

Experimental determination of microwave attenuation and electrical permittivity of double-walled carbon nanotubes

M. Dragoman

**National Research and Development Institute in Microtechnology, Str. Erou Iancu Nicolae 32
B, 077190 Bucharest, Romania**

K. Grenier, D. Dubuc, L. Bary, E. Fourn, and R. Plana

LAAS CNRS, 7 Avenue du Colonel Roche, 31077 Toulouse Cedex 4, France

E. Flahaut

**Centre Interuniversitaire de Recherche et d'Ingénierie des Matériaux, UMR CNRS 5085,
Université Paul Sabatier, 31062 Toulouse, France**

The attenuation and the electrical permittivity of the double-walled carbon nanotubes (DWCNTs) were determined in the frequency range of 1–65 GHz. A micromachined coplanar waveguide transmission line supported on a Si membrane with a thickness of 1.4 μm was filled with a mixture of DWCNTs. The propagation constants were then determined from the S parameter measurements. The DWCNTs mixture behaves like a dielectric in the range of 1–65 GHz with moderate losses and an abrupt change of the effective permittivity that is very useful for gas sensor detection.

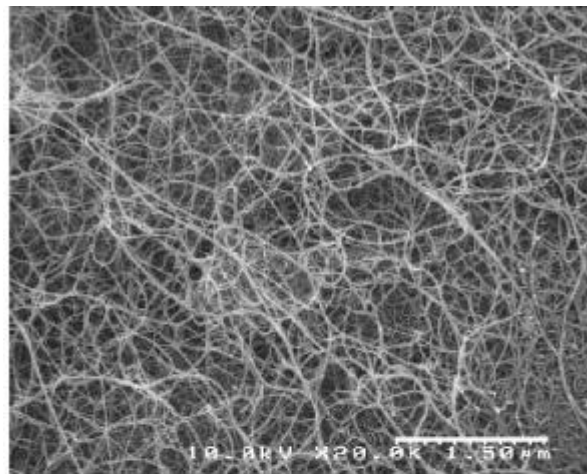
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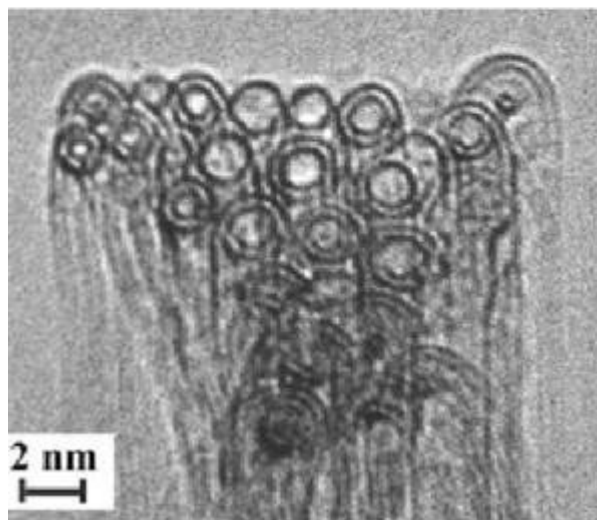
Footnotes

The carbon nanotubes are now extensively studied for various microwave applications (see Ref. [1](#) for an extensive review). Passive devices such as filters,^{[2](#)} and active devices such as oscillators,^{[3](#)} mixers,^{[4](#)} and nanoelectromechanical systems (NEMS)^{[5,6,7,8](#)} are envisaged to be implemented using carbon nanotubes (CNTs) and having impressive performances. These performances originate in the amazing physical properties of the CNTs. Among these properties are the huge Young modulus (~ 1 TPa), ballistic transport at room temperature for nanotubes with the mean-free-path length around 1 μm and where the mobility in semiconducting CNTs could attain even $120\,000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ at a moderate electric field of 10 kV/cm, and the band engineering of CNTs obtained by the rolling of a graphene sheet at different angles, accompanied by applied transverse electric fields or pressure. Moreover, the band gap of the semiconducting CNTs is

