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Density and Viscosity of Binary Mixtures of Diethyl Carbonate with Alcohols at (293.15 to 363.15) K and Predictive Results by UNIFAC-VISCO Group Contribution Method

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Densities and viscosities of the binary mixtures of diethyl carbonate (DEC) with 1-propanol, 1-butanol, and 1-pentanol have been determined at (293.15 to 363.15) K and at atmospheric pressure. Excess molar volumes and viscosity deviations for the binary mixtures were fitted to the Redlich−Kister equation. Dependences of excess molar volumes and viscosity deviations with carbon chain length were discussed. The UNIFAC-VISCO method, based on contribution groups, has been used to calculate the dynamic viscosities of the binary mixtures. The percentage deviations between experimental and predicted values are given.

Introduction

In recent years, interest has been shown in the accumulation of thermodynamic properties of binary mixtures containing esters of carbonic acid. Accurate experimental data on density and viscosity of organic liquids and their binary mixtures are needed in many engineering disciplines. So density (ρ) and viscosity (η) for binary mixtures are important from both the practical and the theoretical viewpoint. As a continuation of the thermodynamic study of the mixture containing dialkyl carbonate,[1] we present here the experimental results of density and viscosity for the binary mixtures of diethyl carbonate with 1-propanol, 1-butanol, and 1-pentanol.

Up to now, Francesconi and Comelli[2] measured excess molar enthalpies and densities of DEC in binary mixtures with seven *n*-alkanols at 298.15 K. Rodriguez et al.[3,5] determined densities and viscosities of diethyl carbonate with alcohols at (293.15, 298.15, 303.15, and 313.15) K. Serious discrepancies were found in the viscosity data for 1-propanol, 1-butanol, and 1-pentanol, especially at higher temperatures. The aim of our work is to cover a wide range of temperatures found in industry about these properties. Measurements of the density and viscosity for the binary mixtures of diethyl carbonate with 1-propanol, 1-butanol, and 1-pentanol were performed at (293.15 to 363.15) K over the whole mole fraction range. The results were used to calculate the excess molar volumes and the deviations in viscosity over the entire mole fraction range for the mixtures. Experimental values were correlated by the Redlich−Kister equation. Comparison between experimental and literature data has also been made.

Experimental Section

Materials. All chemicals used in this study were supplied by Tianjin Reagent Co. except diethyl carbonate, which was supplied by Shanghai Chemical Reagent Co. They were dried over 0.4 nm molecular sieves and particularly degassed by ultrasound prior to their experimental use. The mass fraction purities tested by gas chromatography were as follows: diethyl carbonate (> 0.995), 1-propanol (> 0.996), 1-butanol (> 0.993), and 1-pentanol (0.991).

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

		$\rho/(\mathrm{g \cdot cm^{-3}})$		$\eta/(\mathrm{m \cdot Pas})$	
liquid	T/K	exptl	lit.	exptl	lit.
DEC	293.15	0.97500	0.9747[5]	0.800	0.795[3]
	298.15	0.96897	0.9691[5] 0.96916[4]	0.746	0.749[3]
	303.15	0.96239	0.9634[5]	0.695	0.699[3]
	313.15	0.95232	0.9522[5] 0.95243[6]	0.610	0.623[3]
	323.15	0.93978	0.94077[4]	0.541	
	333.15	0.92869		0.483	
	343.15	0.91633		0.435	
	353.15	0.90502		0.393	
	363.15	0.89371		0.358	
1-propanol	293.15	0.80362	0.8034[5] 0.80364[8]	2.238	2.198[3] 2.188[8]
	298.15	0.79941	0.7995[5] 0.7996[6]	1.981	1.970[3] 1.898[6]
	303.15	0.79527	0.7955[5] 0.7955[6] 0.79548[8]	1.745	1.707[3] 1.676[6] 1.713[8]
	313.15	0.78662	0.7873[5] 0.7875[6] 0.78702[8]	1.381	1.361[3] 1.319[6] 1.378[8]
	323.15	0.77391		1.115	
	333.15	0.76731		0.907	
	343.15	0.75595		0.742	
	353.15	0.74841		0.615	
	363.15	0.73970		0.515	
1-butanol	293.15	0.80982	0.8095[5] 0.80913[10]	2.963	2.941[3] 2.864[10]
	298.15	0.80606	0.8059[5] 0.8057[6] 0.80573[9]	2.619	2.620[3] 2.509[6] 2.5609[9]
	303.15	0.80203	0.8018[5] 0.8119[6] 0.80554[10]	2.298	2.255[3] 2.197[6] 2.273[10]
	313.15	0.79329	0.7945[5] 0.7941[6] 0.79438[9] 0.79432[10]	1.786	1.754[3] 1.706[6] 1.4567[9] 1.681[10]
	323.15	0.78655	0.78670[10]	1.421	1.090[10]
	333.15	0.77781		1.137	
	343.15	0.76703		0.920	
	353.15	0.75752		0.759	
	363.15	0.74938		0.630	
1-pentanol	293.15	0.81451	0.8142[5] 0.81412[7]	4.117	3.979[3] 4.109[7]
	298.15	0.81073	0.8101[5]	3.556	3.347[3]
	303.15	0.80677	0.8070[5] 0.80711[7]	3.060	2.932[3] 2.933[7]
	313.15	0.79905	0.7995[5] 0.79978[7]	2.332	2.256[3] 2.299[7]
	323.15	0.78981	0.79266[10]	1.822	1.304[10]
	333.15	0.78181		1.440	
	343.15	0.77284		1.146	
	353.15	0.76479		0.931	
	363.15	0.75717		0.767	

