

Excess Molar Volumes and Viscosities for Binary Mixtures of Cyclohexanone with Methacrylic Acid, Benzyl Methacrylate, and 2-Hydroxyethyl Methacrylate at (298.15, 308.15, and 318.15) K

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Densities and viscosities at (298.15, 308.15, and 318.15) K are presented as a function of mixture composition for the binary mixtures of cyclohexanone with methacrylic acid, benzyl methacrylate, and 2-hydroxyethyl methacrylate. From the experimental data, the excess molar volumes (V^E) and viscosity deviations ($\delta\eta$) have been calculated. These results have been correlated with the Redlich–Kister type polynomial to derive the coefficients and average absolute deviation (AAD). McAllister's three-body and four-body interaction models were also used to correlate the kinematic viscosities.

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. 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 design of many types of processes and transport equipment in chemical industries. Methacrylic acid (MAA), benzyl methacrylate (BzMA), 2-hydroxyethyl methacrylate (2-HEMA), and cyclohexanone are the key compounds in the manufacturing of the PDCR industries. A series of density and viscosity measurements have been made recently in our laboratory. This paper reports on the binary systems composed of cyclohexanone with methacrylic acid, benzyl methacrylate, and 2-hydroxyethyl methacrylate at (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 data, the excess molar volumes (V^E) and viscosity deviations ($\delta\eta$) have been calculated. These results have been correlated with the Redlich–Kister type polynomial to derive the coefficients and AAD. McAllister's three-body and four-body interaction models were also used to correlate the kinematic viscosities.

Experimental Section

Chemicals. Methacrylic acid (mass fraction 99.5 %) and 2-hydroxyethyl methacrylate (mass fraction 98 %) were Acros Organics products. Cyclohexanone (mass fraction 99.5+ %) was from Riedel-deHaën, and benzyl methacrylate (mass fraction 98 %) was supplied by Lancaster Chemicals. According to gas chromatographic analysis, no impurity peaks were detected. All reagents were used without further purification.

Density Measurement. The densities were determined with a pycnometer having a nominal internal volume of 10 cm³. The internal volume of the pycnometer was calibrated with pure water¹ at each temperature. The sample mixture was prepared by mass with an uncertainty of ± 0.0001 in mole fraction. To minimize evaporation during the sample preparation, the less

volatile 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 a gas chromatography at the end of measurements indicating that there was no change in the compositions.

Viscosity Measurement. 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 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 , in m²·s⁻¹) were obtained from

$$\nu = kt \quad (1)$$

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 $\eta = \rho\nu$.

The measured densities and viscosities of cyclohexanone at (298.15 and 308.15) K were compared with the literature values, and the results are presented in Table 1. It shows that our measurements agree with literature values within the experimental uncertainties.

Results and Discussion

Experimental results for the three binary systems of cyclohexanone with methacrylic acid, benzyl methacrylate, and 2-hydroxyethyl methacrylate are listed in Tables 2 to 4, respectively. Figure 1 illustrates the densities of these three binaries varying with composition at 308.15 K. The densities decrease monotonically

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Table 1. Densities (ρ) and Viscosities (η) of Cyclohexanone at Different Temperatures

T/K	$\rho/g\cdot cm^{-3}$		$\eta/mPa\cdot s$	
	exptl	lit	exptl	lit
298.15	0.9420	0.9418 ^a 0.9410 ^b 0.9420 ^c 0.9425 ^d	1.974	2.0205 ^a 2.0212 ^b 2.2290 ^c 1.9630 ^d
308.15	0.9334	0.9365 ^b 0.9328 ^c 0.9323 ^d	1.632	1.5849 ^b 1.6350 ^c 1.6020 ^d
318.15	0.9242		1.373	

