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Liquid viscosities, excess properties, and viscous flow thermodynamics of triethylene glycol + water mixtures at T = (298.15, 303.15, 308.15, 313.15, and 318.15) K

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ABSTRACT

This paper reports measurements of densities and viscosities for the binary mixtures of triethylene glycol (TEG) + water as a function of composition at T = (298.15, 303.15, 308.15, 313.15, and 318.15) K. Densities were measured using a capillary pycnometer and viscosities were determined with an Ubbelohde capillary viscometer. The experimental results are compared with data published in the literatures. From the experimental data, including density and viscosity values, the excess molar volumes V_m^E, and viscosity deviations $\Delta\nu$, the calculated results are fitted to a Redlich–Kister equation to obtain the coefficients and estimate the standard deviations between the experimental and calculated quantities. The values of V_m^E are negative in the whole composition range, whereas the values of $\Delta\nu$ are positive over the major composition range. From the kinematic viscosity data, enthalpy of activation for viscous flow (ΔH^*) and entropy of activation for the viscous flow (ΔS^*) were calculated.

1. Introduction

Sulfur dioxide (SO_2) is one of the most significant atmospheric pollutants and its emission is directly related to combustion processes, such as the consumption of fossil fuels and burning of biomass [1]. It is known to have crucial effects on human health and environment, and as a consequence, receives more and more attention. Presently, there are several ways of reducing SO_2 emissions from flue gases [2–4]. Limestone-based wet flue gas desulfurization (WFGD) technology has been seen as the most effective method for the control of SO_2 from coal fired boilers [5]. However, WFGD produces the dry product of gypsum, which is difficult to recycle, so that a preferable method has been developed. Recently, more and more organic solvents are used in gas sweetening absorption processes [6,7]. Among numerous organic solvents, alcohols show favorable absorption and desorption properties for acid gases in industrial processes [8,9], for this reason, the alcohol + water systems for SO_2 scrubbing had been paid particular attention in previous works [10–15].

Ethylene glycol (EG) and its similar compounds of various molecular weights showed important chemical uses in the removal of SO_2 from flue gas [4] due to their low vapor pressure, low toxicity, high chemical stability, and low melting point. The primary advantages of triethylene glycol (TEG) may be also related with high solubility and desorption capability for SO_2. The knowledge of physicochemical properties of TEG + water mixtures (TEGW) over a wide range of temperatures is important for practical applications in flue gas desulfurization (FGD) [16]; however, the previous literatures [17–32] show that only the partly mixing properties of TEGW have been reported. Therefore, we have to carry out the measurements for densities and viscosities of TEGW.

This paper is a part of the systematic studies on the physicochemical properties of binary mixtures of EG and its similar compounds [9–15,33,34]. In present work, the densities and viscosities of TEGW are reported in the temperatures range of (298.15 to 318.15) K, over the entire mole fraction range of TEG under atmospheric pressure. From these results, the excess molar volumes (V_m^E), viscosity deviations ($\Delta\nu$), enthalpy of activation for viscous flow (ΔH^*), and entropy of activation for the viscous flow (ΔS^*) were calculated.

2. Experimental section

2.1. Materials

Analytical grade TEG was purchased from Beijing Reagent Company. TEG was dried over 0.4 nm molecular sieves before measurements, and it was degassed by ultrasound just before the experiment. The purity of the sample was checked by density and viscosity determination at 298.15 K. The density and viscosity of TEG at 298.15 K was found to be 1.1192 g cm^{-3} and 35.8 mPa s, in good agreement with the literatures [17–22,27,29]. Bi-distilled water was used in present work.

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2.2. Measurements

Solvent mixtures were prepared by mass using an electronic analytical balance with a precision of ± 0.0001 g (Sartorius BS 224S). The uncertainty in the mole fraction for each binary mixture was estimated to be ± 0.0001.

Densities of pure liquids and their mixtures were determined using a bicapillary pycnometer with a bulb volume of 10 cm^3. The volume of the pycnometer was calibrated as a function of temperature using distilled, deionized, and degassed water at various temperatures of (298.15, 303.15, 308.15, 313.15, and 318.15) K. A thermostatically controlled and well-stirred water bath, which was controlled to ± 0.01 K, was used for all the density and viscosity measurements. The pycnometer filled with liquid was kept in the water bath for 25 min to attain thermal equilibrium. Each experimental density value was an average of at least three measurements; furthermore, the uncertainty of the density measurements was estimated to be ± 0.03%.

The kinematic viscosity in both the pure components and their mixtures was made with a commercial capillary viscometer of the Ubbelohde type, which was calibrated with bi-distilled water and ethanol (HPLC grade) at T = (298.15 to 318.15) K. The flow time was determined with a hand-held digital stopwatch capable of measuring time within ± 0.01 s. The average of sixteen flow times for each fluid was taken for the purpose of the calculation of viscosities.

The kinematic viscosity (ν) was calculated from the following equation

$$\nu = At - \frac{B}{t} \tag{1}$$

where ν denotes the kinematic viscosity; t denotes flow time of liquids; and A and B denote viscometer constants, respectively. A and B are calculated from measurements with the calibration fluids. The absolute viscosity (η) was obtained by multiplying the determined ν by the measured density ($\eta = \nu\rho$). We estimated the uncertainty of the viscosity measurement to be lower than ± 0.3 %.

