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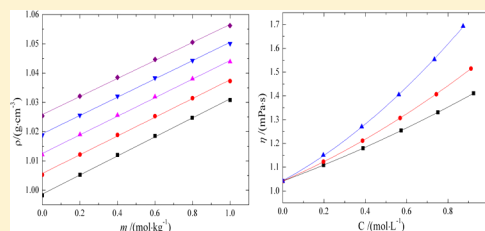
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# Densities and Viscosities of Erythritol, Xylitol, and Mannitol in L-ascorbic Acid Aqueous Solutions at $T = (293.15 \text{ to } 323.15) \text{ K}$

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**ABSTRACT:** The densities and viscosities of ternary solutions (erythritol/xylitol/mannitol + L-ascorbic acid + water) and binary solutions (erythritol/xylitol/mannitol + water), were measured under atmospheric pressure and the temperature range of (293.15 to 323.15) K. The apparent molar volumes ( $V_\phi$ ), the limiting partial molar volumes ( $V_\phi^0$ ) and the limiting partial molar volumes of transfer ( $\Delta_{tr}V_\phi^0$ ) were calculated through densities, respectively. Guimarães equation and extended Jones-Dole equation were applied to correlate the densities and viscosities of erythritol/xylitol/mannitol in L-ascorbic acid aqueous solutions, respectively. The free energy of activation per mole of solvent ( $\Delta\mu_1^{0\neq}$ ) and solute ( $\Delta\mu_2^{0\neq}$ ) were obtained through the viscosity  $B$ -coefficients. These significant parameters are helpful to understand the interactions in sugar alcohols + L-ascorbic acid + water ternary solutions.



## 1. INTRODUCTION

The density and viscosity of multicomponent solutions play a significant role in industrial areas concerning mass transfer, heat transfer, and fluid flow. They are fundamental to develop models for the engineering applications or simulation processes,<sup>1</sup> and also essential to further study the thermodynamic properties and transport properties of solutions. Therefore, studies on multicomponent liquid systems have attracted the increasing attention of many researchers.<sup>2–5</sup>

Recently, “functional sugar alcohols”, as alternative artificial nutritive sweeteners, have been extensively used in food and beverages (including diet drinks) owing to their desirable properties such as good taste, low calorie content, and no tooth decay. Additionally, they are also widely applied in many other products such as cosmetics, explosives, and plasticizers.<sup>6</sup> As a kind of carbohydrate, sugar alcohols are exceptionally important to biological systems and their thermodynamic and transport properties with other bioactive compounds are absolutely of great value.

It is well-known that, L-ascorbic acid (vitamin C, VC for short) is a ubiquitous and indispensable compound in living systems and is very crucial to metabolic processes. It is a sugar acid possessing antioxidant properties which could protect the body from radicals.<sup>7</sup> To our best knowledge, several researchers have studied the thermodynamic properties of sugar alcohols in amino acids or aqueous saccharides solutions,<sup>8–11</sup> but no report is available on the density and viscosity of sugar alcohols in VC aqueous solutions. The scarcity of basic thermodynamic data would hinder the progress in biology, pharmacy, and food industries. Thus in this work, the densities and viscosities of three typical polyalcohols, erythritol/xylitol/mannitol, + VC + water at  $T = (293.15, 303.15, 313.15, 323.15) \text{ K}$  were measured. The Guimarães equation<sup>12</sup> and extended Jones–Dole equation<sup>13</sup> were utilized to correlate densities and viscosities of solutions, respectively. Limiting partial molar volume ( $V_\phi^0$ ), limiting partial

molar volume of transfer ( $\Delta_{tr}V_\phi^0$ ), and the free energy of activation per mole of solvent ( $\Delta\mu_1^{0\neq}$ ) and solute ( $\Delta\mu_2^{0\neq}$ ) were calculated and discussed in terms of solute–solvent interactions.

## 2. EXPERIMENTAL SECTION

**2.1. Materials.** VC is a biochemistry reagent, and butane-1,2,3,4-tetrol (erythritol), (2R,3R,4S)-pentane-1,2,3,4,5-pentol (xylitol) and (2R,3R,4R,5R)-hexane-1,2,3,4,5,6-hexol (mannitol) are food additives. The specifications of VC, erythritol, xylitol, and mannitol have been given in Table 1. All solutions for the

Table 1. Specification of Studied Chemicals

chemical name	molar weight/(g·mol <sup>-1</sup> )	source	mass fraction purity
L-ascorbic acid (VC)	176.12	Tianjin Jiangtian Chemical Reagent Co., Ltd.	≥ 0.997
erythritol	122.12	O’Laughlin (Tianjin) Biotechnology Company	≥ 0.995
xylitol	152.15	Zhengzhou Jianda Chemicals Inc.	≥ 0.990
mannitol	182.17	Tianjin Jiangtian Chemical Reagent Co., Ltd.	≥ 0.990

whole molality range at room temperature were prepared by mass using an analytical balance (FA2204B, Shanghai Jingke, China) with an uncertainty of  $\pm 0.0001 \text{ g}$ , and the uncertainty of molality for all solutions is  $\pm 0.0001 \text{ mol·kg}^{-1}$ . The molality of VC is from (0.0 to 0.4)  $\text{mol·kg}^{-1}$  with an interval of 0.1  $\text{mol·kg}^{-1}$ , while that of erythritol/xylitol/mannitol is from (0.0 to 1.0)  $\text{mol·kg}^{-1}$  with an interval of 0.2  $\text{mol·kg}^{-1}$ .

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**Table 2.** Densities,  $\rho$ , Viscosities,  $\eta$ , and Apparent Molar Volumes,  $V_\phi$ , of Erythritol/Xylitol/Mannitol + VC + Water at Temperature  $T = (293.15, 303.15, 313.15, \text{ and } 323.15)$  K and Pressure  $p = 0.1$  MPa

