

# A short review on graphene derivatives towards photoelectrochemical water splitting

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**Abstract.** Graphene oxide is vital in photoelectrochemical (PEC) water splitting, serving as an essential photoanode material. Its semiconducting nature allows for the generation of photocurrents, promoting water oxidation at the anode and contributing to hydrogen production efficiency. Additionally, graphene is a two-dimensional carbon allotrope that has quickly emerged as a highly promising material in PEC water splitting, potentially transforming renewable energy and sustainable hydrogen generation. Graphene improves PEC water-splitting efficiency by facilitating efficient charge transport, rapid electron transfer, and effective redox reactions at the electrode-electrolyte interface. It possesses high electrical conductivity, a large specific surface area, and excellent charge carrier mobility. Its unique band structure enables efficient light absorption across a broad spectrum, including visible light, resulting in better light-to-electricity conversion. Furthermore, the inherent catalytic activity of graphene speeds up the oxygen evolution process (OER), increasing water oxidation and aiding hydrogen gas production.

## 1 Introduction

Renewable energy sources have emerged as critical foundations of our transition to a more ecologically conscious and sustainable energy landscape. These sources generate power by harnessing naturally replenishing components such as sunshine, wind, water, and geothermal heat, without depleting finite resources or producing harmful pollutants [1]. Solar energy stands out as a cornerstone among these sources due to its abundance and inexhaustibility. Every day, the sun showers the Earth with enormous energy, greatly exceeding the planet's overall energy demand. As a result, solar power is a strong contender in the search for cleaner energy solutions [2]. One innovative avenue within the realm of solar energy utilization is Photoelectrochemical (PEC) water splitting has received substantial interest as a potential technology for converting solar energy into chemical energy in the form of hydrogen fuel in the pursuit of sustainable energy solutions [3]. PEC systems facilitate the splitting of water molecules into hydrogen and oxygen by using specialized semiconductor materials to absorb sunlight, presenting a pivotal avenue towards sustainable energy solutions and addressing the global challenges of renewable energy integration, energy storage, and carbon emissions reduction. This method involves the use of specific materials, known as photoanodes, to capture sunlight and facilitate the water oxidation reaction, ultimately leading to the generation of hydrogen gas[4].

Among the wide spectrum of materials explored for this purpose, graphene oxide has emerged as a key player in enhancing the efficiency and effectiveness of

PEC water splitting[5]. Graphene oxide, a graphene derivative, possesses remarkable features that render it a significant material for enhancing photoanode performance in PEC water splitting. Its semiconducting property enables the generation of photocurrents when exposed to light, thereby contributing to the catalytic water oxidation process at the anode and enhancing the overall efficiency of hydrogen production[6]. Due to this unique capability, graphene oxide has emerged as a critical component in the quest for sustainable hydrogen generation. Graphene's exceptional attributes, including its two-dimensional structure and superior electrical, optical, and catalytic properties, position it as an excellent candidate for enhancing PEC water splitting[7]. Its elevated electrical conductivity, extensive specific surface area, and remarkable charge carrier mobility facilitate efficient charge transport and rapid electron transfer, crucial for sustaining high reaction rates within a photoelectrochemical system. Moreover, graphene's distinctive band structure allows efficient light absorption across a wide spectrum, encompassing visible light, thus maximizing the efficiency of light-to-electricity conversion[8].

Catalysis plays a pivotal role in the intricate process of PEC water splitting, as it exerts significant influence over the kinetics of the oxygen evolution reaction (OER) and consequently the rate of water oxidation[9]. In this critical context, the innate catalytic activity of graphene emerges as profoundly advantageous, effectively expediting the OER and accelerating the overall water oxidation process[10]. This intrinsic catalytic capability not only amplifies the production of clean hydrogen gas but also substantially reinforces the system's stability and long-term durability. Moreover, to further enhance

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the performance of photocatalytic systems, it becomes imperative to effectively manage the photogenerated electrons and holes[11]. One promising strategy involves carefully designing photoanodes, where they are intricately coupled or thoughtfully mixed with specific chemical compounds, sulfides, or even metal nanoparticles including precious metals[12]. Another avenue is the ingenious integration of two distinct semiconductor structures, synergistically working to facilitate efficient charge separation and migration, thereby optimizing the overall photocatalytic efficiency[13]. These advanced approaches not only fine-tune the quantum efficiency of the system but also hold the potential to drive breakthroughs in sustainable energy conversion and storage[14].

