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Rajamani Krishna† [a]
[a] Department of Chemical Engineering, University of Manchester Institute of Science and Technology, Manchester, England.

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A SIMPLIFIED FILM MODEL DESCRIPTION OF MULTICOMPONENT INTERPHASE MASS TRANSFER

RAJAMANI KRISHNA†

Department of Chemical Engineering,
University of Manchester Institute of Science and Technology, Manchester, England.

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Within the framework of the film model description, the interphase mass transfer process in n-component systems is examined and a modified definition for interphase transfer coefficients, based solely on the Generalized Maxwell-Stefan diffusion coefficients, is suggested. The persuasive reasons for the use of these coefficients are shown to be a much simplified additivity of mass transfer resistances formula, freedom from dependence on system thermodynamic non-idealities and the promise of predictability of the coefficients from corresponding binary system parameters. Some consequences of the analysis on fluid-fluid mass transfer operations such as distillation are considered.

It is now well appreciated that the Generalized Maxwell-Stefan (GMS) diffusion equations afford a convenient and correct description of molecular diffusion phenomena in the general case of non-ideal n component fluid mixtures (Lightfoot, *et al.*, 1962; Lightfoot, 1974; Slattery, 1972). In the absence of pressure and thermal gradients and external body forces, these equations may be written as

$$\frac{x_i}{RT} \nabla \mu_i = \sum_{\substack{j=1 \\ j \neq i}}^{n} \frac{x_i \mathbf{N}_j - x_j \mathbf{N}_i}{c \mathcal{D}_{ij}}, \quad i = 1, 2, \ldots n \tag{1}$$

where x_i is the mole fraction of i in the fluid mixture; c is the total molar concentration; $\mathcal{D}_{ij}$ $(i \neq j, \mathrm{i,j} = 1, 2, \ldots, n)$ represents the GMS diffusion coefficients; $\mathbf{N}_i$ is the molar flux of species i with respect to a stationary coordinate frame of reference:

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

In view of the Gibbs-Duhem equation, only $n - 1$ of the Eqs. (1) are independent and therefore an additional relation is required for the calculation of the n molar fluxes $\mathbf{N}_i$. This additional relationship usually takes the form of a linear dependence between the n fluxes $\mathbf{N}_i$ (Krishna and Standart, 1976a)

$$\sum_{i=1}^{n} \lambda_i \mathbf{N}_i = 0 \tag{3}$$

where λ_i constant coefficients appear in the linear dependence.

† Present Address: Koninklijke/Shell-Laboratorium, Amsterdam, Badjuisweg 3, Amsterdam-Noord, The Netherlands.

Equations (1) and (3) provide a complete description of the diffusion process and are sufficient for the calculation of the fluxes N_i, required in design of mass transfer processes. In the following we consider steady-state diffusion across a film of thickness δ, the basis of the film model. By defining a matrix of thermodynamic factors $[\Gamma]$ with elements given by

$$\Gamma_{ij} = \delta_{ij} + \frac{x_i}{x_j} \cdot \frac{\partial \ln \gamma_i}{\partial \ln x_j}, \quad i,j = 1, 2, \ldots, n-1 \tag{4}$$

and a matrix of mass transfer rate factors $[\Phi]$:

$$\Phi_{ii} = \frac{N_i}{\mathscr{k}_{in}} + \sum_{\substack{k=1 \\ k \neq i}}^{n} \frac{N_k}{\mathscr{k}_{ik}}, \quad i = 1, 2, \ldots, n-1 \tag{5}$$

$$\Phi_{ij} = -N_i(1/\mathscr{k}_{ij} - 1/\mathscr{k}_{in}), \quad \underset{i \neq j}{i,j} = 1, 2, \ldots, n-1 \tag{6}$$

where

$$\mathscr{k}_{ij} = c\mathscr{D}_{ij}/\delta, \quad \underset{i \neq j}{i,j} = 1, 2, \ldots, n \tag{7}$$

we may write Eqs. (1) in $n-1$ dimensional matrix notation as (Krishna, 1977)

$$[\Gamma] \frac{\mathrm{d}(x)}{\mathrm{d}\eta} = [\Phi]\,(x) + (\zeta) \tag{8}$$

