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Feature article

Tailoring the interface in graphene/thermoset polymer composites: A critical review

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ABSTRACT

With the emergence of scientific interest in graphene oxide (GO) in recent times, researchers have endeavored to incorporate GO in thermoset polymeric matrix to develop composites with extraordinary set of properties. The current state of research in graphene/thermoset polymer composites is highlighted here with a focus on the role of interface in dictating the overall properties of the composites. Different strategies like covalent and non-covalent functionalization of GO have been discussed with respect to improvement in mechanical, electrical, thermal and rheological properties. In addition, future prospects have been outlined. By assessing the current state of research in graphene/thermoset composites, it is obvious that graphene derivatives are promising materials in enhancing the structural properties of the nanocomposites at extremely low levels of filler loading. This opens new avenues in designing light-weight composites for myriad applications and by tailoring the interfacial adhesion with the polymer, ordered structure can be achieved at macroscopic processing scales.

1. Introduction and background

Graphene, a 2-D monolayer sheet of several densely packed C-atoms with sp^2 hybridization has revolutionized the field of materials science. Theoretically, it was established in 1940 while, in 1962, Boehm was able to successfully extract thin carbon layers from graphite oxide. In 2004, successful preparation and isolation of stable graphene sheets by Nobel laureates Andre Geim and Konstantin Novoselvo led to the realization of outstanding properties of grapheme [1–3]. It showed extraordinary mechanical properties, electrical conductivity, thermal conductivity and optical transparency. Graphene is the source of all graphitic form or in other words, it is the building block (Fig. 1). Graphene in general is expensive compared to other carbon based nanofillers such as carbon nanotubes, nanofibers, etc. Though some of graphene derivatives, such as GO are inexpensive. Different graphene derivatives assist in manipulating the properties of a polymer in developing unique composites. Graphene, graphene nanoplatelets, graphene oxide (GO) and reduced graphene oxide have drawn considerable attention among the researchers owing to their widely different applications. Their highly competing properties such as mechanical stiffness (>1000 GPa), thermal conductivity (as high as 3000 $Wm^{-1} K^{-1}$), high charge mobility, high specific surface area (2600 m^2/g), high electrical conductivity of the order 2×10^3 S cm^{-1} and high thermal stability up to approx. 600 °C, etc. makes them ideal candidates for polymeric nanocomposites [3,4]. The potential applications range from solar cells, sensors, to drug delivery and bone replacements [5–13].

In this review article, the current state of research in graphene based thermoset polymers like epoxy, polyesters, and polyurethane-based composites are highlighted with an objective to provide a comprehensive overview on the structure-property correlationship in this field with a special attention to the role of interface in dictating the properties. The different strategies to synthesize GO and reduced GO, the different strategies to improve the interface with the polymer and the properties have been addressed with respect to the literature available from the last decade.

2. Different synthesis routes

There are different methods, which are being currently employed to synthesize layers of graphene and graphene oxide, such as chemical vapor deposition, mechanical processes, chemical reduction, unzipping of carbon nanotubes, organic synthesis route,

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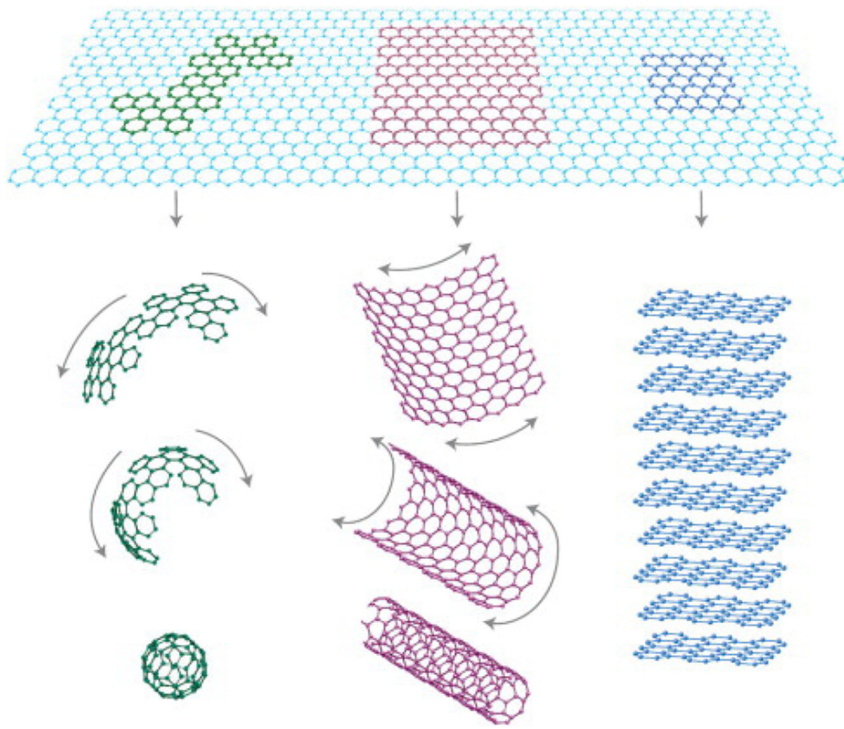

