

## Effect of flocculating agents on solubility behavior of calcium sulfate dihydrate (gypsum) in aqueous sodium chloride system and solution properties at 35 °C

Jignesh Shukla<sup>a</sup>, Tushar J Trivedi<sup>b</sup>, Pankaj Bharmoria<sup>a</sup>  
& Arvind Kumar<sup>a,\*</sup>

<sup>a</sup>Salt and Marine Chemicals Division, CSIR-Central Salt and Marine Chemicals Research Institute, GB Marg, Bhavnagar 364 002, India

Email: arvind@csmcri.org/ mailme\_arvind@yahoo.com

<sup>b</sup>Graduate School of EEWS (Energy Environment Water Sustainability), KAIST, 291 Daehak-ro, Yuseong Gu, Daejeon 305-701, Republic of Korea

Received 17 September 2015; revised and accepted 01 January 2016

The effect of addition of flocculating agents, viz., alum ( $\text{KAl}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$ ) (up to 10 wt%) and polyaluminum chloride ( $[\text{Al}(\text{OH})_a\text{Cl}_b]_n$ ,  $a > 1.05$ ;  $n \sim 15$ ) (up to 6 wt%) on the solubility behavior of calcium sulfate dihydrate (gypsum,  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) in aqueous NaCl solutions has been examined at 35 °C. The solubility of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  decreases with the addition of alum while it increases with the addition of polyaluminum chloride without any significant shift in solubility maximum. Density ( $\rho$ ) and speed of sound ( $u$ ) have also been determined for the quaternary systems ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{alum}/\text{PAC} + \text{H}_2\text{O}$ ) at 35 °C and used to estimate solution isentropic compressibility ( $\kappa_s$ ) and ion-hydration ( $n_h$ ) characteristics. Empirical equations describing the  $s$ ,  $\rho$ ,  $u$  and  $\kappa_s$  as a function of solution composition are presented.

**Keywords:** Solution chemistry, Solubility, Flocculating agents, Gypsum, Density, Speed of sound

Studies on aqueous electrolyte systems are important for understanding ionic equilibrium, ion-solvent and ion-ion interactions.<sup>1,2</sup> Such studies are even more important for developing the processes for recovery of various inorganic salts from the marine resources like sea and subsoil waters and industrial effluents containing different salts. Gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) is one of the important marine salts which is precipitated in large quantities during the solar evaporation of natural brines prior to the production of common salt (NaCl). It finds several applications, for example in cement industry, writing chalk, soil additive, or as a food additive, etc.<sup>3,4</sup> Because of very low solubility of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in water/brines,

the  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  ions present in feed brines precipitate out and form scale in pipes and other production equipment once its saturation limit exceeds the solubility limit.<sup>5</sup> For prevention of scale formation it is necessary to alter the solubility of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  with organic or inorganic additives. Therefore, understanding the effect of various kinds of additives on the solubility behavior of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in brines is of practical importance. Along with the solubility behavior, knowledge of other physical properties of solution such as density, speed of sound and estimated values of compressibility therein is also a necessary prerequisite<sup>6,7</sup> while processing for the industrial applications.

Physical phenomena such as crystallization, phase stability and solubility of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in water and aqueous solutions containing electrolytes have been widely reviewed.<sup>4,8</sup> The solubility behavior of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in brines has been particularly extensively studied in the past.<sup>8-15</sup> We are continuing our research program on measurements and interpretation of aqueous  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  solubility and solution properties in presence of electrolytes and non-electrolytes. Ternary systems ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{H}_2\text{O}$  and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{CaCl}_2 + \text{H}_2\text{O}$ ),<sup>16-19</sup> quaternary systems ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{CaCl}_2 + \text{H}_2\text{O}$ ,  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{MgCl}_2 + \text{H}_2\text{O}$  and  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{H}_2\text{O} + \text{MNO}_3$ , where  $\text{M} = \text{Na}, \text{K}, \text{NH}_4$ )<sup>20-22</sup> and ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{H}_2\text{O} + \text{organic solvents}$ )<sup>23-24</sup> have been investigated in our laboratory in the past few years.

