Density, Viscosity, and Thermal Conductivity of Aqueous Ethylene, Diethylene, and Triethylene Glycol Mixtures between 290 K and 450 K

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The density, viscosity, and thermal conductivity of ethylene glycol + water, diethylene glycol + water, and triethylene glycol + water mixtures were measured at temperatures ranging from 290 K to 450 K and concentrations ranging from 25 mol % glycol to 100 mol % glycol. Our data were generally in agreement with the limited data available in the literature and were correlated using simple empirical expressions and the generalized corresponding states principle (GCSP). The GCSP method, with two adjustable parameters for each property, offers the potential for judicious extrapolation of density and transport property data for all glycol + water mixtures.

Introduction

Aqueous glycol solutions have found widespread application in the manufacture of solvents, hygroscopic agents, lubricants, and conditioning agents. Accurate knowledge of their thermophysical properties is therefore essential in process calculations involving these mixtures. Despite this need, however, literature data on density, viscosity, and thermal conductivity of glycol + water mixtures are generally limited to ambient temperatures.[1−5] The present work extends such data to a wider temperature range and evaluates two models to correlate experimental results and to predict the thermophysical properties of these highly nonideal systems.

Experimental Section

Materials. Reagent grade ethylene glycol (EG), diethylene glycol (DEG), and triethylene glycol (TEG) were purchased from Aldrich Company and used in the experiments without further purification. The stated minimum purity of these reagents was 99.8 mol %, 99 mol %, and 99 mol %, respectively. Glycol + water mixtures were prepared gravimetrically using double distilled water.

Measurements. Densities and viscosities of the mixtures were measured using a pycnometer and a capillary viscometer placed inside a high-pressure view cell. The view cell was pressurized to suppress boiling of the liquids in the pycnometer and viscometer, and it was equipped with glass view ports for visual observation of liquid levels. The pycnometer had graduated markings for volume and was calibrated using pure mercury. The viscometer was calibrated by the manufacturer, and the calibration was validated by measurement of the viscosity of several liquids. Measured viscosities were reproducible to ±1%, and their uncertainty was estimated to be ±2%. The temperature in the view cell was measured with a platinum resistance thermometer, and its uncertainty was estimated to be ±0.1 K. The reproducibility of the density measurements was ±0.1%, and the uncertainty was estimated to be ±0.2%. Additional details relating to the apparatus and experimental procedure are given elsewhere.[6−8]

Thermal conductivity was measured using the relative transient hot-wire method. Since the wire must be insulated for measurements involving electrically conducting liquids, a Pyrex capillary filled with liquid mercury was employed for this purpose, as described in our earlier work.[7−9] The capillary was part of a Pyrex cell placed inside a thermostated high-pressure vessel that kept the temperature in the cell constant within ±0.1 K. The effective length of the wire was obtained by calibration using IUPAC suggested values of the thermal conductivity of water,[10,11] and it was validated by measurement of the thermal conductivity of a second IUPAC reference liquid, dimethylphthalate.[11] Each reported value of the thermal conductivity was obtained by averaging the results of five experiments. The values were reproducible within ±1% and were estimated to have an uncertainty of ±2%. Further details of the experimental apparatus and technique are given elsewhere.[7−9]

Results and Discussion

Tables 1, 3, and 4 list the experimental densities, viscosities, and thermal conductivities of glycol + water mixtures containing (25, 50, and 75) mole % glycol. The temperature range of the measurements was 293 K to 450 K, and the pressures were in the range (0.1 to 2) MPa in order to suppress boiling of the liquids. These pressures were not expected to affect the thermal conductivity[10] to an appreciable extent, within experimental error, and the experimental pressures are not therefore reported in the tables. The thermal conductivity of EG + water at 445 K and $x_1 = 0.25$ and 0.5 could not be measured due to the onset of convection in our apparatus.

