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Thermophysical Study of Ethylene Glycol + H_2O and Ethylene Glycol + (DMF/H_2O) at 298.15 K

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In this research, some thermophysical properties of ethylene glycol with water (H_2O) and two solvent mixtures of dimethylformamide/water (DMF + H_2O) were studied. The densities (ρ) and viscosities (η) of ethylene glycol in water and mixed solvent dimethylformamide (DMF + H_2O) were determined at 298.15 K, t and a range of concentrations from 0.1 to 1.0 molar. The ρ and η values were subsequently used to calculate the thermodynamics of mixing including the apparent molar volume (ϕ_v), partial molar volume (ϕ_v^o) at infinite dilution. The solute-solute interaction is presented by S_v results from the equation $\emptyset_v = \phi_v^0 + S_v\sqrt{m}$. The values of viscosity (B) coefficients and Falkenhagen coefficient (A) of the Jone-Dole equation and Gibbs free energy of activation (ΔG^*) for viscous flow of solution were estimated. The positive values of ϕ_v^0 between 35.58 and 49.908 and S_v values from 12.54 to 8.83 indicate strong solute-solvent interactions.

Keywords: Ethylene glycol, Dimethyl formamide, Density, Viscosity and Activation energy of viscous flow

INTRODUCTION

Solvents play a significant role in chemical preparation and chemical engineering applications [1]. These applications require an understanding of the thermodynamics and transport properties of multi-component systems [2]. Most intermediate or chemical reactions are normally carried out in solutions [3]. Therefore, tests of volumetric and viscometric studies of solutions have been recently used to investigate their physical-chemical properties [4,5]. These properties are also useful to know the properties of liquid solutions [6].

Ethylene glycol is found to have a wide range of applications in aqueous solutions including the separation of bio-molecular mixtures, chemical processes, pharmaceutical applications, and anti-freezing chemicals plasticizers, releasing agents for rubber and scouring agents [7,8]. Accurate knowledge of their thermophysical properties is essential in processes involving these mixtures. In physical chemistry, the solvation of a solute in certain solvents as well as different types of interactions solute-solvent and solvent-solvent are important [9]. Through the preparation of a liquid mixture, molecular interactions alter and the difference in the packing of components becomes evident. For example, if a hydrogen bond network is formed, the mixture properties change at last one of the solvents in a particular manner [10]. Such liquid mixture features would be beneficial in constructing various chemical industry transport and processing industry [11].

These molecules can form micelles via self-associating in aqueous solutions [12], which significantly reduces the interaction between water and hydrocarbon while maintaining hydration in the polar groups. The key contribution to free energy in this phase is to eliminate unfavorable hydrocarbon-water contact between head groups and interactions [13,14]. DMF molecules inter-molecular H-bonding is self-associated while ethylene glycol is a very interesting solvent due to the inclusion of the same molecules

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in the oxygen and hydroxyl groups [15]. Accordingly, ethylene glycol molecules with the same or different O and OH groups form intra- and intermolecular hydrogen bonds [16], despite the compound's intriguing properties and practical importance [17].

Therefore, the present work investigated the experimental data of the viscosities (η) and densities (ρ) at different concentrations (0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9 and 1 M) of ethylene glycol in H_2O and [DMF + H_2O] mixture (5%, 10%, 15%, and 20%) (w/w)) at 298.15 K which were used to calculate the apparent molar volumes ϕ_v, limiting molar volumes at infinite dilution ϕ_v^o, Jone-Dole coefficients (A, B), and Gibbs free energy ΔG^*.

EXPERIMENTAL

Chemicals

Ethylene glycol and Dimethyl formamide (DMF), an aprotic polar liquid with high dielectric constant (purity > 99%), were purchased from Fluka company (USA) and were used without any further purification.

