

# SYNTHESIS, PHYSICOCHEMICAL AND ANTIMICROBIAL EVALUATION OF A TETRADENTATE SCHIFF BASE LIGAND WITH Hg(II), Pb(II), Mn(II), Cd(II), Fe(III) AND Cr(III) METAL COMPLEXES

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**Abstract:** Schiff base ligand derived from the condensation of 2-hydroxybenzaldehyde with ethylenediamine was synthesized and characterized by elemental analysis, M.pt, IR and <sup>1</sup>HNMR. The elemental analysis revealed a 1:2 molar ratio (amine:aldehyde) for the ligand. The complexes formed from the ligand were characterized by elemental analysis, M.pt, solubility, conductivity, IR and electronic spectra. The complexes showed various colours ranging from yellow, brown and golden. The ligand melted at 210°C whereas the complexes melted in the range 200-270°C. This high melting point indicates thermal stability of the compounds. The molar conductivities of the complexes (15.00-40.20 Scm<sup>2</sup>mol<sup>-1</sup>) indicates that they are non-electrolytes in the organic solvents used for the investigation. The comparison of the IR spectra of the Schiff-base and its metal complexes showed that the Schiff base acted as tetradentate ligand. The electronic spectral data indicated d-d transitions in the visible region in some of the complexes. Some metal-ligand charge transfer (MLCT) or vice versa were also observed around 28,600 cm<sup>-1</sup> in the complexes. Antimicrobial results revealed that [Hg(HL)Cl] complex appeared to have promising antimicrobial effect on *S. pyogenes* when compared to the other complexes.

**Key words:** Schiff base, 2-hydroxybenzaldehyde, ethylenediamine, characterization, antimicrobial activity.

## INTRODUCTION

Coordination chemistry has remained significantly enhanced due to the synthesis and characterization of an enormous number of ligands and their transition metal complexes in which the metal is coordinated by sulfur, nitrogen and oxygen. Transition metal complexes has been the subject of interest, because of the complexities related with many variable factors as oxidation states, variety of geometries around the metal ions, donating properties of ligands and their influence on spectral, catalytic and biological properties of metal ion. [1].

The condensation of a diamine derivative with Salicylaldehyde leads to the formation of a Schiff base which possesses a tetradentate structure, indicating that two nitrogen atoms and two oxygen atoms are available for chelation. The ligands known as Salen ligand are analogous to the porphyrin in structural aspects but can be easily prepared [2-3]. Schiff bases of Salicylaldehyde and ethylenediamine act as tetradentate chelating ligands with complexes of rigid structures [4]. Most of the metal complexes are in mole ratio 1:1 (metal:ligand) [5]. The interaction of those donor ligands and metal ions gave complexes of different geometries and these complexes are potentially active biologically [6]. Complexes of some divalent metal ions with Schiff bases derived from Salicylaldehyde and tyrosine have been prepared and investigated by using spectroscopic methods. Results indicated an octahedral structure for the complexes [7-9]. Schiff bases and their complexes have been reported to have many applications in the literature because of their broad variety of biological activities, such as anticancer, antimalarial, antimicrobial, antitubercular, and anti-inflammatory [10]. In view of the numerous applications of the Schiff base ligands and their complexes, the present investigation aims to prepare, characterize and determine the antimicrobial activities of a Schiff base ligand derived from ethylenediamine and 2-hydroxybenzaldehyde and their metal(II) complexes.

