

Density, Viscosity, Excess Molar Volume, and Viscosity Deviation of Three Amyl Alcohols + Ethanol Binary Mixtures from 293.15 to 323.15 K

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Density and kinematic viscosity of (2-methyl-1-butanol + ethanol), (2-methyl-2-butanol + ethanol), and (3-methyl-1-butanol + ethanol) binary solutions were measured over the entire composition range and at seven different temperatures (293.15, 298.15, 303.15, 308.15, 313.15, 318.15, and 323.15) K, at atmospheric pressure. Redlich–Kister-type polynomial equations were used to fit the calculated excess molar volumes and viscosity deviations.

Introduction

In the last years, our investigation group has carried out some diverse experimental studies to determine the physical properties of different substances with a great interest in chemical industry (alcohol or glycols, organic acids, alkanolamines) in pure state or aqueous solution. In this work, continuing with these researches, we have studied the behavior of binary mixtures of amyl alcohol + ethanol over the entire concentration range and at different temperatures.

The amyl alcohols are used fundamentally for the perfumes composition and the synthesis of fruit essences. They are also used as solvents for surfaces and lacquer baths, inks for print, and dyes for wool as well as in the chemical production of photographic and pharmaceutical substances. Finally, they are an intermediate in the production of amyl acetate and other amyl esters. In some of these uses, the knowledge of their physical properties is very important.

Amyl alcohols are miscible in different organic solvents, such as ethanol, diethyl ether, or acetone, and partially miscible in water. Therefore, in this work densities and kinematic viscosities of binary mixtures of (2-methyl-1-butanol + ethanol), (2-methyl-2-butanol + ethanol), and (3-methyl-1-butanol + ethanol) have been determined over the entire concentration range, at temperature of 293.15 K to 323.15 K.

Experimental Section

All solutions were prepared by mass using an analytical balance with an accuracy of ± 0.0001 g. The solutes were Merck reagents of nominal purity > 99.7 %.

The density, ρ , was measured with an Anton Paar DSA 4500 vibration tube densimeter with an accuracy of $\pm 1 \cdot 10^{-5}$ g·cm⁻³. The apparatus allows varying the temperature in the range used in the present study. The uncertainty in the density measurement was $\pm 5 \cdot 10^{-4}$ g·cm⁻³.

The kinematic viscosity, ν , was determined from the transit time of the liquid meniscus through a capillary supplied by

Schott (cap no. I, (0.58 \pm 0.01) mm i.d., $K = 0.009918$ mm²·s⁻²) measured with an uncertainty of ± 0.0005 mm²·s⁻¹ using

$$\nu = K(t - \theta) \quad (1)$$

where t is the efflux time, K is the characteristic constant of the capillary viscometer, and θ is a correction value to correct the final effects.

The glass capillary was connected to a Schott-Geräte AVS 350 automatic Ubbelohde viscometer. An electronic stopwatch with an accuracy of ± 0.01 s was used for measuring efflux times. The viscometer was immersed in a bath, and the precision of the temperature control in all these measurements was ± 0.05 K. Each measurement was repeated at least five times with a maximum deviation of less than 0.4 %. The dynamic viscosity, η , was calculated by multiplying the kinematic viscosity by the corresponding density ($\eta = \nu\rho$).

Finally, the densimeter and the viscometer were calibrated with distilled water and pure ethanol, as is recommended by Marsh.¹ The values measured at the temperature of work are included in Table 1 and are compared with values published by other authors.^{2–8}

Results and Discussion

Experimental densities and kinematic viscosities of binary mixtures of (2-methyl-1-butanol, 2-methyl-2-butanol, and 3-methyl-1-butanol) + ethanol, from $T = (293.15$ to $323.15)$ K, are displayed in Tables 2 to 7. The experimental data show that, for each temperature, these properties increase with amyl alcohol concentration. On the other hand, those values decrease when the temperature increases for a specific mass fraction of amyl alcohol.

