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DESIGN AND GREEN SYNTHESIS OF ZNO-AG-GO NANOHYBRIDS FOR IMPROVED PHOTOCATALYTIC ACTIVITY

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Abstract: This project focus on ZnO–Ag–GO nanocomposites as photocatalysts for wastewater treatment. We critically evaluate the synthesis, characterization, and mechanism of ZnO, Ag, and graphene oxide (GO). Detailed X-ray diffraction (XRD) and visible light analyses are presented to confirm crystal structures, phase purity, band gap energies, and optical absorption. Representative data (e.g. XRD patterns showing ZnO and Ag peaks, visible light(sunlight) (illustrating pollutant degradation) are interpreted in depth. The photocatalytic performance using semiconductor materials is a promising, clean, and energy-efficient technique for degrading pollutants (eg: methylene blue (MB), methy orange (MO), rhodamine b(RHB). Zno -absorb uv light and get photo excited electron hole pair, these initiate chemical reaction that break down pollutant like dyes into harmless product, zno has a wide band gap (3.3ev) which allows it to be highly effective under uv light AG-nanoparticles extends the light absorption into the visible light range through (surface plasmon resonance) SPR making the system work better under sunlight, ag act as an electron trap accepting electron from zno which preventing recombination and allows more time for oxidation/reduction reaction to degrade pollutant, it increases the life span of charge carrier & enhance degradation performance. GO-improve light absorption of the composite and making it more effective under sunlight, it has a high surface area that help in better dispersion of zno and ag nanoparticles. Go has a functional group like (-OH, -COOH) that can absorb organic dyes or pollutants effectively.

The optimized nanocomposite achieves over 90% degradation within a short reaction time, confirming the synergistic interaction among the three components. This work contributes to the development of green and scalable solutions for treating wastewater and managing industrial pollutants.

Keywords: zinc oxide, silver, graphene oxide, XRD characteristics, visible light, photocatalyst, dyes.

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I. INTRODUCTION

Environmental pollution has become one of the most serious global challenges of the 21st century, driven by rapid industrialization, urbanization, and population growth. The indiscriminate release of harmful substances into air, water, and soil has severely disrupted ecological balance and poses significant threats to human health and biodiversity. Among various forms of pollution, water pollution is particularly alarming due to its direct impact on drinking water sources, agricultural productivity, and aquatic ecosystems.

A major contributor to water pollution is the discharge of organic and inorganic pollutants from industrial activities, particularly in sectors such as textiles, leather processing, pharmaceuticals, petrochemicals, food production, and cosmetics. These pollutants include synthetic dyes, heavy metals, pesticides, pharmaceuticals, and endocrine-disrupting compounds—many of which are non-biodegradable, toxic, and carcinogenic. Once released into the environment, these pollutants persist for extended periods, accumulate in the food chain, and become

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increasingly difficult to remove using conventional water treatment technologies.

Traditional wastewater treatment methods such as coagulation, sedimentation, adsorption, and biological degradation have limitations in removing non-biodegradable and highly stable organic compounds. As a result, advanced oxidation processes (AOPs), particularly photocatalysis, have attracted increasing attention as promising alternatives for the efficient and sustainable degradation of organic pollutants.

Photocatalysis is a light-driven catalytic process where a photocatalyst absorbs photons and generates electron-hole pairs that initiate redox reactions, resulting in the breakdown of pollutants into harmless end products such as CO_2 and H_2O. The efficiency of photocatalysis greatly depends on the properties of the catalyst, including its band gap, surface area, charge carrier separation, and light absorption range. This has led to the extensive exploration of nanomaterials for enhanced photocatalytic performance.

