

SYNTHESIS, CHARACTERIZATION AND ANTIMICROBIAL ACTIVITIES OF {FE(II), CO(II), NI(II), CU(II), AND ZN(II)} MIXED LIGAND COMPLEXES SCHIFF BASE DERIVED FROM AMPICILLIN DRUG AND 4(DIMETHYLAMINO)BENZALDEHYDE WITH NICOTINAMIDE

Taghreed Hashim Al-Noor, Manhel Reemon Aziz, Ahmed.T.AL- Jeboori

Department of Chemistry, Ibn -Al-Haithem College of Education for pure science

Baghdad University-Iraq

drtaghreed2@gmail.com

Abstract- New Schiff base ligand (E)-6-(2-(4-(dimethylamino)benzylideneamino)-2-phenylacetamido)-3,3-dimethyl-7-oxo-4-thia-1-azabicyclo[3.2.0]heptane-2-carboxylic acid = (HL) Figure(1) was prepared via condensation of Ampicillin and 4(dimethylamino)benzaldehyde in methanol. Polydentate mixed ligand complexes were obtained from 1:1:2 molar ratio reactions with metal ions and HL, 2NA on reaction with $MCl_2 \cdot nH_2O$ salt yields complexes corresponding to the formulas $[M(L)(NA)_2Cl]$, where $M = Fe(II), Co(II), Ni(II), Cu(II), and Zn(II)$ and NA=nicotinamide.

The ¹H-NMR, FT-IR, UV-Vis and elemental analysis were used for the characterization of the ligand. The complexes were structurally studied through AAS, FT-IR, UV-Vis, chloride contents, conductance, and magnetic susceptibility measurements. All complexes are non-electrolytes in DMSO solution. Octahedral geometries have been suggested for each of the complexes. The Schiff base ligands function as tridentates and the deprotonated enolic form is preferred for coordination. In order to evaluate the effect of the bactericidal activity, these synthesized complexes, in comparison to the uncomplexed Schiff base has been screened against bacterial species, Staphylococcus aureus, Escherichia coli and the results are reported.

Key words:(Ampicillin drug, Nicotinamide, mixed ligand) Complexes, Antibacterial activities, and spectral studies.

I. INTRODUCTION

Schiff bases and its metal complexes have been found to exhibit biological activities including antifungal, antibacterial, antimalarial, antipyretic, larvicidal, and antiviral activities. Besides the biological activities, the metal complexes of Schiff bases are widely used as catalyst in the reactions like carbonylation, hydroformylation, reduction, oxidation, epoxidation and hydrolysis, corrosion inhibitor and as polymers [1].

Cephalosporin is a β -lactam antibiotic, which was first introduced in 1945. Such drugs are among the most widely prescribed antimicrobial classes because of their broad spectrum of activity, low toxicity, ease of administration, and favorable pharmacokinetic profile. The safety profile of cephalosporin's is generally good although a wide range of allergic reactions have been caused by β -lactam antibiotics. It is possible to classify these reactions by the Levine's classification system, which is based on the time of the onset

of the reactions. The frequency of hypersensitivity reactions to cephalosporin's is less than to penicillins. It has been estimated that nearly 75% of fatal anaphylactic reactions result from the administration of penicillin. Of all the injectable cephalosporin's, ceftriaxone, cefuroxime and oral cefaclor are the most frequently reported ones in allergic reactions [2].

Ampicillin is a very important class of β -lactam antibiotics used in therapy because of their specific toxicity towards bacteria. From a coordination chemistry perspective it has been demonstrated that all the β -lactamic antibiotics possess a number of potential donor sites and they are known to interact effectively with several metal ions and organo metallic moieties, originating complexes. Atoms involved in coordination and the structure of these complexes depend on several factors including reaction medium, pH, conformational equilibrium occurring in solution state and nature of the side chain bonded at C6 of the β -lactamic ring [3].

In 2008 Ogunniran and co-workers, [4]. synthesized Mixed ligand metal complexes of ampicillin and chloramphenicol prepared by using Ni(II), Co(II) and Fe(III) metal chloride hexahydrate were reported and characterized based on some physical properties and spectroscopic analysis such as AAS, UV, and IR spectroscopy. The synthesized complexes, in compares to their ligands, were also screened for their antibacterial activity against isolated strains of Escherichia coli, Staphylococcus aureus and Klebsiella pneumonia by using agar diffusion method [4].

In 2009 Pranay Guru, made studies of Ni(II) and Cu(II) complex with ampicillin have been synthesized and characterized. On the basis of elemental analysis and molar conductance, formulas $Ni(C_{16}H_{19}N_3O_4S)MoO_4 \cdot 4H_2O$, and $Cu(C_{16}H_{19}N_3O_4S)MoO_4 \cdot 4H_2O$ have been suggested for the complexes under study. The geometries of the complexes have been proposed on the basis of magnetic moment, electronic and infrared spectral data. Thermogravimetric analysis (TGA) has been carried out to determine the pattern of their decomposition. The crystal system, lattice parameters, unit cell volume and number of molecules in it have been determined by X-ray diffraction data (XRD) [5].

