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PREDICTION OF THE EXCESS ENTHALPIES OF THE CARBON DIOXIDE + HEXANE + TOLUENE SYSTEM AT HIGH TEMPERATURES AND PRESSURES USING THE PENG–ROBINSON EQUATION OF STATE *

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ABSTRACT

Excess enthalpies H^E for the carbon dioxide(1) + n-hexane(2) + toluene(3) system at temperatures between 308.15 and 573.15 K and pressures of 7.50 and 12.50 MPa were predicted using the Peng–Robinson equation of state. Values of the interaction parameters were determined from binary experimental data. Results were compared with ternary H^E data and with predictions previously obtained at the same conditions of temperature and pressure using the Toop equation. The Peng–Robinson equation is shown to provide a good description of ternary H^E values in both the one-phase and two-phase regions.

INTRODUCTION

Excess enthalpies H^E for the carbon dioxide(1) + n-hexane(2) + toluene(3) system have been measured at five temperatures between 308.15 and 573.15 K and at pressures of 7.50 and 12.50 MPa [1–5]. H^E values for the binary systems consisting of carbon dioxide and various hydrocarbons at pressures usually exceeding the critical pressures of both components and over temperature ranges often spanning the critical temperatures of both components have been reported previously by the Brigham Young University group [6–10]. The location of the experimental conditions of measurement for the carbon dioxide(1) + n-hexane(2) + toluene(3) system with respect to the critical points of the pure components and the three binary critical loci

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* Dedicated to Professor James. J. Christensen (deceased 5 September 1987) in memory of his contribution to innovation in calorimetry.

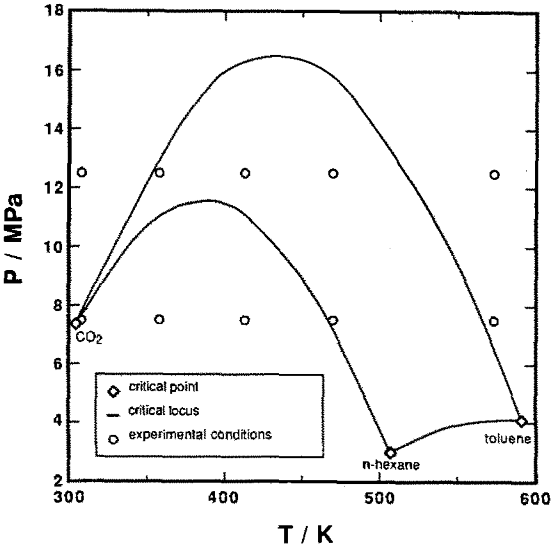

Fig. 1. Plot of p vs. T showing the critical points of the three pure components, the three binary critical loci and the (p,T) coordinate where the experimental data were taken.

[11–13] is given in Fig. 1. Data for the critical locus of the ternary system are not available. It is very likely that the ternary locus consists of a convex, smooth surface stretching from one binary locus to another. The large changes in sign and magnitude of the ternary H^E values have been interpreted as a consequence of changes in the composition of the mixture and in the conditions of temperature and pressure [1–5]. Correlation methods have also been examined and a partial differential approximant has been found to represent most accurately the ternary contribution to the excess enthalpy. Ternary H^E values have also been predicted using Toop's equation [14] from the values of excess enthalpies of the three related binary systems at the same temperature and pressure.

In this study, the predictions of Toop's equation are compared with those obtained using a cubic equation of state, the Peng–Robinson (PR) equation [15], which has been widely used to predict vapor–liquid equilibria (VLE) of binary systems consisting of carbon dioxide and various hydrocarbons at high temperatures and pressures.

