

Densities and Viscosities of Binary Mixtures of *m*-Cresol with Ethylene Glycol or Methanol over Several Temperatures

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Densities and viscosities for the *m*-cresol + (ethylene glycol or methanol) binary systems have been experimentally determined over several temperatures and at normal atmospheric pressure, over the entire mole fraction range. Other mixing properties of interest such as the excess molar volumes and the viscosity deviations have been also obtained for each of the systems. The excess molar volumes, V^E , and viscosity deviations, $\Delta\eta$, have been fitted to the Redlich–Kister equation, and the coefficients and estimate of the standard error values are presented.

Introduction

Physical properties of binary organic mixtures have been extensively studied in the literature of solution chemistry in view of the importance of such data in many areas of science and engineering. Experimental data of physical properties such as density and viscosity in binary mixtures give information on the existence of specific molecular interactions. Transport and thermodynamic properties also provide insight into the molecular arrangement of liquids.¹ Thermophysical properties are required for the design of industrial processes. This paper is part of our systematic program of research on the measurement of physical and transport properties of binary liquid mixtures.

In this work, the densities and viscosities of *m*-cresol + ethylene glycol or methanol mixtures were measured at (308.15 to 353.15) K and (308.15 to 328.15) K, respectively. The results were used to calculate excess molar volumes and deviations in viscosity over the entire mole fraction range for the mixtures. Experimental values were correlated by the Redlich–Kister equation. The root mean square deviations between experimental and calculated values are shown.

Experimental Section

Materials. All of the chemicals used in this study were obtained from Tianjin Reagent Company. They were degassed ultrasonically and dried over 0.4 nm molecular sieves. The mass fraction purities, tested by gas chromatography, were as follows: *m*-cresol (0.990), ethylene glycol (0.994), and methanol (0.995). All chemicals were used without further purification. To reduce the water content, substances were dried over molecular sieves type 4 Å, except methanol, for which type 3 Å was used. Before measurements, pure liquids were degassed ultrasonically.

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 temper-

Table 1. Comparison of Experimental and Literature Values of Densities (ρ) and Viscosities (η) for Pure Compounds

liquid	<i>T</i> /K	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\eta/\text{mPa}\cdot\text{s}$		
		exptl	lit	exptl	lit	
<i>m</i> -cresol	308.15	1.02164	1.02200 ²	7.701		
	313.15	1.01763	1.0160 ³	6.120	6.252 ³ 6.1800 ⁵	
	318.15	1.01360		5.022		
	323.15	1.00956	1.0077 ³ 1.0098 ⁴	4.158	4.111 ³	
	333.15	1.00144	1.0004 ³ 1.0021 ⁶	2.980	3.007 ³	
	343.15	0.99133		2.231		
	353.15	0.98542		1.759		
	ethylene glycol	308.15	1.10188	1.1031 ⁸	11.193	11.050 ⁸
		313.15	1.09833	1.0936 ⁷ 1.0997 ⁸	9.444	9.443 ⁷ 9.244 ⁸ 9.409 ⁹
		318.15	1.09475	1.0967 ⁸	7.972	7.969 ⁸
323.15		1.09116	1.09116 ⁷ 1.0934 ⁸	6.794	6.992 ⁷ 6.712 ⁸	
333.15		1.08394	1.0764 ⁷	5.065	5.060 ⁷ 5.076 ⁹	
343.15		1.07453	1.0676 ⁷	3.879	3.987 ⁷	
353.15		1.06972	1.0600 ⁷	3.058	3.021 ⁷ 3.078 ⁹	
methanol		308.15	0.77692	0.77703 ¹¹	0.476	0.476 ¹¹
		313.15	0.77213	0.77201 ¹⁰ 0.77236 ¹¹	0.445	0.447 ¹⁰ 0.448 ¹¹
		318.15	0.76731		0.421	
	323.15	0.76246		0.395		
	328.15	0.75785		0.372		

ature was controlled automatically within ± 0.01 K of the selected value. Before each series of measurements, the instrument was calibrated at atmospheric pressure with double-distilled 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 calibration was accepted if the measurements were within $\pm 5 \times 10^{-5} \text{ g}\cdot\text{cm}^{-3}$ of the published values. The uncertainty in density measurements was $\pm 5 \times 10^{-5} \text{ g}\cdot\text{cm}^{-3}$. Density measurements were reproducible to $\pm 3 \times 10^{-5} \text{ g}\cdot\text{cm}^{-3}$.

