

Densities and Viscosities of the Quaternary System Mannitol-Sorbitol-D-Glucose-H₂O and Its Ternary Subsystems at 298.15K*

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Abstract To check the applicabilities of the simple density equation and viscosity equation in the semi-ideal solution theory to nonelectrolyte solutions, the densities and viscosities were measured for the quaternary system mannitol-sorbitol-D-glucose-H₂O and its ternary subsystems mannitol-D-glucose-H₂O and sorbitol-D-glucose-H₂O at 298.15K. The results were used to test the applicability of the simple equations for the density and viscosity of the multicomponent nonelectrolyte solution. The agreements between the predicted and measured results are good.

Keywords density, viscosity, mannitol, sorbitol, D-glucose, simple predictive equations

1 INTRODUCTION

Accurate and extensive thermodynamic and transport properties of multicomponent solutions play a very important role in several fields: chemistry and chemical engineering, separation process, waste water treatment, pollution control, oil recovery, and so on. Up to now, extensive measurements have been made in literature for the thermodynamic properties and transport properties of binary aqueous solutions, but relatively few measurements have been made for the multicomponent solutions. Therefore, it is interesting and practically important to develop the simple equations that can completely use the available data for the binary solutions and provide sufficiently accurate predictions for the multicomponent solutions[1—8].

Recently, Hu[1] has presented the extended semi-ideal solution theory from which a new set of simple predictive equations have been established for thermodynamic properties of the multicomponent solutions. It has also been shown that the mixing behavior of viscosities of aqueous solutions obeying the linear isopiestic relation[8—15] is also simple and, therefore, allows for the establishment of the novel predictive equations for the surface tension and viscosity of the multicomponent solutions[5,16] by coupling the simple relation and the thermodynamic relations and the Eyring's absolute rate theory[17]. The proposed equations are expected to hold for different kinds of mixtures such as aqueous and nonaqueous solutions of electrolytes, nonelectrolytes, (electrolyte-nonelectrolyte) mixtures, aqueous solutions of polyelectrolytes and water-soluble neutral polymers, and alloys[1]. These equations have been tested by comparison with the experimental data for strong electrolyte solutions with and without common ions and for the electrolyte solutions involving transition metal chlorides, and the agreements between the measured and predicted properties are in general quite good[5,8,12—16]. However, their applicabilities to the densities and viscosities of multicomponent nonelectrolyte solutions have not yet been tested. Therefore, in this study, the

densities and viscosities were measured for the quaternary system mannitol-sorbitol-D-glucose-H₂O and its ternary subsystems mannitol-D-glucose-H₂O and sorbitol-D-glucose-H₂O at 298.15K, and the results were used to check the applicabilities of the simple equations. The measured data also play an important role in the fields of food engineering[18,19] and in the processing of fragile biological materials[20].

2 EXPERIMENTAL

All the chemicals used in this study were reagent grade and were recrystallized twice using double distilled water. Mannitol, sorbitol, and D-glucose were dehydrated at room temperature under vacuum over CaCl₂ to constant mass[14,21]. Finally, all the chemicals were stored over P₂O₅ in a vacuum desiccator before use.

Solutions were prepared by direct weighing of the double-distilled deionized water and each of the solutes with a precision of $\pm 5 \times 10^{-5}$ g. All weighing was corrected to vacuum. For single solutions in the dilute region, solutions were made by diluting a stock solution by mass. In the concentrated region, solutions were prepared from a known mass of solute and water, respectively. The ternary and quaternary samples were prepared by mixing the binary solutions. All the solutions were prepared afresh before use. Concentrations were expressed in molality ($\text{mol} \cdot \text{kg}^{-1}$). The densities of solutions were measured with a KEM oscillating-tube digital densimeter (DA-505) thermostated to better than ± 0.01 K. The temperature in the measuring cell was monitored with a digital thermometer. The densimeter was calibrated by double-distilled water and dry air. The densities of water at different temperatures were obtained from the literature[22]. The densities of water and air at 298.15K were taken as 0.99701 and $1.18434 \text{kg} \cdot \text{cm}^{-3}$, respectively. The precision of density values was $\pm 5 \times 10^{-5} \text{g} \cdot \text{cm}^{-3}$.

The viscosities were measured using a modified Cannon-Ubbelohde suspended level capillary viscometer.

