

Electromagnetic absorbing properties of graphene–polymer composite shields

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ABSTRACT

A graphene-based composite, consisting of a thermosetting polymeric matrix filled with multilayer graphene microsheets (MLGs), is developed for application in thin radar absorbing materials. An innovative simulation model is proposed for the calculation of the effective permittivity and electrical conductivity of the composite, and used for the electromagnetic design of thin radar absorbing screens. The model takes into account the effects of the MLG morphology and of the fabrication process on the effective electromagnetic properties of the composite. Experimental tests demonstrate the validity of the proposed approach and the accuracy of the developed simulation models, which allow to understand the interaction mechanism between the incident electromagnetic field radiation and the MLG-based composite. Two dielectric Salisbury screen prototypes with resonant frequency at 12 GHz or 12.5 GHz and total thickness of 1.8 mm and 1.7 mm, respectively, are fabricated and tested. The results and technique proposed represent a simple and effective approach to produce thin absorbing screens for application in stealth technology or electromagnetic interference suppression.

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1. Introduction

In recent years, electromagnetic (EM) absorbers have gained a fundamental role in civil and military applications. Their use is a major factor in radar cross section reduction, and they can be employed also to reduce EM interference and electromagnetic compatibility problems caused by the GHz range radiation. One of the first concepts in the design of a radar absorbing material (RAM) was the dielectric Salisbury screen (DSS) [\[1\],](#page--1-0) which is a three layer panel consisting of: a metal plane acting as a reflector, a lossless dielectric layer of thickness equal to a quarter of the wavelength of the radar wave to be absorbed, and a thin lossy layer having the function of absorbing the energy associated to the electromagnetic field.

The main limitation of the DSS is the thickness that, for applications from a few up to 18 GHz, is in the range of centimeters.

To obtain broad-band, lightweight, and thin EM absorbers in the GHz frequency region, significant scientific and technological interest has been focused on custom-tailored RAMs. They are made with polymer or ceramic based composites having a high loss energy, which enables them to absorb the incident radiation and dissipate it as heat [\[1\].](#page--1-0) Carbon fiber reinforced polymeric composites have been widely used for specific application in EM interference suppression and as RAMs [\[2\]](#page--1-0), [\[3\].](#page--1-0)

More recently, several studies have investigated the use of single-walled or multiwalled carbon nanotubes (CNTs) as well

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as carbon nanofibers (CNFs) $[4-12]$, as replacement of more traditional fillers in composites for RAM applications.

In previous studies [\[11–13\],](#page--1-0) the authors have investigated the use of multifiller systems in order to tailor separately the real and imaginary parts of the complex effective permittivity of the composite, with the aim of optimizing RAM absorption properties and minimizing the thickness of DSSs. To this purpose, the use of different conventional micro- and nano-fillers was combined [\[11–13\].](#page--1-0) In fact, it was observed that at radio frequency (RF) the effective electrical conductivity of the composite, which is related to the imaginary part of the complex permittivity, is strongly affected by the filler aspect ratio (FAR), which represents the ratio between the maximum and minimum filler dimensions. For filler volume fractions (FVFs) below the percolation threshold, it results that the effective conductivity at RF reaches values of a few up to ten siemens-per-meter in case of FAR in the range 100–1000. Therefore, nano-fillers like CNTs or micro-fillers like short carbon fibers, having diameter around $7 \mu m$ and length of few millimeters, are particularly suitable for this scope. On the contrary, the real part of the effective permittivity, which is representative of the dielectric polarizability of the material, is not so sensitive to the FAR but it increases proportionally with the FVF, and for low FAR (i.e. below 100) it is characterized by a constant trend over frequency. Therefore, it is evident that the possibility of combining micro- and nano-fillers with different FAR at different dimensional scales represents an opportunity to tailor separately the dielectric properties and the electrical conductivity at RF of the composite.

In this context multilayer graphene microsheets (MLGs) can represent a valid alternative to the combined use of carbon-based micro- and nano-fillers. MLGs are small stacks of graphene sheets, having thickness typically in the range 1– 10 nm, and lateral linear dimensions much greater, varying from about 1 μ m up to 20–25 μ m [\[14,15\]](#page--1-0).

