



Synthesis and Characterization of Some Transition Metal Complexes of Schiff Base Derived From 2, 4 – Dihydroxybenzaldehyde

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Abstract

New N_2O_2 type Schiff base has been designed and synthesized by condensing 2, 4 dihydroxy benzaldehyde and α -naphthylamine in ethanol. Solid metal complexes of the schiff base with Cu(II), Ni(II) and Zn(II) metal ion were synthesized and characterized by elemental analysis, magnetic susceptibility, molar conductance, IR, UV-Vis and 1H NMR spectral studies. The data shows that the complexes have the composition of ML type. The UV-Visible and magnetic susceptibility data of the complexes suggest a square-planar geometry around the central metal ion except Zn (II) complex which has tetrahedral geometry. The redox behaviour of the complexes was studied by Cyclic Voltammetry. The in vitro activities of the compounds were tested against Staphylococcus, Bacillus subtilis, Escherichia Coli and Pseudomonas by well-diffusion method and antifungal of the compound were tested against fungi such as Aspergillus niger, Aspergillus flavus and Rhizoctonia bataicola by minimum inhibitory concentration. All the metal complexes showed stronger antifungal and antimicrobial activities than the free ligand.

Keywords: Schiff base, 2, 4 - dihydroxy benzaldehyde , α -Naphthylamine, Antifungal and Antimicrobial activity.

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1. Introduction

Recently transition metal complexes with salen ligand derived from the condensation of 2, 4 dihydroxy benzaldehyde with amine have been widely studied. The metal complexes have been attractive area of research, in view of their significance as biomimetic catalysts in the process of oxygenation [1-5]. Schiff bases have been widely used as ligands because of

high stability of the coordination compounds and their good solubility in common solvents. The π -system in Schiff bases often imposes a geometrical constriction and affects the electronic structure as well. Thermocouple chemical properties of Schiff bases have attracted much researcher attention in view of their ability to coordinate metal ions, acting as monodentate, bidentate or tetradentate ligands in metal chelates involving a NO or N₂O₂-Schiff's base donor atom sets. These schiff base metal derivatives have considerable interest due to their role as model complexes to biological systems, contributing the knowledge of their structure and behavior [6-7]. Due to chelating nature of azomethine nitrogen atom of Schiff bases, these are well recognized as antibacterial, antifungal, anti tumor, antituberculosis, anticancer, DNA binding and cleaving agents [8]. Because of excellent complexation ability with different metal ions Schiff base ligands have been employed intensively in analytical chemistry. Transition metals are most suitable candidates for the synthesis of cancer combating nonradioactive tools for chemotherapy and diagnosis. Co-ordination chemistry of metal complexes has become of increasing significance in the last few years due to their wide variety of application in supramolecular photochemistry and in medicine [9]. Now we report the structural and synthetic investigation of another set of related Schiff base ligands and their complexes with Cu(II), Ni(II) and Zn(II) ions.

2. Materials and Methods

The chemicals and solvents were purified by standard methods. Elemental analyses were carried out using a Perkin Elmer 2400II elemental analyzer. Molar conductance of the complexes was measured in DMF at room temperature using a systronic conductivity bridge 304. Magnetic susceptibility of the complexes was performed on a Sherwood MSB Mark 1 Gouy balance. Infrared spectral studies were carried out on a shimadzu FT IR 8000 spectrometer using KBR disc. UV-visible spectra were obtained using thermo Spectronic 6 Hexios. NMR spectra were recorded on Bruker DRX-300, 300 MHz NMR spectrometer using TMS as reference. EPR spectra of the Cu (II) complex was recorded in Varian E-112 machine. Cyclic Voltammetric measurements for Cu(II) complex in DMF were carried out on electro chemical analyzer CH instruments using a three electrode cell containing Ag/AgCl reference electrode, Pt-wire auxiliary electrode and glassy carbon working electrode with tetrabutyl ammonium perchlorate as supporting electrode.

