



SYNTHESIS, CHARACTERIZATION AND ANTIMICROBIAL SCREENING OF METAL (II) COMPLEXES DERIVED FROM BENZOFURAN -2-CARBOHYDRAZIDE AND ACETOPHENONE SCHIFF BASE.

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ABSTRACT

The Cu(II), Co(II), Ni(II), Zn(II), Cd(II) and Hg(II) complexes of the Schiff base derived from reaction between benzofuran-2-carbohydrazide and acetophenone have been synthesized. The synthesized complexes have been characterized on the basis of elemental analysis, conductivity data, magnetic properties, IR and ¹H NMR spectral studies. The spectral study shows that Schiff base act as bidentate by coordinating through the azomethine nitrogen and oxygen of the carbonyl groups. Based on these results we have proposed the tentative structure for all of these complexes. The antifungal and antibacterial activities of the ligand and their metal complexes have been screened against fungi *Aspergillus niger* and *Candida albicans* and against bacteria *Escherichia coli* and *Staphylococcus aureus*.

KEYWORDS: Schiff's base, Metal complexes, Spectral studies, Antimicrobial activity.



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INTRODUCTION

Schiff base and its metal complexes have a variety of application in biological and pharmacological areas^{1, 2}. Schiff bases are characterized by the -HC=N-(imine) group. Schiff base and metal complexes play an important role in various biological activities³⁻⁵. Schiff base have wide application in food industry, dye industry, analytical chemistry, catalysis, fungicidal, agrochemical and biological activities⁶⁻⁸. Benzofuran compounds are abundantly present in nature, particularly among the plant kingdom, some of the benzofuran derivatives like 2-nitrobenzofuran, 2-acetylbenzofuran are well known biodynamic agents possessing various pharmacological properties⁹. Benzofuran compounds occur in nature in a variety of structural forms. The seed oil of plant "Egonoki" which is known to contains a benzofuran derivative called "Egonal" and its effective synergist for rotenone pyrethrum against house flies, mosquitoes, aphids and many other insects. Baker's yeast contains a benzofuran derivative which acts as an antioxidant preventing hemorrhagic liver necrosis in rates and hemolysis of red cells in Vitamine-E deficiency. From these points of view, it is important to study different types of transition metal complexes of these biological active ligands. In this paper we synthesis the benzofuran Schiff base derived from the reaction between benzofuran -2-carbohydrazide and acetophenone. Synthesized complexes were characterized on the basis of elemental analysis, molar conductivity, IR, ¹H NMR, electronic spectral studies and magnetic susceptibility measurements. Schiff's base and its metal complexes are screened for their biological activity studies.

MATERIAL AND METHODS

All the chemicals used were of A.R grade. The compound benzofuran-2-carbohydrazide was prepared by the literature method¹⁰. The metal and chloride

contents are determined as per Vogel's procedure¹¹. Elemental analyses were performed on a Perkin-Elmer 240C model at the central drug research institute (CDRI), Lucknow. The IR spectra of the Schiff base and its Cu(II), Co(II), Ni(II), Zn(II), Cd(II), and Hg(II) complexes were recorded in KBr pellets in the region of 4000-350 cm⁻¹ on a perkin-Elmer FT-IR Spectrophotometer. The ¹H NMR spectra were recorded in DMSO-d₆ on Bruker 400 MHz spectrophotometer, using TMS as internal standard. The electronic spectra were recorded on Elico SL - 159 double beam UV-Vis spectrophotometer in the range of 200-1100 nm in DMF (10⁻³M) solution. The Mass spectra were recorded on an Electrospray ionization Mass spectrophotometer at the Indian institute of Technology, Kanpur. Conductance measurements were made on Elico CM- 180 Conductivity bridge in DMF (10⁻³M) solution using a dip-type conductivity cell fitted with a platinum electrode having cell constant 0.1 ohm⁻¹cm²mole⁻¹. The magnetic susceptibility measurement was made on Gouybalance using Hg [Co(NCS)₄] as the calibrant at room temperature. The ESR spectra of Cu (II) complex in polycrystalline state was recorded on Varian E-4X band spectrophotometer using (TCNE) as a 'g' marker (g = 2.00277) at room temperature.

Synthesis of Schiff base: (E)-N'-(1-phenylethylidene)benzofuran-2-carbohydrazide (BCACP).

