Original Research Article

Frequency and Temperature Effect on Dielectric properties of Acetone and Dimethylformamide

Abstract

The dielectric behavior of both acetone and dimethylformamide**(DMF)** 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 for the dielectric constant indicates that at frequency 0.1GHz and temperature 10oC, 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. **The result of calculations of the dielectric constant also compared well with the experimental values for both acetone and DMF at the same frequency temperature of $\mathbf{22.21 \pm 0.04}$ and $\mathbf{40.34 \pm 0.06}$ 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°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 pure dimethylformamide those used in industries for the production of nail polish remover, cleaning of organic chemistry laboratories, fibers, films and surface coatings.** 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 α = polarizability [Cm^2V^{-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 = $8.854 \times 10^{-12}[C^2 J^{-1} m^{-1}]$; E_e =external electric field, produced by the applied voltage **[4].**

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**(1879)** and Mossotti**(1847)** 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 $\mu_i =$ 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 $N = \frac{N_A}{V_M} = \frac{N_A \cdot \rho}{M_r}$ 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 = $6.023 \times 10^{23} mol^{-1}$ [5-7].

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 [8].

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 [9-10]. 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 [11] and decreases with increasing temperature at high frequencies due to free water dispersion [12].

METHOD

The dielectric constant and loss factor of both acetone and dimethylformamidewere calculated theoretically. To do this, the researchers adapted the electrical conductivity σ of both acetone and dimethylformamideat various temperatures in [13-14]. The unit was then converted to $\mathbf{S/m}$ as shown in tables 1 and 2 below.

Table1: **The**electrical conductivity of Acetone at various temperatures

Temperature(°C)	Electric conductivity ($\boldsymbol{\sigma}$) $\mathbf{10^{-3}}\mathrm{S/m}$
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 ($\boldsymbol{\sigma}$) $\mathbf{10^{-3}}\mathrm{S/m}$
10	1.45
20	1.34
30	1.11
40	0.95
50	0.81

The static permittivity ε_s of acetone used in this work was adapted in [15] and that of dimethylformamide was adapted in [16-17]. These values were substitute in this expression $\varepsilon' = \varepsilon_\infty + \frac{1}{\tau}\left(\frac{\varepsilon''}{\omega}\right)$ to obtain the complex permittivity of both acetone and dimethylformamide used in this work as shown below:

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

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°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$^{\mathrm{o}}$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$^{\mathrm{o}}$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 [18-19]. 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	ε_s	ε_∞	$\tau \times 10^{-11} S$
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	ε_s	ε_∞	$\tau \times 10^{-11} S$
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 Debye equations shown below:

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

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

Detail of the derivation of equations (22) and (23) are shown in [20].**The theoretical values of both dielectric constant ε' and the loss factor ε'' were generated using maple-13. The angular frequencies were then varied from $0.1 \leq f \leq 10GHz$ during our calculations.**

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

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:

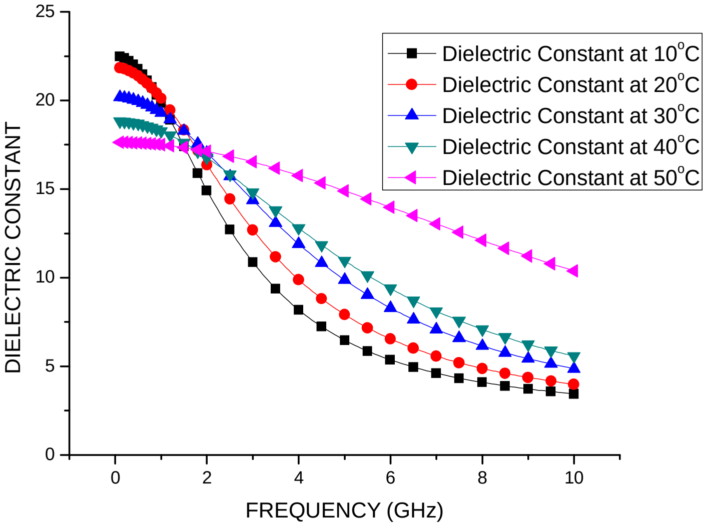

Fig1. Dielectric constant Vs Frequency for Acetone

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).