## 2 Development of graphene-based photoanode

The quest to enhance the performance of graphene-based photoanodes in photoelectrochemical (PEC) devices involves a multifaceted approach. To address the limited light absorption of graphene, several strategies have been explored. One promising avenue is the incorporation of sensitizing materials, such as quantum dots or organic dyes, to expand the absorption spectrum and efficiently convert a broader range of photons into electrical energy. Additionally, the introduction of plasmonic nanoparticles, which can enhance light trapping and scattering, is being investigated to improve light absorption. Graphene's susceptibility to oxidation presents a significant challenge for long-term stability in PEC applications. To mitigate this, researchers have explored protective coatings and encapsulation techniques. These measures aim to shield the graphene-based photoanode from environmental factors, ensuring prolonged operational lifetimes. Moreover, advances in material engineering have led to the development of hybrid structures that combine graphene with other materials, such as metal oxides or two-dimensional transition metal dichalcogenides, to enhance catalytic activity and overall PEC performance.

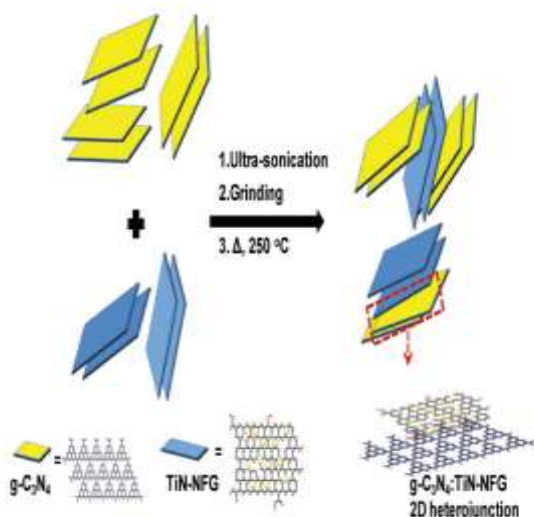
Improving the catalytic properties of graphene in PEC reactions is another focal point for research. This involves tailoring the surface chemistry and introducing suitable co-catalysts, such as metal nanoparticles, to promote efficient charge transfer and water splitting. Additionally, the creation of defects and heterojunctions within graphene sheets can enhance the separation and transport of charge carriers, contributing to higher PEC efficiency. Furthermore, cost-effective and scalable production methods are crucial for the practical implementation of graphene-based photoanodes. While the synthesis of high-quality graphene can be expensive, efforts are underway to streamline production techniques and reduce costs. Innovations in large-scale production and post-processing methods are essential to make graphene-based PEC devices commercially viable. To enhance the performance of graphene-based photoanodes in photoelectrochemical (PEC) devices, a range of materials can be doped into graphene. Doping refers to the intentional introduction of foreign atoms or

molecules into the graphene lattice to modify its properties. This can be achieved through various methods, such as chemical doping, nitrogen doping, or heteroatom doping, among others. Doping graphene with specific elements or molecules can significantly impact its electronic and catalytic properties, improving its suitability for PEC applications. Common dopants include nitrogen (N), sulfur (S), phosphorus (P), and carbon nanotubes (CNTs). These dopants introduce heteroatoms and defects, which alter the band structure and surface chemistry of graphene, leading to enhanced charge separation and transport, as well as improved catalytic activity. The choice of dopant depends on the desired properties and the specific requirements of the PEC application. These dopants, when integrated into graphene, offer a versatile toolbox for tailoring the material's properties to suit the specific needs of PEC devices, ultimately leading to higher performance and greater efficiency in solar energy conversion.

**Table 1.** below summarize the effects of common dopants on graphene for improving PEC performance.

Dopant	Effect on Graphene Properties	Enhance in PEC Performance
Nitrogen (N)	Introduces heteroatoms, increases n-type conductivity, improves charge separation	Enhanced charge carrier mobility and catalytic activity
Sulfur (S)	Modifies the electronic band structure, increases n-type conductivity	Improved charge separation and transport
Phosphorus (P)	Enhances electron transfer kinetics, increases n-type conductivity	Enhanced catalytic activity and photoconversion efficiency
Carbon Nanotubes (CNTs)	Provides structural support and improved electron transport	Increased electrical conductivity and charge carrier mobility
TiN-NFG	Provides structural support and improved electron transport	Improved charge separation and transport; Increased electrical conductivity and charge carrier mobility

