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# Densitometric and Viscometric Study of Diclofenac Sodium in Aqueous Solution in Presence and Absence of Additives at Different Temperatures.

Arun B. Nikumbh*[a], Ganesh K. Kulkarni[a] and Ravindra C. Bhujbal[b]

a = P. G. Department of Chemistry, S. S. G. M. College, Kopargaon-423601. (M. S.) India

b = Department of Chemistry, Hon. B. J. College, Ale, Pune-412411. (M. S.) India

University of Pune, Maharashtra State, India.

**Abstract:**

The measurement of density and viscosity of diclofenac sodium in aqueous solution have been measured and analyzed in presence of additives viz. NaCl, dextrose, KCl, sodium lactate in a range of molarity, m= ($4 \times 10^{-3}$ to $1.99 \times 10^{-2}$) $mol.L^{-1}$, at T = (298.15 to 313.15)K using a pycknometer and Ubbelohde viscometer. The density results were used to evaluate the apparent molar volumes. Viscosity results were used to calculate Jones-Dole viscosity B-coefficient. The simple physical properties like density and viscosity are used to explain the molecular interaction in aqueous solutions as well as in presence of additives. Drug therapy now forms a major aspect of therapeutics. The drug interact with various ions, molecules, biological membranes present in the biological system is an important phenomenon.

**Keywords:** Diclofenac sodium, density, viscosity, apparent molar volume, B-coefficient.

## 1. INTRODUCTION

In solution chemistry the interpretation of molecular interactions are understood with apparent molar volumes, Jones-Dole and Moulik parameters. It is based on theories like Van't Hoff, Gibbs, Debye-Huckel and Onsagar. The simple physical properties like density and viscosity are used to evaluate the molecular interaction in aqueous solutions. Drugs which alter the pain sensitivity or removes pain from body parts are called pain killers or analgesics. Which meet the health needs of majority of the population? They must be effective, safe, and cost effective and meet the medical need of local people.

The drugs in any solid dosage form or suspension when administered will first change into drug solution in body fluids. So, dissolution rate is important factor affecting the rate of absorption. When a drug is more rapidly or completely absorbed from solution, it is very likely that its absorption will be dissolution limited. Viscosity limits the dissolution rate and thereby affects the rapid absorption. Aqueous solution of Na-Salicylate showed its rapid appearance in plasma while the same drug in suspension form failed to reach the target as quickly as with aqueous solution[1].

The parameters like apparent molar volume, density, viscosity and Jone-Dole parameters are useful to focus the solute-solvent interactions and to understand different biochemical reactions at 310.15 K i.e. at body temperature. It also enables to enrich the data at various composition and temperatures.

## 1.1 EXPERIMENTAL:

### 1.1.1 Materials:

Diclofenac sodium of high purity was obtained from Research lab fine chemicals and was used as received. Deionized water with a specific conductance of $< 10^{-6}$ $S.cm^{-1}$ was used for the preparation of solutions at room temperature in a molarity range ($5.5 \times 10^{-3}$ to $1.99 \times 10^{-2}$) $mol.L^{-1}$. The precision of balance used was $\pm 1 \times 10^{-5}$g.

### 1.1.2 Density measurements:

The pycknometer was calibrated by measuring the densities of triple distilled water. The densities of distilled organic liquids like acetone, alcohol, benzene, carbon tetra chloride, aniline, nitrobenzene were evaluated with respect to density of water as a standard. The density was measured with an uncertainty of $\pm$ 1.48 $\times 10^{-4} g.cm^{-3}$.

 

### 1.1.3 Viscosity measurements:

The solution viscosities are measured with an uncertainty of ± 1.48 $\times 10^{-4}$ m.Pa.s by using Ubbelohde viscometer. The temperature of thermostat is maintained to desired temperature, by using demerstat with an accuracy of ± 0.1 K. The flow time will be measured at the accuracy of ± 0.01 s.

