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REVIEW PAPER

The application of ZnO nanoparticles as a photocatalyst for wastewater treatment: A review

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ABSTRACT

Ever-increasing environmental pollutions and water scarcity are highly challenging issues that pose formidable obstacles to human beings on all fronts. Hetero-photocatalytic methods which utilized semiconductors as photocatalysts are highly promising and green technologies for the degradation of recalcitrant organic pollutants which cannot be completely removed by conventional treatment processes. In the view of the current scenario, zinc oxide nanostructures have been demonstrated to be predominant photocatalyst candidates for photodegradation because of their cost-effectiveness, non-toxicity, strong oxidation capability, flexibility in synthesizing, earth-abundance nature, easy crystallization, and high performance in the absorption over an extensive fraction of solar spectrum in comparison with titanium dioxide. Nevertheless, bare zinc oxide possesses several intrinsic limitations, like high recombination rate of the photogenerated charge carriers, limited solar light application, photo corrosion, broad bandgap and limited visible light absorption. Moreover, photocatalysts separation from remediated solutions restricts their large-scale applications. In this review paper, the authors briefly discussed basic principles of the zinc oxide photocatalytic process besides various modifications such as coupling with low bandgap semiconductors like metal and non-metal doping, synthesizing with graphene oxide, or reduced graphene oxide and their integration in magnetic materials to successfully addressing aforesaid disconcerting challenges. Moreover, hybridized photocatalytic and membrane systems are explored. Finally, challenges and future research directions are proposed for giving profound and well-defined insights toward reaching fully exploited zinc oxide-based nanoparticles in the field of water and wastewater treatments.

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INTRODUCTION

Recently, water reuse and recovery have been garnering special attention globally because of the alarming water shortage owing to the climatic variation and inefficient control of water sources such as restricted access to drinkable water sources and imbalance between water supply and demand. It goes without saying that with the burgeoning of economic advancements and population growth availability of clear water becomes challenging [1]. Hence, to ensure the sustainability of water resources, the development of wastewater treatment methods and water reuse would be a highly promising approach. Nevertheless, treatment of recalcitrant organic contaminants still is challenging. Recalcitrant organic contaminants are carbon-dependent chemical compounds that are bio-resistant and have been constantly discharged into the environment. Due to their both

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carcinogenic and bio-resistant nature, they can pose serious adverse effects to human beings. As a result, superior treatment methods are essential to making sure their efficient removal from water sources. In this context, different water remediation approaches have been constantly developed and applied to eliminate recalcitrant organic pollutants from water bodies like coagulation, membrane separation, and adsorption [2]. But such technologies are phase change methods in that changing them from water to solid phase. Thus, further cost and remediation processes are required for the remediation of the secondary contaminants and reproduction of the adsorbents [3]. As a result, advanced oxidation processes (AOPs) have been emerged for the removal of persistent organic contaminants, especially for treating bio-resistant ones. Based on Capelo et al. [4], these processes are operated through in situ productions of extremely powerful oxidants like hydroxyl radicals ($OH^{\cdot}$) by application of Fenton's reagent, hydrogen peroxide (H_2O_2), ozone (O_3), different types of catalyst and UV light, hence result in oxidization of refractory organic matters. Reducing the toxicity of organic materials, operability under ambiance pressure and temperature, mineralizing organic materials to eco-friendly products, and quick decomposition are major benefits of AOPs.

The photocatalytic process as one branch of AOPs can be divided into two broad categories of the homogeneous or heterogeneous photocatalyst. Homogeneous photocatalysis utilizes Fenton's reagent, which integrated H_2O_2 and a ferrous ion for the generation of $OH^{\cdot}$ under ultra-violet radiation at wavelengths higher than 300 nm [5]. On the other hand, heterogeneous photocatalyst uses semiconductor oxides as a photocatalyst [6]. Among different semiconductors, titanium dioxide (TiO_2) has been the most explored semiconductor [7–14]. Because of its desirable chemical stability and low-cost generation, it has been broadly utilized in the photo-degradation of organic materials, like pesticides [15], volatile organic compounds [16], non-biodegradable azo dyes [17], aromatic hydrocarbons [18], saturated hydrocarbons (alkanes) [19] and those with a significant loading of nitrogen-comprising organic compounds [20] using ultra-violet light. As stated by Wang et al. [21], the utilization of titanium dioxide with solar power is extremely limited because of its low quantum performance and broad bandgap (3.2 eV).

Numerous efforts like noble and non-noble metal deposition [22,23], dye sensitization [24], surface modification, generation of nanocomposites [25], and doping [26] have been carried out for increasing titanium dioxide photocatalytic activity and photo response in the visible light area. In comparison with titanium dioxide, zinc oxide (ZnO) is an n-type semiconductor oxide although has not been fully explored [27–31]. Zinc oxide has been suggested as a substitute photocatalyst to titanium dioxide because it has similar bandgap energy however at the same time demonstrates greater absorption performance over an extensive portion of the solar spectrum compared with titanium dioxide [32]. To assess the photosensitization of zinc oxide and titanium dioxide, the photo-degradation of fungicides in the leaching water utilizing zinc oxide and titanium dioxide under solar radiation was explored by Fenoll et al. [33] and discovered nonstoichiometric of zinc oxide making it a favorable photocatalyst in comparison with titanium dioxide.

Even though AOPs have been demonstrated to be highly efficient in the reduction of refractory organic compounds, still there is a long way to fully exploit their potential practically. First and foremost, the cost associated with AOPs is an essential benchmark for the determination of the viability of the process in water remediation systems. The feasibility of high operating costs which may incur because of marked electricity consumption and cost-prohibitive chemicals can remarkably confine the viability of AOPS in water remediation units. Also, the generation of $OH^{\cdot}$ and ions regeneration significantly determines the extent of the degradation/oxidation reaction [34–38]. Without enough generation of $OH^{\cdot}$, the performance of AOPs in the reduction of persistent organic contaminants is reduced. Extensive works have been performed for decades to improve the production degree of $OH^{\cdot}$ to advance AOP remediation effectiveness [39,40]. In addition, there is a conspicuous demand for realizing the toxicity of the intermediates and mineralizing products driven by AOP. Nicotine elimination using Fenton's reagent was applied by Rodriguez et al. [41] to explore the degradation and toxicity of intermediates produced in AOPs. They reported unfinished degradation although all of the nicotine was eliminated and the toxicity of the resultant intermediates (in particular those at the primary level of the oxidation) was 15 times higher than the initial solution.

Because of advancements in the application of the photocatalytic process for degrading persistent organic contaminants, it has been expanded for photocatalytic membrane reactors in a wide range and industrial-scale applications. In heterogeneous photocatalysis, applied semiconductor oxides are generally immobilized on support or suspended in the remediated effluent (slurry-type reactors). The major disadvantage of slurry-type reactors is needing a further process for recovering photocatalyst, which does not occur in the case of an immobilized-type reactor. The combination of photocatalytic and membrane processes is a promising technology that ensures nonstop operation besides excellent photocatalyst recovery [42].

Providing a favorable platform for contaminants adsorption and desorption during the reaction, the better mass transformation of contaminants from bulk liquid phase into photocatalysts active surface sites, and lower pressure drop over the reactor are some of the advantages of photocatalysts suspended in the aqueous media [43]. In order to offset the cost confinements associated with photocatalyst recovery, heterogeneous photocatalysis which uses solar radiation has been recommended as a low-cost process that can be performed without the requirement for an unnatural and expensive radiation source.

Fabrication and conduction of photocatalyst have been absorbing special attention because of the remarkable benefits of the photocatalytic process. Gaining in-depth and full-scale knowledge concerning the characteristics and properties of the zinc oxide photocatalyst can equip researchers with various perceptions to closely scrutinize the unexplored dimensions of the ZnO-driven photocatalytic process which can lead to flourishing large scale application of zinc oxide photocatalysts, especially when hybridized with other remediation technologies.

