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# Preparation and Characterization of complexes with 2-amino pyrimidine and 4-methyl imidazole ligand and Study of Biological screening of the Au (III) Complex

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**Abstract**

The many of metal ions-complexes of Co(II), Ni(II), Cu(II), Zn(II), Cd(II), Hg(II) , and Au(III) have been preparation and characterization using novel azo-Schiff base ligand derived from azo compound namely; **(**1,5-dimethyl-4-((4-methyl-1H-imidazol-2-yl)diazenyl)-2-benzyl-1,2-dihydro-3H-pyrazol-3-one ) with 2-amino pyrimidine. The structures of the new ligand azo-schiff base and their transition metal complexes are characterized using several techniques, including elemental analysis (C.H.N), electronic spectral, IR spectral studies,$^{1}$HNMR,$^{13}$CNMR, magnetic measurements, molar conductance and mass spectra The data show that these complexes have composition of $[ML_2]Cl_2$ where M = Co(II), Ni(II), Cu(II) ,Zn(II),Cd(II) and Hg(II), $[MLCl]Cl_2$ where M=Au(III). The electronic spectral, and magnetic susceptibility data of the complexes suggest octahedral geometry of all complexes, except the Au(III) complex suggest a square planar geometry. In addition, these compounds were also the co-ordination sites are the azomethine nitrogen , azo nitrogen atom and imidazole nitrogen atom of the ligand. The ligand forms shap as natural tridentate manner. The biological screening effect of the Au(III) complex are tested against *against several organisms , staphylococcus aureus, Esherichia coli, Aspegills niger, Aspegills flavus are reported* . all data of biological activaity gave good results about good inhibition for complexes against microbs, the results show the highest inhibitory effect for complex**.**

***Key word:*** Novel azo- schiff base ligand**,** 4-methyl--imidazole, 2-amino pyrimidine **,**Biological screening

## Introduction

New ligands in coordination chemistry were gave new importance in inorganic chemistry field represented by Azo compounds are Containing (-N=N-) group(1), The presence of the unshared electron pair on the nitrogen atom made these compounds of great importance in many areas as a result of the effectiveness shown by these compounds and their derivatives(2). It have significant importance for construction of this well-defined architecture because it used as antibacterial(3), anticancer (4)and antimicrobial evaluation in medicinal and pharmaceutical fields(5-6). Also Schiff bases of chelating compounds containing a group (N=C-) with high activity(7), and its derivatives of 4-aminoantipyrine and its complexes have wide applications both in the medical field, pharmaceutical(8) ,as antitoxtante (9),,antioxidant(10) chemical stimulation(11), in addition to their physiological applications as ,and impartment as antibacterial(12) and antifungal(13). The aim of this paper is to synthesize, characterize and study the biological screening of the gold complex as against several organisms of the new tridentate azo Schiff-base ligand, N-(1,5-dimethyl-4-((E)-(4-methyl-1 H-imidazol-2-yl)diazenyl)-2-phenyl-1H-pyrazol-3(2H)-ylidene)pyrimidin-2-amine and some of its transition metal complexes.

**Materials:** All chemicals were supplied by BHD and Sigma Aldrich and used without further purification.

**Measurement :**
The electro thermal melting point model 9300 was used to measure the melting point of the ligand and its complexes. Elemental analyses were carried out by means of micro analytical unit of 1180 C. H. N

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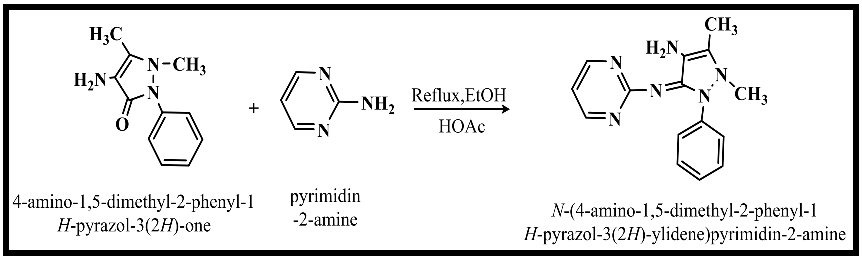
elemental analyzer. Electronic spectra were recorded on Shimadzu spectrophotometer double beam model 1700 Uv-Vis spectrophotometer-FTIR spectra were recorded in KBr disc on FTIR Shimadzu spectrophotometer model 8400 in wave number 4000-400 $cm^{-1}$. $^{1}H$-NMR &$^{13}C$-NMR-spectra in (ppm) unit were operating in DMSO *-d6* as solvent using (Bruker- Ultra Shield 3000 MHz Switzerland).And Mass Spectra were recorded on AB Sciex 3200 QTRAP LC/MS/MS , (Mass range - m/z 5-2000-quad mode and 50-1700- linear ion trap mode). Magnetic susceptibility measurements were carried out on a balance magnetic MSB-MKI using faraday method. The diamagnetic corrections were made by Pascal's constants.

**Preparation of the new Schiff base ligand N-(4 -amino-1,5-dimethyl-2-phenyl-1H- pyrazol-3(2H)-ylidene)pyrimidin-2-amine :**

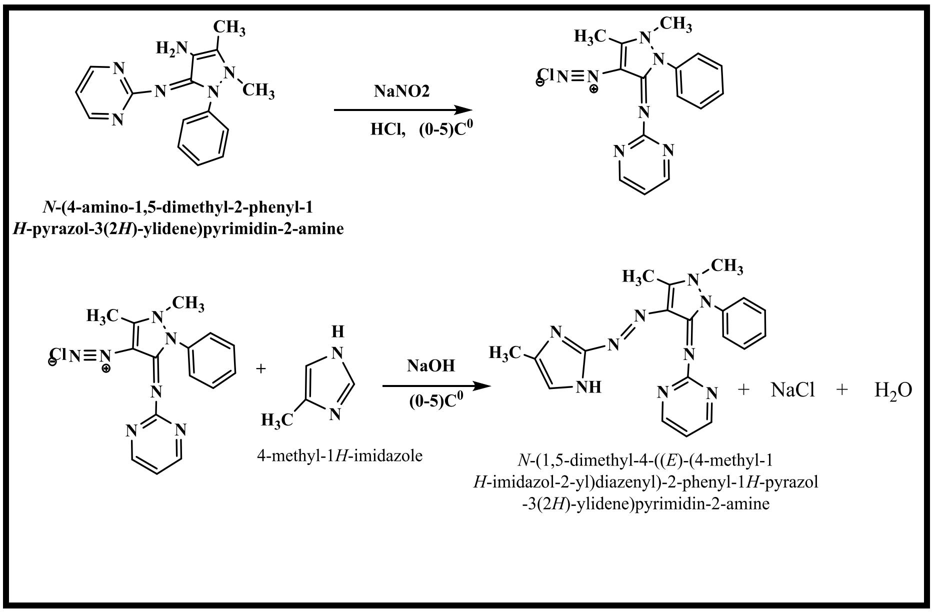
The schiff base ligand is prepared by condensation of 4-amino antipyrine with 2-amine pyrimidine in equimolar (1:1) mole ratio, in absolute alcohol. Few drops of glacial acetic acid were added to the reaction mixture and refluxed for 35 hrs. The product was recrystallized from ethanol, and dried over anhydrous $CaCl_2$. The reaction mixture gave one product.