Schiff base was prepared by condensing benzofuran-2-carbohydrazide (1.76g, 0.01mol) and acetophenone (1.17ml, 0.01mol) in methanol (25 mL). The reaction mixture was refluxed on a water bath for about 4-5 hrs. The Schiff base was separated on removal of 50% of the solvent and cooling to room temperature. The product was filtered out and washed with ethanol and recrystallised from ethanol. The Schiff base was dried in vacuum over the anhydrous CaCl₂. The purity of the Schiff base was checked by TLC.

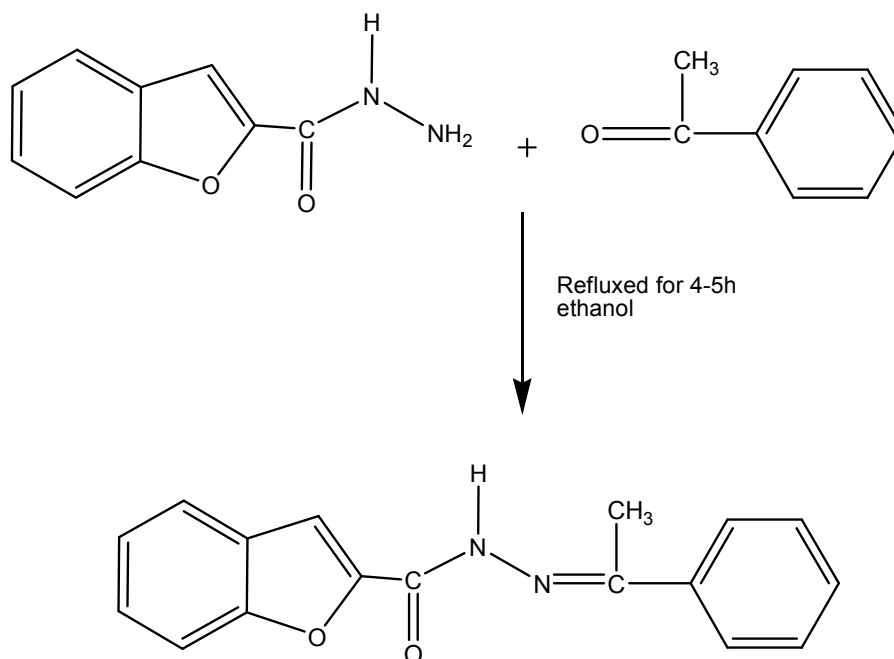


Figure. 1
Synthesis of Schiff base (BCACP)

Mol. Formula: $C_{17}H_{14}N_2O_2$ [BCACP], **Mol. Wt** = 278, **m. p** = 177 °C, **Yield** = 75%

Synthesis of Metal (II) complexes:

Metal complexes were prepared by the mixing of (50 mL) ethanolic solution of metal (II) chlorides with the (50 mL) ethanolic solution of Schiff base in 1:1 (metal: ligand) ratio. The resulting mixture was refluxed on water bath for about 2-3 hours. A light colored product appeared on standing and cooling the above solution. The precipitated complexes were filtered washed with water and recrystallized with ethanol several times and dried under the reduced pressure over anhydrous $CaCl_2$ in desiccator.

Antibacterial and antifungal assays:

The ligand and complexes were screened for their antibacterial and antifungal activity by agar cup plate zone of inhibition technique^{12, 13} against two bacteria *E. coli*, and *S. aureus* and two fungal species *A. niger* and *C. albicans*.

Antibacterial screening using agar-cup plate method:

Peptone 10 g, NaCl 10 g, Yeast extract 5 g, Agar 20 g in 1000 mL of distilled water were used as the medium.

Initially, the stock cultures of bacteria were revived by inoculating in broth media and grown at 37 °C for 18 h. The agar plates of the above media were prepared and wells were made in the plate. Each plate was inoculated with 18 hrs old culture (100 μ l, 10^{-4} cfu) and spread evenly on the plates. After 20 min, the wells were filled with the compound (20mg/ml stock solution). All the plates were incubated at 37 °C for 24 h and the diameter of inhibition zone were recorded. Gentamycine was used as standard.

Antifungal screening using cup-plate method:

Methodology: Potato Dextrose Agar (PDA) 250g of peeled potato were boiled for 20 min and squeezed and filtered. To this filtrate, 20g of dextrose was added and the volume was made up to 1000mL by distilled water.

