

Modeling of Dielectric Relaxation for Lossy Materials at Microwave Frequencies using Polynomial Approaches

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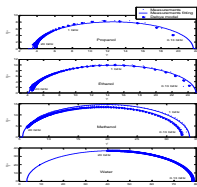
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Graphical abstract



Abstract

This paper reviews the dielectric modeling techniques and features of various dielectric models. In fact, many of the dielectric models have a polynomial characteristic, thus polynomial fitting is proposed in order to estimate the parameters in the dielectric model based on measured data. The coefficients of the polynomial equation were optimized with the measured data using HP 85070 B dielectric probe. Finally, the parameters in the dielectric models can be easily determined, based on the estimated polynomial coefficients. The parameters of the dielectric model were successfully estimated and compared with parameter values in the literature.

Keywords: Relative permittivity; dielectric model; polynomial approaches

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1.0 INTRODUCTION

Dielectric study is important for materials located in the electric field at microwave frequencies, since at microwave frequencies the polar molecules in some dielectric materials become excited. In the industry field, the study of dielectric properties of industrial materials is becoming increasingly critical. This is because the dielectric properties of materials has played an important role in the construction of high-frequency electronic components, the quality of printed circuit board (PCB) substrate, the efficiency of microwave absorption materials, and the performance of dielectric antenna design.

The interaction between dielectric materials with electromagnetic fields can be best described by complex dielectric permittivity, $\epsilon^* = \epsilon' - j\epsilon''$. The real part, ϵ' is the dielectric constant which is the parameter influencing the electric field distribution and the phase of waves traveling through the material. In contrast, the imaginary part, the so-called loss factor, ϵ'' influences the energy absorption or attenuation of the material. Dielectric spectroscopy is a macroscopic study requiring theoretical formulations to represent and describe the dielectric mechanism inside the materials.

The first dielectric expression was derived by Debye [6] based on permanent dipole moments in dielectric materials. However, the polarization molecules in inhomogeneous

insulator materials are complex due to many body interactions between binding molecules. Thus, a lot of the dielectric equation was modified from the ideal Debye theory, such as Cole-Cole (CC) [4], Davidson-Cole (DC) [5], and Havriliak-Negami (HN) [9], as well as Kohlrausch-Williams-Watts (KWW) distribution function, in order to represent the actual properties of the material. Those models are more suitable for lossy materials at microwave frequency range (molecules orientational

relaxation) involves a search for the three parameters (ϵ_s , ϵ_w and τ) as detailed in Section 2 and 3. In order to obtain a calculated value of permittivity closed to the measurement results, some empirical parameter were added to the model to establish a good fit with the measurement data, such as the modified Cole-Cole model [4]. In this study, the polynomial approaches are used to establish the parameters inside the dielectric models based on the measurement results.

2.0 DIELECTRIC MODELS

In general cases, after the dielectric material has been placed in a steady external field, its molecules tend to gravitate exponentially to an equilibrium state as given by [14].

$$f(t) = 1 - \exp(-t/\tau) \quad (1)$$

The relaxation function, $f(t)$ can be related to relative permittivity, ϵ_r material via Laplace transform as

$$\frac{\epsilon_s - \epsilon_\infty}{\epsilon_s - \epsilon_\infty} \int_0^\infty f(t) e^{-j\omega t} dt \quad (2)$$

Thus, in the frequency domain, ideal relative permittivity can be expressed as equation (3) (in Table 1). However, various alternative expression were done to fit the practical permittivity data, such as the Cole-Cole model [4], the Davidson-Cole model [5], and the Havriliak-Negami model [9], as listed in Table 1.

Table 1 Various frequency-domain dielectric models

	Frequency-Domain Formulations
Fgdg Model [6]	$\epsilon_r = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + j\omega h} \quad (3)$ <p>where ϵ_∞ and ϵ_s are the optical permittivity and static permittivity, respectively. Symbol ω and h are the angular frequency and relaxation time.</p>
Cole-Cole Model [4]	$\epsilon_r = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + (j\omega h)^n} \quad (4)$ <p>where n is the empirical constant with $0 < n < 3$</p>
Cole-Davidson Model [5]	$\epsilon_r = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + (j\omega h)^n} \quad (5)$ <p>where n is the empirical constant with $0 < n < 3$</p>
Havriliak & Negami Model [9]	$\epsilon_r = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + (j\omega h)^n} \quad (6)$ <p>where n and m are the empirical constant with $0 < n < 3$ and $0 < m < 3$</p>

In practice, in a large number of heterogeneous systems it is found that the polarization does not decay as equation (1), but it is a response as a stretched exponential function [1].

$$f(t) = \exp(-t/h) \quad (7)$$

The stretched exponential behavior is called Kohlrausch-Williams-Watts (KWW). By using equation (7) as a decay function, equation (2) in the time-domain becomes

$$\epsilon_r = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{h} \int_0^\infty t^{\alpha-1} f(t) e^{-j\omega t} dt \quad (8)$$

For (N+1) relaxation systems, the dielectric properties can be represented by

$$\epsilon_r = \epsilon_\infty + \sum_{n=1}^N \frac{\epsilon_n - \epsilon_{n-1}}{1 + j\omega h_n} \quad (9)$$

3.0 DIELECTRIC MODELS IN POLYNOMIAL EXPRESSIONS

The approximated series of the Debye model provides more degrees of freedom to specify the broadness of experimental relaxation peaks. The polynomial equations listed in Table 2 are used to determine the dielectric parameters ϵ_s , ϵ_∞ , and h .

As mentioned in Section 2, the relative complex permittivity, ϵ_r for materials with only a single relaxation frequency is best described by the Debye or Cole-Cole model. The three widely acceptable representations [4, 13] of the Debye and Cole-Cole model are:

- 1) Cole-Cole plot, equation (12) or (13)
- 2) ϵ_r' versus $-k$ plot equation (10) and
- 3) ϵ_r' versus $\epsilon_r'/\omega k$ plot, equation (11)

Series expansion of Equation (3) is usually used for polynomial fitting of experimental data from which ϵ_s and ϵ_∞ can be estimated easily, once the relaxation time, h is known. The series expansion is described in detail in the Appendix. The real part of (3) can be represented in terms of equation (15). Since heterogeneous mixtures or materials may provide various forms of dispersion, relaxation time, h , can thus (3) be written in the form of equation (17). Similarly, the imaginary part of (3) can be represented in the form of equation (18).

The natural domain of the KWW relaxation function is time domain. In this study, the KWW relaxation function is transformed to frequency domain by expanding the function as a series expression given as equation (19) and (20). This series is only suitable when implemented at low frequencies, as well as short relaxation time, h . This is because the higher series terms in (19) and (20) are weak when the series values are greater than the calculation limit of the software.

The corresponding values of the empirical parameters between the Kohlrausch-Williams-Watts (KWW) function in time domain and the Havriliak-Negami (HN) functions in the frequency domain were found using numerical simulations by Alvarez et al. (1991). The corresponding value of those parameters, obtained from the literature [1] was plotted in Figure 1. In this work, the relationship between those parameters was re-expressed in polynomial form as tabulated in Table 3.

