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Sustainable and Cost-Efficient Production of Micro-Patterned Reduced Graphene Oxide on Graphene Oxide Films

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Abstract: This study tackles the critical demand for sustainable synthesis methods of reduced graphene oxide (rGO), highlighting the environmental drawbacks of conventional chemical processes. We introduce a novel, green synthesis technique involving the irradiation of a 500 eV argon ion beam, which not only facilitates the creation of micro-patterned rGO on a graphene oxide (GO) film but also enables simultaneous material characterization and patterning. By adjusting the irradiation exposure time between 0 and 80 s, we achieve meticulous control over the attributes and the reduction process of the material. The use of X-ray photoelectron spectroscopy (XPS) allows for real-time monitoring of the reduction from GO to rGO, evidenced by a notable reduction in the intensities of C-O, C=O, and O-C=O bonds, and an increase in C-C bond intensities, indicating a significant reduction level. Our research demonstrates the efficient production of eco-friendly rGO using precise, controlled argon ion beam irradiation, proving its advantages over traditional methods. These results contribute to the development of sustainable material science technologies, with potential applications in electronics, energy storage, and more.

Keywords: graphene oxide (GO); argon ion beam irradiation; green synthesis methods; environmental sustainability; reduced graphene oxide (rGO); X-ray photoelectron spectroscopy (XPS); material properties control

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1. Introduction

Graphene oxide (GO), an oxidized form of graphene, has garnered significant attention in materials science and nanotechnology due to its unique properties and potential applications across various fields, including electronics, energy storage, and biomedical devices [1–3]. The synthesis of reduced graphene oxide (rGO) from GO is a critical step in harnessing these properties, as the reduction process helps restore the conjugated graphene-like structure, significantly improving its electrical, thermal, and mechanical characteristics [4–6]. This process aims to eliminate oxygen-containing groups introduced during oxidation, which compromises the graphene's sp^2 bonding and degrades its inherent qualities. Various reduction methods exist, including chemical reduction with agents like hydrazine, thermal reduction under high temperatures, electrochemical reduction via voltage application, and photoreduction using ultraviolet light, each with distinct mechanisms and efficiency levels [7–10].

However, the aforementioned GO reduction methods each come with their own set of drawbacks. Chemical methods pose environmental and health risks due to toxicity and leave residual contaminants that can affect rGO's properties [11]. Thermal reduction can damage the graphene structure and is energy-intensive [12], while electrochemical methods may face scalability challenges and incur high equipment costs [13]. Photoreduction might not fully reduce GO and poses health risks from UV exposure [14]. Across all methods, challenges in achieving consistent reduction, control over the process, and cost-efficiency trade-offs limit their effectiveness and applicability in various applications, underscoring the need for improved reduction techniques.

The use of ion beams for material processing has been explored extensively due to their precision and the ability to induce specific changes at the atomic or molecular level [15,16]. Argon ion beams, in particular, have been utilized for surface cleaning and etching due to their inert nature. Thus, the employment of argon ions at a specific energy level introduces a physical reduction approach via ion beam irradiation [17]. This method is capable of causing structural alterations in materials, such as the elimination of functional groups, promoting cross-linking, and creating defects [18]. Argon ion beam irradiation stands out as an innovative physical method for reducing materials, allowing for precise adjustments in ion energy and exposure to achieve targeted changes in the material's structure without relying on chemical processes [19]. Within this framework, argon ion irradiation presents a promising avenue for reducing the oxygen content in GO and modifying its structure, potentially leading to considerable changes in its properties. The method of reducing GO through argon ion irradiation stands out as both cost-effective and environmentally friendly. Nonetheless, the lack of direct access to specialized studies on the reduction of GO by argon ions complicates the ability to provide detailed insights into the precise mechanisms, the effectiveness, and the outcomes of this approach.

In response to these challenges, our study introduces a pioneering method for synthesizing rGO that avoids the use of toxic-chemical-reducing agents. The approach involved exposing GO samples to argon ion beam irradiation, integrated within XPS analysis commonly employed for cleaning, operating at 500 eV for durations ranging from 0 to 80 s. This span was selected to explore how irradiation time affects the reduction process, with the hypothesis that longer durations would yield a more comprehensive reduction of GO. To our knowledge, while argon ion beam irradiation has been applied in various material processing contexts like surface cleaning and etching, its specific application for reducing GO to rGO using XPS at low energy (500 eV) remains largely unexplored in the literature. Additionally, beyond enabling precise reduction control of GO, this method also allows for simultaneous micro-patterned rGO on GO films, demonstrating its dual functionality in material reduction and design. This positions our study at the forefront of innovative and eco-friendly synthesis techniques for rGO.

