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**Research Article** 

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# Synthesis and structural analogs of novel Schiff bases derived from sulfadoxine with 4-acetyl/benzoyl-1-(4'-nitrophenyl)-3-methyl-2-pyrazolin-5 one their metal complexes and antibacterial activity

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### **ABSTRACT**

4-acetyl/benzoyl-1-(4'-nitrophenyl)-3-methyl-2-pyrazolin-5-ones condensed with sulfadoxine to form Schiff bases. These Schiff bases form complexes type of ML<sub>2</sub>.2H<sub>2</sub>O (M= Mn, Fe, Co, Ni, Cu). Elemental analysis, magnetic susceptibility, electrical conductance, electronic and infrared spectral data suggested octahedral structures for the complexes. All the compounds were tested for their antibacterial activity. The results indicate that the growth of the tested organism was inhibited by most of the compounds. These Schiff bases were characterized by elemental analysis, mass spectra, <sup>1</sup>H NMR, <sup>13</sup>C NMR and FT IR spectra.

**Keywords:** Pyrazolin-5-one, sulfadoxine, Schiff base, Transition metals, Spectroscopy, antibacterial activity.

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### INTRODUCTION

The chemistry of metal complexes with heterocyclic ligands containing nitrogen, sulfur, and/or oxygen has attracted increasing attention. It is well known that these heterocyclic compounds can exhibit bacterial, fungicidal, herbicidal and insecticidal activities in addition to their application as potential drugs. Such heterocyclic ligands, when complexed with metal ions, exhibit enhanced microbiological activities [1,2]. The role of microelements in biochemical processes in well documented [3,4]. However, the mechanisms by which ligands operate are not common to all bioactive molecules. These molecules are expected to possess a certain ability to form complexes with particular metals. The hetero functional biomolecules form complexes with particular metals and their salts, bringing considerable changes to the physicochemical properties of the latter [5].

The coordination behavior of heterocyclic ligands, especially with transition metal ions, has been studied extensively [6-8], Major interest in the complexation of these ligands stems from their suitability as metal-containing model systems, which mimic biologically active system [8]. The presence of donor atoms (N,S,O) at various positions in these molecules enable them to out as multidentate ligands and thus form chelates of diverse structural types with a wide range of metal ions. The interaction of metal ions with biomolecules and the function of metal ions in physiological systems are very complex, and the precise mechanism of these interactions is almost unknown. In the synthetic systems, ligand design based on selective complexation with metal ions is limited to concepts, such as size-matched selectively in macrocycles, drop in stability due to increased size of chelate and steric strain. However, the ligand activity is a combination of steric, electronic, and pharmakinetic factors, and could be understood in the light of chelation theory [5]. In this context, various heterocycles, especially azoles, occupy an important place owing to their versatile bioactivities due to the presence of multifunctional groups. On the basis of our continuation work [9-14] the present investigation was to synthesize selected Mn(II), Fe(II), Co(II), Ni (II) and Cu(II) metal complexes using Sulfadoxine and 4-acetyl/benzoyl-1-(4'-nirophenyl)-3-methyl-2-pyrazolin-5-one as a chelating agent to ascertain their bonding modes and also to study their antibacterial activity.

### Scheme 1

### **EXPERIMENTAL SACTION**

**Chemicals:** All the reagents used were chemically pure or analytical reagent grade. Solvents were purified and dried according to standard procedures.

**Synthesis of Schiff base ligands:** The Schiff base ligands were prepared by the condensation of equimolar amounts of 4-acetyl-1-(4'-nitrophenyl)-3-methyl-2-pyrazolin-5-one or 4-benzoyl-1-(4'-nitrophenyl)-3-methyl-2-pyrazolin-5-one and sulfadoxine in minimum quantity of dioxane. The reaction mixture was refluxed in oil bath for three hours. On cooling the crystallized solid Schiff base separate out. It was then filtered and washed with some hot 1,4 dioxane and dried in air. The structure of Schiff base is shown in Scheme 1.