The different compositions (0.0055 to 0.0199 M) of solutions of pain killers were prepared in NS, DNS, RL and D.W. The densities and viscosities were measured at T = (298.15, 303.15, 308.15, 310.15 & 313.15) K for nine different concentrations. The solvent compositions used are as under,

NS = (0.9g NaCl)/100 ml D.W.

DNS = (5% Dextrose + 0.9 g NaCl)/100 ml D.W.

RL = (0.320 g Sodium Lactate + 0.600 g NaCl + 0.040 g KCl

+ 0.035 g $CaCl_2.2H_2O$) / 100 ml D.W.

D.W. = Pure distilled water.

### 1.1.4 DATA EVALUATION

The apparent molar volume, $\phi_v$, was obtained from the density results using the following equation[2, 3, 4]

$$\phi_v = \frac{1000(\rho_0 - \rho)}{C\rho_0} + \frac{M_2}{\rho} \qquad (1)$$

Where $M_2$, C, $\rho$ and $\rho_0$ are the molar mass of the DS, concentration (mol.$L^{-1}$), and the densities of the solution and the solvent, respectively.

The apparent molar volumes ($\phi_v$) were plotted against the square root of concentration ($C^{½}$) in accordance with the Masson's equation[5]

$$\phi_v = \phi v^{o} + S_v.C^{½} \qquad (2)$$

Where $\phi v^{o}$ is the limiting apparent molar volume and $S_v$ a semi-empirical parameter which depends on the nature of solvent, solute and temperature. Its ($S_v$) value for large organic solutes is not of much significance[6].

The viscosity results for the aqueous solutions of drugs were plotted in accordance with Jones-Dole equation[7]

$$\frac{\eta_r - 1}{C^{½}} = A + BC^{½} \qquad (3)$$

Where $\eta_r = (\eta/\eta_o)$ and $\eta$, $\eta_o$ are viscosities of the solution and solvent respectively, C is the molar concentration. The linear plots for $(\eta_r - 1)/C^{½}$ versus $C^{½}$ were obtained for the DS. The B-coefficients were obtained from the linear plots using the least-square fitting method. The A- coefficient reflects solute-solute interaction[8] and the B-coefficient reflect the solute-solvent interactions. The interactions can also be evaluated from Moulik[9] and Root[10] equations,

$$\eta_r^2 = M + KC^2 \qquad (4)$$

$$(d - d_0)/C = A - B\,C^{½} \qquad (5)$$

## 1.2 RESULTS AND DISCUSSION

The values of the density, apparent molar volume and viscosity of DS in NS, DNS, RL and pure distilled water are listed in table-1. In all sets the densities and viscosities of solutions increases with increase in concentration of solution[11]. At higher temperatures the values of densities and viscosities are smaller. The apparent molar volumes ($\phi_v$) were calculated by using equation (1). The plots of $\phi_v$ with square root of concentration are linear for all the concentrations investigated. Figure-1 shows plots of apparent molar volume $\phi_v$ of DS against square

 

root of concentration over the temperature range 298.15 K to 313.15K for NS. Similar such plots were observed for DNS, RL and DW solvent systems.

The limiting apparent molar volumes ($\phi v^0$) have been calculated from the intercept of linear plots using equation (2). They are listed in table-2. The $\phi v^0$ values provide information regarding the solute-solvent interactions, drug hydrophobicity and hydration properties. Inspection of table-2, shows the positive values of $\Phi_v^0$ for all sets. The positive values of $\phi v^0$ suggests strong solute-solvent interactions[12, 13]. The negative values of $S_v$ obtained from the slope indicate weak solute-solute interactions.

