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Green Photocatalysts in Water Treatment Using Green Materials

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Abbreviations

► Advanced Oxidation Processes (AOPs); ► Conduction band (CB); ► Graphene oxide (GO); ► Localized surface plasmon resonance (LSPR); ► Photocatalytic degradation activity (PCA); ► Valence band (VB)

Definition

Advanced oxidation processes (AOPs) are a promising technology to remove various pollutants such as pharmaceuticals and dyes from wastewaters. The photocatalytic processes are the most promising techniques, especially visible light and solar responsive photocatalysts. The produced photocatalytic radicals are strong oxidants and unselectively react with nearby chemical species. The intermediates undergo higher decomposition in a series of tandem radical reactions resulting in complete decomposition to H_2O, CO_2, and inorganic salts. The reaction necessitates the reactant adsorption and desorption on the surface of the photocatalyst. Experiments should perform under dark conditions to evaluate the absorption effects of the photocatalyst. After that, the reactor is irradiated with UV-vis light and produces radicals. The semiconductor generates e^- and h^+ pairs and radicals under radiation (Ortiz 2019).

In the last 20 years, photocatalysis as an AOPs has been an essential strategy for wastewater treatment (Edla et al. 2015). A photocatalyst needs light irradiation for the structural decomposition of organic pollutants in the photocatalysis process. The photocatalysis efficiency is measured with photocatalytic degradation activity (PCA) (Orlandi et al. 2019). Recent research efforts focus on producing materials capable of efficiently harvesting solar light and its application in green wastewater processes (Fendrich et al. 2019).

Introduction

The significant water pollutions are environmental and global changes, agriculture,

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industrialization, human activities, and deforestation. The water resources nature worsens because of the ongoing growth of chemicals and pollutants in the water bodies, including organic and inorganic pollutants. For example, some heavy metals, mercury, lead, cadmium, arsenic, nickel, and chromium, are the most dangerous inorganic pollutants in industrial wastes (Naushad et al. 2016). Different organic contaminants have been found in numerous water bodies, including coloring agents, organic dyes, oils, pesticides, fertilizers, phenols, hydrocarbons, pharmaceutical waste, and detergents (Sharma et al. 2015).

Several technologies are available to treat polluted water, such as ion exchange, reverse osmosis, adsorption, and biological treatment. Reverse osmosis and ion exchange require operational costs, while biological treatment and adsorption are cost-effective processes. A valuable method for the removal of hazardous pollutants in wastewater treatment is the adsorption process. The big challenge to this process is the cost of materials required for this process. Most adsorption processes use activated carbon as the adsorbent to remove organic contaminants in the environment. However, commercially available activated carbons are costly. Thus, it is necessary to reduce the production cost using new advanced technologies.

Mechanistic Aspects

Fujishima and Honda first reported the photocatalytic behavior of semiconductor materials in 1972 (Fujishima et al. 1972). They investigated the light-current response of TiO2 after UV irradiation. Many researchers have used photocatalytic purification techniques to evaluate photoexcitation processes. Absorption of sufficiently energetic photons by a catalyst causes the excitation of an electron (e_{cb}^-) from the semiconductor valence band (VB) to an empty conduction band (CB), creating a positively charged hole (h_{vb}^+) (Scheme 1). Both of these e_{cb}^- / h_{vb}^+ species also act as charge carriers. A semiconductor requires the bandgap energy to transfer electrons from VB to CB after photon absorption (Fujishima et al. 2008). This energy is required for the photocatalyst activation to produce the e_{cb}^- / h_{vb}^+ pair (Eq. 1). Other photon wavelengths may be absorbed but may not have the energy required to form an electron-hole pair.

$$\text{Semiconductor} + \text{hv} \rightarrow e_{cb}^- + h_{vb}^+ \qquad (1)$$

The amount of e_{cb}^- / h_{vb}^+ pair produced by irradiation penetration in the photocatalyst depends on the following factors: the absorption coefficient of the substance, the wavelength of the radiation source, and the photon diffusion. However, the electrons are unstable in an excited state and tend to return to the ground state in picoseconds during recombination with h_{vb}^+, releasing heat (Eq. 2) (Schneider et al. 2014).

