

Excess Parameters of Binary Mixtures of Nitrobenzene-Dimethyl Sulphoxide (Nb-Dmso)

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Received: 22 March 2022/Accepted 18 October 2022/Published online: 22 October 2022

Abstract: Recently there is an increased quest for alternative forms of electrolytes outside the conventional acid-based electrolytes in battery cell technology because of the health, environmental and cost, implications of acid. The present study explored the possibility of using organic solvent as an alternative electrolyte in battery cell technology by studying those excess parameters which are related to the liquid mixtures of nitrobenzene-dimethyl sulphoxide with a mandate to determine the intensity of the molecular interactions of the binary mixed system. The viscosities, densities, conductivity and pH of the binary solvent were determined using the percentage composition of 0 to 100 % at the following temperature range, 298.15, 303.15, 318.15, 328.15, and 338.15K respectively. Other parameters such as the molar volume (V_m), 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 (d), were determined using the experimental values of the density and viscosity. For the molecular interaction of the mixtures studied, the deviations from the parameters in the composition of the binary mixture and temperature were also investigated. There was a decrease in the pH and an increase in the viscosity and density of the results obtained from the experiment as the system temperature and the mole fractions of nitrobenzene increased. The excess parameters were all negative values indicating the dominance of dipole-dipole interactions within nitrobenzene-dimethyl sulphoxide solvent mixture. The investigation indicated that the solvent mixture

behaviors support the need to use the solvents as an alternative electrolyte in battery cell technology.

Keywords: Electrolytes, battery, excess parameters, nitrobenzene and dimethyl sulphoxide.

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1.0. Introduction

There is an emerging interest in the global community in the applications of binary mixtures in both chemical and biological industries due to their physiochemical, bulk, thermophysical, transport and dielectric properties (Ali and Nain, 2002; Nain, 2008; Dubey and Shama, 2008, and Nowosielki *et al.*, 2022). Studies (Venkatesu and prabhakaruRao, 1999; Nain, 2008; Dubey and Shama, 2008, and Nowosielki *et al.*, 2022), showed that combined solvents mixtures are better in the application when compared to a single pure solvent, and give more practical importance when it comes to chemical and industrial processes because they make available wide range of mixed solvents in varying proportions that allow constant modification for preferred properties of the system. The study of binary solvents uses thermodynamic parameter valuations. Thermodynamics is one of the fundamental subjects that have great importance in science and engineering (Ali and Nain, 2008; Raghuram *et al.*, 2015, and Obowu *et al.*, 2018). Some of the fundamental thermodynamics parameters are volume,

pressure, temperature, enthalpy, entropy, and Gibbs free energy of activation of viscous flow, Grunberg-Nissan interaction parameters, amongst others (Raghuram *et al.*, 2015).

Our quest for binary solvent studies is geared toward a suitable replacement of electrolytes solutions in battery cells with organic solvents that have relatively high density, low viscosity, and high dielectric constant among others. Such solvents will help to produce a better battery capacity which will be easy to recharge, reuse and maintain. Many scientists have studied chemical interactions in many binary organic solvent mixtures based on some parameters such as the molar volume, excess molar volume, and excess density; excess Gibbs free energy of activation to mention a few (Nikam and Neena, 2004, Kapadi *et al.*, 2003). All these excess parameters are computed from the direct experimental measurement of the density and viscosity. The derived experimental data are applied in battery cell technology, process engineering and design applications, biological industries and other related areas.

The solvent mixture characteristics mostly depend on its local structure, expressed in terms of packing density, free volume, or radical distribution function (Mahan *et al.*, 2002, and Gonzalez *et al.*, 2007). The local structures which depend on forces between molecules and their forms cause the volume of molecules to change with compositions. In this work, we used a conductivity meter, pH meter, viscosity meter, density bottles and other related apparatus to determine both the physicochemical and electrochemical properties of the system. The properties deduced included: density, viscosity, excess molar volume, excess viscosity, and excess permittivity, Gibbs free energy of activation of viscous flow and Grunberg-Nissan interaction parameters of the pure and mixed solvent. At temperatures of 298.15, 308.15, 318.15, 328.15, and 338.15 K, the intermolecular interactions of binary mixes (NB-DMSO) and



the excess parameters associated with the mixtures were explored for composition ranges of 0, 15, 25, 50, 75, 85, and 100 percent.

