

Microwave Synthesis, Spectral, Thermal and Antimicrobial Studies of Some Ni(II) and Cu(II) Schiff Base Complexes

A. P. Mishra*, Neha Sharma, Rajendra K. Jain

Synthetic Inorganic & Coordination Chemistry Laboratories,
Department of Chemistry, Dr. H. S. Gour Central University, Sagar, India
Email: *apm19@rediffmail.com, jainrajchem@gmail.com

Received November 30, 2012; revised January 6, 2013; accepted January 24, 2013

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ABSTRACT

Schiff bases have been synthesized by condensing nicotinamide with methyl isobutyl ketone (MKN) and 2-hydroxy acetophenone (HAN). Metal complexes have been prepared by interacting these Schiff bases with metal ions viz. Ni(II), Cu(II) 1:2 (metal:ligand) ratio. These compounds have been synthesized by conventional as well as microwave methods and characterized by elemental analysis, FT-IR, UV-Vis, FAB-mass, ESR, molar conductance, and thermal analysis. FAB mass and thermal data show degradation pattern of the complexes. The complexes are colored and stable in air at room temperature. The structure of the ligands were elucidated by spectral studies which indicate the presence of two or three coordinating groups in ligands which may be oxygen atom of the phenolic -OH group, the nitrogen atom of the azomethine (C=N) group and the oxygen atom of the carbonyl group. The thermal behavior of metal complexes shows that the hydrated complexes loses water molecules of hydration in the first step; followed by decomposition of ligand molecules in the subsequent steps. The solid state electrical conductivity of the metal complexes has also been measured. Solid state electrical conductivity studies reflect semiconducting nature of the complexes. The Schiff bases and metal complexes show good activity against the Gram-positive bacteria; *Staphylococcus aureus* and Gram-negative bacteria; *Escherichia coli* and fungi *Aspergillus niger* and *Candida albicans*. The antimicrobial results also indicate that the metal complexes are better antimicrobial agents as compared to the Schiff bases.

Keywords: Microwave Synthesis; Schiff Base; Thermal Study; Biological Activity

1. Introduction

Schiff bases and their bio-active complexes have been studied extensively over the past decade. Schiff bases provide potential sites for bio-chemically active compounds. The most widely studied metal in this respect is copper(II) which has proved to be beneficial in diseases such as tuberculosis, gastric ulcers, rheumatoid arthritis and cancers [1-4].

Microwave irradiated reactions under solvent free or less solvent conditions are attractive offering reduced pollution, low cost and offer high yields together with simplicity in processing and handling [5-7].

The present investigation aims at the conventional and microwave synthesis, physico-chemical characterization and bio-inorganic studies of Schiff bases involving methyl isobutyl ketone and 2-hydroxy acetophenone with nicotinamide and their metal chelates with Ni(II) and Cu(II).

2. Experimental

All the used chemicals and metal salts were of A.R. grade. Methyl isobutyl ketone and 2-hydroxy acetophenone were obtained from CDH and nicotinamide have been purchased from Sigma-Aldrich. Metal salts were purchased from Loba Chemie. Elemental analyses were performed on an Elemental Vario EL III Carlo Erba 1108 analyzer. FAB-mass spectra were recorded on a JEOL SX 102/DA 6000 Mass Spectrometer using argon/xenon (6 kV, 10 mA) as the FAB gas. The accelerating voltage was 10 kV and the spectra were recorded at room temperature. Electronic spectra (in DMSO) were recorded on Perkin Elmer Lambda-2B-spectrophotometer. Molar conductance measurements were conducted using 10^{-3} M solutions of the complexes in DMSO on Elico-CM 82 Conductivity Bridge at room temperature. Magnetic susceptibility measurements were carried out on a Gouy balance at room temperature using $\text{Hg}[\text{Co}(\text{SCN})_4]$ as the calibrant. FT-IR spectra were recorded in KBr pellets on a Perkin Elmer

*Corresponding author.

RX1 spectrophotometer in wave number region 4000 - 400 cm^{-1} . X-band EPR spectra were recorded on a Varian E-112 spectrometer at room temperature operating at the X-band region with 100 kHz modulation frequency, 5 mw microwave power and 1 G modulation amplitude using TCNE as the internal standard. Thermogravimetric (TG) analysis was done on Mettler Toledo Star Thermal Analyzer at NIPER, Chandigarh. The solid state electrical conductivity has been measured by impedance spectroscopic method using HIOKI 3532 - 50 LCR Hitester at fixed frequency 1 KHz in the temperature range of 297 - 377 K. Microwave assisted synthesis were carried out in open glass vessel on a modified microwave oven model 2001 ETB with rotating tray and a power source 230 V, microwave energy output 800 W and microwave frequency 2450 MHz. A thermocouple device was used to monitor the temperature inside the vessel of the microwave. The microwave reactions were performed using on/off cycling to control the temperature.