The characterization of the synthesized rGO was performed using X-ray photoelectron spectroscopy (XPS), a powerful technique for determining the elemental composition and chemical bonding states within materials. XPS devices equipped with argon ion beam capabilities serve dual purposes: material characterization and modification. This dual functionality means that a single piece of equipment can both synthesize rGO and analyze the chemical composition of materials without the need for separate systems. The XPS analysis provided quantitative insights into the changes in the carbon and oxygen content of the samples, as well as qualitative information on the nature of the chemical bonds present before and after irradiation. The results of the XPS analysis confirmed the presence of both oxygen and carbon in the GO samples, with the relative carbon content increasing as a function of irradiation time. High-resolution spectra revealed the initial presence of C-O, C=O, and O-C=O bonds within the carbon and oxygen spectra. Notably, with the progression of irradiation time, there was a marked decrease in the peak intensity of these bonds, especially the O-C=O bond which culminated in its complete disappearance as shown in C 1s spectra. This observation is indicative of the effective reduction of GO to rGO, achieved through the reduction in oxygen-containing functional groups and the removal of some of them.

This method offers a promising approach to converting GO into rGO, which is in line with sustainable and eco-friendly production principles. By utilizing argon ion beam irradiation, the need for costly and harmful chemicals is eliminated, promoting green chemistry and reducing potential environmental and health risks. Argon ion beam irradiation allows for precise control and potentially faster processing compared to chemical reduction methods, thereby enhancing efficiency and throughput and reducing production time and labor costs. The ability to adjust irradiation time for precise control over the reduction process further demonstrates the method's flexibility, paving the way for its widespread adoption in rGO synthesis.

2. Materials and Method

The graphene oxide (GO) powder was procured from XFNANO, Nanjing, China. It exhibited a purity level of 99.5% and was utilized directly as supplied, without any additional purification processes. Initially, we dispersed 50 mg of GO powder in 10 mL of deionized water. This mixture was subjected to sonication for 30 min, followed by stirring at 800 rpm for 40 min. The drop casting technique was utilized to transfer the resulting GO solution onto a solid substrate, and the resulting film was subjected to irradiation with a 500 eV argon ion beam to produce reduced graphene oxide. X-ray photoelectron spectroscopy (XPS) was employed to characterize the synthesized reduced graphene oxide, and Figure 1 depicts the system setup. Spectra acquisition and characterization were performed using the Thermo-scientific K-alpha X-ray photoelectron spectrometer which manufactured by Thermo Fisher Scientific, Waltham, MA, USA. The samples were loaded into a vacuum of 10^{-8} m bar at room temperature, and the source comprised monochromatic Al kα with a characteristic energy of 1486.6 eV. An electron gun was used to neutralize surface charges in non-conductive materials, while the graphene oxide was exposed to a 500 eV argon ion beam at irradiation times ranging from 0 to 80 s, with a measurement spot size of 400 µm. The survey scan was obtained using a passing energy of 200 eV and a step size energy of 1 eV, while high-resolution spectra were acquired using a passing energy of 50 eV and a step size energy of 0.1 eV.

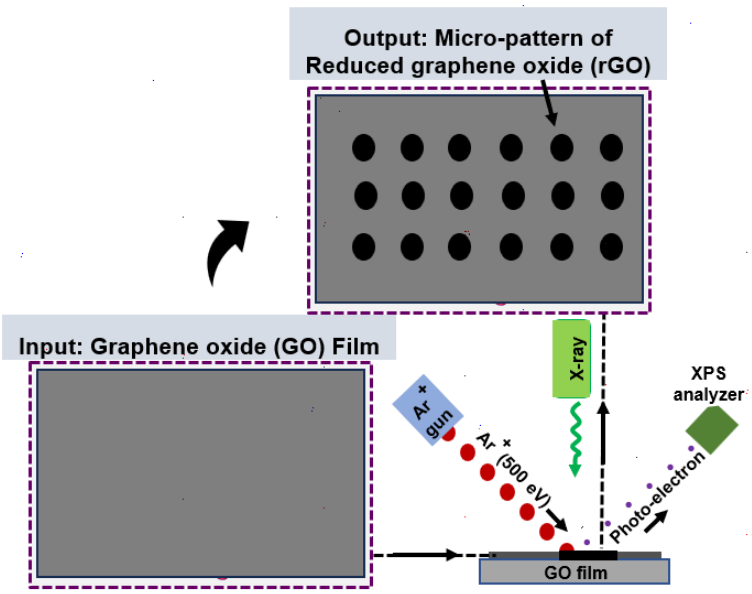

Figure 1. Schematic diagram showing the fabrication of the micro-pattered rGO on GO film using 500 eV argon ion beam.

3. Results and Discussion

X-ray photoelectron spectroscopy (XPS) serves as a critical tool for analyzing the chemical composition and electronic states of materials. Specifically, when exploring how GO is reduced through 500 eV argon ion beam irradiation, XPS offers essential insights into chemical bond alterations and reduction levels. The goal of employing XPS in this scenario is to detail the variations in carbon (C) and oxygen (O) levels on the GO surface, pre- and post-irradiation. This is achieved by scrutinizing the C 1s and O 1s core-level spectra to detect shifts in functional groups from GO to rGO.

