Enthalpic Interaction of D-Sorbitol and D-Mannitol in Water and Aqueous Sodium Halide Solutions at 313.15 K

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The enthalpies of dilution of D-sorbitol and D-mannitol with water and aqueous sodium halide solutions were determined by isothermal microcalorimetry at 313.15 K. Experimental enthalpies of dilution varying with the concentration of the polylols were correlated with the virial expansion equation that was obtained with the McMillan–Mayer theory. Enthalpic interaction coefficients h_2 , h_3 , and h_4 in the equations have been determined, and the values of the pairwise enthalpic interaction coefficient (h_2) are discussed in terms of solute–solute and solute–solvent interactions.

Introduction

D-Sorbitol, a polylol derived from glucose by the enzyme aldose reductase, is a common organic solute in many cells of human body.^{1,2} It can be used as a test substance for measuring liver plasma flow in humans³ and can be recommended as a cost-effective alternative to lactulose for the treatment of constipation in the elderly.⁴ As an important isomer, D-mannitol is also widely used in many fields of medicine because of its beneficial action on the brain,⁵⁻⁹ kidneys,¹⁰ and heart,¹¹ and hence research on its physiological functions in the human body is continuously done.^{12,13} As we all know, the main solvent in the body fluid of humans and animals is water in which various ions coexist. Interactions of sorbitol/mannitol-sorbitol/mannitol. sorbitol/mannitol-water, and sorbitol/mannitol-ions must be in existence and bound up with the thermodynamic character of the systems. Therefore, studying the thermodynamic character of the two polylols in aqueous solutions is as important as researching that of such bioactive substances as amino acids with ions¹⁴⁻¹⁶ in aqueous solutions. In the present work, the dilution enthalpies have been measured, respectively, for D-mannitol and D-sorbitol in pure water and four aqueous sodium halide solutions at 313.15 K, and the homogeneous enthalpic interaction coefficients of the two polylols have been obtained. The results are interpreted in terms of solute-solute and solute-solvent interactions according to the McMillan-Mayer theory.

Experimental Section

Reagents. Both D-mannitol and D-sorbitol were purchased from Aldrich, and the stated purity was better than 99%. They were dried under reduced pressure at 323 K before use. The purity was detected with an examination



Figure 1. Relative apparent molar enthalpy of glycine with water at 298.15 K: +, this work; \times , from ref 19.

of the optical rotation ($[\alpha]^{20}_{D}$). The experimental data determined in this laboratory for D-mannitol and D-sorbitol were $[\alpha]^{20}_{D} = -26.11 \pm 0.20 \ (-26.20 \ in the literature^{17})$ and $[\alpha]_{D}^{20}$ = -1.99 ± 0.02 (-1.98 in the literature¹⁷), respectively. Sodium fluoride (NaF), sodium chloride (NaCl), sodium bromide (NaBr), and sodium iodide (NaI) (analytical reagents, purity >99%, Shanghai Chemical Reagent Company) were recrystalized from distilled water and dried under reduced pressure at 393 K. All of the reagents were stored over P2O5 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 of aqueous sodium halide is 0.500 mol·kg⁻¹. The molality ranges of D-mannitol and D-sorbitol are (0.10 to 0.50) mol·kg⁻¹ and (0.10 to 0.47) mol·kg⁻¹, respectively, with either pure water or a 0.500 mol·kg⁻¹ sodium halide

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Table 1. Enthalpies of Dilution of D-Sorbitol in Water and Sodium Halide Solutions at 313.15 K

