International Journal of Applied Review
www.multiappliedjournal.com
E-ISSN: 3107-8443
Received: 24-04-2026, Accepted: 25-05-2026, Published: 09-06-2026
Volume 2, Issue 2, 2026, Page No. 56-64

Synthesis, spectroscopic characterization, and antibacterial evaluation of Sulfathiazole-derived schiff base complexes of Co(II) and Ni(II)

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DOI: https://doi.org/10.66856/ijar.2026.2.2.2099

Abstract

This study reports the synthesis, spectroscopic characterization, and antibacterial activity of two novel Schiff base ligands and their cobalt (II) and nickel (II) complexes. The ligands, namely L1 = 4-(2-aminobenzylideneamino)-N-(thiazol-2-yl) benzenesulfonamide and L2 = 4-(2-nitrobenzylideneamino)-N-(thiazol-2-yl) benzenesulfonamide, were prepared via condensation reaction of sulfathiazole with two appropriate aromatic aldehydes (2-aminobenzaldehyde and 2-nitrobenzaldehyde). The corresponding metal complexes of the general formula $[M(L)_2Cl_2]$ (where M = Co (II) or Ni (II)) were subsequently synthesized. All synthesized compounds (ligands and complexes) were characterized using a combination of physicochemical methods including molar conductivity measurements, magnetic susceptibility, elemental analysis (CHNS), and spectroscopic techniques such as electronic (UV-Vis) spectroscopy, infrared (FTIR) spectroscopy, atomic absorption spectroscopy, and proton nuclear magnetic resonance (^1H NMR). The obtained results confirmed the proposed octahedral and tetrahedral (square planar or tetrahedral) geometries for the prepared Schiff base complexes. The *in vitro* biological activity of the free ligands and selected complexes was assessment versus two clinically confirmed pathogenic bacterial strains: Gram-negative Escherichia coli and Gram-positive Staphylococcus aureus. The outcomes revealed that the metal complexes exhibit significantly higher inhibitory activity compared to their parent free ligands, with complexes designated as (C1, C2, N1, N2) showing the most potent antibacterial effects.

Keywords: Schiff bases, Cobalt(II) complexes, Nickel(II) complexes, antibacterial activity, Escherichia coli, octahedral geometry

Introduction

In the context of the recent work, Schiff base ligands have been the focus of great interest due to their structural variety and a wide range of physicochemical properties based on which a vast plethora of applications can be envisaged [1–3]. The flexibility in the structure of Schiff bases allows them to act as effective asymmetric inducers and stabilizers for metal complexes in multiple oxidation states [4–6]. In addition, these compounds are essential in regulation of catalytic activity of metals in a large number of synthetically useful transformations. Tetradentate Schiff base complexes are well known to form very stable chelates with dinitrogen and dioxygen donor atoms, so their coordination behavior is reminiscent of that of naturally occurring metalloproteins. Some of them, the azo-azomethine, in particular, have enjoyed wide technological application based on their photophysical and energy-transfer properties. Due to their stability, electrochemical activity, thermochromism and photochromism, Schiff bases are good candidates as precursors for different applications in both industrial and research fields [7–10]. Several reviews report that the organometallic chemistry of Schiff base complexes has a significant impact on their bioactivity [11–18], which highlights the importance of metal centers in biological systems as catalyst in various bioprocesses. The planar nature of the C=N group leads to the adsorption of bonding orbitals and electrons of the highly electronegative heteroatoms on the surface of metal. Thus, this class of compounds has played a vital role in the development of inorganic chemistry and provides a number of biologically active molecules as they serve as flexible substrates in ring-closure, cycloaddition, substitution etc. reactions [19–21]. In terms of chemistry, Schiff bases are condensation products of primary amines and carbonyl compounds (aldehydes or ketones) and the general structure of these molecules is shown in Fig. 1 [22, 23]. Originally described in the reports of the German chemist Hugo Schiff in 1864, they are also described as azomethines, imines or anils [24]. The representative structural feature of these compounds is the azomethine group ($R^1R^2C=NR^3$), where the substituents can be alkyl, cycloalkyl, aryl, or heterocyclic moieties [23,25].

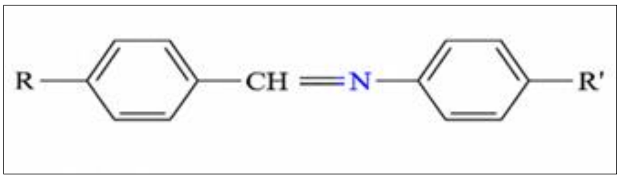


Fig 1: General aromatic Schiff base structure

Importance of Schiff base metal complexes has been well established in the areas of bioinorganic chemistry, supramolecular chemistry, biomedical science and materials science [26]. These complexes give substances of interest to both natural and synthetic oxygen carriers, and active stereospecific catalysts for reduction, oxidation, hydrolysis and modification reactions in inorganic and organic chemistry [28]. Metal–ligand coordination is usually carried out by heteroatoms such as N, S, or O, which form stable coordination complexes. Metals and their complexes have been of considerable medical importance for a time span longer than 5000 years [29]. Additionally, the roles of Schiff base metal complexes as functional pigments, dyes, and agents in polymerization and the pharmaceutical industry merit particular mention [30].

