

(p, ρ, T, x) and viscosity measurements of $\{x_1 n\text{-heptane} + (1 - x_1) n\text{-octane}\}$ mixtures at high temperatures and high pressures

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Abstract

Density and viscosity measurements are reported for $\{x_1 n\text{-heptane} + (1 - x_1) n\text{-octane}\}$ mixtures at $x_1 = (0.2808, 0.5427, 0.7906)$. The measurements of density and viscosity were made with a constant-volume piezometer immersed in a precision liquid thermostat and a capillary flow technique, respectively. Measurements were made at pressures up to 10 MPa. The range of temperature was (293 to 557) K for the density measurements and (298 to 473) K for the viscosity measurements. The total uncertainty of density, viscosity, pressure, temperature, and composition measurements was estimated to be less than 0.06%, 1.6%, 0.05%, 15 mK, and 0.02%, respectively. The effect of temperature, pressure, and concentration on density and viscosity of the binary $\{x_1 n\text{-heptane} + (1 - x_1) n\text{-octane}\}$ mixtures was studied. The measured values of density and viscosity for the pure components and mixtures were compared with those generated by reference equations and prediction techniques for the mixtures. The excess molar volumes V_m^E and the viscosity deviations $\Delta\eta$ were derived using the measured values of density and viscosity for the mixtures and pure components. The viscosity data have been interpreted in terms of the Grunberg–Nissan equation for binary mixtures. The temperature dependence of the Grunberg–Nissan constant was studied using the present viscosity data. The molar excess Gibbs energy of activation for flow ΔE_a was also calculated from our experimental viscosity data for the ($n\text{-heptane} + n\text{-octane}$) mixtures.

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1. Introduction

Transport properties of n -alkane mixtures are required in many industrial applications such as design calculations, heat- and mass-transfer rates, and fluid flow in petrochemical industrial equipment. Knowledge of the thermodynamic (p, ρ, T, x) and transport (viscosity) properties of

n -alkane mixtures is of fundamental importance for the modelling of underground oil flow and in other applications of chemical engineering to develop oil processes, process equipment in the oil and gas industries and oil recovery technology, all of which occur at high temperatures and high pressures. Underground oil comprises the largest reservoirs of n -alkane mixtures currently known. The thermodynamic properties of n -alkane mixtures are also of fundamental importance in geology and mineralogy for geothermal processes, which are responsible for the formation of the oil reservoirs. Thus, there is great practical interest in the thermodynamic and transport properties of n -alkane mixtures at high temperatures and high pressures.

The thermodynamic and transport properties of n -alkane mixtures are of research interest because of the

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need for accurate models to predict density and viscosity values [1–9] and to understand and control these processes that involve *n*-alkane mixtures. Accurate density and viscosity data over a wide range of temperature and pressure are important when modelling the mixture properties. To test and improve the accuracy of the prediction techniques and to develop effective prediction methods, reliable viscosity data for the mixtures and their pure components are required. However, measurements of the density and viscosity of *n*-alkane mixtures have so far been limited to a rather narrow range of temperature, pressure, and concentration with less than satisfactory accuracy. Previous density and viscosity measurements by various authors [10–13] for (*n*-heptane + *n*-octane) mixtures were performed only at atmospheric pressure and temperatures from (283 to 313) K. A survey of the literature reveals the lack of experimental density and viscosity data for (*n*-heptane + *n*-octane) mixtures at high temperatures and high pressures. Recently, the density and viscosity of these mixtures have been measured by Matos *et al.* [12] in the temperature range from (283 to 313) K and at atmospheric pressure over the whole composition range. Measurements of the density and viscosity were performed with the vibrating-tube densimeter and Ubbelohde-type viscometer with capillaries of different diameters, respectively. The uncertainties in density and viscosity measurements were $2 \cdot 10^{-5} \text{ g} \cdot \text{cm}^{-3}$ and 0.2%, respectively. The same techniques were used by Aucejo *et al.* [11] and Chevalier *et al.* [13] to measure the densities and viscosities of (*n*-heptane + *n*-octane) mixtures at $T = 298.15 \text{ K}$ and 0.1 MPa. Cooper and Asfour [10] reported densities and viscosities for (*n*-heptane + *n*-octane) mixtures at $T = 293.15 \text{ K}$ and 0.1 MPa. They also used the same techniques to measure the densities and viscosities. The experimental data were correlated by using a polynomial type equation. Thus, one of the primary objectives of this work was to expand the existing volumetric (p, ρ, T, x) and transport (viscosity) database for this mixture. The present density and viscosity results expand considerably the temperature and pressure ranges over which densities and viscosities for the $\{x_1 n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures are available.

2. Experimental

2.1. Density measurements

The (p, ρ, T, x) relationship of the $\{x_1 n\text{-heptane} + (1 - x_1) n\text{-octane}\}$ mixtures was measured by a constant-volume method. The apparatus and experimental procedure, described previously by Abdulagatov and Azizov [14–21], Azizov and Akhundov [22–24], and Azizov *et al.* [25], was used without modification. Only a brief discussion will be given here. The main part of the apparatus consisted of a piezometer, separating the U-shaped capillary tube with mercury, a liquid thermostat, heaters, a temperature regulator, and a 10 Ω platinum resistance thermometer (PRT-10). The volume of the piezometer at

$T = 293 \text{ K}$ and atmospheric pressure was $V_{293} = (95.545 \pm 0.02) \text{ cm}^3$. The value of V_{293} was previously calibrated from the known density of a standard fluid (pure water) with well-known (uncertainty is 0.0001% at 0.1 MPa in the liquid phase) (p, V, T) values (IAPWS standard, Wagner and Prüss [26]). The piezometer was positioned vertically in the liquid thermostat with volume of 0.02 m^3 . Three heaters were used to regulate the thermostat temperature that was controlled automatically to within 0.02 K by means of the high-precision temperature regulator. The temperature of the thermostat liquid was measured with PRT-10 (ITS-90 temperature scale). The sample temperature was detected with a precision of $\pm 15 \text{ mK}$. The pressure of the mixture was measured with a dead-weight pressure gauge (MP-600) with an uncertainty of 0.015% (maximum uncertainty was 0.05%).

The density of the sample at a given temperature T and pressure p is calculated from the simple relation

$$\rho_i = (m_{\text{tot}} - m_{\text{coll}}) / V_{PT}, \quad (1)$$

where m_{coll} is the mass of the sample extracted from the piezometer and collected during the runs, $i = 1, N$ (N is the number of extractions), m_{tot} is the total mass before extractions, and $V_{PT} = V_{293} + \Delta V_T + \Delta V_P$ is the temperature- and pressure-dependent volume of the piezometer. The temperature dependence of the piezometer volume at fixed pressure was calculated as

$$\Delta V_T = V_{T_0} [1 + 3\alpha(T - T_0)], \quad (2)$$

where V_{T_0} is the volume of piezometer at the initial reference temperature T_0 and $\alpha = 1.3 \cdot 10^{-5} \text{ K}^{-1}$ is the thermal expansion coefficient of the piezometer material. The pressure dependence of the piezometer volume ΔV_P was calculated from the Lave formula (Keyes and Smith [27]) for the cylinder. The uncertainty in piezometer volume V_{PT} at the given temperature and pressure is less than 0.038%. The uncertainty of the mass m of mixture can be estimated to be 0.006%. The experimental uncertainty in the concentration is estimated to be 0.02%. Based on a detailed analysis of all sources of uncertainties likely to affect the determination of density with the present apparatus, the combined expanded ($k = 2$) uncertainty of measuring the density was 0.06%.

To test the apparatus and procedures, the densities of the pure components *n*-heptane and *n*-octane were measured at temperatures from (301 to 516) K and at pressures up to 12 MPa. The results are given in table 1 together with values calculated from the Lemmon and Span [28] reference equation of state (EOS). The agreement between the reference EOS calculations (Lemmon and Span [28]) and the present density results for *n*-heptane and *n*-octane are within 0.038% and 0.022%, respectively (the deviation statistics are given in table 2). This excellent agreement confirms the reliability and accuracy of the present measurements for (*n*-heptane + *n*-octane) mixtures and corrects the operation of the (p, ρ, T) apparatus.

