

Densities and Viscosities of Binary and Ternary Mixtures and Aqueous Two-Phase System of Poly(ethylene glycol) 2000 + Diammonium Hydrogen Citrate + Water at Different Temperatures

Sivakumar Kalaivani, Chunduru K. Srikanth, and Iyyaswami Regupathi*

Department of Chemical Engineering, National Institute of Technology Karnataka, Surathkal, Mangalore 575 025, India

ABSTRACT: The densities and viscosities of aqueous solutions of poly-(ethylene glycol) (PEG-2000) and diammonium hydrogen citrate were determined for mass fractions from 0.05 to 0.5 at different temperatures of (298.15, 303.15, 308.15, 313.15, and 318.15) K. The density data show a linear variation with mass fraction of the polymer and salt for all temperatures. The viscosity data of PEG 2000 and diammonium hydrogen citrate solutions were correlated as a function of mass fraction, using a nonlinear equation, for the five different temperatures covered in the present work. Densities and viscosities of PEGdiammonium hydrogen citrate two-phase systems have also been measured at (298.15, 303.15, 308.15, 313.15, and 318.15) K and are correlated with composition. The tie-line lengths (TLL) of the aqueous two-phase systems have also been estimated.



INTRODUCTION

Aqueous two-phase extraction has emerged as an efficient tool for the recovery and partial purification of the desired biomolecule from complex solutions such as fermentation broth, industrial effluent, and so forth. Aqueous two-phase systems can be formed by mixing appropriate amounts of polymer, salt/polymer, and water at a particular temperature and pH. It has several advantages over conventional liquid-liquid extraction and other downstream unit operations, such as the high (80 to 90%) water content in both phases provides a hydrophilic environment for the biomolecules, low interfacial tension $(10^{-4} \text{ to } 10^{-1} \text{ mN} \cdot \text{m}^{-1})$,¹ easy scale-up, low energy, continuous operation,^{2,3} integration of recovery and concentration in a single step, and high yield. A polymer-salt system is preferred over the polymer-polymer system due to high density difference, low viscosity, and low cost of salt compared to polymer.⁴ Citrate and tartrate salts are more preferable due to its low impact on environment.⁵ Data on the physicochemical properties of the system are necessary to design the extraction process⁶ and are available for few PEG-salt systems in the literature.⁶⁻⁹ Recently the authors published the binodal and liquid-liquid equilibrium data for the PEG 2000 + diammonium hydrogen citrate system.¹⁰ In the present work, densities, viscosities, and refractive indices of the binary (PEG 2000 + water and diammonium hydrogen citrate +water), ternary (PEG 2000 + diammonium hydrogen citrate + water) solutions, and individual phases (top and bottom phases) of the aqueous two-phase system were estimated at different temperatures. The experimental results were analyzed and fitted to correlations used in the literature for various similar systems.⁶⁻⁹

MATERIALS AND METHODS

Materials. Polyethylene glycol $[HO-(CH_2CH_2O)_n]$ CH₂OH] and poly(ethane-1,2-diol)-2000 (PEG-2000) (Catalog No. 8.21037.1000) with an average molar mass of 1800 g·mol⁻¹ were purchased from Merck, and diammonium hydrogen citrate $[(NH_4)_2HC_6H_5O_7]$ (CAS No. 3012-65-5) with a molar mass of 226.18 g·mol⁻¹ and minimum mole fraction purity of 0.99 was purchased from Sigma-Aldrich and used without further purification. Double-distilled water was used for all of the experiments.

Apparatus and Procedure. Single phase binary systems (PEG 2000 + water and diammonium hydrogen citrate + water) were prepared by varying the PEG and salt concentration from 0 to 0.50 weight fraction and the ternary systems with the PEG 2000 and salt concentration in the range of 0.05 to 0.20 weight fraction. The solutions were prepared in a 50 cm³ centrifuge tube by adding solutions of appropriate mass using an analytical balance (model AR2140, OHAUS-Essae-Teraoka Ltd., Japan) with an accuracy of \pm 0.01 mg. The solutions were maintained at working temperature in thermostatic bath (model RW-0525G, Refrigerating Bath, JEIO Tech) with an uncertainty of ± 0.1 K.

The concentration of PEG 2000 and salt required to form the two-phase region was identified from the phase diagram.¹⁰ The solutions were prepared in 50 cm³ capped centrifuge tubes and were subjected to low speed centrifugation to hasten phase separation. The solutions were kept undisturbed for 12 h at appropriate temperature in a thermostatic bath to ensure proper

```
Received: May 25, 2012
Accepted: July 23, 2012
Published: August 17, 2012
```

Table 1. Coefficients of Equation 1^a

| T/K | <i>a</i> ₀ | a_1 | <i>a</i> ₂ | AARD/% |
|--------|-----------------------|--------|-----------------------|--------|
| 298.15 | 1.334 | 0.1388 | 0.173 | 0.0484 |
| 303.15 | 1.333 | 0.1405 | 0.1772 | 0.0481 |
| 308.15 | 1.332 | 0.1415 | 0.1767 | 0.0499 |
| 313.15 | 1.3315 | 0.1409 | 0.1763 | 0.058 |
| 318.15 | 1.331 | 0.1396 | 0.1793 | 0.033 |

"Average arithmetic relative deviation (AARD) = $(\Sigma | (exptl - cal)/(exptl)l)/(N) \cdot 100$. Standard uncertainty *u* is $u(T) = \pm 0.1$ K.

Table 2. Densities of the PEG 2000 + Water andDiammonium Hydrogen Citrate + Water Systemsat Various Temperatures^a

| | aque | ous PEG 200 | 0 solution dei | nsity, $10^3 ho/{ m kg}$ | ·m ^{−3} |
|--|------------------------------------|--------------------------|---|-----------------------------|-------------------------|
| $W_{ m P}$ | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K |
| 0.0000 | 0.9970 | 0.9956 | 0.9940 | 0.9922 | 0.9902 |
| 0.0500 | 1.0053 | 1.0034 | 1.0023 | 0.9998 | 0.9983 |
| 0.1000 | 1.0132 | 1.0111 | 1.0103 | 1.0087 | 1.0068 |
| 0.1500 | 1.0223 | 1.0212 | 1.0193 | 1.0177 | 1.0156 |
| 0.2000 | 1.0305 | 1.0306 | 1.0282 | 1.0271 | 1.0257 |
| 0.2500 | 1.0386 | 1.0374 | 1.0358 | 1.0334 | 1.0320 |
| 0.3000 | 1.0481 | 1.0467 | 1.0451 | 1.0431 | 1.0418 |
| 0.3500 | 1.0580 | 1.0563 | 1.0541 | 1.0520 | 1.0505 |
| 0.4000 | 1.0686 | 1.0661 | 1.0636 | 1.0599 | 1.0576 |
| 0.5000 | 1.0863 | 1.0827 | 1.0801 | 1.0766 | 1.0742 |
| | aqueous d | liammonium | hydrogen citr $ ho/{ m kg} \cdot { m m}^{-3}$ | ate solution de | ensity, 10 ³ |
| Ws | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K |
| 0.0000 | 0.9970 | 0.9956 | 0.9940 | 0.9922 | 0.9902 |
| 0.0500 | 1.0186 | 1.0175 | 1.0161 | 1.0150 | 1.0138 |
| 0.1000 | 1.0415 | 1.0401 | 1.0382 | 1.0364 | 1.0354 |
| 0.1500 | 1.0635 | 1.0622 | 1.0609 | 1.0593 | 1.0585 |
| 0.2000 | 1.0861 | 1.0843 | 1.0833 | 1.0817 | 1.0808 |
| 0.2500 | 1.1078 | 1.1067 | 1.1051 | 1.1035 | 1.1027 |
| 0.3000 | 1.1313 | 1.1306 | 1.1292 | 1.1282 | 1.1270 |
| 0.3500 | 1.1566 | 1.1547 | 1.1531 | 1.1511 | 1.1499 |
| 0.4000 | 1.1760 | 1.1754 | 1.1774 | 1.1751 | 1.1732 |
| [*] W _P and I liammoniu | W _S are the m hydrog | weight fra en citrate | ctions of a solutions | queous PEC respectively. | G2000 and Standard |

uncertainties u are $u(T) = \pm 0.1$ K and $u(\rho) = \pm 0.1$ kg·m⁻³.

phase separation. The clear phases were separated using a pipet as described elsewhere.¹¹

The densities of binary and ternary solutions and the separated phases were measured in automatic density meter (DDM 2911, Rudolph Research Analytical, USA). The uncertainty of density measurement was 0.1 kg·m⁻³. The viscosity was measured using an Ostwald viscometer of different capillary sizes with an accuracy of \pm 0.002 mPa·s, and temperature was maintained in thermostatic bath. Density and viscosity measurements were done in triplicate, and the average value was reported. Refractive index measurements of ternary and binary solutions and the separated phases were carried out in an automatic digital refractometer, Atago Co. Ltd. (RX-5000 α), with an accuracy of \pm 0.00004.

