# A SIMPLIFIED MASS TRANSFER ANALYSIS FOR MULTICOMPONENT CONDENSATION

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### ABSTRACT

This paper considers the problem of calculating the condensation fluxes N; for n-component vapour mixtures, taking account of diffusional interactions in the vapour phase. A simplified method for the flux calculation is suggested requiring the assumption that the matrix of multicomponent mass transfer coefficients [W], defined by the n-1 dimensional matrix relation (N) = [W]( $\Delta x$ ), is constant along the diffusion path. This assumption allows the calculation of the steadystate fluxes N; in an explicit manner without iterations. For conditions of equimolar diffusion, the simplified method developed here coincides with the linearized theory development of Toor and Stewart and Prober. For other cases involving diffusion through an inert gas, for example, the method suggested here is simpler than the Toor-Stewart Prober approach. The accuracy of the simplified method is demonstrated with a few typical examples involving mass transfer in a ternary gas phase. Though condensation in the presence of an inert gas has been treated explicitly here, the developed simplified method should find application in other mass transfer processes such as distillation, absorption and extraction.

## Introduction

The analysis of the mass transfer process during condensation, or evaporation, has been considered by many authors |1 - 32|. The classic works of Ackermann |1| and Colburn, Drew and Hougen |6 - 7| considered the problem of binary vapour condensation. Extensions of the binary analysis to multicomponent mixtures have proceeded in two directions. One set of

+ Current Address: Koninklijke/Shell-Laboratorium, Amsterdam (Shell Research B V) Badhuisweg 3, 1031 CM Amsterdam-N, The Netherlands workers have adopted the pseudo-binary approach to describe multicomponent condensation |18,21,25,26|. In this approach the condensing flux of any component is taken to proportional to its own, intrinsic, driving force. The other set of workers |2,3,8-17,22-24| have properly recognised the characteristics of multicomponent mass transfer processes, described in the pioneering papers of Toor 32 and Stewart and Prober 29, and have modelled the vapour mass transfer process taking account of diffusional coupling effects. Design procedures for multicomponent condensers have been developed using coupled mass transfer rate relations [2,3,9,10,24]. Comparison of available experimental data with such rigorous design procedures |2,3,15| has shown the definite superiority of the rigorous multicomponent models over simpler pseudo-binary models. The work of Bandrowski and Kubaczka 3 has shown that though the model based on an exact matrix method of solution to the Maxwell-Stefan diffusion equations [8,12,13] is statistically superior to the Toor-Stewart-Prober approach, the differences between these two multicomponent models are not significant. Our earlier work, in which we used the ternary mass transfer data of Modine |15| to test various mass transfer models also showed that the Krishna et al approach [8-13] and the Toor-Stewart-Prober methods were of comparable accuracy and decidedly better than a pseudo-binary approach.

The general conclusion to be drawn from the above studies is that the analysis of multicomponent condensation mass transfer must take into account the diffusional coupling effects in the vapour phase. We have earlier shown that coupling effects could be important during condensation of rich vapour mixtures and when the components in the vapour phase differ considerably in molecular size and nature |16|. The logical follow-up is to incorporate these multicomponent mass transfer models in routine design procedures for condensation equipment. One possible stumbling block in the realization of this objective is the fact that the multicomponent mass transfer models involve iterative calculations at any differential position in the condenser. Such iterations are required because the interphase mass transfer coefficients are themselves functions of the interfacial condensation rates: we have to correct the 'zero' flux mass transfer coefficients for high mass transfer rates (see Bird, Stewart and Lightfoot [4] for an excellent discussion on this subject). It would therefore be desirable to develop a simplified method for routine engineering design which would be truly explicit in the mass transfer flux calculation, whilst retaining the essence of the problem, namely coupled diffusion effects. This is the broad

objective of the current communication.

