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High-Pressure Volumetric Behavior of *x* 1,1,1,2-Tetrafluoroethane + (1 − *x*) 2,5,8,11,14-Pentaoxapentadecane (TEGDME) Mixtures

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This paper reports 1017 new *pVT* measurements of the *x* 1,1,1,2-tetrafluoroethane (HFC-134a) + (1 − *x*) 2,5,8,11,14-pentaoxapentadecane (TEGDME) system for x = 0.0, 0.1114, 0.2896, 0.3648, 0.5702, 0.6931, 0.7288, 0.8727, 0.9290, and 1 between 10 and 60 MPa in the temperature range 293.15 K to 373.15 K at 5 MPa and 10 K intervals, respectively. In almost all the measurement range, the density of the pure compressed refrigerant is greater than that of the pure polyether. For temperatures higher than 343.15 K, the isocomposition curves for the mixtures show an intersection point. Similar behavior has been previously found for HFC-134a + triethylene glycol dimethyl ether, carbon dioxide + alkane or toluene systems, and mixtures of R-410A with polyolester lubricants. The excess molar volume is negative over the whole composition range at all temperatures and pressures.

Introduction

The study (measurements and modeling) of the thermophysical properties of refrigerant + lubricant mixtures is necessary for the successful transition to new environmentally alternative refrigerants.[1−5] It is very important to obtain accurate and reliable measurements of the solubilities, densities, and viscosities of these mixtures because they are extremely important for the design of refrigeration compressors. Information on these thermophysical properties for refrigerant + lubricant systems is, however, very scarce.

Continuing our work on measurements and modeling of densities at high pressures of refrigerants, poly(ethylene glycol) dimethyl ethers, dialkyl carbonates, and their mixtures,[6−9] in this work we present new density values for mixtures containing HFC-134a and 2,5,8,11,14-pentaoxapentadecane, also named tetraethylene glycol dimethyl ether (TEGDME), in the compressed liquid phase. We have chosen this polyether because of its good miscibility and solubility with HFC-134a; this means that TEGDME is a good candidate as a lubricant of HFC-134a.

This paper reports new *pVT* measurements for eight binary mixtures of HFC-134a with TEGDME, at temperatures between 293.15 K and 373.15 K and at pressures from 10 MPa to 60 MPa, and excess molar volumes derived from the density results. The volumetric behavior of this mixture provides information on the interactions[6] between the HFC-134a and tetraethylene glycol dimethyl ether molecules and on the packing effect.[10] The *pVT* data presented in this work will be used in a future work, to test the density prediction with different equations of state combined with mixing rules.

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Experimental Section

Materials. HFC-134a (molar mass 102.03 g·mol^{-1}) was obtained from Gazechim Froid with a purity of 99.94% and with a water content not more than 24 ppm. TEGDME (molar mass 222.28 g·mol^{-1}) was obtained from Aldrich with a purity better than 99%.

Measurement Technique. The principle of measurement, the apparatus (Anton Paar DMA60/512P vibrating tube densimeter), and the experimental procedure for the density determination are described in detail in our previous work[7] concerning the *pVT* data of HFC-134a + triethylene glycol dimethyl ether mixtures. Because the lubricant and refrigerant are in two different phases at atmospheric pressure, accurate measurements of these mixtures require specific procedures for the preparation of the samples and the filling of the densimeter. The mixtures are prepared in an additional high-pressure variable-volume cell, containing a stainless steel ball (in order to agitate and homogenize the mixture) and equipped with a piston in order to isolate the lubricant/refrigerant mixture from the pressurizing fluid (oil). This high-pressure cell is pressurized up to a pressure higher than the saturation pressure to ensure that the mixture is in a single-phase.

It is also necessary to implement an isobaric transfer procedure[7,11] to ensure that the densimeter is loaded with the refrigerant + lubricant mixture in a monophasic liquid state and at the expected composition. The control of the temperature of the vibrating tube is performed with a thermoregulated liquid bath (Julabo Paratherm), for which fluctuations are within ±0.01 K. The temperature is measured inside the cell block with an AOIP 5207 thermometer which was calibrated to within ±0.05 K. The pressure is measured with an Hottinger Baldwin Messtechnik (HBM) manometer with an uncertainty of ±0.05 MPa. The calibration parameters of the densimeter cell were determined using vacuum and water as references.[12] The total uncertainty of the density is less than 2×10^{-4} g·cm^{-3}.

