

Synthesis, diagnosis as well as Biological activity studies of some metal ion complexes of Schiff Bases Derived from 4-aminoantipyrine

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Abstract

Polydentate Schiff base ligands L1=orthohydroxy aniline, 4-aminoantipyrin-(N,N⁻ benzilidene) and L2=5-Chloro-2-hydroxyaniline,4-aminoantipyrine-(N,N⁻) benzilide obtained from condensation of benzil and 4-aminoantipyrine with 2-aminophenol or 2-amino 4-chlorophenol in (1:1:1) molar ratio for each component. Ligands employed to synthesis complexes in the general molecular formula [ML1Cl]Cl and [ML2]Cl2 respectively; M= Mn(II), Co(II), Ni(II) and Zn(II). To diagnose the ligands and the complexes the following techniques were adopted: UV-Visible FTIR and 1H-NMR spectroscopies, micro elemental analyses of metal ions and carbon, hydrogen and nitrogen, molar conductivity and magnetic susceptibility. Measurements obtained from the conductivity which was done with DMSO solution indicated that all the complexes are electrolytes with ratio (1:1) for L1 complexes and (1:2) for L2 complexes. The results of the measurements which used in this study showed coordination sites for L1 with central ion were through the phenolic oxygen atom, and two nitrogen atoms of the azomethin groups, L2 in addition to the previous donor atoms, the coordination is carried out by the oxygen atom of the terminal carbonyl group. All complexes are tetra-coordinate with (tetrahedral geometry). Evaluation of biological activity for the compounds tested against Gram (+) staphylococcus aureus and Gram (-) Klebsiella pneumonia.

Keywords: 4-aminoantipyrine, Biological activity, Metal complex, Polydentate, Schiff Base.

Introduction

Schiff base ligands, since they were first prepared and studied in 1864¹ are still of great interest by many researcher. ²⁻⁴ This type of organic compound has wide applications in many different fields from medical^{5,6}, pharmaceutical, industrial, agricultural and others⁷. Polydentate Schiff base is distinguished by its ability to easily attach with metal ions to form more stable coordination compounds, especially when nitrogen atom of C=N group and another donor atom share in coordination with the central atom ^{8,9}. The chemistry of this type of complex has attracted wide attention because of their biological activity,

stability and potential application of many fields such as oxidation catalysis¹⁰ electrochemistry etc.

Antipyrine Schiff base derivatives have been extensively examined because they have shown wide range of applications in various fields like biological, analytical, therapeutic and are also used as precursors in the synthesis of bioactive compounds¹¹. In recent years, many of researchers tended and interest to be published on transition metal complexes derived from antipyrine Schiff base ^{12,13}.

In the present study we report the synthesis of polydentate Schiff bases (L¹ and L²) derived from 4-

Antibacterial Activity

The antibacterial activity of the compounds was tested against Gram (+) bacteria *Staphylococcus aureus* and Gram (-) bacteria *Klebsiella-Pneumoniae* by using the agar diffusion method.

The test solution was prepared by dissolving 10 mg of the prepared materials in (1 ml) of DMSO. A (6mm) diameter blank paper disc was cultivated of 37°C for 24 h. The diameter of the inhibition zone in (mm) was determined and the resulting activity was estimated. A Standard disk of Ciprofloxacin used as a positive control^{16,17}.

Results and discussion

The synthesized Schiff base ligands (L¹ and L²) scheme 1 forms 10 stable complexes with Mn(II), Co(II), Ni(II), Cu(II) and Zn(II) in (1:1) molar ratio (M:L). All the complexes were soluble in most organic solvents and stable in dry air and their melting points ranged from 104 to 258°C. The results obtained from the analytical as well as the physical traits of each compound are recorded in Table 1. The obtained analysis results for the prepared complexes fit in with the suggested formula [ML¹Cl]Cl and [ML²]Cl₂. The conductivity results in DMSO showed that all complexes are electrolytic with (1:1) and (1:2) ratio for L¹ complexes and L² complexes respectively.

The suggested structures of the prepared complexes are shown in Fig. 2

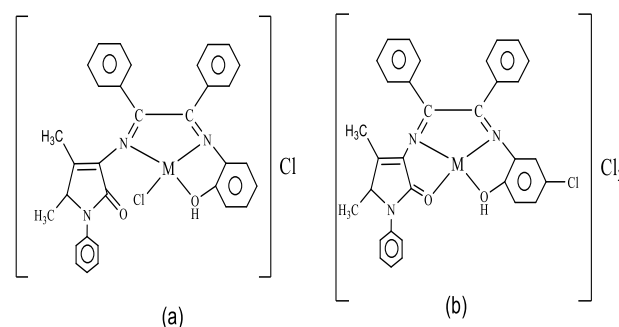


Figure 2. Suggested structure of (a):[ML¹Cl]Cl complexes (b):[ML²]Cl₂ complexes. M=Mn(II), Co(II), Ni(II), Cu(II), Zn(II)

Table 1. Analytical data and physical properties of compounds

NO	Compounds	Color	m.p. (°C)	Analysis Calc. (found)%			M% Calc. (found)	Λ _m ohm ⁻¹ . cm ² .mol ⁻¹
				C	H	N		
L ¹	C ₃₁ H ₂₆ N ₄ O ₂	Dark yellow	130	76.54 (76.24)	5.34 (5.25)	11.52 (10.76)	-----	-----
1	[MnL ¹ Cl]Cl	Light gray	258	68.77 (67.98)	4.80 (4.32)	10.35 (10.13)	10.15 (9.93)	36.1
2	[CoL ¹ Cl]Cl	Brown	245	68.26 (67.80)	4.77 (4.52)	10.27 (9.82)	10.81 (10.53)	40.6
3	[NiL ¹ Cl]Cl	Light brown	147	68.29 (67.55)	4.77 (4.23)	10.28 (9.97)	10.77 (10.23)	34.4
4	[CuL ¹ Cl]Cl	Dark brown	264	67.69 (67.12)	4.73 (3.91)	10.19 (6.38)	11.56 (10.95)	39.9
5	[ZnL ¹ Cl]Cl	Yellow	210	67.46 (67.05)	4.71 (4.62)	10.15 (9.82)	11.85 (11.27)	38.1
L ²	C ₃₁ H ₂₅ N ₄ O ₂ Cl	Dark orange	174	71.46 (71.82)	4.80 (5.00)	10.75 (10.26)		
6	[MnL ²]Cl ₂	Light orange	112	64.64 (64.25)	4.34 (4.15)	9.73 (9.21)	9.54 (9.27)	69.2
7	[CoL ²]Cl ₂	Light brown	110	64.20 (63.88)	4.31 (4.17)	9.66 (9.19)	10.17 (9.82)	73.7
8	[NiL ²]Cl ₂	Light brown	135	64.22 (46.19)	4.31 (5.14)	9.66 (6.55)	10.13 (9.69)	70.11
9	[CuL ²]Cl ₂	Dark brown	104	63.69 (63.20)	4.28 (3.97)	9.58 (9.13)	10.87 (10.56)	82.00
10	[ZnL ²]Cl ₂	Gray	216	63.49	4.26	9.55	11.16	68.11