$m^a$	$C$	$T/K = 293.15$			$T/K = 303.15$			$T/K = 313.15$			$T/K = 323.15$		
		$\rho$	$\eta$	$V_\varphi$	$\rho$	$\eta$	$V_\varphi$	$\rho$	$\eta$	$V_\varphi$	$\rho$	$\eta$	$V_\varphi$
mol·kg <sup>-1</sup>	mol·dm <sup>-3</sup>	g·cm <sup>-3</sup>	mPa·s	cm <sup>3</sup> ·mol <sup>-1</sup>	g·cm <sup>-3</sup>	mPa·s	cm <sup>3</sup> ·mol <sup>-1</sup>	g·cm <sup>-3</sup>	mPa·s	cm <sup>3</sup> ·mol <sup>-1</sup>	g·cm <sup>-3</sup>	mPa·s	cm <sup>3</sup> ·mol <sup>-1</sup>
Erythritol + 0.0000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	0.99821	1.002 <sup>b</sup>		0.99564	0.7977 <sup>b</sup>		0.99222	0.6532 <sup>b</sup>		0.98805	0.5470 <sup>b</sup>	
0.2002	0.1964	1.00526	1.069	86.36	1.00258	0.849	87.07	0.99905	0.690	87.81	0.99480	0.576	88.45
0.4001	0.3860	1.01203	1.134	86.47	1.00924	0.895	87.17	1.00561	0.727	87.91	1.00129	0.604	88.51
0.6001	0.5695	1.01854	1.20	86.57	1.01565	0.947	87.27	1.01190	0.766	88.03	1.00755	0.634	88.57
0.7984	0.7455	1.02474	1.278	86.69	1.02176	1.001	87.37	1.01792	0.806	88.10	1.01353	0.665	88.63
0.9998	0.9185	1.03079	1.358	86.80	1.02773	1.058	87.46	1.02381	0.849	88.18	1.01939	0.698	88.68
Erythritol + 0.1000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.00525	1.042		1.00256	0.828		0.99906	0.677		0.99478	0.565	
0.2005	0.1981	1.01222	1.109	86.48	1.00941	0.877	87.19	1.00580	0.715	87.94	1.00142	0.595	88.68
0.4003	0.3888	1.01889	1.180	86.60	1.01596	0.930	87.32	1.01225	0.753	88.04	1.00777	0.625	88.81
0.5999	0.5731	1.02526	1.254	86.74	1.02223	0.984	87.46	1.01844	0.794	88.16	1.01385	0.656	88.94
0.8000	0.7517	1.03139	1.331	86.88	1.02825	1.040	87.61	1.02439	0.835	88.27	1.01968	0.688	89.08
1.0001	0.9245	1.03728	1.411	87.02	1.03408	1.099	87.69	1.03012	0.881	88.37	1.02522	0.723	89.27
Erythritol + 0.2000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.01212	1.090		1.00933	0.858		1.00569	0.699		1.00131	0.584	
0.2002	0.1991	1.01901	1.164	86.47	1.01610	0.910	87.20	1.01235	0.741	87.96	1.00788	0.614	88.65
0.4001	0.3912	1.02560	1.232	86.61	1.02260	0.965	87.29	1.01874	0.780	88.04	1.01419	0.648	88.72
0.6001	0.5770	1.03193	1.307	86.73	1.02884	1.022	87.39	1.02487	0.823	88.15	1.02025	0.679	88.81
0.7998	0.7563	1.03802	1.390	86.83	1.03483	1.082	87.48	1.03076	0.868	88.23	1.02607	0.713	88.89
1.0000	0.9302	1.04389	1.477	86.92	1.04061	1.146	87.57	1.03645	0.915	88.32	1.03169	0.750	88.97
Erythritol + 0.3000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.01882	1.128		1.01591	0.894		1.01220	0.724		1.00778	0.603	
0.1999	0.2002	1.02561	1.203	86.55	1.02259	0.949	87.25	1.01877	0.766	87.99	1.01426	0.636	88.68
0.4002	0.3938	1.03213	1.278	86.69	1.02902	1.005	87.34	1.02512	0.809	88.03	1.02050	0.670	88.75
0.6001	0.5806	1.03836	1.359	86.83	1.03517	1.062	87.44	1.03120	0.854	88.10	1.02649	0.703	88.82
0.7996	0.7608	1.04434	1.447	86.94	1.04109	1.125	87.53	1.03704	0.901	88.16	1.03225	0.741	88.89
0.9999	0.9357	1.05010	1.539	87.06	1.04681	1.192	87.60	1.04271	0.950	88.21	1.03783	0.776	88.93
Erythritol + 0.4000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.02540	1.167		1.02239	0.922		1.01862	0.751		1.01408	0.623	
0.2000	0.2015	1.03211	1.245	86.62	1.02898	0.979	87.34	1.02511	0.792	88.08	1.02048	0.656	88.73
0.4002	0.3962	1.03853	1.327	86.78	1.03530	1.035	87.47	1.03132	0.837	88.21	1.02663	0.691	88.82
0.6004	0.5843	1.04468	1.415	86.92	1.04135	1.100	87.60	1.03731	0.882	88.27	1.03254	0.724	88.91
0.8000	0.7656	1.05055	1.504	87.06	1.04715	1.166	87.72	1.04301	0.932	88.39	1.03823	0.762	88.95
0.9998	0.9411	1.05620	1.611	87.17	1.05272	1.240	87.82	1.04852	0.987	88.47	1.04374	0.804	88.98
Xylitol + 0.0000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	0.99821	1.002 <sup>b</sup>		0.99564	0.7977 <sup>b</sup>		0.99222	0.6532 <sup>b</sup>		0.98805	0.5470 <sup>b</sup>	
0.2002	0.1959	1.00814	1.081	101.63	1.00540	0.858	102.61	1.00183	0.699	103.60	0.99751	0.582	104.59
0.4000	0.3836	1.01761	1.168	101.76	1.01472	0.917	102.73	1.01099	0.743	103.71	1.00653	0.617	104.70
0.5996	0.5642	1.02667	1.249	101.89	1.02362	0.981	102.86	1.01976	0.792	103.82	1.01516	0.655	104.81
0.7997	0.7382	1.03536	1.343	102.00	1.03215	1.056	102.98	1.02815	0.845	103.94	1.02343	0.695	104.92
1.0002	0.9061	1.04371	1.449	102.11	1.04036	1.125	103.08	1.03624	0.901	104.03	1.03137	0.739	105.03
Xylitol + 0.1000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.00525	1.042		1.00256	0.828		0.99906	0.677		0.99478	0.565	
0.2000	0.1970	1.01507	1.123	101.78	1.01221	0.888	102.77	1.00855	0.723	103.76	1.00414	0.601	104.69
0.4000	0.3863	1.02445	1.212	101.92	1.02144	0.953	102.87	1.01764	0.772	103.83	1.01310	0.639	104.74
0.6002	0.5684	1.03343	1.307	102.03	1.03028	1.021	102.96	1.02634	0.823	103.91	1.02171	0.681	104.77
0.8008	0.7439	1.04206	1.407	102.13	1.03875	1.096	103.08	1.03469	0.881	104.01	1.03000	0.724	104.80
0.9999	0.9115	1.05028	1.515	102.22	1.04684	1.171	103.15	1.04265	0.935	104.07	1.03788	0.764	104.85
Xylitol + 0.2000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.01212	1.090		1.00933	0.858		1.00569	0.699		1.00131	0.584	
0.2004	0.1987	1.02187	1.177	101.84	1.01891	0.921	102.83	1.01511	0.751	103.81	1.01059	0.624	104.80
0.4001	0.3889	1.03115	1.268	101.97	1.02803	0.990	102.95	1.02410	0.801	103.90	1.01944	0.663	104.86
0.6002	0.5720	1.04004	1.362	102.11	1.03677	1.063	103.05	1.03269	0.856	104.01	1.02794	0.705	104.91
0.8008	0.7485	1.04859	1.465	102.19	1.04515	1.139	103.16	1.04097	0.916	104.08	1.03610	0.751	104.98
1.0002	0.9173	1.05674	1.584	102.27	1.05316	1.220	103.24	1.04889	0.977	104.11	1.04390	0.794	105.01

