

Densities and Viscosities of Binary Mixtures of Propylene Glycol Monomethyl Ether Acetate with Methacrylic Acid, Benzyl Methacrylate, and 2-Hydroxyethyl Methacrylate between 298.15 K and 318.15 K

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Densities and viscosities of binary mixtures of propylene glycol monomethyl ether acetate with methacrylic acid, benzyl methacrylate, and 2-hydroxyethyl methacrylate were measured at temperatures from 298.15 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 also used to correlate the kinematic viscosities.

Introduction

The thermophysical properties of a binary mixture such as density and viscosity are very important from practical and theoretical points of view. These properties are useful in design of many types of process and transport equipment in chemical industries. Numerous experiments^{1–7} have been conducted to measure the densities and viscosities for a variety of liquid mixtures in the literature; however, reliable density and viscosity data over wide ranges of composition and temperature are still needed. Methacrylic acid (MAA), benzyl methacrylate (BzMA), 2-hydroxyethyl methacrylate (2-HEMA), and a safety solvent propylene glycol monomethyl ether acetate (PGMEA) are the key compounds in the manufacturing of the pigment dispersed color-resistant industries. The densities and viscosities were measured, in the present study, for the binary mixtures composed of the methacrylates with propylene glycol monomethyl ether acetate at 298.15 K, 308.15 K, and 318.15 K and over the entire composition range. No literature data were found at the 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.

Experimental Section

Methacrylic acid (mass fraction 99.5 %) and 2-hydroxyethyl methacrylate (mass fraction 98 %) were supplied by Acros Organics. Benzyl methacrylate (mass fraction 98 %) was purchased from Fisher Scientific. Propylene glycol monomethyl ether acetate (mass fraction 99 %) was made by the TEDIA Company. The purities of these substances were checked with gas chromatography. No impurity peaks were detected. All reagents were used without further purification. 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

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

T/K	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\eta/\text{mPa}\cdot\text{s}$	
	exptl	lit.	exptl	lit.
298.15	0.8056	0.80575 ^a 0.8060 ^b 0.80576 ^c	2.566	2.571 ^{a,b,c}
303.15	0.8021	0.8022 ^b 0.80201 ^c	2.265	2.271 ^{a,c} 2.263 ^b
308.15	0.7980	0.79821 ^c	1.998	2.000 ^b 1.981 ^c
313.15	0.7945	0.7946 ^b 0.79432 ^c	1.778	1.7734 ^b 1.692 ^c

^a Riddick et al.⁹ ^b TRC Data.¹⁰ ^c Nikam et al.¹¹

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

x_1	$\rho/(\text{g}\cdot\text{cm}^{-3})$ at T/K			$\eta/(\text{mPa}\cdot\text{s})$ 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	1.0040	0.9941	0.9839	1.423	1.202	1.033
0.2000	0.9990	0.9893	0.9794	1.506	1.301	1.105
0.3000	0.9939	0.9841	0.9739	1.560	1.345	1.124
0.3999	0.9891	0.9792	0.9689	1.585	1.348	1.125
0.5000	0.9848	0.9750	0.9648	1.569	1.310	1.100
0.6000	0.9801	0.9703	0.9599	1.499	1.240	1.049
0.7000	0.9758	0.9660	0.9555	1.411	1.168	0.988
0.7999	0.9708	0.9609	0.9505	1.301	1.081	0.929
0.9000	0.9658	0.9560	0.9456	1.175	0.985	0.844
1.0	0.9614	0.9514	0.9407	1.071	0.904	0.777

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

x_1	$\rho/(\text{g}\cdot\text{cm}^{-3})$ at T/K			$\eta/(\text{mPa}\cdot\text{s})$ 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	1.0290	1.0199	1.0109	2.245	1.840	1.544
0.2000	1.0231	1.0138	1.0046	2.123	1.743	1.465
0.3000	1.0170	1.0076	0.9982	1.991	1.638	1.377
0.4000	1.0106	1.0011	0.9915	1.857	1.530	1.289
0.4999	1.0038	0.9942	0.9844	1.694	1.399	1.183
0.6000	0.9966	0.9869	0.9769	1.559	1.286	1.086
0.7000	0.9890	0.9792	0.9690	1.434	1.189	1.007
0.8000	0.9807	0.9707	0.9603	1.327	1.105	0.937
0.9000	0.9719	0.9618	0.9512	1.232	1.028	0.874
1.0	0.9614	0.9514	0.9407	1.071	0.904	0.777

