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Density Measurements under Pressure for the Binary System 1-Propanol + Toluene

Claus K. Zéberg-Mikkelsen* and Simon I. Andersen

Center for Phase Equilibria and Separation Processes (IVCSEP), Department of Chemical Engineering, Technical University of Denmark, Building 229, 2800 Lyngby, Denmark

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The density of the binary system composed of 1-propanol and toluene has been measured under pressure using a vibrating-tube densimeter. The measurements have been performed for four different compositions as well as the pure compounds at four temperatures in the range of (303.15 to 333.15) K and seven isobars up to 30 MPa. The uncertainty of the reported densities is less than 0.05%. The measured data has been used to study the influence of temperature, pressure, and composition on the isothermal compressibility and the isobaric thermal expansivity as well as the excess molar volume, which shows a complex sigmoid behavior involving both positive and negative values. This complex behavior has been interpreted as the result of changes in the free volume due to volume expansion and compressibility as a result of the breaking of hydrogen bonds of the self-associating alcohol molecules but also due to the electron donor−acceptor-type formation of hydrogen bonding between the hydroxyl group of 1-propanol and the π electrons of toluene because aromatic hydrocarbons can act as electron donors.

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1. Introduction

The ability of aromatic compounds to act as electron donors with their π electrons has been investigated by several researchers.[1−4] Because of this ability, weak intermolecular interactions, such as hydrogen bonding, can be formed in mixtures composed of aromatic compounds and polar or associating compounds. In a recent investigation using FTIR spectroscopy, Brinkley and Gupta[4] found clear evidence for the formation of hydrogen bonding between aromatic hydrocarbons and alcohols. They studied qualitatively the degree of hydrogen bonding formation by adding aromatic hydrocarbons to alkane + alcohol mixtures having a very low content of alcohol in order to avoid any influence of self-association of the alcohol.

Alcohols are used in a wide range of industrial applications involving hydrocarbon or petroleum fluids. For example, they are used as cosurfactants in oil-in-water emulsions, as additives in gasoline to reduce pollution effect, as inhibitors to prevent the precipitation of gas hydrates in pipelines, etc. In these systems, complex behavior may be developed because the possibility exists for intermolecular interactions between the alcohols and the aromatic hydrocarbons, which are important constituents of petroleum fluids. To study and understand the behavior of these fluids under operating conditions, experimental property measurements of simplified mixtures can provide valuable information, both from a modeling as well as from a fundamental point of view. One of these properties is the density, which is required in many engineering disciplines as well as to obtain other thermophysical properties.

Several experimental studies of the density of alcohol + aromatic hydrocarbon mixtures have been performed at 0.1 MPa[5−10] as well as under pressure.[11−15] Most of the previous studies involve binary mixtures composed of methanol or ethanol with aromatic hydrocarbons, but to our knowledge, no experimental high-pressure studies exist for binary systems involving 1-propanol and aromatic hydrocarbons. Because of this, the aim of this work is to provide new experimental density data for binary mixtures composed of 1-propanol and toluene at elevated pressure. These data have further been used in a study of the influence of the composition, temperature, and pressure on the excess molar volume as well as on the isothermal compressibility and thermal expansion.

2. Experimental Techniques

The two compounds used in this study are commercially available chemicals with the following purity levels: 1-propanol from Merck with a chemical purity of >99.5% (gas−liquid chromatography) and molecular weight of $M_w = 60.096\ \mathrm{g{\cdot}mol^{-1}}$ and toluene from Rathburn Chemicals with a chemical purity of >99.9% (HPLC distilled grade) and $M_w = 92.141\ \mathrm{g{\cdot}mol^{-1}}$. Before use, 1-propanol was distilled at atmospheric pressure. The condensate (distillate) was collected in a bottle containing molecular sieves in order to remove any remaining water. For toluene, no further purification was performed. The binary mixtures of 1-propanol (1) + toluene (2) were prepared immediately before use by weighing at atmospheric pressure and ambient temperature using a Mettler balance with an uncertainty of 0.001 g. For each mixture, a sample containing 100 g was prepared, which, taking into account the uncertainty of the balance, resulted in an uncertainty in the mole fraction of less than 2×10^{-5}. Before the densimeter equipment was filled with the samples, they were degassed using a Branson 1510 ultrasonic bath in order to avoid any influence of dissolved gas on the measurements. The overall uncertainty in the composition is less than 0.1 mol%, when the uncertainty coming from the weighing of any of the two compounds and the degassing by ultrasound of the samples are taking into account.

