sulfate concentration but increases at constant $MgCl_2$ when the sulfate is increased. We found this behavior to be related to the different polarizability of Cl^- and SO_4^{2-} anions.

Acknowledgment

We thank Mr. P. Perna for his contribution in performing the experimental measurements.

Glossary

a_w	activity of water
G	NRTL binary parameter defined in Table III
G^E	total excess Gibbs energy, J
K_{sp}^0	thermodynamic solubility product of $CaSO_4{\cdot}2H_2O$
$K_{(ij)^0}$	dissociation equilibrium constant for ion pair $(ij)^0$
m_i	true molality of species i
$m_{(ij)^0}$	molality of ion pair $(ij)^0$
N	number of experimental determinations
S_{Ca}	solubility of $CaSO_4{\cdot}2H_2O$, m
S_γ, $S_{S_{Ca}}$	sums of squares
Z	NRTL binary parameter defined in Table III

Greek Letters

$\gamma_{\pm(ij)}$	mean ionic molal activity coefficient of electrolyte (ij)
$\gamma_{(ij)^0}$	molal activity coefficient of ion pair $(ij)^0$
δ	Debye–McAulay parameter defined in Table III
$\delta_{(ij)^0}$	Debye–McAulay binary parameter for ion pair $(ij)^0$
Δ_{Cl^-}	empirical parameter specific for chloride ions
ρ	density
σ_γ, σ_2	square root of estimated variances
$\tau_{(ij)^0}$	NRTL binary parameter for ion pairs

Registry No. $CaSO_4$, 7778-18-9; Na_2SO_4, 7757-82-6; $MgCl_2$, 7786-30-3; gypsum, 13397-24-5.

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Received for review March 21, 1983. Accepted June 28, 1983. Financial support for the research has been provided by the Ministero della Pubblica Istruzione.

Density, Viscosity, and Electrolytic Conductivity of Concentrated Aqueous Electrolyte Solutions at Several Temperatures. Alkaline-Earth Chlorides, $LaCl_3$, Na_2SO_4, $NaNO_3$, NaBr, KNO_3, KBr, and $Cd(NO_3)_2$

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The densities, viscosities, and electrolytic conductivities of concentrated aqueous solutions of alkaline-earth chlorides, $LaCl_3$, Na_2SO_4, $NaNO_3$, NaBr, KNO_3, KBr, and $Cd(NO_3)_2$ have been measured at temperatures from 15 to 55 °C. Temperature dependences of the above-mentioned properties are represented in terms of their thermal coefficients at 25 °C.

Introduction

Density, viscosity, electrolytic conductivity, and their thermal coefficients of electrolyte solutions are physicochemically important properties to understand the nature of solute–solute and solute–solvent interactions and the structural change of solvent induced by the solute. Although a number of studies on the above-mentioned properties have been found in the literature (*1*, *2*), experimental data on concentrated solutions are still lacking for the discussion of solute–solute–solvent interactions. This situation has encouraged us to make a series of systematic measurements of density, viscosity, and electrolytic conductivity of aqueous electrolyte solutions at concentrations from 0.05

Table I. Density and Viscosity of Water

temp/ °C	density[a]/ (kg m^{-3})	viscosity[b]/ (mN s m^{-2})
15	999.101	1.138
20	998.206	1.002
25	997.047	0.8903
30	995.650	0.7975
35	994.036	0.7194
40	992.219	0.6531
45	990.216	0.5963
50	988.038	0.5467
55	985.695	0.5044

[a] Reference *4*. [b] Reference *5*.

Table II. Density ρ, Viscosity η, and Electrolytic Conductivity κ of Several Aqueous Electrolyte Solutions

