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Thermal interface materials with vertically-aligned carbon nanotubes and their thermal properties

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Abstract

We propose a new method to improve the density of carbon nanotubes (CNTs) by compression with silicone rubber and compare their thermal resistance with indium films, which are generally used as a thermal interface material (TIM). As a result, it was found that their thermal resistance corresponded to that of indium films with a temperature gradient method.

1. Introduction

Carbon nanotubes (CNTs) are one of the promising materials for future electronic devices due to their excellent physical properties such as high field effect mobility [1] and high thermal conductivity [2, 3]. Therefore, various applications with CNTs have been suggested, including transistors [1], interconnects [4], thermal bumps [5], and so on. Thermal interface materials (TIM) are used to connect thermally between a heat spreader (HS), and a processor tip is also one of them [6-11]. Although individual CNTs including single-walled CNTs and multi-walled CNTs have extremely high thermal conductivity around 3000 W/mK [2, 3], there are two issues to be addressed to realize CNT-TIM. They are poor CNT density and high thermal resistance at the interface.

As-grown vertically-aligned CNTs are utilized to be directly applied to CNT-TIM after growth on a substrate. Therefore, the density of as-grown CNT decides the number of a heat pass from a heat source to a heat sink, which means it has a large effect on the thermal conductivity of CNT-TIM. Generally speaking, a packing ratio of as-grown CNTs is relatively very low (around several percent) due to difficulties in control of CNT density. Recently, a method to densify CNTs mechanically after CNT growth was reported [10], although uniform densification seemed to be difficult. Furthermore, it is necessary to attach CNT-TIM to a HS and a processor tip with an adhesive layer such as a resin for practical use to maintain reliability of CNT-TIM. Such an adhesive layer, however, leads to high thermal resistance at the interface [10]. Here, we focus on the densification of CNTs.

In this paper, we propose a new method to densify CNT uniform by silicone rubber and compare thermal resistance of CNT-TIM fabricated from a densified CNT sheet with indium films, which are generally used as TIM.

2. Experimental

CNTs were synthesized with a thermal CVD method in a vacuum chamber. Iron and aluminum films were used as a catalyst deposited on a SiO_2/Si substrate with a conventional sputtering method. As the carbon source, a mixture of acetylene and argon gases was introduced into a CVD chamber in which the substrate was placed. Hydrogen was also added during the growth, and the total pressure was 8 kPa. The substrate temperature was approximately 640°C during the CVD process.

After synthesis, CNTs was compressed by the elasticity of the rubber to improve their density. Figure 1 (a-e) indicates a fabrication process of CNT densification. After synthesis of CNTs (a), a film composed of silicone rubber was attached to CNTs while it was kept pulled (b) and removed from the silicon substrate (c). After removal (d), the film with CNTs was snapped back when pulling stopped (e), which led to densification of CNTs. The photograph of the compressed CNT sheet on the film after densification with the 3-inch silicon substrate used to grow CNTs is shown in figure 1(f). One can see clearly that the compression of CNTs was successful. Assuming that the number of CNTs was the same after compression, this implied they were packed more densely. Here, the density of CNTs was estimated to increase three–fold compared with before compression.

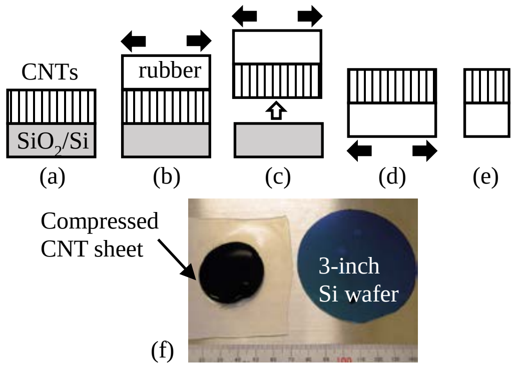

Fig. 1 (a-e) Compression process of CNT sheet. (f) Photograph of CNT sheet after compression and removal from a SiO_2/Si wafer.