Viscosity and Density Measurements

The viscosity values (η) were measured using a suspended level ubbelohodeubbelohde viscometer placed in temperature controlled bath at 298.15 ± 0.01 K under atmospheric pressure. Density values (ρ) for both system solutions were measured using a vibrating tube with a digital Anton-Paar densimeter (Model DMF 60/602). The uncertainties in viscosities and densities measurements were within ± 0.003 cp (centipoise) and ± 1 × 10^{-5} g cm^{-3}, respectively.

RESULTS AND DISCUSSION

The values (ρ) and (η) for the mixtures of [ethylene glycol +H_2O] and [ethylene glycol + (DMF + H_2O)] at 298.15 K are shown in Table 1. The density data were then employed for calculating the apparent molar volumes (ϕ_v), using Masson's equation [18,19].

$$\phi_v = \frac{1}{m}\left[\frac{10^3+mM}{\rho} - \frac{10^3}{\rho_o}\right] \quad (1)$$

Where ρ and ρ_o stand for the solution density and the solvent density, respectively. M represents the molecular mass of the solvent and m is the molality of the solution for which the value is determined using the following relation [20]:

$$m = \frac{1}{\left(\frac{\rho}{c} - \frac{M}{1000}\right)} \quad (2)$$

Here, c represents a molar concentration. The limiting apparent molar volume (ϕ_v) was obtained from the following equation [21]:

$$\emptyset_v = \phi_v^o + S_v\sqrt{m} \quad (3)$$

Where ϕ_v^o is the partial molar volume under infinite dilution and S_v represents experimental slop which stands for a volumetric pairwise interaction coefficient. The ϕ_v^0 indicates the existence of solute-solvent interaction. S_v represents a solute-solute interaction. The plots of ϕ_v against ($\sqrt{m}$) are given in Fig. 1.

Our results show that for the [ethylene glycol + H_2O] and [ethylene glycol + %(H_2O + DMF)] lead to an increase in the values of ϕ_v and ϕ_{Vo} apparent molar volume and partial molar volume for all systems. These results indicate strong solute-solvent interactions [22,23]. The positive and large values of S_v specify the existence of strong solute-solute interaction in

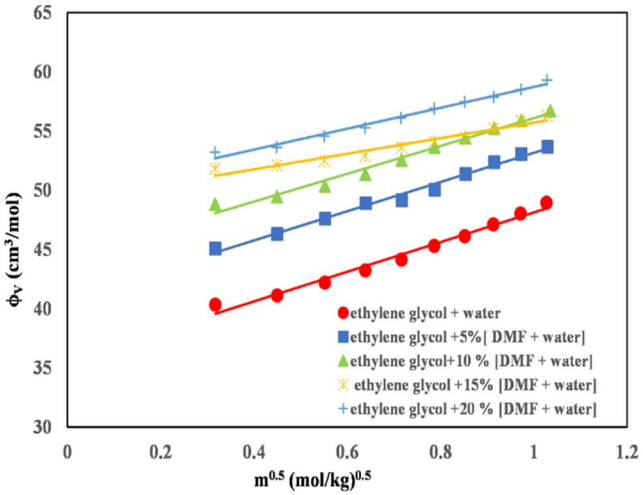

Fig. 1. Masson's plot apparent molar values ϕ_v *vs.* ($\sqrt{m}$) for [ethylene glycol + H_2O] and [ethylene glycol + (H_2O + DMF) mixtures].

the solution. These interactions increase as the system's DMF concentration rises in a system operating at 298.15 K. At a specified temperature composition and form transition state theory, Gibbs free energy of activation is used for viscous flow of solution. ΔG^* (J mol^{-1}) was determined using the following equation [24,25]:

$$\Delta G^* = RT\ln\left(\frac{V_{1,2}\,\eta}{h\,N_A}\right) \quad (4)$$

Accordingly, R, h, and N stand for gas constant, Planck's constant, and Avogadro's constant, respectively. T represents the absolute temperature and the volume of the mole solution $V_{1,2}$ obtained from the following equation [26]:

$$V_{1,2} = \frac{10^3 + mM_2}{\rho\left(\frac{10^3}{M_1} + m\right)} \quad (5)$$

The calculated values of (ΔG^*) are presented in Table 1. This table makes it abundantly evident that the (ΔG^*) magnitudes are large in all systems and tends to increase with increasing concentrations of DMF. These imply that processing viscous flow is difficult because the amount of DMF in the solution increases. This might be related to a strong solute-solvent interacting in a solution [27,28].