$m_{ m i}$	$m_{ m f}$	$\Delta_{ m dil,exptl} H_{ m m}$	$\Delta_{ m dil,calcd} H_{ m m}$	$m_{ m i}$	$m_{ m f}$	$\Delta_{ m dil,exptl} H_{ m m}$	$\Delta_{ m dil,calcd} H_{ m m}$			
$\overline{\mathrm{mol}\mathbf{\cdot}\mathrm{kg}^{-1}}$	$mol \cdot kg^{-1}$	$J \cdot mol^{-1}$	$J \cdot mol^{-1}$	$mol \cdot kg^{-1}$	$mol \cdot kg^{-1}$	$J \cdot mol^{-1}$	$J \cdot mol^{-1}$			
Water										
0.1056	0.0560	-2.41	-2.42	0.3061	0.1619	-10.17	-10.10			
0.1366	0.0730	-3.86	-3.67	0.3429	0.1809	-11.59	-11.53			
0.1708	0.0909	-4.71	-4.98	0.3814	0.2008	-12.86	-13.09			
0.2033	0.1082	-6.23	-6.23	0.4216	0.2217	-14.93	-14.82			
0.2369	0.1258	-7.55	-7.48	0.4690	0.2459	-16.97	-16.96			
0.2720	0.1441	-8.83	-8.80	0.4854	0.2541	-17.70	-17.74			
$NaF (0.5 mol \cdot kg^{-1})$										
0.1064	0.0588	-10.65	-10.71	0.2999	0.1651	-20.64	-20.61			
0.1383	0.0765	-12.39	-12.11	0.3330	0.1829	-22.69	-22.45			
0.1711	0.0946	-13.66	-13.67	0.3654	0.1999	-24.32	-24.43			
0.2037	0.1127	-14.72	-15.01	0.3998	0.2189	-26.33	-26.49			
0.2332	0.1283	-16.96	-16.94	0.4338	0.2369	-28.64	-28.615			
0.2661	0.1466	-18.72	-18.61	0.4700	0.2563	-30.67	-30.86			
	$ m NaCl~(0.5~mol\cdot kg^{-1})$									
0.1412	0.0784	-9.63	-9.57	0.3458	0.1905	-19.67	-19.48			
0.1740	0.0964	-10.86	-10.88	0.3761	0.2067	-21.31	-21.17			
0.2063	0.1138	-12.44	-12.50	0.4095	0.2247	-23.04	-23.11			
0.2406	0.1327	-14.15	-14.13	0.4439	0.2431	-25.08	-25.22			
0.2780	0.1534	-15.95	-15.94	0.4744	0.2594	-26.99	-27.16			
0.3135	0.1735	-18.39	-17.68	0.4984	0.2721	-28.95	-28.77			
			NaBr (0.5	$mol \cdot kg^{-1}$)						
0.1096	0.0597	-2.42	-2.44	0.2854	0.1538	-13.43	-13.46			
0.1430	0.0777	-4.35	-4.43	0.3197	0.1723	-15.59	-15.74			
0.1676	0.09114	-5.97	-5.94	0.3511	0.1887	-17.69	-17.77			
0.1984	0.1077	-7.87	-7.83	0.3867	0.2071	-20.01	-20.15			
0.2248	0.12178	-9.60	-9.56	0.4216	0.2263	-22.47	-22.48			
0.2545	0.1375	-11.40	-11.43	0.4600	0.2460	-24.74	-24.81			
$NaI (0.5 mol \cdot kg^{-1})$										
0.1398	0.0724	-9.29	-9.35	0.3289	0.1690	-22.97	-23.08			
0.1730	0.0895	-11.68	-11.738	0.3605	0.1847	-25.48	-25.53			
0.2031	0.1049	-13.80	-13.85	0.3941	0.2015	-27.99	-28.07			
0.2315	0.1192	-16.23	-16.18	0.4301	0.2191	-30.95	-30.87			
0.2665	0.1368	-18.47	-18.53	0.4545	0.2313	-32.75	-32.69			
0.2974	0.1528	-20.66	-20.73	0.4812	0.2305	-38.51	-38.52			

aqueous solution as the solvent. All of the solutions were degassed with ultrasonic waves and used within 12 h after preparation, and the uncertainty of the concentration was $\pm 0.0002 \text{ mol}\cdot\text{kg}^{-1}$.

Calorimetric Procedure. The enthalpies of dilution were measured with a 2277-204 measuring cylinder supported by a Thermometric 2277 thermal activity monitor¹⁸ (Thermometric, Sweden) at 313.15 K. The performance of the apparatus was checked by measuring the relative apparent molar enthalpy of glycine in water, which is in good agreement with that reported in the literature¹⁹ (Figure 1) (i.e., the accuracy of the calibration was better than 1%). The solutions of sorbitol and mannitol and their solvents were pumped through the mixing-flow vessel of the calorimeter using LKB VS2-10R MIDI dual channel pumps. The variation in flow rates was less than 0.1%. The flow rates were determined by the mass of the samples delivered in 8 min. The variation in rates was less than 0.1% both before and after a complete dilution experiment. Thermal effects of the dilution process have been obtained with the following method.

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

(1) A (aqueous sodium halide solution) + B (aqueous sodium halide solution) - baseline determined;

(2) A (aqueous sodium halide solution) + B (aqueous sorbitol/mannitol solution) – dilution thermal power determined;

(3) A (aqueous sodium halide solution) + B (aqueous sodium halide solution) – baseline reestablished.

