K_2 = association constant shown by Eq. 2, cm^3/mol
L = membrane thickness, cm
$S_{K/Li}$ = ion selectivity

Superscripts

I = compartment I
II = compartment II

Subscripts

K = potassium ion
Li = lithium ion

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R & D NOTES

Computation of Phase Equilibrium: Optimization with Thermodynamic Inconsistency

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In 1979, Gautam and Seider used the Rand method and a new algorithm for phase-splitting to compute the compositions at equilibrium for a mixture of 40 mol % ethylene glycol, 30% lauryl alcohol, and 30% nitromethane at 295 K and 1.013 bar (1 atm). The extended van Laar equation was used with the interaction coefficients for the binary pairs determined by Null (1970), who fit the experimental data of Francis (1956) with three liquid phases at equilibrium. The compositions computed by Gautam and Seider agreed to four significant figures with those of Null and the chemical potentials of the species in the three liquid phases agreed to nearly four significant figures.

However, they obtained the iteration history of the dimensionless Gibbs free energy shown in Figure 1. A single liquid phase is assumed at equilibrium. This is split into two liquid phases with compositions and Gibbs free energies shown in Table 1. As the Rand method adjusts the compositions from iteration 1–6, G/RT *increases*, and the chemical potentials agree to within two significant figures. Phase 2 is split into two liquid phases, with a significant decrease in G/RT. Then, the Rand method reduces G/RT to a minimum after iteration 8, but follows with an *increase* until the convergence criteria (fractional change in $n_{jl} \leq 10^{-3}$) are satisfied.

This spurious behavior can be traced to the thermodynamic inconsistency of the extended van Laar equation. Although the Gibbs-Duhem equation is not satisfied, the Rand method sets the chemical potentials equal (i.e., $\mu_{j1} = \mu_{j2} = \mu_{j3}, j = 1, \ldots, C$), but does not locate a stationary point in the Gibbs free energy surface.

This note is intended to explain the impact of thermodynamic inconsistency upon the performance of optimization procedures that are similar to the Rand method. First, the extended van Laar equation and the UNIQUAC equation are reviewed briefly and additional results presented before the spurious behavior of the Rand method is explained.

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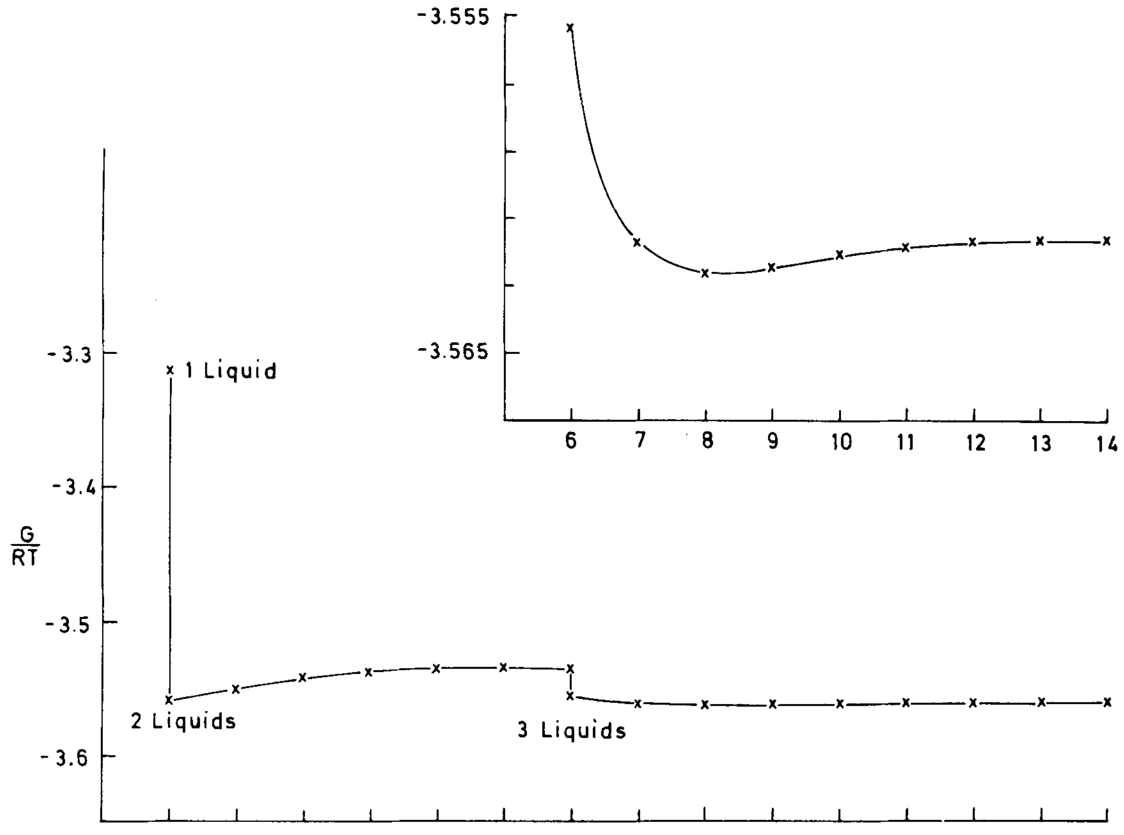

