LETTERS IN HEAT AND MASS TRANSFER 0094-4548/79/0101-0073$02.00/0
Vol. 6, pp. 73-76, 1979

COMPARISON OF MODELS FOR TERNARY MASS TRANSFER

R. Krishna†

Department of Chemical Engineering
University of Manchester Institute of Science and Technology
Sackville Street, Manchester M60 1QD, England

(Communicated by E. Hahne)

ABSTRACT

Methods for predicting ternary mass transport behaviour from information on the transport parameters of the constituent binary pairs are discussed. Experimental data obtained by Modine in a wetted-wall column for mass transfer between a falling film of a binary liquid mixture containing acetone and benzene and a downward flowing vapour-gas mixture containing acetone, benzene and either nitrogen or helium are used to test three predictive transport models:

Model I: a multicomponent 'film' model based on an exact matrix method of solution to the Maxwell-Stefan equations,

Model II: a model utilizing the linearized theory of multicomponent mass transfer and

Model III: a model which assumes that acetone and benzene transfer independently of each other.

For measured inlet conditions at the top of the wetted-wall column, the outlet conditions (temperatures, compositions, flows) and overall rates of transfer predicted by the three models are compared with the experimentally obtained values. It is seen that the predictions of Models I and II are reasonably close to the experimental values whereas Model III shows large deviations. The differences between the predictions of Models I and II are not found to be significant.

It is concluded that for the system studied, diffusional interactions are significant and that a general treatment of multicomponent mass transfer must proceed via non-diagonal matrices of mass transfer coefficients, calculable reasonably accurately by multicomponent 'film' models.

† Present Address: Koninklijke/Shell-Laboratorium, Amsterdam, Badhuisweg 3, 1031 CM Amsterdam-N, The Netherlands.

Ternary Mass Transfer Models

For two-component systems, the diffusion flux of species i in a mixture of i and j is conventionally written as

$$J_i = k^{\bullet}_{ij}(y_{ib} - y_{iI}) \tag{1}$$

where $k^{\bullet}_{ij}$ is the binary mass transfer coefficient corrected for finite rates of transfer [1]. Using a 'film' model, this finite flux mass transfer coefficient can be related to the limiting 'zero flux' mass transfer coefficient, k_{ij}, by

$$k^{\bullet}_{ij} = k_{ij}\frac{\phi}{\exp\phi - 1}; \quad \text{with} \quad \phi = \frac{N_i + N_j}{k_{ij}} \tag{2}$$

The zero flux binary mass transfer coefficient k_{ij} can be calculated for various transfer operations using standard correlations say of the j-factor type:

$$j_D = \frac{k_{ij}\,A}{G_t}\left(\frac{\mu}{\rho\,\mathcal{D}_{ij}}\right)^{2/3} = f(Re) \tag{3}$$

For ternary systems there two independent fluxes and driving forces and the proper generalization of (1), taking account of coupling between species transfers, is in 2-dimensional matrix notation

$$(J) = [k^{\bullet}](y_b - y_I) \tag{4}$$

From a practical engineering view point it is important to be able to estimate the elements $k^{\bullet}_{ij}$ from existing binary correlations, say of type (3), and transport parameters (such as $\mathcal{D}_{ij}$) of the constituent binary pairs of the ternary mixture. In 1964 Toor [2] and Stewart and Prober [3] independently developed a procedure for achieving this. Their method, the linearized theory of multicomponent mass transfer, is based on the assumption that the matrix of diffusion coefficients in ternary mixture, $[D]$, remains constant along the diffusion path. The matrix of mass transfer coefficients is obtained as

$$[k^{\bullet}] = [P]\left[\hat{k}^{\bullet}_i\right][P]^{-1} \tag{5}$$

where $[P]$ is the modal matrix of $[D]$. With a film theory, the pseudo-finite flux mass transfer coefficients, $\hat{k}^{\bullet}_i$, are given as [2,3,4]

$$\hat{k}^{\bullet}_i = \hat{k}_i\frac{\psi_i}{\exp\psi_i - 1}; \quad \text{with} \quad \psi_i = \frac{N_1 + N_2 + N_3}{\hat{k}_i}, \quad i = 1,2 \tag{6}$$

where the pseudo-zero flux mass transfer coefficients $\hat{k}_i$ may be estimated from the binary correlations (3) using the ith eigenvalue $\hat{D}_i$ of $[D]$ in place of the binary diffusivity $\mathcal{D}_{ij}$. For ideal gas mixtures, the matrix of

diffusion coefficients, $[D]$, can be estimated from the constituent $\mathcal{D}_{ij}$ and therefore $[k^{\bullet}]$ can be calculated from binary data alone.

