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Empirical viscous coefficients of dilute solutions of tetraethylene glycol dimethyl ether in 2-propanol and in 2-butanol at different temperatures

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Abstract

This paper reports experimental viscosities and densities of binary mixtures containing tetraethylene glycol dimethyl ether+(2-propanol, 2-butanol) at four different temperatures (288.15, 298.15, 308.15 and 318.15 K), over the mole fraction (x) range of tetraethylene glycol dimethyl ether between $0 \leq x \leq 0.1$ and at atmospheric pressure. The empirical coefficients (β_G, γ_G and $\Delta G_{1,2}^{*0}$) were determined from the viscosity values of the binary mixtures in the dilute zone of tetraethylene glycol dimethyl ether, using the Tamamushi–Isono empirical relationship between the thermodynamic function, $\Delta G_{1,2}^{*0}$, with the mole fraction of tetraethylene glycol dimethyl ether (x) at a particular temperature. The Jones–Dole coefficient (B) was calculated in a graphical way. Both contributions, the energetic and the volumetric ones, to coefficient (B) were obtained. A comparative study of the Jones–Dole coefficient B and the coefficient β_G of the solutions of tetraethylene glycol dimethyl ether in 2-alkanol with tetraethylene glycol dimethyl ether in 1-alkanol data, obtained from literature, was carried out. The results are discussed qualitatively using the rate process Eyring theory, applied to viscous flow.

Keywords: Viscosity; Density; Tetraethylene glycol dimethyl ether; 2-Propanol; 2-Butanol

1. Introduction

This work gives information about thermophysical properties of derivatives of ethylene glycol in aliphatic 2-alcohols. The mixtures of ethers + alcohols are interesting because they are used as gasoline additives [1]. Furthermore, these solutions could be used as new absorbent-refrigeration fluid pairs in absorption systems [2,3]. Thermophysical properties (densities and viscosities) of the binary mixture are needed to design heat absorption processes and to evaluate the performance of the working mixtures.

This paper gives information about the experimental viscosities and densities of binary dilute solutions of tetraethylene glycol dimethyl ether in 2-alkanol (2-propanol, or 2-butanol) at different temperatures, between 288.15 and 318.15 K, over the mole fraction (x) range of tetraethylene glycol dimethyl ether between $0 \leq x \leq 0.1$, at atmospheric pressure. Empirical coefficients, β_G, γ_G, $\Delta G_{1,2}^{*0}$ and energetic and volumetric contributions to Jones–Dole B coefficient are presented for different dilute solutions of the mentioned systems, using the rate process Eyring theory, applied to viscous flow.

2. Theory

From Eyring's theory, the average molar Gibbs free energy of activation for viscous flow of solution, $\Delta G_{1,2}^{*}$ (J mol^{-1}), is given [4] by:

$$\Delta G_{1,2}^{*} = RT\ln\left(\frac{V_{1,2}\eta_{1,2}}{hN}\right) \tag{1}$$

where R is the gas constant, T is the absolute temperature, $V_{1,2}$ is the mean molar volume of the solution, ($\eta_{1,2}$) is the dynamic viscosity of the solution, h is the Planck constant and N is the Avogadro constant.

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The dependence of activation parameters of viscous flow on the solute mole fraction, at a definite temperature, for liquid mixtures can be obtained from the empirical relation [5]:

$$\Delta G^{*}_{1,2} = \Delta G^{*\circ}_{1} + \beta_G x_2 + \gamma_G x_2^2 \qquad (2)$$

where $\Delta G^{*\circ}_{1,2}$ is the activation parameter for the pure solvent, β_G and γ_G are empirical coefficients.

