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Density, Viscosity, Excess Molar Volume, Viscosity Deviation, and Their Correlations for Formamide + Three Alkan-1-ols Binary Systems

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Density and viscosity measurements for formamide + methanol (or + ethanol or + 1-propanol) binary systems over the whole concentration range in the temperature range (30 to 50) °C were made. The experimental results were fitted to empirical equations, whose calculated values are in good agreement with the experimental ones. The data of the mixtures were further used to calculate the excess molar volumes and viscosity deviations. These latter results were fitted by the Redlich−Kister polynomial relation to obtain the coefficients and the standard deviations.

Introduction

Experimental data of properties such as density (ρ) and viscosity (η) at several temperatures over the whole composition range for binary liquid mixtures are useful for a full understanding of their thermodynamic and transport properties, as well as for practical chemical engineering purposes.

This work is a part of an ongoing research program in which we study thermodynamic, transport, and spectroscopic properties of binary mixtures containing formamide as one of the components,[1−3] because little is known about binary mixtures with amides. On the other hand, excess thermodynamic properties and deviations of nonthermodynamic ones of binary liquid mixtures are fundamental for the design of industrial equipment and for the interpretation of the liquid state, particularly when polar components are involved. These properties have also been used as a qualitative and quantitative guide to predict the extent of complex formation in this kind of mixtures.[4−6]

The aim of this work is (i) to report experimental data of density and viscosity for formamide + methanol (or + ethanol or + 1-propanol) binary systems at (30, 35, 40, 45, and 50) °C, (ii) to obtain empirical equations involving both the temperature and mole fraction variables for these properties to facilitate the interpolation of the data, (iii) to use the data of the mixtures to calculate the excess molar volumes and viscosity deviations, and (iv) to fit these last results using the Redlich−Kister polynomial relation to obtain the coefficients and the standard deviations.

García et al.[7] have published similar data for formamide + (C$_1$−C$_5$) alkan-1-ols binary systems at 298.15 K, but we could not find any information about data measured at other temperatures in the available literature.

Experimental Section

Materials. Formamide (Riedel-de Haën, A. R.), methanol (Merck, p.a.), ethanol (Merck, p. GC), and 1-propanol (Merck, p.a.) were used as received, because their purities

were practically 100 mass % (GC). The purity of all the reagents was checked using a HP 6890 gas chromatograph with a TCD detector coupled with a ChemStation. Nitrogen was used as gas carrier.

Procedure. Airtight stoppered bottles were used for the preparation of the mixtures. The mass of the dry bottle was first determined. The less volatile component of the mixture was introduced in the bottle, and the total mass was recorded. Subsequently, the other component was introduced and the mass of the bottle along with the two components was determined. Each mixture was immediately used, after it was well mixed by means of a rotary motion. The compositions of the mixtures were determined from the masses of the two pure components and checked by gas chromatography (using the internal standard method for quantification), before use in the experiments. The pure components were stored over 0.3 nm molecular sieves to prevent water absorption, and their water contents were periodically checked by Karl Fischer titration using an automatic Karl Fischer titrator Mettler DL18. A Mettler Toledo AG245 balance with an uncertainty of ±0.1 mg was used, and the reported mole fractions have an uncertainty within ±0.0001. Densities and viscosities of freshly prepared mixtures were measured with a vibrating tube densimeter KEM DA300 (using degassed bidistilled water and dry air as calibrating substances) and Cannon-Fenske viscometers (calibrated with bidistilled water and benzene in order to determine both viscometer constants), respectively. The mixtures were introduced inside the cells of the densimeter and viscometer with the help of an airtight syringe, and the cells were immediately stoppered in order to prevent evaporation while the samples reached thermal equilibrium. The values of density and viscosity of pure and mixed components reported in this work were obtained by averaging four or five independent measurements. They have an accuracy of ±0.2 kg m^{-3} for density measurements, while the relative error for the viscosity was $\Delta\eta/\eta$ = 0.05. In all cases, a water bath thermostatically controlled to ±0.01 K was used, and the uncertainty in temperature was ±0.05 K. Experimental densities and viscosities, given in Table 1, closely agree with literature values.