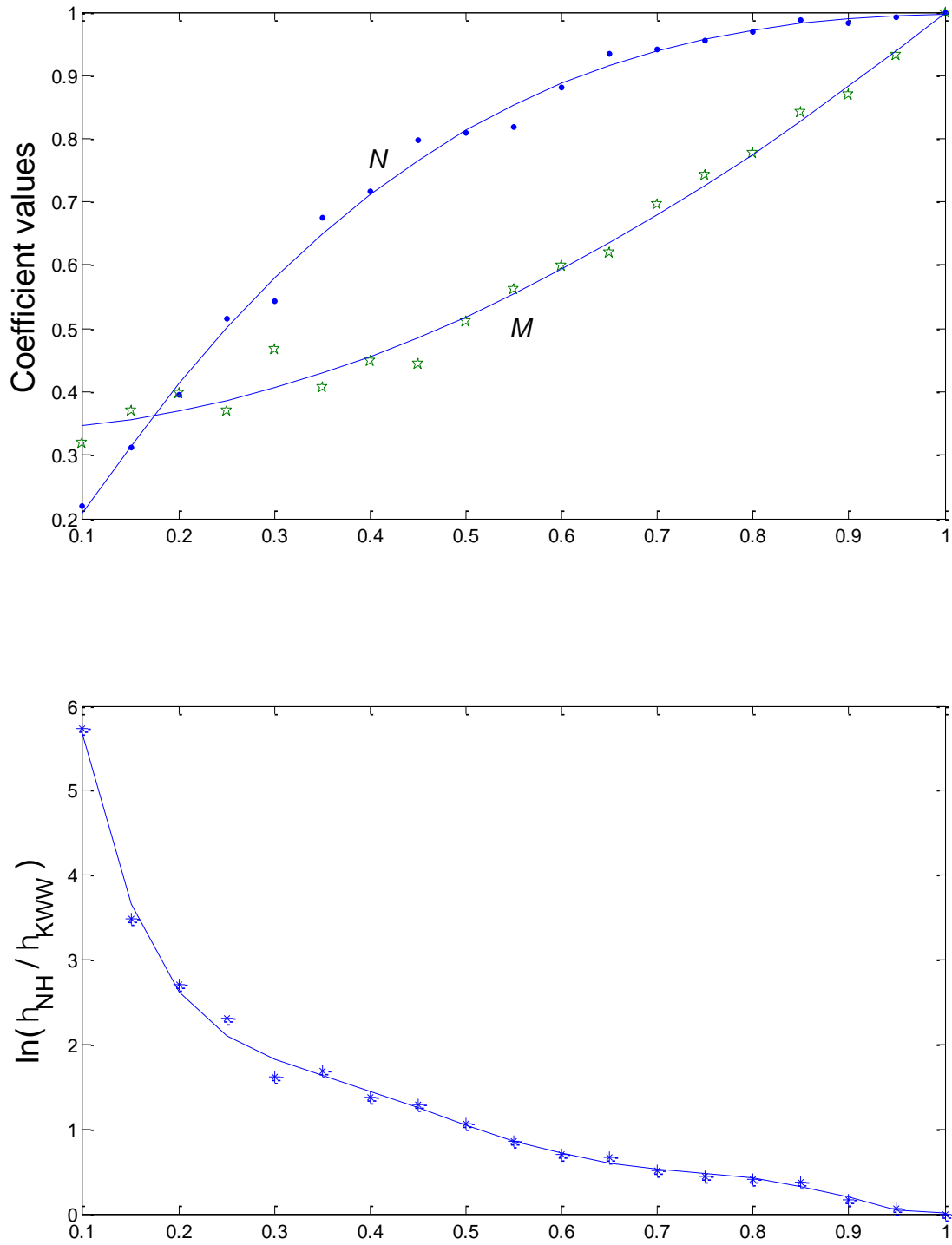


Figure 1 Values of the Havriliak-Negami (HN) fitting parameters as functions of the corresponding β values. The solid line is polynomial fitting and the equations are tabulated in Table 3

Table 2 Various of frequency-domain dielectric models

Polynomial Expressions	Dielectric Expressions
Linear equation $y = 1 + mx + zc$	Debye characteristics $\epsilon_r = 1 + \frac{fk}{\omega^2 + k^2} \quad (10)$ $\epsilon_r = 1 + \frac{fk}{\omega^2 + k^2 + \omega_w^2} \quad (11)$
Quadratic equation $y = 1 + ax^2 + bx + zc$	Debye characteristics $f \left \frac{\epsilon_r - 1}{\omega} \right ^2 = 1 + \frac{fk^2}{\omega^2 + k^2} + \frac{fk^2}{\omega^2 + k^2 + \omega_w^2} \quad (12)$ <hr/> Cole-Cole characteristics $P_1 \left \frac{\epsilon_r - 1}{\omega} \right ^2 = 1 + \frac{fk^2}{\omega^2 + k^2} + \frac{fk^2}{\omega^2 + k^2 + \omega_w^2} + P_2 \frac{\epsilon_r - 1}{\omega} + P_3 \quad (13)$ where $P_1 = 1 - \frac{\Delta}{\epsilon} \tan^2 \frac{\Delta f \omega}{\omega} \frac{N \epsilon d}{2} \frac{\bar{N}}{\bar{O}} \quad (14a)$ $P_2 = 1 - \frac{1}{4} \frac{\Delta f \omega_s}{\epsilon} \frac{z}{\omega^2} \frac{\bar{N}}{\bar{O}} \omega_s^2 + \frac{\Delta f \omega}{\omega} \tan^2 \frac{\Delta f \omega}{\omega} \frac{N \epsilon d}{2} \frac{\bar{N}}{\bar{O}} \quad (14b)$ $P_3 = 1 - \frac{1}{4} \frac{f \omega_s}{\omega} \frac{z}{\omega^2} \sec^2 \frac{\Delta f \omega}{\omega} \frac{N \epsilon d}{2} \quad (14c)$
N-order polynomial equation $y = 1 + \sum_{n=1}^N a_n x^n$	Debye characteristics (Simplexation) Real Part $\epsilon_r' = \epsilon_s + A_1 x + A_2 x^2 + A_3 x^3 + A_4 x^4 + \dots \quad (15)$ where $x = 1/f^2$ and $A_1 = 1/fkd \cdot k^2 f \epsilon_s \omega_w \epsilon ; \quad A_2 = 1/fkd \cdot k^4 f \epsilon_s \omega_w \epsilon ;$ $A_3 = 1/fkd \cdot k^6 f \epsilon_s \omega_w \epsilon ; \quad A_4 = 1/fkd \cdot k^8 f \epsilon_s \omega_w \epsilon^2$ <hr/> Imaginary Part $\frac{\epsilon_r''}{f} = B_1 + B_2 x + B_3 x^2 + B_4 x^3 + B_5 x^4 + \dots \quad (16)$ where $x = 1/f^2$ and $B_1 = 1/fkd \cdot k f \epsilon_s \omega_w \epsilon ; \quad B_2 = 1/fkd \cdot k^3 f \epsilon_s \omega_w \epsilon ;$ $B_3 = 1/fkd \cdot k^5 f \epsilon_s \omega_w \epsilon ; \quad B_4 = 1/fkd \cdot k^7 f \epsilon_s \omega_w \epsilon^2$

