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Communication

Self-assembly of graphene oxide nanosheets in *t*-butanol/water medium under gamma-ray radiation

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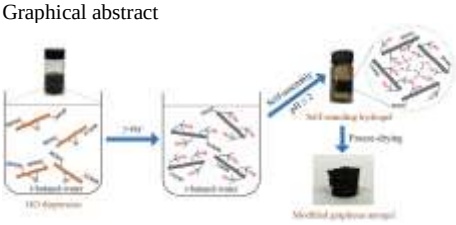
Graphical abstract



Graphene oxide (GO) nanosheets dispersed in strong acidic *t*-butanol/water medium can be reduced and self-assembled into a self-standing graphene hydrogel under *γ*-ray radiation, providing a facile and economical preparation method for hydroxylalkylated graphene-based aerogel.

ABSTRACT
The research on the properties of graphene oxide (GO) in various media has become one of the hottest topics since GO is now the main raw material for graphene-based advanced materials. In this work, the *γ*-ray radiation chemistry effect of GO nanosheets and their self-aggregation behavior in *t*-butanol/water medium were investigated. The results show that GO nanosheets are reduced and hydroxyalkylated simultaneously by the alcohol free radicals produced by the radiolysis of *t*-butanol/water solution under *γ*-ray radiation. The radiation-modified GO nanosheets will self-assemble into a self-standing graphene hydrogel when the pH of the solution is lower than 2. A hydroxyl-functionalized free-standing graphene aerogel is further obtained simply by freeze-drying. This work provides not only a general self-assembly mechanism of GO nanosheets in strong acidic alcohol/water media under high energy radiation, but also a facile and economical preparation method for hydroxyalkylated graphene-based aerogel.

Keywords:
Graphene oxide
Gamma-ray radiation
t-Butanol/water solution
Self-assembly
Hydroxyalkylation
Graphene hydrogel
Graphene aerogel

Graphene oxide (GO) has become the current global research focus since most reported promising graphene-based advanced materials should be originated from GO due to the difficulty and the high cost for acquiring stable graphene nanosheets as the raw material [1-4]. For example, graphene hydrogels/aerogels (GHs/GAs) have been discovered to possess outstanding chemical and physical performance [5-7], and show intriguing application prospect in the areas such as catalysts [8], absorbents [9], and supercapacitors [10]. The current effective synthesis route for GHs/GAs is to make GO nanosheets self-assemble in a hydrothermal reduction system at a high temperature [6,11] or through a chemical crosslinking method [12,13]. Obviously, it is deserved to make clear the compositional or morphological changes in GO nanosheets in various reaction environments in order to control the physicochemical behavior and create novel graphene-based advanced materials.

γ-Ray is a kind of ionizing radiation, which has been widely used for the production of a wide range of functional materials from inorganic nanoparticles [14,15] to macroscopic polymers [16,17] by taking advantage of the diverse radiation chemical effect of matters. Recently, some researchers reported that GO nanosheets dispersed in dimethyl formamide or the aqueous solution of isopropanol and ethylene diamine can be reduced under *γ*-ray radiation [18-20], and even can self-assemble to form GH and GA [21]. Although the reduction of GO has been considered mainly to be attributed to the strong reductive hydrated electron and the scavenging effect of isopropanol or amine on oxidative radical (HO·) generated by the water radiolysis, the reduction and self-assembly mechanisms of GO nanosheets in aqueous systems under *γ*-ray radiation are still unclear since different results from these systems are

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