In recent studies on solar salt production from natural brines it has been observed that  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  ion impurities are dramatically reduced on addition of flocculating agents in saturated brines such as alum or PAC.<sup>25,26</sup> Therefore, in continuation of our research program on solubility studies of gypsum in brines, herein we intend to study the effect of alum and polyaluminium chloride on the solubility behavior of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in aqueous NaCl solutions. We also report the data on solution properties such as density and speed of sound for these systems. The primary data have been used to determine solution isentropic compressibility of the solutions.

## Experimental

Flocculating agent, polyaluminum chloride (PAC powder form, LR grade, CAS No. 1327-41-9) was purchased from National Chemical, assay:  $\text{Al}_2\text{O}_3$  content: 28–30 % (measured in our lab 26.52%). Alum (non-ferric) commercial grade and other inorganic salts such as  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ , NaCl, (>99.5 mol%) obtained from SD Fine Chemicals (Mumbai) were used without further purification after drying in an oven at 70 °C. Solutions were prepared by weight, using an analytical balance with a precision of  $\pm 0.0001$  g (Denver Instrument APX-200) in Millipore grade water. Stock solutions were prepared by adding oven dried NaCl to the solutions containing fixed amounts of PAC or alum. A range of solutions with different NaCl and fixed PAC or alum concentrations saturated with  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  were then made by diluting stock solutions with aqueous PAC or alum solutions and adding excess  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ . The resulting solutions were stirred in a thermostatically controlled water bath. After the solutions were stirred with an electrical paddle for about 24 h, liquid samples were withdrawn periodically and analyzed for different ions as described elsewhere.<sup>16</sup> In brief,  $\text{Ca}^{2+}$  and  $\text{Cl}^-$  concentrations were determined volumetrically using standard EDTA and  $\text{AgNO}_3$  solutions respectively with an error of <0.2%, and  $\text{SO}_4^{2-}$  concentration was determined gravimetrically using barium chloride precipitation as per the standard analytic procedure with an estimated error of <0.1%.

The density ( $\rho$ ) of the solutions was measured with an Anton Paar (model DMA 4500) vibrating-tube densimeter with a resolution of  $5 \times 10^{-2}$  kg m<sup>-3</sup> at 35 °C. The densimeter was calibrated with doubly distilled and degassed water, with dry air at atmospheric pressure, and also against the densities of aqueous NaCl solutions.<sup>27</sup> Sample temperature was controlled by a built-in peltier device to within  $\pm 0.03$  K and the accuracy of the measurements was 0.01%. Reproducibility of the results was confirmed by performing at least three measurements for each sample. Speed of sound ( $u$ ) in the salt solutions was measured at 51600 Hz using a concentration analyzer (model 87, SCM Laboratory Sonic Composition Monitor) based on the sing-around technique<sup>28</sup> at 35 °C with a single transducer cell, immersed in a water bath with temperature controlled to  $\pm 0.01$  °C. The analyzer was calibrated by measurements of speeds of sound in water as a reference<sup>28</sup>, with an

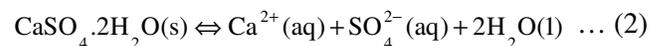
estimated error  $< \pm 0.1$  m·s<sup>-1</sup>. Reproducibility of the results was ensured by performing at least three experiments for each concentration.

