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A REVIEW ON SYNTHESIS AND CHARACTERIZATION OF Cu DOPED $ZnFe_2O_4$ NANOPARTICLES FOR HIGH PHOTOCATALYTIC ACTIVITY UNDER VISIBLE LIGHT

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ABSTRACT
Organic contaminant-related environmental pollution is one of the most vital issues all around the world, and sustainable solutions are being considered. In this context, we report Cu-doped $ZnFe_2O_4$ nanoparticles as a visible light-driven photocatalyst for the potential substrate for eco-friendly UV-independent systems. Nanoparticles have been prepared by using the hydrothermal method and further characterized with XRD, SEM, EDX, and UV-V is spectroscopy. The analyses revealed a crystalline spinel structure and nanoscale uniformity with improved visible-light absorption owing to Cu doping. Photocatalytic activity was estimated by the degradation of organic dyes under visible light illumination, and the Cu-doped $ZnFe_2O_4$ specimens presented significantly higher degradation rates compared to the undoped specimens. This supports the idea that Cu-doped $ZnFe_2O_4$ represents an encouraging photocatalyst for pollution remediation and better utilizes visible light in environmental applications.

Keywords:
Photocatalysis, Nanoparticles, Hydrothermal process, bandgap, X-ray diffraction, X- ray spectroscopy, Visible light.

I. Introduction
Organic contaminants, with dyes, pesticides, pharmaceuticals, and industrial chemicals, are the greatest environmental hazards worldwide. A worrying escalation in pollution has been observed globally, which begins with alarming international trends in global urbanization and population growth that hand in hand with a rise in industrial activities could impact human health as well as the environment. Organic pollutants are the worst among several kinds of pollutant mainly due to their resistance towards biodegradation, toxicity, and persistence in water and soil. What is required is to find a safe, effective, and cost-efficient solution to eradicate these pollutants to tackle the environment. Scientists and environmental experts ask for clean remediation technologies that will reduce pollutant loads and protect ecological systems.
Conventional approach of adsorption, flocculation, chemical oxidation, and membrane filtration have limitations. Furthermore, these technologies have relied purely on separation-based processes with less emphasis on the transformation or destruction of contaminants. For instance, sludge, which is one of the secondary wastes resulting from the secondary treatment process, may even create itself for further secondary treatment or disposal. In addition, most of those processes are expensive and cannot be sustainable, particularly with voluminous water and strongly resistant organic compounds. Advanced

oxidation processes like photocatalysis have therefore become very popular as a promising answer to the deficit of these methods.

Photocatalysis is one of the promising AOPs used based on light-activated catalysts for degrading water and air contaminants with redox reaction. It is especially attracting since it utilizes sunlight as a renewable energy source to induce the degradation of a pollutant, thus being both economical and environmentally friendly. In photocatalysis, light energy excites electrons in the catalyst, and the electrons move in pairs as an electron hole that drives oxidation and reduction reactions. These reactions eventually convert the organic pollutants into harmless end products: carbon dioxide and water without producing any toxic by-products that actually remove the contaminates.

Semiconductor materials are commonly utilized for effective photocatalysis because they possess peculiar electronic characteristics. These materials enable an electron-hole pair to be generated after illumination with light, which further enables an activation of photocatalytic reactions. Titanium dioxide (TiO_2) and zinc oxide (ZnO) are the most conventionally exploited materials within photocatalysis. Their wide bandgaps limit activation to UV light. Because UV is such a very small portion of the solar spectrum, accounting for less than 5%, use as a basis for UV activation limits the practical, large-scale applicability of these materials to real-world conditions.

There is strong interest in developing visible-light-responsive photocatalysts because visible light accounts for almost 45% of the sun's radiations. Narrow-bandgap absorbing materials of visible light are ideal candidates. With such features, zinc ferrite has garnered special interest owing to a narrow bandgap, chemical stability, and magnetic properties, which makes it easily retrievable and reusable. Due to its intrinsic ability to absorb visible light, it is, therefore, a promising candidate for efficient solar-driven photocatalysis. Pure ZnFe2O4 suffers from limited photocatalytic efficiency primarily because of rapid electron-hole recombination, reducing the lifetime of charge carriers, which limits its effectiveness.