Fig. 1. Graphene as a building block of graphitic material. Reproduced by permission from Ref. [13]. Macmillan Publishers Ltd: [Nature], copyright (2007).

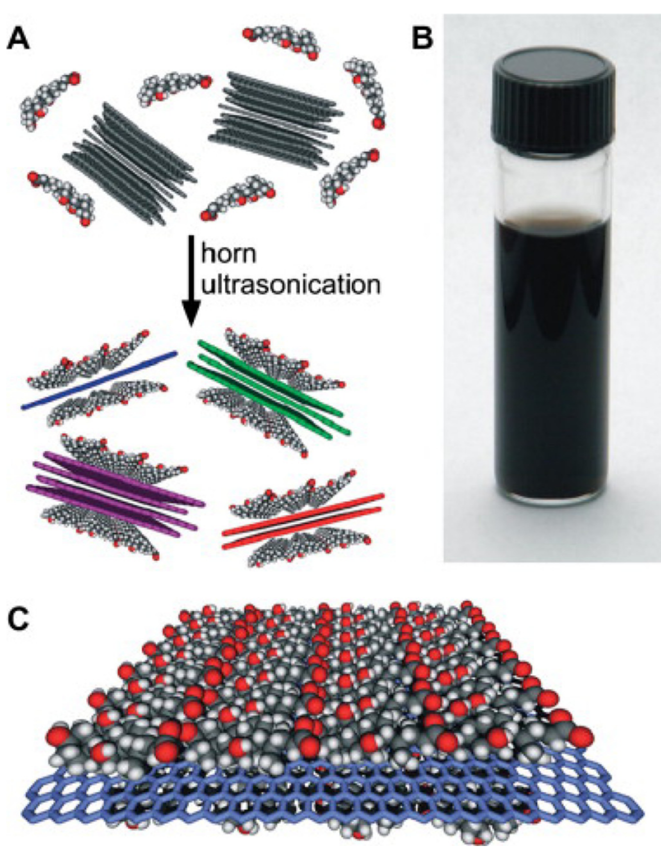

Fig. 2. (A) Schematic illustration of the graphene exfoliation process. Graphite flakes are combined with sodium cholate in aqueous solution. Horn ultrasonication exfoliates few-layer graphene flakes that are encapsulated by SC micelles. (B) Photograph of a 90 μg mL^{-1}graphene dispersion in SC six weeks after it was prepared. (C) Schematic illustrating an ordered SC monolayer on graphene. Reproduced by permission from Ref. [15]. Copyright© 2009 American Chemical Society.

etc. These synthesis techniques are divided depending on the application requisites, in other words, methods are either large or small scale based on purity and precision. Given the brevity of this review, here we will briefly discuss some of the regularly used techniques to synthesize GO and reduced GO and the commonly used characterization techniques to ascertain the reduction process.

2.1. Mechanical process

It involves exfoliation of graphene by different electrical and mechanical means; however, poor yield limits the usage of this technique. Later, researchers attempted to first chemically reduce graphite-to-graphite oxides and then subjected it to exfoliation. Some work in this area also involved exfoliation of graphite in water to produce graphene in presence of a surfactant like sodium dodecyl benzene sulfonate (SDBS). From Raman analysis, no D-band was observed in the spectrum suggesting a defect-free structure, which was further supported with HRTEM. Further electrical study showed reduced conductivity due to the presence of residual surfactant [14].

Efforts were also made to introduce small or guest molecules such as sodium cholate in between the graphite layers by exfoliation and later expanding these layers to prepare high quality graphene sheets (see Fig. 2.). It was found that on annealing at 250 °C sheet resistance of graphene decreased by the order of 2–4 and high optical transmittance was observed [15].

2.2. Chemical vapor deposition (CVD)

Li et al. [16] prepared graphene sheets on copper foils as substrates using a mixture of methane and hydrogen at 1000 °C. The structure was verified using SEM and TEM micrographs, which showed wrinkled structures (Fig. 3). Graphene sheets were also prepared using different surfaces like glass and Si. In another effort