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Original Article

Synthesis, Characterization and Antibacterial Activities of Indole-derived Ligand and its Ni (II), Co (II) and Cr (II) Complexes

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ABSTRACT

Objective: Schiff bases derived from indole frameworks have gained significant attention in medicinal and coordination chemistry due to their versatile donor sites and biological relevance. Their metal complexes often exhibit enhanced pharmacological properties through chelation, which improves lipophilicity and cellular permeability. However, despite their potential, only a limited number of studies have explored the antimicrobial properties of indole-based Schiff base complexes, especially those incorporating Ni(II), Co(II), and Cr(II). Therefore, this research focuses on synthesizing, characterizing, and evaluating new complexes of these metals to help bridge this gap. **Methods**: In this study, a Schiff base ligand, (E)-1-(5-bromoindolin-3-yl)-N-(2-(piperidin-1-yl) ethyl) methanimine (L1), was synthesized from 5-bromoindole-3-carbaldehyde and 1-(2-aminoethyl)piperidine in an ethanolic medium. Its Cr (II), Co (II), and Ni (II) complexes were prepared by refluxing the ligand with the corresponding metal chlorides. The compounds were characterized using FTIR, UV–Vis spectroscopy, molar conductance, and magnetic susceptibility measurements. Their antimicrobial and antifungal activities were evaluated against *Staphylococcus aureus*, *Klebsiella pneumoniae*, *Aspergillus niger*, and *Candida albicans*. **Results**: Spectroscopic and magnetic data confirmed octahedral coordination involving the indole and piperidine. The metal complexes showed higher thermal stability and molar conductance values indicative of $[ML]Cl_2$ electrolytic structures. Antimicrobial assays revealed improved activity of the complexes compared to the free ligand, particularly the Cr (II) and Ni (II) complexes, which exhibited notable inhibitions against *S. aureus* and *A. niger*. **Conclusion**: The study demonstrates that complexation of indole-based Schiff bases with transition metals enhances their physicochemical and biological properties. These findings support the potential of such metal complexes as promising antimicrobial agents and a foundation for designing therapeutic compounds.

1. INTRODUCTION

Azomethine compounds are a prominent class of organic compounds formed by the condensation of primary amines with aldehydes or ketones, resulting in an azomethine (–C=NR) functional group. First reported by Hugo Schiff in 1864, these compounds have since evolved into essential tools in coordination chemistry due to their ease of synthesis, structural diversity, and versatile donor properties (Takeda *et al.*

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2023). The presence of the imine nitrogen, often supplemented by oxygen, sulfur, or halogen donors, allows azomethine compounds to form stable complexes with a wide variety of metals, giving rise to diverse structures and properties (Marinova *et al.* 2025). These unique characteristics underpin their roles in catalysis, materials development, and medicinal chemistry (Kargar *et al.* 2022).

The significance of azomethine compounds in coordination chemistry was cemented in 1889 when Combes reported the first known metal complex of this class, a copper complex formed with an azomethine compound derivative (Rajasekar *et al.* 2025). Throughout the 20th century, Schiff bases became central to coordination studies, aided by the pioneering work of Alfred Werner, whose theories on coordination numbers and geometries provided a structural basis for understanding azomethine metal complexes (Kilic *et al.* 2007). Today, azomethine ligands remain some of the most widely studied in inorganic chemistry, serving as building blocks for systems ranging from enzyme mimics to functional materials (Asatkar *et al.* 2020).

Structurally, azomethine compounds can adopt various coordination modes, from simple bidentate forms, like those derived from salicylaldehyde, to complex tetradentate frameworks such as salen and salophen ligands. These ligands stabilize metals across a spectrum of oxidation states and coordination geometries, often forming square-planar or octahedral complexes (Regueiro *et al.* 2024). Recent studies have highlighted the benefits of halogen and indole substitutions, which can modulate electronic properties and enhance biological activity (Tuna *et al.* 2025). Such modifications not only affect redox behavior but also shift UV-Vis and FTIR spectral features, providing insights into their structure–function relationships (Malav 2024).

Although several indole-based Schiff base complexes have been reported, many of them lack comprehensive structural characterization, making it difficult to establish clear structure-activity relationships. In addition, previous studies have mostly focused on antibacterial properties alone, leaving antifungal screening largely unexplored. The influence of different transition metals such as Ni (II), Co (II), and Cr (II) on the electronic structure and biological performance of these complexes also remains insufficiently understood. These limitations highlight the need for more systematic research to develop well-characterized antimicrobial agents based on indole metal complexes.

