Excess Molar Volume and Viscosity Deviation for the Methanol + Methyl Methacrylate Binary System at $T = (283.15 \text{ to } 333.15)$ K

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Density and viscosity for the methanol + methyl methacrylate (MMA) binary system over the whole concentration range in the temperature range from (383.15 to 333.15) K were measured. From the all experimental data, the excess molar volumes V^E and viscosity deviations $\delta\eta$ were calculated. Excess molar volumes and viscosity deviations were correlated by the Redlich−Kister type equations. Optimally fitted parameters are presented, and the correlation results are in satisfactory agreement with the experimental data.

Introduction

Methyl methacrylate (MMA), which is an important monomer and industrial material is widely used for producing acrylic plastics such as polymethyl methacrylate (PMMA), etc. PMMA with good transparency and weather resistance is used in many fields, and its demand increases year after year. Sharp growth is expected in the attention to industrial production, thermodynamics, and kinetics in the industrial process of PMMA and MMA.[1,2]

The design of an efficient industrial separation process and transport equipment requires a large knowledge of the thermodynamic properties such as density and viscosity of the mixture involved. The excess molar volume V^E and viscosity deviations $\delta\eta$ are often used to describe the intermolecular forces in mixtures, helping us to understand their real behavior and develop models. As we know, detailed thermodynamic data for industrial design cannot be found, and only a few experimental thermodynamic studies such as excess volume at 298.15 K on the binary mixture for methanol and ethanol + MMA are reported in the literature.[3]

In our work, the densities and viscosities of the methanol + MMA binary mixture were measured from (283.15 to 333.15) K. From the experimental results, the excess molar volumes and the viscosity deviations were calculated and correlated by the Redlich−Kister polynomial equation. The effect of temperature on the physical properties (density and viscosity) was analyzed, and the interactions between molecules were discussed in terms of excess molar volume V^E of the binary mixture.

Experimental Section

Materials. Methanol was supplied by Fisher Chemicals Fair Lawn, with mass fraction > 99.9 %. Methyl methacrylate with a mass fraction > 99.0 % was obtained from Yili Fine Chemical Co., Ltd., Beijing, China. The purities of these substances were checked with gas chromatography before use. All reagents were used without further purification.

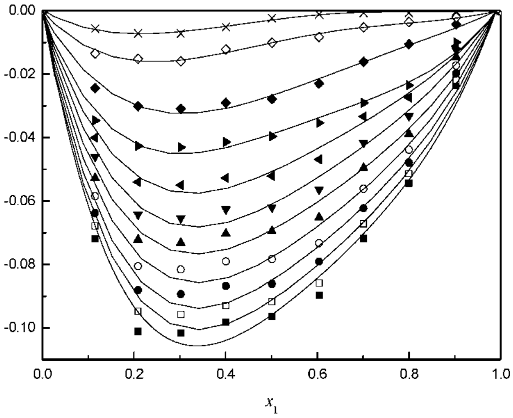

Figure 1. Excess molar volume V^E vs mole fraction x of methanol for methanol (1) + MMA (2): ■, 283.15K; □, 288.15 K; ●, 293.15 K; ○, 298.15 K; ▲, 303.15 K; ▼, 308.15 K; solid arrow pointing left, 313.15 K; solid arrow pointing right, 318.15 K; ♦, 323.15 K; ◇, 328.15 K; ×, 333.15 K. The symbols represent experimental values, and the solid curves represent the values calculated from eq 3.

Apparatus and Procedure. These sample mixtures were prepared by mass in hermetically sealed glass vials. To minimize the evaporation during the sample preparation, the heavier component was charged first. The mass measurements were made by an electronic analytical balance (Sartorius scientific Instrument Co. Ltd., BS124S), accurate to ± 0.0001 g with an uncertainty of ± 0.0001 in mole fraction. The mixtures of the various compositions were prepared fresh, and density and viscosity were measured at the same time.

Densities of the pure liquids and their mixtures were measured by an Anton Paar DMA 5000 densimeter with an accuracy of $\pm\ 0.000001\ \text{g}\cdot\text{cm}^{-3}$. The accuracy of the temperature during the measurements is ± 0.001 K, and the repeatability in the densities for the pure liquids and prepared binary mixtures have been found to be better than $\pm\ 5\cdot10^{-6}\ \text{g}\cdot\text{cm}^{-3}$.

