Density and Viscosity of Binary and Ternary Mixtures of Poly(ethylene glycol) and Poly(acrylic acid, sodium salt) at Temperatures of (288.15 to 318.15) K

Luis A. Minim,*,† Renata C. F. Bonomo,‡ Isabella V. Amaral,† Maycon F. T. Reis,† Ana A. A. Oliveira,† and Valéria P. R. Minim†

Department of Food Technology, Universidade Federal de Viçosa, 36570-000 Viçosa, MG, Brazil, and Laboratory of Process Engineering, Universidade Estadual do Sudoeste da Bahia, 45700-000 Itapetinga, BA, Brazil

The knowledge of physical properties and their dependency upon composition and temperature of aqueous two-phase systems is important in understanding the behavior of agitation and phase separation of such systems. In this work the viscosity and density of binary and ternary mixtures of water solution of polyethylene glycol and sodium polyacrylate were experimentally determined at a polymer concentration range of (0.04 to 0.12) mass fraction and temperature range of (288.15 to 318.15) K. Density followed a linear behavior with temperatures and solute mass fraction. Viscosity followed a quadratic behavior with solute mass fraction and temperature for the ternary mixture. Simple polynomial functions were successfully fitted to the experimental data.

Introduction

Liquid−liquid extraction using aqueous two-phase systems (ATPS) is an interesting alternative to separate, fractionate, and purify biological components, since the systems provide a mild environment which preserves the molecule structure and activity. This technique still presents good resolution, high yield, and easy scaling up.[1] ATPS can be formed under specific thermodynamic conditions by the mixture of aqueous solutions of two polymers or one polymer and one salt. The resulting ternary mixtures consist of two immiscible phases, a polymer-enriched top phase and a salt (or polymer)-enriched bottom phase.

Recently, a new system composed of polyethylene glycol (PEG) and sodium polyacrylate (NaPA) has been used for protein partitioning, presenting advantages such low viscosity, well-marked phases, and recyclable components.[2,3] These systems only separate phases in the presence of small quantities of salts [(150 to 200) mmol·L^{-1}]. The sodium polyacrylate (NaPA) is a negatively charged polymer, nonflammable and nontoxic, that has a high solubility in water.

Extraction using ATPS can be carried out on batch or continuous systems, though the last mode is more appropriate for large volumes. The design and control of the equipment for extraction present difficulties because of the lack of information on the behavior of the thermophysical properties with composition and temperature. Density and viscosity are important variables that affect intense mixing and phase separation.

Previous studies have reported physical properties of aqueous solutions containing PEG or organic salts.[4−9] However, physical properties of PEG/NaPA systems are still scarce, since it is a novel system, and few studies have been done about it.

This work aims to measure the density and viscosity of water solutions of PEG and PEG + NaPA at different temperatures, T, and polymer concentrations, w. Polynomial functions will be adjusted to the data for its prediction.

* Corresponding author. Tel.: +55 31 3899 1617. Fax: +55 31 3899 2208. E-mail: lminim@ufv.br.
† Universidade Federal de Viçosa.
‡ Universidade Estadual do Sudoeste da Bahia.

Table 1. Chemical Characteristics of PEG and NaPA

	PEG	NaPA
state	solid	liquid
average molar mass	∼6000	∼8000
linear formula	$H(OCH_2CH_2)_nOH$	$[CH_2CH(CO_2Na)]_n$
concentration		45 % mass fraction in H_2O
density (mg·mL^{-1})		1.3 at 25 °C
refractive index		1.428 at 20 °C

Experimental Section

Materials. The analytical grade PEG (average molar mass of 6000 g·mol^{-1}) was obtained from Isofar (Brazil) and NaPA (average molar mass of 8000 g·mol^{-1}, 45 % mass fraction) was purchased from Aldrich Co. (USA). Chemical characteristics of PEG and NaPA are provided in Table 1. Deionized water (Millipore, Bedford, MA) was used throughout the experiments.

