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Experimental and Predicted Viscosities of the Ternary Mixture (Hexane + 1,3-Dioxolane + 2-Butanol) at 298.15 and 313.15 K

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Viscosities of the ternary mixture (hexane + 1,3-dioxolane + 2-butanol) have been measured at atmospheric pressure at 298.15 and 313.15 K. Viscosity deviations for the ternary mixture were calculated from experimental data and fitted by the Cibulka equation. To correlate the experimental data of the ternary system, extended Nissan−Grunberg and McAllister equations have been used. The group contribution method proposed by Wu has been used to predict the viscosity of the mixture.

1. Introduction

Experimental viscosity data and methods for the estimation of viscosities of multicomponent mixtures are of great theoretical and practical interest. However, the literature of the correlation and prediction of flow properties for ternary and multicomponent liquid mixtures is rather limited. Recently, some empirical and semiempirical equations for binary mixtures were extended to ternary mixtures by introducing a ternary parameter.[1] Some models have also been developed for the prediction of viscosities of those mixtures, and most of them are based on the group contribution concept.[2,3]

In the last few years, we have reported the study and prediction of transport properties of ternary mixtures[4−7] containing an alkane (cyclohexane or hexane), a cyclic diether (1,3-dioxolane or 1,4-dioxane), and one isomer of butanol (1-butanol or 2-butanol). Continuing this systematic study, we present here experimental viscosity measurements of the ternary mixture hexane + 1,3-dioxolane + 2-butanol at atmospheric pressure at temperatures of 298.15 and 313.15 K. The viscosity data have been used to calculate the viscosity deviations and have then been fit by the Cibulka equation for ternary mixtures.[8] The correlation equations of Nissan−Grunberg[9] and McAllister[10] extended to multicomponent mixtures[1] were applied to the viscosities of the ternary mixture using parameters obtained from the correlation of the constituent binary systems. The group contribution method proposed by Wu,[2] which uses a modified kinetic equation of Eyring[11] and the group contribution method UNIFAC,[12] has been used to predict the viscosity of the ternary system.

2. Experimental Section

The compounds used, hexane (purity better than 99%), 1,3-dioxolane (purity better than 99%), and 2-butanol (purity better than 99%), were obtained from Aldrich. The purities of these compounds were checked by comparing the measured densities with those reported in the literature and also by a chromatographic method using a semicapillary methyl silicone column (o.d. 530 μm) and a flame ionization detector, confirming the absence of other significant compounds, so no further purification was attempted. The 2-butanol was dried with activated molecular sieves (type 0.3 nm from Merck). The pure compound properties, along with literature values at 298.15 K, are given in Table 1.

Kinematic viscosities, ν, of the pure compounds and the ternary mixtures were determined using an Ubbelohde viscosimeter (inner diameter = 0.63 mm, capillary length = 893 mm), with a Schott-Gërate automatic measuring unit model AVS-440, for which the reproducibility of the flow-time measurement is ±0.01 s and the corresponding uncertainty in the kinematic viscosity is $\pm 1 \times 10^{-4}$ $\text{mm}^2\cdot\text{s}^{-1}$. At least four time-flow measurements were performed for each composition and temperature, and the results were averaged. Kinetic energy corrections were applied to the experimental data.

A Schott-Gërate thermostat was used to keep the temperature within ±0.01 K. Densities, ρ, required to calculate absolute viscosities, $\eta = \rho\nu$, were measured with an Anton Paar DMA-58 vibrating tube densimeter. The uncertainty of the density measurements was $\pm 5 \times 10^{-3}$ $\text{kg}\cdot\text{m}^{-3}$, and the uncertainty of calculated absolute viscosities was $\pm 1 \times 10^{-4}$ mPa·s.

The calibration of the viscosimeter was carried out with deionized doubly distilled water, and the value used (η = 0.8902 mPa·s at 298.15 K) was taken from Marsh.[13]

Mixtures were prepared by mass using a Mettler H20T balance. The uncertainty in the mole fraction of the mixtures is estimated to be less than $\pm 1 \times 10^{-4}$.

3. Results and Discussion

The experimental viscosities of the mixture hexane + 1,3-dioxolane + 2-butanol at 298.15 and 313.15 K are shown in Table 2, along with viscosity deviations, $\Delta\eta$, that were determined using the equation

$$\Delta\eta = \eta - \sum_{i=1}^{3} x_i \eta_i \tag{1}$$

where η is the absolute viscosity of the mixture, x_i is the mole fraction of component i, and η_i is the absolute viscosity of pure component i.

