# Densities and Viscosities of Diethyl Carbonate + Toluene, + Methanol, and + 2-Propanol from (293.15 to 363.15) K

# Changsheng Yang,\* Hexi Lai, Zanguang Liu, and Peisheng Ma

Key Laboratory for Green Chemical Technology of State Education Ministry, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, People's Republic of China

Densities and viscosities for diethyl carbonate (DEC) with toluene, methanol, and 2-propanol were determined over several temperatures and at atmospheric pressure. The measurements were carried out over the whole mole fraction, using a vibrating-tube densimeter and an Ubbelohde viscometer. Density and viscosity measurements were used to compute the excess molar volumes,  $V^{\rm E}$ , and viscosity deviations,  $\Delta\eta$ . The excess molar volumes,  $V^{\rm E}$ , and viscosity deviations,  $\Delta\eta$ , have been fitted to Redlich–Kister equation.

### Introduction

Studies on viscosity and density of binary mixtures along with other thermodynamic properties are being increasingly used as tools for the investigation of the properties of pure components and the nature of intermolecular interactions between the liquid mixture constituents. The organic carbonates are attracting an increasing interest due to these chemicals having an extensive variety of applications in the lithium battery technology; for the synthesis of polycarbonates, pharmaceuticals, and agrochemicals; as extractive solvents and solvents for natural and synthetic resins and polymers; and as fuel additives<sup>1</sup>. Also, organic carbonates have been proposed as lubricants of new refrigerants for stationary air conditioners because of their thermal stability, miscibility with HFCs, and lubricity.

This paper is part of our systematic program of research on the measurement of physical and transport properties of binary liquid mixtures containing organic carbonates. In this work, we present density and viscosity data for the binary systems diethyl carbonate + toluene, + methanol, and + 2-propanol from (293.15 to 363.15) K. From the densities and viscosities, excess molar volumes and viscosity deviations are calculated. The Redlich-Kister equation was used to correlate the experimental excess molar volumes and viscosity deviations.

A survey of the literature shows that Lopez et al.<sup>4</sup> measured densities for diethyl carbonate + toluene at (278.15 to 323.15) K. Francesconi et al.<sup>2</sup> measured excess molar enthalpies and densities of DEC in binary mixtures with seven *n*-alkanols at 298.15 K, while Rodriguez et al.<sup>3,5</sup> determined densities and viscosities of DEC with alcohols at 293.15 K, 298.15 K, 303.15 K, and 313.15 K.

## **Experimental Section**

*Materials.* All chemicals used in this study were supplied by Tianjin Reagent Co. except DEC, which was supplied by Shanghai Chemical Reagent Co. DEC was purified by distillation. Other liquids were subjected to no further purification. All were dried over 0.4 nm molecular sieves and particularly degassed by ultrasound prior to their experimental use. The mass fraction purities tested by gas chromatography were as fol-

\* Corresponding author. E-mail: tjuyangchsh@yahoo.com.cn. Fax: +022-27403389. Telephone: +022-27890907.

lows: DEC (> 0.995), toluene (> 0.998), methanol (> 0.998), and 2-propanol (> 0.993). In Table 1, we compare the densities and viscosities determined in this study and the literature data.

Apparatus and Procedure. The densities of the pure components and their mixtures were measured with a high precision vibrating-tube digital density meter (Density/Specific Gravity Meter DA 505, KEM, Japan) whose measurement cell temperature was controlled automatically within  $\pm$  0.01 K of the selected value. Before each series of measurements, the instrument was calibrated at atmospheric pressure with doubledistilled water and dry air. Densities of both water and dry air at the various working temperatures were given by the manufacturer in the instruction manual. The uncertainty in density measurements was  $\pm$  5 × 10<sup>-5</sup> g·cm<sup>-3</sup>. Density measurements were reproducible to  $\pm$  3 × 10<sup>-5</sup> g·cm<sup>-3</sup>.

The liquid mixtures were prepared by mass using a BP210s balance accurate to within  $\pm$  0.01 mg. The average uncertainty in the mole fraction of the mixtures was estimated to be less than  $\pm$  0.0001. The molar excess volumes were calculated from composition-density data with an uncertainty better than  $\pm$  0.002 cm<sup>3</sup>·mol<sup>-1</sup>. All molar quantities were based on the IUPAC relative atomic mass table.

The viscosities of pure liquids and the mixtures were measured at atmospheric pressure and at different temperatures using several Ubbelohde suspended-level viscometers. The viscometer was immersed in a well-stirred water bath (Lauda, Germany) with temperature control to within  $\pm$  0.01 K. An electronic digital stopwatch with a readability of  $\pm$  0.01 s was used for flow time measurement. Experiments are repeated a minimum of four times at each temperature for all compositions, and the results were averaged. The viscosity  $\eta$  of the liquid was then calculated from the following relationship:

$$\nu = \frac{\eta}{\rho} = k(t - \theta) \tag{1}$$

where *t* is the flow time;  $\nu$  is the kinematic viscosity; and *k* and  $\theta$  are the viscometer constant and the Hagenbach correction factor, respectively.

