



Optimization in the Computation of Dielectric Constant of Methanol Using Debye Relaxation Method

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Authors' contributions

This work was carried out in collaboration between all authors. Author TJI designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors MYO and SGA managed the analyses of the study. Author ZJK managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

The Debye relaxation equation and its derivatives were used to analyze dielectric constant and loss factor of pure methanol in megahertz and gigahertz over the temperature range of 10°C to 50°C. This was done in order to ascertain which of the frequency ranges has the highest dielectric constant and least loss factor. The graphs of dielectric constant and loss factor were plotted against the frequency. The results were also used to plot Cole-Cole diagrams. The results revealed that the dielectric constant of methanol is higher at frequency 0.05 GHz and decrease as both frequency and temperature increases. However, as the frequency increases beyond 1.0 GHz the dielectric constant increase as the temperature increases. The results also showed that methanol actually formed a semi-circle which suggested that the solvent indeed exhibits Debye relaxation model.

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There were some discrepancies observed in the Cole-Cole plots which may just be attributed to the distribution of the relaxation times in the methanol. The researchers conclude that dielectric constant of methanol is higher in megahertz than those in gigahertz. The loss factor on the other hand was small at lower frequencies but as the frequency increases the loss factor also increases to a maximum value. The loss factor however decreased when the frequency increases after attaining its maximum.

Keywords: Debye relaxation; dielectric constant; loss factor; megahertz gigahertz; Cole-Cole plot.

1. INTRODUCTION

Methanol is one of the most widely traded chemical commodities in the world today. The estimated global demand for methanol also known as methyl alcohol, wood alcohol, wood naphtha, methyl hydrate, or wood spirit is around 27 to 29 million metric tons [1-2]. Recently, the production capacity of methanol has increased considerably. The coming on stream of new plants in South- America, China and the Middle East is a clear cut example.

Methanol was first produced cheaply as a byproduct of the destructive distillation of wood but today industrial methanol is produced in a catalytic process directly from carbon monoxide, carbon dioxide and hydrogen [3-4]. It is a light, volatile, colourless, flammable liquid with a distinctive odour very similar to that of ethanol [5]. However, unlike ethanol, methanol is highly toxic and unfit for consumption. Methanol is a traditional denaturant for ethanol, the product known as denatured alcohol or methylated spirit. This was commonly used during the prohibition to discourage consumption of bootlegged liquor, and ended up causing several deaths [6-7].

Organic methanol, produced from wood or other organic materials (bioalcohol), has been suggested as a renewable alternative to petroleum-based hydrocarbons [8,1]. Low levels of methanol can be used in existing vehicles with the addition of co-solvents and corrosion inhibitors [9]. Methanol fuel has been proposed for ground transportation. The chief advantage of a methanol economy is that it could be adapted to gasoline internal combustion engines with minimum modification to the engines and to the infrastructure that delivers and stores liquid fuel [10-11].

The solvent is primarily used as feedstock for the manufacture of chemicals and as a fuel for specialized vehicles. At about 40% of methanol is converted to formaldehyde and from there into products as diverse as plastics, plywood, paints,

explosives, and permanent textiles [1]. Other chemical derivatives of methanol include dimethyl ether (DME), which has replaced chlorofluorocarbons as an aerosol spray propellant, and acetic acid. Dimethyl ether can be blended with liquefied petroleum gas (LPG) for home heating and cooking, and can be used as a replacement for transportation diesel fuel [12]. It is because of the increasing demand and the market value of methanol that the researchers have decided to optimize the dielectric constant of methanol. This was done in order to ascertain which frequency value and temperature one can obtain the highest dielectric constant.

2. MATHEMATICAL DERIVATION OF THE DEBYE EQUATIONS

Consider a capacitor that consists of two plane parallel electrodes in vacuum having an applied alternating voltage represented by the equation;

$$V = V_m \cos \omega t \quad (1)$$

Where V = instantaneous voltage, V_m = maximum voltage, ω = angular frequency in radians per second = $2\pi f$. The current through the capacitor, i_1 is given by

$$i_1 = I_m (\cos \omega t + \pi/2) \quad (2a)$$

$$\text{Where } I_m = \frac{V_m}{Z} = \omega C_0 V_m \quad (2b)$$

C_0 = vacuum capacitance, sometimes referred to as geometric capacitance.

