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Double-walled carbon nanotube-based polymer composites for electromagnetic protection

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In this paper, we present a microwave absorber based on carbon nanotubes (CNT) dispersed inside a BenzoCycloButen® (BCB) polymer. The high aspect ratio and remarkable conductive characteristics of CNT give rise to good absorbing properties for electromagnetic protecting in microelectronic devices with very low concentration. In this article, nanocomposites are prepared using a solution-mixing method and are then evaluated and modeled by means of coplanar test structures. First, CNT concentrations are quantified by image processing. The nanocomposites implemented with coplanar test waveguides are then characterized using a vector network analyzer from 40 MHz to 20 GHz. An algorithm is developed to calculate the propagation constant "γ", attenuation constant "α", and relative effective complex permittivity ($\varepsilon_{reff} = \varepsilon_{reff}' - j\varepsilon_{reff}''$) for each CNT concentration. The extracted effective parameters are verified using the electromagnetic FEM-based Ansoft's® high frequency structure simulator (HFSS). Power absorption (PA) of 7 dB at 15 GHz is obtained with only 0.37 weight percent of CNT concentration in the polymer matrix. The resulting engineerable and controllable composite provides consequently a novel degree of freedom to design and optimize innovative microwave components.

Keywords: Carbon nanotubes (CNT), BenzoCycloButen (BCB) polymer, Power absorption

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I. INTRODUCTION

The miniaturization of new RF architectures leads to the integration of more and more RF-microsystems. In order to examine the performance of all those various instruments without electromagnetic interference, it is necessary to develop new shielding and absorbing materials. With the emergence of nanoparticles and nano-objects, a new class of tunable nanocomposites has entered the mainstream broadband market. The use of composite materials based on carbon nanotubes (CNT) appears to be very promising for nanotechnologies. CNT have shown exceptional stiffness [1], remarkable thermal [2], and electrical properties [3]. These advantages make them ideal candidates for the development of multifunctional material systems.

Numerous applications are envisioned for CNT, notably as part of a new class of nanocomposite materials; these nano-objects may be blended into a metal [4], ceramic [5] or, more commonly, polymer matrix [6]. The goal is to obtain a tunable material and to improve the physical properties of the composites. Depending on the application, the novel material could be configured for a low level of percolation threshold (p_c) [7], or to improve mechanical [4], thermal [8], and electrical properties of the matrix [9]. In microwave application, CNT can be utilized (or proposed) for the electrical discharge of bridges microelectromechanical systems (MEMS) where the CNT are added into the dielectric materials [10] and improve the reliability of MEMS. Moreover, the addition of CNT in composites is purposed for broadband microwave absorbing materials [11, 12]. These composites facilitate the protection of the sensitive electrical devices from electromagnetic waves. Indeed, the electromagnetic radiations coming from different sources – electronic circuitry, radar, mobile phone, radios, power lines, etc. – are increasingly present in our environment and could affect the circuits' performances. In 1983, Air Canada did not allow the use of personal computers on its flights for fear that radiated electromagnetic waves will interfere with the onboard navigational computers. This action is still true nowadays, as air plane companies classically recommend switching off all electronics devices during take-off and landing for the purpose of security.

In this paper, the study consists of electromagnetic characterization and electromagnetic modeling of composites based on BenzoCycloButen® (BCB) [13] polymer dispersed with CNT at microwave frequencies (40 MHz to 20 GHz) using coplanar waveguides (CPWs) as test structures. This polymer was chosen because of its frequent use in microwave applications [14], its remarkably low-dissipation factor (0.0008–0.002 at 1 MHz to 10 GHz, respectively), and low relative permittivity (∼2.65).

According to our knowledge, and with the exception of our earlier work [15], this paper is the first report on a concentration characterization by image processing of CNT in BCB. It is shown in Section II that the different concentrations

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were optically characterized and compared with targeted ones. The results show very low deviation (<1%) between measurements and targeted concentrations.

Moreover, this study presents a primary report on the microwaves electrical properties (σ, ε, tan δ) and the power absorption (*PA*) of CNT–BCB nanocomposite at microwave frequencies. The conduction properties, the percolation threshold (p_c), and the *PA* of CNT nanocomposites are compared with other CNT-containing composites, which are described in the literature. A conductivity level of 0.5 S/m is obtained with only 0.37 wt% concentration of CNT. A good dispersion of CNT in the polymer is necessary and enables a low percolation threshold of "$p_c = 0.075$ wt%". As detailed in Section III, having a good conduction in the material will favor the absorption of power inside the material. The measurements results confirm *PA* (as defined in Section IV) for a shielding thickness of 16 μm. Such thickness corresponds indeed to a typical value for microelectronic systems in microwave applications. For higher concentrations (0.37 wt%), the absorption level of CNT nanocomposites may be multiplied by a factor 10.