2.1 Synthesis of Schiff Base

Hot solution of α -Naphthylamine (0.01mmol, 1.43 g) in 50 ml ethanol mixture as 1:1 mole ratio mixed with hot solution of 2,4-dihydroxybenzaldehyde (0.01mmol, 1.38 g) in the same solvent and the reaction mixture was left under heating for 2 hrs [10]. A solid mass was separated and washed with ethanol. Crystallisation was done with ethanol and then dried over CaCl₂. The pale yellow ligand ; L

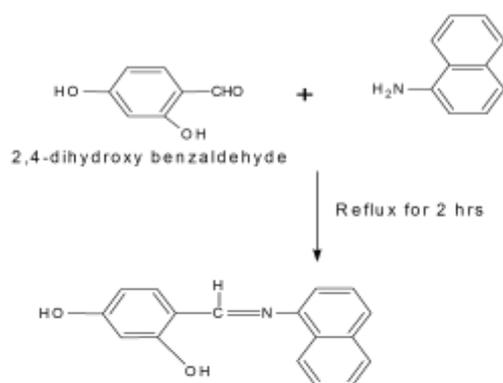


Figure 1. Synthesis of Schiff base Ligand (L).

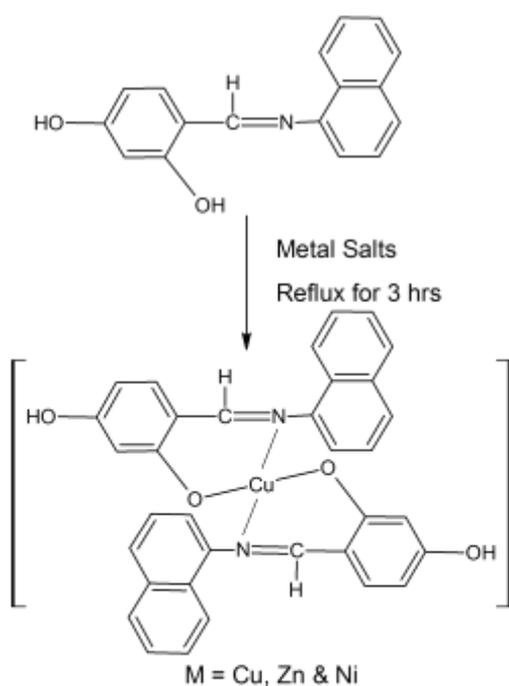


Figure 2. Synthesis of Schiff base Complex derived from L⁵.

(yield 82%) was collected. Synthetic pathway is shown in figure 1.

2.2. Synthesis of metal complexes

All the new complexes were prepared by adding hot ethanolic solution in a 1:1 molar ratio of $\text{Cu}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$, $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ and $\text{Ni}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ (1 mmol) to the hot solution of ligand (1 mmol) in the same

solvent. The solution was stirred with heating for one hour whereupon the complexes precipitated then filtered and left for drying. A solid residue was separated, washed and dried. Crystallization was done with methanol and complexes dried over anhydrous CaCl_2 . Synthesis of metal complex is shown in figure 2.

3. Results and Discussion

Physical characteristics and analysis of carbon, hydrogen and nitrogen contents of the complexes have been determined and listed in table 1. The results of C, H and N percentage are in accordance with the composition suggested for the ligand and their complexes. The obtained analytical data indicated the formation of mononuclear metal complexes.

3.1. Molar Conductance Measurements

The Cu (II), Ni (II) and Zn (II) complexes were dissolved in DMF and the molar conductivities of 10^{-3}M of their solutions were measured at room temperature. The conductance values of the metal complexes are non ionic in nature and non electrolytes. The complexes showed a lower molar conductivity values in the range between 13.54 and 14.33 $\text{ohm}^{-1} \text{cm}^2 \text{mol}^{-1}$ which are in good agreement with those indicated their non-electrolytic behavior. Our results are in good agreement with those reported earlier [11-16]. The data are listed in table 1.

3.2. Infrared Spectra

The IR stretching frequencies for the Schiff bases and their complexes are given in table 2.

Table 1. Analytical data for Schiff base metal complexes.

S.NO	COMPOUNDS	Calculated (Found) %			Metal	ΔM ($\text{Ohm}^{-1}\text{cm}^2$ mol^{-1})
		C	H	N		
1	L	77.56 (77.52)	4.94 (4.93)	5.32 (5.30)	-	-
2	Cu(L)	69.38 (69.34)	4.08 (4.09)	4.76 (4.75)	10.88 (10.86)	13.54
3	Zn(L)	69.26 (69.25)	4.07 (4.08)	4.75 (4.76)	11.03 (11.01)	14.33
4	Ni(L)	70.10 (70.12)	4.12 (4.13)	4.81 (4.82)	9.96 (9.98)	14.12

Table 2. Infrared spectral data for Schiff base metal complexes.

