

5<sup>th</sup> Metallurgical & Materials Engineering Congress of South-East Europe Trebinje, Bosnia and Herzegovina 7-10<sup>th</sup> June 2023



# CONGRESS PROCEEDINGS

## MME SEE

CONGRESS 2023

5<sup>th</sup> Metallurgical & Materials Engineering Congress of South-East Europe Trebinje, Bosnia and Herzegovina 7-10<sup>th</sup> June 2023

# CONGRESS PROCEEDINGS

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### INFLUENCE OF FUNCTIONAL GROUPS ON THERMODYNAMIC PROPERTIES OF BINARY MIXTURES

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The thermodynamic properties of liquid mixtures depend on the geometry, actually on the shape of the molecules and their ability to pack. In addition, the properties also depend on non-covalent interactions like Van der Waals, electrostatic interactions, hydrogen bonding and  $\pi$ - $\pi$  interactions. Each functional group in a molecule influences the geometry of the molecule and the intermolecular interactions between molecules. In this work, the functional groups of double bond and -OH group has been investigated and their influence on the properties of mixtures. The properties of mixtures (1) n-hexane + 1-hexanol and (2) n-hexane + cis-3-hexen-1-ol have been investigated both with experimental measurement and with computation molecular modeling. Experimental measurement considered measurements of density, viscosity, refractive index and speed of sound at a range of temperature from (288.15 to 318.15) K and at complete composition range. Excess molar volume and deviations of viscosity, refractive index and speed of sound has been calculated and Redlich-Kister equation has been used for fitting the data. Considering the molecular modeling, the quantum chemical calculations and molecular dynamic simulations have been performed. Different model systems are used order to investigate the -OH group energy interaction and double bond / single bond intermolecular energy interactions. The molecular dynamic simulations have been performed with the liquid mixtures.

Keywords: binary mixtures, thermodynamic properties, quantum chemical calculations, molecular dynamic simulations

#### Introduction

The behavior of liquid mixtures depends on the thermodynamic and transport properties and in order to understanding the behavior it is required the knowledge of these properties. Properties can be affected by the geometries of molecules and their intermolecular interactions. The properties can be influenced by geometry because of the different sizes and shapes of molecules and their ability to pack with each other or to cause steric repulsions. Intermolecular interactions can be caused by noncovalent interactions like Van der Waals, electrostatic interactions, hydrogen bonding and  $\pi$ - $\pi$  interactions. The functional groups of molecules participate and influence the characteristic properties, as it influences the geometry of the molecule and the non-covalent interactions. The functional groups that are studied in this paper are double bond and -OH group. To analyze these functional groups, two sets of mixtures (1) n-hexane + 1-hexanol and (2) n-hexane + cis-3-hexen-1-ol has been investigated as well as model systems of butane and 2-butene systems.

Mixtures have been experimentally measured, the thermodynamic and transport properties were determined. The same mixtures have been simulated by Molecular dynamics simulations (MD), while the smaller model systems have been used for quantum chemical calculations. Experimental measurement combined with quantum chemical calculations and molecular dynamic simulations connects the macroscopic and microscopic stages and provide insights of interactions at the molecular level. Molecular dynamic simulations can be a powerful tool for analyzing the behavior of liquid binary mixtures at the molecular level. It is a promising tool which can analyze a range of properties, such as the radial distribution functions (RDFs) of the mixture, thermodynamic and transport properties, phase transitions and interactions. RDFs can be used to study the structure of the mixture, including the distribution of different species and the formation of clusters or aggregates. Thermodynamic properties can analyze the effects of temperature and pressure on the mixtures. The interactions between different species in the mixture, including the strength and nature of the interactions can be studied with MD simulations.

This information can be used in order to understand the stability of the mixture and to predict its behavior under different conditions.

Overall, MD simulations help to understand the nature of interactions, which obtain insight into the local structure and dynamics of the mixture.

