Volumetric Properties of Monoethylene Glycol Dimethyl Ether and Diethylene Glycol Dimethyl Ether up to 60 MPa

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In the present article, we report new density data for monoethylene glycol dimethyl ether (MEGDME) and for diethylene glycol dimethyl ether (DEGDME) at several temperatures between 293.15 K and 353.15 K and up to 60 MPa. A vibrating tube densimeter with an uncertainty of $\pm 1 \times 10^{-4}$ g·cm^{-3} was used for density measurements. The experimental density data (182 points) were fitted to the Tait-like equation with standard deviations of less than 2×10^{-4} g·cm^{-3}. Using these correlations, the isobaric thermal expansivity, α_p, the isothermal compressibility, κ_T, and the internal pressure, π, have been obtained for MEGDME and DEGDME. To analyze the trend of these properties with the chain length of polyalkylene glycol dimethyl ethers, their pressure and temperature dependences have been compared with the results found before for triethylene glycol dimethyl ether and for tetraethylene glycol dimethyl ether. The ability of different cubic equations of state to predict the volumetric properties of MEGDME and DEGDME was tested, obtaining the best results with the Peng−Robinson equation of state.

Introduction

Many studies on the polyalkylene glycol dimethyl ethers, CH$_3$O−((CH$_2$)$_2$O)$_n$−CH$_3$, have been carried out in the past years because these fluids are important industrial solvents.[1] Furthermore, some polyether compounds have been proposed as good candidates for absorbents for several refrigerants in absorption refrigeration systems.[2−4] Thus, the R22 + diethylene glycol dimethyl ether pair has been studied by Takeshita et al.[5,6] as the prime working fluid for residential gas-fired absorption heat pumps. For these reasons, the study of thermophysical properties of polyalkylene glycol dimethyl ethers is important and needed in engineering applications. However, the database for thermophysical properties of these compounds, at present, is scarce and limited to narrow temperature and pressure intervals. Most measurements have been made at atmospheric pressure and at temperatures in the range from 283.15 to 323.15 K.

However, there are some papers concerning measurements of volumetric properties for polyalkylene glycol dimethyl ethers at high pressure. Thus, Conesa et al.[7] have published density values at 1 MPa, from 283.15 K to 423.15 K, for monoethylene glycol dimethyl ether (MEGDME), diethylene glycol dimethyl ether (DEGDME), triethylene glycol dimethyl ether (TriEGDME), tetraethylene glycol dimethyl ether (TEGDME), pentaethylene glycol dimethyl ether, and polyethylene glycol 250 dimethyl ether. Density values of MEGDME have been published by Senger[8] between 288.0 and 473.0 K and up to 37 MPa and also by Sharipov and Bairamova[9] between 298.15 and 328.15 K and up to 202 MPa. These last authors have also reported densities of DEGDME in the same pressure and temperature ranges. The compatibility between the density measurements carried out by Senger and by Sharipov and

Bairamova has been analyzed by Cibulka et al. in a review paper,[10] finding that the deviations between the two data sets available for MEGDME[8,9] exceed experimental uncertainties claimed by the authors (3×10^{-4} g·cm^{-3} for Senger and 4×10^{-4} g·cm^{-3} for Sharipov and Bairamova) in the common temperature range. In this sense, additional experimental density values at high pressure with high accuracy are needed. For TEGDME, Svejda et al.[11] have studied the volumetric behavior at 293.15 K between 1 and 10 MPa. All these values[8,9,11] have been analyzed by Cibulka et al. in a review paper,[10] in which a $\rho p T$ Tait correlation and an analysis of the compatibility between the density measurements carried out by different authors are given. Finally, in our previous works[12−15] the density values between 283.15 and 373.15 K and at pressures up to 60 MPa have been reported for TriEGDME and TEGDME.

To complete these experimental[12−15] and theoretical studies[16] of some physical properties of polyethylene glycol dimethyl ethers, in this paper we report new density data (182 experimental points) of MEGDME and DEGDME at temperatures from 293.15 to 353.15 K and at pressures up to 60 MPa. These values have been used to test the prediction ability of three cubic equations of state.