(63.23) (3.94) (9.07) (10.85)

¹H-NMR spectra

The ¹H-NMR spectra of L¹ and L² were registered in DMSO – d⁶ solvent and the ¹H-NMR assignments for the compounds are presented in Table 2 and Fig. 3^{2, 3}

Table 2. The ¹H-NMR data of the ligands

	δ(ppm)	Assignment
L ¹	6.53-7.71	(m,19H, ArH)
	3.14	(S,3H, C-CH ₃)
	2.47	(S,3H, N-CH ₃)
	8.91	(bs, 1H, OH)
L ²	7.15-7.71	(m,19H, ArH)
	3.14	(S,3H, C-CH ₃)
	2.47	(S,3H, N-CH ₃)
	8.91	(bs, 1H, OH)

S=singlets, M=multiplets, Bs=broad signal

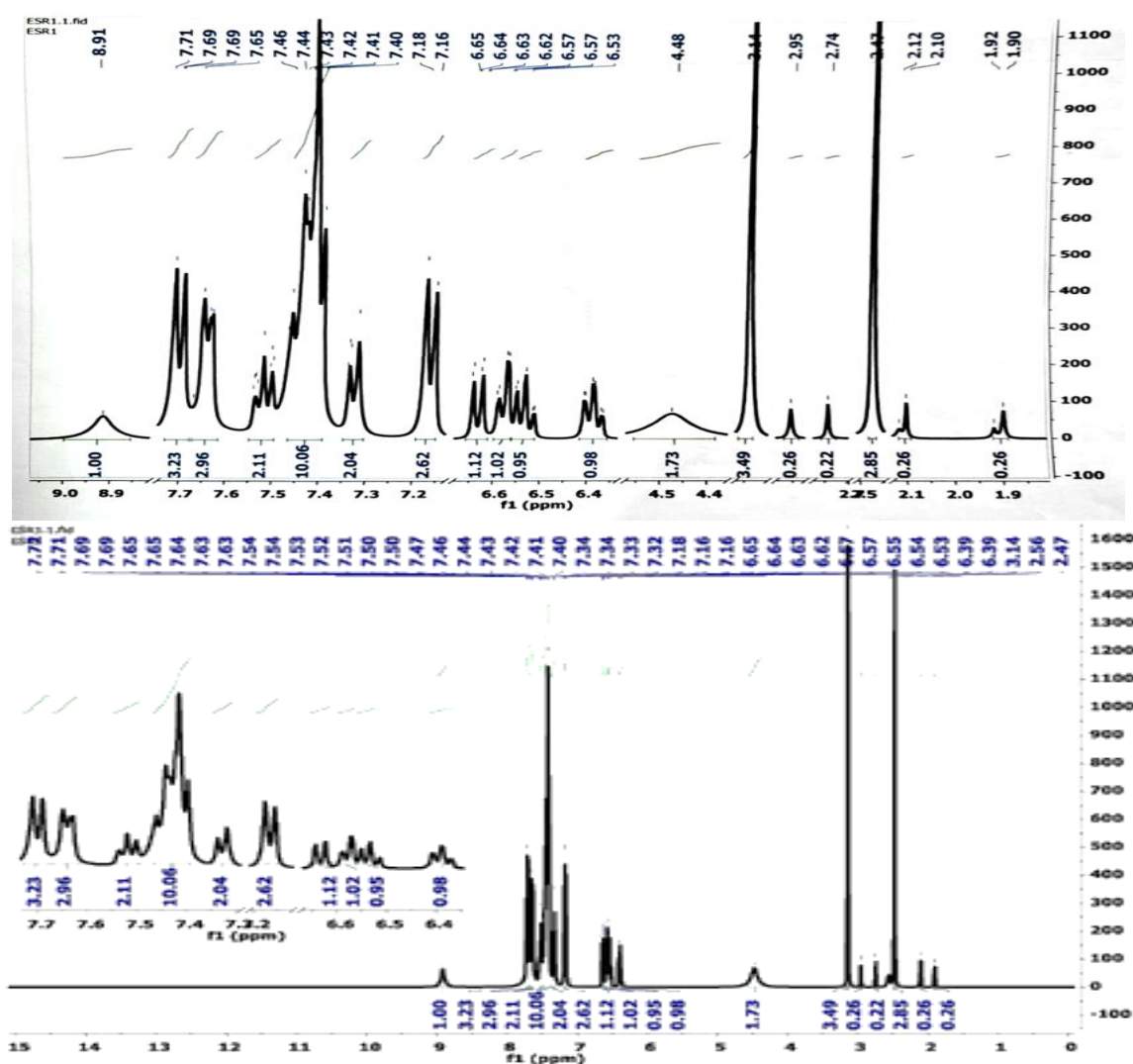


Figure 3. ¹H-NMR spectra of L¹

IR- spectra

Important peaks for all compounds have been diagnosed by IR spectra. The band at 1591 and 1589

cm⁻¹ in IR spectra of L¹ and L² supported the formation of ν(C=N) group^{18,19}, this band shifted to higher frequency in the spectrum complexes^{20, 21}.

