Orientational and dielectric behaviour of N, N-dimethylformamide in different non-polar solvents

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Dielectric behaviour of concentrated solutions of different mole fractions of N, N-dimethylformamide in various non-polar solvents has been studied using precision LCR meter with four terminal liquid dielectric test fixture over a frequency range 20 Hz to 2 MHz at 303.15 K temperature. The dielectric parameters namely dielectric constant (ε'), dielectric loss (ε'') and static dielectric constant (ε_0) have been determined by capacitive measurement method. The limiting dielectric constant (ε_∞) has been determined by Abbe's refractometer for the same solutions at 303.15 K temperature. Kirkwood correlation factor g^{eff} has also been calculated using determined values of ε_0 and ε_∞ to gain information about the orientational behaviour of N, N-dimethylformamide in different non-polar solvent environments.

Keywords: Dielectric constant, Dielectric loss, Kirkwood correlation factor

1 Introduction

Investigation of physical and dielectric properties of organic materials has provided an insight to understand the structural behavior of these materials. In order to understand the interaction of a continuously varying electromagnetic field with a material medium, it is important to know its complex dielectric permittivity over a range of frequency of the applied field at different temperatures. The measured values of complex permittivity at the various frequencies and temperatures are used to evaluate various dielectric parameters like relaxation time, distribution parameter, dipole moment, excess dielectric constant, molar polarization and various thermodynamical parameters like entropy, enthalpy, Gibbs free energy etc. These dielectric parameters provide valuable information about the molecular structure, inter- and intra-molecular rotations and solute-solute and solute-solvent interactions of the materials. Hydrogen bond constitutes a very interesting class of intermolecular interactions. Study of hydrogen bonded molecular interactions is of significant importance in the fields of chemistry, biophysics, pharmacy, molecular biology and biopolymers[1,2]. N, N-dimethylformamide (DMF) is able to form H-bonded network structures with

dipolar aprotic and protic solvents[3] and it is used in the production of acrylic fibers, plastics, pesticides and synthetic leather. It is also used in the manufacture of paint, film and adhesives. Hydrogen bonding ability and large number of applications of N, N-dimethylformamide in the various fields have drawn attention of many researchers to study the structural properties of pure N, N-dimethylformamide and DMF mixed dipolar solvents by dielectric measurements. Dielectric investigation of hydrogen bonded compounds provides valuable information about molecular interactions[4-6]. Sengwa *et al.*[3] studied structure and hydrogen bonding in binary mixtures of N, N-dimethylformamide with some dipolar aprotic solvents like dimethyl sulphoxide and protic solvents like water, ethyl alcohol and ethylene glycol by dielectric characterization. Malathi *et al.*[7] studied mutual interaction of amides with non-polar solvents to understand the conformational stability of protein molecules. Thenappan and Sankar[8] studied hydrogen bonded complexes of alcohols with N, N-dimethylformamide. Dielectric relaxation study of the dilute solutions of binary mixtures of pyridine and N, N-dimethylformamide in benzene was conducted by Kumar *et al.*[9]. Dynamics of the N, N-dimethylformamide was studied by Barthel *et al.*[10]. Orientational behaviour of N, N – dimethylformamide in different non polar solvents has not been studied

much. Many applications of DMF in varieties of fields and less amount of existing literature on the study of orientational dynamics of DMF in different solvent environments through dielectric methods has motivated us to conduct this study. With an aim to understand the dielectric behavior as well as orientational dynamics of the N, N-dimethylformamide in three different non-polar solvents benzene, carbon tetrachloride and 1, 4-dioxane, this study was conducted.

2 Experimental

2.1 Materials and sample preparation

Analytical reagent grade N, N-dimethylformamide (CH_3-CO-NH_2), Benzene(C_6H_6), carbon tetrachloride (CCl_4), and 1,4-dioxane ($C_4H_8O_2$) were procured from E. Merck, Central Drug House [P] Ltd., India and High Purity Laboratory Chemicals Pvt. Ltd., India and were used without further purification. The solutions of different concentrations were made by mixing a certain weight of polar solute N,N-dimethylformamide in non-polar solvents benzene, carbon tetrachloride and 1,4-dioxane using electronic balance of accuracy ±0.0001g. Then the weight fractions of solutions were converted into mole fractions by the following equation:

Mole fraction of solute

$$X_2 = \frac{\left[W_2/M_2 \right]}{\left[W_1/M_1 \right] + \left[W_2/M_2 \right]} \qquad \dots (1)$$

where; W_2 is weight of solute, M_2 is molecular weight of solute, W_1 is weight of solvent and M_1 is molecular weight of solvent.