The calibration of the viscometer was carried out with doubledistilled water and twice-distilled benzene. During the heating, to minimize the evaporation losses, the viscometer's limbs are closed with Teflon caps. During the measurements of flow time,



**Figure 1.** Excess molar volume variation with mole fraction to systems (a) DEC (1) + toluene (2); (b) DEC (1) + methanol (2); (c) DEC (1) + 2-propanol (2) for experimental values:  $\blacksquare$ , 293.15 K;  $\blacklozenge$ , 298.15 K;  $\bigstar$ , 303.15 K;  $\lor$ , 313.15 K; left-facing solid triangle, 323.15 K; right-facing solid triangle, 333.15 K;  $\bigtriangleup$ , 343.15 K;  $\bigtriangledown$ , 353.15 K;  $\square$ , 363.15 K. Literature data: Wankhede et al.<sup>4</sup> to DEC (1) + toluene (2) at  $\bigcirc$ , 298.15 K; Rodriguez et al.<sup>5</sup> to DEC (1) + methanol (2) and DEC (1) + 2-propanol (2) at +, 293.15 K;  $\times$ , 298.15 K; \*, 303.15 K; -, 313.15 K; solid curves, calculated with Redlich–Kister equations; symbols, experimental values.



**Figure 2.** Viscosity deviation variation with mole fraction to systems (a) DEC (1) + toluene (2); (b) DEC (1) + methanol (2); (c) DEC (1) + 2-propanol (2) for experimental values:  $\blacksquare$ , 293.15 K;  $\blacklozenge$ , 298.15 K;  $\bigstar$ , 303.15 K;  $\lor$ , 313.15 K; left-facing solid triangle, 323.15 K; right-facing solid triangle, 333.15 K;  $\bigtriangleup$ , 343.15 K;  $\bigtriangledown$ , 353.15 K;  $\square$ , 363.15 K. Literature data: Rodriguez et al.<sup>3</sup> to DEC (1) + methanol (2) and DEC (1) + 2-propanol (2) at +, 293.15 K;  $\times$ , 298.15 K; \*, 303.15 K;  $\neg$ , 313.15 K; solid curves, calculated with Redlich–Kister equations; symbols, experimental values.

Table 1.	Comparison of Ex	perimental and	Literature Valu	es of Densities	$\rho$ , and $\gamma$	Viscosities $\eta$ ,	, for Pure	Compou	nd

		ρ/g•	cm <sup>-3</sup>	$\eta/1$	mPa•s			$ ho/g\cdot cm^{-3}$		$\eta/\mathrm{n}$	nPa•s
liquid	<i>T</i> /K	exptl	lit	exptl	lit	liquid	T/K	exptl	lit	exptl	lit
diethyl carbonate	293.15	0.97500	0.9747 <sup>5</sup>	0.800	0.795 <sup>3</sup>	toluene	333.15	0.82993	0.82927	0.385	0.38247
	298.15	0.96897	0.9691 <sup>5</sup>	0.746	$0.749^{3}$		343.15	0.82030	$0.8197^{7}$	0.350	$0.3491^{7}$
			$0.96916^4$				353.15	0.81054	$0.8094^{7}$	0.310	$0.3176^{7}$
	303.15	0.96239	0.96345	0.695	$0.699^{3}$		363.15	0.79768		0.286	
	313.15	0.95232	$0.9522^{5}$	0.610	$0.623^{3}$	methanol	293.15	0.79154	$0.7912^{12}$	0.582	$0.591^{3}$
			$0.95243^{6}$						$0.79106^{15}$		$0.577^{15}$
	323.15	0.93978	$0.94077^4$	0.541			298.15	0.78683	$0.7866^{12}$	0.550	$0.553^{3}$
	333.15	0.92869		0.483							$0.538^{13}$
	343.15	0.91633		0.435			303.15	0.78212	$0.7818^{12}$	0.512	$0.514^{3}$
	353.15	0.90502		0.393					$0.7821^{14}$		$0.497^{14}$
	363.15	0.89371		0.358					0.78195 <sup>15</sup>		$0.512^{15}$
toluene	298.15	0.86201	$0.86217^{8}$	0.554	$0.5541^{10}$		313.15	0.77260	$0.7726^{12}$	0.448	$0.450^{3}$
			$0.8609^9$		0.55429				0.7720115		$0.447^{15}$
			0.862311				323.15	0.76297		0.395	
	303.15	0.85832	$0.8565^{7}$	0.525	$0.5171^{7}$		333.15	0.74969		0.351	
			$0.8576^{10}$		$0.5226^{10}$	2-propanol	293.15	0.78528	$0.7894^{5}$	2.382	$2.386^{3}$
			$0.8578^{11}$		$0.5227^{11}$		298.15	0.78069	$0.7809^{5}$	2.069	$2.098^{3}$
	313.15	0.84892	$0.8479^{7}$	0.470	$0.4669^{7}$		303.15	0.77615	$0.7766^{5}$	1.743	$1.763^{3}$
			$0.8482^{10}$		$0.4659^{10}$		313.15	0.76634	$0.7678^{5}$	1.288	$1.325^{3}$
	323.15	0.83875	0.83857	0.423	0.42197		323.15	0.75566		1.002	
			$0.8387^{9}$		$0.4216^{9}$		333.15	0.74551		0.789	
			$0.8390^{11}$		$0.4216^{11}$		343.15	0.73502		0.626	

the caps of the limps were removed. The overall uncertainty of the viscosity measurements is dependent on the temperature control of the viscometer, the time of the flow, and the concentration, which are of the order of  $1 \times 10^{-2}$ ,  $1 \times 10^{-2}$ , and  $3 \times 10^{-4}$ , respectively. The uncertainty of viscosity was within  $\pm 0.003$  mPa·s.

