

Review

Unlocking the Potential of Biomass Residues for the Sustainable Synthesis of Photocatalysts for H₂ Generation

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Abstract

Biomass-derived materials are attracting increasing interest for photocatalytic photoreforming due to their ability to function as photocatalysts, co-catalysts, supports, templating agents, and green reducing agents. This review examines recent advances in the development of biomass-derived photocatalysts, including biochar, hydrochar, carbon quantum dots, graphene-like carbons, heteroatom-doped porous carbons, and biomass-derived graphitic carbon nitride, highlighting the relationships between synthesis strategies, structural properties, and photocatalytic performance. The multifunctional role of biomass-derived materials in enhancing semiconductor dispersion, interfacial charge transfer, and charge-carrier separation is also discussed. Finally, current challenges are identified, including the limited number of intrinsically active biomass-derived photocatalysts, the incomplete understanding of structure-activity relationships, and the lack of standardized evaluation protocols, providing perspectives for the rational design of efficient and scalable photocatalysts for solar-driven photoreforming.

Keywords: waste valorisation; biomass; nanomaterials; nanostructured catalysts; heterogeneous photocatalysts; photoreforming; H₂; circular economy; photocatalysis; waste-to-value

1. Introduction

Hydrogen is a promising clean energy carrier, but its widespread use remains constrained by challenges in production, storage, transportation, and utilization. High-pressure storage, the most common approach, is energy-intensive, costly, and raises safety concerns [1]. The development of efficient and sustainable technologies for green hydrogen production has emerged as a major research priority. Among the emerging approaches, photoreforming has attracted considerable attention because it enables solar-driven H₂ generation through the photocatalytic oxidation of biomass-derived substrates or organic waste. This process requires semiconductor photocatalysts that efficiently convert solar energy into chemical energy by generating, separating, and transferring photogenerated electron-hole pairs to drive surface redox reactions. Therefore, the design and development of advanced photocatalytic materials are essential to improving H₂ production efficiency and supporting the global transition toward a hydrogen-based energy economy [1–6]. Photocatalysis, which enables photon-to-chemical energy conversion under mild reaction conditions, offers an innovative pathway for clean energy production. Since the pioneering work on TiO₂ photoelectrodes in 1972 [7], semiconductor photocatalysts such as metal oxides [8–10] and sulfides [11,12] have been developed.



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Among these, TiO_2 has emerged as a benchmark material in photocatalysis due to its exceptional environmental compatibility, chemical stability, and cost-effectiveness [13–15]. However, its practical application is severely constrained by inherent properties: a wide bandgap (~ 3.2 eV) restricting visible-light utilization, and the rapid recombination of photogenerated electron–hole pairs [13]. More recently, increasing attention has been devoted to metal-free semiconductor photocatalysts, among which graphitic carbon nitride ($\text{g-C}_3\text{N}_4$) has emerged as one of the most versatile and sustainable materials. Its suitable band structure, visible-light responsiveness, chemical stability, and ease of structural and electronic modification have made $\text{g-C}_3\text{N}_4$ a benchmark platform for the design of advanced heterojunctions and high-performance photocatalytic systems for environmental remediation and solar fuel production. The success of this metal-free semiconductor has also stimulated growing interest in the development of carbon-based photocatalysts and other carbon-containing materials as sustainable alternatives to conventional metal-containing photocatalysts.

Despite significant advances in biomass valorisation, the conversion of waste-derived resources into functional photocatalytic materials for heterogeneous light-driven processes is relatively unexplored.

This review focuses on the preparation of semiconductor photocatalysts from waste precursors and their application in photoreforming processes for the simultaneous production of hydrogen and value-added chemicals. Photoreforming involves the light-driven oxidation of an organic substrate, which acts as a hole scavenger, enabling the conversion of organic waste molecules into useful chemical products while promoting hydrogen generation. This emerging approach integrates waste management, sustainable materials development, and renewable energy production, establishing a “two birds with one stone” strategy in which waste streams are transformed into valuable resources for photocatalyst synthesis, hydrogen generation, and added-value chemicals production. Recent studies demonstrating the use of diverse residues as precursors for active photocatalysts highlight the potential of this pathway as a greener and more sustainable alternative for environmental remediation and renewable energy generation.

The aim of this review is to overview the literature on the utilization of biomass-derived wastes as precursors for the preparation of advanced materials employed as photocatalysts in photoreforming processes. This contribution examines waste-derived materials that have been successfully integrated into photocatalyst synthesis, focusing on their role in the development of efficient and sustainable catalytic systems. The transformation of waste into functional photocatalytic materials is inherently eco-friendly, and the use of green methodologies and alternative sustainable technologies will also be highlighted in the following sections.

2. Photoreforming: Reaction of Organics to Obtain Hydrogen

Photoreforming is a photocatalytic redox process in which organic substrates are oxidized while protons are reduced to H_2 under anaerobic conditions and using light-activated semiconductors. As stated in the document, “photoreforming is a photocatalytic reduction-oxidation reaction in which organic substrates are oxidized while protons are simultaneously reduced to generate hydrogen in the absence of oxygen”. Photogenerated holes oxidize organics into smaller intermediates, whereas electrons drive H_2 evolution, enabling simultaneous waste valorization and hydrogen production. This non-selective process, which typically uses alcohols as sacrificial electron donors, proceeds through complex radical pathways and highlights the coupling between organic oxidation kinetics, charge-carrier dynamics, and surface catalytic sites [16].

The photoreforming mechanism of the reaction is compared with photocatalytic water splitting and organic photooxidation in Figure 1b,c. Photoreforming occurs when the semiconductor photocatalyst is irradiated with light of a suitable wavelength to produce active electrons from the valence band to the conduction band, leaving corresponding holes at the valence band. The migration of these photogenerated electrons to the conduction band of the photocatalyst promotes proton reduction, leading to H_2 evolution, while the photogenerated holes in the valence band of the semiconductor induce the oxidation of adsorbed H_2O or electron donors. An inert atmosphere and sacrificial agents are the two necessary conditions for non-selective photoreforming to produce H_2 , as mentioned in Figure 1a. Unlike overall water splitting, photoreforming does not require highly positive valence band potentials because the oxidation of organic substrates is more thermodynamically favourable than water oxidation. Efficient photoreforming must be carried out under anaerobic conditions to prevent oxygen from competing with proton reduction through superoxide formation.

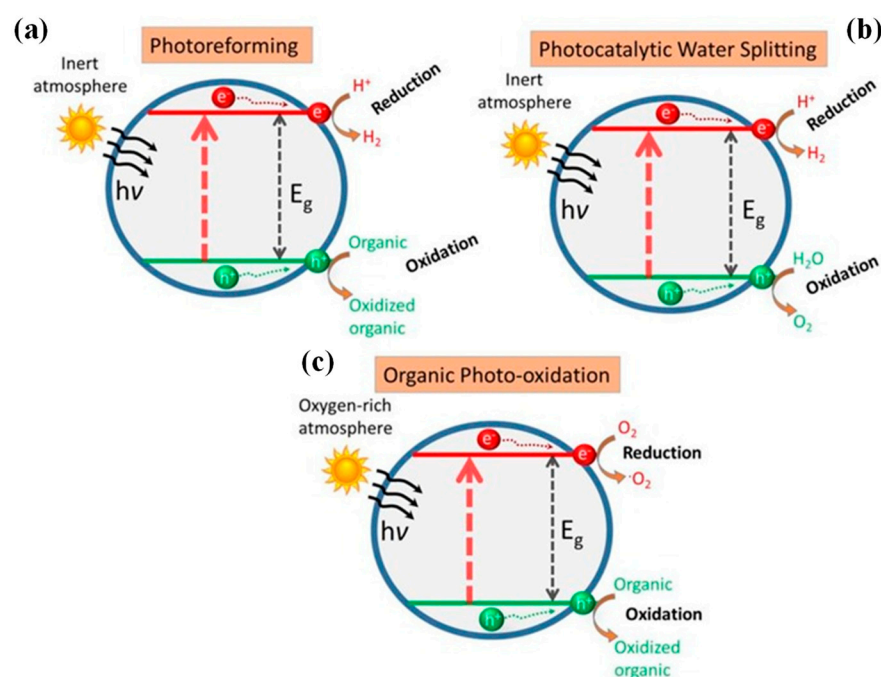


Figure 1. Comparison of photoreforming (a), i.e., the integration of photocatalytic hydrogen generation and organic photooxidation, with photocatalytic water splitting (b) and organic photooxidation processes (c) [17].

Mechanistically, photoreforming resembles water splitting but requires less positive valence band potentials, since organic oxidation is more thermodynamically favorable. Under irradiation, electrons migrate to the conduction band to reduce protons, while holes oxidize water or electron donors. Anaerobic conditions and sacrificial agents are essential to avoid oxygen competing for electrons via superoxide formation. In O_2 -rich media, $\bullet O_2^-$, H_2O_2 , and related species compete with proton reduction and may oxidize sacrificial agents [18–20]. In O_2 -free systems, oxidation proceeds through direct hole transfer or $\bullet OH$ formation. Sacrificial agents suppress recombination and enhance H_2 evolution, but their cost (alcohols, organic acids, Na_2S/Na_2SO_3 , triethanolamine) makes this approach economically unfavorable compared with steam reforming [21].

Lignocellulosic biomass (LCB) is an attractive feedstock due to its hydroxyl-rich, hydrogen-rich, and unsaturated structure, enabling efficient hole scavenging and H_2 generation. Model substrates (glucose, xylose) show higher efficiencies, but research focuses on cellulose, lignin, and raw biomass [22,23]. Most studies still rely on simple alcohols [24],

while direct biomass photoreforming remains challenging due to heterogeneity, competing pathways, $\bullet\text{OH}$ -driven overoxidation to CO_2/CO , and surface fouling [25].

This approach remains a major challenge for future research owing to the complexity and heterogeneous nature of biomass feedstocks. Unlike simple model substrates, biomass-derived feedstocks, especially lignocellulosic materials, are highly heterogeneous and undergo multiple competing reaction pathways. Moreover, the uncontrolled generation of hydroxyl radicals ($\bullet\text{OH}$) promotes overoxidation to CO_2 and CO , while non-ideal adsorption/desorption of reactants and products, together with product accumulation on the photocatalyst surface, further limits the selectivity toward value-added chemicals.

Parallel to its use as a substrate, biomass is increasingly exploited as a precursor for sustainable photocatalyst synthesis. Biomass residues offer renewable, low-cost, and environmentally benign alternatives to chemical precursors, aligning with circular-economy principles. They serve as carbon sources, sacrificial templates, and reducing agents. Conversion into biochar, activated carbon, carbon quantum dots, or porous carbons yields materials with high surface area, hierarchical porosity, abundant functional groups, and enhanced conductivity [26,27], improving light harvesting, charge separation, and adsorption, thus boosting photocatalytic performance [28]. Biomass-derived carbons have been successfully integrated with TiO_2 , ZnO , CdS , $\text{g-C}_3\text{N}_4$, BiVO_4 , and MOFs, enhancing H_2 evolution, pollutant degradation, CO_2 reduction, and biomass photoreforming.

Future directions emphasize fully circular systems where biomass acts both as photocatalyst precursor and photoreforming substrate, reducing costs, minimizing fossil-derived inputs, and improving sustainability. Advances in catalyst design, operando studies, modelling, and AI will accelerate the development of next-generation biomass-derived photocatalysts.

3. Biomass Residues as Renewable Precursors for Sustainable Photocatalyst Synthesis

Biomass wastes encompass a wide range of organic materials derived from various sources, including agriculture, forestry, food industry, and municipalities. Understanding the sources and types of biomass wastes is crucial for identifying opportunities for their utilization and addressing environmental challenges [29]. Considering the vast number of biomass resources reported in the literature, this review is not intended to comprehensively cover all available feedstocks. Rather, it provides an overview of the principal biomass waste sources, focusing on their valorization as sustainable precursors for the synthesis of photocatalytic materials. Figure 2 illustrates the main biomass waste sources and the corresponding categories of biomass residues.

Agriculture and forestry constitute the principal sources of biomass waste, providing abundant residues such as straw, husks, stalks, leaves, animal manure, crop-processing residues, agro-industrial byproducts like bagasse [30–34], and forestry-derived materials including forest residues, logging waste, sawdust, wood chips, and bark [35,36]. Additional biomass waste is generated by various agro-industrial activities, including food and agricultural processing, beverage production, pulp and paper manufacturing, textiles, pharmaceuticals, and chemical sectors whose residues (fruit and vegetable peels, food waste, spent grains, wastewater solids) can be valorized via anaerobic digestion, fermentation, or composting to produce biogas, biofuels, animal feed, and soil amendments [37]. The pulp and paper industry generates black liquor, a lignin- and cellulose-rich byproduct that can be upgraded through gasification or pyrolysis to syngas and bio-oil, improving energy efficiency and reducing emissions [38,39]. Other industrial residues, including cotton waste, fabric scraps, dye wastewater, and pharmaceutical/chemical wastes, also represent viable feedstocks for bio-based materials and bioenergy production, supporting waste reduction and circular-economy strategies. These biomass streams are particularly relevant because of

their structural and chemical richness, which is largely associated with the cellulose, hemicellulose, and lignin components of lignocellulosic biomass, which provide carbon frameworks and functional groups essential for engineering biomass-derived photocatalysts.