Table 1. Data for the Pure Liquids at Several Temperatures

substance	t/(°C)	ρ/(kg m^{-3}) expt	ρ/(kg m^{-3}) lit.	η/(mPa s) expt	η/(mPa s) lit.
formamide	25[a]	1129.75	1129.15[b]	3.34	3.302[b]
	30	1124.7		2.8	
	35	1120.5		2.5	
	40	1116.4		2.2	
	45	1111.9		2.0	
	50	1107.8		1.6	
methanol	25[a]	786.63	786.37[b]	0.553	0.5513[b]
	30	781.9	781.65[f]	0.50	0.510[d]
	30				0.503[f]
	35	777.2	777.10[f]	0.46	0.469[f]
	40	772.5		0.44	0.449[c]
	40				0.456[d]
	45	767.5		0.41	
	50	762.7		0.40	0.403[d]
ethanol	25[a]	785.25	784.93[b]	1.09	1.0826[b]
	30	780.8	780.97[d]	0.95	0.987[b]
	30		780.75[e]		1.003[d]
	35	776.5	776.41[e]	0.87	
	40	772.2	772.03[e]	0.79	0.8309[c]
	40				0.834[d]
	45	767.6		0.72	
	50	763.2		0.63	0.702[d]
1-propanol	25[a]	799.54	799.60[b]	1.95	1.9430[b]
	30	795.6	795.8[e]	1.6	1.725[b]
	35	791.5		1.4	
	40	787.5		1.3	1.4050[c]
	45	783.2		1.1	
	50	779.0		0.92	

[a] Reference 7. [b] Reference 8. [c] Reference 9. [d] Reference 10. [e] Reference 11. [f] Reference 12.

Results and Discussion

Experimental results for the density and viscosity in the temperature range (25,[7] 30, 35, 40, 45, and 50) °C for the systems formamide + methanol, formamide + ethanol, and formamide + 1-propanol are listed in Table 2.

To obtain correlations among density or viscosity with temperature and composition for the [(x_1)formamide + $(1 - x_1)$methanol] (or + ethanol or + 1-propanol) binary systems, the following functional relationships were used:

$$\rho/(\text{kg m}^{-3}) = a_1(x_1) + b_1(x_1)t \qquad (1)$$

$$\eta/(\text{mPa s}) = a_2(x_1) \exp[b_2(x_1)/T] \qquad (2)$$

where a_1, b_1, a_2, and b_2 are parameters dependent on the mole fraction of formamide (x_1) while t and T are Celsius and Kelvin temperatures, respectively.

For this purpose, the densities of the mixtures were plotted against temperature for each binary system, keeping x_1 as a parameter. From these plots, and those of the viscosity against $1/T$, the parameters $a_i(x_1)$ and $b_i(x_1)$ were obtained using, for eq 1, a least-squares method, taking the errors as weight, and a nonlinear regression method based on the Levenberg–Marquardt algorithm,[13] also taking the errors as weight for eq 2. The appropriate number of significant digits was selected taking into account each error coefficient. Figures 1 and 2 show plots of the parameters a_1 and b_1 from eq 1 against the mole fraction of formamide x_1 for the formamide + methanol binary system, respectively. Since the plots for the other properties and systems are similar, they are not presented here.

The resulting equations for the density and viscosity of the three binary systems are collected in Table 3, in which
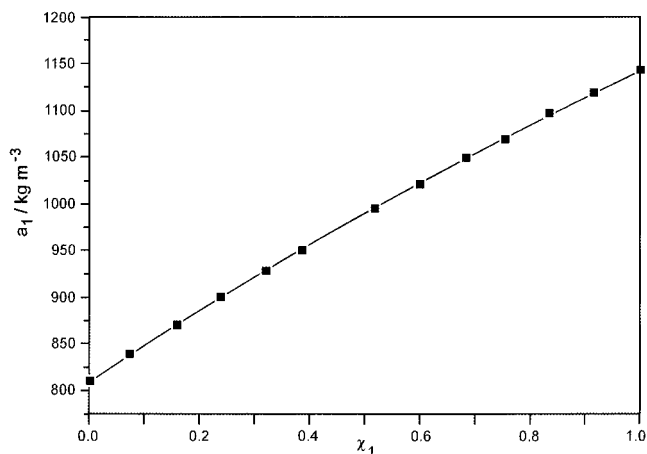
Figure 1. Parameter $a_1(x_1)$ in eq 1 versus mole fraction of formamide for the formamide + methanol binary system.
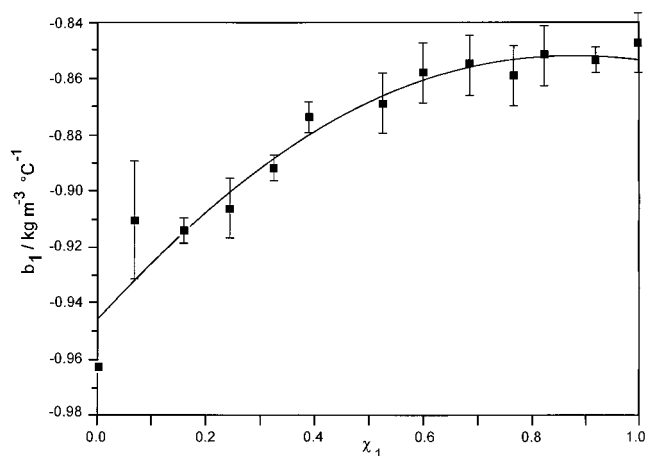
Figure 2. Parameter $b_1(x_1)$ in eq 1 versus mole fraction of formamide for the formamide + methanol binary system.

