Excess Molar Volumes and Viscosities of Diethylene Glycol Diethyl Ether with Dimethyl Carbonate, Diethyl Carbonate, and Propylene Carbonate at (298.15, 308.15, and 318.15) K

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Excess molar volumes, $V_{\rm m}^{\rm E}$, and viscosities, η , have been measured for the binary mixtures diethylene glycol diethyl ether + dimethyl carbonate, + diethyl carbonate, and + propylene carbonate at temperatures of (298.15, 308.15, and 318.15) K and atmospheric pressure over the whole range of mixture compositions. The excess molar volumes are fitted to a Redlich–Kister type polynomial equation to derive the binary coefficients and estimate the standard errors between the experimental and calculated quantities. The experimental results are compared with those of previous studies for binary mixtures containing esters of carbonic acid.

Introduction

Esters of carbonic acid are important fluids that find use as solvents for a variety of extractions of industrial importance, for resins, in the synthesis of pharmaceuticals, and for addressing the problem of agricultural chemicals. Thus, a study of physical property data on binary mixtures containing esters of carbonic acid has attracted considerable interest in the literature (Francesconi and Comelli, 1999; Muhuri and Hazra, 1995; Barthel et al., 1995; Francesconi and Comelli, 1997).

As a part of our systematic program of research on the physicochemical properties of binary liquid mixtures (Pal et al., 1999a,b; Pal and Kumar, 1998; Pal et al., 1998), we present here the experimental results of excess molar volume, $V_{\rm m}^E$, and viscosity, η , at (298.15, 308.15, and 318.15) K and atmospheric pressure for the binary mixtures of diethylene glycol diethyl ether with dimethyl carbonate, diethyl carbonate, and propylene carbonate over the whole mole fraction range. The aim of this work is to provide data for the characterization of the molecular interactions of these mixtures and to examine the effects of replacing methyl by ethyl groups in polyethers.

To our knowledge there are no literature data on these systems.

Experimental Section

Chemicals. Dimethyl carbonate (Spectrochem, Bombay, >98 mass %), diethyl carbonate (Siscochem, Bombay, >99 mass %), and propylene carbonate (Merck-Schuchardt, >99 mass%) were the same as those used in earlier studies (Pal and Kumar, 1998). Diethylene glycol diethyl ether (Merck-Schuchardt, >98 mass %) was dried over FeSO₄ (A. R., BDH) and then fractionally distilled under reduced nitrogen pressure. Prior to measurements, all liquids were stored in contact with molecular sieves type 4A to reduce water content and were partially degassed at low pressure. The results of the measurements of their densities and

Table 1.	Comparison	of Exper	imenta	d Densitie	s, ρ, an	d
Viscositi	les, η , of Pure	Liquids	with L	iterature	Values	at
298.15 K						

	ρ/ ε	g cm ⁻³	η/n	nPa s
liquid	exptl	lit.	exptl	lit.
diethylene glycol diethyl ether	0.9035	0.903 3 ^a	1.268	
dimethyl carbonate	1.0632	1.063 31 ^b 1.063 059 ^c	0.589	
diethyl carbonate	0.9690	0.969 25 ^b 0.969 26 ^d	0.749	
propylene carbonate	1.1988	1.198 83 ^e	2.493	2.4711^{e} 2.51^{f}

 a Roux et al. (1978). b Francesconi et al. (1997). c Negadi et al. (1993). d Riddick et al. (1986). e Muhuri and Hazra (1995). f Barthel et al. (1995).

viscosities at 298.15 K and atmospheric pressure are given in Table 1, together with some values taken from the literature.

Apparatus and Procedure. Excess molar volumes, reproducible to ± 0.003 cm³ mol⁻¹, were measured directly with a continuous dilution dilatometer, as described earlier by Dickinson et al. (1975). Details of its calibration, the experimental setup, and the measuring procedure have been described previously (Pal and Singh, 1994). The mole fraction of each mixture was obtained to an accuracy of 1 \times 10⁻⁴ from the measured apparent masses of the components. All apparent masses were corrected for buoyancy. All molar quantities was based on the relative atomic mass table of 1986 issued by IUPAC (1986). Each run covered just over half of the mole fraction range, giving an overlap between two runs.