## Results and discussion

Experimental results of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  solubility in aqueous-NaCl solutions containing fixed molality of alum or PAC and the solution densities and speed of sounds at 35 °C are given in Table 1. The dependence of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  solubility in NaCl+H<sub>2</sub>O+alum or PAC systems and other solution properties as a function of NaCl concentration has been correlated by means of a polynomial type equation,

$$F(Q) = A_0 + A_1(m\text{NaCl}) + A_2(m\text{NaCl})^2 + A_3(m\text{NaCl})^3 \dots \quad (1)$$

where  $Q$  represents a general measured property (solubility, density, speed of sound) or derived function (isentropic compressibility) and  $m$  is the molality (mol kg<sup>-1</sup>) of NaCl in the solution. The values of the parameters  $A_i$  were evaluated by the method of least squares using non-linear method with all points weighted equally. The parameters  $A_i$  and standard deviations  $\sigma$  are given in Table 2. The solubility pattern of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in pure brines and brines containing different concentration of alum or PAC is compared in Figs 1 and 2 respectively. The solubility of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in brine solutions decreases significantly with increase in alum concentration, whereas in the case of PAC the solubility increases with increase in concentration. The decrease in solubility of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  with the addition of alum may be due to the common ion effect of ( $\text{SO}_4^{2-}$ ). Dissolution of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in water follows the equilibrium:



Excess amount of  $\text{SO}_4^{2-}$  ions will shift the dissolution equilibrium to the left, reducing the concentration of  $\text{Ca}^{2+}$ , and thus effectively reducing the solubility of the solid. The solubility maximum of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  was not altered by the addition of alum and an amount of 10 wt% alum could reduce the solubility higher than 80% with almost linear pattern. In the case of PAC added  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  solutions in brine, interionic forces play a role in enhancing the  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  solubility wherein all the ions in solution, besides possessing tightly-held waters of hydration, tend to attract oppositely-charged ions ("counter-ions") around them. These ion-counterion bodies appear to be almost electrically neutral, which keeps them from interacting with each other (as to

Table 1—Solubility of CaSO<sub>4</sub>·2H<sub>2</sub>O in NaCl+alum/ PAC+H<sub>2</sub>O, solution density ( $\rho$ ) and speed of sound ( $u$ ) at 35 °C

