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Thermodynamics Study of the Interaction of CsCl with D-Glucose in Water from $T = 278.15$ to 313.15 K

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Electromotive forces of cells without liquid junctions K-ISE | CsCl(m_E) | ISE-Cl and K-ISE | CsCl(m_E), glucose(m_S) | ISE-Cl have been measured at 5 K intervals from $T = 278.15$ to 313.15 K, where $m_E = (0.001$ to $0.1)$ mol kg^{-1} and $m_S = (0.01$ to $3.0)$ mol kg^{-1}. The activity coefficients of CsCl in glucose + water solutions can be obtained from these electromotive force data; in the meantime, the interaction parameters of the CsCl + glucose pair in water, g_{EN}, h_{EN}, s_{EN}, and $c_{p,EN}$, can be evaluated. We show that $g_{EN} > 0$, $h_{EN} > 0$, $s_{EN} > 0$, and $c_{p,EN} < 0$ at all four temperatures (except for h_{EN} and s_{EN} at $T = 313.15$ K), and the values of g_{EN}, s_{EN}, and $c_{p,EN}$ vary slightly with temperature, whereas the values of h_{EN} is very sensitive to the temperatures used. These thermodynamic parameters were discussed in terms of a model of the structural interaction and electrostatic interaction and were analyzed by the group additivity principle.

1. Introduction

During the past decade, aqueous solutions of hydrophilic nonelectrolytes have received considerable attention. They are generally characterized by weak nonbonding intermolecular interactions, which are of interest for biological system because of their low specificity.[1] Glucose has been regarded as one of the typical hydrophilic nonelectrolytes resembling urea. The thermodynamic properties of aqueous glucose have been extensively studied.[2−6] Furthermore, some studies have been reported on the thermodynamic properties of water + glucose + electrolyte systems.[7,8] However, no work has been carried out for the glucose + CsCl + water system. In our previous papers, we have studied the thermodynamic properties of some monosaccharide + electrolyte + water ternary systems and the interactions of cesium chloride with D-glucose, D-galactose, D-xylose, and D-arabinose in water at $T = 298.15$ K by several means using an ion selective electrode (ISE),[9] a calorimeter,[10] and a density meter. As part of the systematic studies of the thermodynamics of the interaction of heavy rare alkali metal ions with hydrophilic hydroxyl compounds in water, we report here the interaction parameters of the typical hydrophilic noneletrolyte D-glucose with cesium chloride in water at 5 K intervals from $T = 298.15$ to 313.15 K to examine the dependence of these parameters on temperature. It is expected that such a study will provide more information on the interaction between CsCl and D-glucose in water.

2. Experimental Section

Cesium chloride (mass fraction ≥0.999, made from cesium carbonate; the purification and determination of CsCl have been reported elsewhere[11]) and anhydrous D-glucose (Shanghai Chem. Co., mass fraction ≥0.99) were recrystallized from a water + ethanol mixture and dried under low pressure at $T = 338$ K for 6 h. Cesium chloride was heated at $T = 773$ K to constant mass and stored over P_2O_5 in desiccators. Deionized and doubly distilled water with a conductivity of 1.0×10^{-4} S·m^{-1} to 1.2×10^{-4} S·m^{-1} at $T = 298.15$ K was used throughout this work.

A nonliquid-junction cell was set up with the potassium ion-selective electrode as the working electrode and the chloride ion-selective electrode as the reference electrode to measure the electromotive forces (emfs) of a CsCl + glucose + water solution:

$$\text{K-ISE} \mid \text{CsCl}(m_E) \mid \text{ISE-Cl} \qquad \text{(I)}$$

$$\text{K-ISE} \mid \text{CsCl}(m_E), \text{glucose}(m_S) \mid \text{ISE-Cl} \qquad \text{(II)}$$

where m_E and m_S are the molalities of the electrolyte and saccharide, respectively. The emfs of the cells were measured at $T = (298.15, 303.15, 308.15,$ and $313.15)$ K. These cells were thermostated at each temperature with an accuracy of ±0.05 K. The test solutions were freshly prepared by mass using an automatic electronic balance with a sensitivity of 10 μg. To minimize the concentration gradients in the cell, all of the solutions were stirred continuously by a magnetic stirrer.