Compounds	C=N (cm^{-1})	C=C (cm^{-1})	M-N (cm^{-1})	M-O (cm^{-1})
L	1613	1527	-	-
CuL	1596	1487	448	518
ZnL	1595	1525	416	520
NiL	1594	1457	458	512

The IR spectra of the Schiff base ligand exhibited band in the region 3379 cm^{-1} which were due to stretching frequency of OH groups. The bands in the region 1457-1525 and 1594-1616 were assigned for the presence of C=C and C=N groups [17, 18] respectively. On complexation, it is well strategy to compare the spectra of the free ligand and its complex to account for the chelation site(s). Some positional Schiff have been observed for the above bands namely C=N and C=C in the complexes. New bands were emerged for the complexes which were not seen in the free ligand, these bands were assigned for M-O and M-N. Therefore it can be calculated that (L) ligand binds to the metal ions through

azomethine N and O. The data are listed in table 2.

3.3. Electronic Spectra and Magnetic Moment

The UV –Visible spectra are often very useful in the evaluation of results furnished by other methods of structural investigation. The electronic spectral measurements were used for assigning the stereochemistries of metal ions in the complexes based on the positions and number of d-d transition peaks. The electronic absorption spectra of the Schiff base ligand and its complexes were recorded in DMF solution in the range of 200 to 800 nm regions and the data are presented in table 3. The absorption spectrum of free ligand consist of intense bands centered at 314 nm attributed

Table 3. UV-Visible data of Schiff base metal complexes.

Compounds	Absorption (nm)			
	$\pi \rightarrow \pi^*$	$n \rightarrow \pi^*$	L→M (CT)	d-d
L	258	314	436	-
CuL	256	310	413	625
ZnL	274	322	424	-
NiL	269	358	415	512, 684

to $n \rightarrow \pi^*$ transition of the azomethine group. Another intense band in higher energy region of the spectra of the free ligand was related to $\pi \rightarrow \pi^*$ transition of benzene rings. These transitions are also found in the spectra of the complexes, but they shifted to lower frequencies, confirming the coordination of the ligands to the metal ions. Further, d-d transition of the complex showed a broad band centered at 625 nm for Cu (II) complex Fig.3. This is due to ${}^2B_{1g} \rightarrow {}^2A_{1g}$ transition [19]. The spectra of Ni(II) complex in the visible region at about 512 and 684 nm is assigned to ${}^1A_{1g} \rightarrow {}^2A_{1g}$ and ${}^1A_{1g} \rightarrow {}^1B_{1g}$ transitions, suggesting an approximate square planar geometry of the ligand around the metal ions [20]. Based on these data, a square planar geometry has been assigned to the complexes. These values are comparable with other reported complexes [21].

The magnetic susceptibility measurements have been carried out for Schiff base complexes. The magnetic properties depend on the ground and excited states of the metal complexes. The magnetic moment of the Schiff base Cu (II) complex at room temperature is 1.64 BM [22], corresponding to one unpaired electron. This indicates that the Cu(II) complex is monomeric in nature. The Ni(II) complex has diamagnetic properties

using Gouy balance. The observed magnetic moment also confirms the square planar environment of Ni (II) complex, its conformity with the fact that the known square planar complex of Ni (II) complex is diamagnetic [23].

3.4. Thermal Analysis

Thermal analysis was used to confirm the molecular structure of the complexes. Also, the thermal stability, properties, nature of intermediates and final products of thermal decomposition of coordination compounds can be obtained from thermal analysis [24]. From the TGA curves, the mass loss can be calculated for the single decomposition step and compared with those theoretically calculated for the suggested formula based on analytical and spectral results. The Cu(II) complex with the molecular formula $[CuC_{34}H_{24}N_2O_4]$ was thermally decomposed in a single step. The estimated mass loss of 43.82% [calculated mass loss =44.72%] within the temperature range may be attributed to the loss of aromatic ligand group and amine. The thermal decomposition of the Ni(II) complex with the molecular formula $[NiC_{34}H_{24}N_2O_4]$ proceeds with only one degradation step. The step occurs within the temperature range 210 - 560° with an estimated mass loss 44.57%

