Thermal Conductivity and Processability of Polymer Composites with Nano- and Micro-fusible Fillers

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Abstract
This study aims to develop polymer composites with high thermal conductivity and good processability using nano- and micro-fusible fillers. Polymer composites were prepared by mixing liquid-type epoxy resin with a curing agent, an accelerator, and nano- and micro-fusible fillers. The nano-fusible fillers were a mixture of fusible-powdered epoxy resin with low fusible temperature and carbon nanotubes (CNTs), whereas the micro-fusible fillers were a mixture of fusible-powdered epoxy resin with high fusible temperature and carbon fibers. The nano- and micro-fusible fillers fused during the cure of epoxy matrix and form thermally conductive pathways containing CNTs and carbon fibers in the composites. The viscosity of the uncured composites was measured using a viscometer. Moreover, the thermal conductivity of the cured composites was evaluated using a steady-state method. The effect of addition of the nano- and micro-fusible fillers on the thermal conductivity and viscosity of the composites is discussed.

1. Introduction

Most polymers have very low thermal conductivity. One approach to improving thermal conductivity of polymers is through the addition of micro-conductive fillers, such as ceramics, carbons and metals. Polymer composites with high thermal conductivity offer new possibilities for replacing metal parts in electric systems. The advantages of polymer composites as compared with those made from metals include improved corrosion resistance, lighter weight, and the ability to adapt conductivity properties to suit the application requirements. To use polymer composites as heat sinks in electric systems, new composites must be prepared with a thermal conductivity of 1‑30 W/mK [1]. Although high thermal conductivity can be achieved by including high volume fractions of micro-conductive fillers, this tends to increase the viscosity of the mixture, which must be sufficiently low to be processed into parts in electric systems.

An approach of current interest to improve the thermal conductivity of polymer is the selective addition of nano-conductive fillers. Carbon nanotubes (CNTs) have exceptionally high aspect ratio and high thermal conductivity [2,3]. When CNTs are dispersed in polymer, an interconnecting network is formed which provides a pathway for heat conduction. However, due to their nanoscale dimension and very high aspect ratio, processing of polymer composites with high volume fractions of CNTs is very difficult.

To achieve the high thermal conductivity by the addition of low volume fractions of conductive fillers, synergy effects of nano- and micro-conductive fillers has been addressed by many authors. Sanada et al. [4] fabricated the thermally conductive polymer composites with CNTs and the random close-packed structure of alumina micro particles, and investigated the potential of CNTs to enhance thermal

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conductivity of the composites. The objective of this work is to develop polymer composites with high thermal conductivity and good processability using nano- and micro-fusible fillers. Nano- and micro-fusible fillers contain CNTs and carbon fibers, respectively, and fuse during cure of the polymer matrix and form thermally conductive pathways containing CNTs and carbon fibers in the composite as shown in Figure 1. The polymer composites with nano- and micro-fusible fillers were prepared and experimental measurements of the thermal conductivity and the viscosity of the manufactured composites were carried out.