N-order polynomial equation

$$y = \sum_{n=0}^N a_n x^n$$

N-order polynomial equation

$$y = \sum_{n=0}^N a_n x^n$$

Debye characteristics (Multiple relaxation)

Real Part

$$\chi_r'' = \sum_{s=1}^N C_s x^s = C_1 x + C_2 x^2 + C_3 x^3 + C_4 x^4 + \dots \tag{17}$$

where $x = 1/f^2$ and

$$C_1 = \frac{f_0 d^2}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{2s} \frac{\Gamma(2s)}{\Gamma(2s)} \left(\frac{\omega \tau_1}{2}\right)^{2s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(2s)}{\Gamma(2s)} \left(\frac{\omega \tau_1}{2}\right)^{2s}$$

$$C_2 = \frac{f_0 d^4}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{4s} \frac{\Gamma(4s)}{\Gamma(4s)} \left(\frac{\omega \tau_1}{2}\right)^{4s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(4s)}{\Gamma(4s)} \left(\frac{\omega \tau_1}{2}\right)^{4s}$$

$$C_3 = \frac{f_0 d^6}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{6s} \frac{\Gamma(6s)}{\Gamma(6s)} \left(\frac{\omega \tau_1}{2}\right)^{6s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(6s)}{\Gamma(6s)} \left(\frac{\omega \tau_1}{2}\right)^{6s}$$

$$C_4 = \frac{f_0 d^8}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{8s} \frac{\Gamma(8s)}{\Gamma(8s)} \left(\frac{\omega \tau_1}{2}\right)^{8s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(8s)}{\Gamma(8s)} \left(\frac{\omega \tau_1}{2}\right)^{8s}$$

Imaginary Part

$$\frac{\chi_r''}{f} = \sum_{s=1}^N D_s x^s = D_1 x + D_2 x^2 + D_3 x^3 + D_4 x^4 + \dots \tag{18}$$

where $x = 1/f^2$ and

$$D_1 = \frac{f_0 d}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{2s} \frac{\Gamma(2s)}{\Gamma(2s)} \left(\frac{\omega \tau_1}{2}\right)^{2s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(2s)}{\Gamma(2s)} \left(\frac{\omega \tau_1}{2}\right)^{2s}$$

$$D_2 = \frac{f_0 d^3}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{4s} \frac{\Gamma(4s)}{\Gamma(4s)} \left(\frac{\omega \tau_1}{2}\right)^{4s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(4s)}{\Gamma(4s)} \left(\frac{\omega \tau_1}{2}\right)^{4s}$$

$$D_3 = \frac{f_0 d^5}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{6s} \frac{\Gamma(6s)}{\Gamma(6s)} \left(\frac{\omega \tau_1}{2}\right)^{6s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(6s)}{\Gamma(6s)} \left(\frac{\omega \tau_1}{2}\right)^{6s}$$

$$D_4 = \frac{f_0 d^7}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{8s} \frac{\Gamma(8s)}{\Gamma(8s)} \left(\frac{\omega \tau_1}{2}\right)^{8s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(8s)}{\Gamma(8s)} \left(\frac{\omega \tau_1}{2}\right)^{8s}$$

Kohlrausch-Williams-Watts (KWW) characteristic (Single relaxation)

Real Part

$$\chi_r'' = \sum_{s=1}^N K_s x^s = K_1 x + K_2 x^2 + K_3 x^3 + \dots \tag{19}$$

where $x = 1/f^2$ and f represents the frequency, and

$$K_1 = \frac{f_0 d^2}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{2s} \frac{\Gamma(2s)}{\Gamma(2s)} \left(\frac{\omega \tau_1}{2}\right)^{2s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(2s)}{\Gamma(2s)} \left(\frac{\omega \tau_1}{2}\right)^{2s}$$

$$K_2 = \frac{f_0 d^4}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{4s} \frac{\Gamma(4s)}{\Gamma(4s)} \left(\frac{\omega \tau_1}{2}\right)^{4s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(4s)}{\Gamma(4s)} \left(\frac{\omega \tau_1}{2}\right)^{4s}$$

$$K_3 = \frac{f_0 d^6}{\Delta} \sum_{s=1}^N \gamma_s \tau_1^{6s} \frac{\Gamma(6s)}{\Gamma(6s)} \left(\frac{\omega \tau_1}{2}\right)^{6s} \gamma_s \dots \sum_{s=1}^N \frac{\Gamma(6s)}{\Gamma(6s)} \left(\frac{\omega \tau_1}{2}\right)^{6s}$$

The $\Gamma(x)$ term in (19) is the Gamma function.

Imaginary Part

$$\frac{\chi_r''}{f} = \sum_{s=1}^N L_s x^s = L_1 x + L_2 x^2 + L_3 x^3 + L_4 x^4 + \dots \tag{20}$$

	<p>where $x = 1/f^2$ and</p> $L_1 = 1 - \frac{f^2 d^2 \epsilon''}{\epsilon' \omega} \frac{Y_s}{\omega} \frac{1}{\tau} ;$ $L_2 = 1 - \frac{f^2 d^4 \epsilon''}{3! \epsilon' \omega} \frac{Y_s}{\omega} \frac{1}{\tau} ;$ $L_3 = 1 - \frac{f^2 d^6 \epsilon''}{5! \epsilon' \omega} \frac{Y_s}{\omega} \frac{1}{\tau} ;$ $L_4 = 1 - \frac{f^2 d^8 \epsilon''}{7! \epsilon' \omega} \frac{Y_s}{\omega} \frac{1}{\tau} \dots$
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Table 3 The corresponding relationship between the Havriliak-Negami (HN) parameters (N , M , and h_{HN}) and the stretched exponential (KWW) parameters (τ and h_{KWW})

The corresponding relationship between the parameters of KWW and HN models in polynomial forms	
$\ln \frac{h_{HN}}{h_{KWW}}$	$\frac{N}{M} 446.62150 \tau^6 - 1647.6696 \tau^5 + 2426.5867 \tau^4 - 1819.4324 \tau^3 + 733.33748 \tau^2 - 154.86110 \tau + 5.423989 - 0.38583000$
$N = 0.66878694 \tau^3 + 2.3424665 \tau^2 + 2.7111283 \tau + 0.040639732 - 0.072980000$	
$M = 0.10864371 \tau^3 + 0.76452528 \tau^2 + 0.0030149013 \tau + 0.32962219 - 0.096463000$	

It is easier to calculate the dielectric properties of materials using the KWW function, since the function has only one unknown parameter (τ) compared to the HN function (M , N , and h_{HN}). Moreover, the relaxation time, h_{KWW} , is the actual relaxation value of the materials whilst the values of h_{HN} are normally adjusted to give the best fit for the experimental data. However, for simplicity, the dielectric properties of materials are usually calculated in the frequency domain.

