Densities, Refractive Indices, and Excess Molar Volumes of the Ternary Systems Water + Methanol + 1-Octanol and Water + Ethanol + 1-Octanol and Their Binary Mixtures at 298.15 K

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The densities, refractive indices, and excess molar volumes of the ternary systems water + methanol + 1-octanol and water + ethanol + 1-octanol have been determined at 298.15 K. The excess volumes have been correlated using Redlich–Kister functions.

Introduction

Mixtures of water with methanol or ethanol are among the final products of many chemical reactions, and many studies have been carried out to find suitable solvents for the selective recovery of the alcohol. One potential solvent for this purpose is 1-octanol. In the work described here we determined the densities, refractive indices, and excess molar volumes at 298.15 K of the ternary mixtures over the whole range of compositions. These physical properties are easily measured, and our tabulated data thus allow indirect determination of the composition of arbitrary mixtures. We have found no directly comparable data in the literature, though data are available for the binary mixtures formed by these components (*1–4*) and for other ternary mixtures containing these binary systems (*5, 6*).

Experimental Section

Water was treated by the Millipore process. Methanol and ethanol were purchased from Merck and 1-octanol was purchased from Aldrich with purities of 99.7, 99.5, and 99.5%, respectively; all three were used without further purification. Table I gives density and refractive index measurements for these components together with values obtained from the literature (*7*).

Densites were measured to within ±0.000 01 g/cm^3 using an Anton Paar DMA 60/602 densimeter and refractive indices at 589 nm using an Atago RX-1000 refractometer with a precision of ±0.0001. Temperature was controlled using a Heto Therm ultrathermostat with a precision of ±0.01 °C. All mixtures were prepared by weighing using a Mettler AE 240 balance accurate to within ±0.000 01 g.

Results

The molar volume V of the mixture was calculated as

$$V = \sum_i x_i M_i / d \tag{1}$$

where x_i is the mole fraction of component i in the mixture, M_i its molecular mass, and d the measured density. The excess molar volume is

$$V^E = V - \sum_i x_i V_i \tag{2}$$

where V_i is the molar volume of the pure component i.

The molar refraction, R, was obtained using the Lorentz–Lorenz equation:

$$R = \frac{n_D^2 - 1}{n_D^2 + 2} V \tag{3}$$

where n_D is the refractive index of the mixture. The deviation from a mole fraction average of the molar refraction is given

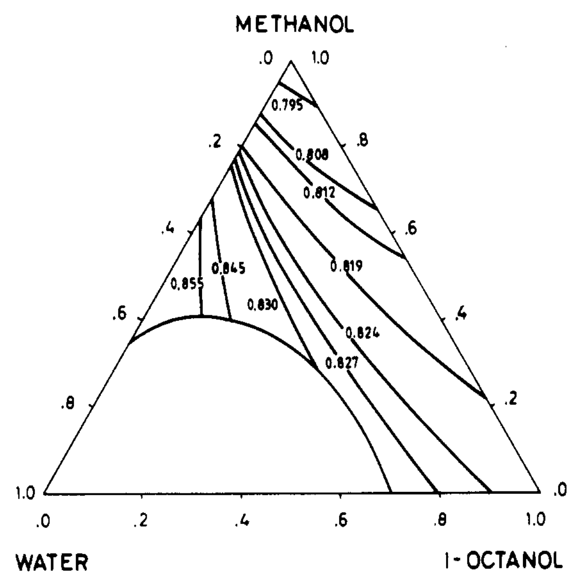

Figure 1. Density isolines for the mixture water + methanol + 1-octanol at 298.15 K.