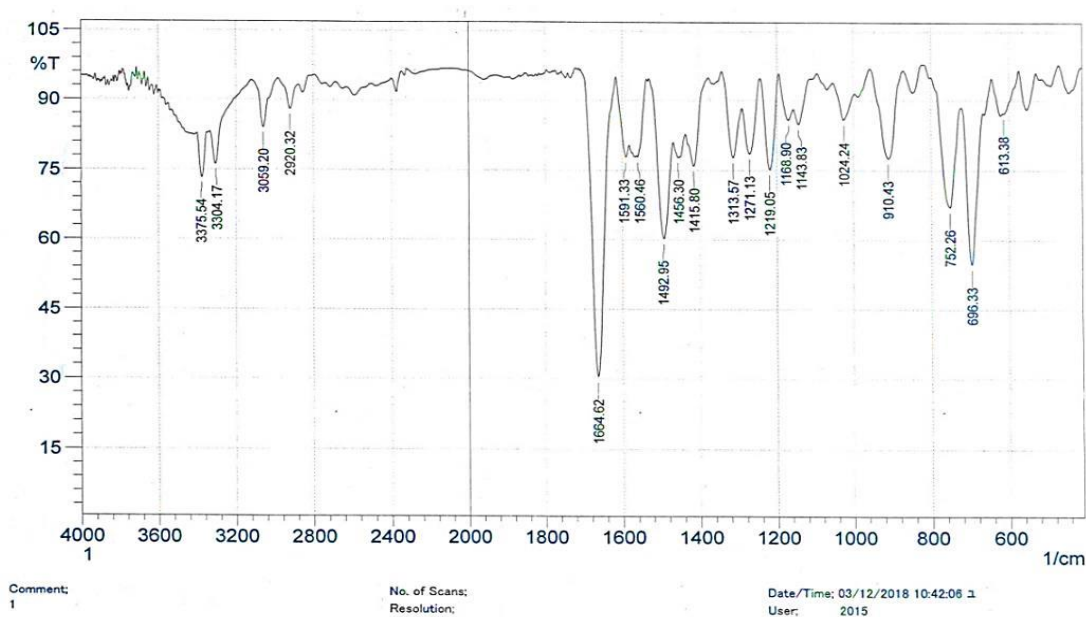
which indicates the participation of the nitrogen atom of the azomethine group in bonding.

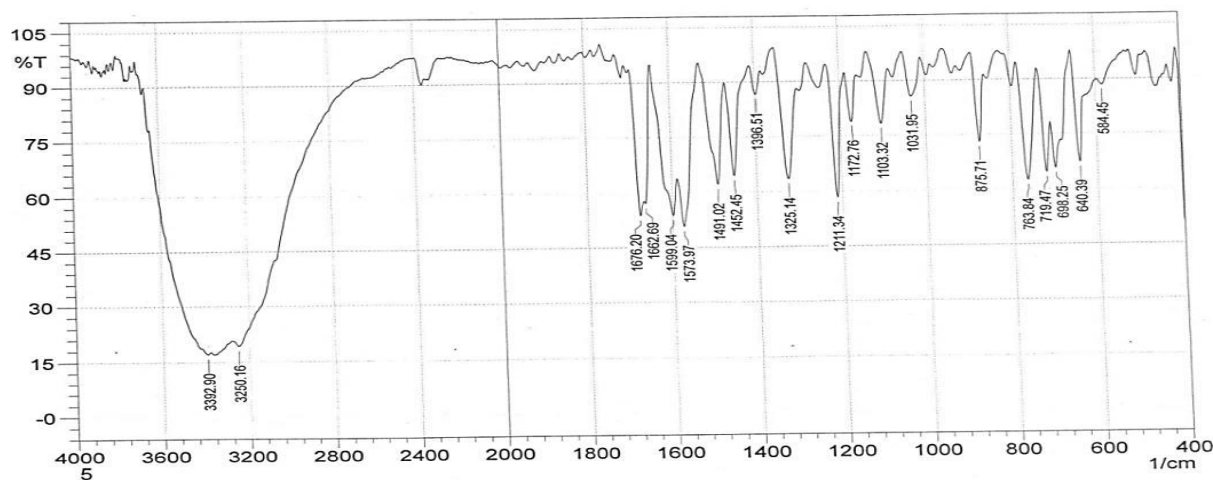
It is clear from the IR spectrum of the free ligand that the bands at 3375 and 3379 cm^{-1} due to phenolic ν (OH) group of L^1 and L^2 respectively this band shift to higher frequencies in the spectrum of the complexes this indicates the coordination of phenolic oxygen to central ion without deprotonating²²⁻²⁵, another important band appeared at 1664 and 1662 cm^{-1} due to ν (C=O) of cyclic keton. In L^1 complexes(1-5) this band remains almost unchanged on complexation, which indicates that the carbonyl oxygen atom of cyclic ketone is not involved in coordination in these complexes.

While in L^2 complexes (6-10) this band shifted to higher frequencies indicating that the carbonyl oxygen atom of cyclic keton considered one of the coordinating sites²³, on the other hand on the spectrum of all complexes a new band was appeared at 503-586 cm^{-1} and 420-466 cm^{-1} due to ν (M-O) and ν (M-N) stretching vibration²⁵. ν (M-Cl) band is not registered because this band is below the spectrophotometer limits. This discussion suggested that L^1 coordinates to metal in tridentate fashion NNO while L^2 is tetradentate fashion NNOO. The results are mentioned in the following Table 3 and shown in Fig.2

