Densities and Viscosities of Binary Mixtures of 1-Butanol with Methacrylic Acid, Benzyl Methacrylate, and 2-Hydroxyethyl Methacrylate between 288.15 K and 318.15 K

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Densities and viscosities of binary mixtures of 1-butanol with methacrylic acid, benzyl methacrylate, and 2-hydroxyethyl methacrylate were measured at temperatures from 288.15 K to 318.15 K. A pycnometer and a Cannon-Fenske routine viscometer were used to determine density and kinematic viscosity, respectively. The excess molar volumes V^E and viscosity deviations $\delta\eta$ were calculated from the experimental data and were correlated by a Redlich−Kister-type polynomial. McAllister's three-body and four-body interaction models were used to correlate the kinematic viscosities. These model parameters were also treated to be temperature dependent.

Introduction

Color filter is one of essential parts of color liquid crystal display, and the pigment dispersed color resist (PDCR) is the most important material for manufacture of this part. In our laboratory, we are interested in investigating the transport properties and storage stability of color resists. The thermophysical properties of a binary mixture such as density and viscosity are useful in the design of many types of process and transport equipment in chemical industries. Methacrylic acid (MAA; 2-methyl-2-propenoic acid), benzyl methacrylate (BzMA; 2-methyl-2-propenoic acid, phenylmethyl ester), 2-hydroxyethyl methacrylate (2-HEMA; 2-methyl-2-propenoic acid, 2-hydroxyethyl ester), and 1-butanol are the key compounds in the manufacturing of the pigment dispersed color resist industries. A series of density and viscosity measurements have been made recently in our laboratory.[1−3] This paper reports on the binary systems composed of 1-butanol with methacrylic acid, benzyl methacrylate, and 2-hydroxyethyl methacrylate at (288.15, 298.15, 308.15, and 318.15) K and over the entire composition range. No literature data were found at comparable conditions. From the experimental results, the excess molar volumes and the viscosity deviations were calculated and were correlated by a Redlich−Kister-type polynomial. McAllister's three-body and four-body interaction models were also used to correlate the kinematic viscosities. Furthermore, these model parameters were treated to be temperature dependent.

Experimental Section

MAA (mass fraction 99 %) was obtained from Sigma-Aldrich. 2-HEMA (mass fraction 98 %) was supplied by Acros Organics. BzMA (mass fraction 98 %) was purchased from Showa Chemical Co. Ltd. 1-Butanol (mass fraction 99 %) was obtained from J. T. Baker. The purities of these substances were checked with gas chromatography before use. All reagents were used without further purification. The densities were determined with a pycnometer having a nominal internal volume of 10 cm^3.

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Table 1. Densities ρ and Viscosities η of 1-Butanol at Different Temperatures

	ρ/g·cm^{-3}		η/(mPa·s)	
T/K	exptl	lit.	exptl	lit.
298.15	0.8056	0.80575[b,e]	2.566	2.571[a]
		0.8060[c]		2.571[b,c]
		0.80576[a]		2.509[d]
		0.8057[d]		2.578[e]
		0.8060[f]		2.570[f]
		0.8059[g]		2.506[g]
		0.8077[h]		2.564[h]
		0.80581[i,j]		2.550[i]
				2.550[j]
308.15	0.7980	0.79821[a]	1.998	1.981[a]
		0.7980[d]		2.000[c]
		0.7985[k,l]		1.929[d]
		0.8002[f]		1.982[k]
		0.7981[g]		1.982[l]
		0.8001[h]		2.000[f]
				1.927[g]
				2.000[h]
318.15	0.7905	0.7902[g]	1.550	1.493[g]

[a] Ref 5. [b] Ref 6. [c] Ref 7. [d] Ref 8. [e] Ref 9. [f] Ref 10. [g] Ref 11. [h] Ref 12. [i] Ref 13. [j] Ref 14. [k] Ref 15. [l] Ref 16.