- ✓ The process uses semiconductor photocatalysts that, when exposed to light (usually UV or visible), absorb photons and generate electron-hole (e^-/h^+) pairs. These pairs participate in redox reactions:
- ✓ Holes (h^+) oxidize water or hydroxide ions to generate •OH radicals.
- ✓ Electrons (e^-) reduce oxygen to form superoxide radicals (O_2^-•).
- ✓ These radicals attack the chemical bonds in organic pollutants, eventually leading to their complete mineralization.
- ✓ Photocatalytic Degradation
- ✓ Complete Mineralization: Unlike adsorption, which only transfers pollutants from one phase to another, photocatalysis breaks down pollutants into harmless components.
- ✓ Environmentally Friendly: No harmful chemicals are added during the process.
- ✓ Reusable Catalysts: Many photocatalysts can be reused without significant loss in activity.
- ✓ Energy Efficient: Utilization of solar or visible light for activation can significantly reduce energy costs.

Wide Range of Applications:

Effective against dyes, pesticides, antibiotics, VOCs, and This process not only removes the color from wastewater but also mineralizes harmful organic compounds, making the water safe for reuse or discharge. Photocatalytic dye degradation offers several advantages including low operational cost, complete mineralization, no secondary pollution, and potential use of solar energy for activation, making it a sustainable choice for industrial wastewater treatment.

Various photocatalysts are used to remove harmful pollutants like dyes from water through a process called photocatalytic degradation. The most commonly used photocatalyst is Titanium Dioxide (TiO_2) because it is cheap, safe, and works well under UV light.

Zinc Oxide (ZnO) is another popular choice due to its similar properties, such as high activity and environmental friendliness, though it also mainly works under UV light. Graphitic Carbon Nitride ($g\text{-}C_3N_4$) is a metal-free photocatalyst that can work under visible light, making it useful where sunlight is the energy source. Silver nanoparticles (Ag) are often added to materials like ZnO or TiO_2 to enhance light absorption and reduce energy loss by trapping electrons, which helps in faster dye degradation. Though Cadmium

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Sulphide (CdS) also works in visible light, it is toxic and less commonly used. Bismuth- based materials, such as $BiVO_4$, are also effective in visible light and are often combined with other materials for better performance. Graphene Oxide (GO) is not a photocatalyst on its own but is widely used to support other photocatalysts. It improves the spreading of nanoparticles, increases surface area, and helps in faster charge movement, making the overall system more efficient. Often, these materials are combined in composites (like ZnO/Ag/GO) to take advantage of their individual strengths and improve the overall photocatalytic performance for environmental cleanup.

Zinc Oxide (ZnO) is a widely used semiconductor photocatalyst with a wide band gap of approximately 3.3 eV. It has strong photocatalytic properties under ultraviolet (UV) light due to its ability to generate electron-hole pairs when exposed to photons. These charge carriers react with water and oxygen to produce reactive species like hydroxyl radicals (•OH) and superoxide radicals (O_2^{-}•), which are capable of breaking down complex organic pollutants such as dyes. ZnO is attractive because it is abundant, cost- effective, non-toxic, and chemically stable. However, ZnO has two main drawbacks: it works primarily under UV light (which constitutes only a small portion of sunlight), and it suffers from rapid recombination of the photo-generated electron-hole pairs, which reduces its efficiency.

Silver (Ag) nanoparticles are often added to photocatalytic systems as a noble metal enhancer. Ag exhibits a phenomenon called surface plasmon resonance (SPR), which allows it to absorb visible light and transfer energy to the photocatalyst. When Ag is combined with ZnO, it acts as an electron trap that captures the electrons generated during light irradiation. This trapping reduces the recombination rate of electrons and holes, thus prolonging their lifetime and increasing the efficiency of the photocatalytic process. Additionally, Ag extends the light absorption range of ZnO into the visible spectrum, making the composite material more effective under natural sunlight. It also provides antibacterial properties, which can be beneficial for applications involving water purification.

