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An alternative linearized theory of multicomponent mass transfer

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There has been increasing interest in the area of multicomponent mass transfer because of the many practical applications in distillation[1, 2], condensation[3] and extraction[4]. The linearized theory of Toor[5] and Stewart and Prober[6] paved the way to the practical solution to a host of multicomponent diffusion problems. With their linearization technique solutions to multicomponent diffusion problems can be written down as straightforward matrix generalizations of standard binary solutions. It may be noted here that for steady-state diffusion in ideal gas mixtures linearization is unnecessary and an exact solution is possible[7].

Multicomponent film models have been used to form the basis of design procedures for distillation[2] and condensation[3, 8] equipment. However, one possible reason why rigorous multicomponent models have not gained widespread usage in industrial practice may be the fact that even for the simplest case of the film model a trial and error procedure is required for the calculation of the transfer fluxes $\mathbf{N}_i$ at any position in the equipment. The interative procedure is necessary because of the influence of the transfer fluxes themselves on the mass transfer coefficients[5–7].

In the present communication we develop an alternative to the Toor–Stewart–Prober linearized theory with the objective of providing a simpler, yet accurate, procedure for calculation of interfacial transfer fluxes. The notations used coincide with those used in our previous publications[2, 7] and only newly introduced symbols are defined in the text.

PRELIMINARIES

The differential continuity relations can be written for an n-component non-reacting system as

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (c_i \mathbf{u}_i) \equiv \frac{\partial c_i}{\partial t} + \nabla \cdot \mathbf{N}_i = 0 \tag{1}$$

$$i = 1, 2, \ldots n$$

where $\mathbf{N}_i$ is the molar flux of component i with respect to a stationary coordinate reference frame and is given by

$$\mathbf{N}_i = c_i\,\mathbf{u}_i, \qquad i = 1, 2, \ldots n. \tag{2}$$

The mixture total flux is obtained by summing eqn (2) over the n species:

$$\mathbf{N}_t = \sum_{i=1}^{n} \mathbf{N}_i = \sum_{i=1}^{n} c_i\,\mathbf{u}_i = c_t \sum_{i=1}^{n} x_i\,\mathbf{u}_i = c_t\,\mathbf{u} \tag{3}$$

where $\mathbf{u}$ is the molar average mixture velocity.

The molar diffusion flux of component i with respect to the molar average mixture velocity, $\mathbf{J}_i$, may be defined as:

$$\mathbf{J}_i \equiv c_i\,(\mathbf{u}_i - \mathbf{u}) \equiv \mathbf{N}_i - x_i\,\mathbf{N}_t. \tag{4}$$

With the definitions (2)–(4), the differential continuity relations (1) may be rewritten as

$$c_t \frac{\partial x_i}{\partial t} + c_t\,\mathbf{u} \cdot \nabla x_i = -\nabla \cdot \mathbf{J}_i \tag{5}$$

$$i = 1, 2, \ldots n - 1$$

where only $n - 1$ of the equations are independent due to the constraints on the mole fractions'

$$\sum_{i=1}^{n} x_i = 1 \tag{6}$$

and on the molar diffusion fluxes

$$\sum_{i=1}^{n} \mathbf{J}_i = 0. \tag{7}$$

To allow calculation of the n molar fluxes $\mathbf{N}_i$, which appear in the mass balance relations for processing equipment, from solution of eqn (5) we require two additional pieces of information: (1) an additional determinancy condition which usually takes the form of a linear dependence between the fluxes $\mathbf{N}_i$ and (2) constitutive relations for the diffusion fluxes $\mathbf{J}_i$.

We consider these in turn.

DETERMINANCY CONDITION

In most practical situations such as those existing in distillation, absorption, condensation, extraction and diffusion with heterogeneous chemical reactions, the additional determinancy condition required may be expressed as a linear constraint on the fluxes $\mathbf{N}_i$:

$$\sum_{i=1}^{n} \lambda_i \mathbf{N}_i = 0. \tag{8}$$

Equations (3), (4) and (6)–(8) may be combined to express the molar fluxes $\mathbf{N}_i$ in terms of the diffusion fluxes $\mathbf{J}_i$ in $n-1$ dimensional matrix notation as

$$(\mathbf{N}) = [\beta]\,(\mathbf{J}) \tag{9}$$

where the elements β_{ij} of the "bootstrap" matrix are given by[2,7]

$$\beta_{ij} = \delta_{ij} - x_i \left[\frac{\lambda_j - \lambda_n}{\sum\limits_{k-1}^{n} x_k \lambda_k} \right], i, j = 1, 2, \ldots n-1. \tag{10}$$

