

Apparent molar volume and coefficients of Jones-Dole equation for the evaluation of ion-solvent interactions of barium chloride in aqueous 1, 4-dioxane and ethylene glycol solutions at T= (298.15 K to 313.15) K and at atmospheric pressure

Braja B.Nanda ^{1,*}, Mamata Pradhan ², Prativa Kar ², Binita Nanda ³

¹P.G. Department of Chemistry, Vikram Deb Autonomous College, Jeypore-764001, Odisha, India

²Department of Chemistry, GIET University, Gunupur, Rayagada, Odisha – 765022, India

³Department of Chemistry, Faculty of Engg. and Technology, (ITER), Siksha 'O' Anusandhan Deemed to be University, Bhubaneswar-751030, Odisha, India

*corresponding author e-mail address: bbnanda.driems@gmail.com | Scopus ID [8431581400](https://orcid.org/0000-0001-8431-5814)

ABSTRACT

The viscosity and density of barium chloride have been measured at T= (298.15-313.15) K in aqueous solutions of 1, 4-Dioxane (1,4-DO) and Ethylene glycol (EG). Jones–Dole equation was used to analyze the viscosity data. The apparent molar volume, V_{ϕ} limiting apparent molar volume, V_{ϕ}^0 are calculated from the density data. Limiting apparent molar expansibilities, E_{ϕ}^0 have been used to describe the temperature dependency of V_{ϕ}^0 . The standard volume of transfer, ΔV_{ϕ}^0 and viscosity B -coefficient of transfer, ΔB of barium chloride from water to aqueous 1, 4-DO and EG solutions were systematizing the different types of interactions in the given solutions. The structure making and breaking capacity of solute in solutions was interpreted with the help of Helper equation. The positive value of $(\partial^2 V_{\phi}^0 / \partial T^2)_p$ suggests the structure making nature for $BaCl_2$ in given solution. It is observed that Friedman-Krishnan co-sphere model explains the increase in the transfer volume of $BaCl_2$ with an increase in 1,4-DO and EG concentrations. The activation parameters of the viscous flow of the given solutions were calculated and interpreted using transition state theory.

Keywords: Apparent molar volume; viscosity B -coefficient; Jones–Dole equation; barium chloride, ion-solvent interactions.

1. INTRODUCTION

Various chemical engineering applications require the idea of thermodynamics and transport properties of the multi component systems. These properties are also useful to know the properties of liquid solutions. The study of physico-chemical properties of barium chloride with EG+ water and 1, 4-DO +water at T=298.15 to 313.15 K is important to know the effect of the ion-solvent interactions between barium chloride and the aqueous solutions of EG and 1, 4-DO. Barium Chloride is one of the water-soluble salts of barium.

Which imparts a yellow-green coloration to a flame so used in fireworks to give a bright green color. Barium chloride is used as a raw material for producing barium nitrate, barium hydroxide and barium carbonate etc. It is widely used in papermaking, plastic industry, dyestuff, rubber, ceramics and petrochemistry etc. It is used to remove sulphate from aqueous salt solution in chlorine-alkali industry.

The EG having both hydrophobic and hydrophilic groups, Therefore, the role of EG in water mixtures is extremely important for the study of many biochemical and complex chemical processes. 1,4-DO is a versatile aprotic solvent. This water miscibility of 1,4-DO is a property which favors several industrial applications. Lewis basic oxygen atom is, able to solvate many inorganic compounds and is used in inks and adhesives applications as a solvent. It is low toxic to aquatic life and is biodegradable [1]. A mixture of non-protic and unassociated solvents such as 1,4-DO gives practical and theoretical interest. Apparent molar volume and viscosity

suggested various types of interactions occurring in solutions [2-3]. The nature of a solute in a binary solvent system is important for the study of their ionic interactions between solute-solute and solute-solvent [4-8].

The thermodynamic properties of non-polar solutes in aqueous solution explained by Frank [9] in terms of the arrangement of water molecules around the solute.

A polar solute in water reveals a tendency to aggregate the molecules and built iceberg formation known as structure promoter and which decreases the order known as structure breaker. This paper is concerned with the relation between these structural ideas and to investigate the interaction of $BaCl_2$ with aqueous solutions of EG and 1,4-DO as these are used for human beings. This paper reflects our experimental data on density and viscosity of barium chloride in EG + water and 1,4-DO + water at T=(298.15 to 313.15) K. Apparent molar volumes (V_{ϕ}) are calculated from density data at T=(298.15 K to 313.15)K and used to estimate limiting apparent molar volume (V_{ϕ}^0), standard partial molar isobaric expansivity (E_{ϕ}^0 and Helper's constant $((\partial^2 V_{\phi}^0 / \partial T^2)_p)$. The viscosity data have been analyzed using the Jones–Dole equation. The values of A and viscosity B -coefficients of Jones-Dole equation and their temperature dependency were also calculated. Further, the analysis of viscosity data was done on the basis of Feakins transition state theory[10]. The parameters were used to the various interactions occurring between the components in the solution mixtures.

2. MATERIALS AND METHODS

2.1. Materials.

1, 4-DO and EG used were of E. Merck “Extra Pure” variety with 99% purity. These were further purified by standard methods [11]. 10 to 30% (v/v) solutions of these compounds were prepared with conductivity water, which served as solvent. AnalaR grade anhydrous barium chloride was dried over anhydrous CaCl₂ in desiccators before use [12]. The sample description is presented in Table 1.

2.2. Instruments and methods.

Solutions of concentration 0.1, 0.075, 0.05 and 0.025 mol dm⁻³ were prepared by weighing the required amount of electrolyte in a weighing bottle and then dissolving it in the required amount of solvent in a 250 mL measuring flask. Rest of solutions of concentration 0.01, 0.0075, 0.005, 0.0025 and 0.001 mol/dm³ was

obtained by dilution. Electronic Anamed balance (India) was used to record the masses of solute and solvents with a precision of ± 0.0001 g. The density (ρ) of mixture solutions were measured at temperatures 298.15K to 313.15 K using a double arm Ostwald-Sprengel type pycnometer of about 25 cm³ capacity calibrated with double distilled water with ± 0.9961 g /cm³ as its ρ at T=298.15 K[13]. The uncertainty in density measurements was less than $\pm 5 \times 10^{-5}$ g cm⁻³. Viscosity was determined with Ostwald viscometer. The viscometer was calibrated using deionized water. The average triplet measurement was taken into account. The viscosity and density measurements were done in the same thermostatic water bath. The uncertainty in the viscosity was found to be less than 0.1%. The viscosity of pure water was taken to be 0.7194×10^{-3} kg m⁻¹ s⁻¹ at 35°C [14].

