

Thermoelectric characteristics of carbon nanotube / polydimethylsiloxane composites fabricated by solvent-less printing

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Abstract

Organic (including carbon nanotubes) thermoelectric materials generally have thin-film shapes. However, to add enough temperature difference to thermoelectric devices, thick thermoelectric materials are indispensable. In this presentation, we report solvent-less printing as a fabrication method of thick organic thermoelectric materials. Millimeter order of thick carbon nanotube/polymer composites with good thermoelectric characteristics were easily formed by this method.

1. Introduction

Organic thermoelectric materials (including carbon nanotube based materials) have been extensively researched due to their mechanical flexibility, light-weight nature, and printing processability [1-11]. In most papers related to organic-based thermoelectrics, materials had thin film-shape and thermoelectric characteristics were mainly investigated in the in-plane direction of the thin film. However, when thermoelectric devices are placed on a heat source, temperature difference is generated along the out-of-plane direction in regards to the device surface. Thus, thermoelectric materials having enough thickness to add temperature difference are required.

Formation of thick materials by printing is generally difficult because drying the solvent causes compression of material-solved solution in out-of-plane direction. As a result, thickness of printed organic materials become several order of thinner than that of before drying printed solution. Thus, developing a printing method which do not include drying the solvent may be an effective way to form thick organic thermoelectric materials by printing.

In this presentation, we report thermoelectric characteristics of organic materials fabricated by solvent-less printing. We used composites consisting of carbon nanotube (CNT) and polydimethylsiloxane (PDMS) as organic materials because CNT/polymer composites have high thermoelectric performance and mechanical flexibility. We used CNT dispersed liquid precursor of PDMS as an ink for printing. This ink become solidified by cross-linking reaction of PDMS induced by catalyst and heat, thus, vaporization of organic solvent do not include in the solidification process. Using this

solvent-less printing process, we succeeded in fabrication of millimeter order of thick CNT/PDMS composites with high thermoelectric performance.

2. Experimental

Figure 1 shows schematic illustration of solvent-less printing of CNT/PDMS composites. We used mixed solution of low molecular weight ($M_w \sim 550$) PDMS and trimethoxymethylsilane (TMMS) as a liquid precursor of cross-linked PDMS. The weight ratio of PDMS and TMMS were 1:3. Single-walled CNTs were dispersed in the liquid precursor by using planetary ball milling. Then a catalyst (dibutyltin dilaurate) was added to the CNT dispersed liquid precursor. Then the CNT/liquid precursor/catalyst solution was printed on a substrate using thick (millimeter order) printing plate. Finally, the printed mixture became solidified through cross-linking reaction of PDMS by heating. We measured thermoelectric characteristics in out-of-plane direction of the solidified CNT/PDMS composites.

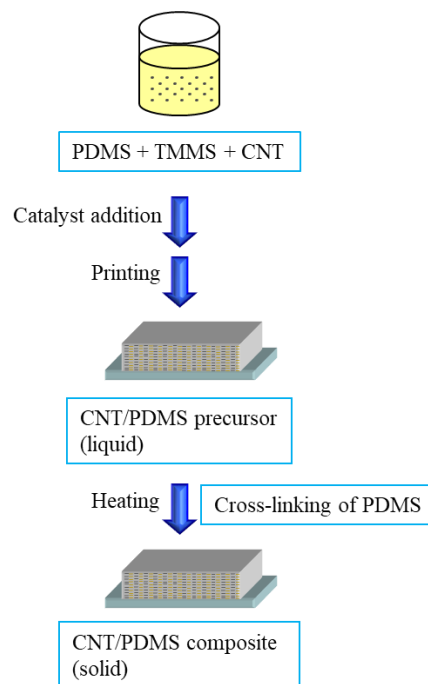


Fig. 1 Procedure of solvent-less printing of CNT/PDMS composites.

3. Results and discussion

Figure 2 shows a photograph of a CNT/PDMS composite fabricated in this study. In our solvent-less printing process, compression of the film thickness owing to drying the solvent do not occur because this process do not include drying the solvent. Thus, solidified CNT/PDMS composites kept almost same thickness as before solidified CNT/PDMS composites. As a result, millimeter order of thick materials are easily obtained by using solvent-less printing (Fig. 2).

Figure 3 shows thermoelectric power factor of CNT/PDMS composites as a function of the concentration of CNTs. The power factor increased with increase in the CNT concentration, and showed peak at approximately 19%. In low CNT concentration region, increase in CNT cause enhancement in the CNT conducting network, which leads increase in electrical conductivity and power factor. On the other hand, in high CNT concentration region, a lot of cracks in the materials caused by lack of binder polymer were observed. This may be the origin of decrease in the power factor in the region over the CNT concentration of 19 wt%.

The maximum power factor in this study was $7 \mu\text{W}/\text{K}^2\text{m}$, which is approximately one order of magnitude higher power factor in out of plane direction of CNT/polymer composites fabricated by ordinarily printing process [12].



Fig. 2 Photograph of solvent-less printed CNT/PDMS pellets.

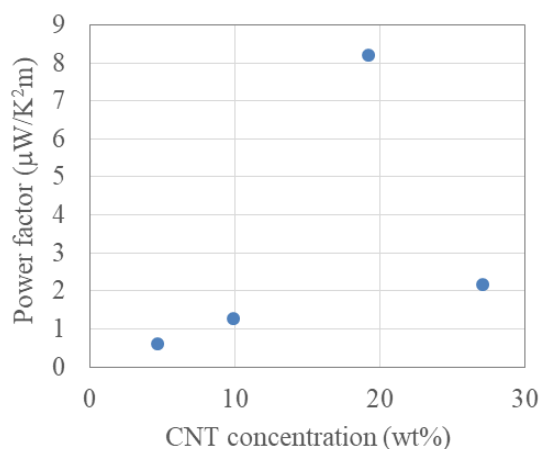


Fig. 3 Power factor as a function of CNT concentration of CNT/PDMS composites.

4. Conclusions

To add enough temperature difference to thermoelectric devices, thick thermoelectric materials are indispensable. We applied solvent-less printing of CNT/polymer composite as a fabrication method of thick organic thermoelectric materials. Millimeter order of thick CNT/polymer composites with good thermoelectric characteristics were easily formed by this method.

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