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Green Synthesis of Benzimidazole Derivatives Using Copper Oxide Nanoparticles: An Efficient Nanocatalytic Approach

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Abstract: *Benzimidazole derivatives represent an important class of nitrogen-containing heterocyclic compounds with extensive pharmaceutical and biological applications due to their remarkable medicinal properties. The present study focuses on the green synthesis of benzimidazole derivatives using copper oxide (CuO) nanoparticles as an efficient heterogeneous nanocatalyst. Copper oxide nanoparticles were synthesized through an eco-friendly plant-mediated route using citrus leaf extract, where phytochemicals present in the extract acted as natural reducing and stabilizing agents during nanoparticle formation.*

The synthesized CuO nanoparticles were characterized using UV–Visible spectroscopy, X-ray Diffraction (XRD), Fourier Transform Infrared Spectroscopy (FTIR), and Scanning Electron Microscopy (SEM). UV–Visible spectroscopic analysis confirmed nanoparticle formation through characteristic absorption in the UV region, while XRD analysis revealed the formation of monoclinic crystalline CuO nanoparticles with nanoscale crystallite dimensions. FTIR studies confirmed Cu–O bond formation along with phytochemical stabilization of the nanoparticles, and SEM analysis indicated spherical and agglomerated nanoparticle morphology.

The synthesized CuO nanoparticles were successfully utilized as reusable heterogeneous catalysts for the condensation reaction between o-phenylenediamine and various aromatic aldehydes to synthesize substituted benzimidazole derivatives. The developed catalytic protocol exhibited several advantages including high product yield, shorter reaction time, operational simplicity, catalyst recyclability, and environmentally benign reaction conditions. The synthesized benzimidazole derivatives were further characterized using FTIR spectroscopy, ^{1}H NMR spectroscopy, and ^{13}C NMR spectroscopy. Overall, the present methodology represents a sustainable, efficient, and eco-friendly synthetic strategy for the preparation of biologically important benzimidazole derivatives.

Keywords: Benzimidazole, copper oxide nanoparticles, green synthesis, nanocatalysis, heterocyclic compounds, medicinal chemistry, CuO nanoparticles.

I. INTRODUCTION

Heterocyclic compounds are an important class of cyclic organic molecules containing carbon atoms along with heteroatoms such as nitrogen, oxygen, or sulfur within the ring structure. Among these compounds, nitrogen-containing heterocycles are particularly significant because of their wide range of biological and pharmaceutical applications.

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Heterocyclic compounds are extensively used in medicinal chemistry, agrochemicals, dyes, polymers, and pharmaceutical industries due to their diverse chemical and biological properties.

Benzimidazole is one of the most important nitrogen-containing heterocyclic systems and consists of a fused benzene ring and an imidazole ring. The benzimidazole nucleus can be represented as:

\text{Benzimidazole = Benzene\ Ring + Imidazole\ Ring}

Benzimidazole exhibits several important chemical characteristics including aromaticity, amphoteric behavior, tautomerism, and resonance stabilization. Unsubstituted benzimidazole undergoes rapid prototropic tautomerism between nitrogen atoms, which contributes to its chemical reactivity and biological activity. Due to these structural and electronic properties, benzimidazole derivatives possess remarkable pharmacological importance.

Benzimidazole derivatives exhibit a wide range of biological activities such as antimicrobial, antiviral, anticancer, antihypertensive, anti-inflammatory, antifungal, and anthelmintic activities. Several therapeutically important drugs contain the benzimidazole nucleus, including omeprazole used as an anti-ulcer agent, thiabendazole as an anthelmintic drug, astemizole as an antihistaminic agent, mebendazole as an antiparasitic compound, and diabazole as an antihypertensive drug. These applications demonstrate the medicinal significance of benzimidazole derivatives in pharmaceutical chemistry.

In recent years, nanocatalysis has emerged as an important approach in organic synthesis because nanocatalysts provide high surface area, enhanced catalytic activity, shorter reaction time, and improved selectivity compared to conventional catalysts. Heterogeneous nanocatalysts are particularly advantageous because they allow easy catalyst recovery, catalyst recyclability, reduced waste generation, eco-friendly synthesis, and simple work-up procedures. These properties make nanocatalysis highly suitable for sustainable organic synthesis and green chemistry applications.

