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Dr. Peeyush Ranjan
 Department of Physics,
 Chaudhary Charan Singh
 University Meerut, Uttar
 Pradesh, India

Volumetric and Viscometric properties Propionic acid in equimolar mixture of (N, N-dimethyl formamide and methanol/ethanol/1-propanol) at 303.15, 313.15, and 323.15

Dr. Peeyush Ranjan

Abstract

The densities, ρ , and viscosities, η , of mixtures of propanoic acid with equimolar mixtures of N,N-dimethyl formamide + methanol/ethanol/1-propanol, over the entire composition range of propanoic acid and including the pure liquids, have been measured at the temperatures T/K = 303.15, 313.15, and 323.15. From this experimental data, the excess molar volume, V_{EmVmE} , deviation in viscosity, $\Delta\eta$, and excess Gibbs energy of activation of viscous flow, ΔG^*E , have been determined at all three temperatures. The influence of temperature on these mixtures has been studied in terms of molecular interactions. The calculated deviation and excess parameters have been fitted to a Redlich–Kister type polynomial and the corresponding standard deviations were also evaluated. Negative values of V_{EmVmE} and positive values of $\Delta\eta$ and ΔG^*E are observed at all temperatures over the entire composition range in the mixtures studied. The observed negative and positive values of various excess and deviation parameters are attributed to the existence of strong interactions, like dipole–dipole interactions, H-bonding between the carbonyl group of acid molecules and hydroxyl group of alcohol groups, geometrical fitting of smaller molecules into the voids created by larger molecules in the liquid mixtures. The strength of these interactions in the mixtures was found to decrease with the rise in temperature and increase with an increase of chain length of the alcohols. The derived partial molar volumes and excess partial molar volumes also support the V_{EmVmE} results.

Keywords: densities, viscosities, dipole–dipole interactions and carbonyl group

1. Introduction

Alkanols are of interesting simple examples of biological and industrial important amphiphilic materials ^[1]. Alkanol are polar molecules and self-associated through hydrogen bonding of their hydroxyl groups ^[2]. Methanol also known as methyl alcohol or wood alcohol with chemical formula CH_3OH . The melting and boiling points are $-97^\circ C$ and $64.7^\circ C$ respectively. It is the simplest alcohol, and is a light, volatile, colourless, flammable liquid with a distinctive odor very similar to, but slightly sweeter than, that of ethanol (drinking alcohol). At room temperature, it is a polar liquid, and is used as an antifreeze, solvent, fuel, and as a denaturant for ethanol. Methanol is used on a limited basis to fuel internal combustion engines. Ethanol, also called ethyl alcohol, pure alcohol, grain alcohol, or drinking alcohol with the structural formula CH_3CH_2OH . It is a volatile, color less liquid that has a slight odor. The melting and boiling points are $-114^\circ C$ and $78.37^\circ C$ respectively. Ethanol is miscible with water and is a good general purpose solvent. The largest single use of ethanol is as a motor fuel and fuel additive Ethanol is the principal psychoactive constituent in alcoholic beverages. It is found in paints, tinctures, markers, and personal care products such as perfumes and deodorants. 1-Propanol or propan-1-ol or 1-propyl alcohol or n-propyl alcohol or n-propanol or simply propanol is a primary alcohol with the formula $CH_3CH_2CH_2OH$. It is a colourless liquid having melting and boiling points are $-126^\circ C$ and 97 to $98^\circ C$ respectively. It is formed naturally in small amounts during many fermentation processes and used as a solvent in the pharmaceutical industry mainly for resins and cellulose esters. The dipole moments of methanol, ethanol and 1-propanol are 1.70 D, 1.69 D and 1.58 D respectively. N, N-dimethyl formamide has large dipole moment 3.82 D ^[3, 4, 5, 6, 8] and in view of this dipole-dipole interactions are expected to play significant role to study the

Correspondence
Dr. Peeyush Ranjan
 Department of Physics,
 Chaudhary Charan Singh
 University Meerut, Uttar
 Pradesh, India

molecular interactions present in the liquid mixtures. The properties of propanoic acid and N, N-dimethyl formamide are discussed in earlier. In the present investigation, we reported densities (ρ) and viscosities (η) of the liquid mixtures of propanoic acid with equimolar mixture of N,N-dimethyl formamide and methanol/ethanol/1-propanol at three different temperatures; $T = 303.15, 313.15$ and 323.15 K over the entire mole fraction range of propanoic acid [9-22]. The excess and deviation properties such as excess molar volume ($E V_m$), deviation in viscosity ($\Delta\eta$) and excess Gibb's free energy of activation of viscous flow (ΔG^*E) have been calculated using experimental data of ρ and η . The variation of $E V_m$, $\Delta\eta$ and ΔG^*E with composition and temperature have been discussed in terms of molecular interactions existing among the molecules of these mixtures.

