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Viscosity prediction of polyethylene glycol-dextran-water solutions used in aqueous two-phase systems

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Abstract

The dynamic viscosities of aqueous polyethylene glycol and dextran, and poly(ethylene glycol)–dextran–water solutions were measured at temperatures of 30, 50 and 70°C. The poly(ethylene glycol) having a relative molecular mass of 8000, and dextran samples with relative molecular masses of 37 500, 494 000, and 2 000 000 were used. A one-parameter Grunberg-like equation proposed earlier by us was used for estimating the values of viscosity of poly(ethylene glycol)–dextran–water solutions. The disposable parameter *a* for this temperature range was calculated as 1.81 for PEG 8000–dextran 37 500–water solutions, as 2.36 for PEG 8000–dextran 494 000–water solutions, and as 2.57 for PEG 8000–dextran 2 000 000–water solutions. It was observed that the relative errors get larger as the relative molecular mass of the dextran increases but vary between 0.00 and 9.37 in absolute value. In view of the results obtained here and before, we may claim that the proposed model works constantly well at different temperatures giving comparable values for the disposable parameter *a*. © 2000 Elsevier Science B.V. All rights reserved.

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

1. Introduction

The extraction of biomolecules using aqueous two-phase systems is important since it allows the separation and purification of these substances in biocompatible surroundings. Aqueous solutions of two polymers, usually poly(ethylene glycol) (PEG) and dextran, are required to form the two aqueous phases. The main advantages of using such systems, see [1], may be summarized as follows:

- Scale-up can be predicted easily and reliably from small laboratory experiments
- Rapid mass transfer and equilibrium is reached by relatively little input of energy in the form of mechanical mixing
- Continuous processing is readily achievable
- The polymers stabilize the enzymes

- Separation can be made selective and rapid
- Separation can be carried out at room temperature due to the rapid separation
- It has proven to be more economical than other separation processes.

Extraction using the two aqueous phases can be carried out in an apparatus where contact is continuous or discontinuous. The phase viscosities are important in designing the contactor used in industrial-scale applications where large volumes of phases must be efficiently handled and separated. Even though the viscosities of the phases are highly important, there have been only a few studies in the literature [2–5].

In Ref. [4], the present author first proposed a Grunberg-like equation [6] to correlate PEG 8000– dextran 580 000–water mixture viscosities, where

the experiments were made at temperature 10° C using equal concentrations of polymers in the range 1-3.5% (w/w).

The main objective of this work is to check the efficiency of the model used in [4] by considering polymers at different temperatures and varying relative molecular masses and concentrations.

The proposed model equation is of the following form

$$\ln \eta_{\text{mixture}} = c_1 \ln \eta_{\text{PEG}} + c_2 \ln \eta_{\text{Dx}} + c_1 c_2 a \tag{1}$$

where

$$c_1 = \frac{c_{\text{PEG}}}{c_{\text{PEG}} + c_{\text{Dx}}}, \quad c_2 = \frac{c_{\text{Dx}}}{c_{\text{PEG}} + c_{\text{Dx}}},$$

 $\eta_{\rm PEG}$ and $\eta_{\rm Dx}$ are, respectively, the dynamic viscosities of PEG-water and dextran-water solutions, $c_{\rm PEG}$ and $c_{\rm Dx}$ are, respectively, the weight percentages of PEG and dextran in solution, and *a* is a disposable parameter.

Derivation of Eq. (1) can be found in our earlier work [4]. We only remark here that Eq. (1) is tailored so as to satisfy $\eta_{\text{mixture}} = \eta_{\text{Dx}}$ if $c_{\text{PEG}} = 0$, $\eta_{\text{mixture}} = \eta_{\text{PEG}}$ if $c_{\text{Dx}} = 0$, and $c_1 + c_2 = 1$.

2. Experimental

2.1. Materials

Dextran samples with relative molecular masses of 37 500, 494 000, and 2 000 000, and PEG which has a relative molecular mass of 8000 were purchased from Sigma (St. Louis, MO, USA). The PEG and dextran concentrations were between 0.5 and 7.0% by weight for PEG–water and dextran–water solutions, and for mixtures of PEG–dextran–water different concentrations of polymers in water representing PEG and dextran compositions of homogeneous systems at temperatures of 30, 50 and 70°C.

