

Dynamic viscosities of binary mixtures of cycloalkanes with primary alcohols at $T = (293.15, 298.15, \text{ and } 303.15) \text{ K}$: New UNIFAC-VISCO interaction parameters

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Abstract

In this work, dynamic viscosities, densities, and speed of sound have been measured over the whole composition range and 0.1 MPa for the binary mixtures (cyclopentane and cyclohexane with ethanol, 1-propanol, and 1-butanol) at several temperatures (293.15, 298.15, 303.15) K along with the properties of the pure components. Excess molar volumes, molar isentropic compression, excess molar isentropic compression, and excess free energy of activation for the binary systems at the above mentioned temperatures, were calculated and fitted to the Redlich–Kister equation to determine the fitting parameters and the root-mean-square deviations. The UNIQUAC equation was used to correlate the experimental viscosity data. The UNIFAC-VISCO method and ASOG-VISCO method, based on contribution groups, were used to predict the dynamic viscosities of the binary mixtures. The interaction parameters of cycloalkanes with primary alcohol ($\text{CH}_{\text{cy}}/\text{-OH}$) have been determined for their application in the predictive UNIFAC-VISCO method.

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Keywords: Viscosity; Density; Speed of sound; Excess molar volume; Molar isentropic compression; Excess molar isentropic compression; Excess free energy of activation; Cyclohexane; Cyclopentane; Ethanol; 1-Propanol; 1-Butanol; UNIQUAC; UNIFAC-VISCO; ASOG-VISCO

1. Introduction

Among all physical properties, the knowledge of the viscosity of liquid mixtures and their dependence with composition and temperature is very important for design the industrial processes. Indeed, viscosity is a fundamental characteristic of substances such as adhesives, lubricants, paintings, *etc.* As an extension of our work concerning dynamic viscosity of binary systems alkanes with alcohols [1–3], in this paper, we show experimental dynamic viscosity, density, and speed of sound data of $\{x_1 \text{ cyclohexane} + (1 - x_1) \text{ ethanol}\}$, $\{x_1 \text{ cyclohexane}$

$+ (1 - x_1) \text{ 1-propanol}\}$, $\{x_1 \text{ cyclohexane} + (1 - x_1) \text{ 1-butanol}\}$, $\{x_1 \text{ cyclopentane} + (1 - x_1) \text{ ethanol}\}$, $\{x_1 \text{ cyclopentane} + (1 - x_1) \text{ 1-propanol}\}$, and $\{x_1 \text{ cyclopentane} + (1 - x_1) \text{ 1-butanol}\}$ at $T = (293.15, 298.15, \text{ and } 303.15) \text{ K}$. Experimental data were used to calculate excess molar volumes, molar isentropic compression, excess molar isentropic compression, and excess free energy of activation over the entire mole fraction range. Viscosity data were correlated using the UNIQUAC [4] equation.

The UNIFAC-VISCO [5,6] and ASOG-VISCO [7] methods have been applied to predict the viscosity of these systems and the results were compared with the experimental data. Both methods are based on the Eyring theory [8] and on group contributions methods. To improve the results of the prediction of the UNIFAC-VISCO method,

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the interaction parameter $\text{CH}_{\text{cy}}/\text{-OH}$ have been determined.

2. Experimental

2.1. Chemicals

The pure components were supplied by Fluka (cyclohexane and cyclopentane) and Merck (1-propanol, 1-butanol, and 1-pentanol). The components were degassed ultrasonically, and dried over molecular sieves Type 4 Å, $4 \cdot 10^{-8}$ cm, that were supplied by Aldrich, and kept in inert argon with a maximum content in water of $2 \cdot 10^{-6}$ by mass fraction. The maximum water contents of the liquids were determined using a Metrohm 737 KF coulometer. Their mass fraction purities were >0.998 for cyclohexane, ethanol, and 1-butanol, >0.990 for 1-propanol and cyclopentane.

2.2. Apparatus and procedure

Samples were prepared by mass using a Mettler AX-205 Delta Range balance with a precision of $\pm 10^{-5}$ g, covering the whole composition range of the mixture.

