

Article

Laser Modification of Graphene Oxide for Efficient Oxygen Evolution Reaction

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Abstract

The Oxygen Evolution Reaction (OER) is the bottleneck in the water splitting reaction since it involves four intermediate steps, constituting the adsorption–desorption of oxygen-based radical groups, and not all of them are energetically favorable. Rapidly growing research interest is focusing on carbon-based materials as novel, highly active and durable non-precious electrocatalysts for the OER, representing a valuable alternative to precious and rare materials with electrochemical properties tuned by defect creation. In this work, we propose a facile and green methodology based on the modification of graphene oxide by laser irradiation to obtain an alternative OER catalyst. GO flakes were chemically and physically modified using pulsed laser irradiation at 532 nm with fluences of 1.5 J/cm² and 2.5 J/cm². Different analyses were carried out to correlate the electrochemical performance with the structural, optical, and morphological properties; after that, we correlated the improvements in the OER with respect to the pristine GO with the increase in OH functional groups obtained by laser treatment. The best-performing sample exhibited an overpotential of 380 mV, comparable to that of catalysts reported in the literature but with the advantage of not being a precious or rare material.

Keywords: graphene oxide; water splitting; OER; laser irradiation



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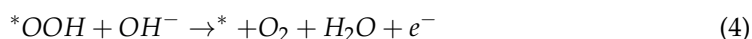
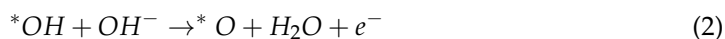
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1. Introduction

Nowadays, water electrolysis is intensively investigated and considered a key process for industrial production of hydrogen because of the zero greenhouse emissions produced and the abundance of water. This process consists of the simple action of dividing water molecules into hydrogen and oxygen by applying an electric potential to a water solution placed between two electrodes [1–3], with a theoretical applied voltage of 1.23 V. The actual process requires an additional applied voltage, called the overpotential, due to the various electrochemical reactions that take place on both electrodes, which are not always energetically favorable. On the negatively charged electrode, the so-called Hydrogen Evolution Reaction (HER) occurs, where, in the first step, a hydrogen atom from a water molecule is adsorbed on the electrode surface, and in the second step, it binds to another

adsorbed atom to be released as H_2 gas [1–3]. On the positively charged electrode, the Oxygen Evolution Reaction (OER) takes place, which is made up of four steps that involve the formation of $^*OH$, $^*O$ and $^*OOH$ groups before the release of O_2 gas [4–6], as described below:



where * represents the active site on the electrode. Not all of these steps require the same amount of additional energy, and depending on the catalyst, the rate-determining step can be Equation (2) or (3) [5,7]. The sluggishness of the OER process represents a bottleneck for water electrolysis in terms of overpotential and performance. Hence, the choice of catalyst for future industrial applications is fundamental in order to obtain great performance, to face the energy crisis and significant climate changes, and to move towards sustainable energy technologies. The state-of-the-art electrocatalysts reported for the OER/HER are based on precious and rare materials, like Pt for the HER and RuO_2 or IrO_2 for the OER [8], whose availability is decreasing and whose production requires expensive processes that are not environmentally friendly. As a consequence, valuable alternatives are required, such as novel, highly active and durable non-precious electrocatalysts, especially for the OER, and various examples are reported in the literature based on transition metals and their oxides [2–4,7–10].

Recently, researchers have shown growing interest in carbon-based materials for water electrolysis, particularly for the OER. In fact, the number of publications (and patents) in Scopus or Web of Science related to water splitting processes involving these materials has reached more than 15,000 in the last 4 years [6,11]. The key factors promoting this interest in carbon-based materials are their multiple advantages and features, such as abundant sources, high electronic conductivity, tunable molecular structures, and environmental compatibility. As a result, the efforts of researchers worldwide revolve around ideas, methods and innovative strategies to design high-performance carbon-based electrocatalysts: single- or multi-heteroatom doping, surface functionalization, structural defect engineering, edge modulation, porous structure and morphology engineering, and carbon hybridization (coupling carbon with other active components, such as metals, non-metals or other types of carbon) [12–17]. Many preparation methods (e.g., biomass pyrolysis, chemical vapor deposition, hydrothermal carbonization [6]) show many disadvantages, such as high temperatures; harsh conditions for realizing a specific composition, structure, or morphology; or relatively expensive chemicals, which can potentially preclude or hinder large-scale production and commercialization.

The carbon-based materials obtained (e.g., doped graphene flakes, quantum dots, reduced graphene oxides) are reported as co-catalysts in combination with metal composites, acting as efficient boosters for electrochemical water splitting [18–20] and sun-driven photoelectrochemical water splitting [21–23], but their performance alone as catalysts has not been studied. Also, defect creation in graphene and nanographene structures is reported to promote the Oxygen Reduction Reaction (the inverse reaction of the OER) by facilitating the adsorption of the intermediate species [24] following the same intermediate step of the OER in reverse order.