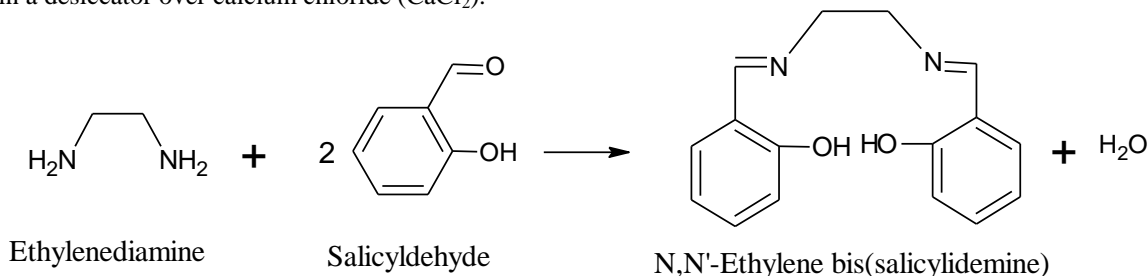
## MATERIALS AND METHODS

All the chemicals and solvents used were of Analar grade and were used without further purification. The metal salts of Mn(II), Hg(II), Pb(II) and Fe(III) were used as chlorides while Cd(II) and Cr(III) salts were used as acetates. Thermo Flash EA CHNS-O elemental analyzer, Griffin melting point apparatus, Fourier Transform Infrared Spectroscopy, UV/Visible Spectrophotometer Model 721, <sup>1</sup>HNMR Bruker/Top Spin 3.2T-D-65536 spectrophotometer and Estick(R) Series

Model EC 500/Conductivity and Temperature Meter were used for the study. Antibacterial activities of the ligand and metal complexes were screened against two strains of Gram positive bacteria (*Staphylococcus aureus*, *Streptococcus pyogenes*) and two Gram negative (*Escherichia coli*, *Klebsiella pneumoniae*) and two fungi (*Aspergillus niger* and *Candida albicans*) using filter paper disc agar diffusion method.

### Synthesis of the Ligand

The ligand was prepared according to literature procedures [11-13]. This was done by the condensation of substituted aromatic aldehydes and the corresponding amines in 1:2 (amine: aldehyde) molar ratio in ethanol (Scheme 1). A 30 ml ethanolic solution of 2-hydroxybenzaldehyde (20 mmol, 2.44 g) was mixed with stirring with 30 ml ethanolic solution of ethylenediamine (10 mmol, 0.601g) after which 2-3 drops of conc. H<sub>2</sub>SO<sub>4</sub> was added. The mixture was refluxed for 3hrs in a quick fit conical flask. After refluxing, the mixture was left to stand for 2-3 days. The solid product obtained was filtered, washed with ethanol and dried in a desiccator over calcium chloride (CaCl<sub>2</sub>).



Scheme: 1.

Synthesis of the Schiff base ligand

### Synthesis of the Metal Complexes

The metal complexes were prepared according to literature procedures [11-13]. Metal salts of Mn(II), Cd(II), Hg(II), Pb(II) Cr(III) and Fe(III) were added to the Schiff base ligand in a mole ratio 1:1 (metal: ligand). A mixture of the Schiff base (HL) under investigation (1 mmol, 0.268g) in 30 ml ethanolic solution and 30 ml ethanolic solution of Mn(II) salt (1 mmol, 0.1979g) were refluxed for 2 hours, after which it was cooled, filtered and the product obtained were dried in a desiccator containing calcium chloride (CaCl<sub>2</sub>). The masses of the products obtained were determined by weighing after drying and percentage (%) yields calculated.

## RESULTS

Table 1: Physical Properties and some Analytical Data of the Schiff Base Ligand and its Metal Complexes

S/No	Compound	Colour	Yield, g(%)	M.P. (°C)	$\Lambda_m$ (Scm <sup>2</sup> mol <sup>-1</sup> )
1	HL	Yellow	2.89 (54)	210	-
2	[Hg(HL)Cl]	Golden	0.54 (54)	290(d)	18.00
3	[Pb(HL) Cl]	Golden	0.57 (56)	235	17.50
4	[Mn(HL)Cl]	Brown	0.326 (91)	200	40.20
5	[Fe(HL)Cl]	Brown	0.268 (74)	230 -232	18.10
6	[Cd(HL)]	Yellow	0.195 (51)	270	15.00
7	[Cr(HL)]	Brown	0.236 (74)	215	18.90

All the compounds are air-stable and were formed in good yields with various colours ranging from yellow, brown and golden. The compounds showed high melting point which indicates that they are stable. However, Hg(HL)Cl decomposed before melting. The ligand is very soluble in most of the solvents used. The complexes were also soluble or slightly soluble in the solvents used. The molar conductivity of the complexes indicated that they are non-electrolytes.

