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RESEARCH ARTICLE

METAL COMPLEXES OF SCHIFF BASE LIGAND 3-(((3-(2-HYDROXYPHENYL)-1-PHENYL-1H-PYRAZOL-4-YL)METHYLENE)HYDRAZONO)INDOLIN-2-ONE: PREPARATION, CHARACTERIZATION AND BIOLOGICAL STUDIES

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ABSTRACT

Metal complexes of Schiff bases derived from isatin monohydrazone and 3-(2-hydroxyphenyl)-1-phenyl-1H-pyrazole-4-carbaldehyde is reported and characterized by elemental analysis, conductance and magnetic measurements, IR, UV-Vis, mass, ¹H NMR, as well as by ESR and XRD studies. The low values of the molar conductance indicate non electrolyte type of complexes. Based on spectral data and magnetic moments, an octahedral geometry may be proposed for Co(II), Ni(II) and Cu(II), complexes while a tetrahedral geometry for Zn(II), Cd(II) and Hg(II) complexes. The Schiff base and its metal complexes have been screened for their in vitro antibacterial and antifungal activities by cup plate method. The DNA cleavage activities of all of the complexes were studied by the agarose gel electrophoresis method. The Schiff base and its metal complexes has been studied for their antioxidant activity. The results of bioassay revealed that, the metal complexes are active than the free ligand and their biological activity increases on complexation.

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INTRODUCTION

Schiff bases forms an important class of organic compounds and are widely studied for various biological applications over the decades (Nirmal *et al.*, 2010). Schiff bases of isatin derivatives have been used to demonstrate a variety of biological activities, such as anti-inflammatory, anti HIV and anti-depressant activities. In addition some Schiff bases show pharmacologically useful activities like anticancer, anti-hypertensive and hypnotic activities (Foziah, 2014). In the present article, we report the synthesis and characterization of a Schiff base derived from isatin monohydrazone and 3-(2-hydroxyphenyl)-1-phenyl-1H-pyrazole-4-carbaldehyde and its metal complexes to gain more information about related structural and spectral properties as well as their biological properties.

Experimental

All chemicals used were of analytical grade. isatin, hydrazine hydrate (99%) were obtained from Fluka and Sisco chemicals.

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Synthesis of Schiff base ligand: 3-(((3-(2-hydroxyphenyl)-1-phenyl-1H-pyrazol-4-yl)methylene)hydrazono)indolin-2-one

The Schiff base has been synthesized by refluxing the equimolar mixture of hot methanolic solution of 3-(2-hydroxyphenyl)-1-phenyl-1H-pyrazole-4-carbaldehyde (0.01 mol, 30 mL) and hot methanolic solution of isatin monohydrazone (0.01 mol, 30 mL) for 6-7 h in presence of catalytical amount of glacial acetic acid. The product obtained after the evaporation of the solvent was filtered, washed with cold methanol to afford Schiff base (HDPPPMHI) as shown in Fig. 1. The purity of the compound has been checked by TLC.

Synthesis of metal complexes

To the hot solution of the Schiff base ligand (0.001 mol) in ethanol (35 mL), a hot ethanolic solution of respective metal chlorides (0.001 mol) in ethanol (15 mL) was added and reaction mixture was then treated with sodium acetate (0.5 g) and the refluxing was continued further for 2 h. The resulting reaction mixture was then decomposed by pouring into distilled water (80-100 mL). The colored complex separated out was collected by filtration, washed with distilled water, then with hot ethanol and dried in vacuum over anhydrous calcium chloride. Elemental analysis data shown in Table 1. The complexes were analyzed for their metal and chloride contents by standard methods (Vogel, 1968).

