Short Articles

Densities and Viscosities of (1-Propanol + 1,2-Dichloroethane), (1-Propanol + Benzaldehyde), (Benzaldehyde + 1,2-Dichloroethane), and (1-Propanol + 1,2-Dichloroethane + Benzaldehyde) Mixtures from $T = 288.15$ K to 313.15 K

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Densities and viscosities of binary and ternary solvent systems which are composed of 1-propanol, 1,2-dichloroethane, and benzaldehyde have been measured in the temperature range (288.15 to 313.15) K and the over entire composition range and under atmospheric pressure. From experimental data, values of viscosity deviations, $\Delta\eta$, have been determined. The calculated binary data have been fitted to the Redlich−Kister equation to determine the appropriate coefficients. Also, the experimental results are used to determine the appropriate coefficients of the Cibulka, Singh, and Nagata equations. The results provide a test of the Grunberg and Nissan equation for correlating the dynamic viscosities of binary and ternary mixtures to mole fractions.

Introduction

Thermodynamic and transport properties are essential in process design and operation. Density and viscosity of multicomponent mixtures are required in many chemical engineering calculations involving fluid flow, heat, and mass transfer.[1,2] It is well-known that the solution of many engineering problems and the study of transport fluid phenomena require the knowledge of the dependence of kinematic viscosity on both temperature and composition of the systems. However, one cannot hope to have direct measurements for the multitude of complex systems of practical interest which justifies the theoretical and practical importance of prediction methods. Although a number of predictive equations[3] are available for estimating thermodynamic excess properties (excess volume, excess enthalpy, and excess free energy) of multicomponent systems, such methods are rarely used for viscosity. Excess molar volumes and viscosity deviation data are fitted to empirical equations.[4−6]

Recently, new models have been developed for the prediction of viscosities of mixtures. Some of them are based on the molecular approach,[7,8] while others are based on the group contribution concept.[9,10] The first models require binary interaction parameters for each binary system present in the multicomponent mixture, but no ternary (or higher) constants are generally needed. Geometrical solution models are considered to predict deviations of the viscosity for the ternary mixture from binary contribution because of dependence of interactions in ternary systems on the interaction in binary systems.[11−14]

Ternary mixtures are by far the easiest multicomponent systems to treat experimentally and clearly represent the most appropriate ground for prediction models.

The experimental data of excess thermodynamic properties of liquid mixtures provide useful information about molecular interactions and can be used to test thermodynamic models.[15−17] In this work, we want to provide a test of the Grunberg and Nissan models for correlating the kinematic viscosities of binary liquid mixtures with mole fractions. Also, this study may be useful in some industrial processes in the future. To this end, viscosities of the ternary mixture (1-propanol + 1,2-dichloroethane + benzaldehyde) and for the binary mixtures (1-propanol + 1,2-dichloroethane), (1,2-dichloroethane + benzaldehyde), and (benzaldehyde + 1-propanol) have been measured at the temperatures (288.15 to 313.15) K. The viscosity data have been used to calculate the viscosity deviations, $\Delta\eta$.

Table 1. Densities ρ and Viscosities η of Pure Components at Several Temperatures

		ρ/g·cm^{-3}		η/mPa·s	
component	T/K	this work	lit.	this work	lit.
1-propanol	288.15	0.8105		2.420	
	293.15	0.8065	0.80428[28]	2.137	2.1970[28]
	298.15	0.8032	0.80021[28]	1.893	1.9470[28]
	303.15	0.7993	0.79642[28]	1.684	1.7260[28]
	308.15	0.7952	0.79227[28]	1.503	1.5420[28]
	313.15	0.7904	0.78730[28]	1.351	1.3790[28]
1,2-dichloroethane	288.15	1.2590		0.870	
	293.15	1.2519	1.25210[29]	0.811	
	298.15	1.2474	1.24560[30]	0.759	0.7805[31]
	303.15	1.2390		0.711	
	308.15	1.2312		0.674	
	313.15	1.2220		0.626	
benzaldehyde	288.15	1.0560		1.704	
	293.15	1.0528	1.04361[32]	1.558	
	298.15	1.0470		1.427	
	303.15	1.0432		1.315	
	308.15	1.0390		1.222	
	313.15	1.0344		1.130	

Experimental Section

Materials. Extra pure 1-propanol (mole fraction > 99 %, Merck), 1,2-dichlorethane, (mole fraction > 99 %, Merck), and benzaldehyde (mole fraction > 98 %, Merck) were used. The purities of all

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Table 2. Densities ρ, Viscosities η, and Deviations of the Viscosity $\Delta\eta$ for the Binary and Ternary System of 1-Propanol (1) + Benzaldehyde (2) + 1,2-Dichloroethane (3) at 288.15 K as a Function of Mole Fraction x_1, x_2

