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EXCESS PARAMETERS OF BINARY MIXTURES OF ACETONITRILE-DIMETHYL SULPHOXIDE

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ABSTRACTS

The excess parameters associated with liquid mixes of acetonitrile-dimethyl sulphoxide were analyzed to estimate the degree of intermolecular interactions in the binary system. The viscosities and densities of the binary system were determined at temperatures of 298.15, 303.15, 318.15, 328.15, and 338.15K, respectively, with a percentage composition of 0 to 100 percent. Using the experimental density and viscosity measurements, the excess molar volumes (V_m^E), excess viscosity (η^E), excess Gibbs' free energy of activation of viscous flow (G^{*E}), and Grunberg-Nissan interaction parameters were calculated. Deviations from these parameters studied in the composition of the mixtures and temperature were discussed in terms of molecular interactions in these mixtures. A comparison of several interaction characteristics was expressed to better understand the intermolecular interactions between acetonitrile and dimethyl sulphoxide. As the temperature of the system and the mole fractions of acetonitrile rise, the viscosity and density of the experimental values drop. According to the findings, the excess parameters were mostly negative, indicating that dipole-dipole interactions predominated in the acetonitrile-dimethyl sulphoxide solvent mixture.

Keywords: Acetonitrile (ACN), Nitrobenzene (NB), Dimethyl Sulphoxide (DMSO), density, viscosity, Excess parameters.

INTRODUCTION

The densities and viscosities of a solvent mixture are important properties, particularly for the chemical project and for the optimization of chemical production. In many industries, studying binary solvent properties has many important roles and also governs their interest in systems such as organic synthesis, ion extraction systems, gas adsorption solvents, mass transfer phenomena, etc (Orjiocha *et al.* 2022). Excess thermodynamic experiments and transport properties for binary mixtures provide valuable knowledge about a deeper understanding of the composition of the molecular liquid and intermolecular interactions (Yang *et al.* 2009). Acetonitrile has vast industrial applications; it is used in the personal care industry, cosmetic products as well as pharmaceutical industries as an active ingredient. It is used also in photographic filmmaking and in battery applications. Acetonitrile has environmental applications and is also used in the textile, electronics as well as petroleum industries. On the other hand dimethyl sulphoxide (DMSO) is equally a very important solvent that has many industrial applications such as in the polymer, paper, cleaning, and pharmaceutical industries. The stability of the solvated complex is been affected by the density of coordinating solvent molecules (Ng.Van *et al.* 1995). In the formulation of electrolytes for industrial electrochemical processes, the viscosity is known as the transport property, and the dielectric constant which is the bulk property is a very important parameter (Sujata & Sunil 2013; Logan et al. 2018).

Researchers have stated that low viscosity property of solvent helps to reduce the ion transport resistance in the solution; whereas the high dielectric constant contributes to the reduction of ion dissociation energy (Matsuda et al. 1981 & Ding et al. 2000). In the chemical manufacturing industry, the thermodynamic properties of binary mixtures have a great deal of application. The insight into thermodynamic properties and volume characteristics of binary solvent mixtures will help in the interpretation of the processes that occur in such mixtures.

Data obtained from the excess molar volumes of constituent mixtures are well needed in electrochemical and engineering processes which covers the field of mass transfer, fluid flow, and heat transfer. The volume output of components of the mixture, such as structural changes, and intermolecular attraction or repulsion between unlike molecules, can be influenced by the behavior of the mixing model (Wang et al. 2004). A thorough analysis of the viscosity behavior of liquid mixtures would provide a deeper understanding of a system's transport properties. The properties susceptible to various kinds of the association are excess molar volume and excess viscosity. Using these properties, molecular packing, molecular motions, and different kinds of intermolecular interactions have been studied. However, the scale, shape, and chemical composition of the component molecules affect these properties (Gurung & Roy 2006; Palaiologou 1996). For several reasons, many aprotic solvents with large dipole moments have been studied by several researchers, including the study of the molecular packaging of such liquids, the energy of the ion-solvation phase, and the solvent effect on reaction rates (Grolier et al. 1991; Wilhelm et al. 1981; Grigoryan et al. 2017). It is equally important to note that researchers have studied the thermodynamic properties of a binary mixture of ACN with DMSO (Grolier et al. 1991). Given this implication, it becomes imperative to us to investigate the binary mixtures of ACN-DMSO to comprehend the connections among these components to obtain and compare their excess properties.