Table 2. continued

$m^a$	$C$	T/K = 293.15			T/K = 303.15			T/K = 313.15			T/K = 323.15		
		$\rho$	$\eta$	$V_\varphi$	$\rho$	$\eta$	$V_\varphi$	$\rho$	$\eta$	$V_\varphi$	$\rho$	$\eta$	$V_\varphi$
mol·kg <sup>-1</sup>	mol·dm <sup>-3</sup>	g·cm <sup>-3</sup>	mPa·s	cm <sup>3</sup> ·mol <sup>-1</sup>	g·cm <sup>-3</sup>	mPa·s	cm <sup>3</sup> ·mol <sup>-1</sup>	g·cm <sup>-3</sup>	mPa·s	cm <sup>3</sup> ·mol <sup>-1</sup>	g·cm <sup>-3</sup>	mPa·s	cm <sup>3</sup> ·mol <sup>-1</sup>
Xylitol + 0.3000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.01882	1.128		1.01591	0.894		1.01220	0.724		1.00778	0.603	
0.2002	0.1998	1.02847	1.215	101.93	1.02539	0.958	102.92	1.02153	0.776	103.90	1.01696	0.643	104.86
0.4002	0.3915	1.03768	1.314	102.06	1.03445	1.030	103.00	1.03044	0.830	103.97	1.02575	0.685	104.89
0.6002	0.5755	1.04648	1.415	102.17	1.04310	1.107	103.11	1.03895	0.886	104.06	1.03417	0.729	104.94
0.8003	0.7526	1.05491	1.527	102.27	1.05140	1.189	103.20	1.04714	0.947	104.11	1.04225	0.778	104.97
1.0006	0.9231	1.06297	1.641	102.40	1.05935	1.270	103.29	1.05498	1.011	104.19	1.05002	0.822	105.01
Xylitol + 0.4000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.02540	1.167		1.02239	0.922		1.01862	0.751		1.01408	0.623	
0.2000	0.2009	1.03495	1.264	102.04	1.03176	0.993	103.03	1.02783	0.801	104.05	1.02315	0.663	105.01
0.3999	0.3936	1.04405	1.368	102.16	1.04071	1.063	103.14	1.03661	0.861	104.18	1.03182	0.708	105.07
0.6000	0.5788	1.05276	1.477	102.29	1.04927	1.150	103.24	1.04500	0.920	104.29	1.04013	0.756	105.12
0.8000	0.7567	1.06109	1.598	102.39	1.05746	1.233	103.33	1.05303	0.983	104.39	1.04809	0.804	105.17
1.0001	0.9279	1.06906	1.726	102.50	1.06529	1.329	103.43	1.06069	1.055	104.51	1.05569	0.855	105.26
Mannitol + 0.0000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	0.99821	1.002 <sup>b</sup>		0.99564	0.7977 <sup>b</sup>		0.99222	0.6532 <sup>b</sup>		0.98805	0.5470 <sup>b</sup>	
0.2003	0.1953	1.01060	1.106	118.96	1.00789	0.875	119.82	1.00433	0.713	120.70	1.00002	0.592	121.67
0.4001	0.3812	1.02232	1.219	119.13	1.01947	0.957	120.01	1.01578	0.774	120.92	1.01134	0.640	121.88
0.5999	0.5589	1.03348	1.345	119.27	1.03047	1.048	120.20	1.02664	0.842	121.13	1.02208	0.693	122.06
0.7997	0.7288	1.04408	1.486	119.44	1.04090	1.151	120.39	1.03696	0.918	121.30	1.03229	0.751	122.23
1.0002	0.8919	1.05422	1.634	119.58	1.05086	1.256	120.58	1.04681	0.996	121.48	1.04203	0.811	122.41
Mannitol + 0.1000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.00525	1.042		1.00256	0.828		0.99906	0.677		0.99478	0.565	
0.2002	0.1965	1.01746	1.151	119.37	1.01465	0.907	120.18	1.01102	0.737	121.04	1.00660	0.608	122.02
0.3990	0.3827	1.02899	1.270	119.51	1.02606	0.989	120.30	1.02231	0.797	121.14	1.01776	0.658	122.11
0.6000	0.5626	1.04006	1.405	119.67	1.03703	1.090	120.42	1.03318	0.868	121.23	1.02851	0.711	122.18
0.8015	0.7348	1.05061	1.554	119.80	1.04747	1.200	120.56	1.04353	0.955	121.35	1.03878	0.778	122.25
0.9705	0.8735	1.05910	1.693	119.89	1.05587	1.295	120.64	1.05187	1.023	121.41	1.04701	0.829	122.33
Mannitol + 0.2002 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.01212	1.090		1.00933	0.858		1.00569	0.699		1.00131	0.584	
0.1983	0.1960	1.02413	1.204	119.48	1.02121	0.938	120.26	1.01744	0.754	121.14	1.01293	0.625	122.07
0.3998	0.3860	1.03571	1.331	119.60	1.03267	1.036	120.39	1.02879	0.834	121.23	1.02417	0.687	122.10
0.5999	0.5660	1.04663	1.469	119.74	1.04349	1.137	120.51	1.03951	0.910	121.32	1.03481	0.746	122.14
0.8029	0.7405	1.05717	1.623	119.88	1.05394	1.249	120.62	1.04986	0.998	121.41	1.04511	0.808	122.17
0.9674	0.8762	1.06532	1.769	120.00	1.06202	1.353	120.72	1.05790	1.069	121.47	1.05312	0.866	122.19
Mannitol + 0.3141 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.01882	1.128		1.01591	0.894		1.01220	0.724		1.00778	0.603	
0.2002	0.1991	1.03085	1.250	119.52	1.02781	0.980	120.32	1.02396	0.789	121.22	1.01940	0.653	122.18
0.3999	0.3885	1.04224	1.387	119.66	1.03908	1.081	120.45	1.03512	0.866	121.31	1.03044	0.713	122.21
0.6001	0.5697	1.05307	1.537	119.80	1.04981	1.188	120.57	1.04574	0.949	121.41	1.04098	0.776	122.26
0.7996	0.7421	1.06334	1.705	119.93	1.05998	1.309	120.68	1.05582	1.039	121.50	1.05100	0.843	122.29
0.9920	0.9013	1.07277	1.888	120.05	1.06934	1.437	120.78	1.06508	1.130	121.59	1.06020	0.912	122.37
Mannitol + 0.4000 mol·kg <sup>-1</sup> VC													
0.0000	0.0000	1.02540	1.167		1.02239	0.922		1.01862	0.751		1.01408	0.623	
0.1993	0.1995	1.03726	1.294	119.67	1.03413	1.017	120.45	1.03023	0.819	121.34	1.02555	0.676	122.27
0.3986	0.3896	1.04850	1.439	119.84	1.04527	1.119	120.57	1.04125	0.902	121.44	1.03647	0.736	122.32
0.6011	0.5739	1.05934	1.600	119.98	1.05601	1.234	120.71	1.05189	0.981	121.53	1.04703	0.800	122.36
0.8045	0.7506	1.06967	1.779	120.14	1.06627	1.362	120.82	1.06204	1.076	121.64	1.05715	0.872	122.39
1.0042	0.9162	1.07928	1.973	120.31	1.07584	1.497	120.94	1.07151	1.175	121.76	1.06660	0.946	122.44

