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# Ionic-Interaction of Potassium Iodide in Edible Oils + DMF System by Viscosity Method

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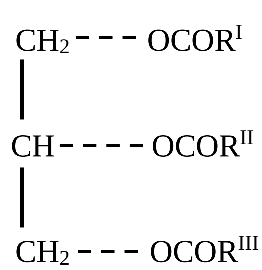
**Abstract**

Ionic-Interactions of Potassium Iodide (KI) in binary solvent systems of edible oil (maize oil and sunflower oil) + N, N-dimethylformamide (DMF) were studied at different temperatures by viscosity method. The viscosities of KI in sunflower oil + DMF and maize oil + DMF were found to increase with the increase in the concentration of edible oil and also with an increase in the concentration of KI from $1.0 \times 10^{-2}$ to $9.0 \times 10^{-2}$ mol.dm$^{-3}$. Ion-ion interaction and ion-solvent interaction were evaluated in terms of *A* and *B* coefficients of the Jones-Dole equation and its applicability checked by the Moulik equation. Ion-solvent interaction was found positive in the edible oil + DMF solvent system. The parameter d*B*/dT was used as a criterion for the evaluation of structure-making or the structure-breaking ability of salt (KI) in the edible oil + DMF solvent system. It was concluded that KI behaves as a structure-breaker in edible oils + DMF solvent system.

**Keywords:** viscosity, edible oils, N, N-dimethylformamide, ion-solvent interaction

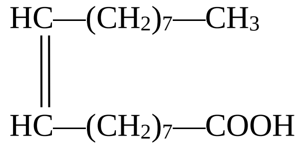
## 1. Introduction

The edible oils are mainly used in cooking but also used as raw material for a wide range of products such as lubricants, soaps, fuels, cosmetics, printing ink, and skincare products (Nik et al., 2005). The important applications of maize oil include margarine, soap, salve, paint, rustproofing for metal surfaces, inks, textiles, nitroglycerine, and insecticides. It is sometimes used as a carrier for drug molecules in pharmaceutical preparations. Maize oil is also a source of biodiesel. Vegetable oils are composed mainly of triglycerides which are glycerol molecules having three long chains of fatty acids attached by ester linkages at the hydroxyl group and are represented by the general structure shown below (Igwe, 2004; Ceriani et al., 2008).

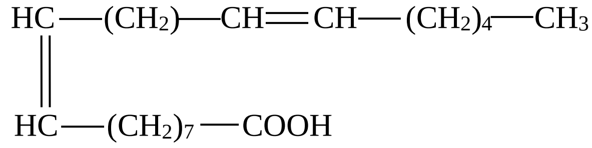



**Triglyceride**

where $R^{I}$, $R^{II}$, and $R^{III}$ are the hydrocarbon chains of fatty acids. The physical and chemical properties of edible oils are largely dependent upon their triglyceride composition. The type and point of attachment of fatty acids on the glycerine molecule affect the triglyceride structure of edible oil. The oxidation occurs at unsaturation sites on the molecule. On the contrary, triglyceride crystallization occurs at a high percentage of saturated fatty acids (Quinchia et al., 2010). The oleic and linoleic acids are two very important unsaturated fatty acids present in the oil.




**Oleic Acid**




**Linoleic Acid**

N, N-dimethylformamide (DMF) is a dipolar aprotic solvent and dissolves most of the edible oils. It is soluble in water and many other organic solvents (Wang et al., 2009). DMF-water systems were used to extract polycyclic aromatic hydrocarbons (PAHs) which are carcinogenic compounds and produced due to incomplete combustion of organic compounds and geochemical processes (Moret & Conte, 2000).

The nature of a solute in a binary solvent system has a fundamental importance in the study of chemical and physical properties of solution regarding their ionic interactions between solute-solute and solute-solvent (Al-Azzawl et al., 1990). In recent years, considerable attention has been given to the behavior of salts in various binary mixtures of solvents (Roy et al., 2010; Mishra et al., 2010; Zamir et al., 2005; Lu et al., 1999; Lu et al., 1995).

