Raman Characterization of Heat Spreading in Carbon Nanotube Film

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Using materials with high thermal conductivity is a matter of great concern in the field of thermal management. Carbon nanotubes (CNTs) are in the spotlight as a good candidate for next generation heat spreading material due to their high thermal conductivity, flexibility in fabrication processes and abundance of raw carbon materials. The high thermal conductivity has been reported for the case of individual CNTs. Since CNTs would be primarily used in the form of networks, clarification of heat transfer chracterisitcs of CNT films is necessary. In addition, since there are several kinds of CNTs which are categorized according to the number of walls and synthesis method, identification of the most appropriate type of CNTs yielding the best heat spreading films would be very important in the field of thermal management. Here, we present our experimental results on two-dimensional thermal conductivity of CNT films. Based on the results, we discuss the possibility of CNT films as a heat spreader and applicable range of the optical method.

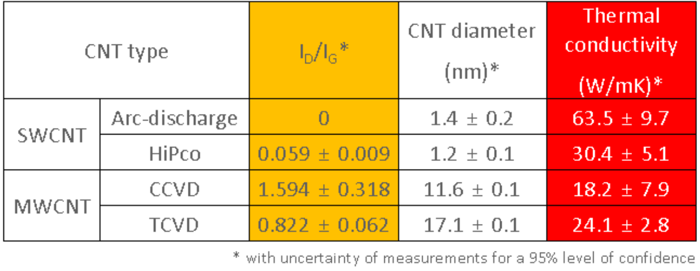
We made suspended CNT films and tested them by using an optical method based on Raman spectroscopy as shown in figure 1. We used four types of CNTs in the present work: single-walled carbon nanotubes (SWCNTs) synthesized by arc-discharge method or HiPCO process and multi-walled carbon nanotubes (MWCNTs) synthesized by catalytic chemical vapor deposition (CVD) or thermal CVD. It is well known that G-peak in Raman spectra is shifted due to the temperature change. The power of the laser used in the Raman spectroscopy is high enough to heat the CNT film. Hence, in this work, we used the G-peak shift and the laser as a temperature sensing probe and a way of heating, respectively, to determine the two-dimensional thermal conductivity of the CNT films. Figure 2 shows experimental results obtained in each step of the Raman characterization. From the energy balance between the heat from the laser and the conduction heat transfer, the film temperature captured by the Raman spectroscopy could be analytically obtained. By comparing the theoretical result with the experimental results, we could determine the thermal conductivity values.

The results are summarized in table 1. Arc-discharge SWCNT films show the best heat spreading capability. However, even the best thermal conductivity was lower than that of aluminum and copper which are widely used materials in the field of thermal management. Even though the as-prepared CNT films are not excellent heat spreaders, several post processes developed to improve the electrical conductance of the CNT films could be helpful in enhancing the thermal conductivity. For that issue, intensive research work would be required. Table 1 shows that the thermal conductivity depends on the crystallinity of the CNTs. We measured the thermal conductivity of CNT films with various transmittances as shown in table 2 and found that the effect of transmittance of films on the value of thermal conductivity is negligible when the light can penetrate the CNT film. However, when the CNT film is not transparent, the determined thermal conductivity seriously deviates from the thermal conductivity of transparent CNT films. In this study, whole cross-sectional area is assumed to be used as heat transfer path. However, when the film is not transparent, laser could not heat the whole cross-sectional area of the film and, due to the overestimated thickness of the film, the thermal conductivity is underestimated. Therefore, the present method is applicable only to transparent films.

In this study, we have reported two-dimensional thermal conductivity of CNT films and discussed applicability of the optical method used in this study. Although the heat spreading performance of the as-prepared CNT films is not satisfactory, there is still room for improvement. It would not be absurd to anticipate that carbon-based heat spreaders would replace metal spreaders in the future.

Figures

Table 1 Thermal conductivity of CNT films.

