# **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.<sup>1–5</sup> 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,<sup>6–9</sup> in this work we present new density values for mixtures containing HFC-134a and 2,5,8,11,14-pen-taoxapentadecane, 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<sup>6</sup> between the HFC-134a and tetraethylene glycol dimethyl ether molecules and on the packing effect.<sup>10</sup> 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.

#### **Experimental Section**

*Materials.* HFC-134a (molar mass 102.03 g·mol<sup>-1</sup>) 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<sup>-1</sup>) 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<sup>7</sup> 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<sup>7.11</sup> 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.<sup>12</sup> The total uncertainty of the density is less than  $2 \times 10^{-4}$  g·cm<sup>-3</sup>.

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Table 1. Experimental Values of Densities, $\rho$ , for x HFC-134a + (1 - x) TEGDME at Different Temperatures, T, an Pressures, p
$a/(a \text{ cm}^{-3})$ at the following values of $TK$

x         ρMPa         293.15         313.15         313.15         323.15         333.15         343.15         353.15         353.15           0         5         10142         1.0050         0.9920         0.9948         0.9948         0.9948         0.9948         0.9959         0.9947         0.9947         0.9959         0.9945         0.9963         0.9454         0.9459         0.9559           15         1.0201         1.0113         1.0025         0.9907         0.9883         0.9764         0.9639         0.9548           25         1.0257         1.0172         1.0066         1.0901         0.9917         0.9883         0.9752         0.9788         0.9774           35         1.0310         1.0227         1.0144         1.0062         0.9982         0.99910         0.9881         0.9774         0.9783         0.97748         0.9774         0.9784         0.9774         0.9884         0.9774         0.9891         0.9774         0.9884         0.9774         0.9884         0.9774         0.9874         0.9774         0.9874         0.9774         0.9875         0.9774         0.9875         0.9774         0.9875         0.9774         0.9875         0.9774         0.9876         0.9774				$\rho/(g\cdot cm^{-3})$ at the following values of <i>TT</i> K									
0         0.1         1.0112         1.018         0.9953         0.9833         0.9740         0.9648         0.9573         0.9573           10         1.0172         1.0082         0.9997         0.9883         0.9870         0.9583         0.9573           20         1.0229         1.0143         1.0025         0.9970         0.9883         0.9764         0.9573           21         1.0227         1.0143         1.0062         0.99970         0.9883         0.9764         0.9863           23         1.0227         1.0172         1.0086         0.9982         0.9960         0.9748         0.9732           30         1.0224         1.0172         1.0091         1.0012         0.9932         0.9838         0.9743           40         1.0335         1.0221         1.0124         1.0081         0.9932         0.9844         0.9752         0.9874         0.9747         0.9844         0.9752         0.9874         0.9774         0.9816           0.1114         1.035         1.0220         1.0172         1.0075         0.9877         0.9794         0.9774         0.9816           0.1114         1.0351         1.0221         1.01071         0.9997         0.9816<	X	<i>p</i> /MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15		
5         1.0142         1.0050         0.9992         0.9983         0.9178         0.9087         0.9537           15         1.0201         1.0113         1.0052         0.9937         0.9853         0.9744         0.9639         0.9537           20         1.0227         1.0172         1.0068         1.0052         0.9950         0.9863         0.9716         0.9639           35         1.0310         1.0227         1.0114         1.0063         0.9950         0.9863         0.9878         0.9778           40         1.0336         1.0227         1.0114         1.0063         0.9963         0.9885         0.9783           45         1.0381         1.0227         1.0141         1.0061         0.9963         0.9886         0.9814           45         1.0381         1.0228         1.0127         1.0126         0.9976         0.9810         0.9814         0.9772         0.9817         0.9819         0.9744         0.9762         0.9814         0.9778         0.9814         0.9778         0.9814         0.9778         0.9814         0.9778         0.9814         0.9778         0.9814         0.9778         0.9814         0.9778         0.9814         0.9778         0.9816	)	0.1	1.0112	1.0018	0.9925	0.9833	0.9740	0.9648	0.9556	0.9469	0.9375		