Apparatus and Procedures. Aqueous solutions of PEG and NaPA with concentrations of (0.04, 0.06, 0.08, 0.10, and 0.12) mass fraction were prepared using an analytical balance (AUX220, Shimadzu, USA) with an uncertainty of ± 0.0001 g. The pH of the solutions was adjusted to 7.0 with 0.1 mol·L^{-1} NaOH (VETEC, Brazil) using a pH meter (Gehaka, PG-100, Brazil) with an uncertainty of ± 0.01. The solutions were prepared adding appropriate amounts of water and stock solutions of PEG (40 % mass fraction) or NaPA (45 % mass fraction) in glass tubes to reach the desired concentrations. Ternary system compositions were set combining all of the binary system concentrations. Since these ATPS only separate phases in the presence of salts, only monophasic systems were obtained. The temperature of the systems were controlled at (288.15, 293.15, 298.15, 303.15, 308.15, 313.15, and 318.15) K. To keep the temperature constant, a constant temperature water bath (Phoenix II, Thermo Electron Corp., Germany) was used, controlled within ± 0.05 K. Data of the physical properties for the binary systems were obtained in triplicate using a complete factorial design with seven levels of T and five levels of w, adding up 105 experiments. For the ternary systems the data were obtained in duplicate, and a complete factorial design

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Table 2. Density ρ and Dynamic Viscosity η for the Binary Mixtures Containing PEG (w_1) + Water at Different Temperatures

	T/K						
w_1	288.15	293.15	298.15	303.15	308.15	313.15	318.15
				ρ/(kg·m^{-3})			
0.04	1007.3 ± 0.1	1004.4 ± 0.9	1004.1 ± 0.4	1000.6 ± 0.1	999.6 ± 0.2	998.2 ± 0.8	996.6 ± 1.0
0.06	1011.1 ± 0.5	1008.4 ± 0.2	1006.8 ± 0.2	1005.0 ± 0.2	1004.3 ± 1.5	1001.2 ± 0.3	999.8 ± 0.3
0.08	1014.4 ± 0.2	1011.6 ± 0.9	1010.5 ± 0.4	1008.4 ± 0.2	1007.0 ± 0.5	1004.5 ± 0.2	1002.6 ± 0.1
0.10	1018.0 ± 0.2	1015.4 ± 0.6	1013.1 ± 0.3	1010.9 ± 0.1	1010.4 ± 0.1	1007.6 ± 0.2	1005.3 ± 0.6
0.12	1021.5 ± 0.5	1018.8 ± 1.3	1017.5 ± 0.2	1014.3 ± 0.8	1012.8 ± 0.3	1010.1 ± 0.2	1007.2 ± 2.1
				η/(mPa·s)			
0.04	2.28 ± 0.01	2.20 ± 0.02	1.99 ± 0.02	1.87 ± 0.05	1.50 ± 0.02	1.37 ± 0.02	1.25 ± 0.02
0.06	3.00 ± 0.03	2.67 ± 0.02	2.29 ± 0.03	2.29 ± 0.04	1.84 ± 0.02	1.71 ± 0.05	1.58 ± 0.02
0.08	3.82 ± 0.02	3.42 ± 0.02	2.89 ± 0.03	2.92 ± 0.05	2.35 ± 0.02	2.15 ± 0.03	1.97 ± 0.05
0.10	4.70 ± 0.02	4.35 ± 0.02	3.64 ± 0.02	3.62 ± 0.04	2.92 ± 0.02	2.65 ± 0.02	2.43 ± 0.02
0.12	6.02 ± 0.02	5.25 ± 0.02	4.57 ± 0.03	4.41 ± 0.06	3.56 ± 0.03	3.15 ± 0.02	3.04 ± 0.03

Table 3. Density ρ and Dynamic Viscosity η for the Binary Mixtures Containing NaPA (w_2) + Water at Different Temperatures

