Microwave Processing and Permittivity Measurement of Thermoplastic Composites at Elevated Temperature

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Abstract

The material properties of greatest importance in microwave processing of a dielectric are the complex relative permittivity $\varepsilon = \varepsilon' - j\varepsilon''$, and the loss tangent, $\tan\delta = \varepsilon''/\varepsilon'$. The real part of the permittivity, ε', sometimes called the dielectric constant, mostly determines how much of the incident energy is reflected at the air-sample interface, and how much enters the sample. This paper shows that the reflection coefficient of a material, $\rho \approx -\frac{(\sqrt{\varepsilon'}-1)}{(\sqrt{\varepsilon'}+1)}$ and the depth of penetration of a dielectric, $D \approx \frac{2}{\omega\sqrt{\mu_0\varepsilon_0\varepsilon'}\tan\delta}$. Therefore the larger the value of the real part of the complex permittivity, the more the incident energy will be reflected by a dielectric but the energy that enters the material will penetrate further than in a dielectric with the same ε'' but lower ε'. However, the most important property in microwave processing is the dielectric loss, ε'' which predicts the ability of the material to convert the penetrating energy into heat. Measurements of ε' and ε'' are therefore critical in the microwave processing of materials with or without primer. The dielectric constant, ε', dielectric loss, ε'', and hence complex relative permittivity, ε and loss tangent, $\tan\delta$, of some commonly used thermoplastics have been measured[1,2,3] at various temperatures and frequencies. These results may be used to determine whether various fibre-reinforced thermoplastic (FRTP) composites are suitable for microwave processing. This paper describes a convenient laboratory based method to obtain ε', ε'' and hence $\tan\delta$. The method employs a network analyser together with a waveguide transmission technique chosen because it provides the widest possible frequency range with high accuracy; the hardware and software of the method is also readily available in the electronic laboratory of the University of Southern Queensland. The required data were collected at a range of elevated temperatures and over a band of frequencies.

Keywords : fibre reinforced thermoplastic composites (FRTP), dielectric constant and dielectric loss.

Introduction

The use of microwave high energy rate joining of FRTP composites has shown promising preliminary results[6,7] and more research is being carried out so that the technology can find its application in manufacturing industries shortly. The ε and the $\tan\delta$ of FRTP composites have rarely been reported in the literature and then at room temperature only. As these electrical properties vary significantly with rising temperature and frequency, values attained at room temperature may not be used to predict the microwave-reactiveness of thermoplastic composites at elevated temperatures. This paper starts by showing the relationship between the depth of penetration of microwave energy into a material under test (MUT) and its complex relative permittivity and loss tangent. It also describes the measurement of ε and $\tan\delta$ of three types of random, 33% by weight, glass-fibre reinforced thermoplastic composites. The three thermoplastic matrices chosen for the study are the three most commonly used thermoplastics, namely, low density polyethylene (LDPE), polystyrene (PS) and nylon 66. The limited heat resistance of the network analyser prevents measurements of the complex relative permittivity being taken at temperatures higher than 100°C. The frequencies selected vary from 2.2 GHz to 12.5 GHz, which cover most industrial microwave applications.

A waveguide transmission technique is a convenient laboratory based method which employs a network analyser to obtain the ε of a length of sample filled waveguide. The ε value was calculated at spot frequencies off-line from manual readout of transmission coefficient, S_{21}, data from the network analyser. Since the transcendental equation used in the off-line calculation had multiple solutions, the broadband S_{21} data was
used as a check on the calculated ε values. From this measurement the sample dielectric constant, dielectric loss and loss tangent were calculated.