^a Riddick et al.² ^b Singh et al.³ ^c Aralaguppi et al.⁴ ^d Nayak et al.⁵**Table 2. Density (ρ) and Viscosity (η) for Cyclohexanone (1) + MAA (2)**

x_1	$\rho/(g\cdot cm^{-3})$ at T/K			$\eta/(mPa\cdot s)$ at T/K		
	298.15	308.15	318.15	298.15	308.15	318.15
0.0	1.0095	0.9994	0.9892	1.267	1.078	0.945
0.1000	1.0020	0.9922	0.9823	1.369	1.159	0.999
0.2000	0.9949	0.9853	0.9756	1.528	1.281	1.101
0.3000	0.9880	0.9787	0.9693	1.659	1.389	1.183
0.4000	0.9815	0.9723	0.9631	1.809	1.508	1.275
0.5000	0.9750	0.9659	0.9567	1.901	1.583	1.330
0.5999	0.9685	0.9595	0.9504	1.949	1.616	1.359
0.7000	0.9620	0.9532	0.9442	1.999	1.651	1.395
0.8000	0.9554	0.9468	0.9377	2.045	1.683	1.424
0.9000	0.9491	0.9405	0.9313	2.037	1.684	1.422
1.0	0.9420	0.9334	0.9242	1.974	1.632	1.373

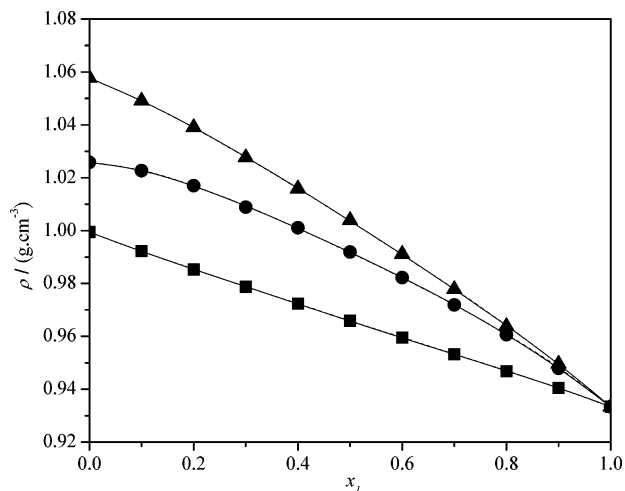
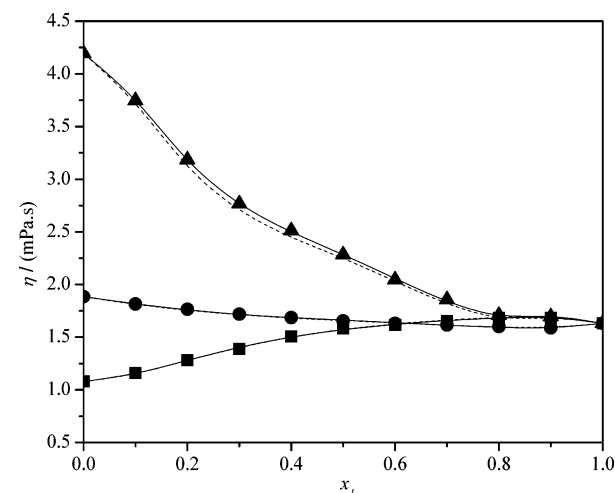
Table 3. Density (ρ) and Viscosity (η) for Cyclohexanone (1) + BzMA (2)

x_1	$\rho/(g\cdot cm^{-3})$ at T/K			$\eta/(mPa\cdot s)$ at T/K		
	298.15	308.15	318.15	298.15	308.15	318.15
0.0	1.0347	1.0258	1.0170	2.302	1.885	1.580
0.1000	1.0318	1.0227	1.0137	2.208	1.816	1.519
0.1999	1.0262	1.0170	1.0079	2.141	1.765	1.484
0.3000	1.0182	1.0089	0.9998	2.079	1.718	1.447
0.3999	1.0100	1.0011	0.9920	2.038	1.687	1.422
0.5000	1.0009	0.9919	0.9828	2.002	1.656	1.398
0.6000	0.9909	0.9822	0.9728	1.972	1.632	1.383
0.7000	0.9808	0.9719	0.9628	1.950	1.617	1.369
0.8000	0.9695	0.9606	0.9515	1.943	1.602	1.356
0.9000	0.9570	0.9479	0.9390	1.934	1.589	1.348
1.0	0.9420	0.9334	0.9242	1.974	1.632	1.373

