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Thermophysical properties of binary mixtures of polyethylene glycol 400 with *n*-propanol

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Densities (ρ), viscosities (η) and refractive indices (n_D) of the binary system polyethylene glycol 400+*n*-propanol have been measured at (293.15, 298.15 and 303.15) K and at atmospheric pressure, over the entire composition range. From experimental results, the values of excess properties have been calculated and fitted to the Redlich-Kister equation. Negative deviations are observed for excess molar volumes, deviation in viscosities and excess internal pressures, while positive deviations are observed in the case of refractive indices and excess Gibbs energies of activation of viscous flow at all of the temperatures and in the entire concentration range. Of the studied models, the Jouyban-Acree model is suitable for describing the viscosity of the PEG 400+*n*-propanol binary system. The refractive index predicted by the Newton-mixing rule shows the best agreement with experimental values. The values of thermodynamic functions of activation viz., $\Delta G^{\#}$, $\Delta H^{\#}$ and $\Delta S^{\#}$, are positive for the PEG 400+*n*-propanol binary system.

Keywords: Solution chemistry, Polyethylene glycol, *n*-Propanol, Thermophysical properties, Excess properties, Binary systems, Density, Refractive indices

The knowledge of thermodynamic and transport properties of environmentally friendly fluids and their mixtures as a function of composition and temperature is of considerable interest for industrial application[1]. Densities and viscosities of multicomponent liquid mixtures are needed for the design of various processes in many chemical and engineering disciplines[2].

The study of the dependence of physical properties on the composition and external conditions of a binary liquid system, is a useful tool in getting information about the structure of various liquids and in studying liquid-liquid interactions[2]. The excess thermodynamic and transport properties for binary mixtures yield important information concerning the molecular liquid structure and intermolecular interactions[3-6].

Polyethylene glycol (PEG) is a nontoxic, highly biodegradable polymer[7]. It is both a good proton donor and proton acceptor[8]. PEG is an important industrial solvent, which may be used in the cleaning of exhaust air and gas streams from industrial production plants because of its favorable properties, such as low vapor pressure, low toxicity, high chemical stability, and low melting point[9]. The main advantage of PEG is also high solubility and desorption capability for SO_2, which leads to lower energy requirements for absorption and regeneration processes[9].

Among the numerous organic solvents, alcohols show favorable absorption and desorption capabilities for the removal of SO_2 in the industrial processes[10].

In this study the densities, viscosities and refractive indices of the binary system of polyethylene glycol 400 (PEG 400) and *n*-propanol were measured at three temperatures (293.15, 298.15 and 303.15) K and at atmospheric pressure, over the entire composition range. A literature survey shows that this system has not been studied till date. From our experimental data, excess molar volumes, deviation in viscosities, deviation in refractive indices, excess internal pressures and excess Gibbs energies of activation of viscous flow, were calculated and correlated by the Redlich-Kister equation[11]. The energies of activation of viscous flow were also calculated.

Experimental

The chemicals *n*-propanol (mass ≥99.5%, CAS 71-23-8, water content ≤0.05%) was supplied by Merck and PEG 400 (CAS 25322-68-3, water content ≤1%) was obtained from Roth. The chemicals were dried over molecular sieves (Fluka type 4 Å). The purity of the components was verified by chromatographic analysis (*n*-propanol mass fraction purity ≥0.995, PEG 400 mass fraction purity ≥0.993).

The binary solutions were prepared by weighing in Adventurer Pro AV 264CM balance at atmospheric pressure and ambient temperature with a precision of $\pm 10^{-4}$ g. The experimental error in mole fraction is estimated to be less than ±0.0001.

The densities were measured using a calibrated glass pycnometer (bulb vol.: 10 cm^3). The volume of the pycnometer was calibrated as a function of temperature using distilled, deionized, and degassed water at various temperatures[12]. The pycnometer filled with liquid was kept in a U-10 Freital thermostatic bath (maintained constant to ±0.05 K) for 20–25 min to reach thermal equilibrium. The density measurements were performed at various temperatures. The estimated uncertainty for density was ±0.0002 g cm^{-3}. The uncertainty in excess molar volume is estimated to be ±5×10^{-4} cm^3 mol^{-1}.

