Densities, Viscosities, and Refractive Indices for Binary and Ternary Mixtures of Acetone, Ethanol, and 2,2,4-Trimethylpentane

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Densities, viscosities, and refractive indices of the ternary system acetone + ethanol + 2,2,4-trimethylpentane at T = 298.15 K and the binary systems acetone + ethanol, acetone + 2,2,4-trimethylpentane, and ethanol + 2,2,4-trimethylpentane were measured at T = (288.15, 298.15, and 308.15) K and atmospheric pressure over the whole composition range. Densities were determined using a vibrating-tube density meter. Viscosities were measured with an automatic Ubbelohde capillary viscometer. Refractive indices were measured using a digital Abbe-type refractometer. Excess molar volumes V^E, deviations in the viscosity from the mole fraction average $\Delta\eta$, and deviations in the refractive index from the volume fraction average Δn_D for the mixtures were derived from experimental data. The binary and ternary data of V^E, $\Delta\eta$, and Δn_D were correlated with liquid composition by using the Redlich−Kister and Cibulka equations.

Introduction

The reformulation of gasoline includes certain oxygenated compounds such as alcohols and ethers. These oxygenated compounds are added to improve the octane rating and pollution-reducing capability of gasoline. This work has been carried out as a part of the project to investigate the thermodynamic behavior of liquid mixtures of the oxygenated compounds included in a hydrocarbon mixture.[1−4]

The objective of the present paper is to determine the densities, viscosities, and refractive indices for the binary and ternary systems formed by two oxygenated compounds, such as acetone and ethanol, as well as 2,2,4-trimethylpentane. The hydrocarbon liquid is the component that generally appears in gasoline. The experimental results are used to calculate excess molar volumes, deviations in the viscosity from the mole fraction average, and deviations in the refractive index from the volume fraction average. The calculated binary data have been fitted to the Redlich−Kister equation to determine the coefficients. For correlating the ternary data, the Cibulka equation was used. As far as we know, no such ternary data are available for the mixtures in the open literature.

Experimental Section

Materials. The chemicals used were of analytical grade, acetone (99.9%) and 2,2,4-trimethylpentane (99.7%) were obtained from Tedia, and ethanol (>99.8%) was obtained from Merck. All chemicals were used without further purification. The purity of all chemicals was checked by gas chromatography. Gas chromatographic analysis showed that the major peak areas are 99.9%, 99.9%, and 99.8% for acetone, ethanol, and 2,2,4-trimethylpentane, respectively. The purity of the solvents was further ascertained by comparing their densities, viscosities, and refractive indices at a temperature of 298.15 K, and the results agreed well with the corresponding values reported in the literature as shown in Table 1.

Apparatus and Procedure. Liquids were dried over activated molecular sieves, type 0.3 nm, from Aldrich. All of the dried liquids were shaken in an ultrasonic oscillator to remove dissolved air. Samples were prepared by mass in a 50 cm^3 Erlenmeyer flask provided with a joint stopper, using a Precisa 262SMA balance with an uncertainty of $\pm 3 \times 10^{-5}$g. Densities were measured with an Anton Paar DMA-5000 vibrating-tube density meter (Anton Paar, Graz, Austria) with an uncertainty of $\pm 5 \times 10^{-6}$g·cm^{-3} in the range of (0 to 3) g·cm^{-3}, thermostatically controlled to within ±0.01 K in the range of (273.15 to 363.15) K. Calibration was performed periodically under atmospheric pressure, in accordance with specifications, using deionized water and dry air. Precautions were taken to prevent evaporation losses or air dissolved during the experiment. The uncertainty of the density measurements was estimated to be $\pm 3 \times 10^{-5}$ g·cm^{-3}. The excess molar volumes were calculated from density data, and the uncertainties were estimated to be within $\pm 5 \times 10^{-3}$ cm^3·mol^{-1}.

The kinematic viscosities were determined with commercial Ubbelohde capillary viscometers (Cannon Instrument Co., State College, PA) of (0.36 and 0.47) mm diameter. The viscometer was kept in a Lauda D20 KP thermostat controlled to ±0.01 K with a proportional-integral-differential regulator. A computer-controlled measuring system (Lauda, Lauda-Königshofen, Germany) with an uncertainty of ±0.01 s was used for flow-time measurement. The range of the flow time for the liquids investigated is varied from (360 to 750) s. The kinematic viscosity (ν) was then obtained from the following relationship

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

where t is the flow time, η is the absolute viscosity, and k and θ are the viscometer constant and the Hagenbach correction, respectively. The k values were 0.001047 ± 0.000001 and 0.002736 ± 0.000002 for the capillary viscometers with 0.36 mm and 0.47 mm diameters, respectively. The value θ, which is dependent on the flow time and the size of capillary, was taken from the tables supplied by the manufacturer. Triplicate measurements of flow times were reproducible to within ±0.02%. The uncertainty

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Table 1. Comparison of Measured Densities, Viscosities, and Refractive Indices of the Pure Components with Literature Values at 298.15 K

	ρ/g·cm^{-3}		η/mPa·s		n_D	
component	this work	lit	this work	lit	this work	lit
acetone	0.78547	0.7844[a]	0.302	0.3029[a]	1.35605	1.35596[a]
		0.78547[b]				
ethanol	0.78517	0.78493[a]	1.084	1.0826[a]	1.35924	1.35941[a]
		0.78502[c]				1.35922[c]
2,2,4-trimethypentane	0.68774	0.68781[a]	0.480	0.4802[d]	1.38900	1.38898[a]

[a] Riddick et al., 1986.[5] [b] *TRC* Tables 1996.[6] [c] Rodriguez et al., 1996.[7] [d] Bouzas et al., 2000.[8]

Table 2. Experimental Densities ρ, Viscosities η, Refractive Indices n_D, Excess Molar Volumes V^E, Deviations in the Viscosity $\Delta\eta$, and Deviations in the Refractive Index Δn_D for the Binary Systems at 288.15 K