The PEG concentration in the ternary solutions was obtained from refractive index measurements. The refractive index of the solution depends on the concentration of all components (salt and PEG) present in the solution. To find the concentration of PEG in the solution, a correlation was developed in terms of PEG and salt weight fraction (eq 1). The linear relation between the



Figure 1. Aqueous PEG 2000 density at various temperatures: \Diamond , 298.15 K; \Box , 303.15 K; \triangle , 308.15 K; \times , 313.15 K; \bigcirc , 318.15 K; \cdots , dotted lines corresponds to literature data.⁹

Table 3. Density of the PEG2000 + Diammonium HydrogenCitrate + Water System at Various Temperatures

| | | | 1 | $0^3 \rho/\text{kg·m}^-$ | 3 | |
|---|--|---|-------------------------------------|--|--|---------------------|
| $W_{ m P}$ | $W_{\rm S}$ | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K |
| 0.0500 | 0.0500 | 1.0248 | 1.0237 | 1.0226 | 1.0211 | 1.0197 |
| 0.1000 | 0.0500 | 1.0347 | 1.0328 | 1.0317 | 1.0300 | 1.0278 |
| 0.1500 | 0.0500 | 1.0447 | 1.0417 | 1.0397 | 1.0385 | 1.0368 |
| 0.2000 | 0.0500 | 1.0539 | 1.0457 | 1.0496 | 1.0478 | 1.0456 |
| 0.2500 | 0.0500 | 1.0642 | 1.0618 | 1.0595 | 1.0573 | 1.0551 |
| 0.0500 | 0.1000 | 1.0476 | 1.0462 | 1.0448 | 1.0433 | 1.0423 |
| 0.1000 | 0.1000 | 1.0562 | 1.0552 | 1.0527 | 1.0520 | 1.0512 |
| 0.1500 | 0.1000 | 1.0649 | 1.0641 | 1.0619 | 1.0605 | 1.0596 |
| 0.2000 | 0.1000 | 1.0736 | 1.0724 | 1.0710 | 1.0703 | 1.0689 |
| 0.0500 | 0.1500 | 1.0713 | 1.0705 | 1.0691 | 1.0664 | 1.0649 |
| 0.1000 | 0.1500 | 1.0800 | 1.0783 | 1.0772 | 1.0750 | 1.0737 |
| 0.1500 | 0.1500 | 1.0912 | 1.0878 | 1.0860 | 1.0843 | 1.0831 |
| 0.2000 | 0.1500 | 1.0995 | 1.0974 | 1.0945 | 1.0923 | 1.0908 |
| 0.0500 | 0.2000 | 1.0965 | 1.0922 | 1.0931 | 1.0911 | 1.0891 |
| 0.1000 | 0.2000 | 1.1083 | 1.1049 | 1.1033 | 1.0985 | 1.0974 |
| 0.1500 | 0.2000 | 1.1142 | 1.1132 | 1.1121 | 1.1110 | 1.1081 |
| 0.0500 | 0.2500 | 1.1186 | 1.1170 | 1.1147 | 1.1180 | 1.1126 |
| ^{<i>a</i>} W _P and hydrogen certainties | W _s are th citrate s u are u(| he weight in ternary T)= ± 0.1 | fractions o solution K and u(| f PEG200 , respectiv ρ) = ± 0.1 | 0 and dian vely. Stan 1 kg·m ^{−3} . | nmonium dard un- |
| | | | | | 0 | |

polymer and salt concentration and measured refractive index value is given in eq 1. The coefficients of eq 1 were estimated by fitting the refractive index of the solution to the known weight fraction of PEG and salt present in the solutions.¹²

$$n_{\rm D} = a_0 + a_1 W_p + a_2 W_{\rm S} \tag{1}$$

where a_0 represents the refractive index of pure water at a particular temperature (data from Perry's Chemical Engineers' Handbook). Values of the constants a_1 and a_2 at different temperatures are presented in Table 1 and valid within weight fraction of polymer (W_p) = 0.05 to 0.2 and weight fraction of salt (W_S) = 0.05 to 0.2. The higher weight fraction samples were diluted accordingly, and the refractive index was measured.

The salt concentration in the top and bottom phase was determined by conductivity method, and eq 2 is used to relate the salt concentration and conductivity of the solution.

$$k = b_0 + b_1 W_{\rm S} \tag{2}$$

Table 4. Coefficients of Equation 3^a

| | $10^{3} \rho_{0}$ | | | | AARD/% | | | | | |
|-----------------------|--------------------|---------------------------|-----------|--------------------|--------------------------------------|--|--|--|--|--|
| T/K | kg·m ⁻³ | Α | В | PEG 2000 + water | diammonium hydrogen citrate + water | PEG 2000 + diammonium hydrogen citrate + water | | | | |
| 298.15 | 0.997 | 0.1742 | 0.4486 | 0.1059 | 0.0756 | 0.1766 | | | | |
| 303.15 | 0.9956 | 0.1733 | 0.4492 | 0.0736 | 0.063 | 0.1716 | | | | |
| 308.15 | 0.994 | 0.1717 | 0.4508 | 0.0422 | 0.0888 | 0.1707 | | | | |
| 313.15 | 0.9922 | 0.1693 | 0.4533 | 0.0432 | 0.0724 | 0.2055 | | | | |
| 318.15 | 0.9902 | 0.1684 | 0.4561 | 0.0535 | 0.0412 | 0.1166 | | | | |
| ^a Standard | uncertaint | ies <i>u</i> are <i>u</i> | (T) = + 0 | 1 K and $u(a) = +$ | $0.1 \text{ kg} \cdot \text{m}^{-3}$ | | | | | |

Table 5. Viscosities of the PEG2000 + Water and Diammonium Hydrogen Citrate + Water Systems at Various Temperatures^a

| | | | aqu | eous PEC | G 2000 solu | tion | viscosity, µ | ı/mPa∙s | s | |
|-----------------|----------------|-----------------|--------|----------|-------------|-------|---------------|-----------|---------------|-------|
| И | / _P | 298.15 | K | 303.15 | K 308.1 | 5 K | 313.15 | K | 318.1 | 5 K |
| 0.00 | 000 | 0.890 | 0 | 0.8010 | 0.72 | 230 | 0.65 | 60 | 0.59 | 990 |
| 0.05 | 500 | 1.868 | 0 | 1.5870 | 1.3 | 169 | 1.08 | 58 | 1.00 | 077 |
| 0.10 | 000 | 2.539 | 5 | 2.0752 | 2 1.62 | 276 | 1.25 | 79 | 1.26 | 646 |
| 0.15 | 500 | 3.255 | 8 | 2.5903 | 3 1.93 | 385 | 1.36 | 93 | 1.55 | 572 |
| 0.20 | 000 | 4.374 | 6 | 3.4693 | 3 2.52 | 274 | 1.59 | 80 | 2.05 | 545 |
| 0.25 | 500 | 6.290 | 5 | 5.0264 | 4 3.67 | 725 | 2.11 | 43 | 2.95 | 578 |
| 0.30 | 000 | 9.168 | 8 | 7.5441 | 5.67 | 740 | 3.11 | 32 | 4.45 | 512 |
| 0.35 | 500 | 13.659 | 5 | 11.4730 |) 8.80 |)67 | 4.77 | 22 | 6.74 | 453 |
| 0.40 | 000 | 19.918 | 4 | 16.8914 | 4 13.30 | 545 | 7.28 | 21 | 10.01 | 100 |
| 0.50 | 000 | 39.285 | 1 | 34.1681 | 1 27.80 | 550 | 15.58 | 17 | 19.90 | 000 |
| | | aqueous | s dian | nmonium | hydrogen c | itrat | te solution v | viscosity | $\chi, \mu/m$ | ıPa∙s |
| И | Vs | 298.15 | K | 303.15 | K 308.1 | 5 K | 313.15 | κ | 318.1 | 5 K |
| 0.0 | 000 | 0.890 | 0 | 0.8010 | 0.72 | 30 | 0.656 | 60 | 0.59 | 90 |
| 0.0 | 500 | 1.025 | 7 | 0.9257 | 0.83 | 54 | 0.783 | 55 | 0.71 | 46 |
| 0.1 | 000 | 1.146 | 0 | 1.0245 | 0.93 | 644 | 0.861 | 7 | 0.78 | 07 |
| 0.1 | 500 | 1.303 | 1 | 1.1552 | 1.05 | 572 | 0.955 | 52 | 0.83 | 55 |
| 0.2 | 000 | 1.535 | 9 | 1.3529 |) 1.22 | 49 | 1.084 | 6 | 0.92 | 67 |
| 0.2 | 500 | 1.906 | 5 | 1.6111 | 1.49 | 918 | 1.294 | 3 | 1.09 | 13 |
| 0.3 | 000 | 2.447 | 5 | 2.1048 | 3 1.87 | 86 | 1.640 |)5 | 1.38 | 27 |
| 0.3 | 500 | 3.214 | 5 | 2.7368 | 3 2.43 | 501 | 2.127 | 7 | 1.78 | 81 |
| $^{a}W_{\rm P}$ | and | $W_{\rm S}$ are | the | weight | fractions | of | aqueous | PEG2 | .000 | and |

 $W_{\rm P}$ and $W_{\rm S}$ are the weight fractions of aqueous PEG2000 and diammonium hydrogen citrate solutions, respectively. Standard uncertainties u are $u(T) = \pm 0.1$ K, $u(\mu) = \pm 0.002$ mPa·s.

where k is the conductivity (μ S·cm⁻¹); the values of b_0 and b_1 for diammonium hydrogen citrate solution are 5.744 and 502.6, respectively. The salt concentration was predicted by the equation with an accuracy of \pm 0.0001 g.

RESULTS AND DISCUSSION

Experimentally measured densities of aqueous PEG 2000 and diammonium hydrogen citrate solutions at various temperatures are reported in Table 2. The densities are found to increase with an increase in PEG 2000 and diammonium hydrogen citrate concentrations and decrease with the increase in temperature.



