Vapor/Liquid Equilibrium Calculations by Constrained Free-Energy Minimization[1]

J. K. Drohm[2] **and A. G. Schlijper**[2]

A method is presented for approximate vapor/liquid equilibrium calculations using an equation of state. The method reduces the number of variables in the free-energy minimization problem by the introduction of suitably chosen additional constraints. It is based on a representation of a multicomponent fluid as a mixture of a small number of weakly polydisperse components. The approximation method can be used to accelerate sequences of vapor/liquid equilibrium calculations, such as are encountered in many engineering applications [plant design, hydrocarbon (oil/gas) reservoir simulation]. In realistic cases computing time has been reduced by up to 65%.

KEY WORDS: equation-of-state calculations; polydisperse fluid; vapor/liquid equilibrium; variational approximation.

1. INTRODUCTION

Analytic equations of state, especially cubic ones, are widely used in the oil industry. Vapor/liquid equilibrium (VLE) calculations with an equation of state (EOS) provide the most accurate and reliable method for predicting the phase behavior of reservoir fluids [1–3]. However, the accurate modeling of a reservoir fluid system generally requires the use of many (20–30) components. Since in practical applications (e.g., reservoir simulation) a large number of consecutive VLE calculations often has to be performed, the large number of components leads to a large expenditure in computer time. Also, problems are frequently encountered with the convergence of the numerical algorithm used to solve the set of equations describing two-phase equilibrium.

---

[2] Koninklijke/Shell Exploratie en Produktie Laboratorium, Rijswijk, The Netherlands.

In this paper we describe an approximation method for vapor/liquid equilibrium calculations with an equation of state. The VLE problem is equivalent to the problem of minimizing the Gibbs free energy. Our approximation method introduces additional constraints into this minimization problem, which leads to the formulation of an "approximate minimization problem" (AMP) with fewer independent variables than the exact minimization problem (EMP). The AMP may then be solved more quickly than the EMP; its solution provides an approximation to the exact equilibrium state. This approximate solution either may be accepted as the final answer or may be used as an initial estimate for a full Newton–Raphson iterative scheme for solving the EMP. Since generally the solution of the AMP is very close to the exact answer, this scheme will then converge quickly.

In Section 2 the AMP is formulated and the general form of the constraints is given, together with some heuristic motivation. In Section 3 it is shown that our approximation follows naturally if the fluid system is considered as a mixture of weakly polydisperse components. In Section 4 we apply our approximation technique in accelerating sequences of VLE calculations. Examples of approximation results are compared with exact results, for a simple artificial fluid system and for a realistic reservoir fluid. Conclusions are formulated in Section 5.

2. THE APPROXIMATE MINIMIZATION PROBLEM (AMP)

In vapor/liquid equilibrium (VLE) calculations for an N-component fluid system at fixed pressure P and temperature T, the thermodynamic quantity of interest is the Gibbs free energy G of the system:

$$G = G^L(\{x_i\}) + G^V(\{y_i\}) \tag{1}$$

which is the sum of contributions G^L from the liquid and G^V from the vapor phase. Here x_i and y_i, respectively, denote the number of moles of component i present in the liquid and vapor phases. The total amount of each component in the system is

$$z_i = x_i + y_i \tag{2}$$

Hence, at fixed P, T, and composition $\{z_i\}$, G may be regarded as a function of N independent variables; to be definite we shall take these to be the x_i, $i = 1,..., N$. The thermodynamic equilibrium state is determined from the condition that G be minimal. This constitutes a minimization problem

(EMP) in the N-dimensional space of the variables x_i. The first-order conditions for this minimum lead to the well-known equations

$$\mu_i^L = \mu_i^V, \qquad i = 1,..., N \tag{3}$$

where μ_i denotes the chemical potential of the ith component.

In this paper we take Eq. (3) as being equivalent to the EMP; the problem of distinguishing among the various sorts of extrema of G, stability problems, etc., is not addressed here [4].