**Synthesis of metal complexes:** For the preparation of the metal complexes, an aqueous solution of the corresponding metal(II) acetate/sulphate (0.05M) and DMF solution of ligand (0.05M) were mixed in the presence of acetate buffer (pH=6.5) and the mixture was digested on a sand bath at 85-90°C for one hour, cooled and the precipitate filtered and then washed with water and finally with DMF to remove excess metal ion and unreacted Schiff bases.

**Physical measurements:** Melting points were taken in one side open capillaries on a melting point apparatus VEEGO VMP-D. Electronic Spectra were recorded in DMF solution on a LAMBDA 19, UV/VIS/NIR ("SICART-CVN" at Vallabh Vidyanagar, Gujarat, India). The thermogravimetric analysis (TGA) was carried out in a dynamic nitrogen atmosphere (20 mL.min<sup>-1</sup>), with a heating rate of 10 °C min<sup>-1</sup> using Shimadzu TGA-50H thermal analyzers. The Mass spectra of all ligands were recorded on a Shimadzu LCMS-2010A. Carbon, Hydrogen and Nitrogen were determined on a Thermo Fisher Flash Elemental Analyzer-1112. The <sup>1</sup>H NMR and <sup>13</sup>C NMR spectra of all the ligands [in deuterated chloroform (CDCl<sub>3</sub>)] were recorded on a BRUKER AVANCE-II 400 spectrometer using TMS [(CH<sub>3</sub>)<sub>4</sub>Si] as internal standard. The Infrared spectra of the ligands studied in the present work were recorded on a Shimadzu FT-IR-8300 of in KBr (Zydus Research Center, Ahmedabad, India).

TGA analysis: Thermo gravimetric analysis of Schiff base ligand and its metal complexes are used to: (i) Get information about the thermal stability of these new complexes, (ii) Decide whether the water molecules (if present) are inside or outside the inner coordination sphere of the central metal ion and (iii) Suggest the general scheme for thermal decomposition of metal complexes. The data are provided in the Table 2. Number of coordinated or lattice water molecule/molecules present in the complexes were calculated from the percentage weight loss of the complexes from the thermograms. Generally, the loss of lattice water will be at a lower temperature than that of coordinated water [15-18]. From the nature of the thermograms and percentage weight-loss, the complexes studied in the present work can be classified in the following three groups [19-21].

The Schiff base and their metal complexes were tested for antibacterial activity against Escherichia coli, Bacillus subtilis, Staphylococcus aureus and evaluated by use of the agar disc diffusion method on the basis of the size of the inhibition zone formed around the paper discs. For each concentration, the mean diameter (mm) of the inhibition zone developed was calculated. The test compounds in measured quantities were dissolved in DMF to get concentrations of 200 and 100 ppm of the compounds. Twenty five milliliter nutrient agar media was poured in each Petri dish. After solidification, 0.1mL of test bacteria were spread over the medium using a spreader. The discs of Whatmann no. 1 filter paper, having the diameter 5.00 mm, were placed at four equidistant places at a distance of 2

cm from the center in the inoculated Petri plates. A filter paper disc treated with DMF served as control and Amoxyciline used as a standard drug. These Petri plates were kept in a refrigerator for 24 hours for pre diffusion. Finally, Petri dishes were incubated for 24 hours at 30°C. The zone of inhibition was calculated in millimeters carefully.

### RESULTS AND DISCUSSION

Schiff bases ligand-L1 (where L1= 4-acetyl-1-(4'-nitrophenyl)-3-methyl-2-pyrazolin-5-one) and ligand-L2 (where L2= 4-benzoyl-1-(4'-nitrophenyl)-3-methyl-2-pyrazolin-5-one) are light yellow colored substance having melting points of 239°C and 248°C respectively. The elemental analysis suggests ML2.2H2O stoichiometry for all metal complexes. The metal complexes are insoluble in water, but soluble in DMF. All the compounds are solid, colored, stable and non-hygroscopic in nature. All the complexes indicates very low molar conductance, which indicated that the complexes are non-electrolytic in nature. The physical, analytical and conductivity data of the Schiff bases and their metal complexes are shown in Table 1.