2.1. Conventional Method for the Synthesis of Schiff Bases

MKN and HAN Schiff bases (**Figure 1**) were synthesized by the condensation of equimolar ratio of methyl isobutyl ketone and 2-hydroxy acetophenone with nicotinamide dissolved in ethanol. The resulting reaction mixture was stirred well, refluxed for 3 - 4 h and then allowed to cool overnight. The coloured solid precipitate of Schiff base obtained was filtered, washed with cold ethanol several times and dried in air at room temperature and finally stored under reduced pressure in a CaCl_2 desiccator. The purity of synthesized compounds was checked by TLC using silica gel G (yield: 76% - 79%).

2.2. Microwave Method for the Synthesis of Schiff Bases

The equimolar (1:1) ratio of methyl isobutyl ketone and 2-hydroxy acetophenone with nicotinamide were mixed thoroughly in a grinder. The reaction mixture was then irradiated by the microwave oven by taking 3 - 4 mL of dry ethanol as a solvent. The reaction was completed in a short time (4 - 5 min) with higher yields. The resulting product was then recrystallized with ethanol, finally dried under reduced pressure over anhydrous CaCl_2 in a desiccator. The progress of the reaction, purity of the product was monitored by TLC using silica gel G (yield: 87% - 89%).

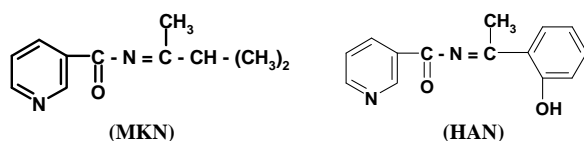


Figure 1. Structure of schiff base ligands.

2.3. Conventional Method for the Synthesis of Metal Complexes

The metal complexes were prepared by the mixing of (50 mL) ethanolic solution of $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ / $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ with the (50 mL) ethanolic solution of Schiff bases (MKN/HAN) in 1:2 (metal:ligand) ratio. The resulting mixture was refluxed on water bath for 6 - 8 h. A coloured product appeared on standing and cooling the above solution. The precipitated complex was, filtered washed with ether and recrystallized with ethanol several times and dried under the reduced pressure over anhydrous CaCl_2 in a desiccator. It was further dried in electric oven at 50°C - 70°C (yield: 60% - 71%).

2.4. Microwave Method for the Synthesis of Metal Complexes

The ligand and the metal salts were mixed in 1:2 (metal: ligand) ratio in a grinder. The reaction mixture was then irradiated by the microwave oven by taking 3 - 4 mL of dry ethanol as a solvent. The reaction was completed in a short time (6 - 9 min) with higher yields. The resulting product was then recrystallized with ethanol and ether and finally dried under reduced pressure over anhydrous CaCl_2 in a desiccator. The progress of the reaction and purity of the product was monitored by TLC using silica gel G (yield: 80% - 85%).

2.5. Biological Activity

The *in-vitro* biological activity of the Schiff bases and their complexes was tested against the bacteria *Escherichia coli* and *Staphylococcus aureus* by disc diffusion method using nutrient agar as medium and streptomycin as control. The antifungal activities of the compounds were also tested by the Well diffusion method against the fungi *Aspergillus niger* and *Candida albicans*, on potato dextrose agar as the medium and miconazole as control. Each of the compounds was dissolved in DMSO and solutions of the concentrations (25, 50 and 100 ppm) were prepared separately. In a typical procedure, a well was made on agar medium inoculated with microorganism. The well was filled with the test solution using a micropipette and the plate was incubated 24 h for bacteria at 37°C and 72 h for fungi at 30°C . During this period, the test solution diffused and the growth of the inoculated microorganism was affected. The inhibition zone was developed, at which the concentration was noted.

3. Results and Discussion

As a result of microwave-assisted synthesis, it was observed that the reaction was completed in a short time with higher yields compared to the conventional method.

In the microwave method homogeneity of reaction mixture was increased by the rotating of reaction platform tray. The confirming of the results was also checked by the repeating of the synthesis process.

