Thermophysical Study of the Binary Mixtures of N,N-

Dimethylacetamide with 1-Propanol and 1-Butanol

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Abstract

Several thermophysical properties such as densities,  $\rho$ , speeds of sound, u, refractive

indices,  $n_D$ , and kinematic viscosities, v, have been measured for the binary mixtures of

N,N-Dimethylacetamide with 1-propanol and 1-butanol over the entire range of

composition, at the temperatures (283.15, 298.15 and 313.15) K and at atmospheric

pressure p = 0.1 MPa. From these experimental data, excess molar volumes,  $V^{E}$ , excess

isentropic compressibilities,  $\kappa_s^E$ , refractive index deviations,  $\Delta n_D$ , and viscosity

deviations,  $\Delta \eta$ , were calculated. The calculated excess or deviation properties were

correlated with the Redlich-Kister equation and the corresponding parameters were

presented. The results obtained, were discussed in terms of structural effects and

specific molecular interactions, and the influence of the alkanol chain length was also

considered.

Keywords: 1-alkanols, N,N-dimethylacetamide, molecular interactions, thermophysical

properties.

#### 1. Introduction

Alkanols are interesting versatile solvents, used in chemical and technological processes; they are inexpensive and easily available at high purity. These polar liquids are self-associated through hydrogen bonding, creating multimers of different degrees [1,2].

When amide and alkanols are mixing different intermolecular interactions take place, resulting in non-ideal behaviour. Hence, the measurement of thermophysical properties such as density, speed of sound, refractive index, and viscosity allows the calculation of excess and deviation properties. From the analysis of sign and magnitude of these properties, it is possible to understand the specific interactions occurring between the molecules. The major goal of our research is to provide thermophysical data and to relate them with molecular size, shape and charge distribution of the mixed compounds.

An extensive survey of the literature has shown that, few studies were available concerning the thermophysical properties for these binary mixtures. Density and viscosity measurements were reported for these mixtures at T = 303.15 K [3]. Another density measurements and calculated excess molar volumes were given only at T =298.15 K by two authors [4,5], and finally the refractive indices and their deviations at T = 298.15 K [5] were reported too. On the other hand, there is more data available regarding the excess enthalpies for these kind of systems at temperatures 298.15 K [4,6,7] and 313.15 K [8], and phase equilibrium for the 1-propanol and N,Ndimethylacetamide mixture (solid-liquid equilibrium [9] and vapour-liquid equilibrium [10]). An earlier study was undertaken [11], to gain thermophysical information about the N,N-dimethylacetamide when it is mixed with methanol or ethanol. We continue this study, by reporting the experimental density, speed of sound, refractive index and viscosity for the binary mixtures of N,N-dimethylacetamide with 1-propanol and 1butanol at temperatures (283.15, 298.15 and 313.15) K and at atmospheric pressure, p =0.1 MPa. The excess molar volume, excess isentropic compressibility, the refractive index deviation and viscosity deviation were also calculated and correlated. The influence of chain length of alcohols has been also studied.

## 2. Experimental

#### 2.1. Materials

Table 1 reports for the chemicals used in this work, the provenance, the mass purity checked by gas chromatography and the water content measured by an automatic titrator Crison KF 1S-2B.

The experimental density, speed of sound, refractive index and kinematic viscosity values, of the pure components at work temperatures and at p = 0.1 MPa are collected in Table 2 along with isobaric expansibility values, calculated from our density measurements, and molar heat capacity values taken from literature [12-13]. For comparison, some values found in the literature [14-31] are also included in this Table.

#### 2.2. *Methods*

The binary mixtures were prepared by weighing appropriate volumes of pure liquids on an electronic balance Sartorius semimicro balance CP225-D within  $\pm 1\cdot 10^{-5}$ g, the uncertainty in the mole fraction was estimated to be 0.0005. To ensure the homogeneity of samples, they were well mixed, and were kept in airtight stopper glass vials, in order to minimize evaporation losses.

The density,  $\rho$ , and the speed of sound, u, of the sample were measured simultaneously by an Anton Paar DSA 5000 densimeter and sound analyzer at 3 MHz. This device automatically corrects the influence of viscosity on density values. The temperature in the cell was maintained stable to  $\pm$  0.005 K. The uncertainties of  $\rho$  and u measurements are 0.1 kg·m<sup>-3</sup> and 0.5 m·s<sup>-1</sup>, respectively.

The refractive index at 589.3 nm sodium wavelength,  $n_D$ , of the sample was measured by an automatic refractometer Abbemat-HP DR. Kernchen, with an uncertainty of  $10^{-5}$ . During the measurement, the sample temperature is controlled by a Peltier device with a stability of  $\pm 0.002$  K.

Finally we measured the kinematic viscosity,  $\nu$ , using an Ubbelohde capillary viscometer. The viscosimeter with the sample was kept within  $\pm$  0.01 K, using a Schoot-Geräte CT 1150/2 thermostat. To determine the flow time, an automatic measuring unit Schoot-Geräte AVS-440 was employed. The corresponding kinetic energy correction was applied to measured time. Once density and kinematic viscosity are known, the dynamic viscosity  $\eta$ , can be calculated using:  $\eta = \rho \cdot \nu$ . The uncertainties of both kinematic and dynamic viscosity determinations were estimated to be 0.005 mm<sup>2</sup>·s<sup>-1</sup>

and 0.005 mPa·s respectively. All details of calibration of these devices can be found in previous papers [32,33].