water⁸ 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

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

x_1	$\rho/(\text{g}\cdot\text{cm}^{-3})$ at T/K			$\eta/(\text{mPa}\cdot\text{s})$ 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	1.0564	1.0467	1.0373	4.764	3.519	2.710
0.2000	1.0457	1.0359	1.0261	3.840	2.900	2.273
0.3000	1.0349	1.0250	1.0151	3.222	2.477	1.969
0.4000	1.0241	1.0142	1.0041	2.719	2.123	1.709
0.5000	1.0135	1.0035	0.9933	2.257	1.790	1.461
0.6000	1.0030	0.9930	0.9826	1.922	1.550	1.284
0.7000	0.9926	0.9825	0.9720	1.651	1.341	1.117
0.8000	0.9822	0.9719	0.9613	1.389	1.147	0.969
0.8999	0.9718	0.9615	0.9508	1.234	1.052	0.889
1.0	0.9614	0.9514	0.9407	1.071	0.904	0.777

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

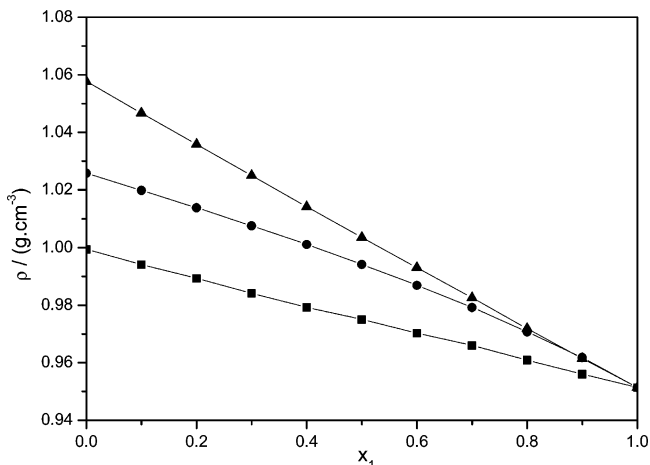
mixture	T/K	A_0	A_1	A_2	A_3	AAD ^a /%
PGMEA + MAA	298.15	-2.2703	-0.8196	0.4823	1.1381	0.01
	308.15	-2.4075	-0.7414	0.2460	0.9252	0.01
	318.15	-2.5490	-0.5549	-0.0365	0.3251	0.02
PGMEA + BzMA	298.15	-1.0907	-0.6353	-0.7367	-0.9597	0.01
	308.15	-1.0111	-0.7399	-0.4828	-0.6684	0.01
	318.15	-0.9728	-0.7443	-0.4130	-0.5802	0.01
PGMEA + 2-HEMA	298.15	-1.2561	-0.0320	-0.2660	0.0199	0.00
	308.15	-1.1919	-0.0441	0.2214	0.2959	0.00
	318.15	-1.1058	-0.1414	0.3119	0.4925	0.00

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

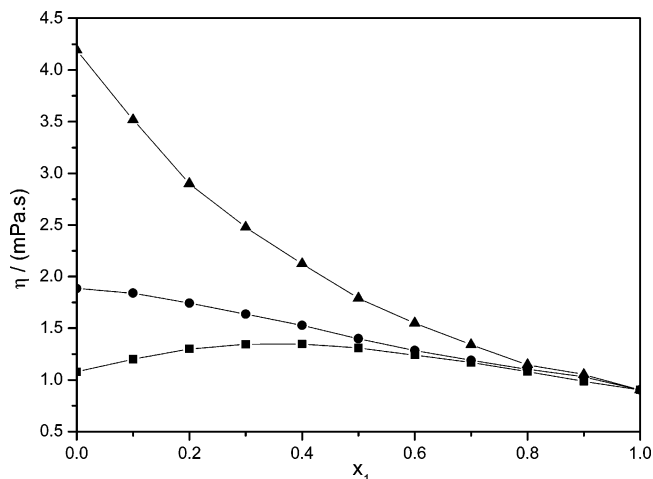
Table 6. Correlated Results of Viscosity Deviation ($\delta\eta$)