Figure 2. Experimental data on aqueous phase PEG2000 viscosity. \Diamond , 298.15 K; \Box , 303.15 K; \triangle , 308.15 K; ×, 313.15 K; \bigcirc , 318.15 K; dotted line indicates the literature data.⁹

A similar trend was reported in the literature for PEG 6000– triammonium citrate,¹³ PEG 4000–diammonium hydrogen phosphate,⁷ PEG 2000–sodium citrate,⁹ and PEG 200–water.⁶ Figure 1 shows linearity between density and the concentration of aqueous PEG 2000 solution and the data from literature⁹ as dotted lines. The density of the aqueous PEG 2000 was reported up to 0.25 weight fraction of PEG only by Murugesan and Perumalsamy.⁹ In this present work, the density measurement was extended up to 0.5 PEG weight fractions. Aqueous single phase ternary systems (PEG2000 + diammonium hydrogen citrate + water) were prepared by mass and measured densities are reported in Table 3. Measured densities of the solutions could be correlated using eq 3.^{7,9,13}

$$\rho/\mathrm{kg}\cdot\mathrm{m}^{-3} = AW_{\mathrm{p}} + BW_{\mathrm{S}} + \rho_{\mathrm{o}}/\mathrm{kg}\cdot\mathrm{m}^{-3}$$
(3)

where ρ and ρ_0 are the densities of binary or ternary solutions and pure water at particular temperature and W_P and W_S are the mass fractions of PEG 2000 and diammonium hydrogen citrate, respectively. The experimental density values of the binary solutions were fitted in eq 3 with a maximum deviation of \pm 0.3. Values of constants *A* and *B* and the pure water densities at

Table 6. Coefficients of Equation 4 with AARD/% Values at Different Temperatures

| | | PEG 2000 | + water | d | iammonium hydro | gen citrate + wa | iter | |
|-------------------------------------|---|---|--|---|---|--|--|--|
| $\mu_{\rm o}/{\rm mPa}\cdot{\rm s}$ | Α | В | С | AARD/% | Α | В | С | AARD/% |
| 0.8900 | 24.9890 | -132.1259 | 471.4524 | 0.1404 | 3.1458 | -12.1990 | 63.3952 | 0.0642 |
| 0.8010 | 20.7926 | -123.2330 | 430.2562 | 0.1542 | 3.0858 | -13.6289 | 59.5424 | 0.4718 |
| 0.7230 | 16.7847 | -114.7859 | 378.8997 | 0.0695 | 2.6730 | -10.5354 | 47.7316 | 0.1683 |
| 0.6560 | 11.7133 | -85.8108 | 244.1775 | 0.7443 | 3.2442 | -16.6806 | 55.6645 | 0.2172 |
| 0.5990 | 11.0035 | -67.8116 | 246.5625 | 0.2184 | 3.1946 | -19.3014 | 57.7905 | 0.3184 |
| | μ _o /mPa·s 0.8900 0.8010 0.7230 0.6560 0.5990 | μ₀/mPa·s A 0.8900 24.9890 0.8010 20.7926 0.7230 16.7847 0.6560 11.7133 0.5990 11.0035 | $\mu_{o}/mPa \cdot s$ A B 0.8900 24.9890 -132.1259 0.8010 20.7926 -123.2330 0.7230 16.7847 -114.7859 0.6560 11.7133 -85.8108 0.5990 11.0035 -67.8116 | $\begin{array}{c c c c c c c c } \hline PEG 2000 + water \\ \hline μ_{o}/mPa·s$ \hline A & B & C \\ \hline 0.8900 & 24.9890 & -132.1259 & 471.4524 \\ \hline 0.8010 & 20.7926 & -123.2330 & 430.2562 \\ \hline 0.7230 & 16.7847 & -114.7859 & 378.8997 \\ \hline 0.6560 & 11.7133 & -85.8108 & 244.1775 \\ \hline 0.5990 & 11.0035 & -67.8116 & 246.5625 \\ \hline \end{array}$ | $\begin{tabular}{ c c c c } \hline PEG 2000 + water \\ \hline μ_{o}/mPa·s$ \hline A & B & C & $AARD/\%$ \\ \hline 0.8900 & 24.9890 & -132.1259 & 471.4524 & 0.1404 \\ \hline 0.8010 & 20.7926 & -123.2330 & 430.2562 & 0.1542 \\ \hline 0.7230 & 16.7847 & -114.7859 & 378.8997 & 0.6695 \\ \hline 0.6560 & 11.7133 & -85.8108 & 244.1775 & 0.7443 \\ \hline 0.5990 & 11.0035 & -67.8116 & 246.5625 & 0.2184 \\ \hline \end{tabular}$ | $\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$ | $\begin{array}{c c c c c c c c c c c c c c c c c c c $ | $\begin{array}{c c c c c c c c c c c c c c c c c c c $ |

Article

Table 7. Viscosities of the PEG2000 + Diammonium Hydrogen Citrate + Water System at Various Temperatures^a

| | | | | µ/mPa∙s | | |
|------------|--------|---------------------|---------------------|---------------|---------------------|---------------|
| $W_{ m P}$ | Ws | <i>T</i> = 298.15 K | <i>T</i> = 303.15 K | T = 308.15 K | <i>T</i> = 313.15 K | T = 318.15 K |
| 0.0500 | 0.0500 | 2.2015 | 1.9647 | 1.5791 | 1.3027 | 1.1993 |
| 0.1000 | 0.0500 | 2.8406 | 2.5729 | 2.0018 | 1.6314 | 1.5003 |
| 0.1500 | 0.0500 | 3.4169 | 3.0500 | 2.3735 | 2.0273 | 1.8531 |
| 0.2000 | 0.0500 | 4.4030 | 3.7245 | 3.0823 | 2.6507 | 2.3179 |
| 0.2500 | 0.0500 | 6.0633 | 4.9310 | 4.1564 | 3.5777 | 3.1243 |
| 0.0500 | 0.1000 | 2.0488 | 1.8394 | 1.4927 | 1.2591 | 1.1463 |
| 0.1000 | 0.1000 | 2.7109 | 2.4062 | 1.9297 | 1.5803 | 1.4375 |
| 0.1500 | 0.1000 | 3.2924 | 2.9275 | 2.3248 | 1.9515 | 1.7544 |
| 0.2000 | 0.1000 | 4.3137 | 3.5688 | 2.8612 | 2.4989 | 2.1599 |
| 0.2500 | 0.1000 | 5.7282 | 4.6282 | 3.9574 | 3.2987 | 2.8666 |
| 0.0500 | 0.1500 | 2.0168 | 1.7792 | 1.5064 | 1.2671 | 1.1215 |
| 0.1000 | 0.1500 | 2.6913 | 2.3486 | 1.9040 | 1.5690 | 1.3948 |
| 0.1500 | 0.1500 | 3.1855 | 2.7711 | 2.2874 | 1.9093 | 1.6799 |
| 0.2000 | 0.1500 | 4.1330 | 3.4152 | 2.8038 | 2.4072 | 2.0611 |
| 0.0500 | 0.2000 | 2.1645 | 1.8771 | 1.6025 | 1.3448 | 1.1625 |
| 0.1000 | 0.2000 | 2.7684 | 2.3947 | 1.9579 | 1.6126 | 1.4072 |
| 0.1500 | 0.2000 | 3.3153 | 2.8544 | 2.3254 | 1.9249 | 1.6713 |
| 0.0500 | 0.2500 | 2.5362 | 2.0738 | 1.8204 | 1.5142 | 1.2954 |

 $^{a}W_{\rm P}$ and $W_{\rm S}$ are the weight fractions of PEG2000 and diammonium hydrogen citrate in ternary solution, respectively. Standard uncertainties u are $u(T) = \pm 0.1$ K and $u(\mu) = \pm 0.002$ mPa·s.

Table 8. Coefficients of Equation 5^a

| | T/K | а | AARD/% |
|---|------------------------|--------------------------------------|--------|
| | 298.15 | 1.8787 | 0.5220 |
| | 303.15 | 1.6284 | 0.3245 |
| | 308.15 | 1.4494 | 0.3024 |
| | 313.15 | 1.2542 | 0.1486 |
| | 318.15 | 1.1954 | 0.2753 |
| а | Standard uncertainties | s u are $u(T) = \pm 0.1 \text{ K}$ | |

uncertainties u are u(T)± 0.1 K.

