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Comparative Study of Solvation Behaviour of Oxidising agents like $KClO_3$, $KBrO_3$ and KIO_3 in Aqueous Solvent Systems at Different Temperatures

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ABSTRACT

The investigation of the solvation trend of oxidizing agents like $KClO_3$, $KBrO_3$ and KIO_3 as electrolytes in aqueous salt solution renders the data suited to interpret ion–ion, solute–solvent, ion-solvent and solvent–solvent interactions and synergy. Apparent molar volumes ($\varnothing_V$) and viscosity B-coefficients for $KClO_3$, $KBrO_3$ and KIO_3 solutions in aqueous 0.5 % KCl ,system have been calculated from density (ρ) and viscosity (η) measurements at 298.15 to 313.15 K using a calibrated bicapillary pycnometer and the simple, yet accurate apparatus known as Ubbelohde viscometer respectively. Jones-Dole equation, Masson's equation, Roots equation and Moulik's equations are implemented to analyse various interactions inter and intra ionic attractions among the ion–ion, ion–solvent and solute–solvent. Additionally the apparent molar volumes of transfer $\Delta\varnothing$(tr) and Rate constant diffusion controlled reaction (k_d) are valuated.

Keywords: $KBrO_3$, $KClO_3$ and KIO_3, Density, Viscosity, B-coefficient, K_d.

INTRODUCTION

Generally, an oxidizing agent donates oxygen atoms to a reactant or substrate and hence the oxidizing agent is also called as an oxygenation reagent or oxygen-atom transfer agent[1] or it also serve as electron acceptors. Apart from oxidising agent, $KBrO_3$ has been used as a food additive, mainly in the bread-making process, flour treatment, as a component of cold-wave hair lotions. Though potassium bromate ($KBrO_3$) is banned for food use, it is used as a flour improver and high riser in bakery industries. It is known as a renal carcinogen and toxic effects of potassium bromate on endocrine glands was studied[2]. $KClO_3$ is used as safe animal husbandry tool[3] for economically important food animals like sheep, cattle, swine and poultry animals. Potassium chlorate is also measured in dietary supplements and flavour enhancing ingredients[4] and also in bottled drinking water or mineral water[5].

Potassium iodate also has been used as a food additive, to prevent iodine deficiency. It may be used to protect against the health risks caused by accumulation of radioactive iodine in the thyroid by administrating and saturating the body with a stable source of iodine in the form of KIO_3 prior to exposure[6]. KIO_3 is the best alternative to potassium iodide KI, as KI has poor shelf life in humid and hot climates[7].The importance of oxidizing agents towards the medical science lead us to undertake the present study.

The extensive information on the transport, thermodynamic and physicochemical properties of oxidizing agents are needed and plays a vital role in multiple industries, biochemical processes, in designing and development of drugs, medicines, vaccines, dyes, marine products and also for thermal treatment and storage of foods. For this perspective, the comparative study of solvation behaviour of oxidizing agents in aqueous solutions of 0.5% KCl, play crucial role not only in generation but systematization of physicochemical information of the studied solute and solvents. The major intent of this study was to assess the effect of molar concentration of solute and temperature on the apparent molar volume, various interaction parameters and solvolysis of oxidizing agent in different solvent systems[8-9].

EXPERIMENTAL

Materials

The water used for the preparation of solutions was deionised and purified by successive distillation. The specific conductance of distilled water was found <5x10^{-6} S.cm^{-1}. The $KClO_3$, $KBrO_3$ and KIO_3 of high purity was obtained from Sigma Aldrich, while KCl from S.D. Fine Lab were vacuum dried and used without further purification. The solutions of molarity range (6.5×10^{-3} to 3.65×10^{-2}) mol.L^{-1} were prepared and the measurements of phytochemical properties were carried out at four different temperatures. The precision of balance used was ± 1×10^{-5} gram.

Density measurements

Calibration of the bicapillary pycnometer was done by measuring the densities of triple distilled water. The densities of $KClO_3$, $KBrO_3$ and KIO_3 solutions in aqueous 0.5% KCl, were measured by the same calibrated pycnometer at 298.15, 303.15, 308.15, and 313.15K temperatures. The density was measured with an accuracy of ±1.48 ×10^{-4} g.cm^{-3}.

