FUDMA Journal of Sciences (FJS)
ISSN online: 2616-1370
ISSN print: 2645 - 2944
Vol. 10 No. 13, July, 2026, pp 1 – 7
DOI: https://doi.org/10.33003/fjs-2026-1013-4792

Structural Modification of Reduced Graphene Oxide Thin Film Via Thermal Annealing

***[1]Nurudeen Siyaka, [2]Daniel Mikanjuola, [1]Yunusa Nuhu and [3]Jibril Idirisu**

[1]Department of Physics, Baze University, Abuja.
[2]Department of Physics, University of Abuja, Abuja.
[3]Department of Chemistry, Baze University, Abuja.

*Corresponding authors' email: nurudeenadinoyi@gmail.com Phone: +2348065159672

Received: 25 Mar. 2026
Accepted: 28 Apr. 2026
Published: 3 Aug. 2026

ABSTRACT
Reduced graphene oxide (rGO) is a promising material for thin film applications owing to its excellent structural, electrical, and mechanical properties. This study investigated the effects of thermal annealing and spin-coating cycles on the structural evolution of chemically reduced graphene oxide thin films. Graphene oxide (GO) was synthesized using the modified Hummers' method, chemically reduced with ascorbic acid, and deposited onto soda lime glass substrates by spin coating using one and two deposition cycles. The deposited films were thermally annealed at 100 °C and 200 °C under a N2/H2 atmosphere. Structural characterization was carried out using scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), and X-ray diffraction (XRD). SEM analysis showed that increasing the annealing temperature improved film continuity, reduced surface agglomeration, and enhanced morphological uniformity, while increasing the number of spin-coating cycles increased the film thickness from approximately 150 nm to 350 nm, resulting in improved substrate coverage. EDS analysis revealed an increase in carbon content from 0.14 at.% to 66.20 at.% and a corresponding decrease in oxygen concentration from 1.87 at.% to 0.02 at.%, indicating progressive deoxygenation. XRD analysis confirmed the transformation from the graphene oxide (001) reflection to the graphitic (002) reflection, demonstrating partial restoration of the sp^2 carbon network and improved crystallographic ordering. The findings indicate that thermal annealing primarily enhances structural ordering, whereas additional spin-coating cycles improve film thickness and surface coverage, providing an effective approach for fabricating high-quality rGO thin films for electronic and energy-related applications.

Keywords: Reduced Graphene Oxide, Graphene Oxide, Thermal Annealing, Spin Coating, Thin Films, SEM, EDS, XRD

INTRODUCTION

Graphene and its associated derivatives have generated significant scientific interest owing to novel characteristics, rendering them highly viable candidates for implementation in optoelectronics, catalysis, sensors, energy storage technology, transparent conductive electrodes, and flexible electronics (Sekhar, 2024; Farooq et al., 2025; Pasindu et al., 2025). Within this class of nanomaterials, reduced graphene oxide (rGO) represents a highly compelling substitute for pristine graphene, as it pairs beneficial graphene-like characteristics with cost-effective, high-volume, liquid-phase manufacturing capabilities. In contrast to graphene produced through chemical vapor deposition (CVD) or mechanical exfoliation, rGO can be manufactured in massive quantities from graphite oxide using straightforward chemical and thermal reduction protocols, making it exceptionally practical for large-area thin-film engineering (Mohammed et al., 2022; Utkan et al., 2023).

Graphene oxide (GO), generated via the intensive oxidation of raw graphite, features a high density of oxygen-bearing functional groups, such as carboxyl, carbonyl, epoxy, and hydroxyl groups, attached across its baseline plane and outer edges (Siklitskaya et al, 2021; Rout, et al., 2026). These oxygen-containing moieties disrupt the continuous sp^2 carbon lattice, which induces poor electrical conductivity but yields outstanding dispersibility within water and alternative polar solvents (Dreyer et al., 2020; Wang et al., 2021). Due to this property, GO functions as an excellent precursor material for liquid-phase deposition approaches, including inkjet printing, spray coating, dip coating, and spin coating. Nevertheless, recovering graphene-like performance metrics depends on the efficient elimination of these oxygen-containing functional groups through thermal, chemical, electrochemical, or hybrid reduction pathways (Rao et al., 2018).

Among the various reduction techniques currently accessible, thermal annealing has established itself as one of the most powerful approaches for elevating the structural alignment of graphene oxide. Annealing drives the desorption of oxygen-bearing functional groups, shrinks the spacing between adjacent layers, rehabilitates sp^2 carbon regions, and heightens graphitic ordering, thereby raising the general structural integrity of the reduced graphene oxide (Guo et al., 2024; Shivang et al., 2025). The degree of these structural transformations is heavily dictated by the specific annealing temperature, the duration of heating, and the prevailing environmental atmosphere. Insufficiently low temperatures frequently culminate in incomplete reduction, whereas excessively elevated temperatures risk generating structural defects or compromising the physical continuity of the thin film (Zhou et al., 2024). Consequently, determining appropriate annealing environments remains a crucial hurdle in maximizing the utility of rGO thin films.

