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DENSITY, VISCOSITY AND ACTIVATION PARAMETERS OF VISCOUS FLOW FOR CETRIMIDE IN ETHANOL + WATER SYSTEM AT 301.5 K

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ABSTRACT

Density and Viscosity of drug Cetrimide (CMD) 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 analysed for the evaluation of partial molar volume, molar excess volume, Gibbs free energy of viscous flow, excess viscosity and A and 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: Cetrimide, Partial molar volume, Gibbs free energy of viscous flow, Excess parameters.

INTRODUCTION

A systematic knowledge of solution behavior of drugs can be of great importance in order to understand their physiological action[1] The thermodynamic properties are the convenient parameter for interpreting solute-solvent interactions in the solution phase, which ultimately explain the excess properties using different interaction parameters.Most of the drugs are organic molecules with both hydrophobic and hydrophilic groups. These molecules often contain certain groups, which are responsible for their acidic, basic or amphoteric properties. Pharmacological properties[2,3] of drugs are highly dependent on the solution behavior.In the present communication an attempt has been made to study density and viscosity measurements of Cetrimide in aqueous ethanol to investigate various types of interactions.

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[4]. Double distilled water is used for preparation of solution mixture. The distillation of water was carried out using a pinch of KMnO4 & 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.

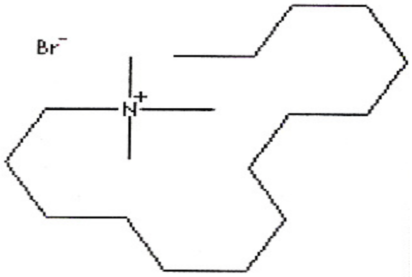
Apparatus and procedure

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 experiments of measurements of flow time of the solution between the two points on the viscometer were performed at least three times for each solution and the average results were noted.

RESULTS AND DISCUSSION



Structure of Cetrimide

Cetrimide is chemically Alkyl tri-methyl ammonium bromide. It is white crystalline powder, free flowing hygroscopic powder. It has faint characteristic odour and bitter soapy taste. It is slightly soluble in water.

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.

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

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

Cetrimide is a salt of alkyl ammonium halide which has bulkier positive ion and negative bromine ion. It is observed that the Φv values decreases with concentration of CMD and increases with increase in percentage of alcohol (table3).

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

$$V^E = \left(\frac{x_1 M_1 + x_2 M_2}{\rho}\right) - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2} \tag{2}$$

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 excess molar volumes calculated for CMD in absence and in presence of additives are negative (Fig 2). We observed that V^E changes parabolically in negative direction with % of ethanol. Over all range of concentration it was observed that V^E values are less in absence of additives and slightly high when NaCl is used. But in presence of sucrose these values are higher in negative direction. This indicates more solubility of it in the mixture. The parabolic shape of V^E against % ethanol is characterized by well defined minima which indicate the presence of complex form between mixing components of solution.

The partial molar volume of mixture containing CMD with and without additives is shown in table 3. These values are positive and changes in parabolic way with the % of ethanol (Fig.1). The higher values of excess viscosities of non-electrolyte in binary system may be due to the presence of larger alkyl chain of CMD.

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),

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

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 hydrocarbon residue of CMD in alcohol results in a considerable amount of hydrophobic hydration. However the further decrease of excess viscosity with increase in percentage alcohol may result because these hydrophobic groups exerts there effect predominantly with increase in alcohol percent (Fig 3). It appears that above 60 % v/v alcohol concentration a loss of hydrophobic hydration takes place which leads to decrease in excess viscosity[5].

The excess Gibb's free energy of activation for viscous flow, ΔG^E was calculated (table 5) for all the system under study using the equation[6,7]:

$$\Delta G^{*E} = RT \{\ln(\eta V/\eta_2V_2)-x_1 \ln (\eta_1V_1/\eta_2V_2)\} \quad (4)$$

Table-1: Density ρ (g cm^{-3}) of CMD 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	0.9998	1.0066	1.0134	1.0201	1.0266
40	0.9757	0.9824	0.9891	0.9956	1.0024
60	0.9492	0.9560	0.9624	0.9694	0.9760
80	0.9097	0.9169	0.9234	0.9302	0.9366
100	0.8176	0.8244	0.8313	0.8380	0.8445

ρ of CMD +0.01 M NaCl

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	1.0008	1.0071	1.0137	1.0207	1.0271
40	0.9760	0.9828	0.9897	0.9963	1.0030
60	0.9496	0.9567	0.9630	0.9697	0.9769
80	0.9105	0.9170	0.9241	0.9309	0.9370
100	0.8184	0.8253	0.8319	0.8381	0.8454