x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s
0.1054	0.0000	1.2110	0.844	−0.189	0.1211	0.5362	1.0904	1.365	−0.282
0.2095	0.0000	1.1664	0.880	−0.313	0.1242	0.6415	1.0735	1.449	−0.320
0.3124	0.0000	1.1218	0.923	−0.430	0.1275	0.7523	1.0548	1.587	−0.309
0.4141	0.0000	1.0784	0.998	−0.513	0.1291	0.8099	1.0454	1.645	−0.316
0.5146	0.0000	1.0324	1.100	−0.567	0.2445	0.1403	1.1116	1.039	−0.364
0.6139	0.0000	0.9860	1.256	−0.565	0.2516	0.2475	1.1001	1.144	−0.388
0.7121	0.0000	0.9428	1.464	−0.509	0.2575	0.3377	1.0832	1.249	−0.391
0.8092	0.0000	0.8954	1.726	−0.398	0.2637	0.4323	1.0629	1.386	−0.368
0.9051	0.0000	0.8435	2.029	−0.243	0.2421	0.5581	1.0464	1.463	−0.396
0.0990	0.9010	1.0388	1.688	−0.327	0.2467	0.6370	1.0322	1.554	−0.399
0.1930	0.8070	1.0218	1.660	−0.397	0.2996	0.1608	1.0973	1.121	−0.390
0.3112	0.6888	0.9977	1.622	−0.488	0.3065	0.2467	1.0780	1.211	−0.405
0.3949	0.6051	0.9808	1.637	−0.511	0.3137	0.3366	1.0587	1.275	−0.451
0.5004	0.4996	0.9564	1.645	−0.550	0.2925	0.4316	1.0531	1.475	−0.323
0.5997	0.4003	0.9329	1.683	−0.557	0.2979	0.5055	1.0378	1.517	−0.370
0.6932	0.3068	0.9168	1.774	−0.508	0.3053	0.6083	1.0179	1.602	−0.410
0.8026	0.1974	0.8754	1.872	−0.459	0.4301	0.1388	1.0492	1.226	−0.463
0.9046	0.0954	0.8431	2.075	−0.302	0.4423	0.2448	1.0269	1.340	−0.485
0.0000	0.1005	1.2330	0.949	−0.031	0.4526	0.3339	1.0080	1.470	−0.469
0.0000	0.2068	1.2072	1.034	−0.063	0.4377	0.4522	0.9915	1.668	−0.378
0.0000	0.2964	1.1865	1.097	−0.099	0.5797	0.0972	0.9898	1.348	−0.526
0.0000	0.3902	1.1660	1.170	−0.129	0.5696	0.2001	0.9809	1.450	−0.523
0.0000	0.4888	1.1464	1.270	−0.138	0.5557	0.2870	0.9755	1.707	−0.340
0.0000	0.5923	1.1255	1.348	−0.174	0.5411	0.3783	0.9649	1.663	−0.462
0.0000	0.7012	1.1076	1.453	−0.188	0.6558	0.0968	0.9570	1.552	−0.440
0.0000	0.8159	1.0867	1.580	−0.188	0.6702	0.1780	0.9347	1.640	−0.464
0.0000	0.9061	1.0706	1.659	−0.208	0.6590	0.2634	0.9360	1.843	−0.337
0.1114	0.2055	1.1637	1.038	−0.230	0.7562	0.0962	0.9132	1.713	−0.434
0.1147	0.3173	1.1348	1.145	−0.251	0.7432	0.1567	0.9134	1.874	−0.320
0.1182	0.4359	1.1150	1.271	−0.262	0.8308	0.0958	0.8796	1.897	−0.365

Table 3. Densities ρ, Viscosities η, and Deviations of the Viscosity $\Delta\eta$ for the Binary and Ternary System of 1-Propanol (1) + Benzaldehyde (2) + 1,2-Dichloroethane (3) at 293.15 K as a Function of Mole Fraction x_1, x_2

x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s
0.1054	0.0000	1.2050	0.785	−0.166	0.1210	0.5368	1.0894	1.253	−0.243
0.2096	0.0000	1.1597	0.808	−0.280	0.1241	0.6420	1.0695	1.326	−0.276
0.3126	0.0000	1.1162	0.850	−0.375	0.1273	0.7527	1.0500	1.437	−0.278
0.4143	0.0000	1.0721	0.911	−0.449	0.1290	0.8103	1.0405	1.494	−0.279
0.5148	0.0000	1.0266	0.998	−0.495	0.2446	0.1406	1.1162	0.963	−0.309
0.6141	0.0000	0.9806	1.138	−0.487	0.2515	0.2479	1.0944	1.050	−0.336
0.7123	0.0000	0.9386	1.309	−0.445	0.2574	0.3383	1.0770	1.147	−0.335
0.8093	0.0000	0.8905	1.534	−0.350	0.2635	0.4329	1.0580	1.263	−0.320
0.9052	0.0000	0.8394	1.795	−0.216	0.2418	0.5587	1.0410	1.329	−0.348
0.0988	0.9012	1.0342	1.535	−0.287	0.2464	0.6375	1.0278	1.410	−0.350
0.1927	0.8073	1.0170	1.505	−0.350	0.2997	0.1611	1.0920	1.028	−0.338
0.3108	0.6892	0.9932	1.468	−0.428	0.3064	0.2471	1.0725	1.082	−0.376
0.3944	0.6056	0.9762	1.475	−0.451	0.3136	0.3371	1.0542	1.180	−0.376
0.5000	0.5000	0.9516	1.482	−0.480	0.2923	0.4322	1.0481	1.334	−0.286
0.5992	0.4008	0.9286	1.501	−0.496	0.2976	0.5060	1.0329	1.384	−0.316
0.6927	0.3073	0.9126	1.588	−0.441	0.3050	0.6088	1.0131	1.452	−0.358
0.8023	0.1977	0.8714	1.669	−0.398	0.4301	0.1391	1.0442	1.115	−0.401
0.9045	0.0955	0.8391	1.839	−0.264	0.4422	0.2452	1.0215	1.231	−0.406
0.0000	0.1007	1.2276	0.885	−0.024	0.4524	0.3344	1.0034	1.313	−0.424
0.0000	0.2072	1.2000	0.959	−0.054	0.4374	0.4527	0.9870	1.515	−0.318
0.0000	0.2969	1.1805	1.017	−0.084	0.5797	0.0974	0.9846	1.219	−0.455
0.0000	0.3909	1.1618	1.084	−0.108	0.5695	0.2005	0.9760	1.314	−0.448
0.0000	0.4894	1.1395	1.167	−0.122	0.5555	0.2874	0.9702	1.526	−0.302
0.0000	0.5929	1.1199	1.241	−0.149	0.5408	0.3787	0.9603	1.504	−0.394
0.0000	0.7017	1.1022	1.335	−0.161	0.6558	0.0970	0.9522	1.386	−0.388
0.0000	0.8163	1.0820	1.444	−0.164	0.6701	0.1783	0.9301	1.473	−0.400
0.0000	0.9063	1.0664	1.517	−0.179	0.6587	0.2637	0.9315	1.637	−0.304
0.1114	0.2059	1.1576	0.967	−0.192	0.7562	0.0964	0.9089	1.493	−0.415
0.1147	0.3179	1.1331	1.059	−0.214	0.7430	0.1569	0.9087	1.657	−0.291
0.1181	0.4366	1.1095	1.172	−0.222	0.8307	0.0959	0.8754	1.687	−0.318

