Experimental Study and Correlation Models of the Density and Viscosity of 2-Methoxy-2-methylpropane with *n*-propanol

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Densities (ρ) and viscosities (η) of the binary systems 2-Methoxy-2-methylpropane with n-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. Belda, Herraez, Emmerling et al. and Gonzalez-Olmos-Iglesias models were used to correlate the densities of binary mixtures. The Mchaweh-Nasrifar-Mashfeghian, Hankinson-Thomson, Yamada-Gunn and Reid et al. models were used to estimate the liquid density of the binary mixtures. Viscosity results were fitted to the equations of Grunberg-Nissan, Heric-Brewer, Jouyban-Acree and McAllister. The thermodynamic activation parameters, free energy, $\Delta G^{\neq}$, enthalpy, $\Delta H^{\neq}$ and entropy, $\Delta S^{\neq}$ for viscous flow have been estimated and their variations with compositions have been discussed.

Keywords: binary liquid mixtures, correlation models, 2-Methoxy-2-methylpropane, n-propanol

The thermophysical properties of liquid mixtures and their analysis in terms of models are important for the design of industrial processes and the search of models capable of correlating the molecular structure and macroscopic properties of liquids [1]. From a theoretical point of view, mixtures of alcohols and ethers are of interest due to the complex structure, a consequence of the self-association of the alcohols, wich is partially destroyed by the active ether molecules, and on the new intermolecular OH-O bonds created [2]. The density of liquids can be determined experimentally, but may be correlated and estimated by different analytical relations [3].

The present study reports experimental density and viscosity data for pure 2-Methoxy-2-methylpropane (MTBE), *n*-propanol and for their binary systems in the 288.15 – 308.15 K temperature range. Different theoretical models were used to estimate the liquid density and also to correlate the density and the viscosity of the binary mixtures.

Experimental part

The chemicals MTBE (mole fraction purity > 0.995) and *n*-propanol (mole fraction purity >0.999) were obtained from Merck. The chemicals were dried over molecular sieves (Fluka type 4 Å). The purity was checked through chromatographic analysis. 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 ± 0.00005 $g \cdot cm^{-3}$. The experimental technique has been previously described [4], 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 [5] 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 (s) is the flowing time of a constant volume liquid through the viscometer capillary. Accuracy of time measurement is ± 0.01s. A and B are characteristic constants of the used viscometer, which were determined by taking bidistilled 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 ± 0.001 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 can be seen in table 1. Densities and

Table 1
EXPERIMENTAL AND LITERATURE VALUES FOR DENSITIES (ρ) AND VISCOSITIES (η) OF THE PURE COMPONENTS

Component	T/K	ρ /(g·cm^{-3})		η /(mPa·s)	
		Experimental	Literature	Experimental	Literature
MTBE	288.15	0.7458	0.7457[6]	0.3905	-
	293.15	0.7408	0.7404[6]	0.3706	0.3711[7]
	298.15	0.7357	0.7357[8]	0.3550	0.3560[7]
	303.15	0.7304	0.7301[9]	0.3387	0.3490[10]
	308.15	0.7243	0.7248[9]	0.3214	0.3245[11]
n-Propanol	288.15	0.8084	-	2.4915	-
	293.15	0.8044	0.80428[12]	2.2304	2.1970[12]
	298.15	0.8007	0.80021[12]	1.9885	1.9700[13]
	303.15	0.7967	0.79642[12]	1.7641	1.7843[14]
	308.15	0.7929	0.79227[12]	1.5540	1.5460[14]