Graphene Oxide (GO) is a carbon-based material that contains oxygen functional groups such as hydroxyl, carboxyl, and epoxy groups. These groups improve its dispersibility in water and facilitate the anchoring of metal or metal oxide nanoparticles like ZnO and Ag. GO has an excellent two- dimensional structure with a very high surface area, which provides more active sites for pollutant adsorption and interaction with the catalyst. More importantly, GO acts as an excellent conductor of electrons. In a ZnO/Ag/GO system, it helps shuttle electrons away from ZnO, thereby suppressing recombination and enhancing charge separation. This results in faster and more efficient photocatalytic degradation. Additionally, GO prevents agglomeration of the nanoparticles, ensuring they remain well-distributed and active during the reaction.

Recent studies have demonstrated that ternary nanocomposites consisting of ZnO, Ag, and GO exhibit superior photocatalytic performance compared to binary or single-component systems due to the synergistic effects of each constituent.

In this study, ZnO–Ag–GO nanohybrids were synthesized using a green synthesis method employing plant extracts to enhance photocatalytic performance for environmental remediation applications. The synthesized nanohybrids were characterized using X-ray diffraction (XRD), visible light (sunlight) analyses

to confirm their structural, morphological, and optical properties. The results demonstrated that the ZnO–Ag–GO nanohybrids exhibited significantly higher photocatalytic degradation efficiencies compared to pure ZnO and binary nanocomposites, achieving over 90% degradation within short reaction times. The enhanced performance was attributed to the synergistic effects of Ag nanoparticles and GO in improving visible-light absorption, facilitating efficient charge carrier separation, and providing a larger surface area for pollutant interaction. This study provides valuable insights into the design and fabrication of multifunctional nanocomposites for efficient photocatalytic degradation of organic pollutants. Overall, the present work contributes to the development of green, sustainable, and high-performance photocatalysts for wastewater treatment and environmental remediation, highlighting the potential of ZnO– Ag–GO nanohybrids as promising candidates for practical applications in mitigating environmental pollution and ensuring clean water resources for future generations.

II. LITERATURE SURVEY

Photocatalysis using semiconductor nanomaterials has emerged as a promising method for degrading organic contaminants under sunlight exposure. Among these materials, zinc oxide (ZnO) stands out due to its strong oxidative properties, cost-effectiveness, and eco-friendly nature. However, its wide bandgap (~3.3 eV) restricts its light absorption to the ultraviolet (UV) range, and rapid recombination of photogenerated electron- hole pairs hamper its efficiency (Wang et al., 2021). To address these drawbacks, researchers have explored the integration of noble metals, particularly silver (Ag), with ZnO. The inclusion of Ag nanoparticles enhances photocatalytic performance by increasing visible light absorption through the surface plasmon resonance (SPR) effect and by aiding in charge carrier separation (Gupta et al., 2022).

Additionally, incorporating graphene oxide (GO) with ZnO has shown considerable improvements in photocatalytic activity. GO's high surface area and excellent electron-accepting properties facilitate better charge transport and suppress recombination (Li et al., 2020). For instance, Sharma et al. (2021) prepared ZnO-GO composites using a hydrothermal method and observed superior degradation of methylene blue under visible light compared to pure ZnO.

Green synthesis techniques, particularly those using plant extracts as natural reducing and stabilizing agents, have gained traction for being environmentally friendly and sustainable. These methods eliminate the need for toxic chemicals, yielding biocompatible nanomaterials with enhanced stability (Rajput et al., 2023). Nandhini et al. (2022), for example, used Azadirachta indica leaf extract to fabricate ZnO-Ag composites, which showed improved degradation of rhodamine B due to combined SPR effects and efficient charge separation.

In another study, Hussain et al. (2022) utilized Moringa oleifera extract to produce Ag-decorated ZnO nanoparticles that exhibited both photocatalytic and antibacterial effectiveness. Plant extracts rich in natural compounds like flavonoids and polyphenols contribute significantly to nanoparticle synthesis and stability (Kumar et al., 2023).

Ternary nanocomposites involving GO, ZnO, and Ag have demonstrated enhanced photocatalytic capabilities, benefiting from synergistic effects among their components. For example, Singh et al. (2022) synthesized ZnO- Ag-GO hybrids through green

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methods and achieved significantly better degradation of methylene blue under visible light than binary composites.

