pρT Measurements and EoS Predictions of Glycol Ethers from (283.15 to 353.15) K at Pressures up to 25 MPa

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New experimental densities in the compressed liquid state are reported for diethylene glycol monobutyl ether and propylene glycol propyl ether over the temperature range (283.15 to 353.15) K and for pressures up to 25 MPa. Density values (144 experimental data points) have been measured with a high-pressure vibrating tube densimeter. These data were used to determine isothermal compressibilities, isobaric thermal expansivities, and internal pressures for these compounds. Furthermore, the predictions for the vapor pressures and for the densities of four glycol ethers with different equations of state (EoS): Soave−Redlich−Kwong, Peng−Robinson, Soave−Redlich−Kwong with volume translation, and Patel−Teja have been analyzed to test the reliability of these EoS.

Introduction

Equations of state are required for the calculations of thermodynamic properties needed in process engineering. For the development of accurate equations of state, a large amount of high-quality thermodynamic data is essential. Information about the pressure and temperature dependencies of such properties allows for thermodynamic modeling based on equations of state, statistical mechanics, and so forth to be checked. Among these properties, density values as a function of temperature and pressure are needed for the development of correlation or prediction models. Moreover, an accurate set of critical parameters and acentric factors for the pure compounds are crucial for several reliable equations of state. Because no single equation has been able to establish unrivaled supremacy, it is interesting to analyze the capability of the equations of state to predict the vapor pressure and the volumetric behavior at high pressure for different compounds.

In this work, the volumetric behavior under pressure of two glycol ethers—diethylene glycol monobutyl ether (DEGBE, 2-(2-butoxyethoxy) ethanol, CAS 112-34-5) and propylene glycol propyl ether (PGPE, 1-propoxy-2-propanol, CAS 1569-01-3)—that contain a hydroxyl group and several ether groups in the same molecule has been studied. These compounds are miscible in a wide range of solvents including water. Their primary use is as a component of liquid cleaners. Because of their fast evaporation rate and excellent ability to solubilize organic soils, their use has increased. The cleaning of exhaust air and gas streams from industrial production plants with the simultaneous recovery of useful materials is taking on increasing importance. It is not only required by legal regulations but also conserves resources by recycling useful materials. Thus, the use of glycol ethers as scrubbing liquids has been suggested because of their favorable properties.[1,2] Moreover, glycol ethers are widely used as detergents by means of forming stable homogeneous solutions with high fractions of water and oil.[3,4] In addition, lubricants, hydraulic fluids, greases, and so forth contain glycol ethers, and the compatibility of certain oil additives is improved and turbidity is corrected by the addition of glycol ether-type compounds.

pρT data at medium and high pressures are needed for the simulation of the behavior of this type of compounds under real operating conditions for different potential applications. It is important to point out that until now there have been no articles in the literature dealing with experimental data at high pressure of DEGBE or PGPE nor any theoretical studies concerning this type of glycol ethers.

In this context, considering the great importance of *pρ*T data with a view toward a complete thermodynamic description and continuing our previous work,[5] the present article reports experimental density values of diethylene glycol monobutyl ether and propylene glycol propyl ether in the temperature range from (283.15 to 353.15) K and at pressures up to 25 MPa. The experimental data were used to determine isothermal compressibilities, isobaric thermal expansivities, and internal pressures for these compounds. Therefore, additional information concerning pressure and temperature dependencies of these properties is also provided.

Furthermore, in this work predictions of the vapor pressures and the volumetric behavior of several glycol ethers (diethylene glycol monomethyl ether, DEGME, 2-(2-methoxyethoxy) ethanol, CAS 111-77-3; diethylene glycol monoethyl ether, DEGEE, 2-(2-ethoxyethoxy) ethanol, CAS 111-90-0, DEGBE and PGPE) have been determined to test the predicted goodness of four equations of state (EoS): Soave−Redlich−Kwong (SRK), Peng−Robinson (PR), Soave−Redlich−Kwong with volume translation (SRK-VT). and Patel−Teja (PT).

