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# Complex Permittivity Determination of Glycerol Using Graphical and Numerical Technique

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**ABSTRACT**

Dielectric properties of materials are important parameters because these properties are related to other physical properties such as moisture content. The Cole – Cole model has been used successfully to describe the experimental data for the dielectric constant of many materials as a function of frequency. From Cole-Cole model, the value for dielectric constant divided into two, $\varepsilon_o$ = low limit frequency dielectric constant and $\varepsilon_\infty$ = high limit frequency dielectric constant. Cole-Cole model also can determine the value for relaxation time of the material, $\tau$. This paper describes the dielectric constant and relaxation time determination of glycerol palm oil based using graphical and numerical methods. The value for dielectric constant was measured by glycerol produced using transesterification process of palm oil and measured at the ambient temperature. Cole-Cole model has been used to describe the experimental data by using graphical and numerical methods. Using both methods, the value of dielectric constant and relaxation time can be determined by the data obtained from the experiment. Then identify the consistency of the dielectric constant and relaxation times obtained from both the methods were discussed. Dielectric measurement has been carried out to obtain experimental data of dielectric constant and relaxation time for glycerol palm oil based. From numerical method, the value for $\varepsilon_\infty$= 4.0, $\varepsilon_o$ = 47 and $\tau$ = 1.62 x10-10s. From graphical method, the value for $\varepsilon_\infty$= 4.0, $\varepsilon_o$ = 42.5and $\tau$ = 1.80 x10-10s. By comparing the result to previous research, both methods show a good approach to estimate the dielectric constant and relaxation time for glycerol palm oil based sample.

## INTRODUCTION

Dielectric properties of materials are important parameters because these properties are related to other physical properties such as moisture content(Bishay 2000). It is a known fact that the value for dielectric properties for many liquids such as glycerol, water and wine(Kuang & Nelson 1997; Watanabe *et al*. 2009; McDuffie *et al*. 1962) can often be described in terms of dielectric constant as a function of frequency for ideal systems. This also applies to solids such as fruits and vegetables(Kuang & Nelson 1997). The Cole – Cole model(Cole & Cole 1941) has been used successfully to describe the experimental data for the dielectric constant of many materials as a function of frequency(Davidson & Cole 1951; McDuffie *et al*. 1962; Bishay



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2000; Salman *et al*. 2014; Said & Varadan 2009). The Cole-Cole model interpreted the dielectric constant mainly on four parameters, , $\varepsilon_o$= low limit frequency dielectric constant and $\varepsilon_\infty$ = high limit frequency dielectric constant, the relaxation time, $\tau$ and an exponent factor $\alpha$ (Bishay 2000; Cole & Cole 1941; Davidson & Cole 1951; Watanabe *et al*. 2009). Other than Cole-Cole model, dielectric constant also be described by Debye equation and Havriliak–Negami equation(Davidson & Cole 1951; Yamada Pittini *et al*. 2008).

Palm oil is one of the most widely used plants where it is produced from oil palm tree. Oil palm plantations abound in the tropical rainforests in Malaysia and Indonesia (Tanaka 2008; Pawlik & Prociak 2012; Chuayjuljit & Sangpakdee 2007). Palm oil is a biological resource that provides the highest return on oil-based hydrocarbons and far more effective than any other commercial crop oil(Arniza *et al*. 2015). Continuous increased in petroleum prices, giving the user a paradigm shift towards using renewable raw materials for industrial and commercial goods. The use of palm oil in glycerol production, which is renewable and biodegradable, making palm oil as an alternative raw material(Tay *et al*. 2011). Furthermore, there is an increased awareness of social responsibility toward the environment to meet the demand for renewable resources and environmentally friendly products(Arniza *et al*. 2015; Xiao *et al*. 2013).