The coefficient, β_G is defined as:

$$\beta_G = \left(\frac{\partial \Delta G^{*}_{1,2}}{\partial x_2}\right)_{x_2 \to 0} \qquad (3)$$

The relationship between the partial molar activation parameters of viscous flow at infinite dilution and the coefficient, β_G, is [6]:

$$\Delta G^{*\infty}_{2} = \Delta G^{*\circ}_{1} + \beta_G \qquad (4)$$

According to Eq. (4), the β_G coefficient of viscous flow can be considered as the difference between the solute partial molar activation parameter of viscous flow at infinite dilution and the corresponding partial molar property of the pure solvent [5].

The relative viscosity of a non-electrolyte solution can be given by [6,7]:

$$\eta_r = \frac{\eta_{1,2}}{\eta_1} = 1 + Bm + Dm^2 \qquad (5)$$

where m is the solution molality in mol kg^{-1} and coefficients B and D are empirical constants, characteristic of a given solute–solvent pair.

Table 1
Comparison of data with literature data for pure liquids at 298.15 K and at atmospheric pressure

Component	$\eta_{1,2}$ (mPa s)		ρ (g cm^{-3})	
	Experimental	Literature	Experimental	Literature
2-Propanol	2.089	2.098[a] 2.0436[b] 1.936[c] 2.045[d]	0.7809	0.78126[b] 0.7809[c] 0.7809[d]
2-Butanol	3.150	3.115[a] 2.998[b] 3.084[f]	0.8023	0.80241[b] 0.80240[e]
TEGDME	3.313	3.394[g]	1.0059	1.0063[g] 1.00653[h]

[a] From Ref. [13].
[b] From Ref. [11].
[c] From Ref. [14].
[d] From Ref. [15].
[e] From Ref. [17].
[f] From Ref. [16].
[g] From Ref. [18].
[h] From Ref. [19].

Table 2
Dynamic viscosity ($\eta_{1,2}$/mPa s) and density (ρ/g cm^{-3}) of dilute solution of DMETEG (2) in 2-PR (1) at different temperatures

x_2	m_2		288.15 K	298.15 K	308.15 K	318.15 K
0.0000	0.0000	$\eta_{1,2}$	2.840	2.089	1.564	1.192
		ρ	0.7891	0.7809	0.7725	0.7635
2.100×10^{-3}	0.0350	$\eta_{1,2}$	2.812	2.074	1.555	1.188
		ρ	0.7909	0.7827	0.7742	0.7653
4.200×10^{-3}	0.0702	$\eta_{1,2}$	2.787	2.059	1.549	1.185
		ρ	0.7920	0.7839	0.7755	0.7665
9.940×10^{-3}	0.1671	$\eta_{1,2}$	2.716	2.022	1.530	1.179
		ρ	0.7963	0.7881	0.7796	0.7706
0.0140	0.2363	$\eta_{1,2}$	2.679	1.999	1.520	1.177
		ρ	0.7988	0.7906	0.7820	0.7731
0.0152	0.2568	$\eta_{1,2}$	2.677	1.996	1.520	1.180
		ρ	0.8013	0.7931	0.7846	0.7757
0.0250	0.4267	$\eta_{1,2}$	2.568	1.934	1.512	1.180
		ρ	0.8060	0.7978	0.7892	0.7802
0.0355	0.6124	$\eta_{1,2}$	2.497	1.941	1.484	1.172
		ρ	0.8114	0.8031	0.7945	0.7855
0.0719	1.2891	$\eta_{1,2}$	2.429	1.860	1.465	1.163
		ρ	0.8332	0.8248	0.8162	0.8071
0.0734	1.3181	$\eta_{1,2}$	2.414	1.883	1.489	1.167
		ρ	0.8321	0.8237	0.8150	0.8061
0.0994	1.8365	$\eta_{1,2}$	2.372	1.830	1.444	1.159
		ρ	0.8442	0.8356	0.8269	0.8178

The B coefficient is mainly related to size and shape effects and also to solute–solvent interactions over the viscous flow. The coefficient D is related to solute–solute interactions, and also reflects that part of the solute solvent interactions which is not included in the viscosity coefficient, B.

