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Experimental and computational study on viscosity and optical dielectric constant of solutions of poly (ethylene glycol) 200

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ABSTRACT

To understand the intermolecular interactions in binary mixtures of poly (ethylene glycol) 200 with ethanolamine, *m*-cresol and aniline respectively, absolute viscosities and refractive indices have been measured at varying concentrations and temperatures. Optical dielectric constant, polarisability, interaction parameter, enthalpy and entropy of viscous flow have been calculated. The viscosity data have been fitted by third order polynomial equation. The experimental data has been used to evaluate molar refraction deviation, deviation in viscosity and excess Gibb's free energy of activation for viscous flow and the results have been fitted to Redlich–Kister polynomial equation. The results reflect the intermolecular interaction and hydrogen bonding to be stronger in the binary mixture of poly (ethylene glycol) 200 with ethanolamine than with *m*-cresol and least with aniline. Different mixing rules have been applied to predict the optical dielectric constant of the liquid mixtures and their relative performances have been analysed.

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1. Introduction

Poly (ethylene glycol) (PEG) is a synthetic polymer that finds several industrial applications. These are frequently used in pharmaceutical and cosmetic fluids as solvents, carriers, humectants, lubricants, binders, bases and coupling agents [1] and also for extraction, separation and purification of biological materials [2,3]. The evaluation of thermodynamic and bulk properties of PEGs and their mixtures with organic solvents may prove quite relevant to the design and implementation of the above mentioned processes, especially for the establishment of reliable correlations and predictions. Poly (ethylene glycols) (PEGs) are polymeric materials whose chains are composed by oxyethylene units and terminated by hydroxyl groups at both ends which makes these materials suitable for synthesis of polyesters and polyurethane [4]. PEG hydroxyl end groups can interact via strong hydrogen bonds (H-bonds), developing relatively extended networks of associated molecules. Interaction with the solvent can perturb this H-bond network. In recent years, attention has been made on the solubility of PEG in both water and organic solvents.

Hence because of the industrial interest, experimental densities, viscosities and refractive indices data of PEG mixtures are important. In the present work binary mixtures of PEG 200 with ethanolamine, *m*-cresol and aniline have been investigated. The solvents used also belong to industrially important class of compounds. Ethanolamine is instrumental for the removal of acidic gases such as CO_2 and H_2S from gas streams in natural gas and petroleum industry and for treating gas streams in chemical production industry. *m*-Cresol, a good solvent for dissolving polymers, has many applications such as plasticizers, gasoline, additives, making explosives, pigments, disinfectants, fumigants and pharmaceutical intermediates. Aniline is used as an accelerator in vulcanization of rubber and also in the manufacturing of synthetic dyes, drugs etc. These solvents chosen have been a subject of considerable interest because mixing of PEG with ethanolamine (an amino alcohol), *m*-cresol (a phenol) and aniline (an amine) leads to H-bond formation due to the presence of reactive hydroxyl group –OH in *m*-cresol, amine group $-NH_2$ in aniline and both groups in ethanolamine. To our knowledge no densities, viscosities and refractive indices data have previously been reported for these mixtures.

Measurement of absolute viscosities and refractive indices of binary mixtures of poly (ethylene glycol) 200 with ethanolamine, *m*-cresol and aniline were performed at 293.15, 303.15 and 313.15 K, under atmospheric pressure. Optical dielectric constant, polarisability and the interaction parameter have been used to interpret the interactions in the mixtures. Enthalpy of activation and entropy of activation for viscous flow at all concentrations have been evaluated for the systems under consideration. The results of the measurements of viscosity were fitted to third order polynomial equation and their average absolute deviations were calculated. Molar refraction deviation, viscosity deviation and excess Gibb's free energy of activation for viscous flow at these three temperatures have been evaluated for the three mixtures over the whole composition range. The results have been fitted to Redlich–Kister polynomial equation. Different

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Table 1
Experimentally measured values of viscosities and refractive indices of pure components and their comparison with literature values.

Compound	T/K	η(mPa·s)		n_D	
		Experimental	Literature	Experimental	Literature
PEG	293.15	63.011	66.79[a]	1.460	1.4626[a]
	303.15	37.670	37.682[b]	1.456	1.4582[a]
	313.15	25.171	25.296[a]	1.452	1.4547[a]
Ethanolamine	293.15	24.124	24.1[c]	1.454	1.4541[d]
	303.15	15.103	15.11[c]	1.449	–
	313.15	10.024	10.02[c]	1.445	–
m-Cresol	293.15	17.887	–	1.540	1.5414[e]
	303.15	9.794	9.806[e]	1.535	–
	313.15	6.127	6.12[f]	1.530	
Aniline	293.15	4.418	4.404[g]	1.588	1.5866[h]
	303.15	3.188	3.19[i]	1.582	1.5814[h]
	313.15	2.469	2.42[i]	1.578	1.5809[h]

[a] Ref. [20].
[b] Ref. [21].
[c] Ref. [22].
[d] Ref. [23].
[e] Ref. [24].
[f] Ref. [25].
[g] Ref. [26].
[h] Ref. [27].
[i] Ref. [28].