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

	ρ	η	V^E	$\Delta\eta$		ρ	η	V^E	$\Delta\eta$		ρ	η	V^E	$\Delta\eta$
x_1	g·cm^{-3}	mPa·s	cm^3·mol^{-1}	mPa·s	x_1	g·cm^{-3}	mPa·s	cm^3·mol^{-1}	mPa·s	x_1	g·cm^{-3}	mPa·s	cm^3·mol^{-1}	mPa·s
							(x_1) DEC + (1 − x_1) 1-Propanol							
							293.15 K							
0.0993	0.82906	1.668	0.0501	−0.428	0.4041	0.89162	1.010	0.1796	−0.648	0.6939	0.93674	0.830	0.1807	−0.411
0.2003	0.85200	1.347	0.1064	−0.603	0.5152	0.91018	0.915	0.2012	−0.582	0.8136	0.95251	0.801	0.1484	−0.267
0.3011	0.87249	1.147	0.1602	−0.658	0.5780	0.91995	0.877	0.2001	−0.530	0.8953	0.96277	0.791	0.0829	−0.160
							298.15 K							
0.0993	0.82424	1.490	0.0847	−0.368	0.4041	0.88622	0.920	0.2114	−0.562	0.6939	0.93088	0.766	0.2120	−0.358
0.2003	0.84698	1.211	0.1398	−0.523	0.5152	0.90462	0.838	0.2292	−0.507	0.8136	0.94653	0.743	0.1734	−0.234
0.3011	0.86735	1.037	0.1849	−0.572	0.5780	0.91427	0.808	0.2303	−0.459	0.8953	0.95659	0.737	0.1185	−0.138
							303.15 K							
0.0993	0.81957	1.328	0.1050	−0.313	0.4041	0.88050	0.838	0.2535	−0.482	0.6939	0.92462	0.709	0.2432	−0.307
0.2003	0.84186	1.089	0.1741	−0.445	0.5152	0.89865	0.771	0.2714	−0.433	0.8136	0.94017	0.690	0.1895	−0.200
0.3011	0.86183	0.938	0.2329	−0.490	0.5780	0.90817	0.745	0.2714	−0.393	0.8953	0.95013	0.685	0.1281	−0.119
							313.15 K							
0.0993	0.81034	1.073	0.1428	−0.231	0.4041	0.87074	0.709	0.2972	−0.360	0.6939	0.91466	0.614	0.2708	−0.232
0.2003	0.83249	0.896	0.2086	−0.330	0.5152	0.88879	0.658	0.3110	−0.326	0.8136	0.93007	0.602	0.2179	−0.151
0.3011	0.85234	0.783	0.2632	−0.365	0.5780	0.89827	0.639	0.3066	−0.296	0.8953	0.94008	0.600	0.1396	−0.091
							323.15 K							
0.0993	0.79804	0.887	0.1012	−0.171	0.4041	0.85781	0.611	0.3273	−0.272	0.6939	0.90180	0.540	0.3029	−0.177
0.2003	0.81974	0.751	0.2142	−0.249	0.5152	0.87596	0.571	0.3330	−0.248	0.8136	0.91735	0.531	0.2368	−0.117
0.3011	0.83943	0.666	0.2877	−0.276	0.5780	0.88537	0.557	0.3386	−0.226	0.8953	0.92747	0.532	0.1468	−0.070
							333.15 K							
0.0993	0.79059	0.739	0.1282	−0.127	0.4041	0.84879	0.529	0.3575	−0.207	0.6939	0.89152	0.478	0.3400	−0.135
0.2003	0.81183	0.636	0.2316	−0.187	0.5152	0.86632	0.500	0.3782	−0.189	0.8136	0.90675	0.473	0.2589	−0.089
0.3011	0.83100	0.572	0.3066	−0.208	0.5780	0.87552	0.490	0.3796	−0.172	0.8953	0.91655	0.474	0.1724	−0.053
							343.15 K							
0.0993	0.77886	0.618	0.1524	−0.093	0.4041	0.83656	0.462	0.4027	−0.156	0.6939	0.87897	0.425	0.3966	−0.104
0.2003	0.79980	0.542	0.2755	−0.138	0.5152	0.85395	0.440	0.4290	−0.144	0.8136	0.89418	0.423	0.3073	−0.069
0.3011	0.81879	0.493	0.3594	−0.156	0.5780	0.86305	0.433	0.4366	−0.131	0.8953	0.90410	0.425	0.1966	−0.041
							353.15 K							
0.0993	0.77055	0.524	0.1829	−0.069	0.4041	0.82650	0.406	0.4895	−0.119	0.6939	0.86815	0.381	0.4566	−0.079
0.2003	0.79085	0.466	0.3272	−0.105	0.5152	0.84354	0.391	0.5113	−0.109	0.8136	0.88316	0.382	0.3447	−0.052
0.3011	0.80927	0.430	0.4290	−0.118	0.5780	0.85242	0.386	0.5204	−0.100	0.8953	0.89295	0.385	0.2180	−0.031
							363.15 K							
0.0993	0.76119	0.448	0.2182	−0.052	0.4041	0.81600	0.361	0.5592	−0.091	0.6939	0.85716	0.345	0.5033	−0.061
0.2003	0.78098	0.405	0.3863	−0.079	0.5152	0.83279	0.350	0.5791	−0.085	0.8136	0.87199	0.347	0.3811	−0.041
0.3011	0.79900	0.378	0.5029	−0.090	0.5780	0.84153	0.347	0.5882	−0.078	0.8953	0.88169	0.350	0.2426	−0.025
							(x) DEC + (1 − x) 1-Butanol							
							293.15 K							
0.1016	0.83056	2.161	0.0870	−0.583	0.4008	0.88538	1.183	0.2347	−0.913	0.6951	0.93220	0.883	0.2029	−0.577
0.2023	0.84989	1.681	0.1671	−0.844	0.5036	0.90242	1.039	0.2462	−0.835	0.7490	0.94008	0.857	0.1877	−0.486
0.2974	0.86723	1.38	0.2203	−0.935	0.5959	0.91715	0.956	0.2249	−0.718	0.8989	0.96115	0.805	0.1118	−0.214
							298.15 K							
0.1016	0.82634	1.909	0.1107	−0.520	0.4008	0.88038	1.074	0.2665	−0.795	0.6951	0.92653	0.814	0.2366	−0.503
0.2023	0.84540	1.504	0.1932	−0.736	0.5036	0.89719	0.948	0.2779	−0.727	0.7490	0.93444	0.793	0.2046	−0.424
0.2974	0.86260	1.254	0.2366	−0.809	0.5959	0.91169	0.877	0.2595	−0.626	0.8989	0.95523	0.749	0.1244	−0.186
							303.15 K							
0.1016	0.82175	1.685	0.1428	−0.450	0.4008	0.87496	0.973	0.3007	−0.683	0.6951	0.92046	0.751	0.2618	−0.433
0.2023	0.84053	1.339	0.2252	−0.635	0.5036	0.89157	0.867	0.3054	−0.624	0.7490	0.92831	0.733	0.2206	−0.364
0.2974	0.85748	1.130	0.2687	−0.691	0.5959	0.90578	0.807	0.2939	−0.536	0.8989	0.94875	0.698	0.1405	−0.159
							313.15 K							
0.1016	0.81255	1.345	0.1789	−0.322	0.4008	0.86512	0.816	0.3639	−0.499	0.6951	0.91036	0.650	0.3115	−0.319
0.2023	0.83122	1.091	0.2567	−0.457	0.5036	0.88163	0.737	0.3639	−0.457	0.7490	0.91823	0.637	0.2592	−0.269
0.2974	0.84788	0.932	0.3189	−0.505	0.5959	0.89578	0.690	0.3463	−0.396	0.8989	0.93869	0.610	0.1552	−0.119
							323.15 K							
0.1016	0.80495	1.097	0.2044	−0.234	0.4008	0.85546	0.697	0.4187	−0.371	0.6951	0.89899	0.569	0.3713	−0.240
0.2023	0.82283	0.906	0.3015	−0.337	0.5036	0.87135	0.636	0.4211	−0.342	0.7490	0.90662	0.559	0.3115	−0.203
0.2974	0.83891	0.785	0.3639	−0.374	0.5959	0.88507	0.600	0.3913	−0.296	0.8989	0.92645	0.540	0.1870	−0.090
							333.15 K							
0.1016	0.79585	0.904	0.2168	−0.167	0.4008	0.84542	0.601	0.4560	−0.274	0.6951	0.88829	0.502	0.4087	−0.181
0.2023	0.81337	0.761	0.3265	−0.244	0.5036	0.86099	0.553	0.4684	−0.254	0.7490	0.89589	0.495	0.3365	−0.152
0.2974	0.82907	0.668	0.4062	−0.275	0.5959	0.87449	0.526	0.4410	−0.221	0.8989	0.91540	0.482	0.2119	−0.068
							343.15 K							
0.1016	0.78494	0.749	0.2119	−0.122	0.4008	0.83369	0.521	0.4934	−0.204	0.6951	0.87615	0.448	0.4410	−0.135
0.2023	0.80209	0.642	0.3463	−0.180	0.5036	0.84910	0.486	0.5058	−0.190	0.7490	0.88354	0.441	0.3862	−0.115
0.2974	0.81760	0.572	0.4311	−0.203	0.5959	0.86240	0.464	0.4860	−0.167	0.8989	0.90311	0.432	0.2242	−0.052
							353.15 K							
0.1016	0.77515	0.630	0.2226	−0.092	0.4008	0.82309	0.457	0.5379	−0.155	0.6951	0.86515	0.402	0.4710	−0.103
0.2023	0.79201	0.549	0.3711	−0.136	0.5036	0.83830	0.430	0.5537	−0.144	0.7490	0.87233	0.396	0.4327	−0.089
0.2974	0.80721	0.496	0.4729	−0.154	0.5959	0.85146	0.414	0.5310	−0.127	0.8989	0.89184	0.390	0.2434	−0.040
							363.15 K							
0.1016	0.76642	0.536	0.2441	−0.066	0.4008	0.81313	0.405	0.5962	−0.116	0.6951	0.85429	0.361	0.5332	−0.080
0.2023	0.78278	0.475	0.4144	−0.100	0.5036	0.82801	0.383	0.6146	−0.110	0.7490	0.86142	0.357	0.4792	−0.069
0.2974	0.79761	0.434	0.5257	−0.116	0.5959	0.84091	0.371	0.5901	−0.097	0.8989	0.88074	0.356	0.2562	−0.030

