Enthalpies of Dilution for *myo*-Inositol in Aqueous Alkali Metal Salt and Alkaline Earth Metal Salt Solutions

Dezhi Sun,* Wenqing Zheng, Xiukui Qu, and Ling Li

College of Chemistry and Chemical Technology, Liaocheng University, Wenhua Road, Liaocheng, Shandong 252059, People's Republic of China

The enthalpies of dilution of *myo*-inositol in aqueous solutions of alkali metal salts and alkaline earth metal salts have been determined with flow-mix isothermal microcalorimetry at 298.15 K. The enthalpic interaction coefficients in the range of salt solutions have been calculated according to the McMillan−Mayer theory. The results show that enthalpic pairwise interaction coefficients h_2 of *myo*-inositol are negative in aqueous alkali metal salt and alkaline earth metal salt solutions. The absolute value of h_2 becomes small with an elongation of the radius of the alkali metal cation while it becomes large with an elongation of the radius of the alkaline earth metal cation. The results are discussed in terms of the different metallic ions, interactions of solute with solute and solute with solvent.

Introduction

myo-Inositol (m-Ino), which comprises 95 % of total free inositols in the human body, is a natural constituent of the diet, totaling approximately 1 g·day^{-1}.[1] m-Ino is widely distributed in nature and is considered a growth factor and an indicator of abnormal metabolism or disease.[2] Kiyoshima et al.[3,4] also found contents of m-Ino and mannitol in both human and rat brain tissues. Inositol levels change in association with ischemic state and edema in animal models.[5,6] m-Ino plays multiple important roles in all organisms.[7] Since the main solvent in the body fluid of humans and animals is water in which there are various ions, interactions of m-Ino−m-Ino, m-Ino−water, and m-Ino ions must exist that depend on the thermodynamic character of the systems. Therefore, study of the thermodynamic character of the polylol in aqueous solutions is as important as research on ions[8−10] that exist in biochemical systems. In our earlier work,[11,12] the dilution enthalpies of D-sorbitol and D-mannitol were determined in aqueous sodium halide solutions and various concentrations of sodium chloride solutions. Information about the interaction coefficients of the virial expansion of an excess thermodynamic property was obtained.[13] In this presentation, the dilution enthalpies have been measured for m-Ino in aqueous solutions of alkali metal salts and alkaline earth metal salts at 298.15 K, and the homogeneous enthalpic interaction coefficients of the polylol have been obtained. The change tendency of pairwise enthalpic interaction coefficients is interpreted in terms of solute−solute and solute−solvent interactions according to the McMillan−Mayer theory.

Experimental Section

Reagents. m-Ino was purchased from Aldrich, and the stated purity was better than 99 %. It was dried under reduced pressure at 323 K before use. Lithium chloride (LiCl), sodium chloride (NaCl), potassium chloride (KCl), magnesium chloride ($MgCl_2$), calcium chloride ($CaCl_2$), strontium chloride ($SrCl_2$), and barium chloride ($BaCl_2$) were all analytical reagents (purity > 99 %) purchased from the Shanghai Chemical Reagent Company (Shanghai, China), recrystallized from distilled water and dried under reduced pressure at 393 K before use. All of the reagents were stored over P_2O_5 in a vacuum desiccator for 72 h at room temperature prior to use. Twice-distilled water was deionized by passing through a quartz sub-boiling purifier before use in the preparation of solutions. Solutions were prepared by mass using a Mettler AE 200 balance, precise to ± 0.0001 g. The molality range of m-Ino used was (0.10 to 0.50) mol·kg^{-1}. The molality of each alkali metal salt in water was 0.200 mol·kg^{-1} and of that each alkaline earth metal salt was 0.100 mol·kg^{-1}. These electrolyte solutions were used as solvents in addition to water. All of the solutions were degassed with ultrasonic waves and used within 12 h of preparation, and the uncertainty of the concentration was ± 0.0002 mol·kg^{-1}.