Beyond thermal processing, the deposition methodology itself exerts a major influence on the morphological and structural properties of rGO thin films. Spin coating stands as one of the most frequently utilized deposition tools because it permits

the fabrication of highly uniform coatings through straightforward processing setups with strong reproducibility. Practical settings like the concentration of the precursor, spin velocity, solvent characteristics, and deposition cycles directly impact the film's thickness, surface distribution, layer homogeneity, and microstructural development (Owoeye, 2023). These structural traits subsequently dictate the efficacy of the thermal annealing step by altering heat propagation, solvent evaporation rates, and the accessibility of oxygen-bearing groups during the reduction phase.

Even though a substantial body of literature has explored either the thermal reduction of graphene oxide or the refinement of spin-coating settings as separate research topics, comparatively few projects have systematically analyzed the coupled impact of annealing temperature and deposition cycles on the structural transformation of chemically reduced graphene oxide thin films. The majority of published research tends to prioritize enhancements in electrochemical performance, electrical conductivity, or optical metrics, leaving a distinct gap in the understanding of how these processing variables collectively shape thin-film topology, elemental distributions, and crystal structures. Furthermore, the precise interplay between the film coverage brought about by multi-cycle spin coating and the structural alterations generated by thermal treatment remains poorly understood, particularly for chemically reduced graphene oxide processed with eco-friendly reduction agents like ascorbic acid (Murphy et al., 2022; Palomba et al., 2022).

Resolving this academic gap is essential because the structural configuration of rGO thin films heavily dictates their performance in future sensing, electronic, and energy-based technologies. A more definitive understanding of how annealing temperatures and deposition repetitions interact to adjust the morphology, atomic composition, and crystallinity of rGO thin films will yield crucial parameters for optimizing high-volume fabrication procedures while preserving low operational costs. In light of this, the present study explores the structural evolution of chemically reduced graphene oxide thin films synthesized via spin coating and subsequent thermal annealing at 100°C and 200°C inside a nitrogen/hydrogen environment. The consequences of modifying the spin-coating iterations and annealing temperatures on the film morphology, elemental distribution, and crystal framework of the resulting rGO thin films were thoroughly analyzed using scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), and X-ray diffraction (XRD). These insights clarify the interactive roles of deposition cycles and thermal annealing in controlling the structural properties of reduced graphene oxide thin films for the scalable manufacturing of graphene-based devices.

MATERIALS AND METHODS

Raw graphite powder was utilized as the starting material for synthesizing graphene oxide (GO). Concentrated sulfuric acid (H_2SO_4), phosphoric acid (H_3PO_4), potassium permanganate ($KMnO_4$), hydrogen peroxide (H_2O_2), ascorbic acid, ethanol, N-methyl-2-pyrrolidone (NMP), and distilled water were employed directly without undergoing supplementary purification steps. Cleaned soda-lime glass pieces were utilized as the baseline substrates for constructing the reduced graphene oxide (rGO) thin films.

Synthesis of Graphene Oxide

Graphene oxide was prepared by employing a modified version of Hummers' oxidation method. A blended acid medium consisting of concentrated sulfuric acid and phosphoric acid (at a 9:1 volume ratio) was formulated and slowly mixed with 0.50 g of graphite powder and 4.50 g of potassium permanganate under unceasing agitation. This oxidation stage was kept at 40-50°C and subsequently transferred to a temperature-regulated water bath at 50°C for a duration of 12 hours to guarantee the comprehensive oxidation of the parent graphite. Following this oxidation window, the chemical blend was permitted to cool down to ambient temperature before distilled water was gingerly introduced to halt the reaction. Hydrogen peroxide (H_2O_2) was subsequently added to convert residual permanganate and secondary manganese oxides into fully soluble manganese ions. The resulting graphene oxide fluid was washed multiple times with distilled water via filtration and centrifugation stages until a chemically stable suspension was successfully isolated. The refined GO was eventually dried inside an oven for subsequent reduction processing.

Chemical Reduction of Graphene Oxide

The as-synthesized graphene oxide was re-dispersed in distilled water to achieve a fluid concentration of 0.15 mg ml^{-1}. Chemical reduction was performed utilizing 0.48 g of ascorbic acid, which served as a green, sustainable reducing agent. This reaction environment was maintained at 80 °C for 1 hour under unceasing magnetic stirring to drive the cleavage of oxygen-bearing functional groups and the restoration of the conjugated sp^2 carbon framework. After completing the reduction step, the resulting black reduced graphene oxide (rGO) fluid was harvested through filtration and centrifugation techniques. The isolated substance was washed multiple times with ethanol and distilled water to flush out remaining chemical impurities before being dried in an oven for subsequent thin-film fabrication.

