

**PROCESSING, MORPHOLOGY AND PROPERTIES OF
GRAPHENE REINFORCED POLYMER NANOCOMPOSITES**

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Abstract

A unique combination of excellent electrical, thermal and mechanical properties has made graphene a multi-functional reinforcement for polymers. The goal of this research has been three-fold: exfoliation of graphite for higher surface area, development of effective strategies for processing and characterization of graphene based polymer composites and understanding their processing, structure and property relationships.

Exfoliated carbon sheets can be obtained from graphite oxide (GO). Functionalized graphene sheets (FGS) are formed by rapid pyrolysis of GO. Despite size reduction and distortion in the flat graphene structure by thermal treatments, FGS have high electrical conductivity and can be melt-processed into polymers. GO can be chemically modified with isocyanate, which improves dispersability in organic solvents and polymers. Although not as thermally stable and electrical conductive as FGS, isocyanate treated GO (iGO) has a larger diameter and is advantageous for retaining high toughness of the composites.

FGS and iGO were incorporated into a range of model polymers. Solvent aided blending led to better dispersion of FGS in thermoplastic polyurethane than melt processing. Via solvent mixing, polyurethane became electrically conductive at even less than 0.5 wt% of FGS. With 3 wt% iGO, tensile modulus was increased up to 10 times and N₂ permeation was reduced by 90%, implying high aspect ratio of exfoliated sheets. Morphology of melt compounded graphite and FGS in poly(ethylene-2,6-naphthalate) was characterized with electron microscopy, X-ray scattering, melt rheology and solid property measurements. Unlike graphite, dispersion of FGS quantified from different routes spreads over a wide range due to structural irregularity and simplified model assumptions. Melt viscoelasticity and electrical properties of polycarbonate were significantly modified by graphite orientation. Flow-induced orientation reduced property gains by graphene dispersion, while quiescent-state annealing restored rigidity and electrical conductivity of the composites. Using melt-state rheological and dielectric measurements, micro-structural evolution of FGS in polystyrene was monitored through annealing. Temporal property changes were analogous to the aging response of colloidal

glasses and also influenced by matrix chain relaxation dynamics. Graphene-based polymer nanocomposites can be a new versatile soft material with numerous advantages. For maximized benefits, composite morphology must be tailored appropriately with understanding of its structure-property relationships.