Viscosity measurements

The six different concentrations (0.0065M to 0.0365M) of solutions of $KClO_3$, $KBrO_3$ and KIO_3 were prepared in aqueous 0.5% KCl, solvent systems. To determine the influence of temperature on viscosity, the time outflow were measured at 298.15, 303.15, 308.15, and 313.15K for all six different concentrations.by using Ubbelohde viscometer. The solution viscosities were measured with an uncertainty of ± 2.4×10^{-4} mPa.s and the flow time will be measured at the accuracy of ± 0.01 s. Demerstat with an accuracy of ± 0.1 K is used to maintain the required temperature of thermostat.

Data evaluation

The measured density data is used to evaluate the apparent molar volumes $\varnothing_V$, using the following equation[10-13].

$$\varnothing_V = \frac{1000(\rho_0 - \rho)}{C\rho_0} + \frac{M2}{\rho} \quad (1)$$

Where, M_2, is the molar mass of the $KClO_3$, $KBrO_3$ and KIO_3, C is the concentration (mol.L^{-1}), and ρ and ρ_0 are the densities of the solution and the solvent, respectively.

The apparent molar volumes ($\varnothing_V$) of all the three oxidising agents were plotted against the square root of selected concentration range ($C^{½}$) in accord with the Masson's equation.[14]

$$\varnothing_V = \varnothing_V^0 + S_v.C^{½} \quad (2)$$

Values of empirical parameters $\varnothing_V^0$ and S_v which depends on temperature and also on the nature of solute, solvent have been obtained from the linear graphs plotted between $\varnothing_V$ and $C^{½}$ The viscosity data for the $KClO_3$, $KBrO_3$ and KIO_3 in aqueous 0.5% KCl, were plotted in accordance with Jones-Dole equation.[15]

$$\eta_r=(\eta/\eta_o)=1+AC^{½}+BC \quad (3)$$

Where $\eta_r = (\eta/\eta_o)$ and η, η_o are viscosities of the solution and solvent respectively, C is the molar concentration. The B-coefficients were obtained from the linear plots using the least-square fitting method. The A-coefficient reflects solute-solute interaction[16-17] and the B-coefficient reflect the solute-solvent interactions. Since in general, A/B <<1, the Jones –Dole equation reduces to,

$$\eta_r = l + \beta.C, \quad (4)$$

The relation between molar concentration and relative viscosity data of these solutions have also been fitted in Moulik equation.

$$\eta_r^2 = M + K\,C^2 \quad (5)$$

The density data of these solutions have also been used to deduce the values of R and S constants using Root's equation.

$$(d-d_0)/C = R - SC^{1/2} \quad (6)$$

The viscosity data have been also employed to determine the diffusion controlled reaction rate constant k_d[18].

$$k_d = \frac{8RT}{3\eta} \quad (7)$$

RESULTS AND DISCUSSION

The densities (ρ) and viscosities (η) values of $KClO_3$, $KBrO_3$ and KIO_3 in aqueous 0.5% KCl, at different temperatures are reported Table 1. It is observed that densities and even viscosities increase with increase in molar concentration while it decrease with increase in temperature for all selected oxidising agents. Similar observations were previously made by[19-22] for other solutions.

The values of apparent Molar Volumes ($\varnothing_V$) and Relative Viscosities (η_r) of $KClO_3$, $KBrO_3$ and KIO_3 in selected solvent systems and at four different temperatures are reported in Table 2. The positive values of $\varnothing_V$ for all three solute systems decrease with increase of concentration in KCl. Derived relative viscosities from the viscosity data are found to increase with increase in concentrations.