In the experiments, the density and viscosity for one composition sample were measured at different temperatures. Densities and viscosities of pure compounds are reported in Table 1 together with the corresponding literature values. For the densities, good agreement was found between the measured and literature values for all these pure substances. There is an

Table 2.	Densities	, ρ, Visco	sities $\eta$ , Exce	ess Molar	Volumes	V <sup>E</sup> , and V	iscosity <b>E</b>	eviations $\Delta I$	for the H	3inary M	ixtures at	Different	: Temperatur	tes A			:	17 E	
	θ	h	ΛE	$\nabla \eta$		θ	μ	ΛE	$\nabla \eta$		θ	h	VE	$\nabla \eta$		θ	μ	ΛE	$\nabla \eta$
x	g•cm <sup>-3</sup>	mPa•s	$cm^{3} \cdot mol^{-1}$	mPa•s	x	g•cm <sup>-3</sup>	mPa•s	$cm^{3} \cdot mol^{-1}$	mPa•s	x	$g^{*}cm^{-3}$	mPa•s	$cm^3 \cdot mol^{-1}$	mPa•s	x	$g \cdot cm^{-3}$	mPa•s	$cm^{3} \cdot mol^{-1}$	mPa•s
				T - 20	0 15 K			(x) Dieth	yl Carbona	lte + (1 –	x) Toluene			T - 23	2 15 V				
0 0000	0 86201	0 554	0 0000	0000	0 6022	0 93018	0 664	-0.0549	-0.006	0 0000	0 82993	0 385	00000	CC = T	0.602	0 89288	0 447	-0.0421	-0.002
0.1014	0.87450	0.573	-0.0366	-0.001	0.7016	0.94028	0.684	-0.0457	-0.005	0.1014	0.84139	0.394	-0.0211	-0.001	0.7016	0.90220	0.452	-0.0327	-0.002
0.1984	0.88594	0.590	-0.0480	-0.002	0.7995	0.94995	0.702	-0.0326	-0.006	0.1984	0.85201	0.403	-0.0358	-0.001	0.7995	0.91111	0.461	-0.0204	-0.002
0.3006	0.9089767	0.608	-0.0595 -0.0619	-0.003	0.8975	0.95936	0.722	-0.0158	-0.004	0.3006	0.86285	0.413	-0.0446 -0.0484	-0.002	0.8975	0.97869	0.471 0.483	0.0000	-0.002
0.5212	0.92172	0.649	-0.0588	-0.005			2	00000	0000	0.5212	0.88509	0.434	-0.0468	-0.002			2	00000	0000
00000	0.010.0		00000	T = 30	3.15 K	101,000			2000	000000			000000	T = 34	3.15 K	0.001.71	00000	1000	
0.0000	0.87046	0.542	0.0000 -0.0306	-0.001	0.7016	0.93452	0.639	-0.0431	c00.0-	0.0000	0.82050	0.358	-0.0201	0.0003	0.7016	16188.0	0.407	-0.0364 -0.0287	-0.003
0.1984	0.88162	0.557	-0.0442	-0.002	0.7995	0.94393	0.657	-0.0322	-0.004	0.1984	0.84179	0.365	-0.0335	-0.0012	0.7995	0.89924	0.415	-0.0181	-0.002
0.3006	0.89306	0.573	-0.0559	-0.003	0.8975	0.95308	0.674	-0.0181	-0.003	0.3006	0.85232	0.373	-0.0414	-0.0020	0.8975	0.90769	0.425	-0.0073	-0.001
0.4024 0.5212	0.90400	0.609	-0.0562	-0.004	1.0000	0.90239	C60.U	0.000	0.000	0.4024 0.5212	0.80248 0.87394	0.382	-0.0434 -0.0423	-0.0021 -0.0028	1.0000	0.91033	0.450	0.000	0.000
				T = 31	3.15 K									T = 35	3.15 K				
0.0000	0.84892	0.470	0.0000	0.000	0.6022	0.91485	0.550	-0.0514	-0.004	0.0000	0.81054	0.310	0.0000	0.000	0.6022	0.87074	0.358	-0.0324	-0.002
0.1014	C6098.0	0.484	C/20.0-	- 0.000	0.7005	0.92460	0.70 0	-0.0416 -0.0301	-0.004	0.1014	0.82153	0.318	-0.020/	-0.001	0.7005	0.87966	0.36/	-0.0254	200.0-
0.3006	0.88339	0.510	-0.0411	-0.003	0.8975	0 94304	6/ 503	-0.0148	-0.004	0.3006	012000	0.334	-0.0320	100.0-	0 8975	0.89652	0 384	-0.0063	-0.001
0.4024	0.89433	0.523	-0.0568	-0.003	1.0000	0.95232	0.610	0.0000	0.000	0.4024	0.85202	0.342	-0.0392	-0.002	1.0000	0.90502	0.393	0.0000	0.000
