

# Thermodynamic Properties of Binary Mixtures of *p*-Xylene with Cyclohexane, Heptane, Octane, and *N*-Methyl-2-pyrrolidone at Several Temperatures

Changsheng Yang, Wei Xu, and Peisheng Ma\*

Key Laboratory for Green Chemical Technology of State Education Ministry, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, PR China

Densities and viscosities for *p*-xylene with cyclohexane, heptane, octane, and *N*-methyl-2-pyrrolidone were determined from (298.15 to 353.15) K and at atmospheric pressure. The measurements were carried out over the whole range of composition, using a vibrating-tube density meter and a Ubbelohde viscometer. Density and viscosity measurements were used to compute the excess molar volumes,  $V^E$ , and viscosity deviations,  $\Delta\eta$ . The excess molar volumes,  $V^E$ , and viscosity deviations,  $\Delta\eta$ , have been fit to the Redlich–Kister equation, and the coefficients and estimates of the standard error values are given.

## Introduction

Studies on the 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 liquid mixture constituents. Accurate experimental data on the density and viscosity of organic liquids and their binary mixtures are needed in many engineering disciplines. Therefore, density,  $\rho$ , and viscosity,  $\eta$ , data of binary mixtures are important from both a practical and theoretical viewpoint.

This is a part of our ongoing program of research on the thermodynamic and transport properties of binary mixtures containing *p*-xylene. This paper presents new experimental data of the density and viscosity from (303.15 to 323.15) K and atmospheric pressure for binary mixtures of *p*-xylene with cyclohexane, heptane, octane, and *N*-methyl-2-pyrrolidone over the whole composition range. 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.

## Experimental Section

**Materials.** All chemicals used in this study were obtained from Tianjin Reagent Company and purified by distillation in which the middle fraction was collected, except for *N*-methyl-2-pyrrolidone. The liquids were dried over 0.4-nm molecular sieves and partially degassed by ultrasound prior to their experimental use. The mass fraction purities tested by gas chromatography were as follows: *p*-xylene (>0.998), cyclohexane (>0.999), heptane (>0.995), and octane (>0.993). The purity of the chemicals used was confirmed by comparing the densities and viscosities with those reported in the literature. In Table 1, we show the densities and viscosities determined in this study as well as those available in the literature, for comparison.

**Apparatus and Procedure.** The densities of the pure components and their mixtures were measured with a high-

**Table 1. Comparison of Experimental and Literature Values of Densities,  $\rho$ , and Viscosities,  $\eta$ , for Pure Compounds**

liquid	T/K	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\eta/\text{m}\cdot\text{Pas}$	
		exptl	lit	exptl	lit
<i>p</i> -xylene	298.15	0.85670	0.85648 <sup>1</sup>	0.6078	0.6045 <sup>3</sup>
			0.85672 <sup>2</sup>		0.6031 <sup>5</sup>
			0.8565 <sup>3</sup>		0.6110 <sup>4</sup>
			0.65663 <sup>4</sup>		
	303.15	0.85235	0.8522 <sup>3</sup>	0.5715	0.5762 <sup>2</sup>
			0.85239 <sup>4</sup>		0.5694 <sup>3</sup>
cyclohexane	313.15	0.84364	0.8434 <sup>3</sup>	0.5080	0.5064 <sup>3</sup>
			0.85239 <sup>2</sup>		
	323.15	0.83490		0.4599	0.4580 <sup>5</sup>
	298.15	0.77382	0.77381 <sup>6</sup>	0.9027	0.9062 <sup>6</sup>
			0.77385 <sup>7</sup>		0.892 <sup>8</sup>
	303.15	0.76910	0.7692 <sup>8</sup>	0.8205	0.813 <sup>8</sup>
heptane	313.15	0.75960	0.7596 <sup>9</sup>	0.6963	0.682 <sup>9</sup>
			0.7595 <sup>10</sup>		0.7027 <sup>10</sup>
	323.15	0.74998	0.75024 <sup>11</sup>	0.5984	0.6051 <sup>11</sup>
			0.7496 <sup>12</sup>		0.6033 <sup>12</sup>
	333.15	0.74023	0.70465 <sup>11</sup>	0.5195	0.5291 <sup>11</sup>
			0.7399 <sup>12</sup>		0.5277 <sup>12</sup>
octane	298.15	0.67990	0.67947 <sup>6</sup>	0.4009	0.4022 <sup>6</sup>
			0.67953 <sup>13</sup>		0.388 <sup>14</sup>
			0.6793 <sup>14</sup>		
	303.15	0.67565	0.67528 <sup>13</sup>	0.3750	0.3691 <sup>3</sup>
			0.6750 <sup>14</sup>		0.3681 <sup>4</sup>
	313.15	0.66708	0.66666 <sup>13</sup>	0.3373	0.3351 <sup>3</sup>
<i>N</i> -methyl-2-pyrrolidone			0.6663 <sup>14</sup>		0.3331 <sup>4</sup>
	323.15	0.65837	0.6574 <sup>15</sup>	0.3074	0.303 <sup>15</sup>
	333.15	0.64953		0.2793	0.2760 <sup>16</sup>
	343.15	0.64047	0.6384 <sup>15</sup>	0.2541	0.2525 <sup>15</sup>
	298.15	0.69988	0.69856 <sup>13</sup>	0.5174	0.5061 <sup>4</sup>
			0.6984 <sup>14</sup>		
	303.15	0.69570	0.69452 <sup>13</sup>	0.4843	0.4813 <sup>13</sup>
			0.6943 <sup>14</sup>		0.4771 <sup>14</sup>
	313.15	0.68746	0.68634 <sup>13</sup>	0.4318	0.431 <sup>13</sup>
			0.6862 <sup>14</sup>		0.4251 <sup>14</sup>
	298.15	1.02794	1.0279 <sup>17</sup>	1.6832	1.687 <sup>17</sup>
			1.02851 <sup>18</sup>		1.656 <sup>21</sup>
	303.15	1.02347	1.02340 <sup>19</sup>	1.5544	
	313.15	1.01455	1.0157 <sup>20</sup>	1.3321	1.322 <sup>21</sup>
			1.01519 <sup>21</sup>		
	323.15	1.00566	1.0054 <sup>20</sup>	1.1602	1.175 <sup>21</sup>
			1.00627 <sup>21</sup>		
	333.15	0.99671	0.9974 <sup>20</sup>	1.0222	1.035 <sup>21</sup>
			0.99741 <sup>21</sup>		
	343.15	0.98775	0.98846 <sup>21</sup>	0.9084	0.921 <sup>21</sup>

\* To whom correspondence should be addressed: E-mail: tjuyangchsh@yahoo.com.cn. Fax: 022-27403389. Tel: 022-27890907.

