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Liquid Densities, Kinematic Viscosities, and Heat Capacities of Some Alkylene Glycol Dialkyl Ethers

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Liquid densities and heat capacities at 1 MPa, and kinematic viscosities at atmospheric pressure of monoethylene glycol diethyl ether, diethylene glycol diethyl ether, diethylene glycol dibutyl ether, triethylene glycol dibutyl ether, and dipropylene glycol dimethyl ether were measured in the temperature range (283.15 to 423.15) K. For each substance, experimental data were correlated with temperature using empirical polynomial equations. The experimental data of kinematic viscosity and heat capacity were used to evaluate the predictive capability of some estimation methods of the literature.

Introduction

The cleaning of exhaust air and gas streams from industrial production plants with the simultaneous recovery of useful materials is taking on increasing importance. Not only is it required by legal regulations, it also conserves resources by recycling useful materials. The use of alkylene glycol dialkyl ethers as scrubbing liquids has been suggested[1] because of their favorable properties such as low vapor pressure, low toxicity, low viscosity, high chemical stability, and low meting point.

Also, in the last years, some authors[2−7] have proposed new organic working pairs containing ethylene glycol ethers as absorbent fluids for absorption heat pumps and chillers to overcome the disadvantages of the classical ammonia + water and water + lithium bromide mixtures.

The thermophysical property data available in the literature for these compounds are meager and very limited in the temperature and pressure conditions. To complete our previous works,[8−12] we report experimental measurements of liquid densities at 1 MPa and kinematic viscosities at atmospheric pressure in the temperature range from (283.15 to 423.15) K and heat capacities at 1 MPa from (312.57 to 421.48) K for monoethylene glycol diethyl ether, diethylene glycol diethyl ether, diethylene glycol dibutyl ether, triethylene glycol dibutyl ether, and dipropylene glycol dimethyl ether. Most of the properties reported in the literature for these substances correspond to densities and viscosities at ambient conditions,[13−23] except the data of Ku and Tu[24] for monoethylene glycol diethyl ether and diethylene glycol diethyl ether in the range (288.15 to 343.15) K and those of Vogel[25] for densities of diethylene glycol diethyl ether in the range (289 to 361) K. Concerning heat capacity, the only data available in the literature are those of Villamañan et al.[26] and Kusano et al.[27] for monoethylene glycol diethyl ether at ambient temperature and Zábranský et al.[28] for diethylene glycol diethyl ether in the range (280 to 310) K.

Experimental Section

Materials. Monoethylene glycol diethyl ether (MEGDEE) (Fluka, 99.5%), diethylene glycol diethyl ether (DEGDEE) (Fluka, >99%), diethylene glycol dibutyl ether (DEGDBE) (Fluka, 98%), triethylene glycol dibutyl ether (TrEGDBE) (Clariant, >99%), and dipropylene glycol dimethyl ether (DPrGDME) (Fluka, >99%) and the reference fluids heptane (Fluka, >99.5%), octane (Panreac, >99.5%), dodecane (Sigma, >99%), tridecane (Sigma, >99%), benzene (Panreac, >99.5%), and ethanol (Panreac, >99.5%) were used without further purification but kept over molecular sieves for several days. They were degassed by ultrasonic treatment (Elma type 480/H-2) immediately before use.

Equipment and Procedure. Densities of the liquids at the pressure 1 MPa were measured with an electronic digital densimeter (Anton Paar DMA 60/512P) connected to a pressure system. The pressure of the system was applied with gas nitrogen with a pressure generator (HiP 50-6-15). To prevent the diffusion of nitrogen into the sample, a 0.75 m tube completely filled with the sample was placed before the densimeter cell. The temperature was regulated by a Julabo F-20 HC thermostat and read by a digital precision thermometer (Anton Paar MKT 100) with a micro RTD probe calibrated in the temperature working range with an accuracy of ±0.01 K.

Before density measurements, the system was heated and evacuated simultaneously for several hours to remove any residual humidity. The sample was carefully introduced into the densimeter, and once the system was filled completely with the sample, it was pressurized at 1 MPa. Liquid water and nitrogen gas were used as reference fluids for the calibration of the densimeter. To check the densimeter and the experimental procedure, the liquid densities of ethanol and octane at 1 MPa pressure were measured in the range (283.15 to 423.15) K and compared with the measurement of Cibulka and Hnedkovsky[29] and TRC tables,[30] respectively. Our experimental values are in good agreement with the literature values, the overall average absolute deviation with both sets of data being less than 0.1%. The uncertainty of our measured densities is $\pm 4 \times 10^{-4}$ g·cm^{-3}.

