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Kinematic Viscosity of Systems Containing Polyethylene Glycol + Salt + Water at 298.2 K

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Kinematic viscosities of binary, ternary, and quaternary aqueous solutions containing poly(ethylene glycol) (PEG) with nominal molecular mass equal to 1000 g·mol^{-1} and salts were determined at 298.2 K and various concentrations. The binary experimental data were used for adjusting the parameters of a Kumar-like equation. The adjusted parameters also allowed the viscosity prediction for multicomponent mixtures, with an overall deviation of 4.8%.

Introduction

Poly(ethylene glycol) (PEG) is a steady and low-toxicity molecule available in diverse molecular weights that presents properties that become possible with its application in the biomedical and biotechnological areas.[1,2] One of the most important characteristics of PEG is the formation of two-phase systems with aqueous solutions of other polymers or salts. Because aqueous two-phase systems (ATPS) are constituted mainly of water, they provide a favorable environment for biomolecules, cells, and cell particles.[3,4] Such systems are composed of two incompatible polymers, for example, dextran and PEG, or a polymer and a salt (generally phosphate or sulfate).[5,6] In comparison to the PEG/dextran systems, PEG/salt systems have the advantages of low cost and higher density and viscosity differences, speeding the process in continuous extractors.[7,8]

Equilibrium data for the systems of interest are necessary for designing liquid−liquid extractors. Many papers containing equilibrium diagrams are available in the literature for systems of polymer/polymer[5,9] and for systems of polymer/salt.[6,10−15] In addition to the knowledge of phase equilibrium, data on the properties of the phases, such as the viscosity, are also necessary for the design of ATPS extraction processes in large-scale applications.[7,8] Chenlo et al.[16] studied the kinematic viscosities of salt solutions at different concentrations and temperatures. Kinematic viscosities of PEGs, PEGs mixtures, and aqueous PEGs solutions were also determined by Cruz et al.[17] and Ninni et al.[18] at different concentrations, molecular masses, and temperatures. Because it is not viable to measure viscosities at all conditions of interest, methods for the estimation of viscosities at various temperatures, concentrations, and solute types are of great practical interest. Numerous empirical and semiempirical methods have been proposed to calculate the liquid viscosity of mixtures,[19−23] but some equations are inappropriate for concentrated and/or multicomponent mixtures.[18]

In the present work, the reformulated Kumar equation[21] was employed to correlate and predict kinematic viscosities of aqueous solutions containing PEG 1000 and different salts at 298.2 K. For each solute employed, parameters were estimated and compared with values available in the literature.[18,21] The parameters obtained allowed the kinematic viscosity predictions of ternary and quaternary mixtures of the solutes studied, presenting an overall deviation of 4.8%.

Experimental Section

Materials. Analytical-grade PEG with nominal molecular mass equal to 1000 g·mol^{-1} was purchased from Sigma (purity >99%) and used without further purification. The salts used were mono- and dibasic potassium phosphates, as well as mono- and dibasic sodium phosphates, and were purchased from Merck (purity >99.8%). Water was distilled, deionized with a Milli-Q water system (Millipore), and used to prepare the solutions. All reagents were characterized according to their water content determined by Karl Fischer titration using a Metrohm device (Switzerland). The values obtained were 0.68 mass % water for PEG 1000, 1.05 mass % and 0.52 mass % water for mono- and dibasic sodium phosphates, respectively, and 0.21 mass % and 1.54 mass % water for mono- and dibasic potassium phosphates, respectively. The water content of the polymer and salts samples was taken into account when calculating the solution concentrations.

Apparatus and Procedure. A set of experimental viscosity data for the binary systems NaH_2PO_4 + water, Na_2HPO_4 + water, and PEG 1000 + water, for the ternary systems PEG 1000 + NaH_2PO_4 + water, PEG 1000 + Na_2HPO_4 + water, PEG 1000 + KH_2PO_4 + water, and PEG 1000 + K_2HPO_4 + water, and for the quaternary system PEG 1000 + KH_2PO_4 + K_2HPO_4 + water were determined in an AMV 200 automatic viscosimeter (Anton Paar) connected to a thermostatic bath (Paar Physica model Viscotherm VT2) at (298.2 ± 0.1) K. The solutions were prepared on a mass basis using an analytical balance (ADAM, model AAA 250 L) accurate to ±0.0001 g. PEG concentrations in the solutions varied up to 40 mass %, and salt concentrations varied up to 30 mass %. The estimated error in the mass fractions was 2 in 10 000. The overall compositions of the ternary and quaternary solutions were selected on the basis of the equilibrium data obtained in the literature[6,24] in order to guarantee that the mixtures were in the one-phase region but close to the