Figure 1. Gibbs free energy as a function of iteration. Extended van Laar equation with Null coefficients. λ = 1 for each iteration.

VAN LAAR EQUATION

The extended van Laar equation for multicomponent systems is:

$$ln\gamma_j = A_j(1 - z_j)^2 \left[1 + C_j z_j \left(z_j - \frac{2}{3}\right) + E_j z_j\right] \qquad (1)$$

$$j = 1, \ldots, C$$

where

$$A_j = \sum_{k=1}^{C} x_k a_{jk}/(1 - x_j);\ E_j = \begin{cases} 0 & A_j B_j \geq 0 \\ -4 & A_j B_j < 0 \end{cases}$$

$$B_j = \sum_{k=1}^{C} x_k a_{kj}/(1 - x_j);\ z_j = \frac{|A_j| x_j}{|A_j| x_j + |B_j|(1 - x_j)}$$

$$C_j = \sum_{k=1}^{C} x_k c_{jk}/(1 - x_j)$$

This relatively simple equation involves only binary interaction coefficients (normally with $c_{jk} = 0$ and $E_j = 0$). The van Laar equation is often used because it gives a good representation and requires much less computation time than more complex equations, such as the UNIQUAC and NRTL equations.

Null (1970) used Eq. 1 to represent the experimental data of Francis (1956) for the ethylene glycol-lauryl alcohol-nitromethane

TABLE 1. COMPOSITION HISTORY USING GAUTAM AND SEIDER (1979b) IMPLEMENTATION OF RAND METHOD WITH PHASE-SPLITTING: ETHYLENE GLYCOL-LAURYL ALCOHOL-NITROMETHANE AT 295 K AND 1.013 bar (1 atm)*

	Composition of Liquid Phase, mol	Composition after Split ($k = 0$) Liquid 1	Liquid 2	Composition prior to Second Split Liquid 1	Liquid 2	Composition after Second Split Liquid 1	Liquid 2	Liquid 3	Composition at Equilibrium Liquid 1	Liquid 2	Liquid 3
van Laar Eq. 1—Null (1970)											
				($k = 6$)						($k = 14$)	
Ethylene Glycol	0.4	0.0734	0.3266	0.0589	0.3411	0.0589	0.3400	0.0011	0.0724	0.3262	0.0014
Lauryl Alcohol	0.3	0.2972	0.0028	0.2847	0.0153	0.2847	0.0144	0.0009	0.2826	0.0162	0.0012
Nitromethane	0.3	0.0230	0.2770	0.0467	0.2533	0.0467	0.1397	0.1136	0.0375	0.1284	0.1341
G/RT**	−3.3127	−3.5588		−3.5351			−3.5553			−3.5617	
UNIQUAC Eq. 16—Coefs. in Table 2											
				($k = 7$)						($k = 19$)	
Ethylene Glycol	0.4	0.3963	0.0037	0.3988	0.0012	0.1483	0.2505	0.0012	0.1693	0.2289	0.00174
Lauryl Alcohol	0.3	0.2997	0.0003	0.2999	0.00007	0.2997	0.0003	0.00007	0.2985	0.0013	0.00013
Nitromethane	0.3	0.1612	0.1384	0.2726	0.0274	0.1376	0.1345	0.0282	0.1390	0.1003	0.06068
G/RT**	−3.6224	−3.6121		−3.6229			−3.6480			−3.6504	

