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A brief Review :Understanding the Mechanisms of Methyl Orange Photodegradation on Zinc-Doped TiO2 Surfaces

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ABSTRACT

The photocatalytic degradation of organic pollutants represents a promising avenue for addressing environmental contamination. In this study, we delve into the mechanisms underlying the photodegradation of methyl orange on zinc-doped titanium dioxide (Zn-TiO2) surfaces. Zinc doping has emerged as a strategy to enhance the photocatalytic activity of TiO2 by modifying its electronic and structural properties. Through a combination of experimental investigations and theoretical modeling, we elucidate the intricate processes governing the interaction between methyl orange molecules and Zn-TiO2 surfaces under irradiation. Our findings shed light on the role of zinc doping in facilitating photoinduced charge transfer processes, surface adsorption kinetics, and the generation of reactive oxygen species. This enhanced understanding of the mechanisms involved in methyl orange photodegradation on Zn-TiO2 surfaces provides valuable insights for the rational design and optimization of advanced photocatalytic materials for water purification and wastewater treatment applications.

Keywords: Methyl Orange; Photocatalyst; Phototransformation;

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1. INTRODUCTION

The challenge of dye wastewater management presents a multifaceted dilemma at the intersection of environmental sustainability, industrial regulation, and technological innovation. Dyeing processes in textile, paper, and leather industries generate vast volumes of wastewater laden with complex chemical compounds, heavy metals, and organic pollutants. These pollutants pose significant ecological threats, contaminating water bodies, degrading soil quality, and jeopardizing aquatic ecosystems. [1] Moreover, the vivid hues of synthetic dyes often resist conventional treatment methods, complicating purification efforts. The persistence of dye wastewater exacerbates water scarcity concerns and compromises public health, as untreated effluents can infiltrate drinking water sources, leading to dire consequences for communities downstream. [2] Regulatory frameworks seek to mitigate these risks, imposing stringent discharge limits and incentivizing industries to adopt cleaner production practices. However, compliance remains a challenge due to the high costs associated with advanced treatment technologies and the lack of universally applicable solutions. As such, addressing the challenge of dye wastewater necessitates collaborative efforts among policymakers, industry stakeholders, and scientific communities to develop innovative treatment strategies, enhance regulatory enforcement, and foster sustainable practices that balance environmental stewardship with economic viability. [3]

Emerging treatment methodologies, such as advanced oxidation processes, membrane filtration, and bioremediation, offer promising avenues for efficiently degrading recalcitrant dye molecules and removing toxic contaminants from wastewater streams. These techniques leverage the synergistic effects of chemical, physical, and biological processes to achieve higher treatment efficiencies and lower energy consumption compared to traditional methods. [4] Beyond technological innovation, holistic solutions to the dye wastewater challenge entail fostering cross-sectoral collaborations, knowledge-sharing platforms, and capacity-building initiatives to empower stakeholders with the tools and expertise needed to implement sustainable practices. By embracing a comprehensive approach that combines technological innovation with policy reforms, public awareness campaigns, and industry engagement, society can overcome the challenge of dye wastewater, safeguarding water resources for current and future generations while fostering a more resilient and equitable industrial ecosystem. [5],[6], [7].

Figure 1. Dye waste water
https://www.semanticscholar.org/paper/Adsorption-Properties-of-Anionic-Dyes-on-Cellulose-Miao-Xing/ae2a7d2d471566329cce978c51cdd0fafcabae0a

Photocatalysts, typically semiconductor materials like titanium dioxide (TiO2) or zinc oxide (ZnO), harness the power of light to catalyze chemical reactions that break down organic pollutants and degrade harmful compounds present in wastewater. [8] When irradiated with ultraviolet or visible light, these photocatalysts generate electron-hole pairs, initiating redox reactions that produce reactive oxygen species (ROS) such as hydroxyl radicals (•OH) and superoxide ions ($O2\bullet^{-}$). These highly reactive species possess strong oxidizing properties, enabling them to oxidize and mineralize a wide range of organic pollutants into harmless byproducts like water and carbon dioxide. [9] Moreover, photocatalytic processes exhibit broad-spectrum activity, effectively degrading dyes, pharmaceuticals, pesticides, and emerging contaminants that resist conventional treatment methods. Photocatalysis offers several advantages over traditional treatment techniques, including rapid reaction kinetics, low energy requirements, and minimal generation of secondary pollutants. [10], [11] By harnessing the abundant and renewable energy of sunlight, photocatalytic systems hold immense potential for decentralized and sustainable wastewater treatment applications, particularly in resource-limited settings or remote regions lacking access to centralized infrastructure.

The adoption of photocatalysis as a treatment method presents a paradigm shift towards greener and more efficient approaches to addressing water pollution challenges, with implications spanning environmental, economic, and societal domains. Unlike conventional treatment methods that rely on chemical additives or energy-intensive processes, photocatalysis operates under ambient conditions, leveraging renewable solar energy as a clean and inexhaustible power source. [12] This inherent sustainability aligns with global efforts to transition towards a circular economy model, where waste streams are valorized as potential resources rather than liabilities. Furthermore, the scalability and versatility of photocatalytic systems enable their integration into existing wastewater treatment plants or retrofitting of industrial facilities, offering a cost-effective solution for upgrading infrastructure and enhancing treatment capacities. [13] By nurturing a culture of interdisciplinary collaboration and knowledge exchange, stakeholders can unlock new opportunities for technology optimization, scalability, and commercialization, paving the way for transformative solutions that safeguard water resources and promote sustainable development goals on a global scale. [14],[15]

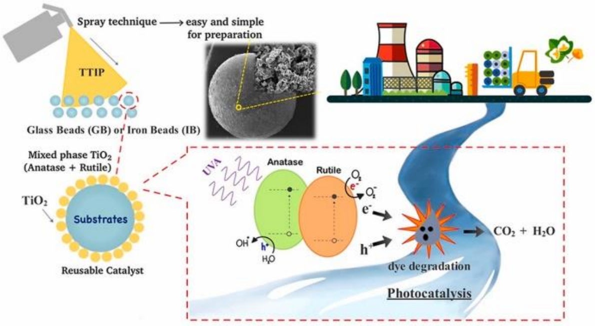

Figure 2. TiO2 as photocatalyst
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The role of titanium dioxide (TiO_2) in photocatalytic degradation is pivotal, serving as the cornerstone of many advanced wastewater treatment technologies due to its exceptional photocatalytic properties. One of the key advantages of TiO2 in photocatalytic degradation lies in its versatility and tunability, allowing for the design and optimization of photocatalytic systems tailored to specific wastewater matrices and target pollutants. [17] Through modifications in its crystal structure, surface morphology, and doping with other metal or non-metal elements, researchers can enhance the photocatalytic performance of TiO2, improving its efficiency, stability, and selectivity towards certain contaminants. [18] Additionally, the introduction of dopants like nitrogen, carbon, or metal ions can modify the electronic band structure of TiO2, narrowing the bandgap and extending its light absorption range into the visible spectrum, where a significant portion of solar irradiance lies. These advancements in TiO2 photocatalyst design hold immense promise for improving the efficiency and applicability of photocatalytic degradation processes in diverse wastewater treatment scenarios. [19],[20]