3.1. Examination of XPS Survey Spectra before and after 500 eV Argon Ion Beam Treatment

The XPS survey spectra of GO, both pre- and post-500 eV argon ion beam treatment, reveal significant shifts in carbon (C 1s) and oxygen (O 1s) peak binding energies and alterations in the carbon to oxygen (C/O) ratio. Initially, from 20 s of irradiation (shown in Figure 2b), there is a distinct rise in the intensity of the carbon peak and a decrease in the intensity of the oxygen peak compared to the scenario with no argon ion exposure (0 s, illustrated in Figure 2a). With continued exposure to argon ions, this pattern becomes more pronounced at longer durations (40 and 60 s, depicted in Figure 2c,d), indicating a continuous reduction process. By the time irradiation reaches 80 s (Figure 2e), this trend culminates in a significant shift, marking a deeper level of reduction.

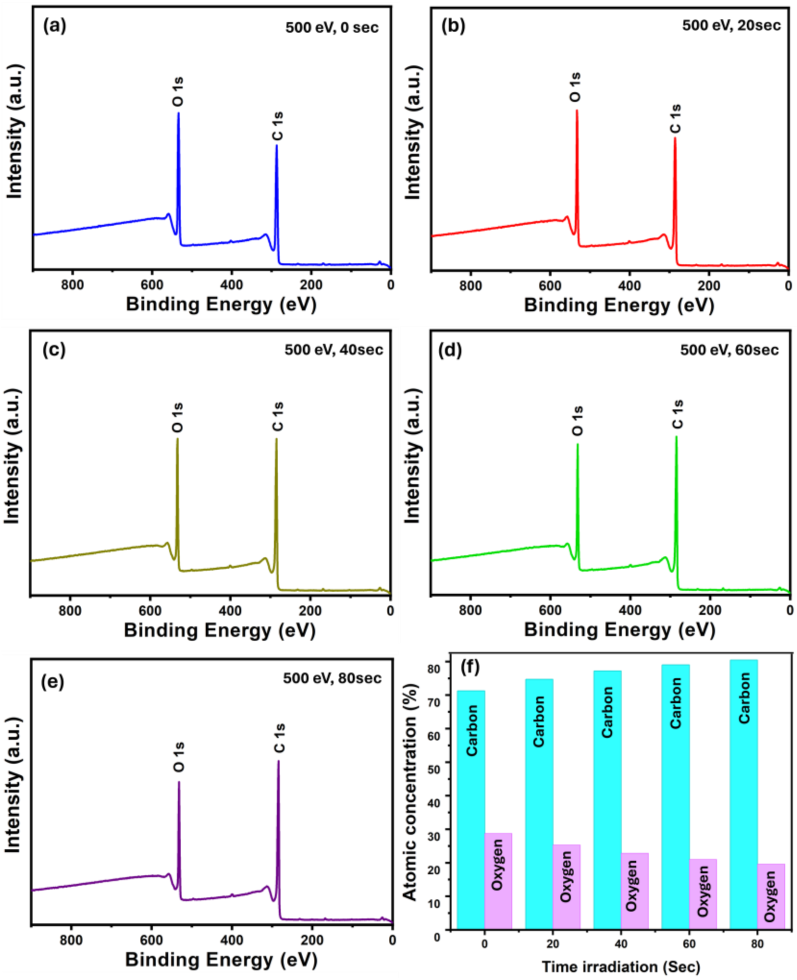

Figure 2. XPS survey spectra of GO before exposure to 500 eV Ar beam irradiation at (**a**) 0 s and after exposure to 500 eV Ar beam irradiation at (**b**) 20 s, (**c**) 40 s, (**d**) 60 s, and (**e**) 80 s time of irradiation. (**f**) Atomic percentage of carbon and oxygen elements at different irradiation times.

Elemental analysis depicted in Figure 2f highlights a notable shift in the C/O ratio from 2.48 at the initial 0 s, with the ratio progressively increasing as the duration of irradiation extends to 4.12 at 80 s. This alteration emphasizes a considerable reduction in the oxygen content on the surface, signaling a shift toward a significantly reduced state of GO. Moreover, the observed increase in carbon content, alongside a decrease in oxygen content from the 20 s to the 80 s, showcases the effectiveness of argon ion beam irradiation in increasing the carbon composition, reducing the oxygen presence, and notably enhancing the C/O ratio of the produced rGO. For clearer insight, Table 1 compiles the binding energy shifts, atomic concentration variations in carbon and oxygen, and the C/O ratio adjustments in GO following varying durations of 500 eV argon ion beam exposure, with minor binding energy shifts across different times due to surface charge influences.

Table 1. The elemental composition including binding energy, atomic concentration of carbon and oxygen, and C/O ratio.