The property y (where $y = \rho$, ln η, or λ) of glycol (1) + water (2) can be described by the following equation:

$$y = w_1 y_1 + w_2 y_2 + (y_1 - y_2) w_1 w_2 (A_4 + A_5 w_1 + A_6 t) \quad (1)$$

where $t = T/\mathrm{K} - 273.15$, w_1 is the mass fraction of glycol, $w_2 = 1 - w_1$, and y_1 and y_2 are the values of ρ, ln η, or λ for pure glycol and pure water, respectively. In the case of ρ and λ, y_1 is given by

$$y_1 = A_1 + A_2 t + A_3 t^2 \quad (2a)$$

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Table 1. Densities of Glycol + Water Mixtures

T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}	T/K	ρ/kg·m^{-3}
Ethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
296.40	1068	297.05	1094	297.35	1106
313.05	1058	313.05	1083	313.05	1094
333.05	1045	333.10	1069	333.10	1080
353.05	1030	353.05	1054	353.05	1065
374.85	1013	376.05	1035	373.45	1049
396.95	995.5	390.25	1023	383.35	1041
417.15	977.4	406.05	1010	393.20	1033
431.55	964.9	423.55	994.5	403.15	1025
		436.65	982.5	404.75	1023
				413.15	1016
				418.35	1012
				431.95	1000
				445.85	987.0
Diethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
294.25	1095	293.45	1111	294.05	1115
313.05	1081	312.95	1097	313.05	1101
333.05	1066	332.95	1082	333.05	1087
353.15	1051	352.95	1066	353.05	1071
381.45	1027	380.05	1043	382.55	1048
392.75	1017	390.25	1035	390.95	1042
409.25	1004	405.35	1023	406.95	1029
426.35	987.6	424.25	1006	424.65	1014
441.65	973.3	443.05	988.2	443.65	996.3
Triethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
293.85	1108	294.05	1120	294.15	1122
313.05	1093	313.05	1105	313.05	1108
333.05	1078	333.05	1089	333.05	1092
353.05	1061	353.05	1073	353.05	1076
379.65	1039	384.25	1046	384.95	1049
389.35	1030	391.35	1041	392.55	1044
405.75	1016	407.85	1028	407.45	1032
423.35	999.2	425.95	1011	425.35	1016
442.45	980.0	443.45	995.0	444.55	999.5

Table 2. Viscosities of Pure Glycols

ethylene glycol		diethylene glycol		triethylene glycol	
T/K	η/mPa·s	T/K	η/mPa·s	T/K	η/mPa·s
295.55	19.1	294.20	35.1	299.65	35.1
313.05	9.74	313.05	15.1	313.05	19.5
333.10	5.24	333.05	7.64	333.10	9.66
353.05	3.13	353.05	4.41	353.05	5.55
372.15	2.06	373.60	2.76	373.50	3.41
392.45	1.44	393.40	1.92	393.30	2.36
412.25	1.07	413.25	1.41	413.25	1.71
428.30	0.867	428.15	1.15	428.10	1.39

In the case of viscosity, y_1 (=ln η) is given by

$$y_1 = A_1 + A_2/(t + A_3) \tag{2b}$$

Functions y_2 ($y_{2\rho}$, $y_{2\eta}$, and $y_{2\lambda}$) were obtained by fitting of literature data[10] between 293 K and 473 K, and they are given below:

$$y_{2\rho} = 1002.17 - 0.116189t - (0.358024 \times 10^{-2})\,t^2 + (0.373\,667 \times 10^{-5})\,t^3 \tag{3a}$$

$$y_{2\eta} = -3.758023 + 590.9808/(t + 137.2645) \tag{3b}$$

$$y_{2\lambda} = 0.570990 + (0.167156 \times 10^{-2})t - (0.609054 \times 10^{-5})\,t^2 \tag{3c}$$

where $y_{2\rho} = \rho$/kg·m^{-3}, $y_{2\eta} = \ln(\eta$/mPa·s), and $y_{2\lambda} = \lambda$/W·m^{-1}·K^{-1}.