	T/K						
w_2	288.15	293.15	298.15	303.15	308.15	313.15	318.15
				ρ/(kg·m^{-3})			
0.04	1037.1 ± 0.6	1035.1 ± 0.4	1033.5 ± 1.3	1030.6 ± 0.2	1029.1 ± 0.1	1027.0 ± 0.2	1026.3 ± 2.2
0.06	1053.55 ± 3.1	1052.69 ± 0.1	1050.40 ± 0.1	1042.69 ± 2.5	1045.79 ± 0.7	1043.82 ± 0.7	1042.02 ± 1.2
0.08	1073.64 ± 0.7	1068.97 ± 0.6	1067.65 ± 0.2	1064.33 ± 0.2	1063.19 ± 0.2	1060.92 ± 0.8	1058.25 ± 1.0
0.10	1090.45 ± 0.5	1086.96 ± 0.9	1084.60 ± 1.7	1081.73 ± 0.9	1079.82 ± 0.9	1077.18 ± 0.1	1077.26 ± 0.9
0.12	1109.17 ± 0.8	1104.37 ± 0.3	1100.82 ± 1.2	1098.81 ± 1.0	1095.94 ± 0.3	1094.57 ± 0.2	1092.69 ± 0.3
				η/(mPa·s)			
0.04	2.66 ± 0.03	2.38 ± 0.04	2.11 ± 0.03	1.88 ± 0.03	1.78 ± 0.02	1.63 ± 0.02	1.51 ± 0.02
0.06	3.24 ± 0.02	2.87 ± 0.03	2.58 ± 0.03	2.33 ± 0.03	2.13 ± 0.02	1.95 ± 0.02	1.77 ± 0.02
0.08	4.22 ± 0.03	3.75 ± 0.02	3.27 ± 0.03	2.97 ± 0.03	2.75 ± 0.03	2.47 ± 0.03	2.28 ± 0.02
0.10	5.69 ± 0.03	5.10 ± 0.04	4.37 ± 0.03	3.95 ± 0.03	3.53 ± 0.03	3.16 ± 0.03	2.91 ± 0.03
0.12	7.05 ± 0.03	6.35 ± 0.04	5.26 ± 0.03	4.82 ± 0.03	4.30 ± 0.02	3.81 ± 0.03	3.53 ± 0.03

was also used with seven levels of T and five levels of w for PEG and for NaPA, adding up 350 experiments. All statistical analyses were performed using the GLM (general linear model) procedure, while fitted functions were obtained by using the REG (REGression) procedure from the SAS statistical package.[10] The suitability of the fitted functions was evaluated by the level of significance (p), the coefficient of determination (R^2), and residual analysis.

Density. Gravimetric determination of the solution density, ρ, at different temperatures was conducted using an analytical balance with an uncertainty of ± 0.0001 g and a standard volumetric pycnometer with 10 cm^3. The pycnometer was previously calibrated with deionized water, at each temperature.[11]

Viscosity. Viscosity, η, measurements were carried out using a modular advanced rheometer system (Haake Mars, Thermo Electron Corp., Germany), with stainless steel concentric cylinders (Z 20), during 3 min at a constant shear rate of 100 s^{-1} with an uncertainty of ± 0.01 mPa·s.

Results and Discussion

A series of experiments were made to determine viscosity, η, and density, ρ, for binary mixtures (PEG + water and NaPA + water) and ternary mixture (PEG + NaPA + water), at different temperatures.

Tables 2 and 3 include the experimental values and respective standard deviations between triplicate measurements for the binary mixtures containing PEG and NaPA, respectively. For all systems, the standard deviations of density and viscosity were smaller than 3.1 kg·m^{-3} and 0.08 mPa·s, respectively. The densities and viscosities of the solutions increased as the mass fraction of PEG and NaPA increased, while a decrease of these

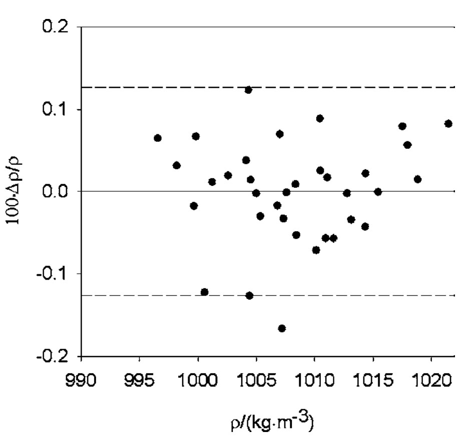

Figure 1. Fractional deviations $\Delta\rho = (\rho_{obs} - \rho_{pred})$ of the observed densities ρ_{obs} of PEG solutions as a function of the predicted density ρ_{pred}. The dashed lines are two standard deviations of the fit.