where the column matrix (ζ) has the elements

$$\zeta_i = -N_i/\mathscr{k}_{in}, \quad i = 1, 2, \ldots, n-1 \tag{9}$$

The set of coupled differential equations (8) may be linearlzed by assuming that the thermodynamic factors Γ_{ij} and the binary mass transfer parameters $\mathscr{k}_{ij}$ are independent of composition (Krishna, 1977); with this assumption the $n-1$ dimensional *linear* matrix differential equation (8) can be solved for the boundary conditions

$$\eta = 0, (x) = (x_b), \text{ the bulk fluid compositions} \tag{10}$$

$$\eta = 1, (x) = (x_I), \text{ the interfacial fluid composition} \tag{11}$$

to give an explicit expression for the fluxes N_i:

$$(N) = [\beta]\,[B_b]^{-1}\,[\Gamma]\,[\Theta]\,\{\exp\,[\Theta] - \ulcorner I \lrcorner\}^{-1}(x_b - x_I) \tag{12}$$

where $[\beta]$ is a matrix of determinancy factors; $[\Theta]$ is given by

$$[\Theta] \equiv [\Gamma]^{-1}[\Phi]; \tag{13}$$

$[B_b]$ is a matrix of inverted binary mass transfer coefficients:

$$B_{bii} = \frac{x_{bi}}{\mathscr{k}_{in}} + \sum_{\substack{k=1 \\ k \neq i}}^{n} \frac{x_{bk}}{\mathscr{k}_{ik}}, \quad i = 1, 2, \ldots, n-1 \tag{14}$$

$$B_{bij} = x_{bi}(1/\mathscr{k}_{ij} - 1/\mathscr{k}_{in}), \quad \underset{i \neq j}{i,j} = 1, 2, \ldots, n-1 \tag{15}$$

In the literature on multicomponent mass transfer (Krishna and Standart, 1976b; Stewart, 1973; Stewart and Prober, 1964; Toor, 1964), it is commonly accepted that the proper definition of the mass transfer coefficients is in terms of the diffusive fluxes J_i:

$$J_i \equiv N_i - x_i \sum_{k=1}^{n} N_i = N_i - x_i N_t, \; i = 1, 2, \ldots, n \tag{16}$$

Thus if, by analogy to the binary system (Bird, Stewart and Lightfoot, 1960), we define multicomponent zero-flux mass transfer coefficients based on the diffusion fluxes in the bulk fluid phase:

$$k_{bij} = \lim_{\text{all } N_i \to 0} \left(\frac{J_{bi}}{x_{bj} - x_{1j}} \right), \; i,j = 1, 2, \ldots, n-1 \tag{17}$$

then the film model yields in view of Eq. (12) the following expression for the matrix of mass transfer coefficients:

$$[k_b] = [B_b]^{-1}[\Gamma] \tag{18}$$

A major drawback of the definition of the matrix of mass transfer coefficients $[k_b]$ is that these coefficients are directly dependent on the system thermodynamics via the thermodynamic factors Γ_{ij}. Thus even if we possess information on the GMS diffusion coefficients $\mathscr{D}_{ij}$, it is not sufficient to be able to calculate k_{bij}; we need additional information on the variation of the activity coefficients with composition of the mixture. The drawback in the definition of $[k_b]$ is exactly analogous to the shortcomings of the Fick's law diffusivity D_{12} in describing binary diffusion. Thus, as shown by Vignes (1966) and Leffler and Cullinan (1970), the GMS diffusion coefficient $\mathscr{D}_{12}$ shows a much more predictable composition dependence than the Fick's law diffusivity. Further, the thermodynamic factor Γ_{ij}, which for a binary system reduces to

$$\Gamma_{11} = 1 + \frac{\partial \ln \gamma_1}{\partial \ln x_1}, \tag{19}$$

accounts for a major portion of the composition dependence of D_{12} and

$$Đ_{12} = D_{12}/\Gamma_{11} = D_{12}/(1 + \partial \ln \gamma_1/\partial \ln x_1) \tag{20}$$

can be related simply to the infinite dilution diffusion coefficients. Vignes (1966) suggests the following empirical relationship

$$Đ_{12} = (D_{12}^0)^{x_2} \quad (D_{21}^0)^{x_1} \tag{21}$$

where D_{12}^0 is the diffusion coefficient of species 1 infinitely diluted in the mixture of 1 and 2; D_{21}^0 is the diffusion coefficient of 2 infinitely diluted in the corresponding mixture. Equation (21) suggests that a plot of log $Đ_{12}$ versus mole fraction x_1 should yield a straight line and in the cases examined by Vignes this is indede found to be the case.