The internal volume of the pycnometer was calibrated with pure water[4] at each temperature of interest. The sample mixture was prepared by mass with an uncertainty of ± 0.0001 in mole fraction. To minimize the evaporation during the sample preparation, the heavier component was charged first. Three loaded pycnometers were immersed in a thermostatic bath (Neslab GP-500), which was controlled to within ± 0.03 K. A precision digital thermometer (model 1560, Hart Scientific) with a thermistor probe was used to read the temperature with an uncertainty of ± 0.015 K. The mixture densities were obtained by averaging the results from these three replications. The uncertainty of reported densities was estimated to be less than ± 0.1 %. The sample compositions were frequently checked with gas chromatography at the end of measurements, indicating that the variations were minimal. The kinematic viscosities ν were measured using Cannon-Fenske routine viscometers (size 75, supplied by Cannon Instrument Co.). The viscometer was placed in a thermostatic water bath (TV-4000, TAMSON), in

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Table 2. Density ρ and Viscosity η for 1-Butanol (1) + MAA (2)

	ρ/(g·cm^{-3})				η/(mPa·s)				V^E/(cm^3·mol^{-1})				$\delta\eta$/(mPa·s)			
	T/K															
x_1	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15
0.0	1.0183	1.0095	0.9994	0.9892	1.447	1.267	1.078	0.945	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
0.1000	0.9963	0.9877	0.9780	0.9681	1.632	1.400	1.174	1.015	0.0030	0.0053	−0.0041	−0.0165	−0.001	0.003	0.006	0.009
0.2000	0.9747	0.9665	0.9574	0.9481	1.818	1.534	1.275	1.090	−0.0273	−0.0424	−0.0825	−0.1094	0.003	0.009	0.015	0.022
0.3000	0.9543	0.9463	0.9377	0.9287	2.009	1.672	1.377	1.160	−0.0945	−0.1231	−0.1839	−0.2197	0.010	0.017	0.025	0.033
0.4000	0.9342	0.9266	0.9181	0.9093	2.200	1.810	1.476	1.227	−0.1844	−0.2175	−0.2790	−0.3166	0.016	0.024	0.032	0.041
0.5000	0.9142	0.9070	0.8986	0.8901	2.392	1.947	1.574	1.294	−0.2722	−0.3063	−0.3532	−0.3876	0.022	0.029	0.037	0.044
0.6000	0.8942	0.8874	0.8790	0.8706	2.576	2.079	1.667	1.355	−0.3297	−0.3707	−0.3989	−0.4290	0.024	0.031	0.038	0.045
0.7000	0.8741	0.8673	0.8591	0.8510	2.756	2.204	1.756	1.413	−0.3332	−0.3915	−0.4090	−0.4364	0.023	0.029	0.036	0.043
0.8000	0.8537	0.8470	0.8392	0.8314	2.935	2.329	1.842	1.466	−0.2704	−0.3497	−0.3688	−0.3953	0.018	0.024	0.031	0.037
0.9000	0.8329	0.8267	0.8191	0.8115	3.111	2.453	1.925	1.515	−0.1482	−0.2258	−0.2494	−0.2711	0.009	0.015	0.020	0.025
1.0	0.8123	0.8056	0.7980	0.7905	3.286	2.566	1.998	1.550	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table 3. Density ρ and Viscosity η for 1-Butanol (1) + BzMA (2)

	ρ/(g·cm^{-3})				η/(mPa·s)				V^E/(cm^3·mol^{-1})				$\delta\eta$/(mPa·s)			
	T/K															
x_1	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15
0.0	1.0412	1.0347	1.0258	1.0170	2.871	2.302	1.885	1.580	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
0.1000	1.0278	1.0213	1.0125	1.0039	2.655	2.123	1.746	1.455	0.0759	0.0670	0.0568	0.0490	−0.250	−0.205	−0.160	−0.116
0.2000	1.0133	1.0068	0.9981	0.9895	2.519	2.021	1.656	1.388	0.1086	0.0997	0.0878	0.0791	−0.429	−0.344	−0.259	−0.174
0.3000	0.9975	0.9909	0.9823	0.9738	2.462	1.958	1.605	1.342	0.1166	0.1093	0.0975	0.0898	−0.547	−0.436	−0.326	−0.216
0.4000	0.9799	0.9733	0.9648	0.9564	2.410	1.919	1.568	1.308	0.1112	0.1039	0.0912	0.0839	−0.622	−0.500	−0.378	−0.256
0.5000	0.9603	0.9537	0.9453	0.9370	2.384	1.868	1.525	1.272	0.0981	0.0892	0.0746	0.0668	−0.678	−0.550	−0.422	−0.294
0.6000	0.9383	0.9316	0.9234	0.9152	2.375	1.834	1.505	1.247	0.0799	0.0692	0.0533	0.0447	−0.725	−0.589	−0.453	−0.317
0.7000	0.9132	0.9066	0.8985	0.8904	2.427	1.863	1.508	1.244	0.0577	0.0470	0.0323	0.0238	−0.753	−0.604	−0.455	−0.307
0.8000	0.8845	0.8779	0.8699	0.8620	2.510	1.982	1.590	1.302	0.0333	0.0256	0.0154	0.0088	−0.713	−0.559	−0.405	−0.250
0.9000	0.8513	0.8446	0.8368	0.8291	2.704	2.168	1.727	1.394	0.0111	0.0084	0.0047	0.0016	−0.513	−0.390	−0.267	−0.144
1.0	0.8123	0.8056	0.7980	0.7905	3.286	2.566	1.998	1.550	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table 4. Density ρ and Viscosity η for 1-Butanol (1) + 2-HEMA (2)

