ACS APPLIED MATERIALS & INTERFACES

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Efficient Natural-Convective Heat Transfer Properties of Carbon Nanotube Sheets and Their Roles on the Thermal Dissipation

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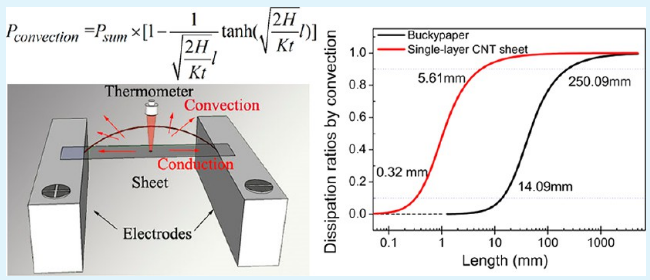
ABSTRACT: In this work, we report our studies related to the natural-convective heat transfer properties of carbon nanotube (CNT) sheets. We theoretically derived the formulas and experimentally measured the natural-convective heat transfer coefficients (H) via electrical heating method. The H values of the CNT sheets containing different layers (1, 2, 3, and 1000) were measured. We found that the single-layer CNT sheet had a unique ability on heat dissipation because of its great H. The H value of the single-layer CNT sheet was 69 W/(m^2 K) which was about twice of aluminum foil in the same environment. As the layers increased, the H values dropped quickly to the same with that of aluminum foil. We also discussed its roles on thermal dissipation, and the results indicated that the convection was a significant way of dissipation when the CNT sheets were applied on macroscales. These results may give us a new guideline to design devices based on the CNT sheets.



KEYWORDS: *carbon nanotube sheets, thermal convection, thermal conduction, thermal managements*

INTRODUCTION

Recently, carbon nanotube (CNT) sheets gotten from superaligned CNT arrays have attracted great attention.[1,2] These unidirectional sheets are transparent and macroscale in length and width, but their thickness is on nanoscale. They have been applied on thermoacoustic loudspeakers,[3] flexible touch screens,[4] CNT bending actuators,[5,6] optically switchable windows,[7] supercapacitors,[8] lithium-ion batteries,[9] and organic solar cells.[10] Among these applications, loudspeakers and bending actuators synthetically utilize the sheets' properties on electricity, thermology and morphology (nanoscale thickness). In these devices, electrical energy first converts to thermal energy, and then to mechanical energy. The heat propagation properties of CNT sheets directly determine the transfer process of thermal energy to mechanical energy. Thereby affect the work of devices.

For examples, Xiao et al. made a thermoacoustic loudspeaker, which only needed to put CNT sheets on two electrodes.[3] When periodic current passed through the sheets, there was a period temperature on them, which caused a period thermal vibration in the surroundings. The intensity of thermal vibration was affected by the heat from sheets to surroundings. Chen et al. fabricated an easy-operable bending actuator based on CNT sheets.[5] In that work, CNT sheets were bonded to polymer. Because of their different thermal expansion coefficients, the actuator would be bent when it was electrically heated. The rate of bending or shape recovery was determined by the rate of thermal dissipation.

In contrast to the rich researches on thermal properties of individual CNT[11−14] and CNT arrays,[15−17] there is few report on the thermal properties of these sheets. Aliev et al. measured the thermal conductivity and thermal diffusivity of these sheets.[18] But for these macroscopically thin and transparent but microscopically spongy sheets, due to their large specific surface (nanoscales on thickness), there should be large heat dissipated by convection. Some researchers have pointed out that environments have a significant effect on dissipation of the individual CNT, for instance, whether it is connected with substrates[19,20] or whether it is in a vacuum.[21] The main object connected with these sheets is usually air during applications. Natural-convective heat transfer coefficients (H) directly show the strength of convection. We need to know H to simulate the temperature distribution of CNT sheets, which has a great effect to electrical behaviors of devices based on these sheets.[22] For the usual solid materials, the H can be gotten through a process of natural cooling in air. But for these sheets, their H values are difficult to obtain by this traditional method of transient states because of their small thermal capacity.