### Analytical data and physical property of Ligands and their metal complexes: Table 1

Compound/ metal	Molecular	M.W	Color	M P	λ <sub>M</sub> *	Yield	Elemental analysis (%) Calculated (Found)					
complexes	Formula	171. 77	Color	( <b>'C</b> )	λ <sub>M</sub> ·	(%)	С	Н	N	S	M	μ <sub>eff</sub> Β.Μ.
Ligand-L <sub>1</sub>	$C_{24}H_{23}N_7O_7S$	553.14	Light Yellow	239	-	78	52.07 (52.11)	4.19 (4.17)	17.71 (17.84)	5.79 (5.81)	-	-
Mn(L <sub>1</sub> ) <sub>2</sub> .2H <sub>2</sub> O	C <sub>48</sub> H <sub>52</sub> MnN <sub>14</sub> O <sub>16</sub> S <sub>2</sub>	1200.08	Brown	-	13.81	59	48.04 (48.11)	4.37 (4.49)	16.34 (16.29)	5.34 (5.33)	4.58	5.59
Fe(L <sub>1</sub> ) <sub>2</sub> .2H <sub>2</sub> O	C <sub>48</sub> H <sub>52</sub> FeN <sub>14</sub> O <sub>16</sub> S <sub>2</sub>	1200.99	Radish Brown	-	09.12	62	48.00 (47.89)	4.36 (4.25)	16.33 (16.28)	5.34 (5.39)	4.65	4.98
Co(L <sub>1</sub> ) <sub>2</sub> .2H <sub>2</sub> O	C <sub>48</sub> H <sub>52</sub> CoN <sub>14</sub> O <sub>16</sub> S <sub>2</sub>	1204.07	Brown	-	12.28	66	47.88 (47.08)	4.35 (4.21)	16.29 (16.31)	5.33 (5.22)	4.89	4.52
Ni(L <sub>1</sub> ) <sub>2</sub> .2H <sub>2</sub> O	$C_{48}H_{52}NiN_{14}O_{16}S_2$	1203.83	Green	-	8.79	67	47.89 (47.28)	4.35 (4.26)	16.29 (16.27)	5.33 (5.54)	4.88	2.87
Cu(L <sub>1</sub> ) <sub>2</sub> .2H <sub>2</sub> O	$C_{48}H_{52}CuN_{14}O_{16}S_2$	1208.69	Dark Brown	-	11.47	59	47.70 (47.29)	4.34 (4.56)	16.22 (16.10)	5.31 (5.36)	5.26	1.99
Ligand-L <sub>2</sub>	C <sub>29</sub> H <sub>25</sub> N <sub>7</sub> O <sub>7</sub> S	615.62	Light Yellow	248	-	74	56.58 (56.46)	4.09 (3.98)	15.93 (15.84)	5.21 (5.17)	-	-
Mn(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	C <sub>58</sub> H <sub>56</sub> MnN <sub>14</sub> O <sub>16</sub> S <sub>2</sub>	1324.22	Brown	-	14.25	65	52.61 (52.50)	4.26 (4.22)	14.81 (14.73)	4.84 (4.41)	4.15	5.38
Fe(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	C <sub>58</sub> H <sub>56</sub> FeN <sub>14</sub> O <sub>16</sub> S <sub>2</sub>	1325.12	Radish Brown	-	11.13	69	52.57 (52.06)	4.26 (4.25)	14.80 (14.52)	4.84 (4.76)	4.21	4.97
Co(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	C <sub>58</sub> H <sub>56</sub> CoN <sub>14</sub> O <sub>16</sub> S <sub>2</sub>	1328.21	Brown	-	9.27	59	52.45 (52.06)	4.25 (4.11)	14.76 (14.66)	4.83 (4.68)	4.44	4.40
Ni(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	C <sub>58</sub> H <sub>56</sub> NiN <sub>14</sub> O <sub>16</sub> S <sub>2</sub>	1327.97	Green	-	10.43	62	52.46 (52.68)	4.25 (4.10)	14.77 (14.19)	4.83 (4.72)	4.42	2.91
Cu(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	C <sub>58</sub> H <sub>56</sub> CuN <sub>14</sub> O <sub>16</sub> S <sub>2</sub>	1332.83	Dark Brown	-	9.44	65	52.27 (52.36)	4.23 (4.42)	14.71 (14.65)	4.81 (4.44)	4.77	2.01