2.0. Materials and Methods

Dimethyl sulphoxide (DMSO) 99 % (AR JHD), nitrobenzene 99% (Qualikems), Distilled water and Roll of magnesium stripe wire. Standard Volumetric Flask, Ubbelohde Viscometer tube, Capillary Pycnometer bottle, Volumetric pipettes, and Retort stand, Electronic weighing balance (adventure OHAUS AR3130) with readability 0.001g, Laboratory water bath (Uniscop SM801A England), Thermometer, Electronic timer, Conductivity Meter (JENCO vision Plus model 3175-307A), pH meter (Jenway model 3510). All the solvents used for this analysis were of analytical grade and were used without further purification.

2.1. Preparation of the binary mixture

The binary NB-DMSO mixes were made in volume percent of 0, 15, 25, 50, 75, 85, and 100 percent. The first sample was prepared by pouring 50 ml DMSO into a 100ml volumetric flask and marked as 0% NB. Another fresh 100ml volumetric flask was obtained and into this was poured 42.5 ml DMSO and 7.5 ml NB. The flask was shaken to homogenize the mixture and the sample was marked as 15% nitrobenzene. The same procedure was repeated for constituting the remaining samples in the order of 25, 50, 75, 85 and 100 % NB. The mole fraction ranged from 0.109 to 1.00.

2.2. Density determination

A 20 ml density bottle was used to determine the density of the binary mixtures. The density of samples marked 0 to 100 % was determined by determining the weight (W_1) of the density bottle using an electronic balance. The sample mixture was poured into the density bottle and the weight (W_2) was measured and taken. The weight of the liquid sample (W_3) was calculated by subtracting W_1 from W_2 . The

density of the mixture was evaluated through equation 1.

$$\rho = \left(\frac{W_2 - W_1}{V} \right) = \frac{W_3}{V} \quad (\text{g/cm}^3) \quad (1)$$

where ρ is density (g/cm^3), W_3 is mass (g) and V is volume (cm^3).

2.3. Viscosity determination

The viscometer tube was properly mounted vertically to determine the viscosity of the mixture at a regulated temperature with the aid of a water bath. About 20 ml of the sample was introduced through the inlet tube of the viscometer to the lower reservoir. A sufficient sample was introduced to bring the liquid level between the marked lines of the lower reservoir. The viscometer holder was fitted to the viscometer tube and lowered into the water bath. The sample was allowed to attain the required bath temperature. The flow time was measured as the liquid sample flowed freely down and crossed the upper and lower marks of the middle bulb of the suction tube.

2.4. Kinematic viscosity

The kinematic viscosity was calculated using equation 2

$$v = kt \quad (\text{cm}^2/\text{s}). \quad (2)$$

where v is the kinematic viscosity (cm^2/s), k is the viscometer constant and t is the flow time (s).

2.5. Dynamic Viscosity Binary Mixture

The dynamic viscosity η (cP) is the converted viscosity, which is calculated using equation 3

$$\eta = v\rho \quad (\text{cP}) \quad (3)$$

where η is the dynamic viscosity (cP), v is the kinematic viscosity and ρ is the density of the liquid.

