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Harnessing synergistic mechanisms in nanophotocatalysts for the degradation of aqueous organic pollutants: A review

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ABSTRACT

Photocatalysis represents an important procedure in the reduction of the environmental hazards posed by persistent organic pollutants and in the support of solar-driven chemical reactions. This review discusses the development of photocatalytic techniques that aimed to enhance light absorption and charge separation efficiency in order to increase the efficiency of photodegradation under solar and visible illumination. Structural, morphological, and spectroscopic analysis were reviewed in order to understand the relationship between the architectural aspects of the developed material and its support in photodegradation. In addition to that, the photocatalytic degradation of organic pollutants was discussed with several factors that affect the process. Enhanced photodegradation efficiency is a result of the specially developed band gap of the developed photocatalyst and increased ROS generation. The current review can help in the discussion and compare the advancement of modern and efficient water treatment techniques.

1. Introduction

In recent decades, wastewater treatment has emerged as a critical global concern due to its importance to public health and ecosystems [1]. Water is an essential resource in numerous industrial operations, including manufacturing, oil and gas processing, and sanitation. These industries are projected to account for roughly 20% of global freshwater consumption, illustrating their significant impact on water demand to support these critical activities [2]. The main contaminants of dyes, such as methylene blue, methyl orange, Congo red, and malachite green, sourced from the textiles, leather, paper, and food industries, can be harmful to the environment [3]. Organic dyes affect water quality, along with the formation of harmful byproducts, making them a cause for concern for environmental sustainability. In addition to, pharmaceutical compounds persist and resist removal from water by conventional methods of wastewater treatment because of their relatively stable structure and high hydrophilicity [4].

The conventional methods, such as sedimentation, biological processes, adsorption, filtration, and membrane processes, have long been practiced for removing contaminants. However, these methods often suffer from low efficacy or serve as a transfer process rather than a complete pollutant removal process. Advanced oxidation processes provide a new range of treatment for removing contaminants from water, and these processes assume a significantly important role for removing pollution from water due to their strong oxidation power, flexibility, and rapid rate of reaction [5]. Advanced Oxidation Processes (AOPs) generate highly reactive oxygen-based species, such as hydroxyl radicals (•OH), sulfate radicals ($SO_4^{\bullet-}$), singlet oxygen ($1O_2$), and high-valent metal species. These reactive mediators can quickly degrade harmful contaminants present in aqueous solutions. This method is especially efficient for persistent contaminants, such as dyes pollutants, pharmacologically active compounds, pesticides, and antibiotics. Classical AOP methods mainly mineralize these contaminants to H_2O, CO_2, and simple organic salts. Based on these techniques, new methods, especially nano-photocatalysis methods, for improving water treatments have appeared [6].

In 1972, Fujishima and Honda found that TiO_2 is capable of promoting the photodissociation of water molecules to evolve hydrogen and oxygen gases, thus laying the foundation for the domain of photocatalytic splitting of water. In 1976, researchers from Canada, along with Carey, discovered that the photocatalysis method can be effectively employed to break down

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highly toxic polychlorinated biphenyls, thus contributing to the domain of semiconductor-assisted photocatalytic decomposition of organic compounds. Today, a vast list of researchers is dedicated to the investigation and development of photocatalysis-based technologies. Photocatalysis is being largely employed for the splitting of water to generate hydrogen, reduction of carbon monoxide, fixation of nitrogen, decomposition of pollutants, sterilization, and disinfection, using solar energy as a non-toxic, eco-friendly energy carrier. On this note, photocatalysis is a far better method to mitigate environmental pollution and scarcity of natural materials [7].

The photocatalytic decomposition of contaminants in the printing and dyeing wastewater is also largely dependent on the factors of light absorption, charge separation, and surface redox sites. Charge separation and transfer play important roles, and sometimes they limit the efficiency of the photocatalytic decomposition process. It is thus a key area of research to optimize charge separation and transfer properties of the photocatalyst to achieve efficient decomposition of printing and dyeing wastewater through photocatalytic oxidation [8].

Metal/metal oxide nanoparticles have found applications of intense importance among the wider nomenclature of nanocatalysts. These nanocatalysts can be categorized based on size, composition, morphology, properties, aggregation, and uniformity. These nanoparticle systems include carbon-based materials, ceramic materials, semiconductor materials, polymeric materials, and metal/metal oxide nanoparticles. Of these, metal/metal oxide nanoparticles stand to gain importance because of their vast applications, such as biomolecule detection and imaging, antibacterial properties, environmental pollutant removal, and bioanalytical applications. Metal/metal oxide nanoparticles can be prepared from metal, metal oxide, or a mixture of metal/metal oxide. They include nanoparticles of materials such as silver, copper, gold, titanium dioxide, iron oxide, and zinc oxide. They can be prepared using methods such as chemical, physical, electrochemical, or photochemical methods [9].

This review offers a thorough investigation of photocatalysis systems and their efficacy for removing organic pollutants by different semiconductor heterojunctions. Additionally, the report offers a scientific discussion of the challenges and environmental implications of organic pollution. The photochemistry of semiconducting materials, carrier transport, and photoreaction processes have been explained clearly. Moreover, a range of innovative methods, such as hydrolysis, precipitation, basic calcinations, sonication-assisted mixture methods, and microwave methods, are being used for preparing nanoscale semiconducting materials and have been extensively analyzed. Additionally, a range of different semiconductor forms, including doping, defect-based materials, optimized morphological materials, and coupled semiconducting materials, have been covered to improve the efficacy of a single-component system. It is observed, however, that this comprehensive review reveals the relevant parameters that affect the mineralization of different organic pollutants by a range of semiconductor heterojunctions. This review discusses the basic strategies available through recent advances to expedite development processes for photocatalysis heterojunction systems.

2. Organic pollutants

The presence of organic pollutants, including dye molecules and pharmaceutical antibiotics, in aquatic systems is a significant global problem due to their effects on human activities and ecosystems [10].

2.1 Dyes Pollutants

Synthetic dyes are particularly resistant to remove because of their robust chemical structure and durability against degradation [11]. Dye compounds, including methyl orange (MO), methylene blue (MB), and rhodamine B (RhB), are widely utilised throughout diverse industrial sectors, encompassing food colouring, pH indicators, cosmetics, medicines, textile colouration, and printing industries [12]. The release of a high quantity of dye molecules to the aquatic system causes serious health and environmental concerns due to the toxicity of these molecules. The presence of organic dyes in wastewater from industries such as textiles, paper, and the garment industry is a contributing factor to environmental deterioration [13].

