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Synthesis and Antibacterial Investigation of Triphenylphosphine-Stabilized Cobalt Schiff Base Complexes

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ABSTRACT
This study reports the preparation, characterisation and antibacterial activity of cobalt (II) Schiff base complexes (C1-C5) bearing triphenylphosphine (PPh_3) as a co-ligand. Five Schiff base ligands (L1 to L5) were prepared by condensation of selected aldehydes and ketones with primary amines or hydrazines under acid catalysis in ethanol. The synthesised compounds were characterised by infrared (FTIR) spectroscopy, ultraviolet-visible (UV-Vis) spectroscopy and magnetic susceptibility measurements. In vitro antibacterial screening was carried out using agar well diffusion method against *Escherichia coli*, *Staphylococcus aureus*, *Klebsiella pneumoniae*, and *Pseudomonas aeruginosa*. FTIR analysis revealed coordination through the azomethine bond with characteristic bands at 1556 and 1684 cm^{-1} with C–N ν (cm^{-1}) between 1306 and 1338 cm^{-1}, and the C–P ν (cm^{-1}) of the PPh_3 observed between 1075 and 1088 cm^{-1}. The UV-Vis absorption peaks of 368, 342, 367 and 377 nm in complexes C1, C2, C3 and C5 respectively were assigned to respective Ligand to Metal Charge Transfer (LMCT). Magnetic susceptibility measurements revealed paramagnetic behaviour in C1, C4 and C5 with chi g values between 1.2 x10 $^{-7}$ and 1.85x10^{-6}, while C2, (-6.3 x10 $^{-7)}$ and C3 (-8 x10 $^{-8}$) were diamagnetic. The antimicrobial results showed that the cobalt (II) complexes were substantially more active than their free-ligand against all four bacterial test organisms. The particularly notable anti-Pseudomonal activity in all five cobalt (II) complexes. The complexes displayed inhibition zone ranging from 20-29 mm, surpassing the standard drug Ofloxacin (23.50 mm) and showing significant enhancement over the free PPh_3, ligands and cobalt salt.

Keywords: Schiff base, triphenylphosphine, co-ligand, antimicrobial activity

INTRODUCTION

Schiff bases stand out because their ease of preparation, structural versatility, and reported capability of coordinating to a wide range of transition metal ions (Aggoun *et al.*, 2020). Schiff bases are particularly attractive due to the flexibility of their electronic and steric properties can be tuned simply by changing the aldehyde or amine starting material. Cobalt(II) occupies a special position among the first-row transition metals With an atomic number of 27 and a ground-state electron configuration of $[Ar]3d^7\ 4s^2$, cobalt in the +2 oxidation state carries a d^7 configuration that allows it to adopt octahedral, tetrahedral and square pyramidal geometries depending on the type of ligands surrounding it(Devi *et al.*, 2022). This flexibility, combined with the rich magnetic and spectroscopic behaviour that comes with d^7 systems, has made cobalt (II) a favourite probe for studying ligand field effects.

Ancillary ligands are those that stabilize the metal centre and tune its electronic and steric properties but are not the primary focus of the reactivity. Triphenylphosphine, commonly written as PPh_3, is one of the most widely used co-ligands in the whole of organometallic and coordination chemistry. PPh_3 acts as a σ-donor via its phosphorus lone pair and significantly as a π-acceptor due to the presence of empty d-orbitals (or σ* orbitals) on the phosphorus atom, which can accept electron density from filled metal d-orbitals(Iaroshenko, 2019). This π-backbonding capability helps to stabilize electron-rich metal centers. It can also distort the coordination geometry, potentially enhancing the biological activity by modifying the complex's shape and lipophilicity (Kumah *et al.*, 2021).