Among carbon-based materials, graphene oxide (GO) and its reduced form have recently attracted attention due to their simple fabrication methods, high availability [25,26] and unique properties provided by edges and functional groups on the surface [27], such

as epoxy, hydroxyl, carboxyl and carbonyl groups. Among the various techniques for modifying GO, pulsed laser irradiation is suitable for altering structural, chemical, and electrical properties in a fast and versatile process [28]. Laser treatment of GO deposited on a substrate has been reported to improve its electrical conductivity [29–31]. Other literature works obtain the graphene-based catalyst layer by laser scribing on a plastic substrate [32,33]. GO irradiation in liquid leads to both a chemical modification (in terms of changing the functional groups) and a physical modification (in terms of size and morphology and crystallinity) with smaller dimensions and eventually holes in the structure [34–37], probably due to release of CO_2 or CO [38,39] as a consequence of the high local temperature reached by the flakes during the laser process [40]. Laser irradiation in liquid is considered a green and facile technique since it works at ambient pressure and temperature; irradiation is directly performed on the liquid solution without the use of additional chemicals and without producing waste [41–43].

The aim of this work is to study the effects of modified carbon materials (using GO as a precursor) as a catalyst in the OER and to prove the effectiveness of using laser irradiation in liquid to produce carbon nanostructures competitive with materials reported in the literature from electrocatalytic, environmental, and commercial perspectives. The proposed method starts with a commercial GO solution and uses laser irradiation to induce controlled morphological and chemical modifications that positively affect the OER without a complex setup or chemical reagents.

2. Experimental Section

2.1. Sample Preparation

A commercial solution of GO flakes in water was purchased from Sigma-Aldrich, St. Louis, MO, USA and was diluted in DI water to obtain a concentration of 0.1 mg/mL.

Pulsed laser irradiation was performed on 5 mL of the diluted GO dispersion using the second harmonic (532 nm) of a Quanta-Ray Pro Series Nd:YAG laser (Spectra-Physics, Milpitas, CA, USA). The solution was placed in a small beaker with a magnetic stirrer and irradiated with 10 ns pulses at a frequency of 10 ns for 45 min with a fluence of 1.5 J/cm^2 or 2.5 J/cm^2 , based on the experimental setup and results of a previous work [34]: a chemical reduction with effects on the functional groups is expected at low fluence, while a morphological modification is also expected at higher fluence. The laser beamline consisted of a tunable beam splitter to modulate the output power and an aperture diameter of 0.5 cm.

In this manuscript, samples obtained from the diluted commercial dispersion will be referred to as $\text{GO-}0.0 \text{ J/cm}^2$, while the ones from the laser-treated dispersion will be referred to as $\text{GO-}1.5 \text{ J/cm}^2$ and $\text{GO-}2.5 \text{ J/cm}^2$.

Nickel foam (NF) substrate (Goodfellow Inc., Delson, Quebec, thickness 1.6 mm, porosity $\geq 95\%$) was cut into rectangles of $2 \times 1 \text{ cm}^2$. Three samples were prepared by dropcasting $100 \mu\text{L}$ of each solution on the bottom 1 cm^2 of the substrate and dried on a hot plate at 95°C for 45 min. The temperature and time were chosen to ensure complete evaporation of the MilliQ water. The reference bare substrate electrode without GO decoration (referred to as *NF-ref*) was prepared by dropcasting only $100 \mu\text{L}$ of MilliQ water.

2.2. Physical and Chemical Characterization

An optical study was conducted using a PerkinElmer LAMBDA 1050+ (PerkinElmer, Shelton, CT, USA) UV-Vis-NIR spectrophotometer. For each sample, the measurement was conducted by placing the solution into a quartz cuvette (1 cm optical path) and measuring the absorbance in the range between 200 nm and 800 nm.

A total of 20 μL of each solution was dropcasted on silicon for XPS analysis and on gold-coated silicon for Raman analysis. A layer of ~ 70 nm of gold is needed to screen the silicon signal during the Raman spectral acquisition and obtain a less noisy signal [44].

X-ray photoelectron spectroscopy (XPS) analysis was conducted using a PHI Genesis XPS (ULVAC-PHI Inc., Chigasaki, Japan) instrument with an $Al\ K\alpha$ X-Ray source (1486.6 eV) and a 180° hemispherical analyzer. Raman analysis was conducted with a HORIBA iHR550 (Horiba, Kyoto, Japan) spectrometer equipped with a He-Ne laser (wavelength 632.8 nm).

Morphological analysis was conducted with a Zeiss SUPRA 25 (Zeiss, Oberkochen, Germany) Scanning Electron Microscope (SEM) on the sample prepared for Raman and electrochemical analyses. In the first case, the gold layer offers a higher contrast between the substrate and GO flakes.

2.3. Oxygen Evolution Reaction Activity

The OER was analyzed in a three-electrode electrochemical cell using aqueous 1 M KOH (Sigma-Aldrich) alkaline electrolyte ($\text{pH} = 14$) with an Admiral Squidstat Plus (Admiral Instruments, Tempe, AZ, USA) potentiostat. The electrolyte is chosen to align with industrial alkaline standards [45].