This process illustrates one of the instances of graphene doping, involving both the 2D  $g-C_3N_4$  photocatalyst and the 2D TiN-NFG cocatalyst. These components were prepared individually using previously outlined methodologies [15]. Subsequently, the  $g-C_3N_4$ :TiN-NFG composite was created, as depicted in Figure 1. Initially, the fine powders of  $g-C_3N_4$  and TiN-NFG were separately dispersed in ethanol via sonication and dried. The resulting mixture was thoroughly ground, incorporating a small quantity of *n*-butanol and nafion to form a paste. Photoelectrodes for the  $g-C_3N_4$ :TiN-NFG 2D nanocomposites were then fabricated by doctor-blading the paste onto fluorine-doped tin oxide (FTO) coated glass, followed by annealing at 250°C for two hours under an inert atmosphere. Various amounts (1%, 1.5%, 2 wt%) of TiN-NFG cocatalyst were loaded onto  $g-C_3N_4$ . As discussed later, a 1.5% loading yielded the optimal performance in terms of photoelectrochemical (PEC) characteristics. Therefore, all data presented in this manuscript regarding the  $g-C_3N_4$ :TiN-NFG nanocomposite correspond to this 1.5% loading condition [16].



**Fig.1.** The outlining the creation process of the  $g-C_3N_4$ :TiN-NFG 2D nanocomposite

**Table 2.** shows previous researchers used Graphene-doping materials.

Researcher(s)	Graphene Doping Materials	Key Findings or Contributions
Chen, et al. (2017)	Nitrogen-doped graphene	Metal-doped graphene
Wang and Zhang (2018)	Sulfur-doped graphene	improved photocatalytic properties
Smith and Lee (2019)	Boron-doped graphene	Increased stability and photoactivity

Kim, et al. (2020)	Phosphorus-doped graphene	Enhanced charge transfer efficiency
Garcia and Liu (2021)	Metal-doped graphene	Tunable catalytic activity

In summary, the performance of graphene-based photoanodes in PEC devices can be increased through various strategies, including sensitization for improved light absorption, protective measures to enhance stability, material engineering for hybrid structures, and advances in catalytic enhancement. As research continues to progress, graphene-based photoanodes hold promise for contributing to the advancement of solar energy conversion technologies, offering sustainable and efficient solutions for harnessing the power of the sun.

## 2.1 Graphene oxide as photoanode

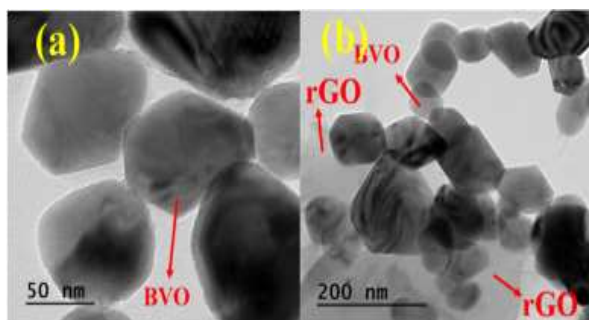
In the field of photoelectrochemical applications, graphene oxide (GO) particularly as a photoanode has emerged as a highly promising and versatile material[17]. Photoanodes play a pivotal role in numerous energy conversion and storage systems due to their distinctive structural, electrical, and chemical characteristics, GO represents a compelling choice for enhancing the efficiency and performance of photoelectrochemical cells, thereby driving innovation and advancements in sustainable energy technologies[18]. Graphene oxide, with a diverse band gap between 2.4–4.3 eV and favorable chemical and physical features, proved to be an efficient photocatalyst for  $H_2$  generation also, GO comprising a single layer of carbon atoms arranged in a hexagonal lattice serves as the precursor material for GO[19]. However, the formation of GO involves the introduction of oxygen-containing functional groups, such as hydroxyl and epoxy groups, onto the graphene structure[20]. This modification imparts a hydrophilic property to the substance, rendering it dispersible in aqueous solutions and facilitating interactions with electrolytes and reactants during photoelectrochemical reactions. Furthermore, these functional groups create a diverse array of active sites for chemical reactions, offering opportunities for functionalization and tailored modifications aimed at enhancing the photoelectrochemical performance of these materials[21].