2.6. Determination of excess molar volume (V_m^E) of binary mixture

The excess molar volume of the binary mixture was determined using equation (4):

$$V_M^E = X_1 M_1 \left(\frac{1}{\rho_m} - \frac{1}{\rho_1} \right) + X_2 M_2 \left(\frac{1}{\rho_m} - \frac{1}{\rho_2} \right) \quad (4)$$

where X_1 and X_2 are the mole fraction of pure constituents NB and DMSO respectively, M_1 and M_2 are the molar masses of pure constituents NB and DMSO



respectively (g/mole), ρ_1 and ρ_2 is the densities of pure constituents NB and DMSO respectively (g/cm^3) while ρ_m is the density of the binary mixture (NB-DMSO) (g/cm^3)

2.7. Determination of Excess Viscosity

This was done using equation 5,

$$\eta^E = n_m - (X_1 n_1 + X_2 n_2) \quad (5)$$

where η^E is the excess viscosity (cP), η_m is the viscosity of the mixture (NB-DMSO), X_1 and X_2 are the Mole fraction of pure constituents of NB and DMSO respectively, η_1 and η_2 is the Viscosity of pure constituents NB and DMSO respectively.

2.8. Excess Gibbs free energy of activation of viscous flow (G^{*E})

This is determined using equation 6

$$G^{*E} = RT[\ln n_m V_m - (X_1 \ln n_1 V_1 + X_2 \ln n_2 V_2)] (\text{Jmol}^{-1}) \quad (6)$$

where, R = the universal gas constant ($\text{J.k}^{-1}.\text{mol}^{-1}$), T is the absolute temperature (k), V_1 and V_2 is the molar volume of pure constituents NB and DMSO (cm^3), V_m is the Molar volume of the mixture (NB-DMSO) (cm^3/mol), η_m is the Viscosity of the mixture (NB-DMSO) (cP), η_1 and η_2 is the Viscosity of pure constituents NB and DMSO respectively (cP).

2.9. Grunberg-Nisan interaction parameter

This was done using equation 7

$$d = \ln n_m - \frac{(X_1 \ln n_1 + X_2 \ln n_2)}{X_1 X_2} \quad (7)$$

where η_1 and η_2 = Viscosity of pure constituents NB and DMSO respectively, X_1 and X_2 = Mole fraction of pure constituents NB and DMSO respectively, η_m = Viscosity of the mixture (NB-DMSO).

2.10. Conductivity

The electrical conductivity meter was used to determine the conductivity by inserting the conductivity electrode into the solution that attained the required temperature, the mixture was stirred and the reading was taken.

3.0. Results and Discussion

Figs. 1 to 6, show plots for the variation of density, viscosity, molar excess volume, excess

viscosity, excess Gibbs free energy of activation for nitrobenzene-dimethyl sulphoxide binary mixture studied at different temperatures versus their mole fractions and the Gruenberg-Nissan interaction parameters of nitrobenzene-dimethyl sulphoxide system.

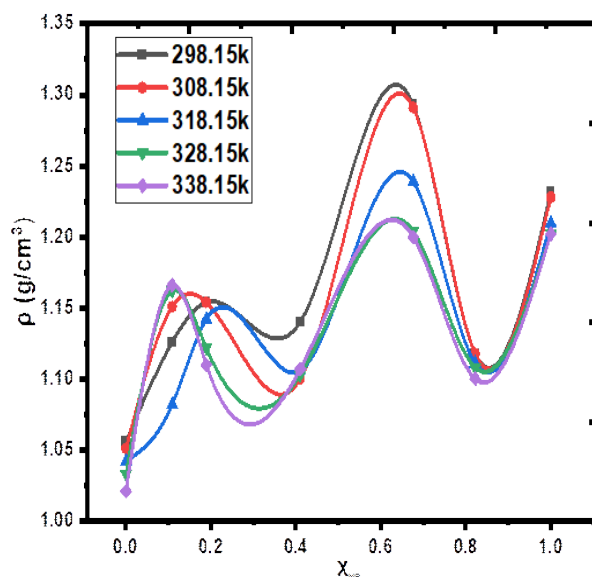


Fig.1: Variation of density with mole fraction for NB-DMSO at different temperatures