The dye effluents contain harmful materials that can cause serious health problems to human beings. When dyes are being processed, about 10-50% of them are lost, and this ends up forming coloured effluents. It is estimated that over 100,000 synthetic dyes are produced yearly, amounting to over 700,000 metric tons, hence generating a considerable amount of wastewater. The concentration of dyes is measured to range from 10 mg/l to 250 mg/l, and a maximum concentration of 800 mg/l is recorded for reactive dyes [14,15]. Methylene Blue (MB) is a widely used industrial dye, known for its frequent use as a model pollutant because of its stability, detectability, and blue colour. It is a serious pollutant because of its carcinogenic properties, toxicity, and ability to cause negative effects on aquatic organisms, as well as human organs such as the liver and kidneys [16].

2.2 Pharmaceutical Pollutants

Hospital water is increasingly being contaminated with residues of pharmaceuticals, especially antibiotics, that have become serious challenges for the environment because of their extensive application for human healthcare, veterinary practice, and agricultural purposes. These compounds are difficult to degrade because of their complexity [17,18]

Recent studies also suggest that a significant percentage of antibiotics is active and unmetabolized, estimated to be between 30% and 80%, and is excreted and eventually enters aqueous systems through sewage. It is estimated that due to the high use of drugs in human and veterinary medicine, these drugs enter the environment either as residues or as metabolites. Most drugs contain high polarization and low vapor pressure, making them highly persistent in aqueous systems. Even now, most of these drugs enter the resultant effluents of wastewater treatments, where these drugs can be incompletely removed. Previous works suggest that as much as 90% of these drugs can be found within the resultant effluents of wastewater treatments [19]. The presence of antibiotics for a prolonged period of time within the public water system can result in negative repercussions, including genetic mutations, the development of pathogen resistance, and possible carcinogenesis. It is, therefore, important that these compounds be removed from wastewater before being allowed to enter natural aquatic bodies [20]

Tetracycline (TC) is a widely used antibiotic drug that is applied for human and veterinary medical purposes, as well as for agricultural and aquaculture practices. Despite its importance for the promotion of health and development for animals raised for agricultural purposes, TC is a serious risk to the environment because of its poor bioavailability, contributing to a substantial excretion of this substance into the environment. For this reason, the total elimination of this antibiotic from aqueous media is a pressing task [21]

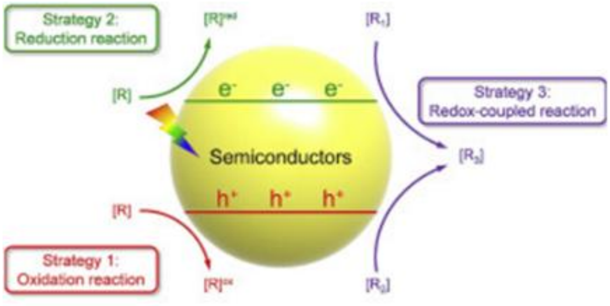
Tetracycline residues are extensively distributed in the worldwide aqueous environment, including surface, groundwater, and drinking water, at concentrations from nanograms to micrograms per liter. Tetracycline is a broad-spectrum antibiotic that affects microbial metabolic processes in natural environmental systems, contributing to the spread of antibiotic resistance genes. It is a potential hazard to the environment and human health [22].

3. Photocatalytic Reaction Mechanism

Photocatalysis is described as light-induced catalysis that catalyzes redox reactions in the presence of a semiconductor photocatalyst. In this process, when photons of energy at or above the bandgap of the semiconductor photocatalyst are incident on it, electrons can be raised from the valence band to the conduction band, and at the same time, an equivalent number of holes are formed in the valence band. Thus, electron–hole pairs are created that act as the key active species in photocatalysis. However, the role of this mechanism depends on the photon absorption capacity and charge carrier transfer of the photocatalytic material.

After excitation, the photogenerated charge carriers diffuse to the photocatalyst surface and undergo redox reactions. Conduction band electrons have strong reducing abilities and can readily react with molecular oxygen to generate reactive oxygen species (ROS), especially superoxide radicals ($O_2^{\bullet-}$). In turn, superoxide radicals can be converted to hydrogen peroxide (H_2O_2) or $^{\bullet}OH$ radicals and contribute to the oxidation of organic materials. Conversely, valence band holes have strong oxidative abilities and can readily oxidize adsorbed hydroxide ions or water molecules on the catalyst surface to generate additional $^{\bullet}OH$ radicals. High reactivity of $^{\bullet}OH$ radicals trigger non-selective oxidation of organic materials to CO_2, H_2O, and non-hazardous products.

The photocatalytic reaction is a complex process that consists of a series of photophysical and photochemical processes, such as photon absorption, charge carrier formation, separation, and transport, surface redox reactions, and mineralization of target molecules. The main impeding factor for the efficiency of a photocatalytic reaction is the high rate of recombination of the photogenerated electrons and holes, resulting in a considerable loss of energy. In this respect, different methods such as metal or non-metal doping, defect tuning, surface modification, or forming a heterojunction have been suggested to overcome this drawback and optimize charge separation, improve the absorption of photons in the visible region, as well as stabilize the photocatalytic system. Photocatalysis can be significantly affected by operational factors such as the intensity, wavelength, and morphologies of the photocatalyst, its surface area, crystallinity, and physicochemical properties of the reaction media. An ideal photocatalyst requires engineered properties such as a desirable band structure, high surface area, and high photoactivity even after long periods of illumination. It is a basic necessity to comprehend the underlying processes of photocatalysis for the rational design of efficient photocatalysts for practical applications such as the purification of polluted media, splitting of water, or solar fuels [23-26].



Advantages *of semiconductor-based photocatalysis: a) visible-light response, b) tunable redox potential, c) phase separation, and d) recyclability.*

Fig. 1 shows the photocatalytic reaction mechanism [27].

4. Synthesis Methods of Nanocomposite Photocatalysts

The approach used in the synthesis of nanocomposites determines the crystalline structure, morphology, specific surface area, and contact at the interface of the photocatalyst, and can be seen to influence photocatalytic activity directly. Many different methods of synthesis have had to be developed in order to allow for homogeneous phase distribution and well-characterized nanoscale structures that can promote charge transfer and light absorption. Hydrothermal and solvothermal processes are widely utilized owing to the fact that they can allow the crystal growth of the nanocomposites under controlled temperature and pressure conditions. In addition, sol-gel methods allow for the mixing of precursors at the molecular level, which can provide stoichiometric control on the nanoscale and achieve an optimal distribution of catalytically active components. Sol-gel methods are very useful when it comes to preparing nanocomposites that can present large surface areas and controlled porosity, aiming at maximizing absorption and catalytic properties.

Thus, coprecipitation is an effective and scalable process that allows for the simultaneous precipitation of different metallic ions in a controlled pH environment, leading to the precipitation of homogeneously mixed precursors that can be calcined to form stable and uniform composites. Similarly, impregnation and wet chemical deposition are common methods for loading secondary phases or co-catalysts onto semiconductor substrates with improved interfacial adhesion and optimized charge separation pathways.

