

HT2007-32815

## ALUMINUM FOIL/CARBON NANOTUBE THERMAL INTERFACE MATERIALS

Baratunde A. Cola, Xianfan Xu, Timothy S. Fisher

*Purdue University School of Mechanical Engineering, West Lafayette, IN 47907  
and Birck Nanotechnology Center, West Lafayette, IN 47907*

### ABSTRACT

Vertically oriented carbon nanotube (CNT) arrays have been simultaneously synthesized at relatively low growth temperatures (i.e.,  $< 700^{\circ}\text{C}$ ) on both sides of aluminum foil via plasma enhanced chemical vapor deposition. The resulting CNT arrays were highly dense, and the average CNT diameter in the arrays was approximately 10 nm, much smaller than in previous work. Also, the CNT arrays were smaller in height than the arrays in previous work. At moderate pressures, the aluminum foil/CNT material achieves resistances as low as  $10 \text{ mm}^2\text{-K/W}$  for relatively smooth and flat interfaces, similar to previous work. However, the aluminum foil/CNT material performs relatively poor for less ideal, rougher interfaces presumably due to the small height and very close packing of CNTs that decreases the materials ability to fill interfacial voids and conform to the geometry of the mating surfaces. It is also possible that the aluminum foil was slightly stiffened during CNT growth (through hydrogen embrittlement), which could further reduce the conformability of the aluminum foil/CNT material.

### NOMENCLATURE

$R$	thermal resistance, $\text{mm}^2\text{-K/W}$
<u>Subscripts</u>	
Al – CNT (CNT – Al)	aluminum/CNT array interface
CNT	effective CNT array property
free CNT tips – solid two	free CNT tips/solid two interface
solid one – free CNT tips	free CNT tips/solid one interface
total	complete aluminum/CNT interface

### INTRODUCTION

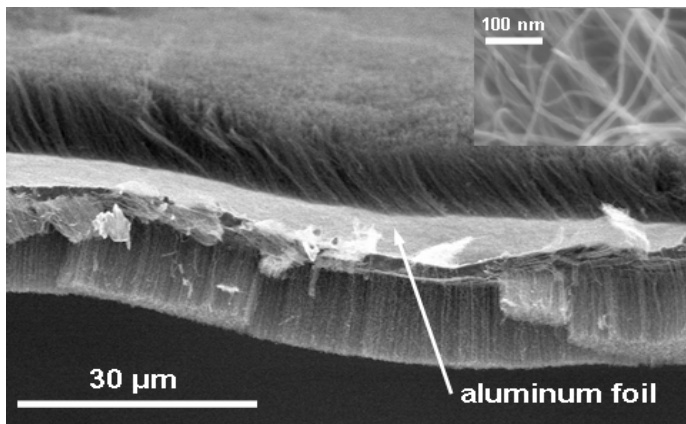
The improvement of thermal interface conductance through the use of directly synthesized carbon nanotube (CNT) arrays has garnered much recent attention [1,2,3,4,5]. Carbon nanotubes are attractive because they can be made to form a dry, removable interface with good thermal conductance. However, direct synthesis of CNT array interfaces can be difficult to implement because of the high temperatures and substrate-preparation steps (i.e., catalyst methods) that are normally required for CNT growth. In prior work, an insertable and conformable copper foil/CNT thermal interface material (TIM) was suggested as a way to overcome such implementation issues, and was measured to achieve low resistances for relatively rough and smooth interfaces [5].

Enabled by recent advancements in low temperature synthesis [6], a CNT TIM that consist of CNT arrays directly and simultaneously synthesized on both sides of aluminum foil has been fabricated. Similar to prior work on CNT-coated copper foil [5], the TIM is insertable and allows temperature sensitive and/or rough substrates to be interfaced by highly conductive and conformable CNT arrays. However, the use of aluminum foil is economical and may prove favorable in manufacturing due to its wide use.