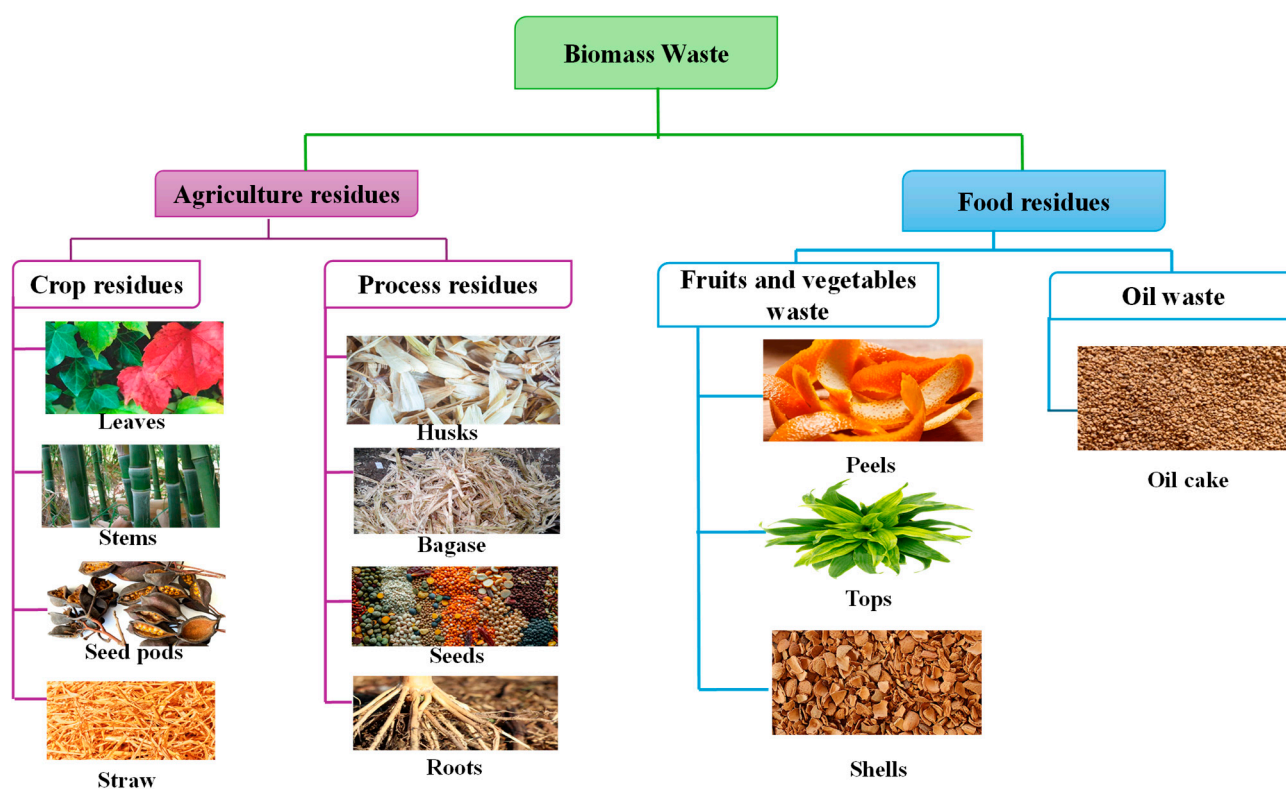


Figure 2. Representative examples of biomass waste resources with potential for photocatalyst synthesis. The categories shown are illustrative rather than comprehensive.

Cellulose, with its highly ordered crystalline domains and abundant hydroxyl groups, offers an ideal precursor for producing carbonaceous supports, porous carbons, and oxygen-rich surface functionalities that enhance charge separation and pollutant adsorption. Hemicellulose, being more amorphous and chemically diverse, decomposes into reactive intermediates that introduce heteroatoms (e.g., O, N) into carbon matrices, improving conductivity and active-site density. Lignin, the aromatic and most recalcitrant fraction, serves as a unique source of graphitic carbon and phenolic structures, enabling the formation of doped carbon frameworks, carbon quantum dots, and hybrid photocatalysts with strong visible-light absorption. Together, these biopolymers provide a renewable, structurally rich platform (see Figure 3) from which biomass-based photocatalysts can be synthesized, allowing the development of sustainable materials with tunable morphology, surface chemistry, and optoelectronic properties. Although lignocellulosic biomass (LCB) exploitation releases CO₂, carbon is continuously recycled through photosynthesis, making LCB a renewable feedstock for fuels and value-added chemicals. Despite its abundance (~1.8 trillion tonnes on land and 4 billion tonnes in marine environments), its utilization remains limited [40]. Cellulose, the main LCB component, is a C₆ polysaccharide composed of $\beta(1\rightarrow4)$ -linked D-glucose units forming highly ordered crystalline fibrils through extensive hydrogen bonding. Its high crystallinity and hydrolysis resistance are balanced by the presence of amorphous regions [41–43]. The increasing interest in LCB valorization is driven by its large annual availability (~13 billion metric tonnes), renewability, carbon neutrality, and versatility. Waste-derived and biomass-inspired materials offer low-cost, eco-friendly alternatives, and several waste-derived/templated photocatalysts have been developed for

photocatalytic applications [44]. The residues can be used to obtain green and sustainable carbonaceous materials such as the semiconductor graphitic carbon nitride (g-C₃N₄). Also, carbon quantum dots can be obtained and used as co-catalysts, forming a composite with a semiconductor and more traditional semiconductors, such as TiO₂ and ZnO, that have been prepared or templated using biowastes.

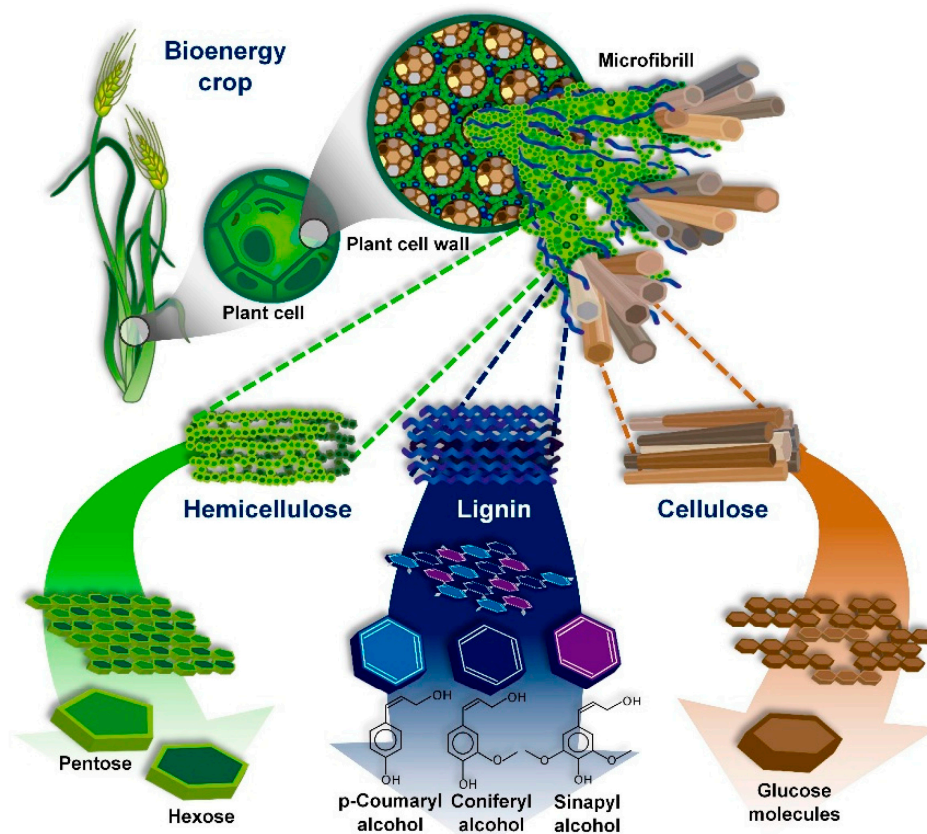


Figure 3. Main structural components of lignocellulosic biomass (LCB) and its biopolymers to be potentially valorised as renewable precursors for photocatalyst synthesis [45].

Lignocellulosic biomass (LCB) combines renewability, carbon neutrality, low ash and sulfur content, wide availability, and low cost, making it an attractive feedstock for sustainable biorefineries. Its conversion enables the production of diverse bio-based products, including nanocellulose, bio-oil, bioethanol, platform molecules, and advanced materials, without competing with food resources [46,47]. However, LCB valorization remains limited, with large amounts of agricultural and forestry residues still discarded or burned, causing environmental impacts and wasting valuable renewable resources [48,49].

LCB is renewable, carbon-neutral, low-ash, and low-sulfur, supporting biorefineries producing nanocellulose, bio-oil, bioethanol, platform molecules, and advanced materials [46,47]. However, large quantities remain underutilized or burned [48,49]. Pretreatment is essential to disrupt the cellulose–hemicellulose–lignin network, improving accessibility and enabling downstream valorization [50,51]. Hemicellulose’s amorphous heteropolysaccharide structure (xylan, galactomannan, glucomannan, xyloglucan) [52,53] supports applications in drug delivery, food, cosmetics, packaging, and papermaking [54,55]. Lignin, composed of phenylpropanoid units (p-coumaryl, coniferyl, sinapyl alcohols) [56,57], provides mechanical strength and is largely burned for energy [58,59], with only ~2% valorized despite its potential as a renewable aromatic feedstock [60,61].

4. Biomass-Derived Photocatalyst

The selection of biomass precursors is a critical factor governing the structural, physicochemical, and activity of biomass-derived photocatalysts. Variations in the relative proportions of cellulose, hemicellulose, lignin, proteins, lipids, and naturally occurring inorganic species (e.g., K, Ca, Mg, Si, Fe, and P) strongly influence carbonization pathways, crystal growth, porosity, surface chemistry, and the electronic structure of the resulting photocatalysts, ultimately affecting their performance for photocatalytic H_2 production through photoreforming [45].

Biomass offers an extraordinarily diverse and renewable pool of precursors for the synthesis of photocatalytic materials, with the range of potential feedstocks being limited primarily by availability and creativity in material design. Lignocellulosic biomass, agricultural and forestry residues, food waste, algae, and other biogenic resources can all be converted into functional photocatalysts through appropriate thermal, hydrothermal, or chemical treatments. This remarkable versatility enables the production of materials with tailored structural, optical, and electronic properties, making biomass a sustainable platform for the development of photocatalysts for solar-driven photoreforming and hydrogen production. Representative examples of biomass feedstocks and the corresponding classes of biomass-derived photocatalysts are summarized in Figure 4.

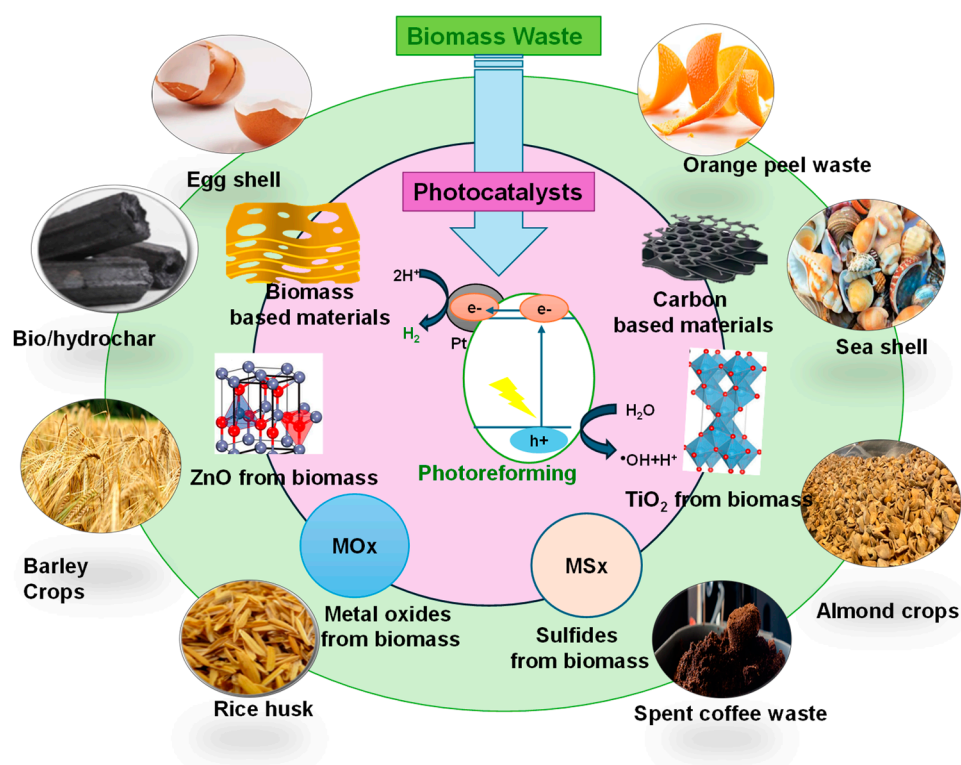


Figure 4. Representative biomass feedstocks and their conversion into biomass-derived photocatalysts for photoreforming applications.

