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Application of an Extended Lee–Kesler Corresponding-States Technique to Prediction of Vapor–Liquid Equilibria in Multicomponent Mixtures Containing Polar Components[1]

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Mixing rules for an extended Lee–Kesler (ELK) corresponding-states method for prediction of thermodynamic properties are presented. The mixing rules, which do not contain adjustable parameters, permit calculation of vapor–liquid equilibria in mixtures containing one or more polar components. While a single heat-of-mixing datum may be included for strongly associating components, generally only pure component properties are used in the calculations. A comparison of calculated bubble-point pressures and equilibrium vaporization ratios to experimental values is made for 26 ternary mixtures (791 points) and 5 quaternary mixtures (175 points). Bubble-point pressures are predicted quite well with ELK (9.1 % AAD for ternaries and 7.9 % for quaternaries). Composition predictions are difficult to compare in multicomponent systems, but ELK predictions appear to be acceptable on both a system-by-system and an overall AAD basis.

KEY WORDS: corresponding states; mixing rules; multicomponent fluids; polar fluids; vapor–liquid equilibrium.

1. INTRODUCTION

The Lee–Kesler (LK) corresponding-states technique has found widespread use for prediction of thermodynamic properties of nonpolar fluids since its introduction by Lee and Kesler [1]. Recently, Wilding and Rowley [2] extended the applicability of the LK method to pure-component polar

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fluids. Wilding et al. [3] used this extended Lee–Kesler (ELK) method to calculate vapor pressures, and Johnson and Rowley [4] developed mixing rules for prediction of vapor–liquid equilibrium (VLE). Results of these previous studies indicate that ELK can be used reliably and accurately to predict VLE in binary mixtures in addition to pure-component compressibility factors, enthalpy departure functions, and vapor pressures. The purpose of this paper is to determine the applicability of the proposed mixing rules for ELK by comparison of predicted and experimental VLE points in multicomponent systems.

2. ELK

ELK can be viewed as a perturbation about a spherically symmetric reference fluid with respect to size/shape, α, and polar, β, parameters. In terms of the compressibility factor, Z, at some reduced temperature, T_r, and pressure, P_r, this perturbation can be written to linear order as

$$Z(T_r, P_r) = Z^{(0)}(T_r, P_r) + \alpha Z^{(1)}(T_r, P_r) + \beta Z^{(2)}(T_r, P_r) \tag{1}$$

where $Z^{(0)}$ represents the compressibility factor of the simple reference fluid, $Z^{(1)}$ represents a deviation of the compressibility factor of a nonpolar, nonspherical reference fluid from that of the simple fluid at the same T_r and P_r, and $Z^{(2)}$ represents the deviation between the compressibility factor of a polar, nonspherical reference fluid and that of the same fluid without polar interactions. As shown by Wilding and Rowley [2], these deviation functions can be obtained easily at any set of reduced conditions from the definitions

$$Z^{(1)} = \left(\frac{Z_1 - Z_0}{\alpha_1}\right) \tag{2}$$

$$Z^{(2)} = \left\{Z_2 - \left[Z_0 + \alpha_2\left(\frac{Z_1 - Z_0}{\alpha_1}\right)\right]\right\} \Big/ \beta_2 \tag{3}$$

where the individual Z_i are computed at the reduced conditions from an equation of state for reference fluid i. ELK uses the same first two reference fluids as the original LK method and water as the third reference fluid. The equations of state and their constants are available for computer implementation [2] and values of the deviation functions have been tabulated [3] for use in hand calculations.

The size/shape parameter, α, is obtained from a correlation as a function of the radius of gyration [2] or from tabulated values [3]. For nonpolar fluids, the correlation produces α values numerically similar to the acentric factor, and the method reduces to the ordinary LK method.

Similarly, β values have been tabulated for common fluids [3], and additional values may be obtained by application of Eqs. (1) through (3) at any single condition where a liquid density is known.

