

Synthesis of Reduced Graphene Oxide Using Tea Extract and Its Application in the Removal of Methylene Blue from Effluents.

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Abstract

An environment friendly, simple and cost-effective method for reduction of graphene oxide (GO) by using green tea extract method is reported. Reduced graphene oxide (RGO) was characterized by X-ray diffraction (XRD), Scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), and UV-visible spectroscopy analysis. RGO has shown to possess remarkable dye disposal property of Methylene blue dye. The Green tea reduced GO material offered a good potential in the removal of dye waste water.

Keywords: Dye adsorbent, Green Reduction, Graphene Oxide, Reduced Graphene Oxide.

Introduction

Graphene is a single layer of sp² hybridized carbon atoms arranged in a honeycomb lattice, which has established significant attention in the scientific community worldwide due to its unique properties

such as electrical, optical, mechanical, thermal, gas barrier, sensors, energy storage and biomedical etc. Recently, numerous scientific reports have described the application of graphene as a good adsorbent. Graphene and its derivatives have opened a new dimension for development of adsorbents for elimination of heavy metals and dyes from wastewater. Numbers of green methods have been reported for the reduction of GO with various green reagents such as alkaloids, amino acids, proteins, polyphenolic compounds, polysaccharides, enzymes and vitamins. While the reduction effectiveness found through green methods is typically lower than that obtained with chemical methods. However, application of such toxic chemicals and reducing agents for large scale remedial applications is possibly harmful to ecosystem and human health. Therefore, to minimize environmental effects, the production of eco-friendly reducing agents has been investigated. So, One and only alternative is the green synthesis of materials which have been broadly used in environmental remediation.

In this work, we proposed environmentally friendly method for the reduction of graphene oxide to reduced graphene oxide (RGO) by using Green tea extract. The GO were synthesized by chemical exfoliation technique and then reduced by green tea extract solution. The synthesized reduced graphene oxide (RGO) was studied to evaluate its ability to adsorb the methylene blue dye.

Experimental

Materials

Graphite fine powder (150 mesh, 99.5%) and Sodium nitrate (NaNO_3) were purchased from CDH. Potassium permanganate (KMnO_4 , 99.5%) was purchased from Merck. Sulfuric acid (H_2SO_4 , 98%) and Hydrogen peroxide (H_2O_2 , 30%) were purchased from Finar Chemicals. Green tea powder was purchased from Apollo Pharmacy. All the chemicals were of analytical grade and used without further purification.

Synthesis of Graphene Oxide

Graphene oxide from graphite was prepared according to modified Hummer's method. 1 g of graphite powder and 0.5 g NaNO_3 were added to 23 mL of concentrated H_2SO_4 in a 500 mL flask. The prepared mixture was stirred in an ice bath. Stir it till homogeneous paste is formed. 3 g KMnO_4 was then added slowly (in little portions) with continues stirring. Then, the flask was

removed from ice bath to maintain the reaction mixture at room temperature. After 2 hr, 46 mL of deionized water was added drop wise in the reaction mixture and temperature was kept to 98 °C using an oil bath for 15 min. Then add 100ml of deionized water and 100 mL of 30% H₂O₂ solution into the reaction mixture. The reaction mixture turned bright yellow. Allow it to stand and decant the supernatant. The supernatant was washed with deionized water and HCl for several times. The residue was centrifuged at 8000 rpm till the pH becomes nearly 5 to 6. The residue was dried in an oven at 50 °C for 72 h. The brown coloured material was obtained.

Synthesis of Green Tea Reduced Graphene Oxide.

Reduced graphene Oxide was prepared by reduction using Green tea extract. Green tea extract was used as a reducing agent because it's an excellent source of polyphenolic compounds. 1 g of tea powder was added to 100 mL of deionized water and boiled at 100 °C for 1 h. After 1 h allow the reaction mixture to attain the room temperature. Then filter the solution. 50 mg GO was added in the solution of green tea extract and heated in an oil bath at 85 °C in a nitrogen atmosphere for 4 h. The obtained solution was filtered and washed several times with ethanol and deionized water to remove excess of polyphenols. The residue was dried in an oven at 50 °C for 24 h. The black coloured product was obtained.

Removal of Methylene blue by RGO.

Exactly 20 mg of reduced graphene oxide added in 100 ml of the methylene blue dye solutions with constant stirring. During the experiment samples were collected at specific time intervals (10, 20, 30, 40, 50 min) for the UV- vis analysis. The absorbance was recorded for methylene blue at 664 nm. The percentage removal of the dye was determined by using the equation given below.

$$\% \text{ of removal} = \frac{(C_i \times C_f)}{C_i} \times 100$$

Where, C_i and C_f are initial and final dye concentrations respectively.