Considering Eyring's theory of rate process applied to viscous flow, the relationship between B and β_G [6,8] is:

$$B = \rho_1(V_1^{\circ} - \bar{V}_2^{\infty}) + \frac{\beta_G M_1}{RT} \qquad (6)$$

where $\bar{V}_2^{\infty}$ is the partial molar volume of the solute at infinite dilution, V_1° is the molar volume, ρ_1 is the

Table 3
Dynamic viscosity ($\eta_{1,2}$/mPa s) and density (ρ/g cm^{-3}) of dilute solution of TEGDME (2) in 2-BU (1) at different temperatures

x_2	m_2		288.15 K	298.15 K	308.15 K	318.15 K
0.0000	0.0000	$\eta_{1,2}$	4.691	3.150	2.179	1.566
		ρ	0.8103	0.8023	0.7939	0.7851
1.80×10^{-3}	0.02433	$\eta_{1,2}$	4.574	3.082	2.157	1.554
		ρ	0.8113	0.8033	0.7948	0.7861
4.60×10^{-3}	0.06235	$\eta_{1,2}$	4.464	3.032	2.132	1.544
		ρ	0.8127	0.8046	0.7962	0.7874
0.0146	0.19989	$\eta_{1,2}$	4.221	2.910	2.069	1.515
		ρ	0.8177	0.8097	0.8012	0.7924
0.0257	0.35587	$\eta_{1,2}$	4.003	2.787	2.024	1.493
		ρ	0.8222	0.8142	0.8057	0.7969
0.0738	1.07499	$\eta_{1,2}$	3.346	2.439	1.834	1.408
		ρ	0.8425	0.8343	0.8257	0.8169
0.0988	1.47907	$\eta_{1,2}$	3.160	2.338	1.777	1.368
		ρ	0.8520	0.8437	0.8351	0.8263

Table 4
Coefficients of Eq. (8) and standard deviations for density of 2-PR + TEGDME and 2-BU + TEGDME systems

	288.15 K	298.15 K	308.15 K	318.15 K
2-PR + TEGDME system				
$a_0 \pm \sigma$	$0.7894 \pm 5 \times 10^{-4}$	$0.7812 \pm 5 \times 10^{-4}$	$0.7728 \pm 5 \times 10 - 4$	$0.7638 \pm 5 \times 10^{-4}$
$a_1 \pm \sigma$	0.70 ± 0.03	0.70 ± 0.03	0.70 ± 0.03	0.70 ± 0.03
$a_2 \pm \sigma$	-1.5 ± 0.3	-1.6 ± 0.3	-1.5 ± 0.3	-1.5 ± 0.3
σ (g cm^{-3})	8.4×10^{-4}	8.4×10^{-4}	9.4×10^{-4}	8.4×10^{-4}
2-BU + TEGDME system				
$a_0 \pm \sigma$	$0.8106 \pm 1 \times 10^{-4}$	$0.8026 \pm 1 \times 10^{-4}$	$0.7941 \pm 1 \times 10^{-4}$	$0.7854 \pm 1 \times 10^{-4}$
$a_1 \pm \sigma$	$0.464 \pm 5 \times 10^{-3}$	$0.461 \pm 5 \times 10^{-3}$	$0.459 \pm 5 \times 10^{-3}$	$0.459 \pm 5 \times 10^{-3}$
$a_2 \pm \sigma$	-0.43 ± 0.02	-0.43 ± 0.03	-0.43 ± 0.02	-0.43 ± 0.02
σ (g cm^{-3})	3.4×10^{-4}	3.4×10^{-4}	3.4×10^{-4}	2.4×10^{-4}

density and M_1 is the molecular weight of pure solvent, respectively.

3. Experimental section

3.1. Materials

2-Propanol (2PR) and 2-butanol (2BU), Biopack pro-analysis, were carefully dried with molecular sieves and purity was verified by GC. Tetraethylene glycol dimethyl ether (TEGDME), Sigma, >99% was used without later purification and was also kept over molecular sieves. Their properties were checked with the published recent values. The values are included in Table 1.

