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SEPARATION PERFORMANCE OF STRUCTURED PACKED COLUMNS: A COMPARISON OF TWO MODELLING APPROACHES

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INTRODUCTION

Structured packings of the corrugated sheet type have become increasingly popular for the application in distillation and absorption columns, due to their very good performance characteristics. Commonly, at sub- and near-atmospheric pressures, structured packings outperform random packings and trays in capacity and/or separation efficiency (see, e.g., Kurtz et al., 1991; Brunazzi, E. and Paglianti, A., 1997). However, these packings are quite expensive, and hence, accurate equipment design is particularly important for the minimisation of investment and operating costs. Such design cannot be realised without sound and predictive process models.

It is widely known, that structured packing performance characteristics depend heavily on the corrugation geometry. Notwithstanding great efforts invested over the last 40 years in studies and modelling of the hydrodynamics and mass transfer in structured packings, determination of the optimal corrugation geometry for a particular separation task is still accomplished mostly by trial-and-error and remains rather art and know-how than rigorous science (see, e.g., Irwin et al., 2004). This explains the large number of packing modifications and the lack of truly predictive models for their performance.

TRADITIONAL MODEL

A number of modelling approaches for the prediction of the structured packed column hydraulics and mass transfer are available in the literature. Most advanced models consider directly actual rates of heat and mass transfer. By far the most frequently used model for the description of the mass transfer in separation columns is based on the *two-film theory* put forward by Lewis & Whitman (1924).

When applying the two-film theory to the modelling of a separation column, the total packing height is subdivided into a sequence of stages of certain height (see Figure 1). At each stage, all the resistance to mass transfer is localised in thin films adjacent to the gas-liquid interface. Mass transfer occurs in these films solely by steady-state molecular diffusion in the direction normal to the interface. Outside the films, in the bulk fluid phases, the ideal mixing is usually assumed, i.e. no temperature and composition

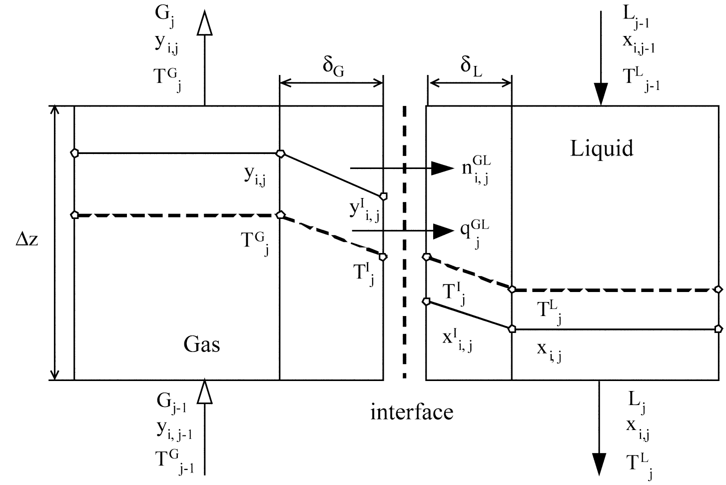

Figure 1. Schematic illustration of the two-film model

gradients exist. The hydrodynamic and mass transfer effects specific for a particular packing geometry are taken into account by applying correlations for the mass transfer coefficients, k_L and k_G. The latter are used to estimate the film thicknesses:

$$\delta_L = \frac{D_L}{k_L}, \quad \delta_G = \frac{D_G}{k_G} \tag{1}$$

Usually, the correlations for the mass transfer coefficients are determined empirically. These coefficients depend on the process hydrodynamic and physicochemical characteristics. Besides, the influence of the corrugation geometry is considered, either explicitly (see, e.g., Olujic et al., 2000) or using the packing specific constants (see, e.g., Zogg, 1973). A detailed discussion of advantages and disadvantages of the two-film theory is given in Shilkin and Kenig (2005).

HYDRODYNAMIC ANALOGY

The two-film model reduces the actual hydrodynamics to the concept illustrated in Figure 1 (stagnant films, ideally mixed bulks). The reason of such simplification is extremely complex two-phase fluid flow in real columns still further complicated due to interfacial heat and mass transfer. Under such conditions, exact localisation of the phase boundaries is hardly possible, and hence, a pure theoretical description of the separation columns by the fundamental differential equations governing momentum, energy and mass transport (i.e. Navier-Stokes, convection-diffusion and heat conduction equations) appears unfeasible in the near future.