0.5212	0.90666	0.539	-0.0545	-0.004 T = 32	3 15 K					0.5212	0.86329	0.352	-0.0378	-0.002 T = 36	3 15 K				
0.0000	0.83875	0.423	0.0000	0.000	0.6022	0.90316	0.491	-0.0465	-0.003	0.0000	0.79768	0.286	0.0000	0.000	0.6022	0.85877	0.328	-0.0296	-0.002
0.1014	0.85049	0.434	-0.0245	-0.001	0.7016	0.91268	0.503	-0.0359	-0.003	0.1014	0.80879	0.293	-0.0178	-0.000	0.7016	0.86785	0.336	-0.0240	-0.001
0.3006	0.87243	0.457	-0.0491	-0.002	0.8975	0.93072	0.527	-0.0135	-0.002	0.3006	0.82957	0.306	-0.0331	-0.001	0.8975	0.88504	0.351	-0.0059	-0.000
0.4024 0.5212	$0.88311 \\ 0.89517$	$0.468 \\ 0.481$	-0.0519 -0.0505	-0.003 -0.003	1.0000	0.93978	0.541	0.0000	0.000	0.4024 0.5212	$0.83971 \\ 0.85116$	0.313 0.322	-0.0348 -0.0315	-0.002 -0.002	1.0000	0.89371	0.358	0.0000	0.000
								(x) Diethy	d Carbonat	e + (1 –	x) Methano	I							
				T = 29	3.15 K					~				T = 31	3.15 K				
0.0000	0.79154	0.582	0.0000	0.000	0.6057	0.94283	0.700	-0.0620	-0.014	0.0000	0.77260	0.448	0.0000	0.000	0.6057	0.92059	0.5371	-0.0396	-0.009
0.1045	0.87469	662.0	+7 CO.O-	-0.007	CTU/.U 0.8000	070700 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	0.747	0070	-0.010	0.2128	0.85364	0.480	-0.0475	-0.003	0008.0	02026-0	100000 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	-0.0245	-0.006
0.2835	0.89208	0.634	-0.0785	-0.010	0.8845	0.96744	0.772	-0.0189	-0.004	0.2835	0.87071	0.490	-0.0524	-0.004	0.8845	0.94493	0.5874	-0.0164	-0.004
0.4025	0.91512	0.655	1620.0-	-0.015	1.0000	0.97500	0.801	0.0000	0.000	0.4025	0.89330	0.503	-0.0506	-0.010	1.0000	0.95232	0.6101	0.0000	0.000
1010.0	+07CC.0	6/0.0	0.0.41	T = 29	8.15 K					10100	12202.0	170.0	0.0401	T = 32	3.15 K				
0.0000	0.78683	0.550	0.0000	0.000	0.6057	0.93711	0.656	-0.0536	-0.013	0.0000	0.76297	0.395	0.0000	0.000	0.6057	0.90928	0.477	-0.0278	-0.008
0.1045	0.83464	0.567	-0.0359 -0.0638	-0.004	0.7015	0.94689	0.696	-0.0480 -0.0345	-0.011	0.1045	0.80945	0.408	-0.0295 -0.0378	-0.002	0.7015	0.91888 0 0.9774	0.493	-0.0260 -0.0183	-0.005
0.2835	0.88665	0.598	-0.0713	-0.009	0.8845	0.96165	0.715	-0.0183	-0.009	0.2835	0.85986	0.432	-0.0396	-0.004	0.8845	0.93343	0.523	-0.0089	-0.001
0.4025	0.90956	0.616	-0.0713	-0.014	1.0000	0.96897	0.746	0.0000	0.000	0.4025	0.88225	0.444	-0.0387	-0.010	1.0000	0.93978	0.541	0.0000	0.000
1010.0	CC076.0	/ 60.0	-0.0041	T = 30	3.15 K					1010.0	0.898/2	0.401	00000-	-0.010 T = 33	3.15 K				
0.0000	0.78212	0.512	0.0000	0.000	0.6057	0.93174	0.611	-0.0492	-0.011	0.0000	0.74969	0.351	0.0000	0.000	0.6057	0.89715	0.426	-0.0154	-0.005
0.1045	0.82980	0.527	-0.0407	-0.003	0.7015	0.94149	0.630	-0.0444	-0.010	0.1045	0.79631	0.363	-0.0227	-0.002	0.7015	0.90690	0.441	-0.0142	-0.003
0.2835	0.80412 0.88142	0.556	-0.0638	-0.004 -0.007	0.8000 0.8845	0.95623	1 CO.U	1 c c 0.0 – 1 7 1 0 0 –	-0.007	0.2120	2002800 0 84711	0.270 0384	-0.0200	cuu.u- 200.00-	0.8845	717179 0	0.466	C0000-	cuu.u-
0.4025	0.90425	0.573	-0.0641	-0.012	1.0000	0.96239	0.695	0.0000	0.000	0.4025	0.86976	0.397	-0.0266	-0.007	1.0000	0.92869	0.483	0.0000	0.000
0.5161	0.92100	0.593	-0.0580	-0.013						0.5161	0.88644	0.410	-0.0223	-0.008					