We construct an approximation to the EMP as follows. Divide the N components of the fluid system into K groups; $i\varepsilon s$ will symbolize that component i belongs to group s. Define "lumped variables" ξ_s, η_s, ζ_s, for $s = 1,..., K$ by

$$\xi_s = \sum_{i\varepsilon s} x_i, \qquad \eta_s = \sum_{i\varepsilon s} y_i, \qquad \zeta_s = \sum_{i\varepsilon s} z_i \tag{4}$$

Next, define a set of additional parameters, "similarity parameters" [5], C_i, for $i = 1,..., N$ by

$$C_i = \frac{y_i/\eta_s - x_i/\xi_s}{z_i/\zeta_s}, \qquad i\varepsilon s \tag{5}$$

Note that the group number s is unambiguous for any component number i since each component belongs only to one group. Also note that for any group s

$$\sum_{i\varepsilon s} z_i C_i = 0 \tag{6}$$

so only $(N-K)$ of the similarity parameters are independent. From Eqs. (2), (4), and (5) we find

$$\frac{x_i}{\xi_s} = \frac{z_i}{\zeta_s} \cdot \left[1 + C_i \left(\frac{\xi_s - \zeta_s}{\zeta_s} \right) \right], \qquad i\varepsilon s$$

$$\frac{y_i}{\eta_s} = \frac{z_i}{\zeta_s} \cdot \left[1 - C_i \left(\frac{\eta_s - \zeta_s}{\zeta_s} \right) \right], \qquad i\varepsilon s \tag{7}$$

The approximation now introduced is based on the assumption that the similarity parameters C_i are constants, i.e., independent of P, T, and $\{z_i\}$. In this case the Gibbs free energy G of the system may be regarded as a function of the K variables ξ_s and the problem of minimizing G by variation of the ξ_s is a K-dimensional problem.

The first-order conditions for the AMP are

$$\left(\frac{\partial G^{\mathrm{L}}}{\partial \xi_s}\right)_{P,T,\{z_i\},\{C_i\}} = \left(\frac{\partial G^{\mathrm{V}}}{\partial \eta_s}\right)_{P,T,\{z_i\},\{C_i\}}, \qquad s = 1,\dots, K \qquad (8)$$

The derivatives in Eq. (8) are analogous to the chemical potentials in Eq. (3) and may be called pseudo-chemical potentials or pseudopotentials; for these we use the notations $\tilde{\mu}_s^{\mathrm{L}}$, $\tilde{\mu}_s^{\mathrm{V}}$. The set of equations

$$\tilde{\mu}_s^{\mathrm{L}} = \tilde{\mu}_s^{\mathrm{V}}, \qquad s = 1,\dots, K \qquad (9)$$

will be taken as being equivalent to the AMP.

The pseudopotentials are easily related to the chemical potentials: with the aid of Eq. (7), we find

$$\tilde{\mu}_s = \sum_{i \in s} \lambda_i \mu_i \qquad (10)$$

with

$$\lambda_i = \frac{z_i}{\zeta_s}\left[1 + C_i\left(\frac{2\xi_s - \zeta_s}{\zeta_s}\right)\right] = \frac{z_i}{\zeta_s}\left[1 - C_i\left(\frac{2\eta_s - \zeta_s}{\zeta_s}\right)\right], \qquad i \varepsilon s \qquad (11)$$

A heuristic motivation for the approach described here is the following.

Simplification of a complicated reservoir fluid description is traditionally accomplished by grouping components together and treating these groups each as one component, the so-called pseudocomponents [6, 7]. Pseudocomponents are endowed with physical properties that are averages of the properties of the constituents. This pseudocomponent approximation is, in essence, based on the assumption that the composition of a group or pseudocomponent is identical in both coexisting phases and is the same as the composition of the group in the total system. This is equivalent to the assumption, in our formulation, that all the similarity parameters are identically equal to zero, as may be observed immediately from Eq. (7). A first improvement on this assumption would be to take the C_i constant, equal to their values for a known two-phase equilibrium situation, which is the approximation we propose here.

3. VAPOR/LIQUID EQUILIBRIUM IN A MIXTURE OF WEAKLY POLYDISPERSE COMPONENTS

In this section we relate the approximation presented in Section 2 to the VLE problem for mixtures of polydisperse components [8–12].