The concept of mixed-ligand or heteroleptic complexes, combining a planar Schiff base with a bulky phosphine, is a powerful strategy in medicinal inorganic chemistry thus its synergistic combination often results in improved lipophilicity, facilitating transport across cell membranes(Odulaja *et al.*, 2026b). Several studies have demonstrated that Co (II) complexes incorporating phosphine ligands exhibit superior cytotoxicity and antimicrobial activity compared to their phosphine-free counterparts. The phosphine ligand can enhance the stability of the complex in physiological media and may also participate in secondary interactions with biological targets, such as DNA backbone phosphates(Amolegbe *et al.*, 2015). Hence, the integration of PPh_3 into the Co (II)-Schiff base architecture is a deliberate design aimed at maximizing biological potency. In this study, PPh_3 is employed as an ancillary ligand alongside the primary Schiff base. Despite the extensive literature on Co (II) Schiff base complexes, the specific combination of a Schiff base derived from 2,4-dinitrophenylhydrazine and acetylpyridine, benzoylpyridine and 3-pyridinecarboxaldehyde has received limited attention, particularly regarding its coordination behaviour with Co(II) in the presence of bulky phosphine ligands like triphenylphosphine. Most studies investigated them as analytical reagents or simple binary complexes, overlooking the potential synergistic effects of mixed-ligand

engineering, there is also a paucity of information on their magnetic properties and antibacterial activities(Yusuf *et al.*, 2021).

Its three phenyl rings give the complex a substantially higher lipophilicity which enables metal-based compounds penetrate bacterial cell membranes more effectively(Odulaja *et al.*, 2026a). The combined effect of PPh_3 as an ancillary ligand on the physicochemical and antimicrobial properties of cobalt (II) complexes carrying the five specific Schiff base ligands targeted in this work has not been reported comprehensively. There is also a notable shortage of data for cobalt(II) PPh_3 Schiff base systems tested against Klebsiella pneumoniae, which is a clinically important Gram-negative pathogen of growing concern in antimicrobial resistance(Murray *et al.*, 2022)

MATERIALS AND METHODS

Preparation of Schiff Base Ligands (L1 to L5)

Five Schiff base ligands were prepared by acid-catalysed condensation in ethanol. For each ligand, the aldehyde (1 mmol dissolved in 10 mL ethanol) and the amine or hydrazine component (1 mmol dissolved in 10 mL ethanol) were combined in a 100 mL round-bottom flask. Three drops of glacial acetic acid were added as catalyst and the mixture was stirred at 200 to 240 revolutions per minute (rpm) for six hours at 70 °C temperature. Products were isolated by vacuum filtration using a Buchner funnel, washed twice with diethyl ether and dried to constant weight in air.

Preparation of Cobalt Precursor

The cobalt(II) PPh_3 complex (CT) was prepared as described by (Odulaja *et al.*, 2024a)with little modification. Solution A: 20 mmol (4.8 g) of $CoCl_2.6H_2O$ was dissolved in 30 mL of ethanol to give a characteristic purple-blue solution. Solution B: 20 mmol (12.6 g) of PPh_3 was dissolved in 30 mL of ethanol at 90 °C using a reflux condenser setup. The cobalt solution was added dropwise to the refluxing PPh_3 solution with continuous stirring, producing a bright blue solution immediately. The reaction mixture was refluxed for a further 30 minutes and then quenched rapidly by immersion in an ice and sodium chloride bath. Sky-blue crystals formed and were collected by filtration, weighed and stored in a sealed vial until needed.

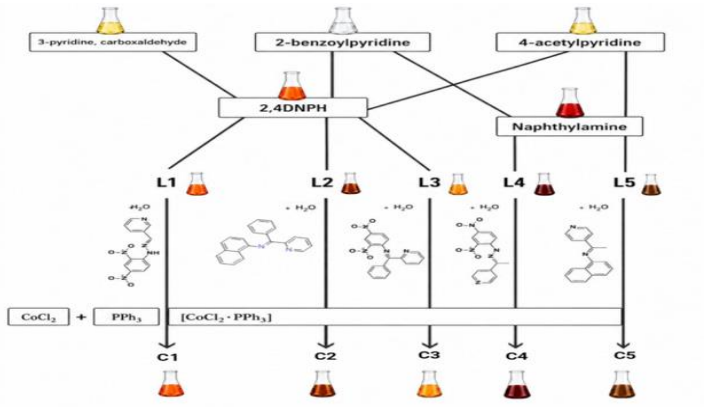

Figure 1: Schematic diagram of the synthesis procedure

Synthesis of Cobalt Complexes (C1- C5)