Application of ELK to other properties is done using appropriate thermodynamic identities which relate those properties to Z. Unfortunately, the same value of β cannot be used for both Z calculations and other properties. Wilding and Rowley [2] showed that a β value derived from a single known vapor pressure (denoted β_{H}) is much better for enthalpy-related properties, including enthalpy and entropy departure functions, fugacity coefficients, and vapor pressure. Tabulated values of β_{H} for use with enthalpic-related properties are also available [3]. Wilding et al. [3] used ELK to calculate vapor pressures and found ELK to be at least as accurate as correlations designed explicitly and exclusively for prediction of vapor pressures of polar fluids. This indicates that with appropriate mixing rules, the method may also be used for VLE calculations.

3. MIXING RULES

The Plocker et al. [5] mixing rules for the Lee–Kesler method (LKP) have found wide acceptance in industry for calculation of VLE. Amazingly, while the LK method itself is not generally applicable to the calculation of properties for polar mixtures, the LKP mixing rules have been used with some success for prediction of VLE for mixtures containing one or more polar components. It does generally fail, however, on very nonideal mixtures, especially when azeotropes are present. An example is shown in Fig. 1; additional examples are also available [6]. It seems appropriate to develop mixing rules for ELK, since the base method itself has been shown to be efficacious for polar fluids.

Recently, mixing rules for ELK applicable to VLE calculations were developed [4]. The following empirical mixing rules were found to be adequate for prediction of binary VLE:

$$T_{\mathrm{cm}}=\left(\frac{T_{\mathrm{cm}}}{P_{\mathrm{cm}}}\right)^{-\eta}\sum_i\sum_j x_i x_j T_{\mathrm{c}ij}\left(\frac{T_{\mathrm{c}ij}}{P_{\mathrm{c}ij}}\right)^{\eta} \tag{4}$$

$$\frac{T_{\mathrm{cm}}}{P_{\mathrm{cm}}}=\sum_i\sum_j x_i x_j \frac{T_{\mathrm{c}ij}}{P_{\mathrm{c}ij}} \tag{5}$$

$$\alpha_{\mathrm{m}}=\left(\frac{T_{\mathrm{cm}}}{P_{\mathrm{cm}}}\right)^{-2/3}\sum_i\sum_j x_i x_j \alpha_{ij}\left(\frac{T_{\mathrm{c}ij}}{P_{\mathrm{c}ij}}\right)^{2/3} \tag{6}$$

$$\beta_{\mathrm{Hm}}=\sum_i\sum_j x_i x_j(\beta_{\mathrm{H}ij}+A_{ij}) \tag{7}$$

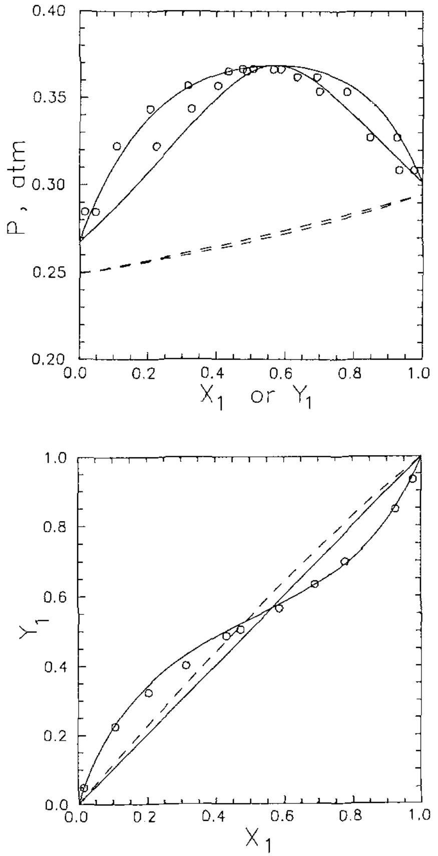

Fig. 1. VLE from experiments (○), ELK predictions (——), and LK predictions (- - -) for the benzene (1) + acetonitrile (2) system at 318.15 K.