In this regard, some novel, eco-friendly, and energy-efficient synthesis techniques have been developed recently, including microwave-assisted synthesis, ultrasonic-assisted techniques, and mechanochemical synthesis. These techniques shorten reaction time, improve dispersion, and develop defect-free surfaces. In all cases, the type of intended nanostructure, content, and specifications of the application determine the method selection. The effective control of synthesis parameters such as temperature, pH, precursor concentration, and calcination conditions is imperative to develop nanocomposite photocatalysts with superior photoactivity, stability, and reproducibility [28-30].

4.1 Hydrolysis Method

Hydrolysis can easily be applied to the synthesis of nanostructured composites of metal oxides. Besides, an aqueous or semi-aqueous process depends on the hydrolysis of metal precursors such as metal alkoxides, chlorides, or metal nitrates. In the synthesis process, metal ions proceed through the hydrolysis reaction to produce metal hydroxides, leading to metal oxide synthesis through a condensation reaction owing to the occurrence of dehydration reactions. The process allows the synthesis of nanostructured materials with controlled morphologies and sizes.

The hydrolysis reactions have two basic steps in the process of nucleation, where the hydrolysed components form nuclei; the second step involves the growth of nuclei to larger-sized particles through Ostwald ripening. These reaction rates can be easily controlled through changes in important factors such as precursor concentrations, hydrolysis ratios, pH, aging times, and temperatures. Controlled hydrolysis reactions can ensure uniform nucleation, prevent large agglomerations of particles, and ensure strong interactions between the components of the composite materials, which are important in achieving high photocatalytic activity.

It can be most effectively applied in the synthesis of heterostructure nanocomposites such as Bi_2MoO_6/TiO_2, Bi_2MoO_6/ZnO, and $Bi_2MoO_6/g\text{-}C_3N_4$, in which optimal distribution and strong binding at the interface are critical in facilitating interfacial charge transfer. In addition, hydrolysis synthesis can be considered an inexpensive and friendly process from an environmental perspective because it usually works under low-temperature and gentle reaction conditions without needing complex devices or poisonous reactants. It must be emphasized, nevertheless, that close control of reaction parameters is important to avoid rapid precipitation, agglomeration, or phase separation, especially when these aspects affect the efficiency of photo-splitting reactions and the long-term stability of the resulting compounds [31,32].

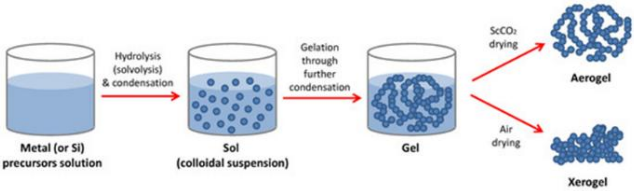


Fig. 2 shows the hydrolysis method [33].

4.2 Precipitation Method

The precipitation process is an easy and reliable synthesis procedure for preparing nano-composite photocatalysts with controlled composition, size, and morphology. In the process, metal ions from a homogeneous solution are precipitated using an appropriate precipitating agent like sodium hydroxide (NaOH), ammonia solution (NH_4OH), or sodium carbonate (Na_2CO_3). In the above process, metal ions from the solution precipitate in the form of insoluble metal hydroxides, carbonates, or basic salts, which further go through the process of dehydration and condensation during thermal treatment to form metal oxides.

The precipitation procedure generally consists of at least three main steps: (i) supersaturation of the solution, (ii) nucleation of solid particles, and (iii) development of the particles through aggregation. Nucleation versus growth: Nucleation and growth are equally important for determining the quality of the crystal and the particle size distribution of the final product. Achieving a well-defined morphology and phase composition in composite materials is highly reliant on controlling several reaction parameters during synthesis, such as the amount of reactants, pH of the reaction, temperature, stirring rates, and the reaction time for the aging process.

Precipitation techniques are widely used for the preparation of nanocomposites having heterojunctions for photocatalytic applications, including Bi_2MoO_6/WO_3, Bi_2MoO_6/ZnO, and $TiO_2/g\text{-}C_3N_4$. Precipitation methods favor the coupling of different semiconductor phases and lead to increased charge transfer at the junctions, which is an important criterion for effective functionality in heterojunctions. In addition, precipitation methods are preferred for easy processing, cost-effectiveness, environmental benefits, and scalability for the large-scale synthesis of the photocatalytic material. However, despite the advantages of this process, control of reaction conditions, such as particle agglomeration, phase separation, and the incomplete formation of the resulting product, remains important for this process. Secondary process steps such as washing, drying, and calcination are required for the removal of ion residuals and for enhancing the photocatalytic properties of the prepared nanocomposite material [34-37].

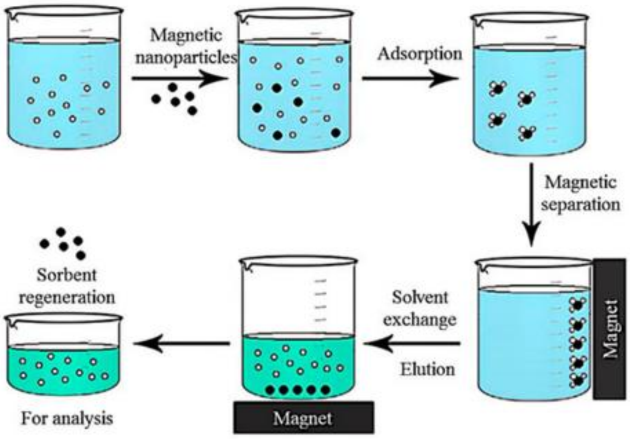


Fig. 3 Shows the Precipitation Method [38].

4.3 Hydrothermal Method

Hydrothermal synthesis can service as an effective synthesis procedure for the fabrication of nanocomposites functioning as photocatalysts with controlled morphology, crystal structure, and phase purity. Hydrothermal synthesis corresponds to the process of precursor crystallization in an aqueous medium under high-temperature and high-pressure conditions inside an autoclave at temperatures above 100ºC. In such cases, the solubility and reactivity of the precursors get enhanced, leading to favourable nucleation and nanoparticle aggregation. The reaction conditions, such as reaction temperature, pressure, pH, precursor concentration, and reaction time, can prominently affect the microstructure and physicochemical properties of the synthesized material.

By employing this method, it becomes easier to develop nanocomposites with strong interfacial interactions and superior heterostructure formation, which can significantly contribute to the charge separation efficiency and enhanced photocatalytic activity. By using the hydrothermal process, it becomes easier to integrate Bi_2MoO_6 with other semiconductor materials such as TiO_2, $g\text{-}C_3N_4$, or WO_3 to produce good heterostructures. In addition, the process can lead to the development of highly crystalline materials with fewer surface defects.

Hydrothermal synthesis is considered green, cost-effective, and flexible. It allows the synthesis of numerous nanostructures, such as nanosheets, nano-rods, and hierarchical nanostructures, without any post-synthesis high-temperature treatment. Thus, it has remained one of the most efficient methods for large-scale synthesis to date [39-42].

