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Synthesis, Photophysical Characterization and Antimicrobial Study of Zn(II) Complexes of N,N'-Bis(salicylidene)ethylenediamine Schiff Base

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ABSTRACT: Zn(II) complex of the Schiff base ligand N,N′-Bis(salicylidene)ethylenediamine (SBL1) was successfully synthesized. XRD and SEM analyses confirmed the formation of a crystalline complex with petal-like morphology, while Raman and UV–Vis spectra indicated coordination of Zn^{2+} to azomethine nitrogen and phenolic oxygen, resulting in enhanced conjugation and ligand-to-metal charge transfer (LMCT) transitions. Photoluminescence studies exhibited dual emission peaks at 542 nm and 604 nm upon excitation at 225 nm, suggesting efficient $\pi \rightarrow \pi^*$, $n \rightarrow \pi^*$ and LMCT processes. Antimicrobial evaluation against Bacillus subtilis and Aspergillus niger demonstrated moderate dose-dependent activity, with inhibition zones increasing with concentration, highlighting the enhanced biological efficacy of the Zn(II) complex.

KEY WORDS: Photophysical Study; Antimicrobial Study; Metal Complex; Schiff Base

I. INTRODUCTION

Schiff base metal complexes have attracted significant scientific interest due to their versatile coordination chemistry and broad range of applications in catalysis, materials science and biomedical research. Among them, complexes of N,N′-Bis(salicylidene)ethylenediamine (salen-type ligands) are particularly notable because of their structural flexibility, thermal stability and ability to coordinate with various transition metal ions. Zinc(II), being a biologically important and nontoxic metal, forms stable complexes with Schiff bases and often enhances their optical and antimicrobial properties.

Recent studies have explored metal complexes of Schiff bases as promising antimicrobial agents due to their enhanced biological efficacy. Obonova et al. synthesized Zn(II) complexes from reduced Schiff bases derived from cyclohexane-1,2-diamine and fluorinated benzaldehydes, reporting significantly higher antibacterial activity, particularly for the para-CF_3 substituted complex. Their results demonstrated a synergistic effect between Zn(II) ion coordination and ligand structure, yielding potency comparable to ciprofloxacin against S. aureus and E. coli [1]. Kargar et al. reported the synthesis of binuclear Zn(II) Schiff base complexes using ONNO-donor tetradentate ligands derived from halogen-substituted salicylaldehydes, confirming their structures through FTIR, 1H NMR, elemental analysis and DFT calculations. The complexes exhibited a centrosymmetric dimeric architecture featuring μ-phenoxo bridges and five-coordinate Zn(II) centers. Antimicrobial screening demonstrated that the Zn complexes possessed significantly enhanced biological activity compared to the free ligands, evidencing a strong metal–ligand synergistic effect [2]. Joseyphus and Nair synthesized Zn(II) complexes of Schiff bases derived from glycylglycine with imidazole-2-carboxaldehyde and indole-3-carboxaldehyde and characterized them by spectroscopic and analyt ical techniques. Their antimicrobial evaluation revealed that the Zn(II) complexes showed stronger antibacterial and antifungal activities compared to the free ligands, particularly against S. aureus, E. coli, K. pneumoniae and A. niger. The study highlighted that zinc coordination significantly enhances biological efficacy, confirming metal–ligand synergism in microbial growth inhibition [3]. Gusev et al. synthesized and structurally characterized Zn(II) Schiff base complexes derived from 4-{(E)-[(2-fluorophenyl)imino]methyl}-5-methyl-2-phenyl-2,4-dihydro-3H-pyrazol-3-one, forming solvate variants through reactions in ethanol and acetonitrile. Their study revealed strong solvent-dependent solid-state fluorescence, supported by UV-Vis, emission spectroscopy and DFT calculations. Bader's topological analysis further clarified electronic structures and non-covalent interactions, demonstrating that solvate environment influences photophysical behavior [4]. Gusev et al. synthesized pyrazolone-based Zn(II) Schiff base complexes that show strong blue photoluminescence both