Calorimetric Procedure. The dilution enthalpies were determined with a 2277-204 measuring cylinder supported by a TAM 2277 thermal activity monitor[14] (Thermometric, Sweden) at 298.15 K. The solutions of m-Ino and their solvents were pumped through the mixing-flow vessel of the calorimeter using VS2-10R MIDI dual-channel pumps. The flow rates were determined from the mass of the samples delivered in 6 min. The variation in flow rates was less than 0.1 % both before and after a complete dilution experiment. Thermal effects of the dilution process were obtained with the following method.

The liquids passing through channels A and B were changed in the following sequence:

(1) A (aqueous salt solution) + B (aqueous salt solution)− baseline determination;

(2) A (aqueous salt solution) + B (aqueous m-Ino solution)− dilution thermal power determination; and

(3) A (aqueous salt solution) + B (aqueous salt solution)− baseline re-established.

The enthalpies of dilution $\Delta_{dil}H_m$ were calculated from the equation[15]

$$\Delta_{dil}H_m = -P(1 + m_iM)/m_i f_2 \qquad (1)$$

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* Corresponding author. Tel.: +86-635-8230614. Fax: +86-635-8239196. E-mail: sundezhisdz@163.com or sundezhi@lcru.edu.cn.

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Table 1. Enthalpies of Dilution of *myo*-Inositol in Water and Alkali Metal Salt Solutions at 298.15 K

m_i	m_f	$\Delta_{dil,exp}H_m$	$\Delta_{dil,cal}H_m$	m_i	m_f	$\Delta_{dil,exp}H_m$	$\Delta_{dil,cal.}H_m$	m_i	m_f	$\Delta_{dil,exp}H_m$	$\Delta_{dil,cal}H_m$
mol·kg^{-1}	mol·kg^{-1}	J·mol^{-1}	J·mol^{-1}	mol·kg^{-1}	mol·kg^{-1}	J·mol^{-1}	J·mol^{-1}	mol·kg^{-1}	mol·kg^{-1}	J·mol^{-1}	J·mol^{-1}
Water											
0.1199	0.0612	45.73	45.76	0.2622	0.1329	88.62	88.51	0.4046	0.2041	128.17	128.26
0.1482	0.0757	54.36	54.31	0.2883	0.1465	96.17	96.23	0.4357	0.2194	135.74	135.78
0.1750	0.0892	63.03	63.06	0.3158	0.1600	104.03	104.11	0.4671	0.2343	143.69	143.66
0.2039	0.1039	71.23	71.28	0.3452	0.1744	112.18	112.12	0.4996	0.2512	150.53	150.58
0.2314	0.1177	79.58	79.42	0.3773	0.1902	121.16	121.14				
LiCl/0.200 mol·kg^{-1}											
0.1208	0.0623	43.64	43.58	0.2580	0.1319	84.62	84.57	0.4032	0.2046	123.88	123.92
0.1466	0.0754	51.83	51.76	0.2873	0.1470	92.05	92.11	0.4286	0.2174	129.77	129.74
0.1745	0.0895	60.09	60.11	0.3172	0.1620	100.58	100.46	0.4629	0.2340	138.79	138.75
0.2029	0.1040	68.80	68.76	0.3463	0.1763	108.52	108.61	0.4968	0.2508	146.46	146.51
0.2292	0.1172	76.66	76.46	0.3753	0.1906	116.75	116.78				
NaCl/0.200 mol·kg^{-1}											
0.1126	0.0565	36.58	36.54	0.2541	0.1267	76.49	76.51	0.3997	0.1970	113.32	113.35
0.1416	0.0709	45.61	45.63	0.2810	0.1400	82.72	82.75	0.4313	0.2133	119.88	119.89
0.1676	0.0839	53.19	53.17	0.3110	0.1549	89.93	89.90	0.4990	0.2464	133.91	133.96
0.1952	0.0977	60.82	60.85	0.3405	0.1687	98.09	98.03				
0.2268	0.1131	69.29	69.32	0.3658	0.1812	104.10	104.05				
KCl/0.200 mol·kg^{-1}											
0.1218	0.0602	39.72	39.70	0.2603	0.1287	77.54	77.51	0.4035	0.1979	112.32	112.22
0.1462	0.0724	46.94	46.92	0.2896	0.1429	85.28	85.29	0.4369	0.2139	119.40	119.35
0.1745	0.0863	55.06	55.04	0.3162	0.1557	91.70	91.74	0.4988	0.2434	132.49	132.51
0.2045	0.1014	62.89	62.83	0.3463	0.1705	98.51	98.46				
0.2302	0.1140	69.61	69.45	0.3758	0.1844	106.26	106.23				