NaCl (mol·kg <sup>-1</sup> )	CaSO <sub>4</sub> (mol kg <sup>-1</sup> )	$\rho$ (kg m <sup>-3</sup> )	$u$ (m s <sup>-1</sup> )	NaCl (mol kg <sup>-1</sup> )	CaSO <sub>4</sub> (mol kg <sup>-1</sup> )	$\rho$ (kg m <sup>-3</sup> )	$u$ (m s <sup>-1</sup> )
1% Alum				3% Alum			
0.0508	0.0223	1009.1	1545.2	0.1009	0.0125	1019.8	1553.8
0.7568	0.0255	1036.0	1578.2	0.7568	0.0165	1041.9	1585.7
1.1604	0.0280	1047.9	1596.8	1.0595	0.0181	1053.3	1598.6
1.6650	0.0305	1063.3	1622.1	1.6144	0.0201	1074.4	1624.8
2.1694	0.0321	1083.3	1643.4	1.9181	0.0215	1082.3	1640.6
2.4721	0.0321	1094.7	1658.8	2.3207	0.0220	1097.1	1659.1
2.9407	0.0318	1110.1	1680.9	2.8757	0.0218	1117.7	1684.3
3.4814	0.0310	1129.5	1707.2	3.2793	0.0204	1133.7	1706.5
4.0361	0.0293	1146.5	1734.2	3.8343	0.0180	1150.7	1730.6
4.4397	0.0275	1162.6	1752.9	3.9856	0.0175	1155.4	1737.9
4.8938	0.0261	1177.7	1775.8	4.6919	0.0145	1182.5	1771.8
6% Alum				10% Alum			
0.0503	0.0115	1040.9	1568.5	0.0727	0.0058	1062.8	1593.2
0.5549	0.0138	1056.5	1589.1	0.5838	0.0060	1075.9	1618.4
1.1099	0.0141	1078.4	1620.5	1.0216	0.0065	1091.5	1637.9
1.4631	0.0143	1089.1	1637.9	1.3621	0.0072	1105.2	1653.6
1.9677	0.0143	1105.4	1659.4	1.8481	0.0080	1121.0	1675.7
2.3712	0.0142	1118.1	1680.4	2.1886	0.0085	1131.2	1688.1
2.7746	0.0141	1133.9	1701.7	2.5770	0.0085	1143.9	1707.7
3.2792	0.0135	1149.3	1724.4	3.0151	0.0082	1157.7	1729.7
3.7332	0.0127	1165.2	1748.3	3.4583	0.0077	1172.3	1745.8
4.0361	0.0119	1174.9	1763.6	3.6790	0.0075	1179.1	1754.2
4.5908	0.0114	1195.7	1786.0	4.2796	0.0070	1199.6	1782.9
1% PAC				3% PAC			
0.0958	0.0406	1002.7	1534.1	0.3392	0.0623	1011.2	1550.3
0.6162	0.0495	1023.5	1558.5	0.7753	0.0668	1028.4	1582.2
1.0660	0.0540	1041.6	1586.1	1.2114	0.0698	1045.3	1600.5
1.5506	0.0565	1060.6	1611.3	1.6475	0.0708	1062.3	1635.4
2.1805	0.0595	1077.9	1641.6	2.0835	0.0708	1078.6	1653.4
2.7135	0.0599	1098.5	1667.6	2.5681	0.0708	1096.1	1687.8
3.2434	0.0599	1117.7	1694.2	3.0042	0.0703	1113.5	1713.2
3.7310	0.0594	1135.8	1718.1	3.4887	0.0679	1131.0	1720.6
4.0702	0.0579	1148.2	1739.1	3.9248	0.0653	1144.9	1756.8
4.4578	0.0562	1163.1	1760.6	4.3125	0.0624	1162.0	1783.4
5.0878	0.0527	1186.8	1793.4	4.6032	0.0608	1172.2	1802.7
6% PAC							
0.0171	0.0651	1019.8	1539.5	2.7926	0.0853	1137.6	1682.6
0.5681	0.0716	1042.4	1563.2	2.9432	0.0847	1143.4	1704.3
1.0130	0.0772	1062.9	1586.2	3.5592	0.0794	1170.4	1726.5
1.5469	0.0816	1085.5	1612.2	4.0640	0.0721	1192.1	1747.7
1.8207	0.0833	1096.4	1638.3	4.3806	0.0655	1204.9	1761.8
2.3819	0.0856	1118.9	1662.9				

Table 2—Parameters  $A_i$  and standard deviations  $\sigma$  of Eq. (1) for the system  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{alum}/\text{PAC} + \text{H}_2\text{O}$  at 35 °C

Additive	$A_0$	$A_1$	$A_2$	$A_3$	$\sigma$
Solubility (mol kg <sup>-1</sup> )					
1% Alum	0.0221	0.0179	0.00571	-0.00311	1.87×10 <sup>-4</sup>
3% Alum	0.0118	0.00715	-0.0017	-8.64×10 <sup>-4</sup>	2.32×10 <sup>-4</sup>
6% Alum	0.0112	0.0064	-0.0056	0.00259	5.87×10 <sup>-5</sup>
10% Alum	0.0059	-0.0014	0.0029	-9.75×10 <sup>-4</sup>	8.44×10 <sup>-5</sup>
1% PAC	0.0397	0.01693	-0.0041	2.54×10 <sup>-4</sup>	6.41×10 <sup>-4</sup>
3% PAC	0.0587	0.01293	-0.0034	2.22×10 <sup>-4</sup>	4.64×10 <sup>-4</sup>
6% PAC	0.0647	0.01299	-0.0004	-5.6×10 <sup>-4</sup>	2.54×10 <sup>-4</sup>
Density, $\rho$ (kg m <sup>-3</sup> )					
1% Alum	1007.8	0034.7	-	-	1.24
3% Alum	1015.8	0035.3	-	-	1.16
6% Alum	1039.0	0033.8	-	-	1.09
10% Alum	1059.2	0032.7	-	-	1.19
1% PAC	1000.9	0036.3	-	-	1.7
3% PAC	0999.0	0037.5	-	-	0.98
6% PAC	1019.0	0042.4	-	-	0.67
Speed of sound, $u$ (m s <sup>-1</sup> )					
1% Alum	1541.90	47.55	-	-	0.92
3% Alum	1549.01	47.46	-	-	0.91
6% Alum	1565.17	48.70	-	-	1.83
10% Alum	1591.93	44.64	-	-	1.58
1% PAC	1528.81	51.62	-	-	2.08
3% PAC	1547.00	58.56	-	-	2.64
6% PAC	1525.08	51.87	-	-	3.05
Isentropic compressibility, $10^{12} \times \kappa_S$ (Pa <sup>-1</sup> )					
1% Alum	409.03	-39.27	1.95	-	1.00
3% Alum	388.02	-37.85	2.11	-	0.63
6% Alum	368.85	-31.05	1.47	-	0.54
10% Alum	404.34	-38.96	1.95	-	1.44
1% PAC	426.37	-42.04	1.95	-	1.61
3% PAC	425.12	-47.01	2.64	-	2.75
6% PAC	416.36	-45.64	2.61	-	2.01

