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Eco-friendly Ball Milling Route for Partially Reduced Graphene Oxide from Natural Coal and Its Electrochemical Diazotization with p-Aminoacetophenone .

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Abstract:

This study included the preparation of a partially reduced graphene oxide (prGO) sheet by grinding coal balls moistened with deionized water droplets. Then, the partially reduced graphene oxide sheet was decorated with para-aminoacetophenone after converting it to diazonium salts by electrochemical preparation using platinum electrodes. The prepared nanocomposites were characterized by spectroscopic methods such as infrared spectra (FT-IR), X-ray diffraction (XRD), scanning electron microscopy (FESEM), atomic force microscopy (AFM), and surface area (BET).

Keywords: coal, graphene, partially reduced graphene oxide, electrochemical decoration of graphene sheets, nanocomposites .

تحضير أوكسيد الكرافين المختزل جزئياً من الفحم الطبيعي بطريقة الطحن الكروي الصديقة للبيئة وتزيين الصفيحة بأملاح الديازونيوم لبارا امينو اسيتوفينون .

علي فاضل أحمد ، غزوان حسن عبد الوهاب ، صفاء محمد رشيد
جامعة تكريت / كلية التربية للعلوم الصرفة - قسم الكيمياء

مستخلص:

تضمنت هذه الدراسة تحضير صفيحة اوكسيد الكرافين المختزل جزئياً عن طريق طحن كرات للفحم المرطب بقطرات من الماء منزوع الأيونات ، بعدها تم تزيين صفيحة اوكسيد الكرافين المختزل جزئياً بمركب بارا امينو اسيتوفينون بعد تحويلة الى املاح الديازونيوم بواسطة التحضير الكهروكيميائي باستخدام اقطاب البلاتين ، وصنفت المركبات النانوية المُحضرة بطرق طيفية مثل أطياف الأشعة تحت الحمراء (FT-IR)، وحيود الأشعة السينية (XRD) ومجهر المسح الإلكتروني (FESEM)، ومجهر القوة الذرية (AFM)، والمساحة السطحية (BET).

الكلمات المفتاحية: الفحم، الكرافين ، اوكسيد الكرافين المختزل جزئيًا، الزخرفة الكهروكيميائية لصفائح الكرافين ، المركبات النانوية.

Introduction

The use of mineral coal has recently been proposed as a low-cost alternative to graphite as a raw material for producing of graphene via chemical exfoliation [3,1] . In addition to its low cost, coal is abundant in many parts of the world. This has led to the development of competitive processes to transform this raw material into a significant resource for the manufacture of new materials [4] . Various grades of coal are abundant and suitable sources for the preparation or conversion into nanomaterials such as graphite, graphene oxide, carbon fibers, and carbon nanoparticles [5,6] . Graphene-based compounds such as graphene oxide and reduced graphene oxide have received considerable attention in recent years. Due to their excellent electrical, mechanical, and thermal properties, they have been considered for use in polymer nanocomposites, energy storage, thin films, and the degradation of organic pollutants [7,8] . Among other applications, several methods have been reported for the production of graphene, such as electrochemical exfoliation [9-11] , arc discharge [12,13] , chemical vapor deposition [14] , and ultrasound [15] .

Experimental

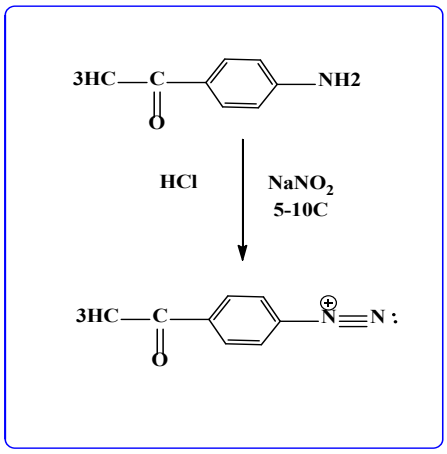
Preparation of Partially Reduced Graphene Oxide Sheets from Natural Coal (prGO)

50 g of natural coal was used and first ground using a mortar and pestle to obtain coal powder. It was then reground using a specially designed nanomill, and humidified with 1ml of deionized water per 10g for ten hours. Partially reduced graphene oxide sheets were obtained from natural coal ground for ten hours (prGO) [16] , as shown in the following reaction scheme:

Scheme (1): Stages of preparation of partially reduced graphene oxide (prGO) nanosheets from mechanical exfoliation of natural coal.

Preparation of diazonium salts

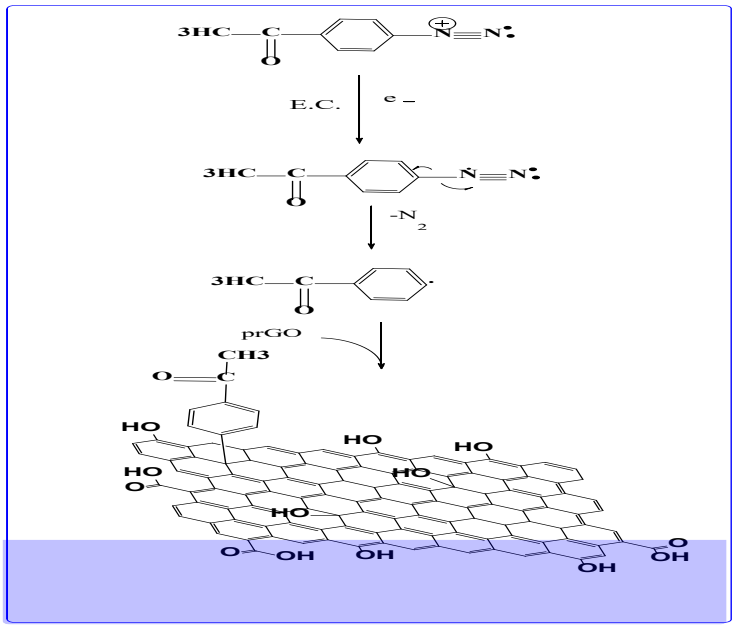
Diazonium compounds were prepared by adding 0.001 mol of different aromatic aniline derivatives to a flask containing an acidic solution (1:1 water: 37% concentrated hydrochloric acid) with the temperature fixed within the range (0-5) °C. In another flask, 0.001 mol of sodium nitrite was dissolved in a suitable amount of distilled water and added to the solution of the first step gradually while keeping the temperature within the range (0-5) °C with stirring for 20-30 minutes in a dark atmosphere. It was not diagnosed but was used directly in the next step [17] . As shown in the following reaction scheme:



Scheme (2): Preparation of diazonium salts of para-aminoacetophenone.

Decorating a partially reduced graphene oxide sheet with para-aminoacetophenone

0.3 g of prGO was placed in 25 mL of water in ultrasonic bath until the solution became clear. This solution was then added to an electrolytic cell containing platinum electrodes at a voltage of 1.6 V under stirring. Diazonium salt solution was then added to the prGO solution in droplets under continuous stirring for 18 hours [18]. As shown in the following reaction scheme :



Scheme (3): Electrochemical decoration of partially reduced graphene oxide sheet with diazonium salt.

