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Research Article

# Antimicrobial potentials of N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine) and its Mn(II), Fe(II), Co(II) and Ni(II) complexes

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## ABSTRACT

A salophen ligand (L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)) was synthesized by refluxing 4-bromobenzene-1,2-diamine with 2-pyridinecarboxaldehyde (1:2 mole ratio, respectively) in ethanol. M(II) complexes (M = Mn, Fe, Co and Ni) of the ligand were synthesized by refluxing the ligand solution with the corresponding M(II) salt solution (1:1 mole ratio) in ethanol. The synthesized compounds were characterized using Fourier transform infrared (FTIR) spectroscopy, CHN elemental analysis, magnetic moments, and molar conductivity. FTIR spectra of the ligand showed a strong band at 1588 $cm^{-1}$ for the C=N stretching vibrations. The band shifted to higher frequency (1596–1603 $cm^{-1}$) in the M(II) complexes signifying complexation. CHN elemental analysis showed strong agreement with theoretical values for the ligand and the M(II) complexes, supporting the proposed formula of the compounds. All the samples were non-electrolyte due to the low molar conductance values (17.12–55.40 $\Omega^{-1}cm^2mol^{-1}$). Effective magnetic moments (3.23–5.97 BM) of the complexes indicated their paramagnetic nature and octahedral geometry. The samples were tested for antimicrobial activity against four bacterial and two fungal isolates. Generally, the complexes were more potent than the free ligand against all the tested isolates. The increasing order of antimicrobial potential of the compounds follows: L < $[Ni(II)(L)_2]$ < $[Mn(II)(L)_2]$ < $[Fe(II)(L)_2]$ < $[Co(II)(L)_2]$.

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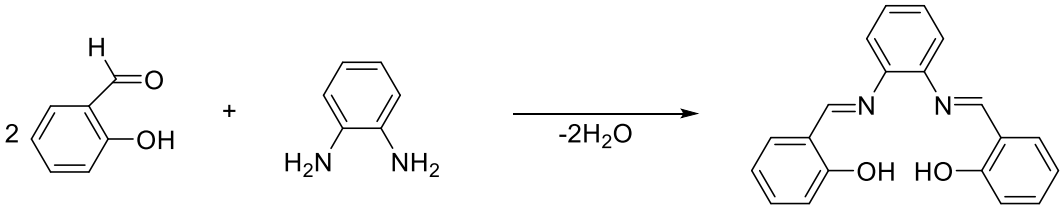
## INTRODUCTION

Conventionally, Schiff bases are a condensation product of aldehyde and primary amine. They are characterized by an imine bond (-C=N-), which is considered hemilabile due to the high binding ability of the nitrogen of the imine group (Sekar *et al.,* 2022). There are various classes of Schiff bases according to the functional group that is attached to its imine group, which include hydrazones, Polymeric, Salicylidene, pyrimidyl, hydrazide-hydrazones, salene and salophen-type among others (Malik *et al.,* 2017). Salophen-type ligands are those in which o-phenylenediamine is employed in place of ethylenediamine in salene-type ligands. These types of ligands are prepared by a typical condensation reaction which involves a reaction between two equivalents of salicylaldehyde or its derivatives and one equivalent of 1,2-phenylenediamine as shown in (Scheme 1) below (Micha *et al.,* 2020).




**Scheme 1:** Synthesis of salophen-type ligand.

Usually, these reactions do not require a catalyst and proceed straightforwardly, but sometimes the products may be hydrolyzed in a reversible manner (Sobel *et al.,* 2020). To overcome this problem, dehydrating agents or molecular sieves (with size of about 3 Å) are used to remove the water molecules produced during the reaction (Erxlenben, 2018). Sometimes template synthesis is also performed to get metal-salen complexes directly first, by preparing a metal-salicylaldehyde complex *in-situ* as templating agent, then ethylenediamine is added to get a target salophen ligand. Although salophen ligands are sensitive to hydrolysis (Scheme 1), their metal complexes are quite stable. Thus, to avoid the hydrolysis of a virgin salophen ligand, their corresponding metal complexes are often used as template for transmetallation reaction (Gavin *et al.,* 2024).