10         1.0172         1.0082         0.9993         0.9814         0.9726         0.9673         0.9583           20         1.0229         1.0143         1.0055         0.9970         0.9883         0.9800         0.9776         0.9593           20         1.0229         1.0143         1.0085         0.0910         0.9813         0.9803         0.9803         0.9803         0.9863         0.9752         0.9675           30         1.0224         1.0021         1.0011         0.03821         0.9743         0.9743           41         1.0311         1.0221         1.0171         1.0142         0.9903         0.9863         0.9904         0.9814         0.9744         0.9744           45         1.0315         1.0220         1.0174         1.0070         0.9993         0.9814         0.9776         0.9910           55         1.0410         1.0332         1.0224         1.01071         1.0022         0.9946         0.9917         0.9846           56         1.0413         1.0351         1.0220         1.0114         1.0021         0.9916         0.9910           1.1114         10         1.0375         1.0223         1.0116         1.0023         0.9973 <td></td> <td>5</td> <td>1.0142</td> <td>1.0050</td> <td>0.9959</td> <td>0.9868</td> <td>0.9778</td> <td>0.9687</td> <td>0.9598</td> <td>0.9513</td> <td>0.9423</td>		5	1.0142	1.0050	0.9959	0.9868	0.9778	0.9687	0.9598	0.9513	0.9423		
15         1.0201         1.0113         1.0025         0.9937         0.9853         0.9764         0.9639           25         1.0257         1.0172         1.0084         1.0001         0.9917         0.9833         0.9776         0.9833           35         1.0257         1.0174         1.0083         0.9982         0.9901         0.9835         0.9778           40         1.0330         1.0227         1.0144         1.0012         0.9936         0.9885         0.9783           40         1.0331         1.0252         1.0172         1.0014         0.9963         0.9885         0.9783           41         1.0331         1.0252         1.0175         1.0050         0.9977         0.9887         0.9794         0.9891           40         1.0335         1.0254         1.0123         1.0013         0.9987         0.9887         0.9794         0.9891           411         1.0335         1.0254         1.0123         1.0013         0.9962         0.9873         0.9794           410         1.0436         1.0253         1.0136         1.0264         1.0932         0.9887         0.9764           411         1.0457         1.0254         1.0123		10	1.0172	1.0082	0.9992	0.9903	0.9814	0.9726	0.9639	0.9557	0.9468		
20         1.0229         1.0143         1.0065         0.0970         0.9883         0.9800         0.9772         0.9675           30         1.0284         1.0000         1.0114         1.0063         0.9950         0.9869         0.9782         0.9675           30         1.0284         1.0072         1.0061         0.9912         0.9861         0.9782         0.9783           40         1.0336         1.0221         1.1042         0.9812         0.9861         0.9882         0.9861         0.9783           50         1.0345         1.0231         1.0212         1.0121         1.0042         0.9863         0.9864         0.9813           50         1.0454         1.0353         1.0275         1.0198         1.0051         0.9974         0.9886           60         1.0453         1.0255         1.0167         0.9925         0.9774         0.9787           51         1.0451         1.0255         1.0168         1.0041         0.9925         0.9872         0.9777           50         1.0451         1.0325         1.0168         1.00471         0.9992         0.9847         0.9866           51         1.0453         1.0473         1.02291		15	1.0201	1.0113	1.0025	0.9937	0.9850	0.9764	0.9679	0.9598	0.951		
25         1.0227         1.0172         1.0081         1.0001         0.9917         0.9833         0.9728         0.9712           35         1.0310         1.0227         1.0144         1.0063         0.9982         0.9901         0.9835         0.9788         0.9712           40         1.0336         1.0227         1.0144         1.0061         1.0012         0.9963         0.9865         0.9783           45         1.0336         1.0228         1.0174         1.0070         0.9993         0.99817         0.9846           55         1.0410         1.0332         1.0228         1.0147         1.0070         0.9993         0.9916         0.9916           1.0141         1.0315         1.0220         1.0124         1.0031         0.9924         0.9764         0.9774           20         1.0375         1.0283         1.0122         1.0167         0.9875         0.9846         0.9774		20	1.0229	1.0143	1.0055	0.9970	0.9883	0.9800	0.9716	0.9639	0.9552		
30         1.0284         1.0200         1.0116         1.0063         0.9950         0.9819         0.9748         0.9748           40         1.0336         1.0254         1.0121         1.0063         0.9952         0.9821         0.9748           40         1.0356         1.0254         1.0121         1.0042         0.9993         0.9917         0.9486           50         1.0355         1.0226         1.0147         1.0070         0.9993         0.9917         0.9486           60         1.0355         1.0226         1.0147         1.0070         0.9993         0.9917         0.9800           21114         1.0345         1.0228         1.0159         1.0076         0.9975         0.9874         0.9774           20         1.0375         1.0285         1.0169         1.0047         0.9982         0.9874         0.9774           20         1.0375         1.0285         1.0169         1.0081         0.9918         0.9774         0.9874           21         1.0434         1.0285         1.0169         1.0977         0.9874         0.9774           30         1.0431         1.0376         1.0291         1.0161         0.9978         0.9877		25	1.0257	1.0172	1.0086	1.0001	0.9917	0.9835	0.9752	0.9675	0.9593		