Table 2. Continued

x_1	ρ	η	V^E	$\Delta\eta$	x_1	ρ	η	V^E	$\Delta\eta$	x_1	ρ	η	V^E	$\Delta\eta$
	g·cm^{-3}	mPa·s	cm^3·mol^{-1}	mPa·s		g·cm^{-3}	mPa·s	cm^3·mol^{-1}	mPa·s		g·cm^{-3}	mPa·s	cm^3·mol^{-1}	mPa·s
(x_1) DEC + (1 − x_1) 1-Pentanol														
293.15 K														
0.1016	0.83180	2.879	0.0975	−0.901	0.3959	0.88031	1.443	0.2721	−1.361	0.7008	0.92863	0.957	0.2593	−0.836
0.2056	0.84916	2.155	0.1830	−1.280	0.5087	0.89840	1.201	0.2903	−1.228	0.7929	0.94302	0.883	0.2038	−0.604
0.2991	0.86458	1.735	0.2357	−1.390	0.6019	0.91321	1.065	0.2773	−1.055	0.9025	0.96001	0.825	0.1080	−0.299
298.15 K														
0.1016	0.82759	2.522	0.1248	−0.748	0.3959	0.87545	1.296	0.2994	−1.148	0.7008	0.92316	0.878	0.2767	−0.708
0.2056	0.84475	1.907	0.2066	−1.071	0.5087	0.89333	1.092	0.3121	−1.035	0.7929	0.93735	0.820	0.2194	−0.508
0.2991	0.85991	1.549	0.2666	−1.166	0.6019	0.90794	0.990	0.2979	−0.875	0.9025	0.95410	0.765	0.1212	−0.255
303.15 K														
0.1016	0.82306	2.206	0.1684	−0.613	0.3959	0.87032	1.170	0.3212	−0.954	0.7008	0.91725	0.809	0.2957	−0.593
0.2056	0.84003	1.690	0.2394	−0.884	0.5087	0.88789	0.994	0.3357	−0.863	0.7929	0.93119	0.755	0.2375	−0.430
0.2991	0.85497	1.386	0.2957	−0.967	0.6019	0.90228	0.891	0.3175	−0.745	0.9025	0.94776	0.712	0.1243	−0.213
313.15 K														
0.1016	0.81472	1.723	0.1957	−0.434	0.3959	0.86057	0.968	0.3683	−0.683	0.7008	0.90630	0.696	0.3248	−0.429
0.2056	0.83126	1.351	0.2648	−0.627	0.5087	0.87771	0.835	0.3738	−0.621	0.7929	0.91985	0.654	0.2648	−0.313
0.2991	0.84578	1.132	0.3230	−0.684	0.6019	0.89167	0.757	0.3593	−0.539	0.9025	0.93600	0.622	0.1419	−0.156
323.15K														
0.1016	0.80516	1.383	0.2235	−0.309	0.3959	0.85051	0.817	0.4109	−0.498	0.7008	0.89584	0.604	0.3677	−0.321
0.2056	0.82148	1.109	0.3035	−0.450	0.5087	0.86745	0.715	0.4249	−0.455	0.7929	0.90938	0.574	0.2921	−0.233
0.2991	0.83587	0.939	0.3630	−0.500	0.6019	0.88134	0.654	0.4019	−0.397	0.9025	0.92540	0.550	0.1671	−0.117
333.15 K														
0.1016	0.79672	1.122	0.2444	−0.220	0.3959	0.84097	0.697	0.4611	−0.364	0.7008	0.88540	0.533	0.4110	−0.236
0.2056	0.81244	0.918	0.3651	−0.325	0.5087	0.85754	0.618	0.4774	−0.335	0.7929	0.89870	0.508	0.3284	−0.173
0.2991	0.82667	0.790	0.4067	−0.363	0.6019	0.87115	0.570	0.4534	−0.294	0.9025	0.91450	0.490	0.1856	−0.086
343.15 K														
0.1016	0.78731	0.919	0.2629	−0.154	0.3959	0.83042	0.599	0.5021	−0.266	0.7008	0.87379	0.472	0.4555	−0.175
0.2056	0.80262	0.766	0.3928	−0.233	0.5087	0.84661	0.538	0.5185	−0.246	0.7929	0.88684	0.453	0.3623	−0.129
0.2991	0.81648	0.670	0.4426	−0.263	0.6019	0.85985	0.500	0.5013	−0.217	0.9025	0.90233	0.439	0.2089	−0.065
353.15 K														
0.1016	0.77885	0.764	0.2784	−0.112	0.3959	0.82082	0.520	0.5478	−0.197	0.7008	0.86323	0.421	0.4952	−0.133
0.2056	0.79374	0.650	0.4227	−0.170	0.5087	0.83658	0.473	0.5733	−0.184	0.7929	0.87604	0.407	0.3922	−0.097
0.2991	0.80726	0.576	0.4777	−0.193	0.6019	0.84957	0.443	0.5485	−0.163	0.9025	0.89131	0.395	0.2161	−0.050
363.15 K														
0.1016	0.77078	0.644	0.2944	−0.082	0.3959	0.81150	0.456	0.5926	−0.149	0.7008	0.85281	0.379	0.5356	−0.101
0.2056	0.78519	0.557	0.4555	−0.126	0.5087	0.82684	0.419	0.6191	−0.140	0.7929	0.86530	0.368	0.4279	−0.075
0.2991	0.79830	0.501	0.5209	−0.144	0.6019	0.83948	0.397	0.5949	−0.124	0.9025	0.88023	0.360	0.2393	−0.038