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Table 1. Experimental Values of Densities, ρ, for x HFC-134a + (1 − x) TEGDME at Different Temperatures, T, and Pressures, p

		ρ/(g·cm^{-3}) at the following values of T/K								
x	p/MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
0	0.1	1.0112	1.0018	0.9925	0.9833	0.9740	0.9648	0.9556	0.9469	0.9375
	5	1.0142	1.0050	0.9959	0.9868	0.9778	0.9687	0.9598	0.9513	0.9423
	10	1.0172	1.0082	0.9992	0.9903	0.9814	0.9726	0.9639	0.9557	0.9468
	15	1.0201	1.0113	1.0025	0.9937	0.9850	0.9764	0.9679	0.9598	0.9511
	20	1.0229	1.0143	1.0055	0.9970	0.9883	0.9800	0.9716	0.9639	0.9552
	25	1.0257	1.0172	1.0086	1.0001	0.9917	0.9835	0.9752	0.9675	0.9593
	30	1.0284	1.0200	1.0116	1.0032	0.9950	0.9869	0.9788	0.9712	0.9631
	35	1.0310	1.0227	1.0144	1.0063	0.9982	0.9901	0.9821	0.9748	0.9669
	40	1.0336	1.0254	1.0172	1.0091	1.0012	0.9932	0.9855	0.9783	0.9704
	45	1.0361	1.0281	1.0201	1.0121	1.0042	0.9963	0.9886	0.9815	0.9739
	50	1.0385	1.0306	1.0226	1.0147	1.0070	0.9993	0.9917	0.9846	0.9771
	55	1.0410	1.0332	1.0252	1.0175	1.0098	1.0023	0.9947	0.9880	0.9805
	60	1.0434	1.0355	1.0278	1.0202	1.0126	1.0050	0.9976	0.9910	0.9838
0.1114	10	1.0315	1.0220	1.0124	1.0031	0.9936	0.9844	0.9752	0.9659	0.9564
	15	1.0345	1.0254	1.0159	1.0067	0.9975	0.9887	0.9794	0.9704	0.9611
	20	1.0375	1.0285	1.0192	1.0103	1.0011	0.9925	0.9834	0.9746	0.9657
	25	1.0405	1.0316	1.0223	1.0136	1.0047	0.9962	0.9872	0.9787	0.9699
	30	1.0433	1.0346	1.0255	1.0169	1.0081	0.9998	0.9911	0.9828	0.9742
	35	1.0461	1.0374	1.0286	1.0201	1.0116	1.0032	0.9947	0.9866	0.9779
	40	1.0488	1.0403	1.0316	1.0232	1.0148	1.0065	0.9983	0.9903	0.9817
	45	1.0515	1.0431	1.0345	1.0263	1.0179	1.0099	1.0017	0.9937	0.9854
	50	1.0541	1.0457	1.0374	1.0291	1.0210	1.0129	1.0050	0.9972	0.9890
	55	1.0567	1.0485	1.0401	1.0321	1.0241	1.0162	1.0083	1.0006	0.9926
	60	1.0593	1.0510	1.0430	1.0349	1.0269	1.0192	1.0114	1.0038	0.9955
0.2896	10	1.0563	1.0460	1.0357	1.0255	1.0151	1.0050	0.9948	0.9847	0.9742
	15	1.0598	1.0497	1.0396	1.0296	1.0196	1.0097	0.9997	0.9899	0.9798
	20	1.0632	1.0533	1.0434	1.0337	1.0237	1.0142	1.0044	0.9948	0.9850
	25	1.0665	1.0568	1.0470	1.0375	1.0278	1.0185	1.0089	0.9995	0.9899
	30	1.0697	1.0601	1.0506	1.0412	1.0317	1.0225	1.0132	1.0039	0.9947
	35	1.0728	1.0634	1.0540	1.0447	1.0355	1.0265	1.0173	1.0083	0.9991
	40	1.0759	1.0666	1.0573	1.0482	1.0392	1.0303	1.0213	1.0125	1.0035
	45	1.0788	1.0697	1.0606	1.0517	1.0428	1.0339	1.0250	1.0164	1.0076
