

Measurement of Selected Thermophysical and Viscosity of Binary Mixtures of Nitromethane with 2-Propanol, 2-Butanol and 2-Pentanol

M. Almasi*

Department of Chemistry, Faculty of Sciences, Ahvaz Branch, Islamic Azad University, Ahvaz, Iran

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The density and viscosity values at $T = (293.15-323.15)$ K for binary mixtures of nitromethane with 2-propanol, 2-butanol and 2-pentanol were measured over the entire composition range. From the experimental data, excess molar volumes, thermal expansion coefficients, excess thermal expansions, partial molar volumes at infinite dilution and deviations in viscosity were calculated. The results were discussed in terms of intermolecular interactions and structure of the studied binary mixtures. Free-volume theory coupled with the Peng-Robinson-Stryjek-Vera equation of state was employed to calculate the viscosities of pure compounds and the binary liquid mixtures. The Agreement between experimental data and theoretical values was satisfactory.

Keywords: Nitromethane, Density, Viscosity, Intermolecular Interactions, Free volume theory

INTRODUCTION

Studying the excess thermodynamic properties is quite important to understand molecular interactions in the mixtures and to develop and test theories of solutions and mathematical models. Nitromethane is an aprotic solvent with high polarity which is commonly used as a solvent in a variety of industrial applications such as extractions, reaction mediums, cleaning solvents, and intermediate inorganic synthesis. It is used widely in the manufacture of pharmaceuticals, pesticides, explosives, fibers, and coatings. Moreover nitromethane is used as a fuel in various internal combustion engines in radio control, control line and free flight model aircrafts [1]. In the mixing process of alkanols with varying chain lengths, interesting behaviors may be observed due to specific interactions, hydrogen bond and hydrophobic effects. Thus, accurate knowledge of their thermodynamic mixing properties has great relevance in theoretical and applied areas of research. The aim of the present work is to continue our previous studies [2,3] on the

binary mixtures containing 2-alknol and different groups of organic compounds. In this work, densities and viscosities over the whole range of compositions, at atmospheric pressure, and in the temperature range (293.15-323.15) K are measured. On the basis of experimental data, excess molar volumes, viscosity deviations, thermal expansion coefficients, excess thermal expansions and partial molar volumes at infinite dilutions for these mixtures are obtained. These properties are studied to characterize the behavior of the corresponding systems and discuss the possible interactions between the components of the mixtures. In addition, we compared the densities and viscosities of the studied mixtures with the same literature data. Agreement between two sets of data was satisfactory [4-6]. The present work also deals with the application of free volume theory to describe the viscosity of fluids. The importance of free-volume theories for the calculation of viscosities owes to the following facts: (i) their equations are simple and involve only few parameters; (ii) the parameters have, almost always, a physical meaning; (iii) they can be used over a wide range of temperature and pressure; (iv) the theories are based on statistical mechanics, therefore, giving them a theoretical background and, (v) they may easily be

*Corresponding author. E-mail: almasi.mohammad@gmail.com

extended to the multicomponent systems [7].

EXPERIMENTAL

The following materials in the liquid state with the mass fraction purities greater than 99% were applied: nitromethane, 2-propanol, 2-butanol and 2-pentanol. All solvents were purchased from Merck and used without further purifications. Table 1 gives the densities and viscosities at $T = 298.15$ K along with the data from the literature [2,4,8].

The densities and viscosities were measured with a fully automated SVM 3000 Anton-Paar Stabinger Viscometer. The viscometer is based on a modified Couette principle with a rapidly rotating outer tube and an inner measuring bob which rotates more slowly. The uncertainty is 1×10^{-4} g cm⁻³ for density measurements and 1% for viscosity measurements. The mixtures were prepared by mass and were kept in special airtight stopper glass bottles to avoid evaporation. The weightings were measured accurately using a Mettler electronic balance with 0.01 mg readability. The uncertainty in the mole fraction was estimated to be less than 1×10^{-4} .