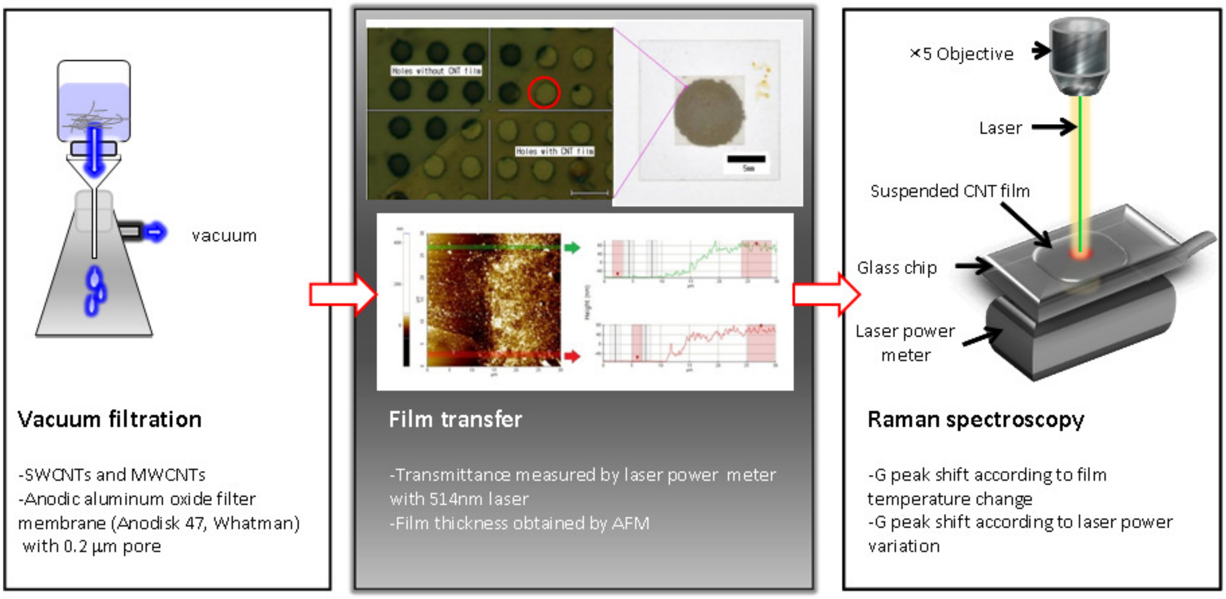


CNT type		I_D/I_G*	CNT diameter (nm)*	Thermal conductivity (W/mK)*
SWCNT	Arc-discharge	0	1.4 ± 0.2	63.5 ± 9.7
	HiPco	0.059 ± 0.009	1.2 ± 0.1	30.4 ± 5.1
MWCNT	CCVD	1.594 ± 0.318	11.6 ± 0.1	18.2 ± 7.9
	TCVD	0.822 ± 0.062	17.1 ± 0.1	24.1 ± 2.8

* with uncertainty of measurements for a 95% level of confidence

Table 2 Effect of transmittance on thermal conductivity.

CNT type	Transmittance (%)	Thermal conductivity (W/mK)
TCVD MWCNT	72.0	23.9
	62.0	22.5
	48.5	21.5
	34.0	24.2
	16.0	20.3
	12.5	24.5
	8.5	24.6
	0.0	0.203



Vacuum filtration

-SWCNTs and MWCNTs
-Anodic aluminum oxide filter membrane (Anodisk 47, Whatman) with 0.2 µm pore

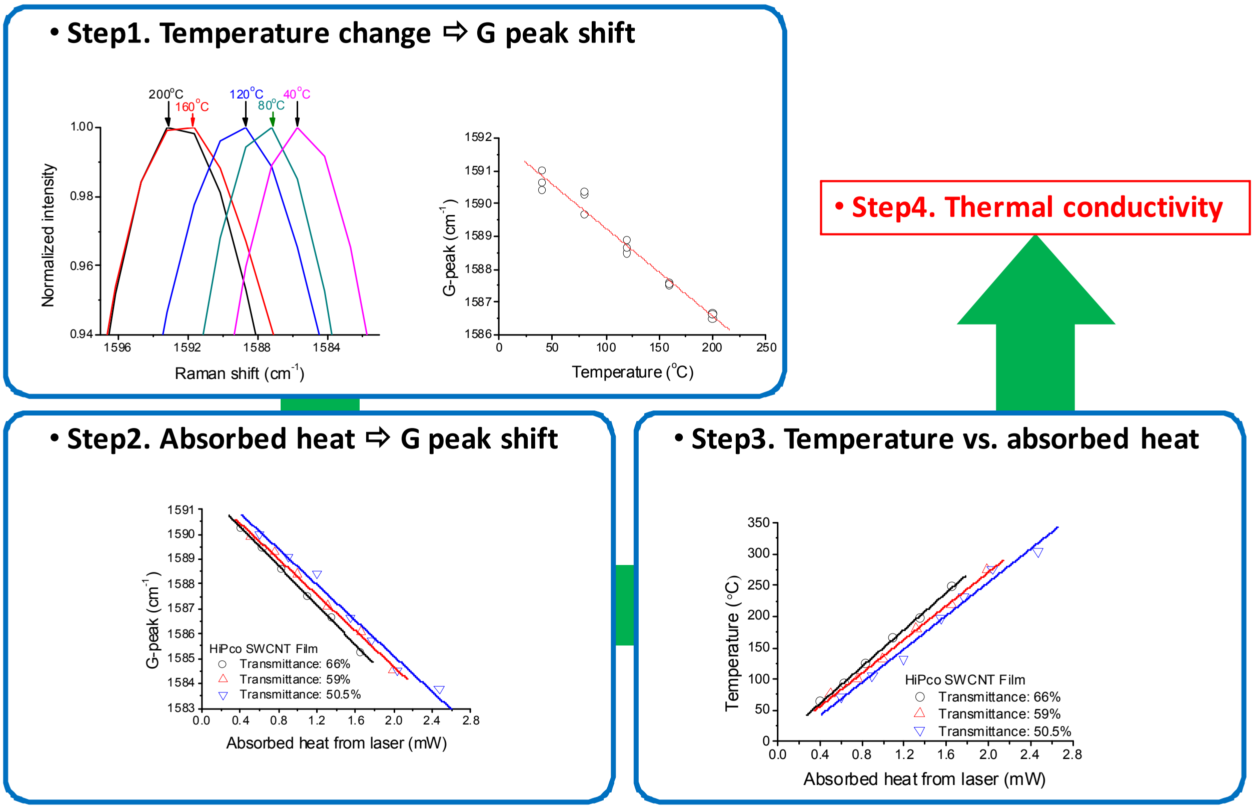
Film transfer

-Transmittance measured by laser power meter with 514nm laser
-Film thickness obtained by AFM

Raman spectroscopy

-G peak shift according to film temperature change
-G peak shift according to laser power variation

Figure 1 Experimental procedures.



- **Step1. Temperature change ⇨ G peak shift**
- **Step2. Absorbed heat ⇨ G peak shift**
- **Step3. Temperature vs. absorbed heat**
- **Step4. Thermal conductivity**

Figure 2 Experiemntal results obtained in each step of Raman characterization.