Viscosities of the pure liquids and their mixtures were measured by the Anton Paar AMVn Measuring Assembly. The measuring system is a combination of a capillary

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10.1021/je800164w CCC: $40.75

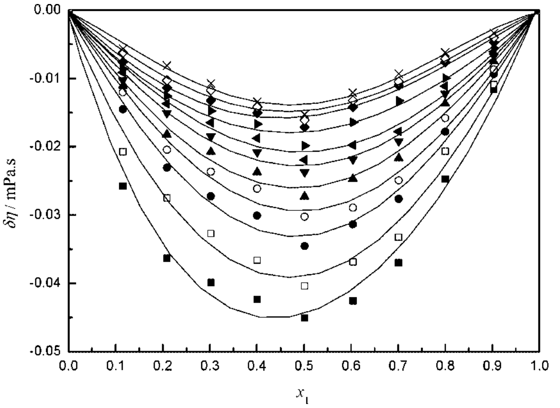

Figure 2. Viscosity deviation $\delta\eta$ vs mole fraction x of methanol for methanol (1) + MMA (2): ■, 283.15 K; □, 288.15 K; ●, 293.15 K; ○, 298.15 K; ▲, 303.15 K; ▼, 308.15 K; solid arrow pointing left, 313.15 K; solid arrow pointing right, 318.15 K; ♦, 323.15 K; ◇, 328.15 K; ×, 333.15 K. The symbols represent experimental values, and the solid curves represent the values calculated from eq 3.

Table 1. Densities ρ and Viscosities η of Methanol and MMA at Different Temperatures

		$\rho/(\mathrm{g \cdot cm^{-3}})$		$\eta/(\mathrm{mPa \cdot s})$	
compounds	T/K	exptl	lit.	exptl	lit.
methanol	283.15	0.800605		0.6781	
	288.15	0.795915		0.6315	
	293.15	0.791218		0.5884	
	298.15	0.786507	0.7864[11] 0.78637[8] 0.78664[12] 0.78654[10] 0.7867[9] 0.78663[7]	0.5509	0.546[11] 0.5513[8] 0.543[12] 0.553[13]
	303.15	0.781778	0.7820[9] 0.7819[13]	0.5154	0.50[13] 0.510[14]
	308.15	0.777028	0.7772[13] 0.7771[16] 0.77735[17]	0.4849	0.46[13] 0.469[15] 0.477[17]
	313.15	0.772238	0.7727[9] 0.7725[13] 0.7723[11]	0.4576	0.44[13] 0.446[11] 0.456[14]
	318.15	0.767326	0.7675[13] 0.76813[17]	0.4324	0.41[13] 0.422[17]
	323.15	0.762567	0.7627[13]	0.4093	0.40[13] 0.403[14]
	328.15	0.757671	0.7578[11]	0.3903	0.369[11]
	333.15	0.752801		0.3724	
MMA	283.15	0.954802		0.6808	
	288.15	0.949097		0.6337	
	293.15	0.943365		0.5899	
	298.15	0.937615	0.93780[3] 0.93763[4] 0.93766[5]	0.5540	0.584[2]
	303.15	0.931837	0.93174[4] 0.93174[6]	0.5248	
	308.15	0.926032	0.92574[4] 0.9257[6]	0.4956	0.492[2]
	313.15	0.920185	0.92026[4]	0.4719	
	318.15	0.914317		0.4497	
	323.15	0.908405		0.4294	
	328.15	0.902468		0.4100	
	333.15	0.896479		0.3938	

(diameter, 1.6 mm) and a diametrically matching ball (diameter, 1.5 mm). The accuracy of the temperature during the viscometer is ± 0.01 K, and the uncertainty of viscosity measurements was estimated to be within ± 0.0001. Afterward, the capillary was cleaned and dried with compressed air, and the viscometer was calibrated with double-distilled water before measuring.

The density and viscosity were measured six times for every sample, and the results were average values. The results were compared between the experimental and the literature values in Table 1. It shows that our experimental values agree with literature values.

