IJARSCT
International Journal of Advanced Research in Science, Communication and Technology
International Open-Access, Double-Blind, Peer-Reviewed, Refereed, Multidisciplinary Online Journal
ISSN: 2581-9429
Volume 6, Issue 5, June 2026
Impact Factor: 8.2

Nanocatalyzed Synthesis of Benzimidazole Derivatives Using Green Synthesized ZnO Nanoparticles: An Eco-Friendly and Sustainable Approach

Supriya Mhatre[1], Gurumeet Wadhawa[2], Yashwant Gaikwad[3], Vitthal Shivankar[4], Aamod Thakkar[5]

[1] Student, P.G. Department of Chemistry, Veer Wajekar College, Phunde, Uran

[2 3] Assistant Professor, Department of Chemistry, Veer Wajekar College, Phunde, Uran

[4] Principal, Raosaheb Ramrao Patil Mahavidyalaya, Savlaj, Tal. Tasgaon, Dist. Sangli, Maharashtra

[5] I/C Principal, Veer Wajekar College, Phunde, Uran

Abstract: *Benzimidazole derivatives constitute an important class of nitrogen-containing heterocyclic compounds that exhibit a wide range of biological and pharmaceutical activities, including antimicrobial, antiviral, anticancer, antihypertensive, anti-inflammatory, and anthelmintic properties. Due to their immense therapeutic significance, the development of efficient and environmentally sustainable synthetic methodologies for benzimidazole derivatives has attracted considerable attention in modern organic chemistry.*

The present study focuses on the green synthesis of zinc oxide (ZnO) nanoparticles using curry leaf extract (Murraya koenigii) and their application as an efficient heterogeneous nanocatalyst for the synthesis of benzimidazole derivatives under solvent-free conditions. The ZnO nanoparticles were synthesized through an eco-friendly route employing plant phytochemicals as reducing and stabilizing agents. The synthesized nanoparticles were characterized using UV–Visible spectroscopy, X-ray diffraction (XRD), Fourier Transform Infrared Spectroscopy (FTIR), and Scanning Electron Microscopy (SEM).

The catalytic activity of ZnO nanoparticles was investigated in the condensation reaction between o-phenylenediamine and various substituted aromatic aldehydes. The reactions proceeded smoothly under mild conditions, affording corresponding benzimidazole derivatives in excellent yields (90–94%) within short reaction times. The catalyst demonstrated good selectivity, operational simplicity, and recyclability up to three cycles.

The developed methodology offers several advantages such as eco-friendly reaction conditions, shorter reaction time, high product yield, easy catalyst recovery, and reduced environmental impact. This study demonstrates the potential of green synthesized ZnO nanoparticles as an efficient and sustainable catalyst for the synthesis of biologically significant benzimidazole derivatives.

Keywords: Benzimidazole, ZnO nanoparticles, green synthesis, heterogeneous catalysis, eco-friendly chemistry, nanocatalyst.

I. INTRODUCTION

1.1 Heterocyclic Compounds and Their Importance

Heterocyclic compounds are organic molecules containing rings composed of carbon atoms along with one or more heteroatoms such as nitrogen, oxygen, or sulfur. Among these, nitrogen-containing heterocyclic compounds occupy a

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significant place in medicinal and synthetic organic chemistry due to their extensive biological activities and occurrence in natural systems.

Nitrogen heterocycles are fundamental structural components of biologically important molecules such as amino acids, nucleic acids, vitamins, alkaloids, and coenzymes. Their wide-ranging pharmacological activities have made them valuable scaffolds in the development of pharmaceuticals, agrochemicals, dyes, polymers, and advanced materials.

Among the various nitrogen-containing heterocycles, benzimidazole and its derivatives have emerged as one of the most important classes of compounds because of their broad spectrum of biological activities.

1.2 Benzimidazole Ring System

Benzimidazole is a bicyclic aromatic heterocyclic compound formed by the fusion of benzene and imidazole rings. The benzimidazole nucleus contains two nitrogen atoms within the imidazole ring, which contribute significantly to its chemical reactivity and biological activity.