Experimental Section

Materials. Diethylene glycol monobutyl ether, $CH_3-(CH_2)_3-O-(CH_2)_2-O-(CH_2)_2-OH$ (Aldrich, puriss, mole

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Table 1. Experimental Densities, ρ, for Diethylene Glycol Monobutyl Ether (DEGBE) and Propylene Glycol Propyl Ether (PGPE) at Different Temperatures T and Pressures p

	T/K							
p/MPa	283.15	293.15	303.15	313.15	323.15	333.15	343.15	353.15
	ρ/g·cm^{-3}							
	DEGBE							
0.10	0.9605	0.9522	0.9438	0.9353	0.9266	0.9178	0.9097	0.9009
1.00	0.9610	0.9527	0.9443	0.9359	0.9272	0.9185	0.9104	0.9017
2.50	0.9619	0.9537	0.9453	0.9369	0.9283	0.9197	0.9116	0.9030
5.00	0.9634	0.9552	0.9469	0.9386	0.9301	0.9216	0.9136	0.9050
7.50	0.9649	0.9567	0.9485	0.9403	0.9318	0.9234	0.9155	0.9071
10.00	0.9663	0.9582	0.9501	0.9419	0.9335	0.9252	0.9174	0.9091
15.00	0.9691	0.9611	0.9531	0.9451	0.9369	0.9287	0.9211	0.9129
20.00	0.9718	0.9639	0.9561	0.9482	0.9401	0.9320	0.9246	0.9166
25.00	0.9744	0.9667	0.9590	0.9512	0.9432	0.9352	0.9280	0.9201
	PGPE							
0.10	0.8951	0.8860	0.8764	0.8669	0.8572	0.8471	0.8375	0.8273
1.00	0.8957	0.8866	0.8772	0.8676	0.8580	0.8479	0.8384	0.8283
2.50	0.8967	0.8877	0.8784	0.8689	0.8593	0.8493	0.8400	0.8299
5.00	0.8985	0.8896	0.8803	0.8710	0.8614	0.8517	0.8424	0.8326
7.50	0.9001	0.8913	0.8822	0.8730	0.8635	0.8539	0.8449	0.8352
10.00	0.9018	0.8931	0.8840	0.8750	0.8656	0.8561	0.8472	0.8377
15.00	0.9050	0.8965	0.8876	0.8788	0.8696	0.8603	0.8518	0.8425
20.00	0.9081	0.8998	0.8911	0.8824	0.8735	0.8643	0.8560	0.8470
25.00	0.9111	0.9029	0.8944	0.8859	0.8772	0.8681	0.8601	0.8513

Table 2. A_i, B_i, and C Coefficients and Standard Deviations s and s^* for Equations 1 and 2

	DEGBE	PGPE
A_0/g·cm^{-3}	1.18872	1.10477
$-A_1 \times 10^3$/g·cm^{-3}·K^{-1}	0.76834	0.55688
$-A_2 \times 10^6$/g·cm^{-3}·K^{-2}	0.1322	0.6479
$s \times 10^4$/g·cm^{-3}	2	2
B_0/MPa	393.2426	366.8335
$-B_1$/MPa·K^{-1}	1.2135	1.1682
$B_2 \times 10^3$/MPa·K^{-2}	0.9387	0.8623
C	0.078697	0.083607
$s^* \times 10^4$/g·cm^{-3}	1	1

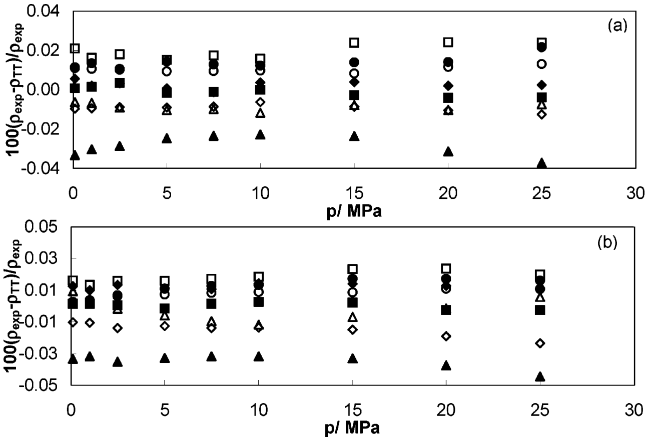

Figure 1. Relative deviation between the experimental density data and those calculated with Tammann−Tait (TT) for (a) DEGBE and (b) PGPE at different temperatures: ◇ 283.15 K; ◆, 293.15 K; ○, 303.15 K; ●, 313.15 K; △, 323.15 K; ▲, 333.15 K; □, 343.15 K; ■, 353.15 K.