II. SAMPLES PREPARATION

A) Fabrication and purification of CNT

As described previously [16], the CNT were prepared by catalytic chemical vapor deposition of a H_2–CH_4 mixture at 1000°C with a CoMo–MgO catalyst. CNT are free from amorphous carbon contamination and the use of an MgO support allows their easy removal [17] without damaging the nanotubes. During the extraction step, the oxides and unprotected metal nanoparticles were dissolved by the addition of an aqueous solution of HCl. The acidic suspension was then filtered on 0.45 μm pore-sized polypropylene membranes (Whatman) and washed with demonized water. The wet nanotubes were then dispersed in T1100 (solvent of BCB) by solvent exchange. Finally, the suspensions were obtained by adding 15.8 g of T1100 to 300 mg of "wet" CNT. The double-walled carbon nanotubes (DWNT) have two concentric walls with an average outer diameter "*d*" of 2 nm and a length "*l*" between 1 and 10 μm [16]; 80% of them have metallic properties, which means that the remaining 20% have semiconducting properties.

B) CNT-polymer blend preparation

The dispersion of the CNT in the matrix system is an important challenge. These nanoparticles exhibit an important specific surface area (ca. 1000 m^2/g) that plays a role in the strong tendency of CNT to form agglomerates. Exploiting the CNT properties in polymers is therefore related to their homogeneous dispersion in the matrix and the separation of the agglomerates, as well as good wetting by the polymer.

CNT were added to the BCB polymer by mechanical mixing (blender) in order to improve the homogeneity of the CNT in the BCB polymer. The morphology and microstructure of the CNT-containing composites was observed by scanning electron microscopy (FEG-SEM) as shown in Fig. 1, and in their "as-prepared" state.

By mixing 22.58 g of BCB, 4.49 g of T1100, and 0.008 g of CNT with the blender, a CNT–BCB composite could be fabricated at a high nanotube concentration (0.37 wt%). Next, in order to obtain low concentrations of CNT, the solvent T1100 and BCB were added in proportion 5:1. Finally, five different concentrations of CNT in BCB polymer (0.05, 0.1, 0.2, 0.3, and 0.37 wt%) were investigated. Using equation (1), we calculated the weight concentration of CNT:

$$\mathrm{CNT(wt\%)} = \frac{m_{CNTs}}{m_{CNTs} + m_{BCB}} * 100, \quad (1)$$

where m_{CNTs} and m_{BCB} are the CNT and the BCB masses, respectively.

Additionally, a composite without CNT (0 wt%) was fabricated and used as a reference sample. Composite solutions (in T1100) were spin coated on silicon wafers and polymerized at 250°C for 1 h in nitrogen flow. Optical microscope images of spin-coated deposits at two different concentrations are compared in Fig. 1.

Both at the lowest and highest CNT concentrations, SEM and optical microscope (Fig. 1) images show agglomerates of CNT. It was observed that the aggregate size increases with the CNT concentration. CNT networks were formed at all concentrations. At the highest CNT concentration (0.37 wt%), large black agglomerates are clearly visible on the surface and may change the surface roughness.

C) Planarization of BCB/DWNT composites

The presence of CNT agglomerates modified the surface roughness, leading to a bad interface for electrical characterization. In order to obtain a clean planar interface, a polishing step (chemical mechanical planarization – CMP) was performed for each sample. In all experiments presented in this paper, a CDP41 CMP tool from Logitech was used for polishing the wafers. By combining 1 psi pressure of mechanical friction (rotating plate) and a flow rate of 100 ml/min of a chemical agent, the surface roughness was decreased by a factor 10. The surface polishing step is illustrated in Fig. 2 where it can be observed that the surface roughness was clearly decreased after polishing. The results are listed in Table 1, before (noted R_a) and after (noted R_p) the polishing step.

The data show a significant increase in the roughness with the CNT concentration. For the highest concentration of CNT (0.37 wt%), the surface roughness was significantly decreased after polishing ($R_a = 4.57$ μm, $R_p = 0.53$ μm). The planarization of the CNT–BCB composites decreased the global surface roughness to 0.5 μm in the worst case.