However, in some cases it is possible to overcome this difficulty by application of the so-called *hydrodynamic analogy* (HA) approach (Kenig, 1997). This approach implies replacement of actual complex two-phase flow by an appropriate combination of geometrically simpler flow patterns. Along these lines, application of the Navier-Stokes, convection-diffusion and heat conduction differential equations becomes possible to describe the transport phenomena in an entire packed column. Recently, Shilkin and Kenig (2005) proposed a HA-based model to describe separation performance of structured packed columns. According to the HA approach, the reproduction of the real gas-liquid flow through the packing layers by a sequence of simplified flow patterns is accomplished in agreement with experimental observations of fluid flow, which can be summarised as follows:

1. gas flow takes place in channels built up by the counter course assemblage of the corrugated sheets into a packing layer (see Figure 2a);
2. there is an interaction between the gas flows in adjacent channels via the open channel side responsible for turbulence in the gas flow;
3. abrupt gas flow redirection at the column walls and at transition between the packing layers (side effects) results into periodic large-scale mixing of the gas phase;
4. liquid generally tends to flow in form of non-wavy films over the surface of corrugated sheets, strictly following the surface patterning;
5. the liquid flow is free of inertial effects and its direction corresponds to the minimal angle built by the packing surface and the vertical axis;

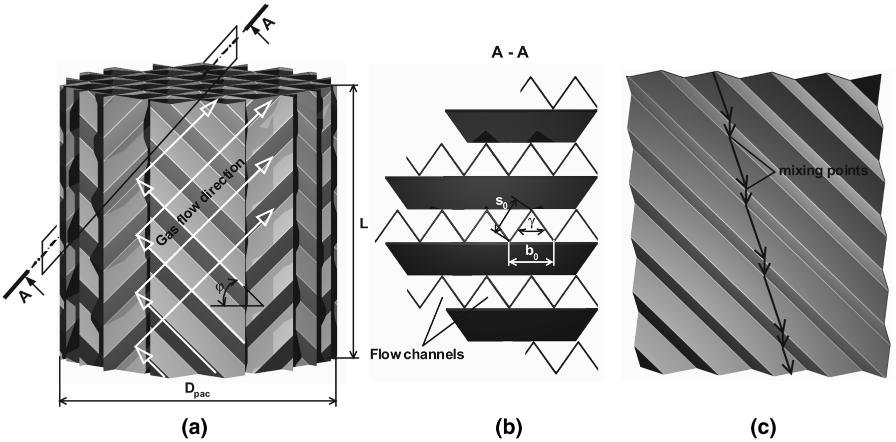

Figure 2. Schematic of the gas flow through a packing layer (a), sectional view illustrating geometry of the flow channels (b) and liquid flow path over the corrugated sheet surface (c)

6. abrupt liquid flow redirection and influence of the intersection points with neighbouring corrugated sheets causes mixing and lateral spreading of the liquid phase (see Figure 2c).

The HA-based physical model represented in Figure 3 comprises all the observed hydrodynamic effects. The actual gas flow in channels illustrated in Figure 2b is approximated by a similar flow in a bundle of parallel circular channels, their number and diameter depending on the structured packing geometric specific area and the corrugation geometry, respectively. The gas flow behaviour in the channels is assumed the same as in structured packing, ranging from laminar to turbulent flow regime. The liquid flows in form of laminar films by gravitation over the inner surface of channels. The channels are inclined at the minimal angle α to fulfil the condition 5. Furthermore, the ratio of wetted to total number of channels is the same as the ratio of effective (wetted) area to the geometric specific area of the packing. The large-scale periodic mixing of both phases due to abrupt flow redirection (conditions 3 and 6) is accounted for by the periodic ideal mixing in the inclined channels. The corresponding lengths of undisturbed fluid flows, z_L and z_G, are derived from the corrugation geometry and the packing layer dimensions,