Table I. Densities d and Refractive Indices n_D of Pure Components at 298.15 K

	d/(g cm^{-3})		n_D	
component	exptl	lit. (*7*)	exptl	lit. (*7*)
water	0.9970	0.997 04	1.3324	1.332 50
methanol	0.7866	0.786 64	1.3264	1.326 52
ethanol	0.7851	0.785 04	1.3592	1.359 41
1-octanol	0.8217	0.822 09	1.4275	1.427 50

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Table II. Densities d, Refractive Indices n_D, Excess Volumes V^E, and δR Values of Binary Systems at 298.15 K and Atmospheric Pressure

x_1	$d/(g{\cdot}cm^3)$	n_D	$V^E/(cm^3{\cdot}mol^{-1})$	$\delta R/(cm^3{\cdot}mol^{-1})$	x_1	$d/(g{\cdot}cm^3)$	n_D	$V^E/(cm^3{\cdot}mol^{-1})$	$\delta R/(cm^3{\cdot}mol^{-1})$
				Water (1) + Methanol (2)					
0.0000	0.786 59	1.3264	0.000	0.000	0.5604	0.895 03	1.3407	−1.016	−0.023
0.0716	0.799 01	1.3288	−0.267	−0.006	0.6063	0.904 93	1.3410	−0.982	−0.023
0.1203	0.807 60	1.3303	−0.423	−0.010	0.6562	0.915 73	1.3411	−0.922	−0.021
0.1612	0.814 96	1.3316	−0.539	−0.013	0.7040	0.926 12	1.3408	−0.842	−0.020
0.2145	0.824 78	1.3331	−0.672	−0.016	0.7484	0.935 81	1.3403	−0.749	−0.018
0.2609	0.833 56	1.3344	−0.772	−0.018	0.7958	0.946 25	1.3394	−0.632	−0.015
0.3054	0.842 19	1.3356	−0.853	−0.020	0.8468	0.957 74	1.3381	−0.487	−0.012
0.3550	0.852 05	1.3369	−0.927	−0.022	0.8984	0.969 87	1.3364	−0.327	−0.009
0.4096	0.863 18	1.3382	−0.986	−0.023	0.9455	0.981 73	1.3346	−0.175	−0.005
0.4584	0.873 33	1.3392	−1.018	−0.024	1.0000	0.997 06	1.3325	0.000	0.000
0.4943	0.880 91	1.3398	−1.028	−0.024					
				Water (1) + Ethanol (2)					
0.0000	0.785 13	1.3592	0.000	0.000	0.5541	0.870 02	1.3624	−1.091	−0.029
0.0770	0.794 69	1.3603	−0.297	−0.011	0.6094	0.882 41	1.3619	−1.096	−0.029
0.1242	0.800 91	1.3610	−0.463	−0.016	0.6599	0.894 72	1.6311	−1.080	−0.028
0.1710	0.807 23	1.3616	−0.605	−0.020	0.7050	0.906 49	1.3601	−1.045	−0.026
0.2239	0.814 54	1.3621	−0.737	−0.023	0.7502	0.918 95	1.3585	−0.982	−0.024
0.2681	0.820 80	1.3625	−0.826	−0.025	0.8069	0.935 38	1.3556	−0.859	−0.021
0.3098	0.826 90	1.3627	−0.894	−0.027	0.8509	0.948 61	1.3524	−0.723	−0.018
0.3610	0.834 72	1.3628	−0.959	−0.028	0.9010	0.964 13	1.3474	−0.522	−0.013
0.3979	0.840 68	1.3629	−0.997	−0.029	0.9511	0.980 24	1.3407	−0.276	−0.007
0.4601	0.851 46	1.3628	−1.046	−0.029	1.0000	0.997 06	1.3324	0.000	0.000
0.5079	0.860 52	1.3626	−1.074	−0.030					
				1-Octanol (1) + Methanol (2)					
0.0000	0.786 61	1.3264	0.000	0.000	0.5427	0.814 37	1.4089	0.137	0.032
0.0488	0.790 89	1.3424	0.091	0.010	0.6017	0.815 69	1.4123	0.120	0.029
0.1025	0.795 08	1.3563	0.154	0.019	0.6369	0.816 41	1.4142	0.110	0.026
0.1513	0.798 44	1.3665	0.185	0.025	0.6860	0.817 34	1.4165	0.095	0.023
0.2029	0.801 57	1.3754	0.200	0.030	0.7406	0.818 28	1.4189	0.078	0.018
0.2541	0.804 29	1.3828	0.202	0.034	0.8021	0.819 24	1.4213	0.056	0.014
0.2945	0.806 20	1.3879	0.198	0.035	0.8306	0.819 66	1.4223	0.046	0.011
0.3485	0.808 44	1.3937	0.187	0.037	0.8831	0.820 36	1.4241	0.027	0.007
0.3942	0.810 11	1.3980	0.176	0.037	0.8263	0.820 89	1.4254	0.014	0.004
0.4505	0.811 90	1.4027	0.161	0.036	1.0000	0.821 67	1.4275	0.000	0.000
0.4960	0.813 19	1.4059	0.149	0.034					
				1-Octanol (1) + Ethanol (2)					
0.0000	0.785 19	1.3592	0.000	0.000	0.5512	0.812 90	1.4114	0.045	0.014
0.0505	0.789 42	1.3677	0.028	0.008	0.5983	0.814 13	1.4137	0.040	0.013
0.0996	0.793 03	1.3747	0.047	0.014	0.6476	0.815 32	1.4159	0.036	0.011
0.1499	0.796 31	1.3810	0.060	0.018	0.7001	0.816 48	1.4180	0.032	0.010
0.1983	0.799 12	1.3863	0.067	0.020	0.7449	0.817 39	1.4197	0.028	0.009
0.2549	0.802 05	1.3917	0.069	0.021	0.7906	0.818 26	1.4213	0.025	0.008
0.2943	0.803 89	1.3951	0.069	0.021	0.8527	0.819 36	1.4233	0.021	0.006
0.3448	0.806 03	1.3990	0.066	0.020	0.9064	0.820 24	1.4249	0.015	0.004
0.3958	0.807 99	1.4025	0.061	0.019	0.9491	0.820 91	1.4261	0.009	0.003
0.4435	0.809 66	1.4056	0.056	0.017	1.0000	0.821 67	1.4275	0.000	0.000
0.4954	0.811 30	1.4085	0.051	0.016					
				1-Octanol (1) + Water (2)					
0.0000	0.997 06	1.3325	0.000	0.000	0.8834	0.824 31	1.4262	−0.006	0.006
0.7410	0.828 47	1.4240	−0.012	0.012	0.9554	0.822 61	1.4270	−0.003	0.003
0.8486	0.825 22	1.4257	−0.008	0.008	1.0000	0.821 67	1.4275	0.000	0.000