Table 1: Densities and Viscosities of $KClO_3$, $KBrO_3$ and KIO_3 solution in 0.5% KCl, at different temperatures

Solute System	Molar Conc. of Solute(C) mol/dm³	Temperatures Density, (ρ)/(g.cm⁻³)				Temperatures Viscosity, (η) / (mPa.s)			
		298.15K	303.15K	308.15K	313.15K	298.15K	303.15K	308.15K	313.15K
$KClO_3$	0.0065	1.00362	1.00198	1.00032	0.99920	0.9143	0.8311	0.7491	0.6757
	0.0105	1.00450	1.00284	1.00120	1.00009	0.9174	0.8351	0.7539	0.6812
	0.0155	1.00560	1.00391	1.00229	1.00117	0.9213	0.8400	0.7598	0.6882
	0.0215	1.00690	1.00519	1.00358	1.00249	0.9259	0.8459	0.7670	0.6966
	0.0285	1.00845	1.00668	1.00513	1.00402	0.9314	0.8528	0.7754	0.7063
	0.0365	1.01010	1.00854	1.00713	1.00572	0.9376	0.8607	0.7850	0.7175
$KBrO_3$	0.0065	1.00431	1.00299	1.00143	0.99946	0.9180	0.8397	0.7615	0.6834
	0.0105	1.00538	1.00407	1.00263	1.00068	0.9231	0.8457	0.7685	0.6902
	0.0155	1.00676	1.00541	1.00398	1.00219	0.9294	0.8532	0.7771	0.7011
	0.0215	1.00839	1.00705	1.00569	1.00402	0.9370	0.8622	0.7875	0.7128
	0.0285	1.01036	1.00898	1.00764	1.00617	0.9460	0.8728	0.7996	0.7263
	0.0365	1.01236	1.01107	1.00991	1.00861	0.9561	0.8848	0.8134	0.7419
KIO_3	0.0065	1.00518	1.00391	1.00264	1.00099	0.9296	0.8461	0.7672	0.6834
	0.0105	1.00666	1.00531	1.00401	1.00238	0.9349	0.8516	0.7742	0.6902
	0.0155	1.00851	1.00711	1.00572	1.00407	0.9417	0.8586	0.7824	0.7011
	0.0215	1.01073	1.00925	1.00775	1.00615	0.9497	0.8669	0.7924	0.7128
	0.0285	1.01333	1.01176	1.01018	1.00855	0.9592	0.8767	0.8042	0.7263
	0.0365	1.01649	1.01476	1.01293	1.01101	0.9699	0.8879	0.8177	0.7419

Table 2: Apparent molar volumes and Relative viscosities of $KClO_3$, $KBrO_3$ and KIO_3 solution in 0.5% KCl, at different temperatures

Solute System	Molar Conc. of Solute(C) mol/dm³	Temperatures Apparent molar volumes, ($\varnothing_V$) /cm³.mol⁻¹				Temperatures Relative viscosities, (η_r)			
		298.15K	303.15K	308.15K	313.15K	298.15K	303.15K	308.15K	313.15K
$KClO_3$	0.0065	121.43	121.61	121.76	121.91	1.0188	1.0095	1.0099	1.0177
	0.0105	121.34	121.54	121.70	121.84	1.0223	1.0131	1.0163	1.0261
	0.0155	121.25	121.45	121.62	121.74	1.0266	1.0179	1.0244	1.0366
	0.0215	121.16	121.37	121.52	121.67	1.0318	1.0239	1.0341	1.0492
	0.0285	121.06	121.28	121.41	121.57	1.0379	1.0312	1.0454	1.0639
	0.0365	120.94	121.14	121.30	121.47	1.0448	1.0396	1.0583	1.0807
$KBrO_3$	0.0065	165.59	165.81	166.06	166.40	1.0231	1.0236	1.0267	1.0293
	0.0105	165.39	165.62	165.89	166.20	1.0286	1.0309	1.0360	1.0396
	0.0155	165.23	165.44	165.71	165.98	1.0356	1.0401	1.0477	1.056
	0.0215	165.03	165.25	165.48	165.74	1.0441	1.0510	1.0617	1.0735
	0.0285	164.85	165.08	165.29	165.53	1.0541	1.0639	1.0782	1.0939
	0.0365	164.62	164.88	165.09	165.29	1.0655	1.0785	1.0966	1.1175
KIO_3	0.0065	212.39	212.67	213.00	213.32	1.0358	1.0312	1.0344	1.0293
	0.0105	212.14	212.43	212.74	213.07	1.0418	1.0381	1.0434	1.0396
	0.0155	211.89	212.17	212.48	212.82	1.0494	1.0466	1.0548	1.0560
	0.0215	211.60	211.88	212.20	212.52	1.0583	1.0567	1.0683	1.0735
	0.0285	211.27	211.60	211.89	212.24	1.0688	1.0687	1.0843	1.0939
	0.0365	210.99	211.26	211.54	211.96	1.0808	1.0824	1.1024	1.1175