2.2 Experimental techniques and measurement

The measurement of dielectric constant (ε') and dielectric loss (ε'') between 20 Hz to 2 MHz frequency range, of the prepared liquid samples were done by Agilent E 4980 precision LCR meter with a four terminal liquid test fixture (Agilent-16452) using the capacitive measurement method[11,12]. Dielectric constant (ε') at 2 MHz frequency was taken as static dielectric

constant (ε_0) of the sample. While the limiting dielectric constant ($\varepsilon_\infty = n_D{}^2$) for same solutions was determined by measuring refractive indices (n_D) using Abbe's refractometer at the wavelength of sodium–D light with accuracy of ±0.0001. The density of the mixtures was determined using a pyknometer by relative measurement method. The pyknometer was calibrated with double distilled water and gave an estimated reproducibility ± 0.0001gcm^{-3}. All measurements were made at 303.15 K and the temperature was controlled thermostatically by using the temperature control water bath with accuracy of 0.1 K.

3 Results and Discussion

The measured and literature values of ε_0 and ε_∞ of the pure N,N-dimethylformamide, benzene, carbon tetrachloride and 1,4-dioxane at 303.15 K are listed in Table 1 for comparison. The experimental values of this study are in good agreement with the literature values.

The Kirkwood correlation factor (g^{eff}) for solutions of polar solute in non-polar solvent has been calculated using equation[18,19]:

$$g^{eff}\mu_2^2 = \frac{9kT(2\varepsilon_0 + \varepsilon_\infty)^2}{4\pi N X_2(\varepsilon_\infty + 2)^2(2\varepsilon_0 + 1)}\left[\frac{V_m(\varepsilon_0 - 1)}{\varepsilon_0} \right. $$
$$\left. - \frac{3V_1X_1(\varepsilon_1 - 1)}{2\varepsilon_0 + \varepsilon_1} - \frac{3V_2X_2(\varepsilon_\infty - 1)}{2\varepsilon_0 + \varepsilon_\infty} \right]$$
$$\dots (2)$$

where, ε_0 is the static dielectric constant of the solution, ε_∞ is the limiting dielectric constant of the solution, ε_1 is the static permittivity of the pure solvent, X_1 is the mole fraction of the solvent, X_2 is the mole fraction of the solute, V_m is the molar volume of the solution, V_1 and V_2 are molar volume of the pure solvent and pure solute respectively. k, T and N are Boltzmann's constant, absolute temperature and Avogadro's number, respectively. While μ_2 is the dipole moment of solute, i.e., N,N-dimethylformamide and it is taken[20] as 3.86 D.

Molar polarization of the solutions is given by[21]:

Table 1 — Comparison of measured and literature values of static dielectric constant (ε_0) and limiting dielectric constant (ε_∞) of N, N-dimethylformamide, benzene, carbon tetrachloride and 1, 4-dioxane at 303.15 K.

Compound	Measured dielectric constant		Literature dielectric constant	
	At 2 MHz frequency	At optical frequency	At 2 MHz Frequency	At optical frequency
N, N-dimethylformamide	36.93	2.0357	36.55[3]	2.0410[4]
Benzene	2.26	2.2455	2.26[13]	2.1933[14]
Carbon tetrachloride	2.22	2.1155	2.24[17]	2.1228[15]
1,4-dioxane	2.21	2.0181	2.26[3]	2.0390[16]

$$P = X_1P_1 + X_2P_2 \qquad \qquad ... (3)$$

From eq. (2) molar polarization of solute is,

$$P_2 = P_1 + \left[\frac{P-P_1}{X_2}\right] \qquad \qquad ... (4)$$

where; P_1 is the molar polarization of the solvent, which is given by:

$$P_1 = \left(\frac{\varepsilon_1-1}{\varepsilon_1+2}\right)V_1 \qquad \qquad ... (5)$$

P is the molar polarization of the solution which is given by:

$$P = \left(\frac{\varepsilon_m-1}{\varepsilon_m+2}\right)V_m \qquad \qquad ... (6)$$

V_m is the molar volume of the solution which is given by:

$$V_m = (w_1M_1 + w_2M_2)/\rho \qquad \qquad ... (7)$$

where; w_1 is weight fraction of pure solvent, M_1 is molecular weight of pure solvent, w_2 is weight fraction of pure solute, M_2 is molecular weight of pure solute and ρ is the density of the solution.