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A thoroughly cleaned and perfectly dried viscometer filled with experimental liquid was placed vertically in a glass sided water thermostat. The experimental temperature was maintained to $(298.15 \pm 0.01)\text{K}$. After attaining thermal equilibrium, the efflux times of flow of liquids were recorded using a digital stop watch with a precision of $\pm 0.01\text{s}$. Triplicate measurements were carried out at each composition. The estimated error of viscosity measurements is within $\pm 10^{-4}\text{mPa}$. The viscosity of solution η is given by Eq.(1).

$$\eta = \eta_0 \frac{d\tau}{d_0\tau_0}, \quad (1)$$

where η_0 is the viscosity of the standard oil, d and d_0 are the densities of the solution and the standard oil, respectively. τ and τ_0 are the flow times of the solution and the standard oil, respectively.

3 RESULTS AND DISCUSSION

3.1 Predictive equations for the density and viscosity of the multicomponent nonelectrolyte solution

In the following section, the variables with the superscript o together with the subscript i were used to denote the quantities of i in the binary solution ($i\text{-H}_2\text{O}$) having the same water activity as that of a multicomponent solution, and those without the superscript o denote the corresponding quantities in the multicomponent solution.

Recently, the semi-ideal solution theory[1] has been presented to describe the thermodynamic behavior of the multicomponent aqueous solutions 1-2-...- $n\text{-H}_2\text{O}$ at constant activity of H_2O . The theory proves that, under the condition of constant activity of H_2O , the average hydration numbers characterizing the ion-solvent interactions have the same values in the mixture as in the subsystems and that the contributions from the ion-ion interactions to the solvent activity are also the same in the mixture as in the subsystems. Therefore, the process of mixing the nonideal electrolyte mixtures $i\text{-H}_2\text{O}$ ($i=1, 2, \dots, n$) at constant activity of H_2O is as simple as that of mixing the ideal mixtures $i\text{-H}_2\text{O}$ ($i=1, 2, \dots, n$) of equal mole fraction of H_2O , so that the changes in thermodynamic properties accompanying the process of mixing the nonideal solutions obey the same linear relations as mixing the classical ideal solutions, including the linear concentration relations, the linear equations for changes in Gibbs free energy, enthalpy, entropy, thermal properties, and volumetric properties. From these linear relations, the linear isopiestic relation[8—16]

$$\sum_i \frac{m_i}{m_i^o} = 1 \quad (2)$$

can be derived theoretically, where m_i and m_i^o are the molalities of the i -th solute in multicomponent aqueous solution 1-2-...- $n\text{-H}_2\text{O}$ and its binary subsystems $i\text{-H}_2\text{O}$ ($i=1, 2, \dots, n$) of equal water activity. If $(\Delta_{\text{mix}}G_i^o, \Delta_{\text{mix}}H_i^o, \Delta_{\text{mix}}S_i^o, \Delta_{\text{mix}}V_i^o)$ and $(\Delta_{\text{mix}}G, \Delta_{\text{mix}}H, \Delta_{\text{mix}}S, \Delta_{\text{mix}}V)$ denote the changes in Gibbs

free energy, enthalpy, entropy, and volume accompanying the process of preparing the subsystems $i\text{-H}_2\text{O}$ ($i=1, 2, \dots, n$) and the system 1-2-...- $n\text{-H}_2\text{O}$ having the same water activity as those of the subsystems $i\text{-H}_2\text{O}$ ($i=1, 2, \dots, n$), then substitution of Eq.(2) to the thermodynamic relationships and the Eyring's absolute rate theory[17] yields the following simple predictive equations for the thermodynamic properties and viscosities of the multicomponent solutions in terms of those of their binary subsystems:

$$\Delta_{\text{mix}}G = \sum_i \frac{m_i}{m_i^o} \Delta_{\text{mix}}G_i^o + RT \sum_i m_i \ln \frac{m_i}{\sum_i m_i} \quad (3)$$

$$\Delta_{\text{mix}}H = \sum_i \frac{m_i}{m_i^o} \Delta_{\text{mix}}H_i^o \quad (4)$$

$$\Delta_{\text{mix}}S = \sum_i \frac{m_i}{m_i^o} \Delta_{\text{mix}}S_i^o + R \sum_i m_i \ln \frac{m_i}{\sum_i m_i} \quad (5)$$

