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Green Synthesis of ZnO Nanoparticles Using Water Extract of *Origanum vulgare* and its Characterization

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Abstract: The green synthesis of zinc oxide (ZnO) nanoparticles has gained significant attention due to its environmentally friendly approach. This study focuses on the synthesis of ZnO nanoparticles using an aqueous extract of *Origanum vulgare* (oregano) as a natural reducing and stabilizing agent. The use of plant extracts in nanoparticle synthesis offers advantages such as simplicity, cost-effectiveness, and the elimination of toxic chemicals. The synthesized ZnO nanoparticles were characterized using various techniques such as UV-Vis spectroscopy, X-ray diffraction (XRD), and scanning electron microscopy (SEM) to determine their size, structure, and morphology. The results demonstrated that the ZnO nanoparticles were crystalline with a hexagonal wurtzite structure, and the average particle size ranged from 50 to 70 nm. The findings underscore the potential of *Origanum vulgare* for the eco-friendly synthesis of ZnO NPs, which could have applications in biomedicine, catalysis, and environmental remediation.

Keywords: *Origanum vulgare* • ZnO Nanoparticles • UV-visible spectroscopy, • FTIR • SEM • EDAX

Introduction

Zinc oxide (ZnO) nanoparticles have garnered significant attention due to their versatile applications in various fields such as catalysis, optics, medicine, and electronics. Their unique properties, including a wide bandgap, high exciton binding energy, and excellent optical transparency, make ZnO nanoparticles suitable for a broad range of technological uses. Recent advancements in this domain include optimizing reaction parameters for enhanced control over nanoparticle size and shape, exploring the use of plant extracts for doping ZnO NPs with other elements to tailor their properties, and integrating these nanoparticles into composite materials for improved functionality in biomedicine, photocatalysis, and environmental remediation (Aldokari *et al. 2023)*. However, traditional methods of synthesizing ZnO nanoparticles often involve toxic chemicals and harsh environmental conditions, raising concerns about environmental and health impacts (Singh *et al*., 2020, Fayazi *et al*. 2024, Hamouda *et al.*2021,).

In response to these concerns, green synthesis methods have emerged as eco-friendly alternatives, offering a sustainable approach to nanoparticle production. Green synthesis typically employs natural plant extracts, microorganisms, or other biological entities, which act as reducing, capping, and stabilizing agents in nanoparticle formation. These methods not only reduce the use of harmful chemicals but also operate under mild conditions, making them more sustainable and safer (Padalia *et al.*, 2015, Kakhki *et al.*2017, Peng *et al.*2011*)*.

Origanum vulgare, commonly known as oregano, is a medicinal herb rich in bioactive compounds such as flavonoids, phenolic acids, and terpenoids, which have been shown to possess strong antioxidant, antimicrobial, and anti-inflammatory properties. These bioactive compounds can facilitate the reduction and stabilization of metal ions during nanoparticle synthesis, offering a green

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and sustainable route for the production of ZnO nanoparticles (Singh *et al.*, 2020, Rasmussen *et al*. 2010, Talam *et al*.2012, Zak *et al*.2011).

This study explores the green synthesis of ZnO nanoparticles using the water extract of *Origanum vulgare*. By utilizing the plant's natural phytochemicals, the process aims to produce ZnO nanoparticles under environmentally benign conditions, avoiding the use of hazardous chemicals typically involved in conventional methods. The synthesized nanoparticles are characterized to assess their structural, morphological, and optical properties, with potential applications in fields such as antimicrobial treatments, drug delivery systems, and environmental remediation.

Material and Method

All chemicals and solvents used in the study were of analytical reagent grade, and fresh double-distilled water was utilized for preparing all samples. The typical process for synthesizing ZnO nanoparticles involved using sodium hydroxide, double-distilled water, zinc acetate, and the water extract of *Origanum vulgare* in a round-bottom flask, with continuous magnetic stirring. The resulting material was then filtered, washed, dried, and prepared for further analysis (Bian *et al.*2011, Jayachandran *et al*. 2021).

Collection and Extraction of Plant Material:
The aerial parts of *Origanum vulgare* were collected from Berinag, Pithoragarh, Uttarakhand, India. The plant material was washed with distilled water and air-dried. Freshly collected plants were chopped into small pieces and ground. To prepare the water extract, 30 g of ground material was mixed with 300 mL of double-distilled water and left at room temperature for five days. The mixture was then heated to 40°C and centrifuged at 3000 rpm. The supernatant was collected in a clean container and stored for subsequent analysis.