The benzimidazole ring system exhibits amphoteric behavior due to the presence of acidic and basic nitrogen atoms. The unsubstituted nitrogen atom undergoes rapid tautomerism, contributing to the compound's unique physicochemical properties.

The structural framework of benzimidazole is highly versatile and serves as a privileged scaffold in medicinal chemistry. Substitution at different positions of the benzimidazole nucleus significantly alters biological activity, thereby enabling the design of diverse therapeutic agents.

1.3 Biological Significance of Benzimidazole Derivatives

Benzimidazole derivatives possess remarkable pharmacological properties and are widely used in medicinal chemistry. Some important biological activities include:

- Antimicrobial activity
- Antiviral activity
- Anticancer activity
- Anthelmintic activity
- Anti-inflammatory activity
- Antihypertensive activity
- Antifungal activity
- Antiulcer activity
- Antidiabetic activity

Several commercially important drugs contain the benzimidazole nucleus, including albendazole, mebendazole, omeprazole, thiabendazole, and pantoprazole.

The importance of benzimidazole compounds increased significantly after the discovery that 5,6-dimethyl-1-(β-D-ribofuranosyl)benzimidazole forms an integral part of Vitamin B12.

1.4 Conventional Methods for the Synthesis of Benzimidazoles

Traditionally, benzimidazole derivatives are synthesized by condensation reactions involving o-phenylenediamine and carboxylic acids, aldehydes, acid chlorides, nitriles, or other carbonyl-containing compounds. Although these methods are effective, many require harsh reaction conditions, toxic reagents, longer reaction times, or environmentally hazardous solvents.

The most common synthetic approach involves the condensation of o-phenylenediamine with aldehydes or carboxylic acids under acidic conditions.

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1.5 Green Chemistry and Nanocatalysis

Green chemistry emphasizes the design of environmentally sustainable chemical processes that minimize waste generation, energy consumption, and hazardous substances. In recent years, nanotechnology has provided significant opportunities for developing eco-friendly catalytic systems.

Nanoparticles exhibit unique properties such as:

- High surface area
- Enhanced catalytic activity
- Better selectivity
- Improved reaction efficiency
- Reusability

Among various nanomaterials, zinc oxide (ZnO) nanoparticles have attracted considerable attention because of their:

- Non-toxic nature
- Low cost
- High catalytic efficiency
- Environmental compatibility
- Thermal stability

ZnO nanoparticles have been successfully employed in various organic transformations including condensation reactions, oxidation reactions, and heterocyclic synthesis.

1.6 Green Synthesis of ZnO Nanoparticles

Conventional methods for nanoparticle synthesis often involve hazardous chemicals and energy-intensive procedures. Green synthesis using plant extracts has emerged as a sustainable alternative.

Plant extracts contain phytochemicals such as:

- Flavonoids
- Alkaloids
- Phenolics
- Terpenoids
- Proteins

These biomolecules act as reducing, stabilizing, and capping agents during nanoparticle synthesis.

In the present study, curry leaf extract (*Murraya koenigii*) was utilized for the green synthesis of ZnO nanoparticles.

1.7 Aim and Objectives of the Study

Aim

To synthesize benzimidazole derivatives using green synthesized ZnO nanoparticles as an eco-friendly heterogeneous catalyst.

Objectives

1. To prepare ZnO nanoparticles using curry leaf extract.
2. To characterize synthesized ZnO nanoparticles using UV–Vis, XRD, FTIR, and SEM techniques.

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3. To synthesize benzimidazole derivatives via condensation reactions.
4. To investigate the catalytic efficiency of ZnO nanoparticles.
5. To evaluate catalyst recyclability and sustainability.
6. To develop an environmentally benign synthetic methodology.

II. LITERATURE REVIEW

Benzimidazole chemistry has been extensively explored because of its diverse biological applications. Numerous synthetic methods have been reported for the preparation of benzimidazole derivatives.

Phillips first developed a classical method involving the condensation of o-phenylenediamine with carboxylic acids in hydrochloric acid under reflux conditions. Later, several modifications were introduced using aldehydes, oxidants, microwave irradiation, Lewis acids, and heterogeneous catalysts.