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Table 1. Kinematic Viscosities, ν, of the Binary Systems at Various Mass Fractions, *w*, and 298.2 K

Na_2HPO_4 + water		NaH_2PO_4 + water		PEG 1000 + water	
w	$\nu/10^{-6}$ m² s⁻¹	w	$\nu/10^{-6}$ m² s⁻¹	w	$\nu/10^{-6}$ m² s⁻¹
0.0148	0.925	0.0318	0.957	0.0368	1.111
0.0276	0.964	0.0594	1.014	0.0695	1.357
0.0420	1.015	0.0896	1.088	0.0976	1.587
0.0545	1.064	0.1989	1.511	0.1324	1.963
0.0679	1.132	0.2997	2.305	0.1499	2.140
0.0987	1.307			0.1791	2.594
				0.2034	3.174
				0.2548	4.316
				0.3083	6.145
				0.3588	8.35
				0.4149	11.29

Table 2. Kinematic Viscosities, ν, of the Ternary Systems at Various Mass Fractions, *w*, and 298.2 K

PEG 1000 (1) + K_2HPO_4 (2) + water			PEG 1000 (1) + KH_2PO_4 (2) + water		
w_1	w_2	$\nu/10^{-6}$ m² s⁻¹	w_1	w_2	$\nu/10^{-6}$ m² s⁻¹
0.0085	0.1811	1.556	0.0238	0.1980	1.723
0.0184	0.1702	1.605	0.0330	0.1632	1.646
0.0345	0.1586	1.750	0.0395	0.1572	1.690
0.0443	0.1425	1.773	0.0609	0.1223	1.751
0.0607	0.1396	2.012	0.0871	0.0975	1.928
0.0822	0.1173	2.149	0.1090	0.0889	2.174
0.1005	0.1097	2.401	0.1363	0.0701	2.465
0.1185	0.0937	2.532	0.1647	0.0663	2.947
0.1371	0.0852	2.786	0.1925	0.0506	3.453
0.1588	0.0732	3.071	0.2228	0.0399	4.064
0.1906	0.0618	3.619	0.2658	0.0352	5.315
0.2190	0.0563	4.299	0.2930	0.0266	6.159
0.2416	0.0478	4.812			
0.2804	0.0383	5.973			
0.3140	0.0322	7.243			

PEG 1000 (1) + Na_2HPO_4 (2) + water			PEG 1000 (1) + NaH_2PO_4 (2) + water		
w_1	w_2	$\nu/10^{-6}$ m² s⁻¹	w_1	w_2	$\nu/10^{-6}$ m² s⁻¹
0.4036	0.0033	10.283	0.2958	0.0576	6.966
0.2919	0.0150	5.692	0.2782	0.0709	6.595
0.2550	0.0222	4.744	0.2363	0.0846	5.263
0.2288	0.0291	4.246	0.2184	0.0975	4.923
0.2174	0.0370	4.249	0.1924	0.1009	4.170
0.1885	0.0414	3.567	0.1612	0.1196	3.617
0.1709	0.0538	3.439	0.1510	0.1287	3.489
0.1495	0.0539	2.971	0.1312	0.1425	3.202
0.1260	0.0610	2.631	0.1049	0.1566	2.787
0.1117	0.0715	2.586	0.0737	0.1809	2.436
0.1026	0.0795	2.572	0.0671	0.1941	2.460
0.0826	0.0837	2.260	0.0435	0.2086	2.181
0.0615	0.0936	2.038	0.0179	0.2310	1.953
0.0461	0.1035	1.928	0.0035	0.2666	2.031
0.0323	0.1143	1.847			
0.0111	0.1384	1.837			
0.0064	0.1536	1.906			

corresponding binodal curves. The experiments were replicated at least 16 times. The standards deviations of the viscosity determinations varied within the range of 1.0×10^{-10} m²·s⁻¹ to 4.6×10^{-9} m²·s⁻¹, with the lowest numbers obtained for the lowest viscosity values. The variation coefficients changed within the range of 0.04 and 0.50% so that the uncertainty of the experimental measurements can be estimated as being not higher than 0.50%.

