CONCENTRATION EFFECT OF SODIUM CHLORIDE ON ENTHALPIC INTERACTION COEFFICIENTS OF *D*-MANNITOL AND *D*-SORBITOL IN AQUEOUS SOLUTION

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The dilution enthalpies of *D*-mannitol and *D*-sorbitol in aqueous sodium chloride solution at various concentrations have been determined by isothermal microcalorimetry at 298.15 K. The homogeneous enthalpic interaction coefficients over a quite large range of concentration of aqueous sodium chloride solutions have been calculated according to the excess enthalpy concept. The results show that enthalpic pairwise interaction coefficients (h_2) of *D*-mannitol and *D*-sorbitol are positive in aqueous sodium chloride solution and become more positive with increase of the concentration of sodium chloride. The results are interpreted in terms of the different conformations of the two polyols, solute–solute and solute–solvent interactions involved by solvent effects.

Keywords: dilution enthalpy, enthalpic pairwise interaction coefficients, D-mannitol, sodium chloride, D-sorbitol, structural interaction

Introduction

D-Mannitol, clinically used as a hyperosmolar solution in the treatment of elevated intracranial pressure in brain trauma, can reduce brain water content by establishing osmotic gradient [1–3]. It is also used as a diuretic agent [4]. As an important isomer of *D*-mannitol, *D*-sorbitol is derived from glucose by the enzyme aldose reductase in many cells of human and animal body [5, 6]. Its function and clinic application are similar to those of *D*-mannitol. On the other hand, it is well known that sodium chloride is a kind of electrolyte that bears an extraordinary important function in life system. So study on the interaction between sodium chloride and *D*-mannitol or *D*-sorbitol in water is valuable for interpreting the influence of electrolyte on some physiological behavior, and helpful in understanding the essence of some biological phenomena [7–9].

It is quite effective to investigate the interaction of electrolytes with non-electrolytes in water through enthalpy effect measurements, and many studies have been carried out in this field [10–12]. On the other hand, isothermal microcalorimetry has become an extremely powerful technique capable of measuring weak interaction of bio-chemical species in solution [13, 14]. In our previous work, we reported the enthalpic interaction of *D*-sorbitol and *D*-mannitol in water and aqueous sodium halide solutions at 313.15 K [15], which revealed the rule about influence of halogen anion on the values of the homogeneous enthalpic pairwise interaction coefficients of *D*-sorbitol and *D*-mannitol. In the present work, we mainly investigate

the influence of concentration of the salt in aqueous solution on the enthalpy interaction parameters, which provide more important information on the interaction of sodium chloride with *D*-mannitol and *D*-sorbitol in water at the constant temperature (298.15 K).

Experimental

Materials

Both *D*-mannitol and *D*-sorbitol were purchased from Aldrich, and the purity was better than 99%. They were dried under reduced pressure at 323 K before used. The purity was detected with examination of optical rotation ($[\alpha]_D^{20}$). The experimental data determined in this laboratory for *D*-mannitol and *D*-sorbitol were $[\alpha]_D^{20}=$ –0.48±0.02 (–0.49 in the literature [16]) and $[\alpha]_D^{20}=$ 1.99±0.02 (–1.98 in the literature [17]) respectively. Sodium chloride (analytical reagent, mass fraction >99%, from Shanghai Chem. Co.) was recrystallized from distilled water and dried under reduced pressure at 353 K. All the reagents were stored over P_2O_5 in a vacuum desiccator for 72 h at the room temperature prior to use.

Twice distilled water was deionized by passing through a quartz sub-boiling purifier before use in preparation of solutions. Both the aqueous salt solutions, which were used as mixed solvents (water+sodium chloride) and the *D*-mannitol or *D*-sorbitol solutions (*D*-mannitol or *D*-sorbitol+sodium chloride+water) were prepared by mass using a Mettler AE 200 balance precise to ±0.0001 g. The molality range of aqueous so-

dium chloride solutions is 0–1.2 mol kg^{-1}. The molality ranges of aqueous *D*-mannitol and *D*-sorbitol solutions are 0.10–0.55 and 0.10–0.58 mol kg^{-1}, respectively. All the solutions were degassed with ultrasonic wave and used within 12 h after preparation to minimize decomposition due to bacterial contamination.