Preparation of prGOCh$_2$ from Ar-prGO

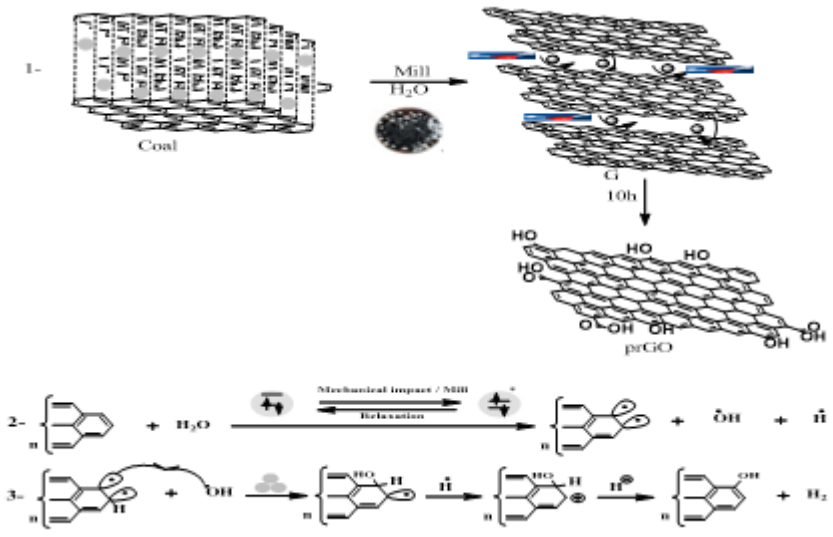
(0.4g) of Ar-prGO dissolved in (10 mL) of methanol was sonicated for 40 min with (0.4 g) of suitable benzaldehydes dissolved in (10 mL) of methanol, to which (10 mL) of 10% sodium hydroxide was added and the mixture was stirred for 24 h at 40°C. After the reaction was completed, the mixture was cooled and the resulting precipitate was filtered and washed with distilled water [19].

Results and Discussion

Compound: Partially reduced graphene oxide (prGO)

The mechanical exfoliation of natural coal is of the free radical type. When the highly ordered coal (carbon aggregate) is ground with strong rollers, shear, collision, and frictional forces occur at the microscopic level. These forces separate the stacked layers of the graphite structure, and the nodes and cracks open, resulting in mechanical exfoliation of thin sheets (graphene-like nanoplatelets) [20] . If a catalyst (water) is introduced during the grinding, oxygen functional groups such as OH, COOH, and C=O are formed on the edges and surfaces, and we obtain graphene oxide (rGO)

or similar materials [21]. Free radicals are formed during grinding. Initially, the C-C and C-H bonds in the carbon structure of natural coal are broken. During strong collisions between the steel grinding balls and the molecules, very high energy is transferred, and the chemical bonds between the carbons are broken, revealing unsaturated sites or active carbon centers. Vacant sites and exposed edges are then formed. These edges contain electron-unsaturated carbon atoms, which act as free radicals. When deionized water is added to the reaction medium, the free radicals react with the added water, forming oxygen groups (OH, COOH, and C=O), leading to the formation of a partially reduced graphene oxide sheet. Free radicals may rearrange the carbon structure to form sp^2 bonds. High-pressure collisions lead to the formation of electrons, and these electrons lead to the separation of sigma or pi bonds in both materials that have been ground, and temporary free radicals are formed on the surface of the graphene and the added material as a result of the two materials being hammered together, so a mutual exchange of free radicals occurs and the bonding occurs.



Scheme (4): Mechanism of preparation of partially reduced graphene oxide (pGO) nanosheets from mechanical exfoliation of coal.

FTIR spectrum diagnosis of partially reduced graphene oxide (prGO) sheet

The FT-IR spectrum showed absorption bands associated with the main active groups in the partially reduced graphene oxide (prGO) structure. The absorption band of the phenolic (O-H) bond stretching at frequencies (1369 cm) and an absorption band at frequencies (1595-1540 cm) are attributed to the C=C and N-H bending. This frequency indicates the presence of carbon-carbon (C=C) double bonds. Absorption bands at frequencies (1095-1010 cm) are attributed to Si-O, and absorption bands at frequencies (500-700 cm) are attributed to the Si-O or C-O bending [22] . The following figure (1) shows the FT-IR spectrum of the partially reduced graphene oxide (prGO) sheet.

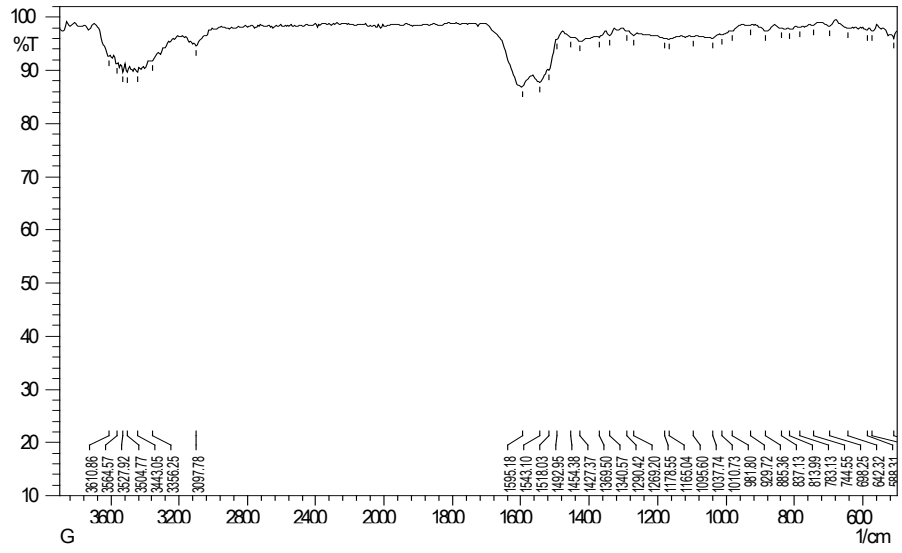


Figure (1): FT-IR spectrum of partially reduced graphene oxide (prGO) nanosheets from mechanical exfoliation of natural coal.

diagnosis XRD to determine the crystal structure of prGO (partially reduced graphite oxide)

The angles 23.44° and 23.50°, with a distance between layers d-spacing 3.79 Å and relative intensity 100% and 50% respectively, show a typical pattern for the presence of a semi-crystalline carbon structure, such as silica or carbon nanotubes.

The angles are 27.06° and 27.13°, with a distance between layers d-spacing 3.29 Å and relative intensities of 76% and 38% respectively. These values indicate the presence of nanocrystals in the material, such as graphene .

