SYNTHESIS, CHARACTERIZATION AND ANTIBACTERIAL ACTIVITIES OF SOME TRANSITION METAL COMPLEXES DERIVED FROM NOVEL LIGAND 2-{(E)-[(3-HYDROXYQUINOXALIN-2-YL)METHYLDEN....

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SYNTHESIS, CHARACTERIZATION AND ANTIBACTERIAL ACTIVITIES OF SOME TRANSITION METAL COMPLEXES DERIVED FROM NOVEL LIGAND 2-{(*E*)-[(3-HYDROXYQUINOXALIN-2-YL)METHYLDENE] AMINO}BENZOIC ACID

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ABSTRACT

The complexes of Mn(II), Co(II), Ni(II), Cu(II) and Zn(II) with a new Schiff base, 2-{(E)-[(3-hydroxyquinoxalin-2-yl)methyldene] amino}benzoic acid (HQMB) formed by the condensation of 2-amino benzoic acid with 3-Hydroxyquinoxaline-2-carboxaldehyde was synthesized and characterized by elemental analysis, conductivity, UV-visible, FT-IR, ^1H NMR,TGA-DTA, Powder X-ray analysis and magnetic susceptibility measurements. All the complexes exhibit 1: 1 metal to ligand ratio. ^1H NMR study shows that Phenolic O-H of ligand participate in complex formation. Coordination of azomethine nitrogen and phenolic oxygen evidenced by IR data from the band of M-O and M-N. Electronic spectral data suggest octahedral geometry for Mn(II), Co(II) complexes while tetrahedral geometry for Ni(II),Cu(II) and Zn(II) complexes. Coordinated and lattice water molecules in the complexes are evidenced by TGA-DTA curve and the presence of the characteristic stretching band in the IR spectrum. Powder X-ray data are evidence that [Ni (HQMB) H_2O]. H_2O, [Cu (HQMB)H_2O]H_2O and [Zn(HQMB)H_2O]H_2O complex exhibit monoclinic crystal system. Ligand and its metal complexes were screened for their antibacterial activity against staphylococcus aureus, klebsiella pneumonia (Gram positive) and Escherichia coli, pseudomonas aerugenosa (Gram negative).It was observed that complexes are more potent bactericides than the ligand.

Keywords: *Schiff bases, 3-Hydroxyquinoxaline-2-carboxaldehyde, 2-amino benzoic acid, Metal Complexes; Spectral Studies.*

INTRODUCTION

Schiff bases are important in molecular processes occurring in biochemistry, material science, catalysis, encapsulation, activation, transport and separation phenomena, hydrometallurgy etc. [1-5]. Quinoxaline based Schiff bases were synthesized and characterized by several workers [6-10]. The synthesized Schiff bases were screened for their anti-inflammatory, anticonvulsant and antimicrobial activities. Quinoxaline based Schiff base metal complexes act as effective catalysts for hydrogenation of benzene [11, 12]. The quinoxaline metal complexes are found to be more active than the free ligand and some side effects may decrease upon complexation. Numerous quinoxaline derivatives are important as biocidal agents, these compounds have the ability to bind and cleave double stranded DNA under physiological conditions. They are used for genomic research and as diagnostic agents in medicinal applications [13-15]. Complexes of Schiff bases derived from 2-amino benzoic acids attract chemists and biologists due to their interesting and useful pharmacological and biological properties [16-20]. Taking into consideration of importance and biological activity of Schiff bases derived from 3-Hydroxyquinoxaline-2-carboxaldehyde and 2-aminobenzoic acid we have prepared novel tridentate ONO donor Schiff base from 3-hydroxyquinoxaline-2-carboxaldehyde and 2-aminobenzoic acid and nature of its complexes studied in detail. We have synthesized the Schiff base, 2-{(*E*)-[(3-hydroxyquinoxalin-2-yl)methyl ene]amino} benzoic acid (HQMB), by condensation reaction (Scheme-1). The Schiff base act as ONO donor dibasic ligand with Mn(II), Co(II), Ni(II), Cu(II) and Zn(II) ions. It forms octahedral complexes with Mn(II) and Co(II) and tetrahedral complexes with Ni(II), Cu(II) and Zn(II) Scheme-2.