# Analysis

For steady-state diffusion in n-component mixtures, the differential equations of continuity reduce to

$$\nabla \bullet \mathbf{N}_{i} = 0 \tag{1}$$

where  $N_i$  represent the fluxes in a stationary coordinate frame of reference. For the most general case of non-ideal fluid mixtures the constitutive relations for the fluxes are given by the Generalized Maxwell-Stefan (GMS) diffusion equations |12|

$$\frac{x_{i}}{RT} \nabla \mu_{i} = \sum_{j=1}^{n-1} \frac{x_{i} N_{j} - x_{j} N_{i}}{c \cdot \theta_{ij}}, \quad i = 1, 2, \dots n$$
(2)

where  $x_i$  represents the constituent mole fraction;  $\mu_i$  is the molar chemical potential of i;  $\vartheta_{ij}$  is the GMS diffusion coefficient of the pair i-j and c is the total mixture molar concentration. In view of the Gibbs-Duhem relation all the n chemical potential gradients are not independent and therefore there are only n-1 independent relations (2). In order to allow calculation of the n interfacial fluxes N<sub>i</sub> we need an additional relationship. For condensation of n-1 vapour species in the presence of an inert gas the additional restriction is

$$N_{n} = 0 \tag{3}$$

making the problem determinate. More general relationships between the fluxes have been considered elsewhere |10,12,14|.

In addition to the fluxes  $\rm N_i$  above, it is customary to define diffusion fluxes  $\rm J_i$  with respect to the molar average mixture velocity:

$$J_{i} \equiv N_{i} - x_{i} \sum_{j=1}^{n} N_{i} = N_{i} - x_{i} N_{t}$$
(4)

where  $N_t$  is the mixture flux in the stationary coordinate reference frame. It is easy to derive the following relation between the fluxes  $N_i$  and  $J_i$  (see reference |12|):

 $(N) = [\beta](J)$ (5)

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where  $[\beta]$  is the bootstrap matrix. It is easy to appreciate from equations (4) and (5) that both  $J_i$  and  $\beta_{ij}$  wary along the diffusion path from the bulk phase to the interface. On the other hand, for diffusion across plane interfaces, the fluxes  $N_i$  are invariant along the diffusion path.

Equations (1), (2) and (4) may be combined to give the following expressions for  $\rm N_i$  and  $\rm J_i$ 

$$(J) = -c [B]^{-1} [\Gamma] (\nabla x) \equiv -c [D] (\nabla x)$$
(6)

and

$$(N) = -c [\beta] [B]^{-1} [\Gamma] (\nabla x)$$
(7)

where [B] is the matrix of inverted diffusion coefficients,  $[\Gamma]$  is the matrix of thermodynamic factors and [D] is the matrix of generalized Fick's law diffusion coefficients. For detailed definitions of these quantities see Table 1 and our earlier published work |12|.

For ideal gas mixtures the matrix of thermodynamic factors reduces to the identity matrix:

 $\Gamma_{i,i} = \delta_{i,i}$  (ideal gas mixture) (8)

and therefore for ideal gas mixtures the matrix of Fick's law diffusivities is given by

 $[D] \equiv [B]^{-1}$  (ideal gas mixture) (9)

and is a function of composition.

In the published literature there are basically two approaches to the integration of the GMS diffusion equations (2) to yield the fluxes  $N_i$ , given by equation (7). We call these Models I and II respectively. In Model I, described by Krishna |12|, we make the following assumptions

i)  $\Gamma_{ij}$  is constant along the diffusion path. This assumption is exact for ideal gas mixtures (see equation (8))

ii) c  $\theta_{ij}$  is also constant along the diffusion path. This assumption is also exactly true for ideal gas mixtures under isothermal conditions.

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The procedure for calculating the fluxes  $N_i$  with the aid of these two assumptions is detailed in Table 1. As is evident from the details given in this Table, a trial and error procedure is involved. Convergence is however rapid as has been reported earlier |8,11|.

In the other method for calculating the fluxes, due to Toor (1964) and Stewart and Prober (1964), we make the following assumption

i) c [D] is constant along the diffusion path. It must be noted that this assumption is not strictly true even for ideal gas mixtures. The advantage of Method I is that the procedure is exact for ideal gas mixtures.