**Table 2. Densities,  $\rho$ , Viscosities,  $\eta$ , Excess Molar Volumes,  $V^E$ , and Viscosity Deviations,  $\Delta\eta$ , for the Binary Mixtures from 298.15 K to 353.15 K**

$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	$\Delta\eta/\text{mPa}\cdot\text{s}$	$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	$\Delta\eta/\text{mPa}\cdot\text{s}$
$(x)p\text{-Xylene} + (1 - x_1)\text{Cyclohexane}$									
$T = 298.15 \text{ K}$									
0.0000	0.77382	0.9027	0.0000	0.000	0.5431	0.81738	0.6326	0.5892	-0.114
0.0809	0.77958	0.8232	0.2532	-0.056	0.6491	0.82654	0.6219	0.5008	-0.095
0.1655	0.78602	0.7570	0.4354	-0.098	0.7603	0.83616	0.6175	0.3707	-0.067
0.2537	0.79296	0.7177	0.5679	-0.112	0.8771	0.84617	0.6139	0.2097	-0.037
0.3457	0.80058	0.6773	0.6243	-0.126	1.0000	0.8567	0.6162	0.0000	0.000
0.4422	0.80868	0.6575	0.6387	-0.119					
$T = 303.15 \text{ K}$									
0.0000	0.76910	0.8205	0.0000	0.000	0.5431	0.81289	0.5868	0.5903	-0.098
0.0809	0.77492	0.7475	0.2514	-0.053	0.6491	0.82209	0.5787	0.5008	-0.080
0.1655	0.78139	0.6992	0.4357	-0.080	0.7603	0.83171	0.5726	0.3755	-0.059
0.2537	0.78836	0.6524	0.5697	-0.105	0.8771	0.84179	0.5701	0.2091	-0.032
0.3457	0.79602	0.6311	0.6256	-0.103	1.0000	0.85235	0.5715	0.0000	0.000
0.4422	0.80415	0.6059	0.6408	-0.105					
$T = 313.15 \text{ K}$									
0.0000	0.75961	0.6963	-0.0015	0.000	0.5431	0.80391	0.5185	0.5873	-0.076
0.0809	0.7655	0.6401	0.2543	-0.041	0.6491	0.81316	0.5108	0.5009	-0.063
0.1655	0.77207	0.5981	0.4372	-0.067	0.7603	0.82286	0.5110	0.3744	-0.042
0.2537	0.77914	0.5739	0.5691	-0.075	0.8771	0.833	0.5052	0.2099	-0.026
0.3457	0.78688	0.5481	0.6246	-0.083	1.0000	0.84364	0.5080	0.0000	0.000
0.4422	0.7951	0.5313	0.6377	-0.082					
$T = 323.15 \text{ K}$									
0.0000	0.74998	0.5984	0.0000	0.000	0.5431	0.79486	0.4638	0.5837	-0.059
0.0809	0.75599	0.5594	0.2538	-0.028	0.6491	0.80419	0.4580	0.4974	-0.051
0.1655	0.76267	0.5260	0.4348	-0.049	0.7603	0.81398	0.4569	0.3697	-0.036
0.2537	0.76983	0.5037	0.5673	-0.059	0.8771	0.82418	0.4567	0.2090	-0.020
0.3457	0.77765	0.4861	0.6236	-0.065	1.0000	0.83490	0.4599	0.0000	0.000
0.4422	0.78598	0.4722	0.6326	-0.065					
$T = 333.15 \text{ K}$									
0.0000	0.74023	0.5195	0.0000	0.000	0.5431	0.78572	0.4186	0.5784	-0.052
0.0809	0.74636	0.4874	0.2523	-0.025	0.6491	0.79514	0.4131	0.4918	-0.048
0.1655	0.75314	0.4646	0.4337	-0.040	0.7603	0.805	0.4109	0.3670	-0.040
0.2537	0.76042	0.4520	0.5627	-0.045	0.8771	0.8153	0.4124	0.2051	-0.028
0.3457	0.76833	0.4338	0.6191	-0.055	1.0000	0.82609	0.4298	0.0000	0.000
0.4422	0.77674	0.4230	0.6295	-0.057					
$(x)p\text{-Xylene} + (1 - x_1)\text{Heptane}$									
$T = 298.15 \text{ K}$									
0.0000	0.5431	0.4009	0.0000	0.000	0.5431	0.77651	0.4807	-0.0889	-0.046
0.0948	0.6491	0.4074	-0.0232	-0.014	0.6491	0.79516	0.5047	-0.0761	-0.044
0.1911	0.7603	0.4176	-0.0383	-0.024	0.7603	0.81474	0.5331	-0.0619	-0.038
0.2880	0.8771	0.4295	-0.0522	-0.033	0.8771	0.83524	0.5714	-0.0315	-0.022
0.3865	1.0000	0.4436	-0.0703	-0.041	1.0000	0.8567	0.6162	0.0000	0.000
0.4856	0.5431	0.4598	-0.0724	-0.046					
$T = 303.15 \text{ K}$									
0.0000	0.67565	0.3750	0.0000	0.0000	0.5861	0.77219	0.4496	-0.0962	-0.041
0.0948	0.69005	0.3820	-0.0243	-0.012	0.6877	0.79083	0.4716	-0.0824	-0.039
0.1911	0.70512	0.3912	-0.0439	-0.021	0.7906	0.81042	0.4979	-0.0696	-0.032
0.2880	0.72075	0.4019	-0.0577	-0.030	0.8949	0.83091	0.5308	-0.0364	-0.020
0.3865	0.73720	0.4154	-0.0771	-0.036	1.0000	0.85235	0.5715	0.0000	0.000
0.4856	0.75424	0.4304	-0.0816	-0.040					
$T = 313.15 \text{ K}$									
0.0000	0.66708	0.3373	0.0000	0.000	0.5431	0.76352	0.4034	-0.1136	-0.034
0.0948	0.68145	0.3440	-0.0280	-0.010	0.6491	0.78215	0.4234	-0.0981	-0.031
0.1911	0.69652	0.3523	-0.0563	-0.018	0.7603	0.80171	0.4440	-0.0787	-0.028
0.2880	0.71212	0.3623	-0.0710	-0.024	0.8771	0.82221	0.4729	-0.0435	-0.017
0.3865	0.72857	0.3740	-0.0960	-0.029	1.0000	0.84364	0.5080	0.0000	0.000
0.4856	0.74557	0.3865	-0.0966	-0.034					
$T = 323.15 \text{ K}$									
0.0000	0.65837	0.3074	0.0000	0.000	0.5861	0.75477	0.3683	-0.1337	-0.029
0.0948	0.67273	0.3136	-0.0349	-0.008	0.6877	0.77341	0.3835	-0.1177	-0.028
0.1911	0.68779	0.3211	-0.0687	-0.016	0.7906	0.79297	0.4041	-0.0940	-0.024
0.2880	0.70338	0.3304	-0.0869	-0.021	0.8949	0.81345	0.4285	-0.0490	-0.015
0.3865	0.71983	0.3408	-0.1160	-0.026	1.0000	0.83490	0.4599	0.0000	0.000
0.4856	0.73682	0.3529	-0.1164	-0.029					