proportional with d^{-1} , the diameter of the CNT, and thus the band engineering of CNTs is easily achieved, making them a strong competitor of AIII-BV semiconductors. A very recent review of all these aspects regarding CNTs is found in Ref. 9. So, the microwave and millimeter wave applications of CNTs could be very promising. A very important application is a gas sensor based on the gas-induced variations in the dielectric properties of CNTs (see Ref. 10 and the references herein). Here, different gases are sensed by the change of the dielectric constant in the microwave range due to the absorption of the gas molecules by CNT mixtures, films, or bundles. Thus, the measurement of the CNT propagation constants (attenuation and electric permittivity) in the microwave and millimeter wave range is a very important issue that constitutes the main aim of this letter. We have to also point out that the measurement of the CNT composites or purified CNTs is an issue of constant interest in applied physics due to the applications described above or their use as microwave absorbers.^{11,12,13} However, due to various composite compositions and due to the circuits in which these composites are introduced to be characterized, very often the measured propagation constants are quite different from one experiment to another. Therefore, we propose an experiment based on a mixture of double-walled CNTs (DWCNTs) displayed in Figs. 1(a)1(b).



(a)



(b)

Fig. 1. (a) SEM of the DWCNTs mixture. (b) SEM of a few DWCNTs where their geometry is evidenced.

The DWNTs are produced by the catalytic decomposition of methane over a CoMo-MgO catalyst.¹⁴ They are free of amorphous carbon contamination, and the use of a MgO support allows its easy removal¹⁵ without damaging the DWNTs. The high purity of the DWNTs has been thoroughly characterized by different techniques including high-resolution transmission electron microscopy (HRTEM),¹⁴ Raman spectroscopy,^{14,16} photoluminescence,¹⁷ electron diffraction,¹⁸ and the electrical properties of individual DWNTs have been investigated with or without applied magnetic fields (up to 35 T).^{19,20} So, using the method of preparation and various characterization methods developed by Flahaut *et al.* we have finally disposed of DWCNTs that are very pure and with known and verifiable electrical, magnetical, and optical properties.

We have introduced the mixture in a coplanar waveguide (CPW) supported on a thin dielectric membrane²¹ (see Fig. 2 for all geometrical values and configurations; DWCNTs have an average diameter of 2 nm and an average length of 10 μm), which, in the absence of the CNT material, displays an almost dielectric constant near 1 and so very low losses in a very wide band of frequencies 1–65 GHz. This almost lossless microwave CPW supported on a dielectric membrane is a high frequency propagating device terminated on both sides by standard 50 Ω lines and realized via the micromachining techniques of a 400 μm thick semi-insulator Si substrate. The line process is divided into three main steps. First, a 1.4 μm thick dielectric membrane, which is composed of both silicon oxide and nitride, is deposited. The line metallization is then performed by gold electroplating into a thick photoresist mold. Finally, the suspended coplanar lines are obtained through silicon substrate micromachining thanks to a KOH bath.²² The mixture of the CNTs was manually introduced in the cavity of the micromachined CPW with the help of a tweezer. A uniform deposition with a high filling rate exceeding 90% of the CNTs near the CPW membrane was then manually performed. Then, the backside of the CPW filled with CNTs was sealed with an adhesive band.

