

# Comparative Studies of the Mixing Effect on the Thermal Effusivity, Compressibility, and Molar Volume for Aqueous Solutions of Alcohols

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**Abstract** Concentration dependences of the thermal effusivity, isentropic compressibility coefficient, and molar volume were investigated experimentally for aqueous solutions of ethanol, 1-propanol, and 2-propanol. The thermal effusivity was determined using a photoacoustic method. The excess molar volume was found from measured densities, while the isentropic compressibility coefficient was calculated based on density and ultrasound velocity measurements. It has been shown that the dependence of the effusivity on concentration, expressed in mass fraction units, is nonlinear in the case of all the alcohols used. Moreover, the location of extreme deviations from linearity for the thermal effusivity,  $\Delta e$ , agrees well with that of characteristic points for the isentropic compressibility coefficient,  $\kappa_S$ , and the excess molar volume,  $V_m^E$ , as a function of the concentration.

**Keywords** Alcohols · Aqueous solutions · Isentropic compressibility coefficient · Molar volume · Photoacoustic · Thermal effusivity

## 1 Introduction

It is known that the physical and chemical properties of binary liquid mixtures may vary significantly by changing their composition. The dependence of many properties on concentration often reveal characteristic maxima, minima, or inflection points, which can be discussed in terms of interactions between components of the mixtures.

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Dedicated to Professor Dr. Stefan Ernst on the occasion of his 75th birthday.

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Let us assume that we mix two liquids, one of them consisting of molecules A and the second of molecules B. The liquids can be characterized by values  $p_A$  and  $p_B$ , respectively, of a selected physical parameter  $p$ . When the molecules do not show any long-range mutual interactions and the interactions between unlike neighbors ( $U_{AB}$ ) and like neighbors ( $U_{AA}$ ) and ( $U_{BB}$ ) are of the same average strength, the mixture is regarded as ideal. It is known that, for some physical parameters of ideal liquids, the linear additivity rule can be applied to calculate the value of the parameter  $p_{\text{mix}}$  for the mixture as a whole. For binary mixtures this rule can be written in a form,

$$p_{\text{mix}} = y_A p_A + (1 - y_A) p_B \quad (1)$$

Here,  $y_A$  is the concentration of liquid A. Obviously, to apply the additivity rule for a given parameter, one should use the proper type of the variable  $y$ .

The situation changes if one deals with a non-ideal mixture. A part of the mixture which consists of molecules associated due to molecular interactions can be considered as a new type of a liquid, AB, with its physical properties different from these of pure liquid A and liquid B. As demonstrated in the work [1], in this case one should expect non-linear dependence of the quantity  $p_{\text{mix}}$  on concentration,  $y_A$ .

In the present article, experimental investigations of the changes in isentropic compressibility, molar volume, and thermal effusivity of aqueous solutions of three monohydrous alcohols: ethanol, 1-propanol, and 2-propanol, accompanying solute concentration changes are reported. The alcohols contain CH, CH<sub>2</sub>, and CH<sub>3</sub> non-polar, hydrophobic groups which are not able to participate in O–H···O hydrogen bonding and hydrophilic OH groups, capable of forming hydrogen bonds with water. Due to the hydrophobic effect, dynamic structures similar to gas clathrate hydrates are created in the solutions. The structure consists of a hydrogen-bonded water network with cavities occupied by a non-polar solute's molecules [2]. The clathrate model is now widely accepted for a description of the hydrophobic hydration effect observed for alcohol–water solutions. It has been proved that, dependent on the alcohol type, the stoichiometry of the alcohol–water semi-clathrates corresponds to alcohol concentrations up to 0.05 mole fraction [3–5]. An increase of alcohol concentration leads to a decay of the clathrate structures because of an insufficient amount of water, and gradually hydrogen bonding effects become dominant over the hydrophobic ones [4,6].

In the last few decades photothermal detection methods have proved to be a useful tool for experimental determination of such thermal parameters as the thermal diffusivity or thermal effusivity for solid, liquid, or gaseous media (see, e.g., [7]). According to the following expressions, both parameters are defined by their dependence on three important physical quantities:

$$e = \sqrt{\rho k c_p} \quad (2)$$

$$a = \frac{k}{\rho c_p} \quad (3)$$

Here  $\rho$  is the density,  $k$  is the thermal conductivity, and  $c_p$  is the specific heat capacity of a sample.

