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Viscosity Behavior of Some Oxygen Containing Compounds

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Abstract Densities (ρ) and viscosities (η) for the binary mixtures of propylene carbonate with tetrahydrofuran (THF), 1,4-dioxane, and acetylacetone were measured over the entire mole fraction range at (288.15, 298.15, and 308.15) K. The results are discussed in the light of intermolecular interactions present among the components.

Keywords Acetylacetone · Binary mixtures · Density · 1,4-Dioxane · Propylene carbonate · Tetrahydrofuran · Viscosity

1 Introduction

The thermodynamic and transport properties of liquid mixtures have been used to understand the molecular interactions between the components of a mixture and also for engineering applications. Densities and viscosities of binary mixtures are important from a theoretical point of view to understand liquid theory. A knowledge of densities and viscosities is required for the design of new processes and the latter for evaluation of mass-transfer phenomena and designing equipment. Viscosity data give valuable information about the nature of interaction forces operating within and

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between molecules. Many engineering problems require quantitative data on the viscosity and density of liquid mixtures.

Propylene carbonate is a highly polar aprotic solvent with a large dipole moment. It shows dipole–dipole interactions. It is used in the production of polycarbonates and polyester elastomers, for separation of carbon dioxide and hydrogen sulfide and refinery aromatics in the oil industry, and as a component of electrolytes in lithium ion rechargeable batteries.

Tetrahydrofuran (THF) is used as a solvent for higher polymers, especially polyvinyl chloride, as a reaction medium for Grignard and metal hydride reactions, and in the synthesis of butyrolactone, succinic acids, and 1,4-butanediol diaceate. 1,4-Dioxane is used as a stabilizer in chlorinated solvents, and a solvent for cellulose acetate, ethyl cellulose, benzyl cellulose, resin, oils, waxes, oil and spirit sol dyes, and many other organic as well as inorganic compounds. Acetylacetone forms organometallic complexes, which are used as gasoline additives, drivers of varnishes, and printer's ink, fungicides, insecticides, colors, etc.

A literature survey reveals that many data are available for the thermodynamic properties of 1,4-dioxane and THF containing liquid mixtures [1–8], but not much work has been carried out on acetylacetone. Also, no measurements have been previously reported for the mixtures considered in this study.

The present paper is a continuation of our study on the thermodynamic properties of liquid mixtures containing propylene carbonate as one of the components [9–14]. We report here densities and viscosities of binary mixtures of propylene carbonate with THF, 1,4-dioxane, and acetylacetone at (288.15, 298.15, and 308.15) K and at atmospheric pressure.

2 Experimental

Propylene carbonate (Merck, >99) was refluxed over anhydrous calcium carbonate and distilled at atmospheric pressure [15]. THF (Spectrochem. Pvt. Ltd.), 1,4-dioxane (SD Fine Chem), and acetylacetone (Spectrochem. Pvt. Ltd.) were distilled at atmospheric pressure. All the liquids were triply distilled. The middle fraction collected off all the liquids was stored over 4 Å molecular sieves. The purity of the purified liquids was checked by comparing the measured densities and viscosities with those reported in the literature [15–18]. The experimental and literature values are shown in Table 1.

Table 1 Densities and viscosities of pure components at 298.15 K

Compound	ρ (g · cm^{-3})		η (mPa · s)	
	Exptl.	Lit.	Exptl.	Lit.
Propylene carbonate	1.19758	1.1978 [16]	2.4997	2.5009 [16]
Tetrahydrofuran	0.88196	0.88193 [17]	0.4615	0.460 [17]
1,4-Dioxane	1.02792	1.0280 [18]	1.1854	1.1825 [18]
Acetylacetone	0.97218	0.9721 [15]	0.6699	–

The binary liquid mixtures were prepared by mixing known masses of pure liquids in air-tight stoppered bottles in order to minimize evaporation losses. All measurements of mass were performed on a Mettler single-pan balance which can be read to the fifth decimal place, with an uncertainty of 0.05 mg.