Table 2: Microanalysis of the Schiff Base Ligand and its Metal Complexes

S/No.	Compounds	Molecular Formula (Molar Mass)	Microanalysis: Found (calculated) %			
			C	H	N	M
1	HL	C <sub>16</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> (268.34)	71.23 (71.55)	5.96 (5.96)	10.51 (10.43)	-
2	[Hg(HL)Cl]	Hg(C <sub>16</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> )Cl	38.30	3.36	5.44	40.20

		(504.84)	(38.03)	(3.16)	(5.54)	(39.81)
3	[Pb(HL) Cl]	Pb(C <sub>16</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> )Cl (510.84)	37.52 (37.58)	3.67 (3.13)	5.05 (5.48)	40.57 (40.52)
4	[Mn(HL)Cl]	Mn(C <sub>16</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> )Cl (325.84)	53.19 (53.50)	3.75 (3.94)	4.02 (4.24)	15.33 (15.40)
5	[Fe(HL)Cl]	Fe(C <sub>16</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> )Cl (359.84)	53.19 (53.35)	5.16 (4.44)	7.20 (7.78)	15.56 (15.90)
6	[Cd(HL)]	CdC <sub>16</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> (380.84)	50.80 (50.41)	4.88 (4.20)	7.10 (7.35)	29.54 (29.25)
7	[Cr(HL)]	Cr(C <sub>16</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub> ) (418.44)	45.40 (45.88)	4.07 (4.20)	7.10 (7.35)	16.23 (16.43)

The Microanalysis of the ligand and the metal complexes are presented in Table 2. Microanalysis (C, H, N) results indicated that the experimental data are in close agreement with the theoretical values in most cases and agrees with the formation of 1:1 (M:L) molar ratio for the complexes.

Table 3: The Relevant Infrared Frequencies (cm<sup>-1</sup>) for the Schiff Base Ligand and its Metal Complexes

S/No	Compounds	v(OH)	v(C=N)	v(C-O)	v(C=C)	M-O	M-N
1	HL	3402s	1635m	1411w	1558w	-	-
2	[Hg(L) Cl]	3406s	1639m	1346b	1411w	856b	509w
3	[Pb(HL) Cl]	3402s	1639m	1492m	1546w	744m	489w
4	[Mn(HL)Cl]	3410s	1620b	1141b	1481b	597w	516w
5	[Fe(HL)Cl]	3402s	1639m	1350b	1411m	821b	489b
6	[Cd(HL)]	3406s	1635m	1485b	1570w	744m	536w
7	[Cr(HL)]	3406b	1639b	1315b	1531b	740w	435b

### IR SPECTRA

The band observed at 3402 cm<sup>-1</sup> in the free ligand was assigned to vO-H band which shifted to higher frequency in the spectra of the metal complexes in the range of 3406-3410 cm<sup>-1</sup>. Previous assignments of 2-(2-(2-hydroxycyclohexa-1,3-dien-1-yl)methylene)aminoethyl)imino)methyl)phenol derivatives have placed the O-H group in the range of 3360-3580 cm<sup>-1</sup> [13]. The azomethine V(C = N) band in the spectrum of the ligand appeared at 1635 cm<sup>-1</sup> as a sharp band. This is similar to the reports by [14] who assigned the v(C = N) stretching vibration in 2-hydroxybenzaldehyde with ethylenediamine at 1636 cm<sup>-1</sup> as a sharp band. On coordination with the metal complexes, this band shifted to higher frequency at 1639 cm<sup>-1</sup> in Hg(II), Pb(II), Fe(III) and Cr(III) complexes indicating that the ligand is coordinated to the metal ion through azomethine nitrogen. The differences in the intensities of these bands may be due to the nature of the metal ions [15].