The cross terms are evaluated by using the following combining rules:

$$T_{cij} = \sqrt{T_{ci} T_{cj}} \tag{8}$$

$$P_{cij} = \frac{8 T_{cij}}{[(T_{ci}/P_{ci})^{1/3} + (T_{cj}/P_{cj})^{1/3}]^3} \tag{9}$$

$$\alpha_{ij} = \frac{1}{2}(\alpha_i + \alpha_j) \tag{10}$$

$$\beta_{Hij} = \frac{1}{2}(\beta_{Hi} + \beta_{Hj}) \tag{11}$$

When calculating Z for the mixture, β_{m} must be determined instead of β_{Hm}. This is done by replacing β_{H} with β in Eqs. (7) and (11) and setting $A_{ij} = 0$.

In order to maintain ELK as a predictive method, the parameters η and A_{ij} were correlated in terms of pure-fluid properties. This allows VLE calculations to be made without using any experimental VLE data to regress adjustable parameters. The parameter η in Eq. (4) represents the effect of different molecular sizes upon the ratio $T_{\mathrm{c}}/P_{\mathrm{c}}$, an effective molecular volume. It is of interest to note that if $\eta = 0$, differences in molecular size have no effect on the pseudocritical temperature; if $\eta = 1$, the temperature mixing rule becomes equivalent to the Van der Waals one-fluid mixing rule for the energy parameter; and if $\eta = 0.25$, a rule similar to LKP is obtained. Rather than choose a constant for η, Johnson and Rowley [4] chose to let the differences in pure-component α values determine η from the correlation,

$$\eta = \frac{1}{N-1} \sum_{i} \sum_{j>i} (x_i + x_j)\, \eta_{ij} \tag{12}$$

where

$$\eta_{ij} = 0.8501 + 0.3177\, |\alpha_i - \alpha_j| - 1.0389\, |\alpha_i \alpha_j| \tag{13}$$

This correlation was obtained from a regression of VLE data on 28 binary mixtures containing only nonpolar components. Its use in Eq. (4) implies that the mixing rule for T_{cm} is independent of any polarity effects. Thus, the parameter β_{Hm} accounts for all polar effects through its mixing rule.

The parameter A_{ij} is seen from Eq. (7) to be an excess polarity term that accounts for specific deviations from an arithmetic mean of pure-component values. Values for A_{ij} were obtained by regression of VLE data for 59 binary mixtures containing polar components with the constraints $A_{ii} = 0$ and $A_{ij} = A_{ji}$. The following correlations were found to represent A_{ij} adequately.

(i) For pairs in which $\beta_{\mathrm{H}} < 0.89$ (or $\beta_{\mathrm{H}} = 1.0$) for both components,

$$\begin{aligned} A_{ij} = {} & -0.01879 - 0.33925\, |\beta_{\mathrm{H}i} - \beta_{\mathrm{H}j}| - 2.8365 \times 10^{-4}\, H_{ij} \\ & - 3.17035 \times 10^{-8}\, H_{ij}^2 \end{aligned} \tag{14}$$

(ii) For pairs containing at least one component with $\beta_{\mathrm{H}} > 0.89$ (excluding $\beta_{\mathrm{H}} = 1.0$),

$$\begin{aligned} A_{ij} = {} & 0.1476 - 0.58985\, |\beta_{\mathrm{H}i} - \beta_{\mathrm{H}j}| - 1.4575 \times 10^{-4}\, H_{ij} \\ & + 1.7735 \times 10^{-8}\, H_{ij}^2 \end{aligned} \tag{15}$$

where H_{ij} is the heat of mixing in $\mathrm{J \cdot mol^{-1}}$ for the i–j binary pair at 0.50 mole fraction. Without loss of accuracy, β_{H} for water was set to unity and it does not contribute to a change from Eq. (14) to Eq. (15). Alcohols is the main class of components that require Eq. (15). The value of H_{ij} can be set to zero for most binary systems without loss of accuracy. However, it was found that including H_{ij} greatly increased the accuracy for systems containing ketones and amines. In this paper we designate calculations made using a single excess enthalpy to obtain A_{ij} as ELK1 and those obtained with all $H_{ij}=0$ as ELK2.

