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Skin Effect Suppression in Infrared-laser Irradiated Planar Multi-walled Carbon Nanotube/ Cu Conductors

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SKIN EFFECT SUPPRESSION IN INFRARED-LASER IRRADIATED PLANAR MULTI-WALLED CARBON NANOTUBE/CU CONDUCTORS

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Abstract

Skin effect suppression in planar multi-walled carbon nanotube (MWCNT)/Copper (Cu) conductors was realized at the 0-10 MHz frequency range through infrared laser irradiation of MWCNTs, which were coated on the surface of the Cu substrate via the electrophoretic deposition (EPD) method. The effect of laser irradiation and its power density on electrical and structural properties of the MWCNT/Cu conductors was investigated using a wavelength-tunable CO_2 laser and then comparing the performance of the samples prepared at different conditions with that of pristine Cu. The irradiation at λ=9.219 μm proved to be effective in selective delivery of energy towards depths close to the interface, compared to the conventional rapid thermal processing (RTP) annealing method. At f=10 MHz, the ac resistance of the laser irradiated MWCNT/Cu conductors was reduced by more than one order of magnitude compared to its original value for the pristine Cu. Impedance measurements and structural characterizations indicate that this technique results in successful implementation of the nanotubes on the surface of the metallic substrate to operate as current channels with saturated skin depths and reduced contact resistance at the interface. Therefore, the limited performance of Cu conductors at high frequencies can be modified. Additionally, it was further demonstrated that the impedance reduction and the suppression of the skin effect in MWCNT/Cu conductors are in direct relation with the irradiated power density and can thus be easily controlled by this parameter.

Introduction

Skin effect is known to be a major limiting factor in the performance of metallic passive components, both in terms of signal propagation and power transmittance. Particularly in the contactless power transmittance systems, the ac resistance in conventional Cu transmission lines tends to increase due to the concentration of the current close to their extremities, leading to undesired power loss and heat generation [1]. Methods such as using hollow conductors [2], Litz wires [3], and Cu/ferromagnetic composites [4-6] have been proposed to suppress the skin effect. However, their effect is limited to a short range of operational frequencies [2]. According to recent studies on the high-frequency (HF) behavior of MWCNTs, they can outperform their Cu counterparts due to their saturated skin depth, which is attributed to their long mean fee paths (momentum relaxation time) [7-9]. Thus, successful formation of MWCNT coatings on the surface of Cu conductors can provide an alternative electrical channel with frequency-independent ac resistance for carriers and, therefore, solve this long-existing limitation. Electrophoretic deposition (EPD), which is a solution-based deposition technique, is considered a cost effective and scalable approach for fabricating durable MWCNT coatings on metallic substrates with predictable structural properties, compared to other techniques such as conventional chemical vapor deposition (CVD) method [11]. However, the electrical properties of the resulting samples are always limited due to the porous structure of the MWCNT coating and its large contact resistance with the substrate [10, 12-14]. Therefore, the EPD process should be followed by a treatment technique to overcome the aforementioned limitations. It should be noted that the traditional methods, such as thermal treatment process using a furnace, may not be applicable for this purpose because they cannot realize selective delivery of heating and extreme levels of temperature to only the MWCNTs. Due to the low melting point of the Cu substrate (~1085 °C), the whole product would be damaged during the treatment [15, 16]**.** Moreover, furnace treatment is considerably time and energy-consuming and inaccurate in terms of throughput at such high temperatures. Laser irradiation, however, could be a more efficient approach for material treatment via precise, controllable, and localized energy delivery through setting the power density and wavelength of the laser in accordance with the absorption depth of the material [17]. In the case of MWCNT/Cu conductors, the low reflectance in the graphitic shells of the nanotubes can be exploited to achieve the complete optical absorption in MWCNT film, and therefore, prevent from extreme heating of the Cu substrate. So far, laser irradiation techniques have been reported to be effective in purification [18] and structural modification [19] in various types of CNT composites**.** Nevertheless, to the best of the authors' knowledge, no detailed practice for the specific effects of laser irradiation on electrical properties of MWCNT/Cu conductors and their relative ac conductivity has been reported.