ρ of CMD +0.01 M Sucrose

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	1.0034	1.0101	1.0168	1.0236	1.0301
40	0.9791	0.9856	0.9925	0.9993	1.0058
60	0.9526	0.9591	0.9662	0.9728	0.9791
80	0.9134	0.9203	0.9267	0.9334	0.9407
100	0.8212	0.8278	0.8347	0.8411	0.8482

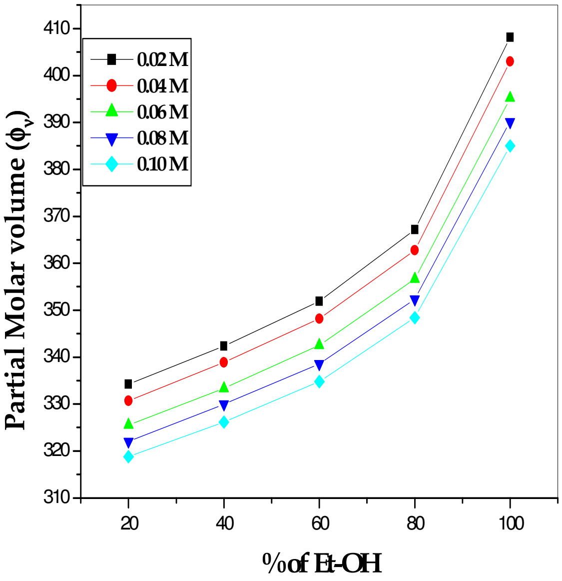


Fig.-1: Variation of Φ_v with % ethanol

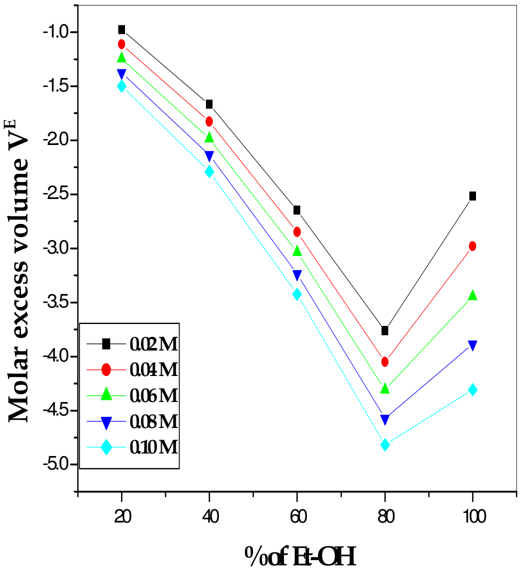


Fig.-2: Variation of V^E with % ethanol

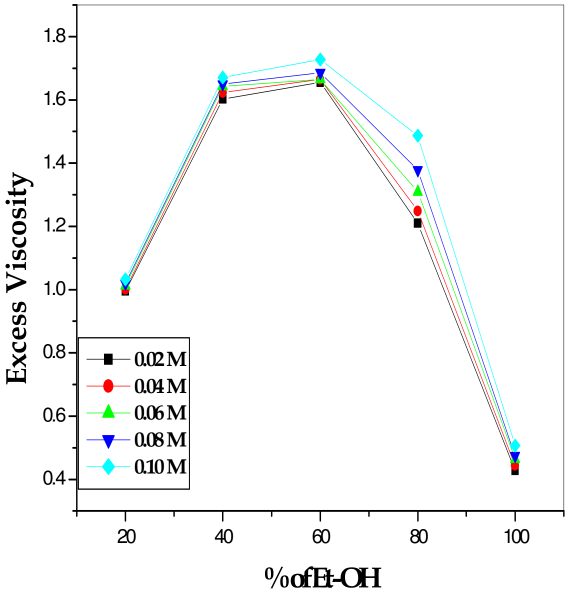


Fig.-3: Variation of η^E with % ethanol

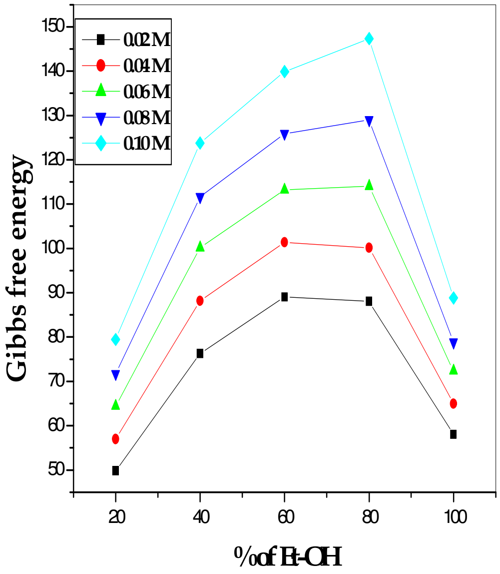


Fig.-4: Variation of ΔG^{*E} with % ethanol

Table-2: Viscosity η (m Pa. s) of CMD 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	1.7138	1.7186	1.7296	1.7368	1.7498
40	2.3449	2.3647	2.3852	2.3919	2.4126
60	2.4337	2.4433	2.4433	2.4643	2.5059
80	2.0475	2.0861	2.1464	2.2158	2.3245
100	1.3774	1.3946	1.4138	1.4239	1.4566

η of CMD +0.01 M NaCl

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	1.7344	1.7386	1.7478	1.7588	1.7686
40	2.3542	2.3871	2.3847	2.4257	2.4437
60	2.4714	2.4937	2.5021	2.5214	2.5375
80	2.0986	2.1287	2.2263	2.3382	2.3583
100	1.4017	1.4166	1.4283	1.4685	1.4877

η of CMD +0.01 M Sucrose

v/v % Et-OH	0.02 M	0.04 M	0.06 M	0.08M	0.10 M
20	1.7508	1.7846	1.7953	1.8165	1.8296
40	2.4101	2.4328	2.4443	2.4784	2.5249
60	2.5821	2.6041	2.6247	2.6476	2.6682
80	2.1875	2.3237	2.3637	2.4812	2.5029
100	1.4318	1.4499	1.4674	1.5654	1.5865

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

V/V % Et- OH	0.02 M (CMD)		0.04 M (CMD)		0.06 M (CMD)		0.08 M (CMD)		0.10 M (CMD)	
	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E
