Infrared Microscopy Thermal Characterization of Opposing Carbon Nanotube Arrays

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Carbon nanotubes (CNTs) have received much recent research interest for thermal management applications due to their extremely high thermal conductivity. An advanced thermal interface structure made of two opposing, partially overlapped CNT arrays is designed for thermally connecting two contact surfaces. The performance of this interface structure is thermally characterized using diffraction-limited infrared microscopy. Significant temperature discontinuities are found at the CNT-CNT contact region, which indicates a large thermal resistance between CNTs. Due to this intertube resistance, the thermal performance of the CNT-based interface structure is far below expectation (with a thermal resistance value about $3.8 \times 10^{-4}$ K m$^2$/W).

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Introduction

Carbon nanotubes (CNTs), a man-made material first reported by Iijima in 1991 [1], are promising for advanced thermal management because of their extremely high thermal conductivity. Hone et al. [2] found that the thermal conductivity of aligned single-walled nanotube (SWNT) ropes is about $250$ W/mK at $300$ K, and that the thermal conductivity of an individual SWNT in the longitude direction ranges from $1750$ to $5800$ W/mK. The thermal conductivities of individual CNTs (multi-walled [3] or single-walled [4]) have also been experimentally tested: at least $3000$ W/mK at room temperature, which is about one order higher than that of the high thermal conductivity materials commonly used for thermal management (for example, copper). Theoretical studies even predict much higher thermal conductivities, such as $6600$ W/mK at room temperature reported by Berber et al. [5].

The technique of using CNTs for thermal management, however, is not straightforward due to the nanoscale nature of CNTs. Early attempts used CNTs as fillers to form high thermal conductivity fluids or composites. Choi et al. [6] measured the effective thermal conductivity of nanotube-in-oil suspensions, and found that with only 1 vol.% of nanotubes, the effective thermal conductivity can be 2.5 times the value of the base fluid. Such an increase in thermal conductivity has never been found previously with any other particles. Biercuk et al. [7] also found that epoxy filled with 1 wt.% of CNTs showed a 70% increase in thermal conductivity at 40 K and 125% at room temperature. Hu et al. [8] proposed the combined use of CNTs and traditional heat conductive fillers for thermal interface materials (TIMs), achieving a thermal conductivity value seven times that of the base fluid, and almost doubling the thermal conductivity of the corresponding TIM composed of only traditional fillers. However, two problems with CNT-based TIMs lead to low thermal conduction efficiencies. One problem is that CNTs are randomly dispersed, and thus only a small portion of CNTs are effectively contributing to heat conduction. The other problem is that heat is not directly conducted from one side to the other through CNTs. CNTs are discontinued by other fillers or the base fluid. The low thermal conductivity of the interstitial media, as well as the contact resistance between those and the CNTs [9–11], degrades the thermal performance of CNT composites.

Most recent interest focuses on growing CNTs directly on a silicon or copper substrate, with the CNTs oriented in the direction of heat conduction (i.e., perpendicular to the substrate). These CNTs are mechanically like an elastic cushion due to their high aspect ratio and mechanical strength [12], and therefore can be used between a thermal expansion mismatched interface (for example, the interface between a CPU die and its heat spreader, or the interface between a heat spreader and a heat sink) as a highly heat conductive interface structure [13–17]. Hu et al. [18] measured the effective thermal conductivity of this CNT layer, including the effects of voids between CNTs, to be about $80$ W/mK, which is one order higher than that of thermal greases (the most widely used TIMs). Unfortunately, the over all thermal resistance across the CNT layer, including CNT contacts, is still too large due to the thermal bottleneck where the CNTs contact the other interface.

The understanding of heat conduction at CNT contacts is very limited. Part of the problem is the lack of effective thermal characterization method capable of resolving temperature distributions across CNT contacts at small scale. In this paper, we attempted to use the diffraction-limited infrared microscopy to measure the heat conduction in a CNT-based thermal interface structure. This CNT structure is formed by growing two opposing CNT arrays from two facing surfaces. The purpose of growing CNT from both surfaces is to avoid the poor CNT-surface contact. A similar struct-

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ture has been made by [19]. This work addresses the remaining question concerning the thermal performance of the CNT-CNT contact region, in which CNTs are discontinued and heat is forced to transport from one CNT to another.

Infrared Microscopy Measurement

Sample Preparation. An ideal interface structure with partial CNT overlaps is difficult to grow directly due to the limitations of the CNT growth process. To grow vertically oriented CNTs, chemical vapor deposition (CVD) or plasma enhanced chemical vapor deposition are generally used. CNTs are assembled with carbon atoms from hydrocarbon gases though a well-controlled oxidation process with proper catalyst. When the CNTs are grown from two facing surfaces close to each other, the CNTs will block the gas flow, shutting themselves off from the carbon source, and thereby prohibiting the growth necessary to form an interface structure with CNT overlaps.

In this study, we make the CNT interface structure using two separate CNT samples (see Fig. 1 for scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images). These CNT samples were grown on the rough side of 15 mm×15 mm single-side-polished silicon substrates using thermal CVD. On the back side of one sample, a tungsten heater was integrated. The length of the CNTs on the heater substrate (CNT1) is about 150 μm and the length of the CNTs on the other chip (CNT2) is about 100 μm, estimated by regular optical microscope. All these CNTs are oriented perpendicular to the silicon substrates, and on average, the diameter of the tubes (d) is about 6±2 nm, and the half-distance between the axes of two tubes (w) is about 8 nm, based on high-magnification SEM and TEM images. Most of the tubes are double walled.