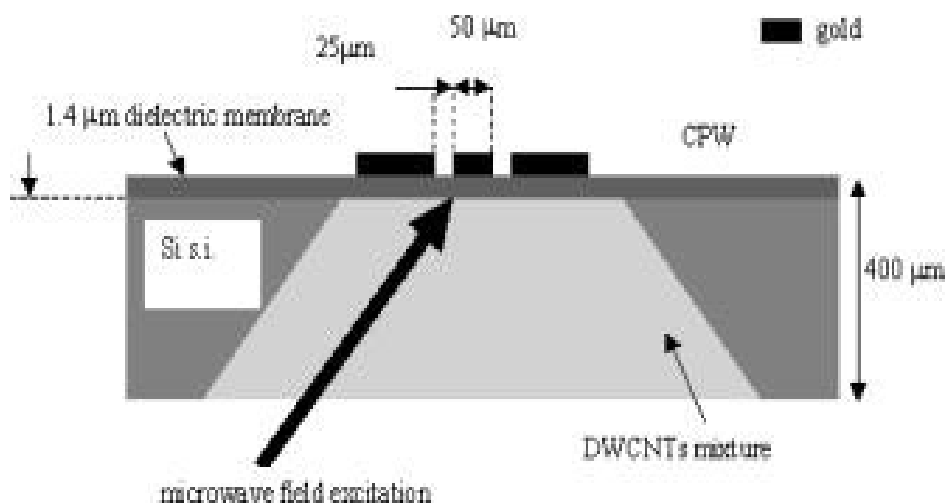


Fig. 2. CPW filled with a DWCNT mixture.

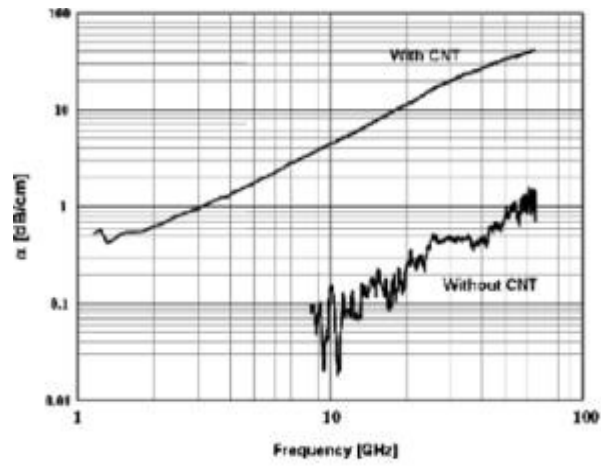
Further, by using calibrated measurements, the propagation constants of DWCNTs were found in the range of frequency encompassing microwave and millimeter waves. In this respect, the S parameters of different CPW lines with the lengths L of 2, 4, and 6 mm were measured using a network analyzer in the range of 1–65 GHz in both cases, i.e., empty and filled with DWCNTs. Using the methodology described in Ref. 22, where the complex propagation constant is determined from the S parameter measurements of two CPW lines having lengths L_1 and L_2 , we have measured and computed several times the S parameters of the CPW not filled and filled with DWCNTs having the lengths mentioned above and compared various computations to verify the accuracy of our results. The attenuation and the relative electrical permittivity are displayed in Fig. 3. The complex propagation formula is given by²³

$$\gamma = \frac{1}{\Delta L} \arg \cosh \left(\frac{1}{2} \frac{S_{11}^{L2} S_{22}^{L1} + S_{11}^{L1} S_{22}^{L2} - \det[S^{L1}] - \det[S^{L2}]}{S_{21}^{L2} S_{12}^{L1}} \right), \quad (1)$$

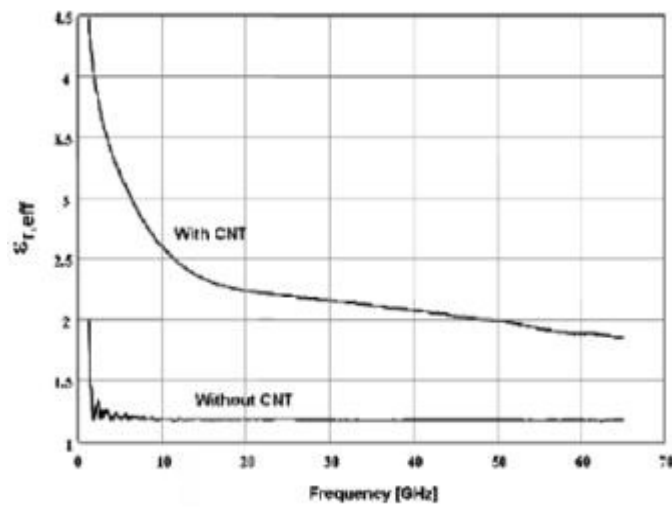
where $\Delta L = |L_1 - L_2|$, the superscript indicates the S parameters for a specific length, and where

$$\gamma = \alpha + i\beta = \alpha + i2\pi f \epsilon_{r,\text{eff}}^{1/2} / c, \quad (2)$$

where α and $\epsilon_{r,\text{eff}}$ are the attenuation and the relative effective permittivity to be determined, f is the microwave frequency, and c is the speed of light.