Apparatus and Procedure. The densities of the pure components and their mixtures were measured with a high precision vibrating-tube digital density meter (Density/Specific Gravity Meter DA 505, KEM, Japan), whose measurement cell temperature was controlled automatically within 0.01 K of the selected value. Before each series of measurements, the instrument was calibrated at atmospheric pressure with double distilled water and dry air. Densities of both water and dry air at the various working temperatures were given by the manufacturer in the instruction manual. The calibration was accepted if the measurements were within 5×10^{-5} g·cm^{-3} of the published values. The uncertainty in density measurements was 5×10^{-5} g·cm^{-3}. Density measurements were reproducible to 3×10^{-5} g·cm^{-3}.

The liquid mixtures were prepared by mass using a BP210s balance accurate to within ± 0.01 mg. The average uncertainty in the mole fraction of the mixtures was estimated to be less than ± 0.0001. The molar excess volumes were calculated from composition-density data with an uncertainty better than ± 0.002 cm^3·mol^{-1}. All molar quantities were based on the IUPAC relative atomic mass table.

The viscosities of pure liquids and the mixtures were measured at atmospheric pressure and at different temperatures using several Ubbelohde suspended-level viscometers. The viscometer was immersed in a well-stirred water bath (Lauda, Germany) with temperature control to within 0.01 K. An electronic digital stopwatch with a readability of 0.01 s was used for flow time measurement. Experiments were repeated a minimum of four times at each temperature for all compositions, and the results were averaged. The viscosity (η) of the liquid was then calculated from the following relationship:

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

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

The calibration of the viscometer was carried out with double distilled water and twice distilled benzene. Care was taken to reduce evaporation during the measurements. The uncertainty in the values is within 0.003 mPa·s.

In the experiment, the density and the viscosity for one composition sample were measured at different temperatures. Densities and viscosities of the pure compounds are reported in Table 1 together with the corresponding literature values. For the densities, good agreement was found between the measured and the literature values. Serious discrepancies were shown in the viscosity data for the alcohols. As shown in Table 1, there is an appreciable difference for the viscosity data among the literature. The viscosity values of DEC and pure alcohols obtained in this study are in good agreement with those of Rodriguez et al.,[3,5] and the average percentage deviation at 16 data points is 1.9 %.

Results and Discussion

The experimental values of density and viscosity for binary mixture at different temperatures and at atmospheric pressure are listed in Table 2. Excess volumes were calculated from our measurements according to the following equation:[11]

$$V^{\mathrm{E}} = (x_1M_1 + x_2M_2)/\rho - x_1M_1/\rho_1 - x_2M_2/\rho_2 \qquad (2)$$

where x_1 and x_2 are mole fractions, M_1 and M_2 are the molar masses, and ρ_1 and ρ_2 are the densities of pure components 1 and 2, respectively. Quantities without subscripts refer to the mixture.

The viscosity deviations were calculated from the following relation:[7]

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

where η is the viscosity of mixtures and η_1 and η_2 are the viscosity of components 1 and 2, respectively. The values of V^{E} and $\Delta\eta$ for each mixture were fitted to the Redlich−Kister polynomial equation:[12]

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

where Y = V^{E} or $\Delta\eta$, A_i are adjustable parameters, and x_1 is the fraction of component 1.

In each case, the optimum number of coefficients A_i was determined from an examination of the variation of the standard deviation:

$$\sigma(Y) = [\sum(Y_{\mathrm{cal}} - Y_{\mathrm{exp}})^2/(n - m)]^{(1/2)} \qquad (5)$$

where n is the total number of experimental values and m is the number of parameters. The excess molar volume data and the viscosity deviations are presented in Table 2. Table 3 lists the values of the parameters A_i together with the standard deviations.

Figure 1 shows the variation of V^{E} with x_1 for the binary mixtures of DEC + 1-propanol, 1-butanol, and 1-pentanol at (293.15 to 363.15) K, and comparison is made with previous literature data.[2,5] Our values are in agreement with those reported by Rodriguez et al.[5] except the excess molar volumes for the system of DEC + 1-pentanol at 313.15 K. Figure 2 shows the graphical variation of $\Delta\eta$ with x_1 for the binary mixtures of diethyl carbonate with the alcohols. There is a good agreement between the measured and literature values at all temperatures.