In the present study, two novel Schiff base ligands were prepared by condensation of sulfathiazole with 2-aminobenzaldehyde and 2-nitrobenzaldehyde to produce ligands L1 and L2, respectively. Their cobalt(II) and nickel(II) complexes were subsequently prepared and characterized using elemental analysis, FT-IR spectroscopy, UV–Vis spectroscopy, molar conductivity measurements, magnetic susceptibility studies, and ^{1}H NMR spectroscopy. Furthermore, the antibacterial activity of the synthesized ligands and their metal complexes was investigated against Gram-positive Staphylococcus aureus and Gram-negative Escherichia coli.

Materials and Instruments

1. Materials

All chemicals and solvents used in this study were of analytical grade and were used without further purification. Sulfathiazole, 2-aminobenzaldehyde, 2-nitrobenzaldehyde, cobalt(II) chloride hexahydrate, nickel(II) chloride hexahydrate, methanol, ethanol, dimethylformamide (DMF), dimethyl sulfoxide (DMSO), chloroform, diethyl ether, and glacial acetic acid were obtained from Fluka, Aldrich, and B.D.H. companies.

2. Instrumental Methods

Melting points were determined using an Electrothermal 9300 apparatus; molar conductivity was measured with a digital conductivity meter (Coring 220) for 10^{-3} M solutions in DMF at 25 °C; FTIR spectra were recorded on Shimadzu FTIR-8400S and Perkin-Elmer spectrophotometers in the range 4000–200 cm^{-1} using KBr/CsI discs; ^{1}H NMR spectra were obtained on a Bruker UltraShield 300 MHz spectrometer in DMSO-d_6 at the University of Al-Bayt, Jordan; elemental analysis (CHNS) was performed using a Eurovector EA 3000 A instrument from Italy; UV-Vis spectra were gauged on a Shimadzu UV-1650 PC spectrophotometer in the range 200–1100 nm for 10^{-3} M solutions in DMF using 1 cm quartz cells; magnetic susceptibility was determined by the Faraday method using a Bruker BM6 at 25 °C; metal content was analyzed using a PYE UNICAM SP9 (Phillips) atomic absorption spectrophotometer.

3. Synthesis of Schiff Base Ligand

3.1 Synthesis of Ligand L1

Sulfathiazole (1.53 g, 6 mmol) was dissolved in 15 mL methanol, then 2-aminobenzaldehyde (0.72 g, 6 mmol) in 5 mL methanol was added dropwise with a few drops of CH_3COOH acid. The mixture was stirred at room temp. for 30 min then refluxed for 3 h. A beige precipitate formed, filtered, washed with ethanol and ether, and dried. Yield: 85% (1.824 g). M.p. 173–175 °C.

3.2 Synthesis of Ligand L2

Sulfathiazole (1.53 g, 6 mmol) in 15 mL methanol was treated with 2-nitrobenzaldehyde (0.9 g, 6 mmol) in 5 mL methanol. Stirred 30 min at room temperature, then refluxed 3 h. The clear yellow solution was concentrated to half volume and cooled. A yellow gum formed, extracted with cold ether, washed with chloroform, and dried. Yield: 92% (2.14 g). M.p. 100–103 °C.

4. Synthesis of Metal Complexes

The cobalt(II) and nickel(II) complexes were prepared by adding a solution of cobalt(II) chloride hexahydrate or nickel(II) chloride hexahydrate (0.237 g, 0.001 mol) in 5 mL of absolute ethanol to a suspension of the appropriate Schiff base ligand, L1 (0.358 g, 0.001 mol) or L2 (0.388 g, 0.001 mol), in 10 mL of absolute ethanol. The reaction mixture was stirred for 3 h at room temperature until a colored precipitate was formed. The resulting solid complex was filtered under reduced pressure, washed with absolute ethanol and chloroform, and dried. The isolated weights and percentage yields of the prepared complexes are listed in Table 1.

