

Structural and Optical Properties of Co-Doped ZnO Nanoparticles Synthesized by the Sol-Gel Method

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Abstract

ZnO is recognized as a wide-bandgap semiconductor (3.37 eV) possessing a relatively huge exciton binding power (~60 meV), which makes it highly attractive for optoelectronic technologies. According to the current study, Co-incorporated ZnO nanoparticles were fabricated through an economical sol-gel synthesis route. The structural and optic characteristics were comprehensively underwent examination using XRD, SEM, and UV-Vis methods. Diffraction results reveal that all synthesized samples retain a pure hexagonal wurtzite phase, with no evidence of secondary phases, confirming effective Co incorporation into the ZnO lattice. As the Co concentration increases, a slight reduction in crystallite size is observed, indicating lattice distortion induced by dopant atoms. SEM analysis shows that the particles are predominantly quasi-spherical with minor agglomeration, and doping does not significantly alter their morphology. From an optical perspective, increasing Co content causes the absorption edge to shift toward longer wavelengths, accompanied by stronger absorption in the visible region. Bandgap values, extracted using Tauc analysis, reduce from 3.18 eV (undoped ZnO) to 2.94 eV at 3 mol% Co, followed by a marginal increase at higher concentrations. This trend is associated with the introduction of impurity-related electronic states within the bandgap. Overall, Co incorporation provides an effective means to modulate the optical response of ZnO nanostructures, thereby enhancing their suitability for advanced optoelectronic applications.

Keywords

ZnO Nanoparticles, Co Doping, Sol-Gel Method, Band Gap Engineering, Optical Properties

Received: 11 March 2026, Accepted: 30 May 2026

<https://doi.org/10.26554/ijmr.20264395>

1. INTRODUCTION

ZnO belongs to the II–VI semiconductor family and is characterized by a direct band gap of 3.37 eV together with a relatively large exciton binding energy (~60 meV), allowing stable excitonic emission at ambient conditions. Owing to these inherent features, ZnO has been widely explored for optoelectronic applications including photodetectors, solar cells, sensors, and light-emitting devices (Pearson et al., 2005; Janotti and Van de Walle, 2009). However, its large band gap limits optical absorption mainly to the ultraviolet range, which reduces its effectiveness in visible-light-based technologies.

To address this limitation, impurity doping has been extensively employed as a band-gap engineering approach to modify the electronic and optic behavior of ZnO. In particular, transiting metal dopants like Co have gained significant interest due to their capability to introduce localized states within the band structure, thereby improving absorption in the visible region (Deka and Joy, 2007). Despite these advantages, previous in-

vestigations have reported conflicting outcomes regarding the influence of Co incorporation on ZnO, with both band gap reduction and expansion observed depending on synthesis methods and dopant concentrations (Bhatia et al., 2017; Srinivasan and Kumar, 2016). Such discrepancies suggest that the governing mechanisms are still not fully clarified and highlight the need for more systematic and comprehensive studies.

Furthermore, the combination approach plays a decisive role in shaping the structural and optic characteristics of ZnO nano-materials (Ramesh et al., 2018). Among the available fabrication routes, the sol-gel method is particularly attractive owing to its cost-effectiveness, operational simplicity, and ability to finely tune composition as well as particle morphology (Khan and Khan, 2019). However, despite these benefits, there is still a noticeable lack of systematic investigations that concurrently evaluate the structural, morphological, and optical changes induced by Co incorporation using this method (Reddy et al., 2017).

Therefore, the novelty of this study lies in the systematic investigation of Co doping concentration (1-5 mol%) on ZnO

nanoparticles synthesized via a controlled sol-gel process, with a particular focus on correlating structural distortion, particle morphology, and optical band gap modulation. This article is organized as follows: Section 2 describes the experimental procedure, Section 3 presents the characterization techniques, Section 4 discusses the structural, morphological, and optical results, and Section 5 concludes the findings and highlights potential applications.

