On an Evaporation Model for a Multicomponent Droplet

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This R and D note is prompted by the statement of Newbold and Amundson (1973) in this journal that the multicomponent diffusion equations (Bird et al., 1960)

$$\frac{dy}{d\xi}_i = \sum_{j=1}^{n} (y_i N_j - y_j N_i)/(D_{ij} C_T) \qquad (1)$$

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

"cannot be solved as it stands." Since the N_i are to be functions of position and C_T and the D_{ij} are to be constant in the cited work of Newbold and Amundson, Equation (1) is a linear, first-order, vector differential equation. The formal closed form solution to this type of equation in terms of the matrix exponential or the matrizant is well known (Amundson, 1966; Frazer et al., 1955). It thus appears that tractable expressions might be obtained without the assumptions of Newbold and Amundson that the binary diffusivity of a component is the same in each of the remaining components (that is, $D_{ij} = D_{im}$, $j = 1, 2, \ldots, n$) and that an inert gas is present in the vapor phase. To this end recall the expression for N_i derived by Newbold and Amundson, namely

$$N_i = r^2 J_i / \xi^2 \qquad (2)$$

Definitions not given herein will be those of Newbold and Amundson, and n will denote the total number of components including inerts.

Substituting Equation (2) into Equation (1) produces

$$\frac{dy}{d\xi}_i = (r/\xi)^2 \sum_{j=1}^{n} (y_i J_j - y_j J_i)/(D_{ij} C_T) \qquad (3)$$

Defining the vector y by

$$y \equiv (y_1, y_2, \ldots, y_n)^T$$

and the constant matrix $M = (m_{ij})$ by

$$m_{ij} = ((\Sigma_k J_k / D_{ik}) - (J_i / D_{ii}))/C_T, \quad j = i$$

$$m_{ij} = -J_i/(D_{ij} C_T), \quad j \neq i$$

permits Equation (3) to be rewritten as

$$\frac{dy}{d\xi} = (r/\xi)^2 My$$

and the further substitution of $\eta = 1/\xi$ results in

$$\frac{dy}{d\eta} = -Mr^2 \qquad (4)$$

Equation (4) has the solution

$$y(\eta) = \exp(-Mr^2\eta)\, y(0) \qquad (5)$$

The boundary conditions of Newbold and Amundson may be written

$$y(0) \equiv y|_{\eta=0} = y|_{\xi=\infty}$$

$$= (P_{A1}, P_{A2}, \ldots, P_{An})^T / P_T \equiv p/P_T$$

$$y(1/r) \equiv y|_{\eta=1/r} = y|_{\xi=r} = [\exp(A - t^{-1}B)]x/P_T \qquad (6)$$

where

$$x \equiv (x_1, x_2, x_3, \ldots, x_n)^T$$

$$P_{Ai}^* = \exp[a_i - (b_i/t)]$$

$$A \equiv \text{diag}(a_i)$$

$$B \equiv \text{diag}(b_i)$$

Subjecting Equation (5) to the boundary conditions, Equations (6), and noting that B and A commute provides

$$e^A e^{-B/t} x = e^{-Mr} p \qquad (7)$$

Obviously iteration will be required to obtain the J_i from Equation (7), but since

$$\frac{\partial e^{-Mr}}{\partial J_i} \neq e^{-Mr} \frac{\partial(-Mr)}{\partial J_i}$$

the usual Newton-Raphson iteration is probably less desirable than the following regula-falsi scheme.

$$J_i = J_{i2} - \frac{(c_2^T c_2)(J_{i1} - J_{i2})}{c_1^T c_1 - c_2^T c_2}$$

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

where, from Equation (7)

$$c \equiv e^A e^{-B/t} x - e^{-Mr} p$$

and the subscript 2 denotes the last previous value while the subscript 1 denotes the penultimate previous value.

Finally one must face up to the problem of computing

$$e^{-Mr} \equiv \sum_{s=0}^{\infty} (-Mr)^s/(s!) \qquad (8)$$

The fact that the droplet radius r is small will certainly help as will a factorization of the type

$$e^{-Mr} = e^{-(Mr/16)(16)}$$

Further, the terms retained when truncating the right side of Equation (8) will normally be nested when the summation is carried out. Swamy (1972) has discussed computation of the matrix exponential.

In summary it is seen that Equation (7) is no more involved than the equations Newbold and Amundson use to obtain numerical values for the J_i. Furthermore Equation (7) is not restricted by two assumptions of Newbold and Amundson recalled in the first paragraph of this note. What has been added to the computation arises because Equation (7) must be solved by an iterative procedure.

LITERATURE CITED

Amundson, N. R., *Mathematical Methods in Chemical Engineering*, Prentice-Hall, Englewood Cliffs, N. J. (1966).