$$c_P = \sum_i \frac{m_i}{m_i^o} c_{P,i}^o \quad (6)$$

$$\gamma_i = \frac{m_i^o}{\sum_i m_i} \gamma_i^o \quad (7)$$

$$\Delta_{\text{mix}}V = \sum_i \frac{m_i}{m_i^o} \Delta_{\text{mix}}V_i^o \quad (8)$$

$$d = \sum_i Y_i / \sum_i (Y_i / d_i^o) \quad (9)$$

with $Y_i = m_i / m_i^o + m_i M_i$,

$$\ln \phi_m = \sum_{i=1}^n \frac{x_i}{x_i^o} \ln \phi_i^o, \quad (10)$$

where γ_i^o , $c_{P,i}^o$, ϕ_i^o , γ_i , $c_{P,i}$, and ϕ are activity coefficient, specific capacity, density, and kinetic viscosity, respectively. Substituting the relationship $\phi = \eta / d$ in Eq.(10) gives

$$\ln \eta = \sum_i \frac{x_i}{x_i^o} \ln \eta_i^o + \left(\ln d - \sum_i \frac{x_i}{x_i^o} \ln d_i^o \right) \quad (11)$$

For the multicomponent electrolyte solutions, it has been shown[6] that the second term on the right hand of Eq.(11) can be omitted so that the equation can be reduced to

$$\ln \eta = \sum_i \frac{x_i}{x_i^o} \ln \eta_i^o \quad (12)$$

For example, coupling Eq.(2) with Eqs.(13) and (14) below and remembering the fact that $a_{\text{H}_2\text{O}} = a_{i,\text{H}_2\text{O}}^o$, we obtain Eq.(8)

$$\Delta_{\text{mix}}V = n_{\text{H}_2\text{O}} (\bar{V}_{\text{H}_2\text{O}} - V_{\text{H}_2\text{O}}) + \sum_i n_i (\bar{V}_i - V_i) \quad (13)$$

$$\Delta_{\text{mix}} V_i^{\circ} = n_{\text{H}_2\text{O}}^{\circ} (V_{\text{H}_2\text{O}} - V_{\text{H}_2\text{O}}) + n_i^{\circ} (\bar{V}_i - V_i), \quad (14)$$

where V and \bar{V} denote molar volume and partial molar volume of pure component, respectively. With the help of $V_{\text{solution}} = \left(n_{\text{H}_2\text{O}} M_{\text{H}_2\text{O}} + \sum_i n_i M_i \right) / d_{\text{solution}}$,

$\Delta_{\text{mix}} V$ and $\Delta_{\text{mix}} V_i^{\circ}$ can be expressed as

$$\Delta_{\text{mix}} V = \frac{1}{d} \left(n_{\text{H}_2\text{O}} M_{\text{H}_2\text{O}} + \sum_i n_i M_i \right) - \frac{n_{\text{H}_2\text{O}} M_{\text{H}_2\text{O}}}{d_{\text{H}_2\text{O}}} - \sum_i \frac{n_i M_i}{d_i} \quad (15)$$

and

$$\Delta_{\text{mix}} V_i^{\circ} = \frac{1}{d_i^{\circ}} \left(n_{\text{H}_2\text{O}}^{\circ} M_{\text{H}_2\text{O}} + n_i^{\circ} M_i \right) - \frac{n_{\text{H}_2\text{O}}^{\circ} M_{\text{H}_2\text{O}}}{d_{\text{H}_2\text{O}}} - \frac{n_i^{\circ} M_i}{d_i}, \quad (16)$$

where $d_{\text{H}_2\text{O}}$ and d_i denote the densities of water and the solutes i , respectively. M is molar mass. From Eqs.(8), (15), and (16), Eq.(9) can be obtained.

3.2 Comparisons with the experimental data

Equations (9), (11), and (12) were tested by comparing with the measured densities and viscosities of the examined systems. The test procedure is briefly described as follows:

(1) Represent the measured densities and viscosities of all the binary solutions by the following polynomials

$$d_{i(\text{Exp})}^{\circ} = 0.99701 + \sum_l A_l (m_i^{\circ})^{1/2} \quad (17)$$

$$\eta_{i(\text{Exp})}^{\circ} = 0.8930 + \sum_l B_l (m_i^{\circ})^{1/2} \quad (18)$$

where $d_{i(\text{Exp})}^{\circ}$, $\eta_{i(\text{Exp})}^{\circ}$, and m_i° represent the density, viscosity, and molality of the binary aqueous solution i -H₂O. The optimum fit was obtained by increasing l until the resulting polynomials are accurate to a few parts in 10⁵ for the densities and in 10⁴ for the viscosities.