PRINCIPLES AND MECHANISM OF ZINC OXIDE PHOTOCATALYSIS

Extensive borderline UV adsorption at room temperature, significant excitation binding energy (60 meV), and wide bandgap (3.37 eV) are some of the main properties of zinc oxide (ZnO) as a semiconductor [44]. Same as titanium dioxide, because of the outstanding optical, mechanical, and electrical characteristics of ZnO it is considered a superior semiconductor [45,46]. Moreover, as a great advantage, it possesses desirable photocatalytic activity, and antibacterial and antifouling features [47]. In comparison with Al_2O_3 and TiO_2, Liang et al. [48] stated that overall costs associated with the generation of zinc oxide are up to 75% lower. Hence, because of the superior benefits of zinc oxide over titanium dioxide, it has been recommended to be utilized in the hetero-photocatalytic process. The hetero-photocatalysis oxidation processes of ZnO include (a) Diffusion of organic contaminants from the liquid phase to the surface of zinc oxide, (b) Surface adsorption of the organic contaminants on the zinc oxide, (c) Oxidation and reduction reactions in the adsorbed phase, (d) Desorption of the products and (e) Elimination of the products from the interface area (see Fig.1) [49].

When zinc oxide is photogenerated using solar light, electrons from the valence band (VB) are transferred to the conduction band (CB) as a result as demonstrated in Eq.1 electron-hole pairs are generated. As demonstrated in Eqs.2-4, they can

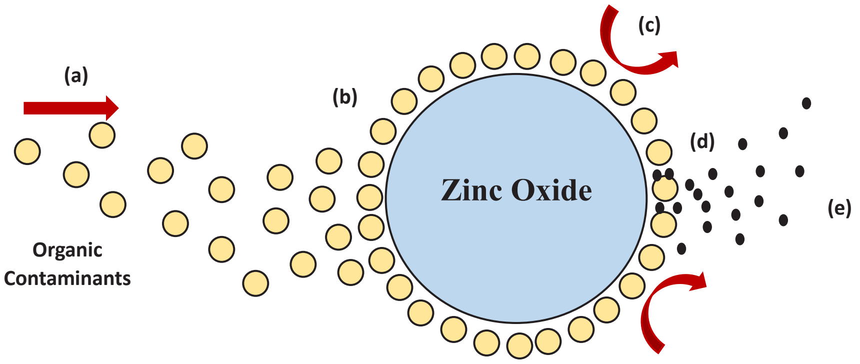

Fig.1. Heterogeneous photocatalytic oxidation steps.

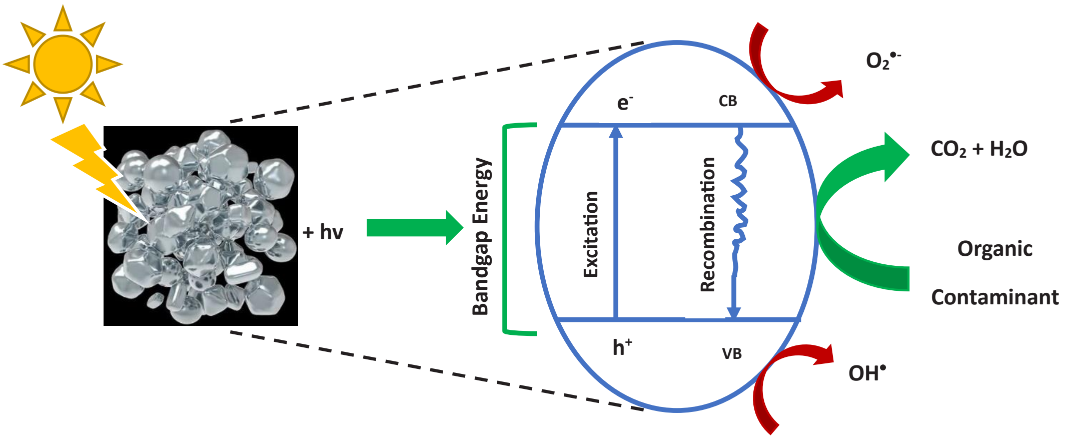

Fig.2. Degrading organic contaminants using zinc oxide and solar light.

move to the zinc oxide surface for contributing to redox reactions, in which electron reacts with O_2 to generate superoxide radical anions then H_2O_2 and the H^+ react with H_2O and hydroxide ions to generate $OH^{\cdot}$ (Eq.5). After that H_2O_2 will react with superoxide radicals to generate $OH^{\cdot}$ (Eqs.7-9). Following that, $OH^{\cdot}$ as potent and non-selective reagents, attacking the contaminants adsorbed on the surface of zinc oxide for the production of intermediate constituents, and finally, they converted to harmless matters like mineral acids, and carbon dioxide, and water (Eq.11). The redox reaction happening during photocatalysis was demonstrated in Fig.2 [50,51].

$$ZnO \xrightarrow{\text{photonic energy}}))\ ZnO(e^-_{(CB)})+(h^+_{(VB)}) \tag{1}$$

$$H_2O+ZnO(h^+_{(VB)})\rightarrow OH^{\cdot}+ZnO+H^+ \tag{2}$$

$$OH^-+ZnO(h^+_{(VB)})\rightarrow OH^{\cdot}+ZnO \tag{3}$$

$$O_2+ZnO(e^-_{(CB)})\rightarrow O_2^{\cdot -})+ZnO \tag{4}$$

$$H^++O_2^{\cdot -}\rightarrow HO^{\cdot}_2 \tag{5}$$

$$2HO^{\cdot}_2\rightarrow O_2+H_2O_2 \tag{6}$$

$$H_2O_2+ZnO(e^-_{(CB)})\rightarrow OH^-+OH^{\cdot} \tag{7}$$

$$O_2^{\cdot -}+H_2O_2\rightarrow O_2+OH^-+OH^{\cdot} \tag{8}$$

$$H_2O_2+\text{photonic energy}\rightarrow 2OH^{\cdot} \tag{9}$$

$$OH^{\cdot}+\text{Organic Contaminants}\rightarrow \text{Intermediates} \tag{10}$$

$$\text{Intermediates}\rightarrow H_2O+CO_2 \tag{11}$$

As stated, in comparison with titanium dioxide, zinc oxide has been demonstrated greater absorption performance over an extensive range of the solar spectrum. The capability to produce photoinduced electron-hole pairs is the major parameter that controlled the catalyst photoactivity. Nevertheless, quick recombination is the main limitation of zinc oxide photocatalyst which disturbs the photogenerated reaction. Moreover, the solar energy conversion efficiency of zinc oxide is heavily influenced by its optical absorption capability, which is related to its broad bandgap energy. Hence, intensive research has been dedicated to enhancing the optical behaviors of zinc oxide by reducing the bandgap energy and preventing the recombination rate. In the following sections, different approaches for synthesizing zinc oxide and the influence of different modification techniques like coupling two semiconductors, doping, and tuning the microstructure to enhance the efficiency of zinc oxide in the photocatalytic process will be studied in-depth.

ZINC OXIDE NANOSTRUCTURE MATERIAL

Zinc Oxide Classification

The application of zinc oxide for the photocatalytic process in different areas is heavily influenced by its nanostructure. During the post-remediation process, photocatalyst recovery and process performance will be improved by using an appropriate zinc oxide nanostructure. Different investigations have been attempted to produce zinc oxide with various types of nanostructures [52,53]. Zinc oxide nanostructures can be classified into 0D, 1D, 2D, and 3D. Nanoneedles, nanotubes, nanowires, nanofibres, and nanorods are different arrays of 1-dimensional zinc oxide. Nanoflowers and nanosheets are 3-dimensional and 2-dimensional nanostructures of zinc oxide, respectively. Because of the significant bulk of electron mobility, desirable mechanical strength, Table.1. Disadvantages and advantages of various zinc oxide nanostructures utilized in photocatalytic applications [61].

