IOSR Journal of Applied Physics (IOSR-JAP)
e-ISSN: 2278-4861.Volume 17, Issue 4 Ser. 1 (Jul. – Aug. 2025), PP 84-93
www.iosrjournals.org

"Synthesis And Characterization of Nitrogen Doped Reduced Graphene Oxied (N-rGO) Exploring Enhanced Properties for Advanced Application"

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Abstract
Nitrogen-doped graphene (N-doped graphene) has gained substantial interest in recent years due to its markedly improved physical, chemical, biological, and materials-related properties compared to pristine graphene. This advancement has opened a promising frontier in the realm of two-dimensional materials science and technology. From a chemical and materials science perspective, this article/ research reviews the latest developments in N-doped graphene, covering common synthesis techniques involving temperature variations, as well as key characterization methods. Additionally, current challenges and future prospects are explored. The review aims to offer valuable insights that may guide future innovations and real-world applications of N-doped graphene.

I. Introduction

Graphene, a two-dimensional (2D) material composed of sp^2-hybridized carbon atoms, has garnered immense attention due to its remarkable properties—such as its flat structure, large surface area, outstanding electrical and optical behaviour, as well as superior mechanical characteristics (with a Young's modulus around 1.0 TPa and fracture strength close to 130 GPa) [7,8]. Since Novoselov and Geim et al. successfully exfoliated graphene in experiments for the first time [9, 10], extensive research has been conducted over the past decade to explore its potential applications stemming from these outstanding properties.

One effective way to modify the structure and behaviour of carbon-based materials is by doping with heteroatoms. In this context, incorporating nitrogen (N) atoms into the graphene lattice has been found to significantly influence its properties and enhance the performance of graphene-derived materials.

The first successful fabrication of nitrogen-doped (N-doped) graphene was reported by Peng et al., who demonstrated its use as an electrocatalyst in the oxygen reduction reaction (ORR) [15]. Their findings showed that N-doped graphene delivers much higher electrocatalytic activity compared to many conventional catalysts. This discovery sparked widespread interest, rapidly pushing N-doped graphene into the research spotlight. Since then, it has evolved quickly, and various synthesis methods have been developed to produce it, including the hydrothermal method [11-14].

II. Methodology: -

2.1 GO Synthesis Method: -

Graphene, discovered in 2004, has garnered significant attention due to its exceptional properties, including high carrier mobility, excellent thermal conductivity, good pore size distribution, superior electrical conductivity, outstanding capacitive performance, and large specific surface area. As an almost completely transparent material, graphene is capable of transmitting up to 97.7% of incident light, making it suitable for various optical and electronic applications. Graphene oxide (GO), an oxidized derivative of graphene, contains abundant oxygen-containing functional groups such as hydroxyl, carboxyl, epoxide, and carbonyl groups. These functional groups impart strong hydrophilicity to GO, allowing it to disperse uniformly and stably in aqueous solutions. GO is typically obtained by the exfoliation of graphite oxide and exists as a monolayer sheet. It exhibits excellent optical properties with a relatively low band gap (~1.17 eV), making it highly suitable for optoelectronic and sensing applications.

Structurally, graphene is a one-atom-thick sheet of carbon atoms arranged in a two-dimensional honeycomb lattice, where each carbon atom is sp^2 hybridized. It is considered the fundamental building block of all graphitic materials. The two-dimensional (2D) structure of GO is particularly advantageous for forming nanocomposites with various transition metal oxides. These composite materials benefit from unique structural characteristics such as strong interfacial bonding, electronic interactions, and synergistic effects, which enhance their performance in applications like sensors, energy storage devices, and optoelectronics.