Fabrication of Reduced Graphene Oxide Thin Films

Thin coatings of reduced graphene oxide were manufactured utilizing the spin-coating method. The dried rGO powder was mixed into 5 mL of N-methyl-2-pyrrolidone (NMP) and subjected to magnetic stirring for 1 hour to establish a uniform, well-blended suspension. Prior to starting the deposition phase, the soda-lime glass substrates were thoroughly cleansed using a piranha solution to eradicate surface impurities and maximize film bonding. The prepared fluid was spread over the clean glass substrates via a spin coater programmed to run at 1000 rpm for 20 seconds, followed immediately by a second stage at 2000 rpm for 30 seconds. Two distinct deposition protocols were tested by applying either a single spin-coating cycle or a double spin-coating cycle. Physical thickness evaluations revealed an average depth of roughly 150 nm for the single-cycle coatings and approximately 350 nm for the double-cycle coatings, verifying that increasing the number of spin-coating passes effectively increased the overall film thickness and elevated the total substrate coverage.

Thermal Annealing Process

The as-deposited rGO thin films were subjected to thermal annealing within a combined nitrogen/hydrogen (N_2/H_2) gas environment to further advance the reduction state and upgrade the structural alignment. The annealing treatments were carried out at 100 °C and 200 °C for a fixed duration of 1 hour. This thermal management step drove the elimination of leftover oxygen-bearing functional groups, compressed the spacing between adjacent layers, and encouraged the partial rebuilding of the graphitic sp^2 matrix, ultimately yielding structurally superior rGO films.

Structural Characterization

Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDS)

The structural topology and atomic composition of the reduced graphene oxide thin films were evaluated using a Scios 3 Focused Ion Beam Scanning Electron Microscope (Scios 3 FIB-SEM) coupled with an Energy-Dispersive X-ray Spectroscopy (EDS) measurement system. SEM images were collected across several different zones of each specimen to analyze film continuity, particle grouping, surface topography, and void formation following the thermal annealing steps. Parallel EDS evaluations were conducted to chart the exact elemental contents of carbon, oxygen, silicon, and any other noticeable elements embedded within the specimens. This atomic concentration data was subsequently leveraged to track the efficiency of the reduction achieved across the different annealing temperatures and spin-coating sequences.

X-ray Diffraction (XRD)

The internal crystal matrix and phase changes of the fabricated rGO thin films were tracked utilizing Bruker D8 ADVANCE and Bruker D8 DISCOVER X-ray diffractometers utilizing with Cu Kα radiation ($\lambda = 1.5406$ Å). Diffraction patterns were captured using a standard reflection orientation, and the collected data were scrutinized to pinpoint changes in lattice patterns driven by thermal annealing and the chosen spin-coating cycles. The resulting diffraction reflections were evaluated to determine structural evolution, levels of graphitic ordering, and modifications in interlayer gap dimensions linked to the progressive reduction of the graphene oxide.

RESULTS AND DISCUSSION

Surface Morphology of rGO Thin Films

The surface topography of the reduced graphene oxide (rGO) thin films manufactured across various spin-coating cycles and annealing temperatures was scrutinized using scanning electron microscopy (SEM). All image sets were recorded with an electron accelerating voltage of 10 kV, a physical working distance spanning roughly 9.0-9.5 mm, and a scale magnification marker of 10 μm. These SEM assessments yielded critical details regarding how the film morphology changes as a direct consequence of thermal annealing configurations and deposition repetitions.

The SEM micrograph of the rGO film assembled via a single spin-coating cycle and annealed at 100°C displays a continuous layer covering on the substrate, paired with relatively delicate surface features. However, this surface also shows multiple clustered particles and distinct localized gaps, which indicates that the thermal energy provided at 100°C was insufficient to completely realign and flatten the graphene sheets after the initial chemical reduction phase. The tiny clusters seen across the layer are believed to stem from partially re-stacked graphene sheets or lingering oxygen-bearing groups that survived the initial reduction step. Analogous trends have been detailed for chemically reduced graphene oxide coatings annealed at lower temperature thresholds, where incomplete deoxygenation acts as a barrier to the full re-establishment of the graphitic lattice (Antidormi et al., 2020; Priyank et al., 2012).