The viscosities ($\eta$) of solutions are listed in table-1. The $\eta$ values increases with concentration and decreases with rise in temperature. This suggests the existence of molecular interactions occurring in the system. The viscosity data have been analyzed by using Jones –Dole equation (3). Where $\eta$ and $\eta_o$ are the viscosities of solute and solvent respectively. Figures-2 shows the variation of $(\eta_r-1)/C^{1/2}$ against square root of concentration over the temperature range 298.15 K to 313.15K for different concentrations for NS. Similar such plots were observed for DNS, RL and DW solvent systems. 'A' is constant independent of concentration and 'B' is Jones-Dole coefficient represents measure of

**Table 1: Density, apparent molar volume and viscosity of diclofenac sodium in NS, DNS, RL and pure distilled water at different temperatures.**

| SET 1 in NS | | | | | SET 2 in DNS | | | | |
|---|---|---|---|---|---|---|---|---|---|
| T /K | C/mole/lit | ρ/(gm/cc) | η/ mPa.s | $\phi_v$/cm$^3$mol$^{-1}$ | T /K | C/mole/lit | ρ/(gm/cc) | η/ mPa.s | $\phi_v$/cm$^3$mol$^{-1}$ |
| 298.15 | 0.008 | 0.99938 | 0.90717 | 24.959 | 298.15 | 0.008 | 1.00613 | 0.95606 | -823.43 |
| | 0.01 | 1.00026 | 0.91046 | -4.9086 | | 0.01 | 1.00892 | 0.97112 | -876.21 |
| | 0.012 | 1.00137 | 0.91401 | -44.21 | | 0.012 | 1.01179 | 0.98393 | -918.39 |
| | 0.014 | 1.00223 | 0.91703 | -54.393 | | 0.014 | 1.01463 | 1.00254 | -946.62 |
| | 0.016 | 1.00336 | 0.91965 | -79.108 | | 0.016 | 1.01757 | 1.02120 | -974.3 |
| | 0.018 | 1.00419 | 0.92209 | -81.599 | | 0.018 | 1.0207 | 1.04089 | -1006.7 |
| | 0.0199 | 1.0052 | 0.92395 | -94.783 | | 0.0199 | 1.02429 | 1.0545 | -1062.8 |
| 303.15 | 0.008 | 0.99792 | 0.81235 | 40.0947 | 303.15 | 0.008 | 1.00502 | 0.85476 | -853.49 |
| | 0.01 | 0.99879 | 0.81557 | 8.18097 | | 0.01 | 1.00785 | 0.86878 | -904.59 |
| | 0.012 | 0.99982 | 0.81767 | -26.629 | | 0.012 | 1.01056 | 0.88020 | -928.88 |
| | 0.014 | 1.00071 | 0.82138 | -41.498 | | 0.014 | 1.01362 | 0.89124 | -971.67 |