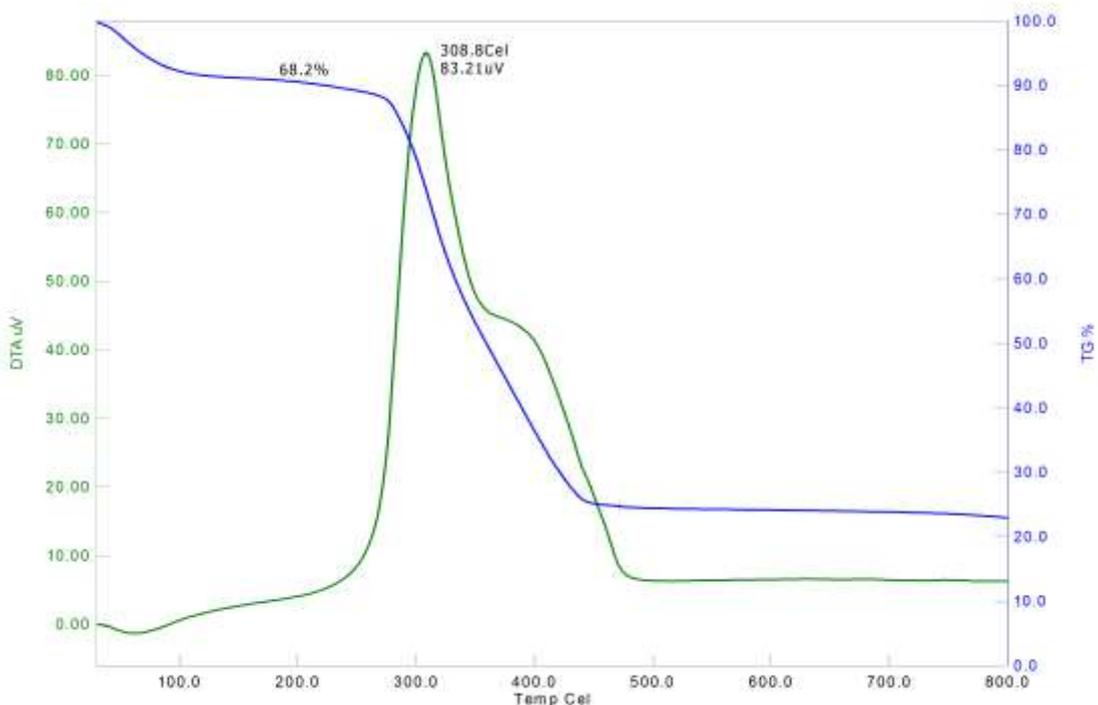


Figure 3. Thermal analysis of CuL.

