



e-ISSN: 2319-8753 | p-ISSN: 2320-6710

IJRSET

International Journal of Innovative Research in
SCIENCE | ENGINEERING | TECHNOLOGY

INTERNATIONAL JOURNAL OF INNOVATIVE RESEARCH

IN SCIENCE | ENGINEERING | TECHNOLOGY

Volume 9, Issue 9, September 2020

ISSN INTERNATIONAL
STANDARD
SERIAL
NUMBER
INDIA

Impact Factor: 7.512

9940 572 462

6381 907 438

ijirset@gmail.com

www.ijirset.com

Synthesis, Characterization And Biological Study Of 3-Acetyl-4-Hydroxy-1-Methyl-2(1H)-Quinolone Hydrazone Based Cu(II) Metal Complex

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ABSTRACT : The present study emphasizes hydrazone Schiff base of 3-acetyl-4-hydroxy-1-methyl-2(1H)-quinolone and its Cu(II) metal complex. The metal complex was synthesized using Schiff base hydrazone synthesized from 4-hydroxybenzhydrazide and 3-acetyl-4-hydroxy-1-methyl-2(1H)-quinolone. Hydrazone complexes were subjected to elemental analysis, UV visible, magnetic susceptibility, FTIR, ¹H NMR spectra, mass spectra, thermal analysis, XRD analysis and antimicrobial activity. IR spectra indicate that free ligand exist in the hydrazone-ketone form in solid states. The ligand behaves as a neutral tridentate with coordination involving the carbonyl of 2(1H) quinolone, hydrazone nitrogen and keto oxygen of amide group that is ONO donor sites. The magnetic and spectral data indicate octahedral geometry for all complexes. The molar conductivity values indicating non electrolytic nature. The mass spectra and analytical data the stoichiometry of metal complex has been found to be the 2:1 ligand to metal ratio. Ea, ΔS, ΔH and ΔG parameters were evaluated for thermal decomposition stages of complexing Horowitz-Metzer and Coats-Redfern methods. X-ray diffraction data suggests triclinic structure of Cu(II) complex. The ligand and their metal complexes have been screened for their antibacterial activity using the Agar cup method at fixed concentration of 1% against microbial strains, E.coli, S. typhi, S. aureus, B. subtilis. The antibacterial screening of ligand and its complex illustrates that ligand and complex seems to be inert towards the gram positive bacteria E. coli and S. typhi. The complex shows higher activity than ligand against gram negative bacteria S. aureus and B. subtilis. Antifungal activity of ligand and complex were tested in vitro against fungi that is A. niger, P. chrysogenum, F. moniliforme, A. flavus by poison plate method using potato dextrose agar medium at fixed (1%) concentration. Ligand does not show fungal activity but its Cu(II) complex shows better activity. It suggests that the incorporation of metal ion in the ligand results in increasing the antimicrobial activity.

KEYWORDS: ONO donor ligand, octahedral complexes, ¹H NMR, Mass, TG-DTA, p-XRD, Biological activity.

I. INTRODUCTION

Schiff base hydrazones are obtained by condensation of aldehydes or ketones with acid hydrazides or hydrazines. They are versatile ligands in coordination chemistry because their wide range of applications [1-5] in catalysis, medicine, corrosion and analytical chemistry as well as functional materials [6]. Synthetic flexibility of these Schiff base hydrazones occupy special place because transition metal complexes of these ligands developed due to chelating capability. Metal complexes of hydrazones proved to have potential applications in luminescence probes [7] and molecular sensor [8]. In continuation of these studies [9-12] using hydrazone ligand bearing ONO donor set, here we report the synthesis, spectroscopic, thermogravimetric and p-XRD analysis of Cu(II) complex using tridentate Schiff base like hydrazone ligand (E)-4-hydroxy-N'-(1-(4-hydroxy-1-methyl-2-oxo-1,2-dihydroquinolin-3-yl)ethylidene)benzohydrazide, derived from the condensation of 3-acetyl-4-hydroxy-1-methyl-2(1H)-quinolone and 4-hydroxybenzhydrazide.

