Comparison between Experimental and Theoretical Estimations of the Thermal Expansion, Concentration Expansion Coefficients, and Viscosity for Binary Mixtures under Pressures up to 20 MPa[†]

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In this work, we have measured the densities of binary mixtures of *n*-dodecane, 1-phenyl-2-methylpropane, and 1,2,3,4-tetrahydronaphthalene for pressures varying from (0.1 to 20) MPa at an average temperature of 25 °C. By a derivative method, we have determined the thermal expansion and concentration expansion coefficients for binary mixtures of equal mass fraction. In addition, viscosities have been measured and compared with theoretical estimates. To accurately predict the thermal expansion and concentration expansion coefficients, the densities of the binary mixtures were calculated using PC-SAFT, Peng-Robinson, and volume translated Peng-Robinson equations of state. The comparison with measured densities showed that PC-SAFT has a better agreement with experimental data than the other equations of state. From calculated densities we evaluated the thermal expansion and concentration expansion variation coefficients. We have found that PC-SAFT gives a suitable prediction for the two derivative properties unlike the two other equations of state. The combination of the model of Lohrenz-Bray-Clark for the viscosity of liquid mixtures and the densities calculated with the three equations of state gave a poor prediction of the viscosities of the binary mixtures.

Introduction

The phenomenon of thermal diffusion was discovered by Ludwig[1] and later more deeply analyzed by Soret.[2] The physical quantity that describes this phenomenon is known as the Ludwig−Soret or Soret coefficient. Since its discovery, numerous investigations and observations have been attributed to this phenomenon in diverse fields such geology, biology, industry, and environment and even in the origin of life.[3]

In the recent years, due to the limited energy resources, the characterization of petroleum reservoirs has gained a lot of interest.[4] In this context especially a knowledge of transport properties of hydrocarbon mixtures such as linear alkanes and organic ring compounds is very important.[5] Different techniques exist to determine the thermal diffusive properties of liquid mixtures.[6] The thermogravitational columns have been validated in a benchmark test for three binary hydrocarbon mixtures.[7] With this technique, the value of the thermal diffusion coefficient of a binary at the stationary state is given by[8]

$$D_T = -\frac{ga^4}{504}\frac{\alpha}{w_0(1 - w_0)\beta\mu}\frac{\partial\rho}{\partial z} \qquad (1)$$

where g is the acceleration of gravity, α is the thermal expansion coefficient of the mixture, β is the concentration expansion coefficient of the mixture, μ is the dynamic viscosity of the mixture, a is the "gap" (distance between the inner cylinder and the exterior cylinder of the column where the fluid is of study), w_0 is the initial mass fraction of the densest component in the binary mixture, and $\partial\rho/\partial z$ is the vertical gradient of density ρ in the column at the stationary state. The parameters α, β, and μ are therefore thermophysical properties needed for the determination of the thermal diffusion coefficient.

The systems of the benchmark of Fontainebleau[7] are composed of binaries 1,2,3,4-tetrahydronaphthalene + dodecane, 1,2,3,4-tetrahydronaphthalene + 1-phenyl-2-methylpropane, and 1-phenyl-2-methylpropane + dodecane (mass fraction of 50 % and mean temperature of 25 °C). They represent binary mixtures for calibration of new setups of thermodiffusion or thermogravitation. Measurements of thermogravitation under high pressure are in progress,[9,10] but thermophysical properties and molecular and thermal diffusion coefficients are known for these systems only at atmospheric pressure. The aim of this work is to provide these properties at high pressure. In this work, we first present the measurements for the thermal expansion and concentration expansion coefficients and for the viscosity of the binaries of the benchmark of Fontainebleau for pressures varying from (0.1 to 20) MPa at 25 °C. The predicted coefficients from the Peng-Robinson, volume translated Peng-Robinson, and PC-SAFT equations of state are compared with the measured values. The Lohrenz-Bray-Clark model was used for the prediction of the viscosities.

