Excess Properties of Binary Mixtures of 2-Methoxy-2-methylpropane with Ethanol at Different Temperatures

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Densities and viscosities of the binary system 2-Methoxy-2-methylpropane + ethanol were measured at 288.15, 293.15, 298.15, 303.15, 308.15 K and atmospheric pressure, over the whole composition range. The excess values of molar volume, viscosity and Gibbs free energy of activation of viscous flow were calculated from experimental measurements. The excess functions of the binary systems were fitted to Redlich-Kister equation. Hind, Grunberg-Nissan, Heric-Brewer, Jouyban-Acree and McAllister models have been used to calculate viscosity coefficients and these were compared with experimental data for the mixtures. Also, the activation energies of viscous flow have been obtained and their variations with compositions have been discussed.

Keywords: binary liquid mixtures, excess properties, 2-methoxy-2-methylpropane, ethanol

The present work is part of our systematic studies on thermodynamic properties for mixtures of great interest in industry. This paper reports densities and viscosities of binary liquid mixtures of 2-Methoxy-2-methylpropane with ethanol at 288.15, 293.15, 298.15, 303.15 and 308.15 K as a function of composition. Knowledge of these mixing properties has relevance in both theoretical and applied areas of research because such results are useful in design and simulation processes. In this sense, there has been an increasing interest in the thermodynamic behaviour of liquid mixtures of the oxygenated compounds included in reformulated gasoline: 2-Methoxy-2-methylpropane (MTBE), *tert*-amyl methyl ether (TAME) and alcohols: methanol, ethanol, 2-Methyl-2-propanol and isobutyl alcohol [1-8].

Experimental part

The chemical MTBE was obtained from Merck (>99.5 mol%) and ethanol was supplied by Chemical Company (>99.3 mol%). The chemicals were dried over molecular sieves (Fluka type 4 A°). The purity was checked through chromatographic and refractive index methods. The mole fractions were determined by weighing and precision of the mole fraction was ± 0.00005. The densities were determined by hydrostatic weighing method of Kohlrausch with the precision of ± 0.00005g/cm^3. The experimental technique has been previously described [9], the temperature of thermostatic water bath being controlled to ± 0.05 K. Viscosities of the pure compounds and of the binary mixtures were determined with an Ubbelohde kinematic viscometer [10] that was kept in a vertical position in a water thermostat. A thermostatically controlled bath (constant to ± 0.05 K) was used. The kinematic viscosity was calculated using the relation:

$$\nu = At - B / t \qquad (1)$$

where:

t is the flowing time (± 0.1s) of a constant volume liquid through the viscometer capillary

A and *B* - characteristic constants of the used viscometer, which were determined by taking water and benzene as the calibrating liquids for correction of kinetic energy deviations.

The dynamic viscosity was determined from the equation:

$$\eta = \nu\rho \qquad (2)$$

where ρ is the density of the liquid. The precision of the viscosity was estimated to be ± 0.0010 mPa·s. In all determinations, triplicate experiments were performed at each composition and temperature, and the arithmetic mean was taken for the calculations of the viscosity.

Results and discussions

The measured densities and viscosities of the pure component liquids present good agreement with the literature values, as seen in table 1.

Densities and viscosities of the binary mixtures of MTBE + ethanol are reported in table 2. The results of this study are in close agreement with the works of Hoga and Tôrres [1] with regard to the system composed of MTBE with ethanol in the temperature interval of 293.15-308.15 K. No experimental data were found for the present mixtures at 288.15 K so a comparison of our values could be made.

The excess molar volumes were calculated from the densities of the pure liquids and their mixtures using the following equation:

$$V^E = [xM_1 + (1-x)M_2]/\rho - [xM_1/\rho_1 + (1-x)M_2/\rho_2] \qquad (3)$$

where *x* and (1-*x*) are the mole fraction of the components, M_1 and M_2 are the molecular masses of the components 1 and 2, and ρ, ρ_1 and ρ_2 are the respective densities of the solution and of the pure components.