#### 3. Results and discussion

Experimental values of density, speed of sound, refractive index, and kinematic viscosity and calculated values of isentropic compressibility, dynamic viscosity, excess molar volume, excess isentropic compressibility, refractive index deviation, and viscosity deviation for the binary systems of N,N-dimethylacetamide with 1-propanol and 1-butanol are collected in Table S1 of the Supplementary material.

### 3.1. Excess molar volume

The excess molar volume (± 10<sup>-9</sup> m<sup>3</sup>·mol<sup>-1</sup>) was calculated from experimental densities of the pure liquids and their mixtures using the following equation:

$$V^{E} = \sum_{i} x_{i} M_{i} \left( \frac{1}{\rho} - \frac{1}{\rho_{i}} \right) \tag{1}$$

where  $\rho$  is the density of the mixture and  $x_i$ ,  $M_i$ , and  $\rho_i$  are, the mole fraction, the molar mass and density of component i, respectively.

The plots of  $V^E$  vs mole fraction  $x_1$ , for the binary mixtures of N,N-dimethylacetamide with 1-propanol and 1-butanol are presented in Fig. 1. The excess molar volume is found to be negative for the binary mixture containing 1-propanol. The magnitude of negative  $V^E$  values decreases when temperature increases. For the mixture involving 1-butanol a sigmoid  $V^E$  behavior is observed. The negative  $V^E$  values are found in the 1-butanol rich region, and they change gradually from negative to positive as the temperature raises, at T=313.15 K the excess molar volume becomes positive over the whole composition range.

The agreement of our excess molar volumes for the system N,N-dimethylacetamide and 1-propanol at T = 298.15 K with those reported by Oba [4] is excellent with an average deviation of 0.0035 cm<sup>3</sup>·mol<sup>-1</sup>. The comparison of our results for N,N-dimethylacetamide and 1-butanol at T = 298.15 K with those of Oba et al. [4] and Ritzoulis and Fidantsi [5], is a little bit less favorable with average deviation of 0.0221 and 0.0359 cm<sup>3</sup>·mol<sup>-1</sup>, respectively. Although it can be outlined that in the study of Ritzoulis and Fidantsi [5]  $V^{\rm E}$  also show a sigmoid behaviour.

The sign of the excess molar volume of a system depends on the relative magnitude of expansion and contraction occurring in the mixing process. If the factors

that cause expansion in volume dominate, then  $V^{E}$  values become positive. On the other hand, if the contractive factors dominate, V<sup>E</sup> values become negative [34]. A negative excess molar volume, suggests the existence of strong interactions, such as heteroassociation through the formation of hydrogen-bond, dipole-dipole interaction, and also the geometrical fitting of one component into the other, due to differences in the molar volumes and free volumes between components [35,36]. While a positive excess molar volume, could be explained by the dispersion forces, dissociation of a component which is associated in pure state, and the unfavorable packing of the molecules [3,35]. Obviously, all these factors are responsible of final balance between positive and negative contributions. For the system N,N-dimethylacetamide + 1propanol the excess molar volume is negative, although the  $V^{E}$  values are not too high, while for the system containing 1-butanol  $V^{E}$  shows a sigmoid behavior. This different behavior could be due to a worst accommodation of the 1-butanol molecules at higher temperatures and at higher mole fractions of N,N-dimethylacetamide along with the existence of stronger dispersion forces and less proton-donating ability to cause heteroassociation.

The comparison of excess molar volumes of the binary systems containing higher chain alkanols with lower alkanols taken from our previous results [11], shows less negative  $V^E$  values. According to several authors [37, 38], the higher alkanols possess less proton-donating ability than the lower ones, and therefore heteroassociation effects are smaller in their mixtures, and  $V^E$  values tend to shift towards positive values.

## 3.2. Excess isentropic compressibility

The isentropic compressibility  $\kappa_S$ , was calculated from density,  $\rho$ , and speed of sound, u, assuming that ultrasonic absorption is negligible, using the Newton-Laplace equation,  $\kappa_S = 1/\rho u^2$ .

Then, the excess isentropic compressibility ( $\pm 10^{-1}$  TPa  $^{-1}$ ) can be determined according to the following relations:

$$\kappa_{\rm S}^{\rm E} = \kappa_{\rm S} - \kappa_{\rm S}^{\rm id} \tag{2}$$

where the ideal isentropic compressibility is given by the expression obtained by Benson and Kiyohara [39]

$$\kappa_{S}^{id} = \sum_{i} \phi_{i} \left[ \kappa_{S,i} + \frac{TV_{i}\alpha_{p,i}^{2}}{C_{p,i}} \right] - T \left( \sum_{i} x_{i} V_{i} \right) \frac{\left( \sum_{i} \phi_{i} \alpha_{p,i} \right)^{2}}{\left( \sum_{i} x_{i} C_{p,i} \right)}$$

$$(3)$$

where,  $\phi_i$  and  $x_i$ , are the volume fraction and the mole faction of component i, in the mixture, T is the temperature and  $V_i$ ,  $\alpha_{p,i}$ ,  $C_{p,i}$ , and  $\kappa_{S,i}$  are respectively the molar volume, the isobaric expansibility, the molar heat capacity at constant pressure, and the isentropic compressibility of pure component i.

The excess isentropic compressibility  $\kappa_S^E$  for both binary systems is plotted as a function of the mole fraction in Fig. 2. The  $\kappa_S^E$  values are negative, and they are more negative for the mixture containing 1-propanol mixture. The  $\kappa_S^E$  minimum value occurs at  $x_1 \approx 0.4$  for both systems at all temperatures. On the other hand, when temperature increases the  $\kappa_S^E$  values decrease, that is, they are more negative.