Reflection Coefficient and Depth of Penetration

The sample materials are all non-conductive and have small loss tangents. When an electromagnetic wave is incident on such a material, a proportion of the energy is reflected at the air-sample interface. The remainder of the wave enters the sample but is progressively attenuated by the polarisation losses which are characterised by the dielectric loss factor, ε''. The reflection coefficient at the air-sample interface, the depth of penetration of the transmitted wave into the surface of the sample, and the loss tangent together determine the suitability of the materials for microwave processing. For a time harmonic wave, the electric field strength within the sample will be $E_o\exp(-\gamma z)$

where z is the distance below the surface;

E_o is the surface value and

γ is the propagation coefficient. The latter is a complex number such that

$$\gamma = \alpha + j\beta \tag{1}$$

where the real part is the attenuation coefficient and the imaginary part is the phase coefficient. Since the sample materials are all non-conductive the propagation coefficient for a plane wave from Maxwells equations [8,9]:

$$\gamma = j\omega\sqrt{\mu_o\varepsilon_0(\varepsilon' - j\varepsilon'')} \tag{2}$$

where ω is the radian frequency;

μ_o is the permeability of free space;

ε_o is the permittivity of free space;

ε' is the dielectric constant and

ε'' is the dielectric loss factor.

The reflection coefficient at the air-sample interface depends on the ratio of the intrinsic impedance of the sample material to that of air. The intrinsic impedance of the sample is :

$$Z_{os} = \frac{j\omega\mu_o}{\gamma} = \frac{\sqrt{\mu_o}}{\sqrt{\varepsilon_o}}\frac{1}{\sqrt{\varepsilon' - j\varepsilon''}} \tag{3}$$

whereas that for air is simply :

$$Z_{os} = \sqrt{\mu_o / \varepsilon_o} \tag{4}$$

The reflection coefficient is then

$$\rho = \frac{Z_{os} - Z_o}{Z_{os} + Z_o} \tag{5}$$

For these materials the loss factor is small ie. $\varepsilon'' << \varepsilon'$ so that to a first approximation :

$$\rho = \frac{-(\sqrt{\varepsilon'} - 1)}{(\sqrt{\varepsilon'} + 1)} \tag{6}$$

The square of the magnitude of this reflection coefficient determines the fraction of microwave energy which is reflected at the air-sample interface. Typically, for thermoplastics $2 < \varepsilon' < 3$, in which case the proportion of energy reflected will vary from 3% to 7%. Materials with a larger dielectric constant will reflect more energy. For the wave which does enter the sample, the field strength amplitudes fall off with distance z into the material as exp($-\alpha z$), where α is the attenuation coefficient introduced earlier. Squaring (2), separating out the real and imaginary parts and solving for the latter :

$$\alpha^2 = \frac{1}{2}\omega^2\mu_o\varepsilon_o\varepsilon'[\sqrt{1 + (\tan\delta)^2} - 1] \tag{7}$$

where $\tan\delta = \varepsilon''/\varepsilon'$ (8)

For these materials the loss tangent is much less than unity, so after using the Binomial Theorem and neglecting higher powers of the loss tangent :

$$\alpha = \frac{1}{2}\omega\sqrt{\mu_o\varepsilon_o\varepsilon'}\tan\delta \tag{9}$$

A more informative expression is obtained if this is expressed in terms of a wavelength :

$$\alpha = \frac{\pi\sqrt{\varepsilon'}}{\lambda_o}\tan\delta \tag{10}$$

where λ_o is the wavelength in air or free space :

$$\lambda_o = 29.98/f(\text{GHz})\ \text{cm} \tag{11}$$

The skin depth is defined as the depth into the material at which the field strength has fallen to (1/e) times its initial value. Hence the depth of penetration is the reciprocal of the attenuation coefficient :

$$D = \frac{\lambda_o}{\pi\sqrt{\varepsilon'}}\frac{1}{\tan\delta} = \frac{\lambda_o\sqrt{\varepsilon'}}{\pi\varepsilon''} \tag{12}$$

If the loss tangent is small, the penetration depth will be many wavelengths, which means that the field strength within the sample will be fairly uniform. Finally, the electromagnetic energy converted into heat per unit volume per second is

$$P = \frac{1}{2}\omega\varepsilon_o\varepsilon''|E|^2 \tag{13}$$

where $|E|$ is the magnitude of the electric field intensity.