All the metal complexes are coloured, solid and stable towards air and moisture at room temperature. They decompose on heating at high temperature, more or less soluble in common organic solvents. The comparison study data of microwave and conventional methods, with analytical and physical data of the compounds are given in the **Table 1**. Analytical data show that metal chelates have 1:2 (metal:ligand) stoichiometry. The observed molar conductance of the complexes in DMSO at room temperature is consistent with electrolytic nature of the complexes of MKN ligand while the complexes of HAN show non-electrolytic nature.

3.1. FAB-Mass Spectrum

The FAB-mass spectra suggested that all the complexes have a monomeric nature. These complexes show molecular ion peaks in good agreement with the empirical formula suggested by elemental analyses.

The FAB-mass spectrum of $[\text{Cu}(\text{MKN})_2(\text{H}_2\text{O})_2]\text{Cl}_2$ complex has been studied as one of the representative case. The peak of appreciable intensity has been observed at m/z value 550, 476, 460, 450, 256, 197 and 91, suggesting the fragmentation pattern. The m/z value 550 corresponds to nearest composition: 550 to $[\text{Cu}(\text{MKN})_2(\text{H}_2\text{O})_2]\text{Cl}_2$, 476 to $[\text{Cu}(\text{MKN})_2(\text{H}_2\text{O})_2]$, 460 to $[\text{Cu}(\text{MKN})_2(\text{H}_2\text{O})]$, 450 to $[\text{Cu}(\text{MKN})_2]$, 256 to $[\text{Cu}]/n(\text{MKN})_2$, 190 to ligand alone and 91 to copper with chelated O and N ligand moiety [8,9].

3.2. IR Spectra

The IR spectra of the complexes were compared with

those of the free ligand in order to determine the involvement of coordination sites in chelation. Characteristic peaks in the spectra of the ligand and complexes were considered and compared.

The IR spectrum of the Schiff base ligand (MKN) exhibits strong band at 1620 cm^{-1} due to (azomethine) ν (C=N) group. It shifts down by $10 - 15\text{ cm}^{-1}$ in the complexes suggesting coordination through azomethine nitrogen, this can be explained by the donation of electrons from nitrogen to the empty d-orbital of the metal atom. A strong band at 1684 cm^{-1} in ligand due to ν (C=O) (amide I band) group has shifted to lower frequency by $10 - 15\text{ cm}^{-1}$ in the complexes. This indicates the involvement of C=O group in coordination. Practically, no shift in the characteristic band position of pyridine nitrogen ($-\text{N}=\text{}$) has been observed, indicating the non-involvement of this donor group in coordination. The IR spectra of complexes exhibit bands around $3460 \pm 10\text{ cm}^{-1}$ and $650 \pm 5\text{ cm}^{-1}$ assignable to coordinate water molecule. The new bands at $544 \pm 6\text{ cm}^{-1}$ and $480 \pm 5\text{ cm}^{-1}$ in the complexes have tentatively been assigned to ν (M-O) and ν (M-N) vibrations in the complexes.

The IR spectra of Schiff base ligand (HAN) shows the band at 1683 cm^{-1} due to ν (C=O) (amide I band) group, it has shifted towards lower frequency $1668 \pm 10\text{ cm}^{-1}$ in the complexes. This indicates the involvement of C=O group in coordination. Characteristic band at about 1618 cm^{-1} due to ν (C=N) azomethine group has shifted towards lower frequency $1600 \pm 5\text{ cm}^{-1}$ in the complexes, suggesting coordination through azomethine nitrogen. A band in ligand spectrum at 1350 cm^{-1} due to phenolic OH (deformation) was found absent in the complexes. A medium intensity ligand band observed at about 1202 cm^{-1} (phenolic C-O) has shifted higher and appeared at $1220 \pm 10\text{ cm}^{-1}$ in the complexes. This suggests the deprotonation of phenolic OH after its chelation with metal ion.

Table 1. The comparative results of conventional and microwave methods, analytical, physical data and magnetic moment values of the compounds.