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Table 1. Densities, ρ, and Viscosities, η, of Pure Components at 298.15 K and 313.15 K and Comparison with Literature Data

	$T = 298.15$ K				$T = 313.15$ K			
	ρ/kg·m^{-3}		η/ mPa·s		ρ/kg·m^{-3}		η/mPa·s	
component	exptl	lit	exptl	lit	exptl	lit	exptl	lit
hexane	655.07	654.84[a]	0.2951	0.294[a]	641.27		0.2552	0.264[a]
1,3-dioxolane	1058.62	1058.66[b]	0.5886		1039.97		0.4985	-
2-butanol	802.20	802.41	3.0427	2.998[a]	789.59	789.7[c]	1.7913	1.7833[c]

[a] Reference 15. [b] Reference 16. [c] Reference 17.

Table 2. Experimental Kinematic Viscosities, ν, Densities, ρ, Calculated Absolute Viscosities, η, and Viscosity Deviations, $\Delta\eta$, of the Ternary Mixture Hexane (1) + 1,3-Dioxolane (2) + 2-Butanol (3)

		$T = 298.15$ K				$T = 313.15$ K			
		ν	ρ	η	$\Delta\eta$	ν	ρ	η	$\Delta\eta$
x_1	x_2	mm^2·s^{-1}	g·cm^{-3}	mPa·s	mPa·s	mm^2·s^{-1}	g·cm^{-3}	mPa·s	mPa·s
0.0510	0.0490	2.3805	0.79989	1.9041	−0.8782	1.5750	0.78674	1.2391	−0.4105
0.0491	0.0985	1.9570	0.80916	1.5835	−1.0826	1.3580	0.79562	1.0805	−0.5080
0.0481	0.8526	0.5743	0.99153	0.5694	−0.2487	0.4929	0.97380	0.4800	−0.1351
0.0500	0.9015	0.5698	1.00663	0.5736	−0.1194	0.4928	0.98858	0.4872	−0.0618
0.1003	0.0510	1.9110	0.79004	1.5098	−1.1322	1.3210	0.77678	1.0261	−0.5451
0.0992	0.0969	1.6120	0.79824	1.2868	−1.2456	1.1610	0.78463	0.9110	−0.6027
0.1013	0.1993	1.2020	0.81650	0.9814	−1.2939	0.9262	0.80225	0.7430	−0.6350
0.1002	0.2920	1.0360	0.83484	0.8649	−1.1859	0.8119	0.82014	0.6659	−0.5940
0.0987	0.4033	0.8494	0.85853	0.7292	−1.0526	0.6853	0.84333	0.5779	−0.5403
0.0991	0.5003	0.7404	0.88029	0.6518	−0.8909	0.6103	0.86462	0.5277	−0.4646
0.0975	0.6045	0.6571	0.90602	0.5953	−0.6960	0.5510	0.88976	0.4903	−0.3697
0.0989	0.6998	0.5987	0.93056	0.5571	−0.4965	0.5096	0.91367	0.4656	−0.2690
0.1016	0.8015	0.5515	0.95855	0.5286	−0.2679	0.4829	0.94091	0.4544	−0.1446
0.0982	0.8510	0.5451	0.97457	0.5312	−0.1532	0.4688	0.95662	0.4485	−0.0918
0.1997	0.0955	1.1890	0.77781	0.9248	−1.3349	0.9010	0.76398	0.6883	−0.6727
0.2019	0.1978	0.9687	0.79482	0.7699	−1.2326	0.7598	0.78041	0.5930	−0.6325
0.1978	0.3019	0.8143	0.81471	0.6634	−1.0949	0.6591	0.79980	0.5271	−0.5700
0.1997	0.4027	0.7056	0.83407	0.5885	−0.9172	0.5856	0.81866	0.4794	−0.4845
0.2005	0.5029	0.6337	0.85502	0.5418	−0.7158	0.5281	0.83905	0.4431	−0.3900
0.2001	0.6002	0.5734	0.87729	0.5030	−0.5169	0.4893	0.86067	0.4211	−0.2868
0.1980	0.7021	0.5318	0.90309	0.4803	−0.2954	0.4599	0.88571	0.4073	−0.1721