**Table 2. (Continued)**

$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	$\Delta\eta/\text{mPa}\cdot\text{s}$	$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	$\Delta\eta/\text{mPa}\cdot\text{s}$
$(x)p\text{-Xylene} + (1 - x_1)\text{Heptane}$									
$T = 333.15 \text{ K}$									
0.0000	0.64953	0.2793	0.0000	0.000	0.5431	0.74592	0.3345	-0.1558	-0.033
0.0948	0.66388	0.2848	-0.0417	-0.009	0.6491	0.76457	0.3537	-0.1379	-0.029
0.1911	0.67895	0.2922	-0.0850	-0.016	0.7603	0.78415	0.3712	-0.1117	-0.027
0.2880	0.69452	0.2997	-0.1038	-0.023	0.8771	0.80464	0.3973	-0.0597	-0.017
0.3865	0.71098	0.3097	-0.1384	-0.028	1.0000	0.82609	0.4298	0.0000	0.000
0.4856	0.72797	0.3201	-0.1394	-0.032					
$T = 343.15 \text{ K}$									
0.0000	0.64047	0.2541	0.0000	0.000	0.5431	0.73691	0.3030	-0.1815	-0.024
0.0948	0.65483	0.2612	-0.0524	-0.005	0.6491	0.75557	0.3166	-0.1592	-0.023
0.1911	0.66989	0.2658	-0.0996	-0.012	0.7603	0.77517	0.3317	-0.1278	-0.021
0.2880	0.68548	0.2730	-0.1263	-0.017	0.8771	0.79569	0.3498	-0.0691	-0.015
0.3865	0.70194	0.2818	-0.1626	-0.020	1.0000	0.81717	0.3783	0.0000	0.000
0.4856	0.71894	0.2915	-0.1643	-0.023					
$(x)p\text{-Xylene} + (1 - x_1)\text{Octane}$									
$T = 298.15 \text{ K}$									
0.0000	0.69990	0.5206	0.0000	0.000	0.6174	0.78605	0.5393	0.0329	-0.040
0.1068	0.71290	0.5185	0.0114	-0.012	0.7152	0.80261	0.5515	0.0252	-0.037
0.2121	0.72639	0.5172	0.0260	-0.024	0.8113	0.81983	0.5678	0.0156	-0.030
0.3156	0.74039	0.5216	0.0348	-0.029	0.9065	0.83791	0.5905	0.0056	-0.017
0.4178	0.75501	0.5273	0.0378	-0.033	1.0000	0.85670	0.6162	0.0000	0.000
0.5184	0.77021	0.5297	0.0380	-0.040					
$T = 303.15 \text{ K}$									
0.0000	0.69572	0.4837	0.0000	0.000	0.6174	0.78176	0.5020	0.0297	-0.036
0.1068	0.70871	0.4829	0.0080	-0.010	0.7152	0.79830	0.5139	0.0224	-0.033
0.2121	0.72217	0.4825	0.0239	-0.020	0.8113	0.81551	0.5299	0.0137	-0.025
0.3156	0.73615	0.4833	0.0332	-0.028	0.9065	0.83352	0.5489	0.0085	-0.014
0.4178	0.75077	0.4919	0.0366	-0.028	1.0000	0.85235	0.5715	0.0000	0.000
0.5184	0.76593	0.4930	0.0367	-0.036					
$T = 313.15 \text{ K}$									
0.0000	0.68750	0.4306	0	0	0.6174	0.77323	0.4483	0.0262	-0.030
0.1068	0.70044	0.4298	0.0059	-0.009	0.7152	0.78972	0.4586	0.0193	-0.027
0.2121	0.71385	0.4298	0.0209	-0.017	0.8113	0.80689	0.4722	0.0115	-0.021
0.3156	0.72778	0.4331	0.0291	-0.022	0.9065	0.82489	0.4895	0.0053	-0.011
0.4178	0.74231	0.4373	0.0336	-0.026	1.0000	0.84364	0.5080	0.0000	0.000
0.5184	0.75745	0.4418	0.0340	-0.029					
$T = 323.15 \text{ K}$									
0.0000	0.67937	0.3864	0.0000	0.000	0.6174	0.76473	0.4057	0.0227	-0.026
0.1068	0.69226	0.3877	0.0017	-0.007	0.7152	0.78117	0.4143	0.0145	-0.025
0.2121	0.70560	0.3903	0.0173	-0.012	0.8113	0.79831	0.4269	0.0105	-0.019
0.3156	0.71947	0.3901	0.0245	-0.020	0.9065	0.81620	0.4409	0.0053	-0.012
0.4178	0.73393	0.3952	0.0313	-0.022	1.0000	0.83490	0.4599	0.0000	0.000
0.5184	0.74901	0.3997	0.0298	-0.025					
$T = 333.15 \text{ K}$									
0.0000	0.67102	0.3495	0.0000	0.000	0.6174	0.75608	0.3753	0.0191	-0.024
0.1068	0.68383	0.3528	0.0077	-0.005	0.7152	0.77247	0.3858	0.0131	-0.021
0.2121	0.69715	0.3552	0.0140	-0.011	0.8113	0.78952	0.3961	0.0084	-0.019
0.3156	0.71096	0.3571	0.0226	-0.018	0.9065	0.80743	0.4138	0.0031	-0.008
0.4178	0.72538	0.3617	0.0270	-0.021	1.0000	0.82609	0.4298	0.0000	0.000
0.5184	0.74041	0.3671	0.0252	-0.024					
$T = 343.15 \text{ K}$									
0.0000	0.66254	0.3139	0.0000	0.000	0.6174	0.74730	0.3329	0.0164	-0.021
0.1068	0.67529	0.3167	0.0081	-0.004	0.7152	0.76364	0.3410	0.0123	-0.019
0.2121	0.68857	0.3164	0.0119	-0.011	0.8113	0.78065	0.3481	0.0075	-0.018
0.3156	0.70232	0.3185	0.0201	-0.016	0.9065	0.79853	0.3588	0.0037	-0.013
0.4178	0.71669	0.3223	0.0248	-0.019	1.0000	0.81717	0.3783	0.0000	0.000
0.5184	0.73168	0.3271	0.0228	-0.020					
$T = 353.15 \text{ K}$									
0.0000	0.65380	0.2840	0.0000	0.000	0.6174	0.73837	0.3016	0.0144	-0.016
0.1068	0.66651	0.2856	0.0065	-0.004	0.7152	0.75470	0.3113	0.0103	-0.011
0.2121	0.67975	0.2877	0.0112	-0.008	0.8113	0.77170	0.3174	0.0060	-0.011
0.3156	0.69348	0.2897	0.0172	-0.011	0.9065	0.78957	0.3264	0.0040	-0.007
0.4178	0.70782	0.2939	0.0205	-0.013	1.0000	0.80822	0.3382	0.0000	0.000
0.5184	0.72278	0.2984	0.0198	-0.014					