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* Double precision—UNIVAC 90/70 computer

** $C^0_{jl} = 0, j = 1, \ldots, 3, l = 1, \ldots, 3$

TABLE 2. INTERACTION COEFFICIENTS FOR THE SYSTEM ETHYLENE GLYCOL (1)–LAURYL ALCOHOL (2)–NITROMETHANE (3) AT 295 K and 1.013 bar (1 atm)

van Laar Eq. 1—Null (1970)—$c_{jk} = 0$		
$a_{12} = 1.496$		$a_{13} = 4.588$
$a_{21} = 3.16$		$a_{23} = 4.68$
$a_{31} = 1.593$		$a_{32} = 2.878$
van Laar Eq. 1—$c_{jk} = 0$		
$a_{12} = -0.8904$		$a_{13} = 3.501$
$a_{21} = 7.510$		$a_{23} = 2.655$
$a_{31} = 1.594$		$a_{32} = 2.457$
UNIQUAC Eq. 2—$z = 10 - r_j, q_j$ from Sorensen and Arlt (1979)		
r_j	$q_j = q_j'$	
2.4088	2.248	
8.8495	7.372	
2.0086	1.868	
$\tau_{12} = 247.2$		$\tau_{13} = 54.701$
$\tau_{21} = 69.69$		$\tau_{23} = 305.52$
$\tau_{31} = 467.88$		$\tau_{32} = 133.19$

system at 295 K and 1.013 bar (1 atm) and obtained the interaction coefficients in Table 2. These give three liquid phases with the compositions in Table 1 and plotted in Figure 2 to show the poor agreement with the experimental data of Francis, especially for the phase concentrated in lauryl alcohol. (Null's Figure 6.17 shows a mistakenly good comparison; but, unfortunately, the calculated compositions are mole percents that were not converted to weight percents when plotted with the experimental data.)

In an attempt to obtain better agreement, we fit the data while biasing the concentrations of the phase concentrated in lauryl alcohol and holding $c_{jk} = 0$ and $E_j = 0$, but without significant improvement. Then, we added mutual solubility data for the binary pairs and obtained much better agreement, Figure 2. However, a negative interaction coefficient is introduced, Table 2.

With the new coefficients, compositions of three liquid phases were computed at equilibrium using the Rand and phase-splitting algorithms (Gautam and Seider, 1979b). A similar iteration history of G/RT was obtained. As previously, the chemical potentials of the three species agreed to nearly four significant figures in the three phases.

UNIQUAC EQUATION

The UNIQUAC equation (Prausnitz et al., 1980) is:

$$ln\gamma_j = ln\frac{\phi_j}{x_j} + \frac{z}{2}q_j ln\frac{\theta_j}{\phi_j} + l_j - \frac{\phi_j}{x_j}\sum_{k=1}^{C} x_k l_k - q_j' ln\left\{\sum_{k=1}^{C}\theta_j'\tau_{kj}\right\} + q_j' - q_j'\sum_{k=1}^{C}\frac{\theta_k'\tau_{jk}}{\sum_{i=1}^{C}\theta_i\tau_{ik}} \quad (2)$$

where

$$l_j = \frac{z}{2}(r_j - q_j) - (r_j - 1) \qquad \theta_j = q_j x_j \Big/ \sum_{k=1}^{C} q_k x_k$$

$$\phi_j = r_j x_j \Big/ \sum_{k=1}^{C} r_k x_k \qquad \theta_j' = q_j' x_j \Big/ \sum_{k=1}^{C} q_k' x_k$$

and the coordination number, z, is typically 10.