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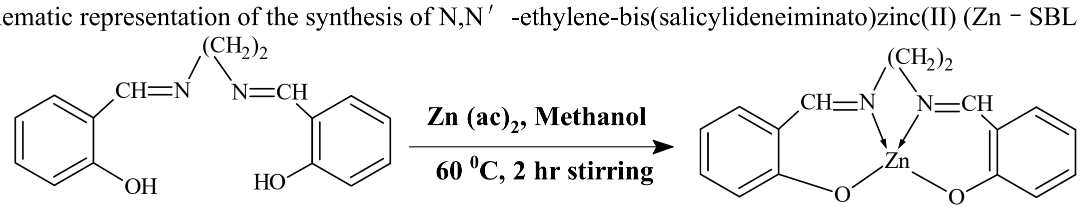
in solution and the solid state, owing to intraligand charge transfer. They constructed organic light-emitting diodes using these complexes, achieving low operating voltages (3.2–4.0 V), brightness up to 11600 cd/m², and external quantum efficiency of ~3.2 %. The high thermal stability and efficient luminescence make these Zn(II) Schiff base complexes promising emitters for purely fluorescent blue OLEDs [5].

In the present work, Zn(II) complex derived from the Schiff base ligand N,N′ -Bis(salicylidene)ethylenediamine was successfully synthesized and characterized through various spectroscopic and analytical techniques, confirming their structural integrity and coordination behavior. The photophysical studies revealed notable optical responses. Furthermore, the antimicrobial evaluation demonstrated that the Zn(II) complexes exhibited enhanced antibacterial and antifungal activity.

II. EXPERIMENTAL DETAILS

The zinc(II) complex of the Schiff base ligand SBL1 was synthesized through a direct complexation route. The previously prepared ligand N,N′ -Bis(salicylidene)ethylenediamine (SBL1) (1.0 mmol) was dissolved in 30 mL of methanol and stirred continuously for 20 minutes to ensure complete solubilization. A methanolic solution of zinc(II) acetate dihydrate, $Zn(CH_3COO)_2 \cdot 2H_2O$ (1.0 mmol in 15 mL MeOH), was added slowly dropwise to the ligand solution under constant stirring. The reaction mixture was then refluxed at 60 °C for 3 hours to promote coordination between the metal ion and the ligand.

A gradual change in solution color from pale yellow to light cream was observed, indicating the formation of the Zn–SBL1 complex. Upon completion of reflux, the mixture was allowed to cool naturally to room temperature. The obtained solid precipitate was isolated by filtration and washed thoroughly with cold ethanol and diethyl ether to remove unreacted metal salts and ligand residues. The product was dried in an oven at 60 °C for 6 hours and subsequently stored in an air-tight desiccator. The resulting complex is a pale yellow, crystalline solid, stable to atmospheric moisture. Figure 1 shows the schematic representation of the synthesis of N,N′ -ethylene-bis(salicylideneiminato)zinc(II) (Zn – SBL1).



bis(salicylidene)ethylene-1,2-diamine (salen) **N,N′-ethylene-bis(salicylideneiminato)zinc(II)**

Figure 1: ***Schematic representation of the synthesis of N,N′ -ethylene-bis(salicylideneiminato)zinc(II) (Zn – SBL1).***

The synthesized Zn–SBL1 was characterized using various spectroscopic and analytical techniques to confirm their structural, morphological and optical properties. The crystalline nature of the materials was examined by X-ray diffraction (XRD) using a Rigaku MiniFlex 600 X-ray Diffractometer, operated with Cu-Kα radiation (λ = 1.5406 Å). The surface morphology was studied using Field-Emission Scanning Electron Microscopy (FE-SEM), performed on a JEOL JSM-7600F FE-SEM instrument. Raman spectroscopy measurements were carried out using a Bruker MultiRAM FT-Raman spectrometer system to analyze vibrational characteristics and confirm structural bonding features. UV–Visible absorption spectra of the ligand and metal complex were recorded using a Shimadzu UV-1800 UV–Vis Spectrophotometer. Photoluminescence (PL) spectroscopy was employed to investigate the emission characteristics of the complex. The PL spectra were recorded on a Fluorescence Spectrophotometer (Model: HITACHI F-7000). The antibacterial study of Zn–SBL1 was tested against Bacillus subtilis and antifungal study was evaluated against Aspergillus niger with corresponding inhibition zone measurements.