The enthalpies of dilution $\Delta_{\rm dil} H_{\rm m}$ were calculated from the equation 20

$$\Delta_{\rm dil}H_{\rm m} = -\frac{P(1+m_iM)}{m_if_2} \tag{1}$$

where *P* is the dilution thermal power (μ W); *M* is the molar mass of one of the polylols; m_i and m_f are, respectively, the initial and final molalities of D-mannitol and D-sorbitol in solvent, water, or aqueous sodium halide solutions; and f_2 is the flow rate of the D-sorbitol or D-mannitol solution. The final molalities, m_f were calculated from the equation

$$m_{\rm f} = \frac{m_{\rm i} f_2}{f_1 (1 + m_{\rm i} M) + f_2} \tag{2}$$

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

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^{21,22}

$$H_m^{\rm E} = \frac{H^{\rm E}}{m} = L_{2\varphi} = h_2 m + h_3 m^2 + h_4 m^3 + \dots \quad (3)$$

 Table 2. Enthalpies of Dilution of D-Mannitol in Water and Sodium Halide Solutions at 313.15K

mi	$m_{ m f}$	$\Delta_{ m dil,exptl} H_{ m m}$	$\Delta_{ m dil,calcd} H_{ m m}$	$m_{ m i}$	$m_{ m f}$	$\Delta_{ m dil,exptl} H_{ m m}$	$\Delta_{ m dil,calcd} H_{ m m}$			
$\overline{\mathrm{mol}\mathbf{\cdot}\mathrm{kg}^{-1}}$	$\overline{\mathrm{mol}\mathbf{\cdot}\mathrm{kg}^{-1}}$	$J \cdot mol^{-1}$	$J \cdot mol^{-1}$	$\overline{\mathrm{mol}\mathbf{\cdot}\mathrm{kg}^{-1}}$	$\overline{\mathrm{mol}\mathbf{\cdot}\mathrm{kg}^{-1}}$	$J \cdot mol^{-1}$	$J \cdot mol^{-1}$			
			Wa	ıter						
0.1024	0.0529	-5.39	-5.40	0.2799	0.1445	-15.55	-15.59			
0.1315	0.0679	-6.03	-6.16	0.3158	0.1599	-18.44	-18.37			
0.1606	0.0819	-9.25	-9.17	0.3465	0.1781	-19.70	-19.60			
0.1920	0.0992	-10.57	-10.64	0.3816	0.1955	-21.95	-21.86			
0.2118	0.1096	-11.59	-11.65	0.4214	0.2156	-24.36	-24.45			
0.2495	0.1291	-13.53	-13.61	0.4460	0.2284	-25.79	-25.23			
$NaF(0.5 mol \cdot kg^{-1})$										
0.1020	0.0532	-11.09	-10.16	0.2797	0.1452	-24.66	-23.74			
0.1322	0.0691	-12.02	-12.13	0.3102	0.1609	-26.20	-26.14			
0.1626	0.0849	-14.40	-14.52	0.3437	0.1779	-28.80	-28.78			
0.1928	0.1006	-16.41	-16.36	0.3723	0.1919	-30.90	-31.07			
0.2224	0.1160	-19.40	-19.21	0.4161	0.2145	-33.90	-34.02			
0.2533	0.1312	-21.81	-21.72	0.4520	0.2328	-36.45	-36.60			
NaCl $(0.5 \text{ mol}\cdot \text{kg}^{-1})$										
0.1139	0.0614	-9.02	-8.95	0.3030	0.1622	-20.85	-20.64			
0.1446	0.0778	-10.52	-10.67	0.3419	0.1828	-23.24	-23.21			
0.1723	0.09294	-12.78	-12.67	0.3746	0.2003	-25.44	-25.39			
0.2061	0.1112	-14.15	-14.20	0.4080	0.2177	-27.63	-27.72			
0.2368	0.1275	-16.26	-16.34	0.4432	0.2361	-30.21	-30.20			
0.2729	0.1465	-18.59	-18.64	0.4779	0.2538	-32.64	-32.77			
$ m NaBr~(0.5~mol\cdot kg^{-1})$										
0.1017	0.0547	-5.17	-5.11	0.2788	0.1492	-18.29	-18.14			
0.1325	0.0714	-7.43	-7.35	0.3114	0.1663	-20.53	-20.54			
0.1612	0.0866	-9.38	-9.47	0.3431	0.1830	-22.93	-22.89			
0.1891	0.1018	-11.44	-11.48	0.3783	0.2015	-25.46	-25.48			
0.2196	0.1178	-13.75	-13.77	0.4270	0.2267	-29.10	-29.10			
0.2461	0.1318	-15.83	-15.74	0.4512	0.2390	-30.84	-30.93			
NaI $(0.5 \text{ mol}\cdot \text{kg}^{-1})$										
0.1129	0.0613	-7.83	-7.84	0.2947	0.1590	-21.04	-21.03			
0.1434	0.0777	-10.13	-10.10	0.3309	0.1779	-23.70	-23.72			
0.1719	0.0931	-12.14	-12.16	0.3632	0.1954	-26.03	-26.05			
0.2000	0.1085	-14.19	-14.17	0.4041	0.2163	-29.30	-29.22			
0.2344	0.1268	-16.64	-16.66	0.4423	0.2364	-32.11	-32.15			
0.2643	0.1427	-18.84	-18.84	0.4677	0.2499	-34.08	-34.12			