Calorimetric procedure

The enthalpies of dilution for *D*-mannitol or *D*-sorbitol in aqueous sodium chloride solutions were measured with 2277-204 Measuring Cylinder supported by Thermometric 2277 Thermal Activity Monitor [18] (Thermometric, Sweden) at 298.15 K. The performance of the apparatus and the details of its construction have been previously described [19]. The solutions of *D*-sorbitol and *D*-mannitol and their solvents were pumped through the mixing-flow vessel of the calorimeter using a VS2-10R MIDI dual channel pumps. The variation of flow rates was less than 0.1%. The flow rates were determined by the mass of the samples delivered in 6 min. The variation in rates was less than 0.1% both before and after a complete dilution experiment. The details of the experiment technique have been described elsewhere [20, 21].

The enthalpies of dilution $\Delta_{dil}H_m$ were calculated from the equation [22]

$$\Delta_{dil} H_m = -\frac{P(1 + m_i M)}{m_i f_2} \quad (1)$$

where P is the dilution thermal power (μW), m_i is the initial molality of the polyol, M is the molar mass of the polyol and f_2 is the flow rate of *D*-sorbitol or *D*-mannitol solutions. The final molality m_f were calculated from the equation

$$m_f = \frac{m_i f_2}{f_1(1 + m_i M) + f_2} \quad (2)$$

where f_1 is the flow rate of diluents (water or aqueous sodium chloride solutions). Uncertainty of $\Delta_{dil}H_m$ calculated according to Eqs (1) and (2) was within ±1% based on data of three iterations.

Results and discussion

According to the McMillan–Mayer theory [23, 24], all the thermodynamic properties of multi-components solutions can be expressed by using a viral expansion in m that relates the non-ideal contributions of any total thermodynamic function to a series of interactions parameters. If aqueous sodium chloride solution is regarded as 'solvent', the excess enthalpy per kg of solvent (H^E) of a solution containing a single *D*-mannitol or *D*-sorbitol at molality m is given by:

$$H^E = h_2 m^2 + h_3 m^3 + h_4 m^4 + \ldots \quad (3)$$

where h_2, h_3, h_4, etc. are enthalpic coefficients representing pairwise and, at least notionally, triplet, quarter, and higher order interactions between solvated solute species. The molar enthalpy change ($\Delta_{dil}H_m$) on diluting a solution of non-electrolytic solute from an initial molality m_i to a final molality m_f can be written as:

$$\Delta_{dil} H_m = H_m^E(m_f) - H_m^E(m_i) \quad (4)$$
$$= h_2(m_f - m_i) + h_3(m_f^2 - m_i^2) + h_4(m_f^3 - m_i^3) + \ldots$$

where $H_m^E(m_i)$ and $H_m^E(m_f)$ are the molar excess enthalpies of the solute in the solutions before and after dilution.

Tables 1–2 give the experimental values ($\Delta_{dil}H_m$), together with the initial and final molalities (m_i and m_f) of the polyols in various aqueous sodium chloride solutions. Tables 3–4 list the coefficients of Eq. (4) that were obtained from least-squares analysis of above results. Since it is difficult to interpret the higher order h coefficients, only the enthalpic pairwise coefficients h_2 is discussed here. The values of h_2 for *D*-sorbitol and *D*-mannitol at 298.15 K in pure water are 28.41, 106.58 J kg mol^{-2}, respectively, which are much smaller than those obtained from the previous work [15] where the values of h_2 for *D*-sorbitol and *D*-mannitol in pure water at 313.15 K are 104.6, 153.2 J kg mol^{-2}, respectively. This difference indicates that more energy will be absorbed when two polyol molecules approach each other at higher temperature. Figure 1 shows the variations of h_2 coefficients for both *D*-mannitol and *D*-sorbitol with the increasing molalities of aqueous
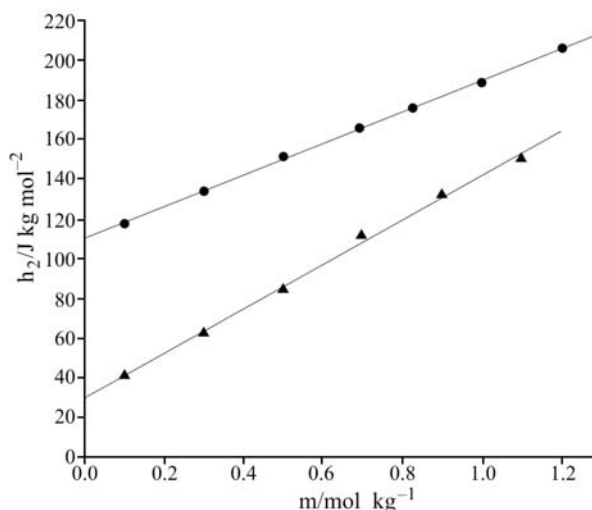