Salophen ligands are generally interesting and excellent coordinating molecules. The imino N is readily coordinating and their metal complexes can exhibit variety of geometry. These ligands and their metal complexes were reported to exhibit a variety of applications such as antibacterial, antifungal, anti-tuberculosis, antimalarial and antiviral (Nazirkar *et al.,* 2019).

For the past two decades, a large number of metal ions have been introduced to salophen to produce a variety of complexes (Atwood and Harvey (2001); Clarke and Storr (2014); Karmakar and Chattopadhyay, (2019)). Further, they have been reported as good candidates for other applications such as in homogenous catalysis, biochemistry, electrochemistry, sensors, molecular magnetism and materials science (Yuan *et al.,* (2022); Gavin *et al.,* (2024)). A number of reviews on salophen ligands and their metal (II) complexes have been published (Cozzi (2004); Baleizão and Garcia (2006); Pessoa and Correia (2019); Yuan *et al.,* (2019)).

The promising biological performance of this class of ligands and their M(II) complexes are well documented. Thus, this work aimed to design and synthesise a new salophen ligand and its M(II) complexes in an attempt to discover a more potent antimicrobial agent.

## MATERIALS AND METHODS

### Materials

All chemicals used in this work were of analytical grade and used without further purification. All glassware were washed with detergent after soaking in conc. $HNO_3$, thoroughly rinsed with distilled water and dried in an oven at 120 ºC. Weighing was conducted using electrical Metler balance model AB54. Infrared spectra were determined using Fourier transform infrared spectrophotometer (FTIR-8400S) in the range 400-4000 $cm^{-1}$. Electrical conductance was measured using Jenway conductivity meter model 4010 range 20-200 µs. Melting points were obtained using microprocessor melting point apparatus (WRS-IB). Magnetic susceptibility was determined using magnetic susceptibility balance MKI Sherwood scientific ltd. Elemental analyses were recorded using Series II CHNS/O 2400 Perkin Elmer. The UV-Vis spectral information was

established using Perkin Elmer UV-Vis Spectrophotometer Lambda-35.

**Methods**

**Synthesis of the salophen ligand (L)**

The salophen ligand (L), which was derived from 4-Bromobenzene-1,2-diamine and 2-pyridenecarbozaldehyde, was synthesized according to Shaban *et al.,* (2024) with slight modification. Exactly 0.214 g of 2-pyridenecarbozaldehyde (2 mmol) was dissolved in boiling ethanol (99%, 30mL) and mixed with a hot solution of 4-Bromobenzene-1,2-diamine (0.187 g, 1 mmol) in 20mL absolute ethanol (99%). The mixture was heated under reflux for about 3 hours with constant stirring, and then cooled to room temperature to allow precipitation. The grey solid formed was filtered off, washed several times with double distilled water and cold ethanol, and dried in a desiccator over $P_2O_5$ for 24 hours. This was later characterized by various spectroscopic techniques before being applied for complex formation.

**Synthesis of the M(II) complexes of the salophen ligand (L)**

M(II) complexes (M= Mn, Fe, Co and Ni) of the synthesized ligands were synthesized according to the method described by Li *et al.,* (2024). In each case, 0.365 g (2 mmol) of the ligand (L) was dissolved in 30 mL of absolute ethanol (99%) and heated to 40 °C on a water bath. 1 mmol of the respective M(II) nitrate (M = Mn, Fe, Co and Ni) solution in ethanol was slowly added into a round bottom flask containing the hot ligand solution. The resulting mixture was stirred for 10 minutes and then reflux for 4 hours at 80 °C. The solution was cooled to room temperature to allow precipitation, followed by filtration. The resulting solid products were then rinsed several times with double distilled water, cold ethanol and diethylether and dried a desiccator over $P_2O_5$ for 24 hours (Li *et al.,* 2024). These were later characterized using various techniques before further application.