Experimental Section

Materials. The 1,2,3,4-tetrahydronaphthalene (99 %), *n*-dodecane (99+ %), and 1-phenyl-2-methylpropane (99 %) were

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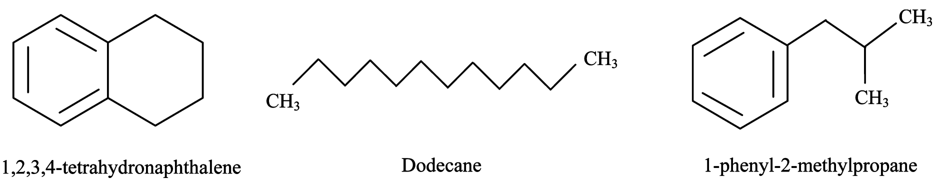

Figure 1. Molecular structures of the components.

Table 1. Pure Component Parameters for PC-SAFT, PR, and vt-PR Equations of State

	PC-SAFT			PR & vt-PR			
component	m_i	σ_i/Å	(ε_i/k_B)/K	T_C/K	P_C/MPa	ω_i	s_{vi}
1-phenyl-2-methylpropane	3.4711	3.9808	288.29	650.15	3.0398	0.3811	−0.0805
1,2,3,4-tetrahydronaphthalene	3.3627	3.8511	322.43	720.15	3.6200	0.3278	0.0146
dodecane	5.3839	3.8669	247.01	658.20	1.8239	0.5734	0.0985

Table 2. Theoretical and Experimental Densities for Binary Mixtures (50 % Mass Fraction and 25 °C)

	exptl	PR		vt-PR		PC-SAFT	
P/MPa	ρ/g·cm^{-3}	ρ/g·cm^{-3}	difference	ρ/g·cm^{-3}	difference	ρ/g·cm^{-3}	difference
1-phenyl-2-methylpropane + dodecane							
0.1	0.7917	0.7518	5.04 %	0.7669	3.13 %	0.7915	0.03 %
4	0.7945	0.7543	5.06 %	0.7695	3.15 %	0.7914	0.39 %
10	0.7986	0.7579	5.10 %	0.7733	3.17 %	0.7973	0.16 %
20	0.8051	0.7632	5.20 %	0.7787	3.28 %	0.8064	−0.16 %
1,2,3,4-tetrahydronaphthalene + dodecane							
0.1	0.8407	0.7745	7.87 %	0.8221	2.21 %	0.8465	−0.69 %
4	0.8434	0.7767	7.91 %	0.8246	2.23 %	0.8500	−0.78 %
10	0.8471	0.78	7.92 %	0.8282	2.23 %	0.8553	−0.97 %
20	0.8530	0.7847	8.01 %	0.8336	2.27 %	0.8636	−1.24 %
1,2,3,4-tetrahydronaphthalene + 1-phenyl-2-methylpropane							
0.1	0.9036	0.9016	0.22 %	0.8745	3.22 %	0.9007	0.32 %
4	0.9061	0.9039	0.24 %	0.8767	3.24 %	0.9043	0.20 %
10	0.9101	0.9073	0.31 %	0.8799	3.32 %	0.9097	0.04 %
20	0.9161	0.9122	0.43 %	0.8846	3.44 %	0.9182	−0.23 %

purchased from Sigma-Aldrich. The molecular structure of the three components is given in Figure 1.

Apparatus. Densities of the binary mixtures were measured with a DMA HPM Anton-Paar densimeter equipped with a mPDS-200V3 unit. The complete experimental assembly has been described in detail elsewhere.[11] The measurement principle is based on the determination of the oscillation period of a U-shaped tube which contains the sample. With this technique, density is linearly related to the square of the measured period of oscillation. As proposed by Lagourette et al.,[11] the two parameters involved are evaluated from the density of a single reference fluid (water) over the whole range of pressure and temperature with the additional knowledge of the temperature dependency of the experimental oscillation period for another reference fluid, vacuum. Taking into account the accuracy of the temperature, the pressure, the period of oscillation measurement for water, vacuum and the studied systems, and the water density accuracy, the overall experimental uncertainty in the reported density values is estimated to be ± 0.5 kg·m^{-3} (i.e., around 0.05 % for density close to water density). This uncertainty is similar to those that are reported in previous studies.[12,13]

A falling body viscometer (semi-automatic Stony Brook Scientific HPHTV-100 viscosimeter) was used to measure the viscosity of the compressed liquids. In this apparatus, a stainless steel cylinder falls through a fluid of unknown viscosity at given conditions of temperature and pressure. The viscosity is a function of the falling time of the density difference between the cylinder and the fluid and of one constant obtained by calibrating the viscometer with a substance of known viscosity and density. We have used toluene[14] as the calibrating fluid. Decane[15,16] has been used to verify the calibration. Each measurement of the falling time was repeated three times at thermal and mechanical equilibrium, and it is reproducible to better than 1 %. The final value is an average of these measurements. The total uncertainty of the obtained viscosity values was estimated to be within 2 %, which is comparable to that estimated by other authors for similar devices, as has been discussed in previous papers.[17,18] Viscosity values at (0.1, 4, and 10) MPa are given in Table 5. The viscosity at 20 MPa was not measured experimentally because the limitation of the viscosimeter is up to 14 MPa.