The kinematic viscosities of the pure components and the mixtures were measured at (298.15, 308.15, and 318.15) K and atmospheric pressure using an Ubbelohde suspended level viscometer. The viscometer was calibrated with thricedistilled water and twice-distilled benzene. Care was taken to prevent evaporation during measurements. The other experimental details have been given previously (Pal and Singh, 1996). After multiplication by density, the dynamic

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Table 2. Excess Molar Volumes, $V_{\mathrm{m}}^{\!\scriptscriptstyle \mathrm{E}}$, for Binary Mixtures at Various Temperature	res
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	$V_{\rm m}^{\rm E}$		$V_{\rm m}^{\rm E}$		$V_{\rm m}^{\rm E}$						
<i>X</i> 1	${\rm cm^3\ mol^{-1}}$	<i>X</i> 1	$\rm cm^3 \ mol^{-1}$	<i>X</i> 1	$\rm cm^3~mol^{-1}$						
Dieth	ylene Glycol	Diethyl E	ther (1) +	Dieth	ylene Glycol	Diethyl E	(1) +	Dieth	ylene Glycol	Diethyl E	ther (1) +
	Dimethyl C	arbonate	(2)		Diethyl Ca	arbonate (2)		Propylene C	Carbonate	(2)
	298	.15 K			298	.15 K			298	.15 K	
0.0158	0.010	0.3712	-0.030	0.0110	-0.005	0.4502	-0.118	0.0156	-0.049	0.3217	-0.800
0.0276	0.022	0.4512	-0.032	0.0325	-0.014	0.4942	-0.120	0.0489	-0.153	0.3379	-0.817
0.0511	0.028	0.5076	-0.034	0.0643	-0.026	0.5310	-0.120	0.0911	-0.272	0.3890	-0.869
0.0617	0.032	0.5751	-0.032	0.0850	-0.034	0.5727	-0.121	0.1412	-0.414	0.4221	-0.910
0.0867	0.030	0.6150	-0.030	0.1197	-0.046	0.6239	-0.117	0.1915	-0.551	0.4307	-0.910
0.1178	0.025	0.7042	-0.024	0.1444	-0.054	0.6645	-0.115	0.1989	-0.570	0.4921	-0.950
0.1543	0.020	0.7609	-0.021	0.1904	-0.068	0.7217	-0.106	0.2177	-0.607	0.5817	-0.941
0.2001	0.011	0.8303	-0.019	0.2342	-0.079	0.7717	-0.097	0.2444	-0.665	0.6800	-0.859
0.2351	-0.009	0.8778	-0.014	0.2863	-0.088	0.8378	-0.071	0.2560	-0.678	0.7526	-0.740
0.2643	-0.015	0.9248	-0.010	0.3420	-0.102	0.8952	-0.050	0.2723	-0.718	0.8648	-0.482
0.2980	-0.019	0.9551	-0.007	0.3601	-0.105	0.9465	-0.026	0.3044	-0.760	0.9806	-0.069
0.3019	-0.020			0.4006	-0.110	0.9752	-0.014				
	308	.15 K			308	.15 K			308	.15 K	
0.0016	0.001	0.3540	-0.042	0.0268	-0.019	0.4505	-0.145	0.0067	-0.027	0.3135	-0.846
0.0243	0.006	0.3950	-0.047	0.0553	-0.036	0.4604	-0.143	0.0186	-0.073	0.3435	-0.889
0.0545	0.010	0.4395	-0.047	0.0964	-0.053	0.4894	-0.143	0.0298	-0.115	0.3697	-0.920
0.0763	0.008	0.4484	-0.050	0.1434	-0.077	0.5204	-0.142	0.0458	-0.178	0.4014	-0.951
0.1031	0.004	0.4980	-0.051	0.1822	-0.087	0.5910	-0.140	0.0609	-0.226	0.4106	-0.960
0.1616	-0.010	0.5727	-0.048	0.2111	-0.095	0.6453	-0.138	0.0843	-0.304	0.4280	-0.973
0.2256	-0.026	0.6396	-0.043	0.2590	-0.107	0.6858	-0.134	0.1136	-0.394	0.4733	-1.000
0.2328	-0.025	0.7139	-0.039	0.2999	-0.119	0.7474	-0.118	0.1577	-0.522	0.5377	-1.007
0.2528	-0.030	0.7848	-0.035	0.3364	-0.130	0.7996	-0.105	0.1989	-0.623	0.6078	-0.983
0.2827	-0.034	0.8498	-0.023	0.3747	-0.139	0.8430	-0.092	0.2127	-0.656	0.6773	-0.917
0.3175	-0.038	0.9213	-0.012	0.3873	-0.143	0.8813	-0.071	0.2206	-0.667	0.7560	-0.784
0.3417	-0.041	0.9620	-0.008	0.4120	-0.144	0.9448	-0.037	0.2429	-0.710	0.8212	-0.650
				0.4202	-0.145	0.9906	-0.010	0.2681	-0.760	0.8952	-0.423
								0.2720	-0.765	0.9388	-0.260
								0.3113	-0.835		
	318	.15 K			318	.15 K			318	.15 K	
0.0177	-0.007	0.3435	-0.062	0.0216	-0.015	0.4062	-0.170	0.0050	-0.040	0.3856	-1.000
0.0407	-0.014	0.4120	-0.065	0.0457	-0.030	0.4433	-0.170	0.0169	-0.079	0.4331	-1.040
0.0573	-0.021	0.5002	-0.063	0.0891	-0.060	0.4883	-0.172	0.0350	-0.160	0.4820	-1.059
0.0784	-0.024	0.5409	-0.061	0.1140	-0.067	0.5265	-0.170	0.0539	-0.249	0.5333	-1.060
0.1057	-0.029	0.6058	-0.060	0.1418	-0.081	0.5992	-0.164	0.0717	-0.308	0.5745	-1.050
0.1329	-0.031	0.6879	-0.057	0.1867	-0.110	0.6632	-0.159	0.1049	-0.427	0.6223	-1.029
0.1664	-0.034	0.7190	-0.056	0.2154	-0.120	0.7285	-0.142	0.1567	-0.584	0.7110	-0.946
0.1974	-0.038	0.7607	-0.053	0.2477	-0.132	0.7616	-0.136	0.2069	-0.697	0.7868	-0.780
0.2398	-0.050	0.8051	-0.051	0.2950	-0.150	0.7826	-0.130	0.2540	-0.800	0.8421	-0.649
0.2677	-0.054	0.8508	-0.044	0.3083	-0.154	0.8206	-0.110	0.2743	-0.841	0.8977	-0.468
0.3049	-0.060	0.9234	-0.034	0.3386	-0.168	0.8644	-0.096	0.3003	-0.893	0.9240	-0.364
				0.3483	-0.168	0.9169	-0.060	0.3414	-0.950		
				0.3781	-0.170	0.9498	-0.040				

