



# Study of thermodynamic and transport properties of aqueous system containing poly(ethylene glycol) dimethyl ether 2000 and poly(propylene glycol) 400



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## ABSTRACT

Mixture of {poly(ethylene glycol) dimethyl ether 2000 (PEGDME2000) + poly(propylene glycol) 400 (PPG400) + water} forms an aqueous two phase system. For this system the binodal curves were measured at two temperatures from which the single phase region was obtained. The density, speed of sound and viscosity measurements were performed in the single phase region for {poly(ethylene glycol) dimethyl ether 2000 (PEGDME2000) + poly(propylene glycol) 400 (PPG400) + water} system and the corresponding binary ((PPG400) + water) solutions at  $T = (293.15, 298.15, 303.15, 308.15, \text{ and } 313.15)$  K. From these measurements, values of the excess molar volume ( $V_m^E$ ) and isentropic compressibility deviation ( $\Delta K_S$ ) were calculated and correlated to the modified Wilson and polymer NRTL models. The viscosity values of binary and ternary solutions were correlated by the Eyring-modified Wilson and Eyring-NRTL models. From the density data for the binary and ternary solutions at dilute range, the apparent specific volumes at infinite dilution values were also determined. Some information regarding the segment–solvent and segment–segment interactions is obtained from these values.

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## 1. Introduction

Since mixture of {poly(ethylene glycol) dimethyl ether 2000 (PEGDME2000) + poly(propylene glycol) 400 (PPG400) + water} forms an aqueous two phase system, thus it may be used in bio-separation processes [1,2]. In this regard physical properties such as density, speed of sound and viscosity are useful. From these properties some information may also be obtained about segment–solvent and segment–segment interactions for this system.

Density and speed of sound of PPG400 aqueous mixtures at different temperatures and polymer concentration ranges have been reported by other workers [3]; but, experimental viscosity data for these solutions are not available in literature. In our previous work [4] density, sound velocity and viscosity of (PEGDME2000 + water) and {PEGDME2000 + poly(ethylene glycol) (PEG400) + water} at  $T = (293.15 \text{ to } 313.15)$  K have been studied completely. As far as we know, these physical properties for ternary system {PEGDME2000 + PPG400 + water} have not been reported in literature. Also for other ternary polymer systems, limited data are available [5] for these physical properties. Thus, the researchers felt the necessity to study these physical properties for aqueous ternary polymer solutions.

In this work, the binodal curves are obtained at two temperatures for aqueous two phase system of {PEGDME2000 + PPG400 + water}. Densities, speed of sounds, and viscosities of binary aqueous solution of PPG400 and ternary solution of {PEGDME2000 + PPG400 + water} in the single phase region were determined experimentally at  $T = (293.15, 298.15, 303.15, 308.15, \text{ and } 313.15)$  K. The excess molar volume ( $V_m^E$ ) and isentropic compressibility deviation ( $\Delta K_S$ ) were computed through density and speed of sound data for the studied binary and ternary solutions at all temperatures. These experimental ( $V_m^E$ ) and ( $\Delta K_S$ ) data of the investigated systems are correlated with modified Wilson [4] and polymer-NRTL [4,6,7] models at all temperatures. These models were presented in our previous work [4]. Also the Eyring-modified Wilson [8] and Eyring-NRTL [9] models were used to correlate the viscosity values of binary and ternary solutions.

## 2. Experimental

### 2.1. Materials

PEGDME2000 and PPG400 were purchased from Merck and Fluka, respectively. By using gel permeation chromatography (GPC, Agilent, USA), previously [10], the number average molar mass and polydispersity of the PEGDME2000 were determined to be  $2305 \text{ g} \cdot \text{mol}^{-1}$  and 1.0579, respectively. Double distilled water and deionized water were used. Densities, speed of sound and viscosity values for PPG400 are

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**Table 1**

A brief summary of the purity of the used materials and the comparison of density ( $d$ ), speed of sound ( $u$ ) and viscosity ( $\eta$ ) of the pure PPG400 at different temperatures with literature at atmospheric pressure.<sup>b</sup>