In this work, a new method of fabricating stable layers of MWCNTs on Cu substrates was developed by using the EPD technique followed by an infrared laser irradiation process. This post-fabrication step proved to be effective in improving the structural and electrical properties of the MWCNT/Cu conductors. MWCNTs were investigated as frequency-independent components in order to reduce the ac resistance and, thus, mitigate the skin effect in Cu conductors. Controllable and effective attachment of the MWCNTs at the interface and their structural modification through using different laser power densities were also investigated in detail.

Experimental Section

Electrophoretic Deposition of MWCNTs

The MWCNTs (Cheap Tubes, Inc.) with diameters of 20-30 nm and lengths of 10-30 μm were used without further treatment. The required solution for EPD was prepared by dispersing 40 mg of MWCNTs into 20 ml of 1,2-dicholoroethane (DCE). The mixture was then ultrasonicated for 3 hrs in order to completely disperse the MWCNTs in the solvent and therefore break the possible entanglements and cross-linking, which are common in pristine MWCNT products. Next, 2 parallel pieces of rectangular copper conductors with similar dimensions were used as the electrodes for MWCNT deposition (Figure 1(a)). For the electrodes, the width and the thickness are 8 mm and 170 μm, respectively. The EPD process was then carried out by applying a 40 V DC voltage to the electrodes, which were kept at a distance of 5 mm for 3 min. During the process, MWCNTs were deposited onto the cathode and formed a rigid film. Afterwards, the sample (the MWCNT-Cu composite structure) was dried in a furnace at 80°C for 24 hrs under atmospheric pressure. The same EPD and drying steps were applied for the other side of the Cu substrate in order to form a MWCNT coating over the whole surface of the substrate. The concentration of the solvent should be kept constant to achieve a constant deposition yield and uniform deposition during the process [20]. In this work, multi-step EPD was employed instead of a single long step, in order to enhance the robustness of MWCNT films while maintaining a constant concentration of MWCNTs in the solvent during the process. The thickness of the resulting MWCNT coating is roughly 30 μm. Further deposition of MWCNTs reaches saturation for repetitions higher than 3 times since the random orientation of the deposited MWCNT coating and its porous structure suppresses the electrical field inside the solution and impedes further deposition [20]. Finally, the uncoated section of the Cu substrate that was outside of the solution during the EPD process was cut and separated from the main body of the samples in order to prepare them for electrical characterization. The finalized length of the MWCNT/Cu conductors was 2 cm.

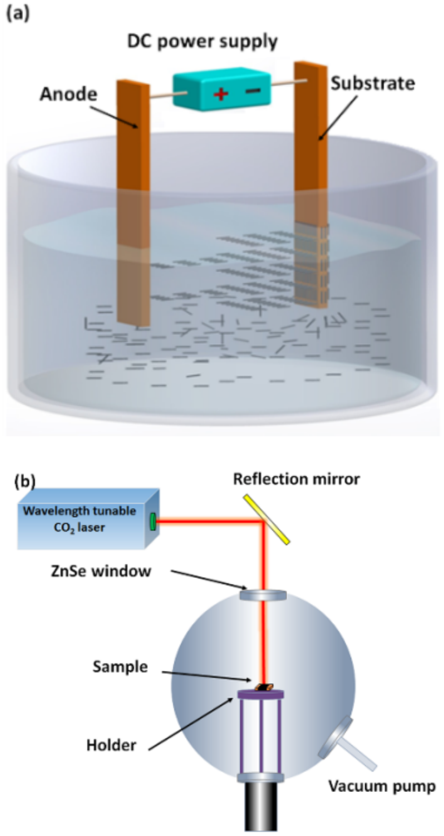

Figure 1. Schematics of the (a) EPD and (b) laser irradiation processes.

Infrared laser irradiation of the samples

Laser irradiation of the samples was then carried out using a wavelength-tunable continuous wave CO_2 laser (PRC Inc., 9.2 ~ 10.9 μm). As shown in Figure 1(b), the laser beam with a Gaussian shape was projected onto the total surface area of the samples located inside a stainless steel vacuum. The samples were irradiated at the pressure of 5×10^{-3} Torr for 3 min. The chamber pressure and irradiation time were kept the same for each experiment in order to achieve comparable results, and the laser power densities were in the range of 48.8-189.3 W/cm^2. The absorption depth of light in MWCNT films is roughly independent of the incident wavelength in mid-infrared range and has been reported previously to approximately be in the range of 20-40 μm [21]. Thus, with respect to the thickness of the fabricated MWCNT coating, a wavelength of 9.219 μm was chosen for the irradiation process in order to realize the beam absorption in depths close to the MWCNT-Cu interface. Table 1 summarizes the power densities applied for the irradiation of the samples. The laser irradiation process with power densities of higher than the one for sample E (189.3 W/cm^2) led to partial separation and rupture of the MWCNT film; therefore, comparative measurements were not possible. Thus, this value can be considered as a threshold for the resulting improvements from the laser irradiation of the MWCNT/Cu conductors.