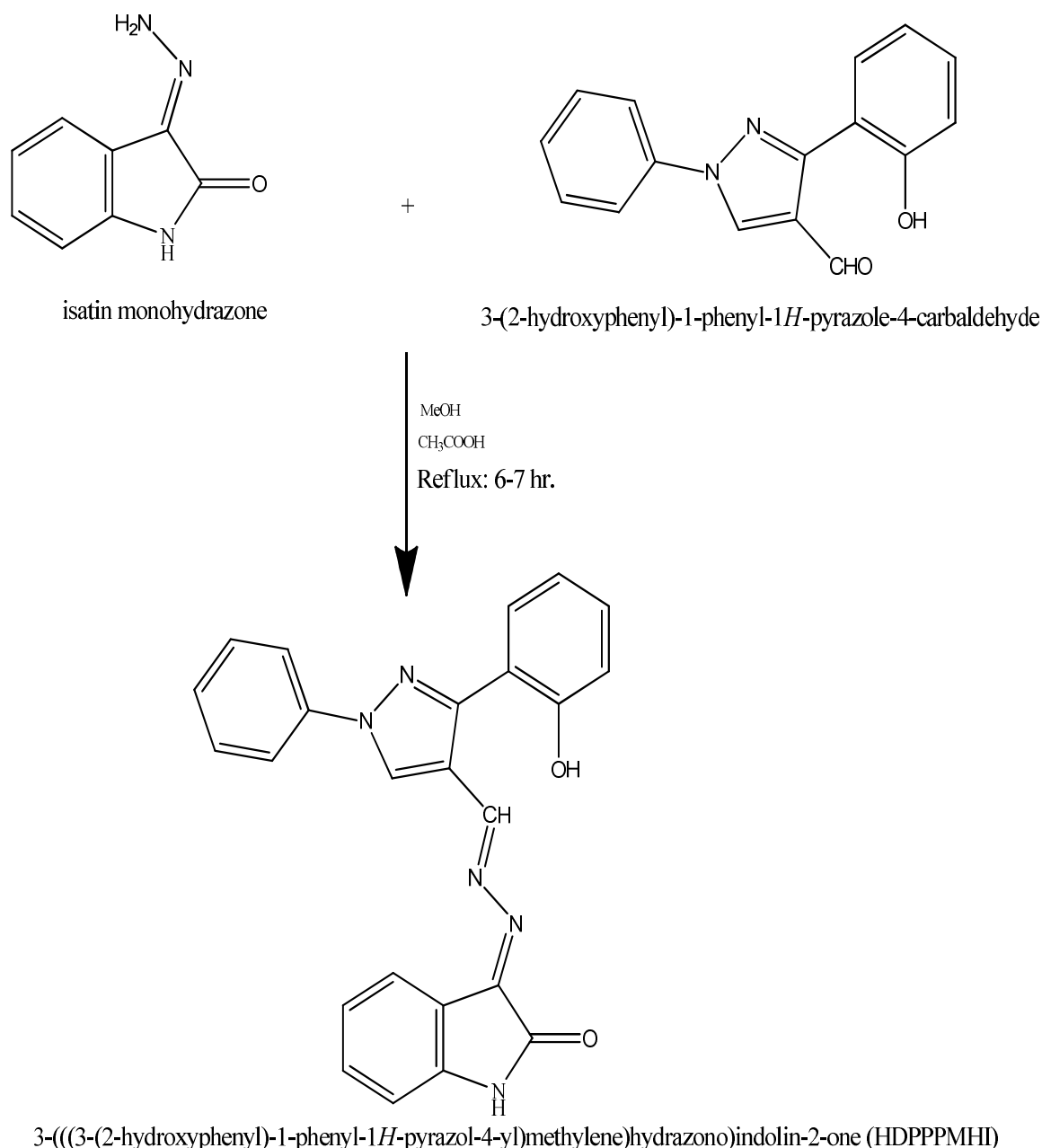


Fig. 1. Schematic representation of Schiff base (HDPPPMHI)

Table 1. Physical and analytical data of the ligand (HDPPPMHI) and its metal (II) complexes

Compound	Molecular formula	Yield (%)	Found (Calculated) %					M	Cl	$\Omega^{-1} \text{ cm}^2 \text{ mol}^{-1}$	μ_{eff} (BM)
			C	H	N						
L	[C ₂₄ H ₁₇ N ₅ O ₂]	89	70.65 (70.75)	4.19 (4.21)	17.00 (17.19)	-	-	-	-	-	
[CoL2]	[CoC ₄₈ H ₃₂ N ₁₀ O ₄]	90	66.00 (66.13)	3.69 (3.70)	16.00 (16.07)	6.68 (6.76)	-	-	19.26	4.77	
[NiL2]	[NiC ₄₈ H ₃₂ N ₁₀ O ₄]	92	66.02 (66.15)	3.68 (3.70)	16.01 (16.07)	6.63 (6.74)	-	-	22.18	3.29	
[CuL2]	[CuC ₄₈ H ₃₂ N ₁₀ O ₄]	80	65.63 (65.78)	3.55 (3.68)	15.82 (15.98)	7.12 (7.25)	-	-	24.61	1.77	
[ZnLCl]	[ZnC ₂₄ H ₁₆ ClN ₅ O ₂]	85	56.75 (56.83)	3.01 (3.18)	13.78 (13.81)	12.70 (12.94)	6.26 (6.38)	-	14.56	-	
[CdLCl]	[CdC ₂₄ H ₁₆ ClN ₅ O ₂]	70	52.00 (52.01)	2.89 (2.91)	12.58 (12.64)	20.19 (20.25)	6.92 (7.01)	-	18.40	-	
[HgLCl]	[HgC ₂₄ H ₁₆ ClN ₅ O ₂]	78	44.71 (44.87)	2.42 (2.51)	10.89 (10.90)	-	-	19.48	-	-	

Physical measurements

Elemental analysis carbon, hydrogen and nitrogen analysis was carried out using a Heracus Carlo Erba 1108 CHN analyzer at STIC, Cochin. The IR spectra of the Schiff base and its Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Hg(II) complexes were recorded in the region of 4000-250 cm⁻¹ on a Perkin Elmer - Spectrum RX-IFTIR spectrophotometer.

The electronic spectra of the Co(II), Ni(II) and Cu(II) complexes were recorded on an ELICO SL-164 double beam UV-visible spectrophotometer in the range of 200-900 nm in DMF (10⁻³ M) solution. Magnetic susceptibility measurements were made at room temperature on a Gouy balance using Hg[Co(NCS)₄] as the calibrant. Molar conductivity measurements were recorded on an ELICO CM-180 conductivity bridge in DMF solution (10⁻³ M) using a dip-type

conductivity cell fitted with a platinum electrode, The $^1\text{H-NMR}$ spectra were recorded in DMSO-d_6 on a Bruker 500MHz spectrophotometer using TMS as an internal standard. The mass spectra were recorded on a JEOL GC mate mass spectrophotometer. The ESR spectrum of the Cu(II) complex in the polycrystalline state was recorded on a Varian-E-4X band EPR spectrophotometer using TCNE as the 'g' marker ($g = 2.00277$) at room temperature. The XRD patterns of the ligand and its Cu(II) complex were recorded on a Rigaku D_{max} X-ray diffractometer using $\text{Cu K } \alpha = 1.5404$ radiation ($\lambda \text{ \AA }^\circ$).