RESULTS AND DISCUSSION

Experimental densities and viscosities for pure materials and the binary mixtures are presented in Table 2. The densities of the binary mixtures decrease with increasing

temperature and increase with increasing carbon chain length. Values of viscosity increase with the increase of carbon chain length and decrease with the rise of temperature. Comparisons of our data and reported values in the literature for similar binary mixtures are shown in supplementary.

Densities and Excess Molar Volumes

On the basis of the experimental values of densities, the excess molar volumes, V_m^E at different temperatures were calculated by the following equation

$$V_m^E = \sum_{i=1}^N x_i M_i (\rho^{-1} - \rho_i^{-1}) \quad (1)$$

where x_i is the mole fraction, M_i is the molar mass of component i , ρ is the density of the mixture, ρ_i is the density of pure component i , and N stands for the number of components in the mixture. Excess molar volumes were fitted to the Redlich-Kister [9] equation:

$$V_m^E = x_1(1-x_1) \sum_{k=0}^N A_k (1-2x_1)^k \quad (2)$$

where x_1 is the mole fraction of nitromethane and A_k is polynomial coefficient. Standard deviations were obtained from

$$\sigma = \left[\sum (V_{\text{exp}}^E - V_{\text{cal}}^E)^2 / (n-p) \right]^{1/2} \quad (3)$$

Table 1. Densities ρ , and Viscosities η , of Pure Liquid Components at $T = 298.15$ K and $P = 0.1$ MPa

Chemical name	Purity	Supplier	ρ		η	
			(g cm ⁻³)		(mPa s)	
				Lit.		Lit.
Nitromethane	>0.99	Merck	1.1303	1.1309 ^[4]	0.62	0.615 ^[4]
2-Propanol	0.997	Merck	0.7811	0.7812 ^[4]	2.08	2.105 ^[8]
2-Butanol	0.995	Merck	0.8027	0.8025 ^[4]	3.04	3.114 ^[8]
2-Pentanol	0.995	Merck	0.8053	0.80524 ^[2]	3.44	3.426 ^[8]

Table 2. Densities, ρ and Viscosities, η for the Liquid Binary Mixtures as a Function of the Mole Fraction x_1 of Nitromethane

Nitromethane + 2-propanol							
ρ (g cm ⁻³)							
x_1	T/K=293.15	T/K=298.15	T/K=303.15	T/K=308.15	T/K=313.15	T/K=318.15	T/K=323.15
0	0.7854	0.7811	0.7768	0.7724	0.7680	0.7634	0.7588
0.0815	0.8056	0.8011	0.7966	0.7919	0.7873	0.7825	0.7777
0.1607	0.8264	0.8216	0.8168	0.8119	0.8071	0.802	0.7969
0.2402	0.8483	0.8432	0.8383	0.8330	0.828	0.8228	0.8174
0.3501	0.8805	0.8752	0.8699	0.8643	0.859	0.8535	0.8479
0.4403	0.909	0.9033	0.8979	0.8919	0.8864	0.8806	0.8748
0.5604	0.9498	0.9438	0.9380	0.9317	0.9259	0.9198	0.9137
0.6500	0.9828	0.9765	0.9705	0.9640	0.9579	0.9517	0.9453
0.7400	1.0183	1.0118	1.0056	0.9989	0.9927	0.9862	0.9797
0.8500	1.0654	1.0587	1.0523	1.0454	1.0389	1.0322	1.0255
0.9402	1.1074	1.1007	1.0941	1.0871	1.0802	1.0734	1.0666
1	1.1372	1.1303	1.1236	1.1167	1.1098	1.1029	1.0961
η (mPa s)							
0	2.42	2.08	1.80	1.56	1.36	1.19	1.05
0.0815	1.80	1.59	1.39	1.22	1.08	0.96	0.86
0.1607	1.46	1.29	1.14	1.02	0.91	0.82	0.74
0.2402	1.25	1.11	0.98	0.88	0.8	0.72	0.65
0.3501	1.04	0.93	0.84	0.76	0.69	0.63	0.57
0.4403	0.92	0.82	0.75	0.69	0.63	0.57	0.52
0.5604	0.80	0.73	0.67	0.62	0.57	0.53	0.49
0.6500	0.72	0.67	0.63	0.58	0.54	0.52	0.47
0.7400	0.68	0.64	0.62	0.57	0.53	0.51	0.47
0.8500	0.67	0.63	0.61	0.56	0.53	0.50	0.47
0.9402	0.66	0.61	0.60	0.55	0.52	0.50	0.48
1	0.65	0.62	0.59	0.56	0.53	0.51	0.49

