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DENSITY AND VISCOSITY STUDIES OF PARACETAMOL IN ETHANOL + WATER SYSTEM AT 301.5 K

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ABSTRACT

Density and Viscosity of drug Paracetamol (PAM) in various aqueous mixtures of ethanol have been determined. These results are further extended for solutes like electrolyte NaCl and non-electrolyte sucrose in the presence of this drug. The density and viscosity data have been analyzed for the evaluation of partial molar volume, molar excess volume, excess viscosity and A & B viscosity coefficients using Jones-Dole equation.

It can be inferred from these studies that this drug acts as a structure-making compound due to hydrophobic hydration of drug molecules. B-coefficients values are found to be positive thereby showing drug solvent interactions. Furthermore these results are correlated to understand the solution behavior of drug.

Keywords: Paracetamol, ethanol+ water System, Excess parameters

INTRODUCTION

The thermophysical properties of liquid systems like density and viscosity are strictly related to the molecular interactions taking place in the system[1].These interactions decides the drug actions i.e. drug reaching to the blood stream its extent of distribution, its binding to receptors and producing physiological actions[2]. The interactions are of different types such as ionic or covalent, charge transfer, hydrogen bonding, ion-dipole and hydrophobic interactions. There are various papers appeared recently which use viscometric method to access thermodynamic parameters of biological molecule and interpreteted the solute-solvent interactions[3-5]. Therefore we decided to study the density and viscometric properties of paracetamol in mixed solvent system.

EXPERIMENTAL

Materials

The binary solvent selected for the study was ethanol + water. Commercial ethyl alcohol is refluxed with CaO for six to eight hours and distilled. Double distilled water is used for preparation of solution mixture. The distillation of water was carried out using a pinch of $KMnO_4$ & KOH in glass quick fit apparatus. The density and viscosity of water and ethanol are measured at 298.15 K and 303.15 K and compared with literature values.

Apparatus and procedure

Densities of liquids and various solutions were measured at 301.5K by using specific gravity bottle of 10 cm^3 capacity. A single pan electronic balance [*Sansui*; model KD-UBED of capacity 120 gm and with a precision of 0.0001 gm] was used for weighing purpose. The weighing was repeated thrice to ensure the accuracy in weights with a little interval of time. The reproducibility of the result was close to hundred percent.

Viscosity measurements were carried out using Ostwald's viscometer with precision ± 0.1 %. The viscometer was clamped vertically in a thermostatically controlled water-bath, whose temperature was maintained constant at 301.5K (± 0.02°C). A fixed volume (10ml) of the solution was delivered into the viscometer.
The viscometer was kept for 30 minutes in the thermostatically controlled water-bath to achieve constant temperature. The experimental measurements of flow time of the solution between two points on the viscometer were performed at least three times for each solution and the average results were noted.

Table-1: Density ρ (g cm^{-3}) **of PAM in binary system at 301.5 K**.

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	0.9960	0.9994	1.0024	1.0054	1.0082
40	0.9721	0.9748	0.9780	0.9810	0.9840
60	0.9457	0.9487	0.9516	0.9545	0.9578
80	0.9061	0.9092	0.9121	0.9151	0.9181
100	0.8142	0.8170	0.8204	0.8229	0.8264

ρ of PAM +0.01 M NaCl

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	0.9968	0.9997	1.0028	1.0059	1.0089
40	0.9726	0.9755	0.9786	0.9816	0.9843
60	0.9460	0.9490	0.9521	0.9553	0.9583
80	0.9070	0.9099	0.9127	0.9157	0.9187
100	0.8147	0.8177	0.8207	0.8239	0.8268

ρ of PAM +0.01 M Sucrose

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	0.9996	1.0028	1.0055	1.0085	1.0116
40	0.9754	0.9785	0.9811	0.9841	0.9874
60	0.9491	0.9521	0.9550	0.9584	0.9612
80	0.9098	0.9129	0.9155	0.9186	0.9217
100	0.8173	0.8204	0.8236	0.8267	0.8295

Table-2: Viscosity η (m Pa. s) **of PAM in binary system at 301.5 K**.