Antibacterial and antifungal activities

The ligand and its complexes were tested against the bacterial species: *Escherichia coli*, *Staphylococcus aureus*, and *Salmonella typhi* and the fungal species: *Aspergillus niger*, *Aspergillus flavus*, and *Cladosporium*. These studies were carried out using cup-plate method (Miller and Rose, 1939). gentamycine and fluconazole were used as the standard antibacterial and antifungal agents. The test organisms were grown on nutrient agar medium in petri plates. The compound was dissolved in DMSO solution and soaked in filter paper disc of 5 mm diameter and 1 mm thickness. The discs were placed on the previously seeded plates and incubated at 37°C and the diameter of inhibition zone around each disc was measured after 24 h for bacterial and 72 h for fungal species.

Antioxidant activity: Radical scavenging activity

DPPH (2,2-diphenyl-1-picrylhydrazyl) scavenging activity was measured by the spectrophotometric method. The stock solutions (1mg/ml) of the ligand and its complexes were diluted to a final concentration of 25, 50, 75 and 100 $\mu\text{g/ml}$ in methanol, DPPH in methanol solution (1ml, 0.1 mmol) was added to 2.5 ml of test solution of different concentration and allowed to react at room temperature. Control without test compound was prepared in an identical manner. In case of blank, DPPH was replaced by ethanol. The reaction was allowed to be completed in the dark for about 30 minutes. Then the absorbance of test mixtures was read at 517 nm. The percentage inhibition was calculated and expressed as percent scavenging of DPPH radical. Butylated hydroxyl anisole (BHA), tertiary butylated hydroxyl quinoline (TBHQ) and ascorbic acid (25, 50, 75 and 100 $\mu\text{g/ml}$) were used as standards. The percentage DPPH inhibition was calculated from the following formula (Rajesh *et al.*, 2013).

$$\% \text{ DPPH inhibition} = [(\text{OD of control} - \text{OD of test}) / (\text{OD of control})] \times 100$$

DNA cleavage experiment

Cleavage reactions were run between the metal complexes and DNA, and the prepared solutions were diluted with loading dye using 1% agarose gel (Sambrook *et al.*, 1989).

Then 10 $\mu\text{g/ml}$ of ethidium bromide was added to the above solution and mixed well. The warmed agarose was poured and clamped immediately with comb to form sample wells. The gel was mounted into electrophoretic tank; enough electrophoretic buffers were added to cover the gel to a depth of about 1 mm. The DNA sample, metal complex and H_2O_2 in 4.84 g Tris base, pH 8.0; 0.5 M EDTA/1 L were mixed with loading dye and loaded into the well of the submerged gel using a micropipette. The electric current was passed into running buffer. After 1–2 h, the gel was taken out from the buffer. After electrophoresis, the gel was photographed under UV transilluminator (280 nm) and documented (Madhavan *et al.*, 2010).

RESULTS AND DISCUSSION

All the complexes are coloured, stable, insoluble in water but soluble in organic solvents like DMF and DMSO. The complexes have characteristic colour (Zamir *et al.*, 2015). The molar conductance of metal complexes was determined at a concentration 1×10^{-3} M in DMF. All the complexes showed very low molar conductance value 19.26 - 24.61 $\text{Ohm}^{-1} \text{ cm}^2 \text{ mol}^{-1}$ which indicates that the complexes are non electrolytic in nature (Geary, 1971). The observed magnetic moment value of 4.77 BM, 3.29 BM and 1.77 BM for the Co(II) , Ni(II) and Cu(II) complexes suggests octahedral geometry (Carlin, 1965).

Magnetic susceptibility measurements

The magnetic susceptibility measurements of the complexes were performed at room temperature. The magnetic moment value for Co(II) complex of the ligand (HDPPPMHI) is 4.77 BM, which is within the range of 4.46-5.53 BM which correspond to high spin octahedral environment around the metal ion (Siddappa *et al.*, 2011). On the other hand Ni(II) and Cu(II) have shown magnetic moment value 3.29 BM and 1.77 BM respectively. Which indicates octahedral geometry for their Ni(II) and Cu(II) complexes (Siddappa and Mallikarjun, 2013).

Electronic spectral studies

Electronic spectra of Co(II) , Ni(II) and Cu(II) complexes were recorded in DMF medium and spectral bands of the complexes are summarized in Table 2. The electronic spectrum of the Co(II) complex exhibits bands at 15673 and 20491 cm^{-1} , assigned to $^4\text{T}_{1g}(\text{F}) \rightarrow ^4\text{T}_{2g}(\text{P})$ and $^4\text{T}_{1g}(\text{F}) \rightarrow ^4\text{A}_{2g}(\text{F})$ transitions respectively, corresponding to octahedral geometry around Co(II) ion (Siddappa *et al.*, 2013). Electronic spectrum of Ni(II) complex in octahedral coordination is $^3\text{A}_{2g}$, the Ni(II) complex exhibits bands in an octahedral field viz., $^3\text{A}_{2g}(\text{F}) \rightarrow ^3\text{T}_{1g}(\text{F})$: (ν_2) 15197, $^3\text{A}_{2g}(\text{F}) \rightarrow ^3\text{T}_{1g}(\text{P})$: (ν_3) 24509 cm^{-1} . The lowest band ν_1 was not observed due to limited range of the instrument used. The observed transition bands lie well within the range of reported values.