Initially the stock cultures were revived by inoculating in broth media and grown at 27°C for 48h. The agar plates of the above media were prepared and wells were made in the plate. Each plate was inoculated with 48 hrs, old culture (100 μ L, 10^{-4} cfu) and spread

evenly on the plate. After 20 min, the wells were filled with a compound (20mg/ml stock solution). All the plates were incubated at 27°C for 48 h and the diameter of inhibition zone were noted. Amphotericin was used as standard.

RESULTS AND DISCUSSION

Analytical data for the complexes indicates 1:1 stoichiometry for all the complexes (Table-1). All the complexes possess high melting point and are stable in air and are partially or insoluble in common organic solvents and soluble in DMF, DMSO and pyridine. The molar conductance of the complexes falls in

the range of 9.95 – 16.85 $\text{ohm}^{-1}\text{cm}^2\text{mole}^{-1}$ in DMF (10^{-3} M) solution. These values suggest non-electrolytic behavior of the complexes¹⁴.

Magnetic properties

The observed magnetic moments values for Co (II) and Ni (II) complexes showed 4.84 and 2.96 BM, respectively. These values are expected for octahedral geometry of Co(II) and Ni(II) complexes^{15, 16}. The Cu (II) complex showed the magnetic moment value of 1.68 BM, suggesting a distorted octahedral geometry around the Cu (II) complex¹⁷. The above magnetic movements values obtained at room temperature are given in Table-1.

Table-1
Elemental analysis, molar conductance and magnetic susceptibility data for the Ligand (BCACP) and its metal complexes.

Ligand / Complexes	Yield (%)	Mol wt	M P (°C)	Elemental analysis found /calculated (%)					\wedge^*_M	μ_{eff} (BM)
				C	H	N	M	Cl		
[C ₁₇ H ₁₄ N ₂ O ₂]	75	278	177	73.16 (73.38)	4.98 (5.03)	9.86 (10.07)	--	--	--	--
[Cu(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂] _n	65	412	>300	49.21 (49.51)	3.18 (3.39)	6.48 (6.79)	15.27 (15.42)	17.08 (17.20)	14.85	1.68
[Co(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂] _n	68	408	290 d	49.88 (50.00)	3.98 (4.16)	6.59 (6.86)	14.26 (14.44)	17.22 (17.37)	11.25	4.84
[Ni(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂] _n	64	407	295 d	49.20 (50.12)	3.26 (3.43)	6.60 (6.87)	14.21 (14.42)	17.33 (17.42)	13.85	2.96
[Zn(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂]	66	414	280 d	49.02 (49.27)	3.16 (3.38)	6.45 (6.76)	15.58 (15.79)	16.98 (17.12)	09.95	--
[Cd(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂]	65	461	>295	44.08 (44.25)	2.96 (3.03)	5.96 (6.07)	24.19 (24.38)	15.12 (15.37)	12.05	--
[Hg(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂]	68	549	>295	37.02 (37.15)	2.98 (3.09)	4.86 (5.10)	-- (36.53)	12.78 (12.91)	16.85	--

* Molar conductance values in $\text{ohm}^{-1}\text{cm}^2\text{mole}^{-1}$.

Electronic spectral studies

The electronic spectra of the Co(II), Ni(II) and Cu(II) complexes were taken in DMF solution (10^{-3} M) at room temperature and spectral data are given in Table -2. The electronic spectra of the Co(II) complex exhibit two bands at 16453 and 20740 cm^{-1} . These two bands are assigned to $^4T_{1g}(F) \rightarrow ^4A_{2g}(F)$ (ν_2) and $^4T_{1g}(F) \rightarrow ^4T_{2g}(F)$ (ν_3) transitions, respectively, in

an octahedral environment¹⁸. The band ν_1 could not be observed due to the limited range of the instrument. However ν_1 would be calculated using band-fitting procedure¹⁹. The octahedral geometry is further supported by the values of ligand field parameters like Dq, B', β , $\beta\%$ and LFSE. All these values are given in Table-2. The reduction in Racah parameter values from free ion value (971) suggests

measure of covalent character of the M-L bond. The hexa coordinated Ni(II) complex exhibit two bands at 15742 and 25820 cm^{-1} , are assignable to ${}^3A_{2g}(F) \rightarrow {}^3T_{1g}(F) (\nu_2)$ and ${}^3A_{2g}(F) \rightarrow {}^3T_{1g}(P) (\nu_3)$ transitions, respectively, in an octahedral environment. The lowest band ν_1 ($10Dq$) could not be observed due to limited range of the instrument used. However, it was calculated by using an equation suggested by