In (2013), Islam and co-workers A new series of four transition metal complexes of a Schiff base derived from salicylaldehyde and glycine, viz. [N-salicylidene glycinate diaqua cobalt (II) dimer] (SGCo)₂, [N-salicylidene glycinate-di-aqua-nickel(II)dimer] (SGN)₂, [N-salicylidene glycinate-aqua-copper(II)] (SGC) and [N-salicylidene glycinate diaqua zinc(II) dimer] (SGZ)₂, Figure (1-54) have been synthesized and characterized through a rapid, simple, and efficient methodology in excellent yield. These compounds were screened for in vitro antibacterial activities against six pathogenic bacteria, such as Shigella sonnei, Escherichia coli, Bacillus subtilis, Sarcina lutea, Staphylococcus aureus and Pseudomonas arioginosa [6].

I. Experimental

A. Chemical

All chemicals used were of reagent grade (supplied by either Merck or Fluka, and used as received., ethanol methanol and dimethylformamide, and KBr, from (B.D.H). ampicillin trihydrate powder) and 4(dimethylamino)benzaldehyde from Riedial-Dehaen DSM (Spain).

B. Instrumentation

UV-Vis spectra were recorded on a (Shimadzu UV-160A) Ultra Violet-Visible Spectrophotometer. IR- spectra were taken on a (Shimadzu, FTI R- 8400S) Fourier Transform Infrared Spectrophotometer (4000- 400) cm⁻¹ with samples prepared as KBr discs. Elemental micro analysis for the ligand was performed on a (C.H.N.S.O) Perkin Elemer 2400. While metal contents of the complexes were determined by atomic absorption (A.A) technique using a Shimadzu AA 620G atomic absorption spectrophotometer. The Chloride contents of complexes were determined by potentiometric titration method using (686-Titro processor-665. Dosimat Swiss) Molecular weight determined by Rast Camphor method, confirming the monomeric nature of the compounds. Conductivities were measured for 10⁻³M of complexes in DMSO at 25°C using (conductivity meter, Jewnary, model 4070). Magnetic measurements were recorded on a Bruker BM6 instrument at 298°K following the Farady's method. Nuclear magnetic resonance spectrum ¹H NMR for ligand was recorded in DMSO-d₆ using a Bruker 300 MHz instrument with a tetra methyl silane (TMS) as an internal standard. The samples were recorded at Queen Mary, university of Al-Albeit, Jordon, Amman. In addition melting points were obtained using (Stuart Melting Point Apparatus). The proposed molecular structure of the complexes was drawing by using chem.office prog, 3DX (2006).

C. Preparation of ligand (HL): [7]

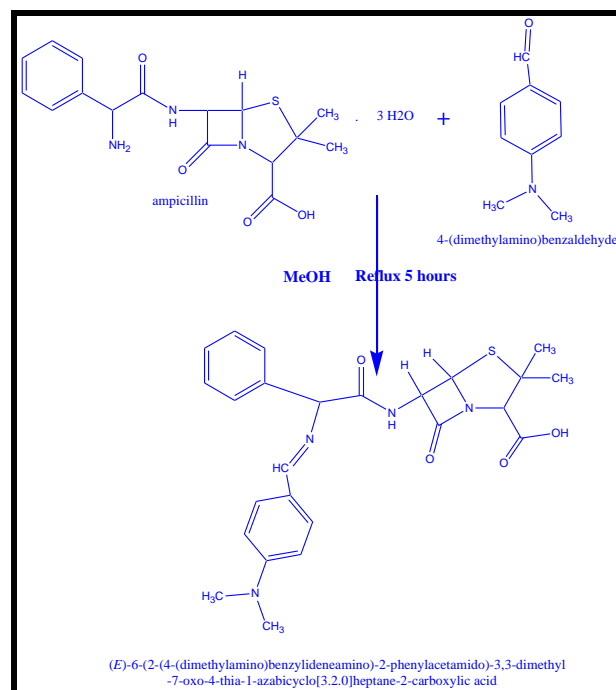
A solution of (Ampi) (0.403 gm, mmole) in methanol (20 ml) was added to a solution of (4DMAB) (0.149 gm, m mole) in methanol (10 ml). The mixture was refluxed for (5 hours) with stirring. The resulting was an deep orange solution allowed to cool and dried at room temperature, then re-crystallization to the precipitate with ethanol, deep orange solid was obtained by evaporation of ethanol during (24 hours) Scheme (1), m. p (88-93° C). Anal. Calc. for ligand (HL) C = 62.48%, H = 5.87%, N = 11.66 %. = Found: C = 62.00%, H = 6.12%, N = 11.86%.

D. Synthesis of [Cu(L)(NA)₂Cl] complex:[7]

A solution of (HL) (0.480gm, 1 mmole) in methanol (10 ml) and a solution of (NA) (0.244 gm, 2mmole) in methanol (10ml), were added to a stirred solution of Cu (II) chloride dihydrate (0.17 gm, 1mmole) in methanol (5 ml).The resulting mixture was heated under reflux for (5 hours). Then the mixture was filtered and the precipitation was washed with an excess of ethanol and dried at room temperature during (24 hours).A green solid was obtained, m. p (260-266° C).