	Disadvantages	Advantages
Nano-thin film	Efficiency is limited by the small surface area	- Does not need post-remediation to eliminate the catalyst - Compatibility on certain substrates
Nanowires	- A lower surface area in comparison with nanoparticles - Growth conditions are significantly limited	- Lower crystallinity and more defects - Does not need post-remediation to eliminate the catalyst - Large efficient surface area in comparison with nano-thin films - Growth could be readily performed on most substrates
Nanoparticles	- Complete recovery of catalyst is challenging - Requiring post-remediation for catalyst elimination - Readily generates agglomerations in solution, which involves decreased efficient surface area	- Excellent efficiency because of their significant surface areas -Easy suspension in a solution

and stability of ZnO-dependent nanomaterial, they have been used in various areas, particularly in photocatalytic, electronic, and photovoltaics. Due to the excellent properties of nanosized zinc oxide quantum dots like significant stability and quantum yield, they can be integrated into a fluorescent probe as a sensing receptor for detecting Cu^{2+} in water [54]. Application in various areas like gas sensing, optoelectronics, and electronics is some of the prevalent potentials of 1-dimensional zinc oxide. According to the mechanical strength experiments of Desai et al. [55] on zinc oxide nanowires, the nanowires have an excellent fracture strain, which renders them a favorable option for nanoscale actuators and sensors. The specific surface area is a crucial parameter that greatly influences photocatalyst activity.

Due to the polar faces and remarkable specific surface area of 2-dimensional zinc oxide [56], its nanosheets are competitive choices in the photocatalytic process. The substantial specific surface area of zinc oxide increases the number of pollutants adsorptions on the active surface of zinc oxide and hence increases the contaminants attacked by $OH^{\cdot}$. The decomposition of pollutants for the production of harmless products will be improved because of the chain reactions which happen at the surface of ZnO. In comparison with 2-dimensional and 1-dimensional nanostructures of ZnO, because of the higher surface-to-volume ratios of 3-dimensional nanoflowers, Luo et al. [57] stated that they demonstrated more sensitivity in gas ethanol sensing. As a result, different morphologies of materials will determine the abilities of sensors. Moreover, compared with nanorods, zinc oxide nanoflowers have better light scattering behaviors [58]. By integrating zinc oxide nanowires on polystyrene spheres that joint each other with a zinc oxide seed layer, nanoflower arrays of zinc oxide have been fabricated by Xie et al. [59]. Hence, by this method, the benefits of 1-dimensional and 3-dimensional zinc oxide materials can be integrated which results in a greater specific surface area. Moreover, this method renders 3-dimensional nanoflowers competitive material for application in solar-related areas like the fabrication of dye-sensitized solar cells. The pros and cons of various zinc oxide nanostructures in photocatalysis applications are demonstrated in Table 1. Because of the significant surface areas of nanoparticles, they have widespread applications in the solar photocatalytic process, since higher contaminants can be readily adsorbed and hence a greater degree of decomposition is obtained. Higher defects and low crystallinity of nanowires are advantageous in photocatalysis applications which, possibly because of the hydroxyl groups bound on the defects like surface and oxygen defects which facilitate the catching of photogenerated electron-hole pairs and hence improve their separation [60].

Synthesizing of Zinc Oxide Nanostructure

For synthesizing zinc oxide nanostructures, different methods are available which can be classified into the vapor phase and solution-dependent methods. Crystallinity, crystal phase, and size are major elements that determine charge carrier separation and bandgap energies. The size of semiconductor and bandgap energy is inversely proportional together [62]. Hence, better control of synthesizing procedures ascertains the performance of the photocatalytic process. Among different synthesizing zinc oxide approaches, the solution-dependent method is the simplest in which energy consumption is very low. Using this method and changing the experimental parameters like reaction conditions, initial materials, and different kinds of solvents, nanostructures morphology and nanostructures sizes can be readily controlled [63]. Solution-dependent methods for synthesizing zinc oxide nanostructures are electrospinning, flux approaches, wet chemical technique, polyol, microwave, electrochemical deposition, solvothermal, microemulsion, precipitation, sol-gel, and hydrothermal [64–67]. For synthesizing zinc oxide nanostructures, the sol-gel is the most appealing one due to its optical properties, excellent compositional homogeneity, easy controlling of morphology and characteristics of nanoparticles, low processing temperature, simple procedure, and desirable repeatability, reliability, and cost-effective production cost [68]. In view of the current scenario, continuous research works have improved the available sol-gel method by utilizing water as a solvent [69]. For example, utilizing modified sol-gel iron-doped zinc oxide nanoparticles was fabricated by Ciciliati et al. [70] which were observed to be similar to prior research [71] that applied ethanol as a solvent. As a result, the modified sol-gel technique was successful for the production of high-quality nanostructure materials however with lower generation cost.

For production of nanostructured materials, vapor phase methods are molecular beam epitaxy (MBE) [72], plasma enhanced chemical vapor deposition (PECVD) [73], metal-organic chemical vapor deposition (MOCVD) [74], chemical vapor deposition [75], physical vapor deposition [76], pulsed laser deposition [77] and thermal evaporation [78].

Some of them are for better controlling of nanostructures growth using metal catalysts. Nevertheless, without adding any metal catalysts zinc oxide nanorods with better optical properties and crystallinity were fabricated by Tang et al. [79]. By changing the initial growth temperature of samples, they managed the growth process of zinc oxide, which result in the generation of various nucleation layers for zinc adsorption. Selecting proper zinc oxide synthesizing approach is primarily based on the dimension of targeted nanostructures. For instance, sol-gel and PECVD approaches have been used for the fabrication of 2-dimensional nanostructures. The PECVD technique has confirmed that in comparison with the sol-gel technique, lower temperatures are enough to generate the films [80].

ENHANCEMENT OF ZINC OXIDE PHOTOCATALYST

Metal or Non-Metal Doping

Because of the significant ionicity at the borderline between ionic semiconductors and the covalent and tetrahedral bonding arrangement of zinc oxide, it has broad bandgap energy. Rocksalt, cubic zincblende, and hexagonal wurtzite are three kinds of zinc oxide crystals [81]. Only at significant pressures, zinc oxide can generate a rocksalt structure. Cubic zincblende can be stabilized by growing zinc oxide on cubic substrates while at ambient conditions hexagonal wurtzite is thermodynamically the most stable [82]. The crystalline structure of zinc oxide was depicted in Fig.4. In general, zinc oxide is an inherently n-type semiconductor with the existence of inherent defects like zinc vacancies (V_{Zn}), zinc interstitials (Zn_i), and oxygen vacancies (V_O) which will heavily influence its optical and electrical behaviors [83]. Electron charge carriers and oxygen vacancies are directly proportional together. The inherent doping asymmetry of zinc oxide by microscopic equilibrium mechanism was explored by Zhang et al. [84] and the results demonstrated that through internal or external dopants n-type zinc oxide can be readily achieved. The main issues for the fabrication of zinc oxide semiconductors are the challenges in achieving a stable and reproducible p-type zinc oxide. Following are the restrictions of asymmetric doping: (1) Electron-hole radiative recombination at the oxygen vacancies performed as the source of the green luminescence was detected, (2) zinc oxide cannot be utilized to dope p-type by intrinsic defects like oxygen interstitials and zinc vacancy and (3) at Zn-rich situations, zinc oxide is n-type. A self-compensation effect can readily happen when zinc oxide is under Zn-rich circumstances. The intrinsic donor defects that are easily accessible from oxygen vacancies, zinc interstitials, or zinc oxide will offset native acceptor defects (zinc vacancies or O_i), hence, facilitating

