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Density, viscosity, surface tension, and molar volume of propylene glycol + water mixtures from 293 to 323 K and correlations by the Jouyban– Acree model

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KEYWORDS

Density; Viscosity; Surface tension; Molar volume; Propylene glycol + water; Binary mixture; Jouyban–Acree model **Abstract** Density, viscosity, surface tension and molar volume of propylene glycol + water mixtures at 293, 298, 303, 308, 313, 318, and 323 K are reported, compared with the available literature data and the Jouyban–Acree model was used for mathematical correlation of the data. The mean relative deviation (MRD) was used as an error criterion and the MRD values for data correlation of density, viscosity, surface tension and molar volume at different investigated temperatures are $0.1 \pm 0.1\%$, $7.6 \pm 6.4\%$, $3.4 \pm 3.7\%$, and $0.4 \pm 0.4\%$, respectively. The corresponding MRDs for the predicted properties after training the model using the experimental data at 298 K are $0.1 \pm 0.2\%$, $12.8 \pm 9.3\%$, $4.7 \pm 4.1\%$ and $0.6 \pm 0.5\%$, respectively for density, viscosity, surface tension, and molar volume data.

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1. Introduction

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Density, viscosity, surface tension and molar volume of liquids are important physicochemical properties (PCPs) which affect mass and heat transfer processes in solutions. So, availability of the related data should be helpful in designing and engineering of such processes. As an example, Tubtimdee and Shotipurk (2011) reported that in the extraction of phenolic compounds from a plant source, employing propylene glycol + water or ethanol + water mixtures require lower temperatures when compared with the mono-solvent extraction

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systems (Tubtimdee and Shotipurk, 2011). It is obvious that using lower temperatures for extraction of thermal liable compounds like phenolics is more valuable.

It has been observed that PCPs of solvent mixtures show deviation from ideal mixing and finding a suitable blend of solvents for a desired amount of PCP requires some experimental determinations, or accurate models. Despite the experimental determination of PCPs, a number of mathematical models have been presented for calculating PCP data. Among the similar models for correlation of PCPs of liquid mixture, the Jouyban–Acree model is perhaps one of the most accurate models (Jouyban et al., 2004a,b,c; 2005a,b,2006; Hasan et al., 2006; Delgado et al., 2012). The model for correlation of PCPs of the binary solvent mixtures at various temperatures is:

$$\ln PCP_{m,T} = x_1 \cdot \ln PCP_{1,T} + x_2 \cdot \ln PCP_{2,T} + J_0 \left[\frac{x_1 \cdot x_2}{T}\right] + J_1 \left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T}\right] + J_2 \left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)^2}{T}\right]$$
(1)

where $PCP_{m,T}$, $PCP_{1,T}$, and $PCP_{2,T}$ are the physicochemical properties under study of the mixture, solvents 1 and 2, respectively; x_1 and x_2 denote mole fractions of the solvents 1 and 2, respectively; *T* is the absolute temperature; and J_i terms are the coefficients of the model which are computed by regressing $(\ln PCP_{m,T} - x_1 \cdot \ln PCP_{1,T} - x_2 \cdot \ln PCP_{2,T})$ against $\frac{x_1 \cdot x_2}{T}$, $\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T}$ and $\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)^2}{T}$ using a no-intercept regression analysis.

The aims of this work are to report density, viscosity, surface tension and molar volume of propylene glycol + water mixtures at different temperatures and to give a predictive model for these properties.

2. Experimental

2.1. Materials

Propylene glycol was purchased from Merck, Germany. The water used in this study was of Ultra pure reagent grade type 1 water, prepared using a Millipore water purifier system (Milli-Q Synthesis, France) with the conductivity of 5.49×10^{-6} S m⁻¹ and TOC < 10 ppb.

