Densities and Viscosities of Poly(ethylene glycol) 4000 + Diammonium Hydrogen Phosphate + Water Systems

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The densities and viscosities of binary and ternary solutions of the poly(ethylene glycol) 4000 (PEG4000) + diammonium hydrogen phosphate + water system were determined at different temperatures [(298.15, 303.15, 308.15, 313.15, and 318.15) K]. The measured density and viscosity data of all the binary and ternary systems were fitted to available empirical correlations, for the corresponding temperatures. The density data show a linear variation with mass fraction of the polymer for all temperatures. The viscosity data of all the solutions were correlated as a function of their mass fraction, using a nonlinear equation, for the five different temperatures covered in the present work. Densities and viscosities of PEG4000−diammonium hydrogen phosphate two-phase systems have been measured at (298.15, 303.15, 308.15, 313.15, and 318.15) K. The tie line lengths (TLL) of the aqueous two-phase systems have also been estimated, and the effect of the physical properties on the TLL is also reported.

Introduction

Aqueous two-phase systems (ATPSs) can be formed either by the addition of two incompatible polymer solutions or one polymer with an inorganic salt above a critical concentration. Aqueous two-phase systems, especially poly(ethylene glycol) (PEG)/salt systems, have been widely used for the separation of biomolecules because of their mild environment, greater selectivity, larger differences in density, lower viscosity, less cost etc., when compared with polymer−polymer systems.[1−3] For the design of extractors, the necessary liquid−liquid equilibrium (LLE) data for few systems only are available in the open literature.[4−10] Data on the physicochemical properties of the phases, such as the viscosity, density, refractive index, etc., at different temperature are also necessary for the efficient design and development of equipment for extraction using ATPS for commercial applications.[11−15] In the present work, an attempt has been made to measure the density, refractive index, and viscosity of the binary and ternary mixtures of the poly(ethylene glycol) 4000 (PEG4000) + diammonium hydrogen phosphate + water based aqueous two-phase systems.

Experimental Section

Materials. Analytical grade poly(ethylene glycol) 4000 (Merck-Schuchardt (Hohenbrunn bei München, Germany), cat. no: 8.07490.1000) with a molar mass average of 4000 (3500 to 4500) and diammonium hydrogen phosphate (Merck, India catalog No: 61757805001730) with a minimum purity of 99 % were used. The polymer and salts were used without further purification. Double distilled, deionized water was used for the present experiments.

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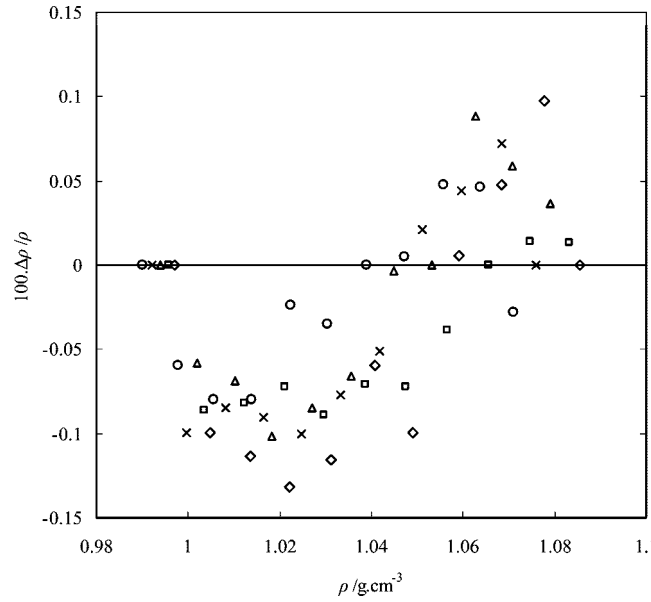

Figure 1. Relative error ($100(\rho - \rho_{cal})/\rho$) between the experimental and predicted density values using eq 1 for aqueous PEG4000: ◇, 298.15 K; □, 303.15 K; Δ, 308.15 K; ×, 313.15 K; ○, 318.15 K.

Apparatus and Procedure. The solutions for the formation of aqueous single-phase (binary and ternary system) and two-phase systems were prepared by mass in 50 cm^3 capped, graduated flasks, using an analytical balance (OHAUS-Essae-Teraoka Ltd., Japan, model AR2140) with an accuracy of 0.1 mg. The prepared aqueous solutions were kept in a Schott-Gerate CT 52 (Germany) thermostatic bath to maintain the appropriate temperature with an uncertainty of ± 0.05 °C. Then the densities and viscosities of the single phase (PEG4000 + water, diammonium hydrogen phosphate + water, and PEG4000

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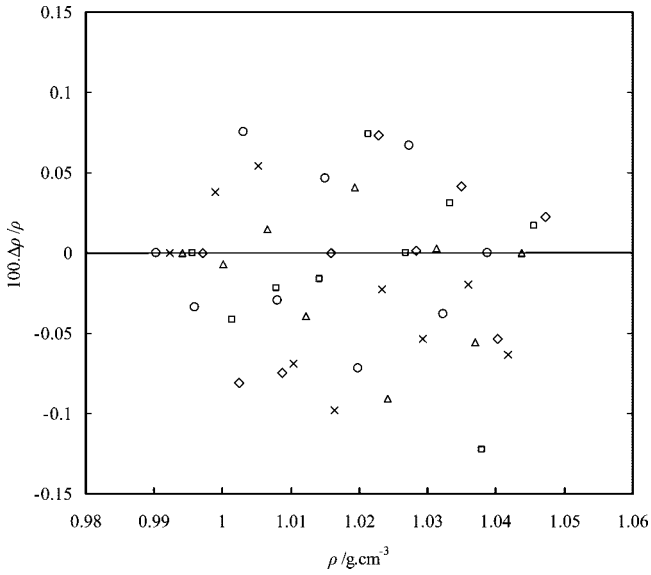

Figure 2. Relative error ($100(\rho - \rho_{cal})/\rho$) between the experimental and predicted density values using eq 1 for diammonium hydrogen phosphate: ◇, 298.15 K; □, 303.15 K; Δ, 308.15 K; ×, 313.15 K; ○, 318.15 K.

