

ELECTRIC FIELD-INDUCED ALIGNMENT OF THERMAL CONDUCTIVE FILLER IN ACRYLIC POLYMER FOR ENHANCED THERMAL CONDUCTIVITY

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Abstract

A recent trend in the integration of electric power devices has caused problems of heat management because of high heat generation from the device. Therefore, studies of new materials having high thermal conductivity have been motivated. Polymer-based composites have been paid attention because of its flexibility and energy-saving productivity. In this study, nitride fillers, which have excellent thermal conductivity and electric insulating property, were used as a component of the polymer-based composites. To achieve high thermal diffusion with a small fraction of the fillers, the fillers were aligned by an electric field. Aluminum nitride (AlN) or boron nitride (BN) particles were dispersed in an acrylic polymer. An array of wire electrodes was used to form widely spread electric field. In-plane and through-plane alignment were carried out with varying arrangements of the electrode arrays. Thermal diffusivity in-plane direction of the AlN and BN composites increased about 1.2- and 1.7-fold by aligning the fillers as comparing with those without the alignment, respectively. To align the filler in through-plane direction, several kinds of the electrode arrangements were examined. As the best result, the through-plane thermal diffusivity of the aligned AlN composites became 30-fold compared to that without the alignment.

1. Introduction

In recent years, miniaturization and integration of electric power devices have progressed with the popularization of electric vehicles. Consequently, heat dissipation from the device has been one of the crucial issues. In general, a metal heatsink contacting directly to the heat source of the device is used for better heat dissipation. However, it is difficult to avoid forming air gaps which are low thermal conductivity, at the contact interface because of the roughness of the surface of the heatsink and the heat source. To fill the air gaps, thermal interface materials (TIM) are used. There are several types of TIM. For example, grease type which is mainly used for heat dissipation of small area such as central processing unit (CPU) of computers, and sheet type which is used for covering a large area such as engine control unit (ECU) of electric vehicles. The sheet type is better than the grease type regarding productivity and usability. TIM is also required having high adhesiveness and flexibility. Therefore, polymer-based composites which include high thermal conductive fillers, have been recognized as one of the promising TIM for the integrated electric power devices because of its processability.

The thermal conductivity of acrylic resin and epoxy resin, is as low as 0.2 W/mK⁽¹⁾. For making high heat conductive composites, fillers with high thermal conductivity, such as carbon materials⁽²⁾ (carbon fibers, carbon nanotubes), metal oxides⁽³⁾ (tin oxide, aluminum oxide), nitrides^(4,5) (aluminum nitride, boron nitride), and metals⁽⁶⁾ (aluminum, copper) have been added. Since nitride fillers have high thermal

conductivity and electric insulation properties, composites including nitride fillers have received much attention. For example, the composites using particles of aluminum nitride (AlN) and boron nitride (BN) represented 14 W/mK (AlN, 67 vol%)⁽⁴⁾, and ~33 W/mK (BN, 78.5 vol%)⁽⁵⁾, respectively. Although high filler content in the composite results in high thermal conductivity, it increases the cost and decreases its flexibility. To overcome the drawback, alignment of fillers in the composite has been studied to achieve high thermal conductivity with a small fraction of fillers⁽⁶⁾. Several kinds of the alignment methods such as mechanical⁽⁷⁾, magnetically⁽⁸⁾ and electrical⁽⁹⁾ methods have been reported.

In this study, an array of wire electrodes was used to form widely spread electric field. Aluminum nitride (AlN) or boron nitride (BN) particles were added as the filler in an acrylic polymer. Because the array of electrodes is covered with a thin dielectric film, the composite does not contact with the array resulting in that smooth surface of the produced composite⁽¹⁰⁾. Alignment of the fillers in-plane and through-plane direction were studied. For the in-plane alignment, the composite was placed on the array electrode. In the case of alignment in through-plane direction, the composite was sandwiched between two array electrodes. The produced composites were evaluated by measuring the thermal diffusivity.

2. Material and Methods

2.1. Array electrode

Schematic illustration and photo of the array electrode are shown in Fig.1. Grooves with 1 mm in width and 1.2 mm in depth at an interval of 1 mm were fabricated on polymethylmethacrylate (PMMA) substrate. Copper wires (diameter:1 mm, length:15 cm) were installed into the grooves, and the surface was sealed with an epoxy resin. The epoxy resin was cured by ultraviolet (UV) light of 365 nm.