viscosity, η , was deduced with a relative error of ± 0.003 mPa s. All the measurements were carried out in a thermostatically controlled, well-stirred water bath with temperature controlled to ± 0.01 K.

Results and Discussion

The experimental results of the excess molar volumes and viscosities of the different binary mixtures for a number of mole fractions at atmospheric pressure and at (298.15, 308.15, and 318.15) K are reported in Tables 2 and 3.

From the measured values of excess molar volumes, the densities of liquid mixtures were calculated using the following equation:

$$\rho = (x_1 M_1 + x_2 M_2) / (V_m^E + x_1 V_1^\circ + x_2 V_2^\circ)$$
(1)

where x_1 and x_2 are the mole fractions, M_1 and M_2 are the molar masses, and V_i° stands for the molar volumes of the pure components, respectively.

The viscosity deviations can be calculated from the following relationship (Aucejo et al., 1996; Ramadevi et al., 1996)

$$\Delta \ln \eta = \ln \eta - [x_1 \ln \eta_1 + x_2 \ln \eta_2]$$
 (2)

where η is the dynamic viscosity of the mixtures and η_1 and η_2 are the viscosities of components 1 and 2.

The values of V_m^E and $\Delta \ln \eta$ at various temperatures for all mixtures were fitted to the Redlich–Kister type equation (Redlich–Kister, 1948)

$$Y = x_1 x_2 \sum_{i=1}^{k} A_i (x_1 - x_2)^{i-1}$$
(3)

where A_i are the polynomial coefficients, which were obtained by fitting the equation to the experimental result with a least-squares regression method with all points weighted equally. The correlated results are given in Table

rubic o. Densities, p, and viscosities, p, for Dinary mixtures at various remperature	Table 3.	Densities , <i>ρ</i> ,	, and Viscosities,	η , for B	Binary M	fixtures at	Various 🛛	Temperature
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<i>X</i> 1	$ ho/{ m g~cm^{-3}}$	η/mPa s	<i>X</i> 1	$ ho/{ m g~cm^{-3}}$	η/mPa s	<i>X</i> ₁	$ ho/{ m g~cm^{-3}}$	η/mPa s	<i>X</i> 1	$ ho/{ m g~cm^{-3}}$	η/mPa s
			Diethy	lene Glycol	Diethyl Ethe	er (1) + Din	nethyl Carb	onate (2)			
					298.	15 K					
0.0000	1.0632	0.589	0.1383	1.0228	0.718	0.4435	0.9633	0.959	0.8183	0.9188	1.189
0.0109	1.0598	0.604	0.1877	1.0109	0.760	0.5274	0.9513	1.021	0.9068	0.9110	1.229
0.0541	1.0460	0.641	0.2422	0.9990	0.808	0.6085	0.9409	1.072	0.9643	0.9063	1.258
0.0935	1.0346	0.677	0.3409	0.9801	0.886	0.7154	0.9289	1.136	1.0000	0.9035	1.268
					308.	15 K					
0.0000	1.0508	0.520	0.1383	1.0118	0.624	0.4435	0.9541	0.811	0.8183	0.9110	0.986
0.0109	1.0473	0.532	0.1877	1.0003	0.654	0.5274	0.9425	0.855	0.9068	0.9034	1.019
0.0541	1.0341	0.563	0.2422	0.9887	0.689	0.6085	0.9325	0.894	0.9643	0.8988	1.037
0.0935	1.0232	0.593	0.3409	0.9704	0.752	0.7154	0.9209	0.942	1.0000	0.8961	1.048
					318	15 K					