Table 1. Sample Description.

Chemical name	Source	Initial mass fraction purity	Purification method	Final mass fraction purity
Barium Chloride	Qualigen	0.99	Used as procured	0.99
Ethylene Glycol	Merck	0.99	Purified by standard methods[11]	0.99
1,4-Dioxane	Merck	0.99	Purified by standard methods[11]	0.99

Table 2. Density (ρ), of BaCl₂ in 10-30% 1,4-dioxane-water and ethylene glycol-water solutions respectively at T=(298.15 to 313.15) K and at atmospheric pressure.

C/ mol L ⁻¹	10 ³ ρ / (kg m ⁻³)											
	T=298.15K			T=303.15K			T=308.15K			T=313.15K		
	1,4-Dioxane+water											
	10%	20%	30%	10%	20%	30%	10%	20%	30%	10%	20%	30%
0.000	1.0004	1.0034	1.0065	0.9983	1.0010	1.0037	0.9963	0.9986	1.0010	0.9942	0.9962	0.9982
0.001	1.0005	1.0036	1.0066	0.9984	1.0011	1.0038	0.9965	0.9988	1.0011	0.9943	0.9963	0.9983
0.003	1.0007	1.0038	1.0068	0.9986	1.0013	1.0040	0.9967	0.9990	1.0013	0.9946	0.9965	0.9985
0.005	1.0011	1.0041	1.0072	0.9990	1.0017	1.0043	0.9970	0.9993	1.0016	0.9949	0.9969	0.9988
0.007	1.0014	1.0045	1.0075	0.9994	1.0020	1.0047	0.9974	0.9997	1.0020	0.9953	0.9972	0.9992
0.010	1.0018	1.0048	1.0078	0.9997	1.0023	1.0050	0.9978	1.0000	1.0023	0.9957	0.9976	0.9995
0.025	1.0039	1.0068	1.0098	1.0019	1.0044	1.0070	1.0000	1.0021	1.0043	0.9979	0.9997	1.0015
0.050	1.0075	1.0103	1.0131	1.0055	1.0079	1.0103	1.0036	1.0056	1.0077	1.0016	1.0032	1.0050
0.075	1.0112	1.0137	1.0164	1.0092	1.0114	1.0137	1.0074	1.0091	1.0111	1.0053	1.0068	1.0084
0.100	1.0148	1.0172	1.0198	1.0129	1.0149	1.0171	1.0111	1.0127	1.0145	1.0091	1.0103	1.0119
	Ethylene glycol +Water											
0.000	1.0151	1.0204	1.0319	1.0071	1.0186	1.0300	1.0054	1.0168	1.0283	1.0037	1.0151	1.0266
0.001	1.0153	1.0205	1.0320	1.0072	1.0187	1.0302	1.0056	1.0170	1.0284	1.0038	1.0153	1.0267
0.003	1.0155	1.0207	1.0322	1.0074	1.0189	1.0304	1.0058	1.0172	1.0286	1.0040	1.0155	1.0269
0.005	1.0159	1.0210	1.0325	1.0078	1.0192	1.0307	1.0061	1.0175	1.0289	1.0044	1.0158	1.0272
0.007	1.0162	1.0214	1.0329	1.0082	1.0196	1.0310	1.0065	1.0179	1.0293	1.0048	1.0162	1.0276
0.010	1.0166	1.0217	1.0332	1.0085	1.0199	1.0314	1.0069	1.0182	1.0296	1.0051	1.0165	1.0279
0.025	1.0188	1.0238	1.0351	1.0107	1.0220	1.0333	1.0091	1.0203	1.0316	1.0074	1.0186	1.0300
0.050	1.0224	1.0272	1.0384	1.0144	1.0255	1.0366	1.0128	1.0238	1.0349	1.0111	1.0221	1.0334
0.075	1.0261	1.0307	1.0417	1.0181	1.0290	1.0400	1.0166	1.0274	1.0383	1.0149	1.0257	1.0368
0.100	1.0298	1.0342	1.0450	1.0219	1.0325	1.0433	1.0203	1.0309	1.0417	1.0187	1.0292	1.0403

Table 3. Apparent molar volumes, V_{ϕ} , of BaCl₂ in 10-30% 1,4-dioxane-water and ethylene glycol-water respectively at T=(298.15 to 313.15) K and at atmospheric pressure.

10 ³ C/ mol m ⁻³	10 ⁶ V_{ϕ} /(m ³ mol ⁻¹)											
	T=298.15K			T=303.15K			T=308.15K			T=313.15K		
	1,4-Dioxane+water											
	10%	20%	30%	10%	20%	30%	10%	20%	30%	10%	20%	30%
0.001	101.2	108.0	112.8	99.9	106.9	111.6	98.6	105.8	110.5	97.3	104.7	109.3
0.003	100.9	107.7	112.4	99.6	106.6	111.3	98.3	105.5	110.2	97.0	104.4	109.1
0.005	100.6	107.4	112.0	99.3	106.3	110.9	98.0	105.2	109.8	96.7	104.1	108.7

Apparent molar volume and coefficients of Jones-Dole equation for the evaluation of ion- solvent interactions of barium chloride in aqueous 1, 4-dioxane and ethylene glycol solutions at T= (298.15 K to 313.15) K and at atmospheric pressure