TOOP'S EQUATION

In a previous paper [16], we examined methods of predicting ternary excess enthalpies from the excess enthalpies of three binary mixtures involved in 42 sets of ternary data. The asymmetric equations with respect to the numbering of components proposed by Toop [14], Scatchard et al. [17]

and Hillert [18] were shown to provide accurate and almost coincident predictions for most systems studied. Toop's equation was shown to be adequate for systems showing a combination of endothermic and exothermic mixing. Since this is the case for the carbon dioxide(1) + n-hexane(2) + toluene(3) system, Toop's equation was used to calculate H^E values for this system. The equation is given by

$$\begin{aligned} H^{\mathrm{E}}\ (\mathrm{J\ mol^{-1}}) = {} & x_1x_2\left\{\sum_0^n A_k(2x_1-1)^k\right\}\Bigg/\left\{\sum_0^m B_l(2x_1-1)^l\right\} \\ & + x_1x_3\left\{\sum_0^{n'} A'_k(2x_1-1)^k\right\}\Bigg/\left\{\sum_0^{m'} B'_l(2x_1-1)^l\right\} \\ & + x_2x_3\left\{\sum_0^{n''} A''_k(x_2-x_3)^k/(x_2+x_3)^k\right\} \\ & \Bigg/\left\{\sum_0^{m''} B''_l(x_2-x_3)^l/(x_2+x_3)^l\right\} \end{aligned} \tag{1}$$

$B_0,\ B'_0,\ B''_0 = 1$

where x_1, x_2 and x_3 are the mole fractions of components 1, 2 and 3 respectively, and A_k, B_l, A'_k, B'_l, A''_k and B''_l are the adjustable coefficients of the n/m, n'/m' and n''/m'' Padé approximants. These approximants are used to represent excess enthalpies for the three binary mixtures involved. A rule was also given in ref. 16 for selecting component 1 for any kind of ternary mixture. A procedure for applying Toop's equation for systems such as carbon dioxide(1) + n-hexane(2) + toluene(3) in which two-phase regions may appear has been developed [1–5].

PENG–ROBINSON EQUATION

The Peng–Robinson equation of state [15] is given by

$$P = \frac{RT}{v-b} - \frac{a\alpha}{v(v+b)+b(v-b)} \tag{2}$$

where

$$a = 0.45724\ (R^2T_c^2/P_c) \tag{3}$$

$$b = 0.07780\ (RT_c/P_c) \tag{4}$$

and α is a dimensionless function of the reduced temperature and acentric factor. For mixtures

$$a\alpha = \sum_i \sum_j x_i x_j (1-\delta_{ij})(\alpha_i a_i \alpha_j a_j)^{\frac{1}{2}} \tag{5}$$

and

$$b = \sum_i x_i b_i \tag{6}$$

The binary interaction parameter δ_{ij} is usually determined from experimental binary data. The PR equation of state with the combination rules given by eqns. (5) and (6) has been successfully used for vapor–liquid equilibrium calculations of carbon dioxide(1) + hydrocarbon(2) mixtures at high temperatures and pressures [19–24]. Occhiogrosso et al. [21] have shown that for VLE calculations of the carbon dioxide(1) + toluene(2), carbon dioxide(1) + *m*-xylene(2) and carbon dioxide(1) + cumene(2) systems better results are obtained when eqn. (6) is substituted by

$$b = \sum_i \sum_j x_i x_j (1 - \eta_{ij})(b_i + b_j)/2 \tag{7}$$

where η_{ij} is a size parameter which accounts for the size difference between carbon dioxide and the aromatic hydrocarbons.

The PR equation of state has been successfully used for other calculations involving carbon dioxide and hydrocarbon mixtures at high temperatures and pressures such as the modeling of supercritical fluid extraction using CO_2 [25,26].

In an effort to improve the calculations of VLE and solubilities at high pressures or for highly non-ideal mixtures, density-dependent combination rules which can be used for cubic equations of state have been proposed in recent years [23–33]. The use of these combination rules renders the equation more complex: more interaction parameters may be required or the equation may become quartic in volume.

