Excess Molar Enthalpies and Excess Molar Volumes of Binary Mixtures Containing Dimethyl Carbonate + Four Butanol Isomers at (288.15, 298.15, and 313.15) K

Romolo Francesconi*

Dipartimento di Chimica "G. Ciamician", Universita' degli Studi, via Selmi 2, I-40126 Bologna, Italy

Fabio Comelli

Centro di Studio per la Fisica delle Macromolecole del CNR, via Selmi 2, I-40126 Bologna, Italy

Excess molar enthalpies, H_m^E, and excess molar volumes, V_m^E, of dimethyl carbonate + 1-butanol, + 2-butanol, + 2-methyl-1-butanol, and + 2-methyl-2-butanol have been determined at (288.15, 298.15, and 313.15) K and atmospheric pressure using a flow microcalorimeter and a digital density meter. Excess properties are positive over the entire experimental conditions, increasing as the temperature increases. Maxima values of H_m^E vary from 2100 up to 2900 J·mol^{-1}. Those of V_m^E vary from 0.57 up to 0.83 cm^3·mol^{-1}. Experimental data have been correlated using Redlich−Kister polynomials, and parameters from least-squares analysis have been reported.

Introduction

Estimations of molecular interactions of binary mixtures and information needed to test existing theories of solutions may be related to the magnitude of excess thermodynamic properties such as excess molar enthalpies, H_m^E, and excess molar volumes, V_m^E.

The thermodynamic study of esters of carbonic acid is arousing an increasing interest owing to their uses in extractions of industrial importance, for many synthetic and natural resins and polymers (Annesini et al.,1984; Merck Index, 1989), in the synthesis of pharmaceuticals (Martindale, 1989), and in agricultural chemistry.

A part of our research program involves the study of binary mixtures containing dialkyl carbonates and a series of organic compounds or structural isomers with the purpose to obtain information on the interactions between molecules (Comelli and Francesconi, 1997; Francesconi et al., 1997).

This paper reports experimental H_m^E and V_m^E for dimethyl carbonate + four butanol isomers, namely, 1-butanol, 2-butanol, 2-methyl-1-propanol, and 2-methyl-2-propanol.

Measurements have been made at (288.15, 298.15, and 313.15) K with the exception for the mixture containing 2-methyl-2-propanol, whose data were carried out only at 313.15 K, its melting point being ≈298.15 K.

No excess enthalpic or volumetric data have been found in the literature on the mixtures studied in this paper.

Experimental Section

Chemicals. Dimethyl carbonate, an Aldrich product, stated purity 99 mol % (from purchaser), was fractionally distilled using the same procedure described previously (Francesconi and Comelli, 1996). Butanol isomers were from Aldrich with the exception of 2-butanol, which was a Merck product, and were used without further purification owing to their high-grade purity.

Purities of all products were checked by using a Hewlett-Packard gas chromatograph model 5890 supplied by an HP (cross-linked 5% ME siloxane) capillary column.

Purities, density values, and comparison with literature are reported in Table 1.

Table 1. Mole Percent Purities, Densities, ρ, and Refractive Indices, n_D, of Pure Components and Comparison with Literature Values at 298.15 K

	ρ/g·cm^{-3}		n_D	
component (purity/mol %)	this paper	lit.	this paper	lit.
dimethyl carbonate (99.8)	1.063 07	1.063 06[a] 1.063 50[b]	1.3667	1.366 7[b]
1-butanol (99.8)	0.805 77	0.805 75[c] 0.805 6[d] 0.805 85[e]	1.3975	1.397 41[c] 1.397 6[d]
2-butanol (99.6)	0.802 53	0.802 41[c] 0.802 3[d]	1.3952	1.395 3[c] 1.395 2[d]
2-methyl-1-propanol (99.5)	0.797 82	0.797 8[c]	1.3938	1.393 89[c]
2-methyl-2-propanol (99.5)	0.76484[f]	0.764 9[c,f]	1.3841[g]	1.384 0[c,g]

[a] Negadi et al. (1993). [b] Garcia de la Fuente et al. (1992). [c] Riddick et al. (1986). [d] Tanaka et al. (1992). [e] Artigas et al. (1994). [f] At 313.15 K. [g] At 300.15 K.