Recent developments have focused on green and sustainable methodologies employing recyclable catalysts and solvent-free conditions.

ZnO nanoparticles have emerged as efficient catalysts in organic synthesis due to their acid-base properties and nanoscale dimensions. Green synthesized ZnO nanoparticles have demonstrated promising catalytic activity in condensation reactions, oxidation reactions, and heterocyclic synthesis.

The use of plant extracts for nanoparticle synthesis has gained increasing attention because it eliminates toxic reducing agents and promotes environmentally sustainable chemistry.

III. EXPERIMENTAL SECTION

3.1 Materials and Chemicals

All chemicals used in this study were of analytical grade and obtained commercially from S.D. Fine Chemicals. The chemicals were used without further purification.

Instruments Used

- UV–Visible Spectrophotometer
- FTIR Spectrophotometer
- X-ray Diffractometer
- Scanning Electron Microscope
- NMR Spectrometer

Spectral Analysis

- ^1H NMR spectra were recorded at 300 MHz.
- ^13C NMR spectra were recorded at 75 MHz.
- Tetramethylsilane (TMS) was used as an internal standard.
- DMSO-d6 and CDCl3 were used as deuterated solvents.

IV. GREEN SYNTHESIS OF ZNO NANOPARTICLES

4.1 Preparation of Curry Leaf Extract

Fresh curry leaves (*Murraya koenigii*) were collected and thoroughly washed with distilled water to remove dust and impurities. The leaves were air-dried at room temperature.

Approximately 10–20 g of curry leaves were boiled in 100 mL distilled water for 10–15 minutes. The solution was cooled and filtered using Whatman filter paper.

The filtrate obtained served as a reducing and stabilizing agent for nanoparticle synthesis.

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4.2 Preparation of Zinc Precursor Solution

A 0.1 M zinc nitrate solution was prepared by dissolving zinc nitrate hexahydrate in distilled water under continuous stirring.

4.3 Synthesis of ZnO Nanoparticles

The curry leaf extract was added dropwise to the zinc nitrate solution under constant stirring at 60–80°C.
The reaction mixture was stirred continuously for 1–2 hours until the formation of a white precipitate indicated nanoparticle formation.

4.4 Purification and Calcination

The reaction mixture was cooled to room temperature and centrifuged.
The precipitate was washed repeatedly with distilled water and ethanol to remove impurities.
The purified precipitate was dried at 80–100°C and calcined at 400–500°C for 2–3 hours to obtain crystalline ZnO nanoparticles.

V. SYNTHESIS OF BENZIMIDAZOLE DERIVATIVES

5.1 General Procedure

In a typical procedure, o-phenylenediamine (10 mmol), substituted aromatic aldehyde (10 mmol), and ZnO nanoparticles (0.8 mol%) were ground together in a mortar and pestle at room temperature for approximately 25 minutes.
After completion of the reaction, 10 mL of water was added to the reaction mixture and the product was filtered.
The crude product was recrystallized using a suitable solvent.
The catalyst was recovered, dried, and reused for subsequent reaction cycles.

5.2 Reaction Mechanism

The reaction proceeds through the following steps:

1. Activation of aldehyde by ZnO nanoparticles.
2. Nucleophilic attack by o-phenylenediamine.
3. Formation of Schiff base intermediate.
4. Cyclization reaction.
5. Oxidative aromatization yielding benzimidazole derivative.

ZnO nanoparticles enhance the reaction rate by increasing the electrophilicity of the aldehyde carbonyl group.

VI. CHARACTERIZATION OF ZNO NANOPARTICLES

6.1 UV–Visible Spectroscopy

UV–Visible spectroscopic analysis confirmed the successful formation of ZnO nanoparticles by exhibiting a characteristic absorption peak in the range of 350–380 nm. The observed absorption band corresponds to the band-gap excitation of ZnO nanoparticles and indicates their semiconducting nature. The optical band-gap energy of the nanoparticles was calculated using the relation E_g = \frac{hc}{\lambda}. A slight blue shift in the absorption spectrum was observed, which suggests the formation of smaller nanoparticles due to the quantum confinement effect.

