Eco-Friendly Synthesis of CuO–Supported Carbon Nanocomposites for Rapid Catalytic Degradation of Methylene Blue

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Abstract

Copper oxide (CuO) nanoparticles were successfully synthesized via a green synthesis route using Vitex negundo Linn leaf extract as a reducing and stabilizing agent. The synthesized nano-CuO was impregnated onto carbon material derived from the stem of Chromolaena odorata to obtain CuO–carbon (CuO–C) nanocomposites. The prepared materials exhibited excellent catalytic performance in the degradation of methylene blue (MB) dye at room temperature. Various concentrations of CuO–C were investigated to optimize the CuO loading for maximum degradation efficiency. The characteristic absorption peaks of MB at 244 nm, 291 nm, and 663 nm disappeared completely after treatment with the CuO–C composite, confirming the effective degradation of the dye. The 5% CuO–C nanocomposite material showed the highest degradation efficiency, attributed to the synergistic interaction between CuO nanoparticles and the carbon support, which enhanced charge transfer and reduced recombination. Additionally, the effect of thermal activation at 50 °C was examined, revealing temperature-dependent catalytic behavior. The adopted synthesis approach is simple, eco-friendly, and cost-effective, demonstrating the potential of CuO–C nanocomposites as efficient catalysts for the removal of organic dyes from wastewater

***Keywords:** CuO nanoparticles; Carbon supported catalyst; Green Synthesis; Catalytic Activity; Dye Degradation*

1. Introduction

Water pollution caused by synthetic dyes is one of the most serious environmental problems faced by industrialized societies today. Dyes such as MB are extensively used in textile, paper, and plastic industries, and their improper discharge into water bodies leads to severe ecological and health hazards due to their toxicity, chemical stability, and resistance to biodegradation. [1&2] Conventional treatment methods—such as adsorption, coagulation, and biological degradation—often prove insufficient for complete mineralization of these persistent organic pollutants. As a result, photocatalysis has emerged as a promising, sustainable, and efficient technique for dye degradation under mild environmental conditions. [3] Among various photocatalysts, copper oxide nanoparticles (nano-CuO) have attracted substantial interest due to their narrow band gap (~1.2–1.9 eV), low cost, abundance, and strong redox properties, making them highly suitable for visible-light-driven photocatalysis. [4&5] However, the practical application of nano-CuO is often limited by their tendency to aggregate, leading to reduced surface area and photocatalytic efficiency. To overcome these drawbacks, carbon-based supports, such as activated carbon (AC), biochar, and graphene derivatives, have been widely employed as stabilizing matrices to improve nanoparticle dispersion, increase adsorption capacity, and facilitate charge transfer during photocatalysis. [6&7] Recent studies have demonstrated that modifying activated carbon with transition metal oxides significantly enhances its catalytic and adsorption performance toward dyes and heavy metals. [8&9] The growing demand for sustainable nanomaterials has shifted attention toward green synthesis routes, which utilize natural plant extracts, biomolecules, and other eco-friendly reducing agents instead of toxic chemicals. [10] Green synthesis not

only minimizes environmental impact but also allows better control over nanoparticle size, morphology, and surface chemistry due to the stabilizing effects of bioactive compounds. Such methods are particularly attractive for producing metal oxide–carbon nanocomposites for photocatalytic wastewater treatment. In this context, the present work focuses on the integration of green-synthesized CuO nanoparticles using Vitex negundo leaf extract and the biomass-derived carbon from Chromolaena odorata plant stems is expected to synergistically combine the adsorption efficiency of carbon with the catalytic activity of nano-CuO, leading to enhanced degradation performance. This study not only contributes to the development of sustainable catalyst, but also offers an eco-friendly strategy for wastewater remediation.

2. Method

2.1. Preparation Of Carbon Material And Nano-CuO Particles

Chromolaena odorata is a plant species abundantly found in various parts of India, particularly in the state of Kerala. The plant stems were collected, thoroughly washed, dried, and carbonized at 150 °C. The resulting carbon material was subsequently treated with a $SnCl_4$ solution, washed thoroughly, and dried at 110 °C for 12 h to remove any adsorbed impurities from the surface. The prepared carbon support is shown in Figure 1.