m/(mol kg^{-1})	15.00 °C	20.00 °C	25.00 °C	30.00 °C	35.00 °C	40.00 °C	45.00 °C	50.00 °C	55.00 °C
				Magnesium Chloride					
				ρ/(kg m^{-3})					
0.050	1003.10	1002.17	1001.00	999.58	997.97		994.16		989.66
0.100	1006.99	1006.05	1004.87	1003.45	1001.83		998.01		993.54
0.500	1037.20	1036.09	1034.79	1033.28	1031.64		1027.82		1023.43
1.000	1072.67	1071.59	1070.04	1068.44	1066.76		1062.91		1058.63
2.000	1138.28	1136.82	1135.26	1133.58	1131.82		1128.00		1123.86
3.000	1194.63	1193.03	1191.36	1189.65	1187.86		1184.05		1180.02
4.000	1247.60	1245.89	1244.19	1242.40	1240.58		1236.76		1232.77
5.000	1298.03	1296.26	1294.50	1292.66	1290.82		1286.99		1283.04
				η/(mN s m^{-2})					
0.050	1.1640	1.0249	0.9110	0.8162	0.7364		0.6104		0.5165
0.100	1.1871	1.0456	0.9296	0.8329	0.7517		0.6236		0.5276
0.500	1.3812	1.2186	1.0847	0.9729	0.8788		0.7293		0.6186
1.000	1.6724	1.4754	1.3139	1.1785	1.0654		0.8849		0.7512
2.000	2.492	2.199	1.950	1.747	1.574		1.302		1.102
3.000	3.805	3.332	2.937	2.616	2.346		1.926		1.617
4.000	6.087	5.271	4.603	4.064	3.615		2.926		2.431
5.000	10.549	9.001	7.752	6.761	5.951		4.726		3.866
				κ/(S m^{-1})					
0.050	0.7802	0.8746	0.9732	1.0739	1.177		1.389		1.610
0.100	1.449	1.624	1.805	1.991	2.181		2.571		2.975
0.500	5.683	6.351	7.042	7.751	8.478		9.959		11.49
1.000	9.226	10.306	11.42	12.56	13.73		16.13		18.59
2.000	12.57	14.07	15.63	17.24	18.89		22.27		25.77
3.000	12.57	14.18	15.84	17.56	19.34		23.04		26.87
4.000	10.75	12.26	13.83	15.49	17.20		20.78		24.54
5.000	8.108	9.389	10.75	12.18	13.69		16.88		20.30
				Calcium Chloride					
				ρ/(kg m^{-3})					
0.050	1003.70	1002.77	1001.59	1000.17	998.55		994.73		990.19
0.100	1008.28	1007.29	1006.10	1004.65	1003.02		999.16		994.64
0.500	1043.62	1042.40	1041.00	1039.37	1037.60		1033.57		1028.97
1.000	1085.34	1083.87	1082.22	1080.40	1078.50		1074.27		1069.58
2.000	1161.89	1159.97	1157.99	1155.85	1153.68		1149.04		1144.13
3.000	1229.87	1227.54	1225.24	1222.81	1220.37		1215.31		1210.19
4.000	1290.63	1287.93	1285.25	1282.51	1279.80		1274.28		1268.72
6.000	1391.93	1388.68	1385.50	1382.30	1379.11		1372.74		1366.38
				η/(mN s m^{-2})					
0.050	1.1572	1.0182	0.9052	0.8107	0.7318		0.6075		0.5140
0.100	1.1729	1.0338	0.9192	0.8242	0.7442		0.6181		0.5232
0.500	1.3051	1.1558	1.0322	0.9285	0.8406		0.7004		0.5963
1.000	1.4940	1.3283	1.1902	1.0741	0.9753		0.8172		0.6982
2.000	2.0053	1.7890	1.6092	1.4569	1.3265		1.1188		0.9602
3.000	2.791	2.488	2.238	2.025	1.844		1.554		1.334
4.000	4.061	3.603	3.225	2.908	2.636		2.207		1.884
6.000	9.794	8.476	7.414	6.541	5.814		4.699		3.884
				κ/(S m^{-1})					
0.050	0.8240	0.9228	1.024	1.128	1.235		1.454		1.681
0.100	1.547	1.730	1.918	2.112	2.309		2.714		3.140
0.500	6.305	7.024	7.752	8.504	9.264		10.82		12.42
1.000	10.67	11.84	13.03	14.26	15.50		18.04		20.63
2.000	15.68	17.35	19.05	20.79	22.56		26.19		29.87
3.000	16.80	18.61	20.46	22.36	24.31		28.26		32.30
4.000	15.39	17.14	18.94	20.80	22.69		26.60		30.61
6.000	9.768	11.13	12.55	14.05	15.60		18.87		22.32
				Strontium Chloride					
				ρ/(kg m^{-3})					
0.050	1006.14	1005.18	1003.99	1002.56	1000.93		997.07		992.54
0.100	1013.19	1012.19	1010.97	1009.51	1007.87		1003.96		999.40
0.500	1067.49	1066.18	1064.67	1062.98	1061.13		1056.93		1052.17
1.000	1131.93	1130.15	1128.36	1126.39	1124.33		1119.79		1114.80
1.500	1192.26	1190.24	1188.15	1185.98	1183.77		1178.87		1173.71
2.000	1239.48	1237.31	1235.12	1232.71	1230.33		1225.18		1219.80
2.500	1288.37	1286.01	1283.51	1280.97	1278.40		1272.99		1267.43
				η/(mN s m^{-2})					
0.050	1.1554	1.0181	0.9055	0.8117	0.7325		0.6082		0.5161
0.100	1.1703	1.0322	0.9188	0.8241	0.7443		0.6186		0.5249
0.500	1.2920	1.1458	1.0246	0.9228	0.8365		0.6997		0.5969
1.000	1.4728	1.3123	1.1785	1.0653	0.9688		0.8151		0.6995
1.500	1.6961	1.5153	1.3643	1.2362	1.1266		0.9514		0.8190
2.000	1.9252	1.7224	1.5523	1.4079	1.2842		1.0862		0.9364
2.500	2.2276	1.9936	1.7971	1.6300	1.4873		1.2580		1.0850

Table II (*Continued*)