Experimental Section

Materials. MEGDME (molar mass, 90.122 g·mol^{-1}) and DEGDME (molar mass, 134.175 g·mol^{-1}) were obtained from Aldrich with a purity of 99.9% and 99.5%, respectively. These chemicals were subjected to no further purification.

Measurement Technique. Densities as a function of pressure and temperature were determined using an Anton Paar DMA 60/512P vibrating tube densimeter. All the devices used for both the filling of the measuring cell and the measurements are shown in Figure 1. First, the vacuum system is connected to valve V$_1$, and then the

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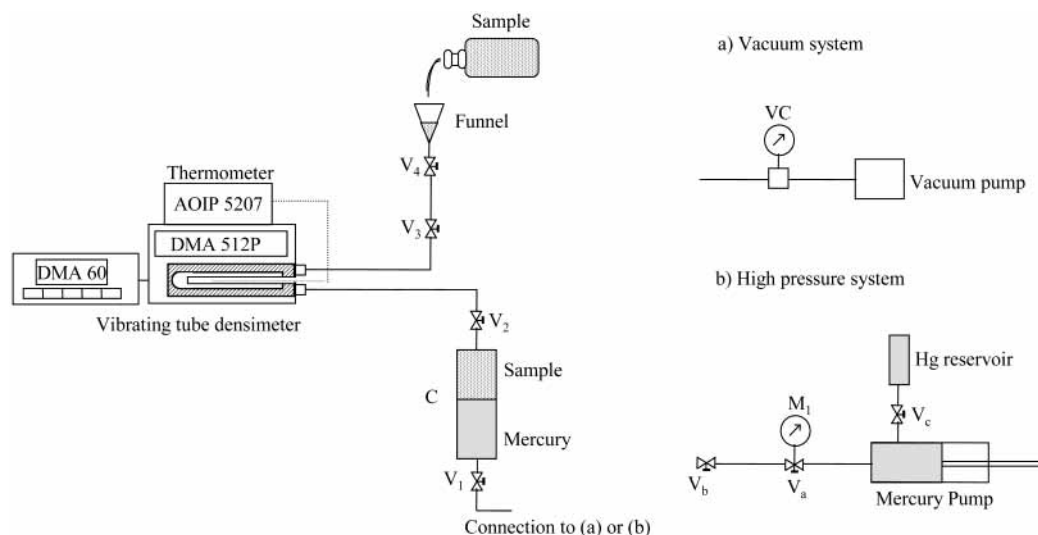‡ Université de Pau et des Pays de l'Adour.

Figure 1. Schematic diagram of the $p\rho T$ apparatus: high-pressure cell (C), vacuometer (VC), and manometer (M_1).

circuit between V_1 and V_4 (containing the high-pressure cell) is evacuated. Once the air is purged between the funnel and V_4, the measurement circuit is filled with the sample under a vacuum. Then, the pressure system is connected to V_1 and the mercury is introduced in the measurement circuit. The capacity between the V_2 and V_3 valves is close to 12 cm³, and the mercury/sample interface is always inside the cell (30 cm³). Finally, the sample is pressurized by using the mercury pump, and as soon as the thermal and mechanical equilibria are reached the data acquisition is performed.

The temperature of the vibrating tube is controlled with a thermoregulated liquid bath (Julabo Paratherm). 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 a Hottinger Baldwin Messtechnik (HBM) manometer with an uncertainty of ±0.05 MPa. The calibration parameters of the densimeter cell were determined using the best of the two calibration methods proposed by Lagourette et al.[17] This method is based on water as a reference substance and also requires the knowledge of the periods of the cell under a vacuum as a function of temperature. For water, the density values reported by Kell and Whalley[18] with an accuracy of 10^{-5} g·cm⁻³ have been used. This method has been employed in previous papers.[13,19] Taking into account the temperature, pressure, and water density accuracies, the total uncertainty in density is 1×10^{-4} g·cm⁻³. Notice here that the uncertainty is better than that of Senger[8] (3 $\times$ 10⁻⁴ g·cm⁻³) or Sharipov and Bairamova[9] (4 $\times$ 10⁻⁴ g·cm⁻³).