Table 3. Viscosities of Glycol + Water Mixtures

T/K	η/mPa·s	T/K	η/mPa·s	T/K	η/mPa·s
Ethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
296.45	3.69	297.05	7.61	297.35	12.3
313.05	2.29	313.05	4.49	313.05	6.97
333.05	1.47	333.10	2.62	333.10	3.88
353.05	0.990	353.05	1.70	353.05	2.42
378.85	0.689	380.05	1.07	373.45	1.62
400.95	0.518	394.25	0.870	383.35	1.37
421.15	0.418	410.05	0.706	393.20	1.18
435.55	0.366	427.55	0.573	403.15	1.02
		440.65	0.500	407.75	0.978
				413.15	0.903
				422.35	0.806
				435.95	0.691
				449.85	0.594
Diethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
294.25	9.79	293.45	22.0	294.05	30.4
313.05	4.88	312.95	9.64	313.05	13.3
333.05	2.76	332.95	5.03	333.05	6.75
353.15	1.76	352.95	3.01	353.05	3.90
384.95	1.03	382.55	1.66	385.05	2.00
396.75	0.912	394.25	1.37	394.95	1.72
413.25	0.743	409.35	1.12	410.95	1.35
430.35	0.610	428.25	0.865	428.65	1.05
445.65	0.521	447.05	0.707	447.65	0.838
Triethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
293.85	19.2	294.05	36.0	294.15	43.2
313.05	8.60	313.05	15.2	313.05	18.3
333.05	4.52	333.05	7.52	333.05	9.04
353.05	2.70	353.05	4.32	353.05	5.19
382.15	1.50	386.75	2.09	387.45	2.41
393.35	1.27	395.35	1.84	396.55	2.11
409.75	1.00	411.85	1.43	411.45	1.68
427.35	0.813	429.95	1.11	429.35	1.30
446.45	0.662	447.45	0.903	448.55	1.03

Table 4. Thermal Conductivities of Glycol + Water Mixtures

T/K	λ/W·m^{-1}·K^{-1}	T/K	λ/W·m^{-1}·K^{-1}	T/K	λ/W·m^{-1}·K^{-1}
Ethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
303.4	0.391	312.3	0.316	298.5	0.269
323.6	0.400	324.4	0.314	324.2	0.272
348.6	0.416	347.7	0.324	347.5	0.277
372.0	0.423	372.8	0.330	371.9	0.281
395.9	0.428	397.0	0.332	396.8	0.280
421.0	0.432	421.7	0.320	419.9	0.279
				441.6	0.276
Diethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
294.4	0.291	295.9	0.235	296.4	0.209
323.4	0.310	322.1	0.244	323.8	0.213
347.4	0.315	347.8	0.249	349.6	0.217
373.0	0.322	371.9	0.258	373.2	0.219
396.0	0.325	395.6	0.257	396.9	0.217
421.5	0.330	422.0	0.256	421.6	0.222
447.5	0.328	447.0	0.253	446.8	0.213
Triethylene Glycol (1) + Water (2)					
$x_1 = 0.25$		$x_1 = 0.50$		$x_1 = 0.75$	
295.0	0.258	295.5	0.210	295.5	0.195
323.8	0.268	324.6	0.216	327.1	0.197
349.1	0.275	349.5	0.219	349.8	0.201
373.8	0.280	373.6	0.220	372.7	0.202
396.4	0.285	396.0	0.221	397.6	0.200
421.3	0.292	421.9	0.221	422.5	0.198
445.0	0.282	448.3	0.216	446.5	0.195

Equation 1 was proposed by Bohne et al.[3] to correlate the thermal conductivity of mixtures as a function of temperature and mass fraction. It is used in this work to correlate all properties. Pure component data required in the calculations are presented in Table 2 in the case of the viscosity, or from previous results reported by us[9,12] in the

