Synthesis of reduced graphene oxide by electrochemical method for electromagnetic interference shielding applications

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ABSTRACT

In this study, graphene oxide (GO) was synthesized via an electrochemical exfoliation method in an $(NH_4)_2SO_4$ *electrolyte solution using two different types of graphite electrodes, namely graphite foil and graphite plate, under an applied potential of 10 V with a controlled current density for 45 minutes. The obtained GO was subsequently subjected to thermal reduction in a nitrogen* (N_2) *atmosphere at 800 °C for 1 hour with a heating rate of 5 °C/min, yielding reduced graphene oxide (rGO). The structural and morphological characteristics of rGO were investigated using scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray diffraction (XRD), Raman spectroscopy, Fourier-transform infrared spectroscopy (FTIR), and energy-dispersive X-ray spectroscopy (EDX). The results revealed that rGO synthesized from graphite foil electrodes exhibited a multilayered structure with a thickness corresponding to 8–15 graphene layers. This morphology is considered highly suitable for the development of materials for electromagnetic interference (EMI) shielding applications.*

Keywords: Graphene oxide (GO); Reduced graphene oxide (rGO); Electrochemical exfoliation; Electromagnetic interference (EMI) shielding.

1. INTRODUCTION

Graphene, a two-dimensional material consisting of a single layer of carbon atoms arranged in a honeycomb lattice, has attracted significant scientific interest owing to its exceptional electrical, thermal, and mechanical properties. With an ultra-high electrical conductivity, large theoretical surface area (~2630 m^2g^{-1}), and remarkable mechanical strength, graphene has been widely investigated for diverse applications including energy storage devices, chemical and biological sensors, conductive coatings, and flexible electronics [1]. Among its derivatives, graphene oxide (GO) and reduced graphene oxide (rGO) have received particular attention, since they can be produced on a large scale at relatively low cost using chemical or electrochemical exfoliation methods [2]. GO contains abundant oxygen-containing functional groups that improve dispersibility in aqueous or polymeric matrices, while rGO partially restores the conjugated π-structure, thereby enhancing electrical conductivity and enabling advanced functional applications [3].

In recent years, electromagnetic interference (EMI) shielding has become an important research focus due to the rapid proliferation of electronic devices, wireless communication systems, and defense technologies. Effective EMI shielding materials must simultaneously exhibit high electrical conductivity, low density, mechanical robustness, and tunable microstructures in order to optimize reflection and absorption mechanisms [4]. Recent studies have reported that multilayered rGO structures can provide efficient multiple reflections, interfacial polarization, and dielectric losses, all of which contribute to enhanced EMI shielding effectiveness [5].

Despite these promising features, large-scale and environmentally friendly production of rGO with controlled structural properties remains a challenge. Chemical oxidation methods, such as the Hummers' method, involve hazardous reagents and generate considerable chemical waste. In contrast, electrochemical exfoliation of graphite has emerged as a green, scalable, and efficient approach to producing GO with high purity and porosity [6]. Furthermore, subsequent thermal reduction can restore electrical conductivity, yielding rGO with favorable structural integrity for EMI shielding applications [7].

In this study, we report on the electrochemical synthesis of graphene oxide using an ammonium sulfate $(NH_4)_2SO_4$ electrolyte with two types of graphite electrodes, namely graphite foil and graphite plate, under an applied potential of 10 V. The obtained GO was thermally reduced in an inert N_2 atmosphere at 800 °C to yield rGO. Comprehensive structural and morphological characterizations were carried out using SEM, TEM, XRD, Raman, FTIR, and EDX techniques. The results indicate that rGO derived from graphite foil electrodes exhibits a multilayered structure consisting of 5 ÷ 10 layers, which is highly promising for the development of advanced EMI shielding materials.