m/ (mol kg^{-1})	15.00 °C	20.00 °C	25.00 °C	30.00 °C	35.00 °C	40.00 °C	45.00 °C	50.00 °C	55.00 °C
κ/(S m^{-1})									
0.050	0.8281	0.9272	1.029	1.134	1.241		1.461		1.687
0.100	1.564	1.749	1.940	2.135	2.333		2.742		3.162
0.500	6.336	7.047	7.778	8.523	9.282		10.83		12.42
1.000	10.72	11.88	13.05	14.27	15.50		18.00		20.55
1.500	13.74	15.19	16.67	18.18	19.71		22.85		26.04
2.000	15.35	16.95	18.58	20.26	21.96		25.43		28.95
2.500	16.30	17.99	19.73	21.51	23.33		27.05		30.79
Barium Chloride									
ρ/(kg m^{-3})									
0.050	1008.33	1007.37	1006.16	1004.73	1003.08		999.18		994.62
0.100	1017.34	1016.31	1015.07	1013.59	1011.91		1007.97		1003.35
0.500	1088.46	1087.09	1085.49	1083.73	1081.83		1077.47		1072.51
0.700	1122.09	1120.55	1118.85	1116.95	1114.92		1110.36		1105.18
1.000	1172.52	1170.82	1168.80	1166.67	1164.46		1159.47		1154.11
1.200	1205.13	1203.20	1201.02	1198.76	1196.34		1191.21		1185.67
1.500	1251.60	1249.35	1247.01	1244.60	1242.04		1236.67		1230.94
η/(mN s m^{-2})									
0.050	1.1534	1.0164	0.9041	0.8105	0.7315		0.6068		0.5141
0.100	1.1646	1.0274	0.9144	0.8204	0.7405		0.6141		0.5208
0.500	1.2643	1.1226	1.0052	0.9062	0.8219		0.6880		0.5868
0.700	1.3186	1.1736	1.0530	0.9513	0.8637		0.7250		0.6200
1.000	1.4123	1.2609	1.1342	1.0272	0.9352		0.7880		0.6759
1.200	1.4821	1.3252	1.1938	1.0824	0.9867		0.8328		0.7155
1.500	1.5968	1.4301	1.2902	1.1714	1.0690		0.9047		0.7794
κ/(S m^{-1})									
0.050	0.8462	0.9461	1.049	1.154	1.261		1.482		1.712
0.100	1.580	1.765	1.953	2.147	2.344		2.748		3.167
0.500	6.515	7.231	7.957	8.704	9.458		11.00		12.57
0.700	8.484	9.391	10.32	11.27	12.23		14.19		16.17
1.000	11.10	12.26	13.44	14.65	15.87		18.35		20.86
1.200	12.56	13.85	15.17	16.52	17.88		20.65		23.44
1.500	14.32	15.78	17.26	18.76	20.30		23.40		26.53
Lanthanum Chloride									
ρ/(kg m^{-3})									
0.050	1010.46	1009.50	1008.34	1006.90	1005.27		1001.43		996.89
0.100	1021.71	1020.72	1019.49	1018.04	1016.38		1012.51		1007.99
0.500	1108.80	1107.45	1105.95	1104.28	1102.48		1098.43		1093.81
1.000	1212.15	1210.50	1208.75	1206.84	1204.92		1200.61		1195.80
1.500	1307.51	1305.55	1303.54	1301.49	1299.38		1294.87		1289.89
2.000	1397.56	1395.39	1393.19	1390.95	1388.66		1383.94		1378.74
2.500	1480.22	1477.89	1475.52	1473.10	1470.63		1465.55		1460.36
3.000	1557.52	1555.08	1552.60	1550.05	1547.49		1542.13		1536.69
η/(mN s m^{-2})									
0.050	1.1762	1.0367	0.9217	0.8261	0.7453		0.6180		0.5241
0.100	1.2107	1.0675	0.9498	0.8516	0.7687		0.6382		0.5409
0.500	1.5159	1.3414	1.1969	1.0759	0.9735		0.8112		0.6896
1.000	2.053	1.818	1.623	1.460	1.322		1.102		0.9379
1.500	2.852	2.522	2.246	2.016	1.824		1.516		1.287
2.000	4.108	3.611	3.199	2.857	2.573		2.120		1.787
2.500	6.069	5.287	4.642	4.114	3.678		2.992		2.492
3.000	9.273	7.977	6.922	6.069	5.371		4.290		3.517
κ/(S m^{-1})									
0.050	1.146	1.282	1.424	1.567	1.714		2.014		2.325
0.100	2.127	2.378	2.636	2.900	3.167		3.715		4.285
0.500	8.081	8.997	9.936	10.89	11.86		13.84		15.88
1.000	12.37	13.75	15.16	16.61	18.07		21.03		24.03
1.500	14.03	15.61	17.23	18.89	20.58		23.99		27.47
2.000	13.74	15.35	17.00	18.68	20.41		23.93		27.48
2.500	12.18	13.69	15.25	16.87	18.51		21.88		25.33
3.000	9.994	11.34	12.74	14.20	15.69		18.78		21.97
Sodium Sulfate									
ρ/(kg m^{-3})									
0.050	1005.63	1004.66	1003.43	1001.96	1000.29		996.39		991.84
0.100	1012.01	1010.96	1009.66	1008.15	1006.45		1002.48		997.86
0.500	1060.39	1058.90	1057.23	1055.37	1053.37		1048.96		1044.04
0.700		1081.54	1079.73	1077.76	1075.66		1071.06		1066.01
1.000		1114.24	1112.20	1110.03	1107.77		1102.94		1097.67
1.200		1135.22	1133.06	1130.80	1128.46		1123.44		1118.13
1.500			1161.60	1159.26	1156.82		1151.68		1146.17

Table II (*Continued*)