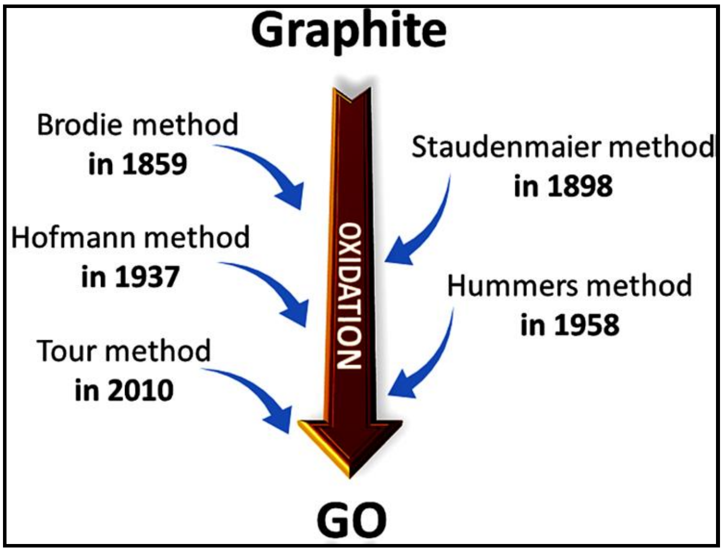

Fig. 3.1: Schematic representation of the major oxidation methods of graphite to graphene Oxide [2].

2.1.1 Tour Method :-

Synthesis of Graphene Oxide (GO):

Graphene oxide (GO) was synthesized using a Tour method. Initially, 180 mL of concentrated sulfuric acid (H_2SO_4) and 20 mL of concentrated phosphoric acid (H_3PO_4) were mixed in a 9:1 (v/v) ratio and stirred at 1000 RPM while maintaining the temperature below 10 °C using an ice bath. The addition of phosphoric acid helps to control excessive oxidation and enhances the safety of the reaction. Subsequently, 2 gm of graphite powder was slowly added to the acid mixture under continuous stirring at the same speed and temperature. After complete dispersion of graphite, 12 gm of potassium permanganate ($KMnO_4$) was gradually added while maintaining the temperature below 10 °C. The mixture turned dark green, indicating the oxidation process had initiated. Once all the $KMnO_4$ was added, the ice bath was removed, allowing the solution to gradually reach room temperature (~22 °C). The mixture was then heated to 40 °C under constant stirring at 1200 RPM. After achieving the target temperature, the heating was stopped, but stirring was continued for 12 hours to ensure complete oxidation. After this, 250 mL of distilled water was slowly added to the reaction mixture to terminate the oxidation process, resulting in an exothermic rise in temperature to around 50–60 °C and a color change to dark brown or yellow. To reduce the residual permanganate and manganese-containing by-products, 15 mL of 30 wt.% hydrogen peroxide (H_2O_2) was added dropwise. This step reduced the manganese ions to soluble manganese sulfate ($MnSO_4$) and other manganese oxides. The addition of H_2O_2 led to vigorous bubbling and a bright yellow color, indicating a high degree of oxidation, as represented by the following reaction:

$$\mathbf{2KMnO_4 + 5H_2O_2 + 3H_2SO_4 \rightarrow 2MnSO_4 + K_2SO_4 + 8H_2O(steam) + 5O_2}$$

After the reaction was complete, the resulting solution was filtered using filter paper to remove metal sulphate ions and other insoluble by-products. The remaining solid residue (filter cake) was washed thoroughly with a 1:10 (v/v) solution of concentrated hydrochloric acid (HCl) and distilled water to eliminate residual metal ions and acids. The washing process was repeated several times using centrifugation at 4000 RPM for 3 hours to ensure the complete removal of impurities. The pH of the supernatant was monitored using a universal indicator until it reached a neutral value, confirming the removal of acidic components. Finally, the obtained solid was dried in a hot air oven at 60 °C overnight. The resulting product was identified as graphene oxide (GO).