Cellulose and hemicellulose generally decompose into oxygen-rich carbon frameworks with abundant surface functional groups that promote pollutant adsorption and facilitate semiconductor anchoring, whereas lignin yields highly graphitized carbons with superior electrical conductivity, enhanced charge transport, and reduced electron–hole recombination. Naturally occurring minerals further contribute to catalyst development by promoting pore formation, acting as structural templates, or enabling the direct synthesis of heteroatom-doped carbons, particularly from nitrogen-rich biomass such as chitosan, algae, and protein-containing residues. The incorporation of heteroatoms modifies the electronic

structure, improves conductivity, and generates defect sites that enhance charge separation and interfacial electron transfer. In addition, the intrinsic hierarchical architecture of many biomass feedstocks, including wood, bamboo, nutshells, leaves, among others, is partially preserved during carbonization, producing interconnected porous networks that improve light harvesting, mass transport, active-site accessibility, and nanoparticle dispersion [62]. Biomass-derived carbon therefore serves not only as a structural support but also as an active photocatalytic component, acting as a photosensitizer, electron reservoir, and conductive matrix that promotes efficient charge separation. Furthermore, the elemental composition of different biomass resources, particularly their carbon, hydrogen, oxygen, and nitrogen contents, determines the chemical functionality of the resulting carbon materials, while careful control of thermochemical conversion conditions and post-treatment strategies enables the tailoring of their catalytic properties [63,64]. Owing to their tunable composition, hierarchical micro-, meso-, and macroporous structures, and the possibility of incorporating functional heteroatoms, biomass-derived carbon materials have emerged as versatile and sustainable photocatalysts with significant potential for solar-driven hydrogen production and other energy-related applications [65].

4.1. Carbon-Based, Biomass-Derived Metal-Free Photocatalysts: Classification, Features, and Roles as Co-Catalysts and Supports

Biomass-derived metal-free materials to be used in photoreforming can be broadly classified into six main categories, as depicted in Figure 5, according to their structural characteristics and synthesis routes: biochar-based photocatalysts, hydrothermal carbons (hydrochars), carbon quantum dots (CQDs), graphene-like biomass-derived carbons, heteroatom-doped porous carbons, and biomass-derived graphitic carbon nitride (g-C₃N₄). Although all these materials originate from renewable biomass feedstocks, they differ significantly in their degree of carbonization, crystallinity, surface chemistry, electronic structure, and photocatalytic performance. Biochars and hydrochars represent the simplest carbonaceous materials and are primarily valued for their adsorption capacity and defect-mediated photocatalytic activity [66]; conversely, carbon quantum dots [67], graphene-like carbons, and heteroatom-doped carbons [68] possess more engineered electronic structures that promote efficient light absorption and charge separation. Biomass-derived graphitic carbon nitride, g-C₃N₄, constitutes a distinct class of polymeric semiconductor that combines the sustainability of biomass precursors with the intrinsic visible-light activity [17]. All of these materials represent a family of metal-free photocatalysts that can be tailored for applications ranging from environmental remediation to solar fuel production.

Biochar-based photocatalysts are produced through the pyrolysis of biomass under oxygen-limited conditions, yielding porous carbonaceous materials with varying degrees of graphitization and abundant surface functional groups. Their photocatalytic activity arises mainly from structural defects, oxygen-containing functional groups, and persistent free radicals that facilitate the generation of reactive oxygen species under light irradiation [69,70]. Pristine biochars generally exhibit limited photocatalytic efficiency because of rapid electron–hole recombination. However, their performance can be substantially improved through activation, defect engineering, and heteroatom doping. Because of their low cost, high stability, and excellent adsorption capacity, biochar-based photocatalysts are widely investigated for wastewater treatment and pollutant degradation. In contrast, hydrothermal carbons, also known as hydrochar, are synthesized via hydrothermal carbonization of biomass in subcritical water, typically at temperatures between 180 and 250 °C [71]. Compared with pyrolytic biochars, hydrochars retain a higher abundance of oxygen-containing functional groups and exhibit a lower degree of aromaticity. These characteristics enhance their affinity for polar contaminants and facilitate interfacial charge transfer during photocatalytic reactions. However, their relatively low electrical

conductivity generally hampers charge transport and limits photocatalytic performance. Consequently, post-synthetic modifications, such as thermal annealing or heteroatom doping, are often employed to improve visible-light absorption, enhance charge-carrier mobility, and ultimately increase photocatalytic efficiency.

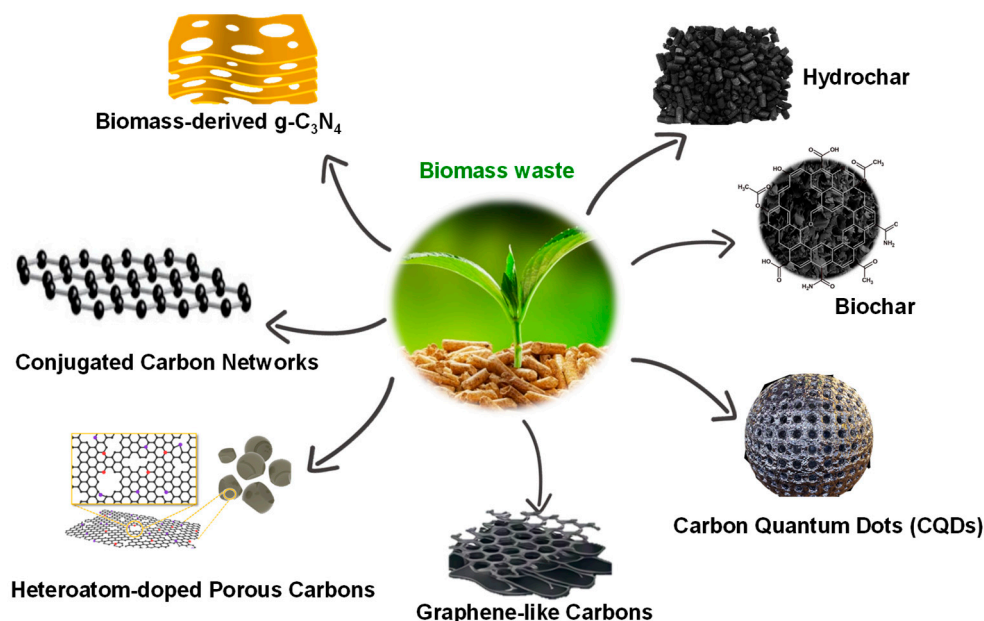


Figure 5. Representative classification of biomass-derived metal-free photocatalysts, highlighting the major material families obtained through different biomass conversion strategies and their corresponding structural characteristics.

Carbon quantum dots (CQDs) are quasi-spherical carbon nanoparticles, typically 2 to 10 nm, that can be synthesized from a wide variety of biomass resources, including fruit peels, coffee waste, lignin, cellulose, and chitosan. They are produced through controlled carbonization. Hydrothermal synthesis is one of the most widely used methods for preparing biomass-derived CQDs because of its simplicity and relatively mild, environmentally friendly conditions. In this process, biomass precursors are dispersed in water and treated in a sealed autoclave (typically at 160–250 °C), where hydrolysis, dehydration, and carbonization lead to the formation of carbon nanoparticles generally smaller than 10 nm. The resulting CQDs are purified by centrifugation, filtration, or dialysis. The resulting CQDs possess abundant oxygen-containing functional groups (e.g., hydroxyl, carbonyl, and carboxyl), which enhance their water dispersibility and provide active sites for further functionalization. Surface passivation or heteroatom doping with elements such as N, S, P, or B is commonly employed to tailor their electronic structure, improve charge separation, broaden visible-light absorption, and enhance photocatalytic activity [72]. Other synthetic approaches include pyrolysis, microwave-assisted synthesis, and solvothermal treatment. Pyrolysis yields more graphitized CQDs with higher electrical conductivity but fewer oxygen-containing functional groups, while microwave-assisted synthesis offers rapid and efficient production. Solvothermal methods enable further control over particle size and surface chemistry. Owing to quantum confinement and abundant edge defects, CQDs exhibit unique optical properties, including tunable photoluminescence, broad visible-light absorption, and excellent electron-transfer capability [73]. These unique properties enable CQDs to serve as efficient photosensitizers, electron reservoirs, and photocatalysts in a wide range of applications, including pollutant degradation, photocatalytic hydrogen evolution, and carbon dioxide reduction [74].

Graphene-like biomass-derived carbons are included among the carbon-based photocatalysts (see Figure 5). They are obtained through high-temperature carbonization that promotes the formation of extended sp^2 -hybridized carbon domains. Their partially graphitic structure provides high electrical conductivity, broad-spectrum light absorption, and rapid electron transport, all of which contribute to improved photocatalytic performance [75]. Unlike conventional biochars, these materials rely on their highly conjugated π -electron networks and defect-rich edges to facilitate charge separation and surface redox reactions. Biomass sources rich in lignin, such as wood and bamboo, are particularly suitable precursors for producing graphene-like carbons. Heteroatom-doped porous carbons represent one of the most advanced classes of biomass-derived metal-free photocatalysts [76]. The incorporation of heteroatoms such as nitrogen, sulfur, phosphorus, or boron into the carbon framework modifies the electronic structure, creates additional defect sites, and enhances visible-light absorption. These dopants facilitate charge separation, increase the density of catalytically active sites, and improve the generation of reactive oxygen species.

Nitrogen-doped porous carbons (NPCs) have attracted considerable attention as metal-free photocatalysts because nitrogen incorporation effectively tailors the electronic structure of carbon frameworks [77]. Depending on its bonding configuration (e.g., pyridinic N, pyrrolic N, graphitic N, and oxidized N), nitrogen can modify the local electron density, create defect sites, and alter the band structure, thereby facilitating visible-light absorption and promoting the separation and migration of photogenerated charge carriers. Nitrogen-doped porous carbons are especially attractive because nitrogen-containing functional groups enhance the electrical conductivity of the carbon matrix while simultaneously introducing active sites for photocatalytic reactions; for instance, pyridinic and pyrrolic nitrogen introduce catalytically active sites that participate in adsorption and interfacial redox reactions. Furthermore, the high surface area and hierarchical porosity of NPCs provide abundant accessible active sites and shorten the diffusion pathways for reactants and charge carriers, ultimately improving photocatalytic efficiency. For these reasons, heteroatom doping has emerged as one of the most effective strategies for enhancing the intrinsic photocatalytic activity of biomass-derived carbons. Besides nitrogen, other heteroatoms such as sulfur, phosphorus, and boron, or their co-doping with nitrogen, can further regulate the electronic structure, optimize charge distribution, and generate synergistic effects that enhance light harvesting, charge transfer, and surface reaction kinetics. As a result, heteroatom-doped porous carbons have demonstrated significantly improved performance in photocatalytic applications, including pollutant degradation, photocatalytic H_2 evolution, and CO_2 reduction.

Biomass-derived graphitic carbon nitride ($g-C_3N_4$) constitutes a distinct subclass of metal-free photocatalysts that combines semiconductor properties with the sustainability of renewable biomass precursors [78]. While conventional $g-C_3N_4$ is synthesized from nitrogen-rich chemicals such as melamine or urea, increasing attention has been devoted to biomass-derived nitrogen sources including chitosan, proteins, amino acids, and algae [79]. Biomass-assisted synthesis often introduces additional porosity, structural defects, and carbon-rich domains that improve visible-light absorption and reduce electron–hole recombination. As a result, biomass-derived $g-C_3N_4$ has shown excellent performance in photocatalytic water splitting, CO_2 reduction, and environmental remediation, making it one of the most promising metal-free photocatalysts for sustainable solar energy conversion [80].

Biomass-derived carbon materials can be synthesized through various routes, schematized in Figure 6, which determine their physicochemical properties. For each method, changes in the physicochemical properties of the as-synthesized carbon materials in response to different conditions are systematically investigated.

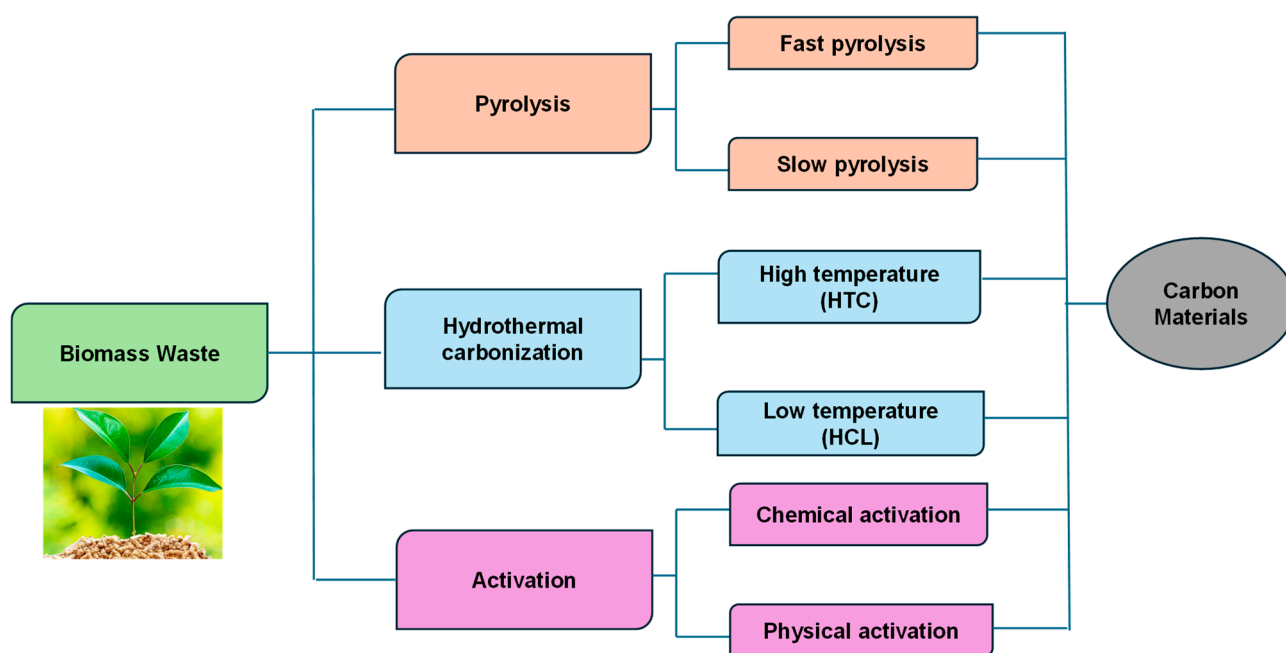


Figure 6. Schematic overview of the main synthetic routes used to produce biomass-derived carbon materials and the key processing parameters influencing their physicochemical properties.

