Effect of Ethylene Glycol and Its Derivatives on the Solubility Behavior of $CaSO_4 \cdot 2H_2O$ in the Aqueous NaCl System and Physicochemical Solution Properties at 35 °C

Manjinder Kaur, K. Srinivasa Rao, Tejwant Singh, V. P. Mohandas, and Arvind Kumar*

Salt and Marine Chemicals Division, Central Salt and Marine Chemicals Research Institute, Council of Scientific and Industrial Research (CSIR), G. B. Marg, Bhavnagar 364002, India

The effect of addition of ethylene glycol (EG), ethylene glycol monomethyl ether (EGMME), and ethylene glycol dimethyl ether (EGDME) on the solubility behavior of $CaSO_4 \cdot 2H_2O$ in aqueous NaCl solutions has been examined at 35 °C. Addition of EG/EG derivatives reduced the solubility of $CaSO_4 \cdot 2H_2O$ in general. Replacement of the hydroxyl group of EG by the methoxy group decreased the solubility of $CaSO_4 \cdot 2H_2O$ in the order: EGDME > EGMME > EG. We also measured precise density (ρ) and speed of sound (u) data for the quaternary systems $CaSO_4 \cdot 2H_2O + NaCl + H_2O$ + EG/EG derivatives at 35 °C. Measurements of speed of sound and density have been used to determine the solution isentropic compressibility (κ_S). Empirical equations describing the solubility, density, speed of sound, and isentropic compressibility in these systems are presented.

Introduction

Accurate and reliable data on solubility and physicochemical solution properties of aqueous multicomponent electrolyte systems are necessary for many industrial processes. Such studies are important for understanding ionic equilibria and ion−solvent and ion−ion interactions in natural waters.[1,2] We are continuing our research program on aqueous electrolyte solutions saturated with $CaSO_4 \cdot 2H_2O$, which is a predominant sparingly soluble electrolyte present in seawater and industrial water systems. It precipitates and forms scale once its saturation limit exceeds a certain threshold limit. Therefore, it is quite important to have accurate data on solubility and other physical properties of systems where $CaSO_4 \cdot 2H_2O$ is an important constituent. In the past a lot of research work has been done on the solubility related problems of aqueous sodium chloride systems saturated with gypsum.[3−6] Research papers of Daniela and Wolfgang and Raju and Atkinson provide a detailed review of the crystallization, phase stability, and solubility of $CaSO_4 \cdot 2H_2O$ in aqueous solution as well as in aqueous solutions of different electrolytes.[7,8] Recently we studied a number of physicochemical properties for the ternary systems ($CaSO_4 \cdot 2H_2O + NaCl + H_2O$ and $CaSO_4 \cdot 2H_2O + CaCl_2 + H_2O$) and quaternary systems ($CaSO_4 \cdot 2H_2O + NaCl + CaCl_2 + H_2O$ and $CaSO_4 \cdot 2H_2O + NaCl + MgCl_2 + H_2O$) and have also examined the effect of pH on the solubility of $CaSO_4 \cdot 2H_2O$ in the system $CaSO_4 \cdot 2H_2O + NaCl + H_2O$.[9−14]

In continuation of the ongoing research work herein we have investigated the effect of organic solvents (ethylene glycol and its derivatives) on the solubility behavior of $CaSO_4 \cdot 2H_2O$ in brines. The investigations on the solubility of inorganic salts and solution properties in mixed water−organic solvent are useful in drowning out precipitation of soluble salts at ambient temperature and obtaining crystals of high purity, desired size, and morphology.[15−19] Ethylene glycol (EG) and its derivates are widely used as solvents and solubilizing agents in many industrial applications.[20−23] EG/EG derivatives can be very effective antisolvents for crystallization of salts from aqueous solutions because of their tendency to bind the water through intermolecular hydrogen bonding.[24−26] There are hardly any investigations on the effect ethylene glycol and its derivatives on the solubility behavior of $CaSO_4 \cdot 2H_2O$ in brines in the literature.[27] Therefore, the aim of the present work is to provide accurate solubility data of gypsum and to provide solution physical properties as an aid toward the assessment of the potential drowning out precipitation using ethylene glycol and its derivatives as a separation technique.