The prepared sample was used as the working electrode, platinum wire as the counter electrode and Ag/AgCl 3.5 M KCl HI5311 (Hanna Instruments, Park East Drive, Tempe, AZ, USA) as the reference electrode. Cyclic voltammetry was performed in the range -1.3 V to 0.8 V with respect to the reference electrode at a scan speed of 50 mV/s in order to activate the sample surface and stabilize the sample. According to previous tests, 5 cycles are sufficient to stabilize the working electrodes. Then linear sweep voltammetry (LSV) was performed at 10 mV/s in the same voltage range as the reference electrode. Electrochemical Impedance Spectroscopy (EIS) was performed at the potential referred to a current of 10 mA using a 10 mV oscillating signal in the range 10 kHz – 1 ns . The measured potential vs the reference electrode (E_{AgCl}) was converted to the reversible hydrogen electrode (E_{RHE}), and then the overpotential η was obtained by correcting the potential drop with the measured current i and the uncompensated resistance $R_0[\Omega]$ (which quantify the electrolyte and the connection resistance) obtained from EIS, where the semicircle intercepts the real axis in the Nyquist plot [46,47]:

$$E_{\text{RHE}}[\text{V}] = E_{\text{AgCl}} + 0.205 + 0.059 \cdot \text{pH} - 1.23 \quad \rightarrow \quad \eta[\text{V}] = E_{\text{RHE}} - iR_0 \quad (5)$$

The overpotential at 10 mA/cm^2 is reported as a key parameter in evaluating electrode efficiency, as it is a standard reported in the literature and allows comparison with literature materials [8,47].

In particular, the lower the overpotential value at 10 mA/cm^2 , the lower the electrical power required for the reaction. The mass activity is defined as 10 mA/cm^2 over the catalyst mass [mg/cm^2], and therefore, catalysts with better performance show lower overpotential values and higher mass activity [8,47].

3. Results and Discussion

3.1. Optical Characterization

At a first glance (Figure 1 Inset), the three solutions present strong differences: the original solution, $\text{GO-}0.0\text{ J/cm}^2$, is dark brown, as is $\text{GO-}1.5\text{ J/cm}^2$, while $\text{GO-}2.5\text{ J/cm}^2$ is transparent. The color transition occurred after ~ 20 min of the laser process. Figure 1 reports absorbance measurements that reveal a peak in the UV region at 220 nm with a small shoulder at $\sim 300\text{ nm}$ for pristine and 1.5 J/cm^2 -irradiated GO samples. The former is associated with the electronic transition $\pi - \pi^*$ due to $\text{C} = \text{C}$ bonds, and the latter

is associated with the electronic transition $n - \pi^*$ attributed to the various functional groups [34,48]. GO-2.5 J/cm² presents a lower intensity, with a significant blue shift in both peaks assigned to the two electronic transitions due to the formation of holes and defects in the flake structures, as already studied theoretically in a previous work [34]. The decrease in the UV peak can be attributed to a reduction in the size of the sp^2 domains, which can result from a physical reduction or chemical sp^3 hybridization, as discussed in the following sections. The spectra after 350 nm present only small artifacts and no relevant peaks.

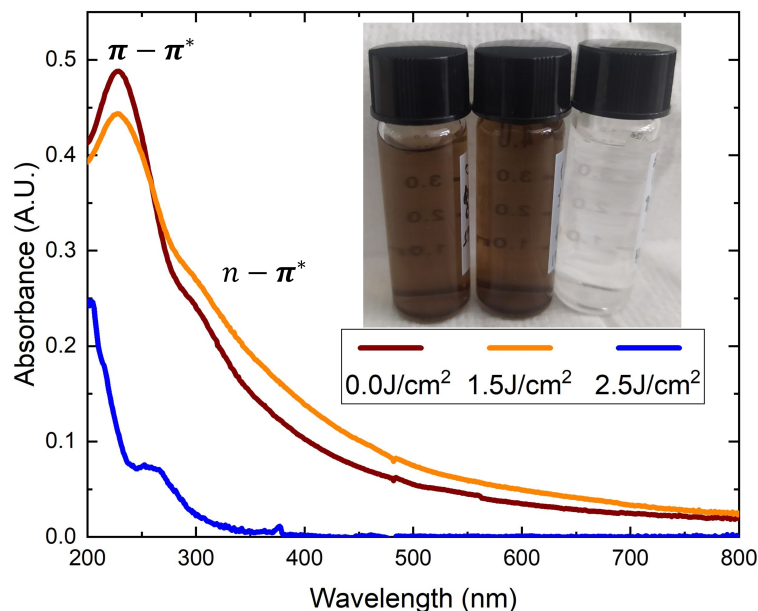


Figure 1. UV-Vis absorption spectra of the three samples with marked electronic transitions [34]. Inset: A photo of the three dispersions: left, GO-0.0 J/cm²; center, GO-1.5 J/cm²; right, GO-2.5 J/cm².

3.2. Raman Spectroscopy

Raman spectra of the three samples were acquired in a wide range, but only the region between 1200 cm^{−1} and 1800 cm^{−1} is reported in Figure 2 since there are no other peaks visible in the extended range. The G peak is related to an ordered graphene-like structure, while the D peak originates from phonon interaction with defects and so represents the disorder [49]; their positions are respectively $G \simeq 1580$ cm^{−1} and $D \simeq 1350$ cm^{−1} in highly ordered graphene. In the reported cases, the D peak is much larger and broader than the G peak, indicating the presence of structural or chemical defects, such as edges or functional groups typical of GO. The noise is lower for the pristine material because bigger flakes are involved in the analysis. The deconvolution reported in Figure 2 shows the real shape of the peaks: D (red lines) and G (blue lines) peaks are evident respectively at ~ 1330 cm^{−1} and ~ 1590 cm^{−1}. This shift in all samples can be attributed to quantum confinement arising from the small size of the crystalline domains within the GO flakes [49]. To complete the picture, a third peak (green lines) is included: it is reported in the literature as D'' [50] and has a controversial nature but is experimentally related to the oxygen present in GO and rGO. This peak shifts from ~ 1540 cm^{−1} in pristine GO to ~ 1480 cm^{−1} in the laser-treated samples, pointing to the decrease in the amount of oxygen in the sample and also the higher crystallinity. The latter is confirmed by a lower I_D/I_G ratio after laser treatment, as reported in Figure 2: GO-0.0 J/cm² has $I_D/I_G = 1.8$, GO-1.5 J/cm² has $I_D/I_G = 1.2$, and GO-2.5 J/cm² has $I_D/I_G = 1.5$.