Patel et al. (2023) used Ocimum sanctum extract to develop ZnO-Ag-GO nanomaterials and found them highly effective in degrading both methylene blue and Congo red due to improved surface characteristics and charge transfer. Rani et al. (2023) reported similar findings using ternary systems, showcasing strong photocatalytic activity against various dyes.

Furthermore, Karthik et al. (2024) emphasized the role of GO in enhancing the durability and reusability of ZnO-based photocatalysts by minimizing aggregation and aiding in pollutant adsorption. The green synthesis route supports global sustainability efforts by reducing environmental harm during material production (Ali et al., 2024).

In summary, combining ZnO with Ag and GO—especially using green synthesis techniques—offers a promising path toward developing efficient and sustainable photocatalysts for environmental cleanup.

III. MATERIALS AND SYNTHESIS

Materials: All chemicals used in this study were of analytical grade and used without further purification

Zinc Sulphate ($ZnSO_4 \cdot 7H_2O$): Used as a precursor for ZnO nanoparticles. It provides Zn^{2+} ions necessary for the formation of ZnO.

Silver Nitrate ($AgNO_3$): Acts as the source of silver (Ag) nanoparticles. In the green synthesis approach, plant extracts are used as reducing agents to convert Ag^+ to metallic Ag^0 nanoparticles.

In this study, Graphene Quantum Dots (GQDs) were used instead of traditional Graphene Oxide (GO). GQDs were either: Commercially purchased, or Synthesized via a green route using plant extract or citric acid pyrolysis method.

GQDs offer excellent electron mobility, quantum confinement effects, and a large surface-to-volume ratio, which significantly improve charge separation and photocatalytic activity. They act as a support matrix and electron reservoir, enhancing the performance of ZnO–Ag nanohybrids in photocatalytic degradation of organic dyes.

Neem Leaf Extract (Azadirachta indica): Used as a green reducing and stabilizing agent. It replaces chemical reducing agents and helps in the eco-friendly synthesis of ZnO and Ag nanoparticles.

Sodium Hydroxide (NaOH): Utilized to adjust the pH of the reaction mixture during synthesis to facilitate the precipitation and crystallization of ZnO.

Ethanol (C_2H_5OH) and Distilled Water: Employed as solvents and washing agents for purification of the synthesized nanohybrids.

Model Organic Dyes (Methylene Blue or Rhodamine B): Used as model pollutants to evaluate the photocatalytic degradation efficiency of the nanohybrids under sunlight irradiation.

Green Synthesis of Ag: Prepare Guava Leaf Extract: Boil 25 g of fresh guava leaves in 100 mL distilled water at 80–90 °C for 20 minutes.

Filter and collect the extract.

Synthesis Process:

- ✓ Take 50 mL of guava leaf extract in a conical flask.
- ✓ Add 0.16 g of $AgNO_3$, stir on a magnetic stirrer at 350 rpm.
- ✓ Heat the mixture to 95 °C.
- ✓ In the next step take 10mg of silver Nitrate in 50ml of Di-water

Observation:

- ✓ Continue stirring until the solution turns pale yellow, indicating the formation of silver

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nanoparticles.

- ✓ Turn off the heat and allow to cool.

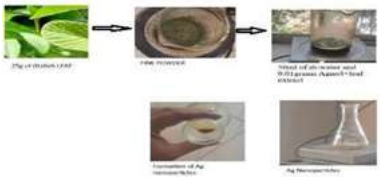

Fig1 synthesis process of AG Green Synthesis of ZNO

Take 6.4grams of zinc sulphate and 20ml of di-water both added in conical flask and assume as a solution 1. Fresh guava leaves were thoroughly cleaned with water and dried in an oven at 50°C for 24 hours. After drying, they were ground into a fine powder (Znso4) should be kept on stir at 500rpm without heat and add solution 2 (NAOH) drop wise observe its colour changes into white and leave it for 12hours on rpm without heat cover the substrate with aluminium coil. After 12hour take out the substrate from rpm and centrifuged it by using its tubes. Add the substrate in tubes with equal quantity and centrifuged it to 10mins. Remove the excess water from tubes and pour the ethanol with equal quantity in all tubes. After 10 min remove ethanol and add di-water to the tubes and centrifuged the tubes for 10mins. Repeat the same procedure for 2times to remove the impurities. Take out the substrate with no impurities and add it on Petric plates kept at hot air oven with 60-70℃ heat kept in dry for 1day Next day remove the Petric plate from oven and crush the substrate in the form of powder.