Table 5. Coefficients A_i of Eqs 1 and 2

j^a	A_1	A_2	A_3	A_4	A_5	A_6	AAD/%	MAD/%
				ρ/kg·m^{-3} Correlation				
1	1127.68	-0.65816	-6.1765×10^{-4}	0.30590	0.13781	-1.8961×10^{-3}	0.038	0.133
2	1132.35	-0.67950	-4.7565×10^{-4}	0.90820	-0.26348	-3.3787×10^{-3}	0.056	0.173
3	1139.48	-0.71040	-4.3663×10^{-4}	1.1712	-0.52694	-3.8797×10^{-3}	0.074	0.215
				ln η/mPa·s Correlation				
1	-3.61359	986.519	127.861	-0.165301	-0.287325	1.10978×10^{-3}	0.80	2.35
2	-3.25001	901.095	110.695	-0.364260	0.334513	9.24070×10^{-4}	1.49	3.21
3	-3.11771	914.766	110.068	-0.727092	1.21086	-1.36642×10^{-3}	0.91	2.71
				λ/W·m^{-1}·K^{-1} Correlation				
1	0.24658	2.5372×10^{-4}	-1.3186×10^{-6}	0.14219	0.38715	-6.6551×10^{-4}	0.64	1.82
2	0.19365	1.9938×10^{-4}	-1.0584×10^{-6}	0.63029	-0.19822	-1.2787×10^{-3}	0.80	2.37
3	0.18890	1.1485×10^{-4}	-8.4807×10^{-7}	0.11107	0.62975	-1.5995×10^{-3}	0.46	2.04

[a] $j = 1$ for ethylene glycol + water, $j = 2$ for diethylene glycol + water, and $j = 3$ for triethylene glycol + water.

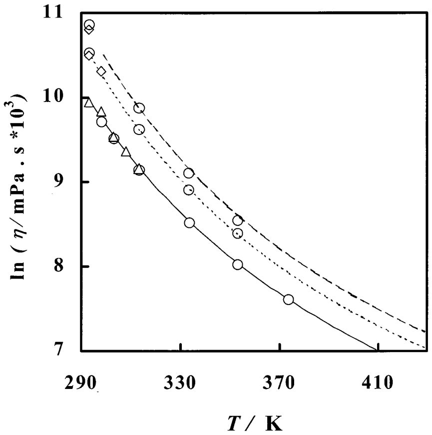

Figure 1. Viscosity of glycols as a function of temperature. Lines represent smoothed data of this work for EG (solid line), DEG (dotted line), and TEG (dashed line). Experimental data of Bohne et al.[3] (○); Tsierkezos and Molinou[1] (△); and Riddick et al.[5] (◇).

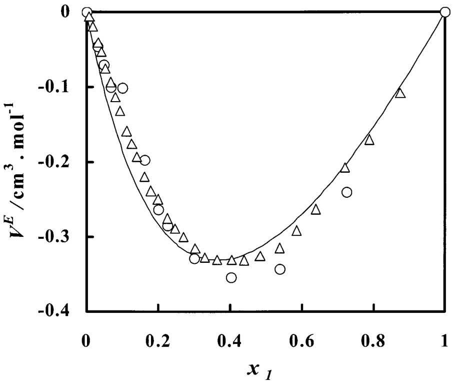

Figure 2. Excess volumes of EG + water mixtures at 303 K. Experimental data of Tsierkezos and Molinou[1] (○) and Lee and Hong[2] (△). Solid line represents the smoothed data of this work.

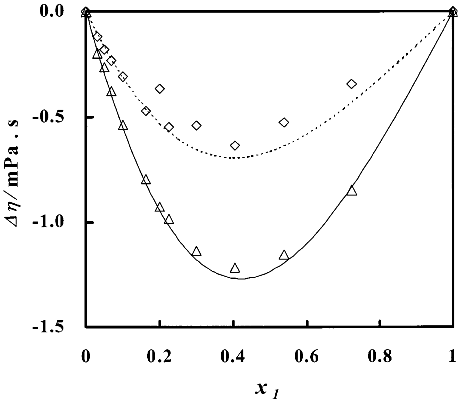

Figure 3. Viscosity deviations for EG + water mixtures. The solid line represents smoothed values at $T = 303$ K, and the dashed line represents values at $T = 313$ K. Experimental data of Tsierkezos and Molinou[1] at $T = 303$ K (◇) and $T = 313$ K (△).

case of density and thermal conductivity. Coefficients of eqs 1 and 2 (A_1 to A_6) obtained by fitting the data are given in Table 5 for each property. The table also lists average absolute deviations (AADs) and maximum absolute deviations (MADs) between experimental data and smoothed (correlated) values. The calculated AAD and MAD are within experimental uncertainty for all three properties.

