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Synthesis, Spectral Characterization and Antibacterial Investigation of Ni(II) Coordination Complexes of Macrocyclic Schiff base ligands Derived from 4-Aminoantipyrine

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ABSTRACT

A series of Ni(II) complexes (NiL1-NiL5) have been synthesized from macrocyclic Schiff base ligands (L1-L5) were obtained from condensation of 4-aminoantipyrine derivative (L) with different diamines and hydrazides. All these compounds were well characterized by elemental, spectral analysis(mass, IR, ¹H-NMR, electronic), magnetic susceptibility, molar conductance and thermal studies. Macrocyclic Schiff bases are tetradentate with N4 donor system around the metal ion.Octahedral geometry have been assigned for all complexes. All ligands and complexes were examined for antibacterial activity and found that complexes were more potent when compared to ligands.

Keywords

4-Aminoantipyrine; Macrocyclic Schiff base; Ni(II) complexes; Spectral analysis; antibacterial activity.

1. INTRODUCTION

Antipyrine it self as named for its temperature reducing nature in humans which firstly found by Knorr. Among various derivatives of pyrazolone, 4-aminoantipyrine is significant owing to its willingly condensing property with various types of carbonyl compounds to obtain a variety of Schiff bases,which could be used in a number of major fields like biology and pharmaceutical industries¹⁻². The Schiff bases are more probable ligands in the formation of complexes with different metals due to its azomethine bond ³⁻⁸. Much work have been reported from many fields on 4-aminoantipyrine,which act as precursor to prepare a large number of various transition metal complexes ⁹⁻¹¹. Specifically Schiff bases derived from 4-aminoantipyrine and their complexes are most concerned because of their utility in the field of biology¹²⁻¹⁷.

Though, 4-aminoantipyrine and its derivatized Schiff bases exhibit fine antimicrobial activity, the activity has improved when it coordinates with metal centers¹⁸⁻²¹. In this context we are motivated to synthesize new Schiff bases from 4-aminoantipyrine as precursor and their Ni(II) complexes to evaluate their antibacterial property.

2. EXPERIMENTAL

2.1. Materials

Nickel(II) chloride, 4-amino antipyrine, orthopthalaldehyde(OPA), 1,4-diamino butane, carbanohydrazide, 4H-1,2,4-traizole-3,5-diamine, 2-amino benzo hydrazide and naphthalene-1,8-diamine were purchased from Aldrich and all the organic solvents such as ethanol, methanol, dichloro methane, DMSO were analytical grade. These solvents distilled and preserved under molecular sieves²². The purity of the compounds was confirmed by TLC using Merck 60F254 silica gel plates.

2.2. Physical Measurements

The percentages of carbon, hydrogen and nitrogen present in ligands and their complexes were determined by using PerkinElmer 2400 Series II CHNS/O analyzer. Buchi-510 melting point machine was used to find melting points. The Mettler-Toledo star system has been employed to perform thermogravimetric studies under a inert atmosphere of dry nitrogen and <10 mg of sample masses utilsed. The FTIR spectra of compounds have been recorded in the range of 4000–200 cm⁻¹ using Perkin Elmer-283 spectrophotometer. Brucker WH 300(400 MHz) and Varian Gemini (100 MHz) spectrometers were used to record NMR spectra(¹H&¹³C NMR). CEC-21-110B, Finningan Mat 1210 and MICROMASS-7070 spectrometers operating at 70 eV using a direct inlet system and VG-Auto-Spec-M mass spectrometer were used for mass spectra. Electronic spectra were obtained with Shimadzu UV-160A, a UV-Visible double beam spectrophotometer with matched quartz cells of path length 1 cm. Conductance measurements were carried out on 10^{-3} M solution of compounds in dichloromethane at 25 ⁰C on Dig sun Digital conductivity meter model DL-909. Gouy balance calibrated with Hg[Co(NCS)₄] was employed for the estimation of magnetic susceptibilities of complexes at normal temperature.