Table 1: Weights and percentage yields of the resulting complexes

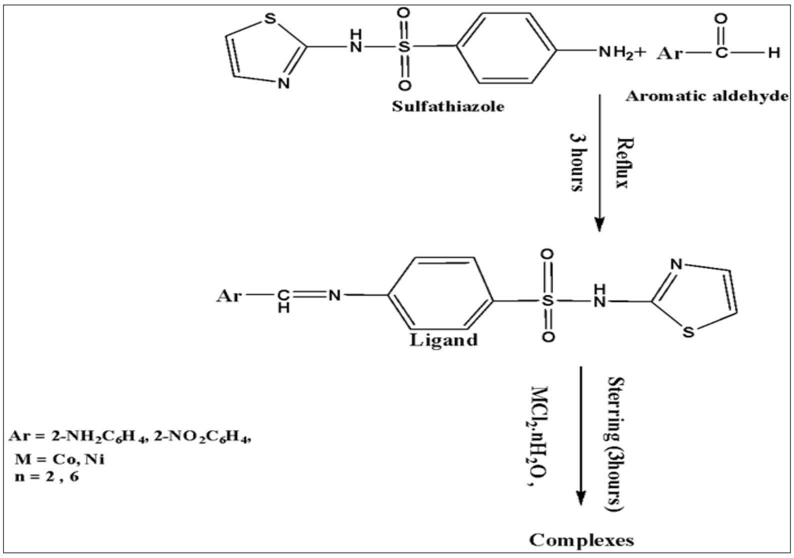
Code	Metal complex	Weights (g)	Yield%
C1	[Co(2ABTBS)$_2$Cl2]	0.228	27
C2	[Co(2NBTBS)$_2$Cl2]	0.2	22
N1	[Ni (2ABTBS)$_2$Cl2]	0.27	32
N2	[Ni (2NBTBS)$_2$Cl2]	0.21	23

5. Biological Evaluation

Two types of pathogenic bacteria were employed: The Gram-positive Staphylococcus aureus and the Gram-negative Escherichia coli. These bacterial isolates were collected from the laboratories of Al-Batool Teaching Hospital in Ninveh Governorate, and identification was confirmed by using Masta staph strips and Api system. The prepared ligands (L1and L2) and their cobalt(II) and nickel(II) complexes were tested for their biological activity towards the aforementioned bacterial strains. Preparation of nutrient agar (solid medium): 38g nutrient agar powder was dissolved in 1000cm^3 of distilled water, sterilized in an autoclave at 121 °C and 15 atm for 15 min allowed cooling at 40–45°C, poured into petri-dishes and left at room temperature to solidify as a semisolid gelatinous layer. In sterile conditions, solutions of each ligand and complex were prepared in DMSO as the solvent at concentrations of 5, 25, 50 and 100 mg/ml and then were sterilized by pasteurization at 63°C for 15 min. The disk diffusion sorptions method was carried out with Whatman N. 1 filter paper discs (6 mm diameter) sterilized by autoclave, dried and then loaded with 4 µL of the solution of the prepared compounds. The 0.1 mL of bacterial suspension was spread on nutrient agar plates with the help of sterile cotton swab, the discs were put on the agar and the plates were incubated at 37°C for 24 h. Two antibiotics, Garamycin and Imipenem, were used as positive controls, while DMSO served as the solvent control. The inhibition zones (diameters) were measured with a millimeter ruler.

Results and Discussion

In this study, two Schiff base ligands, L1 and L2, were synthesized by condensation of sulfathiazole with 2-aminobenzaldehyde and 2-nitrobenzaldehyde, respectively, resulting in the formation of azomethine (C=N) groups. The corresponding cobalt(II) and nickel(II) complexes were subsequently prepared as illustrated in Scheme 1. The purity and composition of the prepared ligands were confirmed by melting point measurements and elemental microanalysis (CHNS).



Scheme 1: General preparation method for the ligands and complexes

1. Characterization of the Ligands and Their Metal Complexes

1.1 Elemental Analysis and Physical Properties of the Ligands

The prepared ligands were characterized by elemental microanalysis of carbon, hydrogen, nitrogen, and sulfur. The experimental values were in good agreement with the calculated values, confirming the proposed molecular formulae of the ligands. The physical properties and elemental analysis data of L1 and L2 are presented in Table 2.

Table 2: Physical properties and elemental analysis data of ligands L1 and L2

Compound	Formula	Color	M.P. (°C)	Yield (%)	C % Found (Calc.)	H % Found (Calc.)	N % Found (Calc.)	S % Found (Calc.)
2ABTBS (L1)	$C_{16}H_{14}N_4O_2S_2$	Honey	173–175	85–87	53.26 (53.61)	3.63 (3.94)	15.71 (15.63)	17.37 (17.89)
2NBTBS (L2)	$C_{16}H_{12}N_4O_4S_2$	Pale yellow	100–103	92	49.50 (49.47)	3.55 (3.11)	13.80 (14.42)	16.15 (16.51)

The agreement between the experimental and theoretical percentages confirms the successful preparation and acceptable purity of the Schiff base ligands. The relatively sharp melting point ranges also indicate good purity of the isolated compounds.