where P is the dilution thermal power (μW), M is the molar mass of one of the polylols, m_i is the initial molality of m-Ino, and f_2 is the flow rate of the m-Ino solutions. Uncertainty of $\Delta_{dil}H_m$ calculated according to eq 1 was within ± 1 % based on calorimetric data of three iterations.

The final molality m_f, which is a necessary parameter in data processing, can be calculated from the equation

$$m_f = m_i\, f_2/[f_1(1 + m_iM) + f_2] \qquad (2)$$

where f_1 is the flow rate of the diluents (water or aqueous sodium halide solutions).

Results and Discussion

The molar excess enthalpy of a solution with molality m can be written as a power series of m with coefficients h_n by[16,17]

$$H^E_m = H^E/m = L_{2\varphi} = h_2m + h_3m^2 + h_4m^3 + ... \qquad (3)$$

where $L_{2\varphi}$ is the relative apparent molar enthalpy. The enthalpic interaction coefficients (h_n), which are related to the McMillan–Mayer coefficients, measure the enthalpic interactions mediated by the solvent occurring between pairs, triplets, and higher order multiplets of solute species. The molar enthalpy of dilution $\Delta_{dil}H_m$ in terms of the excess enthalpy at the initial molality m_i and final molality m_f is

$$\Delta_{dil}H_m = H^{exp}_m(m_f) - H^{exp}_m(m_i) = h_2(m_f - m_i) + h_3(m_f^{\,2} - m_i^{\,2}) + h_4(m_f^{\,3} - m_i^{\,3}) + ... \qquad (4)$$

where $H^{exp}_m(m_f)$ and $H^{exp}_m(m_i)$ are the molar excess (relative apparent molar) enthalpies of the solute in the solutions before and after dilution.

In this work, pure water and the salt solutions are regarded wholly as "solvents". Tables 1 and 2 give the experimental values of dilution enthalpies ($\Delta_{dil,exp}H_m$), which were fitted to eq 4 by a least-squares procedure using Origin 6.0 software with the intercept locked to 0, together with the corresponding calculated values ($\Delta_{dil,cal}H_m$) along with the initial and final molality (m_i and m_f) of m-Ino in different solvents.

Tables 3 and 4 list the coefficients of eq 4 that were obtained from a least-squares analysis of these results. Since it is difficult to interpret the higher order h_n coefficients, only the enthalpic pairwise coefficient h_2 is discussed here. The values for h_2 relate to the measurement of the interaction energy between a pair of solute molecules,[17] which may take place with the coexistent electrolyte in the aqueous solution. Figure 1 shows the variations of h_2 coefficients for m-Ino within water and solutions of three aqueous alkali metal salts. Figure 2 shows similar variations in pure water and aqueous solutions of four alkaline earth metal salts.

From Figures 1 and 2 it can be seen that no matter whether in pure water or in solutions of metal halides, the values of the pairwise enthalpic coefficients, h_2, for m-Ino are negative. These results indicate that energy is released when the molecules of m-Ino approach each other. The value of h_2 for m-Ino in a solution of each salt is different from that in water. This kind of difference can be attributed to the different metal cations.

It can be seen clearly in Figure 1 that the absolute values of h_2 in solutions of the halides of the three alkali metals are all larger than that in pure water. This fact is evidently related to electrostatic interaction of ions with polar groups of the polylol molecule. The curve in Figure 1 also indicates that the algebraic value of h_2 for m-Ino increases with the enlargement of the cations. This important trend might be explained by the structure of the cations and a change of hydration of the solute. As is known, in aqueous solutions, hydration sheaths with a defined number of water molecules are formed around dissolved molecules or ions.[18] Mutual interaction between molecules or ions may cause some water molecules to be squeezed out from their hydration sheaths into the typical bulk water.[19] The lithium ion has the smallest radius of the alkali ions. Therefore, it will exert the largest attractive force on the water molecules. This makes it easier to break the hydration layer around the polylol molecule when two m-Ino molecules approach each other in the LiCl solution than in other alkali chloride solutions at the same concentration. The ability that makes the m-Ino molecule