Materials and Methods

Propanoic acid (PA), N,N-dimethyl formamide (DMF), methanol (MOH), ethanol (EOH) and 1-propanol (POH) used in the present study were the AR grade products from LOBA Chemicals, India and were purified by standard

methods described in the literature [21,22]. The mass fraction purity of liquids obtained is > 0.995 . Before use, the chemicals were stored over 0.4 nm molecular sieves approximately for 72 h to remove water content and degassed. Mixtures are prepared by mass in air tight bottles. The mass measurements are performed with a METTLER TOLEDO (Switzerland make) ABB5-S/FACT digital balance with an accuracy ± 0.01 mg. Densities and viscosities of pure liquids and liquid mixtures have been determined using 5 cm³ two stem double walled Parker & Parker type pycnometer [23] and Ostwald viscometer which is calibrated as described by Subrahmanyam Naidu and Ravindra Prasad [24] using triply distilled water respectively. The reproducibility in the measured parameters density and viscosity are 3 in 105 parts and $\pm 0.2\%$ respectively. The densities and viscosities of pure liquids in this investigation at three different temperatures 303.15, 313.15, and 323.15 are compiled in Table 1 together with the literature data [8, 18, 22, 37-45] available. These results are found to be in good agreement with literature data.

Table 1: Comparison of experimental values of density (ρ) and viscosities (η) of pure liquids with literatures values

Liquid	$\rho/\text{kg m}^{-3}$		$\eta/10^{-3}\text{N.s.m}^{-2}$	
	present work	literature	present work	literature
$T = 303.15$ K				
Methanol	781.6	781.70[8] 781.96[22]	0.513	0.5143[8] 0.515 [22]
Ethanol	779.9	780.66 [8] 780.68 [22]	0.980	0.987 [8]
1-Propanol	795.1	795.65 [8] 795.48 [22]	1.716	1.725[22] 1.705[22]
N,N-dimethyl formamide	939.9	939.8 [37] 939.7 [18]	0.768	0.756[37] 0.752 [38]
Propanoic acid	982.1	982.0 [39]	0.950	0.9498 [39]
$T = 313.15$ K				
Methanol	772.9	772.36 [8] 772.21[22]	0.446	0.448 [8] 0.452 [22]
Ethanol	771.1	772.12[8] 772.13 [22]	0.812	0.823 [8] 0.814 [22]
1-Propanol	787.1	788.69 [8] 787.37 [22]	1.315	1.314 [8] 1.336 [22]
N,N-dimethyl formamide	932.1	930.2 [37] 929.8 [40]	0.678	0.673 [37] 0.664 [38]
Propanoic acid	973.4	971.0[39]	0.848	0.8453[39]
$T = 323.15$ K				
Methanol	764.8	762.7 [41]	0.405	0.395 [41]
Ethanol	766.4	766.1 [41]	0.741	0.7493 [41]
1-Propanol	778.7	779.5 [42]	0.999	1.1004 [42] 1.1208 [43]
N,N-dimethyl formamide	921.6	920.1 [41] 921.14 [44]	0.593	0.614 [41] 0.6128 [45]
Propanoic acid	961.4	959.5 [39]	0.706	0.8102 [39]