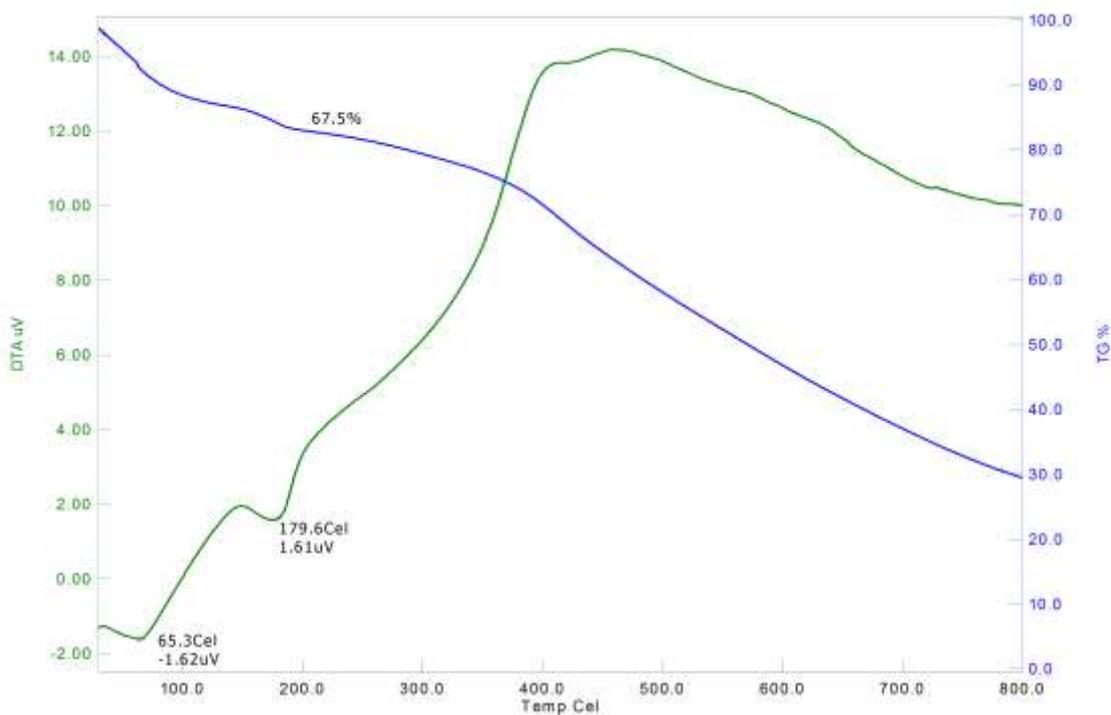


Figure 4. Thermal analysis of NiL⁵.

(calculated mass loss = 45.18%), which was reasonably accounted for the loss of aromatic ligand group and amine group. The last step

did not finish completely. Therefore, last decomposition residue was not determined.

Thermal analysis of [CuL₅] and [NiL₅] complexes are shown in figure 3 and 4.

3.5. ESR spectra

The ESR spectrum of the powdered sample of the Cu (II) complex was recorded at room temperature. The observed value for the Cu (II) complex is a $G=2.194$ [13]. The observed G value is less than 2.3 in agreement with the covalent character of the metal – ligand bond [25]. The trend $g_{\parallel} > g_{\perp} > g_e$ (2.0023) observed for this complex shows that the unpaired electron is localized in dx^2-y^2 orbital of Cu(II) ion and the spectral features are characteristics of axial symmetry; Square planar structure may be assumed for this Cu(II) complex. The anisotropic G values have been calculated by using the equation $G = (g_{\parallel} - 2.002) / (g_{\perp} - 2.002)$. If the G value is less than 4.0, the ligand forming the Cu (II) complex is regarded as strong field ligand while a value of G greater than 4.0 indicates weak field ligand. As $G = 2.194$ for the present complex indicates that the ligand [CuL] was strong field ligands and the metal – ligand bonding in this complex is [26].

3.6. ¹H-NMR

The ¹H-NMR spectrum of ligand [L] in DMSO solvent shows absorption in the region, a multiple one at δ 7.0 -8.0 ppm corresponding to deshielded Ar-H protons. The ¹H-NMR spectrum of the ligand [L] shows a multiple signals observed at 6.9 – 7.2 ppm that may be attributed to aromatic ring protons of naphthalene moiety, while the azomethine proton was observed in the range of 8.24 ppm.

The formation of azomethine group in Schiff base was confirmed by ¹H-NMR spectrum.

3.7. Cyclic Voltammogram

The redox behaviour of Cu (II) complex in DMF containing 10^{-1} M tetramethylammonium percholate was examined by means of cyclic voltammetry [potential range -2.0 to 1.2 V] with different scan rates from 25 to 125 mVs⁻¹. The cyclic voltammogram shows a well defined quasireversible peak for the redox couple Cu (II)/Cu (III) [$E_{p_a} = 0.34$ V]. In the negative potential range of copper shows irreversible cathodic peak at $E_{p_c} = -1.51$ V [Cu (II)/Cu (I)] with the scan rate of 100 mVs⁻¹. The limiting peak to peak separation (E_p) for Cu(III)/Cu(II) process is greater than 59 mV which revealed that the couple is quasireversible. The ratio of anodic and cathodic peak current value is 1:1 demonstrating the simple one-electron process. Further, the anodic peak is shifted towards positive potential value, and the cathodic peak is shifted towards negative potential with a formation of scan rate 25 to 125 mVs⁻¹ which supports quasireversible process. The cyclic voltammogram of the Cu (II) complex, showed two quasi - redox couples. The cyclic voltammetric technique also provides information about interaction between the metal complexes and DNA. Ni (II) and Zn(II) complexes are diamagnetic in nature and also Zn(II) does not have unpaired electron. Due to this, both Ni (II) and Zn (II) complexes do not show cyclic voltammogram.