form a precipitate). No alteration in solubility maximum of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  in solutions with added alum may be due to less perturbation of water by the ions furnished by it. In PAC added solutions, the solubility maximum of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  shifted slightly towards the higher salinities and the solutions formed a gel like structure at PAC concentration >6%.

Density ( $\rho$ ) and speed of sound ( $u$ ) values for the system  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{H}_2\text{O} + \text{alum}/\text{PAC}$  at 35 °C as a function of NaCl concentration are reported in Table 1 and are plotted in Figs 3 and 4, respectively. Both,  $\rho$  and  $u$  were found to increase linearly with an increase in NaCl concentration in the solution and also depended upon the type of flocculating agent in

the solution. The  $\rho$  and  $u$  of solutions were higher for the solutions containing either alum or PAC. Solutions containing 6% PAC became highly dense at higher brine concentrations. Similar behavior was also observed for  $u$  values wherein it was quite high in solutions containing 6% PAC. The density and speed of sound largely depends upon how ions affect the structure of neighboring water molecules in the solution. Normally the small ions cause a high degree of electrostatic ordering of neighboring waters. For larger ions, electrostatic effects are smaller, hence water structure is dominated by water-water hydrogen bonding in the first shell around the ion. The high density in 6% PAC containing solutions

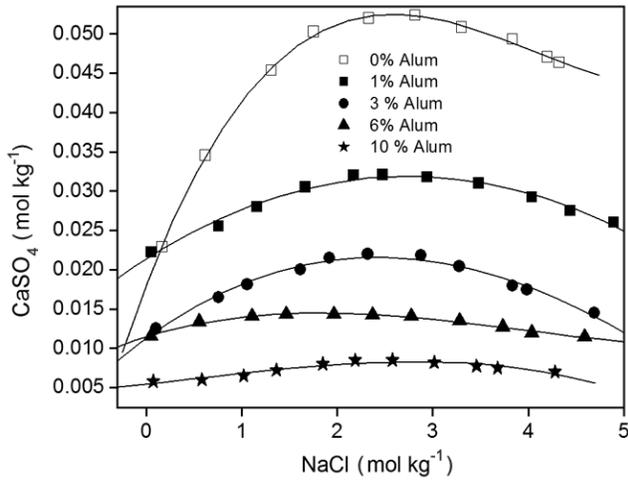


Fig. 1—Comparison of solubility behavior of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  at  $35^\circ\text{C}$  in aqueous  $\text{NaCl}$ -solutions containing coagulant alum at varying concentrations. [Lines are polynomial fit to the experimental data].