	50	1.0818	1.0727	1.0637	1.0548	1.0461	1.0374	1.0287	1.0203	1.0116
	55	1.0846	1.0757	1.0668	1.0581	1.0495	1.0410	1.0325	1.0241	1.0157
	60	1.0874	1.0785	1.0699	1.0613	1.0528	1.0443	1.0359	1.0277	1.0194
0.3648	10	1.0701	1.0593	1.0483	1.0376	1.0267	1.0160	1.0052	0.9945	0.9835
	15	1.0738	1.0632	1.0526	1.0420	1.0315	1.0211	1.0106	1.0002	0.9895
	20	1.0774	1.0671	1.0566	1.0464	1.0360	1.0259	1.0156	1.0055	0.9951
	25	1.0810	1.0708	1.0605	1.0504	1.0404	1.0304	1.0205	1.0106	1.0005
	30	1.0845	1.0744	1.0643	1.0544	1.0446	1.0349	1.0251	1.0153	1.0057
	35	1.0877	1.0779	1.0680	1.0583	1.0486	1.0391	1.0295	1.0201	1.0104
	40	1.0910	1.0813	1.0715	1.0620	1.0526	1.0431	1.0338	1.0245	1.0151
	45	1.0941	1.0846	1.0750	1.0657	1.0563	1.0470	1.0379	1.0288	1.0196
	50	1.0972	1.0878	1.0783	1.0691	1.0600	1.0508	1.0419	1.0330	1.0239
	55	1.1003	1.0910	1.0816	1.0725	1.0635	1.0546	1.0457	1.0369	1.0281
	60	1.1032	1.0939	1.0849	1.0759	1.0670	1.0581	1.0495	1.0408	1.0321
0.5702	10	1.1172	1.1041	1.0910	1.0780	1.0649	1.0516	1.0386	1.0250	1.0116
	15	1.1221	1.1094	1.0966	1.0838	1.0712	1.0585	1.0458	1.0331	1.0200
	20	1.1267	1.1144	1.1019	1.0897	1.0772	1.0649	1.0526	1.0401	1.0278
	25	1.1313	1.1191	1.1069	1.0949	1.0829	1.0710	1.0590	1.0471	1.0350
	30	1.1355	1.1236	1.1118	1.1001	1.0884	1.0767	1.0652	1.0534	1.0419
	35	1.1397	1.1280	1.1164	1.1050	1.0935	1.0822	1.0707	1.0595	1.0482
	40	1.1438	1.1324	1.1208	1.1097	1.0986	1.0874	1.0763	1.0654	1.0543
	45	1.1477	1.1364	1.1252	1.1143	1.1034	1.0924	1.0816	1.0708	1.0601
	50	1.1515	1.1405	1.1293	1.1187	1.1079	1.0972	1.0866	1.0761	1.0655
	55	1.1552	1.1443	1.1334	1.1229	1.1123	1.1021	1.0915	1.0813	1.0709
	60	1.1588	1.1482	1.1375	1.1271	1.1166	1.1063	1.0961	1.0860	1.0760
0.6931	10	1.1528	1.1377	1.1227	1.1074	1.0920	1.0766	1.0609	1.0450	1.0289
	15	1.1589	1.1444	1.1297	1.1149	1.1003	1.0854	1.0706	1.0553	1.0400
	20	1.1646	1.1505	1.1362	1.1222	1.1078	1.0936	1.0793	1.0649	1.0502
	25	1.1701	1.1562	1.1425	1.1287	1.1150	1.1013	1.0874	1.0735	1.0596
	30	1.1754	1.1619	1.1485	1.1351	1.1218	1.1084	1.0949	1.0816	1.0682
	35	1.1804	1.1673	1.1542	1.1411	1.1281	1.1151	1.1021	1.0893	1.0762
	40	1.1853	1.1724	1.1595	1.1469	1.1342	1.1216	1.1089	1.0964	1.0837
	45	1.1900	1.1774	1.1649	1.1524	1.1400	1.1276	1.1152	1.1031	1.0909
	50	1.1946	1.1822	1.1698	1.1576	1.1455	1.1335	1.1214	1.1096	1.0976
	55	1.1990	1.1867	1.1746	1.1627	1.1509	1.1391	1.1272	1.1156	1.1042
	60	1.2032	1.1912	1.1795	1.1677	1.1560	1.1444	1.1329	1.1216	1.1102

