Postgrowth Microwave Treatment to Align Carbon Nanotubes

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We show that a commercial microwave oven can be used after growth to increase alignment of carbon nanotubes (CNTs) and reduce their resistance as thermal and electrical interface materials. Forests of multiwall CNTs were grown directly on both sides of aluminum foils by thermal chemical vapor deposition (CVD) and subsequently exposed to a microwave treatment in air. Scanning electron micrographs revealed enhanced vertical alignment of CNTs after postgrowth microwave treatment. The microwave treatment creates an electric field near the CNT growth substrate that aligns the CNTs orthogonally to the growth substrate. Microwaved CNT forests produced increased mechanical stiffness by approximately 58%, and reduced thermal and electrical contact resistances by 44% and 41%, respectively, compared to as-grown forests. These performance changes are attributed to an increase in the real contact area established at the CNT distal ends because of the enhanced forest alignment. This conclusion is consistent with several prior observations in the literature. This work demonstrates a facile method to enhance the alignment of CNTs grown by thermal CVD without the use of in situ plasma or electric field application. [DOI: 10.1115/1.4023162]

Keywords: carbon nanotube, thermal interface material, electrical interface material, microwave processing, metal substrate, alignment, contact area, thermal chemical vapor deposition

Introduction

Since their discovery, CNTs have garnered significant attention in a wide range of applications due to their superior mechanical, electrical, and thermal properties [1]. It has been reported that CNTs can behave as ballistic electrical conductors and maintain high current densities [2], as well as exhibit high intrinsic thermal conductivities comparable to that of diamond [3,4]. Forests of CNTs have been shown to exhibit excellent mechanical resilience and conformability to surfaces [5–8]. The combination of high mechanical conformability and thermal and/or electrical conductance is a key feature desired for high-performance thermal and electrical interface materials [9–11]. As a result, CNT forests are an ideal choice for such materials and recent efforts have utilized CNT forests to develop novel devices applied at interfaces to improve energy transport [12–15].

Vertical alignment is a key feature for achieving high conductance in CNT forest interface materials [12–14]. Thermal interface materials (TIMs) made from vertically aligned CNT forests in dry contact have produced resistances that range from approximately 8–50 mm^2 K/W [12,13,16–22]. These resistance values are about an order of magnitude lower than the resistances produced by dry CNT TIMs with random orientation of their constituent tubes [23,24]. This is because thermal transport is impeded by increased phonon scattering at CNT–CNT contacts within randomly oriented CNT bundles [25,26]. Enhanced electrical contact conductance has also been achieved using CNT forests [15]. The electrical resistance of two copper plates in contact was measured to decrease by 80% with the addition of a vertical CNT forest between the plates [15]. The enhancements to thermal and electrical contact conductances achieved by using vertical CNT forests have been attributed to the combination of high intrinsic thermal and electrical conductivities of the forests, and the ability of the forest to establish large amounts of true surface contact [12]. Vertical alignment exposes more CNT ends to the contact surface than that which occurs with randomly oriented CNT masses. This enhanced exposure of CNT distal ends is suggested to facilitate enhanced contact between the surface and individual tubes in the forests, which increases the number of parallel paths for energy transport [12].

Some degree of vertical alignment is clearly advantageous for CNT forests used as high-conductance interface materials as concluded in the several studies discussed above. Vertical alignment of CNT forests is usually achieved by crowding that occurs naturally during the growth of dense CNT forests [27], the use of a plasma during growth to selectively etch misaligned tubes [28], or the application of an electric field, with and without plasma exposure, with the electric field lines oriented parallel to the growth substrate [29–32]. The use of a plasma or an applied electric field during growth usually produces forests with straight and aligned CNT tips [29–32]. Forests that align by crowding effects alone tend to exhibit a greater degree of random orientation and entanglement in the tip region as compared to other areas of the forest [33]—note that the entangled region could comprise a substantial portion of the entire height of relatively short forests. The entangled "canopy" of a CNT forest can be removed with additional processing after growth, e.g., plasma etching, to create more alignment [33]. However, achieving a high degree of uniform alignment in CNT forests—especially at the distal ends—without the use of plasma or applied bias during growth, or some sort of postprocessing remains a significant challenge.

