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Effective Binary Diffusion Coefficients in Mixed Solvents

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The diffusion of carbon dioxide through ethanol-water, benzene toluene, and carbon tetrachloride benzene was studied over the entire range of compositions for the solvent pairs. The results were interpreted in terms of an effective binary diffusion coefficient. Two semiempirical correlations were derived with the aid of absolute reaction rate theory to predict the effective binary diffusion coefficient from the binary diffusion coefficients of the solute in the individual pure solvents. These equations, although only crude approximations, reproduced the experimental data for six diffusion systems fairly well, including the highly nonideal solvent mixture ethanol-water.

Many absorption and extraction processes employ mixed solvents instead of a pure solvent. Since the theoretical treatment of such multicomponent diffusion processes on a rigorous basis is very complicated, it is desirable to be able to define and employ an effective binary diffusion coefficient (2) for the diffusing substance in mixed solvents by considering the mixed solvent as a pseudo pure liquid. One would like to have a simple method which permits the estimation of this effective binary diffusion coefficient from the binary diffusion coefficients of the solute in the individual solvents. Several empirical or semiempirical methods have been given in the literature for the diffusion of a dilute liquid component in mixed solvents (*18*, *25*). None of these methods apply to the case where the mixed solvent is highly nonideal.

The objective of this investigation was to develop a suitable approximate method for predicting the effective binary diffusion coefficient. The absorption of carbon dioxide into three mixed solvents, benzene-toluene, CCl_4-benzene, and ethanol-water, was measured and the results compared with a number of predictive methods. The third solvent system, ethanol-water, is known to be a highly nonideal mixture.

PREVIOUS PREDICTIVE RELATIONS

In a binary system, the diffusion coefficient is well defined; it is the mutual diffusion coefficient of the binary system. In multicomponent systems, however, there are many diffusion coefficients, and therefore it is desirable for practical purposes to define an effective binary diffusion coefficient for the component being absorbed or extracted. The idea of an effective binary diffusion coefficient is also useful in the analogies between momentum, heat, and mass transfer where a diffusion coefficient appears in the Chilton-Colburn *j* factor for mass transfer (*5*).

Historically, since most of the experimental work on diffusion has been for binary systems, it is desirable to have some methods which enable the prediction of the effective binary diffusion coefficient from the binary diffusion coefficients of component i in systems $i - j$. For the diffusion of a single gas into a stagnant multicomponent gas mixture, Wilke (*43*) derived from the Stefan-Maxwell equations the following expression for the effective binary diffusion coefficient

$$D_{1m} = (1 - x_1) \Big/ \sum_{j=2}^{n} (x_j/D_{1j}) \qquad (1)$$

where the subscript 1 refers to the diffusing gas and D_{1j} is the mutual diffusion coefficient of the binary system $1 - j$. The same expression was also obtained by Hsu and Bird (*19*) for the evaporation of a pure liquid into a multicomponent gas mixture. Fairbanks and Wilke (*10*) applied this equation with success to the diffusion of toluene vapor into a hydrogen-argon mixture and the diffusion of ethyl propionate into a hydrogen-air mixture.

No satisfactory method has been given in the literature for the prediction of the effective binary diffusion coefficient in a liquid phase. Bhat (*1*) studied the diffusion of some organic acids in ethanol-water mixtures of various compositions at 25°C. and proposed the following correlation:

$$D = 0.0104 \frac{\left(\dfrac{1}{M_A} + \dfrac{1}{M_B}\right)^{1/2}}{\eta (V_A^{1/3} + V_B^{1/3})^2} \qquad (2)$$

Two mistakes were found in this correlation. First, the viscosity data used by Bhat, which he obtained from the International Critical Tables (*20*), were wrong by as much as 70% when compared with the original data source. Second, the line in the plot of his data in the form of D vs. the nonnumerical term of Equation (2) did not pass through origin. Moreover, since the diffusion runs took 24 hr. or longer, the esterification reaction between the diffusing acid and ethanol became a significant factor (which was ignored) in the transport equation.