Studies have been done to determine the dielectric constant for glycerol using Cole – Cole model with some limitation. (Davidson & Cole 1951) has conducted a study using pure glycerol. This study is a preliminary study for the determination of the dielectric constant glycerol using Cole-Cole model. Data obtained from glycerol that can be fitted except at high frequencies. This study also limited by an empirical formula by Debye dispersion equation. (McDuffie *et al*. 1962) has conducted a study using a mixture of glycerol and water. In this study, (McDuffie *et al*. 1962) have been mixed glycerol and water content by measuring the viscosity extrapolated vs temperature for various water concentrations as given by Segar and Oberstar.

The study from (McDuffie *et al*. 1962; Davidson & Cole 1951) will be used as a reference value for this paper. In this research, the value for dielectric constant and relaxation time was determined by glycerol produced using transesterification process of palm oil and measured at the ambient temperature.

## MATERIAL AND METHOD

According to Cole – Cole equation, the complex relative permittivity, $\varepsilon^*$ as a function of frequency is given by

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_o - \varepsilon_\infty}{1 + (j\omega\tau)^{1-\alpha}} \tag{1}$$

Where
$\varepsilon_o$= low limit frequency dielectric constant
$\varepsilon_\infty$ = high limit frequency dielectric constant
$\omega = 2\pi f$ (Angular frequency)
$\tau$= relaxation time
$\alpha$= degree of relaxation distribution

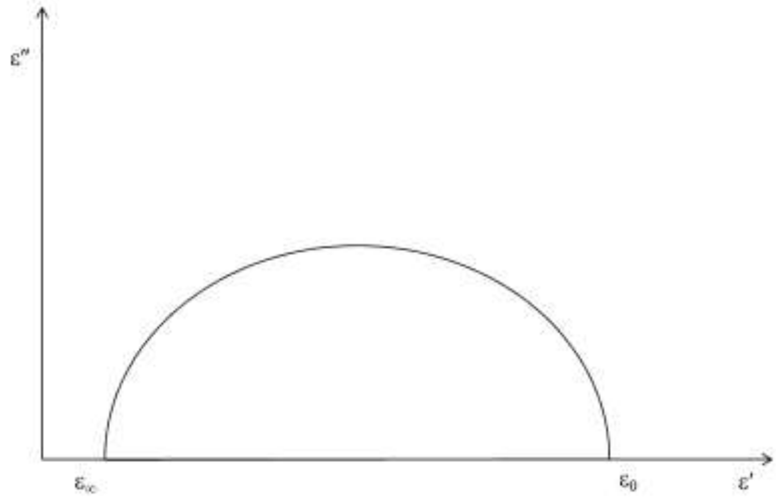


**Fig. 1:** Graph that generate by Cole-Cole model

Fig. 1 shows the semi-circle curve plotted by Cole – Cole model(Davidson & Cole 1951). Two methods have been used to determine the value of dielectric constant that is the numerical method and graphical method. Numerical Method is the method for evaluating the numerical values of the parameters needed in the calculation of the dielectric constant in the Cole – Cole model(Bishay 2000). Graphical method is the plot of the points in the plane which constitute the graph of a given real function or a pictorial diagram depicting interdependence of variables(Sybil P. Parker 2003).

***a. Numerical method:***

In the numerical method, the value for $\varepsilon_0$ and $\varepsilon_\infty$, were obtained from the Cole – Cole graph. According to equation (1), the real and imaginary parts can be represented by

$$\varepsilon^* = \varepsilon' - i\varepsilon'' \qquad (2)$$

From equation (1) and equation (2), Argand diagram can be generated as shown in Fig. 2(Cole & Cole 1941)