The densities of the pure components and their mixtures were measured using a single-arm capillary pycnometer having a bulb volume of approximately 5 cm^3 and a capillary bore with an internal diameter of 0.75 mm. The uncertainty in the density measurements was found to be $5 \times 10^{-5}\ \text{g} \cdot \text{cm}^{-3}$.

The volume (v) of the pycnometer up to the mark in the capillary was determined using triply distilled water at 288.15 K by taking the mass (m) of water and the literature density [19] at 288.15 K using the relation $v = m/\rho$. The density for the next temperature at 293.15 K was determined by measuring an increase in the height of the water level from the mark with the help of a travelling microscope having a least count of 0.001 cm. The increased volume (v_i) of water was calculated from the equation $v_i = \pi r^2 h$, where h is the height of the increased water level and r is the radius of the capillary and is added to the volume v at 288.15 K.

The density of triply distilled water at 293.15 K was calculated using the relation $\rho = m/(v + v_i)$, where m is the mass at 288.15 K. The observed density (0.99821 $\text{g} \cdot \text{cm}^{-3}$) was close to the literature value [19] (0.99826 $\text{g} \cdot \text{cm}^{-3}$). The densities of the mixtures were determined by taking the mass at 288.15 K and using this mass and v_i at each temperature as described above.

Viscosity measurements were performed by using Schott Gerate (AVS 350) viscosity measuring equipment with a series of Ubbelhode viscometers. According to the method suggested by Hagenbach (AVS 350, viscosity measuring system, Introduction Manual Schott Gerate Hofheim, IS Germany 1986), replicate experiments were performed (five times) for each solution and the results were averaged. Several viscometers were used, with capillary diameters from (0.36 to 1.13) mm, for kinematic ranges of (0.3 to 10) $\text{mm}^2 \cdot \text{s}^{-1}$, respectively. In all the measurements, kinetic energy corrections have been taken into account. The experimental repeatability of the viscosity measurements of each solution was found to be ±0.2 %, with an uncertainty of 3×10^{-2}. The details of the density and viscosity measurements are given in previous papers [12–14].

For all the measurements, the temperature was controlled by circulating the water through an ultra-thermostat (JULABO F-25) which has an uncertainty of 0.02 °C.

3 Results and Discussion

The experimental values of density (ρ) and viscosity (η) for binary mixtures of propylene carbonate with THF, 1,4-dioxane, and acetylacetone at (288.15, 298.15, and 308.15) K over the whole composition range are listed in Tables 2, 3, and 4, respectively.

Dynamic viscosities (η) of binary mixtures of propylene carbonate (1) + oxygen (2) were calculated by using densities and flow times from

$$\eta = K\rho\,(t - HC) \tag{1}$$

Table 2 Values of densities (ρ) and viscosities (η) as a function of mole fraction, x_1, for binary mixtures of propylene carbonate (1) + THF (2) at different temperatures

x_1	$\rho \times 10^{-3}$ (kg · m^{-3})			η (mPa · s)		
	288.15 K	298.15 K	308.15 K	288.15 K	298.15 K	308.15 K
0.0000	0.89293	0.88196	0.87085	0.508	0.462	0.426
0.0188	0.90115	0.89025	0.87917	0.524	0.470	0.432
0.0393	0.91003	0.89918	0.88814	0.542	0.479	0.439
0.0603	0.91897	0.90819	0.89718	0.561	0.490	0.448
0.0770	0.92600	0.91527	0.90428	0.575	0.500	0.455
0.0988	0.93505	0.92438	0.91341	0.594	0.513	0.466
0.2117	0.97975	0.96945	0.95845	0.696	0.600	0.541
0.3057	1.01439	1.00406	0.99326	0.794	0.699	0.629
0.4024	1.04772	1.03725	1.02668	0.921	0.831	0.745
0.4992	1.07898	1.06845	1.05796	1.088	0.998	0.890
0.5997	1.10933	1.09874	1.08829	1.317	1.211	1.072
0.7015	1.13816	1.12739	1.11706	1.626	1.473	1.289
0.8033	1.16519	1.15455	1.14402	2.030	1.786	1.540
0.9064	1.19096	1.17968	1.16974	2.556	2.158	1.830
1.0000	1.20867	1.19758	1.18716	3.086	2.500	2.073