As to the calculation of excess enthalpies using an equation of state, it is necessary to evaluate first the enthalpy departure $H - H^\star$ which is given by

$$H - H^\star = RT(z-1) + \int_\infty^v \left\{ T\left(\frac{\partial P}{\partial T}\right)_v - P \right\} \mathrm{d}v \tag{8}$$

where z is the compressibility factor. For a fluid which follows the PR equation of state, eqn. (8) becomes

$$H - H^\star = RT(z-1) + \frac{T(\mathrm{d}a/\mathrm{d}T) - a}{2^{3/2}b} \ln\left(\frac{z + 2.414B}{z - 0.414B}\right) \tag{9}$$

where B is given by

$$B = \frac{bP}{RT} \tag{10}$$

The excess enthalpy may then be calculated in terms of the enthalpy departures as

$$H^{\mathrm{E}} = H - H^\star - \sum_i x_i (H - H^\star)_i \tag{11}$$

Application of eqn. (11) to obtain accurate values of H^E is difficult because it represents a difference between two large and nearly equal terms. Consequently, cubic equations of state other than the PR equation have been used with mixed success to calculate H^E for binary mixtures [34–36]. Although binary interaction parameters are considered to be adjustable parameters, a small error in any term of eqn. (11) due to inadequacies of the equation of state or of the combination rules may lead to large errors in H^E. As far as we know, calculations of H^E in the two-phase regions, which are a common occurrence for systems consisting of carbon dioxide and a hydrocarbon at high temperature and pressure, have never been attempted.

For ternary systems, Gagné et al. [37] have performed calculations for the system carbon dioxide(1) + methane(2) + ethylene(3) using the Benedict–Webb–Rubin and the Redlich–Kwong equations of state at 293.15, 305.15 and 313.15 K and at 1.1, 1.5 and 3.5 MPa. Ternary mixtures at these conditions of temperature and pressure are in the gas state and values of H^E are very small and endothermic (H^E is always lower than 200 J mol^{-1}). Since only binary interaction parameters are required to use equations of state for multicomponent systems, predictions can be made for these kinds of mixtures if the parameters are determined solely from binary data. Gagné et al. used the values of binary interaction parameters obtained from binary data as initial values for the optimization procedure, leading to the interaction parameter values which represent the ternary system.

Despite its success in VLE calculations and supercritical fluid extraction modeling, the PR equation of state has rarely been used to calculate H^E. For instance, Hamam et al. [38] have shown that this equation may adequately represent the excess enthalpies of binary liquid mixtures consisting of an *n*-alkane and a hexane isomer.

Our purpose in this study is to show that the PR equation may be successfully used to predict ternary excess enthalpies for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system at the conditions of temperature and pressure indicated in Fig. 1. Besides the difficulties we have already pointed out in relation to the application of eqn. (11), it should be taken into account that additional difficulties are introduced because of the changes in the sign and magnitude of H^E with temperature, pressure and composition (values of H^E range from 7.1 kJ mol^{-1} to -4.6 kJ mol^{-1}) and because the isotherms for binary and ternary mixtures very often traverse two-phase regions.

RESULTS AND DISCUSSION

Ternary H^E data were determined using CO_2 in one pump of the flow calorimeter and a mixture of *n*-hexane + toluene in the other pump. Three mixtures with *n*-hexane mole fractions of 0.25, 0.50 and 0.75 were used. The

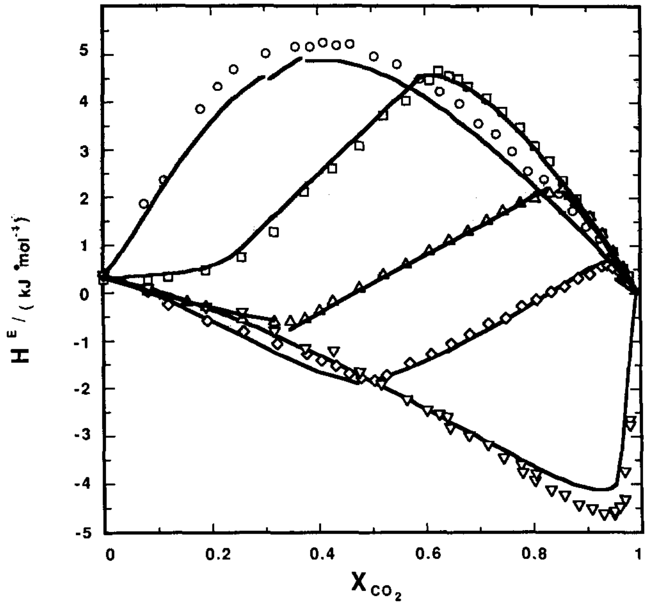

Fig. 2. Plot of H^E against CO_2 mole fraction for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system at 7.50 MPa as a function of temperature when the ratio between the *n*-hexane and toluene mole fractions is three: ▽, 308.15 K; ◇, 358.15 K, △, 413.15 K; □, 470.15 K; ○, 573.15 K; ———, predicted from Toop's equation.