MATERIAL AND METHODS

All the chemicals used were of analytical grade and were used without further purification. The solvent used is DMSO (99%), ACN (99%), and were obtained from Loba Chem Pvt.Ltd. Lithium chloride (LiCl 99%) was obtained from Molychem. Binary mixtures of ACN-DMSO were prepared with proportions of 0, 15, 25, 50, 75, 85, and 100 percent ACN in varying volumes. These were prepared by mixing suitable solvent volumes in regular flasks with caps. The mass measurements were then carried out using AdventureTn OHAUS AR3130 (China) with 0.001g readability. The fractions of the mole vary from 0.192 to 1.00 ACN. At temperatures of 298.15k, 308.15, 318.15, 328.15, and 338.15K, the density of pure solvents and mixtures was measured using a Capillary Pycnometer bottle. Ubbelohde Viscometer tube was used to determine the kinematic viscosity of the pure solvents and mixtures. To maintain and monitor the temperature of the solvent and the mixture at the appropriate temperatures, a Uniscope SM801A laboratory water bath (England) was used. Using a digital stopwatch, the flow time was determined. Equation (1) (Ibezim-Ezeani et al. 2017) measured the kinematic viscosity.

$$v = kt \qquad (1)$$

Where the: v = kinematic viscosity, k = viscometer constant and t = Flow time.

From equation (1) the kinematic viscosity was converted to dynamic viscosities using equation (2)

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

Where η = dynamic viscosity, ρ = the density of the liquid.

Excess properties include: the excess molar volume (V_m^E), excess viscosity (η^E), Excess Gibbs free energy of activation of viscous flow (G^{*E}), and the Grunberg-Nissan interaction parameter (d) was calculated using equation 3 to 7.

$$V^E m = X_1 M_1 \left(\frac{1}{\rho_m} - \frac{1}{\rho_1}\right) + X_2 M_2 \left(\frac{1}{\rho_m} - \frac{1}{\rho_2}\right) \qquad (3)$$

$$\eta^E = \eta_M - (X_1\eta_1 + X_2\eta_2) \qquad (4)$$

$$V_M = \frac{X_1 M_1 + X_2 M_2}{\rho_m} \qquad (5)$$

Where

X_1 and X_2 = Mole fraction of pure constituents of ACN and DMSO respectively

M_1 and M_2 = Molar masses of pure constituents of ACN and DMSO respectively

ρ_1 and ρ_2 = Densities of pure constituents ACN and DMSO respectively

ρ_m = Density of the binary mixture (ACN-DMSO)

η^E = Excess Viscosity (ACN-DMSO)

η_m = Viscosity of the mixture (ACN-DMSO)

η_1 and η_2 = Viscosity of pure constituents ACN and DMSO respectively

$$G^{*E} = RT[\ln\ n_m V_m - (X_1 \ln n_1 V_1 + X_2 \ln n_2 V_2)] \qquad (6)$$

$$d = \ln n(\text{mix}) - \left(\frac{X_1 \ln n_1 + X_2 \ln n_2}{X_1 X_2}\right) \qquad (7)$$

Where, R = the universal gas constant, T = absolute temperature, V_1 and V_2 molar volume of pure constituents ACN and DMSO, V_m = Molar volume of the mixture (ACN-DMSO),

d = Grunberge-Nissan interaction parameters.