	ρ	η		V^E	$\Delta\eta$			ρ	η		V^E	$\Delta\eta$	
x_1	g·cm^{-3}	mPa·s	n_D	cm^3·mol^{-1}	mPa·s	Δn_D	x_1	g·cm^{-3}	mPa·s	n_D	cm^3·mol^{-1}	mPa·s	Δn_D
Acetone (1) + Ethanol (2)													
0.0000	0.79374	1.309	1.36342	0.000	0.000	0.00000	0.5500	0.79668	0.461	1.36301	−0.087	−0.312	0.00074
0.0500	0.79440	1.126	1.36354	−0.034	−0.134	0.00024	0.6000	0.79677	0.438	1.36289	−0.083	−0.286	0.00071
0.1000	0.79486	0.985	1.36360	−0.055	−0.226	0.00041	0.6500	0.79684	0.418	1.36275	−0.077	−0.257	0.00066
0.1500	0.79523	0.873	1.36366	−0.070	−0.290	0.00058	0.7000	0.79686	0.401	1.36261	−0.068	−0.225	0.00061
0.2000	0.79550	0.783	1.36361	−0.078	−0.331	0.00064	0.7500	0.79688	0.386	1.36246	−0.058	−0.192	0.00054
0.2500	0.79575	0.709	1.36357	−0.084	−0.356	0.00071	0.8000	0.79691	0.373	1.36230	−0.049	−0.156	0.00046
0.3000	0.79599	0.649	1.36350	−0.091	−0.367	0.00074	0.8501	0.79690	0.363	1.36214	−0.037	−0.117	0.00039
0.3500	0.79618	0.598	1.36343	−0.094	−0.370	0.00078	0.9000	0.79692	0.353	1.36198	−0.027	−0.078	0.00031
0.4000	0.79634	0.556	1.36334	−0.094	−0.363	0.00079	0.9500	0.79691	0.344	1.36177	−0.015	−0.039	0.00017
0.4500	0.79646	0.518	1.36324	−0.092	−0.352	0.00078	1.0000	0.79687	0.334	1.36152	0.000	0.000	0.00000
0.4999	0.79657	0.488	1.36313	−0.089	−0.334	0.00077							
Acetone (1) + 2,2,4-Trimethylpentane (2)													
0.0000	0.69595	0.537	1.39404	0.000	0.000	0.00000	0.5500	0.72546	0.407	1.37934	0.942	−0.018	−0.00326
0.0499	0.69734	0.519	1.39283	0.208	−0.008	−0.00047	0.6000	0.73017	0.398	1.37774	0.918	−0.017	−0.00330
0.1000	0.69886	0.505	1.39158	0.407	−0.012	−0.00093	0.6500	0.73545	0.389	1.37606	0.871	−0.016	−0.00328
0.1500	0.70068	0.492	1.39031	0.559	−0.015	−0.00137	0.7000	0.74135	0.380	1.37430	0.805	−0.015	−0.00319
0.2000	0.70268	0.479	1.38905	0.696	−0.017	−0.00174	0.7500	0.74784	0.372	1.37243	0.737	−0.013	−0.00303
0.2500	0.70509	0.468	1.38769	0.776	−0.018	−0.00216	0.8000	0.75530	0.364	1.37045	0.629	−0.011	−0.00278
0.3000	0.70768	0.456	1.38640	0.851	−0.020	−0.00244	0.8500	0.76358	0.356	1.36839	0.520	−0.008	−0.00238
0.3500	0.71055	0.446	1.38509	0.907	−0.020	−0.00267	0.9000	0.77312	0.348	1.36624	0.377	−0.006	−0.00179
0.4000	0.71376	0.436	1.38374	0.937	−0.020	−0.00287	0.9500	0.78418	0.341	1.36394	0.197	−0.003	−0.00103
0.4500	0.71731	0.426	1.38233	0.949	−0.020	−0.00304	1.0000	0.79687	0.334	1.36152	0.000	0.000	0.00000
0.5000	0.72115	0.416	1.38086	0.959	−0.020	−0.00318							
Ethanol (1) + 2,2,4-Trimethylpentane (2)													
0.0000	0.69595	0.537	1.39404	0.000	0.000	0.00000	0.5500	0.72322	0.718	1.38350	0.328	−0.244	−0.00130
0.0502	0.69720	0.532	1.39319	0.124	−0.044	−0.00029	0.6000	0.72755	0.755	1.38209	0.317	−0.245	−0.00134
0.1000	0.69876	0.535	1.39241	0.195	−0.079	−0.00047	0.6500	0.73237	0.797	1.38055	0.304	−0.242	−0.00135
0.1500	0.70055	0.542	1.39162	0.242	−0.111	−0.00062	0.7000	0.73781	0.843	1.37884	0.286	−0.234	−0.00136
0.2000	0.70253	0.551	1.39081	0.277	−0.140	−0.00074	0.7500	0.74399	0.894	1.37694	0.262	−0.222	−0.00134
0.2500	0.70471	0.564	1.38996	0.303	−0.166	−0.00085	0.8000	0.75111	0.952	1.37484	0.224	−0.203	−0.00126
0.3000	0.70710	0.580	1.38907	0.322	−0.189	−0.00094	0.8500	0.75927	1.018	1.37249	0.187	−0.175	−0.00112
0.3500	0.70971	0.601	1.38811	0.337	−0.206	−0.00103	0.9000	0.76881	1.095	1.36984	0.138	−0.137	−0.00090
0.4000	0.71260	0.625	1.38709	0.343	−0.221	−0.00111	0.9500	0.78026	1.190	1.36684	0.066	−0.080	−0.00055
0.4500	0.71580	0.652	1.38597	0.341	−0.232	−0.00120	1.0000	0.79374	1.309	1.36342	0.000	0.000	0.00000
0.5000	0.71931	0.682	1.38479	0.338	−0.241	−0.00125							

of the viscosity measurement was estimated to be less than ±0.6%.

Refractive indices, n_D, were measured with an automatic Anton Paar RXA-156 refractometer, which works with the wavelength (589 nm) corresponding to the D line of sodium. The temperature range of this refractometer is from (283.15 to 343.15) K with an uncertainty of 0.03 K. Calibration was performed periodically under atmospheric pressure, in accordance with specifications, using double-distilled water. The uncertainty of the refractive index measurement is 0.00002 units in the range of (1.32 to 1.56) units.

The densities, viscosities, and refractive indices of the binary systems acetone + ethanol, acetone + 2,2,4-trimethylpentane, and ethanol + 2,2,4-trimethylpentane were measured at temperatures of 288.15 K, 298.15 K, and 308.15 K and atmospheric pressure (100.8 ± 0.2) kPa. A set with the compositions varying from 0.05 to 0.95 mole fraction was prepared for each binary system. A total of 55 compositions were measured at 298.15 K for the ternary system acetone + ethanol + 2,2,4-trimethylpentane. An average of at least three measurements was taken for each composition. The uncertainties in the liquid composition and of the investigated temperature were estimated to be $\pm 1 \times 10^{-4}$ and ±0.01 K, respectively.