Table 2. Enthalpies of Dilution of *myo*-Inositol in Water and Alkaline Earth Metal Salt Solutions at 298.15 K

m_i / mol·kg^{-1}	m_f / mol·kg^{-1}	$\Delta_{dil,exp}H_m$ / J·mol^{-1}	$\Delta_{dil,cal}H_m$ / J·mol^{-1}	m_i / mol·kg^{-1}	m_f / mol·kg^{-1}	$\Delta_{dil,exp}H_m$ / J·mol^{-1}	$\Delta_{dil,cal.}H_m$ / J·mol^{-1}	m_i / mol·kg^{-1}	m_f / mol·kg^{-1}	$\Delta_{dil,exp}H_m$ / J·mol^{-1}	$\Delta_{dil,cal}H_m$ / J·mol^{-1}
Water											
0.1199	0.0612	45.73	45.76	0.2622	0.1329	88.62	88.51	0.4046	0.2041	128.17	128.26
0.1482	0.0757	54.36	54.31	0.2883	0.1465	96.17	96.23	0.4357	0.2194	135.74	135.78
0.1750	0.0892	63.03	63.06	0.3158	0.1600	104.03	104.11	0.4671	0.2343	143.69	143.66
0.2039	0.1039	71.23	71.28	0.3452	0.1744	112.18	112.12	0.4996	0.2512	150.53	150.58
0.2314	0.1177	79.58	79.42	0.3773	0.1902	121.16	121.14				
$MgCl_2$/0.100 mol·kg^{-1}											
0.1074	0.0539	38.25	38.26	0.2486	0.1242	80.75	80.76	0.3935	0.1947	118.90	118.82
0.1351	0.0676	47.87	47.85	0.2774	0.1385	88.92	88.84	0.4239	0.2092	126.06	126.17
0.1635	0.0818	56.85	56.88	0.3063	0.1521	97.04	97.06	0.4555	0.2250	132.62	132.54
0.1909	0.0956	64.88	64.83	0.3371	0.1673	104.54	104.51	0.5026	0.2481	142.89	142.76
0.2189	0.1095	72.55	72.58	0.3632	0.1802	111.01	111.09				
$CaCl_2$/0.100 mol·kg^{-1}											
0.1102	0.0554	37.47	37.52	0.2484	0.1237	78.14	78.16	0.3914	0.1934	113.16	113.12
0.1354	0.0680	45.72	45.76	0.2758	0.1370	85.58	85.52	0.4240	0.2094	120.94	120.90
0.1647	0.0824	55.43	55.38	0.3047	0.1512	92.53	92.48	0.4999	0.2460	136.14	136.08
0.1903	0.0949	62.71	62.75	0.3343	0.1654	100.19	100.23				
0.2189	0.1092	70.85	70.81	0.3633	0.1798	106.89	106.96				
$SrCl_2$/0.100 mol·kg^{-1}											
0.1147	0.0581	39.44	39.42	0.2529	0.1272	78.84	78.84	0.4050	0.2024	113.53	113.53
0.1428	0.0721	48.80	48.76	0.2816	0.1416	84.77	84.78	0.4338	0.2161	120.24	120.21
0.1707	0.0860	57.65	57.67	0.3130	0.1571	92.81	92.86	0.4999	0.2485	134.49	134.45
0.1955	0.0986	64.10	64.11	0.3420	0.1714	99.50	99.49				
0.2240	0.1128	70.89	70.86	0.3722	0.1863	106.77	106.77				
$BaCl_2$/0.100 mol·kg^{-1}											
0.1239	0.0616	39.12	39.11	0.2606	0.1288	77.48	77.45	0.4081	0.1998	110.90	110.88
0.1479	0.0735	48.37	48.35	0.2905	0.1434	84.36	84.36	0.4382	0.2145	116.74	116.72
0.1786	0.0886	57.39	57.38	0.3213	0.1580	92.59	92.57	0.5018	0.2453	129.26	129.24
0.2039	0.1010	64.05	64.16	0.3485	0.1710	98.81	98.81				
0.2298	0.1135	70.38	70.32	0.3781	0.1856	104.54	104.55				