**Scheme-1:preparationof the new ligand**

**Preparation of the novel azo- Schiff base ligand:**

N-(4-amino-1,5-dimethyl-2-phenyl-1H-pyrazol-3(2H)-ylidene)pyrimidin-2-amine (2.80g,0.01mol) was dissolved in (3ml conce.HCl,50ml of water),it is then put in cooled bath. Then 40ml of (0.72g,0.01mol) Sodium nitrite(NaNO2)- solution was mixed with the above solution with constant stirring. A cool solution (60ml)10% NaOH with (0.82g,0.01mol) 4-methyl-1H-imidazole was mixed drop wise to the resulting solution with stirring besides to the mixture that was left to 9hr at 0 C◦. Red precipitate was filtered and recrystallized from hot ethanol and then dried in over at 60 C◦ for 14hours.




**Scheme-1:preparation of the novel azo- Schiff base ligand**

**Preparation of metal complexes:**

The metal complexes were prepared by mixing of 40ml ethanol solution of ($CoCl_2.6H_2O$, $NiCl_2.6H_2O$, $CuCl_2.2H_2O$, $ZnCl_2$, $CdCl_2.2H_2O$, $HgCl_2.2H_2O$, $NaAuCl_4.H_2O$) with 40ml ethanol solution of (azo-Schiff base) ligand in (1:2) (metal: ligand ) ratio except the Au(III) complex was (1:1) (metal: ligand ) ratio. the resulting mixture was refluxed for 2h . The product was isolated after reduced of volume by evaporation . It was filtered off , washed with ethanol and dried under vacuum . The complexes obtained are listed in table 1.

**Biological part:** All Chemicals and biological materials were supplied from(Sigma, Difco,) (USA).

**Methods: Determination of antimicrobial activity.**
***Bactra-activity:***

Pathogenic biological, *Esherichia coli,and Staphylococcus aureus* were used to test the antimicrobial activity of the Au(III) complex. The nutrient agar medium was prepared and quantity of 10ml of the medium was poured into the sterilized petri plates and allowed to solidify. The stock solution (0.01 mol) was prepared by dissolving the compound in DMSO and the solutions were serially diluted in order to find the MIC values . The plates were inoculated with spore suspension of pathogenic bactericides. By using the sterilized cork bore, were dug in the center of the culture plates, the test complex solution was added (0.5 ml) to these wells and the plates were incubated at $25^{0}$c for 24 hour. Then the inhibition zone appeared around the wells in each plate was measured and recorded as the cytotoxic effect of the appropriate complex.

***Fungal activity:***

The complex was tested again fungi such as *Aspergillus niger, Aspergillus flavus,* cultured On potato dextrose agar as medium. In atypical procedure, as well was made on the agar medium inoculated with the fungi- the well was filled with the test. Solution using a micropipette and the plate was incubated at 30 $c^{0}$ for 72 hr. During this period, the test solution diffused and growth of the Inoculated fungi was affected.

**Results and Discussion:**

All our complexes are Freely soluble in DMSO, DMF, Methanol , Ethanol and water .Also They are stable in air . The metal complexes were characterized by elemental analysis IR, UV-Vis ,Mass spectra, molar conductivities , magnetic susceptibility, and$^{13}$CNMR, $^{1}$H NMR spectra . The analytical data of the complexes are in agreement with the experimental data .The value reveal that the metal to ligand ratio is (1:2) ratio except the Au(III) complex was (1:1) (metal: ligand ) ratio and are presented in table 1 . The magnetic susceptibility of the chelate complexes at room temperature were consistent with octahedral geometry, except the Au(III) complex suggest a square planar geometry around the central metal ion. Most of chelate complexes prepared in this work showed higher conductivity values of the complexes. This proves that complexes have electrolytic nature.

**Micro analysis:**

The elemental analysis data of1:2 [M:L] ratio complexes except the Au(III) complex was (1:1) (metal: ligand ) ratio showed that the theoretical values are in a good agreement with the found data ,as listed in table(1) . The purity of novel azo-schiff base ligand were tested by TLC technique and C,H ,N elemental analysis .

**Infrared Spectra studies of the novel azo-schiff ligand and its complexes:**

The important infrared spectral bands for the synthesized azo-schiff ligand and its chelate complexes are given in table.2. The IR spectrum of the ligand shows characteristic bands at(1656and 1448) $cm^{-1}$ due to the (C=N)and (N=N) functional groups respectively[14]. .The (C=N) and (N=N) bands in the free ligand shift to (1643-1624 )$cm^{-1}$ and(1429-1408) $cm^{-1}$, respectively for the complexes.. These shifts confirm the coordination of the ligand via the nitrogen of azo methine and the azo groups to metal ions[15]. The absorption band in free ligand observed at 3437$cm^{-1}$ attributed to the υ(NH) group[16].This band remains unchanged in the spectra of their complexes.. The absorption band in ligand azo-schiff observed at (1585) $cm^{-1}$ attributed to the υ(C=N) of the$N_3$ imidazole nitrogen. This band changed in the spectra of their complexes. This suggests that the υ(C=N) group is taking part in coordination[17]. New bands are attributed to υ(M-N) vibrations appearance in all complexes at (534-503) $cm^{-1}$ respectively[18,19].Representative example for their spectra is given in Fig1.