Compounds molecular weight/Colour	Reaction period		Yield (%)		Elemental analysis, found (calcd.) %			$^*\Lambda_m$	$^{\#}\mu_{\text{eff}}$
	CM (hs)	MM (min.)	CM	MM	C	H	N		
MKN 190.0/ Pinkish Creem	4.0	4.9	76	87	69.49 (69.47)	7.28 (7.36)	14.69 (14.73)	-	-
$[\text{Ni}(\text{MKN})_2(\text{H}_2\text{O})_2]\text{Cl}_2$ 545.7/Light Green	7.9	8.9	67	80	48.45 (48.37)	5.75 (5.86)	12.86 (10.26)	125.0	3.09
$[\text{Cu}(\text{MKN})_2(\text{H}_2\text{O})_2]\text{Cl}_2$ 550.5/Sky Blue	7.2	8.2	60	83	47.85 (47.95)	5.78 (5.81)	12.86 (10.17)	110.0	1.92
HAN 240.0/Cream	3.2	4.6	79	89	70.12 (70.00)	5.3 (5.0)	11.45 (11.66)	-	-
$[\text{Ni}(\text{HAN})_2]$ 538.7/Light Green	7.0	8.1	71	84	62.47 (62.37)	4.25 (4.45)	14.39 (10.39)	13.9	3.10
$[\text{Cu}(\text{HAN})_2]$ 543.5/Sky Blue	6.4	6.9	67	85	61.80 (61.82)	4.30 (4.41)	14.39 (10.30)	17.3	1.89

CM = Conventional method, time in hours; MM = Microwave method, time in minutes. $^*\Lambda_m = (\Omega^{-1}\text{ cm}^2\text{ mol}^{-1})$; $^{\#}\mu_{\text{eff}} = \text{B.M.}$

Practically, no shift in the characteristic band position of pyridine nitrogen (-N=) was observed. This indicates the non-involvement of this donor group in coordination. The new bands at $579 \pm 10 \text{ cm}^{-1}$ and $482 \pm 10 \text{ cm}^{-1}$ in the complexes have tentatively been assigned to ν (M-O) and ν (M-N) respectively [10-12].

3.3. Electronic Spectra and Magnetic Moment

The nature of the ligand field around the metal ion has been deduced from the electronic spectra.

The electronic spectrum of Ni(II) complex of MKN shows three bands at 12,406, 22,330, 27,010 cm^{-1} which are assignable to ${}^3\text{A}_{2g} \rightarrow {}^3\text{T}_{2g}$ (F) (ν_1), ${}^3\text{A}_{2g} \rightarrow {}^3\text{T}_{1g}$ (F) (ν_2) and ${}^3\text{A}_{2g} \rightarrow {}^3\text{T}_{1g}$ (P) (ν_3) transition, respectively. The values of various ligand field parameters, 10 Dq, B, β , ν_2/ν_1 , λ and LFSE are as 12,406 cm^{-1} , 808 cm^{-1} , 0.74, 1.8, (-)284 cm^{-1} and 177.8 $\text{kJ}\cdot\text{mol}^{-1}$, respectively. The value of magnetic moment for this complex is 3.09 B.M. These findings are in favor of an octahedral geometry for the Ni(II) complex. In the spectrum of Cu(II) complex a single broad band at 12,314 – 16,570 cm^{-1} has been observed; which is attributable to ${}^2\text{E}_g \rightarrow {}^2\text{T}_{2g}$ transition. The various ligand field parameters viz. 10 Dq, and λ and LFSE have been calculated for this complex and values are; 12314 cm^{-1} , (-) 678 cm^{-1} and 86.05 $\text{kJ}\cdot\text{mol}^{-1}$ respectively. The value of its magnetic moment is 1.92 B.M, hence a distorted octahedral geometry has been suggested for this Cu(II) complex.

The electronic spectrum of Ni(II) complex of HAN shows three bands at 12,658, 22,734, 26,890 cm^{-1} corresponding to transitions ${}^3\text{A}_{2g} \rightarrow {}^3\text{T}_{2g}$ (F) (ν_1), ${}^3\text{A}_{2g} \rightarrow {}^3\text{T}_{1g}$ (F) (ν_2) and ${}^3\text{A}_{2g} \rightarrow {}^3\text{T}_{1g}$ (P) (ν_3) respectively. The value of various ligand field parameter 10 Dq, B, β , ν_2/ν_1 , λ and LFSE are as 12,658 cm^{-1} , 780 cm^{-1} , 0.72, 1.7, (-) 301 cm^{-1} and 181.4 $\text{kJ}\cdot\text{mol}^{-1}$ respectively. The value of magnetic moment for this complex is 3.10 B.M. These findings are in favor of an octahedral geometry for this Ni(II) complex. In spectrum of the copper (II) complex exhibits a single broad band at 12,500 - 13,300 cm^{-1} which is attributable to ${}^2\text{E}_g - {}^2\text{T}_{2g}$ transition. The various ligand field parameters viz 10 Dq, λ and LFSE have been calculated and values are 12,500 cm^{-1} , (-) 578 cm^{-1} and 89.60 $\text{kJ}\cdot\text{mol}^{-1}$ respectively. The value of its magnetic moment is 1.89 B.M. Thus, an octahedral geometry has been suggested for this complex [13-16].