The experimental values of viscosity (η) are used to calculate the excess viscosity (η^E) defined by the equation:

$$\eta^E = \eta - [x\eta_1 + (1-x)\eta_2] \qquad (4)$$

where η_1 and η_2 are the viscosities of pure components.

The excess Gibbs free energy ΔG^{*E} was determinated from the following equation:

$$\Delta G^{*E} = RT[\ln \eta V - (x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2)] \qquad (5)$$

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Component	Temperature (K)	Density ($g\cdot cm^{-3}$)		Viscosity (mPa·s)	
		Exp.	Lit.	Exp.	Lit.
MTBE	288.15	0.7458	0.7457[11]	0.3905	-
	293.15	0.7408	0.7404[11] 0.74065[12]	0.3706	0.3861[13]
	298.15	0.7357	0.7357[8] 0.7356[15] 0.7354[12]	0.3550	0.3354[14] 0.3687[13]
	303.15	0.7304	0.7301[12]	0.3387	0.3490[16]
	308.15	0.7243	0.7248[12]	0.3214	0.3800[17]
Ethanol	288.15	0.7958	-	1.3652	-
	293.15	0.7895	0.78945[18]	1.2206	1.1940[19]
	298.15	0.7857	0.7850[20]	1.1088	1.105[20] 1.0826[21]
	303.15	0.7816	0.7810[22] 0.7808[23]	1.0056	0.9950[23] 1.0102[24]
	308.15	0.7770	0.7762[25]	0.9222	0.8810[25]

Table 1
COMPARISON OF EXPERIMENTAL DENSITIES AND VISCOSITIES OF PURE LIQUIDS WITH LITERATURE VALUES

x	ρ (g/cm^3) Temperature (K)					η (mPa·s) Temperature (K)				
	288.15	**293.15**	**298.15**	**303.15**	**308.15**	**288.15**	**293.15**	**298.15**	**303.15**	**308.15**
0.1005	0.7899	0.7839	0.7799	0.7759	0.7714	1.0872	0.9969	0.9179	0.8281	0.8281
0.1986	0.7840	0.7782	0.7738	0.7693	0.7642	0.8954	0.8227	0.7525	0.7021	0.7021
0.3015	0.7777	0.7721	0.7677	0.7630	0.7576	0.7485	0.6892	0.6470	0.6050	0.6050
0.3979	0.7723	0.7668	0.7623	0.7575	0.7520	0.6355	0.5966	0.5512	0.5213	0.5213
0.4969	0.7677	0.7623	0.7576	0.7526	0.7470	0.5628	0.5246	0.4911	0.4654	0.4654
0.5968	0.7628	0.7576	0.7527	0.7477	0.7420	0.5021	0.4722	0.4430	0.4176	0.4176
0.7005	0.7584	0.7532	0.7483	0.7432	0.7376	0.4576	0.4357	0.4067	0.3854	0.3854
0.8024	0.7543	0.7495	0.7446	0.7394	0.7335	0.4251	0.3999	0.3787	0.3577	0.3577
0.9005	0.7504	0.7454	0.7403	0.7351	0.7294	0.4004	0.3791	0.3581	0.3446	0.3446

Table 2
DENSITIES AND VISCOSITIES OF THE BINARY MIXTURES OF MTBE (x) + ETHANOL AT DIFFERENT TEMPERATURES

where V, V_1 and V_2 are the respective molar volumes of the mixture and of the pure components.

The excess functions of the binary systems can be represented by a Redlich-Kister type equation:

$$X^E = x_i x_j \sum_{k=0}^{3} A_k (2x_i - 1)^k \qquad (6)$$

where:

X^E represents any of the following properties:

V^E, η^E, ΔG^{*E};

A_k

x_i, x_j are the mole fractions of the components i and j, respectively;

denotes the polynomial coefficients.

The values of these coefficients are indicated in table 3 along with the standard deviation, σ, defined by:

$$\sigma = \left[\frac{\sum (X^E_{obs.} - X^E_{calc.})^2}{m-n} \right]^{1/2} \qquad (7)$$

where m is the number of data points and n is the number of estimated parameters.