Methods. The thermal expansion coefficient is given by

$$\alpha = -\rho_0^{-1}(\partial\rho/\partial T)_{P,w} \qquad (2)$$

where ρ_0 is the density of the mixture at the temperature T_0 and $(\partial\rho/\partial T)$ is the derivative of the density with temperature at T_0 and at a mass fraction of 50 %. Figure 2 shows the densities for five temperatures centered on 25 °C and for pressures varying from (0.1 to 20) MPa for the mixture of 1-phenyl-2-methylpropane + dodecane.

The results were fitted by a linear function using the least-squares method. Some obtained values are given in Table 3. Considering the uncertainty of the measurements and the sensitivity of the method of least-squares adjustment, the uncertainty on α is estimated to be ± $(0.3 \cdot 10^{-4})$ K^{-1} for the three systems at all pressures. For 0.1 MPa, our results are in agreement with those published in the literature for the same systems at atmospheric pressure.[19]

Table 3. Theoretical and Experimental Thermal Expansion Coefficient for Binary Mixtures (50 % Mass Fraction and 25 °C)

	exptl	PR		vt-PR		PC-SAFT	
P/MPa	$\alpha/10^{-4}$ K	$\alpha/10^{-4}$ K	difference	$\alpha/10^{-4}$ K	difference	$\alpha/10^{-4}$ K	difference
1-phenyl-2-methylpropane + dodecane							
0.1	9.3	6.06	34.8 %	6.18	33.5 %	9.92	−6.7 %
4	8.9	5.73	35.7 %	5.94	33.3 %	9.68	−8.8 %
10	9.0	5.29	41.2 %	5.64	37.3 %	9.35	−3.9 %
20	8.6	4.70	45.4 %	5.24	39.1 %	8.88	−3.3 %
1,2,3,4-tetrahydronaphthalene + dodecane							
0.1	8.6	5.56	35.4 %	5.90	31.4 %	9.47	−10.1 %
4	8.3	5.28	36.3 %	5.60	32.5 %	9.28	−11.8 %
10	8.4	4.92	41.5 %	5.19	38.2 %	9.02	−7.4 %
20	8.0	4.42	44.7 %	4.62	42.3 %	8.64	−8.0 %
1,2,3,4-tetrahydronaphthalene + 1-phenyl-2-methylpropane							
0.1	8.8	5.76	34.6 %	5.59	36.5 %	9.35	−6.2 %
4	8.5	5.53	35.0 %	5.34	37.2 %	9.18	−8.0 %
10	8.5	5.21	38.7 %	5.01	41.1 %	8.93	−5.1 %
20	8.1	4.76	41.3 %	4.54	44.0 %	8.58	−6.0 %

Table 4. Theoretical and Experimental Concentration Expansion Coefficient for Binary Mixtures (50 % Mass Fraction and 25 °C)

	exptl	PR		vt-PR		PC-SAFT	
P/MPa	β	β	difference	β	difference	β	difference
1-phenyl-2-methylpropane + dodecane							
0.1	0.13	0.266	−104.6 %	0.104	20.0 %	0.130	0.1 %
4	0.13	0.265	−103.8 %	0.103	20.8 %	0.129	0.8 %
10	0.13	0.265	−103.8 %	0.101	22.3 %	0.128	1.5 %
20	0.12	0.264	−120.0 %	0.010	91.7 %	0.127	−5.8 %
1,2,3,4-tetrahydronaphthalene + dodecane							
0.1	0.27	0.335	−24.1 %	0.256	5.2 %	0.273	−1.1 %
4	0.27	0.334	−23.7 %	0.254	5.9 %	0.272	−0.7 %
10	0.26	0.332	−27.7 %	0.252	3.1 %	0.269	−3.5 %
20	0.26	0.329	−26.5 %	0.249	4.2 %	0.265	−1.9 %
1,2,3,4-tetrahydronaphthalene + 1-phenyl-2-methylpropane							
0.1	0.13	0.070	46.2 %	0.153	−17.7 %	0.144	−11.0 %
4	0.13	0.070	46.2 %	0.153	−17.7 %	0.143	−9.9 %
10	0.13	0.068	47.7 %	0.152	−16.9 %	0.141	−8.5 %
20	0.12	0.067	44.2 %	0.151	−25.8 %	0.138	−15.2 %