The synthesized Schiff base ligands (L1–L5) were individually coordinated with the previously prepared [Co $(PPh_3)_2$] precursor complex to obtain the corresponding cobalt complexes. For each reaction, 2 mmol of the metal precursor complex was dissolved in ethanol, followed by the addition of 2 mmol of the corresponding Schiff base ligand. The resulting mixture was stirred nonstop at room temperature using a magnetic stirrer for a minimum duration of 2 hours to allow the reaction proceed to completion after which the products were filtered, washed with diethyl ether and recrystalised with ethanol and then dried with $MgSO_4$

Characterisation Methods

FTIR Spectroscopy

Spectra were recorded as KBr pellets over the range 4000 to 650 cm^{-1} using an Agilent Technologies FTIR spectrometer. Pellets were prepared by grinding approximately 2 mg of sample with 100 mg of dried KBr and pressing under vacuum.

UV-Vis Spectroscopy

Spectra were recorded from 200 to 800 nm in DMSO solution at a concentration of approximately 1×10^{-4} mol/L using a UV-Visible spectrophotometer with 1 cm quartz cuvettes.

Magnetic Susceptibility

Magnetic susceptibility was determined at room temperature by the Gouy balance method at Jabio Laboratory Services.

Antimicrobial Assay

In vitro antimicrobial activity was determined by the agar disc diffusion method. Test organisms were Escherichia coli (EC), Staphylococcus aureus (SA), Klebsiella pneumoniae (KP) and Pseudomonas aeruginosa (PA). All compounds were dissolved in DMSO at a concentration of 1 mg/mL. Filter paper discs (6 mm, Whatman) were impregnated with 20 microlitres of each solution and placed on Mueller Hinton agar plates that had been inoculated with the appropriate test organism. Ofloxacin served as the positive antibacterial control and DMSO served as the negative control. All plates were incubated at 37 °C for 24 hours. Zones of inhibition were measured in triplicate and the mean values recorded (Benzerka *et al.*, 2012).

RESULTS AND DISCUSSION

Fourier Transform Infrared Spectroscopic Analysis

In C1 which carries L1, derived from 3-pyridinecarboxaldehyde and 2,4-DNPH, two C=N stretching bands appear at 1582.5 and 1613.9 cm^{-1}. The presence of two distinct bands in this region suggests that the L1 ligand may coordinate in a bidentate fashion involving both the azomethine nitrogen and the pyridinyl nitrogen of the 3-

pyridinyl moiety. The P-C stretching at 1084.7 cm^{-1} confirms PPh_3 coordination, and the broad N-H/O-H band at 3073 cm^{-1} suggests either hydrogen-bonded water molecules or N-H groups from the 2, 4-DNPH fragment participating in intermolecular interactions.

C2: Which incorporates the 2-benzoylpyridine-derived L2 ligand, a single C=N band appears at 1654.9 cm^{-1}. The ketone-derived Schiff base has a somewhat different electronic environment compared to the aldehyde-derived L1, and the relatively high position of the C=N band at 1654.9 cm^{-1} within the coordinated complex suggests that the cobalt centre in C2 draws less electron density from the azomethine bond than in C1, possibly because the 2-benzoylpyridine framework imposes steric constraints that affect the Co-N bond angle and therefore the degree of π-backbonding. The characteristic P-C stretching at 1075.3 cm^{-1} confirms PPh_3 retention (Fudulu *et al.*, 2020).

C3: derived from L3 which uses 4-acetylpyridine as the carbonyl component, two C=N bands appear at 1586.0 and 1571.1 cm^{-1}. These are the lowest C=N stretching frequencies in the entire series, suggesting the strongest degree of cobalt-nitrogen interaction in this complex. This electronic interpretation is supported by the diamagnetic character of C3, which points to a cobalt(III) centre surrounded by strong-field donors(Khalaf *et al.*, 2026).

C4: Arising from L4 prepared from 4-acetylpyridine and mesitylamine, presents an intriguing case. The absence of observable C=N stretching in the FTIR spectrum is unusual and may indicate that the imine formed from methylamine undergoes hydrolysis or rearrangement during complexation, leaving PPh_3 as the dominant coordinating species, or the C=N band is obscured by overlapping C-N stretching bands at 1306 cm^{-1} (Fernandes *et al.*, 2020). The paramagnetic nature of C4 as revealed by magnetic susceptibility is consistent with a cobalt(II) species where the ligand field provided by the methylamine-derived Schiff base is weaker than for the 2,4-DNPH-bearing analogues.