35         1.0310         1.0227         1.0144         1.0091         1.00912         0.9982         0.9855         0.9783           45         1.0361         1.0281         1.0221         1.0121         1.0042         0.9863         0.9855         0.9346           55         1.0410         1.0322         1.0222         1.0175         1.0098         0.9917         0.9346           55         1.0410         1.0322         1.0222         1.0175         1.0098         0.9917         0.9346           1.1114         10         1.0315         1.0226         1.0122         1.0128         1.0093         0.9976         0.9887         0.9774         0.9872         0.9774           20         1.0375         1.0225         1.0167         0.9775         0.9887         0.9778         0.9872         0.9874         0.9778           35         1.0461         1.0255         1.0167         1.0466         0.9992         0.9972         0.9874         0.9778           36         1.0461         1.0457         1.0252         1.0161         1.0095         0.9993         0.9972           37         1.0481         1.0252         1.0179         1.09991         0.9972         0.9972 <td></td> <td>30</td> <td>1.0284</td> <td>1.0200</td> <td>1.0116</td> <td>1.0032</td> <td>0.9950</td> <td>0.9869</td> <td>0.9788</td> <td>0.9712</td> <td>0.963</td>		30	1.0284	1.0200	1.0116	1.0032	0.9950	0.9869	0.9788	0.9712	0.963		
40         1.03:50         1.02:41         1.0121         1.0012         0.99:52         0.98:55         0.97:53           50         1.0385         1.0206         1.0228         1.0171         1.0070         0.99:83         0.99:17         0.98:60           50         1.0316         1.0228         1.0175         1.0070         0.99:93         0.99:17         0.98:60           60         1.0434         1.0355         1.0278         1.0221         1.0126         1.0050         0.9975         0.98:59           1114         1         0.0315         1.0224         1.0135         1.0011         0.99:52         0.98:34         0.9774         0.9776         0.99:59           1114         1.0345         1.0224         1.0135         1.0061         0.99:82         0.9774         0.9774           25         1.0405         1.0316         1.0223         1.0143         1.0065         0.98:33         0.9033           45         1.0515         1.0431         1.0234         1.0221         1.0121         1.01032         0.99:17         0.98:23           45         1.0516         1.069:1         1.0316         1.0223         1.0116         1.0029         0.0177         0.99:53		35	1.0310	1.0227	1.0144	1.0063	0.9982	0.9901	0.9821	0.9748	0.9669		
30         1.0301         1.0201         1.0121         1.0012         0.3933         0.3933         0.3944           35         1.0410         1.0322         1.0125         1.0070         0.9933         0.9917         0.3944           35         1.0410         1.0322         1.0125         1.0023         0.9947         0.9844           1114         10         1.0315         1.0220         1.0124         1.0031         0.9936         0.9844         0.9772         0.9867           20         1.0375         1.0228         1.0167         0.9975         0.9887         0.9774           20         1.0375         1.0228         1.0167         0.9992         0.9872         0.9787           30         1.0433         1.0346         1.0225         1.0146         1.0092         0.9993         0.9911         0.9826           40         1.0488         1.0403         1.0324         1.0210         1.0116         1.0035         0.9937           50         1.0541         1.0345         1.0321         1.0241         1.0162         1.0983         0.9993           50         1.0541         1.0345         1.0321         1.0241         1.0162         1.0983 <t< td=""><td></td><td>40</td><td>1.0336</td><td>1.0254</td><td>1.01/2</td><td>1.0091</td><td>1.0012</td><td>0.9932</td><td>0.9855</td><td>0.9783</td><td>0.9704</td></t<>		40	1.0336	1.0254	1.01/2	1.0091	1.0012	0.9932	0.9855	0.9783	0.9704		
30         1.0309         1.0229         1.0175         1.0008         1.0239         0.3947         0.39407         0.39407           60         1.0434         1.0355         1.0228         1.0212         1.0126         1.0058         0.0927         0.39407         0.39407         0.39407           1114         10         1.0355         1.0228         1.0217         1.0088         0.0927         0.3957         0.3957         0.3957         0.3957         0.3957         0.3957         0.3957         0.3957         0.3974         0.3774         0.		45	1.0301	1.0206	1.0201	1.0121	1.0042	0.9903	0.9880	0.9815	0.973		
		55	1.0365	1.0300	1.0220	1.0147	1.0070	0.9993	0.9917	0.9840	0.977		
1114         10         1.0315         1.0220         1.0124         1.0031         1.0938         1.09847         0.9752         0.9659           20         1.0375         1.0254         1.0159         1.0067         0.9975         0.9887         0.9774         0.9774           20         1.0375         1.0285         1.0136         1.0011         0.9925         0.9872         0.9771           30         1.0435         1.0164         1.0022         0.9872         0.9777         0.9872         0.9777           30         1.0445         1.0223         1.0161         1.0023         0.9947         0.9866           40         1.0488         1.0403         1.0286         1.0210         1.0165         0.9947         0.9866           50         1.0541         1.0435         1.0243         1.0210         1.0122         1.0006         0.9937           53         1.0550         1.0440         1.0337         1.0224         1.0017         0.9937           54         1.0653         1.0440         1.0337         1.0241         1.0162         1.0083           2896         10         1.0533         1.0434         1.0337         1.0225         1.0114 <td< td=""><td></td><td>60</td><td>1.0410</td><td>1.0352</td><td>1.0252</td><td>1.0175</td><td>1.0038</td><td>1.0025</td><td>0.3347</td><td>0.9880</td><td>0.380</td></td<>		60	1.0410	1.0352	1.0252	1.0175	1.0038	1.0025	0.3347	0.9880	0.380		