Refractive indices, n_D, were determined using an Abbe refractometer (Carl Zeiss, Jena, Switzerland) with a resolution of ±0.0001 refractive unit and are also shown in Table 1. Before measurements, pure liquids were degassed by ultrasound (ultrasonic bath, Hellma, type 460, Milan, Italy), kept in dark bottles, and dried over molecular sieves (Union Carbide, type $^1/_{16}$ in. pellets).

Apparatus and Procedure. Excess molar enthalpies, H_m^E, were determined using a flow microcalorimeter (LKB Produkter, model 2107, Bromma, Sweden).

* To whom correspondence should be sent.

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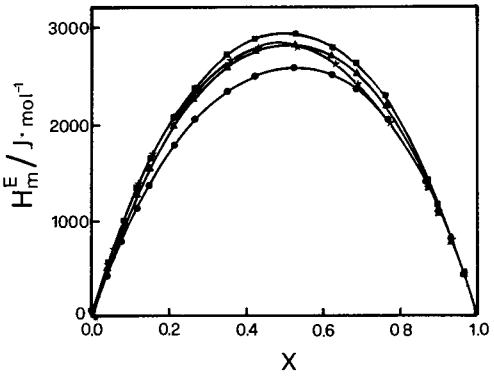

Figure 1. Excess molar enthalpies, H^E_m, of dimethyl carbonate (1) + 1-butanol (2) (●), + 2-butanol (2) (■), + 2-methyl-1-propanol (2) (▲), and + 2-methyl-2-propanol (2) (★) at 313.15 K. Solid curves, calculated by eq 2.

Electrical calibration, details of the equipment, and the operating procedure have been described previously (Monk and Wadso, 1968; Francesconi and Comelli, 1986).

Before measurements, the apparatus performance was checked by measuring the excess molar enthalpies of the standard mixture cyclohexane + hexane for which literature values are known (Gmehling, 1993). The values of H^E_m obtained in this paper were found to be in good agreement (less than 0.5% over most of the mole fraction range of cyclohexane) with those of Gmehling.

Densities, ρ, and excess molar volumes, V^E_m, were determined using an Anton Paar digital density meter (model DMA 60/602, Graz, Austria), as described elsewere (Fermeglia and Lapasin, 1988).

Mole fractions were determined by mass using a digital balance (Mettler, model AE 160) with an accuracy of $\pm 1 \times 10^{-4}$.

The apparatus was calibrated with twice-distilled water and dry air at each mixture. Temperature was controlled by a calibrated digital thermometer (Anton Paar, DT 100-15;100-25;100-40) with an accuracy of ±0.01 K.

The overall precision of the density measurements is estimated to be on the order of 1×10^{-5} g·cm^{-3}, which leads to an error in V^E_m of less than 3×10^{-3} cm^3·mol^{-1}.

Before measurements, the density meter was checked against literature data of the mixture benzene + cyclohexane (Wilhelm, 1985): an agreement of our data of better than 0.5% in the central range of mole fraction of benzene was found.

Results and Discussion

The experimental H^E_m and V^E_m of the four binary mixtures are reported in Tables 2 and 3, respectively, at (288.15, 298.15, and 313.15) K.

The values of V^E_m were derived from densities using the following formula

$$V^E_m = (x_1M_1 + x_2M_2)/\rho - x_1M_1/\rho_1 - x_2M_2/\rho_2 \qquad (1)$$

where x_i, M_i, and ρ_i are the molar fraction, molar mass, and density of component i, respectively.

