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Dielectric behavior of a sintered heterogeneous ternary composite resin/BT/Cu₂O

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Abstract. In this paper, we investigate and model the dielectric behavior of a ternary composite prepared at room temperature with a mixture of epoxy resin (RE), barium titanate (BT) and copper oxide (Cu₂O), sintered at three different temperatures (150°C, 200°C, and 250°C). Time domain spectroscopy (TDS) is used to characterize samples in the range [DC to 2 GHz] by performing a particular study at low frequency (500 MHz). The latter focused on both the sintering and the Cu₂O addition effects on a ternary composite dielectric behavior. These effects were quantified as a function of the BT volume fraction. For this purpose, we used an optimization method based on nonlinear regressions to determine the permittivity, to minimize systematic errors of this dielectric parameter, and to show the effect of Cu₂O on it. Moreover, we attempt to explain the sintering temperature effect on this kind of mixtures through the modified Lichtenecker model. As a matter of fact, the importance of this law is allocated on one hand to the validation and concordance of the experimental results with those of the theory and on the other hand to the temperature effect investigation on the form factor given by the modified Lichtenecker law.

1 Introduction

The combination of different materials in a polymer matrix offers the opportunity to obtain optimal properties [1–4]. In this context, various laws and formalisms have been proposed and used in order to design new materials, their behaviors analysis is necessary [5–11,14,15]. The possibility of processing such heterostructure by using the real parameters, as well as the influence of the inhomogeneities on the electric field behavior in many electronic applications is very essential. The effective permittivity of composites, describing the relationships between the microscopic and macroscopic properties of composites, has been studied through numerous analytical and theoretical formulas, called mixing laws [4,5,12,13]. From a theoretical point of view, the dielectric behavior of either binary or ternary composites was modeled by the modified Lichtenecker law. This law has confirmed its efficiency while taking into account the form factor for the prediction of multiphase permittivity random mixtures [4–6,12,13]. Based on our previous work [5–8], we have also extended our study to highlight the sintering effect on the composite behavior.

In this work, the main element is a composite made of the epoxy resin (RE) as a matrix, barium titanate (BT) and copper oxide (Cu₂O) powders as fillers, and sintered at

different temperatures. The dielectric response of the samples has been studied over a wide range of frequencies up to 2 GHz in order to evaluate their dielectric nature (ϵ , σ) for electronic and microelectronic applications in the high-frequency range. So that, we focused on the effect of Cu₂O and the impact of the sintering temperature variation on the dielectric properties of ternary composites. In parallel to the experimental study and aiming to obtain better performance in predicting the composite dielectric properties, we attempt to investigate the temperature effect that can affect the shape factor of the modified Lichtenecker Law.

2 Experimental protocol

2.1 Samples preparation

The preparation of the specimens is systematically accomplished by the use of a manufacturing protocol that makes it possible to get materials with acceptable and reproducible dielectric characteristics [4–9,12,13]. The method of making the samples consists of mixing the epoxy resin in a liquid state with volume quantities of BT and Cu₂O. These are provided in the powder form with a high purity of 99.98% and 99.99% respectively and with a grain size of less than 3 μm . Several samples were prepared for this study with a 70% fixed volume fraction of the epoxy resin, while the volume fraction of Cu₂O and BT are complementary and varying from 0% to 30% by a step of

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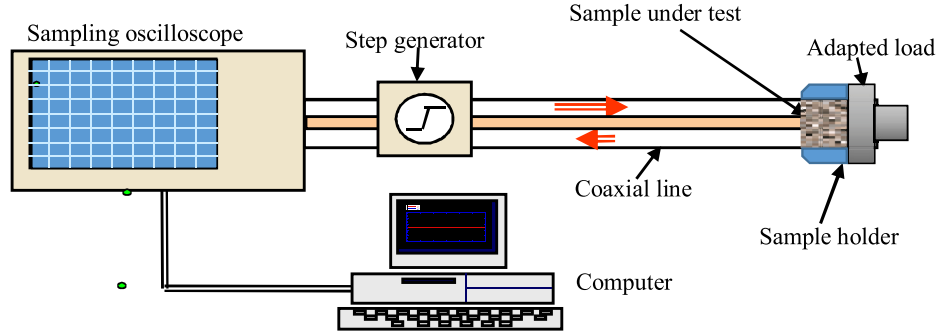


Fig. 1. Measurement setup (TDS).

10%. All samples were dried at room temperature of 27 °C during 48 hours and at an atmospheric pressure in order to improve hardness and to ease sintering at different temperatures (150 °C, 200 °C, and 250 °C) for 30 min. The samples produced are adjusted to the dimensions of the measuring cell used (APC7) and the adapted line method as well.

2.2 Experimental measurement system

The experimental values of the complex permittivity are obtained from measurements made with a time domain reflectometry (TDR) system composed of a very fast HP 54121 pulse generator with a 200 mV amplitude signal and a rise time of 35 ps, and a sampling oscilloscope (HP 84120B). The wave propagates in a standard coaxial line APC7 with a characteristic impedance of 50 Ω. As seen in Figure 1, the sample under test (SUT) of a “ d ” thickness is placed in the measuring cell ended by an adapted load (50 Ω).

2.3 Experimental procedure

The experimental method used is that of the adapted load [4,5,12,13] whose measuring system configuration is shown in Figure 1. This method consists of a processing of the time signal, which is acquired precisely from the TDR system response of the SUT. The first variable on which all calculations are made is the reflection coefficient $\Gamma^*(\omega)$. This is deduced from the Fourier transform applied to the experimental data. This parameter is in fact, the ratio of the signal reflected $V^-(\omega)$ to the incident signal $V^+(\omega)$ and noted in the frequency domain as:

$$\Gamma^*(\omega) = \frac{V^-(\omega)}{V^+(\omega)}, \quad (1)$$

where $V^+(\omega)$ and $V^-(\omega)$ are extracted from the response $V^+(t)$ and $V^-(t)$ of the measured sample.