TABLE 1

Density measurements of *n*-heptane and *n*-octane and the comparison with densities from the reference Equation of State by Span and Lemmon [28]

	ρ (kg · m ⁻³)	ρ (kg · m ⁻³)	$\delta\rho^a$ (%)
	This work	[28]	
<i>T</i> /K	Atmospheric pressure		
	<i>n</i> -Octane		
301.84	695.24	695.30	−0.009
316.49	683.55	683.42	+0.019
329.76	672.69	672.52	+0.025
340.94	663.24	663.22	+0.003
358.76	648.10	648.09	+0.001
	<i>n</i> -Heptane		
302.46	675.91	675.95	−0.006
314.57	665.62	665.60	+0.003
329.71	652.43	652.44	−0.002
342.94	640.61	640.69	−0.013
353.76	630.86	630.80	+0.009
<i>P</i> /MPa	Under pressure		
	<i>n</i> -Octane		
	<i>T</i> = 311.89 K		
4.6800	691.6	691.7	−0.017
10.642	697.4	697.2	+0.029
	<i>T</i> = 367.61 K		
5.8730	648.8	648.8	+0.002
11.461	656.2	656.0	+0.030
	<i>T</i> = 437.97 K		
6.4300	589.6	589.7	+0.020
11.084	599.5	599.6	+0.017
	<i>T</i> = 516.64 K		
5.712	505.4	505.7	−0.053
9.791	525.2	524.9	+0.057
	<i>n</i> -Heptane		
	<i>T</i> = 307.61 K		
4.7600	676.1	676.34	−0.035
11.941	683.2	683.10	+0.015
	<i>T</i> = 387.00 K		
5.7830	610.0	609.9	+0.016
12.460	620.7	620.6	+0.016
	<i>T</i> = 450.12 K		
4.3500	543.7	543.45	+0.046
11.492	564.4	564.20	+0.035
	<i>T</i> = 500.47 K		
6.791	492.7	493.7	−0.195
10.72	512.8	512.3	+0.098

^a $\delta\rho = 100(1 - \rho[28]/\rho^{\text{exp}})$.

2.2. Viscosity measurements

The apparatus and procedures used for the viscosity measurements of the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures has been described in detail in previous papers by Abdulagatov and Azizov [29–34], Abdulagatov *et al.* [35–39], Azizov and Akhundov [40] and were used without modification. Only brief and essential information will be given here. The measurements were made using a capillary-flow method that gives an uncertainty of 1.6% for the viscosity. The main parts of the apparatus consisted of a working capillary with an extension tube, a high temperature and high-pressure autoclave, movable and stationary cylinders, electrical heaters, and a solid red copper block. The capillary together with the extension tube was located in the high-temperature and high-pressure autoclave. The autoclave was placed in the solid red copper block. Two electrical heaters were wound around the surface of the copper block. To generate and measure the pressure, the autoclave was connected with a dead-weight pressure gauge (MP-600) by means of separating vessel. The maximum uncertainty in pressure measurements was 0.05%.

The final working equations for this method are (Abdulagatov and Azizov [29])

$$\eta = U\tau \frac{\rho}{\rho_c} \left(1 - \frac{\rho_c}{\rho_{\text{Hg}}}\right) (1 + \alpha\Delta t)^3 - W \frac{\rho_c}{\tau},$$

$$U = \frac{g\pi R^4 \Delta H_{0,\text{Hg}}}{8LV_c} \text{ and } W = \frac{mV_c}{8\pi L}, \quad (3)$$

where $R = (0.15091 \pm 0.005)$ mm is the inner radius of the capillary, $L = (540.324 \pm 0.005)$ mm is the capillary tube length, τ is the time of flow, α is the linear expansion coefficient of the capillary material, Δt is the difference between experimental temperature and room temperature, $m = 1.12$ (kinetic-energy correction factor [41]) is a constant, $\rho(p, T)$ is the density of the fluid under study at the experimental conditions (p, T), $V_c = 1.2182 \text{ cm}^3$ is the volume of the measuring cylinder, ρ_c is the density of the fluid under study at room temperature and experimental pressure, $\Delta H_0 = (H_1 - H_2)/\ln(H_1/H_2)$, where H_1 and H_2 are the mercury levels at the beginning and ending of the fluid flowing, respectively, at room temperature and atmospheric pressure, ρ_{Hg} is the density of mercury at room temperature and experimental pressure, $\rho_{0,\text{Hg}}$ is the density of mercury at room temperature and atmospheric pressure. Equation

TABLE 2

Deviation statistics between the present measurements of density and calculated with reference EOS by Lemmon and Span [28] for pure components and prediction technique for mixtures by Lemmon and Jacobsen [43]

Fluids	AAD/(%)	Bias/(%)	St. dev./(%)	St. error/(%)	Max. dev./(%)	<i>N</i>
<i>n</i> -Heptane [28]	0.038	−0.001	0.067	0.018	0.098	13
<i>n</i> -Octane [28]	0.022	0.003	0.080	0.029	0.057	13
<i>n</i> -Octane + <i>n</i> -heptane [43]	0.226	0.223	0.080	0.010	0.340	55

(3) was derived from Poiseuille's law with corrections for the temperature effects on the capillary sizes, for mercury and sample densities at the experimental conditions (p, T), and for entrance effects (acceleration of a fluid at the inlet and outlet) on the fluid. The values of the parameters U and W (cell constants) can be also determined by means of a calibration technique. The time of fluid flowing

through the capillary τ was measured with a stopwatch with an uncertainty of less than 0.1 s (0.5%). An electromagnetic device was used to start and stop the watch.

The viscosity was obtained from the measured quantities R^4 , ΔH_0 , L , V_c , τ , ρ_{Hg} , ρ_c , T , P , and m . The accuracy of the viscosity measurements was assessed by analyzing the sensitivity of equation (3) to the experimental uncertainties of the measured quantities [29]. At the maximum measured temperature, the value of the root-mean-square deviations in the viscosity measurements was $\delta\eta = 0.004 \text{ mPa} \cdot \text{s}^{-1}$ ($2 \cdot 10^{-5} \text{ g} \cdot \text{cm}^{-1} \cdot \text{s}^{-1}$). Based on the detailed analysis of all sources of uncertainties likely to affect the determination of viscosity with the present apparatus, the combined expanded ($k = 2$) uncertainty in measuring the viscosity was 1.6% (see Abdulagatov and Azizov [29]). The Reynolds (Re) number occurring during all measurements was less than the critical values ($Re_c = 300$). As one can see from equation (3), to calculate the dynamic viscosity from measured quantities, the values of density of the mixture under study at room temperature and experimental pressure ρ_c , and density at the experimental conditions $\rho(p, T)$ are needed. For this purpose we used the present measurements.

The viscosity of pure components, n -heptane and n -octane, has also been measured at temperatures from (306 to 473) K and at pressures up to 10 MPa in order to verify the accuracy of the method and to correct operation of the present instrument. The measured values of the viscosity for n -heptane and n -octane are given in table 3 together with values calculated using the reference equation by Huber *et al.* [42]. The agreement between these calculated values and the our results for n -heptane and n -octane lie within 0.65% and 0.59%, respectively (the deviation statistics are given in table 4). This good agreement between our data and the values calculated from the reference equation by Huber *et al.* [42] confirms the reliability and high accuracy of the measurements for $\{x_1 n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures and the correct operation of the present instrument.

The samples pure n -heptane and n -octane used in this study were supplied by Novocherkask Chemical Company (Russia) with labeled mole fraction purities of 0.9995 and were used as received without further purification. The purity of the pure n -heptane and pure n -octane was confirmed by g.l.c. analysis. Density of the pure n -heptane (679.42 ± 0.02) $\text{kg} \cdot \text{m}^{-3}$ and pure n -octane (698.51 ± 0.02) $\text{kg} \cdot \text{m}^{-3}$ were also measured at $T = 298.15 \text{ K}$ and at 0.1 MPa with an uncertainty of $2 \cdot 10^{-5} \text{ g} \cdot \text{cm}^{-3}$ and

TABLE 3

Viscosity measurements of n -heptane and n -octane and the comparison with viscosity calculated via the reference equation of Huber *et al.* [42]

T/K	This work	Huber <i>et al.</i> [42]	$\delta\eta^a$ (%)
<i>n</i> -Heptane			
$P = 5 \text{ MPa}$, $\eta/\text{mPa} \cdot \text{s}$			
306.79	0.3779	0.3763	+0.43
337.54	0.2800	0.2786	+0.51
369.35	0.2116	0.2131	−0.69
407.85	0.1595	0.1599	−0.28
449.64	0.1220	0.1202	1.52
473.54	0.1012	0.1022	−0.99
$P = 10 \text{ MPa}$, $\eta/\text{mPa} \cdot \text{s}$			
306.79	0.4013	0.3996	+0.42
337.54	0.2978	0.2964	+0.46
369.35	0.2259	0.2277	−0.79
407.85	0.1718	0.1725	−0.43
449.64	0.1338	0.1321	+1.28
473.54	0.1130	0.1145	−1.33
$P = 0.1 \text{ MPa}$			
298.24	0.3880	0.3885	−0.12
323.93	0.2979	0.2971	+0.26
348.64	0.2354	0.2360	−0.25
<i>n</i> -Octane			
$P = 5 \text{ MPa}$, $\eta/\text{mPa} \cdot \text{s}$			
308.72	0.4786	0.4754	+0.66
349.46	0.3187	0.3188	−0.04
394.81	0.2197	0.2220	−1.06
436.86	0.1672	0.1647	+1.51
467.46	0.1338	0.1333	+0.37
$P = 10 \text{ MPa}$, $\eta/\text{mPa} \cdot \text{s}$			
308.72	0.5043	0.5008	+0.69
349.46	0.3364	0.3365	−0.03
394.81	0.2334	0.2359	−1.07
436.86	0.1799	0.1771	+1.56
467.46	0.1458	0.1455	+0.20
$P = 0.1 \text{ MPa}$			
298.24	0.5080	0.5092	+0.23
323.93	0.3829	0.3837	+0.22
348.64	0.3034	0.3037	+0.10

$$^a \delta\eta = 100(1 - \eta^{\text{exp}}/\eta[42]).$$

TABLE 4

Deviation statistics between the present measurements of viscosity and calculated with reference correlation for pure components by Huber *et al.* [42] and prediction technique for mixtures by Huber [44]

Fluids	AAD/(%)	Bias/(%)	St. dev./(%)	St. error/(%)	Max. dev./(%)	N
<i>n</i> -Heptane [42]	0.65	0.00	0.80	0.21	1.52	15
<i>n</i> -Octane [42]	0.59	0.26	0.78	0.22	1.56	13
<i>n</i> -Octane + <i>n</i> -heptane [44]	0.50	−0.19	0.66	0.06	1.86	122

agreed well (within 0.03%) with the reference values in the literature [28]. The composition was determined gravimetrically.