Among the various techniques, viscometry is one of the most reliable methods for determining the nature of the components by their physicochemical interactions like ionic interactions, covalent bonding, dipole-dipole interactions, charge-transfer phenomenon, hydrogen bonding, and hydrophobic interactions (Khan et al., 2007). The viscosity of salt solutions is needed for the design of numerous industrial processes and, at the same time, provides a useful approach to the solution structure regarding ionic and molecular interactions in different systems (Habibullah et al., 2010). The viscosity of edible oils is one of the important physical properties related to its processing and quality control and is affected by several factors including density, molecular weight, melting point, degree of unsaturation, and temperature (Biswas et al., 2007).

The main aim of this study is to investigate the interaction of salt (KI) with edible oil as both of these serve as nutrients for humans and the nature of their interaction is of considerable interest in determining various changes in their properties and their benefits and hazards for mankind. Therefore present study reveals the characteristic changes related to interactions of salt (KI) with edible oil + DMF solvent system at different temperatures by viscosity method. The ion-ion and ion-solvent interactions have been evaluated by Jones-Dole and Moulik equations. The data obtained by both equations are compared in terms of ion-ion and ion-solvent interactions. The effect of temperature, solvent (DMF), the concentration of salt in edible oils on the properties of the system was further evaluated and the results are explained in terms of structural and electrostatic interactions.

## 2. Experimental

### *2.1 Materials and Methods*

All the glassware used was of Pyrex A-grade quality. Potassium Iodide (KI) of RDH having a percent purity of 99.99% was used for the preparation of different concentrations of salt solutions. N, N-dimethylformamide (DMF) of A. C. S. HPLC grade (Lab-Scan) having a percent purity of 99.98% and density 0.9490 g/cm$^3$ was used as the solvent. Edible oils (sunflower and maize oil) were extracted from their dried, dehulled, and powdered seeds brought from the local market in Karachi, Pakistan. A stock solution of 5.0% v/v for sunflower oil as well as for maize oil was prepared in DMF. Potassium Iodide (KI) solutions with concentration ranging from 1.0 x 10$^{-2}$ to 9.0 x 10$^{-2}$ ± 3.7 x 10$^{-4}$ mol.dm$^{-3}$ were prepared in 1.0, 2.0, 3.0, 4.0 and 5.0% v/v sunflower oil + DMF and maize oil + DMF solvents respectively.

The viscosities of edible oil solutions were measured by using the Ostwald viscometer having Techniconominal constant 0.1 Cs/s capillary ASTM D 445. Three observations were taken to ensure the reproducibility of the measurements. The viscometer was placed inside a glass tube connected with a thermostatic water bath (Circulator, Model YCM-01, Taiwan R. O. C.) having a constant circulation of water throughout the experiment, to maintain the experimental temperatures from 303.15 to 323.15 K with a difference of 5 K respectively. A known volume of solvent and solution, respectively, was placed in the viscometer to attain the required temperature. The time flow between the two marks of the viscometer was recorded with the help of a stopwatch. Densities of solvent and solutions were measured with the help of a relative density bottle having a capacity of 10.0 $cm^3$ at different temperatures. An electronic balance (Model BL-150S, Sartorius, Germany) was used for mass determination. The uncertainty in the experimental data for density and viscosity was found to be in the range of ±1.0%.

*2.2 Statistical Analysis*

Statistical analysis for One Way ANOVA was done by using Minitab software. The reproducibility of the results was checked by taking three replicates of each measurement having probability $p \leq 0.05$.

## 3. Results and Discussion

Viscosities of potassium iodide (KI) having concentration 1.0 to 9.0 x $10^{-2}$ $mol.dm^{-3}$ in edible oil + DMF solvents were studied at different temperatures (303.15 to 323.15 K). Results tabulated in Tables 1 and 2 show that viscosities have increased with the increase in percent composition of edible oils (sunflower and maize oil) and also with the increase in the concentration of salt (KI) in the solution but decreased with the increase in temperature. The results show that the viscosities of KI + edible oils + DMF system decreased with the increase in temperature due to the weakening of the attractive forces between solute and solvent. At higher temperatures, the complex molecular structure breaks down into smaller fragments which can flow more easily and the viscosities of the solutions have decreased. The ability of edible oil molecules to form thermally stable molecular clusters decreased with the increase of temperature. A representative plot showing the effect of concentration of oil and temperature on viscosities of KI in the edible oil + DMF system is shown in Figure 1.