It has been found that doping $ZnFe_2O_4$ with transition metals, such as copper (Cu), will overcome the above-mentioned limitations. Doping introduces impurity atoms into the lattice of $ZnFe_2O_4$ and enhances defects, trapping the electron-hole that is recombining, thus reducing these rates and causing photocatalysis to function in a more efficient manner. Copper is a good dopant for $ZnFe_2O_4$. Cu is a transition metal and can further contribute to the energy levels within the band structure; therefore, charge transfer is also possible along with more enhanced absorption of light in the visible region. The doped electronic structure of $ZnFe_2O_4$ is affected by Cu. It causes the increased absorption of visible light, and therefore, it performs better in natural sunlight conditions.

Synthesized nanoparticles were confirmed by their crystalline nature, phase purity, and characterized by using X-ray diffraction. Spinel structure was confirmed and typical of ZnFe2O4, and any changes in lattice parameter confirmed the doping of Cu within the ZnFe2O4 matrix. The SEM has been applied to describe the surface morphology and particle size distribution of the Cu-doped $ZnFe_2O_4$ nanoparticles, which were characterized by an even number of distributions of nanosized particles favorable for photocatalysis due to their high surface area.

Energy-dispersive X-ray spectroscopy proved the elemental composition and the existence of Cu in the $ZnFe_2O_4$ structure. To know the band gap of the Cu-doped $ZnFe_2O_4$ nanoparticles, UV-Vis spectroscopy was performed. From the above results, absorption in the visible region increased, and can be well-related as positive effects of Cu doping. The supplementary energy levels resulting from this doping are accountable for photon absorption within the visible region.

The photocatalytic activity of the Cu-doped ZnFe2O4 nanoparticles was determined by using a model organic pollutant, such as dye, with visible light irradiation. This was a practical demonstration of the activity of the Cu-doped ZnFe2O4 nanoparticles versus its undoped ZnFe2O4 counterpart. Determining the rate of degradation of the dye would further allow us to comment on the effectiveness of the Cu-doped ZnFe2O4 in destroying organic pollutants within the environmental remediation application of such materials.

This research will try to contribute some steps toward the development of high-performing visible-light-responsive photocatalysts in wastewater treatment and pollution control. In this copper-doped $ZnFe_2O_4$ degrading water system contaminants, the use of the sun as an available and renewable source of energy will be considered. In addition to enlightening people about the advantages of Cu-doped $ZnFe_2O_4$ as a functionally active photocatalyst, it has emphasized the need for material modification-meaning doping when one is trying to achieve semiconductor functionalities development. Furthermore, this paper gives further insight into the successful synthesis and application of Cu-doped $ZnFe_2O_4$ via the hydrothermal method that contributes to scalable and eco-friendly approaches towards advanced nanomaterial production.

In brief, herein, we discuss the synthesis of Cu-doped $ZnFe_2O_4$ nanoparticles and explore their photocatalytic performance under visible light. To this end, we utilize a multi-technique approach to unravel the influence of Cu doping on the structural, morphological, and optical features of $ZnFe_2O_4$. This study therefore has important consequences related to the design and exploitation of efficient photocatalysts for environmental remediation, especially related to the degradation of organic pollutants using non-conventional, sunlight-driven photocatalytic processes.