Metal foils have been used recently as attractive candidate substrates for CNT growth at increased manufacturing scale [34]. Studies have shown that CNT forests grown on both sides of metal foils can produce thermal interface resistances that range from 10 to 90 mm^2 K/W depending on the magnitude of interface roughness and pressure [19,35]. The lower end of this performance range compares favorably with state-of-the-art commercial TIMs [9]. Aluminum foil is an especially attractive metal foil to use for CNT growth and CNT-based TIM applications because of its relatively low cost. However, the reduced growth temperatures required for CNT processing on aluminum foil often result in relatively short and randomly oriented CNTs [36]. Use of in situ

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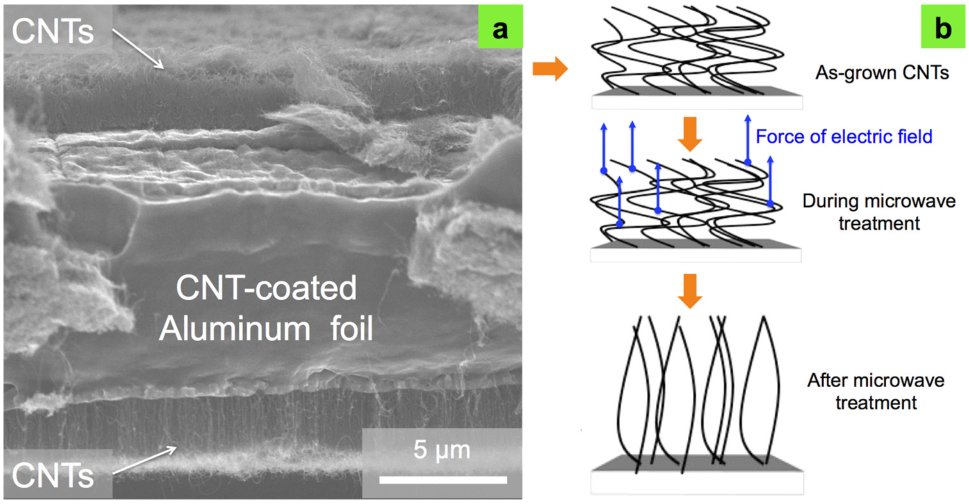

Fig. 1 (*a*) **Scanning electron micrograph of CNT arrays grown on both sides of aluminum foil before microwave treatment. (*b*) Illustration of CNT alignment on one side of the aluminum foil substrates as a result of the electric field applied to the sample during microwave treatment. Electric field forces perpendicular to the substrate pull individual CNTs into alignment.**

plasma or application of electric fields to address this alignment problem can increase manufacturing complexity and costs. Microwave heating is a common process used for drying at large scales in industry [37]. Postgrowth microwave irradiation has been used to improve the quality of CNTs [38] and improve their adhesion to the growth substrate [39]. However, there have been no reports of significantly enhanced vertical alignment in CNT forests treated in a microwave oven after growth.

Here, we introduce a fast and relatively nondestructive microwave treatment after the growth of CNTs on metal substrates to increase vertical alignment of the CNTs (Fig. 1) and reduce their thermal and electrical resistances as interface materials. This technique provides a mean to overcome the relatively poor alignment that often results from thermal CVD growth of CNTs at reduced temperatures on metals such as aluminum and copper [36]. CNTs were observed in a scanning electron microscope (SEM) to become more straight and aligned after microwave irradiation, especially near their distal ends. This change in morphology as a result of the microwave treatment increased the stiffness of CNT arrays synthesized directly on both sides of aluminum foil by approximately 58% and decreased the thermal and electrical resistances of the same CNT-coated foil structure by 44% and 41%, respectively. The double-sided CNT foil structure after microwave posttreatment produced a low-end thermal resistance value of 26 mm^2 K/W and a low-end electrical resistance value of 4.1 Ω for a contact area of 25 mm^2.