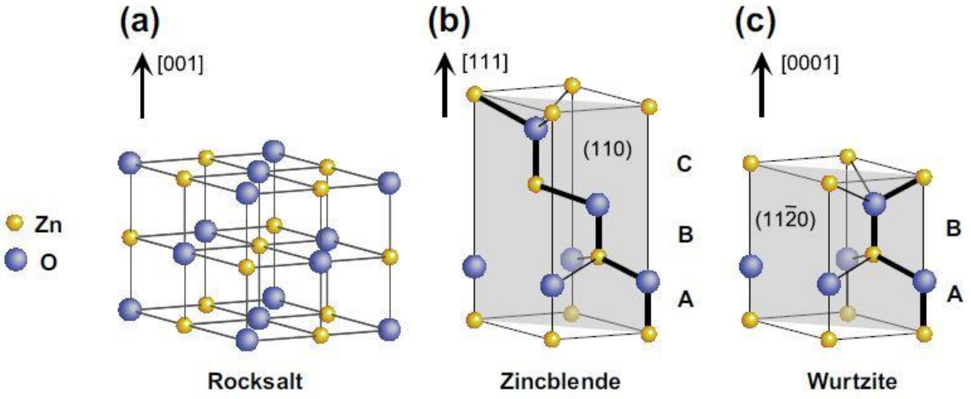

Fig.4. Stick and ball demonstration of zinc oxide crystal structures: (a) cubic rock salt, (b) cubic zincblende, and (c) Hexagonal wurtzite [88].

the generation of n-type instead of p-type zinc oxide [85]. The good quality of p-type zinc oxide is optimum for different utilizations because of its significant radiative stability. By applying the sol-gel technique, the fabrication of good quality Ag-N dual-doped p-type zinc oxide was performed by Duan et al. [86]. Between different fabrication approaches like dual-doping, codoping, and mono-doping, for synthesizing p-type zinc oxide, the dual-doping technique is essential. Also, because of the various features of dopants that enable modifying the optical behaviors of photocatalysts, co-doping has been garnering significant focus [87].

As stated previously, $OH^{\cdot}$ radicals and bandgap energy are remarkably controlled the photocatalysis reactions. The low-charge separation performance is the major disadvantage of zinc oxide semiconductors as photocatalytic material. As a result, modification of chemical and physical characteristics through different doping methods has been applied like integrating impurities like metals or non-metals, to increase the VB energy of zinc oxide and decrease the bandgap energy to the visible-UV area [89]. According to the current investigations, non-metal dopants like N, C, S, and F can increase the zinc oxide bandgap through alternating oxygen vacancies, hence developing higher oxygen vacancy defects on the surface of nanoparticles. To be more specific, because of extrinsically small sizes of carbon, fluorine, oxygen, and nitrogen, they can spread through the lattice interstices and attach to the atoms by the oxidation process [90]. Because of the special electronic characteristics, high chemical resistance, and significant mechanical strength of carbon, it is the main option for a non-metal dopant in semiconductors [91]. Moreover, doping can result in a higher generation rate of hydroxyl radicals which following that the degradation performance of organic contaminants will increase [92]. This is because the dopants can perform as electron scavengers and prevent the recombination of electron-hole pairs [93,94].

Lately, noble metals, rare earth metals, and transition metals besides other metals have demonstrated great benefits in modification of the zinc oxide morphology in photocatalysis employments in addition to other special applications. Metal doping of zinc oxide can remarkably enhance the catalysts photocatalytic activity through rising the trapping site of the photo-generated charge carriers and hence reduce the recombination of photo-generated electron-hole pairs [95]. For reduction of photocatalysts bandgap energy, for application in gas sensing and dye mineralization, metal dopants like aluminum, copper, neodymium, and cerium have been utilized [96,97]. According to the significant light-harvesting performance, in comparison with pure zinc oxide greater volumes of organic matter can be adsorbed on Al-doped zinc oxide [98]. Tin can be utilized to improve the electrical conductivity of zinc oxide via alternating the Zn^{2+} ions without resulting in any large lattice deformity [99]. The integration of metal ions like ferric ions can increase oxygen defects on zinc oxide besides increasing the charge density of zinc oxide which accordingly can motivate greater nanostructure efficiency [100]. The same study performed by Wang et al.[101] and showed that Ag-doped zinc oxide has a higher lattice deficiency and specific surface area. Metaldoped zinc oxide, synthesized via manganese and cobalt using a co-precipitation technique, was applied for the treatment of methyl orange [102]. It was demonstrated that solution pH heavily influenced the decomposition and adsorption of methyl orange onto the heterogeneous catalysts. Based on the results, in an acidic environment like pH 4 using Mn-doped zinc oxide catalyst, the optimum dopant dosage was 12% which yields the highest degradation performance. Despite numerous benefits of metal dopants, in comparison with metal dopants, non-metal dopants present lower costs. Additional research work might be required to investigate the efficiency, properties, and trends of zinc oxide doping comprehensively.

As stated earlier, to enhance the charge separation of the photocatalytic process metal/non-metal doping has been extensively explored. Nevertheless, according to the literature, the decreasing performance of zinc oxide photodegradation can be imputed to over-doping [103] which is possible because the excessive dopants perform as trapping sites for electrons-holes and as a result, prevents the production of $OH^{\cdot}$ which are essential for degrading persistent contaminants.

Synthesizing of Zinc Oxide/Graphene, Challenges and Outlooks

Graphene plays a pivotal role in all regions of material science like metallurgical processes, induction heaters, inks, foams, plastics, glues, varnishes, paints, materials construction, pharmaceuticals, medical monitoring devices, brain chemistry, cancer, biocompatible implants, catalysts, energy storage materials and so on [104]. They have gained significant attention in the field of wastewater remediation by photocatalytic process, because of exceptional characteristics of graphene-dependent nanomaterial like significant thermal stability, excellent electron conductivity, and higher surface area [105]. Among semiconductor nanoparticles, the optoelectronic, catalytic and magnetic properties of graphene nanocomposites have gained substantial attention [106,107]. Nanoparticles are regarded as a stabilizer opposite to the agglomeration of solitary graphene sheets by powerful van der Waals forces between graphene layers rising the surface area for contaminants elimination from aqueous media. Hence, intensive research attempts and innovative methods are required for the production of graphene-dependent nanocomposites [108]. For contaminant elimination, semiconductors often modify the characteristics of graphene structure and hence render it highly active in photocatalytic utilizations [109]. As previously discussed, zinc oxide as a photocatalyst has outstanding properties like pyroelectric and piezoelectric features, great photosensitivity, non-toxicity, biocompatibility, chemical stability, and better photocatalytic activity and powerful oxidation capability. However, it has several intrinsic limitations like absorption in UV area and photo corrosion as a result of modification. Hence, the hybridization of zinc oxide with graphene-dependent nanocomposites is a promising substitute for the mineralization of organic and inorganic pollutants which are existed in water.

Many researchers explored the photoactivity of heterojunction/nanocomposite for degrading organic contaminants and demonstrated that graphene and its by-products play a pivotal function in increasing photocatalytic efficiency [110]. When solar light radiates on zinc oxide and graphene nanocomposite surfaces, electrons are excited from VB to CB of zinc oxide. Graphene sheet in zinc oxide is highly efficient and crucial for grabbing excited electrons in zinc oxide and prevents electron-hole recombination. As a result, it performs as an efficient charge carrier and amended excited electron transformation under solar light radiation. The produced radicals degrade adsorbed contaminants onto the surface of zinc oxide and graphene nanocomposite. The electron transformation through graphene layers as electron receiver and transporter improves the photodegradation activity of the zinc oxide and graphene nanocomposite under solar light radiation. Graphene avoids charge carriers recombination onto the surface of zinc oxide nanoparticles and enhancement in photodegradation rate provides a synergistic effect between zinc oxide nanoparticles and graphene layers which finally increases the degradation of contaminants effectively [111,112].