2. EXPERIMENT

2.1. Materials

Graphite foil (purity ≥ 99,8%, thickness ~ 0,1 mm) and graphite plate (purity ≥ 99,8%, dimensions 10 × 30 × 2 mm) were purchased from Xilong Chemical Co., Ltd. (China) and used directly as anode materials for electrochemical exfoliation without further treatment. A cylindrical graphite rod (purity ≥ 99,9%, diameter 6 mm) was employed as the cathode. Ammonium sulfate $(NH_4)_2SO_4$, analytical grade, ≥ 99%, was used as the electrolyte and prepared as an aqueous solution with a concentration of 0.1 M. All chemicals were of analytical grade and used without additional purification. Deionized (DI) water with a resistivity of 18,2 MΩ·cm was utilized for solution preparation and washing of the products.

2.2. Fabrication of materials

The electrochemical exfoliation process was carried out in a two-electrode configuration using a direct current (DC) power supply at a constant potential of 10 V for 45 minutes under ambient conditions. The resulting suspension was collected and repeatedly washed with deionized water to remove residual electrolyte, followed by vacuum drying at 60 °C. The obtained graphene oxide (GO) was subsequently subjected to thermal reduction in a tubular furnace under a nitrogen (N_2) atmosphere at 800 °C for 1 hour with a heating rate of 5 °C/min, yielding reduced graphene oxide (rGO).

2.3. Research methods

Surface morphologies of materials are analyzed through scanning electron microscopy (SEM, Hitachi S-4800) and transmission electron microscopy (TEM, JEM 2100). Energy Dispersive X-ray analysis (EDX) and elemental mapping are conducted on the same equipment. The Crystal structures of samples are analyzed by X-ray diffraction (XRD, Bruker D8 Advance), Raman spectroscopy (Raman microscope, Thermal scientific DXR3) and Fourier-transform infrared spectroscopy (FTIR, Brucker tensor II). The electrical conductivity of the material was measured using a Keysight Technologies B2900BL system. The electromagnetic shielding performance was evaluated using a vector network analyzer integrated with an N9918A spectrum analysis system, in accordance with Military Standard TCQS 71:2016/VKHCNQS. The measurements focused on determining the transmission loss parameter to elucidate the interaction between the investigated materials and the incident electromagnetic waves.

3. RESULTS AND DISCUSSION

3.1. Study on the characteristics of the material

The morphology of the two types of graphite electrodes, namely graphite foil and graphite sheet, as well as the graphene oxide (GO-F and GO-S) and reduced graphene oxide (rGO-F and rGO-S) materials obtained after electrochemical exfoliation and subsequent thermal reduction, was investigated by scanning electron microscopy (SEM) and is presented in Figure 1.

