



## Electrical Conductance and Ion Pair Formation of Sodium Diethyldithiocarbamate in Acetonitrile-Water Mixtures at 25°C

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### ABSTRACT

The conductance of sodium diethyldithiocarbamate (NaDDC) has been measured in acetonitrile-water mixtures at 25°C where the dielectric constant ( $D$ ) ranges from 46.52 up to 74.66. Using Fuoss-Onsager theory, the data was interpreted to obtain  $\Lambda_o$ ,  $K_A$  and  $\hat{a}$  values. For NaDDC, the ion parameter  $\hat{a}$  (express the solvation) and limiting equivalent conductance  $\Lambda_o$  decrease with decreasing the dielectric constant of the medium until it reaches a minimum and then increases again. The increase in  $\log K_A$  versus  $1/D$  plot shows the dominance of electrostatic interaction. By applying the solvent separated – ion pair model, the association constant ( $K_A$ ) is analyzed.

**Keywords:** Sodium diethyldithiocarbamate, limiting equivalent conductance ( $\Lambda_o$ ), ion association, ion pair.

### INTRODUCTION

This paper aims to illustrate the conductance measurements of NaDDC in acetonitrile-water mixtures at 25°C to shed light on this salt demeanor in mixed solvents. By using the Fuoss-Onsager conductance equation [1], the data was analyzed to explain the electrolyte-solvent interaction.

Dithiocarbamates (DTCs) are widely used in many applications and are produced in large amounts. They play an important role in agriculture [2-6] and in application as linkers in solid-phase organic synthesis (SPOS). [7-10] In industry, DTCs act as accelerators for the vulcanization process of rubber, as antioxidants, work as slimicides in pulp and paper manufacturing as well as in production of sugar, in the treatment of waste water, and as antifoulant for water cooling systems [11, 12]. Dithiocarbamates are utilized to a large extent in a clinical applications such as medicinal chemistry and have some implementations in the treatment of cancer. [13-15]

### EXPERIMENTAL SECTION

#### Materials:

Sodium diethyldithiocarbamate is Analar analytical reagent "BDH", acetonitrile (B.D.H) was purified previously as reported in reference [16]. The specific conductance ( $\kappa_o$ ) for purified acetonitrile was found to be  $7.90 \times 10^{-8} \Omega^{-1} \text{cm}^{-1}$ , density ( $d_{25^\circ\text{C}}$ ) of  $0.7762 \text{ gm/cm}^3$ , viscosity ( $\eta_{25^\circ\text{C}}$ ) =  $0.3409 \times 10^{-2}$  Poise and dielectric constant ( $D_{25^\circ\text{C}}$ ) = 36.61. Conductivity water obtained by passing ordinary distilled water from a tin still over a 60 cm long Elgastat deionizer and guarded against contamination with atmospheric  $\text{CO}_2$  by soda lime tubes. Its specific conductance ( $\kappa_o$ ) amounted to  $(1.51 \times 10^{-6}) \Omega^{-1} \text{cm}^{-1}$ . The dielectric constant values were used as reported in the literatures [16-19].

**Properties of Solvent Mixtures:**

Densities ( $d$ ), viscosities ( $\eta$ ) and dielectric constants of the acetonitrile-water mixtures were used as reported in the literatures [20, 21].

**Conductance Measurements:**

All solutions were prepared by weight. Salt was weighed on microbalance which reads to  $\pm 0.1$  mg. Dilution was carried out successively into the cell by siphoning the solvent by means of weighing pipette. Conductivity Bridge was model Crison GLP31 (Conductivity Measurement error  $\leq 0.5\%$  and the reproducibility:  $\pm 0.1\%$ ) and a cell with bright platinum electrodes was used. The cell constant was  $0.1 \text{ cm}^{-1}$  for dilute solutions. The conductivity meter was calibrated with three KCl conductivity standard solutions (0.001 M, electrical conductivity [EC] =  $147 \mu\text{S cm}^{-1}$  at  $25.0^\circ\text{C}$ ; 0.01M, EC =  $1413 \mu\text{S cm}^{-1}$  at  $25.0^\circ\text{C}$  and 0.1 M, EC =  $12.88 \text{ mS cm}^{-1}$  at  $25.0^\circ\text{C}$ ) supplied by Crison. The temperature is kept constant to within  $\pm 0.1^\circ\text{C}$  using a water ultra thermostat.

**RESULTS AND DISCUSSION**

Conductance of sodium diethyldithiocarbamate at  $25^\circ\text{C}$  in mixtures of acetonitrile and water was measured. The equivalent conductance  $\Lambda$  ( $\Omega^{-1}\text{equiv}^{-1}\text{cm}^2$ ) is calculated at several concentrations (equiv/l). By plotting the extrapolation of  $\Lambda$  against  $C^{1/2}$ , an approximate value of  $\Lambda_0$  was obtained as showing in **Figures (1-7)**. More precise values of the limiting equivalent conductance  $\Lambda_0$  were resulted from the following Fuoss-Kraus-Shedlovsky (F.K.S) equation:

$$1 / \Lambda S_{(z)} = 1 / \Lambda_0 + C \Lambda S_{(z)} f_{\pm}^2 / K_D \Lambda_0^2 \quad (1)$$

Where  $K_D$  is the dissociation constant and  $S_{(z)}$  is a function of  $z$  called Shedlovsky's function that was tabulated by Daggett. The  $z$  value could be determined from the expression:

$$z = \alpha (C\Lambda)^{1/2} / \Lambda_0^{3/2} \quad (2)$$

Where  $\alpha$  is the limiting tangent (Onsager slope). The plot of  $1/\Lambda S_{(z)}$  versus  $(C\Lambda S_{(z)} f_{\pm}^2)$  gives  $1/\Lambda_0$  as the intercept and  $1/K_D \Lambda_0^2$  as the slope.