fraction purity >0.99) and propylene glycol propyl ether, $CH_3-(CH_2)_2-O-CH_2-CH(CH_3)OH$ (Aldrich, puriss, mole fraction purity 0.985) were used without purification except for drying with Union Carbide 0.4 nm molecular sieves. These chemicals as well as water (purified using a Milli-Q Plus system, with a resistivity of 18.2 MΩ·cm at 298.15 K) were partially degassed before use in a Branson 2210 ultrasonic bath.

Experimental Method. The complete experimental assembly of the apparatus and the experimental procedure have been previously described in detail.[6,7] The method is based on the vibrating tube densimeter technique, and the heart of the equipment is a commercial Anton Paar DMA 512P vibrating tube cell. The application range of the DMA 512P cell is from (0 to 70) MPa and from (263.15 to 423.15) K. The temperature inside the cell block was measured with an Anton Paar CKT100 platinum resistance thermometer with an uncertainty of ±0.01 K. The temperature scale employed was ITS-90. The pressure was measured by means of an HBM-PE300 differential pressure gauge. This gauge was calibrated with an uncertainty of ±0.02 MPa.

Among the diverse ways[8] to convert the vibrating periods to densities, in this work the method developed by Lagourette et al.[9] was used. To apply this method, it is necessary to know only the vibrating periods of the evacuated tube in the experimental temperature range and the vibrating period of a reference fluid as a function of temperature and pressure. A classical reference fluid is water, for which precise density data are available as a function of T and p.[10] The total density uncertainty, $\pm 10^{-4}$ g·cm^{-3}, has been estimated using the law of propagation of uncertainty taking into account the accuracy of the temperature, the pressure, the period of oscillation measurements for water, vacuum, and the studied liquids (seven digits frequency counter), the water density accuracy, and the prepared vacuum quality.

Results and Discussion

Densities. Table 1 presents the experimental density measurements of DEGBE and PGPE along eight isotherms from (283.15 to 353.15) K at 10 K intervals and along nine isobars at pressures up to 25 MPa. For both fluids, the experimental $p\rho T$ values were fit with the following modified Tammann−Tait equation:[11]

$$\rho(T, p, C, B) = \frac{\rho(T, p_{\mathrm{ref}}(T))}{1 - C \ln\left(\dfrac{B(T) + p}{B(T) + p_{\mathrm{ref}}(T)}\right)} \tag{1}$$

where $\rho(T, p_{\mathrm{ref}}(T))$ is the temperature dependence of density at the reference pressure (in this case atmospheric pressure) and is given by

$$\rho(T, 0.1\ \mathrm{MPa}) = A_0 + A_1 T + A_2 T^2 \tag{2}$$

The A_i values were determined by smoothing the experimental densities at atmospheric pressure with a standard deviation, s, of 2×10^{-4} g·cm^{-3} for each compound. The denominator of eq 1 changes with both temperature and pressure. The parameter C was assumed to be temperature-independent, and for $B(T)$, the following polynomial function was used:

$$B(T) = B_0 + B_1 T + B_2 T^2 \tag{3}$$

The values of B_i and C for each glycol ether have been determined by fitting with eq 1 all of the experimental data at pressures other than atmospheric. The standard deviations, s^*, from the Tammann−Tait correlations are 1×10^{-4} g·cm^{-3} for both compounds. The set of fitting coefficient values (A_i, B_i, C) and the standard deviations s and s^* are listed in Table 2. Relative deviations between the experimental data and those calculated with the Tammann−Tait equation are presented in Figure 1 for both glycol ethers.

