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# Temperature and concentration dependences of density and Viscosity of aqueous $KBrO_3$ solutions in 0.2% of $NH_4NO_3$

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**Abstract**
The study of Temperature and concentration dependences of density and Viscosity of aqueous $KBrO_3$ solutions in 0.2% of $NH_4NO_3$ provides information useful to elucidate ion–ion, ion–solvent, and solvent–solvent interactions. Apparent molar volumes ($\emptyset_V$) and viscosity B-coefficients for $KBrO_3$ solutions in aqueous 0.2% $NH_4NO_3$ and pure water system have been determined from density (ρ) and viscosity (η) measurements at 298.15 to 313.15 K using a pycnometer and Ubbelohde viscometer respectively. Masson's equation, Jones-Dole equation are used to study various interactions among the ion–ion, ion–solvent, and solvent–solvent. Further diffusion controlled reaction rate constant ($k_d$) are evaluated.

**Keywords:** $KBrO_3$, density, viscosity, B-coefficient

## Introduction
$KBrO_3$ is an oxidizing agent that has been used as a food additive, mainly in the bread-making process, flour treatment. The food additive potassium bromate ($KBrO_3$) is known as a renal carcinogen. Relationship between toxic effects of potassium bromate and endocrine glands was studied [1]. $KBrO_3$ can generate high yields of 8-hydroxydeoxyguanosine DNA adducts, which cause GC>TA transversions in cell-free systems [2]. Batch sorption experiments were performed [3] to examine the influence of various experimental parameters such as contact time, initial bromate concentration, temperature, and pH on the sorption of bromate on nano–$Al_2O_3$. The importances of oxidizing agents towards the medical science lead us to undertake the present study.

It is well known that physicochemical characterization of oxidizing agents plays a crucial role in all the stages associated to design and development of pharmaceutical drugs especially those intended to parenteral administration to cure the various diseases caused by them. In this context, as a contribution to generation and systematization of physicochemical information about oxidizing agent behavior in aqueous solutions of 0.2% $NH_4NO_3$ system, the main goal of this study was to evaluate the effect of concentration and temperature on the apparent molar volume, interaction parameters and solvolysis of oxidizing agent in used solvent systems.

## Experimental
### Materials
$KBrO_3$ of high purity was obtained from Research Lab Fine Chemicals, Mumbai, re-crystallized and then used. Deionized water with a specific conductance of < $10^{-6}$ $S.cm^{-1}$ was used for the preparation of solutions at room temperature in a molarity range ($6.0\times10^{-3}$ to $1.99\times10^{-2}$) $mol.L^{-1}$. The precision of balance used was ± $1\times10^{-5}$g.

### Density measurements
The pycnometer was calibrated by measuring the densities of triple distilled water. The densities of $KBrO_3$ solutions in aqueous 0.2% $NH_4NO_3$ and pure water were measured by bi-capillary pycnometer at different temperatures. The density was measured with an uncertainty of ±$1.48\times10^{-4}$ $g.cm^{-3}$.

### Viscosity measurements
The different compositions (0.0065M to 0.0365M) of solutions of $KBrO_3$ were prepared in aqueous 0.2% $NH_4NO_3$ and pure water solvent systems. The viscosities were measured at 298.15, 303.15, 308.15, and 313.15K temperatures for seven different concentrations. The solution viscosities were measured with an uncertainty of ± $2.4\times10^{-4}$ mPa.s by using Ubbelohde viscometer. The temperature of thermostat is maintained to desired temperature, by using demerstat with an accuracy of ± 0.2 K. The flow time will be measured at the accuracy of ± 0.01 s.

### Data Evaluation
The apparent molar volumes $\emptyset_V$, were obtained from the density results using the following equation [4-7].

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

Where $M_2$, C, $\rho$ and $\rho_0$ are the molar mass of the $KBrO_3$, concentration ($mol.L^{-1}$), and the densities of the solution and the solvent, respectively.

The apparent molar volumes ($\emptyset_V$) were plotted against the square root of concentration ($C^{½}$) in accordance with the Masson's equation [8].

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

Where is the limiting apparent molar volume $\emptyset_V^0$ and $S_v$ a semi-empirical parameter which depends on the nature of solute, solvent as well as temperature. The viscosity results

for the aqueous solutions of $KBrO_3$ in aqueous 0.2% $NH_4NO_3$ and pure water solvent systems were plotted in accordance with Jones-Dole equation [9] Where $\eta_r = (\eta/\eta_o)$ and $\eta$, $\eta_o$ 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 [10] and the B-coefficient reflect the solute-solvent interactions. Since in general, A/B <<1, the Jones –Dole equation reduces to,

$$\eta_r = 1 + \beta.C, \quad (4)$$

The relative viscosity data of these solutions have also been fitted in Moulik equation,

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

The density data of these solutions have also been fitted in Root's equation,

$$(d - d_0) / C = R - SC^{½} \quad (6)$$

where R and S are constants.