More accurate values of  $\Lambda_0$ ,  $J$  (function of  $a$ ),  $\hat{a}$  and  $K_A$  were obtained from Fuoss-Onsager equation [1]. The starting  $\Lambda_0$  value was resulted from Fuoss-Kraus-Shedlovsky equation by the assist of specific computer program that was programmed on an IBM-PC. The desired accuracies in these calculations are  $\pm 0.02$  for  $\Lambda_0$ ;  $\pm 2$  for ( $J$  less than 200),  $\pm 5$  for ( $J$  with values range from 200 to 1000) and  $\pm 10$  for ( $J$  more than 1000).

The standard deviation  $\sigma_\Lambda$  was calculated using the equation [22]:

$$\sigma_\Lambda = \frac{\{\sum (\Lambda \text{ calculated} - \Lambda \text{ observed})^2\}^{1/2}}{(N - 1)^{1/2}} \quad (3)$$

Where  $N$  is the number of experimental points.

The data were analyzed by using the Fuoss – Onsager equation. The outcomes are shown in **Table 1**.

From **Table 1** and **Figure 8**, it is obviously noticed that,  $\Lambda_0$  for sodium diethyldithiocarbamate in acetonitrile-water mixtures were found to exhibit minima at 30 % acetonitrile. This is an unexpected behavior, since on decreasing the dielectric constant, the ionic mobility is hindered and ion-pair formation is more possible, consequently,  $\Lambda_0$  should decrease with the decrease of dielectric constant. The viscosity of the medium will affect the ions mobility. As the viscosity of the medium is increased, the ion mobility will decrease and then the ion mobility will increase as the viscosity of the medium decrease. This explains the unexpected behavior of  $\Lambda_0$  as a function of the dielectric constant and the viscosity of the medium.

A. E. Mahgoub and A. Lasson [23], have reported comparable manner for  $\Lambda_0$  in the condition of Rubidium iodide in mixtures of acetonitrile with water at  $25^\circ\text{C}$ . The values of  $\Lambda_0$  of lithium nitrate and sodium nitrate, in acetonitrile – water mixtures at  $25^\circ\text{C}$  [24], are found to decrease as the percentage of acetonitrile increase in the mixture up to a certain limit and then increase again. This is in accordance with our experimental observations.

M. S. K. Niazi and A. Khan, [25] have reported similar behavior for  $\Lambda_0$  in case of sodium perchlorate and sodium benzoate in acetonitrile – water mixtures. The values of  $\Lambda_0$  for both salts, in the same mixtures, dropped with increasing the percentage of acetonitrile in the mixture, up to 22.7 mole percentage for sodium perchlorate and 10.0 mole percentage for sodium benzoate. Above these limits  $\Lambda_0$  increased with further addition of acetonitrile.

N. G. Foster and E. S. Amis, [26] have reported similar behavior in case of tetraethylammonium picrate in methanol-water and ethanol – water mixtures [27] and for potassium chloride in methanol – water systems [28].

H. Sadek *et al.*, [29] have obtained hassling trend for  $\Lambda_0$  of *s*-alkylisothiuronium iodides in methanol-water mixtures at 25°C.

El-Hammamy *et al.*, [30-34] have gotten the same action for  $\Lambda_0$  in the following: N-N-diphenyl-*s*-alkylisothiuronium bromides in methanol-water at 25°C [30], Co(II) complexes of acetone-thiosemicarbazone halides in methanol-water at 25°C [31], acetylcholine halides and perchlorate in methanol-water at 25°C [32], *s*-acetylthiocholine halides and perchlorate in ethanol-water at 25°C [33] and also in case of *s*-acetylthiocholine halides in methanol-water at 25°C [34].

The variations of  $J$ ,  $\bar{a}$  and  $K_A$  (association constant) with the dielectric constant are shown in **Figures (9-11)**. The initial slight decreases in  $\bar{a}$  for sodium diethyldithiocarbamate, in acetonitrile-water mixtures, may be elucidated according to hydrogen bonding force where the hydrogen bonds strength between ions and water was strongly influenced by adding little quantities of acetonitrile. On accretion of acetonitrile, water-water interactions are broken down and replaced by weaker acetonitrile-water interactions. This was in agreement with the measured endothermic heat of mixing [35] and then a slight increase in  $\bar{a}$  values was observed.

The increase in  $\log K_A$  versus  $1/D$  plot, shown in **Figure 11**, for sodium diethyldithiocarbamate in acetonitrile-water mixtures, can be clarified on the basis that, the dielectric constant decreases with increasing the acetonitrile content as shown in **Table 1**. This increase in  $\log K_A$  versus  $1/D$  plot shows the dominance of electrostatic interaction.

E. Hawlicka *et al.*, [36] have measured the conductance of tetraethylammonium iodide in acetonitrile-water solutions. In the presence of acetonitrile, it was found that tetraethylammonium ion loses its hydrophobic properties and become unsolvated. On the decrease of static dielectric constant of the acetonitrile-water mixtures, the association constant, calculated from the conductivity measurements, rises slowly. Because of the increase of the solution viscosity, the self-diffusion coefficients for both of water and acetonitrile in the presence of  $\text{Et}_4\text{NI}$  detract.

Krishnam Raju *et al.*, [37] have measured the conductance of lithium acetate in 0-90% acetonitrile-water mixtures at 25°C. The association constants were calculated using the Fuoss-Hsia equation with Fernandez-Prini coefficients. On plotting  $\log K_A$  versus  $1/D$  a straight line was obtained in conformity with the Denison-Ramsey ion-pair formation modified theory. Also Walden products were calculated. The results are interpreted in terms of solvent separated ion-pairs and contact ion-pairs.