Holmes et al. (*18*) measured the effective binary diffusion coefficient for the diffusion of toluene in three hydrocarbon mixtures, *n*-hexane-*n*-tetradecane, *n*-hexane-cyclohexane, and cyclohexane-*n*-decane. The concentration of toluene was very low in all the systems; hence the physical properties of the solutions could be assumed to be those of the solvent mixtures. When they applied Equation (1) to their data, deviations of as much as 30% were observed for the *n*-hexane-cyclohexane system. On the other hand they found that the following equation

$$D_{1m}\eta_m = x_2 D_{12}\eta_2 + x_3 D_{13}\eta_3 \tag{3}$$

where the subscript 1 refers to toluene and the subscripts 2 and 3 refer to the two solvents ($x_2 + x_3 = 1$), reproduced all the data within ± 10%.

In a recent article, Lightfoot et al. (*25*) derived a method to treat diffusion in a multicomponent liquid system from the linear phenomenological relations of irreversible thermodynamics. They inverted the flux equations for a multicomponent system to obtain expressions for the force vector:

$$\mathbf{d}_j = -cRT \sum_{i=1}^{n} \left(\frac{x_i x_j}{D'_{ij}} \right) (\mathbf{v}_j - \mathbf{v}_i) \tag{4}$$

In actual applications these authors replaced D'_{ij} by the mutual diffusion coefficients D_{ij} and obtained an equation which is the same as Equation (1). However, since the phenomenological coefficients D'_{ij} are functions of composition, their replacement by the mutual diffusion coefficient is, at best, an approximation. For a nonideal solvent mixture this could introduce a very large error as noted by Holmes.

THEORY

In correlating the mutual diffusion coefficient in binary systems use is sometimes made of the absolute rate theory of diffusion (*14, 35*). One form of this theory was given by Olander (*30*) as

$$\left(\frac{\xi}{k} \right) \left(\frac{D\eta}{T} \right) \left(\frac{V}{N_{Av}} \right)^{1/3} \cong \exp \left(\frac{\Delta F_\eta{}^* - \Delta F_D{}^*}{RT} \right) \tag{5}$$

When the concentration of the diffusing solute is small

$$(\Delta F_\eta{}^*)_{\text{solution}} = (\Delta F_\eta{}^*)_{\text{solvent}}$$

The relation between the free energy of activation for viscous flow and the absolute viscosity of the liquid is

$$\eta = \frac{hN_{Av}}{V} \exp \left(\frac{\Delta F_\eta{}^*}{RT} \right) \tag{6}$$

Olander subdivided each of the free energies of activation in Equation (5) into two parts, the hole-making and the bond-breaking contributions. When the concentration of the diffusing solute is small, the moving solute molecule can be considered to be completely surrounded by solvent molecules; hence the hole-making contribution for diffusion could be assumed to be the same as that for viscous flow. Therefore, for the diffusion of dilute species 1 in species 2

$$\Delta F_\eta{}^* - \Delta F_D{}^* = \Delta F^j{}_{22} - \Delta F^j{}_{12} \tag{7}$$

where the superscript j refers to the bond-breaking contributions and the subscripts refer to the molecular species between which the interaction exists. The bond-breaking contribution $\Delta F^j{}_{22}$ constitutes a fraction of the total free energy of activation for viscous flow $(\Delta F_\eta{}^*)_2$. The ratio between these two, defined as

$$\epsilon \equiv (\Delta F^j{}_{22}) / (\Delta F_\eta{}^*)_2 \tag{8}$$

is not the same for all pure substances, and it may even vary with temperature for a given substance. A rough estimation proposed by Olander is to assume ϵ to be a constant for all pure substances, with its value estimated by Olander to be about 0.5.

With this background in mind the absolute rate theory of diffusion may be extended to the diffusion of dilute component 1 through a mixture of components 2 and 3. The derivation given below is a rather crude approximation; nevertheless, the approach is believed to be reasonable, and the predictive ability of the resulting expressions is good.