Table 3. IR bands of L^1, L^2 and their complexes

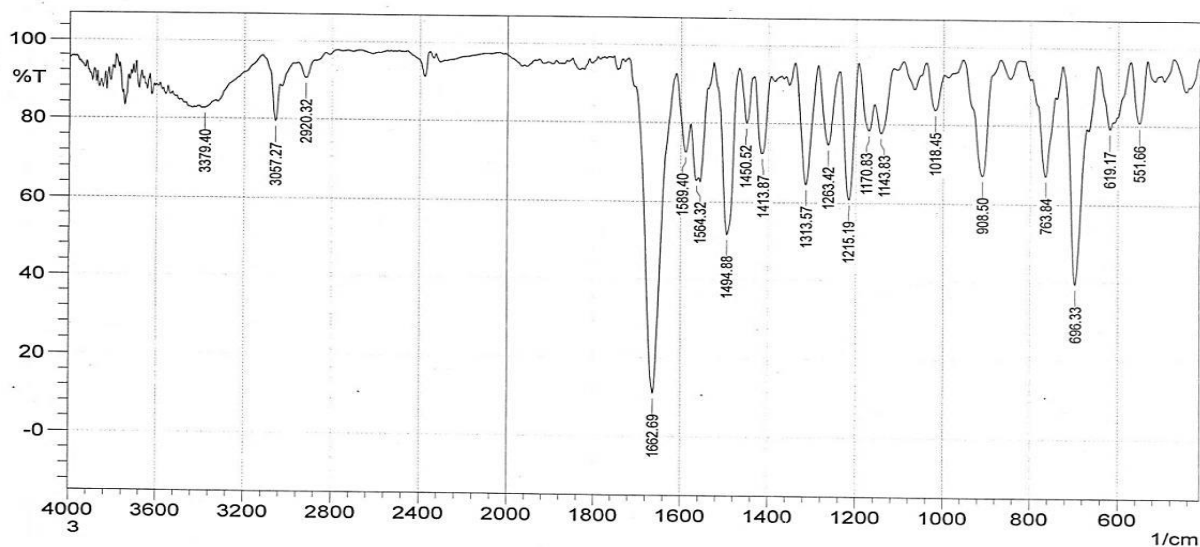
NO.	Compounds	ν (C=N)	ν (O-H)	ν (C=O)	ν (M-O)	ν (M-N)
L^1		1591	3375	1664		
1	[Mn L^1 Cl]Cl	1600	3396	1664	586	466
2	[Co L^1 Cl]Cl	1598	3383	1666	505	440
3	[Ni L^1 Cl]Cl	1599	3392	1662	584	430
4	[Cu L^1 Cl]Cl	1599	3390	1664	507	461
5	[Zn L^1 Cl]Cl	1602	3387	1662	510	445
L^2		1589	3379	1662		
6	[Mn L^2]Cl ₂	1601	3433	1676	503	440
7	[Co L^2]Cl ₂	1600	3391	1675	549	441
8	[Ni L^2]Cl ₂	1599	3394	1672	586	440
9	[Cu L^2]Cl ₂	1605	3406	1675	580	420
10	[Zn L^2]Cl ₂	1601	3393	1674	560	435



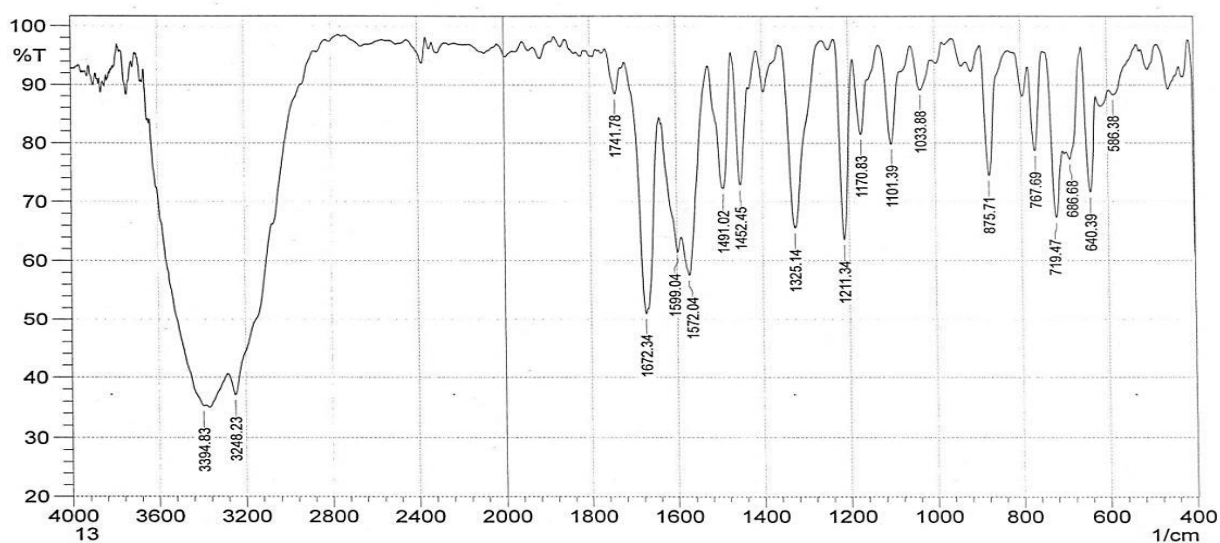


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(b)



(c)



(d)

Figure 2. The IR spectrum of (a)L¹,(b)[NiL¹Cl]Cl,(c) L²,(d)[NiL²]Cl₂

Ultraviolet visible (UV) and magnetic properties

The electronic spectra and magnetic properties are very useful in elucidating structure, the results of both measurements are specified in Table 4. The electronic spectra of the ligands in DMSO show the aromatic intense bands at 39062 cm⁻¹ and 45871 cm⁻¹ assigned to ($\pi \rightarrow \pi^*$) due to transitions relating molecular orbitals situated on the benzene ring of the ligand group and at 31250 and 38759 cm⁻¹ attributed to the ($n \rightarrow \pi^*$) transition due to azomethine groups^{26,27}, which shifted in the spectrum of the complexes which confirms the coordination of ligands with the central ion²⁸⁻³⁰. In the spectrum of all complexes new bands appeared at the region (20876-29673) cm⁻¹ may be attributed to LMCT transition^{31,32}

At room temperature Bohr magneto values of Mn(II) complexes (1 and 6) were (5.71 and 6.10) B.M. supports the tetrahedral arrangement resulting from the presence of the five unpaired electrons which confirms high spin state. The electronic spectra of Mn (II) complexes did not give clear absorption which can indicate to d-d transition. The d-d transitions in the Mn(II) tetrahedral environment is spin-forbidden but no longer parity forbidden, these transitions are ~100times stronger, therefore it is possible to appoint the structure of Mn(II) complexes depending on the results of other measurements namely³³⁻³⁵.