III. CHARACTERIZATIONS AND ELECTROMAGNETIC SIMULATION OF NANOCOMPOSITES

A) Concentration characterization

To quantify the CNT concentration in the composites, this paper presents a first concentration characterization using a standard image processing method. Concentrations of CNT are characterized by evaluating the black point quantity of the optical microscope images as shown in Fig. 1; these black points represent the CNT agglomerate on and into the

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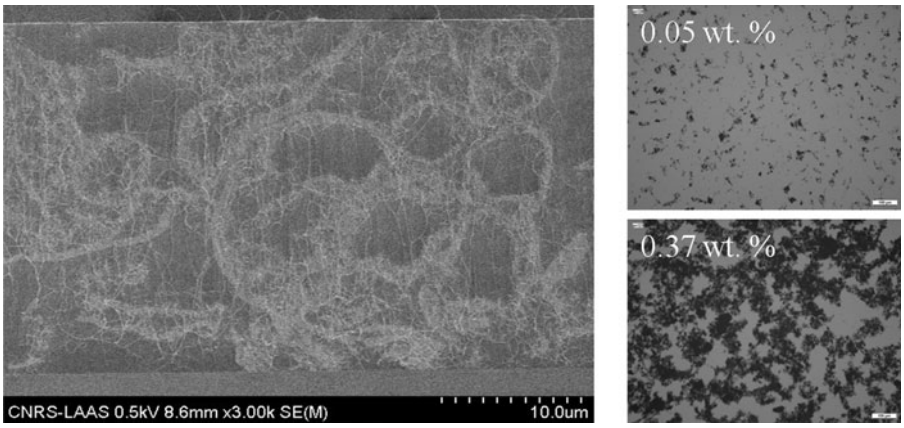


Fig. 1. SEM cross-sectional views of BCB/DWNT composites and optical microscope images of CNT–BCB composites at 0.05 and 0.37 wt%.

BCB layer. At different concentrations (see Fig. 1), the presence of CNT can be more or less important. For high concentration (0.37 wt%), the quantity of black points or the CNTs agglomerate are more important than for low concentration such as 0.05 wt%. The concentrations may be evaluated as a function of quantity of agglomerates using the image processing program ImageJ. The ImageJ 1.38× software calculates the surface black point quantity in percentage (s. %). Figure 3 shows an example of the characterization steps for a CNT concentration of 0.05 wt%.

The process starts with an optical microscope image conversion from color to monochrome image, as shown in Fig. 3(a), as the particles analysis requires a "binary" image (i.e. black and white). The edges need to be exactly defined in order to allow morphological measurements by the software. A "threshold" range has to be set. Pixels, the value of which is included in this range are converted into black, whereas the other ones are converted into white, as shown in Fig. 3(b). Based on this analysis, the quantity of particles is determined in surface percent (s. %), as shown in Fig. 3(c).

The surface percentage is then calculated and integrated via equation (2) in order to extract the weight percentage (wt%) of CNTs [18]:

$$\frac{\Phi_p^m}{\Phi_p^S * 3.25} = \Phi_p^m + \frac{\rho_p}{\rho_m}(1 - \Phi_p^m), \tag{2}$$

where ρ_m and ρ_p are the densities of BCB (0.97 g/cm^3) and CNT (2.36 g/cm^3 [19]), respectively. Φ_p^m is the weight concentration (wt%) and Φ_p^s is the surface concentration obtained with Image J software (s. %). The measurement values and the theoretical concentrations are reported and compared (see Fig. 4).

The excellent agreement between the image processing results and the calculated ones illustrates the accuracy of our calculation technique (deviation $<5\%$). Hence, it can be concluded that the characterization method of CNT concentration with an image processing program is appropriate to characterize the composite after fabrication.

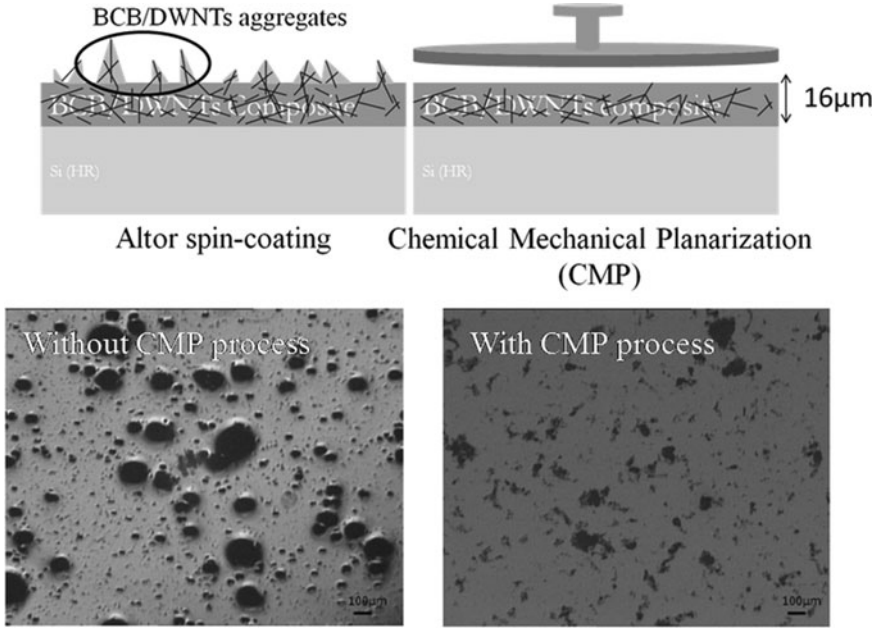


Fig. 2. Polishing process and optical microscope images of BCB/DWNT composite (0.127 wt%): (a) before CMP, and (b) after CMP.