Table 5. Theoretical and Experimental Viscosities for Binary Mixtures (50 % Mass Fraction and 25 °C)

	exptl	PR		vt-PR		PC-SAFT	
P/MPa	μ/mPa·s	μ/mPa·s	difference	μ/mPa·s	difference	μ/mPa·s	difference
1-phenyl-2-methylpropane + dodecane							
0.1	1.09	0.764	29.9 %	0.902	17.2 %	1.098	−0.7 %
4	1.14	0.786	31.1 %	0.930	18.4 %	1.151	−1.0 %
10	1.22	0.818	33.0 %	0.970	20.5 %	1.234	−1.1 %
20	--	0.866	--	1.033	--	1.378	--
1,2,3,4-tetrahydronaphthalene + dodecane							
0.1	1.46	0.965	33.9 %	1.691	−15.8 %	2.079	−42.4 %
4	1.53	0.989	35.4 %	1.746	−14.1 %	2.178	−42.4 %
10	1.64	1.026	37.4 %	1.827	−11.4 %	2.333	−42.3 %
20	--	1.082	--	1.954	--	2.606	--
1,2,3,4-tetrahydronaphthalene + 1-phenyl-2-methylpropane							
0.1	1.33	1.540	−15.8 %	1.156	13.1 %	1.524	−14.6 %
4	1.37	1.580	−15.3 %	1.182	13.7 %	1.586	−15.8 %
10	1.43	1.638	−14.5 %	1.221	14.6 %	1.682	−17.6 %
20	--	1.731	--	1.282	--	1.849	--

The concentration expansion coefficient is given by

$$\beta = \rho_0^{-1}(\partial\rho/\partial w)_{P,T} \tag{3}$$

where w is the mass fraction of the densest component. The temperature is now fixed, and we measure the density on a small interval of concentrations centered at $w_0 = 0.5$. The components of the mixture are weighed on a digital balance with a resolution of 0.01 g. On a total mass of 45 g, the uncertainty on w is estimated to be $\pm\, 2\cdot10^{-4}$. The approach for the determination of β is the same as the one used for the measurement of the thermal expansion α. Figure 3 shows the densities for five mass fractions (of the densest component) for the mixture of 1-phenyl-2-methylpropane + dodecane, and they are presented at different pressures.

Some obtained values of β are given in Table 4. The uncertainty on β is estimated to be ± 0.009 for 1-phenyl-2-methylpropane + dodecane, ± 0.01 for 1,2,3,4-tetrahydronaphthalene + dodecane, and ± 0.008 for 1,2,3,4-tetrahydronaphthalene + 1-phenyl-2-methylpropane. For 0.1 MPa, our results are in agreement with those published in the literature for the same systems at atmospheric pressure.[19]

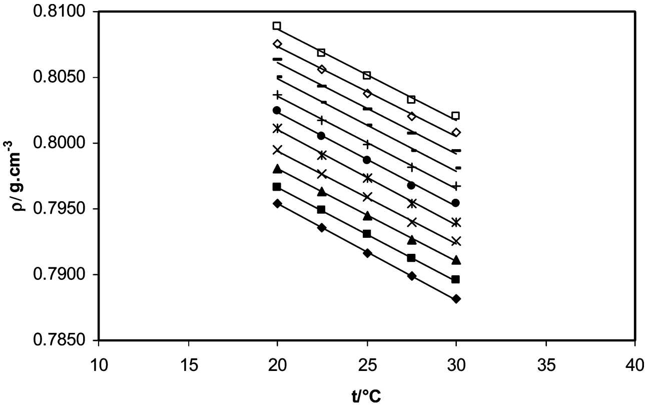

Figure 2. Density of the binary 1-phenyl-2-methylpropane + dodecane as a function of the temperature and for different pressures: ◆, 0.1 MPa; ■, 2 MPa; ▲, 4 MPa; ×, 6 MPa; *, 8 MPa; ●, 10 MPa; +, 12 MPa; -, 14 MPa; - -, 16 MPa; ◇, 18 MP; and □, 20 MPa.