3.3. Scanning Electron Microscopy

The scanning electron microscopy image presented in Figure 3a displays a well-defined polygonal shape for commercial GO, highlighted by the contrast with the gold-coated silicon used as a substrate for Raman measurements. The edge size varies between 100 nm and 2 μm ; moreover, there are zones with well-dispersed flakes and zones with agglomeration as a consequence of dropcasting.

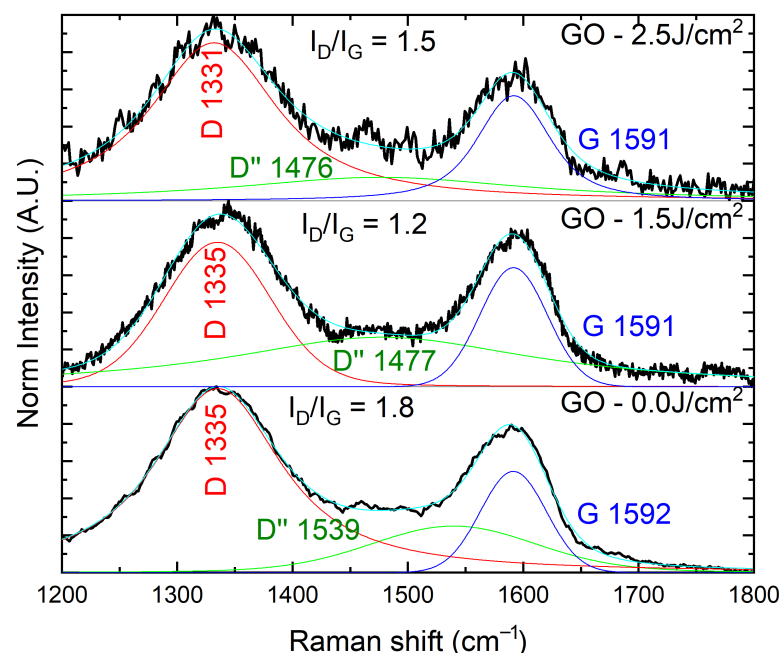


Figure 2. Deconvoluted Raman spectra of the three samples (GO-0.0 J/cm² on bottom, GO-1.5 J/cm² in between, GO-2.5 J/cm² on top) in the region containing typical carbon bands (1200–1800 cm^{−1}). Black line: Experimental data; red line: D peak; blue line: G peak; green line: D'' peak; cyan line: fit sum. The figure reports the peak positions and I_D/I_G ratios for all the samples.

After the laser treatment at 1.5 J/cm² (Figure 3c), the flakes appear jagged, maintaining almost the same size. With the fluence increased to 2.5 J/cm² (Figure 3e), the flakes lose contrast with respect to the background and appear fragmented and smaller. The bigger ones have some holes inside, as previously observed and reported [34]. They mostly appear aggregated in clusters on the order of μm surrounded by small flakes as proof of laser-induced fragmentation.

Figure 3b,c,f show the appearance of nickel foam after the 100 μL dropcasting of the three solutions. They have the same morphology as that observed on the flat substrate, but due to the porous 3D nature of NF, the flakes are uniformly distributed and almost cover the substrate surface, with some flakes partially overlapping.

More SEM images are provided in Supporting Information (Figures S1–S6).

3.4. X-Ray Photoelectron Spectroscopy

To fully understand the surface chemical changes induced by laser irradiation, XPS was conducted on GO samples before and after the laser process. Carbon (C1s)- and oxygen (O1s)-related peaks are reported in Figure 4, while the results of C1s and O1s peak deconvolution are summarized in Table 1 and Table 2, respectively [51,52]. The effects induced by laser irradiation are evident in the C1s peak in Figure 4a,c,e: for the pristine GO, four contributions were identified, associated with C – C, C – O, C = O, and COOH bonds, with binding energies of 284.4 eV, 286.5 eV, 288.8 eV, and 290.4 eV, respectively. By increasing the laser fluence, the relative intensities of the peak components

