Graphene nanoplatelets for thermally conductive polymer nanocomposites via melt reactive extrusion

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Abstract

As the exploitation of graphene in materials for large scale application is still very much limited by the amount and quality of graphenes available, for large scale bulk applications in polymer nanocomposites, multilayer graphenes or graphene nanoplatelets (GNP) are currently the most interesting graphene-based materials. Thermal conductivities for graphenes were reported in the wide range of about 1500 to 6000 W/mK; however, thermal properties of graphene-based materials may decrease dramatically as a function of the number of layers, the density of topological defects, re-hybridization defects as well as on the presence of impurities.

In this paper, different types of GNPs were thoroughly characterized both in terms of chemical/physical properties and in terms of thermal properties of individual flakes. Two selected GNPs were annealed at 1700°C in vacuum for 1 h to reduce defectivity of the graphenic structure, evidencing that thermal annealing can considerably reduce the amount of defects, as consistently proven by Raman measurements, X-ray photoelectron spectroscopy, X-ray diffraction and thermogravimetry. Thermal conductivity improvement of individual GNP upon annealing was confirmed by scanning thermal microscopy, a scanning probe technique which allows to measure thermal properties at the nanoscale and at the same time achieving information on the morphology, with a spatial resolution of a few tens of nanometers, simultaneously with the topography and lateral force maps, thus combining in a single measurement, properties that cannot be observed at the same time with other techniques.

Both pristine and annealed GNPs were used to prepare polymer nanocomposites, via a melt reactive extrusion process. In particular, GNPs were pre-dispersed in cyclic oligomers of polybutylene terephthalate (CBT) followed by catalyzed ring-opening polymerization of CBT in extrusion. This technique allowed to obtain significantly improved dispersion of GNP compared to conventional melt processing methods, thanks to facile distribution of GNP in CBT and further dispersion as a result of high shear applied once viscosity increase during polymerization. Thermal conductivity results showed significant variability as a function of GNP properties, particularly in terms of defectivity, surface area and lateral dimensions (Figure 1). Furthermore, a dramatic two- to three-fold increase in the thermal conductivity of the nanocomposite was observed in the presence of annealed GNP compared to pristine ones (Figure 1), evidencing the importance of using low defectivity nanoflakes. Thermal conductivity of about 1.7 W/mK, i.e. one order of magnitude higher than for pristine polymer, was obtained with 10%wt of annealed GNPs, which is in line with state of the art nanocomposites prepared by more complex and less upscalable in situ polymerization processes.

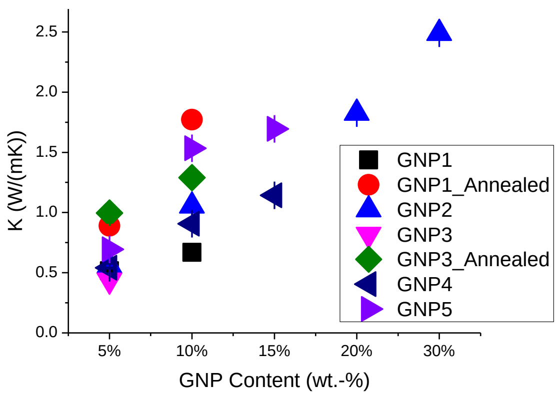

Figure 1: Thermal conductivity values for polymer nanocomposites embedding different types of GNPs or high temperature-annealed GNPs, as a function of % loadings