The liquid mixtures were prepared by weight using a BP210s balance accurate to within ± 0.01 mg. The average uncertainty in the compositions (mole fraction) of the mixtures was estimated to be less than ± 0.0001 . The molar excess volumes

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Table 2. Densities (ρ), Viscosities (η), Excess Molar Volumes (V^E), and Viscosity Deviations ($\Delta\eta$) for the Binary Mixtures at Different Temperatures

	ρ	η	V^E	$\Delta\eta$		ρ	η	V^E	$\Delta\eta$		ρ	η	V^E	$\Delta\eta$
x_1	$\text{g}\cdot\text{cm}^{-3}$	$\text{mPa}\cdot\text{s}$	$\text{cm}^3\cdot\text{mol}^{-1}$	$\text{mPa}\cdot\text{s}$	x_1	$\text{g}\cdot\text{cm}^{-3}$	$\text{mPa}\cdot\text{s}$	$\text{cm}^3\cdot\text{mol}^{-1}$	$\text{mPa}\cdot\text{s}$	x_1	$\text{g}\cdot\text{cm}^{-3}$	$\text{mPa}\cdot\text{s}$	$\text{cm}^3\cdot\text{mol}^{-1}$	$\text{mPa}\cdot\text{s}$
(x) <i>m</i> -Cresol + (1 - x_1) Ethylene Glycol														
$T = 308.15$ K														
0.0000	1.10188	11.193	0.0000	0.000	0.3996	1.05833	11.881	-0.0742	2.084	0.8000	1.03115	8.695	-0.0079	0.296
0.0998	1.08836	11.725	-0.0176	0.880	0.5000	1.05038	11.198	-0.0673	1.751	0.8989	1.02620	8.030	-0.0029	-0.024
0.1999	1.07736	12.317	-0.0687	1.822	0.5967	1.04344	10.373	-0.0472	1.263	1.0000	1.02164	7.701	0.0000	0.000
0.3000	1.06731	12.328	-0.0814	2.183	0.7002	1.03660	9.452	-0.0059	0.704					
$T = 313.15$ K														
0.0000	1.09833	9.444	0.0000	0.000	0.3996	1.05444	9.673	-0.0701	1.557	0.8000	1.02719	7.030	-0.0082	0.245
0.0998	1.08466	9.704	-0.0142	0.591	0.5000	1.04647	9.067	-0.0641	1.285	0.8989	1.02222	6.496	-0.0036	0.040
0.1999	1.07358	10.135	-0.0648	1.356	0.5967	1.03951	8.417	-0.0451	0.956	1.0000	1.01763	6.120	0.0000	0.000
0.3000	1.06347	10.020	-0.0772	1.573	0.7002	1.03266	7.670	-0.0053	0.553					
$T = 318.15$ K														
0.0000	1.09475	7.972	0.0000	0.000	0.3996	1.05055	7.906	-0.0671	1.113	0.8000	1.02323	5.853	-0.0104	0.241
0.0998	1.08098	8.113	-0.0133	0.435	0.5000	1.04256	7.441	-0.0628	0.944	0.8989	1.01822	5.346	-0.0044	0.026