The Redlich−Kister equation

$$Q^E_m = x_1x_2\sum_{k\geq 0} a_k(x_1 - x_2)^k \qquad (2)$$

where $Q^E_m = H^E_m$/(J·mol^{-1}) or V^E_m/(cm^3·mol^{-1}) with all

Table 2. Experimental Values of Excess Molar Enthalpies, H^E_m, of Dimethyl Carbonate + Butanol Isomers at (288.15, 298.15, and 313.15) K

x_1	H^E_m/ J·mol^{-1}	x_1	H^E_m/ J·mol^{-1}	x_1	H^E_m/ J·mol^{-1}
Dimethyl Carbonate (1) + 1-Butanol (2)					
T = 288.15 K					
0.0434	318	0.3525	1943	0.7656	1742
0.0832	619	0.4205	2061	0.8673	1239
0.1198	867	0.5213	2131	0.8970	1010
0.1536	1113	0.6204	2092	0.9289	728
0.2140	1442	0.6853	1978	0.9631	407
0.2663	1668				
T = 298.15 K					
0.0433	327	0.3519	2118	0.7651	1874
0.083	621	0.4199	2251	0.8670	1304
0.1195	871	0.5206	2357	0.8968	1083
0.1532	1119	0.6196	2287	0.9287	796
0.2135	1493	0.6912	2134	0.9630	446
0.2657	1760				
T = 313.15 K					
0.0431	423	0.3509	2344	0.7643	2044
0.0826	792	0.4187	2491	0.8664	1384
0.1190	1101	0.5195	2584	0.8963	1090
0.1526	1368	0.6186	2493	0.9284	803
0.2128	1770	0.6837	2365	0.9629	433
0.2649	2035				
Dimethyl Carbonate (1) + 2-Butanol (2)					
T = 288.15 K					
0.0435	454	0.3533	2372	0.7662	1972
0.0834	828	0.4213	2485	0.8677	1331
0.1202	1139	0.5221	2560	0.8973	1079
0.1540	1394	0.6211	2435	0.9291	800
0.2145	1783	0.6860	2277	0.9633	441
0.2670	2048				
T = 298.15 K					
0.0434	538	0.3528	2549	0.8135	1790
0.0833	964	0.4209	2687	0.8674	1394
0.1199	1308	0.5216	2743	0.8971	1127
0.1537	1576	0.6206	2599	0.9290	828
0.2142	2002	0.6856	2437	0.9632	455
0.2665	2250	0.7658	2088		
T = 313.15 K					
0.0433	560	0.3522	2702	0.7653	2195
0.0831	1005	0.4201	2850	0.8671	1424
0.1196	1359	0.5209	2909	0.8969	1153
0.1534	1644	0.6200	2754	0.9288	825
0.2137	2074	0.6850	2562	0.9631	454
0.2660	2368				
Dimethyl Carbonate (1) + 2-Methyl-1-propanol (2)					
T = 288.15 K					
0.0438	386	0.3547	2140	0.7674	1882
0.0839	722	0.4229	2268	0.8684	1287
0.1208	1018	0.5237	2331	0.8979	1037
0.1549	1292	0.6226	2276	0.9296	748
0.2156	1643	0.6874	2149	0.9635	404
0.2682	1869				
T = 298.15 K					
0.0437	401	0.3541	2366	0.7669	2028
0.0837	778	0.4223	2495	0.8681	1357
0.1206	1075	0.5231	2560	0.8977	1093
0.1545	1359	0.6219	2459	0.9294	810
0.2152	1777	0.6868	2331	0.9634	427
0.2677	2049				
T = 313.15 K					
0.0435	506	0.3532	2591	0.7662	2149
0.0834	926	0.4212	2739	0.8676	1395
0.1201	1278	0.5220	2805	0.8973	1153
0.1540	1550	0.6211	2697	0.9291	816
0.2145	1993	0.6860	2505	0.9633	437
0.2669	2257				
Dimethyl Carbonate (1) = 2-Methyl-2-propanol (2)					
T = 313.15 K					
0.0447	571	0.3595	2658	0.7710	1997
0.0550	691	0.4279	2750	0.8707	1326
0.1230	1368	0.5288	2780	0.9000	1058
0.1576	1670	0.6275	2616	0.9309	775
0.2191	2083	0.6918	2394	0.9642	422
0.2723	2352				