Numerous research attempts have been dedicated to developing different zinc oxide and graphene oxide or reduced graphene oxide nanocomposites. So far, various investigations have demonstrated that the zinc oxide and graphene oxide or reduced graphene oxide heterostructures can significantly enhance photocatalytic efficiency by extending operational lifespan, enhancing charge transportation and separation, and augmenting the absorption of light. Nevertheless, disconcerting challenges must be alleviated to bypass the major technical and operational hurdles that obstruct their application in large scale and industry:

- The generation of zinc oxide and graphene oxide or reduced graphene oxide heterostructures enhanced the electron-hole separation, which is essential for advancements in their photocatalytic efficiency. However, despite outstanding developments that have been carried out for synthesizing zinc oxide and graphene oxide or reduced graphene oxide nanocomposites, still, their well-developed generations with controlled interface, morphology, and size are challenging. This is because these elements are absolutely crucial for effective charge separation. Moreover, decreasing recombination is not enough to the reduction of the cost related to the ultra-violet source. Another problem is a modification of zinc oxide and graphene oxide or reduced graphene oxide nanocomposites to widen photocatalysts operation into the visible region. In order to realize the charge production, transportation, and separation over those nanoscale interfaces and the generation of reactive oxygen species are essential. So far, exploration has concentrated only on enhancing photocatalytic performance on the lab scale.
- The fabrication of graphene oxide or reduced graphene oxide is challenging for graphene-dependent methods. Since currently, there are no measurable and low-cost approaches for the production of huge amounts of graphene and its by-products. Because of the outstanding viability of graphene and its by-products in many areas, their commercialization in the imminent future is highly likely. For better designing of zinc oxide and graphene oxide or reduced graphene oxide, fabrication of graphene oxide or reduced graphene oxide must be the focus of attention in future studies.
- Generally, the combination of reactor design and photocatalyst fabrication still has not been fully understood for applications in wastewater disinfection. Since the photocatalytic stability and efficiency remarkably relies on reactor design, as a result, the capability of the heterogeneous photocatalytic process heavily depends upon the optimum reactor design. Hence, developing, synthesizing, and designing novel zinc oxide and graphene oxide or reduced graphene oxide nanocomposites for application as photocatalysts requires comprehensive knowledge regarding its all dimensions. In this context, for reaching a bright future, trans-disciplinary efforts between engineers and scientists are highly demanded. By this method, it is highly likely to resolve some of the major challenges associated with recyclability, reusability, and the post-recovery of zinc oxide and graphene oxide or reduced graphene oxide nanocomposites.
- Because of the limited efficient lifespan of photocatalysts which is driven by low photo corrosion resistance and low chemical corrosion, highly stable photocatalysts is another feasible challenge in their progress. For ZnO-dependent photocatalysts, in comparison with chemical corrosion, photo corrosion is most critical. So far, various investigations have concentrated on enhancing the photo corrosion resistance of ZnO-dependent photocatalysts. Nevertheless, for zinc oxide and graphene oxide or reduced graphene oxide nanocomposites intensive research attempts are required.
- Synthesizing complicated zinc oxide and graphene oxide or reduced graphene oxide nanocomposites with well-planned structures and various shapes or morphologies is still problematic. Bioinspiration can be an intelligent approach for designing innovative and effective structures with enhanced contaminant-adsorbing and light-catching capacities. In spite of significant advancements in the synthesizing of zinc oxide and graphene oxide or reduced graphene oxide nanocomposites, the synthesized nanocomposites are mainly dependent on zinc oxide nanoparticles integrated into graphene oxide or reduced graphene oxide.

Application of SEM Images for Analyzing Synthesized Catalysts Based on Zinc

For the generation of an enlarged image for analysis, SEM images were applied through scanning of the sample with an electron beam [113]. Fig.5 presents the morphology of the synthesized photocatalysts as carbon nanotubes and rods on the substrate. The photocatalyst was synthesized using the hydrothermal technique. The micrographs of $ZnAl_2O_4$ and $ZnAl_2O_4$/CNT were obtained using SEM analysis are demonstrated in Figs.5 (a,b) and (c,d), respectively. The $ZnAl_2O_4$ micrograph exposed aggregated particles with the morphology of shaped plate type and minor

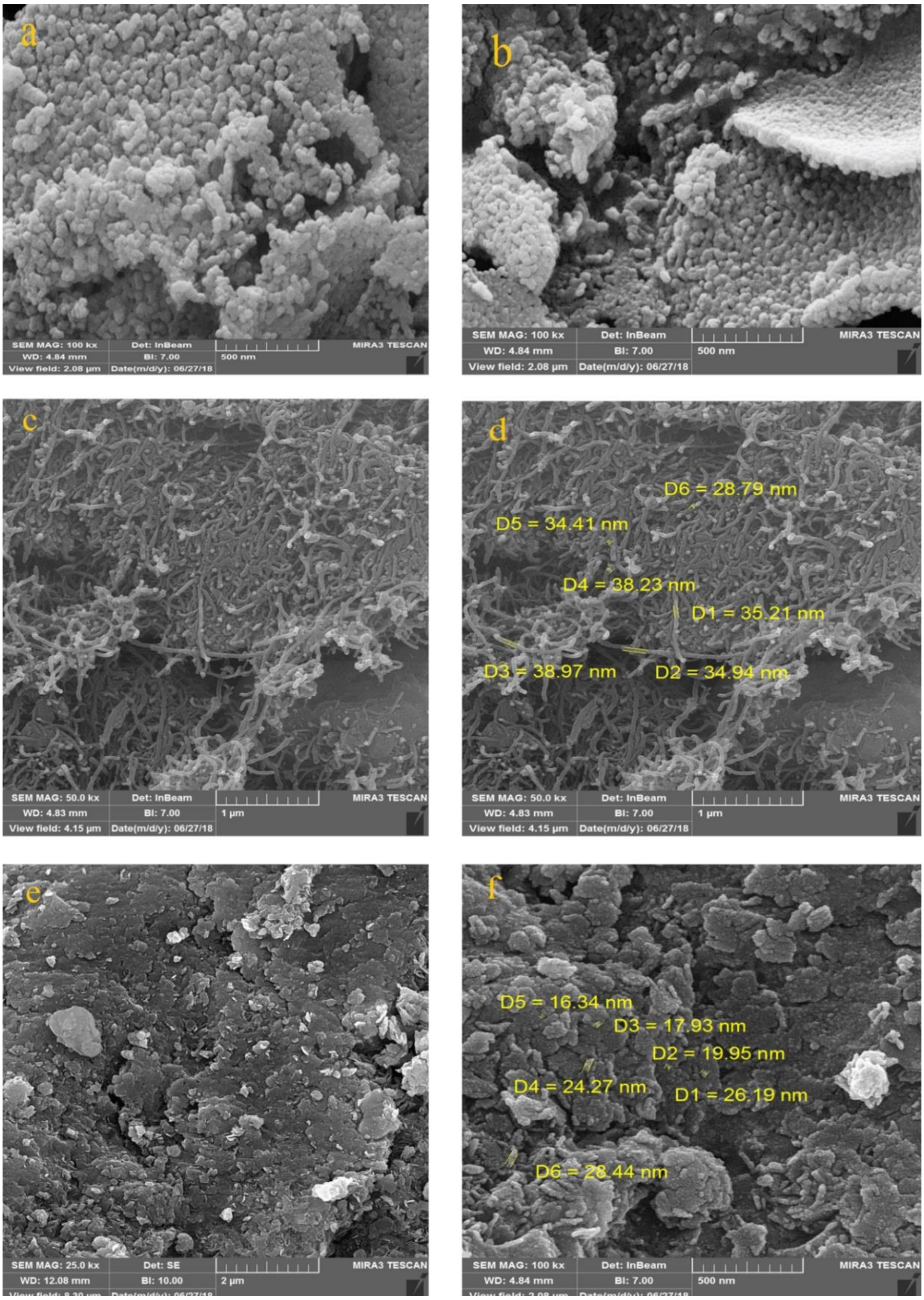

Fig.5. SEM images of the synthesized photocatalysts: (a,b) ZnAl$_2$O$_4$ photocatalyst, (c,d) ZnAl$_2$O$_4$/CNT photocatalyst, (e,f) ZnAl$_2$O$_4$/SiO$_2$ photo catalyst [113]three different types of Zn-based catalysts, including ZnAl2 O4, ZnAl2 O4 /SiO2 and ZnAl2 O4 /CNT (CNT – carbon nanotube.