0.1999	1.06979	8.371	-0.0619	0.989	0.5967	1.03863	6.893	-0.0456	0.681	1.0000	1.01360	5.022	0.0000	0.000
0.3000	1.05961	8.275	-0.0734	1.188	0.7002	1.02870	6.322	-0.0049	0.415					
$T = 323.15$ K														
0.0000	1.09116	6.794	0.0000	0.000	0.3996	1.04664	6.572	-0.0635	0.831	0.8000	1.01925	4.811	-0.0118	0.126
0.0998	1.07728	6.869	-0.0121	0.338	0.5000	1.03863	6.182	-0.0606	0.706	0.8989	1.01422	4.433	-0.0062	0.009
0.1999	1.06598	7.015	-0.0584	0.748	0.5967	1.03163	5.763	-0.0436	0.542	1.0000	1.00956	4.158	0.0000	0.000
0.3000	1.05575	6.894	-0.0702	0.890	0.7002	1.02474	5.270	-0.0055	0.321					
$T = 333.15$ K														
0.0000	1.08394	5.065	0.0000	0.000	0.3996	1.03880	4.817	-0.0578	0.585	0.8000	1.01125	3.488	-0.0145	0.091
0.0998	1.06986	5.133	-0.0106	0.276	0.5000	1.03072	4.450	-0.0555	0.428	0.8989	1.00616	3.219	-0.0079	0.028
0.1999	1.05835	5.087	-0.0531	0.439	0.5967	1.02371	4.145	-0.0430	0.324	1.0000	1.00144	2.980	0.0000	0.000
0.3000	1.04797	4.972	-0.0623	0.533	0.7002	1.01676	3.794	-0.0047	0.189					
$T = 343.15$ K														
0.0000	1.07453	3.879	0.0000	0.000	0.3996	1.02890	3.554	-0.0533	0.333	0.8000	1.00125	2.635	-0.0188	0.075
0.0998	1.06026	3.810	-0.0077	0.096	0.5000	1.02081	3.332	-0.0553	0.277	0.8989	0.99613	2.426	-0.0130	0.029
0.1999	1.04859	3.820	-0.0476	0.270	0.5967	1.01376	3.096	-0.0437	0.200	1.0000	0.99133	2.231	0.0000	0.000
0.3000	1.03814	3.720	-0.0580	0.336	0.7002	1.00679	2.858	-0.0075	0.133					
$T = 353.15$ K														
0.0000	1.06972	3.058	0.0000	0.000	0.3996	1.02339	2.734	-0.0498	0.195	0.8000	0.99550	2.048	-0.0235	0.029
0.0998	1.05521	2.995	-0.0059	0.067	0.5000	1.01521	2.566	-0.0524	0.157	0.8989	0.99032	1.901	-0.0179	0.011
0.1999	1.04324	2.967	-0.0376	0.169	0.5967	1.00810	2.401	-0.0429	0.118	1.0000	0.98542	1.759	0.0000	0.000
0.3000	1.03271	2.882	-0.0517	0.214	0.7002	1.00108	2.237	-0.0090	0.088					
(x) <i>m</i> -Cresol + (1 - x_1) Methanol														
$T = 308.15$ K														
0.0000	0.77692	0.476	0.0000	0.000	0.3999	0.93980	1.778	-0.5977	-1.587	0.7999	1.00299	4.783	-0.2833	-1.472