Table 1. Laser power densities used for the irradiation of MWCNT/Cu conductors.

Sample	**Laser power density (W/cm^2)**
A	48.8
B	96.7
C	142.6
D	163.8
E	189.3

Characterizations

Surface morphologies of the MWCNT/Cu conductors were characterized by a field emission scanning electron microscope (FESEM; Hitachi S4700). Raman spectra of the samples were acquired by an inVia, Renishaw spectrometer equipped with a 50× objective. The focal spot is about 2 μm in diameter. An argon-ion laser (λ=514.5 nm) was used as the exciting source. The impedance parameters of the samples were measured using an LF impedance analyzer (HP, 4192A). The probe/sample contact resistance and the parasitic impedance were also de-embedded from results to observe the sole performance of the samples.

Results and Discussion

Figure 2(a) shows the measured ac resistance and the inductive reactance of the MWCNT/Cu conductors before and after laser irradiation. As is theoretically predicted, the skin effect causes the resistance of Cu to monotonically increase with the increase of frequency. This property, however, appears to be suppressed for all of the laser-irradiated MWCNT coated samples. The most significant reduction in the measured resistance, with respect to the one for ordinary Cu, was observed for sample E, which was irradiated at the highest power density compared to other samples. Unlike the resulting electrical behavior after the laser irradiation, the ac resistance for the sample annealed using the conventional rapid thermal processing (RTP) method at T=800 °C has increased with respect to ordinary Cu, suggesting an increase in the roughness of the metallic substrate, a well-known limitation for furnace annealing techniques [22] which leads to an increase in resistance. The overall resistance reduction rate for each sample was calculated by:

$$\Delta\% = \frac{R_{Cu} - R_{Sample}}{R_{Cu}} \times 100 \,, \qquad (1)$$

where R_{Cu} and R_{sample} are the measured ac resistance values for ordinary Cu and laser irradiated MWCNT/Cu conductors, respectively. As shown in figure 2(b), the reactance of the laser irradiated samples is roughly close to that of ordinary Cu. Since the values of this property are roughly close for the samples, the frequency range is shown in linear mode in order to have a clear illustration. The slight decrease in the reactance curves of the samples irradiated with power densities of higher than 48.8 W/cm^2 can be justified by the introduction of the relatively frequency-independent inductance in nanotubes at the interface, which is caused by their large momentum relaxation time [7, 23].

The overall behavior of the samples was further studied by analyzing the real part of the resistance ($R_{MWCNT\text{-}Cu}$) at the MWCNT-Cu interface to investigate its mechanism. Figure 4 shows the $R_{MWCNT\text{-}Cu}$ plot versus frequency for the sample E which was irradiated with the power density of 189.3 W/cm^2. The interfacial resistance follows a capacitive type of behavior before the laser irradiation process according to its decreasing magnitude. This can be attributed to the porous structure of the MWCNT layer and the considerable density of unfilled voids formed in its morphology, introducing an equivalent dielectric constant at the interface. However, after laser irradiation the interfacial resistance changes to resistive-like behavior with considerably smaller magnitude, indicating densification of the nanotubes, reduction in the amount of voids, and therefore neutralization of the dielectric behavior. Moreover, this value remains roughly constant at frequencies of lower than 3 MHz. These transitions, which were observed for all of the investigated samples, can be attributed to the successful implantation of the MWCNT layer as a less frequency-dependent current channel. The results indicate that the proposed method of laser irradiation of the MWCNTs over the Cu conductors can be applicable to realize the skin effect suppression with a more stabilized inductive behavior that is crucial for achieving the unaffected signal transmission throughout the circuit. These improvements can be easily controlled by the magnitude of laser power density. The effect of laser irradiation with power densities of higher than the one for sample E (189.3 W/cm^2) led to partial separation and rupture of the MWCNT film; therefore, comparative measurements were not possible. Thus, this value can be considered as a threshold for the resulting improvements from the laser irradiation of the MWCNT/Cu conductors.

