Polymer-Carbon Nanotube Sheets for Conformal Load Bearing Antennas

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***Abstract*—We propose a conductive carbon nanotube (CNT) sheet to realize conformal antennas on polymer substrates. Polymer-ceramic composites (rubber-like structures) have good RF (high dielectric constant and low loss tangent) and desirable mechanical properties (conformal, flexible and lightweight). However, there is a challenge in printing metallization circuits on polymer substrates due to their hydrophobic nature. Also, they are associated with low metal-polymer adhesion, causing peeling under stain or tensile stresses. To address these issues, in this paper, we consider the approach of embedding high density vertically-aligned carbon nanotubes within the polymer composite to achieve a CNT sheet having high structural compatibility. We present the fabrication process to achieve high conductivity CNT sheets and construct a sample polymer-CNT patch antenna, yielding a 5.6 dB gain. This is only 0.8 dB lower than that of an ideal patch made of perfect electric conductor (PEC). Strain and tensile tests are also carried out to evaluate electrical performance of the polymer-CNT sheet as it is bent and stretched. Our measurements show that the proposed conductive polymer-CNT sheet is highly flexible and preserves good conductivity under small bending and stretching. The CNT sheet retains acceptable performances even after 100° bending and 13% stretching. The proposed polymer-CNT sheets are well suited for load bearing antenna applications.**

***Index Terms*—Carbon nanotube sheet, load bearing antenna, polymer printing, polymer-ceramic composite, stain and tensile stresses.**

I. Introduction

Conformal lightweight polymer-based materials are important for load-bearing antennas and multilayer RF front-ends for small aircrafts and body-worn applications. There is also interest for multilayer three-dimensional RF front-end architectures with each layer bearing different functionalities. Such conformal antenna and multilayer circuit structure call for a new class of materials with desirable electrical/RF (low loss, high permittivity), mechanical (flexible, lightweight, strong shear and tensile rating) as well as thermal properties.

Among available materials, polymer composites (such as PDMS-D270 and PDMS-MCT composites) are attractive because they are extremely flexible and not as sensitive to large temperature variations. They also have low loss ($\tan\delta < 0.02$) up to several GHz and controllable dielectric constants (relative permittivity of $\varepsilon_r = 3 \sim 20$) [1], [2]. Further, as compared to other materials such as those based on liquid crystal polymers (LCP) [3] and low temperature co-fired ceramics (LTCC) [4], PDMS composites can be processed at room temperature. Specifically, for bonding LCP and LTCC layers, they must be heated (300°C for LCPs and 1000°C for LTCCs) at temperatures that could cause failure to some IC components and fragile wire bonds. With these issues in mind, PDMS composites are well suited for conformal load-bearing antennas and RF systems integration. However, metallization or printing on PDMS composites remains a challenge. Specifically, common lift-off lithography methods using metal evaporation does not work well for PDMS due to poor metal-polymer adhesion [5]. Further, interface incompatibilities can easily cause detachment of the printed layers under bending or tensile stress.

In this paper, we propose a novel polymer-printing technology utilizing carbon nanotube (CNTs) sheets. Specifically, multiwalled carbon nanotubes (MWNTs) are vertically grown out of the polymer surface in a similar way as body hair, but in much higher density (about 3×10^9 nanofibers per cm^2) to form a CNT sheet with nanotube lengths being a few hundred micro-meters. Since the CNTs are embedded into the polymer, they do not peel-off as is the case with metallization typically done via evaporation. More importantly, by controlling the length and density of carbon nanotubes, high surface conductivity is achieved with concurrent mechanical flexibility, high conformality and good structural compatibility. Below, we first describe the equivalent circuit model of the CNT sheet and its fabrication process. Then, a polymer-CNT patch antenna is "printed" and measured as an application example. The flexibility and stability of the polymer-CNT sheet are subsequently evaluated by carrying out strain and tensile tests. At the end of the paper, we also present measurements for two examples of conformal CNT patch antennas on a cylindrical surface.

II. Carbon Nanotube Sheet Printing on Polymers

Carbon nanotubes (CNTs) have drawn significant attention in the RF community due to their superior mechanical properties and potential applications to antennas. Specifically,

Manuscript received March 27, 2009; revised May 21, 2009; accepted January 31, 2010. Date of publication April 22, 2010; date of current version July 08, 2010. This work was supported in part by the U.S. Air Force Research Laboratory.