(2) Determine the compositions (m_i°) of the binary solutions having the same water activity as that of the multicomponent solution of given molalities m_i ($i = 1, 2, \dots, n$) using the osmotic coefficients of the binary solutions mannitol-H₂O[23], sorbitol-H₂O[24], and D-glucose-H₂O[25] and Eq.(2).

(3) Insert the values of $d_{i(\text{Exp})}^{\circ}$ and $\eta_{i(\text{Exp})}^{\circ}$ into Eq.(9), (11), and (12) to yield the predictions for the multicomponent solutions of given m_i ($i = 1, 2, \dots, n$), which were then compared with the corresponding experimental data.

In this article, the average absolute differences between the predicted and measured densities (Δ_d) and viscosities (Δ_{η_j}) over the entire experimental composition range of the multicomponent solution are defined by

$$\Delta_d = \sum_{i=1}^N \left| (d_{\text{Pred}} - d_{\text{Exp}})_i \right| / N \quad (19)$$

$$\Delta_{\eta_j} = \sum_{i=1}^N \left| (\eta_{\text{Pred}} - \eta_{\text{Exp}})_i \right| / N \quad (j = 1, 2), \quad (20)$$

where N is the number of experimental data. For convenience, Δ_{η_1} and Δ_{η_2} were used to represent the average absolute differences between the measured viscosities and the values predicted using Eq.(11) and (12) (denoted by η_1 and η_2), respectively.

Table 1 shows the measured densities and viscosities of the binary solutions mannitol-H₂O, sorbitol-H₂O, and D-glucose-H₂O at 298.15K. It is found that the measured densities and viscosities of D-glucose-H₂O at 298.15K are in good agreement with the literature values[26].

Table 1 The densities and viscosities of the binary systems at 298.15K

	$m, \text{mol}\cdot\text{kg}^{-1}$	$d, \text{g}\cdot\text{cm}^{-3}$	$\eta, \text{mPa}\cdot\text{s}^{-1}$
mannitol-H ₂ O	0.1999	1.00902	0.9818
	0.4002	1.02081	1.0852
	0.5998	1.03184	1.1905
	0.8006	1.04259	1.3113
	0.9995	1.05172	1.4422
sorbitol-H ₂ O	0.5085	1.01167	1.2580
	0.9996	1.05299	1.6121
	1.9508	1.09873	2.3286
	2.9980	1.13174	3.4350
	4.0003	1.16061	5.2208
	4.9970	1.18696	7.8960
	5.9945	1.20521	11.2820
D-glucose-H ₂ O	0.1998	1.01055	1.0019
	0.4998	1.02949	1.1255
	0.8000	1.04670	1.2952
	0.9995	1.05802	1.4188
	1.1990	1.06842	1.5501
	1.5000	1.08361	1.7664
	1.9990	1.10669	2.1932
	2.4992	1.12756	2.7365
	2.9991	1.14630	3.4165
	3.4975	1.16342	4.2353
3.9960	1.17881	5.1830	
4.4990	1.19321	6.2421	
4.9947	1.20631	7.3460	

Table 2 compares the predicted and measured densities for the quaternary system mannitol-sorbitol-D-glucose-H₂O at 298.15K, and the agreement, $\Delta_d = 3.5 \times 10^{-4}$, is good.

Table 2 Comparisons of the predicted and measured densities and viscosities of the quaternary system mannitol(B)-sorbitol(C)-D-glucose(D)-H₂O(A) at 298.15K