3. Results and discussions

3.1. Density measurements

Measurements of the (p, ρ, T, x) relationships of the $\{x_1 n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures were performed at $x_1 = (0.2808, 0.5427, 0.7906)$ for temperatures between (293 and 557) K. The pressure ranged from (0.1 to 10) MPa. The experimental results are presented in tables 5 and 6. Some selected experimental results are shown in figures 1–3 as projections in the ρ - x , ρ - T , and ρ - p planes, together with values reported by other researchers [10–13]. The excess molar volumes were calculated for the $\{x_1 n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures by using the present molar volume data and pure component values calculated from Lemmon and Span [28] reference EOS with the following relation

$$V_m^E = V_{m,\text{mix}} - x_1 V_{m,1} - (1 - x_1) V_{m,2}, \quad (4)$$

where the x_1 is the mole fraction heptane.

The derived values of V_m^E for selected temperature and pressure are given in figure 4 as a function of mole fraction x_1 . Note that the values of V_m^E for the $\{x_1 n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures are negative for measured temperatures and pressures. As one can see from figures 1 and 4, the difference between actual molar volumes of the mixtures V_{mix} and the ideal mixture molar volumes $V_{\text{id}} = x_1 V_{m,1} + (1 - x_1) V_{m,2}$ is small (maximum value at temperature 343.15 and 0.1 MPa is about $0.24 \text{ cm}^3 \cdot \text{mol}^{-1}$). The maximum relative uncertainty δV_m^E in derived values of V_m^E strongly depends on x_1 , V_{mix} , V_1 , V_2 and can be approximately estimated from the following relation

$$\delta V_m^E \cong \delta V_{\text{mix}} V_{\text{mix}} / (V_{\text{mix}} - V_{\text{id}}) + \delta V_1 V_1 / (V_{\text{mix}} - V_{\text{id}}) + \delta V_2 V_2 / (V_{\text{mix}} - V_{\text{id}}), \quad (5)$$

TABLE 5

Experimental densities of $\{x_1 n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures at atmospheric pressure

T/K	$x_1 = 0.2808$ $\rho/(\text{kg} \cdot \text{m}^{-3})$	$x_1 = 0.5427$ $\rho/(\text{kg} \cdot \text{m}^{-3})$	$x_1 = 0.7906$ $\rho/(\text{kg} \cdot \text{m}^{-3})$
293.15	697.8	693.2	688.3
298.15	694.0	689.2	684.2
303.15	689.9	685.0	680.1
308.15	686.1	680.9	675.9
313.15	681.6	676.6	671.6
318.15	677.4	672.7	667.3
323.15	673.2	668.5	663.0
333.15	664.9	659.9	654.4
338.15	661.0	655.6	650.1
343.15	656.8	651.2	645.6
357.15	644.3	638.8	633.0

TABLE 6

Experimental densities of $\{x_1 n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures at high pressures

P/MPa	ρ ($\text{kg} \cdot \text{m}^{-3}$)
$x_1 = 0.2808$ $T = 306.18 \text{ K}$	
2.942	690.3
7.021	693.8
8.826	695.3
$T = 362.65 \text{ K}$	
4.800	646.1
6.720	649.0
9.300	652.5
$T = 374.79 \text{ K}$	
3.256	634.7
8.081	641.5
9.316	643.1
$T = 401.40 \text{ K}$	
4.751	614.2
7.120	618.0
9.310	621.7
$T = 429.34 \text{ K}$	
3.511	585.6
8.237	595.9
9.630	598.7
$T = 451.28 \text{ K}$	
3.900	565.6
5.302	569.0
8.131	576.0
9.621	579.7
$T = 475.85 \text{ K}$	
4.354	539.5
8.492	553.8
10.12	558.1
$T = 494.74 \text{ K}$	
4.250	515.8
6.032	524.3
8.734	535.5
9.220	537.5
$T = 505.34 \text{ K}$	
5.217	507.7
8.904	525.4
9.807	528.4
$T = 525.50 \text{ K}$	
4.771	475.8
5.323	480.4
7.331	494.9
9.533	506.5
$T = 557.23 \text{ K}$	
5.296	430.4
8.238	461.5
9.885	473.3
10.79	479.0

TABLE 6 (continued)

P/MPa	ρ ($\text{kg} \cdot \text{m}^{-3}$)
$x_1 = 0.5427$ $T = 299.24 \text{ K}$	
3.824	691.8
6.865	694.6
9.316	696.4
$T = 331.55 \text{ K}$	
4.119	665.0
5.944	668.1
8.203	670.9
10.10	672.6
$T = 352.61 \text{ K}$	
2.844	648.0
8.237	654.9
10.04	657.0
$T = 380.43 \text{ K}$	
3.110	623.5
5.712	628.4
8.743	633.1
10.23	635.1
$T = 409.59 \text{ K}$	
3.334	598.0
7.257	606.1
9.316	609.6
$T = 424.48 \text{ K}$	
4.903	588.7
7.108	592.8
9.298	597.1
$T = 447.30 \text{ K}$	
4.808	566.0
6.304	569.6
9.463	576.6
$T = 466.78 \text{ K}$	
3.177	538.3
8.649	556.8
9.963	560.6
$T = 483.92 \text{ K}$	
4.703	524.2
7.131	533.5
9.423	541.6
$T = 518.04 \text{ K}$	
4.982	480.1
7.512	496.2
8.983	504.3
$T = 536.57 \text{ K}$	
5.021	449.5
6.931	468.1
9.621	486.1

TABLE 6 (continued)

P/MPa	ρ ($\text{kg} \cdot \text{m}^{-3}$)
$T = 549.34 \text{ K}$	
5.394	431.7
6.766	449.2
8.689	464.6
9.708	471.9
$x_1 = 0.7906$ $T = 307.62 \text{ K}$	
2.608	678.6
7.178	683.0
9.140	684.6
$T = 331.45 \text{ K}$	
4.121	658.7
6.311	662.6
8.523	665.5
10.35	667.1
$T = 365.46 \text{ K}$	
3.432	631.8
8.983	639.4
9.963	640.9
$T = 386.91 \text{ K}$	
3.743	612.4
5.902	616.6
8.793	621.6
10.71	624.7
$T = 401.28 \text{ K}$	
4.100	601.1
6.833	606.2
9.204	610.3
10.51	612.5
$T = 423.26 \text{ K}$	
3.334	578.1
7.433	587.6
9.708	592.4
$T = 466.55 \text{ K}$	
4.570	535.1
7.593	546.3
9.885	553.4
$T = 506.00 \text{ K}$	
5.099	487.7
6.982	499.4
8.492	507.2
9.669	512.6
$T = 555.90 \text{ K}$	
4.982	394.1
6.198	416.1
8.649	445.5
10.24	458.1

where δV_{mix} , δV_1 , and δV_2 are the relative uncertainties in mixture and pure components molar volume determination, respectively. The present results for excess molar

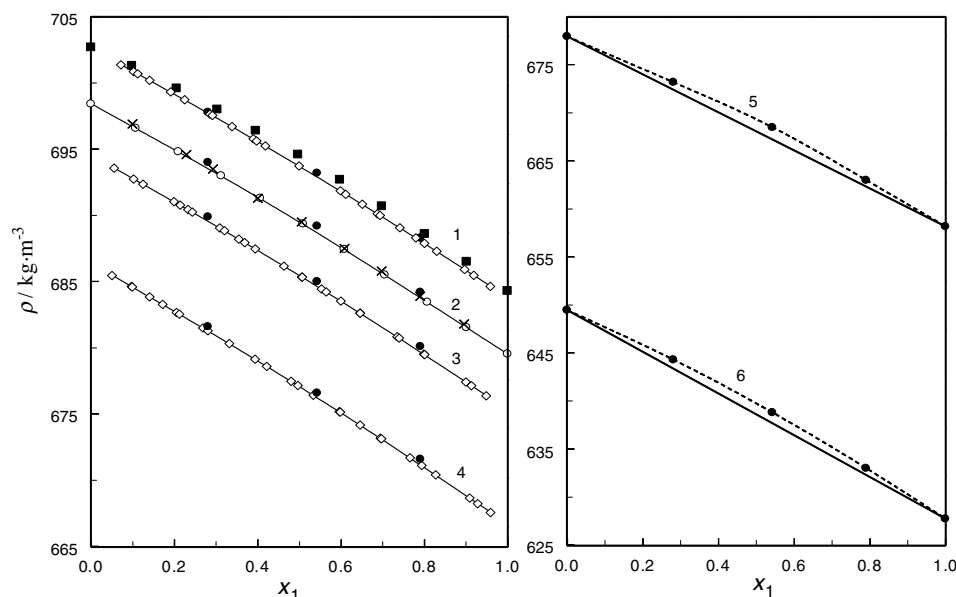


FIGURE 1. Plot of experimental densities ρ for $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures as a function of x_1 at atmospheric pressure for the selected isotherms. ●, this work; ○, Aucejo *et al.* [11]; ■, Cooper and Asfour [10]; ◇, Matos *et al.* [12]; ×, Chevalier *et al.* [13]. The solid and dashed lines are guides for the eye. Isotherms: 1, 293.15 K; 2, 298.15 K; 3, 303.15 K; 4, 313.15 K; 5, 323.15 K; 6, 357.15 K.