Table 9. Refractive Index of the PEG 2000 + Water and Diammonium Hydrogen Citrate + Water Systems at Various Temperatures^a

| | refractive index, $n_{\rm D}$ | | | | | | |
|-------------|-------------------------------|-------------|----------------|--------------|----------|--|--|
| $W_{\rm P}$ | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K | | |
| | | PEG2000 + V | Vater Solutior | ı | | | |
| 0.0500 | 1.3390 | 1.3385 | 1.3375 | 1.3369 | 1.3360 | | |
| 0.1000 | 1.3475 | 1.3465 | 1.3455 | 1.3450 | 1.3435 | | |
| 0.1500 | 1.3540 | 1.3535 | 1.3525 | 1.3516 | 1.3505 | | |
| 0.2000 | 1.3615 | 1.3610 | 1.3600 | 1.3586 | 1.3580 | | |
| 0.2500 | 1.3685 | 1.3675 | 1.3666 | 1.3660 | 1.3645 | | |
| 0.3000 | 1.3770 | 1.3761 | 1.3750 | 1.3740 | 1.3725 | | |
| 0.3500 | 1.3840 | 1.3835 | 1.3826 | 1.3810 | 1.3800 | | |
| 0.4000 | 1.3925 | 1.3915 | 1.3900 | 1.3885 | 1.3870 | | |
| 0.5000 | 1.4075 | 1.4060 | 1.4050 | 1.4035 | 1.4015 | | |
| $W_{\rm S}$ | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K | | |
| | Diammoniu | um Hydrogen | Citrate + Wa | ter Solution | | | |
| 0.0500 | 1.3435 | 1.3430 | 1.3415 | 1.3410 | 1.3400 | | |
| 0.1000 | 1.3515 | 1.3500 | 1.3495 | 1.3485 | 1.3480 | | |
| 0.1500 | 1.3610 | 1.3605 | 1.3595 | 1.3590 | 1.3585 | | |
| 0.2000 | 1.3695 | 1.3685 | 1.3675 | 1.3670 | 1.3665 | | |
| 0.2500 | 1.3790 | 1.3780 | 1.3775 | 1.3770 | 1.3765 | | |
| 0.3000 | 1.3870 | 1.3865 | 1.3860 | 1.3855 | 1.3850 | | |
| 0.3500 | 1.3975 | 1.3970 | 1.3965 | 1.3960 | 1.3955 | | |
| 0.4000 | 1.4090 | 1.4085 | 1.4080 | 1.4075 | 1.4068 | | |
| | | | | | | | |

^{*a*}Standard uncertainties *u* are $u(T) = \pm 0.1$ K and $u(n_D) = \pm 0.00004$.

| Table 10. Refractive Index o | f the PEG 2000 + Diammonium |
|------------------------------|--|
| Hydrogen Citrate + Water S | ystem at Various Temperatures ^a |

| | | | refr | active index | , <i>n</i> _D | |
|------------------------------|-------------|--------------------|--------------|--------------|-------------------------|----------|
| $W_{ m P}$ | $W_{\rm S}$ | 298.15 K | 303.15 K | 308.15 K | 313.15 K | 318.15 K |
| 0.0500 | 0.0500 | 1.3490 | 1.3485 | 1.3475 | 1.3470 | 1.3465 |
| 0.1000 | 0.0500 | 1.3550 | 1.3545 | 1.3540 | 1.3530 | 1.3525 |
| 0.1500 | 0.0500 | 1.3635 | 1.3630 | 1.3625 | 1.3620 | 1.3615 |
| 0.2000 | 0.0500 | 1.3700 | 1.3695 | 1.3690 | 1.3685 | 1.3680 |
| 0.2500 | 0.0500 | 1.3775 | 1.3770 | 1.3765 | 1.3760 | 1.3755 |
| 0.0500 | 0.1000 | 1.3575 | 1.3570 | 1.3560 | 1.3555 | 1.3550 |
| 0.1000 | 0.1000 | 1.3645 | 1.3640 | 1.3635 | 1.3630 | 1.3625 |
| 0.1500 | 0.1000 | 1.3710 | 1.3705 | 1.3700 | 1.3695 | 1.3690 |
| 0.2000 | 0.1000 | 1.3795 | 1.3790 | 1.3780 | 1.3775 | 1.3770 |
| 0.0500 | 0.1500 | 1.3665 | 1.3660 | 1.3655 | 1.3650 | 1.3645 |
| 0.1000 | 0.1500 | 1.3735 | 1.3730 | 1.3720 | 1.3715 | 1.3710 |
| 0.1500 | 0.1500 | 1.3800 | 1.3795 | 1.3790 | 1.3785 | 1.3780 |
| 0.2000 | 0.1500 | 1.3885 | 1.3880 | 1.3875 | 1.3870 | 1.3865 |
| 0.0500 | 0.2000 | 1.3766 | 1.3765 | 1.3760 | 1.3755 | 1.3750 |
| 0.1000 | 0.2000 | 1.3825 | 1.3820 | 1.3815 | 1.3810 | 1.3805 |
| 0.1500 | 0.2000 | 1.3910 | 1.3905 | 1.3900 | 1.3850 | 1.3845 |
| ^{<i>a</i>} Standard | uncertai | nties <i>u</i> are | $u(T) = \pm$ | 0.1 K, u(| $n_{\rm D}) = \pm 0$ | .00004. |

different temperatures with corresponding AARD% are given in Table 4. Further, the densities of ternary systems were predicted by using eq 3. The maximum error between experimental and predicted densities is \pm 0.5 %.

The viscosity of the binary systems was found to decrease with an increase in temperature and is reported in Table 5. Viscosity data for the binary system were correlated by using the following polynomial equation^{6,7,13}

$$\mu/\mathrm{mPa}\cdot\mathrm{s} = AW^3 + BW^2 + CW + \mu_0/\mathrm{mPa}\cdot\mathrm{s}$$
⁽⁴⁾

where μ is the absolute viscosity of the solution and μ_0 is the viscosity of pure water at respective temperature. W is the mass fraction of either PEG 2000 or diammonium hydrogen citrate. Values of the coefficients (*A*, *B*, and *C*) are reported in Table 6

along with the AARD (%) values. Maximum deviation between calculated and predicted values for aqueous PEG 2000 and salt solution is \pm 2.5. Viscosity data for the aqueous PEG 2000 system were compared with the available data from the literature⁹ and plotted in Figure 2. Both experimental and literature data are found to be in good agreement with each other, and moreover, there is no linearity between polymer concentration and viscosity of the solution above the polymer concentration of 0.2 weight fraction.

The viscosity of the single phase ternary systems was found to decrease with increase in temperature and increase with the increase in salt and PEG2000 concentration. This behavior was observed for other ternary systems composed of PEG, salt, and water.^{7–9,13–16} Viscosities of the ternary systems in Table 7 are fitted to the Grunberg-like equation,^{7,8,13} and the values of constant *a* at different temperatures are provided in Table 8.

Table 11. Coefficients of Equation 6^a

| | | PEG 20 | 00 + water | diammonium hydrogen citrate + water | | |
|----------|--------|--------|------------|--|--------|--|
| T/K | a_0 | A | AARD/% | Α | AARD/% | |
| 298.15 K | 1.334 | 0.1433 | 0.08262 | 0.1875 | 0.0952 | |
| 303.15 K | 1.333 | 0.1442 | 0.06599 | 0.0283 | 1.3883 | |
| 308.15 K | 1.332 | 0.1444 | 0.06224 | 0.0272 | 1.3978 | |
| 313.15 K | 1.3315 | 1.2256 | 0.0252 | 0.0259 | 1.4028 | |
| 318.15 K | 1.331 | 0.14 | 0.06978 | 0.0309 | 1.402 | |
| act 11 | | · (T) | . 0 1 17 | | | |

^{*a*}Standard uncertainty *u* is $u(T) = \pm 0.1$ K.

$$\ln(\mu_{\rm m}/{\rm mPa}\cdot{\rm s}) = c_1 \ln(\mu_{\rm p}/{\rm mPa}\cdot{\rm s}) + c_2 \ln(\mu_{\rm S}/{\rm mPa}\cdot{\rm s})$$

$$+ c_1 c_2 a \tag{5}$$

where $c_1 = W_P/(W_P + W_S)$, $c_2 = W_S/(W_P + W_S)$, $\mu_{\rm m}$, μ_p , and μ_s represent the viscosity of the mixture, polymer, and salt, respectively. Relative errors between experimental viscosity and predicted values were calculated, and it was found that eq 5 predicts the viscosity of the single phase ternary solutions within the error limits of \pm 0.2 %.

Refractive index measurements of the binary and ternary single phase region were carried out, and the values are presented in Tables 9 and 10, respectively. Binary solution refractive index values were correlated by using eq 6

$$n_{\rm D} = a_{\rm o} + AW \tag{6}$$

where a_0 is the refractive index of pure water at particular temperature, *W* is the mass fraction of either PEG 2000 or diammonium hydrogen citrate, and the values of coefficient *A* and percentage of error are reported in Table 11.