the chemicals were checked by gas chromatography using a semi capillary methyl silicon column (o.d. 530 μm). The solvents were degassed by ultrasound and dried over molecular sieves (Sigma Union Carbide, 0.4 nm).

Apparatus and Procedures. The mixtures for the measurement of densities and viscosities were prepared just prior to the measurement to prevent any probable oxidation by air (in the case of benzaldehyde), and all the properties were

Table 4. Densities ρ,Viscosities η, and Deviations of the Viscosity $\Delta\eta$ for the Binary and Ternary System of 1-Propanol (1) + Benzaldehyde (2) + 1,2-Dichloroethane (3) at 298.15 K as a Function of Mole Fraction x_1, x_2

x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s
0.1054	0.0000	1.1992	0.732	−0.146	0.1211	0.5363	1.0850	1.152	−0.210
0.2095	0.0000	1.1544	0.747	−0.250	0.1242	0.6416	1.0644	1.219	−0.238
0.3124	0.0000	1.1093	0.783	−0.330	0.1274	0.7524	1.0455	1.315	−0.241
0.4141	0.0000	1.0671	0.836	−0.392	0.1291	0.8100	1.0369	1.372	−0.237
0.5146	0.0000	1.0213	0.911	−0.432	0.2446	0.1404	1.1107	0.889	−0.269
0.6140	0.0000	0.9762	1.035	−0.420	0.2516	0.2476	1.0890	0.970	−0.289
0.7122	0.0000	0.9340	1.176	−0.391	0.2575	0.3379	1.0717	1.058	−0.286
0.8092	0.0000	0.8859	1.369	−0.307	0.2636	0.4325	1.0528	1.149	−0.284
0.9052	0.0000	0.8363	1.602	−0.183	0.2420	0.5583	1.0363	1.211	−0.307
0.0990	0.9010	1.0299	1.404	−0.249	0.2466	0.6371	1.0232	1.292	−0.300
0.1929	0.8071	1.0128	1.373	−0.306	0.2996	0.1609	1.0861	0.941	−0.297
0.3111	0.6889	0.9892	1.337	−0.373	0.3065	0.2468	1.0670	0.996	−0.324
0.3948	0.6052	0.9713	1.333	−0.399	0.3136	0.3368	1.0486	1.093	−0.314
0.5003	0.4997	0.9470	1.341	−0.419	0.2924	0.4318	1.0430	1.214	−0.251
0.5996	0.4004	0.9237	1.362	−0.424	0.2978	0.5056	1.0280	1.274	−0.262
0.6930	0.3070	0.9083	1.423	−0.388	0.3053	0.6084	1.0087	1.324	−0.309
0.8025	0.1975	0.8676	1.490	−0.350	0.4301	0.1389	1.0386	1.012	−0.356
0.9046	0.0954	0.8353	1.639	−0.228	0.4423	0.2449	1.0159	1.148	−0.325
0.0000	0.1006	1.2205	0.821	−0.025	0.4525	0.3341	0.9986	1.201	−0.362
0.0000	0.2069	1.1961	0.895	−0.044	0.4376	0.4523	0.9824	1.337	−0.311
0.0000	0.2965	1.1746	0.947	−0.070	0.5797	0.0973	0.9796	1.110	−0.391
0.0000	0.3904	1.1555	1.008	−0.090	0.5696	0.2002	0.9710	1.199	−0.380
0.0000	0.4889	1.1334	1.080	−0.103	0.5557	0.2871	0.9660	1.357	−0.281
0.0000	0.5924	1.1173	1.151	−0.122	0.5411	0.3784	0.9556	1.343	−0.358
0.0000	0.7013	1.0986	1.232	−0.135	0.6558	0.0968	0.9470	1.225	−0.362
0.0000	0.8160	1.0779	1.328	−0.140	0.6702	0.1781	0.9252	1.334	−0.339
0.0000	0.9061	1.0631	1.394	−0.152	0.6589	0.2634	0.9268	1.452	−0.283
0.1114	0.2056	1.1524	0.894	−0.170	0.7562	0.0962	0.9043	1.381	−0.319
0.1147	0.3175	1.1276	0.979	−0.185	0.7431	0.1567	0.9040	1.473	−0.265
0.1182	0.4361	1.1045	1.078	−0.193	0.8307	0.0958	0.8707	1.509	−0.275