the $a_i(x_1)$ and $b_i(x_1)$ coefficients are presented together with their standard deviations (σ). From any data set, the fitting reduces the correlations to the same equation for density and the same equation for viscosity of pure formamide ($\rho = 1150.4 - 0.86t$, and $\eta = 0.00028 \exp(2800/T)$, respectively). The averages of the percentage differences between calculated and experimental density values corresponding to the different temperatures compare well with experimental values within ±0.03% for formamide, 0.05% for methanol and ethanol, and 0.01% for propanol. For these extreme concentrations and in the same temperature range, the fitting of the viscosity is poorer with average values of 1.1% for formamide, 19.3% for methanol, 19.6% for ethanol, and 2.4% for propanol. However, calculated values for intermediate concentrations compare well within the reported standard deviations given in Table 3.

Equations 9, 10, 13, 14, 17, and 18 (included in Table 3) make it possible to predict the densities and viscosities of these binary systems at any composition and temperature, in the temperature range (25 to 50) °C, since we have included in these correlations the experimental results of García et al.[7] measured at 25 °C. To include this last data set in the correlations, we interpolated their raw values of density and viscosity to our mole fractions reported in Table 2.

Table 2. Experimental Density (ρ) and Viscosity (η) Values at Several Temperatures for Formamide (1) + Alkan-1-ols (2) Binary Systems