The behaviour of  $\kappa_S^E$  is quite well reflected in the  $V^E$  behaviour, that is, the mixture N,N-dimethylacetamide + 1-propanol shows the more negative excess isentropic compressibilities and excess molar volumes. However, for the system containing 1-butanol the  $\kappa_S^E$  and  $V^E$  values are less negative or even positive at some mole fractions and temperatures. Fort and Moore, [35] have proposed that in absence of strong specific interactions components of not too different molecular size mix with positive  $\kappa_S^E$ , while if there are specific interactions like hydrogen bonding and dipole-dipole interactions the isentropic compressibility decreases leading to negative  $\kappa_S^E$ . So, for our mixtures the specific interactions decrease from 1-propanol to 1-butanol. On the other hand, the behaviour of  $\kappa_S^E$  is also closely related with structural effects like interstitial accommodation of molecules with unequal size and shape and changes in the intermolecular free length [40] in the mixture as compared to those in pure state, leading to closer approach of unlike molecules and reduction in compressibility [41, 42].

It is further observed, that  $\kappa_S^E$  values decrease in absolute value with increasing chain length of alkanols, suggesting that, the heteroassociation between molecules decreases with increasing carbon chain from methanol to 1-butanol.

## 3.3. Refractive index deviation

The refractive index deviation ( $\pm$  10<sup>-5</sup>) calculated in terms of the volume fraction,  $\phi_i$ , is given by [43, 44]:

$$\Delta n_{\rm D} = n_{\rm D} - \sum_{\rm i} \phi_{\rm i} n_{\rm D,i} \tag{4}$$

where,  $n_D$  and  $n_{D,i}$  are the refractive index of the mixture, and the refractive index of component i, respectively.

The refractive index deviation is represented against volume fraction in Fig. 3. From the results obtained, it is obvious that  $\Delta n_D$  behaviour for both systems is opposite to  $V^E$  behaviour, as it was stated by Nakata and Sakurai [45] and Brocos et al. [46]. For the mixture N,N-dimethylacetamide + 1-propanol,  $\Delta n_D$  is positive and symmetrical over the whole composition range. As a result of the decrease in volume, the intermolecular free length becomes smaller, and the refractive index increases. On the other hand for the mixture N,N-dimethylacetamide + 1-butanol,  $\Delta n_D$  values exhibit an inversion of sign at temperatures 283.15 and 298.15 K, and they become positive at T = 313.15 K. Further it is observed that  $\Delta n_D$  values decrease when temperature increases for both systems, which suggests that the light propagates faster.

Ritzoulis and Fidantsi [5] reported refractive indices at T = 298.15 K for the mixture N,N-dimethylacetamide and 1-butanol at 289.15 K. The average deviation between their and our data is very low, around 0.000250.

Based on our previous results [11], it can be seen that mixtures containing lower alkanols give more positive  $\Delta n_D$  values than those containing higher chain alkanols.

## 3.4. Viscosity Deviation

Viscosity deviation ( $\pm$  10<sup>-3</sup> mPa·s) in terms of mole fraction,  $x_i$ , was calculated using the following relation:

$$\Delta \eta = \eta - \sum_{i} x_{i} \eta_{i} \tag{5}$$

where,  $\eta$  and  $\eta_i$  are the dynamic viscosity of the mixture, and the dynamic viscosity of the component i, respectively.

Fig. 4 shows the behaviour of viscosity deviation with mole fraction of N,N-dimethylacetamide.  $\Delta \eta$  values are negative for both systems, and become less negative

when temperature increases. Moreover, it is observed that, the magnitude of  $\Delta \eta$  increases in absolute value as the chain length of alkanols increases.

The viscosity depends strongly on both the entropy of mixing which is related to the structure of the mixed components and the enthalpy of mixing [47] that reflects the molecular interactions in the mixture. As it was suggested by Fort and Moore [48], and Vogel and Weiss [47] positive viscosity deviations are obtained in mixtures with strong specific interactions. Whereas mixtures involving no relevant interactions between unlike molecules, or dispersion forces show negative  $\Delta \eta$  values. For our systems the predominant effect is the dissociation of the alkanols in the mixing process, that leads to negative  $\Delta \eta$  values. These conclusions are also drawn by Pikarienen [3] for the interpretation of  $\Delta \eta$ , and they were justified by the calorimetric measurements [6-8], that show an endothermic behaviour.

#### 3.5. Correlation

In order to correlate the calculated excess and deviation properties of the binary mixtures with composition, a Redlich-Kister polynomial equation [49] was applied:

$$Q = x_1 x_2 \sum_{i} A_i (x_1 - x_2)^i$$
 (6)

Q refers to  $V^{\rm E}$ ,  $\kappa_{\rm S}^{\rm E}$ ,  $\Delta n_{\rm D}$ , or  $\Delta \eta$ , and  $x_{\rm i}$ , is the mole fraction, or the volume fraction,  $\phi_{\rm i}$ . The values of coefficients  $A_{\rm i}$  in the above equation have been determined using the least square method. They are listed in Table 3 along with the standard deviation  $\sigma(Q)$ .

### 4. Conclusions

Using experimental density, speed of sound, refractive index and viscosity data some excess and deviation functions for the binary mixtures of N,N-dimethylacetamide with 1-propanol or 1-butanol have been calculated at the temperatures (283.15, 298.15 and 313.15) K, and at atmospheric pressure p=0.1 MPa. The Redlich-Kister polynomial equation was used to correlate the results.  $V^E$  values are negative for N,N-dimethylacetamide with 1-propanol, whereas an inversion of sign at temperatures 283.15 and 298.15 K is obtained for N,N-dimethylacetamide with 1-butanol, being  $V^E$  values finally positive at T=313.15 K.  $\Delta n_D$  values are opposite in sign to  $V^E$  values for both mixtures. On the other hand,  $\kappa_S^E$  and  $\Delta \eta$  values are negative at all temperatures.