Fig. 1 Variation in enthalpic pairwise interaction coefficients (h_2) of *D*-mannitol and *D*-sorbitol with the molality (m) of sodium chloride in aqueous sodium chloride solutions at 298.15 K: ● – *D*-mannitol, ▲ – *D*-sorbitol

Table 1 Enthalpies of dilution of *D*-mannitol in aqueous sodium chloride solutions at 298.15 K

m (NaCl)/mol kg^{-1}	m_i/mol kg^{-1}	m_f/mol kg^{-1}	$\Delta_{dil}H_m$/J mol^{-1}	m_i/mol kg^{-1}	m_f/mol kg^{-1}	$\Delta_{dil}H_m$/J mol^{-1}
	0.0961	0.0428	−3.37	0.3210	0.1410	−15.26
	0.1234	0.0549	−4.90	0.3501	0.1533	−16.93
	0.1545	0.0683	−6.55	0.3820	0.1669	−18.65
0.0000	0.1829	0.0811	−8.19	0.4096	0.1790	−20.16
	0.2123	0.0940	−9.53	0.4386	0.1911	−21.75
	0.2491	0.1099	−11.59	0.4662	0.2016	−23.38
	0.2848	0.1255	−13.32			
	0.0950	0.0447	−2.98	0.3061	0.1432	−13.67
	0.1244	0.0588	−4.52	0.3387	0.1584	−15.13
	0.1523	0.0719	−5.95	0.3604	0.1687	−16.16
0.0998	0.1823	0.0860	−7.43	0.3919	0.1833	−17.93
	0.2146	0.1011	−9.07	0.4269	0.1990	−19.74
	0.2411	0.1136	−10.39	0.5022	0.2328	−24.08
	0.2741	0.1287	−12.00			
	0.0538	0.0252	−1.60	0.2894	0.1337	−15.85
	0.0862	0.0403	−3.83	0.3263	0.1501	−18.00
	0.1156	0.0540	−5.61	0-3601	0.1658	−19.83
0.3000	0.1494	0.0698	−7.62	0.3908	0.1792	−21.88
	0.1827	0.0848	−9.65	0.4186	0.1917	−23.43
	0.2203	0.1023	−11.70	0.4579	0.2086	−26.08
	0.2520	0.1166	−13.63			
	0.1080	0.0524	−6.47	0.3069	0.1439	−20.45
	0.1448	0.0682	−9.53	0.3396	0.1590	−22.64
	0.1727	0.0815	−11.47	0.3712	0.1735	−24.71
0.5003	0.1990	0.0938	−13.20	0.4088	0.1905	−27.28
	0.2248	0.1056	−14.81	0.4352	0.2024	−29.02
	0.2495	0.1174	−16.68	0.4936	0.2290	−32.83
	0.2787	0.1310	−18.58			
	0.0881	0.0403	−5.60	0.3346	0.1511	−25.26
	0.1270	0.0580	−9.15	0.3735	0.1683	−28.09
	0.1680	0.0766	−12.43	0.4128	0.1855	−31.38
0.6992	0.1967	0.0894	−14.60	0.4486	0.2008	−34.07
	0.2278	0.1035	−16.88	0.4817	0.2152	−36.54
	0.2672	0.1213	−20.00	0.4976	0.2223	−37.68
	0.3005	0.1359	−22.68			
	0.0716	0.0327	−6.23	0.3390	0.1522	−26.83
	0.1092	0.0497	−9.95	0.3821	0.1707	−30.53
	0.1458	0.0659	−12.79	0.4179	0.1865	−33.08
0.8262	0.1834	0.0830	−15.39	0.4541	0.2019	−36.02
	0.2219	0.1001	−18.25	0.4821	0.2139	−38.34
	0.2594	0.1169	−20.87	0.5107	0.2273	−40.79
	0.2969	0.1333	−23.91			
	0.0665	0.0293	−5.99	0.3056	0.1322	−26.76
	0.0970	0.0426	−9.13	0.3372	0.1458	−29.00
	0.1273	0.0554	−11.76	0.4026	0.1726	−34.87
1.0000	0.1666	0.0732	−15.02	0.4417	0.1900	−37.90
	0.1963	0.0856	−17.42	0.4814	0.2058	−41.91
	0.2322	0.1011	−20.38	0.5098	0.2183	−44.62
	0.2663	0.1157	−23.37			
	0.0798	0.0348	−7.01	0.3092	0.1328	−30.62
	0.1164	0.0507	−11.00	0.3462	0.1486	−34.25
	0.1430	0.0622	−13.98	0.3839	0.1641	−38.04
1.2034	0.1733	0.0752	−16.69	0.4212	0.1796	−41.53
	0.2067	0.0894	−20.63	0.4378	0.1867	−43.34
	0.2410	0.1041	−23.69	0.4943	0.2090	−49.16
	0.2744	0.1182	−26.88			