The goal of this paper is to analyze the influence of functional groups: double bond and -OH group on thermodynamic properties of binary mixtures. The mixtures of investigation are chosen to be hydrocarbons and alcohols. Liquid binary mixtures are investigated by combining experimental measurements and molecular dynamic simulations, while for quantum chemical calculations model system were used to investigate interactions of double bonds. The focus of this paper is the molecular dynamic simulations and their compatibility with the experimental measurements. Molecular dynamics simulations can give a deeper understanding of the behavior of liquid mixtures at the molecular level, which can make predictions of the behavior at other conditions and help in optimization of processes in industry.

#### Materials and methods

Substances n-hexane (>99 mas%) was provided by Merck and 1-hexanol (98 mas%) and *cis*-3-hexen-1-ol (>98 mas%) were provided by Sigma Aldrich. The details about the experimental preparation are given in previous work (Zarić, et al. 2020). Experimental data of pure substances n-hexane and 1-hexanol are in good agreement with literature data, all compared at temperate of 288.15K (Zarić et al. 2020).

Anton Paar DSA 5000 M density and sound velocity meter was used for measuring the densities of pure substances and their mixture with an accuracy  $\pm 5 \times 10^{-3}$  kgm<sup>-3</sup> for density. Anton Paar Stabinger SVM 3000/G2 viscometer was used for measuring the viscosity with accuracy of  $\pm$  0.1 %.

Theoretical quantum mechanical calculations were performed using Gaussian 09 (version D.01) (Frisch et al. 2013.). The single point energies were calculated creating a potential curve and for each of the investigated system and at minima of each potential curve the optimized geometries and energy of interaction are analyzed. The interaction energy at the CCSD(T)/CBS level was calculated using the extrapolation scheme proposed by Mackie and Dilabio (Mackie et al. 2011.).

Molecular dynamic simulations were performed in GROMACS (Wang et al. 2006, Bussi et al. 2007, Hess, 2008). The results of molecular dynamic simulations depend on the employed force fields, and in this work, the Amber Force Field was applied. For the generating the Amber Force Field, the molecular coordinates are obtained by quantum chemical calculations, and the topology and parameters were generated by ANTECHAMBER and the xleap AMBER tools (Spoel et al. 2005). The box was set up as mixtures of 1000 molecules in a cubic simulation box with dimensions of 6x6x6 nm side length at start. Each box was set up with different sets of concentration of mixtures and at different temperature following the experimental measurements. Each box was originally filled with molecules at random positions with PACKMOL (Martínez et al. 2009, Martínez et al. 2003). The cutoff for van der Waals is set to 1.2 nm and for the short ranged electrostatic interactions, together with Particle-Mesh-Ewald treatment (on a 0.16 grid). The steepest descent minimization of 500 steps and short equilibration runs of 100 ps length were performed at the target temperature using the V-rescale thermostat. For NPT ensemble V-rescale thermostat was used for temperature and the Parrinello-Rahman barostat for pressure. The NPT production runs of 1 ns length at the target temperature and pressure were performed. All bonds hydrogen atoms were constrained by LINCS. Trajectories were saved every 2fs. Visualization and analysis of the MD trajectories was done with Visual Molecular Dynamic (VMD) (Humphrey et al. 1996).

#### **Results and discussion**

The experimental measurements of density and viscosity were done in the whole range of concentration of binary mixture, at a range of temperatures from (288.15 to 318.15) K with a step of 5 K at atmospheric pressure. From density, excess molar volume was calculated:

$$V^{E} = \sum_{i=1}^{N} x_{i} M_{i} \left[ \left( 1/\rho \right) - \left( 1/\rho_{i} \right) \right]$$

$$\tag{1}$$

*N* is the number of components,  $x_i$  is the mole fraction,  $M_i$  is the molecular mass of component i and  $\rho$  and  $\rho_i$  represent the measured densities of a mixture and the pure component i, respectively. From the viscosity, the viscosity deviations,  $\Delta \eta$ , was calculated:

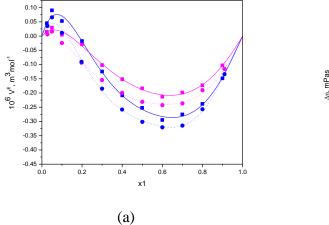
$$\Delta \eta = \eta - \sum_{i=1}^{N} x_i \eta_i \tag{2}$$