The viscosity (η) of the dilute solution of non-electrolytes was calculated as follows [29]:

$$\frac{\eta}{\eta_o} = 1 + BC \quad (6)$$

The values of $\frac{\eta}{\eta_o}$ for all systems were observed and given in Table 1 and plotted against different concentrations of ethylene glycol as in Fig. 2.

Figure 2 indicates that each plot has a linearity greater than 0.9. Based on the Jones-Dole equation, the viscosity data of an aqueous solution at various molarities of [DMF + H_2O] solution were examined [30], Eq. (7):

$$\frac{\frac{\eta}{\eta_0} - 1}{\sqrt{C}} = A + B\sqrt{C} \quad (7)$$

Where the ratio $\frac{\eta}{\eta_0}$ represents the relative viscosity, C represents molar concentration, η and η_o represent the respective viscosities of solution and solvent. From Eq. (6),

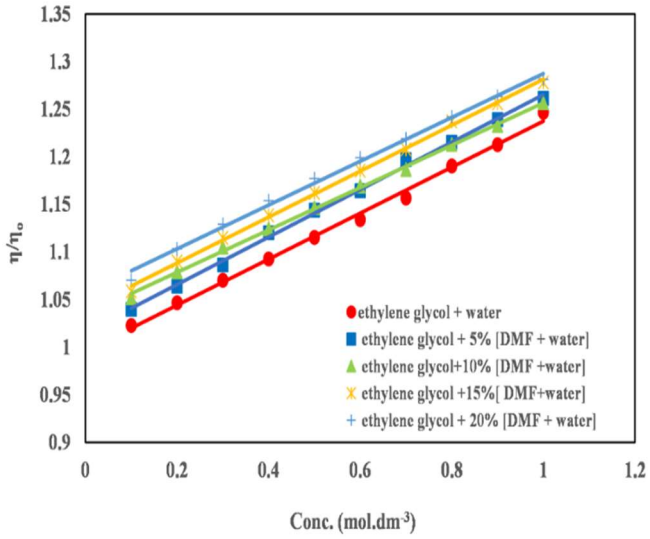

Fig. 2. plot (Conc.) *vs.* (η/η_o) for [ethylene glycol + H_2O] and [ethylene glycol + (DMF + H_2O) mixtures].

the positive values of the Jones-Dole coefficient (B) indicate a strong ethylene glycol in [DMF + H_2O] solutions which reveals the structure-making nature of ethylene glycol in [DMF + H_2O]. A and B coefficients were obtained from the interceptions and slopes of plots $(\eta/\eta_0-1)/\sqrt{C}$ *vs.* $\sqrt{C}$ are depicted in Fig. 3.

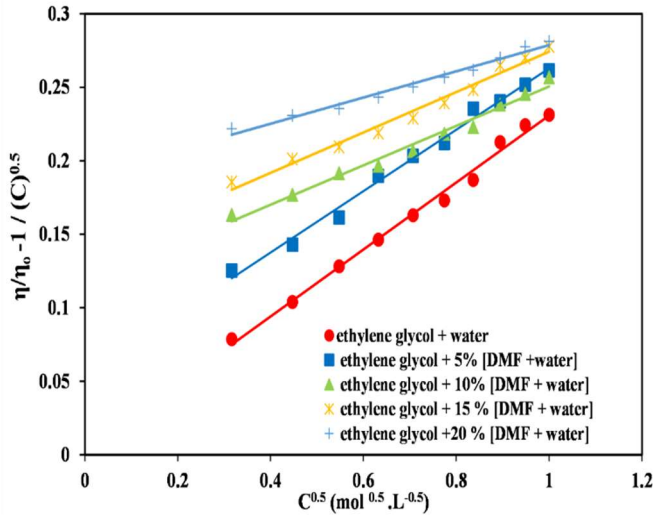

Fig. 3. Jone-Dole plot (η/η_o-1)/√c versus (√c) for [ethylene glycol + H_2O] and [ethylene glycol + (DMF +H_2O) mixtures].