\*  $\lambda_M$  in  $\Omega^{-1}$  cm<sup>-1</sup> mol<sup>-1</sup>

FT-IR Spectra: From the recorded IR spectra, the wave numbers of the following groups are shown. 3140 cm-1 (vC-H stretching of Aromatic), 2923 cm-1 (vC-H stretching of saturated hydrocarbon), 1622 cm-1, (vC=N stretching of azomethine), 1595 cm-1, 1489 cm-1, 1480 cm-1, 1128 cm-1 (characteristic bands of pyrazolin ring) and 1501 cm-1 (Characteristic band of thiazole ring). One of the significant differences to be expected between the IR spectrum of the parent ligand and its metal complex is the presence of more broadened bands in the region of 3450-3100 cm-1 for the metal complex as the oxygen of the O-H group and nitrogen of the C=N azomethine of the ligand forms a coordination band with the metal ion. The ligand band at 1310 cm-1 assigned to υ (C-O), shifts to 1343-1318 cm-1 [22] on complexation, lending further support to the involvement of the nitrogen of the azomethine moiety in the complex formation. The infrared spectra of the ligands show a vO-H (weakly H-bonded) band at 2923 cm-1 [23]. The absence of this band in all the metal complexes indicates the removal of the proton of the hydroxyl group of the pyrazolin ring during the chelation. The sharp intense band at 1622 cm-1 in the ligand can be assigned to  $\nu$ C=N (azomethine). A downward shift ( $\Delta\nu$ =06-35cm-1) in  $\nu$ C=N (azomethine) is observed upon coordination, indicating that the nitrogen of the azomethine group is involved in coordination. All the complexes show a broad band in the region 3200 cm-1 to 3450 cm-1 which may be assigned to vO-H of coordinated water [24]. To account for the octahedral stereochemistry of the metal complexes, the coordination of two water molecules is expected. The bands present at ~514 cm-1 in the Mn(II) complex, ~545 cm-1 in the Fe(II) complex, ~589 cm-1 in the Co(II) complex, ~491 cm-1 in the Ni(II) complex and ~575 cm-1 in the Cu(II) complex respectively may be due to metalnitrogen stretching vibrations [25,26]. A less intense band at ~1618 cm-1 in the spectra of the ligands may be assigned to  $\nu$ C=N (ring) [27]. All the metal complexes do not show shifting in  $\nu$ C=N compared to their respective ligand. This suggests that the nitrogen atom of the thiazole ring has not participated in the coordination. However, in water containing metal complexes, this band is observed as a broad band with some fine structures. This may be due to coupling of the bending mode of coordinated water molecules with  $\nu$ C=N [28].

