Original Research Article

The Dielectric behavior of Acetone and Dimethylformamide in Electric Field

Abstract

The dielectric behavior of both acetone and dimethylformamide were examined in the presence of an electric field. The electrical conductivity of both solvents was used to estimate the complex permittivity of acetone and dimethylformamide. The relaxation time of the solvents at various temperatures was estimated. The experimental static permittivity values together with the estimated complex permittivity, and relaxation time were substituted in the Debye equations to generate the dielectric constant and the corresponding loss factor at various frequencies using maple-13 programme. Result indicates that at frequency 0.1GHz and temperature 10^{o}C, the calculated value for acetone was 22.4637 and that of DMF was 40.3380. This shows that an increase in dielectric constant caused a corresponding decrease in the loss factor, which means an inverse relation between the frequency and temperature. These results compared well with experimental values for both acetone and DMF at the same frequency temperature of 22.21 ± 0.04 and 40.34 ± 0.06 respectively.

Key word: complex permittivity, acetone, dimethylformamide, dielectric, relaxation time

INTRODUCTION

Several studies have been conducted to examine the dielectric behaviour of the mixture of either acetone or dimethylformamide with other solvents or alcohols in the presence of an applied electric field. Notable among them are the dielectric relaxation study of formamide with alcohols [1-2] and the dielectric relaxation study of formamide with some common dipolar aprotic and protic solvents (water, dimethylsulphoxide, N, N- dimethylformamide, acetone, 1, 4-dioxane, mono-, di-, and trihydric alcohols, and homologous series of 2-alkoxyethanol, 2-(2-alkoxyethoxy) ethanol, and ethylene glycol oligomers) binary mixtures at 30^{o}C over the frequency range of 10MHz to 10GHz using time domain reflectometry [3]. However, there are little studies concerning the dielectric relaxation of pure acetone and dimethylformamide. In this work, attempts have been made to study behaviour of pure acetone and dimethylformamide in the presence of an applied electric field using Debye relaxation method.

Whenever a material is exposed to an electric field (as generated by a voltage across electrodes between which the electric is embedded), every different kinds of dipoles become excited even within atomic scales. A local charge imbalance is thus “induced” within the neutral species (atoms or molecules) as the “centers of gravity” for equal amount of positive and negative charges, $\pm q$, become separated by a small distance (d), thus creating a dipole with a dipole moment, $\mu = q.d$, which is related to the “local” or microscopic” electric field (E_{Local}) acting in close vicinity of the species. Thus, the dipole moment can be written as:

$$\mu = \alpha . E_{Local} \qquad (1)$$

Where $\alpha =$ polarizability [Cm^{2}V^{-1}] of the species or material under consideration.

It is necessary to point out that E_{Local} refers to the local field rather than the applied field. This distinction is drawn because the local field will be the vectorial sum of the applied field and the fields generated by the presence of the surrounding charges (i.e. the other dipoles). The question then arises as to how the local may be related to the applied electric field. One of the earliest approaches involves the general relationship between polarization and the applied electric field strength:

$$p = (\varepsilon_r - 1).\varepsilon_o.E_e \qquad (2)$$

Where p = polarization, dipole density [Cm^{-2}], ε_r = relative permittivity and ε_o =electric field constant in vacuum = $8.854 \times 10^{-12}[C^2J^{-1}m^{-1}]$; E_e =external electric field, produced by the applied voltage (Craig, 1995).

The local field was first calculated by Lorentz in 1909 by considering all the electric fields influencing the molecule in the cavity:

$$E_{Local} = E_i + E_e - E_{sph} \qquad (3)$$

E_{Local} = Local electric field

E_i = Internal electric field, caused by interactions with other induced dipole in the sphere

E_e = External electric field, produced by the applied voltage

E_{sph} = Electric field caused by the induced dipoles outside the sphere, causing charges on the surface