Determined values of the static dielectric constant (ε_0), limiting dielectric constant (ε_∞), effective Kirkwood correlation factor (g^{eff}), density (ρ), molar volume (V_m) and molar polarization of solute (P_2) of the solutions of N,N-dimethylformamide in benzene, carbon tetrachloride and 1,4-dioxane are given in Table 2.

Table 2 — Values of the static dielectric constant (ε_0), limiting dielectric constant (ε_∞), effective Kirkwood correlation factor (g^{eff}), density (ρ), molar volume (V_m) and molar polarization of solute (P_2) of different mole fractions of N, N-dimethylformamide (X_2) in benzene, carbon tetrachloride and 1, 4-dioxane.

X_2	ε_0	ε_∞	g^{eff}	P (g/cm^3)	V_m (cm^3/mol)	P_2 (cc)
Benzene						
0.011	2.82	2.2416	1.71	0.867	90.02	680.02
0.015	2.90	2.2392	1.41	0.868	89.98	543.65
0.017	2.94	2.2362	1.34	0.871	89.57	500.35
0.027	3.07	2.2260	1.07	0.874	89.21	379.72
0.030	3.23	2.2186	1.17	0.877	88.89	392.81
0.035	3.34	2.2141	1.12	0.879	88.66	367.27
0.041	3.44	2.2081	1.08	0.882	88.33	342.62
0.048	3.52	2.1992	0.97	0.885	87.99	302.86
0.052	3.77	2.1948	1.08	0.889	87.58	317.66
0.061	3.93	2.1918	1.03	0.891	87.33	294.77
Carbon tetrachloride						
0.010	2.51	2.1094	0.81	1.578	97.20	437.10
0.019	2.73	2.1083	0.99	1.572	97.42	413.02
0.032	2.99	2.0981	0.97	1.564	97.53	360.47
0.042	3.29	2.0943	1.05	1.559	97.57	358.07
0.056	3.37	2.0923	0.86	1.553	97.60	293.46
0.064	3.64	2.0909	0.93	1.549	97.63	297.93
0.073	3.78	2.0822	0.90	1.544	97.69	284.06
0.080	4.16	2.0764	1.02	1.540	97.77	300.93
0.093	4.22	2.0721	0.92	1.533	97.85	270.10
0.111	4.52	2.0692	0.88	1.524	97.94	249.69
1, 4-dioxane						
0.009	2.61	2.0198	1.63	1.021	86.17	598.13
0.017	2.71	2.0215	1.10	1.017	86.40	395.75
0.021	2.74	2.0226	0.96	1.012	86.78	349.44
0.030	2.95	2.0237	0.98	1.007	87.10	335.84
0.039	3.19	2.0249	1.03	1.003	87.34	335.79
0.042	3.30	2.0260	1.04	0.998	87.73	335.25
0.050	3.41	2.0272	0.98	0.992	88.17	313.95
0.056	3.56	2.0283	0.97	0.988	88.45	307.81
0.060	3.63	2.0294	0.95	0.985	88.66	298.85
0.065	3.73	2.0306	0.94	0.980	89.05	294.35

Variation in Kirkwood correlation factor (g^{eff}) with the change in concentration of the N,N-dimethylformamide in non-polar solvents benzene, carbon tetrachloride and 1,4-dioxane is shown in Fig. 1. The g^{eff} value gives an idea about cyclic or linear multimerization of polar molecules in its pure form. However, if the study is conducted for solutions of polar molecules in non-polar solvents, Kirkwood correlation factor (g^{eff}) gives an idea about the solute-solute and solute-solvent associational behaviour of molecules of the polar-non-polar liquid mixtures[4,6]. In present study; the value of Kirkwood correlation factor (g^{eff}) of pure N, N-dimethylformamide is 1.16 and it is in good agreement with the literature value[4]. The g^{eff} values of N,N-dimethylformamide are greater than unity; confirming the formation of H-bonded linear structures with parallel dipole alignments in its pure form[4,22,23]. For dilute solutions of polar molecules in non-polar solvent in the ideal cases; i.e., for solutions of non-interacting spherical polar molecules, the Kirkwood correlation factor (g^{eff}) should be equal 1, independent of the solution concentration. But for solutions of N,N-dimethylformamide in benzene for lower concentration, i.e., below 0.04 mole fraction of N,N-dimethylformamide, the effective Kirkwood correlation factor (g^{eff}) is higher than unity and it decreases non-linearly with concentration, and above this concentration it remains near to unity. This clearly indicates that N,N-dimethylformamide molecules in benzene forms α-multimers at lower concentration and they are converted into β-multimers at higher