detailed procedure and the results for H^E calculations using Toop's equation have been described in refs. 1–5. Figures 2 and 3 show the H^E vs. CO_2 mole fraction diagrams for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system when the ratio between the *n*-hexane and toluene mole fractions is three ($x_2/x_3 = 3$) and the pressures are 7.50 and 12.50 MPa respectively. The experimental points plotted in Figs. 2 and 3 are approximately one-third of the points determined at each pressure. Full lines were obtained using Toop's equation to calculate ternary H^E. Similar figures are obtained for the other two series of H^E determinations with $x_2/x_3 = 1$ and $x_2/x_3 = 1/3$. The discontinuities sometimes observed in the predicted curves are due to the switching from the one-phase region equation to the two-phase region equation.

The accuracy of the predictions is satisfactory. The ratio between the standard deviation for predictions and the maximum absolute value of H^E was found to be 4% or less in most cases. Values for the standard deviation σ_{Toop} between experimental and predicted values of ternary H^E, together with the maximum and minimum values of each set of ternary H^E data (H^E_{max} and H^E_{min}) are given in Table 1.

H^E calculations for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) systems using the Peng–Robinson equation in the one-phase region were carried out as follows. Binary H^E data taken for the three related binary

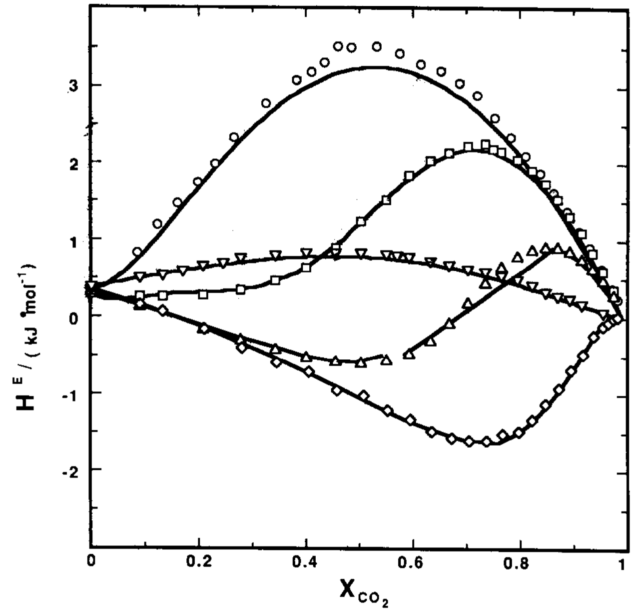

Fig. 3. Plot of H^E against CO_2 mole fraction for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system at 12.50 MPa as a function of temperature when the ratio between the *n*-hexane and toluene mole fractions is three: ▽, 308.15 K; ◇, 358.15 K; △, 413.15 K; □, 470.15 K; ○, 573. 15 K; ———, predicted from Toop's equation.

systems at the same temperatures and pressures at which the ternary data were taken [6–10] were used to obtain values for the binary interaction parameters of the combination rules. Equation (5) was used as combination

TABLE 1

Conditions of determination of H^E data for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system, minimum and maximum values of H^E and standard deviations σ between experimental values and those predicted using Toop's equation and the Peng–Robinson (PR) equation

T (K)	P (MPa)	H^E_{min} (J mol^{-1}),	H^E_{max} (J mol^{-1})	σ_{Toop} (J mol^{-1})	σ_{PR} (J mol^{-1})
308.15	7.50	−4650	140	240	250
	12.50	−50	800	43	30
358.15	7.50	−2100	600	95	130
	12.50	−2100	250	67	130
413.15	7.50	−900	2200	80	160
	12.50	−1000	1000	100	130
470.15	7.50	150	4700	120	160
	12.50	90	2400	84	61
573.15	7.50	300	7100	490	340
	12.50	250	4300	190	150