4. PREDICTION OF VLE

Vapor–liquid equilibrium is more sensitive to the choice of mixing rules than total properties because VLE calculations require determination of partial fugacity coefficients which depend on compositional derivatives of the mixing rules. The VLE calculations reported here were made using the equality-of-partial-fugacities criterion

$$f_i^{\mathrm{L}}=f_i^{\mathrm{V}} \tag{16}$$

Based on Eq. (16), the distribution coefficient, K_i, can be expressed in terms of partial fugacity coefficients as

$$K_i \equiv \frac{y_i}{x_i}=\frac{\phi_i^{\mathrm{L}}}{\phi_i^{\mathrm{V}}} \tag{17}$$

Partial fugacity coefficients can be calculated from the mixture fugacity coefficient using

$$\ln \phi_i=\ln \phi_{\mathrm{m}}-\sum_{k \neq i} x_k\left(\frac{\partial \ln \phi_{\mathrm{m}}}{\partial x_k}\right)_{x_j; j \neq i, k} \tag{18}$$

The composition derivative of the mixture fugacity which appears in Eq. (18) can be written explicitly in terms of mixture departure functions (which are readily calculated with ELK) and derivatives of the four mixing rules with respect to composition:

$$\left(\frac{\partial \ln \phi_{\mathrm{m}}}{\partial x_k}\right)=\frac{1}{T}\left(\frac{H-H^0}{RT_{\mathrm{cm}}}\right)\left(\frac{\partial T_{\mathrm{cm}}}{\partial x_k}\right)+\left(\frac{1-Z}{P_{\mathrm{cm}}}\right)\left(\frac{\partial P_{\mathrm{cm}}}{\partial x_k}\right)$$
$$+\ln \phi^{(1)}\left(\frac{\partial \alpha_{\mathrm{m}}}{\partial x_k}\right)+\ln \phi^{(2)}\left(\frac{\partial \beta_{\mathrm{Hm}}}{\partial x_k}\right) \tag{19}$$

Equations for each of the derivatives in Eq. (19), obtained from the mixing rules used in this study, are available in Ref. 4.

Using the above prescription, Johnson and Rowley [4] computed bubble-point pressures and compositions for 77 binary mixtures containing at least one polar component. Table I summarizes the average absolute percent deviations of pressure (DP) and K values (DK_i) which they obtained. Many of the azeotropic systems were examined point by point, and good agreement with experiment was found in all cases. It is for these very nonideal systems that ELK has its main advantage as can be seen from the typical comparison of ELK and LKP calculations to experimental values in Fig. 1 [6].

Tables II and III show a comparison between ELK and experimental data obtained from Refs. 7 and 8 for ternary and quaternary VLE in mixtures containing at least one polar component. Heats of mixing for use in ELK1 were obtained from Ref. 9. It is interesting to note that the inclusion of a heat of mixing datum does not significantly improve the overall results. Unless very strongly associating components are present the datum is not necessary, and it is much easier to set $H_{ij}=0$, resulting in the following simpler equations for β_H:

(i) For pairs in which $\beta_H < 0.89$ (or $\beta_H = 1.0$) for both components,

$$A_{ij} = -0.01879 - 0.33925\,|\beta_{Hi} - \beta_{Hj}| \tag{20}$$

(ii) For pairs containing at least one component with $\beta_H > 0.89$ (excluding $\beta_H = 1.0$),

$$A_{ij} = 0.1476 - 0.58985\,|\beta_{Hi} - \beta_{Hj}| \tag{21}$$

Comparison of ELK2 (with no heat of mixing data) to LKP on a system-by-system basis indicates that ELK predicts bubble-point pressures better in 25 of the 26 ternaries tested and in 4 of the 5 quaternaries. An example is shown in Fig. 2 for the chloroform + methanol + benzene

