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Densities and Viscosities of Polyethylene Glycol + Salt + Water Systems at 20 °C

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Densities and viscosities of polyethylene glycol (PEG), K_3PO_4, or $(NH_4)_2SO_4$ aqueous single-phase and two-phase systems have been measured at 20 °C. The densities and viscosities were correlated with the compositions and the tie line lengths of the aqueous two-phase systems.

Introduction

Partitioning of biomaterials utilizing aqueous two-phase systems is a new and powerful separation technique with industrial potential. The technique has been applied in biochemistry, biology, and biochemical engineering to separate and purify biological products, biomaterials, cell organs, proteins, and enzymes from the complex mixtures in which they are produced (Albertsson, 1986; Walter et al., 1985). Despite the success of the aqueous two-phase separation technique, data on the properties of phase systems that are necessary for the design of extraction processes and for the development of models that predict phase partitioning are few (Kula et al., 1982).

In this work, a comprehensive set of densities and viscosities of polyethylene glycol (PEG) with various molecular weights and with varying salt concentrations in both the single- and two-phase region were measured at 20 °C. The salts used were potassium phosphate and ammonium sulfate.

Experimental Section

*1. **Materials.*** Polyethylene glycol (PEG) was obtained commercially (Shanghai Chemical Reagent Factory, P. R. China) with five average molecular weights of 1000 (990–1100), 2000 (1900–2200), 4000 (3000–4500), 6000 (5500–7500), and 20000 (18000–20000). The salts were anhydrous potassium phosphate and ammonium sulfate (analytical reagent grade, >99.0%) (Shanghai Chemical Reagent Factory, P. R. China). Distilled, deionized water was used in this research.

*2. **Apparatus and Procedures.*** Using an analytical balance accurate to ±0.1 mg, aqueous solutions were prepared by mass from a stock polymer solution and a stock solution in a 10 cm^3 centrifuge tube and brought into a water bath (20 ± 0.05) °C. The systems were stirred and mixed for 5 min, and then separated at (20 ± 0.05) °C in a centrifuge (4000*g*) for 5 min (Mei, 1988).

Viscosities of the aqueous PEG solutions and of the top phase of the aqueous two-phase systems were determined using an Ostwald viscometer at (20 ± 0.05) °C in a water bath. The flow time was on the order of 120–200 s (Stewede, 1986; Zheng, 1986). Viscosities were accurate to ±0.002 mPa·s. The viscosities of the bottom phase of the aqueous two-phase systems were nearly the same as the viscosity value of water and were not measured.

Table 1. Densities and Viscosities of PEG (1) + H_2O (2) at 20 °C

w_1	$\varrho/(kg{\cdot}m^{-3})$	$100(\varrho - \varrho_{cal})/\varrho$	$\eta/(mPa{\cdot}s)$	$100(\eta - \eta_{cal})/\eta$
		PEG 1000		
0.10	1020.6	−0.28	1.796	−0.90
0.20	1037.9	−0.14	3.374	1.45
0.30	1059.9	0.0	6.300	1.5
0.40	1073.3	0.04	12.015	1.97
0.50	1093.1	0.24	21.878	−2.52
		PEG 2000		
0.10	1020.0	−0.34	2.138	−0.36
0.20	1036.0	−0.33	4.568	0.33
0.30	1054.6	−0.12	9.781	2.20
0.40	1074.1	0.11	22.627	−2.22
0.50	1090.5	0.01		
		PEG 4000		
0.05	1012.9	−0.27	2.222	0.16
0.10	1021.7	−0.17	4.265	0.71
0.15	1030.5	−0.09	7.710	−2.64
0.20	1037.5	−0.19	13.123	0.06
0.25	1046.8	−0.10	19.750	1.68
		PEG 6000		
0.05	1013.7	−0.20	2.283	0.90
0.10	1021.7	−0.17	4.506	−1.88
0.15	1030.5	−0.09	8.074	−1.41
0.20	1039.3	0.01	13.870	0.39
0.25	1046.4	−0.10	20.893	1.92
		PEG 20000		
0.025	1008.7	−0.31	2.325	0.1
0.05	1021.4	−0.32	4.553	−0.49
0.075	1016.8	−0.27	9.043	1.39
0.10	1020.0	−0.34	14.537	−1.83
0.15	1029.8	−0.15	29.532	0.81
0.20	1038.2	−0.12		
0.25	1046.7	−0.09		