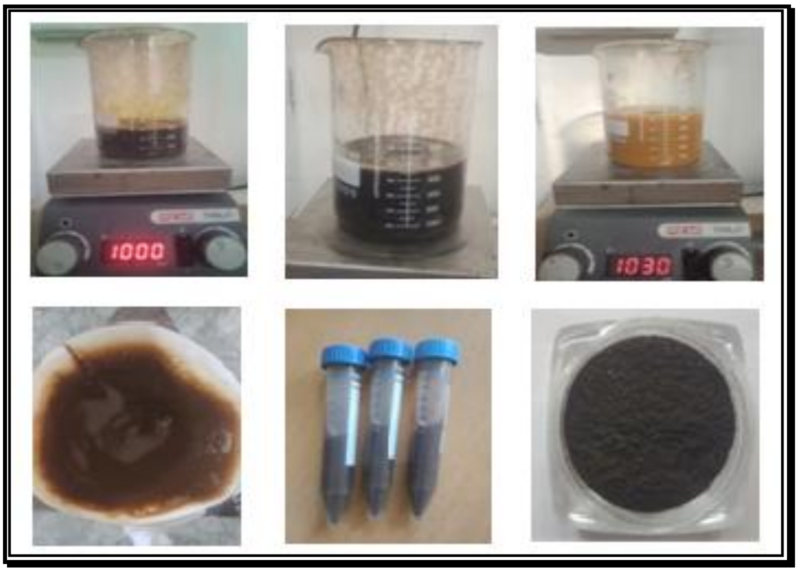

Fig. 3.2: Actual lab work of Graphene Oxide synthesis (Tour method).

2.1.2 Hummers Method:-

Synthesis of Graphene Oxide (GO):

Graphene oxide (GO) was synthesized using a modified Hummers' method. Initially, 6 g of graphite powder was dispersed in 140 mL of concentrated sulfuric acid (H_2SO_4) under continuous stirring in an ice bath to maintain the temperature below 10 °C. After 1 hour, 3 g of sodium nitrate ($NaNO_3$) was gradually added to the mixture. Within a few moments, the entire mass transformed into a thick black slurry. After 2 hours of vigorous stirring, 18 g of potassium permanganate ($KMnO_4$) was added slowly while ensuring the reaction temperature remained below 20 °C. Upon complete addition, the ice bath was removed. Subsequently, 100 mL of distilled water was added carefully, and the suspension was stirred overnight at room temperature to allow complete oxidation. The following day, 15 mL of hydrogen peroxide (H_2O_2, 30 wt. %) was added dropwise at 1500 RPM. This caused the dark brown solution to lighten, indicating the reduction of residual $KMnO_4$ and manganese intermediates. After 30 minutes, an additional 100 mL of distilled water was added to dilute the solution and terminate the reaction. The final mixture was filtered, and the solid residue was washed thoroughly with a 1:10 (v/v) diluted hydrochloric acid (HCl) solution to remove remaining metal ions. The resulting solid was dried in an oven at 60 °C overnight. The final product obtained was graphene oxide (GO).

Fig. 3.4: Actual lab work for Graphene Oxide synthesis (Hummer's method)

2.2 Reduction of Graphene Oxide (GO) Using Ascorbic Acid:

Graphene oxide (GO) was reduced using ascorbic acid (AA) as an environmentally friendly reducing agent. Initially, 400 mg of GO powder was dispersed in 400 mL of distilled water and sonicated or stirred to achieve a uniform suspension. Subsequently, 4 g of ascorbic acid was added to the dispersion, and the mixture was stirred magnetically at 60 °C for 30 minutes to facilitate the reduction process. Following the reduction, the black suspension was centrifuged at 4000 r/s for 40 minutes to separate the reduced graphene oxide (rGO) from the supernatant. To remove any residual ascorbic acid, an excess amount of 30 wt. % hydrogen peroxide (H_2O_2) was added to the black paste, and the mixture was stirred again at 60 °C for 30 minutes. The final black product was collected by centrifugation at 4000 r/s, followed by thorough washing with ethanol and distilled water (three times each) to eliminate impurities. The purified rGO was then dried in a hot air oven at 60 °C overnight to obtain the final reduced graphene oxide powder.