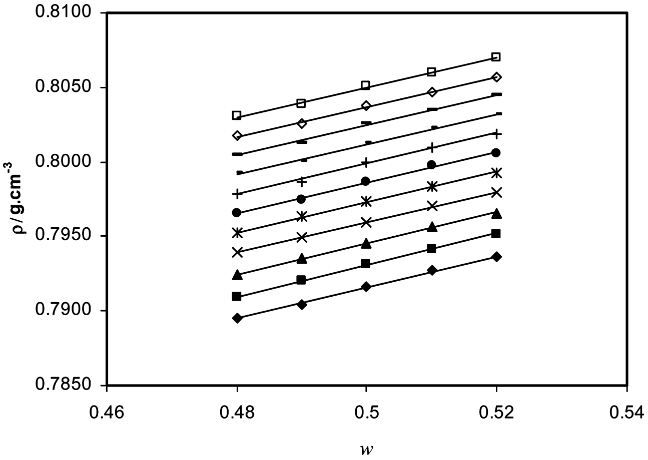

Figure 3. Density of the binary 1-phenyl-2-methylpropane + dodecane at 25 °C as a function of the mass fraction of the densest component and for different pressures: ◆, 0.1 MPa; ■, 2 MPa; ▲, 4 MPa; ×, 6 MPa; *, 8 MPa; ●, 10 MPa; +, 12 MPa; -, 14 MPa; - -, 16 MPa; ◇, 18 MPa; and □, 20 MPa.

Theory

For the evaluation of the density, the compressibility factor of the mixtures has been calculated with different equations of state.

The perturbed chain statistical associating fluid theory equation of state (PC-SAFT) was derived and described in detail by Gross and Sadowski.[20] The compressibility factor Z is the sum of the ideal gas contribution (id), the hard-chain term (hc), and the dispersive part (disp). The effects of multipole interactions and association are not taken into account in this study.

$$Z = 1 + Z^{\mathrm{hc}} + Z^{\mathrm{disp}} \tag{4}$$

The hard chain term was developed by Chapman et al.[21,22] and is based on Wertheim's first-order thermodynamic perturbation theory.[23,24] It contains two pure component parameters: the number of segments m and the diameter of segments σ. The dispersive contribution is calculated from the second-order perturbation theory suggested by Barker and Henderson[25,26] applied to chain molecules. It requires three pure-component parameters: m, σ, and ε/k, the depth of the pair potential (see Table 1 for their values here). The parameters for the mixture are obtained by conventional Berthelot-Lorentz combining rules.

The Peng-Robinson equation of state (PR-EoS) is one of the most popular cubic equations of state and is commonly used in the petroleum industry for hydrocarbon mixtures due to its simplicity and accuracy. The compressibility factor Z in PE-EoS can be written as follows[27]

$$Z = \frac{\bar{V}}{\bar{V} - b_{\mathrm{p}}} - \frac{a_{\mathrm{p}}(T)\bar{V}}{RT(\bar{V}(\bar{V} + b_{\mathrm{p}}) + b_{\mathrm{p}}(\bar{V} - b_{\mathrm{p}}))} \tag{5}$$

The pure component parameters a_{p} (attraction parameter) and b_{p} (covolume) can be calculated from critical properties.[27] The classical van der Waals mixing rules (with all interaction parameters set to zero) are used to compute the parameters of the hydrocarbon mixtures.

The volume translation proposed by Péneloux et al. was used in combination with the PR-EoS.[28] The shift parameters s_{vi} (Table 1) were calculated by the Jhaverl and Youngren[29] approach for hydrocarbons heavier than n-heptane.