III. RESULTS AND DISCUSSION

Figure 2 shows the (a) XRD pattern and (b) SEM images of Zn–SBL1. The XRD pattern (Figure 2a) of the Zn–SBL1 complex displays distinct diffraction peaks at 2θ values of 17.7°, 20.1°, 23.3° and 26.3°. The appearance of these sharp peaks confirms the successful coordination of Zn(II) to the Schiff base ligand (SBL1) and indicates an enhancement in crystallinity [6]. The corresponding SEM image (Figure 2b) reveals a petal-like morphology, consisting of randomly oriented, irregularly sized particles distributed over the surface. This distinctive surface topology suggests the growth of aggregated crystalline domains following metal complexation.

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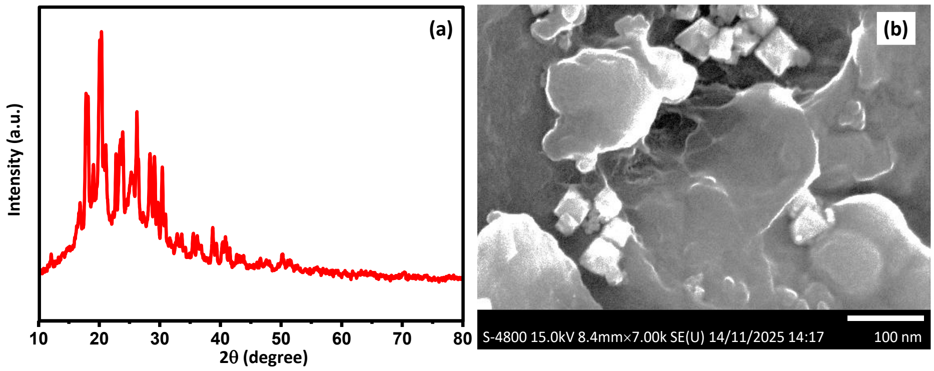

Figure 2: *(a) XRD and (b) SEM image of Zn–SBL1.*

Figure 3 shows the (a) Raman spectra and (b) UV–Vis spectra of Zn–SBL1. The Raman spectrum of the Zn–SBL1 complex (Figure 3a) shows distinct vibrational bands at 1174, 1254, 1348 and 1603 cm^{-1} and 2172 cm^{-1}, which collectively confirm the successful coordination of Zn^{2+} with the azomethine nitrogen and phenolic oxygen atoms of the Schiff base ligand. These bands correspond to C–O stretching, C–N bond vibrations, and C=N stretching modes, respectively. The peak observed at 2172 cm^{-1} suggests symmetric coordination and increased rigidity in the metal–ligand framework [7].

The UV–Vis spectrum of Zn–SBL1 (Figure 3b) exhibits a characteristic dip at 229 nm, followed by a pronounced absorption peak around 255 nm, which is attributed to $\pi \rightarrow \pi^*$ transitions within the aromatic system. A gradual increase in absorbance extending up to 600 nm indicates $n \rightarrow \pi^*$ transitions and ligand-to-metal charge transfer (LMCT) processes, typical of Schiff base Zn(II) complexes. The presence of LMCT bands signifies efficient electronic communication between the ligand orbitals and the Zn(II) center, supporting the observed enhancement in photophysical properties [8].