		-0.206	-0.150	-0.112	-0.090	0.000			-0.148	-0.108	-0.081	-0.065	0.000			-0.106	-0.078	-0.050	-0.047	0.000								
		0.5025	0.4328	0.3547	0.2948	0.0000			0.5805	0.4987	0.4006	0.3335	0.0000			0.6428	0.5434	0.4401	0.3595	0.0000								
T = 323 15 K		0.524	0.517	0.519	0.521	0.541			0.460	0.458	0.461	0.464	0.483			0.407	0.409	0.413	0.417	0.435								
		0.87949	0.90094	0.91258	0.91893	0.93978			0.86810	0.88956	0.90129	0.90767	0.92869			0.85592	0.87730	0.88896	0.89536	0.91633								
	.15 K	0.5905	0.7267	0.8053	0.8494	1.0000		.15 K	0.5905	0.7267	0.8053	0.8494	1.0000		.15 K	0.5905	0.7267	0.8053	0.8494	1.0000								
	T = 323	0.000	-0.175	-0.242	-0.258	-0.254	-0.236	T = 333	0.000	-0.121	-0.169	-0.181	-0.181	-0.168	T = 343	0.000	-0.083	-0.118	-0.128	-0.128	-0.120							
		0.0000	0.1982	0.3379	0.4101	0.4581	0.4898	00000	0.0000	0.2330	0.3996	0.4877	0.5424	0.5674		0.0000	0.2620	0.4452	0.5393	0.6062	0.6428							
		1.002	0.780	0.668	0.604	0.562	0.535		0.789	0.637	0.559	0.515	0.485	0.467		0.626	0.523	0.470	0.440	0.421	0.410							
Propanol	4	0.75566	0.78171	0.80458	0.82695	0.84673	0.86462		0.74551	0.77112	0.79364	0.81577	0.83541	0.85329		0.73502	0.76013	0.78230	0.80417	0.82353	0.84114							
+(1-x)2-		0.0000	0.1015	0.1993	0.3027	0.4029	0.5018		0.0000	0.1015	0.1993	0.3027	0.4029	0.5018		0.0000	0.1015	0.1993	0.3027	0.4029	0.5018							
Carbonate -		-0.621	-0.442	-0.328	-0.25	0.000			-0.527	-0.377	-0.278	-0.219	0.000			-0.424	-0.304	-0.225	-0.177	0.000			-0.287	-0.207	-0.154	-0.122	0.000	
(x) Diethyl	•	0.3075	0.2745	0.2292	0.1880	0.0000			0.3365	0.2988	0.2509	0.2067	0.0000			0.3465	0.3094	0.2616	0.2172	0.0000			0.4083	0.3614	0.2995	0.2493	0.0000	
		0.827	0.791	0.781	0.782	0.800			0.760	0.731	0.726	0.726	0.746			0.700	0.677	0.674	0.676	0.695			0.601	0.588	0.588	0.590	0.610	
T = 293.15  K		$\begin{array}{c} 0.91448 \\ 0.93622 \\ 0.94793 \\ 0.95432 \\ 0.97500 \end{array}$		0.90868	0.93030	0.94194	0.94831	0.96897			0.90271	0.92409	0.93560	0.94189	0.96239			0.89218	0.91363	0.92525	0.93159	0.95232						
	.15 K	0.5905	0.7267	0.8053	0.8494	1.0000		.15 K	0.5905	0.7267	0.8053	0.8494	1.0000		T = 303.15  K	0.5905	0.7267	0.8053	0.8494	1.0000		.15 K	0.5905	0.7267	0.8053	0.8494	1.0000	
	T = 293	0.000	-0.635	-0.814	-0.830	-0.799	-0.724	T=298	0.000	-0.542	-0.687	-0.705	-0.676	-0.614		0.000	-0.414	-0.539	-0.559	-0.538	-0.492	T = 313	0.000	-0.256	-0.347	-0.367	-0.358	-0.330
		0.0000	0.1100	0.1816	0.2269	0.2566	0.2851		0.0000	0.1151	0.2006	0.2506	0.2831	0.3196		0.0000	0.1190	0.2091	0.2679	0.3217	0.3433	0 0000	0.0000	0.1481	0.2595	0.3217	0.3553	0.3950
		2.382	1.586	1.252	1.073	0.946	0.865		2.069	1.393	1.119	0.964	0.860	0.791		1.743	1.223	0.996	0.867	0.782	0.726		1.288	0.964	0.806	0.716	0.657	0.618
		0.78528	0.81295	0.83712	0.86152	0.88088	0.90157		0.78069	0.80812	0.83198	0.85503	0.87534	0.89355		0.77615	0.80327	0.82684	0.84957	0.86948	0.88762		0.76634	0.79312	0.81646	0.83913	0.85920	0.87718
		0.0000	0.1015	0.1993	0.3027	0.4029	0.5018		0.0000	0.1015	0.1993	0.3027	0.4029	0.5018		0.0000	0.1015	0.1993	0.3027	0.4029	0.5018		0.0000	0.1015	0.1993	0.3027	0.4029	0.5018