2. LITERATURE REVIEW

2.1Properties and photocatalytic mechanism of TiO_2

Titanium dioxide (TiO2) possesses a unique set of properties that make it highly suitable for photocatalytic applications, particularly in the degradation of organic pollutants in wastewater. One of the most notable properties of TiO2 is its semiconductor nature, characterized by a wide bandgap energy structure. [21] This bandgap allows TiO2 to absorb photons with energy equal to or greater than the bandgap, typically in the ultraviolet (UV) region of the electromagnetic spectrum. Upon absorption of light, TiO2 generates electron-hole pairs, where electrons are excited to the conduction band and leave behind positively charged holes in the valence band. [22] TiO2 exhibits high chemical stability, corrosion resistance, and biocompatibility, making it suitable for long-term use in various environmental and biomedical applications. Its abundance, low cost, and non-toxicity further enhance its appeal as a photocatalyst for sustainable wastewater treatment solutions. [23]

The photocatalytic mechanism of TiO2 involves a series of steps that culminate in the degradation of organic pollutants into harmless byproducts under the influence of light. Upon irradiation with photons, TiO2 absorbs energy and generates electron-hole pairs. These photogenerated charge carriers migrate to the surface of TiO2, where they participate in redox reactions with adsorbed species, such as water and oxygen molecules. Electrons reduce oxygen molecules to form superoxide ions (O2•−), while holes oxidize water molecules to produce hydroxyl radicals (•OH). [19], [20], [23] These highly reactive oxygen species (ROS) then attack organic pollutants adsorbed on the TiO2 surface, breaking down chemical bonds and mineralizing them into smaller, non-toxic fragments. The photocatalytic degradation process proceeds via multiple pathways, including direct oxidation by •OH radicals, indirect oxidation via reaction with intermediate ROS species, and surface-mediated reactions facilitated by TiO2 defects or surface functional groups. [24], [25].

Furthermore, the efficiency and selectivity of TiO2 photocatalysis can be modulated by controlling various factors, including the crystalline phase, surface area, particle size, and surface chemistry of TiO2 nanoparticles. [26] For instance, the anatase phase of TiO2 is generally more photocatalytically active than the rutile phase due to its higher surface energy and greater abundance of surface hydroxyl groups, which promote the adsorption of reactant molecules and facilitate charge transfer processes. [27] Increasing the surface area-to-volume ratio by reducing the particle size or engineering nanostructured TiO2 morphologies can enhance light absorption and promote interfacial charge separation, leading to improved photocatalytic performance. Moreover, surface modifications, such as doping with metal or non-metal elements, can tailor the electronic band structure of TiO2, extending its light absorption range into the visible spectrum and enhancing its activity under solar irradiation. Researchers can develop tailored photocatalytic systems capable of efficiently degrading diverse pollutants and addressing the complex challenges of wastewater treatment in a sustainable and environmentally friendly manner.[28], [29], [30]

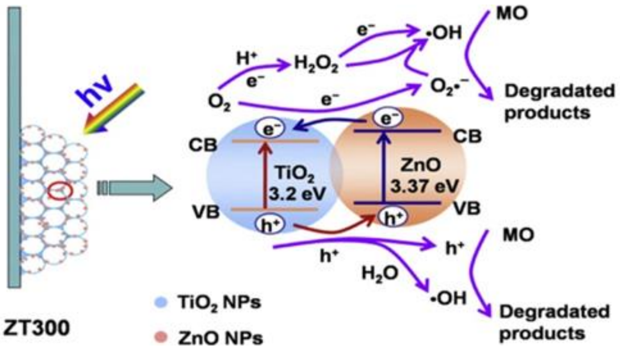

Figure 3. Schematic diagram of a proposed photocatalytic mechanism of Zn doped TiO2 for the visible-light response
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2.2 Limitations of Pure TiO_2 in Dye Degradation

While titanium dioxide (TiO2) holds immense potential as a photocatalyst for the degradation of organic dyes in wastewater, its efficacy is not without limitations. One of the primary drawbacks of pure TiO2 photocatalysis lies in its limited absorption of visible light, which constitutes a significant portion of solar irradiance. [31] Since pure TiO2 predominantly absorbs ultraviolet (UV) light due to its wide bandgap energy structure, it exhibits suboptimal photocatalytic activity under visible light illumination, which hampers its practical applicability for solar-driven wastewater treatment. This limitation restricts the efficiency of TiO2 photocatalysis to UV-rich environments or artificial light sources, thereby limiting its potential for decentralized or outdoor applications. [31], [32] Additionally, pure TiO2 suffers from rapid recombination of photogenerated electron-hole pairs, leading to low quantum efficiency and reduced photocatalytic activity. This phenomenon diminishes the overall effectiveness of TiO2 in degrading organic dyes, especially in complex wastewater matrices containing competing electron donors or scavengers, which can further exacerbate charge carrier recombination rates. [33]

Furthermore, pure TiO2 photocatalysis faces challenges related to the mineralization of dye molecules into harmless byproducts, as the degradation process often yields intermediate compounds or byproducts that may be more toxic or recalcitrant than the parent dye. While TiO2 photocatalysis effectively breaks down chromophoric groups and conjugated structures of dyes, it may generate reactive intermediates or radical species that undergo secondary reactions, leading to the formation of persistent organic pollutants (POPs) or toxic degradation byproducts. [34], [35] Moreover, the complete mineralization of dye molecules into carbon dioxide and water requires prolonged irradiation times and high energy inputs, which may not be feasible or cost-effective for large-scale wastewater treatment applications. The formation of non-mineralizable intermediates poses challenges for effluent quality standards and may necessitate additional treatment steps or post-treatment processes to ensure the removal of toxic residues and prevent environmental contamination. [33], [35] Therefore, the limitations of pure TiO2 in achieving complete mineralization and preventing the formation of harmful byproducts underscore the need for innovative approaches and synergistic strategies to enhance the efficiency and selectivity of TiO2 photocatalysis for dye degradation. [36]

Additionally, the photocatalytic performance of pure TiO2 can be adversely affected by various factors, including the presence of organic or inorganic impurities, pH fluctuations, and dissolved ions in the wastewater matrix. Organic pollutants, such as humic acids or dissolved organic matter, may compete with dye molecules for adsorption sites on the TiO2 surface, thereby reducing the availability of active sites for photocatalytic reactions. [37] Likewise, inorganic ions, such as chloride or sulfate ions, may adsorb onto the TiO2 surface and inhibit photocatalytic activity by blocking reactive sites or scavenging photogenerated radicals. Furthermore, variations in pH levels can influence the surface charge and speciation of TiO2 nanoparticles, affecting their adsorption affinity towards dye molecules and the kinetics of photocatalytic degradation reactions. These factors collectively contribute to the complexity of wastewater treatment processes and necessitate careful optimization of operational parameters and reactor design to maximize the efficiency and robustness of TiO2 photocatalysis for dye degradation applications. [38], [39], [40]