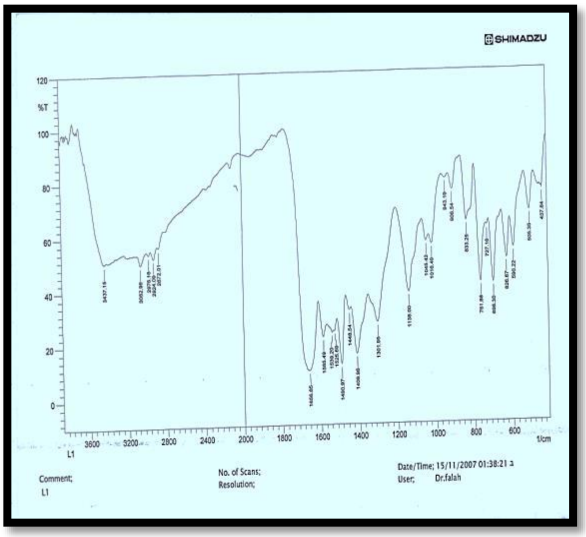



**Fig.1-A : IR spectrum of the azo schiff ligand**

**Table(1):Some physical properties and elemental analysis of azo- schiff base ligand and their metal complexes .**

| No | Formula | M.Wt | M. P $^0$C | Color | yield | Found (Calc.)% | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | | | | C% | H% | N% | M% |
| 1 | L : $C_{19}H_{19}N_9$ | 373.41 | 235-237 | Red | 90% | 61.11 ( 61.21) | 5.13 (5.10) | 33.76 (33.80) | ....... |
| 2 | [Co $C_{38}H_{38}N_{18}$] $Cl_2$ | 876.67 | >300 | Olive | 88% | 52.06 (52.44) | 4.37 (4.50) | 28.76 (28.84) | 6.72 (6.88) |
| 3 | [Ni $C_{38}H_{40}N_{18}O$]$Cl_2$ | 894.44 | >300 | Olive | 92% | 51.03 (51.00) | 4.51 (4.45) | 28.19 (28.33) | 6.56 (6.70) |
| 4 | ($C_{38}H_{38}N_{18}$)] $Cl_2$[Cu | 881.28 | >300 | Olive | 91% | 51.79 (51.60) | 4.35 (4.28) | 28.61 (28.52) | 7.21 (7.18) |
| 5 | [Zn($C_{38}H_{38}N_{18}$)] $Cl_2$ | 883.14 | >300 | Purpl e | 89% | 51.68 (51.60) | 4.34 (4.30) | 28.55 (28.50) | 7.41 (7.42) |
| 6 | [Cd($C_{38}H_{38}N_{18}$)] $Cl_2$ | 930.15 | 298 | Purpl e | 83% | 49.07 (49.00) | 4.12 (4.10) | 27.11 (27.09) | 12.09 (12.03) |
| 7 | [Hg ($C_{38}H_{38}N_{18}$)]$Cl_2$ | 1018.32 | >300 | Purpl | 85% | 44.82 | 3.76 | 24.76 | 19.70 |
| 8 | [Au($C_{19}H_{19}N_9$)] $Cl_3$ | 676.74 | 199-201 | Red | 83% | 33.72 | 2.83 | 18.63 | 29.11 |

**Table. 2: IR absorption bands of the azo- schiff base ligand and their metal complexes in cm$^{-}$**

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| **L= $C_{19}H_{19}N_9$** | 3437 | 1664 | 1656 | 1585 | 1448 | 1138 | --------- |
| **[Co (L )$_2$ ]$Cl_2$** | 3354 | 1664 | 1625 | 1587-1568 | 1421 | 1130 | 505 |
| **[Ni (L )$_2$ ]$Cl_2$ .$H_2O$** | 3352 | 1661 | 1624 | 1591-1545 | 1429 | 1145 | 509 |
| **[Cu (L )$_2$ ]$Cl_2$** | 3437 | 1670 | 1630 | 1593-1542 | 1415 | 1134 | 534 |
| **[Zn (L )$_2$ ]$Cl_2$** | 3435 | 1666 | 1635 | 1593 | 1417 | 1126 | 505 |
| **[Cd (L )$_2$ ]$Cl_2$** | 3419 | 1670 | 1635 | 1593 | 1411 | 1132 | 503 |
| **[Hg (L )$_2$ ]$Cl_2$** | 3442 | 1672 | 1643 | 1587 | 1408 | 1136 | 509 |
| **[Au LCl] $Cl_2$** | 3458 | 1668 | 1635-1627 | 1593-1525 | 1408 | 1143 | 509 |

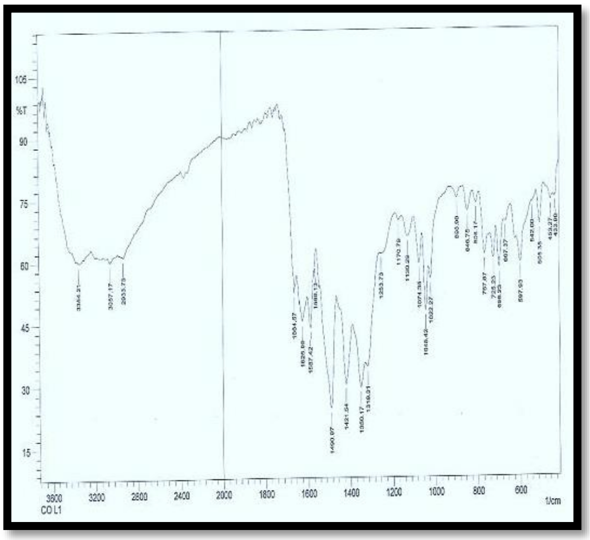


**Fig. 1-B : IR spectrum of Complex with Co (II)**

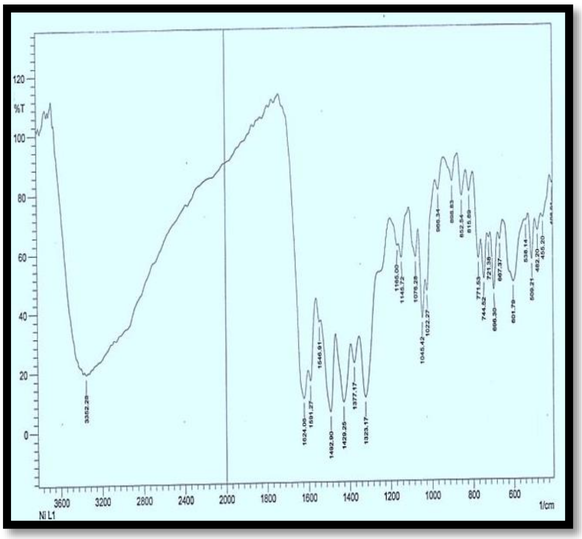


**Fig.1-C: IR spectrum of Complex with Ni(II)**

**Mass spectra:**

The mass spectra of synthesized novel azo- schiff base ligand and its Co(II) complex are recorded at room temperature. All fragments appeared parts of functional groups gave exactly our complexes and their ligands that indicate to suggestion shape ., The obtained molecular ion peaks confirm the proposed formulae for the synthesized compounds. The mass spectrum of Ligand show the molecular ion peak at m/z 373 (6%) compound ($C_{19}H_{19}N_9$) confirm the proposed formulae for the synthesized compound. Also The mass spectrum of the Co(II) complex exhibits the molecular ion peak at m/z 875.8 (0.5%) to the molecular formula ($Co(C_{38}H_{38}N_{18}Cl_2)$ consistent with the molecular weight of the Co(II) complex. (which are in good agreement with their formula as expressed from micro analytical data. The mass spectral data fragmentation of the novel azo-Schiff ligand and Co(II) - complex are shown in schemes 3 and 4..as shown in fig2.

