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Archives of Physics Research, 2013, 4 (2):103-111
(http://scholarsresearchlibrary.com/archive.html)

ISSN : 0976-0970
CODEN (USA): APRRC7

Adiabatic compressibility of aqueous ehylene glycol and copper sulphate mixture at different temperatures

[*]N. S. S. V. Raja Rao[1], V. Brahmajirao[2] and A. V. Sarma[3]

[1]*Godavari Institute of Engineering and Technology, Rajahmundry, A. P., India*
[2]*Department of Nanoscience and Technology,School of Biotechnology, MGNIRSA, Hyderabad, A. P., India*
[3]*Department of Physics, Andhra University, Visakhapatnam, A. P., India*

ABSTRACT

Ultrasonic Velocity (U) and Density (ρ) have been measured using Antonpaar's DMA 5000M in a concentration range of 0.1 – 0.6 M and eleven compositions at 298, 303, 308, 313 and 318K temperatures for a ternary mixture of Aqueous Ethylene Glycol and Copper Sulphate system to study ion- solvent interactions and Eigen-Tomm Ion Pair formation Mechanism (ETIPM). It is observed that the values of density and ultrasonic velocity of any ionic liquid vary with increase in concentration of solutions, mainly due to the change in structure of solvent or solutions as a result of hydrogen bond formation or dissociation or hydrophobic or hydrophilic character of solute, or the solvent-solvent interactions, between Ethylene Glycol and water. Adiabatic compressibility (β) data indicate an ordering interaction leading to the formation of an ionic complex. The ionic complex formation in an ionic system is due to ionic association.

Keywords: Ultrasonic velocity, Density, Adiabatic compressibility, Binary Solvent Mixture(BSM), Ion pairs, ion-solvent interactions, Eigen Tomm Ion Pair Mechanism (ETIPM).

INTRODUCTION

Margaret Robson Wright [1], finds that Ultrasonic and Spectroscopic techniques probably allow a distinction to be made with reasonable certainty between outer and inner-sphere ion pairs, as was expressed by Ohtaki and Radnai [2]. Amongst the many techniques [3], the measurement of speed of sound in solution hitherto was being used to elucidate the structure of the solutions. By taking ultrasonic technique into account, the important consequences of ion solvation, such as reduced volume and the compressibility of the solvent molecules can be studied. The propagation of ultrasonic pulses through a medium can be made use of in the investigation of physical properties of the medium have led to the development of various methods for measuring velocity and attenuation. One such method is by the usage of a vibrating tube made to resonate to an external excitation. The Austrian made ANTONPAAR experimental setup, DSA-5000M is used for the very accurate determination of ultrasonic velocity and density. Adiabatic Compressibility can be calculated from the observed values of ultrasonic velocity and density of mixture at different temperatures.

MATERIALS AND METHODS

All the important precautions for the maintenance [4, 5] of the purity of the liquids used are very carefully implemented. $CuSO_4$ ($5H_2O$), Ethylene Glycol both are analytical reagents(AR) purchased from Merck with Assay

99.8%- 102% and 99.8% respectively. The copper sulphate used is in the form of a penta- hydrate (molecular weight 249.69). Ethylene Glycol is used as binary solvent mixture with water. The mixtures of the desired composition were prepared by weighing on a very accurate digital micro balance, Sartorius CPA -225D, to determine the mass of the electrolyte with a precision of ±0.0001 g. In order to prepare the solutions under investigation in the form of aqueous ionic liquids, a stock solution of 50ml of each concentration is prepared. This stock solution is added to ethylene glycol (redistilled) to makeup ionic liquids of the required composition. Each composition of 5ml is injected into the setup using a syringe.

Range and Accuracy: Ultrasonic velocity: 1000- 2000 m/sec with accuracy of 0.5 m/sec
Density: 0- 3 gram/cc with accuracy of 0. 000005 gram/cc
Temperature: 0- 70^0 C with accuracy of 0.03^{0}C.

Adiabatic compressibility, *β*:
It can be evaluated by the formula,

$$\beta = 1/U^2\rho$$

where, U is ultrasonic velocity, ρ is density of the solution. The data of U and ρ have been given in Tables: 1 and 2. Adiabatic Compressibility is inversely proportional to square of the ultrasonic velocity. The deviations in this parameter may be due to difference in size and shape of the molecules. The compressibility data, presented in Table: 3 indicate an ordering interaction leading to the formation of an ionic complex. The ionic complex formation in an ionic system is due to ionic association. The plots are drawn with concentration as abscissa and β as ordinate.