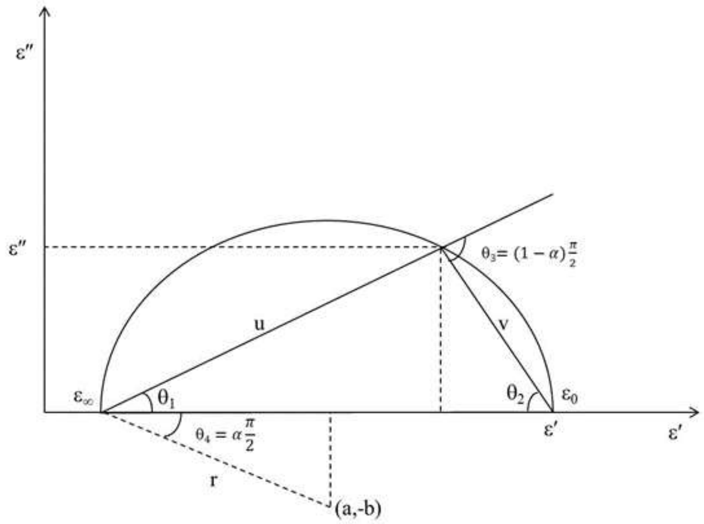


**Fig. 2:** Argand diagram of dielectric constant as given by the Cole – Cole model

Therefore

$$u = \varepsilon^* - \varepsilon_\infty \quad \text{and} \quad v = \varepsilon_\circ - \varepsilon^* = u(j\omega\tau)^{1-\alpha} \qquad (3)$$

***evaluate the accuracy of $\varepsilon_0$ and $\varepsilon_\infty$:***

To evaluate the accuracy of $\varepsilon_0$ and $\varepsilon_\infty$, data were divided into 3 sections as shown in Fig. 3(Bishay 2000).

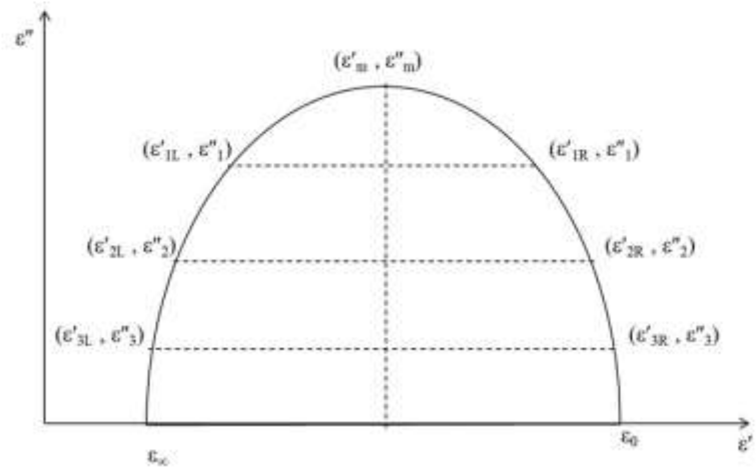


**Fig. 3:** Section divided into three to calculate $\varepsilon_R$ and $\varepsilon_L$

Therefore, the value of $\varepsilon_R$ and $\varepsilon_L$ can be calculated using Equations (4) and (5)

$$\varepsilon_R = \frac{\varepsilon'_{1R} + (\varepsilon''_2 - \varepsilon''_1)(\varepsilon'_{3R} - \varepsilon''_1)}{(\varepsilon''_3 - \varepsilon''_1)} \tag{4}$$

$$\varepsilon_L = \frac{\varepsilon'_{1L} + (\varepsilon'_{2L} - \varepsilon''_1)(\varepsilon'_{3L} - \varepsilon''_{1L})}{(\varepsilon''_3 - \varepsilon''_1)} \tag{5}$$

The value of $\varepsilon_0$ and$\varepsilon_\infty$ must be approximately equal to the value of $\varepsilon_R$ and $\varepsilon_L$.

