## Thermally Conductive Polymer Nanocomposites Based on Graphene-Related Materials

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Abstract : Thermally conductive polymer nanocomposites are of high interest for several applications including lowtemperature heat recovery, heat exchangers in a corrosive environment and heat management in electronics and flexible electronics. In this paper, the preparation of thermally conductive nanocomposites exploiting graphene-related materials is addressed, along with their thermal characterization. In particular, correlations between 1- chemical and physical features of the nanoflakes and 2- processing conditions with the heat conduction properties of nanocomposites is studied. Polymers are heat insulators; therefore, the inclusion of conductive particles is the typical solution to obtain a sufficient thermal conductivity. In addition to traditional microparticles such as graphite and ceramics, several nanoparticles have been proposed, including carbon nanotubes and graphene, for the use in polymer nanocomposites. Indeed, thermal conductivities for both carbon nanotubes and graphenes were reported in the wide range of about 1500 to 6000 W/mK, despite such property may decrease dramatically as a function of the size, number of layers, the density of topological defects, re-hybridization defects as well as on the presence of impurities. Different synthetic techniques have been developed, including mechanical cleavage of graphite, epitaxial growth on SiC, chemical vapor deposition, and liquid phase exfoliation. However, the industrial scale-up of graphene, defined as an individual, single-atom-thick sheet of hexagonally arranged sp2-bonded carbons still remains very challenging. For large scale bulk applications in polymer nanocomposites, some graphene-related materials such as multilayer graphenes (MLG), reduced graphene oxide (rGO) or graphite nanoplatelets (GNP) are currently the most interesting graphene-based materials. In this paper, different types of graphene-related materials were characterized for their chemical/physical as well as for thermal properties of individual flakes. Two selected rGOs were annealed at 1700°C in vacuum for 1 h to reduce defectiveness of the carbon structure. Thermal conductivity increase of individual GNP with annealing was assessed via scanning thermal microscopy. Graphene nano papers were prepared from both conventional RGO and annealed RGO flakes. Characterization of the nanopapers evidenced a five-fold increase in the thermal diffusivity on the nano paper plane for annealed nanoflakes, compared to pristine ones, demonstrating the importance of structural defectiveness reduction to maximize the heat dissipation performance. Both pristine and annealed RGO were used to prepare polymer nanocomposites, by melt reactive extrusion. Thermal conductivity showed two- to three-fold increase in the thermal conductivity of the nanocomposite was observed for high temperature treated RGO compared to untreated RGO, evidencing the importance of using low defectivity nanoflakes. Furthermore, the study of different processing paremeters (time, temperature, shear rate) during the preparation of poly (butylene terephthalate) nanocomposites evidenced a clear correlation with the dispersion and fragmentation of the GNP nanoflakes; which in turn affected the thermal conductivity performance. 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.

**Keywords :** graphene, graphene-related materials, scanning thermal microscopy, thermally conductive polymer nanocomposites

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