Characterization

GO and RGO were characterized using X-ray diffraction (XPERT-PRO diffractometer) equipped with goniometer PW 3050/60 working with Cu(K_α) radiation of wavelength 1.5406 Å°, Scanning

electron microscopy (SEM, Hitachi S-3000N), Fourier transform infrared spectra frequency range of 600 to 4000 cm^{-1} by Bruker FT-IR spectrometer and UV- vis absorption spectra a wavelength range of 200–800 nm by Jasco model-v630 spectrometer.

Result and Discussion

X-ray diffraction pattern (Fig. 1), the appearance of a broad peak at $2\theta = 26.50^\circ$ is due to the formation of few layers of RGO sheets. The reflection peak appeared at $10^\circ 2\theta$ value was attributed to GO. The peak of GO which was absent in the diffraction pattern of RGO suggesting the absolute exfoliation of GO. Another peak at $43^\circ 2\theta$ coincide to the turbostratic band of disordered RGO.

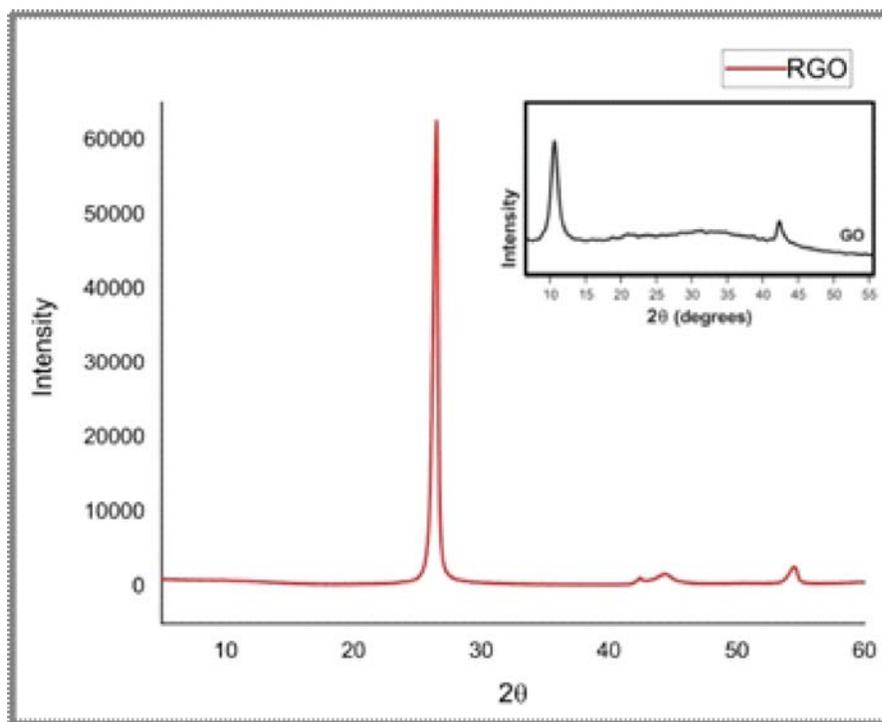


Fig. 1. XRD pattern of RGO

SEM studies (Fig. 2) revealed that the formation of transparent and exfoliated RGO due to the procedure of reduction with green tea extract.

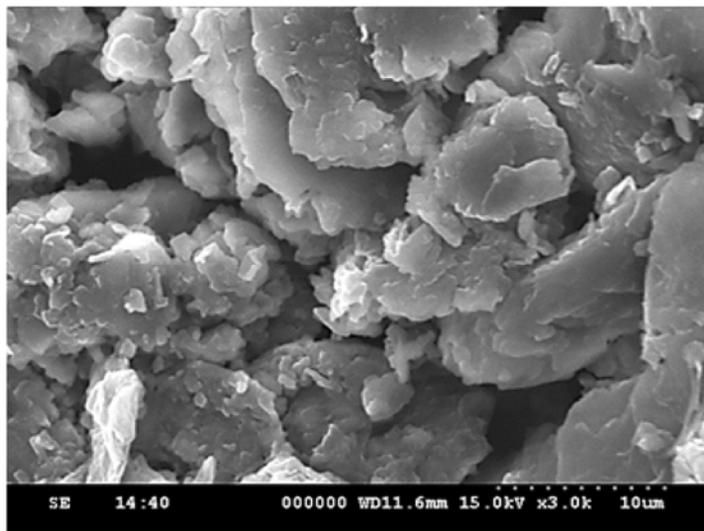


Fig. 2. SEM image of RGO

The peak in UV-visible spectrum at 234 nm is accredited to the π - π^* transition of C=C bonds in GO (Fig. 3). The peak shown at 268 nm is due to green reduction of GO. Although, the GO peak is absent in the RGO spectrum due to rebuilding of graphitic π -electronic conjugated network structure in RGO after reduction.

