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Modelling and simulation of a packed sulphur dioxide absorption unit using the hydrodynamic analogy approach

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The design of reactive gas/vapour-liquid separation units is often based on conventional modelling methods employing rate-based stage concepts. The parameters of such models (e.g., mass transfer coefficients) are determined experimentally, and, most often, significant experimental effort is necessary when new processes are explored. In a novel approach based on hydrodynamic analogies and application of rigorous partial differential transport equations, mass transfer coefficients and hence expensive experimental efforts are not required.
This approach has already been tested for non-reactive distillation and absorption processes. The main focus of this work is its application to reactive absorption. The absorption of sulphur dioxide in aqueous solution of sodium hydroxide is chosen as a test system. The model is successfully validated using experimental data obtained with a lab-scale absorption column.

1. Introduction

Chemical absorption processes are widely used for the purification of industrial gas streams from undesired components, e.g., NO_X or SO_2. Further, they are expected to play a central role in the development of CO_2 capture and storage technologies. In many cases, absorption is performed in columns operated counter-currently and filled with random or structured packings.
Adequate description of chemical absorption usually requires rigorous modelling methods capable of describing mass transfer and chemical reaction rates properly. In the last decades, rate-based models based on an extension of the well-known film theory have become popular (see, e.g., Kenig et al., 2003). However, the mass-transfer correlations included in such models have to be obtained from expensive experimental investigations. Recently, a new modelling approach for structured packed columns has

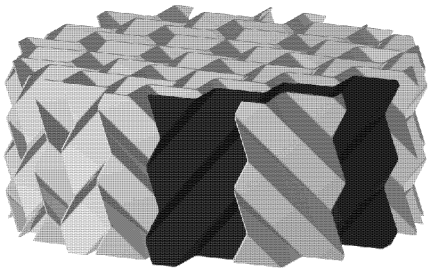

Figure 1. Sketch of a corrugated sheet packing layer, a single sheet highlighted

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been suggested by Shilkin and Kenig (2005). This approach is based on hydrodynamic analogies (HA) between real complex flows in structured packings and more simple (usually film-like) flow patterns in so-called corrugated-sheet packings (see Figure 1). Unlike conventional process models, in the HA model, concentration profiles are determined based on rigorous partial differential transport equations and, therefore, experimental determination of mass transfer correlations is not required.

The HA approach has been validated for different multicomponent distillation processes (Shilkin and Kenig, 2005; Shilkin et al., 2006) and for a reactive stripping process (Brinkmann et al., 2008b). The simulated concentration profiles showed a good agreement with the experimental data, especially at low vapour and gas loads.

This permits an extension of the HA approach towards chemical absorption processes. In our earlier study, the model was applied for the description of absorption units with gas loads changing from moderate values up to the flooding limit, because absorption processes are often operated under such conditions (Brinkmann et al., 2008a). Using a non-reactive test system (absorption of ammonia from air into water), it was found that an adjustment of the gas-phase turbulence description was necessary.

The objective of this work is the application of the HA approach to a particular type of chemical reaction, namely instantaneous reactions at the gas-liquid interface.

2. Modelling

The physical model based on the HA approach for the description of the gas-liquid flow in corrugated sheet packings consists of a bundle of inclined cylindrical channels with identical cross-section. The inner surface of the channels is wetted by a liquid flowing downward, whereas the rest of the volume is occupied by a counter-current gas flow (Figure 2).

The channel inclination angle α is equal to the angle of the liquid-film flow along the corrugated sheets. The number of channels and their radius R_H are determined based on the specific packing surface and corrugation geometry. The gas and liquid phases are assumed to be ideally mixed at regular intervals (z_L and z_G). This is necessary in order to take real mixing in the packing into account. The determination of geometric model parameters is described in detail in previous works (Shilkin and Kenig, 2005; Shilkin et al., 2006).

For the estimation of the film thickness and the velocity profiles, a fully developed two-phase flow in a single channel is considered. Under these conditions, the velocity profiles and film thickness do not depend on the axial coordinate.