Pyrolysis and hydrothermal carbonization (HTC) are the two principal thermochemical routes for converting biomass into carbonaceous materials with tailored properties. Pyrolysis involves heating biomass under an oxygen-free atmosphere, typically between 300 and 1200 °C, leading to the thermal decomposition of cellulose, hemicellulose, and lignin into gaseous products and carbon-rich biochar [81]. The characteristics of the resulting material are strongly influenced by the biomass composition and processing conditions, including temperature, heating rate, residence time, and particle size. In general, slow pyrolysis favours higher biochar yields, whereas fast pyrolysis is optimized for bio-oil production. Increasing the pyrolysis temperature promotes graphitization, electrical conductivity, and surface area, although it generally reduces the carbon yield. In contrast, hydrothermal carbonization (HTC) is carried out in water at relatively low temperatures (180–250 °C) under autogenous pressure, making it particularly suitable for wet biomass without the need for prior drying. During HTC, biomass undergoes hydrolysis, dehydration, decarboxylation, and aromatization reactions to produce hydrochar, which is typically richer in oxygen-containing functional groups and more hydrophilic than pyrolytic biochar. The properties of hydrochar depend mainly on the reaction temperature, residence time, and biomass-to-water ratio, while microwave-assisted HTC has emerged as an attractive alternative due to its shorter processing times and lower energy consumption [82]. To further enhance the performance of biomass-derived carbons, activation treatments are commonly employed to increase porosity, specific surface area, and the density of surface functional groups. Physical activation, usually performed with steam or CO₂, develops pore networks through controlled gasification, whereas chemical activation with agents such as KOH, NaOH, H₃PO₄ or ZnCl₂ is more effective in producing highly porous carbons with exceptionally large surface areas [83]. The activation conditions, particularly temperature, residence time, and activator-to-biomass ratio, determine the final pore structure and surface chemistry, thereby significantly improving the adsorption capacity, electrical conductivity, and photocatalytic performance of the resulting carbon materials.

Representative examples from the literature illustrate the diverse roles of carbon-based biomass-derived materials in photocatalytic photoreforming. Depending on their composition and structure, they can act as photocatalysts (e.g., biomass-derived g-C₃N₄),

co-catalysts coupled with semiconductors, or conductive supports that promote catalyst dispersion, interfacial charge transfer, and efficient charge-carrier separation.

Biomass-derived materials, including biochar, activated carbon, hydrochar, graphene-like carbons, and heteroatom-doped porous carbons, are promising supports for semiconductor photocatalysts owing to their renewable origin, low cost, high surface area, hierarchical porosity, abundant surface functionalities, and good electrical conductivity. Compared with conventional inorganic supports (e.g., SiO_2 and Al_2O_3), these carbonaceous materials offer a sustainable alternative that is fully aligned with the principles of green chemistry and the circular economy. The physicochemical properties of biomass-derived supports play a key role in determining the performance of semiconductor photocatalysts. Their large surface area and interconnected pore network promote the homogeneous dispersion of semiconductor nanoparticles, prevent particle agglomeration, and increase the number of accessible active sites. Simultaneously, oxygen-containing functional groups, including hydroxyl, carbonyl, and carboxyl moieties, provide effective anchoring sites that strengthen the semiconductor–support interface, improve catalyst stability, and facilitate the adsorption and diffusion of biomass-derived substrates and reaction intermediates.

Beyond their structural role, biomass-derived carbon supports actively participate in photocatalytic charge-transfer processes. Their graphitized domains act as conductive pathways and electron acceptors, promoting the migration of photogenerated electrons away from the semiconductor and suppressing electron–hole recombination. Furthermore, heteroatom doping, particularly with nitrogen, sulfur, phosphorus, or boron, tailors the electronic structure of the carbon framework by introducing defect sites, enhancing electrical conductivity, extending visible-light absorption, and promoting charge-carrier separation. Strong semiconductor–carbon coupling may also lead to heterojunction formation, accelerating interfacial charge transfer and improving solar-light harvesting.

These synergistic effects are particularly advantageous for photocatalytic photoreforming, where efficient substrate adsorption, rapid charge separation, and effective photon utilization are essential to maximize hydrogen production. Accordingly, biomass-derived supports have been successfully integrated with a wide range of semiconductor photocatalysts, including TiO_2 , ZnO , CdS , ZnIn_2S_4 , and graphitic carbon nitride ($\text{g-C}_3\text{N}_4$) yielding composites with enhanced photocatalytic activity, stability, and recyclability. Although still in its infancy for biomass photoreforming, biomass-derived supports show great promise for developing efficient photocatalysts for solar hydrogen production and biomass valorization.

Biochar is employed as a conductive support or co-catalyst to improve the performance of semiconductor photocatalysts. Nevertheless, several studies have demonstrated that biochar can exhibit intrinsic photocatalytic activity when its electronic structure is appropriately engineered. In particular, nitrogen-doped biochars prepared from biomass precursors such as bamboo, cellulose, chitosan, and agricultural residues display enhanced visible-light absorption owing to defect states introduced by heteroatom doping. Zerga et al. have prepared a biochar-integrated TiO_2 nanocomposite heterojunction by sol–gel. The hierarchical-functionalized pore framework of biochar had been effectively developed, and the generated biochar had a specific surface area of up to $64 \text{ m}^2 \cdot \text{g}^{-1}$. The biochar/ TiO_2 nanocomposite yielded 5.50 times more H_2 than bare TiO_2 , which was also attributed to the presence of metallic elements via the Schottky barrier (Ni, Mn, Al, Cu, Zn, and Fe) [84].

Biochar has also served as an effective support for CdS/TiO_2 heterojunctions, providing a high-surface-area, conductive scaffold that enabled uniform semiconductor dispersion and strong interfacial contact. Its porous carbon framework facilitated reactant diffusion and accelerated photogenerated electron transport, thereby suppressing charge recombination within the CdS/TiO_2 heterojunction. As a result, the biochar-supported composite

exhibited significantly enhanced glucose photoreforming activity, with improved simultaneous H_2 and organic acid production compared with the unsupported semiconductor [85].

Similarly, biochar obtained from pomelo peel through pyrolysis exhibits high adsorption capacity and a porous carbon structure, suggesting its potential use as a semiconductor support or conductive co-catalyst in photocatalytic systems [86]. Its extended π -conjugated carbon network and abundant oxygen-containing functional groups promote charge separation and the generation of reactive oxygen species, demonstrating that appropriately designed biomass-derived biochars can function as photocatalytically active materials. More broadly, heteroatom-doped biochars obtained from lignocellulosic biomass, including wood, straw, and agricultural residues, have shown measurable photocatalytic hydrogen evolution, with their performance mainly attributed to defect-rich carbon structures, improved visible-light absorption, and enhanced charge transport.

Representative examples include walnut shell-derived activated carbon coupled with TiO_2 nanotubes for methanol photoreforming [87], among others. In these systems, biochar primarily acts as a conductive support that enhances semiconductor dispersion, improves substrate adsorption, facilitates interfacial electron transfer, and suppresses charge recombination, thereby significantly improving hydrogen production. These findings suggest that the greatest potential of biomass-derived biochar in photoreforming lies in its synergistic integration with semiconductor photocatalysts rather than in its use as an independent photoactive material.

Hydrochars, obtained via hydrothermal carbonization (HTC) of lignocellulosic residues under subcritical water conditions, represent a versatile class of carbonaceous materials with tunable surface chemistry and hierarchical porosity. The HTC process typically involves treating raw biomass at moderate temperatures (180–250 °C) in aqueous media, yielding oxygen-rich hydrochars that retain abundant functional groups and aromatic domains, which are advantageous for photocatalytic applications [88]. These hydrochars can act as supports for semiconductor photocatalysts, enhancing charge separation and stability, or serve as electron mediators and sacrificial templates during photoreforming reactions [89]. Their ability to donate electrons and facilitate redox processes makes them particularly effective in photoreforming of biomass-derived substrates, where hydrogen or value-added chemicals are produced under visible-light irradiation. Recent studies have demonstrated that hydrochars integrated with TiO_2 or $g-C_3N_4$ or metal phosphides significantly improve photocatalytic efficiency by promoting interfacial charge transfer and extending light absorption [90].

Biochar and hydrochar are the primary biomass-derived carbon materials obtained by pyrolysis and hydrothermal carbonization, respectively. Biochar is generally more carbonized and electrically conductive, whereas hydrochar contains a higher concentration of oxygen-containing functional groups but typically exhibits lower porosity and conductivity. Both materials are widely employed as photocatalyst supports, carbon precursors, or, after suitable modification, as conductive co-catalysts or metal-free photocatalysts. In contrast, activated carbon (AC) is produced by the physical or chemical activation of carbonaceous precursors, such as biochar or hydrochar. This activation step generates a highly porous structure with a very high specific surface area, making AC an excellent support for semiconductor photocatalysts by enhancing nanoparticle dispersion, reactant adsorption, and interfacial charge transfer. Biomass-derived activated carbon (AC) has been widely employed as a conductive support for semiconductor photocatalysts in photoreforming, owing to its high porosity, large surface area, and ability to facilitate charge transport.

For instance, walnut shell-derived activated carbon (AC) has been employed as a sustainable carbon co-catalyst in TiO_2 -based photocatalytic composites. The AC was prepared by crushing and pre-carbonizing walnut shells at 450 °C, followed by chemical

activation with KOH and a subsequent heat treatment at 800 °C. After acid washing and purification, the resulting porous activated carbon was used as a support for microwave-hydrothermally synthesized TiO₂ nanotubes (TiO₂NT), which were deposited onto the AC surface at different loadings (Figure 7). The high surface area and electrical conductivity of the activated carbon promoted charge separation and electron transport, leading to enhanced photoreforming performance compared with pristine TiO₂ nanotubes [87].

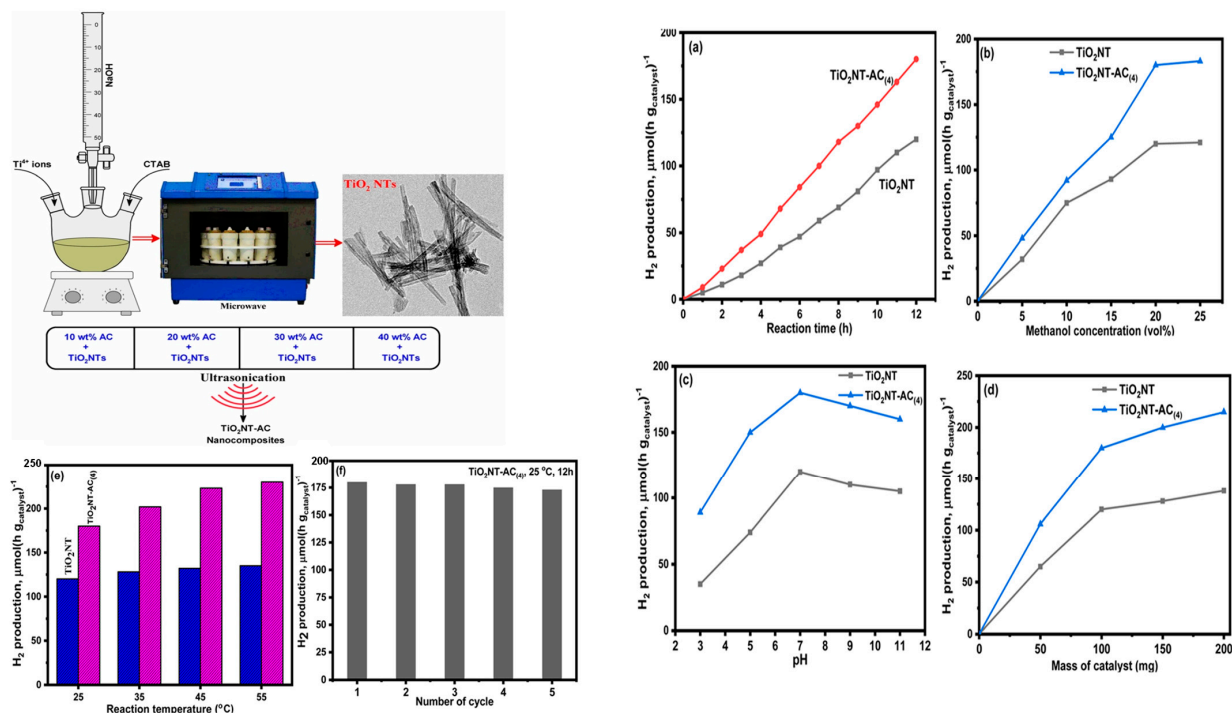


Figure 7. Preparation of walnut shell-derived activated carbon and subsequent deposition of TiO₂ nanotubes to obtain AC/TiO₂NT photocatalytic composites, Photocatalytic hydrogen production over synthesized TiO₂NTs and TiO₂NTs–AC(4) catalysts, (a) effect of reaction time, (b) effect of concentration of methanol, (c) effect of solution pH, (d) effect of mass of catalysts, (e) effect of reaction temperature, and (f) recyclability of the TiO₂NTs–AC(4) catalyst [87].