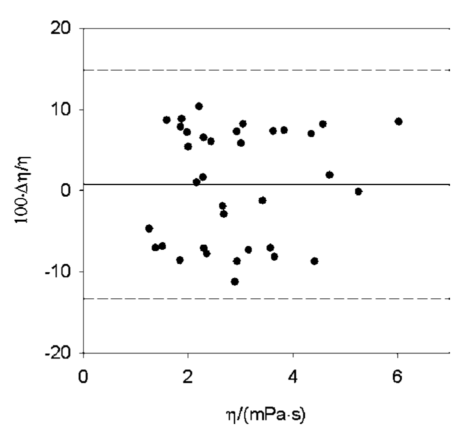

Figure 2. Fractional deviations $\Delta\eta = (\eta_{obs} - \eta_{pred})$ of the observed viscosities η_{obs} of PEG solutions as a function of the predicted viscosities η_{pred}. The dashed lines are two standard deviations of the fit.

Table 4. Density ρ and Dynamic Viscosity η for the Ternary Mixtures Containing PEG (w_1) + NaPA (w_2) + Water at Different Temperatures

		T/K						
w_1	w_2	288.15	293.15	298.15	303.15	308.15	313.15	318.15
					ρ/(kg·m^{-3})			
0.04	0.04	1021.8 ± 0.3	1019.0 ± 0.2	1017.0 ± 0.1	1015.0 ± 0.4	1013.6 ± 0.8	1010.4 ± 2.0	1008.7 ± 0.8
0.04	0.06	1031.1 ± 0.8	1027.8 ± 0.2	1026.7 ± 0.1	1024.1 ± 0.3	1022.3 ± 0.5	1019.8 ± 0.1	1017.0 ± 0.6
0.04	0.08	1039.4 ± 0.3	1036.5 ± 0.1	1034.4 ± 0.2	1031.6 ± 0.1	1030.4 ± 0.3	1027.8 ± 0.1	1024.7 ± 0.1
0.04	0.10	1046.8 ± 0.5	1044.2 ± 0.2	1042.3 ± 0.5	1040.0 ± 0.2	1038.6 ± 0.2	1036.2 ± 0.1	1032.6 ± 0.5
0.04	0.12	1056.1 ± 0.3	1052.6 ± 0.1	1050.9 ± 0.3	1048.3 ± 0.2	1046.5 ± 0.4	1044.6 ± 0.4	1040.3 ± 0.3
0.06	0.04	1023.6 ± 0.1	1020.5 ± 0.5	1018.6 ± 0.4	1017.0 ± 0.2	1017.3 ± 0.5	1014.1 ± 0.5	1012.6 ± 0.1
0.06	0.06	1032.3 ± 0.2	1029.1 ± 0.5	1026.5 ± 0.6	1024.9 ± 0.2	1023.2 ± 0.1	1021.4 ± 0.4	1017.8 ± 0.6
0.06	0.08	1040.2 ± 0.1	1037.8 ± 0.1	1035.5 ± 0.1	1033.4 ± 0.1	1031.7 ± 0.1	1029.6 ± 0.2	1026.2 ± 0.9
0.06	0.10	1049.6 ± 0.4	1046.6 ± 0.3	1044.8 ± 0.2	1042.3 ± 0.2	1040.0 ± 0.4	1037.8 ± 0.1	1033.0 ± 0.4
0.06	0.12	1056.5 ± 0.1	1053.7 ± 0.2	1052.2 ± 0.2	1049.5 ± 0.7	1048.2 ± 0.5	1045.1 ± 0.1	1040.5 ± 0.1
0.08	0.04	1024.5 ± 0.7	1022.6 ± 0.2	1020.8 ± 0.1	1019.0 ± 0.1	1018.7 ± 0.3	1016.3 ± 0.1	1013.1 ± 0.8
0.08	0.06	1034.7 ± 0.1	1031.0 ± 0.3	1029.8 ± 0.8	1027.0 ± 0.1	1025.3 ± 0.2	1023.5 ± 0.6	1019.3 ± 0.1
0.08	0.08	1042.8 ± 0.2	1039.2 ± 0.5	1037.1 ± 0.2	1034.6 ± 0.3	1032.6 ± 0.2	1030.7 ± 0.2	1026.3 ± 0.3