Results

The measured densities of MEGDME and DEGDME are reported in Table 1 along isotherms between 293.15 K and 353.15 K at pressures up to 60 MPa. To correlate our values over all of the temperature and pressure interval, the following Tait-like equation has been used as in previous works:[12,13]

$$\rho(T, p) = \frac{\rho(T, 0.1 \text{ MPa})}{1 - C \ln\left(\dfrac{B(T) + p}{B(T) + 0.1 \text{ MPa}}\right)} \quad (1)$$

where $\rho(T, 0.1 \text{ MPa})$ is the temperature dependence of the

Table 1. Experimental Densities, ρ, at Various Temperatures T and Pressures p

	ρ/g·cm⁻³ at the following T/K						
p/MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15
				MEGDME			
0.1	0.8667	0.8555	0.8444	0.8331	0.8216	0.8099	0.7976
5	0.8711	0.8602	0.8494	0.8385	0.8275	0.8163	0.8048
10	0.8753	0.8648	0.8544	0.8439	0.8332	0.8225	0.8114
15	0.8795	0.8692	0.8591	0.8487	0.8387	0.8283	0.8176
20	0.8832	0.8734	0.8635	0.8534	0.8436	0.8338	0.8232
25	0.8872	0.8774	0.8677	0.8580	0.8484	0.8387	0.8287
30	0.8909	0.8813	0.8717	0.8623	0.8531	0.8435	0.8338
35	0.8944	0.8851	0.8757	0.8664	0.8574	0.8480	0.8386
40	0.8978	0.8885	0.8794	0.8702	0.8617	0.8526	0.8431
45	0.9011	0.8919	0.8831	0.8742	0.8656	0.8566	0.8478
50	0.9043	0.8955	0.8867	0.8778	0.8690	0.8606	0.8520
55	0.9075	0.8986	0.8900	0.8814	0.8733	0.8645	0.8560
60	0.9104	0.9017	0.8933	0.8850	0.8767	0.8683	0.8598
				DEGDME			
0.1	0.9435	0.9337	0.9237	0.9137	0.9036	0.8936	0.8833
5	0.9473	0.9376	0.9279	0.9182	0.9083	0.8985	0.8887
10	0.9509	0.9411	0.9318	0.9227	0.9128	0.9032	0.8939
15	0.9544	0.9450	0.9357	0.9264	0.9173	0.9078	0.8987
20	0.9577	0.9484	0.9394	0.9305	0.9210	0.9123	0.9032
25	0.9609	0.9518	0.9426	0.9342	0.9253	0.9162	0.9076
30	0.9640	0.9552	0.9465	0.9376	0.9289	0.9205	0.9116
35	0.9670	0.9582	0.9497	0.9410	0.9326	0.9242	0.9155
40	0.9698	0.9611	0.9528	0.9444	0.9364	0.9280	0.9195
45	0.9728	0.9640	0.9561	0.9476	0.9398	0.9314	0.9231
50	0.9754	0.9673	0.9588	0.9510	0.9429	0.9348	0.9270
55	0.9782	0.9701	0.9619	0.9539	0.9460	0.9382	0.9304
60	0.9809	0.9732	0.9649	0.9567	0.9491	0.9414	0.9336

density at 0.1 MPa for which we have chosen the following function:

$$\rho(T, 0.1 \text{ MPa}) = \rho_0(T) = A_0 + A_1 T + A_2 T^2 + A_3 T^3 \quad (2)$$

In eq 1, parameter C was assumed to be temperature independent, and for $B(T)$, we have used the following polynomial function:

$$B(T) = B_0 + B_1 T + B_2 T^2 \quad (3)$$

To compare the experimental density values with those obtained with the Tait correlation and with those predicted with several equations of state, we have used the absolute average percentual deviation, AAD, the maximum percen-

Table 2. Obtained Parameters and Deviations for the Density Correlation by Using Equation 1