Table 1 (Continued)

		ρ/(g·cm^{-3}) at the following values of T/K								
x	p/MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
0.7288	10	1.1637	1.1480	1.1322	1.1161	1.0998	1.0834	1.0661	1.0490	1.0317
	15	1.1703	1.1551	1.1398	1.1242	1.1087	1.0931	1.0768	1.0606	1.0443
	20	1.1765	1.1617	1.1468	1.1321	1.1169	1.1019	1.0864	1.0709	1.0556
	25	1.1824	1.1680	1.1535	1.1392	1.1247	1.1102	1.0952	1.0805	1.0658
	30	1.1880	1.1740	1.1599	1.1459	1.1320	1.1180	1.1034	1.0892	1.0752
	35	1.1934	1.1797	1.1660	1.1524	1.1388	1.1253	1.1111	1.0975	1.0839
	40	1.1986	1.1852	1.1718	1.1585	1.1453	1.1321	1.1184	1.1053	1.0919
	45	1.2036	1.1905	1.1773	1.1644	1.1514	1.1385	1.1253	1.1124	1.0997
	50	1.2084	1.1956	1.1827	1.1700	1.1573	1.1447	1.1322	1.1194	1.1068
	55	1.2130	1.2004	1.1878	1.1754	1.1629	1.1507	1.1384	1.1259	1.1137
	60	1.2174	1.2051	1.1929	1.1806	1.1684	1.1563	1.1444	1.1322	1.1202
0.8727	10	1.2182	1.1972	1.1761	1.1543	1.1321	1.1092	1.0858	1.0613	1.0360
	15	1.2283	1.2083	1.1882	1.1676	1.1469	1.1256	1.1042	1.0819	1.0593
	20	1.2376	1.2184	1.1992	1.1797	1.1599	1.1401	1.1200	1.0993	1.0786
	25	1.2463	1.2278	1.2094	1.1907	1.1720	1.1531	1.1343	1.1147	1.0954
	30	1.2546	1.2366	1.2188	1.2009	1.1830	1.1649	1.1470	1.1284	1.1102
	35	1.2622	1.2449	1.2276	1.2103	1.1932	1.1758	1.1584	1.1409	1.1234
	40	1.2695	1.2528	1.2359	1.2192	1.2026	1.1859	1.1692	1.1523	1.1356
	45	1.2765	1.2601	1.2439	1.2276	1.2115	1.1952	1.1791	1.1629	1.1468
	50	1.2832	1.2671	1.2514	1.2355	1.2198	1.2040	1.1884	1.1728	1.1571
	55	1.2896	1.2740	1.2584	1.2430	1.2277	1.2125	1.1973	1.1820	1.1670
	60	1.2957	1.2803	1.2654	1.2503	1.2352	1.2202	1.2055	1.1908	1.1762
0.9290	10	1.2417	1.2169	1.1920	1.1657	1.1391	1.1111	1.0824	1.0514	1.0193
	15	1.2545	1.2311	1.2078	1.1833	1.1589	1.1336	1.1079	1.0806	1.0532
	20	1.2662	1.2438	1.2219	1.1989	1.1759	1.1525	1.1288	1.1043	1.0798
	25	1.2770	1.2555	1.2343	1.2128	1.1912	1.1692	1.1473	1.1245	1.1019
	30	1.2869	1.2664	1.2462	1.2255	1.2048	1.1842	1.1633	1.1420	1.1209
	35	1.2962	1.2765	1.2569	1.2371	1.2175	1.1976	1.1778	1.1577	1.1377
	40	1.3051	1.2860	1.2671	1.2479	1.2290	1.2100	1.1910	1.1720	1.1529
	45	1.3134	1.2949	1.2766	1.2581	1.2397	1.2214	1.2032	1.1848	1.1667
	50	1.3214	1.3032	1.2854	1.2676	1.2499	1.2322	1.2145	1.1969	1.1792
	55	1.3288	1.3114	1.2939	1.2766	1.2593	1.2422	1.2252	1.2080	1.1911
	60	1.3360	1.3190	1.3020	1.2852	1.2683	1.2516	1.2349	1.2185	1.2022
1	5	1.2478	1.2129	1.1759	1.1362	1.0933	1.0446	0.9888	0.9207	
	10	1.2689	1.2372	1.2045	1.1704	1.1343	1.0963	1.0557	1.0119	0.9635
	15	1.2873	1.2583	1.2283	1.1974	1.1657	1.1329	1.0991	1.0635	1.0262
	20	1.3035	1.2763	1.2485	1.2205	1.1914	1.1621	1.1318	1.1008	1.0690
	25	1.3182	1.2925	1.2665	1.2401	1.2132	1.1863	1.1589	1.1307	1.1023
	30	1.3317	1.3072	1.2826	1.2578	1.2324	1.2074	1.1816	1.1557	1.1298
	35	1.3442	1.3206	1.2971	1.2736	1.2496	1.2257	1.2017	1.1775	1.1532
	40	1.3559	1.3332	1.3105	1.2879	1.2651	1.2425	1.2197	1.1968	1.1740
	45	1.3667	1.3449	1.3231	1.3012	1.2795	1.2578	1.2359	1.2142	1.1924
	50	1.3770	1.3558	1.3347	1.3136	1.2926	1.2718	1.2509	1.2300	1.2092
	55	1.3867	1.3662	1.3456	1.3254	1.3051	1.2849	1.2647	1.2446	1.2248
	60	1.3958	1.3759	1.3561	1.3363	1.3165	1.2968	1.2775	1.2582	1.2389

