



The sensitive electrical response of reduced graphene oxide–polymer nanocomposites to large deformation



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ARTICLE INFO

Article history:

Received 14 December 2014
Received in revised form 14 April 2015
Accepted 18 April 2015
Available online 24 April 2015

Keywords:

A. Thin films
B. Plastic deformation
B. Electrical properties
C. Analytical modeling

ABSTRACT

In this paper, the reduced graphene oxide (rGO)–polymer nanocomposite films were prepared by two comparable methods, and their electrical performances at large deformation were measured and analyzed. It is concluded that the gauge factor (GF) value of strain-sensing composite strongly depends on rGO's content and preparing process. The composites with rGO's content between percolation threshold and the content which rGO conducting as a complete network show better sensitive electrical response. What's more, strong interface interaction between conducting filler and matrix benefits the improvement of GF value. The composites we prepared display significant and consistent changes in their GF values when subjected to large deformation, which are suitable for large structural deformation monitoring in advanced civil engineering.

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1. Introduction

Graphene–polymer composites have been studied for several years, and many excellent researches have been reporting on outstanding mechanical reinforcement of polymer with extremely low graphene loadings [1–7]. However, recently, an increasing number of researchers have found that soft, highly stretchable, flexible graphene–polymer conducting composites might be another promising orientation in this field [8–13,28,29]. Taking advantages of high electrical conductivity, optimized dispersion and large aspect ratio of graphene, a small amount of graphene additions in nanocomposites could meet the requirement of electrical response, as well as maintain the ductility and flexibility of polymer matrix. These composites are sensitive to length change and suitable to be applied in a range of fields, such as wearable biosensors, foldable plastic electronics, and large structural deformation monitoring.

There have been some valuable researches on strain-sensing graphene–polymer nanocomposites. Boland et al. [8] infused liquid-exfoliated graphene into natural rubber to create conducting composites which could work at strains exceeding 800% and track dynamic strain. Eswarajah et al. [9] prepared the nanocomposites composed of nitric acid – functionalized graphene and polyvinylidene fluoride (PVDF) by a simple solvent casting technique. The SEM images of nanocomposites with 2 wt% graphene

content revealed the 3D interconnecting conducting network of graphene in the polymer matrix. Wang et al. [11] reported the graphene/poly-dimethylsiloxane (PDMS) nanocomposites as a strain sensor whose GF reaching about 233 at a graphene concentration of 8.33 vol.%, which was measured within a strain range of 2%. Li et al. [13] fabricated the graphene woven fabrics (GWFs) on the PDMS matrix by chemical vapor deposition method. The electrical resistance of GWFs increases exponentially with strain whose gauge factors of $\sim 10^3$ at 2–6% strain and $\sim 10^6$ at 6–10% strain.

It is well known that graphene sheets tend to form agglomerates or restack in water and organic solvents, but graphene oxide can form well-dispersed aqueous colloids [15,30–32]. Hydrophilic graphene oxide, which is electrically insulating, can be converted back to conducting graphene by chemical reduction, for example, by using hydrazine. Thus, reduced graphene oxide is often employed in the solution-based route to prepare graphene–polymer composites.

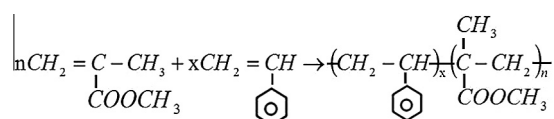
In our study, the reduced graphene oxide–polymer nanocomposite films were prepared by two comparable methods. RGO's content and preparing process were taken into consideration, and systematic experiments on rGO–polymer composite films were carried out. The effect of conductive particles and interfacial interaction on electrical sensitivity of composites were also explored. The conclusions we have achieved could be helpful for explaining the conductive mechanism of composites with large deformation and enhancing the electrical performance and strain sensitivity of graphene–polymer composite.

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2. Experimental details

2.1. Materials

Graphite oxide (Material No. SE2430; Average size: 5 μm) was purchased from The Sixth Element Co. Ltd., Changzhou, China. Styrene–acrylic emulsion (Material No. ps608) was supplied by BASF (China) Co. Ltd., Shanghai. Styrene–acrylic emulsion is a copolymer of styrene and acrylate, which styrene replacing methylmethacrylate partly in polymerization process.



2.2. Preparation of the rGO–polymer composite film

The strain-sensitive material could be prepared by two different methods shown in Fig. 1. The preparing process was designed to explore the effect of rGO's conductivity and interfacial interaction on electrical sensitivity of composites, and it has been demonstrated that the design is effective.