The AC/TiO₂NT nanocomposites were evaluated for photocatalytic H₂ production via methanol photoreforming under simulated solar irradiation. The composite containing 40 wt% activated carbon showed the highest activity, achieving an H₂ yield of ~18 mmol L^{−1} and excellent stability over five consecutive cycles, demonstrating the beneficial role of biomass-derived activated carbon in enhancing photocatalytic performance [87].

Biomass-derived activated carbon (AC) has also been successfully employed as a conductive support for Co₃O₄ photocatalysts in the photoreforming of H₂S-containing wastewater. The highly porous AC framework, prepared from neem leaves by carbonization, promotes the homogeneous dispersion of Co₃O₄ nanosheets, enhances reactant adsorption, and facilitates interfacial charge transfer, thereby suppressing electron–hole recombination and significantly improving H₂ production compared with bare Co₃O₄. Sulfide and sulfite species further enhance hydrogen production by acting as sacrificial electron donors, although excessive sulfide concentrations can inhibit activity through sulfur deposition on the catalyst surface. Moreover, the Co₃O₄/AC composite exhibits excellent recyclability, highlighting the potential of biomass-derived activated carbon as a sustainable support for integrating wastewater remediation with solar-driven hydrogen production [91].

Carbon quantum dots (CQDs) synthesized from biomass precursors via hydrothermal or semi-hydrothermal carbonization have attracted significant attention due to their low-cost, eco-friendly preparation and multifunctional photocatalytic roles. The process

involves treating lignocellulosic or polysaccharide-rich biomass (e.g., cellulose, sawdust, tea waste, walnut shells) under controlled aqueous conditions, producing nanoscale carbons with abundant surface functional groups and tunable band structures [88]. The preparation is relatively simple, as schematised in Figure 8. Carbon quantum dots (CQDs) were synthesized from various biomass wastes using a semi-hydrothermal process under an inert nitrogen atmosphere. Biomass precursors were mixed with distilled water, sealed in porcelain crucibles, and thermally treated at 450 °C for 1 h after a controlled heating ramp. Following cooling, the solid products were dispersed in distilled water, and the suspension was centrifuged to separate the CQDs, which were recovered from the supernatant. This simple and scalable approach enables the direct conversion of diverse biomass residues into CQDs without the use of hazardous chemicals.

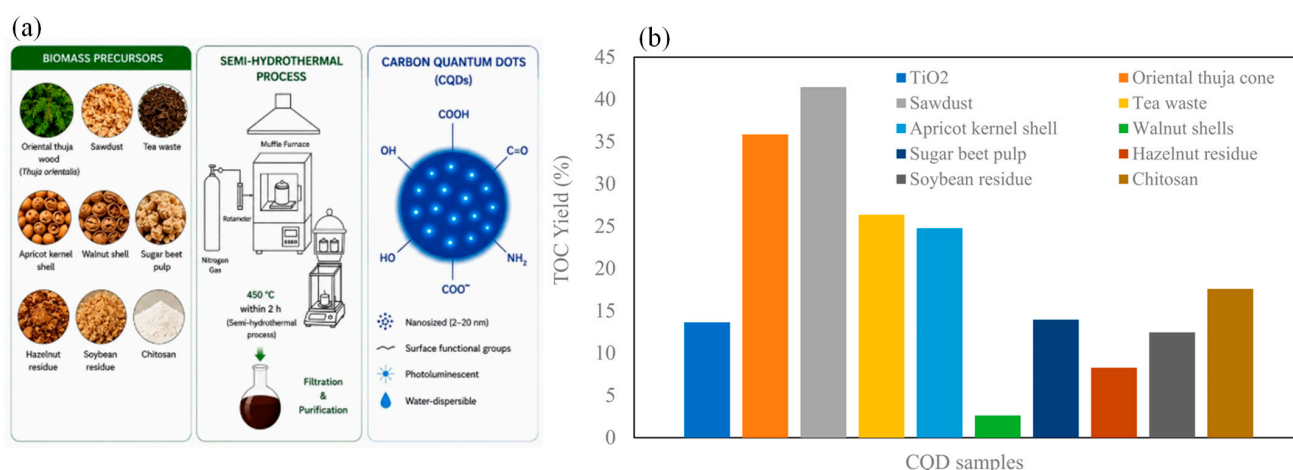


Figure 8. (a) Schematic illustration of the valorization of different biomass wastes into carbon quantum dots (CQDs) through the semi-hydrothermal synthesis process. (b) Time-dependent TOC removal efficiencies of Reactive Black 5 dye by CQDs/TiO₂ systems reported in Ref. [88].

CQDs inherit abundant oxygenated functional groups and aromatic domains that confer strong π – π^* and n – π^* transitions, photoluminescence, and excellent colloidal stability. These features are critical for photocatalytic efficiency, as they enable CQDs to act as electron reservoirs, photosensitizers, and up-conversion agents, thereby extending visible-light absorption and facilitating charge separation when coupled with semiconductors such as TiO₂ or g-C₃N₄. Collectively, biomass-derived CQDs represent a multifunctional platform for solar-driven photoreforming, coupling waste valorization with sustainable hydrogen generation and green chemical synthesis [89]. As a representative case, cellulose-derived CQDs have been synthesized via a hydrothermal route, where cellulose was dispersed in deionized water and subjected to high-temperature (200 °C) and high-pressure conditions in a Teflon-lined autoclave for 6 h. This process enabled the transformation of cellulose into nanoscale carbon structures enriched with oxygenated functional groups. The resulting CQDs were purified through ethanol washing, filtration, and fine syringe filtration, yielding stable colloidal dispersions with strong photoluminescence and surface functionality suitable for photocatalytic applications. CQDs are particularly promising because they can mediate electron transfer, suppress charge recombination, and promote hydrogen evolution or the production of value-added chemicals under solar irradiation. Mechanistic studies further reveal that CQDs enhance photocatalytic disinfection and organic pollutant degradation by generating reactive oxygen species and stabilizing charge carriers [90].

Graphene-like biomass-derived carbons have emerged as a promising class of sustainable photocatalytic materials and attractive alternatives to conventional graphene. Produced by the controlled carbonization and exfoliation of lignocellulosic biomass through

pyrolysis or hydrothermal processes, these materials contain graphene-like graphitic domains but lack the long-range crystallinity and perfect single-layer structure of pristine graphene. Their defect-rich carbon framework, abundant oxygen-containing functional groups, and heteroatom dopants provide active sites for semiconductor anchoring, promote interfacial charge transfer, and enhance reactant adsorption. Combined with their layered structure, good electrical conductivity, tunable surface chemistry, low cost, and renewable origin, these characteristics make graphene-like carbons efficient electron mediators and supports for semiconductor photocatalysts, with growing applications in photoreforming for hydrogen production. Typically, lignocellulosic biomass or agricultural residues are subjected to pyrolysis, hydrothermal carbonization, or chemical activation, followed by exfoliation or graphitization to yield nanosheets with layered structures and abundant oxygenated functional groups [92].

Moustafa et al. reported the preparation of graphene-like carbons from rice straw through washing, drying, chemical activation (H_3PO_4 or KOH), and controlled pyrolysis. The activation sequence and carbonization temperature determine the development of porosity, graphitic domains, and surface chemistry, yielding materials with high surface area, good electrical conductivity, and tunable electronic properties that promote charge separation and light harvesting in photocatalytic applications. When integrated with semiconductors such as TiO_2 , $\text{g-C}_3\text{N}_4$ or transition-metal phosphides, biomass-derived graphene-like carbons act as electron reservoirs, co-catalysts, and conductive supports, thereby suppressing charge recombination and extending visible-light activity [93]. Their role in photoreforming is particularly significant, as they facilitate electron transfer from biomass-derived reductive groups to semiconductor photocatalysts, enabling simultaneous hydrogen evolution and production of value-added chemicals. Recent studies have demonstrated the enhanced photocatalytic activity of semiconductor materials in the presence of graphene-like carbons prepared from biomass carbon stocks for hydrogen generation [94].

Among carbon-based materials employed as co-catalysts and supports for semiconductor photocatalysts, graphene-derived materials have attracted particular attention because of their outstanding electrical conductivity, large specific surface area, and excellent electron-transport capability, facilitating the extraction and transport of photogenerated electrons when coupled with semiconductors [95], suppressing electron-hole recombination, improving semiconductor dispersion, and providing abundant anchoring sites for active phases, so they can play the role of the noble metals (often Pt) used as co-catalysts in photoreforming. As a result, graphene-based materials have been extensively investigated for photocatalytic hydrogen evolution by photoreforming. Graphene oxide (GO) and reduced graphene oxide (rGO) are among the most widely studied members of this family (see Figure 9).

Unlike graphene-like carbons, which are produced directly from biomass through carbonization or hydrothermal processing, rGO is generally obtained by reducing graphite-derived graphene oxide (GO). During this process, a significant fraction of the oxygen-containing functional groups is removed, partially restoring the conjugated sp^2 carbon network while retaining structural defects and residual surface functionalities that are beneficial for photocatalytic applications. In green synthesis approaches, biomass-derived compounds such as polyphenols, flavonoids, sugars, lignin derivatives, and plant extracts are used as sustainable reducing agents, replacing hazardous chemicals such as hydrazine or sodium borohydride. These natural reductants not only restore the electrical conductivity of GO but also preserve surface functionalities that strengthen semiconductor anchoring, facilitate interfacial charge transfer, and promote charge-carrier separation. Consequently, biomass-assisted rGO has emerged as an efficient electron mediator and conductive support for semiconductor photocatalysts, enhancing photocatalytic hydrogen production and

photoreforming while providing a greener route for the synthesis of advanced graphene-based composites.

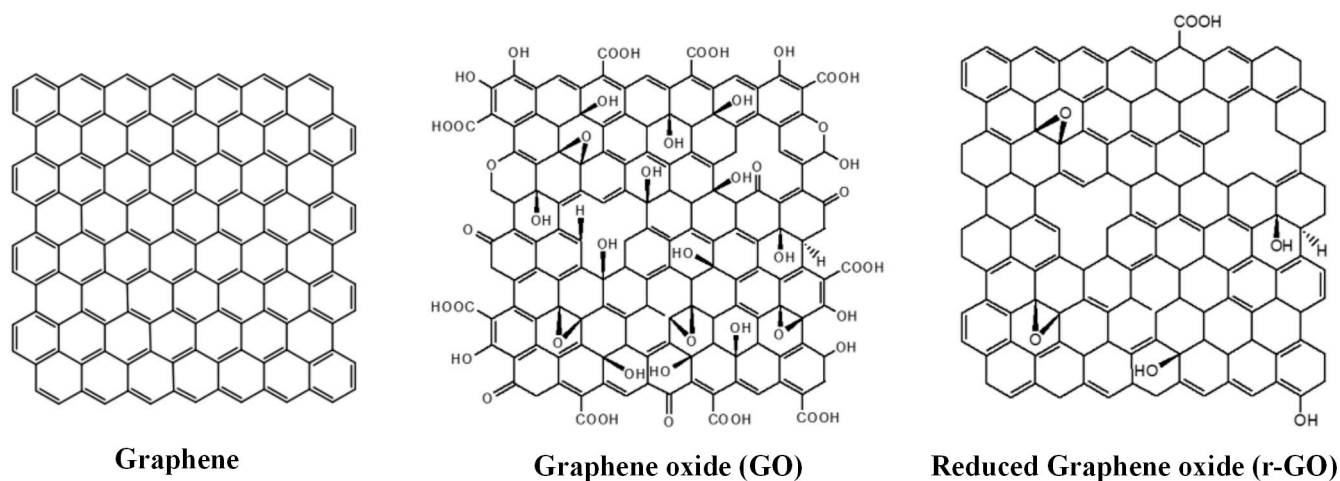


Figure 9. Schematic illustration of the structure of the graphene type-based materials, evidencing the oxygen-containing functional groups present in the materials.

As a representative example, Kasuga et al. synthesized reduced graphene oxide (rGO) from orange peel waste through carbonization followed by hydrazine reduction and subsequently integrated it with TiO₂ by ball milling. The incorporation of biomass-derived rGO substantially improved the photocatalytic H₂ production activity of TiO₂-based composites, with the optimized catalyst reaching hydrogen production rates of about 27 mmol h^{−1} g^{−1}. This enhancement was mainly ascribed to the conductive rGO network, which promotes efficient electron transport, suppresses charge recombination, and facilitates interfacial charge transfer [95].