In this paper, we used a steady-state electrical heating method to measure the H values of the CNT sheets. We theoretically derived the formulas of calculating H, thereby experimentally measured H of the CNT sheets with different layers and so that the buckypaper[23] (up to 1000 layers overlapped together). As a comparison, the H of aluminum foil was also measured using a traditional method. We found that the H of single-layer CNT sheet was twice of aluminum foil in

Received: December 2, 2013
Accepted: February 18, 2014
Published: February 18, 2014

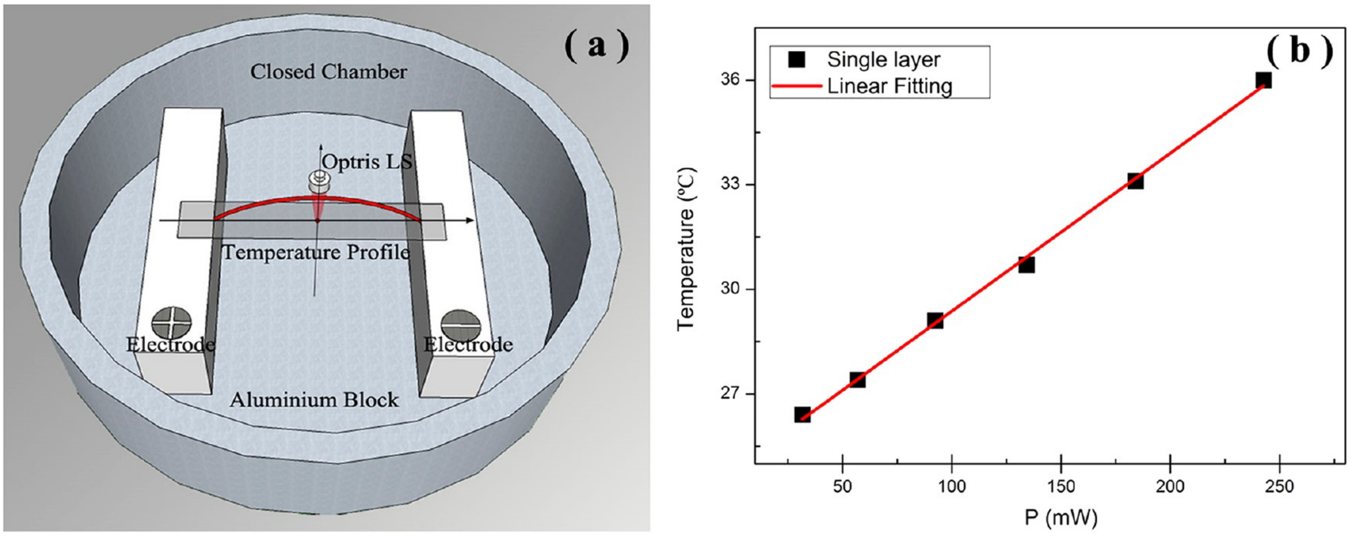


Figure 1. (a) Schematic of the experimental setup for the natural-convective heat transfer coefficient measurements. Keithley 2410 source meter provided working voltage; an Optris LS infrared thermometer measured the temperature; the closed chamber maintained a stable environment and its top was closed in the measurements. (b) Relation between the central temperature of the single-layer CNT sheet and the heating power.

the same environment, and as the layers increased, the H decreased quickly to the same with that of the aluminum foil. We think the alteration of effective contact areas can explain this phenomenon.

■ THEORETICAL BASIS

A self-supporting CNT sheet is spanned between two aluminum blocks which are large enough in volumes and masses. When a small voltage is applied, there will be an increment on temperature owing of joule heating. When its temperature is not much higher than surroundings, heat radiation is much weaker. The main ways of dissipation are conduction and convection. Combining the two effects, the CNT sheet will get an equilibrium temperature. Analyzing the temperature, we can get the information of thermal conductivity and H.