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	1.6786	1.6812	1.6916	1.6958	1.7055
40	2.2650	2.2689	2.2736	2.2862	2.2946
60	2.4087	2.4169	2.4389	2.4506	2.4685
80	1.9528	1.9675	1.9794	1.9905	2.0317
100	1.1286	1.1394	1.1462	1.1579	1.1741

η of PAM +0.01 M NaCl

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	1.6899	1.6906	1.7102	1.7189	1.7205
40	2.2839	2.2922	2.3086	2.3287	2.3307
60	2.4276	2.4352	2.4485	2.4709	2.4815
80	1.9651	1.9781	1.9876	2.0754	2.2299
100	1.2043	1.2243	1.2288	1.2399	1.2412

η of PAM +0.01 M Sucrose

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	1.7134	1.7183	1.7285	1.7478	1.7496
40	2.3266	2.3318	2.3392	2.3563	2.3734
60	2.4952	2.4991	2.5064	2.5217	2.5466
80	2.0885	2.0966	2.1670	2.3082	2.3249
100	1.3283	1.3355	1.3461	1.3461	1.3528

RESULTS AND DISCUSSION

Paracetamol is chemically 4-hydroxyacetanilide. It is white; odorless, crystalline powder. The densities and viscosities of ethanol- water binary mix from 20 % to 100 % range are measured (table 1 and 2) and used for determination of partial molar volume. The partial molar volume Φ_v was obtained from density results using equation 1

$$\phi_V = \frac{1000}{c}\left(\frac{d_0 d}{d}\right) + \frac{M}{d} \qquad \ldots\ldots\ldots 1$$

Where *do* is the density of pure solvent & d is the density of solution, c is molar concentration, *M* is molar mass of drug.

We observed that partial molar volume increases with increase in percentage of ethanol (table3).

The density data was also used to evaluate excess molar volumes (table 3) calculated by using the relation (equation 2).

$$V^E = \left(\frac{x_1 M_1 + x_2 M_2}{\rho}\right) - \left(x_1 V_1 + x_2 V_2\right) \qquad \ldots\ldots\ldots 2$$

Where, ρ is the density of mixture, M_1, x_1, V_1 and M_2, x_2 & V_2 are the molecular weight, mole fraction and molar volumes of ethanol & water respectively.

The trend in partial molar volume and molar excess volume almost remain same when additives sucrose and NaCl is added. The magnitude of excess parameters is slightly higher when we add salt in to it. We selected sucrose, a non-electrolyte and sodium chloride, an electrolyte as additives to know the interaction between the drugs and these additives in binary solvent system.

The values of Φv and V^E show that as the concentration of PAM increases the values of Φv decreases and increases with % of ethanol. The excess molar volume over all concentration range of PAM is found to be negative. Comparing values of V^E of ethanol-water, the values of drug is more in negative direction. It further increases in negative direction with increase in concentration of PAM. The literature survey reveals that excess molar volume of binary system becomes more negative for ethanol and other non-electrolyte[6-7].

The negative values of excess molar volume suggest specific interaction between mixing compound in mixture. It indicates intermolecular hydrogen bonding and also the interstitial accommodation of mixing components because of difference in molar volume.

Viscosity is found to maximum at around 50% (V/V) in aqueous mixtures of alcohol. It seems that some kind of structural organization of water surrounding the hydrocarbon chain of alcohol is the most likely explanation of the observed dependence of viscosity on solvent composition.

The measured values of viscosities of liquid mixtures and those of pure components were used to calculate the excess viscosity η^E(table 4) in the liquid mixtures using the formula (equation 3)

$$\eta^E = \eta mix - (x_1\eta_1 - x_2\eta_2) \qquad \ldots\ldots\ldots 3$$

Where, ηmix, η_1 & η_2 are the viscosities of liquid mixtures, component 1 & 2 respectively and x_1 & x_2 are the mole fractions of component 1 & 2 respectively.

The trend of excess viscosity shows that these values are maximum in presence of additive sucrose where as minimum in absence of any additive. This shows that excess viscosity increases with addition of PAM to ethanol- water system. It further increases in presence of additives but the increase is more pronounced when non-electrolyte sucrose is present compared to electrolyte sodium chloride.

B coefficient values are calculated using Jones-Dole equation -

$$\eta_r = 1 + A\sqrt{c} + B \qquad \ldots\ldots\ldots 4$$

The values of A & B are determined (table5) from the intercept & slope of the lines of plots of $(\eta/\eta_o - 1)$ verses $\sqrt{c}$.