Table III. Parameters and Standard Deviations of the Excess Volume–Composition Curves Fitted to the Data for the Binary Systems[a]

system	A_0	A_1	A_2	A_3	A_4	σ
water + methanol	−4.1148	−0.1325	0.5078	0.6222		0.0001
water + ethanol	−4.2794	−1.0176	−2.2113		1.5959	0.0001
1-octanol + methanol	0.5925	−0.5395	0.5784	−0.5686		0.0001
1-octanol + methanol	0.2005	−0.2190	0.2365			0.0001
1-octanol + water	−0.0625					0.0001

[a] Units of coefficients A_i and σ are cubic centimeters per mole.

by

$$\delta R = R - \sum_i x_i R_i \qquad (4)$$

Table II lists densities, refractive indices, excess molar volumes, and δR values for the binary systems studied. The excess volume and δR data were correlated with Redlich–Kister polynomials (*8*):

$$Y^E = x(1-x)\sum_{k=0}^{k} A_k(2x-1)^k \qquad k = 0, 1, 2, ... \qquad (5)$$

where Y^E is V^E or δR and x is the mole fraction of the first component of the mixture. Curves were fitted by the least-

Table IV. Parameters and Standard Deviations of the δR-Composition Curves Fitted to the Data for the Binary Systems[a]

system	A_0	A_1	A_2	σ
water + methanol	-0.0952			0.0001
water + ethanol	-0.1184		-0.0491	0.0001
1-octanol + methanol	0.1360	-0.0836		0.0001
1-octanol + ethanol	0.626	-0.0653	0.0614	0.0001
1-octanol + water	0.0629			0.0001

[a] Units of coefficients A_i and σ are cubic centimeters per mole.