m_B^o , mol·kg ⁻¹	m_C^o , mol·kg ⁻¹	m_D^o , mol·kg ⁻¹	m_B , mol·kg ⁻¹	m_C , mol·kg ⁻¹	m_D , mol·kg ⁻¹	d_{Exp} , g·cm ⁻³	d_{Cal} , g·cm ⁻³	η_{Exp} , mPa·s	η_1 , mPa·s	η_2 , mPa·s
0.1534	0.1530	0.1522	0.04855	0.05036	0.05395	0.99465	0.99455	0.9912	0.9904	0.9904
0.2185	0.2185	0.2182	0.1023	0.04990	0.06626	1.00342	1.00322	1.0156	1.0142	1.0141
0.2566	0.2563	0.2562	0.0468	0.09985	0.1096	1.00168	1.00148	1.0480	1.0466	1.0465
0.3025	0.3018	0.3018	0.1267	0.1098	0.06558	1.00599	1.00574	1.0685	1.0677	1.0676
0.4232	0.4222	0.4218	0.1518	0.1369	0.1337	1.01669	1.01641	1.1293	1.1276	1.1272
0.5078	0.5068	0.5055	0.1780	0.1522	0.1765	1.02355	1.02316	1.1746	1.1717	1.1712
0.6002	0.5992	0.5966	0.2037	0.1870	0.2080	1.02990	1.02952	1.2295	1.2270	1.2266
0.6376	0.6367	0.6335	0.2365	0.2088	0.1908	1.03136	1.03177	1.2552	1.2515	1.2520
0.7023	0.7015	0.6971	0.2589	0.2215	0.2200	1.03669	1.03627	1.2943	1.2906	1.2901
0.8226	0.8219	0.8148	0.3037	0.2849	0.2315	1.04398	1.04364	1.3765	1.3728	1.3723
0.9035	0.9028	0.8936	0.3568	0.3070	0.2368	1.04895	1.04838	1.4291	1.4258	1.4250
1.0283	1.0276	1.0150	0.3612	0.3136	0.3487	1.05608	1.05545	1.5042	1.5009	1.4998

Tables 3 and 4 are the comparison of the predicted and measured densities for the ternary solutions mannitol-D-glucose-H₂O and sorbitol-D-glucose-H₂O at 298.15K. It can be observed from the fifth and sixth columns that the agreement is quite good, with the Δ_d values being 3.1×10^{-4} and 4.2×10^{-4} , respectively. It was shown that the ternary system mannitol-sucrose-H₂O[23] conforms to Eq.(2), and thus, the literature density data for this system[26] was used to

test Eq.(9). The result is $\Delta_d = 4.9 \times 10^{-4}$. It is also notable that the Δ_d values for the strong electrolyte mixtures, in general, range from 1.2×10^{-4} to 6.0×10^{-4} . For example, the average absolute difference (Δ_d) between the predicted and measured[27—31] densities at 298.15K are 3.8×10^{-4} for NaCl-KCl-H₂O ($I_{max} = 4.4$), 1.9×10^{-4} for KCl-KBr-H₂O ($I_{max} = 5.0$), 2.5×10^{-4} for KCl-CaCl₂-H₂O ($I_{max} = 3.6$), and 3.7×10^{-4} for HCl-KCl-NaCl-H₂O ($I_{max} = 2.2$), in which

Table 3 Comparisons of the predicted and measured densities and viscosities of the ternary system mannitol(B)-D-glucose(C)-H₂O(A) at 298.15K

m_B^o , mol·kg ⁻¹	m_C^o , mol·kg ⁻¹	m_B , mol·kg ⁻¹	m_C , mol·kg ⁻¹	d_{Exp} , g·cm ⁻³	d_{Cal} , g·cm ⁻³	η_{Exp} , mPa·s	η_1 , mPa·s	η_2 , mPa·s
0.1058	0.1057	0.06891	0.03687	0.97960	0.97939	0.9418	0.9413	0.9413
0.1582	0.1580	0.08122	0.07690	0.98895	0.98878	0.9750	0.9737	0.9737
0.2057	0.2054	0.1063	0.09926	0.99285	0.99299	0.9952	0.9936	0.9939
0.2580	0.2575	0.1285	0.1292	0.99882	0.99849	1.0176	1.0166	1.0164
0.3069	0.3062	0.1531	0.1535	1.00336	1.00301	1.0396	1.0380	1.0377
0.3586	0.3576	0.1768	0.1813	1.00818	1.00788	1.0625	1.0613	1.0611
0.4248	0.4233	0.2218	0.2022	1.01338	1.01309	1.0946	1.0930	1.0928
0.4926	0.4905	0.2360	0.2555	1.02009	1.01974	1.1285	1.1266	1.1263
0.5582	0.5552	0.2855	0.2712	1.02476	1.02451	1.1632	1.1615	1.1612
0.6115	0.6078	0.3137	0.2960	1.02898	1.02868	1.1931	1.1909	1.1905
0.6590	0.6545	0.3206	0.3361	1.03294	1.03262	1.2198	1.2175	1.2171
0.7065	0.7012	0.3517	0.3521	1.03635	1.03595	1.2480	1.2454	1.2449
0.7556	0.7493	0.3613	0.3910	1.03996	1.03960	1.2768	1.2742	1.2738
0.8021	0.7948	0.4269	0.3718	1.04260	1.04233	1.3069	1.3034	1.3031
0.8518	0.8433	0.4609	0.3870	1.04598	1.04561	1.3371	1.3346	1.3341
0.9063	0.8964	0.5058	0.3961	1.04942	1.04906	1.3721	1.3693	1.3689
0.9509	0.9398	0.5591	0.3872	1.05216	1.05173	1.4020	1.3986	1.3981
1.0025	0.9899	0.6267	0.3710	1.05515	1.05477	1.4362	1.4328	1.4323