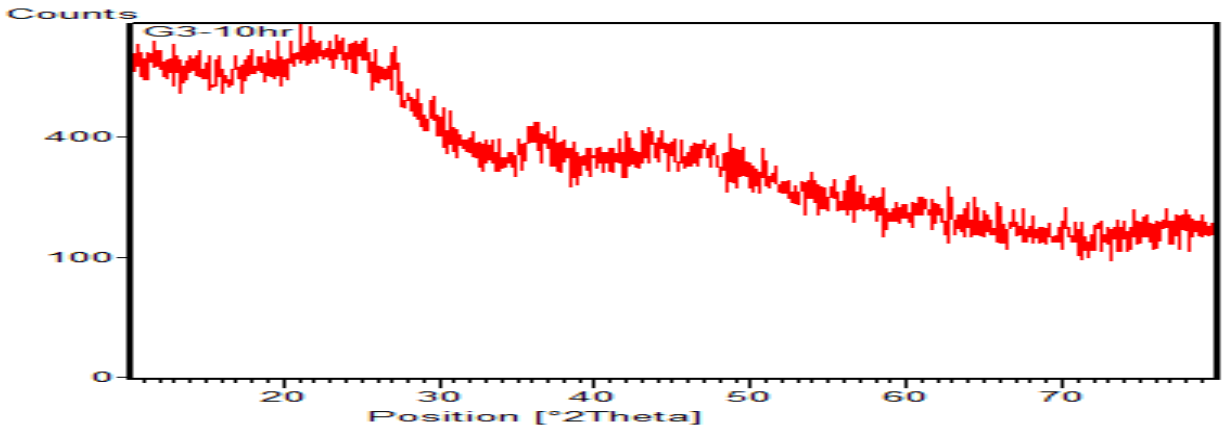


Figure (2) X-ray diffraction (XRD) spectrum of partially reduced graphene oxide sheet.

Scanning Electron Microscope (SEM)

Observing the morphological SEM images of the partially reduced graphene oxide (prGO) sheet, shown in Figure (3), reveals the following:

1. Figure (SEM-a) indicates a large surface area, measured by BET surface area spectroscopy, of 265.98 m^2/g.

2. Figure (SEM-b) shows an evenly distributed porous structure with varying pore sizes, indicating a porous system that allows for good water and chemical adsorption.

3. Figure (SEM-c) shows a network of medium-sized nanopores.

4. Figure (SEM-d) demonstrates the presence of fine nanopores, which enhances microscopic cohesion and reduces the likelihood of microcrack formation.

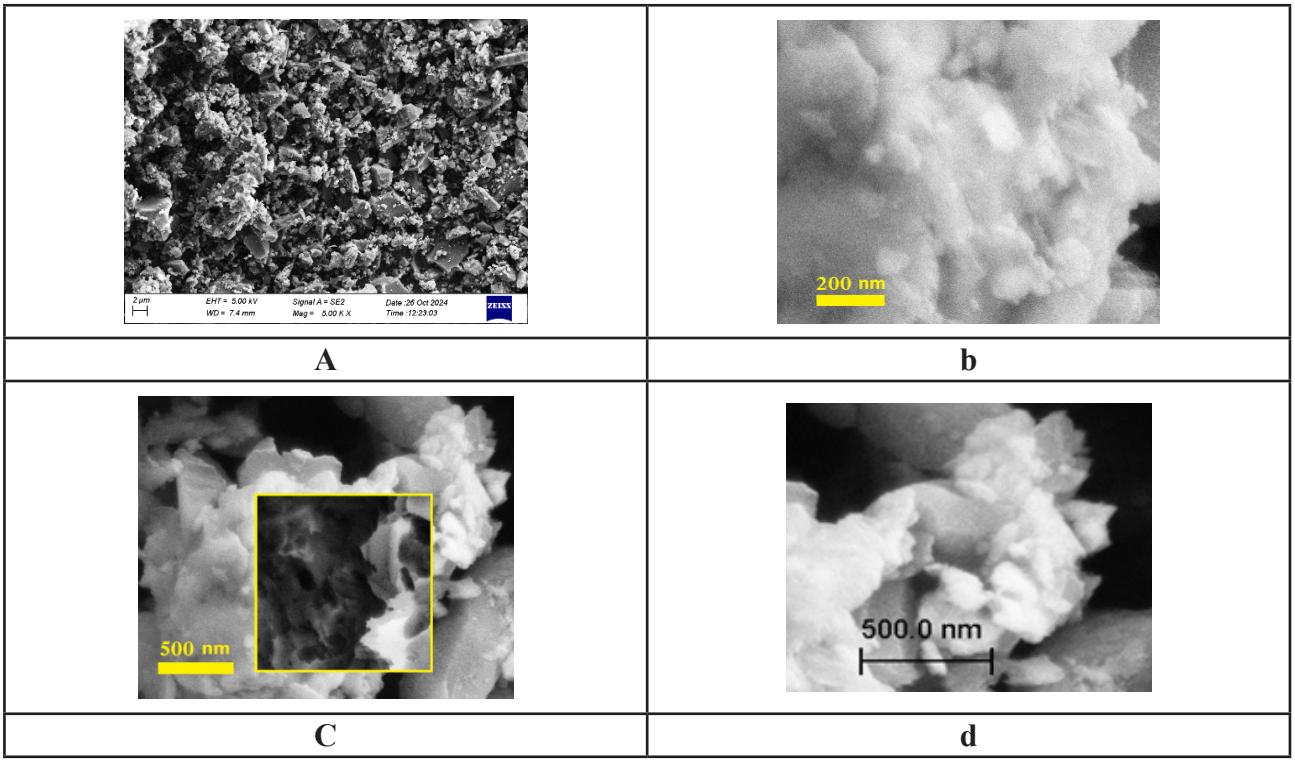


Figure (3): SEM spectrum of partially reduced graphene oxide (prGO) sheet prepared from mechanical exfoliation of natural coal.

Diagnosis of atomic force microscope (AFM) data for nanoscale surface properties

AFM images of partially reduced graphene oxide sheets prepared from grinding coal for over ten hours, shown in Figure (4), show the following:

1. Average nanometer height (Sa): The value reached 4.461 nm. This indicates that the surface has a moderate nano-topography, as shown in Figure (AFM-a), which shows a smooth and moderate distribution of nano-heights on the material's surface.

2. Root mean square height (Sq): 5.944 nm, indicating no excessive surface roughness. Figure (AFM-b) depicts the nano-features as three-dimensional topographic maps, showing a regular distribution of heights and depressions without sharp protrusions.

3. Skewness (Ssk): The skew is positive (+1.359), indicating that the surface is asymmetric, with more peaks than pits. Figure (AFM-c) shows that peaks partially dominate the material's surface compared to pits, reflecting some surface pattern rich in active

contact points.