NMR spectra:  $^1$ H NMR spectrum of ligand  $L_1$ : chemical shifts and multiplicities of the corresponding protons are: (400 MHz, CDCl<sub>3</sub>)  $\delta$  = 1.91 (s, 3H, CH<sub>3</sub>), 1.96 (s, 3H, CH<sub>3</sub>), 2.41 (s, 1H), 3.83 (s, 3H, -OCH<sub>3</sub>), 4.01 (br s, NH), 4.07 (s, 3H, -OCH<sub>3</sub>), 7.62-8.24 (m, Aromatic Protons), 8.39 (s, 1H).  $^{13}$ C NMR Spectrum of ligand  $L_1$ : (400 MHz, CDCl<sub>3</sub>)  $\delta$  = 13.6, 22.5, 48.1, 54.5, 56.5, 118.5, 124.1, 130.8, 143.5, 145.0, 153.1, 153.2, 155.2, 155.9, 161.9, 165.6, 170.8  $^{1}$ H NMR Spectrum of ligand  $L_2$ : chemical shifts and multiplicities of the corresponding protons are: (400 MHz, CDCl<sub>3</sub>)  $\delta$  = 1.91 (s, 3H, CH<sub>3</sub>), 2.43 (s, 1H), 3.82 (s, 3H, -OCH<sub>3</sub>), 3.99 (br s, NH), 4.05 (s, 3H, -OCH<sub>3</sub>), 7.62-8.24 (m, Aromatic Protons), 8.41 (s, 1H).  $^{13}$ C NMR Spectrum of ligand  $L_2$ : (400 MHz, CDCl<sub>3</sub>)  $\delta$  = 22.7, 48.1, 54.5, 56.5, 118.5, 124.1, 128.9, 130.8, 131.5, 134.2, 143.5, 145.0, 153.2, 153.3, 155.1, 156.0, 161.7, 165.5, 170.5

**Mass spectra:** The positive ion mass spectral analysis of ligand  $L_1$  observed at m/z 554.1 ( $M^+$ ), confirms the theoretical molecular weight i.e. 553.14 and for ligand  $L_2$  observed at m/z 616.5 ( $M^+$ ), confirms the theoretical molecular weight i.e. 615.62.

Electronic spectra: The electronic absorption spectra are often very helpful in the evaluation of results furnished by other methods of structural investigation. The electronic spectral measurements were used for assigning the stereo chemistries of metal ions in the complexes based on the positions and number of d-d transition peaks. Both the ligands show two absorption bands between 37000 cm<sup>-1</sup> and 26000 cm<sup>-1</sup>. No absorption was observed in the visible region for the ligand. In the absence of Quantum mechanical calculation, it is not possible to assign the absorption bands to definite electronic transitions with complete certainty. However, it appears reasonable to assign the bands to  $\pi \rightarrow \pi^*$  transitions [29].

The electronic spectrum of the Mn(II) complex exhibits three very low intense bands, one at 16507 cm<sup>-1</sup>, 15209 cm<sup>-1</sup> which may be due to the  ${}^{6}A_{1g} \rightarrow {}^{4}T_{1g}$  (G) transition, another band at 17989 cm<sup>-1</sup>, 18159 cm<sup>-1</sup> assigned to the  ${}^{6}A_{1g} \rightarrow {}^{4}A_{1g}$  (G) transition and the third band at 25786 cm<sup>-1</sup>, 24547 cm<sup>-1</sup> may be assigned to the  ${}^{6}A_{1g} \rightarrow {}^{4}A_{1g}$ , 4E<sub>g</sub>, (G) transition respectively for the Mn(II) ions in an octahedral environment. The  $\mu_{eff}$  (Table 1) value of the complex suggests the 3d<sup>5</sup> spin configuration [30]. The electronic spectrum of Fe(II) complex shows broad bands at 23845 cm<sup>-1</sup> and 24416 cm<sup>-1</sup> respectively which may be assigned to the  ${}^{5}T_{2g} \rightarrow {}^{5}Eg$  transition. The magnetic moment value for both the ligands 4.98 BM and 4.97 BM respectively indicates that the complex is spin-free and it has octahedral geometry [31].