x_1	ρ/(kg m^{-3})						η/(mPa s)					
	25 °C[a]	30 °C	35 °C	40 °C	45 °C	50 °C	25 °C[a]	30 °C	35 °C	40 °C	45 °C	50 °C
						(a) [(x_1)Formamide + (1 − x_1)Methanol]						
0.0000	786.6	781.9	777.2	772.5	767.5	762.7	0.50	0.46	0.44	0.41	0.41	0.40
0.0714	814.8	810.2	806.4	801.8	796.9	792.2	0.63	0.55	0.51	0.48	0.46	0.42
0.1586	847.7	843.7	839.1	834.6	830.0	825.4	0.74	0.63	0.59	0.54	0.51	0.47
0.2422	878.6	874.3	869.8	865.7	860.7	856.2	0.86	0.74	0.68	0.63	0.58	0.54
0.3200	906.8	902.6	898.2	893.8	889.2	884.8	0.99	0.85	0.78	0.72	0.67	0.60
0.3836	929.4	926.1	921.8	917.4	912.9	908.7	1.12	0.95	0.87	0.80	0.73	0.66
0.5273	979.1	974.7	970.5	966.2	961.7	957.4	1.45	1.2	1.10	1.02	0.92	0.80
0.5975	1002.7	997.9	993.7	989.5	985.0	980.8	1.65	1.4	1.2	1.1	1.01	0.86
0.6822	1030.5	1026.4	1022.2	1018.0	1013.5	1009.4	1.93	1.6	1.4	1.3	1.1	0.96
0.7619	1056.1	1051.8	1047.6	1043.4	1038.9	1034.7	2.22	1.8	1.6	1.4	1.3	1.07
0.8279	1076.9	1071.9	1067.6	1063.5	1059.1	1054.9	2.49	2.0	1.8	1.6	1.4	1.2
0.9170	1104.3	1099.5	1095.3	1091.1	1086.7	1082.5	2.90	2.3	2.1	1.9	1.6	1.3
1.0000	1129.7	1124.7	1120.5	1116.4	1111.9	1107.8	3.33	2.8	2.5	2.2	2.0	1.6
						(b) [(x_1)Formamide + (1 − x_1)Ethanol]						
0.0000	786.6	780.8	776.5	772.2	767.6	763.2	1.09	0.95	0.87	0.79	0.72	0.63
0.0716	804.2	799.5	795.2	790.8	786.3	781.9	1.16	0.97	0.89	0.80	0.73	0.64
0.1365	821.2	817.0	812.7	808.5	803.9	799.6	1.24	1.03	0.93	0.85	0.77	0.68
0.2357	848.8	845.1	840.8	836.5	832.0	827.6	1.40	1.1	1.05	0.95	0.86	0.75
0.3142	872.1	868.5	864.2	859.9	855.4	851.2	1.54	1.3	1.1	1.04	0.94	0.83
0.3842	893.9	891.0	886.8	882.5	878.1	873.7	1.69	1.4	1.2	1.1	1.02	0.89
0.4597	918.5	914.8	910.6	906.4	902.0	897.7	1.86	1.5	1.3	1.2	1.10	0.93
0.5389	945.6	941.4	937.3	933.1	928.7	924.5	2.05	1.6	1.5	1.3	1.2	0.98
0.6090	970.7	966.2	962.0	957.9	953.5	949.4	2.23	1.8	1.5	1.4	1.2	1.04
0.6872	999.8	995.8	991.6	987.5	983.1	978.9	2.44	1.9	1.7	1.5	1.3	1.1
0.7741	1033.6	1028.8	1024.7	1020.6	1016.2	1012.0	2.68	2.1	1.8	1.7	1.5	1.2
0.7928	1041.1	1036.1	1031.9	1027.9	1023.4	1019.3	2.73	2.2	1.9	1.7	1.5	1.2
0.9260	1096.4	1091.8	1087.6	1083.5	1078.9	1074.6	3.11	2.6	2.3	2.0	1.8	1.5
1.0000	1129.7	1124.7	1120.5	1116.4	1111.9	1107.8	3.34	2.8	2.5	2.2	2.0	1.6
						(c) [(x_1)Formamide + (1 − x_1)1-Propanol]						
0.0000	799.5	795.6	791.5	787.5	783.2	779.0	1.95	1.6	1.4	1.3	1.1	0.92
0.0961	817.8	813.6	809.5	805.4	801.1	796.9	1.98	1.6	1.4	1.3	1.1	0.92
0.1755	834.3	829.9	825.7	821.7	817.3	813.2	2.07	1.7	1.5	1.3	1.1	0.95
0.2494	850.7	846.6	842.4	838.4	834.0	829.8	2.19	1.8	1.5	1.4	1.2	1.00
0.3192	867.4	863.3	859.2	855.1	850.8	846.7	2.33	1.9	1.6	1.5	1.3	1.05
0.3902	885.6	881.7	877.6	873.6	869.2	865.1	2.48	2.0	1.7	1.6	1.4	1.1
0.4629	906.0	902.5	898.4	894.3	890.0	885.9	2.64	2.1	1.8	1.7	1.5	1.2
0.5342	927.7	924.0	919.9	915.9	911.6	907.4	2.80	2.2	1.9	1.7	1.5	1.3
0.6140	954.4	950.4	946.3	942.2	938.0	933.8	2.96	2.4	2.1	1.9	1.6	1.3
0.6891	982.2	977.8	973.7	969.8	965.5	961.4	3.10	2.6	2.2	2.0	1.7	1.4
0.7654	1013.2	1008.4	1004.3	1000.3	996.0	991.9	3.21	2.7	2.3	2.1	1.8	1.5
0.8114	1033.3	1028.1	1024.3	1020.0	1015.8	1011.6	3.27	2.7	2.4	2.1	1.9	1.5
0.9210	1086.3	1081.3	1077.2	1073.1	1068.8	1064.6	3.34	2.8	2.5	2.2	1.9	1.6
1.0000	1129.7	1124.7	1120.5	1116.4	1111.9	1107.8	3.34	2.8	2.5	2.2	2.0	1.6

[a] Interpolated data from ref 7.