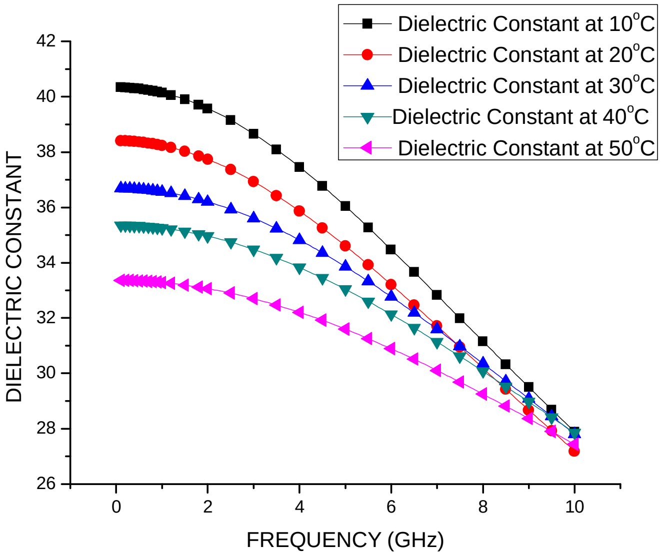

Fig2. Dielectric constant Vs Frequency for Dimethylformamide (DMF)

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°C, 20°C, 30°C, 40°C and 50°C respectively). The dielectric constant of DMF at higher frequencies is also higher at 10°C (i.e. 27.89) but decreases to 27.44 at 50°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:

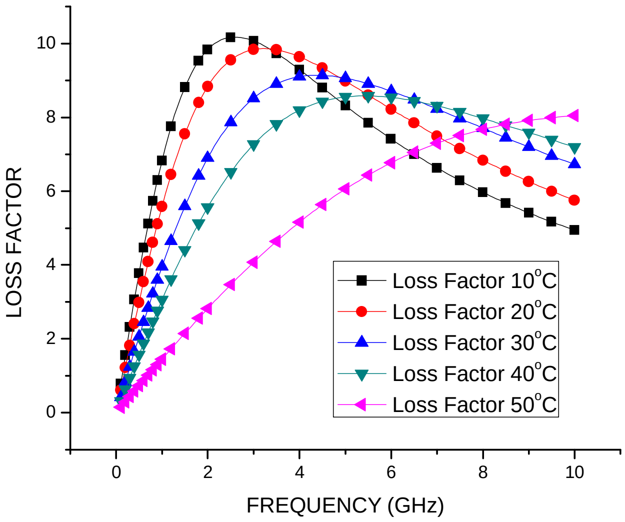

Fig3. Loss factor Vs Frequency for Acetone

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).

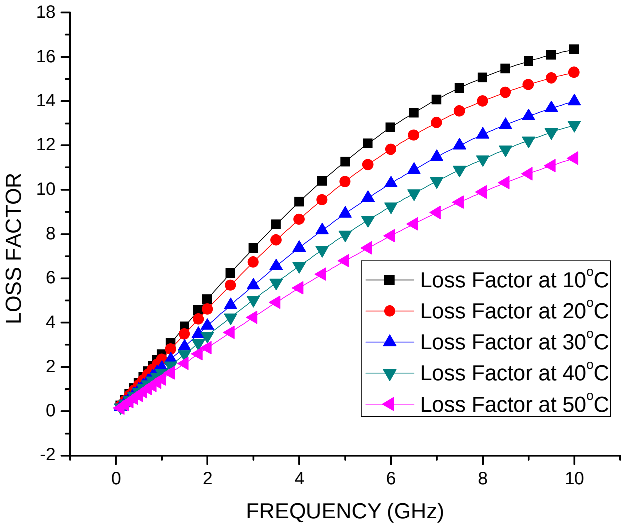

Fig4. Loss factor Vs Frequency for Dimethylformamide (DMF

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 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 [21].

As the temperature increased beyond 10^{o}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 [22] 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^{o}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 [21]

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) [23]. 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 [11] 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 [12]". However, at higher frequency like 10GHz the loss factor of acetone increased as the temperature increase.

Salman, Khalil, and Hazaa [24] 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 [25-27].

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 dielectric constant however, decrease as the both the frequency and temperature increase beyond 0.1GHz and 10oC respectively. For instance, the dielectric constant of acetone at frequency 0.1GHz is 22.4637 and decreased to 3.4427 when the frequency increased to 10GHz and that of dimethylformamide is 40.3380 and has decreased to 27.8893 at frequency 10GHz. Similarly, as the temperature increased from 10oC to 50oC, the dielectric constant of acetone decreased from 22.4637 to 17.6287 and that of the dimethylformamide also decreased from 40.3380 to 33.3592 (see figures 1 and 2 above).**

The loss factor on the other hand has the least value at lower frequencies but increase to reach its maximum as the frequency increases. The loss factor then decreases after attaining its maximum at further increase in the frequency (see figures 3 and 4 above). The loss factor also decrease as the temperature increases from 10^{o}C to 50^{o}C especially within the frequency of $0.1 \leq f \leq 5GHz$ but beyond these frequencies the loss factor increases with increases in the temperatures (see figure 3). This behaviour was only observed in acetone but not dimethylformamide.

The higher value of dielectric constant of both acetone and dimethylformamide at $f = 0.1GHz$ and temperature 10oC suggested that the two solvents are easily polarizable at lower frequencies and temperatures. The result of calculations followed Debye relaxation method.

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