First consider Equation (5). It is reasonable to assume that this equation also holds for the diffusion of dilute component 1 through a mixture of component 2 and 3. Denoting the 2-3 mixture by m, one has

$$\left(\frac{\xi}{k} \right) \left(\frac{D_{1m}\,\eta_m}{T} \right) \left(\frac{V_m}{N_{Av}} \right)^{1/3} \cong \exp \left(\frac{\Delta F_\eta{}^* - \Delta F_D{}^*}{RT} \right)_{1m} \tag{9}$$

The subdivision of the activation free energies should also apply here. A relation corresponding to Equation (7) is then

$$(\Delta F_\eta{}^* - \Delta F_D{}^*)_{1m} = \Delta F^j{}_m - \Delta F^j{}_{1m} \tag{10}$$

Physically, the bond-breaking contribution ΔF^j is considered to be the activation free energy required by a moving molecule to overcome the intermolecular forces and move into a vacant site. In the second term in the right-hand side of Equation (10), $\Delta F^j{}_{1m}$, the moving molecule is the solute 1 while the neighboring molecules are 2 and 3 (since the concentration of solute is very small). It is therefore reasonable to assume that a simple relationship exists between $\Delta F^j{}_{1m}$ and the solvent composition, $\Delta F^j{}_{12}$, and $\Delta F^j{}_{13}$. On the other hand, the term $\Delta F^j{}_m$ involves the interactions between 2-2, 3-3, and 2-3 molecules in a complicated way.

One way to simplify the treatment is to eliminate the viscous-flow activation free energy term from the right-hand side of Equation (10). If one writes Equations (6) and (8) for the solvent mixture and accepts the value of 0.5 for ϵ, the following expression can be obtained:

$$\exp \left(\frac{\Delta F^j{}_m}{RT} \right) \cong \left(\frac{\eta_m V_m}{hN_{Av}} \right)^{1/2} \tag{11}$$

Combining Equations (9), (10), and (11), one gets

$$K \left(\frac{D_{1m}\,\eta_m{}^{1/2}}{T} \right) V_m{}^{-1/6} \cong \exp \left(\frac{-\Delta F^j{}_{1m}}{RT} \right) \tag{12}$$

where

$$K \equiv \xi / (kh^{1/2} N_{Av}{}^{5/6})$$

Similar treatment for the binary systems 1-2 and 1-3 leads to identical expressions, with the subscript m replaced by 2 or 3.

In absolute rate theory the exponential term of the free energy of activation is considered as a jump frequency of the molecules. One might therefore expect the following

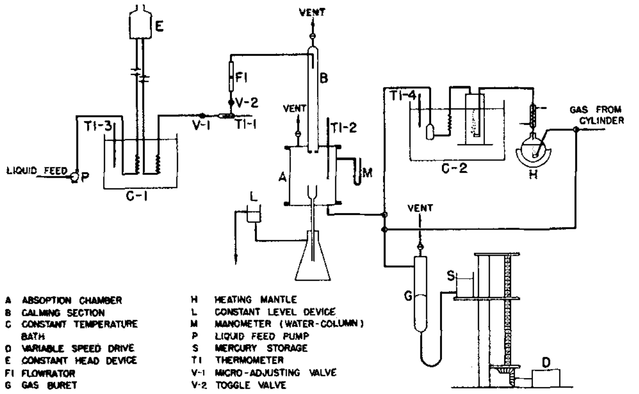

Fig. 1. Schematic flow diagram.

relation to apply at constant temperature:

$$\exp\left(\frac{-\Delta F^{\ddagger}_{1m}}{RT}\right) = x_2 \exp\left(\frac{-\Delta F^{\ddagger}_{12}}{RT}\right) + x_3 \exp\left(\frac{-\Delta F^{\ddagger}_{13}}{RT}\right) \qquad (13)$$

There are, of course, some other possible mixing rules. Combining Equations (12), its equivalent for 1-2, 1-3 binary mixtures, and Equation (13), one has at constant temperature

$$D_{1m}\,\eta_m^{1/2}V_m^{-1/6} \cong x_2D_{12}\eta_2^{1/2}V_2^{-1/6} + x_3D_{13}\eta_3^{1/2}V_3^{-1/6} \qquad (14)$$

Since the variation of the molar volume to the minus one sixth power with respect to the composition of the solvent mixture is small, Equation (14) reduces to

$$D_{1m}\,\eta_m^{1/2} \cong x_2D_{12}\eta_2^{1/2} + x_3D_{13}\eta_3^{1/2} \qquad (15)$$

Equation (15) predicts, for the diffusion of a dilute solute 1 through a liquid mixture of components 2 and 3, that a plot of $D_{1m}\eta_m^{1/2}$ vs. x_2 (or x_3) should yield approximately a straight line. The diffusion coefficients D_{1m}, D_{12}, and D_{13} were assumed to be independent of the solute concentration, which has been assumed to be small in the above derivations.