***Calculate $\alpha$ and $\tau$ values:***

From Fig. 2, values for $\theta_1$ , $\theta_2$ and $\theta_3$ can be deduced by simple trigonometry

$$\theta_1 = \sin^{-1}\left(\frac{\varepsilon''}{u}\right) \quad , \quad \theta_2 = \sin^{-1}\left(\frac{\varepsilon''}{v}\right) \quad \text{and} \quad \theta_3 = \theta_1 + \theta_2 \tag{6}$$

The value for $\alpha$ and $\theta_3$ generated in Fig. 2 can be show as equation 7

$$\theta_3 = (1-\alpha)\frac{\pi}{2} \tag{7}$$

The relationship between v and u in equation 3 can be expressed in the following equations

$$v = u(j\omega\tau)^{1-\alpha} \quad \text{and} \quad \left|\frac{v}{u}\right| = (\omega\tau)^{1-\alpha} \tag{8}$$

Hence, the relaxation time, $\tau$ can be calculate using equation

$$\tau = \frac{1}{\omega}\left[\frac{(\varepsilon_\circ - \varepsilon')^2 + \varepsilon''^2}{(\varepsilon' - \varepsilon_\infty)^2 + \varepsilon''^2}\right]^{\frac{1}{2(1-\alpha)}} \tag{9}$$

***b. Graphical method:***

From Cole – Cole model in equation 1, when the value of $\alpha$=1, the Debye equation describes a dielectric response with a single relaxation time constant as

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_o - \varepsilon_\infty}{1 + (j\omega\tau)} \tag{10}$$

From equation 1, the dielectric constant and loss factor can be written as

$$\varepsilon' = \varepsilon_\infty + \frac{\varepsilon_o - \varepsilon_\infty}{1 + \omega^2\tau^2} \tag{11}$$

$$\varepsilon'' = \frac{(\varepsilon_o - \varepsilon_\infty)\omega\tau}{1 + \omega^2\tau^2} \tag{12}$$

According to equation 11 and 12, Cole – Cole model can be deduce into two linear equation

$$\omega\varepsilon'' = \frac{(\varepsilon_o - \varepsilon_\infty)\omega^2\tau}{1 + \omega^2\tau^2} = \frac{1}{\tau}(\varepsilon_o - \varepsilon') \tag{13a}$$

$$\frac{\varepsilon''}{\omega} = \frac{(\varepsilon_o - \varepsilon_\infty)\tau}{1 + \omega^2\tau^2} = \tau(\varepsilon' - \varepsilon_\infty) \tag{13b}$$

Equations 13a and 13b can be illustrated graphically as in Fig. 4a and 4b. Where the gradient of graph as shown in Fig. 4a and 4b represents the value of $\tau$ and $\frac{1}{\tau}$, respectively. While the intercept at $\varepsilon'$ axis for Fig. 4a and 4b represents the value for and $\varepsilon_0$ and $\varepsilon_\infty$, respectively.