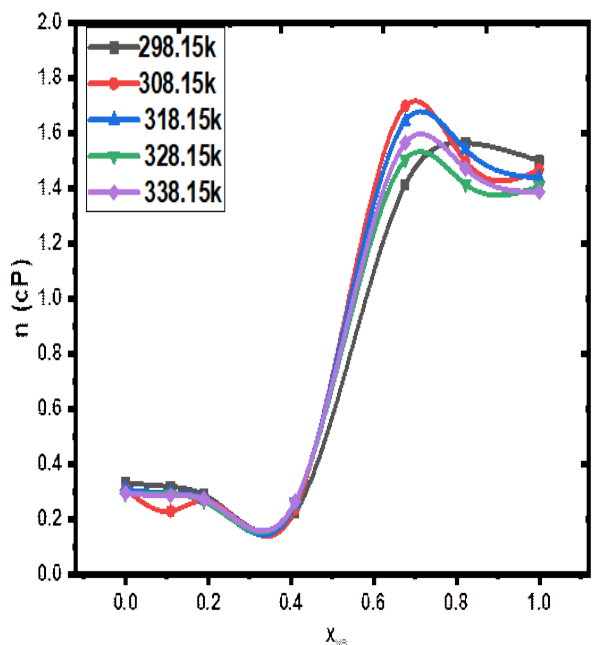


Fig.2: Variation of viscosity with mole fraction for NB-DMSO at different temperatures



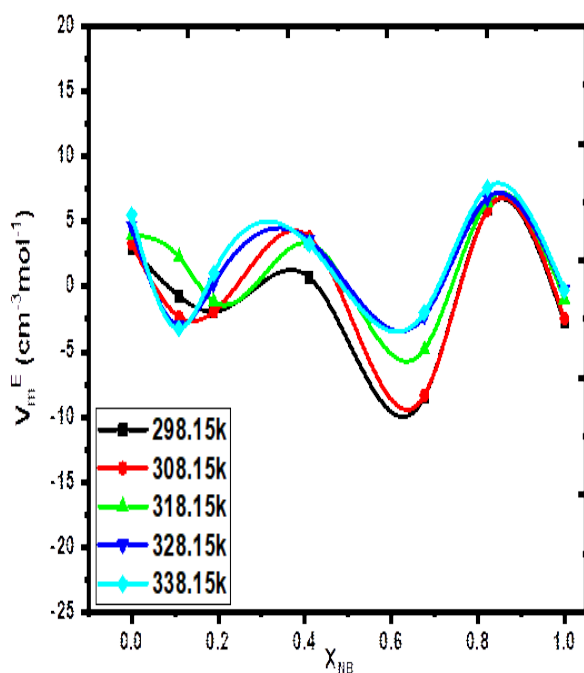


Fig.3: Variation of molar excess volume with mole fraction for NB-DMSO at different temperatures

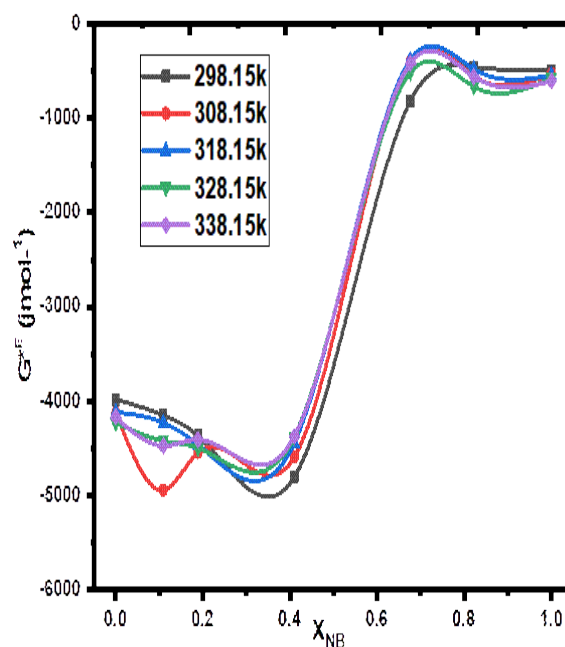


Fig.5: Variation of excess Gibbs free energy with mole fraction for NB-DMSO at different temperatures