mixture	T/K	B_0	B_1	B_2	B_3	AAD ^a /%
PGMEA + MAA	298.15	1.5592	-0.3362	-0.2042	-0.4459	0.3
	308.15	1.2875	-0.6961	-0.2001	-0.2471	0.3
	318.15	0.9596	-0.4211	-0.1115	0.0679	0.4
PGMEA + BzMA	298.15	0.0155	-0.4653	0.8143	0.4278	0.4
	308.15	0.0040	-0.3997	0.6342	0.3395	0.3
	318.15	-0.0007	-0.3440	0.4947	0.2461	0.3
PGMEA + 2-HEMA	298.15	-4.6960	1.9777	-0.3190	-0.4209	0.8
	308.15	-3.0583	1.0460	0.0131	0.1755	0.9
	318.15	-2.0788	0.6532	0.0000	0.1016	0.8

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

**Figure 1.** Density ρ at 308.15 K: ■, PGMEA (1) + MAA (2); ●, PGMEA (1) + BzMA (2); ▲, PGMEA (1) + 2-HEMA (2).

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

**Figure 2.** Viscosity η at 308.15 K: ■, PGMEA (1) + MAA (2); ●, PGMEA (1) + BzMA (2); ▲, PGMEA (1) + 2-HEMA (2).

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 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 which the temperature was regulated to within ± 0.01 K. An electronic stopwatch 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 $\text{m}^2\cdot\text{s}^{-1}$ were obtained from the relation

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

where $k/\text{m}^2\cdot\text{s}^{-2}$ is the capillary constant of viscometer and t/s 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) in $\text{mPa}\cdot\text{s}$ were calculated by using the equation of $\eta = \rho\nu$. Since there are no available data for the methacrylates and propylene glycol monomethyl ether acetate, the measurements of 1-butanol were conducted to test the validity of the experimental 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 propylene glycol monomethyl ether acetate 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 with an increase of mole fraction of PGMEA (x_1) for each system. Figure 2 shows the variations of the absolute viscosities with the mole fraction of PGMEA for these three investigated systems at 308.15 K. In the MAA system, the viscosity increases with the mole fraction of PGMEA, reaching a weak maximum at

Table 7. 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 /%
PGMEA + MAA	298.15	1.5888	1.9568	0.5	1.4323	1.7975	1.7205	0.4
	308.15	1.2603	1.7685	0.2	1.1757	1.4881	1.5590	0.2
	318.15	1.0932	1.4261	0.3	1.0066	1.2691	1.2704	0.3
PGMEA + BzMA	298.15	1.4973	2.0749	1.0	1.5185	1.5483	2.2305	0.6
	308.15	1.2391	1.7315	1.0	1.2654	1.2906	1.8556	0.6
	318.15	1.0503	1.4759	0.9	1.0633	1.1255	1.5587	0.5
PGMEA + 2-HEMA	298.15	1.6673	2.8151	0.7	1.5486	2.0923	3.3745	0.7
	308.15	1.3786	2.2132	0.9	1.2507	1.7580	2.5530	0.9
	318.15	1.1549	1.8021	0.8	1.0582	1.4511	2.0469	0.8