4.0 RESULTS AND ANALYSIS

4.1 Estimation of Parameters Y_s , Y_w and h Using Linear Equation

The static permittivity, Y_s , optical permittivity, Y_w , and relaxation time, h , can be predicted directly from equations

(10) and (11) based on equation (3). In equation (10), the gradient of the line of the $Y_r - k$ plot is the relaxation time, h , whilst Y_s can be determined directly at $k = 1/\tau$. For equation (6.13b), the inverse relaxation time, $1/h$, can be calculated directly from the gradient of variation in Y_r with $Y_r/\tau k$ whilst the intersection between the line and Y_r -axis, when $Y_r/\tau k = 0$, gives the value of Y_w .

By testing (10) and (11), the measured relaxation time, h of bulk water at room temperature, T , was found as illustrated in Figure 2 (a), (b) and the result, which includes the estimated values of Y_s and Y_w were compared with the literature value as available in Table 4. The results show that the Y_r versus k regression is more stable and reliable than the Y_r versus the

$\chi_r^2 / \tau k$ regression estimating the relaxation time, h , of the materials. Linear plots of χ_r^2 versus $\chi_r^2 / \tau k$ for water revealed problems with data below 3 GHz, thus only data above 3 GHz were used in regression calculation. Meanwhile, the higher frequency measurement (2-20 GHz) is required for the accurate value estimation of χ_w for higher lossy materials ($h \sim 10^{12}$) by using the χ_r^2 versus the $\chi_r^2 / \tau k$ plot.

The measured relaxation time of bulk water ranging from 25 °C to 60 °C obtained from equation (10) is plotted as Arrhenius plots (see Figure 3). The linear relationships in Figure 3 indicate that the temperature dependence of the relaxation time can be described by equation (21).

$$\ln(h) = \ln(h_0) - \frac{Q}{R(T - T_0)} \quad (21)$$

Where R is the gas constant ($R = 8.3143 \text{ J/mol K}$) and h_0 is a proportionality factor. The slope in Figure 3 represents the mean activation energy, Q , of bulk water and give 18.8913 kJ/mol compared with the literature value of 18.8 kJ/mol [17]. The estimated value of $\ln(h_0)$ is -33.1045 or $h_0 = 4.1966 \cdot 10^{15} \text{ s}$. Obviously, equations (10) and (21) are quite effective to estimate some microwave parameters, particularly for pure Debye relaxation materials.

Table 4 The estimated values of relaxation time, h , static permittivity, χ_s and optical permittivity, χ_w , of bulk water at various temperatures, T

Temperature ($T \pm 1$) °C	Debye model for water		This work	
	Thrane, 1976	Kaatze et al., 1989	Eq (10)	Eq (11)
25	$h = 18.3766 \text{ ps}$ $\chi_s = 178.5198$ $\chi_w = 14.9$	$h = 18.28 \pm 0.02 \text{ ps}$ $\chi_s = 178.36 \pm 0.05$ $\chi_w = 15.4 \pm 0.2$	$h = 18.3651 \text{ ps}$ $\chi_s = 178.2684$	$h = 18.6841 \text{ ps}$ $\chi_w = 16.9363$
30	$h = 17.6118 \text{ ps}$ $\chi_s = 176.6836$ $\chi_w = 14.9$	$h = 17.31 \pm 0.05 \text{ ps}$ $\chi_s = 176.56 \pm 0.2$ $\chi_w = 15.2 \pm 0.3$	$h = 17.6007 \text{ ps}$ $\chi_s = 176.8199$	$h = 18.0579 \text{ ps}$ $\chi_w = 17.7771$
40	-	$h = 15.82 \pm 0.1 \text{ ps}$ $\chi_s = 173.18 \pm 0.2$ $\chi_w = 14.6 \pm 0.7$	$h = 16.2485 \text{ ps}$ $\chi_s = 174.0768$	$h = 16.7760 \text{ ps}$ $\chi_w = 17.0199$
50	-	$h = 14.75 \pm 0.1 \text{ ps}$ $\chi_s = 169.89 \pm 0.2$ $\chi_w = 14.0 \pm 0.5$	$h = 14.7514 \text{ ps}$ $\chi_s = 170.8523$	$h = 15.7936 \text{ ps}$ $\chi_w = 17.9968$
60	-	$h = 14.01 \pm 0.1 \text{ ps}$ $\chi_s = 166.70 \pm 0.2$ $\chi_w = 14.2 \pm 0.5$	$h = 13.7810 \text{ ps}$ $\chi_s = 168.1510$	$h = 15.6117 \text{ ps}$ $\chi_w = 112.691$