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

x_1	$\rho/(g\cdot cm^{-3})$ at T/K			$\eta/(mPa\cdot s)$ at T/K		
	298.15	308.15	318.15	298.15	308.15	318.15
0.0	1.0671	1.0577	1.0486	5.784	4.194	3.181
0.1000	1.0580	1.0491	1.0405	5.035	3.747	2.881
0.2000	1.0478	1.0390	1.0301	4.220	3.184	2.474
0.3000	1.0364	1.0277	1.0188	3.579	2.768	2.200
0.4000	1.0246	1.0159	1.0071	3.213	2.512	2.012
0.5000	1.0124	1.0039	0.9950	2.928	2.282	1.853
0.6000	0.9995	0.9911	0.9823	2.661	2.045	1.678
0.6999	0.9861	0.9778	0.9691	2.319	1.856	1.540
0.8000	0.9723	0.9638	0.9551	2.089	1.707	1.439
0.9000	0.9581	0.9495	0.9405	2.060	1.695	1.421
1.0	0.9420	0.9334	0.9242	1.974	1.632	1.373

cally with an increase of mole fraction of cyclohexanone (x_1) for each system. Figure 2 shows the variations of the absolute viscosities with the mole fraction of cyclohexanone for these three investigated systems at 308.15 K. In the methacrylic acid system, the viscosity increases with the mole fraction of cyclohexanone, reaching a weak maximum at about $x_1 = 0.9$, and then decreasing slightly to the value of pure cyclohexanone. In the benzyl methacrylate system, the viscosity decreases with the mole fraction of cyclohexanone, reaching a weak minimum at about $x_1 = 0.9$, and then increasing slightly to the value of pure cyclohexanone. However, the viscosities of the 2-hydroxy-

**Figure 1.** Density (ρ) at 308.15 K: ■, cyclohexanone (1) + MAA (2); ●, cyclohexanone (1) + BzMA (2); ▲, cyclohexanone (1) + 2-HEMA (2); —, calculated from eq 4; - - -, calculated from eq 4 with temperature-dependent parameters of eq 10.**Figure 2.** Viscosity (η) at 308.15 K: ■, cyclohexanone (1) + MAA (2); ●, cyclohexanone (1) + BzMA (2); ▲, cyclohexanone (1) + 2-HEMA (2); —, calculated from eq 5; - - -, calculated from eq 5 with temperature-dependent parameters of eq 11.**Table 5. Parameters in Equations 2 and 3 for Density and Viscosity Data of Pure Fluids**

	a_0	$a_1 \times 10^4$	b_0	b_1	$b_2 \times 10^4$
cyclohexanone	1.20745	-8.90	50.2988	-0.2858	4.15
MAA	1.31214	10.20	32.6270	-0.1886	2.80
BzMA	1.29855	-8.85	66.1848	-0.3812	5.60
2-HEMA	1.34284	7.25	318.2490	-1.9081	28.80

ethyl methacrylate system decrease monotonically with the mole fraction of cyclohexanone. In this paper, the measured densities and viscosities for pure fluids are also presented as functions of temperature by the following equations, respectively:

$$\rho = a_0 + a_1(T/K) \quad (2)$$

$$\eta = b_0 + b_1(T/K) + b_2(T/K)^2 \quad (3)$$

where a_0 , a_1 , b_0 , b_1 , and b_2 are the undetermined parameters. The best-fit values of the parameters for pure fluids are given in Table 5. According to eqs 2 and 3 and the parameters of pure fluids in Table 5, the average absolute deviations of the calculated densities and viscosities for pure fluids are approximately close to zero.