Experimental Methods

Sample Fabrication. For CNT fabrication, 99% pure, 10 μm thick aluminum foil (Alfa Aesar, USA) was used as substrate. A catalyst stack of 10 nm Fe/10 nm Al/30 nm Ti/100 nm Ni was deposited on both sides of the aluminum substrates by electron-beam evaporation. The foils were then placed in a thermal CVD furnace (Aixtron Black Magic) for CNT synthesis. Growth was performed at a chamber temperature of 700 °C for 10 min with constant 700 sccm and 100 sccm flow rates for the acetylene and hydrogen gases, respectively. An infrared sensor measured the temperature of the aluminum substrates as 630 °C during growth. CNTs were grown on both sides of the aluminum substrates and the CNT heights were approximately 3–4 μm on the top side and 2–3 μm on the bottom side for each sample (Fig. 1(*a*)). The synthesized CNTs were placed in the center of a commercially available microwave oven (Panasonic NNS-575) set at 2.45 GHz and 1300 W (the maximum power) for 5 min in air to perform the microwave treatment. The samples rotated on the microwave oven stage during the 5 min of treatment. Samples were left to cool immediately following the treatment for approximately 5 min.

Sample Characterization. The CNT mass density was found by weighing the samples before and after CNT growth using a microbalance and calculating the volume of the forests using the measured heights and areas of CNT coverage. The measured CNT mass density of a representative sample was 0.264 ± 0.001 g/cm^3. The mass densities of all tested samples were within ±0.01 g/cm^3 of this value. No change in CNT mass was detectable after microwave treatment, although the mass density decreased because the height of the CNT forests increased after microwave alignment. The CNT morphologies and structure were examined by scanning electron microscopy (SEM, Hitachi 4700) and transmission electron microscopy (TEM, JEOL 4000EX). Raman spectroscopy (Nicolet Almega) was also conducted using a 488 nm laser with a 3 s integration time for 30 accumulations.

Mechanical Testing. Mechanical properties of the CNT forests before and after microwave treatment were characterized using nanoindentation. Nanoindentation was performed in a Hysitron triboindenter using a diamond Berkovich tip. The samples were prepared by bonding one side of the double-sided CNT-coated aluminum foils to rigid glass substrates using superglue and allowing the superglue to dry for at least 24 h. Three to four separately superglued samples of CNT forests of each type were tested to ensure the results were not sensitive to the thickness of the superglue, which might vary between samples. The maximum indentation depth was in the range of 1000–1350 nm for each sample to minimize the change of stiffness due to variation in the maximum depth. The slope of the initial unloading in the load–displacement curves, which are measures of the elastic stiffness of the material [40,41], was averaged over 17 measurements for both microwaved and as-grown CNT samples.

Thermal Testing. Thermal characterization was conducted by a photoacoustic (PA) technique at room temperature. The PA technique has been demonstrated as an effective method to characterize the thermal properties of CNT arrays [18]. It is a nondestructive technique capable of resolving individual component resistances in a multilayer sample. Extended details on the PA characterization system used here can be found in Refs. [18] and [42]. The PA cell is pressurized to apply 135 kPa to the double-sided CNT-coated foil samples, which are placed in dry contact with silver foil on top and a thick quartz backing on the bottom. The corrected phase shift of the PA signal is compared to the output of a heat transfer model and the parameters are adjusted to minimize the difference between the theoretical and experimental data using a nonlinear fitting method. Multiple measurements are performed on each sample to determine a range of probable values for each unknown property. In this case, the unknown properties are: the contact resistance between the CNTs and the silver foil, the layer resistance of the CNT-coated foil, the contact resistance between the CNTs and the quartz, and the thermal diffusivity of the CNT-coated foil. The thermal diffusivity of the CNT-coated foil is allowed to vary to achieve the best match between the theoretical and experimental phase shift. Only the total thermal resistance values are reported in this work, which is the combination of the two contact resistances and the layer resistance, because the uncertainty in fitting for the component resistances was too high to discern meaningful conclusions.

Electrical Testing. Electrical characterization was performed on a probe station using an Agilent E5272A source/monitor unit. Copper metal blocks of 5 × 5 mm were used to make contacts on both sides of double-sided CNT-coated foil samples in order to distribute the pressure of the probes homogeneously and to avoid any possible punch-through while making electrical contact with the CNTs. The force applied to all tested samples was held constant by fixing the sample heights and the number of rotations on a worm screw used to control contact between the probes and the sample.