| | 0.016 | 1.00155 | 0.82365 | -49.566 | | 0.016 | 1.01635 | 0.90844 | -983.19 |
| | 0.018 | 1.0026 | 0.82526 | -67.684 | | 0.018 | 1.01942 | 0.92292 | -1011.4 |
| | 0.0199 | 1.00363 | 0.82699 | -83.234 | | 0.0199 | 1.02281 | 0.93748 | -1057.2 |
| 308.15 | 0.008 | 0.99606 | 0.73446 | 69.1545 | 308.15 | 0.008 | 1.00337 | 0.76936 | -852.37 |
| | 0.01 | 0.99685 | 0.73655 | 39.4769 | | 0.01 | 1.00605 | 0.78267 | -888.93 |
| | 0.012 | 0.99788 | 0.73897 | -0.5881 | | 0.012 | 1.00863 | 0.79379 | -905.16 |
| | 0.014 | 0.99881 | 0.74119 | -22.082 | | 0.014 | 1.0115 | 0.80844 | -937.91 |
| | 0.016 | 0.99966 | 0.74345 | -33.221 | | 0.016 | 1.01431 | 0.81592 | -958.9 |
| | 0.018 | 1.00079 | 0.74496 | -57.682 | | 0.018 | 1.01728 | 0.82981 | -984.41 |
| | 0.0199 | 1.00163 | 0.74713 | -64.553 | | 0.0199 | 1.02105 | 0.84107 | -1052.3 |
| 310.15 | 0.008 | 0.99532 | 0.70476 | 69.2055 | 310.15 | 0.008 | 1.00285 | 0.74073 | -880.76 |
| | 0.01 | 0.99611 | 0.70678 | 39.5056 | | 0.01 | 1.00545 | 0.75193 | -903.73 |
| | 0.012 | 0.99705 | 0.70895 | 6.98968 | | 0.012 | 1.00815 | 0.76243 | -927.73 |
| | 0.014 | 0.99805 | 0.71165 | -20.655 | | 0.014 | 1.01107 | 0.77587 | -961 |
| | 0.016 | 0.99902 | 0.71409 | -39.571 | | 0.016 | 1.01363 | 0.78425 | -963.42 |
| | 0.018 | 0.99978 | 0.71598 | -42.539 | | 0.018 | 1.01644 | 0.79715 | -979.53 |
| | 0.0199 | 1.00047 | 0.71742 | -43.223 | | 0.0199 | 1.02023 | 0.81059 | -1049 |
| 313.15 | 0.008 | 0.99411 | 0.66431 | 83.1746 | 313.15 | 0.008 | 1.00146 | 0.69792 | -845.12 |
| | 0.01 | 0.995 | 0.66662 | 40.5595 | | 0.01 | 1.0041 | 0.70599 | -879.46 |
| | 0.012 | 0.99581 | 0.6691 | 18.799 | | 0.012 | 1.00653 | 0.71570 | -884.93 |
| | 0.014 | 0.99674 | 0.67034 | -5.495 | | 0.014 | 1.00917 | 0.72921 | -904.24 |
| | 0.016 | 0.99777 | 0.6718 | -30.12 | | 0.016 | 1.01235 | 0.73546 | -953.1 |
| | 0.018 | 0.99859 | 0.6754 | -37.521 | | 0.018 | 1.01524 | 0.74826 | -974.99 |
| | 0.0199 | 0.99947 | 0.67845 | -48.369 | | 0.0199 | 1.01885 | 0.75951 | -1035.9 |

