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Molecular Interaction Studies of Ammonium Dihydrogen Phosphate in the Aqueous Solutions of Mono, Di and Triprotic Acids – An Ultrasonic Study

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Abstract

Aqueous solutions of mono, di, and triprotic acids were prepared with different molar percentages (say 10%, 20% and 30%). ammonium dihydrogen phosphate is gradually introduced in the aqueous solutions at different concentrations varied from 0.1M to 0.6M at 303K. Densities, viscosities, and velocity of sound of aqueous solutions are measured and they are used to evaluate the other parameters like adiabatic compressibility (β), free length (L_f), internal pressure (π_i), apparent molar volume (φ_V), apparent molar compressibility (φ_K) and relative Association (R_A) of the solution. Experimental results are analyzed based on molecular associations like hydrogen bonding and dipole-dipole interactions between the component molecules, which revealed that the variation of the parameters with the different molar concentrations of both ammonium dihydrogen phosphate and the acids.

Keywords: Aqueous solutions; Molecular association; Hydrogen bonding; Ultrasonic study.

Introduction

Molecular interaction studies among the component molecules in aqueous medium create interest among many researchers [1-3]. In recent, ammonium dihydrogen phosphate has been used widely for crystal growth because of its nonlinear optical property and molecular interaction between the solvents and crystal surfaces [4-5]. In this paper, we investigate the interactions between ammonium dihydrogen phosphate (ADHP) with increasing concentration in the aqueous solutions of acetic acid, tartaric acid, and orthophosphoric acid. The studies revealed the effect of the impurity that is ADHP in the aqueous solutions. Generally, molecular association occurs between the impurity and the solution, because of the anion-cation present. This selectivity of impurity has been attributed to the orientations of the anion ($H_2PO_4^-$) and the cation (NH_4^+) [6] with the carbonyl oxygen of acetic and tartaric acid. Here a selection of aqueous medium is based on mono, di, and triprotic acids. Acetic acid is the simplest carboxylic acids and it contains acetyl group and then it is ready to release an H^+. Tartaric acid is a diprotic organic acid [7]. Orthophosphoric acid is an

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inorganic acid with non-toxicity, rather weak triprotic acid it can dissociate up to thrice, and giving up an H^+ each time, which usually combines with water molecule [8].

Materials and Methods

Materials and Solution preparation

Ammonium dihydrogen phosphate, tartaric acid, acetic acid, and orthophosphoric acid are the chemicals used in the present investigation and they belong to AR grades acquired from E-Merck (India) with the assay of 99.9% and used without purification. Density, viscosity and ultrasonic velocity of freshly prepared solutions of ammonium dihydrogen phosphate at different molalities (i.e. from 0.1M to 0.6M) in aqueous acetic acid, tartaric acid and orthophosphoric acid solutions at different percentages (say 10%, 20% and 30%) are prepared by weight-dilution method using doubled distilled water at 303K.

Experimental

The ultrasonic velocity of freshly prepared solutions of ammonium dihydrogen phosphate in aqueous acetic acid, tartaric acid, and orthophosphoric acid are measured by a single crystal ultrasonic interferometer with a high degree of accuracy operating frequency of 2 MHz (supplied by Mittal Enterprises, New Delhi). Water is circulated around the double-walled sample holder to maintain the experimental temperature (i.e. 303K). The densities of all compounds are measured by a 10 ml specific gravity bottle calibrated with double distilled water and acetone. An Ostwald's viscometer with 10 ml capacity is used for the viscosity measurements of all the compounds. The viscometer is immersed in fresh conductivity water bath that can be operated at the desired temperature. The flow time of water (t_w) and the flow time of solution (t_s) are measured with a digital stopwatch with an accuracy of 0.01s (RACER HS-10w). Density, viscosity and ultrasonic velocity of the solutions have been computed using the standard relation given by the literature [9-11]. The scientific balance is used for weighting with the accuracy of ± 0.1g.

Theoretical Consideration

Various ultrasonic and solutions parameters are computed using the following equations with the measured values of velocity and density data using.