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Table 2

DENSITIES (ρ) AND VISCOSITIES (η) OF THE BINARY MIXTURES OF MTBE (x) + n-PROPANOL AT DIFFERENT TEMPERATURES AND ATMOSPHERIC PRESSURE

x	ρ /(g·cm^{-3}) T /K					η /(mPa·s) T /K				
	288.15	293.15	298.15	303.15	308.15	288.15	293.15	298.15	303.15	308.15
0.0960	0.8019	0.7982	0.7946	0.7908	0.7869	1.8355	1.6998	1.4701	1.3212	1.1969
0.2003	0.7939	0.7904	0.7866	0.7830	0.7787	1.3738	1.2298	1.1220	1.0387	0.9359
0.3005	0.7864	0.7835	0.7796	0.7755	0.7708	1.0707	0.9691	0.8981	0.8199	0.7687
0.4043	0.7807	0.7765	0.7723	0.7677	0.7628	0.8527	0.7783	0.7280	0.6661	0.6277
0.5088	0.7744	0.7700	0.7655	0.7607	0.7555	0.7145	0.6605	0.6299	0.5956	0.5576
0.5958	0.7690	0.7644	0.7599	0.7551	0.7498	0.6443	0.5718	0.5305	0.4933	0.4674
0.6989	0.7632	0.7586	0.7538	0.7489	0.7433	0.5328	0.4956	0.4624	0.4369	0.4188
0.7966	0.7578	0.7532	0.7485	0.7435	0.7378	0.4724	0.4395	0.4143	0.3967	0.3790
0.8983	0.7520	0.7474	0.7425	0.7376	0.7318	0.4238	0.3951	0.3777	0.3568	0.3431

viscosities of the binary mixtures of MTBE (x) + *n*-propanol are reported in table 2.

The results of this study are in close agreement with the works of Hoga and Torres [15] at temperatures 293.15, 298.15, 303.15 and 308.15 K. No experimental data were found for the present mixtures at 288.15 K.

Densities of the binary mixtures were correlated with composition using the following semi-empirical models:

Belda [16]:

$$\rho = \rho_2 + (\rho_1 - \rho_2)x_1\left(\frac{1+m_1(1-x_1)}{1+m_2(1-x_1)}\right) \qquad (3)$$

Herraez [17]:

$$\rho = \rho_2 + (\rho_1 - \rho_2)x_1^{A+Bx_1+Cx_1^2} \qquad (4)$$

The experimental values of the density were also correlated with composition and temperature by means of the following equations:

Emmerling *et al.* [18]:

$$\rho = x_1\rho_1 + x_2\rho_2 +$$

$$+x_1x_2\begin{bmatrix}P_1 + P_2T + P_3T^2 + (P_4 + P_5T + P_6T^2)(x_1 - x_2) + \\ (P_7 + P_8T + P_9T^2)(x_1 - x_2)^2\end{bmatrix} \qquad (5)$$

The temperature dependence of the densities of each pure component is expressed using equation:

$$\rho_i = A_i + B_iT + C_iT^2 \ (i = 1, 2) \qquad (6)$$

Gonzalez-Olmos-Iglesias [19]:

$$\rho = \sum_{i=0}^{2} A_i x^i \qquad (7)$$

where A_i is a polynomial temperature dependence function as follows:

$$A_i = \sum_{j=0}^{2} A_{ij}T^i \qquad (8)$$

The adjustable parameters of these equations (m_1, m_2, A, B, C, P_1-P_9, A_i, B_i, C_i and A_{ij}) were estimated using the experimental data and a nonlinear regression analysis employing the Levenberg-Marquardt algorithm [20]. Tables 3 and 4 shows the fitting parameters along with the standard deviation (σ) calculated with equation:

$$\sigma = \left[\frac{\Sigma\left(X_{exp} - X_{calc}\right)^2}{m-n}\right]^{1/2} \qquad (9)$$