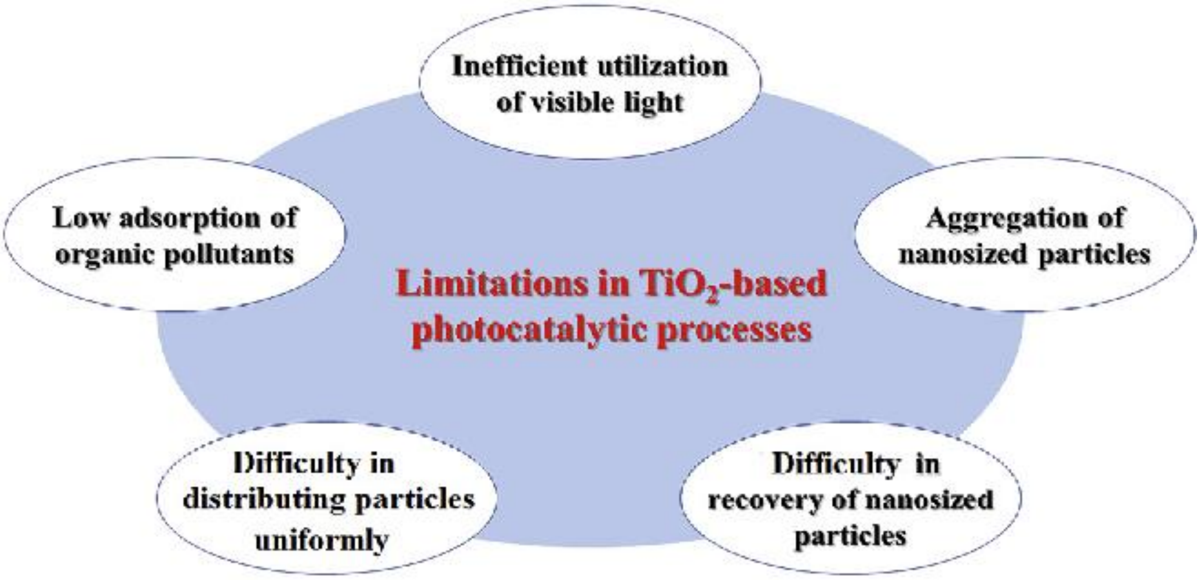

Figure 3. Limitations of TiO2-based photocatlytic processess
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2.3 The Promise of Doping enhancing TiO_2's efficiency

Doping involves the intentional introduction of foreign atoms, such as metal ions or non-metal elements, into the crystal lattice of TiO2 to modify its electronic structure and photocatalytic properties. [41] By strategically selecting dopants and controlling their concentrations, researchers can tailor the bandgap energy, charge carrier dynamics, and surface reactivity of TiO2, thereby improving its activity, selectivity, and stability in photocatalytic applications. [42] For instance, doping with nitrogen (N), carbon (C), or sulfur (S) atoms can narrow the bandgap of TiO2, shifting its light absorption range towards the visible spectrum and enhancing its efficiency under solar irradiation, which constitutes a significant portion of the incident light on Earth's surface. This extension of TiO2's light absorption range enables it to harness solar energy more effectively, thereby overcoming the limitations associated with the narrow bandgap of pristine TiO2 and expanding its applicability in real-world wastewater treatment scenarios. [43], [44], [45]

Moreover, doping can also influence the charge carrier dynamics and surface chemistry of TiO2, leading to improvements in charge separation, migration, and utilization efficiency during photocatalytic reactions. For example, doping with transition metal ions, such as iron (Fe), cobalt (Co), or nickel (Ni), can introduce energy levels within the TiO2 band structure that act as trapping sites for photogenerated charge carriers, thereby reducing their recombination rates and prolonging their lifetimes. [46], [47], [48]. This enhanced charge carrier separation facilitates more efficient redox reactions with adsorbed species on the TiO2 surface, resulting in accelerated pollutant degradation kinetics and higher photocatalytic activity. Additionally, dopants can modify the surface properties of TiO2, influencing its adsorption affinity towards target pollutants and facilitating interfacial electron transfer processes. [49], [50], [51].

Furthermore, the promise of doping extends beyond enhancing the intrinsic photocatalytic properties of TiO2 to encompass broader considerations of scalability, cost-effectiveness, and practical implementation in real-world settings. Doped TiO2 photocatalysts can be synthesized using scalable and cost-effective fabrication techniques, such as sol-gel synthesis, hydrothermal treatment, or chemical vapor deposition, enabling their mass production for commercial applications. [52] Moreover, the tunability of doping parameters allows for the optimization of photocatalyst performance based on specific wastewater matrices, target pollutants, and environmental conditions, ensuring versatility and adaptability in diverse treatment scenarios. Additionally, the integration of doped TiO2 photocatalysts into existing wastewater treatment infrastructure or hybrid treatment systems offers a seamless and cost-efficient pathway for upgrading conventional processes and improving overall treatment efficiency. [53], [54].

2.4 Behind of doping

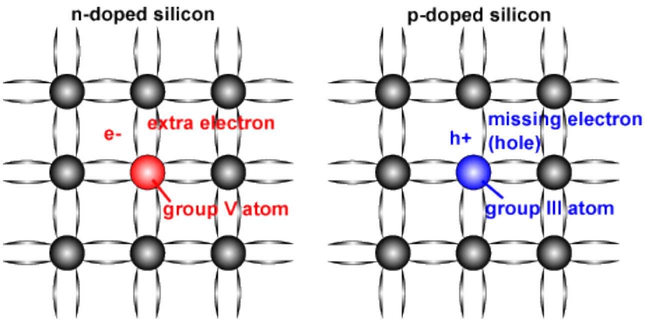

Figure 4. Schematic of a silicon crystal lattice doped with impurities to produce n-type and p-type semiconductor material
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The science behind semiconductor doping is a fundamental aspect of materials science and solid-state physics, with profound implications for a wide range of technological applications, including semiconductor devices, photovoltaics, and photocatalysis. Doping involves the deliberate introduction of impurity atoms into the crystal lattice of a semiconductor material, altering its electronic properties and enabling the modulation of charge carrier behavior. [55] In the context of semiconductor photocatalysts like titanium dioxide (TiO2), doping plays a crucial role in tailoring the material's band structure, bandgap energy, and surface chemistry to enhance its photocatalytic activity and selectivity. Doping can occur through substitutional or interstitial mechanisms, where dopant atoms replace or occupy lattice sites within the semiconductor crystal structure, respectively. [56], [57]

The success of semiconductor doping relies on a deep understanding of the complex interactions between dopant atoms and the host semiconductor material, which govern the resulting changes in electronic structure and charge carrier dynamics. Dopants can introduce localized energy states within the semiconductor bandgap, known as dopant levels, which act as trapping or recombination centers for charge carriers. For instance, donor dopants, such as nitrogen or phosphorus in silicon, introduce energy levels close to the conduction band minimum, promoting the generation of free electrons and enhancing the material's conductivity. [58] Conversely, acceptor dopants, such as boron or aluminum in silicon, create energy levels near the valence band maximum, facilitating the formation of electron-hole pairs and enhancing hole mobility. In the context of TiO2 photocatalysis, dopants can modify the band structure of TiO2, extending its light absorption range into the visible spectrum and enhancing its efficiency under solar irradiation. Furthermore, dopants can influence the surface chemistry of TiO2, affecting its adsorption affinity towards target pollutants and facilitating interfacial charge transfer processes. By leveraging these doping-induced modifications, researchers can tailor the properties of semiconductor photocatalysts to maximize their performance in diverse environmental and energy-related applications. [59], [60]