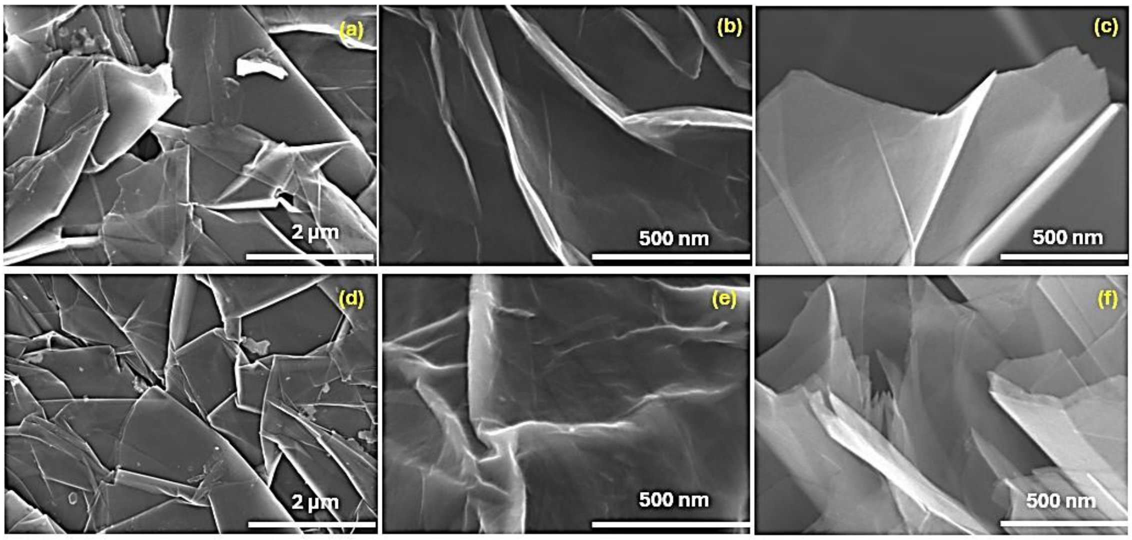


Figure 1. *SEM images of graphite foil (a), GO-F (b), rGO-F (c) and graphite sheet (d), GO-S (e), rGO-S (f).*

Figures 1a and 1d show SEM images of graphite foil and graphite sheet prior to electrochemical exfoliation. The graphite foil exhibits a compact and homogeneous bulk structure with a relatively smooth surface, tightly stacked layers, and only a few cracks or edge defects. In contrast, the graphite sheet displays larger bulk domains with a more irregular surface, less efficient layer stacking compared to the foil, and the presence of grooves and intrinsic defects, making it more susceptible to localized electrochemical attack.

Figures 1b and 1e illustrate the morphology of graphene oxide synthesized from foil electrodes (GO-F) and sheet electrodes (GO-S) after the electrochemical exfoliation process. The GO-F sample appears uniformly exfoliated, with numerous wrinkles and folded edges, forming thin, semi-transparent sheets with well-defined boundaries. In comparison, the GO-S sample exhibits thicker flakes with lower transparency and pronounced multilayer stacking. Therefore, the use of graphite foil as the starting electrode facilitates the preparation of thinner and more uniform GO sheets, which are more suitable for applications requiring high electrical conductivity. Figures 1c and 1f present the morphology of rGO-F and rGO-S, which retain the characteristic morphological features of their corresponding GO precursors.

To determine the thickness and number of layers of the GO-F material, transmission electron microscopy (TEM) analysis was performed, as shown in Figure 2. The results indicate that GO-F is a multilayer material, characterized by alternating semi-transparent regions with fewer than 10 layers and darker regions with more than 10 layers.

The Raman spectra of Graphite-foil, GO-F, and rGO-F samples are presented in Figure 3. The pristine graphite foil exhibits a sharp G band centered at approximately 1580 cm^{-1} and a very weak D band near 1350 cm^{-1}, indicating a highly ordered sp^2-hybridized carbon lattice with minimal defects. A distinct and narrow 2D band around 2700 cm^{-1} further confirms the multilayer crystalline structure of graphite.

In contrast, the GO-F spectrum shows a significant increase in the intensity of the D band and a broadening of the G band, leading to a higher I_D/I_G ratio, which reflects the formation of numerous structural defects and oxygen-containing functional groups introduced during the electrochemical oxidation process. The suppression and broadening of the 2D band indicate the disruption of the graphitic domains and the reduction in crystallite size, characteristics typical of graphene oxide. After thermal reduction, the rGO-F sample demonstrates a partially restored sp^2

network, as evidenced by the sharpening and slight redshift of the G band, along with a more discernible 2D band compared to GO-F.