The CNT sheet is homogeneous and its length is much larger than its width and thickness. The process of heat transfer can be seen as one dimension along its axial direction. When the system gets to the equilibrium state, we can get the following Fourier equation

$$-kwt\frac{dT^2}{dx^2} + 2Hw \times (T - T_0) = UI/2l \tag{1}$$

where U is the voltage of the CNT sheet, I is the current, l is half of the length, w is the width, H is the convective heat transfer coefficient, T is the temperature of the CNT sheet, T_0 is the environment temperature. At the boundary of the CNT sheet, the blocks are two large heat reservoirs. We can get $T(-l,\infty) = T_0$ and $T(l,\infty) = T_0$. Using this boundary condition, we can get the solution of eq 1

$$T(x, \infty) = \frac{P}{2Hw}\left(1 - \frac{\cosh\left(\sqrt{\frac{2H}{Kt}}x\right)}{\cosh\left(\sqrt{\frac{2H}{Kt}}l\right)}\right) + T_0 \tag{2}$$

At the center point, $x = 0$, then we can get

$$T(0, \infty) = \left(1 - \frac{1}{\cosh\left(\sqrt{\frac{2H}{Kt}}l\right)}\right) \times \frac{P_{sum}}{4Hwl} + T_0 \tag{3}$$

where P_{sum} ($P_{sum} = UI$) is the total power of joule heating.

If the test is carried out in vacuum, then $H = 0$, and we can directly calculate K. In fact, when H is near zero, Second-order Taylor expansion of the exponential terms in formula 3, we can get

$$T(0, \infty) = \frac{l}{4Kwt}P_{sum} + T_0 \tag{4}$$

This formula is also the principle of calculating thermal conductivity using electrical heating.[24]

For the CNT sheets used in this work, the typical mass per unit area is 1.5 ug/cm^2. The density of CNT bundles is 1.58 g/cm^3 based on hexagonally closed-packed model.[23] According to closed-packed model, the effective thickness of the single-layer CNT sheet is (1.5 ug/cm^2)/(1.58 g/cm^3) ≈ 10 nm. The thermal conductivity of individual multiwalled CNT is about 1400 W/(m K).[14] Because structure defects and tube−tube interface,[25−27] the thermal conductivity of CNT sheets should be smaller than that of individual multiwalled CNT. In natural convective heat transfer situation, H usually changes between 10 and 50 W/(m^2 K). To reduce the effect of interfacial thermal resistance, l is selected to 20 mm in our experiments. Then we can get that $\cosh((2H/Ktn)^{1/2}l) = [\exp((2H/Ktn)^{1/2}l) + \exp(-(2H/Ktn)^{1/2}l)]/2 \gg 1$ (n = layers of sheets; for $H = 30$ W/(m^2 K), $K = 1000$ W/(m K), $t = 10$ nm, if $n = 1$, $\exp((2H/Ktn)^{1/2}l) = 1.1 \times 10^{15}$; $n = 20$, $\exp((2H/Ktn)^{1/2}l) = 2312$; $n = 500$, $\exp((2H/Ktn)^{1/2}l) = 4.7$) when n is not very large. If $n < 20$, then we can rewrite formula 3 as

$$T(0, \infty) = \frac{P_{sum}}{4Hwl} + T_0 \tag{5}$$

Because $\cosh((2H/Ktn)^{1/2}l) > 1000$, this treatment can ensure the error less than one thousandth. Physical meaning of formula 5 is that nearly all the heat is dissipated by convection when the conduction is very small. Please note that if $n > 500$,