### SYNTHESIS

Plasma-enhanced chemical vapor deposition (PECVD) [7] was used to synthesize the CNT arrays in this study. A tri-layer catalyst [1] configuration (30 nm-Ti/10 nm-Al/2 nm-Fe) was deposited on both sides of 2  $\mu\text{m}$ -thick aluminum foil. The active Fe catalyst layer was only 2 nm to allow the growth of small diameter multiwalled CNTs. Due to the relatively low melting temperature of aluminum ( $\sim 660^{\circ}\text{C}$ ) and to allow the process gases to reach both of its surfaces, the foil was elevated

by ceramic spacers, 1.2 mm in height, on a growth stage set at 650°C. The PECVD process gases were H<sub>2</sub> (40 sccm) and CH<sub>4</sub> (10 sccm), which is more carbon rich than previous synthesis conditions [5] to promote dense low-temperature growth, and the pressure was 10 Torr. A 100 W plasma was formed in the growth chamber and concentrated on a molybdenum shield placed above the aluminum foil, and synthesis was carried out for 10 min. Shielding was necessary during growth to prevent excess heating and foil damage (hardening) due to direct plasma exposure. The temperature on the top of the molybdenum shield was measured with a pyrometer to be 655°C. When the plasma shield was not used and/or when the growth temperature was higher, visible foil damage was noticed and the aluminum foil/CNT material became very stiff and brittle (mostly likely due to exacerbated hydrogen embrittlement of the aluminum). A scanning electron microscope (SEM) is used to image the CNT arrays on the aluminum foil as illustrated in Figure 1. Each array is approximately 10 μm tall, and the average CNT diameter is approximately 10 nm. The CNT density of each array is ~ 10<sup>9</sup> CNTs/mm<sup>2</sup>.

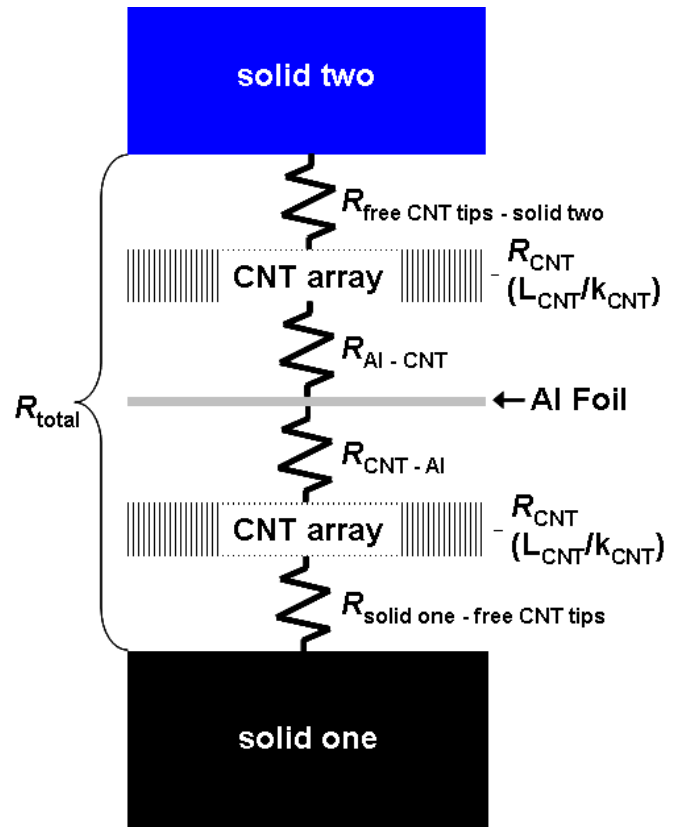


**Figure 1. CNT arrays synthesized on both sides of aluminum foil. The insert is a higher magnification SEM image that illustrates the CNT diameters in the array.**

## RESULTS

A resistive network for the aluminum foil/CNT interface is illustrated in Figure 2. The room-temperature thermal resistance of the complete interface,  $R_{total}$ , was measured for smooth and flat mating solids (silver foil and polished silicon) using a photoacoustic (PA) technique [4]. The PA technique involves periodically heating the sample surface, which is surrounded by a sealed acoustic chamber. The temperature-induced pressure response in the acoustic chamber is measured and used in conjunction with the model of Reference 4 to determine thermal properties. The transient nature of the PA technique allows for precise measurement of the thermal resistance of the aluminum foil/CNT interface (error ~ 1 mm<sup>2</sup>-K/W), and a resistance value of approximately 10

mm<sup>2</sup>-K/W was achieved at an interface pressure of 345 kPa, which compares favorably with results obtained for a copper foil/CNT thermal interface material [5].



