## Increased real contact in thermal interfaces: A carbon nanotube/foil material

Baratunde A. Cola, Xianfan Xu, and Timothy S. Fisher<sup>a)</sup>

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

(Received 18 December 2006; accepted 16 January 2007; published online 2 March 2007)

The thermal performance of an interface material comprised of a metal foil with dense, vertically oriented carbon nanotube (CNT) arrays synthesized on both of its surfaces is characterized for rough and smooth interfaces. The CNT/foil deforms in the interfaces by two mechanisms, CNT deformation and foil deformation, that may significantly increase the number of CNT contact spots on both sides of the foil. As a result, thermal interface resistances less than 10 mm<sup>2</sup> K/W are achieved at moderate pressures and compare very favorably to alternative interface materials and structures. © 2007 American Institute of Physics. [DOI: 10.1063/1.2644018]

Carbon nanotubes (CNTs) have received much attention for their ability to enhance thermal interface conductance.<sup>1–5</sup> Several recent studies have focused on CNT array interfaces directly synthesized on substrates.<sup>2-5</sup> An interface with a CNT array directly synthesized on one side of the interface has been reported to have a resistance as low as 16 mm<sup>2</sup> K/W under moderate pressure, which is approximately an order of magnitude less than that of a bare Si-Cu interface.<sup>2,5</sup> An interface with CNT arrays directly synthesized on both sides has been reported to have a resistance of 4 mm<sup>2</sup> K/W, which is similar to that of a soldered joint.<sup>5</sup> However, in applications where the materials that form the interface cannot be exposed to the temperatures normally required for CNT growth,<sup>6</sup> direct synthesis of CNT array interfaces is challenging, although some progress in lowtemperature synthesis has been reported.<sup>7</sup> In addition, when interface surfaces are relatively rough (e.g., unpolished Cu-Cu interfaces), direct synthesis of CNT arrays that are dense and long enough to fill the interface voids effectively can be difficult. Direct synthesis of CNT array interfaces also faces scalability challenges, given that the synthesis would need to be customized for the geometry and materials used in a particular thermal management scheme.

A CNT thermal interface material (TIM) that consists of CNT arrays directly and simultaneously synthesized on both sides of a metal foil has been fabricated. The TIM eliminates the need for exposing temperature-sensitive materials and devices to normal CNT growth conditions and provides greater conformability to rough interfaces due to foil deformation that increases the number density of contact points between free CNT tips and their opposing substrate, and this part of the interface has been shown in prior work to be the most resistive component in the heat flow path.<sup>4,5</sup> The CNT/ foil TIM is similar to existing state-of-the-art TIMs in that it can be inserted into several different interface configurations; however, it differs in that it is dry, removable, and has an intrinsically high thermal conductivity.

Many parameters affect the performance of metal foils as thermal interface materials.<sup>8</sup> Qualitatively, the thermal resistance of a metal foil interface depends on the thermal and physical properties of the contacting members, foil, and gas gap, the contact geometry, the contact pressure, and the interface temperature. While the foregoing functional dependencies are difficult to resolve analytically, empirical correlations have been developed that match experimental results reasonably well.<sup>8</sup> Experimental observations have revealed the existence of an optimal thickness, for which thermal resistance is a minimum independent of contact pressure, for metal foils used in a given interface.<sup>8</sup> Additionally, the parameter  $k_{foil}/H_{foil}$ , where  $k_{foil}$  and  $H_{foil}$  are the thermal conductivity and the hardness of the metal foil, respectively, has been suggested as a good measure to predict the performance of a metal foil in a given application. Higher  $k_{foil}/H_{foil}$  ratios reduce thermal resistance at the interface.<sup>8</sup>