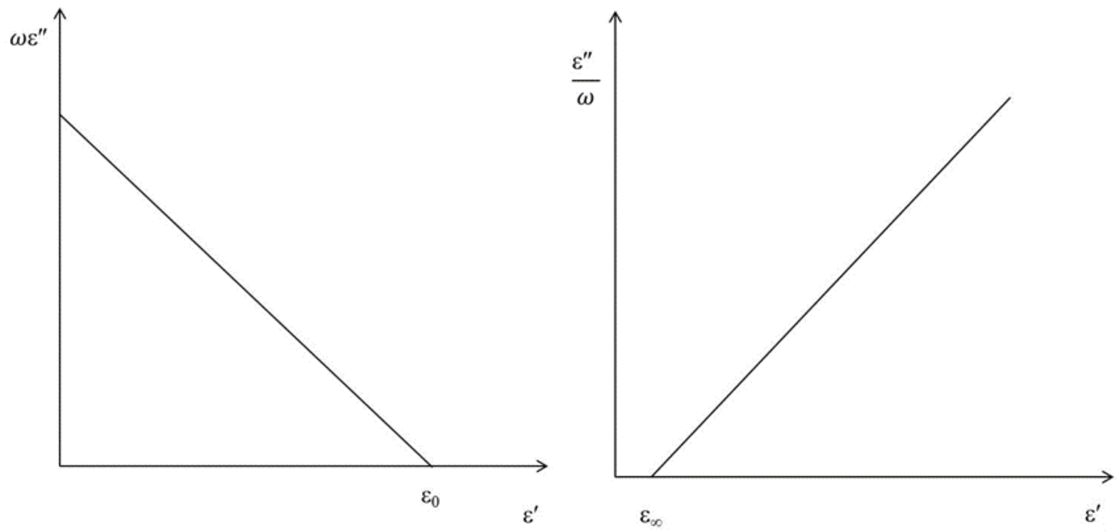


**Fig. 4:** (a) Graph illustrated by equation 13a, (b) Graph illustrated by equation 13b

In this study, the glycerol palm oil based with molecular weight 92.09380 was obtained from FPG Oleochemical Sdn. Bhd. Kuantan, Pahang. The complex permittivity of the glycerol palm oil based was measured in the frequency range between 100 MHz and 20 GHz using Agilent E5071C Network Analyzer in conjunction with Agilent 85070E dielectric probe.

The experimental value of dielectric constant $\varepsilon'$ and loss factor $\varepsilon''$ at room temperature are shown in Fig. 5. To analyse the measured dielectric functions using Cole – Cole model, the real part of the complex permittivity must be plotted on the x-axis and the imaginary part of that on the y- axis. In this paper, after plotting the Cole – Cole graph as in Fig. 6, there are two different types of methods used in determining the dielectric constant value which are numerical method and graphical method.

## RESULT AND DISCUSSION

In determination of dielectric constant and relaxation time for glycerol palm oil based, data obtained from measurement using Agilent E5071C Network Analyzer in conjunction with Agilent 85070E dielectric probe. The dielectric measurement was conducted over a frequency range from 8 GHz to 12 GHz (X-band) at room temperature(Mahmud *et al*. 2017).

### *a. Numerical methods:*

After plotting the experimental value of dielectric constant $\varepsilon'$ and loss factor $\varepsilon''$, in excellent shape obtained graph is shown in Fig. 5.The experimental data as shown in Fig. 5 have been fitted to obtain semi-circle plot as shown in Fig. 6. This fitted data obey the Cole-Cole model graph as in Figure1(Cole & Cole 1941). In numerical method, argand plane generated from experimental data are shown in Fig. 7. According to (Bishay 2000) the values of $\varepsilon_\infty$ , $\varepsilon_0$,$\tau$ and $\alpha$ could obtained from argand plane and calculated using equation (3) to equation (9) in the method and material section. The values of for numerical methods for each parameter was $\varepsilon_\infty = 4.0$, $\varepsilon_0 = 47$, $\tau = 1.62 \text{ x}10^{-10}$ and $\alpha = 0.1937$.