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Digital Object Identifier 10.1109/TAP.2010.2048852

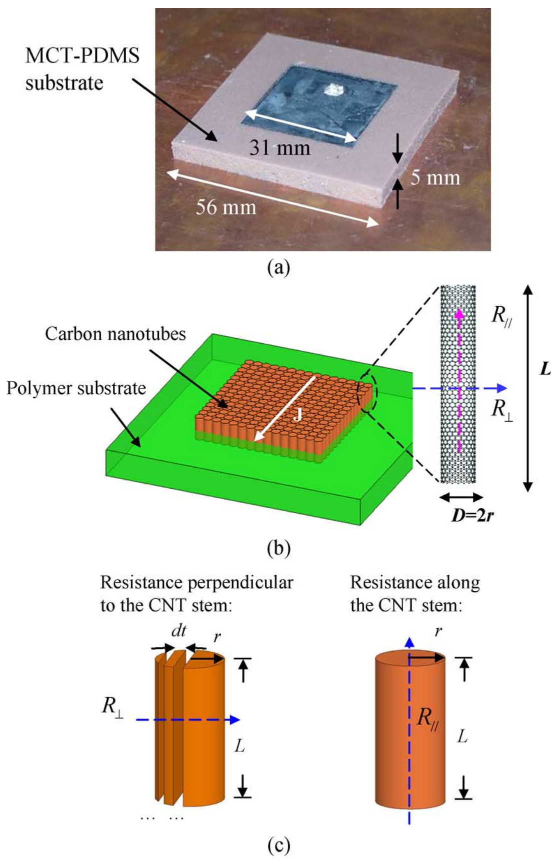


Fig. 1. Illustration of the vertically aligned polymer-CNT sheet (in actual and model form). (a) Printed polymer-CNT patch. (b) Illustrative model of vertically aligned CNTs. (c) Ohm's law for a single CNT.

metallic CNTs do not oxidize [6] and are not susceptible to moisture [7], [8]. CNTs are also stable at high temperatures up to 700°C [6], a desirable characteristic for harsh and high temperature environments. Further, CNTs are attractive in realizing antennas at millimeter wave, even up to optical frequency range [9]–[13]. For example, it has been reported that a single CNT dipole exhibits significantly slower wave velocities ($v_p = 0.02c$, where c is the speed of light) above the relaxation frequency of around 53 GHz [9], [11]. However, so far, reported antennas have exhibited low radiation efficiencies due to their high resistive loss [9], [11]. This is common for all nano-radius wires including CNTs, which have large intrinsic resistance (around 6 $\mathrm{k}\Omega/\mu\mathrm{m}$), viz. too lossy for most microwave applications. However, CNT ensembles or arrays have shown to have improved surface conductivity [14]. For example, nonaligned CNT ensembles have been reported to reduce sheet resistance down to around $20\,\Omega/\mathrm{square}$ [15]. However, even this lower resistance is still too high to realize efficient CNT antennas. Alternatively, e-textiles have been pursued to improve sheet conductivity by weaving the nanotubes within cotton fibers [16]. Nevertheless, flexibility and polymer-CNT adhesion under bending stress are compromised with cotton microfibers. In this paper, we propose to grow vertically aligned CNTs in the polymer substrate [Fig. 1(a)]. Using this approach, we aim to address conductivity and CNT printing issues on polymers.

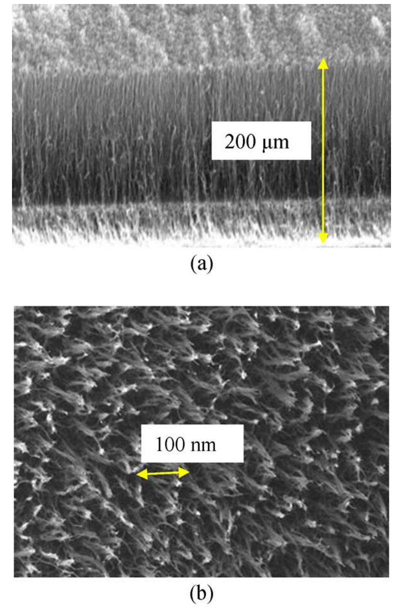


Fig. 2. SEM photograph of the CNT array. (a) Cross-section view of the CNT array. (b) Top view of the CNT array.