Each set of experiments was performed at a fixed electrolyte concentration ranging from 0.001 mol·kg^{-1} to 0.1 mol·kg^{-1}, and the molalities of saccharide were in-

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creased from 0.01 mol·kg^{-1} to 3.0 mol·kg^{-1} by adding solid saccharide. At each point, the emfs were recorded only when the drift was less than 0.1 mV for 5 min. To ensure the stability of the electrode during each experimental process, the electrodes were allowed to stay in solution for only 1 h to 2 h, and after the cell potential of the last sample solution was measured, the electrodes were transferred to a CsCl(aq) solution that had the same molality as that of the solution just measured. The two emfs values of the CsCl(aq) solutions agreed with each other to within 0.3 mV. Some experiments such as those at 0.001 mol·kg^{-1} (the most dilute) and 0.1 mol·kg^{-1} (the most concentrated) CsCl were replicated five times, and the data reported are the average of the replicates that agree with each other to within 0.3 mV.

3. Thermodynamic Relations and Results

The potential of an ISE can be expressed as

$$E = E' \pm \left(\frac{S_i}{Z_i}\right)\ln \alpha_i \tag{1}$$

where α_i is the activity of ion i; the constant E' is the emf of the measuring circuit of the ISE and depends on the activity of the ions in the inner solution and on the type of internal reference electrode of the ISE. The constant S_i refers to the slope of the electrode response to ion i, and Z_i is the charge number of ion i.

For cells (I) and (II), assuming that $S \approx S_+ \approx S_- \approx (S_+ + S_-)/2$ leads to

$$E(\mathrm{I}) = E(\mathrm{I})' + \left(\frac{S}{\upsilon}\right)\left(\frac{Z_+ + Z_-}{Z_+ Z_-}\right)\ln \alpha_{\mathrm{E}}\,(\mathrm{I}) \tag{2}$$

$$E(\mathrm{II}) = E(\mathrm{II})' + \left(\frac{S}{\upsilon}\right)\left(\frac{Z_+ + Z_-}{Z_+ Z_-}\right)\ln \alpha_{\mathrm{E}}\,(\mathrm{II}) \tag{3}$$

where $E(\mathrm{I})$ and $E(\mathrm{II})$ are emfs of cells (I) and (II), respectively, and $E(\mathrm{I})' = E(\mathrm{II})' = E'_+ - E'_-$. Here, α_{E} is the activity of the electrolyte defined as

$$\alpha_{\mathrm{E}} = (m_\pm \gamma_\pm)^{\upsilon} \tag{4}$$

$m_\pm$ and $\gamma_\pm$ are the mean ionic molality and activity coefficients of the electrolyte, and $\upsilon = \upsilon_+ + \upsilon_-$, where υ_+ and υ_- are the stoichiometric numbers of cations and anions.

Subtracting eq 2 from eq 3 and rearranging gives

$$\ln\left\{\frac{\gamma_\pm(\mathrm{II})}{\gamma_\pm(\mathrm{I})}\right\} = \left\{\left(E(\mathrm{II}) - \frac{E(\mathrm{I})}{S}\right)\left(\frac{Z_+ Z_-}{Z_+ + Z_-}\right)\right\} \tag{5}$$

The ratio of the mean activity coefficients of CsCl $\{\gamma_\pm(\mathrm{II})/\gamma_\pm(\mathrm{I})\}$ in the presence of saccharide to those in the absence of saccharide at the same CsCl molality can be calculated from the emf values of cells (II) and (I) according to eq 5. (The values of S can be obtained from experimental data.)

The potassium ion-selective electrode has excellent response to Cs$^+$. Because cesium chloride is a high-purity regent in which the content of impurities, such as Na$^+$ and K$^+$, is very low, there is no interference with the emf measurements of Cs$^+$. Therefore, the potassium ion-selective electrode can be used to obtain the emf values instead of a cesium ion-selective electrode in this work.