In an ideal gas, E_{sph} and E_i are zero. In liquids, neighbouring molecules show a polarizing effect leading to charges on the sphere's boundary, resulting in

$$E_{sph} = -\frac{P}{3.\varepsilon_o} \qquad (4)$$

By combining equations (1), (2), and (3) we obtain for the local field:

$$E_{Local} = E_i + E_e.\left(\frac{\varepsilon+2}{3}\right) \qquad (5)$$

If $E_i = 0, E_{Local}$ is reduced to the Lorentz field(E_L):

$$E_{Local} = E_L = E_e \left(\frac{\varepsilon+2}{3}\right) \qquad (6)$$

According to Clausius and Mossotti we obtain for nonpolar molecules of constant polarizability the following relation:

$$P = N.\mu_i \qquad (7)$$

Where P = polarization, dipole density [Cm^{-2}]; N = number of molecules per volume and μ_i = induced dipole moment

By combining Equations (1) with (7) we obtained

$$P = N.\mu_i = N.\alpha.E_{Local} \qquad (8)$$

Combination of Equations (2), (6) and (8) lead to the Clausius- Mossotti equation for nonpolar molecules (Equation (9)) and (10)

$$\frac{(\varepsilon-1)}{(\varepsilon+2)} = \frac{N.\alpha}{3.\varepsilon_o} \qquad (9)$$

Where $N = \frac{N_A}{V_M} = \frac{N_A.\rho}{M_r}$ is the number of polarizable molecules per unit volume. Therefore, equation (9) can be defined as molar polarizationP_M:

$$P_M = \frac{\varepsilon-1}{\varepsilon+2}.\frac{M_r}{\rho} = \frac{N_A}{3.\varepsilon_o}.\alpha \qquad (10)$$

P_M =molar polarization [m^3mol^{-1}] and N_A = Avogadro's constant = $6.023 \times 10^{23} mol^{-1}$ [4-6].

The Clausius-Mossotti equation was extended by Debye to polar molecules:

$$\frac{\varepsilon-1}{\varepsilon+2}.\frac{M_r}{\rho} = \frac{N_A}{3.\varepsilon_o}.\left(\alpha + \frac{\mu_g^2}{3.k.T}\right) \qquad (11)$$

Where $\varepsilon =$ quasi-static relative dielectric constant; M_r =molecular weight; ρ =density; N_A = Avogadro's constant = $6.023 \times 10^{23} mol^{-1}$; ε_o =electric field constant in the vacuum= $8.854 \times 10^{-12} C^2 J^{-1} m^{-1}$; α =polarizability of the molecule (Cm^2V^{-1}); μ_g =dipole moment in the state of an ideal gas (Cm); k = Boltzmann's constant=$1.38 \times 10^{-23} (JK^{-1})$; T = temperature

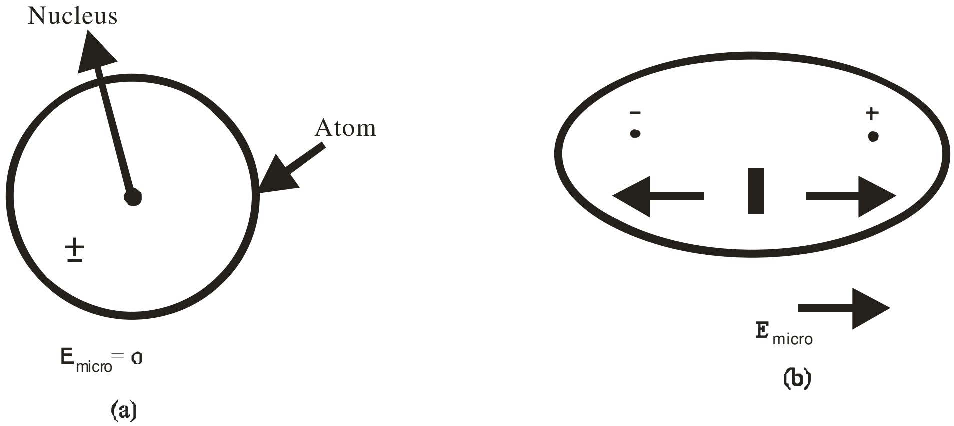
Equation (11) is the Debye equation is valid only for gas under low pressure and highly diluted solutions of polar molecules in nonpolar solvents; dipole-dipole interactions are neglected. Therefore, it is not valid for polar liquids but can be used to estimate quite accurately the dipole moment μ_g of water in highly diluted solution of water in 1,4-dioxane simulating an ideal gas state condition [7].

