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Density and Viscosity of Concentrated Aqueous Solutions of Polyethylene Glycol

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The densities and viscosities of concentrated aqueous solutions of polyethylene glycol (10–50 mass %) have been measured. The polyethylene glycol samples had average molecular masses of 8000, 3350, and 1000. The values of the density from 277 to 298 K show a linear variation with the polyethylene glycol concentration and differ, at most, by 10.07% from those of pure water at the same temperature. In order to estimate the values of dynamic viscosity, an equation is proposed which takes into account the influence of the polyethylene glycol concentration and the temperature in the range 277–313 K. The viscosity of mixtures of polyethylene glycol + magnesium sulfate + water were also studied. A linear relationship exists between the viscosities of the aqueous solutions of polyethylene glycol and the concentration of magnesium sulfate.

Introduction

The extraction of biomolecules using aqueous two-phase systems is important in so much as it allows the separation and purification of these substances in biocompatible surroundings (1, 2). To form the two aqueous phases, aqueous solutions of two polymers are required, usually polyethylene glycol (PEG) and dextran, or one of these polymers and a salt such as MgSO_4 , Na_2SO_4 , $(\text{NH}_4)_2\text{SO}_4$, or KH_2PO_4 . By placing PEG, H_2O , and one of these salts into contact, two phases are formed and an equilibrium concentration is reached which corresponds to the binodal curve.

Extraction using the two aqueous phases can be carried out in an apparatus where contact is continuous (packed columns) or discontinuous (separation by means of equilibrium stages). In both cases, it is essential to know the density and the rheological characteristics of the fluid phases in order to design the contactor.

The system studied in this paper is $\text{PEG} + \text{MgSO}_4 + \text{H}_2\text{O}$; the concentrations of PEG and MgSO_4 have been chosen so that they are included within the existence interval of the binodal curve, according to literature data (3, 4). For the choice of temperature variation, an interval has been chosen in which the denaturation of the biomolecules to be extracted would be negligible, between 277 and 313 K.

Density measurements have not been made by the majority of researchers due to the fact that its value practically coincides with that of water at the same temperature. Tawfik and Teja (5) study the influence of temperature for low-molecular-weight PEG, finding that, for the fitting and interpretation of the experimental data, Rackett's equation is applicable.

The influence of temperature, salt concentration, pH, anionic surfactants, and shear rate on the values of dynamic viscosity has been studied by various researchers on dilute solutions of PEG (6–8), where the equations of Huggins and Kraemer are applicable. However, there are few viscosity data at high PEG concentrations (9).

Materials and Methods

Materials. We have used polyethylene glycols with average molecular masses of 8000 (PEG 8000), 3350 (PEG 3350), and 1000 (PEG 1000), provided by Sigma Chemical Co. Extremely pure $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ (Panreac) has also been

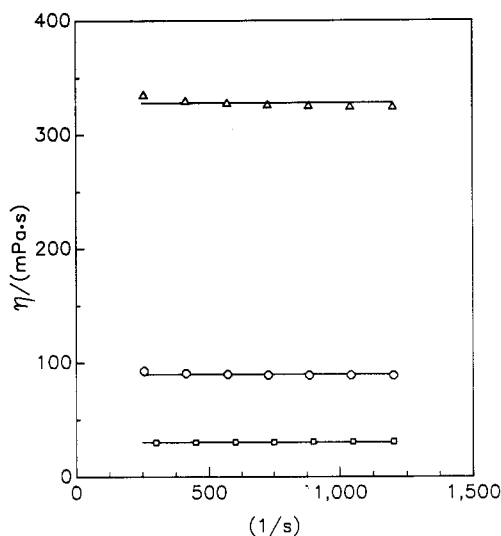


Figure 1. Influence of shear rates on the viscosity, η , for $w(\text{PEG})=0.40$ at 277 K: (Δ) PEG 8000; (\circ) PEG 3350; (\square) PEG 1000.

used. The PEG concentrations are situated between 10 and 50 mass % and those of MgSO_4 between 3 and 20 mass %. These concentrations are included within the existence interval of the binodal curve for the $\text{PEG} + \text{MgSO}_4 + \text{H}_2\text{O}$ system.

Apparatus and Procedures. The solutions were prepared by mass on an analytical balance with ± 0.1 -mg accuracy, using water of Milli-Q quality.

The density measurement is carried out using a 10 cm^3 pycnometer, at temperatures of 277 and 298 K. The temperature control was kept constant within ± 0.1 K.