For the ethylene glycol-lauryl alcohol-nitromethane system, Sorensen and Arlt (1979) present the van der Waals' volume (r_j) and surface area (q_j, q_j') parameters for the pure species. These are given in Table 2 with the binary interaction coefficients (τ_{jk}) obtained by our fit of the data.

With this set of coefficients, compositions of three liquid phases at equilibrium are in fairly close agreement with the experimental data of Francis, as shown in Figure 2. The iteration history of G/RT, using the Rand and phase-splitting algorithms (Gautam and Seider, 1979b), shows a monotonic decrease until the convergence criteria are satisfied, as illustrated in Figure 3 with compositions

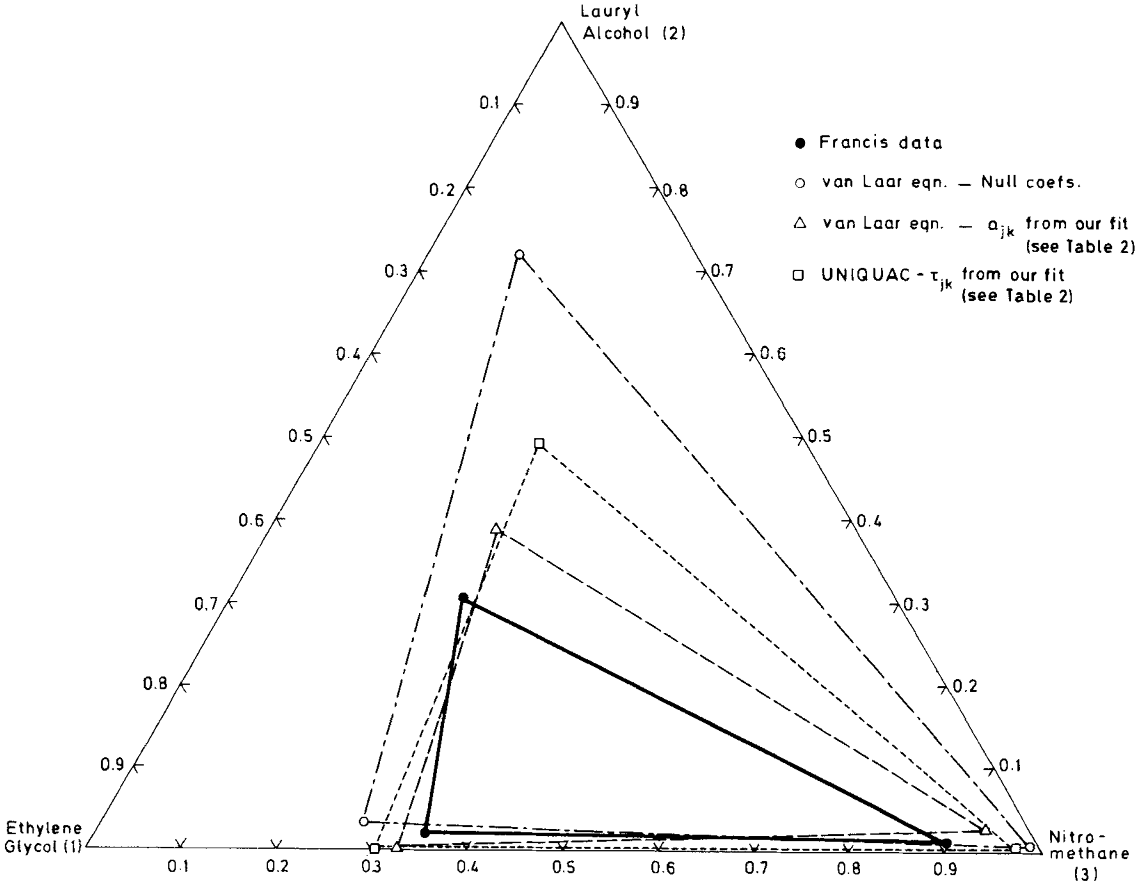

Figure 2. Mole fractions of three liquid phases at equilibrium (295 K and 1.013 bar). Interaction coefficients in Table 1.