The density was measured between (0.1 and 30) MPa in the temperature range of (303.15 to 333.15) K with an

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* Corresponding author. E-mail: czm@kt.dtu.dk; Phone: (-45) 45 25 28 00; Fax: (-45) 45 88 22 58.

10.1021/je049685z CCC: $30.25

Table 1. Experimental Densities ρ for 1-Propanol (1) + Toluene (2) Mixtures vs Temperature *T*, Pressure *P*, and Mole Fraction x_1

T/K	P/MPa	ρ/kg·m^{-3}					
		$x_1 = 0$	$x_1 = 0.200$	$x_1 = 0.400$	$x_1 = 0.600$	$x_1 = 0.800$	$x_1 = 1$
303.15	0.1	857.4	847.5	837.2	825.9	812.4	795.6
303.15	5	861.3	851.5	841.2	829.9	816.4	799.5
303.15	10	865.1	855.3	845.0	833.7	820.2	803.4
303.15	15	868.7	859.0	848.7	837.4	823.8	807.0
303.15	20	872.3	862.5	852.2	840.9	827.3	810.5
303.15	25	875.6	865.9	855.7	844.3	830.7	813.9
303.15	30	878.9	869.2	858.9	847.5	834.0	817.2
313.15	0.1	848.1	838.1	828.0	816.9	803.8	787.6
313.15	5	852.1	842.2	832.0	821.0	807.8	791.6
313.15	10	856.2	846.3	836.1	825.0	811.9	795.7
313.15	15	860.0	850.2	840.0	828.9	815.8	799.5
313.15	20	863.7	853.9	843.7	832.6	819.4	803.2
313.15	25	867.2	857.5	847.3	836.1	823.0	806.7
313.15	30	870.7	860.9	850.7	839.5	826.3	810.1
323.15	0.1	838.6	828.5	818.3	807.4	794.7	779.1
323.15	5	842.9	832.9	822.7	811.8	799.1	783.4
323.15	10	847.2	837.2	827.0	816.1	803.4	787.7
323.15	15	851.2	841.3	831.1	820.2	807.4	791.7
323.15	20	855.1	845.2	835.0	824.1	811.3	795.6
323.15	25	858.9	849.0	838.8	827.9	815.0	799.3
323.15	30	862.5	852.6	842.4	831.5	818.6	802.9
333.15	0.1	829.2	818.7	808.5	797.9	785.6	770.6
333.15	5	833.7	823.3	813.1	802.5	790.2	775.1
333.15	10	838.2	827.9	817.8	807.1	794.7	779.6
333.15	15	842.5	832.3	822.1	811.4	799.0	783.9
333.15	20	846.6	836.4	826.3	815.6	803.1	787.9
333.15	25	850.5	840.4	830.3	819.5	807.0	791.8
333.15	30	854.3	844.2	834.1	823.3	810.8	795.5

Anton-Paar DMA 512P resonance densimeter combined with an mPDS 1000 evaluator unit. The calibration of the densimeter was performed using water and vacuum as described by Lagourette et al.[16] The pressurization of the fluid was performed by an ISCO syringe pump model 260D. However, to reduce the amount of sample required, a high-pressure cell (max pressure 35 MPa) was placed between the densimeter and the pressurizing pump. The uncertainty in the pressure was reported to be 0.5% of the real pressure, except at 0.1 MPa, whereas the uncertainty in the temperature was estimated to be ±0.05 K. The resultant overall uncertainty in the reported density values is estimated to be less than 0.05%.