Viscosities of the binary mixtures and of the pure compounds were determined with an Ubbelohde kinematic viscometer[13], viscosity measuring unit ViscoClock (Schott-Gerate GmbH), that was kept in a vertical position in a thermostatic bath (U10), constant to ±0.05 K. The kinematic viscosity was calculated using the equation,

$$\nu = At - (B/t) \qquad \text{... (1)}$$

where ν is the kinematic viscosity, t (s) is the flow time and, A and B are characteristic constants of the used viscometer. The constants A and B were determined by taking doubly distilled water and benzene as the calibrating liquids. Accuracy of time measurement was ±0.01s. The dynamic viscosity was determined from the relation,

$$\eta = \nu\rho \qquad \text{... (2)}$$

where ρ is the density of the liquid. The precision of the viscosity was found to be ±0.0003 mPas. Each measurement was repeated at least three times, and the results were averaged. The uncertainty in the deviation in viscosity is estimated to be ±4x10^{-4} mPa s.

The refractive indices of pure liquids and their binary mixtures were measured using a thermostatted Abbe refractometer (Atago, model NAR–1T Liquid). Calibration of the instruments was done by measuring the refractive indices of doubly-distilled water and toluene at known temperature. The values of refractive index were obtained using sodium D light. The temperature of the test liquids, between the prism of refractometer during the measurements was maintained to an accuracy of ±0.05 K by circulating water through the jacket around the prism from a controlled thermostatic bath. The temperature was measured with an digital thermometer connected with the prism jacket[14]. The error in the refractive indices measurements was less than 0.0002 units. In all the determinations, triplicate experiments were performed at each composition and temperature.

Results and discussion

The measured densities, viscosities and refractive indices of the pure components are in reasonable agreement with the literature values[1,15-25] (Supplementary data, Table S1). The densities, viscosities and refractive indices of the binary mixtures of PEG 400 + *n*-propanol at (293.15, 298.15 and 303.15) K are reported in Table 1.

PEG 400 density values reported in the literature differ from our experimental data with a maximum of 0.01%. For *n*-propanol, density values published in the literature differ with a maximum 0.06%. Viscosity values reported in the literature differ from our results with a maximum 0.8% for PEG 400 and a maximum 2% for *n*-propanol. For PEG 400, refractive index values published in the literature, differ by less than 0.05% and for *n*-propanol differ by less than 0.1%.

Densities of the pure compounds and binary mixtures were correlated with temperature using the equation[26]:

$$\rho = a_0 + a_1 T \qquad \text{...(3)}$$

where a_0 and a_1, are the estimated parameters and T is the temperature.

Table 1 — Densities (ρ), viscosities (η) and refractive indices (n_D) of the PEG 400 (x_1)+*n*-propanol mixtures at different temperatures[a] and at atmospheric pressure

x_1	ρ (g cm^{-3})[b]			η (mPa s)[c]			n_D[d]		
	293.15	298.15	303.15	293.15	298.15	303.15	293.15	298.15	303.15
0.0990	0.9146	0.9103	0.9070	6.8241	5.6909	5.0301	1.4160	1.4140	1.4121
0.1992	0.9811	0.9771	0.9736	13.7508	11.5274	9.8168	1.4333	1.4312	1.4293
0.2947	1.0245	1.0206	1.0169	23.1393	18.6269	15.7910	1.4422	1.4401	1.4380
0.3978	1.0549	1.0510	1.0472	35.4202	28.2954	23.2308	1.4494	1.4472	1.4453
0.4959	1.0750	1.0713	1.0674	47.5746	38.0319	30.6250	1.4542	1.4521	1.4502
0.6016	1.0909	1.0873	1.0833	62.0674	49.7034	38.9827	1.4581	1.4560	1.4541
0.6851	1.0997	1.0961	1.0920	74.3382	58.6337	45.6716	1.4610	1.4589	1.4570
0.8013	1.1109	1.1074	1.1032	91.3012	71.0846	55.7954	1.4629	1.4608	1.4587
0.9039	1.1193	1.1157	1.1115	105.5938	82.4733	64.3245	1.4641	1.4620	1.4601

Uncertainties, u, are: [a]$u(T)$ = ± 0.05 K; [b]$u(\rho)$ = ±0.0002 g cm^{-3};
[c]$u(\eta)$= ± 0.0003 mPa s; [d]$u(n_D)$= ±0.0002; $u(t)$ = ± 0.01 s.

Viscosities of the pure compounds and binary mixtures were correlated with temperature using Guzman's equation[26]:

$$\eta = \eta_0 e^{\frac{E_a}{RT}} \qquad \text{...(4)}$$

where η_0 and E_a are the adjustable parameters.