Figure 1 Supporting Carbon Material Derived From Chromolaena Odorata Plant Stem

Copper oxide nanoparticles (nano-CuO) were synthesized using Vitex negundo leaf extract. Fresh leaves (10 g) were collected, thoroughly washed, cut into small pieces, and boiled in distilled water for 2 h. The resulting extract was filtered, and the filtrate was mixed with a 0.1 M $CuSO_4$ solution in a 1:9 ratio ($CuSO_4$ solution to plant extract). The mixture was continuously stirred for 24 h, after which the solvent water was evaporated, and the residue was calcined

at 400 °C for 2 h. The formation of nano-CuO from the blue $CuSO_4$ precursor solution using Vitex negundo leaf extract is illustrated in Figure 2.
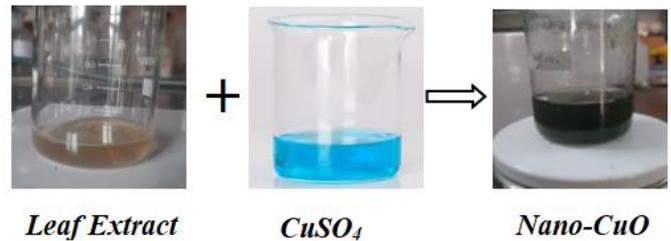

Figure 2 Green Synthesis of Nano-CuO Using Vitex Negundo Leaf Extract and CuSO4

The vitex negundo and Chromolaena odorata plants were shown in Figure 3.

Figure 3 (A) Chromolaena Odorata Plant and (B) Vitex Negundo Plant

2.2. Degradation Studies of MB Dye

The effect of pure carbon and CuO–C on the removal efficiency of MB was investigated. The degradation performance of MB using CuO–C was examined by varying the CuO loading on the carbon matrix. Carbon-supported samples containing 1%, 5%, and 10% nano-CuO were prepared. Each sample was ultrasonicated with the MB dye solution for 5 minutes and subsequently filtered. The residual dye concentration was then measured using a UV–Visible spectrophotometer.

3. Results and discussion

3.1. Results

3.1.1. FESEM Analysis

The FE-SEM image of the green-synthesized CuO nanoparticles is shown in Figure 4. The micrograph clearly reveals that the material consists of well-

defined nanoparticles with an average particle size of approximately 65 nm. Figure 4 FE-SEM Image of Green Synthesized nano-CuO
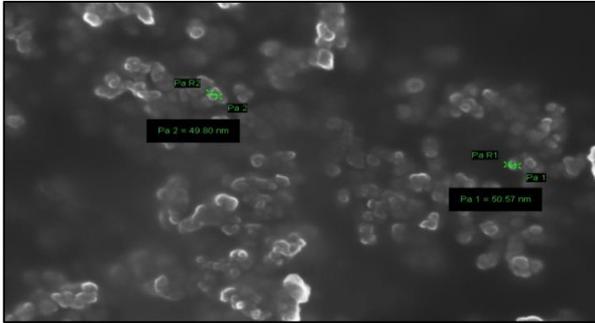

Figure 4 FE-SEM Image of Green Synthesized nano-CuO

3.1.2. FT-IR Analysis

The FT-IR spectrum of the synthesized nano-CuO is presented in Figure 5. The spectrum exhibits characteristic absorption bands at approximately 641 cm^{-1}, 604 cm^{-1}, 530 cm^{-1}, 1628 cm^{-1}, and 3400 cm^{-1}. [11&12]

3.1.3. XRD Analysis

X-ray diffraction (XRD) analysis was performed to investigate the structural properties of the green-synthesized nano-CuO. The XRD pattern of the nano-CuO particles is shown in Figure 6. Prominent diffraction peaks were observed at 2θ values of 35.5°, 38.4°, and 48.5°, which correspond to the characteristic planes of monoclinic CuO. [12] Figure 5 shows The FT-IR spectrum of green synthesized nano-CuO
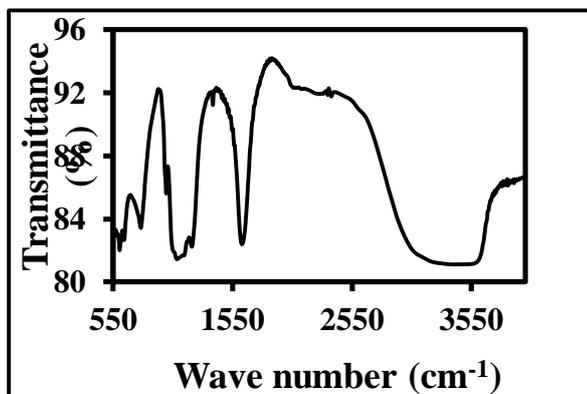