$10^3 C / \text{mol m}^{-3}$	T=298.15K			T=303.15K			T=308.15K			T=313.15K		
	$10^6 V_\phi / (\text{m}^3 \text{mol}^{-1})$											
	1,4-Dioxane+water											
	10%	20%	30%	10%	20%	30%	10%	20%	30%	10%	20%	30%
0.007	100.2	107.0	111.6	98.9	105.9	110.5	97.6	104.8	109.4	96.3	103.7	108.3
0.010	99.8	106.5	111.3	98.5	105.4	110.2	97.2	104.4	109.0	95.9	103.3	107.9
0.025	99.7	106.5	111.1	98.4	105.4	110.0	97.1	104.3	108.9	95.8	103.2	107.8
0.050	99.6	106.4	111.0	98.3	105.3	109.9	97.0	104.2	108.8	95.7	103.1	107.7
0.075	99.5	106.3	110.9	98.2	105.2	109.8	96.9	104.1	108.7	95.6	103.0	107.6
0.100	99.4	106.2	110.8	98.1	105.1	109.7	96.8	104.0	108.6	95.5	102.9	107.5
Ethylene glycol +Water												
0.001	97.9	105.1	111.2	97.1	104.4	109.5	96.2	103.2	108.1	95.4	102.8	106.2
0.003	97.7	104.8	110.9	96.9	104.1	109.2	96.0	102.9	107.8	95.2	102.7	105.9
0.005	97.4	104.5	110.6	96.6	103.8	108.9	95.7	102.6	107.6	94.9	102.4	105.7
0.007	97.0	104.1	110.2	96.2	103.4	108.5	95.3	102.2	107.1	94.5	102.0	105.3
0.010	96.6	103.8	109.9	95.8	103.1	108.2	94.9	101.9	106.8	94.2	101.8	104.9
0.025	96.6	103.7	109.8	95.8	103.0	108.1	94.9	101.8	106.7	94.1	101.7	104.8
0.050	96.5	103.6	109.7	95.7	102.9	108.0	94.8	101.7	106.6	94.0	101.6	104.7
0.075	96.5	103.6	109.6	95.6	102.8	107.9	94.6	101.6	106.5	93.8	101.5	104.6
0.100	96.3	103.5	109.5	95.5	102.7	107.8	94.6	101.5	106.4	93.8	101.4	104.5

Table 4. $\eta_{sp}/c^{1/2}$ of BaCl₂ in 10-30 % (v/v) 1,4-dioxane+water and ethylene glycol + water solvents respectively at T=(298.15 to 313.15) K and at atmospheric pressure.

$10^3 C / \text{mol m}^{-3}$	$10^{-3/2} \eta_{sp} / c^{1/2} / (\text{m}^{3/2} \text{mol}^{-1/2})$											
	T=298.15K			T=303.15K			T=308.15K			T=313.15K		
	1,4-Dioxane+water											
	10%	20%	30%	10%	20%	30%	10%	20%	30%	10%	20%	30%
0.001	0.048	0.058	0.068	0.049	0.058	0.066	0.055	0.062	0.069	0.056	0.061	0.068
0.003	0.065	0.072	0.080	0.064	0.072	0.078	0.063	0.071	0.078	0.064	0.070	0.079
0.005	0.074	0.085	0.090	0.072	0.079	0.088	0.073	0.081	0.090	0.072	0.082	0.089
0.007	0.086	0.088	0.100	0.080	0.088	0.099	0.080	0.087	0.098	0.084	0.089	0.098
0.010	0.094	0.101	0.108	0.093	0.098	0.106	0.090	0.096	0.105	0.091	0.096	0.103
0.025	0.136	0.144	0.152	0.133	0.142	0.150	0.132	0.140	0.148	0.131	0.138	0.146
0.050	0.182	0.191	0.202	0.181	0.189	0.199	0.178	0.186	0.197	0.175	0.184	0.194
0.075	0.219	0.228	0.240	0.215	0.225	0.237	0.211	0.222	0.234	0.208	0.219	0.230
0.100	0.248	0.258	0.273	0.244	0.255	0.269	0.240	0.252	0.266	0.236	0.249	0.262
Ethylene glycol +Water												
0.001	0.061	0.060	0.065	0.060	0.060	0.063	0.057	0.060	0.063	0.057	0.060	0.060
0.003	0.068	0.070	0.072	0.068	0.069	0.071	0.066	0.068	0.071	0.066	0.068	0.069
0.005	0.091	0.093	0.099	0.089	0.092	0.096	0.088	0.091	0.095	0.086	0.089	0.092
0.007	0.105	0.109	0.114	0.104	0.108	0.112	0.103	0.106	0.111	0.102	0.105	0.109
0.010	0.112	0.117	0.122	0.110	0.115	0.120	0.109	0.113	0.118	0.107	0.111	0.116
0.025	0.156	0.163	0.170	0.154	0.160	0.168	0.152	0.157	0.164	0.150	0.155	0.162
0.050	0.206	0.214	0.226	0.203	0.211	0.222	0.200	0.208	0.217	0.197	0.204	0.213
0.075	0.244	0.254	0.268	0.240	0.250	0.263	0.237	0.246	0.257	0.234	0.242	0.253
0.100	0.276	0.287	0.303	0.272	0.283	0.298	0.268	0.279	0.291	0.264	0.274	0.287

Table 5. Limiting apparent molar volumes transfer, $\Delta_i V_\phi^0$ and Viscosity B coefficient of transfer, $\Delta_i B$ from water to different aqueous 1,4-dioxane and ethylene glycol solutions for BaCl₂ at T=(298.15 to 313.15) K and at atmospheric pressure.