m/(mol kg^{-1})	15.00 °C	20.00 °C	25.00 °C	30.00 °C	35.00 °C	40.00 °C	45.00 °C	50.00 °C	55.00 °C
				η/(mN s m^{-2})					
0.050	1.1628	1.0249	0.9113	0.8166	0.7374		0.6114		0.5168
0.100	1.1863	1.0460	0.9298	0.8339	0.7529		0.6240		0.5289
0.500	1.3993	1.2356	1.0999	0.9866	0.8914		0.7405		0.6271
0.700		1.3492	1.2007	1.0761	0.9716		0.8058		0.6830
1.000		1.5478	1.3739	1.2289	1.1074		0.9160		0.7738
1.200		1.7009	1.5055	1.3449	1.2096		0.9978		0.8410
1.500			1.7200	1.5312	1.3731		1.1272		0.9464
				κ/(S m^{-1})					
0.050	0.7138	0.8028	0.8949	0.9896	1.087		1.288		1.495
0.100	1.297	1.459	1.626	1.799	1.975		2.336		2.713
0.500	4.671	5.262	5.871	6.497	7.142		8.457		9.800
0.700		6.633	7.410	8.213	9.031		10.70		12.42
1.000		8.230	9.216	10.24	11.28		13.41		15.58
1.200		9.071	10.18	11.32	12.49		14.89		17.34
1.500			11.23	12.52	13.85		16.58		19.36
				Sodium Nitrate					
				ρ/(kg m^{-3})					
0.050	1002.09	1001.09	999.87	998.42	996.77		992.89		988.32
0.100	1004.99	1003.96	1002.72	1001.23	999.55		995.61		991.00
0.500	1027.47	1026.10	1024.54	1022.80	1020.87		1016.52		1011.60
1.000	1054.28	1052.46	1050.60	1048.54	1046.34		1041.56		1036.28
2.000	1103.85	1101.47	1099.06	1096.56	1093.94		1088.49		1082.65
3.000	1148.94	1146.29	1143.62	1140.76	1137.87		1131.81		1125.46
4.000	1188.07	1185.15	1182.10	1178.97	1175.85		1169.37		1162.67
6.000	1261.22	1257.87	1254.40	1250.95	1247.49		1240.38		1233.13
8.000	1318.96	1315.32	1311.74	1308.08	1304.42		1296.85		1289.21
				η/(mN s m^{-2})					
0.050	1.1403	1.0046	0.8932	0.8005	0.7222		0.5989		0.5068
0.100	1.1419	1.0066	0.8955	0.8028	0.7247		0.6018		0.5096
0.500	1.1583	1.0251	0.9150	0.8225	0.7446		0.6207		0.5280
1.000	1.1914	1.0576	0.9465	0.8530	0.7732		0.6476		0.5522
2.000	1.2923	1.1503	1.0318	0.9323	0.8482		0.7131		0.6102
3.000	1.4317	1.2749	1.1439	1.0336	0.9397		0.7913		0.6785
4.000	1.5943	1.4178	1.2710	1.1476	1.0431		0.8782		0.7517
6.000	2.0287	1.7962	1.6041	1.4440	1.3089		1.0971		0.9378
8.000	2.5323	2.2307	1.9832	1.7783	1.606		1.338		1.139
				κ/(S m^{-1})					
0.050	0.4222	0.4716	0.5224	0.5747	0.6280		0.7378		0.8511
0.100	0.8184	0.9136	1.012	1.112	1.215		1.426		1.645
0.500	3.446	3.840	4.243	4.658	5.080		5.949		6.834
1.000	6.048	6.729	7.428	8.146	8.876		10.37		11.89
2.000	9.880	10.98	12.11	13.26	14.44		16.85		19.29
3.000	12.41	13.80	15.21	16.68	18.15		21.16		24.23
4.000	13.97	15.54	17.15	18.81	20.48		23.91		27.38
6.000	15.61	17.42	19.27	21.18	23.12		27.05		31.07
8.000	15.95	17.86	19.84	21.87	23.91		28.14		32.45
				Sodium Bromide					
				ρ/(kg m^{-3})					
0.050	1003.09	1002.16	1000.97	999.53	997.90	996.06	994.04	991.85	989.50
0.100	1007.14	1006.17	1004.95	1003.49	1001.83	999.97	997.93	995.72	993.35
0.500	1038.27	1037.04	1035.58	1033.91	1032.07	1030.07	1027.89	1025.55	1023.11
1.000	1076.03	1074.50	1072.76	1070.87	1068.83	1066.64	1064.33	1061.87	1059.32
2.000	1147.45	1145.45	1143.24	1140.97	1138.58	1136.07	1133.47	1130.79	1128.03
3.000	1214.33	1211.84	1209.32	1206.66	1203.95	1201.15	1198.28	1195.38	1192.42
4.000	1277.12	1274.28	1271.44	1268.50	1265.54	1262.53	1259.48	1256.41	1253.27
6.000	1392.05	1388.66	1385.25	1381.77	1378.33	1374.89	1371.46	1367.89	1364.46
8.000	1494.20	1490.45	1486.63	1482.75	1478.91	1475.03	1471.17	1467.32	1463.47
				η/(mN s m^{-2})					
0.050	1.1409	1.0053	0.8935	0.8006	0.7225	0.6562	0.5992	0.5496	0.5072
0.100	1.1428	1.0073	0.8956	0.8030	0.7249	0.6587	0.6017	0.5519	0.5095
0.500	1.1601	1.0269	0.9162	0.8240	0.7458	0.6790	0.6213	0.5708	0.5276
1.000	1.1876	1.0547	0.9443	0.8517	0.7722	0.7046	0.6460	0.5948	0.5508
2.000	1.2675	1.1307	1.0157	0.9187	0.8359	0.7648	0.7032	0.6489	0.6023
3.000	1.3824	1.2347	1.1108	1.0059	0.9163	0.8392	0.7724	0.7136	0.6629
4.000	1.5331	1.3682	1.2305	1.1139	1.0143	0.9290	0.8550	0.7896	0.7337
6.000	1.9689	1.7464	1.5626	1.4075	1.2766	1.1650	1.0688	0.9845	0.9128
8.000	2.6418	2.3169	2.0523	1.8325	1.6492	1.4944	1.3627	1.2484	1.1515
				κ/(S m^{-1})					
0.050	0.4547	0.5081	0.5636	0.6206	0.6789	0.7383	0.7990	0.8614	0.9256
0.100	0.8782	0.9814	1.088	1.198	1.309	1.424	1.540	1.660	1.784
0.500	3.891	4.334	4.791	5.261	5.741	6.223	6.722	7.231	7.747