Experimental Section

$CaSO_4 \cdot 2H_2O$ and NaCl (> 99.5 % by mol) obtained from SD Fine Chemicals (Bombay) were used after drying in an oven at 70 °C without further purification. Ethylene glycol (> 99 % by mol) and ethylene glycol monomethyl ether (> 99.5 % by mol) were obtained from Merck KGaA, Germany. Ethylene glycol dimethyl ether (> 99 mol % by mol) was procured from Merck-Schuchardt. All of the organic liquids were used after drying over the 0.4 nm molecular sieves and under vacuum at ambient conditions. Solutions were prepared by weight, using an analytical balance with a precision of ± 0.0001 g (Denver Instrument APX-200) in Millipore grade water. Stock solutions were prepared by adding oven-dried NaCl to the aqueous solutions containing fixed amounts of organic solvent. A range of solutions of different NaCl concentrations saturated with $CaSO_4 \cdot 2H_2O$ were then prepared by diluting the stock solutions of sodium chloride with an aqueous solution of organic solvents and adding excess $CaSO_4 \cdot 2H_2O$. 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.[9] In brief, Ca^{2+} and Cl^- concentrations were determined volumetrically using standard

---

* Corresponding author. E-mail: mailme_arvind@yahoo.com; arvind@csmcri.org.

10.1021/je100336q

Table 1. Molal Solubilities of $CaSO_4 \cdot 2H_2O$ in Aqueous−Organic Solvent−NaCl Solutions, Solution Density ρ, and Speed of Sound u at 35 °C

NaCl	$CaSO_4$	ρ	u	NaCl	$CaSO_4$	ρ	u
$mol \cdot kg^{-1}$	$mol \cdot kg^{-1}$	$kg \cdot m^{-3}$	$m \cdot s^{-1}$	$mol \cdot kg^{-1}$	$mol \cdot kg^{-1}$	$kg \cdot m^{-3}$	$m \cdot s^{-1}$
	EG 5 %				EG 10 %		
0	0.0106	1002.4	1541.7	0	0.0101	1011.7	1566.0
0.4871	0.0301	1019.4	1564.8	0.2925	0.0218	1025.7	1583.1
0.8287	0.0380	1032.4	1585.3	0.7800	0.0326	1043.4	1607.8
1.3651	0.0437	1051.7	1610.8	1.2675	0.0393	1059.6	1628.7
1.7053	0.0467	1067.2	1630.6	1.7538	0.0437	1075.5	1649.8
2.1077	0.0499	1082.3	1652.0	2.2410	0.0461	1090.2	1670.5
2.4376	0.0507	1095.1	1670.3	2.7778	0.0474	1106.6	1693.1
2.9299	0.0505	1109.7	1693.1	3.2154	0.0475	1122.5	1713.8
3.3077	0.0496	1124.7	1709.7	3.7026	0.0461	1137.4	1736.6
3.7812	0.0481	1139.3	1729.5	4.1915	0.0442	1154.0	1754.7
3.9709	0.0472	1146.0	1737.9	4.6786	0.0418	1168.2	1773.2
	EG 15 %				EGMME 5 %		
0	0.0087	1020.0	1581.1	0	0.0101	996.2	1542.1
0.3897	0.0226	1033.6	1597.5	0.4431	0.0231	1013.8	1566.4
0.8769	0.0331	1050.3	1623.4	0.8369	0.0325	1029.4	1590.6
1.3641	0.0387	1066.2	1645.0	1.3294	0.0388	1046.3	1610.8
1.8513	0.0431	1080.8	1664.7	1.7231	0.0424	1061.5	1632.2
2.3402	0.0445	1097.2	1687.2	2.2154	0.0449	1076.9	1654.3
2.8274	0.0456	1112.4	1707.0	2.6581	0.0459	1091.5	1674.0
3.3145	0.0449	1126.3	1725.6	3.0769	0.0458	1106.1	1693.8
3.8017	0.0443	1140.1	1743.8	3.2479	0.0453	1111.8	1703.4
4.2906	0.0426	1156.3	1764.5	3.4465	0.0445	1117.6	1714.8
4.7692	0.0411	1170.0	1781.9	4.1846	0.0418	1142.5	1747.1
	EGMME 10 %				EGMME 15 %		
0	0.0096	996.5	1570.5	0	0.0093	997.9	1588.9
0.2420	0.0156	1007.9	1584.2	0.4185	0.0176	1011.8	1607.8
0.4431	0.0216	1014.1	1595.5	0.9350	0.0251	1030.2	1632.3
1.0338	0.0301	1034.4	1624.1	1.4034	0.0301	1044.9	1650.0
1.6248	0.0350	1053.7	1647.8	1.8462	0.0326	1056.3	1666.9
2.2085	0.0379	1072.2	1675.4	2.3128	0.0350	1071.9	1687.2
2.7573	0.0384	1092.5	1699.1	2.8803	0.0358	1091.7	1711.6
3.2000	0.0379	1107.2	1718.3	3.5692	0.0352	1114.6	1739.3
3.6427	0.0368	1122.1	1738.6	3.8393	0.0348	1122.7	1751.2
4.0376	0.0354	1133.4	1756.3	4.3077	0.0330	1138.8	1768.7
4.4803	0.0338	1148.5	1773.7	4.6034	0.0320	1144.4	1777.5
	EGDME 5 %				EGDME 10 %		
0	0.0083	993.2	1541.6	0	0.0092	993.2	1567.4
0.4586	0.0224	1012.7	1566.9	0.3379	0.0185	1007.9	1585.6
0.8932	0.0301	1028.9	1591.6	0.8208	0.0255	1024.9	1608.3
1.3279	0.0375	1043.7	1610.4	1.3038	0.0305	1042.4	1629.2
1.8103	0.0403	1062.6	1636.6	1.7863	0.0337	1059.3	1652.1
2.2684	0.0433	1077.1	1656.1	2.2684	0.0353	1075.8	1674.4
2.6547	0.0442	1092.2	1674.8	2.7521	0.0356	1091.9	1694.8
3.2581	0.0441	1112.9	1700.1	3.3880	0.0357	1113.6	1719.9
3.5966	0.0437	1123.8	1717.7	3.7179	0.0349	1123.8	1731.7
4.1607	0.0422	1142.4	1740.5	4.2000	0.0342	1139.1	1752.3
4.5624	0.0410	1154.4	1756.0	4.6838	0.0327	1153.8	1771.5
			EGDME 15 %				
0	0.0088	989.34	1577.3	2.8769	0.0320	1089.5	1703.8
0.6940	0.0169	1014.2	1606.3	3.3231	0.0329	1104.8	1720.9
1.0615	0.0209	1027.7	1623.5	3.8684	0.0325	1124.7	1743.7
1.5863	0.0256	1046.5	1648.5	4.0667	0.0323	1130.7	1745.0
2.0820	0.0287	1060.8	1669.7	4.8103	0.0311	1154.3	1776.2
2.3299	0.0300	1071.2	1681.4				