Kinematic viscosities of the liquids at atmospheric pressure were measured with Ubbelohde-type glass capillary tube viscometers (0, 0a, and 0c) using a Schott−Geräte automatic measuring unit (AVS 310) in a thermostated bath (CT 1450/2). The bath was filled with deionized water

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Table 1. Experimental Liquid Densities of Monoethylene Glycol Diethyl Ether (MEGDEE), Diethylene Glycol Diethyl Ether (DEGDEE), Diethylene Glycol Dibutyl Ether (DEGDBE), Triethylene Glycol Dibutyl Ether (TrEGDBE), and Dipropylene Glycol Dimethyl Ether (DPrGDME) at 1 MPa

	ρ/kg·m^{-3}				
T/K	MEGDEE	DEGDEE	DEGDBE	TrEGDBE	DPrGDME
283.15	862.9	917.4	891.6	926.1	913.0
293.15	852.8	907.9	883.2	917.7	903.4
303.15	842.6	898.4	875.0	909.4	894.0
313.15	832.3	889.0	866.8	901.2	884.8
323.15	822.0	879.5	858.5	893.0	875.4
333.15	811.6	870.0	850.4	884.8	866.0
343.15	801.1	860.2	842.1	876.5	856.5
353.15	790.5	850.0	833.7	868.2	846.9
363.15	779.6	840.2	824.9	859.8	837.0
373.15	768.6	830.3	816.5	851.4	827.2
383.15	757.2	820.4	807.9	843.0	817.2
393.15	745.8	810.3	799.4	834.5	807.2
403.15	734.0	800.0	790.8	826.1	797.1
413.15	721.8	789.7	782.1	817.6	786.8
423.15	709.4	779.2	773.3	809.1	776.3

to operate in the temperature range from (283.15 to 343.15) K, and with silicon oil Baysilone M-20 for higher temperatures. The detailed procedure was reported in the previous work of Conesa et al.[11] The fluid used to calibrate the capillaries 0, 0c, and 0a was octane in the temperature range (283.15 to 333.15) K. For higher temperatures, the capillary 0a was calibrated with dodecane. To check the equipment and the procedure, the liquid kinematic viscosities of heptane in the range (283.15 to 323.15) K and tridecane in the range (333.15 to 423.15) K were determined. The maximum deviations of experimental data from the literature data[30] are lower than 0.8%. The uncertainty of the kinematic viscosity data is 4×10^{-3} mm^2·s^{-1}.

The isobaric heat capacity was determined using a heat flux Calvet-type calorimeter (Setaram C-80 II). The detection limit of this calorimeter is 10 μW, and it can operate from ambient temperature to 573 K. The vessels used are made of stainless steel and are of the open type with a special design to avoid the presence of any vapor phase. The heat capacity of a sample was determined using the step by step method. For every measurement the measuring vessel was filled successively with vacuum, water, and the sample while the reference vessel was kept under vacuum. The heating rate was, in all the experiments, 0.3 K·min^{-1}. A more detailed description of the system and procedure was reported by Coxam et al.[31] and Conesa.[12]

To check the equipment and procedure, the liquid heat capacities of ethanol in the range (312.55 to 371.95) K and heptane in the range (312.55 to 421.41) K were determined. The results were compared with the recommended heat capacity data of Zábranský et al.[28] in the same temperature range. The uncertainty of the heat capacity data is 8×10^{-3} J·g^{-1}·K^{-1}.

The deviations of the measured heat capacities from the literature data do not exceed ±0.4%.

Results and Correlations

Density. The liquid densities were measured at 1 MPa from (283.15 to 423.15) K at intervals of 10 K. The experimental densities listed in Table 1 were correlated with the following equation:

$$\rho/\text{kg·m}^{-3} = a_0 + a_1(T/\text{K}) + a_2(T/\text{K})^2 \qquad (1)$$

where ρ is the density, T is the temperature, and the values of the coefficients a_0, a_1, and a_2 are listed in Table 2 for all the substances studied, together with the root-mean-square relative deviation (RMSDs), defined as