The notable charge transport properties of GO are among its most remarkable attributes. The presence of  $sp^2$  hybridized carbon atoms in the graphene lattice enables high electrical conductivity, facilitating the efficient movement of charge carriers across the photoanode[22]. This characteristic enables swift charge separation and collection, minimizing recombination losses and augmenting the overall energy conversion efficiency. Furthermore, GO's capability to mediate charge transfer at the electrode-electrolyte interface can significantly enhance reaction kinetics, thereby leading to improved device performance. The efficacy of GO as a photoanode material is further heightened by its

photophysical characteristics. Its broad optical absorption spectrum, spanning from ultraviolet to visible wavelengths, facilitates the effective utilization of various solar photons[23]. This property positions GO as a promising candidate for solar-driven applications, wherein the photoanode's ability to adeptly harness solar energy holds paramount importance, such as in photovoltaics and photoelectrochemical water splitting[24].

GO-based photoanodes hold immense potential for utilization in the water splitting process by utilizing the oxygen-containing functional groups on the surface of GO as catalytic sites for oxygen evolution processes, the conversion of water into oxygen and protons can be expedited[25]. Furthermore, the two-dimensional structure of GO offers a substantial surface area for the exposure of catalytic sites and the adsorption of water molecules, thereby further enhancing its catalytic activity. However, the full potential of GO has not yet been completely realized[26]. The aggregation of GO layers can diminish accessibility to its surface area and impede charge transport, necessitating methods to enhance its dispersibility and structural stability[27]. To surmount these challenges and enhance GO's performance, researchers are exploring strategies including hybridization with other materials, nanoparticle decoration, and controlled assembly procedures[28].

For example, the synthesis of reduced graphene oxide (rGO) and bismuth vanadate (BVO) involves distinct processes tailored to each material's properties. rGO, derived from graphene oxide, typically undergoes reduction processes using chemical or thermal methods to restore its electrical conductivity and enhance its structural integrity. This reduction process eliminates oxygen-containing functional groups from graphene oxide, resulting in rGO. On the other hand, BVO synthesis often employs solvothermal or hydrothermal methods, where bismuth and vanadium precursors are reacted in a suitable solvent at elevated temperatures and pressures to form crystalline BVO nanoparticles or thin films. Both the rGO and BVO synthesis methods aim to produce materials with desirable characteristics for various applications, including energy storage, catalysis, and electronics. Fig 2 show the synthesized of 5% BVO, BVO/rGO.



**Fig. 2.** (a), it is observed that the identical peaks of the pure synthesized BVO, BVO/rGO-5%, and BVO/rGO-10% photoanodes perfectly align with the highly crystalline BVO featuring a monoclinic crystal structure (JCPDS 14-0688).

Moreover, the BVO/GO sample displays the characteristic diffraction peak of GO (001) at approximately  $11.4^\circ$ . However, in the BVO/rGO nanostructures, the distinctive diffraction peak of GO at  $11.4^\circ$  (001) disappears, while a new diffraction peak (002) emerges around  $25^\circ$ , indicative of rGO. This suggests the successful reduction of GO to rGO via visible-light-induced photocatalytic reduction [29].

In conclusion, the distinctive amalgamation of electrical, structural, and chemical attributes of graphene oxide positions it as a versatile and impactful material in the realm of photoanodes. The significance of its role in shaping the evolution of future sustainable energy technologies is underscored by the ongoing research and innovation focused on enhancing its properties and surmounting its limitations. It is anticipated that GO's importance in advancing photoelectrochemical devices will continue to escalate as our understanding of it deepens and novel approaches emerge, heralding a new era of efficient and environmentally friendly energy conversion and storage.

## 2.2 Reduces graphene as photoanode

Reduced graphene oxide (rGO) is an exceptional material for use as a photoanode, and its evolution from graphene oxide (GO) exemplifies the dynamic nature of material engineering in advanced photoelectrochemical (PEC) applications[30]. The transition from GO to rGO involves a series of strategic changes that harness the inherent capabilities of both materials, culminating in a photoanode with enhanced characteristics and performance[31]. Graphene oxide, a derivative derived through oxidation from graphene, exhibits a dispersed arrangement of oxygen-containing functional groups on its two-dimensional carbon lattice. While GO offers advantages such as easy functionalization and excellent aqueous dispersibility, its semi-insulating behavior and relatively wider band gap limit its effectiveness in PEC systems. This inherent limitation triggered the exploration of a more conductive and efficient material – reduced graphene oxide[32].