Due to their bi-dimensional shape, MLGs can play the role of a nano-filler with a high FAR (intended as the ratio between the lateral dimension of the platelet, and its thickness). At the same time, being the platelet area in the range of several tens up to hundreds microns square, MLGs play the role of a low-FAR micro-filler, having an influence on the real part of the effective permittivity of the composite.

For all the aforementioned reasons, an extensive study concerning the characterization in the entire X and Ku frequency bands (namely from 8 GHz up to 18 GHz) of the effective complex permittivity of MLG-filled polymeric composites has been carried out [\[16\].](#page--1-0) Moreover, a novel simulation model has been developed to predict the complex dielectric permit-tivity of such composites at RF [\[17\].](#page--1-0)

The EM interference shielding properties of graphenebased composites have been investigated in the latest years considering different polymeric systems [\[18–21\]](#page--1-0). Very recently, a graphene/polymer composite film in a sandwich structure has been presented in [\[22\]](#page--1-0) for application as a flexible shield. The composite film was loaded with a high filler content (from 10% wt to 60% wt), and it was proved to provide a high reflection against incident EM fields, with shielding effectiveness up to 27 dB.

In this paper, the authors investigate the feasibility of graphene-based RAMs. In particular thin DSSs, having total thickness below 2 mm, were designed and fabricated at Sapienza Nanotechnology and Nanoscience Laboratory (SNN-Lab). They have a lossy sheet made of an epoxy-based vinyl ester resin filled at 2% wt. MLG, and spacer made either with a commercial Rohacell®IG51 (RC) panel or a polypropylene (PP) layer, in order to obtain a resonant frequency of 12 GHz or 12.5 GHz. The EM design of the absorbing screens is performed by calculation. To this purpose an innovative EM model, accounting for the MLG morphology and the composite production process, is developed to predict the effective permittivity and electrical conductivity of the material.

2. Experimental

2.1. MLG and composite production

MLGs were produced through thermal expansion of a graphite intercalation compound (GIC) [\[16\].](#page--1-0) The starting GIC (Grafguard 160–50 N) was provided by Graftech Inc., and it is characterized by a declared mean lateral size of $350 \mu m$. GIC underwent a thermal shock driven expansion in air at 1150 °C for \sim 5 s, increasing its volume by roughly 200 times, obtaining worm-like graphite.

The resulting expanded graphite was dispersed in acetone. The obtained suspension was tip sonicated using an ultrasonic probe working at the frequency of 20 kHz, running for a time of 20 min, and set in pulse mode (1 s on and 1 s off), thus obtaining MLGs [\[16\].](#page--1-0)

Polymer composites were prepared by using an epoxybased vinyl ester product, kindly provided by Reichhold (DION 9102). This resin has an initial viscosity of 150–200 mPa s, a density of 1.01-1.05 g/cm^3 , and a styrene content around 50% wt.

After the sonication, the MLG-acetone mixture was poured into a beaker containing the liquid vinyl ester resin, which had been previously mixed with 0.2% wt of a Co-based accelerator, and further sonicated for 30 s using a low ultrasound amplitude, with the aim of minimizing bubble formation.

Next, the suspension was magnetically stirred at 200– 250 rpm, in order to remove the solvent in excess. Upon complete evaporation of the solvent, the hardener was added at 2% wt ratio.

The liquid formulation was finally again stirred at 250 rpm until it gained a viscosity suitable for casting, and then it was poured directly in two sets of X and Ku flanges [\(Fig. 1](#page--1-0)(a)), suitable for effective permittivity measurements in the frequency range from 8.2 GHz to 18 GHz.

Finally, the composites were cured in air for 24 h and postcured for another 24 h at 70 \degree C, and after final polishing the filled flanges [\(Fig. 1\(](#page--1-0)b)) were used for electrical permittivity measurements as described in Section 2.3.

2.2. Fabrication of the radar absorbing shield

[Fig. 2](#page--1-0) shows the basic configuration of a Salisbury absorbing screen illuminated by a plane wave with normal incidence. The first layer from the right side consists of a thick metal

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