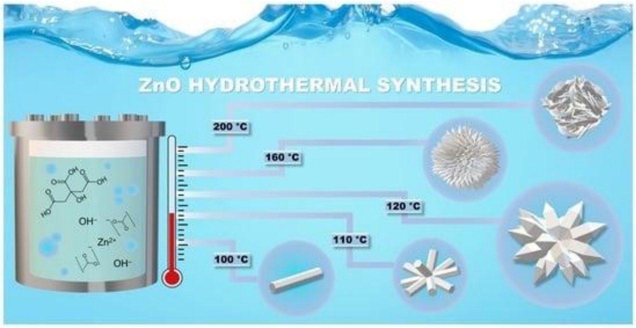


Fig. 4: The hydrothermal method [43].

4.4 Simple Calcination Method

The simple calcination technique is extensively utilized for the synthesis of nanostructured photocatalytic materials owing to its operational ease, scalability, and capacity to yield thermally stable crystalline phases. This method involves the initial preparation of precursor powders or gel-like mixes via the dissolving or co-precipitation of metal salts, followed by drying to eliminate residual solvents. The resultant solid is then subjected to regulated heating in a muffle furnace at a specified temperature, enabling the progressive disintegration of organic wastes and promoting phase transition into the intended oxide structure. Factors like calcination temperature, heating rate, and residence time are pivotal in determining particle size, surface area, and crystallinity. Elevated temperatures generally improve phase purity but can lead to particle development and a decrease in specific surface area, whereas moderate circumstances frequently produce nanoscale domains that are advantageous for photocatalytic processes. Due to its reproducibility and low chemical usage, the calcination method has been successfully employed in the synthesis of semiconductor photocatalysts for dye degradation, hydrogen production, and environmental remediation. This refined approach yields nanomaterials with superior charge separation and increased visible-light responsiveness, presenting a viable pathway for extensive photocatalytic applications. For instance, calcination results in carbon residue. The deposited CNAs underwent calcination in air at temperatures of 300, 400, 500, and 800 °C for durations of 1, 3, 5, and 12 hours, with a ramp-up rate of 20 °C·min−1 and a cooling rate of 2–3 °C·min−1. The selection of parameters was informed by the commonly documented calcination conditions for nanoparticles. Control CNAs were subjected to oxygen plasma treatment (7 W, 500 mTorr, 168 h) for comparative analysis. Optimized circumstances permit equally effective etching in as few as 6 hours [44-47].

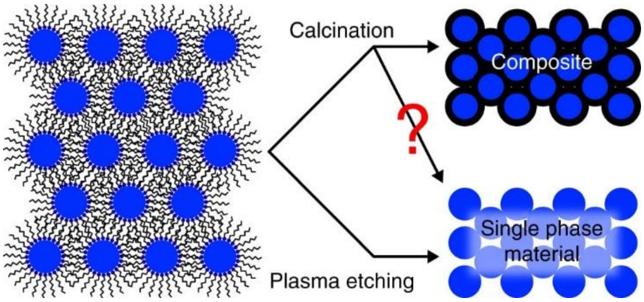


Fig. 5: The Simple Calculation Method.

4.5 Sonication Mixing Method

The sonication-assisted mixing process is an effective and widely adopted procedure in the synthesis of nanocomposite photocatalysts. It makes use of high-frequency acoustic waves in order to ensure uniform distribution and strong interaction between the components. In the process, acoustic cavitation takes place, where bubbles form and collapse in the liquid under exposure to ultrasonic waves, resulting in localized high temperatures and pressure in these bubbles. These changes increase the transfer of matter, prevent agglomeration of particles, and facilitate nucleation, resulting in uniformly dispersed nanoparticles that can be size-controlled.

In fact, because of its benefits in creating interfacial interactions that are essential in achieving charge separation, this synthesis procedure has numerous applications in the fabrication of heterostructure photocatalysts such as Bi_2MoO_6/WO_3, Bi_2MoO_6/TiO_2, and $Bi_2MoO_6/g\text{-}C_3N_4$. In most cases, sonication can be coupled with other synthesis procedures such as hydro-chemical synthesis, sol-gel synthesis, or precipitation in order to enhance the crystallinity, surface area, or overall performance of the resulting photocatalyst. Actually, there are several important factors of sonication that need to be optimized in order to prevent damage or defects to the precursor that can negatively affect the efficiency of the resulting photocatalyst.

The sonication-assisted mixing method presents an efficient, scalable, and green process suitable for the synthesis of complex nanocomposites with advanced physicochemical property profiles and outstanding photocatalytic performance under ultraviolet or visible light illumination. Its capability to generate high-quality heterostructures makes it an essential tool in modern-day research related to photocatalysis [48-51]

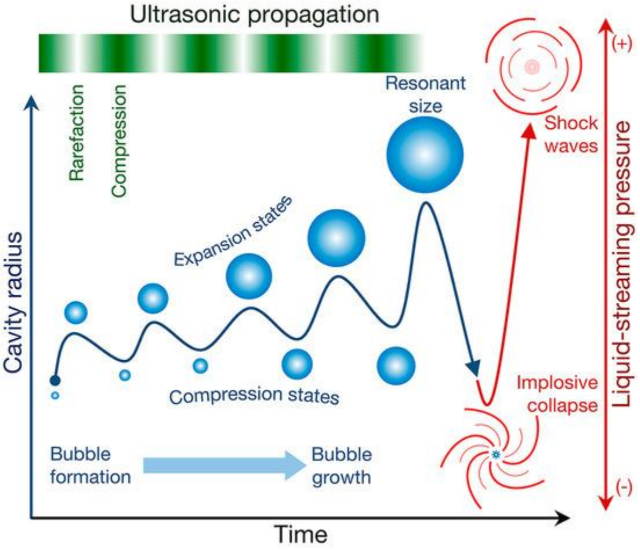


Fig. 6 shows the sonication mixing method [52].

4.6 Microwave Synthesis Method

The microwave-assisted synthesis technique has emerged as an important means of fast and accurate nanocomposite photocatalyst preparation. In comparison to traditional heating processes, microwaves directly involve polar molecules and ions in the reaction mixture, causing uniform volume heating due to dielectric losses. Such heating helps in nucleation and growth and allows for specific control over crystal and morphology properties.

It is especially useful for preparing heterojunction photocatalysts and

Bi_2MoO_6-based composites like $BiVO_4$, Bi_2WO_6, g-C_3N_4/$BiVO_4$, and others. In fact, Bi2MoO6-based composites prepared at lower temperatures and in shorter periods of time exhibit higher photocatalytic efficiency and light absorption due to higher recombination rates of charge carrier particles. In addition, higher recombination rates can be achieved due to higher electron and hole densities in semiconductive materials when higher power microwaves are used.

Optimization of the parameters, such as power of microwaves, irradiation time, solvent polarity, precursor concentration, and pH, is very important in order to get evenly sized particles and to avoid agglomeration and thermal treatment damage. In this process, the advantages of the microwave-assisted method are that it reduces reaction time, it is repeatable, energy and solvent-efficient, and scalable for use in practical applications or for further lab-based investigations. This process follows the principles of green chemistry [53-55].