Another obvious mixing rule, which was used by Eyring and co-workers (*13*) in predicting the viscosity of a liquid mixture, is

$$\Delta F^{\ddagger}_{1m} = x_2\Delta F^{\ddagger}_{12} + x_3\Delta F^{\ddagger}_{13} \qquad (16)$$

Then the final equation corresponding to Equation (15) (again with the small change of $V^{-1/6}$ neglected) is

$$\log(D_{1m}\eta_m^{1/2}) \cong x_2 \log(D_{12}\eta_2^{1/2}) + x_3 \log(D_{13}\eta_3^{1/2}) \qquad (17)$$

The validity of neglecting the molar volume terms in Equation (14) has been tested for the diffusion of carbon dioxide in ethanol-water mixtures in which the value of $V^{1/6}$ ranged from 1.62 for pure water at 25°C. to 1.97 for pure ethanol at the same temperature, both expressed in (cubic centimeters per mole)$^{1/6}$. The predicted values of the effective binary diffusion coefficients by Equations (14) and (15) were about the same over the entire composition range.

Both Equations (15) and (17) are, of course, crude approximations. The value of 5.6 taken for the parameter ξ in the absolute rate theory for diffusion, which appeared in the left-hand side of Equation (5), is only an average value for a number of systems. Therefore, strictly speaking, the constant K is not the same for different systems, and the elimination of K is obtaining Equation (14) was only approximately correct. Furthermore, the assumption that the ratio ϵ defined by Equation (8) is constant for all systems and has a value of approximately 0.5 is even less accurate. For the enthalpy of activation for viscous flow, Bondi (*3*) found that the hole-making contribution amounted to 0.40 to 0.60 of the total enthalpy of activation for hydrocarbons. For primary alcohols the corresponding values are 0.20 to 0.30. He also reported that these values vary slightly with temperature. One would therefore expect less success in a mixed solvent containing associated liquids when Equation (15) or (17) is applied to predict the effective binary diffusion coefficient than in hydrocarbon mixtures.

EXPERIMENTAL

The absorption of gas into a liquid jet was used to obtain the desired diffusion coefficients because of the simplicity and relatively small end effects of the experimental technique. This method was probably first introduced by Matsuyama (*27*) and has been widely used since then (*4, 6, 9, 16, 17, 22, 26, 29, 31, 33, 38, 42, 45, 46*). Most of the previous authors treated their data by applying the penetration theory. A more rigorous solution to the diffusion equation for a liquid jet apparatus was derived in this work to test the validity of the assumptions lying behind the penetration solution.

In the absorption of slightly soluble gases in liquids, the concentration of the solute gas in the liquid phase is so small that the density and viscosity of the liquid phase are practically the same as those of the pure solvent. For an isothermal, steady state flow system in the laminar regime, the diffusion equation in the absence of chemical reactions becomes

$$\mathrm{v} \cdot \nabla c = \nabla \cdot (D\,\nabla c) \qquad (18)$$

The initial and boundary conditions are

$$\begin{aligned} c &= c_o \text{ at the jet nozzle exit} \\ c &= c_i \text{ on the free boundary of the liquid jet} \end{aligned} \qquad (19)$$

where the subscripts *o* and *i* refer to the feed liquid and the gas-liquid interface, respectively. Previous investigators (*6, 16, 38*) showed, for a liquid-phase controlling absorption process in the absence of surface active agents, that c_i can be assumed to be the liquid concentration in equilibrium with the gas phase.

If one temporarily assumes that the depth of penetration of the solute gas in the liquid is small and neglects the curvature of the jet, then Equations (18) and (19) lead to the penetration solution when D_P is a constant:

$$D_P = N^2/16(c_i - c_o)^2\,qL \qquad (20)$$

A more exact analysis of the same problem showed that the penetration theory gave a higher absorption rate with a given value of D. Therefore, when the diffusion coefficient was calculated from experimentally determined absorption rates, the use of the penetration theory led to a smaller value of D than proper. Small corrections (tabulated in reference *41*) were applied to D_P to derive the more exact value of D.