Results and Discussion

Measurements of density were undertaken along nine isotherms between 293.15 K and 373.15 K at 10 K intervals and at pressures up to 60 MPa at 5 MPa intervals. A total of 1017 experimental points were obtained. The results are presented in Table 1 including those of the pure compounds. In a previous work,[9] we have presented the density values measured with the same apparatus up to 353.15 K for pure TEGDME. We have compared both sets of measurements, finding an average absolute deviation (AAD) of 0.01%, which shows the densimeter reproducibility. We also present here new experimental data for this pure compound at 363.15 K and 373.15 K as a function of pressure. Moreover, the values indicated for pure HFC-134a are the ones obtained and discussed in our previous work.[7] In the work concerning the HFC-134a + triethylene glycol dimethyl ether (TriEGDME) mixtures[7] we have compared our values for pure HFC-134a with those of the literature, finding a good agreement with other authors' density values.

In Figure 1, we present a view of the location of the investigated $p\rho T$ surface of pure HFC-134a in a ρT

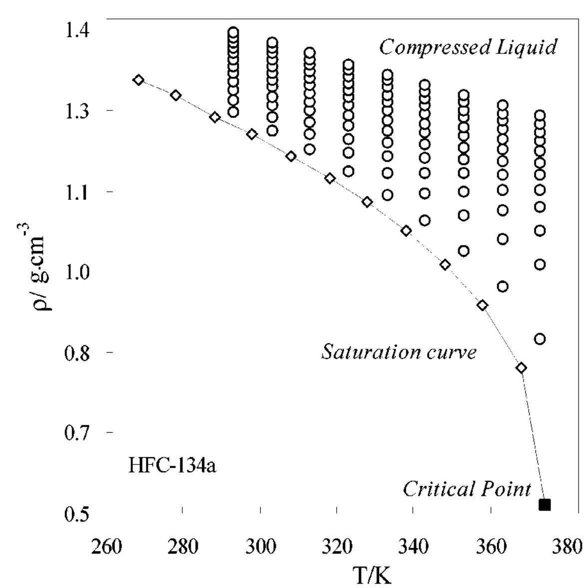

Figure 1. General view of the investigated $p\rho T$ surface of pure HFC-134a in a ρT diagram: ◇, saturation curve; ○, measured densities, our previous work;[7] ■, critical point.