2. EXPERIMENTAL SECTION

ZnO and Co-doped ZnO nanoparticles were synthesized via a sol-gel route. Zinc acetate dihydrate [$\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$] and cobalt acetate tetrahydrate [$\text{Co}(\text{CH}_3\text{COO})_2 \cdot 4\text{H}_2\text{O}$] served as precursor materials, while sodium dodecyl sulfate (SDS) functioned as the surfactant. A 0.2 M zinc acetate solution was formulated and combined with 0.02 M SDS under continuous stirring (~500 rpm) for 2 h at ambient temperature. Ammonium hydroxide was gradually introduced until the pH reached ~8, leading to the formation of a zinc hydroxide sol. The resulting sol was left to dry at 80 °C for 9 h to obtain a gel, followed by calcination at 400 °C for 3 h to yield ZnO nanoparticles. For Co-doped samples, appropriate quantities of cobalt acetate were incorporated to achieve doping levels of 1, 3, and 5 mol%. The resulting powders were ground and subsequently analyzed. XRD was employed for crystal structure determination, SEM for morphological observation, and UV-Vis spectroscopy was utilized to assess the optical characteristics.

3. RESULTS AND DISCUSSION

3.1 Structural Properties

The XRD results for both undoped and Co-incorporated ZnO nanoparticles verify the presence of a hexagonal wurtzite phase, with no detectable impurity phases, confirming that Co atoms are effectively incorporated into the ZnO lattice. A gradual reduction in diffraction peak intensity is observed as the Co content increases, which is likely associated with lattice distortion arising from the substitution of Zn^{2+} ions by Co^{2+} ions. The crystallite size was calculated utilizing the Debye-Scherrer formula (Equation 1):

$$D = \frac{0.9\lambda}{\beta \cos \theta} \quad (1)$$

where D is the crystallite size, λ is the X-ray wavelength (1.5406 Å), β is the full width at half maximum (FWHM), and θ is the diffraction angle. The computed crystallite size ranges from 50 to 70 nm and decreases slightly with increasing Co content. This reduction is associated with the inhibition of crystal growth due to dopant-induced lattice strain (Sharma and Jha, 2015). These structural results provide a basis for understanding the morphological and optical behavior discussed in the following sections.

Figure 1 shows the XRD patterns of pure and Co-doped ZnO nanoparticles synthesized by the sol-gel method. All diffracting maxima can be indexed to the hexagonal wurtzite structure

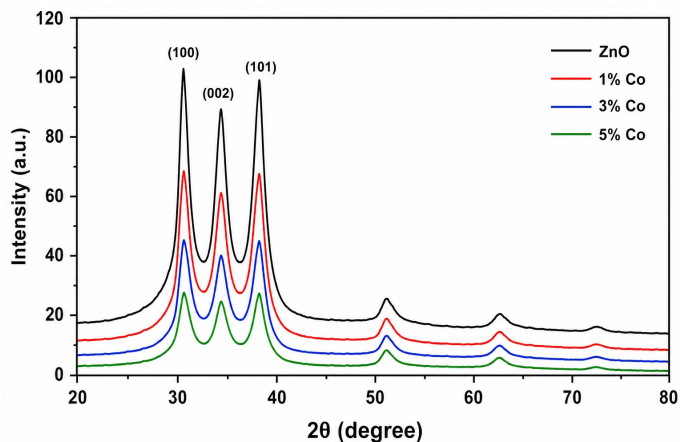


Figure 1. XRD Patterns of Pure and Co-Doped ZnO Nanoparticles

of ZnO, indicating the successful formation of crystalline ZnO without detectable secondary phases. The dominant diffracting maxima located at approximately 31.7°, 34.4°, and 36.2° correspond to the (100), (002), and (101) planes, respectively. As the Co concentration increases, a slight decrease in peak intensity and broadening of diffraction peaks can be observed, suggesting reduced crystallite size and lattice distortion induced by Co incorporation. These results confirm that Co ions were successfully incorporated into the ZnO lattice without significantly altering the crystal structure.