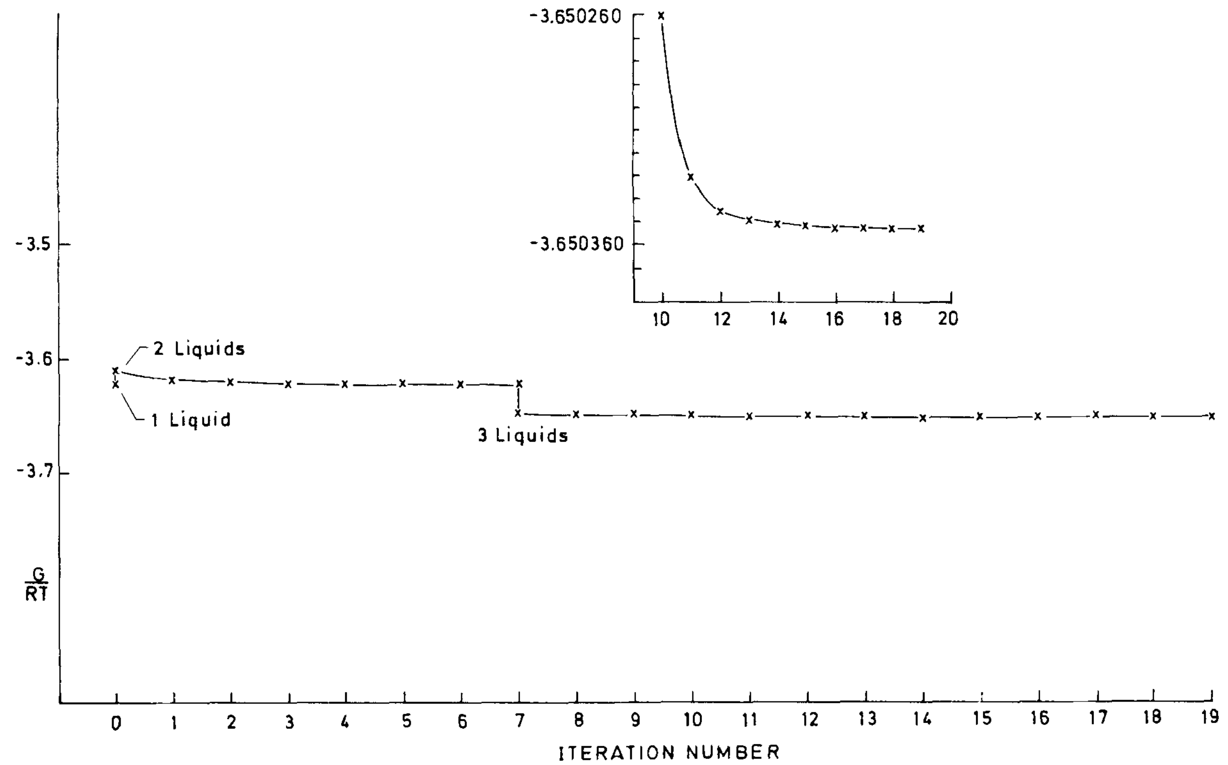

Figure 3. Gibbs free energy as a function of iteration. UNIQUAC equation with coefficients in Table 2. $\lambda = 1$ for all iterations.

in Table 1. At the solution, the chemical potentials of the three species agree to nearly four significant figures. The Gibbs-Duhem equation is satisfied and the equilibrium point is a minimum in G.