Excess molar volumes are positive in the entire composition range for all the systems studied. It can be summarized from our series work[11−13] that V^{E} values may be affected by three factors. The first factor is the specific forces between molecules, such as hydrogen bonds, charge-transfer complexes, breaking of hydrogen bonds, and complexes giving positive excess molar volumes and forming hydrogen and complexes bringing negative values of V^{E}. The second factor is the physical intermolecular forces, including electrostatic forces between charged particles and between permanent dipoles and so on, induction forces between a permanent dipole and an induced dipole, and forces of attraction (dispersion forces) and repulsion between nonpolar molecules. Physical intermolecular forces are usually weak, and the sign of V^{E} values may be positive or negative, but the absolute values are small. The third factor is the structural characteristics of the component, arising from geometrical fitting of one component into the other's structure, due to the differences in shape and size of components and free volume. For diethyl carbonate + alcohols systems, the main contributions to V^{E} are chemical. Diethyl carbonate is a polar liquid with molecules of small size having dipole−dipole interactions in pure liquid. In contrast, alcohols are self-associated through hydrogen bonding. When these two components are mixed, the hydrogen bonding in alcohols are broken, and dipole−dipole interactions between diethyl carbonate with alcohol are formed. Because the interaction strength of the hydrogen is stronger than the dipole interaction, the V^{E} values are positive for all diethyl carbonate with alcohols. With an increase in temperature, the interactions between molecules become weak and the V^{E} values increase.

Table 3. Coefficients of the Redlich−Kister Equation and Standard Deviation for Excess Molar Volumes V^{E} and Viscosity Deviations $\Delta\eta$

T/K	property	A_0	A_1	A_2	A_3	A_4	σ
(x_1) DEC + (1 − x_1) 1-Propanol							
293.15	V^{E}/cm^3·mol^{-1}	0.8100	0.2094	−0.0919			0.0433
	$\Delta\eta$/mPa·s	−2.4068	1.6416	−0.0630	−0.1592	−2.7081	0.0746
298.15	V^{E}/cm^3·mol^{-1}	0.8992	0.1992	0.3136			0.0159
	$\Delta\eta$/mPa·s	−2.0892	1.4253	−0.1000	−0.1841	−2.2396	0.0650
303.15	V^{E}/cm^3·mol^{-1}	1.0757	0.1156	0.2925			0.0213
	$\Delta\eta$/mPa·s	−1.7896	1.2191	−0.0877	−0.1682	−1.8731	0.0546
313.15	V^{E}/cm^3·mol^{-1}	1.2129	0.0036	0.4897			0.0547
	$\Delta\eta$/mPa·s	−1.3434	0.8907	−0.0458	−0.1373	−1.3893	0.0405
323.15	V^{E}/cm^3·mol^{-1}	1.3839	0.2138	0.0061			0.0657
	$\Delta\eta$/mPa·s	−1.0196	0.6581	−0.0724	−0.1155	−0.9404	0.0295
333.15	V^{E}/cm^3·mol^{-1}	1.5086	0.2302	0.1932			0.0202
	$\Delta\eta$/mPa·s	−0.7757	0.4929	−0.0499	−0.1035	−0.6630	0.0204
343.15	V^{E}/cm^3·mol^{-1}	1.7335	0.2440	0.2926			0.0258
	$\Delta\eta$/mPa·s	−0.5893	0.3559	−0.0196	−0.0926	−0.4961	0.0161
353.15	V^{E}/cm^3·mol^{-1}	2.0686	0.1771	0.1964			0.0212
	$\Delta\eta$/mPa·s	−0.4493	0.2721	−0.0157	−0.0907	−0.3482	0.0139
363.15	V^{E}/cm^3·mol^{-1}	2.3444	0.0774	0.2777			0.0352
	$\Delta\eta$/mPa·s	−0.3458	0.1972	−0.0148	−0.0576	−0.2478	0.0085
(x_1) DEC + (1 − x_1) 1-Butanol							
293.15	V^{E}/cm^3·mol^{-1}	0.9622	−0.1886	0.2616	0.5679	−0.0913	0.0198
	$\Delta\eta$/mPa·s	−3.3465	2.0495	−1.2980	0.7508	−0.4851	0.0166
298.15	V^{E}/cm^3·mol^{-1}	1.0987	−0.1253	0.0676	0.3384	0.3728	0.0271
	$\Delta\eta$/mPa·s	−2.9187	1.7084	−0.9759	0.9095	−0.8287	0.0178
303.15	V^{E}/cm^3·mol^{-1}	1.2334	−0.1590	−0.0650	0.2162	0.9004	0.0354
	$\Delta\eta$/mPa·s	−2.5008	1.4524	−0.8664	0.8582	−0.7104	0.0153
313.15	V^{E}/cm^3·mol^{-1}	1.4860	−0.1084	−0.3952	−0.0780	1.4725	0.0412
	$\Delta\eta$/mPa·s	−1.8376	1.0443	−0.5750	0.5373	−0.5380	0.0114
323.15	V^{E}/cm^3·mol^{-1}	1.6945	−0.0844	−0.1088	−0.0466	1.2867	0.0550
	$\Delta\eta$/mPa·s	−1.3705	0.7598	−0.4518	0.3525	−0.3028	0.0070
333.15	V^{E}/cm^3·mol^{-1}	1.8873	−0.1400	−0.3080	0.1680	1.6290	0.0618
	$\Delta\eta$/mPa·s	−1.0207	0.5407	−0.3053	0.2162	−0.1779	0.0075
343.15	V^{E}/cm^3·mol^{-1}	2.0331	−0.0716	0.0168	0.2487	0.8657	0.0262
	$\Delta\eta$/mPa·s	−0.7625	0.3953	−0.2211	0.1261	−0.1233	0.0018
353.15	V^{E}/cm^3·mol^{-1}	2.2163	−0.0823	0.0913	0.3688	0.7011	0.0119
	$\Delta\eta$/mPa·s	−0.5797	0.2893	−0.1617	0.1093	−0.1002	0.0034
363.15	V^{E}/cm^3·mol^{-1}	2.4582	−0.0388	0.2881	0.2041	0.2570	0.0095
	$\Delta\eta$/mPa·s	−0.4409	0.1971	−0.1336	0.0775	−0.0071	0.0046
(x_1) DEC + (1 − x_1) 1-Pentanol							
293.15	V^{E}/cm^3·mol^{-1}	1.1483	0.1161	0.2190	−0.0234	−0.3443	0.0159
	$\Delta\eta$/mPa·s	−6.1756	2.0362	3.9909			0.2272
298.15	V^{E}/cm^3·mol^{-1}	1.2480	0.0742	0.1806	−0.0994	0.0153	0.0230
	$\Delta\eta$/mPa·s	−5.4104	1.7474	3.7530			0.2080
303.15	V^{E}/cm^3·mol^{-1}	1.3344	0.1329	0.3013	−0.6116	0.2491	0.0416
	$\Delta\eta$/mPa·s	−4.6071	1.4316	3.3537			0.1795
313.15	V^{E}/cm^3·mol^{-1}	1.5095	0.1701	0.0414	−0.7631	0.8374	0.0540
	$\Delta\eta$/mPa·s	−3.4974	1.0316	2.7764			0.1478
323.15	V^{E}/cm^3·mol^{-1}	1.7031	0.1466	−0.0634	−0.7617	1.2489	0.0416
	$\Delta\eta$/mPa·s	−2.6208	0.7224	2.1880			0.1140
333.15	V^{E}/cm^3·mol^{-1}	1.8925	0.0602	0.3825	−0.6697	0.6406	0.0355
	$\Delta\eta$/mPa·s	−1.9480	0.5046	1.6804			0.0858
343.15	V^{E}/cm^3·mol^{-1}	2.0721	0.1077	0.3881	−0.6881	0.7606	0.0328
	$\Delta\eta$/mPa·s	−1.4388	0.3433	1.2724			0.0635
353.15	V^{E}/cm^3·mol^{-1}	2.2732	0.1577	0.3367	−0.8477	0.6652	0.0404
	$\Delta\eta$/mPa·s	−1.0721	0.2374	0.9567			0.0468
363.15	V^{E}/cm^3·mol^{-1}	2.4588	0.1306	0.4299	−0.7218	0.5992	0.0365
	$\Delta\eta$/mPa·s	−0.8049	0.1647	0.7215			0.0344