	ρ/(g·cm^{-3})				η/(mPa·s)				V^E/(cm^3·mol^{-1})				$\delta\eta$/(mPa·s)			
	T/K															
x_1	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15	288.15	298.15	308.15	318.15
0.0	1.0747	1.0671	1.0577	1.0486	8.234	5.784	4.194	3.181	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
0.1000	1.0545	1.0471	1.0379	1.0291	7.303	5.188	3.822	2.963	−0.0155	−0.0287	−0.0388	−0.0518	−0.421	−0.295	−0.170	−0.044
0.2000	1.0333	1.0259	1.0169	1.0083	6.437	4.643	3.432	2.692	−0.0273	−0.0445	−0.0598	−0.0817	−0.761	−0.548	−0.334	−0.121
0.3000	1.0109	1.0036	0.9948	0.9863	5.681	4.154	3.109	2.430	−0.0344	−0.0512	−0.0688	−0.0942	−0.987	−0.724	−0.462	−0.199
0.4000	0.9873	0.9800	0.9713	0.9630	5.118	3.738	2.829	2.222	−0.0363	−0.0516	−0.0699	−0.0940	−1.097	−0.817	−0.538	−0.258
0.5000	0.9622	0.9551	0.9466	0.9383	4.633	3.398	2.598	2.051	−0.0334	−0.0473	−0.0656	−0.0858	−1.107	−0.834	−0.561	−0.288
0.6000	0.9357	0.9286	0.9203	0.9121	4.195	3.092	2.399	1.900	−0.0266	−0.0396	−0.0573	−0.0735	−1.039	−0.787	−0.535	−0.283
0.7000	0.9076	0.9006	0.8924	0.8844	3.833	2.834	2.212	1.759	−0.0175	−0.0295	−0.0459	−0.0598	−0.912	−0.690	−0.468	−0.246
0.8000	0.8777	0.8708	0.8629	0.8550	3.534	2.666	2.089	1.657	−0.0081	−0.0182	−0.0319	−0.0449	−0.725	−0.544	−0.363	−0.182
0.9000	0.8460	0.8392	0.8315	0.8238	3.321	2.583	2.018	1.609	−0.0012	−0.0075	−0.0161	−0.0268	−0.445	−0.329	−0.213	−0.097
1.0	0.8123	0.8056	0.7980	0.7905	3.286	2.566	1.998	1.550	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table 5. Correlated Results of Excess Molar Volume V^E

mixture	*T*/K	A_0	A_1	A_2	A_3	σ/(cm^3·mol^{-1})
1-butanol + MAA	288.15	−1.0888	−1.5443	0.4406	0.7721	0.0095
	298.15	−1.2253	−1.5958	0.0005	−0.0130	0.0154
	308.15	−1.4126	−1.2191	0.0067	−0.7560	0.0101
	318.15	−1.5502	−1.1309	−0.0747	−0.9955	0.0091
1-butanol + BzMA	288.15	0.3923	−0.3177	0.1424	−0.2062	0.0015
	298.15	0.3567	−0.3592	0.0965	−0.0742	0.0011
	308.15	0.2983	−0.3965	0.0673	0.0537	0.0012
	318.15	0.2670	−0.4138	0.0219	0.1328	0.0011
1-butanol + 2-HEMA	288.15	−0.1336	0.1010	0.0636	−0.0029	0.0010
	298.15	−0.1893	0.1239	−0.0178	0.0366	0.0012
	308.15	−0.2624	0.1292	−0.0667	0.0446	0.0015
	318.15	−0.343	0.2153	−0.1462	−0.0656	0.0007