RESULTS AND DISCUSSION

Adiabatic compressibility increases with increase in temperature and decreases with increase in concentration. Increases in compressibility were attributed to the formation of hydrogen bonds between solute and solvent molecules [6].

Table: 1. Ultrasonic velocity data (in m/s) of $CuSO_4$ for 6 concentrations (gm.mol/lit or M) in 11 compositions of BSM of EG+WATER at different temperatures

TEMPERATURE: 298K							
% composition of water in BSM	0.1M	0.2M	0.3M	0.4M	0.5M	0.6M	EG+WATER
0	1656.96	1656.96	1656.96	1656.96	1656.96	1656.96	1656.96
10	1682.03	1678.86	1683.11	1683.89	1683.07	1684.95	1682.90
20	1701.18	1701.34	1699.87	1702.50	1701.39	1703.87	1700.79
30	1710.50	1711.55	1711.49	1711.81	1711.53	1712.58	1710.11
40	1708.56	1710.34	1712.12	1711.27	1712.32	1713.38	1707.28
50	1697.20	1699.24	1699.58	1699.79	1702.82	1704.52	1694.97
60	1673.91	1676.15	1675.01	1677.49	1683.54	1683.96	1668.52
70	1642.20	1641.89	1649.12	1648.35	1651.78	1656.29	1629.82
80	1602.65	1603.86	1601.99	1611.99	1616.91	1617.04	1599.91
90	1556.94	1553.21	1565.56	1568.92	1571.40	1580.69	1545.60
100	1506.00	1510.41	1517.35	1521.60	1529.89	1537.35	1497.08
TEMPERATURE : 303K							
0	1645.43	1645.43	1645.43	1645.43	1645.43	1645.43	1645.43
10	1671.04	1667.67	1671.74	1672.77	1671.84	1673.63	1671.80
20	1690.79	1690.82	1689.14	1691.83	1690.78	1693.28	1690.35
30	1700.90	1702.11	1701.94	1702.28	1701.87	1702.76	1700.72
40	1700.48	1702.10	1696.60	1703.07	1703.98	1705.35	1699.45
50	1691.29	1693.14	1693.40	1693.81	1696.48	1698.08	1689.22
60	1670.71	1672.73	1671.70	1673.96	1679.61	1679.99	1657.78
70	1642.15	1641.50	1648.51	1647.85	1651.11	1655.29	1630.77
80	1606.21	1607.41	1605.73	1614.94	1619.50	1619.79	1603.61
90	1564.54	1561.12	1572.48	1575.53	1577.97	1586.65	1553.89
100	1517.54	1521.63	1528.05	1532.00	1539.77	1546.82	1509.33