RESULTS AND DISCUSSION

Density

As the percentage volume of ACN increases and that of DMSO decreases, the density of the ACN-DMSO binary mixture decreases. This decrease in density is due to the inter-molecular order disruption effect and their interaction within the binary mixture (Obowu et al. 2018). The intermolecular dipolar forces within DMSO are disturbed as the volume proportion of ACN in the binary system increases, creating weaker interactions between the DMSO and ACN molecules. Consequently, after mixing, the molecules migrate relatively apart, leading to a decrease in the solvent mixture's density. As the mole fractions of ACN increased from 0.192 to 1.00 and that of DMSO decreases from 1.00 to 0.124, the density of the mixture drops at all temperatures (at 298.15k, 1.051 to 0.801 g/cm; at 308.15 k, 1051 to 0.791 g/cm; at 318.15k. 1.042 to 0.779 g/cm; at 328.15k, 1021 to 0.761 g/cm and at 338.15k, 1.021 to 0.761 g/cm). Furthermore, as temperature increases from 298.15 to 338 .15K, the density of the system ACN-DMSO decreases at different molar volumes and displays a small rise at 318.15k of mole fraction of 0.884 and 328.15k of 0.573-mole fraction and drops at a higher mole fraction of 1.00 (Figure1). However, comparing the density effect of pure ACN on temperature changes from 253.0 K – 233.15 K as shown in the works of literature with the temperature change of mixed ACN-DMSO from 298.15k -338.15k, most of the studies shows that the density of pure ACN drops (826.6kg/m^3 – 735.8kg/m^3) linearly as the temperature increases progressively (Armenskaya & Lanshina 1973; Khimenko & Gritsenko 1980; Davydova 1986; Orjiocha et al. 2022, and Moumouzias 1991). In the ACN-DMSO mixture studied, it was observed that there are fluctuations of little rise and fall in the density at different mole fraction at each temperature which is believed to be the result of DMSO addition (Fig 1). Furthermore, a certain degree of stress is added as the temperature increases which accounts for the change in the system's internal energy. This external effect on the forces of molecular attraction distorts the solvent-solvent dipole's orientation, which is seen at higher device temperatures in the decrease in density.

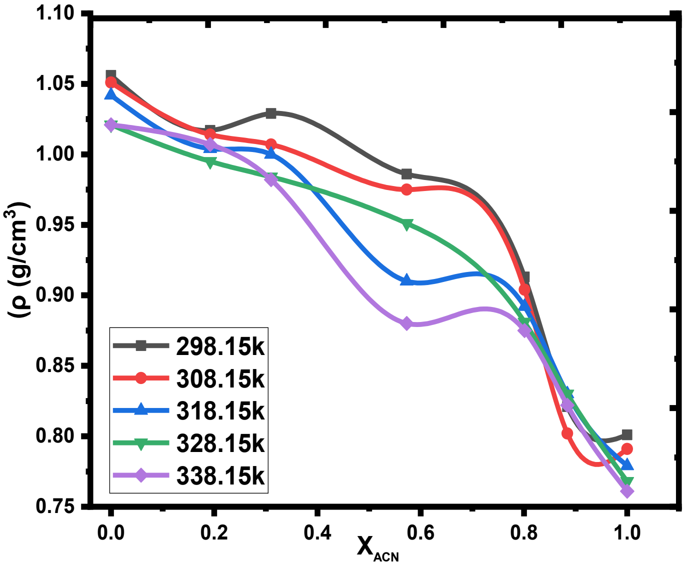

Figure 1: Density at defrent temperature against mole fractions (ACN-DMSO)

Viscosity

The increase in system temperatures will affect molecular interactions within fluid molecules. The increase in temperature of binary mixture (ACN-DMSO) raises the average molecular velocity, leading to a decrease in the amount of time spent in contact with adjacent molecules. The average intermolecular forces decrease at elevated temperature, which accounts for a decrease in the viscosity of the ACN-DMSO mixture with an increase in temperatures (Figure 2). Due to the lower viscosity of the solvents, the viscosity of the binary mixture also decreased as the ratio of ACN in the mixture increased. The viscosity of the ACN-DMSO decreases linearly as the temperature and mole fraction increases but has a little upsurge in viscosity at volume percent of 25% ACN and 75% DMSO which was observed at all temperature. Relating the study to some pieces of literature that studied the viscosity of pure ACN, the viscosity drops consistently as the temperature increases (Davydova et al. 1986); Ritzoulis et al. 1986); Moumouzias et al. 1991, & Dymond 1991). Therefore, these fluctuations are observed at the mole fraction of 0.310ACN and 0.69 DMSO at all temperature may be termed the DMSO effect.