II. EXPERIMENTAL

2.1. Materials and Methods

All the chemical are of analytical grades. 1-methyl aniline, diethyl malonate, diphenyl ether, n-butanol, acetic acid, 4-hydroxybenzhydrazide and metal nitrate salts. All chemicals were perched from SD fine chemicals. Solvents were dried and distilled before use according to standard methods [13]. The precursor 3-acetyl-4-hydroxy-1-methyl-2(1H)-quinolone was prepared by standard method [14]. The metal nitrates were used in hydrated form. Elemental analyses (C,H,N,O,) were performed on Perkin Elmer-2400. IR spectra were recorded on FTIR Spectrophotometer model RZXC Perkin Elmer in the range (400-4000 cm^{-1}), ^1H NMR spectra were recorded on Bruker Avance II at 400 MHz using tetramethyl silane as an internal standard. Electronic spectra were recorded using Shimadzu-1800 spectrophotometer using DMSO as a solvent. The LC-MS spectra were recorded on a Waters, Q-TOF Micro Mass (LC-MS). Conductance were measured on Elico cm-180 Conductometer using 10^{-3}M solution in DMSO. Room temperature magnetic data were collected on a Guoys balance using mercury (II) tetrathiocynato cobaltacetate (II) as a calibrant. Diamagnetic contributions were estimated for each compound by using Pascal's constants. The TG-DTA analysis of Cu(II) complex was performed in an inert nitrogen atmosphere on Perkin Elmer STA 6000. The heating rate was $10^\circ/\text{min}$ and flow rate of nitrogen 50 ml/min. The reference substance used was $\alpha\text{Al}_2\text{O}_3$ in platinum crucible and sample weighed in the range of 4-12 mg. P-XRD studies were carried out with a Bruker AXS D8 Advance was recorded employing Cu k radiation ($\lambda = 1.541 \text{ \AA}$) in the range $0-60^\circ$.

2.2. Antimicrobial activity

The antibacterial activity were performed against microbial strains, *E. coli*, *S. typhi*, *S. aureus*, *B. subtilis* by agar-cup method at fixed concentration of 1% in DMSO. In this method test was performed on nutrient agar cup of 10 mm diameter were borered in the agar plate with stirile cork borer. All solutions were prepared in DMSO (1%) was added on cup, one cup blank DMSO and other for standard reference. The plates were incubated for 24 h at 37°C . The diameters of zones of inhibition for all test compounds were measured and the results compared with penicillium of the same concentration as that of the test compounds under identical conditions.

Antifungal activity of the compounds was evaluated against fungi that is *A. niger*, *P. chrysogenum*, *f. moniliforme*, *A. flavus* by poison plate method using potato dextrose agar medium at fixed (1%) concentration. The plates were incubated at 37°C for 48 h. The diameters of the zone of inhibition for all the test compounds were measured and results compared with standard drug Grisofulvin of same concentration under identical conditions.

2.3. Synthesis of ligand:

The equimolar mixture of 3-acetyl-4-hydroxy-1-methyl-2(1H)-quinolone (0.01 mol) and 4-hydroxybenzhydrazide (0.01 mol) in methanol (30mL) were refluxed in the presence of catalytic amount of glacial acetic acid (5-10 drops) for 3 hours on rotaheating mantal. The reaction mixture was then cooled to room temperature and the solid compound formed was filtered. It was then washed with methanol and dried and recrystallized from mixture of ethanol-DMF. (Yield 85%, M.p. 230°C).