The apparent molar volumes at infinite dilution ($\varnothing_V^0 = V_2^0$) and slopes S_V calculated using Masson equation (2) are reported in Table 3. The $\varnothing_V^0$ values of $KClO_3$, $KBrO_3$ and KIO_3 under investigation in KCl are large and positive suggests presence of strong solute-solvent interactions promotes structure making effect[23].

Table 3: Masson, Moulik, Jone-Dole and Roots parameters of $KClO_3$, $KBrO_3$ and KIO_3 solution in 0.5% KCl at different temperatures

Parameters	Temperature (K)	$KClO_3$	$KBrO_3$	KIO_3
Masson's Parameters				
$\varnothing_V^0$	298.15	121.7	166.2	213.4
	303.15	121.9	166.4	213.7
	308.15	122.1	166.8	214.1
	313.15	122.2	167.2	214.3
S_v	298.15	-4.05	-8.62	-12.8
	303.15	-4.03	-8.34	-12.64
	308.15	-3.89	-8.93	-13.06
	313.15	-3.16	-10.07	-12.42
Moulik Parameters				
K	298.15	57.27	66.49	71.27
	303.15	68.16	86.46	80.91
	308.15	64.00	112.20	108.80
	313.15	86.68	142.70	142.70
M	298.15	1.03	1.05	1.08
	303.15	1.02	1.06	1.07
	308.15	1.03	1.06	1.08
	313.15	1.03	1.07	1.07
Jone-Dole Parameters				
A	298.15	0.03	0.21	0.28
	303.15	0.05	0.19	0.30
	308.15	-0.03	0.19	0.28
	313.15	0.03	0.17	0.29
B	298.15	0.35	0.71	0.54
	303.15	0.93	1.15	0.47
	308.15	1.58	1.60	1.14
	313.15	1.79	2.28	1.16
Roots Parameters				
R	298.15	1.45	0.75	0.74
	303.15	1.46	0.73	0.73
	308.15	1.50	0.73	0.73
	313.15	1.39	0.59	0.59
S	298.15	-5.17	-2.36	-2.36
	303.15	-5.21	-2.31	-2.31
	308.15	-5.50	-2.21	-2.21
	313.15	-4.86	-1.41	-2.36

The Diffusion reaction rate constant (k_d) evaluated by equation (7) and are reported in Table 4. The apparent molar volumes of transfer $\Delta\varnothing$(tr) of $KClO_3$, $KBrO_3$ and KIO_3 and are obtained from the following relation and are included in Table 5.

$\Delta\varnothing V$ (tr) = $\varnothing V$ (of solute in 0.5% salt solutions) – $\varnothing V$ (of solute in DW) (8)

Figure 1 is the plot of $\varnothing V$ ($cm^3.mol^{-1}$) Versus $C^{½}$ ($mol^{1/2}.dm^{-3/2}$) for $KClO^3$ solution in 0.5% KCl at T = 298.15 to 313.15K. $KBrO_3$ and KIO_3 solution in 0.5% KCl also gave the similar linear plots. It is clear that for all the three solutes i.e. $KClO_3$, $KBrO_3$ and KIO_3 in 0.5% KCl solution, the values of $\varnothing V$ ($cm^3.mol^{-1}$) are positive while the listed slope S_v is negative. Since extent of solute-solute interactions are interpreted from the slope S_v, here S_v is negative[24-25], which indicate the strong interaction amongst solute-solute. Secondly there is no specific trend with S_v values either with temperature or concentration, it proposes that the solute-solute interactions are unaffected to change in temperature.