Hence the loss factor and loss tangent are critical in determining the suitability of the material for microwave processing. The dielectric constant is also significant since it determines the proportion of power reflected from the sample.

Calibration of the Network Analyser

The purpose of a vector network analyser is to approximate a perfect measurement system having infinite dynamic range, isolation, and directivity characteristics, no impedance mismatches in any part of the test set-up, with flat frequency response. The 'perfect' network analyser is achieved by measuring the magnitude and phase response of known standard devices, using this data in conjunction with a model of the measurement system to determine error contributions, then measuring a test device and using vector mathematics to compute the actual test device response by removing the error terms. The first step in using the equipment is to input the range of frequencies over which ε is to be measured and to calibrate[10] the equipment itself. Two ports, port 1 and port 2, of the equipment have to be calibrated using three types of termination, namely, short circuit, offset short circuit and matched load. The network analyser was then instructed to calculate the reflection coefficients from the calibration data. The next stage was to connect to the two waveguide to coaxial adaptors together so that the forward and backward transmission coefficients could be calculated. The reflection and transmission coefficients were then saved in a register for later retrieval.

Theory of Waveguide Transmission Technique

The idea of calibration was to get the values of reflection and transmission coefficients with air filled waveguide as standard and then compare with those of a sample filled waveguide. The calibration data was temperature dependent and hence a fresh calibration was required at every temperature. During measurements, fixed lengths of waveguide were filled with the FRTP samples. A range of waveguide sizes was required to provide an almost continous spectrum of frequencies from 2.2 - 12.5 GHz. The transmission coefficient between the reference planes is[11] :

$$S_{12} = \frac{4\Gamma_1\Gamma_0}{(\Gamma_1+\Gamma_0)^2 \exp^{\Gamma_1 L} - (\Gamma_1-\Gamma_0)^2 \exp^{-\Gamma_1 L}} \quad (14)$$

where $\Gamma_0 = j\beta_0$, the propagation coefficient in air filled waveguide

$\Gamma_1 = \alpha_1 + j\beta_1$, the propagation coefficient in sample filled waveguide

L is the sample length in mm.

The reasons for using a range of waveguide sizes and different values of L for each waveguide size would be discussed later. The relative permittivity of the FRTP[11] is :

$$\varepsilon = \varepsilon' - j\varepsilon'' = \varepsilon' (1 - j\tan\delta) \quad (15)$$

For non-magnetic materials the elements of the complex permittivity may be obtained from the propagation coefficient as follows[11] :

$$\varepsilon = [1 + (\beta_1^2 - \alpha_1^2)(a/\pi)^2](c/2af)^2 \quad (16)$$

$$\tan\delta = \alpha_1\beta_1 c^2/(2\pi^2 f^2 \varepsilon') \quad (17)$$

where 'a' is the broad dimension of the waveguide and 'c' is the velocity of electromagnetic waves. Since S_{12} is known from measurement, in principle (14) can be solved for Γ_1, from which the complex permittivity of the sample may be calculated. Sabburg[11], Ness[12] and Ball[13] detailed an iterative technique for getting very accurate estimates of ε′. Very accurate estimates of ε′ may be obtained by noting consecutive frequencies at which the transmission coefficient is a multiple of 90^o. At these frequencies $\beta_1 L \approx n\pi$, where n is the number of half wavelengths in the sample, n = 1/2, 1, 1.5 etc. Substitution into (16) gives Ness's initial estimate[12] :

$$\varepsilon' = [1+ (an/L)^2](150/af)^2 \quad (18)$$

where a and L are in mm and f in GHz. As frequency increases n must increase by one half for each 90^o change in transmission coefficient phase so that it is an even integer at 0^o and an odd integer at 180^o phase angle and it will take on consistent values at consecutive frequencies. If two such frequencies are available within the measurement frequency range and assuming that ε′ is not changing rapidly with frequency (18) yields :