**Antimicrobial Activity Test**

Four bacterial (*Streptococcus pyogenes, Bacillus subtilis, Salmonella typhi* and *Klebsiella pneumoniae*) and two fungal (*Candida albicans* and *Aspergillus fumigatus*) species were obtained from the microbiology laboratory of the Aminu Kano Teaching Hospital, Kano and identified at the post graduate research microbiology laboratory, Department of Microbiology, Bayero University, Kano.

The sensitivity test was carried out by the Well Diffusion Method according to Yusha'u and Salisu, (2011). First, sterile Nutrient Agar (NA) and Potato Dextrose Agar (PDA) were used as bacterial and fungal media, respectively. These were carefully transferred into sterile Petri dishes to an appreciable amount and cooled to solidification at room temperature. The Petri dishes were named according the test organism while position of the wells were marked based on the test concentrations (1000, 500, 250 µg/ml) of the salophen ligand and the M(II) complexes. From the standardized inoculums of each isolate, uniform spreading (using a glass spreader) of 0.1 mL of the bacterial and fungal inoculums was done on the surface of the dried NA and PDA, respectively. The tests were carried out in triplicate. In each case, the different concentrations of the test samples were placed using micro dropper before incubation. For the bacteria, the petri-dishes were kept in an incubator for 24 hours at 37ºC, while for the fungi, the Petri dishes were kept in a cool dry place for 72 hours, and the plates were observed for the presence of zones of inhibition as evidence of antibacterial and antifungal activities, respectively. The degree of sensitivity was determined by measuring the diameter of visible zones of inhibition to the nearest millimeters with respect to each isolate and test concentrations of the samples.

**RESULTS AND DISCUSSION**

**Physico-chemical characterization of the salophen ligand (L) and its M(II) complexes**

The salophen ligand, N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine) and its Mn(II), Fe(II), Co(II) and Ni(II) complexes were successfully synthesized by simple reflux in ethanol. Both the ligand and the complexes were characterized using various analytical and spectroscopic techniques and the following results are obtained.

**Table 1:** Physical properties of the salophen ligand (L) and its M(II) complexes

| Compound | Colour | Melting Point (°C) | %Yield |
|---|---|---|---|
| Ligand (L) | Brown | 140 | 87.40 |
| $[MnLCl_2].2H_2O$ | Grey | 190 | 71.26 |
| $[FeLCl_2].2H_2O$ | Reddish Brown | 200 | 81.29 |
| $[CoLCl_2].2H_2O$ | Green | 260 | 66.64 |
| $[NiLCl_2].3H_2O$ | Greenish-brown | 300 | 66.68 |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)

The salophen ligand (L) was successfully synthesized as a thermally stable light brown amorphous solid (Table 1) in high yield (87.40%). All the M(II) complexes were intensely coloured as shown in (Table 1), attributable to electronic transition within the d-orbitals of the metal ions, charge transfer from metal to ligand and *vice versa* or nature of the ligand (Oliver and Christina, 2021). The melting point of the complexes were higher (190-300 °C) than the salophen ligand (140 °C), indicating a comparably higher thermal stability of the complexes. These observations are in close proximity with the literature (Haque and Salam (2015); Baecker *et al.*, (2020)).

**Table 2: Solubility of the salophen ligand (L) and its M(II) complexes**

| Compound | n-hexane | Diethyl Ether | EtOH | MeOH | $H_2O$ | DMF | DMSO |
|---|---|---|---|---|---|---|---|
| Ligand (L) | IS | SS | SS | SS | IS | S | S |
| $[MnLCl_2].2H_2O$ | IS | IS | S | S | IS | S | S |
| $[FeLCl_2].2H_2O$ | IS | SS | SS | SS | SS | S | S |
| $[CoLCl_2].2H_2O$ | IS | IS | SS | SS | SS | S | S |
| $[NiLCl_2].3H_2O$ | IS | IS | S | S | IS | S | S |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine) **KEY:** S = Soluble, IS = Insoluble and SS = Slightly soluble.