Results and Discussion

The kinematic viscosities for the binary, ternary, and quaternary systems determined in this work are given in Tables 1, 2, and 3, respectively.

Kumar's Equation for Viscosity Correlation in PEG Mixtures. The Kumar expression[25] is one of the equations presented in the literature for calculating the viscosities of solutions as a function of solute concentration. Initially, Kumar's expression was formulated to estimate dynamic viscosities in aqueous and nonaqueous salt solutions.

Table 3. Kinematic Viscosities, ν, of the Quaternary System at Various Mass Fractions, *w*, and 298.2 K

PEG 1000 (1) + K_2HPO_4 (2) + KH_2PO_4 (3) + water			
w_1	w_2	w_3	$\nu/10^{-6}$ m² s⁻¹
0.3291	0.0161	0.0093	7.368
0.2948	0.0181	0.0114	6.026
0.2661	0.0214	0.0119	4.996
0.2367	0.0255	0.0145	4.252
0.2040	0.0322	0.0177	3.541
0.1699	0.0345	0.0187	2.831
0.1391	0.0401	0.0229	2.368
0.1023	0.0519	0.0277	1.963
0.0861	0.0567	0.0315	1.759
0.0625	0.0717	0.0406	1.624
0.0484	0.0899	0.0504	1.604
0.0369	0.1133	0.0628	1.607
0.0282	0.1272	0.0730	1.584
0.0174	0.1425	0.0852	1.559

Lately, the Kumar expression was reformulated[21] to allow the calculation of the kinematic viscosities of aqueous solutions containing salts or organic solutes. The resulting equation is a function of the relative density between the solution and solvent, the mass fraction of solute, and the temperature. Pereira et al.[21] investigated the predictive capacity of the equation for ternary, quaternary, and quinary solutions.

Ninni et al.[18] utilized the reformulated Kumar equation to correlate and predict the kinematic viscosities of aqueous PEG mixtures for different molecular masses, concentrations, and temperatures. This same expression was used in this work to correlate and predict the kinematic viscosities of the aqueous mixtures of PEG and salts. The equations for binary and multicomponent systems are given as follows:

$$\nu_{\text{rel}} = \frac{1}{\rho_{\text{rel}}}\left[\frac{Bw}{1 - Hw}\right] \tag{1}$$

$$\nu_{\text{rel}} = \frac{1}{\rho_{\text{rel}}}\left[\frac{\sum_i B_i w_i}{1 - \sum_i H_i w_i}\right] \tag{2}$$

where the parameters B_i and H_i are interpreted, respectively, as a coefficient of solute−solvent interaction and the number of solvent molecules solvating the solute, both functions of temperature; w is the mass fraction of solute; ν_{rel} and ρ_{rel} are the ratios between the kinematic viscosities and densities of the solution and the corresponding values for the pure solvent, respectively; and i represents the solutes in the multicomponent mixture. The dependence of parameters B and H on temperature is given by polynomials in $(T - T_{\text{ref}})$ as follows:

$$B = B_{\text{ref}} + \sum_n B_n (T - T_{\text{ref}})^n \tag{3}$$

$$H = H_{\text{ref}} + \sum_n H_n (T - T_{\text{ref}})^n \tag{4}$$

where T_{ref} is the reference temperature equal to 298.2 K.

As can be seen, eqs 1 and 2 require information about the densities of pure water and the binary solutions. For this reason, linear equations were fit to experimental

Table 4. Linear Fittings for the Densities of Binary Systems at 298.2 K

	equation: ρ/kg m^{-3} = $a + bw$		
	a	b	R^2 [b]
PEG 1000[a]	995.70	176.30	3.76
Na_2HPO_4	996.56	977.59	5.80
NaH_2PO_4	994.36	818.23	3.40
K_2HPO_4	997.11	912.94	3.71
KH_2PO_4	998.52	746.95	4.80

[a] Experimental density data from ref 26. All other sets of density data were taken from refs 16 and 27. [b] Correlation coefficients such as 0.99998 given as 4.80.

density data of PEG in water[26] and of each salt in water[16,27] at 298.2 K. The coefficients of the linear equations are given in Table 4.