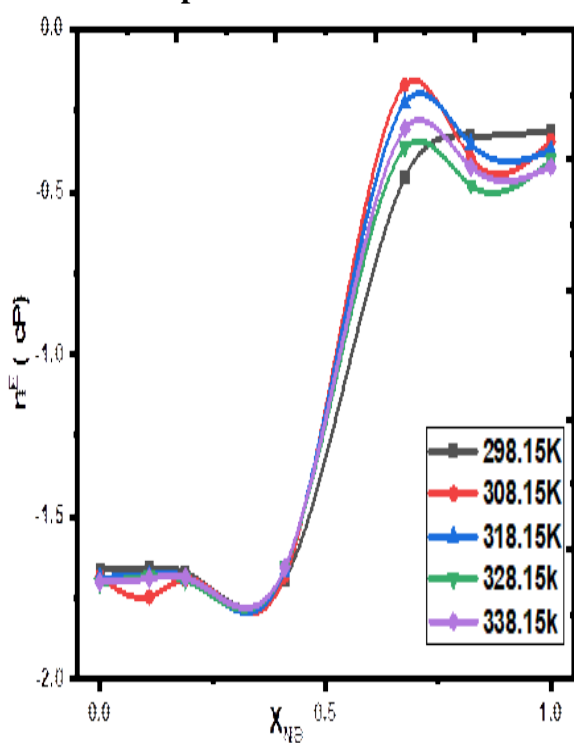


Fig.4: Variation of excess viscosity with mole fraction for NB-DMSO at different temperatures

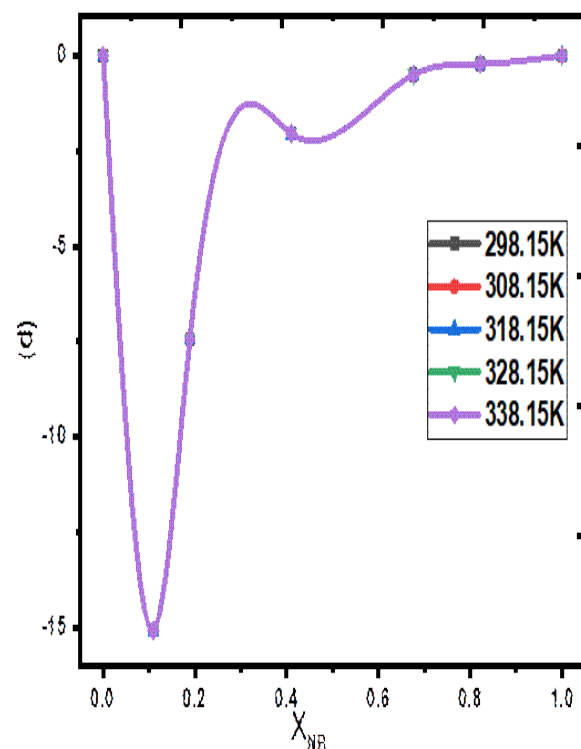


Fig.6: Variation for Gruenberg-Nissan Interaction parameters of NB-DMSO at different temperature