2.3. Synthesis of ligands

Firstly 4-aminoantipyrine derivative (L) synthesized by adding 4-aminoantipyrine (2 mmol), in ethanol (25 mL), little by little with steady stirring to an alcoholic solution (25 mL) of orthophthalaldehyde (1 mmol) (Scheme-1). This reaction mixture was refluxed and evaporated under reduced pressure and kept at room temperature for two days. The yellow coloured precipitate (L) obtained has been filtered, rinsed with diethyl ether solvent and recrystalised from ethanol. In the final step the ethanolic solution of L (2 mmol) was refluxed with diamines (2 mmol) viz., 1,4-diamino butane(L1),



carnohydrazide (L2), 4H-1,2,4-traizole-3,5-diamine(L3), 2-amino benzo hydrazide(L4) and naphthalene-1,8-diamine(L5) correspondingly with the addition of 1g of anhydrous K_2CO_3 for about 5-7 h (Scheme-2). The solvent was reduced to one-third and treated with hot water and kept in refrigerator for 24 h. The solid product obtained was separated by filtration and recrystalised from ethanol.

2.4. Synthesis of Schiff base NI(II) complexes

The newly prepared Macrocyclic Schiff base ligands and metal salt NiCl₂.6H₂O were used as starters for synthesis of complexes. Typical reported method was employed to synthesize all complexes and same procedure used for all ligands because of their resemblance. Ethanolic solution of nickel chloride (5 mmol) was added to a solution of the freshly prepared ligand solution (5 mmol) in ethanol (25 mL) with regular stirring. Further reaction mixture was refluxed on a water bath for few hours until a solid formed. The solids were separated by using suction filteration and rinsing with chloroform, ethanol and dried under vacuum over anhydrous calcium chloride.

2.5. Antibacterial activity

The newly synthesized ligands and complexes have been screened to evaluate In-vitro antibacterial activity in opposition to microbes like *Escherichia coli*, *Klebsiella pneumonia*, *Basillus subtilis*, *Staphylococcus aureus*. The existing antibiotics drugs such as Streptomycin and Rifampicin were used as the standards for current study. This investigation was occurred with help of known reported procedure is so called cup plate method ²³. Microoganisms were cultured in the combination of nutrient agar and nutrient broth (obtained from Hi-media, Mumbai). Sample compound was prepared 10 mg of compound in 10 ml of solvent water, methanol and DMSO. This sample is utilized to make a range of concentrations as 100, 50, 20, 15, 10, 5, 2, 1 µg/ml by addition of solvent. Semi solid agar medium (25 ml) was poured on to uncontaminated Petri dishes and put a side for few hours to allow solidify. Then well grown microorganism about 50 ml was added and distributed equally on to the above arranged agar medium dish with the help of fresh cotton swab. Fine borer was used to make 5 mm wide bores on the agar having cultured microorganism. These bores were further filled with sample solutions of compounds properly with micropipette. Similar steps also used in the preparation of plates for antibiotics. In presence of free air conditions all plates were incubated at the temperature of 37 ⁰C for 24 hours²⁴. Later the zones of inhibition of growth for all compounds and antibiotics were measured and compared. The results were expressed in terms of active and inactive. Further minimum inhibitory concentrations of all active compounds were determined²⁵⁻²⁷.

3. RESULTS AND DISCUSSION

Current work involved synthesis of new macrocyclic Schiff base ligands by condensation of derivative of 4aminoantipyrine with different diamines, diamino hydrazides in presence of base conditions (Scheme1) and their Ni(II) complexes. The carbon, hydrogen and nitrogen percentages in all newly prepared compounds were estimated by using CHN analyzer. The percentage of nickel in all complexes was found by known procedure²⁸. The tentative molecular formulas for all compounds were proposed based on the physical and analytical data (Table-1).