Samples/Time Irradiation	Binding Energy (eV) Oxygen:Carbon	Atomic Concentration (%) Oxygen:Carbon	C/O Ratio
GO (0 s)	534.01:287.33	28.71:71.29	2.48
rGO (20 s)	532.84:286.00	25.28:74.72	2.96
rGO (40 s)	532.32:285.40	22.80:77.20	3.39
rGO (60 s)	532.13:285.09	20.97:79.03	3.77
rGO (80 s)	532.01:284.87	19.54:80.46	4.12

Table 1 demonstrates, through quantitative XPS analysis, an increase in the C/O atomic ratio from 2.48 in graphene oxide to 4.12 in reduced graphene oxide. This highlights the effective conversion of GO to rGO after exposure to various durations of 500 eV argon ion beam irradiation. This finding is consistent with XPS outcomes for rGO created using vitamin C as a reducing agent, further confirming the effectiveness of the reduction process [20].

3.2. High-Resolution Spectra of C 1s Acquired before and after Exposure of Graphene Oxide to 500 eV Argon Ion Beam

The high-resolution XPS C 1s spectra analysis of GO before and after exposure to a 500 eV argon ion beam reveals significant alterations in the chemical composition and bonding, indicative of the reduction process. Initially, the C 1s spectrum of untreated GO (Figure 3a) showcases peaks at approximately 285.16 eV for sp^3 and sp^2 carbon (C-C/C=C), which signifies graphitic domains, 287.38 eV for C-O bonds in epoxy groups, 289.53 eV for C=O bonds present in carbonyl groups, and a peak around 291.32 eV for O-C=O bonds in carboxyl groups [21–23]. Following 20 s of argon ion irradiation (Figure 3b), the reduction starts with diminished oxygen-related peak intensities and increases the graphitic peak, which becomes more pronounced. This trend intensifies with longer irradiation times (40, 60, and 80 s, Figure 3c–e), where a further decrease in oxygen-associated peaks occurs, underscoring the progressive restoration of the sp^2 carbon network (see Table 2). By 80 s, the XPS spectrum aligns closely with that of rGO, marked by minimal oxygen-related peaks and the dominance of graphitic carbon, signifying a thorough reduction.

These spectral changes, observed both quantitatively and qualitatively (see Figure 3 and Table 2), validate the efficacy of argon ion irradiation in reducing GO, showcasing a clear transition towards a structure resembling pristine graphene. This process highlights not only the reduction in oxygen-containing groups but also the enhancement of the graphitic character, affirming the potential of argon ion irradiation as a precise, controlled method for the sustainable production of high-quality rGO.