(a)



(b)

The attenuation α (dB/cm) and the relative effective permittivity $\epsilon_{r,eff}$ are represented in Figs. 3(a)3(b), respectively. It can be seen that the attenuation in the CPW filled with DWCNTs is one magnitude higher than the lowest possible, i.e., when the CPW is empty.

Using the above measured data we have performed an electromagnetic simulation using high frequency structure simulator (HFSS) software to determine the microwave field penetration in the CNT material and implicitly to verify the high filling factor of the nanotubes under the CPW. The result of the simulation is displayed in Fig. 4; in the case of the exciting frequency of 10 GHz, $\epsilon_{eff}=2.6$, $\alpha=4$ dB/cm, $\sigma=1.57$ S/m, and a CPW gold thickness of 2 μ m. In this figure the lightest colors are assigned to the highest field intensities. We can see that the electromagnetic field extends deeply in the CNT material, the penetration depth being greater than 40 μ m, which is 25 times larger than the thin Si membrane. Also from this figure we can see the typical field propagation in the CPW structures where the field is propagating at the air/dielectric interface and where the field

asymmetries are due to the losses in the CNT material under the dielectric membrane.

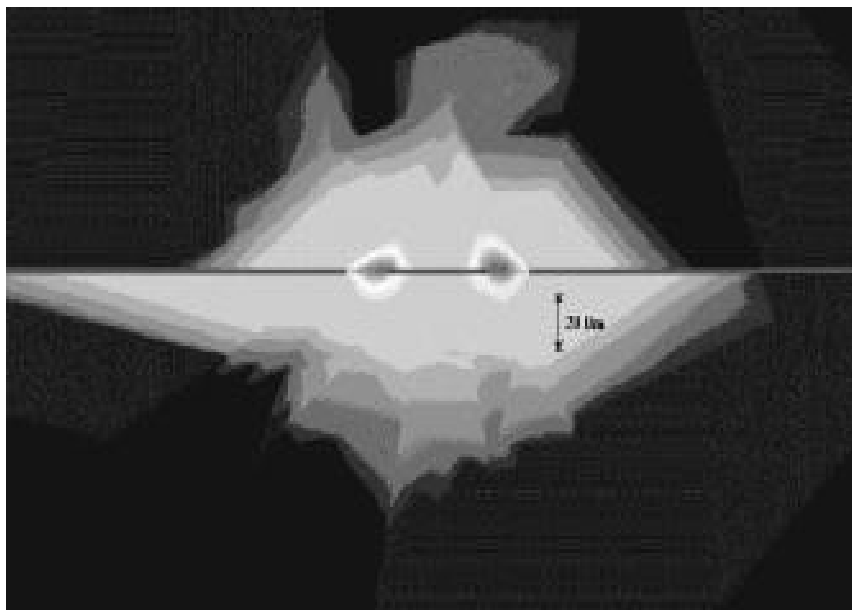


Fig. 4. The microwave electric field distribution in the DWCNT CPW lines at 10 GHz. The three long black lines are the metallic CPW gold lines having a thickness of 2 μm .

The relative effective permittivity of the CPW filled with DWCNTs is varying very abruptly in the range of 1–10 GHz, while in the case of an empty CPW the relative effective permittivity is almost constant and is 1.2 in the range of 1–65 GHz. The abrupt variation of the effective permittivity of the CPW filled with DWCNT, which is about 0.3/GHz, is very useful in the case of gas sensors. Thus, very sensitive integrated gas sensors can possibly be realized with the help of a filled CPW supported on dielectric membranes or resonators based on them.

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FOOTNOTES

^aAuthor to whom correspondence should be addressed; present address: LAAS CNRS, 7 Avenue du Colonel Roche, 31077 Toulouse Cedex 4, France; electronic mail: mdragoma@laas.fr

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