Table I. Summary of Binary VLE Results[a]

	ELK1	ELK2	LKP
DP	8.1	14.7	18.9
DK_1	8.5	14.3	20.0
DK_2	13.7	21.0	28.3

[a] $DP = (100\,\%/N)\sum_{k=1}^{N}(|P_{\text{calc}} - P_{\text{exp}}|/P_{\text{exp}})$.
$DK_i = (100\,\%/N)\sum_{k=1}^{N}(|K_{i,\text{calc}} - K_{i,\text{exp}}|/K_{i,\text{exp}})$.

Table II. Average Absolute Percentage Deviation of VLE Predictions for Ternary Mixtures Containing Polar Components[a]

System	No. pts.	ELK1				ELK2				LKP			
		DP	DK_1	DK_2	DK_3	DP	DK_1	DK_2	DK_3	DP	DK_1	DK_2	DK_3
Methanol + ethanol + water	20	2.2	6.9	9.8	14.6	3.0	6.7	14.1	15.5	14.7	8.6	17.9	28.1
2-Propanone + methanol + water	54	15.2	21.0	18.1	12.7	11.0	12.5	13.2	12.1	13.6	6.8	10.5	18.5
2-Propanone + acetonitrile + water	30	27.1	24.4	14.5	22.3	9.0	36.7	14.2	17.5	18.7	16.8	15.2	26.3
2-Propanone + chloroform + methanol	36	13.6	14.2	9.2	26.9	18.4	18.3	13.8	11.4	8.8	20.6	12.5	19.6
Carbon tetrachloride + benzene + 2-propanol	39	3.9	2.9	4.4	6.2	2.8	3.7	5.2	10.8	26.8	20.5	20.6	44.1
Methyl acetate + 2-propanol + benzene	32	10.1	10.6	13.9	5.8	2.2	6.6	4.8	7.3	18.8	17.4	27.8	23.3
2-Butanone + *n*-heptane + 1-butanol	14	3.5	8.9	13.3	17.0	11.5	12.5	24.3	16.9	22.0	13.9	32.4	26.5
Cyclohexane + toluene + 1-butanol	28	11.9	10.4	10.9	21.9	16.2	11.4	9.7	20.8	29.3	18.3	15.1	28.9
2-Propanone + carbon tetrachloride + benzene	57	4.1	10.5	6.3	10.0	6.0	13.3	7.1	9.9	13.3	21.5	11.6	10.8
2-Propanone + chloroform + 2,3-dimethylbutane	19	3.5	7.1	11.0	4.5	10.2	19.3	39.7	32.9	11.1	16.0	34.2	31.3
Dichloromethane + 2-butanone + water	25	4.9	56.2	117.7	44.9	5.6	59.4	115.1	48.0	10.8	64.3	111.5	63.3
Water + methanol + methyl acetate	32	10.3	25.7	11.6	9.3	6.0	22.4	14.3	6.1	24.1	35.9	17.4	12.9
Ethanol + water + 2-propanol	27	3.6	3.6	4.1	8.6	5.3	3.4	5.7	10.6	20.8	15.3	30.4	14.8