Table 2. Continued

Nitromethane + 2-butanol							
ρ (g cm ⁻³)							
x_1	T/K=293.15	T/K=298.15	T/K=303.15	T/K=308.15	T/K=313.15	T/K=318.15	T/K=323.15
0	0.8067	0.8027	0.7984	0.7941	0.7898	0.7852	0.7806
0.0811	0.8220	0.8178	0.8133	0.8088	0.8044	0.7995	0.7947
0.1594	0.8379	0.8334	0.8287	0.8240	0.8194	0.8144	0.8095
0.2401	0.8556	0.8508	0.8460	0.8411	0.8365	0.831	0.8260
0.3505	0.8824	0.8773	0.8721	0.8669	0.8621	0.8563	0.8510
0.4400	0.9065	0.9012	0.8958	0.8903	0.8853	0.8793	0.8737
0.5600	0.9430	0.9372	0.9315	0.9258	0.9205	0.9141	0.9083
0.6500	0.9740	0.9680	0.9621	0.9561	0.9506	0.9440	0.9379
0.7400	1.0087	1.0025	0.9963	0.9902	0.9844	0.9775	0.9712
0.8500	1.0572	1.0506	1.0444	1.0380	1.0318	1.0247	1.0181
0.9398	1.1028	1.0960	1.0895	1.0828	1.0762	1.0691	1.0626
1	1.1372	1.1303	1.1236	1.1167	1.1098	1.1029	1.0961
η (mPa s)							
0	3.67	3.04	2.54	2.13	1.80	1.54	1.33
0.0811	2.49	2.11	1.81	1.56	1.36	1.19	1.05
0.1594	1.95	1.66	1.46	1.28	1.13	1.01	0.89
0.2401	1.62	1.41	1.24	1.10	0.98	0.88	0.79
0.3505	1.32	1.17	1.03	0.92	0.84	0.76	0.69
0.4400	1.13	1.01	0.90	0.81	0.74	0.68	0.62
0.5600	0.95	0.86	0.77	0.70	0.65	0.60	0.56
0.6500	0.84	0.77	0.70	0.65	0.60	0.56	0.52
0.7400	0.76	0.71	0.65	0.61	0.57	0.53	0.51
0.8500	0.70	0.66	0.61	0.57	0.54	0.52	0.50
0.9398	0.67	0.63	0.60	0.55	0.53	0.50	0.49
1	0.65	0.62	0.59	0.56	0.53	0.51	0.49