0.3027	0.0956	0.9465	0.75864	0.7180	−1.2584	0.7398	0.74457	0.5508	−0.6519
0.3018	0.1992	0.7846	0.77542	0.6084	−1.1162	0.6358	0.76084	0.4837	−0.5864
0.2998	0.3005	0.6839	0.79319	0.5425	−0.9391	0.5660	0.77816	0.4404	−0.5018
0.2969	0.4044	0.6077	0.81297	0.4940	−0.7405	0.5122	0.79744	0.4084	−0.4039
0.2995	0.5026	0.5608	0.83199	0.4666	−0.5198	0.5042	0.81588	0.4114	−0.2701
0.3003	0.5983	0.5135	0.85248	0.4377	−0.3116	0.4447	0.83569	0.3716	−0.1849
0.4017	0.0978	0.7681	0.74196	0.5699	−1.1291	0.6278	0.72771	0.4569	−0.5909
0.4027	0.1974	0.6722	0.75691	0.5088	−0.9430	0.5516	0.74224	0.4094	−0.5081
0.4058	0.2949	0.5916	0.77212	0.4568	−0.7472	0.4993	0.75706	0.3780	−0.4087
0.3998	0.4012	0.5374	0.79171	0.4255	−0.5342	0.4615	0.77617	0.3582	−0.3003
0.3991	0.5007	0.5002	0.81065	0.4055	−0.3119	0.4340	0.79457	0.3448	−0.1861
0.5016	0.0971	0.6502	0.72611	0.4721	−0.9541	0.5420	0.71174	0.3858	−0.5095
0.5016	0.1974	0.5782	0.74058	0.4282	−0.7519	0.4971	0.72583	0.3608	−0.4047
0.4982	0.3017	0.5250	0.75721	0.3975	−0.5359	0.4522	0.74211	0.3356	−0.3004
0.4968	0.4008	0.4889	0.77393	0.3784	−0.3157	0.4248	0.75848	0.3222	−0.1878
0.6042	0.0956	0.5703	0.71110	0.4055	−0.7425	0.4840	0.69664	0.3374	−0.4022
0.6010	0.2020	0.5140	0.72619	0.3733	−0.5224	0.4445	0.71134	0.3162	−0.2907
0.6081	0.2952	0.4761	0.73883	0.3518	−0.2957	0.4147	0.72376	0.3001	−0.1754
0.7003	0.0984	0.5104	0.69875	0.3566	−0.5204	0.4434	0.68413	0.3033	−0.2850
0.7043	0.1986	0.4702	0.71145	0.3345	−0.2857	0.4109	0.69650	0.2862	−0.1664
0.8029	0.1023	0.4674	0.68682	0.3210	−0.2646	0.4106	0.67203	0.2759	−0.1497
0.8511	0.0523	0.4702	0.67539	0.3176	−0.2583	0.4115	0.66084	0.2719	−0.1443
0.8472	0.1021	0.4543	0.68187	0.3098	−0.1546	0.3988	0.66705	0.2660	−0.0919
0.8964	0.0556	0.4548	0.67109	0.3052	−0.1381	0.3997	0.65652	0.2624	−0.0800

The viscosity deviations for the ternary mixture have been fitted by the Cibulka equation

$$\Delta\eta = \Delta\eta_{\text{bin}} + x_1x_2(1 - x_1 - x_2)[B_1 + B_2x_1 + B_3x_2] \quad (2)$$

where

$$\Delta\eta_{\text{bin}} = x_1x_2\sum_{p=0}^{n}A_{p,12}(x_1 - x_2)^p + x_1x_3\sum_{q=0}^{m}A_{p,13}(x_1 - x_3)^q + x_2x_3\sum_{r=0}^{l}A_{p,23}(x_2 - x_3)^r \quad (3)$$

with $A_{p,ij}$ being the binary parameters of a Redlich−Kister-type equation for the constituent binary mixtures, which have already been published,[5,6,14] and x_i being the mole fraction of component i in the ternary data point.

The coefficients, B_p, and the standard deviations, σ, obtained by the least-squares method are shown in Table 3 along with the parameters of the binary mixtures.