Comparison of our viscosity data and literature values is shown in Figure 1 for pure glycols. There is good agreement with literature values at low temperatures and fair agreement at high temperatures. For mixtures, comparisons were not feasible at high temperatures because of the scarcity of literature data. Mixture data at low temperatures were therefore compared in terms of excess molar volume V^E, viscosity deviation $\Delta\eta$, and thermal conductivity deviation $\Delta\lambda$, defined as follows:

$$V^E = x_1 M_1(1/\rho_m - 1/\rho_1) + x_2 M_2(1/\rho_m - 1/\rho_2) \quad (4a)$$

$$\Delta\eta = \eta_m - x_1\eta_1 - x_2\eta_2 \quad (4b)$$

$$\Delta\lambda = \lambda_m - x_1\lambda_1 - x_2\lambda_2 \quad (4c)$$

where the subscripts m, 1, and 2 refer to the mixture, pure glycol, and pure water, respectively, and M is the molecular weight. Smoothed experimental values from eqs 1−3 were used in the comparisons. Figure 2 shows that our V^E curves agree well with literature data in the entire concentration range. Figure 3 demonstrates good agreement with the literature in the case of $\Delta\eta$ at 303 K, and reasonable agreement at 313 K. In the case of the thermal conductivity, our results agree with the data of Bohne et al.[3] in the low-concentration range at all temperatures.

V^E, $\Delta\eta$, and $\Delta\lambda$ were also calculated for all glycol + water mixtures in the temperature range 293 K to 433 K. Excess volumes V^E were found to be negative at all temperatures but became less negative with increasing temperature. This behavior is quite common in completely miscible water + polar organic mixtures[13] and agrees with the data of Tsierkezos and Molinou[1] for EG + water mixtures.

Thermal conductivity deviations were found to be negative for all mixtures studied at all temperatures. Also, $\Delta\lambda$ values became more negative with increasing temperature (as shown in Figure 4) and the relative deviations $\Delta\lambda/\lambda$ were quite large. The behavior of $\Delta\eta$ was more complex. In the case of EG + water, $\Delta\eta$ values were found to be negative

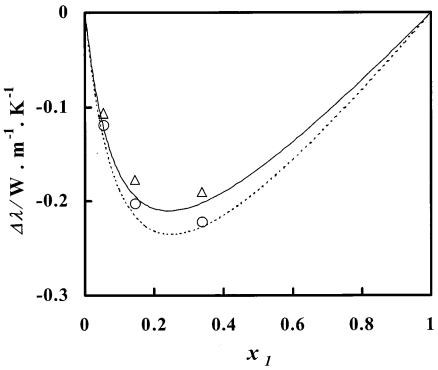

Figure 4. Thermal conductivity deviations for DEG + water. The solid line represents smoothed values at $T = 293$ K, and the dashed line represents values at $T = 433$ K. Experimental data of Obermeier et al.[4] at $T = 293$ K (△) and at $T = 433$ K (○).

between 293 K and 348 K, then became positive in the EG-rich region at higher temperatures, and finally attained positive values at all concentrations at high temperatures. At low concentrations of DEG + water, values of $\Delta\eta$ were negative at 293 K but became positive at higher temperatures. This behavior, however, needs to be verified with additional experiments in the low-concentration regions.