Table 6. Correlated Results of Excess Molar Volume (V^E)

mixture	T/K	A_0	A_1	A_2	A_3	AAD ^a /%
cyclohexanone + MAA	298.15	-0.9678	-0.4029	-0.2193	-0.5227	0.01
	308.15	-1.0605	-0.4032	-0.3245	-0.5980	0.01
	318.15	-1.2933	-0.4607	-0.2241	-0.4687	0.01
cyclohexanone + BzMA	298.15	-0.6343	2.1230	-3.8737	0.9606	0.01
	308.15	-0.6024	1.7613	-3.2705	1.7799	0.01
	318.15	-0.4782	1.8485	-3.5285	1.0890	0.02
cyclohexanone + 2-HEMA	298.15	-1.2117	0.5872	-1.3837	-0.7517	0.01
	308.15	-1.4730	0.4497	-1.5522	-0.1012	0.01
	318.15	-1.6377	0.0813	-2.0634	0.8380	0.01

$$^a \text{AAD}/\% = (100/n) \sum_{k=1}^n |\rho_k^{\text{cal}} - \rho_k^{\text{exp}}| / \rho_k^{\text{exp}}$$

Table 7. Correlated Results of Viscosity Deviation ($\delta\eta$)

mixture	T/K	B_0	B_1	B_2	B_3	B_4	AAD ^a /%
cyclohexanone + MAA	298.15	1.0845	0.1530	-0.2459	0.8818		0.4
	308.15	0.8634	0.0811	-0.2069	0.7664		0.4
	318.15	0.6659	0.0774	-0.1317	0.7550		0.3
cyclohexanone + BzMA	298.15	-0.5340	0.0498	-0.2965	-0.1971		0.2
	308.15	-0.3876	0.0482	-0.3314	-0.3350		0.2
	318.15	-0.2904	0.0314	-0.2629	-0.1116		0.2
cyclohexanone + 2-HEMA	298.15	-3.7734	1.7706	-5.1374	-2.0669	8.2595	0.5
	308.15	-2.5173	0.7711	-3.2429	-1.2303	5.9965	0.3
	318.15	-1.6959	0.5462	-2.4163	-0.9724	4.4874	0.3

$$^a \text{AAD}/\% = (100/n) \sum_{k=1}^n |\eta_k^{\text{cal}} - \eta_k^{\text{exp}}| / \eta_k^{\text{exp}}$$

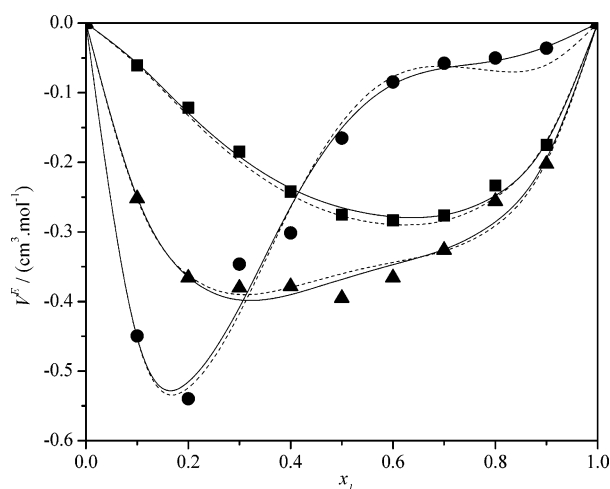


Figure 3. Excess volumes (V^E) at 308.15 K: ■, cyclohexanone (1) + MAA (2); ●, cyclohexanone (1) + BzMA (2); ▲, cyclohexanone (1) + 2-HEMA (2); —, calculated from eq 4; ---, calculated from eq 4 with temperature-dependent parameters of eq 10.