The solubility of the samples was found to be independent of polarity. They were all insoluble in n-hexane and $H_2O$, and readily soluble in DMSO and DMF (Table 2). The samples were invariably sparingly soluble in other solvents. The solubility of the compounds in DMSO provides an avenue to conduct the antimicrobial studies in DMSO since it works perfectly as a blank control for most pathogens showing negligible or no antimicrobial activity (Sani and Dailami, 2015).

**Table 3:** CHN chemical composition of the salphen ligand (L) and its M(II) complexes

| Compound | Elemental Composition: Theoretical (Experimental) | | | |
|---|---|---|---|---|
| | %C | %H | %N | %Metal |
| Ligand (L) | 59.19(60.99) | 3.59(3.94) | 15.34(15.23) | - |
| $[MnLCl_2].2H_2O$ | 41.02(41.54) | 3.25(3.21) | 10.63(10.78) | 10.63(10.42) |
| $[FeLCl_2].2H_2O$ | 40.94(40.64) | 3.25(3.37) | 10.61(10.52) | 06.06(06.53) |
| $[CoLCl_2].2H_2O$ | 40.71(40.39) | 3.23(3.43) | 10.55(10.28) | 11.10(10.99) |
| $[NiLCl_2].3H_2O$ | 39.39(39.75) | 3.49(3.16) | 10.21(10.39) | 10.69(10.27) |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)

The chemical composition of the salophen ligand and the M(II) complexes is presented (in Table 3), which was obtained based on the CHN elemental analysis of the samples. The percentage of C, H, and N in the ligand (L) and the M(II) complexes were compared with calculated values of the proposed molecular formular of the compounds and it was found to be in close proximity (Baecker *et al.*, (2020)). The slight variation in the experimental values compared to the theoretical ones could suggest presence of minor impurities such as surface

adsorbed water molecules (other than water of crystallization) in the samples. The percentage of the metal in each of the complexes were obtained by back calculation based on the percentage residual fraction of the proposed formula of the complexes, after several trial and error. Overall, these data supported by other characterization results are indication of the successful synthesis of the salophen ligand and the M(II) complexes.

**Spectroscopic and other characterization of the salophen ligand (L) and its M(II) complexes**

**FTIR**

The FTIR spectral data of the salophen ligand (L) and the M(II) complexes were compared to confirm complexation. Table 4 shows the FT-IR spectral interpretation of the samples while the spectra are attached as APPENDIX at the end of the manuscript. The band appearing at 1588 $cm^{-1}$ is due to stretching vibration of the imine group in the ligand, which often than not shift to higher frequency to reflect complexation (Bayeh *et al.*, 2020). In this case, the M(II) complexes showed bands at 1596-1603$cm^{-1}$, which strongly suggests the involvement of the imine nitrogen in the formation of the M(II) complexes (Mirza, *et al*., 2014). In the same vein, the (C=N) bond of the pyridine ring in the ligand band showed vibration band at 706 $cm^{-1}$, which shifted to higher frequencies in the range 611–696 $cm^{-1}$, also indicating the participation of the pyridine nitrogen complex formation (Tas *et al.*, 2018).

**Table 4:** IR Spectra of the ligand (L) and the M(II) complexes

| Compound | V(C=N) $cm^{-1}$ | V(M-N) $cm^{-1}$ | V(C=N)py $cm^{-1}$ | V(M-N)py $cm^{-1}$ | V(M-Cl) $cm^{-1}$ | V($H_2O$) $cm^{-1}$ |
|---|---|---|---|---|---|---|
| Ligand (L) | 1588 | - | 706 | - | - | - |
| $[MnLCl_2].2H_2O$ | 1596 | 607 | 637 | 508 | 462 | 3041 |
| $[FeLCl_2].2H_2O$ | 1599 | 515 | 589 | 474 | 452 | 3059 |
| $[CoLCl_2].2H_2O$ | 1599 | 615 | 692 | 533 | 455 | 3059 |
| $[NiLCl_2].3H_2O$ | 1603 | 589 | 696 | 474 | 415 | 3063 |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)