In an ideal dielectric, the current leads the voltage by 90° or $\pi/2$ rads. There is no component of the current in phase with the voltage. If a material of dielectric constant ϵ is now placed between the plates, the capacitance increases to $C_0\epsilon$ and the current is given by

$$i_2 = I_m \cos [\omega t + (\pi/2 - \delta)] \quad (3a)$$

where $\delta = \tan^{-1} \frac{\epsilon''}{\epsilon'}$ (3b)

$\delta =$ loss angle

The usual symbol for the dielectric constant is ϵ_r , but the subscripts was omitted for the sake of clarity, noting that ϵ is dimensionless. The current phasor will now, not be in phase with the voltage but an angle $(90^\circ - \delta)$. The dielectric constant is a complex quantity represented by

$$\epsilon^* = \epsilon' - j\epsilon'' \quad (4)$$

The current can be resolved into two components; the component in phase with the applied voltage is $I_x = V\omega\epsilon''C_0$ and the component leading the applied voltage by 90° is $I_y = V\omega\epsilon'C_0$. This component is the charging current of the ideal capacitor.

The component in phase with the applied voltage gives rise to dielectric loss. The loss angle δ is given by

$$\delta = \tan^{-1} \frac{\epsilon''}{\epsilon'} \quad (5a)$$

Note that $I_x = V\omega C_0 \epsilon''$, but since $C_0 = \frac{\epsilon_0 A}{d}$

$$I_x = V\omega\epsilon'' \frac{\epsilon_0 A}{d} = \frac{V\omega\epsilon_0 \epsilon'' A}{d} = A\omega\epsilon_0 \epsilon'' E \quad (5b)$$

Where $E = \frac{V}{d}$

$$J_x = \frac{I_x}{A} = \frac{A\omega\epsilon_0 \epsilon''}{A} \therefore J_x = \omega\epsilon_0 \epsilon'' E.$$

Note: ϵ' = the real part of the complex dielectric constant

ϵ'' = The imaginary part of the complex dielectric constant

J_x = The current density

The alternating current conductivity is given by

$$\sigma_{ac} = \sigma' + j\sigma'' = \omega\epsilon_0 [\epsilon'' + j(\epsilon' - \epsilon_\infty)] \quad (6)$$

The total conductivity is given by

$$\sigma_T = \sigma_{ac} + \sigma_{dc} = \omega\epsilon_0 \epsilon'' + \sigma_{dc} \quad (7)$$

σ_{dc} is the direct current conductivity

When a direct voltage applied to a dielectric for a sufficiently long duration is suddenly removed, the decay of polarization to zero value is not

instantaneous but takes a finite time. This is the time required for the dipoles to revert to a random distribution, in equilibrium with the temperature of the medium, from a field oriented alignment. Similarly, the build-up of polarization following the sudden application of a direct current takes a finite time interval before the polarization attains its maximum value. This phenomenon is described by the general term "dielectric relaxation".

When a dc voltage is applied to a polar dielectric, let us assume that the polarization builds up from zero to a final value according to an exponential law such that

$$P(t) = P_\infty (1 - e^{-\frac{t}{\tau}}) \quad (8)$$

Where $P(t)$ is the polarization at a time, t and τ is called the relaxation time. τ is a function of temperature and it is independent of the time.

$$\text{If } \omega = 2\pi f, \quad \omega\tau = 2\pi f\tau$$

Differentiating equation (8) with respect to t , yields

$$\frac{dP(t)}{dt} = \frac{P_\infty e^{-t/\tau}}{\tau} \quad (9)$$

Expanding the right hand side of (8) yields

$$P_\infty e^{-t/\tau} = P_\infty - P(t) \quad (10)$$

Which upon dividing both sides by P_∞ yields

$$e^{-t/\tau} = \frac{P_\infty - P(t)}{P_\infty}$$

Substitute equation (10) into (9) we obtained

$$\frac{dP(t)}{dt} = \frac{P_\infty - P(t)}{\tau} \quad (11)$$

This by assuming that the polarization is due to the dipoles yields

$$\frac{dP(t)}{dt} = \frac{P_\infty - P(t)}{\tau} \cong \frac{P_\mu - P(t)}{\tau} \quad (12)$$

Neglecting atomic polarization, the total polarization $P_T(t)$ can be expressed as the sum of the orientation polarization at that instant $P_\mu(t)$ and electronic polarization, P_e which is assumed to attain its final value instantaneously because the time required for it to attain saturation value is in the optical frequency range. Further, it is

assumed that the instantaneous polarization of the material in an alternating voltage is equal to that under dc voltage that has the same magnitude as the alternating voltage at that instant.