Further, it was noticed, that interactions between unlike molecules become weaker and dispersive forces become more important for increased chain alkanol.

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Table 1

Provenance and purity of the liquid compounds.

Chemical name	Source	Purity / mass fraction	Analysis method	Water content / ppm
N,N-Dimethylacetamide	Sigma-Aldrich	0.999	GC	295
1-Propanol	Sigma-Aldrich	0.998	GC	195
1-Butanol	Sigma-Aldrich	0.999	GC	175

Table 2
Thermophysical properties of the pure compounds at p = 0.1 MPa and at work temperatures and comparison of densities, speeds of sound, refractive indices, and dynamic viscosities with literature data at T = 298.15 K.<sup>a</sup>

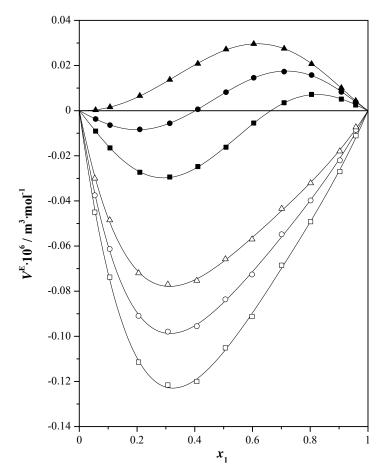
<i>T</i> / K	ρ / k	xg·m <sup>-3</sup>	u / 1	m·s⁻¹	$-C_p / J \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$	o. /1-IZ-l	n	D	$\eta$ / n	nPa·s
	Exptl.	Lit.	Exptl.	Lit.	- C <sub>p</sub> /J·IIIOI ·K	$\alpha_{\rm p}$ / kK <sup>-1</sup> -	Exptl.	Lit.	Exptl.	Lit.
				N,	N-Dimethylacetamide	;				
283.15	950.13		1516.80		170.70 <sup>e</sup>	0.9719	1.442726		1.1683	
298.15	936.29	936.34 <sup>b</sup> 936.28 <sup>c</sup>	1453.68	1455.91 <sup>b</sup> 1455.37 <sup>d</sup>	172.69°	0.9863	1.435794	1.4356 <sup>f</sup> 1.4359 <sup>g</sup>	0.9299	$0.920^{\rm f} \ 0.937^{\rm h}$
313.15	922.43		1393.35		175.46 <sup>e</sup>	1.0011	1.429006		0.7644	
					1-Propanol					
283.15	811.74		1258.06		136.29 <sup>1</sup>	0.9936	1.389271		2.8407	
298.15	799.77	799.66 <sup>i</sup> 799.62 <sup>j</sup>	1206.17	$1206^{i} \\ 1205.93^{k}$	144.18 <sup>1</sup>	1.0085	1.383104	1.3832 <sup>m</sup> 1.38309 <sup>n</sup>	1.9491	1.947° 1.9503 <sup>p</sup>
313.15	787.54		1155.36		$153.09^{1}$	1.0241	1.376864		1.3892	
					1-Butanol					
283.15	817.20		1291.66		$167.94^{1}$	0.9394	1.403162		3.8636	
298.15	806.09	$805.93^{\mathrm{k}}$ $806.06^{\mathrm{q}}$	1240.60	1240.09 <sup>r</sup> 1240.37 <sup>s</sup>	$177.10^{1}$	0.9527	1.397139	1.39729 <sup>t</sup> 1.39720 <sup>u</sup>	2.5733	2.59 <sup>t</sup> 2.563 <sup>u</sup>
313.15	794.44		1190.52		187.29 <sup>1</sup>	0.9667	1.391074		1.7696	

<sup>&</sup>lt;sup>a</sup> Standard uncertainties u are u(T) = 0.005 K for densities and speeds of sound and u(T) = 0.01 K for the rest of properties, u(p) = 0.003 MPa,  $u(\rho) = 0.1$  kg·m<sup>-3</sup>, u(u) = 0.5 m·s<sup>-1</sup>,  $u(n_D) = 10^{-5}$ , and  $u(\eta) = 0.005$  mPa·s.

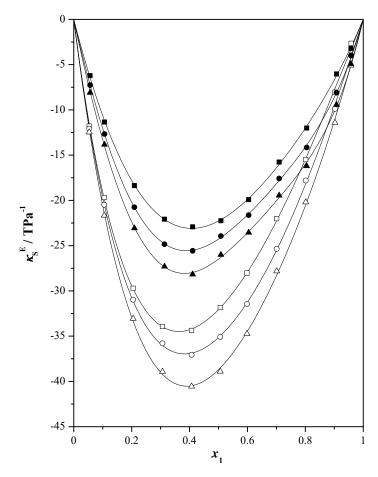
<sup>&</sup>lt;sup>b</sup> Ref. [14]. <sup>c</sup> Ref. [15]. <sup>d</sup> Ref. [16]. <sup>e</sup> Ref. [12]. <sup>f</sup> Ref. [17]. <sup>g</sup> Ref. [18]. <sup>h</sup> Ref. [19]. <sup>i</sup> Ref. [20]. <sup>j</sup> Ref. [21]. <sup>k</sup> Ref. [22]. <sup>l</sup> Ref. [13]. <sup>m</sup> Ref. [23]. <sup>n</sup> Ref. [24]. <sup>o</sup> Ref. [25]. <sup>p</sup> Ref. [26]. <sup>q</sup> Ref. [27]. <sup>r</sup> Ref. [28]. <sup>s</sup> Ref. [29]. <sup>t</sup> Ref. [30]. <sup>u</sup> Ref. [31].