4. Surface area ratio (Sdr) of 0.2745% indicates that the surface has sufficient complexity to promote interaction between the material and organic molecules. Figure (AFM-d) shows fine branching on the material's surface, indicating an increase in structural complexity.

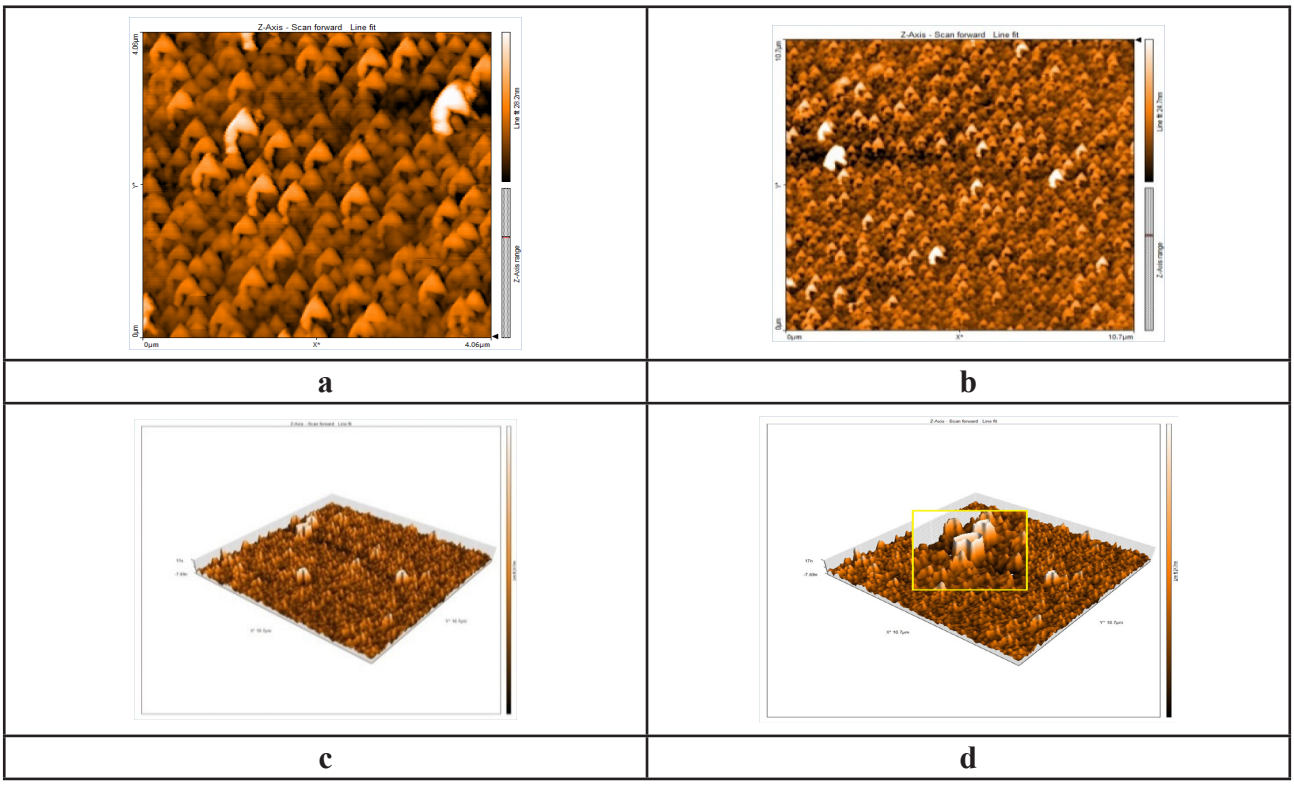
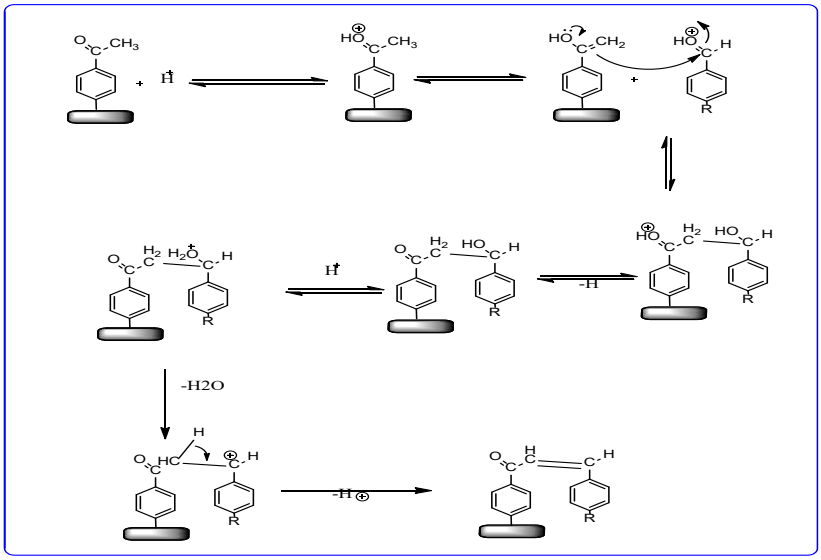


Figure (4): Atomic force microscope (AFM) spectrum of partially reduced graphene oxide (prGO) sheet prepared from mechanical exfoliation of natural coal.

Compounds: Graphene oxide sheet decorated with para-aminoacetophenone

The mechanism of the electroreduction reaction is a free radical mechanism and electrophilic substitution on the surface of partially reduced graphene oxide. When the diazonium salt is placed in an electrolytic cell and an electrical voltage is applied, it will transform into a chemically active free radical. The partially reduced graphene oxide sheet contains electron-rich regions, namely C=C and C-OH bonds, on the sheet lattice. The acetophenone radical attacks these sites via radical

addition to the C=C bond, as shown in the steps for decorating the reduced graphene oxide sheet with acetophenone.



Scheme (5): Proposed mechanism for the preparation of (prGO-Ch$_2$).

FT-IR spectrum of the Ar-prGO composite

The FT-IR spectrum showed absorption bands associated with the main active groups in the Ar-prGO structure. The absorption band for stretching the phenolic (O-H) bond appeared at the wavenumber range of 3412 cm-1, and the absorption band for stretching the (C-H) bond in the aromatic ring structure appeared at the wavenumber of 3089 cm-1. Meanwhile, an absorption band associated with stretching the aliphatic (C-H) bond appeared, and an absorption band at the wavenumber of 1666 cm-1 associated with the (C=O) group. The decrease in the carbonyl band in the aromatic ketones indicates that the C=O bond is affected by the bonding with the surface of the partially reduced graphene oxide sheet or by the resonance effect after covalent bonding with the surface of the prGO sheet. Three absorption bands range Its values are from (1452-1583 cm-1) which are due to the vibration of the aromatic (C = C) bond stretching, and absorption bands at the frequency of (11095-1010 cm-1) are due to Si-O and absorption bands at the frequency

(500-700 cm-1) are attributed to the bending of Si-O or C-O. The following figure (5) shows the FT-IR spectrum of the ((Ar-prGO) compound .