15         1.0345         1.0254         1.0159         1.0067         0.9975         0.9887         0.9794         0.9704           20         1.0375         1.0285         1.0192         1.0103         1.0011         0.9925         0.9834         0.9704           25         1.0405         1.0285         1.0168         1.0042         0.9942         0.9847         0.9784           30         1.0433         1.0346         1.0225         1.0168         1.0062         0.9948         0.9948         0.9948         0.99493           45         1.0515         1.0434         1.0324         1.0116         1.0052         0.9947         0.9983         0.9903           50         1.0547         1.0374         1.02291         1.0122         1.0050         0.9948         0.9972           55         1.0567         1.0485         1.04047         1.0321         1.0162         1.0050         0.9948         0.9847           10         1.0532         1.0433         1.0349         1.02241         1.0164         1.0077         0.9889         0.0114         1.0038         1.0429         1.0192         1.0014         1.0038         1.0273         1.0044         1.0374         1.0255 <t< td=""><td>1114</td><td>10</td><td>1.0434</td><td>1.0333</td><td>1.0278</td><td>1.0202</td><td>0.9936</td><td>0 9844</td><td>0.9752</td><td>0.9659</td><td>0.965</td></t<>	1114	10	1.0434	1.0333	1.0278	1.0202	0.9936	0 9844	0.9752	0.9659	0.965		
20         1.0375         1.0285         1.0192         1.0013         1.0011         0.9925         0.9834         0.9746           25         1.0433         1.0346         1.0225         1.0168         1.0047         0.9962         0.9877         0.9787           30         1.0433         1.0346         1.0225         1.0161         1.0025         0.9947         0.9866           40         1.0488         1.0403         1.0314         1.0045         0.9947         0.9866           45         1.0515         1.0431         1.0345         1.0221         1.0112         1.0050         0.9937           50         1.0567         1.0485         1.0401         1.0321         1.0210         1.0129         1.0083         1.0096           60         1.0593         1.0401         1.0337         1.0226         1.0118         1.0083         0.9948         0.9847           2896         10         1.0533         1.0431         1.0337         1.0226         1.0118         1.0089         0.9948         0.9948         0.9847           2896         1.0667         1.0568         1.0470         1.0375         1.0278         1.0041         0.9948         0.9948         0.9948	.1114	15	1.0345	1 0254	1 0159	1.0051	0.9975	0.9887	0.9792	0.9704	0.961		
25         1.0405         1.0316         1.0223         1.0138         1.0047         0.9992         0.9872         0.9787           30         1.0433         1.0346         1.0252         1.0161         1.0032         0.9947         0.9888           35         1.0461         1.0374         1.0286         1.0211         1.0148         0.0032         0.9947         0.9863           40         1.0488         1.0403         1.0374         1.0222         1.0148         1.0060         0.9983         0.9903           50         1.0571         1.0485         1.0401         1.0321         1.0210         1.0122         1.0063         0.9972           50         1.0567         1.0485         1.0401         1.0321         1.0234         1.0039         1.0070         0.9987         0.9987         0.9987         1.0050         0.9947         0.9887           10         1.0558         1.0410         1.0330         1.0259         1.0161         1.0030         1.0342         1.0033         1.025         1.0131         1.0412         1.0044         0.9484           25         1.0655         1.0568         1.0470         1.0375         1.0278         1.0142         1.0044         0.9		20	1.0375	1.0285	1.0192	1.0103	1.0011	0.9925	0.9834	0.9746	0.965		
30         1.0433         1.0346         1.0255         1.0149         1.0081         0.9981         0.9886           35         1.0441         1.0254         1.0116         1.0032         0.9933         0.9903           40         1.0488         1.0403         1.0316         1.0232         1.0148         1.0055         0.9983         0.9903           45         1.0515         1.0437         1.0243         1.0179         1.0065         0.9982           55         1.0567         1.0485         1.0401         1.0321         1.0241         1.0122         1.0114         1.0038           60         1.0583         1.0400         1.0357         1.0255         1.0151         1.0080         0.9997         0.9899           20         1.0632         1.0353         1.0434         1.0337         1.0237         1.0142         1.0044         0.9948           30         1.0697         1.0606         1.0573         1.0272         1.0131         1.0225         1.0132         1.0032           30         1.0697         1.0606         1.0571         1.0428         1.0333         1.0213         1.0125           40         1.0759         1.0606         1.0571		25	1.0405	1.0316	1.0223	1.0136	1.0047	0.9962	0.9872	0.9787	0.9699		