EXPERIMENTAL SECTION

2.1 Materials and methods:

All chemicals used were of high purity analytical grade. Metal salt $(CH_3COO)_2Mn.4H_2O$(Sdfine), $(CH_3COO)_2$ Co. $6H_2O$(Sdfine), $(CH_3COO)_2Ni.4H_2O$ (Sdfine), $(CH_3COO)_2Cu.2H_2O$ (Sdfine) and $CH_3COO)_2Zn.2H_2O$(Sdfine) were used. Organic solvents like absolute ethyl alcohol, dimethyl sulphoxide and dimethylformamide were supplied by Loba. 2-aminobenzoic acid was supplied by Sigma Aldrich. 3-Hydroxyquinoxaline-2-carboxaldehyde was prepared as previously described [21]. Thin layer chromatography was carried out on silica gel 60/UV254. Melting points of products were recorded in open capillaries on digital melting point apparatus (Optics Technology) and were uncorrected. IR spectra of ligand was recorded on Perkin-Elmer FTIR Spectrophotometer in range 4000-650 cm^{-1} using ATR Instrument. IR spectra of metal complexes were recorded in KBr at 4000-400 cm^{-1} at CFC Balbhim College, Beed. Electronic spectral studies were carried out using a Schimadzu UV-2101 spectrophotometer in the range 200-900 nm at Crystal Growth Laboratory Department of Physics, Milliya College, Beed. ^1H NMR spectra were obtained on a Perkin-Elmer 300 MHz spectrophotometer using TMS as internal standard in DMSO- d_6 as the solvent at National Chemical Laboratory Pune. Elemental analyses were performed on elementar vario EL-III at SAIF Kochi. The percentage of Metal was determined by EDTA complexometric titration.TGA-DTA of complexes studied by Perkin-Elmer Diamond apparatus at STIC Cochin University Kochi. Conductance of complexes were determined in DMSO on conductivity meter Equiptronics model No.EQ665. Magnetic susceptibility was determined on SES instruments Gouy's balance model (EMU-50) at room temperature using copper sulfate as standard experiment performed in laboratory school of the Chemical Sciences SRTM University, Nanded. Mass spectrum of ligand was recorded on Shimadzu Lab solutions Mass spectrometer at IICT Hyderabad. X-ray diffraction patterns of the metal complexes were recorded using Rikagu ultima IV model X-ray diffractometer with goniometer radius 285mm. X-ray generator was operated at 40 KV and 30 mA. Cu Kα (λ =1.54056 A°) radiation was used with Ni filter. The measurements were done for 2θ values from 20° to 80° at a scan rate of 2°/min. All X-ray data recorded at (CNNUM) Center for Nanosciences and

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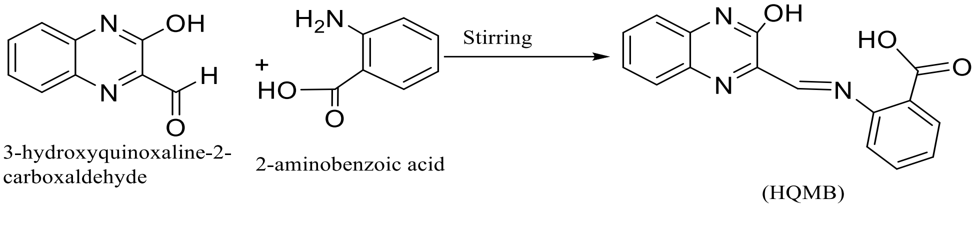
Nanotechnology, University of Mumbai. Antimicrobial activities of the ligand and their complexes were carried out by disc diffusion method using Mueller Hinton agar at department of microbiology Ahmednagar College, Ahmednagar.