The liquid flow in structured packed columns can be assumed laminar, whereas turbulence must be considered in the gas phase under

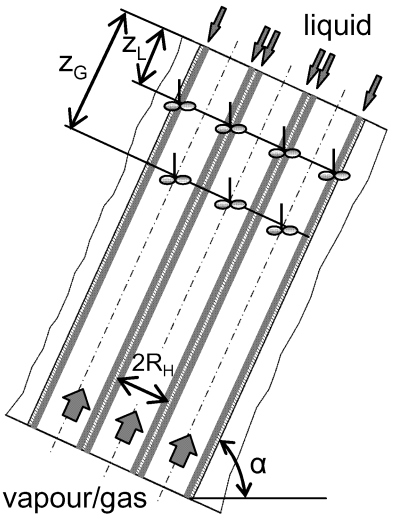

Figure 2. Physical model of the HA for the gas-liquid flow in columns with corrugated sheet packings

certain conditions (see Shilkin et al., 2006; Brinkmann et al., 2008a). Gas and liquid flow are described by a system of Navier-Stokes equations in the film-flow approximation, whereas the Boussinesq approach is adopted for the description of the gas-phase turbulence. Mass and heat transfer phenomena in the liquid and gas phase are described by the partial differential equations including convection, diffusion and heat conduction phenomena. Instantaneous reactions occurring at the gas-liquid interface are implemented into the model using the interfacial boundary conditions, whereas the mass flux continuity incorporates source terms due to reaction.

3. Experimental

Experimental investigations of a structured packed lab-scale absorption column have been performed at BASF SE. The experimental setup is described by Hoffmann et al. (2007) for the measurement of absorption rates in random packings. The experimental conditions are in line with the recommendations given by Hoffmann et al. (2007) to ensure good reproducibility of mass transfer measurements with absorption systems.

Measurements have been performed at different liquid loads (w_L = 10, 20 and 30 $m^3\ m^{-2}\ h^{-1}$) and at F-factors from 1 $Pa^{0.5}$ up to the flooding limit. All measured variables used for this work are shown in Figure 3. The column was filled with a standard corrugated sheet packing with a specific surface of 250 $m^3\ m^{-2}$.

For the model validation, not only a reactive system but also a physical absorption system is taken into account, since real absorption systems often contain both reactive and non-reactive absorbents.

The absorption of ammonia from an ammonia/air mixture into water is selected as *a physical test system*. The mass transfer resistance is distributed between both phases (Billet, 1995); therefore, this system appears appropriate for testing both liquid-phase and gas-phase description in the HA-model.

As a *reactive test system*, the removal of sulphur dioxide from air into an aqueous solution of sodium hydroxide is chosen. The chemical absorption of sulphur dioxide in alkaline solutions can be described by the following two reactions (Hikita et al., 1977):

$$SO_2 + OH^- \rightleftarrows HSO_3^- \quad (1)$$

$$HSO_3^- \rightleftarrows SO_3^{2-} + H_2O \quad (2)$$

However, in strong sodium hydroxide solutions, both reactions are very fast and may together be regarded as an overall instantaneous, irreversible reaction (Schultes, 1998; Hölemann and Górak, 2006) which occurs solely at the gas-liquid interface:

$$SO_2 + 2\,OH^- \rightarrow SO_3^{2-} + H_2O \quad (3)$$

Further, this system is largely dominated by the gas-phase mass transfer.

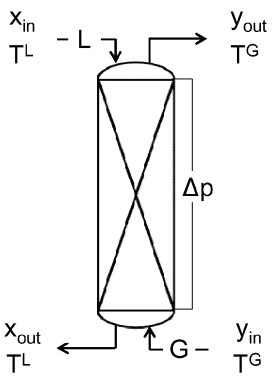

Figure 3. Measured variables in the absorption column

4. Simulation results

As the absorption column is kept isothermal in the experiments and the amount of absorbent in the inlet streams is below 1 mole-% in all experiments, temperature effects are neglected in the simulations.

A parameter required for the proper description of the gas-phase turbulence was estimated in our previous work using the ammonia system as test system (see Brinkmann et al., 2008a). This parameter is assumed to be related only to the packing geometry and valid for both systems handled in this study.

The HA-model is verified using the data of all available experiments. Simulated column profiles at different F-factors together with the experimental data are shown in Figure 4. The calculated outlet concentrations of both absorbents in the gas phase agree well with experimental values for all gas and liquid loads.