**Table 3** Adjusted parameters and standard deviations,  $\sigma(Q)$ , for fitting equation.

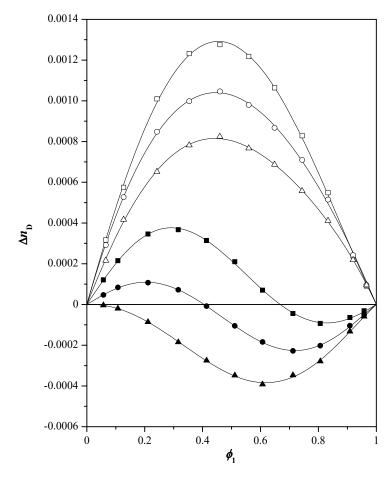
Function	T/K	$A_0$	$A_1$	$A_2$	$A_3$	$\sigma(Q)$
	N,	N-Dimethylacet	amide (1) + 1-p	propanol (2)		
	283.15	-0.4279	0.3114	-0.1904	0.0066	0.0013
$V^{\rm E} \cdot 10^6$ / ${ m m}^3 \cdot { m mol}^{-1}$	298.15	-0.3400	0.2486	-0.1769	0.0278	0.0011
	313.15	-0.2671	0.1956	-0.1501	0.0176	0.0009
	283.15	-128.27	65.48	-33.60	19.80	0.32
$\kappa_{ m S}^{ m E}$ / TPa <sup>-1</sup>	298.15	-140.62	58.47	-34.70	17.61	0.29
	313.15	-154.79	62.97	-34.46	2.97	0.38
	283.15	0.005114	-0.001165	-0.001111	0.000115	0.000014
$\Delta n_{ m D}$	298.15	0.004112	-0.00091	0.000056	0.000126	0.000012
	313.15	0.003216	-0.00076	0.000147	0.000486	0.000007
	283.15	-2.0322	1.6586	-1.2499	0.2441	0.0065
$\Delta\eta$ / mPa·s	298.15	-1.1847	0.9422	-0.5994	0.0844	0.0038
	313.15	-0.6771	0.4566	-0.2292	0.0766	0.0018
	N	,N-Dimethylace	tamide (1) + 1-1	butanol (2)		
	283.15	-0.0668	0.2096	0.0166	-0.0978	0.0002
$V^{\rm E} \cdot 10^6$ / ${ m m}^3 \cdot { m mol}^{-1}$	298.15	0.0302	0.1528	-0.0206	-0.0746	0.0002
	313.15	0.1063	0.0986	-0.0595	-0.0512	0.0002
	283.15	-89.12	33.48	-11.30	-9.44	0.22
$\kappa_{\mathrm{S}}^{\mathrm{E}}$ / TPa <sup>-1</sup>	298.15	-97.13	42.09	-27.86	-28.57	0.19
	313.15	-105.95	48.00	-38.56	-41.15	0.27
	283.15	0.000859	-0.002603	-0.000230	0.001105	0.000006
$\Delta n_{ m D}$	298.15	-0.000378	-0.001982	0.000235	0.000983	0.000003
	313.15	-0.001378	-0.001257	0.000728	0.000556	0.000005
	283.15	-3.3888	2.3349	-1.6992	1.5910	0.0090
$\Delta \eta$ / mPa·s	298.15	-1.9277	1.4492	-0.7491	0.4149	0.0058
	313.15	-1.0500	0.8618	-0.2869	0.0780	0.0026



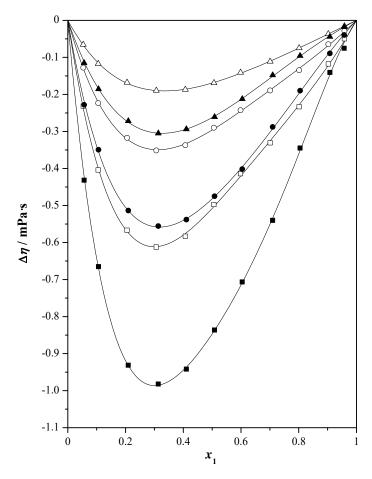
**Figure 1.** Excess volumes,  $V^E$ , for N,N-dimethylacetamide (1) + alkanol (2) at p = 0.1 MPa as a function of mole fraction,  $x_1$ :  $\square$ , 1-propanol at T = 283.15 K;  $\bigcirc$ , 1-propanol at T = 298.15 K;  $\triangle$ , 1-propanol at T = 313.15 K;  $\blacksquare$ , 1-butanol at T = 283.15 K;  $\bigcirc$ , 1-butanol at T = 298.15 K;  $\triangle$ , 1-butanol at T = 313.15 K;  $\bigcirc$ , equation 6.



**Figure 2.** Excess isentropic compressibilities,  $\kappa_S^E$ , for N,N-dimethylacetamide (1) + alkanol (2) at p = 0.1 MPa as a function of mole fraction,  $x_1: \Box$ , 1-propanol at T = 283.15 K;  $\bigcirc$ , 1-propanol at T = 298.15 K;  $\triangle$ , 1-propanol at T = 313.15 K;  $\blacksquare$ , 1-butanol at T = 283.15 K;  $\bigcirc$ , 1-butanol at T = 298.15 K;  $\triangle$ , 1-butanol at T = 313.15 K;  $\longrightarrow$ , equation 6.