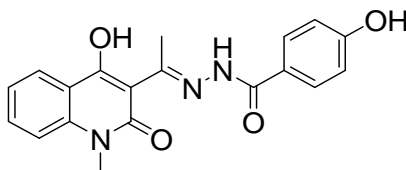
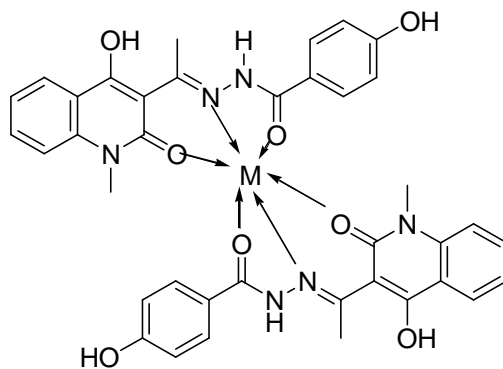


Fig. 1 structure of ligand

2.4. Synthesis of metal complex

Synthesis of Cu(II) metal complex. The metal complex by adding to the hot solution of ligand in methanol (0.02 mol in 25mL), hot methanolic solution of metal salt (0.01 mol in 25 mL) was added drop wise. To this reaction mixture, 10% methanolic ammonia was added to adjust the pH of solution to 7.5 to 8.5. The reaction mixture stirred for 3-5 hours in warm condition on magnetic stirrer to get complex in solid form. The solid complex was filtered off, washed several times with methanol and dried in vacuum over CaCl_2 .



Where M= Cu(II)

Fig. 2 structure of metal complex

III. RESULTS AND DISCUSSION

The Schiff base is soluble in DMF, DMSO and insoluble in common organic solvents. The synthesis of ligand is checked by elemental analysis, FT-IR, ¹HNMR and LC-Mass. The physical characteristics of the Schiff base and its metal complex are given in **Table 1**. The metal complex is soluble in ethanol, methanol and water. The elemental analysis data of the Schiff base and its metal complex is consistent with the calculated results from the empirical formula represent in Fig 2.

3.1. Conductivity measurements

The molar conductivity values for freshly prepared DMSO (10⁻³M) solution of the complex molar indicate that complex is non electrolytic in nature [15].

Table 1 : Physical, Analytical Data of Ligand and its Metal Complexes

Compound	Mol. formula	Colour	M.P. °C	Mol. .Wt	C% Found (Calculated)	H% Found (Calculated)	N% Found (Calculated)	O% Found (Calculated)	Metal Found (Calculated)	μ _(eff) B.M.	Molar conductance Ohm ⁻¹ cm ² mol ⁻¹
HL	(C ₁₉ H ₁₇ N ₃ O ₄)	white	230	351	64.91 (64.96)	5.58 (4.87)	10.04 (11.95)	18.04 (18.21)	--	--	---
(CuL ₂)	(C ₃₈ H ₃₄ N ₆ O ₈ Cu)	Green	>250	766.2	59.96 (59.42)	4.15 (4.46)	11.47 (10.96)	16.35 (16.69)	7.89 (8.28)	1.81	4.1

3.2. Magnetic and Electronic Spectral studies

The electronic spectra of ligand and Cu(II) complex was recorded in DMSO solution 10⁻³M molar concentration. The electronic spectrum of ligand exhibits three absorption transitions at 357.5 nm (27972 cm⁻¹), 289.5 nm (34542 cm⁻¹) and 261.5 nm (38240 cm⁻¹) assigned to the n-π* and π-π* transitions of azomethine, 2-quinolone and 4-hydroxy group respectively.

The Cu(II) complex displayed bands at 376.5 nm (26560 cm⁻¹) and 285 nm (38240 cm⁻¹) which may be assigned to ²B_{1g} → ²E_g transitions and charge transfer band respectively [16,17]. Cu(II) shows magnetic moment 1.79 B.M. shows distorted octahedral geometry [18].

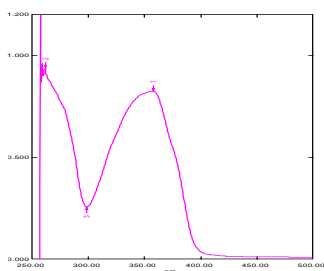


Fig.3 UV spectra of ligand.