The variation of the excess molar volume with the mole fraction of ether is presented in figure 1. We can notice that the excess molar volumes are negative in the whole composition range and at all temperatures. Negative values of excess molar volume were associated with hydrogen bonding and complex formation between ether (non-self-associating component) and alcohol (strong self-associating component) as well as with structural effects [1,26-29] . The effect of temperature on the V^E shows a systematic decrease with temperature for all the mixtures. The excess viscosities are negative over the entire range of mole fractions at all the temperatures as shown in figure 2. The negative excess viscosity was explained by many authors through different forms [5,30-32]. By the addition of MTBE molecules, the alcohol molecules dissociate and

Temperature (K)	Coefficients	V^E (cm^3/mol)	η^E (mPa·s)	ΔG^{*E} (cal/mol)
288.15	A_0	-2.3234	-1.2717	-480.6936
	A_1	0.0335	0.5899	25.1114
	A_2	-1.2864	-0.2845	-13.5347
	A_3	-0.2717	0.0644	-73.9839
	σ	0.0159	0.0032	2.6523
293.15	A_0	-2.3679	-1.0873	-450.4483
	A_1	0.0098	0.5151	42.1903
	A_2	-1.4737	-0.1671	10.1989
	A_3	-0.4341	-0.1317	-198.5108
	σ	0.0168	0.0016	2.2512
298.15	A_0	-2.3888	-0.9708	-454.1618
	A_1	0.0412	0.4277	9.7594
	A_2	-1.5756	-0.1346	-3.5632
	A_3	-0.5350	-0.1539	-215.3652
	σ	0.0178	0.0049	4.3409
303.15	A_0	-2.3974	-0.8331	-419.7282
	A_1	-0.0396	0.2689	-90.7514
	A_2	-1.7573	-0.1571	-41.3265
	A_3	-0.2380	0.1034	36.4296
	σ	0.0196	0.0029	3.2097
308.15	A_0	-2.4337	-0.7445	-410.1345
	A_1	-0.2688	0.2418	-85.7152
	A_2	-2.2197	-0.1493	-31.9160
	A_3	0.0521	0.1556	135.1949
	σ	0.0227	0.0098	3.6545

Table 3
COEFFICIENTS A_k AND STANDARD DEVIATIONS AT DIFFERENT TEMPERATURES

have greater mobility than the pure alcohols due the reduced cohesive forces of alcohol molecules upon mixing [33-35]. If the temperature increases the negative values of excess viscosity decrease. This can be explained by breaking hydrogen bonds and increasing mobility of molecules due of the increase of thermal energy [1].

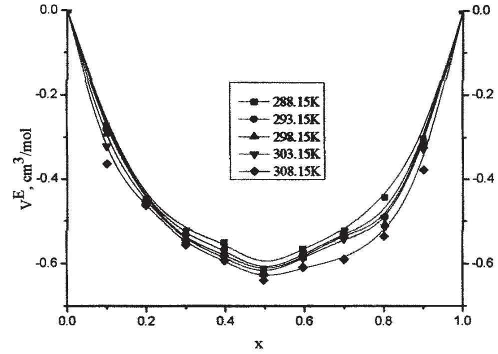

Fig. 1. Molar excess volumes of the MTBE (x) + ethanol system at different temperatures

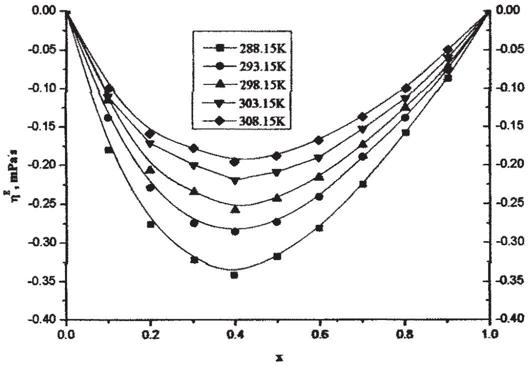

Fig. 2. Excess viscosities of the MTBE (x) + ethanol system at different temperatures