In the case of multicomponent systems, a semiempirical equation was used to calculate the corresponding densities. It requires the densities of the binary solutions (solute + solvent) and is given by

$$\rho_{\text{mix}} - \rho_{\text{w}} = \sum_{i,i \neq \text{w}} (\rho_{i\text{w}} - \rho_{\text{w}}) \tag{5}$$

where ρ_{mix}, ρ_w, and $\rho_{i\text{w}}$ represent the densities of the multicomponent mixture, of pure water, and of the binary aqueous mixtures containing component i, respectively. The summation in eq 5 should be performed on all components i in solution, except for water. Ninni and Meirelles[30] also used eq 5 to estimate the densities of aqueous solutions containing different solutes with very good agreement between experimental and calculated values (average absolute deviation around 0.30%). The applicability of this equation to systems containing polymer + salt + water has already been discussed in the literature. Zafarani-Moatar and co-workers[28,29] suggested the addition of a correction term in eq 5, which involves parameters that must be fit to experimental density data for the multicomponent systems. In these papers, the inclusion of the correction term allowed a further reduction of the average absolute deviation between experimental and predicted densities to values not higher than 0.06%. Unfortunately, experimental density data for the multicomponent systems studied in this work were not found in the literature, except for the quaternary system PEG 1000 + K_2HPO_4 + KH_2PO_4 + water.[28] In this case, eq 5 allows the density prediction for the quaternary system with a deviation of 0.33%. For these reasons, the predictive approach shown in eq 5 above was used to estimate the densities of the multicomponent mixtures. In fact, deviations around 0.50% are still very low and do not affect the prediction of viscosities in a significant way.

By nonlinear regression[31] of eq 1, the values of B and H were obtained for the binary systems PEG1000 + water, Na_2HPO_4 + water, and NaH_2PO_4 + water. Viscosity data for the binary systems K_2HPO_4 + water and KH_2PO_4 + water were taken from Chenlo et al.[16] Predictions of studied systems were also accomplished by using interaction parameters B and H taken from Pereira et al.[21] for all salt solutions (including K_2HPO_4 and KH_2PO_4) and from Ninni et al.[18] for PEG 1000 solutions.

Adjusted parameters B and H obtained in this work and from the literature[18,21] as well as the average absolute deviations (AAD) between experimental and calculated viscosities, estimated by eq 6, are presented in Table 5.

$$\%\text{AAD} = \left[\sum_{i=1}^{n}\left(\frac{|\nu_{\text{exptl},i} - \nu_{\text{calcd},i}|}{\nu_{\text{exptl},i}}\right)\right] \times \frac{100}{n} \tag{6}$$

where n is the number of experimental data of each system.

It is important to consider that, in this work, viscosity data for the systems containing K_2HPO_4 and KH_2PO_4 have not been determined. Therefore, the predicted viscosities for such systems were compared to the experimental data obtained by Chenlo et al.[16] These data[16] were also used by Pereira et al.[21] for the adjustment of parameters B and H.

Figure 1 presents the experimental and calculated kinematic viscosities determined in this work for the binary systems. Viscosity data calculated using parameters from the literature[18,21] are also represented in Figure 1.

As can be seen in Figure 1, there is a large difference between the polymer and salts viscosities, which is advantageous once it facilitates the phase separation in a liquid–

Table 5. Adjusted Parameters *B* and *H* and Average Absolute Deviations (AAD) for the Aqueous Systems Studied at 298.2 K

		parameters from this work			parameters from the literature			
	binary systems	B	H	AAD(%)[a]	B	H	AAD(%)[b]	n
correlation	K_2HPO_4				2.449	1.878	0.21	6
	KH_2PO_4				2.164	1.753	0.26	6
	Na_2HPO_4	3.6148	4.1870	0.31	4.250	3.319	1.64	6
	NaH_2PO_4	2.8525	2.0461	0.30	3.317	2.161	7.72	5
	PEG 1000	8.3017	1.7800	6.53	7.6304	1.6762	8.78	11
	global AAD(%)		*3.41 (n = 22)*			*4.35 (n = 34)*[d] *6.59 (n = 22)*[e]		