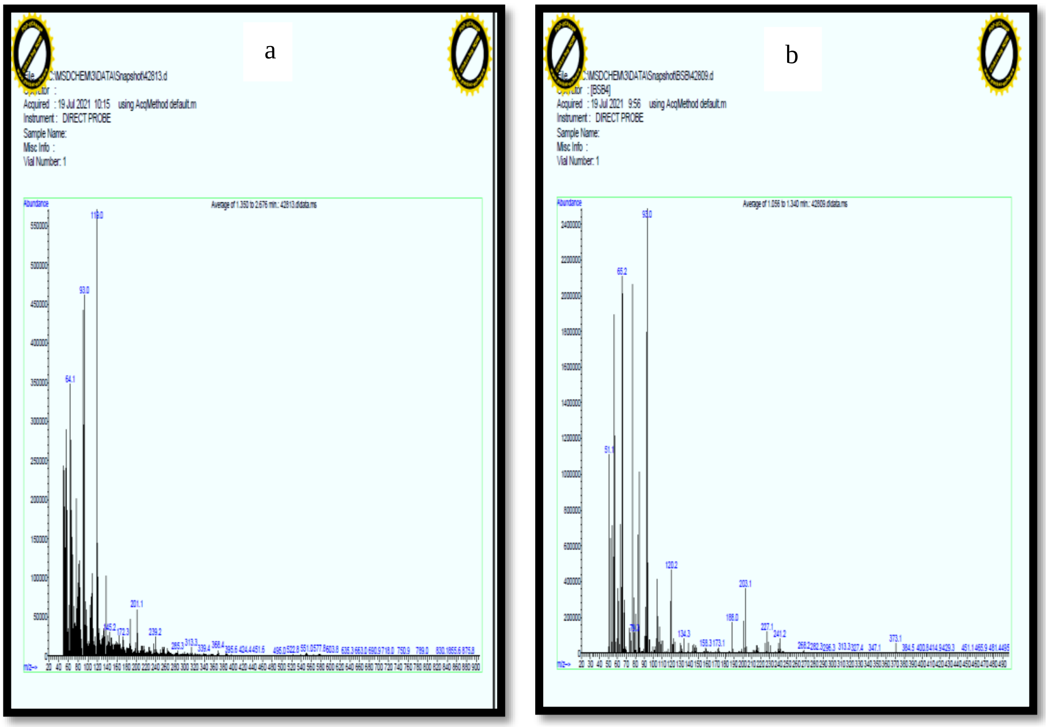
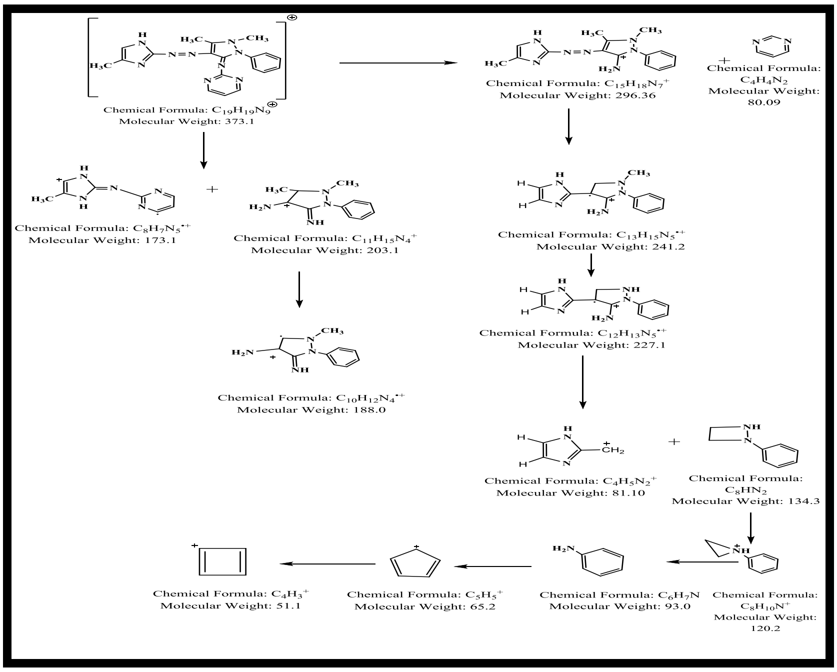
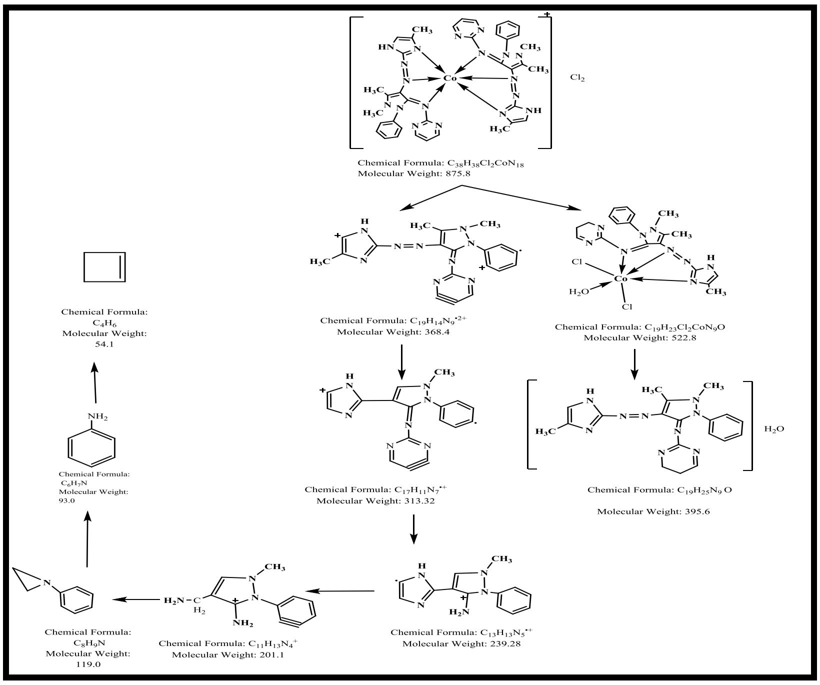



**Fig.2:Mass spectrum:(a) azo-schiff base ligand and (b) Co (II) complex**



**Scheme (3):- Mass spectrum fragmentation of azo -schiff ligand**




**Scheme (4):-Mass spectrum fragmentation of Co- complex**

**[1]HNMRspectra:**

The [1]HNMR spectrum of the novel azo- schiff base ligand shows the following signals : phenyl multiples at (7.3-7.7)ppm, $=C-CH_3$ at 2.7 ppm , $-N-CH_3$ 3.4ppm ,$CH_3$ imidazole ring at 1.8 -NH at 12.8 ppm[20,21,16].There is no appreciable change in all other signals in this complexes. as shown in fig.3.