The slope S of the electrode response was determined by using activity coefficients of CsCl that were calculated by the empirical equation recommended by Hamer and Wu.[12] The typical value of S obtained from the linear regression analysis of the experimental points was (26.21 ± 0.02) mV with a correlation coefficient of 0.9994. Reasonable agreement between the experimental and theoretical values (25.61 mV at $T = 298.15$ K of the Nernst equation) was found, which suggested that the potassium ion-selective electrode behaves in accordance with the Nernst equation in the experimental range.

The ratio of the mean activity coefficients of CsCl in the presence of saccharide to those in the absence of saccharide at the same CsCl molality can be calculated from eq 5 using the value of S and the emf values. The results are given in Tables 1 to 4.

From the Tables, it can be seen that at $T = 298.15$ K the $\gamma_\pm(\mathrm{II})/\gamma_\pm(\mathrm{I})$ ratio of CsCl increased with increasing m_{S} but decreased with increasing m_{E}. However, with the rise in temperature, the dependence of $\gamma_\pm(\mathrm{II})/\gamma_\pm(\mathrm{I})$ on m_{S} became more complicated. It increased first at lower concentration but decreased sharply at higher concentration with increasing m_{S}.

According to the McMillan−Mayer theory,[13] the excess thermodynamic functions can be expressed in terms of pair and triplet interactions between the two solutes. For a solution of an electrolyte (E) and a nonelectrolyte (N) in water, it is given by

$$\begin{aligned} F(m_{\mathrm{N}}, m_{\mathrm{E}}) = {} & F^\circ(m_{\mathrm{N}}, m_{\mathrm{E}}) + F^{\mathrm{EX}}(m_{\mathrm{N}}) + F^{\mathrm{EX}}(m_{\mathrm{E}}) \\ & + 2\upsilon f_{\mathrm{EN}} m_{\mathrm{E}} m_{\mathrm{N}} + 3\upsilon^2 f_{\mathrm{EEN}} m_{\mathrm{E}}^2 m_{\mathrm{N}} + \\ & 3\upsilon f_{\mathrm{ENN}} m_{\mathrm{E}} m_{\mathrm{N}}^2 + \ldots \end{aligned} \tag{6}$$

where $F^\circ(m_{\mathrm{N}}, m_{\mathrm{E}})$ is the total thermodynamic function of the ternary solution when it is assumed that the ions are neutral particles and there are no interactions between the solutes. $F^{\mathrm{EX}}(m_{\mathrm{N}})$ and $F^{\mathrm{EX}}(m_{\mathrm{E}})$ are the excess functions of the binary N + W and E + W solutions, respectively. f_{EN}, f_{EEN}, and f_{ENN} are the pair and triplet interaction parameters that take into account all new sources of nonideality in the ternary solution.

For the transfer function of electrolyte from water to aqueous nonelectrolyte solution, we have from eq 6

$$\begin{aligned} \Delta_{\mathrm{t}}F(\mathrm{W} \rightarrow \mathrm{W + N}) & = F_{\mathrm{E}}\,(m_{\mathrm{N}}, m_{\mathrm{E}}) - F_{\mathrm{E}}^\circ\,(m_{\mathrm{E}}) \\ & = 2\upsilon f_{\mathrm{EN}} m_{\mathrm{N}} + 6\upsilon^2 f_{\mathrm{EEN}} m_{\mathrm{E}} m_{\mathrm{N}} + \\ & \quad 3\upsilon f_{\mathrm{ENN}} m_{\mathrm{N}}^2 + \ldots \end{aligned} \tag{7}$$

where both $F_{\mathrm{E}}(m_{\mathrm{N}}, m_{\mathrm{E}})$ and $F_{\mathrm{E}}^\circ(m_{\mathrm{E}})$ are the partial mole quantities of the electrolyte.