This study contributes to the growing body of research on ligands and their metal complexes, with a particular focus on indole derivatives. Indole-based compounds are known for their wide range of biological activities. By synthesizing, characterizing, and evaluating the biological activities of Ni (II), Co (II), and Cr (II) complexes of indole Schiff base ligands, this research provides valuable insights into the structure–activity relationships of these compounds. The findings will not only advance fundamental understanding of coordination chemistry but may also serve as a foundation for the design of therapeutic agents with improved biological efficacy.

2. METHODS

All chemicals and solvents used in this study were of analytical grade and procured from reputable suppliers. The chemicals were used as purchased without further purification or modification. All glassware used was washed with detergent, rinsed with distilled water, and dried in an oven before use. The equipment includes: Jenway conductivity meter (Model 4010), UV-Visible spectrophotometer (Model T60), Stuart scientific melting point apparatus (SMP1), and FTIR spectrophotometer (Model Cary630, Agilent Technology). The bacterial isolates are: *Klebsiella pneumoniae and Staphylococcus aureus,* and the fungal isolates are: *Aspergillus niger* and *Candida albicans.* All the isolates were obtained and identified at the Microbiology department, Umaru Musa Yar'adua University, Katsina.

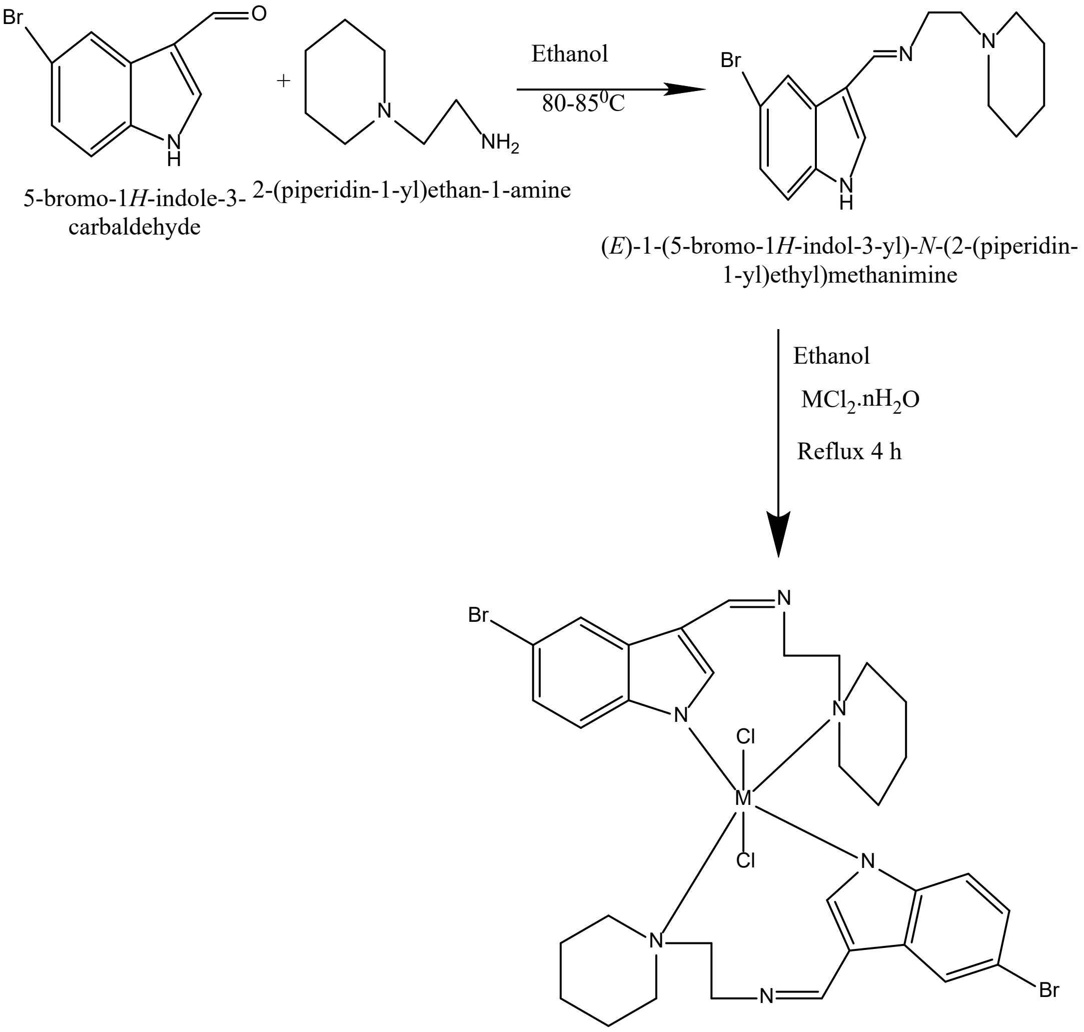
2.1 Preparation of Ligand L1

5-bromo-indole-3-cabaldehyde (0.01 mol, 2.24 g) and 1-(2-aminoethyl) piperidine (0.01 mol, 1.28 g) were combined in a hot ethanolic solution (20 mL) while being continuously stirred. For 6 hours, the mixture was refluxed at 80-85°C with a few drops of strong HCl added. The resulting solution was kept at room temperature overnight (Figure 1). The resulting precipitate was filtered, washed repeatedly with cold ethanol to remove unreacted starting materials, and dried under vacuum (Sulaiman *et al.* 2023).

2.2 Synthesis of Metal (II) Complexes