More recently, Krishna [5] developed a multicomponent mass transfer film model which is based on an exact matrix method of solution to the Maxwell-Stefan equations. They derive an expression for the finite flux multicomponent mass transfer coefficients as

$$[k^{\bullet}] = [k][\Phi]\{\exp[\Phi] - \Gamma_I\}^{-1} \tag{7}$$

which is the exact matrix analogue of (2). The elements of the matrix of zero flux mass transfer coefficients are obtained in terms of the constituent k_{ij} of the binary pairs as:

$$k_{ij} = \frac{k_{j3}\{y_i(k_{3-j,3} - k_{12}) + \delta_{ij}k_{12}\}}{y_1k_{23} + y_2k_{13} + y_3k_{12}}, \qquad i,j = 1,2 \tag{8}$$

The constituent k_{ij} may be estimated from (3) and the matrix $[k^{\bullet}]$ is thereobtainable purely from binary information by use of equations (7) and (8).

When components 1 and 2 in the ternary mixture are infinitely dilute, the matrix $[k^{\bullet}]$ from both the ternary models above simplifies to

$$\begin{bmatrix} k^{\bullet}_{11} & k^{\bullet}_{12} \\ k^{\bullet}_{21} & k^{\bullet}_{22} \end{bmatrix} = \begin{bmatrix} k^{\bullet}_{13} & 0 \\ 0 & k^{\bullet}_{23} \end{bmatrix} \tag{9}$$

For systems which are quite dilute in 1 and 2, one would imagine that relations (9) might represent a good approximation of the ternary behaviour.

It is the object of the work presented here to test the predictive capabilities of the three models described above by comparison with available experimental data. For purposes of this test, the experimental data obtained by Modine [6,7] for ternary mass transfer between a vapour-gas mixture containing acetone(1)-benzene(2)-nitrogen(3)or helium (3) and a binary liquid mixture of acetone and benzene in a wetted-wall column were used. The vapour and liquid streams were in co-current flow. For conditions measured at the top of the column, the binary mass transfer correlations [7] were used to predict the behaviour of the ternary mixture in the column and the conditions at the outlet determined. All the three ternary transport models described above were used for this purpose. The predictions of the three models were compared with the experimental values determined by Modine [6]. The major results obtained by this comparison are described below.

Results, Discussion and Conclusions

The predictions of the non-interacting Model III, taking the cross-mass transfer coefficients as zero (equation (9)) showed large deviations from measured data and in one particular run anticipated the wrong direction of transfer. The Krishna [5] multicomponent model I and the Toor-Stewart and Prober linearized theory model II both predicted the ternary behaviour reasonably well; no significant difference between the predictions of the latter two models was observed. The phenomenon of reverse mass transfer was properly anticipated by models I and II for one particular run 7.

The interesting thing to note about the results is that even though the experimental system was studied in the low vapour concentration range (total composition of acetone and benzene was less than 18 % for any run), the non-interacting model III predictions are hopelessly in error. The approximation suggested by equation (9) must therefore be viewed with extreme caution. The reason for the existence of such large interactions, especially for the helium runs, is that small cross mass transfer coefficients may be multiplied by large composition driving forces to give rise to large deviations from pseudo-binary behaviour described by equation (9).

It may therefore be concluded that the general description of multicomponent mass transfer must include non-zero cross coefficients. The multicomponent film models, I and II, provide a simple means of estimating the elements $k^{\bullet}_{ij}$ at least for the gas phase. The applicability to liquid phase transport must be examined by further experimentation.

References

1. R.B. Bird, W.E. Stewart and E.N. Lightfoot, Transport Phenomena, Wiley, New York (1960)

2. H.L. Toor, A.I.Ch.E.J., 10, 460 (1964)

3. W.E. Stewart and R. Prober, Ind.Eng.Chem.Fundam., 3, 224 (1964)

4. W.E. Stewart, A.I.Ch.E.J., 19, 398 (1973)

5. R. Krishna, Chem.Eng.Sci., 32, 659 (1977)

6. A.D. Modine, Ternary Mass Transfer, Ph.D. Dissertation, Carnegie Institute of Technology, Pittsburgh (1963)

7. A.D. Modine, E.B. Parrish and H.L. Toor, A.I.Ch.E.J., 9, 348 (1963)