For metal foils with CNT-enhanced surfaces, heat flow paths and resulting thermal models become substantially more complicated. In addition to the properties of the metal foil, the effective thermal and physical properties of CNT/ foils depend on, among other factors, CNT density, CNT diameters in the array, and the bonding of the CNTs to the foil. Previous studies have shown not only that CNT arrays conform well in an interface but also that they have relatively high effective thermal conductivities ( $\sim 80 \text{ W/m K}$ ) and can be bonded well to their growth substrate.<sup>2,3,5,9</sup> These CNT array properties can be exploited, through optimization, to create a CNT/foil material whose effective thermal conductivity to effective hardness ratio  $k_{\text{CNT/foil}}/H_{\text{CNT/foil}}$  is greatly increased as compared to a bare metal foil. Both  $k_{\rm CNT/foil}$  and  $H_{\rm CNT/foil}$  are affected by the CNT array properties; however, reducing  $H_{\text{CNT/foil}}$  is expected to be the primary means to increase  $k_{\text{CNT/foil}}/H_{\text{CNT/foil}}$ . Also, CNT/foil characteristics such as the thicknesses of the CNT arrays and the metal foil can be controlled such that the contact geometry allows interfacial void spaces to be filled completely, thus overcoming the resistance to heat flow caused by the roughness of a given interface.

Plasma-enhanced chemical vapor deposition<sup>6</sup> (PECVD) was used to synthesize the CNT arrays in this study. A trilayer catalyst configuration<sup>2</sup> (30 nm Ti/10 nm Al/3 nm Fe) was deposited on both sides of 10  $\mu$ m thick Cu foil. The PECVD process gases were H<sub>2</sub> [50 SCCM (SCCM denotes cubic centimeter per minute at STP)] and CH<sub>4</sub> (10 SCCM), and the growth pressure and temperature were 10 Torr and

<sup>&</sup>lt;sup>a)</sup>Author to whom correspondence should be addressed; electronic mail: tsfisher@purdue.edu



FIG. 1. CNT arrays synthesized on both sides of a 10  $\mu$ m thick Cu foil. The density is ~10<sup>8</sup> CNTs/mm<sup>2</sup>. Both CNT arrays are approximately 50  $\mu$ m in height and the average CNT diameter is approximately 20 nm.

900 °C, respectively. A 200 W plasma was formed in the growth chamber, and CNT synthesis was carried out for 10 min. Figure 1 contains a scanning electron microscope (SEM) image that shows a side view of the two CNT arrays synthesized on the Cu foil. As determined from microscopy (SEM and transition electron microscope) and Raman spectroscopy, the structural characteristics (e.g., prevalence of CNT defects and amorphous C) of the CNT arrays are similar to CNT arrays grown on Si in previous work.<sup>2,3,5,9</sup> Each array is fairly uniform in height (approximately 50  $\mu$ m) and the average CNT diameter is approximately 20 nm. The density of each array is approximately ~10<sup>8</sup> CNTs/mm<sup>2</sup>, similar to previous work.<sup>5</sup>

The room-temperature thermal interface resistance of a CNT/foil TIM and bare 10  $\mu$ m thick Cu foil was measured as a function of pressure using a photoacoustic (PA) technique.<sup>3</sup> The pressure range was chosen to identify the effects of CNT enhancement in a range applicable to the thermal management of electronic components. Two different interfaces, in which the CNT/foil and bare foil were inserted, were assembled to identify the effects of surface roughness on the performance of the CNT/foil TIM. To enable the most accurate PA measurements, both interfaces use Ag for the top substrate in the interface. The Ag is relatively smooth, having an average surface roughness  $R_a$  of 0.06  $\mu$ m and an average peak-to-valley surface height  $R_z$  of 0.4  $\mu$ m, calculated according to Ref. 10. A polished Si base, having  $R_a = 0.01 \ \mu \text{m}$  and  $R_z = 0.09 \ \mu \text{m}$ , was used as the opposing substrate in the first interface (Si-Ag). A Cu base, having  $R_a = 2.8 \ \mu \text{m}$  and  $R_z = 9.3 \ \mu \text{m}$ , was used as the opposing substrate in the second interface (Cu-Ag). The total thermal resistances of the CNT/foil, R<sub>CNT/foil</sub>, and of the bare foil,  $R_{\rm foil}$ , for the two different interface configurations are presented in Fig. 2. For both configurations, the CNT/foil was examined before and after testing to assess any permanent physical changes to the material. For the Si–Ag interface, the CNT/foil's appearance closely resembled the pretest condition upon removal. For the rough Cu-Ag interface, deformation of the CNT/foil was apparent such that its shape matched the interface geometry. In each case, upon separation of the interface, the CNT arrays remained fully intact on the surfaces of the foil. To illustrate its robustness, after removal, the CNT/foil was retested in each interface, and the measured thermal resistances were consistent with the initial tests.

