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Evaluation of viscosities of polymer–water solutions used in aqueous two-phase systems

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Abstract

The dynamic viscosities of dilute aqueous poly(ethylene glycol) and dextran, and poly(ethylene glycol)–dextran–water solutions have been measured. The poly(ethylene glycol) and dextran samples had average molecular masses of 8000 Da and 580 000 Da, respectively. To estimate the values of viscosity of poly(ethylene glycol)–dextran–water solutions, a Grunberg like equation has been proposed which takes into account the influence of poly(ethylene glycol) and dextran concentrations. The relative errors vary between 0.76 and 11.64 in absolute value.

Keywords: Aqueous two-phase system; Viscosity; Poly(ethylene glycol); Dextran

1. Introduction

The extraction of biomolecules using aqueous two-phase systems (ATPS) is important in so much as it allows the separation and purification of these substances in biocompatible surroundings [1]. To form the two aqueous phases, aqueous solutions of two polymers are required, usually poly(ethylene glycol) (PEG) and dextran.

Extraction using the two aqueous phases can be carried out in an apparatus where contact is continuous or discontinuous. The phase viscosities are important to design the contactor used in industrial-scale applications where large volumes of phases must be efficiently handled and separated.

In this study we consider the PEG–dextran–water system. The concentrations of PEG and dextran have been chosen so that they are below the phase separation. In the survey of mixture viscosity equations, most equations are in the form

$$f(\eta) = x_1 f(\eta_1) + x_2 f(\eta_2) \quad (1)$$

where η_1 and η_2 are the viscosities of the two pure components, and x_1 and x_2 can be weight, mole, or volume fractions of components 1 and 2, and $f(\eta)$ is η or $\ln \eta$. Eq. 1 has no disposable parameter.

In this work we propose a similar model to correlate PEG–dextran–water mixture viscosities by considering an equation of the following form

$$\ln \eta_{\text{mixture}} = p(c_1, c_2) \ln \eta_1 + q(c_1, c_2) \ln \eta_2 + ah(c_1, c_2) \quad (2)$$

where η_1 and η_2 are, respectively, the dynamic viscosities of the PEG–water solution and dextran–water solutions, a is a disposable parameter, c_1 and c_2 are, respectively, the weight percentages of PEG and dextran in solution, and p , q , and h are functions to be determined.

Since $\eta_{\text{mixture}}(c_1, c_2)$ should satisfy

$$\begin{aligned}\eta_{\text{mixture}}(0, c_2) &= \eta_2(c_2), \\ \eta_{\text{mixture}}(c_1, 0) &= \eta_1(c_1),\end{aligned}\quad (3)$$

we choose p , q , and h so that

$$p(c_1, 0) = 1, \quad q(c_1, 0) = 0, \quad h(c_1, 0) = 0 \quad (4)$$

$$p(0, c_2) = 0, \quad q(0, c_2) = 1, \quad h(0, c_2) = 0. \quad (5)$$

We may therefore propose the equation

$$\begin{aligned}\ln \eta_{\text{mixture}} &= \frac{c_1}{c_1 + c_2} \ln \eta_1 + \frac{c_2}{c_1 + c_2} \ln \eta_2 \\ &+ a \frac{c_1}{c_1 + c_2} \frac{c_2}{c_1 + c_2}\end{aligned}\quad (6)$$

which satisfies the conditions (3), (4), (5).

2. Experimental

2.1. Materials

Dextran 500 (molecular mass 580 000 Da) and PEG 8000 (molecular mass 8000 Da) were purchased from Sigma (St. Louis, MO, USA). The PEG and dextran concentrations were between 1.0 and 3.5% by weight for PEG–water and dextran–water solutions, and for mixtures of PEG–dextran–water equal concentrations of polymers in the same range were used.

2.2. Apparatus

To measure the dynamic viscosity (η), a rotating concentric cylinder viscometer, Haake Rotovisco RV

3, was used. The apparatus was completed with a Haake circulator bath, capable of maintaining the temperature of tempering vessel to an accuracy of $\pm 0.4^\circ\text{C}$. The temperature was held at about 10°C .

3. Results and discussion

The viscosity data of aqueous PEG and aqueous dextran solutions measured in this work were correlated by using the following equation

$$\ln \eta = P_1 \exp\left(\frac{P_2 + P_3 c}{T(K) - P_4}\right) \quad (7)$$

where η is in mPa s, c is the concentration of the polymer by weight (% w/w) and T is in Kelvin. P_1 , P_2 , P_3 and P_4 are fitted constants for each substance, and their values determined by non-linear regression are given in Table 1.

We define uncertainty as

$$e_i = \left(\frac{\eta_i^{\text{expl}} - \eta^{\text{calc}}}{\eta^{\text{expl}}}\right) 100 \quad (8)$$

where η^{expl} is the viscosity measured and η^{calc} is the viscosity calculated.