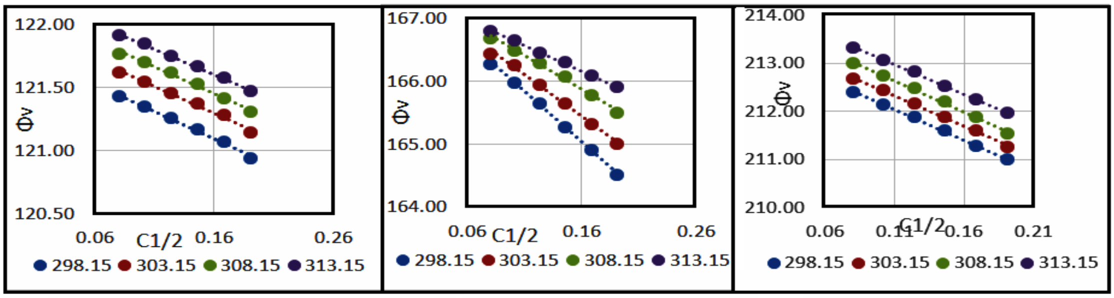
Plots of $(\eta_r - I)/C^{1/2}$ vs $C^{1/2}$ for $KClO_3$, $KBrO_3$ and KIO_3 solution in 0.5% KCl at different temperatures are shown in Fig. 2. The linear plots of $(\eta_r - I)/C^{1/2}$ vs $C^{1/2}$ are obtained at all temperatures with regression coefficients higher than 0.99. The slopes of the plots are positive.

Table 4: Diffusion reaction rate constant kd ($L\ mol^{-1}\ s^{-1}$) values of $KClO_3$, $KBrO_3$ and KIO_3 solution in 0.5% KCl solution

Solvent System	Molar Conc. of (C) mol/dm³	Temperatures 298.15K	303.15K	308.15K	313.15K
		Diffusion reaction rate constant k_d ($L\ mol^{-1}\ s^{-1}$) x 1010			
$KClO_3$	0.0065	7.23	8.09	9.12	10.28
	0.0105	7.21	8.05	9.06	10.19
	0.0155	7.18	8.00	8.99	10.09
	0.0215	7.14	7.95	8.91	9.97
	0.0285	7.10	7.88	8.81	9.83
	0.0365	7.05	7.81	8.70	9.68
$KBrO_3$	0.0065	7.20	8.00	8.97	10.16
	0.0105	7.16	7.95	8.89	10.06
	0.0155	7.11	7.88	8.79	9.90
	0.0215	7.05	7.79	8.68	9.74
	0.0285	6.99	7.70	8.54	9.56
	0.0365	6.91	7.60	8.40	9.36
KIO_3	0.0065	7.11	7.94	8.90	10.16
	0.0105	7.07	7.89	8.83	10.06
	0.0155	7.02	7.83	8.73	9.90
	0.0215	6.96	7.75	8.62	9.74
	0.0285	6.89	7.67	8.49	9.56
	0.0365	6.82	7.57	8.36	9.36

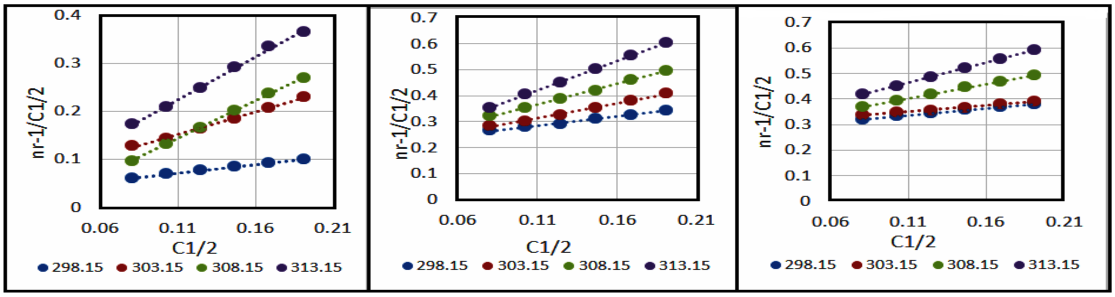
Table 5: The apparent molar volumes of transfer $\Delta\varnothing_V$(tr) of $KClO_3$, $KBrO_3$ and KIO_3 solution in 0.5% KCl solution