The procedure for calculating the fluxes  $N_i$  with the aid of the above assumption of Method II is detailed in Table 2. Again it is evident that a trial and error procedure is involved in the calculation of the fluxes.

Equation (7) provides us with the key assumption to be explored in the current work. Let us denote [W] as the mass transfer coefficient

$$\begin{bmatrix} W \end{bmatrix} \equiv \frac{c}{\delta} \begin{bmatrix} \beta \end{bmatrix} \begin{bmatrix} B \end{bmatrix}^{-1} \begin{bmatrix} \Gamma \end{bmatrix} = \frac{c}{\delta} \begin{bmatrix} \beta \end{bmatrix} \begin{bmatrix} D \end{bmatrix}$$
(10)

If we assume [W] constant along the diffusion path by suitably averaging the elements  $\beta_{ij}$ ,  $B_{ij}$  and  $\Gamma_{ij}$ , then the equation (7) may be directly integrated along the film of thickness  $\delta$  to yield the fluxes N<sub>i</sub> as

$$(N) = [N] (x_0 - x_{\delta})$$
(11)

where  $x_{i0}$  and  $x_{i\delta}$  are the mole fractions at the interface and bulk phase respectively. It is clear that the assumption of constant [W], gives an explicit procedure for calculation of N<sub>i</sub>. The detailed procedure for this Method III is given in Table 3.

If conditions of equimolar diffusion prevail  $(N_t = 0)$ , then the assumption of [N] constant is the same as the assumption of c [D]constant, the basis of the Toor-Stewart-Prober approach; the Methods II and III collapse into each other for this case.

Table 4 compares the predictions of the Methods I, II and III for three illustrative examples A,B and C involving diffusion of two components in the vapour phase in the presence of an non-transferring component ( $N_3 = 0$ ). For these cases the Method I is exact and serves as a standard for comparison.

In the illustrative example A, the binary pair diffusivities are close to each other and the diffusional interactions are weak. All three methods give answers close to one another. It must be remarked that the pseudo-binary approach to this example A will also lead to reasonably accurate predictions.

Examples B and C involve large diffusional interactions. It is interesting to note that even for this case the simplified method III suggested in the current work is able to provide answers close the exact Method I. The predictions of the Toor-Stewart-Prober Method II are closer to the exact Method I than the predictions of Method III, but the differences between Methods II and III are small.

# Concluding Remarks

We have developed a simplified procedure for the calculation of mass transfer fluxes during condensation. The suggested simplified procedure relies on the assumption that the matrix of mass transfer coefficients defined by equation (10), or (11), is a constant along the diffusion path. Calculations with ternary gas mixtures, both for weak and strong diffusion coupling, have shown that the predictions of the simplified procedure are very close to the exact values. The major advantage of the simplified procedure is that no iterations are involved while retaining the essential feature of multicomponent mass transport phenomena, namely the diffusional coupling effect.

The simplified procedure can be easily incorporated into design programs for condensers even for mixtures involving more than 3 components. It does seem necessary to test the procedure further with experimental data.

The method developed here should also be applicable to

- i) evaporation into gas streams
- ii) non-equimolar distillation (see reference [14])
- iii) multicomponent gas absorption
  - iv) multicomponent mass transfer with heterogeneous chemical reaction
  - v) multicomponent extraction.

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### TABLE 1. MODEL I: Multicomponent Film Model due to Krishna (1977)