concentration[18]. Similarly, it is clear from the Fig. 1 that for the solution of N,N-dimethylformamide in 1,4-dioxane for lower concentration, i.e., below 0.04 mole fraction of N,N-dimethylformamide; g^{eff} is higher than unity and it continuously decreases non-linearly with increase in concentration of N,N-dimethylformamide. Above 0.04 mole fraction of N,N-dimethylformamide; it remains very close to unity and this behaviour clearly indicates that N, N-dimethylformamide molecules in 1,4-dioxane also forms α-multimers at lower concentration and they are converted into β-multimers at higher concentration. In both the series of solutions, the molecules of N,N-dimethylformamide associate to form the multimers with parallel as well as with antiparallel dipole moment[22]. The higher values of Kirkwood correlation factor (g^{eff}), i.e., $g^{\text{eff}}>1$ indicates the hetero and self-association of N,N-dimethylformamide molecules but the lower values of Kirkwood correlation factor (g^{eff}), i.e., $g^{\text{eff}} \leq 1$ suggests only the self-association of N,N-dimethylformamide molecules in benzene and 1,4-dioxane[4,22]. While in case of N,N-dimethylformamide molecules in carbon tetrachloride; the values of Kirkwood correlation factor (g^{eff}) is very close to unity and almost constant. This means that there is no dipolar alignment of N,N-dimethylformamide molecules in carbon tetrachloride. The difference in the Kirkwood correlation factor (g^{eff}) values of N,N-dimethylformamide molecules in carbon tetrachloride than in benzene and 1,4-dioxane solvents are due to large difference in their densities. Higher density of CCl_4 in comparison to the densities of benzene and 1,4-dioxane produces large hindrance and there is decrease in g^{eff} values in bulky solvent like carbon tetrachloride. From Table 2, it can be seen that the values of limiting dielectric constant (ε_∞) decreases in benzene and carbon tetrachloride with increase in concentration of N, N-dimethylformamide while in case of 1,4-dioxane; the values are continuously increasing with increase in concentration of N, N-dimethylformamide.

From Table 2, it can be seen that the molar polarization values decrease with increasing mole fraction of DMF in all the three solvents, which indicates solute-solvent interactions of an unassociated solute DMF molecules with the solvent molecules. The trend of molar polarization of solute (P_2) follows the Kirkwood correlation factor. Trivedi et al.[18] have shown similar type of behaviour in molar polarization of the solute (P_2) and Kirkwood correlation factor (g) for the system of amino
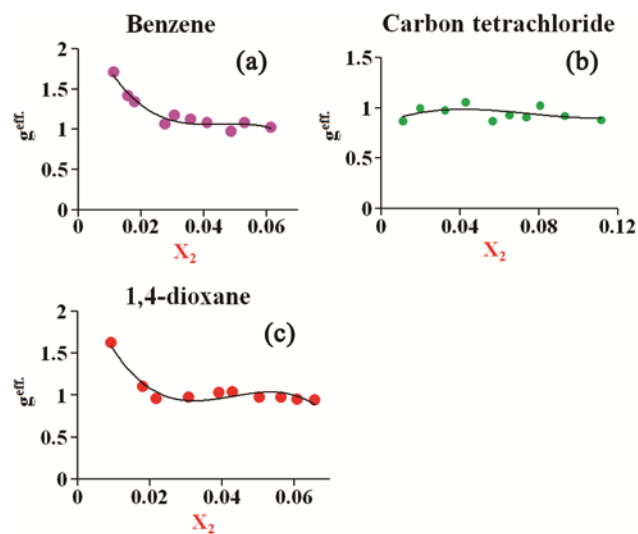

Fig. 1 — The variation in the effective Kirkwood correlation factor (g^{eff}) as a function of the N, N-dimethylformamide mole-fraction (X_2).

substituted pyridines in non-polar solvents benzene, CCl_4 and 1,4-dioxane. The density of the solution of the DMF in benzene increases with increase in mole fraction of DMF, which results in decrease in the molar volume (V_m) of the solution. The decrease in molar volume (V_m) suggests the close packing of the molecules, which indicates the increase in the magnitude of interaction[24]. As compared to benzene;

the density (ρ) and the molar volume (V_m) variations of solutions of DMF in CCl_4 and 1, 4-dioxane exhibits opposite trend with concentration.