The complex dielectric permittivity gives us information on both, the dielectric and the ohmic contribution of the treated material. It is deduced from the admittance ratio expressed as follows:

$$\frac{Y_{in}}{Y_0} = \frac{\sqrt{\epsilon^*} \cdot \tanh(s) + 1}{1 + j \frac{\omega d \tanh(s)}{c \cdot s}}, \quad (2)$$

where “ s ” is expressed as $s = j \frac{\omega d}{c} \sqrt{\epsilon^*}$ and “ ω ” is the angular frequency, “ d ” is the thickness of the sample and “ c ” is the velocity of the light in the vacuum. Y_{in} and Y_0 are respectively the input admittance and the characteristic admittance. The value of Y_{in} can be obtained from the following expression:

$$Y_{in} = \frac{Y_{oc} + Y_0}{1 + Z_{sc} \cdot Y_0}, \quad (3)$$

Y_{oc} and Z_{sc} represent respectively the admittance of the open line and the impedance of the short circuit. The function $(\tanh(s)/s)$ can be evaluated using various numerical techniques [4,5,12,13]. It effectively contributed to solving the transcendental equation (2) in the complex plane, which providing different solutions. The complex permittivity can be expressed from the Debye model by equation (4) where the parameters “ τ ”, “ ϵ_s ”, “ σ ”, and “ ϵ_∞ ” which are respectively the relaxation time, the static permittivity, the static conductivity and the permittivity at the very high frequency.

$$\epsilon^* = \epsilon' - j\epsilon'' = \epsilon_\infty + \frac{\epsilon_s - \epsilon_\infty}{1 + j\omega\tau} - j \frac{\sigma}{\omega\epsilon_0}. \quad (4)$$

The influence of the third term, which represents the ohmic contribution is decisive at low frequency, hence the conductivity of the composite can be expressed by:

$$\sigma_S = 2\pi f \epsilon_0 \epsilon''. \quad (5)$$

3 Results and discussions

3.1 Spectral analysis

3.1.1 Behavior of the permittivity of the composite

The study of the frequency behavior in the wide band up to 2 GHz, allows the detection of possible relaxation phenomena. In order to see the behavior of the complex permittivity (ϵ' , ϵ'') of the ternary composite as a function of the sintering temperature and the frequency, its experimental results are given Figures 2 and 3.

Figure 2 shows the behavior of the permittivity of the sintered ternary composite as a function of the frequency, which exhibits a flat spectrum with a slight decrease in high frequencies. The spectrum dispersions obtained for

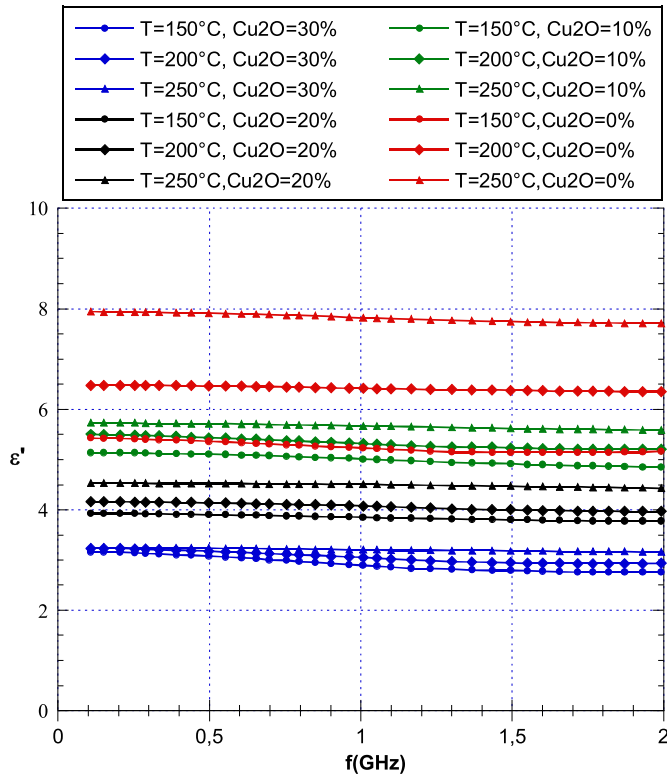


Fig. 2. Variation of ϵ' as function of the frequency.

different temperatures appears to be confounded and show a high contrast while decreasing the concentration of Cu_2O . At this inclusion phase, the increase of the sintering temperature provokes an appreciable deviation of the permittivity value and thus justifying its dielectric contribution advantageously. A graphic illustration of this parameter values obtained as a function of the frequency, the Cu_2O volume fractions, and the sintering temperature is shown in Figure 3. The examination of the curve of this permittivity, which characterizes the conducting side of the composite, reveals to us an opposite phenomenon instead of the dielectric one. This permittivity of the composite with the lack of BT inclusions drops considerably as the temperature increases. On the other hand, the increase addition of BT to the mixture provides a decreasing effect of ϵ'' which is more predominant to that of the temperature rise whose contribution is very little. Otherwise, it can be said that increasing the concentration of Cu_2O in the sintered composite leads to the increase of dielectric losses.

3.1.2 Conductivity behavior of composite

From equation (5), the conductivity of the ternary composite is seen as a function of the frequency, the Cu_2O volume fractions, and the sintering temperature. The results obtained are depicted in Figure 4, from which the curve indicates that both the drop in the Cu_2O concentration and the sintering temperature increase cause the conductivity to go down considerably.