Table 5. Densities ρ, Viscosities η, and Deviations of the Viscosity $\Delta\eta$ for the Binary and Ternary System of 1-Propanol (1) + Benzaldehyde (2) + 1,2-Dichloroethane (3) at 303.15 K as a Function of Mole Fraction x_1, x_2

x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s
0.1056	0.0000	1.1926	0.684	−0.129	0.1211	0.5370	1.0804	1.063	−0.186
0.2099	0.0000	1.1477	0.700	−0.215	0.1241	0.6422	1.0600	1.126	−0.208
0.3129	0.0000	1.1042	0.725	−0.290	0.1273	0.7528	1.0413	1.211	−0.213
0.4146	0.0000	1.0611	0.770	−0.344	0.1290	0.8103	1.0323	1.260	−0.210
0.5151	0.0000	1.0160	0.834	−0.378	0.2448	0.1407	1.1051	0.822	−0.236
0.6144	0.0000	0.9705	0.934	−0.374	0.2517	0.2481	1.0838	0.899	−0.250
0.7125	0.0000	0.9297	1.061	−0.343	0.2575	0.3384	1.0676	0.983	−0.243
0.8095	0.0000	0.8826	1.228	−0.270	0.2637	0.4330	1.0489	1.051	−0.255
0.9053	0.0000	0.8328	1.441	−0.150	0.2419	0.5588	1.0320	1.125	−0.258
0.0989	0.9011	1.0259	1.291	−0.220	0.2465	0.6375	1.0189	1.186	−0.263
0.1927	0.8073	1.0081	1.258	−0.272	0.2999	0.1612	1.0804	0.868	−0.261
0.3108	0.6892	0.9848	1.224	−0.328	0.3067	0.2472	1.0632	0.928	−0.274
0.3945	0.6055	0.9670	1.216	−0.352	0.3137	0.3373	1.0445	1.018	−0.261
0.5000	0.5000	0.9429	1.222	−0.366	0.2924	0.4323	1.0378	1.100	−0.233
0.5993	0.4007	0.9195	1.236	−0.371	0.2977	0.5061	1.0230	1.176	−0.221
0.6928	0.3072	0.9040	1.287	−0.338	0.3051	0.6088	1.0043	1.215	−0.269
0.8023	0.1977	0.8636	1.342	−0.304	0.4304	0.1392	1.0340	0.916	−0.322
0.9045	0.0955	0.8316	1.490	−0.175	0.4424	0.2452	1.0112	1.042	−0.290
0.0000	0.1009	1.2143	0.770	−0.019	0.4525	0.3345	0.9944	1.115	−0.297
0.0000	0.2074	1.1900	0.836	−0.037	0.4375	0.4527	0.9783	1.207	−0.284
0.0000	0.2972	1.1698	0.884	−0.059	0.5800	0.0974	0.9753	1.017	−0.334
0.0000	0.3911	1.1501	0.951	−0.065	0.5697	0.2005	0.9663	1.102	−0.320
0.0000	0.4897	1.1265	1.002	−0.091	0.5556	0.2875	0.9613	1.215	−0.261
0.0000	0.5932	1.1127	1.070	−0.105	0.5409	0.3787	0.9511	1.232	−0.301
0.0000	0.7020	1.0944	1.142	−0.118	0.6560	0.0970	0.9434	1.117	−0.308
0.0000	0.8165	1.0732	1.225	−0.125	0.6702	0.1783	0.9207	1.218	−0.284
0.0000	0.9064	1.0585	1.285	−0.135	0.6588	0.2637	0.9221	1.293	−0.264
0.1115	0.2061	1.1468	0.832	−0.148	0.7563	0.0964	0.8999	1.254	−0.268
0.1148	0.3181	1.1228	0.909	−0.162	0.7431	0.1569	0.8995	1.314	−0.242
0.1182	0.4368	1.1002	1.001	−0.166	0.8308	0.0959	0.8662	1.359	−0.234

measured simultaneously. All binary and ternary mixtures were prepared by mass; the mass measurements were made using an electronic balance (AB 204-N Mettler) accurate to ± 0.01 mg. The uncertainty in mole fractions is estimated to be lower than $\pm\ 2 \cdot 10^{-4}$.

Densities were measured by an Anton Paar DMA-58 vibrating tube density meter. The density meter was calibrated with deionized doubly distilled water and dry air. Ubbelohde viscosimeters (Scott) of relatively long flow times [(60 to 600) s, with water] were used to minimize the kinetic energy correc-