Adiabatic compressibility (β)

$$\beta = (U^2\rho)^{-1} \ldots (N^{-1}\mathrm{m}^2) \tag{1}$$

Where, U is the velocity measured in ms^{-1} and ρ is density measured in kg/m^3.

Free Length (L_f)

$$L_f = K_T\beta^{\frac{1}{2}} \ldots (m) \tag{2}$$

Where, K_T is Jacob's constant and β is the adiabatic compressibility of liquid mixtures measured in $N^{-1}m^2$.

Internal Pressure (π_i)

$$\pi_i = bRT \left[\frac{K\eta}{U}\right]^{\frac{1}{2}} \left[\frac{\rho^{\frac{1}{3}}}{M^{\frac{7}{6}}}\right] \ldots (Pa) \tag{3}$$

Where, T is an absolute temperature in kelvin, R is the universal gas constant, b is the cubic packing fraction factor is assumed to be '2' for all liquid systems, K is constant, η is viscosity of the solution and M is the mass of the solute.

Apparent Molar Compressibility (φ_K)

$$\varphi_K = \frac{1000}{m\rho_0}(\rho_0\beta - \rho\beta_0) + \left[\frac{\beta_0 M}{\rho_0}\right] \ldots (m^5N^{-1}mol^{-1}) \tag{4}$$

Where, m and *M* are the molal concentration and mass of the solute, β and β_0 are the adiabatic compressibilities of the solution and solvent respectively; ρ and ρ_0 are the densities of solution and solvent respectively.

Apparent Molar Volume (φ_v)

$$\varphi_V = \frac{1000}{m\rho_0}(\rho_0 - \rho) + \left[\frac{M}{\rho_0}\right] \ldots (m^3mol^{-1}) \tag{5}$$

Relative Association (R_A)

$$R_A = \frac{\rho}{\rho_0}\left[\frac{U_0}{U}\right]^{\frac{1}{3}} \tag{6}$$

Where, U_0 and U are the velocities of solution and solvent respectively.

Results and Discusion

In this investigation, we have reported the molecular interaction studies between ammonium dihydrogen phosphate and three different aqueous solutions; namely aqueous acetic acid (monoprotic), aqueous tartaric acid (diprotic), aqueous orthophosphoric acid (triprotic). Here the molar percentage of the aqueous medium is varied from 10% to 30%. And molality of the other component ammonium dihydrogen phosphate is varied from 0.1M to 0.6M. The compounds are mixed in the following order; ADHP + aqueous acetic acid, ADHP + aqueous tartaric acid and ADHP + aqueous orthophosphoric acid.

The experimentally obtained values of density, and viscosity and ultrasonic sound velocity of the above

Table 1. Density (ρ), Viscosity (η) and Velocity (U) of Ammonium dihydrogen phosphate in aqueous acetic acid, tartaric acid and orthophosphoric acid at 303K.