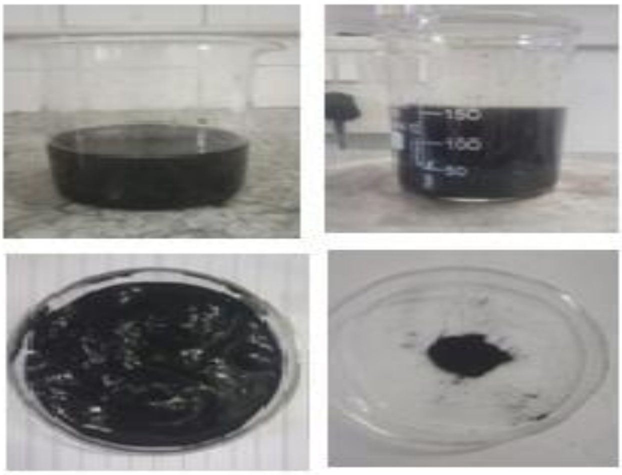

Fig. 3.5: Actual lab work for rGO synthesis.

2.3 Synthesis of Nitrogen-Doped Graphene (N-G) via Hydrothermal Method:

Nitrogen-doped graphene (N-G) was synthesized using a hydrothermal technique starting from graphene oxide (GO) as the precursor. Initially, 0.05 g of GO was dispersed in 50 mL of deionized (DI) water using ultrasonication for 2 hours to ensure uniform dispersion. After that, 20 mL of ammonia solution (25 wt. % in water) was added dropwise to the GO dispersion under constant stirring. The resulting mixture was transferred into a Teflon-lined stainless-steel autoclave and subjected to hydrothermal treatment in a furnace at different temperatures of 160 °C, 180 °C, and 200 °C for 24 hours. After the reaction, the autoclave was allowed to cool naturally to room temperature. The obtained product was collected by centrifugation and thoroughly washed several times with distilled water and ethanol to remove unreacted species and impurities. Finally, the resulting black powder was dried in a vacuum oven at 60 °C for 12 hours to obtain nitrogen-doped graphene.

Fig. 3.6: Actual lab work for N-rGO synthesis.

III. Characterization techniques:

3.1 XRD (X-Ray Diffraction):

X-Ray diffraction is a scattering of X-rays by the atoms of a crystal that produces an interference effect so that the diffraction pattern gives information on the structure of the crystal.

XRD Graphs:-

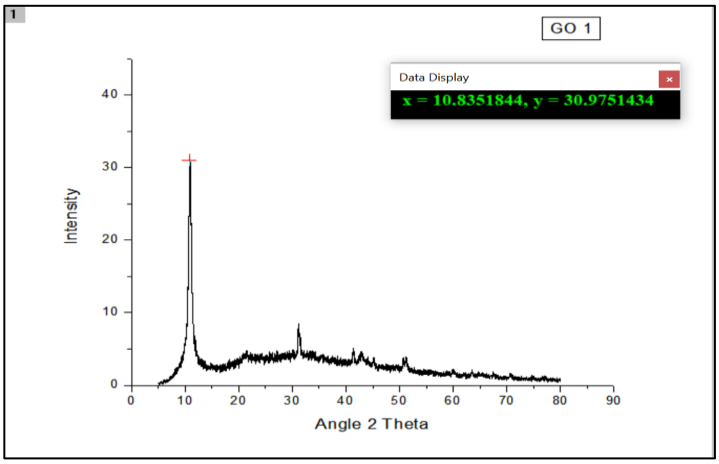

Fig. 4.1: XRD plot for Graphene oxide.

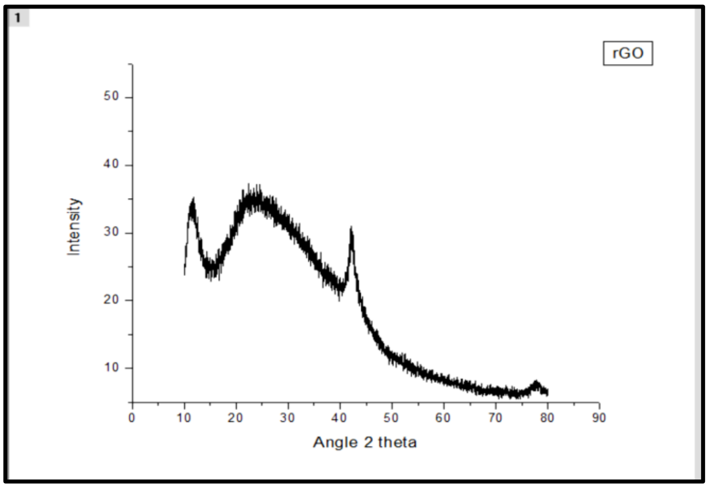

Fig. 4.2: XRD plot for Reduced Graphene oxide.