Densities of the single-phase PEG + salt + water solutions and of the top and bottom phases of the aqueous two-phase systems were determined using density bottles at (20 ± 0.05) °C in a water bath (Stewede, 1986). The volume of the density bottle was approximately 5 cm^3. Densities were accurate to ±0.2 $kg{\cdot}m^{-3}$.

Results and Discussion

The densities and viscosities of various PEG aqueous solutions and the densities of some PEG + salt + water solutions are given in Tables 1–3. The results in Table 1 are plotted in Figures 1–3. The viscosities generally increase with an increase in PEG concentrations or the average molecular weight of PEG. $\ln(\eta/\eta_0)$ values are correlated with PEG concentrations according to (Bill-

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Table 2. Densities of PEG 6000 (1) + H_2O (2) + K_3PO_4 (3) at 20 °C

	$w_1 = 0.00$		$w_1 = 0.05$		$w_1 = 0.10$		$w_1 = 0.20$	
w_3	$\varrho/(\mathrm{kg{\cdot}m^{-3}})$	$100(\varrho - \varrho_{cal})/\varrho$	$\varrho/(\mathrm{kg{\cdot}m^{-3}})$	$100(\varrho - \varrho_{cal})/\varrho$	$\varrho/(\mathrm{kg{\cdot}m^{-3}})$	$100(\varrho - \varrho_{cal})/\varrho$	$\varrho/(\mathrm{kg{\cdot}m^{-3}})$	$100(\varrho - \varrho_{cal})/\varrho$
0.0	1008.0		1017.3	0.16	1024.6	0.11	1037.3	−0.12
0.02							1054.7	−0.12
0.05	1050.4	0.0	1060.8	0.19	1066.6	−0.06	1082.7	−0.06
0.08							1111.3	−0.09
0.10	1097.0	0.05	1107.3	0.15	1116.6	0.16		
0.15	1147.8	0.09	1157.9	0.10	1168.0	0.09		
0.20	1201.8	0.01	1210.7	−0.17	1222.3	−0.14		
0.25	1260.9	−0.13	1272.7	−0.16	1287.6	0.05		

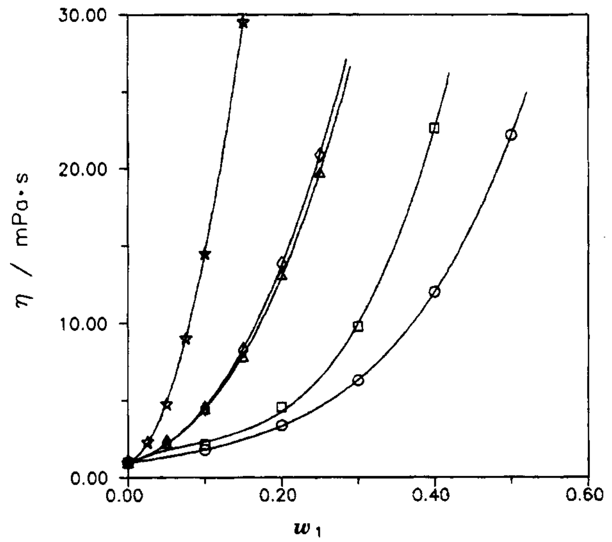

Figure 1. Relationship between the viscosities and mass fraction w_1 for PEG (1) + H_2O (2): (○) PEG 1000; (□ PEG 2000; (△) PEG 4000; (◇) PEG 6000; (☆) PEG 20000.

