

# Hydration number and allied parameters of aqueous solutions of ammonium bromide through ultrasonic study

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## Abstract:

Ammonium bromide, a water-soluble wood preservative used in pharmaceutical preparations, is the focus of this economic study aimed at understanding its physical properties in aqueous solutions. A straightforward and reliable method for examining interactions between molecules in liquids and solutions is ultrasonic research. When ultrasonic waves pass through liquids, they disturb the equilibrium between the molecules of the solute and solvent. Ion-ion and ion-solvent interactions are just two aspects of solvation chemistry that ultrasonic velocity measurement can elucidate. This study investigates the hydration number of aqueous solutions of ammonium bromide at temperatures ranging from 35 to 55°C, assessing both density and ultrasonic velocity across various concentrations. From the adiabatic compressibility, the hydration number and related parameters are derived, all of which are sensitive to changes in concentration and temperature. Thus, we explore how these parameters vary with changes in temperature and concentration, providing insight into the interactions of ions and molecules in the solutions, as well as their molecular structure.

**Keywords:** allied parameters, density, hydration number, molecular interactions, ultrasonic velocity.

**Classification numbers:** 1.3, 2.1, 2.2

## 1. Introduction

Ammonium bromide, a water-soluble wood preservative used in pharmaceutical preparations, is the focus of this economic study aimed at understanding its physical properties in aqueous solutions. We investigate whether Ammonium bromide has a structure-making or -breaking property with water by examining its hydration number and allied parameters such as apparent molal compressibility, apparent molal volume, and molar solvated volume.

When an electrolyte dissolves in water, both volume and compressibility are altered due to solute-solvent interaction. The interaction of ions with water molecules is quantified by the hydration number, representing the number of water molecules surrounding the solute molecules during interaction. Previous research has explored the hydration number, assuming the incompressibility of ions and water molecules in the primary hydration sheaths directly bonded to the ions [1, 2]. The methodology for calculating the hydration number has been documented [3, 4], along with techniques for determining allied parameters such as apparent molal compressibility, apparent molal volume, and molar solvated volume using the electrolyte solution's hydration number [5-7]. For each of these parameters, three formulas were derived using adiabatic compressibility and the number of moles of solute and solvent, corroborated by traditional equations [8].

In this article, we determine the hydration number for aqueous solutions of Ammonium bromide within the temperature range

of 35 to 55°C. From these values, we estimate apparent molal compressibility, apparent molal volume, and molar solvated volume, examining their variations.

## 2. Experimental study

AnalaR grade salts and double-distilled water were used to prepare aqueous solutions of ammonium bromide at various concentrations. The ultrasonic velocity of the solutions was measured using a Mittal type ultrasonic interferometer equipped with a 2 MHz crystal and an accuracy of 0.1 m/s. Density was determined using a 10 ml specific gravity container and a single pan digital balance. Temperature control between 35 and 55°C was achieved using a thermostat accurate to 0.1°C [8].

## 3. Computation

The compressibility method, noted for its accuracy and the insightful data it provides, was employed to estimate the hydration number [9, 10]. Previous studies have established the methodology for calculating the hydration number [11-13], presuming that the free solvent molecules primarily contribute to the solution's compressibility. As every solvent possesses a unique structure, its molecules are bound more or less tightly to the solute during hydration, significantly altering the solvent's surrounding structure. Ultrasonic analysis presents a direct but precise method for determining adiabatic compressibility, which is calculated from the ultrasonic velocity and density measurements. From the

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adiabatic compressibility values, the hydration number is derived [14, 15].

Using the standard relation, the adiabatic compressibility may be calculated by:

$$\beta = \frac{1}{u^2} m^2/N \quad (1)$$

where  $\beta$  is the adiabatic compressibility of a material, a measure of how much the material can be compressed adiabatically (without heat exchange) under the influence of pressure with the units are typically Pa<sup>-1</sup> (inverse of pressure);  $u^2$  is the speed of sound in the material with the units are typically meters per second (m/s);  $\rho$  (rho) is the density of the medium, usually in kilograms per cubic meter (kg/m<sup>3</sup>);  $m^2/N$ : this part represents area per number of particles or units.

The molar hydration number  $n_h$  is calculated using Barnartt's relation [16]:

$$n_h = \frac{1}{n_2} \left[ n_1 - \frac{\beta}{\beta_0} N \right] \quad (2)$$

where  $N$  is the number of solvent molecules in 1000 cc of solvent,  $n_1$  is the number of solvent moles present in 1000 cc of solution of molar concentration  $n_2$ , and  $\beta_0$  is the solvent's compressibility.