As seen in Figure 2, the values of $\Delta\eta$ for all studied systems are negative over the entire range of mole fractions at all temperatures, and the curves are asymmetrical in nature and skewed to the alcohol-rich range. Similar to the excess molar volumes, viscosity is related to the molecular interaction between the components of mixtures as well as of the size and shape of molecules. Positive values of $\Delta\eta$ are indicative of strong interactions whereas negative values indicate weaker interactions.[13] The negative deviations in viscosity support the main factor of breaking of the self-associated alcohols and weak interactions between unlike molecules.

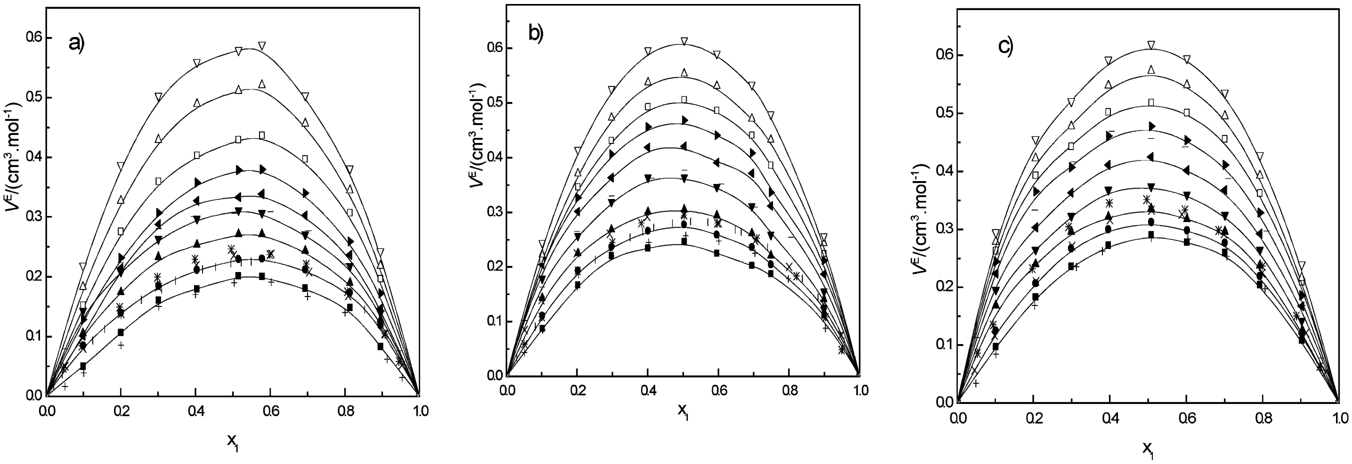

Figure 1. Excess molar volume variation with mole fraction for the following systems: (a) (x_1) DEC + $(1 - x_1)$ 1-propanol; (b) (x_1) DEC + $(1 - x_1)$ 1-butanol; (c) (x_1) DEC + $(1 - x_1)$ 1-pentanol at 293.15 K (■, this work; +, Rodriguez et al.[5]); 298.15 K (● this work, ×, Rodriguez et al.;[5] |, Francesconi and Comelli[2]); 303.15 K (▲ this work; *, Rodriguez et al.[5]); 313.15 K (▼, this work; −, Rodriguez et al.[5]); 323.15 K (left-facing solid triangle, this work); 333.15 K (right-facing solid triangle, this work); 343.15 K (□, this work); 353.15 K (△, this work); 363.15 K (▽, this work); solid curves, calculated with Redlich−Kister equations; symbols, experimental values.

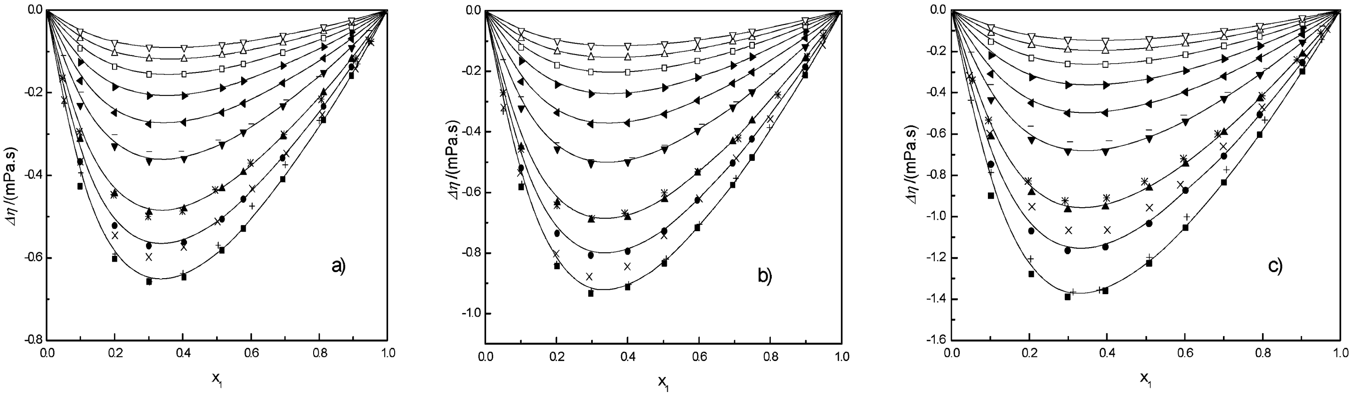

Figure 2. Viscosity deviation with mole fraction for the following systems: (a) (x_1) DEC + $(1 - x_1)$ 1-propanol; (b) (x_1) DEC + $(1 - x_1)$ 1-butanol; (c) (x_1) DEC + $(1 - x_1)$ 1-pentanol at 293.15 K (■, this work; +, Rodriguez et al.[5]); 298.15 K (● this work, ×, Rodriguez et al.[3]); 303.15 K (▲, this work; *, Rodriguez et al.[5]); 313.15 K (▼, this work; −, Rodriguez et al.[5]); 323.15 K (left-facing solid triangle, this work); 333.15 K (right-facing solid triangle, this work); 343.15 K (□, this work); 353.15 K (△, this work); 363.15 K (▽, this work); solid curves, calculated with Redlich−Kister equations; symbols, experimental values.