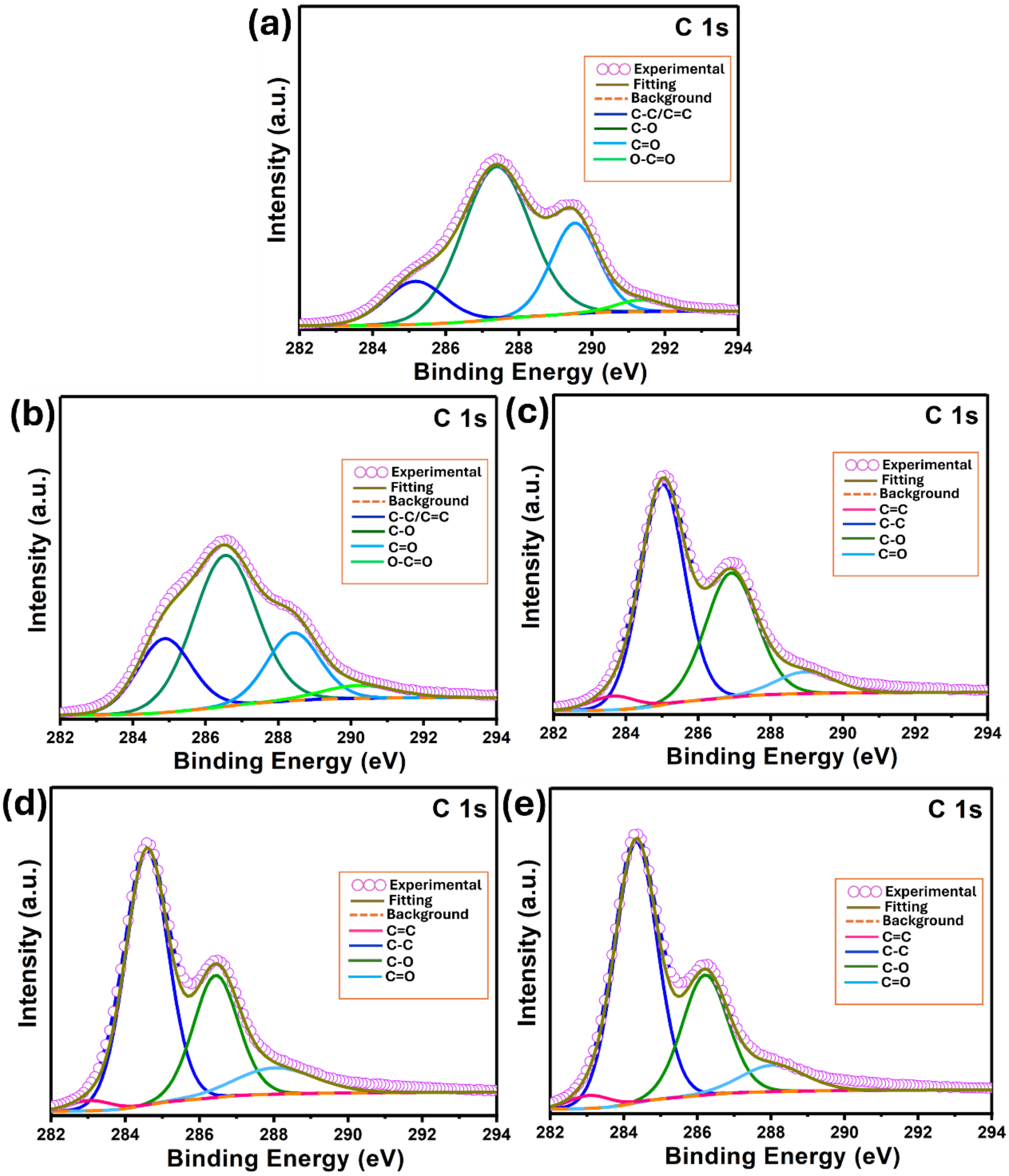

Figure 3. High-resolution X-ray photoelectron spectroscopy (XPS) spectra of carbon obtained from graphene oxide (GO) samples exposed to argon ion beam irradiation for varying durations, as displayed in panels (**a**–**e**), representing irradiation times of 0, 20, 40, 60, and 80 s, respectively.

Table 2. The functional groups in XPS C 1s spectra with binding energy reveal a shift towards a lower binding energy after exposure of graphene oxide (GO) samples to argon ion beam irradiation for varying durations.

Samples/ Time Irradiation	C=C/C-C Binding Energy (eV)	C-O Binding Energy (eV)	C=O Binding Energy (eV)	O-C=O Binding Energy (eV)
GO (0 s)	285.16	287.38	289.53	291.32
rGO (20 s)	284.87	286.54	288.42	290.21
rGO (40 s)	283.65/285.01	286.91	288.95	No peak
rGO (60 s)	283.06/284.58	286.44	288.02	No peak
rGO (80 s)	283.08/284.33	286.21	288.04	No peak

3.3. High-Resolution Spectra of O 1s Acquired before and after Exposure of Graphene Oxide to 500 eV Argon Ion Beam

The high-resolution XPS O 1s spectra of GO provide insightful details into the oxygen-containing functional groups both before and after exposure to a 500 eV argon ion beam, highlighting the dynamic changes in GO's chemical structure through the reduction process. Initially, the O 1s spectrum of GO (Figure 4a) showcases peaks corresponding to various oxygen states: a peak at approximately 533.02 eV is indicative of C=O (carbonyl groups), a peak around 533.8 eV is indicative of C-O (epoxy groups), and another at 535.06 eV is indicative of O-C=O (carboxyl groups), illustrating the diverse oxygen functionalities inherent in GO [24–26]. Upon irradiation for 20 s (Figure 4b), a noticeable shift is observed with a slight reduction in the intensity of these oxygen-related peaks, signaling the onset of the reduction process. As the exposure time extends to 40, 60 s (Figure 4c,d), and eventually 80 s (Figure 4e), the spectra exhibit a progressive decrease in the intensity of these peaks, particularly those associated with more easily reduced oxygen groups such as carboxyl and carbonyl functionalities. By 80 s, the O 1s spectrum markedly approaches that of rGO, with significantly reduced oxygen-related peak intensities (C=O and O-C=O) and a notable shift towards lower binding energies, reflective of the successful removal or transformation of oxygen-containing groups and the partial restoration of the graphene lattice (see Figure 4 and Table 3). This gradual alteration in the O 1s spectra not only evidences the efficacy of argon ion irradiation in reducing GO but also delineates the stepwise reduction pathway, with the extent of oxygen functionality reduction being directly correlated to the duration of argon ion beam exposure, ultimately facilitating the synthesis of rGO with reduced oxygen content and enhanced structural properties akin to pristine graphene.

Table 3. The functional groups in XPS O 1s spectra with binding energy reveal a shift toward lower binding energy after exposure of graphene oxide (GO) samples to argon ion beam irradiation for varying durations.

Samples/Time Irradiation	C=O Binding Energy (eV)	C-O Binding Energy (eV)	O-C=O Binding Energy (eV)
GO (0 s)	532.8	533.63	535.01
rGO (20 s)	531.91	533.23	534.67
rGO (40 s)	530.93	532.67	534.49
rGO (60 s)	530.43	532.24	534.08
rGO (80 s)	530.00	532.02	533.99

Interestingly, the initial presence of a peak at 535 eV in the O 1s spectrum is attributed to water molecules bonded to the hydrophilic surface of GO, which tend to mask the carboxyl group signals and diminish the epoxy group's signal strength. After 20 s of argon ion irradiation, this peak significantly diminishes or disappears due to the desorption of water and surface cleaning by the argon ions. This step marks a critical change in the surface of GO, leading to the emergence of carboxyl peaks at lower energy levels post-water removal, indicating the material's progression towards a structure akin to pristine graphene [25,27].