The experimental results of the density and viscosity of the binary mixtures of NB-DMSO studied under the different temperatures including 298.15, 308.15, 318.15, 328.15 and 338.15 K, respectively were listed and plotted in figure 1&2, as a function of the mole fractions. The results obtained from the density were used to calculate the excess molar volume (V_m^E). It was observed that there was a steady increase in density from 0 to 25% NB (1.056, 1.126, and 1.154 g/cm) which dropped at 50/50 % NB-DMSO (1.140g/cm), and started increasing again from 75 to 100% NB (1.293, 1.118, 1.232 g/cm). This trend is seen as both the mole fraction and temperature increase at NB-DMSO mixture. It was observed that 75 % by volume of NB has the highest density at all temperatures {298.15K (1.293g/cm); 308.15 k (1.291g/cm); 318.15 k (1.239g/cm); 328.15 K (1.204g/cm) and 338.15 K (1.200g/cm)} and 50% volume mixture has the lowest density across all temperature {298.15 K (1.140); 308.15 K (1.100); 318.15 K (1.106); 328.15(1.103); and 338.15(1.107)}. The experimental results of the viscosity value were used to calculate the excess viscosity (η^E), excess Gibbs free energy of activation of viscous flow (G^{*E}) and the Grunberg-Nissan interaction parameter (d). The viscosity of the binary mixture of NB-DMSO decreased linearly from 0 to 50% NB (0.332, 0.319, 0.291, 0.226cP) and has a rapid increase at 75 and 85%NB (1.415 and 1.567 cP), and drops at 100% NB (1.501cP) at the temperature of 298.15 K. At the temperature of 308.15 K the viscosity decreased from 0 to 50% NB (0.310, 0.230, 0.269, 0.239 cP) and increased at 75% (1.700) and started decreasing again from 85% down to 100% NB (1.499 to 1.469cP). At 318.15 K, it decreased from 0 to 50% (0.309, 0.296, 0.279, 0.257 cP), then rapidly increased to 1.646 cP at 75% NB and decreased gently down to 100% NB (1.542cP at 85% and 1.440 cP at 100%). At 328.15 K and from 0 to 50% the viscosity decreases (0.291, 0.292, 0.267 and 0.264cP) while at 75% NB, the viscosity

increased to 1.506 cP, and started again to drop from 1.416 cP at 85% to 1.409 cP at 100%. The viscosity at 338.15 K decreased from 0 to 50 % (0.296, 0.286, 0.275, and 0.265 cP), then at 75% (1.566cP) it increased and drops at 85% (1.472) and 100% (1.389 cP). The viscosity results across all the temperatures studied indicate that 75/25% NB-DMSO mixture has the highest viscosity {298.15 K (1.415cP); 308.15 K (1.700 cP); 318.15 (1.646cP); 328.15 (1.506cP); 338.15K (1.200)} while the 50/50 % by volume has the lowest viscosity {298.15 (0.226 cP); 308.15 K (0.239 cP); 318.15 K (0.257cP); 328.15 K (0.264 cP) and 338.15 K (0.265 cP)}.

3.2.2: Excess parameters

For an ideal mixing, the excess parameters should be zero (Sujata and Sunil, 2013). Excess parameters with a negative or positive value indicate non-ideal behavior in combinations most of the time. From the literature, the excess property is explained as the variation between that of the property of solution thermodynamics and that of an ideal solution under parallel conditions of thermodynamics and that for an ideal solution under parallel conditions of temperature and composition (Ali and Nain, 2002). The variation represents the positive or the negative excess of solution thermodynamics, relative to the ideal solution as shown by Mesquite et al (2014). The non-zero values in excess parameters show evidence of distinct interactions between the components of the mixtures. Researchers have shown the nature and intensity of this interaction basis on the sign and magnitude of excess parameters (Sati *et al.*, 1991).

The results of NB-DMSO system for the excess molar volume (fig.3) shows positive values (0%, 2.985; 50%, 0.724; 85%NB, 5.888) and negative values (15%, -0.713; 25%, -1.918; 75%, -8.473, 100%NB,-2.750) at 298.15k. At the temperature of 308.15k, it shows then positive values at (0%, 3.337; 50%, 3.804; 85%, 5.888) and negative values at (15%, -2.314; 25%, -1.918; 75%, -8.343; 50%,



-2.425). At the temperature of 308.15k, it is positive at (0%, 3.979; 15%, 2.286; 50%, 3.328; 85%, 6.548), and negative at (25%, -1.129; 75%, -4.814; 100%, 0.933). At 318.15k, the excess molar volume is equally positive at (0%, 4.633; 25%, 0.223; 50%, 3.565; 85%, 6.833) while it is negative at (15%, -2.997; 75%, -2.268; 100%, -0.256). Finally at 328.15k, the values are positive at (50%, 5.522; 25%, 1.058; 50%, 3.249; 85%, 7.601) and negative (15%, -3.242; 75%, -1.967 and 100%, -0.256). All these are as the mole fraction and temperature increase. The positive values are attributed to the dispersive forces, while the negative values are attributed to dipole-dipole interactions and the creation of hydrogen bonds (Sujata and Sunil, 2013). The negative values of excess volume (V_m^E) result from strong interactions between unlike molecules of NB-DMSO, while the positive excess volume is due to strong interactions between like molecules of the pure solvents and the mixture. The positive values of excess volume are a result of the rupture of H-bonds and the loosening of dipole interactions (El-Banna, 1997). The positive and negative values of the excess molar volume vary with the percentage volume as the temperature increases (Tables 1-5). The negative value shown on the molar excess volume (V_m^E) is because intermolecular interactions between unlike molecules predominate over intermolecular interactions between like molecules. The dipolar dispersion forces between the NB-DMSO molecules are primarily responsible for the negative V_m^E . (Prakash and Dlip, 2014)