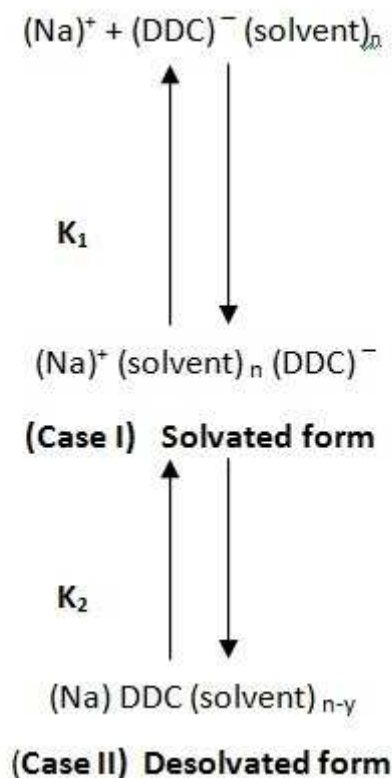
The trend of  $K_A$  in the present work was explained in the light of the  $U$  term as represented in the following equation [38]:

$$\ln K_A = \ln (4 \pi N_A \bar{a}^3 / 3000) + e^2 / \bar{a} D k T + U \quad (4)$$

$$\text{Where } U = \Delta S / k - E_s / k T \quad (5)$$

The factor  $E_s/kT$  introduced by Gilkerson [39] indicates the ion-dipole interaction energy, which differs when ion-pairs are taken into consideration. The entropy term  $\Delta S/k$  refers to the entropy change due to various arrangements of solvent molecules around free ion and ion pairs. From **Table 2**, it is evident that  $U$  increases as the acetonitrile weight percentage increases. This approves that, the entropy term increases with increasing weight percentage of acetonitrile.

Eventually, the solvent separated ion-pair model can be applied [40]. The association steps are illustrated in **Scheme 1**:



Where  $y$  = the number of escaping solvent molecules from solvation

Thus, the association constant  $K_A$  is obtained from the conductance measurements according to the equation:

$$K_A = K \Sigma \frac{(C_{(\text{ion-pairs})})}{(C_{(\text{Na})^+})(C_{(\text{DDC})^- (\text{solvent})_n})} = K_1 (1 + K_2) \quad (6)$$

Where,  $K_1 = 4 \pi N \hat{a}^3 e^b / 3000$  and  $b = e^2 / \hat{a} D T k$ . Then  $K_2$  can be calculated

From the data given in **Table 2**, it was clear that the values of  $K_2$  increase with decreasing the dielectric constant. This means; as the dielectric constant decreases, the ion-pair prefers the desolvated form (II) more than solvated form (I).

**Figure 12** shows that, the **Walden Product**  $\Lambda_0 \eta_0$ , as a function of solvent composition for sodium diethyldithiocarbamate in acetonitrile-water mixtures at 25°C, varies in the usual manner, i.e. decreases with decreasing the dielectric constant. This is in accordance with general finding on several small sizes (Uni–Univalent) [41, 42] and (Bi– Bivalent) [43] systems and can be attributed to ion-solvent relaxation drag [44, 45].

The electrostatic radii  $R^+$  and  $R^-$  were calculated by using the Stokes' equation:

$$R^\pm = 0.8194 \times 10^{-8} / \lambda_0^\pm \eta_0 \quad (7)$$

The application of **Stokes'** equation depends on the determination of the of the ionic conductance. According to **Fuoss** assumption [46], the ionic equivalent conductance for cations and anions in acetonitrile-water mixtures can be calculated as transport number is independent on the solvent composition,. These values are summarized in **Table 3**. **From Table 3** It can be seen easily that, for sodium diethyldithiocarbamate  $R^+$  and  $R^-$  increase on decreasing the dielectric constant. On comparing the summation of electrostatic radii ( $R^+ + R^-$ ) with the closest distance of approach  $\hat{a}$ , which was previously derived using **Fuoss Onsager** equation, one can observe that  $\hat{a}$  is always larger than electrostatic radii ( $R^+ + R^-$ ), obtained from **Stokes'** equation in acetonitrile-water mixtures due to the solvation of ions.

Hughes and Hartly [47], observed the same trend in electrostatic radii for  $\text{Li}^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{N}(\text{C}_2\text{H}_5)_4^+$ ,  $\text{Cl}^-$ ,  $\text{Br}^-$ ,  $\text{I}^-$ ,  $\text{ClO}_4^-$  and picrate ions in alcohol-water and acetone-water mixtures.

Kortium and Welier [48] calculated  $R$ 's for perchlorate ion ( $\text{Pi}$ ) in both alcohol and water solutions. The results were:  $R_{\text{pi}^-}(\text{H}_2\text{O}) = 3.33 \text{ \AA}$ , in  $(\text{MeOH}) = 3.7 \text{ \AA}$  and in  $(\text{EtOH}) = 3.8 \text{ \AA}$ . The authors also found that  $R$ 's for  $\text{Li}^+$  in ethanol-water increases from water to ethanol. On comparing the summation of electrostatic radii ( $R^- + R^+$ ) with the closest distance of approach  $\hat{a}$  which was previously derived using Fuoss Onsager equation,  $\hat{a}$  was always greater except in case of rich methanol content.

This behavior can be explained using **Nightingale's** [49] conclusion, for water and mixed aqueous solvents of high dielectric constant and high viscosity, **Stokes' equation** gives inappropriate small value due to discontinuity of the medium.

**Table 1: Characteristic parameters for sodium diethyldithiocarbamate in acetonitrile-water mixtures at 25°C**

Sodium diethyldithiocarbamate								
Wt %	$D^*$	$10^3/D$	$\Lambda_\infty$	J	$\hat{a}$	$K_A$	$\log K_A$	$\sigma_A$
10	74.66	13.394	142.64	409.11	6.31	66.976	1.8259	0.129
20	70.48	14.188	120.53	393.06	6.10	126.01	2.1004	0.065
30	65.78	15.202	115.18	411.89	5.62	169.31	2.2287	0.114
40	60.20	16.611	124.66	550.54	5.77	246.06	2.3910	0.158
50	55.70	17.953	134.31	777.80	6.39	403.33	2.6057	0.171
60	50.77	19.697	143.84	1220.5	7.75	661.62	2.8206	0.246
70	46.52	21.496	155.35	1641.2	7.88	780.48	2.8924	0.382

$D^*$ : Reference [21]

**Table 2:  $K_2$  and U values for sodium diethyldithiocarbamate in acetonitrile-water mixtures at 25°C**

Sodium diethyldithiocarbamate				
Acetonitrile wt%	$K_A$	$K_1$	$K_2$	U
10	66.976	2.080	31.202	3.472
20	126.01	2.106	58.842	4.092
30	169.31	2.036	82.145	4.421
40	246.06	2.429	100.29	4.618
50	403.33	3.174	126.08	4.845
60	661.62	4.873	134.78	4.911
70	780.48	5.686	136.27	4.922