Dependence of the Excess Molar Volume and Viscosity Deviation with the Carbon Chain Length. The dependences of the excess molar volume with the carbon chain length of alcohols at the equimolar mixture composition are plotted in Figure 3. It can be seen, at the same temperatures, that the excess molar volumes increase with the carbon chain length of the alcohols, and the excess molar volumes at different temperatures show parabolic curves. The increasing of V^E with the carbon chain length of alcohols may be related to the increase of the size of alcohols.

Figure 4 shows the variation of viscosity deviations with the carbon chain length at the equimolar fraction. The dependence of viscosity deviations with the carbon chain length shows curves at the lower temperatures and decreases with carbon chain length. However, at the higher temperatures, the dependence of the viscosity deviation with the carbon chain length is almost a straight line and decreases with the carbon chain length.

Predictive Results by Rodriguez's UNIFAC-VISCO Method. Chevalier et al.[14] and Rodriguez et al.[15,16] have used the UNIFAC-VISCO method, based on contribution groups, to calculate the dynamic viscosities of the mixtures at different temperatures. Interaction parameters carbonate−alcohol and carbonate−alkane have been determined from their experimental data at (293.15, 298.15, 303.15, and 313.15) K by them. These parameters were used directly to predict the viscosity of the binary mixtures diethyl carbonate with 1-propanol, 1-butanol, and 1-pentanol at (293.15 to 363.15) K.

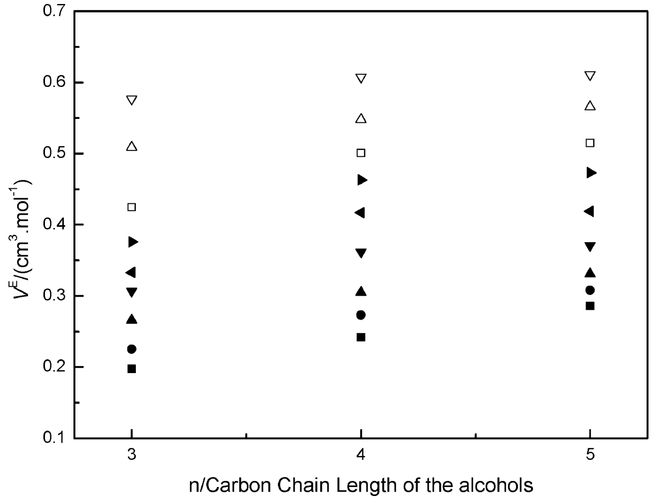

Figure 3. Dependence of the excess molar volume with carbon chain length of the alcohols at the equimolar fraction: ■, 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 313.15 K; left-facing solid triangle, 323.15 K; right-facing solid triangle, 333.15 K; □, 343.15 K; △, 353.15 K; ▽, 363.15 K.

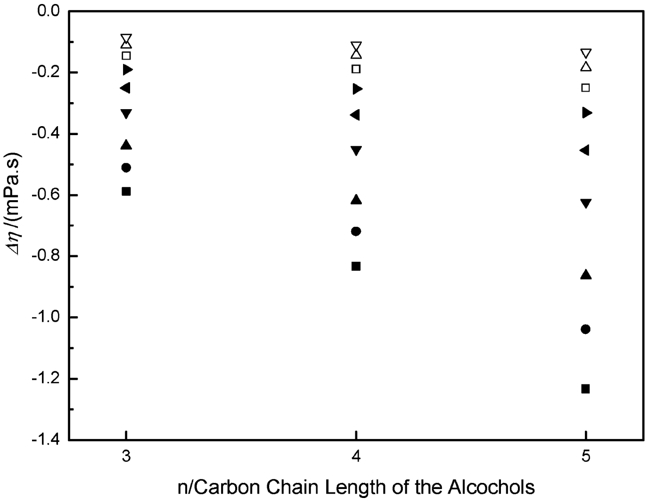

Figure 4. Dependence of the viscosity deviation with carbon chain length of the alcohols at the equimolar fraction: ■, 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 313.15 K; left-facing solid triangle, 323.15 K; right-facing solid triangle, 333.15 K; □, 343.15 K; △, 353.15 K; ▽, 363.15 K.

Table 4. Percent Deviation of Viscosity Calculated from Experimental Values with UNIFAC-VISCO Model for the Binary Mixtures over the Temperature Range (293.15 to 363.15) K

	T/K								
x	293.15	298.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15
				(x_1) DEC + (1 − x_1) 1-Propanol					
0.0993	−4.32	−3.64	−2.43	−1.09	−0.93	0.42	1.76	3.03	4.00
0.2003	−4.83	−4.16	−2.64	−0.66	0.08	2.02	4.13	5.80	7.45
0.3011	−4.06	−3.37	−2.01	0.28	1.62	3.92	6.05	8.21	10.04
0.4041	−3.23	−2.42	−0.98	1.50	3.19	5.24	7.61	9.65	11.67
0.5152	−1.79	−1.10	0.55	2.68	4.12	6.38	8.34	10.52	12.19
0.5780	−1.09	−0.15	1.24	3.26	4.61	6.66	8.51	10.44	12.08
0.6939	−0.17	0.34	1.49	3.26	4.44	6.25	7.62	9.24	10.65
0.8136	0.01	0.35	1.23	2.52	3.15	4.43	5.46	6.75	7.57
0.8953	−0.37	0.05	0.49	1.39	1.81	2.61	3.18	4.06	4.58
				(x_1) DEC + (1 − x_1) 1-Butanol					
0.1016	−1.44	−2.30	−2.31	−0.98	0.47	1.80	2.98	3.47	4.86
0.2023	−2.17	−2.02	−1.99	0.09	1.91	4.01	5.68	6.94	8.85
0.2974	−2.40	−1.61	−0.85	1.14	3.31	5.51	7.70	9.33	11.15
0.4008	−1.26	−0.76	−0.05	2.39	4.86	7.20	9.43	11.20	13.28
0.5036	−0.55	−0.08	0.99	3.56	5.69	7.88	10.11	11.95	13.66
0.5959	0.77	1.19	2.37	4.47	6.29	8.29	10.08	11.81	13.33
0.6951	0.64	1.04	1.95	4.00	5.47	7.07	9.03	10.64	11.62
0.7490	0.87	1.29	2.17	3.97	5.12	6.64	8.06	9.23	10.13
0.8989	−0.07	0.25	0.93	1.63	2.21	3.08	3.66	4.44	5.17
				(x_1) DEC + (1 − x_1) 1-Pentanol					
0.1016	−3.33	−2.53	−1.09	0.01	0.45	1.49	3.09	6.50	4.99
0.2056	−4.02	−3.08	−1.30	0.58	1.93	3.65	5.69	9.50	8.90
0.2991	−2.09	−1.07	0.68	3.23	4.33	6.29	8.58	12.24	12.15
0.3959	0.30	1.03	2.77	4.99	6.62	8.64	10.77	14.06	14.08
0.5087	0.68	1.72	3.36	5.57	7.20	9.15	11.24	14.31	14.52
0.6019	−4.15	−1.29	−1.27	1.23	3.20	5.44	7.71	10.80	11.53
0.7008	−0.87	−0.12	1.27	3.38	4.32	6.47	8.27	10.55	11.24
0.7929	3.77	4.77	5.01	6.21	6.92	8.02	9.10	10.61	10.79
0.9025	−1.46	−1.24	−0.40	0.54	1.15	2.19	2.93	3.84	4.28

