# Densities, Viscosities and Excess Properties of Binary Mixtures of 2-Methoxy-2-methylpropane with iso-Propanol

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Densities ( $\rho$ ) and viscosities ( $\eta$ ) of the binary systems 2-Methoxy-2-methylpropane with iso-propanol were measured at temperatures (288.15, 293.15, 298.15, 303.15 and 308.15) K and atmospheric pressure, over the whole composition range. The density and viscosity of the solutions were correlated with the temperature with a linear equation, respectively with Guzman's equation. The excess values of molar volume ( $V^{E}$ ), viscosity  $(\eta^{E})$  and molar Gibbs energy ( $\Delta G^{*E}$ ) were calculated from experimental measurements. The excess functions of the binary systems were fitted to Redlich-Kister equation. Viscosity results were fitted to the equations of Grunberg-Nissan, Heric-Brewer, Jouyban-Acree and McAllister.

Keywords: binary liquid mixtures, excess properties, 2-Methoxy-2-methylpropane, iso-propanol

Experimental liquid densities and viscosities of pure hydrocarbons and their mixtures are useful in design and simulation processes. The physicochemical properties play an important role in the understanding of several industrial processes [1]. Therefore, experimental measurements are needed to understand the fundamental behaviour of these properties and then to develop new models [2]. This study reports densities and viscosities of binary liquid mixtures of 2-Methoxy-2-methylpropane (MTBE) with iso-propanol at temperatures (288.15, 293.15, 298.15, 303.15 and 308.15) K as a function of composition.

#### **Experimental part**

The chemicals MTBE (mole fraction purity > 0.995) was obtained from Merck and iso-propanol was supplied by Chemical Company (mole fraction purity > 0.997). The chemicals were dried over molecular sieves (Fluka type 4 A). The purity was checked through chromatographic analysis. The mole fractions were determined by weighing and precision of the mole fraction was  $\pm 0.00005$ .

The densities were measurement by hydrostatic weighing method of Kohlrausch [3]. The precision of the density was estimated to be  $\pm$  0.00005 g cm<sup>3</sup>. The temperature of thermostatic water bath being controlled to  $\pm$  0.05 K. Viscosities of the pure components and of the binary mixtures were measurement with an Ubbelohde kinematic viscometer [4]. Temperature constancy was achieved by using a thermostatically controlled bath (constant to  $\pm$  0.05 K). The kinematic viscosity was determined using the relation: (1)

v =

where *t* (s) is the flowing time of a constant volume liquid through the viscometer capillary. Accuracy of time measurement was  $\pm$  0.01s. A and B are viscometer constants and were determined from measurements with the calibration fluids. The dynamic viscosity was calculated from the following equation:

where  $\rho$  is the density of the liquid. The precision of the viscosity was estimated to be  $\pm$  0.002 mPas.

η =

## **Results and discussions**

The measured densities and viscosities of the pure component liquids are compared with the literature values and given in table 1. MTBE densities values reported in the literature differ from our experimental data with a maximum of 0.07%. For iso-propanol, densities values published in the literature differ by less than 0.2% than our results. Viscosity values reported in the literature differ than our results with a maximum 3% for MTBE and with maximum 0.6% for iso-propanol. Densities and viscosities of the binary mixtures of MTBE (x) + *iso*-propanol are reported in table 2.

Densities of the binary mixtures were correlated with temperature using the following equation [15]:

$$\rho = a_0 + a_0 T \tag{3}$$

where  $a_0$ ,  $a_1$  are the equation parameters and T is the temperature.