TEMPERATURE : 308K							
0	1633.63	1633.63	1633.63	1633.63	1633.63	1633.63	1633.63
10	1659.60	1656.18	1660.28	1661.39	1660.39	1662.45	1660.41
20	1680.06	1679.96	1678.27	1680.98	1679.84	1682.40	1679.59
30	1691.11	1692.63	1692.07	1692.33	1691.86	1692.63	1691.14
40	1692.18	1693.59	1690.93	1694.58	1695.46	1696.72	1691.31
50	1684.88	1686.54	1686.72	1687.25	1689.67	1691.16	1683.01
60	1668.89	1668.73	1667.72	1669.88	1675.08	1675.48	1662.53
70	1641.31	1641.11	1647.15	1646.58	1649.68	1653.54	1630.89
80	1608.80	1609.92	1608.47	1616.89	1621.11	1621.48	1606.35
90	1570.78	1567.66	1578.15	1580.93	1583.32	1591.27	1560.94
100	1527.44	1531.35	1537.32	1540.92	1548.23	1554.74	1519.74
TEMPERATURE : 313K							
0	1621.83	1621.83	1621.83	1621.83	1621.83	1621.83	1621.83
10	1648.07	1644.63	1648.76	1649.93	1648.87	1651.01	1648.97
20	1669.24	1669.00	1667.32	1670.04	1668.79	1671.42	1668.70
30	1681.11	1682.78	1681.98	1682.19	1681.63	1682.32	1681.32
40	1683.46	1684.79	1684.71	1685.82	1686.58	1687.74	1682.86
50	1678.02	1679.52	1679.63	1680.28	1682.46	1683.84	1676.39
60	1662.53	1664.19	1663.22	1665.27	1670.02	1670.46	1658.58
70	1639.74	1639.56	1645.04	1644.56	1647.51	1651.06	1630.21
80	1610.44	1611.43	1610.18	1617.88	1621.76	1622.23	1608.10
90	1575.77	1572.89	1582.56	1585.10	1587.43	1594.76	1566.94
100	1535.80	1539.55	1545.09	1548.32	1555.22	1561.21	1528.75
TEMPERATURE : 318K							
0	1609.99	1609.99	1609.99	1609.99	1609.99	1609.99	1609.99
10	1636.46	1632.99	1637.18	1638.39	1637.27	1639.45	1637.51
20	1658.28	1657.92	1656.23	1658.99	1657.63	1660.33	1657.67
30	1670.91	1672.73	1671.72	1671.90	1671.26	1671.88	1671.30
40	1674.50	1675.69	1678.08	1676.77	1677.36	1678.67	1674.09
50	1670.80	1672.14	1672.18	1672.97	1674.88	1676.13	1669.35
60	1657.57	1659.08	1658.14	1660.10	1664.45	1664.94	1654.08
70	1637.45	1637.24	1642.18	1641.81	1644.62	1647.88	1628.73
80	1611.16	1611.96	1610.92	1617.91	1621.48	1622.04	1608.88
90	1579.53	1576.87	1585.79	1588.10	1590.39	1597.12	1571.17
100	1542.86	1546.29	1551.40	1554.32	1560.85	1566.32	1536.30

Table: 2. Density data (in kg/m^3) of $CuSO_4$ for 6 concentrations (gr.mol/lit or M) in 11 compositions of BSM of EG+WATER at different temperatures

TEMPERATURE: 298K							
% composition of water in BSM	0.1M	0.2M	0.3M	0.4M	0.5M	0.6M	EG+WATER
0	1109.558	1109.558	1109.558	1109.558	1109.558	1109.558	1109.558
10	1103.881	1105.570	1107.086	1106.477	1110.255	1111.570	1102.542
20	1098.368	1101.103	1103.158	1104.974	1109.166	1113.092	1095.158
30	1090.749	1094.163	1098.840	1102.078	1108.189	1112.892	1085.094
40	1081.980	1088.678	1088.521	1096.555	1105.544	1111.574	1075.316
50	1072.837	1080.560	1087.477	1091.525	1102.493	1109.212	1064.974
60	1061.966	1071.309	1078.016	1084.237	1098.278	1106.091	1051.506
70	1050.912	1060.552	1072.212	1077.996	1092.368	1103.245	1036.523
80	1039.109	1050.671	1061.671	1071.329	1087.446	1098.620	1027.143
90	1026.831	1038.683	1054.354	1061.648	1080.925	1094.796	1010.938
100	1013.166	1029.484	1045.243	1053.861	1076.090	1090.656	997.241
TEMPERATURE: 303K							
0	1106.066	1106.066	1106.066	1106.066	1106.066	1106.066	1106.066
10	1100.425	1102.099	1103.600	1102.998	1106.734	1108.079	1099.079
20	1094.948	1097.681	1099.721	1101.544	1105.715	1109.638	1091.757
30	1087.430	1090.850	1095.505	1098.748	1104.818	1109.505	1081.811
40	1078.807	1085.479	1085.662	1093.361	1102.269	1108.350	1072.180
50	1069.870	1077.566	1084.458	1088.526	1099.440	1106.142	1062.035
60	1059.271	1068.567	1075.274	1081.465	1095.447	1103.245	1048.863
70	1048.499	1058.124	1069.714	1075.487	1089.823	1100.651	1034.231
80	1037.025	1048.547	1059.550	1069.115	1085.164	1096.318	1025.103
90	1025.059	1036.893	1052.460	1059.715	1078.930	1092.703	1009.281
100	1011.664	1027.916	1043.593	1052.153	1074.283	1088.753	995.844