The influence of the temperature on the density was a linear dependence in the range temperature studied,⁹ whereas several authors in the literature have found nonlinear dependences over large ranges of temperature. In contrast with this behavior, the influence of the temperature on the dynamic viscosity was a nonlinear dependence. For this reason several equations have been used to fit our experimental values: the equation proposed by Cornelissen and Waterman,¹⁰ the Guzman–Andrade's equa-

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Table 1. Review of the Literature Data for the Density and the Viscosity of Water and Ethanol

T/K	water				ethanol			
	$\rho/(g\cdot cm^{-3})$		$\eta/(mPa\cdot s)$		$\rho/(g\cdot cm^{-3})$		$\eta/(mPa\cdot s)$	
	this work	lit (ref)	this work	lit (ref)	this work	lit (ref)	this work	lit (ref)
298.15	0.99710	0.99704 ^a	0.8925	0.8900 ^a	0.7853	0.78525 ^e 0.78549 ^f	1.0912	1.090 ^e 1.084 ^f
303.15	0.99570	0.99565 ^a 0.99560 ^c	0.7990	0.7970 ^c	0.7809	0.78080 ^e 0.78079 ^g	0.9575	0.950 ^e
308.15	0.99410	0.99403 ^a	0.7220		0.7765	0.77650 ^e	0.8758	0.870 ^e
313.15	0.99222	0.99221 ^a 0.99220 ^c	0.6550	0.6560 ^d 0.6530 ^c	0.7723	0.77220 ^e	0.7915	0.790 ^e
318.15	0.99022	0.99021 ^b	0.5950	0.5950 ^a	0.7679	0.76760 ^e	0.7218	0.720 ^e
323.15	0.98798	0.98804 ^a 0.99803 ^b	0.5460	0.5470 ^a	0.7635	0.76320 ^e	0.6323	0.630 ^e

^a Ref 2. ^b Ref 3. ^c Ref 4. ^d Ref 5. ^e Ref 6. ^f Ref 7. ^g Ref 8.**Table 2.** Density ρ for 2-Methyl-1-butanol (1) + Ethanol (2) Mixtures from $T = 293.15$ K to 323.15 K

x_1	$\rho/(g\cdot cm^{-3})$							
	$T = 293.15$ K	$T = 298.15$ K	$T = 303.15$ K	$T = 308.15$ K	$T = 313.15$ K	$T = 318.15$ K	$T = 323.15$ K	
0.0000	0.7897	0.7853	0.7809	0.7765	0.7722	0.7679	0.7635	
0.0268	0.7909	0.7866	0.7823	0.7780	0.7737	0.7694	0.7651	
0.0549	0.7921	0.7879	0.7837	0.7794	0.7752	0.7710	0.7667	
0.0845	0.7934	0.7892	0.7850	0.7809	0.7767	0.7725	0.7683	
0.1156	0.7947	0.7906	0.7864	0.7823	0.7782	0.7741	0.7699	
0.1484	0.7959	0.7919	0.7878	0.7838	0.7797	0.7756	0.7715	
0.1830	0.7972	0.7932	0.7892	0.7852	0.7812	0.7772	0.7731	
0.2584	0.7999	0.7960	0.7920	0.7881	0.7842	0.7803	0.7763	
0.3433	0.8027	0.7988	0.7950	0.7912	0.7873	0.7835	0.7796	
0.4395	0.8055	0.8018	0.7980	0.7943	0.7905	0.7868	0.7829	
0.5495	0.8084	0.8048	0.8011	0.7975	0.7938	0.7901	0.7863	
0.6765	0.8114	0.8079	0.8042	0.8007	0.7970	0.7934	0.7897	
0.8247	0.8145	0.8110	0.8074	0.8039	0.8003	0.7967	0.7932	
1.0000	0.8176	0.8141	0.8105	0.8071	0.8036	0.8001	0.7966	