A. Circuit Model of the Vertically Aligned CNT Sheet

Fig. 1(a) shows a CNT sheet printed on a PDMS composite substrate. Since the CNTs are grown vertically and embedded inside the polymer substrate, the polymer-CNT sheet can be treated as a composite, where the fillers being the carbon nanotubes. The composite's conductivity is, of course, dependent on the CNT density. In accordance with percolation theory [17]–[19], the conductivity of the polymer-CNT sheet, σ, is given by

$$\sigma \propto (P - P_c)^{\alpha} \tag{1}$$

where P is the CNT volume density, P_c is the percolation threshold density, and α is an exponent related to the conductivity. To obtain an explicit expression for the conductivity, we proceed to model the CNT sheet as an array of touching nanowires [Fig. 1(b)]. This simplified model is valid only for high density CNT sheets, and provided the nanotubes are entangled as shown in Fig. 2(b). In this case, the electric current flows perpendicular to the nanotube stems via the vertically touching CNTs.

We can calculate the CNT sheet resistance by using the circuit model in Fig. 3. This is a parallel circuit of resistors, each representing the resistance of individual CNTs in perpendicular direction ($R_\perp$). Referring to Fig. 3, the CNT surface resistance is calculated to be

$$R_s \approx \frac{(N \cdot R_\perp)}{N} = R_\perp \tag{2}$$

where N is a constant related to the CNT density (in essence the number of CNTs). As shown in Fig. 1(c), the resistance along the perpendicular direction can be calculated by adding up the

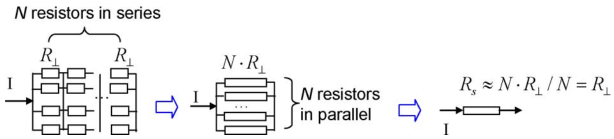


Fig. 3. Model of the CNT array for sheet resistance calculation.

resistance of each segment of length dt and cross-sectional area $L \cdot 2\sqrt{r^2-(r-t)^2}$, giving

$$R_\perp = \int_{-r}^{r} \frac{\rho dt}{L(2\sqrt{r^2-(r-t)^2})} = \frac{\pi}{2} \cdot \frac{\rho}{L} \propto \frac{1}{L} \tag{3}$$

with ρ having the effective resistivity of a single CNT. By comparison, when the current flows along the tube's length, the resistance becomes [see Fig. 1(c)]

$$R_{//} = \rho \frac{L}{\pi r^2} \propto \frac{L}{r^2}. \tag{4}$$

Since $L \gg r$, then $R_{//} \gg R_\perp$, and this is the primary reason for the low $R_\perp$ values, making the proposed CNT arrangement attractive for antennas and RF printing.

As predicted by the above simplified CNT model [Fig. 1(b)] and (3), the CNT density and length are two key parameters to achieve high conductivity. High CNT density (or small spacing distance between CNTs) will increase CNT percolation, leading to higher conductivity. As our fabrication limits for the CNT separation distance are around 100 nm, we tried to compress the CNT arrays to reduce spacing distance down to around 20 nm. Doing so, the corresponding resistance was reduced by 50 times. However, the resulting CNT sample was too small for antenna fabrication. Therefore, as a next step, we will carry out larger area fabrication for the compressed ensembles. Increasing CNT length (L) can also reduce resistance. However, this holds for highly entangled CNT arrays. In practice, the CNT array is less entangled as we increase the length. Therefore, increasing length may not improve conductivity beyond certain point (around $100 \sim 200\ \mu\text{m}$). In the next sections, we focus on the fabrication and characterization of such vertically aligned CNT array. When the nanotubes are about 200 μm tall and spaced about 100 nm apart, the achieved DC sheet resistance of the sheet is about 1 Ω/square. This represents a significant decrease as compared to the single CNT dipole [9] or the non-aligned CNT ensemble [15].

B. Carbon Nanotube Sheet Fabrication Process

The vertically aligned CNTs are synthesized via a chemical vapor deposition (CVD) process [20] (see Fig. 4). The process is as follows: First we sputter an array of ferrous particles on a silicon wafer to serve as catalysts for CNT growth. Next the silicon substrate is placed inside a tube furnace (Thermolyne 79400) and methane gases (CH_4) are blown into the furnace via a carrier argon flow. At high temperature (1000°F), methane gases are decomposed into carbon atoms, aligned along the catalyst particles into cylinder forms. By controlling the furnace temperature (1000°F) and deposition time (2 hours), we can achieve a vertically aligned CNT array on the silicon wafer as shown in Fig. 2.