TEMPERATURE: 308K							
0	1102.551	1102.551	1102.551	1102.551	1102.551	1102.551	1102.551
10	1096.921	1098.601	1100.097	1099.489	1103.148	1104.563	1095.588
20	1091.492	1094.219	1096.242	1098.073	1102.223	1106.145	1088.318
30	1084.068	1087.497	1092.115	1095.357	1101.395	1106.067	1078.479
40	1075.579	1082.224	1082.659	1090.098	1098.941	1105.043	1068.982
50	1066.826	1074.491	1081.361	1085.431	1096.304	1102.987	1059.024
60	1056.469	1065.733	1072.426	1078.597	1092.517	1100.302	1046.133
70	1045.967	1055.573	1067.092	1072.859	1087.152	1097.933	1031.803
80	1034.785	1046.272	1057.262	1066.749	1082.731	1093.879	1022.903
90	1023.102	1034.918	1050.385	1057.602	1076.762	1090.459	1007.425
100	1009.951	1026.137	1041.737	1050.243	1072.281	1086.668	994.236

TEMPERATURE: 313K							
0	1099.014	1099.014	1099.014	1099.014	1099.014	1099.014	1099.014
10	1093.391	1095.075	1096.561	1095.950	1099.606	1101.013	1092.068
20	1088.001	1090.721	1092.729	1094.563	1098.694	1102.613	1084.840
30	1080.662	1084.088	1088.681	1091.912	1097.925	1102.580	1075.100
40	1072.292	1078.912	1079.585	1086.775	1095.500	1101.675	1065.725
50	1063.706	1071.338	1078.185	1082.264	1093.092	1099.751	1055.938
60	1053.571	1062.802	1069.484	1075.633	1089.491	1097.266	1043.294
70	1043.318	1052.904	1064.354	1070.114	1084.364	1095.098	1029.248
80	1033.297	1043.851	1054.828	1064.241	1080.160	1091.292	1020.556
90	1020.974	1032.769	1048.145	1055.323	1074.428	1088.051	1005.387
100	1008.043	1024.170	1039.697	1048.155	1070.103	1084.414	992.420
TEMPERATURE: 318K							
0	1095.459	1095.459	1095.459	1095.459	1095.459	1095.459	1095.459
10	1089.838	1091.532	1092.996	1092.385	1096.091	1097.439	1088.521
20	1084.475	1087.189	1089.179	1091.016	1095.126	1099.045	1081.328
30	1077.208	1080.630	1085.201	1088.420	1094.407	1099.049	1071.674
40	1068.949	1075.539	1076.373	1083.391	1091.674	1098.242	1062.409
50	1060.514	1068.113	1074.937	1079.018	1089.808	1096.434	1052.777
60	1050.583	1059.778	1066.446	1072.574	1086.372	1094.134	1040.361
70	1040.551	1050.115	1061.501	1067.256	1081.464	1092.153	1026.573
80	1029.872	1041.292	1052.256	1061.600	1077.458	1088.569	1018.069
90	1018.681	1030.459	1045.750	1052.889	1071.939	1085.494	1003.183
100	1005.946	1022.022	1037.484	1045.896	1067.764	1082.000	990.417

Table 3. Adiabatic Compressibility data (β in $kg^{-1}ms^2$) of $CuSO_4$ for 6 concentrations in 11 compositions of BSM of EG+WATER at different temperatures