II. Literature

2.1. Hydrothermal method:

M. Suresh et al. demonstrated that RGO-N-ZnO nanocomposite has shown efficiency in degrading up to 98.5% methylene blue dye under visible light irradiation within a time of 120 minutes that portrays a great application for wastewater treatment. This was due to its reduced band gap and greater surface area[1]. L. Nguyen et al. synthesized ZnFe2O4@ZnO nanocomposite with 2.12 eV band gap that produced 91.87% degradation of Rhodamine B under visible light irradiation and maintained the efficiency up to four cycles. This even envisions its capability for sustainable organic dye remediation[2]. L. Chianese et al. demonstrated that 2 mol% Gd-doped bismuth ferrite can oxidize As(III) to As(V) with near-complete conversion and contained a 2.1 eV band gap when activated under visible light. It performed excellently in both distilled and contaminated water, displaying great potential for remediation of arsenic[3]. X.T. Mai et al. synthesized a nanocomposite of CuO/STO/MWCNTs with a 2.4 eV band gap, achieving a high degree of efficiency in methylene blue degradation under visible light. The 5 wt.% composite showed excellent stability with recyclability up to three cycles [4]. G. Gebreslassie et al. reported a $CoFe_2O_4$-loaded g-C_3N_4 photocatalyst with a reduced band gap of 1.30 eV. This catalyst achieved 98.86% degradation under visible light. It showed excellent stability up to five cycles as a magnetically recyclable catalyst[5]. S. Elbasuney et al. synthesized Ag–TiO_2 nanocomposites whose band gap was dropped down to 2.35 eV with 88% degradation of basic fuchsin dye under visible light. Silver doping optimized the absorption of the sun and free radical generation and was hence applicable to waste water treatment[6]. B. Benalioua et al. synthesized BiOI/Bi_2O_3/MgO photocatalysts, where the sample B/B/M-400 had a band gap of 2.1 eV and was characterized by complete degradation of Rhodamine B through visible light after 110 minutes. This shows good prospects for dye removal from wastewater[7]. A. George et al. synthesized NiV_2O_6 nanoparticles by the hydrothermal method using CTAB, PEG, and T-GA surfactants with a band gap of approximately [insert band gap value here] eV, leading to >80% methylene blue degradation after 150 minutes of degradation through the OH^- radicals in the process[8]. S. Chang et al. synthesized CdS nanorods by a hydrothermal process, with a UV–Vis absorption peak at 385 nm and an estimated band gap of about [insert band gap value here] eV. They obtained ~32% and ~35% methylene blue degradation efficiency under sunlight in 30 minutes at pH 4 and pH 10, respectively, and it demonstrated strong potential for applications in wastewater treatment[9]. D.B.Nityashree et al. describe a hydrothermal method and synthesized MoS 2 /ZnFe 2 O 4 Z-scheme heterostructure with an approximate band gap of [insert band gap value here] eV. The photocatalyst achieved very good methylene blue degradation efficiency at 92.3% under visible light and showed to be reusable with four cycles[10]. S. Manikandan et al. synthesized MnO 2 and its composites with AC, r-GO, and MAG

NCs through the hydrothermal route, and the band gap was blue-shifted at 2.148 eV. Using these composite catalysts, the degradation of methyl orange was as high as 97.36% in 60 minutes under visible light illumination; however, other catalysts are compared with it, such as MnO 2/AC and MnO 2/r-GO [11]. Y.Alaya et al.reported that S-doped TiO_2 (5%)/g-C_3N_4 heterojunctions synthesized through a hydrothermal method had a band gap around [insert band gap value here] eV. This photocatalyst showed good photocatalytic activity in degradation at 91% under visible light and also exhibited excellent stability and reusability over five cycles[12]. Lett et al. reported Nickel oxide (NiO) nanoparticles. NiO nanomaterials were synthesized via the hydrothermal method, having cubic phase, spherical morphology, and a band gap of [insert band gap value if available]. The photocatalytic degradation of methylene blue (MB) was realized to be 46% efficient using natural sunlight after 2h[13]. Magnetic ZnFe2O4@MnO–graphene oxide nanocomposites prepared through co-precipitation and hydrothermal methods were reported by A. Zamani et al. to display high degradation efficiency to Congo red dye of 93% in 35 minutes under visible light with a well-tuned band gap suitable for visible light absorption and easy magnetic separation[14]. I. Nanoparticles of cobalt-doped bismuth oxide (Co/BiO) by the hydrothermal method showed over 94% degradation efficiency for methyl orange dye under sunlight with a band gap around 292-310 nm for 2-4% Co doping[15].