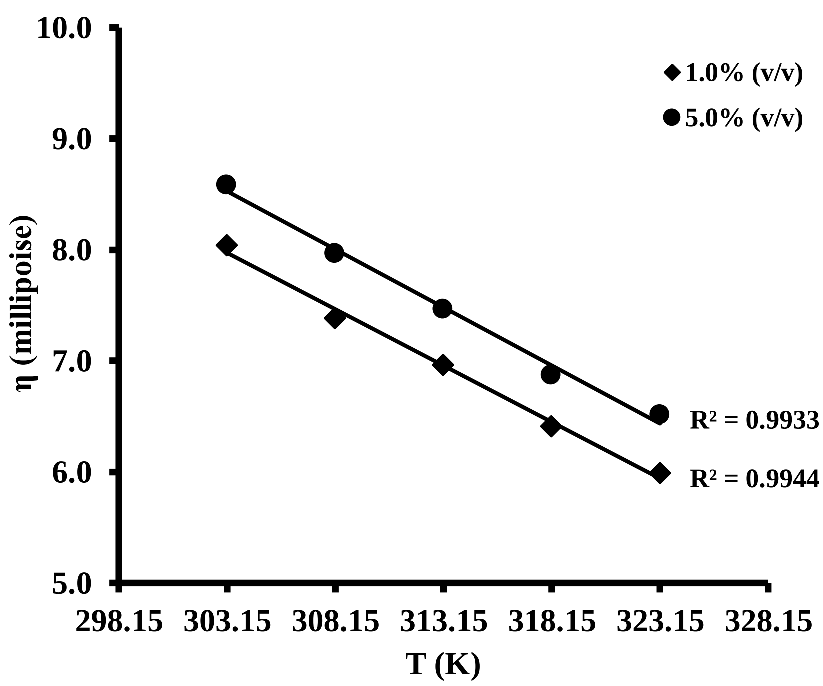



Figure 1. Plot of viscosity (η) of 5 x $10^{-2}$ $mol.dm^{-3}$ KI versus temperature in different percent compositions of maize oil

The results tabulated in Tables 1 and 2 show that the viscosities of KI solution increased with an increase in the concentration of KI in the edible oils + DMF system and also with an increase in percent composition of edible oils. The increase in the concentration of KI (salt) causes an increase in forces of attraction between the molecules which tend to stick together showing an increase in the viscosities of solutions and also with the increase in percent composition of edible oil in the solvent (DMF) because the edible oil is more viscous than DMF and therefore, an increase in the percent composition of oil in the solvent slightly increases the viscosities of the solutions. This behavior may also be due to the structural orientation of edible oils in DMF solvent systems which show ionic interaction with KI. The higher values of viscosities for sunflower oil were observed as compared to maize oil may be due to the difference between saturated and unsaturated components in sunflower and maize oils. Maize and sunflower oil are a mixture of both saturated and unsaturated ingredients. In maize oil, the saturated fatty acids are palmitic acid 8-10% and stearic acid 2-4% whereas unsaturated fatty acids are present as oleic acid 30-50% & linoleic acid 34-56%.

It also contains free acid as oleic acid 1-4% and traces of other acids are also present. Sunflower oil consists mostly of glycerides of oleic acid (25-42%), linoleic acid (52-66%), and saturated acid (5-13%). The change in values of viscosity in the presence and absence of salt is due to the unsaturation of ingredients present in the oil.