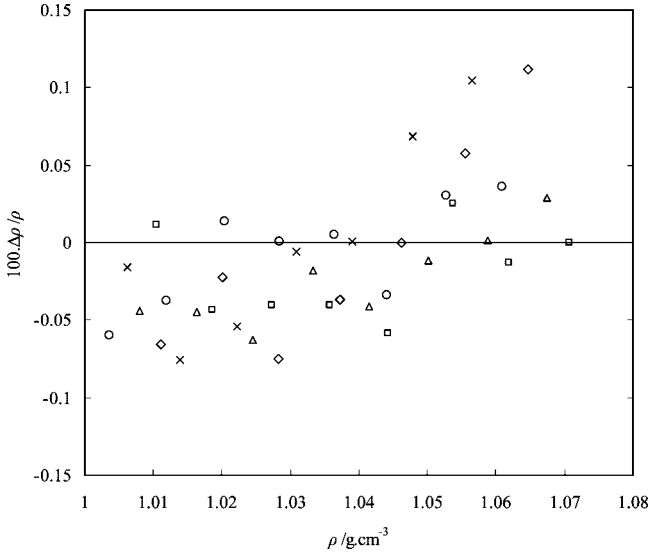

Figure 3. Relative error ($100(\rho - \rho_{cal})/\rho$) between the experimental and predicted density values using eq 2 for PEG4000 + diammonium hydrogen phosphate ($w_S = 0.01$) + water: ◇, 298.15 K; □, 303.15 K; Δ, 308.15 K; ×, 313.15 K; ○, 318.15 K.

+ diammonium hydrogen phosphate +water) and two phases (top and bottom phases formed from the PEG4000 + diammonium hydrogen phosphate + water system) were measured at different temperatures of (298.15, 303.15, 308.15, 313.15, and 318.15) K. The 5 cm^3 glass pycnometer was used to measure the densities of the solution.[15] The densities of pure water at different temperatures were taken from Perry's Chemical Engineers' Handbook.[16] The uncertainty of the density measurements was estimated to be ± 0.0001 g·cm^{-3}. Calibrated Ostwald viscometers of different capillary sizes were used to measure the viscosities of all the above said aqueous PEG4000, salt, and top and bottom phase of the aqueous two-phase systems at (298.15, 303.15, 308.15, 313.15, and 318.15) K in a water bath. The uncertainty of the measurements was ± 0.002 mPa·s. All the measurements for each sample were performed in dublicate (for density and refractive index) and triplicate (for viscosity), and the average values are reported. The uncertainty of the measurements were estimated to be ± 2 %. The details of the experimental procedure were previously described by Murugesan and Perumalsamy (2005).[15]

Table 1. Densities of the PEG4000 (P) + Water and Diammonium Hydrogen Phosphate (S) + Water Systems at Various Temperatures

w_P	ρ / g·cm^{-3}	$100(\rho - \rho_{cal})/\rho$	w_S	ρ / g·cm^{-3}	$100(\rho - \rho_{cal})/\rho$
		298.15 K			
0.0000	0.9970	0.000	0.0000	0.9970	0.000
0.0500	1.0049	−0.100	0.0100	1.0025	−0.081
0.1000	1.0136	−0.114	0.0200	1.0088	−0.075
0.1500	1.0223	−0.132	0.0300	1.0158	0.000
0.2000	1.0313	−0.116	0.0400	1.0228	0.073
0.2500	1.0407	−0.059	0.0500	1.0283	0.002
0.3000	1.0492	−0.100	0.0600	1.0350	0.042
0.3500	1.0591	0.006	0.0700	1.0403	−0.053
0.4000	1.0684	0.048	0.0800	1.0473	0.023
0.4500	1.0778	0.097			
0.5000	1.0856	0.000			
		303.15 K			
0.0000	0.9956	0.000	0.0000	0.9956	0.000
0.0500	1.0035	−0.086	0.0100	1.0015	−0.041
0.1000	1.0123	−0.082	0.0200	1.0079	−0.022
0.1500	1.0212	−0.072	0.0300	1.0142	−0.016
0.2000	1.0297	−0.089	0.0400	1.0213	0.074
0.2500	1.0387	−0.071	0.0500	1.0268	0.000
0.3000	1.0474	−0.072	0.0600	1.0333	0.031
0.3500	1.0565	−0.038	0.0700	1.0379	−0.123
0.4000	1.0656	0.000	0.0800	1.0456	0.017
0.4500	1.0745	0.014			
0.5000	1.0833	0.014			
		308.15 K			
0.0000	0.9940	0.000	0.0000	0.9940	0.000
0.0500	1.0019	−0.058	0.0100	1.0002	−0.007
0.1000	1.0103	−0.068	0.0200	1.0066	0.015
0.1500	1.0184	−0.102	0.0300	1.0123	−0.039
0.2000	1.0270	−0.085	0.0400	1.0193	0.041
0.2500	1.0357	−0.066	0.0500	1.0241	−0.091
0.3000	1.0448	−0.004	0.0600	1.0313	0.003
0.3500	1.0533	0.000	0.0700	1.0369	−0.055
0.4000	1.0627	0.088	0.0800	1.0437	0.000
0.4500	1.0709	0.059			
0.5000	1.0791	0.036			
		313.15 K			
0.0000	0.9922	0.000	0.0000	0.9922	0.000
0.0500	0.9996	−0.100	0.0100	0.9989	0.038
0.1000	1.0081	−0.085	0.0200	1.0053	0.054
0.1500	1.0164	−0.091	0.0300	1.0103	−0.069
0.2000	1.0247	−0.100	0.0400	1.0163	−0.098
0.2500	1.0333	−0.077	0.0500	1.0233	−0.022
0.3000	1.0419	−0.052	0.0600	1.0293	−0.054
0.3500	1.0511	0.021	0.0700	1.0359	−0.020
0.4000	1.0597	0.044	0.0800	1.0417	−0.063
0.4500	1.0684	0.072			
0.5000	1.0760	0.000			
		318.15 K			
0.0000	0.9902	0.000	0.0000	0.9902	0.000
0.0500	0.9977	−0.060	0.0100	0.9959	−0.034
0.1000	1.0057	−0.080	0.0200	1.0031	0.075
0.1500	1.0138	−0.080	0.0300	1.0081	−0.029
0.2000	1.0225	−0.024	0.0400	1.0149	0.046
0.2500	1.0305	−0.035	0.0500	1.0198	−0.072
0.3000	1.0389	0.000	0.0600	1.0273	0.067
0.3500	1.0471	0.005	0.0700	1.0323	−0.038
0.4000	1.0557	0.048	0.0800	1.0387	0.000
0.4500	1.0638	0.047			
0.5000	1.0711	−0.028			