Scheme 1: Synthetic procedure of derivative of 4-amino antipyrene (L)





Scheme 2: Synthetic procedure of Macrocyclic Schiff base ligands (L1-L5)

| Compound | Molecular | Color | Found (Calculated)% | | | |
|---|-----------|--------------------|---------------------|------------|--------------|-------------|
| (Molecular | Weight | (%Yield) | С | н | N | NI |
| Formula) | | | | | | |
| L(C ₃₀ H ₂₈ N ₆ O ₂) | 505 | Yellow(72) | 71.38(71.41) | 5.55(5.59) | 16.61(16.66) | - |
| L1(C ₃₄ H ₃₆ N ₈) | 557 | Light Red(60) | 73.31(73.35) | 6.50(6.52) | 20.10(20.13) | - |
| L2(C ₃₁ H ₃₀ N ₁₀ O) | 559 | Light Orange(71) | 66.63(66.65) | 5.40(5.41) | 25.05(25.07) | - |
| L3(C ₃₂ H ₂₉ N ₁₁) | 568 | Light Brown(68) | 67.70(67.71) | 5.13(5.15) | 27.13(27.14) | - |
| L4(C ₃₇ H ₃₃ N ₉ O) | 620 | Light Yellow(65) | 71.69(71.71) | 5.33(5.37) | 20.32(20.34) | - |
| L5(C ₄₀ H ₃₄ N ₈) | 627 | Cream(63) | 76.63(76.65) | 5.44(5.47) | 17.85(17.88) | - |
| [Ni(L1)Cl ₂] | 686 | Light Green(70) | 59.47(59.50) | 5.27(5.29) | 16.32(16.33) | 8.54(8.55) |
| $(C_{34}H_{36}CI_2N_8Ni)$ | | | | | | |
| [Ni(L2)Cl ₂] | 688 | Green(67) | 58.09(54.10) | 4.37(4.39) | 20.33(20.35) | 8.51(8.53) |
| (C ₃₁ H ₃₀ Cl ₂ N ₁₀ ONi) | | | | | | |
| [Ni(L3)Cl ₂] | 697 | Yellow Green(61) | 55.10(55.12) | 4.15(4.19) | 22.08(22.10) | 8.38(8.42) |
| $(C_{32}H_{29}CI_2N_{11}Ni)$ | | | | | | |
| [Ni(L4)Cl ₂] | 749 | Pale Green(65) | 59.30(59.31) | 4.41(4.44) | 16.80(16.82) | 7.81(7.83) |
| (C ₃₇ H ₃₃ Cl ₂ N ₉ ONi) | | | | | | |
| [Ni(L5)Cl ₂] | 756 | Blackish Green(67) | 63.41(63.52) | 4.51(4.53) | 14.78(14.82) | 7.72 (7.76) |
| (C ₄₀ H ₃₄ Cl ₂ N ₈ Ni) | | | | | | |

| Table 1. F | Physical and anal | vtical data of the Mac | ro cyclic Schiff bases | and their Ni(II) complexe | s |
|------------|---------------------|--------------------------|------------------------|---------------------------|---|
| | ing orour arra arra | y lival aala ol liiv mav | o oyono oonni bacco | | - |