 

**Table 1:** Continued……

| SET 3 In RL | | | | | SET 4 In D.W. | | | | |
|---|---|---|---|---|---|---|---|---|---|
| T /K | C/mole/lit | ρ/(gm/cc) | η/ mPa.s | $\phi_v/cm^3mol^{-1}$ | T /K | C/mole/lit | ρ/(gm/cc) | η/ mPa.s | $\phi_v/cm^3mol^{-1}$ |
| 298.15 | 0.0055 | 1.00064 | 0.90862 | -338.56 | 298.15 | 0.006 | 0.9975 | 0.90112 | 242.033 |
| | 0.007 | 1.0016 | 0.91392 | -335.74 | | 0.008 | 0.99776 | 0.90307 | 228.577 |
| | 0.0085 | 1.00254 | 0.91906 | -331.66 | | 0.01 | 0.99809 | 0.90531 | 213.427 |
| | 0.01 | 1.00346 | 0.92146 | -326.87 | | 0.012 | 0.99847 | 0.90726 | 199.097 |
| | 0.0115 | 1.00439 | 0.92523 | -324.29 | | 0.014 | 0.99887 | 0.90943 | 187.388 |
| | 0.013 | 1.00529 | 0.92757 | -320.04 | | 0.016 | 0.9992 | 0.91214 | 182.984 |
| | 0.0145 | 1.00615 | 0.93314 | -313.96 | | 0.018 | 0.99958 | 0.91507 | 176.734 |
| 303.15 | 0.0055 | 0.99927 | 0.81332 | -333.53 | 303.15 | 0.006 | 0.99621 | 0.80708 | 233.973 |
| | 0.007 | 1.00022 | 0.81749 | -330.44 | | 0.008 | 0.99645 | 0.8089 | 225.109 |
| | 0.0085 | 1.00116 | 0.81924 | -327.37 | | 0.01 | 0.99689 | 0.81042 | 199.609 |
| | 0.01 | 1.00208 | 0.82423 | -323.29 | | 0.012 | 0.99715 | 0.81279 | 197.684 |
| | 0.0115 | 1.00299 | 0.82799 | -319.47 | | 0.014 | 0.99755 | 0.81421 | 186.198 |
| | 0.013 | 1.0039 | 0.83023 | -316.6 | | 0.016 | 0.99787 | 0.81621 | 182.598 |
| | 0.0145 | 1.00475 | 0.83406 | -310.21 | | 0.018 | 0.99826 | 0.8192 | 175.848 |
| 308.15 | 0.0055 | 0.9976 | 0.7361 | -326.75 | 308.15 | 0.006 | 0.99459 | 0.72885 | 232.677 |
| | 0.007 | 0.99855 | 0.73897 | -325.23 | | 0.008 | 0.99487 | 0.73052 | 219.174 |
| | 0.0085 | 0.99947 | 0.7419 | -320.79 | | 0.01 | 0.99527 | 0.73276 | 198.926 |
| | 0.01 | 1.00039 | 0.74429 | -317.76 | | 0.012 | 0.99554 | 0.73453 | 196.324 |
| | 0.0115 | 1.00128 | 0.74888 | -312.97 | | 0.014 | 0.99593 | 0.73648 | 185.78 |
| | 0.013 | 1.00218 | 0.75203 | -310.13 | | 0.016 | 0.99625 | 0.73866 | 182.265 |
| | 0.0145 | 1.00302 | 0.75612 | -303.75 | | 0.018 | 0.99665 | 0.73932 | 175.011 |
| 310.15 | 0.0055 | 0.99683 | 0.70716 | -321.5 | 310.15 | 0.006 | 0.99392 | 0.70027 | 221.082 |
| | 0.007 | 0.99776 | 0.70956 | -318.26 | | 0.008 | 0.99416 | 0.70224 | 215.552 |
| | 0.0085 | 0.99867 | 0.71223 | -313.9 | | 0.01 | 0.99457 | 0.70382 | 195.034 |
| | 0.01 | 0.99958 | 0.7152 | -310.93 | | 0.012 | 0.99487 | 0.7056 | 190.575 |
| | 0.0115 | 1.00049 | 0.71837 | -308.82 | | 0.014 | 0.99521 | 0.70686 | 184.474 |
| | 0.013 | 1.00136 | 0.72254 | -304.14 | | 0.016 | 0.99552 | 0.70789 | 181.768 |
| | 0.0145 | 1.0022 | 0.72519 | -298.4 | | 0.018 | 0.99592 | 0.70944 | 174.578 |
| 313.15 | 0.0055 | 0.99572 | 0.66576 | -320.02 | 313.15 | 0.006 | 0.99283 | 0.66076 | 219.644 |
| | 0.007 | 0.99665 | 0.66875 | -317.17 | | 0.008 | 0.99308 | 0.66215 | 213.265 |
| | 0.0085 | 0.99757 | 0.67161 | -314.25 | | 0.01 | 0.99351 | 0.66358 | 191.206 |
| | 0.01 | 0.99847 | 0.67219 | -310.27 | | 0.012 | 0.99378 | 0.66468 | 189.943 |
| | 0.0115 | 0.99936 | 0.67578 | -306.52 | | 0.014 | 0.99412 | 0.66579 | 183.955 |
| | 0.013 | 1.00026 | 0.67798 | -304.48 | | 0.016 | 0.99445 | 0.66669 | 180.069 |
| | 0.0145 | 1.00111 | 0.68379 | -299.43 | | 0.018 | 0.99483 | 0.66776 | 174.208 |

 