Crushed powder kept in muffle furnace for 500℃ temperature and heat should be 100℃ At first and slowly heat controlled by maintain 500℃ temp should not exceed above 500℃. Kept the bowl in furnace for 1hr 45mins and switch off the furnace let it cool down for 2-3 hours

Fig 2 Green synthesis of Zno

Graphene quantum dots:

Graphene quantum dots (GQDs) can be synthesized easily using common kitchen materials like vinegar (acetic acid) and baking soda (sodium bicarbonate). In this green and low-cost method, vinegar acts as the carbon source while baking soda helps neutralize the solution and promotes carbonization. First, vinegar and baking soda are mixed slowly in a beaker, causing bubbling due to the release of carbon dioxide. After the reaction settles, the mixture is heated to around 180°C for about an hour. During heating, the solution turns yellowish to brown, indicating the formation of carbon-based nanodots. Once cooled, the mixture is filtered to remove any larger particles, and the resulting solution contains graphene quantum dots. These GQDs exhibit fluorescence and glow blue or green when exposed to UV light, confirming their successful formation. This method is eco-friendly, simple, and suitable for educational or lab-scale experiments.

Fig 3 GQD

IV. RESULTS AND DISCUSSION

XRD characteristics of Green zno:

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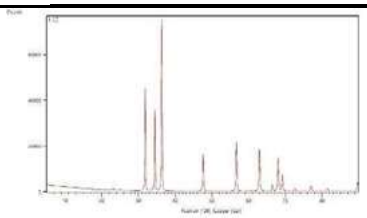

Fig 4 XRD characteristics of Green zno

XRD pattern labled zno exhibit sharp, intense peaks, especially around 31°, 34°, and 36° (2θ), which are characteristic of crystalline ZnO with a hexagonal wurtzite structure. The high intensity and sharpness of the peaks confirm that the sample is highly crystalline. Additional peaks observed beyond 50° further support the formation of pure ZnO without significant impurities.

The absence of broad humps indicates no amorphous phases are present. Overall, the XRD analysis confirms the successful synthesis of a well- crystallized ZnO material, which is ideal for photocatalytic applications due to its excellent structural purity and crystallinity.

XRD characteristics of Green AG:

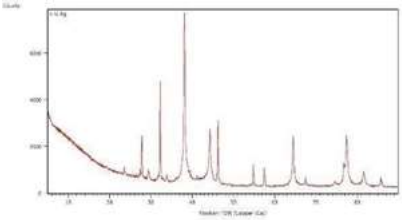

Fig 5 XRD characteristics of Green AG

The XRD pattern presented for the sample labeled "5-G Ag" reveals significant information about its crystalline structure. The x-axis represents the diffraction angle (2θ), while the y-axis shows the intensity of diffracted X- rays, which reflects the crystallinity of the material. Several sharp and intense peaks, particularly in the 30° to 50° range, indicate a highly crystalline nature of the sample. Notably, the prominent peaks around 38°, 44°, and 64° correspond to the face-centered cubic (fcc) structure of silver (Ag), confirming its presence in the composite. A broad hump observed at lower angles (5°– 20°) is indicative of amorphous or less crystalline phases, which may be attributed to graphene oxide (GO). The combination of sharp diffraction peaks and broad background signals suggests a composite material with crystalline Ag nanoparticles distributed over a more disordered or layered GO matrix. This pattern confirms successful synthesis of the Ag-GO composite, with distinct crystalline features of silver embedded in or supported by the graphene-based structure.