The experimental densities and viscosities of pure TEG at the various temperatures are compared with the reported literature values and listed in Table 1.

3. Results and discussion

Experimental densities of TEGWs at T = (298.15, 303.15, 308.15, 313.15, and 318.15) K throughout the whole concentration range are plotted in Fig. 1.

Fig. 1 show that the density values increase with the increasing TEG concentration in TEGWs over the whole concentration range.

Table 1
Comparison of experimental densities (ρ), viscosities (η), of TEG with literature values at various temperatures.

T/K	ρ/(g cm^{-3})		η(mPa s)	
	Exptl.	Lit.	Exptl.	Lit.
298.15	1.1192	1.11966[17], 1.11959[18], 1.11950[19], 1.11984[20] 1.11979[21], 1.11991[22] 1.11976[23], 1.11978[30]	35.8	34.9398[18], 37.26[28]
303.15	1.1151	1.11583[17], 1.11588[18] 1.11585[22]	28.9	29.256[25], 29.224[26]
308.15	1.1118	1.11196[18], 1.11207[20] 1.11196[21], 1.11209[22] 1.11209[24], 1.11259[29] 1.11261[30], 1.1120[31] 1.11210[32]	23.0	21.306[18], 23.841[24] 22.853[25], 22.961[26, 27]
313.15	1.1081	1.10953[18], 1.10802[22] 1.1080[26]	18.3	19.8[33]
318.15	1.1042	1.10427[20], 1.10430[21] 1.10413[22], 1.10430[23]	14.9	

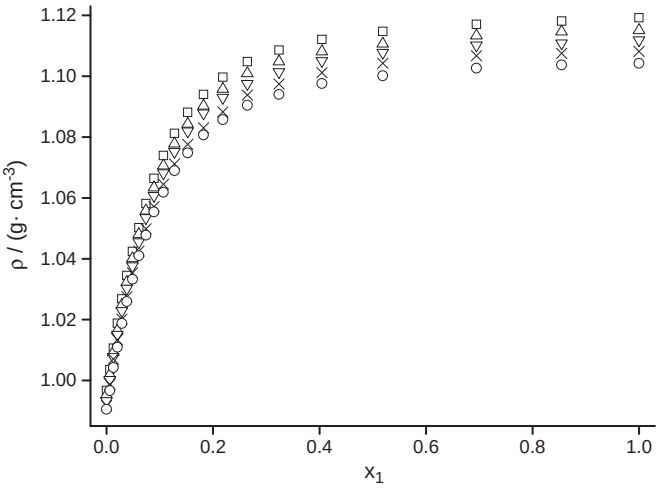

Fig. 1. Experimental densities with mole fraction for TEG (1) + water (2): □, 298.15 K; △, 303.15 K; ▽, 308.15 K; ×, 313.15 K; ○, 318.15 K.

Especially, the values quickly increase in the range of $x_1 = (0$ to $0.26)$. Meanwhile, the density values decrease with the augment of temperatures at the same composition.

The excess mole volume, V_m^E, was calculated from density measurements according to the following equation

$$V_m^E = \frac{x_1 M_1 + x_2 M_2}{\rho_m} - \left(x_1 \frac{M_1}{\rho_1} + x_2 \frac{M_2}{\rho_2}\right) \tag{2}$$

where ρ_m denotes the density of the mixture and x_1, ρ_1, M_1, x_2, ρ_2, and M_2 denote the mole fractions, densities, and molecular weights of pure TEG and pure water, respectively. The dependence of V_m^E at various temperatures is plotted in Fig. 2.

From Fig. 2, V_m^E is negative for all the mixtures over the entire mole fraction range at each temperature, as is common for other completely miscible (water + organic) solvents. The maximum is found at about $x_1 = 0.26$, which indicates that one TEG molecule can combine 3 water molecules to form one tighter complex in the TEGW system. Additionally, these V_m^E values become less negative with the increasing temperatures.

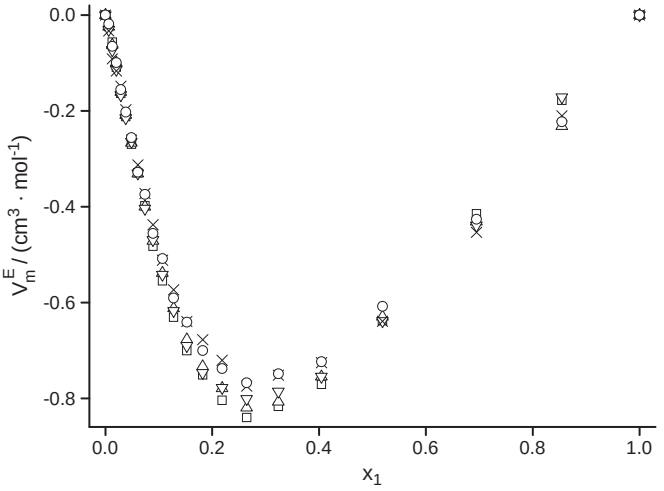

Fig. 2. Excess mole volumes with mole fraction for TEG (1) + water (2): □, 298.15 K; △, 303.15 K; ▽, 308.15 K; ×, 313.15 K; ○, 318.15 K.