3.4. ESR Spectra

The ESR spectra of Cu(II) provide information about the extent of the delocalization of unpaired electron. The X-band ESR spectra of Cu(II) complexes were recorded in the solid state at room temperature, their g_{\parallel} , g_{\perp} , Δg , g_{av} , G have been calculated. The values of ESR parameters g_{\parallel} , g_{\perp} , g_{av} , Δg , G for Cu(II) complex of MKN are 2.2283, 2.0727, 2.1247, 0.1556, 3.2006 respectively.

ESR spectra of the complexes revealed two g values (g_{\parallel} and g_{\perp}). Since the g_{\parallel} and g_{\perp} values are closer to 2 and $g_{\parallel} > g_{\perp}$ suggesting a tetragonal distortion around the Cu(II) ion. The trend $g_{\parallel} > g_{\perp} > g_e$ (2.0023) shows that the unpaired electron is localized in $d_{x^2-y^2}$ orbital in the ground state of Cu(II), spectra are characteristic of axial symmetry. The $g_{\parallel} > 2.3$ is characteristic of an ionic environment and $g_{\parallel} < 2.3$ indicates a covalent environment in metal ligand bonding. The g_{\parallel} values for these complexes are less than 2.3 suggesting the environment is covalent. The exchange coupling interaction between two Cu(II) ions is explained by Hathaway expression $G = (g_{\parallel} - 2.0023)/(g_{\perp} - 2.0023)$. According to Hathaway, if the value G is greater than four ($G > 4.0$), the exchange interaction is negligible; whereas when the value of G is less than four ($G < 4.0$) a considerable exchange coupling is present in solid complex. The G values for these Cu(II) complexes are less than four indicate, considerable exchange interaction in the complexes [17,18].

3.5. Thermal Studies

The thermal behavior of metal complexes shows that the hydrated complexes lose molecules of hydration first; followed by decomposition of ligand molecules in the subsequent steps.

Thermogram of Cu(II) Complex of MKN shows that the weight loss corresponding to two coordinated water molecules occurs between 180°C - 240°C (Remaining wt% obs./cal. 93/93.47). On increasing the temperature above 240°C, a weight loss at faster rate occurs up to 350°C. This corresponds with the decomposition of the ligand moiety. A horizontal zone beyond 350°C suggests the formation of ultimate pyrolysis product (Remaining wt% obs./cal 34/29.89).

Thermogram of Ni(II) Complex of HAN shows that the compound is stable upto 270°C. Above this temperature, a weight loss has been observed in general up to 550°C, indicating decomposition of the ligand in two steps. The observed weight loss between 280°C - 410°C may be due to partial decomposition of ligand part from the complex (Remaining wt% obs./cal 39/36.89). Above 410°C, an inflection occurs in the curve and loss in weight goes up to 510°C. Above this temperature horizontal curve shows no further loss in weight. The metal oxide has been inferred to be the final product (Remaining wt% obs./cal 21/18.33) [19,20].

3.6. Electrical Conductivity

The temperature dependence of the solid state conductivity (σ) of the compounds in their compressed pellet form has been measured at fixed frequency 1 KHz in the temperature range 297 - 377 K. the values of the solid state electrical conductivity of the Schiff base and its complexes increases with increasing temperature and de-

creases upon cooling over the studies temperature range indicating their semiconducting behavior. The general behavior of electrical conductivity follows the Arrhenius Equation:

$$\sigma = \sigma_0 \exp(-Ea/kT)$$

where Ea is the thermal activation energy of conduction, σ_0 is the conductivity constant and k is the Boltzman constant. The plots of σ vs $1000/T$ for all the compounds are found to be linear over a studies temperature range. The room temperature electrical conductivity of all the compounds lies in the range 6.45×10^{-7} - 2.15×10^{-8} ohm⁻¹ cm⁻¹. These values show their semi-conducting nature. The electrical conductivity at room temperature for the complexes of MKN are Cu > Ni and for the metal complexes of HAN are Cu > Ni. The activation energy of the compound lies in the range 0.232 - 0.771 eV [21]. The confirming of the temperature dependence conductivity of the compounds was also checked by the repeating of the conductivity measurements.