TABLE 2

Values of the interaction parameters δ_{ij} and η_{ij} determined from H^{E} data taken for the three binary systems related to the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system

T (K)	P (MPa)	$\delta_{12} \times 10$	$\eta_{12} \times 10$	$\delta_{13} \times 10$	$\eta_{13} \times 10$	$\delta_{23} \times 10$	$\eta_{23} \times 10$
308.15	7.50	−0.6861	−1.684	0.6025	−0.4544	0.03913	−0.1228
	12.50	0.6323	−7.290	1.009	−0.2412	−0.03228	−0.1782
358.15	7.50	0.3866	−0.6263	0.1294	−0.6777	0.1239	−0.0390
	12.50	0.2776	−8.171	0.4971	−0.4958	0.2845	0.1187
413.15	7.50	0.2441	−0.2914	−0.08842	−0.6387	0.1177	−0.03079
	12.50	0.7790	−0.1833	−0.5845	−0.2652	0.04072	0.1033
470.15	7.50	1.417	0.4398	0.3351	−0.5211	0.06516	−0.05403
	12.50	1.338	0.3629	0.5716	−0.1803	0.04195	−0.08069
573.15	7.50	2.203	−0.8046	2.863	0.3286	−0.1509	−0.1340
	12.50	2.277	1.109	0.9095	0.3320	0.06230	0.05180

rule for a. Equation (6) and (7) were used as combination rules for b. Better results were obtained when eqn. (7) was used. Table 2 lists the values of δ_{ij} and η_{ij} obtained by least-squares fitting of binary H^{E} data to eqn. (11) with $i = 2$. Values of a and b for the ternary mixtures were calculated using the values of δ_{ij} and η_{ij} determined solely from binary data. Enthalpy departures for the ternary mixtures were evaluated by substitution of the values of a and b thus obtained into eqn. (9). Values of the ternary excess enthalpies at a fixed condition of temperature and pressure were then predicted from eqn. (11) with $i = 3$.

H^{E} calculations for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system using the Peng–Robinson equation in the two-phase region were evaluated as the sum of two contributions

$$H^{\mathrm{E}} = H_{\mathrm{l}}^{\mathrm{E}} n_{\mathrm{l}} + H_{\mathrm{g}}^{\mathrm{E}} n_{\mathrm{g}} \tag{12}$$

where $H_{\mathrm{l}}^{\mathrm{E}}$ and $H_{\mathrm{g}}^{\mathrm{E}}$ are the excess enthalpies of the liquid and vapor mixtures respectively, and n_{l} and n_{g} are the amounts of components in the liquid and vapor mixtures respectively. Values of $H_{\mathrm{l}}^{\mathrm{E}}$ and $H_{\mathrm{g}}^{\mathrm{E}}$ were obtained as described above for the one-phase region. The mole fractions of the liquid and vapor mixtures in equilibrium (x_{l} and x_{g}) were determined as follows: values of n_{l} and n_{g} are related to x_{l} and x_{g} by the equations

$$x_{\mathrm{l}} n_{\mathrm{l}} + x_{\mathrm{g}} n_{\mathrm{g}} = x(n_{\mathrm{l}} + n_{\mathrm{g}}) \tag{13}$$

$$(1 - x_{\mathrm{l}}) n_{\mathrm{l}} + (1 - x_{\mathrm{g}}) n_{\mathrm{g}} = (1 - x)(n_{\mathrm{l}} + n_{\mathrm{g}}) \tag{14}$$

where x, x_{l} and x_{g} are mole fractions of the same component. Vapor–liquid equilibrium data for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system are not available. In general, two of the boundaries of the two-phase region in the ternary system are known (those corresponding to the two-phase