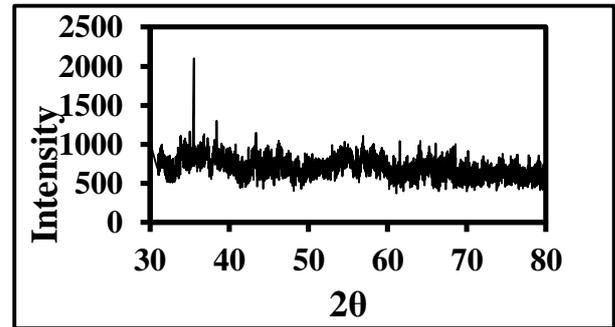**Figure 5** The FT-IR spectrum of green synthesized nano-CuO

Figure 6 The XRD Pattern Of Green-Synthesized Nano-CuO

3.1.4. Degradation Studies

The catalytic degradation of MB using carbon material synthesized from Chromolaena odorata plant stem and various CuO–C nanocomposites is illustrated in Figure 7.
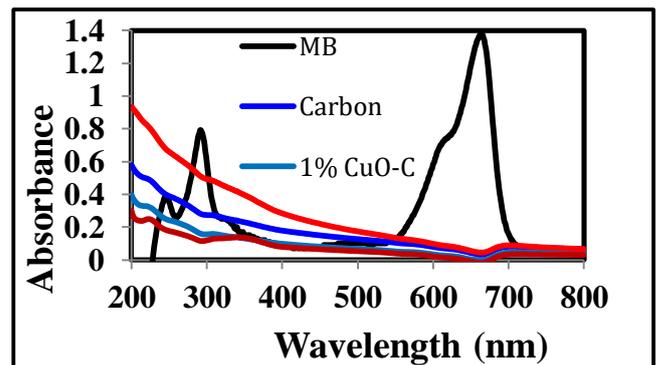

Figure 7 UV–Visible Absorption Spectra Of Methylene Blue (MB), Carbon Material Derived From Chromolaena Odorata Plant Stem, and CuO–C Nanocomposites With Varying CuO loadings (1%, 5%, and 10%)
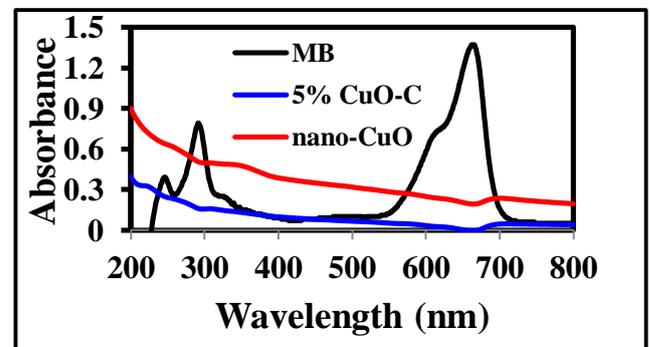

Figure 8 Comparison of the MB Degradation Using Nano-CuO and 5% Nano-CuO Supported On Carbon Material

The catalytic effect of MB degradation was studies using thermally activated nano-CuO supported over carbon. The UV-Vis. Spectrum obtained for the MB degradation at room temperature (RT) and 50 0C is given in Figure 9. Photocatalytic degradation of MB dye is reported elsewhere in the literature, [13&14] but the degradation of MB takes place at RT in our study.
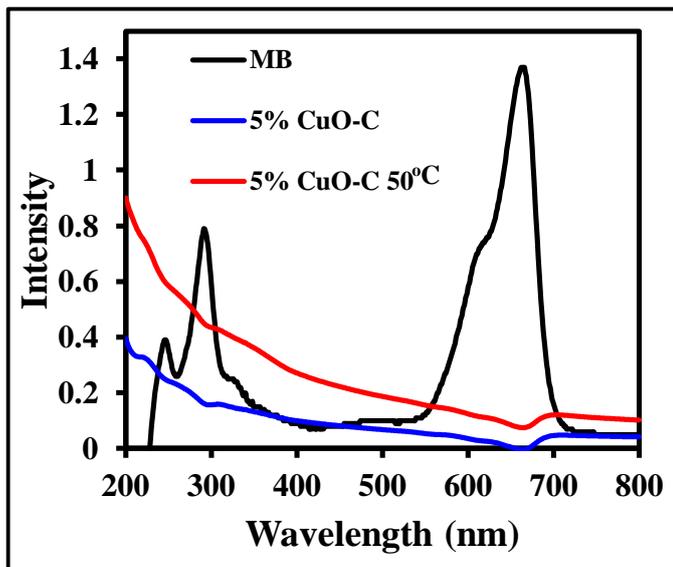