**Table 1. Viscosities of KI solutions in maize oil + DMF system at different temperatures**

| [KI] x $10^2$ (mol.dm$^{-3}$) | Viscosities (millipoise) at different temperatures (K) | | | | |
|---|---|---|---|---|---|
| | **303.15** | **308.15** | **313.15** | **318.15** | **323.15** |
| **1.0% maize oil + DMF system** | | | | | |
| **1.0** | 7.769 | 7.131 | 6.721 | 6.189 | 5.758 |
| **3.0** | 7.875 | 7.250 | 6.767 | 6.292 | 5.837 |
| **5.0** | 8.040 | 7.385 | 6.961 | 6.411 | 5.988 |
| **7.0** | 8.156 | 7.545 | 7.084 | 6.572 | 6.104 |
| **9.0** | 8.300 | 7.655 | 7.187 | 6.657 | 6.220 |
| **2.0% maize oil + DMF system** | | | | | |
| **1.0** | 7.873 | 7.263 | 6.783 | 6.269 | 5.782 |
| **3.0** | 7.969 | 7.359 | 6.862 | 6.389 | 5.918 |
| **5.0** | 8.108 | 7.500 | 7.012 | 6.503 | 6.017 |
| **7.0** | 8.242 | 7.612 | 7.129 | 6.645 | 6.158 |
| **9.0** | 8.394 | 7.801 | 7.360 | 6.830 | 6.303 |
| **3.0% maize oil + DMF system** | | | | | |
| **1.0** | 8.002 | 7.406 | 6.918 | 6.430 | 5.940 |
| **3.0** | 8.128 | 7.130 | 6.990 | 6.496 | 5.982 |
| **5.0** | 8.170 | 7.575 | 7.069 | 6.546 | 6.052 |
| **7.0** | 8.281 | 7.724 | 7.137 | 6.661 | 6.170 |
| **9.0** | 8.411 | 7.816 | 7.222 | 6.724 | 6.309 |
| **4.0% maize oil + DMF system** | | | | | |
| **1.0** | 8.264 | 7.500 | 7.066 | 6.556 | 6.098 |
| **3.0** | 8.364 | 7.640 | 7.115 | 6.635 | 6.186 |
| **5.0** | 8.465 | 7.770 | 6.724 | 6.697 | 6.275 |
| **7.0** | 8.618 | 7.787 | 7.316 | 6.777 | 6.320 |
| **9.0** | 8.657 | 7.973 | 7.374 | 6.854 | 6.436 |
| **5.0% maize oil + DMF system** | | | | | |
| **1.0** | 8.361 | 7.741 | 7.279 | 6.694 | 6.278 |
| **3.0** | 8.490 | 7.853 | 7.380 | 6.820 | 6.395 |
| **5.0** | 8.582 | 7.969 | 7.467 | 6.872 | 6.516 |
| **7.0** | 8.774 | 8.074 | 7.571 | 7.029 | 6.607 |
| **9.0** | 8.941 | 8.244 | 7.804 | 7.147 | 6.735 |

**Table 2. Viscosities of KI solutions in sunflower oil + DMF system at different temperatures**

| [KI] x $10^2$ (mol.dm$^{-3}$) | Viscosities (millipoise) at different temperatures (K) | | | | |
|---|---|---|---|---|---|
| | 303.15 | 308.15 | 313.15 | 318.15 | 323.15 |
| **1.0% sunflower oil + DMF system** | | | | | |
| 1.0 | 14.160 | 13.170 | 12.154 | 11.234 | 10.334 |
| 3.0 | 15.018 | 13.934 | 12.762 | 11.742 | 10.827 |
| 5.0 | 15.660 | 14.630 | 13.260 | 12.110 | 11.175 |
| 7.0 | 16.210 | 15.131 | 13.723 | 12.424 | 11.477 |
| 9.0 | 17.081 | 15.785 | 14.219 | 13.089 | 11.920 |
| **2.0% sunflower oil + DMF system** | | | | | |
| 1.0 | 14.268 | 13.204 | 12.390 | 11.460 | 10.626 |
| 3.0 | 15.110 | 14.020 | 13.160 | 12.138 | 11.230 |
| 5.0 | 15.690 | 14.640 | 13.673 | 12.617 | 11.691 |
| 7.0 | 16.216 | 15.246 | 14.271 | 13.176 | 12.147 |
| 9.0 | 17.917 | 15.883 | 15.080 | 13.841 | 12.800 |
| **3.0% sunflower oil + DMF system** | | | | | |
| 1.0 | 14.515 | 13.363 | 12.663 | 11.847 | 10.913 |
| 3.0 | 14.500 | 14.255 | 13.631 | 12.635 | 11.695 |
| 5.0 | 16.328 | 14.890 | 14.400 | 13.570 | 12.320 |
| 7.0 | 17.100 | 15.493 | 15.026 | 14.176 | 12.927 |
| 9.0 | 18.020 | 16.430 | 15.746 | 14.915 | 13.543 |
| **4.0% sunflower oil + DMF system** | | | | | |
| 1.0 | 14.960 | 13.630 | 12.972 | 12.115 | 11.314 |
| 3.0 | 16.124 | 14.674 | 14.072 | 13.200 | 12.312 |
| 5.0 | 17.183 | 15.518 | 14.925 | 14.044 | 13.120 |
| 7.0 | 18.034 | 16.300 | 15.740 | 14.810 | 13.860 |
| 9.0 | 19.135 | 16.987 | 16.624 | 15.557 | 14.580 |
| **5.0% sunflower oil + DMF system** | | | | | |
| 1.0 | 15.260 | 14.020 | 13.170 | 12.340 | 11.410 |
| 3.0 | 16.683 | 15.240 | 14.260 | 13.460 | 12.405 |
| 5.0 | 17.766 | 16.200 | 15.145 | 14.246 | 13.260 |
| 7.0 | 18.846 | 17.120 | 16.089 | 15.058 | 14.032 |
| 9.0 | 20.370 | 17.992 | 16.910 | 16.232 | 14.515 |

