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Preparation of RGO/Fe_3O_4/poly (acrylic acid) hydrogel nanocomposites with improved magnetic, thermal and electrochemical properties

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HIGHLIGHTS

- A hydrogel nanocomposite composed of RGO/Fe_3O_4/PAA was synthesized successfully.
- Increasing the percentage of iron nanoparticles improved magnetic properties.
- Increasing the percentage of RGO improved thermal and electrochemical capacity.
- The Fe_3O_4 nanoparticles directly affected magnetic properties.

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ABSTRACT

A hydrogel nanocomposite composed of reduced graphene oxide (RGO), iron oxide (Fe_3O_4) nanoparticles, and polyacrylic acid (PAA) was prepared using radical polymerization. Different percentages of RGO, Fe_3O_4, and PAA were used to prepare the nanocomposite. Fourier transform infrared spectroscopy (FTIR) results confirmed the formation of the nanocomposite's chemical structure. X-ray power diffraction (XRD) patterns revealed the principal peak's 2θ value to be 77.39° with the size of the nanocomposite particles estimated at 96 nm.

Results indicated that the electrochemical capacity of the nanocomposites was controlled by the weight percentage of RGO. Increases to the potential scan rate reduced porosity and surface area, thereby decreasing the electrochemical capacity of the nanocomposites. Moreover, increasing the percentage of Fe_3O_4 nanoparticles in the nanocomposites improved their magnetic characteristics and thermal properties. The latter also improved when the RGO percentage increased.

1. Introduction

Graphene is a monolayer material composed of carbon atoms that are connected and bonded in a hexagonal network [1,2]. Graphene is of interest to scientists due to its favorable mechanical [3], electrical [4], thermal [5], and optical [6] features as well as its high surface area [7] and control through chemical agents [8]. Graphene is used as the building block in the fabrication of carbon nanotubes and large fullerenes [9]. Graphene has specific physical features, including the observation of the quantum Hall effect at room temperature [10], very high electron mobility [11], appropriate thermal conductivity [12], high mechanical strength [13], flexibility [14], a high Young's modulus [15], fracture resistance [16,17], the absorption of metal ions [18] and water and soil pollutants [19], and catalytic features [20]. Graphene oxide can be used to produce electrically conductive compounds through the use of reductive methods such as hydrazine [21], dimethyl hydrazine [22], hydroquinone [23] and/or sodium borohydride [24], thermal methods [25], and ultraviolet irradiations [26].

While graphene and its derivatives do not exhibit magnetism, researchers have been able to create magnetic properties by overlaying nanoparticles on the grouping network. These include Fe_3O4 nanoparticles, which are notable for their abundance, non-toxicity, high reactivity, favorable melting point, and ability to modify and remove contaminants. One of the advantages of Fe_3O4 nanoparticles is their ability to use magnetic properties to separate contaminants from adsorbents in a simple and economical fashion [27].

Only a limited number of methods are used to prepare magnetic graphene hybrids [28]. The formation of Fe_3O4–RGO hybrids is usually achieved by the simultaneous reduction of the iron

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chlorides FeCl3·6H2O and FeCl2·4H2O or through the stabilization of the magnetic nanoparticles on RGO's surface [29]. Many nanomaterials, such as CO3O4 [30,31], Cu/Ni alloys, TiO2, Au, and ZnO [32], are absorbed through physical and chemical reactions on RGO due to its large surface area. The use of graphene oxide and RGO in composites is considered an advantage in mechanical characteristics. In order to prevent particles from adhering to each other, low percentages of graphene oxide and RGO are used in the synthesis of polymer nanocomposites, including polyacrylic acid, polystyrene, and silica [33]. Additionally, recent studies have proposed the preparation of magnetic nanocomposites using Fe_3O4 nanoparticles in combination with polymers like polyaniline, polypyrrole, polystyrene, and polyaniline [34]. Carbon-coated magnetic nanoparticles have also received considerable attention recently due to their thermal and chemical resistance [35].

Graphene's potential applications include the construction of very fast transistors [36], data storage [37], liquid crystal tools [38], energy storage [39], solar cells [40], reduction in batteries charging time [41], mass sensors [42], and antibacterial papers [43]. Numerous medical uses have been proposed for magnetic nanocomposites. Incorporating magnetic Fe_3O4 nanoparticles into polymeric hydrogels creates nanocomposites that can be controlled remotely by magnetic fields [44].

The purpose of this study was to prepare RGO/Fe_3O_4/PAA nanocomposites with special magnetic, electrochemical, and thermal features. We used Fe_3O_4 nanoparticles, RGO, and PAA to improve the nanocomposites' magnetic characteristics, capacitance, and thermal impedance, respectively, allowing them to be used in various industries and high-temperature environments.