Thermal resistance of CNT-TIM was measured by a temperature gradient method. This measurement system was composed of CNT-TIM sandwiched with an adhesive layer by two copper blocks, which were a heat sink and a heat source. The differences (ΔT) in temperature of two copper blocks were measured. Their values correspond to the thermal resistance of CNT-TIM including the resistance at the interface.

3. Results

Figure 2 shows the SEM images after CNTs growth. Vertically aligned CNT bundles on a substrate can be clearly observed. The height of CNTs was around 140 μm. Analyzing the SEM images, the packing ratio and diameter of CNTs was estimated to be approximately 2-3 % and 8 nm. In addition, TEM images showed the CNTs to be multi-walled CNTs (not shown).

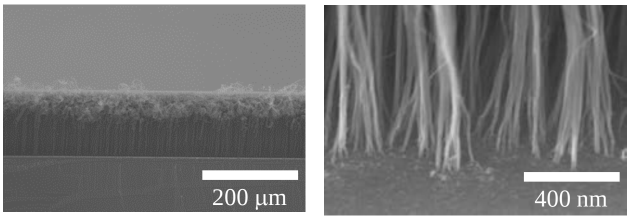

Fig. 2 SEM images of vertically-aligned CNTs bundle synthesized on a SiO_2/Si substrate with a thermal CVD method at 640°C.

To densify CNTs, the compression process was performed as shown in figure 1. The density of CNTs was estimated to increase three–fold compared with before compression. Figure 3 indicates SEM images before (a) and after (b) compression. One can see that the density after compression increased compared with before compression. Furthermore, Raman spectra of CNTs were observed before (c) and after (d) compression shown in figure 1. Raman spectrum of as-grown CNTs indicates multi-walled CNTs growth [12]. After compression, the Raman spectrum was found to be unchanged, which suggested no damage by the compression process.

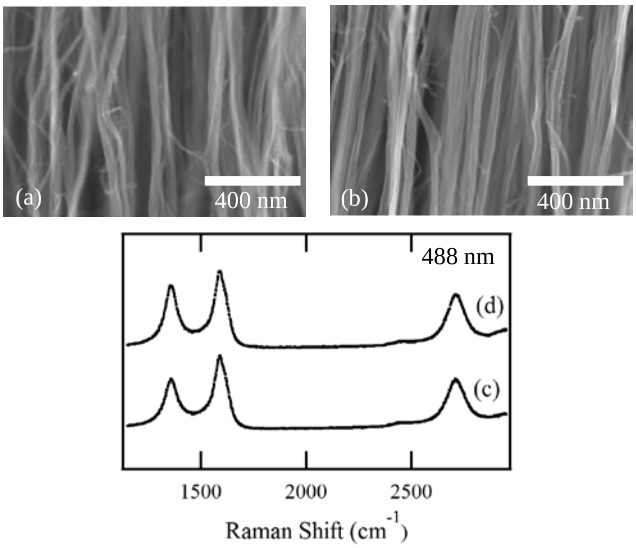

Fig. 3 SEM images and Raman spectra of vertically-aligned CNT bundle before (a and c) and after (b and d) compression.

After densification of CNTs, the differences (ΔT) in temperatures were measured. As a result, ΔT of CNT-TIM was estimated to be 1.06°C, which was comparable to that of indium films (ΔT=1.04°C). This result indicated that the thermal resistance of CNT-TIM including interface resistance between CNT-TIM and metal blocks on both sides was as low as that of indium films. These results help to steadily advance the realization of CNT-TIM application in place of indium TIM. On the other hand, the thermal resistance of CNT-TIM was still high when taking into account the packing ratio of CNTs and individual CNT thermal conductivity. This might originate from the thermal resistance at the interface between CNTs and two metal blocks, which will be our next issue.

4. Conclusions

We proposed a new method to improve the density of carbon nanotubes (CNTs) by compression. After compression, CNTs were observed to be densified three-fold compared with before compression with no damage. Furthermore, the thermal resistance of CNT-TIM was found to be as low as that of indium films generally used as a thermal inter-face material (TIM).

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