The molal hydration number  $n'_h$  is computed through Passynski's equation [17]:

$$n'_h = \frac{N'}{n_2} \left[ 1 - \frac{\beta}{\beta_0} \right] \quad (3)$$

where  $n_2'$  is the molal concentration of the solution and  $N'$  is the number of solvent molecules in 1000 g of the solvent.

Apparent molal volume  $\phi_v$  of the mixture is calculated using the relations:

$$\phi_v = \frac{(V - n_1 V_m)}{n_2} \quad (4)$$

$$\phi_v = V_1 \left[ \frac{(N\Delta\beta)}{n_2\beta_0} \right] - n_h V_m \text{ (molar scale)} \quad (5)$$

$$\phi_v = \frac{\beta_0 n_h \left( \frac{V'}{N'} - V_m \right)}{\Delta\beta} \text{ (molar scale)} \quad (6)$$

$$\phi_v = \frac{1000}{n_2 \rho_0} (\rho_0 - \rho) + \frac{M_2}{\rho_0} \text{ (Traditional formula)} \quad (7)$$

where  $n_1$  is the number of moles of the solvent and  $n_2$  is the number of moles of the solute present in the solution, and  $\Delta\beta = \beta - \beta_0$ .  $V$  is the total volume of the solution,  $V_m$  is the molar volume of the solvent,  $V_1$  is a reference volume per mole,  $n_h$  is the number of moles of component "h",  $V'$  is a volume associated with a specific condition,  $\rho_0$  is the reference density and  $M_2$  is the molar mass or mass of component 2.

The volume of the solution measured in the molal scale is given by:

$$V' = (N'M_1 + n_2'M_2) / \rho$$

where  $M_1$  is the molar mass or mass of component 1,  $M_2$  is the molar mass or mass of component 2,  $n_h$  and  $n'_h$  are found using Eqs. (2) and (3), respectively.

Apparent molal compressibility  $\phi_k$  of the solution is calculated using the relations:

$$\phi_k = \left( \frac{\beta V - n_1 \beta_0 V_m}{n_2} \right) \quad (8)$$

$$\phi_k = -V_m \beta_0 n_h \text{ (molar scale)} \quad (9)$$

$$\phi_k = -n'_h V_m \beta_0 + \phi_v \beta \text{ (molar scale)} \quad (10)$$

$$\phi_k = \frac{1000}{n_2 \rho_0} (\rho_0 \beta - \beta_0 \rho) + \frac{M_2}{\rho_0} \beta_0 \text{ (Traditional formula)} \quad (11)$$

where  $\phi_v$  is calculated using Eq. (7).

Molar solvated volume  $\phi_s$  of the solution is computed from the following equations:

$$\phi_s = \left( \frac{n'_h V_m \beta_0}{\beta} \right) - \left( \frac{n_h V_m \Delta\beta}{\beta} \right) \quad (12)$$

$$\phi_s = \phi_v + n_h V_m \text{ (molar scale)} \quad (13)$$

$$\phi_s = \frac{V n'_h}{n_1} \text{ (molar scale)} \quad (14)$$

$$\phi_s = \frac{V \Delta\beta}{n_2 \beta_0} \text{ (Traditional formula)} \quad (15)$$

where  $\Delta\beta = \beta - \beta_0$ .

Variation of three allied parameters  $\phi_v$ ,  $\phi_k$  and  $\phi_s$  parameters with concentration: The equations for the variation of the three allied parameters  $\phi_v$ ,  $\phi_k$ , and  $\phi_s$  with the square root of molar and molal concentration ( $\sqrt{n_2}$  &  $\sqrt{n_2'}$ ) are

$$\phi_v = \phi_v^0 + n_{hv} \sqrt{n_2} \quad (16)$$

$$\phi_v = \phi_v^{0'} + n'_{hv} \sqrt{n_2'} \quad (17)$$

$$\phi_k = \phi_k^0 + n_{hk} \sqrt{n_2} \quad (18)$$

$$\phi_k = \phi_k^{0'} + n'_{hk} \sqrt{n_2'} \quad (19)$$

$$\phi_s = \phi_s^0 + n_{hs} \sqrt{n_2} \quad (20)$$

$$\phi_s = \phi_s^{0'} + n'_{hs} \sqrt{n_2'} \quad (21)$$

where  $\phi_v^0$ ,  $\phi_k^0$ ,  $\phi_s^0$  and  $\phi_v^{0'}$ ,  $\phi_k^{0'}$ ,  $\phi_s^{0'}$  are the intercepts in molar and molal scale of the linear equations and  $n_{hv}$ ,  $n_{hk}$ ,  $n_{hs}$  and  $n'_{hv}$ ,