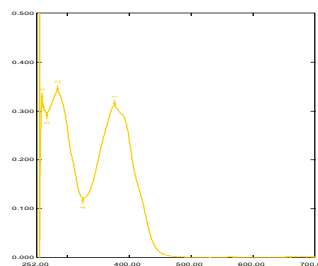


Fig. 4 UV spectra of Cu(II) complex.

3.3. Infrared spectra

IR spectral data of ligand and Cu(II) metal complex are given in **Table 2**. The spectra of ligand display bands in the region 3333, 3178, 1634, 1587, 1610, 1176, 754 cm^{-1} due to enolic -OH group of 4-hydroxy-2(1H)-quinolone, -NH stretch of hydrazone, $>\text{C}=\text{O}$ (quinolone), $>\text{C}=\text{N}$ (azomethine), $>\text{C}=\text{O}$ (amide), aromatic -OH and -NH out of plane bend respectively. The careful investigation of the spectra of the complex and ligand indicate that the bands of -OH, -NH are present in the spectra of complex indicating non coordination with metal ion. They observed at lower frequency in complex may due to its presence in chelate system rather than the open system of the ligand. In complex there is presence of strong sharp band at 1163 cm^{-1} -OH group substitution on aromatic ring this band observed at same stretching frequency as observed in ligand. The bands of $>\text{C}=\text{O}$ (quinolone), $>\text{C}=\text{N}$ (azomethine), $>\text{C}=\text{O}$ (amide) are shifted to lower frequencies by 36 cm^{-1} , 85 cm^{-1} and 51 cm^{-1} respectively on complexation due to their participation in the mode of coordination. This behavior indicates that ligand behaves as neutral tridentate ligand. In Cu(II) complex the bands at 525 cm^{-1} , 466 cm^{-1} which are not present in the spectra of the free Schiff base Hydrazones are assigned to (M-O) and (M-N) bonding [19].

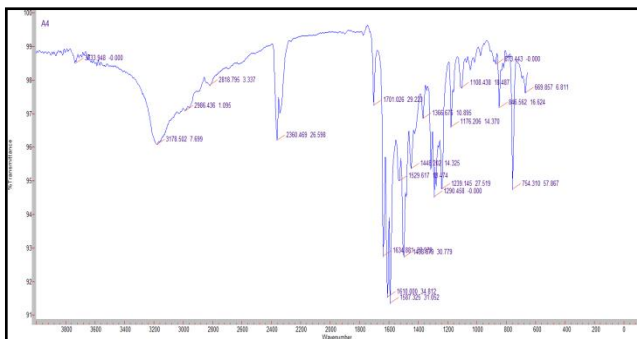


Fig.5 IR spectra of ligand

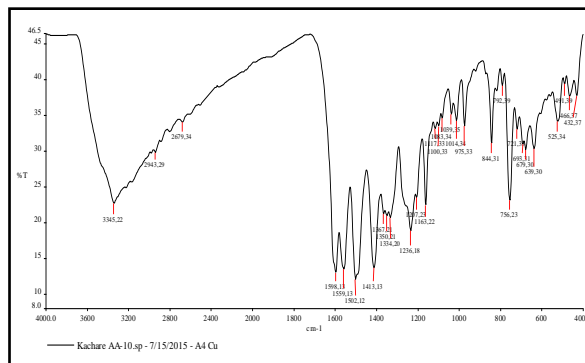


Fig.6 IR spectra of Cu(II) Complex

Table 2
Salient Features of IR Spectral Data of Ligands.
(Assignment of band frequencies to bond vibration modes)

Ligands	$\nu(\text{OH})$ Enolic	$\nu(\text{NH})$ Hydrazone	$\nu(\text{C}=\text{O})$ Quinolone	$\nu(\text{C}=\text{N})$ Azomethine	$\nu(\text{C}=\text{O})$ amide carbonyl	$\nu(\text{OH})$	$\nu(-\text{NH})$ out of plane
Ligand	3333 (b)	3178 (b)	1634 (s)	1587 (b)	1610	1176 (s)	754 (s)
CuL_2	3345 (m)	3205 (s)	1598 (s)	1502 (s)	1559 (s)	1163 (s)	756 (s)