	MEGDME	DEGDME
Coefficients		
A_0/g·cm^{-3}	1.579940	1.239297
A_1/g·cm^{-3}·K^{-1}	-5.0117×10^{-3}	-1.1048×10^{-3}
A_2/g·cm^{-3}·K^{-2}	1.3080×10^{-5}	5.8475×10^{-7}
A_3/g·cm^{-3}·K^{-3}	-1.4616×10^{-8}	-8.7939×10^{-10}
C	0.088502	0.083763
B_0/MPa	403.764	462.568
B_1/MPa·K^{-1}	-1.50558	-1.71578
B_2/MPa·K^{-2}	0.001404	0.001681
Deviation		
σ/g·cm^{-3}	1×10^{-4}	1.4×10^{-4}
AAD/%	1×10^{-2}	1×10^{-2}
DMAX/%	4×10^{-2}	4×10^{-2}
Bias/%	1×10^{-4}	-1×10^{-3}

tual deviation (DMAX), and the average percentual deviation (Bias), which are defined as follows:

$$\text{AAD} = \frac{100}{N} \sum_{i=1}^{N} \left| \frac{\rho_i^{\text{exp}} - \rho_i^{\text{teo}}}{\rho_i^{\text{exp}}} \right| \tag{4}$$

$$\text{DMAX} = \text{Max}\left(100 \left| \frac{\rho_i^{\text{exp}} - \rho_i^{\text{teo}}}{\rho_i^{\text{exp}}} \right| \right) \tag{5}$$

$$\text{Bias} = \frac{100}{N} \sum_{i=1}^{N} \frac{\rho_i^{\text{exp}} - \rho_i^{\text{teo}}}{\rho_i^{\text{exp}}} \tag{6}$$

The Tait correlation parameters, AAD, DMAX, Bias, and standard deviation, σ, obtained with this correlation are shown in Table 2 for both polyethers. Notice that the standard deviation is of the same order as the experimental uncertainty. Equations 1−3 makes it possible to estimate the density at any pT conditions in order to compare the data with those of other authors.

We have compared our experimental values with those determined by Senger[8] and by Sharipov and Bairamova.[9] For MEGDME, an AAD of 0.10% was found between the experimental values of this work and the literature values

of Senger, between 293.4 K and 323.15 K and up to 35 MPa. The data set of Sharipov and Bairamova for DEGDME agrees with our experimental values within an AAD of 0.06% over the temperature range of 293.15−328.15 K and up to 60 MPa. For DEGDME, the AAD with the data set of Sharipov and Bairamova in the same temperature and pressure interval is 0.02%. The obtained deviations between our values for MEGDME and DEGDME and those of Senger and Sharipov and Bairamova are plotted in Figure 2. Our density values for MEGDME are between both data sets.

The Derived Thermodynamic Properties

The isobaric thermal expansivity, $\alpha_p = -(1/\rho)(\partial\rho/\partial T)_p$, the isothermal compressibility, $\kappa_T = (1/\rho)(\partial\rho/\partial p)_T$, and the internal pressure, $\pi = T(\alpha_p/\kappa_T) - p$, for MEGDME and DEGDME were calculated from the Tait-like correlations $\rho(T, p)$ using the parameters of Table 2. The estimated uncertainties are $\pm 0.2 \times 10^{-4}$ K^{-1}, $\pm 0.05 \times 10^{-4}$ MPa^{-1}, and 1%, respectively. Tables 3 and 4 summarize the obtained values for α amd κ as a function of temperature and pressure.

In Tables 3 and 4, it can be seen that the isobaric thermal expansivities and the isothermal compressibilities for MEGDME are higher than those obtained for DEGDME. These results together with those of TriEGDME and TEGDME[13] show that both properties decrease when the length of the polyether $(CH_3O-((CH_2)_2O)_n-CH_3)$ rises. To see this fact, α_p and κ_T have been plotted in Figure 3 versus pressure for the 313.15 K and 333.15 K isotherms for the four studied polyethers, including our previous values[13] for TriEGDME and TEGDME. The isobaric thermal expansivity has an intersection point of isotherms, as shown in Figure 4. The pressure at which α_p is independent of temperature (crossing point) increases when the chain length of polyethylene glycol dimethyl ether rises. In the studied pressure interval, we have clearly observed the intersection point for TEGDME,[13] whereas this point is less apparent for MEGDME.