35         1.0461         1.0374         1.0286         1.0201         1.0116         1.0055         0.9947         0.9863         0.9903           45         1.0515         1.0431         1.0345         1.0263         1.0179         1.0065         0.9993         0.9903           50         1.0547         1.0374         1.02291         1.0210         1.0162         1.0063         1.0066           60         1.0567         1.0485         1.0401         1.0321         1.0249         1.0162         1.0083         1.0066           10         1.0563         1.0401         1.0337         1.0250         0.9972         0.9972           20         1.0653         1.0404         1.04940         1.0336         1.0251         1.0133         0.9979         0.9897         0.9897         0.9897         0.9897         0.9897         0.9897         0.9894         0.0173         1.0275         1.0162         1.0078         0.9948         0.9948         0.9947         0.9917         0.9897         0.9897         0.9897         0.9897         0.9897         0.9897         0.9894         0.0213         1.0125         1.0131         1.0252         1.0121         1.0032         1.0225         1.0123         1.0225		30	1.0433	1.0346	1.0255	1.0169	1.0081	0.9998	0.9911	0.9828	0.9742		
40         1.0488         1.0403         1.0316         1.0232         1.0148         1.0059         0.0993           50         1.0541         1.0437         1.0374         1.0291         1.0210         1.0129         1.0059         0.0907           55         1.0563         1.0485         1.0401         1.0321         1.0241         1.0162         1.0030         0.9972           2896         10         1.0583         1.0460         1.0357         1.0225         1.0151         1.0080         0.9997         0.98899           20         1.0662         1.0568         1.0440         1.0337         1.0237         1.0142         1.0044         0.9948           30         1.0697         1.0601         1.0506         1.0412         1.0317         1.0225         1.0163         1.0043           40         1.0759         1.0666         1.0573         1.0482         1.0392         1.0230         1.0213         1.0250           50         1.0818         1.0727         1.0637         1.0548         1.0491         1.0324         1.0225         1.0164           50         1.0818         1.0727         1.0548         1.0461         1.0374         1.0226         1.0214		35	1.0461	1.0374	1.0286	1.0201	1.0116	1.0032	0.9947	0.9866	0.9779		
45         1.0515         1.0431         1.0345         1.0263         1.0179         1.0029         1.0050         0.9972           55         1.0567         1.0485         1.0401         1.0321         1.0241         1.0162         1.0083         1.0008           60         1.0563         1.0460         1.0357         1.0255         1.0151         1.0050         0.9948         0.9847           15         1.0563         1.0460         1.0357         1.0255         1.0151         1.0050         0.9948         0.9947           25         1.0658         1.0470         1.0375         1.0278         1.0142         1.0044         0.9948           25         1.0665         1.0470         1.0375         1.0278         1.0132         1.0039           35         1.0728         1.0664         1.0577         1.0428         1.0339         1.0213         1.0025           45         1.0788         1.0697         1.0666         1.0577         1.0428         1.0339         1.0225         1.0141         1.0325         1.0243           50         1.0818         1.0777         1.0668         1.0451         1.0411         1.0325         1.0241           50		40	1.0488	1.0403	1.0316	1.0232	1.0148	1.0065	0.9983	0.9903	0.981		
50         1.0541         1.0457         1.0374         1.0210         1.0129         1.0053         0.09972           60         1.0593         1.0510         1.0330         1.0349         1.0269         1.0192         1.0114         1.0038           2896         10         1.0598         1.0467         1.0396         1.0255         1.0151         1.0089         0.9948         0.9847           2896         1.0652         1.0653         1.0437         1.0236         1.0111         1.0047         0.9997         0.9899           20         1.0632         1.0556         1.0470         1.0375         1.0278         1.0666         1.0576         1.0412         1.0317         1.0225         1.0123         1.0083           30         1.0697         1.0666         1.0573         1.0482         1.0392         1.0333         1.0213         1.0125           40         1.0759         1.0666         1.0577         1.0448         1.0491         1.0325         1.0241         1.0235         1.0211         1.0203         1.0214         1.0235         1.0277           3648         10         1.0774         1.0674         1.0784         1.0665         1.0414         1.0354         1.0		45	1.0515	1.0431	1.0345	1.0263	1.0179	1.0099	1.0017	0.9937	0.985		