In view of the success achieved for a binary system by separating the thermodynamic (Γ_{11}) and the kinetic ($Đ_{12}$) factors, one may expect that a similar separation for multicomponent systems would be fruitful; the recent work of Kosanovich and Cullinan (1976) with a quaternary system suggests that this expectation may be fulfilled in many cases.

These developments in the description of liquid diffusion open up the question whether the coefficients k_{bij}, defined by Eqs. (17), are the correct ones for interphase mass transfer. Should we not follow a complete parallel treatment to the corresponding diffusion problem and define the matrix of mass transfer coefficients as

$$[\kappa_b] \equiv [B_b]^{-1} \tag{22}$$

and thus make these coefficients completely free of thermodynamic non-ideality effects? The definition (22) would allow the calculation of the multicomponent κ_{bij} from the GMS coefficients (see Eqs. (7), (14) and (15)). In addition to this advantage, we will show in the following analysis that the coefficients κ_{bij} have other features which suggest their acceptance, in place of the k_{bij}, in describing correctly interphase transport behaviour

Let us consider the specific example of mass transfer in a vapour-liquid system; the analysis should therefore be useful in applications such as distillation, condensation, absorption, etc. We assume that transfer from the liquid phase to the vapour phase yields a positive value for the flux N_i. Further we denote liquid phase mole fractions by x_i and vapour phase mole fractions by y_i. A subscript (x or y) is added to each of the parameters introduced earlier in the paper to distinguish between the corresponding phase values. The interfacial fluxes N_i are, however, phase invariants; for the vapour phase they will be given by Eq. (12) with the simplification that the matrix $[\Gamma]$ reduces to the identity matrix:

$$(N) = [\beta_y]\,[B_{by}]^{-1}\,[\Xi_y]\,(y_I - y_b) \tag{23}$$

Here $[\Xi_y]$ denotes the matrix of correction factors for high mass transfer rates; this matrix takes the form

$$[\Xi_y] = [\Phi_y] \exp [\Phi_y] \{\exp [\Phi_y] - \ulcorner I \lrcorner\}^{-1} \tag{24}$$

Similarly for the liquid phase we can write

$$(N) = [\beta_x] [B_{bx}]^{-1} [\Gamma] [\Xi_x] (x_b - x_l) \tag{25}$$

where the matrix of correction factors for the liquid phase is given by

$$[\Xi_x] = [\Theta_x] \{\exp [\Theta_x] - \ulcorner I \lrcorner\}^{-1} \tag{26}$$

It is common in chemical engineering to assume that equilibrium prevails at the interface. In order to be able to add the resistances in the vapour and liquid phases it is necessary to linearize the vapour-liquid equilibrium relationship. Thus we may write

$$(y_l) = [M] (x_l) + (b) \tag{27}$$

where (b) is a column matrix of 'intercepts'; $[M]$ is the matrix of coefficients for the linearized relationship:

$$M_{ij} = \partial y_i / \partial x_j, \quad i, j = 1, 2, \ldots, n-1 \tag{28}$$

We use an $n - 1$ dimensional linearized relationship (27) in view of the fact that there are only $n - 1$ independent mole fractions.

For equilibrium between vapour and liquid phases we have on equating the fugacities of each component in either phase:

$$\phi_i y_i P = \gamma_i x_i P_i^s, \quad i = 1, 2, \ldots, n \tag{29}$$

where P is the total pressure in the system; P_i^s is the saturation vapour pressure of component i; ϕ_i is the fugaticity coefficient in the vapour phase accounting for vapour phase non-ideality; γ_i is the activity coefficient for component i in the liquid mixture and is a function of the intensive state variables: composition and temperature

$$\gamma_i = \gamma_i(x_1, x_2, \ldots, x_{n-1}, T), \quad i = 1, 2, \ldots, n \tag{30}$$

where we consider only $n - 1$ independent x_i.