The tie lines were determined in triplicate from mixtures of known overall composition above the binodal curve (typically 50 cm^3 in capped, graduated flasks). The detailed experimental procedure for binodal curve, equilibrium concentration measurement for both PEG and salt is available elsewhere.[7−10,15,17] The concentration of PEG in the samples was determined by the

Table 2. Densities and Refractive Index of the PEG4000 (P) + Diammonium Hydrogen Phosphate (S) + Water System at Various Temperatures

w_P	w_S	ρ / g·cm^{-3}	$100(\rho - \rho_{cal})/\rho$	n_D	$100(n_D - n_{Dcal})/n_D$	ρ / g·cm^{-3}	$100(\rho - \rho_{cal})/\rho$	n_D	$100(n_D - n_{Dcal})/n_D$
			298.15 K				303.15 K		
0.0500	0.0100	1.0111	−0.066	1.3432	0.086	1.0105	0.011	1.3428	0.065
0.1000	0.0100	1.0202	−0.022	1.3504	0.052	1.0186	−0.043	1.3492	0.031
0.1500	0.0100	1.0282	−0.075	1.3551	−0.130	1.0272	−0.040	1.3555	−0.076
0.2000	0.0100	1.0373	−0.037	1.3646	−0.015	1.0358	−0.040	1.3637	−0.035
0.2500	0.0100	1.0463	0.000	1.3714	−0.049	1.0442	−0.058	1.3701	−0.067
0.3000	0.0100	1.0555	0.058	1.3802	0.064	1.0537	0.026	1.3791	0.046
0.3500	0.0100	1.0647	0.112	1.3877	0.030	1.0619	−0.013	1.3864	0.013
0.4000	0.0100	1.0738	0.152	1.3945	−0.002	1.0707	0.000	1.3937	−0.019
0.0500	0.0200	1.0180	0.014	1.3451	0.061	1.0166	0.002	1.3446	0.049
0.1000	0.0200	1.0258	−0.068	1.3522	0.027	1.0252	0.008	1.3519	0.016
0.1500	0.0200	1.0348	−0.032	1.3573	−0.155	1.0340	0.022	1.3573	−0.091
0.2000	0.0200	1.0435	−0.019	1.3672	0.033	1.0428	0.039	1.3664	0.023
0.2500	0.0200	1.0492	−0.303	1.3758	0.072	1.0516	0.057	1.3746	0.063
0.3000	0.0200	1.0619	0.089	1.3811	−0.034	1.0599	0.030	1.3800	−0.042
0.0500	0.0300	1.0239	0.000	1.3464	−0.039	1.0227	0.005	1.3454	−0.041
0.1000	0.0300	1.0322	−0.032	1.3545	0.001	1.0307	−0.058	1.3532	0.000
0.1500	0.0300	1.0415	0.035	1.3608	−0.106	1.0399	−0.003	1.3590	−0.107
0.2000	0.0300	1.0498	−0.001	1.3707	0.081	1.0480	−0.049	1.3691	0.081
0.2500	0.0300	1.0571	−0.130	1.3783	0.120	1.0564	−0.069	1.3772	0.120
0.0500	0.0400	1.0310	0.100	1.3482	−0.064	1.0292	0.031	1.3473	−0.057
0.1000	0.0400	1.0385	−0.017	1.3564	−0.024	1.0375	0.006	1.3555	−0.016
0.1500	0.0400	1.0467	−0.050	1.3636	−0.057	1.0570	1.035	1.3628	−0.049
0.2000	0.0400	1.0562	0.025	1.3733	0.129	1.0549	0.024	1.3717	0.065
0.0500	0.0500	1.0371	0.093	1.3511	−0.016	1.0351	0.016	1.3508	0.002
0.1000	0.0500	1.0447	−0.003	1.3582	−0.049	1.0429	−0.069	1.3573	−0.031
0.1500	0.0500	1.0541	0.070	1.3652	−0.082	1.0519	−0.023	1.3644	−0.064
0.0500	0.0600	1.0414	−0.076	1.3543	0.033	1.0411	0.000	1.3537	0.060
0.1000	0.0600	1.0506	−0.019	1.3616	−0.001	1.0498	0.003	1.3604	0.027
0.0500	0.0700	1.0493	0.097	1.3564	0.008	1.0469	−0.032	1.3546	−0.030
0.0500	0.0800	1.0545	0.018	1.3608	0.130	1.0533	−0.006	1.3582	0.102
			308.15 K				313.15 K		
0.0500	0.0100	1.0080	−0.044	1.3416	0.044	1.0063	−0.016	1.3400	0.062
0.1000	0.0100	1.0163	−0.044	1.3482	0.017	1.0138	−0.076	1.3464	−0.038
0.1500	0.0100	1.0245	−0.063	1.3546	−0.085	1.0222	−0.054	1.3546	0.010
0.2000	0.0100	1.0334	−0.018	1.3628	−0.038	1.0308	−0.006	1.3619	−0.015
0.2500	0.0100	1.0415	−0.041	1.3692	−0.065	1.0390	0.001	1.3681	−0.040
0.3000	0.0100	1.0502	−0.012	1.3772	−0.019	1.0479	0.069	1.3754	−0.065