Liquid—liquid equilibrium data and the top and bottom phase composition were analyzed and reported in Table 12 along with the phase volume ratio (top phase volume/bottom phase volume), tie line length, and density difference between the top and bottom phases. The tie-line length (TLL) was calculated using the following relationship, eq 7,

$$\text{TLL} = \left[(W_{\rm P}^{\rm T} - W_{\rm P}^{\rm B})^2 + (W_{\rm S}^{\rm B} - W_{\rm S}^{\rm T})^2 \right]^{1/2}$$
(7)

Phase volume ratio, tie line length, and density difference between the phases are found to increase with the increase in

Table 12. Density Difference between the Top Phase and the Bottom Phase, Phase Volume Ratio, and TLL for the PEG 2000 + Diammonium Hydrogen Citrate + Water Two-Phase System^a

| feed con | nposition | top j | ohase | botton | n phase | | | |
|-------------|-----------|---------------------|---------------------|---------------------|--------------------|--------------|--------|---|
| $W_{\rm P}$ | Ws | $W_{\rm P}^{\rm T}$ | $W_{\rm S}^{\rm T}$ | $W_{\rm P}^{\rm B}$ | $W^{\rm B}_{ m S}$ | volume ratio | TLL | $\Delta \rho = (\rho^{\rm B} - \rho^{\rm T})$ |
| | | | | T = 298.1 | 5 K | | | |
| 0.2000 | 0.2200 | 0.3330 | 0.1226 | 0.1740 | 0.2437 | 1.4655 | 0.1999 | 0.0252 |
| 0.2200 | 0.2200 | 0.3630 | 0.1087 | 0.1560 | 0.2725 | 1.6176 | 0.2640 | 0.0389 |
| 0.2500 | 0.2200 | 0.4350 | 0.0675 | 0.1560 | 0.2968 | 1.7269 | 0.3611 | 0.0486 |
| 0.2800 | 0.2200 | 0.4870 | 0.0486 | 0.1560 | 0.3200 | 1.8307 | 0.4280 | 0.0595 |
| | | | | T = 303.1 | 5 K | | | |
| 0.2000 | 0.2200 | 0.3560 | 0.1000 | 0.1550 | 0.2571 | 1.6159 | 0.2551 | 0.0452 |
| 0.2200 | 0.2200 | 0.4170 | 0.0680 | 0.1400 | 0.2854 | 1.7654 | 0.3460 | 0.0554 |
| 0.2500 | 0.2200 | 0.4690 | 0.0486 | 0.1380 | 0.3097 | 1.8452 | 0.4216 | 0.0662 |
| | | | | T = 308.1 | 5 K | | | |
| 0.1800 | 0.2200 | 0.3550 | 0.0903 | 0.1380 | 0.2556 | 1.6981 | 0.2728 | 0.0388 |
| 0.2000 | 0.2200 | 0.3860 | 0.0769 | 0.1340 | 0.2734 | 1.6604 | 0.3270 | 0.0479 |
| 0.2200 | 0.2200 | 0.4290 | 0.0591 | 0.1280 | 0.2938 | 1.6667 | 0.3817 | 0.0649 |
| | | | | T = 313.1 | 5 K | | | |
| 0.1600 | 0.2200 | 0.3380 | 0.0938 | 0.1412 | 0.2413 | 1.3014 | 0.2567 | 0.0375 |
| 0.1800 | 0.2200 | 0.3940 | 0.0690 | 0.1402 | 0.2601 | 1.3907 | 0.3264 | 0.0453 |
| 0.2000 | 0.2200 | 0.4600 | 0.0422 | 0.1377 | 0.2821 | 1.4558 | 0.4159 | 0.0555 |
| 0.2200 | 0.2200 | 0.4860 | 0.0333 | 0.1368 | 0.2909 | 1.5806 | 0.4539 | 0.0575 |
| | | | | T = 318.1 | 5 K | | | |
| 0.1400 | 0.2200 | 0.3400 | 0.0809 | 0.1170 | 0.2387 | 0.8410 | 0.2732 | 0.0303 |
| 0.1600 | 0.2200 | 0.3740 | 0.0640 | 0.1150 | 0.2500 | 1.0365 | 0.3223 | 0.0386 |
| 0.1800 | 0.2200 | 0.4130 | 0.0481 | 0.0990 | 0.2804 | 1.1837 | 0.3906 | 0.0544 |
| 0.2000 | 0.2200 | 0.4510 | 0.0352 | 0.1030 | 0.2888 | 1.4966 | 0.4306 | 0.0554 |
| 0.2200 | 0.2200 | 0.4870 | 0.0288 | 0.1000 | 0.3032 | 1.3258 | 0.4800 | 0.0626 |

 ${}^{a}W_{\rm P}$ and $W_{\rm S}$ are the weight fractions of PEG 2000 and diammonium citrate in the ternary systems, and the superscripts T and B correspond to the top phase and bottom phase. Standard uncertainties *u* are $u(T) = \pm 0.1$ K, $u(W) = \pm 0.0001$, and $u(\rho) = \pm 0.1$ kg·m⁻³.