3.2. Equipment

Kinematic viscosity measurement of pure liquids and their mixtures were made with an automatic Schott Gerate AVS 400 viscosity-measuring system equipped with a series of Ubbelhode viscosimeters. In all cases, the experiments were generally performed at least in five replicates for each composition and at each temperature and the results were averaged. A thermostated bath constant to 0.02 K was used and the temperatures were read from calibrated thermometers. The overall experimental error in kinematic viscosity was approximately ± 0.003 cSt.

Dynamic viscosity was calculated with the following equation:

$$\eta = k(t_{\mathrm{m}} - f)\rho \qquad (7)$$

where t_{m} is time, k is the viscosimeter constant, f is the Hagenbach correction factor, and ρ is the density.

Densities were determined with an AP digital densimeter, model DMA 45. Calibration was carried out with air and doubly distilled water. The estimated error in the density measurement was approximately $\pm 2 \times 10^{-4}$ g/cm^3.

All weightings were made on a Mettler H20 T balance and the estimated error in mole fraction was $\pm 1.4 \times 10^{-4}$.

4. Results and discussion

The dynamic viscosity ($\eta_{1,2}$) and the density (ρ) values of dilute solutions of tetraethylene glycol dimethyl ether in 2-propanol are included in Table 2. The dynamic viscosity ($\eta_{1,2}$) and the density (ρ) data of dilute solutions of tetraethylene glycol dimethyl ether in 2-butanol (2-BU) are listed in Table 3.

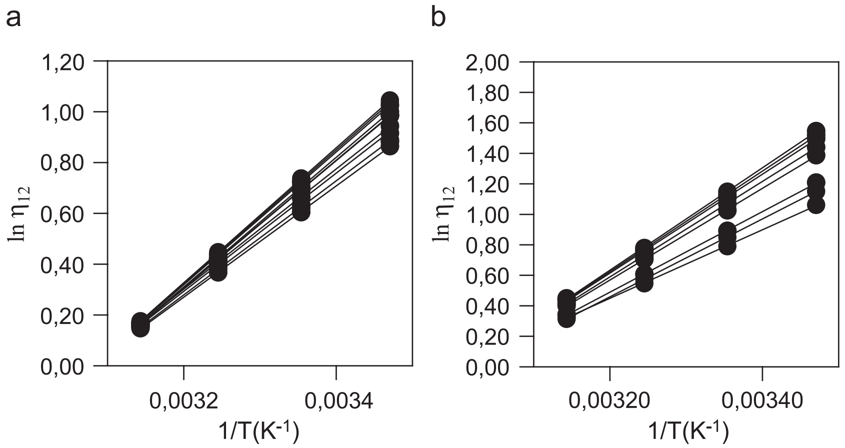

Fig. 1. ln $\eta_{1,2}$ values vs. $1/T$ for the mixture of 2-PR + TEGDME (a) and 2-BU + TEGDME (b), in the studied range of temperature at different mole fraction.

Table 5
Activation energy for viscous flow for 2-PR + TEGDME and 2-BU + TEGDME

2-PR (1) + TEGDME (2)			2-BU (1) + TEGDME (2)		
x_2	ΔE^* (kJ/mol)	σ (ΔE^*) (kJ/mol)	x_2	ΔE^* (kJ/mol)	σ (ΔE^*) (kJ/mol)
0.0000	22.06	± 0.04	0.0000	27.9	± 0.2
2.1×10^{-3}	21.89	± 0.04	1.8×10^{-3}	27.4	± 0.2
4.2×10^{-3}	21.71	± 0.03	4.6×10^{-3}	27.0	± 0.2
9.9×10^{-3}	21.20	± 0.03	0.0146	26.0	± 0.2
0.0140	20.89	± 0.01	0.0257	25.0	± 0.2
0.0152	20.81	± 0.05	0.0738	22.0	± 0.2
0.0250	19.7	± 0.2	0.0988	21.2	± 0.1
0.0355	19.3	± 0.3	0.1979	18.7	± 0.2
0.0719	18.7	± 0.1			
0.0734	18.4	± 0.3			
0.0994	18.2	± 0.1			