Figure 9 UV–Visible absorption Spectra Showing The Catalytic Effect Of Thermally Activated 5% CuO–C Nanocomposite on MB Degradation at RT and 50 °C

3.2. Discussions
3.2.1. Characterization Techniques

The FE-SEM image of the green-synthesized CuO nanoparticles (Figure 4) reveals the formation of nearly uniform, nanosized particles with an average diameter of approximately 50–70 nm. The particles exhibit a predominantly spherical morphology with slight agglomeration, which may be attributed to the high surface energy and intrinsic magnetic interactions among the nanoparticles. The FT-IR spectrum of the green-synthesized CuO nanoparticles (Figure 5) confirms the successful formation of copper oxide through its characteristic vibrational features. The prominent absorption bands observed in the range of 500–700 cm^{-1} correspond to the stretching vibrations of the Cu–O bond. Specifically, the peaks at 530 cm^{-1}, 604 cm^{-1}, and 641 cm^{-1} are

attributed to Cu–O lattice vibrations. The broad absorption band around 3400 cm^{-1} and the peak at 1628 cm^{-1} are associated with O–H stretching and H–O–H bending vibrations, respectively, indicating the presence of adsorbed water molecules on the nanoparticle surface. [15–19]. These observations confirm the formation of pure CuO with minor surface hydroxylation, which is common in green-synthesized metal oxide nanoparticles. The X-ray diffraction pattern of the green-synthesized CuO nanoparticles (Figure 6) exhibits distinct diffraction peaks at 2θ values of 35.5°, 38.4°, 48.5°, 58.3°, and 66.2°, which correspond to the (111), (200), (202), (113), and (311) planes, respectively. These peaks are in good agreement with the standard data for monoclinic CuO (JCPDS card No. 00-001-1117), confirming the formation of crystalline CuO nanoparticles. [16&17]

3.2.2. Degradation Studies of MB

In this study, three main aspects were investigated to understand the effective degradation of methylene blue (MB) dye: (i) optimization of the amount of nano-CuO required, (ii) the synergistic effect of nano-CuO supported on carbon material, and (iii) the effect of thermal activation of nano-CuO. The UV–Visible spectra of MB degradation (Figures 7–9) clearly demonstrate the effective breakdown of MB molecules upon treatment with nano-CuO and CuO–C nanocomposite materials. The characteristic absorption peaks of MB appearing at 246 nm, 292 nm, 613 nm and 664 nm [20] vanished after only 5 minutes of treatment, indicating rapid degradation. When carbon material derived from Chromolaena odorata stem was used alone, partial adsorption of the dye occurred, suggesting that the removal process was primarily adsorption-based. However, the incorporation of nano-CuO into the carbon matrix significantly enhanced the degradation efficiency due to the synergistic interaction between the carbon support and CuO nanoparticles. Figure 7 illustrates the optimization of CuO loading on carbon. The degradation efficiency improved progressively with increasing CuO content up to 5%, beyond which a slight decrease was observed for the 10% CuO–C nanocomposite. This reduction in performance can be attributed to the saturation of active sites on the

adsorbent surface and possible aggregation of excess CuO nanoparticles, which may block pores or occupy active sites, thereby reducing the overall catalytic efficiency. Hence, the 5% CuO–C nanocomposite was found to be the optimal catalyst for MB degradation in this study. Figure 8 compares the degradation efficiency of pure nano-CuO with that of 5% CuO–C nanocomposite. The absorbance value of MB at 664 nm decreased from 1.37 to 0.196 when treated with nano-CuO alone, while it further reduced to nearly 0.0 for the 5% CuO–C nanocomposite, confirming complete degradation. This observation highlights the strong synergistic effect between the carbon support and CuO nanoparticles, where the carbon matrix enhances the surface area and facilitates efficient electron transfer, thereby reducing charge recombination and improving the catalytic activity toward MB degradation. Figure 9 demonstrates the effect of thermal activation on the catalytic behavior of CuO–C nanocomposites. The results reveal that the degradation of MB using 5% CuO–C nanocomposite is more effective at RT compared to 50 °C. The absorbance intensity at 664 nm reduced significantly to 0.075 at room temperature, whereas a slightly higher absorbance value was recorded at 50 °C, indicating comparatively lower degradation efficiency at elevated temperature. This can be attributed to possible catalyst deactivation or nanoparticle agglomeration upon heating, which reduces the number of available active sites for dye degradation.

