



Synergistic enhancement of thermal conductivity in polymer composites filled with self-hybrid expanded graphite fillers



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ABSTRACT

The size of expanded graphite (EG) was controlled using a high-speed crusher because a hybrid of differently sized fillers can induce a synergistic enhancement of the thermal conductivity in polymer composites. We found that the thermal conductivity of a polymer composite filled with both 10 wt% EG and 10 wt% high-speed crusher treated EG (wEG) was synergistically improved by 12.0 and 20.7% compared to that of polymer composites filled with 20 wt% EG and 20 wt% wEG alone, respectively. A three-dimensional (3D) non-destructive analysis using X-ray micro-computed tomography (micro-CT) was applied to explain the synergistic enhancement and to identify the dispersion and 3D network of EG fillers in the composites accurately. According to the non-destructive analysis results, the synergistic enhancement was caused by the formation of efficient thermally conductive pathways due to the hybrid of the differently sized EG and wEG fillers.

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1. Introduction

Electronic devices have become increasingly integrated into ever smaller spaces. It is therefore becoming more critical to dissipate the heat generated efficiently from the insides of devices, as failure adequately to discharge residual heat to the outside environment can cause heat damage or even a fire [1–3]. Particularly, when applying heat-dissipating structures to light-emitting diodes and electronic device housing units, it is necessary to develop polymer composites with high thermal conductivity; and which also have relatively simple processing conditions and a good degree of product flexibility [4]. These advanced thermally conductive polymer composites are typically fabricated by mixing thermally conductive fillers such as metals, ceramics and carbons into a polymer resin [5].

Recently, nano-carbon fillers such as graphene and carbon nanotubes are receiving more attention due to their excellent thermal conductivity between 1950 and 7000 W/m·K [6–11]. However, when a polymer matrix is mixed with nano-carbon fillers, incomplete contact can be produced at the interface between the nano-filler and the polymer matrix, or between the nano-fillers themselves [6,12]. As a result, it has been reported that this incomplete contact can produce interfacial thermal resistance and thermal contact resistance, resulting in phonon scattering and low thermal conductivity near the lower bound of the rule of mixture [6,12–14]. In addition, the high price of nano-carbon fillers compared to other types of thermal conductive fillers is an

obstacle to the commercial use of thermally conductive polymer composites containing nano-carbon fillers.

Expanded graphite (EG) is a carbon material with the unique structure of graphite sheets with interlayer spacings. When filled within polymers, it can become distributed inside the composite in an efficient manner, forming a thermally conductive network. Noh and Kim [15] evaluated the thermal conductivity of thermally conductive polymer composites filled with various carbon fillers such as EG, pitch-based carbon fiber, graphite, graphene nanoplatelet, multi-walled carbon nanotube and carbon black. They reported that the polymer composite filled with EG fillers showed the best performance in terms of thermal conductivity [15]. In addition, given that EG is one of the most cost-effective carbon fillers, it is highly advantageous to use from a commercialization perspective.

It is known that the electrical conductivity of polymer composites rapidly increases at a specific range of filler content, according to percolation theory [16,17]. However, the thermal conductivity of composites filled with EG fillers show a monotonous increase with respect to the filler loading amount [18,19]. Yung and Liem [20] reported that the introduction of hybrid fillers consisting of different particle sizes contributed to the synergistic enhancement of the thermal conductivity of thermally conductive polymer composites and that this synergistic effect was related to the internal structure of the composites. Accordingly, to induce the synergistic enhancement of the thermal conductivity of a composite filled with EG fillers, it can be an effective approach to mix differently sized EG fillers into a polymer matrix.

In this study, inexpensive commercial EG was treated by a high-speed crusher, which is an advantageous method in terms of the

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Table 1
Sample composition

Sample	EG (wt%)	wEG (wt%)	PC (wt%)
E5	5	0	95
E10	10	0	90
E15	15	0	85
E20	20	0	80
w5	0	5	95
w10	0	10	90
w15	0	15	85
w20	0	20	80
E10w10	10	10	80

processing time and cost, to control the size of the EG. The resulting self-hybrid composites filled with different sizes of the same fillers (raw EG and wEG) were fabricated using a typical and cost-effective melt mixing method. Their thermal conductivity values were measured in an effort to investigate the possible synergistic enhancement of the thermal conductivity by the self-hybrid. In addition, a non-destructive X-ray micro-CT method was used to observe the internal structure of the composite to analyze the synergistic effect and to optimize the thermal conductivity of the composites.