Table 3: Physical properties and metal percentages of the prepared Co(II) and Ni(II) complexes

Code	Formula	Color	M.P. (°C)	Yield (%)	Metal % Found (Calc.)
C1	[Co(2ABTBS) Cl]	Reddish brown	191–195	27	6.34 (6.95)
C2	[Co(2NBTBS) Cl]	Dark green	199–202	22	6.96 (6.49)
Code	Formula	Color	M.P. (°C)	Yield (%)	Metal % Found (Calc.)
N1	[Ni(2ABTBS) Cl]	Light beige	250 d*	32	6.12 (6.93)
N2	[Ni(2NBTBS) Cl]	Creamy	217 d	23	5.97 (6.48)

*d = decomposition temperature. $_{22}$ $_{22}$

1.2 Atomic Absorption and Physical Properties of the Metal Complexes

The metal contents of the prepared complexes were determined by atomic absorption spectroscopy. The obtained experimental values were close to the calculated values, supporting the proposed formulas of the complexes. The physical properties and metal percentages are shown in Table 3.

The differences in color between the ligands and their metal complexes indicate the formation of new coordination compounds. The agreement between found and calculated metal percentages supports the proposed stoichiometry of the complexes.

1.3 FTIR Spectra of Ligands L1 and L2

The FTIR spectra of ligands L1 and L2 showed several characteristic bands related to the functional groups present in the Schiff base structures. The FTIR spectra of ligands L1 and L2 exhibited characteristic absorption bands corresponding to the functional groups present in the Schiff base structures. Ligand L1 showed a medium-intensity absorption band at 3433 cm^{-1} attributed to the stretching vibration of the NH_2 group, whereas this band was absent in ligand L2 due to the presence of the nitro substituent instead of the amino group. The sulfonamide NH stretching vibrations appeared at 3226 cm^{-1} for L1 and 3134 cm^{-1} for L2. The azomethine (C=N) stretching bands were observed at 1623 cm^{-1} and 1628 cm^{-1} for L1 and L2, respectively, confirming successful Schiff base formation through condensation reactions between sulfathiazole and the corresponding aldehydes. Moreover, the C=N vibrations of the thiazole ring were observed at 1577 cm^{-1} for L1 and 1573 cm^{-1} for L2. The azomethine bands were shifted to lower frequencies upon complex formation with Co(II) and Ni(II) ions, suggesting the involvement of azomethine nitrogen atoms in coordination.

The appearance of new bands in the low frequency region at about 420–510 cm^{-1} is due to M–N stretching vibrations which is indicative that ligands are coordinated to metal ions via nitrogen donor atoms. These observations support that L1 and L2 act mainly as neutral nitrogen donor ligands, where coordination occurs principally through the azomethine nitrogen, with possible contribution from the thiazole nitrogen depending on the metal ion and complex geometry.

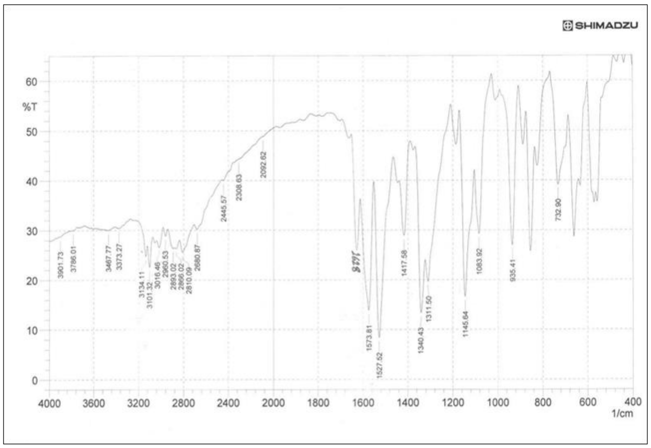


Fig 2: FTIR spectrum of ligand L1

1.4 ^{1}H NMR Spectral Study of the Prepared Ligands

a. ^{1}H NMR Spectrum of Ligand L1

The ^{1}H NMR spectrum of ligand 4NBTBS was recorded in deuterated dimethyl sulfoxide DMSO-d_6, as shown in Figure 3. The spectrum exhibited several overlapped signals in the region δH = 6.84–8.45 ppm, integrating for ten protons. Based on their chemical shift values and integration, these signals were assigned to the aromatic protons of the phenyl and thiazole-containing aromatic systems. A singlet signal was observed at δH = 8.80 ppm, integrating for one proton. This signal was attributed to the azomethine proton HC=N, confirming the formation of the Schiff base linkage between sulfathiazole and

4-nitrobenzaldehyde. In addition, the spectrum showed a relatively broad singlet at δH = 12.75 ppm, corresponding to one proton. This downfield signal was assigned to the sulfonamide NH proton. The broad nature of this signal may be related to hydrogen bonding and the de shielding effect of the sulfonyl group.