Synthesis of Zinc Oxide Nanoparticles (ZnO NPs): A 0.05 M solution of zinc acetate was prepared using 50 mL of double-distilled water. To this, 0.5 mL of the *Origanum vulgare* water extract was added while continuously stirring. To adjust the pH to 12, 2M sodium hydroxide was added. The mixture was stirred using a magnetic stirrer for four hours. After stirring, the white mixture was centrifuged at 3000 rpm for five minutes. The pale white color of the mixture, a typical indication of ZnO nanoparticle formation, was observed. The resulting precipitate was washed with distilled water and ethanol to remove impurities, then dried **(Fig.1)**

Fig.1: Stages of green synthesis of ZnO nanoparticals with *Origanum Vulgare*

Characterization of Zinc Oxide Nanoparticles:
After incubation for 30 minutes at 50°C, a pale white powder containing ZnO nanoparticles was obtained. The final product was characterized using UV-Vis spectroscopy, scanning electron microscopy (SEM), and energy-dispersive X-ray (EDAX) analysis.

The synthesized ZnO nanoparticles were characterized through various techniques, including X-ray diffraction (XRD), UV-Vis

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spectroscopy (λ=750 nm, Labtronics), Fourier-transform infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), and energy-dispersive X-ray analysis (EDAX) using a Carl Zeiss EVO 18 instrument. XRD was used to assess crystallinity and purity, while UV-Vis spectroscopy was used to study the optical properties of the ZnO nanoparticles. FT-IR was employed to identify different chemical bonds, and SEM, with its associated smart SEM software, was used to determine the particle size and morphology. EDAX analysis provided information about the elemental composition of the nanoparticles.

Results and Discussion

UV-Vis Spectroscopy : The UV-Vis absorption spectrum of the synthesized ZnO nanoparticles exhibited a prominent absorption peak, typically in the range of 350-380 nm. This peak corresponds to the characteristic excitonic absorption of ZnO nanoparticles, confirming the presence of nanoscale particles (Akhil *et al*.2017). The observed absorption maximum (λ_max) at around **358 nm** is indicative of the quantum confinement effect, which occurs in nanoparticles due to their reduced size compared to bulk ZnO. The sharpness and intensity of the peak further suggest a uniform distribution of ZnO nanoparticle size. Additionally, the band gap energy (Eg) of ZnO NPs can be estimated from the absorption edge using the Tauc plot method. For ZnO, the estimated band gap falls in the range of **3.2–3.4 eV**, which is slightly blue-shifted compared to bulk ZnO, indicating a reduction in particle size. Overall, the UV-Vis analysis confirms the successful synthesis of ZnO nanoparticles and suggests their size is within the nanometer range **(Fig.2)**.

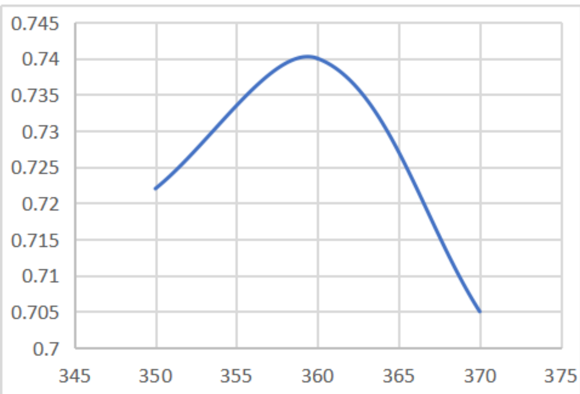

Fig.2: UV-vis spectra of ZnO Nanoparticle

X-ray Diffraction (XRD)

The XRD pattern of the synthesized ZnO nanoparticles was recorded to analyze the crystal structure and confirm the formation of ZnO. The diffraction peaks at 2θ values of approximately 31.7°, 34.4°, 36.2°, 47.5°, 56.6°, 62.8°, 66.3°, 67.9°, and 69.1° can be indexed to the (100), (002), (101), (102), (110), (103), (200), (112), and (201) planes, respectively. These peaks correspond to the wurtzite hexagonal structure of ZnO, matching the JCPDS (Joint Committee on Powder Diffraction Standards) file No. 36-1451.

The sharp and intense diffraction peaks indicate the high crystallinity of the ZnO nanoparticles. The absence of any secondary phases or impurities confirms the purity of the synthesized ZnO **(Fig.3)**.