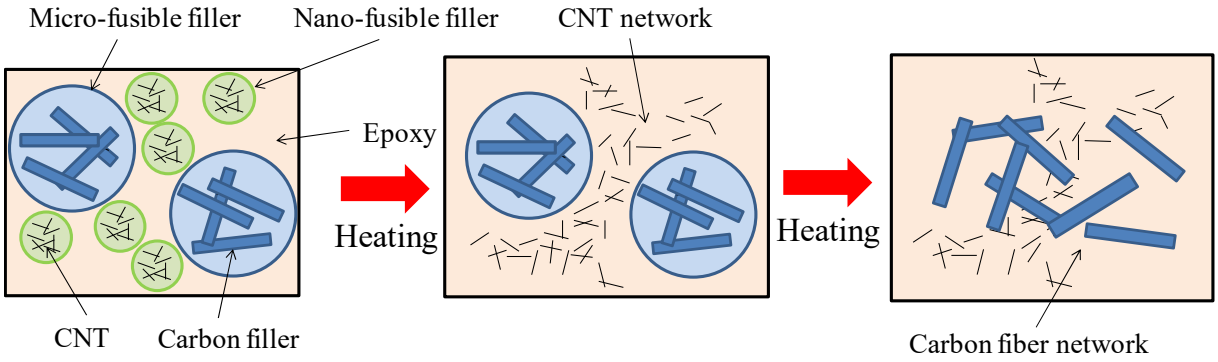

Figure 1. Thermal network structure of nano- and micro-fusible fillers

2. Experimental procedure

2.1. Materials and specimen preparation

Nano- and micro-fusible filler/epoxy composites were prepared by mixing a Epikote 828 liquid-type epoxy resin from Mitsubishi Chemical Co. with a HN-2000 curing agent from Hitachi Chemical Co., Ltd., a BMI12 accelerator from Mitsubishi Chemical Co., and nano- and micro-fusible fillers. The mix ratio of the Epikote 828, the HN-2000 and the BMI12 was 100:80:2 by weight. The nano-fusible fillers were a mixture of YSLV-80XY fusible-powdered epoxy resin from Nippon Steel & Sumikin Chemical Co., Ltd. with fusible temperature of 80°C and VGCF multi-walled carbon nanotubes (CNTs) with diameter of 150nm and length of 8μm from Showa Denko K. K., whereas the micro-fusible fillers were a mixture of YSLV-120TE fusible-powdered epoxy resin with fusible temperature of 120°C and R-A301 carbon fibers with diameter of 8μm and length of 200μm from Teijin Ltd. The CNTs and carbon fibers were used as-received. Nano-fusible fillers had a diameter of 100-250μm and micro-fusible fillers had a diameter of 425-1000μm.

The CNT volume fraction of the composite V_f^{CNT} and the carbon fiber volume fraction of the composite V_f^{C} are given by

$$V_f^{CNT} = \frac{V^{CNT}}{V_T^m} \tag{1}$$

$$V_f^{C} = \frac{V^{C}}{V_T^m} \tag{2}$$

where V^{CNT} and V^{C} are the volume of the CNT and the volume of the carbon fiber, respectively. The volume of the epoxy of the composite V_T^m is given by

$$V_T^m = V_1^m + V_2^m + V_3^m \tag{3}$$

where V_1^m, V_2^m and V_3^m are the volume of the epoxy of the nano- and micro-fusible fillers/epoxy composite, the volume of the epoxy of the nano-fusible filler and the volume of the epoxy of the

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micro-fusible filler, respectively. The CNT volume fraction of the nano-fusible filler V_f^N and the carbon fiber volume fraction of the micro-fusible filler V_f^M are given by

$$V_f^N = \frac{V^{CNT}}{V_2^m + V^{CNT}} \tag{4}$$

$$V_f^M = \frac{V^C}{V_3^m + V^C} \tag{5}$$

The volume fraction of the nano-fusible filler of the composite V_f^{NF} and the volume fraction of the micro-fusible filler of the composite V_f^{MF} are given by

$$V_f^{NF} = \frac{V^{CNT} + V_2^m}{V_T^m + V^{CNT} + V^C} \tag{6}$$

$$V_f^{MF} = \frac{V^C + V_3^m}{V_T^m + V^{CNT} + V^C} \tag{7}$$

The epoxy mixture was heated for 3min at 90°C and stirred with an ARE-310 planetary mixer for 30s at 500rpm to fuse nano-fusible fillers. Furthermore, the epoxy mixture was heated for 3min at 130°C and stirred with the planetary mixer for 30s at 500rpm to fuse micro-fusible fillers. After heating and stirring, the epoxy mixture was degassed, poured into a mold and cured for 5h at 150°C. For comparison, CNT/carbon fiber/mixed epoxy composites were also prepared by mixing the Epikote 828 with the HN-2000, the BMI12, the YSLV-80XY, the YSLV-120TE, the CNTs and the carbon fibers. The epoxy mixture was then degassed, poured into a mold and cured for 2h at 100°C, followed by 4h at 150°C. The cured sample were composite disks of 50mm in diameter and 10-15mm in thickness.