TEMPERATURE: 298K							
% composition of water in BSM	0.1M $x10^{-9}$	0.2M $x10^{-9}$	0.3M $x10^{-9}$	0.4M $x10^{-9}$	0.5M $x10^{-9}$	0.6M $x10^{-9}$	EG+WATER $x10^{-9}$
0	0.3283	0.3283	0.3283	0.3283	0.3283	0.3283	0.3283
10	0.3202	0.3209	0.3188	0.3185	0.3180	0.3169	0.3202
20	0.3146	0.3138	0.3138	0.3123	0.3114	0.3095	0.3157
30	0.3133	0.3120	0.3107	0.3096	0.3080	0.3064	0.3151
40	0.3166	0.3140	0.3134	0.3114	0.3085	0.3063	0.3190
50	0.3236	0.3205	0.3183	0.3170	0.3128	0.3103	0.3268
60	0.3361	0.3322	0.3306	0.3277	0.3212	0.3188	0.3416
70	0.3528	0.3498	0.3429	0.3414	0.3355	0.3304	0.3632
80	0.3747	0.3700	0.3670	0.3592	0.3517	0.3481	0.3803
90	0.4018	0.3991	0.3869	0.3827	0.3746	0.3656	0.4141
100	0.4352	0.4258	0.4155	0.4098	0.3892	0.3879	0.4474
TEMPERATURE : 303K							
0	0.3339	0.3339	0.3339	0.3339	0.3339	0.3339	0.3339
10	0.3254	0.3263	0.3242	0.3240	0.3233	0.3222	0.3255
20	0.3195	0.3187	0.3187	0.3172	0.3164	0.3143	0.3206
30	0.3179	0.3164	0.3157	0.3140	0.3125	0.3109	0.3196
40	0.3206	0.3180	0.3200	0.3153	0.3124	0.3102	0.3229
50	0.3268	0.3237	0.3216	0.3202	0.3160	0.3135	0.3300
60	0.3382	0.3345	0.3325	0.3300	0.3236	0.3212	0.3469
70	0.3537	0.3506	0.3440	0.3424	0.3366	0.3316	0.3636
80	0.3738	0.3691	0.3660	0.3586	0.3513	0.3477	0.3793
90	0.3985	0.3957	0.3842	0.3801	0.3722	0.3635	0.4103
100	0.4292	0.4202	0.4104	0.4050	0.3926	0.3839	0.4278

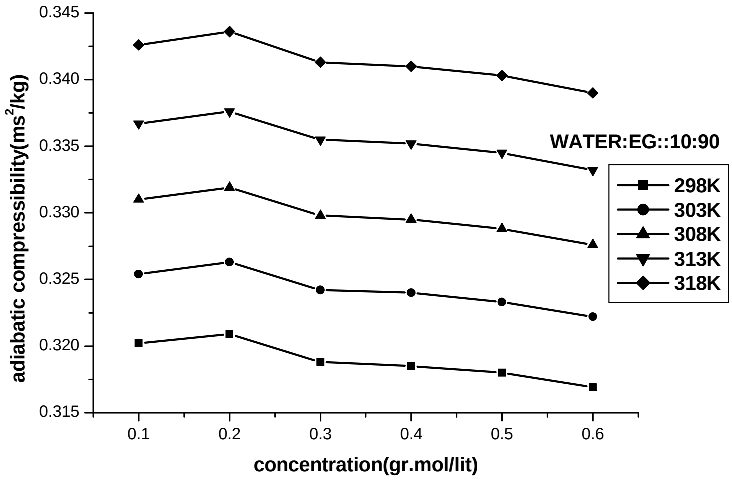
TEMPERATURE : 308K							
0	0.3399	0.3398	0.3399	0.3399	0.3399	0.3399	0.3399
10	0.3310	0.3319	0.3298	0.3295	0.3288	0.3276	0.3311
20	0.3246	0.3238	0.3239	0.3223	0.3215	0.3194	0.3257
30	0.3226	0.3210	0.3198	0.3188	0.3172	0.3156	0.3242
40	0.3247	0.3222	0.3230	0.3194	0.3165	0.3143	0.3270
50	0.3302	0.3272	0.3250	0.3236	0.3195	0.3170	0.3334
60	0.3399	0.3370	0.3353	0.3325	0.3262	0.3237	0.3458
70	0.3549	0.3518	0.3454	0.3438	0.3380	0.3331	0.3644
80	0.3734	0.3688	0.3656	0.3586	0.3514	0.3477	0.3789
90	0.3961	0.3932	0.3823	0.3783	0.3705	0.3622	0.4074
100	0.4244	0.4156	0.4062	0.4010	0.3891	0.3807	0.4355