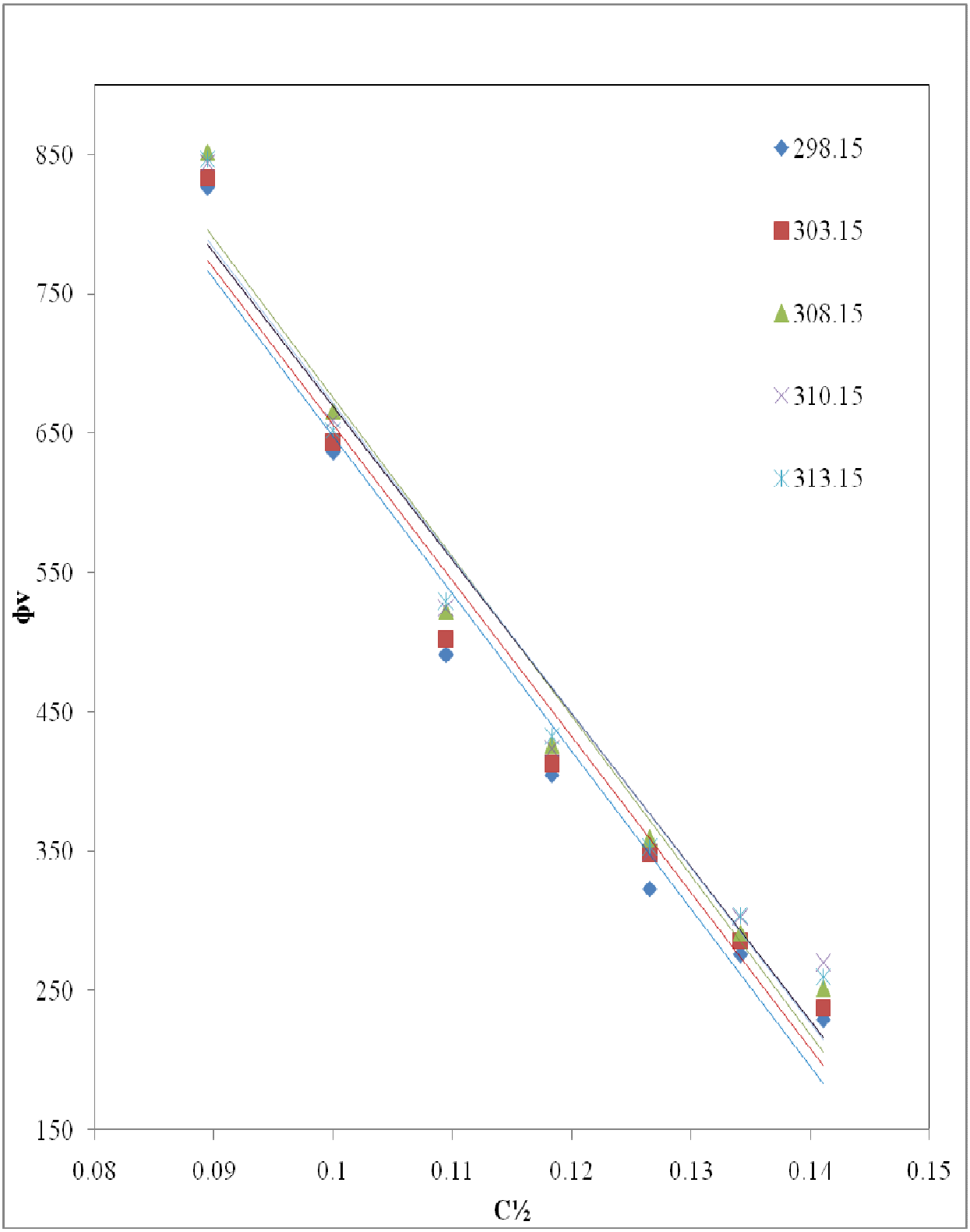


**Figure- 1:** Plot of $\phi_v$ Versus C½ of DS in NS at different temperatures.

 