3.4. The $^1\text{H-NMR}$ Spectra of Ligand and Complex

$^1\text{H-NMR}$ Spectra of ligand and Cu(II) complex were recorded in DMSO over the range 0-20 δ ppm. In ligand signals at 2.74 δ ppm. (s,3H, N=C-CH₃) assigned to azomethine protons, 3.53 δ ppm.(s,3H,N-CH₃), 7.20-8.14 δ ppm (m, 8H, aromatic), 10.23 δ ppm ^1H (s 1H -OH substituted on hydrazide) 11.29 δ ppm ^1H (s broad N-H), 16.76 δ ppm ^1H (broad hump, enolic-OH). In ^1H NMR spectra of complex indicates presence of broad signal at 10.35 δ ppm due -NH proton

of hydrazone indicating non enolization of -NH with amide carbonyl. In complex 9.04 δ ppm observed at ^1H (s 1H - OH substituted on hydrazone). There is slightly upfield shift due to change of environment of free ligand and complexation of ligand with metal. In Cu(II) complex aromatic frequency observed in the range 8.99-6.58 δ ppm. In complex N-CH₃ signal observed at 3.64 δ ppm. In complex azomethine frequency observed at 2.97 δ ppm. (s,3H, N=C-CH₃) here also upfield shift in frequency observed in complex than ligand. In complex there are upfield shifts in NMR frequency than free ligand indicating formation of complex takes place [19].

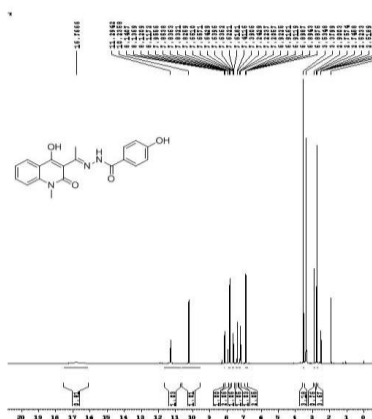


Fig.7 ^1H NMR of Ligand

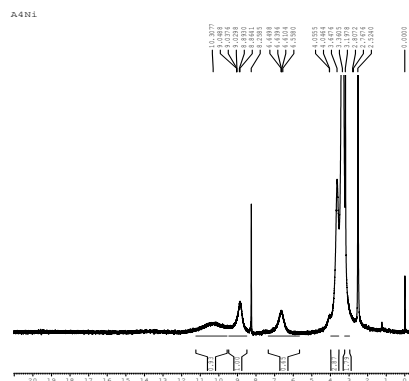


Fig.8 ^1H NMR of Cu(II) complex

3.5. Mass spectra of ligand and Cu(II) complex

Mass spectrum of the ligand and complexes supports its proposed formulation. It reveals the molecular ion peak m/z at 352.2, 353.1 a.m.u. due to (M+1) and (M+2) molecular ion peak, consistent with the molecular weight of the ligand. The mass spectra of the Cu(II) complex molecular ion peak observed at 766.07 m/z which is match with their calculated mass.