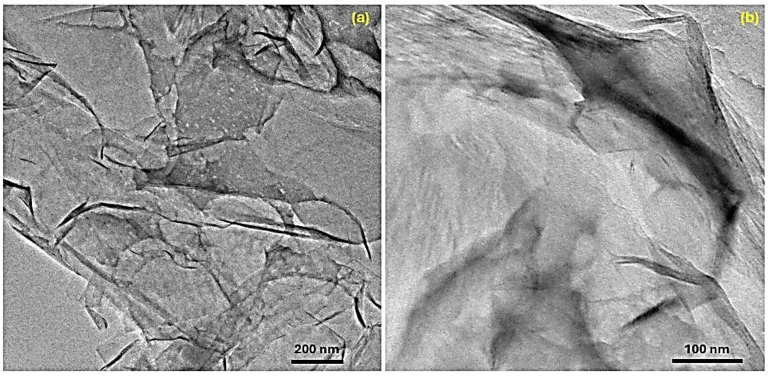


Figure 2. *TEM image of the rGO-F material.*

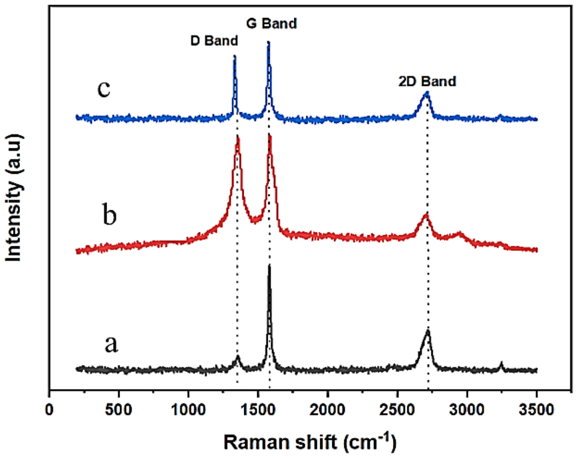


Figure 3. *Raman Spectroscopy of (a) Graphite-foil, (b) GO-F and (c) rGO-F.*

According to the report by Bleu *et al.* [8], the intensity ratio of the 2D band to the G band is closely correlated with the number of graphene layers in the material. Monolayer graphene exhibits an I_{2D}/I_G ratio greater than 1,3, bilayer graphene shows an I_{2D}/I_G ratio in the range of 0,7÷2,2; trilayer graphene has an I_{2D}/I_G ratio of 0,6÷0,7, four-layer graphene presents an I_{2D}/I_G ratio of 0,4÷0,6, and multilayer graphene (5÷10 layers) exhibits an I_{2D}/I_G ratio lower than 0,4. Based on the Raman scattering spectra shown in figure 3, an I_{2D}/I_G ratio of 0,25 was calculated, indicating that the obtained GO consists of approximately 5÷10 layers. The relative intensity ratio of the D band to the G band (I_D/I_G) can be used to evaluate the degree of structural defects in graphene. In this study, the I_D/I_G value of the rGO sample (0,81) is lower than that of the GO sample (0,85), indicating that the reduction process decreases the density of oxygen-containing functional groups and partially restores the conjugated sp^2 network.

The XRD patterns of the GO-F and rGO-F samples (figure 4) reveal distinct differences in their layered structures. The GO-F sample exhibits a broad (002) diffraction peak at 2θ ≈ 25–26°, indicating severe disruption of graphitic order and a low degree of structural ordering due to the presence of abundant defects and oxygen-containing functional groups. This observation is consistent with the Raman results, which show a high D-band intensity and a large I_D/I_G ratio, confirming a high defect density in GO-F. After reduction, the rGO-F sample displays a weakened and further broadened (002) peak, suggesting partial restoration of the graphitic structure while the long-range stacking order remains limited; this interpretation agrees well with the reduced I_D/I_G

ratio and the more pronounced 2D band observed in the Raman spectrum, reflecting partial recovery of the sp^2 carbon network.

These results confirm that thermal reduction effectively removes oxygen functionalities and re-establishes electrical pathways, while retaining a moderate level of defects beneficial for interfacial polarization and electromagnetic interference shielding applications [9].