Chemical name	Source	Molecular formula	Mass fraction purity	Temperature (K)	$d \cdot 10^{-3}/(\text{kg} \cdot \text{m}^{-3})$		$u/(\text{m} \cdot \text{s}^{-1})$		$\eta \cdot 10^3/(\text{Pa} \cdot \text{s})$	
					Exp.	Lit.	Exp.	Lit.	Exp.	Lit.
Poly(propylene glycol) 400	Fluka	$\text{HO}(\text{CH}_2\text{CH}_2(\text{CH}_2\text{O})_n\text{H}$	>0.990	293.15	1.010180	1.007909 <sup>a</sup>	1386.9	1383.51 <sup>a</sup>	88.77	–
				298.15	1.006199	1.003929 <sup>a</sup>	1370.4	1366.70 <sup>a</sup>	64.44	–
				303.15	1.002184	0.999941 <sup>a</sup>	1353.8	1350.10 <sup>a</sup>	47.90	–
				308.15	0.998155	0.995937 <sup>a</sup>	1337.4	1333.64 <sup>a</sup>	36.42	–
				313.15	0.994127	0.991938 <sup>a</sup>	1321.3	1317.20 <sup>a</sup>	28.38	–
Poly(ethylene glycol dimethyl ether) 2000	Merck	$\text{CH}_3\text{O}(\text{CH}_2\text{CH}_2\text{O})_n\text{CH}_3$	>0.990	–	–	–	–	–	–	

<sup>a</sup> Taken from Ref. [3].

<sup>b</sup> Standard uncertainties  $\sigma$  for each variables are  $\sigma(d) = 3.0 \times 10^{-3} \text{ kg} \cdot \text{m}^{-3}$ ;  $\sigma(u) = 0.1 \text{ m} \cdot \text{s}^{-1}$ ;  $\sigma(T(d)) = 10^{-3} \text{ K}$ ;  $\sigma(\eta) = 0.01 \text{ mPa} \cdot \text{s}$ ;  $\sigma(t) = 0.01 \text{ s}$ ;  $\sigma(T(\eta)) = 0.01 \text{ K}$ .

given in Table 1 at different temperatures. These values are compared with the literature values.

## 2.2. Apparatus and procedures

The experimental apparatus employed for obtaining binodal curves is essentially similar to the one used in previous works [10,11]. Clouding point titration method was used to obtain the binodal curves [10]. A thermostat (HETO BIRKERØD, type: 01 TE 623, Denmark) was used to control the temperature within  $\pm 0.05 \text{ K}$  in this method. An analytical balance (Shimadzu, 321-34553, Shimadzu Co., Japan) with a precision of  $\pm 1 \cdot 10^{-7} \text{ kg}$  was used to calculate the composition of the mixture for each point on the binodal curve by mass. The maximum uncertainty in determining the mass fraction of both polymers by using this method was found to be  $\pm 0.002$ .

All solutions were prepared immediately before measurements by mass using an analytical balance (Shimadzu, 321-34553, Shimadzu Co., Japan) with precision ( $10^{-7} \text{ kg}$ ). To minimize the absorption of atmospheric moisture, all the solutions were kept tightly sealed.

A commercial density and sound velocity measurement apparatus (Anton Paar DSA 5000 densimeter and sound velocity analyzer) was used to measure the density and sound velocity of mixtures. In this apparatus the temperature was automatically kept constant within  $\pm 10^{-3} \text{ K}$  because both speed of sound and density are

extremely sensitive to temperature. The experimental uncertainty of density and ultrasonic velocity measurements were  $\pm 3.0 \times 10^{-3} \text{ kg} \cdot \text{m}^{-3}$  and  $\pm 0.1 \text{ m} \cdot \text{s}^{-1}$ , respectively. The calibration of this apparatus was performed at atmospheric pressure with double distilled deionized, and degassed water, and dry air.