2.2. Test methods

Nano- and micro-fusible fillers in the epoxy mixture during heating were observed using a VH5500 digital microscope. Spreading ratio of the nano- and micro-fusible fillers α is defined by following equation

$$\alpha = \sqrt{\frac{S'}{S}} \tag{8}$$

where S is the projected area of the fusible filler before fusing and S' is the projected area of the fusible filler after fusing. Viscosity of the uncured samples was measured using a DV+II Pro viscometer (Brookfield Engineering Laboratories, Inc.) and a SV-100 viscometer (A&D Co., Ltd.). Thermal conductivity of the cured sample was also measured using a HC-110 thermal conductivity tester (Eiko Instruments Co., Ltd.).

3. Results and discussion

Figure 2 shows the micrographs of nano-fusible fillers before and after fusing at 60°C. Although the nano-fusible filler with V_f^N =0.1vol% fused widely, the shape of the nano-fusible filler with V_f^N =0.3vol% did not change.

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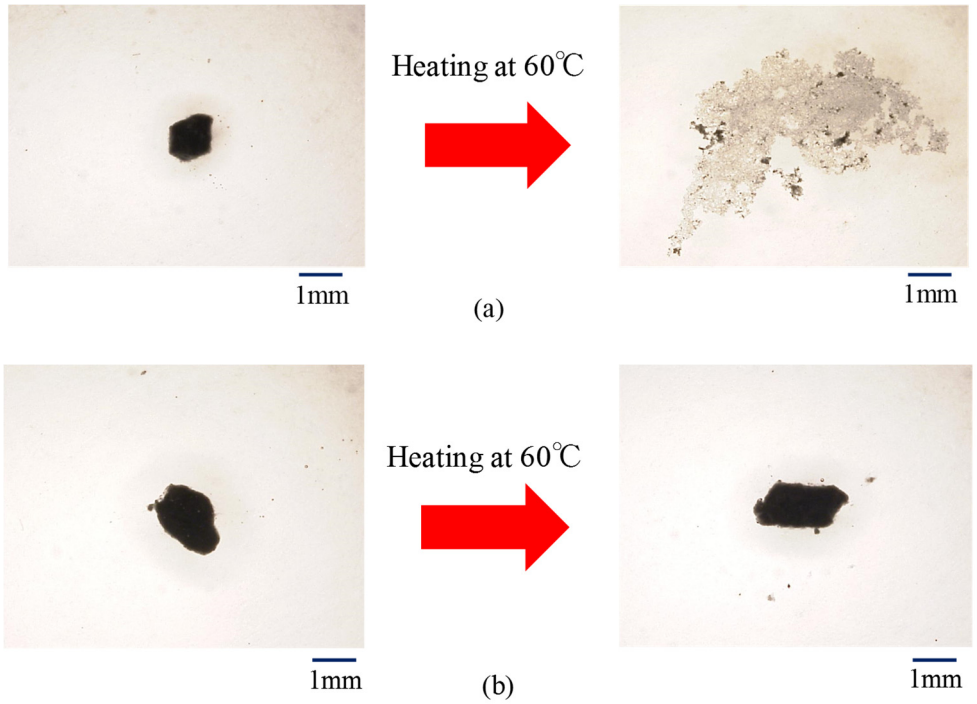

Figure 2. Micrographs of nano-fusible fillers with various V_f^N before and after fusing at 60°C: (a) V_f^N =0.1vol%; (b) V_f^N =0.3vol%.

Figure 3 shows the effect of V_f^N on the spreading ratio of the nano-fusible filler α. The spreading ratio of the nano-fusible filler decreased with increasing V_f^N from 0.05 to 0.3vol%. Above V_f^N =0.3vol%, the spread ratio of the nano-fusible filler converged to a value of α =1.