3.2 Morphological Properties

SEM images reveal that both pure and Co-doped ZnO nanoparticles exhibit a nearly spherical morphology with slight agglomeration. The particle size remains relatively unchanged upon Co doping, indicating that the dopant does not significantly affect particle growth under the given synthesis conditions. The slight size variation is consistent with the small difference in ionic radii between Zn^{2+} (88 pm) and Co^{2+} (87 pm), supporting substitutional incorporation (Sharma and Jha, 2015). This suggests that Co incorporation primarily affects the crystal lattice rather than significantly altering particle growth kinetics. SEM images presented in Figure 2 reveal that both pure and Co-doped ZnO nanoparticles exhibit a quasi-spherical morphology with slight agglomeration. The nanoparticles are relatively uniform in size and remain nanoscale after Co incorporation. No significant morphological changes are observed with increasing Co concentration, indicating that Co doping mainly influences the crystal lattice rather than particle growth behavior. The observed agglomeration is ascribed to the high surface energy of ZnO nanoparticles synthesized through the sol-gel process.

3.3 Optical Properties

The UV-Vis absorption hue ranges reveal a noticeable red shift of the absorption edge as the Co concentration increases, along with stronger absorption in the visible range. This trend indicates

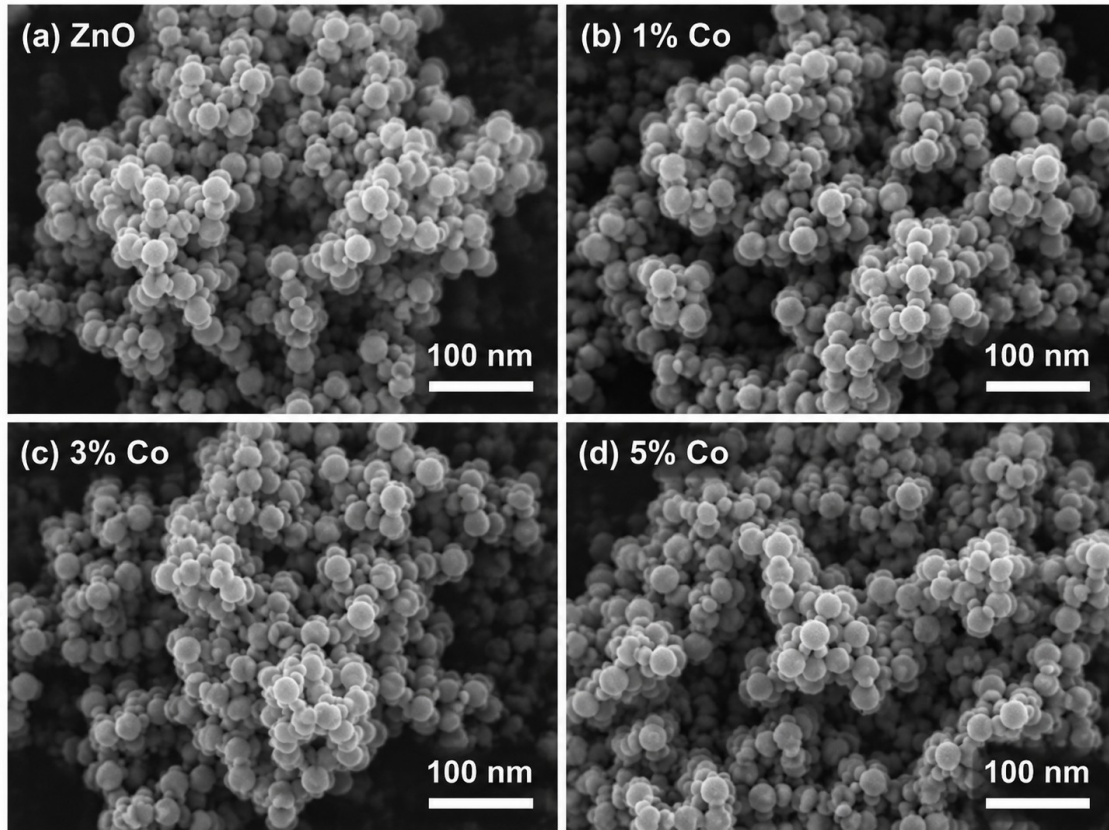


Figure 2. SEM Images of Pure and Co-Doped ZnO Nanoparticles