Table 3. Mole Fractions, x_1, Densities, ρ, and Excess Molar Volumes, V_m^E, of Dimethyl Carbonate + Butanol Isomers at (288.15, 298.15, and 313.15)K

x_1	ρ/g·cm^{-3}	V_m^E/ cm^3·mol^{-1}	x_1	ρ/g·cm^{-3}	V_m^E/ cm^3·mol^{-1}
Dimethyl Carbonate (1) + 1-Butanol					
T = 288.15 K					
0.0250	0.81893	0.049	0.6216	0.96687	0.409
0.0917	0.83414	0.162	0.6499	0.97456	0.397
0.1276	0.84245	0.213	0.7182	0.99342	0.357
0.2131	0.86268	0.311	0.7623	1.00580	0.324
0.3083	0.88590	0.381	0.8593	1.03372	0.226
0.3448	0.89494	0.401	0.9019	1.04631	0.170
0.4443	0.92013	0.429	0.9574	1.06304	0.083
0.5167	0.93895	0.431			
T = 298.15 K					
0.0250	0.81122	0.055	0.6216	0.95579	0.448
0.0917	0.82605	0.180	0.6499	0.96334	0.434
0.1276	0.83416	0.235	0.7182	0.98182	0.389
0.2131	0.85389	0.343	0.76233	0.99397	0.351
0.3083	0.87657	0.420	0.8593	1.02138	0.240
0.34481	0.88539	0.444	0.9019	1.03376	0.177
0.4443	0.91003	0.474	0.9574	1.05022	0.079
0.5167	0.92844	0.475			
T = 313.15 K					
0.0250	0.79938	0.027	0.6216	0.93879	0.533
0.0917	0.81357	0.214	0.6499	0.94609	0.518
0.1276	0.82137	0.278	0.7182	0.96404	0.464
0.2131	0.84030	0.411	0.7623	0.97584	0.420
0.3083	0.86221	0.497	0.8593	1.00250	0.290
0.3448	0.87070	0.526	0.9019	1.01456	0.217
0.4443	0.89446	0.566	0.9574	1.0306	0.103
0.5167	0.91229	0.567			
Dimethyl Carbonate (1) + 2-Butanol (2)					
T = 288.15 K					
0.0452	0.82036	0.145	0.5502	0.94385	0.655
0.0956	0.83161	0.278	0.6251	0.96419	0.621
0.1999	0.8556	0.465	0.7529	1.00029	0.502
0.2779	0.87429	0.565	0.8172	1.01918	0.410
0.3964	0.90376	0.646	0.9162	1.04942	0.217
0.4889	0.92758	0.666	0.9648	1.06484	0.097
T = 298.15 K					
0.0452	0.81197	0.158	0.5502	0.93270	0.723
0.0960	0.82292	0.305	0.6251	0.95270	0.684
0.1999	0.84629	0.518	0.7529	0.98820	0.552
0.2779	0.86452	0.628	0.8172	1.00688	0.446
0.3962	0.89336	0.716	0.9162	1.03668	0.237
0.4889	0.91674	0.736	0.9648	1.05189	0.108
T = 313.15 K					
0.0452	0.79872	0.178	0.5502	0.91556	0.821
0.0960	0.80923	0.344	0.6251	0.93506	0.774
0.1999	0.83171	0.592	0.7529	0.96980	0.617
0.2779	0.84934	0.715	0.8172	0.98803	0.501
0.3964	0.8773	0.817	0.9162	1.01728	0.261
0.4889	0.90023	0.835	0.9648	1.03218	0.119
Dimethyl Carbonate (1) + 2-Methyl-1-propanol (2)					
T = 288.15 K					
0.0452	0.81581	0.091	0.6391	0.96799	0.405
0.1413	0.83845	0.239	0.7107	0.98837	0.361
0.2371	0.86189	0.337	0.7866	1.01042	0.305
0.3306	0.88542	0.399	0.8955	1.04324	0.178
0.4439	0.91487	0.433	0.9312	1.05432	0.125
0.5148	0.93382	0.433	0.9758	1.06840	0.050
T = 298.15 K					
0.0452	0.80790	0.104	0.6391	0.95672	0.458
0.1413	0.82299	0.274	0.7107	0.97674	0.407
0.2371	0.85282	0.384	0.7866	0.9984	0.342
0.3306	0.87582	0.454	0.8955	1.03072	0.197
0.4439	0.90465	0.492	0.9312	1.04163	0.137
0.5148	0.92319	0.494	0.9758	1.05551	0.052
T = 313.15 K					
0.0452	0.79563	0.127	0.6391	0.93933	0.552
0.1413	0.81675	0.336	0.7107	0.95883	0.488
0.2371	0.83876	0.468	0.7866	0.97993	0.409
0.3306	0.86095	0.551	0.8955	1.01151	0.232
0.4439	0.88884	0.594	0.9312	1.02218	0.161
0.5148	0.90678	0.598	0.9758	1.03576	0.061
Dimethyl Carbonate (1) + 2-Methyl-2-propanol (2)					
T = 313.15 K					
0.0214	0.76942	0.100	0.5644	0.90624	0.783
0.0870	0.78394	0.335	0.6417	0.92856	0.724
0.1859	0.80699	0.582	0.7157	0.95060	0.641
0.2788	0.82984	0.722	0.8456	0.99124	0.415
0.3665	0.85231	0.792	0.9076	1.01158	0.270
0.4657	0.87881	0.813	0.9381	1.02185	0.188