2-Butanol + 2-butanone + water	33	16.9	33.7	33.0	22.4	9.4	31.3	14.4	25.2	27.0	30.2	16.9	31.2
Diethyl ether + ethanol + water	33	19.4	88.4	25.7	21.5	20.8	90.4	25.2	22.2	39.3	36.5	13.4	29.4
2-Propanone + 2-butanone + water	37	15.7	17.2	13.2	19.3	6.9	22.5	19.6	15.2	16.4	19.8	14.4	28.5
Cyclohexane + 2-propanol + water	20	23.2	36.0	46.5	23.3	27.3	40.5	50.4	24.0	43.5	53.2	70.8	36.3
Chloroform + methanol + benzene	35	7.0	10.3	7.6	17.0	8.1	20.9	11.6	19.3	31.7	36.6	36.1	30.5
2-Propanone + methanol + 2-propanol	27	12.5	34.0	18.4	22.9	6.6	18.6	13.1	16.1	7.2	23.8	9.0	18.5
2-Propanone + methanol + cyclohexane	21	6.6	31.2	12.9	28.6	22.3	18.9	13.1	16.1	40.4	19.7	28.8	46.8
Carbon tetrachloride + ethanol + benzene	36	1.5	4.9	6.8	8.3	5.0	8.1	11.9	10.6	36.9	22.8	47.6	29.4
Ethanol + chloroform + *n*-hexane	36	5.0	21.1	15.3	15.6	4.8	27.1	12.9	21.5	31.7	42.0	10.6	18.8
2-Propanone + ethanol + *n*-hexane	21	2.8	21.4	9.1	18.2	17.5	15.1	25.5	25.5	35.3	17.0	28.7	31.6
Methyl acetate + 2-propanol + benzene	32	9.5	9.6	16.7	7.7	2.2	6.7	4.8	7.4	18.8	17.4	27.8	22.7
2-Butanone + benzene + 2-propanol	19	10.1	14.7	5.7	11.3	2.9	13.0	5.9	4.0	21.5	21.7	17.6	25.1
Benzene + toluene + 1-butanol	28	43.4	26.1	25.4	64.3	9.5	28.3	30.6	48.1	10.1	29.1	32.5	46.0
Total	791	11.2	20.8	17.4	17.9	9.1	21.4	18.2	17.9	22.2	24.5	25.0	27.2

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[a] See Table I, footnote *a*, for definitions of column heads.

Table III. Average Absolute Percentage Deviation of VLE Predictions for Quaternary Mixtures Containing Polar Components[a]

System	No. pts.	ELK1					ELK2					LKP				
		DP	DK_1	DK_2	DK_3	DK_4	DP	DK_1	DK_2	DK_3	DK_4	DP	DK_1	DK_2	DK_3	DK_4
Dimethylbutane + methanol + 2-propanone + chloroform	5	3.9	23.3	7.3	19.8	10.6	9.5	46.1	7.9	29.0	42.8	29.8	36.9	23.0	47.6	42.6
Methanol + ethanol + methyl acetate + ethyl acetate	4	19.3	8.3	8.0	9.1	6.4	11.5	6.8	4.7	8.2	6.6	7.5	9.9	6.5	6.8	10.0
Ethanol + chloroform + 2-propanone + *n*-hexane	84	4.2	17.9	16.0	17.8	22.7	7.5	17.0	45.9	20.7	38.7	24.5	32.0	40.4	24.1	25.4
1,2-Dichloroethane + 1-propanol + toluene + 2-propanone	24	11.5	13.0	14.5	11.8	14.2	5.5	10.8	12.7	14.9	15.3	10.7	14.0	37.5	24.1	25.4
Cyclohexane + benzene + 2-propanol + 2-butanone	58	4.6	26.5	6.3	20.6	25.9	9.2	31.7	10.7	18.0	16.0	25.1	26.9	17.2	35.8	24.3
Total	175	5.7	20.0	12.2	17.8	21.9	7.9	21.6	27.6	18.9	27.4	22.6	27.3	31.0	31.5	29.8

[a] See Table I, footnote *a*, for definitions of column heads.