0.08	0.10	1050.3 ± 0.1	1048.1 ± 0.3	1045.1 ± 0.4	1043.0 ± 0.6	1041.2 ± 0.2	1038.2 ± 0.1	1034.0 ± 0.3
0.08	0.12	1059.2 ± 0.1	1056.1 ± 0.3	1053.3 ± 0.1	1051.2 ± 0.3	1049.4 ± 0.6	1047.1 ± 0.1	1041.4 ± 0.6
0.10	0.04	1027.5 ± 0.1	1024.8 ± 0.1	1022.7 ± 0.2	1020.3 ± 0.1	1021.0 ± 0.3	1018.0 ± 0.1	1016.3 ± 0.3
0.10	0.06	1035.3 ± 0.3	1033.3 ± 0.7	1030.6 ± 0.8	1028.1 ± 0.5	1026.5 ± 0.2	1024.0 ± 0.1	1021.3 ± 0.1
0.10	0.08	1043.6 ± 0.3	1041.6 ± 0.3	1038.8 ± 0.3	1036.1 ± 0.1	1034.6 ± 0.9	1032.0 ± 0.3	1028.9 ± 0.6
0.10	0.10	1052.6 ± 0.7	1049.9 ± 0.2	1047.7 ± 0.7	1044.1 ± 0.6	1042.9 ± 0.4	1040.6 ± 0.2	1036.7 ± 0.1
0.10	0.12	1061.4 ± 0.7	1058.6 ± 0.4	1055.5 ± 0.1	1053.6 ± 0.1	1051.4 ± 0.1	1048.7 ± 0.1	1044.5 ± 0.2
0.12	0.04	1029.0 ± 0.6	1026.4 ± 0.7	1023.7 ± 0.3	1022.6 ± 0.1	1022.2 ± 0.4	1018.9 ± 0.9	1017.8 ± 0.4
0.12	0.06	1037.8 ± 0.1	1034.5 ± 0.4	1032.1 ± 0.1	1029.7 ± 0.3	1028.2 ± 0.3	1025.6 ± 0.2	1027.3 ± 1.0
0.12	0.08	1045.9 ± 0.2	1043.9 ± 0.1	1040.9 ± 0.5	1038.2 ± 0.2	1036.0 ± 0.1	1034.2 ± 0.5	1029.8 ± 0.1
0.12	0.10	1054.2 ± 0.1	1051.2 ± 0.5	1048.8 ± 0.1	1046.3 ± 0.1	1044.7 ± 0.3	1042.7 ± 0.1	1037.9 ± 0.4
0.12	0.12	1062.7 ± 0.1	1059.8 ± 0.5	1057.3 ± 0.3	1054.8 ± 0.1	1052.9 ± 0.1	1050.7 ± 0.1	1045.1 ± 0.5
					η/(mPa·s)			
0.04	0.04	2.50 ± 0.02	2.40 ± 0.02	2.06 ± 0.03	2.02 ± 0.03	1.67 ± 0.03	1.47 ± 0.02	1.34 ± 0.02
0.04	0.06	2.86 ± 0.02	2.70 ± 0.02	2.27 ± 0.03	2.07 ± 0.03	1.84 ± 0.03	1.69 ± 0.01	1.52 ± 0.02
0.04	0.08	3.17 ± 0.02	2.99 ± 0.019	2.52 ± 0.03	2.32 ± 0.03	2.04 ± 0.03	1.88 ± 0.02	1.69 ± 0.02
0.04	0.10	3.59 ± 0.02	3.32 ± 0.02	2.83 ± 0.03	2.58 ± 0.03	2.29 ± 0.03	2.10 ± 0.02	1.89 ± 0.02
0.04	0.12	4.03 ± 0.02	3.73 ± 0.02	3.18 ± 0.03	2.89 ± 0.02	2.57 ± 0.03	2.37 ± 0.02	2.11 ± 0.02
0.06	0.04	2.96 ± 0.02	2.74 ± 0.02	2.35 ± 0.03	2.08 ± 0.03	1.87 ± 0.03	1.74 ± 0.02	1.56 ± 0.02