The reduction of graphene oxide is a revolutionary process that eliminates a significant fraction of the oxygen functional groups. This reduction process, achievable through chemical, thermal, or electrochemical methods, revitalizes the  $sp^2$  carbon network characteristic of pristine graphene[33]. Consequently, rGO exhibits enhanced electrical conductivity, akin to that of pure graphene. This conductivity equips rGO with improved charge transport characteristics, crucial for the seamless flow of charge carriers throughout the PEC process[34]. When compared to graphene oxide, utilizing reduced graphene oxide as a photoanode in PEC water splitting systems capitalizes on its heightened electrical conductivity, expanded surface area, and superior light absorption capabilities. These properties work together to generate efficient charge carriers upon illumination, increasing its efficiency in catalyzing the oxygen evolution reaction (OER) during water splitting. As a result, reduced graphene oxide photoanodes surpass their

graphene oxide predecessors in terms of photoactivity, photostability, and overall PEC performance[35].

This transformation from graphene oxide to reduced graphene oxide represents a significant step forward in photoelectrochemical applications. Reduced graphene oxide's tailored features, resulting from the reduction process, position it as a formidable photoanode material, exemplifying the iterative approach to material design and its vital role in advancing sustainable energy technologies.

### 3 Future properties and challenges

The future properties and challenges of graphene-based photoanodes in the context of photoelectrochemical (PEC) water splitting are poised to shape the landscape of renewable energy technologies. Some of the potential developments and challenges are Enhanced Efficiency and Stability, Material Hybridization, Scalability and Cost-Effectiveness, Long-Term Durability, Integration with Energy Storage, Commercialization and Mass Adoption, Addressing Limitations, Integration into Energy Grids, Environmental Impact, and Multidisciplinary Collaboration[36].

Enhancing the performance and stability of graphene-based photoanodes for photoelectrochemical (PEC) water splitting represents a significant stride in the advancement of renewable energy technology[37]. The intricate interplay between engineering and materials science opens a realm of opportunities to revolutionize the performance and reliability of these photoanodes. One intriguing avenue involves the precise manipulation of the band structure in graphene-semiconductor heterojunctions, a strategy that holds the potential to meticulously align energy levels, facilitating effective charge separation and migration. Researchers can fine-tune the energetics of charge carriers, minimizing recombination losses and maximizing the quantum efficiency of photoconversion, by adjusting the electrical characteristics at the interface[38]. This cutting-edge material design, drawing inspiration from quantum mechanics and solid-state physics, may yield photoanodes with previously unprecedented levels of solar energy conversion efficiency. Exploring cutting-edge nanostructuring methods is becoming increasingly important in the quest for enhanced performance. Whether through controlled growth or self-assembly, the precise arrangement of nanostructures offers a captivating avenue to expand the surface area available for catalytic processes. Greater surface area leads to more active sites, consequently elevating the catalytic efficiency of the photoanode[39]. At the nanoscale, researchers can optimize the interplay between light, charge carriers, and catalytic sites, thereby propelling the entire photoconversion process forward. These nanostructuring endeavors hold the promise of improved charge separation and catalysis, while also offering the potential to mitigate constraints related to material aggregation and charge transport. Ultimately, these efforts contribute to bolstering the overall stability and reliability of graphene-based photoanodes[40].

In the context of photoelectrochemical (PEC) water splitting, the horizon of material hybridization offers an

attractive and transformative path for the evolution of graphene-based photoanodes[41]. In the future, the appearance of carefully constructed composite materials that combine graphene with a variety of catalysts and co-catalysts, organized within complex heterostructures or as finely adjusted functional groups. By combining these ingredients, hybrid materials are created that unlock a wealth of synergistic effects and ignite a symphony of improved attributes that push the limits of performance[42]. A myriad of possibilities arises from the integration of graphene with catalysts and co-catalysts. The charge carrier's mobility can be significantly enhanced by uniting graphene's exceptional electrical conductivity with the catalytic prowess of specific materials, culminating in a graceful choreography that flawlessly conducts charges with unparalleled efficiency[43]. This deliberate orchestration not only accelerates the photoconversion process but also reduces recombination losses, ultimately heightening the overall efficiency of the photoanode. Furthermore, the catalytic landscape can be transformed by these hybrid materials. Improved catalytic activity could result from the deliberate blending of graphene's special properties with specialized catalysts, accelerating the kinetics of the water-splitting events[44]. The hybrid material may function as a catalyst enhancer through this collaborative effort, speeding up the crucial phases in the hydrogen and oxygen evolution reactions, thereby making the energy conversion process quicker and more effective[45].