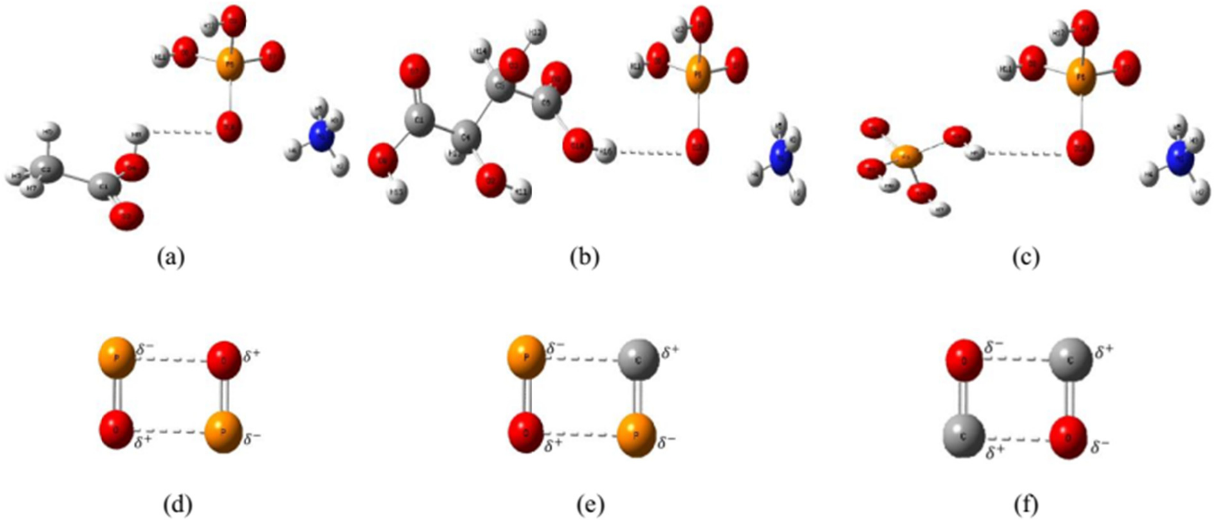
Molality (m) (mol.kg^{-1})	**Density (ρ)** (kgm^{-3})			**Viscosity** (10^{-3}Nsm^{-2})			**Velocity** (ms^{-1})		
	10%	20%	30%	10%	20%	30%	10%	20%	30%
ADHP + Water + Acetic acid									
0.100	1007.2	1013.2	1017.9	1.017	1.210	1.434	1365.7	1378.3	1396.8
0.200	1009.8	1016.2	1021.8	1.060	1.250	1.508	1466.2	1479.3	1507.9
0.300	1012.4	1018.8	1023.9	1.120	1.320	1.561	1507.9	1523.5	1548.9
0.400	1015.8	1021.4	1026.5	1.159	1.391	1.633	1537.0	1549.3	1562.2
0.500	1017.9	1023.9	1029.1	1.190	1.456	1.657	1562.2	1568.1	1573.3
0.600	1020.9	1026.5	1032.5	1.242	1.526	1.680	1569.2	1580.0	1587.3
ADHP + Water + Tartaric acid									
0.100	1020.5	1030.2	1052.6	1.211	1.296	1.648	1529.0	1543.5	1584.5
0.200	1023.5	1034.0	1054.7	1.284	1.363	1.685	1536.8	1557.2	1597.3
0.300	1026.5	1040.0	1056.4	1.325	1.398	1.724	1547.4	1569.6	1619.2
0.400	1028.2	1044.4	1057.7	1.379	1.441	1.795	1553.3	1578.6	1632.7
0.500	1029.1	1049.7	1059.6	1.426	1.483	1.840	1564.4	1586.3	1646.9
0.600	1030.3	1054.2	1062.9	1.496	1.559	1.891	1570.4	1596.9	1662.8
ADHP + Water + Orthophosphoric acid									
0.100	1023.1	1047.9	1102.2	1.205	1.335	2.458	1492.7	1500.0	1564.2
0.200	1026.1	1050.5	1105.7	1.223	1.478	2.559	1519.1	1529.7	1573.7
0.300	1029.5	1053.4	1108.6	1.236	1.555	2.638	1538.2	1546.3	1580.7
0.400	1031.8	1056.4	1110.3	1.267	1.640	2.682	1547.7	1560.4	1593.1
0.500	1034.6	1058.6	1111.8	1.311	1.698	2.774	1554.8	1577.1	1610.1
0.600	1037.2	1059.9	1113.8	1.366	1.724	2.860	1562.7	1589.0	1629.3

three liquid systems at 303K are presented in Table 1. From the measured data, it is observed that the density, viscosity and velocity are increased with increasing molar concentration of both ADHP and the increasing molal concentrations of the aqueous solutions. The density, viscosity and velocity are increased when the concentration of ADHP increases in the aqueous solutions because this serves the possibility of greater interaction between the aqueous solution due to both inter and intramolecular hydrogen bonding. Ammonium dihydrogen phosphate is a dibasic acid since it possesses two replaceable hydrogens, but the replacement is possible only when a suitable base is available to accept these hydrogens. When ADHP dissolves in the water there is a possibility of weak intramolecular hydrogen bonding between two ADHP molecules (O-H…O=P), when ADHP is mixed with aqueous acetic acid there is a possibility of the intermolecular hydrogen bonding (OH…O=C) between two acetic acids because acetic acid itself is a weak acid and exists as dimer.