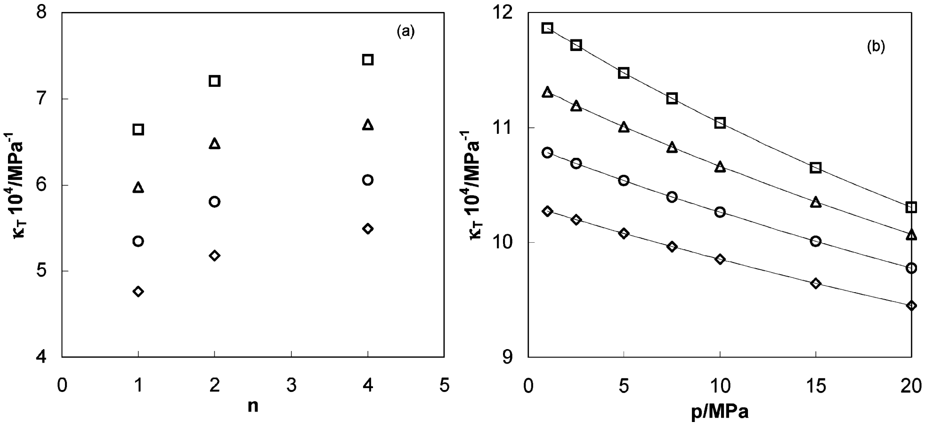

Figure 2. Isothermal compressibility, κ_T, for (a) diethylene glycol monoalkyl ethers, $CH_3-(CH_2)_{n-1}-C_4H_9O_3$, as function of n (DEGME,[5] $n = 1$; DEGEE,[5] $n = 2$; DEGBE,$^{\text{this work}}$ $n = 4$) at 20 MPa and (b) PGPE$^{\text{this work}}$ versus the pressure, p: ◇, 283.15 K; ○, 303.15 K; △, 323.15 K; □, 343.15 K.

The density values available in the literature for both fluids are scarce. To our knowledge, no density values at pressures other than atmospheric exist for these compounds. At atmospheric pressure and different temperatures, our experimental density values agree with the published data.[12−21] For comparison, when the literature density values and our density data are not at the same temperature, interpolated densities using eq 2 have been used. The average absolute deviation (AAD) between our experimental data and the literature values[12−20] at (293.15, 298.15, and 313.15) K is 0.04% for DEGBE, whereas for PGPE the AAD with the literature data[21] at (298.15, 308.15, and 318.15) K is 0.02%.

The density behavior is standard; ρ increases when the temperature decreases or the pressure increases, as shown in Table 1. The density data of DEGBE are lower than those of DEGME[5] or DEGEE,[5] decreasing with the size of the diethylene glycol monoalkyl ether molecule.

Derived Properties. The Tammann−Tait equation with the parameters presented in Table 2 was used to determine the effect of both temperature and pressure on different thermophysical properties. According to their definitions, the isothermal compressibility $\kappa_T = (1/\rho)(\partial\rho/\partial p)_T$, the isobaric thermal expansivity $\alpha_p = -(1/\rho)(\partial\rho/\partial T)_p$, and the internal pressure $\pi = T(\alpha_p/k_T) - p$, were determined. The estimated uncertainties of these properties are $\pm 0.03 \times 10^{-4}$ MPa^{-1}, $\pm 0.2 \times 10^{-4}$ K^{-1}, and ±1MPa, respectively, over the entire pressure and temperature ranges. The temperature dependence of the isothermal compressibilities was calculated from analytical differentiation of the Tammann−Tait equation with respect to pressure, which is certainly the most direct way to obtain reliable isothermal compressibility values.[22] In a similar way, isobaric thermal expansivity data can be determined by analytical calculation. In Figure 2a, the isothermal compressibility values for DEGBE have been plotted together with previous data for DEGME[5] and DEGEE.[5] This Figure shows the increase in the value of this property with the size of the diethylene glycol monoalkyl ether molecule and with the increase in temperature. The κ_T values obtained for PGPE are larger than those of the three diethylene glycol monoalkyl ethers, as seen in Figure 2a and b. The presence of two ether groups in the glycol ether DEGEE ($C_6H_{14}O_3$) versus one in PGPE ($C_6H_{14}O_2$) could explain why PGPE is more compressible than DEGEE. This fact has been also found for polyalkylene glycol dimethyl ethers.[23,24] However, the isothermal compressibility, κ_T, decreases when the pressure increases at constant temperature, about 15% and 11% at 323.15 K over the entire pressure interval for DEGBE and PGPE, respectively. The temperature and pressure trends for the isobaric thermal expansivity are the same as for κ_T, as can be observed in Table 3, which presents κ_T and α_p data for some isotherms.