<sup>a</sup>The symbol  $m$  stands for the molality of erythritol/xylitol/mannitol in pure water or the (VC + water) mixture solvents. <sup>b</sup>The viscosities of pure water at  $T = (293.15, 303.15, 313.15, \text{ and } 323.15) \text{ K}$  refer to the Lange's Handbook of Chemistry.<sup>16</sup> Standard uncertainty: in molality  $u(m) = \pm 1 \cdot 10^{-4} \text{ mol} \cdot \text{kg}^{-1}$ ; in density  $u(\rho) = \pm 5 \cdot 10^{-5} \text{ g} \cdot \text{cm}^{-3}$ ,  $u(T) = \pm 0.03 \text{ K}$ ; in viscosities  $u(\eta) = \pm 1 \%$ ,  $u(T) = \pm 0.05 \text{ K}$ .

**2.2. Density Measurement.** The density measurements of all ternary solutions, binary solutions, and the pure water were performed by means of a vibrating tube density meter DMA 4500 (Anton Paar, Austria) with an uncertainty of  $\pm 5.0 \times 10^{-5} \text{ g} \cdot \text{cm}^{-3}$ . The temperature stability of the density measuring

cell was automatically controlled within  $\pm 0.03 \text{ K}$ . The apparatus was calibrated with double-distilled water and dry air at atmospheric pressure before each series of measurements. The standard values of density for calibration are  $0.998203 \text{ g} \cdot \text{cm}^{-3}$  for water and  $0.001199 \text{ g} \cdot \text{cm}^{-3}$  for dry air at  $293.15 \text{ K}$  and

101.325 kPa.<sup>14</sup> Triplicate measurements of each data were conducted to obtain the average value of density.

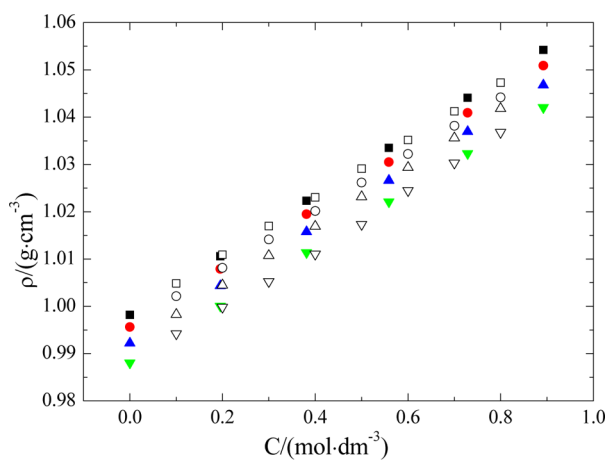
**2.3. Viscosity Measurement.** The viscosities of erythritol/xylitol/mannitol in VC aqueous solutions were measured using an iVisc capillary viscometer (LAUDA, Germany). A thoroughly cleaned and dried viscometer filled with experimental solutions was placed exactly vertical in a glass-sided water thermostat (ET 15S, LAUDA, Germany) controlled to  $\pm 0.05$  K. The efflux time of liquids was recorded automatically by computer software connected to the viscometer until thermal equilibrium. The uncertainty of the time measurement is  $\pm 0.01$  s. Triplicate independent experiments were taken for each sample at a specified temperature, and the deviation was less than 0.2 s. In the case that all the flow times were greater than 100 s and the capillary diameter (0.5 mm) was far less than its length (120 mm), the kinetic energy and the end corrections were negligible. The viscosity  $\eta$  of the solutions was calculated from the following equation:<sup>15</sup>

$$\frac{\eta}{\eta_w} = \frac{\rho t}{\rho_w t_w} \quad (1)$$

where  $\eta$ ,  $\rho$ ,  $t$  and  $\eta_w$ ,  $\rho_w$ ,  $t_w$  are viscosities, densities, and flow times of the solutions and pure water, respectively. The viscosity of pure water was obtained from Lange's Handbook of Chemistry.<sup>16</sup> The uncertainty of the experimental viscosity was  $\pm 1$  %.

### 3. RESULTS AND DISCUSSION

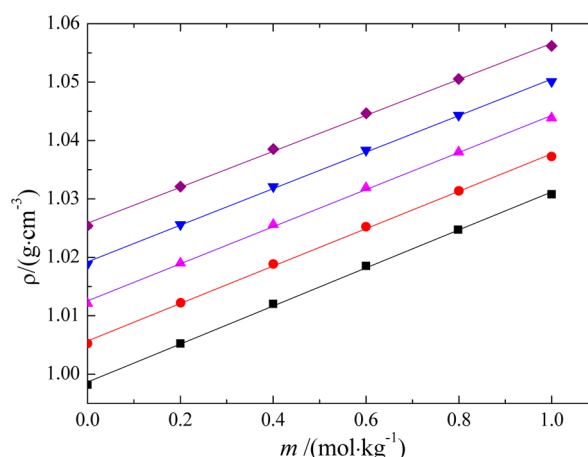
**3.1. The Density of Ternary Solutions.** The experimental densities of erythritol/xylitol/mannitol + VC + water ternary solutions and erythritol/xylitol/mannitol + water binary solutions at  $T = (293.15, 303.15, 313.15 \text{ and } 323.15) \text{ K}$  are listed in Table 2. From Figure 1, the measured densities of



**Figure 1.** Comparison of densities between the experimental data and literature data for mannitol aqueous solutions. Symbols: filled symbols refer to the experimental data and the open symbols stand for the data in the literature.<sup>17</sup> black ■, 293.15 K; red ●, 303.15 K; blue ▲, 313.15 K; green ▼, 323.15 K.

binary solutions (erythritol/xylitol/mannitol + water) show the same trend with the data in the literature<sup>17</sup> at  $T = (293.15, 303.15, 313.15 \text{ and } 323.15) \text{ K}$ .