Table 2. Electronic spectral bands and ligand field parameters of the Co(II) , Ni(II) and Cu(II) complexes in DMF (10^{-3} M) solution

Complexes	Transitions in cm^{-1}			Dq (cm^{-1})	B' (cm^{-1})	β	$\beta\%$	ν_2/ν_1	LFSE (kJ mol $^{-1}$)
	ν_1	ν_2	ν_3						
Co(II) complex	7184	15673	20491	848	974	0.93	6.343	2.18	14.55
Ni(II) complex	9550	15197	24509	955	736	0.70	29.14	1.59	32.74
Cu(II) complex		13698-17241		1546	-	-	-	-	26.51

These values indicate considerable covalent character and support the octahedral geometry for the Ni(II) complex (Siddappa and Nabiya sultana, 2015).

${}^2B_{1g} \rightarrow {}^2A_{2g}$, ${}^2B_{1g} \rightarrow {}^2E_g$ and ${}^2B_{1g} \rightarrow {}^2B_{2g}$ all three transitions lie within the single band in the region $13698\text{--}17241\text{ cm}^{-1}$, have been assigned to charge transfer bands from ligand to metal. The observation favour distorted octahedral geometry around Cu(II) complex (Abbas Noor Al-Shareefi *et al.*, 2013). The ligand field parameters have been calculated using the procedure given by Drago (Drago, 1968).

IR spectral studies

The important IR spectral of the ligand and its metal complexes are given in Table 3. In the IR spectrum of the ligand the broad band observed at 3387 cm^{-1} in the IR spectra of the ligand assigned to $\nu(\text{OH})$, which was found to have disappeared in all their respective complexes, there by indicating deprotonation and formation of metal-oxygen bond (Sanjoy *et al.*, 2015). This is further supported by the shifting of phenolic $\nu(\text{C-O})$ towards higher frequency, indicating the coordination of the phenolate oxygen to metal ion. The band for $\nu(\text{HC=N})$ and $\nu(\text{C=O})$ were observed in the spectra of the ligand at 1589 cm^{-1} and 1682 cm^{-1} which is shifted to a lower frequency by in the complexes indicating the involvement of the azomethine nitrogen and carbonyl oxygen in chelation with the metal ion (Zahid *et al.*, 2015).

However vibrational characteristics of the ring $\nu(\text{N-H})$ and $\nu(\text{C=N})$ of the ketimine moiety remain almost unaffected, indicating the non participation of these groups in coordination (Siddappa and Nabiya sulthana Mayana, 2014). The bands in the regions $518\text{--}569$, $455\text{--}488\text{ cm}^{-1}$ and $362\text{--}392\text{ cm}^{-1}$ are ascribed to $\nu(\text{M-O})$, $\nu(\text{M-N})$ and $\nu(\text{M-Cl})$ vibrations, respectively (Jitendra and sangeeta, 2015).

Mass spectral studies

The GC-MS spectrum of the Schiff base (HDPPPMHI) shows a molecular ion peak at m/z 407, which is equivalent to its molecular weight. The mass spectrum of the Zn(II) complex showed a molecular ion peak at m/z 507, which is the same as that of the molecular weight of the complex. This supports the suggested structure for the complex.

${}^1\text{H-NMR}$ spectral studies

${}^1\text{H-NMR}$ spectra of the ligand (HDPPPMHI) and its Zn(II) complex was recorded in DMSO-d_6 . The signal at δ (10.4) (s, 1H) was assigned to the OH group on has resonated but in the case of Zn(II) which has been disappeared indicating the involvement of phenolate oxygen in the coordination *via* deprotonation (Nayaz *et al.*, 2014). The signal at δ (8.6) (s, 1H) is due to the azomethine group in the ligand, but in case of Zn(II) complex the peak was observed at δ (9.0) (s, 1H) (Saeed and Liyaghati-Delshad, 2015).

Table 3. IR spectral bands of the ligand and its metal complexes (cm^{-1})

Tentative assignments	L	[CoL ₂]	[NiL ₂]	[CuL ₂]	ZnLCl	CdLCl	[HgLCl]
Hydrogen bonded OH group	3358	-	-	-	-	-	-
Indole ring NH	3205	3205	3205	3205	3205	3205	3205
$\nu(\text{C=O})$ ring	1682	1654	1671	1615	1610	1615	1621
$\nu(\text{C=N})$ ring	1605	1605	1605	1605	1605	1605	1605
$\nu(\text{C=N})$ aldimine	1589	1545	1517	1534	1506	1539	1555
$\nu(\text{C-O})$	1210	1254	1258	1249	1270	1292	1249
$\nu(\text{N-N})$	986	991	1002	1101	1034	1034	1106
$\nu(\text{M-N})$	-	456	458	460	480	488	455
$\nu(\text{M-O})$	-	549	536	540	569	518	541
$\nu(\text{M-Cl})$	-	-	-	-	362	380	392