% (v/v)	1,4-Dioxane+Water				Ethylene glycol +Water			
	$10^6 V_\phi^0 / (\text{m}^3 \text{mol}^{-1})$	$10^6 \Delta_i V_\phi^0 / (\text{m}^3 \text{mol}^{-1})$	$B / (\text{m}^3 \text{mol}^{-1})$	$\Delta_i B / (\text{m}^3 \text{mol}^{-1})$	$10^6 V_\phi^0 / (\text{m}^3 \text{mol}^{-1})$	$10^6 \Delta_i V_\phi^0 / (\text{m}^3 \text{mol}^{-1})$	$B / (\text{m}^3 \text{mol}^{-1})$	$\Delta_i B / (\text{m}^3 \text{mol}^{-1})$
T=298.15K								
0	95.5	0	0.2349	0	92.25	0	0.324	0
10	100.9	5.4	0.6939	0.459	97.7	5.45	0.756	0.4319
20	107.7	12.2	0.7271	0.492	104.8	12.55	0.793	0.4683
30	112.4	16.9	0.7648	0.53	110.9	18.65	0.837	0.5129
T = 303.15K								
0	94.15	0	0.2173	0	91.5	0	0.32	0
10	99.6	5.45	0.6843	0.467	96.8	5.3	0.746	0.4209
20	106.6	12.45	0.717	0.5	104.1	12.6	0.779	0.4542
30	111.3	17.15	0.7525	0.535	109.2	17.7	0.824	0.4997

% (v/v)	1,4-Dioxane+Water				Ethylene glycol +Water			
	$10^6 V_\phi^0 / (\text{m}^3 \text{mol}^{-1})$	$10^6 \Delta V_\phi^0 / (\text{m}^3 \text{mol}^{-1})$	$B / (\text{m}^3 \text{mol}^{-1})$	$\Delta_i B / (\text{m}^3 \text{mol}^{-1})$	$10^6 V_\phi^0 / (\text{m}^3 \text{mol}^{-1})$	$\Delta V_\phi^0 / (\text{m}^3 \text{mol}^{-1})$	$B / (\text{m}^3 \text{mol}^{-1})$	$\Delta_i B / (\text{m}^3 \text{mol}^{-1})$
T = 308.15K								
0	92.8	0	0.21	0	90.45	0	0.308	0
10	98.3	5.5	0.6727	0.463	96	5.55	0.738	0.4308
20	105.5	12.7	0.7066	0.497	102.9	12.45	0.769	0.4613
30	110.2	17.4	0.7426	0.533	107.8	17.35	0.801	0.4934
T = 313.15K								
0	91.45	0	0.2097	0	89.64	0	0.297	0
10	97	5.55	0.6613	0.452	95.2	5.56	0.726	0.4288
20	104.4	12.95	0.6982	0.489	101.9	12.26	0.753	0.456
30	109	17.55	0.7313	0.522	105.9	16.26	0.793	0.4961

Table 6. Values of experimental slope (Eq 2), S_v , and Falkenhagen coefficient, A (Eq 12) of BaCl_2 solution in 10-30% 1,4-dioxane-water and glycol-water T=298.15-313.15 K and at atmospheric pressure.

Solvent	% v/v	$-10^9 S_v (\text{m}^{9/2} \text{mol}^{-3/2})$				$10^5 A (\text{m}^3 \text{mol}^{-1})^{1/2}$			
		T= 298.15 K	T= 303.15K	T= 308.15K	T= 313.15K	T= 298.15 K	T= 303.15K	T= 308.15K	T= 313.15K
1,4-Dioxane-water	0	4.92	5.00	5.09	5.14	-0.062	-0.024	-0.012	-0.050
	10	5.54	5.54	5.53	5.53	0.029	0.028	0.027	0.027
	20	5.46	5.48	5.53	5.53	0.029	0.029	0.028	0.028
	30	5.94	5.84	5.78	5.64	0.031	0.031	0.031	0.030
Ethylene glycol-water	0	0.60	0.59	0.59	0.58	0.092	0.108	0.097	0.096
	10	4.86	4.95	5.09	5.15	0.037	0.036	0.035	0.035
	20	4.73	5.13	5.13	4.46	0.037	0.037	0.036	0.036
	30	5.06	5.13	5.22	5.29	0.038	0.037	0.038	0.036

Table 7. The values of coefficients a_0 , a_1 , and a_2 , limiting apparent molar expansibility, E_ϕ^0 and helpers constant values of for BaCl_2 in mixed solutions at T=(298.15 to 313.15) K and at atmospheric pressure.

Solvent	% v/v	$10^6 a_0 / (\text{m}^3 \text{mol}^{-1})$	$-10^6 a_1 / (\text{m}^3 \text{mol}^{-1} \text{K}^{-1})$	$10^6 a_2 / (\text{m}^3 \text{mol}^{-1} \text{K}^{-2})$	$-10^6 E_\phi^0 / (\text{m}^3 \text{mol}^{-1})$				$10^3 dE_\phi^0/dT$
					298.15 K	303.15K	308.15K	313.15K	
1,4-Dioxane-water	0	185.4	0.3313	0.0001	0.272	0.27	0.27	0.269	0.2
	10	268.8	0.8613	0.0010	0.265	0.26	0.245	0.235	2
	20	263.1	0.8213	0.0010	0.225	0.22	0.205	0.195	2
	30	270.1	0.8272	0.0010	0.23	0.22	0.21	0.2	2
Ethylene glycol-water	0	274.0	0.8213	0.0010	0.225	0.22	0.205	0.195	2
	10	76.97	0.3790	0.0004	0.164	0.16	0.157	0.153	7.2
	20	180.8	0.4960	0.0008	0.018	0.01	0.003	0.005	1.6
	30	221.9	0.4020	0.0001	0.342	0.34	0.34	0.339	0.2

Table 8. Isobaric thermal expansion coefficient (α) and solvation number (B/V_ϕ^0) of BaCl_2 in 10-30% 1,4-dioxane-water and glycol-water solutions respectively at T=(298.15 to 313.15) K and at atmospheric pressure.