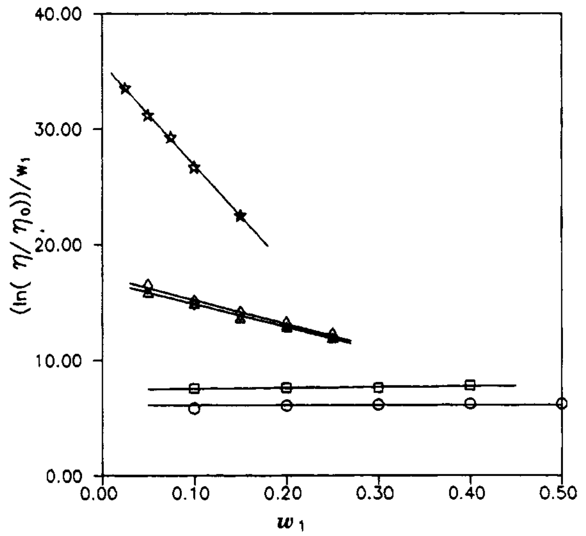

Figure 2. Relationship between $\ln(\eta/\eta_0)$ and mass fraction w_1 for PEG (1) + H_2O (2): (○) PEG 1000; (□ PEG 2000; (△) PEG 4000; (◇) PEG 6000; (☆) PEG 20000.

Table 3. Densities of PEG 6000 (1) + H_2O (2) + $(NH_4)_2SO_4$ (3) at 20 °C

	$w_1 = 0.00$		$w_1 = 0.10$	
w_3	$\varrho/(\mathrm{kg{\cdot}m^{-3}})$	$100(\varrho - \varrho_{cal})/\varrho$	$\varrho/(\mathrm{kg{\cdot}m^{-3}})$	$100(\varrho - \varrho_{cal})/\varrho$
0.0	1008.0		1024.8	0.13
0.05	1031.5	−0.15	1049.2	−0.01
0.10	1061.1	0.08	1077.7	0.12
0.15	1086.7	−0.03	1106.0	0.09
0.20	1116.4	0.03	1137.2	0.19
0.25	1144.9	−0.17	1165.5	−0.12

meyer, 1984)

$$\ln(\eta/\eta_0) = aw_1 + bw_1^{\,2} \qquad (1)$$

in which η is the absolute viscosity of the solution, w_1 is the mass fraction of PEG, and η_0 is the viscosity of water (in this paper, η_0 = 1.0045 mPa·s). The constants a and b are dependent on the average molecular weight of PEG as follows:

PEG 1000 $a = 5.816$ $b = 0.846$

PEG 2000 $a = 7.447$ $b = 0.712$

PEG 4000 $a = 16.845$ $b = -19.992$

PEG 6000 $a = 17.284$ $b = -20.887$

PEG 2000 $a = 35.740$ $b = -88.362$

The average relative deviation is about 1.2%. The densities of various aqueous PEG solutions are very close at the same concentration and independent of the molecular weight of PEG. The densities of aqueous PEG solutions may be correlated by using

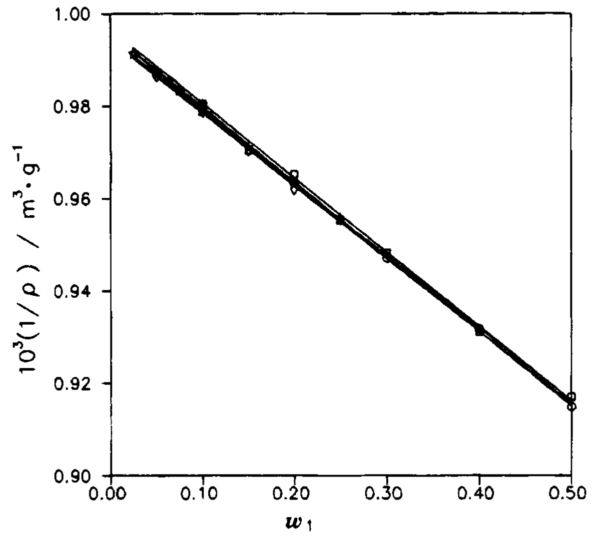

Figure 3. Relationship between the densities and mass fraction w_1 for PEG (1) + H_2O (2): (○) PEG 1000; (□ PEG 2000; (△) PEG 4000; (◇) PEG 6000; (☆) PEG 20000.