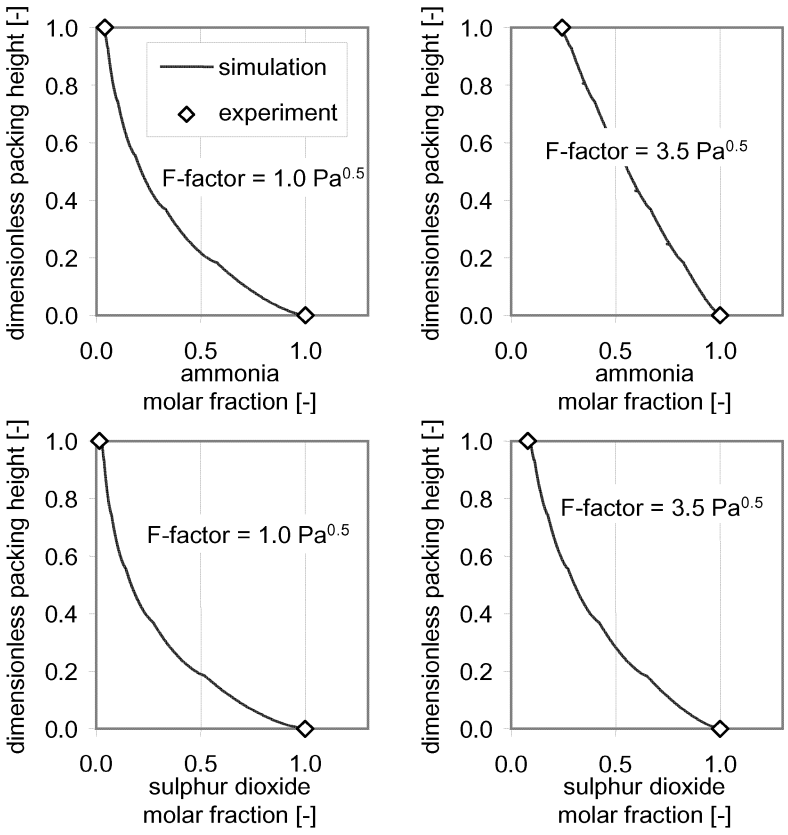

Figure 4. Simulated gas concentration profiles and experimental data normalised against the inlet concentration at different gas loads and $w_L = 20\ m^3\ m^{-2}\ h^{-1}$

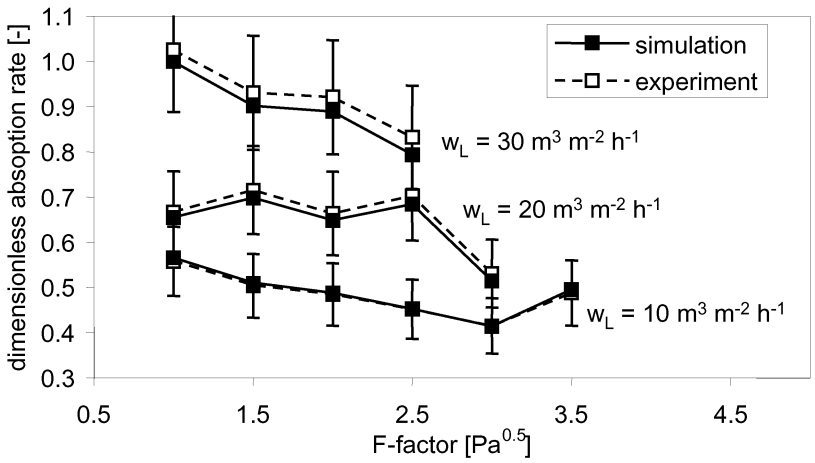

Figure 5. Absorption rate of sulphur dioxide normalised against its simulated maximum value at different gas and liquid loads

The column profiles of ammonia appear less curved than those of sulphur dioxide, especially at high F-factors. This can be explained by the relation between the mass transfer resistances discussed in Section 3, namely, the gas-phase mass-transfer resistance of ammonia at high gas loads is rather low.
Further, the absorption rate of sulphur dioxide under all investigated load conditions is shown in Figure 5. A good agreement between calculated and experimental values is visible for all conditions including high gas and liquid loads.

5. Conclusions

A novel modelling approach based on hydrodynamic analogies (HA) between real and simplified flow conditions has been proposed for the description of gas/vapour-liquid separation units filled with structured packings. In this work, the HA approach is applied for the simulation of absorption processes.
The focus of the study was to extend the model for the description of reactive absorption systems with instantaneous reactions occurring at the gas-liquid interface. It is found that such reactions can be implemented into the model using the interfacial boundary conditions.
For the model validation, a non-reactive and a reactive absorption system are considered. Experimental investigations are carried out in a lab-scale column filled with a standard-type structured packing. Measurements are performed for a broad range of liquid and gas loads.
Using the experimental data, the HA-model is successfully validated. The concentration profiles and absorption rates obtained from simulations are in a very good agreement with the available experimental data of different test systems. Among others, these results confirm the assumption that the turbulence description in the HA-model depends only on the geometry and not on the chemical system.

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