From Eqs. (28)-(30) we obtain

$$M_{ij} = \frac{\partial y_i}{\partial x_j} = \frac{\partial(\gamma_i x_i)}{\partial x_j} \frac{P_i^s}{\phi_i P}$$

$$= x_i \frac{\partial \ln(\gamma_i x_i)}{\partial x_j} \frac{\gamma_i P_i^s}{\phi_i P} = \Gamma_{ij} K_{eqi}, \quad i, j = 1, 2, \ldots, n-1 \tag{31}$$

where we have used K_{eqi} to denote the equilibrium distribution ratio or K value:

$$K_{\mathrm{eq}i} = \frac{\gamma_i P_i^s}{\phi_i P}, \quad i = 1, 2, \ldots, n \tag{32}$$

The activity coefficient in Eq. (32) will have to be evaluated at some suitably averaged composition x_i between the bulk fluid and interface values.

It is interesting to note that the matrix of thermodynamic factors Γ_{ij}, introduced first in Eq. (4) in connection with intraphase diffusion, also plays a role in describing the (linearized) phase equilibria.

If we define a matrix of overall vapour phase mass transfer coefficients $[\kappa_{\mathrm{by0}}]$ ($\equiv [B_{\mathrm{by0}}]^{-1}$) by the interphase rate relationship

$$(N) = [\beta_{\mathrm{y}}][B_{\mathrm{by0}}]^{-1}[\Xi_{\mathrm{y}}](y^* - y_{\mathrm{b}})$$

where (y^*) is a matrix of compositions in equilibrium with the bulk liquid compositions (x_{b}), then the formula for addition of interphase mass transfer resistances can be derived as (Krishna and Standart, 1976a; Toor, 1964):

$$([\beta_{\mathrm{y}}][B_{\mathrm{by0}}]^{-1}[\Xi_{\mathrm{y}}])^{-1} =$$

$$([\beta_{\mathrm{y}}][B_{\mathrm{by}}]^{-1}[\Xi_{\mathrm{y}}])^{-1} + \ulcorner K_{\mathrm{eq}} \lrcorner [\Gamma]\,([\beta_{\mathrm{x}}][B_{\mathrm{bx}}]^{-1}[\Gamma][\Xi_{\mathrm{x}}])^{-1} \tag{34}$$

For distillation operations it is commonly assumed that equimolar counter diffusion prevails and therefore the determinancy matrices $[\beta]$ reduce to $\ulcorner I \lrcorner$, the identity matrix, in both phases. Further, for this case the matrices of correction factors $[\Xi]$ may also be approximated by the identity matrix. With these two simplifications, Eq. (34) reduces to

$$[B_{\mathrm{by0}}] = [B_{\mathrm{by}}] + \ulcorner K_{\mathrm{eq}} \lrcorner [B_{\mathrm{bx}}] \tag{35}$$

which is a remarkable relationship because of the absence of the thermodynamic factor $[\Gamma]$. The matrices $[B_{\mathrm{b}}]$, which may be termed mass transfer resistance matrices, possess a very simple additive property; the vapour and liquid resistances being connected via the equilibrium K values. Since the K values are easily determined, the total resistance $[B_{\mathrm{by0}}]$ can be calculated from the individual phase resistances. For prediction purposes, it is important to note that the coefficients $B_{\mathrm{by}ij}$ and $B_{\mathrm{bx}ij}$ can be related to binary transport coefficients in the fluid phases and are largely free from thermodynamic non-ideality factors (Kosanovich and Cullinan, 1976). In terms of the mass transfer coefficients $\kappa_{\mathrm{b}ij}$ we may write

$$[\kappa_{\mathrm{by0}}]^{-1} = [\kappa_{\mathrm{by}}]^{-1} + \ulcorner K_{\mathrm{eq}} \lrcorner [\kappa_{\mathrm{bx}}]^{-1} \tag{36}$$

In terms of the more conventionally used mass transfer coefficients $k_{\mathrm{b}ij}$ we obtain the addition of resistances formula (Krishna and Standart, 1976a; Toor, 1964):

$$[k_{\mathrm{by0}}]^{-1} = [k_{\mathrm{by}}]^{-1} + \ulcorner K_{\mathrm{eq}} \lrcorner [\Gamma]\,[k_{\mathrm{bx}}]^{-1} \tag{37}$$

which is somewhat more involved than Eq. (35) because the coefficients $k_{\mathrm{bx}ij}$ depend on both transport and thermodynamic system properties and the system hydrodynamics.