3.1 Infrared spectral analysis

The characteristic IR and far IR spectral absorption bands of the ligands were compared with the absorption bands of their Ni(II) complexes in order to under stand the coordination pattern in complexes. The significant absorption frequencies for all compounds are represented in Table-2 and spectra of ligand L1 and its Ni(II) complex were shown in Figure-1 and 2. Two strong intensity bands were found in all ligands spectra in the region of 1608-1565 cm⁻¹ is responsible for stretching frequencies of C=N group confirming the condensation between carbonyl group(L) with the amine group of respective diamines and hydrazides²⁹⁻³⁰. Further it is also supported by absence of a band ~1698 cm⁻¹ attributable to C=O group of L in all ligands³¹. This C=N frequency region of ligands when compared to complexes a considerable decrease by 20-35 cm⁻¹ was noticed for all the complexes is revealing the coordination between metal center and to nitrogen of azomethine group²⁹. Additionally it was also supported by a non-ligand band in the region of 528-506 cm⁻¹ due to metal nitrogen bond (M-N) stretching frequency³². In case of ligands L2 and L4 are having one C=O group and resonated around ~1670 cm⁻¹ while it is moved towards higher frequency region about to 25 cm⁻¹ disclosing the information that non participation of C=O group in the formation of complex. The complexes contains ligands L2, L3 and L4 shown a higher shift owing to N-H frequency when compared to ligands (3160-3144 cm⁻¹) demonstrating no region of 1412-1365 cm⁻¹ and 3086-3045 cm⁻¹ for all ligands and no significant changes were noticed in the case of corresponding complexes signifying that ring atoms are independent of coordination²⁹. In the spectra of all ligands a band was observed in range of 318–309 cm⁻¹ due to two chlorides were coordinated around metal center in trans manner³³.

| Compound | Selected bands(cm ⁺) | | | | |
|--------------------------|----------------------------------|------------------|------------------|-------------------|--------------------|
| | U _{N-H} | U _{C=0} | U _{C=N} | Y _{Ni-N} | U _{Ni-Cl} |
| L | - | 1698 | 1610 | - | - |
| L1 | - | - | 1596, 1576 | - | - |
| [Ni(L1)Cl ₂] | - | - | 1562, 1548 | 521 | 318 |
| L2 | 3155 | 1672 | 1608,1582 | - | - |
| [Ni(L2)Cl ₂] | 3178 | 1685 | 1581, 1562 | 516 | 313 |
| L3 | 3148 | - | 1601,1598,1585 | - | - |
| [Ni(L3)Cl ₂] | 3166 | - | 1585,1574,1561 | 528 | 315 |
| L4 | 3160 | 1665 | 1571,1561 | - | - |
| [Ni(L4)Cl ₂] | 3172 | 1688 | 1548,1538 | 512 | 309 |
| L5 | - | - | 1578, 1565 | - | - |
| [Ni(L5)Cl ₂] | - | - | 1543,1544 | 506 | 311 |

Table 2. IR (in cm⁻¹) spectral data of Macrocyclic Schiff bases and their Ni(II) complexes



Figure 1: Ir spectrum of L1





Figure 2: Ir spectrum of [Ni(L1)Cl₂]

3.2. NMR spectral analysis

The ¹H NMR spectra recorded for all ligands to recognize the chemically different protons present in them. The spectra of all ligands shown a singlet peak in the region of 8.05-8.44 δ attributable to proton of azomethine linkage (HC=N) and it confirms the condensation of OPA with 4-amino antipyrine to form L. Further it is also supported by the dissapearance of a characteristic peak ~9.91 δ due to aldehydic proton. A triplet peak at 3.73 δ (4H, t, CH₂-N=C) and 1.9 δ (4H, t, CH₂-C) was observed in the spectrum of L1 ligand. L2 spectra shown a a singlet peak at 6.12 δ (2H, s, N-NH-) and L3 exhibited a peak as a singlet at 4.75 δ (1H, s, N-H) corresponding to traizole ring. A singlet at 6.62 δ (1H, s, N-NH-) was noticed for L4. Protons of N-CH3 and C-CH3 were gave peaks in the range of 2.71-3.65 δ and 2.16-2.31 δ in case of all ligands. Aromatic protons resonated in range of 6.58-7.92 δ in all ligands as multiplets³⁴.