Table 3. Enthalpies Interaction Coefficients of *myo*-Inositol in Pure Water and Aqueous Alkali Metal Salt Solutions at 298.15 K

solvent	solute	h_2/J·kg·mol^{-2}	h_3/J·kg^2·mol^{-3}	R^2	SD
H_2O	*myo*-inositol	−751.86 (± 46.0)[a]	221.13 (± 106)[a]	0.9999[b]	0.38[b]
LiCl/0.2 mol·kg^{-1}	*myo*-inositol	−691.18 (± 31)[a]	150.65 (± 72)[a]	0.9998[b]	0.26[b]
NaCl/0.2 mol·kg^{-1}	*myo*-inositol	−672.35 (± 52.7)[a]	250.37 (± 124)[a]	0.9998[b]	0.48[b]
KCl/0.2 mol·kg^{-1}	*myo*-inositol	−649.12 (± 25)[a]	199.45 (± 57)[a]	0.9996[b]	0.21[b]

[a] Estimated from triplicate experiments. [b] Parameters for eq 4.

Table 4. Enthalpies Interaction Coefficients of *myo*-Inositol in Pure Water and Aqueous Alkaline Earth Metal Salt Solutions at 298.15 K

solvent	solute	h_2/J·kg·mol^{-2}	h_3/J·kg^2·mol^{-3}	R^2	SD
H_2O	*myo*-inositol	−751.86 (± 46)[a]	221.13 (± 106)[a]	0.9999[b]	0.38[b]
$MgCl_2$/0.1 mol·kg^{-1}	*myo*-inositol	−763.82 (± 27)[a]	352.57 (± 66)[a]	0.9994[b]	0.28[b]
$CaCl_2$/0.1 mol·kg^{-1}	*myo*-inositol	−795.5 (± 38)[a]	473.76 (± 91)[a]	0.9998[b]	0.37[b]
$SrCl_2$/0.1 mol·kg^{-1}	*myo*-inositol	−830.4 (± 50)[a]	630.99 (± 117)[a]	0.9998[b]	0.46[b]
$BaCl_2$/0.1 mol·kg^{-1}	*myo*-inositol	−864.62 (± 76)[a]	710.09 (± 175)[a]	0.9996[b]	0.64[b]

[a] Estimated from triplicate experiments. [b] Parameters for eq 4.

partly dehydrated varies as $Li^+ > Na^+ > K^+$; therefore, the absolute value of the pairwise interaction coefficient (h_2) for m-Ino increases in the order h_2 (LiCl solution) > h_2 (NaCl solution) > h_2 (KCl solution) (Figure 1). In the other words, the larger the size of the alkali metal cation, the weaker is the interaction between it and water molecules, and the more difficult for the m-Ino molecule to dehydrate.

The curve in Figure 2 shows that (1) the absolute values of h_2 are evidently larger than those determined for m-Ino in alkali-metal salts solutions (Figure 1) and (2) the absolute value of h_2 becomes larger with an elongation of the radius of the alkaline earth metal cation at the same positive charge concentration of above-mentioned solutions. This phenomenon might be explained as following. Bivalent alkaline earth metal ions cannot only partly destroy the secondary hydration layer but also affect the primary hydration layer through its stronger interaction with the m-Ino molecule due to its larger electric charge. This makes it easier to break the hydration layer around the polylol molecule when two m-Ino molecules approach each other. So the algebraic value of h_2 for m-Ino in the solution of each of the alkaline earth metal chlorides is less than that in water. The ability to destroy the hydration layer of the polylol molecule should vary as $Mg^{2+} > Ca^{2+} > Sr^{2+} > Ba^{2+}$, only if the radii of the alkaline earth ions are considered, and the absolute value of h_2 for the polylol would increase in the same order. However, the experimental result is opposite (Figure 2). This phenomenon might be interpreted from the dispersion force of ions upon the solute molecules, which can also lead to partial desolvation of solutes, and reorganization of the hydration structure around m-Ino. The larger the size of the cation is, the stronger the dispersion force will be, so the easier the hydration layers around the polylol molecules will be partly disrupted when they approach each other, and the more negative will be the contribution to $h_{2.}$ Consequently, values of h_2 for m-Ino in