C5: Stands apart from the rest of the series by virtue of its 1-naphthylamine component, which brings a large, flat aromatic ring system into the complex. The two C=N bands at 1556.2 and 1654.9 cm^{-1} span the widest frequency range in the series. The lower band at 1556.2 cm^{-1} is particularly low, suggesting very strong metal-nitrogen interaction at one azomethine site. In all, the FTIR data showed that the Schiff base ligands are coordinated through the azomethine nitrogen; second that pyridinyl nitrogen as revealed by the presence of C-N stretching frequency in the range of 1200-1313 cm^{-1} in all the complexes corroborates the coordination through the pyridinyl nitrogen, similarly the appearance of C-P frequency around 1056- 1088 cm^{-1} in all five complexes is an indication of retention of PPh_3.

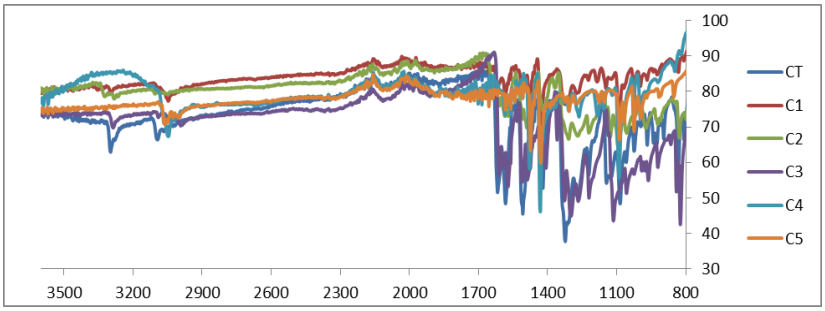

Figure 2: FTIR Spectra of prepared cobalt (II) complexes

Table 1: Summary of FTIR spectral data of ligands and complexes

Functional Group	CT	C1	C2	C3	C4	C5
C=N(Azomethine)ν(cm^{-1})	Absent	1582–1613	1654	1586–1571	Not obs	1556
C–H(Aromatic) ν(cm^{-1})	3274,3063	3073,3267	3088,3276	2996, 3259	3039	3034
C–N ν(cm^{-1})	Absent	1313	1338	1303	1306	1200
C–P ν(cm^{-1})	Absent	1084	1075	1056	1088	1086

UV-VIS Spectroscopy

UV-Visible spectra were recorded for two representative cobalt (II) complexes, C1 and C3, in DMSO solution over the range 300 to 500 nm. C1, an intense sharp absorption rises at approximately 313 to 368 nm with an absorbance of 0.7 at 367 nm. This is assigned to a Ligand to Metal Charge Transfer (LMCT) transition arising from electron donation out of the ligand pi system into the cobalt d- orbitals(Fudulu *et al.*, 2020). The presence of this LMCT band is itself strong indication that the ligand is directly bound to cobalt. For C2, the absorbance rises to a broad maximum at approximately 342 nm with an absorbance value of 1.53, which is assigned to π - π * transitions of the conjugated azomethine chromophore. The absorbance then decreases smoothly to a shoulder at about 403 nm. For C3 the same pattern is seen but with a blue-shifted maximum near 367 nm with absorbance 1.18, compared to 361 nm for C2. This difference reflects the different degree of π conjugation in ligands C2 and C3(Genchi *et al.*, 2023).

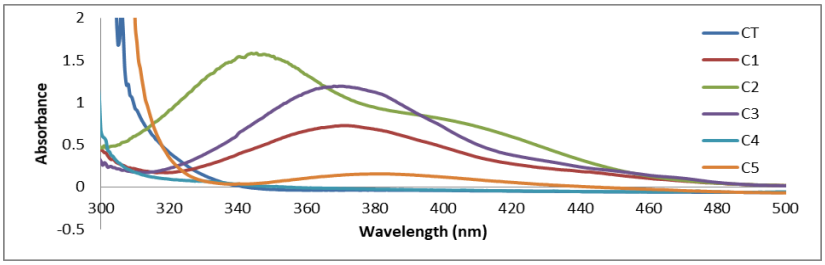

Figure 3: Summary of UV-Vis spetcra of complexes C1-C5.