6.2 X-ray Diffraction (XRD) Analysis

X-ray diffraction analysis confirmed the crystalline nature and phase purity of the synthesized ZnO nanoparticles. The diffraction peaks observed at 2θ values of 31.7°, 34.4°, 36.2°, 47.5°, 56.6°, and 62.8° corresponded to the characteristic planes of the hexagonal wurtzite structure of ZnO. The sharp and intense peaks indicated good crystallinity of the

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nanoparticles. The average crystallite size was calculated using the Debye–Scherrer equation, which confirmed the nanoscale dimensions of the synthesized ZnO particles.

6.3 Fourier Transform Infrared Spectroscopy (FTIR)

FTIR analysis revealed the presence of various functional groups associated with plant-derived biomolecules and Zn–O bonding in the synthesized nanoparticles. The broad absorption band observed in the range of 3200–3500 cm^{-1} was assigned to O–H stretching vibrations. The peak around 1600 cm^{-1} corresponded to C=O and C=C vibrations, while the absorption bands in the range of 1000–1200 cm^{-1} were attributed to C–O stretching vibrations. The characteristic Zn–O stretching vibration observed in the range of 400–600 cm^{-1} confirmed the successful formation of ZnO nanoparticles. The presence of organic functional groups further indicated the role of phytochemicals in the reduction and stabilization of nanoparticles.

6.4 Scanning Electron Microscopy (SEM)

SEM analysis revealed that the synthesized ZnO nanoparticles exhibited spherical, hexagonal, and rod-like morphologies with particle sizes ranging approximately between 20–100 nm. Slight agglomeration of nanoparticles was observed, which may be attributed to high surface energy and the presence of residual phytochemicals on the nanoparticle surface. The SEM images also showed rough and porous surface morphology, which is advantageous for catalytic applications due to the increased effective surface area and enhanced adsorption behavior.

VII. SPECTRAL ANALYSIS OF SELECTED COMPOUNDS

7.1 2-Phenyl-1H-benzimidazole

The FTIR spectrum of 2-Phenyl-1H-benzimidazole showed characteristic absorption bands at 3500, 1718, 1600, 948, and 740 cm^{-1}, confirming the presence of N–H, C=N, and aromatic functional groups. The ^{1}H NMR spectrum recorded in DMSO-d_6 displayed signals at δ 7.20–8.20 ppm corresponding to aromatic protons and a singlet at δ 12.02 ppm attributed to the N–H proton of the benzimidazole ring. The ^{13}C NMR spectrum exhibited signals at δ 115.16, 139.17, 129.97, 129.06, 128.88, 126.50, 122.16, and 151.03 ppm, confirming the aromatic carbon framework of the synthesized compound.

7.2 2-(4-Chlorophenyl)-1H-benzimidazole

The FTIR spectrum of 2-(4-Chlorophenyl)-1H-benzimidazole showed absorption bands at 3548, 1722, 1600, 1450, 1550, and 748 cm^{-1}, indicating the presence of characteristic benzimidazole functional groups. The ^{1}H NMR spectrum recorded in DMSO-d_6 showed aromatic proton signals at δ 7.21–8.18 ppm and doublets at δ 7.53 ppm and 7.66 ppm with coupling constants of J = 8.4 Hz, corresponding to substituted aromatic ring protons. A singlet observed at δ 12.94 ppm confirmed the presence of the N–H proton of the benzimidazole ring.

7.3 2-o-Tolyl-1H-benzimidazole

The ^{1}H NMR spectrum of 2-o-Tolyl-1H-benzimidazole recorded in DMSO-d_6 exhibited multiplet signals in the range of δ 7.82–7.45 ppm corresponding to aromatic protons. A singlet observed at δ 2.58 ppm was assigned to the methyl group attached to the aromatic ring, confirming the successful formation of the o-tolyl substituted benzimidazole derivative.