The bare Cu foil and the CNT/foil TIMs produce very



FIG. 2. (Color online) Thermal resistances of bare foil interfaces,  $R_{\text{foil}}$ , and CNT/foil interfaces,  $R_{\text{CNT/foil}}$ , as a function of contact pressure.

result is expected because of the relatively smooth contacting member surfaces. The plots of Fig. 2 illustrate that the CNT arrays provide greater enhancement to the thermal conductance of the Cu foil in the rougher Cu–Ag interface. A reduction in resistance of approximately 30% is achieved at a contact pressure of 275 kPa. For the smooth Si–Ag interface, enhancement is apparent when sufficient contact pressure is applied, and an approximately 15% reduction in resistance is achieved at a contact pressure of 275 kPa. Moreover, upon closer examination of the interface details, further inferences can be made from the data.

We believe that the CNT/foil material increases conduction in the interface by two major mechanisms, both of which cause an increase in the number density of contact points between free CNT tips and their opposing substrate. The first mechanism is the deformation of the CNT arrays and the second is the deformation of the foil. We postulate that the CNT array deformation is elastic (i.e., there was no evidence of tube buckling), although van der Waals interactions among the tubes can cause them to bundle together after experiencing interfacial compression (mimicking the geometry of the surface asperities), and that the Cu foil deformation is both elastic and plastic as in the case of bare foil. For each interface configuration, the CNT arrays and foil deform concurrently with increased pressure. For the Si-Ag interface, the CNT/foil deforms with increased pressure until a condition exists at which it no longer deforms, and its improved thermal performance over that of the bare foil becomes constant. Even for this smooth interface, the slight deformation of the foil around the surface asperities (primarily on the Ag surface) is apparently sufficient to increase the number density of contact points between free CNT tips and their opposing substrate.

For the relatively rough Cu–Ag interface, the effect of the Cu foil's deformation is more significant due to the larger surface asperities that likely prevent CNTs from initially bridging the interface gap. For this interface, the CNT/foil exhibits significantly better thermal performance than the bare foil because its foil component deforms under high local stress to match the asperities of the interface while the CNTs presumably deform along with it to create substantially more

low thermal resistances in both interface configurations. This contact points. For this interface, a maximum deformation Downloaded 27 Apr 2011 to 130.207.50.192. Redistribution subject to AIP license or copyright; see http://apl.aip.org/about/rights\_and\_permissions



FIG. 3. (Color online) Thermal circuit for the CNT/foil interface. The local resistances sum to give  $R_{\text{CNT/foil}}$ .

extent did not occur in the tested pressure range, and it is expected that under higher pressures, the CNT/foil would continue to conform to the interface, further improving its performance as compared to that of the bare foil.

The two deformation mechanisms of the CNT/foil aid in increasing the number density of contact points between free CNT tips and their opposing substrate. To better illustrate the enhancements that occur at both of the free CNT tip interfaces, the PA method has been used to measure local component resistances within the interface structure. A resistive network for the CNT/foil interface is illustrated in Fig. 3. The interface resistance between a CNT array and its Cu growth substrate ( $R_{CNT-Cu}$ ), approximately 1 mm<sup>2</sup> K/W, and the effective thermal conductivity of CNT arrays synthesized under conditions similar to the ones of this study, approximately 80 W/m K (which corresponds to an intrinsic resistance  $R_{CNT}$  of approximately 1 mm<sup>2</sup> K/W for each CNT array in this study), have been measured in previous work.<sup>5,9</sup> For the CNT/foil material, the combined resistance of both CNT arrays, both CNT-foil interfaces, and the Cu foil  $(<0.3 \text{ mm}^2 \text{ K/W})$  sums to approximately 4 mm<sup>2</sup> K/W. The remaining resistance in the CNT/foil interface is therefore produced by the resistance between the CNT arrays' free tips and the two contacting members  $(R_{\text{Si-CNT}} + R_{\text{CNT-Ag}}$  for the Si-Ag interface and  $R_{Cu-CNT}+R_{CNT-Ag}$  for the Cu-Ag interface).