0.06	0.06	3.20 ± 0.02	2.85 ± 0.02	2.53 ± 0.03	2.28 ± 0.03	2.03 ± 0.03	1.87 ± 0.02	1.69 ± 0.02
0.06	0.08	3.61 ± 0.02	3.28 ± 0.02	2.84 ± 0.03	3.61 ± 0.02	2.29 ± 0.03	2.10 ± 0.02	1.90 ± 0.02
0.06	0.10	3.44 ± 0.08	3.57 ± 0.02	3.18 ± 0.03	2.87 ± 0.02	2.56 ± 0.03	2.34 ± 0.02	2.11 ± 0.02
0.06	0.12	4.50 ± 0.02	3.98 ± 0.03	3.53 ± 0.04	3.19 ± 0.02	2.83 ± 0.03	2.59 ± 0.02	3.24 ± 0.04
0.08	0.04	3.39 ± 0.02	3.01 ± 0.02	2.68 ± 0.03	2.39 ± 0.02	2.11 ± 0.02	1.93 ± 0.02	1.75 ± 0.02
0.08	0.06	3.68 ± 0.02	3.25 ± 0.02	2.87 ± 0.03	2.61 ± 0.02	2.35 ± 0.02	2.12 ± 0.02	1.89 ± 0.02
0.08	0.08	4.05 ± 0.02	3.56 ± 0.02	3.18 ± 0.03	2.82 ± 0.04	2.55 ± 0.02	2.32 ± 0.02	2.09 ± 0.02
0.08	0.10	4.52 ± 0.03	4.00 ± 0.02	3.54 ± 0.03	3.25 ± 0.05	2.83 ± 0.03	2.83 ± 0.03	2.40 ± 0.03
0.08	0.12	5.07 ± 0.03	4.46 ± 0.03	3.91 ± 0.02	3.58 ± 0.04	3.17 ± 0.03	2.88 ± 0.02	2.64 ± 0.02
0.10	0.04	3.84 ± 0.02	3.41 ± 0.02	3.02 ± 0.01	2.67 ± 0.04	2.21 ± 0.04	2.15 ± 0.02	1.97 ± 0.03
0.10	0.06	4.13 ± 0.02	3.63 ± 0.02	3.20 ± 0.02	2.87 ± 0.04	2.54 ± 0.03	2.32 ± 0.02	2.11 ± 0.02
0.10	0.08	4.55 ± 0.02	4.01 ± 0.02	3.55 ± 0.02	3.18 ± 0.04	2.83 ± 0.03	2.58 ± 0.02	2.37 ± 0.03
0.10	0.10	5.06 ± 0.02	4.49 ± 0.02	3.96 ± 0.02	3.54 ± 0.04	3.18 ± 0.03	2.86 ± 0.02	2.62 ± 0.02
0.10	0.12	5.76 ± 0.02	5.08 ± 0.02	4.47 ± 0.02	4.22 ± 0.03	3.57 ± 0.03	3.27 ± 0.02	2.96 ± 0.02
0.12	0.04	4.35 ± 0.02	3.74 ± 0.04	3.38 ± 0.02	2.93 ± 0.03	2.89 ± 0.03	2.40 ± 0.02	2.18 ± 0.02
0.12	0.06	4.65 ± 0.03	4.08 ± 0.02	3.65 ± 0.02	3.20 ± 0.03	2.87 ± 0.03	2.60 ± 0.02	2.39 ± 0.03
0.12	0.08	5.46 ± 0.06	4.53 ± 0.02	4.01 ± 0.02	3.53 ± 0.03	3.22 ± 0.03	2.89 ± 0.02	2.61 ± 0.02
0.12	0.10	5.75 ± 0.03	5.00 ± 0.02	4.45 ± 0.02	3.96 ± 0.03	3.54 ± 0.03	3.22 ± 0.02	2.91 ± 0.03
0.12	0.12	6.58 ± 0.03	5.72 ± 0.02	5.07 ± 0.02	4.58 ± 0.03	4.10 ± 0.03	3.78 ± 0.03	3.58 ± 0.02