55         1.0567         1.0485         1.0401         1.0321         1.0241         1.0122         1.0014         1.0038           2896         10         1.0563         1.0460         1.0357         1.0255         1.0151         1.0050         0.9948         0.9847           120         1.0652         1.0533         1.0434         1.0337         1.0237         1.0142         1.0044         0.9948           25         1.0665         1.0566         1.0470         1.0375         1.0278         1.0183         1.0089         0.9993           35         1.0728         1.0666         1.0573         1.0442         1.0339         1.0213         1.0023           40         1.0759         1.0666         1.0577         1.0428         1.0339         1.0213         1.0125           45         1.0788         1.0697         1.0668         1.0517         1.0428         1.0339         1.0227         1.033           55         1.0846         1.0757         1.0668         1.0528         1.0410         1.0325         1.0211           10         1.0774         1.0661         1.0454         1.0434         1.0325         1.0217           1.0810         1.0778		50	1.0541	1.0457	1.0374	1.0291	1.0210	1.0129	1.0050	0.9972	0.989		
60         1.0593         1.0510         1.0430         1.0349         1.0269         1.0192         1.0114         1.0038           2896         15         1.0558         1.0497         1.0357         1.0255         1.0116         1.0097         0.9997         0.8899           20         1.0632         1.0533         1.0434         1.0375         1.0278         1.0142         1.0044         0.9948           30         1.0697         1.0601         1.0506         1.0412         1.0317         1.0225         1.0132         1.0039           35         1.0728         1.0666         1.0573         1.0428         1.0333         1.0213         1.0288           40         1.0758         1.0666         1.0573         1.0482         1.0339         1.0225         1.0134         1.0287         1.0203           55         1.0846         1.0757         1.0687         1.0641         1.0374         1.0287         1.0221         1.0101         1.0325         1.0211         1.0325         1.0211         1.0325         1.0211         1.0325         1.0211         1.0325         1.0211         1.0325         1.0211         1.0325         1.0211         1.0325         1.0211         1.0253		55	1.0567	1.0485	1.0401	1.0321	1.0241	1.0162	1.0083	1.0006	0.992		
2896         10         1.0563         1.0460         1.0357         1.0255         1.0151         1.0050         0.9948         0.9889           20         1.0632         1.0533         1.0434         1.0337         1.0237         1.0142         1.0044         0.9948           25         1.0665         1.0568         1.0470         1.0375         1.0278         1.0142         1.0044         0.9995           30         1.0667         1.0601         1.0506         1.0412         1.0317         1.0225         1.0133         1.0213           40         1.0759         1.0666         1.0573         1.0442         1.0339         1.0250         1.0164           55         1.0846         1.0757         1.0668         1.0514         1.0428         1.0339         1.0250         1.0244           55         1.0846         1.0757         1.0668         1.0514         1.0434         1.0359         1.0247           60         1.0874         1.0785         1.0669         1.0513         1.0528         1.0443         1.0359         1.0247           10         1.0774         1.0652         1.0470         1.0364         1.0439         1.0251         1.0106		60	1.0593	1.0510	1.0430	1.0349	1.0269	1.0192	1.0114	1.0038	0.995		
15         1.0598         1.0497         1.0396         1.0296         1.0196         1.0097         0.9997         0.8999           25         1.06632         1.0333         1.0470         1.0375         1.0278         1.0185         1.0089         0.9995           30         1.06697         1.0640         1.0375         1.0278         1.0185         1.0089         0.9995           35         1.0728         1.0644         1.0340         1.0447         1.0355         1.0255         1.0132         1.0089           40         1.0758         1.0666         1.0571         1.0482         1.0332         1.0333         1.0213         1.0125           50         1.0846         1.0777         1.0668         1.0581         1.0495         1.0410         1.0325         1.0241           51         1.0738         1.0622         1.0613         1.0267         1.0160         1.0052         0.9945           3648         10         1.0701         1.0566         1.0420         1.0315         1.0211         1.0106         1.0052           20         1.0774         1.0661         1.0564         1.0404         1.0349         1.0251         1.0153           30	.2896	10	1.0563	1.0460	1.0357	1.0255	1.0151	1.0050	0.9948	0.9847	0.974		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		15	1.0598	1.0497	1.0396	1.0296	1.0196	1.0097	0.9997	0.9899	0.979		
25         1.0665         1.0568         1.0470         1.0375         1.0278         1.0185         1.0089         0.9995           35         1.0728         1.0634         1.0540         1.0442         1.0317         1.0225         1.0133         1.0083           40         1.0759         1.0666         1.0573         1.0482         1.0325         1.0213         1.0083           45         1.0788         1.0697         1.0668         1.0517         1.0428         1.0339         1.02250         1.0164           50         1.0846         1.0757         1.0668         1.0581         1.0445         1.0410         1.0325         1.0241           60         1.0874         1.0753         1.0668         1.0420         1.0315         1.0211         1.0106         1.0002           20         1.0774         1.0612         1.0526         1.0442         1.0334         1.0255         1.0010           30         1.0845         1.0704         1.0566         1.0442         1.0344         1.0255         1.0012           25         1.0810         1.0778         1.0665         1.0420         1.0315         1.0251         1.0153           30         1.0845		20	1.0632	1.0533	1.0434	1.0337	1.0237	1.0142	1.0044	0.9948	0.985		