sodium chloride solutions. There is a relatively good linear relation between the pairwise coefficients h_2 and the concentration of sodium chloride aqueous solution. lution in which *D*-mannitol or *D*-sorbitol is diluted. The relations were obtained with the method of least-squares as follows:

$$h_2 \text{ [J kg mol}^{-2}] = 110.37(\pm 0.69^*) + 79.35(\pm 0.93^*)m \text{ (NaCl)} \tag{5}$$

(*D*-mannitol–NaCl–H$_2$O   S.D.**=0.8842)

$$h_2 \text{ [J kg mol}^{-2}] = 29.63(\pm 0.98^*) + 112.34(\pm 1.49^*)m \text{ (NaCl)} \tag{6}$$

(*D*-sorbitol–NaCl–H$_2$O   S.D.**=2.4267)

(*The values in parentheses are the experimental errors; **standard deviation.)

Table 2 Enthalpies of dilution of *D*-sorbitol in aqueous sodium chloride solutions at 298.15 K

m (NaCl)/mol kg^{-1}	m_i/mol kg^{-1}	m_f/mol kg^{-1}	$\Delta_{dil}H_m$/J mol^{-1}	m_i/mol kg^{-1}	m_f/mol kg^{-1}	$\Delta_{dil}H_m$/J mol^{-1}
0.0000	0.1337	0.0651	1.12	0.2798	0.1369	0.26
	0.1580	0.0773	0.94	0.3050	0.1493	0.14
	0.1832	0.0895	0.78	0.3286	0.1612	0.03
	0.2060	0.1008	0.64	0.3572	0.1751	–0.12
	0.2337	0.1143	0.50	0.3908	0.1914	–0.30
	0.2558	0.1250	0.37	0.4253	0.2084	–0.52
0.1000	0.1153	0.0486	0.52	0.3641	0.1506	–4.06
	0.1528	0.0641	–0.20	0.3964	0.1636	–4.68
	0.1900	0.0796	–0.88	0.4302	0.1771	–5.43
	0.2239	0.0933	–1.44	0.4696	0.1928	–6.20
	0.2565	0.1061	–2.06	0.5158	0.2109	–7.32
	0.2928	0.1216	–2.80	0.5615	0.2277	–8.62
	0.3306	0.1370	–3.42			
0.2992	0.0682	0.0293	–0.87	0.3309	0.1400	–9.09
	0.1009	0.0432	–1.92	0.3671	0.1546	–10.31
	0.1228	0.0526	–2.79	0.4076	0.1713	–11.72
	0.1736	0.0740	–4.14	0.4505	0.1885	–13.27
	0.2124	0.0902	–5.40	0.4953	0.2073	–14.95
	0.2490	0.1059	–6.53	0.5285	0.2195	–16.42
	0.2874	0.1220	–7.73			
0.4996	0.0902	0.0379	–3.38	0.2756	0.1141	–11.96
	0.1204	0.0503	–4.86	0.3058	0.1284	–13.10
	0.1517	0.0635	–6.32	0.3410	0.1413	–14.88
	0.1855	0.0790	–7.60	0.3693	0.1519	–16.42
	0.2164	0.0905	–9.15	0.4018	0.1650	–18.07
	0.2452	0.1031	–10.37	0.4465	0.1832	–20.16
0.6971	0.1026	0.0432	–6.14	0.3527	0.1461	–21.45
	0.1382	0.0582	–8.14	0.3960	0.1632	–23.96
	0.1695	0.0707	–10.28	0.4392	0.1805	–26.97
	0.2069	0.0869	–12.34	0.4818	0.1975	–29.39
	0.2423	0.1011	–14.58	0.5326	0.2164	–32.90
	0.2792	0.1159	–17.00	0.5766	0.2348	–35.79
	0.3168	0.1314	–19.22			
0.8999	0.0943	0.0399	–7.10	0.3525	0.1460	–25.50
	0.1326	0.0560	–9.80	0.3887	0.1608	–28.33
	0.1673	0.0704	–12.57	0.4209	0.1733	–30.57
	0.2050	0.0863	–15.20	0.4635	0.1909	–33.72
	0.2390	0.1001	–17.49	0.5185	0.2127	–37.51
	0.2735	0.1143	–19.97	0.5603	0.2280	–40.89
	0.3108	0.1296	–22.40			
1.1000	0.0930	0.0392	–7.36	0.3399	0.1406	–27.16
	0.1288	0.0540	–10.47	0.3791	0.1565	–30.33
	0.1674	0.0701	–13.71	0.4236	0.1742	–34.07
	0.1966	0.0821	–15.92	0.4640	0.1904	–37.04
	0.2273	0.0949	–18.27	0.5010	0.2045	–40.16
	0.2653	0.1103	–21.28	0.5519	0.2242	–44.37
	0.2970	0.1232	–23.81			