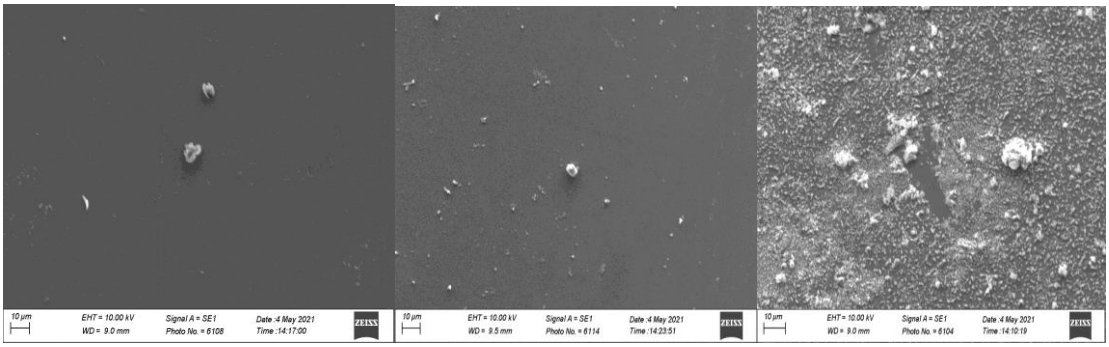
Elevating the annealing temperature to 200°C generated a visibly smoother and far more uniform surface topology. The quantity of prominent particle clusters dropped substantially, and the thin film showed enhanced continuity along with fewer structural flaws. This structural improvement is due to the accelerated elimination of oxygen-bearing functional groups during the high-temperature annealing phase, which stimulates the partial re-establishment of the sp^2 carbon framework and yields superior packing of individual graphene sheets. The lower density of physical discontinuities points to magnified sheet-to-sheet bonding and better adhesion between neighboring graphene layers. Comparable structural evolution was reported by Bamboo (2024), who determined that moderate thermal treatments elevate graphitic alignment by mitigating structural randomness and removing residual oxygen groups.

The film constructed via two spin-coating runs and annealed at 200°C displayed the most consistent and uniform topology among all tested specimens. This thicker film coating (measuring around 350 nm) provided much better physical coverage across the glass substrate, leaving only a few isolated particle clusters visible. When compared directly against the single-cycle specimens, the multi-layer deposition strategy generated a tighter and more continuous film layer, which suggests that the extra spin-coating run primarily served to upgrade substrate coverage rather than acting as a direct driver of the chemical reduction mechanism. This distinction is critical because the overall level of reduction is principally governed by the thermal treatment, whereas multiplying the number of deposition runs mostly acts to adjust film thickness, structural continuity, and topographic uniformity. The enhanced coverage seen in the two-cycle film is therefore credited to the larger mass of deposited material interacting with the subsequent thermal realignment during the annealing stage.

Even though qualitative enhancements in surface topology are distinctly visible, SEM imagery on its own cannot be used to precisely calculate values for surface roughness, structural pore size spreads, or micro-crack densities. Gathering such discrete parameters would necessitate alternative diagnostic tools like atomic force microscopy (AFM) or specialized digital image-processing software. As a result, the current assessment focuses strictly on visible modifications in film continuity, cluster presence, and substrate coverage rather than declaring specific numerical roughness values. This analytical boundary prevents overinterpreting the data while staying fully aligned with the direct structural proofs visible in the SEM micrographs.

On the whole, the SEM findings clarify that thermal annealing and spin-coating sequences exert distinct yet complementary roles on the final topology of rGO thin films. Thermal annealing chiefly assists structural restoration by driving deoxygenation and refining graphitic ordering, whereas increasing the number of spin-coating passes acts to increase film thickness and maximize substrate coverage. The combined impact of these processing variables yields tighter and more uniform rGO films, creating an excellent structural foundation for the subsequent crystallographic and elemental investigations.

.

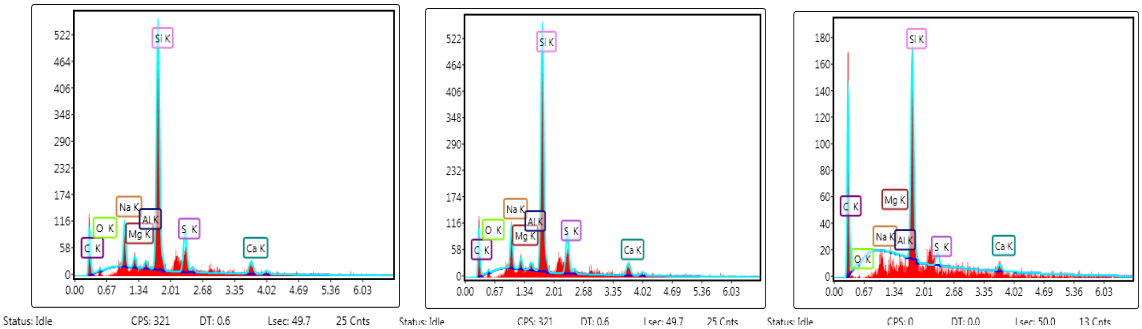


1 cycle, 200 ºC 2 cycles, 100 ºC 2 cycles, 200 ºC

Figure 1: SEM Image of Annealed Reduced Graphene Oxide Thin Film Deposited by the Spin Coater at Two Different Cycles and Temperatures