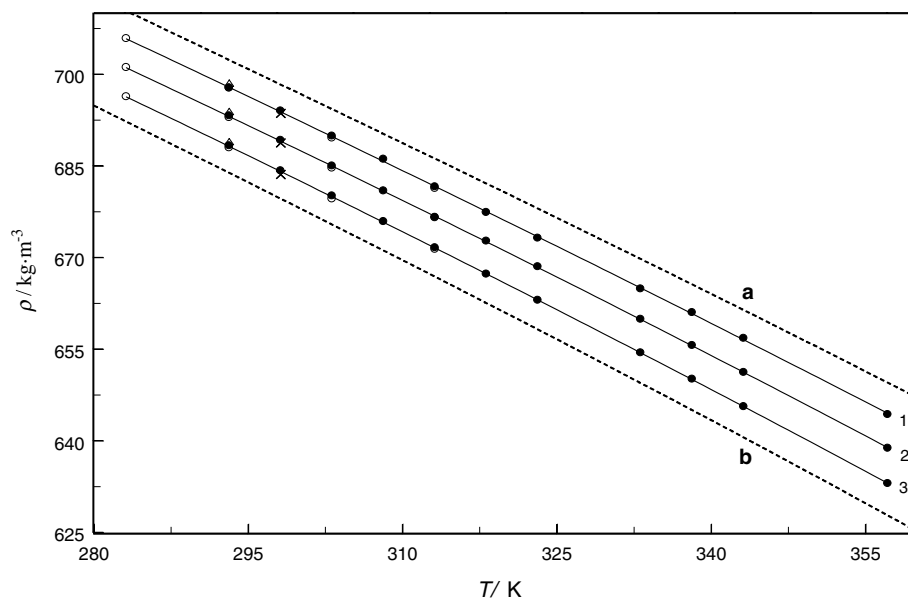


FIGURE 2. Plot of experimental densities ρ for $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures as a function of temperature T at atmospheric pressure for constant mole fraction together with values for the pure components a = $n\text{-C}_8\text{H}_{18}$, b = $n\text{-C}_7\text{H}_{16}$. ●, this work; ○, Matos *et al.* [12]; ×, Aucejo *et al.* [11]; △, Cooper and Asfour [10]; (---), reference EOS calculations (Lemmon and Span [28]) for pure components. 1, $x_1 = 0.2808$; 2, $x_1 = 0.5427$; 3, $x_1 = 0.7906$. The solid lines are guides for the eye.

volumes differ from the data reported by Aucejo *et al.* [11] and Marcos *et al.* [12] by 60%. These deviations are still acceptable because the uncertainty in the V_m^E calculation from equation (5) is about 80% and more, because the relative uncertainty in V_m^E is defined as $\delta V_m^E \cong (V_{\text{mix}} - V_{\text{id}})^{-1}$, where the difference $(V_{\text{mix}} - V_{\text{id}})$ very small and almost zero. This means that the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixture is almost ideal. This is not a surprise because the interaction between the dissimilar molecules of the

components $n\text{-heptane}$ and $n\text{-octane}$ is not different from the interaction between the identical molecules of either $n\text{-heptane}$ or $n\text{-octane}$.

3.1.1. Comparison of densities with the literature data

The present experimental values of density for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures at atmospheric pressure and at temperatures from (293.15 to 313.15) K were compared with the reported data and those from the

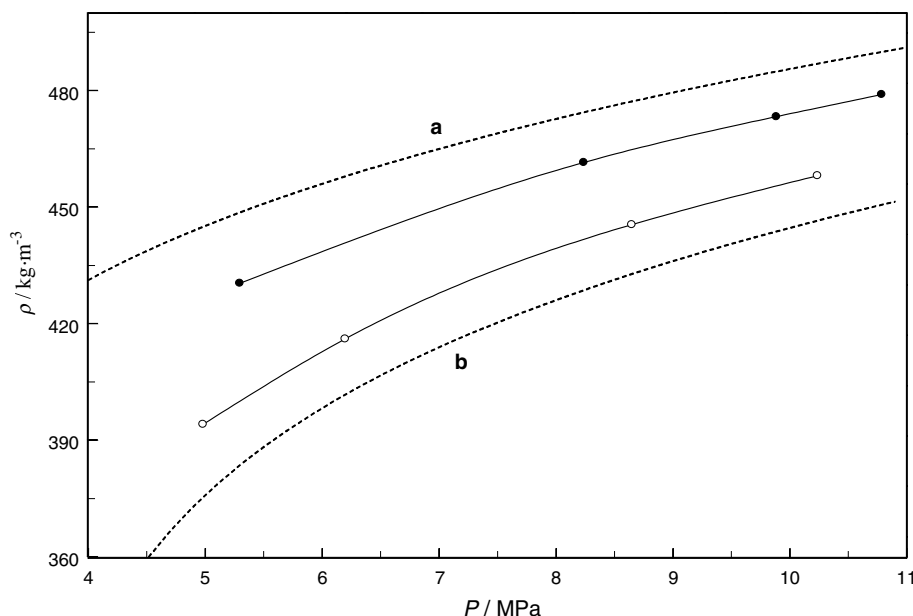


FIGURE 3. Plot of experimental densities ρ of $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures as a function of pressure P at constant compositions and selected isotherms together with values for the pure components a = $n\text{-C}_8\text{H}_{18}$, b = $n\text{-C}_7\text{H}_{16}$. ●, $x_1 = 0.7906$ and $T = 557.23$ K; ○, $x_1 = 0.2808$ and $T = 555.90$ K; (---), reference EOS calculations (Lemmon and Span [28]) for pure components at temperature of 555.15 K. The solid lines are guides for the eye.

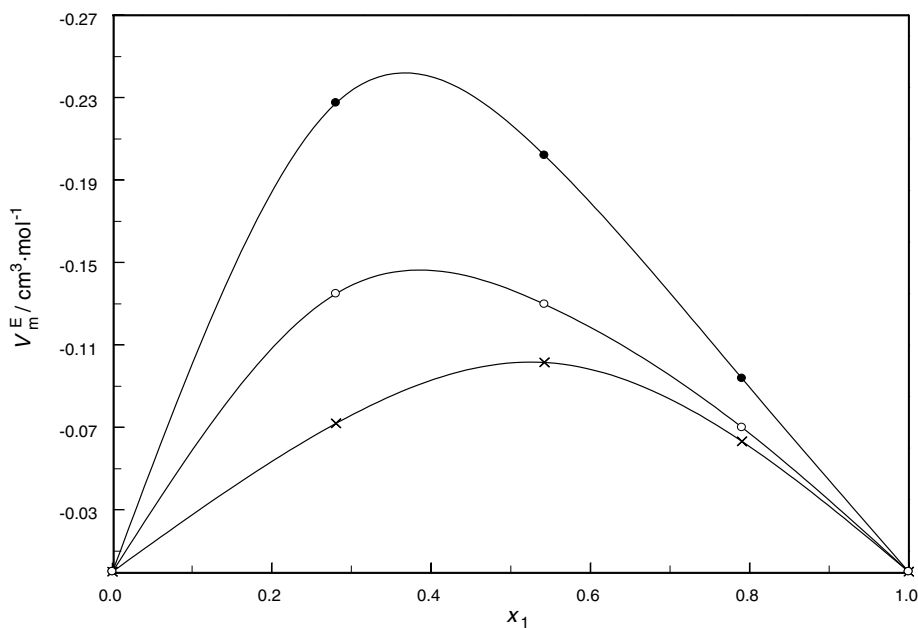


FIGURE 4. Plot of excess molar volumes V_m^E as a function of x_1 for $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ at three selected temperatures and at atmospheric pressure. ×, 293.15 K; ○, 298.15 K; ●, 343.15 K. The solid curves are guides for the eye.

prediction technique. Figures 1–3 show the direct comparison between present densities for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures and those reported by various authors [10–13]. In general, the agreement between all of the available data sets is good. In order to provide quantitative comparison, we used an analytical interpolation procedure for the present data due to the temperature and concentration differences between the various datasets.