E. Synthesis of [Fe(L)(NA)₂Cl], [Co(L)(NA)₂Cl], [Ni(L)(NA)₂Cl], and [Zn(L)(NA)₂Cl] complexes:[7]

The method used to prepare these complexes was similar method to that mentioned in preparation of [Cu(L)(NA)₂Cl] complex in paragraph Scheme (1).



Scheme (1): Schematic representation of synthesis of the ligand (HL)

F. Preparation of Microorganism suspension

A) The micro- organism suspension was prepared by taking 2–4 colonies from all the studied microorganism. Then it was inserted in the physiological solution in 0.85% concentration and was compared with Macferr land tube number 0.5 which is equal to 1.5×10⁸ cell/mm. It is used for Petri dish preparation for the examination of biological activity against the under studied chemical compound.

B) Inhibition Activity Selection for the complexes in studied Microorganism The agar well diffusion method was used to see the effect of under studied chemical complexes on the microorganism growth. This is done by using 20–25 ml from Nutrient agar medium for each Petri dish. The dish was incubated in incubator for 24 hours at (37°C) to make sure that no contamination would occur in the dish.

Bore was made on the cultured medium surface by using cork borer. The chemical complexes were made as 100 m ml per bore and left the central bore containing only DMF. The biological activity for the complexes was defined by measuring the diameter of the inhibition area surrounding each bore in millimeters. [8]

II. Results and Discussion

A. Physical Properties

Ligand (HL) is soluble in (N,N-dimethylformamide (DMF), dimethylsulphoxide (DMSO), methanol (MeOH), ethanol (EtOH), acetone ($\text{C}_3\text{H}_6\text{O}$), 2-propanol ($\text{C}_3\text{H}_8\text{O}$) and completely insoluble in water (H_2O), carbon tetra chloride (CCl_4), benzene (C_6H_6), Petroleum ether and chloroform (CHCl_3). Characterization of Metal Complexes:

Generally, the complexes were prepared by reacting the respective metal salts with the ligands using 1:1:2 mole ratio, i.e. one mole of metal salt: one mole of HL and two moles of nicotinamide. The formula weights and melting points are given in (Table 1 based on the physicochemical characteristics, it was found that all the complexes were non-hygroscopic, stable at room temperature and appears as powders with high melting points. The solubility of the complexes of ligands was studied in various solvents. They are not soluble in water. All complexes are soluble in (DMF) and (DMSO) solvent. The complexes were analyzed for their metal by atomic absorption measurements and chloride contents were determined by standard methods. (Table-1) for all complexes gave approximated values for theoretical values. Molar conductance values of the soluble complexes in DMSO show values ($9.7\text{-}16.2 \Omega^{-1} \text{ cm}^2 \text{ mol}^{-1}$) indicating that they are all non-electrolytic in nature [9].

B. FT-IR spectrum of the ligand (HL)

The (FT-IR) spectrum for the starting material ampicillin, band at $(3446) \text{ cm}^{-1}$ due to $\nu(\text{N}-\text{H})$ primary amine stretching vibration. The bands at $(3269, 3209) \text{ cm}^{-1}$ are due to the $\nu(\text{N}-\text{H})$ secondary amine stretching vibration. The band at $(1774) \text{ cm}^{-1}$ is due to $\nu(\text{C}=\text{O}) \text{ cm}^{-1}$ stretching vibration for (COOH). The band at $(1687) \text{ cm}^{-1}$ stretching vibration is due to $\nu(\text{C}=\text{O})$ for β -Lactam group. The bands at $(1575) \text{ cm}^{-1}$, and $(1384) \text{ cm}^{-1}$ were assigned to stretching vibration (COOH) asymmetric and symmetric stretching vibration, respectively. The bands stretch at (1496) , (3041) , (1170) , and (2970) were assigned to $\nu(\text{C}=\text{C})$ aromatic, $\nu(\text{C}-\text{H})$ aromatic, $\nu(\text{C}-\text{C})$ aliphatic, and $\nu(\text{C}-\text{C})$ stretching vibration respectively. The band at $(1296) \text{ cm}^{-1}$ is due to $\nu(\text{C}-\text{N}) \text{ cm}^{-1}$ stretching vibration. The band at $(1263) \text{ cm}^{-1}$ was assigned to $\nu(\text{C}-\text{O})$ stretching vibration. The band at $(590) \text{ cm}^{-1}$ was assigned to $\nu(\text{C}-\text{S})$ stretching vibration. The spectrum for the starting material 4(dimethylamino)benzaldehyde (DMBA) which exhibits a band to $\nu(\text{C}-\text{N})$ were observed in the $(1371) \text{ cm}^{-1}$. The bands at $(1165) \text{ cm}^{-1}$ and $(2796) \text{ cm}^{-1}$ were assigned to the $\nu(\text{C}-\text{C})$ and $\nu(\text{C}-\text{H})$ aliphatic stretching vibration. The very strong bands due to carbonyl group $\nu(\text{C}=\text{O})$ stretching of (DMBA) were observed in the $(1662\text{-}1600) \text{ cm}^{-1}$ regions, while the bands at $(1548) \text{ cm}^{-1}$ and $(2819) \text{ cm}^{-1}$ were assigned to the $\nu(\text{C}=\text{C})$ aromatic and $\nu(\text{C}-\text{H})$ aromatic stretching vibration respectively.