An ethanolic solution (20 mL) of the corresponding metal salts (0.01 mol) and a hot ethanolic solution (20 mL) of the ligand (6.68 g, 0.02 mol) were mixed together with constant stirring. The reaction mixture was refluxed for 4 hours at 80-85°C. The resulting solution was kept overnight at room temperature (Figure 1). The precipitate formed was filtered, washed repeatedly with cold ethanol to remove unreacted starting materials, and dried under

Figure 1: Reaction scheme for L1 and its metal (II) complexes



Where M= Ni^{2+}, Co^{2+} and Cr^{2+}

vacuum (Suleiman *et al.* 2023).

2.3 Antimicrobial and Antifungal Assay

Antimicrobial activity was tested against *Staphylococcus aureus* and *Klebsiella pneumoniae*, and antifungal activity against *Aspergillus niger* and *Candida albicans*. The agar well diffusion method was used with concentrations of 100, 50, 25, and 12.5 µg/mL. Ciprofloxacin and ketoconazole were used as reference drugs for antibacterial and antifungal screening, respectively.

3. RESULTS

3.1 Physical Properties and Conductivity

The ligand and its metal complexes were obtained in 65 – 80 % yield with decomposition points between 200 – 270 °C, indicating good thermal stability. Color changes upon complexation confirmed metal-ligand interaction. Molar conductance values ranged from 58-198 Ω^{-1} cm^2mol^{-1}, consistent with electrolytic $[ML]Cl_2$ structures (Tables 1 and 2).

3.2 FTIR and Electronic Spectra

The free ligand exhibited a C=N stretch at 1565 cm^{-1}, confirming azomethine formation. Coordination caused shifts in N-H and C-N vibrations and the appearance of new M-N bands at 387-506cm^{-1}, confirming metal-nitrogen bonding (Oruma *et al.*2024) (Table 3)

UV-Vis spectra revealed π-π* transitions near 250 nm and d-d transitions between 400-700 nm, characteristic of octahedral geometries for Ni (II), Co (II), and Cr (II) complexes (Table 4).

Table 1: Physical characteristics of the ligand and its metal (II) complexes.

Ligand/complexes	**Color**	**Yield %**	**Molecular weight**	**M.P/ D. T**	**Molar Conductance**	**μeff (B.M)**
$C_{16}H_{22}BrN_3$	Green	60.53	336.26	238	33	-
(CrL1) Cl_2	Light green	79.73	459.16	270	198	2.6
(CoL1) Cl_2	Pink	74.53	466.10	256	117	5.8
(NiL1) Cl_2	Green	65.79	465.86	260	58	2.8

KEY: M.P. = Melting point, D.T. = Decomposition Temperature, L1= $C_{16}H_{20}BrN_3$

Table 2: Solubility test of the ligand and its metal (II) complexes.