The phenolic C - O appeared at 1411 cm<sup>-1</sup>, this reflects the higher acidity of OH group in HL ligand. Hg(II), Mn(II), Fe(III) and Cr(III) complexes shifted to lower frequency in the range of 1315-1350 cm<sup>-1</sup> on complexation. This observation is in agreement with similar assignments in the literature [16-18]. Pb(II) and Cd(II) shifted to higher frequency at 1492 and 1485 cm<sup>-1</sup> due to participation of oxygen in formation of the C - O - M bond. In the spectrum of the ligand, the (C = C) ring appeared at 1558 cm<sup>-1</sup>. In the spectra of the complexes this band shifted to lower frequency in the range, 1411 - 1550 cm<sup>-1</sup>, except in the spectra of Cd(HL) complex which shifted to higher frequency at 1570 cm<sup>-1</sup> as a weak band. Similar observations reported by [12] assigned C = C at about 1554 cm<sup>-1</sup> for the Cd(II) complex. New bands observed in the lower frequency region in the range of 821 and 613 cm<sup>-1</sup> attributed to (M -O) and (M - N). This indicated that coordination occurred between the ligand and the metal ion through the phenolic oxygen and azomethine nitrogen atom.

Table 4: Electronic Spectra of the Schiff Base Ligand and its Metal Complexes

S/N	Compound	λ <sub>max</sub> nm	λ <sub>max</sub> cm <sup>-1</sup>	Assignment
1	HL	380 390	26315 25641	n - π* n - π*
2	[Hg(HL)Cl]	400	25000	MLCT
3	[Pb(HL)Cl]	390 420	25641 23809	MLCT
4	[Mn(HL)Cl]	430	23255	<sup>6</sup> A <sub>1g</sub> → <sup>6</sup> T <sub>2g</sub> (G)
5	[Fe(HL)Cl]	350 500	28571 20000	MLCT <sup>5</sup> T <sub>2g</sub> (F) → <sup>5</sup> E <sub>g</sub>

6	[Cd(HL)]	410	24390	MLCT
7	[Cr(HL)]	390	25641	MLCT
		440	22727	MLCT
		540	18518	${}^4A_2(F) \rightarrow {}^4T_{1g}(F)$

### ELECTRONIC SPECTRA

The electronic spectra of HL showed absorption band at 26315  $\text{cm}^{-1}$  assigned to  $\pi - \pi^*$  of the aromatic system of benzene ring. The OH phenolic group showed  $n - \pi^*$  absorption at 25641  $\text{cm}^{-1}$  and this absorption interfered with the  $\pi - \pi^*$  absorption resulting in bathochromic or red shift towards the lower wavelength [13]. The  $\pi - \pi^*$  type transition due to C=N chromophore shifted to lower energies on complexation. The electronic spectra of Hg(HL)Cl showed strong absorption band at 25000  $\text{cm}^{-1}$  due to charge transfer transition. Lead (II) complex had absorption at 25641  $\text{cm}^{-1}$  due to charge transfer transition from ligand to metal or metal to ligand. Mn(HL)Cl showed absorption band in the visible region at 23255  $\text{cm}^{-1}$  assigned to  ${}^6A_{1g} \rightarrow T_2(G)$  transitions. The electronic spectra of Fe(III) complex displayed a band around 28571  $\text{cm}^{-1}$  which is due to charge transfer transitions, while the broad band in the range 20000  $\text{cm}^{-1}$  and 20408  $\text{cm}^{-1}$  are assigned to  ${}^5T_{2g}(F) \rightarrow {}^5E_g$  transitions respectively. A strong band occurring at 24390  $\text{cm}^{-1}$  is due to ligand to metal charge transfer transitions. The electronic spectra of the Cr(III) complex compared with the spectra of the corresponding free ligand is associated with the appearance of a broad low intensity band in the range of 22727-18518  $\text{cm}^{-1}$ . This band is attributed to the d-d transition of the chromium ion and is assigned to  ${}^4A_2(F) \rightarrow {}^4T_{2g}(F)$  transitions. The appearance of a strong band at 25641  $\text{cm}^{-1}$  was assigned to ligand - metal charge transfer. These charge transfer transitions probably occurred from the  $\pi$  - orbital of the ligand to the d - orbital of chromium ion.