Underhill and Billing. The β value for Ni(II) complex is less than the Co(II) complex indicating more covalency of M-L bond. The Cu(II) complex show broad asymmetric band in the region 13824-16168 cm^{-1} . The broadness of the band may be due to dynamic Jahn-Teller distortion²⁰. These observations suggest the distorted octahedral structure around Cu(II) ion.

Table-2

Electronic spectral data and ligand field parameters of the Co (II), Ni(II) and Cu(II) complexes

Ligand/Complexes	Transitions in cm^{-1}			Dq cm^{-1}	B' cm^{-1}	β	$\beta\%$	ν_2/ν_1	LFSE kcal/mol e
	ν_1^*	ν_2	ν_3						
$[\text{Co}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)_2\text{Cl}_2]_n$	7653	16453	20743	880	948	0.976	2.36	2.150	15.08
$[\text{Ni}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)_2\text{Cl}]_n$	8150	15742	25820	815	978	0.940	6.153	1.609	27.94
$[\text{Cu}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)_2\text{Cl}_2]_n$	13824 - 18168			1599					27.41

* Calculated values

¹H NMR spectral studies

The ¹H NMR spectra of the Schiff's base and its Zn (II) and Cd (II) complexes are taken in DMSO-d₆. The signal at δ 10.809 (s, 1H) is assigned to amide proton (-CONH). The signal due to (-CONH) shifted downfield in the spectra of the Zn (II) and Cd (II) complexes in the region of δ 10.911 and 10.957 (s, 1H), indicating the coordination of the oxygen of -CONH- with metal ions. The aromatic protons at δ (7.37 - 7.93) (m, Ar H) shifted down field in

the complexes. The signal at δ 3.297 (s, 3H) is assigned to methyl group of acetophenone. Thus the ¹H NMR spectral observations supported the assigned geometry.

Mass spectra studies

The mass spectrum of the Schiff base (Fig-2) showed molecular ion peak at (M+1) m/z 279, which is one mass unit, more than that of the molecular weight of the Schiff base. This supports the structure of the ligand.