30         1.0697         1.0601         1.0540         1.0447         1.0355         1.0225         1.0132         1.0033           40         1.0759         1.06634         1.0540         1.0447         1.0355         1.02265         1.0173         1.0083           40         1.0759         1.0666         1.0573         1.0482         1.0393         1.0213         1.0125           45         1.0788         1.0697         1.06637         1.0548         1.0461         1.0374         1.02267         1.0164           55         1.0846         1.0777         1.0669         1.0517         1.0461         1.0374         1.0252         0.9945           10         1.0701         1.0593         1.0483         1.0376         1.0267         1.0160         1.0052         0.9945           15         1.0774         1.0661         1.0566         1.0464         1.0360         1.0251         1.0106           30         1.0845         1.0744         1.0643         1.0544         1.0446         1.0394         1.0251         1.0163           30         1.0846         1.0779         1.0680         1.0544         1.0446         1.0349         1.0251         1.0431         1.0379<		25	1.0665	1.0568	1.0470	1.0375	1.0278	1.0185	1.0089	0.9995	0.989		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		30	1.0697	1.0601	1.0506	1.0412	1.0317	1.0225	1.0132	1.0039	0.994		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		35	1.0728	1.0634	1.0540	1.0447	1.0355	1.0265	1.0173	1.0083	0.999		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		40	1.0759	1.0666	1.0573	1.0482	1.0392	1.0303	1.0213	1.0125	1.003		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		45	1.0788	1.0697	1.0606	1.0517	1.0428	1.0339	1.0250	1.0164	1.007		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		50	1.0818	1.0727	1.0637	1.0548	1.0461	1.0374	1.0287	1.0203	1.011		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		55	1.0040	1.0737	1.0000	1.0361	1.0495	1.0410	1.0323	1.0241	1.015		
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	3648	10	1.0874	1.0783	1.0099	1.0013	1.0528	1.0445	1.0359	0.0015	1.019		
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	3040	15	1.0701	1.0535	1.0405	1.0370	1.0207	1.0100	1.0052	1 0002	0.983		
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		20	1.0730	1.0032	1.0520	1.0420	1 0360	1 0259	1.0156	1.0055	0.995		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		25	1.0810	1.0708	1.0605	1.0504	1.0404	1.0304	1.0205	1.0106	1.000		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		30	1.0845	1.0744	1.0643	1.0544	1.0446	1.0349	1.0251	1.0153	1.005		
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		35	1.0877	1.0779	1.0680	1.0583	1.0486	1.0391	1.0295	1.0201	1.010		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		40	1.0910	1.0813	1.0715	1.0620	1.0526	1.0431	1.0338	1.0245	1.015		
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		45	1.0941	1.0846	1.0750	1.0657	1.0563	1.0470	1.0379	1.0288	1.019		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		50	1.0972	1.0878	1.0783	1.0691	1.0600	1.0508	1.0419	1.0330	1.023		
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		55	1.1003	1.0910	1.0816	1.0725	1.0635	1.0546	1.0457	1.0369	1.028		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		60	1.1032	1.0939	1.0849	1.0759	1.0670	1.0581	1.0495	1.0408	1.032		
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	.5702	10	1.1172	1.1041	1.0910	1.0780	1.0649	1.0516	1.0386	1.0250	1.011		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		15	1.1221	1.1094	1.0966	1.0838	1.0712	1.0585	1.0458	1.0331	1.020		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		20	1.1267	1.1144	1.1019	1.0897	1.0772	1.0649	1.0526	1.0401	1.027		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		25	1.1313	1.1191	1.1069	1.0949	1.0829	1.0710	1.0590	1.0471	1.035		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		30	1.1355	1.1236	1.1118	1.1001	1.0884	1.0767	1.0652	1.0534	1.041		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		35	1.1397	1.1280	1.1164	1.1050	1.0935	1.0822	1.0707	1.0595	1.048		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		40	1.1438	1.1324	1.1208	1.1097	1.0986	1.0874	1.0763	1.0654	1.054		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		45	1.14//	1.1364	1.1252	1.1143	1.1034	1.0924	1.0816	1.0708	1.060		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		50	1.1515	1.1405	1.1293	1.1187	1.1079	1.0972	1.0866	1.0761	1.065		
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		55 60	1.1552	1.1443	1.1334	1.1229	1.1123	1.1021	1.0915	1.0813	1.070		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	6031	10	1.1000 1.1500	1.1402 1 1977	1.13/3	1.12/1	1.1100	1.1003	1.0901	1.0000	1.076		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0991	10	1.1328	1.13//	1.1221	1.1074	1.0920	1.0700	1.0009	1.0430	1.028		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		20	1.1389	1.1444	1.1287	1.1149	1.1003	1.0804	1.0700	1.0333	1.040		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		25	1.1040	1.1505	1.1302	1.1222	1.10/8	1.0930	1.0793	1.0049	1.050		
