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Synergistic enhancement of thermal conductivity in composites filled with expanded graphite and multi-walled carbon nanotube fillers via melt-compounding based on polymerizable low-viscosity oligomer matrix

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A B S T R A C T

We found that the thermal conductivity of the polycarbonate (PC) composite filled with both 9.9 wt% expanded graphite (EG) and 0.1 wt% multi-walled carbon nanotube (MWCNT) fillers was synergistically improved by 49% compared to that of the PC composite filled with 10 wt% EG alone. In order to induce the optimal internal structure favorable for thermal conduction by enhancing the dispersion of the second MWCNT fillers, we applied a two-step melt-compounding to fabricate composites using a MWCNT masterbatch based on a polymerizable oligomer resin, cyclic butylene terephthalate (CBT), which is characterized by ultra-low viscosity and excellent impregnability during initial melting. Three-dimensional (3D) non-destructive characterization using X-ray micro computed tomography (micro-CT) was utilized to demonstrate the synergistic enhancement and to verify dispersion and 3D thermal network of the fillers in the composites accurately. The synergistic enhancement was significantly affected by the formation of the efficient thermally conductive pathways and dispersion of the second MWCNT fillers.

1. Introduction

As the integration of electronic appliances and electronic circuit boards poses the increased risk of mechanical malfunctions and fires of them, increasing attention is being paid to lightweight, highly-formable polymer composites with high heat dissipation [1,2]. Conventionally, thermally conductive polymer composites are fabricated by filling polymer resins with high-thermal conductivity fillers (e.g., carbon, ceramic or metal) [3–5]. Thermally conductive polymer composites filled with ceramic or metal fillers can provide stable heat dissipation performance, but with the recent trend for weight savings, there has been a growing interest in thermally conductive polymer composites filled with carbon fillers. In a recent study, Noh et al. [6] compared the thermal conductivity of polymer composites filled with various carbon-based fillers such as carbon black, MWCNTs, graphene nanoplatelets, graphite, pitch-based carbon fibers and EG, and reported that EG fillers were the most effective in enhancing the thermal conductivity of composites.

EG is relatively cheap compared to other carbon-based fillers and forms the most effective 3D thermally-conductive network within polymer composites due to the unique structure consisting of graphite sheets with interlayer spacing [6]. Unlike the percolation theory that the electrical conductivity of composites rapidly increases at a certain level of filler content [7], there is a report that the thermal conductivity of composites is monotonously improved with an increasing EG filler content [6]. Incorporation of the second fillers with a large aspect ratio can make a synergistic enhancement in the thermal conductivity of EG filled composites by optimizing the internal filler structure. Yu et al. [8] and Pak et al. [9] reported that incorporating a small amount of carbon nanotubes (CNTs) with a high aspect ratio induced a synergistic enhancement in the thermal conductivity of composites by forming an effective 3D thermally conductive network.

Many studies [10–13] have been conducted on the development of nanocarbon-based polymer composites with high