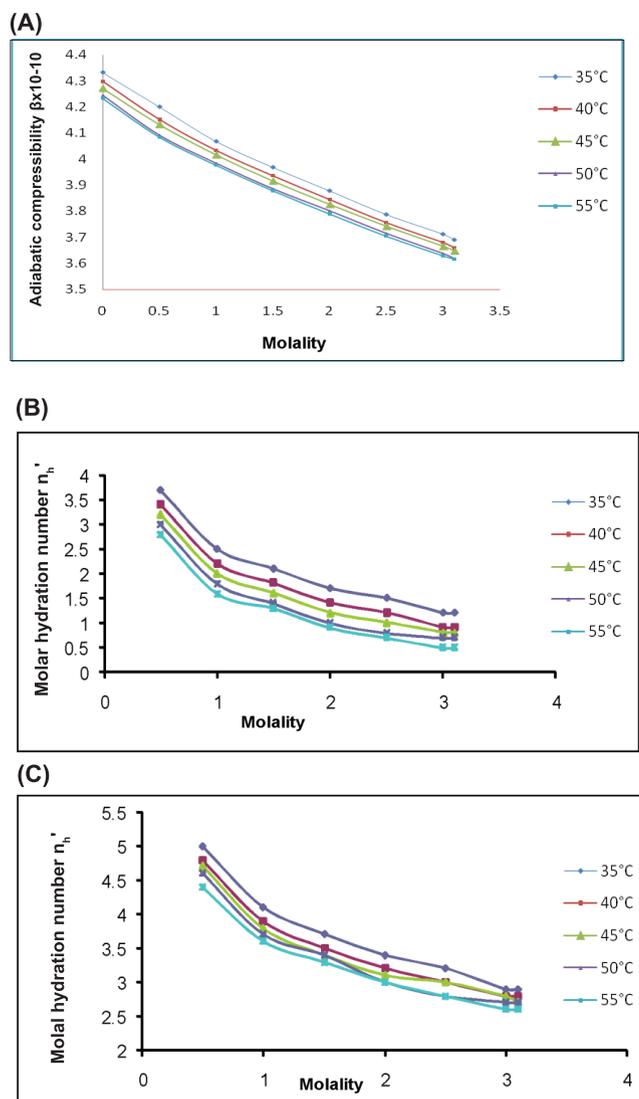
$n_{hk}$ ,  $n_{hs}$  are the slopes in molar and molal scale of the linear equations.

### 4. Results and discussion

Using adiabatic compressibility, the molar and molal hydration numbers of aqueous solutions of ammonium bromide across all concentrations and at various temperatures have been calculated and are presented in Table 1 and Fig. 1.

**Table 1. Adiabatic compressibility, molar and molal hydration numbers ( $n_h$  and  $n_h'$ ) of ammonium bromide solutions.**

Temp °C	Molality ( $n_2$ ) M	Density Kg/m <sup>3</sup>	Velocity m/s	$\beta(^{\circ}E10)$ m <sup>2</sup> /N	$n_2$	$n_1$	$n_h$	$n_h'$
35°C	0.5	1.015	1531.5	4.1991	0.49110	54.518	3.7	5.0
	1.0	1.046	1533.5	4.0666	0.96462	53.543	2.5	4.1
	1.5	1.070	1535.2	3.9670	1.42308	52.66	2.1	3.7
	2.0	1.091	1537.5	3.8769	1.86089	51.645	1.7	3.4
	2.5	1.114	1539.9	3.7859	2.28220	50.671	1.5	3.2
	3.0	1.134	1541.7	3.7104	2.67951	49.576	1.2	2.9
	3.1	1.139	1542.9	3.6886	2.75658	49.357	1.2	2.9
40°C	0.5	1.014	1539.9	4.1605	0.48891	54.275	3.4	4.8
	1.0	1.042	1541.7	4.0373	0.95943	53.255	2.2	3.9
	1.5	1.066	1543.1	3.9398	1.41236	52.263	1.8	3.5
	2.0	1.088	1545.3	3.8487	1.84283	51.144	1.4	3.2
	2.5	1.111	1547.4	3.7599	2.25951	50.167	1.2	3.0
	3.0	1.131	1549.5	3.6835	2.65354	49.096	0.9	2.8
	3.1	1.135	1550.9	3.6622	2.72971	48.876	0.9	2.8
45°C	0.5	1.014	1544.7	4.1319	0.48824	54.201	3.2	4.7
	1.0	1.041	1546.5	4.0155	0.95661	53.098	2.0	3.8
	1.5	1.065	1548.7	3.9149	1.40687	52.060	1.6	3.4
	2.0	1.087	1550.7	3.8254	1.83446	50.912	1.2	3.1
	2.5	1.109	1552.3	3.7411	2.25168	49.993	1.0	3.0
	3.0	1.130	1554.4	3.6643	2.64218	48.886	0.8	2.8
	3.1	1.134	1555.1	3.6469	2.7202	48.706	0.8	2.7
50°C	0.5	1.015	1551.9	4.0909	0.48657	54.016	3.0	4.6
	1.0	1.040	1553.5	3.9832	0.95260	52.875	1.8	3.7
	1.5	1.064	1555.7	3.8853	1.40098	51.842	1.4	3.4
	2.0	1.085	1557.3	3.8006	1.82426	50.629	1.0	3.0
	2.5	1.108	1559.3	3.7139	2.23220	49.560	0.8	2.8
	3.0	1.127	1561.7	3.6368	2.63035	48.667	0.7	2.7
	3.1	1.132	1562.9	3.6168	2.70712	48.472	0.7	2.7
55°C	0.5	1.014	1554.4	4.0839	0.48538	53.883	2.8	4.4
	1.0	1.039	1556.3	3.9759	0.94951	52.703	1.6	3.6
	1.5	1.062	1558.4	3.8783	1.39627	51.668	1.3	3.3
	2.0	1.083	1560.7	3.7894	1.81841	50.467	0.9	3.0
	2.5	1.106	1562.7	3.704	2.22336	49.364	0.7	2.8
	3.0	1.126	1564.6	3.6289	2.60925	48.277	0.5	2.6
	3.1	1.129	1565.1	3.6156	2.68500	48.076	0.5	2.6