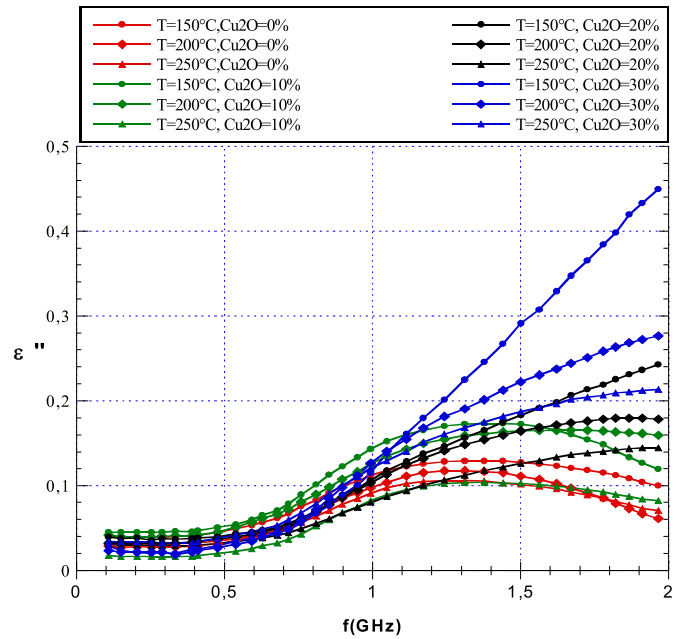


Fig. 3. The behavior of ϵ'' of the composite as a function of the frequency and the sintering temperature.

3.2 Low-frequency analysis

From the results obtained previously in frequency broadband up to 2 GHz, it is seen that the real part of the permittivity ϵ' is almost constant. Thus, its model remains valid and shows best fit to the experimental data in this frequency range. Despite this, the imaginary part of the permittivity ϵ'' has shown stable value at frequencies under 500 MHz. Therefore, the analysis of this parameter is confined to low frequencies and particularly to this upper limit frequency, that serves for conductivity value extraction to all samples under three sintering temperatures.

3.2.1 Modeling of the permittivity of composite

Several laws are proposed and used to study the dielectric properties of composite materials such as electrical module, electrical conductivity, and complex impedance. All these formalisms are able to describe the electrical phenomena present in either simple or complex composite materials. However, one or more of them may appear very able to reveal and demonstrate the physical mechanisms [4–9,12,13]. For the prediction of the effect both phases and forms of inclusions on the electrical properties of composites, other laws have been applied such as Maxwell-Garnet, Wiener, and the law of random mixtures of Lichtenecker.

The dielectric behavior of heterogeneous composite materials has often been subject to modification for various reasons, namely composite constitution, phase morphology, and preparation conditions such as temperature and else. The predictive study of the dielectric properties of this mixture type has carried out to the application of several mixing laws [4–9,12,13]. However, these remain confined to certain domains and specific cases of non-heterogeneous composites and non-random mixtures. Only attempts to develop new models are applied to composite materials that

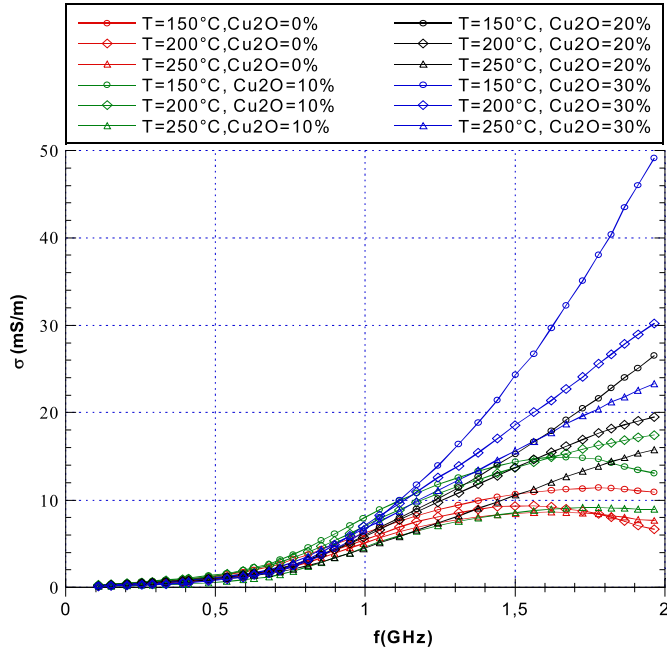


Fig. 4. Variation of the conductivity of the ternary composite as function of the sintering temperature and the frequency.

contain quite high charge concentrations in the host matrix. In contrary, composites with low charges, whose grains have different geometric shapes and randomly distributed are still the discussions object and scientific studies.

In the circumstances of these studies, an alternative solution to the mixing laws such as Wiener, Birchak Hanai, and Böttcher used for a heterogeneous dielectric medium explanation has replaced these theories because they lack a complete and a universal description. This proposed study is based on the Maxwell-Garnett, type formalism as brought up by Wakino et al. [4] and constructed by the modified Lichtenecker law. This probabilistic law, which has answered to the problem of the materials behavior, has allowed not only to evaluate the dielectric constant but also to be extended to the complex permittivity in the range of high frequencies reaching some GHz [4]. The effective dielectric permittivity value of the mixture ϵ_c was deduced from the Lichtenecker law that we express as:

$$\ln(\epsilon_c) = f_1 \cdot \ln(\epsilon_1) + f_2 \cdot \ln(\epsilon_2), \quad (6)$$

with $f_1 + f_2 = 1$. Here f_1 , f_2 , ϵ_1 and ϵ_2 are the volume fractions and the dielectric constants of the charge and the matrix, respectively.

