Multiscale Metacomposites incorporating Carbon Nanotube-Ferromagnetic Microwire Hybrid Fibers

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I. Introduction

The distinguished giant magnetoimpedance/giant stress-impedance effect and soft-magnetic properties of the microwires combined with the high electrical conductivity and remarkable mechanical properties of carbon nanotubes CNT could provide a unique signature in response to electromagnetic (EM) waves and suggest that they can be feasible fillers to constitute metacomposites. Our previous studies have elucidated the benefits of ferromagnetic wires on realizing DNG features. To date, metacomposites containing either Fe-based or Co-based amorphous wires in parallel or orthogonal arrays and hybrid metacomposites derived from them have shown metamaterial characteristics such as negative permittivity and permeability that are tunable with external magnetic field [1]. However, such metacomposites still suffer from the drawback of narrow band for left-handed properties and inefficient transmission tunability. We propose to meet this challenge by acting on multisccale, i.e., three different levels (nano-micro-macro) and fully exploit the intrinsic nano-scale properties of CNTs to transfer them to the metacomposite on the macro-scale.

II. Experimental Details

In the first step, our objective was to grow multi-walled CNT (MWCNT) on microwires by electrophoretic deposition technique (EPD) as an economical and versatile alternative to the conventional chemical vapor deposition process (CVD) [2]. Figure 1c shows the schematic representation of the EPD setup used to grow CNT on the microwires surface. EPD is fundamentally a combination of two processes, electrophoresis and deposition. To prepare the CNTs suspension for the EPD process, the CNTs were chemically oxidized in a mixture of H_2SO_4, $KMNO_4$, $NaNO_3$ and H_2O_2 to introduce carboxyl acid groups on the CNT surface and provide them with a negative surface charge (Fig. 1a).

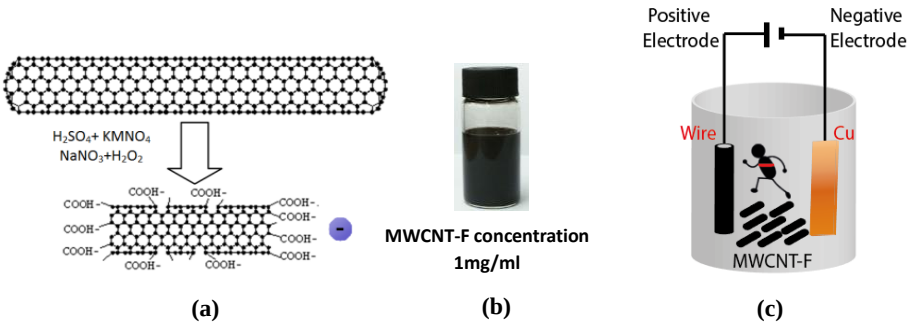

Fig. 1 a EPD deposition of CNTs on wire surface **b** Functionalization of CNTs **c** Dispersed CNT solution

SEM images demonstrate that the functionalized MWCNT were successfully deposited on the surface of the wire by EPD process without application of any binding material (Figure 2b). By a proper selection of EPD parameters (voltage, deposition time), we were able to modify the morphologies of the deposited layers.

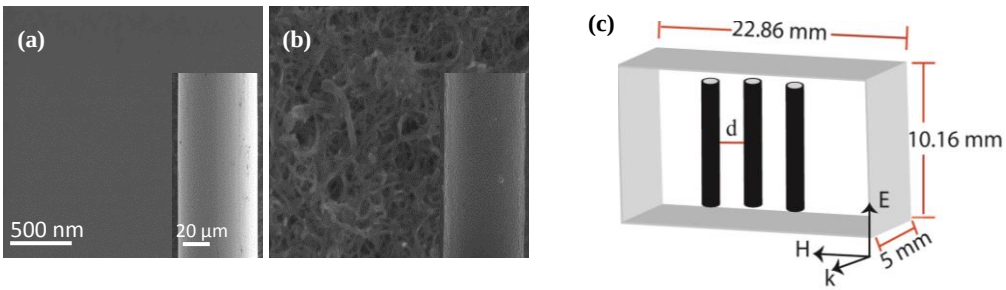

Fig. 2 SEM images of the surface of microwire **a** pristine state **b** CNTS deposited with EPD process; **c** Composite configuration

III. Results and Discussions

Figure 3a shows the complex permittivity ε' and complex permeability μ' of the composites containing the hybrid fibers (configuration shown in Fig. 2b, *d*=3mm) measured by using a WR-90 waveguide in TE10 dominant mode. For the uncoated wire, we can clearly identify a double negative region (DNG) in the range of 10-11GHz without the application of any external stimuli (considered as natural "metacomposite" feature). The DNG region is expanded and shifted to higher frequencies with the incorporation of carbon nanotube coating of 1.73μm. However, with increasing the CNT thickness, the DNG region narrows substantially and moves at frequencies below 10GHz. In this case, the strong interfacial polarization is responsible for the shifting of the plasma frequency f_p to lower frequency, from 10.51GHz for the uncoated wire to 9.61GHz for the hybrid fiber with 3.11μm coating thickness. The sharp peak of μ' for 3.11μm sample, reflects the magnetic coupling between CNT layers and CNT-microwire resulting in the enhancement of the magnetic response for these hybrid fibers.