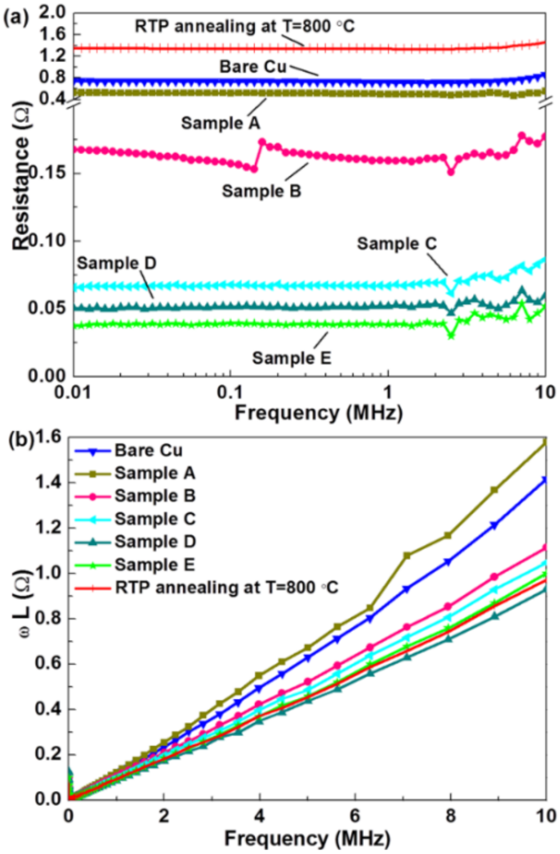

Figure 2. (a) ac resistance and (b) reactance of MWCNT/Cu conductors as function of frequency and the laser power density.

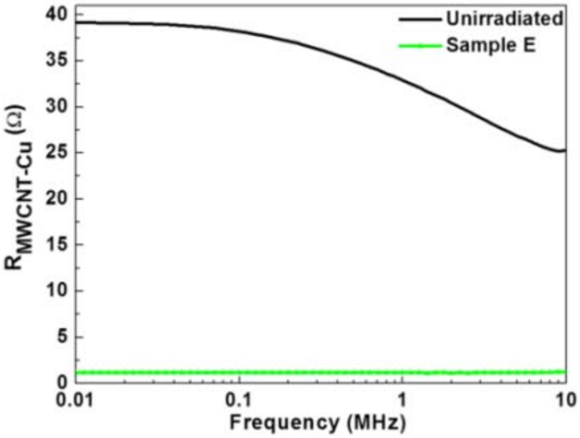

Figure 3. Real part of the MWCNT-Cu interfacial resistance as functions of the frequency before and after the laser irradiation.

The structural properties of the MWCNT films were characterized using Raman spectroscopy as shown in Figure 4. It is observed that as the assigned power density for the laser irradiation process increases, the D-band intensities of the irradiated samples decrease, whereas the G-band intensities become narrower. Since the D-band represents the defect mode in MWCNTs, this observation indicates the reduction of the defects in nanotubes. The D-band to G-band ratio (I_D/I_G) and the full width at half maximum (FWHM) of the D-band of MWCNTs are shown in Figure 4(b) as a function of laser power density. I_D/I_G reflects the ratio of structural defects and degree of disorder in the graphitic walls of nanotubes [24]. As the irradiated power density increases, the I_D/I_G ratio decreases from 1.01 to 0.86, indicating the reduction of dangling bonds and improvement of crystallinity in graphitic shells of the MWCNTs. Furthermore, the FWHM of the D-band also decreases and becomes narrower at higher power densities, and at the highest extent it falls to 82 % of its original value before the laser irradiation process, suggesting the improvement of the structural order and reduction of impurities resulting from the EPD process in MWCNT coatings. Thus, these modifications can also be considered as the results of the laser irradiation process that would further improve the electrical properties of the MWCNT channel. The crystallinity improvement and the reduction of impurities after laser irradiation are potential reasons for the skin effect suppression in the samples, which has also been theoretically expected of defect-free MWCNTs [7].