RAND METHOD

The Rand method, developed by White, Johnson, and Danzig (1958), is an implementation of Newton's method for locating stationary points in the Gibbs free energy surface. A quadratic Taylor Series is used to approximate the Gibbs free energy at $\underline{N}$, a vector of mole numbers in close proximity to the vector of guessed mole numbers, $\underline{n}$:

$$Q\{\underline{N}\} = G\{\underline{n}\} + \underline{\nabla G}^T(\underline{N} - \underline{n}) + (\underline{N} - \underline{n})^T \underline{\underline{\nabla^2 G}}(\underline{N} - \underline{n})/2 \quad (3)$$

$\underline{N}$ is computed at minimum Q subject to mass balance constraints.

Dluzniewski and Adler (1972) and Gautam and Seider (1979a) set:

$$\frac{\partial G}{\partial n_{jl}} = \mu_{jl} \quad (4)$$

Furthermore, to compute the elements of $\underline{\underline{H}} = \underline{\underline{\nabla^2 G}}$, they assumed:

$$\frac{\partial ln \gamma_{kl}}{\partial n_{jl}} = 0 \quad j,k = 1, \ldots, C \quad (5)$$

Hence, for $C = 3$, the submatrices $\underline{\underline{S}}_l$ of the constrained Hessian matrix, $\underline{\underline{H}}^c$, are:

$$\underline{\underline{S}}_l = \left(\frac{\partial^2 G}{\partial n_{jl} \partial n_{kl}}\right) \simeq \begin{matrix} \left(\frac{1}{n_{1l}} - \frac{1}{n_l}\right) & -\frac{1}{n_l} & -\frac{1}{n_l} \\ -\frac{1}{n_l} & \left(\frac{1}{n_{2l}} - \frac{1}{n_l}\right) & -\frac{1}{n_l} \\ -\frac{1}{n_l} & -\frac{1}{n_l} & \left(\frac{1}{n_{3l}} - \frac{1}{n_l}\right) \end{matrix} \quad (6)$$

$$l = 1, \ldots, \mathcal{P}$$

where,

$$\underline{\underline{H}}^c = \begin{pmatrix} (\underline{\underline{S}}_1 + \underline{\underline{S}}_3) & \underline{\underline{S}}_3 \\ \underline{\underline{S}}_3 & (\underline{\underline{S}}_2 + \underline{\underline{S}}_3) \end{pmatrix} \quad (7)$$

Note that $\underline{\underline{S}}_l$ is positive semidefinite and $\underline{\underline{H}}^c$ is positive definite.

With these approximations, the Rand method locates an equilibrium point where $\mu_{j1} = \ldots = \mu_{j\mathcal{P}}$, $j = 1, \ldots, C$. However, when the Gibbs-Duhem equation is not satisfied, a stationary point is not obtained. Rather, at $G_{\min}$, $\mu_{j1} \neq \ldots \neq \mu_{j\mathcal{P}}$, $j = 1, \ldots, C$. The composition at $G_{\min}$ can be computed using the Rand method with numerical first derivatives, $\partial G / \partial n_{jl}$, in the gradient term of Eq. 3. The iteration history of G/RT is monotonically decreasing, and the constrained Hessian matrix is positive definite at the stationary point.

During each iteration of the Rand method, Gautam and Seider define a search direction

$$\underline{\Delta} = \underline{N} - \underline{n} \quad (8)$$

and recommend that new points $\underline{N}'$,

$$\underline{N}' = \underline{n} + \lambda \underline{\Delta} \quad (9)$$

be obtained by adjusting λ to give minimum G, while avoiding negative $\underline{N}'$. However, when the Gibbs-Duhem equation is not satisfied, minimization is counter-productive when G increases to the equilibrium point.

The results for the ternary system with the extended van Laar equation were obtained using the search procedure described by Gautam and Seider, in which λ is reduced from $\lambda_{\max}$ until an estimate for:

$$\frac{dG}{d\lambda} \simeq \sum_{j=1}^{3} \sum_{l=1}^{3} G_{jl} \Delta_{jl} \quad (10)$$

becomes negative, implying that a minimum in the search direction has been passed. But, with $\lambda_{\max} = 1$, this estimate is negative at $\lambda = 1$ for nearly all iterations, although the true derivative is positive. Fortuitously, this poor estimate permits the Rand method to converge to an equilibrium point along a trajectory that increases in G/RT.