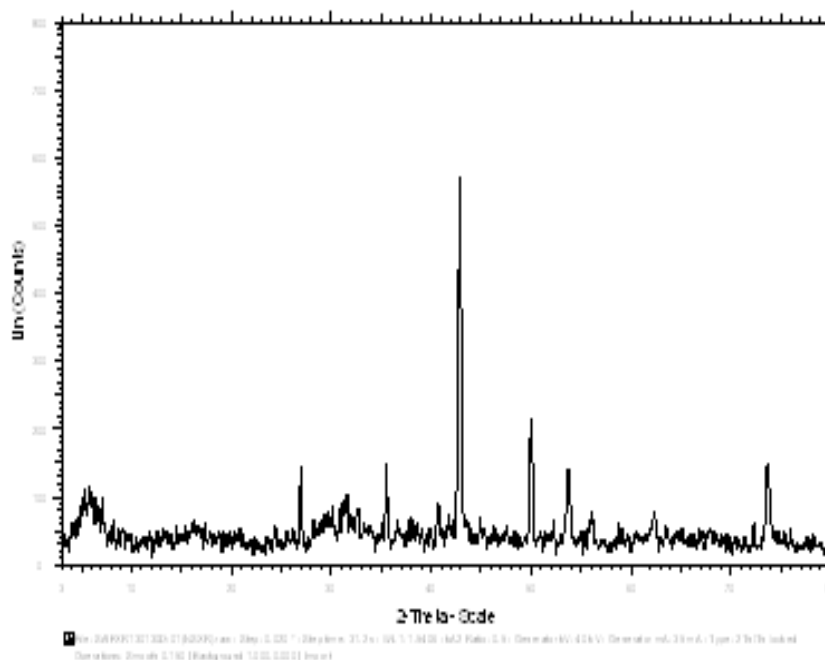


Fig. 2. Powder XRD pattern of [CuL₂] Complex

The peak appeared at δ (9.5) (s, 1H) is due to the hydrogen of -NH in the ligand, but in case of Zn(II) complex the peak was observed at δ (9.5) (s, 1H) (Siddappa and Nabiya sultana, 2014). In ligand Fourteen aromatic protons have been observe in the region δ (6.8-7.9) (m, 14H) as a multiplet, in Zn(II) complex Fourteen aromatic protons have been observe in region δ (7.0-8.2) (m, 14H) as multiplet. On comparing the $^1\text{H-NMR}$ spectra of ligand (HDPPPMHI) and the Zn(II) complex, it was observed that the signals of protons of different functionalities of the ligand have been shifted to the downfield region indicating the coordination of ligand to the metal Zn(II) ion.

ESR spectra of Cu(II) complex

ESR spectrum of copper complex provide information about the extent of the delocalization of unpaired electron. In the present study, the ESR spectrum of copper complex has been recorded a Varian-E-4X band EPR spectrophotometer using TCNE as the 'g' marker ($g = 2.00277$) at room temperature and their g_{\parallel} , g_{\perp} , g_{av} and G values have been calculated. The g_{\parallel} and g_{\perp} values were found to be 2.40514 and 2.07637, respectively. The g_{av} was calculated to be 2.1859. ESR spectrum of the copper complex revealed two g values (g_{\parallel} and g_{\perp}). The trend $g_{\parallel} > g_{\perp}$ shows that the unpaired electron is delocalized in $d_{x^2-y^2}$ orbital in the ground state of metal. The axial symmetry parameter 'G' was determined as $G = (g_{\parallel} - 2.00277) / (g_{\perp} - 2.00277) = 5.4669$, suggesting that there is no exchange interaction in the Cu(II) complex (Haranath *et al.*, 2015).

Powder X-ray diffraction

Powder X-Ray Diffraction (PXRD) spectra of the complexes were recorded on Rigaku D_{\max} X-ray diffractometer, Cu K α radiation. The diffraction spectra of sample was collected with a Cu K α as source (=1.540598 nm) and using θ -2 θ geometry, with a scanning time of 0.5 s and step of 0.03° .

The PXRD spectrum of the Copper complex is shown in Fig. 2 and data are given in Table 4. There are 13 reflections (2 θ) between 26.937 to 73.760 with maxima at $2\theta=42.882$ corresponding to the value of $d=2.107$. The interplanar spacing (d) has been calculated from the positions of intense peaks using Bragg's equation $n\lambda=2d \sin \theta$ (Where $\lambda= 1.54056 \text{ \AA}$). The observed and calculated values of d and are quite consistent. The $h^2+k^2+l^2$ values of the complex were found to be 1, 2, 3, 4, 7. The presence of forbidden number 7 indicates the Cu(II) complex belongs to hexagonal system (Manan Ahamed, 2014).

Antimicrobial evaluation of ligand and its metal complexes

The antimicrobial activity of the Schiff base ligand and Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Hg(II) metal chelates was evaluated against eight microorganisms. The inhibition zones of the Schiff base moiety and Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Hg(II) metal chelates are shown in Table 5. Results indicated that Cu(II) and Zn(II) complexes had greater bacterial and fungal activities than the ligand. The possible mechanisms of action of the test compounds based on their structures, which can bind the microorganism cell walls. The different element binding with Schiff base ligand in different prepared compounds has important role for increases its ability to bind the cell wall. This binding inhibits the biosynthesis of the peptidoglycan layer of bacterial cell walls, which affects penetration through the cell wall. The variety of antimicrobial activity and type of bacteria affected may be due to differences in the ability to bind the cell wall, which is based on the structure of the prepared compound (Omar *et al.*, 2013).

Antioxidant activity: Radical scavenging activity

The newly synthesized compounds were screened for free radical scavenging activity by DPPH method. The results are presented in Fig. 3. The results suggest that the Co(II), Ni(II), Cu(II), Zn(II) complexes have exhibited good scavenging activity. Whereas, Cd(II) and Hg(II) have shown moderate activity.