Table V. Densities d, Refractive Indices n_D, Excess Volumes V^E, and δR Values for Water (1) + Methanol (2) + 1-Octanol (3) at 298.15 K

x_1	x_2	$d/(g\cdot cm^3)$	n_D	$V^E/(cm^3\cdot mol^{-1})$	$\delta R/(cm^3\cdot mol^{-1})$
0.0982	0.9018	0.803 70	1.3297	-0.355	-0.008
0.0880	0.8081	0.806 36	1.3599	-0.118	0.012
0.0785	0.7213	0.809 32	1.3778	-0.012	0.024
0.0684	0.6281	0.812 17	1.3912	0.039	0.032
0.0589	0.5412	0.814 36	1.4003	0.057	0.034
0.0491	0.4506	0.816 19	1.4076	0.063	0.032
0.0392	0.3599	0.817 68	1.4133	0.063	0.028
0.0293	0.2688	0.818 93	1.4179	0.055	0.022
0.0200	0.1834	0.819 94	1.4215	0.041	0.015
0.0104	0.0951	0.820 86	1.4246	0.019	0.007
0.2019	0.7981	0.822 43	1.3329	-0.642	-0.015
0.1809	0.7150	0.817 98	1.3630	-0.328	0.001
0.1606	0.6350	0.817 54	1.3810	-0.178	0.012
0.1407	0.5561	0.817 95	1.3932	-0.093	0.019
0.1200	0.4743	0.818 55	1.4024	-0.036	0.022
0.1016	0.4015	0.819 10	1.4086	-0.003	0.022
0.0798	0.3156	0.819 74	1.4143	0.018	0.020
0.0600	0.2370	0.820 30	1.4186	0.023	0.016
0.0413	0.1635	0.820 78	1.4218	0.020	0.012
0.0220	0.0871	0.821 21	1.4247	0.015	0.006
0.3001	0.6999	0.841 16	1.3357	-0.844	-0.020
0.2680	0.6249	0.828 98	1.3665	-0.469	-0.007
0.2388	0.5568	0.825 05	1.3835	-0.290	0.004
0.2085	0.4863	0.823 22	1.3954	-0.176	0.014
0.1798	0.4194	0.822 32	1.4037	-0.101	0.021
0.1547	0.3609	0.821 88	1.4092	-0.054	0.026
0.1203	0.2805	0.821 60	1.4152	-0.015	0.029
0.0905	0.2109	0.821 54	1.4192	0.002	0.028
0.0609	0.1419	0.821 54	1.4225	0.010	0.023
0.0335	0.0782	0.821 55	1.4249	0.014	0.015
0.3945	0.6055	0.860 09	1.3379	-0.972	-0.023
0.3538	0.5431	0.840 12	1.3683	-0.574	-0.007
0.3134	0.4809	0.332 33	1.3861	-0.373	0.005
0.2766	0.4245	0.828 53	1.3968	-0.250	0.012
0.2392	0.3671	0.826 06	1.4046	-0.150	0.016
0.1978	0.3036	0.824 30	1.4110	-0.068	0.018
0.1565	0.2402	0.823 25	1.4159	-0.022	0.018
0.1191	0.1828	0.822 68	1.4195	-0.006	0.015
0.0831	0.1276	0.822 30	1.4224	-0.002	0.011
0.0415	0.0636	0.821 92	1.4252	0.006	0.006
0.4884	0.5116	0.879 66	1.3396	-1.027	-0.024
0.4640	0.4860	0.862 15	1.3573	-0.795	-0.014
0.4389	0.4597	0.851 62	1.3706	-0.641	-0.007
0.2569	0.2691	0.827 58	1.4108	-0.128	0.010
0.2159	0.2261	0.825 86	1.4148	-0.076	0.010
0.1676	0.1756	0.824 42	1.4187	-0.041	0.010
0.0874	0.0916	0.822 78	1.4236	-0.005	0.008
0.0424	0.0444	0.822 15	1.4258	0.001	0.005
0.7107	0.2893	0.927 58	1.3409	-0.829	-0.020
0.2504	0.1019	0.827 88	1.4200	-0.052	0.009
0.2258	0.0919	0.827 01	1.4210	-0.042	0.009
0.1757	0.01715	0.825 47	1.4228	-0.026	0.008
0.1228	0.0500	0.824 10	1.4245	-0.013	0.007
0.0687	0.0279	0.822 91	1.4259	-0.003	0.004