2.2. Apparatus

A Canon-Fenske type viscometer was used to measure the relative viscosities (Greiner Scientific, NY, USA). Measurements were done at three different temperatures — 30, 50 and 70°C. The immersion heater (Greiner Scientific) used in this study was capable of maintaining the temperature of the water bath to an accuracy of $\pm 0.5^{\circ}$ C. The dynamic viscosities were then calculated using the dynamic viscosities of water at the specified temperatures [7].

3. Results and discussion

Our objective in this work was to check the applicability of Eq. (1) which we proposed earlier for the same polymer system (PEG-dextran). For our purpose we performed our experiments at 30, 50 and 70°C. We considered polymers at varying relative molecular masses and concentrations. Specifically, we used PEG 8000-dextran 37 500-water, PEG 8000-dextran 494 000-water, and PEG 8000-dextran 2 000 000-water solutions with polymer concentrations changing from 0.5 to 7% (w/w). The proposed model gives quite satisfactory results in all cases. We had reported the value of a as 2.09 at 10° C for PEG 8000-dextran 580 000-water solution in our earlier work. Here a is obtained as 1.81, 2.36 and 2.57 for PEG-dextran-water solutions at the different relative molecular masses stated and at temperatures of 30, 50 and 70°C. These values are tabulated in Table 1. The value of the disposable parameter a increases as the relative molecular mass of dextran increases.

Tables 2–4 show that Eq. (1) gives satisfactory results for all polymer solutions studied, and since there is hardly any change in parameter a at the different temperatures values of a in Table 1 are not shown.

The parameter a in Eq. (1) was estimated by using regressional analysis via SPSS.

The uncertainty e_i that appears in Tables 2–4 is defined by

$$e_{i} = \left(\frac{\eta_{i}^{\text{expl}} - \eta^{\text{calc}}}{\eta^{\text{expl}}}\right) \cdot 100$$
(2)

where η^{expl} and η^{calc} , (mPa s), are the measured

Table 1 Disposable parameter a in Eq. (1)

Mixture	а
PEG 8000-dextran 37 500-water	1.81
PEG 8000-dextran 494 000-water	2.36
PEG 8000-dextran 2 000 000-water	2.57

viscosities and the calculated viscosities, respective-ly.

Table 2 displays the experimental and calculated viscosities of PEG 8000–dextran 37 500–water solutions, where it is seen that the uncertainties vary between 0.00 and 6.33 in absolute value. The experimental and calculated viscosities of PEG

8000-dextran 494 000-water solutions are tabulated in Table 3. Here the uncertainties are between 0.00 and 8.54 in absolute value. The relative errors observed for PEG 8000-dextran 2 000 000-water solutions are given in Table 4 with a maximum absolute error of 9.37.

The effect of varying relative molecular masses of

Table 2

Comparison of experimental and correlated viscosities calculated using Eq. (1) for PEG (M_r 8000)-dextran (M_r 37 500)-water solution; viscosities are given in units of mPa s