change, as reported in Table 1. In particular, the sum of $C - O$, $C = O$, and $COOH$ contributions decreases from 54.8% (for pristine GO) to 48.7% (after irradiation at 1.5 J/cm^2) and, finally, to 29.7% (after irradiation at 2.5 J/cm^2), indicating that a reduction process occurred due to a reduction effect. At the highest laser fluence, the signal associated with $COOH$ groups (binding energy $\sim 290 \text{ eV}$) becomes undetectable, and a new signal at lower energies due to C vacancies and/or CH ($\sim 283 \text{ eV}$) appears as a consequence of further flake fragmentation and edge/defect formation, suggesting that the reduction mechanism involves the production and subsequent release of CO_2 or CO [38,39]. This explains the jagged appearance and fragmentation of GO flakes observed by the SEM. The $O1s$ XPS spectra for all the samples are reported in Figure 4b,d,f, and the fitting procedures allow the identification of different contributions associated with $O - C = O$ at $\sim 530 \text{ eV}$, $O = C$ at $\sim 531 \text{ eV}$, $O - C$ bonds (hydroxyls, epoxides) at $532\text{--}532.5 \text{ eV}$, and O bound to an aromatic carbon atom (phenols, OH at the edge in the case of graphene oxide) at about 533.5 eV . The % values obtained for each contribution and the corresponding binding energy values are reported in Table 2: as the fluence is increased, the $C = O$ contribution decreases from pristine GO to laser-treated GO, whereas $C - OH$ functionalities (hydroxyls, phenols, OH at the edge) increase. Therefore, the laser process not only reduces the total amount of oxygen but also induces a change in the relative amounts of the different functional oxygen groups, with a final predominance of OH functionalities.

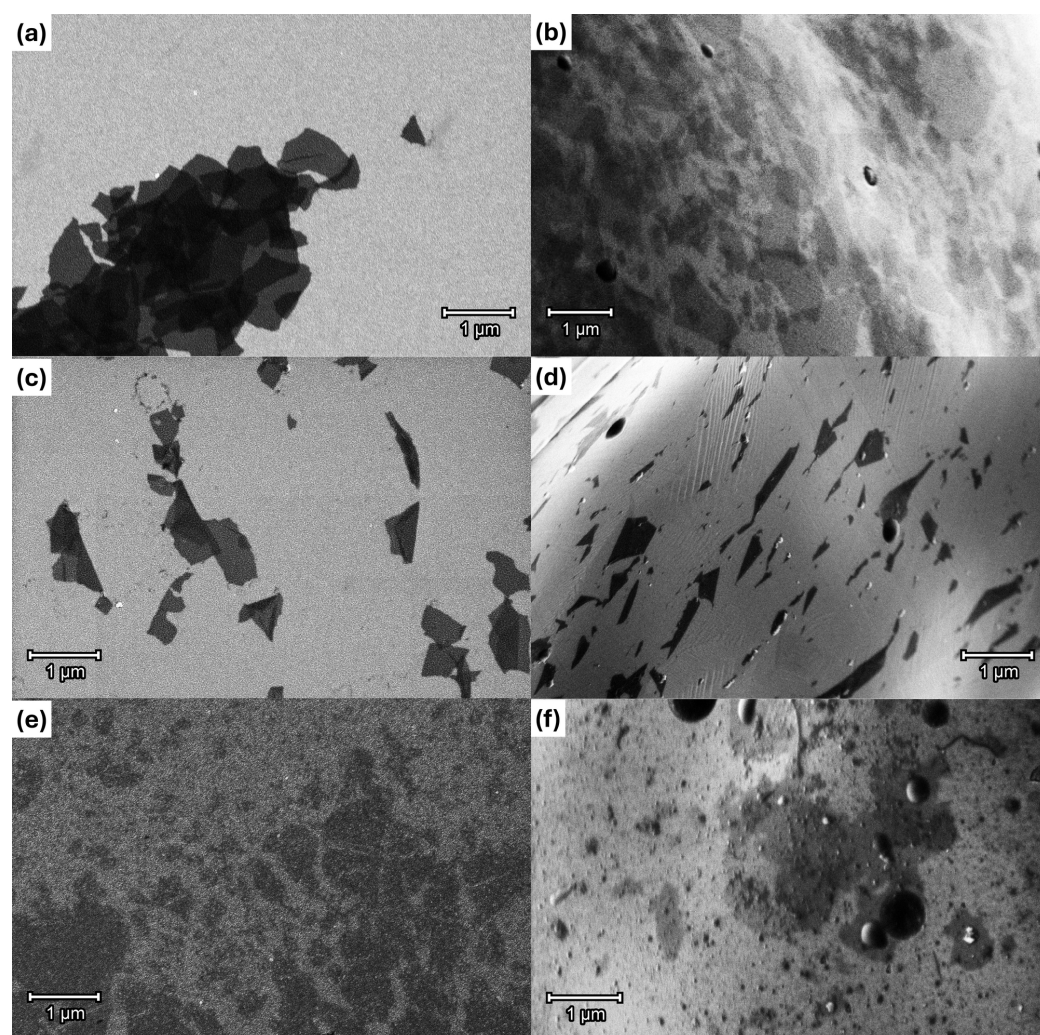


Figure 3. SEM images of $GO-0.0 \text{ J/cm}^2$ on (a) gold-coated silicon and (b) nickel foam. SEM images of $GO-1.5 \text{ J/cm}^2$ on (c) gold-coated silicon and (d) nickel foam. SEM images of $GO-2.5 \text{ J/cm}^2$ on (e) gold-coated silicon and (f) nickel foam.