0.3500	0.0100	1.0587	0.002	1.3856	0.027	1.0564	0.104	1.3844	0.056
0.4000	0.0100	1.0674	0.029	1.3920	0.000	1.0663	0.267	1.3916	0.031
0.0500	0.0200	1.0145	0.011	1.3432	0.036	1.0126	0.009	1.3421	0.053
0.1000	0.0200	1.0226	−0.013	1.3504	0.008	1.0206	0.001	1.3490	0.027
0.1500	0.0200	1.0312	0.008	1.3563	−0.093	1.0286	−0.013	1.3553	−0.072
0.2000	0.0200	1.0397	0.018	1.3656	0.027	1.0367	−0.018	1.3641	0.050
0.2500	0.0200	1.0482	0.027	1.3728	0.000	1.0449	−0.016	1.3712	0.025
0.3000	0.0200	1.0570	0.067	1.3790	−0.027	1.0536	0.039	1.3781	0.000
0.0500	0.0300	1.0202	−0.013	1.3444	−0.047	1.0190	0.043	1.3437	−0.031
0.1000	0.0300	1.0283	−0.040	1.3522	0.000	1.0263	−0.037	1.3514	0.018
0.1500	0.0300	1.0370	−0.010	1.3580	−0.101	1.0352	0.038	1.3572	−0.081
0.2000	0.0300	1.0449	−0.056	1.3671	0.019	1.0429	−0.009	1.3652	−0.032
0.2500	0.0300	1.0538	−0.009	1.3753	0.064	1.0498	−0.132	1.3746	0.089
0.0500	0.0400	1.0266	0.022	1.3464	−0.055	1.0257	0.104	1.3458	−0.039
0.1000	0.0400	1.0348	0.001	1.3541	−0.008	1.0325	−0.025	1.3532	0.010
0.1500	0.0400	1.0539	1.022	1.3612	−0.036	1.0402	−0.069	1.3604	−0.016
0.2000	0.0400	1.0518	0.032	1.3696	0.010	1.0491	0.000	1.3673	−0.041
0.0500	0.0500	1.0322	−0.017	1.3498	0.011	1.0314	0.066	1.3484	0.026
0.1000	0.0500	1.0404	−0.028	1.3560	−0.017	1.0386	−0.024	1.3555	0.001
0.1500	0.0500	1.0490	−0.015	1.3632	−0.044	1.0476	0.059	1.3621	−0.024
0.0500	0.0600	1.0390	0.068	1.3516	0.002	1.0359	−0.086	1.3493	−0.057
0.1000	0.0600	1.0470	0.030	1.3593	0.048	1.0446	−0.032	1.3586	0.066
0.0500	0.0700	1.0443	−0.006	1.3531	−0.006	1.0437	0.088	1.3522	0.009
0.0500	0.0800	1.0509	0.054	1.3564	0.059	1.0491	0.023	1.3541	0.000
			318.15 K						
0.0500	0.0100	1.0036	−0.060	1.3391	0.052				
0.1000	0.0100	1.0119	−0.038	1.3452	−0.049				
0.1500	0.0100	1.0204	0.013	1.3532	0.000				
0.2000	0.0100	1.0283	0.001	1.3592	−0.099				
0.2500	0.0100	1.0364	0.005	1.3674	−0.051				
0.3000	0.0100	1.0441	−0.034	1.3741	−0.076				
0.3500	0.0100	1.0528	0.030	1.3833	0.044				
0.4000	0.0100	1.0609	0.036	1.3906	0.019				
0.0500	0.0200	1.0100	−0.019	1.3413	0.055				
0.1000	0.0200	1.0177	−0.053	1.3481	0.029				
0.1500	0.0200	1.0267	0.045	1.3557	0.003				
0.2000	0.0200	1.0339	−0.043	1.3628	−0.022				
0.2500	0.0200	1.0419	−0.044	1.3697	−0.048				
0.3000	0.0200	1.0502	−0.016	1.3776	0.000				

Table 2. Continued

w_P	w_S	ρ / g·cm^{-3}	$100(\rho - \rho_{cal})/\rho$	n_D	$100(n_D - n_{Dcal})/n_D$	ρ / g·cm^{-3}	$100(\rho - \rho_{cal})/\rho$	n_D	$100(n_D - n_{Dcal})/n_D$
0.0500	0.0300	1.0167	0.052	1.3420	−0.017				
0.1000	0.0300	1.0241	−0.016	1.3503	0.032				
0.1500	0.0300	1.0322	−0.012	1.3564	−0.068				
0.2000	0.0300	1.0398	−0.053	1.3641	−0.019				
0.2500	0.0300	1.0461	−0.214	1.3722	0.028				
0.0500	0.0400	1.0232	0.103	1.3440	−0.013				
0.1000	0.0400	1.0304	0.016	1.3526	0.035				
0.1500	0.0400	1.0383	0.000	1.3591	0.009				
0.2000	0.0400	1.0470	0.066	1.3655	−0.089				
0.0500	0.0500	1.0282	0.000	1.3478	0.064				
0.1000	0.0500	1.0356	−0.063	1.3546	0.038				
0.1500	0.0500	1.0454	0.107	1.3612	0.013				
0.0500	0.0600	1.0330	−0.117	1.3483	−0.007				
0.1000	0.0600	1.0425	0.026	1.3560	0.041				
0.0500	0.0700	1.0405	0.033	1.3508	−0.004				
0.0500	0.0800	1.0458	−0.037	1.3524	0.000				