The (FT-IR) spectrum for the ligand (HL), the displays band at $(3228) \text{ cm}^{-1}$ due to $\nu(\text{N}-\text{H})$ secondary amine stretching vibration, and disappeared the band for the $\nu(\text{N}-\text{H})$ primary amine stretching vibration. The spectrum displays a new band at $(1597) \text{ cm}^{-1}$ is due to $\nu(\text{C}=\text{N})$ stretching vibrations of the ligand. The band at $(1678) \text{ cm}^{-1}$ is due to $\nu(\text{C}=\text{O}) \text{ cm}^{-1}$ stretching vibration for (COOH). The band at $\nu(1678) \text{ cm}^{-1}$ stretching vibration is due to $\nu(\text{C}=\text{O})$ for β -Lactam group overlapping with $\nu(\text{COOH})$ stretching vibrations. The bands at $(1597) \text{ cm}^{-1}$, and $(1373) \text{ cm}^{-1}$ were assigned to stretching vibration (COOH) asymmetric and symmetric stretching

C. FT-IR of spectral data for the Schiff base mixed ligands complexes [Fe(L)(NA)₂Cl](1), [Co(L)(NA)₂Cl](2), [Ni(L)(NA)₂Cl](3), [Cu(L)(NA)₂Cl](4), And [Zn(L)(NA)₂Cl](5):

The spectrum of the (HL) displays a new at $(1597) \text{ cm}^{-1}$ is due to $\nu(\text{HC}=\text{N}-)$ group of the azomethine stretching vibrations of the ligand, on complexation these band has been shifted to higher frequencies (1662) , (1624) , (1624) , (1620) , and $(1662) \text{ cm}^{-1}$ for complexes (1), (2), (3), (4), (5).

The bands at (1597) , and $(1377) \text{ cm}^{-1}$ were assigned to stretching vibration (COOH) asymmetric and symmetric stretching vibration, respectively. on complexation these bands have been shifted to higher frequencies $[(1600)$, (1664) , (1666) , (1668) , and $(1697) \text{ cm}^{-1}$ for $\nu(-\text{COO})_{\text{asym}}$], and lower frequencies $[(1333)$, (1336) , (1334) , (1331) , and $(1337) \text{ cm}^{-1}$ for $\nu(-\text{COO})_{\text{sym}}$] for the compounds (1), (2), (3), (4), and (5) that the coordination with metal was occurred through the oxygen atom of carboxylate ion. The overlap band at $(1678) \text{ cm}^{-1}$ stretching vibration is due to $\nu(\text{C}=\text{O})$ for β -Lactam group, these band has been shifted to lower frequency at $(1662\text{-}1666) \text{ cm}^{-1}$ for complexes showing that the coordination is through the Oxygen atom of β -Lactam group [10-11].

The bands at (497) , (486) , (470) , (489) , and $(474) \text{ cm}^{-1}$ were assigned to $\nu(\text{M}-\text{O})$ for compounds (1), (2), (3), (4), and (5), indicating that to the carbocyclic oxygen, and oxygen of β -Lactam group of the ligand are involved in coordination with metal ions.

The bands at (509) , (532) , (520) , (505) and $(540) \text{ cm}^{-1}$ were assigned to $\nu(\text{M}-\text{N})$ for compounds (1), (2), (3), (4), and (5), respectively, indicating that the nitrogen is involved in coordination with metal ions.

D. (U.V-Vis) Spectrum for the Schiff base ligand:

The electronic spectrum of the ligand has been measured in DMSO solution between 200- 1100 nm at room temperature. In the spectrum of the Schiff base ligand (HL), the absorption band observed at $(212 \text{ nm}) (47169 \text{ cm}^{-1}) (\epsilon_{\text{max}}=1000 \text{ molar}^{-1} \cdot \text{cm}^{-1})$, $(239 \text{ nm}) (41841 \text{ cm}^{-1}) (\epsilon_{\text{max}}=689 \text{ molar}^{-1} \cdot \text{cm}^{-1})$, and high intense absorption peak at $(335 \text{ nm}) (29850 \text{ cm}^{-1}) (\epsilon_{\text{max}}=2114 \text{ molar}^{-1} \cdot \text{cm}^{-1})$, which assigned to $(\pi \rightarrow \pi^*)$, $(\pi \rightarrow \pi^*)$, and $(n \rightarrow \pi^*)$ transition respectively [10-11,16].

E. (U.V-Vis) Spectral data for the Schiff base mixed ligands complexes:

The absorption data for complexes are given in Table (3).