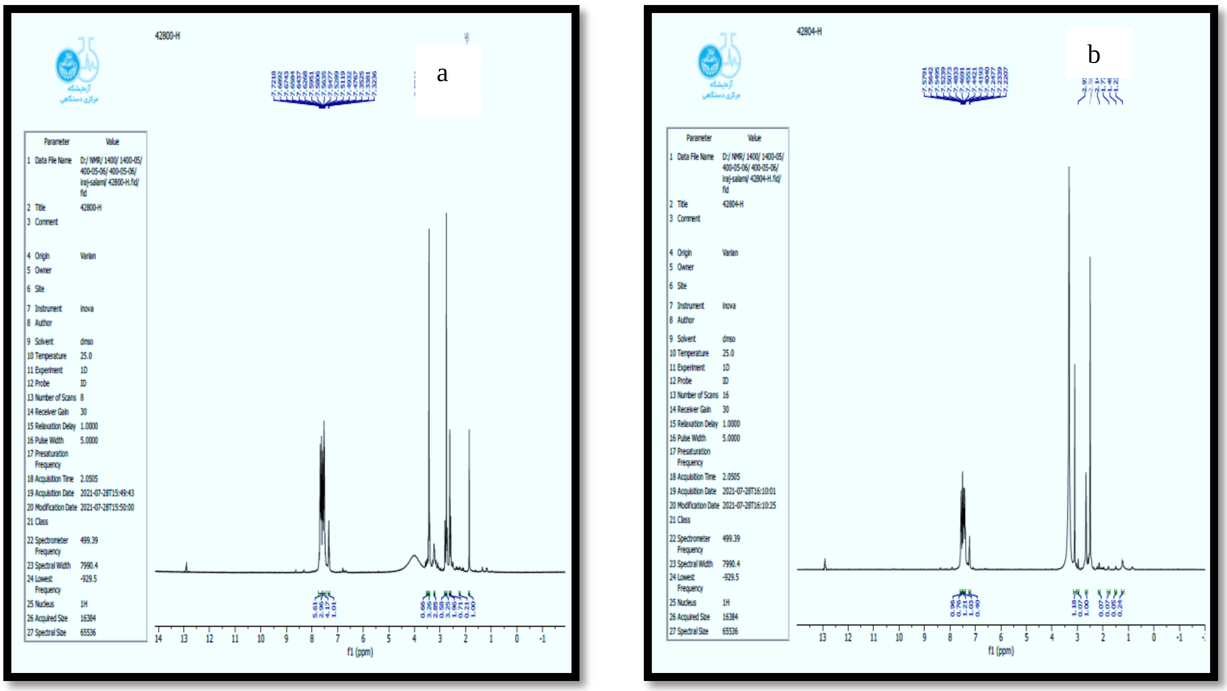



**Fig. 3:[1]H-NMR spectrum:(a)azo-schiff base ligand and (b) Co (II) complex**

**Electronic Spectra:**

The UV-Visible spectra were achieved in Ethanol solution ($10^{-5}$M).The spectrum of novel azo- schiff base ligand observes three bands, the first one at 204nm to ($\pi \rightarrow \pi^*$) transition of the imidazole heterocyclic ring [22],the second band at 327nm to($\pi \rightarrow \pi^*$) transition of benzene ring,which joined with imidazole via azo group[23], while the third band at 380nm to ($n \rightarrow \pi^*$) electronic transmission of ν(-N=N-), this band shifted to higher wave lengths[24].) during the coordination with metals[25].The table (3) refers to electronic transmission of (Co,Ni,Cu and Au)(III) complexes. The electronic spectrum of Co(II) complex displays three bands at 1092 nm (9157 $cm^{-1}$), 510 nm (19607 $cm^{-1}$) and 403 nm (24813 $cm^{-1}$). These three bands are assignable to$^4T_1g_{(F)} \rightarrow {}^4T_2g_{(F)} = v_1$ , $^4T_1g_{(F)} \rightarrow {}^4A_2g_{(F)} = v_2$ , $^4T_1g_{(F)} \rightarrow {}^4T_1g_{(p)} = v_3$ transitions respectively[26]. The electronic spectrum of Ni(II) complex exhibited three absorption bands , at 1092 nm (9157 $cm^{-1}$), 733 nm (13642 $cm^{-1}$) and 529 nm (18903 $cm^{-1}$). These bands may be assigned to $^3A_2g \rightarrow {}^3T_2g_{(F)\ (\upsilon 1)}$ , $^3A_2g \rightarrow {}^3T_1g_{(F)(\upsilon 2)}$ and $^3A_2g \rightarrow {}^3T_1g_{(p)(\upsilon 3)}$ transitions, respectively. The spectrum resemble those reported for octahedral complexes[27]**.** The electronic spectrum of the Cu(II) complex exhibited a single broad asymmetric band around at 520nm (19230 $cm^{-1}$). This band indicates the one transition $^2B_1g \rightarrow {}^2Eg$ (charge transfer)$_{(\upsilon 3)}$ , The broadness of the band may be due to Jahn-Teller distortion. All of these data suggested a distorted octahedral geometry around the Cu(II) ion[28]. The electronic spectrum of Au(III) complex displays one band at 552 nm (18115 $cm^{-1}$), This band is assignable to $^1A_1g \rightarrow {}^1B_1g$ [29]. The table (3) refers to electronic transmission of (Co,Ni,Cu and Au)(III) complexes while (Zn,Cd and Hg) (II) complexes appeared Charge Transfer (M→L,CT),because they are full with electrons ($nd^{10}$) [24]. as shown in Fig. 4.