Table 3. Equations for Density, Viscosity, Excess Volume, and Viscosity Deviation as a Function of Temperature and Composition for the Formamide (1) + Alkan-1-ols (2) Binary Systems

system	eqs	eq no.	σ^a
formamide + methanol	ρ/(kg m^{-3}) = (811.1 + 382.3x_1 − 43x_1^2) + (−0.96 + 0.5x_1 − 0.8x_1^2 + 0.4x_1^3)t	9	0.5
	η/(mPa s) = [0.006 exp(−x_1/0.325)] exp[(1400 + 1400x_1)/T]	10	0.1
	V^E/(kg m^{-3}) = x_1(1 − x_1)[−18 × 10^{-7} − (20 × 10^{-9})t + [−5 × 10^{-7} − (1 × 10^{-9})t](1 − 2x_1) + [(−9 × 10^{-9})t](1 − 2x_1)^2]	11	2 × 10^{-8}
	$\Delta\eta$/(mPa s) = x_1(1 − x_1)[−3.39 + (48 × 10^{-3})t + [1.6 − (21 × 10^{-3})t](1 − 2x_1) + [−1.8 + (21 × 10^{-3})t](1 − 2x_1)^2]	12	0.01
formamide + ethanol	ρ/(kg m^{-3}) = (808.8 + 237x_1 + 104.6x_1^2) + (−0.91 + 0.44x_1 − 0.8x_1^2 + 0.41x_1^3)t	13	0.4
	η/(mPa s) = (0.00106 − 0.00257x_1 + 0.00179x_1^2) exp[(2000 + 2100x_1 − 1300x_1^2)/T]	14	0.3
	V^E/(kg m^{-3}) = x_1(1 − x_1)(−14 × 10^{-7} − (14 × 10^{-9})t)	15	2 × 10^{-8}
	$\Delta\eta$/(mPa s) = x_1(1 − x_1)[−2.09 + (28 × 10^{-3})t + (7 × 10^{-3})t(1 − 2x_1) + [−1.0 + (9 × 10^{-3})t](1 − 2x_1)^2]	16	0.01
formamide + 1-propanol	ρ/(kg m^{-3}) = (819.45 + 195.6x_1 + 29.1x_1^2 + 106.2x_1^3) + (−0.80 − 0.57x_1 + 2.5x_1^2 − 3.69x_1^3 + 1.7x_1^4)t	17	0.23
	η/(mPa s) = (0.00012 + 0.00016x_1) exp[(2900 − 100x_1)/T]	18	0.2
	V^E/(kg m^{-3}) = x_1(1 − x_1)[−66 × 10^{-8} − (9 × 10^{-9})t + [24 × 10^{-8} + (7 × 10^{-9})t](1 − 2x_1)]	19	1 × 10^{-8}
	$\Delta\eta$/(mPa s) = x_1(1 − x_1)[−0.40 + (44 × 10^{-4})t + [−2.8 + (44 × 10^{-3})t](1 − 2x_1) + [0.3 − (8 × 10^{-3})t](1 − 2x_1)^2]	20	0.01

[a] Units of σ: ρ/(kg m^{-3}); η/(mPa s); V^E/(m^3 mol^{-1}); Δη/(mPa s).

The excess molar volume (V^E) and viscosity deviation (Δη) were calculated using the following equations:

$$V^E/(\text{m}^3 \text{ mol}^{-1}) = [(x_1 M_1 + (1 - x_1)M_2)/\rho] - \left(x_1\frac{M_1}{\rho_1} + x_2\frac{M_2}{\rho_2}\right) \quad (3)$$

$$\Delta\eta/(\text{mPa s}) = \eta - \exp[x_1\eta_1 + (1 - x_1)\eta_2] \quad (4)$$

where x_1 is the mole fraction of formamide; M_1 and M_2 are the molar masses of formamide and the alkan-1-ols, respectively; ρ, ρ_1, and ρ_2 are the densities of the mixture, formamide, and the alkan-1-ols, respectively; and η, η_1, and η_2 are the viscosities of the mixture, formamide, and the alkan-1-ols, respectively.

The excess molar volume and viscosity deviation were fitted to these results by means of a Redlich−Kister type
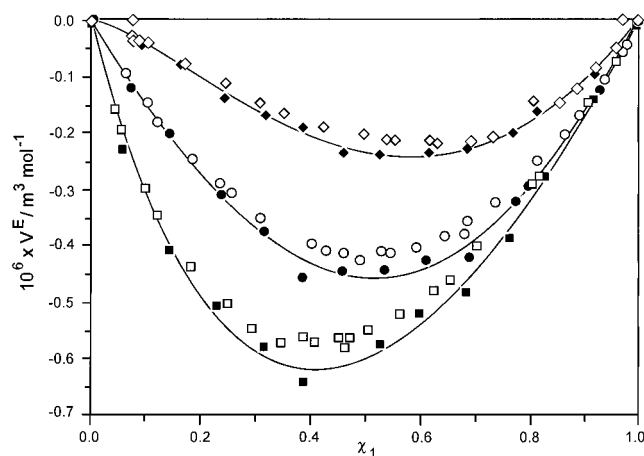
Figure 3. Excess molar volume for (■) formamide + methanol; (●) formamide + ethanol; and (◆) formamide + 1-propanol mixtures at 30 °C as a function of the mole fraction of formamide. Solid lines correspond to the least-squares fit using eq 7. Empty symbols correspond to values of García et al.[7] at 25 °C.