[Fe(L)(NA)₂Cl]:

The (U.V- Vis) spectrum exhibits four peaks, the first small peak at $(267 \text{ nm}) (37453 \text{ cm}^{-1}) (\epsilon_{\text{max}}=1407 \text{ molar}^{-1} \cdot \text{cm}^{-1})$, is due to the ligand field, and the second high peak at $(342 \text{ nm}) (29239 \text{ cm}^{-1}) (\epsilon_{\text{max}}=1346 \text{ molar}^{-1} \cdot \text{cm}^{-1})$ is due to the (C.T), third weak peak at $(817 \text{ nm}) (23980 \text{ cm}^{-1}) (\epsilon_{\text{max}}=9 \text{ molar}^{-1} \cdot \text{cm}^{-1})$

¹) is refers to (⁵T_{2g}→⁵E_g) (d-d), transition. [16]Transitions, similar to those found for distorted octahedral complexes. The doublet is attributed to a Jahn-Teller distortion in the excited state. The room temperature magnetic moment (4. 27 B.M) corresponded to octahedral symmetry.[17]

[Co(L)(NA)₂Cl]:

The (U.V- Vis) spectrum exhibits three peaks , the first high intense peak at (268 nm)(37313 cm⁻¹)(ε_{max} =1488 molar⁻¹.cm⁻¹), is due to the ligand field, the second small peak at (345 nm)(28985 cm⁻¹)(ε_{max} =69 molar⁻¹.cm⁻¹) is due to the (C.T), while the third weak peak at(412 nm)(24271 cm⁻¹)(ε_{max} =20 molar⁻¹.cm⁻¹), which assigned to (⁴T_{1g}→⁴A_{2g}), transition respectively in an octahedral geometry. Also, the values of the magnetic moments 5.11 B.M, may be taken as additional evidence for a high spin octahedral nickel (II) geometry.[17]

[Ni(L)(NA)₂Cl]:

The (U.V- Vis) spectrum exhibits four peaks , the first high intense peak at (267 nm)(37453 cm⁻¹)(ε_{max} =1422 molar⁻¹.cm⁻¹), is due to the ligand field, and the second small peak at (345 nm)(28985 cm⁻¹)(ε_{max} =68 molar⁻¹.cm⁻¹) is due to the (C.T), third and fourth weak peaks at (642 nm)(15576 cm⁻¹)(ε_{max} =4 molar⁻¹.cm⁻¹), and (992 nm)(10080 cm⁻¹)(ε_{max} =9 molar⁻¹.cm⁻¹), which assigned to (³A_{2g}(F) → ³T_{1g}(F) (v2), and (³A_{2g}(F) → ³T_{2g}(F) (v1) transition respectively in an octahedral geometry. [16]

Also, the values of the magnetic moments 3.22 μB. may be taken as additional evidence for octahedral nickel (II) geometry.[17]

[Cu(L)(NA)₂Cl]:

The (U.V- Vis) spectrum exhibits two peaks, the first high broad peak at (279 nm)(35842 cm⁻¹)(ε_{max} =2298 molar⁻¹.cm⁻¹) is due to the ligand field, while the second weak broad peak at (977 nm)(10235 cm⁻¹)(ε_{max} =58 molar⁻¹.cm⁻¹), which assigned to (²E_g→²T_{2g}), transition . Hence the Cu(II) mixed ligand complexes showed distorted octahedral geometry. [16]

[Zn(L)(NA)₂Cl]:

The (U.V- Vis) spectrum exhibits two high peaks, the first high peak at (286 nm)(34965 cm⁻¹)(ε_{max} =1072 molar⁻¹.cm⁻¹), and the second high peak at (342 nm)(29239 cm⁻¹)(ε_{max} =1693 molar⁻¹.cm⁻¹) are due to the (C.T), in an octahedral geometry. this diamagnetic complex show no appreciable absorptions in the region below 26000 cm⁻¹ in DMSO solutions. In accordance with the d¹⁰ electronic configuration of Zn(II).

Also, the values of the magnetic moment Table (3).may be taken as additional evidence³32 for their octahedral structure.Molecular weight determined by Rast Camphor method and were found in accordance with calculated value the range of metal complexes (815-822), [7].

4-¹H NMR spectrum for the ligand (HL):

The ¹H NMR spectrum of (HL), Figure (1)

The signal obtained in range (δ 6.71-7.70) ppm was assigned for doublet due one proton of aromatic ring of phenyl and (δ 7.37) due one proton of benzylidenimin, (S-CH) on the dihydrothiazine ring was observed in the (δ 2.80-3.66) ppm, The signal obtained at (δ 8.76) ppm was assigned for singlet due one proton of (-CH=N) linkage in the ligand. This confirms the formations of imine ligand. This observation was also supported by the FT-IR data of the ligand discussed earlier. The NMR spectral data of (HL) was compared with the spectral data for the similar ligands reported in literatures. Three groups of double peaks given by (CO-CH) and (N-CH) on the beta-lactam ring and (CH=N=C) appeared at (δ 4.86) ppm [17-19].