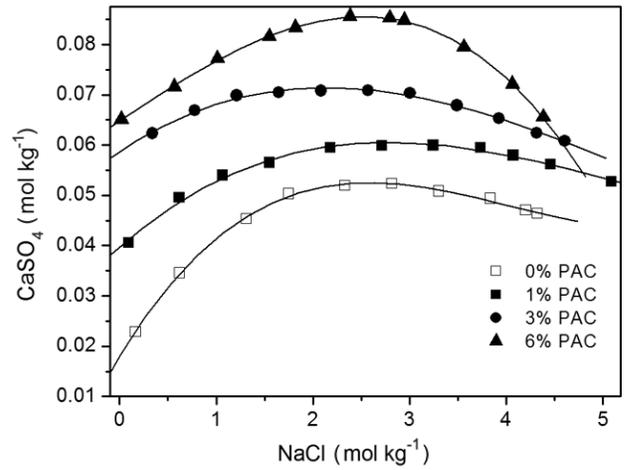


Fig. 2—Comparison of solubility of  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  at  $35^\circ\text{C}$  in aqueous  $\text{NaCl}$ -solutions containing coagulant polyaluminum chloride (PAC) at varying concentrations. [Lines are polynomial fit to the experimental data].

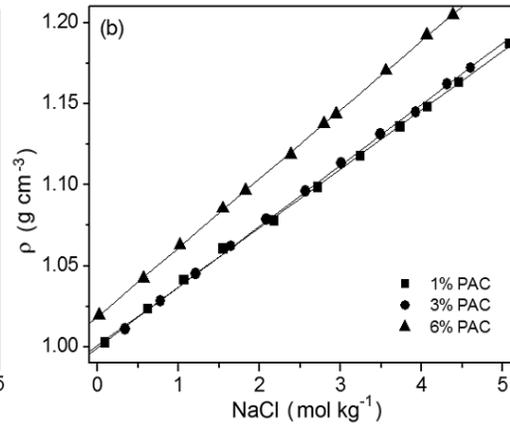
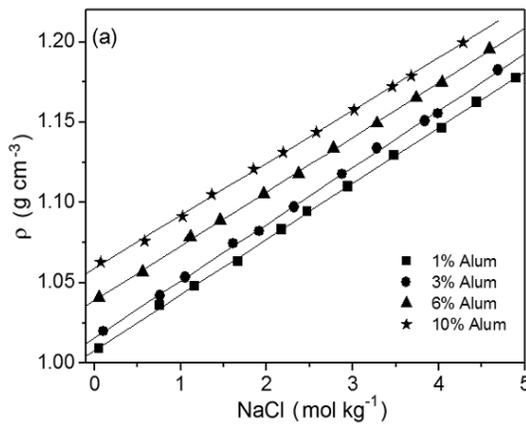


Fig. 3—Density of aqueous- $\text{NaCl}$  solutions saturated with  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  and containing varying amounts of (a) alum, and (b) PAC at  $35^\circ\text{C}$ . [Lines are linear fit to the experimental data].

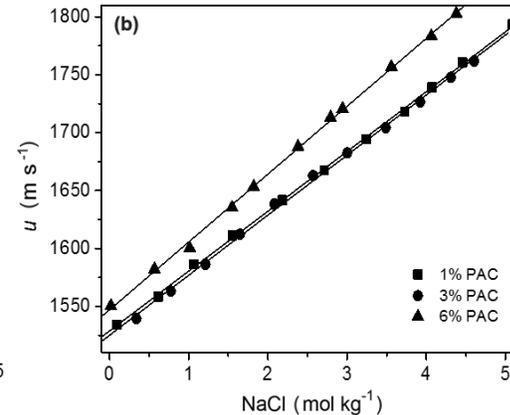
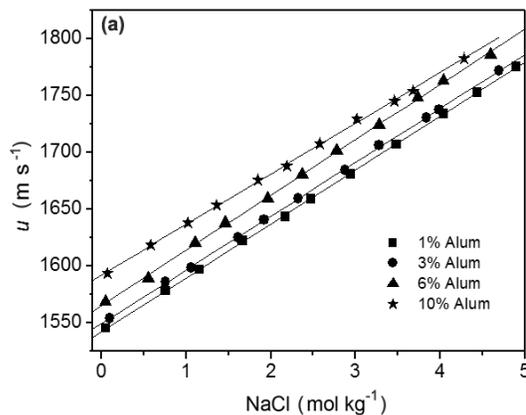