Table 4 Comparisons of the predicted and measured densities and viscosities of the ternary system sorbitol(B)-D-glucose(C)-H₂O(A) at 298.15K

m_B^o , mol·kg ⁻¹	m_C^o , mol·kg ⁻¹	m_B , mol·kg ⁻¹	m_C , mol·kg ⁻¹	d_{Exp} , g·cm ⁻³	d_{Cal} , g·cm ⁻³	η_{Exp} , mPa·s	η_1 , mPa·s	η_2 , mPa·s
0.1036	0.1023	0.05228	0.05069	0.98460	0.98468	0.9890	0.9885	0.9882
0.1602	0.1595	0.08825	0.07164	0.98768	0.98760	1.0148	1.0138	1.0139
0.2125	0.2122	0.1036	0.1088	0.99485	0.99461	1.0356	1.0344	1.0344
0.2563	0.2562	0.0585	0.1976	1.00699	1.00689	1.0363	1.0356	1.0356
0.2569	0.2568	0.1249	0.1320	0.99895	0.99877	1.0565	1.0551	1.0551
0.3129	0.3128	0.1130	0.2002	1.00776	1.00756	1.0728	1.0719	1.0718
0.3132	0.3131	0.1855	0.1276	1.00125	1.00101	1.0950	1.0935	1.0934
0.3135	0.3135	0.2581	0.0555	0.99452	0.99461	1.1159	1.1151	1.1152
0.3140	0.3140	0.1130	0.2005	1.00786	1.00756	1.0731	1.0719	1.0718
0.3142	0.3142	0.1862	0.1280	1.00131	1.00101	1.0952	1.0935	1.0934
0.3150	0.3150	0.2588	0.0559	0.99469	0.99461	1.1158	1.1153	1.1152
0.3560	0.3560	0.1215	0.2351	1.01193	1.01168	1.0930	1.0913	1.0912
0.3567	0.3566	0.2356	0.1210	1.00368	1.00344	1.1261	1.1245	1.1244
0.3572	0.3570	0.2760	0.08068	1.00069	1.00055	1.1370	1.1364	1.1364
0.4023	0.4021	0.1595	0.2433	1.01452	1.01413	1.1235	1.1215	1.1214
0.4030	0.4026	0.2289	0.1742	1.01037	1.01012	1.1436	1.1411	1.1409
0.4037	0.4030	0.3136	0.0893	1.00536	1.00526	1.1665	1.1654	1.1653
0.4876	0.4871	0.2025	0.2852	1.02099	1.02067	1.1736	1.1716	1.1714
0.4885	0.4876	0.3167	0.1714	1.01658	1.01621	1.2055	1.2023	1.2020
0.4900	0.4883	0.4219	0.06646	1.01228	1.01213	1.2320	1.2310	1.2309
0.5281	0.5267	0.1587	0.36914	1.02609	1.02584	1.1825	1.1796	1.1795
0.5287	0.5270	0.2316	0.2965	1.02376	1.02344	1.2021	1.1984	1.1981
0.5289	0.5272	0.3167	0.2115	1.02094	1.02066	1.2236	1.2205	1.2203
0.5298	0.5277	0.4060	0.1225	1.01762	1.01775	1.2458	1.2437	1.2439
0.5306	0.5281	0.4625	0.0662	1.01609	1.01592	1.2615	1.2593	1.2591
0.5819	0.5802	0.2764	0.3053	1.02742	1.02706	1.2389	1.2360	1.2357
0.5825	0.5805	0.4869	0.0956	1.02176	1.02164	1.2915	1.2896	1.2895
0.6551	0.6518	0.3225	0.3312	1.03265	1.03231	1.2871	1.2837	1.2834
0.6555	0.6520	0.4170	0.2372	1.03086	1.03053	1.3094	1.3064	1.3062
0.6563	0.6526	0.5599	0.09502	1.02763	1.02783	1.3425	1.3413	1.3415
0.7018	0.6980	0.3967	0.3042	1.03536	1.03498	1.3143	1.3108	1.3104
0.7025	0.6985	0.3336	0.3668	1.03643	1.03596	1.3286	1.3254	1.3252
0.7029	0.6986	0.4285	0.2726	1.03482	1.03449	1.3360	1.3329	1.3327
0.7036	0.6991	0.5968	0.1053	1.03160	1.03189	1.3752	1.3727	1.3730
0.7678	0.7620	0.3536	0.4110	1.04096	1.04062	1.3528	1.3496	1.3494
0.8291	0.8218	0.4469	0.3788	1.04451	1.04412	1.4065	1.4033	1.4030
0.8552	0.8473	0.4913	0.3605	1.04598	1.04562	1.4306	1.4270	1.4267
0.9129	0.9035	0.5582	0.3511	1.04962	1.04919	1.4761	1.4726	1.4723
0.9618	0.9510	0.6097	0.3482	1.05258	1.05221	1.5137	1.5101	1.5097
1.0185	1.0062	0.6088	0.4050	1.05622	1.05589	1.4698	1.4666	1.4662
1.0188	1.0064	0.2276	0.7815	1.05788	1.05741	1.5085	1.5051	1.5047
1.0195	1.0069	0.4259	0.5858	1.05726	1.05662	1.5450	1.5415	1.5411
1.0202	1.0076	0.8182	0.1982	1.05556	1.05506	1.5860	1.5843	1.5838
1.2528	1.2330	0.6591	0.5843	1.06965	1.06919	1.6905	1.6871	1.6866
1.3721	1.3482	0.6976	0.6628	1.07581	1.07526	1.7716	1.7672	1.7668
1.6285	1.5954	0.7760	0.8352	1.08769	1.08723	1.9529	1.9491	1.9484
1.7620	1.7236	0.8021	0.9391	1.09355	1.09304	2.0520	2.0483	2.0478
1.8522	1.8107	0.8519	0.9778	1.09756	1.09678	2.1262	2.1227	2.1223
2.0582	2.0090	0.9093	1.1214	1.10552	1.10506	2.2991	2.2967	2.2960