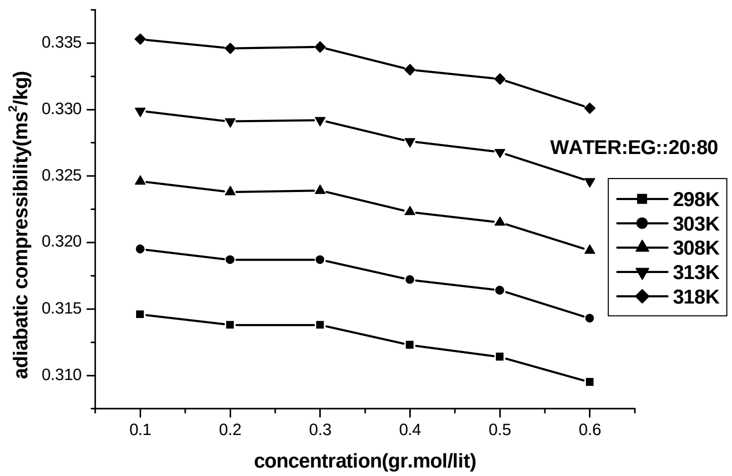
TEMPERATURE : 313K							
0	0.3459	0.3459	0.3459	0.3459	0.3459	0.3459	0.3459
10	0.3367	0.3376	0.3355	0.3352	0.3345	0.3332	0.3368
20	0.3299	0.3291	0.3292	0.3276	0.3268	0.3246	0.3310
30	0.3274	0.3254	0.3247	0.3236	0.3221	0.3205	0.3290
40	0.3291	0.3265	0.3263	0.3238	0.3209	0.3187	0.3313
50	0.3339	0.3309	0.3288	0.3273	0.3232	0.3207	0.3370
60	0.3434	0.3397	0.3380	0.3352	0.3291	0.3266	0.3487
70	0.3565	0.3533	0.3472	0.3455	0.3398	0.3350	0.3656
80	0.3732	0.3689	0.3657	0.3590	0.3520	0.3482	0.3789
90	0.3945	0.3914	0.3809	0.3771	0.3693	0.3614	0.4052
100	0.4206	0.4119	0.4029	0.3980	0.3864	0.3783	0.4312
TEMPERATURE : 318K							
0	0.3522	0.3522	0.3522	0.3522	0.3522	0.3522	0.3522
10	0.3426	0.3436	0.3413	0.3410	0.3403	0.3390	0.3426
20	0.3353	0.3346	0.3347	0.3330	0.3323	0.3301	0.3365
30	0.3325	0.3307	0.3297	0.3287	0.3271	0.3255	0.3341
40	0.3336	0.3311	0.3299	0.3283	0.3256	0.3231	0.3359
50	0.3378	0.3348	0.3327	0.3311	0.3271	0.3246	0.3409
60	0.3464	0.3428	0.3411	0.3383	0.3323	0.3297	0.3513
70	0.3584	0.3553	0.3493	0.3476	0.3419	0.3372	0.3672
80	0.3741	0.3696	0.3662	0.3599	0.3530	0.3492	0.3795
90	0.3935	0.3903	0.3803	0.3766	0.3688	0.3612	0.4038
100	0.4176	0.4092	0.4005	0.3958	0.3844	0.376	0.4278

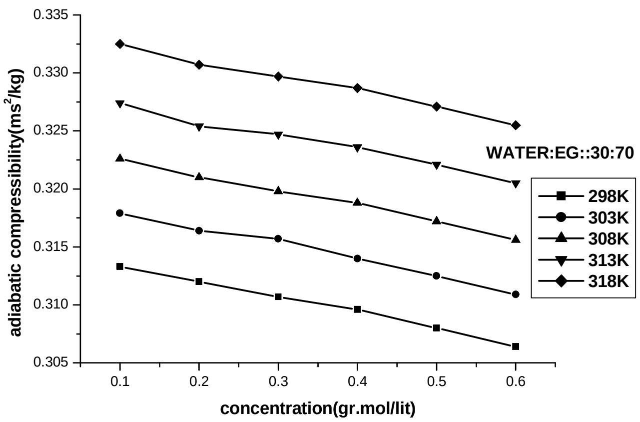
The adiabatic compressibility of aqueous electrolytic solutions is due to about 64% configurational and 36% vibrational compressibility. In dilute solution the adiabatic compressibility is predominantly governed by the configurational compressibility whereas in the concentrated solutions it is due to the vibrational compressibility [7]. In aqueous electrolytic solutions the ion-solvent interactions dominate up to a certain concentration forming a rigid hydration shell and beyond this concentration a transition from ion-solvent to ion-ion interactions occurs leading to the formation of ion-pairs (Solvent-separated, solvent-shared and contact). This critical concentration depends on the nature of the electrolyte. The configurational compressibility contributes to the adiabatic compressibility up to that concentration and beyond that, the vibrational compressibility dominates in the solution. The general assumption of zero compressibility of the primary hydration shell is not collaborated by experimental observation.