From the results presented in figure 4, excess viscosity shows negative values all through, both as the temperature and mole fractions increase. The negative values of η^E suggest the existence of dispersion forces; consequently, the negative deviation of η^E indicates that dipole interactions are more likely in NB-DMSO system (Fort and Moore, 1966).

The excess Gibbs free energy of activation of viscous flow and Grunberg-Nissan interaction

parameters were equally presented in Fig. 5 and 6. The G^{*E} results show negative all through the experiment as both the temperature and mole fraction increase. The negative results of excess Gibbs free energy indicate weak associations of the mixture which translates to a less viscous mixture. Positive deviation in values of excess activation energy of flow indicates definite interactions between the constituents (Sathyanarayana, 2007). The Gibbs free energy of activation of flow is negative across the constitution (NB-DMSO) which indicates the supremacy of dispersion forces.

The sign and magnitude of Grunberg-Nissan interaction parameters are considered the measure of interaction strength. The result in fig. 6, shows negative values all through, in all the temperatures studied both as the mole fraction increases. The values drop negatively down the line as the volume of NB increases and increased linearly as the volume of DMSO decreases back to zero. The negative values of the d-parameter indicate the existence of weak interactions between the unlike molecules NB-DMSO. The positive values of the Grunberg-Nissan interaction parameters indicate that interactions between unlike molecules are strong (Sathyanarayana, 2007)

4.0 Conclusions

Looking at the NB-DMSO excess parameters that were studied, the binary mixture has shown its importance and its possibility in battery cells and other industrial applications based on the nature of their intermolecular interactions. The density and viscosity of both the pure and mixed solvents were measured at different temperatures (298.15, 308.15, 318.15, 328.15 and 338.15k) over the composition range of 0 - 100%. The outcome showed the thermodynamic relationship accorded to heat and fluid flow. The density and viscosity of the entire system increase as the temperature and volume percent increase but have a little drop in density and viscosity at increased volume percent as the temperature increases.



From the results the excess molar volume indicates negative flows which suggests the existence of dispersive forces, and that the dipole interactions are more likely. The excess Gibbs free energy shows negative values which indicate weak molecular interactions between the solvent NB-DMSO. The Grunberg-Nissan interaction parameters which show negative values indicate the existence of weak interaction between the unlike molecules.

From the findings of these excess parameters studied it has shown that the binary mixture of these organic solvents has perfect properties to support as a cell electrolyte with its inherent advantage for green technology, minimal health risk and low cost for commercial purposes when compared to its chemical counterparts.

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Consent for publication

Not Applicable

Availability of data and materials

The publisher has the right to make the data public

Competing interests

The authors declared no conflict of interest. This work was carried out in collaboration among all authors.

Funding

There is no source of external funding

Authors' contributions:

Ibezim-Ezeani, Millicent Uzoamaka and Horsfall, Micheal, design and supervised the research, Orjiocha, Samuel Ibezim, Abugu, Hillary Onyeka, Ogbodo, John Onyebuchi, and Akor Joseph, analyzed the data. Nwanya Assumpta Chinwe, Ibezim-Ezeani, Millicent Uzoamaka edited and reviewed the manuscript and Orjiocha S.I interpreted the data and wrote the manuscript. All authors read and approve the final manuscript.