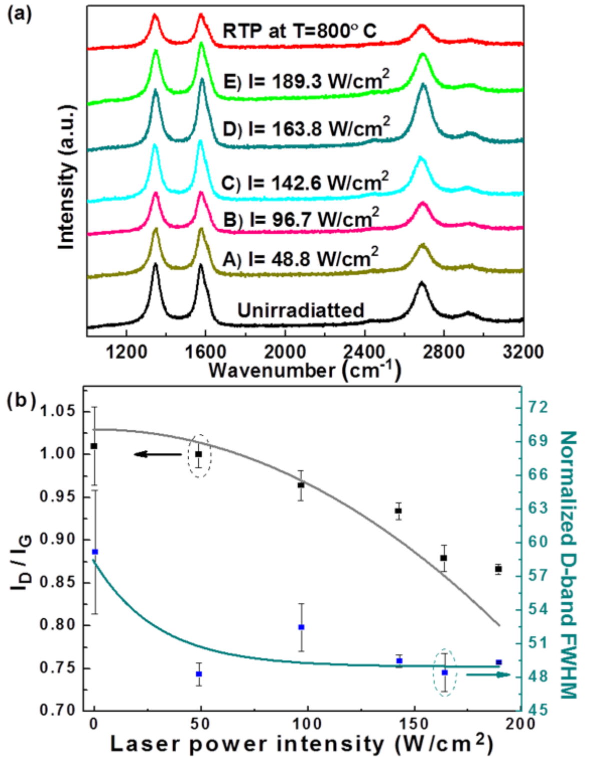

Figure 4. (a) Raman spectra of MWCNT/Cu conductors before and after laser irradiation. (b) I_D/I_G ratio and FWHM of the D-band as functions of the laser power density, along with their fitting curve (The error bars represent the standard deviation.)

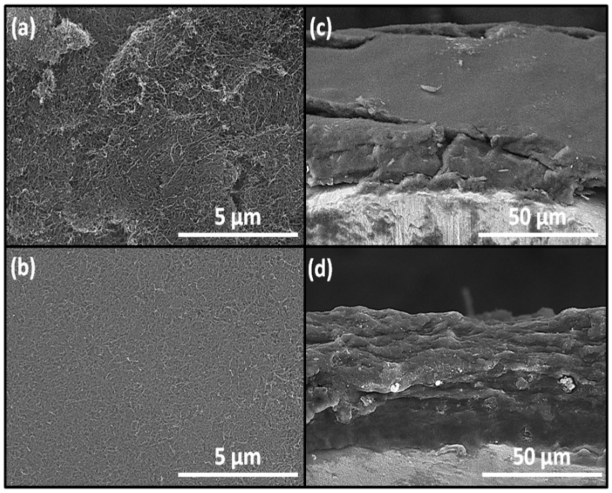

Figure 5. FESEM images of the unirradiated MWCNT coatings (a) before and (b) after laser irradiation with a power intenity of 189.3 W/cm^2. Cross-sectional images of sample E (c) before and (d) after the laser irradiation process.

The effect of the laser irradiation in structural modification of the MWCNTs, both individually and as a whole layer, can be noticed from the FESEM images. Figure 5 compares the morphology of sample E before and after irradiation. As shown in Figure 5(b), the irradiated nanotube film is more uniform and intertwined with a fewer number of unfilled voids compared to the pristine film shown in Figure 5(a). This modification in the structure of the MWCNT coating can be further supported by comparing the cross-sectional images of sample E before and after the laser irradiation process, shown in Figure 5(b) and 5(d), respectively. Moreover, the adhesion between the MWCNT layer and the Cu substrate is also improved. The highly enhanced structural properties of the irradiated sample supports the claims presented for the electrical behavior of the $R_{\mathrm{MWCNT\text{-}Cu}}$ shown in Figure 3.

Conclusions

In conclusion, the skin effect was mitigated in planar MWCNT/Cu conductors at the 0-10 MHz frequency range. The proposed infrared laser irradiation process proved to be advantageous in terms of time consumption and heat delivery selectivity compared to other conventional treatment methods such as the RTP process. At f=10 MHz, the ac resistance is reduced by 94.11 % with respect to the original Cu at power density 189.3 W/cm^2, which is attributed to the presence of the MWCNTs with negligible contact resistance as a less frequency-dependent and low-resistance current channel. The optical energy absorption of the MWCNTs during laser irradiation could explain the realization of their expected electrical behavior. Additionally, it is demonstrated that the laser irradiation process leads to improvement of the crystallinity, purification of the MWCNTs, and formation of a more uniform and interconnected layer that can also be effective in overall reduction of the ac resistance in the prepared samples.

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Biography

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