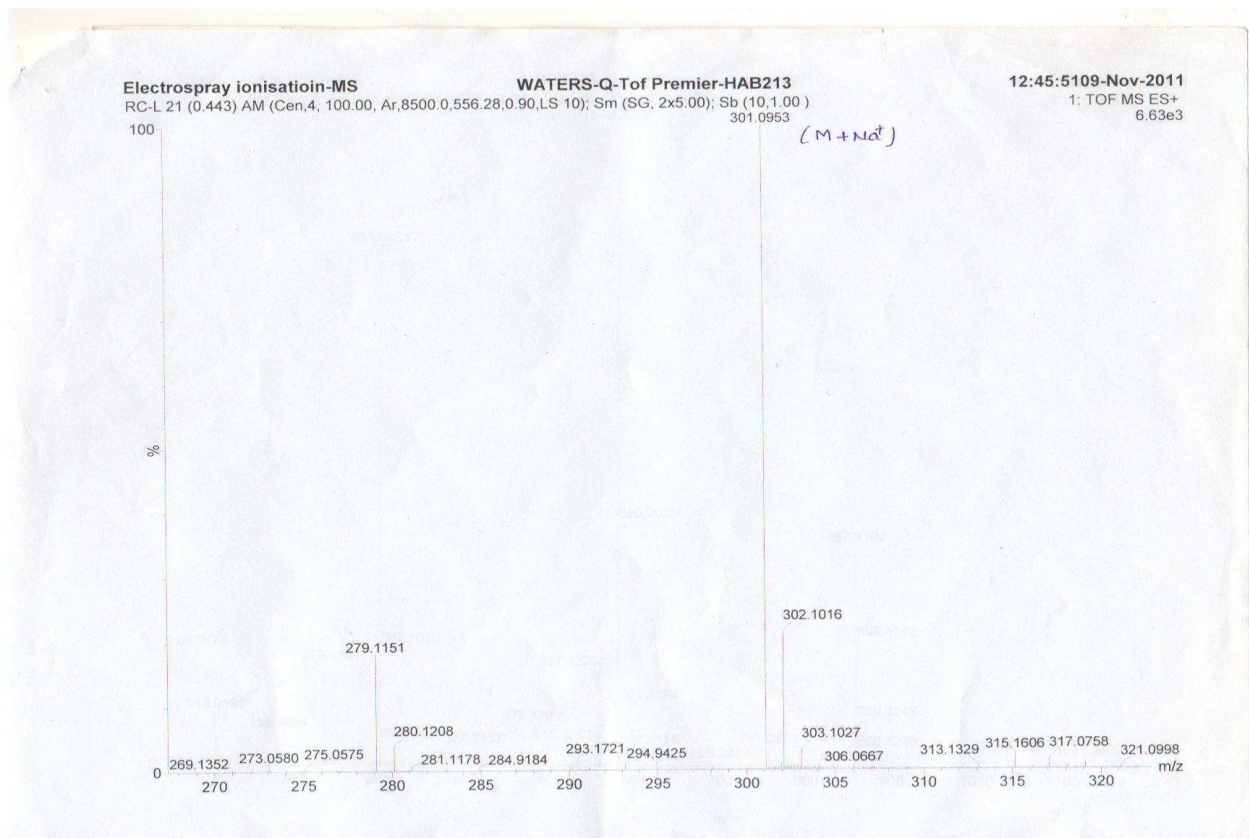


Figure-2
Mass spectra of Ligand [BCACP].

IR spectral studies

The infrared spectral data of the Schiff base and its Cu(II), Co(II), Ni(II), Zn(II), Cd(II), and Hg(II) complexes are shown in Table-3. The IR spectra of the ligand shows a broad band at 3329 cm^{-1} assignable to $\nu(\text{NH})$ of the CONH group. The shifting of this band to higher wave number side in the complexes by about $6\text{-}30\text{ cm}^{-1}$ indicates non-involvement of 'N' of the amide group in the coordination²¹. The strong band observed at 1674 cm^{-1} in free ligand is assigned to $\nu(\text{C}=\text{O})$ stretch of CONH group. This band shifts to lower wave number side by about $7\text{-}72\text{ cm}^{-1}$ in all the metal complexes. This suggests coordination of the carboxyl oxygen with metal ions. Medium to strong

intensity band at 1598 cm^{-1} in the free ligand was assigned to $\nu(\text{C}=\text{N})$ stretch of the azomethine group. This $\nu(\text{C}=\text{N})$ stretch shift to lower wave number side in all the complexes by about $41\text{-}78\text{ cm}^{-1}$ indicating involvement of the azomethine nitrogen in bonding with all the metal ions²².

Metal ligand vibrations are generally observed in the far-IR region and usually give valuable information regarding the bonding of ligand to the metal-ions. The weak intensity non-ligand bands observed in the regions $513\text{-}564\text{ cm}^{-1}$ and $420\text{-}432\text{ cm}^{-1}$ are assigned to $\nu(\text{M}-\text{O})$ and $\nu(\text{M}-\text{N})$ stretching vibration respectively in all the complexes²³.

Table-3
Important IR spectral bands (cm^{-1}) for ligand and its metal complexes

Ligand/Complexes	$\nu(\text{NH})$	$\nu(\text{C}=\text{O})$	$\nu(\text{C}=\text{N})$	$\nu(\text{M}-\text{N})$	$\nu(\text{M}-\text{O})$
$[\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2]$	3329	1674	1598	--	--
$[\text{Cu}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)\text{Cl}_2]_n$	3357	1602	1520	513	431
$[\text{Co}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)\text{Cl}_2]_n$	3325	1666	1556	524	420
$[\text{Ni}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)\text{Cl}_2]_n$	3256	1663	1557	564	431
$[\text{Zn}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)\text{Cl}_2]$	3330	1667	1554	524	430
$[\text{Cd}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)\text{Cl}_2]$	3338	1664	1555	522	432
$[\text{Hg}(\text{C}_{17}\text{H}_{14}\text{N}_2\text{O}_2)\text{Cl}_2]$	3335	1665	1556	526	432