The nth flux $\mathbf{N}_n$ can be obtained from

$$\mathbf{N}_n = -\sum_{i=1}^{n-1} \lambda_i \mathbf{N}_i / \lambda_n. \tag{11}$$

Various special cases may be identified:

(i) *Equimolar diffusion*

$$\mathbf{N}_t = 0; \lambda_1 = \lambda_2 = \lambda_3 = \ldots = \lambda_n; \beta_{ij} = \delta_{ij}. \tag{12}$$

(ii) *Diffusion of $n-1$ species through stagnant species n (Stefan diffusion)*:

$$\mathbf{N}_n = 0; \lambda_1 = \lambda_2 = \lambda_3 = \ldots = \lambda_{n-1} = 0; \beta_{ij} = \delta_{ij} + x_i/x_n. \tag{13}$$

(iii) *Non-equimolar distillation*[2]:

$$\lambda_i \equiv \bar{H}_i^y - \bar{H}_i^x. \tag{14}$$

(iv) *Diffusion with heterogenous chemical reaction with flux rations $\mathbf{N}_i/\mathbf{N}_t$ specified by the stoichiometry of the reaction*:

$$\beta_{ij} = \delta_{ij}/(1 - x_i \mathbf{N}_t/\mathbf{N}_i). \tag{15}$$

Equations (9)–(11) allow the calculation of the n molar fluxes $\mathbf{N}_i$ from knowledge of the $n-1$ diffusion fluxes $\mathbf{J}_i$. The latter can be obtained from the composition gradients in the diffusion path provided we have the proper constitutive relations. These constitutive relations are considered next.

CONSTITUTIVE RELATIONS FOR n-COMPONENT DIFFUSION

For non-ideal mixtures, one of the most convenient forms of the constitutive relations are the generalized Maxwell–Stefan diffusion equations[7]. These may be written as

$$\frac{x_i}{RT} \nabla \mu_i = \sum_{\substack{j=1 \\ j \neq i}}^{n} \frac{x_i \mathbf{N}_j - x_j \mathbf{N}_i}{c_t Ð_{ij}} = \sum_{\substack{j=1 \\ j \neq i}}^{n} \frac{x_i \mathbf{J}_j - x_j \mathbf{J}_i}{c_t Ð_{ij}}, i = 1, 2, \ldots n \tag{16}$$

The eqns (16) may be recast into $n-1$ dimensional matrix notation as follows (see Ref.[7] for detailed derivation and definitions of the various coefficients):

$$(\mathbf{J}) = -c_t\,[B]^{-1}\,[\Gamma]\,(\nabla x) \tag{17}$$

from which we see that the matrix of Fickian diffusion coefficients $[D]$ is given by

$$[D] = [B]^{-1}[\Gamma]. \tag{18}$$

The matrix of thermodynamic factors $[\Gamma]$ reduces to the identity matrix for thermodynamically ideal fluid mixtures.

Combination of eqns (9) and (17) gives the constitutive relations for the $n-1$ molar fluxes $\mathbf{N}_i$ as

$$(\mathbf{N}) = -c_t[\beta][B]^{-1}[\Gamma](\nabla x) = -[W](\nabla x) \tag{19}$$

where we have additionally defined a matrix of transfer coefficients $[\mathrm{W}]$, defined in terms of the molar fluxes $\mathbf{N}_i$:

$$[W] \equiv [\beta]\,[B]^{-1}\,[\Gamma]. \tag{20}$$

LINEARIZATION OF THE MULTICOMPONENT DIFFUSION EQUATIONS

The Toor–Stewart–Prober approach is essentially to assume that $c_t\,[D]$ is independent of the composition. With this assumption the $n-1$ differential continuity relations (5), together with eqns (17) and (18), may be written in matrix notation ($n-1$ dimensional) as

$$\frac{\partial(x)}{\partial t} + \mathbf{u} \cdot (\nabla x) = [D]_a(\nabla^2 x) \tag{21}$$

where the subscript a on the matrix of Fickian diffusion coefficients serves as a reminder that the elements of the matrix have to be evaluated at some averaged composition, normally chosen as the arithmetic average.