- 1. Estimate the Generalized Maxwell-Stefan (GMS) diffusion coefficients  $D_{ij}$ . For ideal gas mixtures these coefficients can be estimated from the kinetic theory; for non-ideal liquid mixtures a procedure such as that suggested by Vignes (1966) is appropriate.
- 2. Calculate the mass transfer coefficients  $k_{ij}$  of the pair i-j in the multicomponent mixture:  $k_{ij} = c \cdot \vartheta_{ij}/\imath$ ; where  $\imath$  is the characteristic length in the film model. This length is given by  $\imath = \imath$ , the film thickness for planar films;  $\imath = r_0 \ln(r_\delta/r_0)$  for cylindrical films and  $\imath = r_0 (1 - r_0/r_\delta)$  for spherical films.  $r_0$  and  $r_\delta$  are the position coordinates along the diffusion path at which the compositions are respectively  $x_{i0}$  and  $x_{i\delta}$ . If  $\imath$  is unknown (as it will be in most practical applications), use an appropriate mass transfer correlation of the form:  $(k_{ij}\imath/r_0 + \vartheta_{ij}) = f(\text{Re}, u/\rho \cdot \vartheta_{ij})$ . Use the multicomponent  $\vartheta_{ij}$  in place of the binary diffusivity in the binary mass transfer correlation.
- 3. At the bulk phase compositions  $x_{i\delta}$ , calculate the matrix  $[B_{\delta}]$  whose elements are given by

$$B_{\delta ij} = \frac{x_{i\delta}}{k_{in}} + \sum_{\substack{j=1 \\ i\neq i}}^{n} \frac{x_{j\delta}}{k_{ij}}; \quad B_{\delta ij} = -x_{i\delta} (1/k_{ij} - 1/k_{in}); \quad i,j = 1,2,...,n-1$$

4. Calculate the matrix of thermodynamic factors  $r_{ij}$  at arithmetic average composition  $x_i$ :

$$\Gamma_{ij} = \delta_{ij} + \frac{x_i}{x_j} = \frac{\partial \ln \gamma_i}{\partial \ln \gamma_j}$$
; where  $\delta_{ij}$  is the Kronecker delta and  $\gamma_i$  is the activity coeff-

icient of component i in solution. For mixtures of ideal gases  $\Gamma_{ij}$  is identically equal to  $\delta_{ij}$  and therefore no assumptions are involved concerning the constancy of  $\Gamma_{ij}$ . This makes MODEL I exact for ideal gas mixtures; for liquid mixtures it may be looked upon as a good approximation.

- Assume that the matrix of correction factors [E] given by [E] = [0]exp[0] + 「1」)<sup>-1</sup> is equal to unity.
- 6. Calculate the n-1 interfacial fluxes  $N_{\frac{1}{2}\delta}$  from the n-1 dimensional matrix equation

 $(N_{\delta}) = [B_{\delta}] [B_{\delta}]^{-1} [\Gamma] [\Xi] (x_0 - x_{\delta})$  where [B] is the matrix of determinancy factors allowing the calculation of the fluxes  $N_i$  from the diffusion fluxes  $J_i$ : (N) = [B](J). For condensation of n-1 vapour components in the presence of an inert gas (n), the elements of [B] are given by  $B_{ij} = \delta_{ij} + x_i/x_n$ . The subscript  $\delta$  indicates that the elements of  $B_{ij}$  are to be calculated at the bulk phase gas compositions  $x_{i\delta}$ .

7. With the estimate of the fluxes N as above calculate the matrix of rate factors [h]

$$\Phi_{ii} = \frac{N_{i\delta}}{k_{in}} + \sum_{\substack{j=1\\j \neq i}}^{n} \frac{N_{j\delta}}{k_{ij}}; \quad \Phi_{ij} = -N_i \quad (1/k_{ij} - 1/k_{in}); \quad i,j = 1,2,...n-1$$

- 8. Calculate  $[0] \equiv [\Gamma]^{-1}[\phi]$  and  $[\Xi] \equiv [0] \exp[0] \{\exp[0] \Gamma_{I_{1}}\}^{-1}$
- 9. Go to Step 6 and repeat head-to-tail iterations for the fluxes  ${\rm N}_{1\delta}$  till convergence is achieved.