**Table 3: Calculation of radii of the ions for sodium diethyldithiocarbamate in acetonitrile-water mixtures at 25°C**

Sodium diethyldithiocarbamate							
Acetonitrile wt %	$\Lambda_\infty \eta_\infty^{(1)}$	$\lambda_\infty^+ \eta_\infty^{(1)}$	$\lambda_\infty^- \eta_\infty^{(1)}$	$R^{+(2)}$	$R^{-(2)}$	$R^+ + R^{-(2)}$	$\hat{a}^{(2)}$
10	1.3979	0.6460	0.7519	1.268	1.090	2.358	6.31
20	1.1703	0.5409	0.6295	1.515	1.302	2.817	6.10
30	1.0504	0.4854	0.5650	1.688	1.450	3.138	5.62
40	1.0521	0.4862	0.5659	1.685	1.448	3.133	5.77
50	1.0060	0.4649	0.5411	1.763	1.514	3.277	6.39
60	0.9422	0.4354	0.5068	1.882	1.617	3.499	7.75
70	0.8902	0.4114	0.4788	1.992	1.711	3.703	7.88

(1)  $\text{ohm}^{-1} \text{equiv}^{-1} \text{cm}^2 \text{p}$   
(2)  $\text{\AA}$

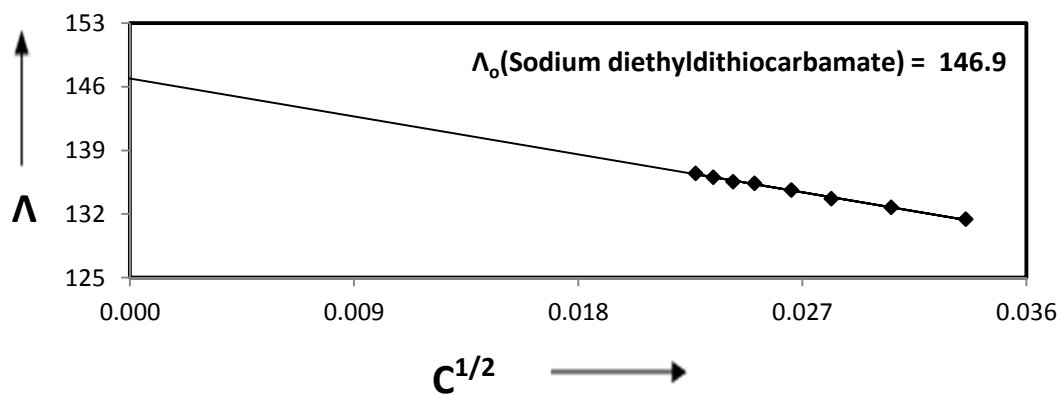


Fig. 1: Conductance of sodium diethyldithiocarbamate in 10% acetonitrile-water mixture at 25°C

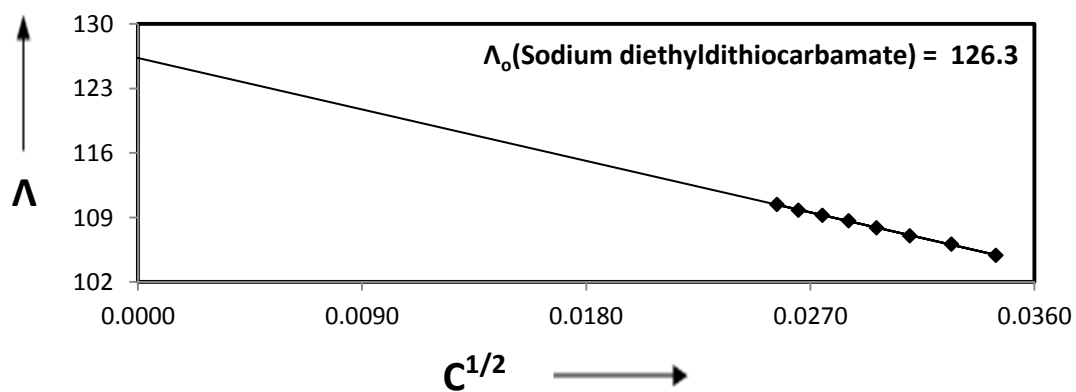


Fig. 2: Conductance of sodium diethyldithiocarbamate in 20% acetonitrile-water mixture at 25°C

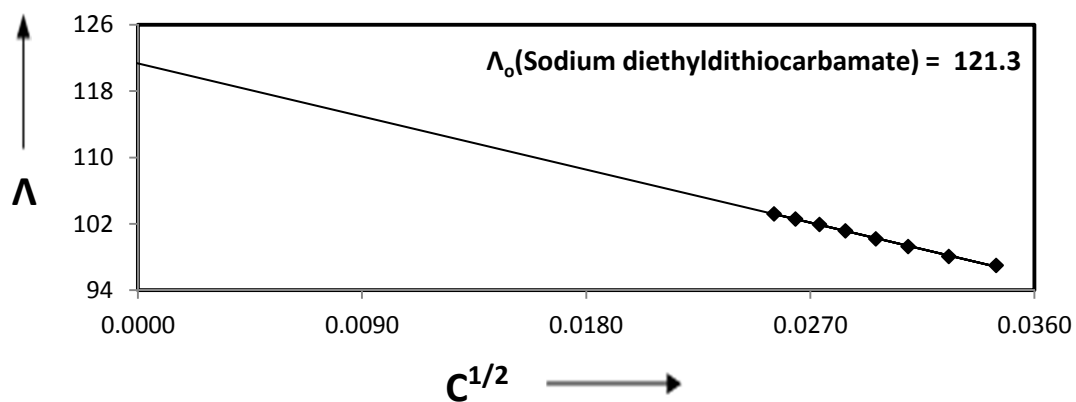


Fig. 3: Conductance of sodium diethyldithiocarbamate in 30% acetonitrile-water mixture at 25°C