Results and Discussion

Experimental density ρ and viscosity η results for the binary system of methanol and MMA over the temperature range T = (283.15 to 333.15) K are reported in Table 2. Excess volumes V^E and viscosity deviations $\delta\eta$ were calculated from the experimental results by the following equations, respectively

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho_M} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2} \tag{1}$$

$$\delta\eta = \eta_M - (x_1 \eta_1 + x_2 \eta_2) \tag{2}$$

where x_i, ρ_i, η_i, and M_i are the mole fraction, density, viscosity, and molar masses of the pure component i, respectively, and the subscript M represents mixture properties. The data of excess molar volume and viscosity deviation were shown in Table 2. The excess volumes V^E and viscosity deviations $\delta\eta$ were correlated by the Redlich−Kister polynomial equation[18–20]

$$Y = x_1 x_2 \sum_{k=0}^{3} A_k (x_1 - x_2)^k \tag{3}$$

where $Y = V^E$ or $\delta\eta$ and the coefficients of A_k are parameters that were obtained by fitting the equations to the experimental values with a least-squares method. The parameters were also treated to be temperature dependent, usually assuming a linear dependence with temperature, as given by the following equation[20]

$$A_k = A_{k0} + A_{k1} T \quad (k = 0 \text{ to } 3) \tag{4}$$

$$B_k = B_{k0} + B_{k1} T \quad (k = 0 \text{ to } 3) \tag{5}$$

where A_{k0}, A_{k1}, B_{k0}, and B_{k1} are the adjustable parameters for the calculated excess volumes and viscosity deviations, which are given in Table 3.

The correlated results for excess volumes and viscosity deviations were also given in Table 3, in which the tabulated standard deviation σ was defined as

$$\sigma = \left[\frac{\sum (Y_{\mathrm{exptl}} - Y_{\mathrm{calcd}})^2}{n - p} \right]^{\frac{1}{2}} \tag{6}$$

where n is the number of data points and p is the number of coefficients. The subscripts exptl and calcd, denoted the experimental and the calculated value, respectively.

As seen from Table 3, the standard deviations of the calculated excess volumes and viscosity deviations approach very well, whether or not considering the effect of temperature variation on the parameters in the Redlich−Kister polynomial equation, in which σ and σ_T are the standard deviations of the calculated excess volumes and viscosity deviations, the former without considering the temperature-dependence and the latter considering the temperature-dependence.

The excess volume V^E and the viscosity deviation $\delta\eta$ of the mixture versus the mole fraction of methanol at (283.15

Table 2. Experimental Densities ρ, Viscosities η, Excess Molar Volumes V^E, and Viscosity Deviations $\delta\eta$ of Methanol (1) + MMA (2)