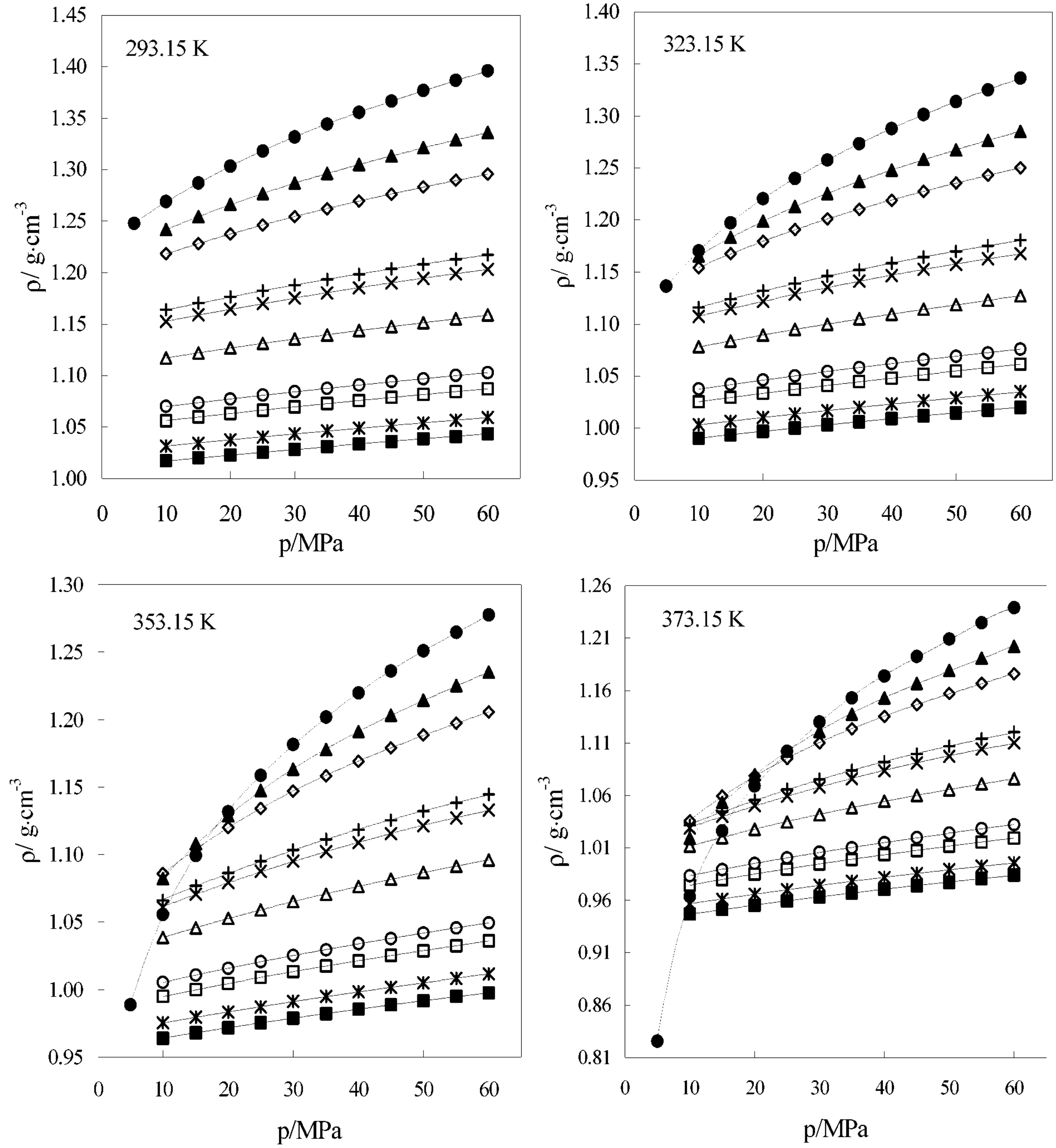

Figure 2. Experimental densities for x HFC-134a + $(1 - x)$ TEGDME mixtures at 293.15 K, 323.15 K, 353.15 K, and 373.15 K versus the pressure for $x = 1.0$ (●), 0.9290 (▲), 0.8727 (◇), 0.7288 (+), 0.6931 (×), 0.5702 (△), 0.3648 (○), 0.2896 (□), 0.1114 (∗), and 0.0 (■). (—) Polynomial fitting for guiding the eye.