With increasing the strength of ADHP from 0.1 to 0.6M, density increases because of the increased availability of replaceable hydrogens of ADHP. Also, there is another factor that is the ionic nature of the solvent (water) which reduces the dimerization of the acetic acid but that enhances the interaction between acetic acid and ADHP. In the beginning the possibility for the interaction between the molecules of ADHP itself is almost negligible because of the less strength of ADHP when compared with acetic acid. But with the gradual increase in the strength of ADHP intra and intermolecular hydrogen bonding are possible. And the intermolecular hydrogen bonding between acetic acid and ADHP is shown in Figure 1(a). The density is also found to be increased linearly with the increasing aqueous percentage of acetic acid form 10% to 30%. The same trend is being observed for both viscosity and velocity of the aqueous acetic acid system. The maximum value of density, viscosity, and velocity is found with 30% of aqueous acetic acid and 0.6M of ADHP.

The measured data for the aqueous tartaric acid with ADHP is similar to the aqueous acetic acid with ADHP system, tartaric acid is a diprotic organic acid and has more possibility of intramolecular hydrogen bonding between two tartaric acid molecules OH…O=C). The greater availability of intermolecular hydrogen bonding between aqueous tartaric acid with ADHP is shown in Figure 1(b). That is why the numerical values that are measured for aqueous tartaric acid with ADHP are greater than the acetic acid with ADHP. The density is also found to be increased linearly with the increasing aqueous percentage of tartaric acid form 10% to 30%. The same trend is being observed for both viscosity and velocity of the aqueous tartaric acid system.

The measured data for the aqueous orthophosphoric


(a) (b) (c)

(d) (e) (f)

Figure 1. Intermolecular hydrogen bonding between aqueous (a) acetic acid and ADHP, (b) tartaric acid and ADHP, (c) orthophosphoric acid and ADHP; dipole-dipole interactions between aqueous (d) ADHP and ADHP, orthophosphoric acid and orthophosphoric acid, (e) ADHP and acetic acid, ADHP and aqueous tartaric acid, (f) acetic acid and acetic acid, tartaric acid and tartaric acid

acid with ADHP is similar to the other two aqueous solutions with ADHP. Orthophosphoric acid is a strong tribasic acid, which has three replaceable hydrogens. It can be removed even with the addition of weaker acid. The triprotic dissociation of orthophosphoric acid, the fact that it's conjugate bases. Both ADHP and orthophosphoric acid are inorganic acids having phosphorous as its theme element. The miscibility between these substances can be observed. For orthophosphoric acid, the intramolecular hydrogen bindings are possible between two orthophosphoric acid molecules (OH…O=P). And strong intermolecular hydrogen bondings are also possible between ADHP and orthophosphoric acid molecules, which are shown in Figure 1(c), which leads to dimeric formation. When ADHP and orthophosphoric acid are mixed in suitable proportions, there is a possibility of the formation of weak – binuclear ionic complexes.

From Table 1, it is evident that the concentration of ADHP with aqueous solutions of acetic acid, tartaric acid, and orthophosphoric acids are varying from 0.1 to 0.6 in terms of mole fraction, whereas the concentration of solvent is fixed at 50ml. In all the systems, the values of the density, viscosity and ultrasonic sound velocity increase with increasing mole concentration of ADHP and mono, di and tri protic acids. The linear variation is observed for density, viscosity, and ultrasonic sound velocity, which depends upon the increasing inter and intramolecular hydrogen bonding and dipole-dipole interaction. The maximum value of density, viscosity, and velocity is found with 30% of aqueous tartaric acid and 0.6M of ADHP.

From Table 2, the values of adiabatic compressibility and free length are decreases and the values of internal pressure increase with increasing concentration of aqueous solutions and ADHP. The adiabatic compressibility has shown inverse trend as compared to ultrasonic sound velocity. The variation in adiabatic compressibility can explain the following two factors; (i) Loss of dipolar association and difference in size and shape of the component molecules leads to increase incompressibility and (ii) Dipole-dipole interaction or hydrogen-bonded complex formation between unlike molecules leads to increase in ultrasonic sound velocity and decrease of compressibility. The compressibility data indicate an ordering interaction leading to the formation of the ionic complex. The complex formation in an ionic system is due to ionic association [12].