On the base of its general sight, the empirical law can be generalized for N-phase composites according to the relation:

$$\ln(\epsilon_c) = \sum_{k=1}^N f_k \cdot \ln(\epsilon_k) \quad \text{with} \quad \sum_{k=1}^N f_k = 1. \quad (7)$$

In the case of a ternary composite (the case of our study), the effective permittivity can be written as follows:

$$\ln(\epsilon_c) = f_{RE} \cdot \ln(\epsilon_{RE}) + f_{BT} \cdot \ln(\epsilon_{BT}) + f_{Cu_2O} \cdot \ln(\epsilon_{Cu_2O}), \quad (8)$$

with $f_{RE} + f_{BT} + f_{Cu_2O} = 1$.

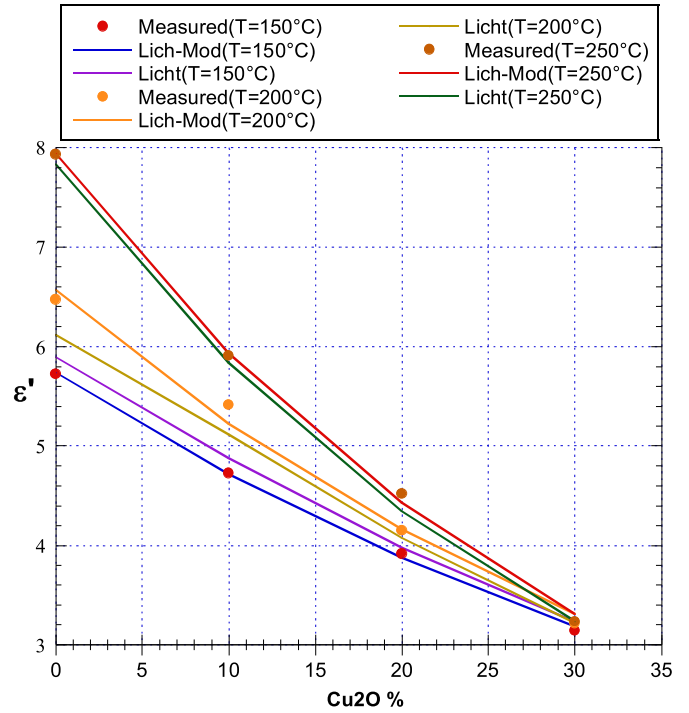


Fig. 5. Modeling with the Lichtenecker and modified Lichtenecker laws of the permittivity as a function of the volume fraction of Cu_2O .

Here (ϵ_{RE}, f_{RE}) , (ϵ_{BT}, f_{BT}) and $(\epsilon_{Cu_2O}, f_{Cu_2O})$ are the effective permittivity and volume fraction of RE, BT, and Cu_2O , respectively.

In this case of a ternary mixture, composed of RE, BT, and Cu_2O materials, the dielectric constant can be calculated through the modified law of Lichtenecker as follows:

$$\epsilon_c = F_s \cdot \epsilon_{RE}^{f_{RE}} \cdot \epsilon_{BT}^{f_{BT}} \cdot \epsilon_{Cu_2O}^{f_{Cu_2O}}. \quad (9)$$

Unknown parameters such as individual permittivity and shape factor can be evaluated using nonlinear regressions (LNR) [16,17]. In the modeling process based on this modified law, it is primordial to look for shape factors for each of the ternary composites made of oxide, titanate and epoxy resin. These factors must be in good coherence with the use of the law in the upper and lower limits of Wiener on one hand and to obtain a model, which describes the dielectric behavior of the mixture accurately on the other hand. As far as the direct and indirect Wiener model is concerned, the effective permittivity must have the upper and lower limits values given, into the following forms:

$$\epsilon_{WD} = f_{RE} \cdot \epsilon_{RE} + f_{BT} \cdot \epsilon_{BT} + f_{Cu_2O} \cdot \epsilon_{Cu_2O}, \quad (10)$$

$$\epsilon_{WID} = 1 / \left(\frac{f_{RE}}{\epsilon_{RE}} + \frac{f_{BT}}{\epsilon_{BT}} + \frac{f_{Cu_2O}}{\epsilon_{Cu_2O}} \right). \quad (11)$$

The resulting composite effective permittivity of the modified Lichtenecker law model must verify the Wiener condition:

$$\epsilon_{WID} \leq \epsilon_c \leq \epsilon_{WD}. \quad (12)$$

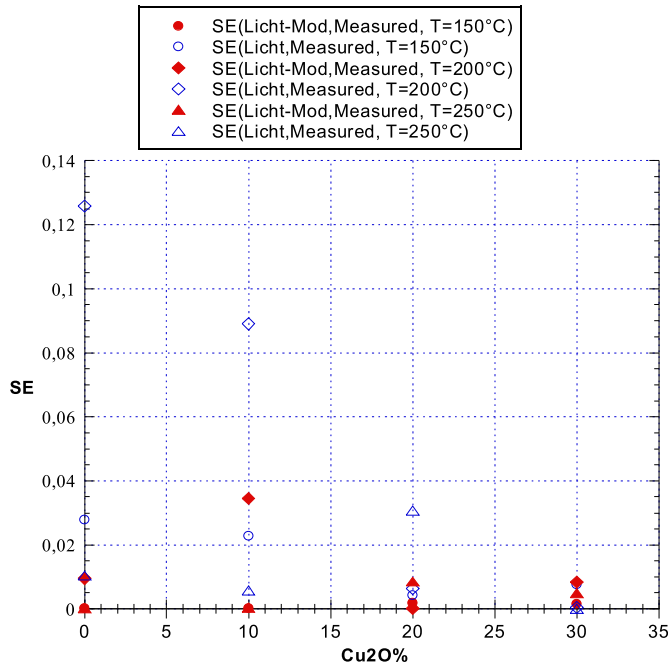


Fig. 6. Representation of the SE as function of the volume fraction of Cu_2O .