$$(1/\varrho)/(\mathrm{m^3{\cdot}kg^{-1}}) = (1/\varrho_0)/(\mathrm{m^3{\cdot}kg^{-1}}) - 1.5 \times 10^{-4} w_1 \qquad (2)$$

The data from Tables 2 and 3 are plotted in Figures 4 and 5. The densities of PEG + salt + H_2O systems could be

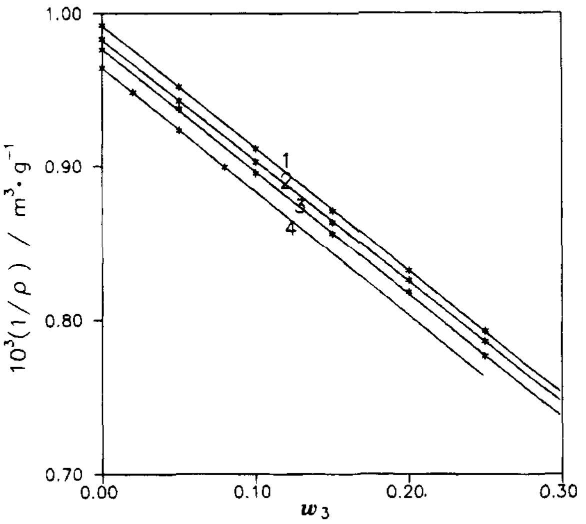

Figure 4. Relationship between the densities of PEG (1) + K_3PO_4 (3) + H_2O (2) and mass fraction w_3 of K_3PO_4 (1, w_1 = 0; 2, w_1 = 0.05; 3, w_1 = 0.10; 4, w_1 = 0.20).

Table 4. Densities and Viscosities of PEG (1) + H_2O (2) + $(NH_4)_2SO_4$ (3) Aqueous Two-Phase Systems at 20 °C

w_1	w_3	TLL	$\eta_{(T)}$/(mPa·s)	$\varrho_{(T)}$/(kg·m^{-3})	$\varrho_{(B)}$/(kg·m^{-3})	$\Delta\varrho$/(kg·m^{-3})
PEG 1000 + $(NH_4)_2SO_4$						
0.135	0.130	0.162	3.311	1093.7	1106.2	12.5
0.139	0.132	0.223	4.459	1091.0	1107.5	16.5
0.170	0.120	0.277	5.589	1090.5	1112.1	21.6
0.150	0.140	0.329	6.121	1088.9	1121.2	32.3
0.210	0.120	0.378	9.578	1089.5	1132.7	43.2
PEG 2000 + $(NH_4)_2SO_4$						
0.120	0.105	0.179	4.038	1082.2	1089.5	7.3
0.139	0.102	0.230	5.053	1081.4	1091.8	9.4
0.140	0.110	0.298	9.146	1078.4	1098.6	20.2
0.170	0.110	0.353	11.587	1082.1	1110.7	28.6
0.200	0.110	0.391	16.218	1083.8	1120.0	36.2
PEG 4000 + $(NH_4)_2SO_4$						
0.090	0.090	0.121	5.623	1067.6	1070.7	3.1
0.120	0.082	0.160	7.647	1069.6	1074.1	4.5
0.130	0.085	0.209	14.506	1070.8	1078.2	7.4
0.140	0.090	0.258	17.708	1071.3	1084.3	13.0
0.170	0.090	0.307	38.182	1076.0	1098.2	22.2
0.190	0.095	0.357	41.187	1077.5	1105.0	27.5
PEG 6000 + $(NH_4)_2SO_4$						
0.110	0.086	0.130	6.870	1068.5	1071.8	3.3
0.080	0.100	0.175	13.587	1068.3	1075.0	6.7
0.100	0.100	0.225	16.589	1072.1	1082.3	10.2
0.114	0.109	0.290	29.073	1075.3	1092.9	17.6
0.170	0.100	0.364	50.121	1078.2	1103.4	25.2
PEG 20000 + $(NH_4)_2SO_4$						
0.080	0.080	0.155	39.622	1061.6	1063.0	1.4
0.109	0.075	0.175	45.592	1061.2	1063.7	2.5
0.100	0.080	0.213		1063.8	1067.5	3.7
0.130	0.075	0.238		1065.0	1071.1	6.1
0.130	0.080	0.270		1067.0	1075.0	8.0