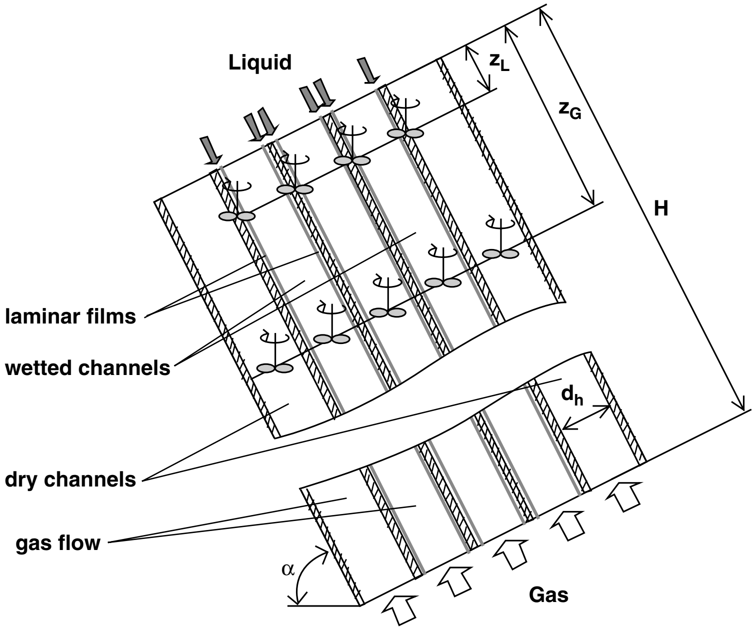

Figure 3. Physical model

D_{pac} and L. The detailed information about the model parameter derivation can be found in Shilkin & Kenig (2005) and Shilkin et al. (2006).

The physical model shown in Figure 3 is assumed to have the same mass transfer efficiency as the actual corrugated sheet structured packing. Contrary to the two-film model (Figure 1) with its main elements representing *stagnant* films, the HA-based model comprises *moving* gas-liquid flow elements and incorporates the experimentally observed hydrodynamic effects.

Heat and mass transport in the suggested model can be described without usage of the transfer coefficients necessary in the two-film model. The mathematical model is based just on a set of partial differential equations covering the hydrodynamics and local transport phenomena. These equations are complemented by the conjugate boundary conditions at the phase interface (Shilkin & Kenig, 2005; Shilkin et al., 2006). A numerical solution yields liquid film thickness and velocity profiles, as well as composition and temperature fields in both phases throughout the packing. These values are used to calculate the average temperature and composition profiles over the packing height.

MODEL VERIFICATION AND COMPARISON

For the verification and comparison of both the traditional and the alternative HA-based model, total reflux distillation data available in the literature are utilised. The results of pilot-scale distillation tests were obtained at the University of Dortmund (Pelkonen, 2001) and at the Nagoya Institute of Technology (Mori et al., 2006). The column in Dortmund was equipped with Montz-Pak A3-500 gauze wire structured packing with an inner diameter of 100 mm and a height of 3 m. Separation of both a binary mixture chlorobenzene/ethylbenzene (CB/EB) and a ternary mixture methanol/acetonitrile/water (MeOH/ACN/WATER) was conducted at ambient pressure with varying feed compositions. Widely used commercial packing Montz-Pak B1-250 was tested at the Nagoya Institute of Technology with the methanol/ethanol/water system (MeOH/EtOH/WATER) in a column of 210 mm inner diameter and the total packing height of about 2.2 m.

For the estimation of the two-film model parameters, different well established correlations for the mass transfer coefficients elaborated at the University of Texas at Austin (Bravo et al., 1985; Rocha et al., 1996) and at the Delft University of Technology (Olujic et al., 2004) are employed. To ensure an adequate comparison, both the two-film and the HA-based model share the same routines and data bases for the calculation of the necessary physical properties (see Shilkin & Kenig, 2005). The same set of operating parameters, namely, flow rate, composition and temperature of the reflux together with the column top pressure, is provided in addition to the corrugation geometry data for both the models.

Plots shown in Figures 4 and 5 compare composition profiles for the binary mixture CB/EB and the ternary mixture MeOH/ACN/ WATER in the column equipped with Montz-Pak A3-500 gauze wire structured packing. In the simulations, the complete surface of the packing is assumed to be wetted, which is appropriate for the organic mixtures investigated.