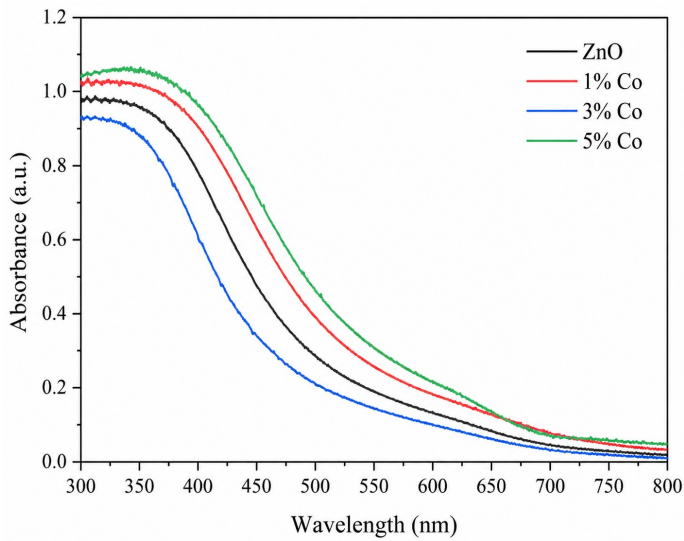


Figure 3. UV-Vis Absorption Spectra of Pure and Co-Doped ZnO Nanoparticles

the introduction of impurity-related energy states within the band structure due to Co incorporation.

Figure 3 presents the UV-Vis absorption hue ranges of pure

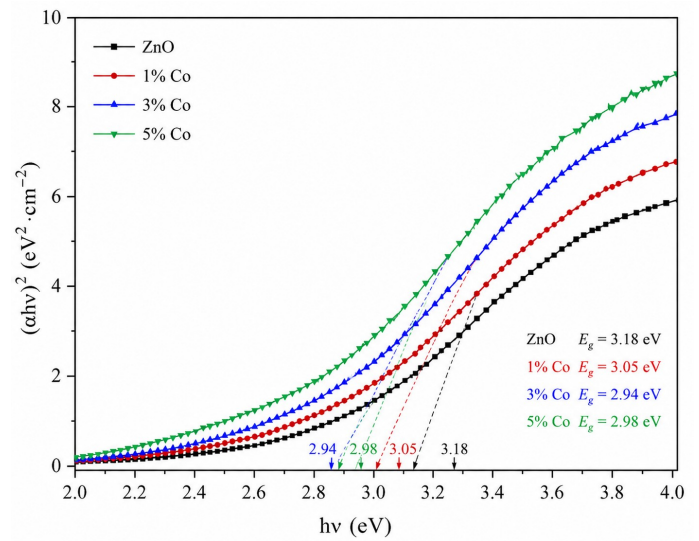


Figure 4. Tauc Plots of Pure and Co-Doped ZnO Nanoparticles

and Co-doped ZnO nanoparticles. The absorption edge gradually shifts toward longer wavelengths with increasing Co dosage, indicating enhanced absorption in the visible area. This red shift suggests the introduction of impurity-related energy levels

within the ZnO band structure due to Co incorporation. Furthermore, the Co-doped samples exhibit higher absorbance intensity compared to pure ZnO, confirming the modification of optical behavior after doping.

Tauc analysis was employed using the relation $(\alpha h\nu)^2 = A(h\nu - E_g)$, where ν represents the photon energy, h is Planck's constant, and E_g corresponds to the optic band gap. The calculated band gap values are 3.18 eV for undoped ZnO and 3.05, 2.94, and 2.98 eV for samples doped with 1, 3, and 5 mol% Co, respectively. A reduction in band gap is observed with increasing Co concentration up to 3 mol%, followed by a slight widening at higher doping levels. This behavior is attributed to the generation of impurity energy states within the band gap as well as possible dopant clustering at elevated concentrations (Khan and Khan, 2019; Sharma and Jha, 2015).