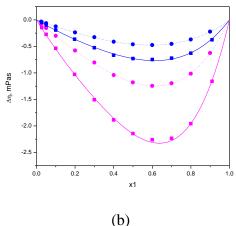
where  $\eta$  and  $\eta$ i are the measured viscosities of a mixture and the pure component i, respectively. For correlating the calculated values of excess molar volume  $V^E$  and viscosity deviations  $\Delta \eta$ . Redlich-Kister (RK) polynomial equation (Redlich et al. 1948) was used

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

where  $Y^E(\Delta Y)$  represents  $V^E$  or  $\Delta \eta$ , and Ap is the the adjustable parameters of the property, and the degree of adjustable parameters (k + 1) is determined applying the F-test.

In Figure 1 the excess molar volume,  $V^E$  (Figure 1a) and viscosity deviation,  $\Delta \eta$  (Figure 1b) for the two binary mixtures: 1-hexanol + n-hexane (square symbol and full lines) and cis-3-hexen-1-ol + n-hexane (circle symbol and dotted lines) are represented. The results are shown for only two temperatures, the lowest: at 288.15 K (violet color) and at the highest, 318.15K (blue color).





**Figure 1** Results of experimental data of (a) excess molar volume  $(V^E)$  and (b) viscosity deviations  $(\Delta \eta)$  for mixtures: 1-hexanol (1) + n-hexane (2) (- $\blacksquare$ -, - $\blacksquare$ -) and cis-3-hexen-1-ol (1) + n-hexane (2) (- $\blacksquare$ -, - $\blacksquare$ -) at 288.15 K ( $\blacksquare$ ,  $\bullet$ ) and at 318.15 K ( $\blacksquare$ ,  $\bullet$ ) and at atmospheric pressure. Symbols represent experimental data, lines represent the calculated values by RK equation (Eq. 3.)

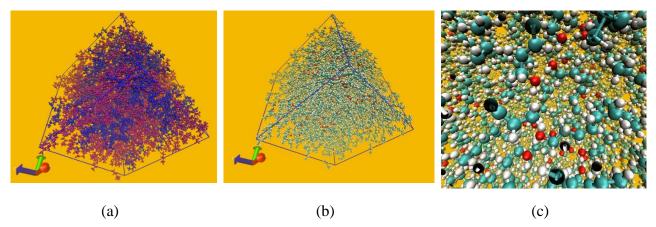
Data in Figure 1a show that the excess molar volume has a typical "S" shaped curve for hydrocarbons and alcohols. The negative values for molar content of alcohol higher than the  $x_I$ =0.2, are the consequence of the self-association of alcohol molecules. This behavior is expected for polar molecules with tendency to form hydrogen bonding between the -OH groups.

At the Figure 1b, the values of viscosity deviations are negative in the whole range of composition.

The values of deviations are different in Figures 1a and 1b, indicating that the interaction energies in the mixture between the cis-3-hexen-1-ol and n-hexane, where the double bonds are present, are stronger.

These interaction energies can be obtained by quantum chemical calculations. The quantum chemical calculations are performed in vacuum. The interactions are performed on model system: for single bond/single bond interactions the butane/butane system, and for double bond/single bond interaction the butane/2-butene system. The results of quantum mechanical calculations are obtained at very accurate CCSD(T)/CBS level. The calculated interaction energies show that the higher energy interactions are in systems of double/single bond (2-butene and butane) with interaction energies between (-2.50 to -2.82) kcal/mol, while the interaction energies in systems of single/single bond (butane dimer) are between (-2.44 to -2.58) kcal/mol. These calculations are confirming the analysis from experiments indicating stronger interactions in the mixture with double/single bond (cis-3-hexen-1-ol + n-hexane) than the mixture with single/single bond (1-hexanol + n-hexane).