Component	T/K	ρ/(g·cm <sup>-3</sup> )		η/(mPa·s)		
		Experimental	Literature	Experimental	Literature	
	288.15	0.7458	0.7457[5]	0.3905	-	
MTBE	293.15	0.7408	0.74065[6]	0.3706	0.3711[7]	
	298.15	0.7357	0.7357[8]	0.3550	0.3560[7]	
	303.15	0.7304	0.7301[6]	0.3387	0.3490[9]	
	308.15	0.7243	0.7248[6]	0.3214	0.3245[10]	
	288.15	0.7912	-	2.7622	-	
iso-Propanol	293.15	0.7871	0.78535[11]	2.4064	2.4140[11]	
	298.15	0.7832	0.78270[12]	2.0546	2.0436[13]	
	303.15	0.7789	0.77712[11]	1.7908	1.7850[11]	
	308.15	0.7745	0.77288[11]	1.5605	1.5510[14]	

Table 1 EXPERIMENTAL AND LITERATURE VALUES FOR DENSITIES (p) AND VISCOSITIES  $(\eta)$  OF THE PURE **COMPONENTS** 

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Table 2
DENSITIES ( $\rho$ ) AND VISCOSITIES ( $\eta$ ) OF THE BINARY MIXTURES OF MTBE (x) + iso-PROPANOL AT DIFFERENT
TEMPERATURES AND ATMOSPHERIC PRESSURE

	ρ /(g·cm <sup>-3</sup> )				η /(mPa·s)					
x		T /K				T /K				
	288.15	293.15	298.15	303.15	308.15	288.15	293.15	298.15	303.15	308.15
0.1007	0.7860	0.7821	0.7783	0.7742	0.7700	1.8579	1.6615	1.4361	1.2741	1.1289
0.1995	0.7807	0.7766	0.7725	0.7684	0.7641	1.3470	1.2007	1.0672	0.9695	0.8726
0.3030	0.7757	0.7715	0.7675	0.7628	0.7579	1.0056	0.9196	0.8267	0.7608	0.6993
0.3981	0.7708	0.7667	0.7626	0.7579	0.7527	0.8111	0.7494	0.6921	0.6387	0.5856
0.4991	0.7664	0.7620	0.7577	0.7530	0.7476	0.6727	0.6194	0.5781	0.5407	0.4958
0.5992	0.7621	0.7575	0.7529	0.7480	0.7427	0.5739	0.5289	0.4956	0.4663	0.4445
0.6993	0.7581	0.7534	0.7486	0.7437	0.7381	0.5142	0.4771	0.4554	0.4228	0.4012
0.8005	0.7538	0.7496	0.7446	0.7396	0.7339	0.4586	0.4374	0.4159	0.3909	0.3660
0.8987	0.7498	0.7458	0.7410	0.7358	0.7299	0.4300	0.4128	0.3769	0.3669	0.3313

The temperature dependence of the viscosities of pure component and binary mixtures is expressed using Guzman's equation [15]:

$$\eta = \eta_0 e^{\frac{E_a}{RT}} \tag{4}$$

where  $\eta_{a}$  and  $E_{a}$  are the adjustable parameters.

The adjustable parameters of these equations were estimated using the experimental data and a nonlinear regression analysis employing the Levenberg-Marquardt algorithm [16]. Table 3 shows the fitting parameters along with the standard deviation calculated with equation:

$$\sigma = \left[\frac{\Sigma (X_{exp} - X_{calc})^2}{m - n}\right]^{1/2}$$
(5)

where X is the value of the analysed property, m is the number of data points, and *n* is the number of estimated parameters.

The small values of the standard deviation show that the equations 3 and 4 correctly estimate the variation of the density and the viscosity of the solutions with the temperature.

## The viscosity data correlation

Several empirical and semi-empirical relations have been used to represent the dependence of viscosity on concentration of components in binary systems:

(1) Grunberg-Nissan [17]:

$$ln\eta = x_1 ln\eta_1 + x_2 ln\eta_2 + x_1 x_2 d$$
(6)

Heric-Brewer [18]: (2)

$$ln\eta = x_1 ln\eta_1 + x_2 ln\eta_2 + x_1 lnM_1 + x_2 lnM_2 -$$

$$-ln(x_1M_1 + x_2M_2) + x_1x_2[\alpha_{12} + \alpha_{21}(x_1 - x_2)]$$
<sup>(7)</sup>