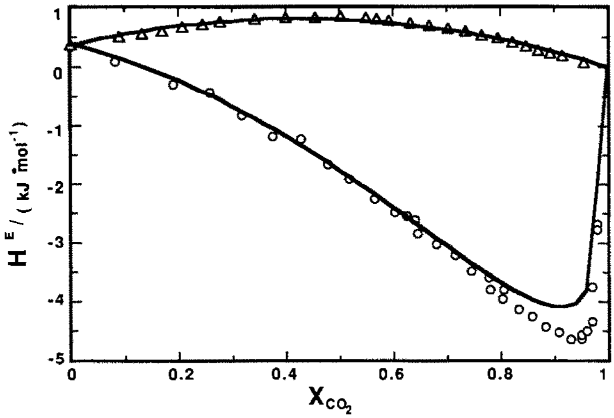

Fig. 4. Plot of H^E against CO_2 mole fraction for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system at 308.15 K as a function of pressure when the ratio between the *n*-hexane and toluene mole fractions is three: ○, 7.50 MPa; △, 12.50 MPa; ——, predicted from PR equation.

regions of the carbon dioxide(1) + *n*-hexane(2) and carbon dioxide(1) + toluene(2) systems respectively) and the other two boundaries are unknown. These boundaries are assumed to be straight lines connecting the limits of the boundaries of the two-phase region in the binary systems carbon dioxide(1) + *n*-hexane(2) and carbon dioxide(1) + toluene(2) at the same temperature and pressure. The intersections between these estimated boundaries and the lines representing the three series of H^E determinations with $x_2/x_3 = 3$, 1 or $\frac{1}{3}$ provide the values of x_l and x_g. This estimation procedure was also employed for predictions made using Toop's equation and is described in refs. 1–5. In some cases the estimated boundaries seem to be adequate. In other cases, the inadequacies of the estimated boundaries seem to be a reason for the high deviations observed between experimental and predicted values of ternary H^E. Two-phase regions appear at 308.15 K and 7.50 MPa, at 358.15, 413.15 and 470.15 K at both 7.50 MPa and 12.50 MPa, and at 573.15 K and 7.50 MPa.

In Figs. 4–8, the same data as shown in Figs. 2 and 3 are regrouped, so that two isobars are collected at each temperature. Full lines in Figs. 4–8 are obtained using the PR equation to calculate ternary H^E. Similar figures are obtained for the other two series of H^E determinations with $x_2/x_3 = 1$ and $x_2/x_3 = \frac{1}{3}$.

The accuracy of the predictions can be considered to be satisfactory. The ratio between the standard deviation for predictions and the maximum absolute value of H^E was found to be 5% or less in most cases. Values for the standard deviation σ_{PR} between experimental and predicted values of ternary H^E are given in Table 1.

Comparison with results obtained using Toop's equation indicates that for six of the ten sets of data studied, PR equation deviations are of the

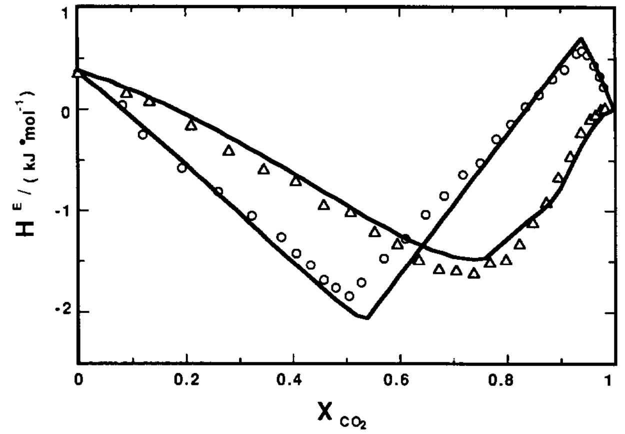

Fig. 5. Plot of H^E against CO_2 mole fraction for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system at 358.15 K as a function of pressure when the ratio between the *n*-hexane and toluene mole fractions is three: ○, 7.50 MPa; △, 12.50 MPa; ———, predicted from PR equation.

same magnitude or lower than Toop's equation deviations. Although the accuracy of the predictions made is not substantially improved by the use of the PR equation of state, one should not forget that the use of Toop's equation is limited to the prediction of H^E, while the PR equation of state enables us to evaluate other thermodynamic properties of the mixture such as vapor–liquid equilibria, solubilities, etc.

