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THE COATING OF REDUCED GRAPHENE OXIDE (rGO): A NOVEL ULTRASONIC-ASSISTED METHOD

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Abstract. *The graphene is one of the most popular materials of our age since its discovery. The graphene and its derivatives have gained much attention in sensor applications because of its features (e.g., electronic conductivity, specific surface area, etc.). However, the coating of graphene is challenging for the researchers especially for Si/SiO2 surfaces due to its surface tension. Many researchers tend to use chemical materials for the coating rGO onto Si/SiO2 such as APTES, TEOS, PEG, HMDS etc. For the purpose, we discovered a novel type ultrasonic-assisted coating method for sensor applications which can be done using any chemicals. To do so, we firstly produced reduced graphene oxide (rGO) from graphite by using Hummer's method and chemical reduction process. Then, we prepared Si/SiO2 samples and put them into plastic container. After that, we put samples into ultrasonic bath and dropped rGO suspension onto samples by using with micro-pipette. After that, the rGO coated samples were dried on hot plate at 100°C. The results showed high potential that rGO can be coated onto Si/SiO2 surfaces with low-cost solution.*

Keywords: *graphene, reduced graphene oxide, coating, ultrasonic-assisted, novel method, Si/SiO2*

1. INTRODUCTION

As a wonder material of our age, the graphene is first discovered and isolated in 2004 by scientists from Manchester University [1]. Its structure consists of sp^2 bonded carbon atoms with honeycomb structure [2]. Since then, the scientists have been trying to produce large scale and high yield graphene for different applications. Especially, nowadays studies have been centered graphene because of its high thermal conductivity, high surface-volume area, high electrical conductivity etc. [3] These features of graphene provide applications in many research areas such as sensor applications, drug studies, environmental monitoring, water quality management and medical applications [3].

One of the most faced challenges is that the coating of graphene for its applications. Many researchers have been trying to figure out defect free coating or transfer of graphene and its derivatives. To solve this problem, the researchers have investigating different kinds of surface functionalization materials such as (3-Aminopropyl) triethoxysilane (APTES) [4], Tetraethyl orthosilicate (TEOS) [5], Poly (Ethylene Glycol) (PEG) [6], Hexamethyldisilane (HMDS) [7]. However, the effects of using of this kind of chemicals is not fully understood. Also, the influences of this materials on biomolecules are not effectively understood for biosensor applications.

To solve the problem, we discovered novel ultrasonic assisted coating method. With this methods, different kind of chemical and equipment usage is prevented. The detailed information is given in the next sections about coating and characterization.

2. THE SYNTHESIS OF REDUCED GRAPHENE OXIDE

2.1. Materials

The graphite powder, L-ascorbic acid, potassium permanganate, sulfuric acid (%96) and hydrogen peroxide were purchased from Sigma-Aldrich (Merck). The whole process was done in 100-1000 class cleanrooms in Nuclear Radiation Detectors Application and Research Center (NÜRDAM) Facility.

2.2. Synthesis of Reduced Graphene Oxide (rGO)

The modified Hummer's Method were used to form graphene oxide from graphite material [8]. As oxidation material, potassium permanganate was used. The mixture that consists sulfuric acid and graphite powder were stirred for three hours at 600 rpm. Then, hydrogen peroxide was used for neutralization agent. In the end of processes, graphene oxide was synthesized, respectively.

For synthesizing of reduced graphene oxide, chemical reduction method was preferred. Graphene oxide and L-ascorbic acid were mixture in glass hardware with DI water and was stirred for 24 hours. After, the mixture was filtered and left to dry for 24 hours at room temperature.

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3. The Coating Method of Reduced Graphene Oxide

The coating of reduced graphene oxide has been challenging for scientist for applications. According to literature, there were many coating types as dip coating [9], spin coating [10], spray coating [11], anodic exfoliation method [12] etc. On the other, these kind of coating techniques require different equipment and chemical processes. For the purpose, we found a low-cost solution for coating. At the beginning, we prepared the FET chip for coating. To do so, we first covered the coating area with Kapton (polyimide) tape (Figure 1). The purpose of using Kapton tape was to prevent spreading of rGO dispersion to other areas.

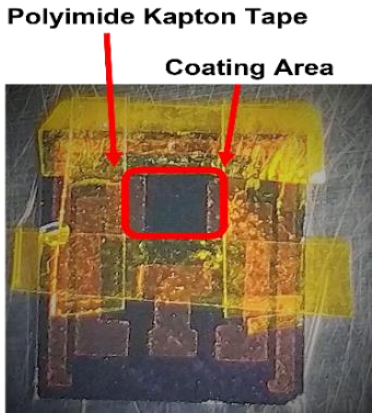

Figure 1. The prepared sample for coating reduced graphene oxide.

Then, we put samples onto plastic container and placed into ultrasonic bath. After that, we pipetted the rGO dispersion with micro pipette as 5 µl. The next step was drop casting of rGO onto coating area. After dropping rGO dispersion, we waited for 5 minutes for the dry of mixture (Figure 2).

Figure 2. The prepared sample for coating reduced graphene oxide.

The last step of coating rGO is annealing of samples on hot plate. In this step, the complete dryness of rGO layer was conducted. When first samples put onto hot plate, the temperature was set to 50°C. And then, the temperature of hot plate gradually (10°C with every 5 minutes) increased until 100°C. After complete dryness of reduced graphene oxide layer, Kapton tapes were removed on samples carefully with help of tweezers.