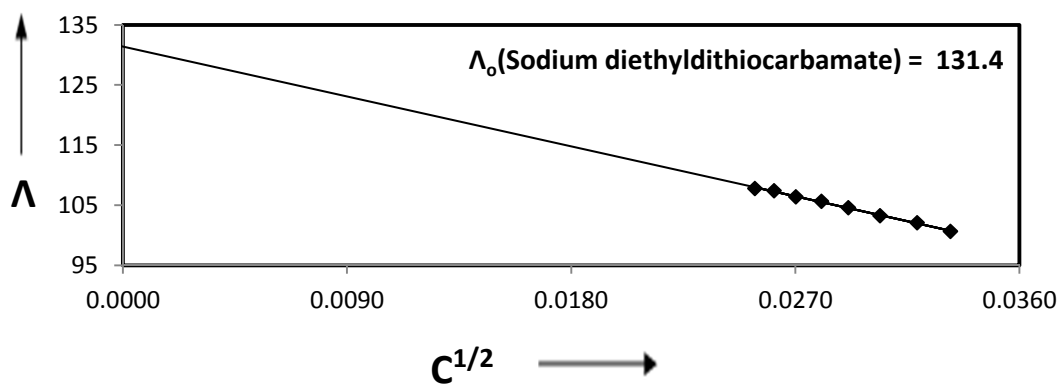


Fig. 4: Conductance of sodium diethyldithiocarbamate in 40% acetonitrile-water mixture at 25°C

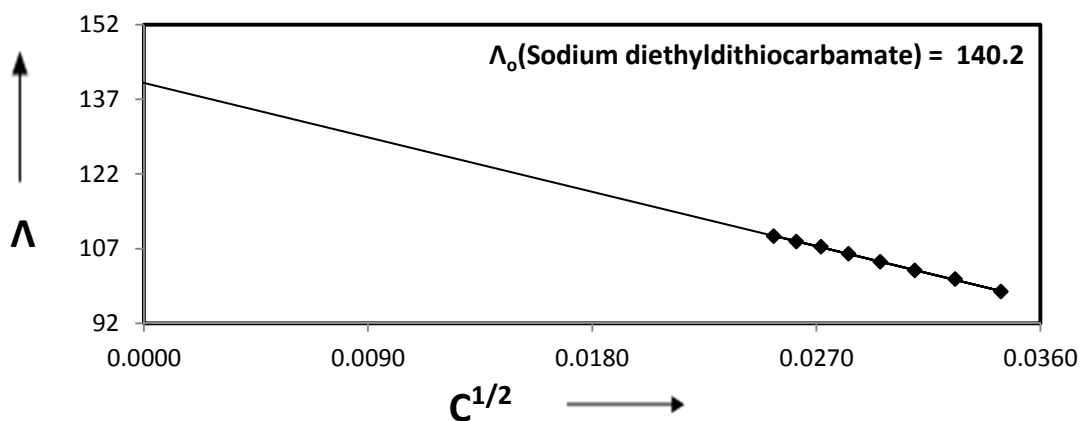


Fig. 5: Conductance of sodium diethyldithiocarbamate in 50% acetonitrile-water mixture at 25°C

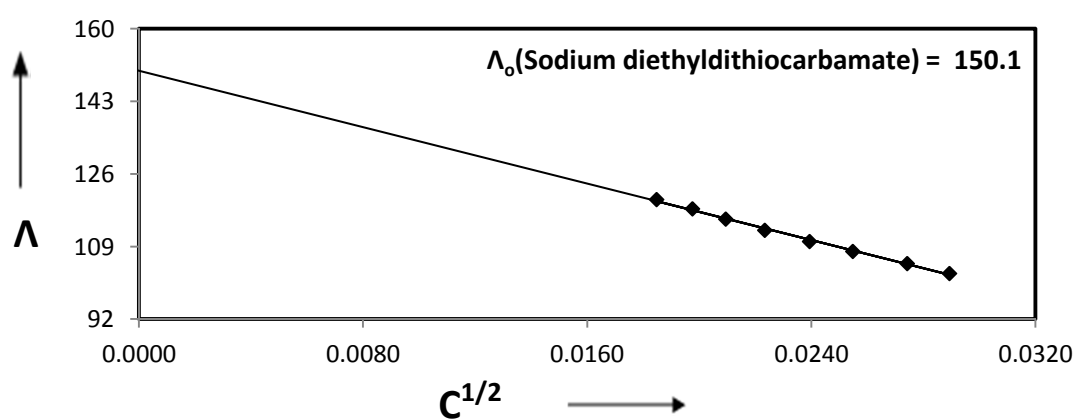


Fig. 6: Conductance of sodium diethyldithiocarbamate in 60% acetonitrile-water mixture at 25°C

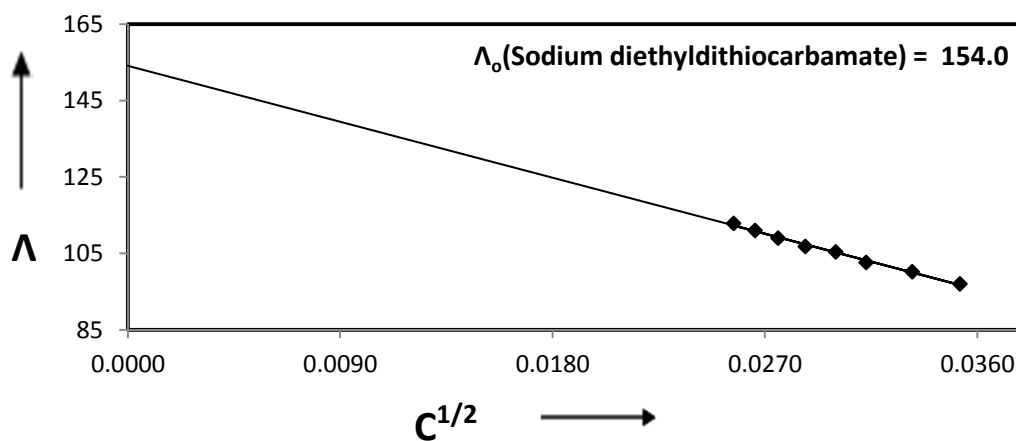


Fig. 7: Conductance of sodium diethyldithiocarbamate in 70% acetonitrile-water mixture at 25°C