Results and Discussion

The experimental values of ρ and η are reported in Tables 2, for (DMF+MOH)+PA, (DMF+EOH)+PA and (DMF+POH)+PA mixtures at different temperatures over the entire mole fraction, x of PA. It has been observed that the density varies monotonically with the concentration of propanoic acid at all temperatures but the viscosity changes non-linearly showing maxima at about $x \approx 0.6$ in the PA rich region as all investigated temperatures in all the systems. The observed maxima (peak) have found to increase with increasing chain length of alcohols. Such deviations may be attributed to specific interactions arising from the formation of complexes among the mixing molecules. This type of behaviour could be attributed to complex formation. The variation of excess molar volume in the present investigation is negative over the entire mole fraction range^[30]. The observed negative $E V_m$ and positive $\Delta\eta$ and ΔG^*E values reveal that strong interactions exist in the unlike molecules of the mixtures. The factors that are mainly responsible for the expansion of molar volume i.e. positive values of $E V_m$ are: (i) Breaking one or both of the components in a solution i.e. loss of dipolar association between the molecules (dispersion forces). (ii) The geometry of molecular structures which does not favour the

fitting of molecules of one component into the voids created by the molecules of other component. (iii) Steric hindrance of the molecules. The negative values of $E V_m$ are due to strong specific interactions such as (iv) association of molecules through the formation of hydrogen bond (or) association due to dipole-dipole interactions (v) accommodation of molecules due to considerable differences in molar volumes. The presence of strong interaction is due to the hydrogen bonding (O...H-O-) between carbonyl group (-C=O) of amide molecules and hydroxyl group (-OH) of alcohol groups. When third component PA is added to the equimolar mixture the existing hydrogen bond between amide and alcohol groups is broken and a new hydrogen bond is formed between unlike molecules of acid and alcohol groups. Moreover all the components of the liquid mixtures studied are polar in nature having dipole moments of methanol $\mu = 1.70$ D, ethanol $\mu = 1.69$ D, 1-propanol $\mu = 1.58$ D, N, N-dimethyl formamide $\mu = 3.82$ and propanoic acid $\mu = 1.75$. Therefore, dipole-dipole interactions are also present in the liquid mixtures investigated. In addition to these, physical interaction such as geometrical fitting of smaller molecules into the voids created by the larger molecules is also favourable for the present systems studied.

Table 2: Experimental values of densities (ρ) and viscosities (η) with mole fraction of PA, X for (DMF + MOH) + PA binary system at 303.15, 313.15 and 323.15 K

x	T= 303.15 K		T= 313.15 K		T= 323.15 K	
	$\rho/\text{kg}\cdot\text{m}^{-3}$	$\eta/10^{-3}\text{N}\cdot\text{s}\cdot\text{m}^{-2}$	$\rho/\text{kg}\cdot\text{m}^{-3}$	$\eta/10^{-3}\text{N}\cdot\text{s}\cdot\text{m}^{-2}$	$\rho/\text{kg}\cdot\text{m}^{-3}$	$\eta/10^{-3}\text{N}\cdot\text{s}\cdot\text{m}^{-2}$
0.0000	892.2	0.674	882.8	0.580	878.3	0.503
0.1142	906.3	0.758	895.9	0.653	887.0	0.550
0.2189	919.3	0.868	908.2	0.729	898.4	0.610
0.324	931.5	1.001	920.4	0.837	911.4	0.696
0.4218	942.2	1.130	931.0	0.940	922.0	0.778
0.5248	952.7	1.267	942.1	1.041	932.4	0.875
0.6225	961.5	1.341	951.6	1.120	940.9	0.919
0.7209	969.4	1.341	960.0	1.115	948.6	0.925
0.8156	976.1	1.273	966.8	1.060	955.0	0.869
0.9074	980.1	1.098	971.1	0.950	958.9	0.767
1.0000	982.1	0.950	973.4	0.848	961.4	0.706

It was observed that, as the temperature of the systems increases the negative excess molar volumes are found to decrease indicating the decrease of interaction between the unlike molecules but the interaction increases from MOH to POH. As the alkanol chain length increases, decreasing the concentration of -OH groups in higher alkanols and thereby lowers dipole moment in higher alkanols which causes weaker interactions. These are the expected results, which

we have observed that the strength of interaction increases as we move from MOH to POH. This is due to the predomination of physical interaction over chemical interaction between the unlike molecules, in other words, geometrical fitting of smaller molecules into the voids created by the larger molecules between the components of molecules of the liquid mixtures. Such similar studies were reported by Harsha Kumar *et al*^[46]. The strength of