The adjustable parameters of Eqs 3 and 4 were estimated using the experimental data and a nonlinear regression analysis employing the Levenberg-Marquardt algorithm[27]. The fitting parameters along with the correlation square coefficient (r^2) and standard deviation (σ) are given in Supplementary data, Table S2.

The values of standard deviation and correlation coefficient, indicate that these two equations are able to correlate well with the experimental density and viscosity values.

Several empirical and semi-empirical equations have been developed to describe the viscosity of liquid mixtures. Some of the proposed equations require a series of adjustable parameters[28] and were therefore classified according to the number of these parameters[29,30]. In order to examine the dependence of viscosity on the concentration of the components in the binary liquid mixtures, several relations based on one, two, three or four adjustable parameters viz., Grunberg-Nissan[31], Heric-Brewer[32], McAllister[33] and Jouyban Acree[34,35] were tested.

The viscosity coefficients were estimated using the experimental viscosity data and a non-linear regression analysis employing the Levenberg-Marquardt algorithm[27].

The calculated parameters and standard deviations (σ) for the above-mentioned relations are shown in Table 2.

From the data presented in Table 2, it can be seen that for the equation with a small number of adjustable parameters, the values of standard deviation are high. These equations do not give good results and cannot be used to correlate the viscosity data. The Jouyban Acree equation (with four adjustable parameters) shows results in good agreement with the experimental values for all temperatures.

Refractive indices of liquid mixtures can be calculated from experimental density data and refractive indices of pure components using different mixing rules[36] viz., Arago-Biot (A-B)[37], Dale-Gladstone (D-G)[38], Newton (Nw)[39], Eyring-John (E-J)[40], Lorentz-Lorenz (L-L)[41,42], Weiner (WR)[43], and Heller (Hr)[44]. The standard deviation values calculated for these models are given in Table 3. From the data presented in Table 3, it can be noticed that there are good concordances between the experimental values and those calculated with all the tested relations. However, the smallest values of standard deviation are obtained with the Newton equation at all temperatures, so this equation is the best.

From the measured densities of the pure liquids and their mixtures, the excess molar volumes (V^E) were computed using the equation:

$$V^E = \frac{[x_1M_1 + x_2M_2]}{\rho} - \left[\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2}\right] \qquad \text{... (5)}$$

where x_1 and x_2 are the mole fractions, M_1 and M_2 are the molecular masses and, ρ, ρ_1 and ρ_2 are the respective densities of the solution and components 1 and 2 respectively.

Table 2 — Parameters for the Grunberg-Nissan, Heric-Brewer, McAllister and Jouyban-Acree equations and standard deviations (mPa s) at different temperatures

Model	Parameters	Temp. (K)		
		293.15	298.15	303.15
Grunberg-Nissan	d	4.53018	4.35208	4.21588
	σ	0.1900	0.1778	0.1854
Heric-Brewer	α_{12}	6.17703	5.94945	5.86313
	α_{21}	-3.63455	-3.47382	-3.57457
	σ	0.0749	0.0688	0.0726
McAllister four body model	η_{1112}	107.76982	83.97546	64.9961
	η_{1122}	37.03294	30.99842	23.39711
	η_{2221}	75.8597	57.63452	50.74696
	σ	0.0271	0.0244	0.0228
Jouyban-Acree	A_0	1257.16015	1233.11835	1206.45873
	A_1	-669.84395	-637.04924	-694.67447
	A_2	478.71399	434.17839	483.92069
	A_3	-324.1299	-320.35094	-274.68985
	σ	0.0130	0.0073	0.0092

Table 3 — Standard deviation for the Arago-Biot, Dale-Gladstone, Newton, Eyring-John, Lorentz-Lorenz, Weiner and Heller equations at different temperatures

Mixing rules	σ		
	293.15 K	298.15 K	303.15 K
A-B	0.0021	0.0021	0.0019
D-G	0.0021	0.0021	0.0019
Nw	0.0018	0.0018	0.0016
E-J	0.0023	0.0023	0.0021
L-L	0.0025	0.0025	0.0023
WR	0.0022	0.0022	0.0021
Hr	0.0023	0.0023	0.0022

The deviations in viscosity ($\Delta\eta$) were calculated from the viscosities of the pure components and mixtures using the relation:

$$\Delta\eta = \eta - [x_1\eta_1 + x_2\eta_2] \quad \ldots (6)$$

where η_1 and η_2 are the viscosities of pure components.