The internal pressure is sensitive to changes of temperature and external pressure as shown in Figure 5, where the internal pressure, π, is plotted against the molar
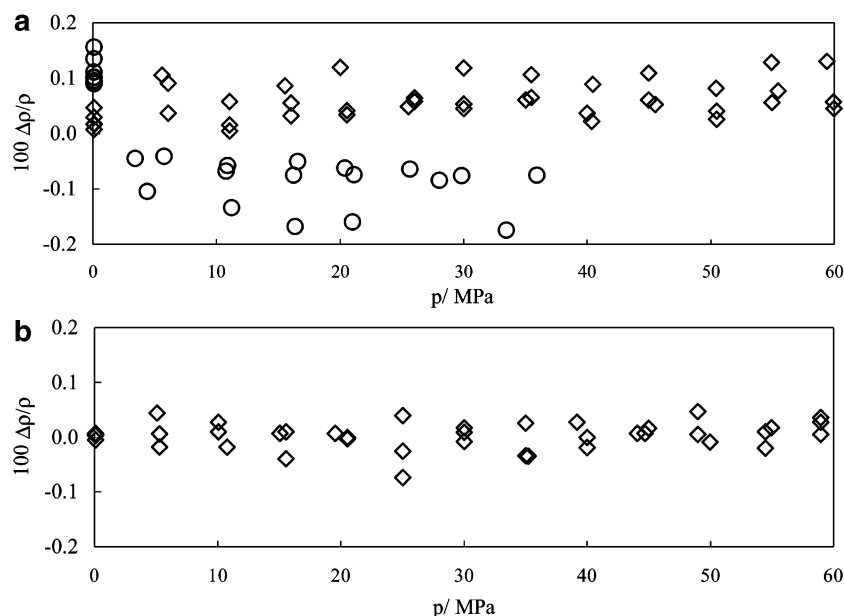

Figure 2. Comparison between the experimental and literature densities, $100(\rho_{\text{lit}} - \rho_{\text{exp}})/\rho_{\text{exp}}$, of MEGDME (a) and DEGDME (b): (○) Sharipov and Bairamova (ref 9); (◇) Senger (ref 8).

Table 3. Isobaric Thermal Expansivity, α_p, at Different Temperatures, T, and Pressures, p

	$\alpha_p/(10^{-4}\ K^{-1})$ at the following T/K				
p/MPa	303.15	313.15	323.15	333.15	343.15
		MEGDME			
0.1	13.0	13.3	13.6	14.2	14.8
5	12.5	12.7	13.1	13.5	14.0
10	12.1	12.3	12.5	12.9	13.3
15	11.7	11.9	12.1	12.4	12.7
20	11.4	11.5	11.6	11.9	12.2
25	11.1	11.1	11.3	11.5	11.8
30	10.8	10.8	10.9	11.1	11.4
35	10.5	10.5	10.6	10.8	11.0
40	10.3	10.3	10.3	10.5	10.7
45	10.1	10.0	10.1	10.2	10.4
50	9.9	9.8	9.8	9.9	10.1
55	9.7	9.6	9.6	9.7	9.8
60	9.5	9.4	9.4	9.5	9.6
		DEGDME			
0.1	10.6	10.8	11.0	11.2	11.4
5	10.3	10.5	10.6	10.8	10.9
10	10.1	10.2	10.3	10.4	10.6
15	9.8	9.9	10.0	10.1	10.2
20	9.6	9.7	9.7	9.8	9.9
25	9.4	9.4	9.5	9.6	9.6
30	9.2	9.2	9.3	9.3	9.4
35	9.0	9.0	9.1	9.1	9.1
40	8.8	8.9	8.9	8.9	8.9
45	8.7	8.7	8.7	8.7	8.7
50	8.5	8.5	8.5	8.5	8.5
55	8.4	8.4	8.4	8.4	8.4
60	8.2	8.2	8.2	8.2	8.2