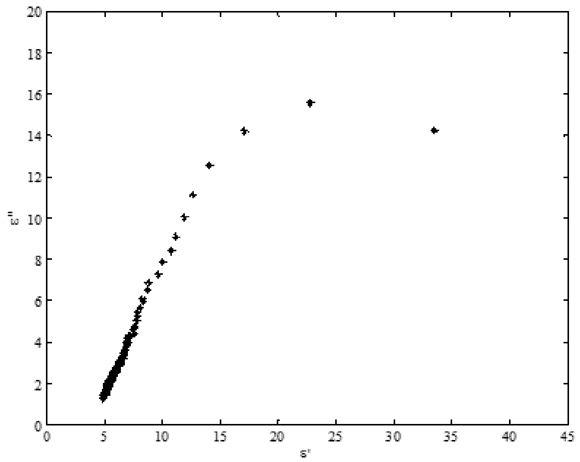

**Fig. 5:** Experimental value for ε″ against ε′

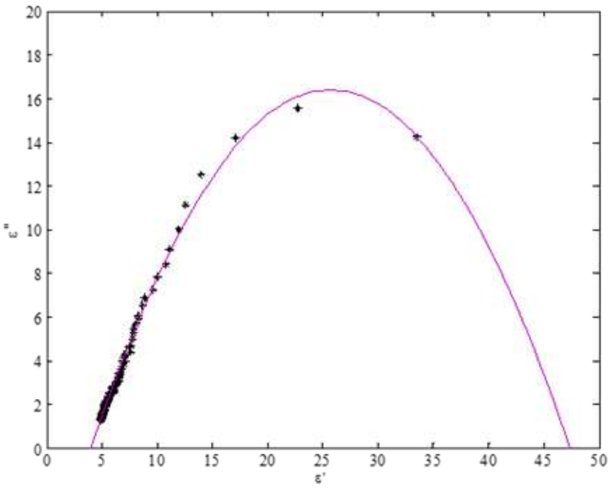

**Fig. 6:** Cole-Cole plot for glycerol palm oil based

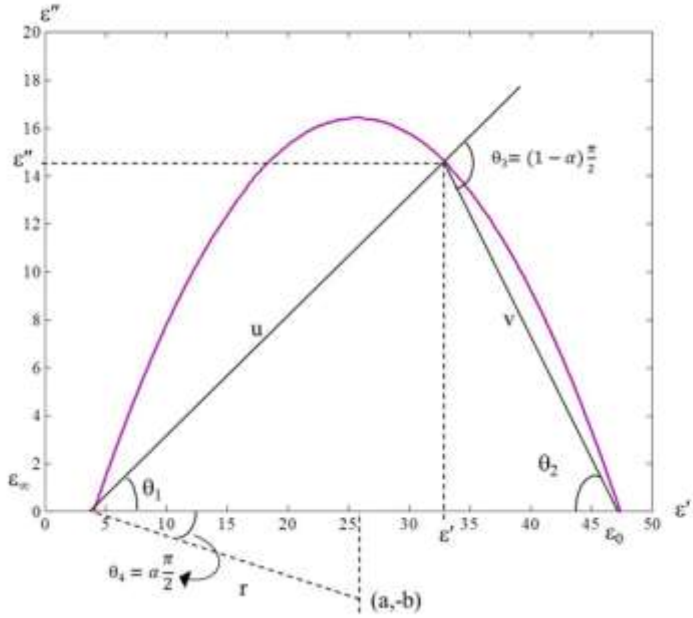


**Fig. 7:** Argand diagram for glycerol palm oil using numerical method

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***b. Graphical method:***

In graphical method, Equations 13a and 13b have been used to produce graph $\omega\varepsilon''$against $\varepsilon'$ and $\frac{\varepsilon''}{\omega}$ against $\varepsilon'$ as shown in Fig. 8a and 8b, respectively.

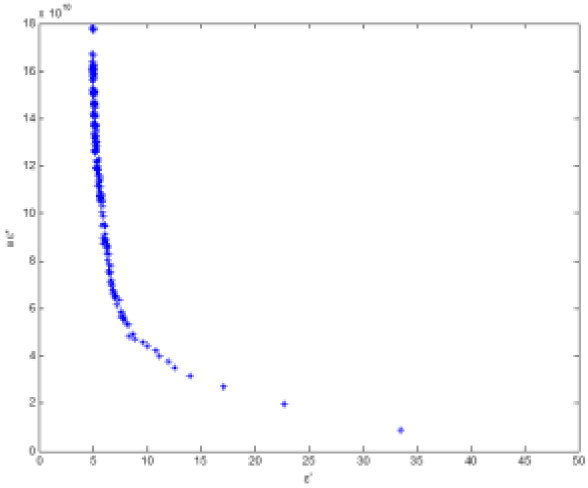
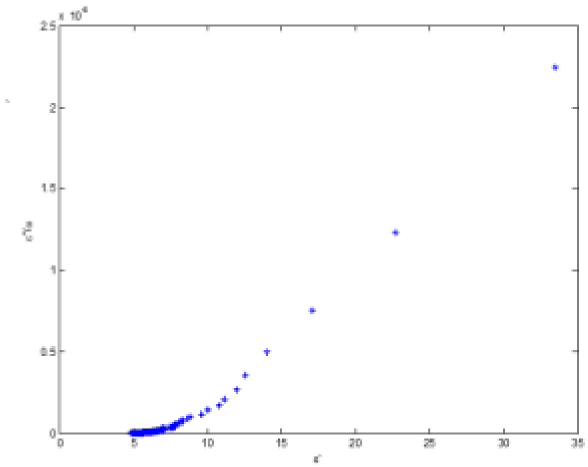