Figure 2 gives the densities of erythritol in different molalities of VC aqueous solutions at  $T = 293.15 \text{ K}$ . From Table 2 and Figure 2, whether for binary or ternary solutions, the densities increase monotonously with the molality of erythritol, xylitol,



**Figure 2.** Densities of erythritol + VC + water ternary solutions at  $T = 293.15 \text{ K}$ : black ■, 0.0 mol·kg<sup>-1</sup>; red ●, 0.1 mol·kg<sup>-1</sup>; pink ▲, 0.2 mol·kg<sup>-1</sup>; blue ▼, 0.3 mol·kg<sup>-1</sup>; purple ◆, 0.4 mol·kg<sup>-1</sup>.

**Table 3.** Fitting Coefficients  $A_1$ ,  $A_2$ ,  $A_3$  of eq 2 for Erythritol/Xylitol/Mannitol + VC + Water Ternary Solutions

$m_{VC}^a$ mol·kg <sup>-1</sup>	$A_1$ g·cm <sup>-3</sup>	$10^4 A_2$ g·cm <sup>-3</sup> ·K <sup>-1</sup>	$A_3$ g·mol <sup>-1</sup>	100 AARD	SD g·cm <sup>-3</sup>
Erythritol					
0	1.1050	-3.6162	0.0347	0.037	0.0004
0.1000	1.1161	-3.7523	0.0338	0.039	0.0005
0.2000	1.1258	-3.8498	0.0334	0.038	0.0005
0.3000	1.1341	-3.9050	0.0329	0.037	0.0004
0.4000	1.1431	-3.9885	0.0321	0.035	0.0004
Xylitol					
0	1.1095	-3.7608	0.0490	0.040	0.0005
0.1000	1.1186	-3.8323	0.0483	0.037	0.0005
0.2000	1.1292	-3.9598	0.0475	0.038	0.0005
0.3000	1.1378	-4.0260	0.0467	0.036	0.0005
0.4000	1.1476	-4.1358	0.0459	0.035	0.0005
Mannitol					
0	1.1086	-3.7298	0.0616	0.038	0.0005
0.1000	1.1169	-3.7793	0.0607	0.039	0.0005
0.2002	1.1265	-3.8727	0.0599	0.038	0.0005
0.3141	1.1359	-3.9652	0.0590	0.036	0.0005
0.4000	1.1445	-4.0347	0.0581	0.036	0.0005

<sup>a</sup>  $m_{VC}$  is the molality of VC aqueous solution.

mannitol, and VC, and decrease monotonously with the temperature of solution. But for the same molality of VC, the densities of the three sugar alcohols decrease in the order: mannitol > xylitol > erythritol. In fact, the larger the molar weight is, the higher the density is. Thus the largest density of mannitol is attributed to its largest molar weight, whereas the smallest density of erythritol is due to its smallest molar weight.

The Guimarães equation has been applied to correlate the experimental density data.<sup>12</sup>

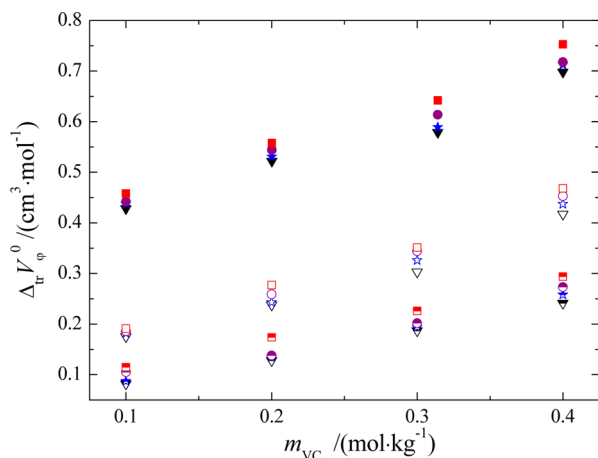
$$\rho = A_1 + A_2 T + A_3 C \quad (2)$$

where  $A_1$ ,  $A_2$ , and  $A_3$  are the empirical constants,  $T$  is the temperature, and  $C$  is the molar concentration of erythritol/xylitol/mannitol in VC aqueous solutions at 293.15 K. The conversion of molality  $m$  to molar concentration  $C$  is done by means of density values. The fitting parameters,  $A_1$ ,  $A_2$ , and  $A_3$  are shown in Table 3 alongside with the values of standard deviation (SD) and the average deviation (AARD).

**Table 4.** Limiting Partial Molar Volumes ( $V_\phi^0$ ) and Limiting Partial Molar Volumes of Transfer ( $\Delta_{tr}V_\phi^0$ ) of Erythritol/Xylitol/Mannitol + VC + Water Solutions at  $T = (293.15 \text{ to } 323.15) \text{ K}$ 