Results and Discussion

Tables 2 to 4 list the experimental densities, viscosities, refractive indices, excess molar volumes, deviations in the viscosity from the mole fraction average, and deviations in the refractive index from the volume fraction average for three binary mixtures acetone + ethanol, acetone + 2,2,4-trimethylpentane, and ethanol + 2,2,4-trimethylpentane at temperatures of 288.15 K, 298.15 K, and 308.15 K. The molar excess volumes, V^E, were calculated from density

Table 3. Experimental Densities ρ, Viscosities η, Refractive Indices n_D, Excess Molar Volumes V^E, Deviations in the Viscosity $\Delta\eta$, and Deviations in the Refractive Index Δn_D for the Binary Systems at 298.15 K

x_1	ρ / g·cm^{-3}	η / mPa·s	n_D	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s	Δn_D	x_1	ρ / g·cm^{-3}	η / mPa·s	n_D	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s	Δn_D
Acetone (1) + Ethanol (2)													
0.0000	0.78517	1.084	1.35924	0.000	0.000	0.00000	0.5500	0.78617	0.409	1.35790	−0.070	−0.245	0.00059
0.0500	0.78559	0.944	1.35919	−0.030	−0.101	0.00015	0.6000	0.78613	0.390	1.35771	−0.066	−0.225	0.00056
0.1000	0.78584	0.829	1.35915	−0.049	−0.177	0.00030	0.6500	0.78607	0.373	1.35753	−0.060	−0.203	0.00053
0.1500	0.78600	0.741	1.35909	−0.060	−0.226	0.00043	0.7000	0.78600	0.358	1.35734	−0.053	−0.179	0.00048
0.2000	0.78608	0.669	1.35899	−0.066	−0.259	0.00051	0.7500	0.78590	0.345	1.35713	−0.044	−0.153	0.00041
0.2500	0.78616	0.610	1.35887	−0.072	−0.279	0.00057	0.8000	0.78584	0.334	1.35692	−0.038	−0.124	0.00034
0.3000	0.78623	0.561	1.35873	−0.077	−0.288	0.00061	0.8501	0.78573	0.325	1.35673	−0.027	−0.094	0.00029
0.3500	0.78624	0.521	1.35857	−0.077	−0.289	0.00062	0.9000	0.78566	0.317	1.35651	−0.020	−0.063	0.00020
0.4000	0.78623	0.486	1.35842	−0.076	−0.285	0.00064	0.9500	0.78557	0.309	1.35629	−0.010	−0.032	0.00011
0.4500	0.78621	0.457	1.35826	−0.074	−0.275	0.00064	1.0000	0.78547	0.302	1.35605	0.000	0.000	0.00000
0.4999	0.78619	0.431	1.35809	−0.072	−0.262	0.00063							
Acetone (1) + 2,2,4-Trimethylpentane (2)													
0.0000	0.68774	0.480	1.38900	0.000	0.000	0.00000	0.5500	0.71573	0.366	1.37379	1.040	−0.016	−0.00360
0.0499	0.68889	0.465	1.38765	0.254	−0.006	−0.00060	0.6000	0.72032	0.358	1.37219	1.007	−0.015	−0.00362
0.1000	0.69036	0.452	1.38634	0.451	−0.010	−0.00111	0.6500	0.72544	0.350	1.37050	0.956	−0.014	−0.00359
0.1500	0.69207	0.440	1.38501	0.614	−0.013	−0.00159	0.7000	0.73116	0.343	1.36870	0.885	−0.012	−0.00351
0.2000	0.69398	0.429	1.38374	0.755	−0.015	−0.00196	0.7500	0.73751	0.336	1.36683	0.804	−0.011	−0.00333
0.2500	0.69624	0.419	1.38240	0.848	−0.017	−0.00234	0.8000	0.74471	0.329	1.36486	0.696	−0.009	−0.00304
0.3000	0.69870	0.409	1.38110	0.931	−0.018	−0.00262	0.8500	0.75289	0.322	1.36279	0.564	−0.007	−0.00261
0.3500	0.70144	0.399	1.37975	0.991	−0.019	−0.00288	0.9000	0.76222	0.315	1.36064	0.407	−0.005	−0.00199
0.4000	0.70451	0.390	1.37833	1.027	−0.019	−0.00313	0.9500	0.77302	0.309	1.35838	0.215	−0.002	−0.00115
0.4500	0.70786	0.382	1.37687	1.051	−0.018	−0.00333	1.0000	0.78547	0.302	1.35605	0.000	0.000	0.00000
0.5000	0.71161	0.374	1.37538	1.052	−0.017	−0.00347							
Ethanol (1) + 2,2,4-Trimethylpentane (2)													
0.0000	0.68774	0.480	1.38900	0.000	0.000	0.00000	0.5500	0.71459	0.615	1.37858	0.379	−0.197	−0.00145
0.0502	0.68884	0.473	1.38810	0.160	−0.037	−0.00035	0.6000	0.71890	0.645	1.37722	0.366	−0.197	−0.00147
0.1000	0.69033	0.476	1.38730	0.245	−0.064	−0.00058	0.6500	0.72373	0.677	1.37573	0.347	−0.196	−0.00148
0.1500	0.69211	0.480	1.38651	0.292	−0.091	−0.00074	0.7000	0.72914	0.715	1.37408	0.327	−0.188	−0.00147
0.2000	0.69407	0.487	1.38569	0.328	−0.114	−0.00090	0.7500	0.73534	0.755	1.37225	0.294	−0.178	−0.00144
0.2500	0.69621	0.497	1.38485	0.359	−0.134	−0.00101	0.8000	0.74237	0.801	1.37020	0.262	−0.162	−0.00137
0.3000	0.69857	0.510	1.38397	0.379	−0.151	−0.00112	0.8500	0.75054	0.854	1.36792	0.217	−0.139	−0.00123
0.3500	0.70117	0.526	1.38304	0.393	−0.165	−0.00120	0.9000	0.76017	0.915	1.36536	0.154	−0.109	−0.00100
0.4000	0.70403	0.543	1.38204	0.399	−0.179	−0.00129	0.9500	0.77159	0.991	1.36248	0.078	−0.063	−0.00062
0.4500	0.70720	0.565	1.38099	0.398	−0.187	−0.00134	1.0000	0.78517	1.084	1.35924	0.000	0.000	0.00000
0.5000	0.71068	0.588	1.37984	0.394	−0.194	−0.00138							

data according to the equation

$$V^E = \sum_{i=1}^{N} x_i M_i \left(\frac{1}{\rho} - \frac{1}{\rho_i}\right) \tag{2}$$

where x_i, M_i, and ρ_i are the mole fraction, molar mass, and density of pure component i, respectively, ρ is the density of the mixture, and N is the number of components.

In the system studied, excess molar volumes are positive for the mixtures acetone + 2,2,4-trimethylpentane and ethanol + 2,2,4-trimethylpentane and negative for the mixture acetone + ethanol over the whole composition range. The values of V^E increase systematically from 288.15 K to 308.15 K over the whole range of mole fraction. The excess molar volume $V^E(x = 0.5)$ increases in the sequence acetone + 2,2,4-trimethylpentane > ethanol + 2,2,4-trimethylpentane > acetone + ethanol. The values of V^E ($x = 0.5$) vary from (−0.089 to 1.154) cm^3·mol^{-1}. In the literature, V^E of the three binary systems in this study had been measured at 298.15 K.[9−12] Figure 1 shows the excess molar volumes for the three systems at $T = 298.15$ K together with the literature values, and our results are in good agreement with their measurements. The curves for ethanol + 2,2,4-trimethylpentane are skewed toward low ethanol mole fraction with the maximum occurring near $x_1 = 0.4$, whereas a similar phenomenon was observed for the curve of molar excess enthalpy.[13]

The dependence of V^E on both composition and temperature for the present mixtures may be explained as a balance between positive contributions (hydrogen bond rupture and dispersive interactions between unlike molecules) and negative contributions (intermolecular dipolar interactions and geometrical fitting between components). In the present investigation, ethanol is strongly self-associated through hydrogen bonding, but acetone and 2,2,4-trimethylpentane do not exhibit this property. The interactions of ethanol and acetone against 2,2,4-trimethylpentane molecules involve mainly the dispersion force giving a positive contribution to V^E. The interactions between ethanol and acetone molecules lead to hydrogen bond effects and/or a weak dispersion-type effect, giving a negative contribution to V^E.