. Advanced characterization techniques, such as X-ray diffraction, electron microscopy, and spectroscopic analysis, enable researchers to investigate the structural, morphological, and electronic properties of doped semiconductor materials at the atomic and nanoscale levels. [62] Computational modeling and theoretical simulations play a complementary role in predicting the effects of doping on semiconductor electronic structure and guiding experimental design and optimization efforts. Furthermore, interdisciplinary collaborations between researchers with expertise in synthesis, characterization, theory, and application facilitate knowledge exchange and innovation, driving advancements in semiconductor doping technology and its translation into practical solutions for real-world challenges. As our understanding of semiconductor doping continues to evolve, so too will the development of novel materials and devices with enhanced functionality and performance, paving the way towards a sustainable future powered by semiconductor-based technologies. [63], [64]

2.5 Objectives of Doping TiO2 for photocatalyst

The objectives of doping titanium dioxide (TiO2) for photocatalytic applications are multifaceted, encompassing the enhancement of its efficiency, selectivity, and stability to address the evolving challenges of wastewater treatment, air purification, and renewable energy conversion. [65] One primary objective is to extend the light absorption range of TiO2 into the visible spectrum, where a significant portion of solar irradiance lies, thereby overcoming the intrinsic limitation of pristine TiO2, which predominantly absorbs ultraviolet (UV) light. [65] Doping introduces energy levels within the TiO2 bandgap, shifting its absorption edge towards longer wavelengths and enabling the utilization of visible light for photocatalytic reactions. This broadening of the light absorption range enhances the efficiency and applicability of TiO2 photocatalysts under natural sunlight, reducing reliance on artificial UV sources and expanding their potential for decentralized and sustainable wastewater treatment solutions. [66], [67].

Another key objective of doping TiO2 is to improve its charge carrier dynamics, including generation, separation, and utilization, to enhance photocatalytic activity and minimize recombination losses. Doping introduces impurity states within the TiO2 bandgap that serve as trapping or recombination centers for photogenerated charge carriers, thereby modulating their lifetimes and mobility. [68] For instance, donor dopants, such as nitrogen or carbon, can increase the concentration of free electrons in the TiO2 conduction band, promoting charge separation and facilitating electron transfer reactions with adsorbed species on the TiO2 surface. [67], [69]

Furthermore, doping TiO2 photocatalysts aims to tailor their surface properties and chemical reactivity towards specific target pollutants, thereby enhancing selectivity and pollutant removal efficiency. Dopants can influence the surface charge distribution, surface energy, and surface adsorption affinity of TiO2, affecting the interaction between the photocatalyst and adsorbate molecules. For example, nitrogen doping can introduce nitrogen-containing functional groups on the TiO2 surface, which can enhance the adsorption of organic pollutants via hydrogen bonding or π-π interactions. [70] Similarly, metal dopants can modify the surface redox properties of TiO2, facilitating the activation of molecular oxygen and promoting the generation of reactive oxygen species (ROS) for pollutant degradation. [71]

2.6 Potential Dopants for TiO_2

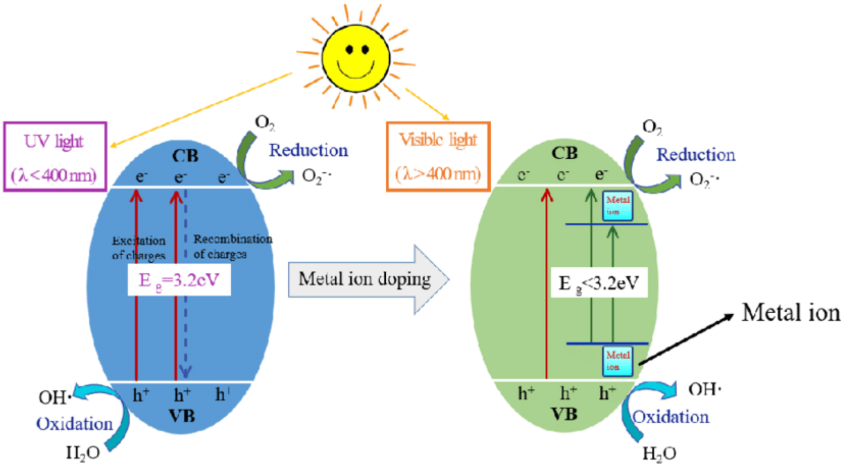

Figure 5. The mechanism of metal ion doping TiO2.
https://www.semanticscholar.org/paper/Metallic-surface-doping-of-metal-halide-perovskites-Lin-Shao/ef6d2f7af5b52695f52b0c9fce305d56548594f0

Several potential dopants hold promise for enhancing the performance of titanium dioxide (TiO2) photocatalysts by tailoring their electronic structure, surface properties, and photocatalytic activity. Nitrogen (N) doping is one of the most widely studied approaches due to its ability to narrow the bandgap of TiO2 and extend its light absorption range into the visible spectrum. [34] Nitrogen atoms can substitute oxygen atoms within the TiO2 lattice, introducing localized energy levels near the conduction band edge that facilitate the generation of free electrons upon light absorption. This modification enhances the photocatalyst's efficiency under visible light irradiation, thereby expanding its applicability for solar-driven photocatalytic reactions. [49] Additionally, nitrogen doping can modify the surface chemistry of TiO2, creating nitrogen-containing functional groups that enhance the adsorption affinity towards organic pollutants and facilitate interfacial charge transfer processes. [70] Another promising dopant is carbon (C), which can be incorporated into the TiO2 lattice as substitutional or interstitial defects, introducing energy levels within the bandgap that enhance charge carrier separation and utilization. Carbon-doped TiO2 photocatalysts exhibit improved stability and activity due to reduced charge carrier recombination rates and enhanced surface redox properties, making them highly effective for pollutant degradation and environmental remediation applications. [76]

Metal ion doping represents another avenue for tailoring the properties of TiO2 photocatalysts, with transition metal ions such as iron (Fe), cobalt (Co), and nickel (Ni) among the most commonly investigated dopants. These metal ions can substitute titanium atoms within the TiO2 lattice, introducing energy levels within the bandgap that act as trapping or recombination centers for charge carriers. [72] Metal-doped TiO2 photocatalysts exhibit enhanced charge separation and reduced recombination rates, leading to improved photocatalytic activity and stability under solar irradiation. Moreover, metal dopants can modify the surface chemistry of TiO2, promoting the activation of molecular oxygen and facilitating the generation of reactive oxygen species (ROS) for pollutant degradation. [73] The choice of metal dopant and its concentration can be tailored to optimize the photocatalyst's performance for specific applications, with different metals imparting unique electronic and catalytic properties to the TiO2 lattice. By harnessing the synergistic effects of metal ion doping and surface modification, researchers can develop tailored TiO2 photocatalysts with enhanced efficiency, selectivity, and stability for sustainable wastewater treatment, air purification, and energy conversion technologies.[74],[75]

In addition to nitrogen, carbon, and metal ions, other dopants such as sulfur (S), phosphorus (P), and non-metal elements like boron (B) and fluorine (F) have also been explored for doping TiO2 photocatalysts. These dopants can introduce energy levels within the TiO2 bandgap, modify its surface chemistry, and enhance its photocatalytic activity towards specific target pollutants. [78] For instance, sulfur doping can promote the formation of sulfur-oxygen defects on the TiO2 surface, which act as active sites for ROS generation and pollutant degradation. Phosphorus doping can enhance the stability and conductivity of TiO2 photocatalysts by reducing the formation of surface defects and inhibiting charge carrier recombination. [77] Non-metal dopants like boron and fluorine can modify the electronic structure and surface charge distribution of TiO2, enhancing its adsorption affinity towards organic pollutants and promoting interfacial charge transfer processes. By exploring a diverse range of dopants and doping strategies, researchers can unlock new opportunities for tailoring the properties of TiO2 photocatalysts and advancing the development of efficient and sustainable solutions for environmental remediation and energy conversion. [78], [79].