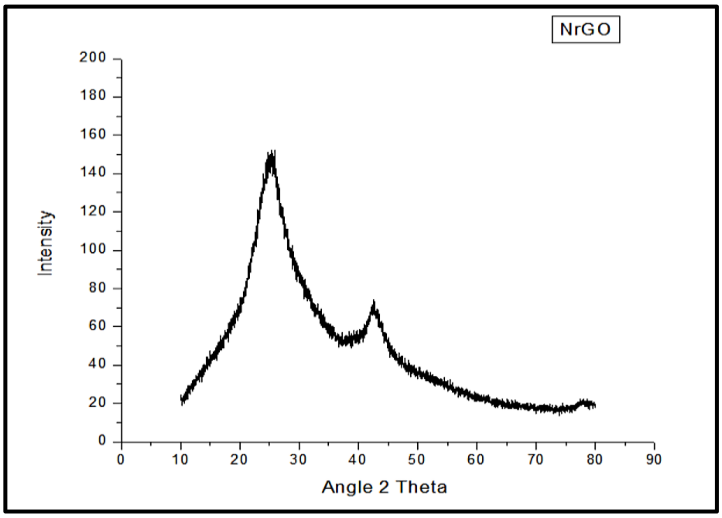

Fig. 4.3: XRD plot for Nitrogen doped reduced Graphene oxide.

3.2 UV-Visible Spectroscopy:-

UV-Visible Spectroscopy is a quantitative technique used to measure how much a chemical substance absorbs light. This is done by measuring the intensity of light that passes through a sample with respect to the intensity of light through a reference sample or blank.

UV-Visible Spectroscopy Graph:

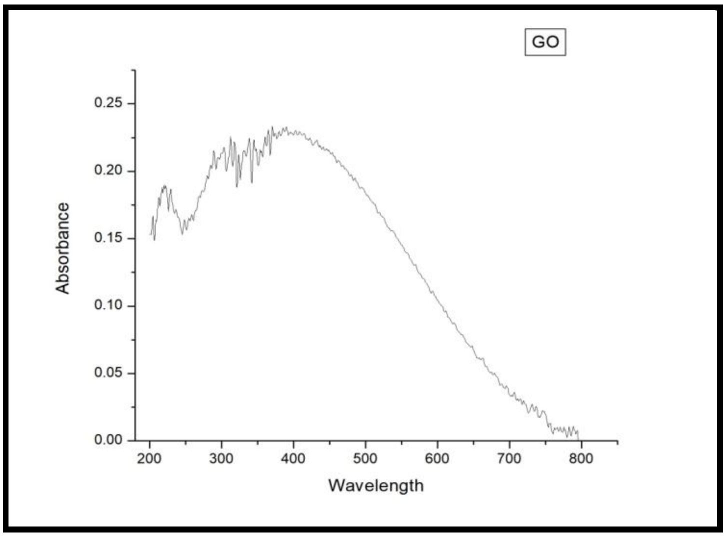

Fig4.4: UV- Visible absorption spectra of Graphene Oxide.