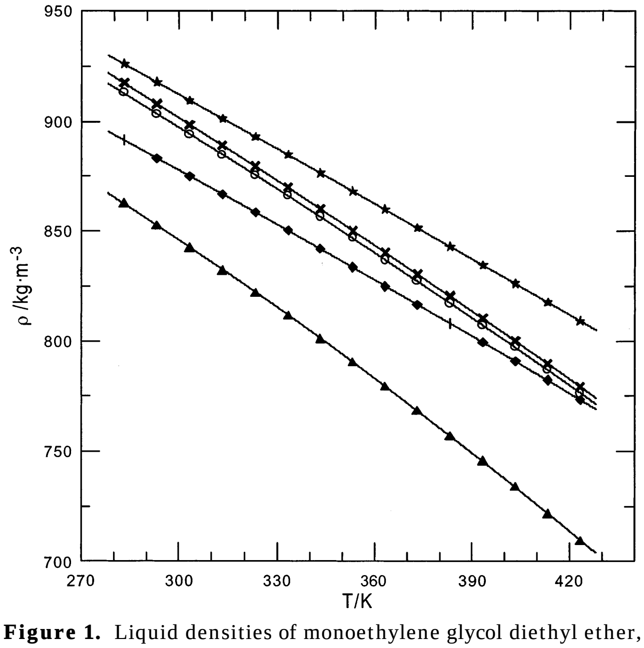

Figure 1. Liquid densities of monoethylene glycol diethyl ether, diethylene glycol diethyl ether, diethylene glycol dibutyl ether, triethylene glycol dibutyl ether, and dipropylene glycol dimethyl ether at 1 MPa: ▲, MEGDEE; ×, DEGDEE; ◆, DEGDBE; ★, TrEGDBE; ○, DPrGDME; —, calculated by eq 1.

Table 2. Parameters of the Liquid Density Correlation (Eq 1) and RMSD Values for Monoethylene Glycol Diethyl Ether (MEGDEE), Diethylene Glycol Diethyl Ether (DEGDEE), Diethylene Glycol Dibutyl Ether (DEGDBE), Triethylene Glycol Dibutyl Ether (TrEGDBE), and Dipropylene Glycol Dimethyl Ether (DPrGDME) at 1 MPa in the Temperature Range (283.15 to 423.15) K

	a_0	a_1	$10^4 a_2$	RMSD
MEGDEE	1066.276	−0.472 333 4	−8.7421	0.04
DEGDEE	1148.966	−0.706 033 5	−3.9603	0.01
DEGDBE	1102.706	−0.680 932 4	−2.3032	0.01
TrEGDBE	1147.426	−0.747 349 8	−1.2329	0.01
DPrGDME	1134.091	−0.654 713 0	−4.4998	0.02

$$\text{RMSD} = 100\left\{\frac{1}{N}\sum_i\left(\frac{X_e - X_c}{X_e}\right)_i^2\right\}^{1/2} \qquad (2)$$

where N, X_e, and X_c are the number of data points, and the experimental and calculated values, respectively. The RMSD values were less than 0.03% for all substances. The experimental and calculated density values are shown in Figure 1.

Viscosity. The kinematic viscosities were measured at atmospheric pressure from (283.15 to 423.15) K at intervals of 10 K. The measurements for MEGDEE were made only up to 353.15 K because its normal boiling point is around 400 K. In Table 3 we report the experimental kinematic viscosities for the substances. The experimental data were fitted as a function of temperature to the equation

$$\ln \nu/\text{mm}^2\text{·s}^{-1} = b_0 + b_1\tau^{-1} + b_2\tau + b_3\tau^2 \qquad (3)$$

where τ is

$$\tau = \frac{T/\text{K}}{298.15} \qquad (4)$$

Table 3. Experimental Kinematic Viscosities of Monoethylene Glycol Diethyl Ether (MEGDEE), Diethylene Glycol Diethyl Ether (DEGDEE), Diethylene Glycol Dibutyl Ether (DEGDBE), Triethylene Glycol Dibutyl Ether (TrEGDBE), and Dipropylene Glycol Dimethyl Ether (DPrGDME) at Atmospheric Pressure