In molecular dynamic simulations the conditions were the same as in the experimental measurements. Three temperatures are simulated, the lowest, 288.15 K, then at the room temperature at 293.15 K, and the highest experimental temperature at 318.15 K. For each temperature, different compositions (3%-97%, 10%-90%, 60%-40% and 80%-20%) have been reached with PACKMOL for both binary mixtures.



**Figure 2** Simulation box by VMD, mixture of 40% cis-3-hexen-1-ol and 60% of n-hexane at T=318.15K (a) Colors by molecule: cis-3-hexen-1-ol - violet color, n-hexane: blue color; (b) Colors by atoms: H – white, C – blue, O – red; (c) Inside the simulation box

Figure 2 shows the representation of simulation box via VMD. The same mixture of 40% cis-3-hexen-1-ol and 60% of n-hexane is represented in three different ways. The first Figure 2a represents the each of the two substances, with two different colors. These colors enable to follow the mixing of the two substances in time. On Figure 2b the same mixture box is represented by coloring the atoms: H atoms white, C atoms blue and O atoms are red color. The Figure 2c represents the Figure 2b zoomed in, insight of the look of inside the box.

The densities after each of the simulation runs are represented in Table 1. Calculating the mean absolute percentage error, all simulated densities are between 0.22 % and 2.21 %. This is in a good agreement and shows a valid representation of the experimental mixture.

**Table 1:** Comparison of density gained by experimental measurements and by simulations at four different composition rate and at three temperatures (288.15 K, 293.15 K, 3118.15 K) and the mean absolute percentage error for both mixtures: 1-hexanol + n-hexane and cis-3-hexen-1-ol + n-hexane

Composition (%)		Experimental measurement	Simulation		Composition (%)		Experimental measurement	Simulation	
1-hexanol	n-hexane	$\begin{array}{c} \rho_{exp} \\ [kgm^{-3}] \end{array}$	$\rho_{sim} \\ [kgm^{-3}]$	% error	cis-3- hexen-1-ol	n-hexane	ρ <sub>exp</sub> [kgm <sup>-3</sup> ]	$\rho_{sim} \\ [kgm^{-3}]$	% error
T = 288.15 K									
3	97	668.49	658.029	1.56	3	97	668.50	658.217	1.54
10	90	679.68	669.806	1.45	10	90	681.63	669.938	1.72
60	40	759.22	754.511	0.62	60	40	759.22	764.802	0.74
80	20	791.16	789.412	0.22	80	20	814.30	806.232	0.99
T = 293.15 K									
3	97	664.00	653.747	1.54	3	97	664.04	652.652	1.71
10	90	675.23	664.914	1.53	10	90	677.15	665.674	1.69
60	40	755.24	750.064	0.69	60	40	770.92	754.685	2.11
80	20	787.41	783.964	0.44	80	20	810.37	801.642	1.08
T = 318.15  K									
03	97	640.91	626.745	2.21	03	97	640.91	628.044	2.01
10	90	652.30	640.878	1.75	10	90	654.05	639.724	2.19
60	40	734.84	728.961	0.80	60	40	749.86	736.31	1.81
80	20	768.16	764.715	0.45	80	20	790.27	777.958	1.56

#### Conclusion

The influence of functional groups, the -OH group and double bond, on properties of liquid binary mixtures have been investigated. Experimental measurements on two binary liquids: 1-hexanol + n-hexane and cis-3-hexen-1-ol + n-hexane were performed. The properties of these mixtures were also investigated by quantum chemical calculations and molecular dynamic simulations. From experimental measurements, the excess molar volume and viscosity deviations were calculated. Difference in the viscosity deviations, indicates difference in the interaction energies in the two mixtures. These have been confirmed with quantum chemical calculations on interaction energies using model systems. The data show that interactions between single and double bods are stronger, than interactions between two single bonds. The MD simulations have been performed on different temperatures and different compositions and all the results from simulations show good agreement of density with the experimental data. The results presented in this work show that the methodology combining two theoretical methods, quantum chemical and molecular dynamics with experimental measurements gives opportunity to obtain, understand and predict data on liquid mixtures.

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