Table 2
Experimentally measured values of viscosities and refractive indices for the systems PEG + ethanolamine, PEG + *m*-cresol and PEG + aniline, at 293.15, 303.15 and 313.15 K with mole fraction of PEG.

x_1	293.15 K		303.15 K		313.15 K	
	η(mPa·s)	n_D	η (mPa·s)	n_D	η(mPa·s)	n_D
PEG + ethanolamine						
0.0000	24.124	1.454	15.103	1.449	10.024	1.445
0.1009	35.203	1.455	23.981	1.452	15.449	1.447
0.2000	53.114	1.457	30.761	1.453	19.874	1.448
0.3012	63.382	1.457	35.526	1.453	22.577	1.449
0.4019	70.624	1.458	40.699	1.454	25.866	1.450
0.5002	70.640	1.458	40.691	1.454	26.145	1.450
0.6009	70.336	1.458	40.417	1.455	26.446	1.451
0.7006	67.537	1.459	40.117	1.455	25.661	1.451
0.8001	67.275	1.459	40.038	1.455	25.332	1.451
0.8999	64.403	1.459	38.572	1.456	25.223	1.452
1.0000	63.011	1.460	37.670	1.456	25.171	1.452
PEG + m-cresol						
0.0000	17.888	1.540	9.794	1.535	6.127	1.530
0.1049	37.922	1.529	18.298	1.525	10.954	1.515
0.2006	50.743	1.518	26.595	1.514	15.646	1.508
0.3017	58.019	1.508	31.579	1.504	19.093	1.498
0.4014	62.401	1.499	35.998	1.494	21.920	1.490
0.5011	64.929	1.491	37.318	1.486	22.842	1.482
0.5996	64.947	1.482	38.045	1.478	23.811	1.474
0.7000	64.014	1.474	38.166	1.470	23.416	1.466
0.8002	63.458	1.468	38.214	1.464	23.425	1.461
0.9000	63.200	1.463	37.697	1.459	23.950	1.455
1.0000	63.011	1.460	37.670	1.456	25.171	1.452
PEG + aniline						
0.0000	4.418	1.588	3.188	1.582	2.469	1.578
0.1019	8.978	1.567	5.792	1.562	4.322	1.556
0.1998	16.678	1.548	10.016	1.543	6.946	1.538
0.3003	26.885	1.533	16.121	1.527	10.020	1.522
0.4010	36.913	1.516	21.008	1.512	13.393	1.508
0.5021	46.661	1.504	26.143	1.499	16.590	1.496
0.5983	52.253	1.493	29.701	1.488	18.999	1.484
0.7249	57.419	1.481	33.784	1.477	21.550	1.472
0.8002	58.932	1.474	35.115	1.467	22.636	1.465
0.9001	61.248	1.465	36.876	1.461	23.991	1.457
1.0000	63.011	1.460	37.670	1.456	25.171	1.452

theoretical models have been used to predict optical dielectric constant of these mixtures using optical dielectric constant of components.

2. Experimental section

2.1. Preparation of mixtures

The analytical grade chemicals PEG200 (Fluka), ethanolamine (Aldrich, purified by redistillation, ≥99.5%), *m*-cresol (Fluka, puriss. p.a., ≥99.7%) and aniline (Sigma-Aldrich, ACS reagent, ≥99.5%) obtained from Sigma-Aldrich Chemicals Pvt. Ltd. were purified by standard procedure discussed by Perrin and Armarego [5]. Solutions have been prepared by mass, in air tight bottles, and measured on electronic balance OHAUS-AR 2104 (Ohaus Corp. Pine Brook, NJ, USA), with an accuracy of 1×10^{-4} g. The possible error in the estimation of mole fraction is less than ±0.0001.

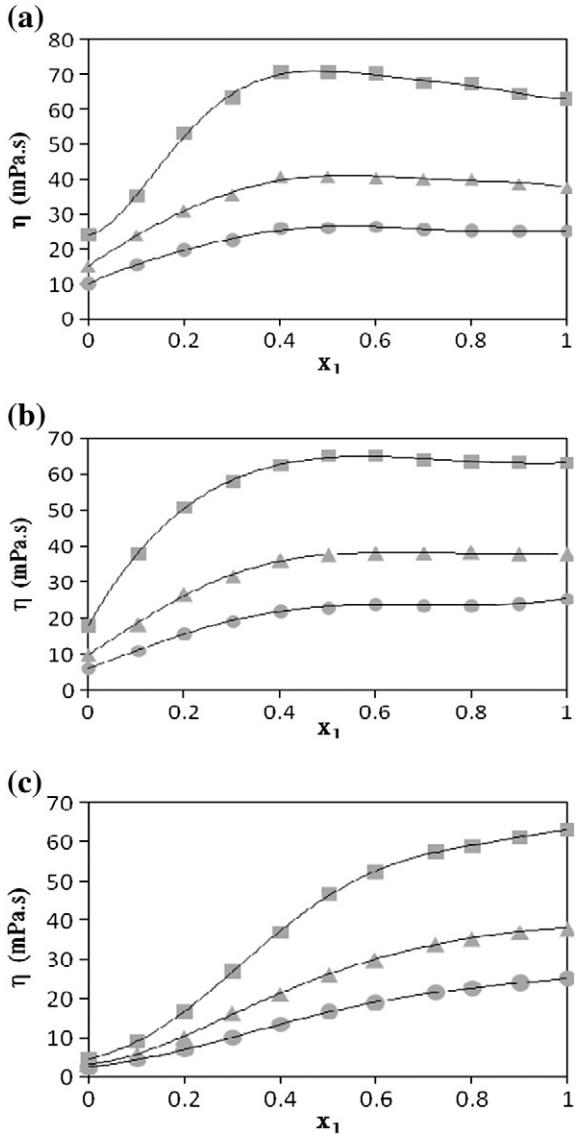

Fig. 1. (a). Absolute viscosity for the system PEG + ethanolamine at ■, 293.15; ▲, 303.15; and ●, 313.15 K with respect to the mole fraction of PEG. (b). Absolute viscosity for the system PEG + *m*-cresol at ■, 293.15; ▲, 303.15; and ●, 313.15 K with respect to the mole fraction of PEG. (c). Absolute viscosity for the system PEG + aniline at ■, 293.15; ▲, 303.15; and ●, 313.15 K with respect to the mole fraction of PEG.

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