3. Results and Discussion

Measurements of the density ρ were performed at four temperatures (303.15, 313.15, 323.15, and 333.15 K) and at seven pressures (0.1, 5, 10, 15, 20, 25 and 30 MPa) for 1-propanol (1) and toluene (2) as well as for four binary mixtures containing 1-propanol mole fractions of 0.200, 0.400, 0.600, and 0.800. For each mixture, 28 measured values were determined. The measured densities are given in Table 1 as a function of temperature *T*, pressure *P*, and mole fraction of 1-propanol x_1.

A comparison of the measured density values of the pure compounds with the values obtained by the correlations of Cibulka and Zikowá[17] and Cibulka and Takagi[18] has been performed. The Cibulka correlations[17,18] have been derived for each pure compound using experimental liquid density values covering wide ranges of temperature and pressure. This correlation method is based on the Tait-type relationship. The required reference density values, $\rho(T, P_{\mathrm{ref}}(T))$, in the Cibulka correlation has been determined for toluene from the expression given by Cibulka and Takagi,[18] whereas for 1-propanol an interpolation of the recommended density values given in the TRC tables[19] at 0.1 MPa was performed to obtain the necessary $\rho(T, P_{\mathrm{ref}}(T))$ values. Any other required compound properties in the Cibulka correlations have been taken from Poling et al.[20]

The obtained deviations between the experimental data in this work and the calculated values[17,18] are shown in Figures 1 and 2 versus pressure for 1-propanol and toluene, respectively. The comparison for toluene reveals excellent agreement between the experimental data in this work and the calculated values[18] resulting in an average absolute deviation (AAD) of 0.01%. For 1-propanol, the agreement between the experimental data and the calculated values[17] is also good, despite the fact that a systematic deviation going from positive to negative values is found; see Figure 1. The obtained AAD is 0.01%.

Nikam et al.[9] have measured the density of 1-propanol + toluene mixtures at 0.1 MPa in the temperature range of (303.15 to 313.15) K. Isothermal modeling of the data of Nikam et al.[9] has been performed to obtain density values corresponding to the compositions studied in this work. The

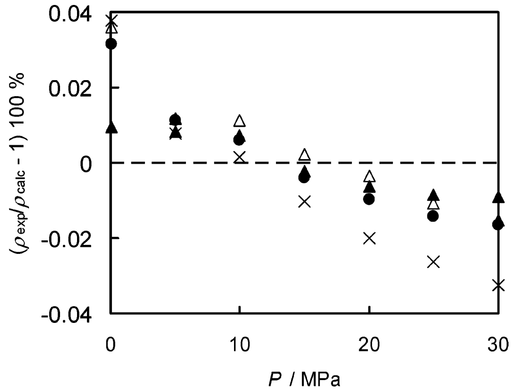

Figure 1. Deviations, $(\rho_{\mathrm{exp}}/\rho_{\mathrm{calc}} - 1)100\%$, between experimental densities ρ_{exp} for 1-propanol and calculated values ρ_{calc} from the correlation of Cibulka and Zikowá[17] vs pressure *P* at 303.15 K (▲), 313.15 K (△), 323.15 K (●), and 333.15 K (×).

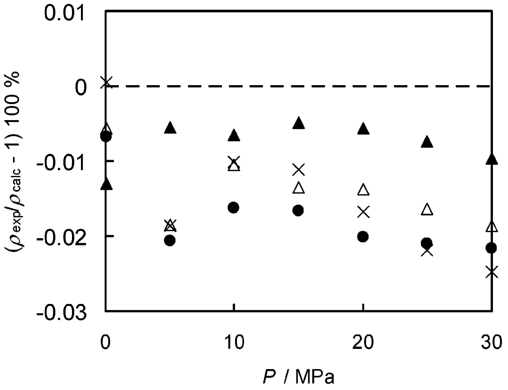

Figure 2. Deviations, $(\rho_{exp}/\rho_{calc} - 1)100\%$, between experimental densities ρ_{exp} for toluene and calculated values ρ_{calc} by the correlation of Cibulka and Takagi[18] vs pressure P at 303.15 K (▲), 313.15 K (△), 323.15 K (●), and 333.15 K (×).