Table 3 Values of densities (ρ) and viscosities (η) as a function of mole fraction, x_1, for binary mixtures of propylene carbonate (1) + 1,4-dioxane (2) at different temperatures

x_1	$\rho \times 10^{-3}$ (kg · m^{-3})			η (mPa · s)		
	288.15 K	298.15 K	308.15 K	288.15 K	298.15 K	308.15 K
0.0000	1.03910	1.02792	1.01655	1.440	1.185	1.029
0.0213	1.04260	1.03146	1.02015	1.429	1.181	1.027
0.0410	1.04589	1.03479	1.02354	1.421	1.177	1.027
0.0636	1.04976	1.03870	1.02751	1.413	1.174	1.020
0.0823	1.05301	1.04199	1.03085	1.408	1.173	1.026
0.0999	1.05612	1.04513	1.03402	1.404	1.172	1.022
0.2020	1.07480	1.06402	1.05309	1.408	1.184	1.028
0.2990	1.09328	1.08269	1.07187	1.450	1.220	1.053
0.3991	1.11262	1.10192	1.09144	1.534	1.286	1.102
0.4991	1.13172	1.12115	1.11070	1.659	1.381	1.171
0.5987	1.15006	1.13940	1.12910	1.826	1.508	1.274
0.6996	1.16740	1.15675	1.14643	2.039	1.671	1.405
0.8049	1.18366	1.17285	1.16259	2.307	1.878	1.571
0.9003	1.19616	1.18545	1.17502	2.589	2.101	1.748
1.0000	1.20867	1.19758	1.18716	3.086	2.500	2.073

Table 4 Values of densities (ρ) and viscosities (η) as a function of mole fraction, x_1, for binary mixtures of propylene carbonate (1) + acetylacetone (2) at different temperatures

x_1	$\rho \times 10^{-3}$ (kg · m^{-3})			η (mPa · s)		
	288.15 K	298.15 K	308.15 K	288.15 K	298.15 K	308.15 K
0.0000	0.98034	0.97218	0.96346	0.741	0.670	0.609
0.0212	0.98424	0.97579	0.96683	0.769	0.692	0.627
0.0438	0.98861	0.97989	0.97069	0.797	0.714	0.646
0.0625	0.99242	0.98348	0.97411	0.821	0.732	0.661
0.0827	0.99668	0.98754	0.97799	0.845	0.751	0.677
0.1026	1.00103	0.99171	0.98200	0.868	0.769	0.693
0.2010	1.02450	1.01455	1.00429	0.978	0.857	0.766
0.2982	1.05005	1.03983	1.02934	1.086	0.944	0.838
0.4028	1.07889	1.06864	1.05814	1.215	1.047	0.922
0.5000	1.10574	1.09561	1.08524	1.359	1.162	1.014
0.6016	1.13267	1.12270	1.11252	1.549	1.313	1.133
0.6985	1.15608	1.14619	1.13615	1.782	1.495	1.277
0.7995	1.17695	1.16695	1.15695	2.093	1.735	1.466
0.9016	1.19311	1.18269	1.17251	2.493	2.040	1.706
1.0000	1.20867	1.19758	1.18716	3.086	2.500	2.073

where K and ρ are a viscometer constant and the density of the mixture, respectively. $(t - HC)$ is the flow time adjusted by the Hagenbach correction factor,

$$HC = E/(Kt^2),$$

The value of $E/K = 70\,500$ is given in the instruction manual provided by the manufacturer. The viscometer constant was calibrated using double-distilled water at 298.15 K. The viscometers with $t > 200$ s were selected for measurements.