		AAD (%) for predictions		
	multicomponent systems	parameters from this work	parameters from the literature	n
prediction	PEG 1000 + K_2HPO_4	7.38[c]	12.86	15
	PEG 1000 + KH_2PO_4	4.21[c]	9.09	12
	PEG 1000 + NaH_2PO_4	1.64	7.60	14
	PEG 1000 + Na_2HPO_4	6.15	5.74	17
	PEG 1000 + K_2HPO_4 + KH_2PO_4	3.93	5.76	14
	global	*4.77*	*8.15*	*72*

[a] AAD values calculated considering the experimental data and parameters obtained from this work. [b] AAD values calculated considering the experimental data from the literature (for systems with K_2HPO_4 and KH_2PO_4)[16] and from this work (for the other binary systems) and parameters from the literature.[18,21] [c] Predictions accomplished using parameters from the literature for salts[21] and from this work for PEG 1000. [d] AAD values calculated considering all 34 experimental points. [e] AAD values calculated considering the experimental points for the binary systems with PEG and sodium phosphates salts.

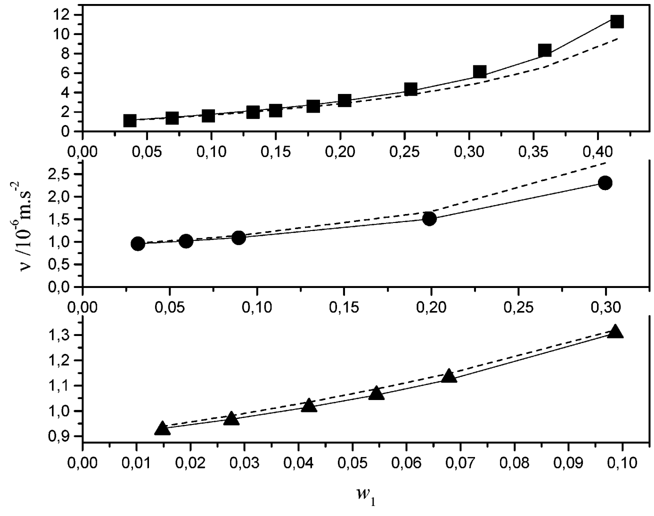

Figure 1. Experimental and calculated kinematic viscosities for binary aqueous systems: ■, PEG 1000; ●, NaH_2PO_4; ▲, Na_2HPO_4; (−) parameters from this work; (- - -) parameters from the literature.

liquid extraction process. In relation to the adjustments, Kumar's equation using the parameters obtained in this work promotes a good description of kinematic viscosities in the range of concentration studied. In fact, the AAD values obtained in this work were lower than the ones obtained using parameters from the literature. It can be explained by the restrict range of concentration used by Pereira et al.[21] for the adjustments of parameters B and H for the binary systems containing sodium phosphates salts ($w \le 0.07$ for Na_2HPO_4 and $w \le 0.13$ for NaH_2PO_4). Because mass fractions up to 0.30 were measured in this work for such systems, parameters B and H were adjusted for a higher range of concentration, providing a better description of the experimental data. Moreover, the average absolute deviations for the binary systems of PEG 1000 were significantly higher in comparison with the AAD values obtained for the salts. This was expected because Kumar's equation was not originally formulated for applications in polymer solutions.

Viscosity Prediction in Multicomponent Systems. The predictive capacity of eq 2 for estimating the viscosities of the ternary and quaternary aqueous systems was investigated. The values of viscosity calculated by eq 2 were compared with the experimental data from Tables 2 and 3. The average absolute deviations (AAD) for predictions are shown in Table 5.