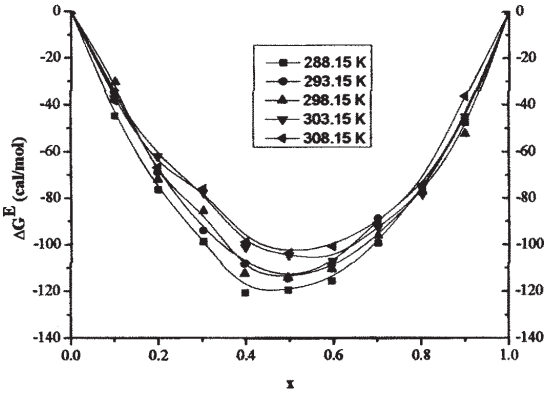

Fig. 3. Molar excess free energies of viscous flow activation of the MTBE (x) + ethanol

Figure 3 shows the dependence of the calculated ΔG^{*E} values upon composition. The excess Gibbs free energy ΔG^{*E} is considered a reliable means of detecting the interaction between molecules [36, 37]. The negative ΔG^{*E} values (fig. 3) suggest the dominance of dispersion interaction between MTBE and ethanol molecules. This type of behaviour has also been observed in other systems [8].

In an attempt to describe the viscosity of binary mixtures we checked the approach proposed by Hind [38]:

$$\eta = x_1^2\eta_1 + x_2^2\eta_2 + 2x_1x_2\eta_{12} \qquad (8)$$

Grunberg-Nissan[39]:

$$\ln\eta = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1x_2d \qquad (9)$$

Heric-Brewer [40]:

$$\begin{aligned}\ln\eta &= x_1\ln\eta_1 + x_2\ln\eta_2 + x_1\ln M_1 + x_2\ln M_2 - \\ &-\ln(x_1M_1 + x_2M_2) + x_1x_2[\alpha_{12} + \alpha_{21}(x_1 - x_2)]\end{aligned} \qquad (10)$$

Jouyban Acree [41,42]:

$$\ln\eta = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1 + x_2\sum\left(\frac{A_j(x_1 - x_2)^j}{T}\right) \qquad (11)$$

The dynamic viscosity of the liquid mixture is designated by η, and η_1, η_2 are the dynamic viscosities, x_1, x_2 are the mole fractions of the pure components 1 and 2 constituting the liquid mixture, M_1, M_2 are the molecular weights, T is the temperature; η_{12}, d, α_{12}, α_{21} and A_j are interaction parameters (viscosity coefficients) and reflect the non-ideality of the system.

We tested the three-body McAllister equation [43]:

$$\begin{aligned}\ln\eta &= x_1^3\ln\eta_1 + 3x_1^2x_2\ln\eta_{12} + 3x_1x_2^2\ln\eta_{21} + x_2^3\ln\eta_2 - \\ &-\ln[x_1 + x_2M_2/M_1] + 3x_1^2x_2\ln[(2 + M_2/M_1)/3] + \\ &+3x_1x_2^2\ln[(1 + 2M_2/M_1)/3] + x_2^3\ln[M_2/M_1]\end{aligned} \qquad (12)$$

and the four-body McAllister equation:

$$\begin{aligned}\ln\eta &= x_1^4\ln\eta_1 + 4x_1^3x_2\ln\eta_{1112} + 6x_1^2x_2^2\ln\eta_{1122} + \\ &+4x_1x_2^3\ln\eta_{2221} + x_2^4\ln\eta_2 - \ln[x_1 + x_2M_2/M_1] + \\ &+4x_1^3x_2\ln[(3 + M_2/M_1)/4] + 6x_1^2x_2^2\ln[(1 + M_2/M_1)/2] + \\ &+4x_1x_2^3\ln[(1 + 3M_2/M_1)/4] + x_2^4\ln[M_2/M_1]\end{aligned} \qquad (13)$$

The McAllister model adjustable parameters are given by η_{12}, η_{21}, η_{1112}, η_{1122} and η_{2221}. The parameters that appear in equations 8-13 were estimated using the experimental viscosity data and a non-linear regression analysis employing the Levenberg-Marquardt algorithm. Table 4 shows the parameters calculated and the standard deviations between experimental values and those obtained using the semi-empirical relations. The data show that there are no important differences between the values obtained with Grunberg-Nissan, Heric-Brewer, Jouyban Acree and McAllister models. However, from the different equations that calculate viscosities of mixtures the Heric-Brewer and the three-body McAllister models show the best agreement with experimental data.