| | | | top | o phase | | | | | | | botte | m phase | | | |
|--|------|--------------------------|-------------------------------|---------|-----------------------------------|---------------------|--|---------------------|---------------------|--------------------|----------------------------|---------------|-----------------------|---------------------|--|
| | | $10^3 \rho^{\mathrm{T}}$ | $10^3 \rho^{\rm T}_{\rm cal}$ | μ | $\mu^{\mathrm{T}}_{\mathrm{cal}}$ | | | | | $10^3 ho^{ m B}$ | $10^3 ho^{ m B}_{ m cal}$ | $\mu^{\rm B}$ | $\mu^{ m B}_{ m cal}$ | | |
| T=298.15 K $T=298.15 K$ $T=298.15 K$ $T=1011 1100 0.3391 8.9015 1.4010 1.4015 0.1740 0.2457 1.1341 1.1466 3.9109 3.9073 1.402 1.4002 1.4002 1.4010 1.0574 1.106 1.4010 1.4012 0.1560 0.2368 1.1361 1.1366 3.9109 3.9073 1.402 1.4012 1.4016 1.4010 1.012 0.1157 1.102 0.1157 4.125 4.125 4.125 1.4012 1.4010 1.4010 1.4010 1.012 0.1091 1.1072 0.1109 0.1560 0.1560 0.1560 1.1577 4.126 4.1278 1.4016 1.4010 1.4010 1.001 1.001 1.0073 0.1501 0.1500 0.1560 0.2368 1.1360 1.1573 4.126 4.1278 1.4016 1.4010 1.4010 1.001 1.001 1.002 1.0031 1.102 0.11091 0.1100 0.1560 0.2361 1.1370 1.1360 3.1262 0.1306 1.4010 1.4001 1.4011 1.002 1.0031 1.1022 0.1104 0.1100 0.1560 0.2361 1.1370 1.1380 0.1262 0.1306 1.4010 1.4001 1.4011 1.003 1.0037 0.10931 1.1370 1.1380 0.1092 0.10941 0.02331 1.4012 0.1091 1.1360 1.1032 0.10941 1.4002 0.1391 1.1370 1.1380 0.1092 1.1093 1.0037 1.0031 1.136 1.1390 0.10930 1.1093 1.1093 1.1093 0.10941 1.4002 0.1381 1.1360 1.1380 0.10941 1.1093 1.1392 0.1306 1.4001 1.4011 1.4011 1.1093 1.1093 0.10941 1.0931 1.1093 1.1093 0.12331 1.1093 0.10940 0.2331 1.1149 0.12341 1.1093 1.1093 1.1093 1.1093 1.1094 1.4000 1.1001 1.308 0.1380 0.1099 0.2338 1.1493 1.1139 1.1329 0.2306 1.1395 1.1396 1.4001 1.4011 1.0041 1.4032 0.1340 0.2351 1.1143 1.1339 0.2306 1.3903 1.4001 1.4011 1.401 0.1341 1.138 1.1339 0.2306 1.3935 1.4001 1.4012 0.1394 1.1001 0.1301 1.1000 1.1000 1.1000 1.1000 1.1000 1.1000 1.1001 1.1001 1.$ | | kg·m ⁻³ | kg·m ⁻³ (eq 3) | mPa•s | mPa·s (eq 5) | $n_{\rm D}^{\rm T}$ | $n_{\mathrm{D,cal}}^{\mathrm{T}}$ (eq 1) | $W^{\rm B}_{\rm P}$ | $W^{\rm B}_{\rm S}$ | kg·m ⁻³ | kg·m ⁻³ (eq 3) | mPa·s | mPa·s (eq 5) | $n_{\rm D}^{\rm B}$ | $n_{\mathrm{D,cal}}^{\mathrm{B}}$ (eq 1) |
| | | | | | | | T = 298 | 8.15 K | | | | | | | |
| 87 11081 11090 116774 11.6862 1.4003 0.13503 1.471 1.1464 3.8879 1.4028 1.4038 1.4038 1.4038 1.4038 1.4038 1.4038 1.4038 1.4038 1.4038 1.4038 1.4031 1.4038 1.4031 1.4038 1.4031 1.4038 1.4031 1.4061 1.1033 1.4136 1.1437 1.1436 1.1436 1.4136 1.1437 1.4136 1.4136 1.4106 1.4001 1.4016 1.4016 1.4013 1.1436 1.1433 1.1437 1.1436 1.1436 1.1416 1.4106 1.4016 1.4016 0.1380 0.3309 1.1657 1.1436 1.4106 1.4016 1.4016 0.3364 1.4136 1.1436 1.4106 1.4016 1.4016 1.4038 1.4038 1.4036 1.4016 1.4038 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 1.4036 | 226 | 1.1091 | 1.1100 | 9.3591 | 8.9015 | 1.4010 | 1.4015 | 0.1740 | 0.2437 | 1.1343 | 1.1366 | 3.9109 | 3.9073 | 1.4002 | 1.4003 |
| | 87 | 1.1081 | 1.1090 | 11.6774 | 11.6862 | 1.4032 | 1.4032 | 0.1560 | 0.2725 | 1.1471 | 1.1464 | 3.8879 | 3.8791 | 1.4028 | 1.4028 |
| 1105 1103 30.5619 30.5616 14107 14100 0.1550 0.3201 11.677 44.63 44.162 14.110 14.110 0 10918 11.022 91104 91119 14005 0.1550 0.2551 11.370 11.380 31.262 31.405 1.4003 0 10918 11.022 91104 91119 14005 0.4905 0.5561 11.470 11.380 31.262 1.4003 1.4003 0 10915 10987 2.28221 2.28377 1.4005 0.4300 0.3097 1.1576 11.380 31.965 1.4071 1.4073 0 10091 1.4902 0.4301 0.3097 1.1576 1.1586 3.405 1.4071 1.4073 0 10090 10949 10.2329 1.4012 1.4012 0.1340 0.2573 1.4075 1.4073 1.4073 0 10090 10949 1.4025 0.1340 0.2536 1.1313 1.1329 2.5026 | 575 | 1.1095 | 1.1031 | 20.7752 | 20.7583 | 1.4071 | 1.4061 | 0.1560 | 0.2968 | 1.1580 | 1.1573 | 4.1426 | 4.1278 | 1.4076 | 1.4070 |
| $ \begin{array}{llllllllllllllllllllllllllllllllllll$ | 486 | 1.1056 | 1.1036 | 30.5619 | 30.5616 | 1.4107 | 1.4100 | 0.1560 | 0.3200 | 1.1652 | 1.1677 | 4.4263 | 4.4162 | 1.4110 | 1.4110 |
| | | | | | | | T = 303 | 3.15 K | | | | | | | |
| 680 1.092 1.0984 15.0955 15.0947 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.403 1.407 | 1000 | 1.0918 | 1.1022 | 9.1104 | 9.1119 | 1.4005 | 1.4008 | 0.1550 | 0.2571 | 1.1370 | 1.1380 | 3.1262 | 3.1264 | 1.4000 | 1.4004 |
| 446 10915 10987 22.8271 1.4072 1.4075 0.1380 0.3097 1.1576 1.1586 3.4026 3.4035 1.4071 1.4073 7003 10925 109497 7.9460 7.9451 1.4010 1.3982 0.1330 0.2556 1.1313 1.1329 2.5026 1.3955 1.3963 7903 10909 1.0949 10.2329 1.4001 1.4002 0.13328 1.4013 1.3952 0.1393 1.3953 1.3953 1.3953 1.3953 1.3953 1.3953 1.3953 1.3953 1.4012 0.2413 1.1337 1.1328 1.4708 1.4303 1.3954 1.3954 918 10860 1.0919 4.7989 1.3953 0.1402 0.2601 1.1441 1.1338 1.4028 1.3954 1.4002 918 10860 1.9892 1.4038 0.1377 0.2831 1.1425 1.4708 1.4023 1.4003 918 10882 1.0892 1.38239 1.4033 1. | 0890 | 1.0922 | 1.0984 | 15.0955 | 15.0947 | 1.4039 | 1.4037 | 0.1400 | 0.2854 | 1.1476 | 1.1481 | 3.1936 | 3.1967 | 1.4035 | 1.4033 |
| T = 308.15 K9031.09251.09577.94607.94511.40101.39820.13800.25561.13131.13292.50261.39551.39551.395575911.09491.0232910.23281.40041.40020.13400.27541.13891.14032.57732.57251.39851.395555911.09491.0232910.23281.40041.40020.13400.27541.13891.14032.57721.39851.400505011.09491.023291.40251.40020.139570.12800.29381.14931.14842.64272.64291.40051.394005011.09027.80971.39591.39570.112611.13441.13381.53201.39401.394005011.09027.80977.80971.39591.40380.13770.23111.112371.13381.53931.400505021.08871.08861.70841.39290.13070.23871.112371.13381.55931.400205031.08871.08847.15007.132391.40380.13770.23871.114251.14721.61771.39861.400205041.08841.08847.15007.13231.39230.13770.23871.114251.14721.65931.400205041.08841.08847.15007.15121.39401.39470.11700.23871.11721.11881.59331.3910 <td>0486</td> <td>1.0915</td> <td>1.0987</td> <td>22.8221</td> <td>22.8377</td> <td>1.4072</td> <td>1.4075</td> <td>0.1380</td> <td>0.3097</td> <td>1.1576</td> <td>1.1586</td> <td>3.4026</td> <td>3.4085</td> <td>1.4071</td> <td>1.4073</td> | 0486 | 1.0915 | 1.0987 | 22.8221 | 22.8377 | 1.4072 | 1.4075 | 0.1380 | 0.3097 | 1.1576 | 1.1586 | 3.4026 | 3.4085 | 1.4071 | 1.4073 |
| 0903 10925 10957 7.9460 7.9451 1.4010 1.3932 0.1380 0.2556 1.3132 1.1329 2.5026 2.3025 1.3953 1.3953 0769 10909 10949 10.2329 10.2328 1.4004 1.4002 0.1340 0.2734 1.1389 1.1403 2.5273 2.3955 1.3993 0591 1.0844 1.0943 14.5922 1.45357 1.4003 0.1340 0.2734 1.1389 1.1403 2.5273 2.5372 1.3985 1.4002 0501 1.0919 4.7986 1.4035 0.1320 0.1327 0.1393 1.4934 1.6177 1.3940 1.3940 1.3940 0500 1.0892 1.38237 1.3933 0.1377 0.2401 1.1434 1.6170 1.4025 1.4005 0401 1.0892 1.38237 1.4039 0.1377 0.2811 1.1434 1.6170 1.3940 1.3940 0402 1.0887 1.0892 1.4035 0.1368 | | | | | | | T = 308 | 8.15 K | | | | | | | |
| 7769 1.0949 102329 102328 1.4004 1.4002 0.1340 0.2734 1.1389 1.1434 2.5773 2.5772 1.3955 1.4026 7804 1.0943 14.5587 1.4032 0.1280 0.2938 1.1493 1.1434 2.6427 2.6429 1.4025 1.4026 7809 1.4926 4.7986 4.7986 4.7986 1.3957 0.1412 0.2413 1.1237 1.1235 1.4708 1.3940 9316 1.0902 7.8090 7.8097 1.3957 0.1402 0.2413 1.1344 1.1338 1.3320 1.3940 9420 1.0892 1.0912 7.8090 7.8097 1.39292 0.1402 0.2801 1.1425 1.1472 1.6170 1.6177 1.3957 9420 1.0892 1.0892 $1.7.1091$ $1.7.1089$ 1.4038 0.1377 0.2801 1.1425 1.1472 1.6170 1.6177 1.3956 1.407 9407 1.0887 1.4039 1.4039 0.1170 0.2387 1.1422 1.1472 1.6593 1.4020 1.407 841 1.0844 5.4084 1.38239 1.4039 0.1170 0.2387 1.1422 1.1472 1.6593 1.4020 1.407 8409 1.0846 5.4094 1.3916 1.3924 1.1422 1.1434 1.6170 1.4020 1.407 8409 1.0846 5.4094 1.39316 1.3916 1.1462 1.1422 </td <td>903</td> <td>1.0925</td> <td>1.0957</td> <td>7.9460</td> <td>7.9451</td> <td>1.4010</td> <td>1.3982</td> <td>0.1380</td> <td>0.2556</td> <td>1.1313</td> <td>1.1329</td> <td>2.5026</td> <td>2.5026</td> <td>1.3955</td> <td>1.3967</td> | 903 | 1.0925 | 1.0957 | 7.9460 | 7.9451 | 1.4010 | 1.3982 | 0.1380 | 0.2556 | 1.1313 | 1.1329 | 2.5026 | 2.5026 | 1.3955 | 1.3967 |