Solvent	% v/v	$-10^3 \times \alpha$				B/V_ϕ^0			
		298.15 K	303.15K	308.15K	313.15K	298.15 K	303.15K	308.15K	313.15K
1,4-Dioxane-water	0	2.844	2.874	2.910	2.937	2.460	2.308	2.263	2.293
	10	2.626	2.560	2.490	2.423	6.880	6.870	6.843	6.818
	20	2.089	2.017	1.940	1.868	6.750	6.726	6.698	6.688
	30	2.046	1.977	1.910	1.835	6.800	6.761	6.739	6.709
Ethylene glycol-water	0	2.439	2.350	2.270	2.175	3.510	3.548	3.400	3.317
	10	1.679	1.653	1.640	1.607	7.740	7.701	7.691	7.627
	20	0.172	0.106	0.030	0.049	7.560	7.481	7.471	7.393
	30	3.084	3.123	3.150	3.201	7.550	7.549	7.429	7.492

Apparent molar volume and coefficients of Jones-Dole equation for the evaluation of ion- solvent interactions of barium chloride in aqueous 1, 4-dioxane and ethylene glycol solutions at T= (298.15 K to 313.15) K and at atmospheric pressure

Table 9. Values of $(\bar{V}_2^0 - \bar{V}_1^0)$, $\Delta\mu_1^{0\ddagger}$, $\Delta\mu_2^{0\ddagger}$, $T\Delta S_2^{0\ddagger}$ and $\Delta H_2^{0\ddagger}$ for BaCl₂ in 1,4-dioxane+ water and ethylene glycol+ water solutions at T=(298.15 to 313.15) K and at atmospheric pressure.

Parameters	1,4-Dioxane + Water				Ethylene glycol + Water			
	298.15K	303.15 K	308.15K	313.15K	298.15K	303.15K	308.15K	313.15K
10% (v/v)								
$10^6 (\bar{V}_2^0 - \bar{V}_1^0)/$ (m ³ mol ⁻¹)	79.58	78.24	76.91	75.57	76.89	75.90	74.94	73.25
$\Delta\mu_1^{0\ddagger}/$ (kJ mol ⁻¹)	65.29	66.07	66.86	67.62	65.73	65.99	66.61	67.44
$\Delta\mu_2^{0\ddagger}/$ (kJ mol ⁻¹)	163.14	163.97	164.53	165.00	173.06	172.76	173.89	174.40
$-T\Delta S_2^{0\ddagger}/$ (kJ mol ⁻¹)	97.85	97.90	97.67	97.38	107.33	106.76	107.27	106.96
$\Delta H_2^{0\ddagger}/$ (kJ mol ⁻¹)	65.29	66.07	66.86	67.62	65.73	65.99	66.61	67.44
20% (v/v)								
$10^6 (\bar{V}_2^0 - \bar{V}_1^0)/$ (m ³ mol ⁻¹)	84.48	83.38	82.26	81.12	82.42	81.58	80.35	79.27
$\Delta\mu_1^{0\ddagger}/$ (kJ mol ⁻¹)	65.94	66.70	67.50	68.26	66.57	67.31	68.12	68.93
$\Delta\mu_2^{0\ddagger}/$ (kJ mol ⁻¹)	159.64	160.65	161.47	162.40	170.47	171.02	172.01	172.26
$-T\Delta S_2^{0\ddagger}/$ (kJ mol ⁻¹)	93.71	93.95	93.97	94.14	103.91	103.72	103.90	103.33
$\Delta H_2^{0\ddagger}/$ (kJ mol ⁻¹)	65.94	66.70	67.50	68.26	66.57	67.31	68.12	68.93
30% (v/v)								
$10^6 (\bar{V}_2^0 - \bar{V}_1^0)/$ (m ³ mol ⁻¹)	86.91	85.76	84.60	83.47	86.68	84.91	83.47	81.54
$\Delta\mu_1^{0\ddagger}/$ (kJ mol ⁻¹)	66.45	67.22	68.02	68.77	67.37	68.19	68.78	69.66
$\Delta\mu_2^{0\ddagger}/$ (kJ mol ⁻¹)	155.73	156.38	157.30	157.93	168.49	169.22	168.51	169.75
$-T\Delta S_2^{0\ddagger}/$ (kJ mol ⁻¹)	89.28	89.16	89.29	89.17	101.12	101.03	99.73	100.09
$\Delta H_2^{0\ddagger}/$ (kJ mol ⁻¹)	66.45	67.22	68.02	68.77	67.37	68.19	68.78	69.66

3. RESULTS AND DISCUSSION

3.1. Volumetric properties.

Apparent molar volume, V_ϕ are very important for analysis of solvation state of barium chloride in aqueous EG and 1,4-DO. So V_ϕ were calculated from the density values using the equation(1)[15, 16] :

$$V_\phi = \frac{M_1}{\rho} + \frac{(\rho^0 - \rho)}{c \rho^0} \times 10^3 \quad (1)$$

In the above equation(1), ρ_0 and ρ are the densities of solvent and solution respectively; M_1 is the molecular weight of solute, c is the concentrations in mol dm⁻³. V_ϕ of a solute molecule is defined as the sum of structural volume of the solute and the changes in the volume of the solute occurred in the solution due to the interaction with solvent. The density and apparent molar volume values are given in Tables 2 and 3 respectively. V_ϕ values of BaCl₂ in given solutions increase in the following order: EG-water<1,4-DO-water. In EG-water solution, the molecules are associated with hydrogen bonding and dipole-dipole interactions, which lead to a decrease the apparent molar volume. So for EG-water system the main factor which affects the apparent molar volume is the H-bonding.

Similarly 1,4-DO is also able to associate by hydrogen bonding, due to etheric oxygen. The strength of interaction decreases in the order EG-water>1,4-DO-water.

The limiting apparent molar volumes (V_ϕ^0) was calculated using Masson equation [17] as follows:

$$V_\phi = V_\phi^0 + S_v \sqrt{c} \quad (2)$$

The intercept and the slope of the plot V_ϕ vs. \sqrt{c} gives the values of V_ϕ^0 and S_v , respectively (Table 5 and 6). The plot of V_ϕ