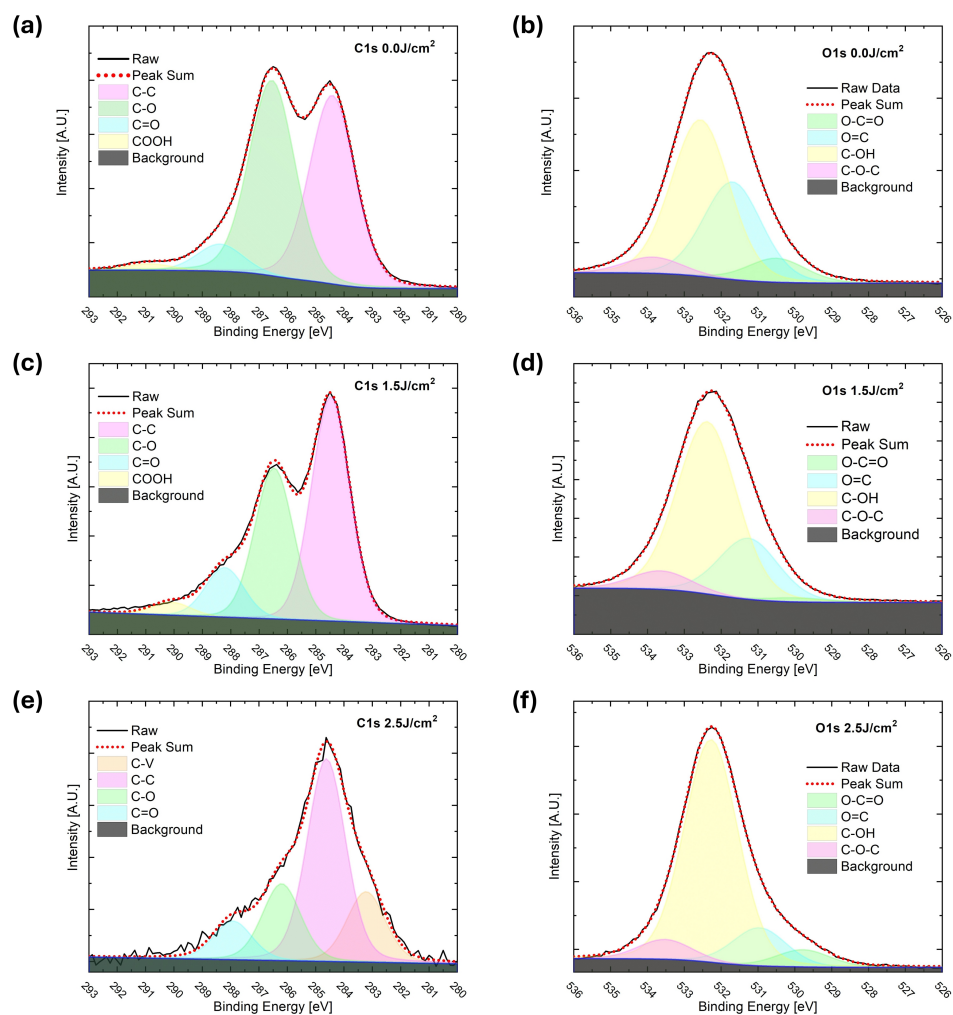


Figure 4. Deconvoluted XPS spectra of: GO-0.0 J/cm² (a) C1s and (b) O1s; GO-1.5 J/cm² (c) C1s and (d) O1s; GO-2.5 J/cm² (e) C1s and (f) O1s.

Table 1. Relative amounts (%) of different contributions resulting from the C1s peak deconvolution and related binding energy (BE) values.

C1s Peak	GO—0.0 J/cm ²		GO—1.5 J/cm ²		GO—2.5 J/cm ²	
Peak Component	BE (eV)	%	BE (eV)	%	BE (eV)	%
Vacancy/CH	-	-	-	-	283.2	18.1
C—C	284.4	45.2	284.5	51.3	284.6	85.1
C—O	286.5	46.8	286.5	34.5	286.2	19.8
C=O	288.8	6.5	288.2	11.3	287.9	9.9
COOH	290.4	1.5	290.1	2.9	-	-

FT-IR analysis was also conducted to obtain information about the functional groups, as reported in Figure S7. The laser-treated sample exhibits the presence of CH₃ and CH₂ in the wavenumber region around 2900 cm⁻¹ and C—OH at 1190 cm⁻¹ and new peaks at 612, 699, 752, and 855 cm⁻¹, which are attributed to out-of-plane C—H bending vibrations in conjugated and aromatic rings. Both GO-1.5 J/cm² and GO-2.5 J/cm² present enhanced features with respect to the pristine sample in the spectral region between 900 and 1400 cm⁻¹, associated with C—O stretching vibrations of alcohols and epoxides, in agreement with XPS results. The emergence of these features indicates the formation of structural defects within the GO flakes. Laser irradiation, in general, induces local heating,

leaving defect and edge sites that are compensated for by interaction with the external environment [37]. Laser irradiation at 2.5 J/cm^2 also induces holes and fragmentation of the flakes, reducing the ordered domain size and, consequently, the previously reported features. Moreover, this kind of morphological effect is reflected in the appearance of peaks around 2900 cm^{-1} , associated with aliphatic $\text{C}-\text{H}$. This feature, in particular, is also observed through XPS, with the appearance of a peak at 283.2 eV associated with vacancies and/or CH .

Table 2. Relative amounts (%) of different contributions resulting from the O1s peak deconvolution and related binding energy (BE) values.