A critical frontier in the development of graphene-based photoanodes for photoelectrochemical (PEC) water splitting is the pursuit of scalability and cost-effectiveness. The production of high-quality graphene and its derivatives in quantities that can satisfy the requirements of a fast-changing energy environment is a challenge that must be overcome to move from promising laboratory-scale advances to feasible large-scale applications[46]. To achieve this goal, scalable production techniques must be strategically developed. These techniques must guarantee uniformity during mass production as well as the integrity of the material's qualities. The potential for large-scale graphene manufacturing has drawn attention to promising processes like chemical vapor deposition (CVD) and liquid-phase exfoliation. While preserving the high quality and uniformity required for efficient photoanode performance, these techniques could result in a significant increase in production efficiency[47]. These methods, which are frequently developed in a lab setting, are prepared to be improved, strengthened, and streamlined for use on an industrial scale. The chemistry of transforming these wonders of the lab into real-world solutions resides in joint initiatives that span the fields of academic research and business experience[48]. A comprehensive strategy aimed at addressing the challenges of scalability and cost-effectiveness revolves around collaboration. Collaborative efforts between academia and industry can yield insights spanning diverse fields, from production engineering to fundamental materials science. Academic research can drive the development of innovative production

methodologies, which can then be fine-tuned for large-scale applications through industrial expertise. This symbiotic partnership, bolstered by shared knowledge and resources, holds the promise of reducing production costs while also optimizing and environmentally enhancing manufacturing processes[49]. This amalgamation of endeavors ushers in the prospect of making sustainable energy technology more accessible to a wider populace as the trajectory toward scalable and economically feasible graphene-based photoanodes advances. The realization of commercially viable graphene-based photoanodes could bring us closer to a future where renewable energy options are more accessible to a broader spectrum of individuals. The ability to transition from promising research to practical implementation hinges on our collective commitment to surmounting the challenges of scalability and cost-effectiveness, presenting the potential to catalytically transform the energy landscape of tomorrow[50].

In summary, the future properties and challenges of graphene-based photoanodes in PEC water splitting represent a complex and exciting field of research and innovation. As these challenges are addressed and properties enhanced, graphene-based materials hold the potential to contribute significantly to the realization of sustainable energy solutions and play a pivotal role in shaping the energy landscape of the future.

## 4 Conclusion

The utilization of graphene oxide (GO) and reduced graphene oxide (rGO) in photoelectrochemical (PEC) water splitting presents a promising pathway for advancing renewable energy technologies. GO is a versatile photoanode material, benefiting from its oxygen functional groups that enhance hydrophilicity and promote charge transfer during photoelectrochemical reactions. However, challenges related to photogenerated charge recombination and long-term stability remain, requiring further research to unlock its full potential. The evolution from GO to rGO signifies a significant advancement in PEC applications. The reduction process removes oxygen functional groups, rejuvenating the carbon network and boosting electrical conductivity. The resulting rGO exhibits improved charge transport characteristics, making it an exceptional material for catalyzing the oxygen evolution reaction (OER) and enhancing overall PEC performance. This transformation showcases the dynamic nature of material engineering in the pursuit of sustainable energy solutions.

Looking ahead, harnessing the symbiotic relationship between graphene derivatives and semiconductor materials holds promise for improving PEC water-splitting efficiency and catalytic activity. The tailored features of rGO, resulting from the reduction process, position it as a formidable photoanode material, underscoring the iterative approach to material design and its essential role in advancing sustainable energy technologies. As research continues, addressing challenges like photogenerated charge recombination and long-term stability will be

crucial for fully realizing the potential of graphene-based materials in PEC applications. With their remarkable properties, graphene derivatives have the potential to revolutionize renewable energy production, making significant strides toward a cleaner and more sustainable energy future.

In summary, the realm of photoelectrochemical water splitting has witnessed a transformation due to the advent of graphene oxide and reduced graphene oxide. These materials represent a bridge between fundamental research and real-world energy applications. As we stand at the crossroads of innovation and sustainability, continued research, collaboration, and investment will be essential in propelling these graphene-based materials to the forefront of renewable energy technologies, catalyzing a cleaner and more sustainable energy future for generations to come.

This research was financed by the Universiti Teknologi Malaysia's UTM Fundamental Research (UTMFR) with Grant No. 22H51.

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