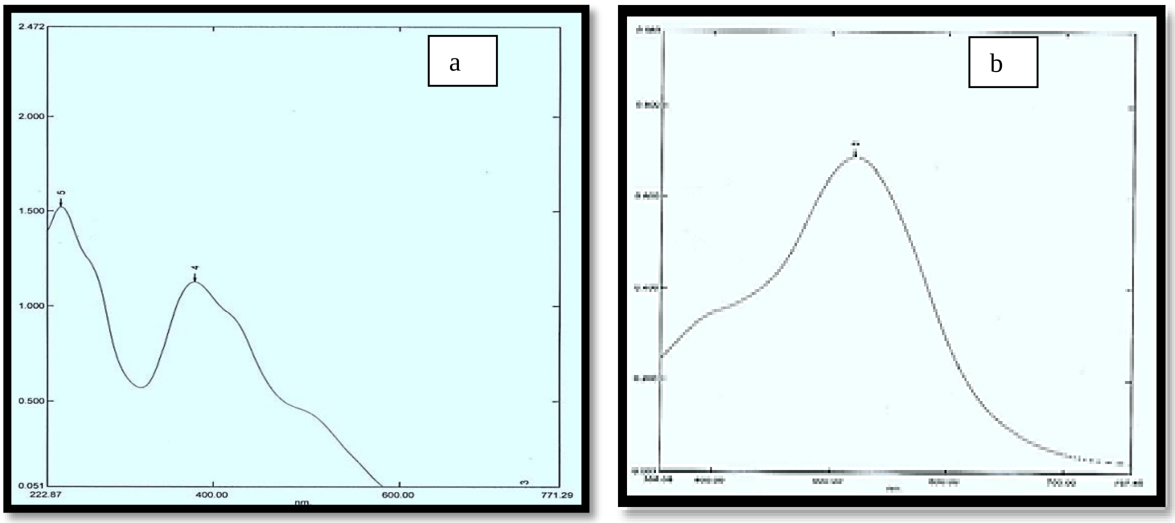



**Fig4-A – Ligand , B:Electronic spectru of azo-schiff base-Copper (II)complex**

**Table 3 : Electronic spectra of the ligand and its complexes**

| Compound | λ(nm) | ύ ($cm^{-1}$) | Transition | Geometry | Hybridization |
|---|---|---|---|---|---|
| L= $C_{19}H_{19}N_9$ | 204<br>327<br>380 | 49019<br>42194<br>26315 | $\pi \rightarrow \pi^*$<br>$\pi \rightarrow \pi^*$<br>$n \rightarrow \pi^*$ | ....... | ....... |
| [Co $(L)_2$ ]$Cl_2$ | 1092<br>510<br>403 | 9157<br>19607<br>24813 | $^4T_1g_{(F)} \rightarrow {}^4T_2g_{(F)} = v_1$ $^4T_1g_{(F)} \rightarrow {}^4A_2g_{(F)}\ v_2$<br>$^4T_1g_{(F)} \rightarrow {}^4T_1g_{(p)} = v_3$ | Octahedral | $sp^3d^2$ |
| [Ni $(L)_2$ ]$Cl_2.H_2O$ | 1092<br>733<br>529<br>392 | 9157<br>13642<br>18903<br>25510 | $^3A_2g_{(F)} \rightarrow {}^3T_2g_{(F)}\ v_1$<br>$^3A_2g_{(F)} \rightarrow {}^3T_1g_{(F)}\ v_2$<br>$^3A_2g_{(F)} \rightarrow {}^3T_1g_{(P)}\ v_3$<br>M.LCT | Octahedral | $sp^3d^2$ |
| [Cu $(L)_2$ ]$Cl_2$ | 520 | 19230 | $^2B_1g \rightarrow {}^2Eg$ | Octahedral | $sp^3d^2$ |
| [Zn $(L)_2$ ]$Cl_2$ | 507 | 19723 | M→L,CT | Octahedral | $sp^3d^2$ |
| [Cd $(L)_2$ ]$Cl_2$ | 547 | 18281 | M→L,CT | Octahedral | $sp^3d^2$ |
| [Hg $(L)_2$ ]$Cl_2$ | 549 | 18314 | M→L,CT | Octahedral | $sp^3d^2$ |

| | | | | | |
|---|---|---|---|---|---|
| [Au LCl] $Cl_2$ | 552 | 18115 | $^1A_1g \rightarrow {}^1B_1g$ | square planar | $dsp^2$ |

**Magnetic measurements:**

The Co(II) complex has a magnetic moment of 4. 1 B.M , which is in agreement with the reported value for octahedral Co(II) complexes(26). .The Ni(II) complex shows a magnetic moment value of 2.85 within the range of (2.9,3.3) B.M (27) suggesting an octahedral environment. The Cu(II) complex shows a magnetic moment value of 1.74 B.M, and consistent with a distorted octahedral geometry (28) .The Zn(II) ,Cd(II),Hg(II),Au(III) are diamagnetic and according to the empirical formulae of complexes, an octahedral geometry is proposed(28) except the Au(III) complex suggest a square planar geometry (29) . Based on the above results, we can deduce the probable structures of the complexes as shown in fig.5.

**Conductivity measurement :**

Molar conductance (Λm) measurements of the metal complexes table(4) carried out using DMSO as solvent at the concentration of $10^{-3}$ M in room temperature. All chelate complexes prepared in this work showed conductivity values ranged between (71.7.80.2-) $s.mol^{-1}.cm^2$ that electrolyte and conductive species( 28) indicating the electrolytic nature (1:2) electrolyte**.** we can deduce the probable structures of the complexes as shown in fig.5.

**Table4:magnetic susceptibility and molar Conductivity data of complexes**

| Compounds | $\mu_{eff}$ (B.M) | Conductivity $S.mol^{-1}. Cm^2$ |
|---|---|---|
| [Co (L) $_2$]$Cl_2$ | 4.1 | 72.1 |
| [Ni (L)] $Cl_2.H_2O$ | 2.85 | 79.7 |
| [Cu (L) $_2$] $Cl_2$ | 1.74 | 80.2 |
| [Zn (L) $_2$] $Cl_2$ | Dia | 75.3 |
| [Cd (L) $_2$] $Cl_2$ | Dia | 74.2 |
| [Hg (L) $_2$]$Cl_2$ | Dia | 80.2 |
| [Au LCl]$Cl_2$ | Dia | 71.7 |

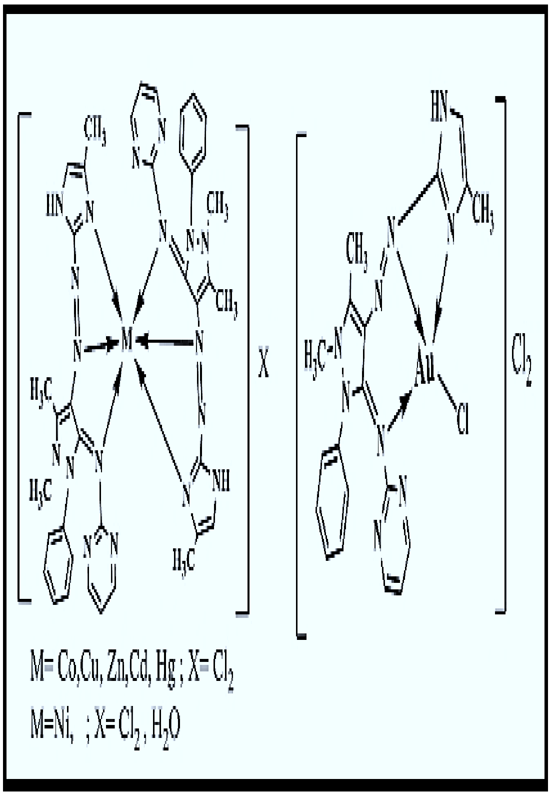



**Fig.5. The proposed structural formula of the complexes**

**Antimicrobial Activity.**

The antimicrobial activity of the investigated compounds were tested in vitro against the bacteria: *Escherichia coli*, *staphylococcus* aureus and fungi**:** A.flevus, *and A. neger* The values of minimum inhibitory concentration (MIC) for the investigated compounds are listed in Fig.6. From this table and growth areas resulting can show it in fig.(7,8) it can be seen that values of MIC indicate that most of the complexes have antimicrobial activity is higher. This increased activity of the metal chelates can be more broadly explained by the overlap between the ligand orbital and sharing of the partial positive charge for the ion of metal with donor groups. Moreover, this increases the non-concentration of π-electron over the entire ring of chelate and reinforces the penetration of the complex into lipid membranes and closing of the metal binding locations in the microorganisms enzymes. Also, the Au(III) complex impede the cell's respiration process and thus prevent the proteins synthesis, which restrains further the organism growth (30-32).