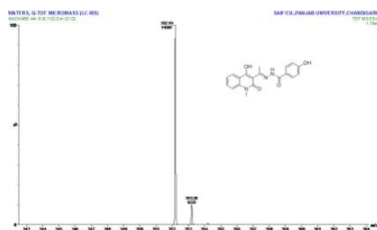


Fig.9 Mass spectra of ligand

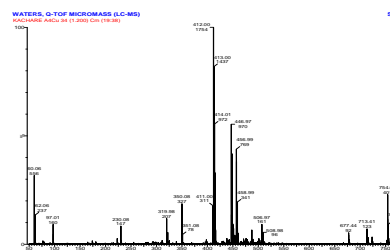


Fig.10 Mass spectra of Cu(II) complex

3.6. Thermogravimetric Studies

The simultaneous TG/DTA analysis of Cu(II) complex was studied

Cu(II) complex of (Fig. 11) decomposes in two steps. It is stable up to 230°C indicating absence of lattice as well as coordinated water molecules. The complex continues to decompose in a first step up to 350°C with weight loss of 58% (calc. wt. loss 58.9%) which is confirmed by sharp endothermic peak in DTA curve at 290.94°C may be due to the decomposition of non coordinated part of the complex. The second step covering the slow step decomposition of the reaction interval 350-750°C with 30% mass loss corresponds to decomposition of actual coordinated part of the complex and above which the residue attains almost constant weight due to formation of CuO as a final product.

The thermal behavior of copper complex in the present study indicates high thermal stability. Decomposition complex is started at relatively higher temperature (~230°C), finally giving a metal oxide residue. Thermogram of complex indicate the absence of coordinated as well as lattice water and exhibit higher thermal stability.

The thermal kinetic parameters ΔS , E_a and Z for non- isothermal decomposition of complex have been calculated by Horowitz-Metzer and Coats-Redfern method from TG-DTA curves (Fig.11) and are presented in **Table 3**.

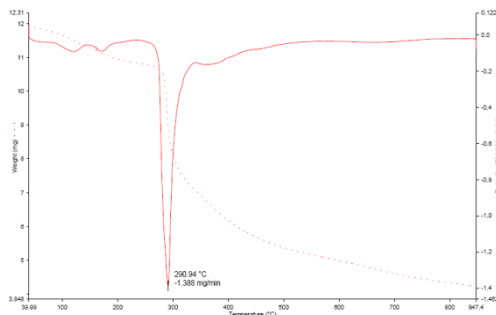


Fig. 11 TG-DTA Curve of Cu(II) Complex

Table 3. Thermodynamic and Kinetic Parameters

Metal complex	Method	Step	Decomp. Temp.	Order of Reaction	Ea(KJ mol ⁻¹)	ΔS(KJ mol ⁻¹)	ΔG(KJ mol ⁻¹)	Z (S ⁻¹)	Correlation Coefficient (r)
Cu(II)	H-M C-R	I	320	0.55	7.43	-162.68	19.62	41425.71	0.997
					6.82	-161.7	19.54	46628.83	0.999
	H-M C-R	II	440	0.55	9.68	-171.11	36.39	31330.3	0.999
					9.95	-170.76	36.33	32685.77	0.997

Generally, with decreasing value of ΔE, the value of Z increases, and higher value of activation energy suggest higher stability [20]. In the present complex, the value of E_a decrease with the increasing value of (Z) i.e. frequency factor indicating that the activated complex have more ordered or more rigid structure than the reactants or intermediate and that the reactions are slower than normal

3.7. X-RAY DIFFRACTION STUDY

The X-ray diffractogram, of Cu(II) complex was scanned in the range 0-60° at wavelength 1.54 Å. The X-ray diffraction pattern of the complex with respect to major peaks having relative intensity greater than 10% have been indexed by using computer program [21]. The above indexing program gives hkl planes, unit cell parameters and volume of the unit cell. The diffractogram and associated data gives 2θ values for each peak, relative intensity and inter planer spacing (d-values). On the basis of X-ray diffraction analysis the standard deviation observed was within the permissible range. The observed density is 0.8251 gcm⁻³ and calculated density is 0.8242 gcm⁻³. The Cu(II) complex is triclinic lattice type having one atom per unit cell. For this complex lattice parameters are a=22.35 Å, b=9.01 Å, c=8.2 Å, α=95°, β=110°, γ= 91.125°, V=1543 Å³. Which satisfies the condition a ≠ b ≠ c and α ≠ β ≠ γ [22].