2.2. Apparatus and procedures

Suitable proportions of the solvents were mixed with each other volumetrically with an uncertainty of 0.1 mL to produce different blends of propylene glycol and water by intervals of 0.10 in volume fraction. The prepared solvent mixtures were placed on a shaking water bath (Memmert, Germany) with set temperature accuracy of 0.1 °C at 293, 298, 303, 308, 313, 318, and 323 K. After at least 2 h and on assurance of equilibrium, the samples were analyzed. Mettler Toledo Densito Portable 30PX Density Meter (USA) was used for density measurements of the equilibrated solvent mixtures. The viscosity of the solvent mixtures was measured using an Ostwald U-tube glass viscometer (Union Scientific Appliances, India)

suspended in a constant temperature water bath. Surface tension of the prepared blends of solvents was determined by drop number method. The accuracy of reported data was 0.0001 (g cm⁻³), 0.0001 (mPag s), and 0.01 (mNg m⁻¹) for density, viscosity, and surface tension, respectively. The lowest and highest standard deviations of the measurements for density were 0.0001 and 0.0008 with an overall relative standard deviation of 0.08%. These values were 0.0032, 0.0719, and 1.32% for viscosity, and 0.1534, 0.9549, and 1.12% for surface tension measurements, respectively. All measurements were done at least in triplicates.

2.3. Computational methods

The Jouyban–Acree model was used as a mathematical model for correlation of the studied PCPs in solvent mixtures. For this purpose, for each property under investigation, it has been trained with experimental data using a no-intercept regression analysis. The molar volumes (V) for binary mixtures are calculated by:

$$V_{m,T} = \frac{x_1 M_1 + x_2 M_2}{\rho_{m,T}}$$
(2)

where M_1 and M_2 are the molar masses of solvents 1 and 2, respectively. The mean relative deviation (MRD) was used as error criterion as:

$$MRD = \frac{100}{N} \sum \frac{|PCP_{\text{Exp}} - PCP_{\text{Cal}}|}{PCP_{\text{Exp}}}$$
(3)

where *N* is the number of data points in each set, PCP_{Exp} and PCP_{Cal} are the experimental and calculated PCPs under investigation.

3. Results and discussion

Experimental and calculated values of density, viscosity, surface tension and molar volume data are listed in Table 1. The behavior of density in mixtures of propylene glycol + water shows a peak at a mole fraction of ~ 0.4 of propylene glycol. The viscosity of the binary mixtures was increased non-linearly with the mole fraction of propylene glycol. The change trend of surface tension in propylene glycol + water mixtures is decreased with the increased mole fraction of propylene glycol. Increasing pattern is observed with the molar volume data of the mixtures with respect to the mole fraction of propylene glycol.

Available experimental data of density, viscosity, and surface tension of water and propylene glycol at different temperatures from the literature (MacBeth and Thompson, 1951; Nakanishi et al., 1967; Hoke and Patton, 1992; Geyer et al., 2000; George and Sastry, 2003; Sun and Teja, 2004; Jiménez and Martínez, 2005) were compared with listed data in Table 1. MacBeth and Thompson (1951) reported the density of propylene glycol + water mixtures at 308 K. Nakanishi et al. (1967) measured densities of aqueous mixtures of glycols at 298.15 K and calculated their excess molar volumes. Surface tensions at 298, 303, 308, 313, 318 and 323 K have been reported by Hoke and Patton (1992). Geyer et al. (2000) published densities of binary mixtures of four diols including propylene glycol at 278.15, 288.15, 298.15, 308.15 and 318.15 K. George and Sastry (2003) measured the density, vis-