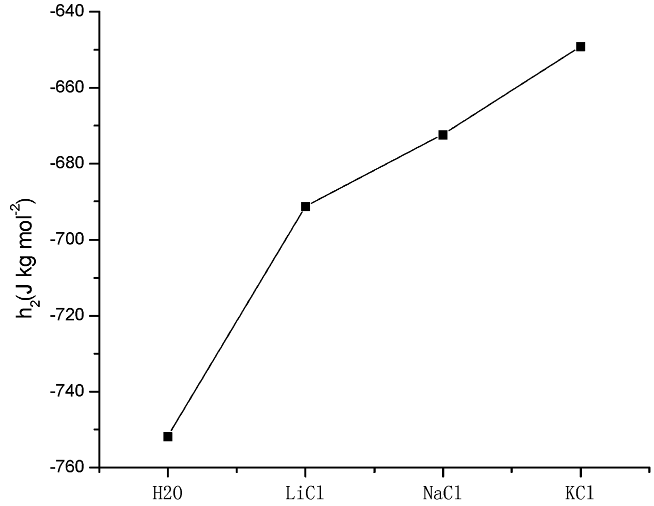

Figure 1. Variation of homogeneous enthalpic pairwise interaction coefficients of *myo*-inositol (■) in pure water and aqueous alkali metal salt solutions (0.200 mol·kg^{-1}) at 298.15 K.

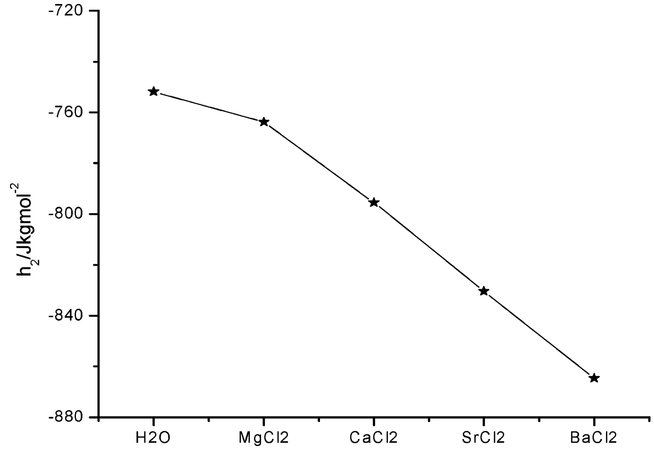

Figure 2. Variation of homogeneous enthalpic pairwise interaction coefficients of *myo*-inositol(★) in pure water and aqueous alkaline earth metal salt solutions (0.100 mol·kg^{-1}) at 298.15 K.

solutions of alkali metal chlorides are less than those in alkaline earth metal chlorides and the barium ion has the strongest dispersion force of the cations, which results in the most negative contribution to h_2 for m-Ino through the dispersion force. Nevertheless, the dispersion force is much weaker than such forces as hydrogen bonding and ion−dipole interactions,[20,21] so the trend of h_2 shown in Figure 1 or Figure 2 is not only dependent on the dispersion force.

Conclusions

Heats of dilution of m-Ino with water and aqueous salt solutions were measured by isothermal flow mixing microcalorimetry at 298.15 K. Experimental enthalpies of dilution that varied with the concentration of the polylols were correlated with a virial expansion equation. Enthalpic interaction coefficients (h_2, h_3, h_4) in the equations were obtained and the values of the pairwise enthalpic interaction coefficient (h_2) have been discussed. The different salt solutions make different contributions to h_2. The change trend of h_2 with respect to the radius of alkali metal ion is opposite to that with the radius of alkaline earth metal ion, which is discussed according to interactions of the polylol with water and the salts.

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