properties was observed as the temperature increased. The values of density and viscosity of the binary mixtures of PEG were close to those found by Mohsen-Nia et al.[12] for aqueous mixtures of PEG molar mass 1000 and 10000 in the temperature range of (298.15 to 328.15) K and PEG concentration range of 0.05 to 0.15. These authors obtained density values from (0.9978 to 1.0250) g·cm^{-3} and (0.9984 to 1.0278) g·cm^{-3} for PEG 1000 and PEG 10000, respectively. For viscosity the values found ranged from (0.509 to 2.27) mPa·s for PEG 1000. The values of PEG 10000 were approximately 20 % higher than the values of PEG 1000. In this work, the values found for both thermophysical properties were higher for the NaPA solutions than PEG solutions (Tables 2 and 3). This result is also found when comparing the values obtained for the binary mixtures of NaPA with the values found by da Silva et al.[9] for binary mixtures of PEG 4000 and lithium sulfate, for which the density values were in the range of (1032.56 to 1129.34) kg·m^{-3} and (999.55 to 1033.72) kg·m^{-3}, respectively. However, viscosity values of

Table 5. Adjusted Models for Calculating Densities and Dynamic Viscosities of Aqueous PEG and NaPA Solutions

$\rho_1 = 1113.971 - 0.392T + 162.156w_1$	$R^2 = 0.989$	(2)
$\rho_2 = 1144.140 - 0.492T + 873.202w_2$	$R^2 = 0.996$	(3)
$\rho_{12} = 1136.599 - 0.463T + 83.866w_1 + 402.441w_2$	$R^2 = 0.995$	(4)
$\eta_1 = 1.26934 + 250.21928w_1 + 153.73914w_1^{\ 2} - 0.80325w_1T$	$R^2 = 0.983$	(5)
$\eta_2 = 1.63307 + 277.31467w_2 + 244.82248w_2^{\ 2} - 0.91661w_1T$	$R^2 = 0.982$	(6)
$\eta_{12} = 91.87297 - 0.55105T + 8.3438 \cdot 10^{-3}T^2 + 72.85144w_1^{\ 2} + 65.38213w_2^{\ 2} - 3.38908Tw_1w_2 + 1100.29252w_1w_2$	$R^2 = 0.990$	(7)

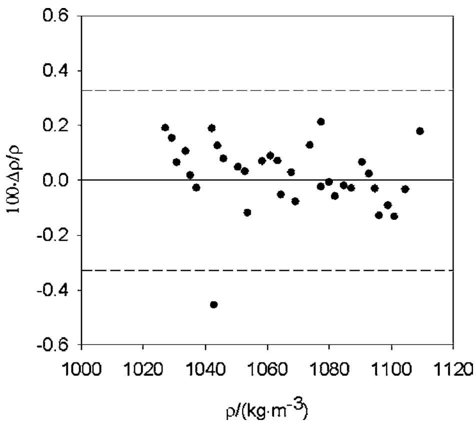

Figure 3. Fractional deviations $\Delta\rho = (\rho_{obs} - \rho_{pred})$ of the observed densities ρ_{obs} of NaPA solutions as a function of the predicted density ρ_{pred}. The dashed lines are two standard deviations of the fit.

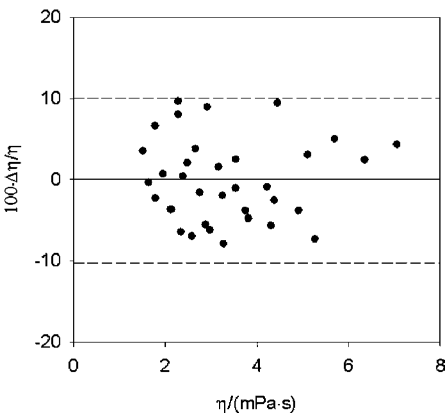

Figure 4. Fractional deviations $\Delta\eta = (\eta_{obs} - \eta_{pred})$ of the observed viscosities η_{obs} of NaPA solutions as a function of the predicted viscosities η_{pred}. The dashed lines are two standard deviations of the fit.

this work are lower than those reported by Ninni et al.[13] for binary mixtures of PEG 8000 and PEG 10000.