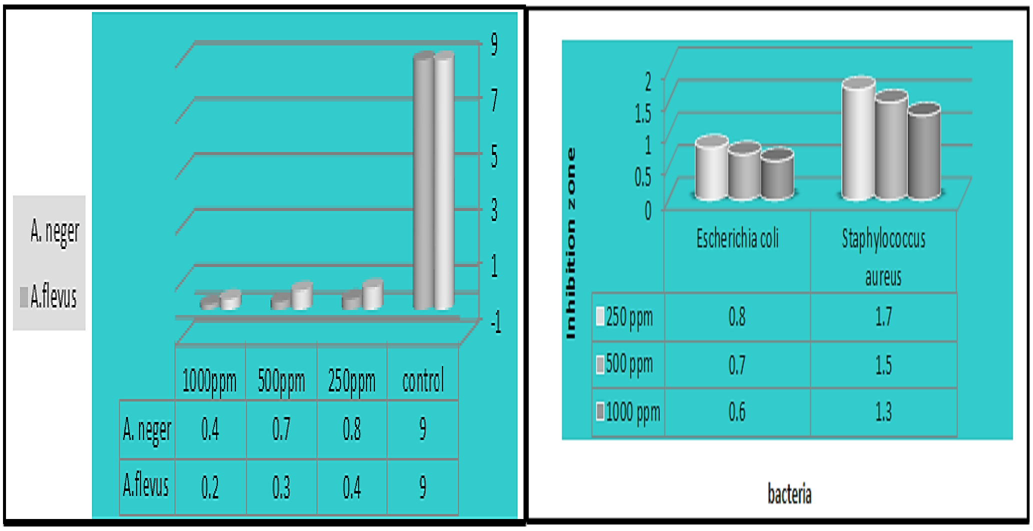



**Fig6:Antimicrobial antifungal and bacteria effect activity for comp**

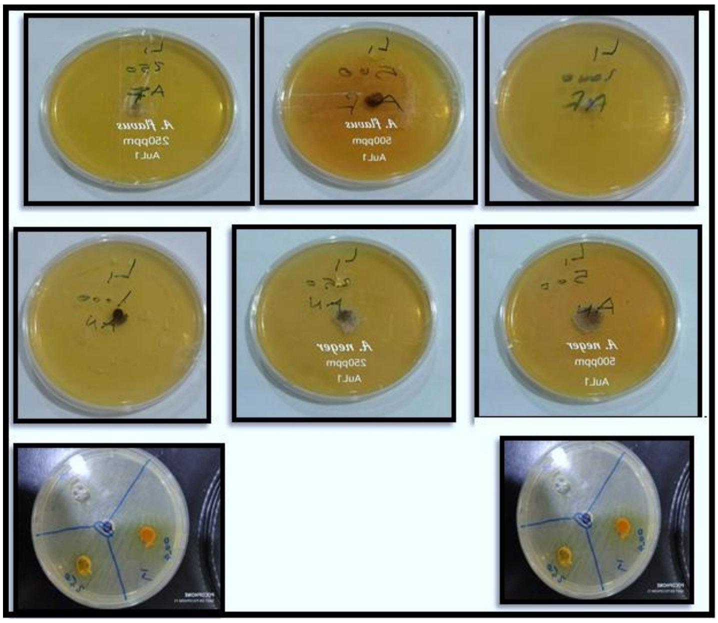


**Fig.7:Growth areas resulting in biological effect (Antimicrobial antifungal ,bacteria)- gold complex-different concentrations**

## Conclusion

In this paper we have explored the synthesis and coordination chemistry of some monomeric complexes obtained from the reaction of tridentate ligand with some metal ions as shown in figure 5.The complexes were determined through physico –chemical and spectroscopic methods. Complexes study via molar ratio has ratio of (M:L) as (1:2) except the Au(III) complex was (1:1) (metal: ligand ) ratio. The biological screening effect of the Au(II)complex are tested against several organisms , *staphylococcus aureus, Esherichia coli, Klebsilla, Aspegills niger, Aspegills flavus are reported* . the results show the highest inhibitory effect for complex more than their ligand , it may be because of ion effects on bacteria.

## References

1. Kribaa, L., Atia, S., Boudehane, A., Bader, R. M. M., & Gherraf, N. (2019). Synthesis, Characterization and Antioxidant Activity Studies of New Azo-Compounds. Journal of Biochemical Technology, 10(1), 85.

2. Mahdi, M. A., Jasim, L. S., & Mohamed, M. H. (2021). Synthesis, Spectral and Biological Studies of Co (III), Ni (II), and Cu (II) Complexes with New Heterocyclic Ligand Derived from Azo dye. Systematic Reviews in Pharmacy, 12(1), 426-434.\
3. Ali, Y., Muhamad Bunnori, N., Susanti, D., Muhammad Alhassan, A., & Abd Hamid, S. (2018). Synthesis, in-vitro and in silico studies of azo-based calix [4] arenes as antibacterial agent and neuraminidase inhibitor: a new look into an old scaffold. Frontiers in chemistry, 6, 210.
4. Sahoo, J., & Paidesetty, S. K. (2018). Biological investigation of novel metal complexes of 2-amino-4-substituted phenylthiazole Schiff bases. Journal of Taibah University medical sciences, 13(2), 142-155.
5. Al-Adilee, K. J., & Dakheel, H. K. (2018). Synthesis, Spectral and Biological Studies of Ni (II), Pd (II), and Pt (IV) Complexes with New Heterocyclic ligand Derived from Azo-Schiff Bases Dye. Eurasian Journal of Analytical Chemistry, 13(5).
6. Tahir, T., Ashfaq, M., Saleem, M., Rafiq, M., Shahzad, M. I., Kotwica-Mojzych, K., & Mojzych, M. (2021). Pyridine Scaffolds, Phenols and Derivatives of Azo Moiety: Current Therapeutic Perspectives. Molecules, 26(16), 4872.
7. Lamia, S. A., Rawa'a ,A. M., Rehab, K. R.,(2021). Applications of biological of Azo-Schiff base ligand and its metal complexes and: A review. MJPS,8(1),
8. Chaudhary, N. K., Guragain, B., Chaudhary, S. K., & Mishra, P. (2021). Schiff base metal complex as a potential therapeutic drug in medical science: A critical review. BIBECHANA, 18(1), 214-230.
9. Nagham Mahmood Aljamali , S Jawad., (2022). Preparation, Spectral Characterization, Thermal Study, and Antifungal Assay of (Formazane - Mefenamic acid)- Derivatives., Egyptian Journal of Chemistry , 411, Volume 65, Issue 2 , February 2022 , *DOI:* 10.21608/EJCHEM.2021.88727.4266
10. ABDELMADJID, A., Haffar, D., Benghanem, F., Ghedjati, S., Toukal, L., Dorcet, V., & Bourzami, R. (2021). Synthesis, Crystal structure, Electrochemical, Theoretical Studies and Antioxidant Activities of New Schiff Base. Journal of Molecular Structure, 1227, 129368.
11. Zhang, Y., & Chu, W. (2021). Cooperation of multi-walled carbon nanotubes and cobalt doped TiO2 to activate peroxymonosulfate for antipyrine photocatalytic degradation. Separation and Purification Technology, 119996.
12. Nagham Mahmood Aljamali.,Synthesis of Antifungal Chemical Compounds from Fluconazole with (Pharma-Chemical) Studying, Research journal of Pharmaceutical, biological and chemical sciences, 2017, 8 (3), 564 -573.
13. Wadher, S. J., Puranik, M. P., Karande, N. A., & Yeole, P. G. (2009). Synthesis and biological evaluation of Schiff base of dapsone and their derivative as antimicrobial agents. International Journal of PharmTech Research, 1(1), 22-33.
14. Miloud, M. M., El-ajaily, M. M., Al-noor, T. H., & Al-barki, N. S. (2020). Antifungal activity of some mixed ligand complexes incorporating Schiff bases. J Bacteriol Mycol, 7(1), 1122
15. Al-Adilee, K. J. (2015). Preparation and Characterization of Some Transition Metal Complexes with NovelAzo-Schiff base Ligand Derived from 2 (E)-(1H-benzo [d] imidazole-2-yly diazenyl)-5-((E)-benzylideneimino) phenol (BIADPI). RESEARCH JOURNAL OF PHARMACEUTICAL BIOLOGICAL AND CHEMICAL SCIENCES, 6(5), 1297-1308.
16. Krishnankutty, K., Ummathur, M. B., & Sayudevi, P. (2009). Schiff bases of 3-(2-thiazolylazo)-2, 4-pentanedione with phenylenediwnines and their metal complexes. Journal of the Indian Chemical Society, 86(4), 325-330.
17. Gülsah C., A. &Tahir T., (2017). Synthesis ,Characterization and Antimicrobial Studies of Some Transition Metal Complexes with azo Ligand Derived from 4-Amino Antipyrine. Res. Chem. Intermed.,DOI 10.1007.
18. Nagham Mahmood Aljamali. 2016. "Synthesis and Biological Study of Hetero (Atoms and Cycles) Compounds", Der Pharma Chemica, 8,6, 40-48.
19. Anupama - B. & GyanaKumara C. ; (2011). synthesis, characterization ,DNA Binding and Autimicrobial Activity of u-amno Autipypyrineeschffroase metal complexes, Research J. of pharm , Biological and chemical . Scienees, 2(4), 140- 146.
20. Al-Salami, B. K., Gata, R. A., & Asker, K. A. (2017). Synthesis spectral, thermal stability and bacterial activity of schiff base