x_1	T/K = 283.15	T/K = 288.15	T/K = 293.15	T/K = 298.15	T/K = 303.15	T/K = 308.15	T/K = 313.15	T/K = 318.15	T/K = 323.15	T/K = 328.15	T/K = 333.15
ρ/(g·cm^{-3})											
0.1152	0.948203	0.942499	0.936771	0.931013	0.925226	0.919404	0.913549	0.907673	0.901723	0.895739	0.889735
0.2092	0.941705	0.936017	0.930303	0.924564	0.918792	0.912999	0.907145	0.901251	0.895325	0.889344	0.883385
0.3024	0.934026	0.928391	0.922727	0.917034	0.911309	0.905566	0.899758	0.893899	0.888029	0.882091	0.876178
0.4008	0.924596	0.919024	0.913423	0.907788	0.902119	0.896436	0.890692	0.884889	0.879081	0.873178	0.867346
0.5011	0.913289	0.907797	0.902273	0.896707	0.891106	0.885499	0.879823	0.874068	0.868343	0.862493	0.856732
0.6041	0.899281	0.893887	0.888436	0.882983	0.877479	0.871944	0.866361	0.860693	0.855055	0.849321	0.843663
0.7013	0.882998	0.877696	0.872375	0.867022	0.861643	0.856222	0.850759	0.845279	0.839741	0.834153	0.828639
0.8003	0.862450	0.857304	0.852138	0.846948	0.841725	0.836468	0.831171	0.825819	0.820417	0.815009	0.809661
0.9033	0.834809	0.829862	0.824900	0.819918	0.814913	0.809896	0.804854	0.799711	0.794611	0.789453	0.784311
η/(mPa·s)											
0.1152	0.6547	0.6127	0.5752	0.5416	0.5126	0.4841	0.4611	0.4391	0.4195	0.4015	0.3857
0.2092	0.6439	0.6057	0.5665	0.5329	0.5046	0.4783	0.4552	0.4335	0.4138	0.3956	0.3812
0.3024	0.6401	0.6003	0.5622	0.5294	0.5012	0.4739	0.4511	0.4297	0.4101	0.3923	0.3766
0.4008	0.6374	0.5962	0.5592	0.5266	0.4973	0.4705	0.4474	0.4261	0.4063	0.3879	0.3718
0.5011	0.6344	0.5922	0.5546	0.5222	0.4928	0.4665	0.4428	0.4212	0.4022	0.384	0.3678
0.6041	0.6366	0.5955	0.5576	0.5232	0.4944	0.4673	0.4435	0.4228	0.4031	0.3849	0.3688
0.7013	0.6419	0.5989	0.5612	0.5269	0.4965	0.4689	0.4441	0.4243	0.4043	0.3857	0.3695
0.8003	0.6539	0.6113	0.5709	0.5357	0.5036	0.4748	0.4493	0.4259	0.4057	0.3871	0.3705
0.9033	0.6667	0.6208	0.5792	0.5425	0.5088	0.4792	0.4529	0.4285	0.4065	0.3882	0.3711
V^E/(cm^3·mol^{-1})											
0.1152	−0.0719	−0.0678	−0.0638	−0.0584	−0.0527	−0.0460	−0.0400	−0.0346	−0.0243	−0.0135	−0.0056
0.2092	−0.1012	−0.0948	−0.0881	−0.0806	−0.0722	−0.0642	−0.0540	−0.0426	−0.0300	−0.0151	−0.0071
0.3024	−0.1016	−0.0958	−0.0893	−0.0816	−0.0732	−0.0653	−0.0550	−0.0431	−0.0309	−0.0159	−0.0072
0.4008	−0.0983	−0.0929	−0.0868	−0.0791	−0.0704	−0.0625	−0.0527	−0.0414	−0.0290	−0.0122	−0.0051
0.5011	−0.0964	−0.0918	−0.0862	−0.0783	−0.0695	−0.0620	−0.0521	−0.0396	−0.0278	−0.0100	−0.0023
0.6041	−0.0897	−0.0859	−0.0791	−0.0732	−0.0651	−0.0564	−0.0469	−0.0353	−0.0230	−0.0082	−0.0011
0.7013	−0.0718	−0.0672	−0.0622	−0.0561	−0.0495	−0.0416	−0.0334	−0.0291	−0.0161	−0.0052	−0.0006
0.8003	−0.0543	−0.0513	−0.0479	−0.0438	−0.0389	−0.0331	−0.0273	−0.0235	−0.0105	−0.0035	−0.0006
0.9033	−0.0237	−0.0218	−0.0197	−0.0172	−0.0145	−0.0124	−0.0111	−0.0099	−0.0042	−0.0020	−0.0001
$\delta\eta$/mPa·s											
0.1152	−0.0258	−0.0207	−0.0145	−0.0120	−0.0111	−0.0103	−0.0092	−0.0086	−0.0076	−0.0062	−0.0056
0.2092	−0.0363	−0.0275	−0.0231	−0.0205	−0.0182	−0.0151	−0.0137	−0.0126	−0.0114	−0.0103	−0.0081
0.3024	−0.0399	−0.0327	−0.0272	−0.0237	−0.0208	−0.0185	−0.0165	−0.0148	−0.0132	−0.0117	−0.0107
0.4008	−0.0423	−0.0366	−0.0301	−0.0262	−0.0237	−0.0208	−0.0188	−0.0167	−0.0150	−0.0142	−0.0134
0.5011	−0.0450	−0.0404	−0.0345	−0.0302	−0.0273	−0.0237	−0.0219	−0.0198	−0.0171	−0.0161	−0.0153
0.6041	−0.0426	−0.0369	−0.0314	−0.0289	−0.0247	−0.0218	−0.0198	−0.0164	−0.0142	−0.0132	−0.0121
0.7013	−0.0370	−0.0333	−0.0276	−0.0249	−0.0217	−0.0192	−0.0178	−0.0133	−0.0110	−0.0105	−0.0093
0.8003	−0.0247	−0.0206	−0.0178	−0.0158	−0.0137	−0.0122	−0.0112	−0.0100	−0.0076	−0.0071	−0.0062
0.9033	−0.0117	−0.0109	−0.0093	−0.0087	−0.0075	−0.0067	−0.0061	−0.0056	−0.0047	−0.0040	−0.0034

