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Simulation of Mass Transfer in Reactive Absorption

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Abstract

The discretization of the column and the films plays a significant role in mass transfer calculations and changes results significantly. The use of special grid distribution for the discretization helps to reduce the computational effort and guarantees reasonable results. The performance of these grid distributions will be demonstrated for a known and solved problem. The differences in calculation result will be shown for a column simulation.

Keywords: mass-transfer, discretization, Stefan-Maxwell, reactive absorption.

1. Introduction

Heat and mass transfer calculations based on Stefan-Maxwell equations are nearly state-of-the-art for chemical absorption. Unfortunately, there are only a few investigations about the discretization depth needed to obtain reasonable results. Here, predictions of a rigorous model are compared with analytical solutions for a well-known problem. In particular, the impact of a different number of film segments, and of different grid distributions, is investigated. Furthermore, a method for choosing a meaningful grid distribution to obtain accurate results with a low number of film segments is proposed, which helps to reduce the computational effort of these calculations.

2. Modeling

The model normally used for simulation of heat and mass transfer in a counter-current column is quite similar to the model of theoretical stages. The column is also divided vertically into segments as can be seen in Figure 1. In each of these non-equilibrium segments, gas and liquid are exchanging heat and mass.

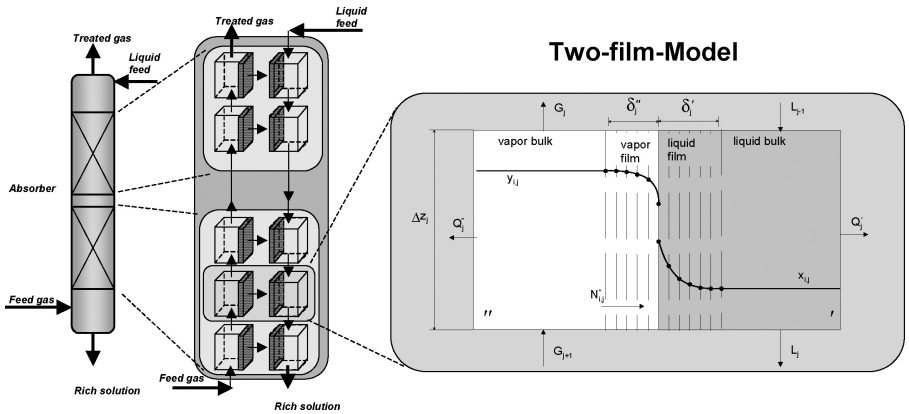

Fig.1. Scheme of the simulation model

The two-film model is used for the description of the heat and mass transfer. In this model, one assumption is that the bulk phases are ideally mixed with uniform concentrations and temperatures. At the interface, vapor and liquid are in equilibrium. The heat and mass transfer resistances are assumed to lie in the film layers separated by the gas-liquid interface. The two films are stagnating so no convective mass transfer, but only diffusive mass transfer, is considered. As with mass transfer, for the case of heat transfer, only the conductive heat and not the convective heat transfer has been considered.
The films are further divided into film segments. With these film segments, it is possible to calculate more accurately the concentration profiles in the film. This is essential for use in reactive systems, where as a consequence of the reaction the mass transfer can be enhanced.
The simulation model allows for chemical reactions in the holdup of the bulk phases, and also within the film. The diffusion of the different components in the film layers is described with the Stefan-Maxwell equations.

3. Influence of Film discretization

For reactive systems it is known that the description of the concentration profiles in the film is essential (cf. Danckwerts (1970) and Astarita et al. (1983)). The description of mass transfer enhanced by chemical reaction is investigated for a solved problem to test the model. Similar investigations are reported from Frank et al. (1995a,b) and Versteeg et al. (1989,1990). Here the aim was to find out the required film discretization. Furthermore, the use of different grid distributions within the film was investigated, since it has been reported (Versteeg et al. (1989)) that they can reduce the computational effort.
To check the results of the mass transfer simulation with chemical reactions a simple, well-known problem (cf. Danckwerts (1970) and Astarita et al. (1983)) was used. The example investigated consists of a gas *A* which is absorbed into a liquid. The liquid is consisting of a solvent *S* and a component *B*, which is able to react with *A*. An irreversible reaction of the following type is considered:

$$A + 2\,B \rightarrow 3\,C \tag{1}$$

The reaction rate r of this reaction is second order. Dependent on the order of the kinetic constant k and the mass transfer coefficient β' in the liquid the mass transfer of the component *A* may be enhanced compared to a physical absorption due to the reaction. A measure of the enhancement is the enhancement factor *E* which is the ratio of the mass transfer rate with chemical reaction to mass transfer rate for a physical absorption.

$$E = \frac{\dot{n}_{chem}}{\dot{n}_{phys}} \tag{2}$$

The influence of the kinetic and the mass transfer coefficient on the enhancement is shown in Figure 2. Here the Hatta number is defined as

$$Ha = \frac{\sqrt{k \cdot c_B \cdot D_A}}{\beta'} \tag{3}$$

and the enhancement for an infinitely fast reaction kinetic (instantaneous reaction, E infinity) for the film model is given by

$$E^{\infty} = 1 + \frac{c_B}{2c_A}\frac{D_A}{D_B} \tag{4}$$

Herein c_A the molar concentration (moles per volume) of A at the interface, c_B the molar concentration of B in the bulk, D_A and D_B the diffusivities of A and B.

