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Thermal Characteristics of Multi-walled Carbon Nanotube and Acrylate Co-polymer Composites

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Abstract

We report the enhancement of positive temperature coefficient (PTC) intensity by improving multi-walled carbon nanotube (MWCNT) dispersion in acrylate co-polymers. Due to insufficient dispersion by conventional magnetic stirrer, PTC intensity of these polymer composites had been limited to 2.5×10^2. By ultrasonic dispersion, we improved MWCNT dispersion in acrylate polymers very well, which leaded to PTC intensity 12-times increase and reached 2.9×10^3.

1. Introduction

Measuring temperature is important for homoiothermic animals, because sickness or an inflammation are appeared as a temperature change. Flexible temperature sensor is promising approach to measure the skin temperature accurately, which leads to health monitoring [1][2]. Especially, polymer-based positive temperature coefficient (PTC) temperature sensor, which is using reduction of conductive pass by thermal expansion of polymers, has gathered considerable attention due to its flexibility and high sensitivity [3].

Previously, we have reported graphite/acrylate copolymer-based PTC temperature sensor [2]. However, this temperature sensor also showed negative temperature coefficient (NTC) effect. In contrast, carbon nanotube (CNT) is known to suppress NTC effect due to its high aspect ratio structure [4]. Also, CNT shows high electrical and thermal conductivity, and a low percolation threshold. One of the biggest problems of CNT composite sensor is poor dispersion due to aggregation, which leads to limit the characteristic of temperature sensor.

In this work, multi-walled CNT (MWCNT)-filled acrylate polymer composites were prepared and their temperature-resistivity characteristic was evaluated. We compared two different dispersion methods (magnetic stirrer and sonication) to investigate CNT-dispersion in acrylate polymer. The electrical resistance was measured as a function of MWCNT concentration for each dispersion method. The surface of MWCNT/polymer composites was observed. The temperature characteristic was also investigated.

2. Experimental

The printed temperature sensor was fabricated as reported previously [2]. The acrylate polymer is composed of two monomers, octadecyl acrylate and butyl acrylate at weight ratio of 17:3. The sharp increase in resistivity is observed around human body temperature by using this weight ratio. The polymer was mixed with MWCNT using a magnetic stirrer for 16 h or a sonicator for 1 h. Temperature sensors were fabricated by printing copolymer with fillers on interdigitate gold electrodes with 400-μm pitch through a 12.5-μm polyimide mask on a hot plate at 60 °C. The temperature sensors were annealed at 100 °C in an oven for 1 h.

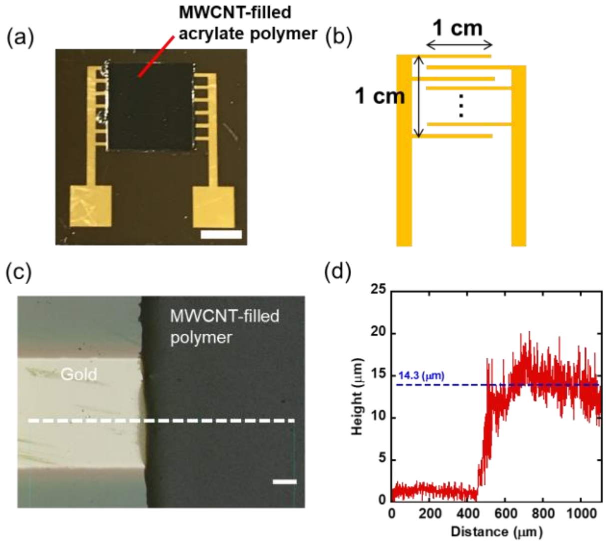

Fig. 1 Printed temperature sensor (8 wt% MWCNT-filled polymer by sonication process). (a) The picture of the temperature sensor. The scale bar corresponds to 5 mm. (b) Structure of interdigit electrodes. W/L= 1 cm/ 400 μm. (c) Optical image of the device. The scale bar corresponds to 100 μm. (d) Height profile of the temperature sensor corresponding to the dashed white line of panel (c).

3. Results and discussion

Figure. 1 shows an image of printed temperature sensor. The MWCNT/acrylate polymer composites were printed on the interdigitate electrodes as shown in Fig. 1 (a) and (b). The polymer was observed by laser microscope (Fig. 1 (c)) and the thickness of the polymer composites was 14.3 μm as shown in Fig. 1 (d).