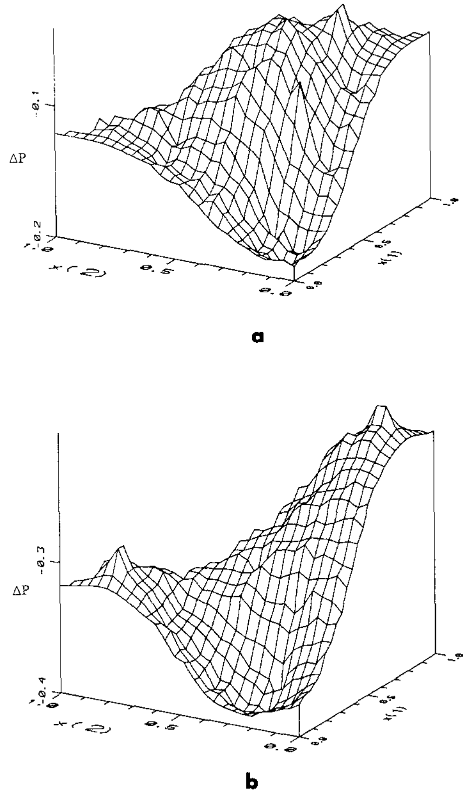

Fig. 2. Fractional deviation surface of predicted bubble-point pressures from experimental values for the chloroform (1) + methanol (2) + benzene (3) system at 328 K using (a) ELK and (b) LKP.

system, in which ELK shows very little error in the chloroform-rich region. While LKP shows a similar qualitative behavior, the whole surface is shifted down with poor agreement even in the chloroform-rich region.

Likewise, ELK predicts *K* values better for most individual systems, although the improvement is not as obvious. In this case, looking at overall percentage errors in *K* values may be somewhat misleading because a relatively small absolute error in a small *K* value may result in a rather large relative percentage error, overshadowing good agreement in other regions. Nor are trends noticeable from tabulated deviations. For example, Figs. 3 and 4 show ELK1 and LKP predicted VLE tie lines that emanate from selected liquid compositions for chloroform + methanol + benzene and carbon tetrachloride + ethanol + benzene mixtures. As expected from

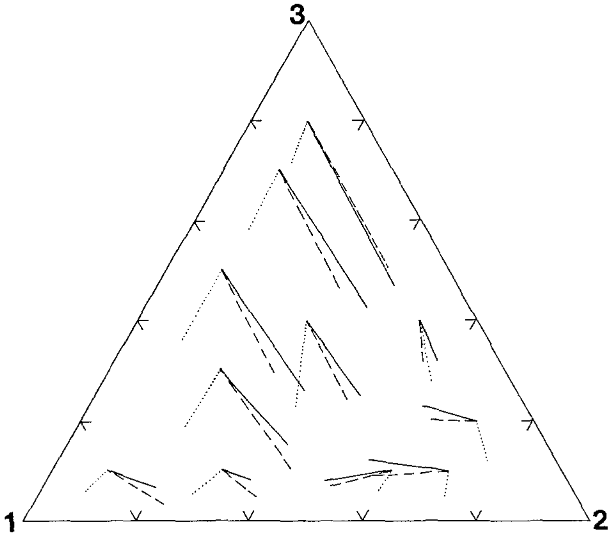

Fig. 3. VLE tie lines obtained from experiment (——), ELK predictions (– – –), and LKP predictions (·········) for chloroform (1) + methanol (2) + benzene (3) mixtures at 328.15 K.

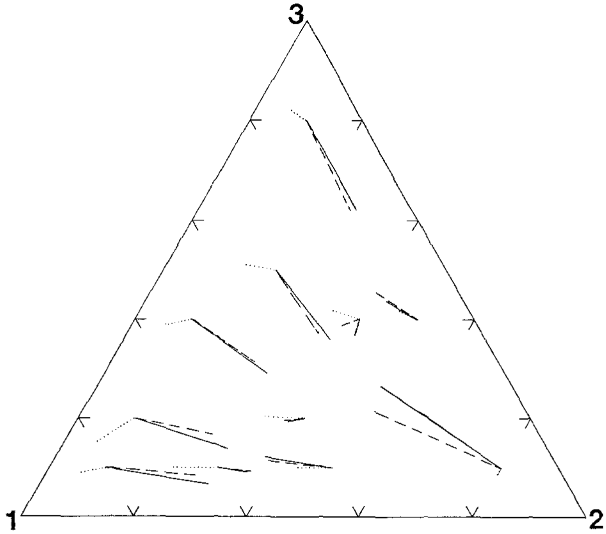

Fig. 4. VLE tie lines obtained from experiment (——), ELK predictions (– – –), and LKP predictions (·········) for carbon tetrachloride (1) + ethanol (2) + benzene (3) mixtures at 323.15 K.