**Fig. 1. (A) Adiabatic compressibility, (B) molar hydration number, and (C) molal hydration number - water + ammonium bromide.**

These variations indicate that ion-solvent interactions exist within the system. The molar and molal hydration numbers decrease with increases in concentration and temperature in aqueous solutions of ammonium bromide. For instance, the molar hydration number declines from 4 to 1 and the molal hydration number from 5 to 3 at 35°C, as detailed in Table 1. A similar pattern of reduction is observed at other temperatures. Temperature influences a slight decrease in both molar and molal hydration numbers.

The reduction in molar hydration numbers with increasing molality is attributed to ion-ion association, consequently decreasing ion-solvent interaction [18]. The number of solvent molecules surrounding an ion also diminishes due to the presence of ion pairs in highly concentrated solutions. Therefore, hydration by water molecules is reduced at higher concentrations, which is reflected by the declining molar

**Table 2. Allied parameters of ammonium bromide solutions.**

Apparent molal volume  $\phi_v$  of ammonium bromide salt in water.

System	Temp. molality	$\phi_v$ (*10 <sup>6</sup> ) m <sup>3</sup> /mole				
		35°C	40°C	45°C	50°C	55°C
Water + ammonium bromide	0.5	24.574	27.414	27.967	31.105	31.223
	1.0	30.838	33.302	35.257	37.713	38.673
	1.5	32.143	35.381	37.399	39.090	39.854
	2.0	34.461	38.155	40.067	42.145	42.677
	2.5	35.840	38.982	40.073	43.172	43.967
	3.0	37.925	40.529	41.776	42.830	45.083
	3.1	38.305	40.862	41.781	42.931	45.178

Apparent molal compressibility  $\phi_k$  of ammonium bromide salt in water.

System	Temp. molality	$\phi_k$ (*10 <sup>14</sup> ) m <sup>5</sup> /N/mole				
		35°C	40°C	45°C	50°C	55°C
Water + ammonium bromide	0.5	-2.875	-2.640	-2.472	-2.317	-2.142
	1.0	-1.942	-1.739	-1.522	-1.368	-1.229
	1.5	-1.645	-1.377	-1.208	-1.104	-1.006
	2.0	-1.352	-1.061	-0.904	-0.755	-0.708
	2.5	-1.166	-0.919	-0.816	-0.604	-0.541
	3.0	-0.934	-0.740	-0.632	-0.577	-0.402
	3.1	-0.908	-0.722	-0.626	-0.573	-0.387