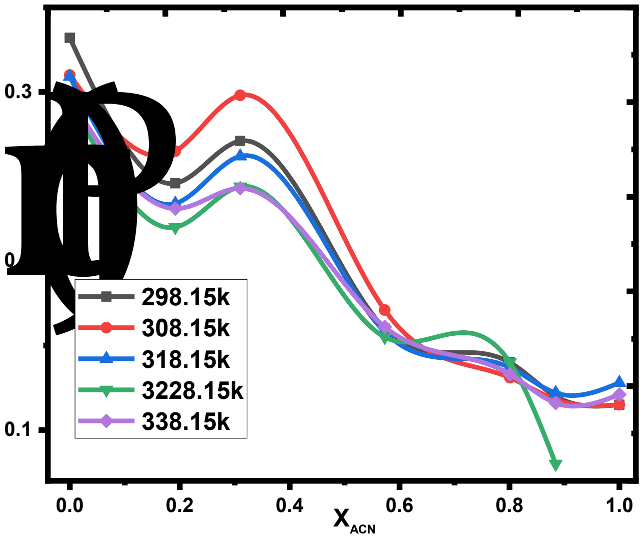

Figure 2: Viscosity of ACN-DMSO at all temperatures against mole fraction.

Excess parameters

Excess properties are the measure of variation between the property values of the thermodynamic solution and that of the ideal solution under parallel temperature and composition conditions. The positive or negative excess of thermodynamics, compared to the optimal solution, reflects this degree of variance (Mesquita et al. 2014). For optimal mixing, excess parameters have zero value. Non-ideal behavior in mixtures is indicated by negative or positive excess parameter values, suggesting the presence of complex interactions between the ACN-DMSO mixtures.

Dispersive forces are attributed to positive excess volume values, whereas dipole-dipole interactions and hydrogen bond formation are attributed to negative values (Sujata & Sunil 2013). Due to strong interactions between unlike molecules, negative values of excess volume occur while positive excess volume occurs from strong interactions between like molecules, suggesting breakup interactions between, unlike mixture molecules. The effects of the hydrogen bond rupture and loosening of dipole interactions are positive values of excess volume (El-Banna, 1997). The non-zero effects of the excess parameter values in this study point to the presence of interactions between the binary mixture components (Figure 3). Based on the sign and magnitude of excess parameters, various workers have explained the nature and strength of these interactions (Sati et al. 1993). In the ACN-DMSO investigation, the disruption of intermolecular dipolar forces in DMSO by the addition of ACN is one factor that may lead to excess molar volume. As the mole fractions increase, the V_m^E values for the mixtures (Figure 3) gives positive results with decreasing values and negative results at an increased volume. This was never the same as the temperature increases. It varies at different mole fractions under different temperatures. At 298.15k temperature, the result was positive, from the mole fractions of 0.00 – 0.192ACN, negative at 0.310 – 0.801mole fraction, 0.884 positive, and negative at 1.00ACN. Again, at the temperature of 308.15k, from 0.00 – 0.310 mole fraction were negative, 0.573 – 0.801 positive, while 0.884-mole fraction is positive and at 1.00 it is negative, at the temperature of 318.15k, it gives positive results at lower mole fractions and negative as the mole fraction increases, (0.00 - 0.573mole fraction was positive while it is negative from 0.801 – 1.00 mole fraction). At 328.15k temperature, it fluctuates; from 0.00 - 0.310 mole fractions it is positive, from 0.573 – 0.884 it is negative, and at 1.00, it is positive. Then at the higher temperature of 338.15k and from 0.00 – 0.573mole fraction is positive, at 0.801 it is negative, and 0.884 – 1.00 it becomes positive. The excess molar volume fluctuates which indicates non-ideal mixing of the binary mixture. The positive outcomes that occur more at the lower mole fraction (0 - 0.310) are due to the influence of dispersive forces that can arise from strong interactions between similar molecules. The negative outcomes that occur at the higher mole fractions (0.573 - 1.00) of ACN are caused by dipole-dipole interactions that demonstrate intense interactions between disparate molecules.