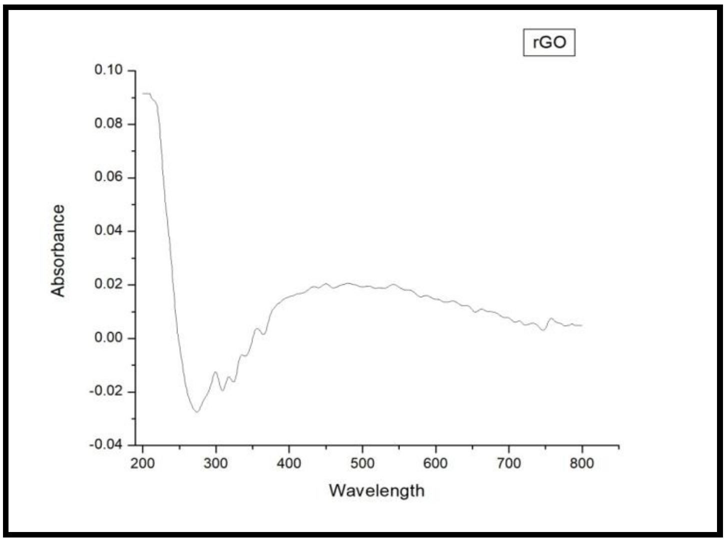

Fig 4.5:-UV- Visible absorption spectra of Reduced Graphene Oxide.

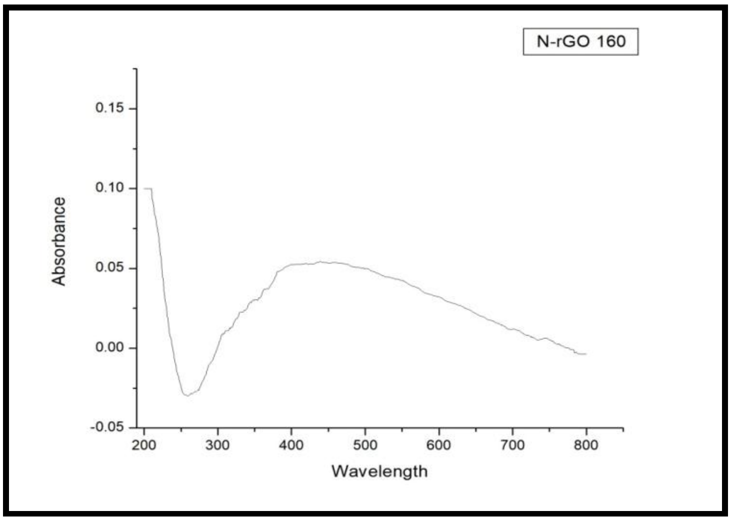

Fig 4.6:-UV- Visible absorption spectra of Nitrogen doped reduced Graphene Oxide 160 °C.

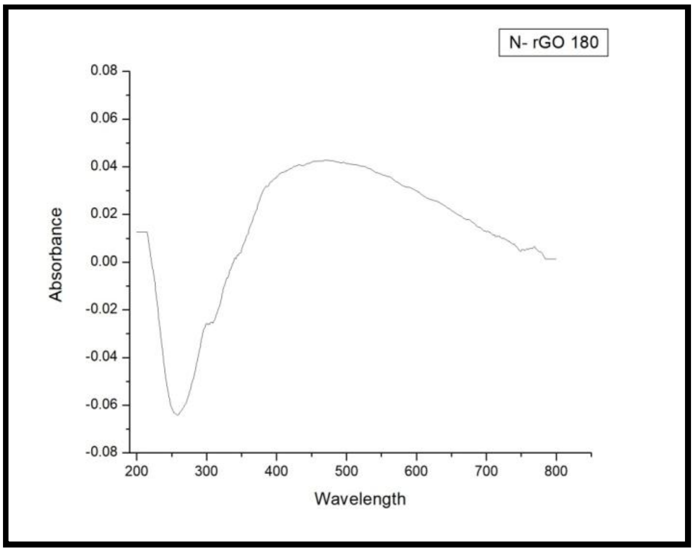

Fig 4.7:-UV- Visible absorption spectra of Nitrogen doped reduced Graphene Oxide at 180 °C.