0.0999	0.83866	0.725	-0.4221	-0.473	0.4998	0.96097	2.367	-0.6056	-1.720	0.8999	1.01319	6.060	-0.1670	-0.918
0.1999	0.88062	0.976	-0.4923	-0.945	0.6001	0.97735	2.958	-0.4992	-1.854	1.0000	1.02164	7.701	0.0000	0.000
0.2998	0.91411	1.319	-0.5954	-1.323	0.6998	0.99184	3.767	-0.4536	-1.765					
$T = 313.15$ K														
0.0000	0.77213	0.445	0.0000	0.000	0.3999	0.93558	1.577	-0.6181	-1.138	0.7999	0.99897	4.007	-0.2953	-0.978
0.0999	0.83409	0.676	-0.4350	-0.336	0.4998	0.95682	2.079	-0.6259	-1.202	0.8999	1.00918	4.978	-0.1737	-0.574
0.1999	0.87622	0.906	-0.5108	-0.674	0.6001	0.97331	2.556	-0.5220	-1.295	1.0000	1.01763	6.120	0.0000	0.000
0.2998	0.90992	1.205	-0.6233	-0.941	0.6998	0.98787	3.228	-0.4765	-1.188					
$T = 318.15$ K														
0.0000	0.76731	0.421	0.0000	0.000	0.3999	0.93138	1.419	-0.6423	-0.842	0.7999	0.99493	3.418	0.3077	0.683
0.0999	0.82948	0.621	-0.4477	-0.259	0.4998	0.95274	1.835	-0.6537	-0.886	0.8999	1.00513	4.153	-0.1787	-0.408
0.1999	0.87178	0.835	-0.5291	-0.506	0.6001	0.96931	2.243	-0.5499	-0.939	1.0000	1.01360	5.022	0.0000	0.000
0.2998	0.90555	1.100	-0.6410	-0.701	0.6998	0.98392	2.789	-0.5043	-0.852					
$T = 323.15$ K														
0.0000	0.76246	0.395	0.0000	0.000	0.3999	0.92714	1.290	-0.6656	-0.610	0.7999	0.99088	2.936	-0.3207	-0.469
0.0999	0.82484	0.588	-0.4605	-0.183	0.4998	0.94857	1.646	-0.6765	-0.630	0.8999	1.00110	3.507	-0.1869	-0.275
0.1999	0.86732	0.783	-0.5482	-0.364	0.6001	0.96528	1.990	-0.5777	-0.663	1.0000	1.00956	4.158	0.0000	0.000
0.2998	0.90114	1.011	-0.6587	-0.512	0.6998	0.97959	2.437	-0.4992	-0.591					
$T = 328.15$ K														
0.0000	0.75785	0.372	0.0000	0.000	0.3999	0.92328	1.180	-0.6917	-0.450	0.7999	0.98718	2.551	-0.3295	-0.338
0.0999	0.82044	0.533	-0.4714	-0.153	0.4998	0.94475	1.459	-0.6993	-0.486	0.8999	0.99741	2.995	-0.1888	-0.208
0.1999	0.86314	0.720	-0.5661	-0.281	0.6001	0.96159	1.788	-0.6031	-0.472	1.0000	1.00593	3.518	0.0000	0.000
0.2998	0.89717	0.933	-0.6841	-0.382	0.6998	0.97607	2.144	-0.5322	-0.430					