Table 3. Density ρ for 2-Methyl-2-butanol (1) + Ethanol (2) Mixtures from $T = 293.15$ K to 323.15 K

x_1	$\rho/(g\cdot cm^{-3})$							
	$T = 293.15$ K	$T = 298.15$ K	$T = 303.15$ K	$T = 308.15$ K	$T = 313.15$ K	$T = 318.15$ K	$T = 323.15$ K	
0.0000	0.7897	0.7853	0.7809	0.7765	0.7722	0.7679	0.7635	
0.0268	0.7906	0.7864	0.7821	0.7778	0.7735	0.7693	0.7649	
0.0549	0.7915	0.7873	0.7832	0.7789	0.7748	0.7706	0.7663	
0.0845	0.7922	0.7882	0.7842	0.7800	0.7760	0.7719	0.7677	
0.1156	0.7930	0.7891	0.7852	0.7811	0.7772	0.7732	0.7691	
0.1484	0.7939	0.7900	0.7862	0.7823	0.7784	0.7744	0.7704	
0.1830	0.7948	0.7910	0.7873	0.7834	0.7796	0.7757	0.7718	
0.2584	0.7970	0.7933	0.7897	0.7859	0.7822	0.7784	0.7746	
0.3433	0.7995	0.7959	0.7923	0.7886	0.7849	0.7812	0.7775	
0.4395	0.8024	0.7988	0.7952	0.7915	0.7879	0.7842	0.7805	
0.5495	0.8053	0.8018	0.7982	0.7946	0.7909	0.7873	0.7837	
0.6765	0.8083	0.8048	0.8013	0.7976	0.7940	0.7905	0.7870	
0.8247	0.8112	0.8077	0.8042	0.8006	0.7971	0.7936	0.7902	
1.0000	0.8139	0.8105	0.8070	0.8035	0.8000	0.7966	0.7933	

Table 4. Density ρ for 3-Methyl-1-butanol (1) + Ethanol (2) Mixtures from $T = 293.15$ K to 323.15 K

x_1	$\rho/(g\cdot cm^{-3})$							
	$T = 293.15$ K	$T = 298.15$ K	$T = 303.15$ K	$T = 308.15$ K	$T = 313.15$ K	$T = 318.15$ K	$T = 323.15$ K	
0.0000	0.7897	0.7853	0.7809	0.7765	0.7722	0.7679	0.7635	
0.0268	0.7905	0.7862	0.7818	0.7775	0.7732	0.7690	0.7646	
0.0549	0.7913	0.7870	0.7827	0.7784	0.7742	0.7700	0.7657	
0.0845	0.7921	0.7879	0.7837	0.7894	0.7752	0.7710	0.7667	
0.1156	0.7929	0.7888	0.7846	0.7804	0.7762	0.7720	0.7678	
0.1484	0.7938	0.7897	0.7855	0.7814	0.7772	0.7731	0.7689	
0.1830	0.7947	0.7906	0.7865	0.7824	0.7782	0.7741	0.7699	
0.2584	0.7966	0.7925	0.7885	0.7844	0.7803	0.7763	0.7721	
0.3433	0.7985	0.7945	0.7905	0.7865	0.7824	0.7784	0.7743	
0.4395	0.8004	0.7965	0.7925	0.7886	0.7846	0.7806	0.7766	
0.5495	0.8024	0.7985	0.7946	0.7907	0.7867	0.7828	0.7789	
0.6765	0.8043	0.8005	0.7967	0.7929	0.7889	0.7851	0.7812	
0.8247	0.8063	0.8026	0.7987	0.7950	0.7910	0.7873	0.7834	
1.0000	0.8083	0.8046	0.8008	0.7971	0.7932	0.7895	0.7857	