20	334.2612	-0.9770	330.6987	-1.1121	325.5738	-1.2455	322.0300	-1.3751	318.8008	-1.4992
40	342.3565	-1.6685	338.8955	-1.8276	333.3433	-1.9845	329.9654	-2.1348	326.1344	-2.2899
60	351.9199	-2.6479	348.1900	-2.8490	342.5476	-3.0357	338.5922	-3.2371	334.7944	-3.4243
80	367.1707	-3.7616	362.7960	-4.0503	356.6296	-4.3071	352.3371	-4.5718	348.3971	-4.8175
100	408.1542	-2.5162	402.9985	-2.9803	395.2026	-3.4434	390.1187	-3.8858	385.0027	-4.3083

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

V/V % Et- OH	0.02 M (CMD)		0.04 M (CMD)		0.06 M (CMD)		0.08 M (CMD)		0.10 M (CMD)	
	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E
20	58.3349	-0.9984	57.9992	-1.1234	57.6331	-1.2525	57.2212	-1.3898	56.8704	-1.5120
40	59.8586	-1.6761	59.4386	-1.8382	59.0124	-2.0010	58.6158	-2.1548	58.2303	-2.3076
60	61.5158	-2.6604	61.0403	-2.8717	60.6475	-3.0558	60.2474	-3.2474	59.7668	-3.4561
80	64.1281	-3.7953	63.7226	-4.0546	63.1920	-4.3379	62.7308	-4.6034	62.3426	-4.8359
100	71.3378	-2.5729	70.7334	-3.0448	70.1982	-3.4869	69.7208	-3.8932	69.0534	-4.3750

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

V/V % Et- OH	0.02 M (CMD)		0.04 M (CMD)		0.06 M (CMD)		0.08 M (CMD)		0.10 M (CMD)	
	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E	Φ_v	V^E
20	339.6183	-1.0539	337.4072	-1.1906	335.2246	-1.3252	332.9724	-1.4607	330.8785	-1.5882
40	348.0874	-1.7545	345.8701	-1.9120	343.4039	-2.0778	340.9724	-2.2401	338.8784	-2.3899
60	357.7360	-2.7541	355.4317	-2.9492	352.5719	-3.1624	350.3338	-3.3538	348.1944	-3.5337
80	372.9085	-3.9168	370.2450	-4.1963	367.7373	-4.4521	365.1463	-4.7155	361.9745	-5.0052
100	414.6380	-2.7704	411.4464	-3.2234	408.0591	-3.6893	405.1118	-4.1126	401.4479	-4.5816