Results and Discussion

The CNTs grown on metal foil in this study were relatively short (2–4 μm) and possessed a low degree of alignment (Fig. 2(*a*)). The poor alignment is attributed to the short forests heights, such that the entangled canopy layer [33] comprises the majority of the forest height. Over 20 as-grown CNT forests with the characteristics shown in Fig. 2(*a*) were treated with microwave processing after growth. Treating the CNTs on metal foil in a microwave after growth consistently resulted in a significant enhancement in vertical alignment (Fig. 2(*b*)). We note that a change in vertical alignment was difficult to observe in SEM when the microwave treatment was applied to highly dense CNT forests with heights greater than about 10 μm because the degree of vertical alignment via the crowding effect [27] was relatively high in these as-grown samples. However, similar reductions in thermal interface resistance were measured for short and tall samples after microwave treatment, which suggest that enhanced alignment occurred in taller CNT forests as well. The correlation between enhanced vertical alignment and reduced thermal interface resistance is discussed further below.

The mechanism of enhanced vertical alignment is ostensibly electric field-induced alignment of the multiwall CNTs, which respond as a metal in the electric field generated by the microwave treatment. The electric field is generated near the growth substrate, parallel to the substrate surface. This field is generated because the microwave irradiation interacts with electrons in the surface of the metal foil and causes the electrons to flow in the plane of the foil surface. The strength of the field attenuates away from its source (the metal surface) into the CNT forest. Dipole moments are generated along the axial direction of individual CNTs in the presence of this electric field because of charge accumulation in the cross-nanotube direction [43,44]. These dipoles generate forces that align CNTs perpendicularly to the substrate surface in the presence of the electric field (Fig. 1(*b*)). This field-induced alignment mechanism is similar to those that result in alignment of CNTs grown under the influence of an applied voltage bias [29–32]. Microwave treatment produces a heating effect, i.e., Joule heating [45], in addition to establishing an electric field that aligns the CNTs. An increase in atomic vibrational energy as a result of heating could assist breaking of van der Waals bonds between neighboring CNTs, which might facilitate CNT motion

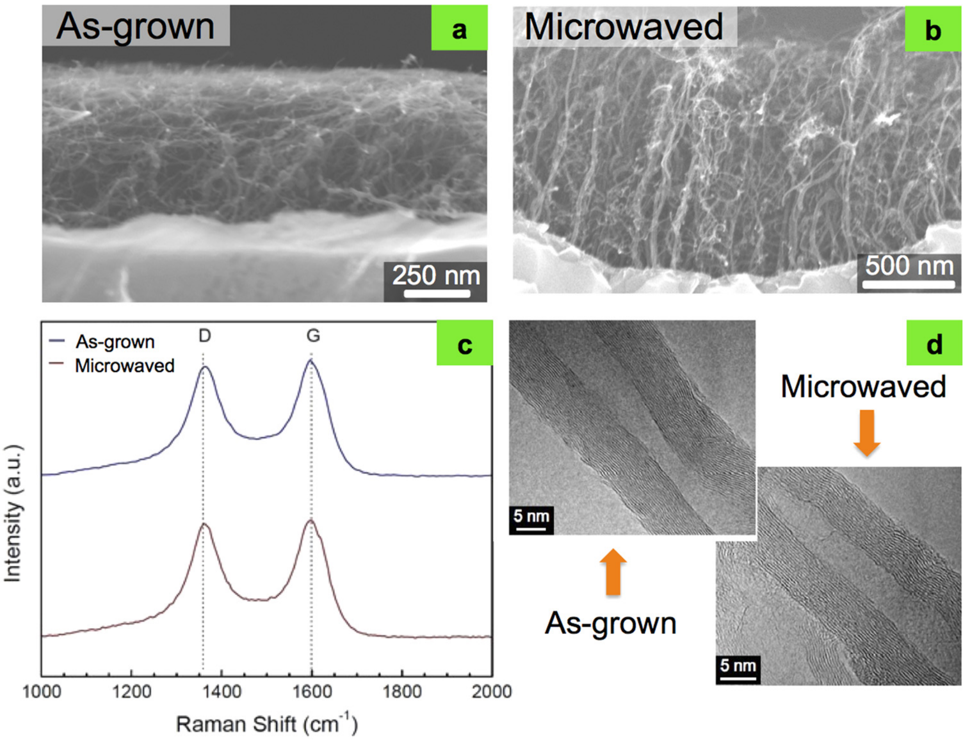

Fig. 2 Morphological characterization of double-sided, CNT-coated foil structures. (*a*) Scanning electron micrograph of CNTs on aluminum foil substrate before microwave treatment. (*b*) Scanning electron micrograph of the CNT sample in frame (*a*) after microwave treatment. (*c*) Raman spectra of CNTs before and after microwave treatment. (*d*) Transmission electron micrographs of a representative CNT before and after microwave treatment.