Table 4. Isothermal Compressibility, κ_T, versus Temperature, T, and Pressure, p

	$\kappa_T/(10^{-4}\ MPa^{-1})$ at the following T/K						
p/MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15
			MEGDME				
5	10.10	10.93	11.87	12.93	13.19	15.52	17.10
10	9.60	10.35	11.19	12.13	12.37	14.38	15.74
15	9.16	9.84	10.59	11.43	11.64	13.41	14.58
20	8.75	9.37	10.06	10.81	11.00	12.57	13.59
25	8.38	8.95	9.57	10.26	10.44	11.83	12.73
30	8.04	8.57	9.14	9.76	9.93	11.17	11.98
35	7.73	8.22	8.74	9.31	9.46	10.59	11.32
40	7.45	7.90	8.38	8.90	9.05	10.07	10.73
45	7.18	7.60	8.05	8.53	8.67	9.60	10.20
50	6.94	7.33	7.74	8.19	8.32	9.18	9.72
55	6.71	7.07	7.46	7.88	8.00	8.79	9.29
			DEGDME				
5	7.71	8.25	8.85	9.49	10.20	10.98	11.82
10	7.40	7.90	8.44	9.03	9.67	10.36	11.11
15	7.12	7.58	8.07	8.61	9.19	9.81	10.48
20	6.85	7.28	7.74	8.23	8.76	9.32	9.93
25	6.61	7.01	7.43	7.88	8.37	8.88	9.43
30	6.38	6.75	7.15	7.56	8.01	8.48	8.98
35	6.17	6.52	6.88	7.27	7.68	8.12	8.57
40	5.98	6.30	6.64	7.00	7.38	7.79	8.21
45	5.79	6.10	6.42	6.75	7.11	7.48	7.87
50	5.62	5.91	6.21	6.52	6.85	7.20	7.56
55	5.46	5.73	6.01	6.31	6.62	6.94	7.27

volume for MEGDME and DEGDME. This derived property, at constant volume, decreases with temperature for the two polyethers analyzed in this work. The same behavior has been found before for TriEGDME and TEGDME.[13] The decrease in the internal pressure when the molar volume diminishes along an isobar is more important than the corresponding decrease in π along an isotherm. This tendency indicates that these compounds behave like an associated or structured fluid.[20] This is in agreement with the previous investigations[21-25] about interactions in polyethers which concluded that they can be considered as hydrogen bonds.

The polyether molecules are polar, with their dipolar momentum increasing as does the length of the chain, and have less conformational flexibility than other molecules, such as normal alkanes, due to the appearance of the anomeric effect molecules which have two atoms of oxygen separated by one or two methylene groups, $-O-CH_2-CH_2-O-$. Kimura and Fujishiro[26] have measured the dipole moments of several polyethers, obtaining 1.59 D, 1.92 D, 2.16 D, and 2.44 D for MEGDME, DEGDME, TriEGDME, and TEGDME, respectively. We have observed that the internal pressure increases with the chain length; this means that this property increases when the dipolar momentum rises, whereas the sequence is the opposite for α_p and κ_T. As the length of the polyether increases, the interaction number grows, and the liquids containing these molecules become more compact, which means less expansible and less compressible. Furthermore, taking into account that $\pi = (\partial U/\partial V)_{T,p}$ it can be concluded that the volume dependence of the internal energy (U) becomes more important when the polyether length increases.