O1s Peak	GO— 0.0 J/cm^2		GO— 1.5 J/cm^2		GO— 2.5 J/cm^2	
Peak Component	BE (eV)	%	BE (eV)	%	BE (eV)	%
$\text{O}-\text{C}=\text{O}$	530.5	8.1	529.9	1.6	529.8	5.8
$\text{O}=\text{C}$	531.7	33.2	531.3	23.9	530.9	12.8
$\text{O}-\text{C}$ (hydroxyls, epoxides)	532.6	53.2	532.4	67.3	532.3	74.8
O —aromatic carbon (phenols, OH at the edge)	533.9	5.5	533.6	7.2	533.5	6.6

3.5. OER Performance

Electrochemical measurements show the strong OER catalytic activity of the laser-treated samples with respect to the nontreated ones and the reference, as shown by the LSV results in Figure 5a. All the curves present a small peak around $\sim 0.1 \text{ V}$ due to nickel substrate oxidation. The reference substrate (NF-ref) has an overpotential of 479 mV at 10 mA/cm^2 , which decreases to 418 mV when the commercial GO is loaded. A further improvement is provided by the laser treatment: GO- 1.5 J/cm^2 reaches 397 mV , and GO- 2.5 J/cm^2 reaches 380 mV .

EIS spectra are acquired at the overpotential referred to a current of 10 mA to study the interface during the OER process. Data are fitted with a simple Randles circuit using the algorithm proposed in [53] and reported in Supporting Information (Figure S8). This equivalent circuit consists of a resistance R_0 that quantifies the electrolyte and connection resistance in series with another resistance R_{ct} related to the charge transfer at the electrode/solution interphase and of a constant phase element in parallel with the two resistances. The latter ($Z_{CPE}(\omega) = 1/Q(i\omega)^\alpha$) is used to model the generalized double-layer capacitance [46].

All the samples present semicircular behavior that starts at $R_0 \sim 1.6 \Omega$. After fitting, they have a common value of $Q \simeq 0.01$, while α increases from 0.70 (pristine) to 0.71 (1.5 J/cm^2) to 0.72 (2.5 J/cm^2), indicating that the nature of the interface differs and tends to the ideal capacitor. The charge transfer resistance R_{ct} decreases from 5.7Ω (pristine) to 4.7Ω (1.5 J/cm^2) to 3.9Ω (2.5 J/cm^2), displayed as smaller semicircles as a sign of increasing reaction kinetics. A Tafel plot is shown in Figure 5b, reporting the log of current density vs the overpotential. The linear zones of the plot are fitted, revealing a slope that decreases from 75 mV/dec (pristine) to 70 mV/dec (1.5 J/cm^2) to 65 mV/dec (2.5 J/cm^2), confirming the faster reaction kinetics [54].

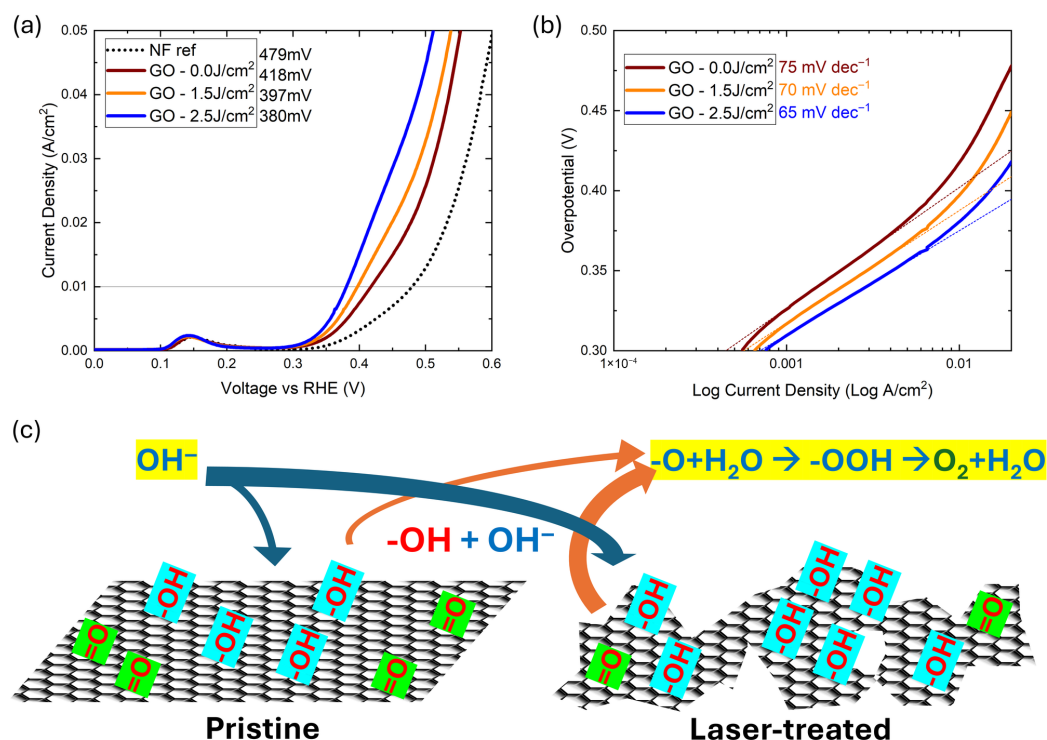


Figure 5. (a) Linear sweep voltammetry with reported values of overpotential at 10 mA/cm^2 (marked by horizontal line); (b) a Tafel plot with linear regions marked and Tafel slopes reported; (c) visual representation of the difference between the pristine and laser-treated GO flakes in terms of active sites for the OER.