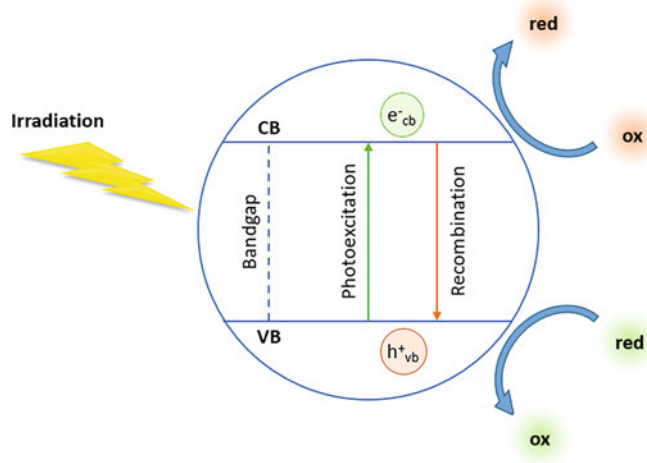
$$e_{cb}^- + h_{vb}^+ \rightarrow \text{Semiconductor} + \text{heat} \qquad (2)$$

Green Photocatalysis

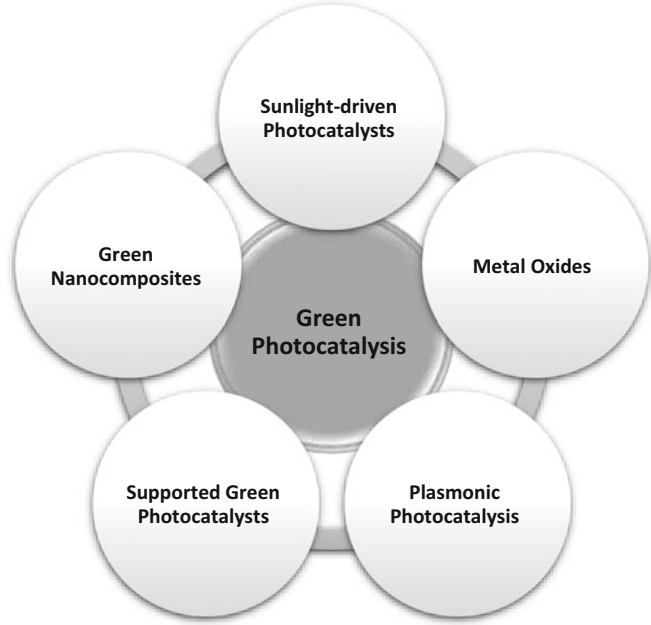
Nowadays, there is a tremendous demand for high-performance technology to treat wastewater. Photocatalysis uses green chemistry principles for wastewater treatment. In addition, the role of green nanomaterials in water and wastewater treatment has significantly increased by altering the nanomaterials features. This section summarizes recent progress in green photocatalysis, including sunlight-driven photocatalysts, metal oxides, plasmonic photocatalysis, supported green photocatalysts, and green nanocomposites in wastewater treatment (Scheme 2).

Sunlight-Driven Photocatalysts

Finding an appropriate photocatalyst to remove wastewater contaminants using solar energy is one of the essential missions. A green photocatalyst would have the preferred bandgap for efficient solar light absorption and water stability throughout the photocatalytic processes. In addition, a green photocatalyst should be cost-



Green Photocatalysts in Water Treatment Using Green Materials, Scheme 1 Photocatalytic excitation leading to separation from charge carriers (e_{cb}^{-} / h_{vb}^{+}) and recombination processes



Green Photocatalysts in Water Treatment Using Green Materials, Scheme 2 Recent progress in green photocatalysis

effective, operate efficiently, be readily accessible, and be environmentally friendly. In recent years, various sunlight-driven nanophotocatalysts were designed and applied as green photocatalytic materials in wastewater remediation. Recent research has shown that transition metal ions are efficient materials for effective photocatalysts (Wang et al. 2008). Table 1 summarizes some influential aspects, which are effective in the photocatalytic process.