Because of the progress in accuracy and precision of photothermal techniques, investigations of various subtle effects prove to be possible. One of such advanced applications seems to be the investigation of variations of thermal parameters accompanying structural changes of concentration in binary liquid mixtures [8–11].

In our previous work [11], the thermal-effusivity dependence on solute concentration has been investigated for a mixture of two liquids: ethylene glycol (component A) and diethylene glycol (component B). The compounds have similar molecular structure:  $\text{OH}(\text{CH}_2\text{-CH}_2\text{-O})_i\text{-H}$ , where  $i = 1$  for A and  $i = 2$  for B. Therefore, it is reasonable to assume that the mean strength of interactions between the like molecules AA, BB, and the unlike molecules, AB, of the solutions should be nearly the same. In other words, this solution seems to be a good model of an ideal liquid mixture. The experimental dependence of the thermal effusivity on concentration obtained in [11] proved to be well represented by Eq. 1, provided that the variable,  $y_A$ , was represented by the mass fraction,  $w_1$ , of a solute. Then, we assumed that any deviation of the thermal-effusivity dependence from the linear Eq. 1 is indicative of the effects resulting from interactions between liquid components of a mixture.

According to our knowledge, the thermal-effusivity dependence on concentration for liquid solutions cannot be predicted by any exact, theoretical model. In this article, we would like to show that, application of the empirical assumption mentioned above to the alcohol–water solutions under study leads to explicable conclusions.

## 2 Samples and Methods

Deionized water and chemical compounds as supplied were used for preparation of solutions. Ethanol (99.8%) was from POCH Poland, 1-propanol (99.5%) and 2-propanol (99.8%) were from Aesar. The mixtures were freshly made by weighing with an uncertainty of 0.0001 g and carefully stirred.

All experimental results needed to determine the quantities examined were obtained at 298.15 K. The density was measured by means of a DMA 602 (Anton Paar) oscillating tube density meter. The temperature of the density measurements was stabilized with an uncertainty of 0.02 K.

The ultrasound propagation speed was measured using an automatic velocity meter (Optel, Poland). The measurements consisted in determining the average time in which an acoustic signal with a frequency between 1 MHz and 10 MHz passed through the sample. The uncertainty of the temperature stabilization was in this case 0.05 K. Based on the density,  $\rho$ , and sound velocity,  $u$ , the isentropic compressibility,  $\kappa_S$ , was determined according to the well-known Laplace's formula,

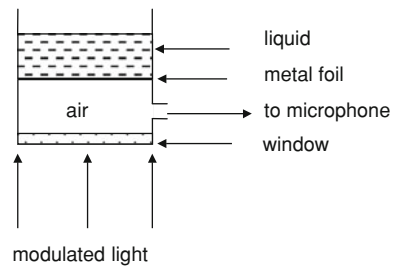
$$\kappa_S = \frac{1}{\rho u^2} \quad (4)$$

From the value of  $\rho$ , the excess molar volume was calculated using the following expression:

$$V_m^E = V_m - \left( x_1 \frac{M_1}{\rho_1} + x_2 \frac{M_2}{\rho_2} \right) \quad (5)$$

**Table 1** Density,  $\rho$ , and speed of ultrasound,  $u$ , for pure alcohols and water at  $T = 298.15$  K

Liquid	$\rho$ ( $\text{kg} \cdot \text{m}^{-3}$ )		$u$ ( $\text{m} \cdot \text{s}^{-1}$ )	
	This work	Literature	This work	Literature
Ethanol	785.08	785.13 [12], 784.93 [13], 785.04 [14]	1141.8	1142.6 [12], 1142.4 [17]
1-propanol	799.51	799.60 [13], 799.7 [14], 800.62 [15]	1206.0	1207.1 [12], 1205.42 [18], 1208.1 [15]
2-propanol	781.03	781.26 [13], 780.9 [14], 781.91 [15], 781.2 [16]	1139.5	1138.3 [12], 1141.3 [15], 1140.1 [16]
Water	–	997.01 [14]	–	1497.08 [19]

**Fig. 1** Geometry of the photoacoustic experiment

where subscripts 1 and 2 stand for the solute and water-solvent, respectively,  $V_m$  is the molar volume of a solution,  $M_1$  and  $M_2$  are the molar masses,  $\rho_1$  and  $\rho_2$  are the densities, while  $x_1$  and  $x_2$  are the mole fractions of a solute and water, respectively.