Table 3

PARAMETERS FOR THE SEMI-EMPIRICAL MODELS OF BELDA AND HERRAEZ AND STANDARD DEVIATIONS AT DIFFERENT TEMPERATURES

Model	Parameters and σ /(g·cm^{-3})	Temperature /(K) 288.15	293.15	298.15	303.15	308.15
Belda	m_1	-0.26059	-0.31205	-0.19791	-0.0663	0.06391
	m_2	-0.40311	-0.42833	-0.30975	-0.1786	-0.04577
	σ	0.00018	0.00044	0.00047	0.00055	0.00068
Herraez	A	0.96678	1.03049	1.04044	1.08014	1.13326
	B	-0.34623	-0.62057	-0.59166	-0.74051	-0.95118
	C	0.30130	0.63203	0.58001	0.72063	0.92715
	σ	0.00012	0.00024	0.00033	0.00029	0.00023

Table 4

VALUES OF PARAMETERS IN THE RANGE 288.15-308.15 K FOR THE EMMERLING *ET AL.* AND GONZALES-OLMOS-IGLESIAS MODELS AND STANDARD DEVIATIONS [1]

	Emmerling *et al.*	
A_1=0.30237	B_1=0.00397	C_1=-8.44128·10^{-6}
A_2=0.85178	B_2=3.69424·10^{-4}	C_2=-2.0115·10^{-6}
P_1=-0.39406	P_2=0.00258	P_3=-4.30446·10^{-6}
P_4=-0.11312	P_5=9.07986·10^{-4}	P_6=-1.7536·10^{-6}
P_7=-0.65732	P_8=0.00384	P_9=-5.35984·10^{-6}
	σ=0.00020	
	Gonzalez-Olmos-Iglesias	
A_{00}=0.84102	A_{01}=4.14485·10^{-4}	A_{02}=-2.03944·10^{-6}
A_{10}=-0.95951	A_{11}=0.00631	A_{12}=-1.09837·10^{-5}
A_{20}=0.39368	A_{21}=-0.00257	A_{22}=4.30004·10^{-6}
	σ =0.00038	

[1] Units: A_i, P_1, P_4, P_7, A_{00}, A_{10}, A_{20}, σ: g·cm^{-3}; B_i, P_2, P_5, P_8, A_{01}, A_{11}, A_{21}: g·cm^{-3}K^{-1}; C_i, P_3, P_6, P_9, A_{02}, A_{12}, A_{22}: g·cm^{-3}K^{-2}

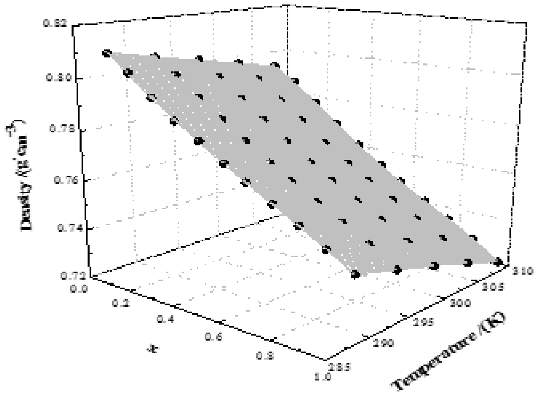

Fig 1. Variation with composition and temperature of experimental (•) and calculated density with Emmerling et al. equation

where X is the value of the analysed property, m is the number of data points, and n is the number of estimated parameters. Figure 1 shows experimental values of density and calculated values with Emmerling *et al.* equation in the 288.15 – 308.15 K temperature range.

The data from tables 3 and 4 show that Herraez model offer the best results to correlate the density with composition. The Emmerling *et al.* model is able to correlate the density with composition and temperature, better than Gonzalez-Olmos-Iglesias model.

Four models Mchaweh-Nasrifar-Mashfeghian (M-N-M) [21,22], Hankinson-Thomson (H-T) [23], Yamada-Gunn (Y-G) [24] and Reid *et al* (R-R) [25] are applied to estimate the liquid densities of the binary mixtures at the range 288.15-308.15 K and atmospheric pressure, over the whole concentration range. This models has been previously described [3].

The critical properties were taken from the literature [26, 27] and are presented in table 5, along with the values of acentric factor (w) [28]. The values of the liquid density calculated with the four models presented in this work have been compared with the experimental values of this property by using equation 9 and the values of the standard deviation are presented in table 6. The M-N-M correlation shows the best results for the mixtures studied in this work at all temperatures investigated. Figure 2 shows experimental values of density and estimated values with the models investigated at 288.15 K.