Isolines at constant values of calculated $\Delta\eta$ from Cibulka's equation (eq 2) for the ternary system at both temperatures have been plotted in Figure 1. Three-dimensional surfaces of $\Delta\eta$ have been represented in Figure 2. As one can see in both Figures, viscosity deviations of the ternary system are negative over the whole composition range at both temperatures, and values increase (less negative values) when the temperature changes from 298.15 to

Table 3. Coefficients of Cibulka's Equation (Eq 2), A_p and B_p, and the Corresponding Standard Deviations, σ, for Viscosity Deviations of the Binary and Ternary Mixtures

system	T K	A_0	A_1	A_2	A_3	A_4	$\sigma(\Delta\eta)$ mPa·s
hexane (1) + 1,3-dioxolane (2)[a]	298.15	−0.2978	0.0702	−0.0446	0.0188		0.0006
	313.15	−0.2515	0.0654	−0.0202	−0.0186		0.0005
hexane (1) + 2-butanol (2)[b]	298.15	−4.5698	3.1302	−1.9632	2.1300	−1.6353	0.0049
	313.15	−2.3666	1.5650	−1.2544	0.7922		0.0050
1,3-dioxolane (1) + 2-butanol (2)[c]	298.15	−3.9257	2.7796	−2.7713	1.8650		0.0099
	313.15	−1.9697	1.2380	−1.1735	0.7938		0.0047

system	T K	B_1	B_2	B_3	$\sigma(\Delta\eta)$ mPa·s
hexane (1) + 1,3-dioxolane (2) + 2-butanol (3)	298.15	12.3814	−10.5730	−11.9753	0.0107
	313.15	5.2790	−4.5582	−4.8846	0.0072

[a] Reference 6. [b] Reference 14. [c] Reference 5.

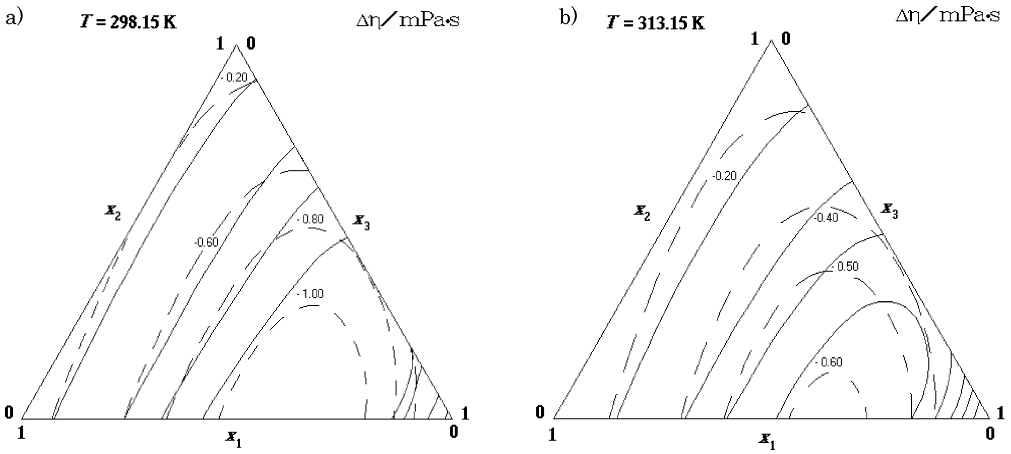

Figure 1. Isolines at constant $\Delta\eta$ for the ternary system hexane (1) + 1,3-dioxolane (2) + 2-butanol (3) correlated with the Cibulka equation (continuous lines) and prediction with the Wu−UNIFAC model with $A = 1$ (dashed lines): (a) at 298.15 K; (b) at 313.15 K.