We may conclude that the PR equation of state approach provides a better description of the behavior of carbon dioxide(1) + *n*-hexane(2) +

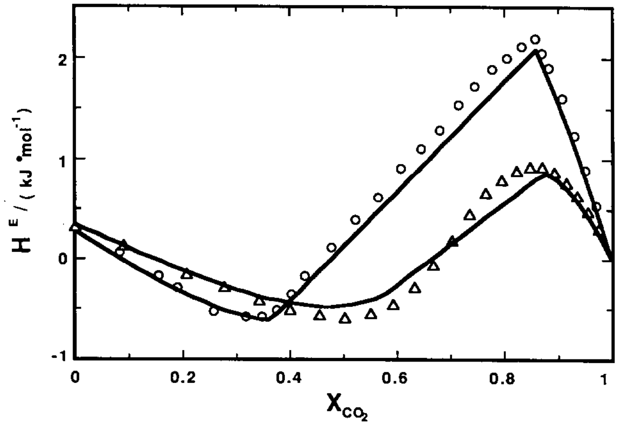

Fig. 6. Plot of H^E against CO_2 mole fraction for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system at 413.15 K as a function of pressure when the ratio between the *n*-hexane and toluene mole fractions is three: ○, 7.50 MPa; △, 12.50 MPa; ———, predicted from PR equation.

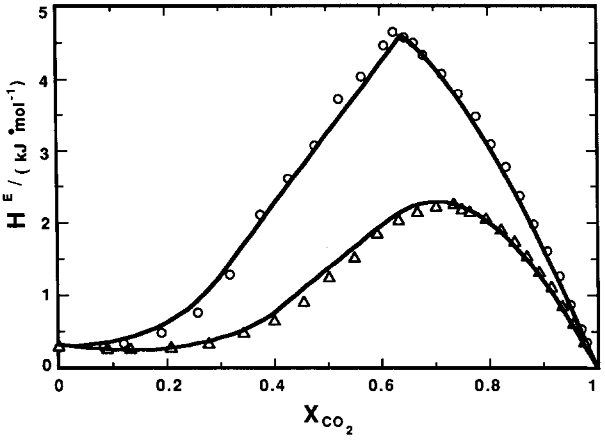

Fig. 7. Plot of H^E against CO_2 mole fraction for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system at 470.15 K as a function of pressure when the ratio between the *n*-hexane and toluene mole fractions is three: ○, 7.50 MPa; △, 12.50 MPa; ———, predicted from PR equation.

toluene(3) mixtures at high temperatures and pressures. Further improvement of this description might be attained by using density-dependent combination rules which have been shown to improve VLE calculation at high pressures.

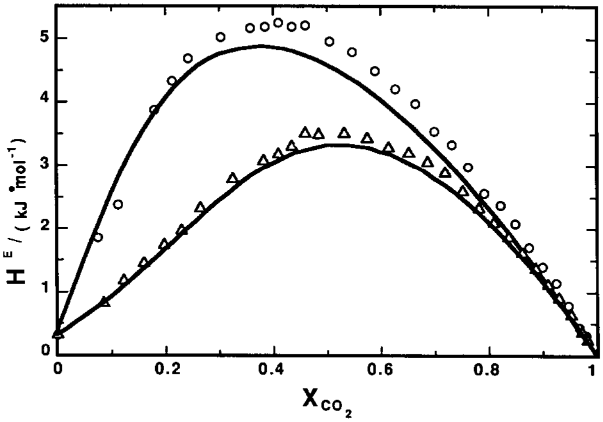

Fig. 8. Plot of H^E against CO_2 mole fraction for the carbon dioxide(1) + *n*-hexane(2) + toluene(3) system at 573.15 K as a function of pressure when the ratio between the *n*-hexane and toluene mole fractions is three: ○, 7.50 MPa; △, 12.50 MPa; ———, predicted from PR equation.

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