In this work a jet not too different in design from those reported in the literature was used; the details can be found in reference *41*. Figure 1 shows the experimental setup. Precautions were taken to have a properly designed orifice and receiver, and measurements were made of the jet surface velocity (by taking high-speed motion pictures of a dye front) and also the jet diameter to insure that the assumptions included in the mathematical model given above were in fact met by the experimental conditions. Entrance and exit effects were calculated and found to be small, but appropriate corrections were included.

A series of runs were made in which carbon dioxide was absorbed into the liquid jet. The gas phase was carbon dioxide saturated with solvent vapor at the experimental temperature, 25°C., while the liquid feed was prepared mixtures of benzene-toluene, benzene-carbon tetrachloride, or ethanol-water. All dissolved inert gas in the solvent was removed before each run. The ethanol used in this investigation was absolute ethyl alcohol and 97% ethanol. The benzene, toluene, and carbon tetrachloride were reagent grade.

TABLE 4. AVERAGE ABSOLUTE DEVIATION OF VARIOUS METHODS FOR PREDICTING THE EFFECTIVE BINARY DIFFUSION COEFFICIENTS

	Systems		Average absolute deviations, %					
	Diffusing solute	Solvents	Equation (1)	Equation (3)	Equation (15)	Equation (17)	Equation (21)	Equation (22)
(A)	Toluene	*n*-hexane-*n*-tetradecane	7.3	2.2	8.8	5.8	35.0	12.8
(B)		*n*-hexane-cyclohexane	21.2	2.0	6.5	8.3	6.3	14.0
(C)		cyclohexane-*n*-decane	3.6	10.0	7.4	7.0	4.8	4.2
(D)	CO_2	benzene-toluene	1.7	1.2	0.6	0.6	1.5	1.8
(E)		CCl_4-benzene	2.4	2.1	1.6	1.8	1.1	1.7
(F)		ethanol-water	23.4	31.9	7.0	10.3	33.4	28.3

Sources of literature values (at 25°C.) used for viscosity, vapor pressure, and carbon dioxide solubility in the mixed solvents were as follows:

	Ethanol-Water	Benzene-Toluene	Carbon tetrachloride Benzene
Viscosity	(*24*)	(*11*)	(*15*)
Vapor pressure	Interpolated from (*7*)	(*34*) and Raoult's law	(*36*) with extrapolated activities
Solubility of carbon dioxide	(*8*), (*21*), (*39*), (*47*), interpolated	(*12*), (*13*) plus assumption of ideality for mixtures	(*12*), (*13*) plus assumption of ideality for mixtures

RESULTS

Effective binary diffusion coefficients for the diffusion of dilute carbon dioxide through the three solvent mixtures as calculated by Equation (20) and then corrected are given in Tables 1, 2, and 3.[*] For comparison, the predicted values of the effective binary diffusion coefficients, from Equations (1), (3), (15), and (17), are also included in these tables. Since in both Equations (15) and (17) the molar volume terms were neglected, as a justification, the effective binary diffusion coefficients have also been predicted by Equation (14) for the ethanol-water mixed solvent system, where the variation of $V^{1/6}$ was the largest among the three systems studied.

The estimated precision of the total gas absorbed N was about ± 1% as was the precision of q and the jet length L. However, the accuracy of the calculated values of the effective binary diffusion coefficients depended greatly upon the accuracy of the solubility data. An error of 1% in the latter would introduce an error of about 2% in the calculated diffusion coefficient. Since considerable divergence was found in the reported solubility data, and since no discussion of the accuracy of the solubility data was given in the original data sources, it is impossible to make definite statements concerning the accuracy of the calculated diffusion coefficients. Some qualitative discussion is given in (*41*) concerning this problem.

METHODS FOR PREDICTING THE EFFECTIVE BINARY DIFFUSION COEFFICIENT

In spite of the crudeness of the predictive equations developed above, the experimental results of this study show that it was possible to predict (with reasonable success) the effective binary diffusion coefficient for the diffusion of a dilute solute in a solvent mixture from the diffusion coefficients of the solute in each of the individual solvents.