The deviation of the viscosity from the mole fraction average, $\Delta\eta$, is given by

$$\Delta\eta = \eta - \sum_{i=1}^{N} x_i \eta_i \tag{3}$$

where η is the absolute viscosity of the mixture and η_i is the absolute viscosity of pure component i. The $\Delta\eta$ values, which are negative over the entire range of composition, increase with increasing temperature. However, the $\Delta\eta$ values in the mixture acetone + 2,2,4-trimethylpentane show little variation with temperature. The values of $\Delta\eta$ follow order acetone + 2,2,4-trimethylpentane > ethanol + 2,2,4-trimethylpentane > acetone + ethanol. The values of $\Delta\eta$ ($x = 0.5$) vary from (−0.334 to −0.015) mPa·s. Figure 2 plots the results of $\Delta\eta$ for the three binary mixtures at

Table 4. Experimental Densities ρ, Viscosities η, Refractive Indices n_D, Excess Molar Volumes V^E, Deviations in the Viscosity $\Delta\eta$, and Deviations in the Refractive Index Δn_D for the Binary Systems at 308.15 K

x_1	ρ / g·cm^{-3}	η / mPa·s	n_D	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s	Δn_D	x_1	ρ / g·cm^{-3}	η / mPa·s	n_D	V^E / cm^3·mol^{-1}	$\Delta\eta$ / mPa·s	Δn_D
Acetone (1) + Ethanol (2)													
0.0000	0.77649	0.905	1.35502	0.000	0.000	0.00000	0.5500	0.77547	0.364	1.35276	−0.048	−0.194	0.00044
0.0500	0.77666	0.793	1.35486	−0.026	−0.080	0.00012	0.6000	0.77530	0.349	1.35251	−0.044	−0.177	0.00040
0.1000	0.77666	0.703	1.35470	−0.038	−0.139	0.00023	0.6500	0.77513	0.335	1.35228	−0.040	−0.160	0.00038
0.1500	0.77661	0.632	1.35454	−0.047	−0.178	0.00033	0.7000	0.77496	0.322	1.35203	−0.036	−0.141	0.00033
0.2000	0.77653	0.575	1.35435	−0.053	−0.204	0.00040	0.7500	0.77477	0.312	1.35177	−0.030	−0.120	0.00027
0.2500	0.77641	0.527	1.35414	−0.056	−0.220	0.00044	0.8000	0.77459	0.303	1.35152	−0.024	−0.097	0.00022
0.3000	0.77628	0.487	1.35391	−0.058	−0.229	0.00045	0.8501	0.77440	0.295	1.35127	−0.016	−0.074	0.00016
0.3500	0.77612	0.453	1.35369	−0.056	−0.231	0.00047	0.9000	0.77423	0.288	1.35102	−0.011	−0.049	0.00009
0.4000	0.77596	0.426	1.35346	−0.055	−0.227	0.00048	0.9500	0.77408	0.281	1.35080	−0.006	−0.025	0.00005
0.4500	0.77579	0.403	1.35324	−0.052	−0.218	0.00048	1.0000	0.77391	0.274	1.35057	0.000	0.000	0.00000
0.4999	0.77563	0.383	1.35300	−0.050	−0.207	0.00046							
Acetone (1) + 2,2,4-Trimethylpentane (2)													
0.0000	0.67941	0.432	1.38395	0.000	0.000	0.00000	0.5500	0.70586	0.332	1.36831	1.144	−0.013	−0.00386
0.0499	0.68044	0.420	1.38256	0.273	−0.004	−0.00063	0.6000	0.71030	0.325	1.36666	1.107	−0.012	−0.00390
0.1000	0.68177	0.408	1.38119	0.490	−0.008	−0.00118	0.6500	0.71527	0.318	1.36495	1.049	−0.011	−0.00387
0.1500	0.68336	0.397	1.37981	0.666	−0.011	−0.00170	0.7000	0.72079	0.312	1.36314	0.976	−0.009	−0.00378
0.2000	0.68516	0.387	1.37848	0.815	−0.013	−0.00212	0.7500	0.72701	0.306	1.36126	0.879	−0.008	−0.00358
0.2500	0.68726	0.378	1.37710	0.925	−0.015	−0.00253	0.8000	0.73399	0.299	1.35931	0.765	−0.007	−0.00324
0.3000	0.68956	0.369	1.37574	1.021	−0.016	−0.00285	0.8500	0.74202	0.293	1.35726	0.614	−0.005	−0.00277
0.3500	0.69221	0.361	1.37434	1.079	−0.016	−0.00314	0.9000	0.75112	0.287	1.35510	0.444	−0.003	−0.00212
0.4000	0.69510	0.353	1.37290	1.127	−0.016	−0.00339	0.9500	0.76168	0.281	1.35285	0.237	−0.001	−0.00124
0.4500	0.69829	0.346	1.37142	1.157	−0.015	−0.00360	1.0000	0.77391	0.274	1.35057	0.000	0.000	0.00000
0.5000	0.70191	0.338	1.36989	1.154	−0.015	−0.00376							
Ethanol (1) + 2,2,4-Trimethylpentane (2)													
0.0000	0.67941	0.432	1.38395	0.000	0.000	0.00000	0.5500	0.70580	0.531	1.37358	0.440	−0.161	−0.00165
0.0502	0.68036	0.424	1.38297	0.198	−0.032	−0.00045	0.6000	0.71011	0.554	1.37227	0.420	−0.162	−0.00167
0.1000	0.68176	0.426	1.38217	0.303	−0.053	−0.00069	0.6500	0.71493	0.580	1.37083	0.397	−0.159	−0.00167
0.1500	0.68352	0.429	1.38138	0.351	−0.074	−0.00087	0.7000	0.72034	0.609	1.36922	0.371	−0.154	−0.00167
0.2000	0.68543	0.434	1.38056	0.395	−0.093	−0.00104	0.7500	0.72654	0.642	1.36745	0.333	−0.145	−0.00162
0.2500	0.68756	0.441	1.37972	0.424	−0.109	−0.00119	0.8000	0.73358	0.678	1.36550	0.293	−0.132	−0.00152
0.3000	0.68988	0.451	1.37885	0.448	−0.123	−0.00130	0.8500	0.74175	0.720	1.36330	0.243	−0.114	−0.00136
0.3500	0.69245	0.462	1.37794	0.462	−0.136	−0.00139	0.9000	0.75144	0.769	1.36085	0.168	−0.089	−0.00110
0.4000	0.69530	0.476	1.37696	0.466	−0.145	−0.00148	0.9500	0.76284	0.828	1.35812	0.089	−0.053	−0.00065
0.4500	0.69844	0.492	1.37593	0.464	−0.153	−0.00154	1.0000	0.77649	0.905	1.35502	0.000	0.000	0.00000
0.5000	0.70191	0.510	1.37480	0.457	−0.159	−0.00160							