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

V/V % Et- OH	0.02 M	0.04 M	0.06 M	0.08 M	0.10 M
			Ethanol-water + (CMD)		
20	0.9965	1.0013	1.0123	1.0195	1.0325
40	1.6029	1.6227	1.6432	1.6499	1.6706
60	1.6553	1.6649	1.6649	1.6859	1.7275
80	1.2101	1.2487	1.3090	1.3784	1.4871
100	0.4278	0.4450	0.4642	0.4743	0.5070
			Ethanol-water + (CMD) + 0.01 M Sucrose		
20	1.0335	1.0673	1.0780	1.0992	1.1123
40	1.6681	1.6908	1.7023	1.7364	1.7829
60	1.8037	1.8257	1.8463	1.8692	1.8898
80	1.3501	1.4863	1.5263	1.6438	1.6655
100	0.4822	0.5003	0.5178	0.6158	0.6369
			Ethanol-water + (CMD) + 0.01 M NaCl		
20	1.0171	1.0213	1.0305	1.0415	1.0513
40	1.6122	1.6451	1.6427	1.6837	1.7017
60	1.6930	1.7153	1.7237	1.7430	1.7591
80	1.2612	1.2913	1.3889	1.5008	1.5209
100	0.4521	0.4670	0.4787	0.5189	0.5381

Table-5: Gibbs free energy of viscous flow ΔG^{*E}(kJ mole^{-1})

V/V % Et- OH	0.02 M	0.04 M	0.06 M	0.08 M	0.10 M
			Ethanol-water + CMD		
20	49.8205	56.9725	64.4216	71.7183	79.4632
40	76.2546	88.1738	100.1813	111.6320	123.7697
60	89.0447	101.3579	113.2090	125.9009	139.9014
80	88.0297	100.1506	114.0186	128.9671	147.3278
100	58.0153	64.9408	72.3814	78.8335	88.8280
			Ethanol-water + CMD+ 0.01 M Sucrose		
20	54.8941	63.8249	71.8270	80.6010	89.0034
40	84.1806	96.6347	108.6055	121.8006	135.9764
60	100.7296	114.5176	128.2735	142.1723	155.9491
80	100.7988	119.7808	133.8930	153.4482	166.4854
100	66.9803	74.7317	82.6047	102.7416	112.2277
			Ethanol-water + CMD + 0.01 M NaCl		
20	51.2205	58.5486	66.0688	75.6477	81.4684
40	77.5489	90.0361	101.0942	117.3757	126.5310
60	91.4752	104.5489	117.1292	133.3747	142.8167
80	91.6276	103.6846	120.2316	141.6300	150.9126
100	61.2662	68.1897	74.8514	87.5429	94.2654

Table-6: A and B coefficient values

V/V % t	(CMD)		(CMD) +0.01 Sucrose		(CMD) +0.01 NaCl	
	A	B	A	B	A	B
20	2.3836	0.0727	2.9281	0.1137	2.5961	0.0864
40	2.5738	0.0808	3.2013	0.1359	2.7248	0.0962
60	2.7950	0.0912	3.3791	0.1452	3.0465	0.1062
80	3.0238	0.1007	4.3651	0.1714	3.5627	0.1257
100	3.5014	0.1192	4.4359	0.1844	3.6909	0.1341

Where, η , η1 & η2 are the viscosities of liquid mixtures, components 1 & 2 respectively, V1 & V2 are the molar volumes of components 1 & 2 respectively, x1 is the mole fraction of first component, R is the gas constant & T is the absolute temperature.

The values are positive and changes parabolically with percentage of ethanol (Fig 4). The maximum Gibbs free energy of viscous flow for ethanol-water is 75.4598 KJ mole^{-1}. The trend in maxima shows that ΔG^{*E} for NaCl is less than ΔG^{*E} for sucrose. This excess free energy increases with increase in concentration of CMD keeping concentration of additive const.

The values of ΔG^{*E} is much higher when CMD and additives are present together. The positive values of ΔG^{*E} represent the size effect of mixing components. It is considered that if ΔG^{*E} is positive there are specific interactions like hydrogen bonding which exists between molecules of mixture[8.]

B coefficient values are calculated using Jones-Dole equation -

$$\eta_r = 1 + A\sqrt{c} + B \quad (5)$$

The values of A & B are determined (table 6) 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 CMD in absence of additives is less and in presence of non-electrolyte sucrose is more. This increases with increase in percentage of alcohol which indicates the structural increases of solution from water to alcohol. The B coefficient of CMD solution reflects the net structural effects of polar groups and hydrophobic benzene ring.

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