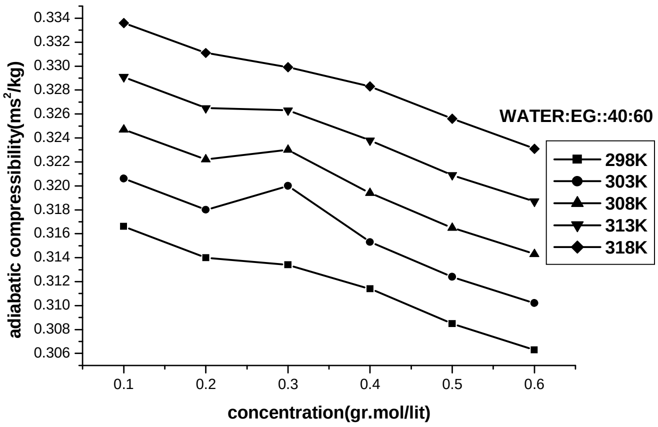
When a Cu^{2+} ion is added to a solvent, certain solvent molecules get attracted towards itself by wrenching the molecule in bulk of the solvent due to the forces of electrostriction. Consequently the available solvent molecules for the next incoming ions get decreased. The compressibility measures the ease with which a medium can be compressed. Thus the compressibility of a solvent is higher than that of solution and it decreases with the increase in concentration of the electrolyte in the solution. With increase in ionic concentration, the electrostrictive forces causes the structure to break [8] and the water molecules surrounding Cu^{2+} are more compactly packed and therefore compressibility decreases with increase in Cu^{2+} concentration. The decrease in adiabatic compressibility indicates the enhancement of the bond strength at that concentration.

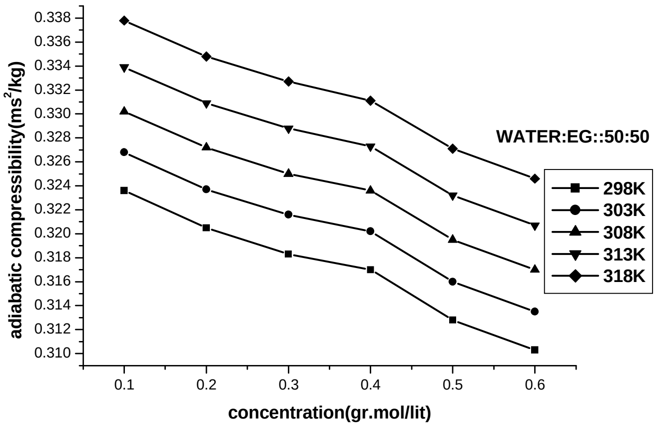
On increasing the salt concentration in the solution, the number of free water molecules around the ion decreases gradually until a situation is reached where all the water molecules are involved in the primary hydration shell of the solute. The adiabatic compressibility at that concentration becomes independent of temperature and assigned as critical adiabatic compressibility. Such a condition may be correlated with the saturation of the primary hydration shell since the water molecules are not compressed further and become independent of temperature. Beyond this concentration, co- spheres of cation and anion start to overlap leading to the formation of ion pairs such as solvent-separated, solvent- shared and contact ion pairs [9].

