K)		
<i>u</i>	Velocity (m/s)		
<i>V</i>	Volume (m <sup>3</sup> )		
<i>x</i>	Axial coordinate (m)		
<i>z</i>	Length of undisturbed mixing (m)		

### Indices

<i>ave</i>	Average
<i>D</i>	Drops
<i>F</i>	Film
<i>G</i>	Gas phase
<i>H</i>	Hydraulic
<i>J</i>	Jet
<i>k</i>	Component <i>k</i>
<i>L</i>	Liquid phase
<i>pac</i>	Packing
<i>PE</i>	Packing element
<i>s</i>	Superficial

### Greek letters

$\alpha$  Film flow angle (°)

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