Compounds	**Ethyl Acetate**	**Acetic Acid**	**DMSO**	**Diethyl Ether**	**Chloroform**	**Distilled Water**	**Ethanol**	**Methanol**
$C_{16}H_{22}BrN_3$,	IS	IS	IS	SS	SS	S	S	SS
(CrL1) Cl_2	IS	IS	IS	SS	SS	S	S	S
(CoL1) Cl_2	IS	IS	IS	SS	SS	S	SS	S
(NiL1) Cl_2	IS	IS	IS	SS	SS	S	S	SS

KEY; L1= $C_{16}H_{22}BrN_3$ S= Soluble, SS=Slightly Soluble, and IS=Insoluble

Table 3: Infrared spectral data of the ligand (L1) and its complexes.

Ligand/complexes	**C=N**	**N-H**	**C-Br**	**C-N**	**C-H**	**M-N**
L1	1565	2631	670	1043/1192	879	-
(CrL1) Cl_2	1565	3220	626	1073/1267	842	387
(CoL1) Cl_2	1565	3242	-	1073/1259	842	387
(NiL1) Cl_2	1565	3250	-	1073/1259	849	506

Table 4: UV Vis for ligand and its metal (II) complexes.

Compounds	**Region (nm)**	**Transition Type**
L1	250–300	π→π*, n→π*
L1+Ni	260–300 400-450	π→π*, n→π* d–d transitions
L1+Cr	260–300 400-450	π→π*, n→π* d–d transitions
L1+Co	260–300 400-450	π→π*, n→π* d–d transitions

Table 5: Antimicrobial activity of Synthesized compounds against *Staphylococcus aureus*

Compounds ID	**100 μg/mL**	**50 μg/mL**	**25 μg/mL**	**12.5 μg/mL**
L1	11.0±1.0	18.0±0.0	14.5±0.5	8.0±0.0
L1+Cr	23±0.0	10.0±1.0	0.0±0.0	0.0±0.0
L1+Co	10.5±0.5	7.5±0.5	7.0±0.0	0.0±0.0
L1+Ni	17.0±0.0	11.0±0.0	8.5±0.5	0.0±0.0
Control (Ciprofloxacin)	22.0±1.0	16.5±0.5	12.5±0.5	9.5±0.5

Table 6: Antimicrobial activity of synthesized compounds against *Klebseilla pneumoniae*

Compounds ID	**100 μg/mL**	**50 μg/mL**	**25 μg/mL**	**12.5 μg/mL**
L1	10.0±1.0	8.5±0.5	7.0±0.0	0.0±0.0
L1+Cr	18.5±0.5	11.5±0.5	7.0±0.0	7.0±7.0
L1+Co	13.5±0.5	11.0±0.0	9.0±0.0	8.0±0.0
L1+Ni	14.0±0.0	9.0±1.0	7.5±0.5	7.0±0.0
Control (Ciprofloxacin)	26.0±0.0	21.0±1.0	17.5±0.5	15.0±0.0

Table 7: Antimicrobial Activity of Synthesized compounds against *Aspergillus niger*.

Compounds ID	**100 μg/mL**	**50 μg/mL**	**25 μg/mL**	**12.5 μg/mL**
L1	11.5±0.5	0.0±0.0	0.0±0.0	0.0±0.0
L1+Cr	16.0±0.0	11.5±0.0	9.0±0.0	7.5±0.0
L1+Co	11.0±0.0	9.0±0.0	7.5±0.5	0.0±0.0
L1+Ni	12.5±0.5	10.5±0.5	8.0±0.0	6.5±0.5
Control (Ketoconazole)	21.0±1.0	16.5±0.5	12.0±0.0	9.5±0.5

Table 8: Antimicrobial activity of synthesized compounds against *Candida albicans*.