Table 6. Densities ρ, Viscosities η, and Deviations of the Viscosity $\Delta\eta$ for the Binary and Ternary System of 1-Propanol (1) + Benzaldehyde (2) + 1,2-Dichloroethane (3) at 308.15 K as a Function of Mole Fraction x_1, x_2

x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s
0.1057	0.0000	1.1834	0.640	−0.122	0.1211	0.5375	1.0754	0.988	−0.162
0.2100	0.0000	1.1407	0.650	−0.198	0.1241	0.6426	1.0558	1.043	−0.184
0.3131	0.0000	1.0972	0.673	−0.261	0.1272	0.7531	1.0371	1.117	−0.190
0.4149	0.0000	1.0549	0.711	−0.307	0.1289	0.8105	1.0280	1.166	−0.183
0.5154	0.0000	1.0095	0.765	−0.336	0.2449	0.1410	1.0996	0.764	−0.212
0.6147	0.0000	0.9640	0.854	−0.330	0.2518	0.2484	1.0786	0.836	−0.220
0.7128	0.0000	0.9243	0.960	−0.305	0.2576	0.3388	1.0629	0.913	−0.212
0.8097	0.0000	0.8773	1.105	−0.240	0.2636	0.4334	1.0433	0.960	−0.236
0.9054	0.0000	0.8275	1.302	−0.122	0.2418	0.5592	1.0278	1.045	−0.221
0.0988	0.9012	1.0214	1.193	−0.195	0.2463	0.6379	1.0146	1.095	−0.231
0.1926	0.8074	1.0034	1.158	−0.242	0.3000	0.1614	1.0749	0.802	−0.234
0.3106	0.6894	0.9801	1.125	−0.289	0.3067	0.2476	1.0584	0.873	−0.228
0.3942	0.6058	0.9627	1.119	−0.306	0.3137	0.3377	1.0398	0.950	−0.221
0.4997	0.5003	0.9378	1.116	−0.322	0.2924	0.4327	1.0336	0.989	−0.231
0.5990	0.4010	0.9155	1.128	−0.324	0.2976	0.5065	1.0186	1.089	−0.187
0.6925	0.3075	0.8999	1.168	−0.295	0.3049	0.6091	0.9992	1.117	−0.236
0.8022	0.1978	0.8589	1.215	−0.263	0.4305	0.1394	1.0285	0.853	−0.276
0.9044	0.0956	0.8273	1.317	−0.174	0.4425	0.2455	1.0065	0.965	−0.248
0.0000	0.1011	1.2065	0.726	−0.019	0.4525	0.3348	0.9898	1.046	−0.238
0.0000	0.2078	1.1826	0.782	−0.038	0.4373	0.4530	0.9743	1.104	−0.250
0.0000	0.2976	1.1629	0.828	−0.055	0.5801	0.0976	0.9706	0.935	−0.288
0.0000	0.3917	1.1441	0.889	−0.059	0.5697	0.2007	0.9618	1.019	−0.267
0.0000	0.4903	1.1210	0.934	−0.083	0.5556	0.2877	0.9571	1.085	−0.251
0.0000	0.5937	1.1078	0.998	−0.092	0.5407	0.3790	0.9469	1.119	−0.269
0.0000	0.7025	1.0910	1.066	−0.100	0.6561	0.0971	0.9386	1.004	−0.281
0.0000	0.8168	1.0685	1.134	−0.112	0.6702	0.1785	0.9166	1.119	−0.236
0.0000	0.9066	1.0530	1.189	−0.120	0.6586	0.2640	0.9175	1.155	−0.250
0.1116	0.2064	1.1411	0.777	−0.134	0.7564	0.0965	0.8957	1.150	−0.218
0.1148	0.3185	1.1168	0.845	−0.147	0.7431	0.1571	0.8952	1.166	−0.234
0.1182	0.4373	1.0953	0.931	−0.147	0.8307	0.0960	0.8618	1.232	−0.198

Table 7. Densities ρ, Viscosities η, and Deviations of the Viscosity $\Delta\eta$ for the Binary and Ternary System of 1-Propanol (1) + Benzaldehyde (2) + 1,2-Dichloroethane (3) at 313.15 K as a Function of Mole Fraction x_1, x_2

x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	x_1	x_2	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s
0.1058	0.0000	1.1749	0.600	−0.103	0.1210	0.5381	1.0691	0.914	−0.146
0.2103	0.0000	1.1335	0.605	−0.173	0.1240	0.6432	1.0510	0.972	−0.158
0.3134	0.0000	1.0890	0.625	−0.228	0.1271	0.7536	1.0327	1.035	−0.169
0.4152	0.0000	1.0474	0.658	−0.269	0.1287	0.8109	1.0234	1.081	−0.161
0.5158	0.0000	1.0040	0.706	−0.294	0.2451	0.1413	1.0938	0.703	−0.192
0.6150	0.0000	0.9567	0.784	−0.287	0.2519	0.2489	1.0728	0.778	−0.191
0.7131	0.0000	0.9189	0.872	−0.270	0.2576	0.3394	1.0571	0.848	−0.183
0.8099	0.0000	0.8701	0.995	−0.218	0.2635	0.4340	1.0371	0.882	−0.215
0.9055	0.0000	0.8215	1.176	−0.107	0.2417	0.5598	1.0231	0.971	−0.191
0.0986	0.9014	1.0151	1.104	−0.175	0.2461	0.6384	1.0102	1.017	−0.198
0.1923	0.8077	0.9978	1.068	−0.218	0.3002	0.1618	1.0689	0.732	−0.216
0.3102	0.6898	0.9738	1.038	−0.257	0.3068	0.2480	1.0533	0.814	−0.194
0.3938	0.6062	0.9565	1.023	−0.279	0.3137	0.3382	1.0350	0.884	−0.187
0.4993	0.5007	0.9340	1.025	−0.285	0.2923	0.4333	1.0281	0.899	−0.218
0.5986	0.4014	0.9109	1.032	−0.286	0.2975	0.5070	1.0131	1.002	−0.166
0.6922	0.3078	0.8948	1.063	−0.263	0.3046	0.6096	0.9949	1.033	−0.206
0.8019	0.1981	0.8536	1.098	−0.237	0.4307	0.1396	1.0224	0.767	−0.261
0.9043	0.0957	0.8236	1.187	−0.156	0.4425	0.2459	1.0018	0.890	−0.215
0.0000	0.1013	1.1980	0.679	−0.012	0.4524	0.3353	0.9844	0.974	−0.196
0.0000	0.2083	1.1736	0.732	−0.028	0.4371	0.4535	0.9698	1.010	−0.225
0.0000	0.2983	1.1539	0.777	−0.042	0.5803	0.0977	0.9649	0.861	−0.248
0.0000	0.3924	1.1367	0.827	−0.052	0.5697	0.2011	0.9566	0.944	−0.225
0.0000	0.4910	1.1155	0.872	−0.070	0.5554	0.2881	0.9525	0.968	−0.246
0.0000	0.5945	1.1022	0.931	−0.078	0.5404	0.3794	0.9417	1.040	−0.222
0.0000	0.7031	1.0855	0.994	−0.085	0.6563	0.0973	0.9330	0.899	−0.265
0.0000	0.8173	1.0631	1.053	−0.100	0.6702	0.1788	0.9121	1.032	−0.195
0.0000	0.9069	1.0497	1.106	−0.104	0.6584	0.2643	0.9121	1.032	−0.242
0.1117	0.2069	1.1357	0.728	−0.112	0.7564	0.0967	0.8916	1.047	−0.190
0.1148	0.3191	1.1109	0.789	−0.126	0.7430	0.1573	0.8910	1.040	−0.226
0.1182	0.4380	1.0895	0.868	−0.126	0.8307	0.0962	0.8574	1.122	−0.168