Elemental Composition and Degree of Reduction (EDS)

The elemental profiles of the reduced graphene oxide (rGO) thin films fabricated across various spin-coating iterations and annealing temperatures were evaluated via Energy-Dispersive X-ray Spectroscopy (EDS). Typical EDS spectra verified that carbon (C) and oxygen (O) stood as the prominent elements present across all specimens, whereas silicon (Si), sodium (Na), magnesium (Mg), aluminum (Al), sulfur (S), and calcium (Ca) originated predominantly from the underlying soda-lime glass substrate or leftover chemical impurities. Because EDS techniques probe both the thin film layer and, in thinner spots, a portion of the support material beneath it, the detected silicon peak must be understood as a direct contribution from the substrate rather than as an inherent element of the graphene-based coating itself.



1 cycle, 100 ºC 2 cycles, 100 ºC 2 cycles, 200 ºC

Figure 2: EDS Analysis of Reduced Graphene Oxide Thin Film Deposited by the Spin Coater at two Different Cycles and Temperatures

The elemental data in table 1 highlighted a steady growth in carbon concentration coupled with a corresponding drop in oxygen levels as the annealing temperature was raised from 100°C to 200°C and as more spin-coating cycles were applied. For the thin film generated via one spin-coating run and annealed at 100°C, the calculated carbon and oxygen levels were 0.14 at.% and 1.87 at.%, respectively. Stepping up the annealing temperature to 200 °C caused the carbon concentration to rise to 15.31 at.% while pushing the oxygen content down to 0.88 at.%, confirming a more thorough removal of oxygen-bearing functional groups during the hotter thermal sequence. A further expansion of the deposition cycles generated a massive jump in carbon concentration, with the double-cycle coating annealed at 100°C demonstrating 40.98 at.% carbon and 0.76 at.% oxygen. The highest degree of chemical reduction was achieved by the double-cycle film annealed at 200°C, which contained 66.20 at.% carbon paired with a mere 0.02 at.% oxygen, confirming comprehensive deoxygenation across the graphene oxide sheets. To offer a more rigorous assessment of reduction performance, the carbon-to-oxygen (C/O) ratio was explicitly determined for each individual specimen (Table 1). Rather than looking at carbon and oxygen atomic percentages in isolation, tracking the C/O ratio yields a far more dependable metric for the restoration of the graphitic carbon matrix because it mirrors the relative removal rate of oxygen groups directly.

Table 1: Surface Elemental Composition and C/O Atomic Ratio of Thermally Reduced Graphene Oxide Samples Determined by XPS

Sample	Carbon (at.%)	Oxygen (at.%)	C/O Ratio
1 cycle – 100 °C	0.14	1.87	0.07
1 cycle – 200 °C	15.31	0.88	17.40
2 cycles – 200 °C	66.20	0.02	3310

This steady increase in the calculated C/O ratio proves that thermal annealing effectively strips away oxygen-bearing functional groups like hydroxyl, epoxy, carbonyl, and carboxyl groups, thereby rehabilitating the continuous conjugated sp^2 carbon lattice of graphene. This behavior matches well with earlier publications reporting that moderate

thermal annealing markedly improves the reduction state of graphene oxide and stimulates structural realignment into graphitic domains (Xu et al., 2025; Sun et al., 2014). The collected EDS spectra also demonstrate that the intensity of the substrate's silicon reflection shrinks relative to the carbon reflection as more spin-coating iterations are added. This structural behavior is fully expected because thicker coatings offer superior surface masking over the glass substrate, which limits the interaction volume of the primary electron beam with the underlying silica. As a result, the reduction of the silicon peak intensity must not be misconstrued as a chemical modification of the glass substrate, but rather as direct physical proof of enhanced film coverage. This finding perfectly matches the physical thickness measurements, which rose from 150 nm for the single-pass coatings up to 350 nm for the double-pass coatings.

Even though these elemental patterns clearly track a progressive chemical reduction, EDS must be treated as a semi-quantitative tool when dealing with thin-film configurations, given that the measured atomic values are influenced by electron beam interaction depths, total film thickness, and substrate background signals. Consequently, the elemental percentages should be evaluated alongside the parallel SEM images and XRD patterns rather than being viewed as an isolated metric. In this research, the rising carbon content, the shrinking oxygen values, the superior film continuity tracked by SEM, and the elevated graphitic ordering proven by XRD collectively verify that pairing thermal annealing with tailored film deposition parameters successfully drives the structural transition of graphene oxide into high-quality reduced graphene oxide.