Among various nanomaterials, copper oxide (CuO) nanoparticles have attracted considerable attention due to their semiconducting properties, high catalytic activity, thermal stability, low toxicity, and excellent redox behavior. CuO nanoparticles are widely used in catalysis, sensors, environmental remediation, and organic transformations. In the present study, CuO nanoparticles were synthesized through a green synthetic approach using citrus leaf extract. Citrus leaf extract contains various phytochemicals such as flavonoids, polyphenols, terpenoids, and alkaloids, which act as natural reducing, stabilizing, and capping agents during nanoparticle synthesis. This plant-mediated synthetic route provides an environmentally friendly, low-cost, and sustainable approach for preparation of copper oxide nanoparticles suitable for nanocatalytic applications.

The primary objective of the present study was to synthesize copper oxide (CuO) nanoparticles using environmentally friendly green chemistry methods and to characterize the synthesized nanoparticles using various spectroscopic and analytical techniques. Another major objective was to utilize the synthesized CuO nanoparticles as efficient heterogeneous nanocatalysts for the preparation of benzimidazole derivatives through sustainable synthetic methodologies.

The study also aimed to develop an environmentally sustainable synthetic route with reduced hazardous chemical usage and improved reaction efficiency. In addition, efforts were made to investigate the catalytic efficiency and recyclability of CuO nanoparticles and to synthesize biologically important heterocyclic compounds possessing potential pharmaceutical applications. The overall objective of the work was to combine green nanotechnology and heterocyclic chemistry for the development of eco-friendly and efficient organic synthesis.

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The materials required for the present study included various analytical grade chemicals such as copper sulfate or copper nitrate, o-phenylenediamine, substituted aromatic aldehydes, ethanol, and distilled water. Copper salts were used as precursor materials for the synthesis of copper oxide nanoparticles, while o-phenylenediamine and aromatic aldehydes served as starting materials for the synthesis of benzimidazole derivatives. Ethanol and distilled water were used as solvents during nanoparticle synthesis and catalytic reactions.

Fresh citrus leaves were used as the biological material for the green synthesis of CuO nanoparticles. The phytochemicals present in the citrus leaf extract acted as natural reducing, stabilizing, and capping agents during nanoparticle formation.

Various analytical instruments and laboratory apparatus were employed for synthesis and characterization studies. A UV–Visible spectrophotometer was used for optical characterization of nanoparticles, while FTIR spectroscopy was utilized for identification of functional groups and confirmation of Cu–O bond formation. Surface morphology and particle size analysis were carried out using a scanning electron microscope (SEM), and crystalline structure determination was performed using an X-ray diffractometer. A magnetic stirrer was used during synthesis procedures for uniform mixing, and a muffle furnace was employed for drying and calcination of the synthesized nanoparticles.

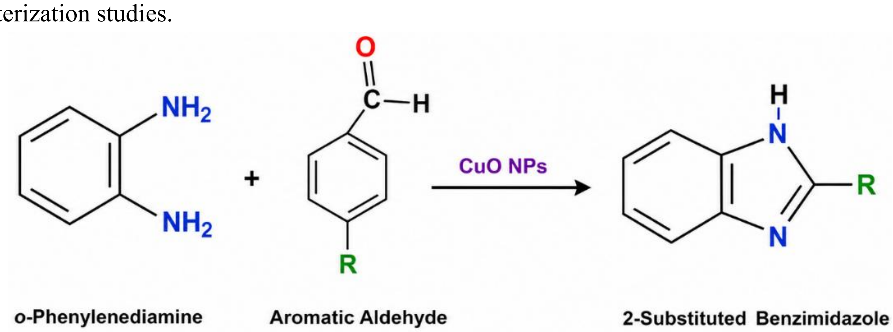
Preparation of Citrus Leaf Extract

Fresh citrus leaves were collected and washed thoroughly with distilled water to remove dust and other impurities. The cleaned leaves were air-dried and chopped into small pieces. The chopped leaf material was then boiled in distilled water for about 15–20 minutes to extract the phytochemical constituents present in the leaves. The obtained extract was filtered using Whatman filter paper and stored for further use in the green synthesis of copper oxide nanoparticles.

Synthesis of CuO Nanoparticles

Copper oxide nanoparticles were synthesized through a green plant-mediated method using citrus leaf extract as a natural reducing and stabilizing agent. Initially, copper sulfate was dissolved in distilled water to prepare the copper salt precursor solution. Citrus leaf extract was then added dropwise into the copper salt solution under continuous stirring conditions. The reaction mixture was heated at 60–80°C to facilitate nanoparticle formation.