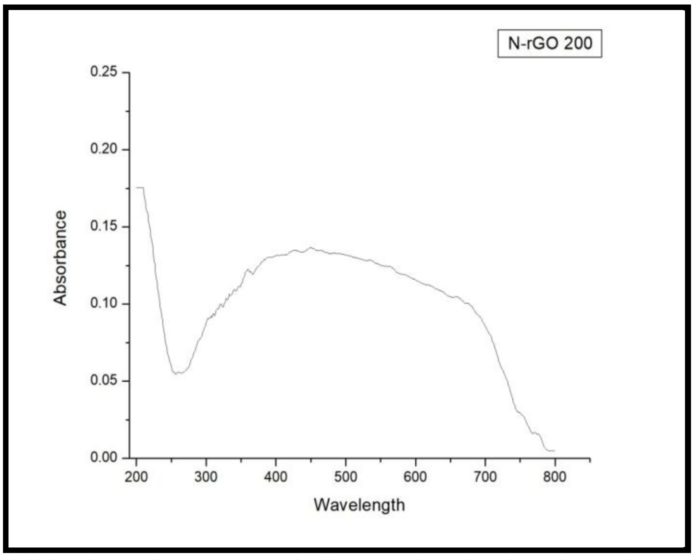

Fig 4.8:- UV- Visible absorption spectra of Nitrogen doped reduced Graphene Oxide at 200 °C.

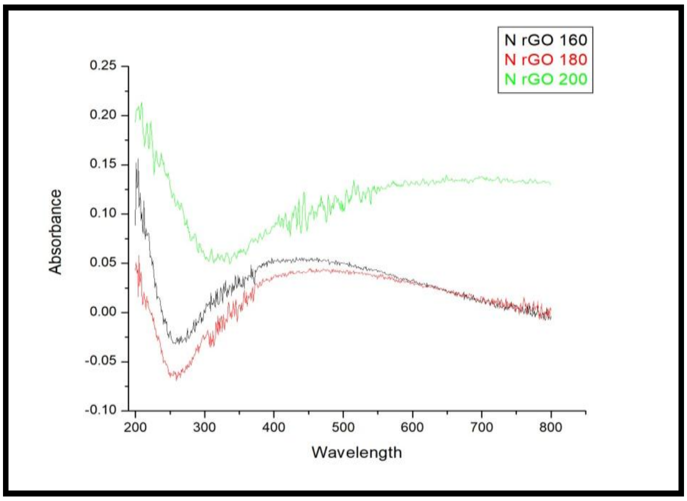

Fig 4.9: UV- Visible absorption spectra plots of all prepared sample.

IV. Result and Discussion

4.1 Result Analysis:-

Figure 5.6 presents the X-ray diffraction (XRD) patterns of graphene oxide (GO) synthesized via the Tour method, reduced graphene oxide (rGO), and nitrogen-doped reduced graphene oxide (N-rGO) samples prepared at 160 °C, 180 °C, and 200 °C. For GO, a distinct diffraction peak was observed at 2θ = 10.96°, while for rGO, a characteristic peak appeared at 2θ = 23.29°, indicating partial restoration of the graphitic structure upon reduction.

In the case of N-rGO samples, the main diffraction peaks were observed at 2θ = 25.32°, with an additional broad peak around 2θ = 43°, which aligns with previously reported findings [5]. These shifts suggest successful nitrogen doping and partial reduction of GO.

The interplanar spacing (d) values were calculated from the XRD data. GO exhibited an interlayer distance of 0.82 nm, while rGO showed a slightly reduced spacing of 0.76 nm. Notably, N-rGO synthesized at 200 °C showed a significantly lower interplanar spacing of 0.356 nm. These results are consistent with literature values. For instance, reported interplanar spacing of 0.88 nm for GO and 0.37 nm for rGO, closely aligning with our measurements. Furthermore, the d-spacing of N-rGO is comparable to that reported in Reference [5], which indicated a value of 4.05 nm, suggesting similar structural evolution upon nitrogen doping.

The optical band gap of GO, rGO, and N-rGO samples synthesized at different temperatures was determined using UV–vis spectroscopy. The absorption spectra, illustrated in Figure (X), reveal that the absorption peaks for GO, rGO, N-rGO (160 °C), N-rGO (180 °C), and N-rGO (200 °C) occur at 388, 413, 433, 442, and 450 nm, respectively. These shifts correspond to a tunable direct optical bandgap ranging from 3.2 eV for GO to 2.7 eV for N-rGO (200 °C) [5]. The progressive redshift and band gap narrowing are indicative of successful nitrogen doping and increased electronic delocalization within the graphene lattice.