Additionally, the reduction in oxygen content modifies the surface properties of rGO, rendering it less hydrophilic than GO. Consequently, changes in the oxygen functional groups on the rGO surface led to a decrease in hydrophilicity and an increase in chemical inertness relative to GO. These changes enhance the suitability of rGO for applications that demand lower reactivity and greater stability.

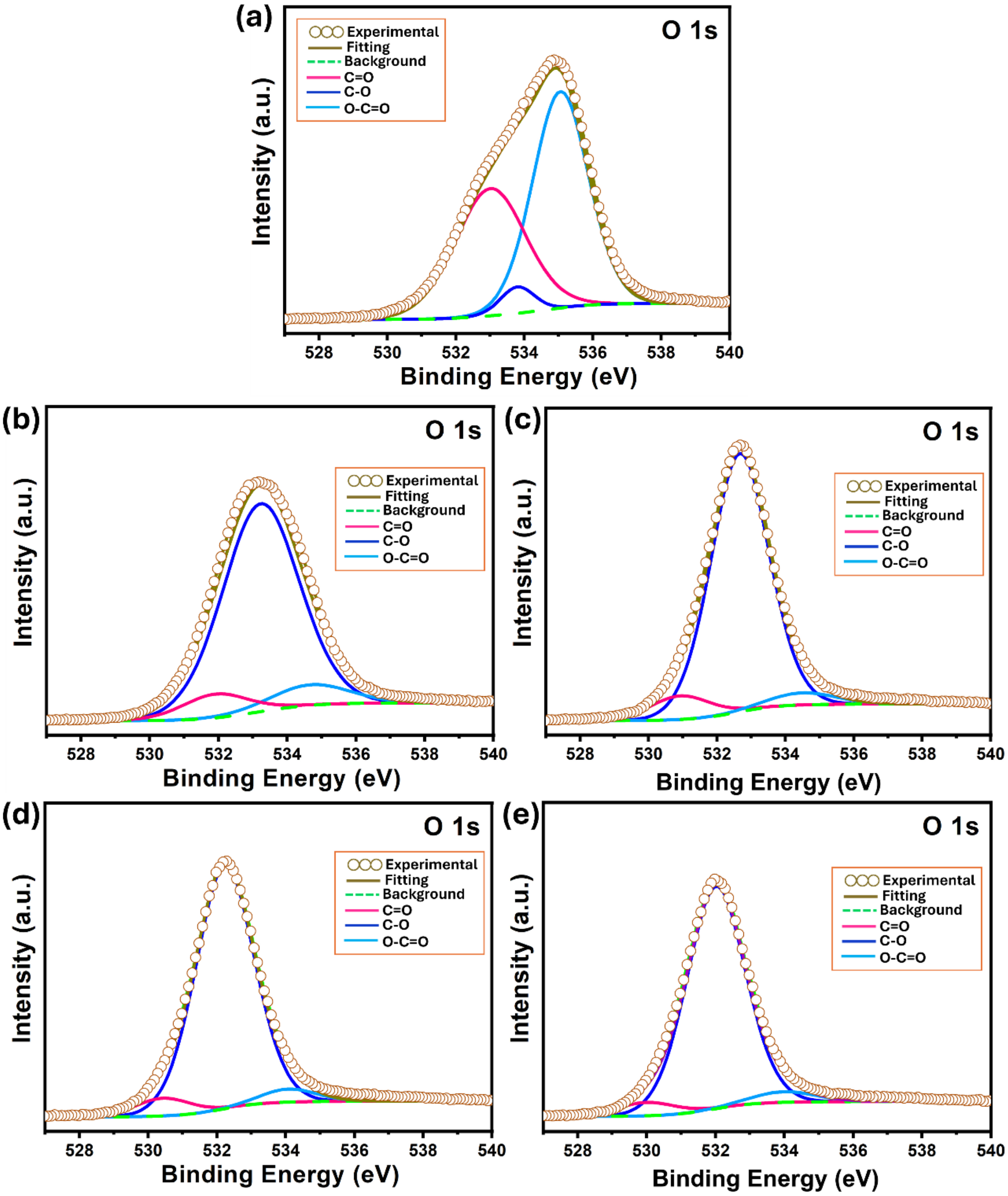

Figure 4. High-resolution X-ray photoelectron spectroscopy (XPS) spectra of O 1s obtained from graphene oxide (GO) samples irradiated by 500 eV argon ions for different durations, as shown in panels (**a**–**e**), representing irradiation times of 0, 20, 40, 60, and 80 s, respectively.