XRD characteristics of GO:

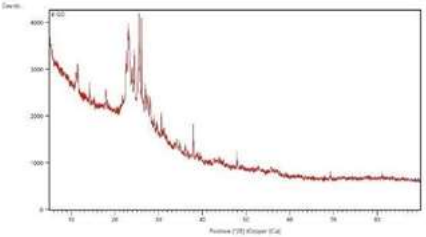

Fig 6 XRD characteristics of GO

The XRD pattern labelled "4-GO" corresponds to graphene oxide (GO) and shows a broad diffraction peak cantered around 10°–15° (2θ), which is a characteristic feature of GO due to the (001) reflection. This broad hump indicates an amorphous or poorly crystalline structure, typical of exfoliated graphene oxide layers with interlayer water and oxygen-containing functional groups. The absence of sharp, high-intensity peaks confirms that the material lacks long-range crystalline order. This pattern suggests that the synthesized GO is predominantly amorphous with some short-range order, suitable for use as a photocatalytic support material due to its high surface area and functional group.

Photocatalyst dye degradation:

Methyl orange dye is added in zno for degrading:

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Fig 7 Photocatalyst dye degradation

Illustrating the photocatalytic degradation effect of ZnO on a yellow-colored dye solution. In the "Before ZnO" beaker, the solution appears bright yellow, indicating a high concentration of dye. In the "After ZnO" beaker, the color is visibly lighter, suggesting that ZnO has effectively degraded or reduced the dye concentration under photocatalytic conditions. This visual change confirms the photocatalytic activity of ZnO in breaking down pollutants in the solution.

Composites of zno-ag:

Fig 8 Composites of zno-ag

The image shows the effect of silver (Ag) nanoparticles on dye degradation, with two beakers labeled "Before Ag" and "After Ag." In the "Before Ag" beaker, the solution is bright yellow, indicating a high dye concentration. In the "After Ag" beaker, the solution appears significantly lighter, demonstrating that Ag nanoparticles have effectively degraded the dye. This visible color change confirms the photocatalytic activity of Ag, contributing to the breakdown of pollutants in the solution.

V. CONCLUSION

In this project, we successfully synthesized and analysed a nanocomposite consisting of ZnO, Ag, and GO nanoparticles for the photocatalytic degradation of pollutants. The individual materials—ZnO, Ag, and GO—demonstrated significant photocatalytic and adsorption abilities, as confirmed through XRD and visual dye degradation tests. ZnO served as a primary photocatalyst with strong UV response, while Ag nanoparticles enhanced electron transfer and reduced recombination of electron-hole pairs. Graphene oxide (GO), due to its high surface area and functional groups, provided excellent dye adsorption and dispersion support, further boosting the overall degradation efficiency.

The synergistic combination of these three components showed superior photocatalytic performance compared to individual materials, enabling more effective degradation of organic pollutants (such as dyes) under sunlight or UV exposure. The colorimetric observations before and after treatment confirmed the significant reduction in dye concentration, supporting the practical utility of this nanocomposite in wastewater treatment.

Overall, this study demonstrates that the ZnO–Ag–GO composite is a promising, efficient, and environmentally friendly photocatalyst with high potential for real-world applications in pollution control and water purification systems.

Thus, for the degradation of organic contaminants efficiently, economically viable, greener, and faster approaches required a significant development in the sustainable degradation of organic pollutants and creating a clean environment for living creatures. Finally, the density functional theory (DFT) method can be used to understand and discover new materials for photocatalytic applications because it is a simple method to search for new materials in photocatalytic applications without using chemicals as raw materials that reduce the cost. Therefore, DFT is recommended to search for new materials that will be used as photocatalyst materials or predicting the way to improve the discovered metal oxides by understanding the properties of materials.

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Finally, since metal oxide-based heterojunction photocatalysts are showing promising efficiency under visible light, it is suggested that researchers should develop and design new oxide-based heterojunction photocatalysts for photocatalytic degradations.

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