Important results

The enthalpic pairwise interaction coefficients of *D*-mannitol and *D*-sorbitol in infinite dilution NaCl solution obtained from Eqs (5) and (6), 110.37 and 29.63 J kg mol^{-2}, are in good agreement with the values of 106.58 and 28.41 J kg mol^{-2} obtained from Eq. (4) in this work. It can be seen from Tables 3 and 4 that the values of pairwise enthalpic coefficients (h_2) for the two polyols are all positive over the whole concentration of the salt aqueous solutions investigated, and the value of h_2 for *D*-sorbitol is less than that for *D*-mannitol in aqueous solutions. From Fig. 1 it can be seen that the enthalpic pairwise interaction coefficients of the two polyols become more positive with the increase of salt concentration in the mixed solvents, and the increasing trend of the coefficients for *D*-sorbitol is more positive than that of *D*-mannitol.

Why h_2 is positive? It is generally accepted that the h_2 coefficients are attributable to the interaction between two molecules of the diluted component, which is quite sensitive to solvent variation. The interaction of ions with a non-electrolyte consists of electrostatic and structural interactions [12]. Lilley *et al.* [25] considered that the interactions between electrolyte and non-electrolyte consist of three effects: (a) partial

desolvation of solutes that interact with each other; (b) solvent reorganization in the neighbourhood of the hydrophobic groups of the non-electrolyte; (c) electrostatic interaction. The first two effects belong to the class of structural interactions, which has been used to explain the concentration dependence of transfer functions of electrolytes [26, 27]. Desonyers *et al.* [28] gave a general discussion for structural interaction, and thought that in most cases structural interaction should make positive contributions to the enthalpy interaction parameters (h_2). The last effect, electrostatic interaction, is to be discussed in the following section.

In the present system, it can be assumed that the interaction of sodium chloride with the polar groups in polyols (–OH) is mainly electrostatic [12], accompanied by the desolvation of the solutes, and the interaction of sodium chloride with the non-polar groups (–CH) in polyols. The electrostatic interaction of the electrolyte with the polar groups of a polyol molecule results in a negative contribution [12] to the enthalpy parameters (h_2), but would be partly counteracted by the positive contribution from the desolvation of the solutes. Next, more detailed analysis of an important change tendency and evident differences shown by Tables 3–4 and Fig. 1 is conducted.

Table 3 Enthalpic interaction coefficients of *D*-mannitol in aqueous sodium chloride solutions

m (NaCl)/mol kg^{-1}	h_2/J kg mol^{-2}	h_3/J kg^2 mol^{-3}	h_4/J kg^3 mol^{-4}	R^a	$S.D.^b$	m_i^c/mol kg^{-1}
0.0000	106.58	−35.84	38.74	0.9999	0.073	0.10–0.47
0.0998	117.74	−67.17	71.76	0.9999	0.055	0.10–0.50
0.3000	134.23	−71.91	75.84	0.9999	0.070	0.05–0.46
0.5003	151.77	−50.72	35.62	0.9999	0.089	0.11–0.50
0.6992	166.11	−51.33	36.43	0.9999	0.124	0.09–0.50
0.8262	176.28	−108.6	111.3	0.9998	0.199	0.07–0.51
1.0000	189.11	−117.2	121.9	0.9999	0.151	0.07–0.51
1.2034	206.57	−70.94	56.84	0.9998	0.179	0.08–0.51

^aR – correlation coefficients for Eq. (4), $^bS.D.$ – standard deviation, ^{c}concentration range