At the 2nd step we transfer the CNT sheet onto the polymer substrate using a two-stage curing process. First a thin PDMS composite layer is spin-coated onto the CNT sheet as displayed in Fig. 5. After curing, the CNTs are implanted inside the thin polymer layer to form a polymer-coated CNT sheet. The polymer-coated CNT sheet is then detached from the silicon

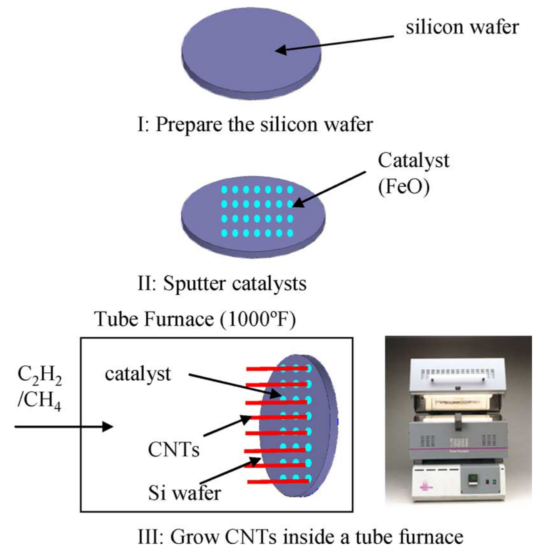


Fig. 4. Process for growing vertically aligned CNTs.

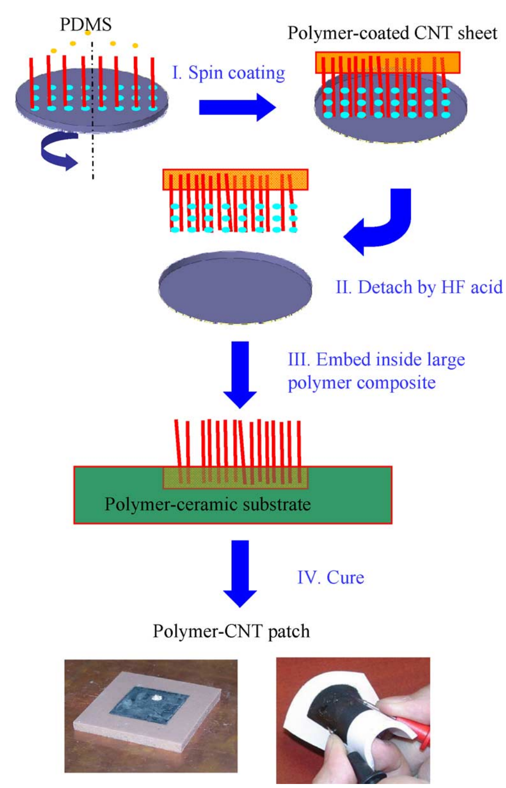


Fig. 5. Process for transferring the CNT sheet onto the polymer composites.

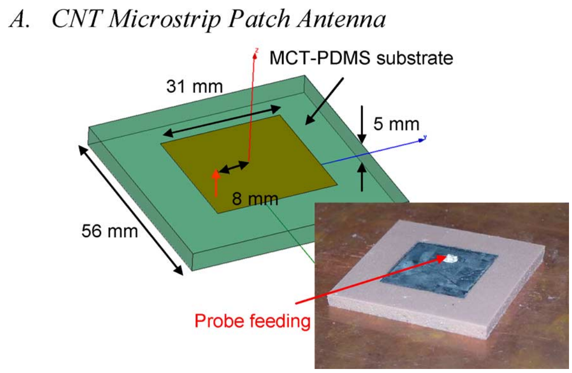


Fig. 6. Simulation model and fabricated polymer-CNT patch.

wafer by dissolving SiO_2 on the Si surface using hydrofluoric (HF) acid. During this process, we observe a $\sim 2\%$ shrinkage of the polymer, further increasing the CNTs density and improving conductivity. In the final (and 3rd) step, we embed the polymer-coated CNT sheet into a larger customized polymer-ceramic substrate for antenna realization. During this curing stage, the polymer-ceramic substrate cross-links with the coated polymer, leading to a strongly bonded CNT sheet on the polymer-ceramic substrate.