**Table 2. (Continued)**

$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	$\Delta\eta/\text{mPa}\cdot\text{s}$	$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\eta/\text{mPa}\cdot\text{s}$	$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	$\Delta\eta/\text{mPa}\cdot\text{s}$
$(x)p\text{-Xylene} + (1 - x_1)N\text{-Methyl-2-pyrrolidone}$									
$T = 303.15 \text{ K}$									
0.0000	1.02347	1.5544	0.0000	0	0.5836	0.91947	0.8327	-0.7443	-0.148
0.0947	1.00571	1.3594	-0.2502	-0.102	0.6854	0.90273	0.7753	-0.6887	-0.105
0.1893	0.98837	1.2386	-0.4562	-0.130	0.7888	0.88585	0.7001	-0.5374	-0.079
0.2858	0.97107	1.1290	-0.6191	-0.144	0.8987	0.86904	0.6291	-0.3081	-0.047
0.3838	0.95382	1.0195	-0.7271	-0.158	1.0000	0.85235	0.5715	0.0000	0
0.4830	0.93656	0.9203	-0.7622	-0.159					
$T = 313.15 \text{ K}$									
0.0000	1.01455	1.3321	0.0000	0.000	0.5836	0.91056	0.7296	-0.7507	-0.122
0.0947	0.99679	1.1732	-0.2549	-0.081	0.6854	0.89386	0.6862	-0.6953	-0.081
0.1893	0.97945	1.0701	-0.4643	-0.106	0.7888	0.87702	0.6190	-0.5418	-0.063
0.2858	0.96214	0.9754	-0.6282	-0.121	0.8987	0.86027	0.5579	-0.3113	-0.038
0.3838	0.9449	0.8894	-0.7376	-0.126	1.0000	0.84364	0.5080	0.0000	0.000
0.4830	0.92768	0.8070	-0.7757	-0.127					
$T = 323.15 \text{ K}$									
0.0000	1.00566	1.1602	0.0000	0.000	0.5836	0.90168	0.6521	-0.7626	-0.099
0.0947	0.98788	1.0234	-0.2586	-0.070	0.6854	0.88499	0.6084	-0.7043	-0.072
0.1893	0.97053	0.9373	-0.4712	-0.090	0.7888	0.86816	0.5517	-0.5453	-0.056
0.2858	0.95321	0.8571	-0.6367	-0.103	0.8987	0.85146	0.4998	-0.3125	-0.035
0.3838	0.93597	0.7795	-0.7472	-0.112	1.0000	0.83490	0.4599	0.0000	0.000
0.4830	0.91877	0.7084	-0.7867	-0.114					
$T = 333.15 \text{ K}$									
0.0000	0.99671	1.0222	0.0000	0.000	0.5836	0.89271	0.5828	-0.7721	-0.094
0.0947	0.97893	0.9019	-0.2647	-0.064	0.6854	0.87603	0.5437	-0.7109	-0.072
0.1893	0.96155	0.8316	-0.4788	-0.078	0.7888	0.85921	0.4947	-0.5459	-0.060
0.2858	0.94423	0.7602	-0.6474	-0.093	0.8987	0.84256	0.4504	-0.3108	-0.042
0.3838	0.92699	0.6940	-0.7592	-0.101	1.0000	0.82609	0.4298	0.0000	0.000
0.4830	0.9098	0.6321	-0.7991	-0.104					
$T = 343.15 \text{ K}$									
0.0000	0.98775	0.9084	0.0000	0	0.5836	0.88372	0.5173	-0.7901	-0.082
0.0947	0.96995	0.8009	-0.2705	-0.057	0.6854	0.86704	0.4921	-0.7258	-0.053
0.1893	0.95256	0.7371	-0.4897	-0.071	0.7888	0.85022	0.4476	-0.5548	-0.043
0.2858	0.93523	0.6810	-0.6618	-0.076	0.8987	0.83361	0.4081	-0.3171	-0.026
0.3838	0.91798	0.6209	-0.7753	-0.084	1.0000	0.81717	0.3783	0.0000	0
0.4830	0.90082	0.5686	-0.8197	-0.084					
$T = 353.15 \text{ K}$									
0.0000	0.97876	0.8127	0.0000	0.000	0.5836	0.87466	0.4761	-0.8035	-0.060
0.0947	0.96094	0.7219	-0.2766	-0.046	0.6854	0.85797	0.4466	-0.7344	-0.041
0.1893	0.94354	0.6637	-0.5011	-0.059	0.7888	0.84116	0.4072	-0.5582	-0.031
0.2858	0.92619	0.6119	-0.6758	-0.065	0.8987	0.82456	0.3726	-0.3129	-0.016
0.3838	0.90893	0.5610	-0.7910	-0.070	1.0000	0.80822	0.3382	0.0000	0.000
0.4830	0.89178	0.5127	-0.8374	-0.071					