The values of densities and viscosities for all the binary mixtures are found to decrease with mole fraction and with increasing temperatures. The trend in both properties are the same, as, THF < acetylacetone < 1,4-dioxane. The graphical variation of densities and viscosities for all the binary mixtures at 298.15 K is shown in Figs. 1 and 2, respectively. The increase in the values of densities and viscosities for binary mixtures with the addition of propylene carbonate indicates a low level of interactions in these mixtures.

The molar volumes of propylene carbonate, THF, 1,4-dioxane, and acetylacetone at 298.15 K are (85.25, 81.76, 85.72, and 109.28) cm^3 · mol^{-1}, respectively. Thus, in the case of acetylacetone mixtures, the difference between the molar volumes of the two components might allow both components to fit into each other's structure due to which negative viscosity deviations may be observed. But in the case of THF and 1,4-dioxane, no such behavior seems to be present.

Propylene carbonate is a highly polar ($\mu = 4.94$ D at 20 °C) aprotic solvent. It shows higher dipole–dipole interactions which are responsible for self-association of

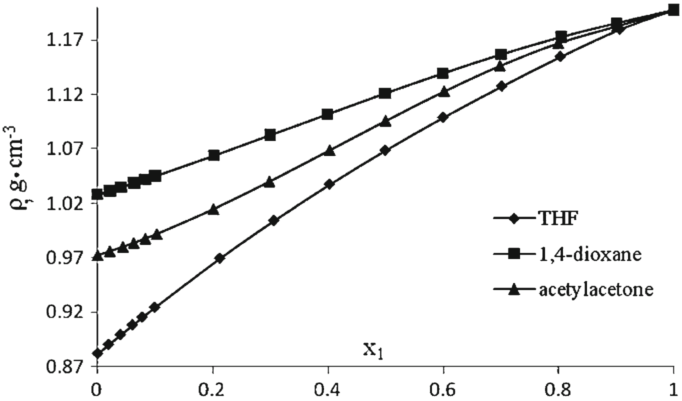

Fig. 1 Densities (ρ) for binary mixtures of propylene carbonate with THF, 1,4-dioxane, and acetylacetone mixtures at 298.15 K

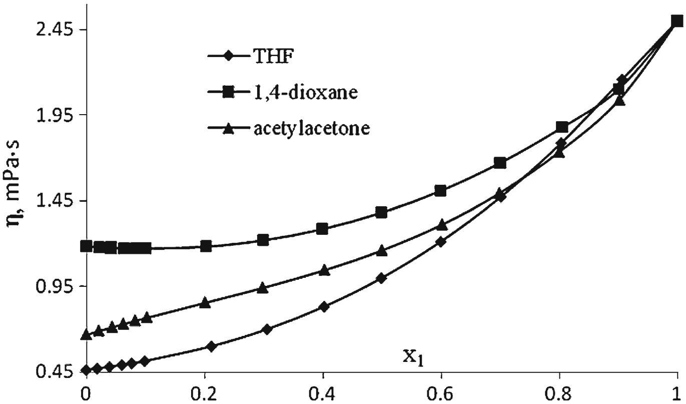

Fig. 2 Viscosities (η) for binary mixtures of propylene carbonate with THF, 1,4-dioxane, and acetylacetone mixtures at 298.15 K

propylene carbonate molecules. In the case of the propylene carbonate + 1,4-dioxane system, the poor unlike dipole–dipole interactions between CO groups of the propylene carbonate and the oxygen of 1,4-dioxane, and the capability of 1,4-dioxane molecules to break the polymer chains of propylene carbonate molecules may be the probable reason behind the interactions. The same kind of interactions is also expected in the case of THF mixtures. A similar behavior was reported by Geisler and co-workers [20] for binary mixtures of the DMSO and 1,4-dioxane system.

Acetylacetone is capable of showing keto-enol tautomerism and propylene carbonate has one carbonyl group. Hence, in the case of acetylacetone mixtures, a small amount of hydrogen bonding seems to be possible.

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