2.2 Synthesis of 2-{(E)-[(3-hydroxyqu inoxalin-2-yl)methyl dene]amino} benzoic acid (HQMB)(L):

The aqueous solution of 3-Hydroxyquinoxaline-2-carboxaldehyde (2.50gm 0.001) kept on stirrer pad. Ethanolic solution of 2-amino benzoic acid (1.37 gm 0.001mol) was added to this drop wise while the solution was stirred. The solution was added till the precipitation of Schiff base was completed (**Scheme-1**). The dark yellow compound thus obtained was filtered, washed with ethanol and dried in *vacuo* over anhydrous $CaCl_2$.

The characterization data for the Ligand HQMB given below.

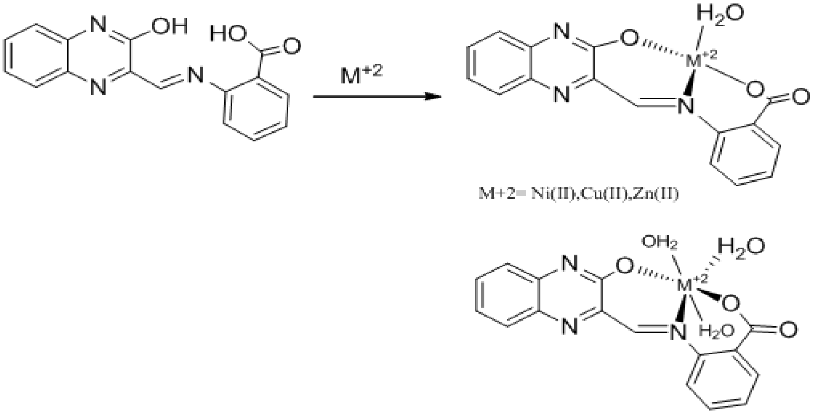
2.2a HQMB (L): Yield: 70%. $C_{16}H_{11}N_3O_3$: M.p. (°C): 258. Anal. Found: C, 65.53; H, 3.78; N, 14.33%; Calc.: C, 65.07; H, 3.01; N, 14.07%. IR (cm $^{-1}$): 3340w, 2964w, 2883w, 2700-3600w,1670m, 1586s, 1525 vs,1413s, 1221m, 1018s, 753vs (vs, very strong; s, strong; m, medium; w, weak).UV-visible in Ehanol [λ max nm(cm^{-1})]: 298(33557), 400 (25000), ^{1}H NMR (δ, ppm in dmso-d $_6$): 12.54 (s, 1H); 11.68 (s, 1H); 9.78 (s, 1H); 6.5-7.94(m, 8H); ESI-MS in 90% ehanol m/z: 295.



Scheme-1

***2.3* Synthesis of complexes of HQMB:**

The Schiff base HQMB(0.01 mol, 0.293 gm) was dissolved in 50 mL ethanol to this solution metal salt (0.01mol; manganese acetate tetra hydrate 0.245gm,Cobalt acetate hexa hydrate 0.249gm, Nickel acetate 0.249gm, Cupric acetate 0.219gm, Zinc acetate 0.198gm) in 20 mL water was added. The solution was refluxed for half an hour instantaneous formation of a precipitate was observed, and the refluxing was continued for one more hour to ensure complete precipitation. The precipitate formed was filtered, washed with ethanol and kept in desiccators.



Scheme-2

The characterization data for the complexes are given below.

2.3a [Mn(HQMB)(H₂O)₃]H₂O (1): [Yield:45%], Colour: Yellow, Analysis: Calculated for $C_{16}H_{15}N_3O_6Mn$: C,48.99; H, 3.65; N, 9.52; Mn,12.45 Found: C, 48.97; H, 3.60; N, 9.08; Mn,12.07 IR / cm-1: 3340w, 2500-3700w, 1666vs, 1576m,1517m, 1413m, 1050 m,513m, 425m, (vs, very strong; s, strong; m, medium; w, weak).UV-visible in DMSO [λ max nm(cm^{-1})]: 280(24800), 360 (23500), Conductivity in 10^{-3} Molar DMSO (λ m): 7.50 Mho cm^2 M^{-1}, Magnetic moment(μ $_{\text{eff}}$):5.78.