precision vibrating-tube digital density meter (density-specific gravity meter DA 505, KEM, Japan) whose measurement cell temperature was controlled automatically within  $\pm 0.01 \text{ K}$  of the selected value. Before each series of measurements, we calibrated the instrument at atmospheric pressure with doubly 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 the 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  $\pm 0.01 \text{ mg}$ . The average uncertainty in the compositions (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 of 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 tem-

peratures using several Ubbelohde suspended-level viscometers. The viscometer was immersed in a well-stirred water bath (Lauda, Germany) with temperature control to within  $\pm 0.01 \text{ K}$ . The efflux time was measured with a handheld digital stopwatch capable of measuring time to within 0.01 s. Experiments were 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

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

where  $\eta$  is the absolute (dynamic) viscosity,  $\rho$  is the density,  $t$  is the flow time,  $\nu$  is the kinetic viscosity, and  $k$  and  $\theta$  are the viscometer constant and the Hagenbach correction factor, respectively.

The calibration of the viscometer was carried out with doubly distilled water and doubly distilled benzene. Care was taken to reduce evaporation during the measurements. The uncertainty in the values is within  $\pm 0.003 \text{ mPa}\cdot\text{s}$ .

In the experiment, the density and viscosity for one composition sample were measured at different tempera-

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

T/K	property	A <sub>0</sub>	A <sub>1</sub>	A <sub>2</sub>	A <sub>3</sub>	A <sub>4</sub>	$\sigma$
(x)p-Xylene + (1 - x <sub>1</sub> )Cyclohexane							
298.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	2.467	-0.970	0.123	0.147	0.240	0.0021
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.466	0.221	-0.149	0.057	0.049	0.0020
303.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	2.469	-0.961	0.213	0.135	0.061	0.0023
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.406	0.174	-0.025	0.101	-0.158	0.0015
313.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	2.460	-0.954	0.227	0.104	0.110	0.0022
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.315	0.158	-0.002	0.039	-0.172	0.0014
323.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	2.447	-0.973	0.194	0.138	0.177	0.0023
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.247	0.115	-0.071	0.005	0.038	0.0006
333.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	2.426	-0.969	0.221	0.109	0.116	0.0019
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.220	0.040	-0.016	-0.022	-0.170	0.0010
(x) p-Xylene + (1 - x <sub>1</sub> )Heptane							
298.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.319	-0.169	0.028	0.201	0.000	0.0019
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.244	0.016	0.482	-0.643	0.225	0.0004
303.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.348	-0.165	-0.013	0.156	0.051	0.0019
	$\Delta\eta/\text{mPa}\cdot\text{s}$	0.001	0.007	-0.251	0.203	-0.092	0.0004
313.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.416	-0.141	-0.030	0.096	0.099	0.0025
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.206	0.234	-0.212	0.079	0.001	0.0004
323.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.497	-0.168	-0.078	0.161	0.204	0.0026
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.212	0.498	-1.029	1.064	-0.419	0.0001
333.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.586	-0.167	-0.137	0.129	0.272	0.0031
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.214	0.419	-0.922	1.100	-0.488	0.0006
343.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-0.691	-0.169	-0.089	0.150	0.183	0.0029
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.220	0.658	-1.169	0.767	-0.062	0.0003
(x)p-Xylene + (1 - x <sub>1</sub> )Octane							
298.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	0.151	-0.048	-0.027	0.029	-0.100	0.0024
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.152	-0.063	-0.069	0.033	0.077	0.0006
303.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	0.146	-0.081	-0.087	0.142	-0.002	0.0066
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.137	-0.045	-0.029	0.010	0.044	0.0008
313.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	0.132	-0.071	-0.084	0.114	-0.045	0.0058
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.113	-0.044	-0.040	0.032	0.061	0.0002
323.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	0.118	-0.0715	-0.115	0.133	0.019	0.0026
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.100	-0.043	0.010	-0.005	-0.026	0.0005
333.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	0.103	-0.043	-0.125	0.023	0.087	0.0065
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.093	-0.040	-0.024	0.013	0.072	0.0006
343.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	0.0936	-0.0348	-0.143	0.018	0.151	0.0078
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.080	-0.009	-0.017	-0.095	-0.020	0.0003
353.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	0.080	-0.038	-0.120	0.038	0.1812	0.0029
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.057	-0.012	0.012	-0.015	-0.027	0.0005
(x)p-Xylene + (1 - x <sub>1</sub> )N-Methyl-2-pyrrolidone							
303.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-3.066	-0.170	-0.301	-0.056	0.434	0.0033
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.635	0.184	0.380	0.351	-1.080	0.0019
313.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-3.108	-0.141	-0.267	-0.082	0.374	0.0035
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.509	0.187	0.257	0.214	-0.782	0.0025
323.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-3.154	-0.139	-0.222	-0.051	0.350	0.0033
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.443	0.161	0.259	0.162	-0.761	0.0015
333.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-3.203	-0.116	-0.131	0.000	0.268	0.0036
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.411	0.098	0.246	0.103	-0.834	0.0015
343.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-3.281	-0.104	-0.056	-0.016	0.176	0.0035
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.339	0.098	0.240	0.202	-0.697	0.0019
353.15	V <sup>E</sup> /cm <sup>3</sup> ·mol <sup>-1</sup>	-3.344	-0.081	-0.024	0.064	0.252	0.0035
	$\Delta\eta/\text{mPa}\cdot\text{s}$	-0.271	0.119	0.143	0.154	-0.414	0.0013