In this study, we used this law as a basic tool for modeling the permittivity of the mixture. The results obtained from this process using the modified Lichtenecker and Lichtenecker models are shown in Figure 5 for sintering temperatures of 150 °C, 200 °C, and 250 °C, respectively.

The modeling results demonstrate a correlation between both measurements and the Lichtenecker model since this one has provided data with a very good approximation to the measurement ones. The evaluation of the quadratic error between the measurements and the different models' results have made it possible to present the best-suited model to the measurements on one side and has revealed the effect of the grains on the other hand. Quadratic error (SE) is typically used in statistics to give a numerical value to the square difference between the values estimated by the model and the actual or measured values. The smaller or larger the SE is the closer or farther the model data are to actual data, thus proving or disproving the theoretical model.

For a large number of measurements, the SE varies from one measurement point to another. This conducts to a huge amount of data to be collected and consequently to be difficult to analyze. In this case, a use of the mean square error (MSE) as tool of calculation is recommended for data compression and optimal measurements reasons. The value of the MSE is obtained from the following expressions:

$$MSE = \frac{SE}{n} = \frac{\sum_{i=1}^n (\epsilon' - \epsilon'_{iModel})^2}{n}, \quad (13)$$

where “ n ” is the number of measured points, $\epsilon'_{iMeasure}$ and ϵ'_{iModel} are the values of the measured and the estimated permittivity by model respectively.

Table 1. MSE values as a function of the sintering temperature for both models.

Temperature (°C)	MSE (Measured-Modified Lichtenecker)	MSE (Measured-Lichtenecker)
150	0.00090	0.015575
200	0.013075	0.055300
250	0.003475	0.011650

At high BT concentrations, the effect of the shape factor appears as a minimum square error of the modified Lichtenecker model, which means that the effect of BT grains is considerable. On the other hand, at high concentrations of Cu_2O , the effect of the morphology is hardly noticeable. It is noted that the combination of the modified Lichtenecker and Lichtenecker law and the quadratic error make it possible to show the effect of morphology on the dielectric behavior of multiphase composites. The obtained values of the squared error SE for both Lichtenecker and the modified Lichtenecker laws models are presented as function of the concentration of Cu_2O and the sintering temperature in Figure 6.

The squared error (SE) has been carried out for the models of the usual and modified Lichtenecker laws using expressions (7) and (9) respectively. This is aiming to highlight the model that matches well the experimental results. The error (SE) estimated for the models mentioned above is computed for different Cu_2O concentrations and under the three sintering temperatures. The resulting curves illustrated in Figure 6 indicate the best fitting of the modified Lichtenecker law model to the experimental data since a minimum SE value is achieved with.

In order to be able to see the most suitable model, we calculated the different MSE values for the three sintering temperatures values shown in Table 1. The MSE values show that the proposed modified Lichtenecker model gives the best fit for the experimental results. This mean of error evaluation is carried out for both models mentioned before and under the three sintering temperature conditions. The results given in the table above show clearly the accuracy of the modified Lichtenecker law model.

Figure 7 below shows a comparison of the permittivities measured with those given by the different models (modified Lichtenecker, Wiener direct and indirect Wiener). By looking at the curves, we observe that the measured permittivity values and those obtained from the model of the modified Lichtenecker law are bordered by the direct and indirect Wiener models verifying the inequality of Wiener equation (12).

Figure 7 shows that the modified Lichtenecker law satisfies the direct and indirect Wiener conditions and gives us a good prediction of the permittivity of the multiphase composites.

3.2.2 Behavior of the shape factor with temperature

Due to the imperfections encountered by the samples in their manufacturing process, measurement errors accumulations occurred in the estimate of the shape factors, this

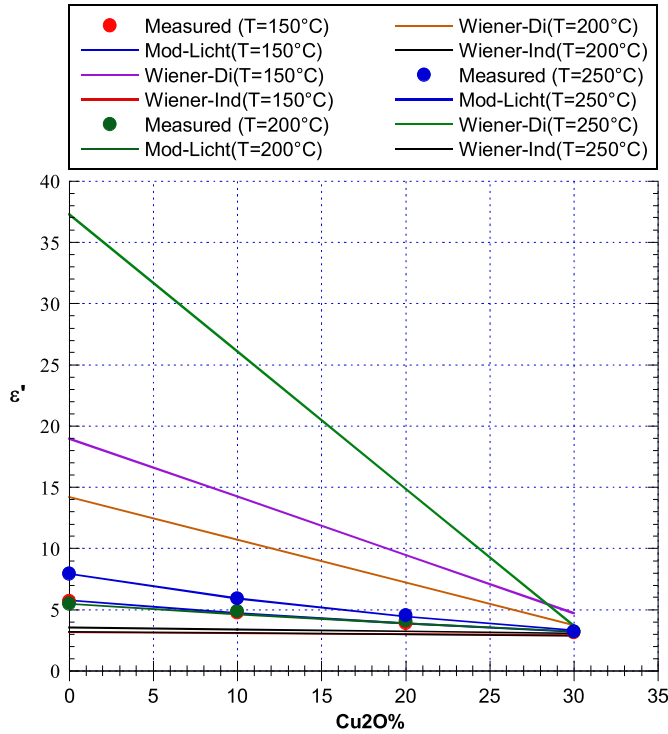


Fig. 7. Representation of the permittivity as a function of the volume fraction of Cu_2O with the different models at $T = 150^\circ\text{C}$, $T = 200^\circ\text{C}$, and $T = 250^\circ\text{C}$.

parameter, though small is its value (varying around 1), evolves nonlinearly. For an optimal shape factor estimation, a nonlinear regressions (LNR) calculation based approach was applied to the modified Lichtenecker model through equation (9) accordingly. Then, we get through the optimal solutions of the unknown parameters such as the actual permittivity of each constituents, and the shape factor as well. The latter is represented in Figure 8, as a function of the temperature.