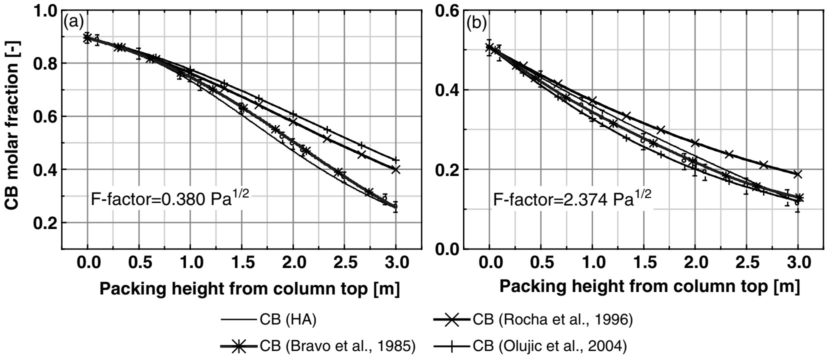

Figure 4. Predicted (lines) versus measured (circles) CB/EB composition profiles in a column with Montz-Pak A3-500 structured packing

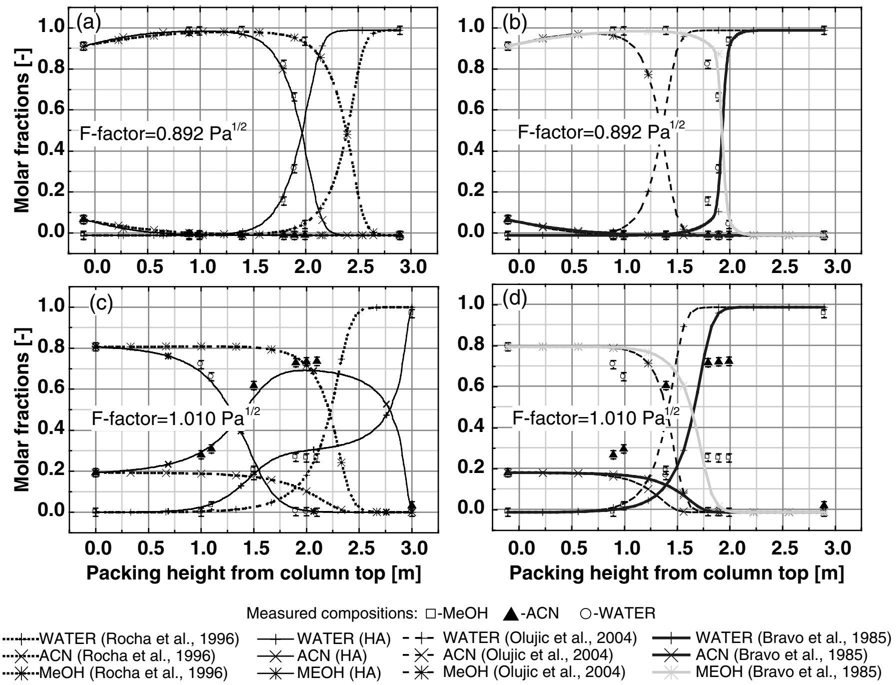

Figure 5. Predicted versus measured composition profiles (MeOH/ACN/WATER) in a column with Montz-Pak A3-500 structured packing

For the low F-factor (Figure 4a), a very good agreement between experimental and simulated profiles is evident for the two-film model with Bravo et al. (1985) correlations for the mass transfer coefficients and the HA-based model. Olujic et al. (2004) correlations perform better at the higher F-factor (Figure 4b), whereas the Rocha et al. (1996) correlations generally tend to somewhat underpredict the measured values in the whole range of the investigated gas-liquid loads. It is worth noting that the Rocha et al. (1996) correlations represent a more recent development of the Bravo et al. (1985) model, extended for the description of both gauze wire and metal sheet structured packings.

Figure 5 demonstrates a comparison for the ternary mixture MeOH/ACN/WATER for two different feed compositions. To give a more clear view, the same set of experimental data is presented in two plots, namely, Figure 5a,b (corresponding F-factor = 0.892 $Pa^{1/2}$) for a lower and Figure 5c,d (corresponding F-factor = 1.010 $Pa^{1/2}$) for a higher ACN feed concentration. Obviously, both the two-film and the HA-based model are capable of predicting the compositions in the column reboiler. Again, the correlations by Bravo et al. (1985) reproduce the experimental composition profiles better than the more recent equations by Rocha et al. (1996). In the case of a higher ACN concentration (see Figure 5c,d), application of the Olujic et al. (2004) correlations also yields a satisfactory agreement with experimental data. However, none of the correlations tested are able to reproduce the ACN accumulation in the lower part of the column, which is accurately predicted with the HA-based model (Figure 5c).