We also used the correlations reported by other researchers [10–13] to compare with our data. All reported data at temperatures from (293.15 to 313.15) K lie between 0.01% and 0.08%, which is very close to the estimated uncertainty of our experimental results (0.06%). The agreement between the data of Matos *et al.* [12], Chevalier *et al.* [13], and Aucejo *et al.* [11] and the present data lies within 0.044%, 0.051%, and 0.065%, respectively, while the data by Cooper

and Asfour [10] deviate from the present results within 0.077%, which is still close to the uncertainty of our data. Our data were also compared with the values calculated from a mixture model developed by Lemmon and Jacobsen [43]. The agreement between both predicted and measured values of density lie within 0.23% (deviation statistics see table 2) at temperatures up to 365 K. This agreement is still good because the mixture model by Lemmon and Jacobsen [43] has no adjustable parameters and was developed without using any experimental data for $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures. Lemmon and Jacobsen [43] claimed uncertainty of the calculated values of density from predicting model to be 0.2%.

3.2. Viscosity measurements

The viscosity measurements for $x_1 = (0.2808, 0.5427, 0.7906)$ for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures have been performed over the temperature range from (298 to 473) K at pressures up to 10 MPa. All experimental viscosity data were obtained as a function of temperature at three isobars (0.1, 5, and 10) MPa. The experimental temperature, viscosity, pressure, and composition values for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures are presented in table 7. Some selected experimental viscosity data for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures as an example of the present results are shown in figures 5–7 in the η - x , η - T , and η - p projections together with values reported by other authors. The correlation between the viscosity of liquid mixtures η_{mix} and that of the pure components η_1 and η_2 is very complicated, and no satisfactory theories taking into account the deviations from the viscosities of ideal mixtures η_{id} have been developed so far. When interaction between the different molecules of the components is not appreciably different from the interaction of identical molecules, the mixture is considered to be ideal. As for the viscosity of liquid mixtures consisting of dissimilar molecules, the viscosity deviations $\Delta\eta$ arising from the difference between the viscosity of the mixture η_{mix} and those of the components is

$$\Delta\eta = \eta_{\text{mix}} - x_1\eta_1 - (1 - x_1)\eta_2. \quad (6)$$

The values of viscosity deviations $\Delta\eta$ are negative, $\Delta\eta < 0$, if a weak bond is formed between the components $n\text{-heptane}$ and $n\text{-octane}$. If a strong interaction is formed between the components, $\Delta\eta > 0$. The larger is the value of $\Delta\eta$, the stronger is the interaction between the molecules of the components. The differences in the shape and size of the molecules can also play an important role in this respect. From the present viscosity data for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures and the corresponding values for the pure components calculated with the correlation by Huber *et al.* [42], the values of viscosity deviations $\Delta\eta$ were calculated using the relation (6). The derived values of the viscosity deviations $\Delta\eta$ for selected temperature and pressure are given in figure 8 as a function of concen-

TABLE 7

Experimental viscosities of $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures

T/K	$\eta/(\text{mPa} \cdot \text{s})$		
	$P = 0.1 \text{ MPa}$	$P = 5 \text{ MPa}$	$P = 10 \text{ MPa}$
$x = 0.2808$			
298.39	0.467	0.493	0.521
303.85	0.441	0.465	0.492
312.71	0.400	0.424	0.446
321.71	0.362	0.384	0.405
333.54	0.322	0.341	0.360
344.39	0.291	0.308	0.325
354.08	0.267	0.283	0.299
363.71	0.245	0.261	0.276
373.51		0.241	0.255
382.99		0.223	0.237
393.49		0.206	0.220
413.60		0.178	0.191
432.62		0.156	0.168
455.63		0.133	0.145
473.46		0.118	0.130
$x = 0.5427$			
298.15	0.436	0.461	0.487
303.39	0.414	0.435	0.459
312.74	0.374	0.396	0.419
321.21	0.338	0.359	0.380
333.73	0.300	0.319	0.338
344.40	0.273	0.290	0.307
352.62	0.255	0.270	0.286
363.74	0.232	0.247	0.260
373.97		0.227	0.240
383.18		0.212	0.224
394.51		0.195	0.206
403.23		0.183	0.193
415.96		0.167	0.177
424.55		0.158	0.167
433.62		0.148	0.158
452.73		0.129	0.140
463.18		0.120	0.131
473.10		0.112	0.123
$x = 0.7906$			
298.22	0.408	0.435	0.459
305.74	0.379	0.402	0.425
312.75	0.352	0.373	0.395
323.82	0.314	0.334	0.353
335.62	0.281	0.298	0.316
344.21	0.260	0.276	0.292
353.20	0.239	0.255	0.270
364.62	0.216	0.232	0.246
373.19		0.217	0.230
384.55		0.198	0.211
395.52		0.183	0.195
407.72		0.167	0.180
424.45		0.149	0.160
435.62		0.138	0.149
455.30		0.120	0.132
472.52		0.107	0.118

tration at temperature 298.15 K together with the data reported by Aucejo *et al.* [11]. Note that the values of $\Delta\eta$ for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures are negative for measured temperatures and pressures and therefore a weak bond is formed between the $n\text{-heptane}$ and $n\text{-heptane}$

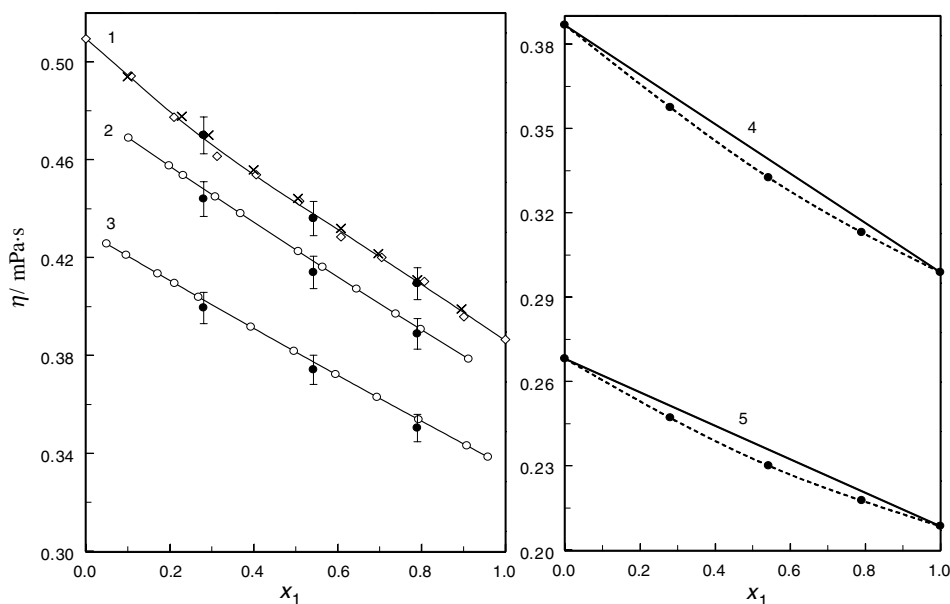


FIGURE 5. Plot of measured values of viscosity η for $\{x_1n\text{-heptane} + (1-x_1)n\text{-octane}\}$ mixtures as a function of mole fraction x_1 along the selected isotherms and at atmospheric pressure together with values reported by other authors from the literature. ●, this work; ○, Matos *et al.* [12]; ◇, Aucejo *et al.* [11]; ×, Chevalier *et al.* [13]. 1, $x_1 = 0.2808$; 2, $x_1 = 0.5427$; 3, $x_1 = 0.7906$. Isotherms: 1, 298.15 K; 2, 303.15 K; 3, 313.15 K; 4, 323.15 K; 5, 363.15 K. The solid and dashed lines are guides for the eye.