Coffee waste has also emerged as an important biomass precursor for photocatalyst preparation due to its high carbon content, intrinsic heteroatoms, and porous organic matrix, which can be transformed into functional carbonaceous materials through pyrolysis or hydrothermal treatment, as schematised in Figure 10. These coffee-derived carbons exhibit enhanced surface area, tunable electronic properties, and strong affinity for semiconductor nanoparticles, enabling improved charge separation and light-harvesting efficiency in photocatalytic systems. Waste coffee silverskin (CSS), a by-product of coffee roasting, has emerged as a sustainable precursor for the synthesis of graphene oxide-like carbon materials through a low-cost and environmentally friendly ozone oxidation process. After carbonization, nitrogen functionalization, and ozone treatment, the resulting silver skin graphene oxide (SGO) exhibited metal-free photocatalytic activity, achieving H₂ production rates of 24.34 mmol g^{−1} h^{−1} in ultrapure water and 27.50 mmol g^{−1} h^{−1} in secondary treated wastewater under sunlight irradiation, demonstrating its potential for sustainable solar hydrogen production [63].

Recent advances in sustainable materials science have underscored the importance of life cycle assessment (LCA) in evaluating the environmental footprint of graphene production. A comparative LCA conducted by Bahmei et al. revealed that graphene synthesized from biomass waste via KOH – assisted pyrolysis is substantially more sustainable than conventional graphene obtained through the Hummers method, which relies on strong oxidants and generates hazardous effluents [96]. The biomass-derived route not only valorizes agricultural and forestry residues but also reduces reliance on fossil-based precursors, thereby aligning with circular economy principles. Among the feedstocks assessed, poplar wood waste exhibited the lowest environmental impact, reflecting its favorable composition and availability. However, the study identified electricity consumption during

pyrolysis and the use of KOH as critical environmental hot spots, highlighting the need for process optimization and greener activation strategies. These findings emphasize that while biomass waste offers a promising sustainable precursor for graphene, further improvements in energy efficiency and chemical utilization are essential to fully realize its environmental advantages [97].

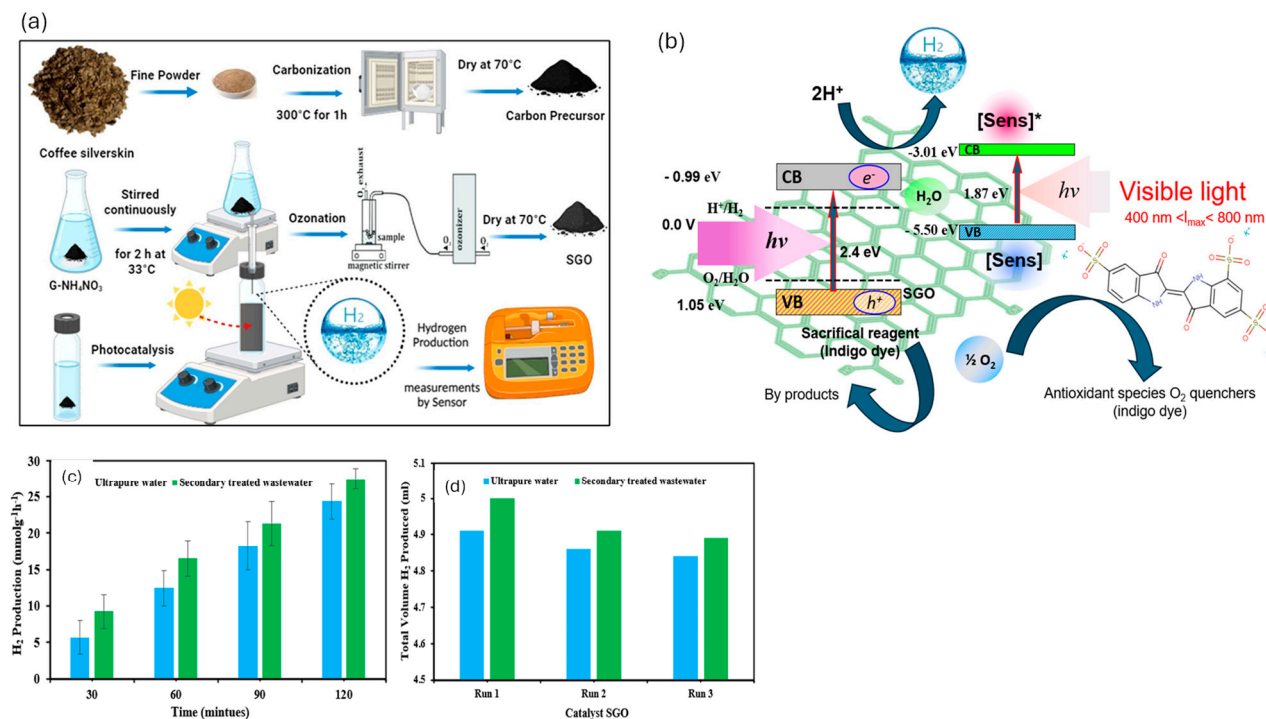


Figure 10. (a) Preparation route of silver skin graphene oxide (SGO) from waste coffee silverskin (CSS) via carbonization, nitrogen functionalization, and ozone treatment. (b) Proposed mechanism for photocatalytic hydrogen evolution by SGO under sunlight irradiation. (c) H₂ production versus reaction time. (d) reusability of the photocatalyst [63].

While most biomass-derived carbon materials primarily serve as conductive supports, co-catalysts, or electron mediators for semiconductor photocatalysts, graphitic carbon nitride (g-C₃N₄) represents a notable exception. As an intrinsically active metal-free semiconductor, biomass-derived g-C₃N₄ has emerged as one of the most promising sustainable photocatalysts owing to its earth-abundant nitrogen-rich precursors, suitable band-edge positions, visible-light activity, and tunable electronic structure. Unlike other biomass-derived carbon materials, g-C₃N₄ is capable of directly driving photocatalytic reactions without requiring an additional semiconductor, making it a versatile platform for solar-driven hydrogen production and biomass photoreforming.

Typically prepared via thermal polycondensation of nitrogen-rich biomass residues (e.g., amino acids, proteins, or agricultural wastes), g-C₃N₄ exhibits a layered structure with conjugated heptazine units that facilitate charge transport and redox reactions. Beyond conventional synthesis, microwave-assisted chemical modification of g-C₃N₄ has been shown to tailor surface functionalities and band structures, enabling efficient organic degradation, hydrogen production, and selective alcohol oxidation under visible light [98]. It can also be used with r-GO as a co-catalyst, exhibiting enhanced H₂ evolution from triethanolamine solutions [99]. Its versatility also allows for the preparation of heterojunction materials to outperform the activity of the single semiconductors. For instance, García-López et al. reported the use of Nb₂O₅/g-C₃N₄ composites in the photoreforming of triethanolamine and other organics in aqueous suspension. The integration of thermo-etched g-C₃N₄ with

Nb₂O₅ markedly improved photocatalytic photoreforming performance through enhanced interfacial charge separation. The optimized composite achieved hydrogen production rates of ca. 46 mmol h^{−1} g^{−1}, representing a 4–5-fold increase over pristine g-C₃N₄ with Nb₂O₅ alone, with an apparent quantum efficiency of approximately 14% under UV LED irradiation [96]. These studies, among others, are examples of how g-C₃N₄, when combined with semiconductor oxides and co-catalysts, provides excellent results for solar-driven photoreforming and hydrogen production. The presence of graphitic carbon domains facilitates electron transport, while pyridinic and graphitic nitrogen species create catalytically active sites that promote charge separation and surface redox reactions, enabling photocatalytic H₂ production and pollutant degradation, albeit with lower activity than conventional semiconductor photocatalysts [67].

To facilitate a clearer comparison of the different biomass-derived photocatalysts discussed in this section, we have included a systematic summary table that compiles the essential synthesis parameters for each material. This table provides an organized overview of the material type, biomass feedstock, synthesis method, resulting properties, and corresponding references, offering readers a structured and accessible way to evaluate the various preparation strategies and their outcomes (Table 1).

Table 1. Systematic Comparison of Biomass-Derived Photocatalyst Preparation Methods.

Material	Biomass Feedstock	Synthesis Method	Resulting Properties	Ref.
carbon quantum dots	Sucrose	Microwave-assisted	Homogeneous, simultaneous, and rapid heating, resulting in a uniform size distribution	[67]
	Citric acid	Microwave-assisted	Uniform size distribution	[67]
	Resoles (phenol/formaldehyde resins) and silica spheres	Template method	Mild conditions, tailorable sizes and properties	[67]
	Glucose, chitin, and chitosan	Hydro-/solvothermal method	High quantum efficiency, simple operation, and low cost	[67]
biochar-integrated TiO ₂	Sewage sludge	Sol–gel	High specific area	[84]
Biochar support for CdS/TiO ₂	Wood chips	Hydrothermal	High specific area	[85]
AC/TiO ₂ nanotubes	Walnut shell	Pulverization followed by chemical activation	High specific area	[87]
Co ₃ O ₄ -AC	Neem leaves	Sonication then Hydrothermal treatment	Large specific surface area	[91]
CQDs	Cellulose	Hydrothermal	Quasi-spherical morphology (7 nm)	[89]
Graphene-like carbons	Rice straw	Chemical Activation followed by pyrolysis	Charge separation	[96]

Table 1. Cont.

Material	Biomass Feedstock	Synthesis Method	Resulting Properties	Ref.
TiO ₂ -rGO	Orange peels	Pyrolysis followed by reduction	High specific area	[95]
SGO	Waste coffee silverskin (CSS)	Carbonization, nitrogen functionalization, and ozone treatment	Enhanced surface area and tunable electronic properties	[63]
Nb ₂ O ₅ /g-C ₃ N ₄	Melamine	Thermal treatment	High specific area	[96]
TiO ₂	Olea europaea	Biotemplate	Macroporous photocatalyst	[100]
Carbon-decorated polymer carbon nitride	Melamine on organic yeast	One-pot surface-assisted polymerization	Morphology tuning and composition tailoring	[101]
Cu(OH) ₂ -Ni(OH) ₂ /TiO ₂	Corn straw	Renewable biomass sacrificial agent	Non-noble metal photocatalysts with natural biomass sacrificial agents	[102]
TiO ₂ -Ce	Coffee Waste	Free solvent, one-pot templated reaction	Morphology control	[103]
g-C ₃ N ₄	Yeast	One-pot surface-assisted polymerization	Controlling morphology and electronic structure	[104]
g-CNQDs/TiO ₂	Formamide and urea	Thermal-chemical etching process	Strong blue emission	[105]

4.2. Biomass as a Sustainable Sacrificial Template for the Synthesis of Photocatalysts

Beyond serving as a carbon precursor, biomass has emerged as an effective sacrificial template for the sustainable synthesis of advanced photocatalytic materials. Owing to its naturally occurring hierarchical architectures, diverse morphologies, and rich chemical composition, biomass can direct the formation of porous and nanostructured photocatalysts with controlled size, morphology, and surface properties. During synthesis, inorganic precursors infiltrate or coat the biomass framework through physical adsorption or chemical interactions. Subsequent thermal treatment converts the precursor into the desired photocatalyst while the organic template is removed by pyrolysis or calcination, leaving behind a replica of the original biological structure. In some cases, the biomass is partially carbonized and becomes an integral component of the final material, providing conductive carbon domains that enhance photocatalytic performance. In other cases, only trace amounts of biomass-derived components remain in the final material; nevertheless, these components can influence the final photocatalytic features of the material. This sacrificial templating strategy represents a green and cost-effective alternative to conventional synthetic templates, which often require expensive reagents and additional chemical etching steps for removal.

The remarkable structural diversity of biomass offers unique opportunities to tailor photocatalyst morphology. Natural templates such as wood, bamboo, cotton, leaves, pollen, and agricultural residues feature ordered vascular channels, fibrous networks, and hierarchical pores that can be faithfully replicated during synthesis [106]. These bio-inspired architectures yield materials with high specific surface areas and interconnected porosity, enhancing light scattering, reactant diffusion, and active-site exposure in photoreforming systems [107]. This template-driven approach is especially important for cellulose- and lignocellulose-based platforms, which have been widely developed into hybrid, supported, and porous photocatalysts for hydrogen evolution and biomass valorization [108,109].

Recent studies and reviews also show that hierarchical porous designs and biomass-derived carbon can further strengthen photocatalytic performance by improving structure control and facilitating efficient reforming of biomass into fuels and chemicals [60,110]. This can be explained because when partial carbonization occurs, the resulting carbon matrix acts as an electron-conducting network that promotes charge transport, suppresses electron–hole recombination, and strengthens the interaction between the semiconductor and the support, leading to enhanced photocatalytic efficiency.

Numerous semiconductor photocatalysts have been synthesized using biomass as sacrificial templates, including TiO_2 , ZnO , Fe_2O_3 , WO_3 , BiVO_4 , and graphitic carbon nitride ($\text{g-C}_3\text{N}_4$). For example, wood and cellulose have been extensively employed to fabricate TiO_2 monoliths and hollow porous structures that preserve the original vascular channels of the biomass, resulting in improved light harvesting and mass transport [111]. Similarly, cotton fibers and bacterial cellulose have been used to prepare one-dimensional semiconductor architectures with enhanced crystallinity and charge separation. In other systems, biomass-derived activated carbon obtained after template removal remains incorporated into the final composite, forming semiconductor/carbon heterojunctions that facilitate electron transfer and significantly improve hydrogen evolution during biomass photoreforming. These studies demonstrate that biomass-templated photocatalysts outperform analogous materials synthesized without biological templates owing to their superior structural organization and more efficient utilization of photogenerated charge carriers.