Excess volumes (V^E) and viscosity deviations ($\delta\eta$) were calculated from the experimental results by the following equations, respectively:

$$V^E = V_M - (x_1 V_1 + x_2 V_2) \quad (4)$$

$$\delta\eta = \eta_M - (x_1 \eta_1 + x_2 \eta_2) \quad (5)$$

where x_1 and x_2 are the mole fractions; V_1 and V_2 are the molar volumes; and η_1 and η_2 are the viscosities of components 1 and 2, respectively. The subscript M represents mixture properties. The excess volumes and viscosity deviations were correlated by a Redlich–Kister type polynomial:⁶

$$V^E/(\text{cm}^3 \cdot \text{mol}^{-1}) = x_1 x_2 \sum_{k=0}^3 A_k (x_1 - x_2)^k \quad (6)$$

$$\delta\eta/(\text{mPa} \cdot \text{s}) = x_1 x_2 \sum_{k=0}^4 B_k (x_1 - x_2)^k \quad (7)$$

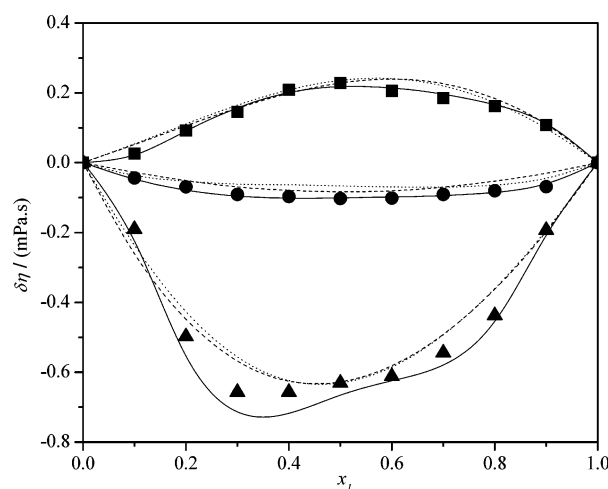


Figure 4. Viscosity deviations ($\delta\eta$) at 308.15 K: ■, cyclohexanone (1) + MAA (2); ●, cyclohexanone (1) + BzMA (2); ▲, cyclohexanone (1) + 2-HEMA (2); —, calculated from eq 5 with temperature-dependent parameters of eq 11; ---, calculated from eq 8 with temperature-dependent parameters of eq 12; ···, calculated from eq 9 with temperature-dependent parameters of eqs 13 and 14.

The coefficients of A_k and B_k were obtained by fitting the equations to the experimental values with a least-squares method. Tables 6 and 7 give the coefficients together with the average absolute deviation (AAD) of the calculated densities and viscosities, respectively.

The variations of V^E and $\delta\eta$ with the mole fraction of cyclohexanone at 308.15 K are presented in Figures 3 and 4, respectively. Figure 3 shows that the excess molar volumes are negative over the entire composition range for all the investigated binaries. It implies that volume contraction takes place when cyclohexanone mixes with the methacrylates. Figure 3 also shows that the V^E profile of cyclohexanone + benzyl methacrylate system is asymmetric and sigmoid with initial negative values followed by close to zero in the cyclohexanone rich region. Figure 4 illustrates that the viscosity deviations are positive in cyclohexanone + methacrylic acid, negative in cyclohexanone + 2-hydroxyethyl methacrylate, and nearly

Table 8. Correlated Results of McAllister's Models

mixture	T/K	three-body model			four-body model			
		ν_{12}	ν_{21}	AAD ^a /%	ν_{1112}	ν_{1122}	ν_{2221}	AAD ^a /%
cyclohexanone + MAA	298.15	2.3241	1.8904	0.8	2.2361	2.1268	1.6230	0.8
	308.15	1.9079	1.5294	0.9	1.8172	1.8476	1.3424	0.8
	318.15	1.6490	1.2837	0.9	1.5855	1.5065	1.1691	0.8
cyclohexanone + BzMA	298.15	1.9583	1.9856	0.4	1.9388	2.0473	1.9972	0.1
	308.15	1.6427	1.6643	0.4	1.6054	1.7243	1.6678	0.2
	318.15	1.4155	1.4121	0.4	1.3738	1.4950	1.3962	0.2
cyclohexanone + 2-HEMA	298.15	2.1943	3.2345	1.8	2.0726	2.7667	3.6117	1.8
	308.15	1.7442	2.6109	1.4	1.6220	2.3205	2.7495	1.2
	318.15	1.4953	2.0612	1.2	1.3721	1.9344	2.1462	1.0