Consider a mixture of K components, with mole fractions $\hat{\zeta}_s$, $s = 1,..., K$, and suppose that each component is polydisperse in nature. By this we mean that each component consists of a mixture of various "species"; we assume that these species can be labeled by a variable ω that takes values in some label set Ω. Ω is supposed to be a measure space equipped with a suitably chosen a priori measure μ. For instance, Ω may be the set of real numbers R with the Lebesgue measure or the set of natural numbers N with the counting measure. The "internal composition" of each polydisperse component is then given by a density function $F_s(\omega)$ with

$$\int_\Omega F_s(\omega)\, d\mu(\omega) = 1 \tag{12}$$

In the context of an equation of state of the Van der Waals type, such as the EOS of Soave [13] or of Peng and Robinson [14], such a polydisperse component is characterized by two functions $a_s(\omega)$ and $b_s(\omega)$, which act as generalizations of the EOS constants a_i and b_i characteristic of a pure component [8, 15]. (This is assuming that there are no binary interaction coefficients.) The VLE problem for such a system then takes the following general form [15]:

$$\log \frac{\hat{\eta}_s\, g_s(\omega)}{\hat{\zeta}_s\, f_s(\omega)} = \phi_s(\omega) \tag{13}$$

$$\hat{X}\hat{\zeta}_s f_s(\omega) + \hat{Y}\hat{\eta}_s\, g_s(\omega) = \hat{\zeta}_s F_s(\omega) \tag{14}$$

Here $\hat{\zeta}_s$ is the mole fraction of component s in the liquid phase and $f_s(\omega)$ is the density function describing its internal composition; $\hat{X}$ is the molar liquid fraction; and $\hat{\eta}_s$, $g_s(\omega)$, and $\hat{Y}$ are the corresponding quantities for the vapor phase. The function ϕ_s is related to a difference of chemical potentials excluding contributions from the ideal mixing terms [10]. Its specific form depends on the EOS, but a general feature is that the dependence on s and ω is derived entirely from the dependence on $a_s(\omega)$ and $b_s(\omega)$ (cf. Ref. 15, Appendix II).

At fixed P, T, $\hat{\zeta}_s$, and $F_s(\omega)$, Eqs. (13) and (14) have to be solved for $\hat{X}$, $\hat{\zeta}_s$, $f_s(\omega)$, $\hat{Y}$, $\hat{\eta}_s$, and $g_s(\omega)$, taking into account the normalization conditions:

$$\hat{X} + \hat{Y} = \int_\Omega f_s(\omega)\, d\mu(\omega) = \int_\Omega g_s(\omega)\, d\mu(\omega) = \sum_s \hat{\zeta}_s = \sum_s \hat{\eta}_s = 1 \tag{15}$$

Let us now assume that each component in the mixture is only weakly polydisperse in nature, i.e., let

$$a_s(\omega) = a_s{}^0 + \varepsilon_s \alpha_s(\omega), \qquad b_s(\omega) = b_s{}^0 + \delta_s \beta_s(\omega) \tag{16}$$

with ε_s and δ_s small parameters. If $\varepsilon_s = \delta_s = 0$ for all s, then each component is monodisperse, $\phi_s(\omega)$ is independent of ω, and the mixture is equivalent to an ordinary mixture of K components. The VLE problem then has the solution $\hat{X}^0$, $\hat{\xi}_s^{\,0}$, $\hat{Y}^0$, $\hat{\eta}_s^{\,0}$ together with

$$f_s^{\,0}(\omega) = F_s(\omega); \qquad g_s^{\,0}(\omega) = F_s(\omega) \tag{17}$$

We may now look for solutions of Eq. (13) as series expansions in ε_s, δ_s. A straightforward calculation yields that if we take

$$a_s^{\,0} = \int F_s(\omega)\, a_s(\omega)\, d\mu(\omega), \qquad b_s^{\,0} = \int F_s(\omega)\, b_s(\omega)\, d\mu(\omega) \tag{18}$$

to first order the only effect of the polydispersity of the components is on their internal composition in the coexisting phases (cf. Ref. 10):

$$
\begin{aligned}
f_s(\omega) &= F_s(\omega) \cdot \left[1 - \frac{\hat{Y}^0 \hat{\eta}_s^{\,0}}{\hat{\zeta}_s} \left\{ \varepsilon_s \alpha_s(\omega) \frac{\partial \phi_s}{\partial a_s} + \delta_s \beta_s(\omega) \frac{\partial \phi_s}{\partial b_s} \right\} \right] + O(\varepsilon^2) \\
g_s(\omega) &= F_s(\omega) \cdot \left[1 + \frac{\hat{X}^0 \hat{\xi}_s^{\,0}}{\hat{\zeta}_s} \left\{ \varepsilon_s \alpha_s(\omega) \frac{\partial \phi_s}{\partial a_s} + \delta_s \beta_s(\omega) \frac{\partial \phi_s}{\partial b_s} \right\} \right] + O(\varepsilon^2)
\end{aligned}
\tag{19}
$$

The derivatives that occur in these expressions are to be evaluated at $\varepsilon_s = \delta_s = 0$.