tures. Densities and viscosities of pure compounds are reported in Table 1 together with the corresponding literature values.

## Results and Discussion

The experimental values of density and viscosity for binary mixtures at different temperatures and atmospheric pressure are listed in Table 2. Excess volumes were calculated from our measurements according to the following equation

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

where  $x_1$  and  $x_2$  are mole fractions,  $M_1$  and  $M_2$  are the molar

masses, and  $\rho_1$  and  $\rho_2$  are the densities of pure components 1 and 2, respectively. Quantities without subscripts refer to the mixture.

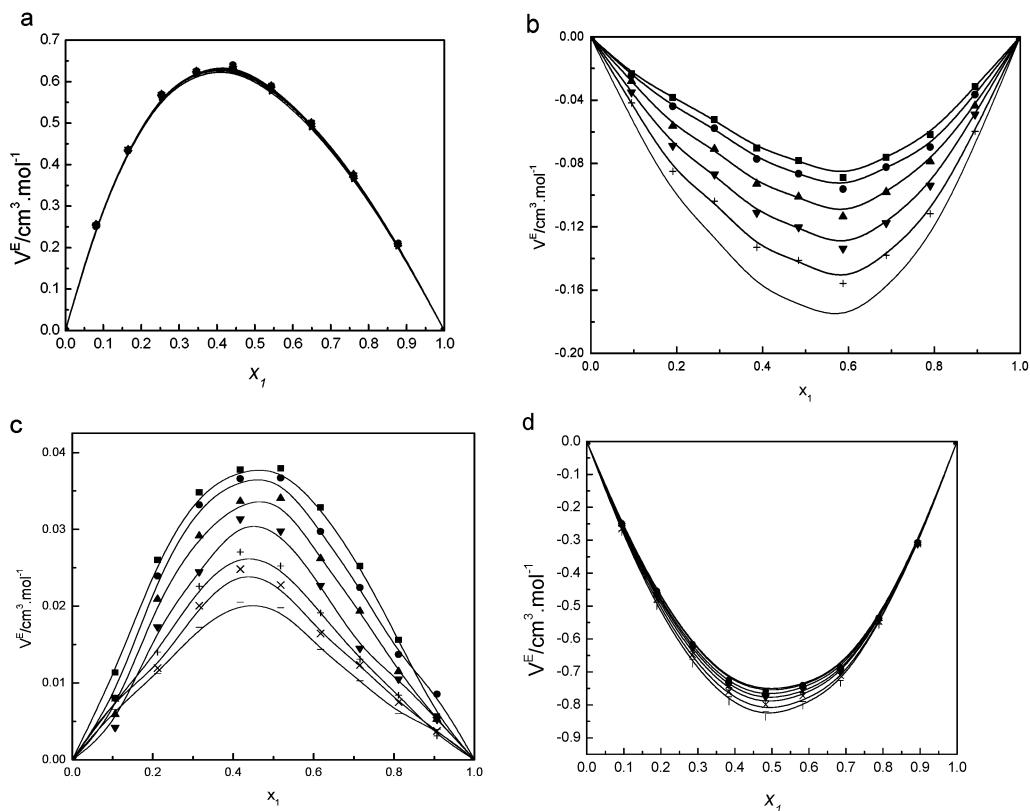
The viscosity deviations were calculated from the following relation

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

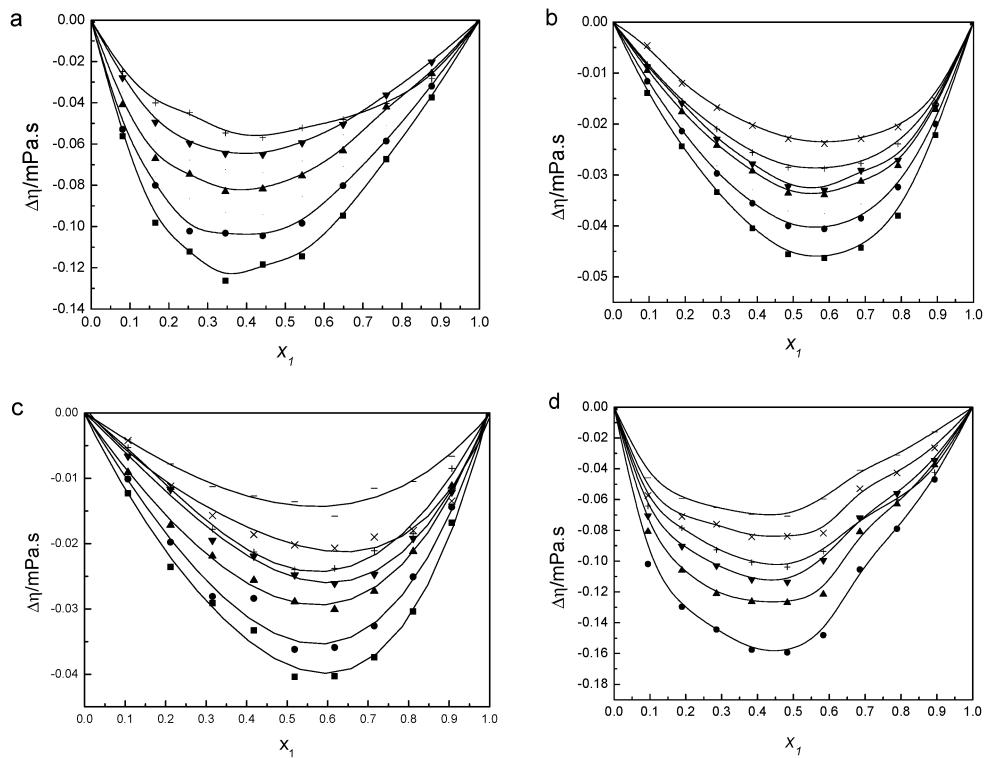
where  $\eta$  is the viscosity of the mixtures and  $\eta_1$  and  $\eta_2$  are the viscosities of components 1 and 2, respectively.