$m_{VC}^a$ mol·kg <sup>-1</sup>	T/K = 293.15		T/K = 303.15		T/K = 313.15		T/K = 323.15	
	$V_\phi^0$ cm <sup>3</sup> ·mol <sup>-1</sup>	$\Delta_{tr}V_\phi^0$ cm <sup>3</sup> ·mol <sup>-1</sup>	$V_\phi^0$ cm <sup>3</sup> ·mol <sup>-1</sup>	$\Delta_{tr}V_\phi^0$ cm <sup>3</sup> ·mol <sup>-1</sup>	$V_\phi^0$ cm <sup>3</sup> ·mol <sup>-1</sup>	$\Delta_{tr}V_\phi^0$ cm <sup>3</sup> ·mol <sup>-1</sup>	$V_\phi^0$ cm <sup>3</sup> ·mol <sup>-1</sup>	$\Delta_{tr}V_\phi^0$ cm <sup>3</sup> ·mol <sup>-1</sup>
Erythritol								
0.0000	86.24		86.96		87.71		88.39	
0.1000	86.32	0.08	87.05	0.09	87.82	0.11	88.50	0.11
0.2000	86.36	0.13	87.09	0.13	87.85	0.14	88.56	0.17
0.3000	86.42	0.18	87.15	0.19	87.92	0.21	88.61	0.22
0.4000	86.48	0.24	87.22	0.26	87.99	0.28	87.68	0.29
Xylitol								
0.0000	101.50		102.48		103.48		104.46	
0.1000	101.68	0.18	102.66	0.18	103.66	0.18	104.66	0.20
0.2000	101.74	0.24	102.73	0.25	103.74	0.26	104.74	0.28
0.3000	101.80	0.30	102.81	0.33	103.82	0.34	104.82	0.36
0.4000	101.92	0.42	102.92	0.44	103.93	0.45	104.93	0.47
Mannitol								
0.0000	118.79		119.60		120.49		121.47	
0.1000	119.22	0.43	120.04	0.44	120.94	0.45	121.93	0.46
0.2002	119.31	0.52	120.13	0.53	121.04	0.55	122.03	0.56
0.3141	119.37	0.58	120.19	0.59	121.11	0.62	122.12	0.65
0.4000	119.49	0.70	120.31	0.71	121.21	0.72	122.23	0.76

<sup>a</sup> $m_{VC}$  is the molality of VC aqueous solution.

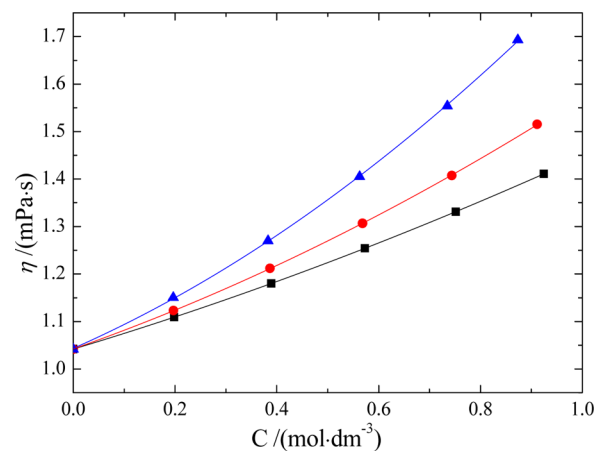
**Figure 3.** Transfer partial molar volumes of erythritol/xylitol/mannitol + VC + water solutions at different temperatures: black ▼, 293.15 K; blue ★, 303.15 K; purple ●, 313.15 K; red ■, 323.15 K. Symbols: filled symbols, mannitol; open symbols, xylitol; semifilled symbols, erythritol.

The standard deviation (SD) and the average deviation (AARD) are calculated as follows:

$$SD = \left[ \sum_{i=1}^n (y_{exp,i} - y_{calc,i})^2 / (n - m) \right]^{1/2} \quad (3)$$

$$AARD = \frac{1}{n} \sum_{i=1}^n \left| \frac{y_{exp,i} - y_{calc,i}}{y_{exp,i}} \right| \quad (4)$$

where  $n$  is the total number of experimental data point and  $m$  is the number of parameters.  $y_{exp,i}$  and  $y_{calc,i}$  refer to the experimental values and the calculated values, respectively. From Table 3, the maximum values of AARD and SD are 0.040 % and 0.0005 g·cm<sup>-3</sup>. Within the margin of error, eq 2 is

**Figure 4.** Viscosities of erythritol/xylitol/mannitol in 0.1 mol·kg<sup>-1</sup> VC aqueous solutions at  $T = 293.15 \text{ K}$ : ■, erythritol; red ●, xylitol; blue ▲, mannitol.

accurate enough to correlate the densities of erythritol/xylitol/mannitol + VC + water ternary solutions.

**3.2. Volumetric Properties.** The experimental densities of erythritol/xylitol/mannitol + VC + water solutions at  $T = (293.15 \text{ to } 323.15) \text{ K}$  (given in Table 2) are used to calculate apparent molar volume ( $V_\phi$ ) according to the following equation:<sup>18</sup>

$$V_\phi = \frac{M}{\rho} - \frac{1000(\rho - \rho_0)}{m\rho\rho_0} \quad (5)$$

where  $M$  is the molar weight of the solute,  $m$  is the molality of erythritol/xylitol/mannitol in solvent,  $\rho$  and  $\rho_0$  are the densities of the solution and the solvent, respectively. The calculated apparent molar volumes,  $V_\phi$ , are also included in Table 2.

It can be found that apparent molar volumes,  $V_\phi$ , have a good linear relationship with the molar concentration of the solute.



**Table 5. Viscosity  $B$  and  $D$  Coefficients of eq 8 for Erythritol/Xylitol/Mannitol + VC + Water Ternary Solutions at  $T = (293.15$  to  $323.15)$  K**