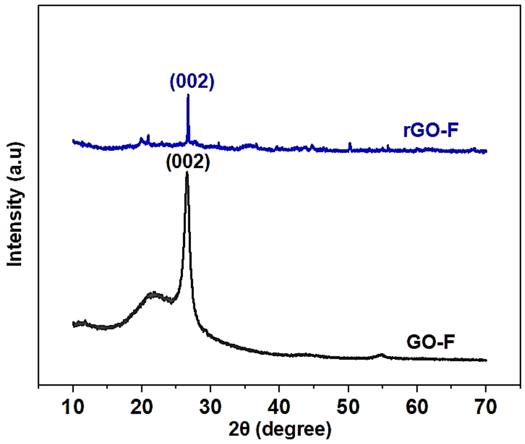


Figure 4. *XRD Spectroscopy of GO-F and rGO-F.*

Consistent with the Raman results, which revealed the increase of oxygen-containing functional groups and structural disorder in GO-F, the FTIR analysis further elucidates the chemical transformation from graphite to GO and subsequently to rGO (Figure 4). The FTIR spectrum of pristine graphite foil shows no significant absorption bands, reflecting a highly ordered sp^2 network (Figure 5). In the case of using H_2SO_4 as the electrolyte, previous studies have demonstrated that oxidation occurs excessively, leading to a high density of defects and oxygen functionalities, which severely disrupt the graphitic sp^2 domains and negatively affect the electrical conductivity of the resulting rGO. In contrast, when $(NH_4)_2SO_4$ is employed, the oxidation process proceeds in a more moderate manner, as evidenced by the characteristic bands at ~3316 cm^{-1} (O–H stretching), ~1634 cm^{-1} (C=O stretching of carbonyl/carboxyl), and ~1122 cm^{-1} (C–O stretching of epoxy/alkoxy). After thermal reduction, these bands are markedly attenuated, confirming the effective removal of oxygen functionalities and the partial restoration of graphitic sp^2 domains. Therefore, the use of $(NH_4)_2SO_4$ as the electrolyte provides better control over the oxidation degree, enabling the preparation of rGO-F with a balanced structure between conductive sp^2 domains and defects, which is highly suitable for applications requiring high electrical conductivity [7].

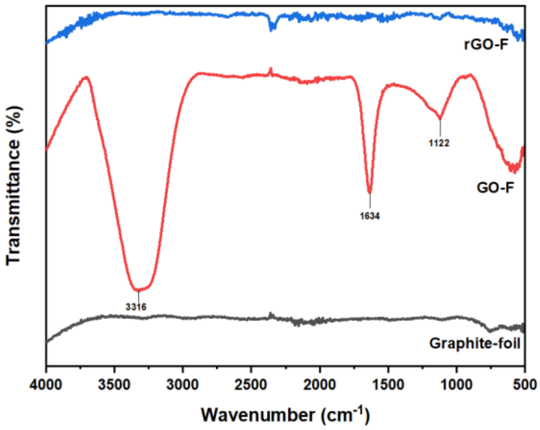
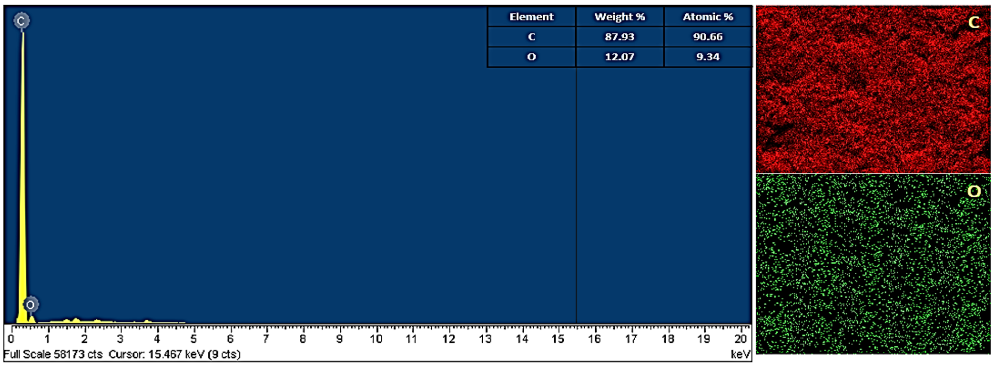