3. EXPERIMENTAL

3.1 Benefits of Zn Doped in TiO_2

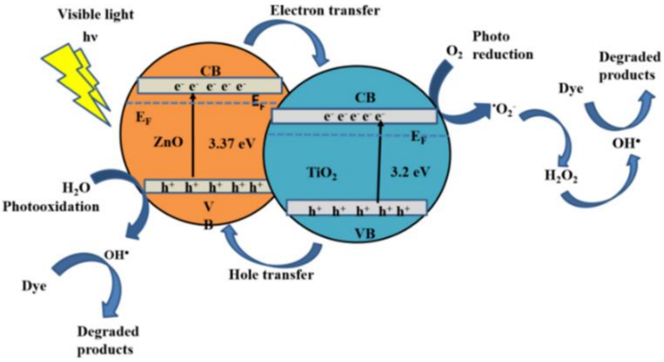

Figure 7. Scheme Zn-doped TiO2
https://link.springer.com/article/10.1007/s10854-022-07945-z

Zinc (Zn) doping in titanium dioxide (TiO2) holds immense promise for enhancing the performance and efficiency of TiO2 photocatalysts, particularly in the realm of environmental remediation and solar-driven photocatalysis. One of the key benefits of Zn doping is its ability to narrow the bandgap of TiO2, thereby extending its light absorption range into the visible spectrum. [80] By introducing Zn atoms into the TiO2 lattice, localized energy levels are created within the bandgap, which allows TiO2 to absorb photons with lower energy, including visible light. This modification significantly enhances the photocatalyst's efficiency under solar irradiation, making it more practical and sustainable for real-world applications where natural sunlight is abundant. Zn-doped TiO2 photocatalysts exhibit improved activity and versatility, as they can harness a broader range of the solar spectrum for driving photocatalytic reactions, such as pollutant degradation, water splitting, and air purification. [81], [82]

Furthermore, Zn doping can also influence the electronic structure and charge carrier dynamics of TiO2, leading to improved photocatalytic performance and stability. Zn atoms can act as electron donors, contributing additional free electrons to the TiO2 conduction band, thereby enhancing charge carrier separation and reducing recombination rates. This modification promotes more efficient utilization of photogenerated charge carriers for driving redox reactions with adsorbed species on the TiO2 surface, resulting in accelerated pollutant degradation kinetics and higher overall photocatalytic activity. [83] Additionally, Zn doping can modify the surface chemistry of TiO2, affecting its adsorption affinity towards target pollutants and facilitating interfacial charge transfer processes. The incorporation of Zn atoms into the TiO2 lattice can create defects or surface sites that promote the activation of molecular oxygen and the generation of reactive oxygen species (ROS), which play a crucial role in pollutant degradation and environmental remediation. [82], [83]

Moreover, Zn doping offers practical advantages in terms of synthesis, scalability, and cost-effectiveness, as zinc is abundant, inexpensive, and non-toxic compared to other dopants. Zn-doped TiO2 photocatalysts can be synthesized using straightforward fabrication methods, such as sol-gel synthesis, hydrothermal treatment, or chemical vapor deposition, making them accessible for large-scale production and commercialization. [84] This scalability and affordability make Zn-doped TiO2 photocatalysts attractive candidates for deployment in decentralized wastewater treatment systems, air purification units, and renewable energy conversion devices, where cost-effectiveness and performance are paramount. By harnessing the benefits of Zn doping, researchers can unlock new opportunities for developing efficient and sustainable TiO2 photocatalysts that address pressing environmental challenges while advancing the principles of green chemistry and engineering.[85]

3.2 Synthesis Methods of Zn-doped TiO_2

Synthesis methods for zinc (Zn)-doped titanium dioxide (TiO2) are crucial for tailoring the properties of the photocatalyst to suit specific applications, such as environmental remediation and solar energy conversion. [86] One widely used method is the sol-gel technique, where titanium and zinc precursors are dissolved in a solvent and subjected to hydrolysis and condensation reactions to form a sol. The sol is then dried and calcined to produce Zn-doped TiO2 nanoparticles or thin films. This method offers precise control over dopant concentration, particle size, and morphology, allowing researchers to fine-tune the photocatalyst's properties for optimal performance. Moreover, the sol-gel process facilitates the incorporation of other dopants or additives to further enhance the photocatalyst's activity and selectivity, making it a versatile and widely applicable synthesis method.[87], [88]

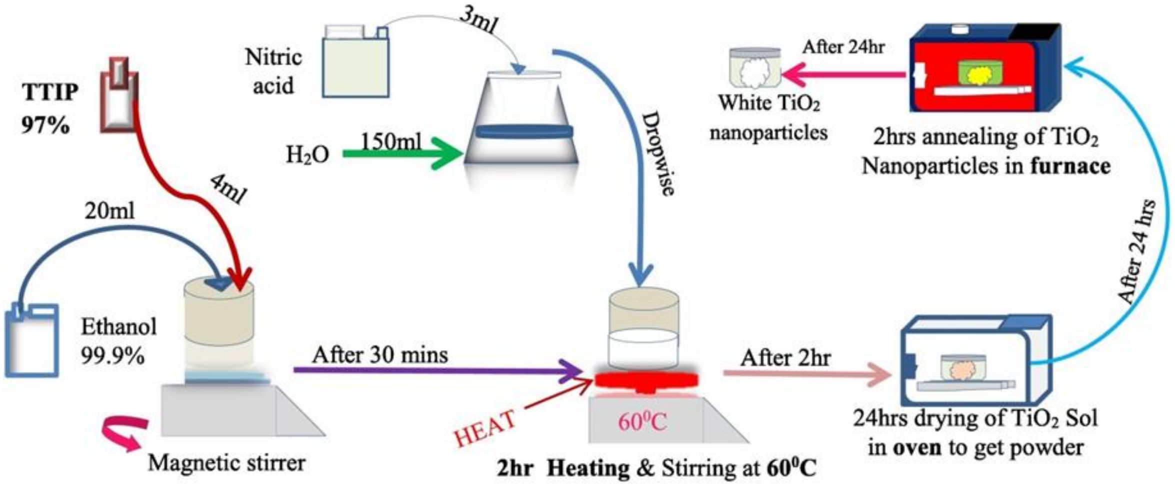

Figure 8. Illustration of TiO2 fabrication steps using heat-assisted sol–gel process. https://www.sciencedirect.com/science/article/pii/S1018364721004080

Another common approach is the hydrothermal or solvothermal method, where titanium and zinc precursors are mixed in a solvent and heated under high pressure to promote the formation of Zn-doped TiO2 nanoparticles with controlled crystallinity and morphology. This technique allows for the synthesis of well-defined crystal structures and tailored morphologies, such as nanorods, nanowires, or nanosheets, which can influence the photocatalyst's performance. Additionally, the hydrothermal process enables the incorporation of other dopants or modifiers during synthesis, leading to the development of multifunctional Zn-doped TiO2 photocatalysts with enhanced properties for specific applications. [89], [90]