Table 3. Isobaric Thermal Expansivity, α_p, and Isothermal Compressibility, κ_T, for DEGBE and PGPE at Several Temperatures T and Pressures p

	$\alpha_p \times 10^4$/K^{-1}			$\kappa_T \times 10^4$/MPa^{-1}		
p/MPa	293.15	323.15	343.15	293.15	323.15	343.15
			DEGBE			
1.00	8.9	9.2	9.4	10.52	11.31	11.87
2.50	8.8	9.1	9.3	10.44	11.19	11.72
5.00	8.7	9.0	9.2	10.31	11.01	11.48
7.50	8.6	8.9	9.0	10.18	10.83	11.25
10.00	8.5	8.8	8.9	10.06	10.66	11.04
15.00	8.4	8.6	8.7	9.83	10.35	10.65
20.00	8.2	8.4	8.5	9.62	10.07	10.30
			PGPE			
1.00	6.6	7.9	8.9	8.41	10.41	12.22
2.50	6.5	7.8	8.8	8.30	10.24	11.98
5.00	6.4	7.6	8.6	8.11	9.96	11.60
7.50	6.3	7.4	8.3	7.94	9.69	11.24
10.00	6.2	7.3	8.2	7.77	9.45	10.91
15.00	6.0	7.0	7.8	7.45	8.99	10.30
20.00	5.8	6.7	7.5	7.17	8.57	9.76

The internal pressure values clearly decrease with the size of the diethylene glycol monoalkyl ether molecule, as presented in Figure 3a at different pressures. This means that when the number of methylene groups (nonpolar) increases the internal pressure decreases. This behavior is quite general, and it has also been found for other compounds.[5,24] An inspection of the last Figure shows that the pressure dependence of the internal pressure is higher for the longest diethylene glycol monoalkyl ether, DEGBE. The π values for PGPE are lower than those of DEGBE, ranging over the entire temperature and pressure intervals from (332 to 366) MPa and from (361 to 392) MPa, respectively. A plot of the internal pressure of PGPE as a function of pressure for several temperatures is shown in Figure 3b. For both glycol ethers, when the pressure increases along an isotherm, the internal pressure values increase. This increase is about 2% for both DEGBE and PGPE at 303.15 K, as shown in Figure 3a and b.

Equations of State. As stated before, an important activity has been displayed in developing semiempirical equations of state, and no single equation has been able to establish unrivaled supremacy.[25,26] In previous papers,[23,27,28] it was found that the most popular models for phase