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

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

PGMEA + MAA							
$A_0^0 = 1.8856$	$A_1^1 = -0.0139$	$B_0^0 = 10.5073$	$B_1^1 = -0.0300$	$\nu_{12}^0 = 9.0196$	$\nu_{12}^1 = -0.0250$	$\nu_{1112}^0 = 7.3156$	$\nu_{1112}^1 = -0.0198$
$A_1^0 = -4.7977$	$A_2^1 = 0.0133$	$B_1^0 = 0.8248$	$B_2^1 = -0.0042$	$\nu_{21}^0 = 9.9278$	$\nu_{21}^1 = -0.0267$	$\nu_{1122}^0 = 9.4230$	$\nu_{1122}^1 = -0.0256$
$A_2^0 = 8.2239$	$A_3^1 = -0.0259$	$B_2^0 = -1.6002$	$B_3^1 = 0.0046$	AAD ^c /% = 0.6		$\nu_{2221}^0 = 8.9240$	$\nu_{2221}^1 = -0.0240$
$A_3^0 = 13.3425$	$A_4^1 = -0.0407$	$B_3^0 = -7.9570$	$B_4^1 = 0.0257$			AAD ^c /% = 0.6	
AAD ^a /% = 0.01		AAD ^b /% = 0.5					
PGMEA + BzMA							
$A_0^0 = -2.8410$	$A_1^1 = 0.0059$	$B_0^0 = 0.2561$	$B_1^1 = -0.0008$	$\nu_{12}^0 = 8.1039$	$\nu_{12}^1 = -0.0222$	$\nu_{1112}^0 = 8.2522$	$\nu_{1112}^1 = -0.0225$
$A_1^0 = 0.9782$	$A_2^1 = -0.0055$	$B_1^0 = -2.2794$	$B_2^1 = 0.0061$	$\nu_{21}^0 = 11.3168$	$\nu_{21}^1 = -0.0310$	$\nu_{1122}^0 = 8.0091$	$\nu_{1122}^1 = -0.0218$
$A_2^0 = -5.5301$	$A_3^1 = 0.0162$	$B_2^0 = 5.5719$	$B_3^1 = 0.0160$	AAD ^c /% = 1.2		$\nu_{2221}^0 = 12.1203$	$\nu_{2221}^1 = -0.0332$
$A_3^0 = -6.5882$	$A_4^1 = 0.0190$	$B_3^0 = 3.1513$	$B_4^1 = -0.0091$			AAD ^c /% = 0.9	
AAD ^a /% = 0.01		AAD ^b /% = 0.4					
PGMEA + 2-HEMA							
$A_0^0 = -3.6514$	$A_1^1 = 0.0080$	$B_0^0 = -43.6028$	$B_1^1 = 0.1309$	$\nu_{12}^0 = 10.0410$	$\nu_{12}^1 = -0.0280$	$\nu_{1112}^0 = 8.5320$	$\nu_{1112}^1 = -0.0235$
$A_1^0 = 1.6172$	$A_2^1 = -0.0055$	$B_1^0 = 21.7131$	$B_2^1 = -0.0665$	$\nu_{21}^0 = 17.7913$	$\nu_{21}^1 = -0.0503$	$\nu_{1122}^0 = 12.6324$	$\nu_{1122}^1 = -0.0351$
$A_2^0 = -8.8219$	$A_3^1 = 0.2889$	$B_2^0 = -5.0166$	$B_3^1 = 0.0159$	AAD ^c /% = 1.3		$\nu_{2221}^0 = 21.5983$	$\nu_{2221}^1 = -0.0616$
$A_3^0 = -7.0157$	$A_4^1 = 0.0236$	$B_3^0 = -8.2569$	$B_4^1 = 0.0266$			AAD ^c /% = 1.3	
AAD ^a /% = 0.00		AAD ^b /% = 1.7					

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

about $x_1 = 0.4$, and then decreasing slightly to the value of pure PGMEA. However, the viscosities of the other two systems decrease monotonically with the mole fraction of PGMEA. Excess volumes (V^E) and viscosity deviations ($\delta\eta$) were calculated from the experimental results by the following equations, respectively:

$$V^E = V_M - (x_1 V_1 + x_2 V_2) \quad (2)$$

$$\delta\eta = \eta_M - (x_1 \eta_1 + x_2 \eta_2) \quad (3)$$

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:¹²

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

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

The coefficients of A_k and B_k were obtained by fitting the equations to the experimental values with a least-squares method. Tables 5 and 6 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 PGMEA at 308.15 K are presented in Figures 3 and 4, respectively. Figure 3 shows that the excess molar volumes

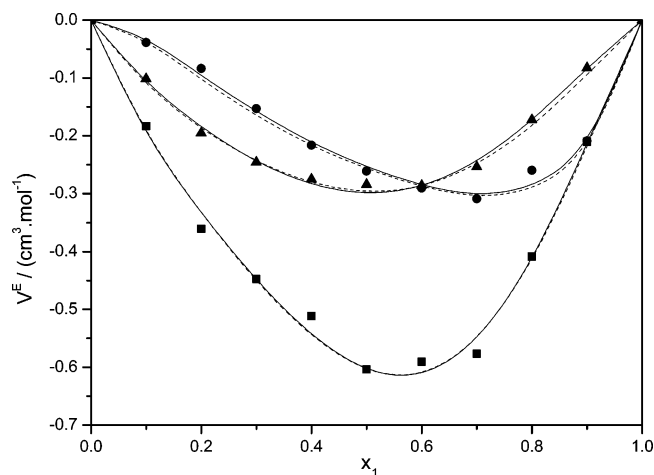


Figure 3. Excess volumes (V^E) at 308.15 K: ■, PGMEA (1) + MAA (2); ●, PGMEA (1) + BzMA (2); ▲, PGMEA (1) + 2-HEMA (2); —, calculated from eq 4 with temperature-specific parameters; --, calculated from eq 4 with temperature-dependent parameters of eq 8.