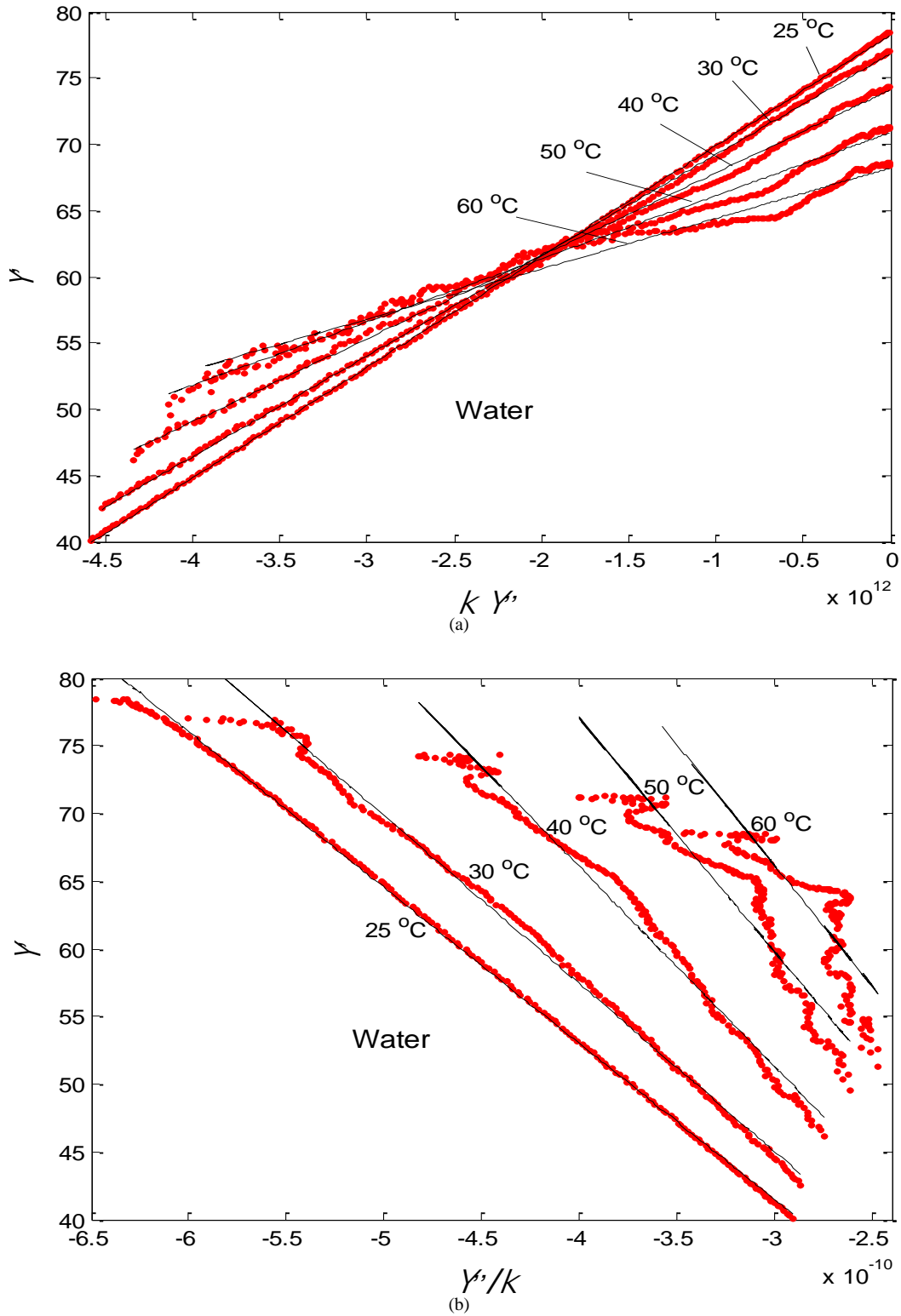


Figure 2 (a) (b) Variation in γ_r^t with γ_r^t , which plotting slope represents the relaxation time, η , of water at 25°C, 30°C, 40°C, 50°C and 60°C

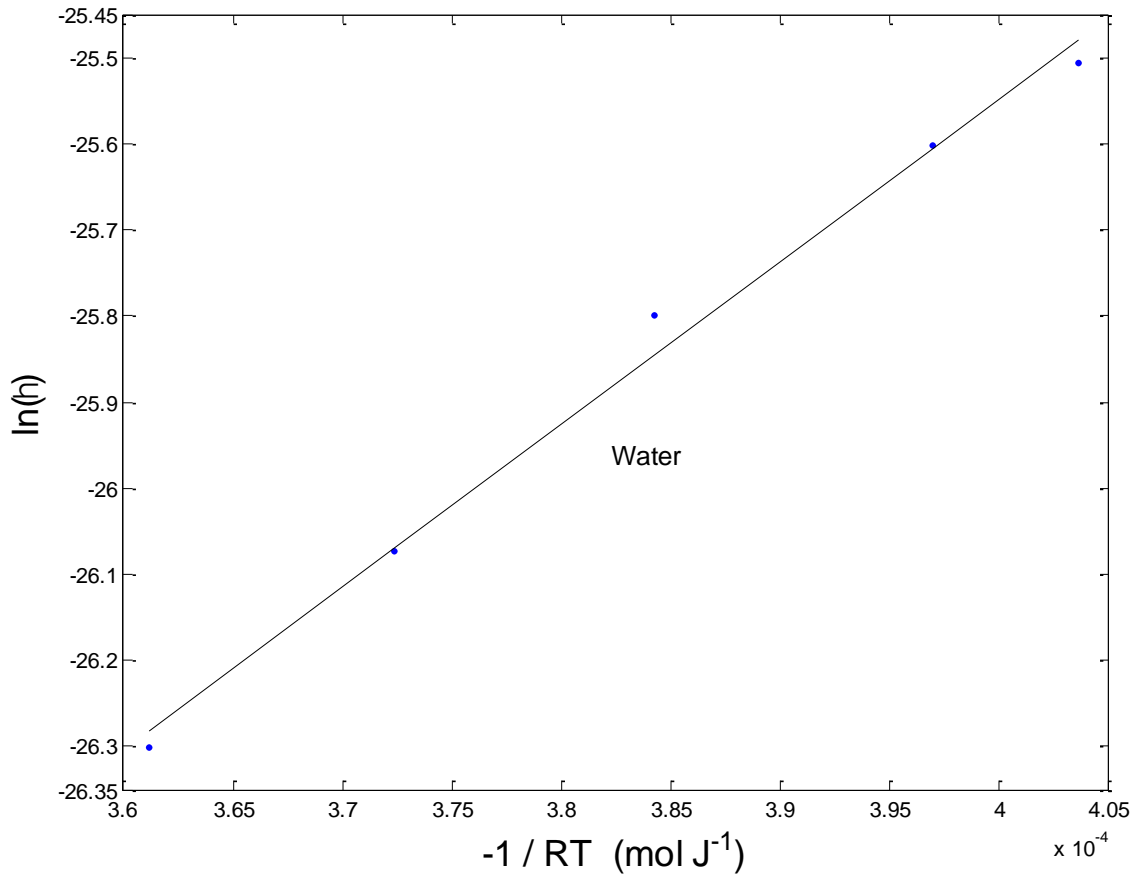


Figure 3 Arrhenius plots for bulk water

4.2 Estimation of Parameter γ_s Using Higher Order Polynomial Equation

The higher-order polynomial fitting of Debye models (15) and (16) was applied on pure water. There are 4, 5, and 6 order polynomial fitting that were tested and those polynomial coefficients are compared with the original Debye model [22] at $(25 \pm 1)^\circ\text{C}$ as tabulated in Table 5 and plotted in Figure 4. There are no typical rules to determine the order of polynomial to give the best fit with the experimental data. As long as the fitting line is matched with the experimental data, the order of polynomial is considered an appropriate choice. In our experience, the first few terms of coefficients are usually attributed to curve scope at low frequencies, and provide accurate values with mathematical calculation and conversely as shown in Table 5.

In this work, the dielectric constant, γ_r of water as a function of frequency, f and temperature, T , was formulated as equation (22) with the aid of equation (15):

$$\begin{aligned} \gamma_r = & 1 + 0.291531740265 T - 8.26619558508 \times 10^{-5} T^2 \\ & + 5.587521507307 \times 10^{-12} T^3 - 4.234240810053 \times 10^{-24} T^4 + 7.041326579374 \times 10^{-21} T^5 - 3.67162865847 \times 10^{-19} f^2 \\ & + 2.348175849507 \times 10^{-45} T^3 - 5.237541347645 \times 10^{-43} T^2 + 4.191922013075 \times 10^{-41} T + 1.160604385157 \times 10^{-139} f^4 \\ & + 6.503772847655 \times 10^{-66} T^3 - 1.089718588133 \times 10^{-63} T^2 + 6.179833452506 \times 10^{-62} T + 1.372700865828 \times 10^{-60} f^6 \end{aligned} \quad (22)$$