To measure the dynamic viscosity (η), a rotating concentric cylinder viscometer, Contraves Rheomat 108 E/R, is used. The apparatus is completed with a thermostated bath, capable of maintaining the temperature to an accuracy of ± 0.1 K. The reproducibility of the viscosity data is estimated to be $\pm 1\%$.

In all the experiments, the viscosity is independent of the shear rate. For this reason, the average value of all the viscosity data obtained for shear rates included between 250 and 1250 s^{-1} has been taken. In Figure 1 the viscosities for

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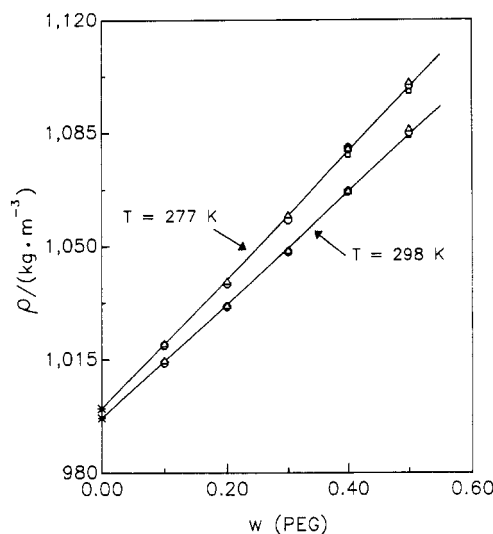


Figure 2. Variation of density with the PEG concentration: (Δ) PEG 8000; (○) PEG 3350; (□) PEG 1000; (*) water.

Table 1. Densities of Aqueous PEG Solutions

<i>w</i>	<i>T</i> /K	$\rho/(\text{kg}\cdot\text{m}^{-3})$		
		PEG 8000	PEG 3350	PEG 1000
0.50	277	1100.7	1099.5	1097.7
	298	1086.2	1084.9	1084.1
0.40	277	1080.1	1080.3	1078.0
	298	1067.1	1066.9	1066.5
0.30	277	1059.7	1058.0	1058.4
	298	1048.7	1048.2	1048.5
0.20	277	1039.2	1038.3	1038.2
	298	1031.7	1031.3	1031.2
0.10	277	1020.0	1019.4	1019.2
	298	1014.8	1014.0	1014.1
0	277	1000.0		
	298	997.07		

different shear rates obtained for 40 mass % PEG 8000, PEG 3350, and PEG 1000 at 277 K are shown.

Results and Discussion

Densities. The results obtained at 277 and 298 K are presented in Table 1 (along with the pure water densities from the literature) and are shown in Figure 2. The density values are practically independent of the molecular mass of the PEG and vary in a linear manner with its mass fraction, *w*.

$$\rho/(\text{kg}\cdot\text{m}^{-3}) = 1000.0 + 198.20w \quad T = 277 \text{ K} \quad (1)$$

$$\rho/(\text{kg}\cdot\text{m}^{-3}) = 997.07 + 174.41w \quad T = 298 \text{ K} \quad (2)$$

Equations 1 and 2 reproduce the results with maximum absolute deviations (MAD) of 0.15% and 0.18%, respectively.

Viscosities. Influence of the PEG Concentration and Temperature. In Table 2 the viscosity results are presented. In all cases the viscosity increases with molecular mass and PEG concentration and decreases with temperature.

To analyze the influence of temperature on η , the equation proposed by Lee and Teja has been used (10):

$$\eta/(\text{mPa}\cdot\text{s}) = A \exp\{B/(T/K - C)\} \quad (3)$$

where *A*, *B*, and *C* are the three parameters of the model and *T* is the temperature. By nonlinear regression of eq 1, the values of *A*, *B*, and *C* are obtained, which are shown in Table 3 and correspond to different PEG 3350 concentrations. Given

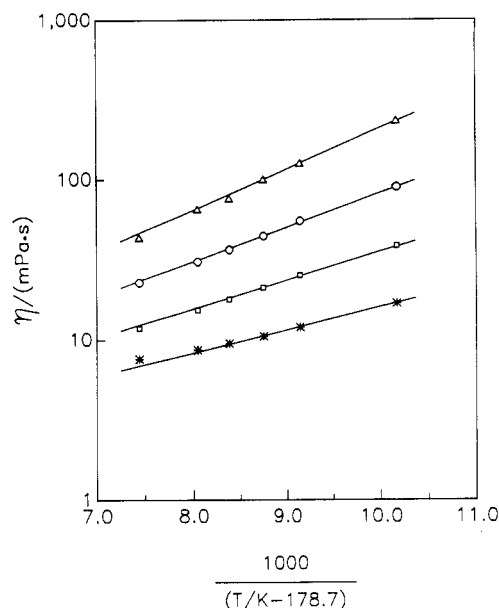


Figure 3. Lee and Teja (10) equation test for PEG 3350: (Δ) *w*=0.50; (○) *w*=0.40 (□) *w*=0.30; (*) *w*=0.20.