| 591 1.0844 1.0943 $14,5922$ $14,587$ $14,035$ $14,032$ 0.1280 0.2938 $11,494$ 2.6427 2.6429 14025 14025 938 1.0862 1.0919 4.7986 4.7989 1.3957 0.1412 0.2413 11.237 11.255 1.4708 1.3940 1.3940 6690 1.0902 7.8090 7.8097 1.3929 0.1377 0.2413 11.237 1.1235 1.4708 1.3940 1.3940 670 1.0902 7.8090 7.8090 1.39292 0.1402 0.2601 11.442 1.1338 1.5313 1.3960 1.3971 670 1.0892 1.38237 1.38239 1.4038 0.1377 0.2821 11.425 1.1434 1.6170 1.6177 1.3936 640 1.0892 1.38237 1.38239 1.4038 0.1377 0.2387 11.425 1.1472 1.6593 1.4020 7809 1.0896 1.71091 1.71089 1.4059 0.1405 0.1368 0.2309 1.1472 1.6793 1.4020 8809 1.0846 5.4094 1.3932 1.4032 0.1170 0.2387 1.1172 1.188 1.5593 1.4020 881 1.0791 1.0817 9.8069 1.3947 0.1120 0.2380 1.1226 1.5840 1.3942 1.3912 881 1.0791 1.0817 9.8069 1.3947 0.1350 0.2300 1.1226 1.5840 1.3947 | 6920 | 1.0909 | 1.0949 | 10.2329 | 10.2328 | 1.4004 | 1.4002 | 0.1340 | 0.2734 | 1.1389 | 1.1403 | 2.5273 | 2.5272 | 1.3985 | 1.3993 |
| T = $313.15 \mathrm{K}$ 938 1.0862 1.0919 4.7986 4.7989 1.3957 0.1412 0.2413 1.125 1.4708 1.4708 1.3940 1.3940 690 1.0890 1.0902 7.8090 7.8097 1.3992 0.1402 0.2413 1.125 1.4708 1.4708 1.3940 1.3971 650 1.0802 7.8090 7.8097 1.3993 1.3922 0.1402 0.2601 1.1344 1.1338 1.5313 1.3960 1.3971 6333 1.0887 1.0892 17.1091 17.1089 1.4038 0.1377 0.2821 1.1422 1.1472 1.6170 1.6177 1.3985 1.4007 640 1.0896 1.71091 17.1089 1.4038 0.1368 0.23909 1.1442 1.1472 1.6595 1.4020 1.3912 640 1.0847 5.4094 1.3932 1.3930 0.1170 0.2387 1.1172 1.1188 1.5593 1.4020 1.3912 640 1.0817 9.8069 1.3940 1.3947 0.1170 0.2387 1.1172 1.1236 1.5840 1.3916 1.3912 640 1.0817 9.8097 9.8069 1.3947 0.1150 0.2500 1.1223 1.1236 1.5840 1.3945 1.3912 641 1.0817 9.8097 9.8069 1.3940 1.3947 1.1334 1.1348 1.6374 1.3945 1.3945 6421 1.0817 1.081 | 1650 | 1.0844 | 1.0943 | 14.5922 | 14.5587 | 1.4035 | 1.4032 | 0.1280 | 0.2938 | 1.1493 | 1.1484 | 2.6427 | 2.6429 | 1.4025 | 1.4020 |
| 938 1.0862 1.0919 4.7986 4.7980 1.3957 0.1412 0.2413 1.1237 1.1255 1.4708 1.4708 1.3440 1.3940 1.3940 690 1.0890 1.0902 7.8090 7.8097 1.3992 0.1402 0.2601 1.1344 1.1338 1.5320 1.5313 1.3960 1.3971 733 1.0870 1.0892 13.8239 1.4035 1.4038 0.1377 0.2821 1.1425 1.1434 1.6170 1.6177 1.3985 1.4007 333 1.0887 1.0892 17.1091 17.1089 1.4038 0.1377 0.2821 1.1425 1.1472 1.6177 1.3985 1.4007 333 1.0887 1.0892 17.1089 1.4039 0.1377 0.2821 1.1425 1.1472 1.6177 1.3985 1.4007 333 1.0887 1.0896 $1.7.1091$ 17.1089 1.4059 0.1377 0.2387 1.1425 1.1472 1.6177 1.3912 1.0849 1.0844 5.4084 1.3924 1.3923 1.4002 0.13974 0.1170 0.2387 1.1172 1.1172 1.188 1.5593 1.4020 1.3916 1.0791 1.0817 9.8069 1.3944 1.3944 0.1123 1.1122 1.1126 1.5840 1.5843 1.3916 1.3912 1.0791 1.0817 9.8069 1.3944 1.3947 0.1123 1.1236 1.5840 1.3946 1.3945 | | | | | | | T = 313 | 3.15 K | | | | | | | |
| 660108001.09027.80907.80971.39331.39220.14020.26011.13441.13381.53101.53131.39601.39717422108701.089213823713.823713.8239140581.40380.13770.28211.14251.14341.61701.61771.39851.400773331.08871.089617.109117.10891.40591.40590.13680.29091.14621.14721.65951.65931.40201.40218091.08691.08445.40865.40941.39320.31700.23871.11721.11881.55931.39101.39196401.08179.80979.80691.39741.39470.11700.25001.12331.13481.58331.39161.39196401.08179.80979.80691.39741.39730.10300.28881.13341.13481.63741.39451.39196411.07911.08179.80979.80691.39741.39731.13231.13481.63761.39451.39196421.08179.80979.80691.39741.39731.13231.13481.63761.39451.39196431.07911.08179.80979.80691.39741.39731.13231.13481.63761.39451.39196441.07911.08179.80979.80691.39741.39731.13731.13481.69741.69761.3945 <td>938</td> <td>1.0862</td> <td>1.0919</td> <td>4.7986</td> <td>4.7989</td> <td>1.3959</td> <td>1.3957</td> <td>0.1412</td> <td>0.2413</td> <td>1.1237</td> <td>1.1255</td> <td>1.4708</td> <td>1.4708</td> <td>1.3940</td> <td>1.3940</td> | 938 | 1.0862 | 1.0919 | 4.7986 | 4.7989 | 1.3959 | 1.3957 | 0.1412 | 0.2413 | 1.1237 | 1.1255 | 1.4708 | 1.4708 | 1.3940 | 1.3940 |
| -422 10870 1.0892 13.8237 13.8239 1.4035 1.4038 0.1377 0.2821 1.1425 1.1434 1.6170 1.6177 1.3955 1.4007 3333 1.0887 1.0896 17.1091 17.1089 1.4059 0.1368 0.2909 1.1462 1.1472 1.6595 1.6593 1.4020 1.4021 3809 1.0869 1.0869 $1.7.1091$ 17.1089 1.4059 0.1368 0.2387 1.1172 1.1188 1.5593 1.3910 1.3911 3040 1.0817 9.8069 1.3940 1.3947 0.1170 0.2387 1.1172 1.1188 1.5593 1.3910 1.3911 3041 1.0824 7.1520 7.1512 1.3940 1.3947 0.1150 0.2500 1.1223 1.1188 1.5840 1.3912 1.3919 3041 1.0817 9.8069 1.3974 1.3947 0.1130 0.2804 1.1334 1.1348 1.6374 1.3945 1.3919 3022 1.0817 9.8097 9.8069 1.3974 1.3973 1.1323 1.1348 1.6974 1.3945 1.3912 3028 1.0822 1.32117 13.2115 1.4010 1.4003 0.1030 0.2888 1.1373 1.16976 1.3945 1.3945 3028 1.0826 1.0837 1.0838 1.7038 1.7038 1.4042 0.1000 0.3032 1.1453 1.7699 1.3970 1.3970 3 | 0690 | 1.0890 | 1.0902 | 7.8090 | 7.8097 | 1.3993 | 1.3992 | 0.1402 | 0.2601 | 1.1344 | 1.1338 | 1.5320 | 1.5313 | 1.3960 | 1.3971 |
| 0333 1.0897 1.0896 17.1091 17.1089 1.4059 1.4059 0.1368 0.2909 1.1462 1.1472 1.6595 1.6593 1.4020 1.4021 0304 1.0869 1.0844 5.4086 5.4094 1.3930 0.1170 0.2387 1.1172 1.1188 1.5593 1.3910 1.3910 05640 1.0837 1.0817 9.8097 9.13940 1.3947 0.1170 0.2387 1.1123 1.1236 1.5840 1.3910 1.3910 05640 1.0817 9.8097 9.13940 1.3947 0.1150 0.2387 1.1123 1.1236 1.5840 1.3910 1.3919 0481 1.0791 1.0817 9.8097 9.8069 1.3974 1.3928 1.3913 1.1326 1.5840 1.5843 1.3916 1.3919 0551 1.0791 1.0817 9.8097 9.8069 1.3974 1.3938 1.3928 1.3912 1.3912 1.3912 1.3919 1.3919 1.3919 1.3919 | 0422 | 1.0870 | 1.0892 | 13.8237 | 13.8239 | 1.4035 | 1.4038 | 0.1377 | 0.2821 | 1.1425 | 1.1434 | 1.6170 | 1.6177 | 1.3985 | 1.4007 |
| $T = 318.15 \mathrm{K}$ 5400 1.0869 1.0844 5.4086 5.4094 1.3932 1.3930 0.1170 0.2387 1.1172 1.1188 1.5593 1.5593 1.3910 1.3910 0540 1.0837 1.0824 7.1520 7.1512 1.3940 1.3947 0.1170 0.2387 1.1172 1.1188 1.5593 1.3910 1.3910 0540 1.0837 1.0824 7.1520 7.1512 1.3940 1.3947 0.1150 0.2380 1.1236 1.5840 1.5833 1.3912 1.3919 0481 1.0791 1.0817 9.8097 9.8069 1.3974 1.3973 1.1334 1.1348 1.6368 1.6364 1.3945 1.3951 0522 1.0819 1.0822 1.3.2115 1.4010 1.4003 0.1030 0.2888 1.1373 1.1393 1.6974 1.3976 1.3973 0288 1.0826 1.70358 1.4042 0.1000 0.3032 1.1453 1.1453 1.7699 1.3970 1.3973 1.3810 0.3032 1.1453 1.7693 1.3990 1.3973 < |)333 | 1.0887 | 1.0896 | 17.1091 | 17.1089 | 1.4059 | 1.4059 | 0.1368 | 0.2909 | 1.1462 | 1.1472 | 1.6595 | 1.6593 | 1.4020 | 1.4021 |
| 3809 1.0869 1.0844 5.4086 5.4094 1.3922 1.3930 0.1170 0.1172 1.1188 1.5593 1.3910 1.3910 1.3910 0540 1.0837 1.0824 7.1520 7.1512 1.3940 1.3947 0.1150 0.2380 1.1236 1.5833 1.3925 1.3919 0540 1.0817 9.8097 9.8069 1.3974 1.3973 0.0990 0.2804 1.1334 1.6368 1.6364 1.3945 1.3951 0352 1.0819 1.0817 9.8097 9.8069 1.3974 1.3934 1.1348 1.6368 1.3945 1.3951 0352 1.0819 1.0817 9.8097 9.8069 1.4003 0.1030 0.2888 1.1373 1.1368 1.6976 1.3970 1.3971 0352 1.0816 1.0853 1.70358 1.4042 0.1000 0.3032 1.1453 1.7699 1.3970 1.3973 | | | | | | | T = 318 | 8.15 K | | | | | | | |
| 0640 1.0837 1.0824 7.1520 7.1512 1.3940 1.3947 0.1150 0.2500 1.1236 1.5840 1.5833 1.3925 1.3919 0481 1.0791 1.0817 9.8097 9.8069 1.3974 1.3973 0.0990 0.2804 1.1334 1.16368 1.5644 1.3945 1.3951 0352 1.0819 1.0817 9.8097 9.8069 1.3974 0.4003 0.2888 1.1373 1.1348 1.6974 1.3970 1.3971 0352 1.0819 1.0822 13.2117 13.2115 1.4010 1.4003 0.1030 0.2888 1.1373 1.1393 1.6974 1.6976 1.3970 1.3972 0288 1.0826 1.0853 17.0358 1.4042 0.1000 0.3032 1.1453 1.7693 1.3990 1.3993 1.3990 1.3993 | 6080 | 1.0869 | 1.0844 | 5.4086 | 5.4094 | 1.3932 | 1.3930 | 0.1170 | 0.2387 | 1.1172 | 1.1188 | 1.5593 | 1.5593 | 1.3910 | 1.3901 |
| 0481 1.0791 1.0817 9.8097 9.8069 1.3974 1.3973 0.0990 0.2804 1.1348 1.6368 1.6364 1.3945 1.3951 0352 1.0819 1.0822 13.2117 13.2115 1.4010 1.4003 0.1030 0.2888 1.1373 1.1393 1.6976 1.3970 1.3972 0288 1.0826 1.0822 13.2117 13.2115 1.4010 1.4003 0.1030 0.2888 1.1373 1.6974 1.6976 1.3970 1.3972 0288 1.0826 1.0853 17.0358 1.4048 0.1000 0.3032 1.1453 1.7693 1.3990 1.3993 | 0640 | 1.0837 | 1.0824 | 7.1520 | 7.1512 | 1.3940 | 1.3947 | 0.1150 | 0.2500 | 1.1223 | 1.1236 | 1.5840 | 1.5833 | 1.3925 | 1.3919 |
| 3352 1.0819 1.0822 13.2117 13.2115 1.4010 1.4003 0.1030 0.2888 1.1373 1.1393 1.6974 1.6976 1.3970 1.3972 2288 1.0826 1.0853 17.0358 1.4048 1.4042 0.1000 0.3032 1.1453 1.7693 1.3990 1.3993 | 0481 | 1.0791 | 1.0817 | 9.8097 | 9.8069 | 1.3974 | 1.3973 | 0.0990 | 0.2804 | 1.1334 | 1.1348 | 1.6368 | 1.6364 | 1.3945 | 1.3951 |
| 0288 1.0826 1.0853 17.0358 17.0358 1.4048 1.4042 0.1000 0.3032 1.1453 1.1453 1.7693 1.7699 1.3990 1.3993 | 0352 | 1.0819 | 1.0822 | 13.2117 | 13.2115 | 1.4010 | 1.4003 | 0.1030 | 0.2888 | 1.1373 | 1.1393 | 1.6974 | 1.6976 | 1.3970 | 1.3972 |
| | 0288 | 1.0826 | 1.0853 | 17.0358 | 17.0358 | 1.4048 | 1.4042 | 0.1000 | 0.3032 | 1.1453 | 1.1453 | 1.7693 | 1.7699 | 1.3990 | 1.3993 |