Table 4. X-ray diffraction data of [CuL₂] Complex

Peak no.	2 θ	θ	Sin θ	Sin ² θ	$h^2+k^2+l^2$	hkl	d Value		a in \AA
							Cal	obs	
1	26.937	13.468	0.23291074	0.054247412	1	1 0 0	3.30726	3.305987	3.305
2	35.539	17.769	0.30518841	0.093139971	1.71694773	1 1 0	2.52403	2.523031	3.305
3	42.882	21.441	0.36554294	0.133621641	2.46318923	1 1 0	2.10727	2.106455	3.305
4	50.039	25.0195	0.42292668	0.178866984	3.29724455	1 1 1	1.82134	1.820646	3.305
5	53.804	26.902	0.45246583	0.204725335	3.77391892	2 2 0	1.70245	1.701785	3.305
6	73.760	36.880	0.60014104	0.360169275	6.63938179	-	1.28354	1.283030	3.305

Table 5. The antimicrobial activity of ligand and its metal (II) complexes evaluated by (mm)

Compounds	Zone of inhibition in mm					
	Antibacterial activity			Antifungal activity		
	<i>E. coli</i>	<i>S. aureus</i>	<i>S. typhi</i>	<i>A. flavus</i>	<i>A. niger</i>	<i>Cladosporium</i>
L	9	10	11	11	8	6
[CoL ₂]	12	14	16	15	21	17
[NiL ₂]	22	17	19	22	13	14
[CuL ₂]	26	27	24	24	26	22
[ZnLCl]	25	28	23	26	24	24
[HgLCl]	22	24	21	23	20	18
[CdLCl]	20	23	20	22	23	21
Gentamycine	28	30	26	-	-	-
Fluconazole	-	-	-	27	29	25