Table 2. Continued

Nitromethane + 2-pentanol							
ρ (g cm ⁻³)							
x_1	T/K=293.15	T/K=298.15	T/K=303.15	T/K=308.15	T/K=313.15	T/K=318.15	T/K=323.15
0	0.8093	0.8053	0.8012	0.7970	0.7927	0.7884	0.7840
0.0810	0.8220	0.8177	0.8134	0.8091	0.8046	0.8001	0.7956
0.1599	0.8354	0.8309	0.8264	0.8219	0.8174	0.8127	0.8080
0.2402	0.8506	0.8458	0.8411	0.8365	0.8318	0.8270	0.822
0.3501	0.8741	0.8691	0.8641	0.8591	0.8542	0.8492	0.8439
0.4399	0.8961	0.8908	0.8859	0.8810	0.8757	0.8701	0.8646
0.5600	0.9303	0.9247	0.9192	0.9137	0.9082	0.9027	0.8970
0.6500	0.9605	0.9546	0.9489	0.9432	0.9374	0.9316	0.9257
0.7399	0.9955	0.9894	0.9833	0.9775	0.9714	0.9654	0.9593
0.8500	1.0467	1.0404	1.0341	1.0277	1.0214	1.0149	1.0084
0.9400	1.0976	1.0910	1.0846	1.0779	1.0712	1.0645	1.0578
1	1.1372	1.1303	1.1236	1.1167	1.1098	1.1029	1.0961
η (mPa s)							
0	3.97	3.28	2.72	2.28	1.93	1.65	1.42
0.0810	2.56	2.21	1.90	1.66	1.42	1.23	1.09
0.1599	1.97	1.73	1.52	1.31	1.1	0.98	0.88
0.2402	1.63	1.45	1.27	1.07	0.92	0.82	0.74
0.3501	1.30	1.18	1.02	0.89	0.78	0.71	0.63
0.4399	1.09	1.00	0.88	0.79	0.70	0.64	0.58
0.5600	0.88	0.81	0.74	0.67	0.62	0.58	0.53
0.6500	0.76	0.70	0.66	0.62	0.58	0.54	0.51
0.7399	0.67	0.63	0.60	0.58	0.54	0.52	0.49
0.8500	0.62	0.59	0.58	0.56	0.52	0.50	0.47
0.9400	0.61	0.58	0.57	0.55	0.52	0.49	0.47
1	0.65	0.62	0.59	0.56	0.53	0.51	0.49

Where V_{exp}^E and V_{cal}^E are the experimental and calculated data, respectively. Values of A_k and standard deviations are presented in supplementary. For clarity reasons, only the variations of excess molar volumes as a function of the mole fraction at $T = 298.15$ K for all the studied mixtures are presented in Fig. 1. For all the studied systems, excess molar volumes are positive and increase as temperature increases. In the present liquids, molecules of 2-alkanols are self-associated through intermolecular hydrogen bonding (hydrogen is attached directly to an electronegative element (oxygen)), causing hydrogen to acquire a significant positive charge. Oxygen is not only significantly negative, but also has at least one "active" lone pair with a high density of negative charge. Therefore, some interactions occur between the hydrogen of OH group of one alcohol and the oxygen of the hydroxyl group of another molecule, leading to the formation of hydrogen bonds. This phenomenon is called self-association. Nitromethane has special properties such as dipolar aprotic solvent, large dipole moment and weak or probably negligible H-bonds.

In the mixing of 2-alkanols with nitromethane different resultant contributions to the volumes of the mixtures can be expected. The disruption of the associated structure of the alcohols through the breaking of hydrogen bonds and steric hindrance effects will result in an increase in volume. On the other hand, a decrease in volume would result from dipole-dipole and specific interactions between the unlike molecules. It is clear that the prevailing factors are steric hindrance and bond-breaking effects for mentioned mixtures. In the higher chain length alkanols, the interaction contributions are negligible because of chain length hindrance. The magnitude of positive V_m^E values follows the sequence: 2- propanol < 2-butanol < 2-pentanol. Increase of excess molar volumes with temperature can be expected since hydrogen bonds are more easily broken at higher temperatures. Influence of temperature on the excess molar volumes is presented graphically in Fig. 2.

Partial molar property at infinite dilution $\bar{V}_{m,i}^0$ is of interest since at the limit of infinite dilution the solute-solute interactions disappear and the only interactions present are solute-solvent interactions. So, partial molar volume at infinite dilution for the studied mixtures was calculated by the well-known relations [10]. The calculated partial molar

volumes at infinite dilution for binary mixture nitromethane + 2-pentanol are presented in Supplementary. According to the results, partial molar volumes at infinite dilution for nitromethane and 2-alkanols are slightly greater than the corresponding molar volumes of the pure materials. This could indicate that interaction between nitromethane and alcohols is not very favorable. $\bar{V}_{m,i}^0$ increases with chain-length of alkanols and slightly increases with temperature.