The experimental values of refractive indices are used to calculate the deviation of refractive indices (Δn_D) defined by the equation:

$$\Delta n_D = n_D - [x_1 n_{D1} + x_2 n_{D2}] \quad \ldots (7)$$

where n_D, n_{D1} and n_{D2} refer to the refractive index of the mixture and the pure components 1 and 2, respectively.

The excess internal pressure (P_i^E) was calculated using the relation:

$$P_i^E = P_i - [x_1 P_{i1} + x_2 P_{i2}] \quad \ldots (8)$$

where P_i, P_{i1} and P_{i2} are the internal pressures of the mixtures and the pure components, respectively, which can be obtained from the Buchler-Hirschfelder-Curtis equation of state[45].

The excess Gibbs energies of activation of viscous flow (ΔG^E) were calculated using the equation:

$$\Delta G^E = RT[ln(\eta V) - x_1 ln(\eta_1 V_1) - x_2 ln(\eta_2 V_2)] \ldots (9)$$

The excess molar volumes (V^E), deviation in viscosities ($\Delta\eta$), deviation of refractive indices (Δn_D) and excess internal pressure (P_i^E) were correlated with the Redlich-Kister equation[11].

The values of the polynomial coefficients of the Redlich-Kister equation, along with the standard deviation (σ), are presented in Table S3 (Supplementary data).

The experimental excess properties and the fitting curves calculated from the Redlich-Kister equation are presented in Figs 1-5.

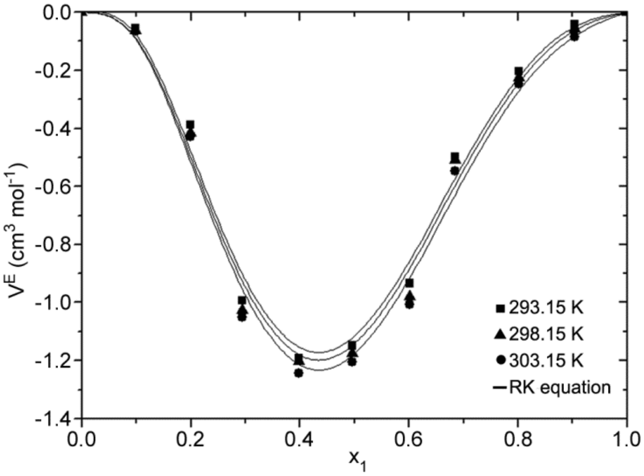

Fig. 1 — Experimental values of excess molar volume as a function of PEG 400 molar fraction x_1.

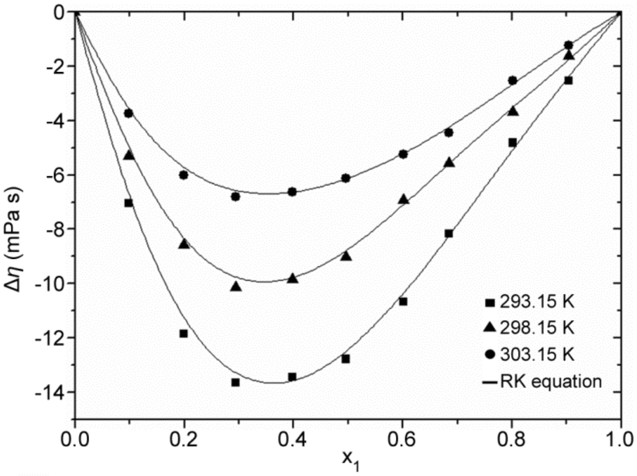

Fig. 2 — Experimental values of deviation in viscosity as a function of PEG 400 molar fraction x_1.

The excess molar volumes for PEG 400+*n*-propanol are negative in the entire composition range and at all investigated temperatures. The negative contributions are a consequence of the following effects: (*i*) strong intermolecular interactions attributed to the charge-transfer complex, dipole-dipole and dipole-induced

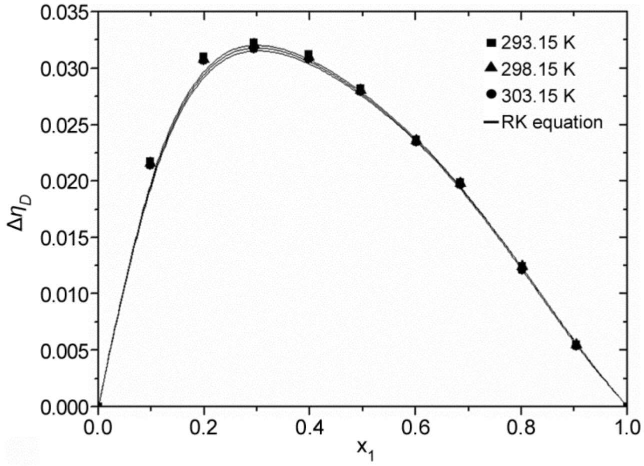

Fig. 3 — Experimental values of deviation of refractive index as a function of PEG 400 molar fraction x_1.