Conclusion

The present study successfully demonstrated the effective degradation of methylene blue (MB) dye using carbon material derived from the stem of Chromolaena odorata and green-synthesized copper oxide (CuO) nanoparticles prepared with Vitex negundo leaf extract. The synergistic interaction between nano-CuO and the carbon support significantly enhanced the catalytic performance, leading to rapid and efficient dye degradation. The synthesis route adopted is simple, energy-efficient, and environmentally friendly. The prepared materials were thoroughly characterized using FE-SEM, FT-IR, and XRD analyses, confirming the formation of nanosized, crystalline CuO with good dispersion on the carbon surface. Among the composites tested, the 5% CuO nanocomposite exhibited the highest degradation efficiency toward MB dye. Notably, the degradation occurred rapidly at room temperature without the need for external photo or thermal activation, highlighting the potential of this catalyst as a sustainable and cost-effective material for wastewater treatment. The results suggest that the green-synthesized CuO–carbon composite can serve as a promising candidate for the removal of organic dyes and other contaminants from industrial effluents.

References

[1]. Vanitha, K., Jibrail, K., & Sie, Y. L. (2018). Efficiency of Various Recent Wastewater Dye Removal Methods: A Review. Journal of Environmental Chemical Engineering, 6(4), 4676-4697. doi.org/ 10.1016/ j.jece. 2018. 06. 06.

[2]. Mustafa, T. Y., Tushar, K. S., Sharmeen, A., Ang, H. M. (2014). Dye and its Removal from Aqueous Solution by Adsorption: A Review. Advances in Colloid and Interface Science, 209, 172-184. doi.org/ 10.1016/ j.cis.2014.04.002.

[3]. Akpan, U. G., & Hameed, B. H. (2011). Parameters Affecting the Photocatalytic Degradation of Dyes Using TiO_2-Based Photocatalysts: A Review. Journal of Hazardous Materials, 170(2–3), 520-529. doi.org/10.1016/j.jhazmat.2009.05.039.

[4]. El-Trass, A., El-Shamy, H., El-Mehasseb, I., & El-Kemary, M. (2012). CuO Nanoparticles: Synthesis, Characterization, Optical Properties and Interaction with Amino Acids. Applied Surface Science, 258(7), 2997-3001. doi.org/10.1016/j.apsusc.2011.11.025.

[5]. Archita, B., & Ahmaruzzaman, M. (2018). Microwave Assisted Facile and Green Route for Synthesis of CuO Nanoleaves and Their Efficacy as a Catalyst for Reduction and Degradation of Hazardous Organic Compounds. Journal of Photochemistry and Photobiology A: Chemistry, 353, 215-228. doi.org/ 10.1016/j.jphotochem.2017.11.018.

[6]. Gupta, V. K., Kumar, R., Nayak, A., Saleh, T.

A., & Barakat, M. A. (2013). Adsorptive Removal of Dyes from Aqueous Solution onto Carbon Nanotubes: A Review. Advances in Colloid and Interface Science, 193, 24-374. doi. org/10. 1016/ j.cis. 2013. 03.003.

[7]. Guillermo, C. Q., Cristian, G. R., María, V. L. R., María, D. P. F. P., Inmaculada, V. G. Shu, Z., Zhang, X., & Chen, X. (2025). Carbon-Based Metal Oxide Nanocomposites for Water Treatment by Photocatalytic Processes. Environmental Research, 279, 121724-121732.doi.org/10.1016/j.envres.2025.121724.

[8]. Akinnike, A., Oluranti, A., Edith, A., & Ezinne, I. (2024). The Role of Catalyst in the Adsorption of Dye: Homogeneous Catalyst, Heterogeneous Catalyst, and Advanced Catalytic Activated Carbon, Critical Review. Desalination and Water Treatment, 320, 100780-100796. doi.org/ 10.1016/ j.dwt.2024. 100780.

[9]. Josiane, P., Marcela, F. S., Marcelo, F. V., Rosangela, B. & Angelica M. S. V. (2017). Synthesis and Impregnation of Copper Oxide Nanoparticles on Activated Carbon through

[10]. Green Synthesis for Water Pollutant Removal. Materials Research, 21, 2-11. doi:10.1590/1980-5373-mr-2016-0460.