7.4 2-p-Tolyl-1H-benzimidazole

The ^{1}H NMR spectrum of 2-p-Tolyl-1H-benzimidazole displayed a broad singlet at δ 12.81 ppm corresponding to the N–H proton of the benzimidazole ring. Aromatic proton signals appeared at δ 8.06, 7.56, 7.36, and 7.19 ppm, while the singlet observed at δ 2.38 ppm was attributed to the methyl group attached to the para position of the aromatic ring. These spectral observations confirmed the successful synthesis of the p-tolyl substituted benzimidazole derivative.

DOI: 10.48175/IJARSCT-35660

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VIII. RESULTS AND DISCUSSION

8.1 Synthesis of Benzimidazole Derivatives

The catalytic efficiency of ZnO nanoparticles was evaluated using the condensation reaction between o-phenylenediamine and various substituted aromatic aldehydes.

The reactions proceeded efficiently under mild solvent-free conditions and afforded excellent yields within short reaction times.

8.2 Reaction Data

Entry	Aldehyde Substituent	Product	Time (min)	Yield (%)
1	H	2-Phenylbenzimidazole	10	92
2	p-NO_2	2-(4-Nitrophenyl)benzimidazole	10	94
3	p-CH_3	2-(4-Methylphenyl)benzimidazole	10	92
4	p-OMe	2-(4-Methoxyphenyl)benzimidazole	15	90
5	m-NO_2	2-(3-Nitrophenyl)benzimidazole	20	91
6	p-C_2H_5	2-(4-Ethylphenyl)benzimidazole	15	90
7	p-Cl	2-(4-Chlorophenyl)benzimidazole	10	90
8	Dichloro	Dichlorobenzimidazole derivative	20	91

The results indicate that both electron-donating and electron-withdrawing substituents effectively participate in the reaction.

Electron-withdrawing groups such as nitro substituents showed slightly higher yields due to enhanced electrophilicity of the aldehyde carbonyl group.

8.3 Catalyst Reusability Study

The recyclability of ZnO nanoparticles was investigated using the model reaction between benzaldehyde and o-phenylenediamine.

The catalyst was recovered by filtration, washed, dried, and reused.

Reusability Results

Cycle	Yield (%)
First	95
Second	88
Third	71

Although a gradual decrease in activity was observed, the catalyst retained appreciable catalytic efficiency after three cycles.

8.4 Advantages of the Developed Methodology

The present methodology offers several advantages:

- Environmentally friendly process
- Solvent-free reaction conditions
- Shorter reaction times
- High product yields
- Easy catalyst recovery
- Recyclable catalyst
- Mild reaction conditions

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- Reduced waste generation
- Operational simplicity

IX. CONCLUSION

The present investigation successfully demonstrates an eco-friendly and sustainable methodology for the synthesis of benzimidazole derivatives using green synthesized ZnO nanoparticles as an efficient heterogeneous catalyst.

ZnO nanoparticles were synthesized successfully using curry leaf extract through a green synthesis approach. The synthesized nanoparticles were characterized using UV–Visible spectroscopy, XRD, FTIR, and SEM analyses, confirming their nanoscale crystalline structure and catalytic potential.

The catalytic studies revealed that ZnO nanoparticles effectively promote the condensation reaction between o-phenylenediamine and substituted aromatic aldehydes under solvent-free conditions. The methodology afforded excellent yields within short reaction times while maintaining high selectivity.

The catalyst demonstrated appreciable recyclability and retained significant activity over multiple cycles. The developed protocol aligns well with the principles of green chemistry by minimizing hazardous chemicals, reducing solvent usage, and enabling catalyst reuse.
Overall, the study highlights the immense potential of green synthesized ZnO nanoparticles in sustainable organic synthesis and provides an efficient route for preparing biologically important benzimidazole derivatives with promising applications in medicinal chemistry and pharmaceutical research.

X. FUTURE SCOPE

Future investigations may focus on:

1. Large-scale industrial synthesis of benzimidazole derivatives.
2. Development of more efficient recyclable nanocatalysts.
3. Exploration of biological activities of synthesized compounds.
4. Use of alternative plant extracts for nanoparticle synthesis.
5. Application of ZnO nanocatalysts in multicomponent reactions.
6. Investigation of antimicrobial and anticancer properties of synthesized derivatives.
7. Computational studies and molecular docking investigations.

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