The ionic interaction of salt in solutions was evaluated in terms of Jones-Dole (Jones et al., 1929) coefficients expressed by the relation (Khan et al., 2005, 2006):

$$\eta_r = 1 + AC^{1/2} + BC \quad (1)$$

$$\frac{\eta_{sp}}{C^{1/2}} = A + BC^{1/2} \quad (2)$$

where $\eta_r$ is the relative viscosity of the solution which is derived by the $\eta/\eta_o$, $\eta$ is the viscosity of the solution, $\eta_o$ is the viscosity of the solvent, *C* is the concentration of salt, $\eta_{sp} = \eta_r - 1$ is the specific viscosity of the solution, *A* coefficient is the ion-ion interaction and coefficient *B* is ion-solvent interaction (Ibemesi, 1989; Ibesmi and Igwe, 1989). The coefficients *A* and *B* of Jones-Dole equation were obtained by plotting $\eta_{sp}/C^{1/2}$ versus $C^{1/2}$ as shown in Figure 2. *A* coefficient is related to the long-range interionic forces or ion-ion interaction while the *B* coefficient is related to the interaction between the ions and the solvent and is interpreted as a measure of the structure making and structure breaking tendency of an electrolyte in solution (Sundaramurthy et al., 2010).
The viscosity data were also analyzed by Moulik equation to evaluate ionic interactions:

$$\eta_r^2 = M + KC^2 \quad (3)$$

where *M* and *K* coefficients are constants (Roy et al., 2010). The applicability of the Moulik equation was obtained by plotting $\eta_r^2$ versus $C^2$ as shown in Figure 3.

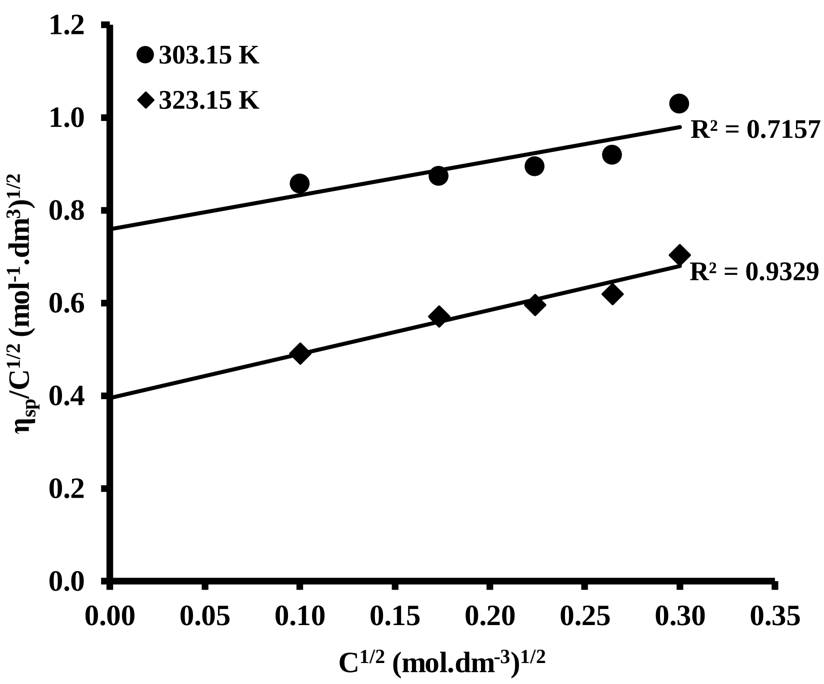



Figure 2. Plot of $\eta_{sp}/\sqrt{C}$ vs $\sqrt{C}$ for KI solutions in 1.0% sunflower oil at different temperatures