The energies of activation of viscous flow for the binaries studied were calculated using the following equations [44]:

$$\eta = \frac{hN}{V}e^{\frac{\Delta G^{\neq}}{RT}} \qquad (14)$$

$$\Delta G^{\neq} = \Delta H^{\neq} - T\Delta S^{\neq} \qquad (15)$$

where $\Delta G^{\neq}$, $\Delta H^{\neq}$ and $\Delta S^{\neq}$ are the free energy, enthalpy and entropy of activation of viscous flow.

The plots of ln η vs $1/T$ were found to be linear in the temperature range 288.15 – 308.15 K, as such the values of $\Delta H^{\neq}$ and $\Delta S^{\neq}$ were obtained by the corresponding slopes and the intercepts. With $\Delta G^{\neq}$ and ΔS values as input in eq. (15), the corresponding values of $\Delta G^{\neq}$ were also calculated. The values of activation energies are listed in table 5 as a function of composition. The value of $\Delta H^{\neq}$ for ethanol is more than two times higher than that for MTBE, indicating that association and dipole-dipole interactions increase the value of $\Delta H^{\neq}$. The value of $\Delta S^{\neq}$ is negative for MTBE and positive for ethanol and shows that overall molecular order due to activated complex formation increases for ether (nonassociating component) but

Table 4
PARAMETERS FOR THE SEMI-EMPIRICAL RELATIONS OF HIND, GRUNBERG-NISSAN, HERIC-BREWER, JOYBAN-ACREE AND MCALLISTER AND STANDARD DEVIATIONS AT DIFFERENT TEMPERATURES

Temperature (K)	Equation					
	Hind	Grunberg-Nissan	Heric-Brewer	Jouyban-Acree	McAllister three body model	McAllister four body model
288.15	$\eta_{12}=0.22084$ $\sigma=0.048$	$d=-1.06587$ $\sigma=0.0061$	$\alpha_{12}=-0.85729$ $\alpha_{21}=0.00498$ $\sigma=0.0051$	$A_0=-306.1974$ $A_1=26.57368$ $A_2=-6.01668$ $A_3=-37.15695$ $\sigma=0.0055$	$\eta_{12}=0.4273$ $\eta_{21}=0.64224$ $\sigma=0.0051$	$\eta_{1112}=0.41569$ $\eta_{1122}=0.523$ $\eta_{2221}=0.7689$ $\sigma=0.0055$
293.15	$\eta_{12}=0.23928$ $\sigma=0.037$	$d=-0.99234$ $\sigma=0.0063$	$\alpha_{12}=-0.78388$ $\alpha_{21}=-0.0366$ $\sigma=0.0067$	$A_0=-291.7331$ $A_1=35.33309$ $A_2=-6.50953$ $A_3=-99.48859$ $\sigma=0.0044$	$\eta_{12}=0.40178$ $\eta_{21}=0.60862$ $\sigma=0.0067$	$\eta_{1112}=0.39542$ $\eta_{1122}=0.4856$ $\eta_{2221}=0.72484$ $\sigma=0.0072$
298.15	$\eta_{12}=0.23626$ $\sigma=0.029$	$d=-0.9902$ $\sigma=0.0097$	$\alpha_{12}=-0.78193$ $\alpha_{21}=-0.09959$ $\sigma=0.0094$	$A_0=-295.1487$ $A_1=19.34083$ $A_2=-0.21825$ $A_3=-107.7064$ $\sigma=0.0087$	$\eta_{12}=0.37051$ $\eta_{21}=0.57508$ $\sigma=0.0094$	$\eta_{1112}=0.36633$ $\eta_{1122}=0.45726$ $\eta_{2221}=0.67487$ $\sigma=0.0101$
303.15	$\eta_{12}=0.244$ $\sigma=0.024$	$d=-1.2886$ $\sigma=0.0277$	$\alpha_{12}=-0.72182$ $\alpha_{21}=-0.12586$ $\sigma=0.0056$	$A_0=-279.3901$ $A_1=-30.51563$ $A_2=-18.49643$ $A_3=17.54495$ $\sigma=0.0062$	$\eta_{12}=0.35152$ $\eta_{21}=0.54593$ $\sigma=0.0056$	$\eta_{1112}=0.34354$ $\eta_{1122}=0.44413$ $\eta_{2221}=0.62515$ $\sigma=0.0057$
308.15	$\eta_{12}=0.23855$ $\sigma=0.023$	$d=-0.90139$ $\sigma=0.0067$	$\alpha_{12}=-0.693$ $\alpha_{21}=-0.06026$ $\sigma=0.0069$	$A_0=-276.1155$ $A_1=-26.6554$ $A_2=-12.08444$ $A_3=65.4502$ $\sigma=0.0071$	$\eta_{12}=0.34031$ $\eta_{21}=0.5002$ $\sigma=0.0069$	$\eta_{1112}=0.33245$ $\eta_{1122}=0.41487$ $\eta_{2221}=0.57557$ $\sigma=0.0074$