During the reaction, the color of the solution gradually changed from blue to green and finally to dark brown, indicating the formation of copper oxide nanoparticles. The reaction mixture was allowed to age for several hours to complete nanoparticle precipitation. The obtained precipitate was separated by filtration and repeatedly washed to remove impurities and unreacted substances. Finally, the product was dried at 80–100°C and calcined at 400–500°C to obtain crystalline CuO nanoparticles. The formation of crystalline copper oxide nanoparticles was subsequently confirmed through characterization studies.



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II. CHARACTERIZATION OF CUO NANOPARTICLES

UV–Visible Spectroscopy

UV–Visible spectroscopic analysis of the synthesized copper oxide nanoparticles showed a broad absorption band in the range of 250–350 nm. The observed absorption confirmed the successful formation of CuO nanoparticles and indicated charge transfer transitions occurring between O^{2-} ions and Cu^{2+} ions within the semiconductor lattice. The optical properties of the nanoparticles were further studied by calculating the band gap energy using the relation:

where (E_g) represents band gap energy, (h) is Planck's constant, (c) is the speed of light, and (\lambda) is the wavelength of absorption. The calculated band gap confirmed the semiconducting nature of the synthesized CuO nanoparticles.

X-Ray Diffraction (XRD) Analysis

X-ray diffraction analysis was carried out to determine the crystalline structure and phase purity of the synthesized nanoparticles. The analysis follows Bragg's law:

n\lambda = 2d\sin\theta

Characteristic diffraction peaks observed at 2θ values of 32.5°, 35.5°, 38.7°, 48.7°, and 53.5° corresponded to the crystal planes (-110), (111), (111), (-202), and (020), respectively. These diffraction peaks confirmed the monoclinic crystal structure of CuO nanoparticles. The sharp peaks indicated good crystallinity, whereas peak broadening confirmed the nanoscale dimensions of the synthesized particles. The average crystallite size was calculated using the Debye–Scherrer equation:

III. FTIR ANALYSIS

FTIR spectroscopy was employed to identify functional groups associated with the synthesized nanoparticles and to confirm Cu–O bond formation. The broad absorption bands observed between 3200–3500 cm^{-1} corresponded to O–H stretching vibrations of hydroxyl groups present in phytochemicals and adsorbed moisture. Peaks appearing in the range of 1600–1650 cm^{-1} were attributed to C=O and C=C stretching vibrations of organic compounds present in the citrus leaf extract. The characteristic absorption band observed between 500–600 cm^{-1} corresponded to Cu–O stretching vibrations, confirming the formation of copper oxide nanoparticles. The presence of organic functional groups indicated phytochemical stabilization and capping of the nanoparticles.

Scanning Electron Microscopy (SEM)

SEM analysis revealed that the synthesized CuO nanoparticles possessed predominantly spherical morphology with agglomerated particle distribution and rough surface texture. The nanoparticles exhibited nanoscale dimensions with slight agglomeration due to high surface energy and intermolecular interactions. The rough surface morphology and nanosized structure provide enhanced surface area, which is advantageous for catalytic applications and improved reaction efficiency in organic synthesis.

IV. SYNTHESIS OF BENZIMIDAZOLE DERIVATIVES

The synthesis of substituted benzimidazole derivatives was carried out through the condensation reaction between o-phenylenediamine and various aromatic aldehydes in the presence of CuO nanoparticles as heterogeneous nanocatalysts. The general reaction can be represented as:

In the general experimental procedure, o-phenylenediamine and the selected aromatic aldehyde were mixed thoroughly, followed by the addition of CuO nanoparticles as the catalyst. The reaction mixture was ground using a mortar and pestle for about 20–30 minutes under solvent-free conditions to facilitate efficient mixing and reaction progress. After completion of the reaction, the product was separated by filtration and purified by recrystallization to obtain pure

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benzimidazole derivatives. The CuO nanocatalyst was recovered after the reaction and reused for subsequent synthetic cycles, demonstrating catalyst recyclability and sustainability.

The proposed reaction mechanism involves several sequential steps. Initially, the aromatic aldehyde undergoes condensation with one amino group of o-phenylenediamine to form an intermediate. This is followed by Schiff base formation through imine linkage generation. Subsequently, intramolecular cyclization occurs, leading to ring closure and formation of the benzimidazoline intermediate. In the final step, oxidation and aromatization take place to produce the stable benzimidazole derivative. The CuO nanoparticles facilitate the reaction by providing active catalytic sites, enhancing reaction efficiency, and accelerating the cyclization and oxidation processes under mild and environmentally friendly conditions.