These first-order corrections to $f_s(\omega)$ and $g_s(\omega)$ generate second-order corrections to $\hat{X}$, $\hat{Y}$, $\hat{\xi}_s$, and $\hat{\eta}_s$.

Given the expressions for $f_s(\omega)$ and $g_s(\omega)$ in Eq. (19) it is a natural progression to look for solutions of the polydisperse VLE problem with

$$f_s(\omega) = F_s(\omega) \cdot \left[1 - \hat{Y} \frac{\hat{\eta}_s}{\hat{\zeta}_s} C_s(\omega) \right], \quad g_s(\omega) = F_s(\omega) \cdot \left[1 + \hat{X} \frac{\hat{\xi}_s}{\hat{\zeta}_s} C_s(\omega) \right] \tag{20}$$

with

$$C_s(\omega) = \frac{g_s(\omega) - f_s(\omega)}{F_s(\omega)} = \varepsilon_s \alpha_s(\omega) \frac{\partial \phi_s}{\partial a_s} + \delta_s \beta_s(\omega) \frac{\partial \phi_s}{\partial b_s} \tag{21}$$

and to determine $\hat{X}$, $\hat{Y}$, $\hat{\xi}_s$, $\hat{\eta}_s$ from the requirement that the (polydisperse) free energy should be a minimum. A detailed discussion of this approximation and its application will appear elsewhere [16].

For our present purpose (see Section 4) we introduce an additional simplification: in the expression for $C_s(\omega)$ in Eq. (21) we neglect the dependence on P, T, $\{\hat{\xi}_s\}$, $\{\hat{\eta}_s\}$ of the coefficients $\partial \phi_s / \partial a_s$ and $\partial \phi_s / \partial b_s$; with this assumption $C_s(\omega)$ is a *fixed* function that describes to first order the effect

of the polydisperse nature of the components in the mixture. Assuming that $C_s(\omega)$ is fixed, it may be calculated by means of the first equality of Eq. (21) from any known VLE situation.

The relationship to our approximation in Section 2 is now obvious: a polydisperse component corresponds to a group of actual, pure components; if ω takes discrete values, Eq. (21) corresponds to Eq. (5) and Eq. (20) has its counterpart in Eq. (7). [Note that Eq. (7) is in terms of mole numbers rather than mole fractions.] Thus, our approximation may be motivated by taking the point of view that in a complex fluid mixture, such as a reservoir oil, it is possible to distinguish groups of components, each of which can adequately be described as one weakly polydisperse component.

4. APPLICATION AND EXAMPLES

4.1. Application

We have applied the approximation method described in Section 2 as a means of accelerating sequences of VLE calculations. Such sequences are encountered, for instance, in numerical reservoir simulation, where the total number of VLE calculations may be of the order of 10^6.

The problem, then, is to determine VLE at conditions P, T, $\{z_i\}$, knowing the equilibrium results at another set of conditions (which we refer to as calibration conditions, referred to by means of a superscript ^{0}) P^0, T^0, $\{z_i^0\}$. Equation (3) may be solved for the new set of conditions by means of a Newton–Raphson iterative scheme [17–20], but the success and efficiency of this method depend on the availability of good initial estimates. Such estimates are obtained by solving the approximate problem AMP [Eq. (9)] at P, T, $\{z_i\}$ first.

From the known equilibrium data $\{x_i^0\}$, $\{y_i^0\}$ at calibration conditions, ξ_s^0, η_s^0, and ζ_s^0 are calculated by Eq. (4), and C_i^0 by Eq. (5). Obviously, with $C_i = C_i^0$, at calibration conditions the AMP yields the exact VLE data; this is why we speak of calibration of the approximation.