	ν/mm^2·s^{-1}				
T/K	MEGDEE	DEGDEE	DEGDBE	TrEGDBE	DPrGDME
283.15	0.9502	1.835	3.361	5.418	1.409
293.15	0.8223	1.502	2.692	4.157	1.199
303.15	0.7281	1.269	2.210	3.299	1.030
313.15	0.6478	1.092	1.857	2.696	0.9028
323.15	0.5844	0.9556	1.590	2.253	0.8013
333.15	0.5319	0.8477	1.383	1.923	0.7188
343.15	0.4865	0.7598	1.219	1.666	0.6503
353.15		0.6910	1.077	1.464	0.5931
363.15		0.6331	0.9680	1.298	0.5507
373.15		0.5773	0.8720	1.166	0.5060
383.15		0.5320	0.7954	1.045	0.4689
393.15		0.4930	0.7282	0.9490	0.4365
403.15		0.4592	0.6699	0.8680	0.4067
413.15		0.4290	0.6207	0.7979	0.3813
423.15		0.4023	0.5778	0.7369	0.3587

Table 4. Parameters of the Kinematic Viscosity Correlation (Eq 3) and RMSD Values for Monoethylene Glycol Diethyl Ether (MEGDEE), Diethylene Glycol Diethyl Ether (DEGDEE), Diethylene Glycol Dibutyl Ether (DEGDBE), Triethylene Glycol Dibutyl Ether (TrEGDBE), and Dipropylene Glycol Dimethyl Ether (DPrGDME) in the Temperature Range (283.15 to 423.15) K at Atmospheric Pressure

	b_0	b_1	b_2	b_3	RMSD
MEGDEE	−5.193 61	4.595 17	−0.154 42	0.496 77	0.21
DEGDEE	−26.973 66	13.860 94	18.095 59	−4.659 63	0.37
DEGDBE	−22.209 12	12.795 18	13.645 09	−3.329 37	0.30
TrEGDBE	−33.995 15	18.243 64	22.732 07	−5.671 93	0.41
DPrGDME	−19.892 59	10.522 17	12.822 30	−3.348 97	0.26

The coefficients b_0, b_1, b_2, and b_3 and the root-mean-square relative deviations (RMSDs) are listed in Table 4 for all the substances studied.

The maximum and minimum values of RMSD are respectively 0.4% for TrEGDBE and 0.2% for MEGDEE. Figure 2 shows the experimental and calculated values of the kinematic viscosity for the different substances as a function of the temperature. It can be observed in this figure that the kinematic viscosity increases with the length of the ether chain.

The percentage deviations of the kinematic viscosity values for DEGDEE and DEGDBE measured at 298.15 K by Pal and Sharma[20] from those calculated with eq 3 are +2.3% and −0.22%, respectively. The experimental data of Ku and Tu[24] for DEGDEE in the range (293.15 to 343.15) K are in very good agreement with our experimental data except the value at 313.15 K that deviates +2.4% from our data. Nevertheless, the deviations of data reported by these authors for MEGDEE in the same range are 8% below our data.

In the literature there are many methods for the prediction of the dynamic viscosity, but there are few for the kinematic viscosity. One of them was developed by Shing et al.[32] to predict the kinematic viscosity of petroleum mixtures. This method uses a single viscosity value at 310.93 K to make predictions at other temperatures. We calculated the kinematic viscosity of all the alkylene glycol dialkyl ethers in the temperature range studied using this method with the reference viscosity value calculated by eq 3. Figure 3 shows the graphical representation of the experimental and the predicted values using the Shing et al.[32] method for all the substances studied in this work.

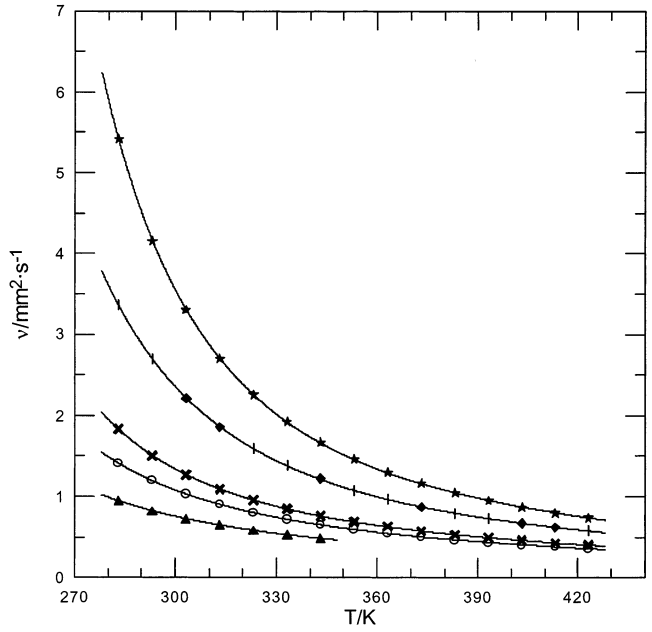

Figure 2. Kinematic viscosities of monoethylene glycol diethyl ether, diethylene glycol diethyl ether, diethylene glycol dibutyl ether, triethylene glycol dibutyl ether, and dipropylene glycol dimethyl ether at atmospheric pressure: ▲, MEGDEE; ×, DEGDEE; ◆, DEGDBE, ★, TrEGDBE; ○, DPrGDME; —, calculated by eq 3.