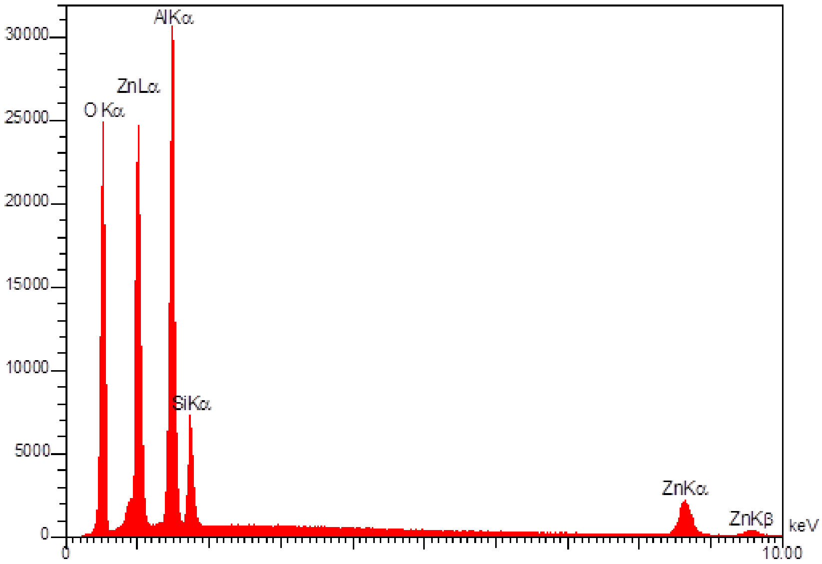

Fig.6. EDX spectrum of $ZnAl_2O_4/SiO_2$ photocatalyst [113]three different types of Zn-based catalysts, including ZnAl2 O4, ZnAl2 O4 / SiO2 and ZnAl2 O4 /CNT (CNT – carbon nanotube.

aggregates on the surface of larger clusters [114]. The SEM patterns of $ZnAl_2O_4$/CNT and $ZnAl_2O_4/SiO_2$ Fig.5 (e,f) demonstrated the existence of plate-like accumulations with unequal surface and porous structures. The catalysts morphology heavily relies upon the utilized synthesis procedure. Motloung et al. [114] studied zinc aluminate powders with rod-like-needles morphology prepared using citrate sol-gel method and Du et al. [115] explored the $ZnAl_2O_4$ powders synthesized with polyhedral morphology using the solid-state technique.

According to the SEM images of the samples, the particles of $ZnAl_2O_4$ dependent catalysts are almost spherical with narrow particle size distribution. Through comparison between pictures, it is obvious that in comparison with SiO_2, the size of the carbon nanotube nanoparticles is smaller and as a result, its surface area is higher, which augments the carbon nanotube photoactivity efficiency. These findings are accepted through the BET outcomes which are shown in the next part. Both synthesized Zinc dependent catalysts demonstrated a solid affinity for accumulation with plate-like morphology [113].

Elemental Analysis of the Synthesized Catalysts Using EDX Approach

For qualitative elemental analysis and chemical characterization of samples, the EDX method was utilized [116,117]. The zinc-dependent catalysts were containing oxygen, silicon, carbon, Aluminum, and zinc. Figs. 6 and 7 depicted the EDX spectra of $ZnAl_2O_4/SiO_2$ and $ZnAl_2O_4$/CNT nanocomposites. It is evident that the existence of Zn, Al, O_2, and Si elements in the $ZnAl_2O_4/SiO_2$ nanocomposites and Zn, Al, O_2, and C in the $ZnAl_2O_4$/CNT nanocomposite are indicative of synthesizing these materials and the lack of other ones.

BET Outcomes of Synthesized Samples

At the temperature of -196 °C and using nitrogen adsorption, the specific surface area and pore volume of the prepared catalysts were ascertained. Physical features like volume and porosity of cavities besides specific surface area were determined by BET analysis. According to the N_2 adsorption-desorption isotherms of the two $ZnAl_2O_4$/CNT and $ZnAl_2O_4/SiO_2$ samples, the former possess more maximum surface area than later. Table 2 shows the cavity volume, cavity size, and surface area of the two samples. The specific surface area of the $ZnAl_2O_4$ catalyst was 80 m^2/g. With the addition of carbon nanotube to the catalyst composition, the surface area of the $ZnAl_2O_4$ is remarkably raised, which is crucial because of its critical role in reaching the reactive molecules in the catalytic substances. Moreover, with increasing pore size the BET surface area of the sample was decreased (see Table 2) [113].x

Based on the outcomes, the higher surface area

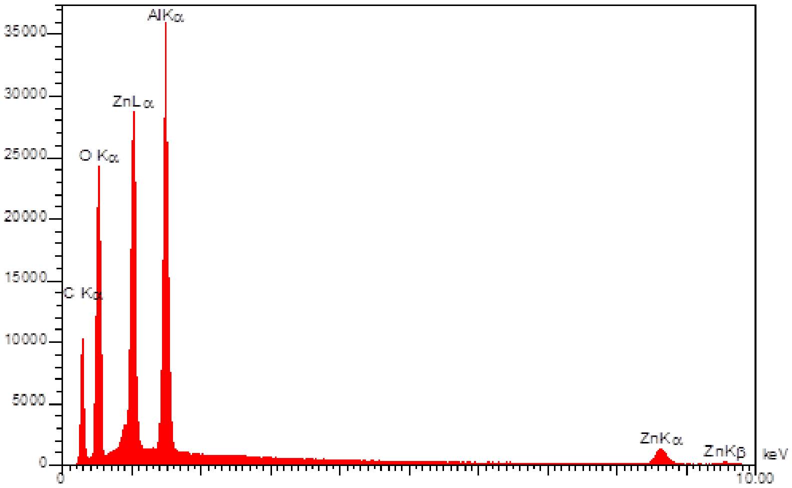

Fig.7. EDX spectrum of ZnAl$_2$O$_4$/CNT photocatalyst [113]three different types of Zn-based catalysts, including ZnAl2 O4, ZnAl2 O4 /SiO2 and ZnAl2 O4 /CNT (CNT – carbon nanotube.

Table 2. Comparing BET outcomes in synthesized samples [113].

Sample	Pore Size(nm)	Pore Volume (Cm^3/g)	Surface Area (m^2/g)
$ZnAl_2O_4$/CNT	8.46	0.427	201.81
$ZnAl_2O_4/SiO_2$	31.0	0.773	94.59

facilitates and increases reaction or absorption sites throughout the photocatalytic reaction and enhances the photoactivity. In comparison with $ZnAl_2O_4$ /SiO$_2$, the $ZnAl_2O_4$ /CNT powder showed a higher BET surface area and smaller pore volume and size. Ballarini et al. [118] studied the catalytic efficiency of Pt-$ZnAl_2O_4$ on the n-butane dehydrogenation reaction and concluded that the $ZnAl_2O_4$ catalysts have demonstrated significant BET surface area and highest catalytic activity. In this study, the significant surface area and porous structure of $ZnAl_2O_4$/CNT are of utmost importance for catalytic applications.