Variation in dielectric constant (ε') and dielectric loss (ε'') of solutions of N,N-dimethylformamide in benzene, carbon tetrachloride and 1,4-dioxane over a frequency span of 20 Hz to 2 MHz is shown in Fig. 2(a-e). From these figures, it can be seen that
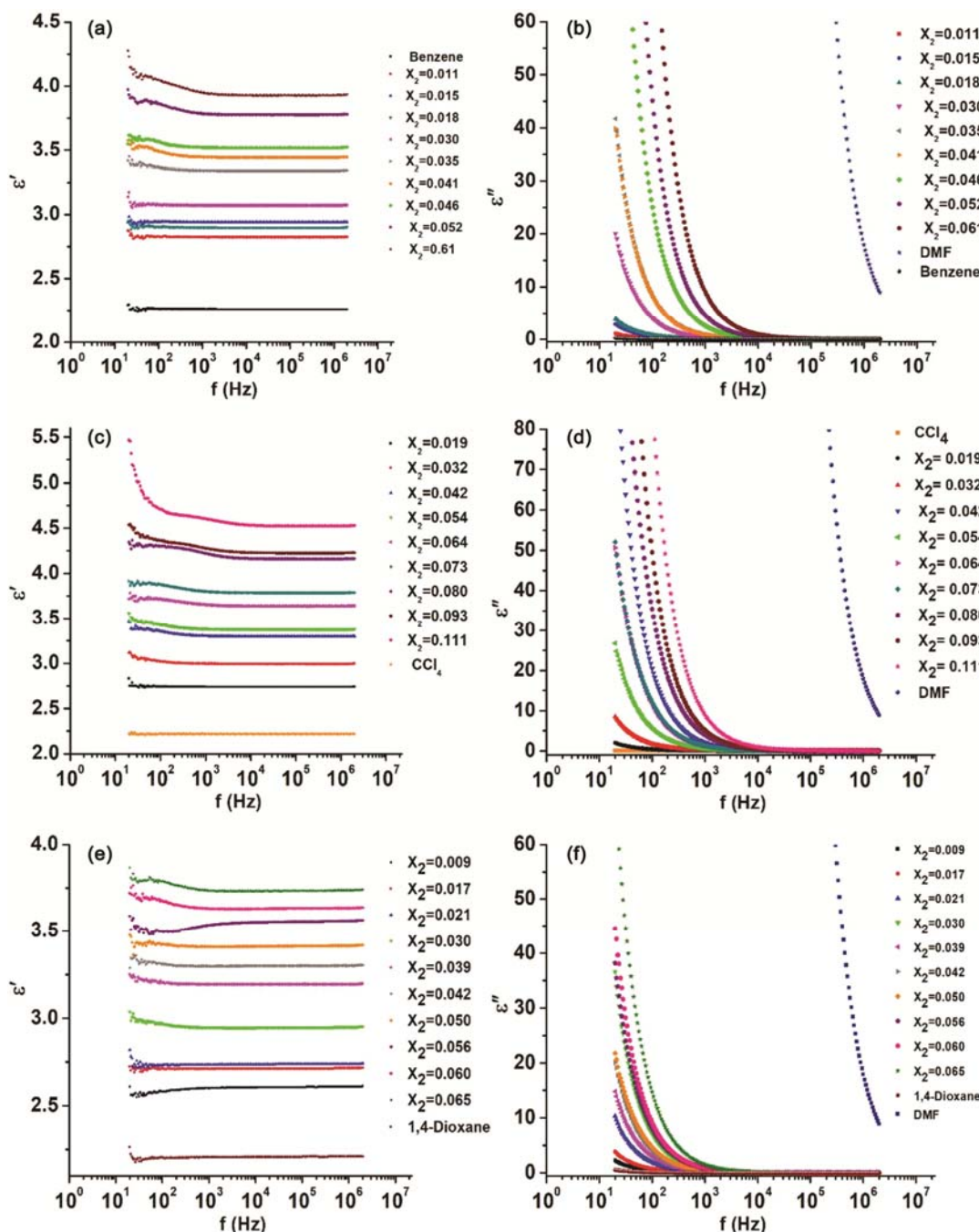