Table 4 shows the experimental data of density and viscosity for the ternary mixtures; it was observed that these properties increased as PEG and NaPA concentration increased. It was observed that polymer concentration had a greater influence than temperature over density and viscosity. This fact was also reported in other works.[9,12,14]

Polynomial models for the thermophysical properties ρ and η as a function of T and w were fitted to the experimental data. The general quadratic model, eq 1, was first analyzed, and the nonsignificant parameters were eliminated on the basis of the t-test(Student) and $p > 0.05$. Table 5 shows the predictive models based on the master model (eq 1) for ρ and η of the binary and ternary mixtures.

$$\psi = \beta_0 + \beta_1 T + \beta_2 w_1 + \beta_3 w_2 + \beta_4 T^2 + \beta_5 {w_1}^2 + \beta_6 {w_2}^2 + \beta_7 T w_1 + \beta_8 T w_2 + \beta_9 w_1 w_2 \quad (1)$$

In eq 1, Ψ is the thermophysical property, and the subscripts 1 and 2 refer to the solutes PEG and NaPA, respectively.

Figures 1 to 6 show the relative deviations between observed and predicted values of ρ and η. The agreement between experimental and predicted values for the density and viscosity was very good for all of the systems studied. In all cases, the determination coefficient (R^2) was superior to 0.98. The fractional deviation values were lower than those reported by González-Tello et al.[4] and Ninni et al.,[13] which were approximately 17.5 % for viscosities lower than 10 mPa·s.

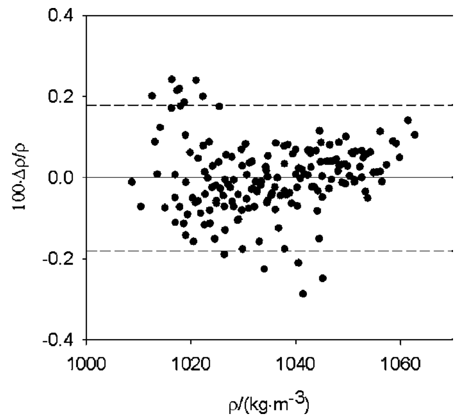

Figure 5. Fractional deviations $\Delta\rho = (\rho_{obs} - \rho_{pred})$ of the observed densities ρ_{obs} of ternary solutions as a function of the predicted density ρ_{pred}. The dashed lines are two standard deviations of the fit.

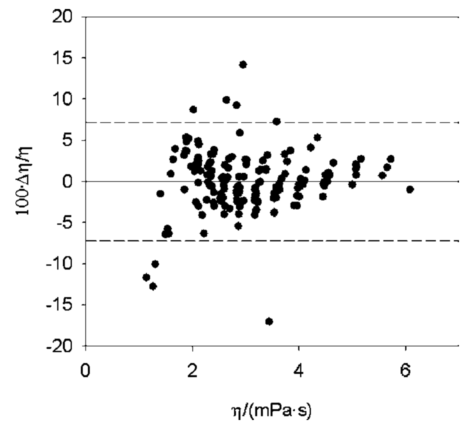

Figure 6. Fractional deviations $\Delta\rho = (\eta_{obs} - \eta_{pred})$ of the observed viscosities η_{obs} of ternary solutions as a function of the predicted viscosities η_{pred}. The dashed lines are two standard deviations of the fit.

Conclusions

In this paper, the effect of T and w on the density and viscosity of binary and ternary mixtures of water solutions containing PEG and/or NaPA were studied. The physical property values increased with the increase of solute mass fraction and decrease in temperature for all systems studied. For viscosity, significant decreases were observed with the addition of small masses of water. The simple polynomial functions gave good agreement with the experimental data, and thus the estimation of ρ and η using the models developed in this work is recommended, considering the range of T and w investigated.

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