towards enhanced vertical alignment. However, such possible effects as a result of heating require further study, especially since the substrate temperature during microwave treatment could not be measured in this study.

It is important to note that severe electrical discharges (i.e., sparks) were observed when the postgrowth microwave treatment was applied to CNT forests grown on silicon and silicon dioxide substrates. These discharges were due to the accumulation of charge on the surface of the silicon and silicon dioxide substrates as reported previously [38]. Therefore, the applicability of the postgrowth microwave alignment technique is limited. CNT forests on aluminum substrates were found to be ideal for microwave treatment post growth; although some light sparking occurred occasionally at the corners of the aluminum foil substrates. Small sections of the corner of the foils would burn when this sparking occurred, but the bulk of the samples remained undamaged. Generally, CNTs on metal substrates are expected to work well with the microwave processing. We propose that the high electrical conductivity of aluminum (and metals generally) facilitates the dissolution of charge buildup on the substrate surface. In addition, the CNTs promote effective heat transfer away from the aluminum, which reduces substrate heating and might also reduce the accumulation of surface charge.

Raman spectroscopy was conducted before and after microwave treatment to determine if any significant structural changes to the CNTs resulted from microwave irradiation (Fig. 2(*c*)). All CNT forests displayed characteristic peaks at 1350 cm^{-1} and 1580 cm^{-1} known as the D and G bands, respectively, which provide a qualitative assessment of the inherent "diamond" and "graphene" like bonding present in the samples. For CNT forests, a D/G band peak greater than 1 would indicate a higher density of defects in the CNTs than if the D/G band peak were less than 1. No detectable differences in peak location or peak intensity were observed in the Raman spectra of as-grown and microwaved CNT forests (Fig. 2(*c*)). A slight increase in the defect density of microwave-treated CNTs was reported in a prior study [39]; however, the as-grown CNTs in this prior work were more defective than the CNTs here, which would make them more prone to chemical modification from microwave irradiation in air.

Several studies [38,46–48] have demonstrated microwaves as a purification technique for ridding CNTs of defects and contaminants such as moisture and oxygen [45]. The relatively short microwave exposure times used here could explain why we did not observe such changes to CNT quality in this study. Furthermore, the air atmosphere would provide a source of oxygen, and possibly moisture, which would replenish any oxygen removed by microwave irradiation. Heating in an oxidizing environment can burn away CNT outer walls [49]. Therefore, TEM was utilized to further examine the impact of the microwaves on individual CNTs (Fig. 2(*d*)). Under our microwave parameters, a decrease in the number of walls from 24 to 17 was observed, which corresponds to a reduction in outer CNT diameter from 30 to 22 nm. Further study is required to elucidate any effect reduced CNT diameter might have on the process of postgrowth microwave alignment.

The alignment of CNT forests, especially near the free tips of CNTs has been shown to affect the stiffness response of the forests in nanoindentation testing [50]. This is because increased tip alignment creates more contact area between the CNT forest and the head of the indenter. The stiffness responses of the microwaved and as-grown samples were evaluated by nanoindentation testing. The stiffness of as-grown samples was $12.0 \pm 2.8\ \mu\mathrm{N/nm}$, and the stiffness of microwaved samples was $28.6 \pm 5.6\ \mu\mathrm{N/nm}$, which is more than twice the stiffness of as-grown samples (Fig. 3(*a*)). This result indicates that microwaved CNT forests produced more true contact area than as-grown forests when interfaced with the indenter head.