By substituting equations (21) and (22) into (10), yields

$$\gamma_r = \frac{E_0 \exp\left(-\frac{E_a}{RT}\right) \left[0.291531740265 T - 8.26619558508 \times 10^{-5} T^2 + 5.587521507307 \times 10^{-12} T^3 - 4.234240810053 \times 10^{-24} T^4 + 7.041326579374 \times 10^{-21} T^5 - 3.67162865847 \times 10^{-19} f^2 + 2.348175849507 \times 10^{-45} T^3 - 5.237541347645 \times 10^{-43} T^2 + 4.191922013075 \times 10^{-41} T + 1.160604385157 \times 10^{-139} f^4 + 6.503772847655 \times 10^{-66} T^3 - 1.089718588133 \times 10^{-63} T^2 + 6.179833452506 \times 10^{-62} T + 1.372700865828 \times 10^{-60} f^6 \right]}{E_0} \quad (23)$$

where T is the temperature (in $^\circ\text{C}$) and $\gamma_s = 1 + 0.291531740265 T - 8.26619558508 \times 10^{-5} T^2 + 5.587521507307 \times 10^{-12} T^3 - 4.234240810053 \times 10^{-24} T^4 + 7.041326579374 \times 10^{-21} T^5 - 3.67162865847 \times 10^{-19} f^2 + 2.348175849507 \times 10^{-45} T^3 - 5.237541347645 \times 10^{-43} T^2 + 4.191922013075 \times 10^{-41} T + 1.160604385157 \times 10^{-139} f^4 + 6.503772847655 \times 10^{-66} T^3 - 1.089718588133 \times 10^{-63} T^2 + 6.179833452506 \times 10^{-62} T + 1.372700865828 \times 10^{-60} f^6$. Nevertheless, equations (22) and (23) are only valid for $25^\circ\text{C} \leq T \leq 60^\circ\text{C}$ and $0.13 \text{ GHz} \leq f \leq 20 \text{ GHz}$. The comparison between equations (22) and (23) with measured data is illustrated in Figure 5.

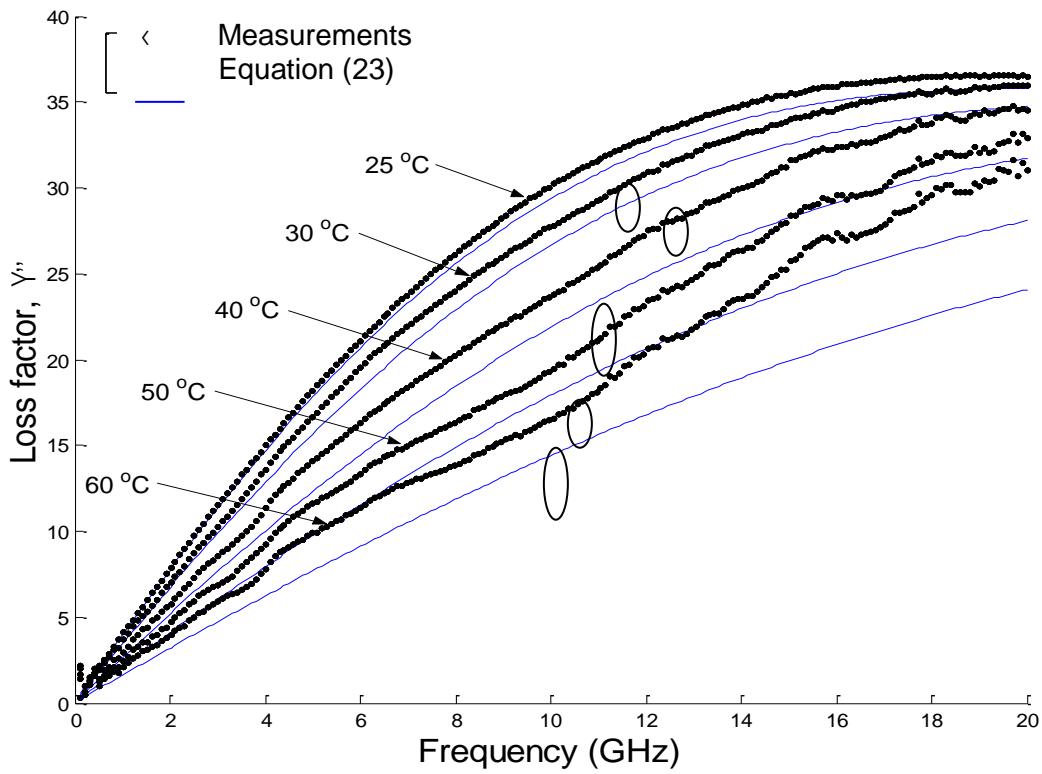
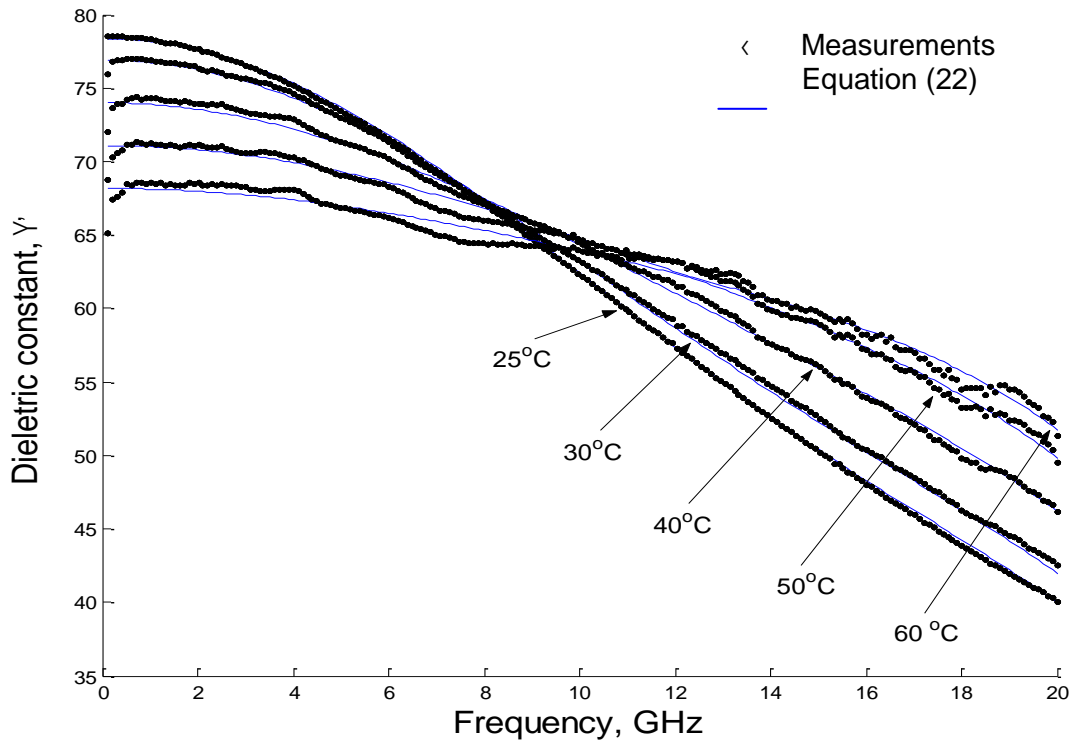


Figure 5 Variation in γ'_r and γ''_r with frequency for water at 25°C, 30°C, 40°C, 50°C and 60°C

Table 5 Comparison between the polynomial fitting approach and the Debye model for water at $(25 \pm 1)^\circ\text{C}$