A suspended level Ubbelohde type capillary viscometer was used to measure the viscosity of the solutions which was mounted in water thermostat (Julabo, MD-18 V, Germany) and was calibrated with water at five different temperatures,  $T = (293.15 \text{ to } 313.15) \text{ K}$ . An electronic stopwatch with a resolution of 0.01 s was applied to measure the flow time of a constant volume of liquid through capillary. For each solution an average of at least four readings of flow time was taken with a variation not exceeding  $\pm 0.1 \text{ s}$ . Controlling the temperature of the thermostat bath was within  $\pm 0.01 \text{ K}$ . The measured viscosities were found to be accurate within  $\pm 0.01 \text{ mPa} \cdot \text{s}$ . The dynamic viscosity,  $\eta$ , was calculated by the following relation:

$$\eta = dK(f - \theta) \quad (1)$$

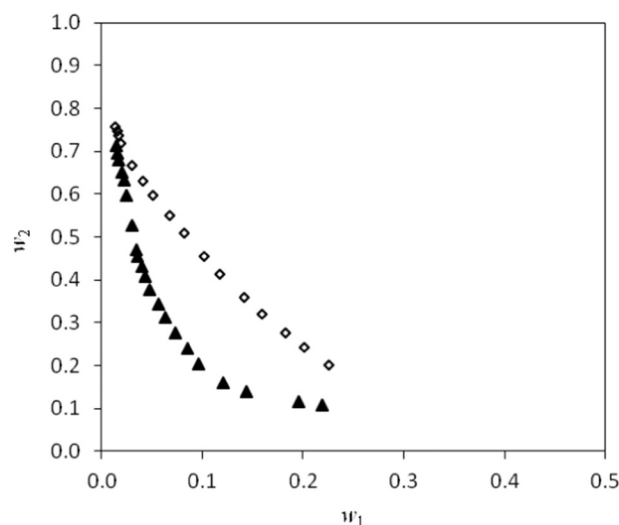
where  $f$ ,  $K$ ,  $\theta$  and  $d$  are the flow time, the viscometer constant, the Hagenbach correction factor and the density, respectively. Distilled water was used to determine the viscometer constant,  $K$ , by calibrating at working temperatures using viscosity values from the literature [12]. The values  $\eta = (1.002, 0.890, 0.797, 0.719 \text{ and } 0.653) \times 10^{-3} \text{ Pa} \cdot \text{s}$  [12] were used for viscosity of water at  $T = (293.15, 298.15, 303.15, 308.15 \text{ and } 313.15) \text{ K}$ , respectively. The dynamic viscosity determination uncertainty was estimated to be  $\pm 1\%$ .

**Table 2**

Experimental binodal data in mass fractions,  $w_i$ , for the {PEGDME2000 (1) + PPG400 (2) + Water (3)} system at  $T = (298.15 \text{ and } 313.15) \text{ K}$  at atmospheric pressure.<sup>a</sup>

$T/\text{K} = 298.15$		$T/\text{K} = 313.15$	
$100w_1$	$100w_2$	$100w_1$	$100w_2$
1.38	75.81	1.41	71.31
1.50	74.74	1.56	69.40
1.63	73.69	1.71	67.92
1.90	71.94	2.02	65.10
2.97	66.74	2.25	63.33
4.07	63.00	2.47	59.72
5.04	59.81	2.97	52.78
6.75	55.01	3.38	46.89
8.13	50.91	3.55	45.51
10.13	45.54	3.97	43.16
11.72	41.36	4.30	40.74
14.11	35.89	4.77	37.63
15.90	32.09	5.63	34.31
18.20	27.71	6.32	31.33
20.13	24.29	7.26	27.66
22.53	20.22	8.54	23.90
		9.65	20.35
		12.04	15.99
		14.41	13.87
		19.55	11.67
		21.86	10.73

<sup>a</sup> Standard uncertainties  $\sigma$  for each variables are  $\sigma(w_1) = \sigma(w_2) = 0.002$ ;  $\sigma(T) = 0.05 \text{ K}$ .



**Fig. 1.** Plot of the mass fraction of polymer (1) against the mass fraction of polymer (2) to show binodal curves for the (PEGDME2000 (1) + PPG400 (2) + water (3)) two-phase system:  $\diamond$ ,  $T = 298.15$ ;  $\blacktriangle$ ,  $T = 313.15$ .

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