In the first method, the integration of filler and matrix was done before the reduction of graphene oxide composite film (rGO-1 composite film). A typical procedure of the first method is as follows: A certain amount of graphite oxide powder was added into deionized water to form 2.0 mg/ml graphene oxide (GO) dispersion accompanied by ultrasonic treatment. After the GO dispersion was diluted into various concentrations, they were mechanically

blended with styrene–acrylic emulsion to fabricate different mixtures whose GO accounting varies between 0.2 and 5 wt%. Then, the GO-emulsion mixtures were subjected to 50 °C drying to form GO composite films after complete solvent evaporation. Finally, these GO composite films were reduced in the pure hydroiodic (HI) acid at 100 °C for 12 h, then washed by water and dried out.

As for the second method, the greatest difference compared with the first one, is that the reduction process of GO took place before blending with the emulsion (rGO-2 composite film). A typical procedure is as follows: After GO dispersion was prepared as mentioned above, Sodium dodecyl-benzene sulfonate (SDBS) accounting for 1 wt% of the GO colloid was added, stirred to form a uniformly dispersion accompanied by sufficient ultrasonication. Then, the dispersion was mixed with hydrazine hydrate (0.2% by volume), and reduced at room temperature for 1–2 h before being allowed to have a high-temperature reaction in a water bath at 80 °C for another 12 h. The integration process of rGO composite film refers to the similar process of GO composite. As a contrast, rGO without SDBS was also prepared. The rGO-1 and rGO-2 composite films could hardly be distinguished by exterior appearance while the GO composite films presented light color and some degree of transparency.

2.3. Characterization

The degree of reduction was investigated by XPS (X-ray photoelectron microscopy, PHI 5000C ESCA System, USA) and Raman spectroscopy (Jobin-Yvon HR 800, France). AFM (SPA-300HV, Japan) was used to examine the shape and magnitude of rGO. The surface morphology of composite films was viewed by SEM (FEI Quanta 200 FEG electron microscopy). The electrical

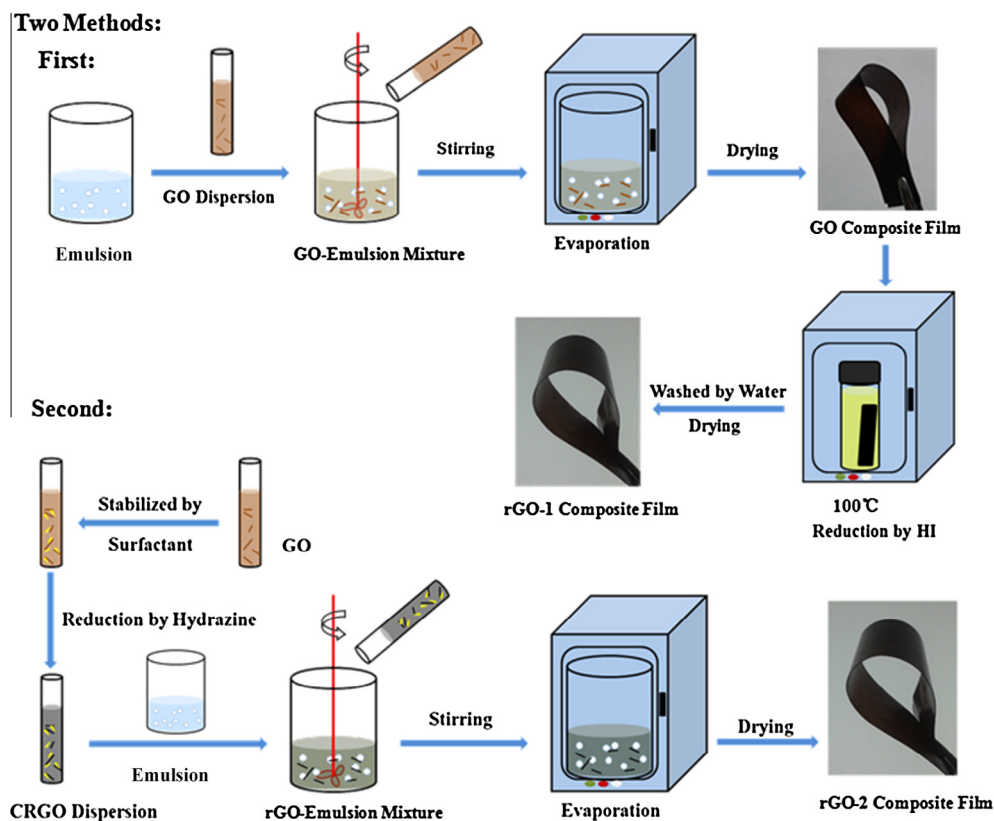


Fig. 1. Schematic diagram of the two methods on preparing the rGO–polymer composite films. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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