Table 5. Kinematic Viscosity ν for 2-Methyl-1-butanol (1) + Ethanol (2) Mixtures from $T = 293.15$ K to 323.15 K

x_1	$\nu/(mm^2 \cdot s^{-1})$						
	$T = 293.15$ K	$T = 298.15$ K	$T = 303.15$ K	$T = 308.15$ K	$T = 313.15$ K	$T = 318.15$ K	$T = 323.15$ K
0.0000	1.5248	1.3895	1.2261	1.1278	1.0250	0.9399	0.8282
0.0915	1.6616	1.5003	1.3164	1.1983	1.0785	0.9797	0.8586
0.1753	1.7965	1.6110	1.4077	1.2717	1.1367	1.0251	0.8958
0.2524	1.9181	1.7130	1.4954	1.3437	1.1960	1.0735	0.9368
0.3235	2.0393	1.8161	1.5851	1.4194	1.2589	1.1264	0.9833
0.3894	2.1715	1.9295	1.6846	1.5037	1.3306	1.1865	1.0363
0.4505	2.3219	2.0592	1.7984	1.5994	1.4108	1.2547	1.0957
0.5605	2.6732	2.3576	2.0569	1.8187	1.5954	1.4116	1.2327
0.6567	3.0617	2.6912	2.3483	2.0654	1.8039	1.5898	1.3881
0.7416	3.4735	3.0445	2.6577	2.3298	2.0281	1.7823	1.5575
0.8170	3.9200	3.4317	2.9992	2.6223	2.2786	1.9989	1.7485
0.8844	4.4525	3.8929	3.4069	2.9705	2.5770	2.2582	1.9780
0.9451	5.1446	4.4938	3.9396	3.4295	2.9727	2.6012	2.2830
1.0000	6.1084	5.3379	4.6898	4.0827	3.5374	3.0938	2.7215

Table 6. Kinematic Viscosity ν for 2-Methyl-2-butanol (1) + Ethanol (2) Mixtures from $T = 293.15$ K to 323.15 K

x_1	$\nu/(mm^2 \cdot s^{-1})$						
	$T = 293.15$ K	$T = 298.15$ K	$T = 303.15$ K	$T = 308.15$ K	$T = 313.15$ K	$T = 318.15$ K	$T = 323.15$ K
0.0000	1.5248	1.3895	1.2261	1.1278	1.0250	0.9399	0.8282
0.0915	1.6373	1.5026	1.3351	1.2300	1.1181	1.0234	0.9013
0.1753	1.7587	1.6159	1.4413	1.3257	1.2038	1.1016	0.9680
0.2524	1.8920	1.7353	1.5507	1.4202	1.2880	1.1775	1.0318
0.3235	2.0360	1.8619	1.6642	1.5172	1.3729	1.2528	1.0948
0.3894	2.1889	1.9939	1.7827	1.6183	1.4605	1.3288	1.1586
0.4505	2.3495	2.1331	1.9057	1.7240	1.5510	1.4063	1.2230
0.5605	2.6943	2.4320	2.1672	1.9485	1.7406	1.5669	1.3544
0.6567	3.0673	2.7555	2.4472	2.1873	1.9396	1.7335	1.4877
0.7416	3.4593	3.0960	2.7398	2.4340	2.1422	1.9029	1.6203
0.8170	3.8599	3.4413	3.0378	2.6838	2.3442	2.0708	1.7507
0.8844	4.2629	3.7838	3.3322	2.9269	2.5417	2.2324	1.8749
0.9451	4.6727	4.1243	3.6216	3.1621	2.7308	2.3839	1.9910
1.0000	5.1087	4.4741	3.9086	3.3870	2.9085	2.5214	2.0940