The equations (21) can be diagonalized by use of the similarity transformation[5,6], reducing the multicomponent diffusion equations to a series of uncoupled equations, $n-1$ in number, each of which corresponds to an equivalent binary problem but with the binary diffusivity replaced by D'_{ia}, the ith eigenvalue of the matrix $[D]_a$. Thus if the binary solution exists, the multicomponent solution can be written straightforwardly.

For steady state diffusion along a film of thickness δ with the boundary conditions

$$\begin{aligned} r &= 0 \qquad x_i = x_{io}, \\ r &= \delta, \qquad x_i = x_{i\delta}, \end{aligned} \tag{22}$$

the molar fluxes N_i are obtained using the Toor–Stewart–Prober technique as

$$(N) = [\beta]_o[k^\bullet](x_o - s_\delta) \tag{23}$$

where the matrix of finite flux mass transfer coefficient $[k^\bullet]$ is given by

$$[k^\bullet] = [P][k'][P]^{-1} \tag{24}$$

where the diagonal matrix of pseudo-binary finite flux mass transfer coefficients is given by

$$k_i'^\bullet = k_i' \frac{\phi_i'}{\exp \phi_i' - 1}, \quad i = 1, 2, \ldots n-1 \tag{25}$$

with the pseudo-binary zero flux mass transfer coefficients given by

$$k_i' \equiv c_t D'_{ia}/\delta, \quad i = 1, 2, \ldots n-1 \tag{26}$$

and the pseudo-binary dimensionless mass transfer rate factors:

$$\phi_i' \equiv N_t/k_i', \quad i = 1, 2, \ldots n-1. \tag{27}$$

Unless conditions of equimolar diffusion ($N_t = 0$) prevail the calculation of the fluxes N_i requires a trial and error procedure. The same problem exists for unsteady state mass transfer under conditions of finite mass transfer rates. The crux of the problem is the convective mass transfer term, the second term of the left hand side of eqn (21).

In developing our proposed linearization technique, we note that the assumption of constant $[D]$ in the Toor–Stewart–Prober approach is approximate *even for ideal gas mixtures*. The elements D_{ij} are strong functions of composition for highly nonideal fluid mixtures. At this stage in the theoretical development there is no reason to suppose that the elements D_{ij} are any less composition dependent than the elements W_{ij}, defined by eqn (20). We shall proceed further and explore the consequences of assuming constant W_{ij} (in practice the elements will have to be evaluated at some averaged composition).

The assumption of constant W_{ij} allows the eqns (1) to be written as

$$\frac{\partial(x)}{\partial t} = [W]_a(\nabla^2 x) \tag{28}$$

where the subscript a on the matrix [W] again serves as a reminder that the elements have to be evaluated at some average composition. The differential eqn (28) can be diagonalized in exactly the same manner as described by Toor and Stewart and Prober but the solution is much simpler than for the corresponding eqn (21) resulting from the Toor–Stewart–Prober treatment.

For steady-state diffusion along a film of thickness δ, the fluxes N_i can be calculated from

$$(N) = \frac{c_t}{\delta}[W]_a(x_o - x_\delta) = \frac{c_t}{\delta}[\beta]_a[B]_a^{-1}[\Gamma]_a(x_o - x_\delta). \tag{29}$$

The penetration model for finite transfer rates is obtained as

$$(N) = 2c_t\left(\frac{[W]_a}{\pi t_c}\right)^{1/2}(x_o - x_\delta) \tag{30}$$

where t_c is the contact time between the phases. The square root of the matrix $[W]_a$ can be evaluated by the use of Sylvester's theorem.

It is clear on comparison of eqn (29) and the corresponding set of relations for calculation of N_i using the Toor–Stewart–Prober technique, eqns (23)–(27), that the current linearization technique gives an explicit expression for the transfer fluxes without the need for iterations.

NUMERICAL EXAMPLE

To test the accuracy of the linearization technique developed in this paper we shall consider a numerical example involving non-equimolar distillation in the system pentane–2 (1)–ethanol (2)–water (3). The problem is to calculate the interfacial transfer fluxes N_i at a point in a vapour–liquid contacting apparatus under the conditions of vapour phase diffusion controlled transfer. The specified conditions are as follows:

Compositions (mole fractions) at either ends of diffusion path: $x_{1o} = 0.630$; $x_{1\delta} = 0.590$; $x_{2o} = 0.165$; $x_{2\delta} = 0.095$.