Table II (*Continued*)

m/(mol kg^{-1})	15.00 °C	20.00 °C	25.00 °C	30.00 °C	35.00 °C	40.00 °C	45.00 °C	50.00 °C	55.00 °C
1.000	7.145	7.934	8.750	9.584	10.44	11.30	12.18	13.07	13.98
2.000	12.45	13.79	15.18	16.58	18.02	19.48	20.95	22.45	23.97
3.000	16.37	18.12	19.91	21.74	23.61	25.50	27.43	29.36	31.32
4.000	19.06	21.10	23.20	25.35	27.52	29.72	31.96	34.27	36.52
6.000	21.43	23.86	26.32	28.87	31.47	34.10	36.76	39.49	42.18
8.000	20.97	23.54	26.23	28.95	31.77	34.66	37.55	40.53	43.52
				Potassium Nitrate					
				ρ/(kg m^{-3})					
0.050	1002.34	1001.36	1000.16	998.73	997.09		993.20		988.64
0.100	1005.46	1004.47	1003.24	1001.76	1000.08		996.15		991.54
0.500	1029.92	1028.61	1027.11	1025.40	1023.50		1019.24		1014.33
1.000	1058.62	1057.00	1055.22	1053.26	1051.14		1046.48		1041.28
1.500	1085.53	1083.61	1081.60	1079.46	1077.13		1072.16		1066.68
2.000	1111.35	1109.36	1107.15	1104.77	1102.29		1097.02		1091.33
2.500		1133.15	1130.82	1128.27	1125.68		1120.13		1114.22
3.000		1156.23	1153.82	1151.14	1148.42		1142.70		1136.54
				η/(mN s m^{-2})					
0.050	1.1339	0.9990	0.8889	0.7964	0.7193		0.5964		0.5047
0.100	1.1300	0.9967	0.8870	0.7957	0.7184		0.5967		0.5062
0.500	1.1029	0.9784	0.8750	0.7884	0.7156		0.5993		0.5115
1.000	1.0853	0.9678	0.8696	0.7866	0.7174		0.6038		0.5182
1.500	1.0803	0.9669	0.8716	0.7910	0.7235		0.6128		0.5278
2.000	1.0845	0.9735	0.8802	0.8004	0.7334		0.6246		0.5404
2.500		0.9855	0.8927	0.8138	0.7469		0.6376		0.5540
3.000		1.0022	0.9095	0.8319	0.7636		0.6543		0.5696
				κ/(S m^{-1})					
0.050	0.5111	0.5682	0.6265	0.6863	0.7470		0.8710		0.9973
0.100	0.9761	1.085	1.196	1.311	1.426		1.661		1.902
0.500	4.120	4.574	5.037	5.507	5.985		6.955		7.942
1.000	7.297	8.083	8.887	9.703	10.53		12.21		13.90
1.500	9.962	11.02	12.09	13.19	14.30		16.54		18.79
2.000	12.28	13.56	14.86	16.18	17.52		20.22		22.93
2.500		15.70	17.19	18.70	20.22		23.29		26.38
3.000		17.59	19.23	20.89	22.59		25.96		29.35
				Potassium Bromide					
				ρ/(kg m^{-3})					
0.050	1003.38	1002.45	1001.27	999.85	998.22		994.37		989.82
0.100	1007.56	1006.59	1005.40	1003.94	1002.31		998.43		993.86
0.500	1040.79	1039.60	1038.21	1036.58	1034.79		1030.69		1025.92
1.000	1080.44	1079.00	1077.47	1075.62	1073.63		1069.24		1064.33
2.000	1154.56	1152.74	1150.79	1148.68	1146.44		1141.47		1136.17
3.000	1222.65	1220.52	1218.28	1215.82	1213.26		1207.85		1202.08
4.000	1285.41	1283.01	1280.37	1277.65	1274.87		1269.07		1263.02
				η/(mN s m^{-2})					
0.050	1.1341	0.9993	0.8884	0.7962	0.7187		0.5963		0.5046
0.100	1.1296	0.9961	0.8862	0.7947	0.7177		0.5960		0.5047
0.500	1.0961	0.9721	0.8692	0.7827	0.7095		0.5928		0.5048
1.000	1.0640	0.9495	0.8529	0.7716	0.7021		0.5908		0.5064
2.000	1.0248	0.9229	0.8352	0.7609	0.6969		0.5930		0.5134
3.000	1.0113	0.9162	0.8342	0.7639	0.7029		0.6036		0.5270
4.000	1.0165	0.9253	0.8461	0.7781	0.7189		0.6221		0.5471
				κ/(S m^{-1})					
0.050	0.5505	0.6126	0.6761	0.7407	0.8067		0.9415		1.079
0.100	1.059	1.177	1.298	1.422	1.547		1.804		2.065
0.500	4.872	5.389	5.916	6.450	6.996		8.096		9.222
1.000	9.260	10.19	11.15	12.11	13.09		15.06		17.06
2.000	17.24	18.84	20.48	22.12	23.80		27.12		30.47
3.000	24.14	26.26	28.39	30.56	32.72		37.03		41.38
4.000	29.93	32.42	34.94	37.48	40.00		45.03		50.14
				Cadmium Nitrate					
				ρ/(kg m^{-3})					
0.050	1008.81	1007.83	1006.61	1005.16	1003.49	1001.63	999.60	997.40	995.03
0.100	1018.38	1017.35	1016.07	1014.57	1012.87	1010.98	1008.91	1006.67	1004.27
0.500	1092.75	1091.21	1089.54	1087.75	1085.70	1083.53	1081.21	1078.72	1076.09
1.000	1180.81	1178.77	1176.57	1174.27	1171.85	1169.35	1166.72	1163.92	1161.01
1.500	1263.39	1260.89	1258.31	1255.66	1252.89	1250.06	1247.05	1243.96	1240.79
2.000	1340.85	1338.05	1335.14	1332.16	1329.10	1326.01	1322.71	1319.34	1315.93
3.000	1483.19	1479.75	1476.25	1472.72	1469.15	1465.54	1461.95	1458.14	1454.31
4.000	1603.14	1599.23	1595.61	1591.72	1587.81	1583.85	1579.89	1576.01	1572.01
5.000	1713.72	1709.48	1705.21	1700.85	1696.51	1692.15	1687.85	1683.63	1679.11