are negative over the entire composition range for all the investigated binaries. It implies that volume contraction takes place when PGMEA mixes with the methacrylates. The volume contraction in PGMEA + MAA is slightly greater than those in PGMEA + BzMA and PGMEA + 2-HEMA. Figure 4 illustrates that the viscosity deviations are positive in PGMEA + MAA, negative in PGMEA + 2-HEMA, and nearly zero in PGMEA + BzMA, over the entire composition range.

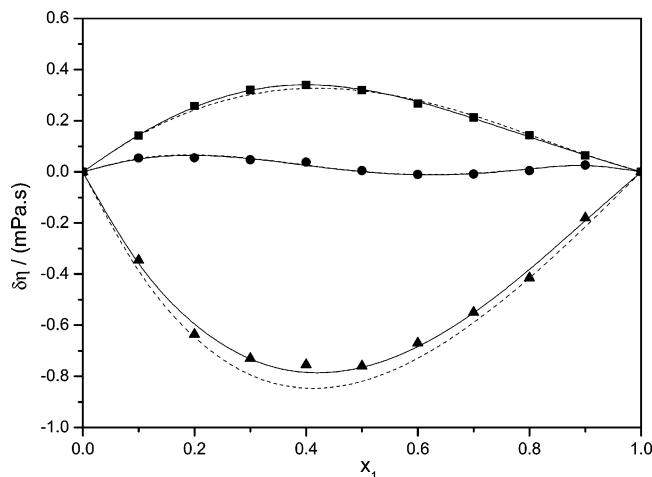


Figure 4. Viscosity deviations ($\delta\eta$) at 308.15 K: ■, PGMEA (1) + MAA (2); ●, PGMEA (1) + BzMA (2); ▲, PGMEA (1) + 2-HEMA (2); —, calculated from eq 5 with temperature-specific parameters; - -, calculated from eq 5 with temperature-dependent parameters of eq 9.

McAllister's multi-body interaction model¹³ was widely used to correlate kinematic viscosity (ν) data. The three-body McAllister model was defined as

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2x_2 \ln \nu_{12} + 3x_1x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln[x_1 + x_2(M_2/M_1)] + 3x_1^2x_2 \ln[(2 + M_2/M_1)/3] + 3x_1x_2^2 \ln[(1 + 2M_2/M_1)/3] + x_2^3 \ln(M_2/M_1) \quad (6)$$

and the four-body McAllister model was given by

$$\ln \nu = x_1^4 \ln \nu_1 + 4x_1^3x_2 \ln \nu_{1112} + 6x_1^2x_2^2 \ln \nu_{1122} + 4x_1x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 - \ln[x_1 + x_2(M_2/M_1)] + 4x_1^3x_2 \ln[(3 + M_2/M_1)/4] + 6x_1^2x_2^2 \ln[(1 + M_2/M_1)/2] + 4x_1x_2^3 \ln[(1 + 3M_2/M_1)/4] + x_2^4 \ln(M_2/M_1) \quad (7)$$

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.

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

$$A_k = A_k^0 + A_k^1 T/K \quad (k = 0 \text{ to } 3) \quad (8)$$

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

$$\nu_{ij} = \nu_{ij}^0 + \nu_{ij}^1 T/K \quad (ij = 12 \text{ or } 21) \quad (10)$$

$$\nu_{iii} = \nu_{iii}^0 + \nu_{iii}^1 T/K \quad (iii = 1112 \text{ or } 2221) \quad (11)$$

$$\nu_{1122} = \nu_{1122}^0 + \nu_{1122}^1 T/K \quad (12)$$

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 PGMEA + MAA, PGMEA + BzMA, and PGMEA + 2-HEMA are given in Table 8. As seen from Tables 5 to 8, the values of AAD are approximately within the experimental uncertainty, regardless of the parameters treating as temperature-specific or temperature-dependent in the Redlich–Kister type polynomial and the McAllister's multi-body interaction model.

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