$$^a \text{AAD}/\% = (100/n) \sum_{k=1}^n |\nu_k^{\text{cal}} - \nu_k^{\text{exp}}|/\nu_k^{\text{exp}}$$

Table 9. Temperature-Dependent Parameters in the Redlich–Kister Type Polynomial and the McAllister's Model

Cyclohexanone + MAA							
$A_0^0 = 3.9081$	$A_0^1 = -0.0163$	$B_0^0 = 7.3207$	$B_0^1 = -0.0209$	$\nu_{12}^0 = 12.2682$	$\nu_{12}^1 = -0.0334$	$\nu_{1112}^0 = 11.2907$	$\nu_{1112}^1 = -0.0306$
$A_1^0 = 0.4709$	$A_1^1 = -0.0029$	$B_1^0 = 1.2729$	$B_1^1 = -0.0038$	$\nu_{21}^0 = 10.7064$	$\nu_{21}^1 = -0.0298$	$\nu_{2221}^0 = 11.4378$	$\nu_{2221}^1 = -0.0313$
$A_2^0 = -0.1810$	$A_2^1 = -0.0002$	$B_2^0 = -1.9541$	$B_2^1 = 0.0057$			$\nu_{2221}^0 = 8.3878$	$\nu_{2221}^1 = -0.0227$
$A_3^0 = -1.3658$	$A_3^1 = 0.0027$	$B_3^0 = 2.7427$	$B_3^1 = -0.0063$				
AAD ^a /% = 0.01		AAD ^b /% = 0.4		AAD ^c /% = 1.1		AAD ^c /% = 1.0	
Cyclohexanone + BzMA							
$A_0^0 = -2.9766$	$A_0^1 = 0.0078$	$B_0^0 = -4.1566$	$B_0^1 = 0.0122$	$\nu_{12}^0 = 10.0321$	$\nu_{12}^1 = -0.0271$	$\nu_{1112}^0 = 9.9999$	$\nu_{1112}^1 = -0.0272$
$A_1^0 = 6.1509$	$A_1^1 = -0.0138$	$B_1^0 = 0.3274$	$B_1^1 = -0.0009$	$\nu_{21}^0 = 10.5309$	$\nu_{21}^1 = -0.0287$	$\nu_{2221}^0 = 11.0923$	$\nu_{2221}^1 = -0.0301$
$A_2^0 = -8.8759$	$A_2^1 = 0.0173$	$B_2^0 = -0.8140$	$B_2^1 = 0.0017$			$\nu_{2221}^0 = 10.4392$	$\nu_{2221}^1 = -0.0284$
$A_3^0 = -0.7216$	$A_3^1 = 0.0065$	$B_3^0 = -1.5321$	$B_3^1 = 0.0043$				
AAD ^a /% = 0.02		AAD ^b /% = 0.2		AAD ^c /% = 0.8		AAD ^c /% = 0.7	
Cyclohexanone + 2-HEMA							
$A_0^0 = 5.1237$	$A_0^1 = -0.0213$	$B_0^0 = -34.6476$	$B_0^1 = 0.1038$	$\nu_{12}^0 = 12.7750$	$\nu_{12}^1 = -0.0356$	$\nu_{1112}^0 = 12.5124$	$\nu_{1112}^1 = -0.0347$
$A_1^0 = 8.1886$	$A_1^1 = -0.0254$	$B_1^0 = 19.9695$	$B_1^1 = -0.0615$	$\nu_{21}^0 = 21.1121$	$\nu_{21}^1 = -0.0600$	$\nu_{2221}^0 = 14.3029$	$\nu_{2221}^1 = -0.0396$
$A_2^0 = 8.8030$	$A_2^1 = -0.0340$	$B_2^0 = -45.8300$	$B_2^1 = 0.1370$			$\nu_{2221}^0 = 26.1288$	$\nu_{2221}^1 = -0.0751$
$A_3^0 = -24.5267$	$A_3^1 = 0.0796$	$B_3^0 = -18.4291$	$B_3^1 = 0.0552$				
		$B_4^0 = 64.8194$	$B_4^1 = -0.1901$				
AAD ^a /% = 0.01		AAD ^b /% = 1.0		AAD ^c /% = 1.7		AAD ^c /% = 1.8	