appreciable difference for the viscosity data among the literatures for methanol. The viscosity values of methanol obtained in this study are in good agreement with those of Rodriguez et al.<sup>3</sup>

## **Results and Discussion**

The experimental values of density and viscosity for binary mixture at different temperatures and at atmospheric pressure are listed in Table 2. Excess molar volumes were calculated from our measurements according to the following equation:<sup>16</sup>

$$V^{\rm E} = \frac{x_1 M_1 + x_2 M_2}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2}$$
(2)

where  $x_1$  and  $x_2$  are mole fractions;  $M_1$  and  $M_2$  are the molar masses; and  $\rho_1$  and  $\rho_2$  are the densities of pure components 1 and 2, respectively. Quantities without subscripts refer to the mixture.

The viscosity derivations were calculated from the following relation:

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

where  $\eta$  is the viscosity of mixtures and  $\eta_1$  and  $\eta_2$  are the viscosity of components 1 and 2, respectively. The values of  $V^{\rm E}$  and  $\Delta \eta$  for each mixture were fitted to the Redlich–Kister polynominal equation:<sup>17</sup>

$$Y = x_1(1 - x_1) \sum_{i=0}^{n} A_i (2x_1 - 1)^i$$
(4)

where  $Y = V^{E}$  or  $\Delta \eta$ ,  $A_i$  are adjustable parameters, and  $x_1$  is the mole fraction of component 1.

In each case, the optimum number of coefficients  $A_i$  was determined from an examination of the variation of the standard derivations:

$$\sigma(Y) = \left[\sum (Y_{\rm cal} - Y_{\rm exp})^2 / (n - m)\right]^{(1/2)}$$
(5)

where n is the total number of experimental values and m is the number of parameters.

The excess molar volume data and the viscosity derivations are presented in Table 2, they are shown graphically in Figures 1 and 2, and comparison is made with literature data. Our values are in disagreement with those reported by Rodriguez et al. for the binary mixtures of DEC + toluene and + methanol. It may be that the values of  $V^{\rm E}$  and  $\Delta \eta$  for these two binary mixtures are very small and only larger one order quantitatively than their uncertainties. Table 3 lists the values of the parameters  $A_i$ together with the standard deviations.

It can be seen from the Figure 1 that  $V^{\rm E}$  values for DEC + toluene and methanol over the whole composition range are negative. The absolute values of  $V^{\rm E}$  are small and decrease with temperature rising. However, for diethyl carbonate + 2-propanol, the excess molar volumes are positive and large, increasing with temperature. It can be summarized from our series of work<sup>16–18</sup> that  $V^{\rm E}$  values may be affected by three factors. The first factor is the specific forces between molecules, such as hydrogen bonds, and charge–transfer complexes, breaking of hydrogen bonds and complexes giving positive excess molar volumes, and forming of hydrogen bonds and complexes bringing negative values of  $V^{\rm E}$ . The second factor is the physical intermolecular forces, including electrostatic forces between charged particles and between permanent dipoles and so on, induction forces between a permanent dipole and an

Table 3.	<b>Coefficients of the Redlich</b>	-Kister Equation and Standar	d Deviation for Excess Molar	Volumes and Viscosity	<b>Deviations of Mixtures</b>
		1			