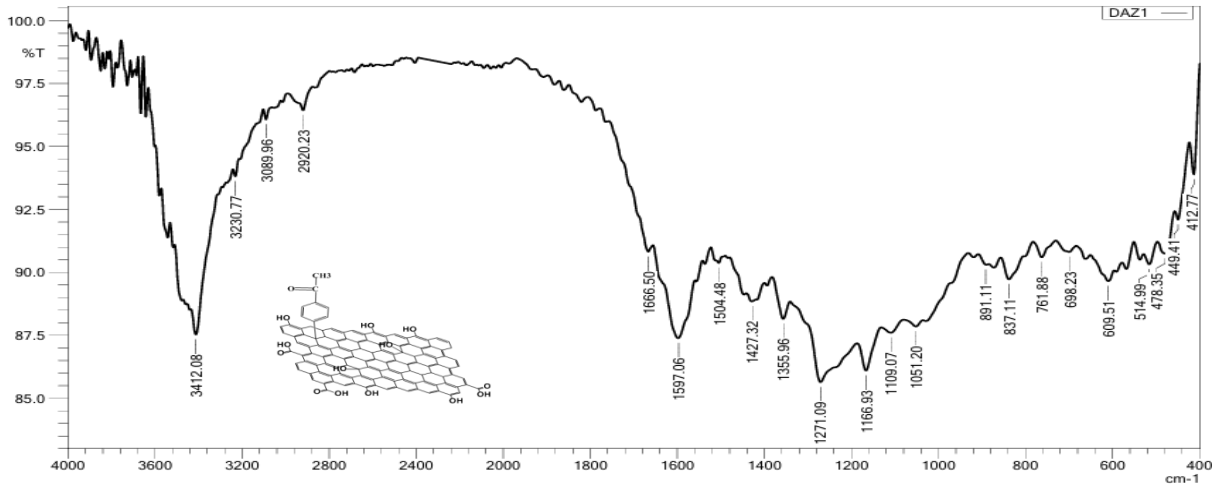


Figure (5): FT-IR spectrum of the prepared (Ar-prGO) compound.

XRD spectrum of the Ar-prGO composite

The XRD spectrum of the prepared Ar-prGO composite showed angle values of (2Ɵ) at 32.4431 and 46.0505 with a grain size of D=149.9 number of layers n=76.16, and spacing between them d=1.96, as shown in Figure (6).

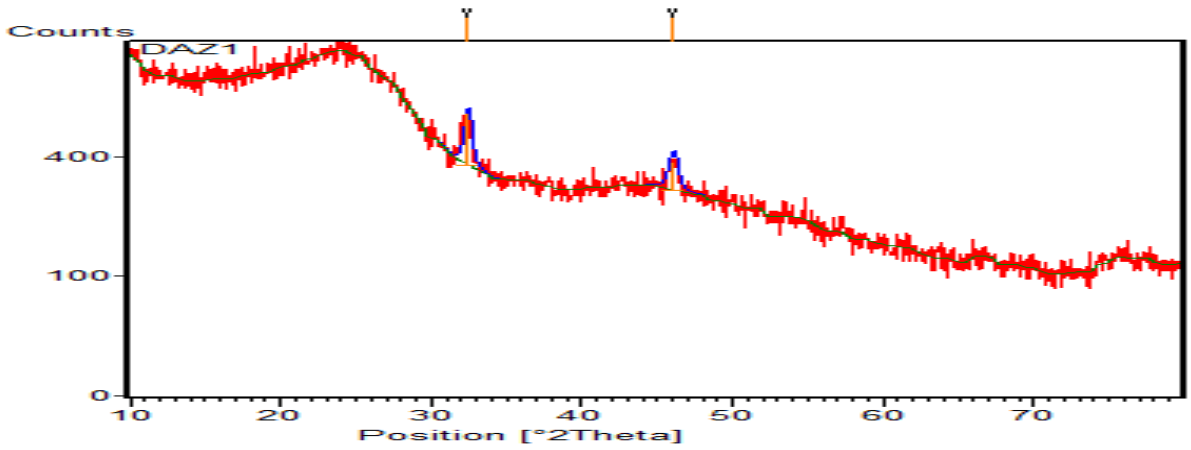


Figure (6): XRD spectrum of the prepared (Ar-prGO) composite.

Scanning Electron Microscope (SEM) of the Ar-prGO Composite

Observing the morphological SEM images of the prepared Ar-prGO composite, shown in Figure (7), it is evident that:

1. Figure (SEM-a) shows the surface covered with a dense, uniformly distributed nanodots, in addition to some larger clusters. This reflects the presence of a thin layer of organic molecules bound to the surface via the electrochemical reaction of diazonium salts. The fine dots can be attributed to covalent anchoring sites spread across the surface, while the larger clusters reflect an increased decoration density.

2. Figure (SEM-b) shows thin, wrinkled prGO sheets with folded edges. Acetylated acetophenone molecules are concentrated at the edges due to increased chemical activity and the absence of large cracks. This indicates the integrity of the prGO after decoration.

3. Figure (SEM-c) shows a wider perspective, showing the partially reduced graphene oxide sheet covered with irregularly distributed, hollow particles. This indicates that the decoration process was not uniform, but rather focused on... The chemically active sites on the prGO surface are located. This distribution is a natural feature of diazonium reactions on graphene and its oxides, where the reactions are concentrated at the active sites, leading to variations in the decoration density.

4. The SEM-d image shows the general morphology of the prGO sheets, which appear as overlapping, broken flakes with dimensions between 0.5 and 2 microns. Small, irregular particles can be observed distributed on the surfaces and edges. These particles reflect the immobilization of organic groups on the sheet surface after the electrochemical decoration process, indicating the success of the surface decoration process for the prGO sheet compared to undecorated samples, which typically exhibit relatively smoother and more homogeneous sheets.