The experimental values of the density and speed of ultrasound for pure liquids are compared with reported values available in the literature in Table 1. The uncertainties in the measured values of the density were estimated to be  $5 \times 10^{-2} \text{ kg} \cdot \text{m}^{-3}$ , and for the speed of ultrasound,  $1 \text{ m} \cdot \text{s}^{-1}$ .

The photoacoustic (PA) technique was used for the thermal-effusivity experimental determination. The PA signal was generated in a PA cell due to the periodic heat flow from a sample, after absorption of periodically modulated light. Temperature changes in the air filling the cell caused periodic pressure changes which could be detected by the use of an acoustic receiver. In the experiment described, the PA signal amplitude was measured by means of an open PA cell [20] with microphonic registration. The experimental arrangement is schematically presented in Fig. 1. A detailed description of the experimental setup, together with values of material parameters necessary for determination of the effusivity from Eq. 2, are presented in our previous work [11].

It has been shown in the previous work [11] that the thermal effusivity of the liquid examined can be determined from the expression,

$$e_x = D \left[ \frac{1}{R_1} \sqrt{\left\{ \left( \frac{e_r}{D} + 1 \right)^2 + 1 - R_1^2 \right\}} - 1 \right] \quad (6)$$

**Table 2** Comparison of the experimental and calculated thermal effusivities for pure liquids at  $T = 298.15$  K

Liquid	$e_{\text{exp}}^a$ ( $\text{W} \cdot \text{s}^{0.5} \cdot \text{K}^{-1} \cdot \text{m}^{-2}$ )	$\rho$ ( $\text{kg} \cdot \text{m}^{-3}$ )	$c_p$ ( $\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ )	$k$ ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )	$e_{\text{calc}}^b$ ( $\text{W} \cdot \text{s}^{0.5} \cdot \text{K}^{-1} \cdot \text{m}^{-2}$ )	$\Delta e^c$ (%)
Water	–	997.048	4179.8	0.6062	1589	–
Ethanol	581	785.04	2437.7	0.167	565	2.8
1-propanol	549	799.7	2394.5	0.154	543	1.5
2-propanol	538	780.9	2604.2	0.135	523	3.8

<sup>a</sup>  $e_{\text{exp}}$ —value of thermal effusivity determined from PA measurements, this work

<sup>b</sup>  $e_{\text{calc}}$ —value of thermal effusivity calculated from Eq. 2. Values of density ( $\rho$ ), thermal conductivity ( $k$ ), and specific heat capacity ( $c_p$ ) have been taken from [14]

<sup>c</sup>  $\Delta e$  (%) =  $100(e_{\text{exp}} - e_{\text{calc}})/e_{\text{calc}}$

where  $R_1 = Q_x/Q_r$  is the ratio of photoacoustic amplitudes:  $Q_x$  is obtained in the case of the liquid mixture placed behind the metal foil,  $Q_r$  is measured with water behind the foil, and  $e_r$  is the thermal effusivity of water. The parameter  $D$  depends only on the properties of the metal foil used and is defined by

$$D = b_s^{-1} e_s l_s \quad (7)$$

Here,  $b_s$ ,  $e_s$ , and  $l_s$  are the thermal diffusion length, thermal effusivity, and the thickness of the thin metal foil closing the PA cell, respectively.

The uncertainty of the thermal effusivity deviation has been estimated as  $8 \text{ W} \cdot \text{s}^{0.5} \cdot \text{K}^{-1} \cdot \text{m}^{-2}$ . The total uncertainty for determination of the absolute value of effusivity is  $20 \text{ W} \cdot \text{s}^{0.5} \cdot \text{K}^{-1} \cdot \text{m}^{-2}$ .

### 3 Results and Discussion

The values of thermal effusivities for pure liquids obtained from the PA measurement agree well with those calculated from Eq. 2, as can be seen from Table 2.