T/K	property	$A_0$	$A_1$	$A_2$	$A_3$	$A_4$	σ						
		(x)	Diethyl Carbonate +	-(1-x) Toluene									
298.15	$V^{\rm E}/{\rm cm^3 \cdot mol^{-1}}$	-0.2377	0.0427	-0.0681	0.1531		0.011						
	$\Delta \eta$ /mPa·s	-0.0222	-0.0178				0.001						
303.15	$V^{\dot{E}}/cm^3 \cdot mol^{-1}$	-0.2276	0.0558	-0.0535	0.0459		0.008						
	$\Delta \eta$ /mPa•s	-0.0185	-0.0165				0.001						
313.15	$V^{\dot{E}}$ /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.2232	0.0474	-0.0100	0.0603		0.006						
	$\Delta \eta$ /mPa•s	-0.0147	-0.0142				0.002						
323.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.2029	0.0635	0.0004	0.0187		0.007						
	$\Delta \eta$ /mPa•s	-0.0101	-0.0132				0.001						
333.15	$V^{\dot{E}}$ /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.1901	0.0712	0.0365	0.0159		0.003						
	$\Delta \eta$ /mPa•s	-0.0060	-0.0103				0.001						
343.15	$V^{\dot{E}}$ /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.1708	0.0702	0.0314	0.0299		0.003						
	$\Delta \eta$ /mPa•s	-0.0029	-0.0068				0.001						
353.15	$V^{\acute{\mathrm{E}}}$ /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.1514	0.0606	0.0050	0.0617		0.002						
	$\Delta \eta$ /mPa•s	-0.0078	-0.0103				0.001						
363.15	$V^{\acute{\mathrm{E}}}$ /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.1345	0.0512	0.0042	0.0496		0.004						
	$\Delta\eta/mPa$ ·s	-0.0082	-0.0123				0.002						
		$(x) \square$	Diethyl Carbonate +	(1 - x) Methanol									
293.15	$V^{\text{E}/\text{cm}^3 \cdot \text{mol}^{-1}}$	-0.2987	0.0970	-0.1222	0.2219		0.015						
	$\Delta \eta$ /mPa•s	-0.0598	-0.0313	0.0210	0.0753		0.008						
298.15	$V^{\acute{\mathrm{E}}}$ /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.2711	0.1632	-0.0350	-0.0577		0.017						
	$\Delta \eta$ /mPa•s	-0.0516	-0.0150	-0.0151	-0.0271		0.010						
303.15	$V^{\acute{\mathrm{E}}}/\mathrm{cm}^{3}\cdot\mathrm{mol}^{-1}$	-0.2358	0.0842	-0.1022	0.1277		0.014						
	$\Delta \eta$ /mPa•s	-0.0496	-0.0279	0.0411	0.0542		0.006						
313.15	$V^{\acute{\mathrm{E}}}/\mathrm{cm}^{3}\cdot\mathrm{mol}^{-1}$	-0.1872	0.0902	-0.1218	0.0726		0.008						
	$\Delta \eta$ /mPa•s	-0.0348	-0.0150	0.0082	0.0115		0.009						
323.15	$V^{\acute{\mathrm{E}}}$ /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.1385	0.0612	-0.0985	0.1305		0.007						
	$\Delta \eta$ /mPa•s	-0.0341	-0.0129	0.0330	0.0270		0.009						
333.15	$V^{\acute{\mathrm{E}}}/\mathrm{cm}^{3}\cdot\mathrm{mol}^{-1}$	-0.0871	0.0588	-0.0664	0.1281		0.008						
	$\Delta \eta$ /mPa·s	-0.0247	0.0081	0.0099	-0.0150		0.006						
(x) Diethyl Carbonate + $(1 - x)$ 2-Propanol													
293.15	$V^{\rm E}/{\rm cm^3 \cdot mol^{-1}}$	1.1451	0.4496	0.5721	-0.4826	-0.4546	0.021						
	$\Delta \eta$ /mPa•s	-2.8924	1.7721	-1.1319	2.0741	-2.2033	0.025						
298.15	$V^{\acute{\mathrm{E}}}/\mathrm{cm}^{3}\cdot\mathrm{mol}^{-1}$	1.2657	0.4377	0.6069	-0.3733	-0.5468	0.029						
	$\Delta \eta$ /mPa•s	-2.4565	1.4708	-0.8682	1.8120	-2.0670	0.023						
303.15	$V^{\acute{\mathrm{E}}}/\mathrm{cm}^{3}\cdot\mathrm{mol}^{-1}$	1.3695	0.3111	0.3136	-0.0444	-0.0872	0.020						
	$\Delta n/mPa \cdot s$	-1.9632	1.1626	-0.7662	1.2768	-1.2678	0.015						
313.15	$V^{\rm E}/{\rm cm^{3} \cdot mol^{-1}}$	1.5610	0.3955	0.8115	-0.3491	-0.7504	0.035						
	$\Delta n/mPa \cdot s$	-1.3147	0.7518	-0.5132	0.6514	-0.5844	0.007						
323.15	$V^{\rm E}/\rm cm^3 \cdot mol^{-1}$	1.9546	0.3160	0.8410	-0.4189	-0.6476	0.039						
	$\Delta n/mPa \cdot s$	-0.9374	0.5158	-0.3611	0.4027	-0.3561	0.007						
333.15	$V^{E}/cm^{3}\cdot mol^{-1}$	2.2789	0.2620	1.0084	-0.4483	-0.9150	0.040						
	$\Lambda n/mPa\cdots$	-0.6701	0.3481	-0.2403	0.2669	-0.2274	0.006						
343.15	$V^{\rm E}/{\rm cm^3 \cdot mol^{-1}}$	2.5538	0.2621	0.8519	-0.5554	-0.6880	0.045						
2.0110	$\Lambda n/mPa\cdots$	-0.4776	0.2332	-0.1712	0.1765	-0.1191	0.005						
		00	0.2002	0.1.7.12	0.1,00	0	0.000						

induced dipole, and forces of attraction (dispersion forces) and repulsion between nonpolar molecules. Physical intermolecular forces are weak usually, and the sign of  $V^{\rm E}$  values may be positive or negative. The third factor is the structural characteristics of component, arising from geometrical fitting of one component into the other's structure, due to the differences in shape and size of components and free volume. Because of the interaction between the polarity of DEC and  $\pi$  electron in aromatic ring, the values of excess molar volumes are negative for the DEC + toluene mixture. For DEC + alcohols systems, the main contributions to  $V^{\rm E}$  are chemical. DEC is a small polar liquid with dipole-dipole interactions in pure liquid. In contrast, alcohols are self-associated through hydrogen bonding. When these two components are mixed, the hydrogen bonding in alcohols is broken, and dipole-dipole interactions between DEC with alcohol are formed. Because the interaction strength of the hydrogen is stronger than the dipole interaction, the  $V^{\rm E}$ values are positive for DEC with 2-propanol, whereas the sign of excess molar volumes is negative for the DEC + methanol mixture. The steric effect caused by the two additional methylene groups in 2-propanol had to greatly reduced its hydrogen bonding pattern as compared to methanol. Also, the larger size of 2-propanol hindered the molecular packing between the two components.

Figure 2 shows viscosity deviations for these three binary mixtures, plotted against mole fraction together with the fitted curve, obtained from the Redlich–Kister equation. The viscosity deviations for these systems at selected temperatures are negative over the entire composition. For the DEC + toluene and methanol mixtures, the absolute viscosity deviations are very small.

As shown in Figures 1 and 2, there are large difference between the experimental values with literature values<sup>3,5</sup> to the excess molar volumes and the viscosity deviation for DEC + methanol mixture. The reason is the values of these two properties are very small. The little difference in the values of densities and viscosities will result in the larger changes in the excess volumes and deviations of viscosity.

# Conclusion

Densities and viscosities of the binary systems of DEC + toluene, + methanol, and + 2-propanol have been measured at several temperatures and for the whole composition range. The excess molar volumes and viscosity deviations were computed. The computed quantities have been fitted to the Redlich–Kister equation. Excess molar volumes and viscosity deviation show a systematic change with increasing temperature.  $V^{\rm E}$  values are negatives for DEC + toluene and methanol; however,  $V^{\rm E}$  values

are positive over the entire composition range for DEC + 2-propanol. The deviations of viscosity,  $\Delta \eta$ , are negative for all the mixtures over the entire composition range and become less negative with increasing temperature. The effect of interactions of components on excess molar volumes and viscosity deviation is discussed.