Table 3. Coefficients of Equation 1

T	ρ_o	PEG4000		diammonium hydrogen phosphate	
K	g·cm^{-3}	A	AARD/%[a]	A	AARD/%[a]
298.15	0.9970	0.1772	0.0701	0.6252	0.0436
303.15	0.9956	0.1750	0.0489	0.62244	0.0360
308.15	0.9940	0.1693	0.0515	0.6207	0.0314
313.15	0.9922	0.1675	0.0583	0.6273	0.0523
318.15	0.9902	0.1625	0.0368	0.6066	0.0452

[a] AARD = $(1/N)\sum_{i=1}^{N}((100\rho^{exp} - 100\rho^{cal})^2/100\rho^{exp})^{0.5}$.

Table 4. Coefficients of Equations 2 and 3

	density (eq 2)				n_D (eq 3)			
T/K	A	B	ρ_o/g·cm^{-3}	AARD/%	a_1	a_2	a_o	AARD/%
298.15	0.1725	0.6085	0.9970	0.0605	0.1491	0.2341	1.3320	0.0582
303.15	0.1722	0.6144	0.9956	0.0607	0.1490	0.2213	1.3315	0.0495
308.15	0.1676	0.5986	0.9940	0.0606	0.1474	0.2113	1.3309	0.0338
313.15	0.1629	0.6064	0.9922	0.0508	0.146849	0.2118	1.3297	0.0360
318.15	0.1607	0.5988	0.9902	0.0450	0.1469	0.1957	1.3290	0.0341

refractive index method using an Abbe Refractometer (Advance Research Instruments Co., New Delhi, Model R-4) having a precision of ± 0.0001. The concentrations of diammonium hydrogen phosphate in the top and bottom phases were determined through an analytical procedure using hydrochloric acid as titrant.[17]

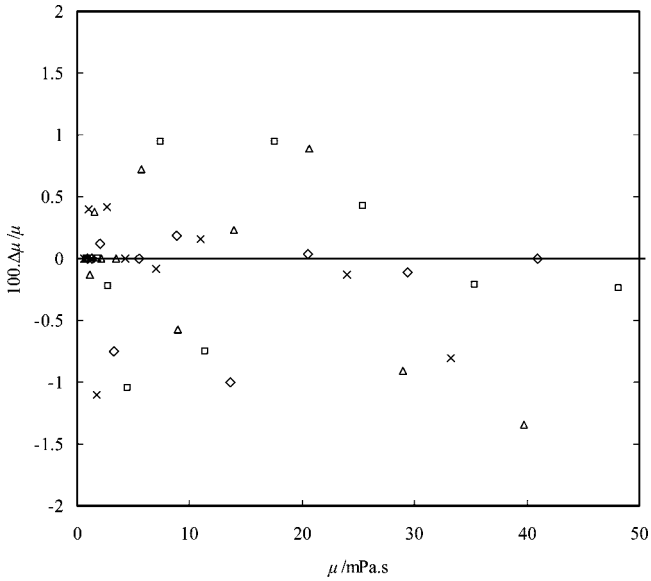

Figure 4. Relative error $(100(\mu - \mu_{cal})/\mu)$ between the experimental and predicted viscosity values using eq 4 for aqueous PEG4000: ◇, 298.15 K; □, 303.15 K; Δ, 308.15 K; ×, 313.15 K; ○, 318.15 K.

Results and Discussion

The measured densities of the aqueous solutions of PEG4000 and diammonium hydrogen phosphate are given in Table 1. The densities are found to decrease with an increase in temperature and increase with an increase in PEG4000 and diammonium hydrogen phosphate concentrations. The densities for the binary systems could be correlated by using the following equation[8]

$$\rho/\text{g·cm}^{-3} = Aw + \rho_o/\text{g·cm}^{-3} \quad (1)$$

where ρ is the density of the solution; ρ_o is the density of pure water at the corresponding temperature; and w is the mass fraction of PEG4000/diammonium hydrogen phosphate. The constant "A" values for both the PEG4000 and diammonium hydrogen phosphate systems along with corresponding average absolute relative deviation (AARD) are reported in Table 3, and the relative deviations are shown in Figure 1 and 2, respectively.

The densities and refractive index[15] of the PEG4000 + diammonium hydrogen phosphate system + water systems (Table 2) could be expressed as follows

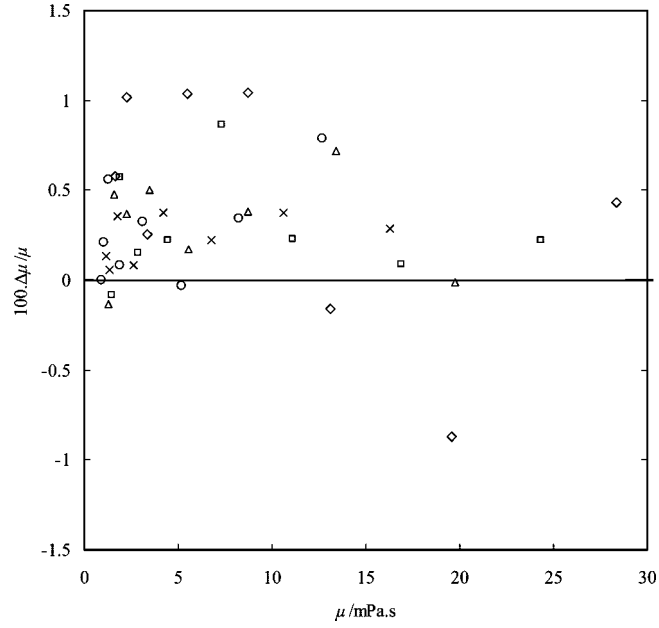

Figure 5. Relative error $(100(\mu - \mu_{cal})/\mu)$ between the experimental and predicted viscosity values using eq 5 for aqueous solution for diammonium hydrogen phosphate. ◇, 298.15 K; □, 303.15 K; Δ, 308.15 K; ×, 313.15 K; ○, 318.15 K.