were calculated from composition–density data with an uncertainty better than $\pm 0.002 \text{ cm}^3\cdot\text{mol}^{-1}$. 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 controlled to within ± 0.01 K. An

electronic digital stopwatch with a readability of ± 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 η of the liquid was then calculated from the following relationship:

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

Table 3. Coefficients of the Redlich–Kister Equation and Standard Deviation for Excess Molar Volumes and Viscosity Deviations of Mixtures

<i>T</i> /K	property	<i>A</i> ₀	<i>A</i> ₁	<i>A</i> ₂	<i>A</i> ₃	<i>A</i> ₄	σ
(x) <i>m</i> -Cresol + (1 - <i>x</i> ₁) Ethylene Glycol							
308.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-0.265	0.522	0.202	-0.642		0.0467
	$\Delta\eta$ /mPa·s	6.886	-9.616	1.958	5.088	-8.203	0.1744
313.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-0.253	0.505	0.206	-0.658		0.0478
	$\Delta\eta$ /mPa·s	5.093	-7.217	1.889	5.094	-6.767	0.2656
318.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-0.245	0.471	0.195	-0.626		0.0495
	$\Delta\eta$ /mPa·s	3.644	-5.092	2.869	3.449	-7.084	0.1338
323.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-0.234	0.447	0.175	-0.621		0.0472
	$\Delta\eta$ /mPa·s	2.807	-3.845	1.221	2.314	-4.056	0.1439
333.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-0.215	0.391	0.145	-0.570		0.0486
	$\Delta\eta$ /mPa·s	1.832	-2.216	-0.771	0.817	0.854	0.1282
343.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-0.207	0.348	0.119	-0.590		0.0474
	$\Delta\eta$ /mPa·s	1.087	-1.539	0.626	1.639	-1.933	0.0442
353.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-0.191	0.283	0.080	-0.569		0.0409
	$\Delta\eta$ /mPa·s	0.644	-0.956	0.438	0.848	-1.220	0.0553
(x) <i>m</i> -Cresol + (1 - <i>x</i> ₁) Methanol							
308.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-2.406	0.522	0.971	1.903	-3.582	0.1654
	$\Delta\eta$ /mPa·s	-7.386	-2.906				0.3211
313.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-2.496	0.535	0.884	1.951	-3.484	0.1776
	$\Delta\eta$ /mPa·s	-5.057	-1.601				0.1150
318.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-2.609	0.479	0.942	2.131	-3.538	0.1784
	$\Delta\eta$ /mPa·s	-3.689	-0.980				0.0679
323.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-2.700	0.561	1.121	2.048	-3.894	0.1417
	$\Delta\eta$ /mPa·s	-2.597	-0.582				0.0634
328.15	<i>V</i> ^E /cm ³ ·mol ⁻¹	-2.809	0.532	1.070	2.200	-3.712	0.1598
	$\Delta\eta$ /mPa·s	-1.948	-0.335				0.0455

where *t* is the flow time; ν is the kinematic viscosity; and *k* and θ are the viscometer constant and the Hagenbach correction factor, respectively.

According to the viscosity of the mixtures, two types of viscometers at different diameters and lengths were used. For the system of *m*-cresol + ethylene glycol, the viscometer with the capillaries (0.8 to 0.9) mm in diameter and 100 mm in length was used. For the system of *m*-cresol + methanol, one of the capillaries was (0.8 to 0.9) mm in diameter and 100 mm in length for molar fraction of *m*-cresol from (0.6 to 1.0); the other one was (0.2 to 0.3) mm in diameter and 120 mm in length for molar fraction from (0 to 0.5). The calibration of the viscometer was carried out with double-distilled water and standard oil (supplied by the National Bureau of Standards). During the heating, to minimize the evaporation losses, the viscometer's limbs are closed with Teflon caps. During the measurements of flow time, the caps of the limbs were removed. The overall uncertainty of the viscosity measurements is dependent on the

equilibrium stability of the viscometer, the time of the flow, and the change of concentration, which are of the order of 1×10^{-2} , 1×10^{-2} , and 3×10^{-4} , respectively. The uncertainty of viscosity results was within ± 0.003 mPa·s.

In the experiment, the density and viscosity for one composition sample were measured at different temperatures. In Table 1, the pure components are compared with recent published density and dynamic viscosity values.

Result and Discussion

Excess volumes and viscosity deviations were calculated from our measurements according to the following equations:¹²