0.0000	1.0377	0.472	0.1383	1.0000	0.546	0.4435	0.9440	0.697	0.8183	0.9020	0.831
0.0000	1 0343	0.480	0 1877	0.9888	0.571	0 5274	0.9327	0.727	0.9068	0.8945	0.857
0.0541	1 0216	0.502	0 2422	0.9776	0.604	0.6085	0.9230	0.761	0.9643	0.8901	0.872
0.0011	1 0110	0.502	0.2422	0.9598	0.651	0.0000	0.9116	0.798	1 0000	0.8874	0.881
0.0000	1.0110	0.020	0.0100	0.0000	0.001	0.7101	0.0110	0.700	1.0000	0.0074	0.001
			Dieth	ylene Glyco	ol Diethyl Eth	her $(1) + Die$	ethyl Carbo	nate (2)			
					298.	15 K					
0.0000	0.9690	0.749	0.1858	0.9530	0.863	0.4971	0.9309	1.041	0.8683	0.9099	1.214
0.0221	0.9670	0.763	0.2214	0.9502	0.890	0.5997	0.9246	1.092	0.9296	0.9069	1.239
0.0409	0.9653	0.776	0.2775	0.9459	0.923	0.6836	0.9198	1.133	0.9694	0.9050	1.255
0.0945	0.9606	0.812	0.3669	0.9395	0.972	0.7747	0.9148	1.170	1.0000	0.9035	1.268
					308.	15 K					
0.0000	0.9577	0.658	0.1858	0.9429	0.741	0.4971	0.9221	0.879	0.8683	0.9023	1.009
0.0221	0.9558	0.667	0.2214	0.9403	0.758	0.5997	0.9162	0.917	0.9296	0.8994	1.027
0.0409	0.9543	0.675	0.2775	0.9363	0.784	0.6836	0.9116	0.947	0.9694	0.8975	1.040
0.0945	0.9499	0.699	0.3669	0.9302	0.822	0.7747	0.9069	0.979	1.0000	0.8961	1.048
					318.	15 K					
0.0000	0.9438	0.578	0.1858	0.9305	0.643	0.4971	0.9115	0.747	0.8683	0.8932	0.849
0.0221	0.9421	0.584	0.2214	0.9281	0.656	0.5997	0.9060	0.775	0.9296	0.8905	0.862
0.0409	0.9407	0.592	0.2775	0.9245	0.677	0.6836	0.9018	0.801	0.9694	0.8887	0.871
0.0945	0.9368	0.609	0.3669	0.9190	0.707	0.7747	0.8975	0.825	1.0000	0.8874	0.881
0.0010	010000	01000				(1) + D			110000	010011	01001
			Diethy	lene Glycol	Diethyl Ethe	r(1) + Prop	bylene Carb	onate (2)			
					298.	15 K					
0.0000	1.1988	2.493	0.2136	1.0976	2.225	0.4970	1.0065	1.833	0.8704	0.9256	1.377
0.0130	1.1914	2.474	0.2570	1.0811	2.179	0.5874	0.9838	1.717	0.9264	0.9158	1.316
0.0542	1.1693	2.418	0.3304	1.0556	2.080	0.6907	0.9606	1.589	0.9628	0.9096	1.289
0.1089	1.1424	2.353	0.4267	1.0259	2.940	0.7876	0.9410	1.474	1.0000	0.9035	1.268
0.1594	1.1198	2.288									
					308.	15 K					
0.0000	1.1897	2.041	0.2136	1.0894	1.827	0.4970	0.9988	1.504	0.8704	0.9181	1.148
0.0130	1.1824	2.024	0.2570	1.0730	1.782	0.5874	0.9763	1.404	0.9264	0.9083	1.101
0.0542	1.1605	1.982	0.3304	1.0477	1.702	0.6907	0.9531	1.306	0.9628	0.9022	1.071
0.1089	1.1339	1.923	0.4267	1.0182	1.589	0.7876	0.9335	1.219	1.0000	0.8961	1.048
0.1594	1.1115	1.875									
					318.	15 K					
0.0000	1.1780	1.718	0.2136	1.0793	1.506	0.4970	0.9895	1.227	0.8704	0.9095	0.957
0.0130	1.1709	1.703	0.2570	1.0631	1.466	0.5874	0.9671	1.152	0.9264	0.8997	0.922
0.0542	1.1495	1.658	0.3304	1.0379	1.390	0.6907	0.9442	1.076	0.9628	0.8935	0.901
0.1089	1.1233	1.605	0.4267	1.0086	1.297	0.7876	0.9248	1.010	1.0000	0.8874	0.881
0.1594	1.1012	1.555									