In the electronic spectrum of the Co(II) complexes exhibited two low energy peaks at 8352, 8812 cm<sup>-1</sup>; 16854, 16938 cm<sup>-1</sup> and a strong high energy peak at 19754, 20012 cm<sup>-1</sup>, which can be assigned to the transitions  ${}^4T_{1g}(F) \rightarrow {}^4T_{2g}(F)$ ,  ${}^4T_{1g}(F) \rightarrow {}^4A_{2g}(F)$  and  ${}^4T_{1g}(F) \rightarrow {}^4T_{2g}$  (P) for a high spin octahedral geometry. The magnetic measurement of Co(II) complex display in Table 1 magnetic moments is in the octahedral range. The Ni(II) complex exhibited three bands at 10541 cm<sup>-1</sup>, 16215 cm<sup>-1</sup> and 25974 cm<sup>-1</sup> for ligand  $L_1$  and 11017 cm<sup>-1</sup>, 16157 cm<sup>-1</sup> and 26338 cm<sup>-1</sup> for ligand  $L_2$  which are attributed to the  ${}^3A_{2g} \rightarrow {}^3T_{2g}(F)$  (v1);  ${}^3A_{2g} \rightarrow {}^3T_{1g}(F)$  (v2) and  ${}^3A_{2g} \rightarrow {}^3T_{1g}(p)$  (v3) transitions respectively indicating octahedral geometry around Ni(II) ion. Ni(II) complex showed the magnetic moment values in the range of 2.90 to 3.00 B.M suggesting consistency with their octahedral environment [32]. For the Cu(II) complex with  $D_{4h}$  symmetry, three spin allowed transitions  ${}^2B_{1g} \rightarrow {}^2A_{1g}$  (v1),  ${}^2B_{1g} \rightarrow {}^2B_{2g}$  (v2) and  ${}^2B_{1g} \rightarrow {}^2E_{2g}$  (v3) are possible but the electronic spectrum of the Cu(II) complex displayed two bands in the region of 13000 cm<sup>-1</sup> to 14000 cm<sup>-1</sup> and 20000 cm<sup>-1</sup> to 22000 cm<sup>-1</sup>. The third transition could not be observed which may be due to very close energy values of the different states. Absence of any spectral bands below 10000 cm<sup>-1</sup> rules out the possibility of a tetrahedral structure of the complexes and also suggests a distorted octahedral geometry of the complexes [33].

### Thermo gravimetric analysis

Thermo gravimetric analysis of Schiff base ligand and its metal complexes are used to: (i) Get information about the thermal stability of these new complexes, (ii) Decide whether the water molecules (if present) are inside or outside the inner coordination sphere of the central metal ion and (iii) Suggest the general scheme for thermal decomposition of metal complexes. In the present investigation, heating rates were suitably controlled at 10°C min<sup>-1</sup> under nitrogen atmosphere and the weight loss was measured from the ambient temperature up to ~1000°C [27-29]. The data are provided in the Table 2. Number of coordinated or lattice water molecule/molecules present in the complexes were calculated from the percentage weight loss of the complexes from the thermograms. Generally, the loss of lattice water will be at a lower temperature than that of coordinated water [30-33]. From the nature of the thermograms and percentage weight-loss, the complexes studied in the present work can be classified in the following three groups [34-36].

Thermo analytical results of metal complexes: Table 2

Compounds	Stage-I [140-210°C]	Stage-II [210-400°C]	Stage-III [400-900°C]	
Compounds	Mass Lose Calc. (Obs.)	Mass Lose Calc. (Obs.)	Mass LoseCalc. (Obs.)	
$Mn(L_1)_2.2H_2O$	3.00 (2.94)	90.18 (90.02)	6.57 (6.71)	
$Fe(L_1)_2.2H_2O$	3.00 (2.97)	90.11 (90.15)	6.64 (6.54)	
$Co(L_1)_2.2H_2O$	2.98 (2.88)	90.54 (90.32)	6.22 (6.51)	
$Ni(L_1)_2.2H_2O$	2.99 (2.87)	91.89 (91.44)	4.87 (4.71)	
$Cu(L_1)_2.2H_2O$	2.98 (2.91)	90.2 (90.12)	6.58 (6.29)	
$Mn(L_2)_2.2H_2O$	2.71 (2.65)	91.16 (91.05)	5.96 (5.86)	
$Fe(L_2)_2.2H_2O$	2.71 (2.68)	91.10 (91.08)	6.02 (6.11)	
$Co(L_2)_2.2H_2O$	2.71 (2.81)	91.49 (91.26)	5.64 (5.67)	
$Ni(L_2)_2.2H_2O$	2.71 (2.69)	92.71 (92.11)	4.41 (4.35)	
$Cu(L_2)_2.2H_2O$	2.70 (2.71)	91.17 (91.06)	5.96 (5.68)	
Assignment	Loss of two coordinated water molecules	Loss of two Schiff base ligand molecules	Metal Oxide/Metal	