These observations provide an obvious indication that the ligand actually act tetradentally and coordinate to the metal (II) ions via (1) the two azomethine N and (2) the two pyridine hetero N. Further analysis of the spectra revealed two new bands in the M(II) complexes, which were originally absent in the spectrum of the ligand. These new bands were in the range of 474-533 $cm^{-1}$ and 515-615 $cm^{-1}$ assigned to the stretching vibrations of the metal-nitrogen (pyridine) and metal-nitrogen (imine) bonds, respectively (Salem *et al.*, 2021). These new bands further support the argument that the ligand coordinated to the M(II) ions in a tetradentate manner. Shakir *et al.*, (2009) assigned a set of medium intensity bands in the range of 410–460 $cm^{-1}$ to (M–Cl) bonds stretching vibrations. Conventionally, broad and strong intense bands in the range of 3041–3093 $cm^{-1}$ are assigned to the stretching vibration of the O-H bonds in water molecules present as water of crystallization in the M(II) complexes (Conti *et al.*, 2025).

**Table 5:** Electronic transition of the salophen ligand (L) and its M(II) complexes

| Compound | $\pi \longrightarrow \pi^*$ (nm) | $n \longrightarrow \pi^*$ (nm) |
|---|---|---|
| Ligand (L) | 232 | 336 |
| $[MnLCl_2].2H_2O$ | 247 | 334 |
| $[FeLCl_2].2H_2O$ | 218 | 359 |
| $[CoLCl_2].2H_2O$ | 240 | 358 |
| $[NiLCl_2].3H_2O$ | 247 | 350 |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)

The UV–Vis spectra of the salophen ligand (L) and the M(II) complexes were summarized (in Table 5). The electronic transition of the samples were recorded in DMSO at a wavelength of 200 to 700 nm. Two absorption bands at 232 and 336 nm were assigned to intra ligand $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ transitions, which showed slight shift to higher wavelength of 218-247 nm for $\pi \rightarrow \pi^*$ and 334-359 nm for $n \rightarrow \pi^*$ transition upon complexation. Both Tas

*et al.* (2018) and Wu *et al.,* (2020) reported similar observations. This, yet, serves as further evidence in support of the successful complexation of the the ligand to the M(II) ions.

**Table 6:** Conductivity of the salophen ligand (L) and its M(II) complexes

| Compound | Concentration ($Moldm^{-3}$) | Specific Conductance ($Ohm^{-1}\ cm^{-1}$) | Molar Conductance ($Ohm^{-1}\ cm^{2}\ mol^{-1}$) |
|---|---|---|---|
| $[MnLCl_2].2H_2O$ | $1.0\times10^{-3}$ | $17.12\times10^{-6}$ | 17.12 |
| $[FeLCl_2].2H_2O$ | $1.0\times10^{-3}$ | $16.02\times10^{-6}$ | 16.02 |
| $[CoLCl_2].2H_2O$ | $1.0\times10^{-3}$ | $39.30\times10^{-6}$ | 39.30 |
| $[NiLCl_2].3H_2O$ | $1.0\times10^{-3}$ | $55.40\times10^{-6}$ | 55.40 |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)

The molar conductance of the M(II) complexes were measured in DMSO at room temperature (Table 6). All the complexes are non-electrolyte based on the low molar conductance values of 16.02-55.40 $\Omega^{-1}cm^2mol^{-1}$ (Joshaghani *et al.* (2008)).

**Table 7:** Magnetic Susceptibility result of salophen ligand (L) and its M(II) complexes

| Compound | Xg($gmol^{-1}$) | Xm($gmol^{-1}$) | $\mu_{eff}$ (BM) | n | Magnetic Property |
|---|---|---|---|---|---|
| $[MnLCl_2].2H_2O$ | $3.04\times10^{-5}$ | $1.49\times10^{-2}$ | 5.97 | 5 | Paramagnetic |
| $[FeLCl_2].2H_2O$ | $2.21\times10^{-5}$ | $1.09\times10^{-2}$ | 5.09 | 4 | Paramagnetic |
| $[CoLCl_2].2H_2O$ | $1.34\times10^{-5}$ | $6.64\times10^{-3}$ | 3.98 | 3 | Paramagnetic |
| $[NiLCl_2].3H_2O$ | $8.83\times10^{-6}$ | $4.37\times10^{-3}$ | 3.23 | 2 | Paramagnetic |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)