against \sqrt{c} were found to be linear with negative slopes. V_ϕ^0 values measure the ion-solvent interaction as it is unaffected by ion-ion interaction, whereas S_v represents the interactions between the solutes and co-solutes. At very low concentration, each ion of the electrolyte is covered by the sphere of the solvent particles so remains away from other ions. The S_v values are found to be negative. Table 6 shows the effect of temperature and organic solvent concentrations on the values of S_v . It indicates that the interaction of BaCl₂ molecules and ion-solvent interaction are decreased with the increase of temperature and increased with the organic solvent concentration. The V_ϕ^0 values (Table 5) are high and positive. It is understood that the density and shear viscosity of alcohol depends on the number of -OH groups. Since the hydrophilicity increases with the presence of -OH group, the addition of -OH group to the alcohol increases the electrostatic attraction interaction between water and alcohol due to intermolecular H-bonding. Therefore the observed decreased value of V_ϕ^0 in all the solvents suggest that the association is driven by H-bonding. It also indicates that V_ϕ^0 values decreases as the volume fraction of organic solvent increases. It may be due to the decrease in ion-solvent interactions. The limiting apparent molar volumes (V_ϕ^0) of a salt in aquo-organic solvent can be affected by polarity and dielectric constant of the solvent[18]. It is observed that the (V_ϕ^0) values of BaCl₂ in organic solvents decrease with decrease in polarity of solvent and with an increase in temperature. Again the (V_ϕ^0) values of BaCl₂ in aqueous 1,4-DO are higher than aqueous EG. This is due to strong ion-solvent interactions in it which are strengthened at low temperatures.

The temperature dependency of V_{ϕ}^0 values of BaCl_2 solution can be written by the following Equation[19]:

$$V_{\phi}^0 = a_0 + a_1 T + a_2 T^2 \quad (3)$$

Here T is temperature of solution in Kelvin. Coefficient a_0 , a_1 , and a_2 in Eq.(3) are calculated using the least-squares fitting method at $T=298.15-313.15$ K (Table 7). The Standard partial molar isobaric expansivity (E_{ϕ}^0), measures the ion-solvent interactions, solvation behavior and also finds the change of volume with temperature. So E_{ϕ}^0 can be obtained by differentiating Eq.(3) w.r.t T :

$$E_{\phi}^0 = (\partial V_{\phi}^0 / \partial T)_p = a_1 + 2a_2 T \quad (4)$$

The E_{ϕ}^0 values are shown in Table 6. On increasing temperature, some water molecules may be escaped from hydration layers of ions. This would cause a reduction in the apparent molar volume and consequently E_{ϕ}^0 values become negative. It is clear that E_{ϕ}^0 values increase with increase in temperature, which shows the absence of caging effect. The structure promoting/breaking tendency of salt in aqueous solution can be discussed with the help of Helper equation[16] on the following basis:

$$(\partial E_{\phi}^0 / \partial T)_p = (\partial^2 V_{\phi}^0 / \partial T^2)_p = 2a_2 \quad (5)$$

The structure making/ breaking ability of the solute in the solution can be determined using the sign of $(\partial E_{\phi}^0 / \partial T)_p$ value.

The positive and negative value of $(\partial E_{\phi}^0 / \partial T)_p$ shows the structure making and breaking ability of solute in solution respectively. The $(\partial E_{\phi}^0 / \partial T)_p$ values are shown in. Table 7. The positive value of $(\partial E_{\phi}^0 / \partial T)_p$ indicates that BaCl_2 predominantly acts as solvent structure maker in aquo-organic solvents over $T=298.15-313.15$ K. For the aqueous solutions, voids may not be easily acquired due to strong inter-molecular H-bonds between the H_2O molecules[20].

The values of V_{ϕ}^0 of a solute in aqueous solution may be obtained from four contributions

$$V_{\phi}^0 = V_{\text{Int}} + V_{\text{Coul}} + V_{\text{Str}} + V_{\text{Hb}} \quad (6)$$

Where V_{Int} is intrinsic volume, V_{Coul} is the volume caused by coulombic interactions of water-organic solvents, V_{Str} is the volume due to the structure of water and V_{Hb} is the volume contribution due to H- bonding in solutions.

Eq. 6 can also be represented as:

$$V_{\phi}^0 = V_{\text{Int}} + V_{\text{Hy}} \quad (7)$$

Where V_{Hy} is the interactions between solute and solvent due to hydration and hydrophilic effect of the molecules. Temperature has negligible effect on the values of V_{Int} so the decrease in V_{ϕ}^0 by increasing temperature occurs with the hydration.

Isobaric thermal expansion coefficient (α) was determined from the limiting apparent molar volumes data according to the Eq.8 .

$$\alpha = (E_{\phi}^0 / V_{\phi}^0) \quad (8)$$

The solvation number (B/V_{ϕ}^0) can also be used to determine the solvation process of a solute. The values of α and B/V_{ϕ}^0 are given in Table 8. Larger the value of B/V_{ϕ}^0 indicates that the solute is more solvated[21]. The B/V_{ϕ}^0 values of BaCl_2 in mixed solvents indicate that, the B/V_{ϕ}^0 of the solute are more than 2.2 and decrease at higher temperature and increase in the concentration of organic content. Some water molecules leave from the solvated

sphere of the ions due to squeezing in solvated volume. This is due to strong ion-solvent interactions.

Limiting the apparent molar volume of transfer, ΔV_{ϕ}^0 from water to aqueous solutions of 1,4-DO and EG have been estimated by the following formula[22, 23]

$$\Delta_i V_{\phi}^0 = V_{\phi}^0(\text{aqueous 1,4-DO or EG}) - V_{\phi}^0(\text{water}) \quad (9)$$

$\Delta_i V_{\phi}^0$ represents the solute-cosolute interaction because it is free from solute-solute interaction[27].

It has been observed that the $\Delta_i V_{\phi}^0$ values (Table 5) are positive at $T=298.15-313.15$ and increase with an increase in the concentration of both DO and EG in water respectively. In aqueous solution, the effect of concentration on the thermodynamic properties of a solute can be determined by interaction of hydration co-sphere. As per the model proposed by Friedmann and Krishnan[24], when two solute particles in the same solvent come closer, their co-spheres overlap displacing some co-sphere materials and results in a change in the volume of solution. The value of $\Delta_i V_{\phi}^0$, will be positive or negative depending on the dominant interactions between the nature of solute and co-solute. The positive value of $\Delta_i V_{\phi}^0$ is due to ionic and negative values of $\Delta_i V_{\phi}^0$ is due to non-ionic or nonpolar. The predominance of the hydrophobic-hydrophobic and ion-hydrophobic group interactions result in positive values of $\Delta_i V_{\phi}^0$ and it reduces the effect of electrostriction of hydration co-sphere of BaCl_2 and 1,4-DO and EG. This effect increases with increase in the concentration of DO and EG in water (Fig.1- 4).