(3) McAllister four body model [19]:

$$ln\eta = x_1^4 ln\eta_1 + 4x_1^3 x_2 ln\eta_{1112} + + 6x_1^2 x_2^2 ln\eta_{1122} + 4x_1 x_2^3 ln\eta_{2221} + x_2^4 ln\eta_2 - ln[x_1 + x_2(M_2/M_1)] + 4x_1^3 x_2 ln[(3 + M_2/M_1)/4] +$$

 $+ 6x_1^2x_2^2ln[(1 + M_2/M_1)/2] + 4x_1x_2^3ln[(1 + 3M_2/M_1)/4] +$ 

$$+x_{2}^{4}ln[M_{2}/M_{1}]$$

(8)

## (4) Jouyban Acree [20,21]:

 $ln\eta = x_1 ln\eta_1 + x_2 ln\eta_2 + x_1 x_2 \sum_{j=0}^{n} \left(\frac{A_j (x_1 - x_2)^j}{\tau}\right)$ (9) In these equations  $\eta$ , and  $\eta_1$ ,  $\eta_2$  are the dynamic viscosities of the liquid mixtures and of the pure components 1 and 2,  $x_1$ ,  $x_2$  are the mole fractions,  $M_1$ ,  $M_2$ are the molecular masses, T is the temperature;  $d, \alpha_{12}$ ,  $\alpha_{21}$ ,  $\eta_{1112}$ ,  $\eta_{1122}$ ,  $\eta_{2221}$  and  $A_j$  are interaction parameters (viscosity coefficients) and reflect the non-ideality of the system.

The parameters that appear in equations 6-9 were estimated using the experimental viscosity data and a nonlinear regression analysis employing the Levenberg-Marquardt algorithm [16]. Table 4 shows the parameters calculated and the standard deviations between experimental values and those obtained using the semiempirical relations, calculated with equation 5. The data show that no model can not correctly estimate the excess viscosity at all temperatures investigated. A comparison of the calculated and the experimental viscosities shows that the Jouyban - Acree model produces accurate results for 288.15 K, 293.15 K, 303.15 K and 308.15 K, while for 298.15 K the Heric-Brewer model is the best to describe viscosities.

## Excess properties

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

x	<i>a</i> 0	a1·10 <sup>4</sup>	$\sigma/\text{g-cm}^{-3}$	η0 <sup>.</sup> 10 <sup>4</sup>	Ea/kJ-mol <sup>-1</sup>	$\sigma/mPas$
0.0000	1.03104	-8.32	0.00017	4.06	21.15	0.014
0.1007	1.01604	-7.98	0.00015	8.20	18.52	0.015
0.1995	1.01933	-8.28	0.00007	16.7	16.04	0.005
0.3030	1.03124	-8.86	0.00034	35.1	13.56	0.005
0.3981	1.03048	-9.00	0.00044	56.9	11.89	0.004
0.4991	1.03522	-9.32	0.00040	69.1	10.97	0.004
0.5992	1.04065	-9.66	0.00027	106.8	9.53	0.004
0.6993	1.04474	-9.94	0.00031	113.9	9.12	0.003
0.8005	1.04126	-9.96	0.00047	150.4	8.20	0.004
0.8987	1.03742	-9.96	0.00065	90.0	9.28	0.008
1.0000	1.05382	-10.7	0.00039	205.3	7.06	0.001