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Table 1. Measured roughness before (Ra) and after polishing (Rp) for each CNT concentration

Concentrations (wt%)	*Ra* (μm)	*Rp* (μm)
0.05	0.85	0.06
0.10	0.98	0.05
0.20	1.75	0.08
0.30	5.25	0.21
0.37	4.57	0.53

Therefore, the following section will concentrate on the electromagnetic characterization and electromagnetic simulations.

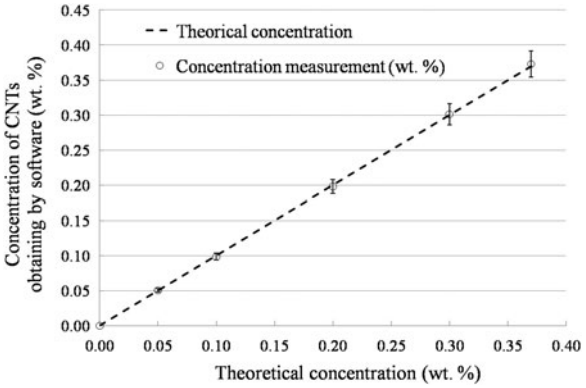


Fig. 4. Theoretical concentrations versus measured concentrations with Image J software.

B) Electromagnetic characterization and electromagnetic simulations

To characterize the nanocomposites in the microwave regime, CPW transmission lines were designed and realized on various 16 μm BCB-composite layers (over a silicon substrate) featuring the different concentrations presented in Table 1. Using a vector network analyzer coupled to an on-wafer probe station, measurements were completed in a range from 40 MHz up to 20 GHz. For each BCB-composite, a line–line method [20] using three lines with different lengths (600, 800, and 1000 μm) was developed in order to extract the dielectric constant and losses of the various BCB composites. S-parameters characterization was presented in a previous study [15] and demonstrated a good repeatability between identical samples. The data of one of the samples are plotted in Fig. 5 (solid).

This study presents the complex propagation constant "γ" and effective parameters that can be extracted from *S*-parameter measurement using (3). The relative parameters (ε_{reff}, σ_{reff}) include the different layers (silicon substrate, BCB or composite, metallic line, and air). In our case, CNTs

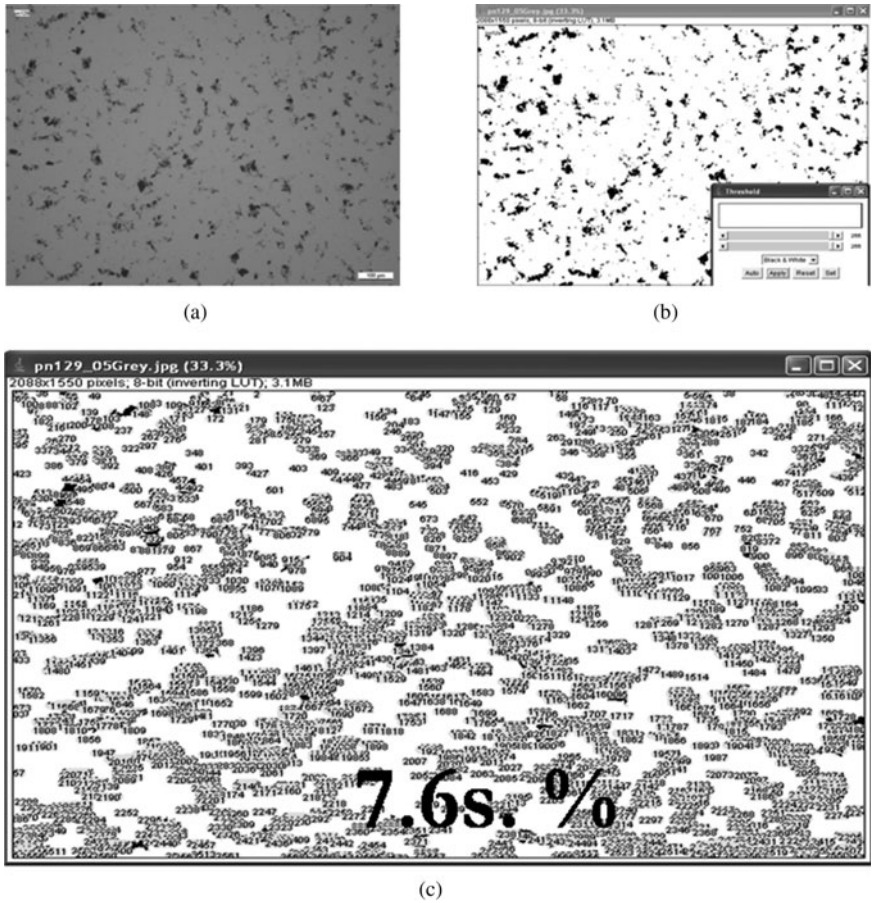


Fig. 3. Concentration characterization of CNT–BCB composites of 0.05 wt%: (a) monochrome optical microscope image, (b) tuning of threshold contrast (black point), and (c) particle analysis results in surface concentration (% s).