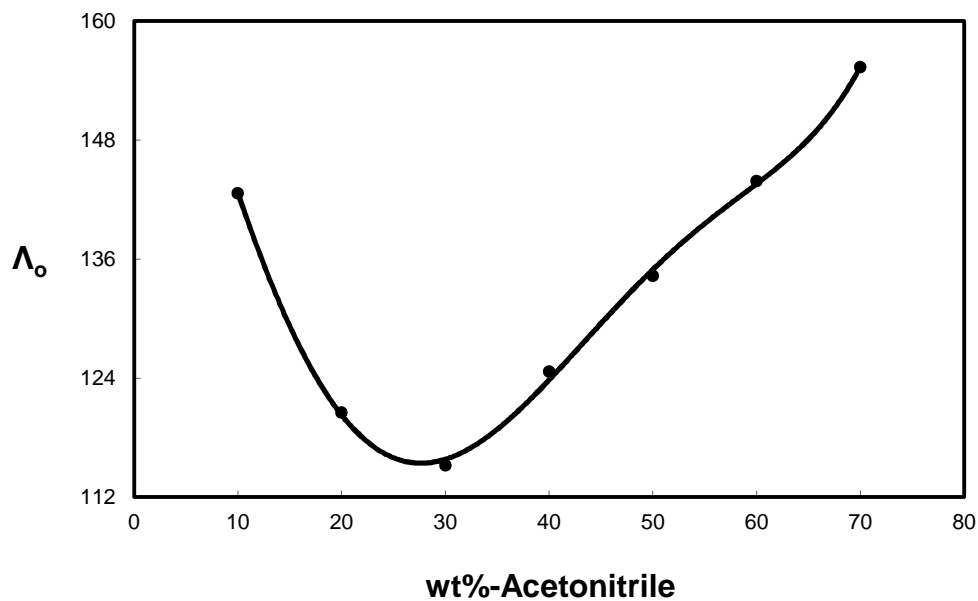


Fig. 8: Variation of  $\Lambda_0$  of sodium diethyldithiocarbamate with the composition of solvent mixtures

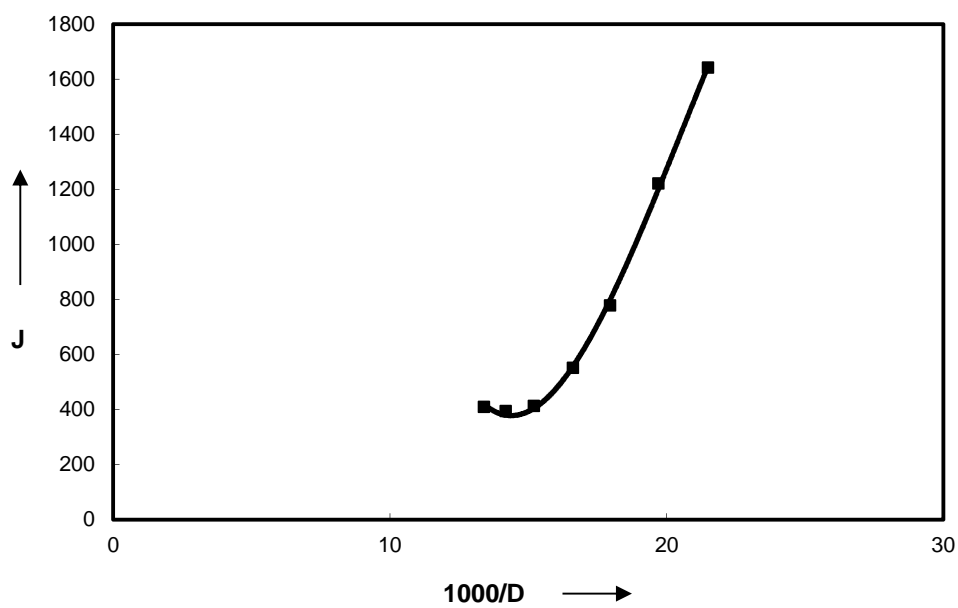


Fig. 9: Variation of  $J$  with dielectric constant for sodium diethyldithiocarbamate in acetonitrile-water mixtures



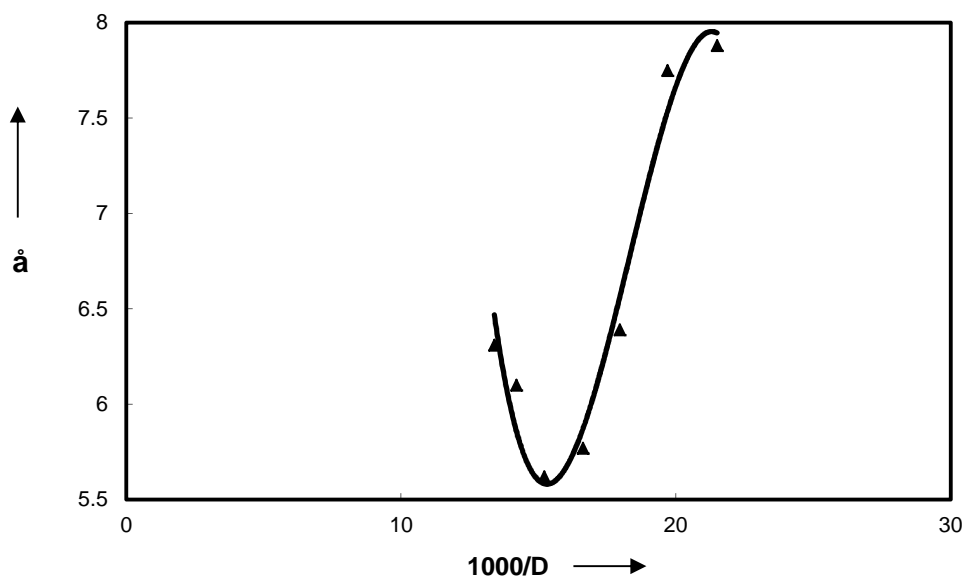


Fig. 10: Variation of  $\alpha$  with dielectric constant for sodium diethyldithiocarbamate in acetonitrile-water mixtures