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 initial molality m_i and final molality m_f is

$$\Delta_{\rm dil} H_{\rm m} = H^{\rm expti}_{\rm m}(m_{\rm f}) - H^{\rm expti}_{\rm m}(m_{\rm i})$$

= $h_2(m_{\rm f} - m_{\rm i}) + h_3(mf^2 - m_{\rm i}^2) + h_4(mf^3 - m_{\rm i}^3) + \dots (4)$

where $H^{\text{exptl}}_{\text{m}}$ (m_{f}) and $H^{\text{exptl}}_{\text{m}}$ (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 aqueous sodium halide solutions are regarded wholly as "solvents". Tables 1 and 2 give the experimental values of dilution enthalpies $(\Delta_{dil,exptl}H_m)$, to which eq 4 was fit with a least-squares procedure, together with the corresponding calculated values $(\Delta_{dil,calcd}H_m)$ along with the initial and final molalities $(m_i \text{ and } m_f)$ of D-sorbitol or D-mannitol in different solvents.

Table 3 lists the coefficients of eq 4 that were obtained from a least-squares analysis of these results. Because it is difficult to interpret the higher-order *h* coefficients, only the enthalpic pairwise coefficient h_2 is discussed here. The values of h_2 are the measurement of the interaction energy between a pair of solute molecules,²² which may take place with a coexisting electrolyte in aqueous solution. Figure 2 shows the variations of h_2 coefficients for both D-mannitol and D-sorbiol within water and four aqueous sodium halide solutions.

From Figure 2, we can see that in water or in aqueous sodium halide solutions the values of the pairwise enthalpic coefficient, h_2 , for both D-mannitol and D-sorbitol are positive. These results indicate that energy is needed to destroy the aqueous sheath of each polylol molecule when two molecules of D-mannitol or D-sorbitol approach each other. The value of h_2 for D-mannitol is less than that for D-sorbitol in aqueous solutions. This kind of difference can



Figure 2. Variation of homogeneous enthalpic pairwise interaction coefficients of \Box , D-sorbitol and \bigcirc , D-mannitol in water and aqueous sodium halide solutions (0.500 mol kg⁻¹) at 313.15 K.

Table 3	. Enthalpies	and Interaction	Coefficients	of D-Sorbitol	and D -Mannito	ol in Pure	Water and	l Aqueous S	sodium
Halide 8	Solutions at	313.15 K							