tions. For each specified composition, five readings were taken for the flow time with variations not exceeding ± 0.01 s. The viscosities of all mixtures were calculated from the average flow time, while the viscosimeter constants were determined by using values from Marsh[18] for the water viscosity together with the corresponding flow times measured by the means of this viscosimeter. The uncertainties of the density and viscosity measurements were ± $8 \cdot 10^{-4}$ g·cm^{-3} and ± $6 \cdot 10^{-3}$ mPa·s. The viscosimeter was held in a Heidolph water bath whose temperature was controlled to within ± 0.01 K. The details of

Table 8. Coefficients of the Redlich−Kister's (A_p) of Equation 2 and Standard Deviations, σ, for Deviation Viscosity of Binary Systems at (288.15 to 313.15) K

T	A_0	A_1	A_2	A_3	σ
K	mPa·s	mPa·s	mPa·s	mPa·s	mPa·s
		1-Propanol + 1,2-Dichloroethane			
288.15	−2.2033	−0.6325	−0.2727	0.1893	0.0605
293.15	−1.9171	−0.5143	−0.2942	0.0793	0.0458
298.15	−1.6813	−0.4493	−0.2370	0.1443	0.0250
303.15	−1.4861	−0.6325	−0.1205	0.3611	0.0165
308.15	−1.3299	−0.5143	−0.0827	0.5583	0.0310
313.15	−1.1723	−0.4301	−0.0491	0.4915	0.0475
		Benzaldehyde + 1,2-Dichloroethane			
288.15	−0.5193	−0.1688	−1.1618	−1.6562	0.1528
293.15	−0.4478	−0.1457	−0.9835	−1.4657	0.1263
298.15	−0.3677	−0.1496	−0.8898	−1.1546	0.1103
303.15	−0.3062	−0.1541	−0.8009	−1.0235	0.0943
308.15	−0.2719	−0.0963	−0.7394	−0.9583	0.0817
313.15	−0.2299	−0.1009	−0.6171	−0.8551	0.0645
		1-Propanol + Benzaldehyde			
288.15	−2.0823	−0.4560	−2.1603	0.8192	0.1649
293.15	−1.8298	−0.3789	−1.8787	0.7143	0.1491
298.15	−1.6003	−0.3240	−1.6233	0.6066	0.1202
303.15	−1.4257	−0.3516	−1.2260	0.9263	0.0900
308.15	−1.2252	−0.1946	−1.2684	0.4513	0.0895
313.15	−1.0931	−0.1585	−1.1511	0.3821	0.0782

the methods and techniques of the measurements have been described earlier.[19−23] Data processing and curve fitting were made by Sigma plot ver. 8.2.160.

Results and Discussion

Tables 1 to 7 list the experimental kinematic viscosities and densities of the three pure solvents and their 33 ternary mixtures, along with their compositions expressed as mole fractions (x_i), at different temperatures. The viscosity deviations, $\Delta\eta$, for binary and ternary mixtures were determined using the equation

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

where η is the absolute viscosity of the mixture; η_i is the absolute viscosity of pure component i; x_i is the mole fraction component i; and N is the number of components in the mixture. The viscosity deviations for binary mixtures were fitted to a Redlich−Kister equation[24]

$$\Delta Y_{12} = x_1 x_2 \sum_{n=0}^{p} A_n (x_1 - x_2)^n \tag{2}$$

where ΔY is viscosity deviation and A_n are fitting coefficients which can be obtained by the least-squares method. For each isotherm, the standard deviations, σ, can be calculated from the following equation

$$\sigma = \left(\frac{1}{m - p} \sum_{i=1}^{m} (Y_{\text{exp}} - Y_{\text{cal}})^2 \right)^{1/2} \tag{3}$$

In this equation, m is the number of experimental data; p is the number of parameters; and Y_{exp} and Y_{cal} are the experimental and calculated property values, respectively. The values A_n and σ for the Redlich−Kister equation are given in Table 8.