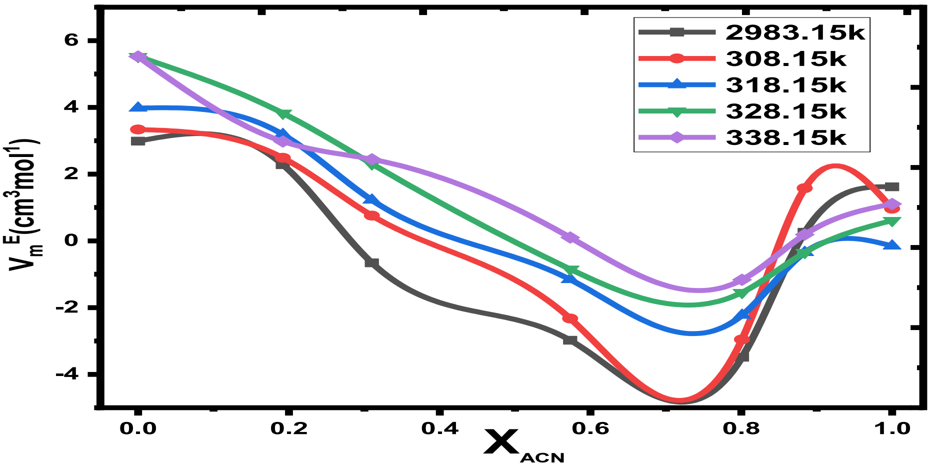

Figure 3: Molar excess volume versus mole fraction for ACN-DMSO.

Intermolecular interactions between different molecules tend to prevail over intermolecular interactions between similar molecules. The substitutional accommodation of one variable in the structure of the other is not sufficiently favorable. Thus, the positive and negative values of V_m^E are primarily responsible for dipolar-dipole interactions and dispersion forces between ACN-DMSO molecules (Prakash & Dlip, 1994).

Excess viscosity, according to Fort and Moore (1966), the negative values of η^E indicate the presence of dispersion forces; hence, the negative deviation of η^E indicates that ACN-DMSO system dipole interactions are more likely. From the findings excess viscosity (ACN-DMSO) was all negative as the mole fractions increased from 0.192 to 1.00 and were the same at all temperatures (Figure 4), showing the possibility of dipole activity in the system.

The help of excess Gibbs free energy of activation of viscous flow (G^{*E}) and the Grunberg-Nissan interaction parameter (d) is necessary to understand the forces acting between the unlike molecules of ACN-DMSO. The findings indicate that in all mole fractions and temperatures, excess Gibbs free energy was all negative (Figure 5). The negative values of excess Gibbs free Energy of viscous flow specify the dominance of dispersion forces characterized by a decrease in the internal energy of viscous flow; revealing the occurrence of the weak interface in the systems. This demonstrates a weak relation between ACN-DMSO molecules that can trigger a less viscous mixture. Positive deviations in the values of excess flow for the energy of activation suggest particular interactions between the mixture components (Sathyanarayana et al. 2007).

Grunberg–Nissan interaction parameter proves the strength of interactions between the molecular systems. From the system (ACN-DMSO) studied, the d-parameters are negative at the first two-mole fraction of 0.192, 0.310 and positive at the mole fraction of 0.573 to 1.00 (Figure 6) the negative values of the d-parameter as observed shows the existence of weak interactions between the ACN-DMSO molecules and the positive values indicates that the interactions between disparate molecules of ACN-DMSO are stronger. This result has a close relationship with some solvent works done by some researchers (Sathyanarayana et al, 2007).