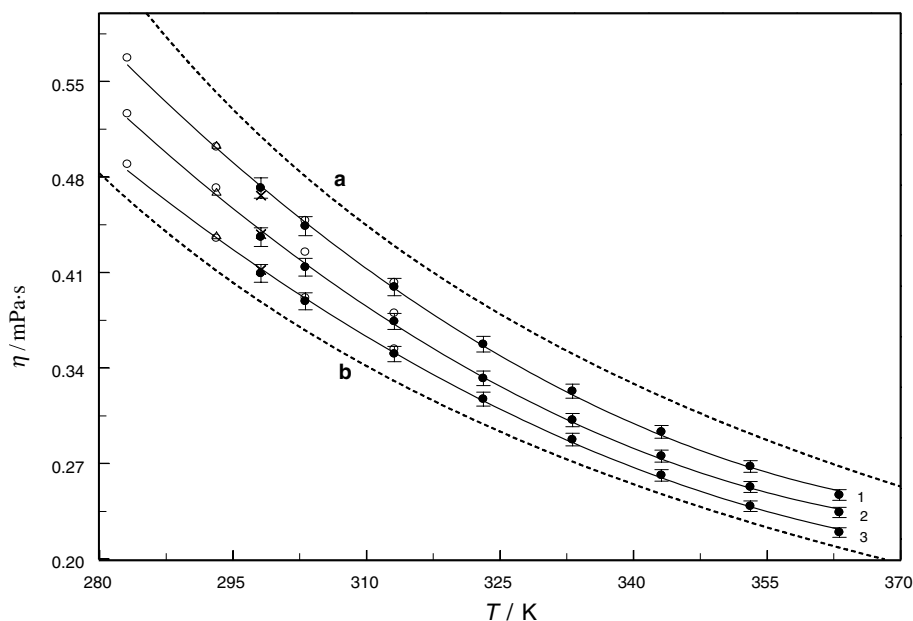


FIGURE 6. Plot of measured values of viscosity η for $\{x_1n\text{-heptane} + (1-x_1)n\text{-octane}\}$ mixtures as a function of temperature T along three selected values of mole fraction and at atmospheric pressure together with values reported by other authors from the literature. ●, this work; ○, Matos *et al.* [12]; ×, Aucejo *et al.* [11]; △, Cooper and Asfour [10]; (---), Huber *et al.* [42] correlation for pure components a = $n\text{-C}_8\text{H}_{18}$, b = $n\text{-C}_7\text{H}_{16}$. 1, $x_1 = 0.2808$; 2, $x_1 = 0.5427$; 3, $x_1 = 0.7906$. The solid curves are guides for the eye.

molecules. The maximum relative uncertainty $\delta(\Delta\eta)$ in viscosity deviations determination can be estimated as

$$\delta(\Delta\eta) \cong \delta\eta_{\text{mix}}\eta_{\text{mix}}/(\eta_{\text{mix}} - \eta_{\text{id}}) + \delta\eta_1\eta_1/(\eta_{\text{mix}} - \eta_{\text{id}}) + \delta\eta_2\eta_2/(\eta_{\text{mix}} - \eta_{\text{id}}), \quad (7)$$

where $\delta\eta_{\text{mix}}$, $\delta\eta_1$, and $\delta\eta_2$ are the relative uncertainties in the determination of the viscosity of the mixture and its

pure components respectively. As one can see from equation (7), the values of $\delta(\Delta\eta) \approx (\eta_{\text{mix}} - \eta_{\text{id}})^{-1}$. As figures 5 and 8 show, the difference between η_{mix} and the ideal mixture viscosity η_{id} for $\{x_1n\text{-heptane} + (1-x_1)n\text{-octane}\}$ is small (within 0.014 mPa·s at $T = 298.15$ K and 0.1 MPa), and therefore the uncertainty in viscosity deviations $\Delta\eta$ estimated from equation (7) is very large. For

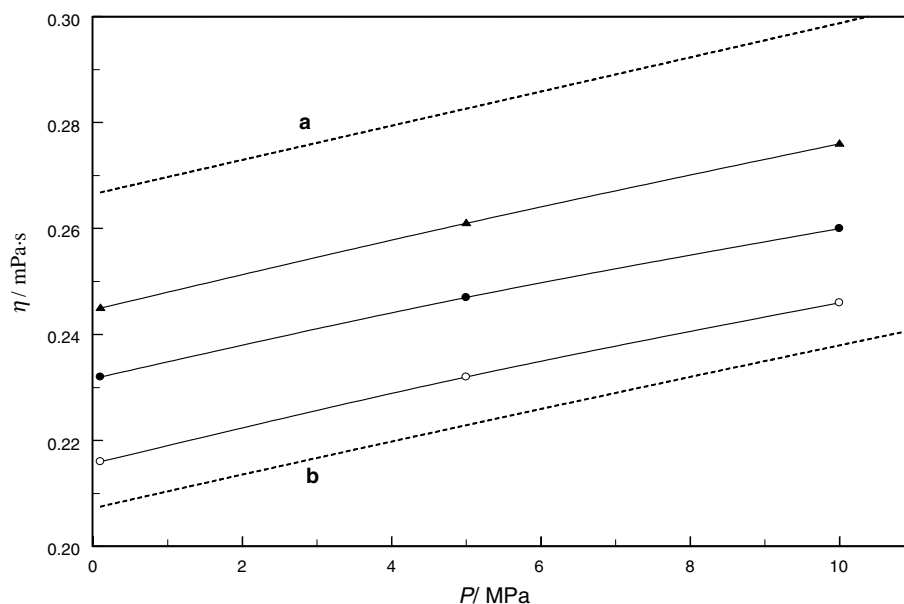


FIGURE 7. Plot of measured values of viscosity η for $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures as a function of pressure P at various temperatures and constant mole fraction together with values for pure components calculated with Huber *et al.* [42] correlation (dashed lines at 363.70 K) a = $n\text{-C}_8\text{H}_{18}$, b = $n\text{-C}_7\text{H}_{16}$. ●, $x_1 = 0.5427$ and $T = 363.74$ K; ○, $x_1 = 0.7906$ and $T = 364.62$ K; ▲, $x_1 = 0.2808$ and $T = 363.71$ K. The solid curves are guides for the eye.

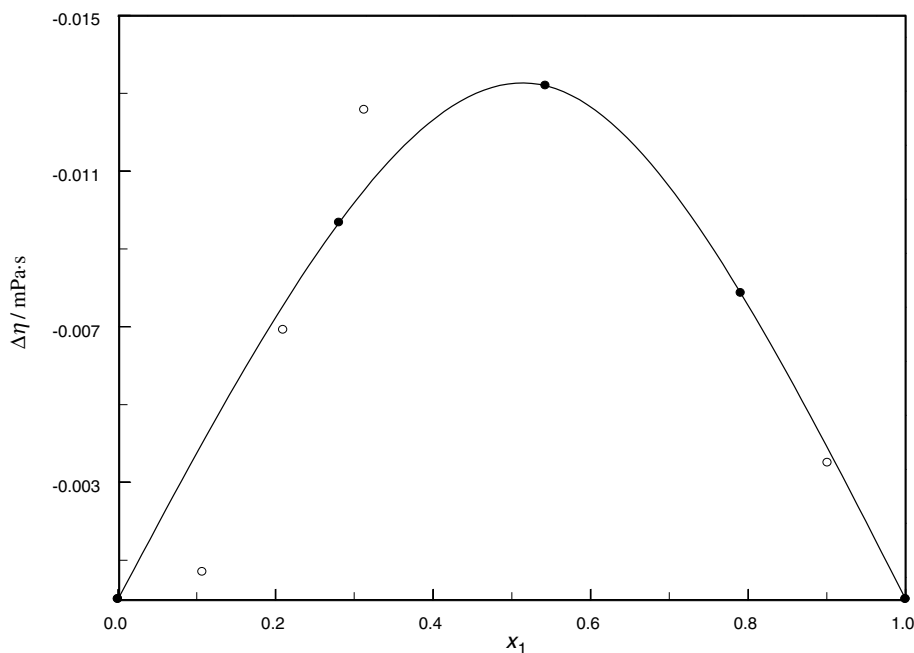


FIGURE 8. Plot of the viscosity deviations $\Delta\eta$ for $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ as a function of mole fraction x_1 at $T = 298.15$ K and 0.1 MPa together with values reported by Aucejo *et al.* [11]. ●, this work; ○, Aucejo *et al.* [11]. The solid curve is a guide for the eye.

example, the minimum uncertainty in $\Delta\eta$ at temperature 298.15 K and at 0.1 MPa is within 58%. The derived values for $\Delta\eta$ were compared with the values reported by Aucejo *et al.* [11] and Matos *et al.* [12]. The differences reach up to 50 %. This remains acceptable because, as was shown above, the maximum uncertainty in $\Delta\eta$ determination lies within 60%.

Grunberg and Nissan [1] proposed the following expression for the viscosity of a mixture

$$\ln(\eta_{\text{mix}}/\eta^\theta) = x_1 \ln(\eta_1/\eta^\theta) + x_2 \ln(\eta_2/\eta^\theta) + x_1 x_2 G(T), \quad (8)$$

where $G(T)$ is the Grunberg–Nissan constant as a function of temperature; $\eta^\theta = 1.0 \text{ mPa} \cdot \text{s}$. This equation has been

recommended also by Irving [3] to represent viscosity data for binary mixtures. Dymond and Young [45,46] and Zeberg-Mikkelsen *et al.* [47] successfully used this equation to describe the experimental viscosity data for hydrocarbon mixtures. In the present work, we applied equation (8) to the viscosity data for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures. The values of the Grunberg–Nissan constant were determined as a function of temperature (see figure 9). The concentration dependence of the Grunberg–Nissan constant for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures is almost zero at each temperature studies. As figure 9 demonstrates, the Grunberg–Nissan constant is negative and monotonically decreases with increasing temperature. For mixtures with $n\text{-alkane } n < 12$ where n is the carbon number, the values of G are negative [45,46]. For the ideal mixtures, the values of the Grunberg–Nissan constant are zero ($G = 0$). The linear relation $G(T) = 0.1565545 - 0.0005565(T/K)$ represented the temperature dependence of $G(T)$ for the present viscosity data. The present viscosity data in the temperature range from (298 to 373) K agree with values calculated from equation (8) to within 0.47% when $G(T)$ is taken as a function of temperature.