dx.doi.org/10.1021/am405491t | ACS Appl. Mater. Interfaces 2014, 6, 3075−3080

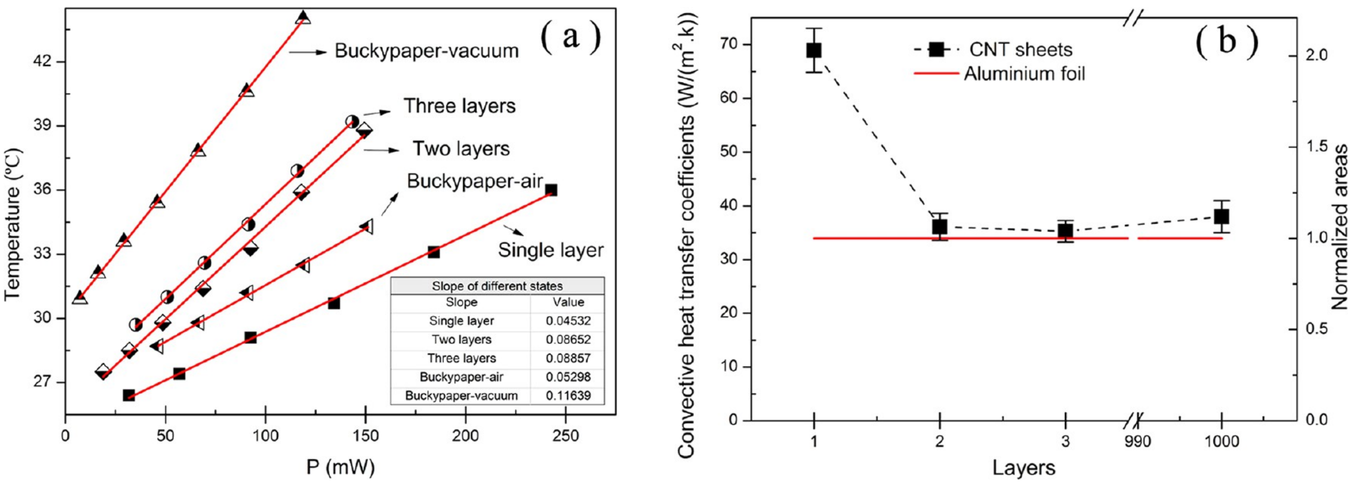


Figure 2. (a) Relations between the central temperature and the heating power for the CNT sheets; (b) H values of the CNT sheets containing different layers. Red line denotes the H of the aluminum foil.

this approximation can bring great error, so one has to calculate H according to frmula 3.

On the basis of formula 5, when the sheet and environment are selected, H, l, w, and T_0 are identified, and $T(0,\infty)$ is related only to P_{sum}. There are two methods to calculate H. (1) Using single $T(0,\infty)$, P_{sum}, and T_0, we can get H. (2) We can get a series of $T(0,\infty)$ by changing P_{sum}. Linear fitting $T(0,\infty)$ and P_{sum}, H can then be gotten via the slope. This method can decrease test errors due to average multiple data. In our experiments, we used the second method to calculate H.

So, in our experiments, we only need to change P_{sum} to get a series of central point's temperature. For CNT sheets (1, 2, and 3 layers), we use formula 5 to get H. For the buckypaper, its conduction is large. First, we need to calculate its thermal conductivity based on formula 4 in a vacuum, then combined it with formula 3 to obtain H.

■ EXPERIMENTAL SECTION

The transparent and conductive CNT sheets were drawn out from superaligned CNT arrays.[28] Their typical density of per unit area was about 1.5 ug/cm^2. The apparent thickness of single-layer CNT sheet was about several micrometers but was difficult to be measured exactly because of its aerogel-like feature. After densified using ethanol the thickness could be tens of nanometers. All the CNTs formed the sheets were multiwalled CNTs. Figure 1a schematically illustrates the experimental setup. The sheet without any treatments was spanned between two aluminum blocks and cut into 4 mm in width by the laser. Alcohols and silver paste were both applied on the sheet/block interface to decrease the interfacial thermal resistance. The distance between blocks was 40 mm, and the height of blocks was 12 mm. Keithley 2410 source meter was used to supply working power. An Optris LS infrared thermometer whose temperature and spatial resolution were 0.1 K and 1 mm was employed to measure the temperature at the center of the sheet. The highest temperature increment was less than 15 K. In this situation, the radiation was weak, and most of the heat that dissipated to the air was by the convection. The entire equipment was placed in a closed chamber, which ensured all the measurements were in the same environment. This measurement is noncontact and the radiation is weak, we can use formula 5 to calculate H for single-layer, two-layer, and three-layer CNT sheets based on the above method.

The buckypaper was gotten from 1000-layer CNT sheets overlapped together. It was made by the continuous spinning process with the specific equipment. The thickness of the buckypaper was 70 um. First, we measured the thermal conductivity of the buckypaper in a vacuum chamber using the same equipment except for a thermocouple instead of the Optris LS infrared thermometer. By calculating according to formula 4, the thermal conductivity of buckypaper was 153 W/(m K). Combined with formula 3, the H of the buckypaper could be obtained.

We need to note that in our experiments all the emissivity coefficients of the sheets were set to 0.95. The emissivity coefficient of the buckypaper was calibrated with a thermocouple by measuring the temperature on the same point and it was 0.95. For few-layers CNT sheets, they could not be directly calibrated with a thermocouple because of their vulnerability. We used the similar structure as shown in Figure 1a to determine the emissivity of the few-layers CNT sheets but one of the aluminum blocks was heat, we measured the temperature neighbor the aluminum block, and found only when the value of the emissivity coefficient was set to be larger than 0.9 we could get the reasonable temperature. On the other hand, the displayed temperature had little change when the value was larger than 0.9. So in our experiments, the values of emissivity for all the CNT sheets were set to 0.95.