Figures 2 and 3 show the experimental and predicted kinematic viscosities (ν_{exptl} and ν_{calcd}, respectively) calculated by eq 2. Predictions using parameters from the literature were compared to the predictions accomplished using parameters from this work. (In the case of systems containing potassium phosphates, predictions were made using parameters from this work for PEG 1000 and from the literature for the salts.) In all cases, we can observe the good agreement between experimental and predicted values, mainly for the low viscosity values, which corresponds to the experimental points with low PEG concentrations. It is important to emphasize that such AAD values obtained in this work are similar to those reported in the literature for the viscosity prediction of multicomponent mixtures (AAD values varied within the range of 2.7 to 20%).[18,32,33] However, predictions with good accuracy were obtained by Pereira et al.[25] employing the modified Kumar equation for estimating the viscosities of multicomponent mixtures. However, only low-molecular-mass solutes (salts, glycerin, monosaccharides, etc.) were employed at relatively low solute concentrations (up to 15 mass %).

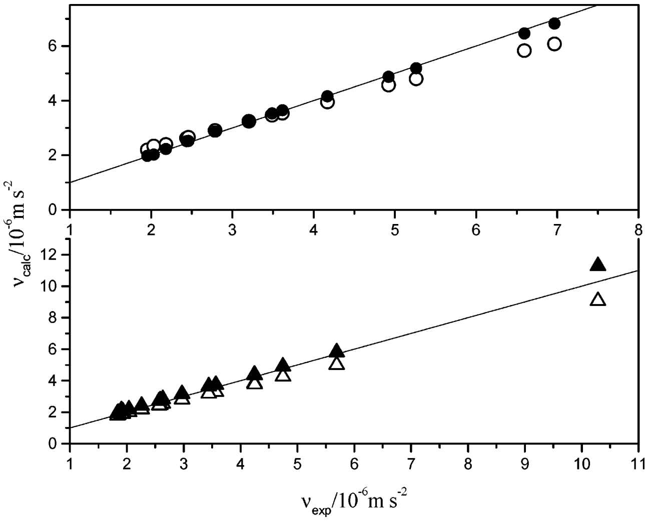

Figure 2. Experimental and calculated kinematic viscosities for ternary aqueous systems of PEG 1000 + NaH_2PO_4: ●, parameters from this work; ○, parameters from the literature.Experimental and calculated kinematic viscosities for ternary aqueous systems of and PEG 1000 + Na_2HPO_4: ▲, parameters of this work; △, parameters from literature.

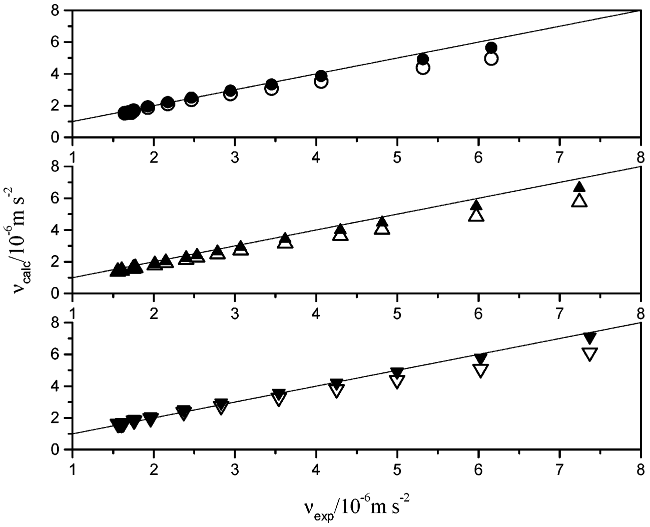

Figure 3. Experimental and calculated kinematic viscosities at 298.2 K for ternary and quaternary aqueous systems. Parameters from this work (solid symbols) and parameters from the literature (open symbols): ●, PEG 1000 + KH_2PO_4; ▲, PEG 1000 + K_2HPO_4; ▼, PEG 1000 + KH_2PO_4 + K_2HPO_4.

Conclusions

In general, the results obtained in this work, either in correlation or in prediction, were satisfactory. The parameters of the reformulated Kumar equation obtained by the correlation for binary systems allowed a good prediction of viscosity data for the ternary and quaternary systems studied. The AAD values were low in comparison with the ones reported in the literature. The approach suggested in the present work can help in the estimation of viscosity values for ATPS used in the purification of biomolecules.

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