derived from selective amino acid and their complexes. Pelagia Research Libray, 8(3), 4-12.
21. Dakore, S. D., Kamble, V., & Bisal, P. (2017). Synthesis and characterization of biologically active N2O2 type of Novel Schiff base metal complexes derived from 4-aminoanty pyrine using TEA. J. Chem Studies , 5(1), 110-113.
22. Nassar, M. Y., Ahmed, I. S., Dessouki, H. A., & Ali, S. S. (2018). Synthesis and characterization of some Schiff base complexes derived from 2, 5-dihydroxy acetophenone with transition metal ions and their biological activity. Journal of Basic and Environmental Sciences, 5, 60-71.
23. Al Zoubi, W., Al-Hamdani, A. A. S., Ahmed, S. D., & Ko, Y. G. (2018). A new azo-Schiff base: Synthesis, characterization, biological activity and theoretical studies of its complexes. Applied Organometallic Chemistry, 32(1), e3895.
24. Mahmoud, W. A., Musa, A., & Obaid, N. H. (2017). Preparation and Characterization of New Azo Ligand N-[(1-(4-(4, 5-Dimethyl-1h-Imidazol-2-Yl) Diazenyl) Phenyl-3-(Trifluoromethyl)] Aniline with Some Metal Complexes Ions. Acta Chimica Pharmaceutica Indica, 7(1), 1-12.
25. Rajaa Abdul Ameer Ghafil, Nour A Alrazzakb, Nagham Mahmood Aljamali. Synthesis of Triazole Derivatives via Multi Components Reaction and Studying of (Organic Characterization, Chromatographic Behavior, Chem-Physical Properties)., Egyptian Journal of Chemistry. Vol. 63, No. 11, pp. 4163 - 4174 (2020). DOI: 10.21608/EJCHEM.2020.23541.2399 .
26. Raut, K. T., & Shirote, P. J. (2012). Synthesis and characterization of novel amide derivatives of nitro imidazole. Der Pharma Chemica, 4(4), 1435-1439.
27. Al-Adilee, K. J., & Dakheel, H. K. (2018). Synthesis, Spectral and Biological Studies of Ni (II), Pd (II), and Pt (IV) Complexes with New Heterocyclic ligand Derived from Azo-Schiff Bases Dye. Eurasian Journal of Analytical Chemistry, 13,5.
28. Al-Adilee, K. J., Abedalrazaq, K. A., & Al-Hamdiny, Z. M. (2013). Synthesis and spectroscopic properties of some transition metal complexes with new azo-dyes derived from thiazole and imidazole. Asian Journal of Chemistry, 25(18), 10475.
29. Mahmoud, W. A., Musa, A., & Obaid, N. H. (2017). Preparation and Characterization of New Azo Ligand N-[(1-(4-(4, 5-Dimethyl-1h-Imidazol-2-Yl) Diazenyl) Phenyl-3-(Trifluoromethyl)] Aniline with Some Metal Complexes Ions. Acta Chimica Pharmaceutica Indica, 7(1), 1-12.
30. Chaudhary, N. K., & Mishra, P. (2017). Metal complexes of a novel Schiff base based on penicillin: characterization, molecular modeling, and antibacterial activity study. Bioinorganic chemistry and applications, 2017.
31. Mohammed, L. A., Mehdi, R. T., & Ali, A. A. M. (2018). Synthesis and Biological Screening of the Gold Complex as Anticancer and Some Transition Metal Complexes with New Heterocyclic Ligand Derived from 4-Amino Antipyrine. Nano Biomed. Eng, 10(3), 199-212.
32. Nagham Mahmood Aljamali , Nemah Sahib Muhammed. . Chemo - Spectral and Biological Studying of New Ligands ., Research Journal of Pharmaceutical, Biological and Chemical Sciences ., May – June, 2017 , RJPBCS ,8,(3) ,Page No. 674
33. Morad, F. M., Elajaily, M. M., & Ben Gweirif, S. (2007). Preparation, physical characterization and antibacterial activity of Ni (II) Schiff base complex. Journal of Science and Its Applications, 1, 72-78.
34. Rabab Mahdi Ubaid Mahmood, Rajaa Abdul Ameer Ghafil., Synthesis and Characterization some Imidazolidine Derivatives and Study the Biological Activity ,Annals of R.S.C.B., ISSN: 1583-6258, Vol. 25, Issue 3, 2021,Pages. 569 – 584