The problem of solving the AMP at the new conditions, knowing its solution at calibration conditions, is now completely analogous *qualitatively* to the original problem, but it differs *quantitatively* in that it entails only K instead of N variables. Consequently any algorithm that has been designed for sequential VLE calculations may be applied to the approximate calculation; it will perform faster since the problem has been reduced in size.

From the values for ξ_s and η_s so obtained, values for the x_i and y_i are found by means of Eq. (7), using $C_i = C_i^0$. These, then, may be used as

initial estimates to solve the exact VLE problem. Alternatively, if the change in conditions is not too large, the approximation results may be accepted as the final answer, thus saving even more computing time. For this the following criterion is used: a complete VLE calculation is performed and the approximation is recalibrated only if one of the following conditions is violated [3]:

$$|\zeta_s - \zeta_s^0| \leqslant \Delta\zeta_{\max}, \qquad s = 1,..., K$$

$$|P - P^0| \leqslant \Delta P_{\max} \tag{22}$$

$$|T - T^0| \leqslant \Delta T_{\max}$$

As default values of the tolerances in Eq. (22) we use $\Delta\zeta_{\max} = 0.10$, $\Delta P_{\max} = 1000$ kPa, and $\Delta T_{\max} = 20°$C, but the pressure tolerance in particular may often be set at a much higher value.

The procedure described above has been implemented in the algorithm for VLE calculations of Kao, which is described in detail elsewhere [18]. The calculations discussed below were performed with this algorithm, using the equation of state of Soave, on a Univac 1106/2 computer.

4.2. Examples

By way of illustration we present some results for two fluid systems.

Fluid A (Table I) is an artificial nine-component system which is an obvious candidate for representation by three polydisperse components or groups. The approximation was calibrated at 1000 kPa and 300 K and approximation results at various conditions were compared with the exact VLE data. Figure 1 shows the deviation (approximation result — exact

Table I. Fluid A Composition and Component Grouping

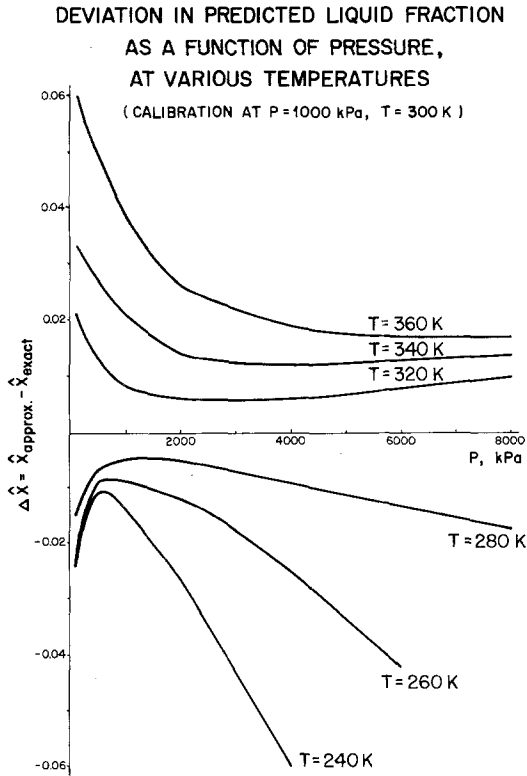| Group	Component	Mole fraction
I	Methane	0.40
II	N-Butane	0.10
	N-Pentane	0.10
	N-Hexane	0.10
III	N-Decane	0.06
	N-Undecane	0.06
	N-Dodecane	0.06
	N-Tridecane	0.06
	N-Tetradecane	0.06

Fig. 1. Illustration of the accuracy of the variational approximation for fluid A.

result) in the molar liquid fraction at various pressures and temperatures. The deviation at $300\,K$ is not shown: it is less than 0.0005 over the entire pressure range from 1000 to 8000 kPa.

Variation in system composition has also been considered. Illustrative results are given in Fig. 2. The deviation in liquid fraction is shown as a function of pressure, for various mixtures of fluid A and methane. The symbol x denotes the number of moles of methane that have been added to 1 mol of fluid A.

The approximation is shown to be accurate over a wide range of pressures and compositions; it is more sensitive to temperature variations, however.