**Fig. 8:** (a) Graph $\omega\varepsilon''$against $\varepsilon'$ for glycerol palm oil based.




(a)

**Fig. 8:** (b) Graph $\frac{\varepsilon''}{\omega}$ against $\varepsilon'$ for glycerol palm oil based.

From data plotting in Fig. 8(a), measurement below 3 x $10^8$ Hz could be fitted(McDuffie *et al*. 1962). After linear regression the best value for $\varepsilon_0$ obtained from the graph was 42.5. In Fig. 8(b), data plotting was less than 2.5 x $10^8$ Hz. After the linear regression for data plotting in Fig. 8 (b), the best value for $\varepsilon_\infty$ was 4.0. The value for relaxation time,$\tau$ could obtained from the gradient for both graph $\omega\varepsilon''$against $\varepsilon'$ and $\frac{\varepsilon''}{\omega}$ against $\varepsilon'$. All data were obtained from numerical method and graphical methods are tabulated in Table 1.

**Table 1:** The result for dielectric constant of glycerol palm oil based

| Method | $\varepsilon_\infty$ | $\varepsilon_0$ | $\tau$ (sec) | $\alpha$ | Remark |
|---|---|---|---|---|---|
| Numerical Method | 4.0 | 47 | 1.62 $\times 10^{-10}$ | 0.1937 | At ambient temperature |
| Graphical Method | 4.0 | 42.5 | 1.80 $\times 10^{-10}$ | | At ambient temperature |
| (Cole & Cole 1941) | 5.0 | 63 | - | 0.21 | At -50 $^0$C |
| (Davidson & Cole 1951) | 4.1 | 63.9 | 1.25 $\times 10^{-4}$ | - | At -50 $^0$C |
| (McDuffie *et al*. 1962) | 4.0 | 54.3 | 158 $\times 10^{9}$ | - | At -19.5 $^0$C and water fraction 5.04 |

Table 1, clearly shows that the value of $\varepsilon_\infty$ for both numerical method and graphical method are the same value that is 4.0. This value is approximately the same as known value(Davidson & Cole 1951; McDuffie *et al*. 1962). The $\varepsilon_0$ value from numerical method is 47 and graphical method is 42.5. These values are far deviated from the known values(McDuffie *et al*. 1962; Davidson & Cole 1951; Cole & Cole 1941). The $\varepsilon_0$ value for (Davidson & Cole 1951) and (Cole & Cole 1941)was 63.9. The value was measured at the -50 $^0$C. The $\varepsilon_0$ value for (McDuffie *et al*. 1962) was 54.3 and it measured at -19.5 $^0$C. From the interpretation Table 1, the $\varepsilon_0$ values decrease when the temperature increase. $\tau$ values from both methods is approximately close. However, these values which are 1.62 x$10^{-10}$ s and 1.80 x$10^{-10}$ s both did not satisfy the known value(McDuffie *et al*. 1962; Davidson & Cole 1951).

***Conclusion:***

The results obtained for glycerol palm oil based agree reasonably with the experimental data. The numerical values of the $\varepsilon_{0,}$ $\varepsilon_\infty$ and $\tau$ calculated by using numerical method and graphical method were almost consistent. The $\varepsilon_\infty$ value also consistent with the known value. For $\varepsilon_0$ values, compare to the knowns value, the $\varepsilon_0$ values decrease when the temperature increase. The study confirms that the Cole-Cole model is an adequate approach for the calculation of the dielectric constant of glycerol palm oil based. The glycerol palm oil based in this study is used in production in polyurethane palm oil based.

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