Graphite Nanoplatelets Dispersion by Melt Reactive Extrusion for the Preparation of Thermally Conductive Polymer Nanocomposites

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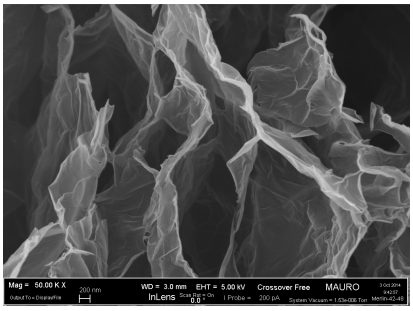
Thermally conductive polymer nanocomposites are of great interest in substituting metal components in all those applications where corrosion resistance, lightweight and processability are required. Nanoparticles with extremely high thermal conductivity (Carbon Nanotubes, Graphene and Graphene Nanoplatelets, hexagonal Boron Nitride) are expected to confer a huge improvement to polymer thermal properties.
Carbon nanotubes (CNT) have been widely used to increase the thermal conductivity of polymeric materials, with different results, mainly limited by the poor thermal contact between nanoparticles [1]. More recently, Graphene Nanoplatelets (GNP) have become an interesting option due to their geometry, which allows to obtain an higher contact between nanoparticles [2,3,4]. However, it is critical to obtain a good dispersion degree of GNP in polymer matrix, especially during melt mixing.
In this work, a GNP with an expanded structure has been melt mixed with cyclic butylene terephthalate (CBT). This oligomer is known to have a very low viscosity that can allow to a complete filling of the expanded structure, which is a preliminary to the obtainment of a good exfoliation degree, and the consequent improvement in the thermal conductivity of the nanocomposite.

CBT (CBT 100, from Cyclics Corporation - USA) were mixed with 10% of FLG024 (from AVANZARE, Spain. BET = 210 ± 12 m^2/g; $D_{90} \leq 205$ µm; $D_{50} \leq 39$ µm) for 5 minutes at 190°C and 100rpm. In a different extrusion process, CBT and GNP were mixed at 245°C and 100rpm for 5 minutes, then Butyl Tin Chloride Dihydroxide was added, as trans-esterification catalyst, and the mixing continued for 10 minutes with the formation of polybutylene terephthalate (pCBT) nanocomposites. Materials were characterized by microscopy (FESEM) and thermal conductivity measurement (Transient Plane Source, HotDisk AB).

A representative FESEM micrographs of FLG024 is reported in Figure 1, showing the expanded GNP structure



References

[1] Z. Han, A. Fina, Prog. Polym. Sci. 36 (2011) 914.
[2] A. Balandin, Nature Materials 10 (2011) 569.
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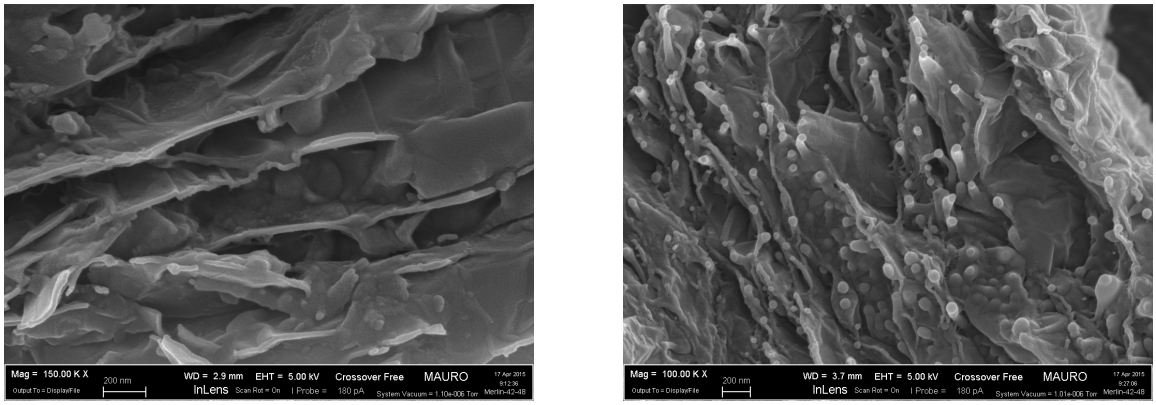
[4] C.C. Teng et al., Carbon 49 (2011) 5107.

Figure 1: FESEM micrograph of FLG024

After melt mixing, CBT is filling the volume between expanded GNPs (Figure 2a), evidencing good wettability of FLG024 by the low viscosity CBT. After CBT polymerization, pCBT is still localized between GNP (Figure 2b), causing little or no separation of GNPs from the original expanded structure.



(a) (b)

Figure 2: FESEM on CBT + 10% FLG024 (a) and pCBT + 10% FLG024 (b)

Thermal conductivity results show that the addition of FLG024 allows to obtain a significant increase in the thermal conductivity of the nanocomposites respect to the pure polymer, both in CBT and pCBT, as visible in Figure 3. In pCBT, the thermal conductivity improvement is slightly higher than in CBT, which is likely due to the partial disaggregation of GNPs into dispersed platelets.

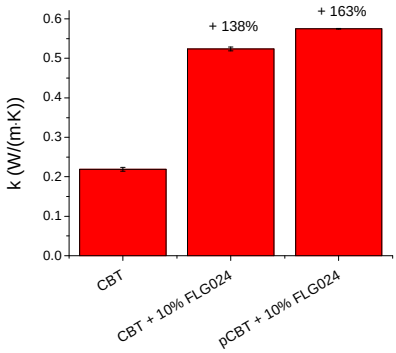


Figure 3: Thermal conductivity results for CBT and its GNP nanocomposites

Melt mixing of GNPs in CBT is a convenient process to allow mixing of a low viscosity polyester oligomer in GNP, and subsequently carry out its polymerization into a high molecular weight polyester, possibly exploiting the increase in viscosity to progressively disrupt the expanded GNP structure into a dispersed platelets morphology. .
Thermal conductivity of the nanocomposites prepared are encouraging and will require further efforts in optimizing polymerization conditions, viscosity of the polymer and imposed shear stress to further improve dispersion of the GNPs. [5]

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