III. RF and Mechanical Performance Evaluation

We now proceed to characterize the RF and mechanical properties of a fabricated CNT antenna. Specifically, a sample polymer-CNT patch antenna was designed and fabricated as shown in Fig. 6. We then measured the CNT-polymer antenna performance and compared it with simulations for a perfectly electric conductor (PEC) patch on a similar substrate. This was followed by strain and tensile tests to examine the flexibility and stability of the CNT sheet under load bearing conditions. An evaluation of CNT patch under conformal installation was also carried out.

A. CNT Microstrip Patch Antenna

Referring to Fig. 6, we show a 31 mm $\times$ 31 mm CNT sheet on a 56 mm $\times$ 56 mm PDMS-MCT substrate, having a dielectric constant of $\varepsilon_r = 3.8$ and loss tangent of $\tan\delta = 0.015$ at the resonant frequency of 2.25 GHz. The patch was fed by a coaxial probe soldered to the CNT sheet by conductive epoxy. The polymer-CNT patch was then measured on a 150 mm $\times$ 150 mm ground plane at the Ohio State University-ElectroScience (OSU-ESL) anechoic chamber. The measurement data were then compared with HFSS simulations for perfectly electric conductor (PEC) patch and a finite conductivity patch (surface resistivity: 0.9 Ω/square). As shown in Fig. 7, the measured gain (blue solid curve) agrees well with the finite conductivity patch (dashed curve), verifying the low sheet resistivity (0.9 Ω/square) of the polymer-CNT sheet. Further, the radiation pattern [Fig. 7(c)] is that of typical patch antenna. More specifically, the measured gain of the CNT patch was 5.6 dB, i.e., only 0.8 dB lower than that of the simulated PEC patch (dotted curve) of the same dimensions and substrate dielectric properties. Indeed this is a very good RF performance for practical applications.

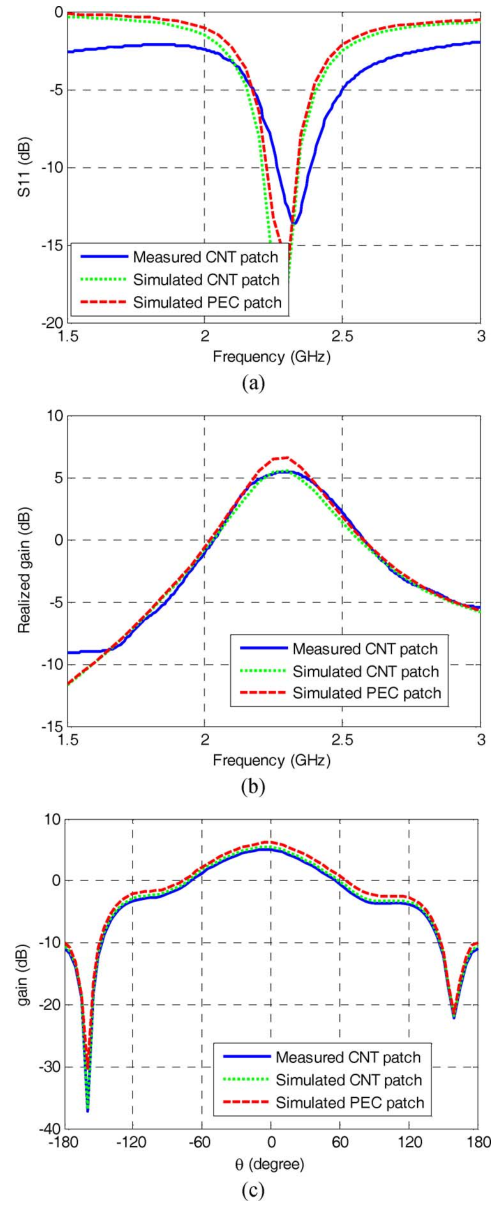
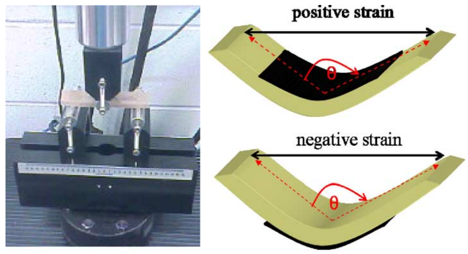


Fig. 7. Measured RF performance of the polymer-CNT patch antenna. (a) Reflection coefficient (S11). (b) Realized gain. (c) E-plane pattern.