which the temperature was regulated to within ± 0.01 K. An electronic stop watch was used to measure the flow times. Triplicates or more measurements of flow times were reproducible within ± 0.2 % or less. The kinematic viscosities ν were obtained from the relation

$$\nu = kt \tag{1}$$

where k is the capillary constant of viscometer and t is the flow time. The viscometer was calibrated with double-distilled water at each working temperature, and the capillary constant at each specific temperature was determined by averaging 10 calibration runs. The uncertainty of viscosity measurements was estimated to within ± 1.0 %, and the values of absolute viscosities η were calculated by using the equation of $\eta = \rho\nu$. Since there are no available data for the methacrylates, the measurements of 1-butanol were conducted to test the validity of the experimental

Table 6. Correlated Results of Viscosity Deviation $\delta\eta$

mixture	T/K	B_0	B_1	B_2	B_3	σ/(mPa·s)
1-butanol + MAA	288.15	0.0905	0.0788	−0.062	−0.0274	0.001
	298.15	0.1124	0.0705	−0.0175	0.0394	0.002
	308.15	0.1411	0.0526	−0.0227	0.0786	0.001
	318.15	0.1820	0.0403	0.0257	0.1070	0.001
1-butanol + BzMA	288.15	−2.6875	−0.8576	−2.6130	−1.6843	0.023
	298.15	−2.2511	−1.1513	−1.4437	0.0513	0.012
	308.15	−1.6609	−0.7979	−0.9533	0.0769	0.007
	318.15	−1.1789	−0.5602	−0.5872	0.4852	0.004
1-butanol + 2-HEMA	288.15	−4.5958	1.0003	−0.6337	−1.8178	0.016
	298.15	−3.1788	−0.1766	−0.1051	−0.0808	0.011
	308.15	−2.0469	0.0424	0.0712	−0.5675	0.012
	318.15	−1.3329	−0.0745	0.6108	−0.4402	0.015

procedure. Table 1 compares the experimental results with the literature values. It shows that our measurements agree with literature values within the experimental uncertainties.

Results and Discussion

Experimental results for the three binary systems of 1-butanol with methacrylic acid, benzyl methacrylate, and 2-hydroxyethyl methacrylate are listed in Tables 2 to 4, respectively. Excess volumes V^E and viscosity deviations $\delta\eta$ were calculated from the experimental results by the following equations, respectively:

$$V^E = V_M - (x_1V_1 + x_2V_2) \tag{2}$$

$$\delta\eta = \eta_M - (x_1\eta_1 + x_2\eta_2) \tag{3}$$

where x_i, V_i, and η_i are the mole fraction, molar volume, and viscosity of the pure component i, respectively. The subscript M represents mixture properties. The excess volumes and viscosity deviations were correlated by a Redlich−Kister-type polynomial:[17]

$$V^E/(\mathrm{cm^3\cdot mol^{-1}}) = x_1x_2\sum_{k=0}^{3}A_k(x_1 - x_2)^k \tag{4}$$

$$\delta\eta/(\mathrm{mPa\cdot s}) = x_1x_2\sum_{k=0}^{3}B_k(x_1 - x_2)^k \tag{5}$$

The coefficients of A_k and B_k were obtained by fitting the equations to the experimental values with a least-squares method. The correlated results for excess volumes and viscosity deviations are given respectively in Tables 5 and 6, in which the tabulated standard deviation σ was defined as

$$\sigma = \left[\frac{\sum(Y_{exp} - Y_{cal})^2}{n - p}\right]^{1/2} \tag{6}$$

where Y refers to V^E or $\delta\eta$, n is the number of data points, and p is the number of coefficients. The subscripts, exp and cal, denote the experimental value and the calculated value, respectively.