The molar excess Gibbs energy of activation for flow ΔE_a can be derived from the present experimental viscosity data for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures from the relation

$$\ln(\eta_{\text{mix}} V_{\text{mix}}^*) = x_1 \ln(\eta_1 V_1^*) + (1 - x_1) \ln(\eta_2 V_2^*) + \frac{\Delta E_a}{RT}, \quad (9)$$

where ΔE_a is the molar excess activation energy of viscous flow; $V_{\text{mix}}^* = V_{\text{mix}}/hN_A$, $V_1^* = V_1/hN_A$, $V_2^* = V_2/hN_A$, $N_A = 6.02252 \cdot 10^{26} \text{ kmol}^{-1}$ is the Avogadro's constant,

$h = 6.6256 \cdot 10^{-34} \text{ J} \cdot \text{s}^{-1}$ is the Planck's constant; R is the gas constant. This relation is derived from Eyring's theory of the dynamic viscosity [48] for pure fluids. Coursey and Heric [2] successfully applied the equation (9) to viscosity of $n\text{-alkane}$ mixtures at $T = 298.15 \text{ K}$. The values of ΔE_a calculated with the present viscosity and density data for the mixture and pure components as a function of mole fraction for three selected temperatures is shown in figure 10. As one can see from figure 10, over the considered temperature range the values of ΔE_a are negative. This implies that the viscosity of the mixtures is reduced compared with that of an ideal mixture. The maximum values of the $|\Delta E_a|$ is about $25 \text{ J} \cdot \text{mol}^{-1}$, which means the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixture is a very weakly interacting system. The mixing law (9) represents our viscosity results for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixture within 0.8% to 1.0%.

3.2.1. Comparison of viscosities with literature data

The present experimental values of viscosity for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures at atmospheric pressure were compared with the data and correlations reported by other authors in the literature. figures 5 and 6 demonstrate the direct comparison of our viscosity data with those reported by various authors in the literature [10–13]. As one can see from these figures, the agreement between various data sets at low temperatures and atmospheric pressure is good. These figures illustrate that our data are consistent with most literature values at various temperatures and concentrations. The quantitative comparison of our measurements with the data reported by other authors was difficult due to the temperature and

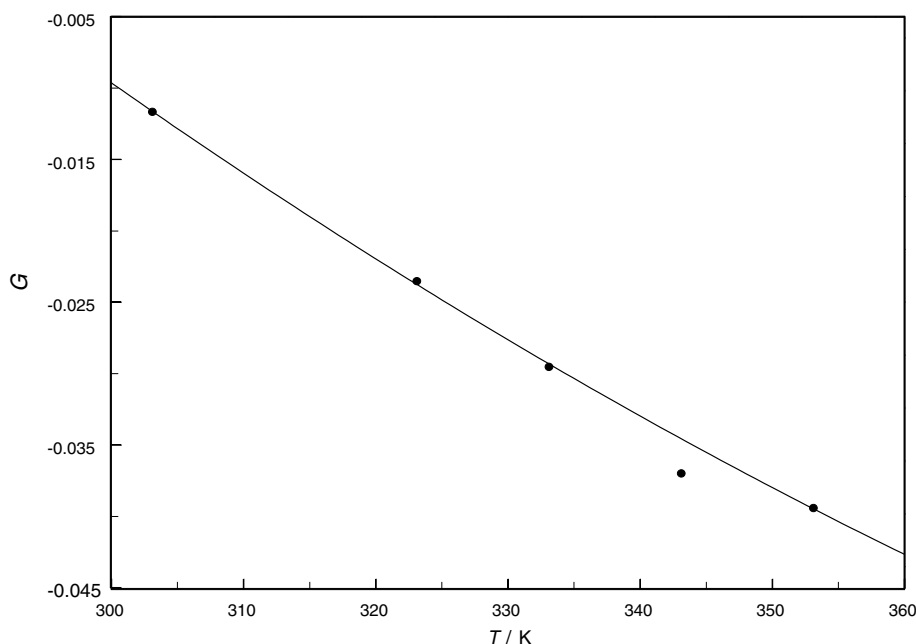


FIGURE 9. Plot of the Grunberg–Nissan constant as a function of temperature at atmospheric pressure for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixture.

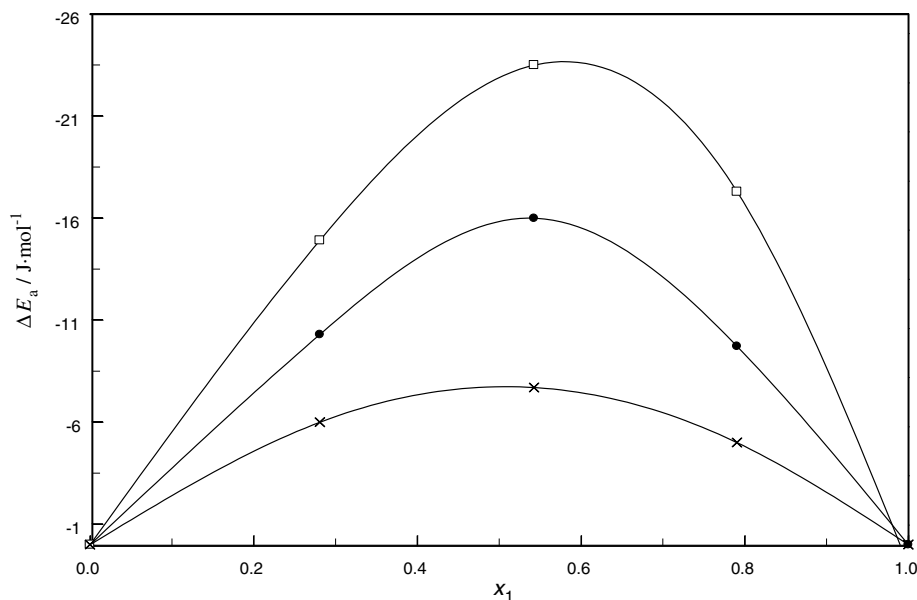


FIGURE 10. Plot of the molar excess activation energy of viscous flow ΔE_a as function of x_1 for $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures at three selected temperatures and at atmospheric pressure. ●, 298.15 K; ×, 303.15 K; □, 325.15 K.

concentration differences between the various datasets and therefore, a regression analysis was employed for our data. We also used the correlations reported by other researchers [10–13] to compare with present data. The relative percentage deviations between our viscosity results for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures and those reported by others were calculated. All reported data at temperatures from (293.15 to 313.15) K lie between 0.5% and 0.9%, which is lower than the experimental estimated uncertainty of the present results (1.6%). Our data were analytically extrapolated to low temperature (to 293.15 K) in order to compare with the data by Cooper and Asfour [10] and the agreement lies within 1.39%, which is acceptable. Excellent agreement within 0.51% and 0.62% is found between our measurements and those reported by Chevalier *et al.* [13] and Aucejo *et al.* [11], respectively at $T = 298.15$ K, while the data of Matos *et al.* [12] agree

within 0.88% at $T = 303.15$ and $T = 313.15$ K. The maximum deviation of 1.09% is found at $T = 303.15$ K and $x_1 = 0.5427$. Our results were also compared with the values calculated from an extended corresponding states model by Huber [44] and the agreement is excellent (average absolute deviation, AAD = 0.50%, see table 4). The deviation plot is given in figure 11. These good agreements confirm the reliability and consistency of our results.

Assael *et al.* [49–51] developed a method to predict the dense fluid viscosity coefficients for binary n -alkane mixtures over extended ranges of temperature and pressure. The values of viscosity predicted from this model were compared with our data. The deviations at atmospheric pressure and at low temperatures (up to 363 K) lie within (2 to 3)%, while at high pressures and high temperatures the deviations reached 4% to 5%. This is good because the model has no adjustable parameters.

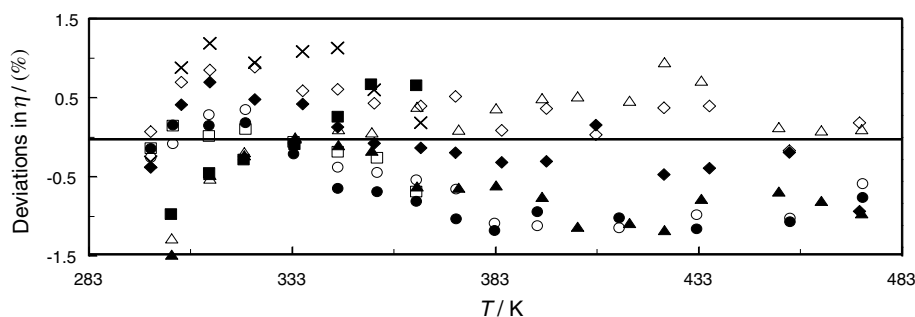


FIGURE 11. Plot of percentage density deviations, $\delta\eta = 100(\eta_{\text{exp}} - \eta_{\text{cal}})/\eta_{\text{exp}}$, of the experimental viscosities for $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures against temperature. The $\delta\eta$ was determined from the values calculated from an extended corresponding states model by Huber [44]. ●, 10 MPa ($x_1 = 0.2808$); ○, 5 MPa ($x_1 = 0.2808$); □, 0.1 MPa ($x_1 = 0.2808$); ▲, 10 MPa ($x_1 = 0.5427$); △, 5 MPa ($x_1 = 0.5427$); ■, 0.1 MPa ($x_1 = 0.5427$); ◆, 10 MPa ($x_1 = 0.7906$); ◇, 5 MPa ($x_1 = 0.7906$); ×, 0.1 MPa ($x_1 = 0.7906$).