3.7. Antimicrobial Activities

The *in-vitro* Antimicrobial activity of the synthesized Schiff base ligands and their corresponding metal complexes on selected bacteria *E. coli* and *S. aureus* and two fungi *A. niger* and *C. albicans* was carried out. All of the tested compounds showed good biological activity against microorganism. On comparing the biological activity of the Schiff base and its metal complexes with the standard bactericide and fungicide, it is show that the some metal complexes have good activity as compared to the standard but all the complexes are more active than their respective ligands. The higher inhibition zone of metal complexes than those of the ligands can be explained on the basis of Overtone's concept and Chelation

theory. On chelation, the polarity of the metal ion will be reduced to greater extent due to the overlap of the ligand orbital and partial sharing of the positive charge of the metal ion with donor groups. Further, it increases the delocalization of π -electrons over the whole chelating ring and enhances the penetration of the complexes into lipid membranes and blocking of the metal binding sites in the enzymes of microorganisms. There are other factors which also increases the activity are solubility, conductivity and bond length between the metal and ligand [22-25].

The bactericidal and fungicidal investigation data of the compounds are summarized in **Tables 2** and **3**. The results of the investigations account for the antipathogenic behavior of the compounds and this efficacy is positively modified on complexation.

4. Conclusion

In the present research studies, our successful efforts are synthesis of some newly compounds from the conventional as well as microwave methods. These synthesized compounds Characterized by various physicochemical and spectral analyses. In the result of microwave-assisted synthesis, it has been observed that the reaction time decreased from hours to minutes and availability of the product within better yields compared to the classical method. FAB-mass and thermal data shows degradation pattern of the complexes. Thermogravimetric studied of the complexes also helped to characterize of the complexes. Electrical conductivity data suggest that all the complexes fall in the semiconducting range. The antimicrobial data show that the metal complexes to be more biological active compared to those parent Schiff base ligand against all pathogenic species. The compounds also inhibit the growth of fungi and bacteria to a greater

Table 2. Antibacterial screening data for the ligands and their complexes.

Comp.	<i>E. coli</i>						<i>S. aureus</i>					
	Diameter of inhibition zone (mm)			% Activity index			Diameter of inhibition zone (mm)			% Activity index		
	25	50	100	25	50	100	25	50	100	25	50	100
MKN	12	14	17	55	58	61	11	14	19	61	64	79
Ni(II)	16	20	24	73	83	86	13	16	19	72	73	79
Cu(II)	17	20	25	77	83	89	15	19	21	83	86	88
HAN	15	17	21	68	71	75	13	17	19	72	77	80
Ni(II)	15	17	20	68	71	71	13	16	18	72	73	75
Cu(II)	18	22	26	82	92	93	15	19	21	83	86	88
Streptomycin (Standard)	22	24	28	100	100	100	18	22	24	100	100	100

$$\% \text{ Activity index} = \frac{\text{Zone of inhibition by test compound (diameter)}}{\text{Zone of inhibition by standard (diameter)}} \times 100 .$$

Table 3. Antifungal screening data for the ligands and their complexes.

Compound	Diameter of inhibition zone (mm); Concentration in ppm					
	<i>A. niger</i>			<i>C. albicans</i>		
	25	50	100	25	50	100
MKN	12	15	20	11	15	20
Ni(II)	14	18	25	16	18	21
Cu(II)	18	22	28	19	22	27
HAN	14	18	21	13	16	19
Ni(II)	16	18	23	16	19	22
Cu(II)	18	20	25	17	19	23
Miconazole (Standard)	20	24	30	22	24	29

extent as the concentration is increased. The Schiff base ligands were found to be biologically active and their metal complexes displayed enhanced antimicrobial activity against one or two strains. Chelation tends to make the ligand act as more powerful and potent bactericidal agent. Further chelation can help in MDR problems.

5. Acknowledgements

We are thankful to I. I. T. Mumbai for ESR analysis. We also acknowledge SAIF, CDRI Lucknow for micro analysis and spectral analyses. Thanks are also due to the Head, Department of Chemistry, Botany and Physics, Dr. Hari Singh Gour University, Sagar (M.P.) for providing Laboratory facilities.

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