$$[1+ (an_1/L)^2]f_2^2 = [1+ (an_2/L)^2]f_1^2 \quad (19)$$

Imposing the appropriate relationship between n_1 and n_2, the result is a quadratic equation which gives an estimate of either one of these numbers. Rounding the solutions to the nearest odd or even integer or integer plus one half as appropriate, a reliable value of n is obtained, with the aid of which ε′ may be estimated from (18); the initial estimate of tan δ is much less critical and Ness[6] suggestion is adequate. The initial estimates of ε′ and tan δ are used to calculate Γ_1 and S_{21}. The calculated values of S_{21} were compared with the measured ones and ε′ adjusted until the phase angles agree. After that tan δ is adjusted until the magnitudes match. The cycle is repeated until error is minimum. The process was cumbersome and tedious and computer software based on Newton's method was therefore developed to speed up the procedure. The software requested the input of values of transmission coefficients and of those of three frequency markers[7], as well as the broadside waveguide dimension for the initial estimate of ε′ for the sample and the sample length, L. An accurate initial estimate of ε′ is important because it enables the iteration process to converge to the required result from the infinite number of solutions. Sometimes, the software failed to estimate the initial value of ε′ and the programme did not converge. In this situation, a manual estimate was required and this was achieved by adding or deducting a small value, depending on the fibre reinforcement, to the values of the ε′ of the unreinforced thermoplastic. For example, if the ε′ value of low density polyethylene (LDPE) is 2.25 then a small value, say, 0.25, is added to it to make the ε′ value of 33% glass fibre reinforced low density polyethylene LDPE/GF(33%) 2.5 which is then input to the programme. The software then calculated the values of ε′ at a range of frequencies dedicated to that particular type of waveguide.

Waveguide Sizes and Sample Lengths

In order to cover the frequency range from 2.2 to 12.5 GHz, four different sizes of waveguides were used, namely : WR90, WR159, WR229 and WR340. Taking waveguide WR340 as an example, the recommended range of frequencies[14] for dominant (TM_{10}) mode propagation was from 2.2 to 3.3 GHz. The wavelength in the sample depends on the sample relative permittivity or dielectric constant, ε′, as[9] :

$$\lambda_g = \frac{\lambda_o}{\sqrt{\varepsilon'}} \frac{1}{\sqrt{1-(\frac{\lambda_o}{2a\sqrt{\varepsilon'}})^2}} \quad (20)$$

where λ_o is the free space wavelength;
and 'a' is the broad side waveguide dimension.
For each waveguide size, the sample length should be selected so that it is at least three quarters of a wavelength long at the highest frequency of operation. This requires prior knowledge of the approximate value of the dielectric constant.

Results of Waveguide Transmission Technique

In order to verify that the method is accurate, materials with known dielectric constant values were measured; air and teflon were chosen for this purpose and their dielectric constants were measured at room temperature. Figure 1 shows that dielectric constants for air and teflon were one and two respectively at room temperature. This coincides very well with data in the literature,[15,16] and verifies the equipment performance. The dielectric loss for the two materials could not be reliably measured as negative values were recorded; their losses were too low. The loss through the waveguide wall was of the same order as the loss of the materials and some compensation should be included into the calculation programme to generate reasonable results. Figure 1 also illustrates the dielectric constants of nylon 66 , low density polyethylene (LDPE), glass fibre-reinforced LDPE and glass fibre-reinforced nylon 66. The dielectric constants and loss of acrylic at room temperature (25^{o}C) and at a frequency 3 GHz measured by the set-up matched the published data[15,16] being 2.6 and 0.015 respectively.

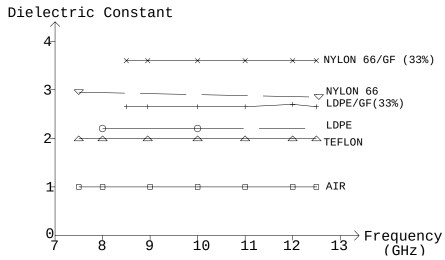

Figure 1. : Dielectric Constants of Air, Teflon, LDPE/GF(33%), Nylon 66/GF(33%), Nylon 66 at Room Temperature at Microwave Frequencies.