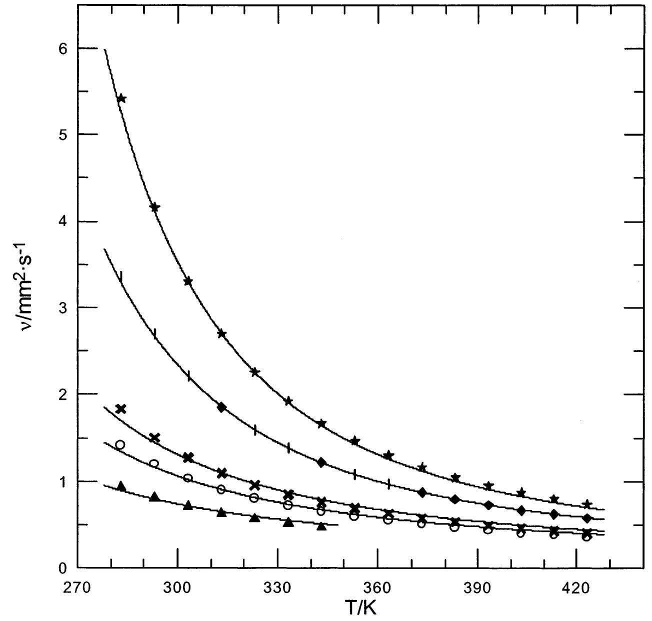

Figure 3. Kinematic viscosities of monoethylene glycol diethyl ether, diethylene glycol diethyl ether, diethylene glycol dibutyl ether, triethylene glycol dibutyl ether, and dipropylene glycol dimethyl ether at atmospheric pressure: ▲, MEGDEE; ×, DEGDEE; ◆, DEGDBE; ★, TrEGDBE; ○, DPrGDME; —, calculated by the Shing et al. method.[36]

The RMSD values between our experimental and predicted data for MEGDEE, DEGDEE, DEGDBE, TrEGDBE, and DPrGDME are 3.7%, 6.0%, 0.76%, 3.1%, and 6.2%, respectively. The predictions for MEGDEE, DEGDEE, and DPrGDME are lower than the experimental data for temperatures below 313.15 K. For DEGDBE and TrEGDBE the calculated values are lower than experimental data in all the range of temperatures.

Heat Capacity. The experimental liquid heat capacities from (312.57 to 421.48) K at 1 MPa are listed in Table 5.

Table 5. Experimental Liquid Heat Capacities of Monoethylene Glycol Diethyl Ether, Diethylene Glycol Diethyl Ether, Diethylene Glycol Dibutyl Ether, Triethylene Glycol Dibutyl Ether, and Dipropylene Glycol Dimethyl Ether at 1 MPa

	c_p/kJ·kg^{-1}·K^{-1}				
T/K	MEGDEE	DEGDEE	DEGDBE	TrEGDBE	DPrGDME
312.57	2.239	2.152	2.091	2.075	2.026
322.47	2.247	2.165	2.111	2.088	2.050
332.37	2.271	2.178	2.132	2.111	2.075
342.27	2.292	2.191	2.155	2.131	2.098
352.18	2.315	2.211	2.179	2.152	2.128
362.08	2.337	2.224	2.200	2.178	2.157
371.98	2.372	2.249	2.230	2.206	2.183
381.88	2.395	2.274	2.261	2.227	2.216
391.78	2.422	2.294	2.292	2.247	2.248
401.68	2.453	2.324	2.315	2.278	2.279
411.58	2.483	2.347	2.346	2.301	2.305
421.48	2.513	2.372	2.372	2.333	2.350