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

Grunberg-Nissan [29]:

$$\ln\eta = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1x_2d \qquad (10)$$

Heric-Brewer [30]:

$$\ln\eta = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1\ln M_1 + x_2\ln M_2 - \\ -\ln(x_1M_1 + x_2M_2) + x_1x_2[\alpha_{12} + \alpha_{21}(x_1 - x_2)] \qquad (11)$$

Table 5

CRITICAL PROPERTIES FOR THE COMPONENTS

Compound	T_c (K)	ρ_c (g·cm^{-3})	ω
MTBE	497.10	0.268	0.266
n-propanol	536.8	0.275	0.407

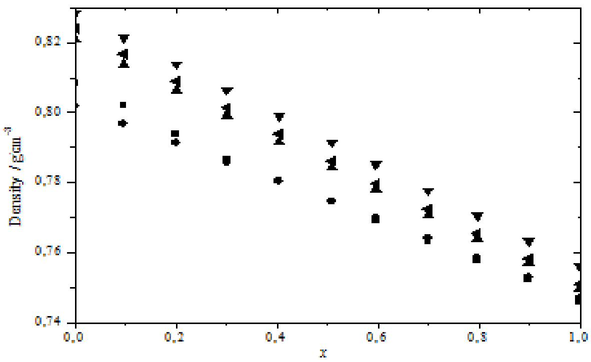

Fig. 2. Comparison of the experimental density (■) with estimation density: MNM (•), HT (▲), RR (▼), YG (◀) at 288.15 K

Jouyban Acree [31,32]:

$$\ln\eta = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1x_2\sum_{j=0}^{n}\left(\frac{A_j(x_1-x_2)^j}{T}\right) \qquad (12)$$

McAllister three body model [33]:

$$\ln\eta = x_1^3\ln\eta_1 + 3x_1^2x_2\ln\eta_{12} + 3x_1x_2^2\ln\eta_{21} + \\ + x_2^3\ln\eta_2 - \ln[x_1 + x_2\,M_2/M_1] + \\ +3x_1^2x_2\ln\left[\frac{(2 + M_2/M_1)}{3}\right] + 3x_1x_2^2\ln\left[\frac{(1 + 2\,M_2/M_1)}{3}\right] + \\ + x_2^3\ln[M_2/M_1] \qquad (13)$$

McAllister four body model [33]:

$$\ln\eta = x_1^4\ln\eta_1 + 4x_1^3x_2\ln\eta_{1112} + 6x_1^2x_2^2\ln\eta_{1122} + \\ +4x_1x_2^3\ln\eta_{2221} + x_2^4\ln\eta_2 - \\ \ln[x_1 + x_2(M_2/M_1)] + 4x_1^3x_2\ln[(3 + M_2/M_1)/4] + \\ +6x_1^2x_2^2\ln[(1 + M_2/M_1)/2] + \;4x_1x_2^3\ln[(1 + 3M_2/M_1)/4] + \\ +x_2^4\ln[M_2/M_1] \qquad (14)$$

In these equations η_1, and η_1, η_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, α_{12}, α_{21}, A_j, η_{12}, η_{21}, η_{1112}, η_{1122} and η_{2221} are interaction parameters (viscosity coefficients) and reflect the non-ideality of the system.