Fig. 4—Speed of sound of aqueous- $\text{NaCl}$  solutions saturated with  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  and containing varying amounts of (a) alum, and (b) PAC at  $35^\circ\text{C}$ . [Lines are linear fit to the experimental data].

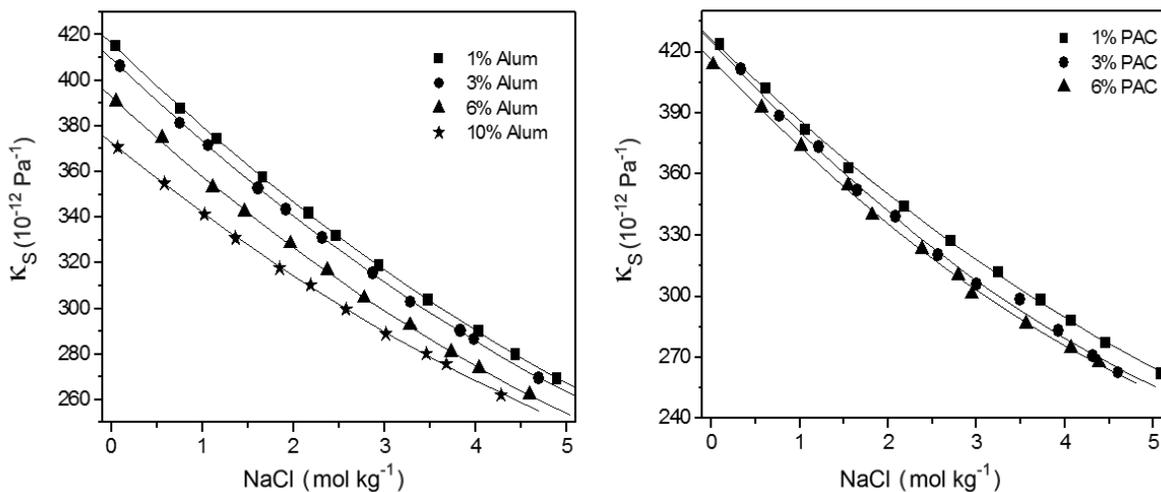


Fig. 5—Isentropic compressibility of aqueous-NaCl solutions saturated with  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  and containing varying amounts of (a) alum, and (b) PAC at  $35^\circ\text{C}$ . [Lines are polynomial fit to the experimental data].

may be due to breaking of normal water structure and formation of dense ion solvated networked structure. The composition dependence of  $\rho$  and  $u$  has been fitted to Eq. 1. The parameters  $A_i$  and standard deviations  $\sigma$  are given in Table 2. Assuming negligible ultrasonic absorption under the experimental conditions, we derived the solution isentropic compressibility  $\kappa_S$  from the  $u$  and  $\rho$  values by using the Newton–Laplace equation,

$$\kappa_S = 1/u^2 \rho \quad \dots (3)$$

Figure 5 show a comparison of  $\kappa_S$  of the  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{NaCl} + \text{H}_2\text{O} + \text{alum/PAC}$  system. The composition dependence of  $\kappa_S$  has been fitted to Eq. 1, and the parameters and standard deviations are provided in Table 2. Addition of alum/PAC in water decreased the solution compressibility in general. The  $\kappa_S$  decreased with the increase of alum/PAC concentration. When compared at the same concentration of alum or PAC in the solution, the solutions having alum were found less compressible. The compressibility of aqueous electrolyte solutions comprises configurational as well as vibrational effects with the former effect playing a dominant role in dilute solutions.<sup>29</sup> At higher concentrations the isentropic compressibility is largely determined by the compression of the hydration shell of the ions. Therefore, it can be assumed that alum perturbs the bulk water structure and weakens its hydrogen bonding to a larger extent as compared with PAC, thereby making the solutions more rigid.