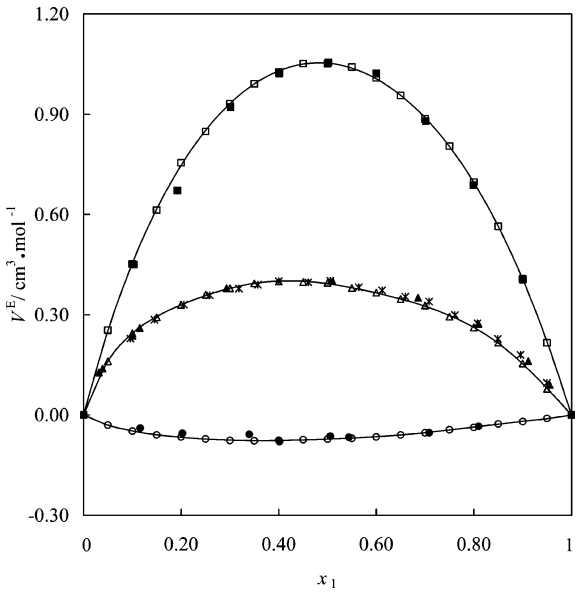

Figure 1. Variation of excess molar volume V^E with mole fraction x_1 for the binary systems at $T = 298.15$ K: ○, acetone + ethanol; ●, acetone + ethanol from Wei et al.;[9] □, acetone + 2,2,4-trimethylpentane; ■, acetone + 2,2,4-trimethylpentane from Papaloannou et al.;[10] △, ethanol + 2,2,4-trimethylpentane; ▲, ethanol + 2,2,4-trimethylpentane from Kretschmer et al.;[11] *, ethanol + 2,2,4-trimethylpentane from Blanco et al.[12] Solid curves were calculated from the Redlich−Kister equation.

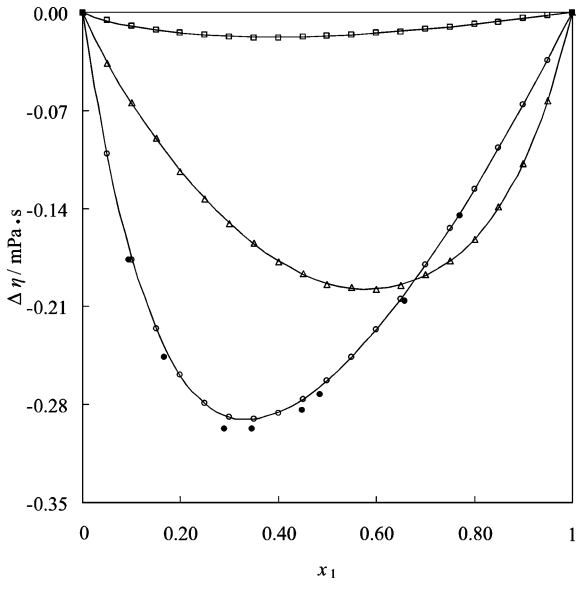

Figure 2. Variation of deviation in the viscosity $\Delta\eta$ with mole fraction x_1 for the binary systems at $T = 298.15$ K: ○, acetone + ethanol; ●, acetone + ethanol from Wei et al.;[9] □, acetone + 2,2,4-trimethylpentane; △, ethanol + 2,2,4-trimethylpentane. Solid curves were calculated from the Redlich−Kister equation.

298.15 K together with the literature values[9] for the system acetone + ethanol. In the high acetone region, our viscosity data agree well with the literature values. However, in the low acetone region, our data give somewhat higher values than the literature data.

The deviations in refractive index Δn_D were calculated from the volume fraction average as suggested by Brocos

Table 5. Binary Coefficients of the Redlich−Kister Equation from T = 288.15 K to T = 308.15 K and Ternary Coefficients of the Cibulka Equation at T = 298.15 K for V^E, $\Delta\eta$, and Δn_D

ΔQ_{ij}	T/K	$a_0 \times 10^2$	$a_1 \times 10^2$	$a_2 \times 10^2$	$a_3 \times 10^2$	$a_4 \times 10^2$	σ
			Acetone (1) + Ethanol (2)				
V^E/cm^3·mol^{-1}	288.15	−35.92	10.88	−12.75	13.78		0.0015
	298.15	−28.86	10.84	−11.93	14.61		0.0013
	308.15	−20.12	10.40	−11.41	14.03		0.0010
$\Delta\eta$/mPa·s	288.15	−133.55	78.34	−45.63	38.33	−15.80	0.0008
	298.15	−104.46	60.63	−43.25	27.15		0.0007
	308.15	−82.83	49.50	−32.62	18.45		0.0007
Δn_D	288.15	0.308	−0.088	0.076	−0.003	0.109	1.0×10^{-5}
	298.15	0.249	−0.079	0.051			0.9×10^{-5}
	308.15	0.185	−0.075	0.008	−0.035		0.9×10^{-5}
			Acetone (1) + 2,2,4-Trimethylpentane (2)				
V^E/cm^3·mol^{-1}	288.15	382.81	−24.81	80.11			0.0059
	298.15	420.37	−27.92	86.30			0.0041
	308.15	461.91	−27.02	84.90			0.0044
$\Delta\eta$/mPa·s	288.15	−7.67	2.39	−3.23	2.92		0.0005
	298.15	−6.92	3.46	−1.95			0.0003
	308.15	−5.83	3.60	−0.74			0.0004
Δn_D	288.15	−1.269	−0.412	−0.462	−0.302	0.114	1.5×10^{-5}
	298.15	−1.390	−0.501	−0.423	−0.161	−0.173	1.2×10^{-5}
	308.15	−1.502	−0.513	−0.448	−0.207	−0.130	1.1×10^{-5}
V^E/cm^3·mol^{-1}	288.15	382.81	−24.81	80.11			0.0059
	298.15	420.37	−27.92	86.30			0.0041
	308.15	461.91	−27.02	84.90			0.0044
$\Delta\eta$/mPa·s	288.15	−7.67	2.39	−3.23	2.92		0.0005
	298.15	−6.92	3.46	−1.95			0.0003
	308.15	−5.83	3.60	−0.74			0.0004
Δn_D	288.15	−1.269	−0.412	−0.462	−0.302	0.114	1.5×10^{-5}
	298.15	−1.390	−0.501	−0.423	−0.161	−0.173	1.2×10^{-5}
	308.15	−1.502	−0.513	−0.448	−0.207	−0.130	1.1×10^{-5}
			Ethanol (1) + 2,2,4-Trimethylpentane (2)				
V^E/cm^3·mol^{-1}	288.15	134.18	−15.75	70.57	−38.62		0.0051
	298.15	157.23	−21.10	49.07	−58.72	78.93	0.0064
	308.15	182.73	−30.83	49.87	−84.04	111.39	0.0078
$\Delta\eta$/mPa·s	288.15	−96.16	−23.86	−22.72	−24.52	22.53	0.0005
	298.15	−77.55	−19.74	−17.10	−15.48	−19.20	0.0008
	308.15	−63.53	−16.26	−10.65	−12.25	−22.60	0.0007
Δn_D	288.15	−0.501	−0.232	−0.255	−0.101	−0.250	6.0×10^{-5}
	298.15	−0.560	−0.187	−0.298	−0.166	−0.311	0.8×10^{-5}
	308.15	−0.642	−0.204	−0.321	−0.113	−0.355	1.4×10^{-5}