**Table 2:** The interaction parameters of diclofenac sodium solutions at various temperatures.

| Sets | T /K | **Masson equation** | | **Roots equation** | | **Jone-Dole equation** | | **Moulik equation** | |
|---|---|---|---|---|---|---|---|---|---|
| | | $\Phi_v^0$ | $S_v$ | A | B | A | B | M | K |
| Set 1 | 298.15 | 1777 | -11301 | 11.31 | -1.462 | -0.277 | 2.801 | 0.990 | 109.8 |
| | 303.15 | 1774 | -11181 | 11.18 | -1.456 | -0.435 | 3.439 | 0.972 | 105.1 |
| | 308.15 | 1817 | -11418 | 11.39 | -1.496 | -0.307 | 3.791 | 0.984 | 101.6 |
| | 310.15 | 1770 | -11012 | 10.98 | -1.448 | -0.358 | 3.129 | 0.980 | 109.5 |
| | 313.15 | 1781 | -11097 | 11.05 | -1.456 | -0.315 | 3.038 | 0.985 | 121.1 |
| Set 2 | 298.15 | 6077 | -41834 | 32.79 | -5.897 | -2.625 | 19.54 | 0.816 | 560.0 |
| | 303.15 | 5922 | -40818 | 41.68 | -5.728 | -2.515 | 18.33 | 0.818 | 508.5 |
| | 308.15 | 5902 | -40537 | 41.32 | -5.697 | -2.468 | 17.89 | 0.823 | 481.6 |
| | 310.15 | 5819 | -39930 | 40.67 | -5.608 | -2.459 | 17.87 | 0.823 | 490.0 |
| | 313.15 | 5871 | -40256 | 40.95 | -5.654 | -2.372 | 17.16 | 0.827 | 469.6 |
| Set 3 | 298.15 | 1614 | -11594 | 14.18 | -1.448 | -0.173 | 3.25 | 1.006 | 280.2 |
| | 303.15 | 1646 | -11772 | 14.38 | -1.480 | -0.443 | 4.411 | 0.978 | 276.3 |
| | 308.15 | 1630 | -11633 | 14.19 | -1.459 | -0.733 | 5.794 | 0.950 | 290.3 |
| | 310.15 | 1613 | -11503 | 14.02 | -1.439 | -0.891 | 6.394 | 1.028 | 301.9 |
| | 313.15 | 1612 | -11506 | 14.01 | -1.437 | -0.730 | 5.521 | 1.023 | 286.2 |
| Set 4 | 298.15 | 333.8 | -1200 | -0.02 | 1.24 | 0.008 | 1.219 | 1.012 | 114.2 |
| | 303.15 | 313.3 | -1049 | -0.008 | 1.169 | 0.010 | 1.154 | 1.012 | 107.7 |
| | 308.15 | 306.6 | -1001 | -0.003 | 1.134 | 0.022 | 1.175 | 1.014 | 113.5 |
| | 310.15 | 285.0 | -836.5 | 0.027 | 0.89 | 0.037 | 0.918 | 1.014 | 97.5 |
| | 313.15 | 280.4 | -808.8 | 0.031 | 0.866 | 0.043 | 0.687 | 1.012 | 81.14 |

 