In the particular case of the Gibbs free energies, eq 7 is reduced to[14−16]

$$\begin{aligned} \Delta_{\mathrm{t}}G_{\mathrm{E}}(\mathrm{W} \rightarrow \mathrm{W + N}) & = -F\Delta E \\ & = \mu_{\mathrm{E}}(m_{\mathrm{N}}, m_{\mathrm{E}}) - \mu_{\mathrm{E}}^\circ(m_{\mathrm{E}}) \\ & = 2\upsilon g_{\mathrm{EN}} m_{\mathrm{N}} + 6\upsilon^2 g_{\mathrm{EEN}} m_{\mathrm{E}} m_{\mathrm{N}} + \\ & \quad 3\upsilon g_{\mathrm{ENN}} m_{\mathrm{N}}^2 \end{aligned} \tag{8}$$

where $\mu_{\mathrm{E}}(m_{\mathrm{N}}, m_{\mathrm{E}})$ and $\mu_{\mathrm{E}}^\circ(m_{\mathrm{E}})$ are the chemical potentials of the electrolyte in the E−N−W ternary solution and the E−W binary solution, respectively, F is the Faraday constant, and ΔE is the difference in electromotive forces. In the present case, $\Delta E = E(\text{cell II}) - E(\text{cell I})$. The higher-order terms have been neglected in eq 8. The pair and triplet interaction parameters can be obtained from a least-

Table 1. Ratios[a] of the Ionic Activity Coefficient of CsCl in the Presence ($\gamma_{\pm}^{\mathrm{II}}$) and Absence ($\gamma_{\pm}^{\mathrm{I}}$) of D-Glucose at Different m_E and m_S at T = 298.15 K

	m_E/mol·kg^{-1}					
m_S/mol·kg^{-1}	0.001	0.005	0.01	0.05	0.08	0.1
	$\gamma_{\pm}^{\mathrm{II}}/\gamma_{\pm}^{\mathrm{I}}$					
0.01	1.021	1.021	1.019	1.017	1.017	1.015
0.03	1.037	1.033	1.029	1.029	1.025	1.023
0.07	1.055	1.051	1.045	1.039	1.035	1.032
0.1	1.071	1.059	1.055	1.049	1.045	1.035
0.3	1.096	1.088	1.081	1.075	1.065	1.053
0.7	1.132	1.121	1.115	1.125	1.121	1.124
1.0	1.163	1.158	1.147	1.132	1.121	1.104
1.5	1.262	1.250	1.248	1.234	1.194	1.172
2.0	1.365	1.294	1.275	1.270	1.253	1.194
3.0	1.505	1.352	1.297	1.279	1.270	1.255

[a] The error was less than 0.002.

Table 2. Ratios[a] of the Ionic Activity Coefficient of CsCl in the Presence ($\gamma_{\pm}^{\mathrm{II}}$) and Absence ($\gamma_{\pm}^{\mathrm{I}}$) of D-Glucose at Different m_E and m_S at T = 303.15 K

	m_E/mol·kg^{-1}					
m_S/mol·kg^{-1}	0.001	0.005	0.01	0.05	0.08	0.1
	$\gamma_{\pm}^{\mathrm{II}}/\gamma_{\pm}^{\mathrm{I}}$					
0.01	1.008	1.006	1.006	1.004	1.002	1.002
0.03	1.013	1.011	1.010	1.008	1.003	1.004
0.07	1.044	1.023	1.017	1.010	1.010	1.010
0.1	1.048	1.036	1.027	1.023	1.013	1.011
0.3	1.061	1.052	1.037	1.025	1.017	1.015
0.7	1.076	1.033	1.047	1.031	1.019	1.017
1.0	1.086	1.065	1.041	1.019	1.017	1.017
1.5	1.103	1.049	1.027	1.017	1.011	1.008
2.0	1.123	1.035	1.023	1.015	1.002	0.998
3.0	1.148	1.027	1.015	1.013	0.998	0.994

[a] The error was less than 0.002.

Table 3. Ratios[a] of the Ionic Activity Coefficient of CsCl in the Presence ($\gamma_{\pm}^{\mathrm{II}}$) and Absence ($\gamma_{\pm}^{\mathrm{I}}$) of D-Glucose at Different m_E and m_S at T = 308.15 K

	m_E /mol·kg^{-1}					
m_S/mol·kg^{-1}	0.001	0.005	0.01	0.05	0.08	0.1
	$\gamma_{\pm}^{\mathrm{II}}/\gamma_{\pm}^{\mathrm{I}}$					
0.01	1.010	1.006	1.004	1.002	1.002	1.002
0.03	1.015	1.012	1.010	1.008	1.006	1.004
0.07	1.027	1.021	1.015	1.013	1.013	1.010
0.1	1.037	1.031	1.019	1.017	1.015	1.012
0.3	1.047	1.037	1.025	1.017	1.017	1.015
0.7	1.057	1.045	1.033	1.021	1.025	1.023
1.0	1.069	1.055	1.045	1.025	1.017	1.012
1.5	1.083	1.065	1.029	1.019	1.010	1.002
2.0	1.102	1.041	1.019	1.013	1.002	0.996
3.0	1.138	1.025	1.013	1.010	0.994	0.985