Table 4 (Continued)

m_B^o , mol·kg ⁻¹	m_C^o , mol·kg ⁻¹	m_B , mol·kg ⁻¹	m_C , mol·kg ⁻¹	d_{Exp} , g·cm ⁻³	d_{Cal} , g·cm ⁻³	η_{Exp} , mPa·s	η_1 , mPa·s	η_2 , mPa·s
2.2035	2.1489	0.9637	1.2090	1.11150	1.11062	2.4338	2.4311	2.4304
2.3617	2.3013	1.0265	1.3010	1.11696	1.11646	2.5912	2.5881	2.5876
2.5298	2.4634	1.2298	1.2658	1.12282	1.12202	2.7816	2.7787	2.7775
2.6014	2.5325	1.5885	0.9860	1.12396	1.12339	2.8836	2.8808	2.8798
2.8032	2.7274	1.6253	1.1460	1.13086	1.13033	3.1207	3.1182	3.1172
2.9190	2.8394	1.7631	1.1243	1.13436	1.13380	3.2751	3.2726	3.2710
3.1065	3.0210	1.8924	1.18076	1.14028	1.13966	3.5372	3.5347	3.5331
3.2689	3.1787	1.9468	1.2856	1.14552	1.14487	3.7810	3.7784	3.7770
3.4060	3.3120	2.0587	1.3101	1.14968	1.14895	4.0043	4.0021	4.0003
3.5263	3.4291	2.1949	1.2947	1.15299	1.15234	4.2136	4.2108	4.2090
3.6519	3.5516	2.2860	1.3284	1.15670	1.15604	4.4413	4.4382	4.4364
3.8267	3.7225	2.4306	1.3580	1.16165	1.16105	4.7796	4.7758	4.7739
4.0265	3.9183	2.5565	1.4305	1.16768	1.16684	5.1876	5.1898	5.1861
4.2008	4.0895	2.8219	1.3423	1.17185	1.17122	5.5869	5.5846	5.5818
4.3668	4.2531	2.9768	1.3538	1.17646	1.17568	5.9816	5.9783	5.9749
4.5291	4.4134	3.1365	1.3570	1.18065	1.17992	6.3880	6.3845	6.3807
4.8029	4.6848	3.3836	1.3844	1.18768	1.18690	7.1095	7.1117	7.1071
4.9689	4.8499	3.6489	1.2883	1.19130	1.19056	7.5926	7.5958	7.5910
4.9691	4.8502	3.4620	1.4708	1.19209	1.19118	7.5638	7.5670	7.5608

I_{max} is the maximum ionic strength[12]. These results indicate that Eq.(8) holds well for both the multicomponent electrolyte solutions and the multicomponent nonelectrolyte solutions.