Table 7. Kinematic Viscosity ν for 3-Methyl-1-butanol (1) + Ethanol (2) Mixtures from $T = 293.15$ K to 323.15 K

x_1	$\nu/(mm^2 \cdot s^{-1})$						
	$T = 293.15$ K	$T = 298.15$ K	$T = 303.15$ K	$T = 308.15$ K	$T = 313.15$ K	$T = 318.15$ K	$T = 323.15$ K
0.0000	1.5248	1.3895	1.2261	1.1278	1.0250	0.9399	0.8282
0.0915	1.5522	1.4246	1.2658	1.1694	1.0673	0.9799	0.8715
0.1753	1.6131	1.4819	1.3220	1.2213	1.1174	1.0257	0.9127
0.2524	1.6775	1.5419	1.3815	1.2749	1.1661	1.0700	0.9528
0.3235	1.7481	1.6077	1.4476	1.3327	1.2177	1.1172	0.9961
0.3894	1.8311	1.6859	1.5230	1.3985	1.2772	1.1705	1.0456
0.4505	1.9316	1.7785	1.6087	1.4737	1.3460	1.2315	1.1022
0.5605	2.1884	2.0061	1.8112	1.6525	1.5077	1.3739	1.2344
0.6567	2.5104	2.2811	2.0519	1.8635	1.6935	1.5377	1.3854
0.7416	2.8877	2.5971	2.3282	2.1026	1.8994	1.7188	1.5485
0.8170	3.3143	2.9584	2.6408	2.3675	2.1269	1.9171	1.7215
0.8844	3.7958	3.3721	2.9915	2.6601	2.3802	2.1336	1.9041
0.9451	4.3509	3.8414	3.3834	2.9814	2.6556	2.3629	2.0943
1.0000	5.0041	4.3550	3.8147	3.3305	2.9366	2.5896	2.2863

tion,¹¹ and others. The best one is a polynomial equation:

$$\eta/(mPa \cdot s) = \sum_{n=0}^2 a_n T^n / K \quad (2)$$

where a_n are the fitting parameters whose values vary with the mole fraction.

The excess molar volumes of mixtures, V^E , were calculated from density measurements by applying the following equation:¹²

$$V^E/(cm^3 \cdot mol^{-1}) = \sum_{i=1}^2 x_i M_i \left(\frac{1}{\rho_m} - \frac{1}{\rho_i} \right) \quad (3)$$

where x_i represents the molar fraction of the component i in

the mixture, ρ_i represents the density of the i th pure component, and ρ_m is the measured mixture density.

On the other hand, experimental viscosities were used to calculate the dynamic viscosity deviation, $\Delta \ln \eta$, defined by

$$\Delta \ln \eta/(mPa \cdot s) = \ln \eta_{12} - (x_1 \ln \eta_1 + x_2 \ln \eta_2) \quad (4)$$

where η_1 and η_2 are the viscosity of the pure components, and η_{12} is the viscosity of the mixture.

Finally, a Redlich-Kister-type equation is applied to correlate the excess molar volume and the viscosity deviation. For a binary system, the Redlich-Kister equation has the following expression:

$$Y = x_1 x_2 \sum_{i=0}^n A_i (1 - 2x_2)^i \quad (5)$$

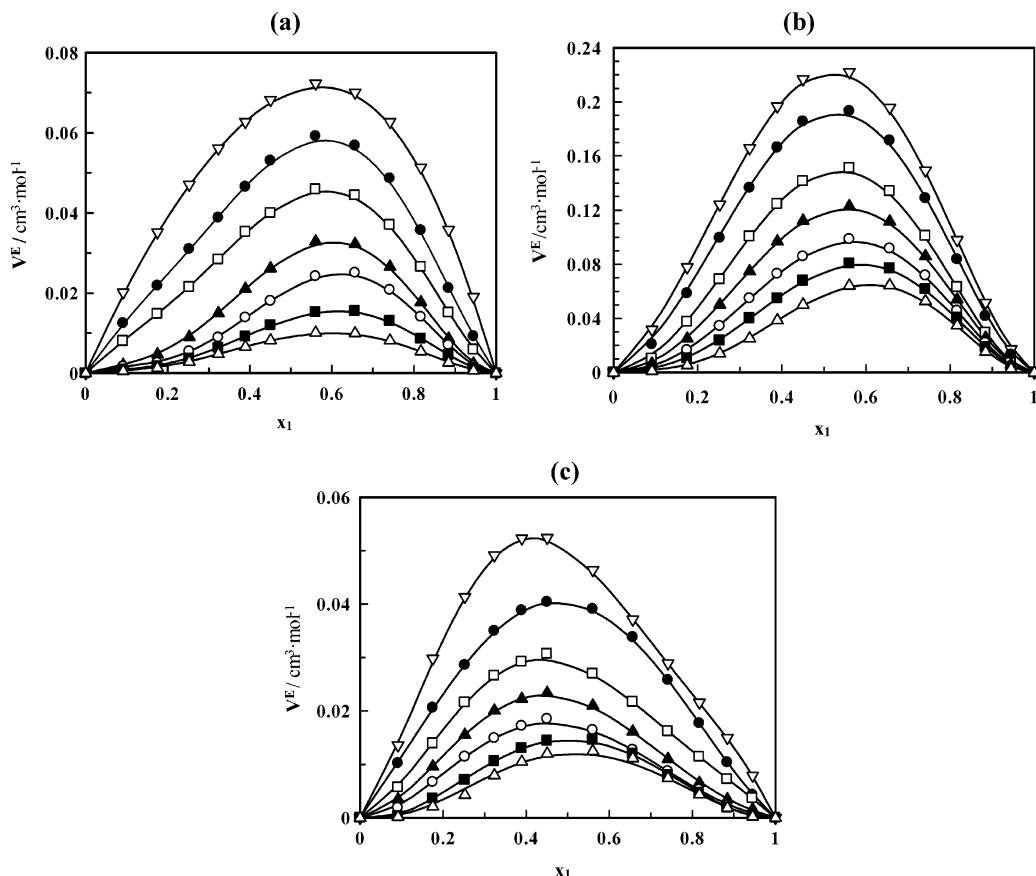


Figure 1. Excess molar volumes, V^E , for (a) 2-methyl-1-butanol (1) + ethanol (2), (b) 2-methyl-2-butanol (1) + ethanol (2), and (c) 3-methyl-1-butanol (1) + ethanol (2): ∇ , 293.15 K; \bullet , 298.15 K; \square , 303.15 K; \blacktriangle , 308.15 K; \circ , 313.15 K; \blacksquare , 318.15 K; \triangle , 323.15 K; —, Redlich-Kister fit curves.