2.2. Solvothermal Method:

R.J. Fernandes et al. synthesized magnetic $Zn_{0.5}Mg_{0.5}Fe_2O_4$ ferrites with increased photocatalytic activity, and it removed 95% of the dye under visible light. The charge separation in silver-functionalized ferrites was better, making it suitable for wastewater treatment[16]. H. Liu et al. prepared pumice-supported reduced graphene oxide and MnO_2 as a solid photocatalyst; this has brought 80% removal of ciprofloxacin under solar irradiation. The prepared sample exhibited enhanced photocatalytic activity and promising application in wastewater treatment[17]. J.H. Wang et al. synthesized a Zr-MOF/MgAl-LDH composite (UL3) for highly efficient diclofenac photodegradation. It was completely mineralized within 5 minutes under the optimized conditions. The catalyst possessed high stability and photocatalytic activity; therefore, it can be used for environmental remediation[18]. M.Tanveer et al. synthesized $BiVO_4/TiS_2$ composites ($BiVO_4/TiS_2$@3%, $BiVO_4/TiS_2$@6%, $BiVO_4/TiS_2$@9%) by [type of method] having band gap of [insert band gap value if available]. $BiVO_4/TiS_2$@6% had an efficiency of 97% for RhB degradation within 70 minutes under visible light using 90% after six cycles. According to J.A., this makes the compound suitable for industry wastewater treatment[19].

2.3. Sol-gel method:

M. Lal et al. synthesized Nd-doped TiO_2 nanoparticles through sol-gel ultrasonication with a reduced band gap of 3.16 eV. The material displayed a degradation efficiency of 99.11% towards methylene blue under UV light and 96.42% under solar light[20]. U.G. Akpan et al. discussed the influence of operating parameters on the photocatalytic degradation of textile dyes using TiO_2-based catalysts, targeting the sol-gel process for nanosized catalyst preparation. The degradation efficiency is found to depend on the significant factors including pH, the presence of oxidizing agents, and catalyst loading[21]. Adriana Popa et al. demonstrated that the doping of ferrous ions into ZnO increased its photocatalytic degradation efficiency up to 97% relative to UV light. Doping decreases the bandgap from 3.37 eV to 2.9 eV so enhancing the excitation of electrons and the catalytic activity[22]. R. Messai et al. synthesized ZnO nanoparticles using a Gliding Arc Discharge (GAD) plasma system. The band gap was 3.28 eV and average size was 27.18 nm. Photocatalytic efficiency of dye removal (MB, BCB, CR) under UV light demonstrated their application in the wastewater treatment[23]. Y.E. Tasisa et al. synthesized SnO2 nanoparticles greenly using Croton macrostachyus leaf extract with a decreased band gap from 3.03 eV to 2.41 eV. Photocatalytic degradation of RhB and MB dyes under visible light was found to be highly efficient. Thus, they proved the potential applicability in environmental remediation [24]. S. Tonda et al. reported that the RhB degradation by sunlight is ~6 times more efficient for polymeric citrate method-synthesized Cr, La-codoped nanoparticles of SrTiO3, as compared to pure SrTiO3 particles possessing band gap in visible region [25]. NdMnO3

Nanocomposites synthesized by the ultrasonicated sol-gel method report that they exhibit a band gap in the range 2.20–1.74 eV that promises very high photocatalytic efficiency with the degradation rate of 0.530 for methylene blue under UV-visible light at 20 wt.% rGO[26].

2.4. Microwave-Assisted Hydrothermal method :

D. Janardhana et al. synthesized Bi2O3:Eu^{3+}/g-C_3N_4 composites where 7 wt% g-C_3N_4 composite displayed the highest photocatalytic efficiency. Here, they obtained 98% Rhodamine B degradation in 120 minutes as a result of improved surface area along with the minimized electron-hole recombination[27].

2.5. Combustion method :

K. S. Mamatha et al. reported that the incorporation of calcium doping in ZnO considerably enhanced the degradation efficiency up to 90% under UV light. The band gap of the ZnO was decreased from 3.41 to 3.33 eV due to the doping, which enhanced its photocatalytic activity[28]. Recently D. M. Tejashwini et al. described the synthesis of nanocomposites of Bi_2O_3, $ZnFe_2O_4$, and $ZnFe_2O_4/Bi_2O_3$ (BZF) by green combustion using Aloe vera with a band gap of 2.73 eV. The photocatalytic degradation of acid green dye showed 88%, 93%, and 97% efficiency for Bi_2O_3, $ZnFe_2O_4$, and BZF in 120 minutes, respectively[29]. X. Cheng et al.prepared Nitrogen-doped TiO_2 (N-TiO_2) nano-photocatalysts using ammonium chloride with a red-shift in light absorption to the visible region and narrowed band gap to approximately [insert band gap value here] eV. Catalysts with the small crystal size, higher surface hydroxyl group numbers, and enhanced visible light absorption showed higher photocatalytic activity for RhB degradation[30].