The magnetic moment (B.M) values **(**Table 7**)** of the M(II) complexes determined at room temperature were found to be 5.97, 5.09, 3.98 and 3.23 B.M for the Mn(II), Fe(II), Co(II) and Ni(II) complexes, respectively. These values are consistent with high spin octahedral geometries showing five, four, three and two unpaired electrons, respectively (Ferenc *et al.,* (2020); Sunjuk, *et al.,* (2022)).

Based on the above characterization data, the following structures for the ligand and the M(II) complexes are proposed.

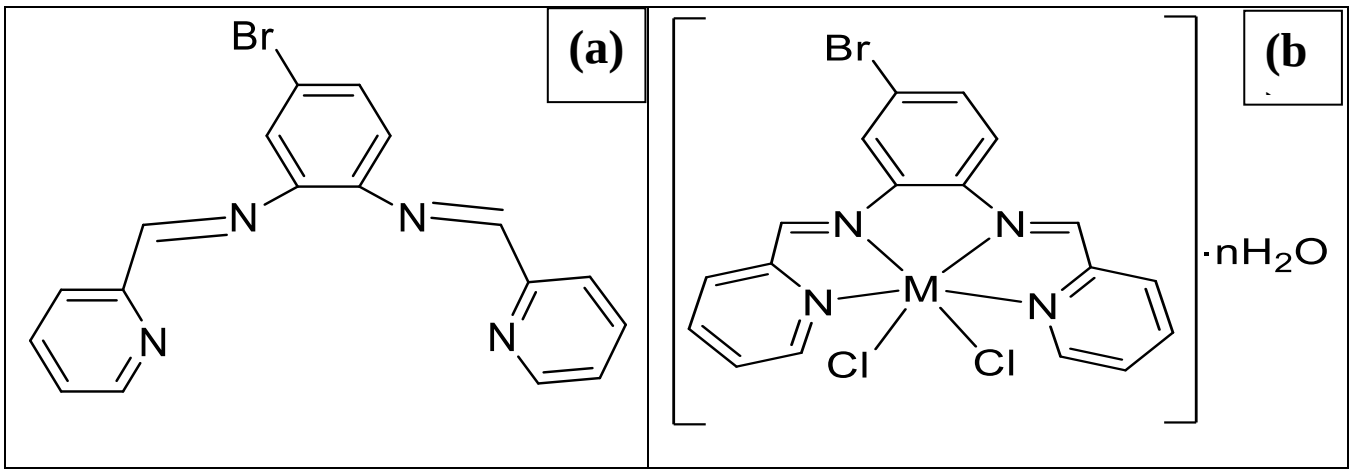



**Figure 1**: Proposed structure of (a) the salopen ligand (L) and (b) the M(II) complexes (M = Mn, Fe, Co or Ni)

**Antimicrobial Activity of the ligand and its M(II) complexes**

The *in-vitro* antimicrobial activity of the salophen ligand (L) and its M(II) complexes (subsequently referred to as the samples in this section) were tested against two gram positive (*Streptococcus pyogenes* and *Bacillus subtilis*) bacteria, two gram negative (*Salmonella typhi* and *Klebsiella pneumoniae*) bacteria and two fungal isolates (*Candida albicans* and *Aspergillus fumigatus*). Three concentrations of 1000, 500 and 250 µg/ml of the samples in DMSO were tested in triplicates and the mean value of diameter of growth inhibition zones (in mm) were recorded as antimicrobial activity of the samples (Sani *et al.*, 2017). Ciprofloxacin (for bacteria) and ketoconazole (for fungi) were employed as positive controls for comparison (Table 8).