We observed positive values for all the systems. The B coefficient for PAM in absence of additives is less and in presence of non-electrolyte sucrose is more. The increase in value of B with increase in percentage of alcohol indicates the structural organization of solution from water to alcohol. The B coefficient of PAM solution reflects the net structural effects of polar groups and hydrophobic benzene ring.

Table 3a- Φ_v in $cm^3\ mol^{-1}$ and V^E in $cm^3\ mol^{-1}$ of PAM in binary mixture at 301.5 K

V/V %	0.02 M (PAM)		0.04 M (PAM)		0.06 M (PAM)		0.08 M (PAM)		0.10 M (PAM)	
Et- OH	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E
20	151.3805	-0.9007	150.3522	-0.9690	149.4536	-1.0289	148.5630	-1.0884	147.7390	-1.1437
40	155.0115	-1.5821	154.1540	-1.6469	153.1469	-1.7233	152.2117	-1.7945	151.2849	-1.8653
60	159.3406	-2.5432	158.3344	-2.6330	157.3708	-2.7192	156.4160	-2.8049	155.3401	-2.9018
80	166.3349	-3.6156	165.2026	-3.7414	164.1538	-3.8584	163.0792	-3.9786	162.0152	-4.0980
100	184.9739	-2.2813	183.7082	-2.4749	182.1887	-2.7082	181.0834	-2.8786	179.5527	-3.1153

Table 3b- Φ_v in $cm^3\ mol^{-1}$ and V^E in $cm^3\ mol^{-1}$ of PAM +0.01 M NaCl in binary mixture at 301.5 K

V/V %	0.02 M (PAM)		0.04 M (PAM)		0.06 M (PAM)		0.08 M (PAM)		0.10 M (PAM)	
Et- OH	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E
20	58.5806	-0.9173	58.6388	-0.9754	58.2536	-1.0376	58.0683	-1.0996	57.8843	-1.1596
40	60.0555	-1.5944	60.0920	-1.6646	59.6814	-1.7388	59.4991	-1.8103	59.3540	-1.8733
60	61.7563	-2.5525	61.7818	-2.6424	61.3479	-2.7350	61.1233	-2.8306	60.9512	-2.9181
80	64.3683	-3.6529	64.4386	-3.7708	63.9877	-3.8838	63.7782	-4.0043	63.5701	-4.1240
100	71.6879	-2.3164	71.7047	-2.5244	71.1815	-2.7296	70.8448	-2.9501	70.6480	-3.1441

Table 3c- Φ_v in $cm^3\ mol^{-1}$ and V^E in $cm^3\ mol^{-1}$ of PAM +0.01 M sucrose in binary mixture at 301.5 K

V/V %	0.02 M (PAM)		0.04 M (PAM)		0.06 M (PAM)		0.08 M (PAM)		0.10 M (PAM)	
Et- OH	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E
20	340.9047	-0.9007	339.8888	-0.9753	339.0807	-1.0962	338.0751	-1.1572	336.9420	-1.2207
40	349.4391	-1.5821	348.1929	-1.6635	347.4869	-1.8029	346.4309	-1.8756	345.1715	-1.9558
60	359.0505	-2.5432	357.9232	-2.6473	356.8402	-2.8266	355.3926	-2.9299	354.5467	-3.0123
80	374.3780	-3.6156	373.1119	-3.7685	372.1790	-4.0022	370.8871	-4.1282	369.6042	-4.2535
100	416.8638	-2.2813	415.1422	-2.4985	413.6367	-2.9350	411.7914	-3.1494	410.7557	-3.3374

Table- 4.-Excess Viscosities (η^E) : - (mPas)

Ethanol-water + PAM

V/V % Et- OH	0.02 M	0.04 M	0.06 M	0.08 M	0.10 M
20	0.9613	0.9639	0.9743	0.9785	0.9882
40	1.5230	1.5269	1.5316	1.5442	1.5526
60	1.6303	1.6385	1.6605	1.6722	1.6901
80	1.1154	1.1301	1.1420	1.1531	1.1943
100	0.1790	0.1898	0.1966	0.2083	0.2245