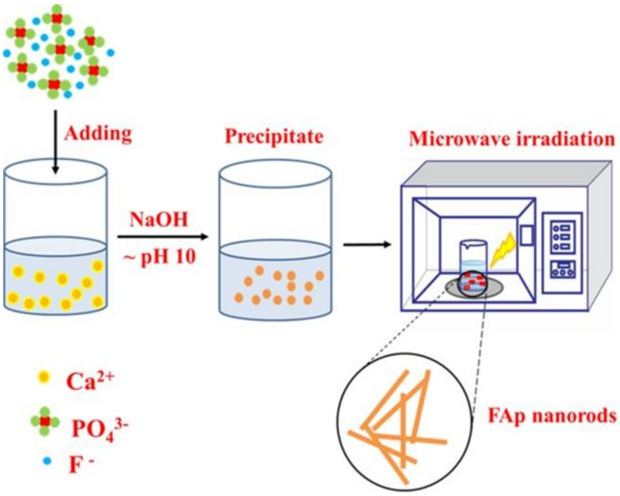


Fig.7 shows the microwave synthesis method [56].

5. Nanocomposite Photocatalysts

Photocatalytic methods have garnered considerable attention due to their widespread applications in solar power generation, pollutant removal, and organic transformations. In photocatalysis, the application of highly efficient and stable semiconductor photocatalysts is required. In addition to making use of solar light, it is important to develop catalysts that can work in the visible light range instead of semiconductors like Bi_2MoO_6, WO_3, ZnO, or Fe_3O_4 [57]

This can be explained on the grounds of inherent properties possessed by semiconductor materials. In this regard, semiconductors are known to have the following properties: (i) cost-effectiveness, (ii) non-toxicity, (iii) higher specific surface area that translates into an expanded contact area for reaction, (iv) large absorption spectra accompanied by higher absorption coefficients to allow for better light absorption over large ranges, (v) adjustable properties that can be controlled or altered in different ways such as downsizing, doping, and making use of sensitizers to promote photonic properties, (vi) compatibility with multi-electron transfer, thus making it easy for multi-electron transfer processes to occur, and (vii) stable operation for an extended period of time and accompanied by low degradations in photocatalytic properties after cycles of use. In addition, semiconducting particles that are separated from materials in systems involving filtration or centrifugation, or those fixed in fluid beds in which particles are suspended in fluid that flows in circular motion, retain considerable photocatalytic properties after many catalytic cycles [58]

By definition, photocatalysis can be described as the promotion of catalytic reaction rates when exposed to light. In practice, photocatalysis has promising applications within the secondary methods of wastewater treatment. Specifically, it can be very beneficial in the treatment of contaminated water sources containing persistent and toxic chemicals of anthropogenic origin. In this case, sufficient exposure of the photocatalytic surface to light and contact with the liquid must be achieved [59].

5.1 Doping of Single Photocatalysts

Doping is a useful means for improving the photocatalytic activities of individual semiconductor photocatalysts, in which foreign atoms are introduced into the crystal lattice of semiconductors. By this means, the band gap structure of semiconductors can be optimized, and light absorption and charge-carrier separation can be increased. In undoped semiconductors like TiO_2, ZnO, and WO_3, photocatalytic activities are only present in the UV range, owing to large band gap energies. However, in doped semiconductors, additional levels are formed inside the band gap, which can be excited only when exposed to the visible light range.

Metal dopants (for example, Fe, Cu, Mn) create donor levels in the lower energy side of the conduction band, which increases the excitation of electrons and facilitates charge transport. Non-metal dopants (for example, N, C, S), on the other hand, affect the valence band by hybridization of orbitals and decrease the band gap and light absorption [60-62].

5.2 Defect Engineering

Defect engineering consists of intentionally incorporating defects and irregularities in the crystal structure, such as vacancies and lattice defects, to promote photocatalytic reaction rates better. Oxygen vacancies in bismuth semiconductors and other semiconducting materials can act as midgap states, increase the absorption of visible light, and increase charge separation. Oxygen vacancies, in particular, are very effective in improving the amount of reactive oxygen species (ROS) and, as such, can be very advantageous in decomposing organic pollutants, in the generation of hydrogen gas, and in CO_2 reduction. Nonetheless, excessive defects can serve as recombination sites and must be carefully controlled. Defect engineering is an important tool for improving the efficiency of single and hybrid photocatalysts [63,64].

5.3 Morphology Optimization

Morphology optimization aims at intentionally adjusting the shape, size, and surface topography of photocatalytic materials for efficiency improvement. In bismuth-containing semiconductive materials and similar photocatalysts, optimized morphologies include nanosheets, nanoplates, nanorods, and hierarchical nano systems that can enhance the photocatalytic materials' surface area and active sites, as well as light harvesting capacity. Strategies in particle morphology that can favor charge separation and transportation in photocatalysts, as well as recombination of electron and hole couples and the yield of reactive oxygen species (ROS), include hydrothermal treatment and template-based methods, and such morphology control is of substantial importance in improving photocatalytic efficiency when irradiated with visible light [65,66].

6. Semiconductor Heterojunctions

A semiconductor heterojunction can be generally described as the junction that exists between two different semiconductor materials and has different band gap energies and electronic structures. In modern optoelectronic and photocatalytic applications, this junction is very important because it determines the charge carrier separation and transfer processes. By combining two semiconducting materials that have different levels of conduction and valence bands, it is possible to create an electric field at the junction point of the semiconductors. In turn, this electric field can be used to separate the charge carrier holes and electrons in opposite directions in a semiconductor material. Type II semiconductor heterojunctions are commonly used in photocatalysis because of their efficiency in achieving charge carrier separation in semiconductors. In addition, superior semiconductor topographies like Z-scheme and S-scheme semiconductor heterojunctions have been developed to improve the redox potential and efficiency of such systems. Semiconductor heterojunctions must be carefully and intentionally synthesized for use in materials applications in environmental remediation and solar energy conversion [67-69].

