

#### Green-Synthesized ZnO-CuO Nanocomposites for Photocatalytic Degradation of Pharmaceutical Pollutants in Wastewater

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## Abstract

Pharmaceutical pollutants such as antibiotics and analgesics represent a significant challenge in modern wastewater treatment due to their persistence, bioaccumulation potential, and resistance to conventional remediation techniques. This study investigates the green synthesis of zinc oxide–copper oxide (ZnO–CuO) nanocomposites using *Azadirachta indica* (neem) leaf extract and their application for the photocatalytic degradation of pharmaceutical contaminants under visible light irradiation. Characterization using XRD, SEM, UV-Vis spectroscopy, and FTIR confirmed the successful formation and stability of the nanocomposites. The photocatalytic tests, using diclofenac as a model pollutant, revealed a degradation efficiency of 92% within 90 minutes. Furthermore, the nanocomposites demonstrated high reusability, retaining over 85% activity after five cycles. These findings highlight the environmental and technological promise of green nanomaterials for sustainable water purification applications.

**Keywords:** Green synthesis; ZnO–CuO nanocomposites; Photocatalysis; Pharmaceutical pollutants; Wastewater treatment

### **Graphical Abstract**

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## 1. Introduction

Pharmaceutical residues—including non-steroidal anti-inflammatory drugs (NSAIDs), antibiotics, and anticonvulsants—are increasingly detected in aquatic systems, often at concentrations ranging from ng/L to  $\mu$ g/L due to inadequate removal in conventional wastewater treatment plants [1,2]. Diclofenac, a widely used NSAID, has been listed by the World Health Organization as a high-priority emerging contaminant due to its persistence, endocrine-disrupting potential, and ecological toxicity [3].

Recent advances have explored semiconductor-based photocatalysis as a robust alternative to traditional treatment methods [4]. ZnO, with its high electron mobility and redox potential, and CuO, with its narrow bandgap energy, have individually shown promise. However, their limitations—such as rapid electron-hole recombination and limited visible light absorption—necessitate composite strategies. ZnO–CuO heterojunctions offer improved photocatalytic efficiency due to enhanced charge separation and extended light absorption [5].

#### Research Gap.

While previous studies have demonstrated the potential of ZnO–CuO systems, most rely on chemical synthesis methods involving hazardous solvents or energy-intensive steps [6,7]. These approaches contradict green chemistry principles and hinder scalability and environmental compliance.

#### Novelty Statement.

This study is the first to synthesize ZnO–CuO heterojunction nanocomposites using a single-step, plant-mediated approach with *Azadirachta indica* extract, enabling dual-functionality as both reducing and stabilizing agent. The green-synthesized nanocomposites exhibited high photocatalytic efficiency and reusability, outperforming many chemically synthesized counterparts.

#### Paper Organization.

The remainder of this paper is structured as follows: Section 2 outlines the materials, green synthesis route, and characterization techniques. Section 3 presents the results, including structural features, photocatalytic efficiency, kinetic analysis, and mechanistic insights. Section 4 concludes the study and outlines future research directions.



**Special notice:** Figures and Tables used in the main body should not exceed a total of nine (09). Any additional figures or tables should be moved to the supplementary material and labelled accordingly (e.g., Table S1, Figure S1). Their presence should be stated in the main text for clarity

## 2. Materials and Methods

### 2.1 Materials

All chemicals, including zinc nitrate hexahydrate, copper sulfate pentahydrate, and diclofenac sodium, were of analytical grade and procured from certified suppliers. Neem leaves were collected from the University of Lagos campus.

### 2.2 Green Synthesis of ZnO–CuO Nanocomposites

Fresh neem leaves were washed, air-dried, and boiled in distilled water to extract bioactive compounds. The filtered extract was added dropwise to a mixed solution of Zn and Cu salts under constant stirring. The pH was adjusted to 9 using NaOH. The resulting precipitate was aged, filtered, washed, and calcined at 400°C.

#### 2.3 Characterization

X-ray diffraction (XRD) was performed to determine the crystalline structure. Scanning electron microscopy (SEM) was used for morphology. UV-Vis spectroscopy provided optical absorption data, while FTIR analysis confirmed the presence of functional groups from plant phytochemicals.

#### 2.4 Photocatalytic Degradation Study

A 10 ppm diclofenac solution was irradiated with visible light in the presence of the catalyst (0.5 g/L). Aliquots were sampled at regular intervals and analyzed via UV-Vis spectrophotometry at 276 nm.



### 2.5 Reusability Test

The catalyst was recovered, washed, and reused across five degradation cycles to assess its stability.

### 3. Results and Discussion

### 3.1 Structural and Morphological Characterization

XRD patterns revealed distinct peaks for ZnO and CuO phases, confirming nanocomposite formation. SEM images showed quasi-spherical particles with average sizes below 50 nm. FTIR spectra exhibited bands corresponding to O-H, C=O, and M-O bonds, indicating capping by phytochemicals.

#### **3.2 Photocatalytic Activity**

The nanocomposites achieved 92% degradation of diclofenac in 90 minutes. The photocatalytic performance was attributed to the enhanced charge separation and broadened light absorption of the heterojunction system.

#### 3.3 Reusability and Stability

The catalyst maintained above 85% efficiency after five cycles, indicating good stability. Minimal changes in structural morphology were observed via post-reaction SEM analysis.

#### **3.4 Mechanistic Insights**

The degradation pathway likely involves reactive oxygen species (ROS) generation, primarily  $\cdot$ OH and O2 $\cdot$ -, which attack the diclofenac molecule. The green synthesis not only reduces environmental burden but also introduces surface functionalities that may enhance pollutant adsorption.



## 4. Conclusion

This study successfully demonstrated the green synthesis of ZnO–CuO nanocomposites using *Azadirachta indica* leaf extract. The materials exhibited high photocatalytic degradation efficiency and excellent reusability. Their eco-friendly synthesis and effectiveness suggest strong potential for application in sustainable wastewater treatment technologies.

## Ethics and Reproducibility

This research did not involve human or animal subjects. All experimental methods were performed in accordance with relevant guidelines and regulations. Data and analytical codes are available upon request. The authors commit to open data and reproducibility standards endorsed by JCAS.

## **Conflict of Interest**

The authors declared that there is no conflict of interest.

## **Author's Declaration**

All authors have contributed significantly to the conceptualization, methodology, analysis, and writing of this manuscript. All authors have read and approved the final version. The manuscript is original, has not been published elsewhere, and is not under consideration by any other journal and will accept all liability for any claims about the content.

#### Acknowledgments

### References

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[2] World Health Organization. (2020). Pharmaceuticals in drinking water: *A global overview*. <u>https://www.who.int/water\_sanitation\_health/publications/pharmaceuticals-in-drinking-water</u>

### **Supplementary Materials**



Supplementary files include raw UV-Vis spectra, MATLAB scripts for kinetic modeling, and high-resolution images. These materials are submitted as a compressed ZIP file and referenced in the main text as Supplementary Figures S1 to S3.

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