The Bohr magneto values of Co (II) complexes (2 and7) at (4.15 and 4.33) B.M. which refers to the tetrahedral structure of these complexes, their

electronic spectra exhibit one transition ν_3 $^4A_{2(F)} \rightarrow ^4T_{1(P)}$ consisting of two humps located at 14857 and 15083 cm⁻¹ and 16826 and 16477 cm⁻¹ respectively, the fission of this band which caused by Jahn-Teller type of the tetrahedral structure in the excited state³⁶⁻³⁸, the other two bands ν_1 and ν_2 are below the spectrophotometer limits.

The Bohr magneto values of Ni(II) complexes (3and8) were found to be (3.91 and 3.77) B.M. and its electronic spectrum also recorded a clear absorption band ν_3 at (14772 and 14084) cm⁻¹ respectively due to $^3T_{1(F)} \rightarrow ^3T_{1(P)}$ transition in tetrahedral geometry³⁹⁻⁴¹, the other two bands ν_1 and ν_2 are located in the lower part of the electronic spectrum which is below the scale limits, the obtained magnetic moment values of Cu (II) complexes (4 and9) found to be (2.44 and 2.59) B.M. this confirms the existence of unpaired electron in the electronic spectrum of these complexes a wide band was seen at 13973 and 14992 cm⁻¹ respectively attributed to $^2T_2 \rightarrow ^2E$ transition which suggests the tetrahedral structure of these complexes⁴².

The electronic spectra of Zn(II) complexes (5 and 10) show only a band at 28255 and 28385 cm⁻¹ respectively which represents the charge transfer spectra.

The Zn(II) complexes are expected to be diamagnetic and their geometry is most probably similar to the Co(II), Ni(II) and Cu(II) complexes based on the rest of results for the other measurements in particular the results of metal content and IR spectra^{43,44}.

Table 4. Results of magnetic moment and UV-vis

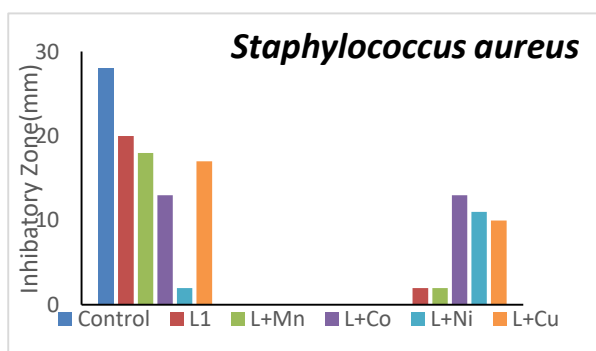
No	Compounds	μ_{eff} B.M.	Band maxima λ cm ⁻¹	Structure	Assigned transition
L ¹	C ₃₁ H ₂₆ N ₄ O ₂		39062,31250		
1	[MnL ¹ Cl]Cl	5.7	28248	Td	C.T
2	[CoL ¹ Cl]Cl	4.15	14857,16826,23255	Td	$^4A_{2(F)} \rightarrow ^4T_{1(P)}$, C.T
3	[NiL ¹ Cl]Cl	3.91	14772,26231	Td	$^3T_{1(F)} \rightarrow ^3T_{1(P)}$, C.T
4	[CuL ¹ Cl]Cl	2.44	13973,27781	Td	$^2T_2 \rightarrow ^2E$, C.T
5	[ZnL ¹ Cl]Cl	Dia	28255	Td	C.T
L ²	C ₃₁ H ₂₅ N ₄ O ₂ Cl		45871,38759		
6	[MnL ²]Cl ₂	6.10	29673	Td	C.T
7	[CoL ²]Cl ₂	4.33	15083,16477,20876	Td	$^4A_{2(F)} \rightarrow ^4T_{1(P)}$, C.T
8	[NiL ²]Cl ₂	3.77	14084,23277	Td	$^3T_{1(F)} \rightarrow ^3T_{1(P)}$, C.T
9	[CuL ²]Cl ₂	2.59	14992,24277	Td	$^2T_2 \rightarrow ^2E$, C.T
10	[ZnL ²]Cl ₂	Dia	28385	Td	C.T

Antibacterial activity

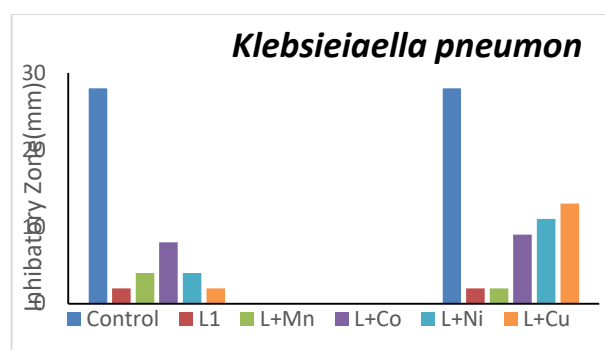
The evaluation of biological activity for the ligands and complexes with 1×10^{-3} M concentrations was studied against selected types of G(+) bacteria *Staphylococcus aureus* and G(-) bacteria *Klebsiella pneumoniae*. The obtained results have been tabulated and shown in Table 5, Fig. 4 and Fig. 5 and compared with a standard drug (Ciprofloxacin). The evaluation showed that the ligands and their complexes have an activity described as less than effective of ciprofloxacin against the bacteria used in this research.

Table 5. The bacterial examination results of L¹, L² and their complexes.

No.	Compound	Staphylococcus aureus (mm)	Klebsiella pneumonia (mm)
L ¹	C ₃₁ H ₂₆ N ₄ O ₂	20	2
1	[MnL ¹ Cl]Cl	18	4
2	[CoL ¹ Cl]Cl	13	8
3	[NiL ¹ Cl]Cl	2	4
4	[CuL ¹ Cl]Cl	17	2
L ²	C ₃₁ H ₂₅ N ₄ O ₂	2	2
	Cl		
6	[MnL ²]Cl ₂	2	2
7	[CoL ²]Cl ₂	13	9
8	[NiL ²]Cl ₂	11	11
9	[CuL ²]Cl ₂	10	13
Control	Ciprofloxacin	28	28



(a) *Staphylococcus aureus*



(b) *Klebsiella pneumoniae*

Figure 4. Antibacterial activity of compounds agents (a) *Staphylococcus aureus* (b) *Klebsiella pneumoniae*