Sample	Interlayer distance (d)	Crystallite Size (D)
GO	0.82 nm	12.17 nm
rGO	0.38 nm	0.80 nm
N-rGO (200°C)	0.36 nm	0.77 m

Table 5.1: The interlayer distance and crystallite size obtained from the XRD analysis.

Samples	Bandgap (E_{bg})
Graphene oxide	3.1 eV
Reduced Graphene oxide	3.0 eV
Nitrogen doped Reduced Graphene oxide (160°C)	2.9 eV
Nitrogen doped Reduced Graphene oxide(180°C)	2.8 eV
Nitrogen doped Reduced Graphene oxide (200°C)	2.7 eV

Table 5.2: The Bandgap obtained from the UV- Visible analysis.

4.2 Discussion:

The incorporation of nitrogen nanoparticles onto the surface of graphene oxide (GO) has a pronounced effect on the interplanar spacing of the resulting nanocomposites. In particular, the N-rGO (200 °C) composite exhibits a noticeable reduction in interlayer distance compared to pristine GO. This decrease in interplanar spacing is attributed to the successful anchoring and uniform dispersion of nitrogen nanoparticles on the GO surface, as well as their possible intercalation between the GO layers. The reduction in interlayer spacing is expected to positively influence the photocatalytic performance of the N-rGO (200 °C) composites. A smaller interplanar gap may correspond to an increased surface area, offering more active sites for photocatalytic reactions. Additionally, the closer proximity of the GO layers can facilitate more efficient charge transfer between GO and the nitrogen-doped regions, thereby enhancing the overall photocatalytic activity of the composite material [5].

V. Conclusion and Future scope

5.1 Conclusion:

X-ray diffraction (XRD) analysis revealed a reduction in the interplanar spacing of nitrogen-doped reduced graphene oxide (N-rGO) composites compared to pristine GO. This decrease indicates the successful incorporation and anchoring of nitrogen nanoparticles onto the GO surface. The reduced interlayer distance in the N-rGO composites suggests effective dispersion of nitrogen species and partial intercalation into the GO layers. These structural modifications are expected to enhance the photocatalytic performance of the composites, implying that N-rGO may exhibit superior photocatalytic activity compared to both GO and rGO. Among the samples, N-rGO synthesized at 200 °C exhibited the highest nitrogen content and the lowest optical band gap, approximately 2.7 eV. Furthermore, a trend was observed in which increasing the hydrothermal synthesis temperature led to a progressive decrease in the optical band gap of N-rGO. This reduction in band gap is attributed to the introduction of nitrogen-induced defect states, which create compensating energy levels within the band structure and shift the conduction band edge, thereby narrowing the band gap and enhancing light absorption efficiency.

5.2 Future Scope:

The observed changes in interplanar spacing in rGO and N-rGO composites suggest their potential for enhanced photocatalytic activity. This highlights the importance of future investigations focusing on the photocatalytic performance of these materials and their application in diverse areas such as environmental remediation, water purification, and energy conversion. Further research may also aim to optimize the synthesis process to improve the anchoring efficiency of nitrogen nanoparticles onto the GO surface, which could lead to a further reduction in interlayer spacing and enhancement of the composites' photocatalytic capabilities.

Nitrogen-doped reduced graphene oxide (N-rGO) has recently emerged as a promising material for a wide range of applications, including energy storage, electrochemical devices, fuel cells, sensors, and water treatment. Its exceptional properties—such as high electrical conductivity, excellent electronic characteristics, and strong electrocatalytic activity—make it especially suitable for these advanced technologies.

In particular, N-rGO shows great potential for membrane-based water treatment technologies, including ultrafiltration and nanofiltration systems. These technologies are especially beneficial in rural and resource-limited areas. The integration of graphene oxide into membrane structures can significantly enhance their performance by improving selectivity, increasing permeability, and imparting strong anti-fouling properties, thereby making them more efficient and durable for long-term use in water purification applications.

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