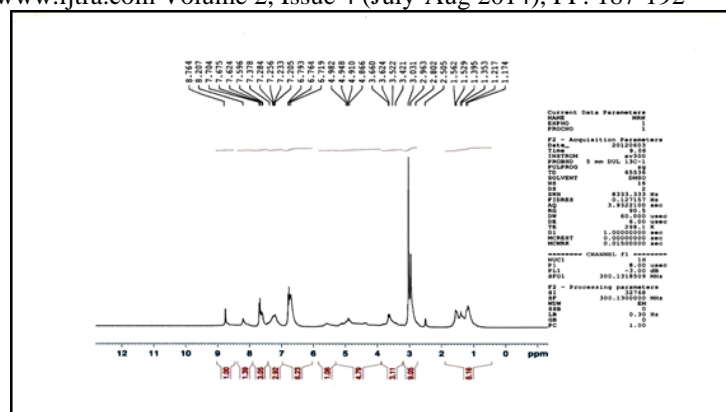


Figure (1): ¹H NMR spectrum of the ligand (HL) in DMSO-d₆

V. Antimicrobial activity

The in vitro antimicrobial screening results are given in Table 4, Chart (1). On the basis of observed zones of inhibition, all the metal-mixed ligand complexes are active against all six tested organisms which in fact are in agreement with the literature [20-23].

REFERENCES

- [1] Badma Priya D, Santha Lakshmi S, (2014), "Synthesis, Spectral and Antimicrobial Investigation of some Ternary Schiff Base Transition Metal Complexes", International Journal of Chem Tech Research, Vol.6, No.1, pp 87-94.
- [2] Sweetman, C. Sean, (2009), "Antibacterials". (36th Ed.). London: Pharmaceutical Press. Pp. 218-9.
- [3] R. Di Stefano, M. Scopelliti, C. Pellerito, T. Fiore, R. Vitturi, M.S. Colomba, P. Gianguzza, G.C. Stocco, M. Consiglio, L. Pellerito, (2002)," Organometallic complexes with biological molecules XVII. Triorganotin (IV) complexes with amoxicillin and ampicillin", Journal of Inorganic Biochemistry 89, 279-292.
- [4] Ogunniran, K. O., Ajanaku, K. O., James, O. O., Ajani, O. O., Adekoya, J. A. and Nwinyi, O. C (2008). Synthesis, characterization, antimicrobial activity and toxicology study of some metal complexes of mixed antibiotics. African Journal of Pure and Applied Chemistry, 2(7). pp69 -074.
- [5] Pranay Guru, (2009), "Studies of two complexes with ampicillin", International Journal of ChemTech Research, Vol.1, No.3, pp 461-463.
- [6] M. N. Islam, , S. M. S. Shahriar, M. K. Islam, M. Jesmin, M. M. Ali, J. A. Khanam (2013),International Letters of Chemistry, Physics and Astronomy, 5 , pp 12-20.
- [7] Sunil Joshi, Vatsala Pawar , And V. Uma, 2011 Research Journal Of Pharmaceutical, Biological And Chemical Sciences, RJPBCS Volume 2 Issue 1.,
- [8] Vignolo M.; Surlant F.; Holgado A.P., and Oliver G.,(1993). Antibacterial activity of Lado bacillus strains iso lated from dry fermented sausage; Journal Of App. Bac. 75:pp 344-349.
- [9] Geary W. J., (1971).The Use of Conductivity Measurements in Organic Solvents for the Characterisation of Coordination Compounds. Journal Of coord. Chem. Rev. 7, 81-122.
- [10] Nakamoto K.; (1996) "Infrared spectra of Inorganic and coordination compounds "4ED th ; J. Wiley and Sons, New york,
- [11] Juan R. Anaconda And Ibranyellis Rodriguez, "Synthesis And Antibacterial Activity Of Cephalexin Metal Complexes"(2004) , Journal Of. Coord. Chem., Vol. 57, No. 15, 15 October, pp. 1263-1269.
- [12] Aldo Caiazzo, Shadi Dalili, Christine Picard, Mikio Sasaki, Tung Siu, and Andrei K.Yudin, (2004)New methods for the synthesis of heterocyclic compounds, ; Journal Of Pure Appl. Chem., Vol. 76, No. 3, pp. 603-613.