In this study, we proposed a theoretical model in polynomial form indicating the degree of dependence of the shape factor on temperature and expressed as:

$$F_s(T) = p_1 \cdot T^2 + p_2 \cdot T + p_3, \quad (14)$$

where the coefficients p_1 , p_2 , and p_3 of the polynomial given by equation (14) have their values chosen from among their pair for allowing best fit of the Lichtenecker curve with the experimental one and are as follows: $p_1 = 4.59 \cdot 10^{-6}$; $p_2 = -6.699 \cdot 10^{-4}$; $p_3 = 0.8028$.

From these values, it appears that the serves as an adjustment of this factor for better curve smoothing, coefficient p_1 , although low in value, contributes with p_3 , which is constant to the improvement of the shape factor for relatively high temperatures. While p_2 is a negative value that by replacing the factor F_s by its new value in equation (9), we obtain:

$$\epsilon_C = F_s(T) \cdot \epsilon_{RE}^{f_{RE}} \cdot \epsilon_{BT}^{f_{BT}} \cdot \epsilon_{Cu_2O}^{f_{Cu_2O}}. \quad (15)$$

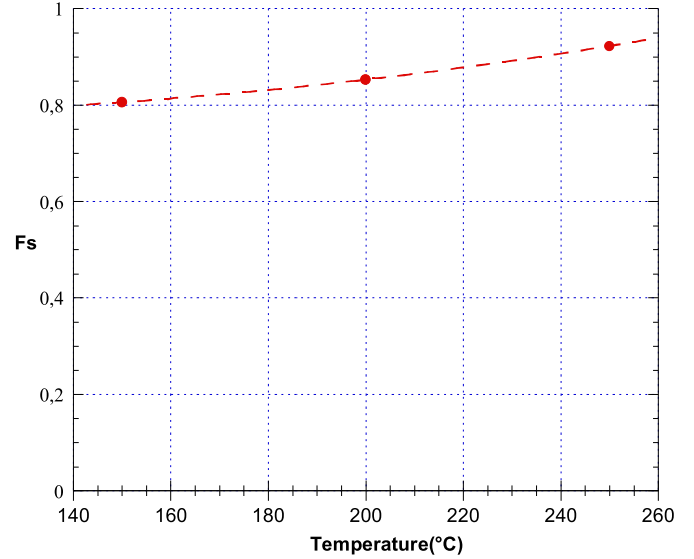


Fig. 8. Variation of the form factor as function of the sintering temperature.

It is noted that this model is related to the sintering temperature and may take another form or mathematical model depending on whether the morphology of the composite or the effect of temperature on the composite would require it. In the current study case, this factor usually less than or equal to unity, is located between 0.81 and 0.93. Therefore, the sintering process has an effect on the morphology and has led to a slight increase of the shape factor; following a nonlinear tendency. The extraction of parameters from the modified Lichtenecker model allowed us to obtain, besides to this parameter, each mixture's constituent permittivity under every sintering temperature. The results obtained are shown in Figure 9.

The figure shows that the titanate permittivity evolves increasingly as a function of the sintering temperature. On the other hand, the copper oxide permittivity goes down slightly. Despite all of these remarks, the sintering process has shown no effect on the permittivity and remains stable towards the temperature variations. These components joined together in a binary or a ternary combination will get their permittivity exhibited to alterations caused by sintering. For this purpose, merit figures (FoM) are widely used to compare composites with modified properties. The latter clearly shows the effect of each phase on the permittivity behavior of the binary composite (BT-RE and Cu_2O -RE). It is a question here of describing the relative improvement of the dielectric constant in a given matrix of polymer (RE), for a volume fraction (f_2) of the filler used, through the FoM calculation, which is defined as follows [18]:

$$FoM = \frac{\epsilon'_c - \epsilon'_1}{\epsilon'_1} \cdot \frac{1}{f_2}, \quad (16)$$

where ϵ'_c and ϵ'_1 are the dielectric constant of the binary composite and the polymer matrix respectively. For the comparison purpose, we calculated the FoM values of the RE-BT and RE- Cu_2O binary composites as a function of the sintering temperature and the results are depicted in Figure 10.

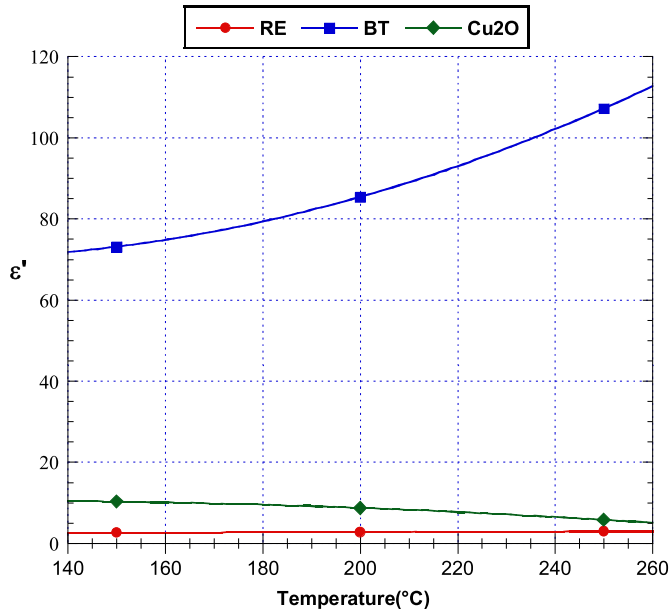


Fig. 9. Variation of the permittivity of BT, Cu₂O, and RE as function of the sintering temperature.