On the other hand, the capabilities of the equations of state of Soave−Redlich−Kwong (SRK), Peng−Robinson (PR), Patel−Teja (PT), and SRK with volume translation of Peneloux (SRK-VT) for density prediction of these compounds have been checked. The values of the critical
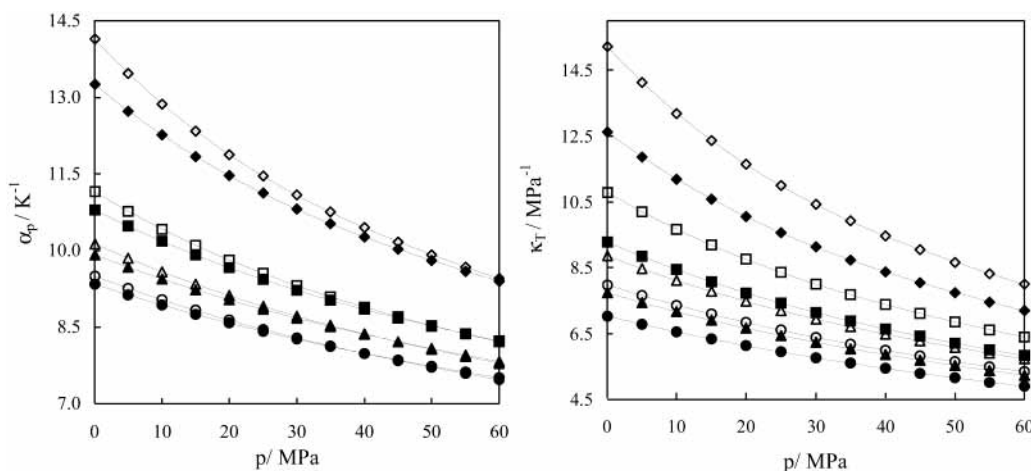

Figure 3. Temperature and pressure dependence of the isobaric thermal expansivity, α_p, and of the isothermal compressibility, κ_T: (◆, ■) values from this work for MEGDME and DEGDME; (▲, ●) values for TriEGDME and TEGDME reported by Comuñas et al. (ref 13). Dark and white symbols are the values at 313.15 K and 333.15 K, respectively.
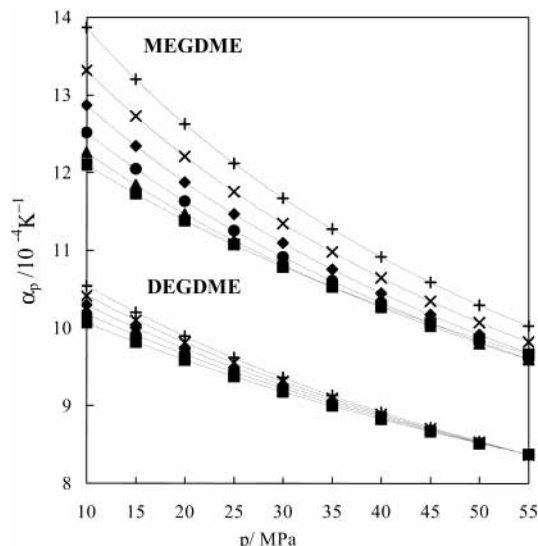
Figure 4. Variation of the isobaric thermal expansion coefficient, α_p, with pressure for MEGDME and DEGDME at different temperatures: ◆, 293.15 K; ■ 303.15 K; ▲, 313.15 K; ●, 323.15 K; ×, 333.15 K; +, 343.15 K.

pressure, p_c, critical temperature, T_c, and acentric factor, ω, needed to apply the equations of state have been obtained from the literature. Quadri and Kudchadker[27] have reported the experimental critical temperature and pressure for MEGDME (T_c= 539.2 K and p_c = 3.86 MPa, respectively). For DEGDME, we have used the experimental critical temperature reported by Morton et al.,[28] T_c= 617 K. The acentric factor values (ω = 0.455 for MEGDME and 0.595 for DEGDME, respectively) have been obtained from the experimental vapor pressure values published by Stull.[29] The critical pressure for DEGDME has been estimated by using the group contribution methods of Joback[30] (p_c = 2.94 MPa), since experimental p_c data are not available. We have used this group contribution method because this model better reproduces the experimental critical pressure of the other polyether (MEGDME).