correlated as follows:

$$(1/\varrho)/(\mathrm{m^3{\cdot}kg^{-1}}) = (1/\varrho_0)/(\mathrm{m^3{\cdot}kg^{-1}}) - 1.5 \times 10^{-4}w_1 - c \times 10^{-3}w_3 \quad (3)$$

where ϱ is the density of the solution, ϱ_0 is the density of the solvent (in this paper, the solvent is a phosphate buffer, ϱ_0 = 1008.0 kg·m^{-3}), w_1 is the mass fraction of PEG, w_3 is the mass fraction of salt, the units of 1.5 × 10^{-4} are m^3·kg^{-1}, and c is a constant, with the value 0.80 m^3·kg^{-1} for K_3PO_4 and 0.48 m^3·kg^{-1} for $(NH_4)_2SO_4$.

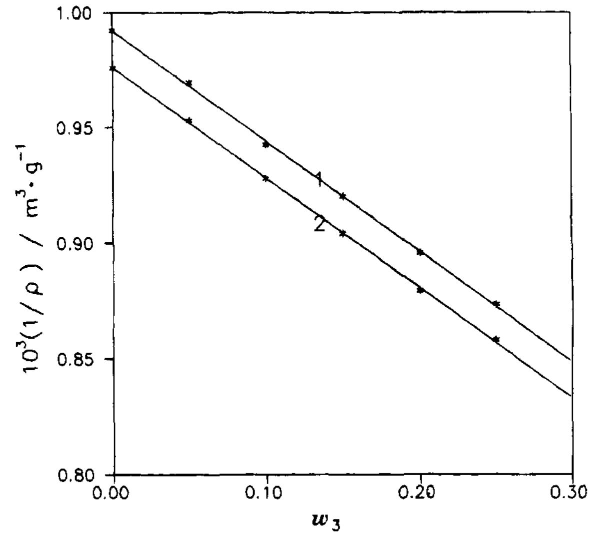

Figure 5. Relationship between the densities of PEG (1) + $(NH_4)_2SO_4$ (3) + H_2O (2) and mass fraction w_3 of $(NH_4)_2SO_4$ (1, w_1 = 0; 2, w_1 = 0.10).

Table 5. Densities and Viscosities of PEG (1) + H_2O (2) + K_3PO_4 (3) Aqueous Two-Phase Systems at 20 °C