$$\rho/\mathrm{g{\cdot}cm^{-}} = Aw_P + Bw_S + \rho_o/\mathrm{g{\cdot}cm^{-3}} \quad (2)$$

$$n_D = a_o + a_1 w_P + a_2 w_S \quad (3)$$

where w_P and w_S are the mass fractions of polymer and salt, respectively. Constants A, B, a_0, a_1, and a_2 and the pure water densities at different temperatures with corresponding AARD are given in Table 4, and the deviations are shown in Figure 3.

The viscosity data for the binary (Table 5) and ternary (Table 7) systems could be represented in the following form of correlations.[13,17]

$$\mu/\mathrm{mPa{\cdot}s} = Aw_P^3 + Bw_P^2 + Cw_P + \mu_o/\mathrm{mPa{\cdot}s} \quad (4)$$

$$\mu/\mathrm{mPa{\cdot}s} = Aw_S^3 + Bw_S^2 + Cw_S + \mu^o/\mathrm{mPa{\cdot}s} \quad (5)$$

Table 5. Viscosities of the PEG4000 (P) + Water and Diammonium Hydrogen Phosphate (S) + Water Systems at Various Temperatures

PEG4000 (P) + water			diammonium hydrogen phosphate (S) + water		
w_P	μ/mPa·s	$100(\mu - \mu_{cal})/\mu$	w_S	μ/mPa·s	$100(\mu - \mu_{cal})/\mu$
		298.15 K			
0.0000	0.894	0.000	0.0000	0.894	0.000
0.0500	1.352	0.000	0.0100	0.910	−0.387
0.1000	2.047	0.119	0.0200	0.933	−0.037
0.1500	3.275	−0.746	0.0300	0.952	−0.162
0.2000	5.442	0.000	0.0400	0.970	−0.482
0.2500	8.817	0.185	0.0500	0.996	0.000
0.3000	13.567	−1.003	0.0600	1.017	−0.092
0.3500	20.484	0.040	0.0700	1.043	0.275
0.4000	29.412	−0.115	0.0800	1.063	0.000
0.4500	40.941	−0.001			
0.5000	56.401	1.974			
		303.15 K			
0.0000	0.801	0.000	0.0000	0.801	0.000
0.0500	1.233	0.000	0.0100	0.825	0.163
0.1000	1.788	−0.002	0.0200	0.845	−0.152
0.1500	2.774	−0.218	0.0300	0.870	0.053
0.2000	4.475	−1.050	0.0400	0.894	0.161
0.2500	7.397	0.941	0.0500	0.913	−0.398
0.3000	11.424	−0.746	0.0600	0.937	−0.345
0.3500	17.548	0.949	0.0700	0.964	0.000
0.4000	25.364	0.425	0.0800	0.990	0.143
0.4500	35.372	−0.212			
0.5000	48.151	−0.244			
		308.15 K			
0.0000	0.723	0.000	0.0000	0.723	0.000
0.0500	1.100	−0.128	0.0100	0.736	−0.153
0.1000	1.493	0.380	0.0200	0.746	−0.693
0.1500	2.175	0.000	0.0300	0.766	−0.015
0.2000	3.460	0.000	0.0400	0.782	0.000
0.2500	5.679	0.722	0.0500	0.797	−0.047
0.3000	8.954	−0.570	0.0600	0.816	0.272
0.3500	13.888	0.230	0.0700	0.830	0.049
0.4000	20.670	0.892	0.0800	0.845	−0.264
0.4500	28.929	−0.906			
0.5000	39.735	−1.340			
		313.15 K			
0.0000	0.656	0.000	0.0000	0.656	0.000
0.0500	1.009	0.398	0.0100	0.670	−0.250
0.1000	1.266	0.000	0.0200	0.687	0.029
0.1500	1.698	−1.103	0.0300	0.702	−0.267
0.2000	2.646	0.413	0.0400	0.718	−0.316
0.2500	4.299	0.000	0.0500	0.738	0.069
0.3000	6.981	−0.081	0.0600	0.757	0.293
0.3500	10.992	0.159	0.0700	0.773	0.000
0.4000	17.002	2.704	0.0800	0.789	−0.401
0.4500	23.936	−0.129			
0.5000	33.260	−0.804			
		318.15 K			
0.0000	0.599	0.000	0.0000	0.599	0.000
0.0500	0.859	0.021	0.0100	0.612	0.138
0.1000	0.994	−0.151	0.0200	0.622	−0.294
0.1500	1.260	0.000	0.0300	0.636	−0.190
0.2000	1.920	0.777	0.0400	0.648	−0.334
0.2500	3.165	−0.494	0.0500	0.664	0.113
0.3000	5.365	0.466	0.0600	0.677	0.000
0.3500	8.587	−0.564	0.0700	0.690	−0.176
0.4000	13.237	−0.609	0.0800	0.706	0.155
0.4500	19.522	−0.599			
0.5000	27.680	−0.621			

$$\ln(\mu_m/\mathrm{mPa{\cdot}s}) = c_1 \ln(\mu_P/\mathrm{mPa{\cdot}s}) + c_2 \ln(\mu_S/\mathrm{mPa{\cdot}s}) + c_1 c_2 a \quad (6)$$

where

$$c_1 = w_P/(w_P + w_S)$$

$$c_2 = w_S/(w_P + w_S)$$

where μ is the absolute viscosity of the solution and μ_o is the viscosity of water at the corresponding temperature. The coefficients of the polynomial eqs 4 and 5 are determined by regression analysis. The details are given in Table 6, and the deviations are shown in Figures 4 and 5, respectively. The value of constant "a" in eq 6 and the corresponding AARD for different temperatures are given in Table 7, and the respective deviations are shown in Figure 6.