The dependence of density on mole fraction of tetraethylene glycol dimethyl ether was studied with a polynomial equation, for the 2-PR + TEGDME and 2-BU + TEGDME systems.

$$\rho(x_2) = \sum_{i=0}^{n} a_i x_2^i \tag{8}$$

The results of this correlation procedure are listed in Table 4, together with the standard deviations at each investigated temperature, for 2-PR + TEGDME and 2-BU + TEGDME systems, respectively.

Fig. 1 shows the graphic of ln $\eta_{1,2}$ vs. $1/T$ for the mixture of 2-PR + TEGDME and 2-BU + TEGDME, in the studied range of temperature at different mole fraction (see Tables 2 and 3). Full lines match the values obtained by fitting experimental data by Arrhenius equation

$$\eta_{1,2} = A e^{-\Delta E^*/RT} \tag{9}$$

where A is a constant and ΔE^* is the activation energy for viscous flow, R is the gas constant, and T is the temperature.

Table 6
The β_G, γ_G and $\Delta G^{*\,0}_{1,2}$ parameters of Eq. (2) at different temperatures

(kJ mol^{-1})	288.15 K	298.15 K	308.15 K	318.15 K
2-PR + TEGDME				
β_G	− 5.3 ± 0.8	− 1.7 ± 0.9	1.4 ± 1.4	3.6 ± 0.3
$\Delta G^{*0}_{1,2}$	15.07 ± 0.01	14.85 ± 0.01	14.60 ± 0.01	14.43 ± 0.01
γ_G	55 ± 8	30 ± 9	13 ± 8	3 ± 3
2-BU + TEGDME				
β_G	− 11.89 ± 1.0	− 8.5 ± 0.8	− 4.3 ± 0.5	− 1.1 ± 0.5
$\Delta G^{*0}_{1,2}$	16.69 ± 0.01	16.3 ± 0.01	15.96 ± 0.01	15.63 ± 0.01
γ_G	59 ± 10	47 ± 8	26 ± 5	11 ± 5

The ln $\eta_{1,2}$ values of the 2-PR + TEGDME and 2-BU + TEGDME systems decrease when the temperature increases and decrease too when the molar fraction increases.

It can be seen in Tables 2 and 3 that the values of dynamic viscosity decrease while the mole fraction of tetraethylene glycol dimethyl ether increases. All the dilute solutions of 2-PR + TEGDME and 2-BU + TEGDME systems have less viscosity than the pure alcohol (solvent). It is possible that the solute (TEGDME) destroys the solvent structure, the bonds between solvent molecules are broken, then the viscosity of the solution is smaller than that of the solvent [9]. This behaviour was also observed in dilute solutions of TEGDME in 1-alcohols [10]. If the viscosity of the solution is higher than that of the solvent, it would indicate an increase in the net structure of the solvent, in this case the solute–solvent bonds can be formed [9].

The dynamic viscosity values increase with the molar volumes of the solvent, then the mixture values for 2-PR + TEGDME are lower than the values for 2-BU + TEGDME.

From Table 5, it can be seen that the values of activation energy of viscous flow of 2-PR + TEGDME system are smaller than the values of the 2-BU +TEGDME system. In both systems, the values of activation energy are smaller than the values of the pure alcohol, since the energy of activation for viscous flow is related to the work required to form a hole in the liquid, the experimentally