diagram. These experimental values could be used to validate the new equations of state[13,14] formulated for the thermodynamic properties of this refrigerant. We have plotted in Figure 2 the experimental densities for x HFC-134a + $(1 - x)$ TEGDME mixtures versus the pressure for different x values, at four temperatures, 293.15 K, 323.15 K, 353.15 K, and 373.15 K. For some isotherms the density has an intersection point of isocomposition lines. For the highest temperatures and the lowest pressures the density of some mixtures becomes greater than that for the pure HFC-134a. The crossing point appears, as can be seen in Figure 2, at a pressure (crossover pressure) that is higher at higher temperatures. For example, at 353.15 K the crossing point appears around 15 MPa, and at 373.15 K it appears around 25 MPa. This behavior has also been found in our previous work concerning the HFC-134a + triethylene glycol dimethyl ether mixture. This is probably due to the fact that the polyether is less compressible than the refrigerant. Similar behavior has been found by Kiran et al.[15] and by Pöhler et al.[16] for mixtures containing carbon dioxide and by Cavestri and Schafer[17] for R-410A with four polyolester lubricants.

The excess molar volumes, V_m^E, at each pressure p and temperature T, can be calculated from our experimental density values using the relation

$$V_m^E(T,p,x) = V_m(T,p,x) - (xV_{m,1}(T,p) + (1 - x)V_{m,2}(T,p)) \quad (1)$$

where $V_m = M_m/\rho$ ($M_m = xM_1 + (1 - x)M_2$, where M_i is the molar mass of component i) is the molar volume of the mixtures at each pressure and temperature and $V_{m,i} = M_i/\rho_i$ is the molar volume of compound i (ρ_i is the density

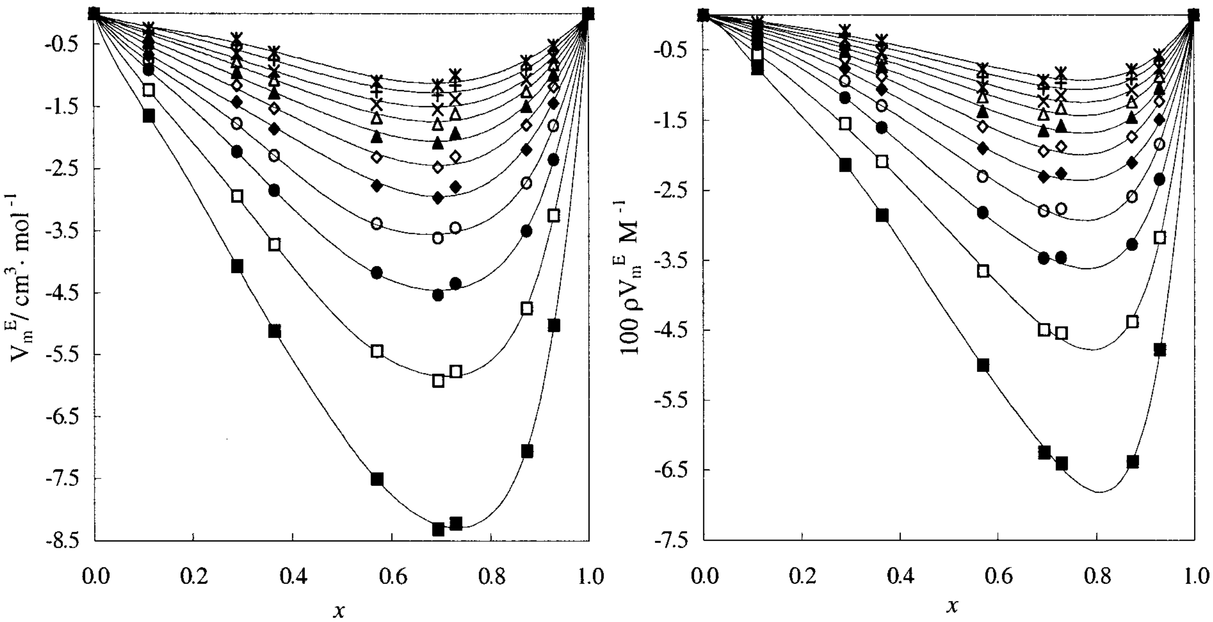

Figure 3. Pressure (p) and refrigerant molar fraction (x) dependencies of (a, left) the experimental excess volumes V_m^E (cm^3·mol^{-1}) and (b, right) the $\rho V_m^E/M$ (%) at 363.15 K: ■, 10 MPa; □, 15 MPa; ●, 20 MPa; ○, 25 MPa; ◆, 30 MPa; ◇, 35 MPa; ▲, 40 MPa; △, 45 MPa; ×, 50 MPa; +, 55 MPa; ∗, 60 MPa. (—) Polynomial fitting for guiding the eye.