The ¹³C NMR spectra exhibited a signal in the range of 153.1-165.8 δ supporting the presence of imine carbon of azomethine group in all the ligands. It confirming the condensation between OPA with amine group of 4-amino antipyrine during the formation of L and next synthesis of ligands upon condensation of L with corresponding diamines & hydrazides³⁴. Further the spectrum of L contains C=O group is observed at 162.4 δ but it was absent in the spectra of all ligands is also confirmation for the formation of all macrocyclic Schiff base ligands. A signal in the range of 156.5-168.2 δ due to C=O group of amide found for the ligands L2 and L4. N-CH3 and C-CH3 signals were noticed in the region of 33.4-38.3 and 8.1-12.5 δ correspondingly in case of all ligands. The peaks corresponding to aromatic carbons are observed in the range of 116.3-148.2 δ . The ¹H & ¹³C NMR data of all the ligands are given in Table-3 and spectra of ligand L1 are shown in Figure-3 and 4 in that order.



Figure 3: ¹H NMR specterum of L1



13C



Figure 4: ¹³C NMR spectra of L1

| Ligand | ¹ H NMR peak position (δ ppm) | 13C NMR peak position (δ ppm) |
|----------------|---|--|
| ΟΑΑΡ | 8.16(2H, s, CH=N), 7.53-7.65(4H, m, Ar- H), 6.62-6.88(10H, m, Ar-H), 2.71(6H, s, N-CH ₃), 2.16(6H, s, C-CH ₃). | 12.5(2C, C- <u>C</u> H ₃), 33.4(2C, N-CH ₃), 112.2(2C, =C-N-), 123.1, 124.5, 130.1, 130.8, 132.5, 134.6, 135.2 (18C, Ar-C), 148.3(2C, =C-N-), 161.2 (2C, C=O), 164.5 (2C, CH=N). |
| L1 | 8.05(2H, s, CH=N), 7.51-7.70(4H, m, Ar- H), 6.70-6.95(10H, m, Ar-H), 3.73(4H, t, CH ₂ -N=C), 2.72(6H, s, N-CH ₃), 2.21(6H, s, C-CH ₃), 1.9(4H, t, CH ₂ -C) | 9.8(2C, C- <u>C</u> H ₃), 29.1(2C, C- <u>C</u> H ₂ -C), 36.5(2C, N-CH ₃), 47.1(2C, C- <u>C</u> H ₂ -N=), 109.8(2C, =C-N-), 123.5, 124.1, 130.6, 130.9, 132.3, 134.2, 136.8 (18C, Ar-C), 145.2(2C, =C-N-), 153.1(2C, C=N), 162.8(2C, CH=N). |
| L ₂ | 8.23(2H, s, CH=N), 7.55-7.68(4H, m, Ar- H), 6.60-6.86(10H, m, Ar-H), 6.12(2H, s, N-H), 3.65(6H, s, N-CH ₃), 2.25(6H, s, C- CH ₃). | 9.1(2C, C- <u>C</u> H ₃), 37.1(2C, N-CH ₃), 110.1(2C,=C-N-), 122.2, 123.5, 129.8, 130.5, 132.6, 134.8, 136.8 (18C, Ar-C),146.1(2C,=C-N-), 147.2(2C, C=N), 156.5(1C, C=O), 163.6 (2C, CH=N). |
| L ₃ | 8.44(2H, s, CH=N), 7.58-7.72(4H, m, Ar- H), 6.66-6.88(10H, m, Ar-H), 4.75(1H, s, N-H), 2.76(6H, s, N-CH ₃), 2.27(6H, s, C- CH ₃). | 9.6(2C, C- <u>C</u> H ₃), 34.1(2C, N-CH ₃), 111.4(2C, =C-N-), 121.9, 124.6, 130.8, 131.8, 132.5, 134.2, 137.1 (18C, Ar-C), 143.4(2C,=C-N-), 150.1(2C, -C=N) 153.3(2C, =C-N-), 164.1 (2C, CH=N). |
| L ₄ | 8.35(2H, s, CH=N), 7.55-7.92(8H, m, Ar- H), 6.68-7.2(10H, m, Ar-H), 6.62(1H, s, N- H), 2.80(6H, s, N-CH ₃), 2.26(6H, s, C- CH ₃). | $\begin{array}{l} 8.9(2C,\ C-\underline{C}H_3),\ 34.1(2C,\ N-CH_3),\ 107.5-112.0(2C,=C-N),\ 121.1,\\ 123.5,\ 124.2,\ 127.1,\ 127.8,\ 128.1,\ 130.2,\ 131.6,\ 132.5,\ 134.6,\\ 135.8,\ 137.1,\ 146.2(24C,\ Ar-C),\ 147.1-148.2(2C,=C-N),\ 151.6-153.6(2C,\ C=N)\ 164.7\ (2C,\ CH=N),\ 168.2(1C,\ C=O).\\ \end{array}$ |
| L ₅ | 8.21(2H, s, CH=N), 7.50-7.71(10H, m, naphthalene+Ar-H), 6.58-6.72(10H, m, Ar- H), 2.76(6H, s, N-CH ₃), 2.31(6H, s, C- CH ₃). | 9.1(2C, C- <u>C</u> H ₃), 38.3(2C, N-CH ₃), 112.2(2C, =C-N), 123.1, 124.2, 130.1, 132.3, 132.8, 135.8, 137.1 (18C, Ar-C), 145.2(2C,=C-N-),153.4(2C, CH=N), 165.8 (2C, CH=N), 116.3, 121.2, 127.2, 129.1,138.1,148.2(10C, naphthalene ring). |