4, in which the tabulated standard deviation was defined as

$$\sigma = \left[\sum (Y_{\rm exp} - Y_{\rm cal})^2 / (n - k)\right]^{1/2}$$
(4)

where *n* is the number of measurements and *k* is the number of estimated parameters. *Y* refers to $V_{\rm m}^{\rm E}/{\rm cm^3}$ mol⁻¹ or $\Delta \ln[\eta/{\rm mPa~s}]$.

Figures 1 and 2 show V_m^E and $\Delta \ln \eta$ data for the three mixtures at 298.15 K. We have not reported the experimental data at (308.15 or 318.15) K, as the curves are similar. The excess molar volumes are negative over the entire range of composition for the systems diethylene glycol diethyl ether + diethyl carbonate and + propylene carbonate. For the system diethylene glycol diethyl ether

+ dimethyl carbonate, a change of sign occurs at 298.15 and 308.15 K. The binary mixtures with propylene carbonate show strongly negative V_m^E values that increase with an increase in temperature while, for the mixtures with dimethyl carbonate or diethyl carbonate, V_m^E shows a negligible temperature dependence. This behavior may be compared with the V_m^E results of the mixtures of diethylene glycol dimethyl ether with esters of carbonic acid (Pal and Kumar, 1998): a marked increase in V_m^E at all three temperatures is evident here. With the replacement of a methyl group by an ethyl group in the polyether chain, V_m^E becomes more negative with a rise in temperature. The negative values of V_m^E suggest a specific intermolecular interaction occurring in the mixing process. This is very important with propylene carbonate. The temperature



Figure 1. Comparison of the excess molar volumes, V_{m}^{E} , of different mixtures at 298.15 K: \bigcirc , dimethyl carbonate; \triangle , diethyl carbonate; \Box , propylene carbonate. Solid curves were calculated from eq 3 using coefficient A_i of Table 4.