		h_2	h_3	h_4		
solvent	solute	$\overline{{ m J}{\cdot}{ m kg}{\cdot}{ m mol}^{-2}}$	J·kg ² ·mol ⁻³	$\overline{J\cdot kg^3\cdot mol^{-4}}$	R^2	SD
H ₂ O	D-sorbitol	104.6 a(+2.3)	-68.4 a(+5.8)	76.7 a(+5.2)	^b 0.9994	^b 0.36
	D-mannitol	153.2 $a(\pm 5.2)$	-106.5 $a(\pm 8.9)$	130.3 $a(\pm 11.5)$	^b 0.9989	^b 0.28
NaF/0.5 mol·kg $^{-1}$	D-sorbitol	67.5 $a(\pm 3.2)$	93.2 $a(\pm 6.6)$	-55.9 $a(\pm 5.3)$	^b 0.9991	^b 0.26
	D-mannitol	107.7 $a(\pm 2.4)$	127.8 $a(\pm 9.5)$	-128.7 $a(\pm 2.3)$	$^{b}0.9974$	^b 0.54
NaCl/0.5 mol·kg^{-1}	D-sorbitol	93.9 $a(\pm 2.3)$	$1.6 a(\pm 0.6)$	$35.8 \\ a(\pm 2.9)$	^b 0.9996	^b 0.14
	D-mannitol	116.6 $a(\pm 3.3)$	$24.5 \\ a(\pm 2.3)$	$0.09 \\ a(\pm 0.03)$	^b 0.9995	$^{b}0.21$
NaBr/0.5 mol·kg ⁻¹	D-sorbitol	108.2 $a(\pm 4.6)$	67.2 $a(\pm 3.5)$	-60.5 $a(\pm 2.5)$	^b 0.9992	^b 0.28
	D-mannitol	160.4 a(+2.3)	-5.1 $a(\pm 0.5)$	0.46 a(+0.23)	^b 0.9999	^b 0.09
NaI/0.5 mol·kg ⁻¹	D-sorbitol	115.0 a(+5.3)	-290.8	292.3 a(+2.8)	^b 0.9995	^b 0.23
	D-mannitol	173.4 $a(\pm 3.2)$	-45.2 $a(\pm 2.3)$	48.2 $a(\pm 1.6)$	^b 0.9999	^b 0.04

^a From the regression analyses procedures used to fit eq 4 with the enthalpy of dilution data. ^b Parameters for eq 4.

be attributed to their different conformation.¹⁷ There may be more opportunities for mannitol molecules to form intermolecular hydrogen bonds because all four secondary hydroxyl groups (–OH) stretch in different directions and 2- and 3-OH in sorbitol molecule can form intramolecule hydrogen bonds. Therefore, the interaction between a pair of D-mannitol molecules is stronger than that between a pair of D-sorbitol molecules.

It can also be seen in Figure 2 that the value of h_2 for both D-mannitol and D-sorbitol increases positively with the radius of the anion in aqueous sodium halide solutions. As is known, in aqueous solutions, hydration sheaths with defined water molecules are formed around dissolved molecules or ions.²³ Mutual interaction between the molecules or ions of the salts may cause some water molecules to be squeezed out from the hydration sheaths into the typical bulk water.²⁴ The fluoride ion has the smallest radius and the largest surface charging density in the halide ions; therefore, it will exert the largest attractive force on the water molecules. This makes it easier to break the hydration layer around polylol molecules when two manitol/sorbitol molecules approach each other, so the value of h_2 for manitol/sorbitol in NaF solution is even less than that in water. The ability to destroy the hydration layer of the polylol molecule varies as $F^- > Cl^- > Br^- >$ I⁻; therefore, the values of h_2 increase in the same order (Figure 2) for the two polylols. However, the dispersion force of ions upon two solute molecules can lead to the partial desolvation of solutes and the reorganization of the hydration structure around sorbitol and mannitol. Because the dispersion force is not strong enough to destroy the primary hydration structure of salt ions, the partial desolvation effect can destroy only the secondary hydration layer structure^{25,26} and make some negative contribution to enthalpic interaction parameters. The larger the size of the anion, the stronger the dispersion force becomes, so the more the hydration layer around the polylol molecule will be disrupted, and the more the negative contribution to h_2 will be. Therefore, the iodide ion has the strongest dispersion force in halide ions, which results in the most negative contribution to h_2 for sorbitol and mannitol through the dispersion force. Nevertheless, the dispersion force is much

weaker than forces such as hydrogen bonding and iondipole interactions, so the trend of h_2 in Figure 2 is not decided by it.

Conclusions

Enthalpies of dilution of D-sorbitol and D-mannitol with water and aqueous sodium halide solutions were measured by isothermal flow-mixing microcalorimetry at 313.15 K. Experiential enthalpies of dilution varying with the concentration of the polylols were correlated with the virial expansion equation. Enthalpic interaction coefficients h_2 , h_3 , and h_4 in the equations are obtained, and the values of the pairwise enthalpic interaction coefficient h_2 have been discussed. The different structures of D-sorbitol and Dmannitol make a contribution to their different values of h_2 . Anions F⁻, Cl⁻, Br⁻, and I⁻ exert markedly different influences on the values of the homogeneous enthalpic pairwise interaction coefficients of sorbitol and mannitol, and the change in the trend of h_2 with the radius of the anion is discussed according to interactions of the polylol with water and the salts.

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