The viscosity deviations for 1-propanol + 1,2-dichloroethane, benzaldehyde + 1-propanol, and benzaldehyde + 1,2-dichloroethane mixtures are plotted in Figures 1 to 3, respectively. The Grunberg and Nissan interaction model[25] has been used to correlate the dynamic viscosity of binary mixtures with mole fractions used as

$$\eta = \exp(x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12}) \tag{4}$$

where G_{12} is a parameter which is proportional to the interchange energy. The standard deviation and adjustable parameter of this equation for 1-propanol + 1,2-dichloroethane, benzaldehyde +

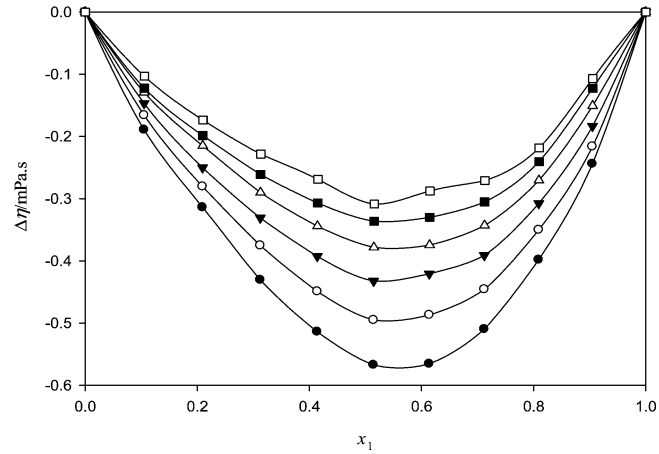

Figure 1. Viscosity deviations, $\Delta\eta$, for the binary mixture 1-propanol (1) + 1,2-dichloroethane (3) against 1-propanol mole fraction, x_1, at: ●, 288.15 K; ○, 293.15 K; ▲, 298.15 K; Δ, 303.15 K; ■, 308.15K; □, 313.15 K.

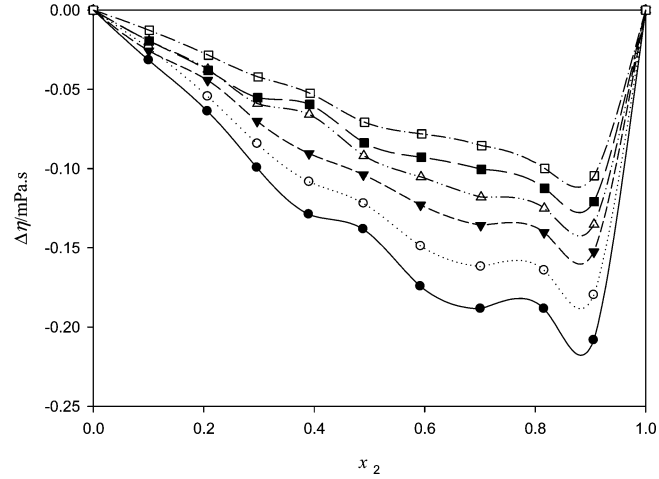

Figure 2. Viscosity deviations, $\Delta\eta$, for the binary mixture benzaldehyde (2) + 1,2-dichloroethane (3) against benzaldehaye mole fraction, x_2, at: ●, 288.15 K; ○, 293.15 K; ▲, 298.15K; Δ, 303.15 K; ■, 308.15 K; □, 313.15 K.

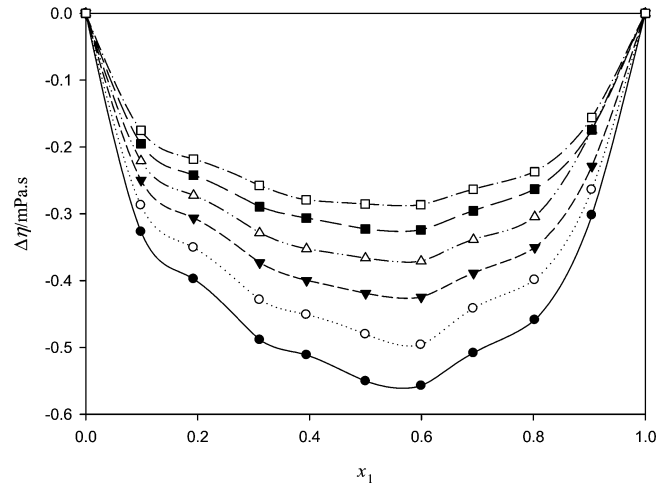

Figure 3. Viscosity deviations, $\Delta\eta$, for the binary mixture 1-propanol (1) + benzaldehyde (2) against 1-propanol mole fraction, x_1, at: ●, 288.15 K; ○, 293.15 K; ▲, 298.15 K; Δ, 303.15 K; ■, 308.15 K; □, 313.15 K.