2. Materials and methods

2.1. Materials

Graphite, sulfuric acid (98%; H_2SO_4), potassium permanganate ($KMnO_4$), hydrogen peroxide (30%; H_2O_2), sodium nitrate ($NaNo_3$), and hydrochloric acid (37%; HCl) were used to prepare graphene for this study. Hydrazine monohydrate (N_2H_2) was used to reduce the graphene oxide. Acrylic acid ($C_3H_4O_2$), tetramethylethylenediamine ($C_6H_{16}N_2$), potassium persulphate ($K_2S_2O_8$), iron oxide nanoparticles (Fe_3O_4), and an *N*, *N′*-methylene-bis-acrylamide (99%) cross-linker were used to prepare the hydrogel nanocomposites. All required compounds and materials were purchased from Merck in Germany.

The present study used a Gemini BV Sigma2-15 centrifuge for purifying the solution, an ultrasonic bath, and a Sonicket device model Banelin to homogenize the solution. A PerkenElmer Spectrum 400 FTIR spectrometer, Shimadzu XRD-6000 x-ray diffractometer, and Philips XL30 electron microscope were used to identify the obtained products, and a cyclic voltammetry (CV) device model Autolab PGSTAT-12/30/302 was used to determine their electrochemical characteristics. A VSM device model Mdkft was used to determine the samples' magnetic features, and a TGA device model TGA-PL-Polymer laboratory was applied to determine their thermal features.

2.2. Methods

2.2.1. Synthesize graphene oxide

Hummers method' was used to synthesize graphene oxide. First, 2 gr graphite (500 mesh) and 50 ml concentrated sulfuric acid (98%) was poured into a 250-ml beaker and stirred; 2 gr sodium nitrite were then added. Following this, an ice-water bath for 1 h at 0 °C. Next, 7.3 g potassium permanganate was gradually added to the mixture over the course of 2 h. The temperature was increased to 35 °C, and the mixture was stirred for 2 h. After this, 46 ml double-distilled water was added to the reaction container, and the mixture was stirred for an additional 30 min at 90 °C. The reaction was stopped by adding 140 ml double distilled-water and 16 ml hydrogen peroxide (3%). The resulting compound was graphite oxide.

The graphite oxide suspension was placed in a 35-kHz ultrasonic bath for 30 min to produce monolayer graphene oxide, which was then washed three times with a hydrochloric acid (3%) solution and filtered. The obtained suspension was washed three times with double-distilled water and filtered again. The brown-colored solid material obtained through this process was dried in an oven at 40 °C for 24 h.

2.2.2. Preparation RGO

To do so, platelets were extracted from the graphene oxide colloid suspension and mixed with distilled water in a balloon at a ratio of 3 mg/ml. The hydrazine monohydrate (1 ml per every 3 mg graphene oxide) was immediately added. The mixture was placed in an oil bath for 12 h at 80 °C. Finally, a black powder RGO was obtained.

2.2.3. Preparation poly (acrylic acid) hydrogel

Acrylic acid (2 ml) was resolved in 10 ml distilled water and transferred to a reactor equipped with a 300-rpm stirring device where the mixture was stirred. After the mixture was homogenized, 0.1 gr potassium persulfate initiator, 0.06 gr *N*, *N′*-methylene-bis-acrylamide cross-linker, and a small amount of tetramethylethylenediamine catalyst were added. The mixture was stirred again. A room-temperature pure nitrogen gas flow was used to eliminate dissolved oxygen. After 1 h, 16.2 ml sodium hydroxide 1 M was added to the mixture to partially neutralize carboxylic groups in the resulting polyacrylic acid. Ethanol (200 ml) was then added to the mixture, which was left to settle in a dry place for 12 h to dehydrate the gel. Following this, the remaining ethanol was isolated and 200 ml fresh ethanol was added again. The mixture was then left to perfectly freeze for 24 h. Finally, the gel particles were dehydrated, filtered, and dried in an oven for 10 h at 45 °C (see Figs. 1 and 2).

2.2.4. Preparation RGO/Fe_3O_4/poly (acrylic acid) hydrogel nanocomposites

In the present study, six nanocomposites were prepared with different weight percentages: 0.03 gr and 0.05 gr graphene oxide;

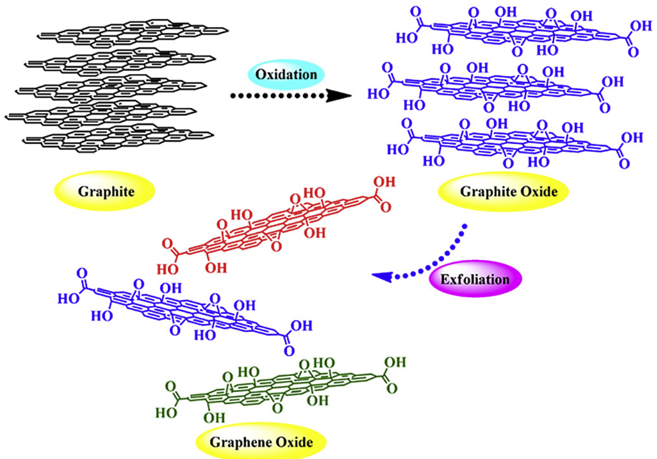

Fig. 1. Preparation graphene oxide from graphite.