Table 2
Comparison of experimental and correlated viscosities calculated using Eq. 7

Concentration (%, w/w)	η^{expl}	η^{calc}	e_i
<i>PEG</i>			
1.0	1.40	1.34	4.29
1.5	1.46	1.48	-1.37
2.0	1.58	1.62	-2.53
2.5	1.78	1.79	-0.56
3.0	1.95	1.97	-1.03
3.5	2.21	2.17	1.81
<i>Dextran</i>			
1.0	1.72	1.75	-1.74
1.5	2.17	2.12	2.30
2.0	2.47	2.54	-2.83
2.5	3.13	3.07	1.82
3.0	3.69	3.70	-0.27
3.5	4.45	4.46	-0.22

Table 1
Best-fit parameterization from correlation based on Eq. 7

	PEG–water solution	Dextran–water solution
P_1	0.80	0.64
P_2	50.17	92.00
P_3	30.06	54.17
P_4	127.88	138.00

Table 3
Comparison of experimental and correlated viscosities calculated using Eq. 6

PEG concentration (% w/w)	Dextran concentration (% w/w)	η^{expl}	η^{calc}	e_i
1.0	1.0	2.39	2.61	-9.20
1.5	1.5	2.84	3.00	-5.63
2.0	2.0	3.19	3.33	-4.39
2.5	2.5	3.95	3.98	-0.76
3.0	3.0	4.96	4.52	8.87
3.5	3.5	5.99	5.29	11.64

Eq. 7 represents the experimental results of polymer solutions very well with a maximum uncertainty of at most 4.29%. The same model equation also works well for more concentrated aqueous PEG solutions [2,3] which is in a good agreement with our observation. Table 2 shows the experimental and

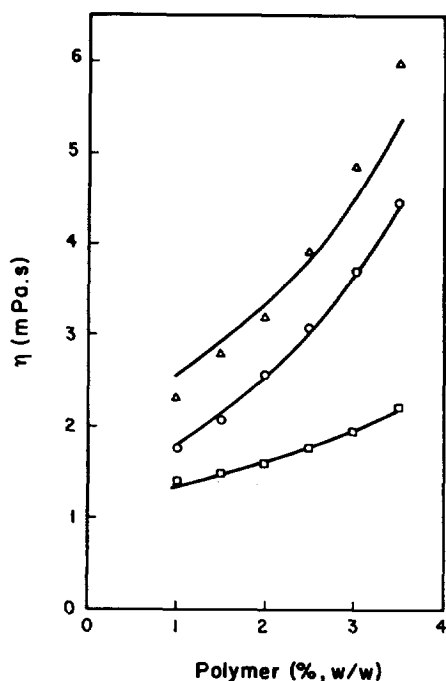


Fig. 1. Dynamic viscosity of: (□) PEG 8000, (○) dextran 500, and (Δ) a mixture of equal concentrations of dextran 500 and PEG 8000 in water ($T = 10 \pm 0.4^\circ\text{C}$). Correlated viscosities are given by the solid lines.

calculated viscosities, and the uncertainties defined in Eq. 8.

Eq. 6, which we use for the viscosity correlation of the PEG–dextran–water solution, resembles the equation

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + 2Gx_1x_2 \quad (9)$$

given by Grunberg and Nissan [5]. Here x_1 and x_2 are the mole fractions of components 1 and 2, and G is a parameter. Eq. 9 has been successfully applied to binary liquid mixtures. One can easily see that, by setting $w_1 = c_1/(c_1 + c_2)$ and $w_2 = c_2/(c_1 + c_2)$, Eq. 6 reduces to

$$\ln \eta_{\text{mixture}} = w_1 \ln \eta_1 + w_2 \ln \eta_2 + aw_1w_2. \quad (10)$$

The parameter a in Eq. 10 has been estimated as 2.0904 by minimizing the function

$$S^2 = \sum_{i=1}^6 (\eta^{\text{expl}} - \eta^{\text{calc}})^2. \quad (11)$$

We have observed that the viscosity of a mixture of dextran and PEG is higher than the sum of the increments of the separate polymers (Fig. 1). This is probably because polymer molecules in the mixture have a greater opportunity for interaction, resulting in more disturbances of the flow. Johansson et al. [4] observed the same phenomena for the same polymer mixture at 22°C .

Table 3 displays the experimental and calculated viscosities of PEG–dextran–water solutions. It is seen that the uncertainties vary between 0.76 and 11.64 in absolute value. The agreement between calculated and experimental values is demonstrated in Fig. 1.

A similar analysis may be carried out to study biphasic mixtures. Further results in this direction will be reported soon for ATPS with polymers having different molecular masses.

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