3.3. Molar conductance, magnetic susceptibility and electronic spectral studies

All Ni(II)complexes were subjected to molar conductance study in dichloromethane solvent and their values were lie in between 11.5-14.3 ohm⁻¹cm²mol⁻¹(Table-4) indicating that all are non-electrolytes³⁵. The magnetic susceptibilities have been determined for all complexes using a Gouy balance at atmospheric conditions and are noticed in the range of 4.85-5.08 B.M. (Table-4) confirming the octahedral structure around metal center³⁶. The electronic spectral data are tabulated in the Table-4 and electronic spectrum of [Ni(L1)Cl₂] is presented in Figure-5. All the five complexes exhibited three bands at 982-1026 nm, 624-702 nm, 361-385 nm are assigned for the d-d transitions are ${}^{3}T_{2g}(F) \leftarrow {}^{3}A_{2g}(F)$, ${}^{3}T_{1g}(F) \leftarrow {}^{3}A_{2g}(F)$ and ${}^{3}T_{1g}(P) \leftarrow {}^{3}A_{2g}(F)$ respectively supporting the octahedral geometry of these complexes³⁷.



| Schiff base Ni(II) | λ _{max} (nm) | Λм | μ _{eff} |
|--------------------------|-----------------------|---|------------------|
| complex | | (Ω ⁻¹ cm ² mol ¹) | (B.M.) |
| [Ni(L1)Cl ₂] | 367, 702, 1012 | 14.3 | 4.87 |
| [Ni(L2)Cl ₂] | 361, 656, 982 | 13.6 | 4.92 |
| [Ni(L3)Cl ₂] | 373, 624, 998 | 12.4 | 4.85 |
| [Ni(L4)Cl ₂] | 381, 628, 987 | 11.5 | 5.08 |
| [Ni(L5)Cl ₂] | 385, 682, 1026 | 13.2 | 5.01 |

Table 4. Electronic spectral, molar conductance and magnetic data of Ni(II) complexes



Figure 5: Electronic spectrum of [Ni(L1)Cl₂]



The TG-DTG analysis revealed that two fine significant decomposition curves are present in all the thermograms of Ni(II) complexes. The initial stage at ~300 $^{\circ}$ C due to breaking of the complex by the dissemble of organic moiety is later supported by exothermic peak in the DTG curve and next curve above 500 $^{\circ}$ C is a result of metal oxidation. No weight loss was found in the temperature range of 70-125 $^{\circ}$ C and no endothermic peak observed in the DTG curve of these complexes confirming that absence of lattice water³⁸. All complexes are also not contains any coordinated water is evidenced by the nonappearance of a weight loss curve(in TGA) and endothermic peak (in DTG curve) in between 150-200 $^{\circ}$ C⁴⁰. The TG-DTG curves of Ni(II) complex of L2 is shown in Figure-6.