Note: The  $B_{ij}$  coefficients are related to  $B_{ij}$  by:  $B_{ij} = \frac{\ell}{c} B_{ij}$ 

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- Estimate the matrix of Fick's law diffusion coefficients [D] from the kinetic theory 1. (for gases) or from experiments (for liquids). For gases, use eq (9) with arithmetic avg  $x_i$ .
- Calculate the eigenvalues  $D_i$  of the matrix [D], which is assumed to be constant along the 2. diffusion path.
- Calculate the pseudo-mass transfer coefficients  $k_i$  from: 3.  $\hat{k}_i = c \hat{D}_i/\ell$  or from appropriate binary mass transfer correlation  $(\hat{k}_i \ell/c\hat{D}_i) = f(Re, S\hat{c}_i)$ where  $D_i$ , the ith eigenvalue replaces the binary diffusion coefficient in that phase.
- 4. Assume the correction factors  $\hat{\Xi}_i$  to be unity 5. Calculate  $\hat{k}_i^{\bullet} = \hat{k}_i \hat{\Xi}_i$  where  $\hat{k}_i^{\bullet}$  represent the high flux pseudo mass transfer coefficients. The superscript black dot is used to indicate high flux coefficients.
- 6. Obtain the matrix of high flux multicomponent mass transfer coefficients

$$\begin{bmatrix} \mathbf{k}^{\bullet} \end{bmatrix} = \sum_{i=1}^{n-1} \quad \tilde{k}_{i}^{\bullet} \frac{\pi \{\begin{bmatrix} D \end{bmatrix} - \tilde{D}_{j}^{\bullet} \mathbf{\Gamma}_{j} \}}{\pi \{\tilde{D}_{i} - \tilde{D}_{j} \}} \text{ where } \pi \text{ denotes product over } n-2 \text{ factors excepting } j = i.$$

- 7. Calculate the mass transfer rates  $N_{i\delta}$  from  $(N_{\delta}) = [\beta_{\delta}][k^{\bullet}](x_{0} x_{\delta})$
- 8. Re-estimate the mass transfer correction factors for pseudo species E; from

$$\tilde{z}_{i} = \frac{\tilde{\phi}_{i} \exp \tilde{\phi}_{i}}{(\exp \tilde{\phi}_{i} - 1)} \text{ where } \tilde{\phi}_{i} \text{ is given by } \tilde{\phi}_{i} = \frac{\sum_{i=1}^{N} N_{i} c_{i}}{\tilde{k}_{i}}$$

9. Go to Step 5 and repeat head-to-tail iterations till convergence is achieved.

#### TABLE 3. MODEL III. Multicomponent Film Model, this work

- 1. Estimate the GMS diffusion coefficients  $\Phi_{ij}$  in an appropriate manner. For ideal gas mixtures  $\mathbf{D}_{ii}$  correspond exactly with the binary diffusion coefficients and are independent of composition. For liquid mixtures  $\boldsymbol{\vartheta}_{i\,i}$  are functions of composition and therefore some averaging is involved.
- 2. Calculate  $k_{ii}$  from knowledge of i or from appropriate binary mass transfer correlation using the GMS diffusion coefficients  $D_{ij}$  in place of the binary diffusivity.
- 3. At arithmetic average composition x, obtain the matrix [B] , whose elements will be given by relations analogous to those for MODEL I.
- At arithmetic average composition  $x_i$  calculate the matrix of thermodynamic factors  $r_{ii}$ 4. as discussed for MODEL I.
- 5. Calculate the matrix of determinancy factors  $\beta_{ij}$  at suitably averaged composition. For condensation of n-1 vapour species in the presence of inert gas n,  $\beta_{ij} = \delta_{ij} + x_i/x_n$ .
- The interfacial fluxes N<sub>16</sub> are then given by  $(N_{\delta}) = [\beta] [\theta]^{-1} [\Gamma] (x_0 - x_{\delta})$

Note: The  $B_{ij}$  coefficients are related to  $B_{ij}$  by:  $B_{ij} = \frac{\ell}{c} B_{ij}$ 

#### TABLE 4. Comparison of Predictions of MODEL I, II and III with Illustrative Examples

<u>Illustrative Example A</u>. Adapted from Sherwood (1937). Absorption of NH<sub>3</sub> by H<sub>2</sub>O in presence of Air. Ammonia is diffusing from an NH<sub>3</sub>-Air mixture into water under a total pressure of 20265 N/m<sup>2</sup>.