2.3b [Co(HQMB)(H₂O)₃]H₂O (2): [Yield:70%], Colour: Brick red, Analysis: Calculated for $C_{16}H_{15}N_3O_6Co$: C,48.55; H, 3.62; N, 9.44; Co,13.24 Found: C, 47.08; H, 3.45; N, 9.32; Co,13.02 IR / cm-1: 3292w, 2500-3600w, 1656s, 1580vs, 1526m, 1410m, 1149m, 537m,437m, (vs, very strong; s, strong; m, medium; w, weak).UV-visible in DMSO [λ max nm(cm^{-1})]: 365(27397), 495 (20202),685(14598), Conductivity in 10^{-3} Molar DMSO (λ m): 10.3 Mho cm^2 M^{-1}, Magnetic moment(μ $_{\text{eff}}$):4.85.

2.3c [Ni(HQMB)(H₂O)]H₂O (3): [Yield:63%], Colour: Brown, Analysis: Calculated for $C_{16}H_{11}N_3O_4Ni$: C,48.58; H, 3.62; N, 9.44; Ni,13.19 Found: C, 48.32; H, 3.20; N, 9.22; Ni,13.02 IR / cm-1: 3286w, 2500-3600w, 1648w, 1590vs, 1567m, 1413m, 1140m, 526m,450m, (vs, very strong; s, strong; m, medium; w, weak).UV-visible in DMSO [λ max nm(cm^{-1})]: 364(27472), 400 (25000),545(18349), Conductivity in 10^{-3} Molar DMSO (λ m): 17.50 Mho cm^2 M^{-1}, Magnetic moment(μ $_{\text{eff}}$):3.35.

2.3d [Cu(HQMB)(H₂O)]H₂O (4): [Yield:82%], Colour: Black, Analysis: Calculated for $C_{16}H_{11}N_3O_4Cu$: C,51.54; H, 2.97; N, 11.27; Cu,17.04 Found: C, 50.54; H, 2.12; N, 11.20; Cu,17.03 IR / cm-1: 3346w, 2800-3700w, 1579m, 1529vs, 1435w, 1413m, 1300s, 1145m, 550m,410m, (vs, very strong; s, strong; m, medium; w, weak).UV-visible in DMSO [λ max nm(cm^{-1})]: 365(27397), 465 (20205),560(17600), Conductivity in 10^{-3} Molar DMSO (λ m): 8.00 Mho cm^2 M^{-1}, Magnetic moment(μ $_{\text{eff}}$):2.13.

2.3e [Zn(HQMB)(H₂O)]H₂O (5): [Yield:72%], Colour: Orange, Analysis: Calculated for $C_{16}H_{11}N_3O_4Zn$: C,51.29; H, 2.96; N, 11.21; Zn,17.46 Found: C, 51.12; H, 2.50; N, 10.50; Zn,16.94 IR / cm-1: 3334w, 2800-3700w, 1649w, 1549s, 1524vs, 1414m, 1130m, 963s, 510m,410m, (vs, very strong; s, strong; m, medium; w, weak).UV-visible in DMSO [λ max nm(cm^{-1})]: 350(28571), Conductivity in 10^{-3} Molar DMSO (λ m): 6.54 Mho cm^2 M^{-1}, Magnetic moment(μ $_{\text{eff}}$):0.0.

2.4 Antimicrobial Activity:

The in vitro biological screening effects of the investigated compounds were tested against the bacteria: *staphylococcus aureus, klebsiella pneumonia* (Gram positive) and *Escherichia coli, pseudomonas aerugenosa* (Gram negative) by disc diffusion method [22] using Mueller Hinton agar as the medium. The stock solution (100 μg/ml) was prepared by dissolving the compounds in DMSO. In a typical procedure [23] well was made on the agar inoculated with microorganisms. The well was filled with the test solution using micropipette and plates were then kept in a refrigerator for 30-45 minutes and then incubated at 37°C for 18-24 hours. After incubation, diameter of zone of complete inhibition (Including diameter of the well) was measured in mm using an antibiotic zone reader.