The average measured thermal resistances of double-sided CNT foil TIMs before and after the microwave treatment were 46 mm^2 K/W and 26 mm^2 K/W, respectively, indicating a 44% decrease in resistance (Fig. 3(*b*)). As mentioned previously, these samples were measured with an applied pressure of 135 kPa. The significant reduction in thermal resistance is attributed to the improvement in vertical alignment from the microwave treatment because vertical alignment brings more individual CNTs into contact with the interface surfaces [12]. So better vertical alignment is shown here to correlate with increased stiffness and interfacial contact area, and increased thermal conductance by providing more CNT pathways for thermal transport.

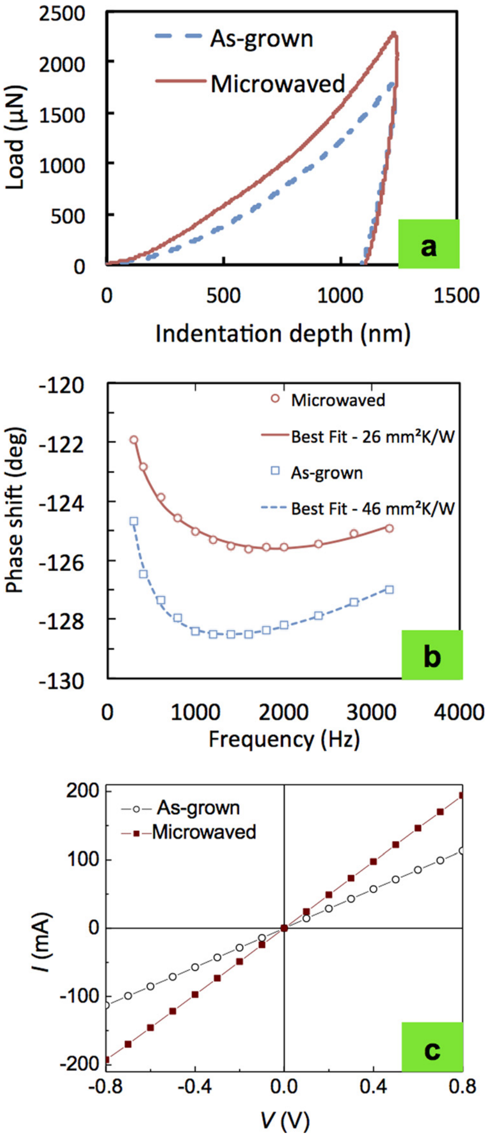

Fig. 3 Mechanical, thermal, and electrical testing of double-sided, CNT-coated foil structures. (*a*) Load–displacement curve for CNT samples before and after microwave treatment. (*b*) Measured phase shift in PA measurement of CNT samples before and after microwave treatment. (*c*) Current–voltage characteristics of CNT samples before and after microwave treatment.

The current–voltage (*I*–*V*) characteristics of as-grown and microwaved double-sided CNT foil structures are shown in Fig. 3(*c*). It can be seen clearly that the microwave treatment of double-sided CNT foil structures exhibits less resistance than that of as-grown structures. The extracted values of the resistance are 4.1 Ω for microwave-treated structures, and 7.0 Ω for as-grown structures. These results indicate that the electrical contact resistance of as-grown structures is reduced by 41% after the microwave treatment. Therefore, the microwave treatment can be used effectively to reduce electrical contact resistance (in addition to thermal resistance), and appears to be a useful technique to be exploited for CNT based interface materials grown on metal substrates.

Conclusions

Forests of CNTs were fabricated on both sides of aluminum foil substrates using thermal CVD. A rapid microwave technique that resulted in a significant increase in vertical alignment of CNTs was applied after growth with little change to the defect density of the CNTs. Vertically oriented forces, caused by dipole moments in the CNTs that were generated by the microwave-induced electric field near the substrate, were the ostensible causes of this enhanced alignment. Such field-induced alignment of CNTs has been a frequent observation in the literature. The enhanced vertical alignment resulted in a greater "free-tip" contact area, and enhanced thermal (by 44%) and electrical (by 41%) contact conductances in CNT-foil interface materials. The enhanced contact at the distal ends of CNTs was further confirmed by a 58% increase in the stiffness response of microwaved CNT forests compared to as-grown forests. The simple microwave technique presented here provides a rapid and scalable postgrowth process that can tune and enhance the effectiveness of CNT-based interface materials.

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