interactions in the mixtures follows the order (DMF + MOH) + PA < (DMF + EOH) + PA < (DMF + POH) + PA. The above discussion is also supported by the observed positive values of $\Delta\eta$ and ΔG^*E . Generally, negative values of $\Delta\eta$ indicate the presence of dispersion forces or mutual loss of specific interactions in like molecules operating in the systems arising due to weak intermolecular interactions and positive values of deviation in viscosity indicate the presence of strong interactions [25, 26]. The sign and magnitude of $\Delta\eta$ depend on the combined effect of factors such as molecular size, shape, and intermolecular forces. From the above observations it has

been clear that as the temperature increases the interaction between unlike molecules decreases. The deviation/excess properties have been fitted to Redlich-Kister type polynomial [29] equation (3.28). The coefficients A_i (A_0 to A_4) are determined using the least square method. The corresponding standard deviations (Δ) EY Δ are calculated using the relation 3.29. The coefficients, A_i and the standard deviations, σ of all the liquid mixtures have been presented in Table 3. The experimental viscosity data of all of these liquid mixtures have been correlated with four viscosity models, those of [30, 31, 32, 34]. The Katti and Chaudhri model was found to be in good agreement with the experimental values.

Table 3: Coefficients A_i of Redlich-Kister type polynomial equation (3.28) and the corresponding standard deviations, σ (3.29) of all the systems under investigation

property	A_0	A_1	A_2	A_3	A_4	σ
(DMF+MOH)+PA						
$T = 303.15$ K						
$V_m^E / 10^{-5} m^3 \cdot mol^{-1}$	-0.212	0.152	-0.112	0.085	0.067	0.001
$\Delta\eta / 10^{-3} N \cdot s \cdot m^{-2}$	1.684	-1.834	0.075	1.194	-0.998	0.007
$\Delta G^{*E} / kJ \cdot mol^{-1}$	4347.49	-3147.14	125.71	1380.25	-1897.60	15.47
$T = 313.15$ K						
$V_m^E / 10^{-5} m^3 \cdot mol^{-1}$	-0.167	0.221	-0.142	0.024	0.116	0.001
$\Delta\eta / 10^{-3} N \cdot s \cdot m^{-2}$	1.237	-1.415	-0.137	1.095	-0.436	0.005
$\Delta G^{*E} / kJ \cdot mol^{-1}$	3946.00	-2985.68	-630.10	1995.54	-626.08	12.42
$T = 323.15$ K						
$V_m^E / 10^{-5} m^3 \cdot mol^{-1}$	-0.145	0.237	0.065	0.208	0.128	0.002
$\Delta\eta / 10^{-3} N \cdot s \cdot m^{-2}$	0.981	-1.225	-0.153	1.118	0.670	0.004
$\Delta G^{*E} / kJ \cdot mol^{-1}$	3830.37	-3353.06	-668.67	2795.34	-1949.20	15.35
(DMF+EOH)+PA						
$T = 303.15$ K						
$V_m^E / 10^{-5} m^3 \cdot mol^{-1}$	-0.234	0.205	-0.271	0.029	0.202	0.001
$\Delta\eta / 10^{-3} N \cdot s \cdot m^{-2}$	1.880	-1.507	-0.044	0.479	-0.436	0.011
$\Delta G^{*E} / kJ \cdot mol^{-1}$	4413.56	-2421.62	141.39	-78.65	-611.31	20.74
$T = 313.15$ K						
$V_m^E / 10^{-5} m^3 \cdot mol^{-1}$	-0.202	0.169	-0.276	0.134	0.344	0.001
$\Delta\eta / 10^{-3} N \cdot s \cdot m^{-2}$	1.410	-1.104	-0.003	0.560	-0.778	0.004
$\Delta G^{*E} / kJ \cdot mol^{-1}$	4042.90	-2085.25	346.74	442.86	-2174.14	12.54