Table 1. Concentration (c), Molality (m), Density, Viscosity, Apparent Molar Volume, Relative Viscosity, and Gibbs Free Energy of Activation for Viscous Flow for [Ethylene Glycol + H_2O] and [Ethylene Glycol + %(DMF+ H_2O) Mixtures] at 298.15 K

Ethylene glycol + H_2O

Conc. (mol dm^{-3})	m (mol kg^{-1})	$\sqrt{m}$ (mol kg^{-1})$^{0.5}$	ρ (g cm^{-3})	ϕ_V (cm^3 mol^{-1})	η (cp)	$\eta_r = \eta/\eta_o$	$\sqrt{C}$ (mol$^{1/2}$ dm$^{-3/2}$)	ΔG^* (J mol^{-1})	$(\eta/\eta_o-1)/\sqrt{c}$
0.0	0.00000	0.00000	0.99704	0.00000	0.89013			60529.78	
0.1	0.10071	0.31735	0.99921	40.33742	0.91225	1.02272	0.31623	60595.69	0.07858
0.2	0.20227	0.44974	1.00121	41.10834	0.93147	1.04644	0.44721	60652.87	0.10384
0.3	0.30478	0.55207	1.00295	42.18701	0.95259	1.07017	0.54772	60714.61	0.12811
0.4	0.40831	0.63900	1.00448	43.22114	0.97248	1.09251	0.63246	60772.55	0.14627
0.5	0.51286	0.71614	1.00585	44.13947	0.99271	1.11524	0.70711	60830.72	0.16297
0.6	0.61878	0.78663	1.00689	45.29235	1.00935	1.13394	0.77459	60879.93	0.17292
0.7	0.72578	0.85193	1.00865	46.11571	1.02937	1.15643	0.83666	60934.87	0.18697
0.8	0.83421	0.91335	1.00865	47.10694	1.05942	1.19019	0.89443	61016.83	0.21264
0.9	0.94401	0.97160	1.00924	48.03039	1.07945	1.21269	0.94868	61072.50	0.22421
1.0	1.05531	1.02728	1.00966	48.93977	1.10957	1.24653	1.0000	61145.10	0.23128

Ethylene glycol + 5% [DMF +H_2O]

Conc. (mol dm^{-3})	m (mol kg^{-1})	$\sqrt{m}$ (mol kg^{-1})$^{0.5}$	ρ (g cm^{-3})	ϕ_V (cm^3 mol^{-1})	η (cp)	$\eta_r = \eta/\eta_o$	$\sqrt{C}$ (mol$^{1/2}$ dm$^{-3/2}$)	ΔG^* (J mol^{-1})	$(\eta/\eta_o-1)/\sqrt{c}$
0.0	0.00000	0.00000	0.99728	0.00000	0.92558			64100.47	
0.1	0.10073	0.31738	0.99898	45.06959	0.96224	1.03961	0.31623	64192.53	0.12526
0.2	0.20243	0.44992	1.00042	46.30772	0.98469	1.06396	0.44721	64246.13	0.14302
0.3	0.30519	0.55244	1.00159	47.58847	1.00742	1.08639	0.54772	64299.80	0.16145
0.4	0.40914	0.63964	1.00248	48.91324	1.03665	1.12003	0.63246	64368.50	0.18978
0.5	0.51409	0.71700	1.00363	49.15692	1.05869	1.14381	0.70711	64417.81	0.20338
0.6	0.62041	0.78766	1.00435	50.03667	1.07758	1.16422	0.77459	64459.87	0.21201
0.7	0.72969	0.85421	1.00458	51.37762	1.10791	1.19698	0.83666	64528.11	0.23544
0.8	0.83755	0.91512	1.00482	52.36688	1.12459	1.21501	0.89443	64564.56	0.24039
0.9	0.94812	0.97371	1.00512	53.06344	1.14667	1.23886	0.94868	64612.01	0.25178
1.0	1.06011	1.02962	1.00537	53.66973	1.16761	1.26148	1.0000	64656.26	0.26148