RESULTS AND DISCUSSIONS

All the complexes are having different colors, non-hygroscopic solids. They are soluble in DMSO, slightly soluble in DMF, insoluble in ethanol, methanol and benzene. The molar conductance values indicate non-electrolytic nature [24]. μ_{eff} value of Mn(II) complex was found to be 5.78 B.M. as expected, to high spin d^5 system [25-27] for octahedral complexes. The room temperature magnetic moment of the cobalt (II) complex is 4.85 B.M. which indicates octahedral geometry around a cobalt atom. The Ni (II) complex exhibits a magnetic moment value of 3.35 B.M. which observed for tetrahedral Ni (II) complexes. The magnetic moment of the copper (II) complex was 2.13 B.M. suggest tetrahedral geometry [28]. for copper complex with sp^3 hybridization.

3.1 Infrared spectra:

The IR spectrum of the ligand compared with that of the complexes to know the changes during complex formation. Schiff base υ(C=N) showed band at stretching frequency 1670 cm^{-1} in the free ligand, after complexation υ(C=N) band observed below 1670 cm^{-1}. The symmetric υ (COO-) stretching [29] observed around 1410-1440 cm^{-1}. All the complexes showed a broad absorption band around 3200-3400 cm^{-1} due to the presence of the water molecule, υ(C-H) and υ(Ar-H) stretching observed in the same region. The broad band at 2700- 3700 cm^{-1} in the spectra of complexes indicates presence of lattice/ coordinated water [30] this fact also indicated by the results of elemental analysis and TGA-DTA of these complexes [31]. Furthermore, in these complexes, two non-ligand bands in the region 510-550 cm^{-1} and 410-440 cm^{-1} can be assigned to the stretching modes of the metal to ligands bonds, υ (M-O) and υ (M-N) respectively [32].

3.2 ^{1}H NMR spectra:

The phenolic -OH signal at δ 12.54 ppm(s, 1H) observed in the spectrum of the ligand does not observed in spectrum of the Zn (II) complex, indicating the participation of the phenolic -OH group in chelation with proton displacement. The signal due to azomethine proton at 8.99 ppm (s, 1H) gets shifted to lower δ value upon complexation, due to the donation of the lone pair of electrons by the nitrogen to the central metal atom, resulting in the formation of a coordinate linkage (M -N). The aromatic and quinoxaline protons resonate as a number of complex multiplets in the region δ 6.00-8.00 ppm (m, 8H). Singlet for water molecule does not seen in the spectrum it may be overlap with H_2O in DMSO.

3.3 Electronic absorption spectra:

Electronic spectra of the Schiff base and metal complexes were taken in DMSO (≈5×10^{-4} molar) in the range 50000-11111 cm^{-1}. The absorption maxima are listed in the **Table I.** The spectrum of Mn(II) contains bands at 23500 and 24800 cm^{-1}, which corresponds to charge transfer transitions [33]. The three spin allowed electronic transitions are expected for octahedral Cobalt(II) complexes. The shoulder band observed at 27397 cm^{-1}, 20202 cm^{-1} and 14598 cm^{-1} can be assigned to transition from $^4T_1g(F)\rightarrow{}^4T_1g(P)$, $^4T_1g(F)\rightarrow{}^4A_2g(F)$ and $^4T_1g(F)\rightarrow{}^4T_2g(F)$ respectively**.** Intensities of these bands are very low expected for the octahedral complexes [34]. The Nickel(II) complex exhibits, bands at 27472, 25000 and 18349 cm^{-1} which can be assigned to $^3T_1g(F)\rightarrow{}^3T_1g(P)$, $^3T_1g(F)\rightarrow{}^3A_2g(F)$ and $^3T_1g(F)\rightarrow{}^3T_2g(F)$ transitions respectively expected for octahedral complexes of Ni(II) [35-37]. The Copper(II) complex exhibits ligand to metal charge transfer transitions at 27397 cm^{-1} and 20205 cm^{-1} while transition $^2T_{2g}\rightarrow{}^2E_g$ shows absorbance [38] at 17600 cm^{-1}.