Fig. 2 — (a, c, e) Plots of dielectric constant (ε') versus log f and (b, d, f) dielectric loss (ε'') versus frequency (f) for concentrated solutions of N, N-dimethylformamide in benzene, carbon tetrachloride and 1, 4-dioxane.

dielectric constant (ε') increases with increase in concentration of N,N-dimethylformamide in all the three solvents. For $0 < X_2 < 0.03$; dielectric constant (ε') values remain constant over the entire frequency range. But for $X_2 > 0.03$; dielectric constant (ε') values in the low frequency region increase with decrease in the frequency. The increase in dielectric constant (ε') values with decrease of frequency in low frequency region is certainly due to the electrode polarization (EP) effect. This effect represents the formation of electric double layer (EDL) capacitances at interface of the dielectric material and the metallic electrode surfaces in plane geometry by accumulation of long distance drifted ions and free charges in the solutions of polar liquids in the non-polar solvents[25-28]. The formation of EDLs is an evidence of the presence of ions and free charges in the polar liquid[26,27,29,30]. For this dipolar liquid, the strength of EP effect decreases with increase of frequency which is due to decrement in long range movement of free charges in the liquid dielectric material under the increase of fast reversal changes developed in the applied ac electric field. The frequency value at which the EP effect completely disappears is found to increase with increase in concentration of N,N-dimethylformamide in all the three solvents. From Fig. 2(b,d,f); it can be seen that the dielectric loss (ε'') values of pure solvents are independent of frequency and they are very close to zero. But as N,N-dimethylformamide of varying amount is added into the non-polar solvents; dielectric loss (ε'') values exhibit strong dependence on the frequency in the low frequency region. From the plots; it can be seen that the frequency at which the constant value of loss (ε'') is attained, increases with increase in concentration of N,N-dimethylformamide. Rapid increase in dielectric loss values with decrease in frequency is due to the ionic conductivity effect. The ε'' spectra of mixtures of N,N-dimethylformamide in non-polar solvents decrease non-linearly with the increase of frequency from 20 Hz to 2 MHz, which confirms non-Ohmic type ionic conduction mechanism in these liquid materials. Dielectric loss (ε'') of pure N,N-dimethylformamide increases rapidly with decrease in frequency range over entire frequency range as shown in Fig. 3. This clearly suggests highly polar nature of N, N-dimethylformamide and presence of high amount of free charge contaminants in it.
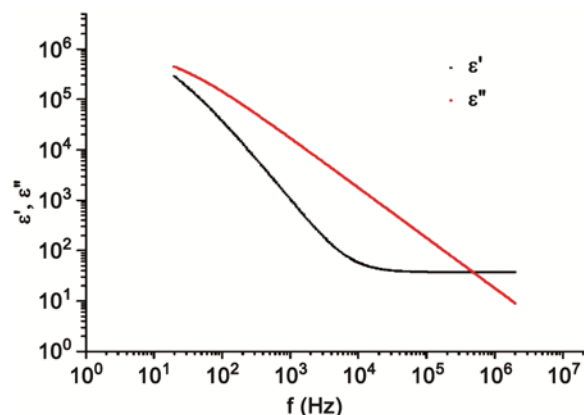

Fig. 3 — Plot of dielectric permittivity (ε') and dielectric loss (ε'') of pure N, N-dimethylformamide versus frequency (f).
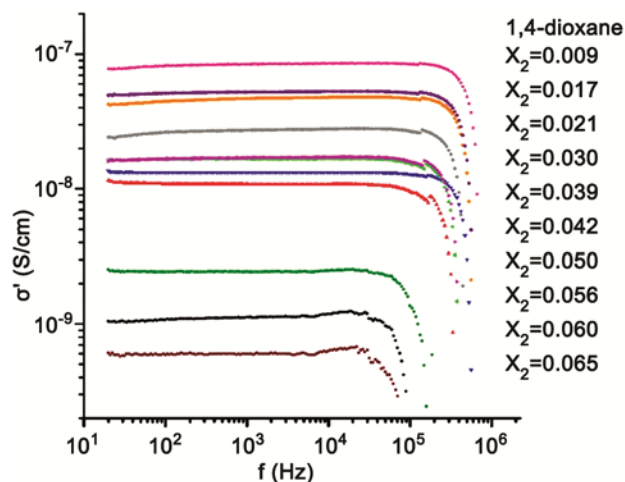

Fig. 4 — Frequency dependent real part (σ') of complex ac conductivity of DMF in 1, 4-dioxane mixtures at various mole fractions of DMF (X_2) at 303.15 K.