Polarization Mechanism

Dielectrics are materials which have no free charges; all electrons are bound and associated with the nearest atoms. An external electric field causes a small separation of the centres of the electron cloud and the positive ion core so that each infinitesimal element of volume behaves as electric dipole. The cumulative effect of these microscopic displacement results in a net polarization due to setting of induced dipole moment (electronic and ionic polarization) or due to the rotation of permanent electric dipoles already present in the materials (orientation polarization). Thus when external electric field is applied induced charges appears on the surface of the dielectric.

Electronic Polarization

In non polar dielectrics there is zero dipole moment under normal condition (i.e. when $F = 0$). However, when electric field is applied electronic polarization occurs due to the response of individual atoms in the dielectric to the local electric field (E_{micro} or E_{loc}) at the atom. Thus the centre of negative charge (electron cloud) is displaced in the direction of opposite electric to field and centre of positive charge (nucleus) is displaced in the direction of electric field. Consequently the atoms acquire an induced dipole moment, due to the shift of electron charge cloud of the atom with respect to the positive nucleus under the influence of electric field (fig. a and b).



(a)

(b)

In figure (a), the positive and negative charges coincide (under normal state$E.F. = 0$). *Whereas in figure (b), the induced dipole moment when* $E.F$ *is applied.* $p = q1$. *This induced dipole moment vanishes as soon as electric field is removed.*

Ionic Polarization

When an external electric field is applied to the molecule positive and negative ions are displaced in the opposite direction until the ionic bonding forces stop the process. Such a shift of the ions relative to other oppositely charged neighbours results in an induced dipole moment proportional to the local field at the ions and the corresponding polarization is known as ionic polarization.

Orientation Polarization

This type of polarization occurs only in polar dielectrics. Such dielectrics have permanent dipole moment (i.e. the positive and negative charge centre does not coincide even when no field is applied). In polar dielectrics, under normal conditions (i.e. when $E.F. = 0$) the dipoles are randomly oriented, so that the net dipole moment and thus the polarization is zero.

However, when an external electric field is applied, the field exerts torque on the molecular dipole moments and tends to rotate them in the direction of field. The contribution of this process of orientation of permanent dipoles to the polarization, is called the orientation polarization and is given by

$$P_o = \frac{NP_p^2 E}{3kT} \quad (12)$$

Where P_p is the permanent dipole moment of each molecule and N is the number of molecules per unit volume.

Orientation Polarization $\alpha_o = \frac{P_p^2}{3kT}$ (13)

From equation (12)

$$P_o \alpha \frac{1}{T} \quad (14)$$

Thus orientation polarization P_o is dependent on temperature and decreases with increase in temperature. This is because at higher temperature the thermal energy will increase which will try to disorient the dipoles.

Space Charge Polarization

The fourth type of polarization is called space charge polarization and occurs due to the accumulation of charges at the electrodes or at the interfaces in a multiphase material. The ions diffuse over appreciable distances in response to the applied field, giving rise to a redistribution of charges in the dielectric medium.

Total Polarization

The total polarization of a dielectric material is the sum of the contribution from all the other polarization mechanism

$$P_T = P_e + P_i + P_o + P_s \qquad (15)$$

Where P_T is the total polarization of a multiphase dielectric material

$P_e$is the orientation polarization

$P_i$is ionic polarization

$P_o$is the orientation polarization

$P_s$is the space charge polarization

In case of polar dielectric i.e. dielectric having ionic bonding and permanent dipoles-dielectric possess orientation as well as electronic and ionic polarization mechanism

$$P_T = P_e + P_i + P_o \quad (16)$$

However, in case of ionic and nonpolar dielectric, if the nonpolar dielectric-dielectric possesses both electronic and ionic polarization i.e.

$$P_T = P_e + P_i \qquad (17)$$

But if the non-polar dielectric have no ionic bonding i.e. elemental solid dielectric (ESD)- dielectric exhibit only electronic polarization i.e.

$$P_T = P_e \qquad (18)$$

Factors Affecting the Dielectric Constant of Materials

Effect of the Frequency on the Dielectric Constant

The frequency-dependent trend of the dielectric properties can provide useful information about the material characteristics. In theory, electric conduction and various polarization mechanisms (including dipole, ionic, and Maxwell-Wagner) contribute to the dielectric loss factor [8-9]. For moist dielectric materials, ionic conductivity plays a major role at lower frequencies $(e.g < 200MHz)$, whereas both ionic conductivity and dipole rotation of free water play a combined role at microwave frequencies.

