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Viscometric Behavior of Binary Mixtures of Butan-2-one with Benzene at T = (303.15, 313.15, and 323.15) K

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ABSTRACT: Viscosities of binary liquid mixtures of butan-2-one with benzene were measured at a number of mole fractions at T = (303.15, 313.15, and 323.15) K and atmospheric pressure. The viscometric behavior is discussed on the basis of structural and geometrical effects between the components. In addition, kinematic viscosities were calculated using the UNIFAC-VISCO model. The predicted kinematic viscosities are in good agreement with the experimental ones.

■ INTRODUCTION

The nature of the interaction among polar and nonpolar liquid mixtures is important for understanding their behavior in analytical applications such as conductometric, potentiometric, and electroanalytical techniques that work in mixed solvents, chromatographic applications, etc. ^{1–4} These data are also useful in process engineering design applications and other related areas and have drawn considerable attention from many investigators. ^{5–20} In this work, which is a part of our research on the accumulation of physical property data of binary organic liquid mixtures, we present here viscosity data for binary mixtures of butan-2-one with benzene at T=(303.15, 313.15, and 323.15) K. The density data for the same system were reported previously. ²³

The aprotic and protophilic liquid butan-2-one (dipole moment, $\mu_1=2.8$ D; relative permittivity at 20 °C, $\varepsilon^{20^{\circ}\mathrm{C}}=18.5$)²⁷ herein is considered as the solute, while the apolar, aprotic, and nonprotophilic liquid benzene ($\mu_1=0$ D, $\varepsilon^{20^{\circ}\mathrm{C}}=2.28$)²⁷ is considered as the solvent. Both liquids are widely used as solvents in scientific studies and industrial applications. The experimental data are interpreted using some previous observations for the binary systems. In addition, kinematic viscosities predicted using the UNIFAC—VISCO model^{28,29} are compared with the experimental ones.

In the literature, there are reports on the viscometric behavior of binary systems of butan-2-one with benzene at temperatures other than those reported in this work, $^{30-36}$ and Jayalakshmi and Reddy 37 have reported data at T=303.15 K. However, no experimental viscosity data for mixtures of butan-2-one with benzene under comparable conditions at T=(313.15) and (323.15) K are available.

■ EXPERIMENTAL SECTION

Butan-2-one (Merck, 0.98 mass fraction purity) and benzene (BDH, 0.99 mass fraction purity) were used without further treatment. The solvent purity was ascertained by comparing the densities and viscosities of the liquids with the available literature data (Table 1).

Table 1. Comparison of Experimental Densities $(\rho_{\rm exp})$ and Viscosities $(\eta_{\rm exp})$ of Pure Liquids with Literature Values at $T=303.15~{\rm K}$

	ρ	$ ho_{ m exp}/{ m g\cdot cm}^{-3}$		$\eta_{ m exp}/{ m mPa}$ · s		
liquid	exp	lit	ref	exp	lit	ref
butan-2-one	0.7946	0.7946	40	0.367	0.366	40
		0.7948	41		0.372	42
benzene	0.8681	0.8683	41	0.564	0.562	40
		0.8684	43		0.560	43

All of the binary mixtures were prepared by mass on an analytical balance (B204-S, Mettler Toledo, Greifensee, Switzerland) with an uncertainty of \pm 0.0001 g and operating in a drybox. Mixtures were made just before use and were completely miscible over the whole composition range. Caution was taken to prevent evaporation of the samples after preparation. The uncertainty in the mole fraction was estimated to be less than $\pm 1 \cdot 10^{-4}$. The densities of liquids were measured using a 25 mL specific gravity bottle that had previously been calibrated with redistilled water with an average uncertainty of 0.0002 $g \cdot cm^{-3}$. The uncertainties in the viscosities were on the order of ± 0.005 mPa·s. An A-type Ostwald viscometer previously calibrated with redistilled water was used to measure the viscosities. An electronic digital stopwatch with an uncertainty of \pm 0.01 s was used for flow time measurements. A transparent glasswalled thermostatic water bath was used, and the uncertainty in the temperature during the measurements was \pm 0.05 K. For all of the mixture compositions and the pure solvents, triplicate measurements were performed, and the average of these values was considered in all calculations.