Figure 4 shows the frequency dependent real part of complex ac conductivity of DMF in 1, 4-dioxane at various mole fraction of DMF (X_2) at 303.15 K. The similar spectra are observed for solutions of DMF in carbon tetra chloride and benzene. σ' spectra have dc ionic conductivity (σ_{dc}) plateau in low frequency region, and a non-linear decrease with increase of frequency in EP affected high frequency region. From these spectra dc ionic conductivities of all the solutions are determined[27] and they are presented in Table 3. It is observed that the values of dc conductivity vary between 10^{-7} to 10^{-9} S/cm and it continuously increases with increase in the mole fraction of DMF (X_2) in all the three solvents. σ_{dc} of solutions of DMF in all the three non-polar solvents over the entire concentration range is in order of $\sigma_{dc,\text{Carbon tetrachloride}} > \sigma_{dc,\text{benzene}} > \sigma_{dc,\text{1, 4-dioxane}}$.

Table 3 — The values of dc conductivity of different mole fractions of N, N-dimethylformamide (X_2) in benzene, carbon tetrachloride and 1, 4-dioxane.

Benzene		Carbon tetrachloride		1, 4-dioxane	
X_2	σ_{dc}(S/cm)	X_2	σ_{dc}(S/cm)	X_2	σ_{dc}(S/cm)
0.011	1.13×10^{-9}	0.010	1.14×10^{-9}	0.009	1.10×10^{-9}
0.015	1.15×10^{-9}	0.019	2.14×10^{-9}	0.017	2.44×10^{-9}
0.017	3.22×10^{-9}	0.032	9.51×10^{-9}	0.021	1.08×10^{-8}
0.027	3.68×10^{-9}	0.042	1.11×10^{-8}	0.030	1.32×10^{-8}
0.030	4.32×10^{-9}	0.056	2.97×10^{-8}	0.039	1.66×10^{-8}
0.035	2.25×10^{-8}	0.064	6.68×10^{-8}	0.042	1.69×10^{-8}
0.041	4.50×10^{-8}	0.073	7.16×10^{-8}	0.050	2.70×10^{-8}
0.048	4.53×10^{-7}	0.080	2.13×10^{-7}	0.056	4.63×10^{-8}
0.052	1.37×10^{-7}	0.093	2.83×10^{-7}	0.060	5.21×10^{-8}
0.061	2.52×10^{-7}	0.111	4.95×10^{-7}	0.065	8.37×10^{-8}

4 Conclusions

At lower concentration of N, N-dimethylformamide in benzene and 1, 4-dioxane; the N, N-dimethylformamide molecules are found to form α-multimers and they are converted into β-multimers at higher concentration. There is no dipolar alignment of N, N-dimethylformamide molecules in CCl$_4$. Different values of Kirkwood correlation factor of N, N-dimethylformamide in benzene, 1, 4-dioxane and carbon tetrachloride may be attributed to the large difference of the densities of the solvents. The trend of molar polarization of solute (P_2) follows the Kirkwood correlation factor (g^{eff}) for all the mixture range in non-polar solvents. The increase in molar volume (V_m) in benzene suggests the close packing of molecules and increment in the magnitude of interaction. Increase in dielectric constant values (ε′) with decrease in frequency in the lower frequency region for solutions of concentration $X_2 > 0.03$ indicates the formation of EDL (Electric double layer) at the interface of the electrode and the dielectric material. The constant values of dielectric constant (ε′) in the high frequency region indicate that there is no movement of free charges due to variation in the applied ac electric field. The frequency value at which the EP (electrode polarization) effect completely disappears is found to increase with increase in concentration of N, N-dimethylformamide in all the three solvents. σ_{dc} of solutions of DMF in all the three non-polar solvents over the entire concentration range is in order of $\sigma_{dc,\text{Carbon tetrachloride}} > \sigma_{dc,\text{ benzene}} > \sigma_{dc,\text{1, 4-dioxane}}$.

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