Expansivity coefficients are related to the interaction volume of liquids and mixtures. The excess amount may reflect the molecular orientation and packing of the mixtures. To discuss more about the interactions and effects governing the mixtures, thermal expansion coefficient and excess thermal expansion for the studied systems were calculated using

$$\alpha^E = \alpha_p - \phi_1\alpha_1 - \phi_2\alpha_2 \quad (4)$$

where α_p is the thermal expansion coefficient of the mixture and α_i is the thermal expansion of pure state. The volume fraction ϕ_i is defined by

$$\phi_i = \frac{x_i V_i^*}{\sum_{i=1}^2 x_i V_i^*} \quad (5)$$

where V_i^* is the molar volume of pure states. Calculated values of α_p are increased with the increase of temperature. The main microscopic reason for this phenomenon is hydrogen bond rupture. As the temperature rises, rupture of hydrogen bonds becomes more important. Values of α_p at various temperature increase with the increase of nitromethane concentration in the mixture. These results are explained in terms of packing effects. Packing effects take place as a consequence of the accommodation of nitromethane molecules in the hydrogen-bonded structure of the alcohol. This phenomenon only can appear at a high enough mole fraction of alcohol, since there must be enough alcohol molecules to create this hydrogen-bonded structure. So, the Packing effects take place at high concentration of alcohols. This effect is graphically shown in Fig. 3. α^E

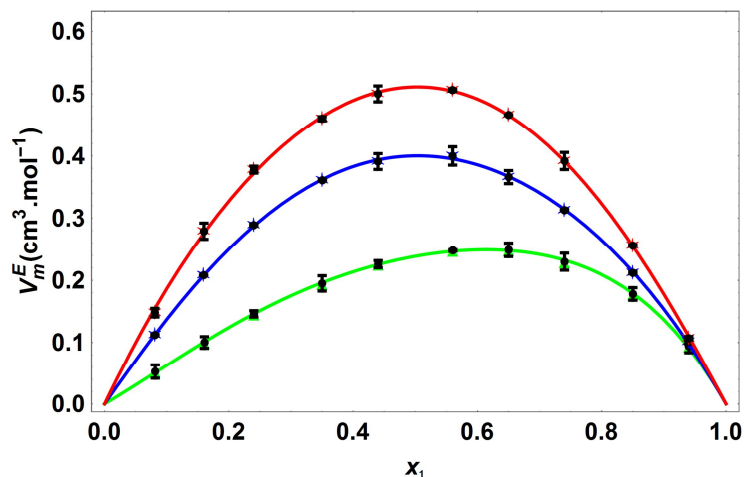


Fig. 1. Excess molar volumes V_m^E vs. mole fraction of nitromethane for binary mixtures of nitromethane with (from below to up) 2-propanol, 2-butanol, 2-pentanol, respectively at 298.15 K. The solid curves follow the Redlich-Kister equation. The symbols are measured data.

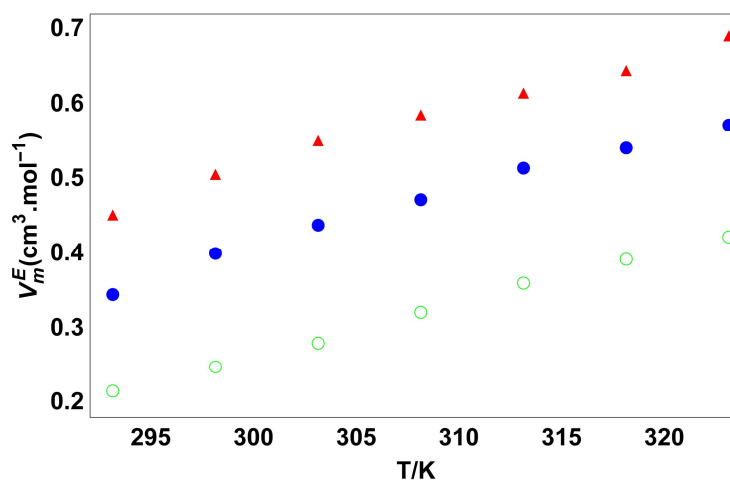


Fig. 2. Excess molar volumes at equimolar concentration ($x \approx 0.56$) for binary mixtures of nitromethane + 2-propanol (\circ), 2-butanol (\bullet), 2-pentanol (\blacktriangle) at various temperatures.

values for binary mixtures of nitromethane + 2-pentanol are shown in Fig. 4. Positive values correspond to the breakdown of the molecular association or complex in the mixtures. The increase of α^E values for binary mixtures with increasing temperature indicates more destruction of

order during the mixing process. The calculated values are in agreement with our excess molar volumes.