**Figure 3.** Refractive index deviations,  $\Delta n_D$ , for N,N-dimethylacetamide (1) + alkanol (2) at p = 0.1 MPa as a function of volume fraction,  $\phi_1$ :  $\square$ , 1-propanol at T = 283.15 K;  $\bigcirc$ , 1-propanol at T = 298.15 K;  $\triangle$ , 1-propanol at T = 313.15 K;  $\blacksquare$ , 1-butanol at T = 283.15 K;  $\bigcirc$ , 1-butanol at T = 298.15 K;  $\triangle$ , 1-butanol at T = 313.15 K;  $\bigcirc$ , equation 6.



**Figure 4.** Viscosity deviations,  $\Delta \eta$ , for N,N-dimethylacetamide (1) + alkanol (2) at p = 0.1 MPa as a function of mole fraction,  $x_1$ :  $\square$ , 1-propanol at T = 283.15 K;  $\bigcirc$ , 1-propanol at T = 298.15 K;  $\triangle$ , 1-propanol at T = 313.15 K;  $\blacksquare$ , 1-butanol at T = 283.15 K;  $\bigcirc$ , 1-butanol at T = 298.15 K;  $\triangle$ , 1-butanol at T = 313.15 K;  $\bigcirc$ , equation 6.

# **Supplementary material**

**Table S1.** Experimental densities,  $\rho$ , speeds of sound, u, isentropic compressibilities,  $\kappa$ s, refractive indices,  $n_D$ , kinematic viscosities,  $\nu$ , absolute viscosities,  $\eta$ , and calculated excess volumes,  $V^E$ , excess isentropic compressibilities,  $\kappa_S^E$ , refractive index deviations,  $\Delta n_D$ , and viscosity deviations,  $\Delta \eta$ , for the binary mixtures N, N-dimethylacetamide (1) + 1-alkanol (2) at p = 0.1 MPa and at work temperatures <sup>a</sup>

$x_1$	$\phi_1$	$\rho/\mathrm{kg}\cdot\mathrm{m}^{-3}$	$V^{\rm E} \cdot 10^6$ / ${ m m}^3 \cdot { m mol}^{-1}$	<i>u</i> / m·s <sup>-1</sup>	κ <sub>S</sub> / TPa <sup>-1</sup>	$\kappa_{\mathrm{S}}^{\mathrm{E}}$ /	$n_{ m D}$	$\Delta n_{ m D}$	ν/ mm <sup>2</sup> ·s <sup>-1</sup>	η/mPa·s	Δη/ mPa·s
			N	I,N-Dimethyla	cetamide (1) + 1	-propanol (2	) at $T = 283.15$ ]	K			
0.0000	0.0000	811.74		1258.06	778.36		1.389271		3.4995	2.8407	
0.0535	0.0654	821.29	-0.0450	1277.92	745.59	-11.80	1.393084	0.000317	3.0681	2.5198	-0.2314
0.1052	0.1271	830.14	-0.0738	1295.36	717.91	-19.68	1.396640	0.000575	2.7234	2.2608	-0.4040
0.2053	0.2424	846.50	-0.1114	1326.96	670.90	-29.71	1.403238	0.001010	2.2808	1.9307	-0.5667
0.3066	0.3539	862.03	-0.1216	1355.99	630.91	-33.93	1.409421	0.001232	1.9903	1.7157	-0.6122
0.4070	0.4595	876.62	-0.1200	1382.80	596.58	-34.37	1.415110	0.001276	1.7990	1.5770	-0.5830
0.5061	0.5593	890.27	-0.1051	1407.39	567.09	-31.84	1.420386	0.001218	1.6814	1.4969	-0.4974
0.5984	0.6486	902.47	-0.0912	1429.51	542.24	-28.02	1.425006	0.001064	1.5798	1.4257	-0.4142
0.7010	0.7438	915.41	-0.0685	1452.64	517.69	-22.02	1.429860	0.000829	1.4612	1.3376	-0.3307
0.8014	0.8333	927.58	-0.0492	1475.05	495.49	-15.49	1.434364	0.000549	1.3660	1.2671	-0.2333
0.9025	0.9198	939.31	-0.0270	1496.83	475.17	-8.05	1.438678	0.000239	1.2920	1.2136	-0.1178
0.9583	0.9661	945.55	-0.0111	1506.97	465.70	-2.65	1.441005	0.000091	1.2565	1.1881	-0.0499
1.0000	1.0000	950.13		1516.80	457.47		1.442726		1.2296	1.1683	
			N	I,N-Dimethyla	cetamide (1) + 1	-propanol (2	) at $T = 298.15$ ]	K			
0.0000	0.0000	799.77		1206.17	859.45		1.383104		2.4371	1.9491	
0.0535	0.0654	809.10	-0.0375	1224.56	824.21	-12.08	1.386842	0.000292	2.1838	1.7669	-0.1277
0.1052	0.1271	817.77	-0.0613	1241.03	793.97	-20.48	1.390328	0.000527	1.9792	1.6185	-0.2234
0.2053	0.2424	833.82	-0.0909	1270.78	742.66	-30.98	1.396724	0.000848	1.7063	1.4227	-0.3172
0.3066	0.3538	849.10	-0.0979	1298.57	698.41	-35.79	1.402744	0.000998	1.5137	1.2853	-0.3513
0.4070	0.4594	863.49	-0.0955	1324.89	659.76	-37.06	1.408356	0.001046	1.3864	1.1971	-0.3372