Application of the approximation as an acceleration method has been tested in a realistic case, a 28-component representation of an actual reservoir fluid (Table II). Figure 3 shows the liquid fraction as a function of
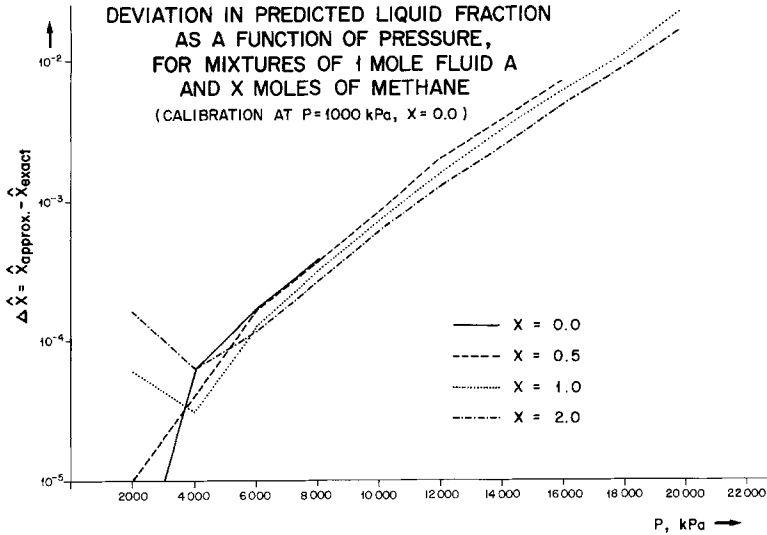
Fig. 2. Illustration of the accuracy of the variational approximation for fluid A.

pressure, at the reservoir temperature of 394 K, for various mixtures of this fluid B and nitrogen. To illustrate the accuracy of the approximation in this case, Fig. 3 also shows approximation results based on calibration at 15,000 kPa, 394 K, and $x = 0.50$. Again, for this realistic fluid mixture the approximation is shown to be accurate over a wide range of conditions.

The amount of computing time that is saved by using our method depends on whether approximated results are acceptable if conditions do not differ too much from the last calibration conditions. Demanding exact answers in all cases {i.e., setting $\Delta \zeta_{max}$, ΔP_{max}, and ΔT_{max} [cf. Eq. (22)] equal to zero}, generation of the data for Fig. 3—which involved 61 VLE calculations—took 74% of the computing time required by the original algorithm. For individual VLE calculations this percentage ranged from 35 to 90, depending on the change in conditions between successive calculations.

The approximate calculation typically takes 10–15% of the time of an exact calculation. Thus it is very advantageous to forego the final iteration procedure whenever the approximation results are expected to be of acceptable accuracy. Particularly in reservoir simulation a large number of VLE calculations are performed within a relatively narrow range of conditions; in such circumstances most calculations will be performed in approximation only, and accordingly execution time will be reduced substantially.

Table II. Fluid B Composition and Component Grouping

Group	Component	Mole fraction
I	Nitrogen	0.0108
II	Methane	0.2268
III	Carbon dioxide	0.0068
	Ethane	0.0898
	Propane	0.0928
	I-Butane	0.0127
	N-Butane	0.0452
	I-Pentane	0.0139
	N-Pentane	0.0249
	N-Hexane	0.0304
IV	*N*-Heptane	0.0200
	N-Octane	0.0100
	N-Nonane	0.0100
	N-Decane	0.0100
	N-Undecane	0.0100
	N-Dodecane	0.0200
	N-Tridecane	0.0200
	N-Tetradecane	0.0200
V	*N*-Pentadecane	0.0300
	N-Hexadecane	0.0300
	N-Heptadecane	0.0250
	N-Octadecane	0.0200
	Eicosane	0.0200
	N-C24	0.0200
	N-C28	0.0159
	Naphthalene	0.0200
	Biphenyl	0.0500
	Terphenyl	0.0950

5. CONCLUSION

We have presented a method for approximate vapor/liquid equilibrium calculations with an equation of state. The method reduces the number of independent variables by introducing additional constraints into the free energy minimization problem. It is based on the assumption that a complex fluid mixture may be viewed as consisting of a small number of weakly polydisperse components.