In this case, the adiabatic compressibility decreases with the increase in concentration of ADHP. And it attains a gradual increase to a higher concentration of the salt (ADHP). It means ion-solvent interaction increases at a given composition [1]. This indicates that there is a significant interaction between the molecules of both ADHP and the aqueous solutions. It can be taken as an indication of the formation of complexes. The addition of interacting molecules breaks up the molecule clustering of the other, releasing several dipoles-dipole interactions is shown in Figure 1(c)-1(f).

The Figure 1(d) represents the dipole-dipole

Table 2. Adiabatic Compressibility (β), Free Length (L_f) and Internal Pressure (π_i) of ammonium dihydrogen phosphate in aqueous acetic acid, tartaric acid and orthophosphoric acid at 303K.

Molality (mol.kg^{-1})	Adiabatic Compressibility (β)			Free Length (L_f)			Internal Pressure (π_i)		
	(kgm^{-3})			(10^{-3}Nsm^{-2})			(ms^{-1})		
	10%	20%	30%	10%	20%	30%	10%	20%	30%
	ADHP + Water + Acetic acid								
0.100	5.323	5.195	5.035	4.554	4.499	4.429	24.983	25.446	25.977
0.200	4.607	4.497	4.304	4.236	4.186	4.095	24.615	24.974	25.667
0.300	4.344	4.229	4.071	4.114	4.059	3.982	24.949	25.292	25.764
0.400	4.167	4.079	3.992	4.029	3.986	3.943	25.296	25.749	26.026
0.500	4.026	3.972	3.926	3.960	3.934	3.911	25.771	26.187	26.350
0.600	3.978	3.902	3.844	3.937	3.899	3.870	26.265	26.710	26.435
	ADHP + Water + Tartaric acid								
0.100	4.192	4.074	3.939	4.041	3.984	3.917	25.463	24.081	23.065
0.200	4.137	3.988	3.867	4.015	3.942	3.882	26.158	24.518	23.374
0.300	4.069	3.913	3.803	3.981	3.904	3.849	26.486	24.656	23.639
0.400	4.008	3.860	3.744	3.952	3.878	3.819	26.914	24.864	23.873
0.500	3.971	3.816	3.698	3.933	3.856	3.796	27.278	25.061	24.067
0.600	3.936	3.759	3.645	3.916	3.827	3.768	27.860	25.509	24.509
	ADHP + Water + Orthophosphoric acid								
0.100	4.387	4.241	3.708	4.134	4.065	3.801	25.693	24.510	22.862
0.200	4.223	4.068	3.652	4.056	3.981	3.772	26.661	25.447	23.429
0.300	4.105	3.970	3.610	3.999	3.933	3.750	27.401	25.874	23.848
0.400	4.046	3.888	3.549	3.970	3.892	3.718	28.051	26.365	24.228
0.500	3.998	3.798	3.469	3.947	3.847	3.676	28.588	26.584	24.462
0.600	3.948	3.737	3.382	3.922	3.815	3.630	29.029	26.909	24.885

Table 3. Apparent Molar Volume (φ_v), Apparent Molar Compressibility (φ_K) and Relative association (R_A) of ammonium dihydrogen phosphate in aqueous acetic acid, tartaric acid and orthophosphoric acid at 303K.