Figure 5 shows graphically the viscosities and the calculated values, obtained from the UNIFAC-VISCO method, for the binary mixtures diethyl carbonate with 1-propanol, 1-butanol, and 1-pentanol at (293.15 to 363.15) K.

The predicted values of viscosity were compared with the experimental values. The percentage deviations (*E* %) were calculated:

$$E\,\% = (\eta_{\text{exp}} - \eta_{\text{calc}}) \times 100/\eta_{\text{exp}} \tag{6}$$

where η_{exp} and η_{calc} are the experimental values of viscosity and calculated values, respectively. Table 4 lists the percentage deviation. It can be seen that good results are obtained while the UNIFAC-VISCO method is used to calculated the dynamic viscosities of the binary mixtures of diethyl carbonate with 1-propanol, 1-butanol, and 1-pentanol at (293.15, 298.15, 303.15, and 313.15) K. The average absolute errors at 36 data points are within 2.0 %; however, the predictive results are worse for the temperatures at (333.15, 343.15, 353.15, and 363.15) K. The average errors were in excess of 6.0 %. The higher the temperatures are, the larger are the percentage deviations. One reason is that the group interaction parameters used have been obtained from the experimental viscosity data in the literature at (293.15, 298.15, 303.15, and 313.15) K. On the other hand, the values of viscosity at higher temperatures are small comparatively. All these result in the worse prediction of values.

Conclusion

In this paper, the densities and viscosities of the binary systems of DEC + 1-propanol, + 1-butanol, and + 1-pentanol have been experimentally determined in temperature range (293.15 to 363.15) K and for the whole composition range. Comparisons with literature data have been made for the pure substances and the binary mixtures. For the densities, good agreement was found between the measured and the literature values. Serious discrepancies were shown in the viscosity data for the alcohols.

The excess molar volumes and viscosity deviations are computed and have been fitted to Redlich−Kister equation.

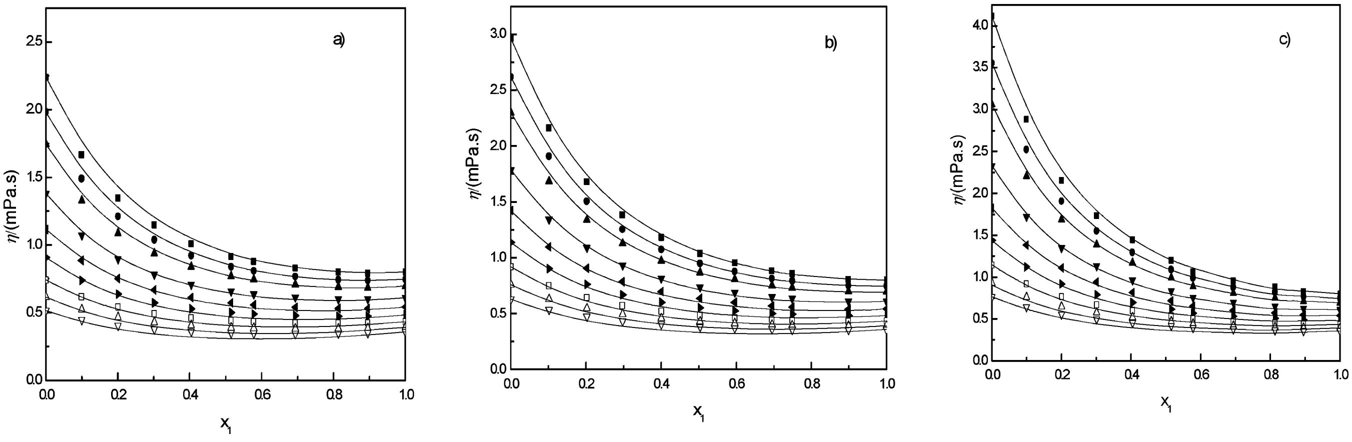

Figure 5. Experimental viscosity data *η* with mole fraction for the binary mixtures of diethyl carbonate with (a) 1-propanol, (b) 1-butanol (2), and (c) 1-pentanol (2) at ■ 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 313.15 K; left-facing solid triangle, 323.15 K; right-facing solid triangle, 333.15 K; □, 343.15 K; △, 353.15 K; ▽, 363.15 K; solid curves, calculated by using the UNIFAC-VISCO method; symbols, experimental values.

Excess molar volumes and viscosity deviation show a systematic change with increasing temperature. V^E values are positive over the entire composition range for all studied systems and increase with increasing temperature. The deviations of viscosity ($\Delta\eta$) are negative for all the mixtures over the entire composition range and become less negative with increasing temperature.

The excess molar volumes increase with carbon chain length of the alcohols and show parabolic curves at the different temperatures. The plot of the viscosity deviations with carbon chain length shows curves at the lower temperatures and decrease with carbon chain length. However, at the higher temperatures, the dependences of the deviation with carbon chain length are almost straight line and decrease with the carbon chain length.

The UNIFAC-VISCO predictive method has been used to obtain calculated values of the viscosity at different temperatures. Due to the interaction parameters directly obtained from literature, which were calculated by experimental values at (293.15, 298.15, 303.15, and 313.15) K, the accurately predicted results are obtained over this temperature range. However, the worse results are obtained at (333.15, 343.15, 353.15, and 363.15) K.

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