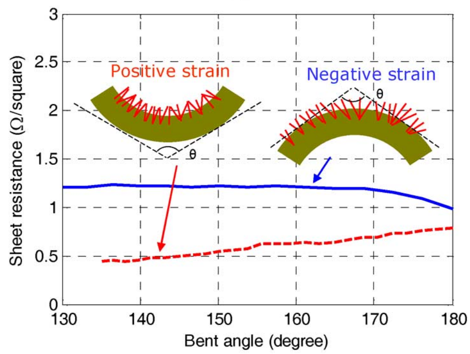
B. Strain and Tensile Tests

As load bearing antennas are subject to vibration and temperature change, they are usually deformed by bending or stretching. Therefore, it is critical to characterize their electrical properties under strain and tensile stresses. Specifically, it is desirable that the CNT sheet conductivity remains stable as the substrate it bent or stretched. Here, we only measure the CNT sheet DC resistance under different stain and tensile stresses (which is related to the RF performance of the CNT surface).

Fig. 8(a) shows the measurement setup for the bending test. As seen, we clamped a sample polymer-CNT sheet at the two ends and exerted an external force using a universal test machine to deform the polymer substrate. The deformation was evaluated by the degree of bent angle (θ). The DC sheet resistance was subsequently measured and recorded as the sample was deforming. Fig. 8(b) gives the measured sheet resistance versus ap-



(a)



(b)

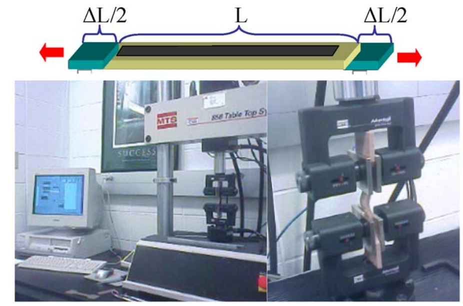
Fig. 8. DC sheet resistance versus strain. (a) Measurement setup. (b) DC sheet resistances versus strain.

plied strain. We observed that the DC sheet resistance was fairly stable within a large range of angles up to $+/-130°$. As expected, the resistance decreases when positive strain is applied since the CNTs are pushed towards each other. In contrast, negative strain further separates the CNT "hairs," leading to higher resistance. Since the rate of decrease/increase in resistance is slow, we expect that RF performance degradation will also be comparatively slow.

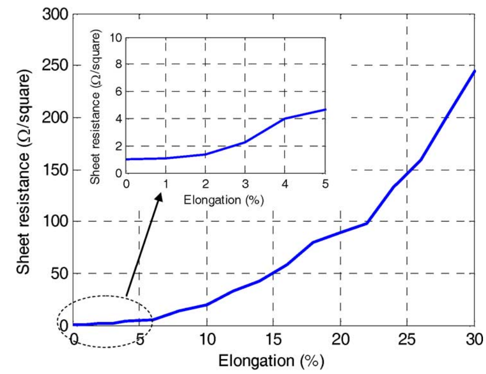
It is important to characterize the conductivity of the polymer-CNT sheet under stretching condition as well (not just bending). To do so, we employed the setup shown in Fig. 9(a) and measured the DC sheet resistance as the sample was elongated. In this setup, the degree of elongation is defined as $\Delta L/L$, where L is the original length of the sample and ΔL refers to the length after stretching. Obviously, stretching led to a decrease in the CNTs density, leading to an increase in resistance as depicted in Fig. 9(b). This behavior also agrees with percolation theory that predicts an exponential relationship between conductivity and CNT density. Nevertheless, the sample resistance was fairly stable within 2% of elongation, a typical maximum stretching for most practical applications. In the future, we expect to improve the CNT sheet conductivity under large strain and tensile stresses by sputtering metal nanoparticles on the nanotubes and by inter-dispersing horizontal CNTs into the vertically aligned CNTs.