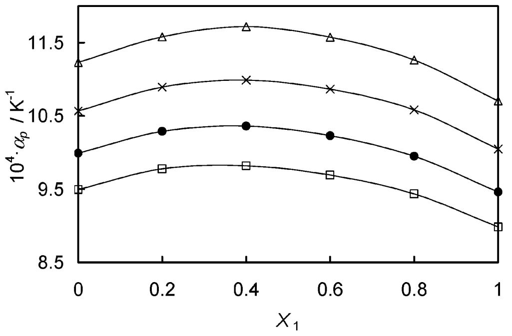

Figure 3. Isobaric thermal expansivity α_P vs the mole fraction of 1-propanol x_1 at 323.15 K under 0.1 MPa (Δ), 10 MPa (×), 20 MPa (●), and 30 MPa (□).

comparison between the densities of this work and those derived from the reported values of Nikam et al.[9] resulted in an AAD of 0.05% with the highest deviation (0.13%) found for toluene at 313.15 K.

The density increases with increasing pressure and decreasing temperature for all mixtures, and the density behavior varies monotonically with composition; see Table 1. An examination of the density values reveals a general pattern consistent with observations found for different liquid systems.[21−26] For a given mixture, the variation of the density with pressure at constant temperature is concave with $(\partial\rho/\partial P)_T$ but is associated with a negative second-order derivative. In addition, no curvature of the density at constant pressure can be detected over the small temperature range for which the experimental measurements were performed.

By numerical analysis of the experimental densities, other quantities can be derived, such as the isothermal compressibility defined as

$$\kappa_T = \frac{1}{\rho}\left(\frac{\partial\rho}{\partial P}\right)_T \tag{1}$$

or the isobaric thermal expansivity defined as

$$\alpha_P = -\frac{1}{\rho}\left(\frac{\partial\rho}{\partial T}\right)_P \tag{2}$$

These two quantities have been derived for each mixture and are reported in Tables 2 and 3 (given as Supporting Information). The isothermal compressibility has been derived by applying the following Tait-type equation

$$\rho(T, P)/\text{kg}\cdot\text{m}^{-3} = \frac{\rho(T/\text{K}, 0.1\ \text{MPa})/\text{kg}\cdot\text{m}^{-3}}{1 - C\ln\left(\frac{B/\text{MPa} + P/\text{MPa}}{B/\text{MPa} + 0.1\ \text{MPa}}\right)} \tag{3}$$

where B an C are adjustable parameters. These parameters were determined by fitting the isothermal density data of each mixture. The experimental densities of each isotherm were represented within an AAD of 0.003%. The uncertainty for the isothermal compressibility is estimated to be 1.0%. Because the isobaric densities within the considered temperature range decrease linearly with the temperature, the slope of each isobar is equal to the derivative $(\partial\rho/\partial T)_P$ in eq 2. The uncertainty of the isobaric thermal expansivity is estimated to be 0.5%.