The diffusion controlled reaction rate constant $k_d$ can be evaluated by using the viscosity data as

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

The evaluated values are used to predict whether the solvolysis is fast or slow process.

## Results and Discussion

The values of the densities ($\rho$) and viscosities ($\eta$) of $KBrO_3$ in 0.2% $NH_4NO_3$ solvent systems and temperatures are reported Table-1. The table-1 reveals that densities and viscosities of $KBrO_3$ solutions under investigation decrease with increase in temperature and increases with increase in concentration. Such observations were previously made by Comesana *et al.* [11], Lee *et al.* [12, 13] and Nikumbh *et al.* [14] for other solutions.

Apparent Molar Volumes ($\emptyset_V$) and Relative Viscosities ($\eta_r$) of $KBrO_3$ in 0.2% $NH_4NO_3$ solvent systems and temperatures are reported in table-2. The positive values of $\emptyset_V$ decrease with concentration in 0.2% $NH_4NO_3$ and pure water solvent systems. The relative viscosities are found to increase with concentrations.

The apparent molar volumes at infinite dilution ($\emptyset_V^0 = V_2^0$) and slopes $S_V$, calculated using Masson equation (2) are given in table-3. The $\emptyset_V^0$ values of $KBrO_3$ under investigation in 0.2% $NH_4NO_3$ and in pure water solvent systems are large and positive suggests presence of strong solute-solvent interactions promotes structure making effect [15].

It is clear that the values of $\emptyset_V$ ($cm^3.mol^{-1}$) are positive and more or less similar in water and in salt solutions at different temperatures. The slope $S_v$ is negative for $KBrO_3$ solution in 0.2 % $NH_4NO_3$ and in pure water. Since $S_v$ is measure of solute-solute interactions [16, 17]. These results indicate that there is presence of strong solute-solute interactions. $S_v$ values do not change systematically with change in temperature, and hence it suggests that the solute-solute interactions are insensitive to change in temperature.

The Diffusion reaction rate constant ($k_d$) evaluated by equation 7 and are reported in table-4. These values reveals that the solvolysis of $KBrO_3$ in aqueous electrolyte studied is diffusion controlled rather than activated controlled process.

## Conclusions

In the present report, physicochemical properties of $KBrO_3$ solutions in water and 0.2%$NH_4NO_3$ solutions at different temperatures are systematically presented. It has been observed that there exist strong solute– solvent interactions in these systems. The values of $\emptyset_V^0$ are positive suggest presence of ion-solvent interactions. The Moulik, Roots and Jones-Dole reduced equation are verified for $KBrO_3$ solutions in 0.2%$NH_4NO_3$ solvent systems. The solvolysis of $KBrO_3$ in water and 0.2%$NH_4NO_3$ solutions at different temperatures is diffusion-controlled process.

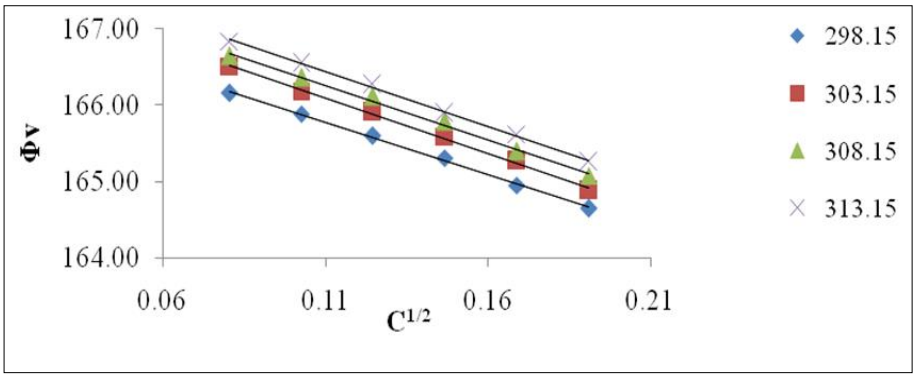
The $S_v$ values listed are all negative. There is no specific trend with $S_v$ values either with temperature or concentration or % composition. The negative $S_v$ values have been previously reported in aqueous solutions of electrolytes.

**Table 1:** Densities and Viscosities of $KBrO_3$ solution in 0.2%$NH_4NO_3$ and distilled water at different temperatures.