Table II (*Continued*)

m/(mol kg^{-1})	15.00 °C	20.00 °C	25.00 °C	30.00 °C	35.00 °C	40.00 °C	45.00 °C	50.00 °C	55.00 °C
				η/(mN s m^{-2})					
0.050	1.1578	1.0199	0.9069	0.8129	0.7336	0.6664	0.6085	0.5582	0.5151
0.100	1.1748	1.0357	0.9215	0.8263	0.7459	0.6780	0.6193	0.5680	0.5243
0.500	1.3177	1.1664	1.0412	0.9360	0.8471	0.7712	0.7057	0.6484	0.5992
1.000	1.5363	1.3629	1.2187	1.0974	0.9940	0.9059	0.8296	0.7627	0.7055
1.500	1.8111	1.6068	1.4368	1.2935	1.1714	1.0669	0.9770	0.8979	0.8302
2.000	2.1563	1.9116	1.7082	1.5367	1.3912	1.2667	1.1596	1.0660	0.9859
3.000	3.1145	2.7460	2.4426	2.1887	1.9745	1.7924	1.6372	1.5020	1.3871
4.000	4.536	3.964	3.496	3.110	2.789	2.521	2.288	2.093	1.926
5.000	6.555	5.664	4.949	4.364	3.882	3.480	3.143	2.855	2.613
				κ/(S m^{-1})					
0.050	0.7405	0.8270	0.9163	1.008	1.101	1.196	1.293	1.392	1.493
0.100	1.367	1.525	1.688	1.854	2.024	2.196	2.373	2.556	2.738
0.500	5.007	5.567	6.138	6.722	7.314	7.916	8.522	9.146	9.764
1.000	7.546	8.371	9.221	10.08	10.96	11.84	12.74	13.64	14.55
1.500	8.702	9.658	10.64	11.64	12.66	13.69	14.73	15.78	16.83
2.000	8.988	9.995	11.03	12.09	13.16	14.25	15.37	16.47	17.59
3.000	8.229	9.216	10.23	11.28	12.36	13.45	14.57	15.70	16.84
4.000	6.956	7.863	8.813	9.796	10.81	11.85	12.90	14.00	15.10
5.000	5.631	6.443	7.300	8.197	9.131	10.09	11.08	12.10	13.15

mol kg^{-1} to near saturation and at temperatures from 15 to 55 °C. In a previous paper (*3*) were presented the experimental data for alkali-metal chlorides, $MgSO_4$, $NiSO_4$, and $ZnSO_4$ in aqueous solutions. This paper deals with the data for alkaline-earth chlorides, $LaCl_3$, Na_2SO_4, $NaNO_3$, NaBr, KNO_3, KBr, and $Cd(NO_3)_2$; the temperature dependences of density, viscosity, and conductivity are represented in terms of their thermal coefficients at 25 °C.

Experimental Section

Reagents. All the chemicals used were reagent grade (Wako Pure Chemicals) and were recrystallized once except $CaCl_2$, $SrCl_2$, and $LaCl_3$, which were recrystallized twice from redistilled water. These electrolytes were dried to appropriate form, and the water content and impurities were determined by chemical analysis.

Solutions were prepared on the molal basis with conductivity water redistilled from a quartz still and boiled just before preparation of solution to expel dissolved air. All the solutions freshly prepared were passed through a G4 sintered glass filter and transferred to measuring cells.

Density Measurements. The densitometers used were of the Weld type, the volume being 25 cm^3 and the diameter of a capillary tube being 0.7 mm. Calibration of the densitometer was made with the density of pure water (Table I) (*4*) at each experimental temperature. The density data listed in Table II are mean values obtained from duplicate measurements by using two densitometers, the difference between duplicate measurements being less than 0.01 kg m^{-3}.

Viscosity Measurements. The viscometers used were of the Ubbelohde type with two different capillary diameters: one (A) for solutions of lower viscosities and the other (B) for those of higher viscosities. Absolute viscosity η was calculated from the relative viscosity by using the relation $\eta = \eta_0\eta_r$, where η_0 is the viscosity of a standard solution and η_r is the relative viscosity of a sample solution. The viscosity of pure water (*5*) at each experimental temperature shown in Table I was used as a standard for viscometer A. When viscometer B was used, however, a solution of the highest concentration of a given electrolyte at a given temperature, the viscosity of which could be measured with viscometer A, was used as a secondary standard, because the flow time of viscometer B for pure water (about 60 s at 55 °C) was too short to be measured accurately. The secondary standards used were 2 mol kg^{-1} solutions of $MgCl_2$ and $CaCl_2$ and a 1 mol kg^{-1} solution of $LaCl_3$. The flow time was determined with a precision of 0.01 s by using a two-channel digital counter with an electrooptical meniscus detection device (Fuso Seisakusho Model 920). In each experimental run, flow times ranged from 200 to 900 s and the difference in flow time by repeated measurements was less than 0.03 s. Kinetic energy correction was made for the relative viscosity according to the equation (*6*)

$$\eta_r = \frac{\rho_1 t_1}{\rho_2 t_2}\left[1 + D\left(\frac{1}{t_2^2} - \frac{1}{t_1^2}\right) + D^2\left(\frac{1}{t_2^4} - \frac{1}{t_1^2 t_2^2}\right)\right] \quad (1)$$

where

$$D = mV^2/(\pi^2 R^4 h_m g) \quad (2)$$

The symbols in eq 1 and 2 have the following meanings: ρ, density; t, flow time; h_m, mean height of the liquid column in the viscometer; R, radius of the viscometer capillary; V, volume of a liquid flowing through the tube in time t; m, numerical coefficient ($\simeq 1$) introduced to take account of the particular shape of the capillary ends; g, acceleration due to gravity (subscripts 1 and 2 refer to the sample and standard solutions, respectively).