We can express the total polarization, $P_T(t)$ as

$$P_T(t) = P_\mu(t) + P_e \quad (13)$$

The final value attained by the total polarization is

$$P_T = \epsilon_0(\epsilon_s - 1)E \quad (14)$$

Under steady voltages;

$$P_e = \epsilon_0(\epsilon_\infty - 1)E \quad (15)$$

where ϵ_s and ϵ_∞ are the dielectric constants under direct voltage and at infinity frequency respectively. Also note that Maxwell's relation holds

$$\epsilon_\infty = n^2$$

Substituting (14) and (15) into (13) to have

$$P_\mu = \epsilon_0 E(\epsilon_s - \epsilon_\infty) \quad (16)$$

Representing the alternating electric field as

$$E = E_{max} e^{j\omega t} \quad (17)$$

Substituting (16) into (12), we have

$$\frac{dP(t)}{dt} = \frac{\epsilon_0(\epsilon_s - \epsilon_\infty)E - P(t)}{\tau} \quad (18)$$

Substituting (17) into (18); yields

$$\frac{dP(t)}{dt} = \frac{1}{\tau} [\epsilon_0(\epsilon_s - \epsilon_\infty)E_{max} e^{j\omega t} - P(t)] \quad (19)$$

By using the idea of solving linear equations by the use of integrating factors,

$$P(t) = \frac{\epsilon_0(\epsilon_s - \epsilon_\infty)E_m e^{j\omega t}}{1 + j\omega\tau} + C e^{-t/\tau} \text{ or}$$

$$P(t) = C e^{-t/\tau} + \epsilon_0 \frac{(\epsilon_s - \epsilon_\infty)E_m e^{j\omega t}}{1 + j\omega\tau} \quad (20)$$

Where C, is a constant. At time, t sufficiently large when compared with τ , the first term on the right hand side of equation (20) becomes so small that it can be neglected, so the solution for P (t) becomes

$$P(t) = \epsilon_0 \frac{(\epsilon_s - \epsilon_\infty)E_m e^{j\omega t}}{1 + j\omega\tau} \quad (21)$$

Substituting (15), (17) and (20) into (14) and let

$$P(t) = P_\mu(t) = \epsilon_0 \frac{(\epsilon_s - \epsilon_\infty)E_m e^{j\omega t}}{1 + j\omega\tau};$$

We have upon simplification

$$P(t) = \left[\epsilon_\infty - 1 + \frac{(\epsilon_s - \epsilon_\infty)}{1 + j\omega\tau} \right] \epsilon_0 E_m e^{j\omega t} \quad (22)$$

Equation (22) shows that P (t) is a sinusoidal function with the same frequency as the applied voltage. The instantaneous value of flux density, D is given by

$$D(t) = \epsilon_0 \epsilon^* E_m e^{j\omega t} \quad (23)$$

But flux density is also equal to

$$D(t) = \epsilon_0 E_m e^{j\omega t} + P(t) \quad (24)$$

Equate equation (23) to (24);

$$\epsilon_0 \epsilon^* E_m e^{j\omega t} = \epsilon_0 E_m e^{j\omega t} + P(t) \quad (25)$$

Substitute (24) into (25) and simplify

$$\epsilon_0 \epsilon^* E_m e^{j\omega t} = \epsilon_0 E_m e^{j\omega t} + \left[\epsilon_\infty - 1 + \frac{(\epsilon_s - \epsilon_\infty)}{1 + j\omega\tau} \right] \epsilon_0 E_m e^{j\omega t}$$

Dividing each term by $\epsilon_0 E_m e^{j\omega t}$, the last equation reduces to

$$\epsilon^* = 1 + \left[\epsilon_\infty - 1 + \frac{(\epsilon_s - \epsilon_\infty)}{1 + j\omega\tau} \right] \quad (26)$$

Substitute equation (4) into (26)

$$\epsilon' - j\epsilon'' = 1 + \left[\epsilon_\infty - 1 + \frac{(\epsilon_s - \epsilon_\infty)}{1 + j\omega\tau} \right] \quad (27)$$