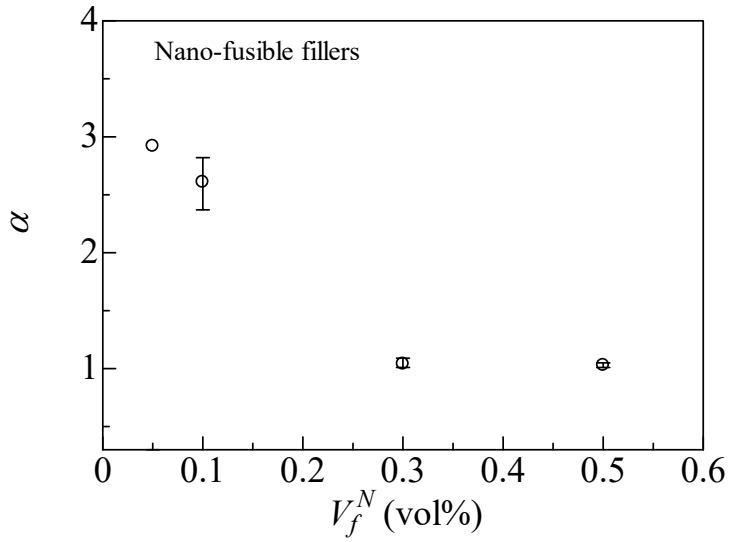

Figure 3. Spreading ratio versus CNT volume fraction of nano-fusible fillers.

Figure 4 shows the micrographs of micro-fusible fillers before and after fusing at 120°C. The shape of the micro-fusible filler with V_f^M =10 and 30vol% did not change.

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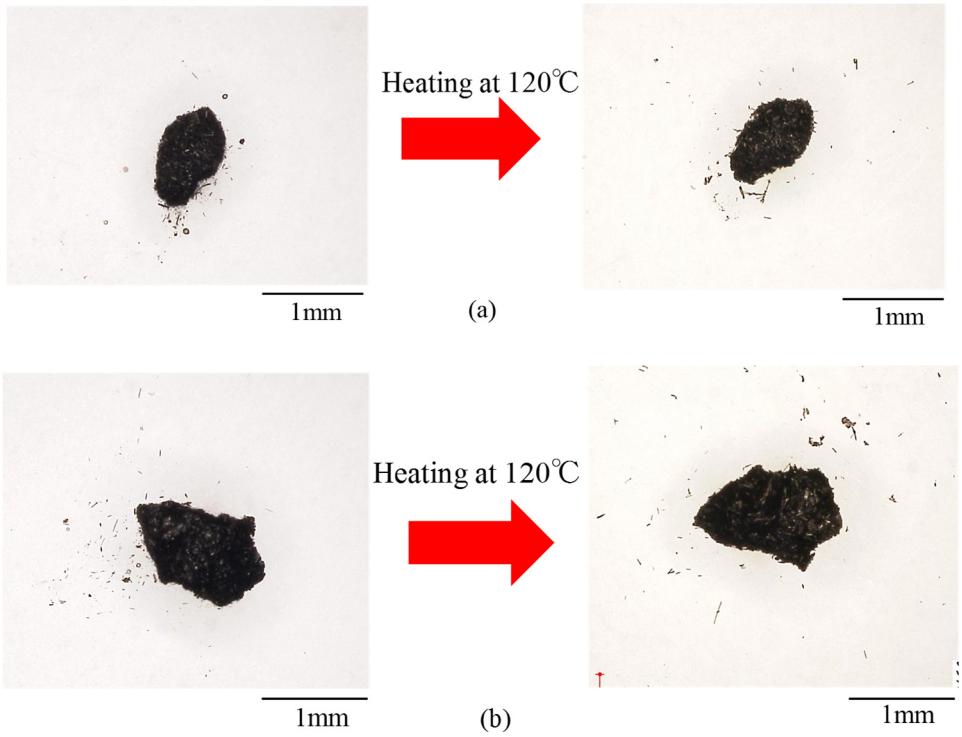

Figure 4. Micrographs of micro-fusible fillers with various V_f^M before and after fusing at 120°C: (a) V_f^M =10vol%; (b) V_f^M =30vol%.

Figure 5 shows the effect of V_f^M on the spreading ratio of the micro-fusible filler. The spreading ratio of the micro-fusible filler was constant with increasing V_f^M from 10 to 50vol%. Therefore, the mixing process was required to spread the nano- and micro-fusible fillers widely.