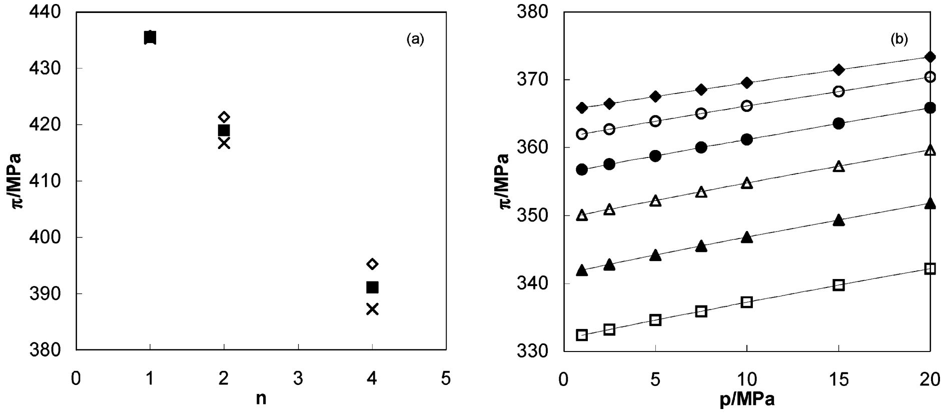

Figure 3. Internal pressure, π, for (a) diethylene glycol monoalkyl ethers, $CH_3-(CH_2)_{n-1}-C_4H_9O_3$, against n (DEGME,[5] $n = 1$; DEGEE,[5] $n = 2$; DEGBE,[this work] $n = 4$) at 303.15 K for different pressures: ×, 1 MPa; ■, 10 MPa; ◇, 20 MPa and (b) PGPE[this work] against the pressure, p, at different temperatures ◆, 293.15 K; ○, 303.15 K; ●, 313.15 K; △, 323.15 K; ▲, 333.15 K; and □, 343.15 K.

Table 4. Molecular Weight M_w, Critical Temperature T_c, Critical Pressure p_c, and Acentric Factor ω, for the Glycol Ethers under Study

component	M_w/g·mol^{-1}	T_c/K	p_c/MPa	ω
DEGME	120.15	672[34]	3.67[34]	0.5157
DEGEE	134.17	670[34]	3.167[34]	0.5737
DEGBE	162.23	692.3[35]	2.79[35]	0.6476
PGPE	118.17	605.1[37]	3.051[37]	0.4720

Table 5. AAD(%) between the Experimental Vapor Pressure Data and Those Predicted Using Several EoS for Pure Glycol Ethers

fluid	ref	PT	PR	SRK
DEGME	38	3.2	5.8	2.2
DEGEE	39, 40, 43	11.8	7.5	7.9
DEGBE	40	7.1	6.8	8.6

equilibria, Soave−Redlich−Kwong[29] (SRK) and Peng−Robinson[30] (PR), give reasonable predictions for the volumetric behavior at high pressure for some pure compounds. Other available equations that correlate the phase equilibria with accuracy similar to that of the SRK or PR EoS and improve the predictions of volumetric properties at high pressure in the liquid region are Soave−Redlich−Kwong with volume translation[31] (SRK-VT) and Patel−Teja[32] (PT). The SRK-VT EoS is a modification of the SRK EoS, which does not affect the calculations of vapor pressures, and the PT EoS is an extension of the Soave,[29] Peng−Robinson,[30] and Schmidt−Wenzel[33] equations. These EoS (PR, SRK, SRK-VT, and PT) can be applied if the critical parameters (critical temperature, T_c, and critical pressure, p_c) and the acentric factor (ω) of the pure components are available. The PT EoS requires, in addition to T_c and p_c, two new parameters to characterize each particular fluid. Patel and Teja[32] have generalized their original equation correlating these new parameters as a function of the acentric factor.

The critical properties and the acentric factor of the pure compounds under study (DEGME, DEGEE, DEGBE, and PGPE) are reported in Table 4. In this work, the available[34,35] experimental critical temperatures and pressures have been considered. In the case of DEGME and DEGEE, Wilson et al.[34] have measured the critical temperatures and pressures, whereas Teja and Rosenthal[35] have reported the experimental critical values for DEGBE. For PGPE, the experimental T_c and p_c values reported by VonNierderhausern et al.[37] were used. The acentric factor, ω, for DEGME, DEGEE, and DEGBE was determined, taking into account the Antoine coefficients for the vapor pressures available in the literature.[38−40] The acentric factor for PGPE has been calculated using the Lee−Kesler method,[41] taking into account the experimental critical temperature and pressure[37] and the experimental boiling point (T_b = 422.95 K)[42] due to the nonexistence of experimental vapor pressure. The Lee−Kesler method has been selected because it better reproduces the acentric factor values of the others glycol ethers under study.