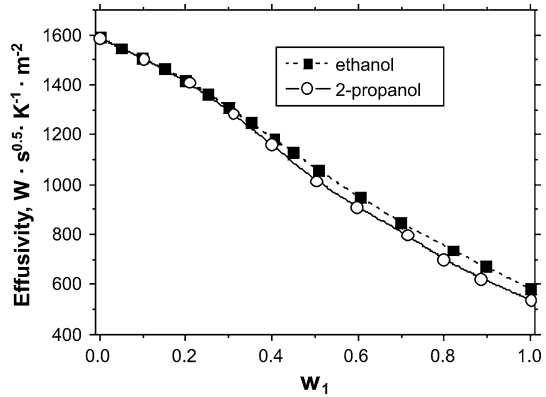
Figure 2 shows two examples of the experimental thermal-effusivity dependence on concentration which are evidently non-linear. In Table 3, the experimental values of thermal effusivities are presented for the solutions under investigation.

Based on the data collected in Table 3, the relative deviations of the effusivity from linearity,  $\Delta e/e_w$ , have been calculated from the expression,

$$\frac{\Delta e}{e_w} = \frac{e_{\text{exp}} - (w_1 e_1 + w_2 e_w)}{e_w} \quad (8)$$

Here,  $e_w$  is the thermal effusivity of pure water,  $e_{\text{exp}}$  is the experimentally determined effusivity for the solution examined,  $w_1$  is the mass fraction of the solute, and  $w_2$  is the mass fraction for the water solvent, while  $e_1$  is the effusivity for the pure solute. The selection of mass fraction as a concentration unit is purely empirical, as explained at the end of the Sect. 1. The dependences of the thermal-effusivity deviations for the three monohydrous alcohol–water solutions on the alcohol concentration are illustrated in Fig. 3.

**Fig. 2** Experimental dependence of the thermal effusivity on the mass fraction of the solute for ethanol–water and 2-propanol–water solutions. Points in this figure correspond to experimental data, line is “for eye” only. Remark is valid for all subsequent figures

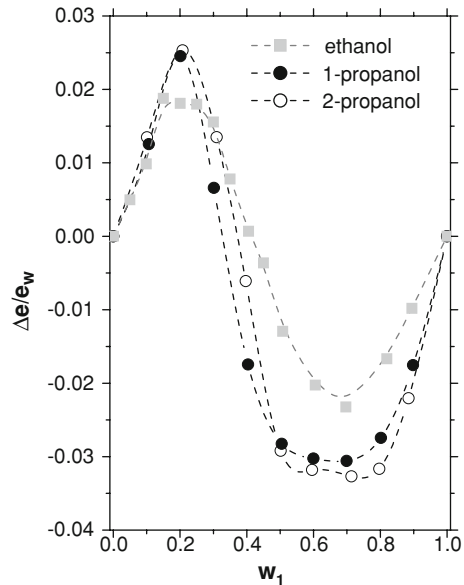


**Table 3** Values of experimental thermal effusivity,  $e$ , for the solutions investigated at  $T = 298.15$  K ( $w_1 =$  mass fraction of alcohol)

Ethanol–water		1-propanol–water		2-propanol–water	
$w_1$	$e$ ( $W \cdot s^{0.5} \cdot K^{-1} \cdot m^{-2}$ )	$w_1$	$e$ ( $W \cdot s^{0.5} \cdot K^{-1} \cdot m^{-2}$ )	$w_1$	$e$ ( $W \cdot s^{0.5} \cdot K^{-1} \cdot m^{-2}$ )
0	1589	0	1589	0	1589
0.0499	1546	0.1068	1498	0.1014	1504
0.0998	1505	0.2012	1419	0.2079	1411
0.1481	1468	0.3015	1286	0.3098	1285
0.2005	1416	0.4039	1141	0.3971	1162
0.2566	1366	0.5046	1019	0.5017	1015
0.3001	1311	0.5996	917	0.5957	911
0.3573	1249	0.6991	813	0.7135	800
0.4052	1182	0.8025	711	0.7967	701
0.4472	1130	0.8982	627	0.8846	624
0.5077	1057	1	549	1	538
0.6053	947				
0.6969	850				
0.8022	737				
0.8945	673				
1	582				