2. Experimental

2.1. Materials

EG powder was provided by Hana Chemtech (ES 250 B5D, Hana Chemtech, Seoul, Korea). The EG has a unique structure consisting of graphite sheets with interlayer spacings which effectively enhances the thermal conductivity of composites filled with EG filler. The EG had a carbon content of 95% and an expansion coefficient of $250 \text{ cm}^3/\text{g}$. Average size of the EG was larger than $300 \mu\text{m}$. To control the size of the EG, a high-speed crusher (WB-1, Osaka Chemical Co. Ltd., Osaka, Japan) was used to treat it at a speed of 25,000 rpm for 30 s at room temperature. The crusher crushes samples instantly from hard to soft, such as ceramics, cereals, chips of wood, clothes, crude drugs, medicinal herbs and minerals, based on its super-high-speed rotation and strong motor power of 700 W. A linear polycarbonate (PC) resin (LUPOY PC 1300-03, LG Chemistry Co., Gyeonggi-do, Korea) which was designed for extrusion or injection, was used as the matrix.

The Vicat softening point was 151° , as measured under conditions of $50^\circ/\text{hr}$ and a 50 N load according to ASTM D 696. The density of the resin was 1200 kg/m^3 , as measured according to ASTM D 792. The melt flow rate was $3 \text{ g}/10 \text{ min}$, as measured according to ASTM D 1238.

2.2. Composite fabrication

The prepared EG and/or wEG and PC resin samples were weighed to the target contents as shown in Table 1. They were put into a HAAKE Rheomix internal Mixer (HAAKE™ Rheomix OS Lab Mixers, Thermo Scientific Inc., Marietta, GA, USA) heated to 260°C and mixed at a constant screw speed of 60 rpm for 30 min, and then pelletized. The fabricated pellets were put into a mold ($2.5 \text{ cm}^2 \times 2 \text{ mm}$) for the thermal conductivity tests. The pellets were pressed using a heating press (D3P-30J, Daheung Science, Incheon, Korea) at a pressure of 15 MPa under a temperature of 260°C for 15 min and were then quenched to 30°C using cold water.

2.3. Characterization

2.3.1. EG filler

For an analysis of the surface functional groups of the EG fillers, Fourier transform infrared spectroscopy (FT-IR, Nicolet 6700, Thermo Scientific, MA, USA) and X-ray photoelectron spectroscopy (XPS, K-Alpha, Thermo Scientific, Massachusetts, USA) were used. The FT-IR spectra were measured in the range of $1000\text{--}4000 \text{ cm}^{-1}$ at a resolution of 16 cm^{-1} . The XPS spectra were obtained with an Al X-ray source at a pressure of $1 \times 10^{-8} \text{ Pa}$. To evaluate the defect level of the EG fillers, Raman spectroscopy (LabRAM HR 800, HORIBA Jobin Yvon, Japan) was used along with a 514 nm Ar ion laser.

2.3.2. Morphology

The fabricated composites were fractured using liquid nitrogen to obtain the samples. The EG, wEG and composite samples were coated with platinum for 120 s under a vacuum using a sputter-coating machine (Ion Sputter E-1030, Hitachi High Technologies, Tokyo, Japan). In this case, 10 kV of electricity was applied to the coated samples under a nitrogen vacuum using a field emission scanning microscope (FE-SEM, Nova NanoSEM 450, FEI Corp., OR, USA).

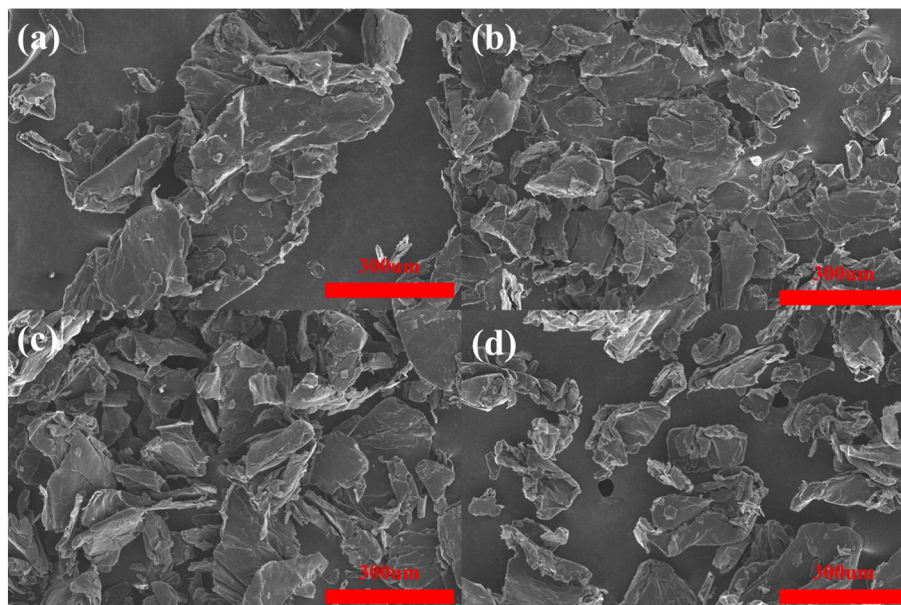


Fig. 1. SEM images of EG fillers with respect to treatment time of the high-speed crusher. (a) Raw EG, (b) 10 s, (c) 20 s, and (d) 30 s.

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