The variations of V^E and $\delta\eta$ with the mole fraction of 1-butanol for these three investigated systems at 308.15 K are presented in Figures 1 and 2, respectively. Figure 1 shows that the excess molar volumes are asymmetric and positive in the 1-butanol + BzMA system, asymmetric and negative in the 1-butanol + MAA system, and negative but nearly zero in the 1-butanol + 2-HEMA system over the entire composition range. They imply that volume expansion takes place when 1-butanol mixes with benzyl methacrylate and that the volume contraction in the 1-butanol + MAA system is greater than in the 1-butanol + 2-HEMA system. The above results can be discussed in terms of several effects that may be simply divided into dispersion forces and hydrogen bond contributions. The positive V^E values may be attributed to the dispersion forces contribution between 1-butanol and BzMA molecules. On the other hand, the negative V^E values for binary mixtures of 1-butanol with MAA or 2-HEMA may be attributed to hydrogen bond formation through dipole−dipole interactions between 1-butanol and MAA or 2-HEMA molecules. Figure 2 illustrates that the viscosity deviations are negative in 1-butanol + BzMA as well as in

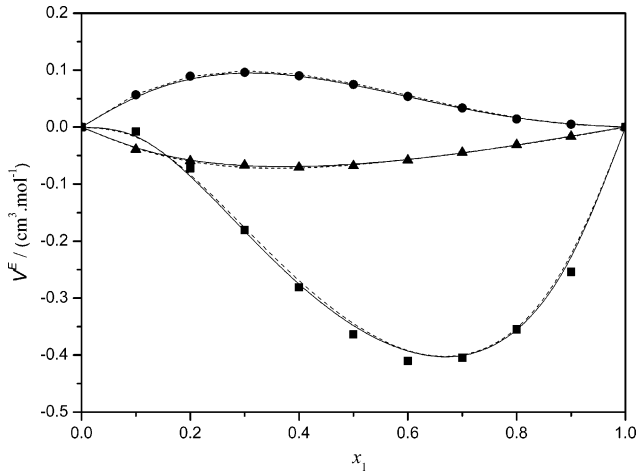

Figure 1. Excess volumes (V^E) at 308.15 K: ■, 1-butanol (1) + MAA (2); ●, 1-butanol (1) + BzMA (2); ▲, 1-butanol (1) + 2-HEMA (2); —, calculated from eq 4; - - -, calculated from eq 4 with temperature-dependent parameters of eq 9.

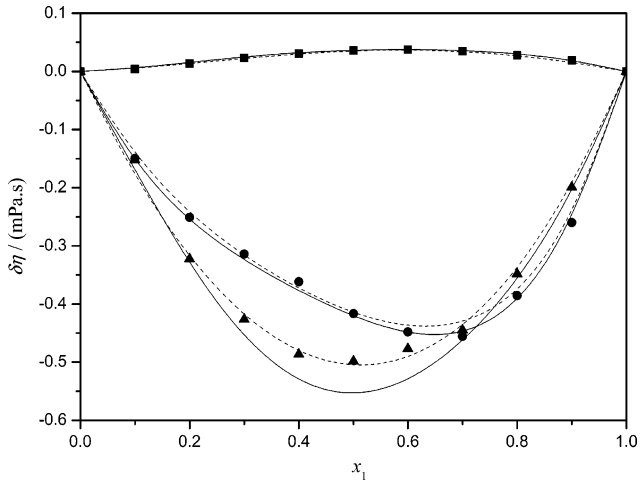

Figure 2. Viscosity deviations ($\delta\eta$) at 308.15 K: ■, 1-butanol (1) + MAA (2); ●, 1-butanol (1) + BzMA (2); ▲, 1-butanol (1) + 2-HEMA (2); —, calculated from eq 5 with temperature-dependent parameters of eq 10; - - -, calculated from eq 8.