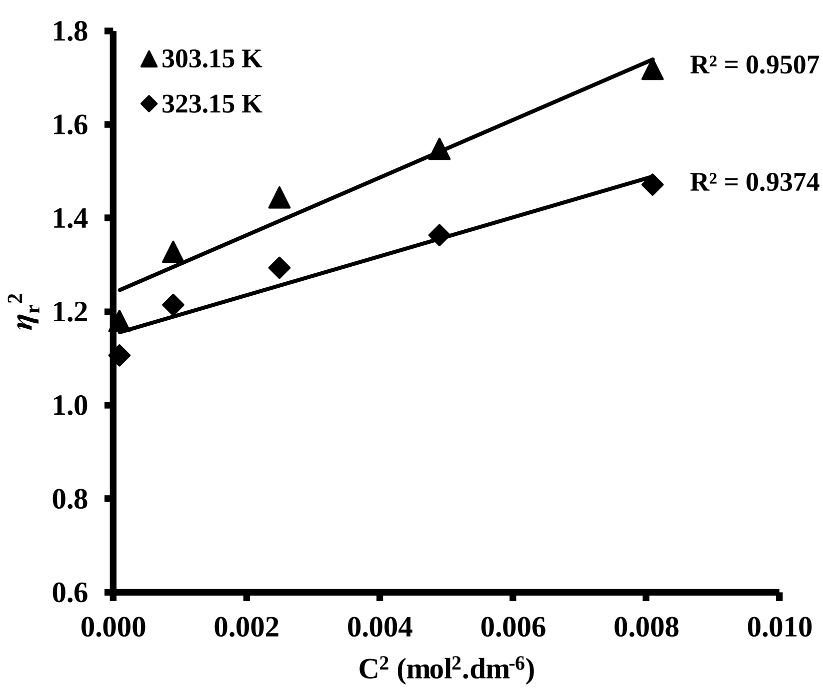



Figure 3. Plot of $\eta_r^2$ vs $C^2$ for KI solutions in 1.0% sunflower oil at different temperatures

The values of *A* and *B* coefficients of KI solutions in maize oil + DMF and sunflower oil + DMF are tabulated in Table 3. The values of *A* coefficient are positive at all temperatures and in all percent compositions of edible oil in the DMF solvent. This shows the presence of significant ion-ion interactions at low temperatures (303.15 K) and decreased at high temperature (323.15 K) with little variation in all percent compositions of solvent. *A* coefficient expresses the complete or incomplete dissociation and ion association of salt in edible oils + DMF solvent system. It is often ignored in dilute solutions where ion-ion interactions are not so prominent. The ion-solvent interaction in terms of the *B* coefficient of Jones-Dole equation can be explained based on Stokes and Mills equation:

$$\eta^E + \eta^A + \eta^D = \eta BC \tag{4}$$

where $\eta^E$ and $\eta^A$ are the viscosity increments due to the size and shape of ions and orientation of solvent molecules around the ions respectively, $\eta^D$ is the decrement as a result of the distortion of the solvent structure by ions.

**Table 3. *A* and *B* coefficients of KI solutions in different percent compositions of maize oil and sunflower oil + DMF system at different temperatures**