ethylenediaminetetraacetic acid (EDTA) and $AgNO_3$ solutions, respectively. The SO_4^{2-} concentration was determined gravimetrically using barium chloride as a precipitating agent.

The density (ρ) of the solutions was measured with an Anton Paar (model DMA 4500) vibrating-tube densimeter with a resolution of $5 \cdot 10^{-2}$ $kg \cdot m^{-3}$ 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 NaCl(aq),[28] with an accuracy of 0.01 %. The temperature of the apparatus was controlled to within ± 0.03 K by a built-in peltier device. The reproducibility of the results was confirmed by performing at least three measurements for each sample.

The speed of sound (u) in the salt solutions was measured at 51 600 Hz using a concentration analyzer (model 87, SCM Laboratory Sonic composition monitor) based on the sing-around technique[29] at 35 °C with a single transducer cell, immersed in a water bath with temperature controlled to ± 0.01 °C. The analyzer was calibrated by measurements of speeds of sound in water as a reference, and the error was estimated to be less than ± 0.1 $m \cdot s^{-1}$.[29] Measurements were carried out in a specially designed sample jar of low volume capacity. Sample jars were provided with an airtight Teflon covering to keep the samples moisture free during measurements. To check the reproducibility of the results, not less than three experiments were performed for each concentration.

Table 2. Parameters A_i and Standard Deviations σ of Equation 1 for the System $CaSO_4 \cdot 2H_2O$ + NaCl + $MgCl_2$ at 35 °C