The parameters that appear in equations 10-14 were estimated using the experimental viscosity data and a non-linear regression analysis employing the Levenberg-Marquardt algorithm [20]. Table 7 shows the parameters calculated and the standard deviations between experimental values and those obtained using the semi-empirical relations, calculated with equation 9. A comparison of the calculated and the experimental viscosities shows that the Jouyban – Acree model produces accurate results for 288.15 K and 293.15 K, the Grunberg -

Table 6

STANDARD DEVIATION FOR PREDICTION OF LIQUID DENSITY OF BINARY SOLUTIONS USING DIFFERENT THEORETICAL MODELS

System	Model	σ/ (g·cm^{-3})				
		Temperature / (K)				
		288.15	293.15	298.15	303.15	308.15
MTBE + *n*-propanol	M-N-M	0.0027	0.0031	0.0036	0.0040	0.0044
	H-T	0.0093	0.0086	0.0050	0.0066	0.0062
	R-R	0.0167	0.0155	0.0144	0.0135	0.0130
	Y-G	0.0118	0.0106	0.0096	0.0087	0.0082

Table 7
PARAMETERS FOR THE SEMI-EMPIRICAL RELATIONS OF GRUNBERG-NISSAN, HERIC-BREWER, JOYBAN-ACREE AND MCALLISTER AND STANDARD DEVIATIONS AT DIFFERENT TEMPERATURE

T/K	Equation				
	Grunberg-Nissan	Heric-Brewer	Jouyban-Acree	McAllister three body model	McAllister four body model
288.15	d = -1.2356 σ = 0.0258	α_{12} = -1.16387 α_{21} = 0.25868 σ = 0.0167	A_0 = -348.51979 A_1 = 124.50581 A_2 = -57.92124 A_3 = -143.52363 σ = 0.0153	η_{12} = 0.52733 η_{21} = 0.82202 σ = 0.0168	η_{1112} = 0.46792 η_{1122} = 0.70926 η_{2221} = 1.03695 σ = 0.0164
293.15	d = -1.27915 σ = 0.0198	α_{12} = -1.20703 α_{21} = 0.17585 σ = 0.0148	A_0 = -372.39118 A_1 = 125.91851 A_2 = -22.74446 A_3 = -217.07618 σ = 0.0100	η_{12} = 0.47062 η_{21} = 0.76033 σ = 0.0148	η_{1112} = 0.43544 η_{1122} = 0.615 η_{2221} = 0.97673 σ = 0.0157
298.15	d = -1.24169 σ = 0.0066	α_{12} = -1.16941 α_{21} = 0.16031 σ = 0.0174	A_0 = -354.94094 A_1 = 40.10968 A_2 = -110.4724 A_3 = 31.02467 σ = 0.0133	η_{12} = 0.44341 η_{21} = 0.7067 σ = 0.0174	η_{1112} = 0.38694 η_{1122} = 0.64283 η_{2221} = 0.84407 σ = 0.0122
303.15	d = -1.18253 σ = 0.0240	α_{12} = -1.11014 α_{21} = 0.13957 σ = 0.0225	A_0=-346.70897 A_1=54.26727 A_2= -85.92162 A_3= -28.18212 σ = 0.0238	η_{12} = 0.41824 η_{21} = 0.65967 σ = 0.0225	η_{1112} = 0.37292 η_{1122} = 0.58365 η_{2221} = 0.79252 σ = 0.0218
308.15	d = -1.04686 σ = 0.0201	α_{12} = -0.97449 α_{21} = 0.14451 σ = 0.0174	A_0 = -314.00262 A_1 = 42.18914 A_2 = -62.89559 A_3 = 15.35676 σ = 0.0187	η_{12} = 0.40572 η_{21} = 0.62221 σ = 0.0174	η_{1112} = 0.36598 η_{1122} = 0.54163 η_{2221} = 0.74752 σ = 0.0170

Nissan model is better for 298.15 K, while for 303.15 K and 308.15 K the four-body McAllister model is the best to describe viscosities.