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Table 2. Composition and Experimental Viscosities ($\eta_{\rm exp}$) for Binary Mixtures of Butan-2-one (1) with Benzene (2) at $T=(303.15,\,313.15,\,{\rm and}\,\,323.15)~{\rm K}$

	$\eta_{ m exp}/{ m mPa}$ · s				
x_1	T = 303.15 K	T = 313.15 K	T = 323.15 K		
0	0.564	0.498	0.442		
0.1010	0.534	0.473	0.422		
0.1999	0.508	0.448	0.403		
0.2988	0.478	0.427	0.384		
0.3969	0.452	0.409	0.369		
0.4996	0.432	0.393	0.354		
0.5988	0.417	0.378	0.343		
0.6993	0.401	0.367	0.333		
0.7931	0.389	0.355	0.324		
0.8961	0.376	0.345	0.316		
1	0.367	0.334	0.306		

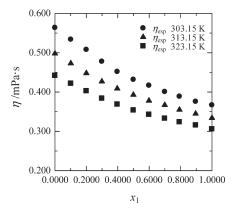


Figure 1. Experimental viscosities for the system butan-2-one (1) + benzene (2) at T = (303.15, 313.15, and 323.15) K.

■ RESULTS AND DISCUSSION

The binary compositions and experimental viscosities $(\eta_{ ext{exp}})$ at all of the investigated temperatures are summarized in Table 2. Values of $\eta_{\rm exp}$ for the binary mixtures of butan-2-one and benzene as a function of the mole fraction of butan-2-one (x_1) at T = (303.15,313.15, and 323.15) K are shown in Figure 1. It can be observed that upon the addition of butan-2-one to benzene, the viscosity decreases slowly up to a substantial concentration of ketone. The viscosity of a mixture strongly depends on the structures and bond enthalpies of the liquids and consequently on the molecular interactions between the components of the mixture.³⁸ The positive values of $\eta_{\rm exp}$ can be attributed to the dispersion-type interaction between butan-2-one and benzene.³⁹ Butan-2-one, like all other linear ketones, has a stiff propanone skeleton and is less inflexible than other linear organic solvents. 10 However, a butan-2-one molecule has a light molecular structure relative to the other linear ketones. Therefore, in the lowconcentration region, a butan-2-one molecule may be able to orient itself into a less flow-resistant arrangement as a result of the dispersion force created from the apolar organic solvent benzene, resulting in a decrease in the viscosity. The inflexible nature of butan-2-one becomes the major influencing force in the high-concentration region. The viscosity values reveal that the magnitude of the positive $\eta_{\rm exp}$ values decreases with increasing temperature [i.e., $(d\eta_{exp}/dT)_p$ is negative].

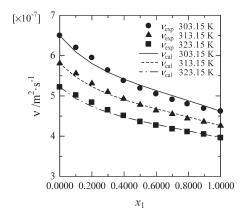


Figure 2. Kinematic viscosities for the butan-2-one (1) + benzene (2) system at T = (303.15, 313.15, and 323.15) K.

Table 3. Comparison of the Kinematic Viscosities Predicted Using the UNIFAC—VISCO Model and the Experimental Values

T/K	AAD/%	$ E_{\nu,i} _{\mathrm{max}}/\%$
303.15	1.21	2.4
313.15	0.64	1.7
323.15	0.70	2.1

In Figure 2, kinematic viscosities predicted using the UNIFAC-VISCO model are compared with the experimental values. Experimental kinematic viscosities ($\nu_{\rm exp}$) were calculated from the experimental viscosities $\eta_{\rm exp}$ (Table 2) and the densities $\rho_{\rm exp}$ obtained in our previous study²³ using the following equation:

$$\nu_{\rm exp}/{\rm m}^2 \cdot {\rm s}^{-1} = \frac{(\eta_{\rm exp}/{\rm Pa} \cdot {\rm s})}{(\rho_{\rm exp}/{\rm kg} \cdot {\rm m}^{-3})} \tag{1}$$