For the aluminum foil, we used a traditional method to measure H. In brief, a piece of aluminum foil whose area and thickness were 1×1 cm^2 and 0.58 mm was merely supported by two porous foams. Then it was heated by a laser to 338 K (the room temperature was 299 K). After closing the laser, the temperature of aluminum foil dropped to room temperature by convection according to the exponential decay. On the basis of the lumped capacitance model, the exponential decay coefficient was mC_p/HS, where m, C_p, and S are the mass, thermal capatance per unit mass, and the surface area, respectively. By exponentially fitting the temperature−time curve, we could get the value of the decay coefficient. H could then be obtained.

■ RESULTS AND DISCUSSION

Figure 1b shows the relation of the single-layer CNT sheet between the central temperature and the heating power. The fluctuation of the temperature was less than 0.3 K, and the error bars were covered by the black symbols in the figure. From it we can see that temperature is linear to heating power. So heat conduction equation is linear too, and we can neglect the effect of heat radiation in our experiments, which is nonlinear to heating power.

Using the same method, we measured the relations between central temperature and heating power for multilayer CNT sheets (Figure 2a). The results were stable and repeatable. From it, we can see that the single-layer sheet has the slowest heating rate despite its thermal conduction is the weakest. This hints single-layer sheet has a great H. For the buckypaper, the heating rates vary largely from whether it is put in air. We can

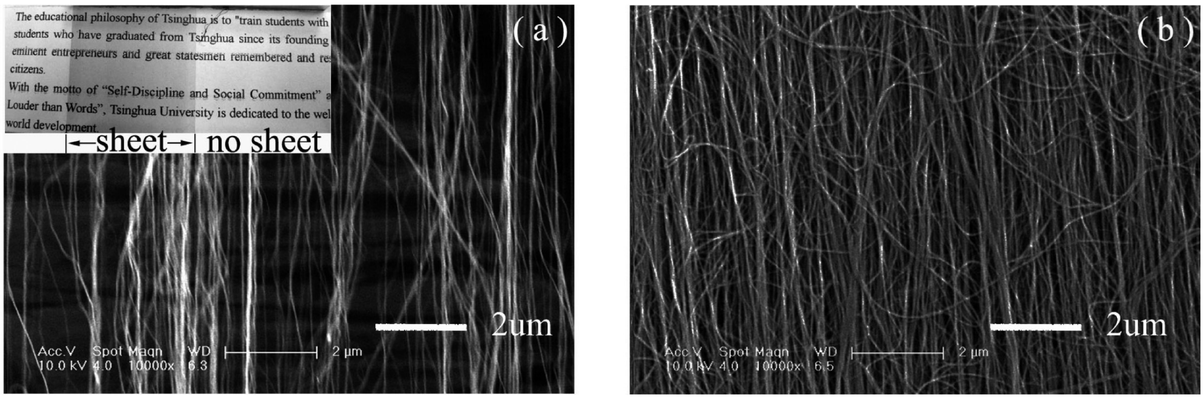


Figure 3. SEM images of (a) the single-layer CNT sheet (background is the substrate of aluminum.) and (b) the buckypaper. The inset in a) is the optical image of the transparent single-layer CNT sheet.