3.2. Viscosity B-coefficient.

Viscosity of barium chloride solution having concentration 0.001 to 0.1 mol dm^{-3} in mixed solvents was measured at $T=(298.15$ to $313.15)$ K. The viscosity was determined from the formula: $\eta/\eta_0 = (t \times \rho) / (t_0 \times \rho_0)$; where ρ , t , η are the density, time and absolute viscosity of solution, while ρ_0 , t_0 and η_0 are same quantity for the solvent respectively. According to the Jones-Dole equation [25] the viscosity varies with concentration 'c' as given,

$$\frac{\eta}{\eta_0} = 1 + A\sqrt{c} + Bc \quad (10)$$

The constant A is the Falkenhagen coefficient which related to the long range interionic forces [26, 27], and the term $Ac^{1/2}$ is predominant in very dilute solutions. The constant B is known as viscosity B -coefficient is related to ion-solvent interactions and is a measure of the structure making and /or breaking ability of solute in solutions [28]. Jones-Dole parameters were derived by using the following equation;

$$\left(\frac{\eta}{\eta_0} - 1\right) = \eta_{sp} = A\sqrt{c} + Bc \quad (11)$$

$$\left(\frac{\eta}{\eta_0} - 1\right) / \sqrt{c} = A + B\sqrt{c} \quad (12)$$

The values of $\eta_{sp}/c^{1/2}$ at different temperature are reported in Table 4. The values of viscosity B -coefficients and A were determined from the slopes and intercepts of the linear plots of $\eta_{sp}/c^{1/2}$ vs. $c^{1/2}$ by a least square method. The values are tabulated in Table 4. The variation of $\eta_{sp}/c^{1/2}$ with molarities 'c' shows that the viscosity decreased with rising in temperature. The viscosity increases with an increase in the concentration of BaCl_2 is due to the sticking of solute molecules with solvents together by forces of attraction. The A coefficient values are found to be positive for BaCl_2 at all temperature indicating the presence of significant ion-ion interactions. Stokes and Mills equation [15] explains the ion-

Apparent molar volume and coefficients of Jones-Dole equation for the evaluation of ion- solvent interactions of barium chloride in aqueous 1, 4-dioxane and ethylene glycol solutions at T= (298.15 K to 313.15) K and at atmospheric pressure

solvent interactions in terms of viscosity B -coefficient. The viscosity B -coefficient has two contributions: such as positive and negative. The tendency of solute to attract the water molecule is due to positive contribution, while the opposite tendency is due to negative contribution. Table 5 shows the value of viscosity B -coefficient is positive throughout all experimental temperatures and decrease with increase in temperature but increase regularly at concentrated mixtures. The positive value of viscosity B -coefficient indicates the presence of ion-solvent interaction, and this is further strengthened at concentrated mixtures and weakened at a higher temperature. The B -coefficient is higher in EG-water than 1, 4-DO-water showing higher ion-solvent interaction in EG-water. The increasing value of B -coefficient in all solutions due to the increase in volume percentage of organic content shows the presence of ion-solvent interaction and structure-promoting nature of solute in the higher mass fraction of organic content. It has also been reported the structure-promoting and structure-breaking tendency of solute in the solution can be predicted from thermal coefficient (dB/dT) i.e., the temperature dependency of viscosity B -coefficient [29]. It is found that the values of viscosity B -coefficient decrease with temperature (negative value dB/dT), indicating that $BaCl_2$ in a given solution mixture acts as a structure-maker which is in agreement with the conclusion of Helper equation [16].

Viscosity B -coefficients of transfer $\Delta_t B$ from water to different aqueous 1,4-DO and EG solutions have been determined as follows:

$$\Delta_t B = B(\text{aqueous solution of 1, 4-DO or EG}) - B(\text{water}) \quad (13)$$

The $\Delta_t B$ values are presented in Table 5 and graphically represented in Fig. 1-4 which supports the result discussed above. As given by Feakins *et al* [10] the viscosity data have been analyzed on the basis of transition state theory and the free energy of activation of viscous flow, $\Delta\mu_2^{0\ddagger}$ of solution per mole of solute can be determined by the following equation.14

$$\Delta\mu_2^{0\ddagger} = \Delta\mu_1^{0\ddagger} + RT(1000B + \bar{V}_2^0 - \bar{V}_1^0)/\bar{V}_1^0, \quad (14)$$

where \bar{V}_1^0 and \bar{V}_2^0 are the partial molar volumes of the solvent and solute respectively. R is universal gas constant and T is temperature in kelvin. The free energy of activation of viscous flow for pure solvent or solvent mixture, $\Delta\mu_1^{0\ddagger}$ is given by the following equation [10, 30]:

$$\Delta\mu_1^{0\ddagger} = \Delta G_1^{0\ddagger} = RT \ln(\eta_0 \bar{V}_2^0 / h N_A) \quad (15)$$

Where $h = 6.625 \times 10^{-34}$ J s, N_A is Avogadro's number and other symbols have their usual meanings. The values of $\Delta\mu_1^{0\ddagger}$ and $\Delta\mu_2^{0\ddagger}$ are given in Table 8, which shows that the values of $\Delta\mu_1^{0\ddagger}$ are almost constant at all the studied temperature and concentration of solvent whereas $\Delta\mu_2^{0\ddagger}$ values depend on the values of B and $(\bar{V}_2^0 - \bar{V}_1^0)$. The $\Delta\mu_2^{0\ddagger}$ values are found to be positive under the experimental conditions implies that the viscous flow becomes difficult when the temperature and concentration of $BaCl_2$ and organic solvent increase in the solutions.

According to Feakins *et al*, the values of $\Delta\mu_2^{0\ddagger}$ is greater than $\Delta\mu_1^{0\ddagger}$ for solutes having positive B -coefficients implies stronger ion-solvent interactions, due to the formation of transition state associated with the breaking of intermolecular forces in the solvent structure.