Table 5:  ${}^1\text{H}$  Nuclear Magnetic Resonance Data of the Ligand in (ppm)

Compounds	Molecular Formula (Molar Mass)	Phenolic proton $\delta$ (OH)	Azomethine $\delta$ (-HC=N)	Aromatic proton $\delta$ (C - H)	Solvent (DMSO)
HL	$\text{C}_{16}\text{H}_{16}\text{N}_2\text{O}_2$ (268.34)	13.38	8.60	6.88 - 7.43	2.51 - 3.93

The hydrogen proton NMR of 2-hydroxybenzaldehyde with ethylenediamine (HL) ligand showed the following characteristic chemical shifts. The singlet signal at  $\delta$ 13.38 ppm is characteristics of proton of the O-H group. This is in close agreement with reports by [19]; [14]. The hydroxy proton appears as a singlet at 13.2 ppm of a similar Schiff base ligand. The singlet signal at  $\delta$ 8.60 ppm suggested the attribution of the azomethine proton (-HC=N) group. A multiplet signals in the range of 6.88-7.43 ppm was assigned to aromatic proton (C-H). [20] also observed peaks in the range of 6.8-7.6 ppm due to aromatic hydrogen of a similar ligand. The DMSO proton showed a multiplet signals in the range of  $\delta$ 2.51-3.93 ppm respectively.

Table 6: Antimicrobial Activities of the Schiff Base Ligand (HL) and its metal Complexes

Compounds	Concentration (mg/ml)	Antimicrobial activity with zone of inhibition (mm)					
		<i>Staphylococcus aureus</i>	<i>Streptococcus Pyogenes</i>	<i>Escherichia coli</i>	<i>Klebsiella pneumonia</i>	<i>Candida albicans</i>	<i>Aspergillus niger</i>
HL	400	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	300	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	200	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	100	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
[Hg(HL)Cl]	400	24.67±0.58 <sup>b</sup>	27.67±0.58 <sup>b</sup>	18.0±0.00 <sup>b</sup>	20.0±0.00 <sup>b</sup>	31.67±0.58 <sup>b</sup>	17.0±0.00 <sup>b</sup>
	300	20.33±0.58 <sup>c</sup>	22.67±0.58 <sup>c</sup>	13.33±0.58 <sup>c</sup>	15.33±0.58 <sup>c</sup>	27.67±0.58 <sup>c</sup>	12.33±0.58 <sup>c</sup>
	200	16.67±0.58 <sup>d</sup>	19.0±0.00 <sup>d</sup>	9.67±0.00 <sup>d</sup>	11.67±0.58 <sup>d</sup>	24.0±0.00 <sup>d</sup>	9.67±0.58 <sup>d</sup>
	100	12.0±0.00 <sup>e</sup>	14.33±0.58 <sup>e</sup>	7.0±0.00 <sup>e</sup>	8.67±0.00 <sup>e</sup>	19.67±0.00 <sup>e</sup>	7.33±0.58 <sup>e</sup>
[Pb(HL)Cl]	400	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	300	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	200	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	100	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
[Mn(HL)Cl]	400	23.0±0.00 <sup>f</sup>	24.00±0.00 <sup>f</sup>	22.0±0.00 <sup>f</sup>	15.0±0.00 <sup>f</sup>	26.67±0.58 <sup>f</sup>	16.0±0.00 <sup>f</sup>
	300	18.0±0.00 <sup>g</sup>	20.00±0.00 <sup>g</sup>	18.0±0.00 <sup>g</sup>	11.0±0.00 <sup>g</sup>	22.0±0.00 <sup>g</sup>	12.0±0.00 <sup>g</sup>
	200	18.67±0.58 <sup>h</sup>	15.67±0.58 <sup>h</sup>	14.33±.58 <sup>h</sup>	9.0±0.00 <sup>h</sup>	18.0±0.00 <sup>h</sup>	9.0±0.00 <sup>h</sup>