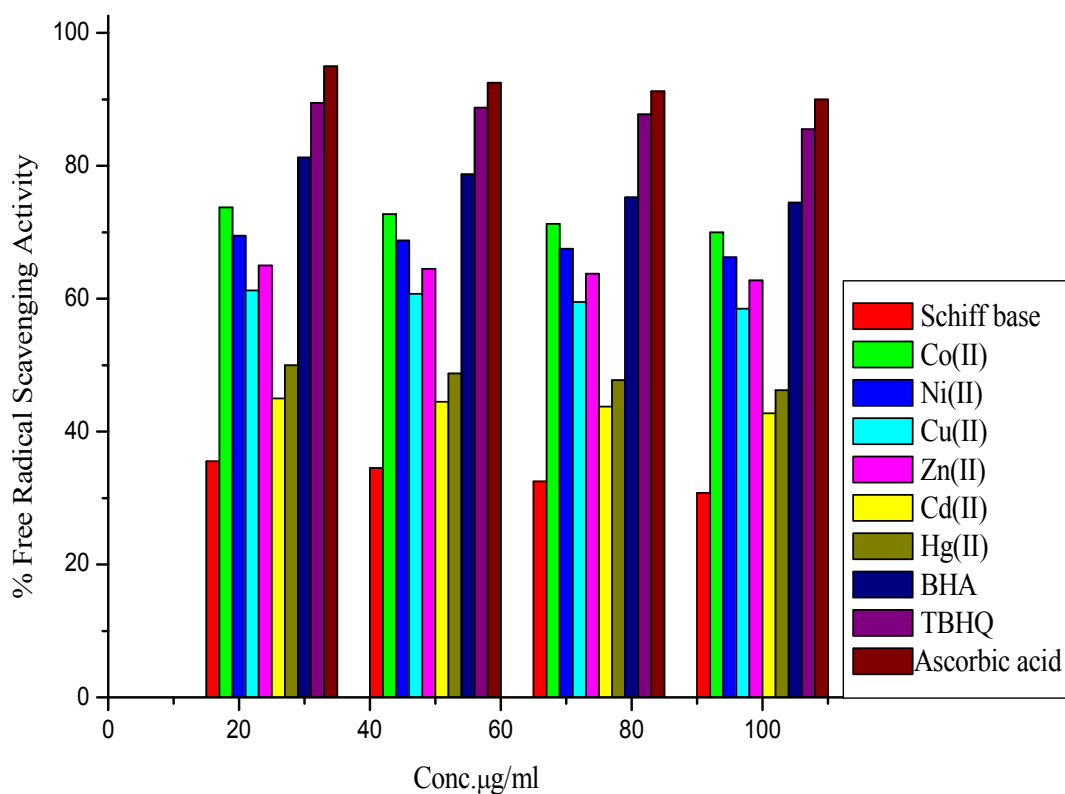


Fig. 3. Antioxidant results of Schiff base/complexes

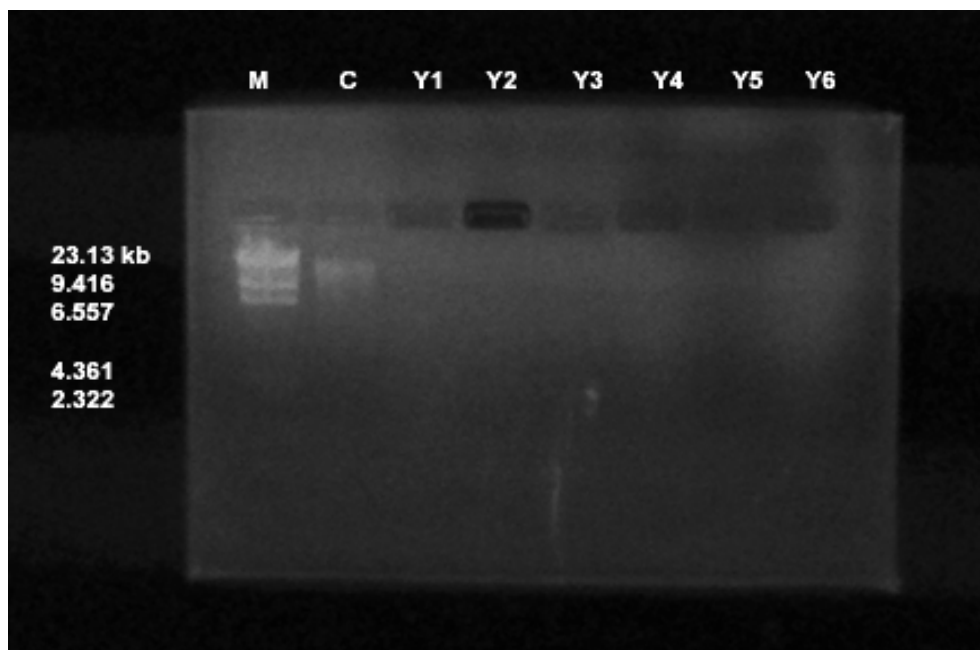


Fig. 4. Gel picture showing the cleavage analysis of samples

DNA cleavage efficiency

The Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Hg(II) complexes were studied for their DNA cleavage activity by agarose gel electrophoresis method against DNA of Calf-thymus Fig. 4. A comparative study of gel electrophoresis analysis of the control DNA with the metal complexes clearly revealed that the control DNA does not show any cleavage whereas Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Hg(II) complexes (lanes Y1-Y6 respectively) have shown complete cleavage of DNA which is evidenced by diminishing in intensity of the lanes.

Conclusion

A Schiff-base ligand and its Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Hg(II) complexes were synthesized and well characterized by elemental analyses and spectral studies. Physical and analytical data suggest that the Schiff base acts as tridentate ligand towards metal ions via azomethine-N, deprotonated-O of phenol and carboxylato oxygen. The results of these studies revealed that the free ligand and its metal complexes showed significant biological potency. On the basis of above mentioned studies we proposed the octahedral

geometry for the Co(II), Ni(II) and Cu(II) complexes and tetrahedral geometry for the Zn(II), Cd(II) and Hg(II) complexes (Fig. 5 and 6).

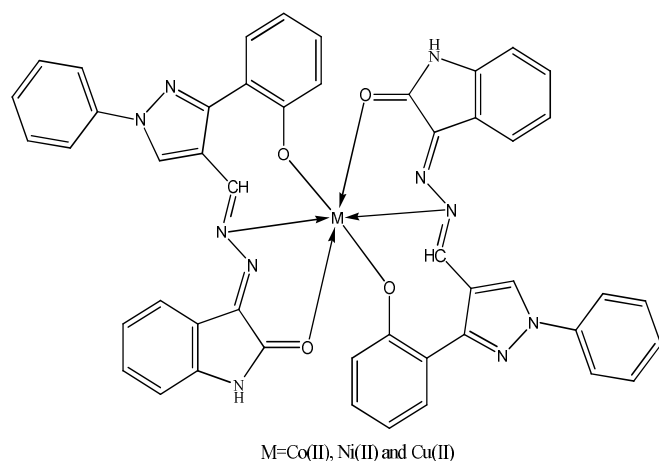


Fig. 5. Proposed structure of Co(II), Ni(II) and Cu(II) metal complexes (octahedral)

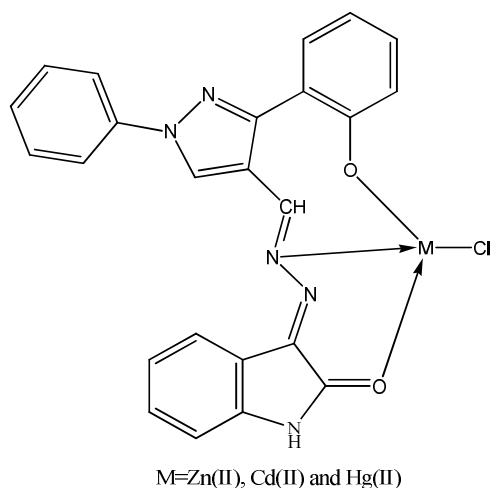


Fig. 6. Proposed structure of Zn(II), Cd(II) and Hg(II) metal complexes (tetrahedral)

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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REFERENCES