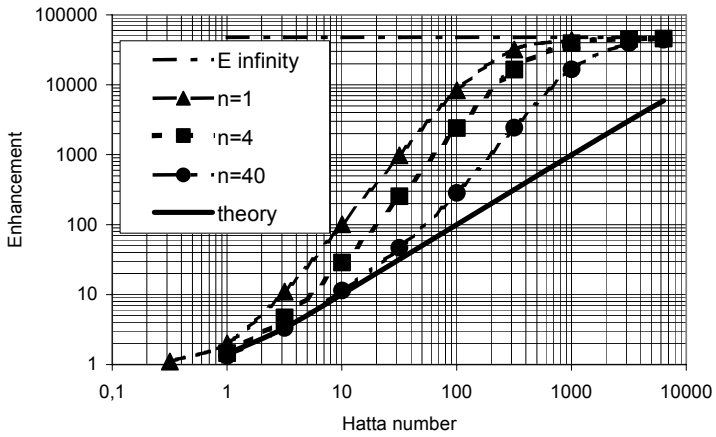

Fig.2. Enhancement for different numbers of film segments

In Fig. 2 the full line represents the results of the numerical solution of the exact differential equations. The lines with the symbols show calculation results with the model for different numbers (1, 4 and 40) of equidistant film segments. As can be seen not even 40 equidistant film segments are sufficient to describe the enhancement correctly for the complete Hatta-range given in Fig. 2. Only 2 points are described well, independent of the number of films: The physical absorption (E=1) and the instantaneous reaction (E=E^{∞}). For the rest the enhancement predicted with the model is significantly overestimated.

For the practical use of the model this has 2 severe consequences:

1. The use of the model to evaluate reaction kinetics will lead to reaction kinetics, which are too low.
2. The use of literature kinetics will in general (also dependent on other physical properties) result in an overestimation of the enhancement.

In addition, the use of very large numbers of film segments is unfavourable for column simulations or simulations of large flow sheets in terms of convergence stability and computational effort.

In Figure 3 the concentration profiles of a simulation with 40 film segments is shown for a Hatta number of 101. As can be seen although the number of film segment is quite high only a few segments lie within the sharp decrease of component A at the interface. Therefore, the description of the concentration gradient at the interface will not be very accurate. The best option would be to have an adaptive grid distribution with a high number of grid in the film region where the biggest changes occur. Unfortunately, there is as yet no information available on this method.

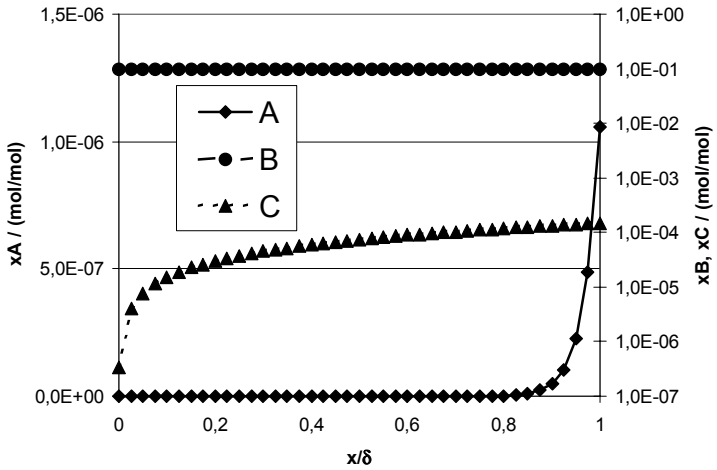

Fig. 3. Concentration profile for Ha=101

Alternatively an unsymmetrical grid distribution with more grids near to the interface will help. One possible kind of grid distributions is

$$x_i = \delta \left(\frac{i}{n} \right)^{1/m} \tag{5}$$

with x_i the position of the i-th grid in the film with the total thickness δ. n denotes the number of films, and m is a grid distribution parameter. For m equal to one, this results in an equidistant grid distribution. For higher numbers of m, the grid distribution will shift towards the interface.
If m is chosen in the following way

$$m = Ha \cdot \ln 2 \tag{6}$$

then a sufficiently high proportion of all grids should lie within the region with the biggest changes.
In Figure 4 it is shown that with this method, the theoretical enhancement curve is described well, even though only 6 film segments are used.