	100	10.67±0.58 <sup>l</sup>	11.67±0.58 <sup>l</sup>	10.33±0.58 <sup>l</sup>	7.0±0.00 <sup>l</sup>	14.0±0.00 <sup>l</sup>	7.0±0.00 <sup>l</sup>
[Fe(HL)Cl]	400	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	300	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	200	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	100	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
[Cd(HL)]	400	10.67±0.58 <sup>l</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	17.0±0.00 <sup>j</sup>	14.0±0.00 <sup>j</sup>
	300	8.0±0.00 <sup>j</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	13.33±0.00 <sup>k</sup>	11.0±0.00 <sup>k</sup>
	200	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	10.0±0.00 <sup>l</sup>	7.67±0.58 <sup>l</sup>
	100	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>	7.0±0.00 <sup>m</sup>	0.00±0.00 <sup>a</sup>
[Cr(HL)]	400	16.67±0.58 <sup>d</sup>	17.0±0.00 <sup>j</sup>	24.0±0.00 <sup>j</sup>	17.0±0.00 <sup>j</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	300	13.67±0.58 <sup>k</sup>	13.33±0.58 <sup>k</sup>	20.0±0.00 <sup>k</sup>	13.0±0.00 <sup>k</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	200	10.33±0.58 <sup>l</sup>	10.0±0.00 <sup>l</sup>	16.33±0.58 <sup>l</sup>	9.33±0.58 <sup>l</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
	100	7.33±0.58 <sup>m</sup>	7.0±0.00 <sup>m</sup>	12.0±0.00 <sup>m</sup>	7.0±0.00 <sup>l</sup>	0.00±0.00 <sup>a</sup>	0.00±0.00 <sup>a</sup>
Cipro	400	20.67±0.58 <sup>n</sup>	35.00±0.00 <sup>n</sup>	18.00±0.00 <sup>n</sup>	17.33±0.58 <sup>m</sup>	-	-
Amph. B		-	-	-	-	22.33±0.58 <sup>n</sup>	32.00±0.00 <sup>m</sup>

Different superscript letters along the same column are significantly (P<0.05) different.

The Schiff base ligand derived from 2-hydroxybenzaldehyde with ethylenediamine was not susceptible against all the tested microorganisms. Hg(HL)Cl complex showed high activity on *S. aureus* at 400 mg/ml (24.67±0.58 mm), the activity observed is higher than the standard drug (ciprofloxacin 20.67±0.58 mm). At 400 mg/ml (18.0±0.00 mm), Hg(HL)Cl complex displayed equal inhibition zone with the standard drug (18.0±0.00 mm) on *E. coli*. The complex was also susceptible on *K. pneumoniae* at 400 mg/ml (20.0±0.00 mm) more than the standard drug (17.33±0.58 mm). This results indicated that Hg(HL)Cl complex has stronger activity. Pb(HL)Cl complex showed no activity against all the tested microorganisms at all concentrations. Mn(II) complex showed activity against all the tested organisms at all concentrations. This is in close agreement with results reported by [21] who observed that the activity of Mn(II) complex showed better efficiency against same bacterial species. At 400 mg/ml the complex showed a high inhibition zones on both Gram positive, *S. aureus* (23.0±0.00 mm) and Gram negative, *E. coli* (22.0±0.00 mm) organisms more than the standard drug (20.67±0.58, 18.0±0.00 mm). The fungal specie *C. albicans* at 400 mg/ml (31.67±0.58 mm) showed more superior activity as compared with the standard drug (Amph. B 22.33±0.58 mm). The results indicated that at a high concentration, the complex is more susceptible than at a low concentration.