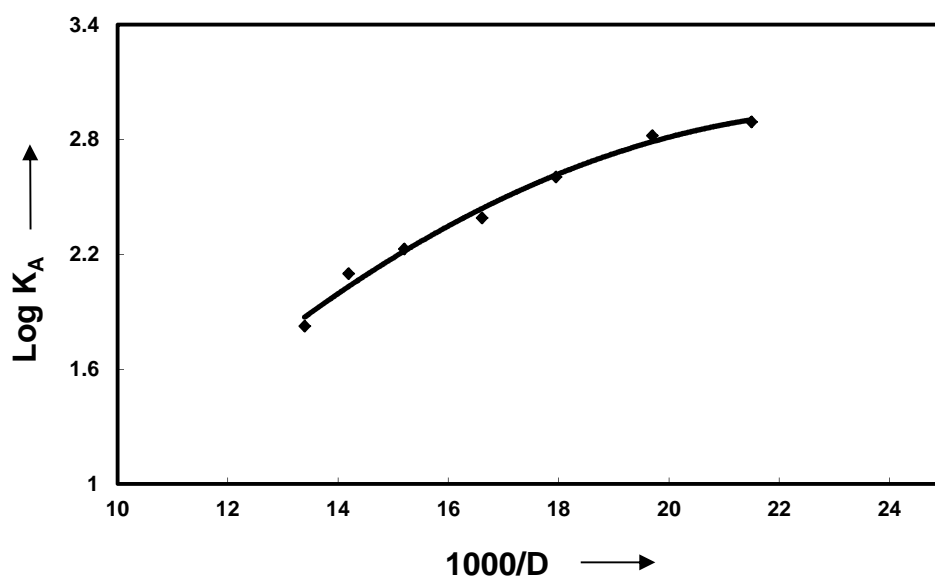


Fig. 11: Variation of  $\log K_A$  with dielectric constant for sodium diethyldithiocarbamate in acetonitrile-water mixtures

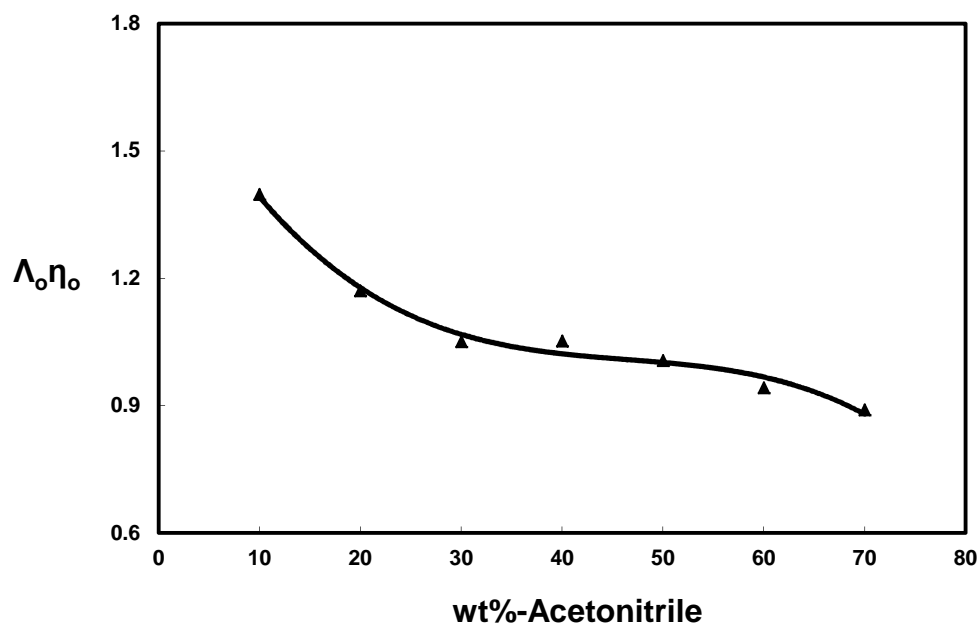


Fig. 12: Variation of Walden Product with the composition of solvent mixtures

#### CONCLUSION

This paper aims to shed light on the association process of the organic compound: sodium diethyldithiocarbamate (NaDDC); which has been interpreted by solvent separated – ion pair model. Moreover, the limiting equivalent conductance ( $\Lambda_0$ ) of NaDDC in mixtures of acetonitrile-water at 25°C was discussed in details. By plotting the extrapolation of  $\Lambda$  against  $C^{1/2}$ , preliminary values of  $\Lambda_0$  were estimated. Consequently, more precise values of  $\Lambda_0$  were resulted by applying Fuoss-Kraus-Shedlovsky equation. Finally, more accurate values of  $\Lambda_0$ ,  $J$  (function of  $a$ ),  $\hat{a}$  and  $K_A$  were obtained from Fuoss-Onsager equation. A comparison between the summation of electrostatic radii ( $R^+ + R^-$ ) and the closest distance of approach  $\hat{a}$  was done; it was observed that  $\hat{a}$  is always larger than electrostatic radii ( $R^+ + R^-$ ) due to the solvation of ions.