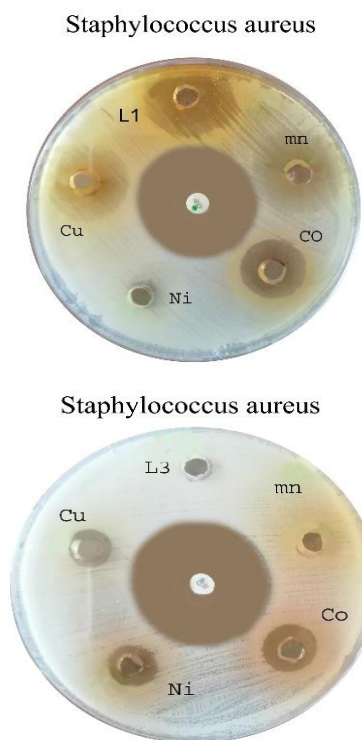


Figure 5. Antibacterial activity of the compounds

Conclusion

New complexes of Schiff base ligands were prepared and diagnosed by various physical and chemical methods. The data revealed that (L¹) ton in [ML¹Cl] Cl acts as a polydentate ligand and coordinated with the central atoms through the oxygen atom of Phenol group and nitrogen atoms of the azomethin groups, (L²) in [M L²] Cl₂ coordinated through oxygen atom of terminal carbonyl

group in addition to the atoms listed above and all complexes classified as mononuclear with tetrahedral geometry. The ligands and their complexes were examined against both Gram (+) and Gram (-) bacteria, the test results were compared with the standard drugs and showed that the ligands and complexes have an activity described as less than the drug.

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Author's Declaration

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are ours. Furthermore, any Figures and images, that are not ours, have been included with the necessary permission for re-publication, which is attached to the manuscript.
- No animal studies are present in the manuscript.
- No human studies are present in the manuscript.
- No potentially identified images or data are present in the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee at University of Mosul.

Author's Contributions Statements

A.M.M. Carried out the experiment and analysed all parameters, I.A.S. Collected the samples and taking

measurements, K.H.N. Conceived the idea, supervised the project and wrote the manuscript

References

1. Qin W, Long S, Panunzio M, Biondi S. Schiff bases: A short survey on an evergreen chemistry tool. *Molecules*. 2013 Oct 8; 18(10): 12264-89. <https://doi.org/10.3390/molecules181012264>
2. Al-Hialy QH, Al-Nama KS. Synthesis, Antibacterial Activity and DFT Calculation of Co (II) and Ni (II) Schiff Bases Complexes Derived from Acenaphthenequinon and Phenylenediamine. *Egypt J Chem*. 2021 Nov 1; 64(11): 6429-38. <https://dx.doi.org/10.21608/ejchem.2021.78906.3856>
3. Jona P, Gnana V GK, Sobana I R. Synthesis, Characterization Andbiological Screening of Some Sulpha Pyridine Schiff Base Transition Metal Complexes, *Int J Sci Technol Res*. 2019; 8(11): 2728-2735.
4. Nartop D, Öğütçü H. Synthesis of new unsymmetrical Schiff bases as potential antimicrobial agents. *Sinop Üniversitesi Fen Bilimleri Dergisi*. 2020 Jun 6; 5(1): 13-25. <https://doi.org/10.33484/sinopfdb.640297>
5. Sridevi G, Arul Antony S, Angayarkani R. Schiff base metal complexes as anticancer agents. *Asian J Chem*. 2019; 31(3): 493-504. <https://doi.org/10.14233/ajchem.2019.21697>
6. Mohammed ER, Saied SM, Saleh MY. Synthesis, Characterization and Biological Evaluation Study of Cephalixin (Ceph) and Isatin Schiff base and Its Complexation with Some Divalent Metal Ions. *Egypt J Chem*. 2022 Jul 1; 65(7): 595-603. <https://doi.org/10.21608/ejchem.2021.106994.4914>
7. Maity D. Biological Applications of Schiff base Metal Complexes-A Review. *Int J Res Analytic Rev*. 2019; 6(2): 471-8.
8. Solati H, Sahebalzamani M, Adhami Moghadam F. Effect of Family-Based Care Training by Tele-nursing on Emotional Reactions in Mothers of Children with Bone Marrow Transplantation. *J. Mazandaran Univ Med Sci*. 2021 Jan 10; 30(192): 156-61.
9. NARTOP D, ÖĞÜTCÜ H. Synthesis of new unsymmetrical Schiff bases as potential antimicrobial agents. *Sinop Üniversitesi Fen Bilimleri Dergisi*. 2020 Jun 6; 5(1): 13-25. <https://doi.org/10.33484/sinopfdb.640297>
10. Salih KS, Shraim AM, Al-Mhini SR, Al-Soufi RE, Warad I. New tetradentate Schiff base Cu (II) complexes: synthesis, physicochemical, chromotropism, fluorescence, thermal, and selective catalytic oxidation. *Emergent mater*. 2021 Apr;4: 423-34. <https://doi.org/10.1007/s42247-021-00183-9>
11. Jima'a RB, Shaalan ND. Synthesis, Characterization, and Biological Activity of New Metal Ion Complexes with Schiff Base Derived from 2-Acetylthiophene and Isatin. *Egypt J Chem*. 2022 Dec 1; 65(132): 1409-19. <https://doi.org/10.21608/ejchem.2022.124768.5552>
12. Teran R, Guevara R, Mora J, Dobronski L, Barreiro-Costa O, Beske T, et al. Characterization of antimicrobial, antioxidant, and leishmanicidal activities of Schiff base derivatives of 4-aminoantipyrine. *Molecules*. 2019 Jul 24; 24(15): 2696. <https://doi.org/10.3390/molecules24152696>
13. Jailani AK, Gowthaman NS, Kesavan MP. Synthesis, Characterisation and Biological Evaluation of Tyramine derived Schiff base Ligand and Its Transition Metal (II) Complexes. *Karbala Int J*

