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Effects of graphene concentration, relative density and cellular morphology on the thermal conductivity of polycarbonate–graphene nanocomposite foams

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ABSTRACT

The thermal conductivity of polycarbonate–graphene nanocomposite foams was studied as a function of relative density, developed cellular structure and graphene concentration. Two types of supercritical CO_2 foaming processes were employed to obtain foams with a wide range of relative densities and cellular morphologies. The thermal conductivity of unfoamed nanocomposites increased in more than two times upon addition of 5 wt% graphene. Foaming led to lowered thermal conductivity values, as low as 0.03 W/(m K), with thermal conductivity being mainly controlled by relative density and in a lower extent by graphene concentration. Higher thermal conductivities were obtained with increasing relative density and cell size, as well as with increasing graphene concentration, especially in those cases where improved graphene dispersion was achieved with foaming. Thermal conductivity values displayed a better fit when using a three-phase model when compared to the two-phase model previously proposed for polymer composite foams.

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1. Introduction

Polymers are well known for being materials that are intrinsically thermally and electrically insulating [1,2]. As foaming further enhances the insulating behavior of polymers by means of incorporating an important void fraction, the study of the heat transfer of polymer foams has become one of their most important fields of research, backed up by the vast number of applications and uses that these materials offer as thermal insulators [3,4]. However, many applications could benefit from the use of polymers with enhanced thermal conductivity, as for instance when used as heat sinks in electric or electronic systems [5] and in electrical wires for heat dissipation [6]. Besides the use of conductive polymers, which are expensive and commonly display a relatively low thermal stability and poor mechanical performance, the thermal conductivity of insulating polymers has been traditionally enhanced by the addition of thermally conductive fillers, including graphite, carbon black, carbon fibres, ceramic or metal particles [7–9].

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Previous studies have shown that the thermal conductivity enhancement due to the addition of conductive nanofillers differs depending on the type of nanofiller and polymer matrix [9–12]. Therefore the selection of the type of filler is crucial. It has been stated that significant scatter of data are typically reported for thermal conductivity of fillers. This is caused by several factors including filler purity, crystallinity, particle size and measurement method [9]. Among conductive nanofillers, graphene nanoplatelets have been recently considered to enhance the thermal conductivity of polymers [13,14]. Despite the intrinsically high thermal conductivity of graphene nanoplatelets, the thermal conductivity enhancement in polymers has been shown to be quite limited, even at high graphene volume fractions [15].

Most of the studies published regarding the heat conduction of polymer composites with carbon-based nanoparticles and particularly graphene nanoplatelets have considered the use of theoretical models [15,16], as it has been the case for unfilled polymer foams [17–20]. Efforts to enhance the thermal conductivity of polymers, especially thermoset-based, by means of adding conductive carbon nanoparticles have been reported [21–28]. It has been shown that the addition of graphite or graphene-like nanoplatelets to epoxy results in linear increases in thermal conductivity with increasing nanoparticles concentration, reaching enhancements of four times the thermal conductivity of unfilled epoxy with adding 5 wt% graphene or as high as twenty times at 40 wt% graphene loading [21]. Similarly, the formation of strong interfacial interactions between matrix and nanoparticle, for instance by means of silane-crosslinking, has been shown to enhance thermal conductivity [23–28].

Scarce experimental work has been dedicated to the study of the thermal conductivity of polymer foams containing carbon-based nanoparticles. In one of our works we studied the thermal conductivity behavior of polypropylene–carbon nanofibres (PP–CNF) composite foams [29]. It was found that carbon nanofibres were not efficient in increasing the thermal conductivity of PP-based foams, its value resulting almost constant and independent of CNF's volume fraction, which was related to the possible partial rupture, poor dispersion and aggregation of CNFs during processing, limiting the possibility of intimate contact between the nanofibres, crucial to attain significant thermal conductivity improvements [30]. Similarly, Shen et al. [31] demonstrated that for PS–CNF nanocomposite foams the addition of increasingly higher concentrations of CNFs only slightly enhanced the thermal conductivity, which was related to the presence of insulating polymer between the conductive nanofibres, limiting thermal conduction. In the same way, Zheng and co-workers [32,33] have recently demonstrated that the thermal conductivity of microcellular polyetherimide-graphene nanocomposite foams tends to decrease gradually with reducing cell size, reaching values as low as 0.036 W/(m K) even for GnP contents as high as 7 wt %, typical of thermally insulating polymer foams. Authors showed that these low thermal conductivities were due on the one hand to the fact that it was not possible to establish a thermally conductive network by physical contact between the graphene nanoplatelets and, on the other, carbon-based graphene acted as IR absorber and reflector, reducing thermal radiation.

Due to the lack of studies regarding the thermal conductivity behavior of polymer nanocomposite foams based on polycarbonate (PC), which is one of the most used thermoplastic polymers in electric and electronic applications, in the current study we investigate the effects of foaming process variables, the resulting cellular morphology and graphene nanoplatelets concentration of PC–graphene nanocomposite foams on their thermal conductivity.

2. Experimental

2.1. Materials and compounding

Polycarbonate used in the current study (Lexan 123R, SABIC; Sittard, Netherlands) has a density of 1.2 g/cm^3 and a melt flow index (MFI) of 17.5 g/10 min measured at 300 °C and 1.2 kg according to ISO 1133. Graphene nanoplatelets (GnP; XG Sciences, Inc.; Michigan, U.S.A.) had 6 to 8 nm thickness with a 15 μm average platelet lateral dimension and a bulk density of 2.2 g/cm^3, as reported by the manufacturer. PC–GnP nanocomposites containing 0.5, 2 and 5 wt% GnP, respectively named for now on PC05, PC2 and PC5, were prepared by melt-mixing using a Brabender Plasti-Corder internal mixer followed by compression moulding, as explained in our previous work [34].

2.2. Foaming

Polymer foaming processes based on the use of CO_2 as physical blowing agent have been vastly considered in both industrial and research fields [35]. Among these, batch foaming using supercritical CO_2 ($scCO_2$), where foaming can be done in both 1-step by means of sudden pressure drop at relatively high temperatures (also known as the pressure-quench method [36]) or in 2-steps where, after a first step of $scCO_2$ saturation at high pressure and low temperature, foaming is usually done by heating in a second stage the saturated CO_2–polymer sample, has been considered as a very versatile process to obtain from very low density foams with highly anisotropic cellular structures to microcellular or even nanocellular polymer foams [37–42], hence enabling the preparation of foams with a wide density range and different cellular structures.

With this in mind, two types of foaming processes were used in the current study: 1-step and 2-step foaming. In both cases, compression-moulded PC–GnP composite circular-shaped discs were saturated with supercritical CO_2 and then expanded (foamed) via $scCO_2$ dissolution. These samples are marked as "1-step" [34]. In the case of "2-step" foaming, after $scCO_2$ dissolution stage inside a high pressure vessel at 80 °C with a total dissolution time of 210 min, discs containing CO_2

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