#### REFERENCES

- [1] RM Fuoss; L. Onsager, *J. Phys. Chem.*, **1957**, 61, 668; RM Fuoss, *J. Amer. Chem. Soc.*, **1959**, 81, 2659.
- [2] C Rafin; E Veignie; M Sancholle; D Postal; C Len; P Villa; G Ronco, *J. Agric. Food. Chem.*, **2000**, 48, 5283.
- [3] W Chen-Hsien, *Synthesis*, **1981**, 622
- [4] T Mizuno; I Nishiguchi; T Okushi; T Hirashima, *Tetrahedron Lett.*, **1991**, 32, 6867.
- [5] YS Chen; I Schuphan; JE Casida, *J. Agric. Food Chem.*, **1979**, 27, 709.
- [6] C Len; D Postal; G Ronco; P Villa; C Goubert; E Jeufrault; B Mathon; H Simon, *J. Agric. Food Chem.*, **1997**, 45, 3.
- [7] P Morf; F Raimondi; HG Nothofer; B Schnyder; A Yasuda; JM Wessels; TA Jung, *Langmuir*, **2006**, 22, 658.
- [8] A McClain; YL Hsieh, *J. Appl. Polym. Sci.*, **2004**, 92, 218.
- [9] BP Bongar; VS Sadavarte; LS Uppalla, *J. Chem. Res. (S)*, **2004**, 9, 450.
- [10] AD Dunn, WD Rudorf. Carbon Disulphide in Organic Chemistry, Ellis Horwood: Chichester, U. K., **1989**, 226.
- [11] PC Wales, in: PE Helliker (Ed.), Evaluation of Methyl Isothiocyanate as a Toxic Air Contaminant, California Environmental Protection Agency, Sacramento, **2002**, p. 1.
- [12] L Monser; N Adhoum, *Sep. Purif. Technol.*, **2002**, 26, 137.
- [13] L Ronconi; C Marzano; P Zanella; M Corsini; G Miolo; C Macca; A Trevisan; D Fregona, *J. Med. Chem.*, **2006**, 49, 1648.
- [14] W Walter; KD Bode, *Angew. Chem. Int. Ed. Engl.*, **1967**, 6, 281.
- [15] GH Elgemeie; SH Sayed, *Synthesis*, **2001**, 1747.
- [16] MAH Daghash, M.Sc. thesis in Chemistry, Faculty of Science, Alexandria University, Egypt, **2013**.
- [17] RA Robinson, RH Stokes. "Electrolyte Solutions", Butterworths, 2<sup>nd</sup> ed, London, **1959**.
- [18] NH El-Hammamy; MM El-Kholy; GA Ibrahim; AI Kawana, *J. Adv. Appl. Sci. Res.*, **2010**, 1(3), 168.
- [19] NH El-Hammamy; AA Hasanien; MF Amira; FM Abd El-Halim, *J Electrochem. Soc.*, India, **1984**, 33, 43.
- [20] A D'Aprano; RM Fuoss, *J. Phys. Chem.*, **1969**, 73, 400.

- [21] MSK Niazi; J Ali, *Bull. Chem. Soc. Japan*, **1990**, 63, 3619.
- [22] GW Brown, *Am. J. Dis. Child.*, **1982**, 136, 937.
- [23] AE Mahgoub; A Lasson, *Acta Chem. Scand. A.*, **1975**, 29, 537.
- [24] AM Sanchez; C Moran; C Quintana; A Vivo, *J. Solution Chem.*, **1989**, 18(10), 993.
- [25] MSK Niazi; A Khan, *J. Chem. Soc. Pak.*, **1992**, 14(4), 265.
- [26] NG Foster; ES Amis, *Z. Phys. Chem. Neufolge*, **1956**, 7, 360.
- [27] R Whoton; ES Amis, *Z. Phys. Chem. Neufolge*, **1958**, 17, 300.
- [28] NG Foster; ES Amis, *Z. Phys. Chem. Neufolge*, **1955**, 3, 366.
- [29] H Sadek; AM Hafez; NH El-Hammamy, *Pak. J. Sci. Res.*, **1977**, 29, 100.
- [30] NH El-Hammamy; AA Hasanein; HA Mahmoud; FM Abd El-Halim, *J. Chem. Soc. Pak.*, **1986**, 8, 125.
- [31] NH El-Hammamy; SA El-Shazly; AI Kawana; FM Abd El-Halim, *J. Chem. Soc. Pak.*, **1991**, 13, 236.
- [32] NH El-Hammamy; AM Ahmed; FF El-Bardisy; FM Abd El-Halim, *Alex. Eng. J.*, **1989**, 28, 867.
- [33] NH El-Hammamy; AI Kawana; FF El-Bardisy; FM Abd El-Halim, *Bull. Electrochem.*, **1995**, 11, 253.
- [34] NH El-Hammamy; HM El-Kashlan; HM Moharam, *Asian J. Chem.*, **2013**, 25(4), 2221.
- [35] DA Armitage; MJ Blandamer; MJ Foster; NJ Hidden; KW Morcom; MCR Symons; MJ Wootten, *Trans. Faraday Soc.*, **1968**, 64, 1193.
- [36] E Hawlicka; R Grabowski; W Reimschüssel, *Berichte der Bunsengesellschaft für physikalische Chemie*, **2010**, 94(2), 158.
- [37] K Raju; V. Maniaiah; B. Sethuram; TN Rao, *Physics and Chemistry of Liquids: An International Journal*, **2006**, 22(3), 163.
- [38] F Accascina; A D'Aprano; R Triolo, *J. Phys. Chem.*, **1967**, 71, 3469.
- [39] WR Gilkerson, *J. Chem. Phys.* **1956**, 25, 1199.
- [40] DF Evans; P Gardam, *J. Phys. Chem.*, **1969**, 73, 158.
- [41] JE Lind; RM Fuoss, *J. Phys. Chem.*, **1961**, 65, 1001.
- [42] RW Kunze; RM Fuoss, *J. Phys. Chem.*, **1963**, 67, 911.
- [43] G Atkinsen; S Petrucci, *J. Phys. Chem.*, **1964**, 86, 7.
- [44] RM Fuoss, *Proc. Nat. Acad. Sci. U.S.A.*, **1959**, 45, 807.
- [45] R Zwanzig, *J. Phys. Chem.*, **1963**, 38, 1603-1605.
- [46] RM Fuoss, *J. Amer. Chem. Soc.*, **1958**, 80, 5059.
- [47] OL Hughes; H Hartly, *Phil. Mag.*, **1933**, 15, 610.
- [48] G Kortium; A Welier, *Z. Nature. Forsch.*, **1950**, 590.
- [49] ER Nightingale, *J. Phys. Chem.*, **1959**, 63, 1381.