organic solvent	A_0	A_1	A_2	A_3	σ
		Solubility/mol·kg^{-1}			
EG 5 %	0.0119	0.0397	−0.0131	0.0014	0.0014
EG 10 %	0.0116	0.0322	−0.0091	0.0008	0.0010
EG 15 %	0.0099	0.0330	−0.0096	0.0009	0.0010
EGMME 5 %	0.0104	0.0325	−0.0093	0.0008	0.0005
EGMME 10 %	0.0101	0.0264	−0.0077	0.0007	0.0006
EGMME 15 %	0.0094	0.0212	−0.0052	0.0004	0.0002
EGDME 5 %	0.0089	0.0313	−0.0088	0.0008	0.0007
EGDME 10 %	0.0101	0.0235	−0.0068	0.0006	0.0007
EGDME 15 %	0.0086	0.0140	−0.0022	0.0006	0.0003
		Density, ρ/kg·m^{-3}			
EG 5 %	1003.0	36.5			1.9
EG 10 %	1016.0	32.9			1.9
EG 15 %	1022.3	31.3			1.4
EGMME 5 %	999.8	34.2			2.2
EGMME 10 %	999.1	33.3			1.6
EGMME 15 %	998.4	32.4			1.9
EGDME 5 %	996.7	35.2			1.9
EGDME 10 %	996.7	34.1			1.9
EGDME 15 %	990.5	34.4			1.1
		Speed of Sound, u/m·s^{-1}			
EG 5 %	1539.3	56.8	−1.6		1.9
EG 10 %	1568.1	48.4	−1.0		1.7
EG 15 %	1580.8	48.1	−1.3		1.2
EGMME 5 %	1543.9	51.9	−0.8		2.0
EGMME 10 %	1572.6	48.3	−0.7		1.5
EGMME 15 %	1589.4	44.2	−0.6		1.5
EGDME 5 %	1542.4	54.2	−1.6		1.3
EGDME 10 %	1568.2	49.2	−1.3		1.1
EGDME 15 %	1575.4	48.0	−1.3		1.3
		Isentropic Compressibility, $10^{12} \cdot \kappa_S$/Pa^{-1}			
EG 5 %	420.9	−45.2	3.0		0.9
EG 10 %	400.6	−37.2	2.1		1.4
EG 15 %	391.7	−35.1	2.0		0.9
EGMME 5 %	420.5	−43.0	2.7		1.3
EGMME 10 %	404.6	−36.8	1.8		1.3
EGMME 15 %	396.2	−34.0	1.7		1.1
EGDME 5 %	422.2	−43.7	2.8		1.1
EGDME 10 %	408.4	−38.8	2.3		1.0
EGDME 15 %	406.5	−37.4	2.1		0.5

Results and Discussion

Experimental results of solubility and solution properties (density and speed of sound) of $CaSO_4 \cdot 2H_2O$ in aqueous−organic solvent−NaCl solutions at 35 °C are given in Table 1. The composition dependence of $CaSO_4 \cdot 2H_2O$ solubility and other solution properties in NaCl + H_2O + EG/EG derivatives systems was correlated by means of a polynomial-type equation,

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

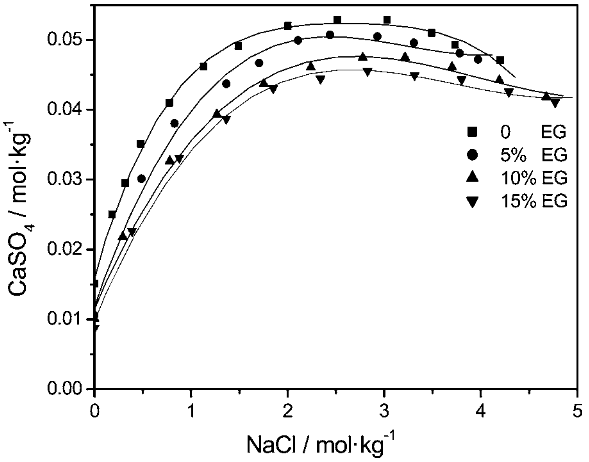

Figure 1. Solubility of $CaSO_4 \cdot 2H_2O$ in aqueous NaCl solutions at 35 °C having different concentrations of ethylene glycol (EG). Lines are a polynomial fit to the experimental data.

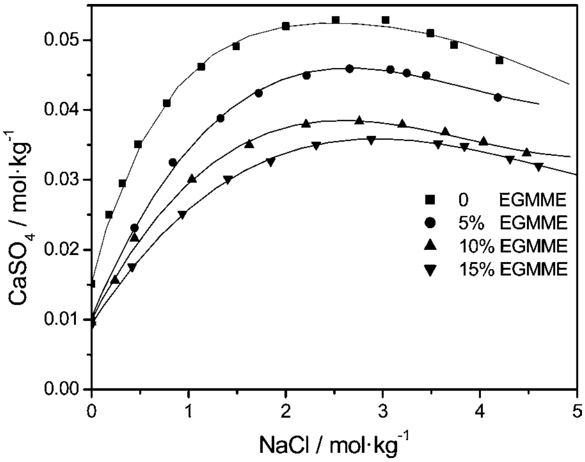

Figure 2. Solubility of $CaSO_4 \cdot 2H_2O$ in aqueous NaCl solutions at 35 °C having different concentrations of ethylene glycol monomethyl ether (EGMME). Lines are a polynomial fit to the experimental data.