6.1 Type I Heterojunction

Type I heterojunctions, or straddling gap heterojunctions, are created when the minimum point of the conduction band (CB) and the top of the valence band (VB) of one semiconductor are located within the band gap of another semiconductor. For Type I heterojunctions, the semiconductor material with the smaller band gap is surrounded by the material that has the larger band gap. Consequently, both electrons and holes are confined in the same area. Exciton recombination can be facilitated due to this confining effect, which can be useful in some optoelectronic devices like LEDs and lasers [70]

In photocatalytic systems, Type I are relatively less favourable for achieving optimal charge separation because of the localization of charge carriers in the same semiconductor region. Therefore, this type of HJs commonly shows higher recombination rates and lower photocatalytic efficiencies. Despite such demerits, Type I can be utilized in combination with different frameworks or modifications on the semiconductor surface to improve charge diffusion and light absorption properties. Nonetheless, it is generally important to have in-depth insights into band alignments and charge carrier phenomena in Type I for optimal device functionality of multifaceted semiconductors [71]**.**

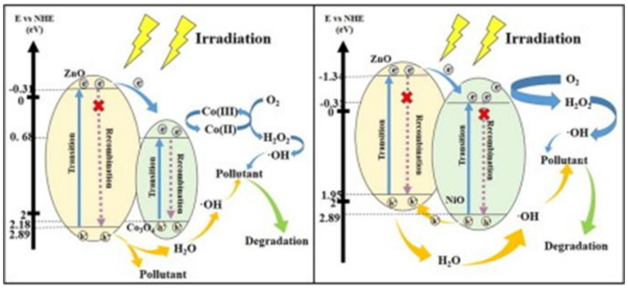


Fig. 8 shows the type I heterojunction [72]

6.2 Traditional Type II Heterojunction

Bi_2MoO_6: Despite being an Aireville-type oxide semiconductor featuring a relatively small band gap of around 2.6–2.8 eV and possessing excellent chemical properties and layered nanosheet morphology favoring charge transfer, Bi_2MoO_6 suffers from poor photocatalytic properties due to fast recombination of the photogenerated electrons and holes. In this situation, Bi_2MoO_6 materials are commonly combined with other semiconductors to create type-II heterojunctions [73]

In a standard type-II heterojunction, the CB edge of one material is higher in energy than that of the other, and the VB edge is relatively lower. Upon irradiation with visible light, the photoexcited electrons are transferred from the CB of Bi_2MoO_6 to the CB of the nearby semiconductor (for instance, WO_3, TiO_2, or ZnO), and the holes are transferred in the counter direction, from the VB of the counterpart to Bi_2MoO_6. By this means, the charge carriers are separated in space, and the ROS species like •OH and $•O_2^-$ are generated for the degradation of organic contaminants [74,75]

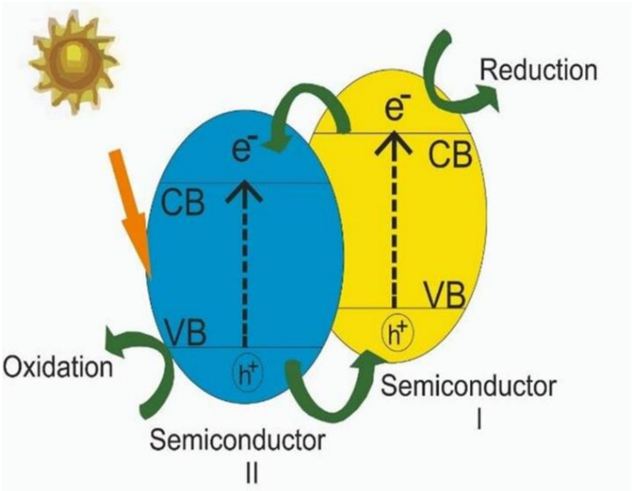


Fig. 9 shows the traditional Type II heterojunction [74]

6.3 S-Scheme Heterojunction

Bi_2MoO_6, which is a semiconductor that can be activated in the visible light range and possesses an Aurivillius phase and a relatively small band gap, has been widely researched for photocatalytic applications. Nonetheless, it suffers from the issue of fast recombination of the charge carrier pairs that are generated for photocatalysis. The design of an S-scheme type of heterojunction is an effective means of countering this problem in photocatalytic materials, and this approach can be achieved in an effective manner for Bi_2MoO_6 to promote the charge separation process and maintain substantial redox properties. In this case, the electric field in the Bi_2MoO_6-based S-scheme type of heterojunction and band bending at the junction help in achieving the recombination of low-energy hole and electron charge carrier pairs, and it also helps in keeping the highly energetic electrons in the CB of the reduction photocatalyst and highly potent oxidative VB holes in Bi_2MoO_6 [76-79]

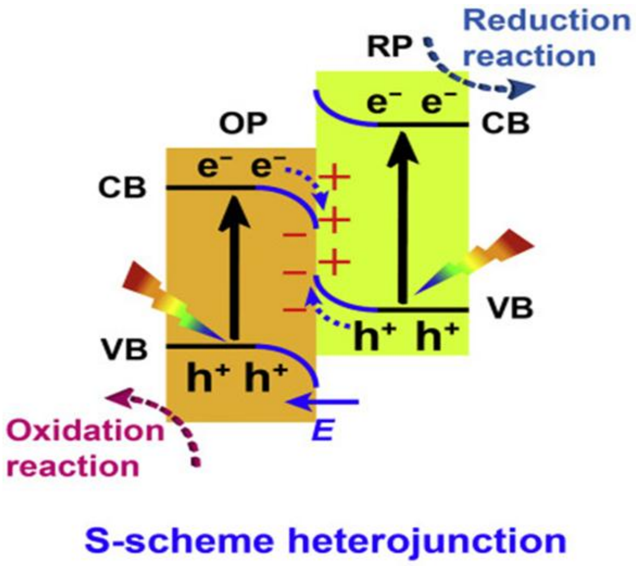


Fig. 10 shows the S-Scheme heterojunction [80].

6.4 p–n Heterojunction

Bi_2MoO_6, an n-type semiconductor that owns a super-thin band gap and an Aurivillius phase, has gained much attention for its applications in photocatalysis under visible light. Nonetheless, its photocatalytic process is sometimes hindered by the fast recombination of charge carriers. In that case, the combination of Bi_2MoO_6 and an n-type semiconductor (for example, TiO_2, g-C_3N_4, ZnO, or WO_3), which forms the p-n junction, can be very effective in improving the charge carrier separation and interfacial charge transfer. In this way, when Bi_2MoO_6 and the n-type semiconductor are closely contacted, Fermi level equilibration happens, and then band bending occurs at the junction, and an inward-built electric field in the n-type materials to the p-type materials is created [81,82]

Both semiconductors give rise to electron–hole pair generations when irradiated with visible light. Based on the direction of the potential difference, the transported electrons in the CB of Bi_2MoO_6 migrate towards the CB of the n-type semiconductor, and the holes in the VB of the n-type semiconductor migrate towards the VB of Bi_2MoO_6. Thus, charge transfer in both directions leads to the increased lifetime of charges and prevents recombination. Thus, due to band alignment and the electric field at the junction, the redox cycle at the catalyst surface increases. Here, the electrons at the catalyst surface reduce the adsorbed O_2 to •O_2– species, while the holes at the catalyst surface convert the organic pollutants or H_2O molecules to •OH radicals. Thus, Bi_2MoO_6-based p-n junctions demonstrate improved photocatalytic efficiency and increased utilization of visible light compared to individual semiconductors [83].