$$^a \text{AAD}/\% = (100/n) \sum_{k=1}^n |\rho_k^{\text{cal}} - \rho_k^{\text{exp}}|/\rho_k^{\text{exp}}. \quad ^b \text{AAD}/\% = (100/n) \sum_{k=1}^n |\eta_k^{\text{cal}} - \eta_k^{\text{exp}}|/\eta_k^{\text{exp}}. \quad ^c \text{AAD}/\% = (100/n) \sum_{k=1}^n |\nu_k^{\text{cal}} - \nu_k^{\text{exp}}|/\nu_k^{\text{exp}}$$

zero in cyclohexanone + benzyl methacrylate, over the entire composition range.

McAllister's multi-body interaction model⁷ was widely used to correlate kinematic viscosity (ν) data. The three-body McAllister model was defined as

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln[x_1 + x_2(M_2/M_1)] + 3x_1^2 x_2 \ln[(2 + M_2/M_1)/3] + 3x_1 x_2^2 \ln[(1 + 2M_2/M_1)/3] + x_2^3 \ln(M_2/M_1) \quad (8)$$

and the four-body McAllister model was given by

$$\ln \nu = x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln \nu_{1112} + 6x_1^2 x_2^2 \ln \nu_{1122} + 4x_1 x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 - \ln[x_1 + x_2(M_2/M_1)] + 4x_1^3 x_2 \ln[(3 + M_2/M_1)/4] + 6x_1^2 x_2^2 \ln[(1 + M_2/M_1)/2] + 4x_1 x_2^3 \ln[(1 + 3M_2/M_1)/4] + x_2^4 \ln(M_2/M_1) \quad (9)$$

where ν_{12} , ν_{21} , ν_{1112} , ν_{1122} , and ν_{2221} are model parameters. The calculated results are presented in Table 8. As seen from Table 8, the values of AAD are approximately the same, regardless of whether the three-body or the four-body model was used.

The parameters in eqs 6 to 9 were also treated to be temperature dependent, as given by the following equations, respectively:

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

$$B_k = B_k^0 + B_k^1 T/K \quad (k = 0 \text{ to } 4) \quad (11)$$

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

$$\nu_{iii} = \nu_{iii}^0 + \nu_{iii}^1 T/K \quad (iii = 1112 \text{ or } 2221) \quad (13)$$

$$\nu_{1122} = \nu_{1122}^0 + \nu_{1122}^1 T/K \quad (14)$$

where A_k^0 , A_k^1 , B_k^0 , B_k^1 , ν_{ij}^0 , ν_{ij}^1 , ν_{iii}^0 , ν_{iii}^1 , ν_{1122}^0 , and ν_{1122}^1 are the undetermined parameters. The best-fit values of the parameters together with the average absolute deviations of the calculated densities, viscosities, and kinematic viscosities for cyclohexanone + MAA, cyclohexanone + BzMA, and cyclohexanone + 2-HEMA are given in Table 9. As seen from Tables 6 to 9, the values of AAD from the temperature-specific parameters in the Redlich–Kister type polynomial or the McAllister's multi-body interaction model are smaller than those from the

temperature-dependent parameters. These calculated results are also illustrated in Figures 1 to 4.

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