The partial molar volumes, 1 mV of component 1 (propanoic acid) and, 2 mV of component 2 (N, N-dimethyl formamide + methanol/ethanol/1-propanol) in the mixtures over the entire composition range are calculated by using the relations 3.30 to 3.33. The excess partial molar volumes, 1 E mV and, 2 E mV are calculated using the relations 3.34 and 3.35. From the above table, the values of 1 mV and 2 mV for both the components in the mixtures are lesser than their respective molar volumes in the pure state. The variation of excess partial molar volumes, 1 E mV (PA), 2 E mV (Equimolar mixture DMF+MOH/EOH/POH) with mole fraction of PA for (DMF+MOH)+PA, (DMF+EOH)+PA and (DMF+POH)+PA mixtures. This suggest that there is a contraction of volume occurs in the binary mixtures, indicating the presence of strong interactions between the unlike molecules of the mixtures as observed in E Vm. The partial molar volumes, 1 mV Δ , 2 mV Δ and excess partial molar volumes 1 E mV Δ 2 E mV Δ of the components at infinite dilution are obtained using the equations 3.36 and 3.37. From the above table these values are negative concluded that strong interactions exist in the unlike molecules of the components. The magnitude

of excess partial molar volumes at infinite dilution also follow the order (DMF+POH)+PA > (DMF+EOH)+PA > (DMF+MOH)+PA which supports the trends of E Vm values very well those observed in these mixtures. The dynamic viscosities of the investigated liquid mixtures have been calculated using the [35, 36, 37, 38] relations. The theoretical values of viscosity of the (DMF+MOH)+PA, (DMF+EOH)+PA and (DMF+POH)+PA liquid mixtures calculated using the above theories are compiled. Table 4 presents the values of the interaction parameters along with the standard deviations, σ . The terms G12, H12, Wvis and $\Delta 12$ are adjustable parameters representing the binary interactions. The variation of these parameters with composition follows the order (DMF+POH)+PA > (DMF+EOH)+PA > (DMF+MOH)+PA at constant temperature. Further the interaction parameter values are found to decrease with an increase in temperature of all the systems studied. These results are in good agreement with the results derived from the excess properties. [34] reported positive values of interaction parameter corresponding to systems with negative excess molar volumes. This is in good agreement with our results. The estimated values of σ

are smaller indicating that experimental values of viscosities are well correlated by all the four viscosity models. ^[40-46].

Table 4: Various interaction parameters calculated from eqns. [(3.47)-(3.53)] and the corresponding standard deviations ($\sigma/10^{-3} \text{ N}\cdot\text{s}\cdot\text{m}^{-2}$)

T/K	G ₁₂	σ	H ₁₂	σ	W _{vis} /RT	σ	Δ_{12}	σ
(DMF+MOH)+PA								
303.15	1.682	0.109	0.002	0.131	1.677	0.107	1.740	0.108
313.15	1.446	0.079	0.001	0.097	1.448	0.076	1.504	0.078
323.15	1.277	0.071	0.001	0.082	1.292	0.069	1.335	0.071
(DMF+EOH)+PA								
303.15	1.808	0.103	0.002	0.120	1.772	0.100	1.831	0.103
313.15	1.538	0.068	0.001	0.084	1.510	0.066	1.562	0.068
323.15	1.449	0.061	0.001	0.071	1.428	0.059	1.473	0.061
(DMF+POH)+PA								
303.15	1.833	0.126	0.002	0.128	1.784	0.123	1.838	0.126
313.15	1.607	0.068	0.002	0.073	1.564	0.066	1.613	0.068
323.15	1.596	0.063	0.001	0.066	1.556	0.061	1.601	0.062

Conclusions

The densities and viscosities of liquid mixtures of propanoic acid with equimolar mixture of N, N-dimethyl formamide and methanol/ethanol/1-propanol have been measured over the entire composition range at T = 303.15, 313.15 and 323.15 K and the properties like, E mV, $\Delta\eta$, ΔG^*E , V_m , $V_{m,1}$, $V_{m,2}$ have been computed from the experimental results. The values of E_m are negative and $\Delta\eta$, ΔG^*E are positive at all temperatures studied, indicating the presence of strong interactions (hydrogen bonding O...H-O-) such as interactions between carbonyl group(-C=O) of amide molecules and hydroxyl group(-OH) of alcohol groups and also the intermolecular interactions between carbonyl group(-C=O) of acid molecules and hydroxyl group(-OH) of alcohols, dipole-dipole interactions and geometrical fitting of smaller molecules into the voids created by the larger molecules in the investigated systems. The strength of interactions follows the order (DMF+MOH) +PA < (DMF+EOH) +PA < (DMF+POH) +PA. The strength of interactions is also studied with the variation of temperature. The experimental viscosity values are compared with the viscosity values obtained from different empirical relations and these are in good agreement with the experimental values.

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