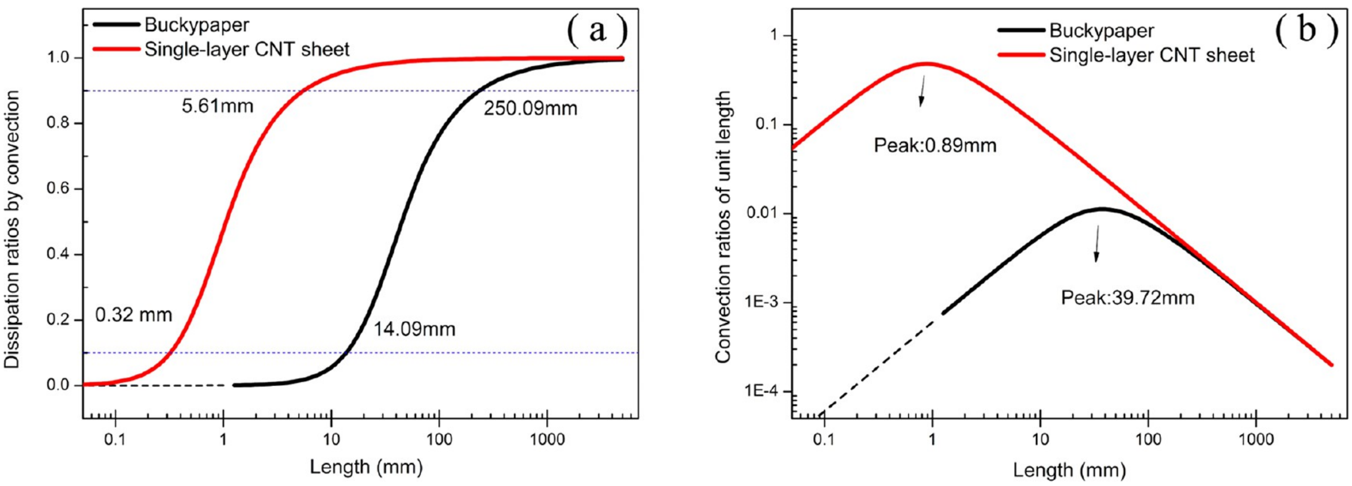


Figure 4. Dissipation ratios by convection for the single-layer CNT sheet and the buckypaper. Dotted line is extended result by theoretical calculations. (The buckypaper thickness is 60um. To meet the one-dimensional hypothetical, the length should be much larger than thickness. We take the minimum length as 1 mm.) (a) Dissipation ratios by convection; (b) dissipation ratios by convection per unit length.

also see that air has a great effect on its equilibrium temperature. All of these indicate air has a significant effect on heat dissipation of the sheets. Buckypaper has more thermal conductive channels than two and three layers' CNT sheets. Higher conduction should be the reason of the slower heating rate in air.

From the slopes of the CNT sheets in Figure 2a, we got the H values. The H of the single-layer sheet is 69 W/(m^2 K) which is nearly double of aluminum foil, and when more than one layer is overlapped, the H drops to the same with aluminum foil (Figure 2b).In Figure 2b, the error bars means uncertainty in groups of test results. The H of the CNT sheets we have gotten has a big difference to the estimated H of individual CNT which ranges from 1×10^3 to 1×10^5 W/(m^2 K).[29] But our results here can compare the H of general solids. This shows the principle of convective heat transfer strength for CNT sheets is more similar to general solids.

We would like to give a reasonable explanation for our results based on enlargements of effective contact areas. For the CNT sheets, there is a big van der Waals force between CNTs. When multilayer CNT sheets overlapped together, the CNT sheets attract together closely, and the sheets become dense because of the Van de Waals force, which is a long-range interaction. From the SEM images as shown in Figure 3, we could see this phenomenon. The microstructures of the single-layer CNT sheet and the buckypaper have a big difference. The single-layer CNT sheet is thin and loose and the buckypaper is thick and dense. For the single-layer CNT sheet, the thin and loose structure provides gaps to make air flow inside. Part of air between CNTs also can flow to take the heat away due to the thin and loose structure. So not only the external CNTs but also internal CNTs are beneficial to exchange heat with surrounding air. But for the buckypaper, small gaps bring great challenge to the air flow between CNTs. Only the air directly contacted with the external CNTs can take amount of heat away., So only the external CNTs favor to exchange heat with air.[30] We think the increment of H for the single-layer CNT sheet is caused by the internal CNTs that directly contacts with the flow air. This would make the cooling areas increase. We define the total cooling areas as the effective contact areas. We can then get the enlargement rates between the effective contact areas and the outmost surface through dividing H of multilayer CNT sheets by the H of aluminum foil. In Figure 2b, the right vertical axis represents the enlargement rates of effective areas. We can see that the enlargement rate of single-layer CNT sheet is about two. When the layers increase, the enlargement rate rapidly decreases to one.