Table S1. Continued

-												
-	$x_1$	$oldsymbol{\phi}_1$	$ ho/\mathrm{kg}\cdot\mathrm{m}^{-3}$	$V^{\mathrm{E}} \cdot 10^6 / \mathrm{m}^3 \cdot \mathrm{mol}^{-1}$	$u / \text{m} \cdot \text{s}^{-1}$	κ <sub>S</sub> / TPa <sup>-1</sup>	$\kappa_{\rm S}^{\rm E}$ / TPa <sup>-1</sup>	$n_{ m D}$	$\Delta n_{ m D}$	v/ mm <sup>2</sup> ·s <sup>-1</sup>	η / mPa·s	Δη/ mPa·s
_	0.5061	0.5593	876.99	-0.0836	1349.24	626.37	-35.08	1.413554	0.000980	1.3037	1.1433	-0.2900
	0.5984	0.6485	889.06	-0.0726	1370.98	598.42	-31.44	1.418141	0.000868	1.2334	1.0966	-0.2426
	0.7010	0.7438	901.88	-0.0548	1393.79	570.77	-25.36	1.423005	0.000710	1.1590	1.0453	-0.1893
	0.8014	0.8332	913.93	-0.0398	1414.75	546.67	-17.80	1.427520	0.000515	1.0917	0.9977	-0.1346
	0.9025	0.9198	925.56	-0.0220	1436.03	523.93	-9.89	1.431810	0.000242	1.0424	0.9648	-0.0645
	0.9583	0.9661	931.74	-0.0085	1445.81	513.43	-3.99	1.434104	0.000096	1.0132	0.9440	-0.0284
	1.0000	1.0000	936.29		1453.68	505.42		1.435794		0.9932	0.9299	
				N	,N-Dimethyla	cetamide (1) + 1	-propanol (2	) at $T = 313.15$	K			
	0.0000	0.0000	787.54		1155.36	951.25		1.376864		1.7640	1.3892	
	0.0535	0.0654	796.67	-0.0300	1172.46	913.12	-12.47	1.380490	0.000216	1.6196	1.2903	-0.0655
	0.1052	0.1270	805.17	-0.0485	1188.16	879.75	-21.65	1.383903	0.000417	1.4979	1.2061	-0.1174
	0.2053	0.2423	820.96	-0.0720	1216.51	823.09	-33.06	1.390150	0.000652	1.3314	1.0930	-0.1679
	0.3066	0.3537	836.04	-0.0770	1243.55	773.48	-38.93	1.396089	0.000782	1.2057	1.0080	-0.1896
	0.4070	0.4593	850.26	-0.0753	1268.96	730.39	-40.55	1.401637	0.000824	1.1150	0.9480	-0.1869
	0.5061	0.5591	863.63	-0.0658	1292.78	692.83	-38.90	1.406784	0.000767	1.0467	0.9040	-0.1690
	0.5984	0.6484	875.58	-0.0570	1313.59	661.89	-34.75	1.411360	0.000687	0.9987	0.8744	-0.1409
	0.7010	0.7437	888.29	-0.0434	1335.31	631.37	-27.82	1.416199	0.000557	0.9459	0.8402	-0.1110
	0.8014	0.8332	900.25	-0.0321	1356.36	603.79	-20.20	1.420720	0.000411	0.9040	0.8138	-0.0747
	0.9025	0.9197	911.78	-0.0180	1376.83	578.56	-11.42	1.425038	0.000219	0.8649	0.7886	-0.0367
	0.9583	0.9660	917.92	-0.0073	1386.51	566.70	-5.08	1.427328	0.000095	0.8442	0.7749	-0.0156
	1.0000	1.0000	922.43		1393.35	558.40		1.429006		0.8287	0.7644	
				N	N,N-Dimethyla	acetamide (1) +	1-butanol (2)	at $T = 283.15 \text{ k}$	<b>C</b>			
	0.0000	0.0000	817.20		1291.66	733.46		1.403162		4.7279	3.8636	
	0.0565	0.0571	824.87	-0.0091	1305.34	711.49	-6.22	1.405542	0.000121	3.9758	3.2795	-0.4318
	0.1063	0.1073	831.62	-0.0165	1317.73	692.51	-11.35	1.407622	0.000215	3.5016	2.9120	-0.6651
	0.2094	0.2112	845.53	-0.0273	1341.88	656.82	-18.37	1.411864	0.000346	2.8005	2.3679	-0.9313