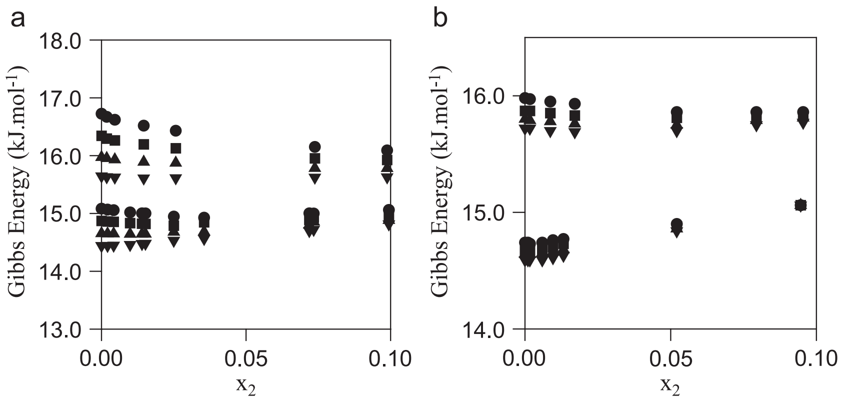

Fig. 2. $\Delta G^*_{1,2}$ vs. x_2 of the systems: (a) 2-PR + TEGDME and 2-BU + TEGDME; (b) 1-PR + TEGDME and 1-BU + TEGDME; ● 288.15 K, ■ 298.15 K, ▲ 308.15 K, ▼ 318.15 K.

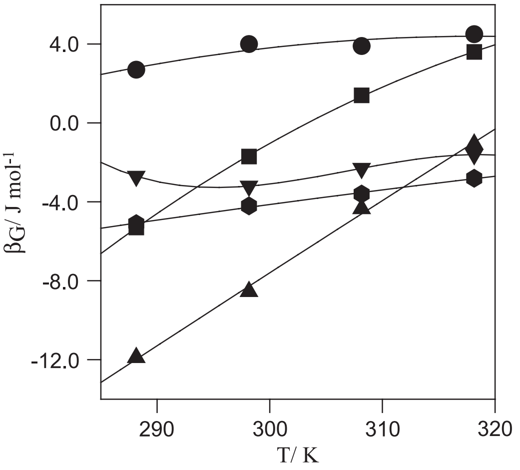

Fig. 3. β_G parameters for systems: 2-PR + TEGDME (■), 2-BU + TEGDME (▲), 1- PR + TEGDME (●), 1-BU + TEGDME (▼), and 1-PE + TEGDME (◆).

observed activation energy could mean that the work decreases while the mole fraction of tetraethylene glycol dimethyl ether increases. The values of activation energy for viscous flow of the pure solvents are some fractions of the energy of vaporisation, considering Eyring's theory of rate process [4] applied to viscous flow, and can be represented by equation: $\Delta E^* = \Delta H/n$, where $n = 2.0$ for 2-PR and $n = 1.8$ for 2-BU at 298.15 K, then the activation energy increases with the molecular weights of the solvent. The values of the energy of vaporisation were taken from Ref. [11].

The $\Delta G^*_{1,2}$ values have been calculated from experimental dynamic viscosity of dilute solutions of TEGDME in 2-alkanol using Eq. (1). Fig. 2 shows the plot of $\Delta G^*_{1,2}$ vs. x_2 of dilute solutions of tetraethylene glycol dimethyl ether in 2-alkanol and the solutions of TEGDME in 1-alkanol, data were obtained from Ref. [11]. The change of $\Delta G^*_{1,2}$ values with the mole fraction is greater in 1-propanol than in 2-propanol and the behaviour is different. The values of $\Delta G^*_{1,2}$ of the 2-butanol + TEGDME system are larger than the $\Delta G^*_{1,2}$ values of 1-butanol + TEGDME, but the behaviour is similar.

The β_G, γ_G and $\Delta G^{*0}_{1,2}$ parameters of TEGDME in 2-alkanol, listed in Table 6, were calculated from the Tamamushi–Isono empirical relation [4]. Fig. 3 shows the β_G values vs. temperature for the system of TEGDME+(2-PR, or 2-BU) and the β_G values taken from literature for the systems of TEGDME in three 1-alkanols (1-propanol (1-PR), 1-butanol (1-BU) and 1-pentanol (1-PE)) [12]. The β_G values for TEGDME in 2-alkanol and in 1-alkanol increase with the temperature, but the change of the β_G values of TEGDME in 2-alkanol is larger than in 1-alkanols.