X-ray Diffraction (XRD) Analysis

The internal crystal matrix and structural phase development of the reduced graphene oxide (rGO) thin films engineered under various spin-coating runs and annealing temperatures were investigated via X-ray diffraction (XRD), as displayed in Figure 3. The recorded diffraction spectra reveal a progressive structural transition from an oxidized graphene oxide state toward a more highly aligned graphitic framework as both the annealing temperature and film depth are increased. The XRD profile of the single-cycle coating annealed at 100°C displays a wide, diffused diffraction peak centered at $2\theta = 10.8^{o}$, which matches the characteristic (001) crystal plane reflection of graphene oxide. This specific peak is a hallmark of oxidized graphite, confirming the presence of dense oxygen-bearing functional groups and trapped water molecules lodged between neighboring graphene planes. Utilizing Bragg's law:

$$d = \frac{\lambda}{2\sin\theta} \quad (1)$$

where $\lambda = 1.5406\,Å$ (via Cu Kα radiation), the baseline interlayer gap space is calculated to be 0.82 nm. This value is significantly larger than the standard interlayer spacing of highly crystalline graphite (0.335 nm), confirming that the aggressive oxidation process pushed apart the graphitic stacking layers and expanded the internal lattice. The broad shape of this diffraction peak also indicates poor overall crystallinity and substantial structural disorder, which are standard structural features of graphene oxide materials.

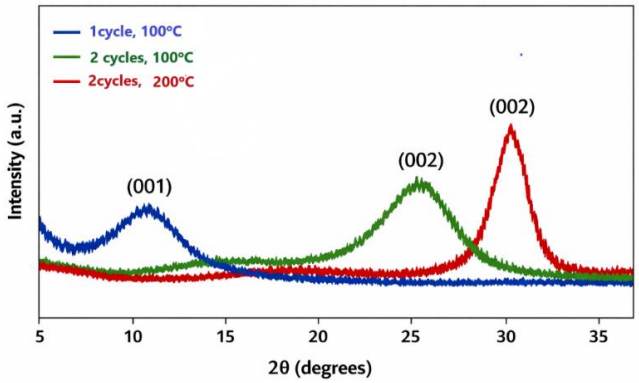


Figure 3: XRD Patterns of Reduced Graphene Oxide Thin Film Deposited by the Spin Coater for Two Different Cycles and Annealing Temperature

Upon raising the annealing temperature to 200°C while keeping a single spin-coating cycle, the primary diffraction reflection migrates away from the GO-associated (001) peak and relocates toward the traditional graphitic (002) reflection situated at $2\theta = 25.5°$. Concurrently, this new peak becomes visibly more intense and sharply defined, proving a partial recovery of graphitic ordering brought about by the thermal treatment. The calculated interlayer gap distance shrinks down to roughly 0.35 nm, demonstrating that thermal annealing successfully expels the oxygen-bearing functional groups, which enables adjacent graphene sheets to restack much closer together. Even so, the relatively wide profile of this peak shows that the material still harbors structural defects and turbostratic orientation errors, which are widely documented in chemically reduced graphene oxide samples processed at moderate annealing temperatures.

An additional upgrade in lattice crystallinity is observed for the thin film generated via two spin-coating iterations and annealed at 200°C. The (002) crystalline peak becomes distinctly sharper and far more intense, accompanied by a small migration toward higher diffraction angles. The narrowing of this diffraction peak indicates an expansion in average crystallite size along with superior stacking alignment of the graphitic sheets, while the heightened peak intensity mirrors a more regular stacking framework throughout the rGO layout. This improved crystallinity is due to the combined action of thermal annealing and expanded film thickness. Thermal annealing drives steady deoxygenation and structural repairing of the sp^2 carbon matrix, whereas the supplementary spin-coating cycle creates a thicker, more uniform film (350 nm) that ensures better substrate masking and encourages a more homogeneous structural realignment during the heat treatment.

Table 2: XRD Analysis of Reduced Graphene Oxide Samples Under Different Thermal Treatment Conditions

Sample	Main Peak	20(°)	Plane d-spacing (nm)	Structural Interpretation
1 cycle, 100 °C	(001)	10.8	0.82	Graphene oxide with expanded interlayer spacing
1 cycle, 200 °C	(002)	25.5	0.35	Partial restoration of graphitic domains
2 cycles, 200 °C	(002)	30.2	0.30	Improved graphitic ordering and increased crystallinity