squares method, and for each curve the number and significance of the coefficients were determined with an F test. The values of the coefficients and standard deviations of eq 5 for all the binary mixtures and both properties studied are listed in Tables III and IV.

Table VI. Densities d, Refractive Indices n_D, Excess Volumes V^E, and δR Values for Water (1) + Ethanol (2) + 1-Octanol (3) at 298.15 K

x_1	x_2	$d/(g\cdot cm^3)$	n_D	$V^E/(cm^3\cdot mol^{-1})$	$\delta R/(cm^3\cdot mol^{-1})$
0.0985	0.9015	0.797 50	1.3609	-0.375	-0.013
0.0878	0.8037	0.802 35	1.3775	-0.213	-0.002
0.0782	0.7160	0.806 08	1.3886	-0.126	0.006
0.0684	0.6262	0.809 32	1.3974	-0.072	0.012
0.0584	0.5350	0.812 06	1.4045	-0.037	0.017
0.0493	0.4508	0.814 19	1.4099	-0.016	0.019
0.0392	0.3585	0.816 18	1.4148	-0.001	0.019
0.0295	0.2695	0.817 84	1.4187	0.005	0.017
0.0198	0.1813	0.819 27	1.4220	0.006	0.014
0.0101	0.0921	0.820 54	1.4249	0.002	0.008
0.1954	0.8046	0.810 57	1.3620	-0.670	-0.021
0.1750	0.7205	0.811 21	1.3785	-0.422	-0.008
0.1548	0.6374	0.812 71	1.3903	-0.272	0.003
0.1350	0.5561	0.814 31	1.3990	-0.179	-0.011
0.1156	0.4760	0.815 76	1.4058	-0.116	0.016
0.0977	0.4025	0.816 94	1.4109	-0.072	0.019
0.0797	0.3284	0.818 01	1.4151	-0.039	0.020
0.0572	0.2355	0.819 20	1.4195	-0.013	0.019
0.0392	0.1614	0.820 07	1.4224	-0.004	0.015
0.0189	0.0778	0.820 96	1.4252	-0.003	0.008
0.2965	0.7035	0.824 93	1.3627	-0.874	-0.026
0.2654	0.6296	0.820 70	1.3798	-0.552	-0.013
0.2355	0.5587	0.819 49	1.3914	-0.366	-0.003
0.2061	0.4889	0.819 30	1.4000	-0.249	0.005
0.1770	0.4199	0.819 47	1.4065	-0.168	0.011
0.1488	0.3529	0.819 75	1.4116	-0.109	0.014
0.1187	0.2818	0.820 10	1.4161	-0.060	0.016
0.0890	0.2113	0.820 48	1.4197	-0.027	0.015
0.0653	0.1549	0.820 81	1.4221	-0.012	0.013
0.0386	0.0916	0.821 19	1.4246	-0.006	0.009
0.3990	0.6010	0.840 87	1.3631	-0.998	-0.029
0.3579	0.5392	0.830 75	1.3806	-0.621	-0.015
0.3170	0.4776	0.826 51	1.3927	-0.411	-0.003
0.2765	0.4165	0.824 40	1.4014	-0.278	0.006
0.2390	0.3601	0.823 26	1.4076	-0.189	0.012
0.2000	0.3012	0.822 52	1.4126	-0.118	0.016
0.1661	0.2501	0.822 13	1.4162	-0.073	0.018
0.1195	0.1800	0.821 84	1.4203	-0.033	0.017
0.0801	0.1207	0.821 72	1.4231	-0.014	0.014
0.0427	0.0643	0.821 66	1.4253	-0.002	0.009
0.4940	0.5060	0.857 81	1.3629	-1.067	-0.030
0.4391	0.4497	0.840 08	1.3827	-0.630	-0.014
0.3922	0.4018	0.833 37	1.3939	-0.431	-0.004
0.3427	0.3510	0.829 36	1.4025	-0.296	0.003
0.2961	0.3032	0.826 93	1.4085	-0.203	0.007
0.2496	0.2557	0.825 25	1.4132	-0.131	0.010
0.2010	0.2059	0.824 03	1.4171	-0.077	0.010
0.1485	0.1521	0.823 12	1.4206	-0.039	0.009
0.1004	0.1028	0.822 51	1.4232	-0.018	0.007
0.0554	0.0567	0.822 06	1.4253	-0.003	0.004
0.5954	0.4046	0.879 17	1.3621	-1.097	-0.029
0.5413	0.3679	0.854 68	1.3802	-0.683	-0.016
0.4937	0.3355	0.844 50	1.3913	-0.504	-0.008
0.3020	0.2053	0.828 28	1.4139	-0.139	0.005
0.2418	0.1643	0.826 10	1.4178	-0.079	0.004
0.1789	0.1215	0.824 53	1.4210	-0.049	0.003
0.1156	0.0785	0.823 32	1.4237	-0.029	0.001
0.0553	0.0376	0.822 33	1.4258	-0.003	-0.000
0.6985	0.3015	0.904 75	1.3600	-1.051	-0.027
0.6650	0.2871	0.882 31	1.3721	-0.807	-0.023
0.2642	0.1140	0.827 65	1.4194	-0.078	0.005
0.2147	0.0927	0.826 10	1.4214	-0.055	0.005
0.1477	0.0637	0.824 87	1.4237	-0.029	0.005
0.0847	0.0366	0.823 07	1.4255	-0.011	0.003