2.2. Nitride fillers, matrix and composite

AlN and BN particles, both were 5 μm in diameter, were used as fillers. A matrix was prepared by mixing a high crosslinking binder and a low viscosity acrylate at a volume ratio of 3:7 and adding photoinitiator at the volume ratio of 1:10 with the matrix for curing in ultraviolet. Each of particles was added to the matrix at 15 wt% and stirred by vortexing to make AlN composite and BN composite.

2.3. Electric field-induced alignment in-plane direction

Mixed AlN composite or BN composite was uniformly spread on polyethylene terephthalate (PET) film (38 μm in thickness), which was placed upon the array electrode. For alignment of in-plane direction, the wire electrodes in the array were alternately connected to the high-voltage and the ground. Then, the AC voltage (4 kV and 4 kHz) was applied for 10 min. During application of the electric field, the PET film oscillated in the electric field direction by a slide mechanism (distance: 4.0 mm, speed: 0.4 mm/s) to spatially average the electric field affecting in the composite⁽⁸⁾. After the electric field application, the composite was cured by irradiation of UV of 365 nm. The composites without the voltage application were also prepared.

2.4. Electric field-induced alignment through-plane direction

To apply the electric field in through-plane direction, two electrodes were arranged as shown in Fig.2. Three arrangements, (a) plate vs. plate, (b) array vs. plate, and (c) array vs. array were studied by combining a plate electrode and the array electrode. In the case of (c), the lower electrode and the upper electrode were placed so that they face each other and cross at right angles to form the grid. This is because the high electric field is generated at the intersection of the electrodes. The distance between the upper and lower electrodes was adjusted to 3 mm.

AlN composite was uniformly spread on the PET film, which was placed on the lower electrode. AC voltage (5 kV and 4 kHz) was applied to the lower electrode for 10 min, and the upper electrode was grounded. After applying the AC voltage, the composite was cured by UV light.

2.4. Evaluate the thermal conductive property of the composites

To evaluate the thermal conductive property of the composites, thermal diffusivity was measured by a laser flash method using a xenon flash analyzer (1.06 μm , LHA447, Netzsch). A square film, 10 mm each side, was cut out from the produced composites. Thermal diffusivity of the film in through-plane direction was determined well-defined method as described in previous studies^(11,12). The in-plane method was used to measure the thermal diffusivity in-plane direction⁽¹³⁾. In the in-plane method, the temperature detection position was different from the position of the laser irradiation. At the detection point, the heat transferred from the laser irradiation spot was detected. Then, the in-plane thermal diffusivity was calculated by the time lag between the laser spot and the detection point. The thermal

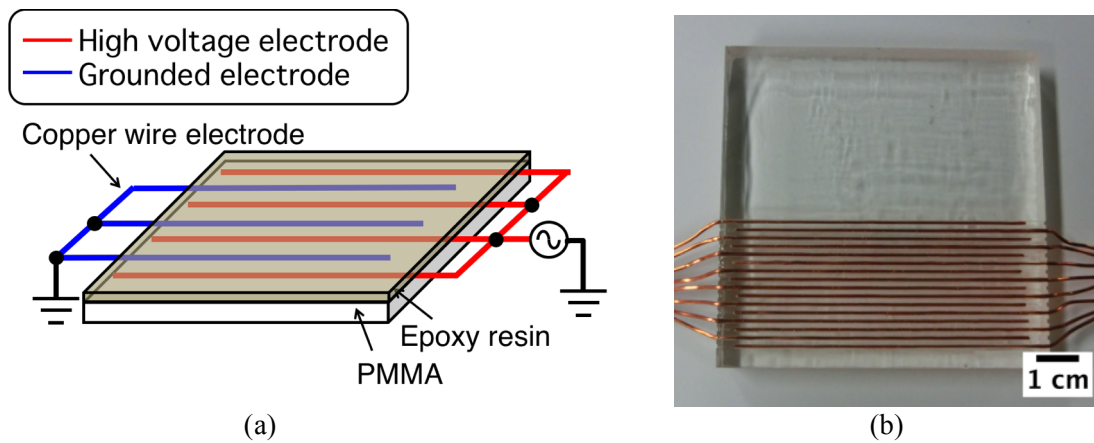


Fig.1. Schematic illustration (a), and photo (b) of the array electrode.