Ethylene glycol + 10% [DMF +H_2O]

Conc. (mol dm^{-3})	m (mol kg^{-1})	$\sqrt{m}$ (mol kg^{-1})$^{0.5}$	ρ (g cm^{-3})	ϕ_V (cm^3 mol^{-1})	η (cp)	$\eta_r = \eta/\eta_o$	$\sqrt{C}$ (mol$^{1/2}$ dm$^{-3/2}$)	ΔG^* (J mol^{-1})	$(\eta/\eta_o-1)/\sqrt{c}$
0.0	0.00000	0.00000	0.99992	0.00000	0.94882			64155.39	
0.1	0.10049	0.31700	1.00124	48.80841	0.99779	1.05161	0.31623	64276.86	0.16321
0.2	0.20202	0.44947	1.00242	49.44933	1.02389	1.07912	0.44721	64337.95	0.17692
0.3	0.30464	0.55194	1.00338	50.36551	1.04835	1.10489	0.54772	64394.10	0.19151
0.4	0.40846	0.63911	1.00412	51.35757	1.06682	1.12437	0.63246	64435.56	0.19664
0.5	0.51361	0.71667	1.00456	52.54963	1.08745	1.14611	0.70711	64481.95	0.20663
0.6	0.62011	0.78747	1.00482	53.64412	1.10932	1.16916	0.77459	64530.67	0.21839
0.7	0.72787	0.85315	1.00506	54.45098	1.12586	1.18659	0.83666	64566.76	0.22302
0.8	0.83725	0.91501	1.00516	55.23447	1.15089	1.21297	0.89443	64621.02	0.23811
0.9	0.94792	0.97361	1.00521	55.90051	1.16984	1.23294	0.94868	64661.38	0.24554
1.0	1.07231	1.03552	1.00529	56.71124	1.19233	1.25665	1.0000	64708.39	0.25665

Table 1. Continued

Ethylene glycol + 15% [DMF +H_2O]

Conc. (mol.dm^{-3})	m (mol kg^{-1})	$\sqrt{m}$ (mol kg^{-1})$^{0.5}$	ρ (g cm^{-3})	ϕ_V (cm^3 mol^{-1})	η (cp)	$\eta_r = \eta/\eta_o$	$\sqrt{C}$ (mol$^{1/2}$ dm$^{-3/2}$)	ΔG^* (J mol^{-1})	$(\eta/\eta_o-1)/\sqrt{c}$
0.0	0.00000	0.00000	1.00216	0.00000	0.98386			64239.73	
0.1	0.10029	0.31669	1.00317	51.82743	1.04159	1.05867	0.31623	64378.58	0.18554
0.2	0.20163	0.44903	1.00411	52.12675	1.07229	1.08989	0.44721	64448.26	0.20111
0.3	0.30414	0.55148	1.00496	52.49639	1.09666	1.11465	0.54772	64501.87	0.20933
0.4	0.40773	0.63854	1.00571	52.91199	1.12006	1.13844	0.63246	64552.36	0.21889
0.5	0.51283	0.71612	1.00623	53.61352	1.14314	1.16189	0.70711	64601.64	0.22895
0.6	0.61872	0.78659	1.00674	54.08853	1.16622	1.18535	0.77459	64649.93	0.23929
0.7	0.72632	0.85224	1.00721	54.47842	1.18829	1.20779	0.83666	64695.24	0.24836
0.8	0.83536	0.91398	1.00733	55.21669	1.21702	1.23699	0.89443	64754.17	0.26496
0.9	0.94585	0.97255	1.00739	55.85861	1.23593	1.25621	0.94868	64792.24	0.27007
1.0	1.05774	1.02846	1.00748	56.34004	1.25725	1.27787	1.0000	64834.41	0.27787