Table 4. Smoothing Coefficients, A_{i} , and Standard Deviations, $\sigma(Y(x))$, of Eq 3 for the Binary Mixtures at Various Temperatures

Y(x)	A_1	A_2	A_3	A_4	A_5	$\sigma(Y(x))$					
Diethylene Glycol Diethyl Ether (1) + Dimethyl Carbonate (2)											
298.15 K											
$V_{\rm m}^{\rm E}/{ m cm^3mol^{-1}}$	-0.134	0.065	0.094	-0.566	0.425	0.003					
$\Delta \ln[\eta/mPa s]$	0.589	-0.162	0.084	-0.049		0.003					
		308	.15 K								
$V_{ m m}^{ m E}/ m cm^3mol^{-1}$	-0.195	0.019	0.007	-0.254	0.299	0.002					
$\Delta \ln[\eta/\text{mPa s}]$	0.514	-0.131	0.106	-0.114		0.003					
		318	.15 K								
$V_{ m m}^{ m E}/ m cm^3mol^{-1}$	-0.260	0.034	0.012	-0.192	-0.283	0.003					
$\Delta \ln[\eta/\text{mPa s}]$	0.435	-0.135	0.012	0.054		0.003					
Diethylene	Glycol Di	iethyl Et	her (1) +	Diethyl	Carbona	te (2)					
		298	.15 K								
$V_{\rm m}^{\rm E}/{ m cm^3mol^{-1}}$	-0.480	-0.108	-0.012	0.073		0.001					
$\Delta \ln[\eta/\text{mPa s}]$	0.271	-0.078	0.005	0.003		0.002					
		308	.15 K								
$V_{ m m}^{ m E}/ m cm^3mol^{-1}$	-0.582	-0.036	-0.113	-0.038		0.003					
$\Delta \ln[\eta/\text{mPa s}]$	0.227	-0.021	-0.040	0.040		0.001					
_		318	.15 K								
$V_{\rm m}^{\rm E}/{ m cm^3mol^{-1}}$	-0.693	0.050	-0.192	-0.215	0.166	0.003					
$\Delta \ln[\eta/\text{mPa s}]$	0.193	-0.052	-0.055	0.069		0.002					
Diethylene G	lycol Die	thyl Eth	er (1) + I	Propylen	e Carbon	ate (2)					
		298	.15 K								
$V_{ m m}^{ m E}/ m cm^3mol^{-1}$	-3.780	-0.422	-0.199	-0.005	0.487	0.004					
$\Delta \ln[\eta/mPa s]$	0.121	-0.139	0.133	-0.018	-0.451	0.002					
308.15 K											
$V_{ m m}^{ m E}/ m cm^3mol^{-1}$	-4.029	-0.393	-0.152	0.062	-0.229	0.003					
$\Delta \ln[\eta/\text{mPa s}]$	0.116	-0.170	-0.028	0.150		0.003					
_		318	.15 K								
$V_{\rm m}^{\rm E}/{ m cm^3mol^{-1}}$	-4.259	-0.360	-0.580	0.027	-0.473	0.004					
$\Delta \ln[\eta/\text{mPa s}]$	-0.004	-0.126	0.030	0.114		0.003					

coefficient $(\partial V_{m}^{E}/\partial T)_{P}$ is negative for all the mixtures over the whole mole fraction range.

Figure 2 shows that the deviation in viscosity $\Delta \ln \eta$ is positive over the entire composition range for all systems, showing an inversion of sign for mixtures of diethylene glycol diethyl ether with propylene carbonate at all temperatures. The viscosities of all three binary mixtures decrease with the increase of temperature. The η results of the mixtures at all three temperatures follow the sequence dimethyl carbonate < diethyl carbonate < propylene carbonate. At any particular temperature, as x_1



Figure 2. Comparison of viscosity deviations, $\Delta \ln \eta$, of different mixtures at 298.15 K: \bigcirc , dimethyl carbonate; \triangle , diethyl carbonate; \square , propylene carbonate. Solid curves were calculated from eq 3 using coefficient A_i of Table 4.

increases, the η values of dimethyl carbonate and diethyl carbonate increase whereas that for propylene carbonate decreases. A further comparison of data at different temperatures shows that the temperature coefficient $(\partial \eta / \partial T)_P$ is decreasing for the three mixtures with increasing temperature.

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