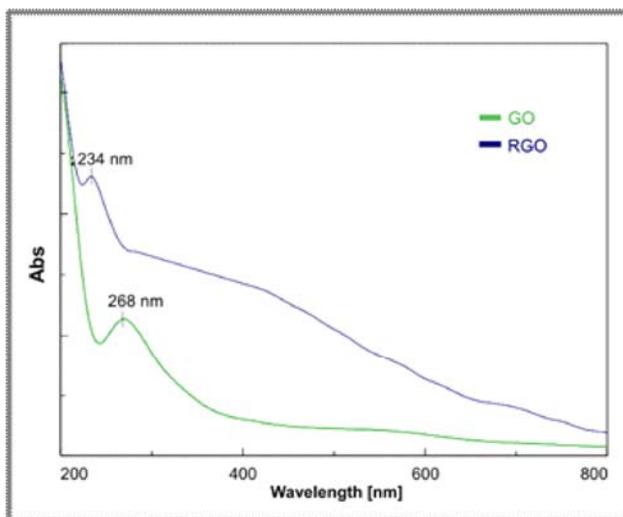


Fig. 3. UV-visible spectra of GO and RGO.

FT-IR spectra of GO and RGO (Fig. 4). The existence of oxygen functional groups confirm the

successful exfoliation of GO. The typical peaks of GO were observed at 1650 cm^{-1} (C=O stretching) and at 1040 cm^{-1} (C–O stretching vibration), with a broad peak at 3270 cm^{-1} (O–H stretching vibrations). The disappearance of peak at 1040 cm^{-1} in RGO spectra shows a decrease of the C–O stretching vibrations. A broad peak at 3270 cm^{-1} is compressed in the RGO due to removal of hydroxyl groups. This observation supports the reduction of RGO.

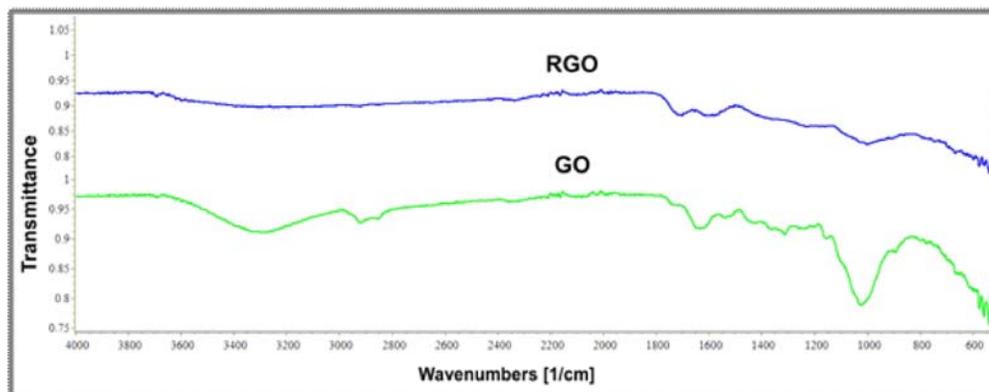


Fig. 4. FT-IR spectra of GO and RGO

Dye removal property of RGO was explored with methylene blue dye without interaction to either sunlight or UV using of 20 mg of RGO (Fig. 5). It proposes comprehensive elimination of the dyes in a time span of 50 min.

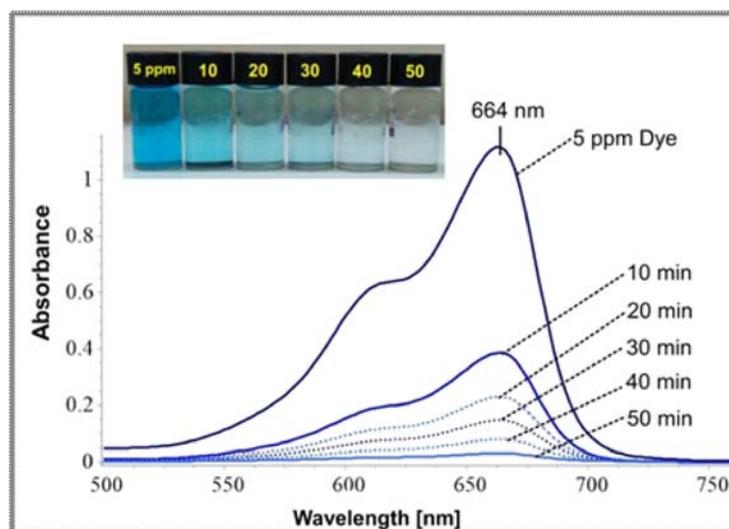


Fig. 5. Removal of Methyleneblue in the presence of RGO

Conclusion

X-ray diffraction, SEM imaging, FT-IR and UV-vis spectroscopic techniques clearly reveal the efficient reduction of few layered graphene from GO using green tea extracts. The synthesized RGO was shown to possess suggestively good dye removal properties. The simple and effective method of reduction makes it possibly attractive for the low-cost synthesis of graphene and related materials.

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