The results obtained for the density prediction with several equations of state are presented in Table 5. For MEGDME, the best results (AAD = 0.7%, DMAX = 1.0%, and Bias = −0.5%) are obtained by using the PR equation of state (EOS), and for DEGDME the PT EOS (AAD =

Table 5. Obtained Results for Density Prediction with Several Equations of State

liquid	AAD/%	Bias/%	DMAX/%
	SRK		
MEGDME	10.3	10.3	11.0
DEGDME	14.6	14.6	15.9
	SRK-VT		
MEGDME	9.0	−9.0	10.1
DEGDME	12.0	−12.0	13.2
	PR		
MEGDME	0.7	−0.5	1.0
DEGDME	4.5	4.5	6.2
	PT		
MEGDME	4.8	−4.8	6.1
DEGDME	3.3	−3.3	5.3

3.3%, DMAX = 5.3%, and Bias = −3.3%) yields the best results. The results found for DEGDME with the PT and PR equations of state are very similar. Contrarily, for MEGDME the AAD for PT (4.8%) is higher than that found with PR (0.7%). The density values predicted for both polyethers with the PT EOS are higher than the experimental data, whereas with the PR EOS the theoretical values are higher than the experimental ones for MEGDME and lower for DEGDME. Similar results have been found in previous work[16] for TriEGDME and TEGDME. The highest AADs are obtained with the SRK EOS, which predicts density values that are always lower than the experimental data. With the SRK-VT EOS, the AADs are slightly lower than those found with SRK, and the theoretical values are always higher than the experimental ones.

Conclusions

The pressure and temperature dependences of the volumetric properties of MEGDME and DEGDME have been determined over wide temperature (293.15 to 353.15 K) and pressure (0.1 to 60 MPa) ranges. Using previous results on TriEGDME and TEGDME, it seems that the isobaric thermal expansivities and the isothermal compressibilities decrease when the length of the polyether rises. For the internal pressure, the sequence with the chain is the opposite. This means that when the length of polyethers rises the liquids become less expansible and less compressible but the volume dependence of the internal energy becomes bigger. It is also shown that the isobaric thermal
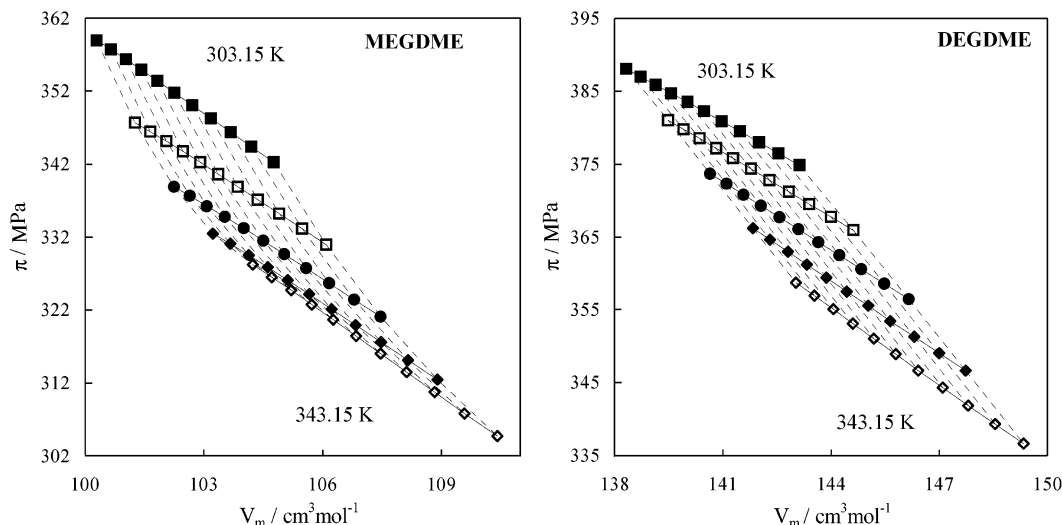

Figure 5. Variation of the internal pressure, π, with molar volume at different temperatures and pressures: ■, 303.15 K; □, 313.15 K; ●, 323.15 K; ◆, 333.15 K; ◇, 343.15 K; dashed lines, isobars from 5 to 55 MPa at 5 MPa intervals.

expansivity has a crossover point of isotherms, which occurs at a pressure higher for TEGDME than for MEGDME. The predictions of the density with the SRK, PR, and PT equations of state have been checked, and the best results are obtained with the PR EOS for MEGDME and with PT for DEGDME.

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