Table 4 shows the average absolute deviations of various estimation methods when applied to the diffusion of dilute carbon dioxide in the three solvent systems studied in this work and to the diffusion of dilute toluene in the three hydrocarbon mixtures studied by Holmes et al. (*18*). The average absolute deviations for the following ideal relations are also included in this table:

$$D_{1m} = x_2 D_{12} + x_3 D_{13} \tag{21}$$

$$\log (D_{1m}) = x_2 \log (D_{12}) + x_3 \log (D_{13}) \tag{22}$$

It can be seen from Table 4 that, for systems A through E, where the mixed solvents were nearly ideal solutions, Equations (3), (15), and (17) all reproduced the experimental data fairly well, while Equation (1) failed to reproduce the data of system B. For systems C through E the difference between the binary diffusion coefficients of the dilute solute in pure solvents are small, and even Equations (21) and (22) could be used to approximate the effective binary diffusion coefficients. Since the precision of the experimentally determined diffusion coefficients was estimated to be about ± 3% for systems D and E and was about ± 5% for systems A through C (*18*), it was impossible to tell which one of the three Equations, (3), (15), or (17), was better for these five systems.

For the diffusion of dilute carbon dioxide in ethanol-water mixtures, where the two solvents formed a highly nonideal solution, only Equations (15) and (17) derived in this work gave a fairly good estimate of the effective binary diffusion coefficients, and Equation (15) seemed to be slightly better than Equation (17). The maximum deviations of these two equations were −13.8 and −18.7%, respectively. The large deviations occurred at high ethanol concentrations where a serious discrepancy between the sources of the solubility data was noted.

The true test of a semiempirical equation such as Equation (15) or (17) is whether or not it can be applied to a large number of systems. Aside from the experimental data of Bhat (*1*), which have already been discussed and found to be unsuitable for a general correlation, the only other data found in the literature for the diffusion of a dilute nonelectrolytic solute in a highly nonideal liquid mixture was the diffusion of carbon dioxide in methanol-water mixtures studied by Hikata et al. (*17*). However, the reported value of the diffusion coefficient of carbon

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[*] Tabular material has been deposited as document 8083 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or 35-mm microfilm.

dioxide in 99% methanol at 25°C., 8.02×10^{-5} sq. cm./sec., is believed to be high as compared with 3.36×10^{-5} for the diffusion of carbon dioxide in *n*-butanol determined by the same authors and 3.66×10^{-5} for the diffusion of carbon dioxide in ethanol determined in this work. It was, therefore, not possible to test Equation (15) or (17) from their data. Further testing will have to await the arrival of new diffusivity data, and this, in turn, will be predicated on the development of reliable solubility data.

In summary, Equations (15) and (17) derived in this work, although only crude approximations, were found to be better than existing methods for predicting the effective binary diffusion coefficient for diffusion in the systems tabulated in Table 4. Further experimental work is necessary to test the generality of these two equations.

NOTATION

a = activity
c = concentration
D = mutual diffusion coefficient in a binary system at given composition, subscripts are sometimes used to indicate the components when necessary
D_{1m} = effective binary diffusion coefficient for the diffusion of species 1 through a mixed solvent
D'_{ij} = phenomenological coefficients in Equation (4)
$\mathbf{d}_j$ = force vector for species $j = c_j \nabla \mu_j$ for ordinary diffusion
h = Planck constant
K = $\equiv \xi/(kh^{1/2}N_{Av}^{5/6})$
k = Boltzman constant
L = length of the liquid jet
M = molecular weight
N = amount of gas absorbed per unit time
N_{Av} = Avogadro number
q = liquid flow rate
R = gas constant
T = absolute temperature
t = time variable
V = molar volume
$\mathbf{v}$ = velocity vector with respect to fixed coordinates
x = mole fraction
x = coordinate variable

Greek Letters

ϵ = $\equiv \Delta F^j/\Delta F_\eta^*$
η = absolute viscosity
μ = chemical potential
ξ = average number of nearest neighbors with respect to which the solute molecule moves ($\cong 5.6$)
ΔF_η^* = free energy of activation for viscous flow
ΔF_D^* = free energy of activation for diffusion
ΔF^j_{ik} = bond-breaking contribution of the free energy of activation for viscous flow or diffusion (interaction between *i*-*k* molecules)

Subscripts

A,B = components in a binary
i = gas-liquid interface
$i(j)$ = species $i(j)$
m = mixed solvent
o = jet nozzle exit ($z = 0$)
P = penetration theory solution
1,2,3 = species 1, 2, or 3

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