| **Oil (%v/v)** | ***A* (dm$^3$.mol$^{-1}$)$^{1/2}$ coefficients at different temperatures (K)** | | | | |
|---|---|---|---|---|---|
| | **303.15** | **308.15** | **313.15** | **318.15** | **323.15** |
| **Maize oil + DMF system** | | | | | |
| **1.0** | 0.0655 | 0.1245 | 0.1327 | 0.1766 | 0.1868 |
| **2.0** | 0.0726 | 0.0858 | 0.1125 | 0.1229 | 0.1282 |
| **3.0** | 0.0635 | 0.0424 | 0.0298 | 0.0413 | 0.0503 |
| **4.0** | 0.0717 | 0.0696 | 0.0601 | 0.0519 | 0.0505 |
| **5.0** | 0.1057 | 0.0908 | 0.1299 | 0.1303 | 0.1618 |
| **Sunflower oil + DMF system** | | | | | |
| **1.0** | 0.7585 | 0.7372 | 0.5573 | 0.4767 | 0.3994 |
| **2.0** | 0.7097 | 0.6665 | 0.6402 | 0.5770 | 0.5333 |
| **3.0** | 0.7743 | 0.6700 | 0.7924 | 0.8659 | 0.7485 |
| **4.0** | 0.9817 | 0.8205 | 0.9926 | 1.0281 | 1.0421 |
| **5.0** | 1.1263 | 1.0162 | 0.9998 | 1.0876 | 1.0594 |
| | ***B* (dm$^3$.mol$^{-1}$) coefficients at different temperatures (K)** | | | | |
| **Maize oil + DMF system** | | | | | |
| **1.0** | 0.6452 | 0.6512 | 0.6621 | 0.6776 | 0.6909 |
| **2.0** | 0.6492 | 0.6639 | 0.6824 | 0.7034 | 0.7279 |
| **3.0** | 0.6692 | 0.6730 | 0.6884 | 0.7097 | 0.7300 |
| **4.0** | 0.6773 | 0.6801 | 0.6906 | 0.7108 | 0.7414 |
| **5.0** | 0.6825 | 0.6932 | 0.7104 | 0.7138 | 0.7459 |
| **Sunflower oil + DMF system** | | | | | |
| **1.0** | 0.7166 | 0.7792 | 0.8121 | 0.8325 | 0.8618 |
| **2.0** | 0.8545 | 1.0110 | 1.1473 | 1.2137 | 1.2522 |
| **3.0** | 1.2492 | 1.2510 | 1.3003 | 1.3276 | 1.3497 |
| **4.0** | 1.3017 | 1.3215 | 1.3447 | 1.3629 | 1.3904 |
| **5.0** | 1.3459 | 1.3635 | 1.4029 | 1.4530 | 1.7713 |

Results tabulated for edible oils + DMF in Table 3 show a rapid increase in *B* coefficient with the increase in percent composition of edible oil in DMF solvent indicating lower structural order of edible oils gradually decreases with the addition of DMF. *B* coefficient would be positive when $\eta^E + \eta^A > \eta^D$. *B* coefficient of Jones-Dole relation is a measure of effective solvodynamic volume of solvated ions, the size and shape of solute as well as structural effects induced by solute-solvent interactions which make a major contribution to relative viscosity. When a solute dissolves in a solvent, a hole is made in the liquid with a rupture of intermolecular bonds, and the solute is inserted in that hole. Some of the solvent molecules get attached to the ions because of ion-solvent interactions and cause an increase in the viscosity of the solution showing a positive contribution to the viscosity *B* coefficient.

Generally positive *B* coefficient indicates structure making capacity for strongly hydrated solutes and structure breaking for weakly hydrated solutes. But in the case of large hydrophobic solutes, only *B* coefficient is not used to indicate structure making/breaking. An improvement of the *B* coefficient is to use its first derivative over temperature (dB/dT) because its sign is more indicative for the evaluation of structure making/breaking. The values of the *B* coefficient (ion-solvent interaction) in the edible oil + DMF system show an increasing trend with the increase in temperature showing the structure-breaking behavior of KI in edible oils + DMF solvent system. The values of dB/dT are positive showing that the KI behaves as a structure-breaker in edible oils + DMF. As DMF is a dipolar aprotic solvent and anion ($I^-$) was poorly solvated while cation ($K^+$) was strongly solvated in DMF (Gill et al., 1985). Potassium iodide also behaves as a structure breaker in dimethylacetamide-water mixtures (Singh et al., 1977).

The increase in values of *B* coefficient for KI in edible oil + DMF with the rise of temperature was observed due to large distortion ($\eta^D$) of regular solvent structure in presence of $K^+$ and $I^-$ ions. The values of ion-solvent interactions as a function of temperature are positive because $\eta^E$ is constant and $\eta^A$ will decrease slowly so $\eta^E + \eta^A > \eta^D$. The validity of the Moulik equation and the values of ion-solvent interaction also support the data evaluated from the Jones-Dole's relation.

## 4. Conclusions

Ionic interactions were studied by different relations that are Jones-Dole and Moulik equation for salt (KI) in edible oils + DMF solvent system. The nature of potassium iodide in terms of structure making or breaking was studied in edible oils + DMF solvent system. It was concluded that KI behaves as a structure-breaker in sunflower oil + DMF and maize oil +DMF solvent systems.

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