Table 5
VALUES OF $\Delta G^{\neq}$, $\Delta H^{\neq}$AND $\Delta S^{\neq}$ FOR THE BINARY MIXTURE OF MTBE (x)+ETHANOL

x	$\Delta G^{\neq}$ (cal/mol) Temperature (K)					$\Delta H^{\neq}$ (cal/mol)	$\Delta S^{\neq}$ (cal/mol·K)
	288.15	**293.15**	**298.15**	**303.15**	**308.15**		
0.0000	3025.74	3021.87	3017.99	3014.12	3010.24	3249.01	0.77
0.1005	2955.63	2955.55	2955.46	2955.38	2955.29	2960.56	0.02
0.1986	2890.55	2894.50	2898.44	2902.38	2906.32	2663.38	-0.79
0.3015	2834.68	2844.93	2855.18	2865.43	2875.68	2244.04	-2.05
0.3979	2785.61	2797.23	2808.84	2820.46	2832.07	2116.26	-2.32
0.4969	2754.12	2767.39	2780.65	2793.92	2807.18	1989.65	-2.65
0.5968	2728.40	2743.28	2758.17	2773.06	2787.95	1870.41	-2.98
0.7005	2715.20	2731.05	2746.91	2762.77	2778.62	1801.37	-3.17
0.8024	2704.36	2721.41	2738.46	2755.51	2772.55	1721.86	-3.41
0.9005	2704.56	2726.70	2748.84	2770.97	2793.11	1428.75	-4.43
1.0000	2719.89	2742.20	2764.52	2786.84	2809.16	1433.72	-4.46

decreases in case of ethanol due to the breaking of H-bonds. The values $\Delta H^{\neq}$and $\Delta G^{\neq}$ are positive for all the binary mixtures while the values of $\Delta S^{\neq}$are negative for all binary mixtures except for the mixture concentrated in ethanol. The positive $\Delta S^{\neq}$ value shows less overall molecular order due to activated complex formation for viscous flow.

Conclusions

The densities and viscosities of binary mixtures of MTBE + ethanol were measured experimentally at 288.15 K, 293.15 K, 298.15 K, 303.15 K and 308.15 K over the entire composition range. From these results, the excess values of molar volume, viscosity and Gibbs free energy of activation of viscous flow have been computed and fitted to the Redlich-Kister equation. The energies of activation of viscous flow for these binaries mixtures were also calculated and were discussed. Hind, Grunberg-Nissan, Heric-Brewer, Jouyban Acree and McAllister models have been used to calculate viscosity coefficients and these were compared with experimental data for the mixtures. Our results reveal that there are no important differences between the values obtained with Grunberg-Nissan, Jouyban Acree and the four-body McAllister models. The Heric-Brewer and the three-body McAllister models show the best agreement with experimental data.

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