Table No. I: UV-visible spectral data of Metal complexes in DMSO (10^{-5} mol l^{-1})

Ligand/ Complex	λ max nm	cm^{-1}	Tentative assignment	Ligand/ Complex	λ max (nm)	cm^{-1}	Tentative Assignment
L	298	33557	π→π*	**3**	364	27472	$^3T_1g(F)\rightarrow{}^3T_1g(P)$
	400	25000	n→π*		400	25000	$^3T_1g(F)\rightarrow{}^3A_2g(F)$
1	280	24800	Charge Transfer		545	18349	$^3T_1g(F)\rightarrow{}^3T_2g(F)$
	360	23500	Charge Transfer	**4**	365	27397	Charge Transfer
2	365	27397	$^4T_1g(F)\rightarrow{}^4T_1g(P)$		498	20205	Charge Transfer
	495	20202	$^4T_1g(F)\rightarrow{}^4A_2g(F)$		560	17600	$^2T_{2g}\rightarrow{}^2E_g$
	685	14598	$^4T_1g(F)\rightarrow{}^4T_2g(F)$	**5**	350	28571	Charge Transfer

3.4 Thermal analysis:

Thermal stability of HQMB metal complexes was investigated using TGA DTA under nitrogen atmosphere with a heating rate 10^0C per minute and weight loss was measured. TGA data in the temperature range 40-700 ^{0}C is given in Table II. All complexes decompose at higher temperature compared to free Schiff base ligand suggesting coordination of Schiff bases to metals. The initial weight loss upto 100 ^{0}C in all these complexes corresponds to the loss of hydrated water molecules [39]. For Mn(II), Co(II), complexes the thermal degradation was given below.

$$[M(HQMB)(H_2O)_3]H_2O \xrightarrow[\text{-}H_2O\text{ Lattice}]{\text{below }100^oC} [M(HQMB)(H_2O)_3]$$

$$\xrightarrow[\text{-}3H_2O\text{ Coord.}]{150\text{-}270^oC} M(HQMB) \xrightarrow{\text{Above }400^oC} MO_2$$

M=Mn(II),Co(II)

For Ni(II),Cu(II) and Zn(II) complexes the thermal degradation was given below:

$$[M(HQMB)(H_2O)]H_2O \xrightarrow[\text{-}H_2O\text{ Lattice}]{\text{below }100^oC} [M(HQMB)(H_2O)]$$

$$\xrightarrow[\text{-}H_2O\text{ Coord.}]{160\text{-}260^oC} M(HQMB) \xrightarrow{\text{Above }400^oC} MO_2$$

M=Ni(II), Cu(II), Zn(II)

The Mn(II) complex loses their weight by 0.05 mg (4.30%) at temperature 81.24 ^{0}C corresponding to one lattice water molecule, it shows weight loss of 0.032 mg (12.91%) at temperature 270.20 ^{0}C corresponds to three coordinated water molecules. The Co(II) complex lose their weight by 0.039 mg (4.26%) at temperature 61.64 ^{0}C corresponding to one lattice water molecule, it shows weight loss of 0.128 mg (12.79%) at temperature 157.13 ^{0}C corresponds to three coordinated water molecules. The Ni(II) complex loses their weight by 0.08 mg (4.67%) at temperature 72.34 ^{0}C corresponding to one lattice water molecule, it also shows weight loss of 0.09 mg (4.78 %) at temperature 160.45 ^{0}C corresponds to a loss of one coordinated water molecule.The Cu(II) complex loses their weight by 0.10 mg (5.59%) at temperature 74.65^{0}C corresponding to one lattice water molecule, it also shows weight loss of 0.128 mg (4.67%) at temperature 221.39 ^{0}C corresponds to a loss of one coordinated water molecule. The Zn(II) complex loses their weight by 0.092 mg (4.58%) at temperature 77.14 ^{0}C corresponding to one lattice water molecule, it also shows weight loss of 0.101 mg (4.61%) at temperature 258.81 ^{0}C corresponds to a loss of one coordinated water molecule.

