



High-performance and multifunctional epoxy composites filled with epoxide-functionalized graphene



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ABSTRACT

Uniform dispersion and strong interfacial adhesion are two critical prerequisites for application of graphene in the polymer composites. To equip the composites with multifunctional character, as little damage as possible to intrinsic structure of pristine graphene (*pG*) is also normally required. With these ends in view, an epoxide-functionalized graphene (G-EP) was designed, synthesized, and applied for developing the high-performance and multifunctional epoxy composites. Since the structure and properties of *pG* are largely inherited and the introduced epoxide moieties ensure the good dispersion and covalent connection to epoxy network, G-EP exhibits an outstanding filler efficiency compared with *pG* and other graphenic fillers previously explored in epoxy matrix. The resultant composite with only 1.0 wt% G-EP acquires 116% and 96% increments in tensile strength and Young's modulus, respectively, relative to neat epoxy. Furthermore, the electrical percolation threshold is down to 0.16 vol% (0.33 wt%) G-EP, and the thermal conductivity (TC) reaches up to 3.138 W/(m K) at 10 wt% G-EP that corresponds to a TC increment of 189% per 1.0 wt% G-EP loading or 370% per 1.0 vol% loading. A new type of high-performance, multifunctional but much lightweight epoxy composites is thus in prospect.

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1. Introduction

Epoxy is a well established thermoset polymer with a wide variety of applications ranging from electronics to aerospace. Despite the favorable mechanical, adhesive, and chemical and corrosion resistance properties, technical progress has put forward requirements of higher performance and multifunction on the epoxy materials. For example, in the electronics industry, the progress in miniaturizing and integrating the device components has created a need for epoxy materials with high heat dissipation ability when they are applied as the thermal interface materials [1]. In the aviation industry, there are practical demands on the electrostatic dissipation capacity and enhanced mechanical properties when the epoxy materials are used as the structural elements of large high-speed aerocrafts [2]. Filling epoxy with graphene seems to be a promising solution in meeting these objectives. The distinctive one-atom-thick planar structure along with low density and excellent mechanical, thermal, and electrical properties renders graphene an ideal nanofiller for developing the high-performance and multifunctional polymer composites [3,4]. What is more attractive is that graphene can be produced by exfoliation of graphite [5,6], affording cost advantage over carbon nanotube (CNT) that is another sort of important carbon nanofiller but produced by chemical synthesis.

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It is well known that the practical performance of polymer composites depends not only on the inherent properties of fillers but, even more importantly, on their uniform dispersion in and strong interfacial interaction with polymer matrix. For the nanofiller of pristine graphene (*pG*), however, its highly cohesive van der Waals energy (5.9 kJ/mol) makes it hardly possible to be well dispersed in most of polymer matrices including epoxy matrix [7]. On the other hand, the weak interfacial adhesion between *pG* and polymer matrix, resulting mainly from the atomically smooth surface and chemical inertness of graphene, makes the load transfer from matrix to graphene inefficient. Fortunately, surface functionalization of graphene in either a covalent or noncovalent manner is found effective in improving both dispersion and integration of graphene in the matrix [8–10].

In comparison, the covalently functionalized graphene is usually more powerful than the noncovalently functionalized one in terms of enhancing the mechanical properties of the resultant composites [3,4,8]. The most common strategy for covalent functionalization begins with graphene oxide (GO) as the starting material, which has abundant oxygen-rich functional groups with the C:O ratio up to 2:1 and proves to be an eminent platform for numerous chemical transformations [10–13]. However, owing to the excessive disruption of π -orbital structure of graphene, GO suffers from significant loss in electrical/thermal conductivity and thermal stability. Even after the tedious reduction processes, large defect population still remains (C:O ratio of 2.8–23.9:1) and intrinsic properties of *pG* are only partially restored [11]. Toxicity and expense of chemicals used in the preparation and reduction of GO are also issues of severe concern. For all that, the polymer/graphenic filler composites reported hitherto are prepared mostly from GO, reduced GO (*rGO*), or their derivatives. As a result, the physical properties of the obtained composites are far below the anticipated potentials. There still exists a large space to further improve the composite properties by taking some measures. For instance, a functionalized graphene that largely reserves the structure and properties of *pG* but can be tightly integrated into the matrix is taken as the graphenic filler.