For mass transfer on a tray, we define the number of transfer units in either fluid phase and the overall unit as

$$\mathrm{NTU}_{\mathrm{y}ij} \equiv \frac{\kappa_{\mathrm{by}ij}\mathscr{a}v}{G};\ \mathrm{NTU}_{\mathrm{x}ij} \equiv \frac{\kappa_{\mathrm{bx}ij}\mathscr{a}v}{L};\ \mathrm{NTU}_{\mathrm{y0}ij} \equiv \frac{\kappa_{\mathrm{by0}ij}\mathscr{a}v}{G} \tag{38}$$

where $\mathscr{a}$ is the interfacial area per unit volume of dispersion; v is the total volume of dispersion on the tray; G is the molar flow rate of the vapour upwards through the dispersion; L is the molar flow of the liquid. The additivity formula in terms of the NTUs can be derived as

$$[\mathrm{NTU}_{\mathrm{y0}}]^{-1} = [\mathrm{NTU}_{\mathrm{y}}]^{-1} + \ulcorner S \lrcorner [\mathrm{NTU}_{\mathrm{x}}]^{-1} \tag{39}$$

where $\ulcorner S \lrcorner$ represents a diagonal matrix of stripping factors;

$$S_i = K_{\mathrm{eq}i}\,\frac{G}{L},\ i = 1, 2, \ldots, n-1 \tag{40}$$

The calculation of the overall number of transfer units, necessary for the calculation of the distillation efficiencies, is highly simplified with the use of the $\kappa_{\mathrm{b}ij}$ coefficients.

For vapour-liquid transfer in packed columns, we generally work in terms of the heights of transfer units (HTU). We define the HTU for vapour phase transfer as

$$[\mathrm{HTU}_{\mathrm{y}}] \equiv [\kappa_{\mathrm{by}}]^{-1} aA/G = [B_{\mathrm{by}}]\,aA/G \tag{41}$$

where a is the interfacial area per unit volume of packed column; A is the cross sectional area of column. The definitions of the corresponding heights in the liquid phase and the overall set may be written in analogous manner. With these definitions the additivity formula takes the form

$$[\mathrm{HTU}_{0\mathrm{y}}] = [\mathrm{HTU}_{\mathrm{y}}] + \ulcorner S \lrcorner [\mathrm{HTU}_{\mathrm{x}}] \tag{42}$$

It is clear from Eqs. (39) and (42) that in a multicomponent mixture, the distribution of resistances between the liquid and vapour phases can be different for each component. Thus in a ternary mixture, for example, we may write

$$\mathrm{HTU}_{0y11} = \mathrm{HTU}_{y11} + S_1\mathrm{HTU}_{x11} \tag{43}$$

$$\mathrm{HTU}_{0y12} = \mathrm{HTU}_{y12} + S_1\mathrm{HTU}_{x12} \tag{44}$$

$$\mathrm{HTU}_{0y21} = \mathrm{HTU}_{y21} + S_2\mathrm{HTU}_{x21} \tag{45}$$

$$\mathrm{HTU}_{0y22} = \mathrm{HTU}_{y22} + S_2\mathrm{HTU}_{x22} \tag{46}$$

and therefore if component 1 is very much more volatile in comparison to component 2:

$$S_1 \gg S_2 \tag{47}$$

then the transfer of component 1 will be liquid phase controlled and that of component 2 will be vapour phase controlled.

In the spirit of Eqs. (14) and (15), we may also relate the multicomponent heights of transfer units to the corresponding binary pairs as

$$\mathrm{HTU}_{xii} = x_{bi}HTU_{xin} + \sum_{\substack{k=1\\k\neq i}}^{n} x_{bk}HTU_{xik}, \quad i = 1, 2, \ldots, n-1 \tag{48}$$

and

$$\mathrm{HTU}_{xij} = -x_{bi}(HTU_{xij} - HTU_{xin}), \quad \underset{i\neq j}{i, j} = 1, 2, \ldots, n-1 \tag{49}$$

where the HTU_xs refer to the heights of transfer unit of the corresponding binary pairs, these units being defined in terms of the mass transfer coefficients $\mathscr{k}_{xij}$. If the HTUs are defined in terms of the transfer coefficients k_{bij}, then the simple predictive relations (48) and (49) do not follow because of the need to take the thermodynamic factors separately into account.