For the two-phase system of PEG4000 + diammonium hydrogen phosphate + water, the densities and viscosities of the top and bottom phases are measured and listed in Table 8, along with tie line compositions and tie line lengths (TLL). The tie line lengths are estimated using the following relationship[15,19]

$$\mathrm{TLL} = [(w_{P(T)} - w_{P(B)})^2 + (w_{S(T)} - w_{S(B)})^2]^{1/2} \quad (7)$$

It can be seen from the Table 8 that the TLL increases with increasing temperature and PEG composition. The density difference between the phases ($\Delta\rho$) and the viscosities of the top phase (μ_T) and the bottom phases (μ_B) are also given in Table 8. The density difference between the phases and the

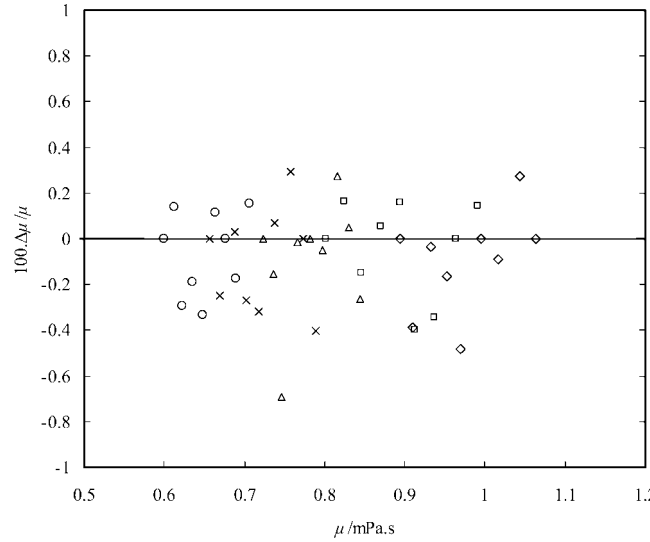

Figure 6. Relative error ($100(\mu - \mu_{cal})/\mu$) between the experimental and predicted viscosity values using eq 6 for PEG4000 + diammonium hydrogen phosphate ($w_S = 0.01$) + water: ◇, 298.15 K; □, 303.15 K; Δ, 308.15 K; ×, 313.15 K; ○, 318.15 K.

Table 6. Coefficients of Equations 4 and 5

T/K	A	B	C	μ_o/mPa·s	AARD/%
		PEG 4000−water (eq 4)			
298.15	436.2853	−18.5741	9.0038	0.8940	0.3802
303.15	417.6579	−37.9367	9.4876	0.8010	0.4351
308.15	393.9901	−57.7835	9.4822	0.7230	0.4698
313.15	370.5622	−73.2400	9.7221	0.6560	0.5264
318.15	335.7812	−75.1239	8.1218	0.5990	0.3912
		diammonium hydrogen phosphate + water (eq 5)			
298.15	1.1331	2.4675	1.9074	0.8940	0.1793
303.15	1.0068	1.0835	2.2541	0.8010	0.1769
308.15	0.2065	2.0073	1.3861	0.7230	0.1867
313.15	1.0466	2.1769	1.5177	0.6560	0.2032
318.15	1.0013	1.0174	1.2368	0.5990	0.1556

Table 7. Viscosities of the PEG4000 (P) + Diammonium Hydrogen Phosphate (S) + Water System at Various Temperatures