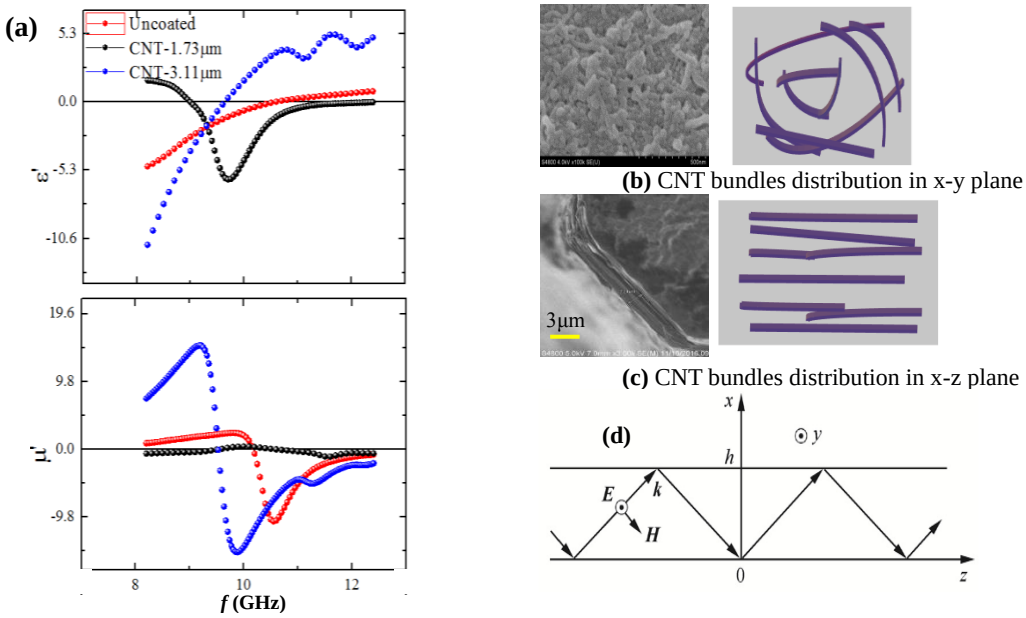

Fig. 3 a ε' and μ' spectra of composites containing fibers with different CNT coating thickness; **b, c** Schematic diagram of split closed path made of CNT bundles; **d** Propagation of TE_{10} wave rectangular waveguide

In order to explain the role of carbon nanotubes on left-handed properties of our composite system, we should take a close look at the morphology of the coating. From Fig. 2b and Fig. 3b we can appreciate the random nature of the CNT coating, consisting of numerous bundles intertangled and with disordered arrangement. These CNT bundles construct closed paths. Although the path is closed in the x-y plane (Fig.3a), splits exist between bundles in x-z plane (Fig.3c). The split closed path made of CNT bundles is analogous to split-ring resonator (SRR), which is the conventional unit of artificial negative permeability material [3]. Base on magnetization and circuit, effective permeability of SRR will have a negative real part when the frequency of the incident wave is in a special range above the resonance frequency of SRR. On the other hand, in the waveguide measurement, magnetic field vector H has a component perpendicular to the plane of these split closed paths, while electrical field vector E is parallel to split closed paths [3] as shown in Fig. 3(d). The amount of split closed paths through thickness direction is limited for a single layer. Multilayer CNT film is "easier to behave" negative μ' than monolayer CNT coating, which is proved by our results when the thickness of the coating is 3.11μm.

IV. Conclusions

We successfully prepared composites containing fibers CNT-ferromagnetic microwire hybrid fibers by EPD process. The metacomposite properties of CNT-microwire hybrid fibers depend affinely on the morphology of deposited CNT layer; a house-of-card structure is in favor of negative permeability. Split closed paths consisting of CNT bundles along with the induced interfacial polarization are proposed to explain effects of CNT on the dispersion of constitutive parameters, respectively, of the hybrid fibers.

References

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