The presence of this LMCT band is itself strong confirmation that the ligand is directly bound to cobalt. For C2, the absorbance rises to a broad maximum at approximately 342 nm with an absorbance value of 1.53, which is assigned to π - π * transitions of the conjugated azomethine chromophore. The absorbance then decreases smoothly to a shoulder at about 403 nm. For C3 the same pattern is seen but with a blue-shifted maximum near 367 nm with absorbance 1.18, compared to 361 nm for C2. This difference reflects the different degree of π conjugation in ligands C2 and C3.

The complex of L4 prepared from 4-acetylpyridine and mesitylamine, presents an interesting case. The absence of absorption in C4 is in attributable to obscured C=N band possibly overlapped by the PPh_3, moreso the UV Vis spectrum of C4 is very similar to CT corroborating the possibility of PPh_3 as the dominant coordinating(Ghosh *et al.*, 2018). The paramagnetic nature of C4 as revealed by magnetic susceptibility is consistent with a cobalt(II) species where the ligand field provided by the methylamine-derived Schiff base is weaker than for the 2,4-DNPH-bearing analogues. For C5, the absorbance rises to a broad maximum at approximately 377 nm with an absorbance value of 0.15, which is assigned to π - π * transitions of the conjugated azomethine chromophore(Genchi *et al.*, 2023; Raj, 2015) The absence of clearly resolved d-d bands in the 400 to 700 nm region in all spectra is consistent with low-spin cobalt(II) or cobalt(III) in a strong ligand field, where d-d transitions are Laporte-forbidden and consequently very weak (Djekić *et al.*, 2006)

Magnetic Susceptibility

Complexes C1, C4 and C5 showed positive chi g values and are paramagnetic, indicating the presence of one or more unpaired electrons in the cobalt (II) centre. The particularly large chi g value for C5 points to high-spin octahedral cobalt (II) with three unpaired electrons showed positive chi g values and are therefore paramagnetic, meaning they have unpaired electrons while complexes C2 and C3 showed negative chi g values and are diamagnetic, in the context of cobalt chemistry this points to low-spin cobalt (III) with a d^6 configuration. The formation of cobalt(III) is consistent with the well-known tendency of cobalt(II) Schiff base complexes to be oxidised by atmospheric oxygen during synthesis, especially when strong-field nitrogen donor ligands and a pi-acceptor phosphine are both present (Özçelik *et al.*, 2013). The variation in magnetic behaviour across the series makes sense in terms of the different ligand field strengths of L1 to L5: the hydrazine-type ligands L1 to L3 carrying the electron-withdrawing 2,4-dinitrophenyl group impose a stronger ligand field and are more likely to push the cobalt towards lower spin states (forced pairing) or oxidation state changes as suspected with C2 andC3, while the imine-type ligands L4 and L5 impose a weaker field and favour the retention of cobalt(II) (Biswas *et al.*, 2010; Çapan *et al.*, 2026a)