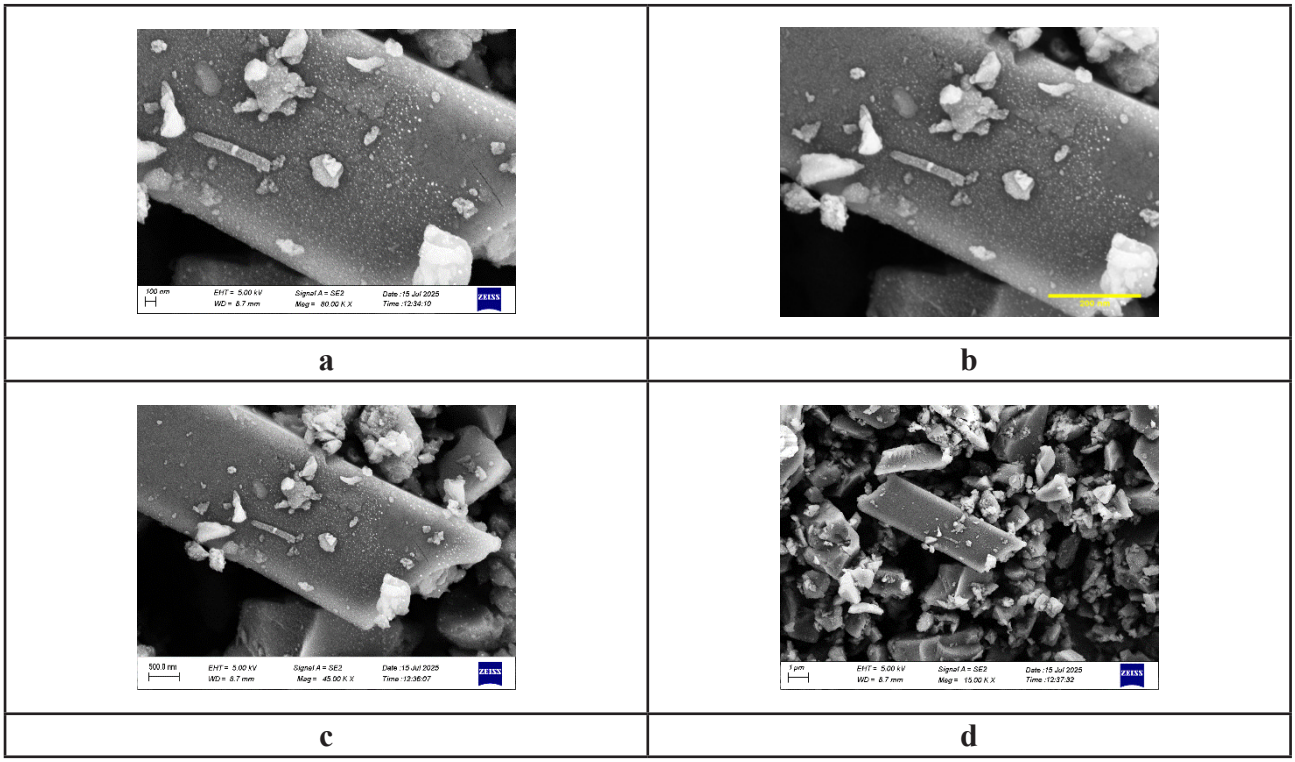


Figure (7): SEM spectrum of the prepared (Ar-prGO) composite

Atomic Force Microscopy (AFM) of the Ar-prGO composite

The AFM images of the prepared Ar-prGO composite [23] , shown in Figure (8), show the following:

1. Average nanometer height (Sa): The value reached 37.54 nm, indicating electrochemical decoration of the prGO sheet surface, and that the reaction occurred on the sheet surface (image AFM-a).

2. The root mean square height (Sq) was 48.91 nm, indicating that the surface is rough due to electrochemical decoration of the prGO sheet surface. Image AFM-b shows nanoscale features in the form of three-dimensional topographic maps, showing a regular distribution of peaks and valleys without sharp protrusions.

3. Skewness (Ssk): The skewness is positive (+1.010), indicating that the surface is asymmetric, with more peaks than pits. Image (AFM-c) shows that the peaks partially dominate the surface of the material compared to the pits, reflecting a surface pattern rich in active contact points.

4. Surface area ratio (Sdr): 3.598%, indicating that the surface has enhanced

covalent interactions between the sheet surface and the organic molecules. Image (AFM-d) shows the presence of slightly thickened, elevated peaks.

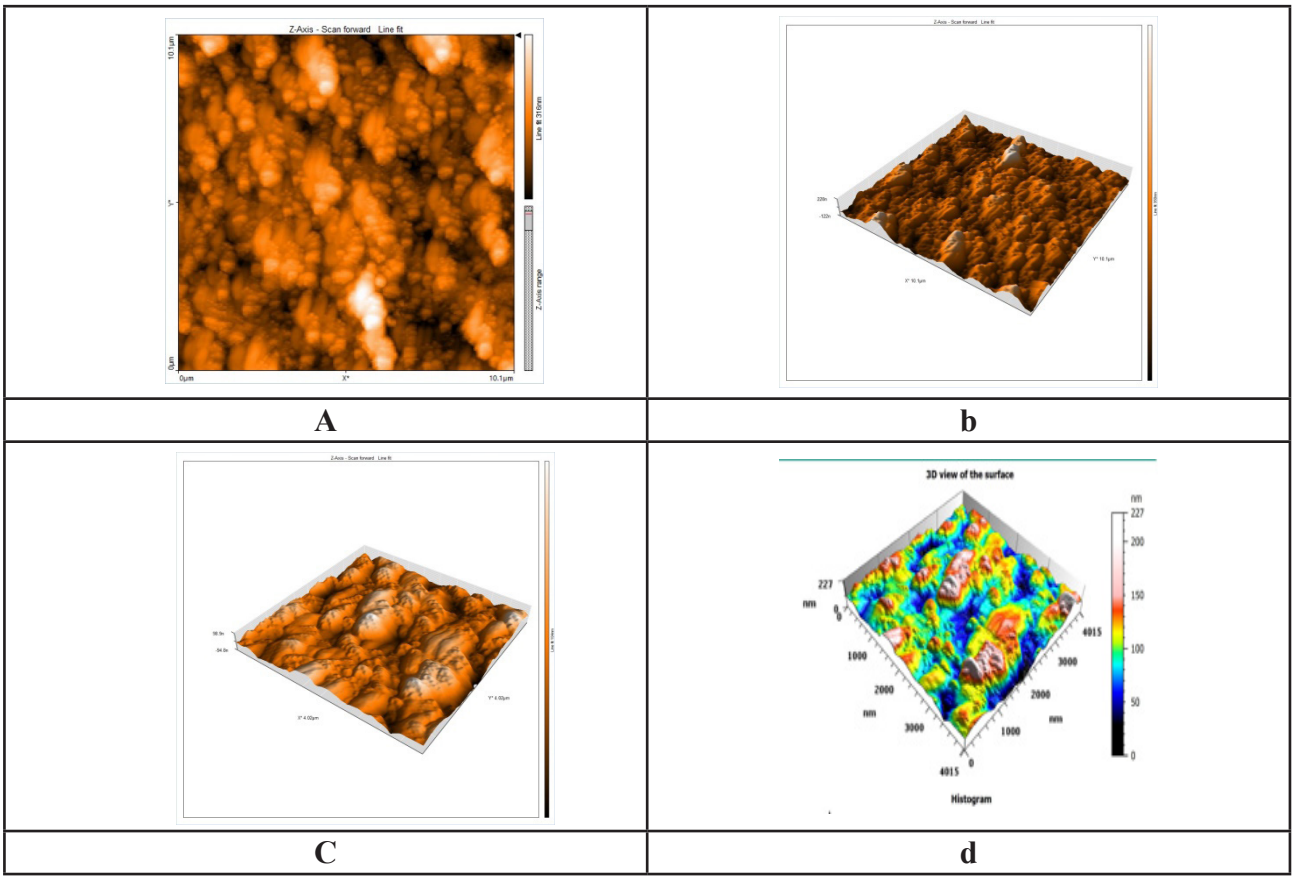


Figure (8): Atomic force microscope (AFM) spectrum of the prepared (Ar-prGO) composite.