Ethanol is a compound with the chemical formula  $CH_3CH_2OH$ . This alcohol is known to reveal hydrophobic or hydrophilic behavior in water solutions, depending on the concentration. A distinct maximum of the effusivity deviation in the low concentration region (at  $w_1 = 0.22$ , i.e.,  $x_1 = 0.1$  mole fraction) and a minimum located at high concentrations ( $w_1 = 0.6$ , i.e.,  $x_1 = 0.4$ ) can be seen in Fig. 3. Such a shape of the effusivity dependence can be explained if one considers the long known fact that at low concentrations the specific heat capacity for this solution is higher than that for pure water [21]. According to the present state of knowledge, positive values

**Fig. 3** Relative deviation of thermal effusivity from linearity for ethanol (grey squares), 1-propanol (solid circles), and 2-propanol (empty circles) water solutions versus mass fraction of the solute



of the excess molar heat capacity at high dilution is a typical feature of hydrophobic interactions [2]. It may therefore be concluded that the maximum effusivity deviation for the ethanol–water solution should be assigned to hydrophobic interactions.

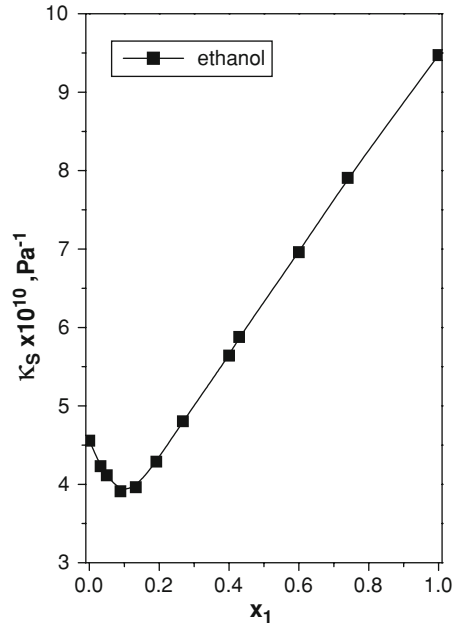
Another typical feature of alcohol solutions is the presence of a distinct minimum in the dependence of the isentropic compressibility on mole fraction [3]. It should be noted that the location of this minimum for ethanol–water solutions (at  $x_1 = 0.1$  in Fig. 4) agrees well with the location of the maximum deviation of the effusivity from linearity.

The high negative value of the excess molar volume of ethanol–water solutions (Fig. 5) can be considered as a manifestation of strong interactions between the solution components. The location of the minimum ( $x_1 = 0.4$ ) is determined by the concentration at which a significant change in the type of mutual interactions between components of the solution occurs. In this concentration region ( $0.3 < x_1 < 0.4$ ), a minimum of the deviation curve for the thermal conductivity of ethanol–water solutions is also observed [22, 23].

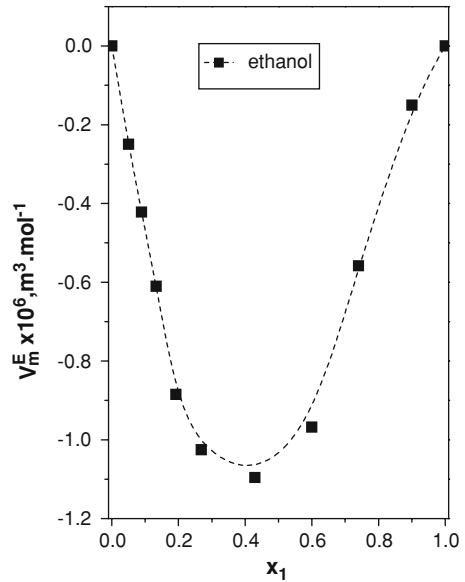
Summing up the knowledge collected about aqueous solutions of ethanol, a conclusion can be drawn that the dependence of the effusivity deviation from linearity may be accounted for mainly by changes in the volumetric heat capacity,  $\rho c_p$ , at low concentrations, and in the thermal conductivity at higher concentrations.