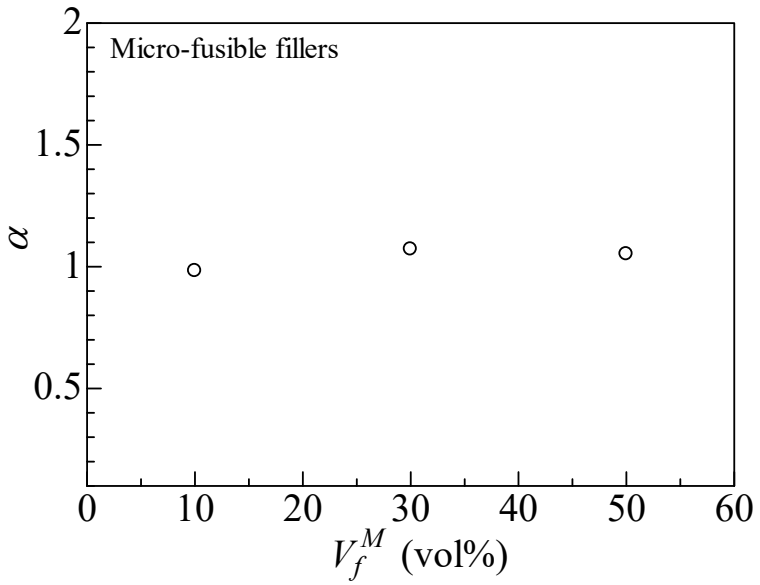

Figure 5. Spreading ratio versus carbon fiber volume fraction of micro-fusible fillers.

The measured thermal conductivity of the nano- and micro-fusible fillers/epoxy composites and CNT/carbon fiber/mixed epoxy composites with V_f^C =20vol% and V_f^{CNT} =3vol% are presented in Figure

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6. The mixed epoxy is composed of the Epikote 828, the HN-2000, the BMI12, the YSLV-80XY and the YSLV-120TE. For the same viscosity, the thermal conductivity of nano- and micro-fusible fillers/epoxy composites was higher than that of CNT/carbon fiber/mixed epoxy composites. Nano- and micro-fusible fillers facilitated the formation of thermally conducting pathways of CNTs and carbon fibers through the composite to result in increased thermal conductivity.

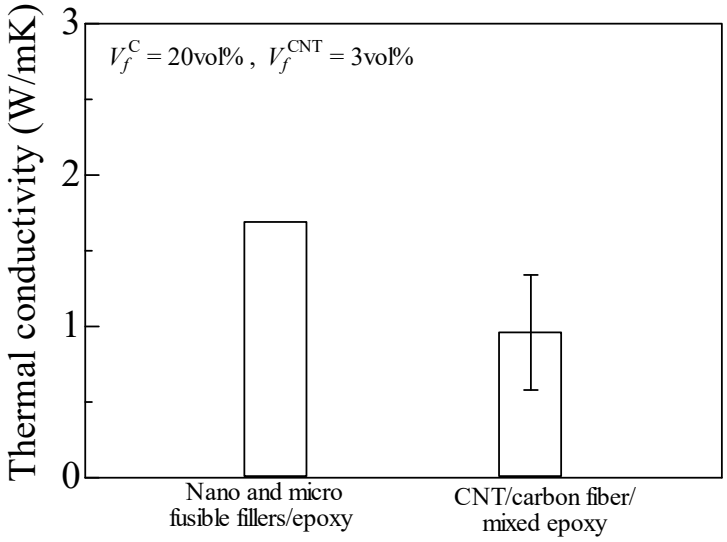

Figure 6. Thermal conductivities of the nano- and micro-fusible fillers/epoxy and CNT/carbon fiber/mixed epoxy composites.

3. Conclusions

The composites with the nano- and micro-fusible fillers were fabricated and the potential of nano- and micro-fusible fillers to increase the thermal conductivity and decrease the viscosity of the composites was investigated. The results demonstrated that high therrmal conductivity and good processability can be achieved by using the nano- and micro-fusible fillers.

Acknowledgments

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