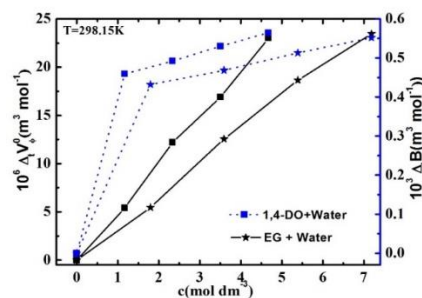


Figure 1. Plot of Limiting apparent molar volumes transfer, $\Delta_t V_\phi^0$ and Viscosity B coefficient, $\Delta_t B$ of transfer against molarity from water to different aqueous 1,4-DO and EG solutions for $BaCl_2$ at 298.15 K. (.....) line represents $\Delta_t B$ and (—) line represents $\Delta_t V_\phi^0$ values respectively.

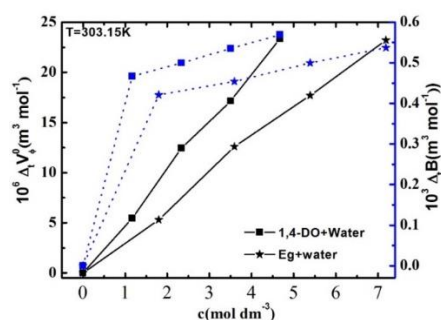


Figure 2. Plot of Limiting apparent molar volumes transfer, $\Delta_t V_\phi^0$ and Viscosity B coefficient, $\Delta_t B$ of transfer against molarity from water to different aqueous 1,4-DO and EG solutions for $BaCl_2$ at 303.15 K. (.....) line represents $\Delta_t B$ and (—) line represents $\Delta_t V_\phi^0$ values respectively.

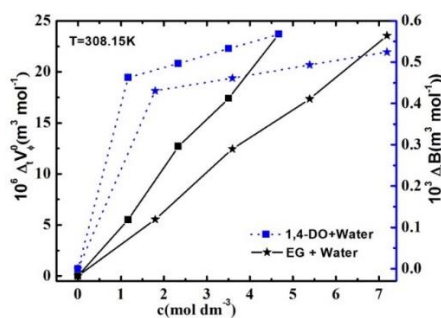


Figure 3. Plot of Limiting apparent molar volumes transfer, $\Delta_t V_\phi^0$ and Viscosity B coefficient, $\Delta_t B$ of transfer against molarity from water to different aqueous 1,4-DO and EG solutions for $BaCl_2$ at 308.15 K. (.....) line represents $\Delta_t B$ and (—) line represents $\Delta_t V_\phi^0$ values respectively.

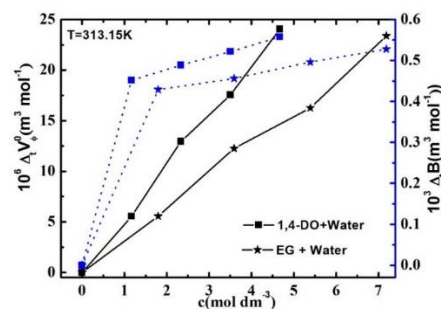


Figure 4. Plot of Limiting apparent molar volumes transfer, $\Delta_t V_\phi^0$ and Viscosity B coefficient, $\Delta_t B$ of transfer against molarity from water to different aqueous 1,4-DO and EG solutions for $BaCl_2$ at 313.15 K. (.....) line represents $\Delta_t B$ and (—) line represents $\Delta_t V_\phi^0$ values respectively.

The positive value of $\Delta\mu_2^{0\ddagger}$ indicates that $BaCl_2$ behaves like a structure maker in aqueous 1, 4-DO and EG.

Further greater the value of $\Delta\mu_2^{0\ddagger}$, the greater is the structure-making tendency. The entropy of activation, $\Delta S_2^{0\ddagger}$ and the enthalpy of activation, $\Delta H_2^{0\ddagger}$ of BaCl_2 in the mixed solvents can be determined [10]:

$$\Delta S_2^{0\ddagger} = -d(\Delta\mu_2^{0\ddagger})/dt \quad (16)$$

$$\Delta H_2^{0\ddagger} = \Delta\mu_2^{0\ddagger} + T\Delta S_2^{0\ddagger} \quad (17)$$

4. CONCLUSIONS

The measurement of viscosity, density, apparent molar volumes of solutions of BaCl_2 in aqueous DO and EG solvents are reported at $T=(298.15-313.15)$ K. The viscosity B coefficient was positive at all temperatures, indicating a structure making behaviour of BaCl_2 in studied solutions and this type of interaction is more favorable at concentrated mixtures and reversed at a higher temperature. The negative value of thermal coefficient (dB/dT) may presumably due to the strong ion-solvent interactions at lower temperatures. V_ϕ^0 values of BaCl_2 in the solutions increase with

$\Delta S_2^{0\ddagger}$ can also be found from the negative slope obtained from the plot of $\Delta\mu_2^{0\ddagger}$ vs. T . The values of $\Delta H_2^{0\ddagger}$ are given in Table 9. It has been found that the values of $\Delta S_2^{0\ddagger}$ are negative and that of $\Delta H_2^{0\ddagger}$ are positive at all the ranges of temperature which says that the transition state is associated with breaking of bonds.

decrease in temperature so ion-solvent interactions are favorable at low temperatures. The negative value of E_ϕ^0 and the positive values of Helper's constant of BaCl_2 in the mixture prove that BaCl_2 in the studied aquo-organic solvents acts as a structure-maker. Further the positive value of $\Delta\mu_2^{0\ddagger}$ proves the structure-making ability of BaCl_2 and positive value of $\Delta H_2^{0\ddagger}$ at all ranges of temperature implies that transition state is associated with the breaking of bonds.

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6. ACKNOWLEDGEMENTS

The author (BBN) thanks to the Principal of Vikram Deb Autonomous College and HOD Mr. Ranjan Kumar Pradhan for providing the infrastructure as an initiation for the improvement of research activities in the P.G. Department of Chemistry



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