The Lohrenz et al.[30] liquid viscosity of the fluid mixtures formulation is as follows

$$\mu_{\mathrm{f}} = \mu^* + \frac{(\zeta^4 - 10^{-4})}{\psi} \tag{6}$$

where

$$\mu^* = \frac{\sum_{i=1}^{n} x_i \mu_i \sqrt{M_i}}{x_i \sqrt{M_i}} \tag{7}$$

and

$$\zeta = 0.1023 + 0.023364\rho_{\mathrm{r}} + 0.05833\rho_{\mathrm{r}}^2 - 0.40758\rho_{\mathrm{r}}^3 + 0.0093324\rho_{\mathrm{r}}^4 \tag{8}$$

and

$$\psi = \frac{(\sum_{i=1}^{n} x_i T_{\mathrm{C}i})^{1/6}}{(\sum_{i=1}^{n} x_i M_i)^{1/2} (\sum_{i=1}^{n} x_i P_{\mathrm{C}i})^{2/3}} \tag{9}$$

μ_i is the viscosity, $T_{\mathrm{C}i}$ is the critical temperature, $P_{\mathrm{C}i}$ is the critical pressure, and M_i is the molecular weight of the ith component. ρ_{r} is the reduced density of the mixture.

Results and Discussion

The mixture densities have been compared between measured and calculated (PR, vt-PR, and PC-SAFT) densities for pressures varying from (0.1 to 20) MPa. Comparison is given in Table 2. As one may notice, the PC-SAFT equation of state proves to be more efficient for predicting the densities than the other equations of state. For the vt-PR-EoS, the disagreement with the experimental densities is the same for the three binaries. PR-EoS gives good results only for the 1,2,3,4-tetrahydronaphthalene + 1-phenyl-2-methylpropane system (mixture of aromatics), which is logical being given that cubic equations are not suitable for long alkanes unlike equations of the SAFT type that contain a term of chain. We can observe that the performances of the equations of state are independent of the pressure, presumably because the temperature is not very high.

The thermal expansion and concentration expansion coefficients have been calculated (PR, vt-PR, and PC-SAFT) from the densities, that calculated for temperatures centered on 25 °C and for mass fractions centered on 50 %, and with the same methodology that was used for experimental data. The comparisons between the measured and the calculated thermal expansion coefficients and the concentration expansion coefficients for different pressures [from (0.1 to 20) MPa] are given in Table 3 and Table 4, respectively.

The PC-SAFT equation of state gives a suitable result for the prediction of the two derivative properties α and β. PR and vt-PR equations of state are unsuitable for the prediction of the two derivative properties.

Using the Lohrenz-Bray-Clark model for the viscosity of mixtures (eq 6) and the calculated densities (PR, vt-PR, and PC-SAFT), it was possible to evaluate viscosities for each binary mixture and from each equation of state. The comparison between the measured and the calculated viscosities for pressures going from (0.1 to 20) MPa is given in Table 5.

The Lohrenz-Bray-Clark model combined with the PR or vt-PR equation of state is unsuitable for the prediction of the viscosity of the three binaries. When combined with the PC-SAFT equation of state for the density, the agreement with experimental data is good only for the 1-phenyl-2-methylpropane + dodecane system (see Table 5). Being given the good character for the prediction of densities with the PC-SAFT equation of state (Table 2), we can conclude that the Lohrenz-Bray-Clark model, as applied here, is not adapted for the prediction of viscosities of the studied binaries.

Conclusions

In thermogravitational experiments, the thermal expansion and concentration expansion coefficients and the viscosity of mixtures are necessary properties for the determination of the thermal diffusion coefficient. We have measured the densities of binaries of dodecane, 1-phenyl-2-methylpropane, and 1,2,3,4-tetrahydronaphthalene for pressures going from (0.1 to 20) MPa for temperatures centered on 25 °C and for concentrations centered on 50 %. By a derivative method, we have determined the thermal expansion and concentration expansion coefficients at 25 °C and 50 % mass fraction. Viscosities have been directly measured. To accurately predict the thermal and concentration expansion coefficients, an attempt was made first to calculate the densities of the binaries using PC-SAFT, Peng-Robinson, and volume translated Peng-Robinson equations of state. The comparisons with measured densities show that PC-SAFT has better agreement with experiments than the other equations of state. From calculated densities, we have evaluated the thermal expansion and concentration expansion coefficients. It is found that the PC-SAFT gives suitable results for the two derivative properties unlike the two other equations of state. The combination of the model of Lohrenz-Bray-Clark for the viscosity of liquid mixtures and the densities calculated with the three equations of state proved to be inefficient for the prediction of the viscosities of the binaries.

Supporting Information Available:

All the density data have been reported. This material is available free of charge via the Internet at http://pubs.acs.org.

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