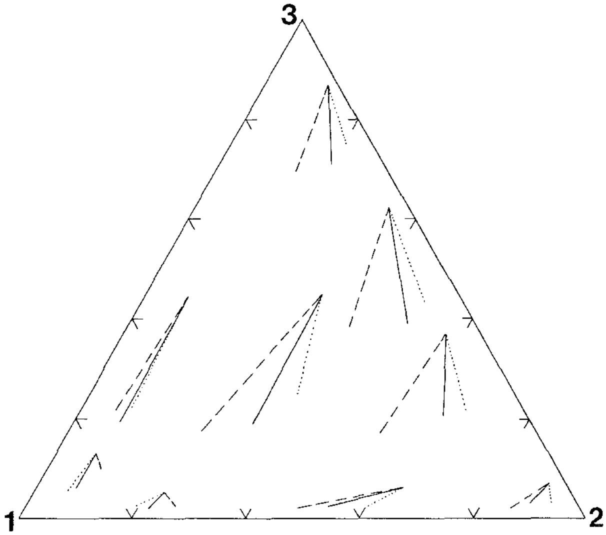

Fig. 5. VLE tie lines obtained from experiment (——), ELK predictions (– – –), and LKP predictions (·········) for 2-propanone (1) + methanol (2) + 2-propanol (3) mixtures at 328.15 K.

Table II, ELK1 does significantly better, but the fact that the LKP tie lines actually extend in the wrong direction in most regions is not apparent from the tabulated percentage errors. Similarly, ELK errors for the 2-propanone + methanol + 2-propanol system are comparable to the LKP errors for the above systems, but the tie lines shown in Fig. 5 exhibit qualitatively better trends.

5. CONCLUSIONS

In this work, ELK VLE predictions have been examined in ternary and quaternary mixtures using recently proposed mixing rules. While the mixing rules permit input of a binary heat-of-mixing datum for strongly associating mixtures, overall predictions are not significantly improved. For ease of use, Eqs. (20) and (21), which utilize only pure-component data, seem preferable.

The results of this study indicate that ELK achieves a significant improvement in bubble-point pressures over LKP. The improvement in K values is also significant but not nearly as substantial. As was the case in the binary systems, polar systems that exhibit large nonidealities including azeotropes were particularly difficult to predict with the original LK method; it is for these types of systems that the extended Lee–Kesler method is particularly valuable.

REFERENCES

1. B. I. Lee and M. G. Kesler, *AIChE J.* **21**:510 (1975).
2. W. V. Wilding and R. L. Rowley, *Int. J. Thermophys.* **7**:525 (1986).
3. W. V. Wilding, J. K. Johnson, and R. L. Rowley, *Int. J. Thermophys.* **8**:717 (1987).
4. J. K. Johnson and R. L. Rowley, *Fluid Phase Equilib.* **44**:255 (1989).
5. U. Plocker, H. Knapp, and J. Prausnitz, *Ind. Eng. Chem. Process Des. Dev.* **17**:324 (1978).
6. J. K. Johnson, M.S. thesis (Brigham Young University, Provo, Utah, 1987).
7. J. Gmehling and U. Onken, *Chemistry Data Series, Vapor-Liquid Equilibrium Data Collection, Vol. 1* (DECHEMA, Frankfurt, 1977), Parts 1 and 2a.
8. J. Gmehling, U. Onken, and W. Arlt, *Chemistry Data Series, Vapor-Liquid Equilibrium Data Collection, Vol. 1* (DECHEMA, Frankfurt, 1978), Part 2b.
9. J. J. Christensen, R. W. Hanks, and R. M. Izatt, *Handbook of Heats of Mixing* (Wiley, New York, 1982).