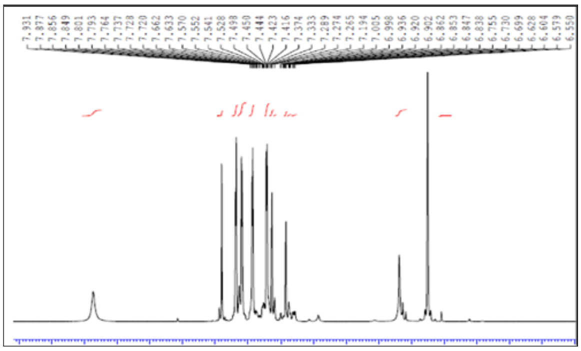


Fig 3: ^{1}H NMR spectrum of ligand L1 in DMSO-d_6

b. ^{1}H NMR Spectrum of Ligand L2 (2NBTBS)

The ^{1}H NMR spectrum of ligand L2 (2NBTBS) was recorded in deuterated dimethyl sulfoxide DMSO-d_6, as shown in Figure 4. The spectrum exhibited a group of multiplet signals in the region δH = 5.85–8.31 ppm, corresponding to ten aromatic protons. This assignment was supported by the integration values and the chemical shift range expected for aromatic protons. The appearance of the aromatic signals as overlapped multiplets may be attributed to the close chemical environments of the aromatic protons and, to some extent, to the limited purity of the isolated compound. In addition, the spectrum showed a distinct singlet signal at δH = 8.88 ppm, integrating for one proton. This signal was assigned to the azomethine proton HC=N, confirming the formation of the Schiff base linkage through condensation between sulfathiazole and 2-nitrobenzaldehyde. Furthermore, a relatively broad singlet appeared at δH = 12.60 ppm, integrating for one proton. This downfield signal was attributed to the sulfonamide NH proton. Its broad nature and high chemical shift are consistent with the presence of hydrogen bonding and the electron-withdrawing effect of the sulfonyl group.

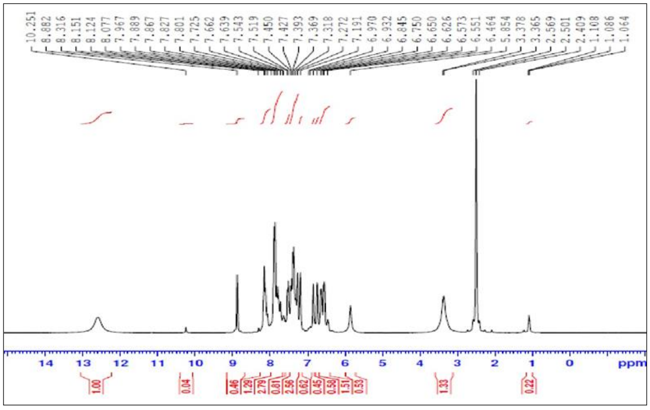


Fig 4: ^{1}H NMR spectrum of ligand L2 (2NBTBS) in DMSO-d_6

1.5 Molar Conductivity Measurements

Molar conductivity is a useful parameter in coordination chemistry to determine whether a metal complex behaves as an ion or nonelectrolyte in solution. Low values of molar conductance in polar organic solvents like DMF, DMSO are indicative of non-electrolytic nature of complexes, whereas higher values indicate ionic dissociation in solution [31, 32]. In this work the molar conductivities of the novel Co(II) and Ni(II) complexes were recorded in DMF at 25° C at 10-3 M concentration. The values obtained were very low suggesting that the complexes are nonelectrolytic. This behavior suggests that the chloride anions are closely associated with the metallic centers and are trapped in the coordination sphere rather than existing as free counter ions. Typical values of molar conductance for different types of electrolytes in few solvents are given in Table 4.

Table 4: Typical molar conductance values of different electrolyte types at 10^{-3} M in various solvents

Solvent	**1:1 electrolyte**	**2:1 electrolyte**	**3:1 electrolyte**	**4:1 electrolyte**	**Non-electrolyte**
Water	120	240	360	480	0
Ethanol	35–45	70–90	120	160	0–20
Nitromethane	75–95	150–180	220–260	290–330	0–20
Acetonitrile	120–160	220–300	340–420	500	0–30
Dimethylformamide (DMF)	65–90	130–170	220–240	300	0–30
Dimethyl sulfoxide (DMSO)	30–40	70–80	—	—	0–20

The molar conductance values of the prepared complexes were measured in DMF at a concentration of 10^{-3} M and at 25 °C after thermal equilibrium was reached. The obtained results are listed in Table 6**.** The measured values were very low for all prepared complexes, indicating that these compounds behave as non-electrolytes or very weakly conducting species in solution.

This behavior confirms that the complexes do not dissociate into free ions in DMF. Therefore, the Cl ions are most likely coordinated directly to the metal-centers and are located inside the coordination sphere rather than existing as free counter ions. These results are consistent with the suggested neutral formulas of the prepared complexes.

The conductivity data are also complementary to the results obtained from elemental analysis, electronic spectra, FT-IR spectra, atomic absorption measurements, and magnetic susceptibility. Together, these data support the suggested geometrical structures of the synthesized metal complexes. The results are shown in Table 5.