Some representative examples can be given, for instance, ordered macroporous photocatalyst fabrication with *Olea europaea* leaves used as a biotemplate to make TiO_2 -based photocatalysts later coupled with $\text{g-C}_3\text{N}_4$ and Pt [100], and yeast-derived biomass carbon was used in a surface-assisted polymerization route to form biomass carbon-decorated polymeric carbon nitride [101]. Also, Lin et al. report that corn straw was employed as a renewable biomass sacrificial agent for photocatalytic H_2 production using a non-noble-metal $\text{Cu}(\text{OH})_2\text{-Ni}(\text{OH})_2/\text{TiO}_2$ photocatalyst. The study demonstrated that the nature and pretreatment of the biomass significantly influence hydrogen evolution. In particular, urea pretreatment of corn straw markedly enhanced H_2 production by partially removing lignin, modifying the biomass microstructure, and improving the accessibility of the carbohydrate fraction [101]. These findings highlight that the physicochemical characteristics of biomass substrates, in addition to catalyst composition, play a crucial role in determining the efficiency of biomass photoreforming.

The intrinsic hierarchical organization of biomass, ranging from the micro- to the nanoscale, provides a unique scaffold for directing the formation of functional materials with tailored pore structures and high surface areas. It is well reported in the literature that biomaterials and biomass have been extensively explored as sustainable templates for the synthesis of ordered and hierarchical porous materials across a wide range of applications, including zeolites, metal oxides, carbon materials, and other functional porous solids. In particular, recent advances have demonstrated the effectiveness of biomass-templating strategies in controlling the crystallization, pore architecture, and textural properties of zeolites, highlighting the growing importance of renewable biological templates in the design of advanced porous materials [112].

Unlike conventional synthetic templates, biomass-derived structures provide naturally occurring three-dimensional morphologies, hierarchical organization, and abundant surface functional groups that can direct the nucleation and growth of inorganic materials. For instance, microorganisms, in particular, have attracted considerable attention because of their uniform size, structural diversity, ease of cultivation, and renewable nature. Their cell surfaces contain a variety of functional groups capable of binding metal ions, making them excellent scaffolds for the controlled synthesis of nanostructured photocatalysts [113].

Among microbial templates, *Escherichia coli* has been extensively investigated owing to its widespread availability, low cost, and well-defined rod-shaped morphology. In one representative approach, *E. coli* cells were employed as sacrificial templates for the synthesis of $\text{TiO}_2/\text{Au}/\text{TiO}_2$ hollow heterostructures. Initially, Au^{3+} ions were adsorbed onto the bacterial cell wall through electrostatic interactions and subsequently reduced by biomolecules present on the cell surface or by bacterial metabolites, leading to the in situ formation of Au nanoparticles [114]. A TiO_2 layer was then deposited around the bacteria-Au composite, and a final calcination step removed the organic template while preserving the hollow architecture. The resulting $\text{TiO}_2/\text{Au}/\text{TiO}_2$ microcapsules combined the localized surface plasmon resonance (LSPR) of Au nanoparticles with whispering gallery mode (WGM) optical effects, resulting in enhanced light harvesting and improved photocatalytic activity. This bio-templating approach demonstrates how microorganisms can simultaneously act as structural templates and reaction platforms for nanoparticle formation, providing a sustainable route for the fabrication of highly efficient photocatalytic materials while facilitating the recovery of valuable metals and minimizing the use of synthetic templating agents.

Coffee waste has also been successfully employed as a sustainable biotemplate for the synthesis of mesoporous TiO_2 and Ce-doped TiO_2 photocatalysts for solar hydrogen production. In particular, the Ce-doped composites prepared through a solvent-free, coffee waste-templated route exhibited enhanced photocatalytic performance for hydrogen photoreforming [115]. The incorporation of cerium species significantly improved the reduction capability of TiO_2 and the coffee waste biotemplate gave the material the optimal physico-chemical features to promote optimum H_2 production from aqueous ethanol without the use of noble-metal co-catalysts. The highest hydrogen evolution rates were achieved at an optimal Ce loading of approximately 3 wt.%, with the Ce-doped titania composites outperforming pristine benchmark TiO_2 [103].

The preparation procedure of the biotemplated photocatalyst architecture is schematised in Figure 11.

Furthermore, the photocatalysts exhibited a remarkable apparent quantum efficiency (AQE), highlighting their effective utilization of solar photons for hydrogen generation and demonstrating the potential of biomass-templated synthesis as a sustainable alternative to conventional preparation routes (see Table 2).

Table 2. Photoreforming results using TiO_2 -Ce nanocomposites [104].

Sample	H_2 [$\mu\text{mol h}^{-1} \text{g}^{-1}$] No Pt	AQE [%] No Pt	H_2 [$\mu\text{mol h}^{-1} \text{g}^{-1}$] with Pt	AQE [%] with Pt
TiO_2 CW	47.0	0.01	6288	2.0
TiO_2 CW-Ce 0.5%	110	0.03	2286	0.80
TiO_2 CW-Ce 1%	181	0.06	1484	0.50
TiO_2 CW-Ce 3%	312	0.10	540	0.20
TiO_2 CW-Ce 5%	90.0	0.03	288	0.08
TiO_2 Huntsman	0.00	0.00	8200	2.6

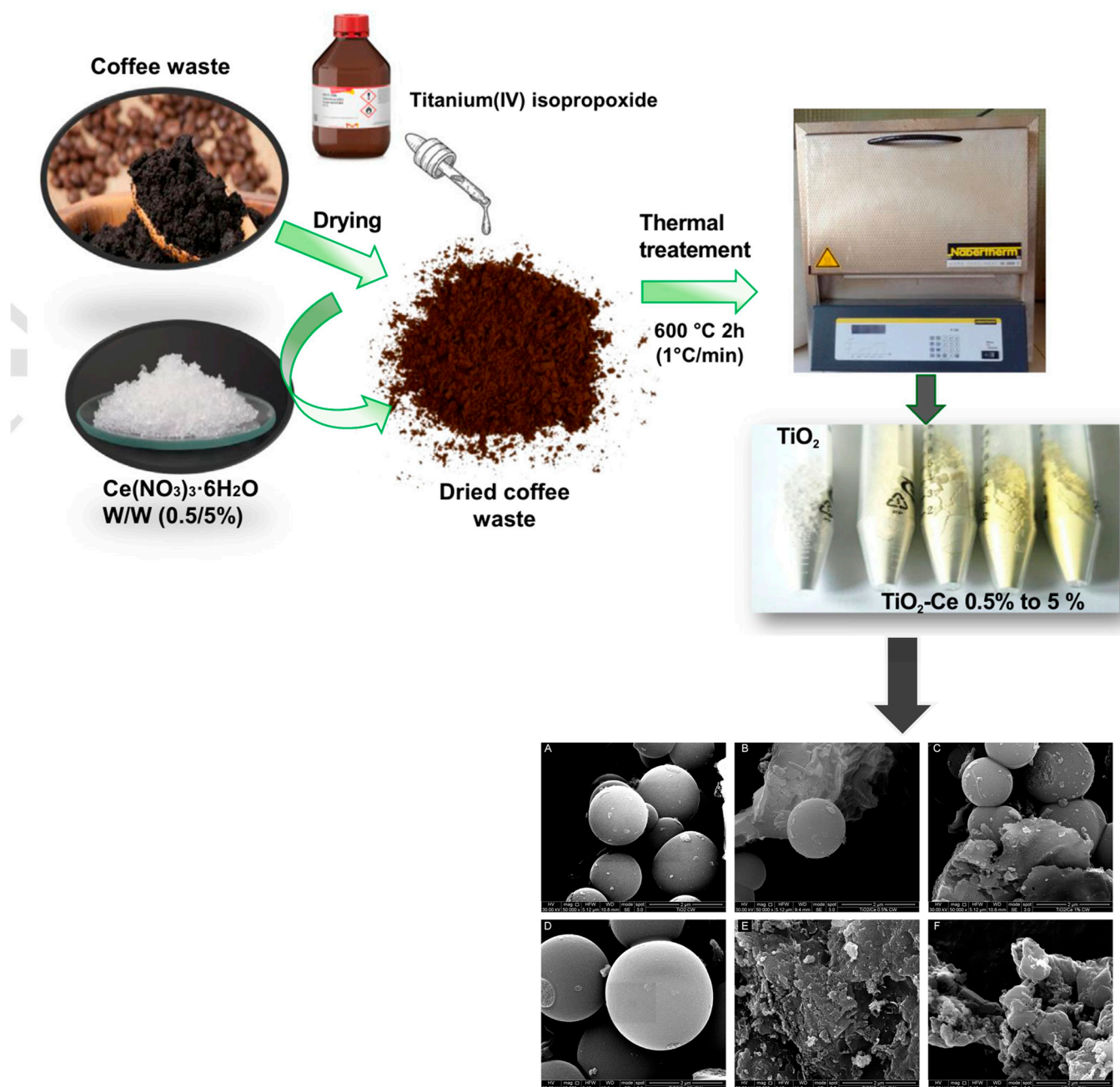


Figure 11. Schematic illustration of the synthesis process for Ce-TiO₂ nanocomposites, photo and SEM images of the obtained materials.

Biomass can also act as an effective template for controlling the morphology and electronic structure of polymeric carbon nitride. For example, yeast has been used as a structure-directing agent to synthesize carbon-modified g-C₃N₄ (CCN_x) through a one-pot surface-assisted polymerization strategy. Hydrogen-bond-mediated interfacial confinement between melamine and yeast promotes the formation of porous, thin-layer nanosheets while suppressing particle aggregation. Simultaneously, yeast-derived carbon extends visible-light absorption and enhances charge delocalization and separation, leading to a markedly improved photocatalytic H₂ evolution rate of 2704 μmol g^{−1} h^{−1}, substantially higher than that of pristine g-C₃N₄ [101]. These findings demonstrate that biomass can simultaneously function as both a carbon source and a morphology-directing template, providing a simple and sustainable strategy for engineering highly efficient g-C₃N₄-based photocatalysts [44,104,116].

In another recent example, olive leaves have been employed as biotemplates for the synthesis of hierarchical TiO_2 photocatalysts, enabling faithful replication of both the external leaf morphology and the internal porous architecture. The biotemplated structure promoted the formation of mesoporous TiO_2 with enhanced visible-light absorption, attributed to structural defects and oxygen vacancies introduced during the templating process. Subsequent photodeposition of Au and Pt nanoparticles preserved the leaf-like architecture, with Pt exhibiting a finer and more homogeneous dispersion than Au. The schematic preparation method of those catalysts is reported in Figure 12.

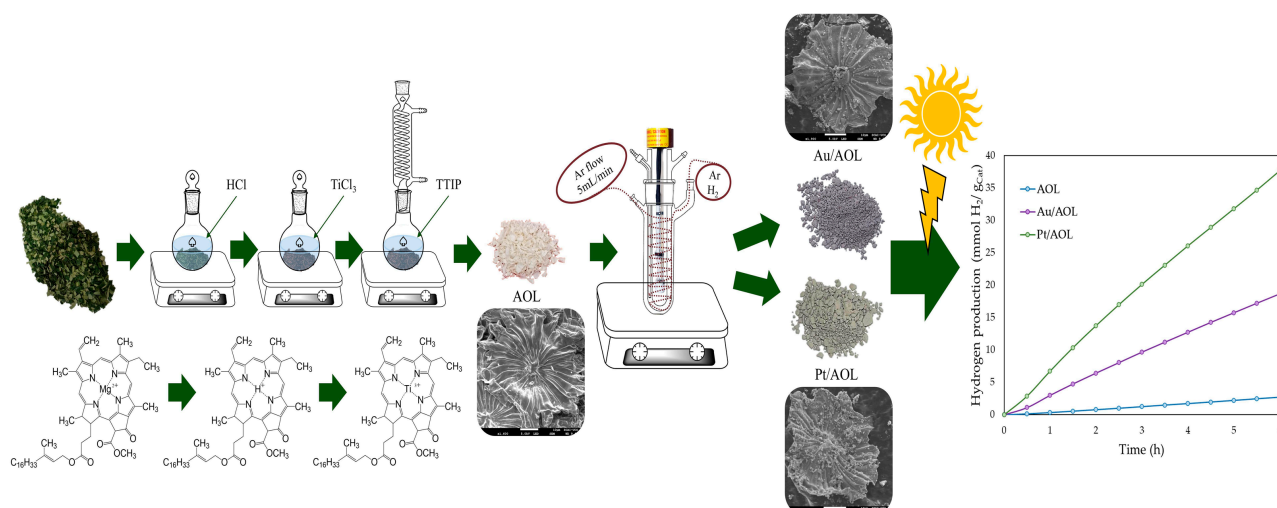


Figure 12. Schematic illustration of the biotemplated synthesis of olive leaf-derived hierarchical TiO_2 photocatalysts and subsequent photodeposition of Au and Pt nanoparticles for glycerol photoreforming [117].

The resulting biotemplated photocatalysts showed markedly enhanced glycerol photoreforming activity, with Pt/ TiO_2 achieving up to a 23-fold increase in H_2 production under simulated solar irradiation owing to improved charge separation through strong metal-support interactions and Schottky junction formation [118]. To enable a clearer comparison of the photoreforming performance of different biomass-derived photocatalysts, Table 3 compiles the key operational and performance indicators reported in recent studies. The table summarizes the material type, biomass feedstock, hole scavenger, Pt loading, irradiation system, and resulting H_2 productivity, along with the corresponding references. This comparative overview highlights the diversity of biomass-derived materials and provides a direct benchmark against conventional photocatalysts, facilitating a more critical assessment of their efficiency and practical applicability in solar-driven hydrogen production.