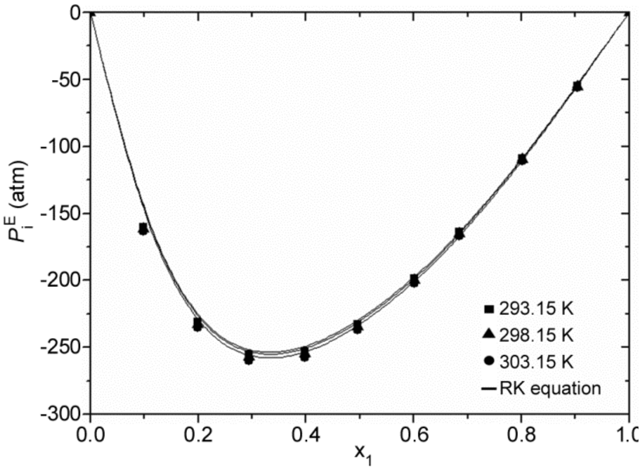

Fig. 4 — Experimental values of excess internal pressure as a function of PEG 400 molar fraction x_1.

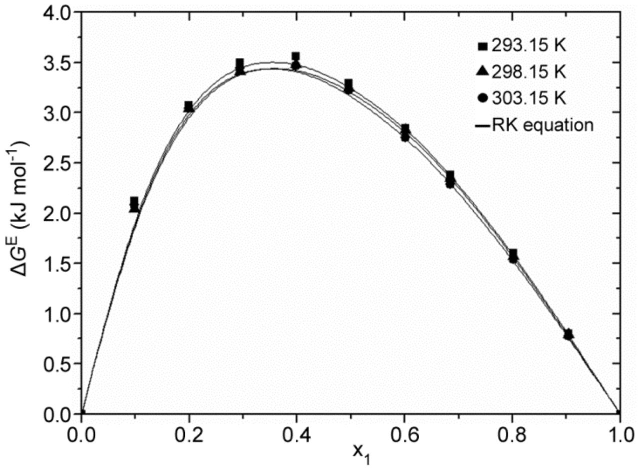

Fig. 5 — Experimental values of excess Gibbs energies as a function of PEG 400 molar fraction x_1.

dipole interactions and H-bonding between unlike molecules finally leading to the more efficient packing in the mixture than in the pure liquids; (*ii*) structural effects which arise from suitable interstitial accommodation giving more compact structure of mixtures[15].

Our results indicate the presence of strong molecular interactions in the mixtures attributed to dipole-dipole and H-bonding between unlike molecules. Both the components have good hydrogen bonding ability and polar nature allowing dipole-dipole interactions[20].

The structure of *n*-propanol alone, as well as in the mixture with PEG 400, is determined by interactions through the -OH groups. *n*-Propanol as a monohydroxyl alcohol forms mainly linear associates where the molecules of PEG 400 are involved in cross-linking H-bonds giving negative values of V^E [20].

Chen *et al.*[46] reported that in polar organic solvents PEG molecules have a helical polar structure with gauche –C–C– bonds, *trans* –O–C– bonds and hydrogen bonds between the solvent molecules and the oxygen atoms in PEG chain. Volume contractions that determine negative values of excess molar volume can also be attributed to large differences in the size of molecules of solution components. The molar volumes of *n*-propanol and PEG 400 at 298.15 K, are 75.2 and 356.3 cm^3 mol^{-1}, respectively. The molar volume of PEG 400 is nearly five times larger as compared to *n*-propanol. The smaller propanol molecules may fit into the voids created by larger PEG 400 molecules, reducing the volume of the mixture and resulting in negative values of molar excess volume. With a rise in temperature, the negative values of excess molar volumes increase. More negative values of excess volume at higher temperatures show that in these systems the structural effects are more important than the effect of molecular interactions on excess molar volumes.