Cd(II) complex showed activity against *S. aureus*, *C. albicans* and *A. niger*. High activity displayed on *C. albicans* at all concentrations. The Cd(II) complex was more susceptible on fungal specie than the bacteria while chromium complex was effective against bacteria, but on fungi they showed no activity at all concentrations. The Cr(III) complex exhibited activity against all the four bacterial isolates comprising both Gram positive and Gram negative organisms. At 400 mg/ml (24.0±0.00 mm) and 300 mg/ml (20.0±0.00 mm), high zones of inhibition were observed on *E. coli* when compared to the standard drug (18.0±0.00 mm). Such enhanced activity of metal chelates is due to the lipophilic nature of the metal ions in the complexes. The increase in activity with concentration is due to the effect of metal ions on the normal metabolic function of the cell. The action of the compounds may involve the formation of azomethine bonds with the active centre of the cell constituents, resulting in interference with the normal function of the cell (Achut *et al.*, 2010).

## CONCLUSION

All the compounds are air-stable and were formed in good yields with various colours. The compounds showed high melting point which indicates that they are stable. The molar conductivity of the complexes indicated that they are non-electrolytes. From the IR results, the Schiff base acted as tetradentate ligand and coordinated with the metal ion through the phenolic oxygen and azomethine nitrogen atoms. The electronic spectral measurements of the complexes in methanol suggested the complexes to be five coordinated. The antimicrobial results revealed that Hg(HL)Cl complexes appeared to have promising antimicrobial effect on the *S. pyogene* when compared to the other complexes. The antifungal activity of the metal complexes also revealed that Hg(HL)Cl complex showed the highest activity on *C. albicans*, followed by Mn(HL)Cl. Based on the results obtained, the following structure was proposed for the complexes.

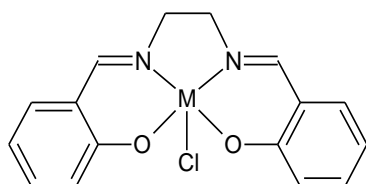


Figure 1: The proposed structure of the complexes  
M=Mn(II), Cd(II), Hg(II), Pd(II), Fe(III) and Cr(III)