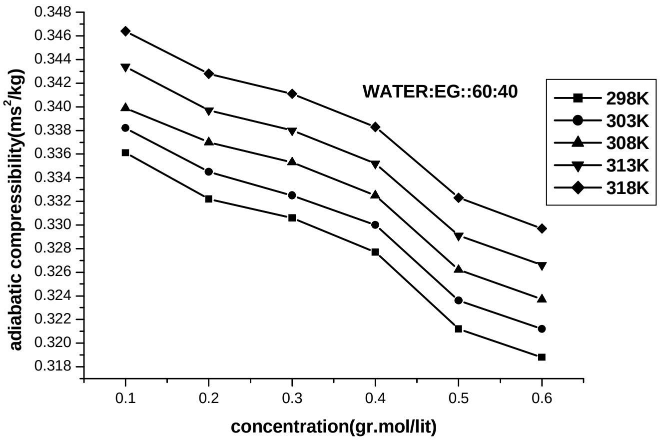


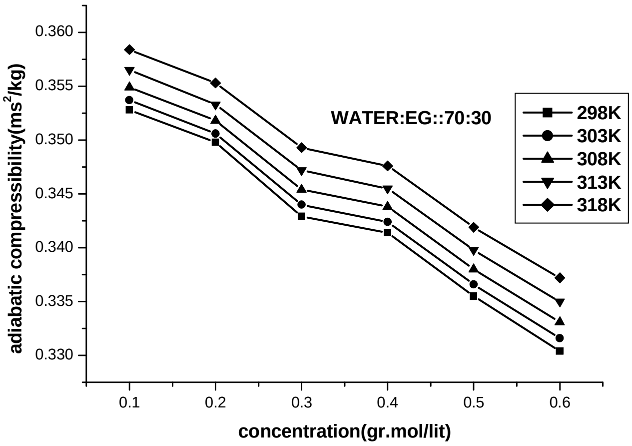


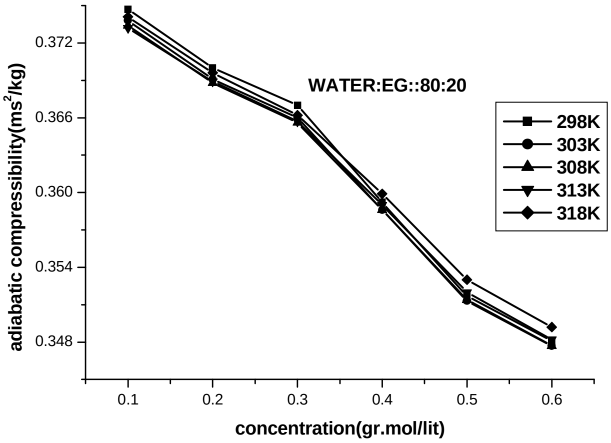


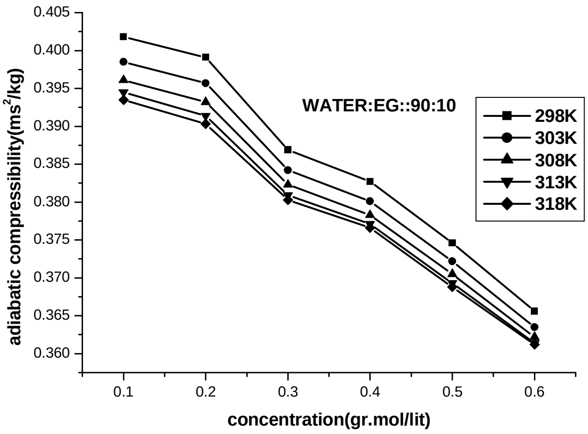

adiabatic compressibility(ms²/kg)
concentration(gr.mol/lit)
WATER:EG::40:60
298K
303K
308K
313K
318K


adiabatic compressibility(ms²/kg)
concentration(gr.mol/lit)
WATER:EG::50:50
298K
303K
308K
313K
318K


adiabatic compressibility(ms²/kg)
concentration(gr.mol/lit)
WATER:EG::60:40
298K
303K
308K
313K
318K


WATER:EG::70:30
298K
303K
308K
313K
318K
adiabatic compressibility(ms^2/kg)
concentration(gr.mol/lit)


WATER:EG::80:20
298K
303K
308K
313K
318K
adiabatic compressibility(ms^2/kg)
concentration(gr.mol/lit)


WATER:EG::90:10
298K
303K
308K
313K
318K
adiabatic compressibility(ms^2/kg)
concentration(gr.mol/lit)

CONCLUSION

The adiabatic compressibility is rendered sensitive not only to the ionic and molecular interactions but also dependent on the dipolar behaviour of the solvent molecules. This makes it a precious tool to probe into solvent-solvent, Ion- solvent, and ion- ion interactions precisely and divulges valuable conceptions to understand the ion pair formation mechanism. Many authors tried to compute the hydration compressibility [10] by using the Passynskii model. This model is based on the assumption that the solvent molecules solvating ions were fully compressed by the electrical forces of ions. As a result, the compressibility of the solvent molecules in the hydration shell is assumed to be zero. However this fact is not collaborated by the experimental confirmations [11,12].

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