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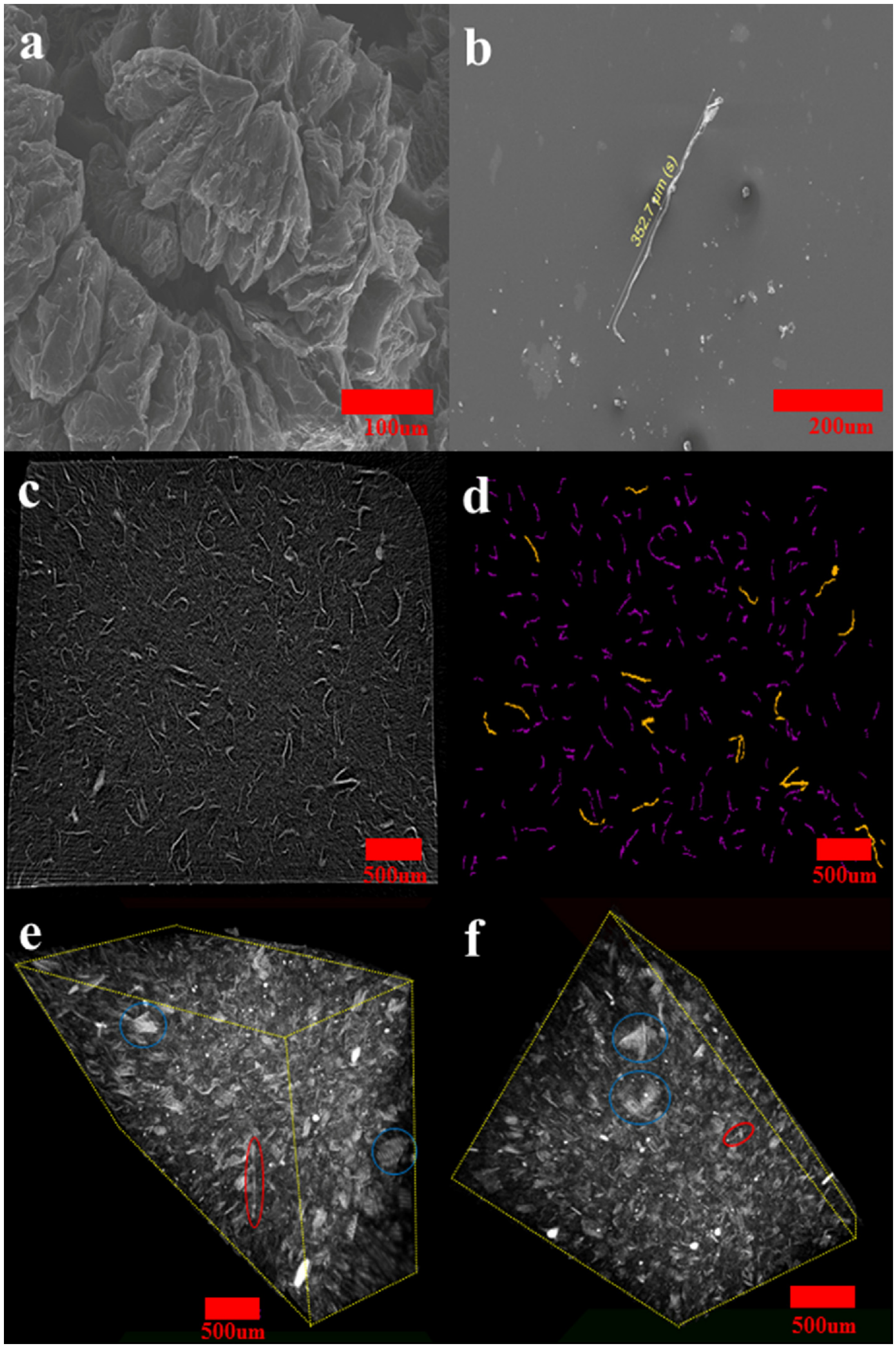

Fig. 1. SEM images of (a) EG and (b) MWCNT, and micro-CT (c) 2D, (d) reconstruction, 3D (e) side-view and (f) top-view images of composites filled with both EG 9.9 wt% and MWCNT 0.1 wt%.

thermal conductivity and heat dissipation because CNTs and graphene turned out to have a thermal conductivity of 3000–6000 and 4000–7000 W/m·K, respectively. To our disappointment, however, the experimentally reported thermal conductivity values of nanocarbon filled polymer composites mostly remain near the lower bound given by the rule of mixtures [14]. The low values of thermal conductivity are related to the interfacial thermal resistance between the nanocarbons and polymer matrix [9,14]. To be specific, the reason for low thermal conductivity is that phonons acting as heat transfer media are scattered at the interface between the nanocarbon filler and the polymer matrix, which is maximized by the introduction of nano-sized fillers. However, the nano-sized materials with a large aspect ratio are useful as the second fillers for the effective formation of 3D thermally conductive networks in polymer composites. Furthermore, the dispersion of the second fillers is known as an important parameter in determining effectiveness of the 3D thermally conductive networks [15].

In this study, a synergistic enhancement in thermal conductivity of polymer composites was discussed in relation to the incorporation of MWCNTs as the second fillers which were expected to exert a synergistic effect in optimizing the thermal