Dynamic Viscosities

The viscosity deviation was calculated by

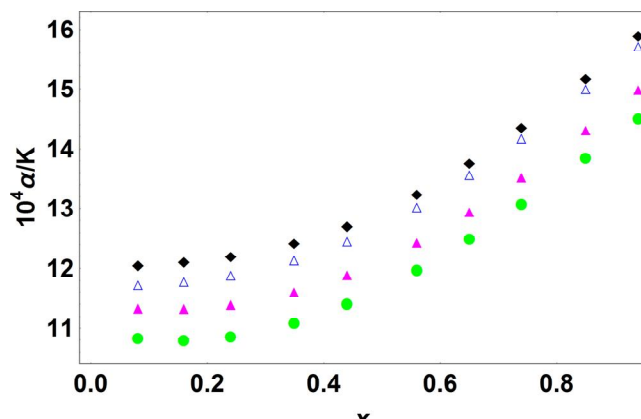


Fig. 3. Experimental thermal expansion coefficients (α_p) of nitromethane + 2-pentanol at temperatures (●) 298.15 K, (▲) 303.15 K, (Δ) 308.15 K, (◆) 313.15 K.

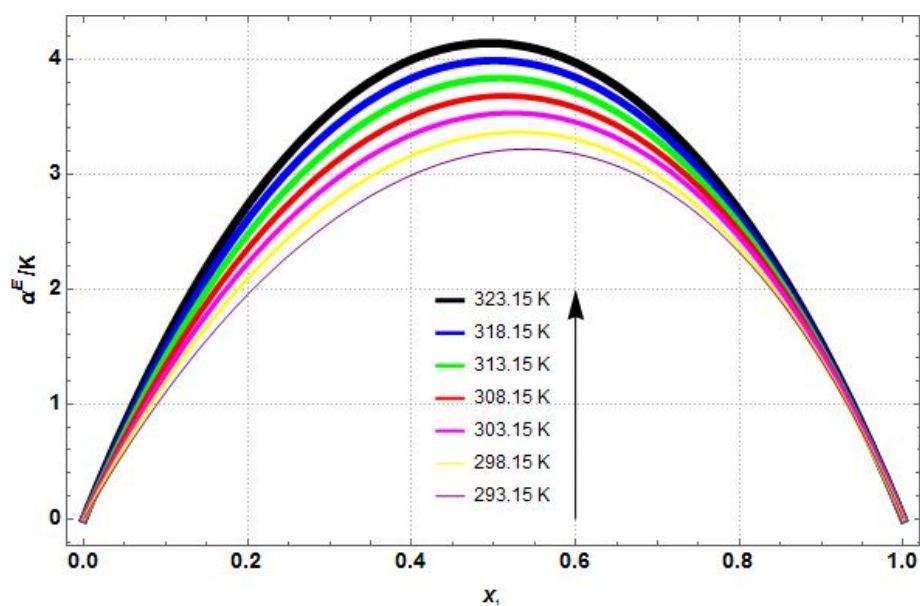


Fig. 4. Excess thermal expansion for binary mixtures of nitromethane + 2-pentanol at various temperatures.

$$\Delta\eta = \eta - x_1\eta_1 - x_2\eta_2 \quad (6)$$

where η is the mixture viscosity, η_1 and η_2 are the viscosities of pure components. The values of viscosity deviations were correlated by the Redlich-Kister equation and adjustable coefficients are reported in supplementary.