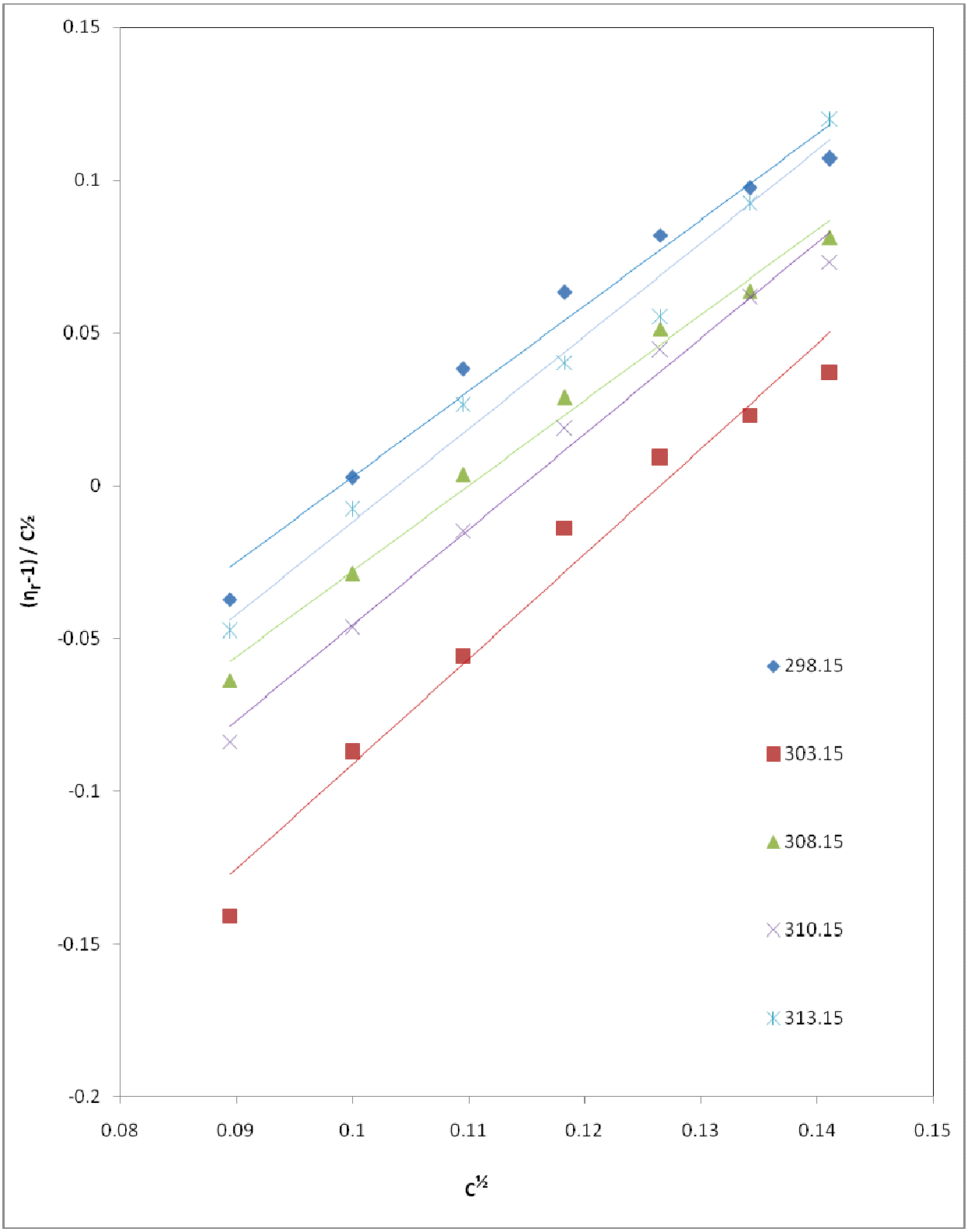


**Figure- 2:** Plot of $(\eta_r-1)/C^{½}$ Versus C½ of DS in NS at different temperatures.

order and disorder introduced by solute into the solution; positive B-coefficient shows strong alignment of solvent towards solute and is related to the effect of the solute on the structure of water [14, 15]. The strong interaction immobilizes the neighboring solvent molecules and presents large obstruction to viscous flow of solution thereby increasing viscosity. Thus the present system behaves as structure maker.

 

The parameters are obtained from Masson equation, Root equation, Jone-Dole equation and Moulik equation. The 'A' coefficients of Root's equation for set 1 to 3 are positive; while for set 4 are positive as well as negative. The positive values show strong solute-solute interactions with additives, while negative values show weak interactions. The 'B' coefficients are negative for set 1 to 3 and are positive for set 4. M and K coefficients are positive in all solvent systems & at all temperatures. M values are of low magnitudes & K values are of higher magnitudes. These models satisfy the investigated systems.

## 1.3 CONCLUSIONS:

From densitometric and viscometric studies of aqueous solutions of DS and in presence of additives as NaCl, KCl, Dextrose, Sodium lactate etc. at different temperatures shows,

1. All the values of $\phi v^0$ at all temperatures are positive and higher; suggest the strong solute-solvent interactions in aqueous solution, that may have the implication for the permeation of those molecules through biological membranes. All the $S_v$ values are negative and are very small, suggesting weak solute-solute interactions.

2. The positive values of Jones-Dole coefficient 'B' at all temperatures indicates water structuring. Positive values of B suggesting strongly hydrated solute. Which indicates structure promoting tendency. (i. e. kosmotropes).

3. In aqueous solution (set-4) the values of apparent molar volume are positive and decrease with the extent of hydrogen bonding. Lower the apparent molar volume values, stronger are the hydrogen bonds.

4. Root's & Moulik equations are found to be obeyed for DS in presence and absence of additives.

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