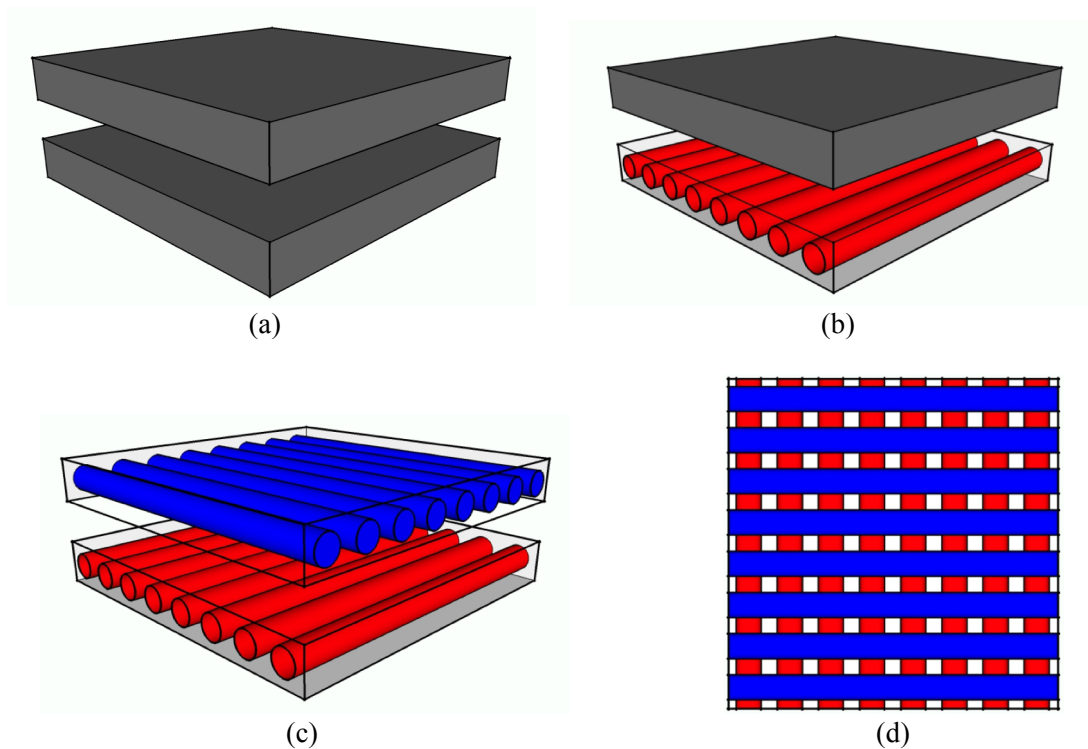


Fig.2. Schematic illustrations of electrode arrangement. (a) Plate vs. plate, (b) array vs. plate, (c) array vs. array, and (d) top view of array vs. array.

diffusivity measurement was carried out three times for the same sample, and the average of values was defined as the thermal diffusivity of the composites.

3. Results and discussion

3.1. In-plane thermal diffusivity of the electrical aligned composite

The in-plane thermal diffusivities of the composites are shown in Table 1. The thermal diffusivity of AlN composite with and without electric field was 1.6 mm²/s and 1.3 mm²/s, respectively. In the case of BN composite, the thermal diffusivity with and without electric field was 2.2 mm²/s and 1.3 mm²/s, respectively. The in-plane thermal diffusivity improved by about 1.2-fold in AlN composite and 1.7-fold in BN composite due to electric field-induced alignment. In this work, we used pure AlN and BN. By surface treatment of fillers, thermal diffusivity of composite would enhance more⁽¹⁴⁾.

Fig.3 shows microscopic images of BN composite before and after the application of the voltage. By applying the voltage, BN particles formed structures as pearl chains and were oriented in the electric field direction. This implies that the electrical alignment of the nitride fillers, AlN and BN, in the composite increased the thermal diffusivity.

3.2. Through-plane thermal diffusivity of the electrical aligned composite

Fig.4 shows the thermal diffusivity in the through-plane direction of AlN composite. Without voltage application, the thermal diffusivity was 0.22 mm²/s. When the voltage applied in each of the electrode arrangement, the thermal diffusivity became (a) 0.20 mm²/s, (b) 0.50 mm²/s, (c) 7.65 mm²/s. In (c), the through-plane thermal diffusivity increased significantly, about 30-fold. However, the value of each

Table 1. In-plane direction thermal diffusivity of AlN particle composite and BN particle composite

| Filler | Thermal diffusivity [mm ² /s] | |
|--------|------------------------------------------|--------------------------|
| | Without voltage application | With voltage application |
| AlN | 1.3 | 1.6 |
| BN | 1.3 | 2.2 |