- [13] Taghreed .Al-Noor, Ibtisam Dawood and Ibtihaj. Malih, Synthesis, (Spectroscopic and Antibacterial) Studies of Tin (II) and Lead (Anthranilic acids and nicotinamide) Complexes, (2012)Journal of International Journal for Sciences and Technology Vol. 7, No. 3, September pp 32-42
- [14] Dhivya Priya D., Akila E., Usharani M. And. Rajavel R., (2012) Synthesis, Spectral And Biological Activity Of Mixed Ligand Schiff Base Complexes Derived From Salicylaldehyde, Journal of International Journal Of Pharmacy &Technology,(IJPT), April- Vol. 4 , Issue No.1, pp 4067-4078.
- [15] Fayad,N.K.,Taghreed H. Al-Noor and Ghanim F.H, (2012)Synthesis, Characterization, And Antibacterial Activities Of Manganese (II), Cobalt(II), Iron (II), Nickel (II), zinc (II) And Cadmium(II) Mixed- Ligand Complexes Containing Amino Acid(L-Valine) And Saccharin, Journal of Advances in Physics Theories and Applications, Vol 9, pp1-12.
- [16] Lever A.B.P., (1984) “Inorganic Electronic spectroscopy“, Elsevier, New York.
- [17] Dutta R. L. and Syamal A.,(1996) Elements of Magnatochemistry, 2nd Ed., East west press, New Delhi,
- [18] Sinn E. and Harris C.M., Coord. Chem. Rev., (1969), 4(4), pp.391-422.
- [19] Atmaram K. Mapari, and Kiran V.. Mangaonkar , International Journal of Chem Tech Research, Jan-Mar (2011), Vol.3, No.1, pp 477-482
- [20] Fayad N.K. , Taghreed H. Al-Noor and F.H Ghanim,(2012). Synthesis ,characterization and antibacterial activity of mixed ligand complexes of some metals with 1-nitroso-2-naphthol and L-phenylalanine , Journal of Chemistry and Materials Research, Vol 2, No.5,pp18-29
- [21] Chaudhary Rakhi and Shelly, (2011) Synthesis, Spectral and Pharmacological Study of Cu(II), Ni(II) and Co(II)Coordination Complexes, Res. J. Chem. Sci., 1(5), pp1-5.
- [22] Shriver & Atkins (1999) “Inorganic Chemistry”, 3rd Ed.,, Freeman.
- [23] Taghreed H. Al-Noor, Sajed. M. Lateef and Mazin H. Rhayma, (2012)Synthesis, Characterization, Schiff Base Phenyl 2-(2-hydroxybenzylideneamino)benzoate and its complexes with La(III), Cr (III), and Pr (III), *Journal of Chemical and Pharmaceutical Research*, 4(9): pp 4141-4148.

Compounds	M. wt Calc.	M. wt Rats method	Color	Yield %	M. p ° c (de) ° c	$\Lambda_m \Omega^{-1} \text{cm}^2 \text{mol}^{-1}$	Metal%	Cl%
							theory (exp)	
[Fe(L)(NA) ₂ Cl] C ₃₇ H ₃₉ ClFeN ₈ O ₆ S	815.11	824.6	Brown	75	174-186	16.2	6.85	4.35 (4.30)
[Co(L)(NA) ₂ Cl] C ₃₇ H ₃₉ ClCoN ₈ O ₆ S	818.20	837.2	Violet	86	279-292	10.5	7.20	4.33 (4.37)
[Ni(L)(NA) ₂ Cl] C ₃₇ H ₃₉ ClNiN ₈ O ₆ S	817.96	838.0	Yellow	83	300 Dec.	11.4	7.18	4.33 (4.35)
[Cu(L)(NA) ₂ Cl] C ₃₇ H ₃₉ ClCuN ₈ O ₆ S	822.81	832.1	Green	71	262-270	9.7	7.72	4.31 (4.28)
[Zn(L)(NA) ₂ Cl] C ₃₇ H ₃₉ ClZnN ₈ O ₆ S	824.68	836.5	Brown	72	218-224	14.3	7.93	4.30 (4.36)

Table (1): The physical properties of the compounds

M. wt = Molecular Weight , Λ_m = Molar Conductivity , dec. = decomposition , Calc.= calculation

Table (3): Electronic Spectral data, magnetic moment, of the studied compounds

Compound	$\lambda \text{ nm}$	$\tilde{\nu} \text{ cm}^{-1}$	$\epsilon_{\text{max}} \text{ molar}^{-1} \cdot \text{cm}^{-1}$	Assignments	$\mu_{\text{eff}} (BM)$
AmpH	264	37878	640	$\pi \rightarrow \pi^*$	
	327	30581	786	$n \rightarrow \pi^*$	
NA	220	45454	1548	$\pi \rightarrow \pi^*$	
	261	38314	1104	$\pi \rightarrow \pi^*$	
4DMAB	208	48007	338	$\pi \rightarrow \pi^*$	-
	342	29239	493	$n \rightarrow \pi^*$	
	336	29761	1961	$n \rightarrow \pi^*$	
LH	212	47169	1000	$\pi \rightarrow \pi^*$	-
	239	41841	689	$\pi \rightarrow \pi^*$	
	335	29850	2114	$n \rightarrow \pi^*$	
[Fe(L)(NA) ₂ Cl]	267	37453	407	L-F	4.27
	342	29239	1346	C-T	
	817	23980	9	$^5T_{2g} \rightarrow ^5E_g$	
[Co(L)(NA) ₂ Cl]	268	37313	1488	L-F	5.10
	345	28985	69	C-T	
	990	10101	9	$^4T_{1g} \rightarrow ^4A_{2g}$	
[Ni(L)(NA) ₂ Cl]	267	37453	1422	L-F	3.22
	345	28985	68	C-T	
	642	15576	4	$^3A_{2g}(F) \rightarrow ^3T_{1g}(P)(v3)$	
	759	13227	8	$^3A_{2g}(F) \rightarrow ^3T_{1g}(F)(v2)$	
	992	10080	9	$^3A_{2g}(F) \rightarrow ^3T_{2g}(F)(v1)$	
[Cu(L)(NA) ₂ Cl]	279	35842	1198	L,F	1.82
	977	10235	58	$^2E_g \rightarrow ^2T_{2g}$	
[Zn(L)(NA) ₂ Cl]	286	34965	1072	C-T	0.0
	342	29239	1693	C-T	