Debye model for water at $(25 \pm 1)^\circ\text{C}$			
Literature (Thrane, 1976) $\gamma_w = 14.9; g = 10$ $h = 18.3766 \times 10^{12};$ $s = 1/\gamma_s \times \omega^2$	This work		
	Coefficients of 4 order polynomial fitting	Coefficients of 5 order polynomial fitting	Coefficients of 6 order polynomial fitting
$\gamma_s = 178.5198$	$\gamma_s = 178.4222$	$\gamma_s = 178.4714$	$\gamma_s = 178.4756$
$f_b \omega^2 = 8 f_l \omega^2 = 12.0393 \times 10^{19}$	$A_1 = 2.09533 \times 10^{19}$	$A_1 = 2.16143 \times 10^{19}$	$A_1 = 2.18826 \times 10^{19}$
$f_b \omega^4 = 8 f_l \omega^4 = 15.6492 \times 10^{40}$	$A_2 = 5.81889 \times 10^{40}$	$A_2 = 7.23914 \times 10^{40}$	$A_2 = 8.25943 \times 10^{40}$
$f_b \omega^6 = 8 f_l \omega^6 = 11.5649 \times 10^{60}$	$A_3 = 1.13358 \times 10^{60}$	$A_3 = 2.19206 \times 10^{60}$	$A_3 = 3.39696 \times 10^{60}$
$f_b \omega^8 = 8 f_l \omega^8 = 14.3349 \times 10^{81}$	$A_4 = 9.78302 \times 10^{82}$	$A_4 = 4.17349 \times 10^{81}$	$A_4 = 1.04033 \times 10^{80}$
$f_b \omega^{10} = 8 f_l \omega^{10} = 11.2008 \times 10^{101}$		$A_5 = 3.35478 \times 10^{102}$	$A_5 = 1.79994 \times 10^{101}$
$f_b \omega^{12} = 8 f_l \omega^{12} = 13.3263 \times 10^{122}$			$A_6 = 1.28057 \times 10^{122}$
$f_b \omega^2 = 8 f_l \omega^2 = 13.8747 \times 10^9$	$B_1 = 3.92047 \times 10^9$	$B_1 = 3.92160 \times 10^9$	$B_1 = 4.05296 \times 10^9$
$f_b \omega^3 = 8 f_l \omega^3 = 11.0733 \times 10^{29}$	$B_2 = 1.20988 \times 10^{29}$	$B_2 = 1.22504 \times 10^{29}$	$B_2 = 2.22363 \times 10^{29}$
$f_b \omega^5 = 8 f_l \omega^5 = 12.9733 \times 10^{50}$	$B_3 = 3.62133 \times 10^{50}$	$B_3 = 3.94714 \times 10^{50}$	$B_3 = 2.47755 \times 10^{49}$
$f_b \omega^7 = 8 f_l \omega^7 = 18.2362 \times 10^{71}$	$B_4 = 7.25554 \times 10^{71}$	$B_4 = 9.68369 \times 10^{71}$	$B_4 = 1.94559 \times 10^{69}$
$f_b \omega^9 = 8 f_l \omega^9 = 12.2815 \times 10^{91}$	$B_5 = 6.25084 \times 10^{92}$	$B_5 = 1.35806 \times 10^{91}$	$B_5 = 8.07950 \times 10^{90}$
$f_b \omega^{11} = 8 f_l \omega^{11} = 16.3200 \times 10^{112}$		$B_6 = 7.69585 \times 10^{113}$	$B_6 = 1.64109 \times 10^{110}$
$f_b \omega^{13} = 8 f_l \omega^{13} = 11.7507 \times 10^{132}$			$B_7 = 1.28868 \times 10^{131}$

4.3 Estimation of Parameters γ_s, γ_w , and Using Second Order Polynomial Equation

It is hard to determine the value of parameter, γ_w , especially for lower and multiple relaxation time materials. For lower single relaxation time materials, like water, this problem can be solved by using a greater measured data range or measuring the material at a sufficiently higher frequency. The size of the data range can be considered based on the uniformity distribution of measured data and parabola shape in the Cole-Cole plots (χ_r versus χ_r^2) as shown in Figure 6.

Equation (12) is an ideal concept, but in actual experiments, some molecules have much broader dispersion lines and their absorption maxima are distinctly below the ideal concept. In this case, the Cole-Cole characteristics [Equation (13)] are suggested. Obviously, the measured data of χ_r^2 as a function of χ_r^2 can be easily expressed by the second order of polynomial equation. This approach was applied on four standards of lossy liquids, propan-1-ol, ethanol, methanol and water. The quadratic fitting expression, based on measured data for each liquid is tabulated in Table 6.

Besides that, the Debye parameters can be found from the root values of equations (12) or (13). The maximum root value of equations (12) or (13) is represented as γ_s , while the minimum value is assumed as γ_w . The empirical constant, of the Cole-Cole model can also be estimated using (13) by comparing the calculated coefficient (P_1, P_2 and P_3) with corresponding fitting coefficient (a_1, a_2 and a_3) as shown in Table 8. The fitting coefficients values (a_1, a_2 and a_3) in Tables 6 and 7 were compared with the calculated values using (14a), (14b), and (14c), where the constant parameters (γ_s and γ_w) in those equations were obtained from the literature as listed in Table 7. The more complete the parabola shape in the Cole-Cole plots, the more accuracy there will be in the predicted parameters. The $q \times g \times t \times c \times n \times e \times q \times o \times r \times c \times t \times k \times u \times q \times p \times t \times g \times u \times w \times n \times v \times u \times q \times h \times v \times j$ for the four liquids are available in Table 7.

Another important factor in modeling is the uniformity or weight distribution of measured data. From Figure 6, it is evident that a higher weight of data occurs at higher frequency for propan-1-ol and ethanol as well as methanol. For propan-1-ol liquid, the measured data of χ_r^2 has a higher density of value between ~ 4 to ~ 5 for the frequency range 0.13 GHz to 20 GHz.

This condition may influence the tendency of polynomial fitting. In addition, the small variation in γ_r with the greatest frequency range will give a lower sensitivity. The phenomena may provide a large number of relative errors in modeling. Therefore, in this work, the fitting measured data acquired by

using formulas (15) and (11) for propan-1-ol and ethanol are 0.13 GHz - 1.5 GHz and 0.13 GHz - 3 GHz, respectively. For methanol, the data range 0.13 GHz - 68 GHz was utilized and the results are shown in Table 7.