**Figure 2. Resistive network for the aluminum foil/CNT interface.**

The aluminum foil/CNT material was also tested in a less ideal interface (i.e., rougher, wavier, and less flat) and over a larger area (1 x 1 cm) using a one-dimensional reference bar technique [1], and a resistance value of approximately 90 mm<sup>2</sup>-K/W was measured at 345 kPa. We attribute the relatively poor performance in the less ideal, rougher interface to three characteristics of the aluminum foil/CNT material fabricated in this study that prevent a significant increase in real contact area; small CNT array heights, a very large density of small diameter CNTs in the arrays, and possible stiffening of the aluminum foil during CNT growth. The small heights hamper the ability of the CNT arrays to completely fill the interfacial voids, especially in highly rough and/or warped areas of contact. For closely packed, small diameter CNTs, tube-to-tube van der Waals interactions are strong such the CNTs aid each other in reaming ridged, which causes the CNT array to be relatively stiff and to perform like a macro material. Stiffening of the aluminum foil during CNT growth would further impede the aluminum foil/CNT materials ability to conform in the interface. These effects are less significant in the smooth interface because only modest (in comparison) aluminum

foil/CNT material deformation is necessary to enhance real contact area. After both tests, the interfaces were separated, and the CNT arrays remained firmly attached to the aluminum foil, indicating the good adhesion provided by the reported growth technique.

## CONCLUSIONS

The room-temperature thermal resistance of an aluminum foil/CNT interface material was measured to achieve a low resistance of approximately 10 mm<sup>2</sup>·K/W for smooth and flat interface, which compares very favorably with previous results [5]. The conductance enhancement was diminished for the same aluminum foil/CNT material in a less ideal interface due primarily to small CNT array heights that prevent complete spanning of the interface gaps and highly dense small diameter CNTs that through van der Waals interactions create ridged CNT arrays. It is also possible that the aluminum foil is stiffened from exposure to the CNT growth environment (in particular, hydrogen at high temperatures); thus, synthesis should be extended to even lower temperatures and plasmas formed from other gases should be explored (e.g., argon). The performance of the aluminum foil/CNT material in a given interface is a function of several parameters (e.g., CNT density, length, and diameter, and foil thickness) that require further study in order to identify the ones that are most readily adjusted to maximize thermal conduction in a particular interface configuration.

## ACKNOWLEDGMENTS

We gratefully acknowledge the financial support of the Air Force Research Laboratory. We thank Sriharsha Aradhya for reference bar measurements. The lead author is grateful for personal support from the Intel and Purdue University.

## REFERENCES

- [1] Xu, J., and Fisher, T. S., 2006, "Enhanced Thermal Contact Conductance using Carbon Nanotube Array Interfaces," *IEEE Trans. Comp. Pack. Tech.* 29(2), pp. 261-267.
- [2] Xu, J., and Fisher, T. S., 2006, "Enhancement of Thermal Interface Materials with Carbon Nanotube Arrays," *Int. J. Heat Mass Trans.* 49, pp. 1658 -1666.
- [3] Tong, T., Zhao, Y., Delzeit, L., Kashani, A., Majumdar, A., and Meyyappan, M., 2005, "Vertically Aligned Multi-walled Carbon Nanotube Arrays as Thermal Interface Materials and Measurement Technique," Proc. ASME International Mechanical Engineering Congress and Exposition, Orlando, Florida, IMECE2005-81926, pp. 1-7.
- [4] Cola, B. A., Xu, J., Cheng, C., Hu, H., Xu, X., and Fisher, T. S., 2007, "Photoacoustic Characterization of Carbon Nanotube Array Interfaces," *J. Appl. Phys.* 101, 054313.
- [5] Cola, B. A., Xu, X., and Fisher, T. S., 2007, "Increased Real Contact in Thermal Interfaces: A Carbon Nanotube/foil Material," *Appl. Phys. Lett.* 90, 093513.
- [6] Amama, P. B., Ogebule, O., Maschmann, M. R., Sands, T. D., Fisher, T. S., 2006, "Dendrimer-assisted Low-temperature Growth of Carbon Nanotubes by Plasma-enhanced Chemical Vapor Deposition," *Chem. Commun.*, pp. 2899-2901.
- [7] Maschmann, M. R., Amama, P. B., Goyal, A., Iqbal, Z., Gat, R., and Fisher, T. S., 2006, "Parametric Study of Synthesis Conditions in Plasma-enhanced CVD of High-quality Single-walled Carbon Nanotubes," *Carbon* 44, pp. 10-18.