- Abbas Noor Al-Shareefi, Salih Hadi Kadhim, Waleed Abbas, J. 2013. Synthesis and study of Fe(III), Co(II), Ni(II) and Cu(II) complexes of new Schiff's base ligand derived from 4-amino antipyrine. *J. Appl. Chem.*, 2 (3): 438-446.
- Carlin, R. L. 1965. Transition metal chemistry, 2nd edn, Marcel Decker, New York.
- Drago, R. S. 1968. Physical Methods in Inorganic Chemistry, Reinhold Publishing Corporation, New York.
- Foziah, A. 2014. Spectroscopic elucidation, conductivity and activation thermodynamic parameters studies on Pt(IV), Au(III) and Pd(II) 1,5-dimethyl-2-phenyl-4-[(thiophen-2-ylmethylene)-amino]-1,2-dihydro-pyrazol-3-one Schiff base complexes. *Int. J. Electrochem. Sci.*, 9: 398 – 417.
- Geary, W. J. 1971. The use of conductivity measurements in organic solvents for the characterization of coordination compounds. *Coord. Chem. Rev.*, 7 (1): 81–122.
- Haranath. B. V., Venkateswararao, A. and Ashok, M. 2015. Synthesis, characterization and biological properties of Schiff base and transition metal complexes, *Int. J. Chem. Pharm. Sci.*, 3(1): 1453–1457.
- Jitendra, S. and Sangeeta, A. 2015. Lanthanide (III) complexes of Schiff bases of dithiocarbamate derivatives: synthesis, spectral characterization and biological evaluation. *Int. J. Adv. Eng. Global Tech.*, 3(3): 387-397.
- Madhavan sivasankaran, N., Dasan, A. and Raphael selwin, J. 2010. Synthesis, characterization, antifungal, antibacterial and DNA cleavage studies of some heterocyclic Schiff base metal complexes. *J. Saudi Chem. Soc.*, doi:10.1016/j.jscs.2010.11.002.
- Manan Ahmed, 2014. Synthesis and structural analysis of copper (II) glutathione complexes via Cu-S linkage. *World Appl. Sci. J.*, 29 (11): 1357-1362.
- Miller, R. E. and Rose, S. B. 1939. Studies with the agar cup-plate method: III. The influence of agar on mercury antiseptics. *Journal of Bacteriol.*, 38 (5): 539–547.
- Nayaz, A., Riaz, M., Altaf, A. and Madhulika, B. 2014. Synthesis, characterisation, and biological evaluation of Zn(II) complex with tridentate (NNO Donor) Schiff base ligand, *Int. J. Inorg. Chem.*, 1-5.
- Nirmal, R., Ajay babu, C. and Prasad rao, M. 2010. Synthesis and antimicrobial evaluation of novel Schiff bases analogue of 3-(4-amino) phenylimino) 5- fluorindolin-2-one. *Int. J. Pharm. Bio sci.*, 1 (3): 1-8.
- Omar, B. I., Mahmoud, A. M. and Moamen, S. R. 2013. Spectroscopic and Characterization of Medical and Physical Applications of Metal Complexes of Schiff bases in nano-sized Shape: Mg(II), Ca(II) and Ba(II) Schiff base Complexes, *Int. J. Innovative Res. Sci. Eng. Tech.*, 2(11): 6355- 6370.
- Rajesh, M., Gopi Krishna, S. and Sanjeev, R. 2013. In vitro antiinflammatory and antioxidant activity of leaf extracts of datura metal. *Asian J. Pharm. Clin. Res.*, 6 (4): 146-149.
- Saeed, J. and Liyaghati-Delshad, M. 2015. Synthesis, physical characterization and antimicrobial activity of copper (II) and cobalt (II) complexes with new Schiff base ligand containing thiocarbonylhydrazide. *J. Novel Appl. Sci.*, 4 (2): 135-139.
- Sambrook, J., Fritsch, E. F. and Maniatis, T. (1989). Molecular cloning, a laboratory manual, 2nd edn. Cold Spring Harbour Laboratory, New York.
- Sanjoy, S., Dhiraj, B. and Biswajit, S. 2015. Cu(II) complexes of an ionic liquid-based Schiff base [1-{2-((2-hydroxybenzylidene)amino)ethyl}-3-methylimidazolium] PF6: Synthesis, characterization and biological activities. *J. Serb. Chem. Soc.*, 80 (1): 35–43.
- Siddappa, K., Mallikarjun, K., Chandrakant P. Tukaramreddy, 2011. Synthesis, physico-chemical and antimicrobial studies of N1', N6'-bis((2-hydroxyquinolin-3-

- yl)methylene)adipohydrazide and their metal (II) complexes. *J. Chem. Pharm. Res.*, 3 (6): 780-788.
- Siddappa, K. and Mallikarjun, K. 2013. Synthesis, characterization and antimicrobial studies of N1-(1-(2-Hydroxy-5-methylphenyl)ethylidene)-2-oxo-2H-Chromene-3-carbohydrazide and its metal complexes. *J. Appl. Chem.*, 2 (3): 405-414.
- Siddappa, K. and Nabiya sultana, M. 2014. Synthesis, spectroscopic characterization and biological evaluation studies of Schiff base derived from 5-bromo-3-hydrazonoindolin-2-one with 5,5- dimethylcyclohexane-1,3-dione and its metal complexes. *International J. Res. Chem. Environ.*, 4: 78-84.
- Siddappa, K. and Nabiya sultana Mayana, 2014. Synthesis, spectroscopic characterization, and biological evaluation studies of 5-bromo-3-(((hydroxy-2-methylquinolin-7-yl)methylene)hydrazono)indolin-2-one and its metal (II) complexes. *Bioinorg. Chem. Appl.*, 1-11.
- Siddappa, K. and Nabiya sultana, M. 2015. Synthesis, structural characterization, antimicrobial activity and DNA cleavage of transition metal (II) complexes derived from 5-bromo-3-hydrazonoindolin-2-one, 3-((2-aminoethyl)amino)quinoxalin-2(1H)-one and 2,2-dimethyl-1,3-dioxane-4,6-dione. *Int. J. Cur. Res.*, 7 (1): 11349-11354.
- Siddappa, K., Sunilkumar, B. M. and Manikprabhu, D. 2013. (E)-3-2-(1-(2,4-dihydroxyphenyl)ethylideneamino)phenyl)-2- methylquinazoline-4(3H)-one Schiff base and its metal complexes: a new drug of choice against methicillin-resistant *Staphylococcus aureus*. *Bioinorg. Chem. Appl.*, 1-8.
- Vogel, A. I. 1968. A Text book of quantitative inorganic analysis, 3rd Edn.; Longman: London.
- Zahid, K., Zahida, T. M., Muhammad Asad, K. T., Khalid, M. K., Lubna, I. and Mehreen, L. 2015. Synthesis, characterization, in-vitro antimicrobial and antioxidant activities of Co⁺², Ni⁺², Cu⁺² and Zn⁺² complexes of 3-(2-(2-hydroxy-3-methoxybenzylidene)hydrazono)indolin-2-one, *J. Bas. Appl. Sci.*, 11: 125-130.
- Zamir ahmed, M., Saleem, M., Riaz, M., Farkhana, K. and Saima Irm, M. 2015. New series of complexes of molybdenum with biologically active Schiff bases. *Am. J. Pharm. Health Res.*, 3(1): 292-300.