For all compositions, the isothermal compressibility as well as the isobaric thermal expansion increases with increasing temperature and decreases with increasing pressure; see Tables 2 and 3. This is the result of changes in the free volume of the liquid and may also be related to the Brownian motion of the molecules because when a fluid is pressurized the molecular distance decreases, resulting in a less compressible and less expansive fluid. With increasing temperature, the Brownian motion of the molecules increases because of a larger free volume and distance, resulting in a more expansive fluid as well as in a decrease in the density of the fluid. Within the considered T, P conditions, it can be seen that 1-propanol is more compressible and less expansive with increasing temperature and pressure than toluene. The reason may be due to the self-association of alcohol molecules but may also be due to the shape and packing of the molecules. In 1-propanol, the self-association results in the formation of multimers, which make the fluid less expansive because of hydrogen bonding but more compressible because of a more loose packing of the molecules. The isothermal compressibility increases with the composition of 1-propanol, but as the pressure is increased, the isothermal compressibility of the mixtures becomes more similar. In the case of the isobaric thermal expansivity, the variation with the composition of 1-propanol is nonmonotonical with a maximum found at a mole fraction of 1-propanol of $x_1 = 0.400$; see Figure 3. The thermal expansivity of the mixture containing a mole fraction of 1-propanol of $x_1 = 0.800$ is similar to that of toluene. The higher thermal expansivity of the mixtures is a result of the disruption of the ordered molecular structure within the liquid and the breaking of hydrogen bonds within the formed 1-propanol multimers because the volume of the multimers is generally smaller than that of the sum of the single molecules.

4. Excess Molar Volume

The experimental densities have also been used to calculate the excess molar volume of the mixtures. The excess molar volume at each T, P condition was calculated by the following relation, which is commonly used to estimate excess molar volumes of binary mixtures[6−10,24,27]

$$v^{\text{E}}_{\text{mix}}/\text{cm}^3\cdot\text{mol}^{-1} = \frac{(x_1M_{\text{w},1}/\text{g}\cdot\text{mol}^{-1} + x_2M_{\text{w},2}/\text{g}\cdot\text{mol}^{-1}}{\rho_{\text{mix}}/\text{g}\cdot\text{cm}^{-3}} - \left(\frac{x_1M_{\text{w},1}/\text{g}\cdot\text{mol}^{-1}}{\rho_1/\text{g}\cdot\text{cm}^{-3}} + \frac{x_2M_{\text{w},2}/\text{g}\cdot\text{mol}^{-1}}{\rho_2/\text{g}\cdot\text{cm}^{-3}}\right) \tag{4}$$

where x is the mole fraction and M_w is the molecular weight. The calculated excess molar volumes are reported in Table 4 (Supporting Information). As it can be seen, the excess volume for the binary 1-propanol + toluene system is small compared with excess molar volumes reported for

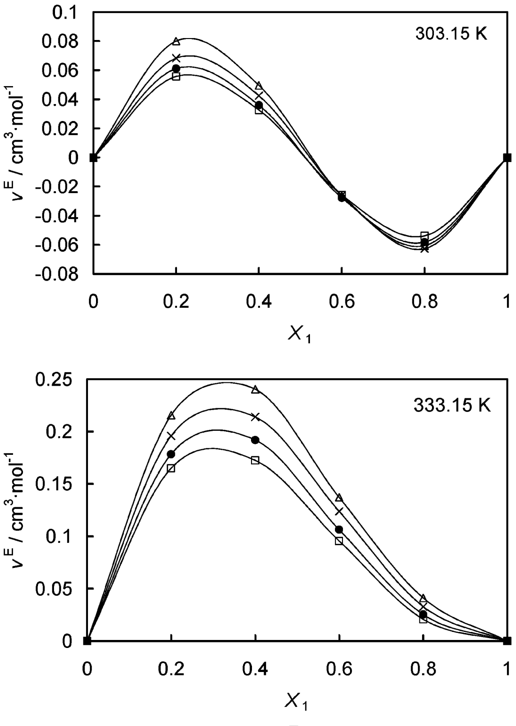

Figure 4. Excess molar volume V^E versus the mole fraction of 1-propanol x_1 at 303.15 K and 333.15 K under 0.1 MPa (△), 10 MPa (×), 20 MPa (●), and 30 MPa (□).

the asymmetrical binary system dimethyl carbonate + *n*-octane[27] or the associative system water + methanol.[28] However, a similar order of magnitude for the excess molar volume has been reported for other alcohol + aromatic hydrocarbon systems.[8,9,29−31]