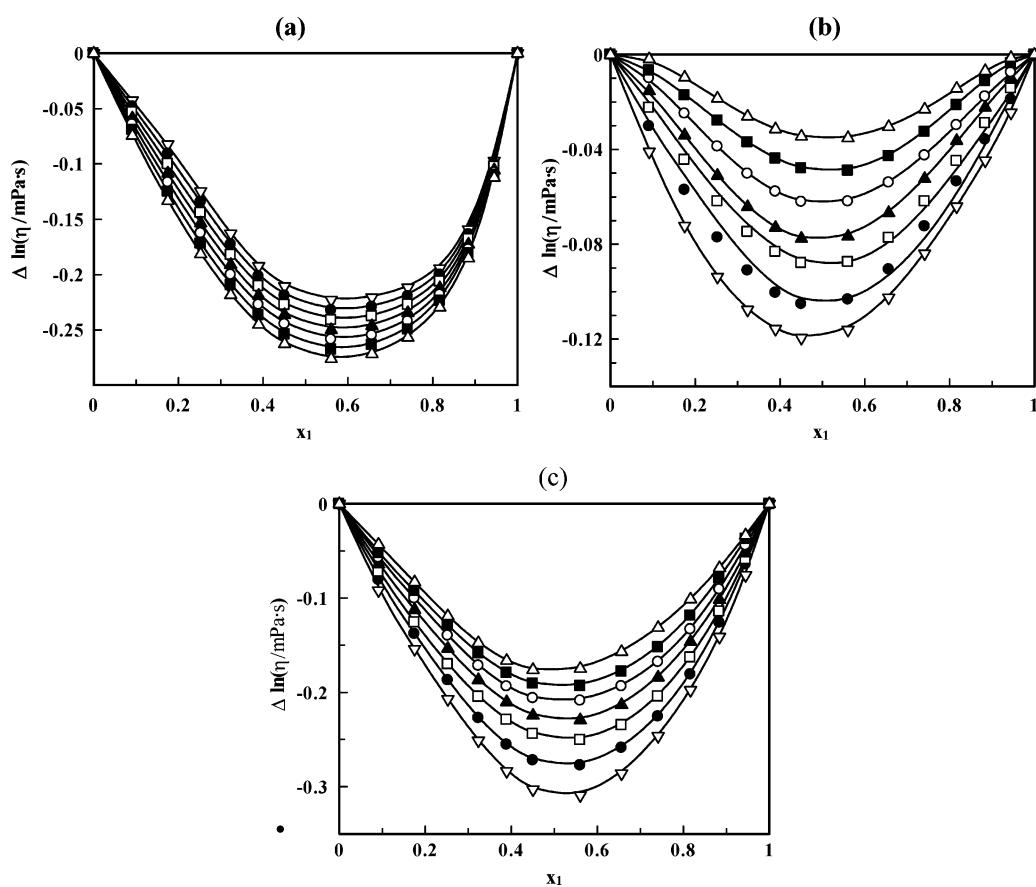


Figure 2. Viscosity deviations, $\Delta \ln(\eta)$, for (a) 2-methyl-1-butanol (1) + ethanol (2), (b) 2-methyl-2-butanol (1) + ethanol (2), and (c) 3-methyl-1-butanol (1) + ethanol (2): ∇ , 293.15 K; \bullet , 298.15 K; \square , 303.15 K; \blacktriangle , 308.15 K; \circ , 313.15 K; \blacksquare , 318.15 K; \triangle , 323.15 K; —, Redlich-Kister fit curves.

Table 8. Adjustable Parameters (A_i) with the Standard Deviations (σ_{st}) for Excess Molar Volumes (V^E)

T/K	A_0	A_1	A_2	A_3	σ_{st}^a
2-Methyl-1-butanol + Ethanol					
293.15	0.28266	-0.08621	0.02542	0.02358	0.022
298.15	0.22630	-0.12553	-0.07851	0.14438	0.027
303.15	0.17431	-0.11138	-0.08625	0.13182	0.016
308.15	0.11956	-0.12484	-0.10857	0.14584	0.014
313.15	0.08541	-0.11626	-0.07154	0.14107	0.016
318.15	0.05481	-0.06646	-0.04748	0.07547	0.009
323.15	0.03710	-0.03731	-0.03509	0.04168	0.005
2-Methyl-2-butanol + Ethanol					
293.15	0.89567	-0.15602	-0.72951	0.18039	0.038
298.15	0.77331	-0.18615	-0.68333	0.19935	0.015
303.15	0.59806	-0.20306	-0.63099	0.21421	0.043
308.15	0.47989	-0.23482	-0.51492	0.23672	0.036
313.15	0.37648	-0.24994	-0.40939	0.26098	0.041
318.15	0.30148	-0.26094	-0.32305	0.28065	0.045
323.15	0.23186	-0.28193	-0.24491	0.32837	0.042
3-Methyl-1-butanol + Ethanol					
293.15	0.20134	0.10699	-0.06754	-0.14586	0.023
298.15	0.16263	0.01914	-0.07966	-0.00089	0.013
303.15	0.11624	0.04888	-0.07079	-0.07546	0.034
308.15	0.09050	0.03890	-0.08405	-0.05036	0.020
313.15	0.07063	0.02448	-0.07353	-0.03439	0.022
318.15	0.05930	-0.00323	-0.07643	-0.00386	0.017
323.15	0.04904	-0.01004	-0.06360	0.00294	0.051

^a $\sigma_{st} = [\{\sum(V_{\text{cal}}^E - V_{\text{exp}}^E)^2\}/(N - n)]^{1/2}$, where N is the number of data points and n is the number of coefficients.