Simplifying further produces

$$\epsilon' = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + j\omega\tau} + j\epsilon'' \quad (28)$$

Setting $\epsilon'' = \frac{(\epsilon_s - \epsilon_\infty)\omega\tau}{1 + \omega^2\tau^2}$ and substituting into equation (28) gives

$$\epsilon' = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + \omega^2\tau^2} \quad (29)$$

From equation (28);

$$j\epsilon'' = \epsilon' - \epsilon_\infty - \frac{\epsilon_s - \epsilon_\infty}{1 + j\omega\tau} \quad (30)$$

Substituting equation (29) into (30); we have

$$\varepsilon'' = \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{1 + \omega^2\tau^2} \quad (31)$$

Recall equation (5),

$$\begin{aligned} \tan \delta = \frac{\varepsilon''}{\varepsilon'} &= \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{1 + \omega^2\tau^2} \bigg/ \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2\tau^2} \\ &= \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{\varepsilon_\infty\omega^2\tau^2 + \varepsilon_s} \end{aligned} \quad (32)$$

Equations (29) and (31) are known as Debye equations and they describe the behaviour of polar dielectrics at various frequencies.

3. MATHEMATICAL DERIVATION OF THE RELAXATION TIME

Recall equation (31) and differentiating it with respect to $\omega\tau$ using quotient rule i.e.

$$\frac{d\varepsilon''}{d\omega\tau} = \frac{(1 + \omega^2\tau^2)(\varepsilon_s - \varepsilon_\infty) - (\varepsilon_s - \varepsilon_\infty)\omega\tau \cdot 2\omega\tau}{(1 + \omega^2\tau^2)^2}$$

Simplifying the above expression gives

$$\begin{aligned} \frac{d\varepsilon''}{d\omega\tau} &= (\varepsilon_s - \varepsilon_\infty) \left[\frac{1 + \tau^2\omega^2 - 2\omega^2\tau^2}{(1 + \omega^2\tau^2)^2} \right] \\ &= (\varepsilon_s - \varepsilon_\infty) \frac{(1 - \omega^2\tau^2)}{(1 + \omega^2\tau^2)^2} \\ \frac{d\varepsilon''}{d\omega\tau} &= \frac{(\varepsilon_s - \varepsilon_\infty)(1 - \omega^2\tau^2)}{(1 + \omega^2\tau^2)^2} \end{aligned} \quad (33)$$

According to Agilent [13] Basics of Measuring the Dielectric properties of materials, a material that has a single relaxation frequency as exhibited by the Debye relation will appear as a semicircle with its centre lying on the horizontal $\varepsilon'' = 0$ axis and the peak of the loss factor occurring at $\frac{1}{\tau}$. Assuming that it is lying below the horizontal $\varepsilon'' = 0$ axis and then equating equation (33) to zero.

$$\frac{d\varepsilon''}{d\omega\tau} = \frac{(\varepsilon_s - \varepsilon_\infty)(1 - \omega^2\tau^2)}{(1 + \omega^2\tau^2)^2} = 0 \quad (34)$$

$$(\varepsilon_s - \varepsilon_\infty)(1 - \omega^2\tau^2) = 0 \quad (35)$$

In equation (35), if $(\varepsilon_s - \varepsilon_\infty) \neq 0$, then

$$(1 - \omega^2\tau^2) = 0 \quad (36)$$

Solving for $\omega\tau$ in equation (36), we have

$$\begin{aligned} \omega^2\tau^2 &= 1 \\ \Rightarrow \omega\tau &= 1 \end{aligned} \quad (37)$$

Recall

$$\omega = 2\pi f_r \quad (38)$$

Where f_r is the relaxation frequency. Now substituting equation (38) into equation (37) and then solve for τ , we have

$$\begin{aligned} 2\pi f_r \tau &= 1 \\ \tau &= (2\pi f_r)^{-1} \end{aligned} \quad (39)$$

The values of the complex and static permittivity of methanol used in this work was adapted from National Physical Laboratory Report MAT 23 [14]. The experimental data of both complex and static permittivity as adapted in [14] were solved in the Debye relaxation equations (29) and (31) using Maple-13 and results were generated as shown in Table 1 below:

Table 1. The static permittivity ε_s , complex permittivity ε_∞ and relaxation frequency

| Temperature $^{\circ}\text{C}$ | ε_s | ε_∞ | f (GHz) |
|--------------------------------|-----------------|----------------------|---------|
| 10 | 35.74 | 5.818 | 2.262 |
| 20 | 33.64 | 5.654 | 2.822 |
| 30 | 31.69 | 5.450 | 3.490 |
| 40 | 29.85 | 5.251 | 4.283 |
| 50 | 28.19 | 5.224 | 5.175 |

Source: NPL Report MAT, 2012 [14]

4. RESULTS AND DISCUSSION

The experimental data for static ε_s and complex ε_∞ permittivity substituted in equations (29) and (31). An algorithm was written and maple-13 was used to generate results for dielectric constant ε' and loss factor ε'' . The obtained in this work were discussed based on the already existing theories. The effect of the frequency for the dielectric constant and loss factor of methanol for different temperatures are shown in the Fig. 1 and Fig. 2. The results of our work are also used to plot Cole-Cole diagrams as shown in Fig. 3a to Fig. 3e.

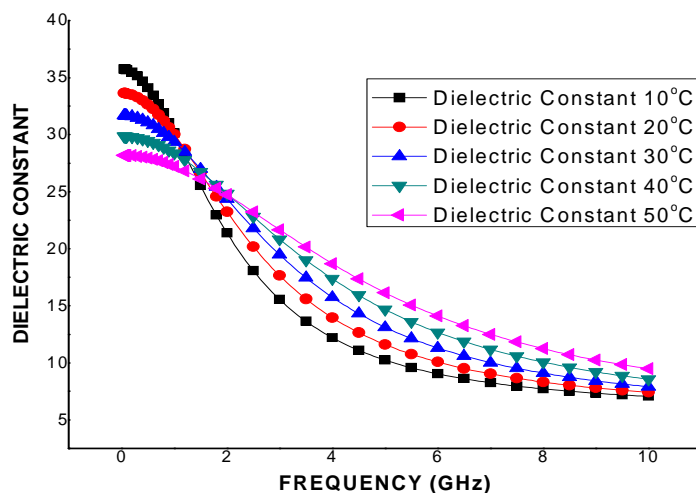


Fig. 1. Graph of the dielectric constant of methanol against the frequency. The dielectric constant is higher at frequency 0.05 GHz and temperature 10°C. The dielectric constant then decrease as both frequency and temperature increases

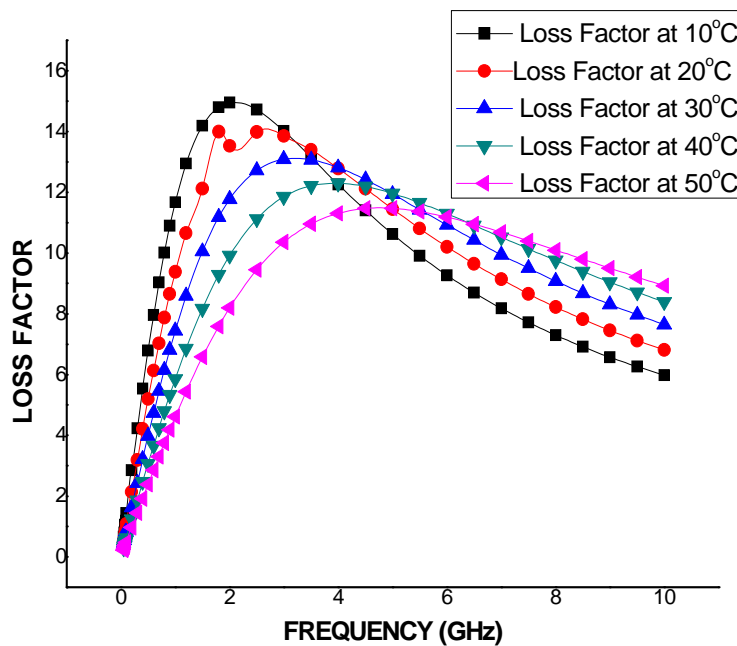


Fig. 2. Graph of the loss factor against the frequency. The loss factor of methanol is small at low frequency (i.e. 0.66, 0.50, 0.46, 0.29 and 0.22) for temperature 10°C, 20°C, 30°C, 40°C, and 50°C respectively but as the frequency increases the loss factor also increases to a maximum value of 14.95, 14.00, 13.09, 12.30 and 11.45 respectively. The value however decreased to 5.97, 6.81, 7.64, 8.40 and 8.92 when the frequency increased to 10 GHz after attaining its maximum