Table (4): Biological activity of the Schiff bases mixed ligands complexes

(Zone of inhibition (mm))

Compound	<i>E-coli</i>	<i>Pseudomona</i>	<i>Staphylococcus aureus</i>	<i>Klebsiella pneumoniae</i>	<i>Salmonella Typhi</i>	<i>Acinetobacte baumannii</i>
Control	5	7	5	6	5	5
[Fe(L)(NA) ₂ Cl]	0	15	0	15	0	20
[Co(L)(NA) ₂ Cl]	14	24	14	14	25	16
[Ni(L)(NA) ₂ Cl]	16	17	0	13	14	16
[Cu(L)(NA) ₂ Cl]	0	18	0	12	14	21
[Zn(L)(NA) ₂ Cl]	16	18	0	13	14	20

Observation	Report
Inhibition zone > 15mm	Highly active
Inhibition zone >10mm	Moderately active
Inhibition zone > 5mm	Slightly active
Inhibition zone ≤5-0 mm	Inactive

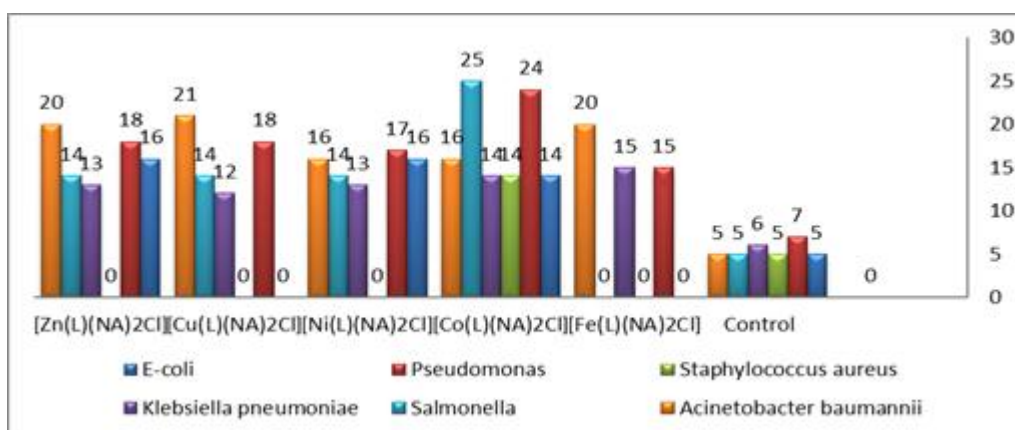


Chart (1): Graphic of biological effects of some of the studied complexes

Compound	$\nu(\text{N-H})$ primary amine	$\nu(\text{N-H})$ Secondary amide	$\nu(\text{C=O})$ β -lactam	ν (HC=N-)	ν_{as} COO	ν_{s} COC	$\nu(\text{C=C})$ arom.	$\nu(\text{C-C})$ aliph	$\nu(\text{C-N})$	$\nu(\text{C-O})$	$\nu(\text{C-S})$	$\nu(\text{C-H})$ arom	ν (C-H) aliph	$\nu(\text{M-N})$	$\nu(\text{M-O})$
HL	-----	3228 overlap	1678 overlap	1597 overlap	1597 overlap	1373	1531	1168	1284	1230	570	3062	2924	-----	-----
Nicotine amide	3367, 3159	-----	-----	-----	-----	-----	1593	1124	1201	-----	-----	3061	2787	-----	-----
[Fe(L)(NA) ₂ Cl]	3414	3232, 3190	1662	1662 overlap	1600 overlap	1333	1535	1166	1315	1230	572	3060	2880	509	474
[Co(L)(NA) ₂ Cl]	3317	3267, 3194	1666 overlap	1624	1664	1336	1577	1199	1296	1242	573	3078	2900	532	489
[Ni(L)(NA) ₂ Cl]	3408	3221, 3267	1666	1624	1666	1334	1581	1145	1292	1199	578	3078	2900	520	478
[Cu(L)(NA) ₂ Cl]	3402	3228	1666	1620	1668	1331	1570	1149	1292	1199	572	3066	2950	505	478
[Zn(L)(NA) ₂ Cl]	3298 overlap	3298 overlap	1662	1662 overlap	1697	1337	1531	1195	1292	1249	573	3062	2970	540	470

Table (2): Data from the Infrared Spectra for the Metal Complexes (cm⁻¹).