Table 9. Adjustable Parameters (A_i) with the Standard Deviations (σ_{st}) for Viscosity Deviations $\Delta \ln \eta$

T/K	A_0	A_1	A_2	A_3	A_4	A_5	σ_{st}^a
2-Methyl-1-butanol + Ethanol							
293.15	-0.87716	0.24511	0.10848	0.97736	-0.65281	-0.41636	0.024
298.15	-0.91233	0.25205	0.05839	0.87535	-0.61862	-0.30806	0.021
303.15	-0.94653	0.25316	0.00014	0.79883	-0.57498	-0.22241	0.027
308.15	-0.98189	0.25847	-0.03723	0.70474	-0.56877	-0.11219	0.017
313.15	-1.01568	0.26406	-0.08409	0.63791	-0.54259	-0.05656	0.023
318.15	-1.05326	0.26392	-0.11839	0.57374	-0.51976	0.00542	0.023
323.15	-1.08623	0.26324	-0.16775	0.51997	-0.49137	0.06357	0.016
2-Methyl-2-butanol + Ethanol							
293.15	-0.47829	-0.04891	0.06741	-0.09476	-0.08262	0.18887	0.004
298.15	-0.42195	-0.01802	0.14754	-0.10650	-0.07456	0.18655	0.005
303.15	-0.35681	0.04228	0.13562	-0.07647	-0.01671	0.11736	0.004
308.15	-0.31477	-0.00609	0.17877	0.02608	-0.00126	0.00567	0.021
313.15	-0.25265	0.00550	0.15629	0.03452	0.01496	-0.00657	0.011
318.15	-0.19810	0.02746	0.16382	-0.02488	0.01756	-0.00437	0.045
323.15	-0.14254	0.01698	0.12873	0.02429	0.04309	-0.03354	0.009
3-Methyl-1-butanol + Ethanol							
293.15	-1.24232	0.14839	0.32949	0.24449	-0.52793	-0.28276	0.015
298.15	-1.11307	0.12457	0.16745	0.38250	-0.22838	-0.48621	0.007
303.15	-1.00106	0.14249	0.10273	0.15489	-0.14036	-0.19556	0.009
308.15	-0.92045	0.11307	0.17871	0.20629	-0.19057	-0.24262	0.010
313.15	-0.84103	0.06370	0.15849	0.43235	-0.10386	-0.52720	0.008
318.15	-0.77887	0.05860	0.18066	0.32280	-0.07602	-0.43759	0.006
323.15	-0.71376	-0.00770	0.24651	0.31991	-0.08850	-0.26950	0.005

^a $\sigma_{st} = [\{\sum(\ln(\eta)_{\text{cal}} - \ln(\eta)_{\text{exp}})^2\}/(N - n)]^{1/2}$, where N is the number of data points and n is the number of coefficients.

where Y represents the viscosity deviation or the excess volume; x_1 and x_2 are the mole fractions of the amyl alcohol and ethanol, respectively. In this equation, $n = 3$ for the excess molar volume, and $n = 5$ for the viscosity deviation. The coefficients, A_i , for the excess volume and the viscosity deviation are presented in Tables 8 and 9.

Equation 5 fits satisfactorily the deviation values calculated from experimental data of density and viscosity (Figures 1 and 2). When we analyze the results obtained concerning the influence of mixture composition upon the excess volumes, we can observe that these deviations are positive in all cases. However, the viscosity deviations for all systems are negative over the entire composition range.

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