The documented phase transition from the (001) reflection of raw graphene oxide to the definitive (002) reflection of reduced graphene oxide confirms the successful reduction of the precursor and the steady return of graphitic domains. Corresponding structural phase shifts have been reported by Pei and Cheng (Ahmed et al. (2024), and Hamdane et al. (2024), who similarly linked the eradication of the (001) peak and the subsequent appearance of the (002) reflection to the expulsion of oxygen functional groups and the restoration of conjugated sp^2 carbon networks during thermal processing. The crystalline trends from the XRD data show excellent alignment with the parallel SEM and EDS evaluations detailed previously. SEM data showed enhanced layer continuity and a drop-in surface particle clustering post-annealing, while EDS data tracked a steady rise in carbon values alongside a heavy drop in oxygen metrics. The collective data from these distinct characterization tools confirm that thermal annealing accelerates the reduction sequence by purging oxygen functionalities, compressing the interlayer gap space, and stimulating graphitic structural ordering. Furthermore, the increased film thickness obtained through two spin-coating cycles improves surface coverage without directly influencing the chemical reduction process, highlighting the complementary roles of deposition and annealing in determining the structural quality of the rGO thin films. It must be noted that, despite the notable upgrades in lattice alignment post-annealing, the diffraction reflections remain quite broad when compared directly against highly crystalline natural graphite. This experimental finding demonstrates that the synthesized coatings still retain a certain degree of structural randomness and are composed of nanocrystalline graphitic patches rather than perfectly continuous, flawless graphite. Such material behavior is entirely expected for chemically reduced graphene oxide processed at relatively modest annealing windows (100-200 °C), where complete graphitization is not anticipated to occur. As a result, the structural development tracked in this work should be viewed as a partial recovery of graphitic alignment, which is fully sufficient to enhance the structural performance of the thin films while maintaining the distinct benefits of low-temperature device processing.

CONCLUSION

Reduced graphene oxide (rGO) thin films were effectively engineered through the chemical reduction of graphene oxide precursors followed by spin coating and subsequent thermal annealing steps. The collected data prove that increasing the annealing temperature from 100°C to 200°C significantly upgraded the structural properties of the films by driving more thorough deoxygenation and a partial return of the graphitic phase. SEM imagery revealed a smoother, more continuous film morphology, while EDS tracking verified a rise in carbon concentration accompanied by a parallel drop in oxygen content as the annealing temperature went up. XRD characterization further tracked the successful phase transformation from the characteristic graphene oxide (001) plane reflection to the graphitic (002) plane reflection, confirming the progressive rebuilding of the sp^2 carbon lattice. In parallel, expanding the number of spin-coating iterations increased the physical thickness and maximized substrate masking without altering the underlying chemical reduction dynamics. This research project was bounded specifically to evaluating the structural traits of the rGO thin films using SEM, EDS, and XRD, however, the direct correlations linking these observed structural transformations to the resulting optical or electrical characteristics of the films could not be mapped out. Future research efforts should incorporate Raman spectroscopy, atomic force microscopy (AFM), optical transmission profiling, and electrical conductivity testing to supply a more exhaustive mapping of the structure-property relationships within these optimized rGO thin films.

REFERENCES

Ahmad, F., Mohammed A., Lease, J., Zheng, A., Lim T., Subota, T., Andou, Y. (2024). Structural Metamorphosis of Graphitic Derivatives Produced from Fractionated. *ACS Sustainable Resource Management.* https://doi.org/10.1021/acssusresmgt.3c00048

Antidormi, A., Roche, S., Colombo, L. (2020). Impact of oxidation morphology on reduced graphene oxides upon thermal annealing. *Journal of Physics: Materials.*1(3), 2020. https://doi.org/10.1088/2515-7639/ab5ef2.

Bamboo L., Ahmad F., Mohammed A., Lease, J., Zheng, A., Tsubota, T., Andou, Y. (2024). Structural Metamorphosis of Graphitic Derivatives Produced from Fractionated. *ACS Sustainable Resource Management*. 1(1). https://doi.org/10.1021/acssusresmgt.3c00048

Carotenuto, M., Longo G. (2022). A Brief Review: The Use of L-Ascorbic Acid as a Green Reducing Agent of Graphene Oxide. *Materials (Basel)*.15(18), 6456. https://doi.org/10.3390/ma15186456.

Farooq, N., Rehman, Z., Hareem, A., Masood, R., Ashfaq, R., Fatimah, I., Hussain, S., Ansari, S., & Parveen, N. (2024). Graphene Oxide and Based Materials: Synthesis, Properties, and Applications -A Comprehensive Review. *MatSci Express*. 1. 185-231. https://doi.org/10.69626/mse.2024.0185.

Giang T., Le, J., Manyam, P., Opaprakasit, N., Chanlek, N., Grisdanurak, P. (2018). Divergent mechanisms for thermal reduction of graphene oxide and their highly different ion affinities. *Diamond and Related Materials.* Volume 89, 2018, Pages 246-256. https://doi.org/10.1016/j.diamond.2018.09.006

Guo M., Yuan H., Ni K., Ye C., Pan F., Xiong J., Zhu Y. (2025). Structural Repair of Reduced Graphene Oxide Promoted by Single-Layer Graphene. *Adv Sci (Weinh)*. 12(7). https://doi.org/10.1002/advs.202410088.

Kanishka K., De Silva, H., Hsin-Hui H., Rakesh J., Masamichi Y. (2020). Restoration of the graphitic structure

by defect repair during the thermal reduction of graphene oxide. *Carbon*. Volume 166. https://doi.org/10.1016/j.