In the case of 1-propanol with a molecular structure  $\text{CH}_3(\text{CH}_2)_2\text{OH}$  and its isomer, 2-propanol with a molecular structure  $\text{CH}_3\text{CHCH}_3\text{OH}$ , the dependence on concentration of the three quantities investigated is similar, as seen from Figs. 3, 6, and 7. The experimentally obtained numerical data for the thermal effusivity, density, speed of sound, and isentropic compressibility are shown in Tables 3, 4, and 5, respectively. The isentropic compressibility for pure water,  $\kappa_S = 4.475 \times 10^{-10} \text{ Pa}^{-1}$  has been

**Fig. 4** Adiabatic compressibility coefficient for ethanol–water solution versus alcohol mole fraction



**Fig. 5** Excess molar volume for ethanol–water solution versus alcohol mole fraction

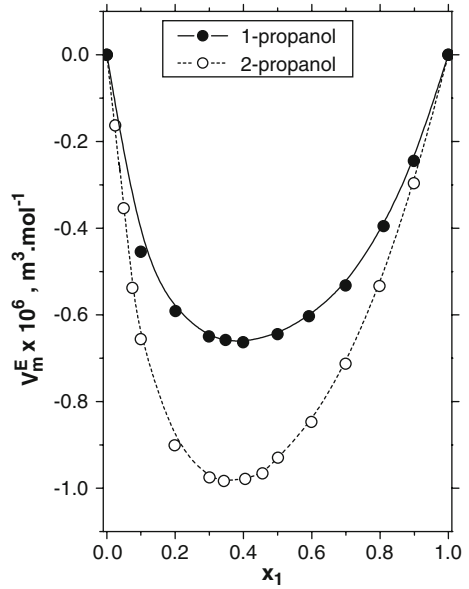


calculated from literature data for the water density and ultrasound velocity given in Table 1.

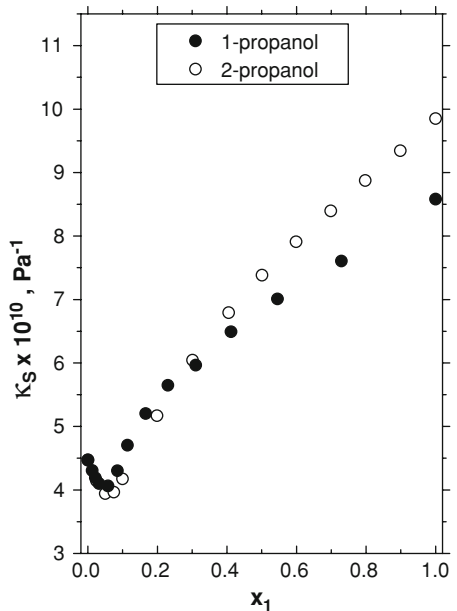
As in the case of ethanol, the locations of extremes on the plot of deviations from linearity for the effusivity agree with the location of minima of the adiabatic



**Fig. 6** Excess molar volume for 1-propanol–water and 2-propanol–water solutions versus mole fraction of the solute



**Fig. 7** Isentropic compressibility for 1-propanol–water and 2-propanol–water solutions versus mole fraction of the solute



compressibility coefficient and excess molar volume, respectively. Slightly deeper extremes are observed with 2-propanol than for 1-propanol for both  $\Delta e$  and  $V_m^E$ .

Table 6 gives a comparison between locations of extremes for the dependences investigated. As seen from Table 6, for the solutions examined, the values in column 2

**Table 4** Density ( $\rho$ ), speed of sound ( $u$ ), and isentropic compressibility ( $\kappa_S$ ) for 2-propanol–water mixtures at  $T = 298.15$  K ( $x_1$ —mole fraction)

2-propanol–water			
$x_1$	$\rho$ (kg · m <sup>-3</sup> )	$u$ (m · s <sup>-1</sup> )	$\kappa_S \times 10^{10}$ (Pa <sup>-1</sup> )
0.0124	988.58		
0.0250	984.03	1565.1	4.149
0.0400	977.88		
0.0504	973.96	1613.1	3.946
0.0625	969.45		
0.0754	964.44	1615.7	3.972
0.0872	959.54		
0.0999	953.85	1584.2	4.177
0.1985	913.67	1454.2	5.175
0.3011	881.32	1369.2	6.053
0.3434	870.46		
0.4052	856.47	1310.6	6.797
0.4556	846.54		
0.5011	838.39	1270.8	7.386
0.5986	823.48	1238.4	7.919
0.6986	810.65	1210.9	8.413
0.7980	799.72	1186.3	8.885
0.8992	789.89	1163.0	9.360
1	781.03	1139.5	9.860

agree with these in column 3 while those in column 5 are compatible with the values in column 6.