Table 6 The fitting expressions of $f(\gamma_r, t) \epsilon^2$ as a function of γ_r for four standard lossy liquids at $(25 \pm 1)^\circ\text{C}$

Samples	Quadratic fitting between γ_r and γ_r $f(\gamma_r, t) \epsilon^2 = a_1 \gamma_r^2 + a_2 \gamma_r + a_3$
Propan-1-ol	$f(\gamma_r, t) \epsilon^2 = 0.78294794403552 \gamma_r^2 + 0.752800611883 \gamma_r + 0.50581728768$
Ethanol	$f(\gamma_r, t) \epsilon^2 = 0.91140405224695 \gamma_r^2 + 6.637788111836 \gamma_r + 97.000514651388$
Methanol	$f(\gamma_r, t) \epsilon^2 = 1.0661317094666 \gamma_r^2 + 2.092128800077 \gamma_r + 20.66792634657$
Water	$f(\gamma_r, t) \epsilon^2 = 0.95057011678464 \gamma_r^2 + 7.938401661636 \gamma_r + 262.09787960974$

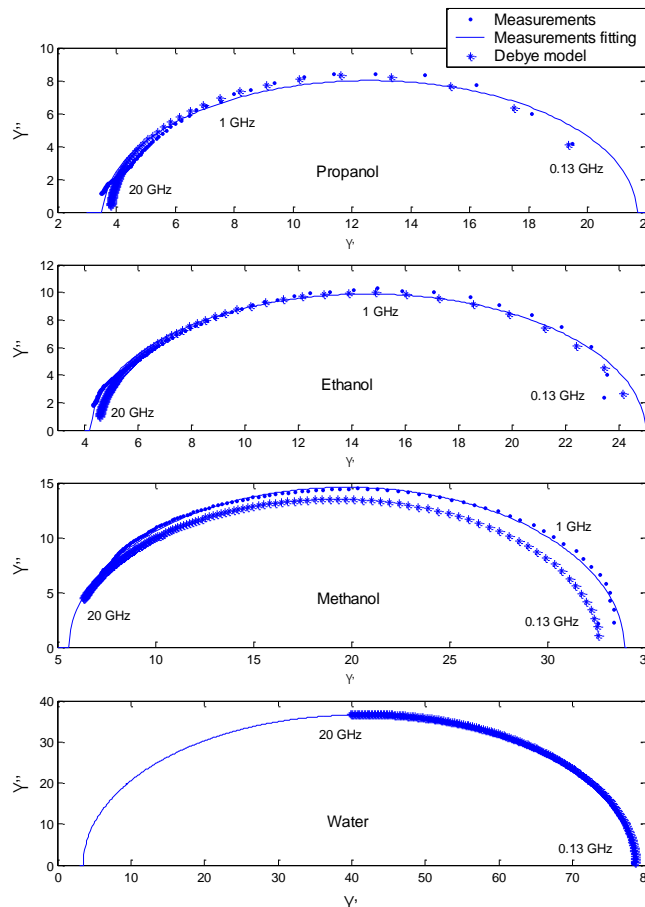


Figure 6 Cole-Cole plots for four standard lossy liquids for a frequency range of 0.13 - 20 GHz at room temperature $(25 \pm 1)^\circ\text{C}$

Table 7 The comparison of the fitting parameters for the Debye model and the roots of the 2nd order equation (13) for Propan-1-ol, Ethanol, Methanol, and Water.

Materials	Roots of 2 nd order Eq (13) [using Table 6]	Fitting [using Eq (15)]	Fitting [using Eq (11)]	Debye model		Fitting coefficient [using Table 6]
Propan-1-ol	$\gamma_s = 21.66109$ $\gamma_w = 3.567664$	$\gamma_s = 20.141$	$\gamma_s = 20.142$ $h = 268.93$ ps	$\gamma_s = 20.44$ $\gamma_w = 3.8$ $h = 320.685$ ps (Grant et al., 1989)	$\gamma_s = 77.67$ $\gamma_w = 124.24$	$a_3/a_1 = 77.28$ $a_2/a_1 = 25.23$
Ethanol	$\gamma_s = 24.96384$ $\gamma_w = 4.263356$	$\gamma_s = 24.052$	$\gamma_s = 24.336$ $h = 141.54$ ps	$\gamma_s = 24.5$ $\gamma_w = 4.52$ $h = 165.442$ ps (Grant et al., 1989)	$\gamma_s = 110.7$ $\gamma_w = 129.02$	$a_3/a_1 = 106.4$ $a_2/a_1 = 29.23$
Methanol	$\gamma_s = 33.86837$ $\gamma_w = 5.612804$	$\gamma_s = 33.593$	$\gamma_s = 33.859$ $h = 49.091$ ps	$\gamma_s = 32.616$; $\gamma_w = 5.5849$ $h = 47.4451$ ps (Using NBS data)	$\gamma_s = 182.2$ $\gamma_w = 138.2$	$a_3/a_1 = 191.0$ $a_2/a_1 = 39.67$
Water	$\gamma_s = 78.47778$ $\gamma_w = 3.513441$ $h = 8.3651$ ps	$\gamma_s = 78.476$	$\gamma_s = 78.276$	$\gamma_s = 78.5198$; $\gamma_w = 4.9$ $h = 8.3766$ ps (Thrane, 1976)	$\gamma_s = 384.7$ $\gamma_w = 183.42$	$a_3/a_1 = 275.7$ $a_2/a_1 = 81.99$

Table 8 The estimated empirical constant, N of the Cole-Cole model using (13) by comparing the calculated coefficient (P_1 , P_2 and P_3) with the corresponding fitting coefficient (a_1 , a_2 and a_3)

Materials	Estimated values of	Calculated values of Coefficients in Equation (13)		Fitting values of Coefficients (in Table 7)
		Using ω and s from literatures (in Table 7)	Using ω and s from roots of Eq (13) (in Table 7)	
Propan-1-ol	0.912	$P_1 = 0.7823$ $P_2 = 177.67$ $P_3 = 177.67$	$P_1 = 0.7823$ $P_2 = 177.28$ $P_3 = 177.28$	$a_1 = 0.7830$ $a_3/a_1 = 177.28$
Ethanol	0.969	$P_1 = 0.9112$ $P_2 = 110.74$ $P_3 = 110.74$	$P_1 = 0.9112$ $P_2 = 110.643$ $P_3 = 110.643$	$a_1 = 0.9114$ $a_3/a_1 = 110.64$
Methanol	1	$P_1 = 1$ $P_2 = 1182.16$ $P_3 = 1182.16$	$P_1 = 1$ $P_2 = 1190.10$ $P_3 = 1190.10$	$a_1 = 1.0611$ $a_3/a_1 = 1191.0$

5.0 CONCLUSION

The approximated series of the dielectric model provides more degrees of freedom to specify the broadness of experimental relaxation peaks. The polynomial approach is implemented to reduce the difficulty of finding q , w , v , n , v , j , g , n , f , k , g , n , g , e , v , t , k , e , θ , u , r , c , t , c , o , g , v , g , t , u , 0 . In this paper, a polynomial fitting was used to estimate the Debye or Cole-Cole parameters. In this work, four types of methods were suggested:

- Using an N -order polynomial fitting, equation (15) was used to estimate the value of Y_s . This procedure is applied particularly on the multiple dispersion lossy materials.
- Applying the linear polynomial fitting of equation (10) to estimate the values of Y_s and h . This method can be used for both single and multiple dispersion lossy materials.
- Rooting the second order polynomial fitting of equations (12) and (13) to estimate values of Y_s and ω , as well as determine the empirical constant, N from the coefficients of (13). The techniques are quite accurate for single dispersion lossy materials with non-Debye characterization.
- Determining the values of Y_s and ω from the coefficients of second order polynomial equation (13). This rule is only effective for single dispersion lossy materials with pure or approximated pure Debye characterization.
- The corresponding values of empirical parameters between the Kohlrausch-Williams-Watts (KWW) and Havriliak-Negami (HN) functions were re-expressed in polynomial form.

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