Figure 6: TGA-DTG curves of [Ni(L2)Cl₂]



3.5. Mass spectral analysis

The mass spectra of macrocyclic Schiff base ligands and their Ni(II) complexes have been showed different molecular ion peaks with different intensities. The molecular ion peak resulting of M^++1 pattern for all ligands observed at 506(L), 558(L1), 560(L2), 569(L3), 621(L4), and 628(L5) correspondingly. The mass spectra of complexes contains molecular ion peaks at m/z (M^+) 687 (M^++1 , for comp-NiL1), 690 (M^++2 , for comp-NiL2), 697 (M^+ , for comp-NiL3), 750(M^++1 , for comp-NiL4), and 756 (M^+ , for comp-NiL5). This data is good agreement with the corresponding molecular formulae. The mass spectra of ligand L1 and its Ni(II) complex are shown in Figure-7 & 8.



Figure 7: Mass spectrum of L1





4. ANTIBACTERIAL STUDIES

In the current study, all ligands and their Ni(II) complexes have been screened to evaluate their antibacterial activity in against of some selected bacteria. Results revealed that the complexes were exhibited more activity when compared to the free ligands. The increased activity of metal complexes could be explained by the fact that the affect of metal ion chelation on the usual cell metabolism. As believed by Tweedy's chelation theory³⁹⁻⁴⁰, chelation between metal ion and ligand considerably reduces the polarity of the metal ion since incomplete sharing of its positive charge with donor groups and plausible π -electron delocalization above the entire chelation. Hence lipophillic nature of metal ion enhances



which resulting in to its entry in to cell through lipid layers. All the all five complexes were shown good activity against four different strains of bacteria. Furthermore, all active ligands and complexes were also used to estimate their minimum inhibitory concentration (MIC) values and are given in Table-5. From the results, it is noticed that the Ni(II) complexes were displayed good bacteriostatic activity even at low concentrations when compared to standard antibiotics like Streptomycin and Ampiciline where as poor activity than Rifampicine at same concentrations.

| | Basillus Staphylo | | Escherichia | Klebsiella | |
|--------------------------|-------------------|---------------|-------------|------------|--|
| | subtilis | Coccus aureus | coli | pneumonia | |
| L1 | 25 | 28 | 31 | 22 | |
| L2 | 24 | 31 | 33 | 30 | |
| L3 | 26 | 27 | 29 | 26 | |
| L4 | 28 | 32 | 26 | 25 | |
| L5 | 32 | 26 | 23 | 23 | |
| [Ni(L1)Cl ₂] | 05 | 06 | 05 | 07 | |
| [Ni(L2)Cl ₂] | 04 | 04 | 02 | 05 | |
| [Ni(L3)Cl ₂] | 03 | 05 | 04 | 06 | |
| [Ni(L4)Cl ₂] | 04 | 09 | 06 | 03 | |
| [Ni(L5)Cl ₂] | 08 | 05 | 05 | 08 | |
| Streptomycin | 02 | 10 | 12 | 15 | |
| Ampicillin | 15 | 13 | 16 | 18 | |
| Rifampicin | 08 | 0.21 | 0.23 | 0.21 | |

Table 5. MIC of the macrocyclic Schiff bases, their metal complexes and existing antibiotics

5. CONCLUSION

New macrocyclic Schiff base Ni(II) complexes from 4-aminoantipyrine based ligands have been synthesized. In all the complexes ligand coordinates via four donor nitrogen atoms of azomethine group to metal ion. All complexes are non-electrolytic and para in nature. According to the results of elemental and spectral studies octahedral structures were assigned tentatively for all the complexes. All newly prepared compounds subjected to antibacterial study and these Ni(II) complexes were shown medium to good activity when compared to the uncoordinated ligands.

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