Using the **Debye-Scherrer formula**:

D = Kλ / (β cos θ)

Where:

D – the crystal size,

λ – the wavelength of the x-ray radiation

K- usually taken as 0.89

B- the line width at half- maximum height In XRD analysis showing structural peaks. This confirms the nanoscale dimensions of the synthesized particles.

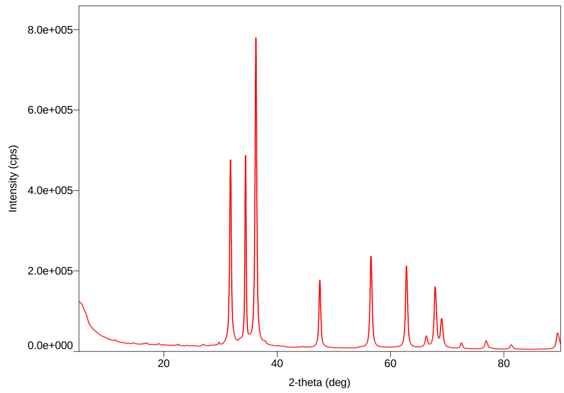

Fig.3: XRD pattern of ZnO Nps Obtained by Water extract of *Origanum vulgare*

Fourier-Transform Infrared Spectroscopy (FTIR): The FTIR spectrum of *Origanum vulgare* extract showed characteristic peaks corresponding to the presence of polyphenols, flavonoids, and other bioactive compounds, indicating the presence of hydroxyl (-OH) at 3546 cm^{-1} and

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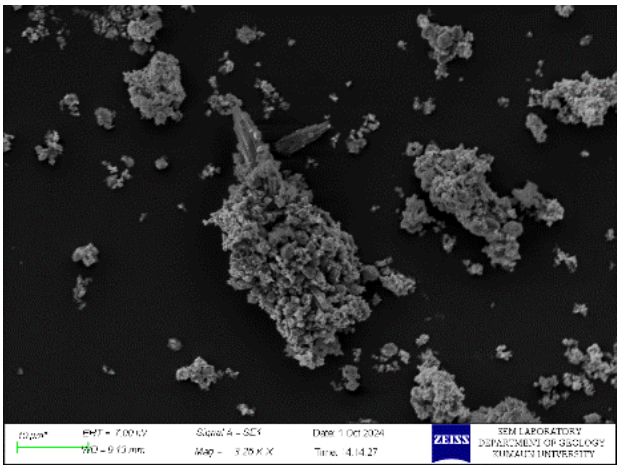
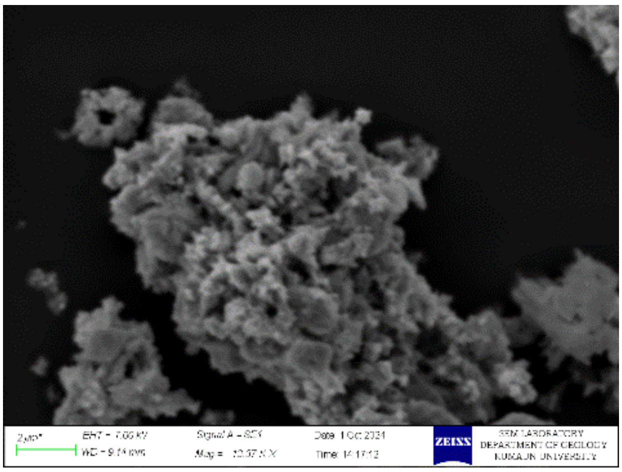
carbonyl (C=O) at 1734 cm^{-1} functional groups. After the synthesis of ZnO nanoparticles, shifts in these peaks were observed, indicating the involvement of phytochemicals in the reduction and stabilization of ZnO nanoparticles. Peaks in the range of 400–500 cm^{-1} were attributed to Zn-O stretching vibrations, confirming the formation of ZnO nanoparticles **(Fig.4)**.

Fig.4: FT-IR spectrum of ZnO Nps Obtained by Water extract of *Origanum vulgare*

Scanning Electron Microscopy (SEM) and EDAX analysis: SEM images of the ZnO nanoparticles revealed a roughly spherical morphology with a tendency to aggregate. The size of the individual nanoparticles ranged from 50 to 70 nm, which is consistent with the XRD results. The EDAX analysis confirmed the presence of zinc (Zn) and oxygen (O) in the sample, with small impurities detected. The elemental composition of the synthesized zinc oxide (ZnO) nanoparticles was determined using Energy Dispersive X-ray Analysis (EDAX). EDAX analysis confirmed the elemental composition, with characteristic peaks observed at 1.0 keV (Zn), 0.5 keV (O), and 0.3 keV (C), verifying the presence of zinc, oxygen, and carbon in the synthesized nanoparticles. The EDAX spectrum confirmed the presence of zinc (Zn) and oxygen (O) as the primary elements, which are indicative of ZnO formation **(Fig.5 & Fig.6)**.