Metal Oxides

Nanostructures based on metal oxide semiconductors were used for green photocatalytic applications toward wastewater treatment. The most critical conditions for a green effective photocatalytic material include the required bandgap, morphology, increased surface area, chemical stability, and reusability. Some metal oxides such as TiO_2, Cu_2O, ZnO, SnO_2, and WO_3 with these

Green Photocatalysts in Water Treatment Using Green Materials, Table 1 The determining factors affecting the photocatalytic process

Determining factor		Description	References
Catalyst	Amount	Increasing the number of catalysts increases active sites in the photocatalytic reaction, resulting in more reactive radicals in the photodegradation process	Reza et al. (2017)
	Morphology	The morphology directly links the catalyst surface and organic pollutants. Photons on the photocatalyst surface control the photocatalytic activity rate. Therefore, the reaction proceeds more rapidly by the photocatalysts with different morphologies	Zhu et al. (2006)
	Surface area	Materials with more significant surface areas can create several active sites on the photocatalyst surface, creating numerous reactive radicals for an effective photodegradation process	Ameen et al. (2012)
	Dopants	Some strategies can fabricate TiO_2 nanomaterials to absorb photons at low energy efficiency. These strategies are made up of bandgap modification through introducing metals and nonmetals in photocatalyst composition	Rajeshwar et al. (2008)
Pollutant	Concentration	The pollutant adsorption on the photocatalyst surface is directly proportional to the pollutant concentration. The degradation rate decreases with a growing pollutant concentration, though the required amount of photocatalyst would be preserved	Akpan and Hameed (2009)
	Nature	The nature of pollutants determines the photodegradation degree in a wastewater matrix. In the high concentration of the contaminant, some photocatalysts cannot remove the pollutant. In addition, it may saturate the photocatalyst surface and prevent the formation of active radicals, resulting in reduced photocatalyst effectiveness	Mills et al. (1993)
Conditions	Temperature	Performing reactions at temperatures between 0 °C and 80 °C resulted in efficient photocatalytic performance. However, above the temperature of 80 °C, the photocatalyst will promote the electron-hole pairs' undesirable recombination and decrease photocatalyst activity	Mamba et al. (2014)
	pH	The photocatalyst surface potential varies by changing the solution pH. The contaminant adsorption on the photocatalyst surface initiates a change in the photodecomposition rate	Davis et al. (1994)
	Irradiation	The photodegradation rate is inversely related to the light intensity at high light intensities as the formation of excitons hinders the undesirable recombination of electron-hole pairs. Besides, the recombination of electron-hole pairs happens at the photocatalyst surfaces by increasing the light power, and thus the photocatalytic activity reduces in the reaction mixture	Asahi et al. (2001)

favorable features have the same photocatalytic efficiency. These green materials excite photogenerated charge carriers to create holes, oxidizing organic substances (Maeda 2011). Nanostructures based on metal oxide semiconductors are efficiently activated with sunlight, UV-visible light, or a mixture of these light irradiations.

The electron-hole pairs generated by photoexcitation of charge carriers are efficiently involved in the oxidation and chemical reaction to destroy the organic pollutants. The metal oxide activity as photocatalyst is due to the formation of hydroxyl radicals through the chemical oxidation of OH^- reactive species and reactive superoxide radicals by reducing O_2. Organic pollutants are affected by these radical reactive species to nontoxic byproducts.

Several metal oxides in nanoscale forms, such as TiO_2, ZnO, WO_3, and Fe_2O_3, were used in water treatment (Fendrich et al. 2020). Zinc oxide (ZnO), a direct bandgap and n-type semiconductor, has appeared to be a hopeful metal oxide for efficient environmental remediation.