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

where *x*₁ and *x*₂ are mole fractions; *M*₁ and *M*₂ are the molar

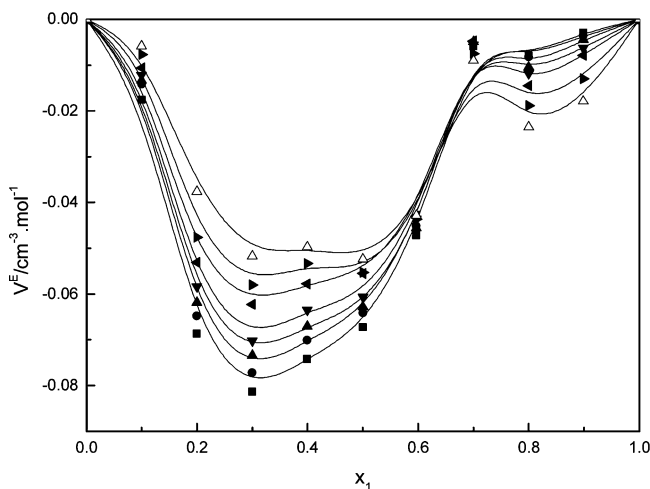


Figure 1. Excess volumes, *V*^E, vs the mole fraction, *x*₁, for system *m*-cresol (1) + ethylene glycol (2) at different temperatures *T*: ■, 308.15 K; ●, 313.15 K; ▲, 318.15 K; ▼, 323.15 K; left-facing solid triangle, 333.15 K; right-facing solid triangle, 343.15 K; △, 353.15 K; solid curves, calculated with Redlich–Kister equations; symbols, experimental values.

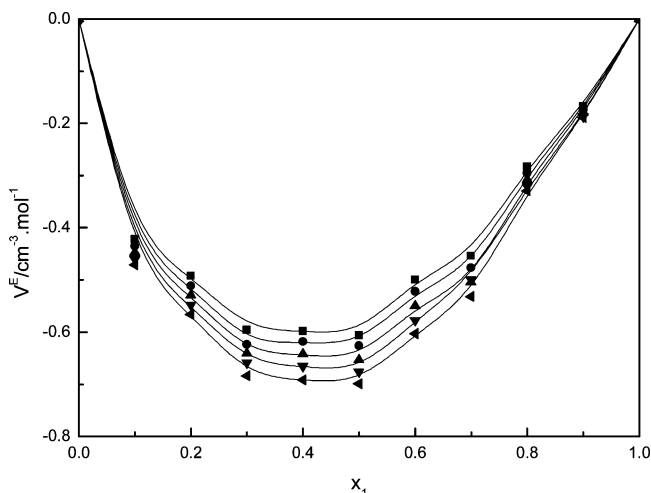


Figure 2. Excess volumes, *V*^E, vs the mole fraction, *x*₁, for system *m*-cresol (1) + methanol (2) at different temperatures *T*: ■, 308.15 K; ●, 313.15 K; ▲, 318.15 K; ▼, 323.15 K; left-facing solid triangle, 328.15 K; solid curves, calculated with Redlich–Kister equations; symbols, experimental values.

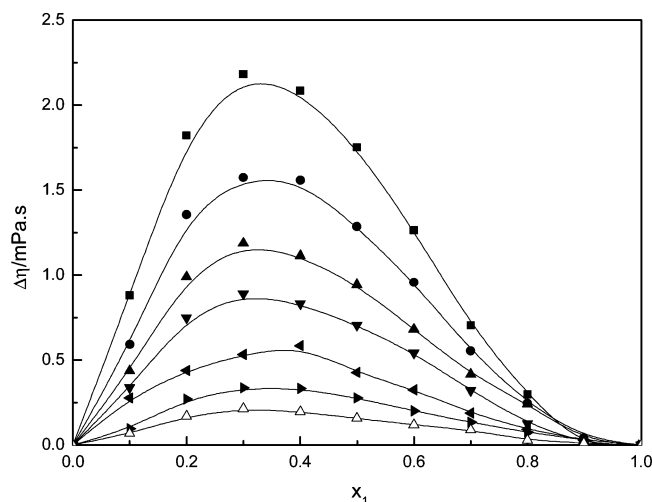


Figure 3. Deviation of viscosity, $\Delta\eta$, vs the mole fraction, x_1 , for system *m*-cresol (1) + ethylene glycol (2) at different temperatures T : ■, 308.15 K; ●, 313.15 K; ▲, 318.15 K; ▼, 323.15 K; left-facing solid triangle, 333.15 K; right-facing solid triangle, 343.15 K; △, 353.15 K; solid curves, calculated with Redlich–Kister equations; symbols, experimental values.

masses; and ρ_1 and ρ_2 are the densities of pure components 1 and 2, respectively. Quantities without subscripts refer to the mixture.