To assess and compare the performances of the different EoS, the absolute average percentual deviation AAD, the maximum percentual deviation MD, and the average percentual deviation BIAS, have been used according to their definitions, given in a previous paper.[27] The deviations between the available experimental[38−40,43] and predicted vapor pressure values for pure glycol ethers are presented in Table 5. The lowest AADs are obtained by using PT and SRK equations of state for DEGME, with PT and PR EoS for DEGBE. For DEGEE, a disagreement is observed among the experimental vapor pressures reported by Akhundov et al.[43] and those published by Stull[39] and Gardner et al.[40] Thus, if Akhundov et al.[43] data are not considered, then the absolute average percentual deviations are reduced for PT, PR, and SRK EoS to 4.9%, 6.2%, and 6.8%, respectively, with PT EoS giving the best predictions.

To study the abilities of these EoS in relation to volumetric predictions at high pressure, the experimental data reported in this work and those from our previous paper[5] have been considered. The AAD, MD, and BIAS obtained from PR, SRK, SRK-VT, and PT $p\rho T$ predictions for the glycol ethers are listed in Table 6 over wide temperature and pressure ranges. This Table shows that when the volume translation is considered for SRK EoS the density predictions are remarkably improved (about 14%), except for DEGBE. As can also be seen in Table 6, the best results (AADs lower than 3%) are obtained with the PT EoS for all pure compounds with the exception of DEGBE, for which the best predictions are those of the PR EoS (1.1%). From the PT EoS predictions, taking into account the BIAS values, it can be observed that the predicted densities are always smaller than the experimental ones, except for DEGBE for which theoretical values are always higher than the measured data. The SRK-VT theoretical values are always higher than the experimental ones for the glycol ethers under study. However, the temperature and pressure dependencies of the density are well predicted by all EoS.

It is interesting to mention that Yan et at.[36] have recently estimated the critical temperature for three of

Table 6. AAD(%), MD(%), and BIAS(%) between the Density Experimental Data and Those Predicted from (283.15 to 353.15) K and (0.1 to 25) MPa Using Several EoS

	ref	N_p[a]	PR	SRK	SRK-VT	PT
DEGME						
AAD			8.4	18.3	3.1	2.9
MD	5	72	9.7	19.2	4.0	4.3
BIAS			8.4	18.3	−3.1	2.9
DEGEE						
AAD			8.3	18.2	6.3	1.5
MD	5	72	9.8	19.3	7.4	3.2
BIAS			8.3	18.2	−6.3	1.5
DEGBE						
AAD			1.1	11.5	19.6	8.5
MD	this work	72	2.7	12.9	20.8	10.4
BIAS			1.0	11.5	−19.6	−8.5
PGPE						
AAD			6.2	16.4	2.8	1.7
MD	this work	72	7.6	17.4	4.1	3.2
BIAS			6.2	16.4	−2.8	1.7

[a] N_p is the number of experimental points.

these compounds (DEGME, DEGEE and DEGBE) using four group-contribution methods; they found deviations in T_c up to 43 K. This deviation in T_c has a great influence on the vapor pressures estimations. For example, if the experimental critical temperature is used for DEGEE, then a deviation in vapor pressure of 4.9% is reached with PT, as already pointed out above. When using the estimated T_c (627.4 K), the deviation is more than 180%. Moreover, the density predictions at different pressures become worse when going from 1.5% to 4.1% for DEGEE using PT.

Conclusions

$p\rho T$ data for DEGBE and PGPE have been measured over the ranges $283.15 \le T/\mathrm{K} \le 353.15$ and $0.1 \le p/\mathrm{MPa} \le 25$. The AADs between our experimental density data at atmospheric pressure and the literature data are lower than or equal to 0.04%. The parameter values for the Tammann−Tait equation have been provided. The internal pressure values decrease with the size of the diethylene glycol monoalkyl ether molecule. Four equations of state (PR, SRK, SRK-VT, and PT) have been tested for pure glycol ethers (DEGME, DEGEE, DEGBE, and PGPE) over broad temperature and pressure ranges. The best $p\rho T$ predictions are obtained with the PT EoS for all pure compounds (AADs lower than 3%) with the exception of DEGBE; the best predictions for this compound are those of PR EoS (1.1%).

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