The unique features of this metal oxide make it suitable for environmental applications. ZnO can generate reactive oxidants, including hydroxyl and superoxide reactive radicals (Belhaj et al. 2020). Zinc oxide also has high chemical and thermal strength and low toxicity, displaying optimum environmental compatibility. Finally, concerning economic considerations, ZnO is potentially cheaper than commercial TiO_2.

Plasmonic Photocatalysis

In recent years, plasmonic photocatalysts have grown exponentially owing to their enhanced photodecomposition effectiveness under visible light and sunlight irradiation and outstanding charge transport properties (Liang et al. 2018). Homogeneous dispersing metal nanoparticles produced these materials to the semiconductors' bodies. A Schottky barrier and localized surface plasmon resonance (LSPR) are achieved as distinct features (Wang and Astruc 2014). Under visible light irradiation, the charge carriers efficiently separate and transfer by these features. The LSPR is an essential characteristic of plasmonic photocatalysis, demonstrating strong oscillation on metal nanoparticle surfaces.

Supported Green Photocatalysts

Since discovering the photocatalytic influence by Fujishima and Honda (1972), abundant investigations were suggested to synthesize TiO_2 catalysts of diverse forms. The TiO_2 catalyst in nanodimensions has an excellent ratio of surface area to volume. It can promote effective charge separation and trap at the photocatalyst surface. The supported TiO_2 nanophotocatalysts have an improved oxidation ability compared to the same bulk TiO_2 photocatalysts. While the TiO_2 nanophotocatalysts show significant enhancement in chemical and physical properties, the size and morphology of nanoparticle is the main challenge in the industrial water treatment (Yu et al. 2002).

Carbon nanomaterials possess excellent physicochemical properties and have been studied extensively as solid support to remove pollutants from wastewater. While keeping the promising features of carbon-based materials, nanocomposites were efficiently developed to incorporate common photocatalytic species for effective light-derived water remediation. Fullerenes, graphene oxide (GO), and carbon nanotubes support photocatalytic materials such as TiO_2, SnO_2, ZnO, etc. Carbon-based nanomaterials afford a strong reduction ability to destroy harmful pollutants in wastewaters. Graphene-supported TiO_2 nanophotocatalysts displayed outstanding photocatalytic activity under UV light irradiation (Williams et al. 2008). The advance of this green nanocomposite was confirmed with various materials for better exercise. Graphene and its chemical derivatives display exceptional catalytic performance in the presence of multiple pollutants because of their extraordinary physicochemical properties, increased electron-accepting capacity, and improved electronic properties. These properties cause that graphene-based nanocomposite emerges as superior materials for photocatalytic remediation by tuning the appropriate features to affect the photocatalyst activity.

Alternative adsorbent materials should be of low cost, have high adsorption capacity, be available in large quantities, and regenerate. One of the best green materials for alternative adsorbents and photocatalyst immobilization is agricultural wastes. In the last decades, numerous researches on applying agricultural wastes have been conducted for wastewater treatment as alternative adsorbents. Various aspects of the use of low-cost adsorbents were reported in the literature (Levchuk et al. 2018). Nevertheless, with the growing published articles and the complication of the environmental remediation processes, it is required to afford updated data and logical discussions on agricultural waste as effective alternative adsorbents.

Green Nanocomposites

Green nanocomposites are used as an alternative method to increase responsibility for photocatalysts in various applications. Nanocomposites

have several advantages, such as being eco-friendly and it does not make second contaminants. TiO_2 and ZnO are chemically stable, producing an active charge during irradiation by light with suitable wavelengths (Ashouri et al. 2019). Zinc oxide is an n-type semiconductor with high excitation binding energy, bandgap width, biocompatibility, and it is more active when irradiated by visible light. The irradiation process for ZnO will create an electron/hole by gaining the electron energy to excite the conduction band (Singh et al. 2020). The electron at the conduction band moves quickly to the valence band by the recombination processes. The heterostructure method is a practical approach to control and minimize recombination rate in ZnO, consequently increasing the charge carrier lifetime (Ma et al. 2017).