### Acknowledgement

Council of Scientific & Industrial Research, New Delhi, India, is gratefully acknowledged for supporting the research under Chemical Science Cluster (CSC0203) project of Twelfth Five Year Plan.

### References

- Robinson R A & Stokes R H, *Electrolyte Solutions*, 2nd Edn, (Butterworths, London) 1970.
- Jenkins H D B & Marcus Y, *Chem Rev*, 95 (1995) 2695.
- Singh N B & Middendorf B, *Prog Cryst Growth Charact Mater*, 53 (2007) 57.
- Freyer D & Voigt W, *Monatsh Chem*, 134 (2003) 693.
- Hill A E & Wills J H, *J Am Chem Soc*, 60 (1938) 1647.
- Bharmoria P, Gupta H, Mohandas V P, Ghosh P K & Kumar A, *J Phys Chem B*, 116 (2012) 11712.
- Bharmoria P, Gehlot P S, Gupta H & Kumar A, *J Phys Chem B*, 118 (2014) 12734.
- Raju K U G & Atkinson G, *J Chem Eng Data*, 35 (1991) 361.
- Cameron F K & Seidell A, *J Phys Chem*, 5 (1901) 643.
- Bock E, *Can J Chem*, 39 (1961) 1746.
- Marshall W L & Slusher R, *J Phys Chem*, 70 (1966) 4015.
- Marshall W L & Slusher R, *J Chem Eng Data*, 13 (1968) 83.
- Furby E, Glueckauf E & McDonald L A, *Desalination*, 4 (1968) 264.
- Susarla V R K S & Sherwood D, *J Indian Chem Soc*, (1997), 24.
- Ostroff A G & Metler A V, *J Chem Eng Data*, 11 (1966) 346.
- Kumar A, Mohandas V P, Susarla V R K S & Ghosh P K, *J Sol Chem*, 33 (2004) 995.
- Kumar A, Mohandas V P, Sanghavi R & Ghosh P K, *J Sol Chem* 34 (2005) 333.
- Kumar A, Sanghavi R & Mohandas V P, *J Sol Chem*, 35 (2006) 1515.
- Shukla J, Mohandas V P & Kumar A, *J Chem Eng Data*, 53 (2008) 2797.

- 20 Kumar A, Sanghavi R & Mohandas V P, *J Chem Eng Data*, 52 (2007) 902.
- 21 Kumar A, Shukla J, Dangar Y & Mohandas V P, *J Chem Eng Data*, 55 (2010) 1675.
- 22 Trivedi T J, Shukla J & Kumar A, *J Chem Eng Data*, 59 (2014) 832.
- 23 Kaur M, Rao K S, Singh T, Mohandas V P & Kumar A, *J Chem Eng Data*, 55 (2010) 4704.
- 24 Trivedi T J, Pandya P & Kumar A, *J Chem Eng Data* 58 (2013) 773.
- 25 Kumar A, Mukhopadhyay I, Ghosh P K, Mohandas V P, Shukla J & Sanghavi R J, *US Pat*, (2011) US 8, 021, 442, B2.
- 26 Mukhopadhyay I, Ghosh P K, Mohandas V P, Hamidani A U, Susarala, V R K S & Sanghavi R J, *US Pat*, (2012) US8282690 B2 and EP1928569 B1.
- 27 Surdo A L, Alzola E M & Millero F J, *J Chem Thermodyn*, 14 (1982) 649.
- 28 Del Grosso V A & Mader C W, *J Acoust Soc Am*, 52 (1972) 1442.
- 29 Sile W M, Donfor A R & Litovitz T A, *J Chem Phys* 44 (1966) 3712.