Tables V and VI list densities, refractive indices, excess molar volumes and δR values for the systems water + methanol + 1-octanol and water + ethanol + 1-octanol, respectively.

Figures 1 and 2 show the density and refractive index isolines for the ternary mixture water + methanol + 1-octanol at 298.15 K; these isolines are limited to the miscible region of the ternary mixture, the boundaries of which were established previously (9). Since the refractive index and-

Table VII. Parameters and Standard Deviations of the Excess Volume–Composition Surfaces Fitted to the Data for the Ternary Systems[a]

system	A	B_1	B_2	B_3	C_1	C_2	C_3	D_1	D_2	D_3	σ
water + methanol + 1-octanol	3.6121	−3.3100	−4.2043	−0.8944	−17.3551	6.4303	6.5305	−7.9618	−1.0085	−12.3591	0.003
water + ethanol + 1-octanol	5.1479	1.4337	−2.7274	−4.1612	−10.4407	1.7379	11.3323	−12.6561	−1.8198	−13.1419	0.003

[a] Units of coefficients A, B_i, C_i, and D_i and σ are cubic centimeters per mole.

Table VIII. Parameters and Standard Deviations of the δR–Composition Surfaces Fitted to the Data for the Ternary Systems[a]

system	A	B_1	B_2	B_3	C_1	C_2	C_3	σ
water + methanol + 1-octanol	−0.0858	−0.2709	−0.1429	0.1280				0.002
water + ethanol + 1-octanol	0.0174	−0.7316	−0.3762	0.3554	−1.5990	−1.4783	2.0406	0.002

[a] Units of coefficients A, B_i, C_i, and σ are cubic centimeters per mole.