[11]. Josiane Peternelaa, Marcela Fernandes Silvaa, Marcelo Fernandes Vieiraa, Rosângela Bergamascoa, Angélica Marquetotti Salcedo Vieira

[12]. Siavash I., (2011). Green Synthesis of Metal Nanoparticles Using Plants. Green Chemistry, 13 (10), 2638-2650. doi. org/ 10. 1039 /C1GC15386B.

[13]. Aliakbar, D. K., Mahbobeh, S., Marketa, J., & Pavel, M. (2020). Synthesis, Characterization, and Antibacterial Activity of Copper(II) Oxide Nanoparticles Prepared by Thermal Decomposition. Journal of Surface Investigation: X-ray, Synchronotron and Neutron Techniques, 14, 961-964.doi.org/10.1134/S1027451020050109.

[14]. Fernandes, D. M., Silva, R., Hechenleitner, A. A. W., Radovanovic, M., Melo, M. A., &

Pineda, E. A. G. (2009). Synthesis and Characterization of ZnO, CuO and a Mixed Zn and Cu Oxide. Materials Chemistry and Physics. 115 (1), 110-115. doi.org/ 10.1016 /j.matchemphys.2008.11.038.

[15]. Asfaw, N., Said, M., Hulugirgesh, D. W., Abera, D. A. & Minbale, G. (2023). Enhanced Photocatalytic Degradation of Methylene Blue Dye Using Eco-friendly Synthesized rGO@ZnO Nanocomposites, Scientific Reports, 13, 22234. doi.org/ 10.1038/s41598-023-48826-7.

[16]. Biniyam, A. B., Ali, A., Desalew, Getahun, W. D., Derese, M. M., Kedir, S. M., Jia-Yaw, C., & Wubshet, M. G. (2023). Enhanced Photocatalytic Degradation of Methylene Blue Dye via Valorization of A Polyethylene Terephthalate Plastic Waste-Derived Metal-Organic Framework-Based ZnO@Co-BDC Composite Catalyst, Nanoscale Advances, 7, 3834-3845. doi: 10.1039/ d4na01071j.

[17]. Prasanta, K. R., Samarpita, S., Ashish, K. S., Iohborlang, M. U., Rashmi, R. D., Ashim, J. T., & Vijay V. (2014). CuO Nanorods: A Potential and Efficient Adsorbent in Water Pollution, RSC Advances, 4, 40580-40587. doi.org/10.1039/C4RA04619F.

[18]. Muhammad, H. S., Maimona, S., Saima, M., Muhammad, S., Asma, A., Sumreen, H., Saba, S., Arif, M. K., Abeer, H., Graciela, D. A. Q. & Elsayed, F. A. A. (2024). Biofabrication of Copper Oxide Nanoparticles Using Dalbergia Sisso Leaf Extract for Antibacterial, Antibiofilm and Antioxidant Activities, Scientific Reports, 14, 31867.doi.org/10.1038/s41598-024-83199-5.

[19]. Elango, M., Deepa, M., Subramanian, R. & Mohamed, A. M. (2018). Synthesis, Characterization, and Antibacterial Activity of Polyindole/Ag–Cuo Nanocomposites by Reflux Condensation Method, Polymer-Plastics Technology and Engineering, 57(14), 1440-14doi:10.1080/03602559.2017.1410832.

[20]. Angel, S. J., & Kalaichelvan, G. (2025). Synthesis and Characterization of CuO

Nanoparticles from Bioleached Copper through Modified and Optimized Double Precipitation Method, ACS Omega, 10(10), 10193-10198. doi. org/ 10.1 021/ acsomega. 4c09064.

[21]. Chandan, T., Indranirekha, S., Moushumi, H., & Manash, R. D., (2014). Reduction of aromatic nitro compounds catalyzed by biogenic CuO nanoparticles, RSC Advances, 4, 53229-53236. doi.org/ 10.1039 /C4RA 10397A.

[22]. Dinh, V. P., Huynh, T. D. T., Le, H. M., Nguyen, V. D., Dao, V. A., Hung, N. Q., Tuyen, L. A., Lee, S., Yi, J., Nguyen, T. D., & Tan, L. V. (2019). Insight into the Adsorption Mechanisms of Methylene Blue and Chromium(III) from Aqueous Solution onto Pomelo Fruit Peel. RSC Advances, 9, 25847-25860. doi: 10.1039/c9ra04296b.