ESR Spectrum of Cu(II) Complex:

The ESR spectrum of the Cu(II) complex (Fig-3) in a polycrystalline state was recorded at room temperature. The g_{11} and g_{\perp} values were found to be 2.173 and 2.035 respectively. The g_{av} value was calculated to be 2.081. The spectrum showed asymmetric bands with two g values. The trend $g_{11} > g_{\perp} >$

(2.00277), indicating that the unpaired electron lay predominately in the dx^2-y^2 orbital with possibly mixing of dz^2 orbital because of the low symmetry²⁴. The axial symmetry parameter 'G' is determined as $G = (g_{11} - 2.00277) / (g_{\perp} - 2.00277) = 5.31$ found to be more than 4 suggesting very weak or no interaction in the solid state²⁵.

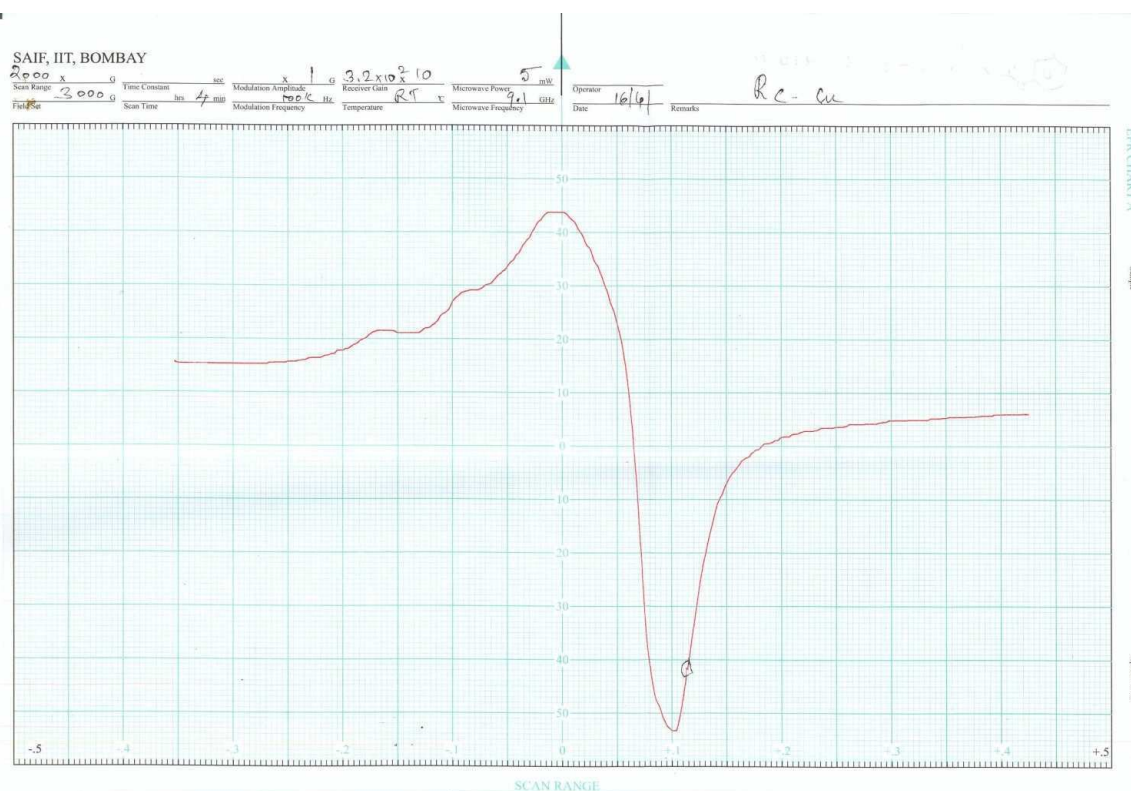


Figure-3
ESR spectrum of Cu(II) complex.