$m_{VC}^a$	$T$	$B$	$D$		SD		$m_{VC}^a$	$T$	$B$	$D$		SD
mol·kg <sup>-1</sup>	K	dm <sup>3</sup> ·mol <sup>-1</sup>	dm <sup>6</sup> ·mol <sup>-2</sup>	100 AARD	mPa·s		mol·kg <sup>-1</sup>	K	dm <sup>3</sup> ·mol <sup>-1</sup>	dm <sup>6</sup> ·mol <sup>-2</sup>	100 AARD	mPa·s
Erythritol							Xylitol					
0.0000	293.15	0.3090	0.0834	0.096	0.0018			313.15	0.3354	0.1062	0.100	0.0021
	303.15	0.2917	0.0675	0.117	0.0022			323.15	0.3194	0.0800	0.103	0.0018
	313.15	0.2697	0.0613	0.048	0.0009	0.3000	293.15	0.3629	0.1410	0.061	0.0010	
	323.15	0.2492	0.0551	0.062	0.0011		303.15	0.3408	0.1260	0.080	0.0015	
0.1000	293.15	0.3092	0.0798	0.007	0.0001			313.15	0.3302	0.1065	0.079	0.0013
	303.15	0.2873	0.0719	0.030	0.0005			323.15	0.3158	0.0862	0.103	0.0022
	313.15	0.2627	0.0671	0.068	0.0014	0.4000	293.15	0.3750	0.1509	0.093	0.0016	
	323.15	0.2502	0.0553	0.073	0.0012		303.15	0.3366	0.1488	0.152	0.0026	
0.2000	293.15	0.2997	0.0865	0.144	0.0029			313.15	0.3131	0.1309	0.126	0.0024
	303.15	0.2860	0.0798	0.041	0.0007			323.15	0.3072	0.1018	0.057	0.0012
	313.15	0.2737	0.0619	0.095	0.0018	Mannitol						
	323.15	0.2505	0.0579	0.108	0.0018	0.0000	293.15	0.4647	0.2712	0.076	0.0017	
0.3000	293.15	0.3008	0.0938	0.074	0.0015			303.15	0.4338	0.2350	0.096	0.0020
	303.15	0.2813	0.0787	0.107	0.0017			313.15	0.4122	0.1974	0.113	0.0024
	313.15	0.2723	0.0651	0.031	0.0006			323.15	0.3767	0.1842	0.055	0.0011
	323.15	0.2608	0.0494	0.082	0.0015	0.1000	293.15	0.4586	0.2908	0.133	0.0026	
0.4000	293.15	0.3005	0.1075	0.135	0.0027			303.15	0.4140	0.2659	0.129	0.0026
	303.15	0.2728	0.0982	0.099	0.0019			313.15	0.3745	0.2425	0.243	0.0046
	313.15	0.2491	0.0886	0.100	0.0016			323.15	0.3411	0.2245	0.167	0.0036
	323.15	0.2408	0.0700	0.158	0.0025	0.2002	293.15	0.4578	0.2835	0.221	0.0043	
Xylitol								303.15	0.4294	0.2582	0.136	0.0028
0.0000	293.15	0.3626	0.1399	0.191	0.0032			313.15	0.3971	0.2393	0.248	0.0050
	303.15	0.3454	0.1191	0.161	0.0029			323.15	0.3616	0.2162	0.284	0.0055
	313.15	0.3184	0.1098	0.115	0.0022	0.3141	293.15	0.4641	0.3105	0.191	0.0035	
	323.15	0.2944	0.1010	0.092	0.0017			303.15	0.4248	0.2742	0.118	0.0022
0.1000	293.15	0.3647	0.1453	0.040	0.0009			313.15	0.4125	0.2332	0.045	0.0009
	303.15	0.3420	0.1237	0.041	0.0008			323.15	0.3887	0.1997	0.069	0.0014
	313.15	0.3224	0.1063	0.080	0.0017	0.4000	293.15	0.4769	0.3000	0.114	0.0021	
	323.15	0.3118	0.0842	0.119	0.0021			303.15	0.4496	0.2505	0.114	0.0023
0.2000	293.15	0.3563	0.1463	0.210	0.0037			313.15	0.4174	0.2149	0.189	0.0038
	303.15	0.3467	0.1229	0.032	0.0006			323.15	0.3852	0.1966	0.057	0.0012

of erythritol/xylitol/mannitol molecules and the alkyl chain of VC molecules. The overlap of hydration cospheres of  $-\text{OH}$  group and  $-\text{OH}$  group result in a net increase in volume, while the overlap of hydrophilic–hydrophobic group and hydrophobic–hydrophobic group lead to a net decrease in volume. Thus among the above three interactions, only the first one contributes positively to the  $\Delta_{\text{tr}}V_{\varphi}^0$  values, whereas the hydrophilic–hydrophobic and hydrophobic–hydrophobic interactions contribute negatively to the  $\Delta_{\text{tr}}V_{\varphi}^0$  values.

From Table 4 and Figure 3, the  $\Delta_{\text{tr}}V_{\varphi}^0$  values of three sugar alcohols are positive, indicating that the hydrophilic–hydrophilic interactions surpass the hydrophilic–hydrophobic and hydrophobic–hydrophobic interactions. The  $\Delta_{\text{tr}}V_{\varphi}^0$  values are found to increase with the increase of the temperature or the molality of VC, which suggests that this predominant effect becomes more distinct when the temperature or the molality of VC increases. It is noted that from Figure 3, the  $\Delta_{\text{tr}}V_{\varphi}^0$  values of three sugar alcohols increase in the order erythritol < xylitol < mannitol, which coincides with the trend of  $V_{\varphi}^0$ . This can be rationalized in that mannitol with a longer alkyl chain is less solvated by water molecules, which leaves it more freedom to interact with VC molecules. Moreover, mannitol has the most  $-\text{OH}$  groups while erythritol has the least  $-\text{OH}$  groups. The more the  $-\text{OH}$  groups are, the stronger the hydrophilic–hydrophilic interaction or the hydrogen bonding is, which leads to the larger values of the limiting partial molar volume and the transfer partial molar volume.<sup>21,22</sup> Thus it can be concluded that the VC molality, solution temperature, and solute structure all have some bearing on the  $\Delta_{\text{tr}}V_{\varphi}^0$ .

**3.3. Viscometric Properties.** The experimental viscosities of erythritol/xylitol/mannitol + VC + water solutions at  $T = (293.15 \text{ to } 323.15) \text{ K}$  are given in Table 2 and the data of erythritol/xylitol/mannitol in  $0.1 \text{ mol}\cdot\text{kg}^{-1}$  VC aqueous solutions at  $T = 293.15 \text{ K}$  are depicted in Figure 4. The viscosities increase nonlinearly with the increase of the molar concentration of erythritol/xylitol/mannitol. And from Table 2, viscosities increase in direct proportion to the molality of VC but in inverse proportion to temperature. This is because when the molality of VC increases, the distance between molecules would be shortened and the interaction between solute and solvent molecules would be enhanced, which naturally leads to the increasing of viscosity. But with the increasing temperature, the molecular motion becomes much more rapid, and then the viscosity decreases. And it can be also observed from Figure 4 that for the same molality of VC, the viscosities of erythritol + VC + water solutions are the smallest, while the viscosities of mannitol + VC + water solutions are the largest mainly because of its highest molar weight.

The relative viscosities of erythritol/xylitol/mannitol in VC + water mixtures could be correlated by the extended Jones-Dole equation.<sup>13</sup>

$$\eta_r = \frac{\eta}{\eta_0} = 1 + BC + DC^2 \quad (8)$$

where  $\eta_r$  is the relative viscosity;  $\eta$  and  $\eta_0$  are the viscosities of ternary solutions (erythritol/xylitol/mannitol + VC + water) and the mixed solvents (VC + water), respectively.  $B$  and  $D$  are the constants at a given temperature and qualified with some physicochemical characteristics of solutions. Usually  $B$ -coefficient stands for solute–solvent interactions and  $D$ -coefficient seems to account for solute–solute interactions.<sup>23</sup> The fitting parameters  $B$  and  $D$  of viscosity are obtained by the least-squares deviations and

**Table 6.**  $\Delta\mu_1^{0\neq}$ ,  $\Delta\mu_2^{0\neq}$  of Erythritol/Xylitol/Mannitol + VC + Water at  $T = (293.15 \text{ to } 323.15) \text{ K}$