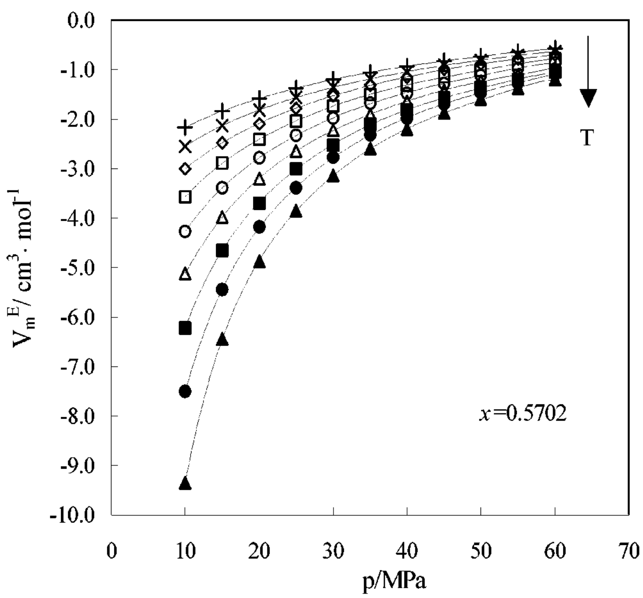

Figure 4. Temperature (T) and pressure (p) dependencies of V_m^E for x HFC-134a + (1 − x) TEGDME mixtures with x = 0.5702: +, 293.15 K; ×, 303.15 K; ◇, 313.15 K; □, 323.15 K; ○, 333.15 K; △, 343.15 K; ■, 353.15 K; ●, 363.15 K; ▲, 373.15 K. (—) Polynomial fitting for guiding the eye.

of component i). Figure 3a shows the variations of V_m^E versus the refrigerant molar fraction, at 363.15 K and at different pressures. The quantity $\rho V_m^E/M_m$ relative to the unit volume is represented versus the HFC-134a composition in Figure 3b. In Figure 4, we have represented the excess molar volume at the closest composition to the equimolar fraction against the pressure for different temperatures. At all the temperatures and pressures this excess property is negative and asymmetrical toward high refrigerant composition. At fixed composition and temperature, V_m^E is more negative when the pressure decreases, and at fixed composition and pressure V_m^E becomes more negative when the temperature increases. This behavior is very similar to that of HFC-134a + triethylene glycol dimethyl ether. The strong negative values of V_m^E are in agreement with the previous remarks of Tseregounis and Riley[18] about the high degree of interaction in the mixture of HFC-134a with tetraethylene glycol dimethyl ether. In fact, other mixtures of HFCs with ethers present strong interactions between the unlike molecules, for example HFC236ea or HFC236fa with dimethyl ether, as Bobbo et al.[19,20] have concluded from VLE measurements. Nevertheless, in the case of excess molar volumes of HFC-134a with long molecules such as 2,5,8,11,14-pentaoxapentadecane, the free-volume or packing effects could give an important negative contribution.[10]

Conclusions

The volumetric behavior of HFC-134a + polyalkylglycol mixtures has been analyzed in wide temperature and pressure ranges. In the literature, there are very few data at high-pressure concerning the densities of these mixtures. In our previous work, we have analyzed the HFC-134a + triethylene glycol dimethyl ether mixtures, and in the present article we have reported the experimental density (pVT) and excess volume results for the compressed liquid HFC-134a + tetraethylene glycol dimethyl ether system. For the two systems the density has a crossover point of the isocomposition lines at higher temperatures. The study displays the high degree of interaction between the lubricant and refrigerant molecules. It is hoped that the data will aid the formulation of new correlations and the test of new models of refrigerant−lubricant mixtures.

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