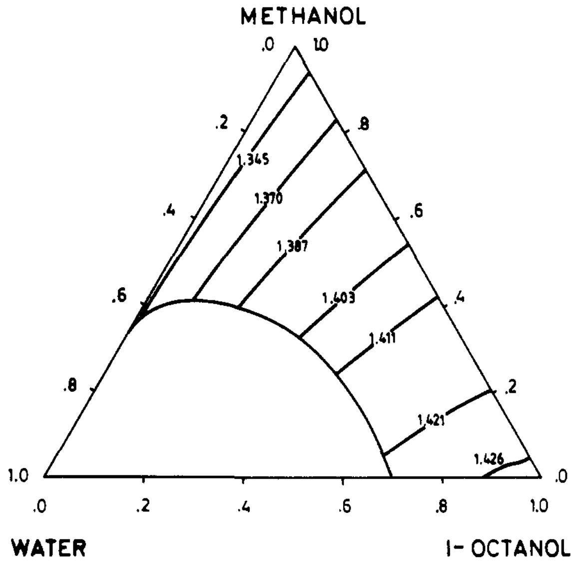

Figure 2. Refractive index isolines for the mixture water + methanol + 1-octanol at 298.15 K.

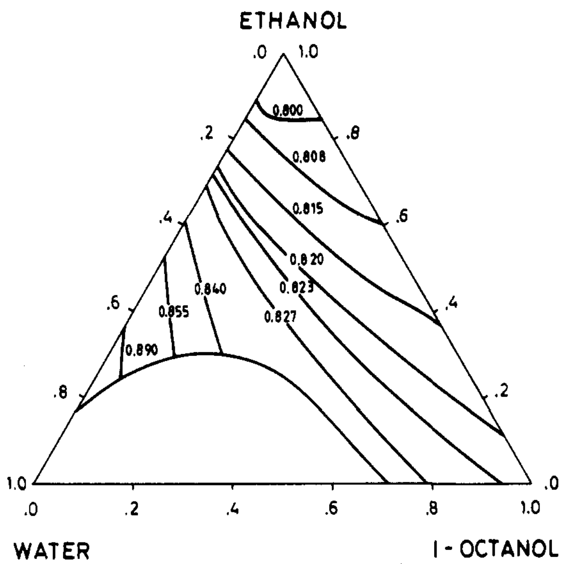

Figure 3. Density isolines of the mixture water + ethanol + 1-octanol at 298.15 K.

density isolines are approximately orthogonal to each other, measurements of the refractive index and density allow considerably accurate determination of the composition of an arbitrary water + methanol + 1-octanol mixture. Figures 3 and 4 show the density and refractive index isolines for the water + ethanol + 1-octanol system.

The excess volume and δR data for the systems water + methanol + 1-octanol and water + ethanol + 1-octanol were

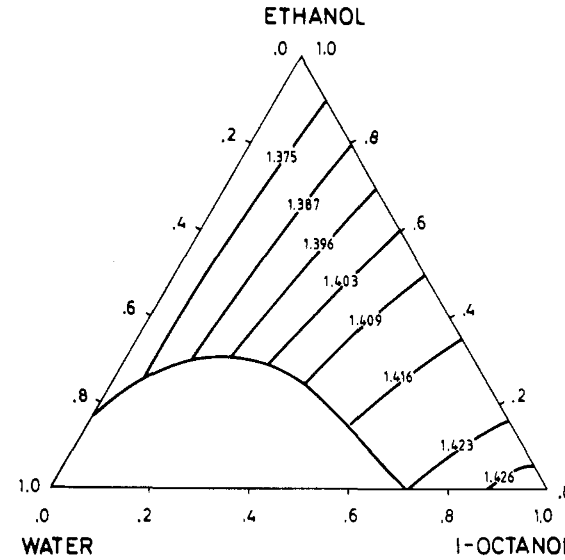

Figure 4. Refractive index isolines for the mixture water + ethanol + 1-octanol at 298.15 K.