The viscosity deviations were calculated from the following relation:¹³

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

where η is the viscosity of mixtures and η_1 and η_2 are the viscosity of components 1 and 2, respectively. The values of V^E and $\Delta\eta$ for each mixture were fitted to the Redlich–Kister equation:¹⁴

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

where $Y = V^E$ or $\Delta\eta$, A_i are adjustable parameters, and x_1 is the 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 derivation:

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

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

The excess molar volume data and the viscosity deviations are presented in Table 2. Table 3 lists the values of the parameters A_i together with the standard deviations.

The V^E against x_1 plots of *m*-cresol with ethylene glycol or methanol are shown in Figure 1 and Figure 2, respectively. It can be seen from Figure 1 that V^E values for the *m*-cresol + ethylene glycol over the whole composition range are negative. The absolute values of V^E are small and decrease with temperature rising, and variations of V^E with molar fraction are not regular. For the system of *m*-cresol + methanol, the excess molar volumes are negative too and parabolic composition dependence is found, but absolute values of V^E are large and increase with temperature rising.

Figure 3 shows viscosity deviations for the binary mixture of *m*-cresol with ethylene glycol, plotted against mole fraction together with the fitted curve, obtained from the Redlich–Kister

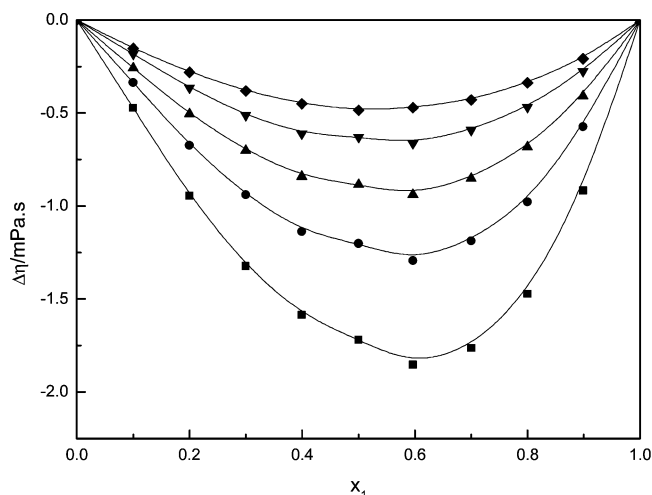


Figure 4. Deviation of viscosity, $\Delta\eta$, vs the mole fraction, x_1 , for system *m*-cresol (1) + methanol (2) at different temperatures T : ■, 308.15 K; ●, 313.15 K; ▲, 318.15 K; ▼, 323.15 K; left-facing solid triangle, 328.15 K; solid curves, calculated with Redlich–Kister equations; symbols, experimental values.

equation. The viscosity deviations at selected temperatures are positive over the entire composition, and the maximum occur about at $x = 0.3$. $\Delta\eta$ values decrease with temperature rising.

Figure 4 shows the relation of the viscosity deviations with mole fraction for the system *m*-cresol with methanol. The viscosity deviations are negative, and the curves are skewed to the *m*-cresol-rich range.

Conclusion

Densities and viscosities for the *m*-cresol + (ethylene glycol or methanol) binary systems have been experimentally determined over several temperatures and at normal atmospheric pressure. The excess molar volume and viscosity deviations were correlated using the Redlich–Kister polynomial equation. Excess molar volumes and viscosity deviation show a systematic change with increasing temperature. The excess molar volumes for these two binary systems were negative over the whole composition range and at all temperatures. The deviations of viscosity for *m*-cresol + ethylene glycol were positive, and the values for *m*-cresol + methanol are negative.

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