The combination of TiO_2 with ZnO nanoparticles improves photocatalytic activities (Gupta et al. 2020). TiO_2 particles have higher efficiency because they respond to the visible spectrum with bandgap 3.2 eV (Al-Zahrani et al. 2020). It is applied as a photocatalyst due to its good optical properties, low cost, and high chemical stability. The catalyst was used for photodegradation of chemical or organic pollutants in wastewater under visible irradiation. Doping TiO_2 with metals or nonmetal ions will increase the UV-vis light absorbance capacity or reactivity (Qu et al. 2018). ZnO/TiO_2 nanohybrids as a photocatalyst were synthesized using a hydrothermal, solid-state, sol-gel method and TiO_2 commercial for pollutant degradation (Singh et al. 2020). ZnO/TiO_2 thin films prepared by the hydrothermal method were utilized for orange G degradation (Binupriya et al. 2008). However, green, appropriate, and efficient photocatalysts can be easily integrated into wastewater plants and reused for real applications. Biocompatibility, recyclability and the reusability in successive cycles are among the principles of these photocatalytic materials.

The materials widely used to produce nanocomposites include granular activated carbon, cellulose, silica, polymers, and sands (Zhao et al. 2011). Polymeric materials are principally a convenient choice owing to their controllable pore space and their outstanding mechanical strength. The resulting polymeric nanocomposite has nanoparticles' characteristic properties, whereas the polymer supports afford increased stability and processability. The nanoparticles can be supported on cellulose, porous resins, chitosan, and other solid supports (Wu et al. 2009). The choice of polymeric materials is usually determined by thermal and mechanical properties, chemical functional groups, hydrophobic, hydrophilic behavior, chemical stability, biocompatibility, and electronic properties.

Advances and Challenges in the Photocatalytic Water Treatment

Degussa P-25 TiO_2 is generally applied in wastewater treatment as a green photocatalyst. This photocatalyst is used to compare photoactivity under various conditions (Serpone et al. 1996). The Degussa P-25 TiO_2 fine particles were used in a slurry form, with an increased volumetric generation rate of ROS as proportional to the number of surface-active sites. The immobilization of photocatalysts into an inert substrate decreases active catalyst sites and mass transfer limitations. The catalyst immobilization makes the operation more complex such that the photonic activation cannot occur due to lack of photon penetration (Pozzo et al. 1997). Therefore, the application of TiO_2 catalyst in the slurry type is usually preferred. An additional process step of catalysts post-separation with the slurry TiO_2 system is needed. The separation process is vital to avoid photocatalyst loss and introduce the TiO_2 contamination as a new pollutant in the treated wastewater. The recovery of photocatalyst is reached over a combination with common sedimentation, cross-flow filtration, or membrane filtrations. Several significant operating issues through TiO_2 in the slurry form still stay even with a membrane integration development. These include the membrane types, pore size, recyclability, and fouling. Numerous studies have employed micron size for catalyst immobilization to increase surface contact with pollutants and avoid membrane fouling. These strategies

include catalyst immobilization on mesoporous clays, activated carbon, fibers, or the membrane.

Conclusion

Green photocatalytic materials have great potential to reduce contaminants in wastewater treatment. The green and safe methods of producing these green materials are crucial to wastewater treatment. The extensive use of photocatalysts in the adsorption and degradation of pollutants results in environmental remediation. Production of new green materials of high efficiency and low cost is currently a hot topic. However, most results are based on experimental tests on the laboratory scale. Therefore, many questions may need to be answered in mass production and field application. A green photocatalyst for water and wastewater treatment has requirements such as safety to the environment, the impossibility of leakage in water, recyclability, the commerciality of raw materials, and preferably having a natural origin photocatalyst. In addition to the above, one of the essential features of a green photocatalyst is its ability to excite with visible and solar light, facilitating its use on an industrial scale. Thus, recent researchers' efforts have focused on using green photocatalysts under visible light irradiation.

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