Table 4 Enthalpic interaction coefficients of *D*-sorbitol in aqueous sodium chloride solutions

m (NaCl)/mol kg^{-1}	h_2/J kg mol^{-2}	h_3/J kg^2 mol^{-3}	h_4/J kg^3 mol^{-4}	R^a	$S.D.^b$	m_i^c/mol kg^{-1}
0.0000	28.41	−43.51	42.04	0.9999	0.0053	0.13–0.43
0.1000	40.84	−29.32	34.27	0.9998	0.0411	0.12–0.56
0.2992	62.29	−30.49	41.09	0.9999	0.0589	0.07–0.53
0.4996	84.4	−24.82	35.65	0.9999	0.0675	0.10–0.45
0.6971	111.7	−20.56	20.64	0.9999	0.1098	0.10–0.58
0.8999	132.3	−26.46	22.84	0.9998	0.1364	0.10–0.56
1.1000	150.7	−35.78	28.86	0.9999	0.1062	0.10–0.55

^aR – correlation coefficients for Eq. (4), $^bS.D.$ – standard deviation, ^{c}concentration range

Pairwise enthalpic interaction coefficient of polyol and concentration of sodium chloride

The values of h_2 for both *D*-mannitol and *D*-sorbitol increase positively with elevation of concentration of sodium chloride solutions, which can be attributed to the influence of salt on the interaction of polyol. As is known, in aqueous solutions, hydration sheaths with defined water molecule are formed around dissolved molecules or ions [29]. Mutual interaction between the molecules or ions of the electrolyte may cause some water molecules to be squeezed out from the hydration sheaths into the typical bulk water [30]. So, the direct interaction between two solute molecules provokes partial dehydration of ions near them, which is an endothermic process. Another endothermic effect is resulted from the removal of a number of water molecules from the *D*-mannitol or *D*-sorbitol hydration shells. Therefore the partial dehydration of solutes gives positive contribution to h_2. The larger the concentration of sodium chloride is, the stronger the associating force of ion or polyol molecule with water molecules is and the more difficultly the water molecules depart from the solute ion or polyol molecule, and the more positive the contribution from desolvation to h_2 will be. Consequently, pairwise enthalpic interaction coefficients (h_2) of *D*-mannitol and *D*-sorbitol become more positive with the elevated concentration of sodium chloride.

The discrepancies of pairwise enthalpic interaction coefficients of the two isomers molecule in aqueous sodium chloride solution

The value of h_2 for *D*-sorbitol is less than that for *D*-mannitol in aqueous solutions. This kind of difference can be attributed to their different conformation [16, 17]. There may be more opportunities for *D*-mannitol molecules to form intermolecular hydrogen bond, because all the four secondary hydroxyl groups (–OH) stretch to different directions while 2- and 3-OH in *D*-sorbitol molecule can form intra-molecule hydrogen bond. So interaction between a pair of *D*-mannitol molecules is stronger than that between a pair of *D*-sorbitol molecules [15].

On the other hand, it can be seen from Fig. 1, that the increasing rate of the h_2 coefficient ($\mathrm{d}h_2/\mathrm{d}m$ (NaCl)) for *D*-sorbitol is larger than that of D-mannitol. Similarly, considering the difference of h_2 is mainly ascribed to the difference in the conformation of the two polyols. When two molecules of *D*-sorbitol approach each other, the intramolecular hydrogen bonds might be partly broken. Larger value of $\mathrm{d}h_2/\mathrm{d}m$ (NaCl) for *D*-sorbitol means that it is more difficult to break the intra-molecular hydrogen bond in higher concentration of the salts than that in lower concentration of the salt.

Conclusions

Heats of dilution of *D*-sorbitol and *D*-mannitol with water and different concentrations of aqueous sodium chloride solutions were measured by isothermal flow-mixing microcalorimetry at 298.15 K. Experiential enthalpies of dilution varying with the concentration of the polyols were correlated with virial expansion equation. Enthalpic interaction coefficients, h_2, h_3, h_4, in the equations were gotten and the values of pair-wise enthalpic interaction coefficient h_2, have been discussed. The subtle difference in molecular conformations of *D*-sorbitol and *D*-mannitol makes contribution to their different values of h_2. The salt concentration exert markedly different influences on the values of the homogeneous enthalpic pairwise interaction coefficients of *D*-sorbitol and *D*-mannitol, and the change trend of h_2 with the concentration of sodium chloride is discussed according to interactions of the polyol with water and sodium chloride.

Acknowledgements

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