Table 6. Parameters of the Liquid Heat Capacity Correlation (Eq 5) and RMSD Values for Monoethylene Glycol Diethyl Ether (MEGDEE), Diethylene Glycol Diethyl Ether (DEGDEE), Diethylene Glycol Dibutyl Ether (DEGDBE), Triethylene Glycol Dibutyl Ether (TrEGDBE), and Dipropylene Glycol Dimethyl Ether (DPrGDME) at 1 MPa in the Temperature Range (312.57 to 421.48) K

	c_0	10^3c_1	10^6c_2	RMSD
MEGDEE	2.254 43	−2.048 36	6.32	0.14
DEGDEE	2.537 78	−3.678 54	7.81	0.11
DEGDBE	1.859 64	−0.675 77	4.50	0.13
TrEGDBE	1.850 11	−0.538 52	3.98	0.13
DPrGDME	1.507 72	0.734 60	2.93	0.11

For each substance, heat capacity data were fitted as a function of temperature to the equation

$$C_p/\text{kJ}\cdot\text{kg}^{-1}\cdot\text{K}^{-1} = c_0 + c_1(T/\text{K}) + c_2(T/\text{K})^2 \qquad (5)$$

The coefficients c_0, c_1, and c_2 and the root-mean-square relative deviations (RMSDs) are listed in Table 6 for all the substances studied.

The agreement between the experimental and calculated values from eq 5 is very good with an overall average RMSD value of 0.12%. The experimental and calculated values of the heat capacity are represented graphically as a function of the temperature in Figure 4. In this figure, it can be seen that the heat capacity values for MEGDEE are much higher than the values for the other substances studied.

The deviations of the heat capacities for MEGDEE reported by Villamañan et al.[26] and Kusano et al.[27] at 293.15 K and 298.15 K, respectively, from the values extrapolated using eq 5 are ±0.5%.

The methods of Sterning−Brown,[33] Rowlinson,[34] and Rowlinson−Bondi,[34] based on the corresponding-states principle, were used to predict the liquid heat capacities. Since the critical temperature and the acentric factor are needed in these calculations and they were not available, several methods were used to estimate them. The values of the critical temperature T_c and the acentric factor for each substance were estimated using the group-contribution methods of Jobak[35] and Han and Peng,[36] respectively. The values of the molecular weight M, the acentric factor ω, and the critical parameter T_c calculated for each substance are listed in Table 7.

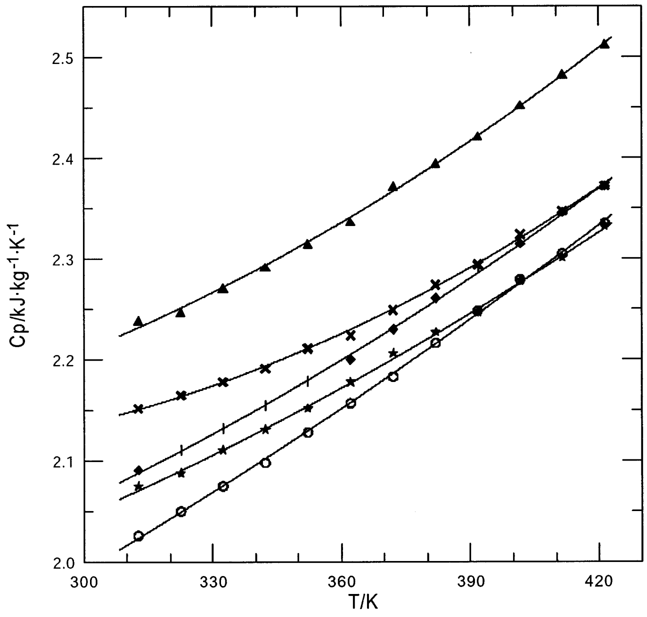

Figure 4. Liquid heat capacities of monoethylene glycol diethyl ether, diethylene glycol diethyl ether, diethylene glycol dibutyl ether, triethylene glycol dibutyl ether, and dipropylene glycol dimethyl ether at 1 MPa: ▲, MEGDEE; ×, DEGDEE; ◆, DEGDBE; ★, TrEGDBE; ○, DPrGDME; —, calculated by eq 5.

Table 7. Molecular Weight *M*, Acentric Factor *ω*, and Critical Temperature T_c for Monoethylene Glycol Diethyl Ether (MEGDEE), Diethylene Glycol Diethyl Ether (DEGDEE), Diethylene Glycol Dibutyl Ether (DEGDBE), Triethylene Glycol Dibutyl Ether (TrEGDBE), and Dipropylene Glycol Dimethyl Ether (DPrGDME).