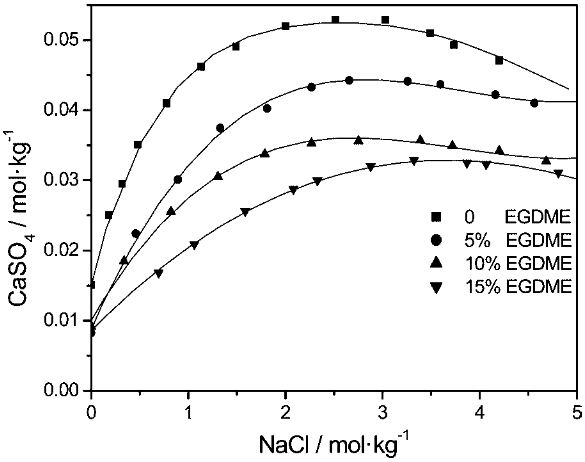

Figure 3. Solubility of $CaSO_4 \cdot 2H_2O$ in aqueous NaCl solutions at 35 °C having different concentrations of ethylene glycol dimethyl ether (EGDME). Lines are a polynomial fit to the experimental data.

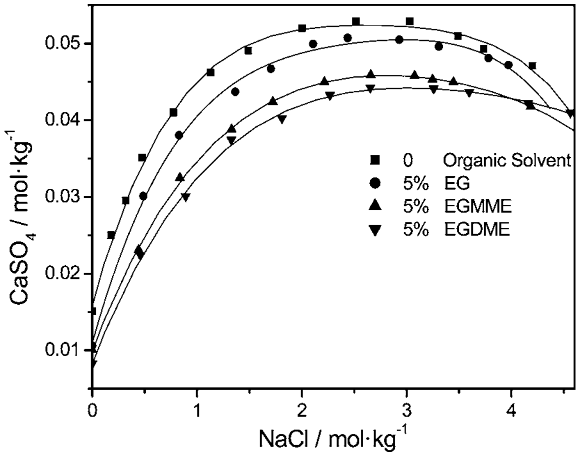

Figure 4. Comparison of the solubility of $CaSO_4 \cdot 2H_2O$ in aqueous NaCl solutions at 35 °C having similar concentrations of EG/EG derivatives. Lines are a polynomial fit to the experimental data.

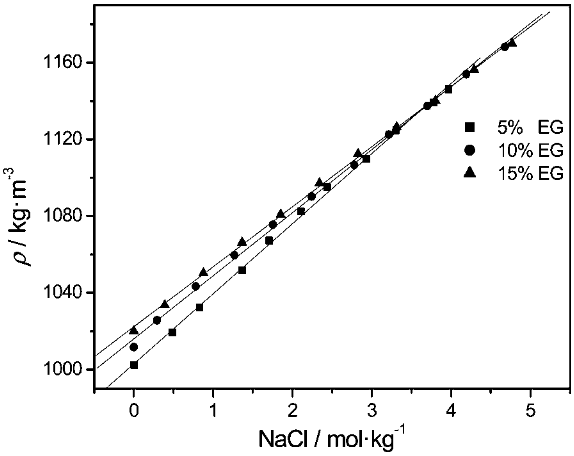

Figure 5. Density of aqueous NaCl solutions saturated with $CaSO_4 \cdot 2H_2O$ having different concentrations of ethylene glycol (EG) at 35 °C. Lines are a linear fit to the experimental data.

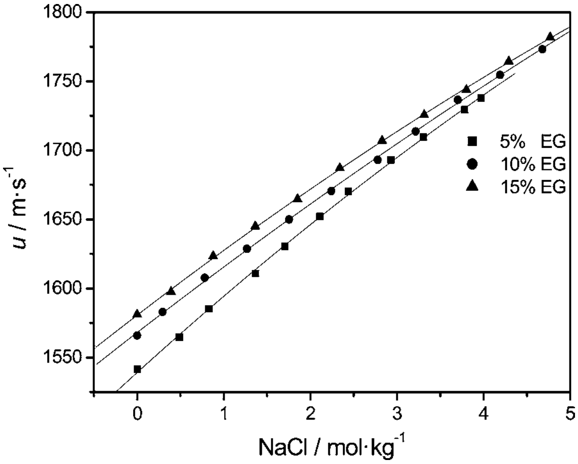

Figure 6. Speed of sound in aqueous NaCl solutions saturated with $CaSO_4 \cdot 2H_2O$ having different concentrations of ethylene glycol (EG) at 35 °C. Lines are a polynomial fit to the experimental data.