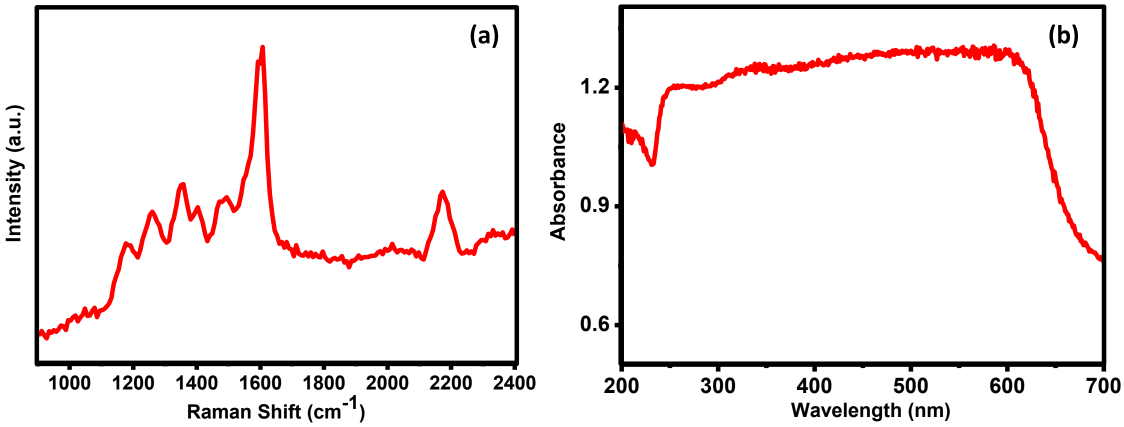

Figure 3: *(a) Raman spectra and (b) UV-Vis spectra of Zn–SBL1.*

Figure 4 shows the PL spectra of Zn–SBL1. The PL spectrum of the Zn–SBL1 complex recorded at an excitation wavelength of 225 nm reveals two significant emission peaks centered at 542 nm and 604 nm. The strong emission band at 542 nm corresponds to a green luminescent transition, which can be attributed to ligand-centered $\pi \rightarrow \pi^*$ relaxation processes enhanced through coordination with the Zn^{2+} ion. The second emission peak observed at 604 nm falls within the orange-red region and arises from $n \rightarrow \pi^*$ transitions and possible ligand-to-metal charge transfer relaxation pathways [9]. This suggests that Zn–SBL1 possesses strong fluorescence capability and may be applicable in photoactive materials, sensing or optoelectronic devices.

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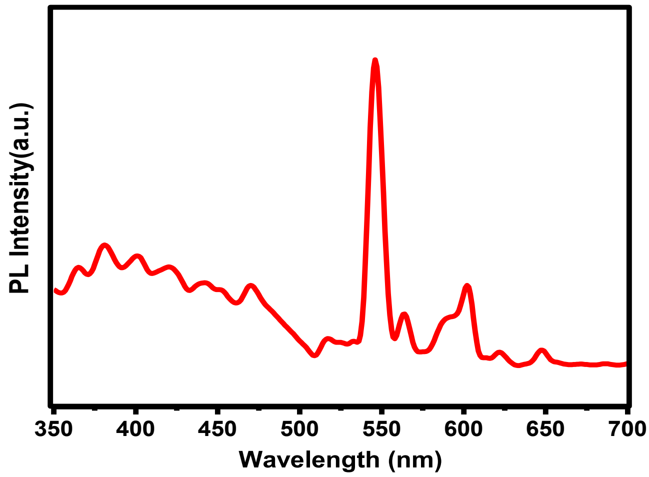