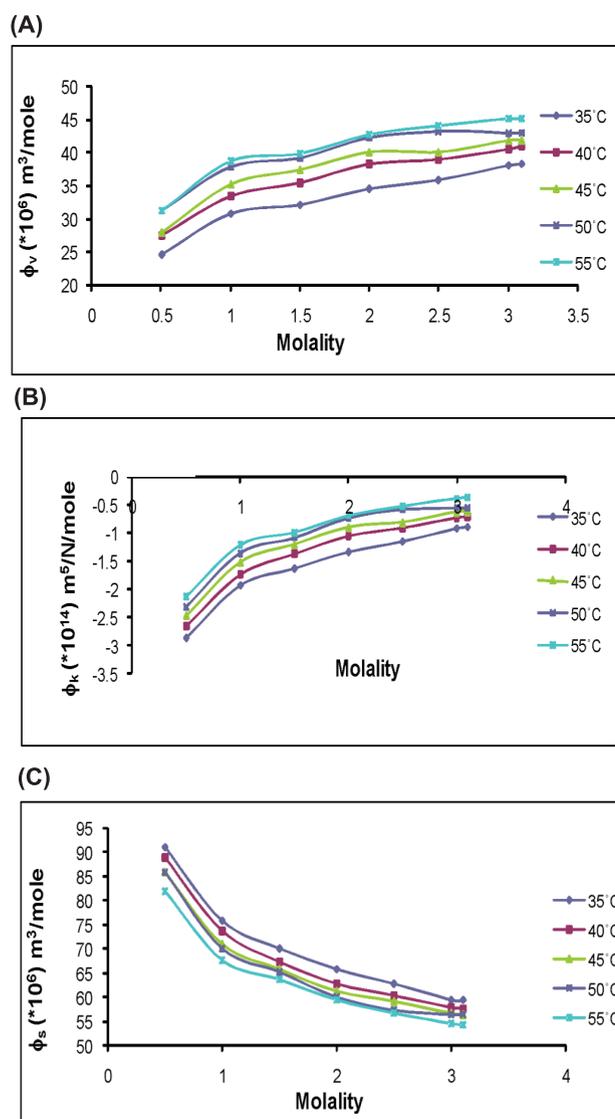
Molar solvated volume  $\phi_s$  of ammonium bromide salt in water.

System	Temp. Molality	$\phi_s$ (*10 <sup>6</sup> ) m <sup>3</sup> /mole				
		35°C	40°C	45°C	50°C	55°C
Water + ammonium bromide	0.5	90.950	88.832	88.842	85.686	81.825
	1.0	75.665	73.750	70.887	69.940	67.719
	1.5	70.112	67.415	65.689	65.107	63.630
	2.0	65.685	62.836	61.231	59.929	59.396
	2.5	62.748	60.356	59.174	57.405	56.738
	3.0	59.490	57.748	56.572	56.428	54.580
	3.1	59.268	57.663	56.441	56.422	54.310

hydration number values [19]. Regarding temperature, a slight decrease in molar hydration number may be due to the disruption of the hydration sphere around ions.

The computed values of the above parameters for ammonium bromide solutions are provided in Table 2 and Fig. 2. Apparent molal volume is observed to increase with ammonium bromide at all temperatures in the aqueous solutions. The rate of change of  $\phi_v$  with concentration is more pronounced at lower concentrations than at higher concentrations. Concerning temperature, the values of

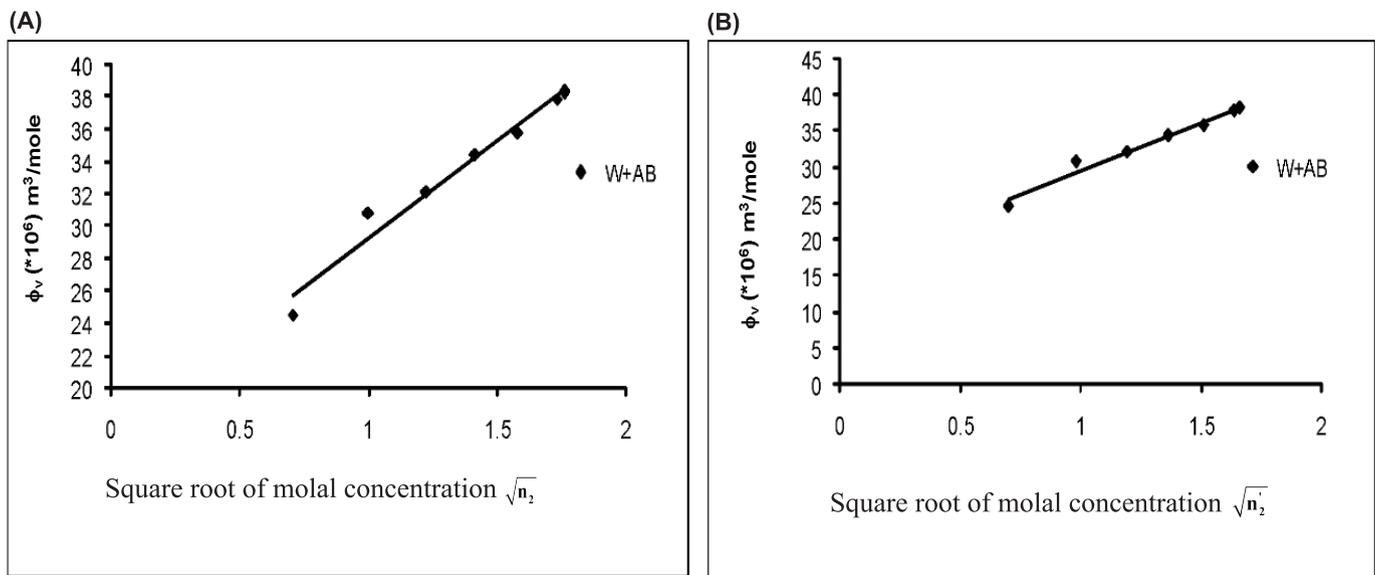
$\phi_v$  increase in the case of ammonium bromide solutions. Apparent molal compressibility values are negative, and this parameter increases with both concentration and temperature. Molar solvated volume decreases with increases in concentration and temperature, mirroring the trends seen with the hydration number. It is evident that the hydration number significantly influences interactions and, consequently, the value of  $\phi_s$  [20]. In ammonium bromide solutions, the hydration number has a more substantial impact on the molar solvated volume than the apparent molal volume. Thus, molality affects the three allied parameters-apparent molal volume, apparent molal compressibility, and molar solvated volume-predominantly at lower concentrations, while temperature has a minimal effect on these values.