PEG-water		Dextran-water		PEG-dextran-water		ei
c_{PEG} (%, w/w)	$\eta_{ m PEG}$	c_{Dx} (%, w/w)	$\eta_{ m Dx}$	$\eta_{_{ m mixture}}^{_{ m expl.}}$	$\eta_{_{ m mixture}}^{_{ m calc.}}$	
$T = 30^{\circ} \text{C}$						
5.0	1.33	0.5	0.56	1.38	1.43	-3.64
5.0	1.33	1.0	0.61	1.51	1.50	0.82
4.0	1.12	1.5	0.66	1.42	1.39	1.87
4.0	1.12	2.0	0.72	1.48	1.45	2.55
3.0	0.94	2.5	0.78	1.39	1.35	2.58
3.0	0.94	3.0	0.85	1.46	1.40	4.27
2.0	0.77	3.5	0.93	1.36	1.32	2.63
2.0	0.77	4.0	1.01	1.44	1.38	3.89
1.0	0.64	4.5	1.10	1.34	1.30	2.76
1.0	0.64	5.0	1.18	1.43	1.37	3.63
0.5	0.58	6.0	1.39	1.52	1.47	3.06
0.5	0.58	7.0	1.63	1.75	1.70	3.05
$T = 50^{\circ} \text{C}$						
5.0	0.80	0.5	0.39	0.83	0.87	-4.65
5.0	0.80	1.0	0.42	0.98	0.92	5.77
4.0	0.69	1.5	0.46	0.91	0.88	2.22
4.0	0.69	2.0	0.50	0.94	0.92	1.86
3.0	0.58	2.5	0.55	0.88	0.88	0.00
3.0	0.58	3.0	0.58	0.91	0.91	0.00
2.0	0.49	3.5	0.61	0.86	0.85	0.40
2.0	0.49	4.0	0.67	0.91	0.90	1.84
1.0	0.41	4.5	0.75	0.86	0.88	-2.24
1.0	0.41	5.0	0.79	0.91	0.91	0.00
0.5	0.39	6.0	0.89	0.98	0.95	3.31
0.5	0.39	7.0	1.03	1.08	1.08	0.00
$T = 70^{\circ} \text{C}$						
5.0	0.51	0.5	0.27	0.53	0.56	-6.33
5.0	0.51	1.0	0.38	0.58	0.59	-1.90
4.0	0.44	1.5	0.32	0.56	0.58	-3.17
4.0	0.44	2.0	0.33	0.58	0.60	-2.74
3.0	0.38	2.5	0.35	0.54	0.57	-5.68
3.0	0.38	3.0	0.38	0.58	0.59	-3.11
2.0	0.33	3.5	0.41	0.54	0.57	-5.84
2.0	0.33	4.0	0.44	0.58	0.60	-4.17
1.0	0.28	4.5	0.47	0.54	0.56	-2.70
1.0	0.28	5.0	0.50	0.58	0.58	0.00
0.5	0.27	6.0	0.58	0.61	0.62	-1.44
0.5	0.27	7.0	0.65	0.69	0.69	0.88

PEG on the disposable parameter a while keeping the relative molecular mass of dextran fixed is a topic for future research. It would also be very interesting to check the validity of Eq. (1) for other polymer systems.

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Table 3

Comparison of experimental and correlated viscosities calculated using Eq. (1) for PEG (M_r 8000)-dextran (M_r 494 000)-water solution; viscosities are given in units of mPa s

PEG-water		Dextran-water		PEG-dextran-water		ei
c_{PEG} (%, w/w)	$\eta_{ m PEG}$	c_{Dx} (%, w/w)	$\eta_{ m Dx}$	$\eta_{_{ m mixture}}^{_{ m expl.}}$	$\eta_{_{ m mixture}}^{_{ m calc.}}$	
$T = 30^{\circ} \text{C}$						
5.0	1.33	0.5	0.68	1.41	1.52	-7.86
5.0	1.33	1.0	0.83	1.33	1.79	4.67
4.0	1.12	1.5	1.04	1.72	1.75	-1.91
4.0	1.12	2.0	1.31	2.18	1.99	8.54
3.0	0.94	2.5	1.61	2.14	2.16	-0.71
3.0	0.94	3.0	1.92	2.54	2.42	4.59
2.0	0.77	3.5	2.31	2.64	2.68	-1.32
2.0	0.77	4.0	2.72	3.09	3.02	2.35
1.0	0.64	4.5	3.22	3.28	3.41	-3.96
1.0	0.64	5.0	3.74	3.84	3.87	-0.72
0.5	0.58	6.0	4.83	4.68	4.85	-3.67
0.5	0.58	7.0	6.22	6.29	6.15	2.23
$T = 50^{\circ} \text{C}$						
5.0	0.80	0.5	0.45	0.94	0.92	1.84
5.0	0.80	1.0	0.54	1.11	1.04	6.32
4.0	0.69	1.5	0.67	1.11	1.09	1.52
4.0	0.69	2.0	0.84	1.32	1.25	5.70
3.0	0.58	2.5	1.02	1.32	1.35	-1.96
3.0	0.58	3.0	1.20	1.59	1.51	5.35
2.0	0.49	3.5	1.45	1.63	1.69	-3.52
2.0	0.49	4.0	1.60	1.92	1.82	5.10
1.0	0.41	4.5	2.06	2.04	2.18	-6.96
1.0	0.41	5.0	2.28	2.40	2.38	0.94
0.5	0.39	6.0	2.89	2.93	2.92	0.02
0.5	0.39	7.0	3.68	3.89	3.67	5.66
$T = 70^{\circ} \text{C}$						
5.0	0.51	0.5	0.31	0.58	0.59	-2.14
5.0	0.51	1.0	0.37	0.68	0.67	1.33
4.0	0.44	1.5	0.46	0.66	0.71	-7.76
4.0	0.44	2.0	0.55	0.82	0.80	2.34
3.0	0.38	2.5	0.67	0.84	0.88	-5.09
3.0	0.38	3.0	0.79	0.99	0.98	0.16
2.0	0.33	3.5	0.93	1.04	1.10	-5.93
2.0	0.33	4.0	1.08	1.20	1.23	-2.42
1.0	0.28	4.5	1.28	1.40	1.38	1.48
1.0	0.28	5.0	1.47	1.47	1.55	-5.28
0.5	0.27	6.0	1.88	1.78	1.92	-7.57
0.5	0.27	7.0	2.37	2.32	2.38	-2.36