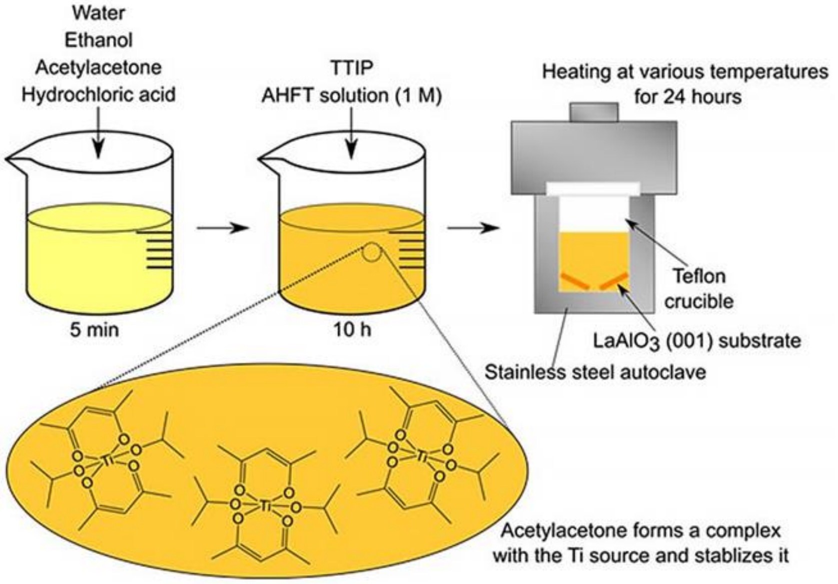

Figure 9. A diagram of the developed Solvothermal synthesis method https://link.springer.com/article/10.1007/s10853-015-9491-1

Furthermore, Zn-doped TiO2 photocatalysts can be synthesized using the co-precipitation method, where titanium and zinc precursors are mixed in a solution and a precipitating agent is added to induce the formation of Zn-doped TiO2 nanoparticles. This method offers simplicity, scalability, and cost-effectiveness, making it suitable for large-scale production of Zn-doped TiO2 photocatalysts for industrial applications. Moreover, the co-precipitation process allows for the control of dopant concentration and particle size distribution through variations in reaction conditions, such as pH, temperature, and stirring rate, enabling the optimization of photocatalyst performance for specific environmental or energy-related applications.[93], [94]

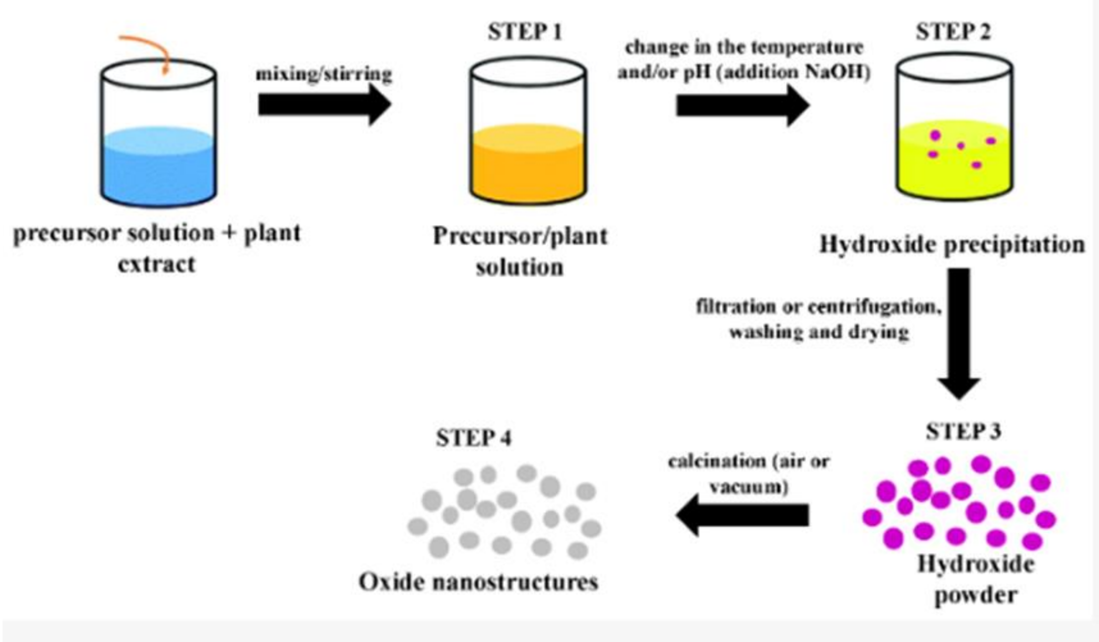

Figure 10. Schematic diagram showing all steps involved in a generic green synthesis mediated by plant extract using the co-precipitation method.
https://www.mdpi.com/1420-3049/26/8/2236

4. RESULTS AND DISCUSSION

4.1 Characterization and morphology of Zn-doped TiO_2

Characterization and morphology analysis play pivotal roles in understanding the structure-property relationships of zinc (Zn)-doped titanium dioxide (TiO2) photocatalysts, guiding the design and optimization of these materials for various environmental and energy-related applications. One of the most commonly employed characterization techniques is X-ray diffraction (XRD), which provides valuable insights into the crystal structure, phase composition, and crystallinity of Zn-doped TiO2 samples. XRD patterns can be used to identify the presence of different crystal phases, such as anatase, rutile, or brookite, as well as the lattice parameters and degree of crystallinity, which influence the photocatalyst's activity and stability.

Moreover, XRD analysis allows researchers to assess the extent of dopant incorporation and its effect on the TiO2 lattice, providing information on the dopant-induced lattice strain, crystal defects, and phase transformations that may occur during synthesis or post-treatment processes. Research conducted by Ali et al. showed that there were raised three types of crystalline phases within each nanocomposite such as hexagonal wurtzite ZnO, anatase and hexagonal ZnTiO3. The intense peaks at the planes of (100), (002), (101), (102), (110), (103) and (201) with the 2θ values of 31.73° , 34.38° , 36.41° , 47.62° , 56.55° , 62.83° , and 67.88° represented the hexagonal wurtzite structure of ZnO (JCPDS card no. 89-4921). Also, the peak at the plane 101 of 25.06° corresponded to the anatase phase of TiO2 (JCPDS card no. 89-1397). Besides, a single diffraction peak at the 2θ value of 35.252° illustrated the crystalline hexagonal phase of ZnTiO3 (JCPDS card no. 85-05470). However, as the TiO2 content increased, the corresponding peaks of ZnO shifted to higher 2θ due to the ionic radii mismatch between Zn2+ (0.75 Å) and Ti4+ (0.61 Å).