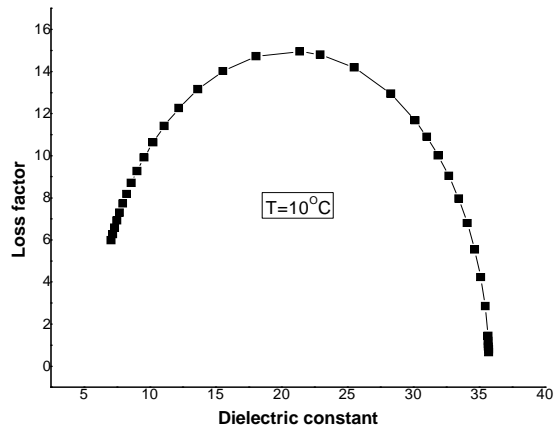


Fig. 3a. A Cole-Cole plot at temperature 10°C

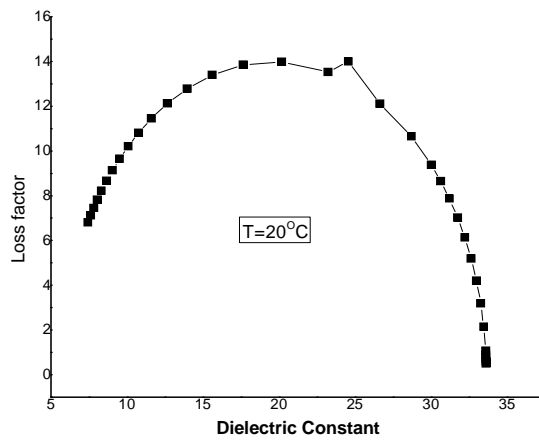


Fig. 3b. A Cole-Cole plot at 20°C

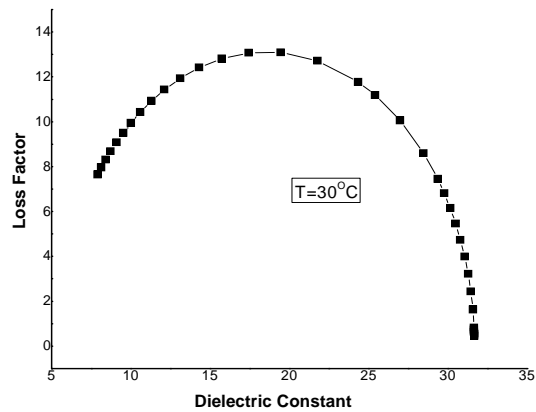


Fig. 3c. A Cole-Cole plot at 30°C

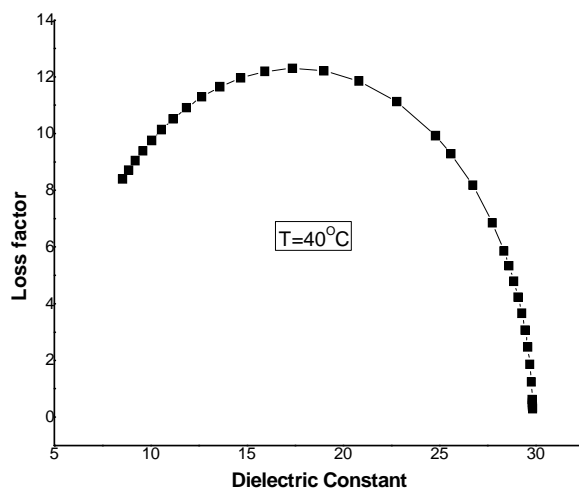


Fig. 3d. A Cole-Cole plot at 40°C

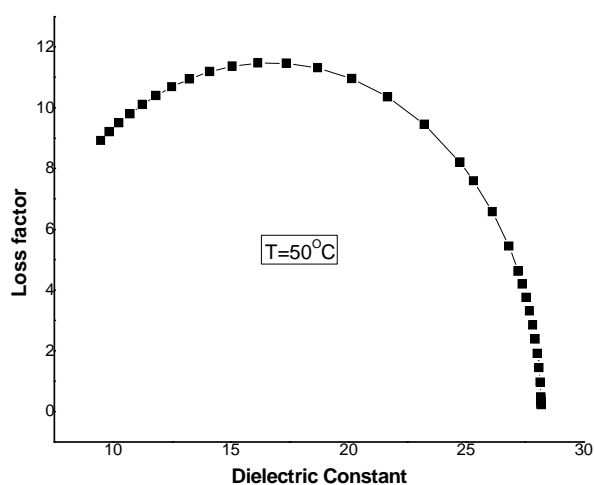


Fig. 3e. A Cole-Cole plot at 50°C

Fig. 3a to Fig. 3e shown in the above, are graphs of loss factor ϵ'' against the dielectric constant ϵ' known as Cole-Cole plot. The points on these graphs actually formed a semi-circle. However, there are some discrepancies in all the figures as shown in the above.

4.1 Discussion

The dielectric constant ϵ' and the loss factor ϵ'' of methanol were fitted using the Debye relaxation model. The computation was done within the frequency range of 500 MHz

to 10 GHz and temperature range of 10°C to 50°C. The parameters such as temperature and frequency were varied in order to ascertain which value of the frequency and temperature one can obtain the highest value of the dielectric constant. The graph of the dielectric constant ϵ' against the frequency and that of the loss factor ϵ'' against the frequency were then plotted. The results of the loss factor ϵ'' and dielectric constant ϵ' obtained in this work were used to plot Cole- Cole diagrams as shown in Fig. 3a to Fig. 3e above.

The Cole-Cole plots indeed formed a semi-circle which suggest that methanol exhibits Debye relaxation model. The discrepancies observed in our plots may just be due to distribution of relaxation times in methanol.

The dielectric constant of methanol was found to be higher at 0.05 GHz and temperature 10°C (see Fig. 1). However, as both the frequency and temperature increases beyond 0.05 GHz and 10°C, the dielectric constant continue to decrease steadily until it reaches 1.0 GHz. Beyond this frequency the dielectric constant of methanol decreases as the frequency increases. The dielectric constant however increased as the temperature increases at higher frequency values.

The higher value of the dielectric constant ϵ' at low frequencies as observed in Fig. 1 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 1.0 GHz may be due to the fact that the dipoles cannot follow up the rapid variation of the applied field [15].