- Modern Sci. 2020; 6(2): 15.
<https://doi.org/10.33640/2405-609X.1637>
14. Putaya HA, Ndahi NP, Bello HS, Mala G, Osunlaja AA, Garba H. Synthesis, characterization and antimicrobial analysis of Schiff bases of o-phenylenediamine and 2-aminopyridine-3-carboxylic acid with ofloxacin and their metal (II) complexes. *Int J Biol Chem Sci.* 2020 Apr 3; 14(1): 263-78.
<https://doi.org/10.4314/ijbcs.v14i1.22>
 15. Al-Jiboury MM, Al-Nama KS. Preparation, Characterization and Biological Activities of some Unsymmetrical Schiff Bases Derived from m-phenylenediamine and their Metal Complexes. *Rafidain J Sci.* 2019; 28(2): 23
<https://doi.org/10.33899/rjs.2019.159965>
 16. Jassim SA, Khaleel AM. Characterization and synthesis of new Schiff base compound from levofloxacin and l-cysteine with its cu (ii) and pt (iv) complexes and estimation antibacterial and antifungal activities. *Biochem. Cell Arch.* 2021 Apr 2; 21.
 17. Elshafie HS, Sadeek SA, Camele I, Mohamed AA. Biochemical characterization of new gemifloxacin schiff base (GMFX-o-phdn) metal complexes and evaluation of their antimicrobial activity against some phyto-or human pathogens. *Int J Mol Sci.* 2022 Feb 14; 23(4): 2110.
<https://doi.org/10.3390/ijms23042110>
 18. Ibraheem IH, Sadiq AS, Al-Tameemi M, Alias MF. Synthesis, Spectral Identification, Antibacterial Evaluation and Theoretical Study of Co, Fe, Rh and Pd Complexes for 2-benzoylthiobenzimidazol. *Baghdad Sci J.* 2022 Dec 1; 19(6): 1326-1326.
<https://doi.org/10.21123/bsj.2022.6704>
 19. Wahba O, Hassan AM, Naser A, Hanafi A. Preparation and spectroscopic studies of some copper and nickel Schiff base complexes and their applications as colouring pigments in protective paints industry. *Egypt J Chem.* 2017 Feb 1; 60(1): 25-40.
<https://doi.org/10.21608/ejchem.2017.517.1000>
 20. Aljamali N, Hadi M, Mohamad M, Salih L, Aljamali N. Review on imine derivatives and their applications. *Int J Photochem.* 2019 Jul 17; 5: 20-32.
 21. Hussein KA, Mahdi S, Shaalan N. Synthesis, Spectroscopy of New Lanthanide Complexes with Schiff Base Derived From (4-Antipyrinecarboxaldehyde with Ethylene Di-Amine) and Study the Bioactivity. *Baghdad Sci J.* 2023 Apr 1; 20(2): 0305-0305.
<https://doi.org/10.21123/bsj.2022.7088>
 22. Patai Ed S, *The Chemistry of Carbone-Nitrogen Double Bond*, Jhon Wiley and Sons, New York., 1972. 235 – 253.
 23. Aliyu HN, Suleiman Z. Spectrophotometric analysis on anticonvulsant bis (N-isatin benzene-1-hydroxy-2-iminato) Mn (II), Fe (II) and Co (II) complexes. *Global Adv Res J Microbiol.* 2012; 1(5): 079-83.
 24. Dawood MN, Jamel HO. Synthesis, Characterization and Evaluation of Biological Activity of new Ligand Derived from 4-Aminoantipyrine, 2-Mercaptobenzoxazole and its Complexes with Some Metal Ions. *HIV Nursing.* 2022 Nov 4; 22(2): 2981-93.
 25. Khan Z, Maqsood ZT, Tanoli MA, Khan KM, Iqbal L, Lateef M. Synthesis, characterization, in-vitro antimicrobial and antioxidant activities of Co²⁺, Ni²⁺, Cu²⁺ and Zn²⁺ complexes of 3-(2-(2-hydroxy-3-methoxybenzylidene) hydrazono) indolin-2-one. *J Basic Appl Sci.* 2015; 11: 125.
<https://doi.org/10.6000/1927-5129.2015.11.17>
 26. Boulechfar C, Ferkous H, Delimi A, Djedouani A, Kahlouche A, Boublia A, et al. Schiff Bases and Their Metal Complexes: A review on the history, synthesis, and applications. *Inorg Chem Commun.* 2023 Jan 26; 110451.
<https://doi.org/10.1016/j.inoche.2023.110451>
 27. Mirza AS, Nema B, Teli SA, Dar DA, Khalid S. Noval 3d Series transition metal Complexes with Schiff base AEBTC (2-Thiophenecarboxaldehyde and 2-Amino-6-Ethoxybenzothiazole): Synthesis, Characterization, and Biological Importance. *Bulletin of Environment, Pharmacol. Life Sci.* 2023 March; 12(4): 27-35.
 28. Ajaz A, Shaheen MA, Ahmed M, Munawar KS, Siddique AB, Karim A, et al. Synthesis of an amantadine-based novel Schiff base and its transition metal complexes as potential ALP, α -amylase, and α -glucosidase inhibitors. *RSC Adv.* 2023; 13(5): 2756-67.
<https://doi.org/10.1039/D2RA07051K>
 29. Hussaini SY, Abdulkadir M, Panda NA, Fagge II, Danjaji HI, Sani S. Synthesis, Analysis and Antibacterial Studies of Co (II) and Ni (II) Schiff Base Complexes Derived from 2, 4-Dinitrophenylhydrazine and Benzaldehyde. *Sch Int J Chem Mater Sci.* 2023; 6(3): 47-52.
<https://doi.org/10.36348/sijcms.2023.v06i03.002>
 30. Pathan AH, Naik GN, Bakale RP, Machakanur SS, Gudasi KB. Ligational behavior of new mononucleating NOO ethyl pyruvate Schiff base towards 3d metal (II) ions: an emphasis on antiproliferative and photocleavage property. *Appl Organomet Chem.* 2012 Mar; 26(3): 148-55.
<https://doi.org/10.1002/aoc.2831>
 31. Arumugam A, Shanmugam R, Munusamy S, Muhammad S, Algarni H, Sekar M. Study of the Crystal Architecture, Optoelectronic Characteristics, and Nonlinear Optical Properties of 4-Amino Antipyrine Schiff Bases. *ACS omega.* 2023 Apr 17; 8(17): 15168-80.
<https://doi.org/10.1021/acsomega.2c08305>
 32. Zeyrek CT, Elmali A, Elerman Y, Svoboda I. Crystal structure and magnetic exchange interaction in a binuclear copper (II) schiff base complex with a bridging m-phenylenediamine ligand. *Zeitschrift für Naturforschung B.* 2005 Feb 1; 60(2): 143-8.
<https://doi.org/10.1515/znb-2005-0203>