**Fig. 2. (A) Apparent molal volume, (B) apparent molal compressibility, and (C) molar solvated volume of water + ammonium bromide.**

**Table 3. Variation of allied parameters due to hydration with molality at 35°C for water + ammonium bromide.**

Molality	Apparent molal volume $\phi_v$ (*10 <sup>6</sup> ) m <sup>3</sup> /mole				Apparent molal compressibility $\phi_k$ (*10 <sup>14</sup> ) m <sup>3</sup> /N/mole				Molar solvated volume $\phi_s$ (*10 <sup>6</sup> ) m <sup>3</sup> /mole			
	Equations											
	4	5	6	7	8	9	10	11	12	13	14	15
0.5	24.585	23.902	24.78	24.574	-2.874	-2.904	-2.907	-2.875	91.707	90.950	91.713	90.950
1.0	30.833	30.362	31.210	30.838	-1.942	-1.962	-1.980	-1.942	76.579	75.665	76.574	75.665
1.5	32.143	32.058	32.211	32.143	-1.645	-1.648	-1.651	-1.645	70.262	70.112	70.262	70.113
2.0	34.465	34.879	34.538	34.461	-1.352	-1.334	-1.358	-1.352	65.902	65.685	65.834	65.685
2.5	35.836	35.566	36.071	35.840	-1.166	-1.177	-1.182	-1.166	63.136	62.748	63.152	62.748
3.0	37.928	37.745	37.291	37.925	-0.934	-0.942	-0.895	-0.934	58.393	59.490	58.496	59.490
3.1	38.307	37.523	37.974	38.305	-0.908	-0.942	-0.888	-0.908	58.567	59.268	58.756	59.269



**Fig. 3. (A)  $\sqrt{n_2}$  and (B)  $\sqrt{n_2}$  vs.  $\phi_v$  in water at 35°C.**

Validity of three equations for  $\phi_v$ ,  $\phi_k$  and  $\phi_s$  from hydration studies: From the hydration studies, the three parameters  $\phi_v$ ,  $\phi_k$  and  $\phi_s$  are calculated using three sets of equations and their values are compared with those obtained by traditional equations. This comparison, for values at 35°C, is presented in Table 3. It is evident from the table that the values calculated from the three equations are in good agreement with those obtained from the traditional equations. This concordance suggests that the assumptions made by Passynski, Barnartt, and the authors are valid, thereby affirming the correctness of the methodology adopted for deriving these equations.

Variation of three allied parameters  $\phi_v$ ,  $\phi_k$  and  $\phi_s$  parameters with concentration: The variation of the three allied parameters  $\phi_v$ ,  $\phi_k$  and  $\phi_s$  with the square root of molar and molal concentration ( $\sqrt{n_2}$  &  $\sqrt{n_2}$ ) are found to be linear. The variation of  $\phi_v$ ,  $\phi_k$  and  $\phi_s$  and molar solvated volume with concentration are studied and the corresponding plots of  $\phi_v$  vs.  $\sqrt{n_2}$  and  $\sqrt{n_2}$ ,  $\phi_k$  vs.  $\sqrt{n_2}$  and  $\sqrt{n_2}$ ,  $\phi_s$  vs.  $\sqrt{n_2}$  and  $\sqrt{n_2}$  are constructed at 35°C and presented in Figs. 3 to 5. The values of slope, intercept, and correlation coefficient at 35°C in molar and molal scale for the three allied parameters are given in Table 4. The correlation coefficient is found to be 0.99 suggesting that the linear equations provide the best fit, thus indicating their appropriateness for modelling these relationships.