Assume that the diffusion takes place through a stagnant gas layer 1 mm thick at an average temperature of 328.15 K. At one point in the apparatus the gas contains 3% NH<sub>3</sub> by volume and the concentration of NH<sub>3</sub> in the water is so low that the partial pressure of NH<sub>3</sub> over the solution may be neglected at the position under consideration. The bulk gas is dry. Allowing for water vaporization, calculate the rate of diffusion of NH<sub>3</sub>.

<u>Data</u>: the vapor pressure of water at 328.15 K is 7359 N/m<sup>2</sup>. If we label NH<sub>3</sub> = 1, Water = 2, Air = 3, then the diffusivities of the binary pairs at the system pressure and temperature are

 $-D_{12} = 147 \text{ mm}^2/\text{s}; +D_{13} = 107.5 \text{ mm}^2/\text{s}; +D_{23} = 124.5 \text{ mm}^2/\text{s}.$ 

Taking  $r = r_0$  as the interface and  $r = r_{\delta}$  as the bulk gas phase

 $x_{10} = 0.0, x_{20} = 0.36315, x_{1\delta} = 0.03, x_{2\delta} = 0.0$ 

Result. The fluxes N1 and N2 calculated using the three models are summarized below

	N <sub>1</sub> , mmol/ m <sup>2</sup> /s	N <sub>2</sub> , mmo1/ m <sup>2</sup> /
MODEL I	- 21.1	414.0
MODEL II	- 21.1	414.0
MODEL III	- 20.8	407.5

Illustrative Example B. Adapted from Modine (1963). Ternary Mass Transfer in a Wetted-Wall Col. A falling liquid film consisting of acetone (1) and benzene (2) is brought into contact

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with a downward flowing turbulent gas mixture of acetone, benzene and helium (3). Calculate the rates of transfer of acetone and benzene for the following conditions:

compositions at the ends of the diffusion path:  $x_{10} = 0.082$ ,  $x_{20} = 0.118$  $x_{10} = 0.116$ ,  $x_{20} = 0.030$ 

Binary gas diffusivities 
$$D_{12} = 4 \text{ mm}^2/\text{s}$$
;  $D_{13} = 41 \text{ mm}^2/\text{s}$ ;  $D_{23} = 39 \text{ mm}^2/\text{s}$ 

total system pressure =  $101325 \text{ N/m}^2$ ; temperature = 301.1 K; film thickness  $z = \delta = 1.34 \text{ mm}$ . <u>Result</u>. The fluxes N<sub>1</sub> and N<sub>2</sub> calculated using the three models are summarized below.

	$N_1$ , mmol/m <sup>2</sup> / s	N <sub>2</sub> , mmo1/m <sup>2</sup> /s
MODEL I	13.4	64.2
MODEL II	14.1	63.6
MODEL III	14.1	63.5

<u>Illustrative Example C.</u> Adapted from Sherwood, Pigford and Wilke (1975). Ternary Condensation. Calculate the condensation rates of  $NH_3$  and Water vapour in the presence of inert  $H_2$  for

the following conditions. Label  $NH_3 = 1$ , Water vapour = 2,  $H_2 = 3$ .

 $x_{10} = 0.455$ ,  $x_{20} = 0.195$ ,  $x_{1\delta} = 0.3$ ,  $x_{2\delta} = 0.4$ .  $D_{12} = 29.4$  mm<sup>2</sup>/s,  $D_{13} = 113$  mm<sup>2</sup>/s,  $D_{23} = 130$  mm<sup>2</sup>/s. Film thickness = 10 mm, pressure 340400 N/m<sup>2</sup>, temperature = 366.38 K. Result.

N <sub>2</sub> , mmo1/ m <sup>-</sup> / s
- 179.2
- 178.1
- 176.3