The general conclusion to be drawn from the analysis presented in this communication is that the simplest description of interphase mass transfer is obtained in terms of the coefficients κ_{bij}, or equivalently the B_{bij}, defined in terms of the Generalized Maxwell-Stefan diffusion coefficients. The multicomponent transport parameters can be estimated from the corresponding binary ones with relations such as Eqs. (14) and (15) or (48) and (49). The logical and consistent way to estimate the constituent binary parameters such as HTU_{ij} and $\mathscr{k}_{ij}$ is to use published binary mass transfer correlations with the multicomponent $\mathscr{D}_{ij}$ in place of the binary D_{ij}.

In the spirit of Eq. (17), we may formally define our recommended mass transfer coefficients κ_{bij} as

$$\kappa_{bij} = \lim_{\substack{\text{all } N_i \to 0;\\ \Gamma_{ij} \to \delta_{ij}}} \left(\frac{J_{bi}}{x_{bj} - x_{Ij}}\right), \quad \mathrm{i, j} = 1, 2, \ldots, n-1$$

LIST OF SYMBOLS

a	interfacial area per unit volume of packed column
$\mathscr{a}$	interfacial area per unit volume of dispersion on tray
A	cross sectional area of packed column
$[B]$	matrix of mass transfer resistances
(b)	column matrix of intercepts
c_i	molar density of species i in mixture
c	total mixture molar density
$\mathscr{D}_{ij}$	Generalized Maxwell-Stefan diffusion coefficient
D_{ij}	Fick's law diffusion coefficient in the binary pair $i-j$
D_{ij}^0	Fick's law diffusion coefficient for i infinitely dilute in j
G	total flow rate of vapour mixture
HTU	height of transfer unit for binary distillation
[HTU]	matrix of heights of transfer units for multicomponent distillation
$\ulcorner I \lrcorner$	identity matrix
J_i	diffusion flux of species i relative to molar average mixture velocity
$[k]$	matrix of multicomponent mass transfer coefficients
$\mathscr{k}_{ij}$	mass transfer coefficient of binary pair $i-j$
K_{eqi}	equilibrium distribution ration, K value for species i
L	molar flow rate of liquid mixture
$[M]$	matrix of coefficients in linearized equilibrium relationship
n	number of components in mixture
N_i	molar flux of species i relative to stationary coordinate frame of reference
N_t	mixture molar flux relative to stationary coordinate frame of reference
[NTU]	matrix of numbers of transfer units
P	total system pressure
P_i^s	saturation (vapour) pressure of component i

LIST OF SYMBOLS–*continued*

R	gas constant
S_i	stripping factor for component i
T	absolute temperature
u_i	diffusion velocity of component i
v	volume of dispersion on tray
x_i	mole fraction in fluid mixture; specifically in interphase transfer it refers to the liquid phase
y_i	mole fraction in vapour phase

Greek Symbols

$[\beta]$	determinancy matrix for calculation of n fluxes N_i from $n-1$ independent fluxes J_i
γ_i	activity coefficient of species i in solution
$[\Gamma]$	matrix of thermodynamic factors
δ	thickness of film, stagnant layer adjacent to the interface
δ_{ij}	Kronecker delta
ζ_i	elements given by Eq. (9)
η	dimensionless distance coordinate, normalized with respect to the film thickness
$[\Theta]$	matrix defined by Eq. (13)
$[\kappa]$	matrix of (recommended) mass transfer coefficients
λ_i	coefficients in linear dependence relationship for the N_i
μ_i	molar chemical potential of species i
$[\Xi]$	matrix of correction factors for finite mass transfers
$[\Phi]$	matrix of mass transfer rate factors, defined by Eqs. (5) and (6)
ϕ_i	vapour phase fugacity coefficient for species i

Operational Symbols

∇	gradient operator
Σ	summation

LIST OF SYMBOLS–*continued*

Matrix Notation

( )	column matrix with $n-1$ elements
[ ]	matrix of dimension $(n-1)\times(n-1)$
⌜ ⌟	diagonal matrix with $n-1$ non-zero elements

Subscripts

b	bulk phase property
I	interfacial property or parameter
n	pertaining to n-th component
0	overall coefficient
t	pertaining to total mixture
x	pertaining to liquid phase
y	pertaining to vapour phase

Superscripts

*	equilibrium value
0	infinite dilution value

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