Table 2. Viscosities of Aqueous PEG Solutions

<i>w</i>	<i>T</i> /K	$\eta/(\text{mPa}\cdot\text{s})$		
		PEG 8000	PEG 3350	PEG 1000
0.50	277	968.1	232.3	65.0
	288	515.5	126.2	
	293		99.9	
	298	325.5	76.5	24.3
	303		65.3	
0.40	313	181.6	43.5	
	277	328.0	89.7	30.3
	288		55.0	
	293		44.3	
	298	126.5	36.4	13.2
0.30	303		30.7	
	313		22.7	
	277	108.4	38.4	15.5
	288		25.2	
	293		21.1	
0.20	298	50.9	17.9	9.2
	303		15.3	
	313		11.8	
	277	38.5	16.8	8.6
	288		12.0	
0.10	293		10.5	
	298	20.2	9.5	6.7
	303		8.6	
	313		7.6	
	277	12.7	7.9	6.7
	288	9.7		
	298	8.9	6.1	5.2
	313	7.4		

Table 3. Eq 3 Parameter Values for PEG 3350

<i>w</i>	<i>A</i>	<i>B</i>	<i>C</i>	<i>B</i> _{cal}
0.50	0.5445	585.0	180.4	586.5
0.40	0.5931	487.3	180.0	493.7
0.30	0.5723	396.3	182.9	409.2
0.20	0.6569	340.4	171.4	330.2

that the values of *A* and *C* seem to oscillate around an average value (*A* = 0.5917 ± 0.0414 and *C* = 178.7 ± 4.3), these values have been taken in order to recalculate *B* (*B*_{cal}). In Figure 3, the values of η calculated from eq 3 are shown, along with the proposed modifications for the three parameters. Equation 3 reproduces the experimental results with a MAD of 8.1% and an average absolute deviation (AAD) of 2.2%. Hence, only *B* seems to be influenced by the PEG concentration. Of the tested relations, Figure 4, that which best

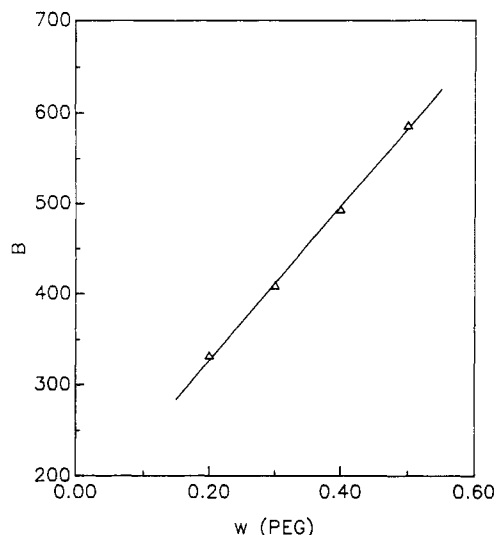


Figure 4. Variation of B , eq 3, with the PEG 3350 concentration.

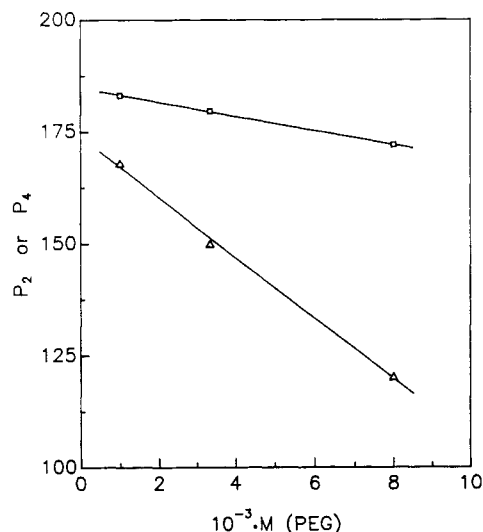


Figure 5. Variation of P_2 and P_4 , eq 5, with the PEG molecular mass: (Δ) P_2 ; (\square) P_4 .