Table 7. Correlated Results of McAllister's Models

		three-body model			four-body model			
mixture	T/K	ν_{12}	ν_{21}	AAD[a]·10²	ν_{1112}	ν_{1122}	ν_{2221}	AAD[a]·10²
1-butanol (1) + MAA (2)	288.15	3.1185	2.3253	0.04	3.3233	2.6976	2.0524	0.04
	298.15	2.5297	1.9104	0.06	2.6648	2.2123	1.7143	0.04
	308.15	2.0384	1.5728	0.06	2.1354	1.8027	1.4231	0.04
	318.15	1.6665	1.3224	0.09	1.7420	1.4726	1.2267	0.06
1-butanol (1) + BzMA (2)	288.15	2.1360	2.4414	1.89	2.1328	2.8421	2.2210	0.46
	298.15	1.5837	2.0103	1.33	1.7188	2.1416	1.8293	0.50
	308.15	1.3261	1.6598	1.24	1.4522	1.7235	1.5349	0.39
	318.15	1.1768	1.3733	1.01	1.2641	1.4327	1.2957	0.59
1-butanol (1) + 2-HEMA (2)	288.15	3.8669	5.5214	0.68	3.6827	4.9928	5.7604	0.48
	298.15	2.7611	4.1409	0.39	2.8039	3.4952	4.3440	0.24
	308.15	2.2251	3.1360	0.38	2.2529	2.7117	3.2377	0.28
	318.15	1.8239	2.5213	0.54	1.9243	1.9931	2.7102	0.48

[a] $AAD = (1/n)\sum_{k=1}^{n}|v_k^{cal} - v_k^{exp}|/v_k^{exp}$.

Table 8. Temperature-Dependent Parameters in the Redlich−Kister Type Polynomial and the McAllister's Model

1-Butanol (1) + MAA (2)

$A_0^0 = 3.4447$	$A_0^1 = -0.0157$	$B_0^0 = -0.7880$	$B_0^1 = 0.0030$	$\nu_{12}^0 = 15.4757$	$\nu_{12}^1 = -0.04341$	$\nu_{1112}^0 = 14.8606$	$\nu_{112}^1 = -0.0408$
$A_1^0 = -6.3074$	$A_1^1 = 0.0163$	$B_1^0 = 0.4664$	$B_1^1 = -0.0013$	$\nu_{21}^0 = 10.7621$	$\nu_{21}^1 = -0.0297$	$\nu_{1122}^0 = 12.8675$	$\nu 1122^1 = -0.0359$
$A_2^0 = 4.7615$	$A_2^1 = -0.0154$	$B_2^0 = -0.8005$	$B_2^1 = 0.0026$	AAD[a]·10² = 1.14		$\nu_{2221}^0 = 9.9445$	$\nu_{2221}^1 = -0.0274$
$A_3^0 = 18.1476$	$A_3^1 = -0.0607$	$B_3^0 = -1.2968$	$B_3^1 = 0.0044$			AAD[a]·10² = 1.81	
$\sigma = 0.0136$ cm³·mol⁻¹		$\sigma = 0.002$ mPa·s					

1-Butanol (1) + BzMA (2)

$A_0^0 = 1.6450$	$A_0^1 = -0.0043$	$B_0^0 = -17.4542$	$B_0^1 = 0.0512$	$\nu_{12}^0 = 8.4202$	$\nu_{12}^1 = -0.0228$	$\nu_{1112}^0 = 10.3546$	$\nu_{1112}^1 = -0.0289$
$A_1^0 = 0.6212$	$A_1^1 = -0.0033$	$B_1^0 = -4.5924$	$B_1^1 = 0.0124$	$\nu_{21}^0 = 12.5283$	$\nu_{21}^1 = -0.0352$	$\nu_{1122}^0 = 11.3834$	$\nu_{1122}^1 = -0.0309$
$A_2^0 = 1.2669$	$A_2^1 = -0.0039$	$B_2^0 = -21.3091$	$B_2^1 = 0.0657$	AAD[a]·10² = 2.22		$\nu_{2221}^0 = 10.8195$	$\nu_{2221}^1 = -0.0301$
$A_3^0 = -3.5045$	$A_3^1 = 0.0115$	$B_3^0 = -20.1266$	$B_3^1 = 0.0655$			AAD[a]·10² = 2.04	
$\sigma = 0.0016$ cm³·mol⁻¹		$\sigma = 0.016$ mPa·s					

1-Butanol (1) + 2-HEMA (2)