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#### Table 3

PARAMETERS IN EQS. 3 and 4 FOR DENSITY AND VISCOSITY DATA AND STANDARD DEVIATION OF MTBE (x) + iso-PROPANOL

1	Equation						
T/K	runberg-Nissan	Heric-Brewer	McAllister four	Jouyban-Acree			
8			body model				
			$\eta_{1112} = 0.48784$	$A_0 = -506.42166$			
8	d=-1.74376	$\alpha_{12} = -1.67009$	$\eta_{1122} = 0.56258$	A1=150.95522			
288.15	$\sigma = 0.0452$	$a_{21} = 0.56991$	$\eta_{2221} = 0.9742$	$A_2 = 28.56382$			
8		$\sigma = 0.0075$	$\sigma = 0.0071$	$A_3 = 47.92936$			
				$\sigma = 0.0070$			
			$\eta_{1112} = 0.47681$	$A_0 = -501.92068$			
	d=-1.67321	$a_{12} = -1.59955$	$\eta_{1122} = 0.48989$	$A_1 = 112.9452$			
293.15	$\sigma = 0.0455$	$a_{21} = 0.55266$	$\eta_{2221} = 0.92034$	$A_2 = 80.62897$			
		$\sigma = 0.0148$	$\sigma = 0.0119$	$A_3 = 155.98116$			
				$\sigma = 0.0081$			
			$\eta_{1112} = 0.42089$	$A_0 = -467.0135$			
	d = -1.57252	$\alpha_{12} = -1.49889$	$\eta_{1122} = 0.51649$	$A_1 = 151.960/2$			
298.15	$\sigma = 0.04123$	$\alpha_{21} = 0.50921$	$\eta_{2221} = 0.78359$	$A_2 = -12.05233$			
		$\sigma = 0.0107$	$\sigma = 0.0115$	$A_3 = 7.93651$			
		1 40000		$\sigma = 0.0120$			
1	7-10200	$a_{12} = -1.40889$	$\eta_{1112} = 0.40309$	A0=-401.20005			
202.15	a = -1.9588	$a_{21} = 0.40000$	$\eta_{1122} = 0.47010$	A1-94.44007			
505.15	0 - 0.0008	0 - 0.0088	$\eta_{2221} = 0.73700$	A2-13.24908			
			0 - 0.0094	$\sigma = 0.0055$			
			#1110 = 0.35422	$4_0 = -424.5406$			
8	d = .140241	$a_{12} = -1.32889$	$\eta_{112} = 0.33422$ $\eta_{112} = 0.48450$	$A_1 = 140, 20022$			
308.15	$\sigma = 0.0321$	$a_{21} = 0.37334$	$\eta_{1122} = 0.64639$	$A_2 = -71,79359$			
500.15	0 0.0521	a = 0.0135	$\sigma = 0.0120$	$A_3 = -10572996$			
		0 0.0155	0 0.0120	$\sigma = 0.0119$			

 $V^{\rm E} = \left[ xM_1 + (1-x)M_2 \right] / \rho - \left[ xM_1 / \rho_1 + (1-x)M_2 / \rho_2 \right]$ (10)

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  $\rho$ ,  $\rho_1$  and  $\rho_2$  are the densities of the solution and respective of the pure components.

The experimental values of viscosity  $(\eta)$  are used to calculate the excess viscosity  $(\eta^{E})$  defined by the equation:

$$\eta^{\rm E} = \eta - \left[ x \eta_1 + (1 - x) \eta_2 \right] \tag{11}$$

where  $\eta_1$  and  $\eta_1$  are the viscosities of pure components. The excess molar Gibbs energies of activation of viscous

flow,  $\Delta G^{\neq \tilde{e}}$ , were obtained from following equation:

$$\Delta G^{*E} = RT[ln(\eta V) - xln(\eta_1 V_1) - (1 - x)ln(\eta_2 V_2)]$$
(12)

where *R* is the universal constant of gases, *T* is the absolute temperature, *V* is the molar volume of the mixture and  $V_1$  and  $V_2$  are the respective molar volumes of the pure components.

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

$$X^{E} = x_{i} x_{j} \sum_{k=0}^{3} A_{k} (2x_{i} - 1)^{k}$$
(13)

where  $X^{E}$  represents any of the following properties:  $V^{E}$ ,  $\eta^{E}$ ,  $\Delta G^{*E}$ ,  $x_{i}$ ,  $x_{j}$  are the mole fractions of the components *i* and *j*, respectively, and  $A_{k}$  denotes the polynomial coefficients.

The values of these coefficients are indicated in table 5 along with the standard deviation calculated with equation 5.

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 [22-26]. The effect of temperature on the V<sup>E</sup> shows a systematic decrease with temperature for all the mixtures. As an illustration, figure 1 shows experimental (eq. 10) and calculated values (equation 13, continuous curve) of V<sup>E</sup> at all temperature.