		$T = 298.15$ K		$T = 303.15$ K		$T = 308.15$ K		$T = 313.15$ K		$T = 318.15$ K	
w_P	w_S	μ/mPa·s	$100(\mu - \mu_{cal})/\mu$	μ/mPa·s	$100(\mu - \mu_{cal})/\mu$	μ/mPa·s	$100(\mu - \mu_{cal})/\mu$	μ/mPa·s	$100(\mu - \mu_{cal})/\mu$	μ/mPa·s	$100(\mu - \mu_{cal})/\mu$
0.0500	0.0100	1.637	0.576	1.447	−0.080	1.267	−0.132	1.140	0.132	0.942	0.000
0.1000	0.0100	2.231	1.015	1.920	0.572	1.593	0.476	1.338	0.058	1.042	0.207
0.1500	0.0100	3.369	0.253	2.836	0.153	2.228	0.369	1.741	0.358	1.290	0.560
0.2000	0.0100	5.482	1.035	4.459	0.225	3.459	0.504	2.638	0.081	1.910	0.085
0.2500	0.0100	8.730	1.041	7.288	0.864	5.560	0.169	4.224	0.374	3.101	0.322
0.3000	0.0100	13.136	−0.158	11.073	0.226	8.692	0.383	6.767	0.224	5.171	−0.031
0.3500	0.0100	19.557	−0.868	16.866	0.087	13.427	0.717	10.589	0.373	8.242	0.345
0.4000	0.0100	28.332	0.431	24.315	0.219	19.748	−0.014	16.274	0.285	12.698	0.786
0.0500	0.0200	1.751	−0.471	1.547	−0.078	1.340	0.014	1.188	−0.841	0.974	−0.060
0.1000	0.0200	2.299	−0.419	1.990	0.386	1.639	−0.026	1.388	0.289	1.070	0.272
0.1500	0.0200	3.399	−0.289	2.870	0.318	2.255	0.538	1.769	0.490	1.298	0.129
0.2000	0.0200	5.400	0.298	4.411	0.126	3.407	−0.060	2.619	−0.098	1.894	0.031
0.2500	0.0200	8.485	0.382	7.055	0.059	5.418	−0.004	4.137	0.347	3.037	0.576
0.3000	0.0200	12.762	0.011	10.698	0.074	8.420	0.547	6.573	0.456	5.012	0.377
0.0500	0.0300	1.801	−0.579	1.586	−0.236	1.366	−0.125	1.222	−0.065	0.977	−0.969
0.1000	0.0300	2.348	−0.764	2.027	−0.006	1.679	0.381	1.420	0.403	1.083	−0.120
0.1500	0.0300	3.437	0.307	2.879	0.195	2.256	0.101	1.780	0.184	1.305	0.000
0.2000	0.0300	5.360	0.701	4.356	0.007	3.372	0.003	2.611	0.263	1.878	0.057
0.2500	0.0300	8.293	0.414	6.931	0.689	5.298	0.108	4.037	−0.099	2.968	0.539
0.0500	0.0400	1.822	−0.093	1.604	0.000	1.370	−0.082	1.230	−0.234	0.987	−0.016
0.1000	0.0400	2.401	0.346	2.093	1.992	1.695	0.442	1.431	−0.230	1.094	−0.069
0.1500	0.0400	3.443	0.562	2.871	−0.052	2.262	0.447	1.789	0.048	1.310	0.064
0.2000	0.0400	5.261	0.240	4.306	0.151	3.328	−0.033	2.594	0.354	1.870	0.633
0.0500	0.0500	1.824	0.011	1.598	−0.125	1.363	−0.137	1.235	−0.016	0.985	−0.207
0.1000	0.0500	2.407	0.000	2.059	−0.018	1.697	0.260	1.447	0.000	1.106	0.308
0.1500	0.0500	3.405	−0.264	2.865	0.199	2.244	−0.005	1.794	0.065	1.319	0.472
0.0500	0.0600	1.816	0.168	1.592	−0.240	1.357	−0.035	1.229	−0.921	0.983	−0.069
0.1000	0.0600	2.404	−0.089	2.065	0.086	1.698	0.200	1.462	0.154	1.109	0.272
0.0500	0.0700	1.797	−0.401	1.596	0.190	1.347	−0.011	1.224	−0.834	0.980	−0.082
0.0500	0.0800	1.789	−0.004	1.579	−0.659	1.336	0.000	1.217	−1.104	0.982	0.090
		$a = 1.8093$		$a = 1.6436$		$a = 1.5066$		$a = 1.3386$		$a = 1.070$	
		AARD = 0.406 %		AARD = 0.277 %		AARD = 0.211 %		AARD = 0.313 %		AARD = 0.255 %	

Table 8. Densities and Viscosities of the PEG4000 (P) + Diammonium Hydrogen Phosphate (S) + Water Aqueous Two-Phase System at Various Temperatures

			ρ/g·cm^{-3}			μ/mPa·s	
w_P	w_S	TLL	ρ_T	ρ_B	$\Delta\rho$	μ_T	μ_B
			$T = 298.15$ K				
0.2000	0.0800	29.7994	1.0689	1.1030	0.0342	16.670	1.344
0.2000	0.1000	35.1796	1.0731	1.1206	0.0475	23.200	1.465
0.2000	0.1200	40.7605	1.0766	1.1354	0.0589	30.671	1.634
0.2000	0.1400	45.8035	1.0802	1.1513	0.0711	36.559	1.844
0.2000	0.1600	50.5910	1.0829	1.1664	0.0835	47.335	2.052
			$T = 303.15$ K				
0.2000	0.0800	31.3331	1.0659	1.1004	0.0351	16.255	1.210
0.2000	0.1000	36.2829	1.0715	1.1177	0.0468	22.500	1.348
0.2000	0.1200	41.9110	1.0745	1.1301	0.0563	27.614	1.458
0.2000	0.1400	46.8800	1.0783	1.1457	0.0681	34.897	1.631
0.2000	0.1600	50.5518	1.0797	1.1614	0.0823	42.147	1.834
			$T = 308.15$ K				
0.2000	0.0800	33.1874	1.0636	1.0935	0.0304	13.373	1.064
0.2000	0.1000	39.5818	1.0686	1.1101	0.0420	18.648	1.138
0.2000	0.1200	43.8888	1.0716	1.1260	0.0549	24.601	1.262
0.2000	0.1400	49.2954	1.0754	1.1423	0.0675	30.560	1.392
0.2000	0.1600	52.8308	1.0789	1.1591	0.0808	37.051	1.551
			$T = 313.15$ K				
0.2000	0.0800	30.2737	1.0610	1.0871	0.0261	9.478	0.849
0.2000	0.1000	39.5774	1.0663	1.1033	0.0371	13.443	0.892
0.2000	0.1200	46.8080	1.0702	1.1226	0.0524	18.225	1.012
0.2000	0.1400	51.5656	1.0743	1.1394	0.0659	22.961	1.116
0.2000	0.1600	54.8485	1.0777	1.1556	0.0780	26.144	1.194
			$T = 318.15$ K				
0.2000	0.0800	33.5649	1.0604	1.0869	0.0266	10.014	0.786
0.2000	0.1000	40.5147	1.0648	1.1027	0.0378	13.476	0.851
0.2000	0.1200	46.4890	1.0687	1.1199	0.0513	17.694	0.910
0.2000	0.1400	51.3599	1.0731	1.1353	0.0622	22.318	1.004
0.2000	0.1600	53.7886	1.0756	1.1543	0.0787	25.420	1.091

viscosities of both top and bottom phases increases with an increase in the tie line length and decreases with an increase in temperature.

Conclusions

In the present study, the densities, refractive index, and viscosities of the binary and ternary mixtures of PEG4000 + diammonium hydrogen phosphate + water based aqueous two-phase systems were measured and correlated for five different temperatures, (298.15, 303.15, 308.15, 313.15, and 318.15) K. TLL were calculated, and the density and viscosity of the top and bottom phases were measured for different temperatures.

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