Table 5: Molar conductivity values of the prepared complexes at 10^{-3} M in DMF at 25 °C

Code	**Complex**	**ΛM (ohm^{-1} cm^2 mol^{-1})**
C1	[Co(2ABTBS) Cl]	0.172
C2	$[Co(2NBTBS)_2Cl_2]$	0.427
N1	$[Ni(2ABTBS)_2Cl_2]$	0.424
N2	$[Ni(2NBTBS)_2Cl_2]$	0.185

The molar conductivity values of the prepared Co(II) and Ni(II) complexes were measured in DMF at a concentration of 10^{-3} M and at 25 °C. The obtained values were very low, the observed behavior of the complexes implies that they function as non-electrolytes in an aqueous environment. This phenomenon indicates that the chloride ions are directly coordinated to the metallic centers and are situated within the coordination sphere, rather than being present as unbound counter ions. These results support the proposed neutral formulas of the prepared complexes.

1.6 Elemental Microanalysis

Elemental analysis is a necessary instrumental method for confirming the empirical formula and purity of novel Schiffbase ligands and their metal derivatives. The found values of C, H, N and S by the elemental analysis also agree with the molecular formula of the synthesized compounds calculated from the molecular weight [33, 34]. In the present work, the values of element analysis of ligands L1 and L2 were very close to the calculated values. For ligand L1, the found percentages of C, H, N, and S were close to those expected for $C_{16}H_{14}N_4O_2S_2$, while ligand L2 showed elemental composition consistent with $C_{16}H_{12}N_4O_4S_2$. These results confirm the successful formation and acceptable purity of the Schiff base ligands. The physical properties and elemental analysis data are provided in Table 6.

Table 6: Physical properties and elemental microanalysis data of the prepared ligands and selected complexes

Compound. No	**Formula**	**Color**	**M.P. °C**	**Yield %**	**C % Found (. (Calc**	**H % Found (. (Calc**	**N % Found (. (Calc**	**S % Found (. (Calc**
L1	$C_{16}H_{14}N_4O_2S_2$	Honey	175–173	87–85	53.26 (53.61)	(3.94) 3.63	15.71 (15.63)	17.37 (17.89)
L2	C_{16} H_{12} N_4 O^4 S_2	Pale yellow	103–100	92	49.50 (49.47)	(3.11) 3.55	13.80 (14.42)	16.15 (16.51)
Compound. No	Formula	Color	M.P. °C	Yield %	C % Found (. (Calc	H % Found (. (Calc	N % Found (. (Calc	S % Found (. (Calc
C1	Co(2ABTBS) Cl2] [2	Reddish brown	*d 232	27	35.61 (35.87)	(2.63) 2.42	10.47 (10.46)	11.42 (11.97)
N1	[Ni(2ABTBS) ₂Cl2]	Light beige	d 242	24	30.87 (31.25)	(2.62) 2.57	(9.11) 9.35	10.36 (10.43)
C2	Co(2NBTBS) Cl2] [2	Dark green	d 264	78	34.30 (33.97)	(2.14) 2.78	(9.90) 9.88	11.47 (11.34)
N2	[Ni(2NBTBS) Cl]	Light brown	d 313	44	29.43 (29.37)	(1.85) 1.29	(8.56) 8.47	(9.80) 9.30

The elemental analysis values of the free ligands showed good agreement with their calculated molecular formulae. For ligand L1, the found percentages of C, H, N, and S were close to the theoretical values expected for the formula $C_{16}H_{14}N_4O_2S_2$**.** Similarly, ligand L2 showed elemental composition consistent with the proposed formula $C_{16}H_{12}N_4O_4S_2$**.** These results confirm the successful formation of the ligands.

The small difference between the found and calculated values could be due to residual solvent, partial decomposition on drying, or some complexes being hygroscopic. In general, the results of the elemental microanalysis are in support of the suggested stoichiometry of the synthesized compounds.

1.7 Atomic Absorption Spectroscopy

The metal contents of the synthesized Co(II) and Ni(II) complexes were ascertained by atomic absorption spectroscopy. This technique is commonly used in coordination chemistry to verify the percentage of metal ions and to the suggest stoichiometric of metal complexes [35, 36]. The Co(II) and Ni(II) ions were successfully coordinated by the Schiff base ligands as indicated by the experimentally found values of metal which were very close to the calculated ones. These findings are in favor of the suggested formulas of the synthesized complexes and are consistent with the values obtained from FT-IR, electronic spectra, molar conductivity and magnetic measurements. The physical characteristics, the metal content are given in Table 7.