<i>x</i> ₁	$\rho \times 10^{-3} \ (\text{kg m}^{-3})$		$\eta \times 10^{-3}$ (Pa s)		$\sigma \times 10^{-3} ({\rm N \ m^{-1}})$		$V (\mathrm{cm}^3 \mathrm{mol}^{-1})$	
	Expt	Cal	Expt	Cal	Expt	Cal	Expt	Cal
T = 293 (K)								
0.000	0.9978	0.9978	1.003	1.003	72.88	72.88	18.04	18.04
0.027	1.0051	1.0062	1.434	1.277	63.11	68.17	19.44	19.43
0.058	1.0136	1.0147	1.975	1.667	57.61	63.50	21.07	21.10
0.095	1.0227	1.0229	2.780	2.242	51.61	58.93	23.01	23.12
0.141	1.0312	1.0304	3.826	3.115	47.60	54.57	25.38	25.61
0.197	1.0365	1.0368	5.325	4.484	43.72	50.56	28.41	28.74
0.269	1.0408	1.0410	9.235	6.694	42.26	47.15	32.31	32.74
0.364	1.0427	1.0424	12.021	10.344	41.40	44.66	37.55	38.03
0.495	1.0425	1.0409	18.332	16.462	40.50	43.57	44.87	45.36
0.688	1.0401	1.0390	29.494	27.263	39.55	43.80	55.75	56.36
1.000	1.0353	1.0353	57.571	57.571	38.57	38.57	73.50	73.50
T = 298 (K)								
0.000	0.9958	0.9958	0.976	0.976	69.06	69.06	18.08	18.08
0.027	1.0031	1.0040	1.170	1.228	59.80	64.87	19.48	19.46
0.058	1.0113	1.0123	1.569	1.582	55.66	60.71	21.12	21.11
0.095	1.0202	1.0204	2.346	2.095	51.22	56.65	23.06	23.12
0.141	1.0282	1.0278	3.331	2.857	49.04	52.79	25.45	25.60
0.197	1.0333	1.0340	4.529	4.021	46.93	49.29	28.49	28.70
0.269	1.0376	1.0381	6.757	5.842	45.68	46.39	32.40	32.69
0.364	1.0393	1.0395	9.864	8.726	44.72	44.45	37.66	37.96
0.495	1.0389	1.0379	14.508	13.277	43.72	44.01	45.01	45.29
0.688	1.0367	1.0361	22.908	20.648	42.68	45.22	55.92	56.33
1.000	1.0323	1.0323	39.436	39.436	41.31	41.31	73.71	73.71
T = 303 (K)								
0.000	0.9938	0.9938	0.937	0.937	65.59	65.59	18.11	18.11
0.026	1.0007	1.0018	1.026	1.165	60.27	61.83	19.52	19.48
0.058	1.0086	1.0099	1.396	1.481	56.74	58.10	21.17	21.12
0.095	1.0166	1.0177	1.976	1.931	52.26	54.48	23.13	23.12
0.140	1.0242	1.0248	2.727	2.585	49.66	51.05	25.53	25.58
0.197	1.0294	1.0308	3.513	3.560	48.34	47.97	28.58	28.66
0.268	1.0329	1.0348	5.086	5.037	47.37	45.49	32.52	32.63
0.363	1.0347	1.0359	7.424	7.276	47.83	44.00	37.80	37.90
0.495	1.0342	1.0341	10.524	10.600	46.72	44.11	45.18	45.24
0.688	1.0335	1.0319	16.043	15.505	44.66	46.12	56.07	56.36
1.000	1.0276	1.0276	26.852	26.852	43.46	43.46	74.05	74.05
T = 308 (K)								
0.000	0.9918	0.9918	0.862	0.862	64.46	64.46	18.15	18.15
0.026	0.9983	0.9996	0.956	1.063	59.89	60.87	19.57	19.51
0.057	1.0063	1.0074	1.154	1.339	56.46	57.31	21.21	21.14
0.095	1.0136	1.0150	1.560	1.728	52.49	53.83	23.19	23.12
0.140	1.0207	1.0220	2.201	2.287	50.28	50.55	25.60	25.56
0.196	1.0253	1.0277	3.160	3.108	48.16	47.61	28.67	28.63
0.268	1.0288	1.0315	4.091	4.328	47.22	45.25	32.63	32.58
0.363	1.0317	1.0324	5.698	6.132	46.65	43.88	37.88	37.84
0.494	1.0302	1.0304	7.847	8.718	45.54	44.11	45.32	45.19
0.687	1.0285	1.0279	12.054	12.336	44.83	46.28	56.31	56.38
1.000	1.0231	1.0231	20.267	20.267	43.96	43.96	/4.3/	/4.3/
T = 313 (K)								
0.000	0.9884	0.9884	0.761	0.761	63.93	63.93	18.21	18.21
0.026	0.9951	0.9961	0.848	0.932	58.41	60.40	19.63	19.56
0.058	1.0029	1.0038	1.069	1.166	54.09	56.90	21.28	21.19
0.095	1.0103	1.0112	1.421	1.492	51.70	53.48	23.26	23.16
0.140	1.0167	1.0181	1.863	1.956	50.73	50.23	25.70	25.59
0.196	1.0233	1.0237	2.563	2.627	49.81	47.32	28.73	28.65
0.268	1.0244	1.0275	3.526	3.608	47.18	44.97	32.77	32.59
0.363	1.0294	1.0285	4.640	5.028	45.99	43.58	37.96	37.83

Table 1	Mole fraction of propylene glycol (x_1), experimental and calculated (by Eqs. ()()()()(4)–(7)) data of density (ρ), viscosity (η),
surface te	ension (σ), and molar volume (V) of propylene glycol + water binary mixtures at different temperatures.