Table 4. Least-Squares Parameters, a_k, and Standard Deviations, $\sigma(Q_m^E)$, Eq 2, of Dimethyl Carbonate + Butanol Isomers

function	a_0	a_1	a_2	a_3	a_4	$\sigma(Q_m^E)$[a]
Dimethyl Carbonate (1) + 1-Butanol (2)						
T = 288.15 K						
H_m^E/J·mol^{-1}	8506.4	655.55	2883.4	1635.0	−1908.9	9.0
V_m^E/cm^3·mol^{-1}	1.7289	−0.0327	0.3123			0.0015
T = 298.15 K						
H_m^E/J·mol^{-1}	9425.6	802.47	886.16	2104.8		8.6
V_m^E/cm^3·mol^{-1}	1.9083	−0.0856	0.2528			0.0015
T = 313.15 K						
H_m^E/J·mol^{-1}	10282	713.27	2963.2	303.08	−2327.9	9.1
V_m^E/cm^3·mol^{-1}	2.2685	−0.0767	0.3538			0.0031
Dimethyl Carbonate (1) + 2-Butanol (2)						
T = 288.15 K						
H_m^E/J·mol^{-1}	10235	148.01	1733.3	782.56		8.6
V_m^E/cm^3·mol^{-1}	2.6522	−0.0966	0.5130	−0.2008		0.0026
T = 298.15 K						
H_m^E/J·mol^{-1}	10973	−105.10	2306.2			8.9
V_m^E/cm^3·mol^{-1}	2.9341	−0.1696	0.5287	−0.1489		0.0018
T = 313.15 K						
H_m^E/J·mol^{-1}	11658	7.0614	1776.6	−549.10		9.6
V_m^E/cm^3·mol^{-1}	3.3355	−0.2574	0.5297	−0.1307		0.0011
Dimethyl Carbonate (1) + 2-Methyl-1-butanol (1)						
T = 288.15 K						
H_m^E/J·mol^{-1}	9291.7	642.68	3672.4	711.97	−2866.5	8.9
V_m^E/cm^3·mol^{-1}	1.7376	−0.0663	0.3522			0.0017
T = 298.15 K						
H_m^E/J·mol^{-1}	10220	382.45	3183.2	1265.3	−2870.9	9.3
V_m^E/cm^3·mol^{-1}	1.9768	−0.1120	0.3628			0.0019
T = 313.15 K						
H_m^E/J·mol^{-1}	11222	332.57	2251.5	−266.50	−1524.7	9.0
V_m^E/cm^3·mol^{-1}	2.3910	−0.1816	0.4143			0.0030
Dimethyl Carbonate (1) + 2-Methyl-2-propanol (2)						
T = 313.15 K						
H_m^E/J·mol^{-1}	11180	−665.86	1896.8			9.0
V_m^E/cm^3·mol^{-1}	3.2307	−0.4530	0.7158	−0.2330		0.0020

[a] In J·mol^{-1}, or cm^3·mol^{-1}.