As the temperature increased beyond 10°C the dielectric constant decreases especially at the frequency range of $0.05 \leq f \leq 1.0 \text{ GHz}$. This decrease in the dielectric constant as the result of increase 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. This suggest a freezing of electric dipoles at low temperature [16] 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 results have shown a sudden increase in the dielectric constant as the temperature increased beyond 10°C between the frequency ranges of 1.2 GHz to 10 GHz. This increase in the dielectric constant as the result of an increase in the frequency may be due to the permanent electric dipoles possessed by methanol [15].

The loss factor on the other hand has a small value at frequency 0.05 GHz and increases to reach its maximum value as the frequency increases. It then decreases steadily after attaining the maximum value (see Fig. 2). 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) [13]. But as temperature increases the loss factor decreases especially within the frequency range of 0.05 GHz to 4.5 GHz. This behaviour exhibited by methanol is contrary to the statement made by Guan et al. [17] 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 [18]". However, beyond the 4.5 GHz, the loss factor of methanol increased as the temperature increase in line with what Guan and Wang said.

5. CONCLUSION

The dielectric constant of methanol has been optimized by varying the frequency and temperature. The computation was done using the Debye equation and its derivatives within the frequency range of $0.05 \leq f \leq 10 \text{ GHz}$. The results from our computation reveals that methanol has the highest dielectric constant at 0.05 GHz at temperature 10°C. However, as both the frequency and temperature increases the dielectric constant decreases continuously except for values beyond 1.0 GHz. At frequency 1.2 GHz and above the dielectric constant value increased as the temperature increases. This suggests that the dielectric constant of methanol is easily polarizable at lower frequency and temperature.

The loss factor ϵ'' was also plotted against the dielectric constant ϵ' in order to decide which relaxation model methanol obeyed. The results showed that methanol at those temperatures fit in Debye relaxation model ($\alpha = 0$). This is because all Cole-Cole plots that are shown in Figs. 3a to 3e formed a semi-circle.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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