w_1	w_3	TLL	$\eta_{(T)}$/(mPa·s)	$\varrho_{(T)}$/(kg·m^{-3})	$\varrho_{(B)}$/(kg·m^{-3})	$\Delta\varrho$/(kg·m^{-3})
PEG 1000 + K_3PO_4						
0.130	0.105	0.178	3.387	1106.5	1154.2	47.7
0.140	0.110	0.256	4.898	1098.1	1167.5	67.4
0.150	0.112	0.319	6.777	1093.8	1177.9	85.1
0.150	0.122	0.382	9.628	1093.0	1196.0	103.0
0.180	0.115	0.405	10.235	1092.0	1202.0	110.0
0.205	0.110	0.429	11.818	1092.2	1212.2	120.0
0.220	0.110	0.470	13.969	1093.5	1241.2	147.7
PEG 2000 + K_3PO_4						
0.130	0.085	0.192	5.129	1091.0	1126.1	35.1
0.140	0.090	0.263	7.763	1087.6	1141.6	54.0
0.140	0.100	0.318	11.111	1083.0	1156.5	73.5
0.150	0.107	0.347	14.396	1085.2	1171.5	86.3
0.200	0.090	0.370	15.099	1085.9	1180.3	94.4
0.220	0.100	0.425	18.944	1087.9	1230.4	142.5
PEG 4000 + K_3PO_4						
0.090	0.081	0.122	6.036	1083.9	1103.8	20.1
0.110	0.080	0.162	12.596	1082.0	1110.7	28.7
0.120	0.088	0.233	18.201	1081.2	1125.0	43.8
0.135	0.090	0.289	29.297	1078.5	1133.5	55.0
0.190	0.080	0.325	35.958	1079.1	1152.0	72.9
0.190	0.090	0.368	40.057	1080.5	1184.5	104.0
PEG 6000 + K_3PO_4						
0.090	0.080	0.151	9.333	1079.2	1106.9	27.7
0.100	0.084	0.184	12.589	1078.6	1116.8	38.2
0.110	0.090	0.235	19.621	1079.0	1130.3	51.3
0.150	0.080	0.283	29.767	1079.8	1147.9	68.1
0.170	0.080	0.328	35.019	1080.4	1164.9	84.5
0.190	0.090	0.388	39.483	1076.3	1212.5	136.2
PEG 20000 + K_3PO_4						
0.100	0.064	0.130	29.184	1075.0	1089.5	14.5
0.110	0.064	0.155	45.711	1075.4	1093.2	17.8
0.125	0.064	0.187		1076.2	1100.5	24.3
0.150	0.064	0.217		1079.1	1111.6	32.5

The viscosities of the top phase and densities of the top and bottom phases for some PEG + salt + water two-phase systems are listed in Tables 4 and 5. The density differences between phases and the viscosities of the top phases generally increase with the increase of the TLLs (TLL is the tie line length, TLL = $[(w_{1(T)} - w_{1(B)})^2 + (w_{3(T)} - w_{3(B)})^2]^{1/2}$). The data of the phase diagrams come from Mei (1988) and Albertsson (1986) for all systems. In Figures 6–9, the common logarithm of the density differences between phases and of the viscosities of the top phase show linear relationships with log(TLL).

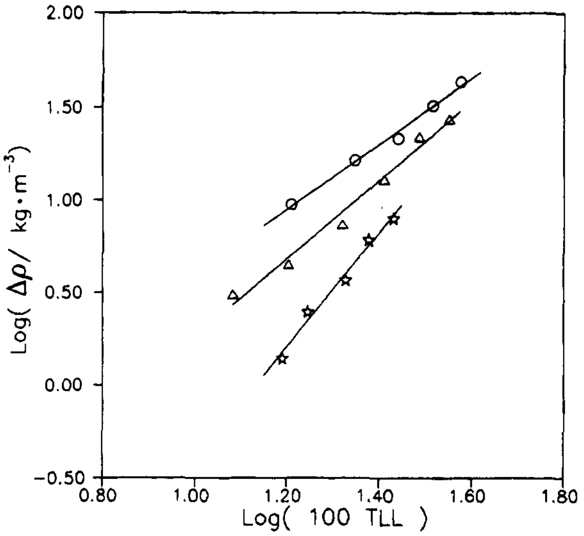

Figure 6. Relationship between log(Δϱ) and log(TLL) for PEG + $(NH_4)_2SO_4$ + H_2O two-phase systems: (○) PEG 1000; (△) PEG 4000; (☆) PEG 20000.