V. RESULTS AND DISCUSSION

Entry	Aldehyde	Product	Time (min)	Yield (%)
1	Benzaldehyde	2-Phenylbenzimidazole	10	92
2	p-Nitrobenzaldehyde	2-(4-Nitrophenyl)benzimidazole	10	94
3	p-Methylbenzaldehyde	2-(4-Methylphenyl)benzimidazole	10	92
4	p-Methoxybenzaldehyde	2-(4-Methoxyphenyl)benzimidazole	15	90
5	p-Chlorobenzaldehyde	2-(4-Chlorophenyl)benzimidazole	10	90

Results and Discussion

The catalytic activity of CuO nanoparticles was significantly influenced by the nature of substituents present on the aromatic aldehydes. Aromatic aldehydes containing electron-withdrawing groups such as $–NO_2$ and –Cl exhibited increased reaction efficiency and improved product yields due to enhanced electrophilic character of the carbonyl carbon. In contrast, electron-donating groups such as $–CH_3$ and $–OCH_3$ also provided excellent yields, although a slight increase in reaction time was observed because of reduced electrophilic activation. Overall, the developed catalytic system demonstrated excellent substrate compatibility and efficient synthesis of substituted benzimidazole derivatives under mild conditions.

The recyclability of the CuO nanocatalyst was also investigated to evaluate its practical applicability and sustainability. The fresh catalyst provided a product yield of 95%, while the first and second reuse cycles afforded yields of 88% and 71%, respectively. The catalyst retained significant catalytic activity even after multiple reaction cycles. The slight reduction in catalytic efficiency after repeated use may be attributed to nanoparticle agglomeration and gradual surface deactivation of active catalytic sites.

Characterization of Benzimidazole Derivatives

FTIR spectroscopic analysis confirmed the successful formation of benzimidazole derivatives through characteristic functional group absorptions. The broad absorption bands observed between 3400–3200 cm^{-1} corresponded to N–H stretching vibrations, while peaks in the range of 1600–1620 cm^{-1} were assigned to C=N stretching vibrations of the benzimidazole ring. Additional peaks observed between 1500–1580 cm^{-1} corresponded to aromatic C=C stretching vibrations, and bands in the range of 1250–1300 cm^{-1} were attributed to C–N stretching vibrations. These spectral features confirmed successful cyclization and formation of the benzimidazole nucleus.

The ^1H NMR spectra of the synthesized compounds exhibited characteristic signals supporting benzimidazole ring formation. Signals observed between δ 12.5–13.0 ppm corresponded to the NH proton of the benzimidazole ring, while aromatic proton signals appeared in the range of δ 7.0–8.2 ppm. These spectral characteristics confirmed aromatic conjugation and successful synthesis of substituted benzimidazole derivatives.

Further structural confirmation was obtained through ^{13}C NMR analysis. Characteristic signals observed between δ 150–155 ppm were assigned to the C=N carbon of the benzimidazole ring, while aromatic carbon signals appeared between δ 120–140 ppm. These observations confirmed the aromatic heterocyclic framework and supported the proposed molecular structures of the synthesized compounds.

Advantages of the Present Method

The developed synthetic methodology offers several important advantages over conventional synthetic approaches. Synthetic advantages include high product yields, shorter reaction times, mild reaction conditions, and broad substrate scope. Environmental advantages of the method involve eco-friendly synthesis, recyclable catalyst system, reduced solvent usage, and minimal waste generation. In addition, the CuO nanocatalyst exhibited high catalytic activity, easy catalyst recovery, good reusability, and operational simplicity, making the process economically and environmentally beneficial.

Conclusion

The present study successfully demonstrated the green synthesis of copper oxide nanoparticles using citrus leaf extract through an environmentally friendly synthetic approach. Characterization studies using UV–Visible spectroscopy, FTIR, SEM, and XRD analyses confirmed the successful formation of CuO nanoparticles with nanoscale crystalline structure. The synthesized CuO nanoparticles acted as efficient heterogeneous catalysts for the synthesis of benzimidazole derivatives under mild and sustainable reaction conditions.

The developed methodology provided several advantages including high product yields, shorter reaction times, catalyst recyclability, and environmentally sustainable conditions. Spectroscopic characterization studies further confirmed the successful formation of substituted benzimidazole derivatives. Overall, the present work highlights the importance of nanotechnology and green chemistry in heterocyclic synthesis, medicinal chemistry, and sustainable organic transformations.

Future Scope

Future investigations in this field may focus on detailed biological screening of the synthesized benzimidazole derivatives, including anticancer and antimicrobial activity studies. Further research may also emphasize catalyst optimization, enhancement of catalyst recyclability, and industrial-scale green synthesis for pharmaceutical and medicinal applications.

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