Solvent System	Molar Conc. of KBrO^3 (C) mol/dm^3	Temperatures 298.15K	303.15K $\Delta\varnothing_V$(tr) /cm^3.mol^{-1}	308.15K	313.15K
$KClO_3$	0.0065	-0.06	-0.10	-0.12	-0.03
	0.0105	-0.11	-0.13	-0.16	-0.07
	0.0155	-0.12	-0.13	-0.19	-0.12
	0.0215	-0.15	-0.17	-0.22	-0.18
	0.0285	-0.24	-0.15	-0.27	-0.19
	0.0365	-0.20	-0.20	-0.29	-0.28
$KBrO_3$	0.0065	-0.65	-0.76	-0.82	-0.93
	0.0105	-0.66	-0.65	-0.68	-0.77
	0.0155	-0.68	-0.64	-0.6	-0.67
	0.0215	-0.75	-0.6	-0.51	-0.49
	0.0285	-0.80	-0.6	-0.42	-0.37
	0.0365	-0.83	-0.57	-0.37	-0.21
KIO_3	0.0065	-0.17	-0.23	-0.08	-0.13
	0.0105	-0.23	-0.24	-0.13	-0.17
	0.0155	-0.24	-0.24	-0.17	-0.26
	0.0215	-0.29	-0.27	-0.22	-0.29
	0.0285	-0.36	-0.27	-0.25	-0.36
	0.0365	-0.40	-0.3	-0.39	-0.42



1. $KClO_3$ in 0.5 % KCl 2. $KBrO_3$ in 0.5 % KCl 3. KIO_3 in 0.5 % KCl

Fig. 1. Plots of $\varnothing_V$ (cm^3.mol^{-1}) versus C^{½} (mol1/2.dm$^{-3/2}$) for $KClO_3$, $KBrO_3$, KIO_3 in 0.5% KCl at T = 298.15 to 313.15K



1. $KClO_3$ in 0.5 % KCl 2. $KBrO_3$ in 0.5 % KCl 3. KIO_3 in 0.5 % KCl

Fig. 2. Plots of (ηr-1)/C^{½} vs C^{½} for $KClO_3$, $KBrO_3$, KIO_3 in 0.5% KCl at T = 298.15 to 313.15K

Sharma, Rani, R., Kumar, A., & Bamezai,[26] observed that during ionic-ionic interactions, co-sphere of two ionic species overlaps and it adds to increase the volume of solution while decrease in volume is noted due to overlapping of co-sphere of hydrophilic-hydrophobic groups or ion hydrophobic groups The negative values of $\Delta\varnothing_V$ (tr) have been reported in terms of weaker ion-ion and ion-hydrophilic group interactions than the ion-hydrophobic interaction. It results in an decrease in volume. Co-sphere overlap model developed by Gurney[27] is utilised for the better understanding of various inter ionic attractions. It is also interpreted that the properties of solvent molecule in the hydration co-sphere depend on the nature of solute species[28-29].

CONCLUSION

In the present research article, solvolysis and transport properties of $KClO_3$, $KBrO_3$ and

KIO_3 solutions in 0.5% salt solutions at different temperatures and concentrations are methodically reported.

The effect of temperature on the $\varnothing_V$ has been reported in terms of ion-solvent interactions.

It has been concluded that in all the three solute systems, there exist strong solute– solvent interactions in these systems.

The $\varnothing_V^0$ values reported in present study are found to be positive suggest presence of ion-solvent interactions.

The large and positive $\varnothing_V^0$ values for all the three solute systems has been employed to predict the presence of strong solute-solvent interactions promotes structure making effect .

The Moulik, Roots and Jones-Dole reduced equation are verified for $KClO_3$, $KBrO_3$, and KIO_3 solutions in these solvent systems.

The positive K_d value interprets that solvolysis of $KClO_3$, $KBrO_3$, and KIO_3 in 0.5% salt solutions at different temperatures and concentrations is diffusion controlled process rather than activated controlled process.

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Conflicts of Interest

The authors declare no conflict of interest.

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