It should be noted that the unit for the viscosities was changed in the calculations. As can be seen from the figure, the kinematic viscosities obtained using the UNIFAC—VISCO model are in good agreement with the experimental ones. The relative errors $(E_{\nu,i})$ and the absolute average differences (AAD), values of which are shown in Table 3, were computed using the following equations:

$$E_{\nu,i} = \frac{\nu_{\exp,i} - \nu_{\text{cal},i}}{\nu_{\exp,i}} \times 100 \%$$
 (2)

$$AAD = \frac{1}{N_p} \sum_{i=1}^{N_p} |E_{\nu,i}|$$
 (3)

where i denotes the i-th experimental data value and $N_{\rm p}$ is the number of experimental data values at each temperature. At each of the temperatures studied, the maximum error was observed at around 0.2 mol fraction of butan-2-one but was only about 2.0 %. Therefore, the UNIFAC—VISCO model seems to have a sufficient predictability for the system over the temperature range studied.

■ CONCLUSIONS

Viscosities for binary mixtures of butan-2-one with benzene have been measured at several temperatures over the whole composition range. A systematic change with increasing temperature was observed for $\eta_{\rm exp}$. Interactions between the components in

binary mixtures have been used to interpret the observed viscometric behavior. Furthermore, the kinematic viscosities predicted using the UNIFAC—VISCO model are in good agreement with the experimental ones.

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■ REFERENCES

- (1) Rowlinson, J. S.; Swinton, F. L. Liquids and Liquid Mixtures; Butterworth Scientific: London, 1982.
- (2) Dominey, L. A. Current State of the Art on Lithium Battery Electrolytes. In *Lithium Batteries: New Materials, Developments, and Perspectives*; Pistoia, G., Ed.; Elsevier: Amsterdam, 1994; pp 137–165.
- (3) Sadek, P. C. *The HPLC Solvent Guide*, 2nd ed.; Wiley-Interscience: New York, 2002.
- (4) Reichardt, C., Solvents and Solvent Effects in Organic Chemistry, 3rd ed.; Wiley-VCH: Weinheim, Germany, 2003.
- (5) Joshi, S. S.; Aminabhavi, T. M.; Shukla, S. S. Densities and viscosities of binary liquid mixtures of anisole with methanol and benzene. *J. Chem. Eng. Data* **1990**, *35*, 187–189.
- (6) Lee, M. J.; Wei, M. C. Densities and viscosities of 2-butanone/dibutyl ether, 2-picoline/2-butanone, and 2-picoline/water mixtures. *J. Chem. Eng. Data* **1992**, *37*, 209–212.
- (7) Aminabhavi, T. M.; Patil, V. B.; Aralaguppi, M. I.; Phayde, H. T. S. Density, viscosity, and refractive index of the binary mixtures of cyclohexane with hexane, heptane, octane, nonane, and decane at (298.15, 303.15, and 308.15) K. J. Chem. Eng. Data 1996, 41, 521–525.
- (8) Fiege, C.; Joh, R.; Petri, M.; Gmehling, J. Solid—liquid equilibria for different heptanones with benzene, cyclohexane, and ethanol. *J. Chem. Eng. Data* **1996**, *41*, 1431–1433.
- (9) Lee, M. J.; Lin, T. K.; Pai, Y. H.; Lin, K. S. Density and viscosity for monoethanolamine + 1-propanol, + 1-hexanol, and + 1-octanol. *J. Chem. Eng. Data* **1997**, *42*, 854–857.
- (10) Tamura, K. Excess molar enthalpies and excess molar heat capacities of (2-butanone plus cyclohexane, or methylcyclohexane, or benzene, or toluene, or chlorobenzene, or cyclohexanone) at T = 298.15 K. *J. Chem. Thermodyn.* **2001**, *33*, 1345–1353.
- (11) Ouyang, G.; Huang, Z.; Ou, J.; Wu, W.; Kang, B. Excess molar volumes and surface tensions of xylene with 2-propanol or 2-methyl-2-propanol at 298.15 K. J. Chem. Eng. Data 2003, 48, 195–197.
- (12) Al-Jimaz, A. S.; Al-Kandary, J. A.; Abdul-Latif, A. H. M. Densities and viscosities for binary mixtures of phenetole with 1-pentanol, 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol, and 1-decanol at different temperatures. *Fluid Phase Equilib.* **2004**, 218, 247–260.
- (13) Cerdeirina, C. A.; Gonzalez-Salgado, D.; Romani, L.; Delgado, M. d. C.; Torres, L. A.; Costas, M. Towards an understanding of the heat capacity of liquids. A simple two-state model for molecular association. *J. Chem. Phys.* **2004**, *120*, 6648–6659.