where Q represents a general measured property (solubility, density, speed of sound) or derived function (isentropic compressibility) and m is the concentration (mol·kg^{-1}) of NaCl in the solution. The values of the parameters A_i were evaluated by the method of least-squares using the nonlinear method with all points weighted equally. The parameters A_i and standard deviations σ are given in Table 2. Variation in the solubility of $CaSO_4 \cdot 2H_2O$ with the increasing amount of EG derivatives in aqueous- NaCl solutions is shown in Figures 1 to 3. All of the organic solvents used show an antisolvent effect wherein the solubility decreases with the increase in concentration of organic component in all of the systems. Figure 4 shows a comparison of change in solubility of $CaSO_4 \cdot 2H_2O$ in different systems at an equal concentration of organic solvent (5 % by weight). The replacement of the hydroxyl group of EG by the methoxy group decreased the solubility of $CaSO_4 \cdot 2H_2O$ in the order: EGDME > EGMME > EG. Comparatively stronger hydrogen bonding between EGDME and water molecules[25] is responsible for the lower solubility of $CaSO_4 \cdot 2H_2O$ in the NaCl + H_2O + EGDME system. A lesser decrease in solubility of $CaSO_4 \cdot 2H_2O$ in the NaCl + H_2O + EG system is due to the comparatively weaker hydrogen bonding interactions between EG and water molecules[25] because of the self-associated structure of EG. The addition of organic solvent did not shift the solubility maximum of $CaSO_4 \cdot 2H_2O$.

Density (ρ) and speed of sound (u) values for the system $CaSO_4 \cdot 2H_2O$ + NaCl + H_2O + EG/EG derivatives at 35 °C as a function of NaCl concentration and at different organic solvent concentrations are given in Table 1. The density increases linearly with an increase in electrolyte concentration in the solution. Addition of EG in the solution increased the density of the solution, whereas EGMME or EGDME decreased the solution density. A representative plot of density variation as a function of NaCl concentration in the system $CaSO_4 \cdot 2H_2O$ + NaCl + H_2O + EG at 35 °C is shown in Figure 5. The speed of sound is significantly higher in the systems containing organic solvent when compared to the aqueous salt system. This sharp increase in speed of sound in the water-rich region is due to changes in the water structure as a consequence of interaction between water and EG/EG derivatives.[30] The speed of sound increases monotonically with the increase in NaCl concentration in the solution (Figure 6 shows the variation in speed of sound as a function of NaCl concentration in the system $CaSO_4 \cdot 2H_2O$ + NaCl + H_2O + EG at 35 °C). The composition dependence of density and speed of sound are well-fitted to eq 1. The parameters A_i and standard deviations σ are given in Table 2. Assuming negligible ultrasonic absorption under the experimental conditions, we derived the solution isentropic compressibility κ_S from the u and ρ values by using the Newton–Laplace equation

$$\kappa_S = 1/u^2\rho \qquad (2)$$

Figure 7a–c shows a comparison of κ_S of the $CaSO_4 \cdot 2H_2O$ + NaCl + H_2O + EG/EG derivatives systems. The composition dependence of κ_S is fitted to eq 1, and the parameters and standard deviations are presented in Table 2. The addition of EG/EG derivatives decreases the solution compressibility in general. By increasing the concentration of EG/EGMME in the solution from (5 to 10 and 10 to 15) % the magnitude of

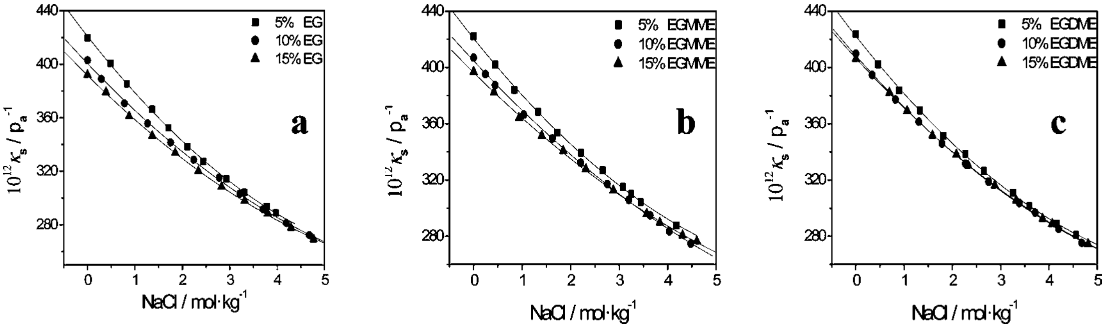

Figure 7. Isentropic compressibility of aqueous NaCl solutions saturated with $CaSO_4 \cdot 2H_2O$ having (a) EG; (b) EGMME; and (c) EGDME at 35 °C. Lines are a polynomial fit to the experimental data.