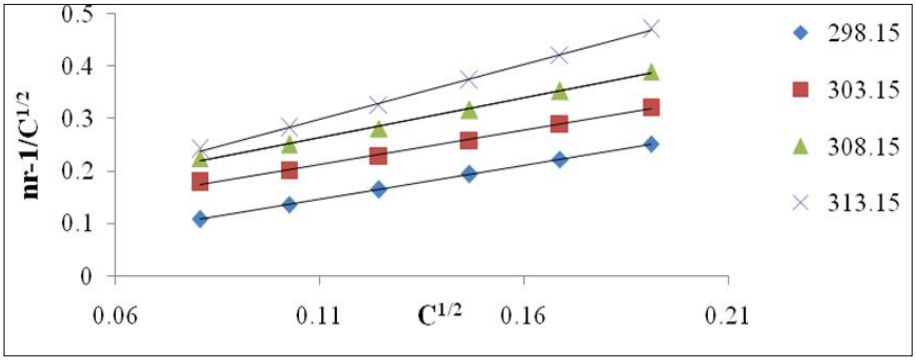
| Solvent System | Molar Conc. of $KBrO_3$ (C) mol/dm$^3$ | Temperatures | | | | Temperatures | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | 298.15K | 303.15K | 308.15K | 313.15K | 298.15K | 303.15K | 308.15K | 313.15K |
| | | Density, ($\rho$) / (g.cm$^{-3}$) | | | | Viscosity, ($\eta$) / (mPa.s) | | | |
| 0.2% $NH_4NO_3$ | 0.0065 | 1.00096 | 0.99957 | 0.99817 | 0.99669 | 0.9074 | 0.8290 | 0.7508 | 0.6717 |
| | 0.0105 | 1.00237 | 1.00096 | 0.99956 | 0.99801 | 0.9121 | 0.8342 | 0.7564 | 0.6779 |
| | 0.0155 | 1.00411 | 1.00270 | 1.00115 | 0.99974 | 0.9179 | 0.8406 | 0.7633 | 0.6856 |
| | 0.0215 | 1.00621 | 1.00482 | 1.00327 | 1.00172 | 0.9251 | 0.8483 | 0.7716 | 0.6949 |
| | 0.0285 | 1.00866 | 1.00729 | 1.00568 | 1.00407 | 0.9332 | 0.8573 | 0.7813 | 0.7057 |
| | 0.0365 | 1.01162 | 1.01010 | 1.00843 | 1.00675 | 0.9427 | 0.8675 | 0.7924 | 0.7181 |
| Distilled water | 0.0065 | 0.99908 | 0.99764 | 0.99590 | 0.99432 | 0.8962 | 0.8165 | 0.7330 | 0.6593 |
| | 0.0105 | 1.00033 | 0.99880 | 0.99718 | 0.99563 | 0.8973 | 0.8199 | 0.7389 | 0.6628 |
| | 0.0155 | 1.00190 | 1.00029 | 0.99880 | 0.99731 | 0.8986 | 0.8244 | 0.7462 | 0.6673 |
| | 0.0215 | 1.00373 | 1.00213 | 1.00073 | 0.99929 | 0.9003 | 0.8298 | 0.7551 | 0.6726 |
| | 0.0285 | 1.00596 | 1.00420 | 1.00299 | 1.00170 | 0.9022 | 0.8359 | 0.7652 | 0.6788 |
| | 0.0365 | 1.00844 | 1.00677 | 1.00557 | 1.00436 | 0.9044 | 0.8430 | 0.7769 | 0.6859 |

**Table 2:** Apparent molar volumes and Relative viscosities of $KBrO_3$ solution in 0.2%$NH_4NO_3$ and distilled water at different temperatures.

| Solvent System | Molar Conc. of $KBrO_3$ (C) mol/dm$^3$ | Temperatures | | | | Temperatures | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | 298.15K | 303.15K | 308.15K | 313.15K | 298.15K | 303.15K | 308.15K | 313.15K |
| | | Apparent molar volumes, ($\emptyset_V$) /cm$^3$.mol$^{-1}$ | | | | Relative viscosities, ($\eta_r$) | | | |
| 0.2% $NH_4NO_3$ | 0.0065 | 166.60 | 166.86 | 167.11 | 167.20 | 1.0087 | 1.0144 | 1.0181 | 1.0195 |
| | 0.0105 | 166.25 | 166.49 | 166.77 | 166.91 | 1.0140 | 1.0206 | 1.0256 | 1.0288 |
| | 0.0155 | 165.92 | 166.20 | 166.44 | 166.65 | 1.0205 | 1.0285 | 1.035 | 1.0406 |
| | 0.0215 | 165.58 | 165.85 | 166.10 | 166.32 | 1.0284 | 1.0379 | 1.0463 | 1.0546 |
| | 0.0285 | 165.22 | 165.44 | 165.71 | 165.93 | 1.0375 | 1.0489 | 1.0594 | 1.0710 |
| | 0.0365 | 164.84 | 165.11 | 165.32 | 165.63 | 1.0480 | 1.0614 | 1.0745 | 1.0898 |
| Distilled Water | 0.0065 | 166.92 | 167.19 | 167.50 | 167.73 | 1.0020 | 1.0084 | 1.0119 | 1.0142 |
| | 0.0105 | 166.63 | 166.90 | 167.17 | 167.41 | 1.0032 | 1.0128 | 1.0199 | 1.0230 |
| | 0.0155 | 166.33 | 166.58 | 166.89 | 167.12 | 1.0047 | 1.0182 | 1.0300 | 1.0338 |
| | 0.0215 | 166.01 | 166.24 | 166.57 | 166.79 | 1.0065 | 1.0248 | 1.0422 | 1.0468 |
| | 0.0285 | 165.70 | 165.91 | 166.19 | 166.45 | 1.0087 | 1.0325 | 1.0563 | 1.0620 |
| | 0.0365 | 165.34 | 165.57 | 165.86 | 166.11 | 1.0111 | 1.0412 | 1.0725 | 1.0795 |