Photoluminescence Spectroscopy (PL) Analysis

The photoluminescence spectroscopy analysis is not only a significant approach for the realization of the processes happening on the material surface but also is considered essential for exploring the photoelectric, optical, and electrical features of semiconductor nanomaterial. The enhancement in the photoactivity and other nanoparticles stemming from a single up-conversion of graphene quantum dots or carbon has been explored by numerous researchers [119,120]. Changing the graphitic carbon nitride using GO or rGO and their photoluminescent and photocatalytic activity was investigated by Aleksandrzak et al. [121]. According to the photoluminescence spectroscopy analysis, electron-hole recombination can be explored. It can be seen that the peak intensities reported in this experiment were lower, hence the recombination of the electron-hole pairs is reduced and the separation performance is augmented. This will cause increasing photocatalytic mineralization of the contaminants. The photoluminescence spectroscopy spectrum of $ZnAl_2O_4/SiO_2$ and $ZnAl_2O_4$/CNT nanocomposites with excitation wavelength at 340 nm presents an intense peak at 650 nm wavelength and the former is higher than later nanocomposites which indicates that the $ZnAl_2O_4$/CNT sample possess greater separation performance and lower electron-hole recombination. As a result, photodegradation

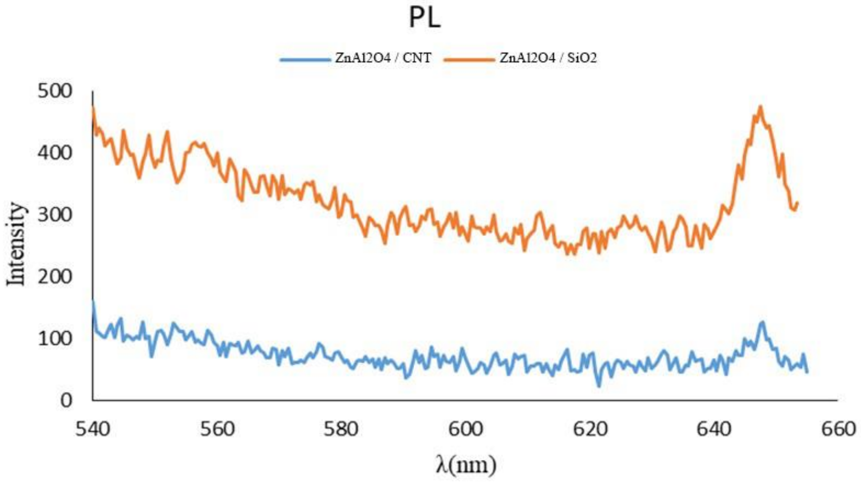

Fig.8. Photoluminescence spectrum of $ZnAl_2O_4/SiO_2$ and $ZnAl_2O_4$/CNT [113]three different types of Zn-based catalysts, including ZnAl2 O4, ZnAl2 O4 /SiO2 and ZnAl2 O4 /CNT (CNT – carbon nanotube.

of pollutants is increased using the $ZnAl_2O_4$/CNT sample. It must be mentioned that the photoluminescence spectroscopy spectra of pure $ZnAl_2O_4$ are not presented in Fig.8, however, based on the findings of other researchers [122], it was evident that compared with peaks of $ZnAl_2O_4/SiO_2$ and $ZnAl_2O_4$/CNT sample at 650 nm, it has a more intense peak, which indicates that in comparison with the modified ones it possesses significant electron-hole recombination and hence lower photoactivity.

MAGNETIC PHOTOCATALYSTS AND THEIR APPLICATIONS IN ZINC OXIDE

Despite the outstanding efficiency of photocatalysts in alleviating environmental-related challenges, their separation and recycling from the reaction solution are still problematic [123]. For the separation of applied photocatalysts from remediated solutions, filtration and centrifugation techniques are utilized. Nevertheless, these strategies are time-prohibitive and costly which significantly limits the application of photocatalysts in a wide range [124]. As a solution for overcoming this challenge, several researchers suggested the immobilization of photocatalysts into an inactive substrate. Even though this approach reduces particle accumulation and promotes photocatalysts separation, the photoactivity is highly feasible to reduce because of the decreasing photocatalyst active sites [125,126]. To bypass this issue, photocatalysts are integrated with magnetic substances for separation and recovering the applied photocatalysts in wide ranges [127]. Besides better ability in the separation of magnetic materials, they can demonstrate remarkable photodegradation activities, which render them highly efficient and promising materials in photocatalyst-enabled operations [128]. In this context, different magnetic materials like Fe_3O_4/ZnO [129], Fe_2O_3/ZnO/TiO_2 [130], Fe_2O_3/ZnO [131], etc. were fabricated. Fig.9 presents different categories of magnetic photocatalysts dependent on zinc oxide.

The broad bandgap is considered as the major disadvantage of several magnetically separable photocatalysts synthesized utilizing zinc oxide, which renders them active only under ultra-violet light sources [132]. Solar energy involves only 5% of ultra-violet light, which renders it inefficient in performing photocatalytic reactions when zinc oxide is applied as photoactive material [133]. Hence it is crucial to apply highly efficient approaches for raising the performance of photocatalysts activated with sunlight [134]. As a result, intensive research works have been carried out for the fabrication of magnetically recoverable visible-light-driven photocatalysts utilizing zinc oxide which includes (a) bandgap engineering by the generation of defect states and doping of metal and non-metal elements, (b) incorporation with small bandgap semiconductors and (c) sensitization with dyes, organic compounds, and polymers [135]. Besides excellent magnetic features, various magnetic substances are better sensitizers and possess better visible light activity, because of their limited bandgaps, however, their photoactivity is negligible

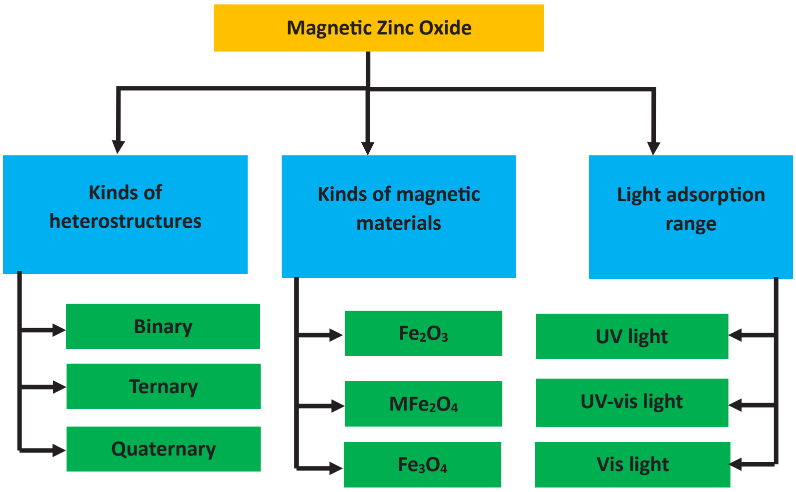

Fig.9. Different categories of magnetic photocatalysts are dependent on zinc oxide.

due to the rapid recombination of photogenerated electron-hole pairs [136].

The stability and recoverability of the magnetically separable photocatalysts driven by zinc oxide is one key parameter that precisely determines their design as efficient photocatalysts. Nevertheless, several reported photocatalysts are not effective enough under light irradiation, due to the significant rate of photo corrosion. Hence, still intensive research efforts are required for the protection of magnetic photocatalysts utilizing various methods without compromising photoactivity. Moreover, profound explorations concerning the application of earth-abundant materials in the framework of the magnetically separable photocatalysts must be performed for the fabrication of highly efficient photocatalysts with low cost which is economically feasible for their industrialization.

HYBRIDIZED PHOTOCATALYTIC AND MEMBRANE SYSTEMS

The combination of membrane and photocatalysis methods is a competitive and highly promising approach due to the fact that it does not need complex procedures for photocatalysts recovery after water remediation. To improve the absorption of reactants and photons, this integrated method utilizes immobilized nanostructured photocatalysts hence the catalyst suspension is obviated [137]. Because of the remarkable effectiveness of nanostructured photocatalysts in the degradation of recalcitrant organic contaminants into easily biodegradable matters, they have garnered significant attention from the research community. For numerous applications like Congo red dye remediation [138], fungicides in leaching water (for example fludioxonil and cyprodinil) [33], pharmaceutically active compounds (for instance diclofenac) [139], and the elimination of endocrine compound (like estrone) [140], integrated photocatalytic and membrane systems like nanofiltration and ultrafiltration have been broadly explored and expanded. So far, a remarkable enhancement in photocatalytic membrane reactors (PMR) has been occurred in which instead of ultra-violet radiation, sunlight is harvested. This alternation of the light source is highly desirable because of its extensive application in regions where sunlight is easily available and electricity cost is lower. Although the integrated photocatalytic-membrane technology is highly competitive and promising for water remediation purposes, the presence of several technical limitations which require to be efficiently bypassed before it can be conducted in a wide range is inevitable. (1) For exploring the stability of polymer membranes in photocatalytic membrane reactors, relatively limited investigations have been implemented. (2) Only limited works regarding the kinetic models and mass transformation restrictions that heavily influence the efficiency of the separation in photocatalytic membrane reactors have been done. (3) Rather than synthetic samples, explorations on the efficiency of photocatalytic membrane reactors must be performed utilizing