Table 7: Physical properties and metal percentages of the prepared metal complexes

Code	Formula	Color	% Yield	(. Metal % Found (Calc
C1	[Co(2ABTBS) Cl]	Reddish brown	27	(6.95) 6.34
C2	$[Co(2NBTBS)_2Cl_2]$	Dark green	22	(6.49) 6.96
N1	$[Ni(2ABTBS)_2Cl_2]$	Light beige	32	(6.93) 6.12
N2	$[Ni(2NBTBS)_2Cl_2]$	Light brown	23	(6.48) 5.97

The agreement between the experimental and calculated metal percentages confirms the successful coordination of Co(II) and Ni(II) ions with the Schiff base ligands. These results, together with FT-IR, electronic spectra, molar conductivity, and magnetic susceptibility data, support the proposed stoichiometries of the synthesized metal complexes.

1.8 Electronic Spectra of the Ligands

The free Schiff base ligands L1 and L2 were subjected to the electronic spectra in DMF. The spectra exhibited absorption bands, which can be mainly assigned to the intraligand electronic transitions (π→π* and n→π*). These transitions occur at the aromatic rings, azomethine group, thiazole ring and sulfonamide nucleus of the ligand moieties. L1 was similarly found to have absorption peaks at 26385 and 33222 cm^{-1}, which can be attributed to the n→π* and π→π* transitions, respectively. Moreover, a high-energy band is generated at 42194 cm-1 and is assigned to the ligand -based electronic transitions. Ligand L2 presented at 17953 and 31746 cm^{-1} a couple of absorption bands, along with a high energy one at 41666 $cm^{-}1$. The lower energy band for L2 may be due to the electron withdrawing effect of the nitro group, which affects the electron distribution in the ligand molecule. The electronic spectral data of the ligands are presented in Table 8.

Table 8: Electronic spectral data of ligands L1 and L2

Ligand	Band 1 cm^{-1}	Band 2 cm^{-1}	Charge transfer / high-energy band cm^{-1}
L1	26385	33222	42194
L2	17953	31746	41666

1.9 Electronic Spectra of the Metal Complexes

The UV-Vis spectra of the prepared Co(II) and Ni(II) complexes in DMF were measured at 25 °C utilizing 10^{-3} M concentrations. The complexes experience shifts in position and intensity of ligand-based bands in comparison to the free ligands, indication the coordination of the ligands to the metal ions. Furthermore, new bands also emerge in the visible region, associated with d–d transitions, and the bands of higher energy are considered as charge-transfer transitions. The C1 complex showed a band at 17513 cm^{-1} along with a charge-transfer band at 27027 cm^{-1}, confirming a square-planar geometry. The C2 complex showed a band at 16339 cm^{-1} and a charge transfer band at 28490cm^{-1} indicative of a tetrahedral geometry. Regarding one precious issue for the Ni(II) complexes, N1 revealed an absorption band at 15047 cm−1 and a charge transfer band at 32467 cm−1 assigned to a tetrahedral environment. N2, on the other hand, showed three bands at 9460, 17421 and 29746 cm^{-1} thought to be salary of an octahedral Ni(II) complex. These electronic spectral findings and magnetic susceptibility measurements justified the suggested geometries for the synthesized complexes. The results are given in Table 9.

Table 9: Electronic spectral data of complexes

Compound	Geometry	ν_1 / n→π* (cm^{-1})	ν_2 / π→π* (cm^{-1})	ν_3 (cm^{-1})	C.T. (cm^{-1})
C1	Sq. planar	—	—	17513	27027
C2	Tetrahedral	—	—	16339	28490
N1	Tetrahedral	—	—	15047	32467
N2	Octahedral	9460	17421	29746	31746

1.10 Magnetic Measurements

Magnetic measurements are widely used for characterizing transition metal complexes, as they provide essential information about electronic configuration, oxidation state, and spin state (high-spin or low-spin) based on the number of unpaired electrons. A complex is paramagnetic if the central atom contains unpaired electrons and diamagnetic if all electrons are paired. When paramagnetic centers are separated by diamagnetic ligands, they are magnetically diluted with no interaction between magnetic moments; otherwise, interactions may lead to ferromagnetism (parallel alignment) or antiferromagnetism (antiparallel alignment) (39.40). Magnetic susceptibilities of the prepared complexes were gauged at 25 °C, with diamagnetic corrections using Pascal's constants. The values of gram susceptibility (χg), molar susceptibility (χM), atomic susceptibility (χA), diamagnetic correction factor (D), and effective magnetic moment (μeff) for the prepared complexes are presented in Table 10.

Table 10: Magnetic measurement values for selected prepared complexes at 25 °C

Code	χg × 10^{-6}	χM × 10^{-6}	D × 10^{-6}	χA × 10^{-6}	μeff (B.M.)	Suggested geometry
C1	2.786	2362	329.3	2691	2.53	Square planar
C2	6.892	6249	371.44	6619	3.97	Tetrahedral
N1	9.085	7687	329.3	8016	4.37	Tetrahedral
N2	3.354	3040	371.44	3411	2.85	Octahedral

Magnetic susceptibility metrics were used to support the proposed geometries of the prepared Co(II) and Ni(II) complexes. The Co(II) complex C1 showed an effective magnetic moment of 2.53 B.M., implies a square-planar environment, whereas C2 showed a magnetic moment of 3.97 B.M., which is more coherent with a tetrahedral arrangement. For the Ni(II) complexes, N1 exhibited a magnetic moment of 4.37 B.M., supporting a tetrahedral geometry with orbital contribution, while N2 showed a value of 2.85 B.M., consistent with an octahedral environment. These results are in agreement with the electronic spectral data and support the proposed structures of the prepared complexes.