The first material under test was 33% glass-fibre reinforced low density polyethylene [LDPE/GF(33%)]. Figure 2 shows the ε′ values of LDPE/GF(33%) over a range of frequencies and at temperatures varying from 25^{o}C to 95^{o}C. As there was no published data for the values of ε′ of LDPE/GF(33%), simulated data of the material was used to verify the authenticity of the measured data. The values[15, 16] of ε′ of LDPE (2.25) and glass fibre (3.78) at 25^{o}C and at 3 GHz were employed to generate the ε′ of LDPE/GF(33%) at the same physical conditions. It was assumed that the mixture was homogeneous and the ε′ of the material could be calculated by proportion as follows: 2.25 x 67/100 + 3.78 x 33/100 = 2.755.
The data measured by the technique at the same

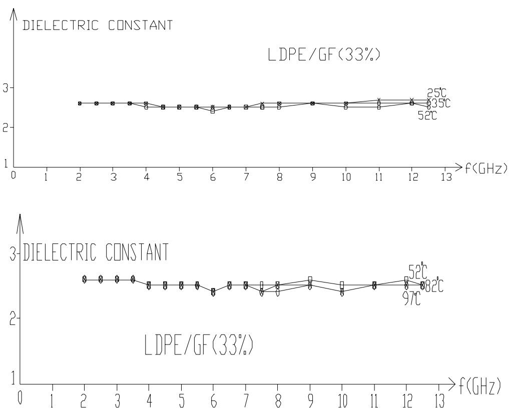

Figure 2. : Values of Dielectric Constant of LDPE/GF(33%) Against Frequencies (2 - 12.5 Ghz).

physical condition was 2.6. By similar arguement, the ε″ could also be simulated and it was found to be 0.0018 and LDPE/GF(33%) should therefore be a low loss material and its loss tangent was not able to be measured accurately by this method.
The next material to be studied was PS/GF(33%). Figure 3 depicts that the ε′ values of PS/GF(33%) do not change much with frequency and temperature but the ε′ values tend to be lower at higher temperatures, especially towards the higher frequency end. ε′ and ε″ of PS/GF(33%) were similarly simulated from published data[17] and they were found to be 2.92 and 0.0021 respectively at 25^{o}C and at 3 GHz. On the other hand, the measured values at the same conditions were 2.90 and 0.0136 respectively. The measured value of ε′ was very near to the simulated one and seemed to be reasonably accurate ; while, the measured value of ε″ was much higher than the simulated one (6.5 times) and it was 7.6 times higher than that of the simulated LDPE/GF(33%). Since the ε″ of PS was low and was very near to that of LDPE, it was therefore believed that the values obtained were unreliable and needed further investigation and study.

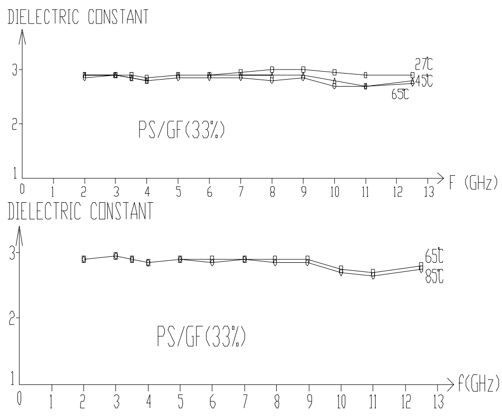

Figure 3. : Values of Dielectric Constant of PS/GF(33%) Against Frequencies (2 - 12.5 GHz) .