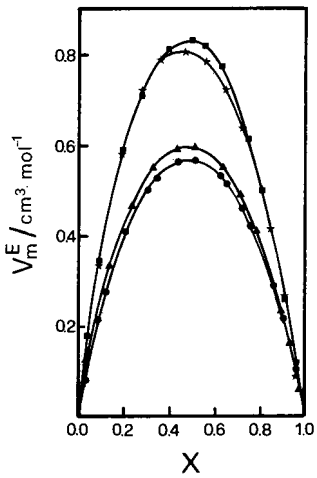

Figure 2. Excess molar volumes, V_m^E, of dimethyl carbonate (1) + 1-butanol (2) (●), + 2-butanol (2) (■), + 2-methyl-1-propanol (2) (▲), and + 2-methyl-2-propanol (2) (★) at 313.15 K. Solid curves, calculated by eq 2.

points weighted equally was fitted to the data by the least-squares method. The adjustable parameters, a_k, and the standard deviations, $\sigma(Q_m^E)$, are listed in Table 4.

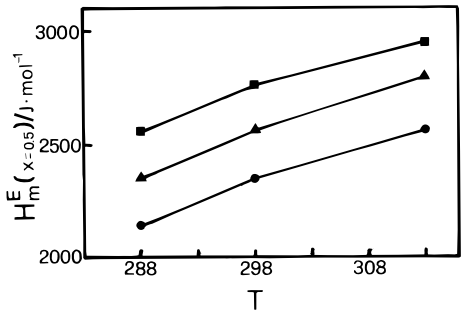

Figure 3. Values of equimolar H^E_m ($x = 0.5$) as a function of the test-temperature of dimethyl carbonate + butanol isomers. (●), (■), (▲) refer to dimethyl carbonate + 1-butanol, + 2-butanol, or + 2-methyl-1-propanol, respectively.

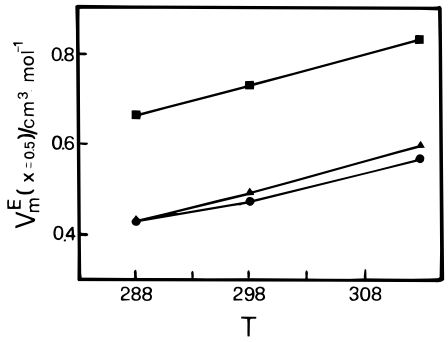

Figure 4. Values of equimolar V^E_m ($x = 0.5$) as a function of the test temperature of dimethyl carbonate + butanol isomers. (●), (■), (▲) refer to dimethyl carbonate + 1-butanol, + 2-butanol, or + 2-methyl-1-propanol, respectively.

Figures 1 and 2 show H^E_m and V^E_m data for the four mixtures at 313.15 K. We have not reported the experimental data at (288.15 or 298.15) K to avoid overlapping of the curves. Plots of equimolar $H^E_m(x_1 = 0.5)$ or $V^E_m(x_1 = 0.5)$ as a function of temperature are represented in Figures 3 and 4.

The large positive values of H^E_m are consistent with the well-known associations of pure alkanols, owing to strong hydrogen bonding between molecules.

Figure 1 shows an increase of H^E_m in the order 1-butanol → 2-methylpropanol → 2-butanol, that is, with the increase of branching in the carbon atom carrying the −OH group.

Also, the H^E_m's of the binary mixtures dimethyl carbonate + methanol or + ethanol, or + 1-propanol, previously obtained (Comelli and Francesconi, 1997) show the same increase with the increased molecular size and match with the H^E_m values of this paper.

The complex molecular interactions in these systems make impossible even a qualitative interpretation of the results. In fact, in addition to the H-bond association of the alkanols, the strong dipole−dipole interactions between carbonate molecules and the H-bond association of alkanols with the carbonate affect considerably the properties of the mixtures with endothermic as well as exothermic contributions. Moreover, branching of aliphatic groups may alter their electron donor character and increase their steric hindrance even if these effects are less important than the above-mentioned interactions.

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