Effect of the Temperature on the Dielectric Constant

Temperature of a material has a significant effect on the dielectric properties. Generally, the loss factor increases with increasing temperature at low frequencies due to ionic conductance [10] and decreases with increasing temperature at high frequencies due to free water dispersion [11].

METHOD

The electrical conductivity σ of both acetone and dimethylformamideat various temperatures were adapted in [12-13]. The unit was then converted to s/m as shown in tables 1 and 2 below.

Table1: the electrical conductivity of Acetone at various temperatures

Temperature(°C)	**Electric conductivity (σ) 10^{-3}s/m**
10	4.34
20	3.39
30	2.34
40	1.78
50	0.83

Table2: the electrical conductivity of Dimethylformamide (DMF) at various temperatures

Temperature(°C)	**Electric conductivity (σ) 10^{-3}s/m**
10	1.45
20	1.34
30	1.11
40	0.95
50	0.81

To obtain the complex permittivity the electrical conductivity was then substituted in equation (19) and the loss factor was estimated

$$\varepsilon'' = \frac{\sigma}{\omega \varepsilon_o} \qquad (19)$$

σ = Electrical conductivity of the material
ω = Angular frequency
ε_o = Permittivity of free space = $8.854 \times 10^{-12} F/m$

The complex permittivity for acetone at 10°C calculated as shown below:

$$\varepsilon' = \varepsilon_\infty + \frac{1}{\tau}\left(\frac{\varepsilon''}{\omega}\right) = 22.21 = \varepsilon_\infty + \frac{1}{9.22 \times 10^{-11}}\left(\frac{0.78}{6.284 \times 10^{8}}\right)$$

$$\varepsilon_\infty = 22.21 - 13.52 = 8.69$$

And that of dimethylformamide at 20^{o}C was also calculated as follows:

$$\varepsilon' = \varepsilon_\infty + \frac{1}{\tau}\left(\frac{\varepsilon''}{\omega}\right) = 38.45 = \varepsilon_\infty + \frac{1}{1.0747 \times 10^{-11}}\left(\frac{0.24}{6.284 \times 10^8}\right)$$

$$\varepsilon_\infty = 38.45 - 35.54 = 2.91$$

The relaxation time τ derived by Bloembergan et al (1948) shown in equation (20) for the magnetic relaxation in terms of the correlation time τ_c, which was found to be closely related to Debye's theory of dielectric dispersion in polar liquids was used to calculate the relaxation time for both acetone and DMF in this work.

$$\tau = \frac{4\pi\eta a^3}{kT} \qquad (20)$$

η = Viscosity of the solvent

a = Radius of solute molecule

k = Boltzmann constant

T = The temperature

The relaxation time of acetoneτ at 10^{o}C is shown below:

$$\tau = \frac{4\pi\eta a^3}{kT} = \frac{4 \times 3.242 \times 4.25 \times 10^{13} \times (0.62)^3}{1.38 \times 10^{23} \times 10}$$

$$= 9.22 \times 10^{-11} s$$

The relaxation time at 20^{o}C for DMF is shown below:

$$\tau = \frac{4\pi\eta a^3}{kT} = \frac{4 \times 3.242 \times 4.72 \times 10^{11} \times (1.71)^3}{1.38 \times 10^{23} \times 20}$$

$$= 1.0747 \times 10^{-11} s$$

The solute radius a (effective Born radius) of DMF and acetone was adapted in [14-15]. The Viscosity of acetone and DMF was calculated using the expression below:

$$\eta = V_o e^{\epsilon_a}/_{RT} \qquad (21)$$

V_o = Absolute viscosity

ϵ_a = Activation energy

R = Universal gas constant

For example the viscosity of acetone is given by

$$\eta = V_o e^{\epsilon_a}/_{RT} = 0.000316 \times e^{9.28 \times 10^4}/_{8.314 \times 283}$$

$$= 4.25 \times 10^{13}$$

The static and complex permittivities obtained from our calculations are shown in tables (3) and (4) below:

Table 3: Parameters for Acetone

Temperature °C	$\boldsymbol{\varepsilon_s}$	$\boldsymbol{\varepsilon_\infty}$	$\boldsymbol{\tau \times 10^{-11} S}$
10	22.25	8.69	9.22
20	21.13	4.55	4.05
30	20.20	3.34	3.12
40	18.83	2.70	2.07
50	17.63	1.32	1.43

Table 4: Parameters for dimethylformamide (DMF)

Temperature °C	$\boldsymbol{\varepsilon_s}$	$\boldsymbol{\varepsilon_\infty}$	$\boldsymbol{\tau \times 10^{-11} S}$
10	40.34	3.51	1.117
20	38.45	2.91	1.075
30	36.69	3.10	0.93
40	35.35	2.98	0.85
50	33.36	3.00	0.76

The values of $\boldsymbol{\varepsilon}_\infty$ and $\boldsymbol{\varepsilon}_s$ as shown in the tables 3 and 4 above were substituted in the real and imaginary part of Debye equations shown below:

$$\varepsilon' = \varepsilon_\infty + \frac{(\varepsilon_S - \varepsilon_\infty)}{(1+\omega^2\tau^2)} \tag{22}$$

$$\varepsilon'' = \frac{(\varepsilon_S - \varepsilon_\infty)\omega\tau}{(1+\omega^2\tau^2)} \tag{23}$$

Equations (22) and (23) are the real and imaginary part of Debye equation. Detail of the derivation of equations (22) and (23) are shown in [16].

RESULTS AND DISCUSSION

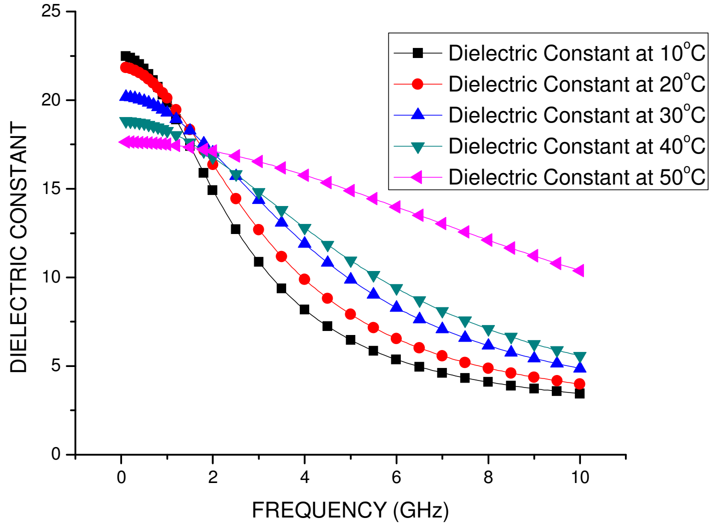
The dielectric constant and loss factor of acetone and dimethylsulphoxide (DMF) were computed within the temperature range of 10°C to 50°C and frequency range $0.1 \leq f \leq 10GHz$. The results of our computation are interpreted based on the existing theories.

A. Dielectric constant

Effect of Frequency at constant Temperature

The effect of frequency on the dielectric constant and its variation as a function of temperature at different frequencies for Acetone and Dimethylformamide are shown graphically below:

Fig1. Dielectric constant Vs Frequency for Acetone



The dielectric constant of Acetone is higher at low frequencies and decreases along the temperature line (i.e. 22.48, 21.84, 20.19, 18.82 and 17.63 for temperatures 10^{o}C, 20^{o}C, 30^{o}C, 40^{o}C and 50^{o}C respectively). However, at higher frequency like $f = 10.0GHz$ the reverse is the case, the dielectric constant increase steadily from 3.44 at 10^{o}C to 10.38 at 50^{o}C (see figure1 above).