**Table 4. Values of slope, intercept and correlation co-efficient in molar and molal scale for the three allied parameters.**

Values of  $n_{hv}$ ,  $\phi_v^0$ ,  $n'_{hv}$ ,  $\phi_v^{0'}$  and correlation co-efficient  $r$  at 35°C.

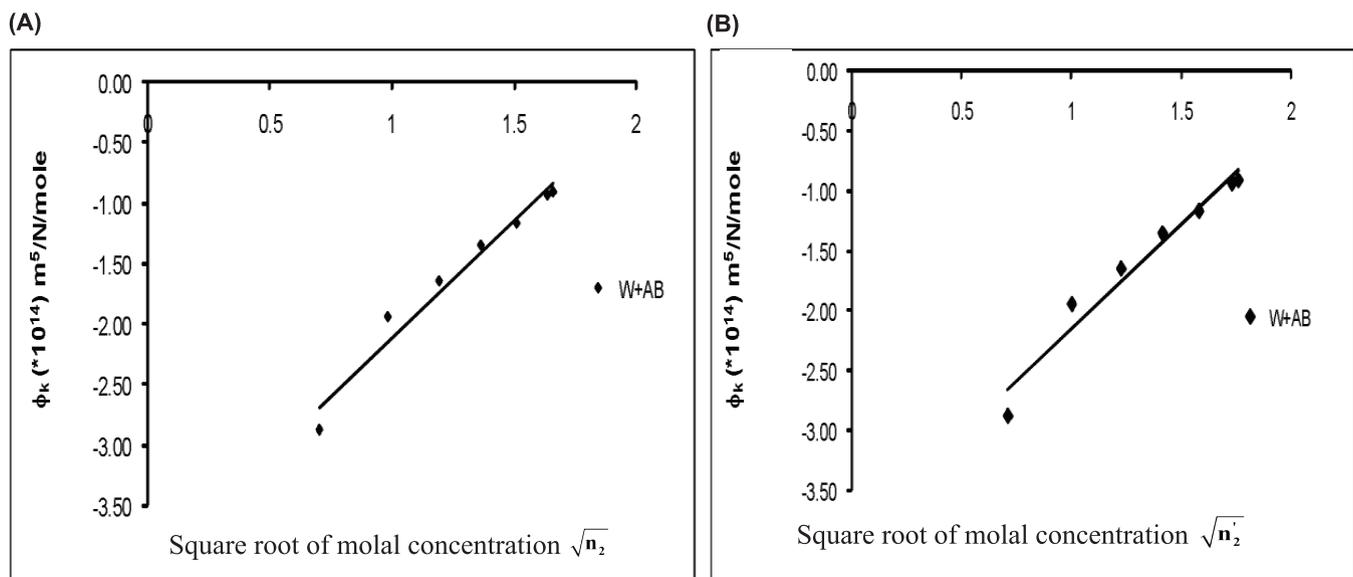
System	Molar scale			Molal scale		
	Slope $n_{hv}$ (*10 <sup>6</sup> m <sup>3</sup> /M <sup>2</sup> )	Intercept $\phi_v^0$ (*10 <sup>6</sup> m <sup>3</sup> /mole)	Correlation co-efficient $r$	Slope $n'_{hv}$ (*10 <sup>6</sup> m <sup>3</sup> /M <sup>2</sup> )	Intercept $\phi_v^{0'}$ (*10 <sup>6</sup> m <sup>3</sup> /mole)	Correlation co-efficient $r$
Water + ammonium bromide	13.255	016.308	0.987	12.0242	017.260	0.985

Values of  $n_{hk}$ ,  $\phi_k^0$ ,  $n'_{hk}$ ,  $\phi_k^{0'}$  and correlation co-efficient  $r$  at 35°C.