1.11 Antibacterial Activity Measurements

The antibacterial activity results showed that the free Schiff base ligands L1 and L2 exhibited weak to moderate activity against the tested bacterial strains. Ligand L1 showed inhibition only at the highest concentration, with inhibition zones of 5 mm against E. coli and 10 mm against Staphylococcus aureus. Ligand L2 displayed slightly higher activity, especially against Staphylococcus aureus, with inhibition zones of 10 and 15 mm at 50 and 100 mg/mL, respectively. The Co(II) and Ni(II) complexes generally showed enhanced antibacterial activity compared with the corresponding free ligands. This improvement may be attributed to chelation, which increases the lipophilic character of the complexes and facilitates their penetration through the bacterial cell membrane. Among the prepared complexes, C1 and N2 showed the highest inhibition against E. coli at 100 mg/mL, while C1 exhibited the highest activity against Staphylococcus aureus. However, the activity of all synthesized compounds remained lower than that of the standard antibiotic imipenem. The outcomes are presented in Table 11.

Table 11: Antibacterial activity of ligands and complexes

. Compound No	**(Conc. (mg/mL**	***E. coli***	***Staph. aureus***
L1	5	---	---
	25	---	---
	50	---	---
	100	5	10
L2	5	---	---
	25	---	---
	50	---	10
	100	7	15
C1	5	---	7
	25	---	13
	50	14	17
	100	18	21
C2	5	---	---
	25	---	---
	50	11	11
	100	15	14
N1	5	---	---
	25	---	---
	50	10	11
	100	15	16
N2	5	---	---
	25	12	8
	50	15	10
	100	18	14
Garamycin	10	15	21
.Compound No	(Conc. (mg/mL	*E. coli*	*Staph. aureus*
Imipenem	10	25	36
DMSO	---	---	---

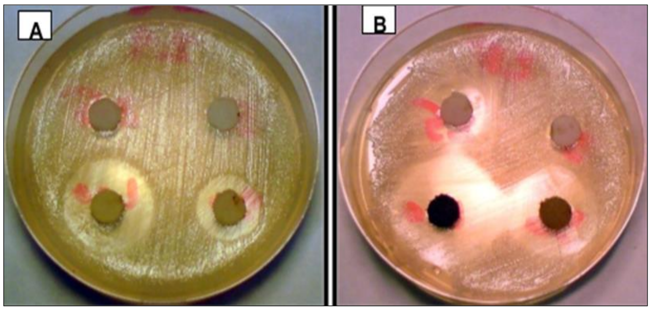


Fig 5: Antibacterial activity of complexes: A) N1 and B) C1

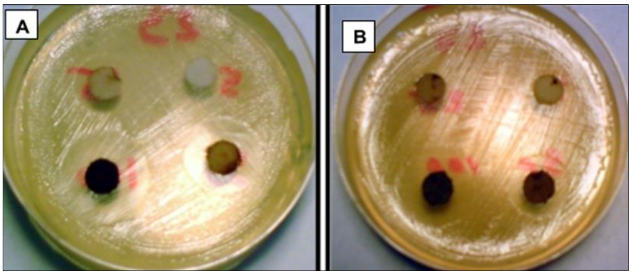


Fig 6: Antibacterial activity of complexes: A) N2 and B) C2

Conclusion

In conclusion, two ligands derived from sulfathiazole, L1 and L2, were successfully synthesized through condensation reactions with 2-aminobenzaldehyde and 2-nitrobenzaldehyde, respectively. Their cobalt(II) and nickel(II) complexes were also prepared and characterized using different physicochemical and spectroscopic techniques.

FTIR and ^{1}H NMR spectral data confirmed the formation of the azomethine group and supported the proposed structures of the ligands. The shifts notice in the FTIR spectra after complexation, together with the appearance of M–N bands, indicated coordination of the ligands to the metal ions through nitrogen donor atoms. Molar conductivity measurements confirmed the non-electrolytic nature of the complexes, suggesting that chloride ions are coordinated within the inner coordination sphere. Atomic absorption, electronic spectra, and magnetic susceptibility data supported the proposed formulas and geometries of the prepared complexes. The antibacterial activity results showed that the metal complexes exhibited higher inhibitory activity than the free ligands against both Escherichia coli and Staphylococcus aureus, although their activity remained lower than that of Imipenem.

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