**Table 8:** Antibacterial activity of the salophen ligand (L) and its M(II) Complexes

| **Compound** | **Zone of Inhibition (mm)** | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | ***S. pyogenes*** | | | ***B. subtilis*** | | | ***S. typhi*** | | | ***K. pneumoniae*** | | |
| | **1000** | **500** | **250** | **1000** | **500** | **250** | **1000** | **500** | **250** | **1000** | **500** | **250** |
| | **(µg/ml)** | | | **(µg/ml)** | | | **(µg/ml)** | | | **(µg/ml)** | | |
| Ligand (L) | 16 | 15 | 10 | 16 | 14 | 11 | 17 | 15 | 10 | 16 | 12 | 08 |
| $[MnLCl_2].2H_2O$ | 21 | 18 | 13 | 20 | 18 | 13 | 21 | 18 | 13 | 20 | 17 | 11 |
| $[FeLCl_2].2H_2O$ | 20 | 18 | 12 | 19 | 16 | 13 | 21 | 19 | 14 | 19 | 17 | 10 |
| $[CoLCl_2].2H_2O$ | **22** | **18** | **13** | **20** | **16** | **12** | **20** | **17** | **13** | **20** | **18** | **13** |
| $[NiLCl_2].3H_2O$ | 20 | 19 | 14 | 19 | 15 | 12 | 19 | 18 | 13 | 20 | 17 | 12 |
| Ciprofloxacin (500µg/ml) | | 30 | | | 28 | | | 25 | | | 31 | |
| Negative Control (DMSO) | | 00 | | | 00 | | | 00 | | | 00 | |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)

Good antibacterial activity was observed against all the tested isolates at high concentrations (1000 and 500 µg/ml) while moderate activity was observed at the lower concentration of 250 µg/ml. Overall, all the complexes showed great activity against all the tested organisms (*Streptococcus pyogenes*, *Bacillus subtilis*, *Salmonella typhi* and *Klebsiella pneumoniae)* at all concentrations (1000, 500 and 250 µg/ml). Generally, the mechanism of action of M(II) complexes is based on the ability of the metal to penetrate and cause some damage to the cell membrane of the bacteria or fungi (Evangelinou *et al.*, (2014); Arif *et al.*, (2018)). Other literature suggested generation of reactive oxygen species when the imine N of the ligand interact with the microbial cell components, and eventually damaging the microorganism (Arif *et al.*, (2018)).