The XPS findings indicate that irradiation with 500 eV argon ions is an effective method for converting GO into rGO, significantly reducing oxygen-containing groups and reinstating the essential graphitic configuration. This technique not only proves the effectiveness of argon ion irradiation in producing graphitic rGO but also facilitates fine control over the material's properties, enhancing its versatility for a wide range of applications, from electronics to composite materials. The capability to tailor the duration of irradiation allows for the customization of rGO properties, highlighting the significance of argon ion irradiation in optimizing the material for sophisticated applications, thereby leveraging graphene's inherent attributes. Real-time monitoring with XPS confirms the efficacy of the reduction, ensuring detailed characterization. Additionally, argon ion patterning transcends mere reduction; it enables the creation of micro-scale rGO patterns directly on substrates, bypassing the need for conventional lithography or etching techniques, thus offering further application flexibility. Overall, our method distinguishes itself by permitting the precise adjustment of rGO characteristics through controlled irradiation times, which is a novelty in the field.

Furthermore, this research introduces argon ion beam irradiation as a sustainable approach for producing reduced graphene oxide (rGO), highlighting its benefits in terms of controlled synthesis and environmental friendliness. Incorporating argon ion beam irradiation into XPS equipment offers an economical avenue for large-scale production by combining material characterization with modification processes. A comparative analysis provided in Table 4 outlines how our method compares with other reduction techniques [20], focusing on the conditions of reduction and the carbon-to-oxygen ratio (C/O ratio) as key metrics for assessing efficiency.

Table 4. Comparison of the reaction conditions and the carbon to oxygen (C/O) ratio obtained through XPS analysis which exhibits the varying reduction efficiencies of rGO achieved through different reduction approaches.

Method	Reduction Conditions	(C/O) Ratio
Chemical reduction	Vitamin C, 80 °C	rGO = 1.80, rGO = 2.96
UV reduction	Xenon arc lamp, 24 h	rGO = 1.80, rGO = 2.80
Microwave reduction	800 W, 2 h, atmospheric	rGO = 1.80, rGO = 1.86
Argon ions beam	500 eV, 80 s	rGO = 2.48, rGO = 4.12

The carbon-to-oxygen ratio (C/O) ratio serves as an indicator of reduction efficiency across various methods, where a higher (C/O) ratio signifies a reduction in oxygen content within rGO. According to the data presented in Table 4, microwave reduction exhibits lower efficiency, with a (C/O) ratio of only 1.86. In comparison, UV reduction and chemical reduction using vitamin C demonstrate better performance, achieving (C/O) ratios of 2.96 and 2.80, respectively. Notably, the (C/O) ratio for argon ion beam reduction (as explored in our study) is 4.12, showcasing its superior capability to eliminate oxygen-containing groups and secure a more complete reduction. Consequently, argon ion beam reduction stands out as the most effective method for achieving a high degree of reduction efficiency. The results showcase that this method not only matches the reduction efficiency seen in conventional approaches but also stands out for its eco-friendliness and safety, avoiding the use of hazardous chemicals and high temperatures. This technique enables precise adjustments in the reduction and patterning processes, supports scalable manufacturing while keeping operational costs and throughput in mind, and produces high-quality rGO suitable for a wide range of uses.

4. Proposed Mechanism

Figure 5 illustrates the proposed mechanism for the reduction of GO to rGO using a 500 eV argon ion beam. The transformation of GO into reduced rGO through 500 eV argon ion (Ar+) beam irradiation is a multifaceted process involving physical and chemical interactions and possibly localized thermal effects. These interactions contribute to the effective removal of oxygen-containing groups, leading to the synthesis of high-quality reduced graphene oxide [19]. The interactions between argon Ar+ ions irradiation and GO that lead to the production of rGO involve complex processes at the atomic and molecular levels. Under argon ion beam exposure, the physical impact on the GO surface results in sputtering, predominantly removing lighter atoms like oxygen due to their lower binding energy compared to carbon atoms. This process significantly reduces the oxygen content on the GO surface. The energetic argon ions transfer their kinetic energy to the GO material through elastic and inelastic collisions at the atomic level [28]. The energy imparted by the Ar+ ions to the GO surface can break the bonds of oxygen-containing functional groups such as hydroxyl (–OH), epoxy (C–O), carbonyl (C=O), and carboxyl (–COOH) groups. This breaking of bonds is primarily due to the collisions that provide enough energy to overcome the bond dissociation energies of these functional groups, leading to their removal or transformation into molecules such as H_2O, CO, and CO_2 [29]. This process facilitates the strategic rearrangement of carbon atoms into the sp^2 hybridized lattice that typifies graphene, indicating a significant restructuring towards its characteristic structure.