Figure 4: *PL spectra of Zn–SBL1.*

Figure 5 (a) shows the antibacterial activity of Zn–SBL1 towards the Bacillus subtilis for different doses (D1: 20 □g, D2: 40 □g, D3: 60 □g, D4: 80 □g, D5: 100 □g) and Figure 4(b) shows the antifungal activity of Zn–SBL1 towards the Aspergillus niger for different doses (D1: 20 □g, D2: 40 □g, D3: 60 □g, D4: 80 □g, D5: 100 □g, D6:120 □g). The antibacterial activity of Zn–SBL1 against Bacillus subtilis was evaluated at five different doses ranging from 20 µg to 100 µg (D1–D5) (Figure 5a, Table 1). The observed zones of inhibition gradually increased from 6 mm at 20 µg to 7 mm at 40–100 µg, indicating that the complex exhibits moderate antibacterial efficacy with a slight dose-dependent enhancement. This suggests that Zn coordination improves the ligand's ability to interact with the bacterial cell wall, possibly disrupting cell membrane integrity [10]. Similarly, the antifungal activity of Zn–SBL1 against Aspergillus niger was tested across six doses ranging from 20 µg to 120 µg (D1–D6) (Figure 5b, Table 1). The inhibition zones increased from 7 mm at 20–40 µg to 8 mm at 60–120 µg, demonstrating a mild but noticeable dose-dependent antifungal effect. The results indicate that the Zn–SBL1 complex is more effective against A. niger compared to B. subtilis, which may be attributed to enhanced interaction with fungal cell membranes and possible interference with intracellular processes [11].

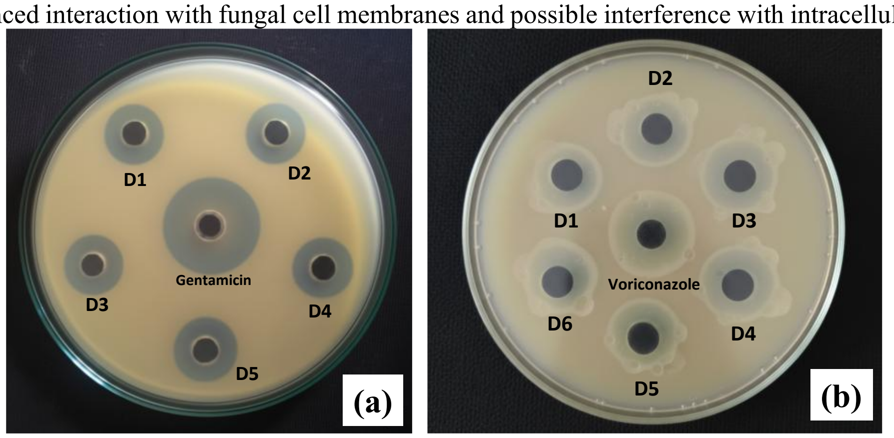

Figure 5: (a) ***Antibacterial activity of Zn–SBL1 towards the Bacillus subtilis for different doses (D1: 20 µg, D2: 40 µg, D3: 60 µg, D4: 80 µg, D5: 100 µg), (b) Antifungal activity of Zn–SBL1 towards the Aspergillus niger for different doses (D1: 20 µg, D2: 40 µg, D3: 60 µg, D4: 80 µg, D5: 100 µg, D6:120 µg).***

Table 1: Antimicrobial activity of Zn-SBL1.

Sample	**Zone of inhibition (mm)**				
	20 µg	**40 µg**	**60 µg**	**80 µg**	**100 µg**
	D1	**D2**	**D3**	**D4**	**D5**
Bacillus subtilis					
Zn–SBL1	**6**	**7**	**7**	**7**	**7**
Aspergillus niger					
Zn–SBL1	**7**	**7**	**8**	**8**	**8**

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IV. CONCLUSION

In conclusion, Zn(II) complex of N,N′ -Bis(salicylidene)ethylenediamine was successfully synthesized and their structural, optical and biological properties were systematically investigated. XRD and SEM studies revealed improved crystallinity and unique petal-like morphology, while Raman and UV–Vis analyses confirmed stable coordination through azomethine nitrogen and phenolic oxygen atoms, leading to significant LMCT and enhanced photophysical responses. Photoluminescence studies showed dual emission peaks at 542 nm and 604 nm, indicating multiple radiative relaxation pathways. Antimicrobial assays demonstrated that Zn–SBL1 exhibits moderate antibacterial and antifungal activities, with dose-dependent enhancement, confirming the role of metal coordination in improving bioactivity. These findings suggest that the Zn–SBL1 complex possesses multifunctional potential, making it suitable for applications in optoelectronics, sensing and antimicrobial materials.

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