System	Molar scale			Molal scale		
	Slope $n_{hk}$ (*10 <sup>14</sup> m <sup>5</sup> /N/M <sup>2</sup> )	Intercept $\phi_k^0$ (*10 <sup>14</sup> m <sup>5</sup> /N/mole)	Correlation co-efficient $r$	Slope $n'_{hk}$ (*10 <sup>14</sup> m <sup>5</sup> /N/M <sup>2</sup> )	Intercept $\phi_k^{0'}$ (*10 <sup>14</sup> m <sup>5</sup> /N/mole)	Correlation co-efficient $r$
Water + ammonium bromide	01.915	-04.022	0.983	01.734	-03.880	0.980

Values of  $n_{hs}$ ,  $\phi_s^0$ ,  $n'_{hs}$ ,  $\phi_s^{0'}$  and correlation co-efficient  $r$  at 35°C.

System	Molar scale			Molal scale		
	Slope $n_{hs}$ (*10 <sup>3</sup> m <sup>3</sup> /M <sup>2</sup> )	Intercept $\phi_s^0$ (*10 <sup>6</sup> m <sup>3</sup> /mole)	Correlation co-efficient $r$	Slope $n'_{hs}$ (*10 <sup>3</sup> m <sup>3</sup> /M <sup>2</sup> )	Intercept $\phi_s^{0'}$ (*10 <sup>6</sup> m <sup>3</sup> /mole)	Correlation co-efficient $r$
Water + ammonium bromide	-030.962	109.151	-0.979	-028.009	106.823	-0.975



**Fig. 4. (A)  $\sqrt{n_2}$ , (B)  $\sqrt{n_2}$  vs.  $\phi_k$  in water at 35°C.**

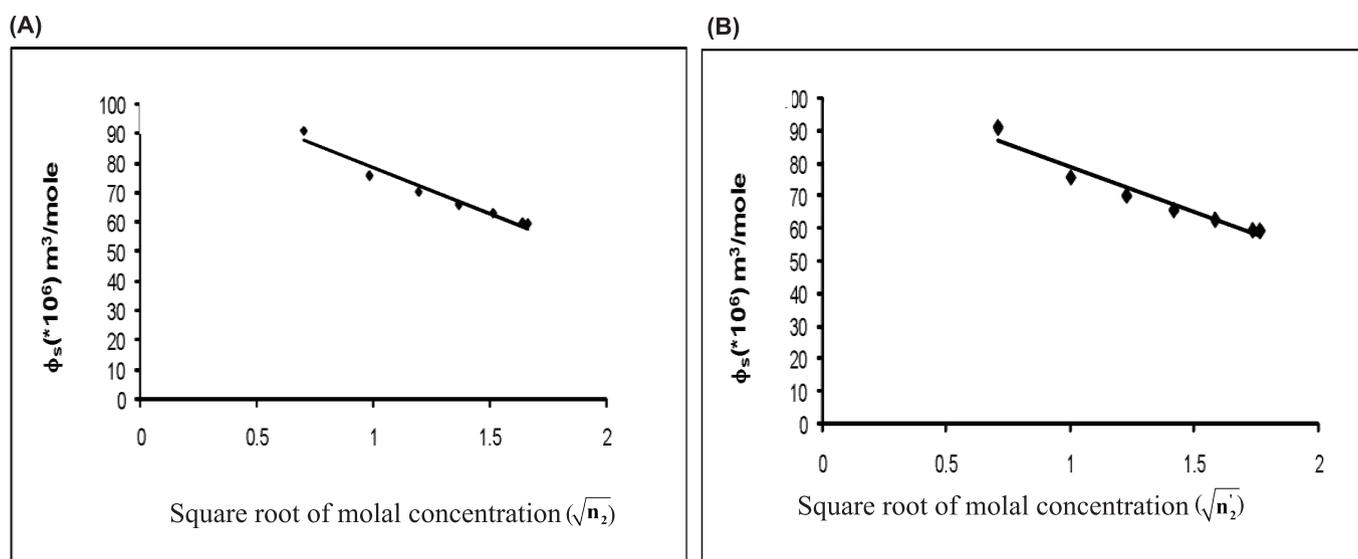


Fig. 5. (A)  $\sqrt{n_2}$ , (B)  $\sqrt{n_2}$  vs.  $\phi_s$  in water at 35°C.

## 5. Conclusions

The hydration number and some related allied parameters for ammonium bromide at various temperatures are reported. Variations in adiabatic compressibility and related parameters against concentration and temperature reveal the existence of molecular association among solute and solvent molecules. The results of the hydration number and their allied parameters indicate the molecular interactions taking place in the solution, and the trends of these parameters are in good agreement with the structure-promoting behaviour of the solutes.

### CRediT author statement

J.H. Rakini Chandrasekaran: Conceptualisation, Data curation, Investigation; S. Nithiyantham: Methodology, Formal analysis, Original draft preparation, Visualisation.

### COMPETING INTERESTS

The authors declare that there is no conflict of interest regarding the publication of this article.

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