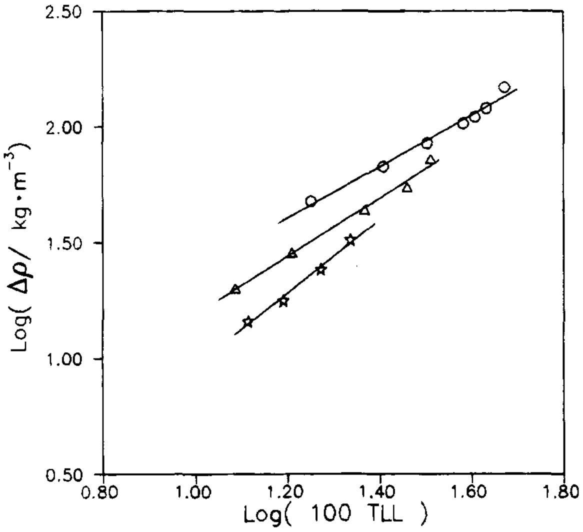

Figure 7. Relationship between log(Δϱ) and log(TLL) for PEG + K_3PO_4 + H_2O two-phase systems: (○) PEG 1000; (△) PEG 4000; (☆) PEG 20000.

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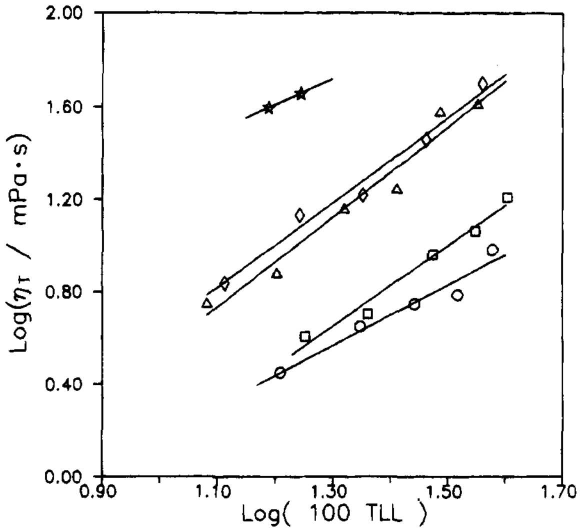

Figure 8. Relationship between log(η_T) and log(TLL) for PEG + $(NH_4)_2SO_4$ + H_2O two-phase systems: (○) PEG 1000; (□) PEG 2000; (△) PEG 4000; (◇) PEG 6000; (☆) PEG 20000.

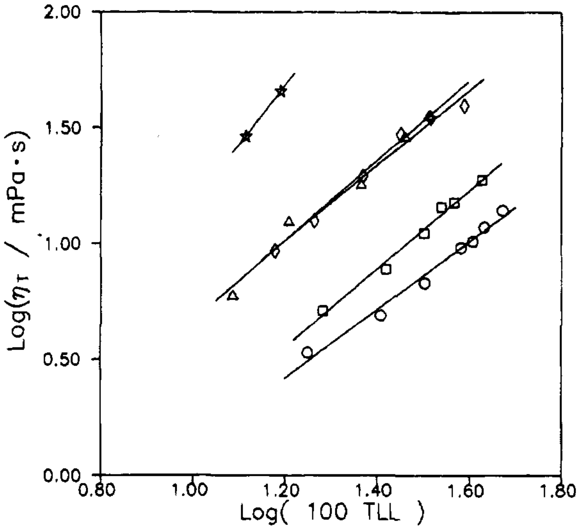

Figure 9. Relationship between log(η_T) and log(TLL) for PEG + K_3PO_4 + H_2O two-phase systems: (○) PEG 1000; (□) PEG 2000; (△) PEG 4000; (◇) PEG 6000; (☆) PEG 20000.

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