First, we investigated the electrical characteristics of temperature sensor with several MWCNT concentrations. As shown in Fig. 2, the electrical characteristic was decreasing as the MWCNT concentration increased. Also, fig. 2 indicated the resistance of the sensor dispersed by sonication (blue) was higher compared to that dispersed by magnetic

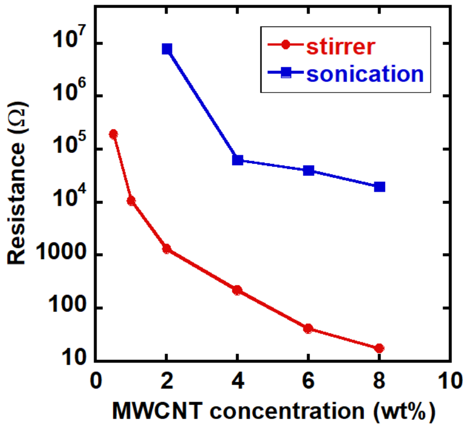

Fig. 2 The relation between electrical resistance and MWCNT content for the MWCNT/acrylate polymer composites fabricated by stirrer or sonication, respectively.

stirrer (red). In case of polymer composites mixed by a stirrer for a long time, the electrical resistance was dramatically decreasing as filler content increases, and it showed 0.22 kΩ at 4 wt% MWCNT concentration. However, in the case of polymer composites fabricated by a sonication, the electrical resistance was gradually decreasing and it showed 63 kΩ at 4 wt% MWCNT concentration. This increase in electrical resistance of polymer composites is due to the damage by the sonication process. MWCNT fillers are cut by the sonication process and the aspect ratio of the fillers is decreasing, which causes the conductive pass lower.

Figure. 3 shows the SEM images of the surfaces of the polymer composites. Compared to two SEM images, the surface of polymer composites fabricated by stirrer possessed larger mass of conductive fillers than that fabricated by sonication. These large masses indicate that MWCNT dispersion in polymers by stirrer is less sufficient than that by sonication.

Next, we investigated the effect of temperature on the electrical resistance. The temperature sensor was placed on a hotplate and the hotplate temperature was changing in the range from 30 °C to 40 °C. Figure. 4 shows the effect of temperature on the electrical resistance about various conditions; fabricated by stirrer and by sonication, and different MWCNT concentration. Fabricated by the stirrer process, the temperature sensor showed 2.5×10^2 PTC intensity at 4 wt% MWCNT concentration. In contrast, the temperature sensor using polymer composites fabricated by sonication showed 2.9×10^3 PTC intensity, which is 12 times higher than that by stirrer. Using sonication is enhancing MWCNT dispersion in acrylate polymer and it causes high PTC intensity in spite of the sacrifice of conductivity in room temperature.

4. Conclusions

In this work, we investigated the MWCNT dispersion in acrylate polymer using different method; stirrer and sonication, in order to fabricate the temperature sensor which has high PTC intensity. The temperature sensors composed of

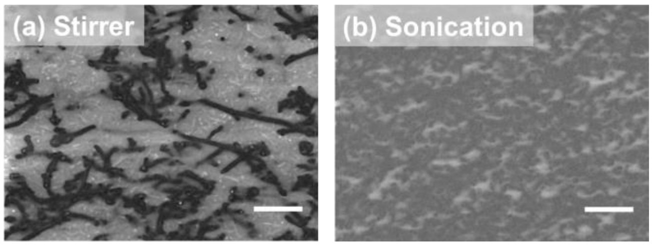

Fig. 3 SEM image of the surface of the printed 4 wt% MWCNT-filled copolymers fabricated by (a) stirrer and (b) sonication. The scale bar corresponds to 10 µm.

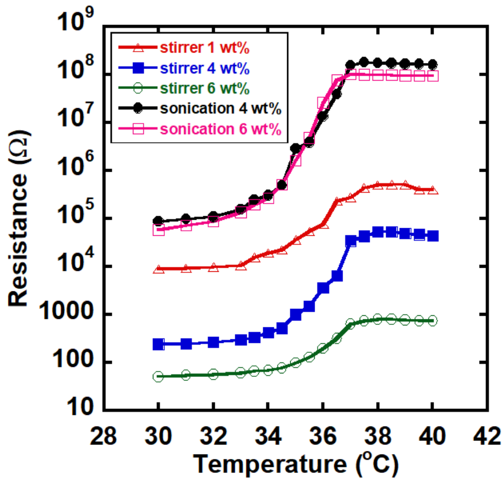

Fig. 4 The effect of temperature on the electrical resistance of various polymer composites; 1, 4, or 6 wt% MWCNT-filled polymer composites fabricated by stirrer and 4 or 6 wt% MWCNT-filled polymer composites by sonication.

MWCNT-filled acrylate polymer were fabricated on a flexible polyimide substrate by printing process and the thickness of polymer composites was 14.3 µm. Polymer composites dispersed by stirrer showed lower electrical resistance than those by sonication at the same MWCNT concentration, but the dispersion in polymers is insufficient, which caused lower PTC intensity. At 4 wt% MWCNT concentration the PTC intensity by sonication process reached 2.9×10^3, which was 12 times higher than that by stirrer process.

Acknowledgements

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References

[1] R. C. Webb *et al.*, Nat. Mater. **12** (2013) 1078.
[2] T. Yokota *et al.*, Proc. Natl. Acad. Sci. **112** (2015) 14533.
[3] H. P. Xu *et al.*, J. Appl. Phys. **107** (2010) 034105.
[4] X. J. He *et al.*, Appl. Phys. Lett. **86** (2005) 1.