From this figure, it can be said, that the BT *FoM* increases with sintering temperature rises. This clearly expresses the effect of BT on the permittivity of the composite, which is more important than that of Cu₂O, which is weak. Actually, the *FoM* shows the importance of the effect of the sintering temperature on the permittivity of the BT compared to that of Cu₂O. In all cases, the values of the *FoMs* of the RE-BT binary composite are higher and grow linearly as a function of the sintering temperature. On the other hand, the RE-Cu₂O composite has almost fixed *FoM*.

3.2.3 Study of the static conductivity of composite

Figure 11 shows the variation of the conductivity as a function of the volume fraction of the Cu₂O. It is found that the sintered samples at 150 °C and 200 °C exhibit a static conductivity, which increases gradually to a maximum of 2.56 mS/m at 150 °C with a 20% concentration of and 2.36 mS/m at 200 °C for a concentration of 20%. On the other hand, the sintered samples with a temperature of 250 °C exhibit a diminishing of static conductivities that drop down from 1.98 to 1.08 mS/m while increasing the concentration of the Cu₂O.

Figure 12 shows the variation of the static conductivity as a function of the sintering temperature. It is observed that for all Cu₂O concentrations at the temperature of 150 °C, the differences between the static conductivities are low. At a temperature of 200 °C, the values of this conductivity are close to each other and the deviations between themselves are imperceptible. Whereas, at a temperature of 250 °C, the composites exhibit conductivity's values with a noticeable change from 1 and 2.6 mS/m. This is related to the concentration of the BT whose decrease of which causes this difference to deepen, i.e. it increases with increasing Cu₂O concentration. It is noted

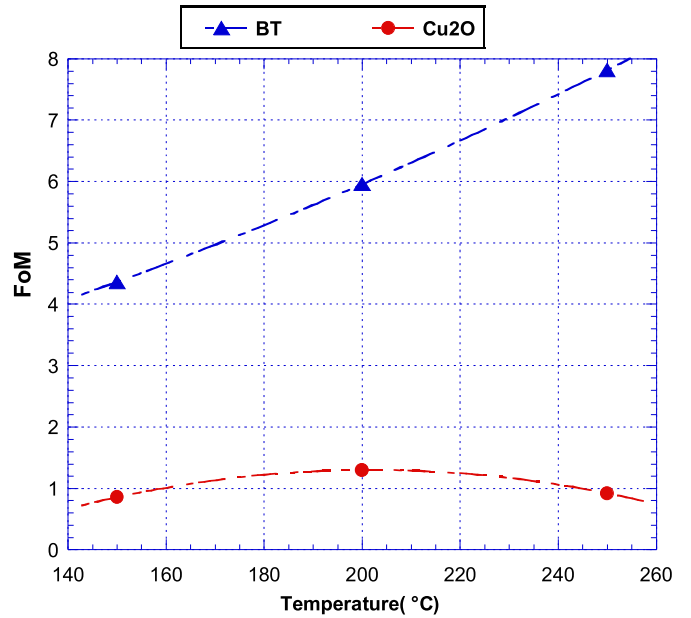


Fig. 10. *FoM* (Figure of merit).

that the high concentrations of the copper oxide increase the static conductivity of the composite for the temperature 250 °C.

3.2.4 Effect of sintering temperature

In Figure 13, the ϵ' results obtained are presented as a function of the sintering temperature in order to see the effect of the latter on the dielectric properties of the prepared composite materials. Deviations of the permittivity are observed for fixed percentages of Cu₂O at different sintering temperatures. Consequently, it is seen that a drop off of the Cu₂O concentration while the sintering temperature increases the difference between the permittivity values becomes more perceptible. Indeed, the passage of the volume fraction of Cu₂O from 30% to 0% led to the growth of the effective permittivity of the composites to almost 180% of its initial value under the sintering temperature of 150 °C, and to 200% and 250% of this value for sintered composites at 200 °C and 250 °C, respectively. The effect of the sintering temperature on the composite effective permittivity clearly appears in the permittivity of high BT concentrations samples. It reaches an increase of 113% for a 30% BT concentration when the sintering temperature increases from 150 °C to 200 °C and 138% for a sintering rate ranging from 150 °C to 250 °C. This is considered as an eminent effect for the improvement of the composite permittivity.

Figure 14 shows how the increase sintering temperature weakens the dielectric losses of the composites for different concentrations of Cu₂O and making it reaching its minimum value at the temperature of 250 °C for all the samples. On the other hand, the dielectric losses curves show a tendency change towards increasing values of $\tan(\delta)$, while increasing Cu₂O concentration and decreasing the sintering temperature. This confirms once again that BT titanate contributes in the same way as sintering temperature in reducing dielectric losses in composites.

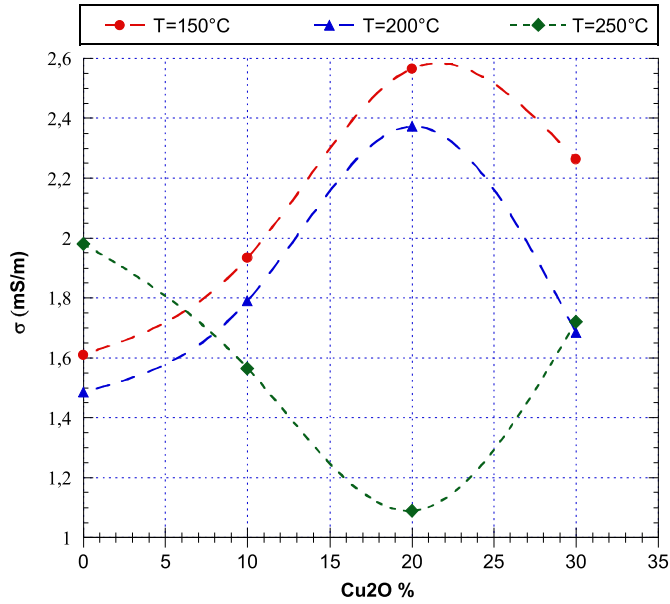


Fig. 11. Variation of the static conductivity as function of the volume fraction of Cu_2O .