4. Conclusions

Density and viscosity for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ binary mixtures at $x_1 = (0.2808, 0.5427, 0.7906)$ have been measured with a constant-volume piezometer and capillary flow techniques, respectively. Measurements were made at pressures up to 10 MPa and at temperatures from (293 to 557) K for the density and from (298 to 473) K for the viscosity. The present viscosity results for the mixtures are in excellent agreement (within 0.5%) with the values predicted by an extended corresponding states model by Huber [44], while our density results deviate from the values predicted by a mixture model developed by Lemmon and Jacobsen [43] within 0.23% (the uncertainty in calculating values of density is 0.2%). The excess molar volumes V_m^E and viscosity deviations $\Delta\eta$ were derived using the measured values of density and viscosity for the mixtures and pure components. The small negative deviations of the mixture molar volumes and viscosities from the ideal mixture values were found. The temperature dependence of the Grunberg–Nissan constant was studied using the present viscosity data. The Grunberg–Nissan constant is negative and monotonically decreases with increasing temperature. The values of the molar excess Gibbs energy of activation for flow ΔE_a as a function of temperature were calculated from the present experimental viscosity data for the $\{x_1n\text{-heptane} + (1 - x_1)n\text{-octane}\}$ mixtures. Over the temperature and density ranges considered, the values of ΔE_a are negative. This implies that the viscosity of the mixture is reduced compared with that of the ideal mixture.

References

- [1] L. Grunberg, A.H. Nissan, *Nature* 164 (1949) 799–800.
- [2] B.M. Coursey, E.L. Heric, *Mol. Phys.* 13 (1967) 287–291.
- [3] J.B. Irving, N.E.L. Report No. 631, National Engineering Laboratory, East Kilbride, Glasgow, 1977.
- [4] A.S. Teja, P. Rice, *Chem. Eng. Sci.* 36 (1981) 7–10.
- [5] R.P. Chhabra, T. Sridhar, *Chem. Eng. J.* 40 (1989) 39–43.
- [6] R.P. Chhabra, *AIChE J.* 38 (1992) 1657–1661.
- [7] H. Liu, W. Wang, Ch.-H. Chang, *Ind. Eng. Chem. Res.* 30 (1991) 1617–1624.
- [8] S.P. Tan, H. Adidharma, B.F. Towler, M. Radosz, *Ind. Eng. Chem. Res.*, in press.
- [9] A.J. Queimada, S.E. Quinones-Cisneros, I.M. Marrucho, J.A.P. Coutinho, E.H. Stenby, *Int. J. Thermophys.* 24 (2003) 1221–1239.
- [10] E.F. Cooper, A.A. Asfour, *J. Chem. Eng. Data* 36 (1991) 285–288.
- [11] A. Aucejo, M.C. Burguet, R. Muñoz, J.L. Marques, *J. Chem. Eng. Data* 40 (1995) 141–147.
- [12] J.S. Matos, J.L. Trenzado, E. Gonzalez, R. Alcalde, *Fluid Phase Equilib.* 186 (2001) 207–234.
- [13] J.L.E. Chevalier, P.J. Pettrino, Y.H. Gaston-Bonhomme, *J. Chem. Eng. Data* 40 (1990) 206–212.
- [14] I.M. Abdulagatov, N.D. Azizov, *J. Sol. Chem.* 32 (2003) 573–599.
- [15] I.M. Abdulagatov, N.D. Azizov, *Int. J. Thermophys.* 24 (2003) 1581–1610.
- [16] I.M. Abdulagatov, N.D. Azizov, *J. Sol. Chem.* 33 (2004) 1305–1331.
- [17] I.M. Abdulagatov, N.D. Azizov, *J. Chem. Thermodyn.* 36 (2004) 829–843.
- [18] I.M. Abdulagatov, N.D. Azizov, *Fluid Phase Equilib.* 216 (2004) 189–199.
- [19] I.M. Abdulagatov, N.D. Azizov, *High Temp. High Press.* 35/36 (2003/2004) 477–498.
- [20] I.M. Abdulagatov, N.D. Azizov, *J. Chem. Thermodyn.* 36 (2004) 17–27.
- [21] I.M. Abdulagatov, N.D. Azizov, *Chem. Geol.*, in press.
- [22] N.D. Azizov, T.S. Akhundov, *Russ. J. Appl. Chem.* 12 (1997) 1955–1959.
- [23] N.D. Azizov, T.S. Akhundov, *Russ. J. Appl. Chem.* 70 (1997) 1867–1871.
- [24] N.D. Azizov, T.S. Akhundov, *Russ. J. High Temp.* 36 (1998) 385–390.
- [25] N.D. Azizov, T.S. Akhundov, L.A. Azizova, *Russ. J. High Temp.* 34 (1996) 973–977.
- [26] W. Wagner, A. Pruß, *J. Phys. Chem. Ref. Data* 31 (2002) 387–535.
- [27] F.G. Keyes, L.B. Smith, *Proc. Am. Acad. Arts Sci.* 68 (1933) 505–521.
- [28] E.W. Lemmon, R. Span, *J. Chem. Eng. Data*, in press.
- [29] I.M. Abdulagatov, N.D. Azizov, *J. Chem. Eng. Data* 48 (2003) 1549–1556.
- [30] I.M. Abdulagatov, N.D. Azizov, *J. Chem. Eng. Data* 49 (2004) 1444–1450.
- [31] I.M. Abdulagatov, N.D. Azizov, *Ind. Eng. Chem. Res.* 44 (2005) 416–425.
- [32] I.M. Abdulagatov, N.D. Azizov, *Int. J. Thermophys.* 26 (2005) 593–635.
- [33] I.M. Abdulagatov, N.D. Azizov, *J. Sol. Chem.* 34 (2005) 645–685.
- [34] I.M. Abdulagatov, N.D. Azizov, *Thermochim. Acta* 439 (2005) 8–20.
- [35] I.M. Abdulagatov, A.B. Zeinalova, N.D. Azizov, *Fluid Phase Equilib.* 227 (2005) 57–70.
- [36] I.M. Abdulagatov, N.D. Azizov, *J. Sol. Chem.* 33 (2004) 1501–1520.
- [37] I.M. Abdulagatov, A.B. Zeinalova, N.D. Azizov, *J. Chem. Thermodyn.* 38 (2006) 179–189.
- [38] I.M. Abdulagatov, A.B. Zeinalova, N.D. Azizov, *Electrochim. Acta* 439 (2005) 8–20.
- [39] I.M. Abdulagatov, A.B. Zeinalova, N.D. Azizov, *J. Mol. Liquids*, in press.
- [40] N.D. Azizov, T.S. Akhundov, *Russ. J. Phys. Chem.* 71 (1997) 1955–1959.
- [41] J.F. Swindells, J.R. Coe Jr., T.B. Godfrey, *J. Res. NBS* 48 (1952) 1–31.
- [42] M.L. Huber, A. Laesecke, H.W. Xiang, *Fluid Phase Equilib.* 224 (2004) 263–270.
- [43] E.W. Lemmon, R.T. Jacobsen, *Int. J. Thermophys.* 20 (1999) 825–835.
- [44] M. Huber, An extended corresponding states model: In development at NIST (personal communication).
- [45] J.H. Dymond, K.J. Young, *Int. J. Thermophys.* 1 (1980) 331–344.
- [46] J.H. Dymond, K.J. Young, *Int. J. Thermophys.* 2 (1981) 237–247.
- [47] C.K. Zeberg-Mikkelsen, A. Baylaucq, G. Watson, C. Boned, *Int. J. Thermophys.* 26 (2005) 1289–1302.
- [48] S. Glasstone, K.J. Laidler, H. Eyring, *The Theory of Rate Processes, the Kinetics of Chemical Reactions, Viscosity, Diffusion, and Electrochemical Phenomena*, McGraw-Hill, New York, 1941.
- [49] M.J. Assael, E. Charitidou, J.H. Dymond, M. Papadaki, *Int. J. Thermophys.* 13 (1992) 237–249.
- [50] M.J. Assael, J.H. Dymond, M. Papadaki, P.M. Patterson, *Int. J. Thermophys.* 13 (1992) 659–669.
- [51] M.J. Assael, J.H. Dymond, M. Papadaki, *Fluid Phase Equilib.* 75 (1992) 287–297.