Vapour phase diffusion coefficients of binary pairs: $\mathcal{D}_{12} = 7.27\ \text{mm}^2/\text{s}$; $\mathcal{D}_{13} = 14.4\ \text{mm}^2/\text{s}$; $\mathcal{D}_{23} = 20.9\ \text{mm}^2/\text{s}$

Film thickness. $\delta = 10\ \mu\text{m}$

Temperature $T = 346$ K; *Pressure* $p = 100$ kPa

Partial molar enthalpies in the vapour and liquid phases (MJ/kmol). $\bar{H}_1^y = 38$; $\bar{H}_2^y = 50.6$; $\bar{H}_3^y = 47$; $\bar{H}_1^x = 15.5$; $\bar{H}_2^x = 10.1$; $\bar{H}_3^x = 5$.

The fluxes N_i calculated using eqn (29), with the parameters evaluated at the arithmetic averaged composition, are as follows:

$$N_1 = 4.62\ \text{mol/s/m}^2;\ N_2 = 3.04\ \text{mol/s/m}^2;\ N_3 = -5.40\ \text{mol/s/m}^2.$$

As a comparison to test the accuracy of the method, the exact solution to the problem was also obtained using the method described elsewhere[2, 7]. The exact solution leads to the values given below:

$$N_1 = 4.6\ \text{mol/s/m}^2;\ N_2 = 3.03\ \text{mol/s/m}^2;\ N_3 = -5.4\ \text{mol/s/m}^2$$

which shows that linearized solution if of excellent accuracy. In the two sets of calculations given above, the non-equimolar transport processes caused by differences in molar heats of vaporizations are properly taken into account. If on the other hand, we assume that conditions of equimolar diffusion prevail (as is traditionally done in text-book treatments of distillation mass transfer), the molar fluxes are obtained using the model given in Ref. [2] as:

$$N_1 = 3.23\ \text{mol/s/m}^2;\ N_2 = 2.74\ \text{mol/s/m}^2;\ N_3 = -5.97\ \text{mol/s/m}^2$$

which shows that the assumption $N_t = 0$ significantly underestimates the molar flux of volatile pentane-2, as might be expected from physical considerations.

The example chosen above to demonstrate the accuracy of the linearization technique developed here is fairly typical of a large class of systems showing moderate to high diffusional interaction phenomena. The linearization technique was also tested for a variety of other ternary gas diffusion problems involving Stefan diffusion. The results of these investigations have been reported in detail elsewhere[9]. These results confirm the conclusions reached above regarding the accuracy of the linearization technique.

CONCLUDING REMARKS

We have considered a linearization technique for the solution of multicomponent diffusion problems. The technique consists in assuming that the product of the bootstrap matrix $[\beta]$ and the matrix of Fickian diffusion coefficients $[D]$ is constant along the diffusion path. Clearly if equimolar diffusion prevails ($\beta_{ij} = \delta_{ij}$), then this assumption reduces to the Toor–Stewart–Prober assumption of constant $[D]$. It has been emphasized in the discussions that there is no theoretical or experimental reason to suppose that the matrix $[W]$ is any less constant than the matrix $[D]$. The accuracy of the method has been demonstrated by means of numerical examples given in this paper and elsewhere[9]. Due to the fact that the current linearization technique leads to a non-iterative procedure for the calculation of the fluxes N_i, the incorporation of the procedure into standard design procedures for processing equipment could take place easily.

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NOTATION

$[B]$	matrix of inverted diffusion coefficients with elements given by:

$$B_{ii} = \frac{x_i}{\text{Đ}_{in}} + \sum_{\substack{k=1\\k\neq 1}}^{n}\frac{x_k}{\text{Đ}_{ik}}, \quad i = 1, 2, \ldots n-1$$

$$B_{ij} = -x_i(1/\text{Đ}_{ij} - 1/\text{Đ}_{in}), \quad \substack{i,j\\i\neq j} = 1, 2, \ldots n-1$$