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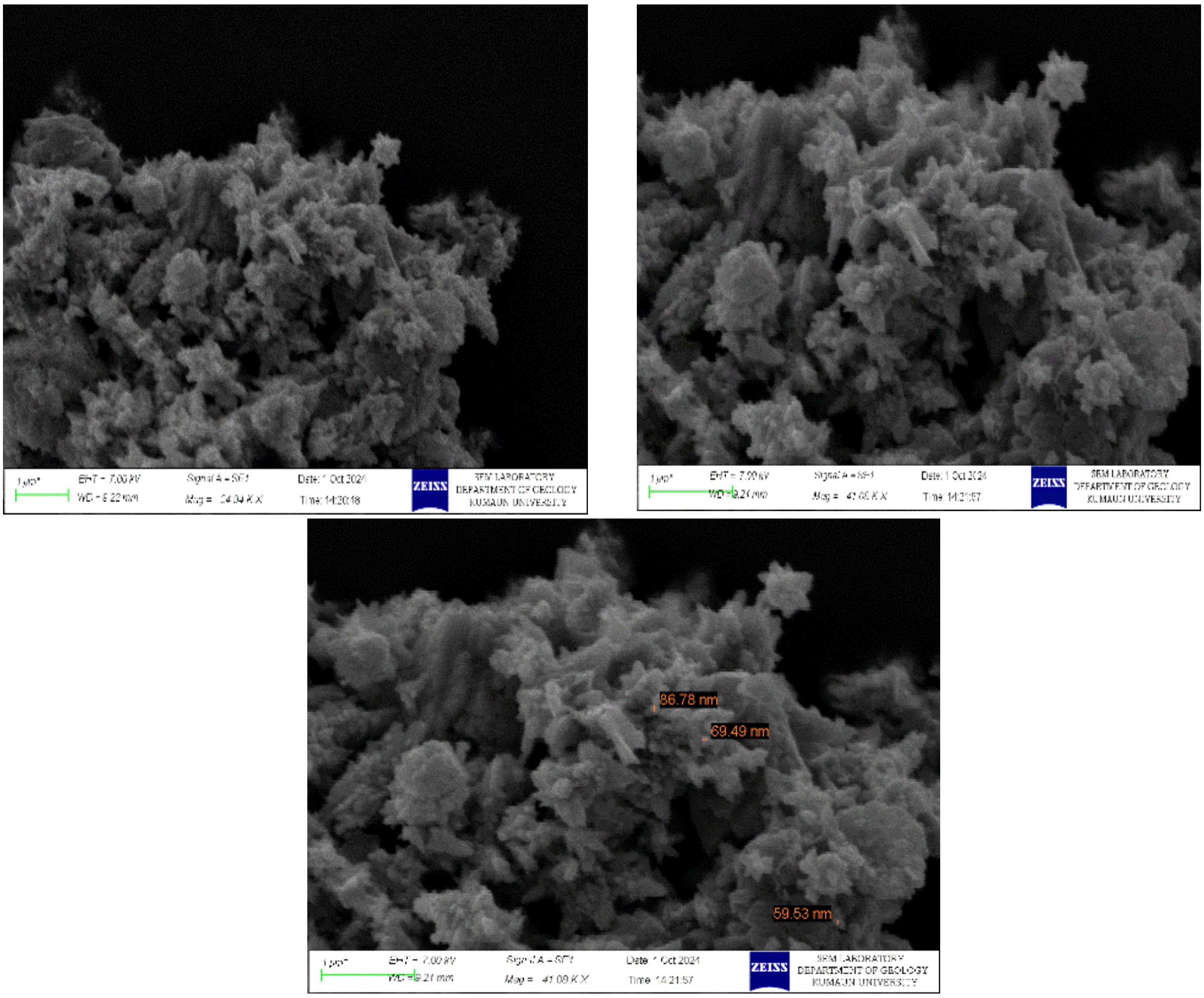
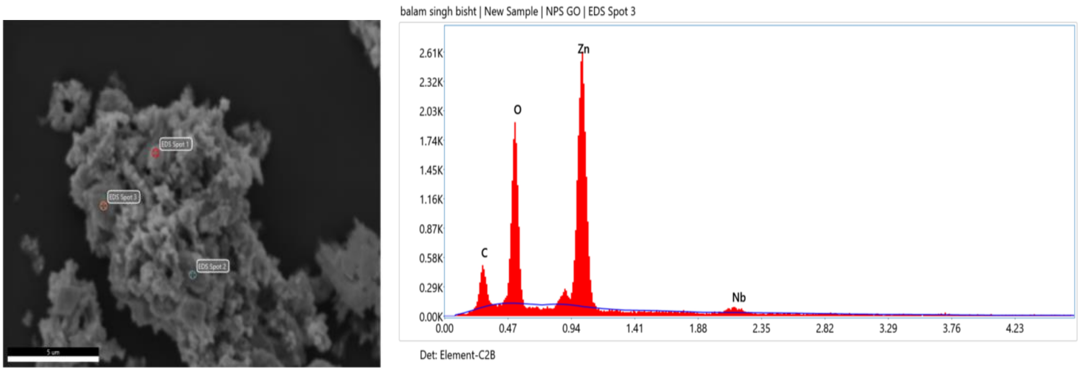