Table 1 continued								
<i>x</i> ₁	$ ho imes 10^{-3} \ (\mathrm{kg} \ \mathrm{m}^{-3})$		$\eta \times 10^{-3}$ (Pa s)		$\sigma \times 10^{-3} ({\rm N \ m^{-1}})$		$V (\mathrm{cm}^3 \mathrm{mol}^{-1})$	
	Expt	Cal	Expt	Cal	Expt	Cal	Expt	Cal
0.494	1.0257	1.0266	6.449	7.001	45.15	43.73	45.53	45.18
0.687	1.0233	1.0242	9.269	9.632	44.07	45.73	56.60	56.42
1.000	1.0197	1.0197	15.136	15.136	43.30	43.30	74.62	74.62
T = 318 (K)							
0.000	0.9840	0.9840	0.721	0.721	62.67	62.67	18.29	18.29
0.026	0.9912	0.9914	0.784	0.878	56.54	59.29	19.70	19.64
0.057	0.9989	0.9988	0.964	1.092	53.44	55.93	21.35	21.25
0.094	1.0065	1.0060	1.230	1.387	50.30	52.64	23.33	23.21
0.139	1.0125	1.0126	1.705	1.805	49.37	49.52	25.78	25.63
0.196	1.0151	1.0179	2.116	2.404	48.71	46.71	28.93	28.68
0.267	1.0200	1.0214	2.869	3.271	47.81	44.45	32.87	32.62
0.362	1.0216	1.0221	4.185	4.508	45.41	43.12	38.20	37.87
0.493	1.0177	1.0198	5.161	6.197	44.59	43.29	45.83	45.26
0.686	1.0142	1.0168	7.955	8.387	44.12	45.32	57.06	56.63
1.000	1.0114	1.0114	12.844	12.844	43.07	43.07	75.23	75.23
T = 323 (K)							
0.000	0.9715	0.9715	0.675	0.675	62.02	62.02	18.53	18.53
0.027	0.9826	0.9790	0.701	0.820	58.20	58.82	19.89	19.89
0.058	0.9917	0.9865	0.907	1.015	55.32	55.66	21.53	21.51
0.095	1.0009	0.9938	1.055	1.284	54.03	52.58	23.50	23.49
0.140	1.0065	1.0006	1.569	1.659	53.47	49.68	25.99	25.93
0.197	1.0107	1.0063	1.961	2.193	53.27	47.12	29.13	29.00
0.269	1.0160	1.0102	2.697	2.953	51.52	45.15	33.09	32.95
0.364	1.0159	1.0117	3.759	4.020	50.00	44.18	38.53	38.22
0.495	1.0119	1.0106	4.802	5.437	48.04	44.86	46.21	45.63
0.688	1.0082	1.0095	7.319	7.207	47.20	47.67	57.50	57.00
1.000	1.0069	1.0069	10.691	10.691	46.47	46.47	75.57	75.57

cosity, speed of sound and dielectric constants of propylene glycol + water mixtures and some other alkanediol + water mixtures at 298, 308, 318, 328 and 338 K. Density, viscosity and thermal conductivity of aqueous mixtures of propylene glycol, dipropylene glycol and tripropylene glycol at different temperatures (290–460 K) have been reported by Sun and Teja (2004). There are good agreements between available data from the literature and newly generated data in this work. Density of propylene glycol + water mixtures at 293.15, 298.15, 303.15, 318.15, and 323.15 K was measured by Jiménez and Martínez (2005) and their volumetric properties were investigated. Our measured values differ from the published literature values by no more than 0.002. The measured data cover a wider temperature range and also could be used to evaluate the reproducibility of the data measured in different laboratories.