4. Influence of discretization on column simulation

In the following, a column simulation of the operating data of Pintola et al. (1993) will be discussed. Pintola et al. investigated the CO_2 absorption into an aqueous MEA solution in a packed column (3 beds with 4,6 m 2" pall rings, diameter 1,9 m). Of this data set only the first one will be taken into account. For a symmetrical, non-equidistant grid distribution with 6 film segments, Kucka (2002) investigated the agreement between simulation and experimental data. In his investigations he found out that 12 segments for the column height should be sufficient. The agreement between his simulation (6,5 ppm CO_2 in the treated gas) with the operational data (8 ppm CO_2) is excellent.

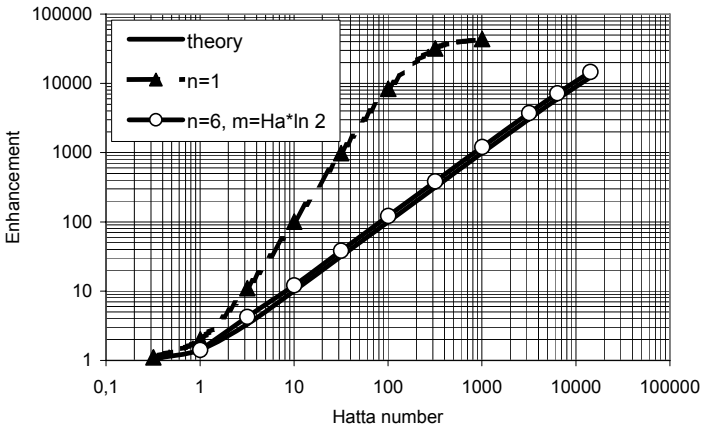

Fig.4. Enhancement for 6 film segments with variable grid distribution.

Since the model used here is similar, the reproduction of this result with a similar (but slightly different) grid distribution was possible and resulted in 5,6 ppm CO_2 at the top of the column. However, in contradiction to the results of Kucka's investigations a higher number of height segments decreased the concentration at the top until the equilibrium value of 4 ppm CO_2 at the top was reached (compare Fig. 5).

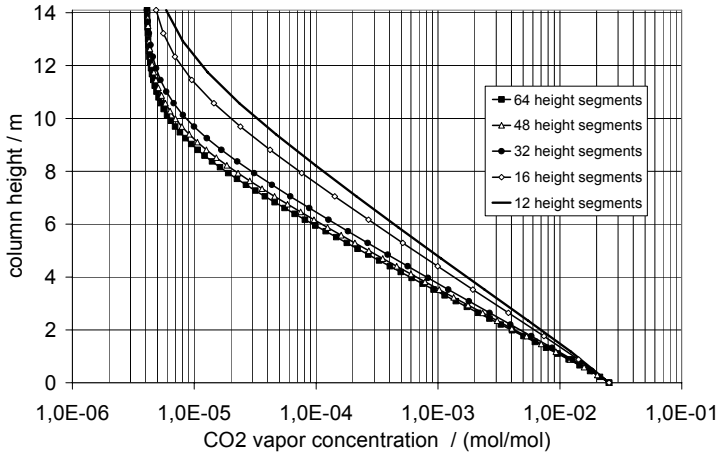

Fig.5. Column profiles for different numbers of height segments (6 film segments)

Of course there is also an influence of the film distribution. In this example the Hatta number is about 50. According to eq. 6 a grid distribution with m=35 should be used. With this grid distribution it was possible to get a good description of the gradient at interface of CO_2. As can be seen in Figure 6 the profiles with this grid distribution show a completely different CO_2 treated gas specification (54 ppm with 32, 41 ppm with 64 and 39 ppm with 80 height segments). This result shows again that the wrong grid distribution will lead to a overestimation of the enhancement. Unfortunately, the more accurate calculation result is not in agreement with the operational data.

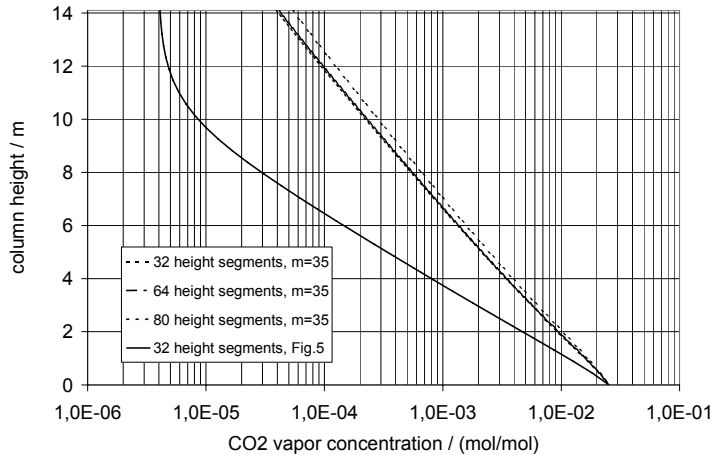

Fig.6. Column profiles for different discretizations (6 film segments)

5. Conclusions

The importance of the discretization on the results of mass transfer calculations with a Stefan-Maxwell approach has been demonstrated. A grid distribution was presented which helps to reduce computational effort and guarantees reasonable results.

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