The second term on the right hand of Eq.(11) represents the contribution from the change in density accompanying the mixing process at constant water activity, which can be examined by comparing the predictions of Eq.(11) and (12). The results from the last three columns of Table 2 show that the agreement between the predicted and measured viscosities of the quaternary solution mannitol-sorbitol-D-glucose-H₂O at 298.15K is good. The values of Δ_{η_1} and Δ_{η_2} are 2.6×10^{-3} and 2.8×10^{-3} , respectively, suggesting that the effect of the above-mentioned density term is small. The last three columns of Tables 3 and 4 compare the predicted and measured viscosities of the ternary solutions mannitol-D-glucose-H₂O and sorbitol-D-glucose-H₂O at 298.15K. The values of Δ_{η_1} and Δ_{η_2} are $(2.2 \text{ and } 2.5) \times 10^{-3}$ and $(2.5 \text{ and } 3.2) \times 10^{-3}$, respectively, indicating that the predictions are in good agreement with the experimental results and that the addition of the density term $\left(\ln d - \sum_i \frac{x_i}{x_i^o} \ln d_i^o \right)$ has little effect on the predicted results, which may be due to the fact that the volume change accompanying the process of mixing of binary nonelectrolyte solutions at

constant water activity shows the ideal mixing behavior[1]. Note that the Δ_{η_2} values for the strong electrolyte mixtures, in general, range from 0.8×10^{-4} to 5.0×10^{-4} . For example, the average absolute differences (Δ_{η_2}) between the predicted and measured[28, 30,31] viscosities at 298.15K are 2.9×10^{-3} for NaCl-KCl-H₂O ($I_{max}=4.4$), 0.8×10^{-3} for HCl-KCl-H₂O ($I_{max}=4.0$), 2.5×10^{-3} for KCl-CaCl₂-H₂O ($I_{max}=3.6$), and 2.5×10^{-3} for HCl-KCl-NaCl-H₂O ($I_{max}=4.0$)[6], indicating that Eq.(12) holds well for both the multicomponent electrolyte solutions and the multicomponent nonelectrolyte solutions. However, because the isopiestic measurements are simple but time consuming, and up to now relatively few isopiestic measurements have been reported for the binary nonelectrolyte solutions, the practical applicability of these equations is limited.

It is clear that the values of Δ_d for the examined electrolyte and nonelectrolyte solutions are less than 5×10^{-4} , whereas those of Δ_{η_2} are more than 8×10^{-4} . Therefore, we recommend to predict the densities of the multicomponent solutions using Eq.(9), and then use Eq.(11) or Eq.(12) to predict the viscosities. We do not recommend to use Eq.(11) along with the experimental data for d_i^δ , η_i^o , and η to predict the densities of the multicomponent solutions because this means

that the Δ_d values will be likely larger than 1.0×10^{-3} for the above multicomponent solutions.

4 CONCLUSIONS

The densities and viscosities were measured for the quaternary system mannitol-sorbitol-D-glucose-H₂O and its ternary subsystems mannitol-D-glucose-H₂O and sorbitol-D-glucose-H₂O and at 298.15K. The results were used to test the applicabilities of the simple equations for the density and viscosity of the multicomponent nonelectrolyte solution. The comparison results are $3.1 \times 10^{-4} \leq \Delta_d \leq 4.210^{-4}$ and $2.2 \times 10^{-3} \leq \Delta_\eta \leq 3.2 \times 10^{-3}$. These results together with those for the multicomponent electrolyte solutions[6,13,15] show that Eq.(9), (11), and (12) can yield good predictions for the densities and viscosities of the multicomponent electrolyte solutions and the multicomponent nonelectrolyte solutions from the data of their binary solutions but do not involve mixing parameters, indicating that these simple equations can make use of the information on the binary solutions, avoid much complexity in calculation of mixture properties, and provide good predictions for the multicomponent solutions.

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