Table 3. Temperature-Specific and Temperature-Dependent Parameters in the Redlich−Kister Equation for V^E and $\delta\eta$ of the Methanol (1) + MMA (2) System

properties	T/K	A_0	A_1	A_2	A_3	σ	σ_T
V^E/cm^3·mol^{-1}	283.15	−0.3887	0.1326	−0.1955	0.2495	0.0023	0.0085
	288.15	−0.3687	0.1204	−0.1755	0.2468	0.0023	0.0048
	293.15	−0.3433	0.1108	−0.1634	0.2436	0.0020	0.0027
	298.15	−0.3140	0.0979	−0.1445	0.2373	0.0021	0.0030
	303.15	−0.2796	0.0868	−0.1287	0.2274	0.0019	0.0035
	308.15	−0.2466	0.0975	−0.1041	0.1661	0.0019	0.0043
	313.15	−0.2035	0.0904	−0.1060	0.1318	0.0016	0.0040
	318.15	−0.1557	0.0050	−0.1263	0.1547	0.0007	0.0032
	323.15	−0.1053	0.0703	−0.0645	0.0845	0.0007	0.0023
	328.15	−0.0396	0.0519	−0.0617	0.0308	0.0005	0.0107
	333.15	−0.0124	0.0376	−0.0277	−0.0044	0.0004	0.0053
$\delta\eta$/mPa·s	283.15	−0.1776	0.0040	−0.0319	0.1211	0.0006	0.0042
	288.15	−0.1536	−0.0096	−0.0140	0.0986	0.0011	0.0013
	293.15	−0.1317	−0.0015	0.0108	0.0421	0.0010	0.0028
	298.15	−0.1173	−0.0041	0.0130	0.0306	0.0010	0.0034
	303.15	−0.1033	−0.0003	0.0097	0.0278	0.0009	0.0035
	308.15	−0.0899	−0.0056	0.0039	0.0349	0.0006	0.0035
	313.15	−0.0821	−0.0071	0.0055	0.0340	0.0007	0.0027
	318.15	−0.0700	0.0069	−0.0039	0.0106	0.0009	0.0023
	323.15	−0.0609	0.0160	−0.0016	−0.0039	0.0008	0.0017
	328.15	−0.0579	0.0119	0.0094	−0.0028	0.0007	0.0009
	333.15	−0.0534	0.0090	0.0152	0.0009	0.0008	0.0021
$A_{00} = -2.6352$	$A_{01} = 0.0078$	$A_{10} = 0.6036$	$A_{11} = -0.0017$	$A_{20} = -1.0876$	$A_{21} = 0.0032$	$A_{30} = 1.8241$	$A_{31} = -0.0054$
$B_{00} = -0.8461$	$B_{01} = 0.0024$	$B_{10} = -0.1539$	$B_{11} = 0.0005$	$B_{20} = -0.1408$	$B_{21} = 0.0005$	$B_{30} = 0.6960$	$B_{31} = -0.0021$

to 333.15) K were plotted in Figures 1 and 2, respectively. In Figure 1, it shows that the excess molar volumes V^E are asymmetric and negative over the entire composition range and that the excess volume V^E values decrease with increasing temperature. Comparing our experimental data with the data of Zhang et al.,[3] the excess volume V^E values are both negative for methanol + MMA at 298.15 K. The experimentally derived molar excess volume was correlated satisfactorily by the Redlich−Kister equation, which represents a further contribution to results from works reported previously in the literature.

The above results can be discussed in terms of several effects that may be simply divided into dispersion forces and hydrogen bond contributions. The negative V^E values for binary mixtures may be attributed to a hydrogen bond between methanol and MMA molecules. The decrease of V^E values with the increase of the temperature may be attributed to dispersion forces, which implies that volume expansion takes place. However, the hydrogen bonds are stronger than dispersion forces; that is, the hydrogen bond was slightly dominate, and the excess molar volumes V^E are still negative. In Figure 2, it illustrates that the viscosity deviations are negative over the entire composition range, and the minimum existed at $x_1 = 0.5$ within the whole region of experimental temperatures. The viscosity derivations $\delta\eta$ decreased slightly with an increase in temperature from (283.15 to 333.15) K.

Conclusions

A series of new experimental data of density and viscosity for the mixture of methanol + MMA were measured over the entire range of compositions from (283.15 to 333.15) K. Density and viscosity were found to decrease with increasing temperature. Viscosity was more sensitive than density to changes in temperature or composition. Reasonable correlations were achieved by fitting the excess molar volumes and the viscosity deviations to the Redlich−Kister polynomial equation. The correlation procedure included the functionality with both temperature and composition. Estimated coefficients and standard deviation values are also presented. The results show the parameters, and taking no account of temperature yielded better calculated results than considering temperature did for the investigated systems.

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Received for review March 7, 2008. Accepted May 5, 2008. This work was supported financially by the Program of Instrument and Equipment Development of Chinese Academy of Sciences (No. Y2005013), the National Natural Science Foundation of China (Grant No. 20436050), and the National 863 Program of China (Grant No. 2006AA06Z376).

JE800164W