According to Eq. (4), β_G is equal to the difference: $\Delta G^{*\infty}_2 - \Delta G^{*0}_1$. The solute partial molar activation parameter of viscous flow at infinite dilution does not contain any contributions from the changes of solvent–solvent interactions caused by the molecule of solute depending only on the solute–solvent interactions. Thus, if $\Delta G^{*\infty}_2 < \Delta G^{*0}_1$, it could be said that the solute–solvent bonds are lighter or similar than the solvent–solvent bonds in the transition state, according to the vicosity transition state theory, and the solution viscosity can be smaller than that of the pure solvent.

The Jones–Dole coefficient (B) has been obtained from a plot of $(\eta_r - 1)/m$ vs. molality (m), when $m \rightarrow 0$. With the β_G values at each temperature, the second term on the right hand in Eq. (6) was calculated and through the difference with respect to B, the contribution due to the molar volume differences was obtained.

A plot of B coefficient vs. T and also the contributions of both volumetric and energetic terms to B for the 2-PR + TEGDME and 2-BU + TEGDME systems can be seen in Fig. 4.

All the values of the B coefficient of TEGDME + 2-PR and TEGDME + 2-BU are negative and increase when the temperature increases. The negative B coefficient shows a break solvent structure. The B coefficients decrease with increase in the size and molecular weights of the alcohol. The contribution corresponding to the molar volume differ-

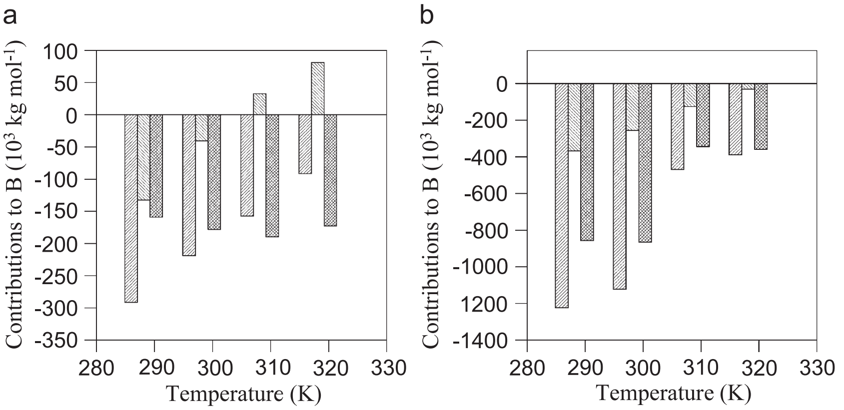

Fig. 4. The Jones–Dole B coefficients (▨), $M\beta_G/RT$ term (▧) and the difference term (▩) at different temperatures for the systems: 2-PR + TEGDME (a) and 2-BU + TEGDME (b).

ence of 2-PR + TEGDME is almost constant with the temperature and the values are negative. The values of 2-BU + TEGDME increase with the temperature and all of them are negative. The energetic contribution terms of the 2-PR + TEGDME system are of a different sign when the temperature increases, but in the 2-BU + TEGDME all the values are negative. The energetic contribution terms increase when the temperature increases in both systems. In addition, from Fig. 4, it may be concluded that the contribution corresponding to the molar volume difference is more important than the energetic contribution terms to *B* coefficient values.

5. Conclusions

Dilute solutions of 2-PR + TEGDME and 2-BU + TEGDME systems have less viscosity than the pure alcohol (solvent). It is possible that the solute (TEGDME) destroys the solvent structure, the bonds between solvent molecules are broken, then the viscosity of the solution is smaller than that of the solvent. The contribution corresponding to the molar volume difference is more important than the energetic contribution terms to *B* coefficient values in both studied systems.

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