The values of  $V^E$  and  $\Delta\eta$  for each mixture were fit to the Redlich-Kister polynomial equation.

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



**Figure 1.** Excess volume,  $V^E$ , versus mole fraction,  $x_1$ , for *p*-xylene (1) + (a) cyclohexane (2), (b) heptane (2), (c) octane (2), and (d) *N*-methyl-2-pyrrolidone (2) at different temperatures  $T$ : ■, 298.15 K; ●, 303.15 K; ▲, 313.15 K; ▼, 323.15 K; +, 333.15 K; ×, 343.15 K; –, 353.15 K. Solid curves, calculated with the Redlich-Kister equation; symbols, experimental values.



**Figure 2.** Deviation of viscosity,  $\Delta\eta$ , versus mole fraction,  $x_1$ , for *p*-xylene (1) + (a) cyclohexane (2), (b) heptane (2), (c) octane (2), and (d) *N*-methyl-2-pyrrolidone (2) at different temperatures  $T$ : ■, 298.15 K; ●, 303.15 K; ▲, 313.15 K; ▼, 323.15 K; +, 333.15 K; ×, 343.15 K; –, 353.15 K. Solid curves, calculated with the Redlich-Kister equation; symbols, experimental values.

where  $Y = V^E$  or  $\Delta\eta$ ,  $A_i$  represents the 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 deviation

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

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, and they are shown graphically in Figures 1 and 2. Table 3 lists the values of the  $A_i$  parameters together with the standard deviations.

The variations of  $V^E$  and  $\Delta\eta$  with the mole fraction of *p*-xylene for the four systems studied at different temperatures are presented in Figures 1 and 2, respectively. It can be seen from Figure 1 that  $V^E$  values are positive for the *p*-xylene + cyclohexane, + octane system and negative for the *p*-xylene + heptane, *N*-methyl-2-pyrrolidone systems. The absolute  $V^E$  values for binary mixtures of *p*-xylene + octane, + heptane are much smaller than those for the other two systems. For *p*-xylene with cyclohexane and *N*-methyl-2-pyrrolidone, the absolute  $V^E$  values show a negligible temperature dependence. The dependence of excess molar volume on the temperature for the system of *p*-xylene with heptane decreases with increasing temperature. However, for the binary system of *p*-xylene with octane, the  $V^E$  values increase with temperature.

For *p*-xylene + alkane mixtures, there are no specific interactions between unlike molecules. The main contribution arises from the structure. Hence, the  $V^E$  values are very small for *p*-xylene + heptane, + octane. However, the excess molar volumes of the system *p*-xylene + heptane are negative, whereas those of the system *p*-xylene + octane are positive, which may be related to the different molecular packing due to the different molecular chain lengths of the alkanes. With the *p*-xylene + cyclohexane mixtures, the large, positive  $V^E$  values indicate a preponderance of the expansive effects associated with the rupture of the homomolecular interactions. It also suggests weak heteromolecular interactions.

Similar to self-association in pyridine and quinoline,<sup>22,23</sup> *N*-methyl-2-pyrrolidone is self-associating through hydrogen bonding. The negative values of  $V^E$  can be attributed to interactions of electron donor–acceptor type interactions between *p*-xylene and *N*-methyl-2-pyrrolidone.

From Figure 2, it can be seen that  $\Delta\eta$  values for the four binary systems analyzed in this work are negative over the entire range of composition and decrease in absolute value with increasing temperature. The  $\Delta\eta$  values for *p*-xylene with heptane and octane are almost identical and very small. The absolute value of the viscosity deviation is found to vary in the following order: *N*-methyl-2-pyrrolidone > cyclohexane > heptane ≈ octane.

## Conclusions

New experimental values of density and viscosity for the system of the binary mixtures of *p*-xylene + cyclohexane, heptane, octane, and *N*-methyl-2-pyrrolidone were determined from (298.15 to 353.15) K and atmospheric pressure. The excess molar volume and viscosity deviations were correlated using the Redlich–Kister polynomial equation. The excess molar volume,  $V^E$ , values are positive for the *p*-xylene + cyclohexane + octane system and negative for the *p*-xylene + heptane, *N*-methyl-2-pyrrolidone systems.

The viscosity deviations,  $\Delta\eta$ , are negative over the entire range of composition and decrease in absolute value with increasing temperature.