The surface area of the prepared Ar-prGO composite was calculated and can be illustrated as follows

BET surface area of the Ar-prGO composite

- Specific surface area (as, BET): 74.999 m^2/g
- Langmuir surface area (SA): 108.81 m^2/g
- Total pore volume (TPV): 0.1203 cm^3/g
- Average pore diameter (APD): 6.41 nm, reflecting an ideal (mesoporous) nanostructure that enables physical and chemical interactions with other compounds.
- Pore diameter according to the BJH method (BJH pore diameter rp): 1.72

nm, indicating the presence of small pores indicative of a nanostructure.

The reaction of chalcones between partially reduced graphene oxide sheets decorated with acetophenone and benzaldehydes

FT-IR spectrum of the (pr-GO-Ch2) complex

The FT-IR spectrum showed absorption bands associated with the main active groups in the (Ar-prGO) structure. The absorption band for the stretching of the phenolic (O-H) bond appeared at the wavenumber range of 3423 cm^{-1}, and the absorption band for the stretching of the (C-H) bond in the aromatic ring structure appeared at the wavenumber of 3089 cm-1. Meanwhile, an absorption band associated with the stretching of the aliphatic (C-H) bond appeared, and an absorption band at the wavenumber of 1654 cm^{-1} associated with the (C=O) group appeared, with the carbonyl band descending in the chalcone. Three absorption bands with values ranging from (1429 cm^{-1}) to (1654 cm^{-1}) appeared. 1544) which is due to the vibration of the aromatic (C = C) bond, and absorption bands at the frequency of (11095-1010 cm^{-1}) are due to Si-O and absorption bands at the frequency (500-700 cm^{-1}) are attributed to the bending of Si-O or C-O. The following figure (9) shows the FT-IR spectrum of the compound (pr-GO-Ch2).

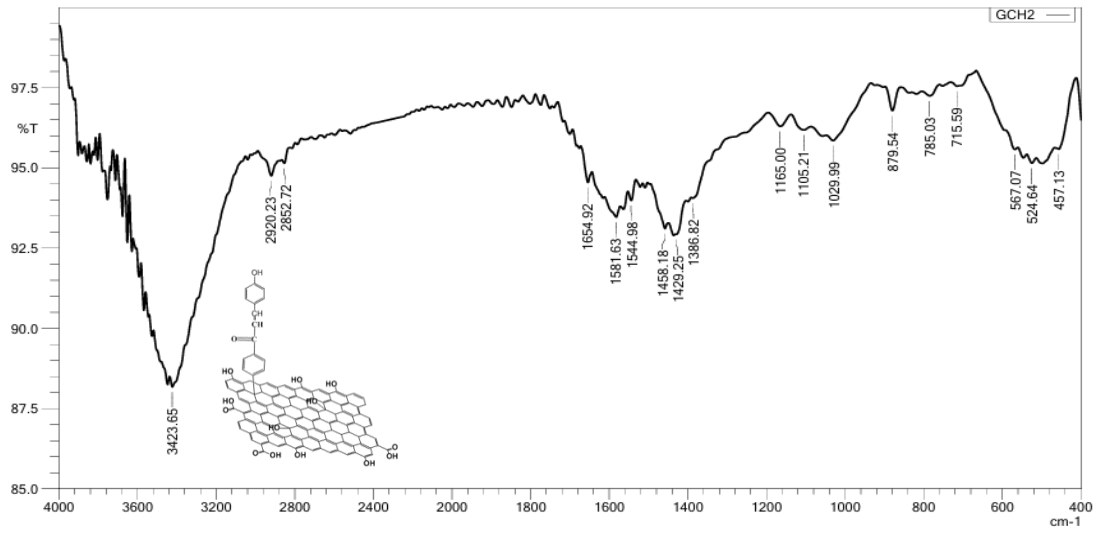


Figure (9): FT-IR spectrum of the (prGO-Ch2) compound.

XRD spectrum of the (prGO-Ch2) composite

The XRD spectrum of the prepared (prGO-Ch2) composite showed angle values of (2Θ) at 27.8209, 30.3331, 37.9665, 40.0331, 46.8512, and 48.4924 with a grain size of D= 167 , a layer number of n= 52 , and an interlayer spacing of d= 3.20 as shown in Figure (10).

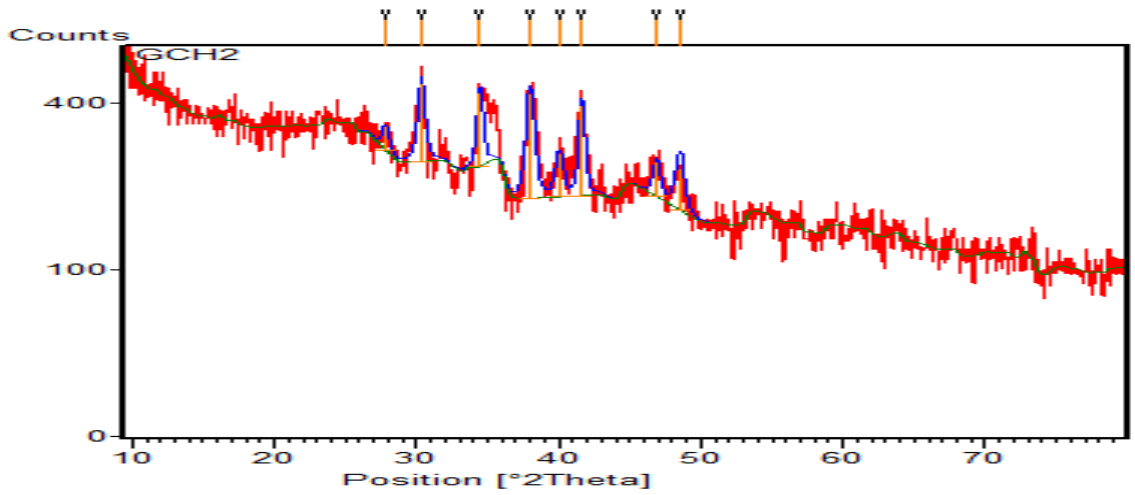


Figure (10): XRD spectrum of the (prGO-Ch2) compound.