C. Conformal CNT Patch Antennas

We examined two conformal CNT patches mounted on a cylinder surface [21]. As shown in Fig. 10, we attached a polymer-CNT patch on a metal cylinder (80 mm in diameter and 160 mm in length). Referring to Fig. 10(a), the patch is bent along the E-plane, implying that the current flows along the circumferential direction. Alternatively [see Fig. 10(b)], when

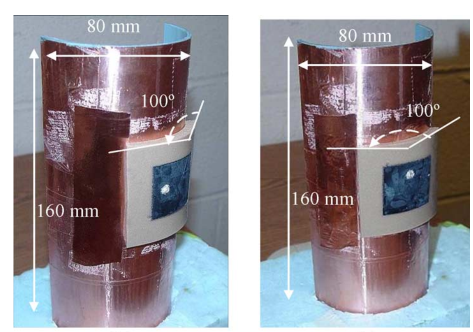


(a)



(b)

Fig. 9. DC sheet resistance versus tensile stresses. (a) Measurement setup. (b) DC sheet resistances versus tensile stresses.



(a) (b)

Fig. 10. Photograph of the cylindrically mounted polymer-CNT patch antennas. (a) E-plane bending. (b) H-plane bending.

the patch is bent in the E-plane, the current flows along the axial direction. We remark that bending led to a 13% stretching in this case. Therefore, the E-plane resonance frequency was decreased from 2.25 GHz to 1.95 GHz. This frequency shift can be justified by taking into consideration Young's modulus of the polymer substrate and the geometry of the platform.

The measured reflection coefficient and radiation patterns of the conformal CNT patches were identical to the simulated PEC patch on the same cylinder surface. However, we are most interested in the gain performance as stretching changes the CNT patch conductivity. Indeed, as shown in Fig. 11(a), the E-plane bent CNT patch had a gain of 1.7 dB at 1.95 GHz, viz. 3 dB lower than that of a simulated PEC patch in the same bent configuration. This is because the CNT surface resistivity was increased since the bending (100°) and stretching (13% elongation) reduced the nanotube density. The corresponding H-plane CNT patch had a larger measured gain of 2.9 dB at 2.25 GHz,

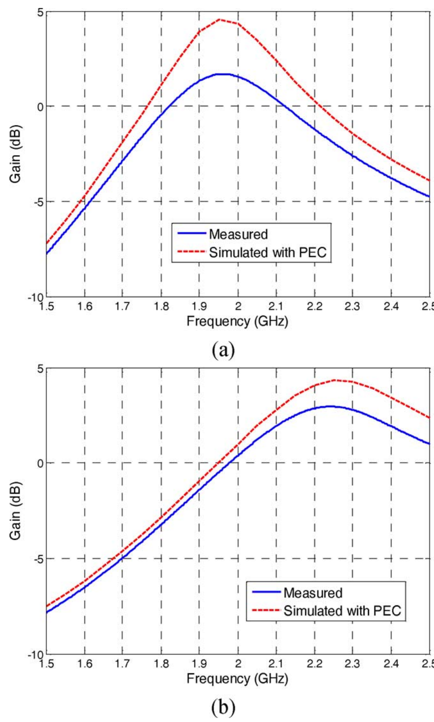


Fig. 11. Measured broadside gain of the conformal CNT patches mounted on the cylinder shown in Fig. 10. (a) E-plane bending. (b) H-plane bending.

viz. 1.5 dB lower than the simulated PEC patch on the same cylinder [Fig. 11(b)]. This larger gain is due to that the radiating currents flow vertical along the unbent direction of the CNT patch. For most practical applications, the antennas will not be subjected to such large bending or stretching. Therefore, we expect higher antenna radiation gain as the strain and bending will be smaller.

IV. Conclusion

We presented a flexible, lightweight and conductive polymer-CNT sheet for conformal load bearing antennas and RF circuits. The CNT sheet is realized by growing a high density of aligned CNTs (200 μm tall and 100 nm apart) to practically form a conducting sheet on the polymer matrix. As the CNTs are implanted within the polymer substrate, the approach leads to structural integrity under stress, strain and bending.

In this paper, we described the fabrication process and presented a circuit model to calculate the CNT sheet conductivity. The measured patch resistivity was only 0.9 Ω/square (as compared to 20 Ω/square reported with other CNT ensembles [15]. A sample polymer-CNT patch antenna was fabricated and measured to exhibit a gain of 5.6 dB, viz. only 0.8 dB less than that of a simulated ideal patch. Mechanical tests were also carried out to demonstrate the flexibility of the polymer-CNT sheets. Further, two conformal CNT patch antennas were fabricated and measured. The latter had an acceptable gain performance for practical applications, making the proposed polymer-CNT sheets suitable for conformal load bearing antennas.