	M/g·mol^{-1}	ω	T_c/K
MEGDEE	118.18	0.451	571.37
DEGDEE	162.23	0.608	629.63
DEGDBE	218.34	0.769	677.57
TrEGDBE	262.00	0.902	703.00
DPrGDME	162.23	0.608	611.84

Table 8. RMSD (%) of the Heat Capacity Values Predicted by the Sterning−Brown, Rowlinson, and Rowlinson−Bondi Methods from the Experimental Data for Monoethylene Glycol Diethyl Ether (MEGDEE), Diethylene Glycol Diethyl Ether (DEGDEE), Diethylene Glycol Dibutyl Ether (DEGDBE), Triethylene Glycol Dibutyl Ether (TrEGDBE), and Dipropylene Glycol Dimethyl Ether (DPrGDME).

	Sterning−Brown	Rowlinson	Rowlinson−Bondi
MEGDEE	3.69	5.89	3.62
DEGDEE	3.46	5.64	2.06
DEGDBE	1.57	3.59	0.70
TrEGDBE	2.36	4.40	0.32
DPrGDME	0.50	2.50	1.37

The RMSD values between experimental and predicted data are summarized in Table 8. The maximum RMSD values of all studied substances for the Sterning−Brown, Rowlinson, and Rowlinson−Bondi methods are 3.7, 5.9, and 3.6%, respectively, and correspond always to MEGDEE. The predictions with the Rowlinson method are always lower than our experimental values for all the substances studied. For DEGDBE and TrEGDBE the calculated values with the Rowlinson−Bondi method are in very good agreement with our experimental data, while for MEGDEE and DEGDEE they are lower. The predictions for DPrGDME using the Sterning−Brown method are in good agreement with our experimental data, while for the other substances they are lower than experimental values. In Figure 5 the experimental and predicted values of liquid heat capacity for DPrGDME are shown for the three methods.

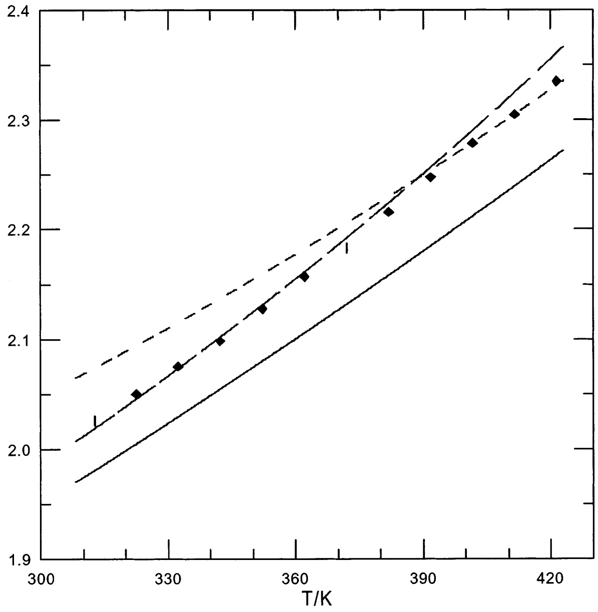

Figure 5. Experimental and predicted liquid heat capacities of dipropylene glycol dimethyl ether: ◆, experimental; —, Rowlinson method; - - - - - -, Rowlinson−Bondi method; — — —, Sterling−Brown method.

Conclusions

In this paper, we have completed our previous works over the measurement of thermophysical properties of alkylene glycol dialkyl ethers. We reported experimental liquid densities and liquid heat capacities at 1 MPa, and kinematic viscosities at atmospheric pressure of monoethylene glycol diethyl ether, diethylene glycol diethyl ether, diethylene glycol dibutyl ether, triethylene glycol dibutyl ether, and propylene glycol dimethyl ether in the temperature range 283.15−423.15 K. The experimental data for each substance studied were correlated with the temperature using polynomial equations.

The experimental data of kinematic viscosity and heat capacity were used to evaluate the predictive capability of some estimation methods. Although the Shing et al.[32] method was developed for the prediction of kinematic viscosities of petroleum mixtures, the deviations of the prediction values from the experimental data for the alkylene glycol dialkyl ethers are always lower than 6.2%. The methods of Sterning−Brown,[33] Rowlinson,[34] and Rowlinson−Bondi[34] to predict the heat capacity were used. The best predictions were obtained with the Rowlinson−Bondi method with deviations lower than 3.6%.

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