- (14) Rathnam, M. V.; Mohite, S.; Kumar, M. S. S. Viscosity, density, and refractive index of some (ester plus hydrocarbon) binary mixtures at 303.15 K and 313.15 K. *J. Chem. Eng. Data* **2005**, *50*, 325–329.
- (15) Al-Kandary, J. A.; Al-Jimaz, A. S. Viscosities, densities, and speeds of sound of binary mixtures of benzene, toluene, o-xylene, m-xylene, p-xylene, and mesitylene with anisole at (288.15, 293.15, 298.15, and 303.15) K. J. Chem. Eng. Data 2007, 52, 664–664.
- (16) Clara, R. A.; Marigliano, A. C. G.; Solimo, H. N. Density, viscosity, refractive index, excess molar enthalpy, viscosity, and refractive index deviations for the (1-butanol + 2-butanone) binary system at $T=303\,$ K. A new adiabatic calorimeter for heat of mixing. *J. Chem. Thermodyn.* **2008**, 40, 292–297.
- (17) Song, C. Y.; Shen, H. Z.; Zhao, J. H.; Wang, L. C.; Wang, F. A. Densities and viscosities of binary mixtures of vitamin K₃ with benzene, toluene, ethylbenzene, *o*-xylene, *m*-xylene, and *p*-xylene from (303.15 to 333.15) K. *J. Chem. Eng. Data* **2008**, *5*3, 1110–1115.
- (18) Yang, J. H.; Dai, L. Y.; Wang, X. Z.; Chen, Y. Q. Densities and viscosities of binary mixtures of methyl 4-chlorobutyrate with aromatic hydrocarbons at *T* = (298.15 to 318.15) K. *J. Chem. Eng. Data* **2009**, 54, 2332–2337.
- (19) Kabir-ud-Din; Koya, P. A. Effects of solvent media and temperature on the self-aggregation of cationic dimeric surfactant 14–6–14, 2Br studied by conductometric and fluorescence techniques. *Langmuir* **2010**, *26*, 7905–7914.
- (20) Kabir-ud-Din; Koya, P. A. Micellar properties and related thermodynamic parameters of the 14–6–14, 2Br gemini surfactant in water + organic solvent mixed media. *J. Chem. Eng. Data* **2010**, 55, 1921–1929.
- (21) Chowdhury, M. A.; Majid, M. A.; Saleh, M. A. Volumetric and viscometric behaviour of binary systems: (1-hexanol + hydrocarbons). *J. Chem. Thermodyn.* **2001**, 33, 347–360.
- (22) Saleh, M. A.; Habibullah, M.; Ahmed, M. S.; Uddin, M. A.; Uddin, S. M. H.; Uddin, M. A.; Khan, F. M. Excess molar volumes and viscosities of some alkanols with cumene. *Phys. Chem. Liq.* **2006**, *44*, 31–43.
- (23) Adhikhari, A. B.; Rahman, I. M. M.; Uddin, M. A.; Hasegawa, H.; Majid, M. A. Volumetric behavior of the binary mixtures of methyl ethyl ketone with n-hexane, cyclohexane, and benzene at T = (303.15, 313.15, and 323.15) K. J. Chem. Eng. Data 2009, 54, 1138–1141.
- (24) Das, K. N.; Habibullah, M.; Rahman, I. M. M.; Hasegawa, H.; Uddin, M. A.; Saifuddin, K. Thermodynamic properties of the binary mixture of hexan-1-ol with m-xylene at T = (303.15, 313.15, and 323.15) K. J. Chem. Eng. Data 2009, 54, 3300–3302.
- (25) Habibullah, M.; Das, K. N.; Rahman, I. M. M.; Hasegawa, H.; Uddin, M. A.; Saifuddin, K. Volumetric and viscometric behavior of the binary system: (Hexan-1-ol + p-Xylene). *J. Chem. Eng. Data* **2010**, 55, 5311–5313.
- (26) Habibullah, M.; Das, K. N.; Rahman, I. M. M.; Uddin, M. A.; Saifuddin, K.; Iwakabe, K.; Hasegawa, H. Density and viscosity of the binary mixtures of hexan-1-ol with isomeric xylenes at T = (308.15 and 318.15) K and atmospheric pressure. *J. Chem. Eng. Data* **2010**, *55*, 5370–5374.
- (27) Smallwood, I. M. Handbook of Organic Solvent Properties; Halsted Press: New York, 1996.
- (28) Chevalier, J. L.; Petrino, P.; Gaston-Bonhomme, Y. Estimation method for the kinematic viscosity of a liquid-phase mixture. *Chem. Eng. Sci.* 1988, 43, 1303–1309.
- (29) Gaston-Bonhomme, Y.; Petrino, P.; Chevalier, J. L. UNIFAC—VISCO group contribution method for predicting kinematic viscosity: Extension and temperature dependence. *Chem. Eng. Sci.* **1994**, *49*, 1799–1806.
- (30) Donald, M. B.; Ridgway, K. The binary systems benzene—ethyl methyl ketone and benzene—cyclohexane. *J. Appl. Chem.* **1958**, 8, 403–407
- (31) Teller, A. J.; Walsh, T. J. Viscosities of benzene—methyl ethyl ketone systems. *J. Chem. Eng. Data* **1959**, *4*, 279–283.
- (32) Mussche, M. J.; Verhoeye, L. A. Viscosity of ten binary and one ternary mixtures. *J. Chem. Eng. Data* 1975, 20, 46–50.