The thermodynamic activation parameters for viscous flow have been calculated using Eyring equation [34]:

$$\eta = \frac{hN}{V} exp\left(\frac{\Delta G^{\neq}}{RT}\right) \tag{15}$$

where η is viscosity of a liquid mixtures, *h* is Planck's constant, *N* is Avogadro's number, *V* is the molar volume of the solution, *R* is general gas constant, *T* is temperature and $\Delta G^{\neq}$ is the molar Gibbs energy of activation for the viscous flow process. Combining with

$$\Delta G^{\neq} = \Delta H^{\neq} - T\Delta S^{\neq} \tag{16}$$

we can write,

$$ln\left(\frac{\eta V}{hN}\right) = \frac{\Delta H^{\neq}}{RT} - \frac{\Delta S^{\neq}}{R} \tag{17}$$

where $\Delta H^{\neq}$ and $\Delta S^{\neq}$ are enthalpy and entropy of activation of viscous flow.

From the experimental viscosity and density data, plots of ln$(\eta V/hN)$ vs $1/T$ were found to be linear in the temperature range 288.15 to 308.15 K, as such the values of $\Delta H^{\neq}$ and $\Delta S^{\neq}$ were obtained by the corresponding slopes and the intercepts. By substituting these values in equation 16, the values of $\Delta G^{\neq}$ were also calculated at different temperatures. The values of thermodynamic functions of activation are listed in table 8 as a function of composition.

The values of $\Delta H^{\neq}$ are more than three times higher for *n*-propanol than that for MTBE, indicating that association and dipole-dipole interactions increase the value of $\Delta H^{\#}$. The value of $\Delta S^{\#}$ is negative for MTBE and positive for *n*-propanol. The positive value indicated that rupturing of hydrogen bonds formed through OH groups of alcohol in the activation process for viscous flow occurs, resulting in the structural disorder [35]. The values of $\Delta H^{\#}$ and $\Delta G^{\#}$ are positive for all the binary mixtures while the values of $\Delta S^{\#}$ are negative for all binary mixtures except for the mixtures concentrated in *n*-propanol.

Table 8
VALUES OF $\Delta G^{\#}$, $\Delta H^{\#}$ AND $\Delta S^{\#}$ FOR THE BINARY MIXTURES OF MTBE (x) + n-PROPANOL

x	$\Delta S^{\neq}$ (J·mol^{-1}K^{-1})	$\Delta H^{\neq}$ (J·mol^{-1})	$\Delta G^{\neq}$ (J·mol^{-1})				
			T/K				
			288.15	293.15	298.15	303.15	308.15
0.0000	18.8	20436.5	15018.6	14924.6	14830.6	14736.6	14642.6
0.0960	13.7	18189.0	14234.6	14166.0	14097.4	14028.8	13960.2
0.2003	9.1	16282.4	13665.0	13619.5	13574.1	13528.7	13483.3
0.3005	4.7	14575.3	13210.6	13186.9	13163.2	13139.6	13115.9
0.4043	-1.8	12344.4	12854.0	12862.9	12871.7	12880.5	12889.4
0.5088	-5.6	10950.5	12567.0	12595.0	12623.1	12651.1	12679.2
0.5958	-9.3	9656.9	12347.3	12394.0	12440.7	12487.3	12534.0
0.6989	-13.6	8300.3	12223.1	12291.2	12359.2	12427.3	12495.4
0.7966	-15.3	7766.8	12172.3	12248.8	12325.2	12401.7	12478.1
0.8983	-16.5	7389.4	12156.3	12239.0	12321.8	12404.5	12487.2
1.0000	-18.7	6910.6	12178.3	12269.7	12361.1	12452.5	12543.9

Conclusions

The densities and viscosities of binary mixtures of MTBE + *n*-propanol were measured experimentally at 288.15, 293.15, 298.15, 303.15 and 308.15 K over the entire composition range. Belda and Herraez models have been used to correlate the densities with concentration and Emmerling *et al.* and Gonzalez-Olmos-Iglesias models were used to correlate the densities of binary mixtures with concentration and temperature.

Four models are tested to estimate the liquid densities of the binary mixtures. The Mchaweh-Nasrifar-Mashfeghian model show the best result.

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 and four-body McAllister models are better. The activation energies of viscous flow have been estimated and discussed.

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