decrease in compressibility is nearly the same; however, in the system containing EGDME, the decrease in compressibility is very small when going from (10 to 15) %. The increase in electrolyte concentration also decreases the solution compressibility significantly, which may be due to the weakening of hydrogen bonding of bulk water, thereby making the solutions more rigid.

Literature Cited

(1) Robinson, R. A.; Stokes, R. H. *Electrolyte Solutions*, 2nd ed.; Butterworths: London, 1970.

(2) Jenkins, H. D. B.; Marcus, Y. Viscosity B-Coefficients of Ions in Solution. *Chem. Rev.* **1995**, *95*, 2695–2724.

(3) Cameron, F. K.; Seidell, A. Solubility of Gypsum in Aqueous Solution of Certain Electrolytes. *J. Phys. Chem.* **1901**, *5*, 643–654.

(4) Bock, E. On the solubility of anhydrous calcium sulphate and of gypsum in concentrated solutions of sodium chloride at 25, 30, 35, 40, 45, and 50 °C. *Can. J. Chem.* **1961**, *39*, 1746–1751.

(5) Marshall, W. L.; Slusher, R. Thermodynamics of Calcium Sulfate Dihydrate in Aqueous Sodium Chloride Solutions, 0−1100. *J. Phys. Chem.* **1966**, *70*, 4015–4027.

(6) Susarla, V. R. K. S.; Sherwood, D. Solubility of Calcium Sulpfate in the Binary NaCl-H_2O and Ternary NaCl-$MgSO_4$-H_2O Systems. *J. Indian Chem. Soc.* **1997**, 24–26.

(7) Daniela, F.; Wolfgang, V. Crystallization and Phase Stability of $CaSO_4$ and $CaSO_4$-Based Salts. *Monatsh. Chem.* **2003**, *134*, 693–719.

(8) Raju, K. U. G.; Atkinson, G. The Thermodynamics of “Scale” Mineral Solubilities. 3. Calcium Sulfate in Aqueous NaCl. *J. Chem. Eng. Data* **1991**, *35*, 361–367.

(9) Kumar, A.; Mohandas, V. P.; Susarla, V. R. K. S.; Ghosh, P. K. Ionic Interactions of Calcium Sulfate Dihydrate in Aqueous Sodium Chloride Solutions: Solubilities, Densities, Viscosities, and Electrical Conductivities at 30 °C. *J. Solution Chem.* **2004**, *33*, 995–1003.

(10) Kumar, A.; Mohandas, V. P.; Sanghavi, R.; Ghosh, P. K. Ionic Interactions of Calcium Sulfate Dihydrate in Aqueous Sodium Chloride Solutions: Solubilities, Densities, Viscosities, and Electrical Conductivities, and Surface Tensions at 35 °C. *J. Solution Chem.* **2005**, *34*, 333–342.

(11) Kumar, A.; Sanghavi, R.; Mohandas, V. P. Experimental Densities, Speeds of Sound, Isentropic Compressibility and Shear Relaxation Time of $CaSO_4 \cdot 2H_2O + CaCl_2 + H_2O$ and $CaSO_4 \cdot 2H_2O + NaCl + H_2O$ Systems at Temperatures 30 and 35 °C. *J. Solution Chem.* **2006**, *35*, 1515–1524.

(12) Kumar, A.; Sanghavi, R.; Mohandas, V. P. Solubility Pattern of $CaSO_4 \cdot 2H_2O$ in the System $NaCl + CaCl_2 + H_2O$ and Solution Densities at 35 °C: Nonideality and Ion Pairing. *J. Chem. Eng. Data* **2007**, *52*, 902–905.

(13) Kumar, A.; Shukla, J.; Dangar, Y.; Mohandas, V. P. Effect of $MgCl_2$ on Solubility of $CaSO_4 \cdot 2H_2O$ in the Aqueous NaCl System and Physicochemical Solution Properties at 35 °C. *J. Chem. Eng. Data* **2010**, *55*, 1675–1678.