ΔQ_{123}	C_0	C_1	C_2	σ
V^E/cm^3·mol^{-1}	−3.4532	5.3601	6.7000	0.0047
$\Delta\eta$/mPa·s	0.7825	−0.6426	−0.1511	0.0022
Δn_D	0.0071	−0.0348	−0.0452	6.0×10^{-5}

et al.[14] and is given by

$$\Delta n_D = n_D - \sum_{i=1}^{N} \phi_i n_{Di} \tag{4}$$

and

$$\phi_i = \frac{x_i V}{\sum_{i=1}^{N} x_i V_i} \tag{5}$$

where n_D, n_{Di}, and ϕ_i are the refractive index of the mixture, the refractive index of pure component i, and the volume fraction of component i, respectively. V and V_i are the molar volume of the mixture and the molar volume of component i, respectively, and N is the number of components. The Δn_D values, which decrease with increasing temperature, are negative for the systems acetone + 2,2,4-trimethylpentane and ethanol + 2,2,4-trimethylpentane and positive for the system acetone + ethanol over the whole composition. The values of Δn_D (x = 0.5) follow the order acetone + ethanol > ethanol + 2,2,4-trimethylpentane > acetone + 2,2,4-trimethylpentane. The values of Δn_D (x = 0.5) vary from −0.00165 to 0.00078. Figure 3 shows the results of Δn_D as a function of mole fraction for the three binary systems at 298.15 K.

The mixing functions V^E, $\Delta\eta$, and Δn_D were represented mathematically by the Redlich−Kister equation[15] for correlating the experimental data

$$\Delta Q_{ij} = x_i x_j \sum_{k=0}^{m} a_k (x_i - x_j)^k \tag{6}$$

where ΔQ_{ij} refers to V^E/cm^3·mol^{-1}, $\Delta\eta$/mPa·s, or Δn_D for each i−j binary pair, x_i is the mole fraction of component i, and a_k represents the coefficients. The values of coefficients a_k were determined by a multiple regression analysis based on the least-squares method and are summarized along with the standard deviations between the experimental and fitted values of the respective functions in Table 5. The standard deviation is defined by

$$\sigma = \left[\sum_{i=1}^{n} \frac{(\Delta Q_i^{exptl} - \Delta Q_i^{calcd})^2}{n - p} \right]^{1/2} \tag{7}$$

where n is the number of experimental points and p is the

Table 6. Binary Coefficients of McAllister's Multibody-Interaction Equations and Standard Deviations, σ, for Kinematic Viscosities

	three-body			four-body			
T	ν_{12}	ν_{21}	$10^4\sigma$	ν_{1112}	ν_{1122}	ν_{2221}	$10^4\sigma$
K	mm²·s⁻¹	mm²·s⁻¹	mm²·s⁻¹	mm²·s⁻¹	mm²·s⁻¹	mm²·s⁻¹	mm²·s⁻¹
			Acetone (1) + Ethanol (2)				
288.15	0.5050	0.6031	36	0.4732	0.5691	0.7615	20
298.15	0.4561	0.5406	36	0.4235	0.5240	0.6622	11
308.15	0.4209	0.4791	27	0.3924	0.4705	0.5822	9
			Acetone (1) + 2,2,4-Trimethylpentane (2)				
288.15	0.5611	0.6287	6	0.5159	0.5943	0.6571	4
298.15	0.5150	0.5687	4	0.4767	0.5352	0.5984	4
308.15	0.4780	0.5184	7	0.4448	0.4863	0.5493	3
			Ethanol (1) + 2,2,4-Trimethylpentane (2)				
288.15	1.1357	0.7575	79	1.1767	0.9922	0.7249	24
298.15	0.9734	0.6761	55	1.0087	0.8585	0.6530	15
308.15	0.8368	0.6076	48	0.8643	0.7530	0.5899	17

number of adjustable parameters. The σ values lie between 0.0010 cm³·mol⁻¹ and 0.0078 cm³·mol⁻¹, between 0.0003 mPa·s and 0.0008 mPa·s, and between 0.000006 and 0.000015 for V^E, $\Delta\eta$, and Δn_D, respectively.

McAllister's multibody interaction model[16] is widely used for correlating the kinematic viscosity of binary mixtures with mole fraction. The three-body model is defined as

$$\ln \nu = x_1^3 \ln \nu_1 + 3x_1^2 x_2 \ln \nu_{12} + 3x_1 x_2^2 \ln \nu_{21} + x_2^3 \ln \nu_2 - \ln\left(x_1 + \frac{x_2 M_2}{M_1}\right) + 3x_1^2 x_2 \ln\left(\frac{2 + M_2/M_1}{3}\right) + 3x_1 x_2^2 \ln\left(\frac{1 + 2M_2/M_1}{3}\right) + x_2^3 \ln\left(\frac{M_2}{M_1}\right) \quad (8)$$

and the four-body model is given by

$$\ln \nu = x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln \nu_{1112} + 6x_1^2 x_2^2 \ln \nu_{1122} + 4x_1 x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 - \ln\left(x_1 + \frac{x_2 M_2}{M_1}\right) + 4x_1^3 x_2 \ln\left(\frac{3 + M_2/M_1}{4}\right) + 6x_1^2 x_2^2 \ln\left(\frac{1 + M_2/M_1}{2}\right) + 4x_1 x_2^3 \ln\left(\frac{1 + 3M_2/M_1}{4}\right) + x_2^4 \ln\left(\frac{M_2}{M_1}\right) \quad (9)$$

where ν, ν_1, and ν_2 are the kinematic viscosities of the mixture and the viscosities of pure components 1 and 2, respectively. ν_{12}, ν_{21}, ν_{1112}, ν_{1122}, and ν_{2221} are the model parameters. Table 6 records the calculated results with the standard deviation defined as eq 7. It is shown that McAllister's four-body equation gave a better result for those three systems. The σ/mm²·s⁻¹ values for this model lie between 0.0003 and 0.0024, and the largest σ value corresponds to the ethanol + 2,2,4-trimethylpentane system at 288.15 K.