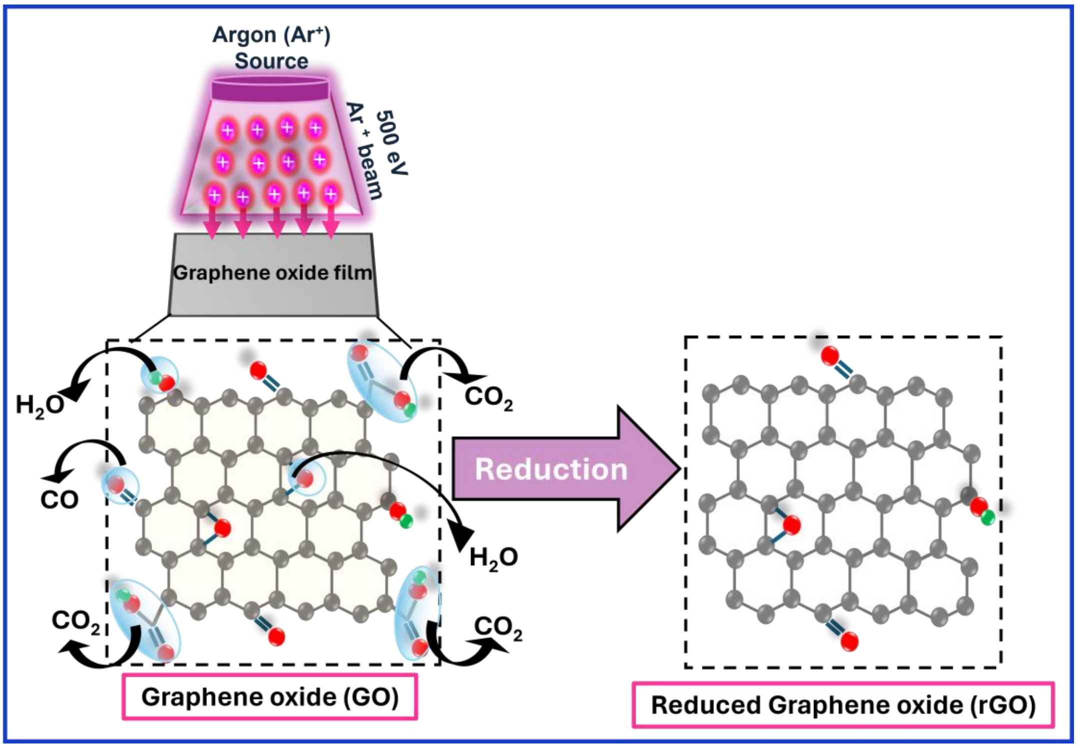

Figure 5. The schematic diagram has been created to illustrate the proposed mechanism for the reduction of GO to rGO using a 500 eV argon ions beam.

Besides direct bond breaking, the energy transferred from the Ar+ ions during the irradiation can cause localized heating of the GO material and generate secondary electrons, which break additional bonds of oxygen-containing functional groups [30]. By tuning the argon ion beam energy and exposure time, the degree of reduction can be precisely controlled. Longer irradiation times lead to a more complete removal of oxygen-containing groups and a higher C/O ratio, as evidenced by the XPS data presented in the study. The produced rGO, with reduced oxygen content and restored graphene structure, demonstrates the method's accuracy in materials engineering.

5. Conclusions

This study showcases the innovative use of a 500 eV argon ion beam for the dual purpose of surface cleaning and the reduction of GO to rGO, marking a significant advancement over traditional methods that rely on toxic chemicals. Ar ions enable selective patterning, allowing for the creation of micro-scale rGO patterns without traditional lithography or chemical etching. X-ray photoelectron spectroscopy (XPS) analysis of the irradiated GO revealed a systematic increase in the carbon content and a corresponding decrease in the oxygen content with prolonged irradiation times. Specifically, C 1s high-resolution XPS spectra highlighted the diminishing presence of O-C=O signals post-irradiation, confirming the effective reduction of GO. Additionally, changes in the intensities of C=C/C-C, C-O, and C=O signals in C 1s and O 1s high-resolution spectra were observed across different irradiation periods. The process not only efficiently targets and removes oxygen-containing functional groups, leading to the formation of carbon–carbon bonds and the restoration of the sp^2 hybridized graphene lattice, but also transitions the material towards graphitic carbon. This enhances the electrical conductivity and mechanical strength of the resulting rGO, as demonstrated by the detailed XPS spectra analysis. Although further research is required to fully understand the subtle impacts of the argon ion beam technique, it emerges as an innovative, environmentally conscious, and precise method for generating high-quality rGO. Looking ahead, further exploration of the method's scalability and sustainability in producing high-quality rGO is warranted. Future research should focus on optimizing irradiation conditions and ion dosage, extending the approach to other 2D materials for wider applications, and integrating it with existing synthesis processes. Advancements in argon ion beam technology could enhance cost-effectiveness and energy efficiency for industrial use, paving the way for innovative and sustainable material synthesis in various applications.

Author Contributions: Conceptualization, T.F.Q.; Methodology, S.A., T.F.Q. and A.M.A.; Software, A.M.A.; Validation, A.M.A. and S.B.; Formal analysis, S.T.H. and D.S.; Investigation, N.A.; Resources, N.A., S.B. and D.S.; Data curation, T.O.O. and S.T.H.; Writing—original draft, S.A. and A.M.A.; Writing—review & editing, T.F.Q. and T.O.O.; Supervision, T.F.Q.; Project administration, T.F.Q.; Funding acquisition, T.F.Q. All authors have read and agreed to the published version of the manuscript.

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