Compounds ID	**100 μg/mL**	**50 μg/mL**	**25 μg/mL**	**12.5 μg/mL**
L1	9.5±0.5	7.0±0.0	0.0±0.0	0.0±0.0
L1+Cr	14.0±0.0	11.5±0.0	8.0±0.0	0.0±0.0
L1+Co	9.0±0.0	6.5±0.5	0.0±0.0	0.0±0.0
L1+Ni	15.0±1.0	11.0±0.0	9.0±0.0	7.5±0.5
Control (Ketoconazole)	22.0±1.0	16.5±0.5	12.5±0.5	9.5±0.5

3.3 Magnetic Susceptibility

Magnetic moment values (2.8-4.3 B.M.) suggested high-spin octahedral configurations: Ni (II) (2.9 B.M.), Co (II) (3.5 B.M.), and Cr (II) (4.3 B.M.). These data support paramagnetic, high-spin behavior typical of these ions in octahedral fields. (Suemitsu *et al.* 2024)

3.4 Antimicrobial and Antifungal Activities

The ligand displayed moderate inhibition against *S. aureus* (11.0 ± 1.0 mm), while metal complexes showed enhanced activity. The Cr (II) complex demonstrated strong inhibition against *K. pneumoniae* (18.5 ± 0.5 mm), and Ni (II) complex exhibited high antifungal potency against *C. albicans* (15.0 ± 1.0 mm). Overall, complexation improved antimicrobial efficacy compared to the free ligand (Table 5-8).

4. DISCUSSION

The ligand L1 appeared as a green crystalline solid with a sharp melting point at 238 °C, indicating purity (Bandgar *et al.* 2010). Its metal complexes with Cr (II), Co (II), and Ni (II) exhibited distinct colors—such as light green, pink, and dark brown-reflecting changes in ligand field due to coordination. The decomposition temperatures of the complexes (200-270 °C) were significantly higher than those of the ligand, suggesting enhanced thermal stability due to chelation, which increases structural rigidity (Shakir *et al.* 2009). The good yields (65-80%) further confirm the efficiency of the synthesis. These changes in physical appearance and stability upon metal coordination are consistent with the behavior of ligands known to form thermally stable, colored complexes with transition metals (Chandra *et al.* 2009).

The molar conductance measurements of the synthesized metal complexes were performed in distilled water due to their good aqueous solubility. The values obtained were in the range typically associated with 1:2 electrolytes, suggesting the presence of two dissociable chloride ions in solution. This observation indicates that the chloride ions are present as outer-sphere counterions rather than being coordinated to the metal center, confirming the formulation of the complexes as $[ML]Cl_2$, where L represents the bidentate Schiff base ligand. The high molar conductance values further support the ionic nature and potential dissociation of the complexes in aqueous medium, which is essential for their possible application in biological systems (Geary 1971).

The magnetic susceptibility measurements of the complexes compared well with their corresponding spin-only values. For Ni (II), the measured values of 2.9 B.M. agree with the spin-only value of 2.83 B.M., confirming two unpaired electrons in a high-spin octahedral geometry. For Co (II), the observed range of 3.5 B.M. matches closely with the spin-only value of 3.87 B.M., consistent with three unpaired electrons in a high-spin octahedral environment, though with less orbital contribution than commonly reported for Co (II). For Cr (II), the measured values of 4.3 B.M. approach the spin-only value of 4.90 B.M., indicating four unpaired electrons and supporting a high-spin octahedral geometry. These results confirm that all three metal complexes predominantly adopt high-spin octahedral structures, with slight variations in orbital contribution depending on the ligand environment (Suemitsu *et al.* 2024).

Solubility testing revealed that all ligands and their metal complexes were soluble in polar protic solvents such as distilled water, ethanol, and methanol, but insoluble in DMSO and other less polar or aprotic solvents such as chloroform, diethyl ether, ethyl acetate, and acetic acid. The water solubility of the compounds, particularly the metal complexes, is likely due to their ionic nature and the presence of hydrophilic functional groups in the ligand framework. This behavior is consistent with the properties of metal complexes bearing outer-sphere chloride ions, which enhance solubility in aqueous environments (Chandra *et al.* 2010). The aqueous solubility of the compounds is advantageous for biological evaluations, especially antimicrobial and antifungal studies, as it ensures better diffusion and bioavailability in physiological media.

The UV-Vis spectrum of the ligand L1 showed a maximum absorbance of 3.95 at 250 nm, attributed to π-π^* transitions within the aromatic and azomethine moieties. Upon coordination with Ni (II), Cr (II), and Co (II), slight variations in absorbance were observed, particularly in the 250–400 nm range, indicating metal–ligand interactions. Additional weak bands in the visible region (400–700 nm) were consistent with d–d transitions and ligand-to-metal charge transfer (LMCT), confirming complex formation. Based on the spectral features and the nature of the transition metals, the Ni (II) and Co (II) complexes are likely to adopt an octahedral geometry, while the Cr (II) complex also favors an octahedral arrangement due to its d^3 configuration and observed electronic transitions. These findings are consistent with those of Akash *et al.* (2019), who reported similar spectral behavior and geometrical preferences for Schiff base-metal complexes.