It should be also stressed that although the nature of the concentration effect is similar for all the solutions studied, the magnitude of the deviation for ethanol solutions is apparently smaller than that for the propanols (see Fig. 3), probably due to contributions from additional CH<sub>2</sub> (1-propanol) or CH<sub>3</sub>CH (2-propanol) groups.

Hence, generalizing the former conclusion for ethanol, the location of extremes in the plot of  $\Delta e/e_w$  may be attributed to changes in the volumetric heat capacity,  $\rho c_p$ , at low concentrations, and to changes in the thermal conductivity  $k$ , at higher concentrations.

## 4 Conclusions

It has been shown that the obtained positive deviations of the effusivity from linearity in aqueous solutions of alcohols at low concentrations can be assigned to hydrophobic interactions. Such interactions in alcohol solutions were confirmed in many research studies, among other things, by investigating volumetric [3], elastic [4], or dielectric [24] properties. As confirmed by mass and IR spectrometry [6,25], for higher con-

**Table 5** Density ( $\rho$ ), speed of sound ( $u$ ), and isentropic compressibility ( $\kappa_S$ ) for 1-propanol–water mixtures at  $T = 298.15$  K ( $x_1$ —mole fraction)

1-propanol–water			
$x_1$	$\rho$ (kg · m <sup>-3</sup> )	$u$ (m · s <sup>-1</sup> )	$\kappa_S \times 10^{10}$ (Pa <sup>-1</sup> )
0.0123	992.31	1530	4.305
0.0221	988.28	1554	4.190
0.0323	983.86	1575	4.097
0.0503	975.73	1593	4.039
0.0579	972.24	1592	4.058
0.0864	959.16	1535	4.427
0.1139	947.04	1499	4.699
0.1666	926.07	1442	5.193
0.2307	905.04	1397	5.662
0.3103	884.41	1376	5.972
0.4117	863.88	1336	6.485
0.5454	843.27	1300	7.017
0.7297	822.58	1263	7.621
1	799.51	1206	8.600

**Table 6** Location of extremes for the relative thermal effusivity deviation,  $\Delta e/e_w$ , in comparison with locations of the characteristic points for the excess molar volume,  $V_m^E$ , and isentropic compressibility coefficient,  $\kappa_S$ 

Solute	$\Delta e/e_w$ (maximum)		$\kappa_S$ (minimum)	$\Delta e/e_w$ (minimum)		$V_m^E$ (minimum)
	1	2	3	4	5	6
	$w_1$	$x_1$	$x_1$	$w_1$	$x_1$	$x_1$
Ethanol	0.22 <sup>a</sup>	0.10 <sup>b</sup>	0.1 <sup>a</sup> , 0.1 [18], 0.1 [3]	0.63 <sup>a</sup>	0.4 <sup>b</sup>	0.4 <sup>a</sup> , 0.35 [3]
1-propanol	0.19 <sup>a</sup>	0.06 <sup>b</sup>	0.05 <sup>a</sup> , 0.045 [18], 0.05 [4]	0.65 <sup>a</sup>	0.36 <sup>b</sup>	0.4 <sup>a</sup> , 0.4 [4]
2-propanol	0.21 <sup>a</sup>	0.07 <sup>b</sup>	0.06 <sup>a</sup>	0.68 <sup>a</sup>	0.39 <sup>b</sup>	0.37 <sup>a</sup>

<sup>a</sup> Present work, experiment<sup>b</sup> Recalculated from column on the left

centrations, above the concentration corresponding to the minimum for the effusivity deviation curve, processes related with the formation of hydrogen bonds with water and association of alcohol molecules are predominant. As seen in Table 6, the excess molar volume minima are located at the same concentration as those for the thermal effusivity. It seems therefore that, effusivity deviations from linearity are good parameters for evidence of such interactions between the molecules in water mixtures. Further investigations are in progress.

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