[a] The error was less than 0.002.

squares analysis of the experimental data. The results are given in Table 5, together with their standard deviations. Here we are interested only in the pair parameters that are most important in dilute solution.

The pair free-energy interaction parameter g_{EN} was analyzed as a function of thermodynamic temperature T using a least-squares routine, and the best representation of the data was found to be[17,18]

$$g_{EN} = a + bT + cT^2 \tag{9}$$

The coefficients a = 98893.8 J·kg·mol^{-2}, b = −640.249 J·kg·mol^{-2}·K^{-1}, and c = 1.03796 J·kg·mol^{-2}·K^{-2} were acquired with a standard deviation of the fit of 1.31.

Table 4. Ratios[a] of the Ionic Activity Coefficient of CsCl in the Presence ($\gamma_{\pm}^{\mathrm{II}}$) and Absence ($\gamma_{\pm}^{\mathrm{I}}$) of D-Glucose at Different m_E and m_S at T = 313.15 K

	m_E /mol·kg^{-1}					
m_S/mol·kg^{-1}	0.001	0.005	0.01	0.05	0.08	0.1
	$\gamma_{\pm}^{\mathrm{II}}/\gamma_{\pm}^{\mathrm{I}}$					
0.01	1.012	1.012	1.010	1.008	1.006	1.006
0.03	1.021	1.017	1.015	1.012	1.013	1.010
0.07	1.029	1.027	1.025	1.021	1.019	1.017
0.1	1.035	1.033	1.029	1.025	1.029	1.025
0.3	1.045	1.041	1.039	1.031	1.031	1.017
0.7	1.062	1.055	1.047	1.015	1.021	1.012
1.0	1.083	1.069	1.037	1.012	1.015	1.008
1.5	1.106	1.057	1.029	1.010	1.008	1.006
2.0	1.127	1.051	1.021	1.006	1.002	0.998
3.0	1.144	1.041	1.016	1.004	0.996	0.994

[a] The error was less than 0.002.

Table 5. Gibbs Free Energy Pair Interaction Parameters (g_{ES}) for the CsCl + Glucose + Water System from T = 298.15 to 313.15 K

T/K	298.15	303.15	308.15	313.15
	g_{EN}/J·kg·mol^{-2}			
	265.66 ± 11.6	186.03 ± 15.3	155.37 ± 9.8	179.53 ± 12.1

Table 6. Pair Interaction Parameters for the CsCl + Glucose in Water System from T = 298.15 to 313.15 K

T/K	s_{EN}/J·kg·mol^{-2}	h_{EN}/J·kg·mol^{-2}	$c_{p,EN}$/J·kg·mol^{-2} K^{-1}
298.15	21.3	6625	−618
303.15	10.9	3505	−629
308.15	~0	332	−639
313.15	−9.8	−2891	−650

The pair interaction parameters are related to each other in the usual way.[19] From eq 9, it follows that

$$s_{EN} = -(b + 2cT) \tag{10}$$

$$h_{EN} = a - cT^2 \tag{11}$$

$$c_{p,EN} = -2cT \tag{12}$$

Values of pair interaction entropic, enthalpic, and isobaric capacity parameters at different temperatures can be calculated from eqs 10 to 12. They are given in Table 6.

4. Discussion

It can be seen from Tables 5 and 6 that $g_{EN} > 0$ at the temperatures concerned and decreased with increasing temperature. This means that the relation between glucose and CsCl is thermodynamic repulsion and glucose is being salted out by CsCl. Both s_{EN} and h_{EN} are positive (except at T = 313.15 K), and the values decreased with rising temperature. The values of $c_{p,EN}$ are negative and decreased slowly with increasing temperature. Among these interaction parameters, h_{EN} is the most sensitive to the temperatures used.