## Literature Cited

- (1) Yu, L.; Li, Y. Excess Molar Volumes of Sulfolane in Binary Mixtures with Six Aromatic Hydrocarbons at 298.15 K. *Fluid Phase Equilib.* **1998**, *147*, 207–213.
- (2) Serrano, L.; Silva, J. A.; Farelo, F. Densities and Viscosities of Binary and Ternary Liquid Systems Containing Xylene. *J. Chem. Eng. Data* **1990**, *35*, 288–291.
- (3) 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.
- (4) Serrano, L.; Silva, J. A.; Farelo, F. Densities and Viscosities of Binary and Ternary Liquid Systems Containing Xylenes. *J. Chem. Eng. Data* **1990**, *35*, 288–291.
- (5) Kashiwagi, H.; Makita, T. Viscosities of Twelve-Hydrocarbon Liquid in the Temperature Range 298–348 K at Pressures up to 110 MPa. *Int. J. Thermophys.* **1982**, *3*, 289–305.
- (6) Wu, J.; Asfour, A. Densities and Excess Molar Volumes of Eight n-Alkane Binary Systems at 293.15 and 298.15 K. *Fluid Phase Equilib.* **1994**, *102*, 305–315.
- (7) Benson, G. C.; Murakami, S.; Lam, V. T. Molar Excess Enthalpies and Volumes of Cyclohexane-isomeric Decalin Systems at 298.15 K. *Can. J. Chem.* **1970**, *48*, 211–218.
- (8) Aminabhavi, T. M.; Patil, V. B.; Aralaguppi, M. I.; Phayde, H. T. S. Density, Viscosity, and Refractive Index of the Binary Mixtures of Cyclohexane with Hexane, Heptane, Octane, Nonane, and Decane at (298.15, 303.15, and 308.15) K. *J. Chem. Eng. Data* **1996**, *41*, 521–525.
- (9) Aminabhavi, T. M.; Gopalakrishna, B. Density, Viscosity, Refractive Index, and Speed of Sound in Binary Mixtures of 2-Ethoxyethanol with n-Alkanes (C<sub>6</sub> to C<sub>12</sub>), 2,2,4-Trimethylpentane, and Cyclohexane in the Temperature Interval 298.15–313.15 K. *J. Chem. Eng. Data* **1995**, *40*, 632–641.
- (10) Dymond, J. H.; Young, K. J. Transport Properties of Nonelectrolyte Liquid Mixtures. V. Viscosity Coefficients for Binary Mixtures of Benzene Plus Alkanes at Saturation Pressure from 283 to 393. *Int. J. Thermophys.* **1981**, *2*, 237–247.
- (11) Padua, A. A. H.; Fareleira, J. M. N. A.; Calado, J. C. G. Validation of an Accurate Vibrating-Wire Densimeter: Density and Viscosity of Liquid Over Wide Range of Temperature and Pressures. *Int. J. Thermophys.* **1996**, *17*, 781–803.
- (12) Tanaka, Y.; Hosokawa, H.; Makita, T. Viscosity and Density of Binary Mixtures of Cyclohexane with n-Octane, n-Hexadecane under High Pressures. *Int. J. Thermophys.* **1991**, *12*, 245–264.
- (13) Matos, J. S.; Trenzado, J. L.; Gonzalez, E.; Alcalde, R. Volumetric Properties and Viscosities of the Methylbutanoate + n-Heptane + n-Octane Ternary System and Its Binary Constituents in the Temperature Range from 283.15 to 313.15 K. *Fluid Phase Equilib.* **2001**, *190*, 207–234.
- (14) Aminabhavi, T. M.; Aralaguppi, M. I.; Gopalakrishna, B.; Khinnavar, R. S. Densities, Shear Viscosities, Refractive Indices, and Speeds of Sound of Bis(2-methoxyethyl) Ether with Hexane, Heptane, Octane, and 2,2,4-Trimethylpentane in the Temperature Interval 298.15–318.15 K. *J. Chem. Eng. Data* **1994**, *39*, 522–528.
- (15) Baylauq, A.; Boned, C.; Dauge, P. Measurements of the Viscosity and Density of Three Hydrocarbons and the Three Associated Binary Mixture Versus Pressure and Temperature. *Int. J. Thermophys.* **1990**, *18*, 3–23.
- (16) Knapstad, B.; Skjøelsvik, P. A.; Oeye, H. A. Viscosity of Pure Hydrocarbons. *J. Chem. Eng. Data* **1989**, *34*, 37–43.
- (17) Pal, A.; Bhardwaj, R. K. Excess Molar Volumes and Viscosities for Binary Mixtures of 2-Propoxyethanol and of 2-Isopropoxyethanol with 2-Pyrrolidinone, *N*-Methyl-2-pyrrolidinone, *N,N*-Dimethylformamide, and *N,N*-Dimethylacetamide at 298.15 K. *J. Chem. Eng. Data* **2002**, *47*, 1128–1134.
- (18) Domańska, U.; Lachwa, J.; Letcher, T. M. Densities, Excess Molar Volumes, and Excess Molar Enthalpies of (*N*-Methyl-2-pyrrolidone + Ketone) at T = 298.15 K. *J. Chem. Eng. Data* **2002**, *47*, 1446–1452.
- (19) Kumari, P. G.; Radhamma, M.; Sekhar, G. C.; Rao, M. V. P. Excess Volumes and Speeds of Sound of *N*-Methyl-2-pyrrolidone with Chloroethanes and Chloroethenes at 303.15 K. *J. Chem. Eng. Data* **2002**, *47*, 425–427.
- (20) Aguila-Hernandez, J.; Gomez-Quintana, R.; Murrieta-Guevara, F.; Romero-Martinez, A.; Trejo, A. Liquid Density of Aqueous Blended Alkanolamines and *N*-Methylpyrrolidone as a Function of Concentration and Temperature. *J. Chem. Eng. Data* **2001**, *46*, 861–867.
- (21) Henni, A.; Hromek, J. J.; Tonitwachwuthikul, P.; Chakma, A. Volumetric Properties and Viscosities for Aqueous *N*-Methyl-2-pyrrolidone Solutions from 25 °C to 70 °C. *J. Chem. Eng. Data* **2004**, *49*, 231–234.

- (22) Ramachandran, D.; Rambabu, K.; Krishnan, K. M.; Venkateswarlu, P.; Raman, G. K. Volume of Mixing, Speed of Sound, and Viscosity of Butyl Acetate with Xylenes at 303.15 K. *J. Chem. Eng. Data* **1995**, *40*, 815–817.
- (23) Moreirus, A. F. Gorgia, J.; Lugo, L.; et al. Experimental Densities and Dynamic Viscosities of Organic Carbonate + n-Alkane or p-Xylene Systems at 298.15K. *Fluid Phase Equilib.* **2003**, *204*, 233–243.

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