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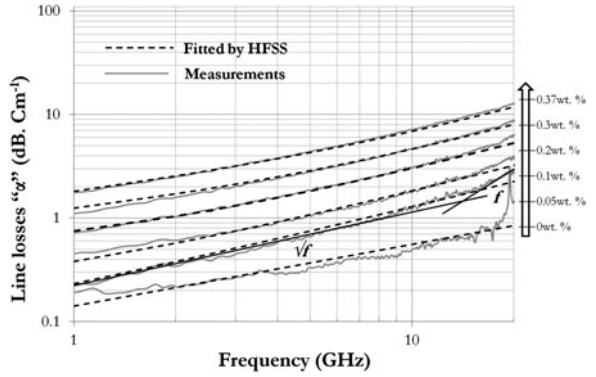


Fig. 5. Line losses measurement (solid) and simulate (dotted line) for 0, 0.05, 0.1, 0.2, 0.3, and 0.37 wt% of CNT concentration.

are randomly aligned (as manifested in Fig. 1) and we will treat the nanocomposite as an isotropic effective medium.

From "γ" we can easily deduce the line losses "α" and the relative effective permittivity "ε_{reff}" of the CPWs using equation (4). To determine the effective parameters (ε_{eff}, σ_{eff}), the electromagnetic simulator (high frequency structure simulator – HFSS) was used to fit between measurements with simulations.

As a reference, the HFSS simulations show good agreement with measurements in the case of a BCB-polymer layer without CNT. The supplier's parameters were integrated ($\tan\delta = 0.0008$ and $\varepsilon_{eff} = 2.65$) in HFSS simulator, and the excellent agreement between simulation and characterization illustrates the accuracy of the electrical parameter extraction, as shown in Figs 5 and 6. By tuning the effective parameters (ε_{eff}, $\tan\delta$) of BCB polymer within the HFSS software, the simulations were fitted with measurements for each CNT's concentration. For an improved correlation between simulations and measurements, an iterative method is finally performed.

We then monitor the impact of the CNTs concentration on the electromagnetic properties of the resulting composite [15].

Data from Figs 5 and 6 show the results of electromagnetic simulations of the line losses and relative effective permittivity, respectively:

$$\gamma l = \alpha l + j\beta l = \mathrm{Argch}\left(\frac{1 - S_{11}^2 + S_{21}^2}{2.S_{21}^2}\right), \tag{3}$$

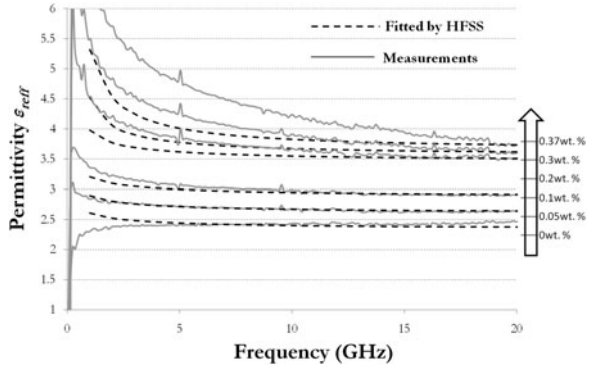


Fig. 6. Permittivity measurement (down stroke) and simulate (dotted line) for 0, 0.05, 0.1, 0.2, 0.3, and 0.37 wt% of CNT concentration.

$$\gamma l = \alpha l + j\left(\frac{2\pi}{c} f \sqrt{\varepsilon_{reff}}\right) l, \tag{4}$$

where l is the line length of the CPW, f is the frequency, and c is the velocity of light in free space.

For 0 wt%, Fig. 5 shows that the line losses vary as a function of $\sqrt{f}$ in the range from 1 to 10 GHz and as a function of f in the range of 10–20 GHz. This corresponds to the resistive (α_c) and dielectric (α_d) losses, respectively. When we doped the BCB polymer with the CNT in BCB polymer, the metallic losses increase. Consequently, α varies as a $\sqrt{f}$ in the range 1–10 GHz. At 0.37 wt%, for example, the lineic losses are multiplied by a factor of 17 (11.9 dB/cm) compared to pure BCB (0.7 dB/cm) over the whole frequency range. This demonstrates the impact of metallic CNT in BCB polymer. Indeed, 80% of CNT are metallic and 20% are semiconductors.