TGA Analysis of metal complexes: In the present investigation, heating rates were suitably controlled at 10°C min under nitrogen atmosphere and the weight loss was measured from the ambient temperature up to ~1000°C [34-36]. The thermograms of this group of metal complexes show three stage decomposition. All the metal complexes do not show weight loss below 120°C, and they indicates the absence of lattice water in the metal complexes. The first stage decomposition is obtained in the temperature range 140-210°C. The % weight loss in this range corresponds to the loss of two coordinated water molecules [37-40]. The second stage decomposition is obtained in the temperature range 210-400°C. The % weight loss in this range corresponds to % weight loss of two Schiff base ligands. The third stage decomposition range is obtained in the temperature range 400-900°C. The % weight loss in this range corresponds to % weight loss of the metal oxide residue. On the basis of TGA and analytical data, all Mn(II), Fe(II), Co(II), Ni(II) and Cu(II) complexes studied in the present work correspond to the [ML<sub>2</sub>.2H<sub>2</sub>O] group.

### **Antibacterial activity**

The Schiff base ligand was found to be biologically active (Table 3). It is known that chelation tends to make ligands act as more powerful and potent bactericidal agents [41]. The values indicate that the metal complexes had a higher antibacterial activity than the free ligand. Such increased activity of the metal complexes can be explained on the basis of the overtone concept [42] and chelation theory [43]. According to the overtone concept of cell permeability, the lipid membrane that surrounds the cell favors the passage of only lipid soluble materials, due to which liposolubility is an important factor controlling the antimicrobial activity. On chelation, the polarity of the metal ion is reduced to a great extent due to the overlap of the ligand orbital and the partial sharing of the positive charge of the metal ion with donor groups. Furthermore, it increases the delocalization of electrons over the whole chelate ring and enhances the lipophilicity of the complex. This increased lipophilicity enhances the penetration of the complex into the lipid membrane and blocks the metal binding sites on the enzymes of the microorganism.

Antibacterial activity of ligand and its metal complexes: Table 3

	Zone of inhibition in mm(concentration in ppm)							
compound	E. 0	coli	B. su	btilis	S. aureus			
	100	200	100	200	100	200		
Ligand-L <sub>1</sub>	6	13	7	13	6	12		
$Mn(L_1)_2.2H_2O$	8	15	9	14	8	14		
$Fe(L_1)_2.2H_2O$	9	15	10	17	9	15		
$Co(L_1)_2.2H_2O$	9	16	11	15	8	13		
$Ni(L_1)_2.2H_2O$	14	21	12	18	12	23		
Cu(L <sub>1</sub> ) <sub>2</sub> .2H <sub>2</sub> O	13	19	11	19	13	21		
Ligand-L <sub>2</sub>	6	13	7	13	6	12		
Mn(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	8	15	9	14	8	14		
$Fe(L_2)_2.2H_2O$	9	15	10	17	9	15		
Co(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	9	16	11	15	8	13		
Ni(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	14	21	12	18	12	23		
Cu(L <sub>2</sub> ) <sub>2</sub> .2H <sub>2</sub> O	13	19	11	19	13	21		
Amoxyciline	17	28	16	22	14	29		

### Scheme 2

$$H_3CO$$
  $OCH_3$   $OCH_$ 

Where, M=Mn, Fe, Co, Ni, Cu  $R=-CH_3, -C_6H_5$ 

## **CONCLUSION**

On the basis of the results obtained from elemental analysis, infrared and electronic spectra, TGA analysis and magnetic susceptibility measurements it is clear that octahedral complex of type  $[ML_2(H_2O)_2]$  are formed. Antibacterial activity leads to the following conclusions: 1. The metal complexes show more activity than the ligands against the tested bacteria. 2. The antibacterial activity of the Cu(II) and Ni(II) complexes have higher activity than the other complexes.

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