BIOLOGICAL ACTIVITY**Antibacterial and antifungal activity**

The synthesized Schiff's base and its metal complexes were tested for their antimicrobial activity against selected bacteria *E. coli* and *S. aureus* and two fungi *A. niger* and *C. albicans* was carried out. From the table-4 it is clear that Cu(II), Co(II) and Zn(II) complexes show moderate activity against bacteria *E. coli*, while Hg(II) complex show high activity against bacteria *E. coli*. The Ni(II), Co(II) and Zn(II) complexes show moderate activity, while Hg(II)

and Cu(II) complexes show high activity against bacteria *S. aureus*. These results show that the metal complexes have higher activity compared with parent ligand. In the case of fungicidal activity most of complexes and ligand show in activity against fungi *A. niger* and *C. albicans*, but in the case of Hg(II) and Cu(II) complexes show high activity against fungi *C. albicans* and *A. niger*. The activity of the complexes may be due to the presence of metal ions in the complexes.

Table-4
Antibacterial and antifungal activity results of ligand and its metal complexes(20mg/ml concentration)

Ligand/ complexes	Zone of inhibition in mm			
	Antibacterial activity		Antifungal activity	
	<i>E. coli</i>	<i>S. aureus</i>	<i>A. niger</i>	<i>C. albicans</i>
[C ₁₇ H ₁₄ N ₂ O ₂]	7	4	0	0
[Cu(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂] _n	11	12	12	20
[Co(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂] _n	9	7	0	0
[Ni(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂] _n	6	5	0	0
[Zn(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂]	10	7	0	0
[Cd(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂]	3	2	0	0
[Hg(C ₁₇ H ₁₄ N ₂ O ₂)Cl ₂]	12	13	21	27
Gentamycine	26	25	--	--
Amphotericin	--	--	08	10

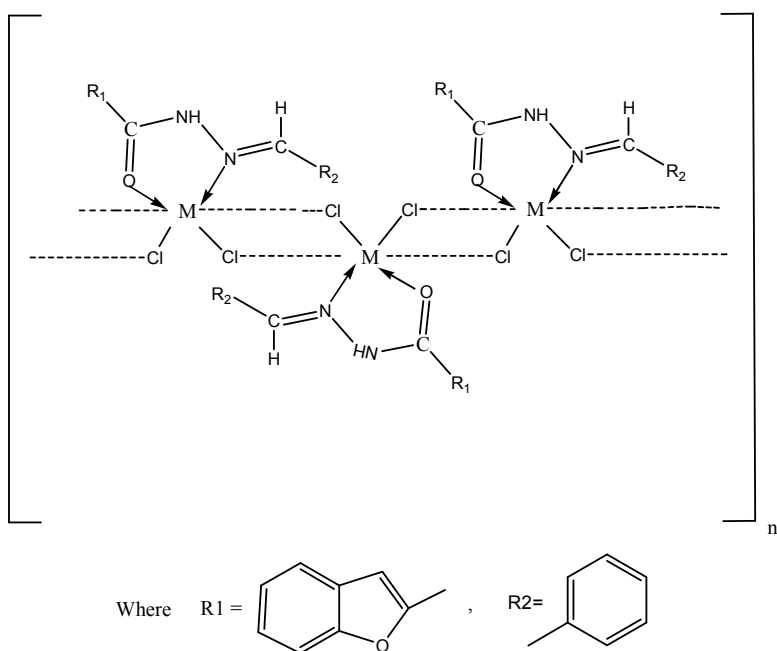


Figure-4
Suggested structure of Cu(II) ,Co(II) and Ni(II) complexes

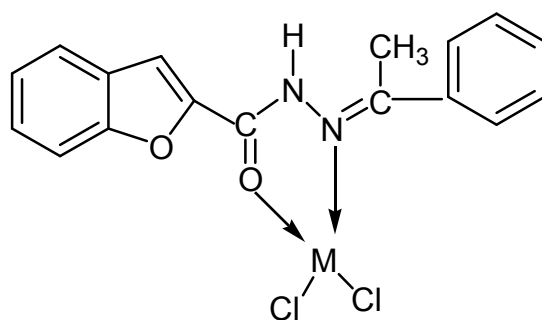


Figure.5
Suggested structure of Zn(II), Cd(II) and Hg(II) complexes.

CONCLUSION

Based on the analytical data, IR, conductivity data, magnetic properties and spectral studies we assigned the possible structures for all the complexes. Cu(II), Co(II) and Ni(II) complexes are chloride bridged polymeric in nature while Zn(II), Cd(II) and Hg(II) complexes are monomeric tetrahedral in nature. The Schiff base and its metal complexes have been screened for their antimicrobial activities. The results show that some of the complexes exhibit good activity as compared to the ligand.

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