Figure 5. *FTIR spectra of Graphite-foil, GO-F and rGO-F.*

The EDX-mapping results of the rGO-F sample, as presented in Figure 6, show the presence of C Kα and O Kα peaks with a composition of 87,93 wt% carbon and 12,07 wt% oxygen, along with a homogeneous distribution of both elements across the entire surface. This uniform distribution confirms the effective deoxygenation process while retaining a small amount of oxygen functionalities, which provide beneficial defect sites for interfacial polarization. Such characteristics contribute to enhanced electrical conductivity and highlight the potential of rGO-F for high-performance electromagnetic interference (EMI) shielding applications.



Element	Weight %	Atomic %
C	87.93	90.66
O	12.07	9.34

Figure 6. *EDX spectra of rGO-F.*

3.2. Evaluation of the electrical conductivity of the material

The electrical conductivity of graphite foil, GO-F, and rGO-F samples were evaluated from their current-voltage (I-V) characteristics (Figure 7). All samples exhibited linear I-V curves passing through the origin, indicating an ohmic contact behavior. However, the slope of the curves, which reflects the electrical conductivity, varied significantly among the samples. The graphite foil displayed the highest conductivity owing to its highly ordered sp^2 carbon lattice and minimal structural defects (the lowest resistance R=1,79 Ω). In contrast, GO-F showed the lowest conductivity because the introduction of abundant oxygen-containing functional groups (–OH, –COOH, –C=O) during electrochemical oxidation disrupted the delocalized π–electron network, leading to severe carrier scattering (the highest resistance R=29,9 Ω). After thermal reduction at 800 °C, rGO-F exhibited a markedly improved conductivity compared with GO-F, due to the partial restoration of the conjugated sp^2 structure and the removal of most oxygen functionalities (resistance R=14,1 Ω). Nevertheless, residual defects and remaining oxygen atoms limit their conductivity relative to pristine graphite. The intermediate electrical performance of rGO-F suggests a favorable balance between electrical conduction and structural defects, which can enhance interfacial polarization and energy dissipation-beneficial features for electromagnetic interference (EMI) shielding applications [10].

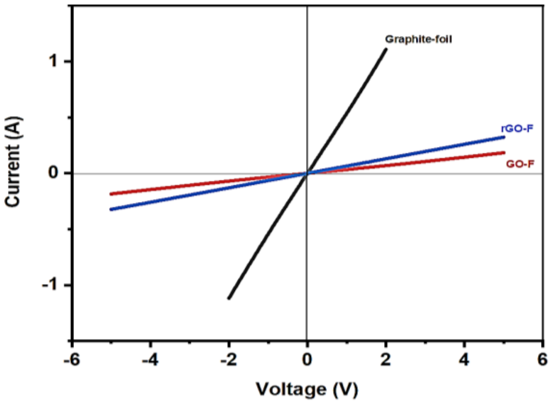


Figure 7. *Comparison of electrical conductivity among graphite foil, GO-F, and rGO-F samples based on current-voltage (I-V) measurements.*

The electromagnetic shielding performance of GO-F and rGO-F in the 8÷12 GHz range is shown in Figure 8 and Table 1. Across the entire X-band, rGO-F exhibits higher transmission loss than GO-F, with S_{21}values of -5.20 to -7.43 dB compared with -2.70 to -3.82 dB for GO-F, indicating stronger attenuation of electromagnetic waves. This trend is consistent with the shielding effectiveness curves in Figure 8, where rGO-F maintains lower SE values throughout the measured frequency range. The enhanced shielding of rGO-F is attributed to its higher electrical conductivity after reduction, which increases induced currents and Joule losses, whereas the lower conductivity of GO-F limits its attenuation capability despite contributions from dielectric polarization.