Table 9. Adjustable Parameters (G_{12}) of Equation 4 and Standard Deviations for the Binary Mixtures of (1-Propanol + 1,2-Dichloroethane) and (Benzaldehyde + 1,2-Dichloroethane) and (1-Propanol + Benzaldehyde)

T	G_{12}	σ
K	mPa·s	mPa·s
1-Propanol + 1,2-Dichloroethane		
288.15	−1.1338	0.0214
293.15	−1.1322	0.0195
298.15	−0.1369	0.0345
303.15	−0.1065	0.0355
308.15	−1.1289	0.0189
313.15	−1.1155	0.0134
Benzaldehyde + 1,2-Dichloroethane		
288.15	−0.1649	0.0387
293.15	−0.1634	0.0368
298.15	−0.1369	0.0345
303.15	−0.1065	0.0355
308.15	−0.1091	0.0336
313.15	−0.06817	0.0342
1-Propanol + Benzaldehyde		
288.15	−1.2140	0.0307
293.15	−1.1936	0.0296
298.15	−1.1548	0.0261
303.15	−1.1229	0.0257
308.15	−1.0879	0.0266
313.15	−1.0668	0.0266

1-propanol, and benzaldehyde + 1,2-dichloroethane are given in Table 9. The viscosity deviations of viscous flow for the ternary mixture have been fitted to the Cibulka,[4] Singh,[5] and Nagata and Sakura[6] equations. The Cibulka,[4] Singh,[5] and Nagata and Sakura[6] are shown in eqs 5, 6, and 7, respectively.

Cibulka equation[4]

$$\Delta Y = \Delta Y_{12} + \Delta Y_{13} + \Delta Y_{23} + x_1x_2x_3(B_1 + B_2x_1 + B_3x_2) \quad (5)$$

Singh equation[5]

$$\Delta Y = \Delta Y_{12} + \Delta Y_{13} + \Delta Y_{23} + B_1x_1x_2x_3 + B_2x_1(x_2 - x_3) + B_3x_1^2(x_2 - x_3)^2 \quad (6)$$

Nagata and Sakura equation[6]

$$\Delta Y = \Delta Y_{12} + \Delta Y_{13} + \Delta Y_{23} + B_1x_1x_2x_3 \quad (7)$$

where in all equations B_1, B_2, and B_3 are fitting parameters. The fitted parameters, standard deviations obtained by the least-squares method, are given in Table 10.

According to Kauzman and Eyring,[26] the viscosity of a mixture strongly depends on the entropy of the mixture, which is related to liquid structure and enthalpy (and consequently to molecular interactions between the components of the mixture). So, the viscosity deviations are functions of interactions as well as the size and shape of molecules. Vogel and Weiss[27] affirm that mixtures with strong interactions between different molecules ($H^E < 0$ and negative deviations from Raoult's law) present positive viscosity deviations, whereas for mixtures with positive deviations of Raoult's law and without strong specific interactions, the viscosity deviations are negative. The breaking of hydrogen bonding of alcohols and dipole−dipole specific interactions cause the mixture to flow more hardly.

Deviations of viscosity, $\Delta\eta$, for the systems 1-propanol + 1,2-dichloroethane, benzaldehyde + 1-propanol, and benzaldehyde + 1,2-dichloroethane over the entire range of mole fractions and all temperatures are negative. The magnitudes of $\Delta\eta$ for binary systems in this investigation show that propanol + 1,2-dichloroethane ≈ 1-propanol and benzaldehyde > benzaldehyde + 1,2-dichloroethane. This means that viscosity behavior corresponds to a system in which there is an associated component: (solute−solute) complexes are formed and have high stability and (solute−solvent) interaction weakening this effect. The negative value of $\Delta\eta$ can be interpreted qualitatively by considering the strength of intermolecular hydrogen bonding, molecular size, and shapes of the components. The OH−Cl and OH−O interactions increase the viscosity, but the effect is not as important as the breaking of associations.

Table 10. Coefficients of the Cibulka Equation 4, Singh Equation 5, and Nagata Equation 6, B_P, and σ (Standard Deviations) for Deviation Viscosity of the Ternary System 1-Propanol + 1,2-Dichloroethane + Benzaldehyde at (288.15 to 313.15) K

	B_1	B_2	B_3	σ
equation	mPa·s	mPa·s	mPa·s	mPa·s
		T = 288.15 K		
Cibulka	−4.461	16.672	9.810	0.0543
Singh	3.7212	0.3076	2.859	0.0615
Nagata	4.5466	--	--	0.0731
		T = 293.15 K		
Cibulka	0.6796	5.660	2.8022	0.0409
Singh	3.2755	0.2690	2.584	0.0498
Nagata	4.0194	--	--	0.0612
		T = 298.15 K		
Cibulka	−2.634	11.574	6.551	0.0324
Singh	3.0089	0.21303	1.7855	0.0391
Nagata	3.5329	--	--	0.0471
		T = 303.15 K		
Cibulka	− 2.6571	10.755	6.205	0.0232
Singh	2.6671	0.18442	1.5202	0.0325
Nagata	3.1153	--	--	0.0397
		T = 308.15 K		
Cibulka	−2.0820	9.342	4.903	0.0199
Singh	2.3948	0.12370	1.3010	0.0300
Nagata	2.7663	--	--	0.0343
		T = 313.15 K		
Cibulka	−2.0813	8.486	4.919	0.0209
Singh	2.1868	0.13314	0.9792	0.0292
Nagata	2.4825	--	--	0.0328

Conclusion

New experimental values of density and viscosity for the systems ethyl valerate (1-propanol + 1,2-dichloroethane), (1-propanol + benzaldehyde), (benzaldehyde + 1,2-dichloroethane), and (1-propanol + 1,2-dichloroethane + benzaldehyde) from T = 288.15 K to 313.15 K were measured. The viscosity deviations were correlated using the Redlich−Kister equation, and a good agreement was observed. The coefficients of the Cibulka, Singh, and Nagata and Sakura equations seemed appropriate.

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