Ethanol-water + PAM + 0.01 M Sucrose

V/V % Et- OH	0.02 M	0.04 M	0.06 M	0.08 M	0.10 M
20	0.9961	1.0010	1.0112	1.0305	1.0323
40	1.5846	1.5898	1.5972	1.6143	1.6314
60	1.7168	1.7207	1.7280	1.7433	1.7682
80	1.2511	1.2592	1.3296	1.4708	1.4875
100	0.3787	0.3859	0.3965	0.3965	0.4032

Ethanol-water + PAM + 0.01 M NaCl

V/V % Et- OH	0.02 M	0.04 M	0.06 M	0.08 M	0.10 M
20	0.9726	0.9733	0.9929	1.0016	1.0032
40	1.5419	1.5502	1.5666	1.5867	1.5887
60	1.6492	1.6568	1.6701	1.6925	1.7031
80	1.1277	1.1407	1.1502	1.2380	1.3925
100	0.2547	0.2747	0.2792	0.2903	0.2916

Table -5:-A and B coefficient values:-

V/V % Et- OH	PAM		PAM +0.01 Sucrose		PAM +0.01 NaCl	
	A	**B**	**A**	**B**	**A**	**B**
20	0.3236	0.0274	2.1309	0.0741	0.9155	0.0404
40	0.4373	0.0310	2.1746	0.0772	1.1091	0.0447
60	0.4588	0.0368	2.3833	0.0864	1.0139	0.0527
80	0.5431	0.0417	2.6188	0.1033	1.2662	0.0656
100	0.7249	0.0503	2.6873	0.1094	1.6994	0.0777

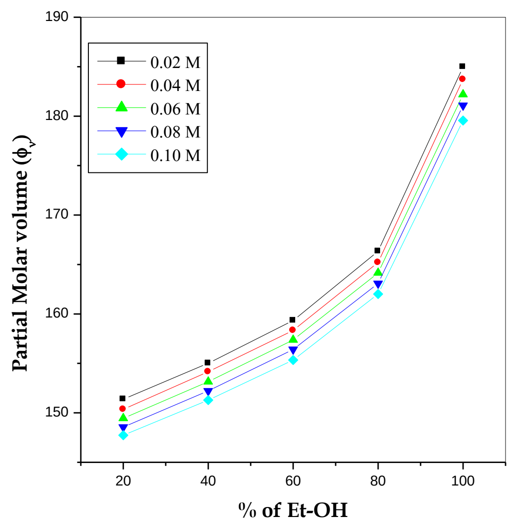

Fig. 1-Variation of Φ_v with % ethanol

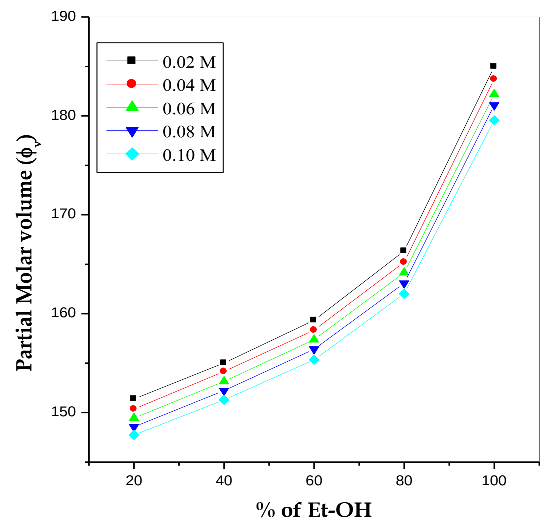

Fig. 2-Variation of V^E with % ethanol

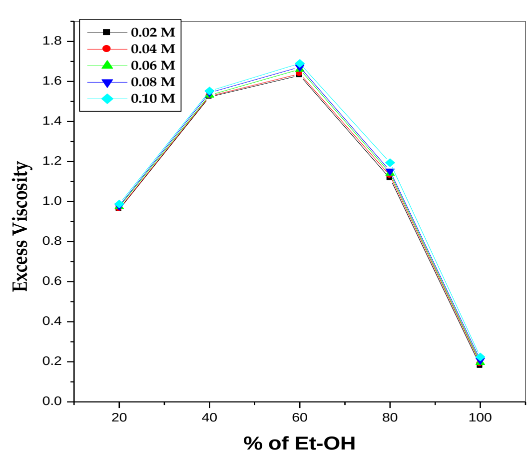

Fig.3-Variation of η^E with % ethanol

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