Fig2. Dielectric constant Vs Frequency for Dimethylformamide (DMF)

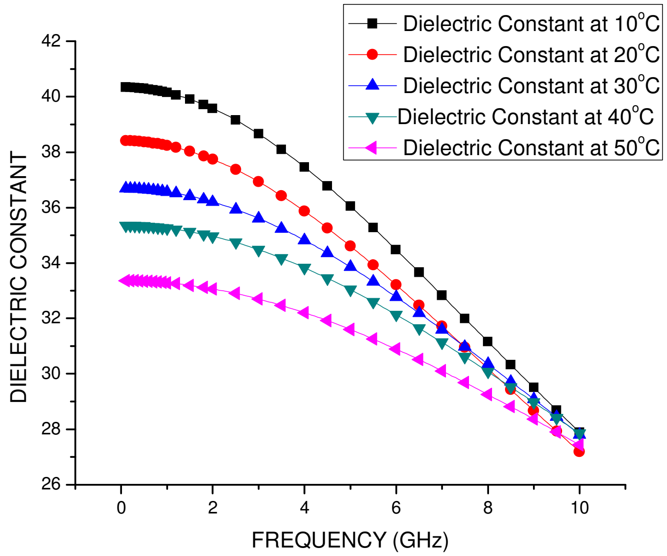


Figure 2 showed the behaviour of dielectric constant of DMF against the frequency. The dielectric constant of DMF also is higher at low frequencies (i.e.

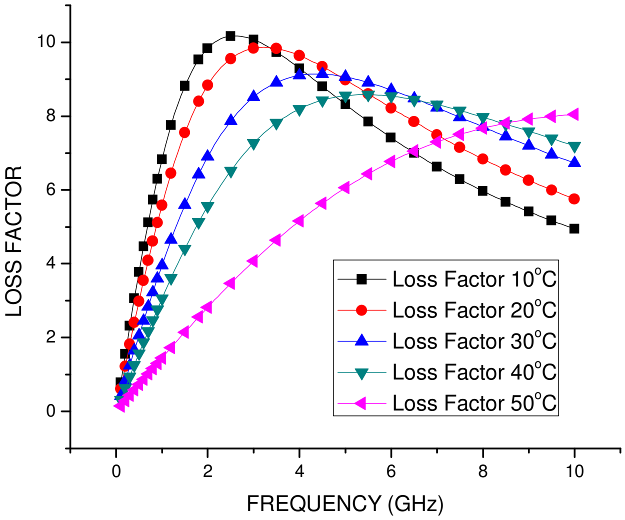
40.34, 38.41, 36.69, 35.35, and 33.36 for temperatures 10^{o}C, 20^{o}C, 30^{o}C, 40^{o}C and 50^{o}C respectively). The dielectric constant of DMF at higher frequencies is also higher at 10^{o}C (i.e. 27.89) but decreases to 27.44 at 50^{o}C unlike the case in Acetone.

B. The loss factor

Effect of frequency

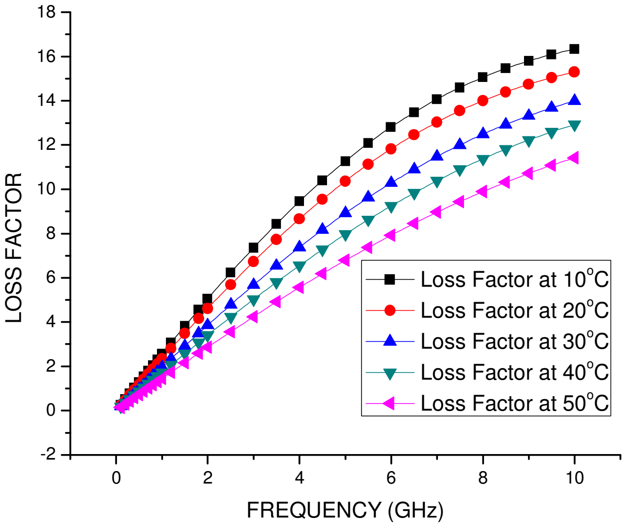
The effect of temperature on loss factor as a function of frequency is shown in figures 3 and 4 below. The loss factor values decreases attaining minimal and then increases sharply. The loss factor of both acetone and DMF are smaller at low frequency and increased sharply attaining its maximum. This value then decreased almost constant for the solvents as shown below:

Fig3. Loss factor Vs Frequency for Acetone



The loss factor of Acetone is small at low frequency (i.e. 0.78, 0.61, 0.42, 0.32 and 0.15) but as the frequency increases the loss factor also increases to a maximum value of 10.17, 9.84, 9.14, 8.58 and 8.05 respectively. The value however decreased after attaining its maximum (see figure 3).

Fig4. Loss factor Vs Frequency for Dimethylformamide (DMF)



DMF has the smallest value of loss factor (i.e. 0.23, 0.24, 0.20, 0.17, and 0.15) at low frequency as compared with acetone but as the frequency increases, the loss factor increases to 16.33, 15.30, 13.99, 12.93 and 11.42 respectively.

DISCUSSIONS

Figure1 and 2 revealed that the dielectric constant of both acetone and DMF is the function of the frequency, temperature, and loss factor. This is because at low frequency and temperature the value of dielectric constant is higher and as the frequency and temperature increased the dielectric constant of both solvents decreases. The results also showed that an increase in dielectric constant caused a corresponding decrease in the loss factor.

The higher value of the dielectric constant ε' at low frequencies as observed in figure1 and 2 above may be due to the overall conductivity which consists of different conduction mechanisms. The most prevalent one in moist materials is the ionic conductivity which varies inversely proportional to the frequency. The decrease in the dielectric constant as frequency increases for all the temperatures may be due to the fact that the dipoles cannot follow up the rapid variation of the applied field [17].

As the temperature increased beyond 10°C the dielectric constant decreases especially at the frequency range of $0.1 \leq f \leq 9GHz$. This decreased in the dielectric constant as the result of increased in the temperature may be due to the relaxation time which has been found to be fast at high temperature and increases dramatically at low temperatures, suggesting a freezing of electric dipoles at low temperature [18] and because of orientation polarization which depend on temperature and decreases with increase in temperature. This is because at higher temperature the thermal energy will increase which will try to disorient the dipoles. The only exception to this is DMF that have shown a sudden increase as the temperature increased beyond 10°C at frequency$10GHz$. The increase in the dielectric constant as the result of an increase in the frequency especially at 10GHz may be due to the permanent electric dipoles possess by DMF [17]

The loss factor on the other hand has a small value at frequency 0.1GHz and increases to reach its maximum value as the frequency increases. It then decreases steadily after attaining the maximum value (see figures 3 and 4 above). This behaviour exhibited by the loss factor may be due to the contribution from all the four types of polarization (i.e. space charge, dipole, ionic and electronic) (Agilent, 2006). But as temperature increases the loss factor decreases especially at frequency 0.1GHz. This behaviour exhibited by both acetone and DMF is contrary to the statement made by Guan et al [10] which said “Generally, the loss factor increases with increasing temperature at low frequencies due to ionic conductance and decreases with increasing temperature at high frequencies due to free water dispersion [11]”. However, at higher frequency like 10GHz the loss factor of acetone increased as the temperature increase.

Salman, Khalil, and Hazaa [19] observed that at higher frequencies, only the ionic and electronic polarizations contribute. The decrease in dielectric constant as the result of increase in the frequency means that, the response of the permanent dipoles decreases as the frequency increases and the contribution of the charge carriers (ions) towards the dielectric constant decreases [20-22].

CONCLUSION

The Debye equation and its derivatives have been used to compute the dielectric constant and loss factor of acetone and dimethylformamide. The computation was done within the frequency range of $0.1 \leq f \leq 10GHz$. The results from our computation revealed that both acetone and dimethylformamide have its maximum dielectric constant at $f = 0.1GHz$ and temperature 10^{o}C.

The higher value of dielectric constant of both acetone and dimethylformamide at $f = 0.1GHz$ and temperature 10^{o}C is because the two solvents are easily polarizable.

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