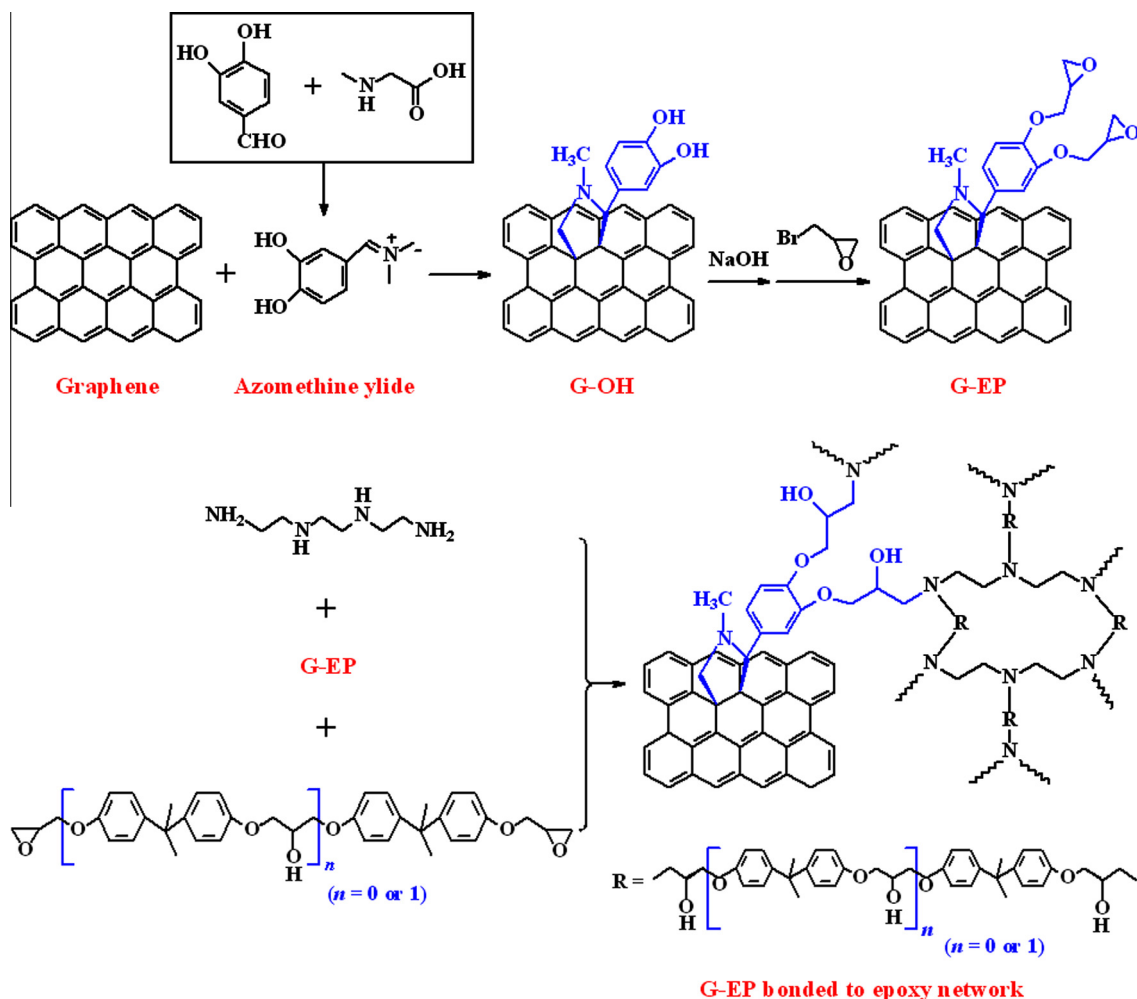


Fig. 1. Synthesis of G-EP and its application in epoxy composite.

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