The approximation method may be applied as a means of accelerating sequences of vapor/liquid equilibrium calculations, such as are encountered

PHASE SPLIT ISOTHERMS AT 394 K FOR VARIOUS MIXTURES OF
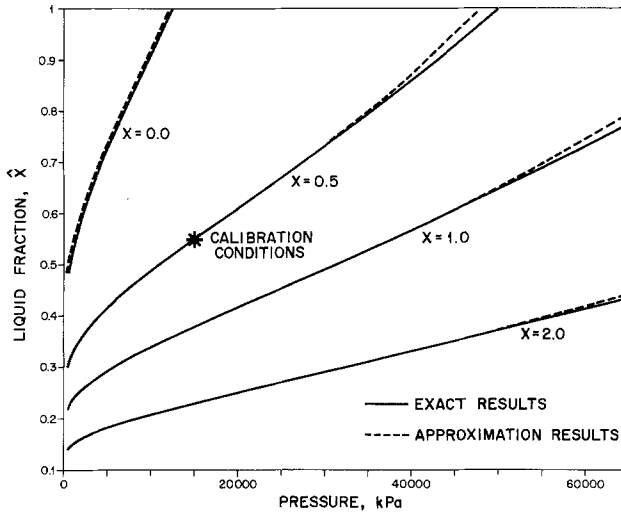1 MOLE OF FLUID B AND X MOLES OF NITROGEN

Fig. 3. Illustration of the accuracy of the variational approximation for
fluid B.

in numerical reservoir simulation. Computing time for an (exact!)
equilibrium determination may be reduced by a factor of up to three, in
realistic cases.

An interesting question is how to identify in a complex fluid mixture
groups of components that may adequately be described as one weakly
polydisperse component each. This is currently being investigated.

REFERENCES

1. L. Yarborough, in *Equations of State in Engineering*, Advances in Chemistry Series, K. C. Chao and R. L. Robinson, eds. (Am. Chem. Soc., Washington, D. C., 1979), pp. 385–439.
2. L. C. Young and R. E. Stephenson, *Soc. Petr. Eng. J.* **32**:727 (1983).
3. L. X. Nghiem and Y. K. Li, Paper SPE 13517, presented at the SPE 1985 Reservoir Simulation Symposium, Dallas, Tex. Feb. 10–13, 1985.
4. M. L. Michelsen, *Fluid Phase Equil.* **9**:1 (1982).
5. A. G. Schlijper, Paper SPE/DOE 12633, presented at the SPE/DOE Fourth Symposium on Enhanced Oil Recovery, Tulsa, Okla. Apr. 15–18, 1984.
6. K. H. Coats, Paper SPE 10512, presented at the Sixth SPE Symposium on Reservoir Simulation, New Orleans, La., Jan. 31–Feb. 3, 1982.
7. K. C. Hong, Paper SPE/DOE 10691, presented at the SPE/DOE Third Symposium on Enhanced Oil Recovery, Tulsa, Okla., Apr. 4–7, 1982.
8. J. A. Gualtieri, J. M. Kincaid, and G. Morrison, *J. Chem. Phys.* **77**:521 (1982).
9. J. J. Salacuse and G. Stell, *J. Chem. Phys.* **77**:3714 (1982).

10. J. M. Kincaid, G. Morrison, and E. Lindeberg, *Phys. Lett.* **96A**:471 (1983).
11. J. G. Briano and E. D. Glandt, *Fluid Phase Equil.* **14**:91 (1983).
12. M. T. Rätzsch and H. Kehlen, *Fluid Phase Equil.* **14**:225 (1983).
13. G. Soave, *Chem. Eng. Sci.* **27**:1197 (1971).
14. D. Y. Peng and D. B. Robinson, *Ind. Eng. Chem. Fund.* **15**:59 (1976).
15. R. L. Cotterman and J. M. Prausnitz, *Ind. Eng. Chem. Process. Des. Dev.* **24**:434 (1985).
16. A. G. Schlijper, SPE Reservoir Engineering (in press).
17. Y. Hirose, Y. Kaware, and M. Kudih, *J. Chem. Eng. Jpn.* **11**:150 (1978).
18. J. Kao, Paper SPE 7605, presented at the 53rd Annual Fall Technical Conference and Exhibition of the Society of Petroleum Engineers of AIME, Houston, Tex., Oct. 1–3, 1978.
19. L. Arselineau, G. Bogdanic, and J. Vidal, *Fluid Phase Equil.* **3**:273 (1979).
20. M. L. Michelsen, *Fluid Phase Equil.* **9**:21 (1982).