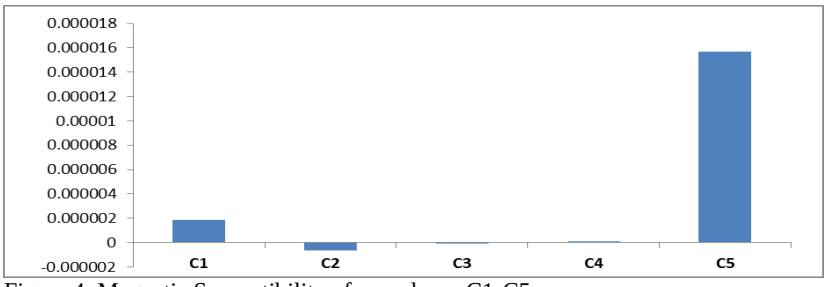

Figure 4: Magnetic Susceptibility of complexes C1-C5

Table 2: Summary of magnetic susceptibility of C1-C5 and interpretation

Cobalt complex	chi g value	Magnetic Character	Electron Interpretation	Proposed Spin State	Possible Structural Implication
C1	1.85E-06	Paramagnetic	Presence of unpaired electron	Low-spin Co(II), d7	Low spin cobalt configuration
C2	-6.3E-07	Diamagnetic	No unpaired electron	Low-spin Co(III), d^6	Possible octahedral low spin geometry
C3	-8E-08	Slightly diamagnetic	Nearly balanced electron pairing	Low-spin Co(III), d^6	Borderline magnetic behavior
C4	1.2E-07	Weakly paramagnetic	Very low magnetic moment	Low-spin Co(II), d7 1 unpaired electron	Possible distorted geometry
C5	1.57E-05	Strongly paramagnetic	Highest number of unpaired electron	High-spin Co(II), d7 3 unpaired electrons	Strong high spin cobalt

In Vitro Antimicrobial Activity

The cobalt (II) complexes showed dramatically enhanced activity across all four bacterial organisms compared to the free ligands, the cobalt salt and PPh_3 alone. Zones of inhibition ranged from 20 to 29 mm, placing several of the complexes at a level that matches or exceeds the ofloxacin positive control.

Against EC, C3 was the most active compound in the entire study at 29 mm, comfortably exceeding ofloxacin at 26 mm. C1 and C2 were also highly active at 27 and 26 mm

respectively. Against SA, C3 was again the standout compound at 27 mm compared to 24 mm for ofloxacin, with C1 very close behind at 26 mm. Against KP, C1 was the most active at 26 mm, exceeding ofloxacin at 22 mm, with C3 and C4 at 25 and 24 mm also performing well.

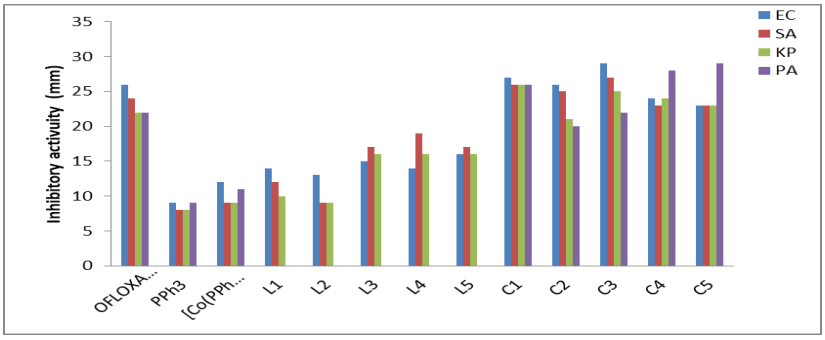

Figure 4: Inhibitory activities of ligands and their complexes

The most remarkable result of the entire study came from the activity against PA. All five free ligands were completely inactive against this organism, yet every single one of the five cobalt (II) complexes produced a detectable and substantial zone of inhibition. C5 was the most active compound against PA at 29 mm, and C4 followed closely at 28 mm. Both values substantially exceed the ofloxacin control of 22 mm. C1 gave 26 mm, C3 gave 22 mm and C2 gave 20 mm. This pattern leaves no doubt that the cobalt PPh_3 framework is absolutely essential for activity against Pseudomonas aeruginosa (Çapan *et al.*, 2026b; Odulaja *et al.*, 2024b).

CONCLUSION

This project investigated the synthesis, characterisation and in vitro antimicrobial activity of five mixed-ligand cobalt (II) schiff base complexes bearing triphenylphosphine as an ancillary ligand, restricting the study entirely to cobalt chemistry. The synthesis route involved first preparing five Schiff base ligands L1 to L5 and the [Co $(PPh_3)_2$] independently, then combining them at room temperature to give the five complexes C1 to C5.

Characterisation by FTIR, UV-Vis spectroscopy and magnetic susceptibility provided consistent evidence of complex formation. The antimicrobial results showed that the cobalt (II) complexes are substantially more active than their free-ligand counterparts against all four bacterial test organisms. The particularly notable finding of anti-Pseudomonal activity in all five cobalt (II) complexes, despite low activity in the corresponding free ligands, highlights the critical role of the metal PPh_3 framework in enabling membrane penetration of the complex against this intrinsically resistant pathogen.

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