Table 4. Eq 5 Parameter Values

PEG	P_1	P_2	P_3	P_4
8000	1.3311	120.2	1143.0	172.0
3350	0.5265	150.0	886.1	180.0
1000	0.3571	168.0	637.8	183.1

reproduces our results is

$$B = 156.2 + 853.3w \quad r^2 = 0.9987 \quad (4)$$

Combining eqs 3 and 4 to obtain the influence of temperature and concentration on η gives

$$\eta/(\text{mPa}\cdot\text{s}) = P_1 \exp\left(\frac{P_2 + P_3w}{T/K - P_4}\right) \quad (5)$$

where P_1 , P_2 , P_3 , and P_4 are the parameters of the model. Admitting that eq 3 is applicable to the remaining PEGs (PEG 8000 and PEG 1000), the results presented in Table 4 have been calculated by nonlinear regression.

With these values of P_1 , P_2 , P_3 , and P_4 , the experimental results of $\eta \geq 10 \text{ mPa}\cdot\text{s}$ with an AAD of 3.5% and a MAD of $\pm 10\%$ are reproduced. However, in the case of $\eta < 10 \text{ mPa}\cdot\text{s}$, the AAD is 17.5%.

In Figures 5 and 6, the variation of P_1 , P_2 , P_3 , and P_4 with the molecular mass, M , of the PEG is shown. In Figure 5 P_2

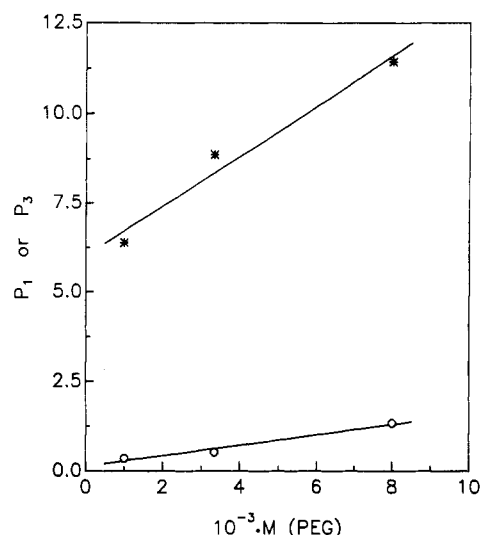


Figure 6. Variation of P_1 and P_3 , eq 5, with the PEG molecular mass: (\circ) P_1 ; ($*$) $10^{-2}P_3$.

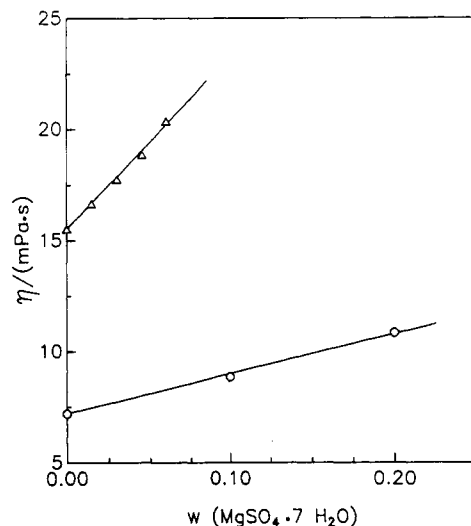


Figure 7. Variation of viscosity with the concentration of $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ for PEG 1000 at 277 K: (Δ) $w = 0.30$; (\circ) $w = 0.10$.

Table 5. Influence of the Concentration of $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$

PEG	T/K	$w(\text{PEG})$	$w(\text{MgSO}_4 \cdot 7\text{H}_2\text{O})$	$\eta/(\text{mPa}\cdot\text{s})$
8000	277	0.30	0	108.4
			0.015	115.2
			0.030	121.2
	298	0.30	0	50.9
			0.015	52.8
			0.030	55.0
1000	277	0.30	0	15.5
			0.015	16.7
			0.045	18.9
	0.30	0.10	0.030	17.8
			0.060	20.3
			0.10	7.2
			0.10	8.9
			0.200	10.9

and P_4 decrease with the molecular mass; however, in Figure 6, P_1 and P_3 increase with the molecular mass. Although these relations are approximate, they can be useful for the estimation of the viscosity corresponding to PEG with molecular masses which differ from those measured in this work.

Influence of Salt Concentration. The viscosity values in the homogeneity zone of the binodal curve have been determined with the aim of calculating the increase in the

viscosity as a consequence of a decrease in water concentration. The results obtained are shown in Table 5. A linear relationship exists between the viscosity and the concentration of MgSO_4 at both temperatures. In Figure 7, the results are shown for PEG 1000.

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