Molality (mol.kg^{-1})	Apparent molar Volume ($-\varphi_V$)			Apparent molar Compressibility ($-\varphi_K$)			Relative Association (R_A)		
	(kgm^{-3})			(10^{-3}Nsm^{-2})					
	10%	20%	30%	10%	20%	30%	10%	20%	30%
	ADHP + Water + Acetic acid								
0.100	302.35	363.72	411.80	1.025	0.568	2.411	1.044	1.047	1.047
0.200	164.41	197.15	225.79	3.137	3.851	4.961	1.022	1.026	1.025
0.300	118.43	140.26	157.64	3.011	3.506	4.120	1.015	1.018	1.018
0.400	97.49	111.81	124.85	2.745	3.038	3.321	1.012	1.015	1.017
0.500	82.27	94.54	103.03	2.501	2.670	2.794	1.009	1.013	1.016
0.600	73.65	83.2	89.83	2.189	2.364	2.476	1.010	1.014	1.016
	ADHP + Water + Tartaric acid								
0.100	438.39	537.61	622.51	10.981	12.659	14.439	1.019	1.025	1.029
0.200	234.48	288.18	327.05	5.841	6.858	7.660	1.020	1.026	1.029
0.300	166.51	203.67	227.88	4.174	4.880	5.370	1.021	1.027	1.030
0.400	129.20	158.10	177.02	3.303	3.820	4.206	1.020	1.027	1.030
0.500	105.18	130.14	143.08	2.727	3.162	3.453	1.019	1.027	1.029
0.600	89.67	111.84	121.14	2.341	2.748	2.966	1.019	1.026	1.028
	ADHP + Water + Orthophosphoric acid								
0.100	464.99	718.66	864.93	9.164	11.907	17.958	1.029	1.053	1.063
0.200	247.78	372.57	450.31	5.477	6.886	9.369	1.026	1.048	1.059
0.300	176.74	258.23	310.05	4.103	4.967	6.429	1.026	1.048	1.057
0.400	138.41	201.31	236.86	3.255	3.969	4.994	1.026	1.047	1.054
0.500	116.43	165.53	190.29	2.728	3.378	4.148	1.027	1.046	1.051
0.600	101.44	140.14	160.1	2.380	2.928	3.605	1.028	1.045	1.047

interaction between ADHP with ADHP, ADHP with aqueous orthophosphoric acid and aqueous orthophosphoric acid with aqueous orthophosphoric acid molecules, Figure 1(e) represents the dipole-dipole interaction between ADHP with aqueous acetic acid and ADHP with aqueous tartaric acid molecules and Figure

1(f) represents the dipole-dipole interaction between aqueous acetic acid with aqueous acetic acid and aqueous tartaric acid with aqueous tartaric acid. The suggested hydrogen-bonded association between the component molecules is due to the greater forces of interaction between solute and solvent molecules. If the interaction is greater then there will be a decrease in the free length in the mixture [13]. It is evident in Table 2.

Intermolecular free length in two-phase mixtures can be used to find the attraction between the selected molecules. An increase in the concentration of ADHP leads to a decrease in the gap between two species (ADHP + aqueous solutions). The intermolecular free length is found to be a major factor in finding the nature of the variation of sound velocity in liquid mixtures. The intermolecular free length decreases with an increase in ultrasonic velocity and this pointed out that, there is a strong interaction between the ion and solvent molecules. This supports the close packing of molecules at higher concentrations. From Table 2 it is also been found that the internal pressure increases with an increase in the concentration of both ADHP and aqueous percentage of acetic acid, tartaric acid, and orthophosphoric acid because the hydrogen replacing the ability of both ADHP and aqueous systems selected for the present study increases gradually. This is due to complex formation between the two-phase mixture in the form of dipole-dipole and hydrogen bonding association.

The vales of the apparent molal volume are all negative over the entire range of molality and increasing aqueous percentage of the acetic acid, tartaric acid, and orthophosphoric acid. The values of apparent molal volume decrease with the increasing concentration of the salt (ADHP). These observations imply that the negative values of apparent molal volume in the two-phase mixture indicate the presence of ion-solvent interaction. The decrease in apparent molal volume is due to ion-solvent, and the negative values point toward electrostrictive solvation of ion [4].

The values of apparent molal compressibility are negative in the entire range of molalities, which are given in Table 3. The values of apparent molal compressibility gradually increase with the increasing concentration of the ADHP, additionally the relative association of aqueous solution, which indicates electrostriction and hydrophilic interaction accruing in these systems, thereby indicating solute-solvent interaction. The breaking up of the solvent molecules by adding solute to it and subsequently the solvation of ions by the free solvent molecules influences the relative association. The former effect results in a decrease while the latter increases the values of the relative association. In this study, the relative association increase with the concentration of aqueous solutions in ADHP suggests that ion-solvent interaction is varied and it is maximum at a higher concentration of ADHP.

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