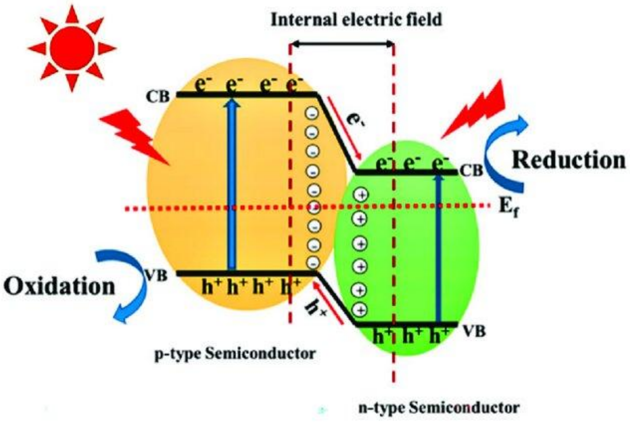


Fig. 11 shows the p–n heterojunction [83]

6.5 Z-Scheme Heterojunction

Bi_2MoO_6 has fast recombination rates of the photo-created charge carriers, making it less favourable for photocatalysis. In recent years, the Z-scheme type of heterojunction has been an effective approach in improving charge separation efficiency while retaining high redox potential. In the Z-scheme type of heterojunction, two semiconductors are combined based on well-designed band alignment such that the photogenerated electrons in the CB of the oxidation photocatalyst are made to recombine in the VB of the reduction photocatalyst, preferably via an internal solid phase (like MWCNTs, rGO, and precious materials), or directly. However, this characteristic charge transfer process adapts perfectly to the Z-scheme process in photosynthesis, keeping the highly energetic electrons in the CB of one semiconductor and the highly oxidative holes in the VB of another semiconductor. In this Z-scheme setup, better photocatalytic efficiency can be achieved compared to type-II counterparts, which normally decrease charge potential owing to interfacial carrier transfer [84,85].

Bi_2MoO_6-based Z-scheme photocatalytic systems, in combination with semiconducting materials like g-C_3N_4, WO_3, and TiO_2, demonstrate higher efficiency in charge separation, wider absorption ranges of visible light, and higher resistance to irradiation. In addition, taking advantage of the internal electric field and Fermi level matching at the heterojunction, directional charge transfer and prevention of electron-hole pair recombination are achieved. In addition, the strong oxidation and reduction abilities of separated charge carriers can promote the formation of reactive radicals such as •O_2^- and •OH, playing an important role in the photodegradation of persistent organic pollutants and the hydrogen evolution reaction. Further advancements of Bi_2MoO_6-based Z-scheme photocatalysts are expected to be on the horizon for the next-generation photocatalytic systems driven under visible light. Improvements in interface modulation, defects, and conductive bridges are expected to allow them to better contribute in terms of efficiency and stability for wide photocatalytic applications [86-88].

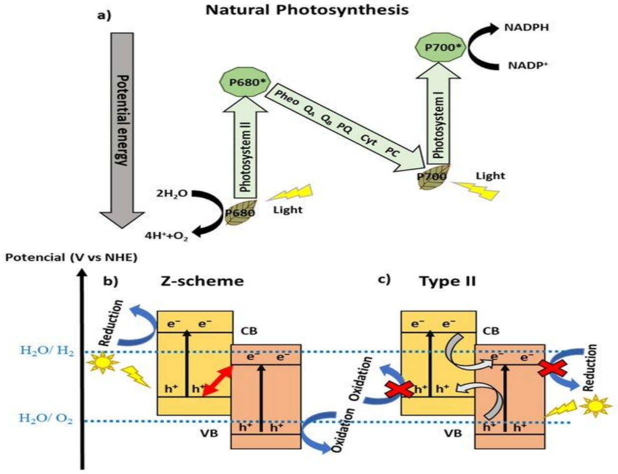


Fig. 12 shows the Z-Scheme heterojunction [89].

6.6. Schottky Heterojunction

In Schottky heterojunctions, the Schottky barrier at the metal-semiconductor contact originates from the equilibration of the Fermi levels in the two materials. Schottky barriers lead to band bending at the semiconductor/metal contact and cause charge spillover due to the potential difference between the two materials. Later, this charge spillover causes the generation of an internal electric field in the semiconductor, which further leads to superfast directional transport of charge carriers and reduces electron-hole pair recombination in the photocathode materials. In photocatalytic and photoelectrochemical systems, Schottky junctions are known to act as efficient channels for charge transfer between different materials and enhance the overall reaction rates in such systems. Ag, Au, and Pt are generally preferred in Schottky junctions due to their superior conductivity and possible localized surface plasmon resonance effects of the metal, which further increase light absorption in the photocathode materials, especially in the visible range of the electromagnetic spectrum. In addition to this, Schottky junctions allow for the use of semiconductors as an electron sink for collecting photogenerated charge in the photocathode materials, while holes are retained in the semiconducting materials [90-92].

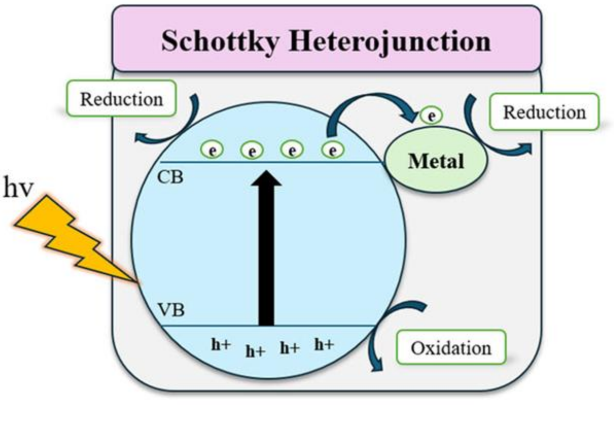


Fig. 13 shows the Schottky heterojunction [93].

7. Photocatalytic degradation

Photocatalytic degradation can be described as an advanced oxidation process that uses nanocomposites as a catalysts for the treatment and remediation of contaminants in contaminated sources of water. In this process, photocatalytic materials such as TiO_2, Bi_2S_3, and ZnO are utilized to produce electron-hole couples, which in turn act as catalysts in producing superoxide and hydroxyl radicals that easily decompose contaminants in the water into less harmful products. Based on current developments, the efficiency of this process can be improved using nanotechnology and photocatalytic couples, making photocatalysis an effective means for pursuing sustainable methods for the treatment of contaminated water sources [94,95].

8. Factors Influencing the Photocatalytic Reaction

Many important factors are involved in influencing photocatalytic efficiency, and absorption compared to the band gap of catalysts is of important concern. Care must be taken in optimizing catalyst loading in order to reduce light scattering. Solution pH and temperature conditions affect charge and adsorption properties of the catalyst, whereas temperature conditions influence charge pair recombination and reaction rates. Also, adsorption equilibrium and rates of reaction generally follow pseudo-first-order kinetics [96,97].