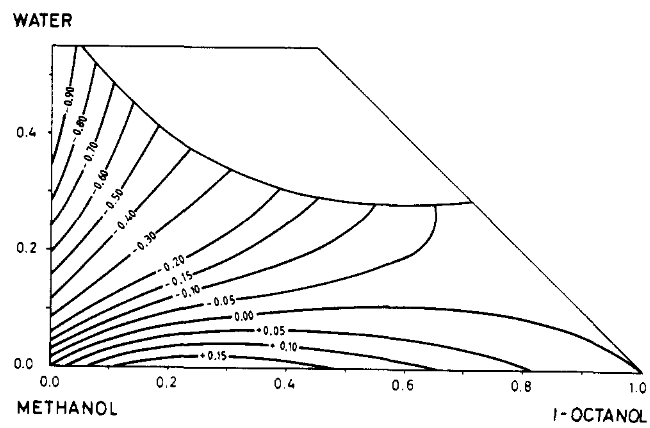

Figure 5. Excess volume isolines for the mixture water + methanol + 1-octanol at 298.15 K.

fitted with Redlich–Kister equations for ternary mixtures (*8*):

$$Y^E_{123} = Y^E_{12} + Y^E_{32} + Y^E_{31} + x_1x_2x_3(A + B_1(x_1 - x_2) + B_2(x_3 - x_2) + B_3(x_3 - x_1) + C_1(x_1 - x_2)^2 + C_2(x_3 - x_2)^2 + C_3(x_3 - x_1)^2 + ...) \quad (6)$$

where x_i is the mole fraction of component i in the ternary mixture and

$$Y^E_{ij} = x_ix_j\sum_k A_{kij}(x_i - x_j)^k$$

where the number of terms and coefficients A_{kij} is the same

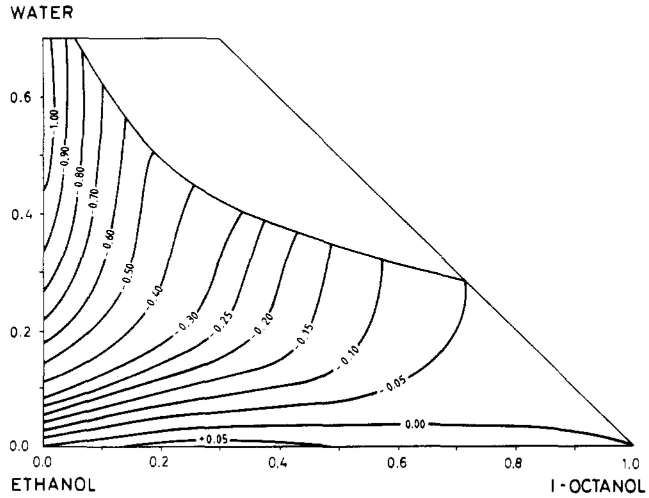

Figure 6. Excess volume isolines for the mixture water + ethanol + 1-octanol at 298.15 K.

as that of the Redlich–Kister equation for the binary mixture of components i and j (Tables III and IV). The values of the coefficients and standard deviations of eq 6 for the two ternary systems studied are listed in Tables VII and VIII for V^E and δR, respectively. Figures 5 and 6 show the excess molar volume isolines for these mixtures.

Discussion

The binary mixture water + methanol was negative V^E values with a minimum of -1.028 cm^3/mol and the binary mixture 1-octanol + methanol positive V^E values with a maximum of 0.202 cm^3/mol. The V^E of the ternary system water + methanol + 1-octanol varies between these limits.

The binary mixture water + ethanol has slightly more negative V^E values than water + methanol, with a minimum of -1.096 cm^3/mol. The binary mixture 1-octanol + ethanol has positive values lower than those of 1-octanol + methanol, with a maximum of 0.069 cm^3/mol. The V^E of the ternary system water + ethanol + 1-octanol is only positive for the low water contents.

The δR data follow the same general pattern as described above for V^E. However, the relatively small values of δR make its use difficult.

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