The measurement results and electromagnetic simulations from HFSS of the RF permittivity (ε_{reff}) are given in Fig. 6 for every CNT–BCB composite. The increase of CNT concentration affects the permittivity parameter. At 20 GHz, the real part of ε_{reff} ranges from 2.1 (pure BCB) to 3.8 (0.37 wt%).

By definition, ε_{reff} is composed of a real and an imaginary parts (5). The imaginary part of permittivity is proportional to effective conductivity, to refer to equation (6):

$$\varepsilon_{reff} = \varepsilon' - j\varepsilon'' = \left(\frac{c_0}{j2\pi f}\right), \tag{5}$$

$$\sigma_{reff} = 2\pi f \varepsilon_0 \varepsilon'', \tag{6}$$

where c_0 and ε_0 are, the speed of light and the dielectric permittivity in vacuum, respectively.

For the BCB polymer without CNT, the imaginary part (ε'') is presented in Fig. 7, it is approximately zero (0.03 at 5 GHz and 0.11 at 20 GHz), in all the frequency ranges. This means that BCB polymer is a good dielectric material for microwave applications. By addition of CNT, the ε'' parameter increases by factor 12 (1.39 at 20 GHz) with 0.37 wt% of CNT. This variation of ε'' confirms the conductive behavior of the polymer.

Deduced from S-parameter characterizations, the relative parameters (ε_{reff}, ε'') were extracted and demonstrated modification depending on the CNT doping of the BCB polymer. It is observed that the concentrations in CNT change the electrical properties of BCB.

C) Electrical parameters extraction of nanocomposites

The electric parameters of nanocomposites are studied using the HFSS simulation software. After modeling and fitting between measurements and simulations, conductivity (σ_{eff}), permittivity (ε_{eff}), and loss tangent ($\tan\delta$) values are extracted from the HFSS library. The effective parameters indicate the real value of nanocomposites for each concentration.

Primary, the effective conductivity variances "σ_{eff}" according to the weight percent of CNT is reported in Fig. 8. The results demonstrate a low percolation of nanocomposites doping in CNT (8) [21]:

$$\sigma = (p - p_c)^t \quad \text{for } p > p_c, \tag{7}$$

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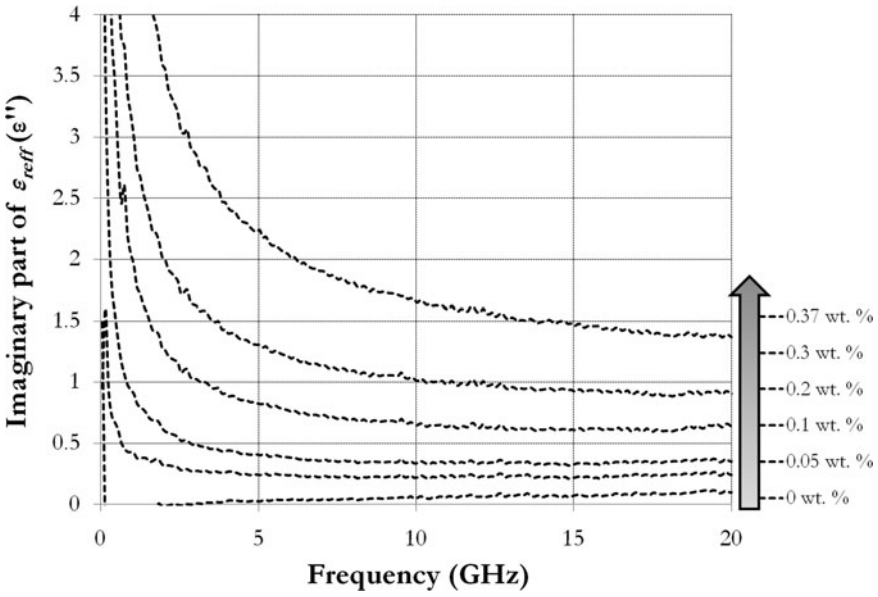


Fig. 7. The imaginary part of ε_{reff} measurement for 0, 0.05, 0.1, 0.2, 0.3, and 0.37 wt% of CNT concentration.

where p is the weight fraction of CNTs, p_c corresponds to percolation threshold, and t to the critical exponent [21].

A conductivity of three orders of a magnitude higher than the pure BCB is extracted for the 0.37 wt% sample (0.0001 up to 0.5 S/m).

An abrupt percolation threshold "p_c" of the conductivity was observed at 0.075 wt% CNT, indicating the existence of percolating paths via connecting CNT. At this stage, the conductivity of the nanocomposite is controlled by the conducting CNT. The conductivity is due to the formation of a network of CNT within the BCB matrix. Barrau *et al.* [6] have presented an optimum percolation threshold of 0.08 wt% obtained with suspensions of CNTs. Our results contribute and confirm the quality of nanocomposite BCB/CNT that we can fabricate.