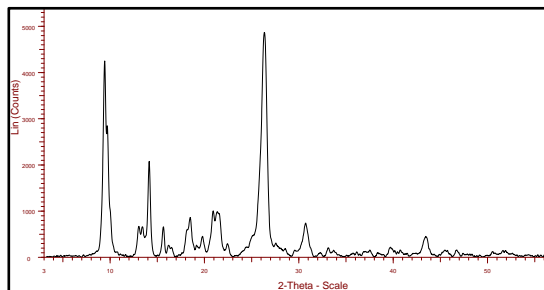


Fig. 12 X-ray Diffractogram of Cu(II) complex

IV. BIOLOGICAL ACTIVITY OF THE COMPOUNDS

4.1. In vitro antibacterial activity of the compounds

The antimicrobial activity of the ligand and complex were tested against the standard microbial strains, *Escherishia coli*, *Salmonella typhi*, *staphylococcus aurus*, *Bacillus substilis* by agar cup method at fixed concentration of 1% in

DMSO. The test was performed on nutrient agar Cup of 10 mm diameter were bored in the agar plate with sterile cork borer. All solutions were prepared in DMSO(1%) was add on cup, One cup for DMSO as blank and other for standard reference penicillium was also placed on the seeded nutrient agar. Then the plates were shifted to incubator at 37°C and incubated for 24 hours. Activity measured in diameter (mm). The results obtained are presented in **Table 4**

Inspection of the data revealed that complex and ligand lack the activity towards the Gram-negative bacteria *E. coli* and *S. typhi*. On the other hand, ligand and complex shows activity against Gram-positive bacteria *S. aureus* and *B. subtilis*.

Table 4: Report for Antibacterial testing.

Medium - Nutrient Agar Dose of compound - 1%	Method- Agar cup method cup size - 10 mm			
compound	<i>Bacillus subtilis</i> (mm)	<i>Staphylococcus aureus</i> (mm)	<i>Escherichia coli</i> (mm)	<i>Salmonella typhi</i> (mm)
Ligand(L)	14	12	--	--
(CuL ₂)	25	21	--	--
Penicillium	14 mm	20 mm	36 mm	28 mm

4.2. In vitro antifungal activity of the compounds

Compound were screened in vitro against *Aspergillus niger*, *Penicillium chrysogenum*, *fusarium moniliforme*, *Aspergillus flavus*, by poison plate method with potato dextrose agar media. The compound were tested at the 1% concentration in DMSO and compared with control.

Grisofulvin was prepared as standard reference plate. The fungal suspension was spot inoculated on the plates prepared using compound with nicrome wire loop. The plates were incubated at room temperature for 48 hours. The results obtained are presented in **Table 5**. Ligand does not show antifungal activity but its complex shows appreciable activity. Antifungal activity of complex increased several times on being coordinated with metal ions. Cu(II) complex shows more than 90% reduction of fungal growth for all fungi.

Table 5 : Report for Antifungal testing

Compound	<i>Aspergillus niger</i>	<i>Penicillium chrysogenum</i>	<i>Fusarium Moniliforme</i>	<i>Aspergillus flavus</i>
Ligand	+ve	+ve	+ve	+ve
(CuL ₂)	RG	-ve	-ve	-ve
Grisofulvin	-ve	-ve	-ve	-ve

Legends- +ve - Growth -(Antifungal Activity absent)
 -ve - No growth (More than 90 % reduction in growth Antifungal activity present)
 RG - Reduced Growth.

V. CONCLUSION

In the light of above discussion we have proposed octahedral geometry for both complex. On the basis of physicochemical and spectral data discussed above, one can assume that the ligand behaves as, ONO tridentate, coordinating via quinolone carbonyl, azomethine nitrogen and amide oxygen in complex. The mass spectra of ligand and its metal complex are in great accordance with calculated and observed. Thermogravimetric studies of Cu(II) complex revealed that complex is rigid and stable. p-XRD study of Cu(II) complex shows triclinic lattice type. The complex is biologically active and having greater activity compared to free ligand.

VI. ACKNOWLEDGEMENTS

The authors gratefully thanks the staff of department of chemistry Dr. B.A.M.U. Aurangabad. Further, the authors thank CSIR New Delhi for providing JRF fellowship.



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