TABLE 3. RESULTS OF UNIDIRECTIONAL SEARCH WITH MINIMIZATION WHEN $G\{\underline{N}\} < G\{\underline{n}\}$ AND MAXIMIZATION OTHERWISE; $0 \leq \lambda \leq 4$. FIBONACCI SEARCH BEGINNING AT ITERATION 8 FOR VAN LAAR EQ. 1 WITH NULL COEFFICIENTS

Iteration k	$G\{\underline{N}\}/RT$		$G\{\underline{N}'\}/RT$
7	−3.5616		
8	−3.5626	$\lambda_{min} = 3.99$	−3.5648
9	−3.5618	$\lambda_{max} = 3.99$	−3.5516
10	−3.5602	$\lambda_{min} = 3.1$	−3.5706
11	−3.5705	$\lambda_{max} = 3.99$	−3.5472
⋮	⋮		⋮
⋮	⋮		⋮

To improve the efficiency, we sought to carry out the unidirectional search more accurately, but were uncertain whether to search for a minimum or maximum in the $\underline{\Delta}$ direction. We developed a strategy based upon our observations for the glycol-lauryl alcohol-nitromethane system. Let $G\{\underline{N}\}$ be the Gibbs free energy after each iteration of the Rand method. For the ternary system, we observed that if $G\{\underline{N}\} < G\{\underline{n}\}$, a minimum occurs with $\lambda > 0$, and if $G\{\underline{N}\} > G\{\underline{n}\}$, a maximum occurs with $\lambda > 0$. This suggested that the sign of $G\{\underline{N}\} - G\{\underline{n}\}$ determine whether to search for a maximum or minimum point.

We tested such an algorithm using the van Laar equation with Null coefficients, beginning with iteration 8, and using a Fibonacci search in the range $0 \leq \lambda \leq 4$. The results in Table 3 show that during iteration 8, G/RT is reduced to −3.5648 with $\lambda_{min} = 3.99$, whereas during iteration 9, G/RT is increased to −3.5516 with $\lambda_{max} = 3.99$. Subsequent iterations similarly reversed from minimization to maximization and back, with many more iterations to converge to the equilibrium point. Hence, we recommend that with thermodynamically inconsistent models the unidirectional search be abandoned, leaving $\underline{N}' = \underline{N}$; i.e., $\lambda = 1$.

CONCLUSIONS

(1) Dluzniewski and Adler (1972) and Gautam and Seider (1979a) take $\partial G/\partial n_{jl} = \mu_{jl}$ in the Rand method. They obtain convergence to an equilibrium point, even when the Gibbs-Duhem equation is not satisfied (i.e., $\partial G/\partial n_{jl} \neq \mu_{jl}$). When $\partial G/\partial n_{jl}$ is computed numerically, the Rand method converges to a minimum in the Gibbs free energy (but, the chemical potentials of the species are unequal).

(2) Numerical experiments for the ethylene glycol-lauryl alcohol-nitromethane system, with a thermodynamically inconsistent model, show that the Rand algorithm is more efficient with $\lambda = 1$ than using a unidirectional search designed to locate a minimum or maximum in the search direction.

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NOTATION

a = binary interaction coefficient, van Laar equation
A = see Eq. 1
B = see Eq. 1
c = binary interaction coefficient, van Laar Eq. 1
C = number of chemical species, Eq. 1
E = see Eq. 1
G = Gibbs free energy
G_{jl} = partial molal Gibbs free energy for species j in phase l
G^0_{jl} = Gibbs free energy of formation in standard state for species j in phase l
$\underline{\underline{H}}$ = Hessian matrix
$\bar{l}$ = see UNIQUAC Eq. 2
n_{jl} = moles of species j in phase l
n_ℓ = moles of phase l
$\underline{N}$ = moles at minimum Q
$\underline{N}'$ = moles after unidirectional search
$\bar{\mathcal{P}}$ = number of phases
q,q' = van der Waals' surface area parameter
Q = quadratic approximation of G surface
r = van der Waals' volume parameter
R = universal gas constant
$\underline{\underline{S}}$ = diagonal submatrix of $\underline{\underline{H}}$ (unconstrained Hessian matrix)
T = absolute temperature
$\underline{x}$ = vector of mole fractions
z = see Eq. 1; coordination number in UNIQUAC equation