Conductivity Measurements. The electrolytic conductivity was measured by the four-electrode cell method (*7*) at a frequency of 125 Hz. No frequency dependence of the conductivity was observed at 60 and 1000 Hz. Pyrex glass cells with different cell constants of 12.95, 50.97, and 139.45 cm^{-1} were used according to the conductivity of solution. The cell constant was determined by using the conductivity of a potassium chloride solution whose composition was 71.1352 or 7.419 13 g of KCl in 1000 g of solution (*8*).

Temperature Control. All the measurements were carried out in a thermostated water bath regulated by a homemade temperature controller with a sensitivity of ±0.001 °C. The temperature of the bath was measured with a quartz crystal thermometer (Hewlett-Packard Model 2801). Temperature fluctuation in the bath during the measurement was less than ±0.01 °C at each experimental temperature.

Results

The experimental values of density ρ, viscosity η, and electrolytic conductivity κ of aqueous solutions of $MgCl_2$, $CaCl_2$, $SrCl_2$, $BaCl_2$, $LaCl_3$, Na_2SO_4, $NaNO_3$, NaBr, KNO_3, KBr, and $Cd(NO_3)_2$ are summarized in Table II.

Table III. Thermal Expansivity, $-10^4[(1/\rho)(d\rho/d\theta)]$, at 25 °C

salts	m/(mol kg^{-1})									
	0.05	0.1	0.5	1.0	2.0	3.0	4.0	5.0	6.0	8.0
$MgCl_2$	2.57	2.57	2.69	2.85	2.85	2.84	2.81	2.78		
$CaCl_2$	2.57	2.61	2.89	3.18	3.54	3.86	4.21		4.61	
$SrCl_2$	2.59	2.63	2.99	3.35	3.70					
$BaCl_2$	2.61	2.67	3.07	3.50						
$LaCl_3$	2.56	2.62	2.86	3.01	3.19	3.23				
Na_2SO_4	2.66	2.76	3.32							
$NaNO_3$	2.66	2.71	3.22	3.75	4.49	4.83	5.19		5.50	5.53
NaBr	2.60	2.65	3.00	3.37	3.90	4.28	4.55		4.95	5.15
KNO_3	2.62	2.68	3.12	3.54	4.11					
KBr	2.58	2.61	2.89	3.13	3.53	3.84	4.14			
$Cd(NO_3)_2$	2.64	2.72	3.21	3.81	4.40	4.76	4.75	5.05		

Table IV. Thermal Coefficient of Viscosity, $10^2[(1/\eta)(d\eta/d\theta)]$, at 25 °C

salts	m/(mol kg^{-1})									
	0.05	0.1	0.5	1.0	2.0	3.0	4.0	5.0	6.0	8.0
$MgCl_2$	-2.29	-2.28	-2.26	-2.26	-2.31	-2.43	-2.62	-2.88		
$CaCl_2$	-2.29	-2.28	-2.20	-2.13	-2.06	-2.07	-2.16		-2.61	
$SrCl_2$	-2.28	-2.26	-2.18	-2.10	-2.03					
$BaCl_2$	-2.28	-2.26	-2.15	-2.06						
$LaCl_3$	-2.28	-2.27	-2.22	-2.20	-2.35	-2.75				
Na_2SO_4	-2.28	-2.28	-2.26							
$NaNO_3$	-2.29	-2.27	-2.21	-2.16	-2.11	-2.11	-2.12		-2.19	-2.28
NaBr	-2.29	-2.28	-2.21	-2.15	-2.09	-2.06	-2.07		-2.17	-2.36
KNO_3	-2.28	-2.27	-2.17	-2.08	-1.96					
KBr	-2.28	-2.27	-2.18	-2.08	-1.93	-1.82	-1.97			
$Cd(NO_3)_2$	-2.28	-2.27	-2.21	-2.18	-2.19	-2.28	-2.44	-2.63		

Table V. Thermal Coefficient of Conductivity, $10^2[(1/\kappa)(d\kappa/d\theta)]$, at 25 °C

salts	m/(mol kg^{-1})									
	0.05	0.1	0.5	1.0	2.0	3.0	4.0	5.0	6.0	8.0
$MgCl_2$	2.04	2.03	1.98	1.97	2.02	2.13	2.33	2.59		
$CaCl_2$	2.00	1.99	1.91	1.85	1.81	1.83	1.93		2.32	
$SrCl_2$	2.01	1.99	1.90	1.83	1.78					
$BaCl_2$	1.98	1.95	1.85	1.77						
$LaCl_3$	2.00	1.97	1.90	1.88	1.96	2.24				
Na_2SO_4	2.09	2.09	2.11							
$NaNO_3$	1.97	1.96	1.93	1.90	1.88	1.89	1.90		1.95	2.01
NaBr	1.99	1.98	1.93	1.88	1.84	1.82	1.83		1.90	2.06
KNO_3	1.88	1.88	1.85	1.82	1.76					
KBr	1.89	1.88	1.79	1.72	1.60	1.51	1.44			
$Cd(NO_3)_2$	1.97	1.95	1.88	1.85	1.89	2.02	2.19	2.40		

The temperature dependence of density ρ, viscosity η, and conductivity κ could be represented with a reasonable accuracy by empirical equations of a polynomial type

$$Y_\theta = Y_{25} + a_1(\theta - 25) + a_2(\theta - 25)^2 + a_3(\theta - 25)^3$$

$$Y \equiv \rho, \eta, \kappa$$

where Y_θ and Y_{25} mean the given physical quantitiy (ρ, η, and κ) at Celsius temperature θ and 25 °C, respectively. From these empirical equations the following thermal coefficients of each quantity at 25 °C were calculated and they are given in Tables III–V:

thermal expansivity

$$\alpha = -\frac{1}{\rho}\frac{d\rho}{d\theta}$$

thermal coefficient of viscosity

$$\beta = \frac{1}{\eta}\frac{d\eta}{d\theta}$$

thermal coefficient of conductivity

$$\gamma = \frac{1}{\kappa}\frac{d\kappa}{d\theta}$$

Acknowledgment

I thank Dr. Y. Takahashi and Dr. K. Takahashi of this Institute for analysis of chemicals and construction of the temperature controller, respectively.