The inset Fig. 8 shows the percolation scaling law between $\log \sigma$ and $\log(p - p_c)$ where the solid line corresponds to the best-fitted line.

This law leads to a low percolation threshold (0.075 wt%) and critical value of $t = 1.24$ less than the classical used one ($t = 2$). The exponent t reflects the dimensionality of the systems, the values 1.3 and 1.93 corresponding to two and three dimensions, respectively. As reported by Barrau *et al.* [22], a value of t around 1.44 has been observed in Polyepoxy. The low value of t would not be the sign of a two-dimensional network but rather a consequence of weakly connected parts in the networks.

To define the electrical behavior of the composite, the loss tangent ($\tan \delta$), effective permittivity (ε_{eff}), and the effective conductivity (σ_{eff}) are defined for each concentration, as shown in Table 2.

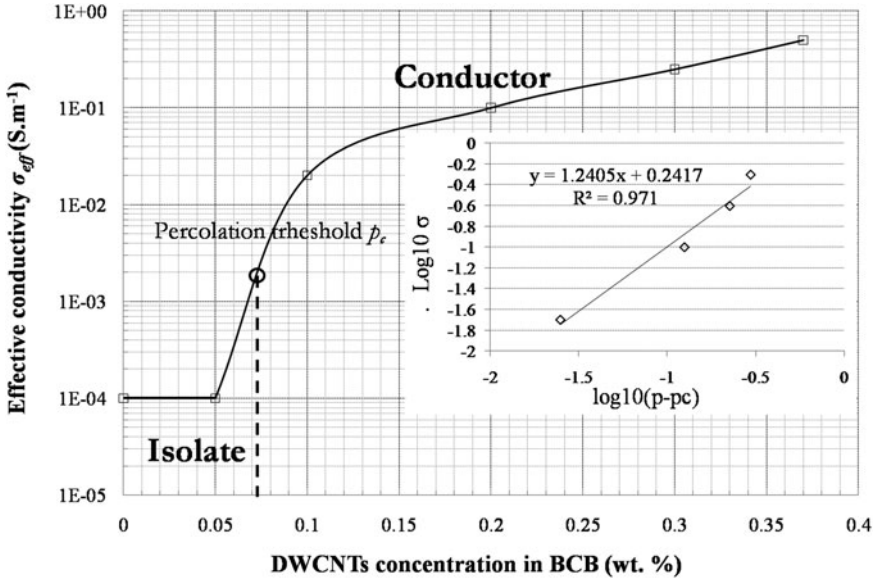


Fig. 8. Effective conductivity dependence on the CNT (in linear scale and log10–log10 scale).

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Table 2. Electrical parameters for each concentration by extraction of HFSS simulator

Concentrations of CNT in BCB (wt%)	tan ô	σ_{eff} (S/m)	ε_{eff}
0	0.0008	0.0001	2.3
0.05	0.075	0.0001	2.8
0.1	0.12	0.02	3.3
0.2	0.2	0.1	4.6
0.3	0.32	0.25	4.8
0.37	0.5	0.5	4.8

For a doping of 0.2 wt%, the ε_{eff} increases two times compared to the one without CNT. For a higher concentration (>0.3 wt%), the effective permittivity saturates and reaches a maximum value of 4.8. This behavior will be explained below.

Loss tangents are increased by a factor of 100 under the percolation threshold "p_c". Above p_c, tan δ behavior is linear.

The explanation of this is based on the following equation:

$$\tan \delta = \varepsilon''/\varepsilon'. \tag{8}$$

The imaginary part of the permittivity (ε'') is related to the energy dissipation (or loss) within the medium and the real part (ε') is related to the stored energy within the medium. At the percolation threshold, the loss tangent is dominated by the imaginary part, composed of conductor losses (α_c) and dielectric ones (α_d) in the composite. At this point, the nanocomposite becomes a conductor composite. After the stationary regime ($p > p_c \sim 0.2$ wt%), the loss tangent is stabilized by increasing the real part of permittivity. Once this point is reached, the composite begins to store energy in the matrix. In microwave applications, the storage of energy is critical for shielding. Therefore, the following section will concentrate on the electromagnetic protection parameter, keeping in mind the conductivity 0.5 s/m and loss tangent of 0.5 for 0.37 wt% in CNT.