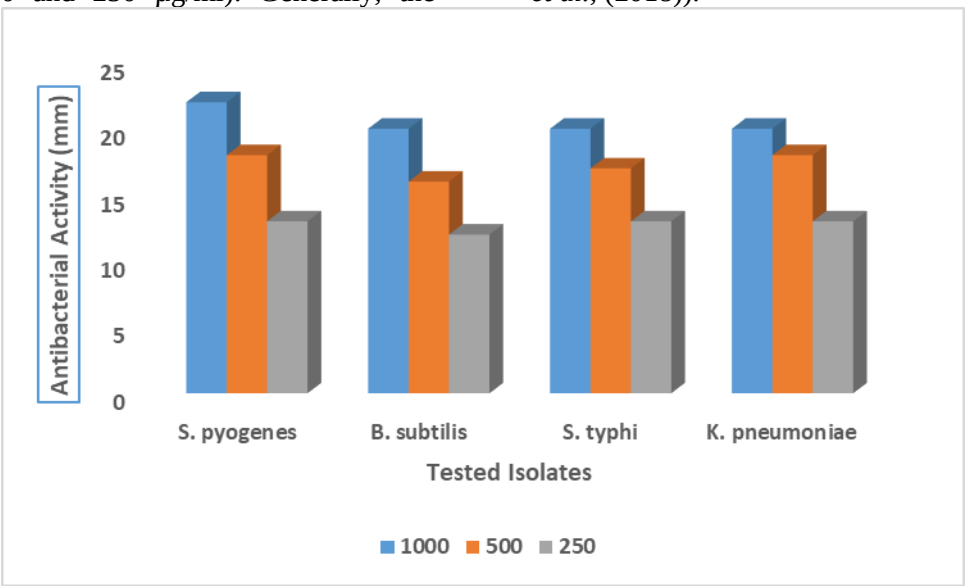



**Figure 2**: Graphical representation of the antibacterial activity trend for the Co(II) complex

All the samples show good antibacterial activity, with Co(II) complex performing comparably higher (Table 8). The activity observed over the tested samples can be explained through different perspectives. According to Sani and Dailami (2015), antimicrobial activity of complexes was explained on the basis of ancient rules; the Overtone's concept (Sani *et al.,* 2017), and the Tweedy's chelation theory (Tweedy, 1964; Mounika *et al.,* 2010). The Overtone's concept of cell permeability suggests that the lipid membrane that surrounds the cell favors the passage of only lipid-soluble materials due to which, lipophilicity is an important factor controlling the antimicrobial activity. Chelation theory postulates that the polarity of the metal ion will be reduced to a greater extent due to the overlap of the ligand orbital and partial sharing of the positive charge of the metal ion with donor groups, which increases the delocalisation of π-electrons over the whole chelate ring and enhances the lipophilicity of the metal complexes (Sani and Dailami, 2015). This increased lipophilicity enhance the penetration of the metal complexes into lipid membranes and thus blocking the various metabolic activities of microorganisms. Further, the higher antimicrobial activity observed for the metal complexes can be attributed to the involvement of the M(II) ions in disrupting the normal cell processes (Sani *et al.,* 2017).

**Table 9:** Antifungal activity of the ligand (L) and its M(II) complexes

| **Compound** | **Zone of Inhibition (mm)** | | | | | |
|---|---|---|---|---|---|---|
| | ***Candida albicans*** | | | ***Aspergillus fumigatus*** | | |
| | **1000** | **500** | **250** | **1000** | **500** | **250** |
| | | **(µg/ml)** | | | **(µg/ml)** | |
| Ligand ($L^b$) | 16 | 14 | 08 | 16 | 12 | 08 |
| $[MnLCl_2].2H_2O$ | 19 | 15 | 10 | 19 | 16 | 12 |
| $[FeLCl_2].2H_2O$ | 20 | 15 | 12 | 18 | 14 | 10 |
| $[CoLCl_2].2H_2O$ | 19 | 16 | 12 | 20 | 16 | 12 |
| $[NiLCl_2].3H_2O$ | 18 | 15 | 11 | 19 | 17 | 11 |
| Ketoconazole (Control) (500µg/ml) | | 29 | | | 26 | |
| Negative Control (DMSO) | | 00 | | | 00 | |

L = N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)

The antifungal activity of the salophen ligand (L) and the M(II) complexes (Table 9) revealed that the free ligand exhibited slightly lower antifungal activity compared to the M(II) complexes (Table 9). Interstingly, the Co(II) complex also showed the highest antifungal activity against *Candida albicans*, while the cobalt(II) complex was particularly more effective against *Aspergillus fumigatus*. Overall, all the samples showed good antibacterial and antifungal activity against the tested isolates, and, hence, this calls for further investigation and possible application of the ligand and the M(II) complexes in the future.

**CONCLUSION**

A new salophen ligand, (N,N'-(4-bromo-1,2-phenylene)bis(1-(pyridin-2-yl)methanimine)) and its metal(II) complexes (Mn, Fe, Co and Ni) were successfully synthesized and characterized using various spectroscopic and analytical techniques. The results suggested that the ligand is tetradentate and coordinated to the central metal ion through the imine and pyridine nitrogen. Molar conductance values showed that the complexes were non-electrolytic. The CHN elemental analysis lead to the proposed structures of both the salophen ligand and the M(II) complexes. Magnetic susceptibility and UV-vis spectra of all the M(II) complexes suggested octahedral geometries. All the M(II) complexes showed potential antimicrobial activities against the tested isolates. The complexes showed comparably higher antimicrobial potentials than the free salophen ligand under the same experimental conditions. Thus, further investigation of the compounds is desirable to

gain more insight into their crystal structures and biological activities.

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