**Plot A:** Plot of $\emptyset_V$ (cm$^3$.mol$^{-1}$) Versus C$^{½}$ (mol$^{1/2}$.dm$^{-3/2}$) for $KBrO_3$ in 0.2% $NH_4NO_3$ at T=298.15 to 313.15K.




**Plot B:** Plot of $(\eta_r-1)/C^{½}$ vs C$^{½}$ for $KBrO_3$ in 0.2% $NH_4NO_3$ solvent system at different temperatures

**Table 3:** Masson ($\emptyset_V^0$, $S_v$,) Moulik (M, K) Jone-Dole (A, B) and Roots (R, S) parameters of $KBrO_3$ solution in 0.2%$NH_4NO_3$ and distilled water at different temperatures.

| Parameters | Temp (K) | 0.2% $NH_4NO_3$ | Distilled water | Parameter | Temp (K) | 0.2% $NH_4NO_3$ | Distilled Water |
|---|---|---|---|---|---|---|---|
| $\emptyset_V^0$ | 298.15 | 167.8 | 168.0 | A | 298.15 | 0.00 | 0.083 |
| | 303.15 | 168.1 | 168.4 | | 303.15 | 0.07 | 0.297 |
| | 308.15 | 168.4 | 168.7 | | 308.15 | 0.10 | 0.549 |
| | 313.15 | 168.3 | 168.9 | | 313.15 | 0.07 | 0.59 |
| $S_v$ | 298.15 | -15.85 | -14.28 | B | 298.15 | 1.29 | -0.005 |
| | 303.15 | -15.83 | -14.73 | | 303.15 | 1.31 | -0.017 |
| | 308.15 | -16.13 | -14.85 | | 308.15 | 1.51 | -0.035 |
| | 313.15 | -14.36 | -14.59 | | 313.15 | 2.08 | -0.036 |
| M | 298.15 | 1.02 | 1.005 | R | 298.15 | 0.35 | 0.313 |
| | 303.15 | 1.04 | 1.021 | | 303.15 | 0.35 | 0.302 |
| | 308.15 | 1.04 | 1.036 | | 308.15 | 0.38 | 0.272 |
| | 313.15 | 1.05 | 1.037 | | 313.15 | 0.45 | 0.317 |
| K | 298.15 | 60.44 | 13.79 | S | 298.15 | 0.02 | 0.01 |
| | 303.15 | 73.17 | 50.33 | | 303.15 | -0.02 | 0.004 |
| | 308.15 | 88.33 | 94.62 | | 308.15 | -0.19 | 0.249 |
| | 313.15 | 111.10 | 102.30 | | 313.15 | -0.60 | 0.086 |

**Table 4:** Diffusion reaction rate constant $k_d$ (L $mol^{-1}$ $s^{-1}$) values of $KBrO_3$ solution in 0.2 % $NH_4NO_3$.

| Solvent System | Molar Conc. of $KBrO_3$ (C) mol/$dm^3$ | Temperatures | | | |
|---|---|---|---|---|---|
| | | 298.15K | 303.15K | 308.15K | 313.15K |
| | | Diffusion reaction rate constant $k_d$ (L $mol^{-1}$ $s^{-1}$ ) x $10^{10}$ | | | |
| 0.2% $NH_4NO_3$ | 0.0065 | 7.32 | 8.15 | 9.16 | 10.41 |
| | 0.0105 | 7.28 | 8.10 | 9.09 | 10.32 |
| | 0.0155 | 7.23 | 8.04 | 9.01 | 10.21 |
| | 0.0215 | 7.18 | 7.97 | 8.92 | 10.08 |
| | 0.0285 | 7.12 | 7.89 | 8.81 | 9.94 |
| | 0.0365 | 7.05 | 7.79 | 8.69 | 9.78 |

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