33. Al-Shammari WA, Lateef SM. Synthesis, structural, thermal and biological studies of ligand derived from anthrone with 4-aminoantipyrine and its metallic complexes. Chem Methodol. 2022; 6: 548-59. <https://doi.org/10.22034/CHEMM.2022.339536.1499>
34. Dilawer Issa K, Rasul Braiem R. Green and highly efficient synthetic approach for the synthesis of 4-aminoantipyrine Schiff bases. Chem Rev Let. 2022 Aug 1; 6(1): 2-6. <https://doi.org/10.22034/crl.2022.347066.1171>
35. Shokohi-Pour Z, Chiniforoshan H, Momtazi-Borojeni AA, Notash B. A novel Schiff base derived from the gabapentin drug and copper (II) complex: Synthesis, characterization, interaction with DNA/protein and cytotoxic activity. J Photochem Photobiol B: Biol. 2016 Sep 1; 162: 34-44. <https://doi.org/10.1016/j.jphotobiol.2016.06.022>
36. Tapabashi NO, Taha NI, El-Subeyhi MN. Synthesis and Characterization of Some New Azo-Schiff Bases as Energy Rich Candidate Compounds Derived from 1, 5-Diaminoanthraquinone by Fusion Method. Kirkuk Univ J Sci Stud. 2021 Jun 1; 16(2): 51-63. <https://doi.org/10.32894/kujss.2021.168662>
37. Al-Labban HM, Sadiq HM, Aljanaby AA. Synthesis, Characterization and study biological activity of some Schiff bases derivatives from 4-amino antipyrine as a starting material. J Phys Conf Ser. 1294, 052007:1-8. <https://doi.org/10.1088/1742-6596/1294/5/052007>
38. Lever AP. Inorganic electronic spectroscopy. Studies in physical and theoretical chemistry. 1984; 33.
39. Teran R, Guevara R, Mora J, Dobronski L, Barreiro-Costa O, Beske T, et al. Characterization of antimicrobial, antioxidant, and leishmanicidal activities of Schiff base derivatives of 4-aminoantipyrine. Molecules. 2019 Jul 24; 24(15): 2696. <https://doi.org/10.3390/molecules24152696>
40. Adithya Krishnan M, Saranyaparvathi S, Raksha C, Vrinda B, Girish CG, Kulkarni NV, et al. Transition Metal Complexes of 4-Aminoantipyrine Derivatives and Their Antimicrobial Applications. Russian J Coordin. Chem. 2022 Nov; 48(11): 696-724. <https://doi.org/10.1134/S1070328422110082>
41. Rakhi C, Shelly S. Spectral and pharmacological study of Cu (II), Ni (II) and Co (II) coordination complexes. Res J Chem Sci. 2011; 1(5): 87-90.
42. Seleem HS, Mostafa M, Stefan SL, Abdel-Aziz E. Structural diversity of 3d complexes of an isatinic quinolyl hydrazone. Res J Chem Sci. ISSN. 2011; 2231:606X.
43. Kulkarni PA, Habib SI, Saraf VD, Deshpande MM. Synthesis, spectral analysis and antimicrobial activity of some new transition metal complexes derived from 2, 4-dihydroxy acetophenones. Res J Pharm Biol Chem Sci. 2012; 3: 107-13.
44. Mahmoud WH, Refaat AM, Mohamed GG. Zinc complexes of novel Schiff bases derived from 1, 8(-Diaminonaphthalene: Synthesis, characterization and biological activity evaluation. 2018; 8(11): 53-61

تحضير، تشخيص فضلاً عن دراسة الفعالية البيولوجية لمعقدات بعض من ايونات الفلزات مع قواعد شيف المشتقة من 4 - امينو انتي بايرين

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الخلاصة

قواعد شيف متعددة السن ذوات الصيغة $L^1 =$ اورثو هايدروكسي انيلين - ٤ - امينو انتيبيرين (N',N بنزليدين) تم تحضيرها من تكثيف البنزائل و٤-امينو انتيبيرين مع ٢-امينو فينول - ١، ٢ امينو ٤ كلورو فينول بنسبة مولية (١:١:١) لكل مكون. واستخدمت هذه الليكاندات لتحضير المعقدات ذوات الصيغة $[ML^1 Cl]Cl$ و $[ML^2]Cl_2$ حيث ان $Zn(II)$ $M = Mn(II), Co(II), Ni(II)$. تم استخدام التقنيات التالية لتشخيص الليكاندات والمعقدات: مطيافية الأشعة فوق البنفسجية والأشعة تحت الحمراء - المرئية وطيف الرنين النووي المغناطيسي للبروتون $^1H NMR$ والتحليل الدقيق لايونات العناصر والكربون والهيدروجين والنترجين بالإضافة الى التوصيلية الكهربائية المولارية وقياسات الحساسية المغناطيسية. دلت القياسات التي تم الحصول عليها من التوصيلية الكهربائية المولارية والتي تم اجرائها بمحلول DMSO ان جميع معقدات (L^1) كانت الكتروليتية بنسبة (١:١) بينما معقدات (L^2) كانت الكتروليتية بنسبة (1:2). كما اظهرت نتائج القياسات المستخدمة في هذه الدراسة ان مواقع التناسق لليكاند (L^1) مع الأيون المركزي كانت من خلال ذرة الاوكسجين الفينولية وذرتين نيتروجين لمجموعتي الازوميثين اما بالنسبة لـ (L^2) فبالإضافة للذرات المانحة المذكورة سابقا يتم التناسق مع ذرة الاوكسجين لمجموعة الكربونيل الطرفية. جميع المعقدات اظهرت التناسق الرباعي بشكل هندسي رباعي السطوح ايضا تمت دراسة الفعالية البيولوجية للمركبات المحضرة ضد البكتريا موجبة الكرام ($Staphylococcus aureus$ (+) وسالبة الكرام (-) *Klebsiella pneumonia*

الكلمات المفتاحية: 4-aminoantipyrine ، النشاط البيولوجي ، المركب المعدني ، Polydentate ، قاعدة شيف.