equation[14] with the same fitting procedure used for the density and viscosity:

$$Y^E = x_1(1 - x_1)\sum_{j=0}^{n} a_j(1 - 2x_1)^j \tag{5}$$

where Y^E represents either V^E or $\Delta\eta$.

Since the coefficients a_j are functions of temperature, they were plotted against this variable in order to obtain the equations that can represent each property in the studied temperature range. We propose the following dependence with temperature (t, in °C) for these coefficients:

$$a_j = \sum_{i=0}^{m} a_{ji}t^i \tag{6}$$

Using this temperature dependence, eq 5 can be rewritten as follows:

$$Y^E = x_1(1 - x_1)\sum_{j=0}^{n}\sum_{i=0}^{m} a_{ji}t^i(1 - 2x_1)^j \tag{7}$$

The parameters that best fitted this equation are summarized in Table 3, together with the standard deviation σ, defined as

$$\sigma = [\sum(Y^E_{\exp} - Y^E_{\text{calc}})^2/(N - p)]^{1/2} \tag{8}$$

where N and p are the numbers of experimental points and of parameters, respectively. The choice of the appropriate number of constants in eq 7 was based on the variation with n and m of the standard error of the estimated value. Their significant digits were determined taking into account each error coefficient.

Figures 3 and 4 show the excess molar volumes and the viscosity deviations plotted against the mole fraction of formamide for the formamide + methanol, formamide + ethanol, and formamide + 1-propanol systems at 30 °C, respectively. The data reported by García et al.[7] at 25 °C are also included in these plots for comparison. The characteristic behavior observed in these figures remains the same for the other temperatures, and consequently, their plots are not shown.
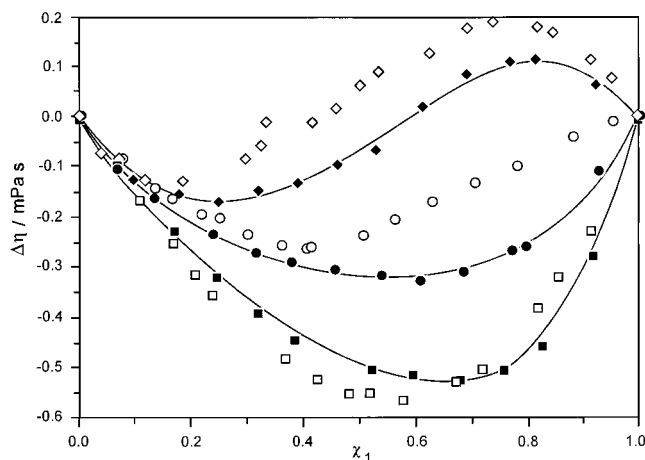
Figure 4. Viscosity deviation for (■) formamide + methanol; (●) formamide + ethanol; and (◆) formamide + 1-propanol mixtures at 30 °C as a function of the mole fraction of formamide. Solid lines correspond to the least-squares fit using eq 7. Empty symbols correspond to values of García et al.[7] at 25 °C that were calculated by these authors with the following equation: $\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2)$.

Figure 3 shows that excess molar volumes are always negative for the three systems at 30 °C and for any composition. From this plot we can conclude that greater negative V^E values are obtained when the chain length of the alkan-1-ols diminishes, which is in agreement with the conclusions obtained by García et al.[7] Additionally, when temperature is increased, the V^E values become more negative for the three systems. Figure 4 shows that the viscosity deviation is negative for formamide + methanol and formamide + ethanol systems, and sigmoid for the formamide + 1-propanol system, also in agreement with García et al.[7] This behavior remains unaltered for all the temperatures studied and for the three systems.

An increment of temperature diminishes the self-association in the pure components and also the heteroassociation formamide−alcohol in the mixtures, because of the increase of the thermal energy. This leads to more negative values of V^E and less negative ones of $\Delta\eta$, as observed.

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