Generalized Corresponding States Method

In addition to the empirical correlations discussed above, the two-reference-fluid generalized corresponding states principle (GCSP) of Teja et al.[14−18] was also used to correlate the data. The GCSP method relates the quantities $Z_C V_R$, $\ln(\eta\xi)$, or $\lambda\phi$ of the mixture to the properties of two reference fluids r1 and r2 at the same reduced temperature T_R and reduced pressure P_R as follows:

$$Z_C V_R = x_1(Z_C V_R)^{(r1)} + x_2(Z_C V_R)^{(r2)} \tag{5a}$$

$$\ln(\eta\xi) = x_1 \ln(\eta\xi)^{(r1)} + x_2 \ln(\eta\xi)^{(r2)} \tag{5b}$$

$$\lambda\phi = x_1(\lambda\phi)^{(r1)} + x_2(\lambda\phi)^{(r2)} \tag{5c}$$

in which

$$\xi = V_C^{2/3} T_C^{-1/2} M^{-1/2} \tag{6a}$$

$$\phi = V_C^{2/3} T_C^{-1/2} M^{1/2} \tag{6b}$$

In the above equations, Z is the compressibility and V is the volume. The subscript C denotes the critical point, and superscripts r1 and r2 denote the properties of two reference fluids. These equations can be extended to mixtures using

$$V_C = \Sigma\Sigma x_i x_j V_{Cij} \tag{7a}$$

$$T_C V_C = \Sigma\Sigma x_i x_j T_{Cij} V_{Cij} \tag{7b}$$

$$Z_C = \Sigma x_i Z_{Ci} \tag{7c}$$

where subscripts i or ii denote pure component properties. When $(i \neq j)$

$$V_{Cij} = (1/8)(V_{Ci}^{1/3} + V_{Cj}^{1/3})^3 \theta_{ij} \tag{8a}$$

$$T_{Cij} V_{Cij} = (T_{Cii} T_{Cjj} V_{Ci} V_{Cj})^{1/2} \psi_{ij} \tag{8b}$$

θ_{ij} and ψ_{ij} are binary interaction parameters which must be obtained by fitting experimental data. Critical properties (experimental or estimated values) of pure glycols and water required in the calculations were obtained from the literature.[19] Other required properties were calculated from eqs 2 and 3. It was found that density and viscosity could be correlated better using mass fraction based mixing rules, whereas thermal conductivity could be better correlated using mole fraction based mixing rules. Binary interaction coefficients determined using the GCSP model are listed in Table 6 together with the corresponding AAD and MAD. The results demonstrate that it is possible to correlate all data within experimental uncertainty using only two adjustable parameters per binary system for each property.

Table 6. GCSP Correlations of Density, Viscosity, and Thermal Conductivity of Glycol + Water Mixtures

	no of data points	AAD/%	MAD/%	θ_{12}	ψ_{12}
		Density[a]			
EG + H_2O	30	0.08	0.28	1.0857	1.0166
DEG + H_2O	27	0.11	0.32	1.1755	1.0026
TEG + H_2O	27	0.11	0.27	1.2299	0.9394
		Viscosity[a]			
EG + H_2O	30	2.07	4.34	1.0166	1.0148
DEG + H_2O	27	1.86	3.51	0.8057	0.7818
TEG + H_2O	27	3.16	8.31	2.2424	1.7868
		Thermal Conductivity[b]			
EG + H_2O	19	0.91	2.06	1.4019	1.3209
DEG + H_2O	21	1.25	3.33	2.0654	1.6111
TEG + H_2O	21	0.50	2.23	2.5218	1.8538

[a] Mass fraction based mixing rules. [b] Mole fraction based mixing rules.

Conclusions

Densities, viscosities, and thermal conductivities of EG + water, DEG + water, and TEG + water were measured at temperatures ranging from 290 K to 450 K and glycol concentrations from 25 to 100 mol %. Our data generally agreed with available literature data within experimental uncertainty. The temperature behavior of the excess properties was also examined, and large deviations from ideal behavior were observed. The data were correlated using an empirical relationship and also using the GCSP method. The GCSP method required only two parameters per binary system to correlate each property over the entire range of temperatures studied. This suggests that the method can be used for interpolation and judicious extrapolation of transport property data for glycol + water mixtures.

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