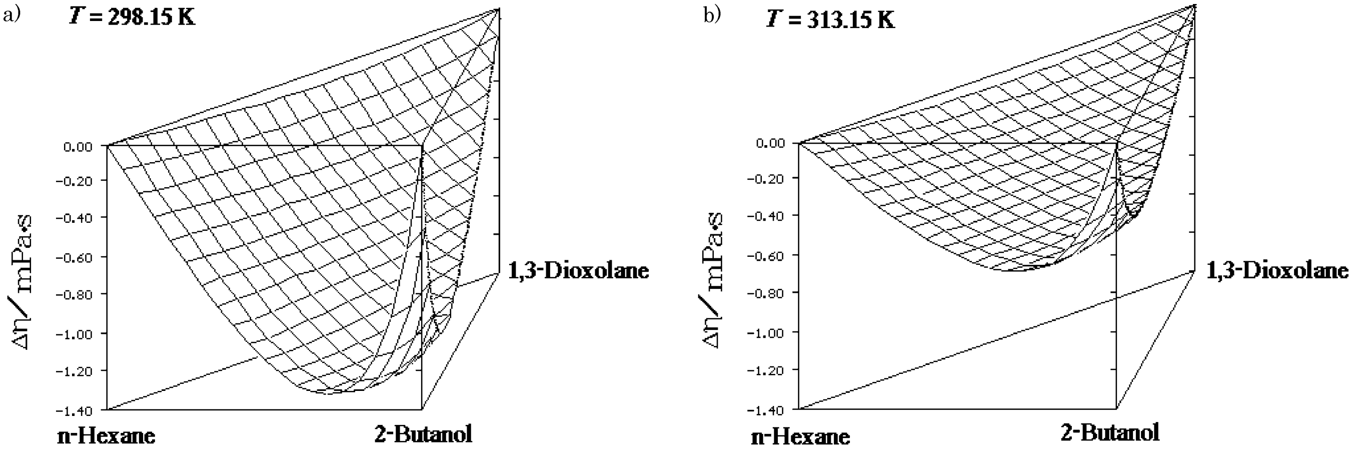

Figure 2. Three-dimensional surfaces of $\Delta\eta$ for the ternary system hexane (1) + 1,3-dioxolane (2) + 2-butanol (3) correlated with Cibulka equation: (a) at 298.15 K; (b) at 313.15 K.

313.15 K. This behavior is similar to that reported in previous papers for similar ternary mixtures.[4−7] We have concluded that the main effect in the viscosity deviations of the mixtures is the breaking of the self-interactions in the compounds during the mixing process: in this case, the hydrogen bonding of 2-butanol and the dipole−dipole interactions of 1,3-dioxolane. It would also be expected that the presence of new OH−O interactions in the mixture increases the viscosity, but according to experimental results, the effect is not as important as the breaking of self-interactions. Consequently, viscosity deviations of the mixture are negative, which means that our system has an easier flow than the pure liquids.

If we compare these values with viscosity deviations obtained for the ternary system[5] containing cyclohexane instead of hexane, we can realize that viscosity deviations are more negatives for the system with hexane. Values are particularly more negative when mole fractions of alkane and 2-butanol are important; however, for ternary mixtures with small mole fractions of 2-butanol, the differences are not relevant. This indicates that the main difference between the two ternary systems is due to the different

Table 4. Adjustable Parameters and Standard Deviations of the Nissan−Grunberg and McAllister Equations for the Viscosities of the Binary and Ternary Mixtures

T/K	equation	A_{ij}	A_{ji}	$\sigma(\eta)$/mPa·s
	Hexane (1) + 1,3-Dioxolane (2)			
298.15	Nissan−Grunberg	−0.5129		0.0010
	McAllister	35.3324	37.1099	0.0010
313.15	Nissan−Grunberg	−0.5068		0.0009
	McAllister	30.9432	32.3950	0.0005
	Hexane (1) + 2-Butanol (2)[a]			
298.15	Nissan−Grunberg	−2.4514		0.0336
	McAllister	45.2162	58.1791	0.0092
313.15	Nissan−Grunberg	−1.9858		0.0205
	McAllister	38.5137	46.4801	0.0040
	1,3-Dioxolane (1) + 2-Butanol (2)			
298.15	Nissan−Grunberg	−2.0130		0.0943
	McAllister	53.2206	59.2543	0.0241
313.15	Nissan−Grunberg	−1.5590		0.0478
	McAllister	43.7293	50.2199	0.0137

T/K	equation	A_{ijk}	$\sigma(\eta)$/mPa·s
	Hexane (1) + 1,3-Dioxolane (2) + 2-Butanol (3)		
298.15	Nissan−Grunberg	0.0499	0.0531
	McAllister	39.6359	0.0273
313.15	Nissan−Grunberg	0.5188	0.0257

[a] Reference 18

behavior of binary mixtures of alkanes + butanol, with viscosity deviations much more negatives for the systems with hexane instead of cyclohexane.