T/K	$A_0$	$A_1$	$A_2$	A3	σ			
$V^{E/(cm^3 mol^{-1})}$								
288.15	-1.42448	-0.38836	-0.36626	0.74191	0.0096			
293.15	-1.50329	0.14184	-1.50645	-1.36073	0.0143			
298.15	-1.67504	0.39666	-1.50695	-1.76559	0.0272			
303.15	-1.75569	0.31418	-1.8631	-1.30431	0.0296			
308.15	-1.86465	0.16925	-2.42789	-0.68731	0.0265			
			η <sup>E</sup> /(mPa·s)					
288.15	-3.61066	2.44702	-1.73786	1.14151	0.0058			
293.15	-3.07647	2.00605	-1.29541	0.99288	0.0039			
298.15	-2.49878	1.64372	-1.1680	0.71116	0.0062			
303.15	-2.09962	1.26925	-0.89552	0.74686	0.0027			
308.15	-1.7479	1.1030	-0.79883	0.35759	0.0059			
			∆G* <sup>E</sup> /J·mol <sup>-1</sup>					
288.15	-4013.0805	1222.1292	229.43882	414.37841	14.5			
293.15	-3972.7653	914.58541	638.57279	1272.5612	16.3			
298.15	-3681.7214	1245.6801	-130.57694	30.83315	26.5			
303.15	-3546.5708	766.18517	66.30362	1123.9093	11.2			
308.15	-3303.8753	1226.7292	-612.63269	-815.48617	25.9			

Table 5COEFFICIENTS A, AND STANDARDDEVIATIONS,σ, OF THE EXCESSFUNCTIONS OF THEMTBE (x) + iso-PROPANOL SYSTEM



Fig. 1. Excess molar volumes versus mole fraction of (x) MTBE + iso-propanol at:(■)288.15 K, (•) 293.15 K, (▲) 298.15 K, (▼) 303.15 K, (◀) 308.15 K.



Fig. 3. Excess free energy of activation versus mole fraction of (x) MTBE + iso-propanol at: (■) 288.15 K, (•) 293.15 K, (▲) 298.15 K, (▼) 303.15 K, (◀) 308.15 K

The excess viscosities are negative over the entire range of mole fractions at all the temperatures. The negative excess viscosity was explained by many authors through different forms [26-28]. By the addition of MTBE molecules, the alcohol molecules dissociate and have greater mobility than the pure alcohols due the reduced cohesive forces of alcohol molecules upon mixing [29-31]. 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 [22]. Figure 2 shows experimental (equation 11) and calculated values (equation 13, continuous curve) of  $\eta^{E}$ .

Negative values were observed for excess molar Gibbs energies of activation of viscous flow of the all the binary mixtures and at all temperatures studied. The  $\Delta G^{\neq E}$  parameter may be considerable a valid measure to detect the presence of interaction between molecules [32,33].

Negative values of  $\Delta G^{\#E}$  correspond to the existence of solute-solute association [34]. Figure 3 shows experimental (equation 12) and calculated values (equation 13, continuous curve) of  $\Delta G^{\#E}$  at all temperature.

## Conclusions

The densities and viscosities of binary mixtures of MTBE + *iso*-propanol were measured experimentally at temperatures (288.15, 293.15, 298.15, 303.15 and 308.15) K over the entire composition range. From these results, the excess values of molar volume, viscosity and Gibbs energie of activation of viscous flow have been computed and fitted to the Redlich-Kister equation, each with low



Fig. 2. Excess viscosities versus mole fraction of (x) MTBE + isopropanol at: (■) 288.15 K, (◆) 293.15 K, (▲) 298.15 K, (▼) 303.15 K, (◀) 308.15 K

standard deviations. Negative deviations in the range of measured mole fraction were observed for excess molar volumes, for the excess dynamic viscosity and for the excess molar Gibbs energies of activation of viscous flow at all of the temperatures.

The density of the solutions studied in this paper can be correctly estimated at different temperatures using a linear equation, and the viscosity of these solutions conforms Guzman's equation.

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. The results of these correlations showed that Jouyban Acree model is best for all temperatures investigated except temperature 298.15 K, where Heric-Brewer is better.

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