The mass of catalyst material was estimated taking into account the initial GO concentration of 0.1 mg/mL , so the sample $\text{GO}-0.0 \text{ J/cm}^2$ consists of just $10 \text{ }\mu\text{g/cm}^2$ of material. The catalyst mass for all the samples was also estimated at $10 \text{ }\mu\text{g/cm}^2$; hence, a mass activity of 1 A/mg is reported in Figure S9 compared with some literature data [8]. All samples in this work are located in the high part of the graph, thanks to the low amount of used material, and show overpotential values within the literature range. As suggested by the low intensity of UV-Vis spectra and SEM analysis, the fragmentation and hole formation during laser irradiation with 2.5 J/cm^2 induce a mass decrease since some carbon is released as CO_2 or CO [38,39]. Even if the mass is overestimated in the laser-treated samples, there is an overpotential improvement and faster observed kinetics, especially in the $\text{GO}-2.5 \text{ J/cm}^2$ sample, demonstrating the effectiveness of the process on a commercial material that is already performing, with the advantage of using a very low mass.

Unlike conventional metal-based catalysts, where well-defined active centers govern the reaction pathway [4,7], the OER on graphene oxide is expected to proceed through a heterogeneous distribution of catalytic sites associated with oxygen functional groups and structural defects, as in other reactions [24,55,56]. In this framework, laser irradiation induces two key modifications: (i) a redistribution of oxygen-containing functional groups, with a relative increase in hydroxyl ($-\text{OH}$) species at the expense of carbonyl ($=\text{O}$) groups, and (ii) pronounced fragmentation of the flakes, resulting in a higher density of edges and defect sites. These changes are not independent but rather mutually reinforcing in determining catalytic performance. From a mechanistic standpoint, the increase in hydroxyl functional groups appears to play a central role in facilitating the initial adsorption step of OH^- ions, which is widely recognized as one of the critical stages in the OER pathway under alkaline conditions [4,7]. Hydroxyl groups can act as anchoring sites, promoting the formation and stabilization of $^*\text{OH}$ intermediates through hydrogen bonding and local electronic effects. This stabilization may lower the activation energy associated with the

first proton-coupled electron transfer step, thereby accelerating the overall reaction kinetics. In contrast, carbonyl groups, while capable of stabilizing oxygenated intermediates, may lead to an excessive binding strength that hinders the subsequent transformation into $^*OOH$ species, effectively limiting the turnover rate. Therefore, the laser-induced shift in functional group distribution toward hydroxyl-rich surfaces can be interpreted as an optimization of intermediate binding energies, in line with the Sabatier principle. In parallel, the increase in defect density and edge exposure significantly contributes to the catalytic enhancement. Defects in the sp^2 carbon lattice are known to introduce localized electronic states that can serve as high-energy active sites, facilitating both charge transfer and reactant adsorption [24,55,56]. Edges, in particular, provide undercoordinated carbon atoms that are more reactive than basal plane atoms, thus promoting the formation of key intermediates such as $^*O$ and $^*OOH$. The combined presence of defects and hydroxyl groups likely generates a cooperative environment in which functional groups modulate the local electronic structure of defect sites, enhancing their intrinsic activity. Based on these considerations, a plausible reaction scenario can be proposed in which hydroxyl-functionalized defect sites act as the primary catalytic centers. In this model, OH^- adsorption occurs preferentially at or near $-OH$ -enriched regions (Figure 5c), followed by sequential oxidation steps facilitated by adjacent defect sites. The spatial proximity between functional groups and structural irregularities may enable an efficient relay of charge and intermediates across the surface, thereby lowering kinetic barriers along the reaction pathway. It is important to note, however, that the proposed mechanism remains speculative in the absence of direct experimental evidence of reaction intermediates. No operando spectroscopic measurements or density functional theory (DFT) calculations were performed in this study to explicitly identify the active sites or quantify the adsorption energies of OER intermediates. Furthermore, the intrinsic heterogeneity of GO surfaces makes it challenging to isolate the contribution of individual functional groups. Nevertheless, the strong correlation observed between hydroxyl group enrichment, defect density, and improved electrochemical performance provides compelling indirect support for the proposed interpretation.

4. Conclusions

The present work demonstrates that pulsed laser irradiation represents an effective and versatile strategy to tune both the chemical composition and morphology of graphene oxide (GO), ultimately enhancing its electrocatalytic activity toward the Oxygen Evolution Reaction (OER). The observed improvement, leading to an overpotential as low as 380 mV at 10 mA/cm² using less than 10 µg/cm² of material, can be rationalized by considering the synergistic interplay between surface functional groups and defect-induced active sites. These findings highlight the critical role of functional group engineering in carbon-based electrocatalysts and suggest that the relative balance between hydroxyl and carbonyl species can be strategically tuned to optimize OER activity. More broadly, this work opens new perspectives for the design of metal-free catalysts through controlled structural and chemical modification, with laser processing emerging as a scalable and environmentally friendly approach. Future studies combining theoretical modeling and operando characterization will be essential to achieve a deeper understanding of the reaction mechanism and to further refine the design principles of graphene-based catalytic systems.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/cryst16050319/s1>: supplementary.pdf: file containing SEM images, FT-IR spectra, EIS and literature comparison.

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