We also can analyze the differences with the mixture[6] containing 1-butanol instead of 2-butanol. Viscosity deviations are more negative for the mixture with 2-butanol, as we have observed for the constituent binary mixtures; values are more negative for the mixtures hexane + 2-butanol and 1,3-dioxolane + 2-butanol than for the mixtures containing 1-butanol. This agrees with the fact that hydrogen bonds in secondary butanol are not as strong as in primary butanol and their breaking is easier during the mixing process.

The extension to multicomponent mixtures of the correlation equations of Nissan−Grunberg and McAllister proposed by Canosa et al.[1] have been applied to the studied system.

Nissan−Grunberg:

$$\ln(\eta) = \sum_{i}^{n} x_i \ln(\eta_i) + \sum_{i}^{n}\sum_{j>i}^{n} x_i x_j A_{ij} + \sum_{i}^{n}\sum_{j>i}^{n}\sum_{k>j}^{n} x_i x_j x_k A_{ijk} \quad (4)$$

McAllister:

$$\ln(\eta V) = \sum_{i}^{n} x_i^{3} \ln(\eta_i V_i) + 3\sum_{i}^{n}\sum_{j\neq i}^{n} x_i^{2} x_j \ln(A_{ij}) + 6\sum_{i}^{n}\sum_{j>i}^{n}\sum_{k>j}^{n} x_i x_j x_k \ln(A_{ijk}) \quad (5)$$

In each equation, A_{ij} are the binary correlation parameters, and A_{ijk} is the ternary correlation parameter. V and V_i are, respectively, the molar volume of the mixture and of the pure components. The parameters A_{ij} are estimated by adjusting the data of the binary mixtures with the nonextended equations. Then, these parameters are included in the extended equation, and A_{ijk} is determined by fitting the ternary mixture data. Correlation parameters and standard deviations in absolute viscosity, obtained with these equations, are presented in Table 4. The best correlation for all of the binary systems and also for the ternary one is obtained when the McAllister equation is used.

Table 5. Relative Root Mean Square Deviation (RMSD$_r$) of the Wu Model for the Ternary System

		RMSD$_r$(%)	
system	T/K	$A = 1$	$A = 2.45$
hexane (1) + 1,3-dioxolane (2) + 2-butanol (3)	298.15	15.90	29.89
	313.15	12.94	20.37

Wu used the following modified viscosity equation of Eyring et al.[10] to predict the viscosity of liquid mixtures:

$$\eta = \frac{hN}{V}\exp\left[\frac{(x_i G_i^{*}) - \dfrac{G^{E}}{A}}{RT}\right] \quad (6)$$

where h is Planck's constant, N is Avogadro's number, V is the molar volume of the liquid mixture, G_i^{*} is the Gibbs energy of activation of the viscous flow of pure liquid i, G^{E} is the Gibbs excess energy, A is an empirical factor that can be equal to 1 or 2.45 according to the original work of Wu[2], T is the absolute temperature, and R is the gas constant. G_i^{*} can be obtained from the corresponding viscosity and molar volume of the pure components using eq 6 with $x_i = 1$ and $G^{E} = 0$.

In the Wu model, G^{E} can be estimated with different group contribution methods. In this paper, we have used the UNIFAC parameters proposed by Gmehling et al.[12]

Table 5 shows the relative root mean square deviation (RMSD$_r$) values for the ternary mixture at both temperatures, defined by

$$\mathrm{RMSD_r} = \left[\frac{1}{m}\sum\left(\frac{\eta_{\mathrm{calcd}} - \eta_{\mathrm{exptl}}}{\eta_{\mathrm{exptl}}}\right)^{2}\right]^{1/2} \quad (7)$$

where m is the number of experimental points and η_{calcd} and η_{exptl} are the calculated and experimental absolute viscosity, respectively.

In Table 5, we can see that viscosity predictions are not very far from experimental values. Differences between experimental and predicted viscosities are similar to those reported in previous papers. We have realized that UNIFAC predictions work better with a value of $A = 1$ for the ternary mixtures containing 2-butanol, whereas for the systems with 1-butanol predictions with $A = 2.45$ are closer to experimental values. Consequently, in this paper, we have plotted viscosity predictions of the UNIFAC model with $A = 1$ for the ternary mixture along with experimental measurements in Figure 1.

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