For all mixtures, the excess molar volume increases with increasing temperature, and the largest numerical values of the excess molar volume are found at 0.1 MPa. As the pressure is increased, the numerical value of the excess molar volume decreases. This results in a crossing of the excess molar volume isobars when the excess molar volume of a mixture goes from negative to positive values with increasing temperature, as it can be seen from the reported values for the mixture containing 80 mol % 1-propanol. The behavior of the excess molar volume versus composition is shown in Figure 4 for various isobars at 303.15 K and 333.15 K. The plot at 303.15 K shows a sigmoid behavior of the excess molar volume with composition for this binary system. Similar behavior has been found for several alcohol + toluene systems.[9,29−31] As the temperature increases, this behavior disappears. In the toluene-rich region for all T, P conditions considered in this work, the excess molar volume is positive whereas in the 1-propanol-rich region, the excess molar volume is negative, but as the temperature increases, the excess molar volume becomes positive; see Figure 4.

Although the thermal expansivity of the mixtures is higher than for 1-propanol, indicating that a volume expansion in the mixtures occurs because of the breaking of hydrogen bonds in 1-propanol, the negative excess molar volumes found in the 1-propanol-rich region may be qualitatively explained as the result of the formation of weak hydrogen bonding interactions between 1-propanol and toluene due to electron donor−acceptor-type interactions between the hydroxyl group of the alcohol and the π electrons of the aromatic hydrocarbon ring structure. In the toluene-rich region, the positive values are the result of volume expansion due to the breaking of the hydrogen bonds within the self-associating alcohol molecules. Similar discussions and explanations of the sigmoid behavior of the excess molar volume with composition for this binary system as well as other binary alcohol + toluene systems can be found in the literature.[8,9,29−31]

The explanation for the increase in the excess volume with increasing temperature is due to the increase in the thermal expansivity as a result of the enlargement of the free volume and also to Brownian motion, which will result in the weakening or breaking of hydrogen bonds in alcohol multimers as well as the interactions between alcohol and toluene molecules, leading to a larger volume of the mixture. Furthermore, the physical forces start to prevail over the chemical interactions (hydrogen bonding), which are short-range forces. The decrease in the numerical value of the excess volume with increasing pressure (Figure 4) is due to the fact that when a fluid is pressurized its compressibility decreases with increasing pressure because of a reduction in the free volume within the liquid as a result of a decrease in the molecular distance.

A comparison of the excess molar volumes estimated in this work with those reported by Nikam et al.[9] at 0.1 MPa for T = 303.15 K and T = 313.15 K reveals that although the same trend in the behavior is found for the excess molar volume versus composition, differences between the calculated excess values of this work and those reported by Nikam et al.[9] are found. However, the agreement can be considered satisfactory by taking into account the experimental uncertainty of the density values.

5. Conclusions

The density of toluene and 1-propanol as well as four of their binary mixtures has been measured in the temperature range of (303.15 to 333.15) K and up to 30 MPa with an uncertainty of <0.05%. These data have been used to calculate the excess molar volume and to derive the isothermal compressibility and the isobaric thermal expansivity. As a result of the breaking of hydrogen bonds within the self-associating 1-propanol, the thermal expansivity of the mixtures is higher than the values of the two pure compounds, indicating a larger expansion in the mixture volumes. Despite this, plots of the excess molar volume as a function of composition, temperature, or pressure reveal a complex sigmoid behavior involving both positive and negative values. This has been interpreted as a result of the changes in the free volume due to volume expansivity and compressibility but also due to the possibility that toluene has the ability to act as an electron donor with its π electrons (negative excess volume).

Supporting Information Available:

Isothermal compressibility and isobaric thermal expansivity values for each mixture. Calculated excess molar volumes. This material is available free of charge via the Internet at http://pubs.acs.org.

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Received for review September 2, 2004. Accepted November 3, 2004. Financial funding for C.K.Z.-M. is provided by a Talent project from the Danish Technical Research Council (STVF) (contract no. 26-03-0063).

JE049685Z