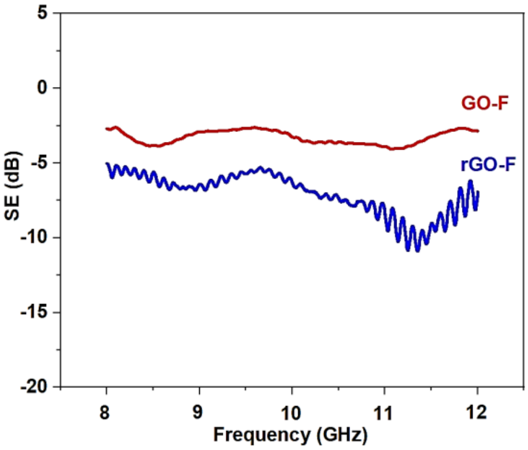


Figure 8. *Electromagnetic shielding effectiveness in the X-band (8÷12 GHz).*

Table 1. *Transmission loss in the frequency range of 8÷12 GHz.*

No	Frequency	Transmission loss, S_{21} [dB]
GO-F	8 GHz	-2,70
	9 GHz	-2,93
	10 GHz	-3,28
	11 GHz	-3,82
	12 GHz	-2,83
rGO-F	8 GHz	-5,20
	9 GHz	-6,45
	10 GHz	-5,50
	11 GHz	-7,43
	12 GHz	-6,82

4. CONCLUSIONS

In this study, graphene oxide (GO) was successfully synthesized via electrochemical exfoliation using graphite foil and graphite sheet electrodes in $(NH_4)_2SO_4$ electrolyte, followed by thermal reduction to obtain reduced graphene oxide (rGO). SEM and TEM analyses revealed that rGO-F derived from graphite foil exhibited thinner and more uniform multilayer sheets (5÷10 layers), while rGO-S obtained from graphite sheet showed thicker and less transparent flakes with significant multilayer stacking. Raman and FTIR spectroscopy confirmed the introduction of oxygen-containing functional groups during electrochemical oxidation and their subsequent removal upon thermal reduction, leading to partial restoration of graphitic sp^2 domains. EDX analysis further demonstrated the effective deoxygenation of rGO-F, with a high C/O ratio and homogeneous elemental distribution. Collectively, these results indicate that electrochemical exfoliation in $(NH_4)_2SO_4$ is a suitable strategy for producing high-quality rGO with a balance of conductive sp² networks and defect sites, making rGO-F a promising candidate for high-performance electromagnetic interference (EMI) shielding applications.

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TÓM TẮT

Nghiên cứu, chế tạo vật liệu graphen oxit khử bằng phương pháp điện hóa, định hướng ứng dụng làm vật liệu che chắn sóng điện từ

Trong nghiên cứu này, oxit graphene (GO) được tổng hợp bằng phương pháp bóc tách điện hóa trong dung dịch chất điện ly $(NH_4)_2SO_4$ *sử dụng hai loại điện cực graphite khác nhau, bao gồm điện cực dạng foil và điện cực dạng tấm, ở điện thế 10 V với mật độ dòng điện được kiểm soát trong thời gian 45 phút. Vật liệu GO thu được sau đó được khử nhiệt trong môi trường khí trơ* N_2 *ở 800 °C trong 1 giờ với tốc độ gia nhiệt 5 °C/phút, tạo thành oxit graphene khử (rGO). Các đặc trưng cấu trúc và hình thái của rGO được phân tích bằng kính hiển vi điện tử quét (SEM), kính hiển vi điện tử truyền qua (TEM), nhiễu xạ tia X (XRD), quang phổ Raman, phổ hồng ngoại biến đổi Fourier (FTIR) và phổ tán xạ năng lượng tia X (EDX). Kết quả cho thấy rGO tổng hợp từ điện cực graphite dạng foil có cấu trúc đa lớp với độ dày tương ứng từ 5 đến 10 lớp graphene. Hình thái này được đánh giá là phù hợp cho định hướng chế tạo vật liệu che chắn sóng điện từ (EMI).*

Từ khóa: Graphen; Graphen oxit khử; Phương pháp điện hóa; Che chắn sóng điện từ.