Table 4

Comparison of experimental and correlated viscosities calculated using Eq. (1) for PEG (M_r 8000)-dextran (M_r 2 000 000)-water solution; viscosities are given in units of mPa s

PEG-water		Dextran-water		PEG-Dx-water		e _i
c_{PEG} (%, w/w)	$\eta_{ m PEG}$	c_{Dx} (%, w/w)	$\eta_{ m Dx}$	$\eta_{_{ m mixture}}^{_{ m expl.}}$	$\eta_{_{ m mixture}}^{_{ m calc.}}$	
$T = 30^{\circ}C$						
5.0	1.33	0.5	0.70	1.50	1.55	-3.28
5.0	1.33	1.0	0.93	1.69	1.79	-6.33
4.0	1.12	1.5	1.31	2.05	1.95	5.04
4.0	1.12	2.0	1.71	2.52	2.28	9.37
3.0	0.94	2.5	2.21	2.56	2.62	-2.36
3.0	0.94	3.0	2.75	3.02	3.06	-1.20
2.0	0.77	3.5	3.42	3.62	3.61	0.43
2.0	0.77	4.0	4.06	4.08	4.13	-1.24
1.0	0.64	4.5	4.88	5.15	4.95	3.97
1.0	0.64	5.0	5.66	5.75	5.63	2.12
0.5	0.58	6.0	7.47	7.52	7.37	2.02
0.5	0.58	7.0	10.11	9.95	9.81	1.42
$T = 50^{\circ} \text{C}$						
5.0	0.80	0.5	0.48	0.92	0.94	-2.34
5.0	0.80	1.0	0.63	1.10	1.10	0.00
4.0	0.69	1.5	0.86	1.22	1.22	0.00
4.0	0.69	2.0	1.09	1.45	1.42	1.82
3.0	0.58	2.5	1.38	1.59	1.62	-2.31
3.0	0.58	3.0	1.73	1.87	1.90	-1.55
2.0	0.49	3.5	2.05	2.14	2.21	-3.06
2.0	0.49	4.0	2.47	2.50	2.55	-2.07
1.0	0.41	4.5	2.95	2.96	3.02	-1.82
1.0	0.41	5.0	3.48	3.52	3.48	0.98
0.5	0.39	6.0	4.41	4.61	4.39	4.59
0.5	0.39	7.0	6.02	5.85	5.88	-0.56
$T = 70^{\circ} \text{C}$						
5.0	0.51	0.5	0.34	0.59	0.61	-2.75
5.0	0.51	1.0	0.45	0.74	0.71	3.79
4.0	0.44	1.5	0.53	0.82	0.77	5.49
4.0	0.44	2.0	0.71	0.96	0.91	4.69
3.0	0.38	2.5	0.85	1.01	1.04	-2.84
3.0	0.38	3.0	1.05	1.18	1.20	-2.25
2.0	0.33	3.5	1.28	1.37	1.42	-3.87
2.0	0.33	4.0	1.62	1.63	1.69	-3.75
1.0	0.28	4.5	1.82	1.84	1.90	-2.93
1.0	0.28	5.0	2.11	2.17	2.15	0.68
0.5	0.27	6.0	2.73	2.83	2.74	3.35
0.5	0.27	7.0	3.62	3.63	3.57	1.72

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