Figure 3. The prepared sample for coating reduced graphene oxide.

Accordingly, we simplified the coating of reduced graphene oxide with novel type ultrasonic-assisted method. The main advantage of this coating method was reducing cost, avoiding usage of different equipment, and avoiding usage of chemicals mentioned in introduction part.

4. The Characterization of Reduced Graphene Oxide

After completion of reduced graphene oxide coating, the characterizing analyses were done by X-Ray Diffraction (XRD) and Fourier Transform Infrared Spectroscopy (FTIR).

4.1. The X-Ray Diffraction (XRD) Analysis

Figure 4 shows XRD result of graphite, graphene oxide and reduced graphene oxide after coating. The one sharp peak at 25.72° was observed for graphite material as primary material for synthesis reduced graphene oxide. After oxidation of graphite, the shift was observed and towards 9.84° because of oxygen containing groups [13]. After reduction of graphene oxide, the peak was observed shift back to 25.18°. This means that, the graphite material was successfully oxidized and then successfully reduced to reduced graphene oxide by removing hydroxyl groups (-OH). The similar shift amounts were compatible with the literature for graphite, graphene oxide and reduced graphene oxide [14–16]. Also, the peak at 44° was labeled as amorphous hexagonal carbon peak for rGO [17].

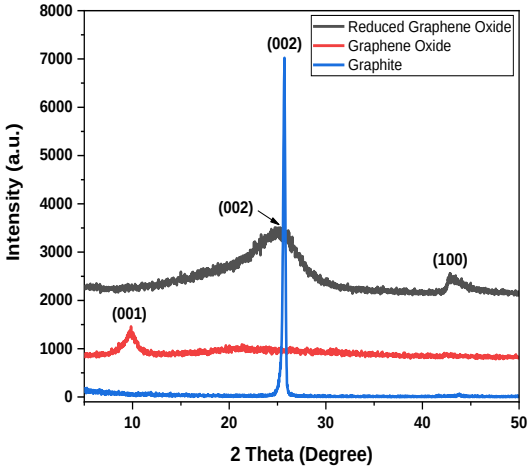

Figure 4. The XRD pattern of graphite, Graphene oxide and reduced Graphene oxide.

4.2. Fourier Transform Infrared (FTIR) Analysis

The synthesized graphene oxide and reduced graphene oxide's FTIR spectra were given in Figure 5 and Figure 6. In both figures, the peaks around at 1050 cm^{-1}, 1550 cm-1, 1750 cm^{-1} and 3400 cm^{-1} were determined, respectively. In Figure 5, the peaks that appeared clearly showed -C-O alkoxy group, -C=O stretching and -OH hydroxyl groups [14,15,18].

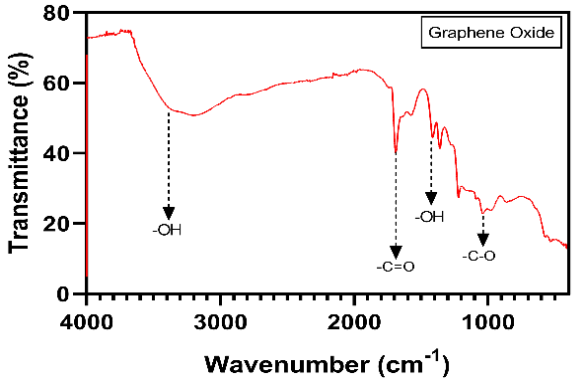

Figure 5. The FTIR spectra of synthesized graphene oxide.

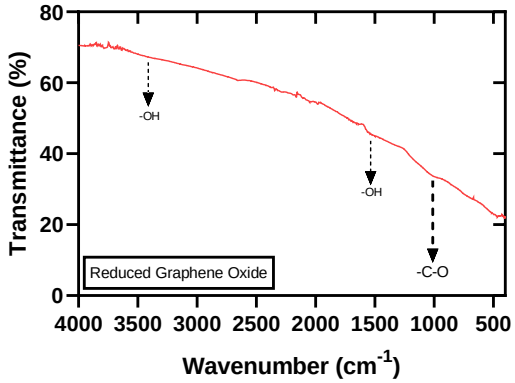

Figure 6. The FTIR spectra of synthesized reduced graphene oxide.

With having these groups proves that graphite structure successfully oxidized to graphene oxide. After reduction process with L-ascorbic acid, the structure of synthesized reduced graphene oxide showed visibly removal of -OH hydroxy groups (Figure 6) [19]. Consequently, the graphite powder was effectively converted into graphene oxide and then to reduced graphene oxide. The XRD analysis of graphene oxide and reduced graphene oxide also showed coherent information with FTIR results.

5. Conclusion

In conclusion, we discovered a novel coating method of reduced graphene oxide for Si/SiO2 surfaces for biosensor applications. To do so, we synthesized graphene oxide from graphite flakes. After, the synthesized graphene oxide was turned into reduced graphene oxide by chemical reduction process. Next part was the coating of rGO onto Si/SiO2 structure. After completion of coating reduced graphene oxide with ultrasonic assisted method, characterization of reduced graphene has been done. The FTIR and XRD analyses showed high yield production of reduced graphene oxide for sensor applications.

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