The experimental densities, viscosities, refractive indices, excess molar volumes, deviations in the viscosity from the mole fraction average, and deviations in the refractive index from the volume fraction average for the ternary system acetone (1) + ethanol (2) + 2,2,4-trimethylpentane (3) at a temperature of 298.15 K are listed in Table 7. The derived properties, V^E/cm³·mol⁻¹, $\Delta\eta$/mPa·s, and Δn_D, of the ternary system were correlated

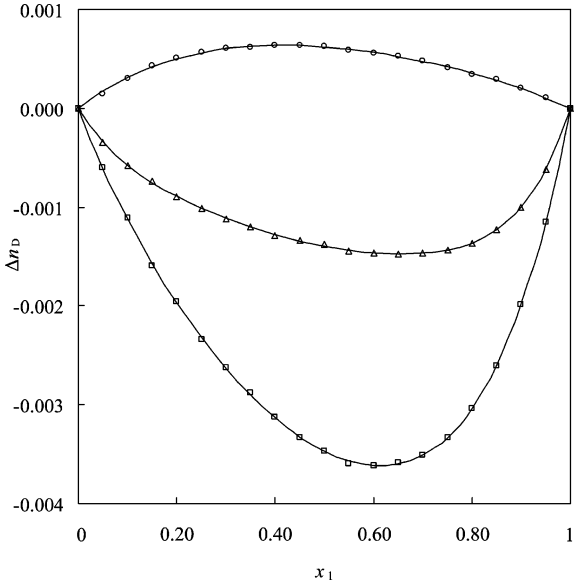

Figure 3. Variation of deviation in the refractive index deviation Δn_D with mole fraction x_1 for the binary systems at $T = 298.15$ K: ○, acetone + ethanol; □, acetone + 2,2,4-trimethylpentane; △, ethanol + 2,2,4-trimethylpentane. Solid curves were calculated from the Redlich−Kister equation.

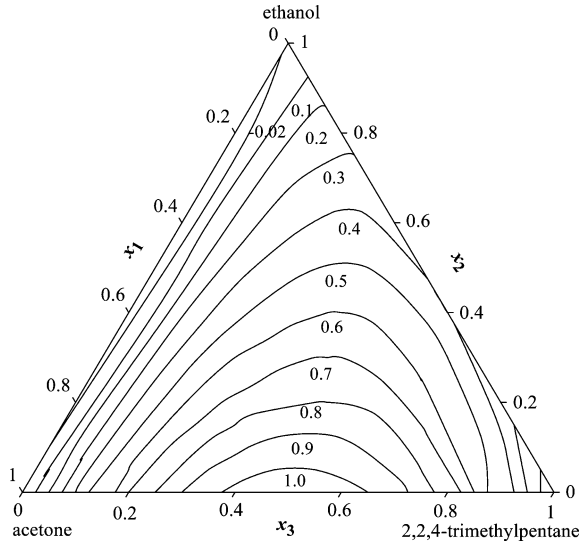

Figure 4. Curves of constant V^E/cm³·mol⁻¹ for the ternary system acetone (1) + ethanol (2) + 2,2,4-trimethylpentane (3) at 298.15 K.

Table 7. Experimental Densities ρ, Viscosities η, Refractive Indices n_D, Excess Molar Volumes V^E, Deviations in the Viscosity $\Delta\eta$, and Deviations in the Refractive Index Δn_D for the Ternary System Acetone (1) + Ethanol (2) + 2,2,4-Trimethylpentane (3) at 298.15 K

		ρ	η		V^E	$\Delta\eta$				ρ	η		V^E	$\Delta\eta$	
x_1	x_2	g·cm^{-3}	mPa·s	n_D	cm^3·mol^{-1}	mPa·s	Δn_D	x_1	x_2	g·cm^{-3}	mPa·s	n_D	cm^3·mol^{-1}	mPa·s	Δn_D
0.0500	0.9000	0.77182	0.864	1.36230	0.074	−0.151	−0.00057	0.3000	0.5500	0.75177	0.495	1.36621	0.306	−0.264	−0.00148
0.0500	0.8000	0.75083	0.753	1.36758	0.224	−0.201	−0.00132	0.3000	0.4500	0.73644	0.466	1.37004	0.478	−0.232	−0.00213
0.0500	0.7000	0.73546	0.673	1.37179	0.333	−0.221	−0.00162	0.3000	0.3500	0.72457	0.443	1.37322	0.627	−0.195	−0.00246
0.0502	0.5999	0.72383	0.612	1.37522	0.401	−0.221	−0.00172	0.3000	0.2500	0.71525	0.427	1.37588	0.737	−0.151	−0.00266
0.0500	0.5000	0.71466	0.563	1.37803	0.445	−0.210	−0.00174	0.3000	0.1500	0.70767	0.416	1.37814	0.830	−0.101	−0.00274
0.0500	0.4001	0.70730	0.523	1.38041	0.466	−0.190	−0.00167	0.3000	0.0500	0.70141	0.408	1.38011	0.904	−0.049	−0.00274
0.0500	0.3000	0.70122	0.495	1.38247	0.476	−0.157	−0.00153	0.4000	0.5500	0.77283	0.466	1.36101	0.084	−0.275	−0.00040
0.0500	0.2001	0.69622	0.477	1.38426	0.458	−0.115	−0.00137	0.4000	0.4500	0.75202	0.438	1.36558	0.350	−0.243	−0.00166
0.0500	0.1000	0.69206	0.464	1.38591	0.407	−0.067	−0.00111	0.4000	0.3500	0.73670	0.420	1.36932	0.550	−0.200	−0.00239
0.0500	0.0501	0.69031	0.459	1.38670	0.357	−0.042	−0.00096	0.4000	0.2599	0.72491	0.405	1.37244	0.710	−0.155	−0.00278
0.1000	0.8500	0.77209	0.771	1.36217	0.066	−0.205	−0.00049	0.4000	0.1499	0.71549	0.397	1.37505	0.851	−0.102	−0.00302
0.1000	0.7500	0.75110	0.679	1.36730	0.233	−0.236	−0.00135	0.4001	0.0499	0.70782	0.392	1.37725	0.974	−0.047	−0.00318
0.1000	0.6500	0.73579	0.615	1.37145	0.347	−0.240	−0.00171	0.5000	0.4501	0.77289	0.414	1.36057	0.107	−0.249	−0.00047
0.1000	0.5500	0.72400	0.566	1.37483	0.443	−0.228	−0.00186	0.5000	0.3501	0.75231	0.396	1.36497	0.388	−0.206	−0.00185
0.1000	0.4500	0.71477	0.527	1.37762	0.506	−0.207	−0.00189	0.4999	0.2501	0.73696	0.386	1.36862	0.620	−0.156	−0.00260
0.1000	0.3500	0.70736	0.492	1.37997	0.541	−0.182	−0.00185	0.5000	0.1500	0.72511	0.378	1.37166	0.814	−0.104	−0.00310
0.1000	0.2500	0.70125	0.473	1.38198	0.564	−0.140	−0.00179	0.5000	0.0499	0.71567	0.374	1.37421	0.973	−0.047	−0.00340
0.1000	0.1500	0.69620	0.457	1.38377	0.560	−0.096	−0.00164	0.6000	0.3500	0.77297	0.377	1.36010	0.126	−0.208	−0.00057
0.1002	0.0500	0.69205	0.448	1.38543	0.516	−0.044	−0.00138	0.6000	0.2500	0.75256	0.366	1.36434	0.430	−0.158	−0.00203
0.2000	0.7500	0.77244	0.627	1.36184	0.064	−0.270	−0.00038	0.6000	0.1501	0.73723	0.360	1.36788	0.688	−0.104	−0.00292
0.2000	0.6500	0.75147	0.569	1.36678	0.266	−0.268	−0.00137	0.5999	0.0501	0.72536	0.357	1.37084	0.904	−0.046	−0.00348
0.2000	0.5500	0.73605	0.526	1.37077	0.421	−0.251	−0.00189	0.7000	0.2500	0.77305	0.350	1.35964	0.145	−0.156	−0.00070
0.2000	0.4500	0.72431	0.495	1.37402	0.532	−0.221	−0.00216	0.7000	0.1500	0.75269	0.341	1.36375	0.484	−0.105	−0.00222
0.2001	0.3500	0.71499	0.469	1.37675	0.625	−0.187	−0.00227	0.7004	0.0500	0.73753	0.340	1.36715	0.757	−0.046	−0.00320
0.2000	0.2500	0.70752	0.451	1.37909	0.686	−0.145	−0.00225	0.8000	0.1500	0.77304	0.328	1.35912	0.173	−0.100	−0.00087
0.2001	0.1500	0.70133	0.437	1.38106	0.735	−0.098	−0.00224	0.8000	0.0500	0.75289	0.324	1.36308	0.529	−0.044	−0.00251
0.1999	0.0501	0.69620	0.427	1.38283	0.757	−0.048	−0.00213	0.9000	0.0500	0.77314	0.312	1.35859	0.189	−0.038	−0.00110
0.3000	0.6500	0.77269	0.533	1.36146	0.070	−0.286	−0.00035								