The values of deviation in viscosity are negative at all investigated temperatures. The positive values are an indication of specific interactions[47,48].

The negative values of deviation in viscosity of the studied system indicate that the strength of specific interactions is not the only factor influencing the deviation in viscosity of liquid mixtures. The molecular size and shape of the components also play an important role[21]. The inclusion of propanol molecules in the interstices of PEG molecules reduces the surface available for friction, increasing mobility and thus decreasing the viscosity of the mixture.

The negative values of deviation in viscosity decrease with an increase in temperature. An increase in temperature decreases self-association and also the association between unlike components because of the increase in thermal energy[17]. This fact explains the less negative values of deviation in viscosities as the temperature is increased.

The experimental values of the deviations of refractive indices are positive at all temperatures. This kind of behaviour indicates that the dispersion interactions between the unlike molecules in the mixture are stronger than in the pure components[49-51]. Increasing the temperature produces a small decrease in refractive index deviation, which indicates a slight reduction of the dispersion forces in the propanol+PEG system.

The negative values of excess internal pressure in the entire composition range and at all temperatures indicate strong intermolecular interactions[52]. This is in accordance with the negative values of excess molar volume.

The excess Gibbs energies of activation of viscous flow are positive over the whole range of composition and at all temperatures. The sign of ΔG^E values is considered as a reliable criterion for detecting or excluding the presence of interaction between different molecules[53]. The positive value indicates the existence of strong attractive interactions[54]. Positive ΔG^E may be attributed to specific interactions like H-bonding and charge transfer[47,55].

The energies of activation of viscous flow were calculated using the equation:

$$\eta = \frac{hN}{V} exp\left(\frac{\Delta G^{\#}}{RT}\right) \quad \ldots (10)$$

where η is the viscosity of a liquid mixture, h is Planck's constant, N is Avogadro's number, V is the molar volume of the solution, R is the universal gas constant, T is temperature and $\Delta G^{\#}$ is the molar Gibbs energy of activation of viscous flow.

The plots of $\ln(\eta V/hN)$ versus $1/T$ are linear in the temperature range 293.15 to 303.15 K and the values of $\Delta H^{\#}$ and $\Delta S^{\#}$ were obtained from the corresponding slopes and intercepts. From these results the values of $\Delta G^{\#}$ were also calculated. The values of thermodynamic functions of activation of viscous flow and the correlation square coefficient (r^2) are listed in Table S4 (Supplementary data), as a function of composition.

The values of $\Delta G^{\#}$ and $\Delta H^{\#}$ are positive for the binary system of PEG 400+*n*-propanol suggesting specific interactions, like H-bonding, between solution components. These values increase with the PEG 400 concentration of solution at temperature constant. The values of free energy at constant concentration increase if the temperature increases. The values of the activation entropy of viscous flow are positive over the whole mole fraction range. The positive $\Delta S^{\#}$ values are indicative of less overall molecular order due to activated complex formation for viscous flow.

In summary, the densities, viscosities and refractive indices of binary mixture of PEG 400 with *n*-propanol were measured experimentally at temperatures between 293.15 and 303.15 K over the entire composition range. From experimental results, the values of the excess molar volume, deviation in viscosity, deviation of refractive index, excess internal pressure and excess Gibbs energies of activation of viscous flow, have been calculated and fitted to the Redlich-Kister equation. Negative deviations were observed for excess molar volumes, deviation in viscosities and excess internal pressures, while positive deviations were observed for refractive indices and excess Gibbs energies of activation of viscous flow at all the studied temperatures over the entire concentration range. Grunberg-Nissan, Heric-Brewer, four-body McAllister and Jouyban Acree models have been used to calculate viscosity coefficients and these were compared with experimental data for the mixtures. The results of these correlations showed that Jouyban Acree model is suitable to describe viscosities of the binary mixture studied in this work.

The refractive indices were compared with the predicted results from different mixing rules (Arago-Biot, Gladstone-Dale, Newton, Eyring-John, Lorentz-Lorenz, Weiner and Heller). The refractive index predicted by Newton rule, showed the best agreement with the experimental values. The energies of activation of viscous flow were calculated. The values of $\Delta H^{\#}$, $\Delta G^{\#}$ and $\Delta S^{\#}$ are positive at all the temperatures and in the entire concentration range.

Supplementary data

Supplementary data associated with this article are available in the electronic form at http://www.niscair.res.in/jinfo/ijca/IJCA_57A(8-9)1144-1150_SupplData.pdf.

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