(14) Shukla, J.; Mohandas, V. P.; Kumar, A. Effect of pH on the Solubility of $CaSO_4 \cdot 2H_2O$ in Aqueous NaCl Solutions and Physicochemical Solution Properties at 35 °C. *J. Chem. Eng. Data* **2008**, *53*, 2797–2800.

(15) Mullin, J. W. *Crystallization*, 2nd ed.; Butterworths: London, 1972; Chapter 8, pp 263−265.

(16) Budz, J.; Karpinski, P. H.; Mydlarz, J.; Nyvlt, J. Salting-out precipitation of cocarboxylase hydrochloride from aqueous solution by addition of acetone. *Ind. Eng. Chem. Prod. Res. Dev.* **1986**, *25*, 657.

(17) Titiz-Sargut, S.; Sayan, P.; Avcı, B. Influence of citric acid on calcium sulfate dihydrate crystallization in aqueous media. *Cryst. Res. Technol.* **2007**, *42*, 119–126.

(18) Rashad, M. M.; Baioumy, H. M.; Abdel-Aal, E. A. Structural and spectral studies on gypsum crystals under simulated conditions of phosphoric acid production with and without organic and inorganic additives M. *Cryst. Res. Technol.* **2003**, *38*, 433–439.

(19) Song, X.; Sun, S.; Fan, W.; Yu, H. Preparation of different morphologies of calcium sulfate in organic media. *J. Mater. Chem.* **2003**, *13*, 1817–1821.

(20) Queste, S.; Bauduin, P.; Touraud, D.; Kunz, W.; Aubry, J. M. Short chain glycerol 1-monoethers-a new class of green solvosurfactants. *Green Chem.* **2006**, *8*, 822–830.

(21) Bauduin, P.; Renoncourt, A.; Kopf, A.; Touraud, D.; Kunz, W. Unified concept of solubilization in water by hydrotropes and cosolvents. *Langmuir* **2005**, *21*, 6769–6775.

(22) Dumez, A. G. Pharmaceutical uses of the glycols and their derivatives. *J. Am. Pharm. Assoc.* **1939**, *28*, 416–421.

(23) Barton, D. H.; Ollis, W. D., Eds. *Comprehensive Organic Chemistry*; Pergamon: Oxford, 1979; Vol. 1.

(24) Douhéret, G.; Pal, A.; Høiland, H.; Anowi, O.; Davis, M. I. Thermodynamic properties of (ethan-1,2-diol + water) at the temperature 298.15 K I. Molar volumes, isobaric molar heat capacities, ultrasonic speeds, and isentropic functions. *J. Chem. Thermodyn.* **1991**, *23*, 569–580.

(25) Douhéret, G.; Davis, M. I.; Hernandez, M. E.; Flores, H. Excess Molar Volumes of Some Polyether + Water Systems. *J. Indian Chem. Soc.* **1993**, *70*, 395–401.

(26) Hazra, D. K.; Das, B. Viscosities and Excess Molar Volumes for 2-Methoxy-1-ethanol + Water at Different Temperatures. *J. Chem. Eng. Data* **1993**, *38*, 361–363.

(27) Kan, A. T.; Fu, G.; Tomson, M. B. Effect of Methanol and Ethylene Glycol on Sulfates and Halite Scale Formation. *Ind. Eng. Chem. Res.* **2003**, *42*, 2399–2408.

(28) Surdo, A. L.; Alzola, E. M.; Millero, F. J. The P.V.T. Properties of Concentrated Aqueous Electrolytes. I. Densities and apparent molar volumes of NaCl, Na_2SO_4, $MgCl_2$ and $MgSO_4$ solutions from 0.1 $mol \cdot kg^{-1}$ to saturation and from 273.15 to 323.15 K. *J. Chem. Thermodyn.* **1982**, *14*, 649–662.

(29) Del Grosso, V. A.; Mader, C. W. Speed of Sound in Pure Water. *J. Acoust. Soc. Am.* **1972**, *52*, 1442–1446.

(30) Douhéret, G.; Reis, J. C. R.; Davis, M. I.; Fjellanger, I. J.; Høiland, H. Aggregative processes in aqueous solutions of mono- to tetraethylene glycol dimethyl ether at 298.15 K. *Phys. Chem. Chem. Phys.* **2004**, *6*, 784–792.

Received for review April 8, 2010. Accepted September 3, 2010. Financial support to T.S., SRF from CSIR (31/28(110) 2009 EMR-I), is acknowledged.

JE100336Q