8.1 Effect of Dosage

The main factor that affects the efficiency of the photocatalytic process is the amount of catalyst used in the reaction. If the amount of catalyst used is low, the number of active sites and photon absorption will be limited, leading to decreased efficiency in the removal of pollutants. Increasing the amount of the catalyst will lead to increased absorption of photons and additional sites for the adsorption of pollutants. High amounts of catalyst, beyond the optimal amount, may lead to scattering of light and aggregation of particles in the photocatalyst, which may reduce the efficiency of the photocatalytic process [98,99].

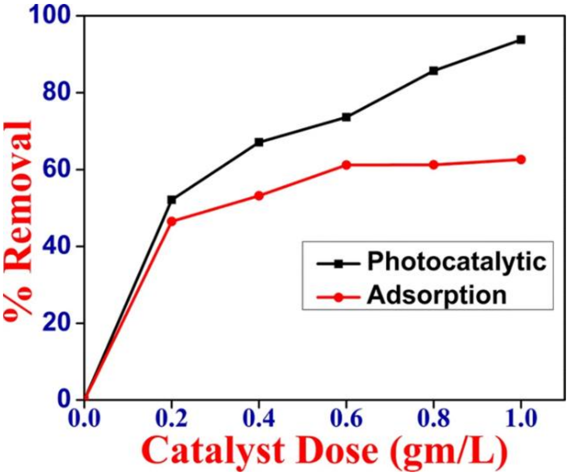


Fig. 14 Shows the Effect of Dosage [100].

8.2 Effect of Concentration

The amount of the contaminant in the solution has a large influence on the photocatalytic degradation reaction rate. In lower concentrations of contaminants, the availability of active sites on the catalyst surface compared to the amount of contaminant molecules in the solution makes it easier for adsorption to take place. However, as the contaminant concentration increases, the efficiency of contaminant degradation reduces due to lower amounts of reactive species and adsorption [101].

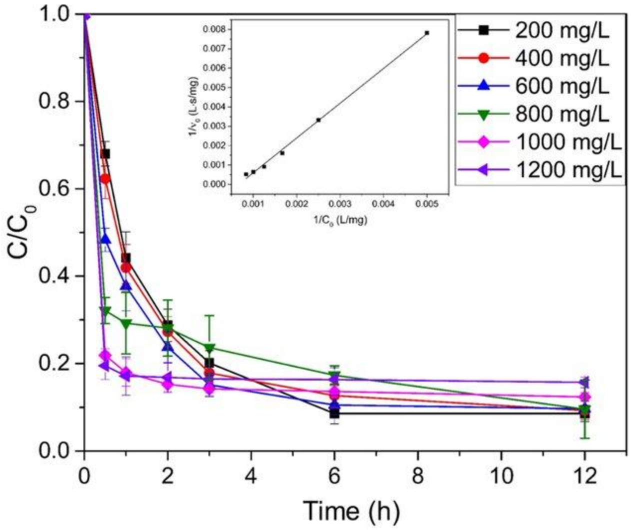


Fig. 15 Shows the Effect of Concentration [102].

8.3 Effect of pH

pH in the solution has been shown to greatly influence the photocatalytic degradation process due to the effects of pH on the charge of the photocatalytic materials and ionization of the molecules of pollutants. In acidic or basic environments, the potential difference between the catalyst and the pollutants changes, and this may act as an accelerator or inhibitor of the process of adsorption. In addition, pH conditions influence the role of •OH and O_2^- radicals in the photocatalytic process, as it increases or accelerates the generation of •OH radicals in the basic conditions [103,104].

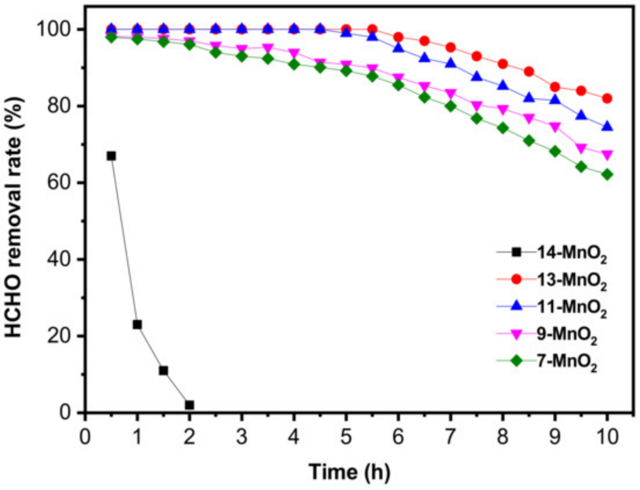


Fig. 16 shows the effect of pH [105].

8.4 Effect of Light

Intensity and wavelength of light are very important factors in determining photocatalytic efficiency, as they control the rate of excitation of electrons in the band gap of semiconducting materials. For the creation of electron-hole pairs for redox reactions, photons of energy or above the band gap energy are required. Higher light intensities increase charge carrier and radical formations until such a point that saturation is attained, beyond which recombination or heating may prevail. Further, the use of the same wavelength of light as that of absorption in the catalyst (UV or visible light range) optimises the use of the light sources for effective photocatalysis [106,107].

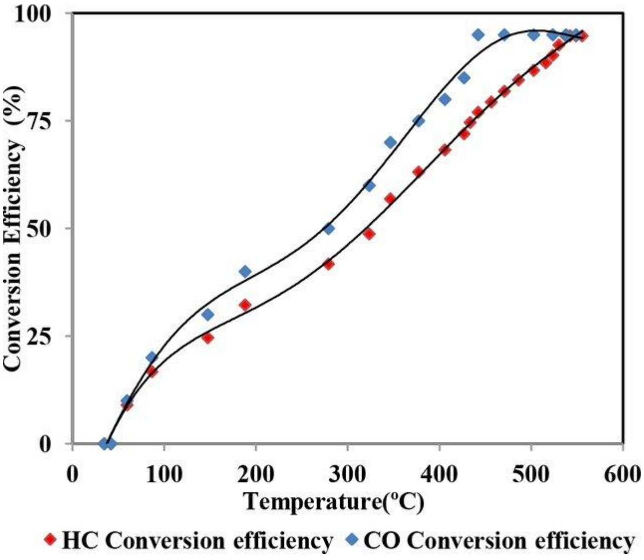


Fig. 17 shows the effect of light [108].

9. Conclusion

Results of this work clearly confirm that the strategic design and engineering of photocatalytic materials can promote the efficiency and reliability of the photodegradation process. Based on the combination of optimized nanostructures and band gap settings and improved charge transfer channels, higher efficiency and operation reliability of the synthesized photocatalytic materials were achieved during solar and visible light irradiation. In addition to practical efficiency and reliability, this work can contribute to the scientific understanding of light-induced redox reactions and may be important for the further development of next-generation photocatalysts. Further improvement of such developments may be vital for the advancement of clean water technologies and light-based environmental solutions.

Declaration of Competing Interest

The authors declare no conflicts of interest.

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This study didn't receive any specific funds.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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