This can be interpreted in terms of the structural interaction and electrostatic interaction models. It is generally believed that the interaction of ions with a nonelectrolyte consists of electrostatic and structural interactions. The concept of structural interactions was first introduced by Frank and Robinson.[20] For the studied solution here, the structural interaction between CsCl and glucose consists principally of the partial desolvation of solutes that interact with each other and the solvent reorganization in the neighborhood of the hydrophobic groups of the nonelectrolyte. In ref 19, it was discussed in detail and now

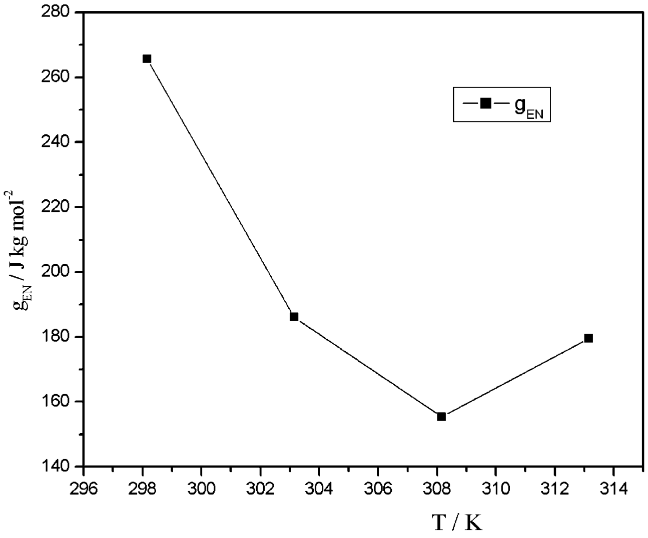

Figure 1. Temperature dependence of the Gibbs free energy interaction parameter (g_{EN}) for the CsCl−glucose pair in water.

has been widely accepted that in most cases structural interaction should make positive contributions to h_{EN}, s_{EN}, and g_{EN}.

The interaction between glucose and CsCl should include four types of interactions:

Cs$^+$−R (R represents hydrophobic alkyl groups);

Cl$^-$−R;

Cs$^+$−O; and

Cl$^-$−O (O represents hydrophilic groups such as −OH, C=O and −O−).

The interaction of the Cs$^+$−O pair, which is mainly electrostatically attractive in nature, contributes a negative value to h_{EN} and $g_{EN.}$ However, this negative contribution would be partially counteracted by the positive contribution from the Cs$^+$−R pair interaction that is mainly a structural interaction. However, the interaction of the Cl$^-$−R pair, in which structural interaction is the leading part, contributes a positive value to h_{EN} and g_{EN}. This interaction, as pointed out by Desnoyers and et al.,[14] decreases rapidly with temperature. The interaction of the Cl$^-$−O pair is mainly electrostatically repulsive and contributes a positive value to h_{EN}, s_{EN}, and g_{EN}. The electrostatic interaction of Cs$^+$ and Cl$^-$ with polar groups in the saccharide is the dominant interaction in the systems studied. The two interactions have opposite effects on h_{EN} and g_{EN}, and the experimental results suggest that the values of h_{EN} and g_{EN} are determined by the interaction of the Cl$^-$−O pair. These are in agreement with the suggestion of Kelley and Lilley[21] that the experimental free-energy pair interaction parameters are fairly well represented by the electrostatic interaction only.

In addition, it is very interesting that the value of g_{EN} decreases rapidly with temperature from $T = 298.15$ to 308.15 K but then increases slightly at 313.15 K (Figure 1). This demonstrates that the Gibbs free energy interaction parameter g_{EN} is lower in the temperature zone that approaches a human being's natural temperature than in other ones. We do not know the meaning of this phenomenon with regard to physiological action in the body, but it is important and warrants further careful consideration.

Heat capacity is a very sensitive probe for the study of structural interactions.[19] Negative values mean that the interaction of glucose with CsCl in water results in a reduction in the structure of the solution.

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