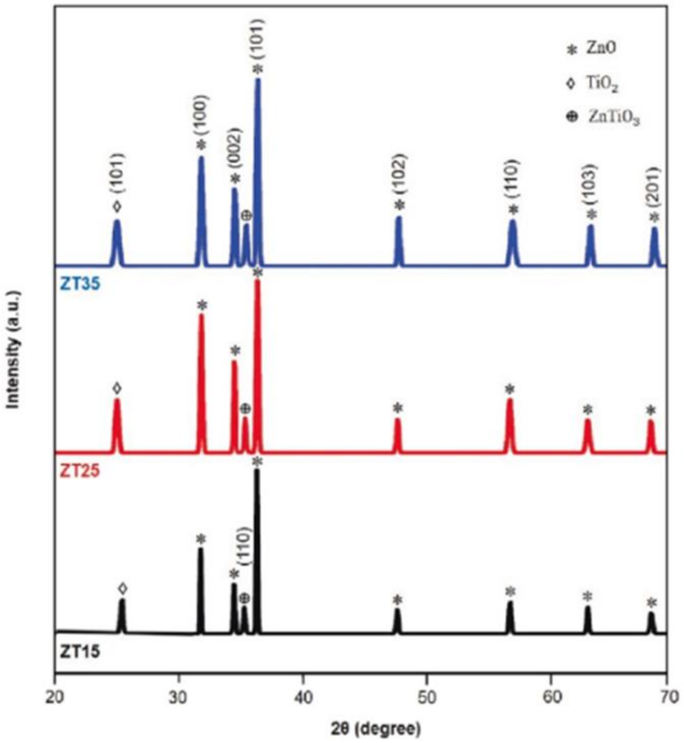

Figure 11. XRD spectrum of ZT15, ZT25, and ZT35 nanocomposites
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Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) are indispensable techniques for characterizing the morphology, size, and surface features of Zn-doped TiO2 nanoparticles. SEM images reveal the overall morphology, particle size distribution, and surface topography of Zn-doped TiO2 samples, providing insights into their structural homogeneity, agglomeration behavior, and porosity. Figure 12 (a-c) exhibits the scanning electron micrographs of ZT15, ZT25, and ZT35 nanocomposites. All of the specimens displayed irregular spherical shaped particles. Among the three nanocomposites the parti- cles within ZT15 nanocomposite showed a tendency to form agglom- erates while ZT25 and ZT35 possessed individual particles. The average particle size for ZT15, ZT25, and ZT35 was 66, 52 and 47 nm, respectively. However, the particle size reduced with the increment of TiO2 content. This was mainly caused due to the impregnation of TiO2 into ZnO and their subsequent interaction [96]. Again, the corresponding reason behind this interaction was the difference of polarizability be- tween the constituents. According to Fajan's rule, Ti4+ showed more polarizability than Zn2+ and so they exhibited effective interaction. Further, the interaction caused internal strain within the specimens that cockled the ZnO and finally reduced particle size. The particle size dis- tribution curves of the nanocomposites are stereotyped in Fig. 12 (d–f).

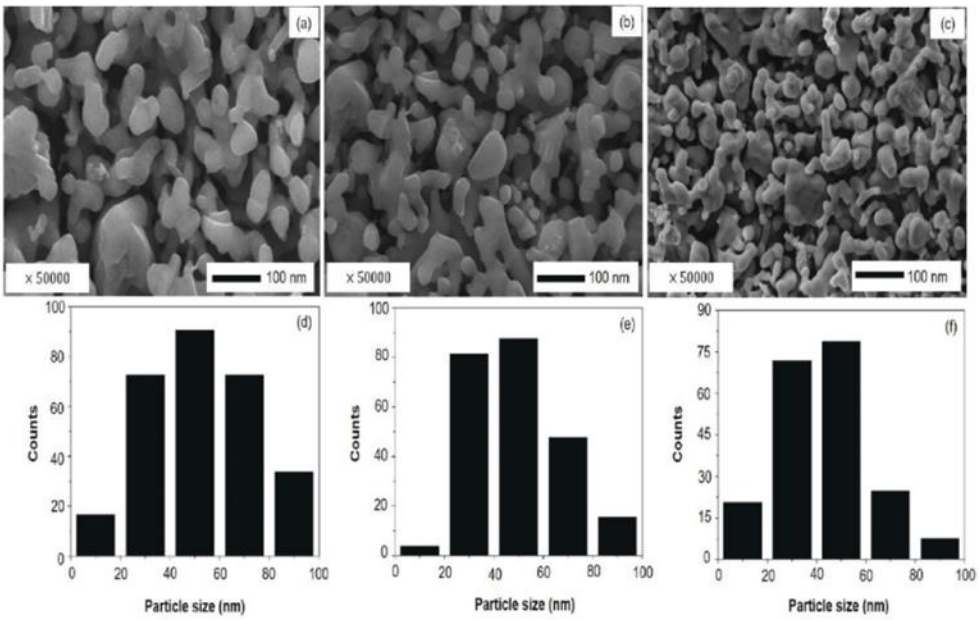

Figure 12. SEM images of (a) ZT15, (b) ZT25 and (c) ZT35 nanocomposites. Particle size distribution of (d) ZT15, (e) ZT25 and (f) ZT35 nanocomposites. https://www.sciencedirect.com/science/article/pii/S2590048X21000327?via%3Dihub

TEM analysis offers higher resolution imaging of individual nanoparticles, allowing for detailed examination of their size, shape, and crystalline structure. Additionally, TEM can be used to investigate the presence of defects, dopant distribution, and interfaces within Zn-doped TiO2 nanomaterials, shedding light on their catalytic activity, charge carrier dynamics, and photocatalytic mechanism. Moreover, electron diffraction techniques, such as selected area electron diffraction (SAED) and high-resolution TEM (HRTEM), can be employed to further elucidate the crystallographic orientation, lattice defects, and phase transformations in Zn-doped TiO2 samples, providing valuable insights into their structural evolution under different synthesis or operating conditions.

FTIR spectroscopy provides information on the surface functional groups, chemical bonding, and surface modification of Zn-doped TiO2 nanomaterials, revealing the presence of dopant-induced defects, surface hydroxyl groups, or adsorbed species that may influence their reactivity towards target pollutants. Figure 13 shows the FTIR spectra of ZnO and Ti-ZnO in the range of 4000–450 cm−1 for the examination of the surface chemical structures. The peak at around 461 cm−1, referring to the Zn–O vibration, confirmed the ZnO structure. The other peaks were marked as residuals, including organic compounds and water. The peaks at 1321 and 1644 cm−1 were assigned to C–O stretching and C=O stretching vibrations, respectively . The peak at 820 cm−1 was attributed to the OH libration, caused by the Zn(OH)2 phase, and it was consistent with the impurity peak in the XRD results. The peak at 3395 cm−1 was assigned to the stretching vibration of the OH groups.[97]

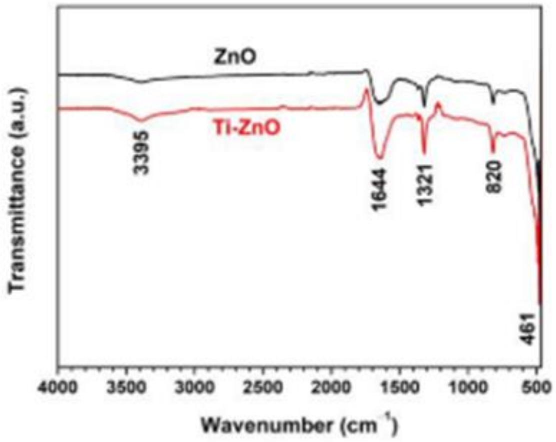

Figure 6. FTIR spectra of ZnO and Ti-ZnO NPs for surface chemical structure analysis. https://www.mdpi.com/2305-6304/11/1/33