#### Literature Cited

- Pardo, J. M.; Tovar, C. A.; Cerdeireina, C. A.; Carballo, E.; Romani, L. Excess molar volumes and excess molar heat capacities of (dimethyl carbonate, or diethyl carbonate + *n*-heptane) at several temperatures. *J. Chem. Thermodyn.* **1999**, *31*, 787–796.
- (2) Francesconi, R.; Comelli, F. Excess molar enthalpies, densities, and excess molar volumes of diethyl carbonate in binary mixtures with seven *n*-alkanols at 298.15 K. J. Chem. Eng. Data 1997, 42, 45–48.
- (3) Rodriguez, A.; Canosa, J.; Dominguez, A.; Tojo, J. Dynamic viscosities of diethyl carbonate with linear and secondary alcohols at several temperatures. J. Chem. Eng. Data 2004, 49, 157–162.
- (4) Lopez, E. R.; Lugo, L.; Comunas, M. J. P. Temperature dependence of the excess molar volume of (dimethyl carbonate, or diethyl carbonate + toluene) from *T*= 278.15 K to 323.15 K. *J. Chem. Thermodyn.* 2000, *32*, 743–754.
- (5) Rodriguez, A.; Canosa, J.; Tojo, J. Density, refractive index, and speed of sound of binary mixtures (diethyl carbonate + alcohols) at several temperatures. J. Chem. Eng. Data 2001, 46, 1506–1515.
- (6) Rodriguez, A.; Canosa, J.; Tojo, J. Physical properties of binary mixtures (dimethyl carbonate + alcohols) at several temperatures. J. Chem. Eng. Data 2001, 46, 1476–1486.
- (7) Assael, M. J.; Dalaouti, N. K.; Dymond, J. H. The viscosity of toluene in the temperature range 210 to 370K. *Int. J. Thermophys.* 2000, 21, 291–299.
- (8) Fermeglia, M.; Lapasin, R. Excess volumes and viscosities of binary mixtures of organics. J. Chem. Eng. Data 1988, 33, 415–417.
- (9) Malhotra, R.; Woolf, L. A.Temperature and density dependence of the viscosities of octane and toluene. J. Chem. Eng. Data 1997, 42, 1254–1260.

- (10) Exarchos, N. C.; Tasioula-Margar, M.; Demetropoulos, I. N. Viscosities and densities of dilute solutions of glycerol trioleate + octane, + *p*-xylene, + toluene, and + chloroform. *J. Chem. Eng. Data* **1995**, 40, 567–571.
- (11) Assael, M. J.; Papadaki, M.; Wakeham, W. A. Measurements of the viscosities of benzene, toluene, and *m*-xylene at pressure up to 80 MPa. J. Thermophys. **1991**, 12, 449–457.
- (12) Comelli, F.; Francesconi, R. Isothermal vapor-liquid equilibria measurements, excess molar enthalpies, and excess molar volumes of dimethyl carbonate + methanol, + ethanol, and + propan-1-ol at 313.15 K. J. Chem. Eng. Data **1997**, 42, 705–709.
- (13) Aminabhavi, T. M.; Patil, V. B.; Banerjee, K. Thermodynamic interactions in binary mixtures of ethenylbenzene with methanol, ethanol, butan-1-ol, pentan-1-ol, and hexan-1-ol in the temperature range 298.15–308.15 K. J. Chem. Eng. Data 1999, 44, 1291–1297.
- (14) Rived, F.; Roses, M.; Bosch, E. Densities, refractive indices, absolute viscosities, and static dielectric constants of 2-methylpropan-2-ol + hexane, + benzene, + propan-2-ol, + methanol, + ethanol, and + water at 303.2 K. *J. Chem. Eng. Data* **1996**, *40*, 1111–1114.
- (15) Djojoputro, H.; Ismadji, S. Density and viscosity of binary mixtures of ethyl-2-methylbutyrate and ethyl hexanoate with methanol, ethanol, and 1-propanol at (293.15, 303.15, and 313.15) K. J. Chem. Eng. Data 2005, 50, 1343–1347.
- (16) Yang, C.; Ma, P.; Zhou, Q. Excess molar volumes and viscosities of binary mixtures of sulfolane with benzene, toluene, ethylbenzene, *p*-xylene, *o*-xylene, and *m*-xylene at 303.15 and 323.15 K and atmospheric pressure. J. Chem. Eng. Data **2004**, 49, 881–885.
- (17) Yang, C.; Ma, P.; Jing, F. Tang, D. Excess molar volume, viscosity and heat capacities for the mixtures of ethylene glycol-water from 273.15 K to 353.15 K. J. Chem. Eng. Data 2003, 48, 836–840.
- (18) Yang, C.; Xu, W.; Ma, P. Thermodynamic properties of binary mixtures of *p*-xylene with cyclohexane, heptane, octane, and *N*-methyl-2-pyrrolidone at several temperatures. *J. Chem. Eng. Data* **2004**, *49*, 1794–1801.

Received for review October 7, 2005. Accepted December 27, 2005.

JE050414D