- (33) Yadava, R. R.; Yadava, S. S. Viscometric study of molecular interactions in binary liquid mixtures of polar and nonpolar solvents. *Indian J. Chem., Sect. A* **1981**, 20, 221–224.
- (34) Ratkovics, F.; Palágyi-Fényes, B. The effect of keto—enol tautomerism on the properties of methyl ethyl ketone + benzene mixtures. Fluid Phase Equilib. 1984, 16, 99–116.
- (35) Schrodt, J. T.; Akel, R. M. Binary liquid viscosities and their estimation from classical solution thermodynamics. *J. Chem. Eng. Data* **1989**, *34*, 8–13.
- (36) Petrino, P. J.; Gaston-Bonhomme, Y. H.; Chevalier, J. L. E. Viscosity and density of binary liquid mixtures of hydrocarbons, esters, ketones, and normal chloroalkanes. *J. Chem. Eng. Data* **1995**, *40*, 136–140.
- (37) Jayalakshmi, T.; Reddy, K. Viscosities of binary liquid mixtures of polar—apolar and polar—polar systems at 303.15 K. *J. Chem. Sci.* **1985**, *94*, 457–460.
- (38) Kauzmann, W.; Eyring, H. The viscous flow of large molecules. *J. Am. Chem. Soc.* **1940**, *62*, 3113–3125.
- (39) Fort, R. J.; Moore, W. R. Viscosities of binary liquid mixtures. *Trans. Faraday Soc.* **1966**, 62, 1112–1119.
- (40) Riddick, J. A.; Bunger, W. B.; Sakano, T. Organic Solvents: Physical Properties and Methods of Purification, 4th ed.; Wiley-Interscience: New York, 1986.
- (41) TRC Databases for Chemistry and Engineering—TRC Thermodynamic Tables; Texas Engineering Experiment Station, Texas A & M University System: College Station, TX, 1998.
- (42) Martínez, S.; Garriga, R.; Pérez, P.; Gracia, M. Densities and viscosities of binary mixtures of butanone with butanol isomers at several temperatures. *Fluid Phase Equilib.* **2000**, *168*, 267–279.
- (43) Oswal, S.; Rathnam, M. V. Viscosity data of binary mixtures: ethyl acetate + cyclohexane, + benzene, + toluene, + ethylbenzene, + carbon tetrachloride, and + chloroform at 303.15 K. *Can. J. Chem.* **1984**, 62, 2851–2853.