The resulted equations for density, viscosity, surface tension and molar volume calculations using the Jouyban–Acree model are:

$$\ln \rho_{m,T} = x_1 \cdot \ln \rho_{1,T} + x_2 \cdot \ln \rho_{2,T} + 27.820 \left[\frac{x_1 \cdot x_2}{T} \right] - 30.537 \left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T} \right] + 30.476 \left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)^2}{T} \right]$$
(4)

$$\ln \eta_{m,T} = x_1 \cdot \ln \eta_{1,T} + x_2 \cdot \ln \eta_{2,T} + 926.206 \left[\frac{x_1 \cdot x_2}{T}\right] - 606.410 \left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T}\right]$$
(5)

$$\ln \sigma_{m,T} = x_1 \cdot \ln \sigma_{1,T} + x_2 \cdot \ln \sigma_{2,T} - 183.307 \left[\frac{x_1 \cdot x_2}{T}\right] + 197.808 \left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)}{T}\right] - 456.916 \left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)^2}{T}\right]$$
(6)

$$\ln V_{m,T} = x_1 \ln V_{1,T} + x_2 \ln V_{2,T} + 264.365 \left[\frac{x_1 x_2}{T}\right] - 101.545 \left[\frac{x_1 x_2 (x_1 - x_2)}{T}\right] + 62.243 \left[\frac{x_1 \cdot x_2 \cdot (x_1 - x_2)^2}{T}\right]$$
(7)

where ρ , η , σ and V are indicators of density, viscosity, surface tension and molar volume; 1, 2, and m subscripts stand for propylene glycol, water and their mixtures, respectively. The coefficient of determination (R^2) values for Eqs. (4)–(7) are 0.992, 0.965, 0.916, and 0.999, respectively.

All of these PCPs were correlated perfectly using the Jouyban–Acree model with overall MRD values of $0.1 \pm 0.1\%$, $7.6 \pm 6.4\%$, $3.4 \pm 3.7\%$, and $0.4 \pm 0.4\%$ for density, viscosity, surface tension, and molar volume data, respectively. The model could be trained using measured data at 298 K and the PCP data at other temperatures and solvent compositions could be predicted by employing PCP data of mono-solvents at each temperature of interest, i.e. PCP_{1,T} and PCP_{2,T}. The resulted MRD values for predicted density, viscosity, surface tension, and molar volume data (N = 66) are $0.1 \pm 0.2\%$,

 $12.8 \pm 9.3\%$, $4.7 \pm 4.1\%$ and $0.6 \pm 0.5\%$, respectively. The Jouyban-Acree model also produced the most accurate results among other similar models for representing the solubility of drugs in mixed solvents (Jouvban-Gharamaleki et al., 1999), solubility of solutes in binary mixtures of supercritical fluids (Jouyban et al., 2005b), acid dissociation constants of analytes in binary mixtures (Jouyban et al., 2005e), electrophoretic mobility of charged species in capillary electrophoresis (Jouyban-Gharamaleki et al., 2000) and retention factors of analytes in high performance liquid chromatography (Jouyban et al., 2005d). All these solutions PCP have been represented using the universal form of the Jouyban-Acree model, i.e., Eq. (1), whereas different algorithms were used in the literature for representing these PCPs. Training of these models (including the Jouyban-Acree model) is their main drawback which restricts their practical application. To overcome this restriction, two solutions could be employed; (a) training the model by a minimum number of experimental data and predicting the unmeasured data points (Jouyban et al., 2004a, 2005a,c) and (b) employing the globally trained version of the model (Jouyban et al., 2011) to predict the PCPs.

4. Conclusion

As a conclusion, the PCP data of propylene glycol + water mixtures are reported at 293 to 323 K and the model constants of the data are computed. Using these constants, it is possible to predict the PCP data in all solvent compositions of propylene glycol + water at various temperatures using the interpolation technique. The trained equations, i.e. Eqs. (4)–(7), are valid for propylene glycol + water mixtures. It is possible to include some descriptors to represent the effects of the solvent properties to provide global versions of the model. Such models have been reported for viscosity (Jouyban et al., 2011) and density (Jouyban et al., 2012) of mixed solvents at various temperatures.

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