Table S1. Continued

<i>x</i> <sub>1</sub>	$oldsymbol{\phi}_1$	$ ho/\mathrm{kg}\cdot\mathrm{m}^{-3}$	$V^{\rm E} \cdot 10^6 / \text{m}^3 \cdot \text{mol}^{-1}$	$u / \mathbf{m} \cdot \mathbf{s}^{-1}$	$\kappa_{\rm S}$ / Tpa <sup>-1</sup>	$\kappa_{ m S}^{ m E}$ /	$n_{ m D}$	$\Delta n_{ m D}$	v/ mm <sup>2</sup> ·s <sup>-1</sup>	$\eta$ / mPa·s	Δη/ mPa·s
0.3134	0.3157	859.45	-0.0294	1365.22	624.27	-22.08	1.416019	0.000367	2.3699	2.0368	-0.9821
0.4106	0.4132	872.37	-0.0248	1386.23	596.53	-22.92	1.419824	0.000314	2.0805	1.8150	-0.9419
0.5083	0.5110	885.29	-0.0162	1407.48	570.21	-22.25	1.423589	0.000210	1.8722	1.6574	-0.8362
0.6042	0.6068	897.92	-0.0055	1428.07	546.09	-19.92	1.427239	0.000070	1.7025	1.5287	-0.7064
0.7096	0.7118	911.79	0.0035	1450.52	521.26	-15.77	1.431280	-0.000044	1.5474	1.4109	-0.5401
0.8046	0.8063	924.31	0.0072	1472.55	498.93	-12.01	1.434969	-0.000093	1.4609	1.3503	-0.3447
0.9078	0.9087	937.94	0.0051	1495.60	476.64	-6.03	1.439050	-0.000064	1.3606	1.2762	-0.1406
0.9575	0.9579	944.51	0.0025	1507.54	465.86	-3.23	1.441029	-0.000031	1.2784	1.2075	-0.0754
1.0000	1.0000	950.13		1516.80	457.47		1.442726		1.2296	1.1683	
			N	N,N-Dimethyla	acetamide (1) +	1-butanol (2)	at $T = 298.15$ H	ζ			
0.0000	0.0000	806.09		1240.60	806.03		1.397139		3.1923	2.5733	
0.0565	0.0571	813.56	-0.0037	1254.00	781.65	-7.25	1.399393	0.000047	2.7684	2.2523	-0.2281
0.1063	0.1074	820.14	-0.0064	1265.68	761.14	-12.67	1.401375	0.000084	2.4986	2.0492	-0.3494
0.2094	0.2114	833.69	-0.0083	1289.06	721.86	-20.75	1.405418	0.000107	2.0578	1.7156	-0.5136
0.3134	0.3160	847.28	-0.0056	1311.32	686.37	-24.84	1.409426	0.000072	1.7737	1.5028	-0.5555
0.4106	0.4135	859.92	0.0006	1331.09	656.34	-25.57	1.413115	-0.000008	1.5821	1.3605	-0.5380
0.5083	0.5113	872.58	0.0082	1350.26	628.58	-23.94	1.416798	-0.000105	1.4475	1.2631	-0.4749
0.6042	0.6070	884.99	0.0146	1369.91	602.12	-21.63	1.420419	-0.000184	1.3320	1.1788	-0.4016
0.7096	0.7120	898.63	0.0173	1391.67	574.57	-17.58	1.424433	-0.000228	1.2455	1.1192	-0.2879
0.8046	0.8065	910.94	0.0158	1413.36	549.55	-14.16	1.428112	-0.000202	1.1648	1.0611	-0.1899
0.9078	0.9088	924.33	0.0083	1435.78	524.80	-8.10	1.432165	-0.000104	1.0734	0.9922	-0.0892
0.9575	0.9580	930.78	0.0039	1445.62	514.10	-3.98	1.434119	-0.000051	1.0315	0.9601	-0.0396
1.0000	1.0000	936.29		1453.68	505.42		1.435794		0.9932	0.9299	

Table S1. Continued

$x_1$	$oldsymbol{\phi}_1$	$ ho/\mathrm{kg}\cdot\mathrm{m}^{-3}$	$V^{\mathrm{E}} \cdot 10^6 / \mathrm{m}^3 \cdot \mathrm{mol}^{-1}$	$u / \text{m·s}^{-1}$	κ <sub>S</sub> / TPa <sup>-1</sup>	$oldsymbol{\kappa}_{ ext{S}}^{ ext{E}}$ /	$n_{ m D}$	$\Delta n_{ m D}$	v/ mm <sup>2</sup> ·s <sup>-1</sup>	η / mPa·s	Δη/ mPa·s
			1	N,N-Dimethyl	acetamide (1) +	1-butanol (2)	at $T = 313.15 \text{ I}$	ζ			
0.0000	0.0000	794.44		1190.52	888.10		1.391074		2.2275	1.7696	
0.0565	0.0572	801.76	0.0003	1203.41	861.25	-8.10	1.393240	-0.000004	1.9930	1.5979	-0.1149
0.1063	0.1075	808.18	0.0016	1214.40	839.01	-13.84	1.395132	-0.000020	1.8278	1.4772	-0.1855
0.2094	0.2114	821.45	0.0066	1236.94	795.65	-23.07	1.399007	-0.000086	1.5671	1.2873	-0.2718
0.3134	0.3160	834.77	0.0137	1257.94	757.03	-27.31	1.402876	-0.000185	1.3771	1.1496	-0.3050
0.4106	0.4136	847.19	0.0208	1276.81	724.05	-28.17	1.406488	-0.000275	1.2549	1.0631	-0.2938
0.5083	0.5114	859.64	0.0272	1294.70	693.98	-26.01	1.410124	-0.000348	1.1608	0.9979	-0.2608
0.6042	0.6071	871.87	0.0296	1313.44	664.86	-23.56	1.413711	-0.000392	1.0898	0.9502	-0.2121
0.7096	0.7121	885.33	0.0275	1334.46	634.29	-19.45	1.417738	-0.000347	1.0258	0.9082	-0.1481
0.8046	0.8065	897.47	0.0207	1355.64	606.31	-16.21	1.421387	-0.000279	0.9639	0.8651	-0.0957
0.9078	0.9088	910.66	0.0101	1376.92	579.20	-9.44	1.425415	-0.000132	0.8925	0.8128	-0.0443
0.9575	0.9580	917.01	0.0044	1386.32	567.41	-4.92	1.427354	-0.000059	0.8608	0.7894	-0.0177
1.0000	1.0000	922.43		1393.35	558.40		1.429006		0.8287	0.7644	

<sup>&</sup>lt;sup>a</sup> Standard uncertainties u are u(T) = 0.005 K for densities and speeds of sound and u(T) = 0.01 K for the rest of properties, u(p) = 0.003 MPa,  $u(x_1) = 0.0001$ ,  $u(\rho) = 0.1$  kg·m<sup>-3</sup>, u(u) = 0.5 m·s<sup>-1</sup>,  $u(n_D) = 1 \cdot 10^{-5}$ ,  $u(\nu) = 0.005$  mm<sup>2</sup>·s<sup>-1</sup>, and  $u(\eta) = 0.005$  mPa·s.