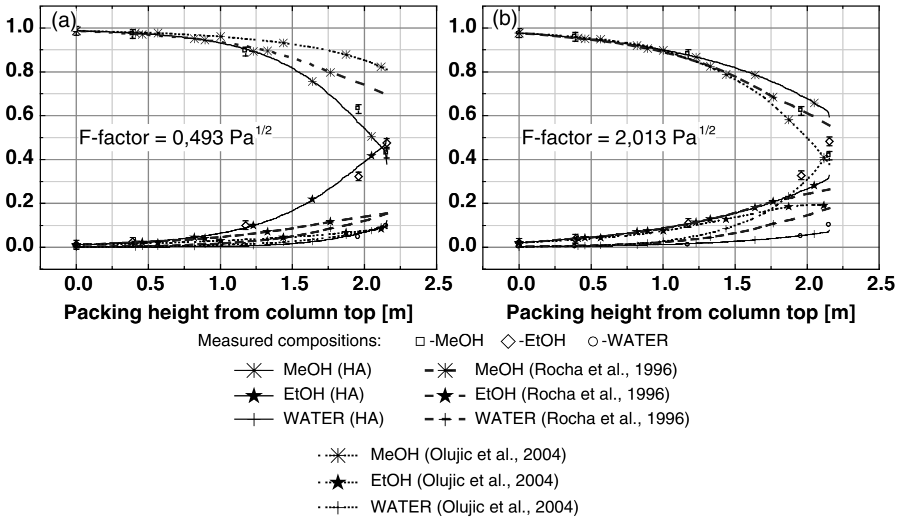

Figure 6. Predicted versus measured composition profiles (MeOH/EtOH/WATER) in a column with Montz-Pak B1-250 structured packing

A comparison between the measured and predicted composition profiles for the Montz-Pak B1-250 metal sheet packing for two different F-Factors is shown in Figure 6. Since the tested correlations maintain their validity within a certain range of operating conditions near the loading point, application of the two-film model results in a poor performance for the low gas-liquid load (see Figure 6a). On the contrary, nearly perfect agreement with experimental data is obtained with the HA-based model. For the higher F-factor (see Figure 6b), the best fit to the measured MeOH profile is achieved with the Olujic et al. (2004) correlations. For the other components, however, discrepancy between the calculated and measured data is smaller with the Rocha et al. (1996) correlations. The HA-based model somewhat underpredicts the packing mass transfer efficiency in the lower part of the packing. This can be attributed to the change in the flow conditions, specifically to the liquid built-up in the bottom of the packing and formation of new flow patterns (e.g., drops, bubbles), which are not considered in the developed hydrodynamic analogy.

CONCLUSIONS

A new model for the prediction of the structured packed columns efficiency based on the hydrodynamic analogy approach is presented and compared with the traditional two-film model. The comparison is carried out using experimental data on total reflux distillation in columns equipped with structured packings of different type. The experiments are conducted with both binary and ternary mixtures and cover a broad range of operating conditions.

The hydrodynamic-analogy-based model is more rigorous and physically consistent than the two-film model. It comprises Navier-Stokes equations, convection-diffusion and heat conduction equations to describe the transport phenomena, thus avoiding usage of the transport coefficients. Application of these fundamental equations becomes possible due to a reasonable replacement of the actual two-phase flow in the packings by a combination of geometrically simpler flow patterns.

The accuracy of the two-film model is demonstrated to be strongly affected by the applied correlations, test systems and operating conditions. Among the tested mass transfer correlations, the best fit to the experimental data is obtained with Bravo et al. (1985) and Olujic et al. (2004) equations for the gauze wire and metal sheet structured packing, respectively. On the contrary, the model based on the hydrodynamic analogy approach demonstrates a stable performance over a broad range of F-factors, test systems and for the both investigated packing types. Some deviations from the experimentally measured composition profiles appear only in the lower part of the packing, when the flow conditions close to the loading point are considered. They are attributed to the change in the flow conditions, which have not yet been accounted for in the physical model developed.

The developed hydrodynamic-analogy-based model can also be used for the determination of the mass transfer correlations for the two-film model, thus avoiding/minimising expensive experimental pilot-plant studies.

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