Acknowledgment

Authors are very grateful to P. Ramesh of The Ohio State University Mechanical Engineering Department for his help with mechanical tests.

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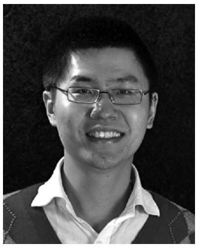

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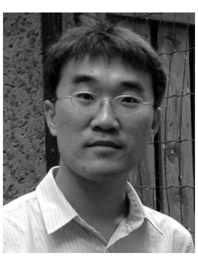

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After one year of research in PPG coating, he began Ph.D. studies on material engineering at the University of Dayton, Dayton, OH. His areas of research focus on the growth of carbon nanotube with controlled lengths onto varying kinds of substrates and he also has strong interests in instrumentation, electrospinning, membrane, electrical chemistry and carbon nanotube dry adhesive applications. In September 2009, he followed his supervisor and began performing research at Case Western Reserve University, Cleveland, OH.

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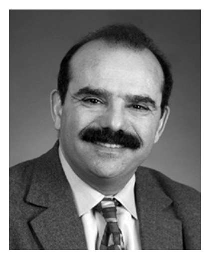

John L. Volakis (S'77–M'82–SM'89–F'96) was born on May 13, 1956 in Chios, Greece and immigrated to the U.S.A. in 1973. He received the B.E. degree (*summa cum laude*) from Youngstown State University, Youngstown, OH, in 1978, and the M.Sc. and Ph.D. degrees from the Ohio State University, Columbus, in 1979 and 1982, respectively.

He started his career at Rockwell International (1982–84), now Boeing Phantom Works. In 1984 he was appointed Assistant Professor at the University of Michigan, Ann Arbor, becoming a full Professor in 1994. He also served as the Director of the Radiation Laboratory from 1998 to 2000. Since January 2003, he is the Roy and Lois Chope Chair Professor of Engineering at the Ohio State University, Columbus, and also serves as the Director of the ElectroScience Laboratory. His primary research deals with antennas, computational methods, electromagnetic compatibility and interference, propagation, design optimization, RF materials, multi-physics engineering and bioelectromagnetics. He has published over 280 articles in major refereed journals, nearly 500 conference papers and 20 book chapters. He coauthored the following five books: *Approximate Boundary Conditions in Electromagnetics* (Inst. Elect. Eng., London, 1995), *Finite Element Method for Electromagnetics* (IEEE Press, New York, 1998), *Frequency Domain Hybrid Finite Element Methods in Electromagnetics* (Morgan & Claypool, 2006), *Computational Methods for High Frequency Electromagnetic Interference* (Verlag, 2009) and edited the *Antenna Engineering Handbook* (McGraw-Hill, 2007). He has also written several well-edited coursepacks on introductory and advanced numerical methods for electromagnetics, and has delivered short courses on antennas, numerical methods, and frequency selective surfaces.

Dr. Volakis was elected Fellow of the IEEE in 1996, and is a member of the URSI Commissions B and E. He received the University of Michigan (UM) College of Engineering Research Excellence award in 1998 and the UM, Department of Electrical Engineering and Computer Science Service Excellence Award in 2001. He is listed by ISI among the top 250 most referenced authors; He graduated/mentored nearly 60 Ph.D. students/post-docs, and coauthored with them 12 best paper awards at conferences. He was the 2004 President of the IEEE Antennas and Propagation Society and served on the AdCom of the IEEE Antennas and Propagation Society from 1995 to 1998. He also served as Associate Editor for the IEEE TRANSACTIONS ON ANTENNAS AND PROPAGATION from 1988–1992, *Radio Science* from 1994–97, and for the *IEEE Antennas and Propagation Society Magazine* (1992–2006). He currently serves as an associate editor for the *J. Electromagnetic Waves and Applications* and the *URSI Bulletin*. He chaired the 1993 IEEE Antennas and Propagation Society Symposium and Radio Science Meeting in Ann Arbor, MI., and co-chaired the same Symposium in 2003 at Columbus, Ohio.