The CNT interface structure is made by pressing the two pieces of the CNT samples together, as illustrated in Fig. 2. After removing the pressure, the two samples remain mechanically bonded to each other due to the strong van der Waals force between CNTs. The final distance between the two substrates, without any press, is about 200 μm, and therefore the CNT overlap region L is estimated to be around 50 μm.

Experiment Setup and Method. The experiment system is given in Fig. 3. The CNT interface structure is attached to a copper cold plate using thermal grease. The cold plate is cooled by a thermo-electrical cooler and a water-cooling heat sink. The integrated heater on the other side of the CNT sample is connected using two probes. This whole structure, as shown in part (a), is then mounted on a stage with precisely controlled translational and rotational movement, and placed under an infrared microscope, as shown in part (b). The IR microscope features a 256×256 InSb focal plane array, with a detection wavelength ranging from 3 to 5 μm and a temperature sensitivity of 0.1 K. The spatial resolution of the IR microscope is about 2 μm, which is close to the diffraction limit at the given wavelength. The surfaces exposed to the IR microscope, including the CNT sample to be examined, the copper cold plate, and the thermal grease in between, are coated with a thin layer of amorphous carbon to achieve relatively uniform emissivity.

The experimental system needs a two-step calibration process prior to the measurement. The first step is to map the radiance intensity, including the effects of radiance from ambient, with absolute temperatures. This step is done by testing a standard blackbody at given temperatures. The second step is to map out the emissivity of the carbon coating, which may not be exactly uniform. During this calibration step, the thermo-electrical cooler is operated in its heating mode. The whole structure, including the copper plate and the CNT sample, is uniformly heated and reaches a stable final temperature that can be measured by the thermocouples attached to the backside of the copper plate. The temperature uniformity of the test surface is checked under this isothermal condition, and the calibrated radiance is used as a reference to determine the emissivity of the carbon coating, including the effect of surrounding radiance. The relationship between the reference radiance and the effective emissivity has been given in [20].
Attertube thermal resistance is of tube overlap, respectively. The equation for estimating this in-area $A_e = 1.4 \text{ K m}^2/\text{W}$, based on an effective tube heat exchange surface drop is found at the end of the CNT-CNT overlap region. This along the axial direction, and that the contact resistance at the hot region and the cold region, the temperature distributions are.

The data indicate that the thermal resistance of the interface structure is far below expectation. The problem is the large intertube contact resistance between the CNTs, which results in a significant temperature drop at the end of the CNT-CNT contact region when the CNT interface structure is parallel to the axial direction, and that the contact resistance at the CNT growth surface is small. However, an apparent temperature drop is found at the end of the CNT-CNT overlap region. This temperature drop implies a significant CNT intertube thermal resistance in the CNT overlap region, which is estimated to be about $1.4 \text{ K m}^2/\text{W}$, based on an effective tube heat exchange surface area $A_e = \pi d L$, where $d$ and $L$ are the tube diameter and the length of tube overlap, respectively. The equation for estimating this intertube thermal resistance is

$$R_t = \frac{T_1 - T_2}{P/\phi A_t} (A_r/A_t)$$

(1)

where $T_1$ and $T_2$ are the temperatures of CNT1 and CNT2, $P$ is the heater power, $\phi$ is the tube volume fraction, $A_t$ is the area of the CNT interface structure, and $A_r = \pi d^2/4$ is the cross-sectional area of a single tube.

Due to the large intertube thermal resistance, the thermal performance of the CNT interface structure is far below expectation. The data indicate that the thermal resistance of the interface structure ($R_t$) is about $3.8 \times 10^{-4} \text{ K m}^2/\text{W}$ according to

$$R_t = \frac{T_1 - T_2}{P/\phi A_t}$$

(2)

Conclusions

An interface structure made by pressing two pieces of vertically oriented CNTs against each other is characterized using diffraction-limited IR microscopy. It is found that the thermal performance of the CNT interface structure is far below expectation. The problem is the large intertube contact resistance between the CNTs, which results in a significant temperature drop at the end of the CNT-CNT contact region when the CNT interface structure is subject to a constant out-of-plane heat flux. Many experimental and modeling efforts are still needed to understand the mechanism of thermal coupling between CNTs.

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Nomenclature

- $A_e$: effective heat transfer area per tube, $\text{nm}^2$
- $A_t$: cross-sectional area of a single tube, $\text{nm}^2$
- $A_s$: area of the sample, $\text{cm}^2$
- $d$: diameter of a nanotube, $\text{nm}$
- $L$: length of the tube overlap region, $\mu\text{m}$
- $P$: heater power, $\text{W}$
- $R_t$: thermal resistance between CNTs, $\text{K m}^2/\text{W}$
- $T_1$: temperature of CNTs on the hot side, $\text{K}$
- $T_2$: temperature of CNTs on the cold side, $\text{K}$
- $w$: distance between two adjacent nanotubes, $\text{nm}$

References