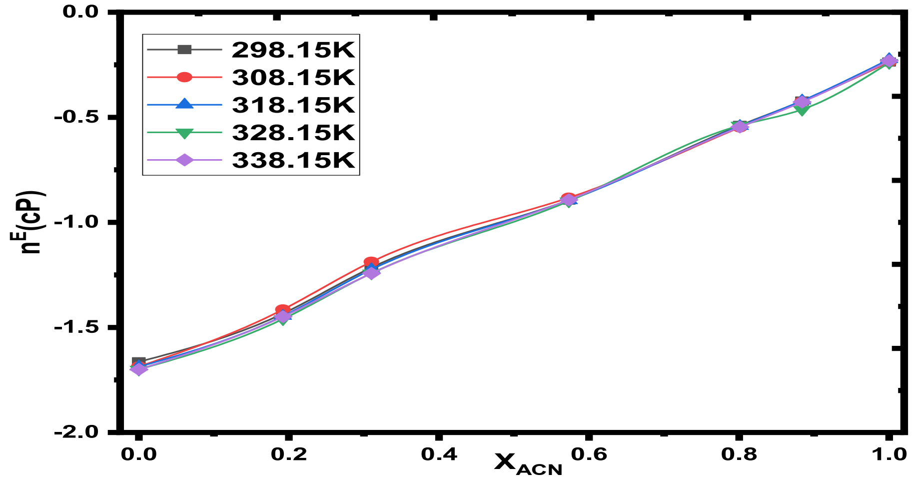

Figure 4: Excess viscosity against mole fraction of ACN-DMSO at all temperature

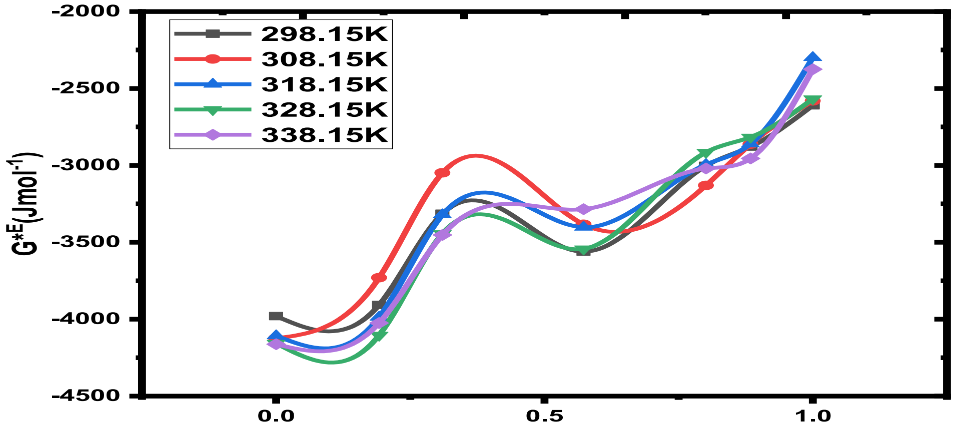

Figure 5: Plot of Gibbs free energy of activation against molar fraction of ACN-DMSO

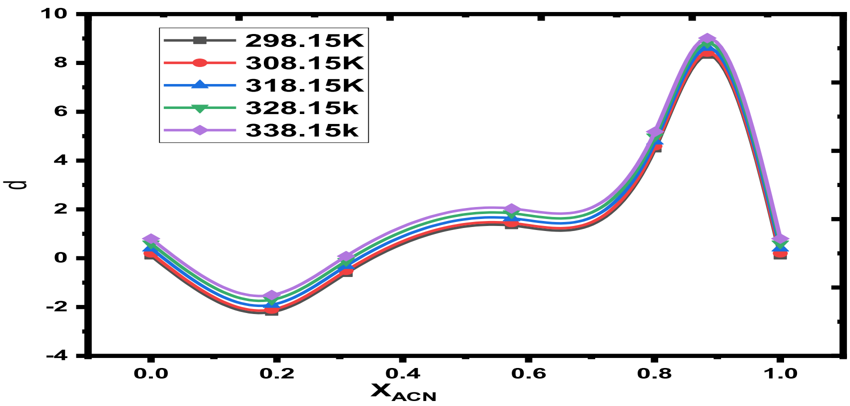

Figure 6: d-parameters against mole fraction of ACN-DMSO

CONCLUSION

The Excess parameters studies of ACN-DMSO binary mixtures have revealed their usefulness in understanding the nature of intermolecular interactions between two liquids. Some parameters such as density and viscosity of pure solvents and binary liquid mixture were measured at temperature (298.15, 308.15, 318.15, 328.15, and 338.15k) over the composition range of 0 - 100%. This provides statistics on thermodynamic properties associated with heat and fluid flow. The density and viscosity of the system decrease as the temperature, as well as the mole fractions increase with a little upsurge in viscosity volume percent of 25:75 ACN-DMSO. Dipolar and dispersion forces in ACN-DMSO molecules are principally responsible for negative values of V_m^E and η^E, and it appears that intermolecular interactions between unlike molecules predominate over that of like molecules. The perusal of the sign and magnitude of G^{*E} and d-parameters indicated that interactions between dissimilar molecules in the binary systems studied followed the normal order.

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