35       1.1734       1.1015       1.1455       1.1551       1.1216       1.0044       1.0949       1.0010         35       1.1804       1.1673       1.1542       1.1411       1.1281       1.1151       1.1021       1.0893         40       1.1853       1.1724       1.1595       1.1469       1.1342       1.1216       1.1089       1.0964         45       1.1900       1.1774       1.1649       1.1524       1.1400       1.1276       1.1152       1.1031         50       1.1946       1.1822       1.1698       1.1576       1.1455       1.1335       1.1214       1.1096         55       1.1900       1.1774       1.1698       1.1576       1.1455       1.1335       1.1214       1.1096		20 20	1.1754	1.1302	1.1420	1.1207	1.1130	1.1013	1.08/4	1.0733	1.039		
55         1.1004         1.1075         1.1342         1.1411         1.1201         1.1151         1.1021         1.0093           40         1.1853         1.1724         1.1595         1.1469         1.1342         1.1216         1.1089         1.0964           45         1.1900         1.1774         1.1649         1.1524         1.1400         1.1276         1.1152         1.1031           50         1.1946         1.1822         1.1698         1.1576         1.1455         1.1335         1.1214         1.1096           55         1.1966         1.1827         1.1276         1.1276         1.1277         1.1477		30	1.1734	1.1019	1.1400	1.1331	1.1210	1.1004	1.0949	1.0010	1.008		
45         1.1900         1.1774         1.1649         1.1524         1.1400         1.1210         1.1005         1.0904           45         1.1900         1.1774         1.1649         1.1524         1.1400         1.1276         1.1152         1.1031           50         1.1946         1.1822         1.1698         1.1576         1.1455         1.1335         1.1214         1.1096           55         1.1900         1.1967         1.1746         1.1570         1.1305         1.1214         1.1096		40	1 1 9 5 9	1 1794	1.1542	1 1/60	1.1201	1 1916	1 1021	1.0055	1 0 8 2		
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		40	1 1000	1.1724	1.1355	1.1405	1.1342	1 1976	1 1159	1 10304	1 000		
55 1.1000 1.1068 1.1070 1.1770 1.1700 1.1700 1.1010 1.1217 1.1070		50	1 19/6	1 1 1 8 9 9	1 1608	1 1576	1 1 4 5 5	1 1 2 2 5	1 1914	1 1006	1 007		
JJ LIMMU LIMD7 LI740 LID77 LIMMU LIMMU LIMMU LI777 LIT56		55	1,1990	1.1867	1 1746	1 1697	1 1509	1,1391	1.1279	1,1156	1 10/		
60 1/2032 1/1012 1/705 1/677 1/560 1/444 1/1220 1/1916		60	1 2039	1 1019	1 1795	1 1677	1 1560	1 14/1	1 1320	1 1916	1 1 1 1 1		

Table	1 (	(Continue	ed)
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		$ ho/(g\cdot cm^{-3})$ at the following values of $T/K$								
X	<i>p</i> /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
0.0000	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	1.2002	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	1.1092	1.14/3	1.1245	1.1019
	30	1.2809	1.2004	1.2462	1.2255	1.2048	1.1842	1.1033	1.1420	1.1209
	30	1.2902	1.2703	1.2009	1.2371	1.2173	1.1970	1.1770	1.1377	1.1577
	40	1.3031	1.2000	1.2071	1.2479	1.2290	1.2100	1.1910	1.1720	1.1329
	45	1.3134	1.2.949	1.2700	1.2301	1.2.397	1.2214	1.2032	1.1040	1.1007
	55	1.3214	1.3032	1 2030	1.2070	1.2400	1.2.522	1.2145	1.1303	1.1752
	60	1 3360	1 3190	1 3020	1 2852	1 2683	1 2516	1 23/9	1 2185	1 2022
1	5	1 2478	1 2129	1 1759	1 1362	1.2003	1 0446	0.9888	0.9207	1.2022
1	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,<sup>9</sup> 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.<sup>7</sup> In the work concerning the HFC-134a + triethylene glycol dimethyl ether (TriEGDME) mixtures<sup>7</sup> 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  $\rho T$ 



**Figure 1.** General view of the investigated  $p\rho T$  surface of pure HFC-134a in a  $\rho T$  diagram:  $\diamond$ , saturation curve;  $\bigcirc$ , measured densities, our previous work;<sup>7</sup>  $\blacksquare$ , 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 ( $\bullet$ ), 0.9290 ( $\blacktriangle$ ), 0.8727 ( $\diamond$ ), 0.7288 (+), 0.6931 (×), 0.5702 ( $\triangle$ ), 0.3648 ( $\bigcirc$ ), 0.2896 ( $\Box$ ), 0.1114 (\*), and 0.0 ( $\blacksquare$ ). (-) Polynomial fitting for guiding the eye.

diagram. These experimental values could be used to validate the new equations of state<sup>13,14</sup> 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.<sup>15</sup> and by Pöhler et al.<sup>16</sup> for mixtures containing carbon dioxide and by Cavestri and Schafer<sup>17</sup> for R-410A with four polyolester lubricants.

The excess molar volumes,  $V_{\rm m}^{\rm E}$ , at each pressure p and temperature *T*, can be calculated from our experimental density values using the relation

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

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



**Figure 3.** Pressure (*p*) and refrigerant molar fraction (*x*) dependencies of (a, left) the experimental excess volumes  $V_m^{\text{E}}$  (cm<sup>3</sup>·mol<sup>-1</sup>) and (b, right) the  $\rho V_m^{\text{E}}/M$  (%) at 363.15 K:  $\blacksquare$ , 10 MPa;  $\Box$ , 15 MPa;  $\blacklozenge$ , 20 MPa;  $\bigcirc$ , 25 MPa;  $\blacklozenge$ , 30 MPa;  $\diamondsuit$ , 35 MPa;  $\blacktriangle$ , 40 MPa;  $\triangle$ , 45 MPa;  $\times$ , 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;  $\diamond$ , 313.15 K;  $\Box$ , 323.15 K;  $\circ$ , 333.15 K; △, 343.15 K;  $\blacksquare$ , 353.15 K;  $\bullet$ , 363.15 K;  $\blacktriangle$ , 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<sup>18</sup> 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.<sup>19,20</sup> 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.<sup>10</sup>

#### 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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