The last material to be discussed is nylon 66/GF(33%) where the higher the values of the temperature and frequency , the lower the values of the dielectric constant as depicted in figures 4 and 5 respectively. The simulated value of ε′ for nylon 66/GF(33%) at 10 GHz and at 25^{o}C is 3.27 while that procured by measurement is 3.71; the measured value is 12% higher. The trends of the values of the measured dielectric constant for the three materials were the same and they matched with the trend for most materials eg water[17]. Figures 6 and 7 illustrate the change of loss tangent values for nylon66/GF(33%) with frequencies and temperatures respectively. The simulated value of tan δ of nylon

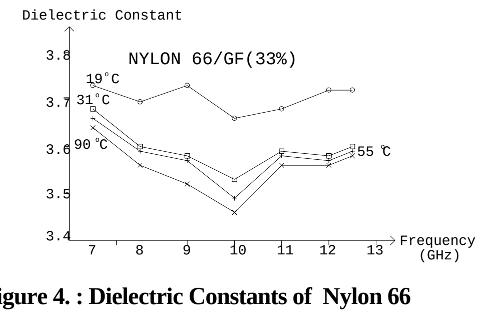

Figure 4. : Dielectric Constants of Nylon 66 /GF(33%)at Elevated Temperatures over a Certain Frequencies.

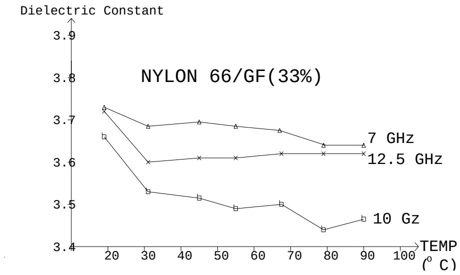

Figure 5. :Dielectric Constants of Nylon66 /GF(33%) at Different Frequencies over a Range of Temperatures.

66/GF(33%) at the same ambient conditions is 0.0072 and the measured one is 0.0149; the measured value is 52% larger. The trend shows that the higher the temperature the higher the values of the dielectric loss factor but there is no such a trend with frequency. Since nylon 66 is a relatively higher loss material, it was reckoned that the values of the loss tangents obtained were reliable. Referring to figure 7, at a frequency of 7 GHz and at a temperature of 90 degrees Celcius, the dielectric loss factor is at maximum. At this point, the maximum amount of microwave energy penetrated is converted to heat to facilitate the joining process but on the other hand, the dielectric constant was quite high and microwave energy penetration to the material is not too good. A compromise value of tan δ and ε' should therefore be chosen to obtain the greatest microwave energy penetration and maximum energy conversion to heat.

Conclusion

From the measured data of LDPE/GF(33%), PS/GF(33%) and nylon 66/GF(33%), it is found that at higher temperatures and higher frequencies, the materials become less reflective and permit

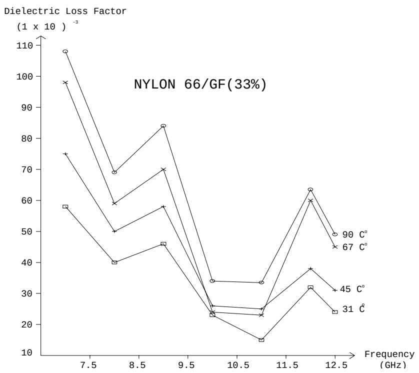

Figure 6. : Dielectric Loss Factors of Nylon 66/ 66/ GF(33%) at Different Frequencies over a Frequencies.

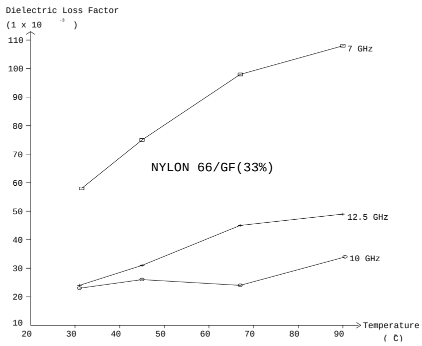

Figure7. : Dielectric Loss Factors of Nylon GF(33%) at Elevated Temperatures over Certain Range of Temperatures.

more microwave energy to enter; at the same time, their dielectric loss factors increase with temperature and frequency and enable more of the absorbed energy to be converted to heat. Therefore, within limits, higher temperatures and higher frequencies are more suitable for microwave-assisted joining of the three materials. The results implied that in microwave processing of thermoplastic composites, higher frequencies should be employed and that the temperature of the welds of the materials should increase evenly and gradually with processing time.

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