The resistances at the free surfaces of the bare foil (same as  $R_{\text{foil}}$ ) and the free surfaces (i.e., free CNT tips) of the CNT/foil are illustrated in Fig. 4 for both interface configurations. Clearly, the thermal resistance at the contacting member interfaces is greatly reduced by the presence of the CNT arrays. A reduction in resistance greater than 50% is observed for both interface configurations at moderate pressure. These results suggest that the CNT/foil configuration is highly effective in increasing the number density of contact points between free CNT tips and their opposing substrate and thus provides an effective means of increasing the real contact area in a thermal interface.



FIG. 4. (Color online) Thermal resistance between the two free surfaces of the samples. For the bare foil, the resistance is the same as  $R_{\text{foil}}$ . For the CNT/Foil the resistance is the sum of the two free CNT tip interface resistances.

In summary, a CNT/foil TIM has been fabricated, that is capable of providing high conformability to the surface roughness that is inherent to every real interface. This TIM reduces the effects of surface roughness on thermal transport across an interface, and accordingly, resistances near 10 mm<sup>2</sup> K/W are attained for rough and smooth interfaces at moderate pressures. Furthermore, the free-tip interface resistances achieved by the CNT/foil are significantly lower than previous measurements.<sup>4,5</sup> The performance of the CNT/foil in a given interface is a function of several parameters (e.g., the CNT density, length, and diameter, and the foil material and 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. The CNT/ foil of this study was not optimized for the interfaces to which it was applied. We expect that a CNT/foil TIM optimized for a particular interface application will produce significantly superior results.

The authors gratefully acknowledge financial support from the Air Force Research Laboratory. The lead author (B.A.C.) is grateful for personal support from the Intel Foundation and the Purdue University Graduate School.

<sup>1</sup>M. J. Biercuk, M. C. Llanguno, M. Radosavljevic, J. K. Hyun, A. T.

- Johnson, and J. E. Fischer, Appl. Phys. Lett. 80, 2767 (2002).
- <sup>2</sup>J. Xu and T. S. Fisher, IEEE Trans. Compon. Packag. Technol. **29**, 261 (2006).
- <sup>3</sup>J. Xu and T. S. Fisher, Int. J. Heat Mass Transfer **49**, 1658 (2006).
- <sup>4</sup>T. Tong, Y. Zhao, L. Delzeit, A. Majumdar, and A. Kashani, *Third ASME Integrated Nanosystems Conference* (ASME, New York, 2004), Paper No. NAN02004-46013.
- <sup>5</sup>B. A. Cola, J. Xu, C. Cheng, H. Hu, X. Xu, and T. S. Fisher, J. Appl. Phys. 101, 060704 (2007).
- <sup>6</sup>M. R. Maschmann, P. B. Amama, A. Goyal, Z. Iqbal, R. Gat, and T. S. Fisher, Carbon **44**, 10 (2006).
- <sup>7</sup>P. B. Amama, O. Ogebule, M. R. Maschmann, T. D. Sands, T. S. Fisher, Chem. Commun. (Cambridge) **27**, 2899 (2006).
- <sup>8</sup>M. M. Yovanovich, AIAA Pap. 72, 283 (1972).
- <sup>9</sup>X. Hu, A. A. Padilla, J. Xu, T. S. Fisher, and K. E. Goodson, J. Heat Transfer **118**, 1109 (2006).
- <sup>10</sup>Surface Texture, Surface Roughness, Waviness and Lay (American Society of Mechanical Engineers, New York, 2003), AMSE B46.1-2002, pp. 1–13.