The FTIR spectral analysis of the ligand reveals important features that confirm its structure and coordination mode upon complexation with Cr(II), Co(II), and Ni(II) ions. The presence of a strong absorption band at 1565 cm⁻¹ in the free ligand and the metal complexes corresponds to the azomethine (C=N) stretching vibration, indicating the formation of the Schiff base and suggesting that the azomethine nitrogen does not participate in coordination (Adhao & Wagh 2024). A broad N-H stretch is observed at 2631 cm⁻¹ in the free ligand, which shifts to a higher range (3220-3250 cm^{-1}) in the metal complexes, implying involvement of the indolic nitrogen in coordination through deprotonation (Oruma *et al.* 2024). Additionally, the disappearance of the C–Br band at 670 cm⁻¹ in the metal complexes and its shift to 626 cm⁻¹ in the Cr complex supports complexation-induced electronic changes. Bands attributed to the C-N stretching vibration shift from 1043/1192 cm⁻¹ in the free ligand to 1073/1259-1267 cm^{-1} in the complexes, which further indicates coordination through nitrogen atoms (Mana *et al.* 2025). Most conclusively, new bands appearing in the far-infrared region at 387 cm^{-1} for Cr (II) and Co (II) complexes, and 506 cm^{-1} for the Ni (II) complex, are assigned to M-N stretching vibrations, confirming that both the indole nitrogen and the piperidine nitrogen are involved in bidentate coordination with the metal ions (Sunjuk *et al.* 2023; Juyal *et al.* 2024).

The antimicrobial screening of the synthesized ligand and its metal complexes against *Klebsiella pneumoniae* and *Staphylococcus aureus* revealed enhanced activity upon metal coordination. For *K. pneumoniae*, metal complexes such as L1+Cr (18.5 ± 0.5 mm) and L3+Ni (14.5 ± 0.5 mm) showed increased inhibition compared to their parent ligand and maintained activity at lower concentrations, indicating improved bacterial membrane permeability through chelation. While ciprofloxacin exhibited superior potency (26.0 ± 0.0 mm), some complexes showed moderate efficacy, suggesting their potential as alternative agents. Against *S. aureus*, ligand L1+Cr alone exhibited strong inhibition (23.0 ± 1.0 mm), comparable to fluconazole (22.0 ± 1.0 mm). These observations agree with Hamad *et al.* (2024), who reported that metal complexation improves membrane interaction and bioavailability of antimicrobial agents.

The antifungal evaluation against *Aspergillus niger* and *Candida albicans* further demonstrated that metal complexation significantly enhances the ligands' activity. For *A. niger*, L1 showed little to no activity on its own, but upon coordination, especially with Cr (II), inhibition improved markedly, as seen in L1+Cr (16.0 ± 0.0 mm). These complexes maintained activity across multiple concentrations, unlike the ligand alone. Similarly, against *C. albicans*, L1+Ni (15.0 ± 1.0 mm) and L1+Cr (14.0 ± 0.0 mm) demonstrated improved efficacy over their parent ligand, with activity sustained at lower doses. Although ketoconazole remained the most potent (22.0 ± 1.0 mm at 100 μg/mL), the enhanced activity of Ni (II) and Cr (II) complexes suggests promising antifungal potential. This is consistent with findings by Hamad *et al.* (2024), who emphasized that chelation increases lipophilicity and cellular uptake, thereby improving the antifungal performance of bioactive metal complexes against resistant fungal strains.

5. CONCLUSION

A new indole-derived Schiff base and its Cr (II), Co (II), and Ni (II) complexes were successfully synthesized and characterized. Spectral and magnetic data confirmed bidentate coordination through imine and indolic nitrogen atoms, leading to octahedral geometries. The complexes showed improved antimicrobial and antifungal activity compared to the free ligand, especially those containing chromium and nickel. These results demonstrate that metal complexation significantly enhances the biological potential of indole-based Schiff bases, making them promising candidates for future drug development studies.

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Conflict of interest

The authors declare that there is no conflict of interest regarding the publication of this manuscript.

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