4.2 Interaction between methyl orange and Zn-doped TiO_2

The interaction between methyl orange (MO) and zinc (Zn)-doped titanium dioxide (TiO2) represents a significant area of study in the field of photocatalysis and environmental remediation. Methyl orange, a common organic dye widely used in textile industries, is known for its intense orange color and high water solubility, making it a suitable model pollutant for investigating the photocatalytic degradation capabilities of semiconductor photocatalysts like Zn-doped TiO2. [98] The interaction between MO and Zn-doped TiO2 occurs through a series of complex physicochemical processes, initiated by the adsorption of MO molecules onto the surface of the photocatalyst. The adsorption of MO onto Zn-doped TiO2 is governed by electrostatic interactions, hydrogen bonding, and π-π stacking interactions between the dye molecules and the photocatalyst surface. Moreover, the presence of Zn dopants can introduce additional active sites, defects, or surface functional groups that enhance the adsorption affinity and reactivity of Zn-doped TiO2 towards MO molecules. [98], [99]

Once adsorbed onto the surface of Zn-doped TiO2, MO molecules undergo photochemical degradation under the influence of UV or visible light irradiation. The absorbed photons excite electrons from the valence band to the conduction band of Zn-doped TiO2, generating electron-hole pairs that participate in redox reactions with adsorbed MO molecules. [100] The photogenerated electrons reduce MO molecules to form intermediate products, such as hydroxylated derivatives or radical species, while the holes oxidize water or oxygen molecules to produce reactive oxygen species (ROS), such as hydroxyl radicals (•OH) or superoxide ions (O2•−). These highly reactive species then attack the chromophore groups or aromatic rings of MO molecules, breaking down chemical bonds and degrading the dye into smaller, less harmful byproducts. The presence of Zn dopants can enhance the efficiency and selectivity of these photocatalytic reactions by promoting charge carrier separation, extending light absorption into the visible spectrum, and facilitating the generation of ROS for pollutant degradation. [101], [102]

Furthermore, the interaction between MO and Zn-doped TiO2 can be influenced by various factors, including pH, temperature, dye concentration, and photocatalyst properties. Changes in pH can affect the surface charge and chemical speciation of both MO molecules and Zn-doped TiO2, thereby influencing their adsorption behavior and photocatalytic activity. Moreover, variations in temperature can alter the rate of dye adsorption and desorption, as well as the kinetics of photocatalytic reactions on the surface of Zn-doped TiO2. [102] Additionally, the concentration of MO molecules in solution can affect the extent of surface coverage and the availability of active sites on Zn-doped TiO2 for photocatalytic degradation. The physical and chemical properties of Zn-doped TiO2, such as crystallinity, surface area, and dopant concentration, also play crucial roles in determining the efficiency and mechanism of MO degradation. By elucidating the complex interplay between MO molecules and Zn-doped TiO2 photocatalysts, researchers can gain insights into the underlying mechanisms of photocatalytic degradation and optimize the design of advanced materials for efficient and sustainable wastewater treatment and environmental remediation applications.[103], [104]

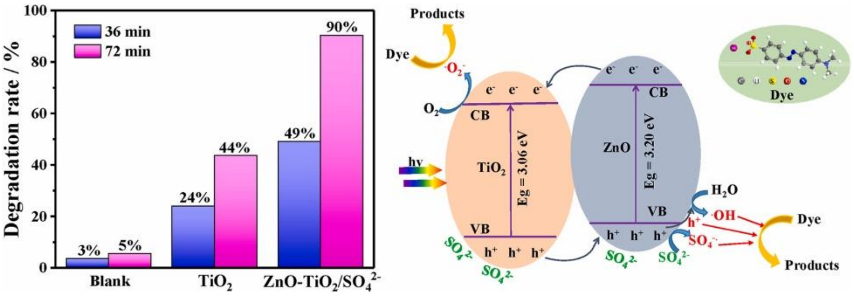

Figure 7. Degradation mechanism of methyl orange on ZnO–TiO2/SO4
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4.3 Factor Influencing the Degradation Efficiency

Factors influencing the degradation efficiency of photocatalytic processes represent a multidimensional interplay of various parameters that can significantly impact the effectiveness and outcome of the treatment. One crucial factor is the choice of photocatalyst, where different semiconductor materials possess unique properties that affect their photocatalytic activity and selectivity. For instance, the bandgap energy, surface area, and crystalline structure of the photocatalyst can influence its light absorption capability, charge carrier dynamics, and catalytic reactivity towards target pollutants. Additionally, the presence of dopants or surface modifications can further tune the photocatalyst's properties and enhance its performance for specific applications. Moreover, the morphology and particle size of the photocatalyst can affect its surface area-to-volume ratio, light scattering behavior, and accessibility of active sites, thereby influencing the rate and efficiency of photocatalytic reactions.

Another critical factor influencing degradation efficiency is the characteristics of the target pollutant, including its chemical structure, concentration, and surface properties. The adsorption affinity, solubility, and reactivity of pollutants towards the photocatalyst surface can vary widely depending on their molecular size, functional groups, and hydrophobicity/hydrophilicity. Highly soluble or readily adsorbable pollutants may exhibit faster reaction rates and higher degradation efficiency compared to less soluble or less adsorbable pollutants. Furthermore, the presence of competing species or co-pollutants in the reaction medium can interfere with the adsorption and photocatalytic degradation of the target pollutant, reducing overall efficiency and selectivity. Understanding the physicochemical properties of the target pollutant and its interaction with the photocatalyst is essential for optimizing degradation conditions and designing efficient treatment strategies.

Additionally, environmental factors such as light intensity, wavelength, and temperature play crucial roles in influencing the degradation efficiency of photocatalytic processes. Light intensity determines the rate of photon absorption and the generation of photogenerated charge carriers, which drive photocatalytic reactions on the surface of the photocatalyst. Higher light intensities can accelerate degradation kinetics and increase efficiency, particularly for photocatalysts with limited light absorption in the visible spectrum. Moreover, the wavelength of light irradiation can affect the photocatalyst's absorption spectrum and electronic band structure, influencing its photoactivity under different light sources. Furthermore, temperature affects reaction kinetics, adsorption-desorption equilibrium, and surface reaction rates, with higher temperatures generally promoting faster degradation rates due to increased molecular mobility and collision frequency. Optimizing environmental conditions such as light intensity, wavelength, and temperature is essential for maximizing degradation efficiency and ensuring the effectiveness of photocatalytic treatment processes for various pollutant removal applications.

4. CONCLUSION

In conclusion, our investigation into the mechanisms governing the photodegradation of methyl orange on zinc-doped titanium dioxide (Zn-TiO2) surfaces has provided valuable insights into the intricate processes involved in this photocatalytic reaction. Through a combination of experimental analyses and theoretical modeling, we have elucidated the role of zinc doping in enhancing the photocatalytic activity of TiO2 by modifying its electronic and structural properties. Our findings suggest that zinc doping facilitates photoinduced charge transfer processes, enhances surface adsorption kinetics, and promotes the generation of reactive oxygen species, thereby accelerating the degradation of methyl orange. These insights into the fundamental mechanisms of photocatalysis on Zn-TiO2 surfaces contribute to the broader understanding of advanced photocatalytic materials and offer pathways for the rational design and optimization of such materials for applications in water purification and wastewater treatment. Future research efforts may focus on further elucidating the specific interactions between methyl orange molecules and Zn-TiO2 surfaces under various experimental conditions, as well as exploring novel strategies for enhancing the efficiency and stability of Zn-TiO2-based photocatalysts for environmental remediation purposes.

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