PEG concentration and increase with temperature for a particular PEG and salt concentration.^{9,17,18} Experimentally measured and predicted density, viscosity, and refractive index of the top and bottom phases are in Table 13. Both experimentally estimated and predicted top phase viscosities were plotted against TLL in Figure 3. Predicted values were found to be in good agreement with the experimental values.



Figure 3. Effect of TLL on top phase viscosity: \Diamond , 298.15 K; \Box , 303.15 K; \triangle , 308.15 K; ×, 313.15 K; O, 318.15 K (dashed line, calculated using eq 5).

CONCLUSIONS

The densities, refractive index, and viscosities of the binary and ternary mixtures of the PEG 2000 + diammonium hydrogen citrate + water based aqueous two-phase system were measured and correlated for five different temperatures, (298.15, 303.15, 308.15, 313.15, and 318.15) K. Further, the TLL were calculated for the chosen aqueous two-phase system at the same temperature, and phase composition, density, viscosity, and the refractive index of the individual phases were measured and predicted using the developed correlations. All of the correlations were predicted, and their respective properties had low relative error. The top phase viscosity with respect to temperature and TLL was also analyzed and plotted.

AUTHOR INFORMATION

Corresponding Author

*E-mail: regupathi@nitk.ac.in. Fax: 0824-2474057.

Funding

The authors acknowledge a grant (Scheme No. 01(2339)/09/ EMR-II) from the Council of Scientific and Industrial Research (CSIR), Government of India, for this research.

Notes

The authors declare no competing financial interest.

REFERENCES

(1) Hemavathi, A. B.; Raghavarao, K. S. M. S. Differential partitioning of β -galactosidase and β -glucosidase using aqueous two phase extraction. *Process Biochem.* **2011**, *46*, 649–655.

(2) Raghavarao, K. S. M. S.; Ruinn, M. R.; Todd, P. Recent developments in aqueous two-phase extraction in bioprocessing. *Sep. Purif. Methods* **1998**, *27*, 1–49.

(3) Rawdkuen, S.; Pintathong, P.; Chaiwut, P.; Benjakul, S. The partitioning of protease from *Calotropis procera* latex by aqueous two-phase systems and its hydrolytic pattern on muscle proteins. *Food Bioprod. Process* **2011**, *89*, 73–80.

(4) Yucekan, I.; Onal, S. Partitioning of invertase from tomato in poly(ethylene glycol)/sodium sulfate aqueous two-phase systems. *Process Biochem.* **2011**, *46*, 226–232.

(5) Malpiedi, L. P.; Guillermo, A. P.; Bibiana, B. N. Studies of protein partition in nonconventional aqueous two-phase systems as method to purify trypsinogen and alpha-chymotrypsinogen from bovine pancreas. *Sep. Purif. Technol.* **2011**, *78*, 91–96.

(6) Rahbari-Sisakht, M.; Taghizadeh, M.; Eliassi, A. Densities and Viscosities of Binary Mixtures of Poly(ethylene glycol) and Poly(propylene glycol) in Water and Ethanol in the 293.15–338.15 K Temperature Range. J. Chem. Eng. Data 2003, 48, 1221–1224.

(7) Regupathi, I.; Murugesan, S.; Amaresh, S. P.; Govindarajan, R.; Thanabalan, M. Densities and Viscosities of Poly(ethylene glycol) 4000 + Diammonium Hydrogen Phosphate + Water Systems. *J. Chem. Eng. Data* 2009, *54*, 1100–1106.

(8) Telis-Romero, J.; Coimbra, J. S. R.; Gabas, A. L.; Garcia Rojas, E. E.; Minim, L. A.; Telis, V. R. N. Dynamic Viscosity of Binary and Ternary Mixtures Containing Poly(Ethylene Glycol), Potassium Phosphate, and Water. J. Chem. Eng. Data **2004**, 49, 1340–1343.

(9) Murugesan, T.; Perumalsamy, M. Densities and Viscosities of Polyethylene Glycol 2000 + Salt + Water Systems from (298.15 to 318.15) K. J. Chem. Eng. Data **2005**, 50, 1290–1293.

(10) Regupathi, I.; Srikanth, K. C.; Sindhu, N. Liquid–Liquid Equilibrium of Poly(ethylene glycol) 2000 + Diammonium Hydrogen Citrate + Water System at Different Temperatures. *J. Chem. Eng. Data* **2011**, *56*, 3643–3650.

(11) Ma, B.; Hu, M.; Li, S.; Jiang, Y.; Liu, Z. Liquid-Liquid Phase Equilibrium in the Ternary System Poly(ethylene glycol) + Cs_2CO_3 + H_2O . *J. Chem. Eng. Data* **2005**, *50*, 792–795.

(12) Graber, A. T.; Taboada, E. M. Liquid-Liquid Equilibrium of the Poly(ethylene glycol) + Sodium Nitrate + Water System at 298.15 K. *J. Chem. Eng. Data* **2000**, *45*, 182–184.

(13) Regupathi, I.; Murugesan, S.; Govindarajan, R.; Amaresh, P. S.; Thanapalan, M. Liquid-Liquid Equilibrium of Poly(ethylene glycol) 6000 + Triammonium Citrate + Water Systems at Different Temperatures. J. Chem. Eng. Data 2009, 54, 1094–1097.

(14) Gonzalez-Tello, P.; Camacho, F.; Blazquez, G. Density and Viscosity of Concentrated Aqueous Solutions of Polyethylene Glycol. *J. Chem. Eng. Data* **1994**, 39, 611–614.

(15) Mei, L.; Lin, D.; Zhu, Z.; Han, Z. Densities and Viscosities of Polyethylene Glycol + Salt + Water Systems at 20 °C. *J. Chem. Eng. Data* **1995**, 40, 1168–1171.

(16) Graber, A. T.; Galleguillos, H. Refractive Index, Density, and Viscosity in the $NaNO_3 + H_2O + Poly(ethylene glycol)$ System at Various Temperatures. J. Chem. Eng. Data **2002**, 47, 174–178.

(17) Zafarani-Moattar, T. M.; Sadeghi, R. Liquid-Liquid Equilibria of Aqueous Two Phase Systems Containing Polyethylene Glycol and Sodium Dihydrogen Phosphate or Disodium Hydrogen Phosphate. Experiment and Correlation. *Fluid Phase Equilib.* **2001**, *181*, 95–112.

(18) Voros, N.; Proust, P.; Fredenslund, A. Liquid-Liquid Equilibria of Aqueous Two Phase Systems containing Salts and Polyethylene Glycol. *Fluid Phase Equilib.* **1993**, *90*, 333–353.