Kinematic viscosities were determined using an automatic viscosimeter Lauda PVS1 with two Ubbelohde capillary microviscosimeters of 0.4 mm and 0.53 mm diameter. Gravity fall is the principle of measurement on which this

viscosimeter is based. The capillary is maintained in a D20KP LAUDA thermostat with a resolution of 0.01 K. The capillaries are calibrated and accredited by the company. The uncertainty of the capillary diameter is ± 0.005 mm. In order to verify the calibration, the viscosity of the pure liquids were compared with data recently published (table 1). The uncertainty in the viscosimeter measurement is ± 0.001 mPa · s. The equipment has a control unit PVS1 (Processor Viscosity System) that is a PC-controlled instrument for the precise measurement of fall time, using standardized glass capillaries, with an accuracy of 0.01 s.

The densities and the speed of sound of the pure liquids and mixtures were measured using an Anton Paar DSA-5000 digital vibrating tube densimeter. Uncertainty in density measurement is $\pm 2 \cdot 10^{-6}$ g · cm⁻³, and for the speed of sound is ± 0.1 m · s⁻¹.

3. Results and discussion

Dynamic viscosity, density, speed of sound, excess molar volume and molar isentropic compression, excess molar isentropic compression, and excess free energy of activation for the binary systems $\{x_1$ cyclohexane + $(1 - x_1)$ ethanol $\}$, $\{x_1$ cyclohexane + $(1 - x_1)$ 1-propanol $\}$, $\{x_1$ cyclohexane + $(1 - x_1)$ 1-butanol $\}$, $\{x_1$ cyclopentane + $(1 - x_1)$ ethanol $\}$, $\{x_1$ cyclopentane + $(1 - x_1)$ 1-propanol $\}$, and $\{x_1$ cyclopentane + $(1 - x_1)$ 1-butanol $\}$ at $T = (293.15, 298.15$

TABLE 1

Comparison of density ρ , and viscosity η , with the literature data for pure components at $T = 298.15$ K

| Component | $\rho/(\text{g} \cdot \text{cm}^{-3})$ | | $10^3 \eta/(\text{Pa} \cdot \text{s})$ | | $u/(\text{m} \cdot \text{s}^{-1})$ | | $C_{p,i}^*/(\text{J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1})$ |
|--------------|--|--|--|--|------------------------------------|--|--|
| | Experimental | Literature | Experimental | Literature | Experimental | Literature | Literature |
| Cyclohexane | 0.77392 | 0.7738 ^a 0.77389 ^c | 0.887 | 0.888 ^a 0.883 ^b | 1254 | 1255 ⁱ | 156.04 ^l |
| Cyclopentane | 0.73955 | 0.73969 ^d 0.73947 ^e | 0.416 | 0.416 ^c | 1206 | 1206 ^e | 130.62 ^c |
| Ethanol | 0.78546 | 0.7854 ^f | 1.082 | 1.082 ^f | 1145 | 1143 ^j 1145 ^k | 112.03 ^m |
| 1-Propanol | 0.79958 | 0.7996 ^c | 1.915 | 1.927 ^g 1.898 ^h | 1206 | 1206 ^j | 145.21 ⁿ |
| 1-Butanol | 0.80575 | 0.80575 ^c | 2.571 | 2.571 ^c | 1240 | 1240 ^j | 177.10 ^o |

^a Petrino et al. [9].

^b Aminabhavi et al. [10].

^c Riddick et al. [11].

^d Guzman et al. [12].

^e Pereiro et al. [13].

^f Nikam et al. [14].

^g Aminabhavi et al. [15].

^h Aminabhavi et al. [16].

ⁱ Rodriguez et al. [17].

^j Rodriguez et al. [18].

^k Iloukhani et al. [19].

^l Cerdeirina et al. [20].

^m Peleteiro et al. [21].

ⁿ Cerdeirina et al. [22].

^o Troncoso et al. [23].

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