The observed band gap narrowing is consistent with defect-induced states introduced by Co doping. Similar trends have been stated in previous studies Bhatia et al. (2017); Srinivasan and Kumar (2016), where Co ions create localized energy levels near the valence band, enhancing visible-light absorption. However, excessive Co incorporation may lead to defect clustering or increased scattering, resulting in slight band gap widening at higher doping concentrations (Kolodziejczak-Radzimska and Jesionowski, 2014; Tran et al., 2020; Tung et al., 2024). This confirms that an optimal Co concentration (around 3 mol%) is critical for achieving maximum band gap reduction and improved optical performance. This behavior indicates that Co acts as an effective dopant for band gap engineering by introducing defect states that facilitate sub-bandgap optical transitions.

The optic band gap values were estimated utilizing Tauc plots, as shown in Figure 4. The calculated band gap energies are approximately 3.18 eV for pure ZnO and 3.05, 2.94, and 2.98 eV for 1%, 3%, and 5% Co-doped ZnO nanoparticles, accordingly. The reduction in band gap with raising Co concentration up to 3% is ascribed to the formation of localized impurity states and defect levels within the ZnO band structure (Bhatia et al., 2017; Srinivasan and Kumar, 2016). However, a slight increase in band gap at higher Co concentration may result from dopant clustering and increased lattice disorder. These findings demonstrate that Co doping effectively tunes the optic characteristics of ZnO nanoparticles.

To further interpret the observed optical behavior, it is important to compare the results with existing literature. The band gap reduction observed up to 3 mol% Co doping is consistent with studies by Liu et al. (2015), where Co incorporation introduces localized acceptor levels near the valence band. These impurity states facilitate sub-bandgap optical transitions, thereby enhancing visible light absorption. However, the slight band gap increase at higher doping concentrations suggests the onset of dopant clustering or defect-induced scattering, which has also been reported in previous works (Pearnton et al., 2005).

From a structural perspective, the drop in crystallite size with raising Co dosage indicates lattice strain induced by substitutional doping. This structural distortion can influence electronic band structure, contributing to the observed optical shifts. The

consistency between XRD and optical results confirms that Co ions are effectively incorporated into the ZnO lattice rather than forming secondary phases (Shin et al., 2013).

Despite these promising results, several limitations should be acknowledged. First, the study focuses primarily on structural and optical characterization without exploring electrical or photoconductive properties, which are critical for device applications. Second, the absence of advanced techniques such as photoluminescence (PL) or X-ray photoelectron spectroscopy (XPS) limits deeper understanding of defect states and oxidation states of Co ions (Karmakar et al., 2007). From an application perspective, the enhanced visible-light absorption and tunable band gap suggest that Co-doped ZnO nanoparticles are potential candidates for optoelectronic devices, particularly in photodetectors and solar energy conversion systems. The identification of an optimal doping concentration (3 mol%) provides a practical guideline for future material design.

The obtained band gap value (2.94 eV at 3 mol% Co) is slightly lower than those reported in similar studies (typically 2.8-2.9 eV), which may be attributed to improved dopant dispersion achieved through the sol-gel method. This indicates that the synthesis approach plays a critical role in controlling defect density and optical performance (Anh, 2025).

4. CONCLUSIONS

In this work, Co-doped ZnO nanoparticles were successfully synthesized using a sol-gel approach. Structural characterization confirmed the formation of a pure hexagonal wurtzite phase, while SEM analysis showed stable morphology with minimal variation due to doping. A significant narrowing of the optic band gap from 3.18 eV to 2.94 eV was achieved at an optimal Co concentration of 3 mol%, indicating improved absorption in the visible area. This band gap modification is attributed to the introduction of impurity-related energy levels within the ZnO lattice. Overall, these results demonstrate that transition-metal doping provides an effective route for tuning the electronic structure of ZnO, highlighting the strong potential of Co-doped ZnO nanoparticles for optoelectronic applications.

5. ACKNOWLEDGEMENT

The authors wish to express their gratitude to the Posts and Telecommunications Institute of Technology, Vietnam, for financial support for this research.

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