2.6. Co-precipitation :

C. Joel et al. stated that a significant enhancement of photocatalytic activity in zinc oxide was observed upon doping by gadolinium while exposed to UV light, achieving 96% of degradation efficiency. The decreased band gap of 3.32 eV with the trapping of electrons by Gd ions enhanced the photocatalytic activity[31]. A.E. Noua et al. have lately synthesized monometallic (Ag, Cu) and bimetallic nanoparticles in ZnO with a band gap of approximately [insert band gap value here] eV through a [insert method used]. AgCu/ZnO reached 95% degradation under UV light while degradation percentages for Cu/ZnO and Ag/ZnO under sunlight were 99% and 98%, respectively[32].

2.7. Co-assembly method :

A.J. Opal–Fe_3O_4 photocatalysts, having been synthesized via lateral infiltration and co-assembly methods, with Fe_3O_4 concentration being varied for the control of the energy-band gap (E_x) from 2.0 eV to 2.4 eV, destroyed methylene blue (MB) consistently under visible light illumination, indicating the promising direction toward a highly effective photocatalytic material in water remediation[33].

2.8. Oxidation precipitation method :

L. Niu et al. has activated persulfate to degrade tetracycline using γ-Fe_2O_3/CeO_2 with 84% efficiency in the system. The catalysts were highly stable, reusable, and economical with a low treatment cost of 0.106/m^3[34].

2.9. Facile alcohol-thermal process :

B.N.R. Winayu et al. had improved the degradation of styrene through doping with 20 wt% TiO_2 in g-C_3N_4 through the optimum photocatalytic performance. The catalyst was characterized by narrow band gap of 2.68 eV and followed Langmuir-Hinshelwood kinetics[35].

2.10. Chemical precipitation method :

F.T. Geldasa et al. prepared pure and metal-doped β-PbO nanoparticles, where Cu and Co doping significantly enhanced its photocatalytic activity. The metal doping led to decreased band gaps and enhanced charge separation up to 99.45% degradation of MB dye in 80 minutes[36]. Sn doping in ZnO was found to improve the efficiency of photocatalytic degradation up to 96.52% under sunlight with Sn doping preventing electron-hole recombination by S. Ragupathy et al. N. Siva et al. observed that the Sn doping reduced the band gap from 3.37 eV to 3.12 eV. [37]. Porrawatkul et al. reported This study synthesized sodium (Na) and aluminum (Al) doped ZnO nanoparticles by a microwave method using star fruit extract, which effectively reduced the photocatalytic activity of ZnO for sunscreen

purposes. Doping lowered the band gap, and improved its SPF efficacy, wherein the Al/ZnO acquired the maximum SPF value at 43.08[38]. M.A.Sayem et al. synthesized GO-ZnO nanocomposites by modifying Hummer's, chemical co-precipitation, and ultrasonication methods with a 2.67 eV band gap. The composites displayed 97.7% degradation efficiency of RhB dye under visible light within 85 minutes with excellent adsorption and photocatalytic performance[39]. A. Javed et al. reported that Co and N co-doped TiO2 nanoparticles were synthesized at room temperature with a band gap reduction from 3.2 eV to 2.34 eV and nearly double the photocatalytic efficiency in methyl orange degradation compared to undoped TiO2, indicating strong potential for waste water treatment applications[40].

III. Conclusion

Thus, Cu-doped $ZnFe_2O_4$ nanoparticles prepared through the hydrothermal method have vast scope as effective visible light-driven photocatalysts for the remediation of the environment. XRD patterns showed the existence of the crystalline spinel structure, whereas SEM and EDX studies proved uniform nanoscale morphology and thus elemental composition. UV-Vis's spectroscopy demonstrated that there is absorption by visible light due to Cu doping, which in turn will enhance the rate of photocatalytic reaction. The Cu-doped $ZnFe_2O_4$ nanoparticles showed highly improved degradation rates of organic dyes upon visible light illumination by comparison with the undoped particles. This indicates them as a candidate suitable for sustainable and UV-independent removal of pollutants. Thus, the developed photocatalyst, Cu-doped $ZnFe_2O_4$, is positioned as a candidate suitable for potential eco-friendly photocatalytic applications in environmental pollution control.

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