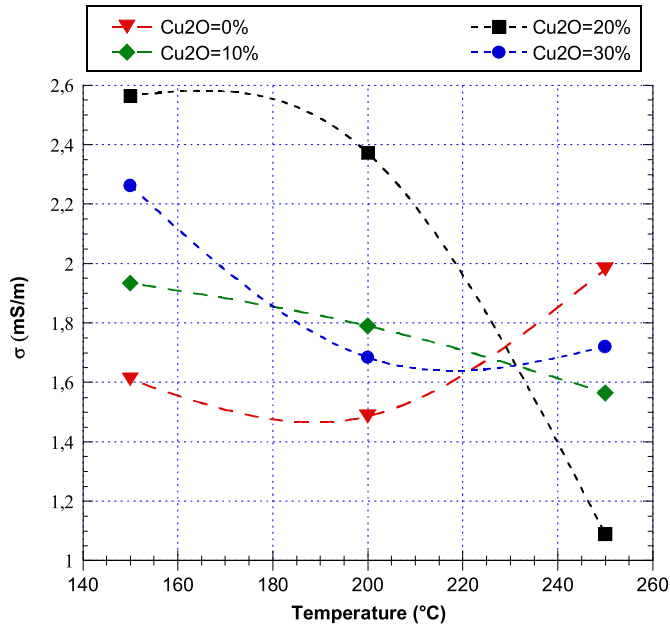


Fig. 12. Variation of the static conductivity as function of the sintering temperature.

4 Conclusion

In this study, a new composite material (RE- Cu_2O -BT) is prepared based on BT and Cu_2O powders dispersed in a matrix of epoxy resin, sintered at different temperatures in order to study the effects of Cu_2O inclusion, morphology, and sintering, on the dielectric properties of the BT-epoxy resin composite. Its dielectric characterization is carried out by the use of the time domain spectroscopy (TDS) method. For a prediction of the dielectric properties, a

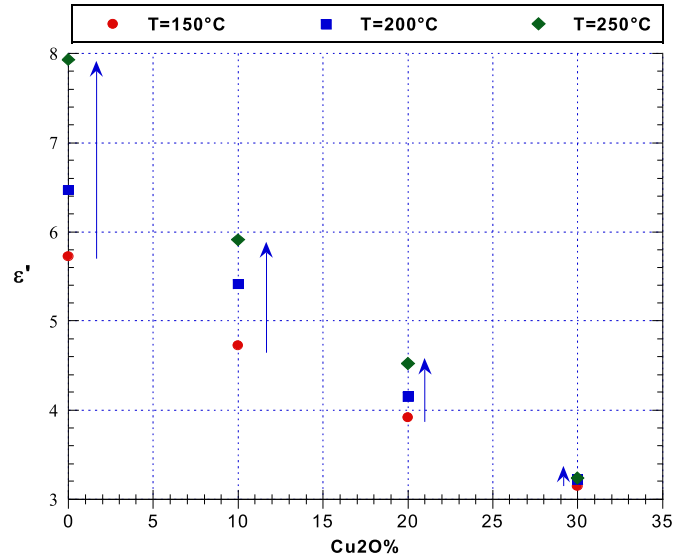


Fig. 13. Presentation of the deviations of the permittivity for fixed percentages of Cu_2O for different sintering temperatures.

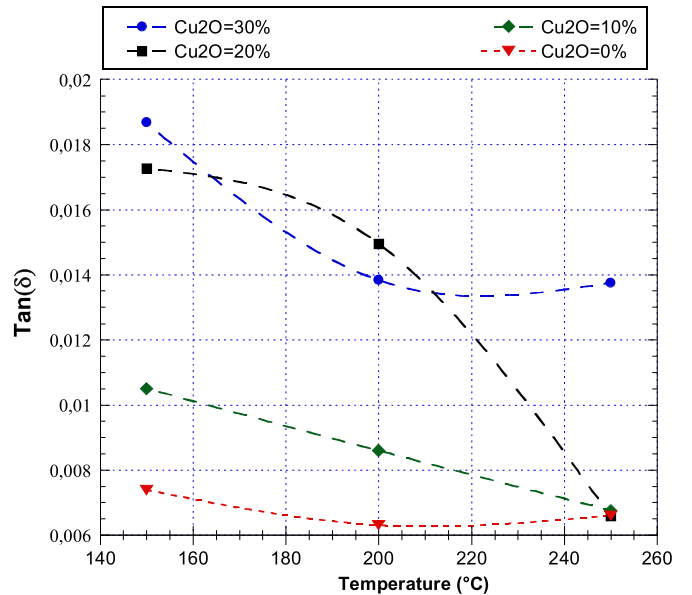


Fig. 14. Variation of $\text{Tan}(\delta)$ as function of the sintering temperature.

modeling was done using the two Lichtenecker parametric models and for which the parameters of both of them are calculated using nonlinear regressions. As a result of the quantification of these parameters and the use of nonlinear regressions, a better numerical optimization was obtained compared to the conventional approaches and an improved prediction of the dielectric behavior of the ternary composites has been achieved. The latter has shown good agreement with the experimental results and in addition, it has made in evidence the effect of the shape of the grains. The study concludes that the sintering temperature and the BT loading rate greatly increase the dielectric constant of the

composite. On the other hand, the effect of Cu_2O appears very influent in the static conductivity whereas its grains shape effect on the permittivity is imperceptible. As a matter of fact, the result we should point it out is the temperature which has considerably increased the permittivity of the composite independently from the volume fractions of the fillers. The dielectric characteristics obtained so far with the studied ternary composites show an acceptable improvement that can meet the needs of certain electronic and microelectronic applications at high frequencies.

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