Figure 5 indicates the viscosity deviations for the mixtures of nitromethane with 2-alkanol at $T = 298.15$ K. It is clear that the viscosity deviations are negative throughout the whole concentration range. The negative values of viscosity deviations for the binary mixtures investigated suggest that the viscosities of associates formed between unlike

molecules are relatively less than those of the pure components. Deviation values become more negative as the alkyl chain length increases and temperature decreases.

Free Volume Theory

In the free volume theory [11-13], the viscosity is expressed as the sum of two terms

$$\eta = \eta_0 + \Delta\eta^{res} \quad (7)$$

η_0 is the viscosity in the dilute gas limit, while the residual viscosity $\Delta\eta^{res}$ describes deviations from the dilute gas. η_0 was calculated as

$$\eta_0 = 40.785 \frac{\sqrt{M_w T}}{V_c^{2/3} \Omega^*} F_c \quad (8)$$

Ω^* is the reduced collision integral and F_c is given by

$$F_c = 1 - 0.275\omega + 0.059035\mu_r^4 + \chi \quad (9)$$

where ω is the acentric factor, μ_r is dimensionless dipole moment and χ is a correction for the influence of hydrogen bonds. The residual viscosity is described by a generalized dumbbell model

$$\Delta\eta^{res} = \frac{\rho N_a \zeta L^2}{M} \quad (10)$$

where ρ is the density, ζ is the friction coefficient, and N_a is Avogadro's number and L^2 is an average quadratic length related to the size of the molecule. Moreover, the free-volume fraction is calculated by

$$f_v = \left(\frac{RT}{E} \right)^{3/2} \quad (11)$$

It has been assumed that $E = E_0 + pM/\rho$ where E_0 is related to the energy barrier that the molecule has to exceed in order to diffuse. On the other hand, the residual viscosity is depend on the free volume fraction through the exponential relation

$$\Delta\eta^{res} = A \exp\left(\frac{B}{f_v}\right) \quad (12)$$

where B is the characteristic of the free-volume overlap. ζ can be written as

$$\zeta = \zeta_0 \exp\left(\frac{B}{f_v}\right) \quad (13)$$

ζ_0 is a friction coefficient related the mobility of the molecule. By combining Eqs. (10) and (13)

$$\Delta\eta^{res} = \frac{\rho N_a L^2 \zeta_0 \exp\left(\frac{B}{f_v}\right)}{M} \quad (14)$$

The friction coefficient ζ_0 is related to the energy of dissipation E by the expression [15]

$$\zeta_0 = \frac{E}{N_a b_f} \left(\frac{M}{3RT} \right)^{0.5} \quad (15)$$

where b_f is the dissipation length of the energy E . By inserting Eqs. (14) and (15) to Eq. (10)

$$\Delta\eta^{res} = \rho l \left(\frac{E_0 + (PM/\rho)}{\sqrt{3RTM}} \right) \exp \left[B \left(\frac{E_0 + (PM/\rho)}{RT} \right)^{1.5} \right] \quad (16)$$

Where $l = L^2 / b_f$. The final equation includes three adjustable parameters related to the structural and energetic properties of the fluid: l , E_0 and B . The theory can be applied to the mixtures by employing mixing rules for the three parameters. We applied a linear compositional mixing rule of the Lorentz type for the three parameters of the dense fluid term:

$$E_{0,mix} = \sum_{i=1}^n E_{0,i} x_i \quad (17)$$

$$B_{mix} = \sum_{i=1}^n B_i x_i \quad (18)$$

$$l_{mix} = \sum_{i=1}^n l_i x_i \quad (19)$$

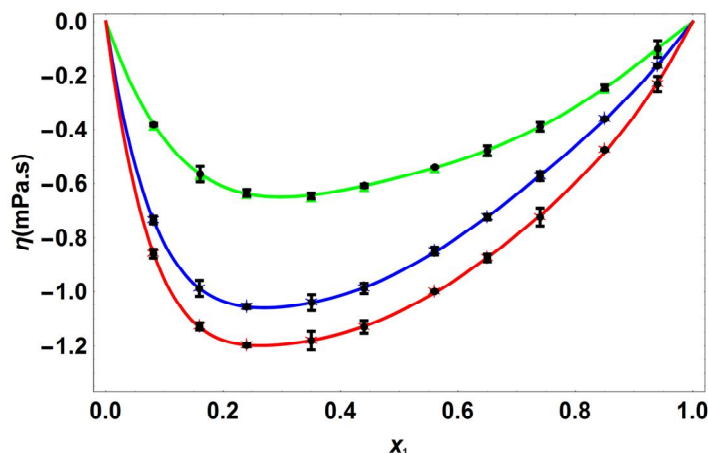


Fig. 5. Viscosity deviations $\Delta\eta$ vs. mole fraction of nitromethane for binary mixtures of nitromethane with (from up to below) 2-propanol, 2-butanol, 2-pentanol at 298.15 K. The solid curves follow the Redlich-Kister equation. The symbols are measured data.

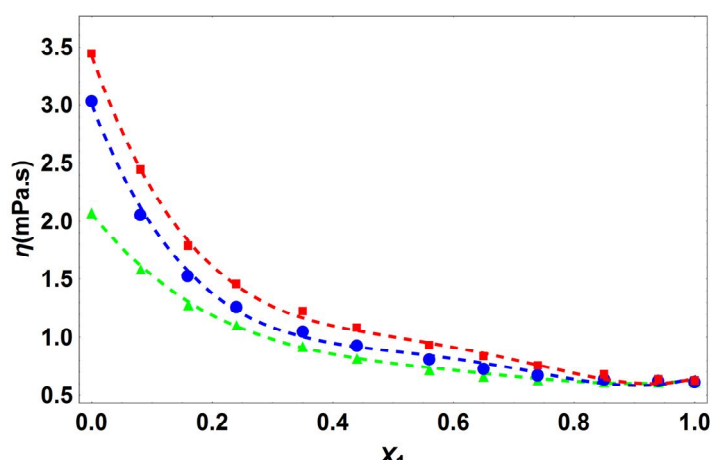


Fig. 6. Experimental and calculated viscosities of nitromethane with (\blacktriangle) 2-propanol, (\bullet) 2-butanol, (\blacksquare) 2-pentanol at 298.15 K. The solid curves follow the free volume theory. The symbols are measured data.

Here, it should be noted that no binary adjustable parameters are used for the viscosity of mixtures. Three viscosity parameters were fitted to viscosity data of the pure fluid at several temperatures, whereas the densities were obtained from Peng-Robinson-Stryjek-Vera (PRSV) equations of state. Adjustable parameters of the free volume

theory associated with the PRSV equation are presented in Table 3 along with AAD. Comparison between the calculated viscosities and experimental data are shown in Fig. 6. The agreement between the experimental and calculated values is good.

Table 3. Optimized Parameters of the Free-Volume Theory Along with Average Absolute Deviation (AAD%)

Compound	$\alpha \times 10^3$	B $\times 10$	L _v	ADD (%)
Nitromethane	27.50	1.60	0.724	3.81
2-Propanol	390.27	3.09	0.043	2.98
2-Butanol	334.6	2.96	0.034	2.06
2-Pentanol	243.91	2.51	0.021	1.43

CONCLUSIONS

This study reports the densities and viscosities for binary mixtures of nitromethane + 2-alkanols. From the obtained data, excess molar volumes, thermal expansion coefficients, excess thermal expansions, partial molar volumes at infinite dilution and viscosity deviations were calculated. Excess molar volumes are positive and viscosity deviations are negative over the full range of composition for all studied mixtures. Analyses suggest that there is no strong interaction due to disruption of associated structures of alcohols through the breaking of hydrogen bonds and steric hindrance effects. Free volume theory coupled with Peng-Robinson-Stryjek-Vera equation of state has been used for modeling the viscosities of pure state and binary mixtures. Good agreement with available experimental data and calculated values was found in all cases and average absolute deviation was within the experimental uncertainties.

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