Registry No. Magnesium chloride, 7786-30-3; calcium chloride, 10043-52-4; strontium chloride, 10476-85-4; barium chloride, 10361-37-2; lanthanum chloride, 10099-58-8; sodium sulfate, 7757-82-6; sodium nitrate, 7631-99-4; sodium bromide, 7647-15-6; potassium nitrate, 7757-79-1; potassium bromide, 7758-02-3; cadmium nitrate, 10325-94-7.

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Received for review January 25, 1983. Accepted June 20, 1983.

Solubility in the System $CO(NH_2)_2$–NH_3–H_3PO_4–H_2O at 0, 25, and 50 °C

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The quaternary system $CO(NH_2)_2$–NH_3–H_3PO_4–H_2O was studied at 0, 25, and 50 °C to determine the solubility isotherms in the pH range 1-7. No interaction occurred between urea and the ammonium phosphates, but at low pH the urea-phosphoric acid adduct formed. The solid phase contained urea and mono- or diammonium phosphate throughout most of the pH range studied. An equation was developed to predict P_2O_5 and N solubility over the pH range 2-5.5 and temperature range 0-50 °C.

Introduction

Several papers concerning the solubility of urea in ammonium orthophosphate solutions (*1-4*), as well as its solubility in ammonium polyphosphate solutions (*5*), have appeared in the literature. In the present report the phase relationships in the system urea-ammonia-orthophosphoric acid-water at 0, 25, and 50 °C over a wide pH range (1-7) are reported. This system provides an excellent liquid fertilizer with a high content of plant nutrients.

Experimental Section

Reagent-grade mono- and diammonium phosphate and recrystallized reagent urea were employed in the study. Saturated solutions of mono- and diammonium phosphate were prepared. The solutions were adjusted to the desired pH by adding H_3PO_4 or anhydrous ammonia and then saturating with urea. This saturation was performed at 5 °C above the experimental temperature to assure that the solid phase contained well-formed crystals when the solutions were adjusted to the desired temperature. The solutions (in plastic bottles) were rotated at 4 rpm in a water bath at 25 ± 0.05 °C. The approach to equilibrium was followed by periodic petrographic examination of the solid phase. When it was determined that a salt was absent, a few grams of the missing salt was added and equilibration continued. When all the expected solid phases were present, the liquid phase was sampled for composition and pH. This was repeated at 1-week intervals until two consecutive samplings produced the same results. The 0 °C data were obtained from solutions stored in a cold room at 0.0 ± 0.5 °C and agitated manually at various intervals. The sampling at 50 ± 0.1 °C was at 1-week intervals, except for those solutions at low pH (<3) where hydrolysis began to occur. Here, sampling was conducted daily until consecutive duplicate results were obtained.

Table I. Solubility of Urea in Ammonium Orthophosphate

		liquid-phase composition, %				
solution no.	pH	total N	urea N	NH_3 N	P_2O_5	solid phases[a]
			0 °C			
1	2.04	19.0	17.6	1.4	17.28	A, D
2	2.21	19.7	18.4	1.3	17.12	A, C, D
3	2.80	19.0	17.6	1.4	12.30	A, C
4	3.15	19.0	17.5	1.5	10.55	A, C
5	4.87	18.7	17.0	1.7	8.36	A, C
6	5.42	18.5	16.4	2.1	9.14	A, C
7	6.23	16.1	10.2	5.9	20.15	A, B, C
8	7.24	17.4	13.6	3.8	11.16	B, C
			25 °C			
1	1.86	23.8	22.7	1.1	21.87	A, D
2	2.17	24.2	22.6	1.6	19.01	A, C, D
3	2.59	23.8	22.3	1.5	15.19	A, C
4	3.08	23.9	22.1	1.8	11.19	A, C
5	4.67	23.9	22.0	1.9	9.13	A, C
6	5.24	23.7	21.7	2.0	9.73	A, C
7	6.05	20.5	15.3	5.2	18.90	A, B, C
8	7.02	23.1	19.4	3.7	8.93	B, C
			50 °C			
1	1.42	25.2	24.3	0.9	25.76	A, D
2	1.93	27.1	25.6	1.5	21.21	A, C, D
3	2.10	27.9	26.3	1.6	19.03	A, C
4	2.45	28.1	26.1	2.0	16.07	A, C
5	2.81	28.2	26.3	1.9	13.29	A, C
6	3.22	28.8	26.9	1.9	11.67	A, C
7	4.07	28.2	26.2	2.0	10.23	A, C
8	4.96	29.1	26.9	2.2	9.97	A, C
9	5.21	28.8	26.4	2.4	10.53	A, C
10	5.35	28.9	26.6	2.3	10.78	A, C
11	6.09	26.3	22.1	4.1	16.52	A, B, C
12	7.09	30.1	28.4	1.7	5.43	B, C

[a] A = $NH_4H_2PO_4$; B = $(NH_4)_2HPO_4$; C = $CO(NH_2)_2$; D = $CO(NH_2)_2 \cdot H_3PO_4$.