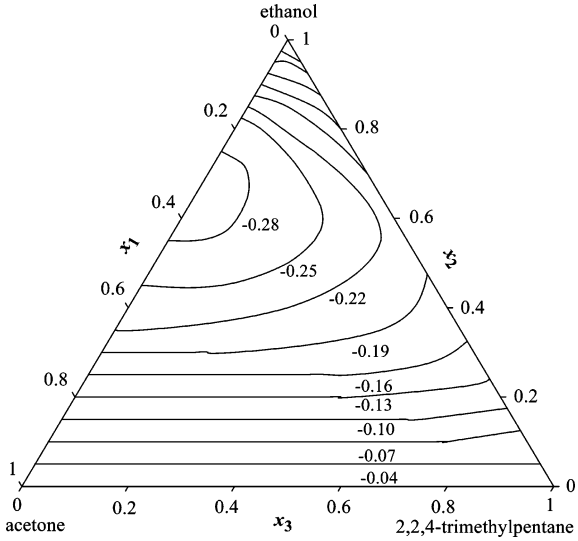

Figure 5. Curves of constant $\Delta\eta$ for the ternary system acetone (1) + ethanol (2) + 2,2,4-trimethylpentane (3) at 298.15 K.

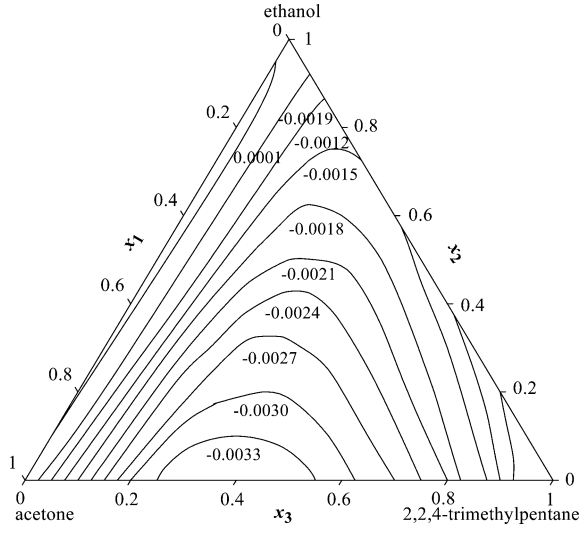

Figure 6. Curves of constant Δn_D for the ternary system acetone (1) + ethanol (2) + 2,2,4-trimethylpentane (3) at 298.15 K.

respectively using the equations

$$\Delta Q_{123} = \Delta Q_{bin} + x_1 x_2 x_3 \Delta_{123} \quad (10)$$

and

$$\Delta Q_{bin} = \Delta Q_{12} + \Delta Q_{13} + \Delta Q_{23} \quad (11)$$

where ΔQ_{123} refers to V^E, $\Delta\eta$, or Δn_D for ternary mixtures, $x_3 = 1 - x_1 - x_2$, and ΔQ_{ij} is the binary contribution of each $i-j$ pair to the V^E, $\Delta\eta$, or Δn_D given by eq 6 with the parameters shown in Table 5. The ternary contribution term Δ_{123} was correlated using the expression suggested by Cibulka[17]

$$\Delta_{123} = C_0 + C_1 x_1 + C_2 x_2 \quad (12)$$

The ternary parameters C_0, C_1, and C_2 were determined with the optimization algorithm similar to that for the binary parameters. The parameters C_0, C_1, and C_2 and the corresponding standard deviations are given in Table 5. The σ values are 0.005 cm^3·mol^{-1} 0.002 mPa·s, and 0.00006 for V^E, $\Delta\eta$, and Δn_D, respectively. The curves of constant V^E, $\Delta\eta$, and Δn_D at 298.15 K were calculated from eqs 10 to 12 and were plotted in Figures 4 to 6, respectively. As can be expected, the ternary system shows positive values of V^E at almost all compositions (Figure 4), except at compositions close to the binary system acetone + ethanol where a change in sign occurs. The maximum V^E value is found near $x_1 = 0.5$ of the binary system acetone + 2,2,4-trimethylpentane. Figure 5 shows negative values for ternary $\Delta\eta$, with a minimum value near $x_1 = 0.3$ of the binary system acetone + ethanol. Figure 6 shows the

negative values for ternary Δn_D at almost all compositions, except at compositions close to the binary system acetone + ethanol where a change in sign occurs. The minimum Δn_D value is found near $x_1 = 0.6$ of the binary system acetone + 2,2,4-trimethylpentane.

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