IV. NANOCOMPOSITE BCB/CNT FOR MICROWAVE APPLICATIONS

In the previous section, we observed that the addition of CNT within a BCB polymer changes the electrical behavior and increases the linear losses. For microwave applications, losses are generally avoided in the systems so as not to lose the transmission signal. However, these drawbacks can be an advantage for some other applications such as electromagnetic shielding. Our nanocomposite may be a new material for absorbing electromagnetic waves for the microelectronic components. *PA* that is related to the parameters of the material is defined by

$$PA = 20 \log_{10} |e^{\gamma}|. \tag{9}$$

The *PA* presented in Fig. 9 varies over the frequency range and also for different concentrations of CNT. For the low concentration (0.05 wt%), the low conductivity does not result in significant absorption of the electromagnetic wave through the composite. This is confirmed by the low value of 1 dB at 12 GHz of the *PA* in the composite. When the concentration increases (0.1–0.37 wt%), the *PA* in the composite increases. For the 0.37 wt%, the *PA* reaches 9 dB at 20 GHz instead of 1 dB without CNT. Compared with other studies on composites, one advantage of the proposed composite is the very low-percolation threshold "p_c" with the low polymer thickness (16 μm) while keeping a good *PA* of 9 dB. In addition, we obtain a good conductivity and further improvements could be made by increasing the concentration of CNTs within the polymer or/and by increasing the composite layer thickness. Our study, however, is focalized on the microelectronic scale and for microwave application (Table 3).

At a fixed frequency, the contribution of CNTs is very significant in terms of electromagnetic absorption. For expected *PA* at a fixed frequency, the concentration of nanocomposites can be consequently predicted and fabricated. For example, for 5 dB of *PA* at 15 GHz, a 0.3 wt% concentration is necessary to fulfill the specifications. We have thus obtained an

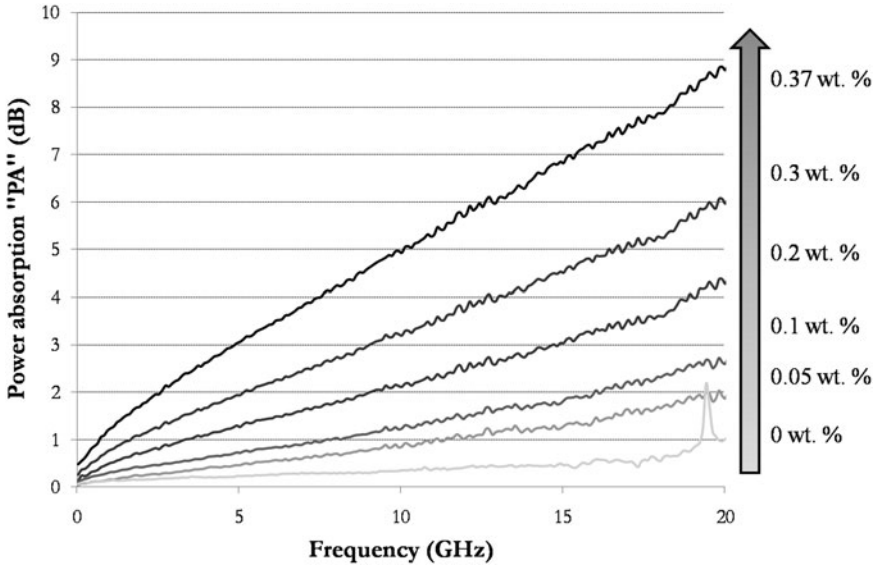


Fig. 9. *PA* for BCB/CNT containing 0, 0.05, 0.2, 0.3, and 0.37 wt% of inclusion.

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Table 3. Summary of reported properties of carbon nanotubes composite

Reference	Our	[11]	[23]
CNT concentrations, (wt%)	0.075	0.5	8
Thickness	16 μm	1 – 2 mm	1.4 mm
Conductivity	0.5 S/m	1 S/m	Unreported
Power absorption	9 dB at 20 GHz	11 dB at 20 GHz	15 dB at 18 GHz

engineerable configurability of nanocomposites for RF applications.

V. CONCLUSIONS

This paper presents a study of CNT–BCB-based composites for microwave applications. Our research started from experimental results to equivalent modeling. For good dispersion of CNT into the BCB polymer matrix, we used purified and filtered double-walled CNTs obtained from the CIRIMAT laboratory to obtain a high quality of CNT. Their suspension in the BCB solvent (T1100), combined to a mechanical dispersion method (mixer) improved the uniformity mixing of the nanocomposites. These techniques allowed the production of composite materials from 0 to 0.37 wt%. For high concentrations, however, CMP was used to reduce the surface roughness caused by the agglomerates of CNT. CPWs on different composites were then fabricated; this permitted us to characterize losses induced by CNTs. The majority presence of metallic CNTs increased the linear losses, which implied an increase in conductivity of the material. An iterative simulation allowed the extraction of electrical parameters (σ_{eff}, ε_{eff}, and tan δ) of each composite. The change of effective conductivity identified a percolation threshold of 0.075 wt%. This very low value was obtained by using a liquid suspension of DNTCs. These nanocomposites could be implemented for electromagnetic shielding of electronic systems at the micrometer scale. The small thickness and compatibility with integrated circuits' technologies correspond to strong advantages for their use as new absorbent materials in microelectronics.

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