c_i	molar concentration of species i
c_t	mixture solar concentration, $c_t = \sum_{i=1}^{n} c_i$
Đ_{ij}	generalized Maxwell–Stefan diffusion coefficient for binary pair i–j. For gas mixture, $\text{Đ}_{ij} = \mathcal{D}_{ij}$
$[D]$	matrix of Fickian diffusion coefficients
D'_i	ith eigenvalue of Fickian matrix $[D]$
$\mathcal{D}_{ij}$	Vapour phase diffusivity of binary pair i–j
$\bar{H}_i$	partial molar enthalpy of i in mixture
$\mathbf{J}_i$	molar diffusion flux of i with respect to molar average reference velocity
$[k^\bullet]$	matrix of finite flux mass transfer coefficients
$[k'^\bullet]$	diagonal matrix of pseudo-binary mass transfer coefficients
k'_i	pseudo-binary zero-flux mass transfer coefficients
n	number of components in mixture

$\mathbf{N}_i$	molar flux of i with respect to stationary coordinate reference frame
$\mathbf{N}_t$	mixture total molar flux
p	total system pressure
$[P]$	modal matrix of [D]
r	distance coordinate along diffusion path
R	gas constant
t	time
T	temperature
$\mathbf{u}_i$	diffusion velocity of species i in mixture
$\mathbf{u}$	molar average mixture
$\mathbf{u}$	molar average mixture velocity
$[W]$	matrix of total diffusion coefficients defined by eqn (20)
x_i	mole fraction of i in mixture

Greek symbols

$[\beta]$	bootstrap matrix with elements given by eqn (10)
γ_i	activity coefficient of species i in solution
$[\Gamma]$	matrix of thermodynamic factors with elements given by

$$\Gamma_{ij} = \delta_{ij} + \frac{x_i \partial \ln \gamma_i}{x_j \partial \ln x_j}, \; i,j = 1, 2, \ldots n-1$$

δ	film thickness
δ_{ij}	Kronecker delta
λ_i	coefficients in linear dependence relation (8)
μ_i	molar chemical potential of species i in mixture
ϕ_i'	pseudo-binary dimensionless mass transfer rate factor

Matrix notation

( )	column matrix with $n-1$ elements
[ ]	$n-1$ dimensional square matrix
$[\ ]^{-1}$	$n-1$ dimensional inverted square matrix
$\lceil\ \rfloor$	diagonal matrix with $n-1$ non-zero elements

Superscript

x	pertaining to liquid phase in vapour–liquid transfer
y	pertaining to vapour phase in vapour–liquid transfer
•	corresponding to conditions of finite transfer mass transfer rates
′	pseudo-binary parameter

Subscripts

a	coefficient evaluated at average composition
n	pertaining to nth species
o	parameter evaluated at position $r = r_o$
t	pertaining to mixture
δ	parameter evaluated at position $r = \delta$

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Enhancement factor in gas–liquid reactions in presence of significant gas side mass transfer resistence for a second order reaction†

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The relationship between the enhancement factor E for gas liquid reactions and various experimental conditions (hydrodynamic and kinetic) has been investigated extensively for a variety of chemical reactions, as summarised by Astarita[1] and Danckwerts[3]. Here we shall restrict our attention to the second order reaction

$$A(g) + zB(l) \rightarrow \text{products} \qquad (1)$$

with the intrinsic kinetics

$$-r = kAB. \qquad (2)$$

For this case, the computation of the numerical value of the enhancement factor is quite straightforward if the absorption process is liquid phase controlled, i.e. if the resistance to mass transfer lies entirely in the liquid phase. For cases where appreciable gas phase resistance is present, an iterative procedure has been suggested for calculating the rate of gas absorption[3] which is ideally suited for machine computation. In the present paper, a new procedure is evolved for the latter case which makes it possible to calculate the gas absorption rate in a straightforward manner through the use of a single generalised graph. Such a procedure is valuable for a quick calculation of the gas absorption rates without recourse to machine computations.

A brief outline of the iterative procedure is as follows. The concentration and the partial pressure of A at the gas liquid interface are related through Henry's law as

$$P_i = HA_i \qquad (3)$$

where H is the Henry's law constant. Equating the transport of A across both the films we get the absorption flux as

$$\bar{R} = k_G(P_b - P_i) = k_L A_i E. \qquad (4)$$

The enhancement factor on the liquid side, E, is a known function of E_i and M, and may be expressed as

$$E = E(E_i, M) \qquad (5)$$

where

$$E_i = 1 + D_B B_0/(zD_A A_i) \qquad (6)$$

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