$m_{\text{VC}}^a$ mol·kg <sup>-1</sup>	$T$ K	$\Delta\mu_1^{0\neq}$ kJ·mol <sup>-1</sup>	$\Delta\mu_2^{0\neq}$ kJ·mol <sup>-1</sup>
Erythritol			
0.1000	293.15	9.41	59.97
	303.15	9.16	58.37
	313.15	8.94	56.20
	323.15	8.76	55.58
0.2000	293.15	9.54	58.39
	303.15	9.27	57.87
	313.15	9.05	57.43
	323.15	8.87	55.31
0.3000	293.15	9.64	58.22
	303.15	9.39	56.93
	313.15	9.16	56.92
	323.15	8.98	56.48
0.4000	293.15	9.75	57.86
	303.15	9.49	55.47
	313.15	9.28	53.41
	323.15	9.09	53.32
Xylitol			
0.1000	293.15	9.41	69.46
	303.15	9.16	68.09
	313.15	8.94	66.95
	323.15	8.76	66.94
0.2000	293.15	9.54	67.96
	303.15	9.27	68.32
	313.15	9.05	68.37
	323.15	8.87	67.63
0.3000	293.15	9.64	68.42
	303.15	9.39	67.13
	313.15	9.16	67.24
	323.15	8.98	66.71
0.4000	293.15	9.75	69.61
	303.15	9.49	66.17
	313.15	9.28	64.49
	323.15	9.09	65.10
Mannitol			
0.1000	293.15	9.41	84.40
	303.15	9.16	80.44
	313.15	8.94	76.82
	323.15	8.76	73.75
0.2002	293.15	9.54	83.77
	303.15	9.27	82.04
	313.15	9.05	79.51
	323.15	8.87	76.25
0.3141	293.15	9.65	83.91
	303.15	9.40	80.75
	313.15	9.17	81.01
	323.15	8.98	79.52
0.4000	293.15	9.75	85.22
	303.15	9.49	83.74
	313.15	9.28	81.34
	323.15	9.09	78.67

<sup>a</sup>  $m_{\text{VC}}$  is the molality of VC aqueous solution.

are shown in Table 5. From Table 5, the maximum values of AARD and SD are 0.284 % and 0.0055 mPa·s, respectively.

According to the Feakin's transition state theory,<sup>24</sup> the viscosity of erythritol/xylitol/mannitol + VC + water solutions can be investigated to analyze the free energy of activation per



mole of solvent ( $\Delta\mu_1^{0\neq}$ ) and the free energy of activation per mole of solute ( $\Delta\mu_2^{0\neq}$ ). The  $B$ -coefficient can be expressed as following equation

$$B = (\bar{V}_1^0 - \bar{V}_2^0) + \bar{V}_1^0 \left( \frac{\Delta\mu_2^{0\neq} - \Delta\mu_1^{0\neq}}{RT} \right) \quad (9)$$

where  $\bar{V}_1^0 (= \sum x_i M_i / \rho_0)$  is the mean molar volume of the solvent and  $\bar{V}_2^0 (= V_\varphi^0)$  is the standard partial molar volume of the solute at infinite dilution. The  $x_i$  and  $M_i$  specify the mole fraction and molar weight of water and VC in mixtures (VC + water), and  $\rho_0$  is the density of mixtures (VC + water).

The free energy of activation per mole of solvent ( $\Delta\mu_1^{0\neq}$ ) and the free energy of activation per mole of solute ( $\Delta\mu_2^{0\neq}$ ) can be calculated through following equations:<sup>25</sup>

$$\Delta\mu_1^{0\neq} = RT \ln \frac{\eta_0 \bar{V}_1^0}{h N_A} \quad (10)$$

$$\Delta\mu_2^{0\neq} = \Delta\mu_1^{0\neq} + \frac{RT}{\bar{V}_1^0} [B - (\bar{V}_1^0 - \bar{V}_2^0)] \quad (11)$$

where  $R$  is gas constant,  $N_A$  is Avogadro's number,  $h$  is Planck's constant, and  $\eta_0$  is the viscosity of the solvent. The calculated values of  $\Delta\mu_1^{0\neq}$  and  $\Delta\mu_2^{0\neq}$  at different temperatures are summarized in Table 6.

From Table 6, the values of  $\Delta\mu_2^{0\neq}$  of erythritol/xylitol/mannitol are positive and higher than the values of  $\Delta\mu_1^{0\neq}$  at various temperatures in (0.1, 0.2, 0.3 and 0.4) mol·kg<sup>-1</sup> VC + water mixture solvents, which suggests that the solute–solvent interactions exist and hinder the formation of a transition state in the presence of VC.<sup>26</sup> It is observed that the  $\Delta\mu_2^{0\neq}$  values decrease with the increase of temperature for a given solute, implying that high temperature favors the formation of the transition state, while for a given molality of VC, the  $\Delta\mu_2^{0\neq}$  values decrease in the order mannitol > xylitol > erythritol, which coincides with the trend of  $V_\varphi^0$  and  $\Delta_{tr}V_\varphi^0$ . This reveals that erythritol is the most favorable to the formation of a transition state among the three sugar alcohols.

#### 4. CONCLUSIONS

In the present work, densities and viscosities of erythritol/xylitol/mannitol in VC aqueous solutions have been measured at  $T = (293.15, 303.15, 313.15, \text{ and } 323.15)$  K under atmospheric pressure. A series of volumetric properties—apparent molar volume  $V_\varphi$ , limiting partial molar volume  $V_\varphi^0$ , and limiting partial molar volume of transfer  $\Delta_{tr}V_\varphi^0$ , have been obtained by density data. Positive  $\Delta_{tr}V_\varphi^0$  suggests that the hydrophilic–hydrophilic interactions take the predominant role, and when the temperature or the molality of VC increases, the values of  $\Delta_{tr}V_\varphi^0$  become larger. For erythritol/xylitol/mannitol, the number of the –OH groups in the solute molecules is a determining factor conditioning the strength of the hydrophilic–hydrophilic interactions. In addition, the densities and viscosities of erythritol/xylitol/mannitol in VC aqueous solutions are fitted well with the Guimarães equation and the extended Jones-Dole equation, respectively. The viscosity  $B$ -coefficients are applied to obtain the free energy of activation per mole of solvent ( $\Delta\mu_1^{0\neq}$ ) and the free energy of activation per mole of solute ( $\Delta\mu_2^{0\neq}$ ) and the results show that both the temperature and the solute molar weight have certain effect on the  $\Delta\mu_2^{0\neq}$ .

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##### Notes

The authors declare no competing financial interest.

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