Table No. II: Thermo gravimetric analysis data below 700 ^{0}C

Complex	Temp. Range,^{0}C	Wt. Loss In mg	% Wt. Loss	Fragment Loss	The nature of Fragment
1	81.24	0.015	4.30	1H_2O	Lattice water
	270.20	0.032	12.91	3H_2O	Coordinated Water
2	61.64	0.039	4.26	1H_2O	Lattice water
	157.13	0.128	12.79	3 H_2O	Coordinated water
3	72.34	0.08	4.67	1H_2O	Lattice water
	160.45	0.095	4.78	1H_2O	Coordinated Water
4	74.65	0.102	4.59	1H_2O	Lattice water
	221.39	0.128	4.67	1H_2O	Coordinated Water
5	77.14	0.092	4.58	1H_2O	Lattice Water
	258.81	0.101	4.61	1H_2O	Coordinated Water

3.5 Powder X-ray diffraction:

In the absence of single crystal, X-ray powder data are especially useful to deduce accurate cell parameters. X-ray diffraction patterns of the metal complexes of Ni (II), Cu (II) and Zn (II) were recorded in 2θ= 20-80^0 range. The diffraction pattern reveals the crystalline nature of complexes. The indexing procedures were performed using Material Analysis Using Diffraction (MAUD) software (Luca Lutterotti). The density and particle size of metal complexes calculated cell parameters of the complexes are shown in Table III.

Table No. III: Crystallographic data of the complexes

Complexes	[Ni(L)(H_2O)]H_2O	[Cu(L)(H_2O)]H_2O	[Zn(L)(H_2O)]H_2O
Empirical formula	$C_{16}H_{11}N_3O_4Ni$	$C_{16}H_{11}N_3O_4Cu$	$C_{16}H_{11}N_3O_4Zn$
Formula Weight	367.97	372.82	374.69
Temperature (^{0}K)	298	298	298
Wavelength (A^0)	1.5418	1.5418	1.5418
Crystal System	Monoclinic	Monoclinic	Monoclinic
Space group	C1	C1	C1
2θ range	20-80^0	20-80^0	20-80^0
Unit cell dimensions(A^0)	a =5.31	a = 5.07	a = 9.08
	b =9.21	b = 8.77	b = 17.73
	c =14.40	c = 13.77	c = 10.20
Angles in degree	α =102.11	α = 90.09	α = 89.95
	β =90.20	β = 90.12	β = 124.58
	ϒ =90.10	ϒ = 89.97	ϒ = 90
Volume(A^3)	2245.72	2245.75	2345.24
Limiting indices	0≤ h ≤ 3	0≤ h ≤ 2	0≤ h ≤ 4
	0≤ k ≤ 6	0≤ k ≤ 3	0≤ k ≤ 5
	1≤ l ≤ 4	1≤ l ≤ 5	1≤ l ≤ 4
Density	1.23	1.24	1.25
Z	4	4	4

3.6 Antibacterial activities:

The free ligands and its respective metal complexes were screened against *staphylococcus aureus, klebsiella pneumonia* (Gram positive) and *Escherichia coli, pseudomonas aerugenosa* (Gram negative). The results are quite promising. In vitro antibacterial activity of the Schiff base HQMB and its complexes are presented in **Table IV**. Ciprofloxacin used as positive standards and DMSO used as solvent to dissolve metal complexes. The relative zone of inhibition of HQMB and its metal complexes shown graphically in **Figure 1.** All complexes are highly active against *E.coli* as compared to Schiff base ligand. Cu (II) complex moderately active against *K. pneumonia.* All complexes are moderately active against *S.aureus* and *P.aerugenosa.*