$A_0^0 = 1.8946$	$A_0^1 = -0.0070$	$B_0^0 = -35.8943$	$B_0^1 = 0.1092$	$\nu_{12}^0 = 18.2522$	$\nu_{12}^1 = -0.0516$	$\nu_{1112}^0 = 17.4720$	$\nu_{1112}^1 = -0.0491$
$A_1^0 = -0.9163$	$A_1^1 = 0.0035$	$B_1^0 = 9.3368$	$B_1^1 = -0.0301$	$\nu_{21}^0 = 28.3340$	$\nu_{21}^1 = -0.0813$	$\nu_{1122}^0 = 24.2421$	$\nu_{1122}^1 = -0.0695$
$A_2^0 = 2.0145$	$A_2^1 = -0.0068$	$B_2^0 = -11.8677$	$B_2^1 = 0.0391$	AAD[a]·10² = 2.55		$\nu_{2221}^0 = 30.8801$	$\nu_{2221}^1 = -0.0890$
$A_3^0 = 0.5538$	$A_3^1 = -0.0018$	$B_3^0 = -11.8363$	$B_3^1 = 0.366$			AAD[a]·10² = 2.92	
$\sigma = 0.0019$ cm³·mol⁻¹		$\sigma = 0.042$ mPa·s					

[a] $AAD = (1/n)\sum_{k=1}^{n}|v_k^{cal} - v_k^{exp}|/v_k^{exp}$.

1-butanol + 2-HEMA but nearly zero in 1-butanol + MAA over the entire composition range.

McAllister's multi-body interaction model[18] was widely used to correlate kinematic viscosity ν data. The three-body McAllister model was defined as

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln[x_1 + x_2(M_2/M_1)] + 3x_1^2 x_2 \ln[(2 + M_2/M_1)/3] + 3x_1 x_2^2 \ln[(1 + 2M_2/M_1)/3] + x_2^3 \ln(M_2/M_1) \quad (7)$$

and the four-body McAllister model was given by

$$\ln \nu = x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln \nu_{1112} + 6x_1^2 x_2^2 \ln \nu_{1122} + 4x_1 x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 - \ln[x_1 + x_2(M_2/M_1)] + 4x_1^3 x_2 \ln[(3 + M_2/M_1)/4] + 6x_1^2 x_2^2 \ln[(1 + M_2/M_1)/2] + 4x_1 x_2^3 \ln[(1 + 3M_2/M_1)/4] + x_2^4 \ln(M_2/M_1) \quad (8)$$

where ν_{12}, ν_{21}, ν_{1112}, ν_{1122}, and ν_{2221} are model parameters. The calculated results are presented in Table 7. The values of AAD are approximately within the experimental uncertainty, regardless of whether the three-body or the four-body model was used. As seen from Table 7, the four-body model yielded better calculated results than the three-body model did for those three investigated systems.

These parameters in eqs 4, 5, 7, and 8 were also treated to be temperature dependent, as given by the following equations, respectively:

$$A_k = A_k^0 + A_k^1 T/\mathrm{K} \quad (k = 0 \text{ to } 3) \quad (9)$$

$$B_k = B_k^0 + B_k^1 T/\mathrm{K} \quad (k = 0 \text{ to } 3) \quad (10)$$

$$\nu_{ij} = \nu_{ij}^0 + \nu_{ij}^1 T/\mathrm{K} \quad (ij = 12 \text{ or } 21) \quad (11)$$

$$\nu_{iiij} = \nu_{iiij}^0 + \nu_{iiij}^1 T/\mathrm{K} \quad (iiij = 1112 \text{ or } 2221) \quad (12)$$

$$\nu_{1122} = \nu_{1122}^0 + \nu_{1122}^1 T/\mathrm{K} \quad (13)$$

where A_k^0, A_k^1, B_k^0, B_k^1, ν_{ij}^0, ν_{ij}^1, ν_{iiij}^0, ν_{iiij}^1, ν_{1122}^0, and ν_{1122}^1 are the undetermined parameters. The best-fit values of the parameters together with the standard deviations σ of the calculated excess volumes and viscosity deviations and the average absolute deviations of the calculated kinematic viscosities for 1-butanol + MAA, 1-butanol + BzMA, and 1-butanol + 2-HEMA are given in Table 8. As seen from Tables 5, 6, and 8, the standard deviations of the calculated excess volumes and viscosity deviations are approximately within the experimental uncertainty, regardless of the parameters treating as temperature-specific or temperature-dependent in the Redlich−Kister-type polynomial. However, from Tables 7 and 8, the McAllister's

multi-body interaction model with temperature-specific parameters yielded better calculated results than with temperature-dependent parameters did for those three investigated systems.

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