Single-layer CNT sheet has great H and demonstrates better ability on convective heat transfer. We further analyze the role of convection on the total thermal dissipation. We can get the dissipated power by conduction through

$$P_{\text{conduction}} = |-KS\nabla T|_{x=l}| + |-KS\nabla T|_{x=-l}| = \frac{P_{\text{sum}}}{\sqrt{\frac{2H}{Kt}}l}\tanh\left(\sqrt{\frac{2H}{Kt}}l\right) \quad (6)$$

T is the temperature profile shown in formula 2. The dissipation ratio by convection, which shows the percentage of convection, can be obtained through

$$\frac{P_{\text{convection}}}{P_{\text{sum}}} = 1 - \frac{P_{\text{conduction}}}{P_{\text{sum}}} = 1 - \frac{1}{\sqrt{\frac{2H}{Kt}}l}\tanh\left(\sqrt{\frac{2H}{Kt}}l\right) \quad (7)$$

We know that thermal conductance is inversely proportional to length (larger thermal resistance), and convection is proportional to length (bigger surface). So for the CNT sheets with different lengths, the dissipation ratios by conduction or convection will have a big difference. We need the thermal conductance of the single-layer sheet to calculate the dissipation ratios by convection. But the precise thermal conductance of it is hard to measured directly because of its ultrathin thickness, which makes some heat dissipated not by conduction and takes difficulties on the temperature measurement in vacuum. If we do not consider scattering between layers of the buckypaper, each layer has the same conductance. The conductance of the single-layer sheet can be obtained from the thermal conductance of the buckypaper divided by the layers. For comparison purpose, the dissipation ratios of the buckypaper are also calculated.

The dissipation ratios by convection of the single-layer CNT sheet and the buckypaper with different length are calculated on the basis of formula 7 and shown in Figure 4a. Specially, the Kt of the single-layer sheet is 153 W/(m K) × 70 um/1000. From the results, we can see that when the length is less, convection takes smaller portion of dissipation for both of them. With the increase of length, the roles of convection become important. For the single-layer sheet, when its length is less than 0.32 mm, less than 10% heat is dissipated by convection; when the length is more than 5.61 mm, more than 90% heat is dissipated by convection. This result also confirms that our approximation was reasonable on calculating H (length was 40 mm in our experiments), and we must consider convection on thermal analysis for the CNT sheets in macro applications. For the buckypaper, the positions of convection ratios equaling 10 and 90% correspond to the length 14.09 and 250.09 mm, respectively. Some researchers have pointed out that buckypaper possesses huge potential on chip cooling because of its great thermal conductivity.[24,31] Our results here show that the convection is an additional important way for the thermal managements (for PCs and smart phones, the length of cooling channels is about 100 mm). For the macro applications of buckypapers, we may take advantage of both conduction and convection.

For some applications, one focuses not only on the total heat transfer quantity but also on their intensity. For example, for the thermoacoustic loudspeaker, one needs more heat transfer into air per unit length to get a bigger sound pressure. We can obtain this intensity through the convection ratios dividing the length. Dissipation ratios by convection of unit length are shown in Figure 4b. There is a peak in the curves. The peak positions of the single-layer CNT sheet and the buckypaper are 0.89 and 39.72 mm, respectively. As seen in the curves, the position of the peak shifts to the right with the thickness of the sheets increase, but the peak height reduces. This peak gives us a chance to optimize the sheet size to get the biggest unit length's dissipation by convection.

CONCLUSION

In summary, we have developed a steady-state method to measure the convective heat transfer coefficient H of suspend CNT sheets. This method is noncontact and nondestructive to samples and may also be used to characterize the thermal properties of other thin films. Using this method, we find H of the single-layer CNT sheet is twice of aluminum foil in the same environment, and as the layers increase, the H drops quickly to the same with aluminum foil. We think the effective contact areas are the origin of these results. The single-layer CNT sheet are thin and loose, the internal gaps make for air flow inside, so the effective areas are enlarged. The analysis to the dissipation ratios by convection indicates that convection is the main dissipation approach for the CNT sheets on macro scales. There is a maximum convection ratio of unit length for different length sheets. The maximum for single-layer CNT sheet is located about 0.89 mm. These results may give us a new guideline to design the thermal devices based on the CNT sheets.

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by National Basic Research Program of China (2012CB932301) and the Natural Science Foundation of China (51173098, 10721404).

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