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## REFERENCES

1. Hanaa A. E., Azza A. S. and Ahmed A. S. (2019). Synthesis, Spectral Characterization, Thermal and In vitro Antimicrobial Studies for Novel Co(II) and Ni(II) Complexes of (N,N'-(1,2-Phenylene)Bis(2-Aminobenzamide). *Journal of Advances in chemistry*. 16: 5360 – 5374.
2. Larry, Y. (2003). Chemistry and Biology of Salicylideneimine and Related Compounds. *Chemical Acta.*, **103**: 4283-4306.
3. Neelima, M., Kavitan, P. and Dinesh, K. (2013). An overview of biological aspects of Schiff base metal complexes. *International Journal of Advanced Research and Technology*. **2**(8):52-56.
4. Reedijk, J. (1987). Heterocyclic Nitrogen - Donor Ligands A: *Comprehensive Coordination Chemistry: The Synthesis, Reactions, Properties and Application of Coordination Compounds*. Eds.; Pergamon Press; Oxford. pp.1324-1232.
5. Shakir, M., Azam, M., Parveen, S., Khan, A. U. and Firdaus, F. (2009). Synthesis and spectroscopic studies on complexes of N,N-bis(2-pyridinecarboxaldimine)-1,8-diaminonaphthalene (L); DNA binding studies on Cu(II) complexes, *Spectro chemical Acta, Part A*, **71**:1851-1856.
6. Raman, N., Ravichandran, S. and Thangaraja, C. (2004). Copper(II), cobalt(II), nickel(II) and zinc(II) complexes of Schiff base derived from benzyl-2,4-dinitrophenylhydrazone with aniline., *Journal of Chemical Sciences.*, **116**:215-225.
7. Anacona, J. R. and Gladys, D. S. (2005). Synthesis and antibacterial activity of cefotaxime metal complexes. *Journal of Chilly Chemical Society*. **50**(2):447 - 450.
8. Rajib, L. D., Mahnya, M., Lovely, R. and Jay, D. M. (2008). Synthesis, spectroscopic studies, crystal structure and complexation reaction of N-(2 or 4 hydrocylphenyl) benzaldimine. *Indian Journal of Chemistry*. **47**:207 - 273.
9. Yoshikawa, Y., Ueda, E., Kawabe, K., Miyake, H., Takino, T., Sakurai, H. and Kojima, Y. (2012). Development of new insulinomimetic Zinc(II) picolinate complexes with a Zn(N<sub>2</sub>O<sub>2</sub>) coordination mode: structure characterization, *in-vitro*, and *in-vivo* studies., *Journal of Inorganic Biochemistry.*, **88** (68): 91-99.
10. Maihub A. A., Sofian S. M, Awin A. A., Belaid A. K., Bensaber S. M., Hermann, A. and Abdul M. G. (2018). Antimicrobial activity of some pyrazolidin-3-one Schiff base derivatives and their complexes with selected metal ions. *Journal of Pharmacology and Clinical Trials*. **1**: (1), 55-64.
11. Achut, S. M., Amarnath, N. J., Sarika, M. J. and Trimbak, K. C. (2010). Synthesis, characterization and thermal study of some transition metal complexes of an assymetrical tetradentate Schiff base ligand. *Journal of Serbian Chemical Society*, **75**(3):349-459.
12. Nevin, T. and Memet, S. (2009). Synthesis and Chacterization of Co(II), Ni(II), Cd(II) and Cu(II) complexes of Bis-Schiff Bases obtained from 1,8-Diaminonaphthalene., *Journal of Chemical Society.*, **31**(4):564-567
13. Tawfiq, A. A. (2011). Synthesis and Characterization of Some Divalent Transition Metal Complexes of Schiff Bases Derived from Salicylaldehyde Diamine Derivatives., *Al-Mustansiriyah Journal of Science.*, **21** (1): 104-113.
14. Rafeye, R. and Anita, A. (2015). Synthesis and Characterization of New Schiff Bases of Ethylenediamine and Benzaldehyde Derivatives, along with their Iron Complexes., *Journal of Applied Chemical Research.*, **9** (2): 59-65.
15. Hamil, A. M. Abdelkarem, M. Hemmet M. and El-ajaily M. M. (2012). Synthesis of a New Schiff Base: 2-[2-(E)-(2-hydroxyphenyl)-ethylidene]aminoethyl) ethanimidoyl] phen., *International Journal of Chemical Technology and Research.*, **4**(2):682-685.
16. Andrzej, O., Damain, R., Jaroslaw, C. and Barbara, B. (2015). Coordination polymers and molecular structures among complexes of mercury (II) halides with selected 1-benzoylthioureas., *Polyhedron* **90** : 47-57.
17. Sunil, J., Vatsala, P. and Uma, V. (2010). Antibacterial and Antioxidant properties of Mn(II), Co(II), Ni(II) and ZN(II) Complexes of Schiff base derived from Cephalexin. *Research Journal of Pharmaceutical, Biological and Chemical Sciences*. **2**(1):61-70.
18. Mostafa, M. H. K., Eman, H. I., Gehad, G. M., Ehab, M. Z. and Ahmed, B. (2012). Synthesis and characterization of a novel Schiff base complexes and their application in determination of iron in different types of natural water. *Open Journal of Inorganic Chemistry*. **2**:13-21.
19. Ahmad, F. A. B., Hadariah, B., Karimah, K. and Mazatulikhma, M. Z. (2011). Synthesis, Characterization and Neurotoxic Effect of Schiff Base Ligand and their Complexes. *The Malaysian Journal of Analytical Science.*, **15**(1): 93-100.
20. Pedro, E. A., Mirian, P. S., Sandra, R. and Edward, R. D. (2006). Synthesis, Characterization, and Spectroscopic Studies of Tetradentate Schiff Base Chromium (III) Complex., *Polyhedron*, doi: 10.1016/ Journal of polyhedron, pp.11005.
21. Voguri, H. B., Venkateswararao, K., Podisetty, H., Badde, S. and More, A. (2017). Binuclear Manganase(II) complexes of a new Schiff base as ligand: Synthesis, Structural Characterization, and antibacterial Activity., *International Journal of Chemical and Technology Research.*, **10** (5):136-143.