Table 3. Comparative Performance Indicators of Biomass-Derived Photocatalysts in Photoreforming Systems.

Material	Biomass Feedstock	Hole Scavenger	Pt Wt	Irradiation System	H_2 Productivity	Ref.
SGO	Waste coffee silverskin (CSS)	(wastewater)	-	Sunlight	$27.50 \text{ mmol g}^{-1} \text{ h}^{-1}$	[63]
Biochar-integrated TiO_2	sewage sludge	Glycerol (5%)	-	Xenon 35 W HID lamp	$2.523 \text{ } \mu\text{mol/g h}$	[84]
Biochar support for CdS/TiO_2	Wood chips	Glucose (50 mg/L)	-	300 W xenon lamp	$12.77 \text{ mmol g}^{-1} \text{ h}^{-1}$	[85]

Table 3. Cont.

Material	Biomass Feedstock	Hole Scavenger	Pt Wt	Irradiation System	H ₂ Productivity	Ref.
AC/TiO ₂ nanotubes	walnut shell	Methanol (0.15 mol/L)	-	500 W xenon lamp	18.000 mmol ⁻¹ g ⁻¹ h ⁻¹	[87]
Co ₃ O ₄ -AC	Neem leaves	Sulphide wastewater (0.2 M)	-	Solar irradiation	~70 mL/h	[91]
TiO ₂ -rGO	Orange peels	Water and ethanol (9:1, molar ratio)	1%	4 UV LED	27 mmol h ⁻¹ g ⁻¹	[95]
Graphene-like carbons	Rice straw	MB dye solution (10 mg/L)	-	Visible halogen lamp (2000 W)	32.718 µmol g ⁻¹ h ⁻¹	[93]
C ₃ N ₄	Melamine	Triethanolamine (0.27 M)	1%	2 UV-LED	3.117 µmol g ⁻¹ h ⁻¹	[99]
TiO ₂ (P25)	Commercial	Triethanolamine (0.27 M)	1%	2 UV-LED	51,839 µmol g ⁻¹ h ⁻¹	[99]
TE-g-C ₃ N ₄	Melamine	Triethanolamine (0.27 M)	1%	2 UV-LED	9 mmol g ⁻¹ h ⁻¹	[96]
g-C ₃ N ₄	Melamine	Triethanolamine (0.27 M)	1%	2 UV-LED	1 mmol g ⁻¹ h ⁻¹	[96]
Nb ₂ O ₅ /g-C ₃ N ₄	Melamine	Triethanolamine (0.27 M)	1%	2 UV-LED	46 mmol h ⁻¹ g ⁻¹	[96]
TiO ₂	Olea europaea	Glycerol 10%	1%	UV LED	3.053 µmol·g ⁻¹ ·h ⁻¹	[100]
carbon-decorated polymer carbon nitride	Melamine on organic yeast	Triethanolamine (10%)	3%	300 W xenon lamp	2704 µmol g ⁻¹ ·h ⁻¹	[101]
Cu(OH) ₂ -Ni(OH) ₂ /TiO ₂	Corn straw	Corn straw liquid solution	0.5%	300 W xenon lamp	1.08 mmol g ⁻¹ ·h ⁻¹	[102]
TiO ₂ -Ce	Coffee Waste	Ethanol (0.39 M)	1%	2 UV LEDs	6.288 µmol g ⁻¹ ·h ⁻¹	[103]
TiO ₂ (Hatsman)	Commercial	Ethanol (0.39 M)	1%	2 UV LEDs	8.200 µmol g ⁻¹ ·h ⁻¹	[103]
g-CNQDs/TiO ₂	Formamide and urea	Methanol (10%)	1%	Visible light	≈460 µmol g ⁻¹ h ⁻¹	[105]
Cu ²⁺ /biochar/TiO ₂	Corn Straw	Bio-sacrificial (corn straw extracts)	0.5%	300 W xenon lamp	6.72 µmol g ⁻¹ h ⁻¹	[119]

4.3. Biomass-Derived Reducing Agents for the Green Synthesis of Photocatalysts

In addition to serving as precursors for carbonaceous photocatalysts and as structure-directing templates, biomass-derived materials have increasingly been exploited as sustainable reducing agents for the green synthesis of semiconductor photocatalysts and their composites. Conventional reduction methods typically employ hazardous chemicals, such as hydrazine, sodium borohydride, or ethylene glycol, for the reduction of metal ions or graphene oxide (GO), raising concerns related to toxicity, environmental impact, and processing costs. In contrast, biomass extracts are naturally enriched in reducing compounds, including polyphenols, flavonoids, reducing sugars, proteins, amino acids, organic acids, and other antioxidant molecules, which can efficiently donate electrons to metal precursors under mild reaction conditions. Besides acting as reducing agents, these biomolecules also function as capping and stabilizing species, preventing nanoparticle aggregation, promoting homogeneous nucleation and growth, and enabling fine control over particle size, morphology, crystallinity, and surface chemistry. Furthermore, through coordination with metal cations and selective adsorption on specific crystal facets, biomass-derived molecules can influence crystal growth, leading to hierarchical nanostructures with enhanced dispersion of active components. Residual surface functionalities originating from the biomass may also improve hydrophilicity, facilitate reactant adsorption, and strengthen interfacial interactions between the semiconductor and co-catalyst, ultimately promoting charge separation and photocatalytic activity. Consequently, biomass-mediated synthesis has emerged as a versatile and environmentally benign strategy for photocatalyst fabrication, fully consistent with the principles of green chemistry.

A broad range of renewable feedstocks, including tea leaves, coffee waste, fruit peels, flower and leaf extracts, lignin, cellulose, starch, chitosan, and agricultural residues, has

been successfully employed as reducing agents for the preparation of photocatalytic materials, as schematised in Figure 13. Among the most extensively investigated applications is the green synthesis of noble metal nanoparticles (Au, Ag, and Pt) supported on semiconductor photocatalysts, in which phytochemicals reduce metal ions without the need for additional chemical reductants [118–120].

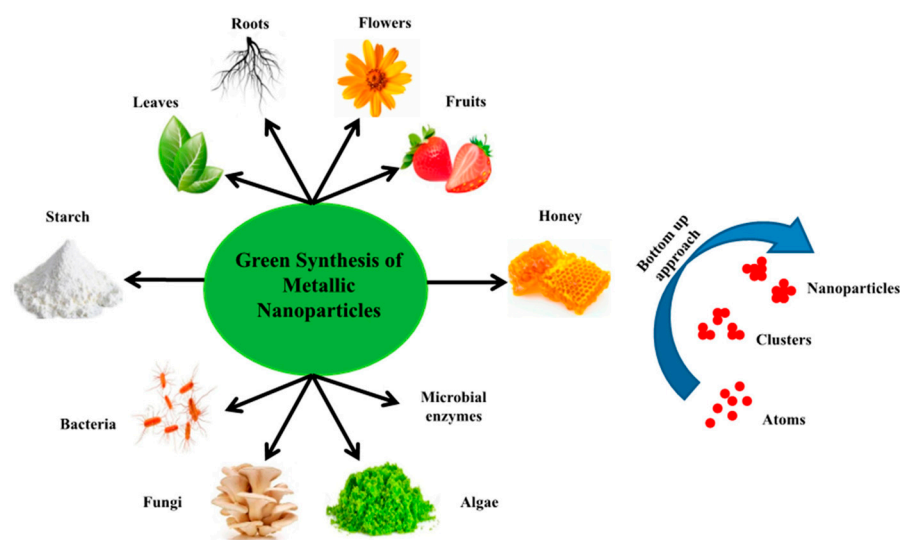


Figure 13. Different types of green synthesis used for the preparation of metal nanoparticles, as reported in [120].

The resulting metal nanoparticles generally exhibit high dispersion and intimate interfacial contact with semiconductor supports, favouring the formation of Schottky junctions that efficiently trap photogenerated electrons and suppress electron–hole recombination. In the case of Au and Ag nanoparticles, the localized surface plasmon resonance (LSPR) effect further extends light absorption into the visible region, enhancing solar energy utilization [121].

Although the use of biomass-derived metallic nanostructures has been largely restricted to photocatalytic pollutant degradation, their application in photoreforming remains scarcely explored. Given that these nanostructures enhance photocatalytic performance through improved charge separation, accelerated interfacial electron transfer, and increased light harvesting, they hold considerable promise for H₂ production via biomass photoreforming. Their integration into photoreforming photocatalysts therefore represents an attractive avenue for future research.

Another important application of biomass-derived reducing agents is the preparation of reduced graphene oxide (rGO)-based photocatalysts. Polyphenol-rich plant extracts, glucose, lignin derivatives, starch, cellulose hydrolysates, and ascorbic acid have been successfully employed to replace hydrazine in the reduction of graphene oxide. Unlike harsh chemical reductants, these biomass-derived compounds restore the conjugated sp² carbon network while preserving oxygen-containing functional groups that improve the interfacial coupling between rGO and semiconductor nanoparticles [27].

For example, Reduced graphene oxide (rGO) was successfully synthesized through a green reduction route using phytochemical extracts from *Citrus grandis* (pomelo) and *Tamarindus indica* (tamarind) as natural reducing agents. The biomass-derived extracts effectively removed oxygen-containing functional groups, restoring the graphitic structure and yielding graphene nanosheets with electrical conductivities approximately four orders of magnitude higher than those of graphene oxide. This simple and environmentally

friendly approach demonstrates the potential of fruit extracts for the sustainable, large-scale production of rGO that can be used in photoreforming [122].

As a result, biomass-reduced rGO can be integrated with TiO_2 , CdS, ZnIn_2S_4 , and $\text{g-C}_3\text{N}_4$, where it acts, as demonstrated by alternatively reduced GO, as an efficient electron mediator, accelerating charge transport and suppressing charge-carrier recombination. These green-synthesized composites can demonstrate enhanced photocatalytic performance in hydrogen evolution and biomass photoreforming, highlighting the potential of biomass-assisted reduction for the sustainable fabrication of advanced photocatalysts.

Despite these promising developments, the application of biomass-mediated reduction to the synthesis of photocatalysts specifically designed for photoreforming remains relatively limited. Most state-of-the-art photoreforming systems containing Pt, Au, Pd, or Ni co-catalysts are still prepared by conventional methods such as photodeposition, impregnation, or chemical reduction. Nevertheless, biomass-assisted reduction represents a highly promising alternative, enabling the preparation of well-dispersed co-catalysts under mild and environmentally friendly conditions while eliminating hazardous reducing agents. The integration of renewable biomass-derived reductants into photocatalyst synthesis is therefore expected to play an increasingly important role in the development of sustainable materials for hydrogen production by photocatalysis [123].

By integrating renewable biomass-derived reductants into photocatalyst synthesis, researchers can design eco-friendly materials that couple waste valorization with solar-driven hydrogen production. This strategy is expected to play an increasingly important role in the development of sustainable photocatalytic systems, bridging the gap between laboratory innovation and scalable green energy technologies.

5. Conclusions

Biomass has emerged as a highly versatile and sustainable platform for the development of photocatalysts for solar-driven photoreforming. Biomass can fulfil multiple roles, ranging from carbon precursor and templating agent to reducing agent, co-catalyst, and support for semiconductor photocatalysts. This multifunctionality, together with the abundance, renewability, and low cost of biomass resources, makes biomass-derived materials attractive for the design of environmentally benign photocatalytic systems capable of simultaneously producing hydrogen and valorizing waste streams.

Significant progress has been achieved in the synthesis of biomass-derived photocatalysts, including biochar, hydrochar, carbon quantum dots, graphene-like carbons, heteroatom-doped porous carbons, and biomass-derived graphitic carbon nitride. These materials exhibit tunable structural, optical, and electronic properties that can be tailored through the choice of biomass precursor, synthesis route, and post-treatment strategy. Biomass-derived materials have also demonstrated considerable potential as conductive supports, templating agents, and green reducing agents, enabling improved semiconductor dispersion, enhanced interfacial charge transfer, efficient charge separation, and environmentally friendly synthesis routes that avoid hazardous chemicals.

Despite these advances, the application of biomass-derived materials to photocatalytic photoreforming remains at a relatively early stage compared with their extensive use in photocatalytic hydrogen evolution and environmental remediation. Many biomass-derived materials are still primarily employed as supports or conductive additives rather than as intrinsically active photocatalysts, with the possible exception of $\text{g-C}_3\text{N}_4$, and systematic correlations between biomass composition, carbon microstructure, surface chemistry, and photocatalytic performance remain insufficiently understood. Furthermore, comparisons between different biomass sources and synthesis strategies are often complicated by the lack of standardized photocatalytic testing protocols.

Future research should focus on establishing rational design principles that correlate biomass composition and processing conditions with photocatalytic performance. Particular attention should be devoted to heteroatom engineering, defect modulation, hierarchical pore design, and interface engineering to optimize light harvesting, charge transport, and catalytic activity. The development of noble-metal-free photocatalysts, scalable and low-energy synthesis routes, and integrated biomass valorization strategies will be essential for improving the economic and environmental sustainability of these materials. In addition, life-cycle assessment and techno-economic analysis should become integral components of future studies to evaluate the real sustainability of biomass-derived photocatalysts beyond laboratory-scale demonstrations.

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