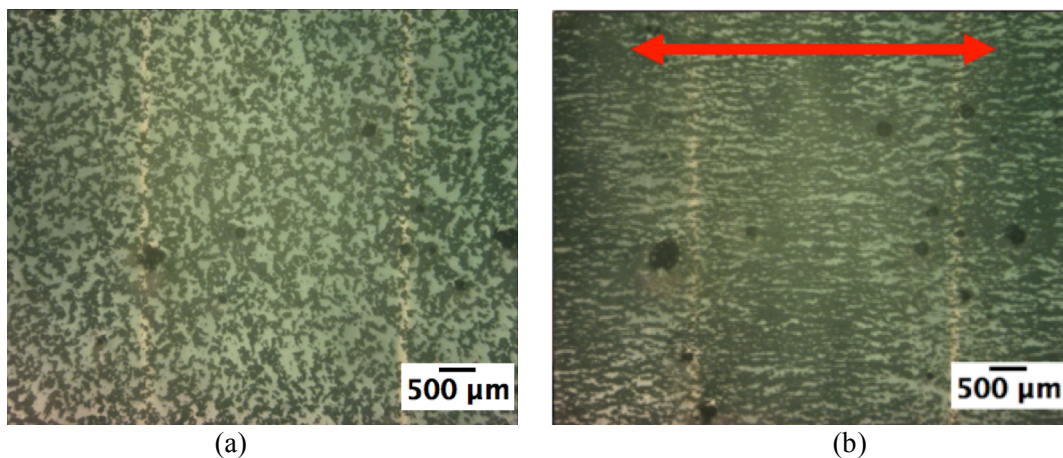


Fig.3. Microscope images of BN particle composite. Before applying voltage (a), and after applying voltage for 10 min (b). (Red arrow indicates direction of the electric field.)

thermal diffusivity measurements varied largely (0.68, 0.59, 0.66, 9.79, 19.6, and 14.6 mm²/s). Because the electric field gradient induced by the electrode array was not as uniform as the plate electrode, the region, where the fillers formed the pearl chain orientation, was not uniform in the whole composite. Therefore, the region containing the aligned fillers and the region without the aligned filler would exist all over the composite, which made the variation of thermal diffusivity large. To verify morphology of composite, we will investigate cross-sectional images of composite by scanning electron microscopy(SEM) and X-ray computed tomography(CT).

4. Conclusion

To manufacture high thermal conductive polymer-based composites with a small fraction of the fillers, nitride fillers, AlN and BN of 5 μm in diameter, were aligned in the acrylic polymer matrix by applying the electric field in-plane or through-plane direction. The in-plane thermal diffusivity increased 1.2-fold and 1.7-fold for AlN and BN composite as comparing with no-electric field one, respectively. The through-plane thermal diffusivity increased about 30-fold by using vertically arranged two array electrodes. Electric-induced alignment is effective for the improvement of both in-plane and through-plane direction thermal conductive property of polymer-based composites.

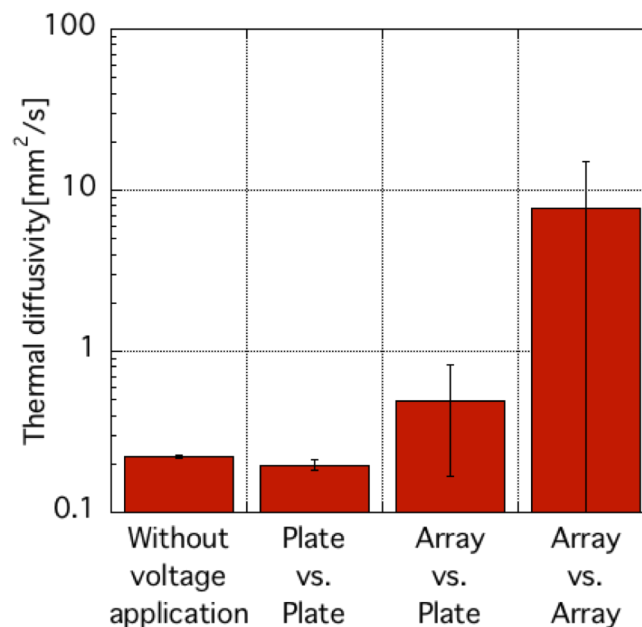


Fig.4. Through-plane thermal diffusivity of the composite obtained with electrode arrangement.

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