Ethylene glycol + 20% [DMF +H_2O]

Conc. (mol dm^{-3})	m (mol kg^{-1})	$\sqrt{m}$ (mol kg^{-1})$^{0.5}$	ρ (g cm^{-3})	ϕ_V (cm^3 mol^{-1})	η (cp)	$\eta_r = \eta/\eta_o$	$\sqrt{C}$ (mol$^{1/2}$ dm$^{-3/2}$)	ΔG^* (J mol^{-1})	$(\eta/\eta_o-1)/\sqrt{c}$
0.0	0.00000	0.00000	1.00448	0.00000	1.02468			64334.77	
0.1	0.10008	0.31635	1.00534	53.22415	1.09653	1.07012	0.31623	64500.64	0.22175
0.2	0.20126	0.44862	1.00611	53.62868	1.13046	1.10323	0.44721	64574.28	0.23083
0.3	0.30364	0.55104	1.00663	54.57348	1.15678	1.12892	0.54772	64630.05	0.23538
0.4	0.40724	0.63815	1.00705	55.28391	1.18231	1.15383	0.63246	64683.13	0.24322
0.5	0.51217	0.71566	1.00728	56.08667	1.20604	1.17699	0.70711	64731.82	0.25031
0.6	0.61849	0.78644	1.00734	56.90689	1.22852	1.19893	0.77459	64777.46	0.25685
0.7	0.72616	0.85215	1.00742	57.46231	1.24897	1.21889	0.83666	64818.18	0.26162
0.8	0.83521	0.91389	1.00751	57.86483	1.27222	1.24158	0.89443	64863.68	0.27009
0.9	0.94558	0.97241	1.00762	58.51132	1.29443	1.26326	0.94868	64906.31	0.27755
1.0	1.05745	1.02832	1.00767	59.31246	1.31281	1.28119	1.0000	64941.14	0.28119

The amounts of A and B have explained in Table 2.

The values of the A coefficients are positive, as shown in Table 2, indicating strong solute-solvent interactions. A and B coefficients formerly presented as an empirical term, that is dependent on solute-solvent interaction and the relative size of a solute and solvent molecules. Positive viscosity B coefficient levels describe a solute's hydrogen-bonded interactions with solvents. The positive values of ϕ_V^o and S_v indicate strong solute-solvent interactions [31].

CONCLUSIONS

The results of the current study show that ethylene glycol (0.1-1) M concentration added to mixtures of 5%, 10%, 15%, and 20% (w/w) [DMF + H_2O] causes the dissociation of H-bonded structures, which predominates over strong interactions in the mixtures. This is shown by the variation of the derived parameter value of the B-coefficient, which also shows the presence of solute-solvent interaction. This is also evident from the value positive of ϕ_V^o. The positive B values

Table 2. Values of Jones-Dole Coefficient (A, B) for Ethylene Glycol in H_2O and %(DMF + H_2O) Mixtures at 298.15 K

	η/η_o *vs. C*	$(\eta/\eta_o\text{-}1)/\sqrt{c}$ *vs.* $\sqrt{c}$			
%(DMF + H_2O) mixtures	B ($dm^3\ mol^{-1}$)	B ($dm^3\ mol^{-1}$)	A ($dm^{3/2}\ mol^{-1/2}$)	ϕ_V^o	S_v
0%	0.2417	0.2281	0.0028	35.588	12.540
5%	0.2494	0.2088	0.0540	40.830	12.334
10%	0.2321	0.1345	0.1163	44.338	11.755
15%	0.2455	0.1375	0.1367	49.134	6.591
20%	0.2373	0.0892	0.1895	49.908	8.830

also suggest that ethylene glycol in its various molarities serves as a structural component in solvent mixes.

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