Mohammed S., Pegah S. M., Kenta N., Garrett St., Michael G., Ilkeun L., Mahesh R. N., Ashok M. (2022). Synthesis of pristine graphene-like behaving rGO thin film: Insights into what really matters, *Carbon*, Volume 186, Pages 437-451, ISSN 0008-6223, https://doi.org/10.1016/j.carbon.2021.10.011.

Murphy, B., Apollo, N., Unegbu, P.,Posey, T., Rodriguez, N.;Hendricks, Q., Cimino, F., Richardson, G., Vitale, F. (2022). Vitamin C-reduced graphene oxide improves the performance and stability of multimodal neural microelectrodes. *iScience,* 25 (7). 2022 https://doi.org/doi.org/10.1016/j.isci.2022.104652

Owoeye V, Adewinbi S., Salau A., Orelusi A., Adeoye A, Akindadelo A. (2023). Effect of precursor concentration on stoichiometry and optical properties of spray pyrolyzed nanostructured NiO thin films. *Heliyon*. 9(1). https://doi.org/10.1016/j.heliyon.2023.e13023.

Palomba M, Carotenuto G, Longo A. (2022). A Brief Review: The Use of L-Ascorbic Acid as a Green Reducing Agent of Graphene Oxide. *Materials (Basel)*. 15(18), 2022. https://doi.org/10.3390/ma15186456

Pasindu V, Yapa P, Dabare S, Munaweera I. Multifunctional transition metal oxide/graphene oxide nanocomposites for catalytic dye degradation, renewable energy, and energy storage applications. *RSC Adv.* 2025 Sep 12;15(40):33162-33186. https://doi.org/10.1039/d5ra04806k

Priyank V., Neelkanth M., Guan-Yu C., Zeyang L., Angela M., Jeffrey C. (2016). New insights into the thermal reduction of graphene oxide: Impact of oxygen clustering. *Carbon*, Volume 100, 2016. https://doi.org/10.1016/j.carbon.2015.12.087.

Rao, S., Upadhyay, J., Polychronopoulou, K., Umer, R., & Das, R. (2018). Reduced Graphene Oxide: Effect of Reduction on Electrical Conductivity. *Journal of Composites Science*, *2*(2), 25. https://doi.org/10.3390/jcs2020025

Rout, P., Roy, S., Acharya, P. (2026). Graphene oxide–modified fly ash pellet lightweight concrete: Microstructure–property relationships and transition to structural-grade performance. *Innovative Infrastructure Solutions*. 11. https://doi.org/10.1007/s41062-026-02727-6.

Sekhar C. (2024). Application of graphene/graphene-oxide: A comprehensive review[J]. *AIMS Materials Science*, 2025, 12(3): 453-513. https://doi.org/10.3934/matersci.2025023

Shivang S., Akshay K., Naveen Y., Mohit K., Sharma, B. (2025). Defect evolution and structural transformation in graphene oxide during hydrogen assisted thermal annealing, *Ceramics International*, Volume 51, Issue 26, 49836-49844. https://doi.org/10.1016/j.ceramint.2025.08.222.

Siklitskaya A, Gacka E, Larowska D, Mazurkiewicz-Pawlicka M, Malolepszy A, Stobiński L, Marciniak B, Lewandowska-Andrałojć A, Kubas A. (2021). Lerf-Klinowski-type models of graphene oxide and reduced graphene oxide are robust in analyzing non-covalent functionalization with porphyrins. *Sci Rep.* 11(1):7977. https://doi.org/10.1038/s41598-021-86880-1.

Sun, P., Wang, Y., Liu, H., Wang, K., Wu, D., Xu, Z., Zh,u H. (2014). Structure evolution of graphene oxide during thermally driven phase transformation: is the oxygen content really preserved? *PLoS One*. 9(11). https://doi.org/10.1371/journal.pone.0111908.

Utkan, Guldem, Y., Gorkem, T., Beste, C., Ozturk, Tarik T. (2023). Production of Reduced Graphene Oxide by Using Three Different Microorganisms and Investigation of Their Cell Interactions, *ACS Omega AmericanChemical Society*, 8(34). https://doi.org/10.1021/acsomega.3c03213

Xu, X., Huang, J., Miao, G., Yan, B., Chen, Y., Zhou, Y., Zhang, Y., Zhang, X., & Cai, W. (2025). Visualizing Thermal Reduction in Graphene Oxide. *Materials*, *18*(10), 2222. https://doi.org/10.3390/ma18102222

Zhou, B., Cai, Z., Wen, J., Liu, Huajie. (2024). Engineering Thermally Reduced Graphene Oxide for Synchronously Enhancing Photocatalytic Activity and Photothermal Effect. *ACS Applied Bio Materials,* 7 (9) 6249-6260. https://doi.org./10.1021/acsabm.4c00862.