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

Frazier, R. A., W. J. Duncan, and A. R. Collier, *Elementary Matrices*, Cambridge Univ. Press, England (1955).
Newbold, F. R., and N. R. Amundson, "A Model for Evaporation of a Multicomponent Droplet," *AIChE J.*, **19**, 22 (1973).
Swamy, M. N. S., "On a Formula for Evaluating e^{At} when the Eigenvalues of A are not Necessarily Distinct," *Matrix Tensor Quarterly*, 23, 67 (1972).

Manuscript received May 14, 1973; revision received June 21 and accepted July 23, 1973.

Note on Light Transmission through a Polydisperse Dispersion

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A recent paper of McLaughlin and Rushton (1973) presented an analysis of light transmission through a dense dispersion of spherical particles when only the unscattered light is received by the detector. They numerically generated samples from various drop size distributions and, using a relation for the probability of a light ray (in a parallel beam) not striking any of the drops, found that the total light transmission is a unique exponential function of the group al, where a is the interfacial area per unit volume, and l is the path length through the dispersion. They confirmed this numerically for four different drop size distributions. It may be shown that this result is theoretically exact for all drop size distributions, subject to certain idealizations. Actually, Calderbank (1958) showed this, in a rather elegant fashion, but used a distribution-free argument that obscured the fact that the result is independent of a drop size distribution. The Calderbank equation is equal to that obtained by McLaughlin and Rushton. The following derivation simplifies but generalizes the analyses of Otvos et al. (1957) and Gumprecht and Sliepcevich (1953a, 1953b).

Assuming, as did McLaughlin and Rushton, a beam of parallel rays, and that scattered light (diffracted or refracted) is not received by the detector, the effective scattering cross section of a single drop of radius r is

$$K_a \pi r^2 \tag{1}$$

where K_a is the total-scattering coefficient. If the path length is l, the number of particles per unit volume is n, and their probability density distribution of radius r is $p(r)$, the average number of drops $\bar{q}$ that lie along any particular path is

$$\bar{q} = \int_0^\infty K_a \pi r^2 n l p(r)\, dr \tag{2}$$

Since $4\pi r^2 N p(r) dr$ is just the interfacial area per unit volume contributed by drops having radii between r and $r + dr$, if K_a is a constant (an acceptable assumption for large drops when also $K_a = 1$), this becomes

$$\bar{q} = \frac{1}{4} K_a a l \tag{3}$$

We now assume that the drops occur randomly and independently on a light path. The number present is then Poisson distributed (this assumes no interference between drops, as did McLaughlin and Rushton), or

$$p(q) = \frac{\bar{q}^q \exp(-\bar{q})}{q!} \tag{4}$$

The probability of light transmission on a path, and therefore in a beam, is the probability that $q = 0$, or $\exp(-\bar{q})$. Combining this and Equation (3), and letting $K_a = 1$, we obtain for the light attenuation

$$f = \exp\left(-\frac{1}{4} a l\right) \tag{5}$$

Defining N_T as $al/6$, as did McLaughlin and Rushton, the group in the exponential in Equation (5) is just $1.5N_T$, which was found numerically by them.

NOTATION

- a = interfacial area per unit volume of dispersion
- f = ratio of received to incident light intensity, or the probability of light not being scattered from a path
- K_a = total scattering coefficient
- l = length of light path through dispersion
- n = number of drops per unit volume of dispersion
- N_T = transmission number of McLaughlin and Rushton
- $p(r)$ = probability density distribution of drop radius
- $p(q)$ = probability of there being q drops on a light path
- q = number of drops on a light path
- $\bar{q}$ = average number of drops on a light path
- r = drop radius

LITERATURE CITED

Calderbank, P. H., "Physical Rate Processes in Industrial Fermentation, Part I: The Interfacial Area in Gas-Liquid Contracting with Mechanical Agitation," *Trans. Instn. Chem. Engrs.*, **36**, 443 (1958).
Gumprecht, R. O., and C. M. Sliepcevich, "Scattering of light by large spherical particles," *J. Phys. Chem.*, **57**, 90 (1953).
——————., "Measurements of particle sizes in polydispersed systems by means of light transmission combined with differential settling," *ibid.*, **57**, 95 (1953).
McLaughlin, C. M., and J. H. Rushton, "Interfacial Areas of Liquid-Liquid Dispersions from Light Transmission Measurements," *AIChE J.*, **19**, 817 (1973).
Otvos, J. W., H. Stone, and W. R. Harp, "Theory of Radiant Energy Absorption by Randomly Dispersed Discrete Particles," *Spectrochimica Acta*, **9**, 148 (1957).

Manuscript received August 10, 1973, and accepted September 4, 1973.