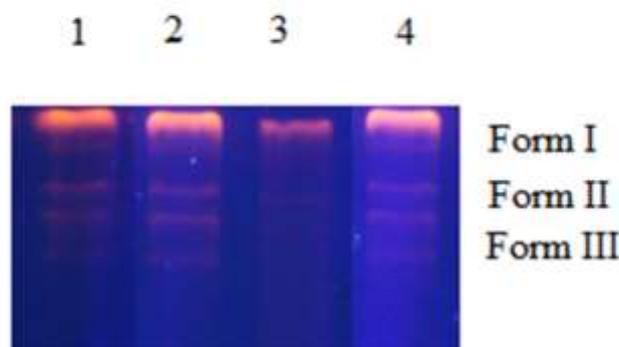


Figure 5. DNA Cleavage studies of complexes, DNA alone (Lane 1), CuL (Lane 2), ZnL (Lane 3) and NiL (Lane 4) Schiff base metal complexes.

3.8. DNA cleavage Activity

The DNA cleavage experiment was conducted using pUC18 DNA by electrophoresis with the corresponding metal complex in the presence of H_2O_2 as an oxidant. The cleavage of pU18 DNA induced by the metal complexes in the presence of H_2O_2 is shown in figure 5. In the absence of the complex (Lane 1), DNA remains in the supercoiled form. Incubation of DNA with Cu(II) complex (Lane 2) leads to its conversion to form II and form III. This indicates that Cu(II) complex has the ability to cleave pUC18 DNA in the presence oxidant H_2O_2 . The probable reason may be the oxidation of reoxyribose moiety by hydroxyl free radicals followed by the hydrolytic cleavage of the sugar phosphate backbone. The cleavage efficiency was measured in terms of the ability of the complex to convert the supercoiled form to open circular form. The ability of nickel and Zinc complexes (lane 3 and lane 4) is less when compared to the Cu

(II) complex. In the absence of H_2O_2 the synthesized complexes did not show any effect towards the cleavage the cleavage of DNA.

3.9. Antimicrobial Activity

For *in vitro* antimicrobial activity, the investigated compounds were tested against the bacteria such as Escherichia Coli, Bacillus Subtilis, pseudomonas and staphylococcus and fungi Aspergillus niger, Aspergillus flavus and Rhizoctonia bataicola. The ligand and their complexes were tested by minimum inhibitory concentration and values of the investigated compounds are summarized in table 4 and 5. The values indicate that most of the complexes have higher antimicrobial activity than the free ligands. Such increased activity of the metal chelates can be explained on the basis of chelation theory. On chelation, the polarity of the metal ion will be reduced to a 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

Table 4. Antibacterial activity of the Schiff base ligand and its metal complexes (Minimum inhibitory concentration $\times 10^{-2}$ M).

Compound	Escherichia Coli (mm)	Bacillus subtilis (mm)	Pseudomonas (mm)	Staphylococcus (mm)
L	4	3	4.5	4.2
CuL	15	15.2	16	15.4
ZnL	10	11	13	11
NiL	8	7	8.5	8.3

Table 5. Antifungal activity of the Schiff base ligand and its metal complexes (Minimum inhibitory concentration $\times 10^{-2}$ M).

Compounds	Aspergillus Niger (mm)	Aspergillus Flavus (mm)	Rhizoctonia Bataicola (mm)
L	5.2	4.3	4.8
CuL	16.2	16	16.5
ZnL	11.1	11.3	11.7
NiL	9.3	9.6	9.2

increases the delocalization of π - electron over the whole chelate ring and enhances the penetration of the complexes into lipid membranes and blocking the metal binding sites in the enzymes of microorganisms. These complexes also disturb the respiration process of the cell and thus block the synthesis of proteins, which restricts further growth of the organisms [27].

4. Conclusion

Azomethine ligand and their complexes were prepared and characterized. Analytical data, molar conductance measurements, magnetic susceptibility, IR, UV-Vis and $^1\text{H-NMR}$ spectral studies suggest the square planar geometry for Cu(II) and Ni(II) complexes except Zn(II) complex. The ligands coordinate to the metal ion via, the nitrogen atom of the amine, the oxygen atom of the aldehyde group, the azomethine $-\text{N}$ and the oxygen atom of the OH group in chelation

centers. Conductance data reveal that the complexes are non-electrolytes. The thermal data confirmed the suggested formula based on spectral results. The synthesized complexes were more active against bacteria and fungi than the ligands, thus giving a new thrust of these compounds in the field of metallo-drugs (bio-inorganic chemistry). Also metal complexes of such type are of interest especially due to their potential as biocides and nematocides with unique electrical and magnetic properties. [28]

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