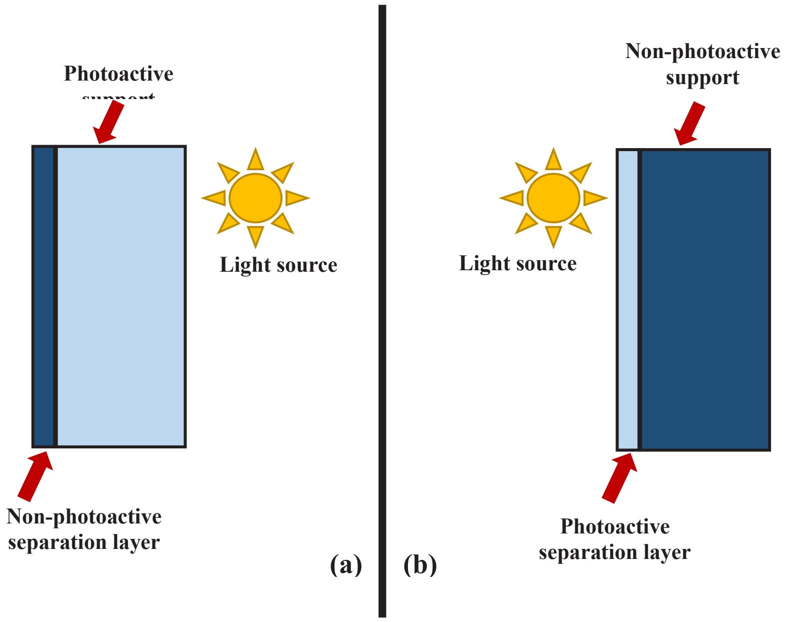

Fig.10. Designs of photocatalytic membrane reactors with photocatalytic membranes (a) membrane with non-photoactive layer deposited on a porous photoactive support (b) membrane with photoactive layer deposited on a porous non-photoactive support [141].

real-world water samples in order to study the real circumstances, like the influences of colloidal particles against the stability of photocatalytic effectiveness. Following are some of the great benefits of photocatalytic membrane reactors with an immobilized photocatalyst on a membrane substrate [141,142]:

(a) Because of the degradation of organic pollutants and improved hydrophilicity of the modified membrane, the membrane fouling phenomenon is alleviated to some extent.

(b) Pollutants must be degraded, either in feed or in permeate.

(c) Highly stable flux and low degree of flux-reduction.

(d) Additional complicated photocatalyst recovery processes are eliminated.

Because of outstanding features of photocatalytic membranes like simultaneous photocatalytic oxidation and separation in comparison with traditional membranes, super hydrophilicity, anti-microbial, and anti-fouling they are extensively applied in various separation systems [143,144]. As it can be seen in Fig.10 there are two types of photocatalytic membrane reactors: (a) membrane with a non-photoactive layer deposited on photoactive porous support and (b) membrane with a photoactive layer deposited on non-photoactive porous support. Type-a membrane designs are less efficient because photodegradation is performed on the permeate side and hence decreasing permeate water quality [42]. Because organic compounds can be efficiently degraded prior to transformation across the membrane, hence increasing permeate per flux and fouling alleviation are considered the major benefits of the type-b membrane design. Immobilizing photocatalyst particles on membranes is challenging. Photocatalytic membranes can experience destruction to some extent after a long time of radiation exposure [145]. Hence, exploration of durability, and UV light and OH^{-} resistance of polymer membranes has to be performed for overcoming polymer membranes destructions.

In the suspended reactor units during the mineralization process nanoparticles are suspended in the wastewater. As a result, for catalyst recycling, its separation and recovery are crucial stages. Through the application of a suspension system and zinc oxide photocatalyst immobilization, under sunlight, Hu et al. [146] studied the decoloration of methyl orange. According to their results, after 6 h of radiation utilization, zinc oxide immobilized on a polymeric membrane only 74% of decoloration was obtained, while under 40 min of sunlight radiation in a slurry type reactor around

98% of methyl orange decoloration was obtained. This is due to the fact that in comparison with a photocatalyst immobilized on a membrane, the slurry-type reactor presents a significant surface area of the photocatalyst. Both immobilized and suspended systems have their own merits and demerits, as a result, specific applications will determine their selection.

CHALLENGES AND FUTURE OUTLOOKS

It is a widely held opinion that in comparison with traditional techniques the application of solar-enabled photocatalytic processes is capable to work more effectively for degrading persistent organic contaminants. To improve the futuristic viability of such a novel method, numerous attempts are required to effectively surpass several associated challenges. Photodegradation explorations should be conducted utilizing the real organic contaminant instead of synthetic or alternative ones. According to the literature, many research works were implemented utilizing dye as sample organic contaminants for the photodegradation process. Nevertheless, in comparison with other organic contaminants like endocrine-disrupting constituents and pesticides, dyes can be readily eliminated and their elimination can be readily obtained. A profound realization of the decomposition mechanisms and the interaction between the photocatalysts and other organic contaminants is still highly demanded. In addition, still many enhancements might be performed for developing nanostructures. In most conditions, the photocatalysts greatly controlled the performance of photocatalytic reactions. Although for fabrication of high-quality photocatalysts with respect to longer charge carrier lifetime, small particle size, and higher surface area, recent methods are highly capable and still intensive research is required in this region. Moreover, it is highly recommended that researchers additionally explore the efficiency and quality of photocatalytic membranes when photocatalytic membrane reactors are selected. There are some disconcerting challenges associated with the fabrication of membranes-based photocatalytic processes, like production cost, photocatalysts leaching from supports, and the sustainability of polymer membranes. Furthermore, the photocatalytic activity of recycled photocatalyst and loss and recovery of photocatalyst throughout post-remediation are some of the operational issues which require to be effectively overcome. Hence, for the prediction of process optimal conditions, kinetics, and quantum yield besides enhancing and confirming the mathematical models in photocatalytic-driven processes for wastewater remediation applications, intensive research efforts are required.

CONCLUSION

Zinc oxide photocatalyst with several special characteristics such as many active surface sites and enhanced specific surface area streamline contaminants absorption and empowers reactions of photogenerated charge carriers with different types of contaminants is a well-worth photocatalyst. Nevertheless, limited visible light absorption and photo corrosion significantly confined the photocatalytic activity of zinc oxide. Different graphene-dependent nanomaterials have been utilized as a support material to improve the photodegradation activity of various photocatalysts. Because of their structural characteristics and features, different techniques were applied to fabricate graphene-dependent nanomaterial and zinc oxide/graphene-dependent nanocomposites. The synthesized nanocomposites effectively degrade contaminants. The integration of graphene-dependent nanocomposites with photoactive zinc oxide enhanced its photodegradation capability against different organic and inorganic contaminants like bacterial disinfection, heavy metal ions, antibiotics, phenolic compounds, and dyes. The proper fabrication of nanocomposites was desirable for contaminants mineralization and zinc oxide reusability is needed to enhance by modifying the band gap in order to it become transferable to visible light aided photocatalyst area. Graphene-dependent nanomaterial remarkably increased the functionality of zinc oxide/graphene-dependent nanocomposites including increasing stability, improving charge separation, and adsorption capacity besides augmenting light absorption range. However still in real-world applications contribution of advanced mechanisms is extremely crucial for the effective application of nanocomposites for contaminants mineralization which will be dramatic progress towards enhancing the performance of multifunctional nanocomposites.

CONFLICT OF INTEREST

The authors declare no conflicts of interest.

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