Scanning Electron Microscope (SEM) (prGO-Ch2)

Observing the morphological SEM images of the partially reduced graphene oxide (prGO-Ch2) sheet, shown in Figure (11), reveals the following:

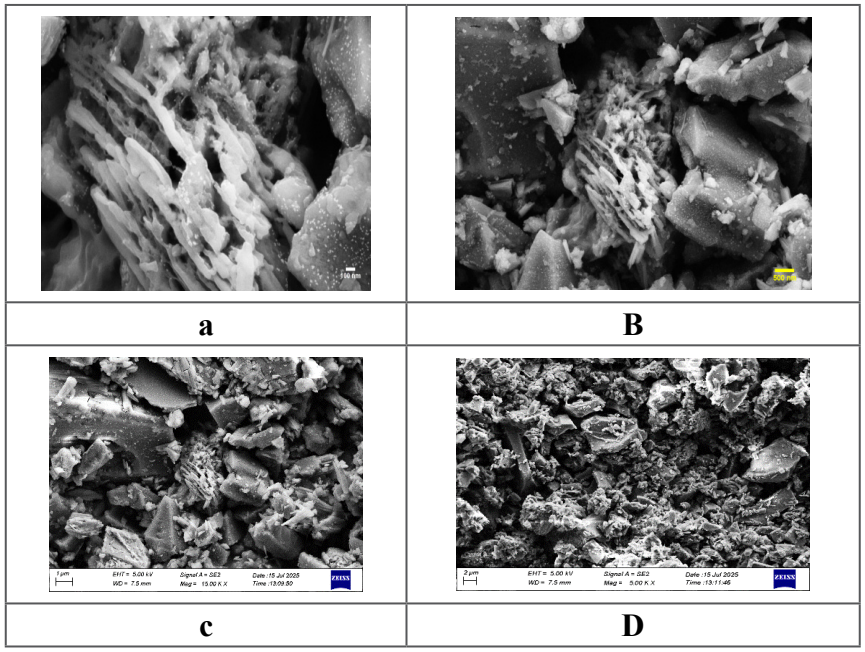
1. Figure (SEM-a) indicates a large surface area, measured by BET surface area spectroscopy, of 117.54 m^2/g.

2. Figure (SEM-b) shows an evenly distributed porous structure with varying pore sizes, indicating a porous system that allows for good adsorption of water and chemicals.

3. Figure (SEM-c) shows a network of medium-sized nanopores

4. Figure (SEM-d) demonstrates the presence of fine nanopores, which enhances microscopic cohesion and reduces the likelihood of microcrack formation.

Figure (11): Scanning electron microscope (SEM) (prGO-Ch2).



Atomic Force Microscope (AFM) of the (prGO-Ch2) composite:

The AFM images of the prepared (Ar-prGO) composite, shown in Figure (12), showed the following:

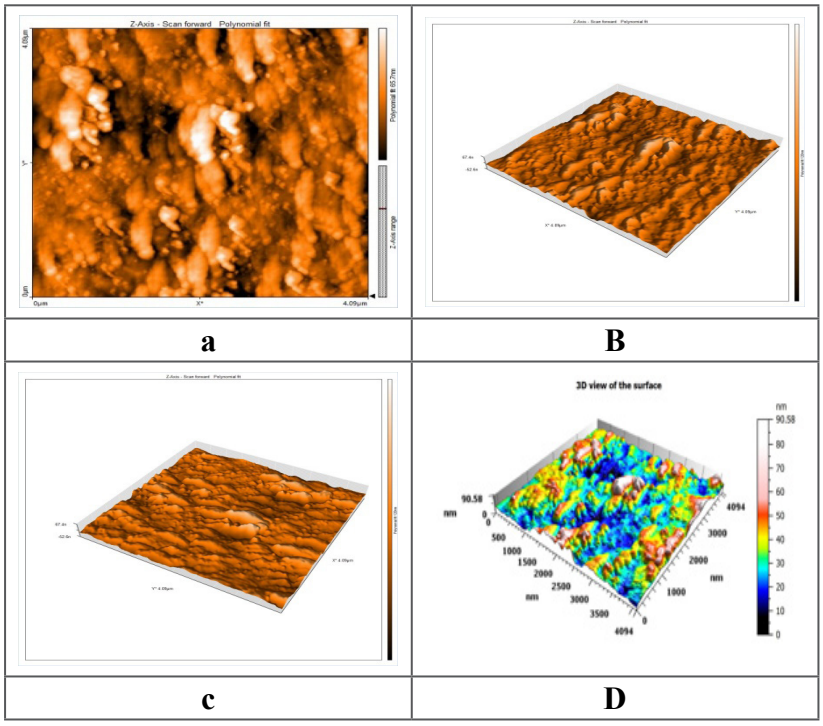
1. Average nanometer height (Sa): 12.83 nm

2. Root mean square height (Sq): 18.60 nm

3. Skewness (Ssk): Positive (+2.392)

4. Surface area ratio (Sdr): 1.223%, indicating that the surface enhanced the covalent interaction between the sheet surface and the organic molecules.

Figure (12): Atomic force microscope (AFM) of the ((prGO-Ch2) compound.



The surface area of the prepared (prGO-Ch2) composite was calculated and can be illustrated as follows

BET Surface Area :

• Specific Surface Area (as, BET): 117.54 m^2/g, indicating a lower surface area than that of prGO, meaning that the decoration occurred on a larger area of the sheet.

• Langmuir Surface Area (SA): 167.86 m^2/g, supporting the previous result with a higher index.

• Total Pore Volume (TPV): 0.1252 cm^3/g, indicating the presence of a porous system suitable for decoration reactions on the sheet surface.

• Average Pore Diameter (APD): 4.26 nm.

Conclusions

Partially reduced graphene oxide (prGO) sheets were prepared by ball milling, a physical method from natural coal. Traditional methods for preparing graphene oxide compounds, including the modified Hammer method, Staudenmayer method, and Tor method, which use environmentally harmful chemicals and strong oxidizing agents, were avoided. Therefore, the mechanical grinding method was used, which is considered a safe, green, and fast method of preparation compared to common traditional methods. After that, the partially reduced graphene oxide sheet was decorated with para-aminoacetophenone compound after converting the latter to diazonium salt and placing it in an electric cell, placing platinum electrodes, applying a voltage of 1.6 V, and decorating the partially reduced graphene oxide sheet. The accuracy and precision of the prepared nanocomposites (prGO, Ar-prGO, prGO-Ch2) were verified using spectroscopic measurements using infrared spectra (FT-IR), X-ray diffraction (XRD), scanning electron microscope (FESEM), and microscope. Atomic force microscopy (AFM) and surface area (BET) spectroscopic results have shown that ball milling of natural coal using deionized water has given excellent results in obtaining partially reduced graphene oxide sheets which can be used in many applications including corrosion, as electrical conductors, adsorption, pharmaceutical delivery, cement mortar and other applications. Nanostructures resembling sheets have

been prepared resulting from the covalent interaction of aromatic systems with partially reduced graphene oxide sheets.

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