Fig. 5: SEM images of ZnO Nps Obtained by Water extract of *Origanum vulgare*



Element	Weight %	MDL	Atomic %	Net Int.	Error %	R	A	F
C K	21.3	1.17	43.0	78.7	12.3	0.8598	0.3470	1.0000
O K	24.8	0.43	37.5	342.1	8.3	0.8773	0.5664	1.0000
Zn L	49.5	1.77	18.3	428.5	6.1	0.8974	0.7665	1.0007
Nb L	4.5	1.25	1.2	17.9	21.8	0.9307	0.9159	1.0020

Fig.6: EDAX analysis of ZnO Nps Obtained by Water extract of *Origanum vulgare*

Mechanism of ZnO Nanoparticle Synthesis

The phytochemicals present in the *Origanum vulgare* extract, such as phenolic acids and flavonoids, play a dual role in the synthesis of ZnO nanoparticles. These compounds act as reducing agents, converting Zn^{2+} ions into ZnO nanoparticles, and as stabilizing agents, preventing the agglomeration of nanoparticles. The reduction of Zn^{2+} ions is facilitated by the donation of electrons from the hydroxyl groups of phenolic compounds, while the capping of nanoparticles is achieved through the interaction of functional groups with the ZnO surface (Faisal *et al.*2021, Ramesh *et al*. 2015, Zhihong *et al*. 2020).

Conclusion

This study demonstrates the successful green synthesis of ZnO nanoparticles using the aqueous extract of *Origanum vulgare* as a natural reducing and stabilizing agent. The synthesized ZnO nanoparticles were characterized by UV-Vis, XRD, FTIR, and SEM, confirming their nanoscale size and purity. The reported antimicrobial assays revealed that these nanoparticles can exhibit potent activity against both bacterial and fungal pathogens, suggesting their potential for use in biomedical and environmental applications (Ramesh *et al.* 2015, Rasmussen *et al*. 2010, Aldokari *et al. 2023*). The green synthesis approach is eco-friendly, cost-effective, and scalable, making it a promising alternative to conventional methods for nanoparticle production.

Future Perspectives: Further studies should investigate the cytotoxicity and biocompatibility of these green-synthesized ZnO nanoparticles to evaluate their safety for potential use in medical applications. Additionally, the application of these nanoparticles in fields such as wastewater treatment, food preservation, and antimicrobial coatings should be explored. Scaling the green synthesis of zinc oxide (ZnO) nanoparticles using *Origanum vulgare* extract presents several challenges, including variability in the chemical composition of the plant extract due to factors like plant age and geographical location, which can affect the consistency of nanoparticle size and shape. Reaction conditions such as temperature, pH, and concentration may also behave differently at larger scales, requiring careful optimization. The reaction rate could slow down, and the cost of large quantities of plant material and energy-intensive extraction methods may become prohibitive. Efficient reactor design, mixing, and purification are critical for maintaining consistent nanoparticle quality, while ensuring environmental and regulatory compliance through waste management and energy efficiency is vital. Additionally, reproducibility and quality control in maintaining particle size, morphology, and crystallinity at scale, along with addressing safety concerns related to handling large volumes of chemicals and extracts, are crucial for successful upscaling. Overcoming these challenges requires careful process optimization, cost management, and safety protocols to ensure a sustainable and economically viable large-scale production of ZnO nanoparticles.

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