Greek Letters

γ = liquid-phase activity coefficient
$\underline{\Delta}$ = vector in search direction ($= \underline{N} - \underline{n}$)
$\bar{\theta}$ = molal average van der Waals' surface area
λ = parameter of unidirectional search, Eq. 10
μ_{jl} = chemical potential of species j in phase l ($= G^0_{jl} + RT\, ln\{\gamma_{jl} x_{jl}\}$)
ϕ = molal average van der Waals' volume
τ = binary interaction coefficient, UNIQUAC equation

Subscripts

j = species counter
k = species counter
l = phase counter

Superscripts

c = constrained derivative
T = matrix transpose

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The Influence of Suspended Particle Size Distribution in Deep-Bed Filtration

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Deep-bed filtraton has been used in municipal water treatment since 1890. Many studies have been done on the influence of different independent variables on filter performance. Although from both observations and theory it has been found that the size of suspended particles is one of the most important physical variables influencing deep-bed filtration, size is not often measured because the techniques available are time consuming and expensive (O'Melia and Ali, 1978).

Yao et al. (1970) were the first to study the particle size effects in filtration. Based on their clean-bed filter studies they stated: "There exists a critical size at which the suspended particles have a minimum removal efficiency. This critical suspended particle size is in the order of 1 μm" (Figure 1). O'Melia and Ali (1978), in their detailed study of monodimensional latex suspension, showed that the size effect continues throughout the period of effective filtration, and 1 micron particles always showed the poorest removal. They also developed a mathematical model taking into account the particle size influence on filter removal efficiency.

In reality, the suspension has a wide spectrum of particle sizes. As shown in Figure 2, removal efficiency of different sizes of particles in the suspension would be different. It is also possible that the removal efficiency of particles of a particular size could be altered by the presence of other particles of different sizes. Therefore a detailed study of the influence of particles of different sizes on the removal efficiency of a particle of a particular size would be a useful addition to the earlier works on particle size effect.

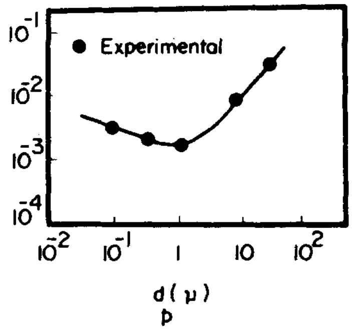

Figure 1. Collector efficiency of particles of different sizes (Yao et al., 1970).

EXPERIMENT

Laboratory-scale filter experiments were performed using artificial suspension of pollen grains of different sizes (Table 1). A known dose of

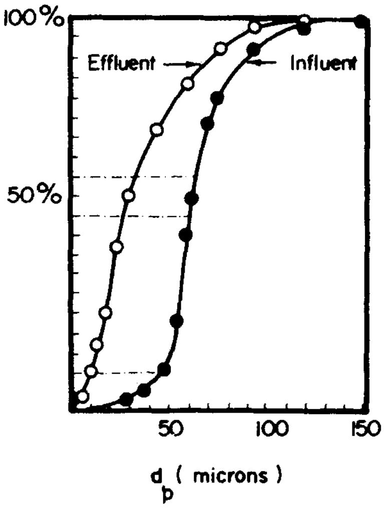

Figure 2. Filter influent and effluent size distribution (suspension used: effluent from municipal wastewater treatment; V_o = 7 $m^3/m^2 \cdot h$; filter media = glass beads of 2.5 mm) (Al Alousi, 1979).