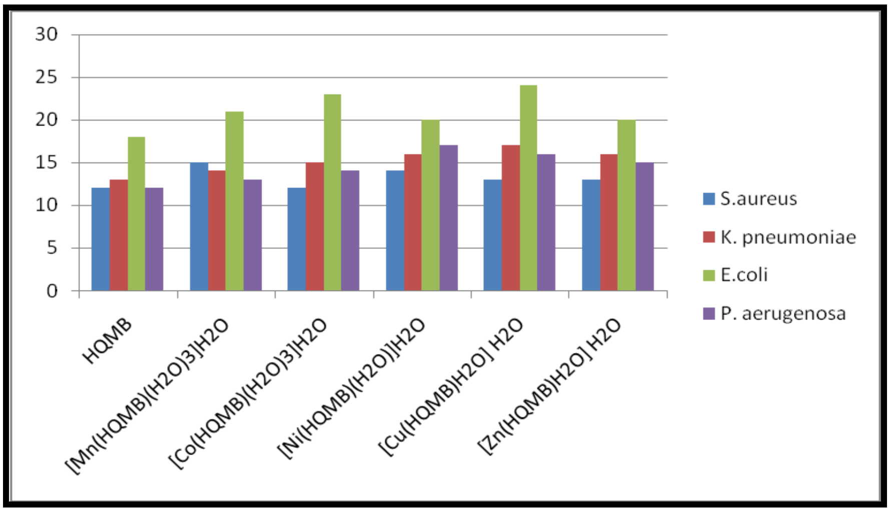


Fig. 1: Relative zone of inhibition of HQMB and its metal complexes

Table No. IV: Results of Antimicrobial activity of the HQMB and its metal complexes.

Compound	Zone of inhibition in (mm)			
	Gram positive		Gram Negative	
	S.aureus	*K. pneumoniae*	*E.coli*	*P. aerugenosa*
HQMB	12	13	18	12
$[Mn(HQMB)(H_2O)_3]H_2O$	15	14	21	13
$[Co(HQMB)(H_2O)_3]H_2O$	12	15	23	14
$[Ni(HQMB)(H_2O)]H_2O$	14	16	20	17
$[Cu(HQMB)H_2O]\ H_2O$	13	17	24	16
$[Zn(HQMB)H_2O]\ H_2O$	13	16	20	15
Ciprofloxacin*	33	34	33	23
DMSO	8	7	7	6

*Standard; Minimum inhibitory concentration of standard, ligand and metal complexes in (100 µg/ml); 8-12mm poor activity, 13-17 mm, moderate activity, 18-20 mm and above good activity.

CONCLUSION

The elemental analysis and magnetic moment measurements suggest the metal ion in all the complexes to be in the +2 oxidation state with molecular formulae, $[Mn(HQMB)(H_2O)_3]H_2O$, $[Co(HQMB)(H_2O)_3].H_2O$, $[Ni(HQMB)H_2O].H_2O$, $[Cu(HQMB)H_2O]H_2O$ and $[Zn(HQMB)H_2O]H_2O$. All complexes are found to have metal to ligand molar ratio of 1:1.^{1}HNMR study shows that Phenolic O-H of ligand takes part in complex formation. Coordination of azomethine nitrogen and phenolic oxygen is evidenced by IR data from the band of M-O and M-N. Electronic spectral data suggest octahedral geometry for Mn(II), Co(II) complexes while tetrahedral geometry for Ni(II), Cu(II) and Zn(II) complexes. Coordinated and lattice water molecules in the complexes are evidenced by TGA-DTA curve and the presence of the characteristic stretching band in the IR spectrum. Powder X-ray data are evidence that Ni(II), Cu(II) and Zn(II) complex exhibit monoclinic crystal system. All complexes are highly active against *E.coli* as compared to Schiff base ligand. Cu(II) complex moderately active against *K.pneumonia.* All complexes are moderately active against *S.aureus.*

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