Preparation, Characterization and Study Biological Activity of some New Metal Chelate Complexes Derived from Ligand Azo–Schiff for HetrocyclicThiazole

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ABSTRACT

This Work included the preparation of the ligand 4-[{(z)-4-florobenzylidene)amino}-2-(thiazol-2-yldiazenyl)pheno]] from the preparation of the Schiff base by reacting para-aminophenol with 4-Flurobenzaldehyde to obtain schiff base as a first step. After that, the diazonium salt resulting from 2-amino thiazolewas coupling with Schiff base as a second step. Above is a component that pairs in an alcoholic base circumference to obtain the azo-Schiff derivative. Also, nine chelating complexes were prepared for the ions of each (Mn(II), Fe (II), Co (II), Ni (II), Cu (II), Zn (II), Pd (II), Cd (II), Hg (II). The results show that the mole ratio (metal - ligand) is (1: 2) and for all the complexes under study except for the palladium complex, whose molar ratio is (1:1). The ligand and its complexes were characterized by (C.H.N), molar conductance, magnetic susceptibility, FTIR, UV-Vis, HNMR, and mass spectroscopy. Based on the obtained results, an octahedral geometrywereproposed for all chelate complexes except for the palladium complex, which has square planar geometry around central metal ion. The research also included a study of the biological effect of this ligand and its metal complexes on the growth of two types of bacteria by using the solvent DMSO and the technique of spreading on the media of the blemishes as anti-bacterial Staphylococcus represented by bacteria positive for Gram stain (Gram positive bacteria) and bacteria Escherichia coli represented by bacteria negative for (Gram negative) Bacteria cause many common diseases.Where it was found that this ligand and its metal complexes have different biological efficacy as antagonists in inhibiting the growth of bacteria. The study has shown that the ligand and the prepared metal complexes have biological activities and when compared with the drugs Fluconazole and Cephalexin it is revealed that they can be used as medicines in the field of medicine.

KEYWORDS

Azo-Schiff, HetrocyclicThaizole, Antibacterial Activity.

Introduction

Azo - Schiff base complexes contain both azo and azomethine groups. The compositions of azo dyes differ according to each of the nitrogenous and paired components [1], Azo compounds are distinguished by their bright and clear colors. Their molecular structures and compensating groups differ on the aromatic rings, including yellow, red, orange, blue, and green [2]. They are used as indicators in analytical chemistry [3]. As it was characterized by strong sensitivity to acids, bases, and colors, it was clearly distinguished, and it also played an important role in the field of green chemistry [4]. As for the biological effect, it was used as antibacterial and fungal agents. It entered into the manufacture of medicines and medicinal drugs as anticancer [5,6], either in the field of Harmonic chemistry has played a major role in the formation of stable canine complexes [7,8]. This type of organic compounds was discovered by the scientist Chris (Greiss) and he called it this name in 1860, and they are the compounds containing in their chemical structures the azo group (N = N) (Azo group), linked on either side by two similar or different groups of aliphatic or Aromatically homogeneous or heterogeneous, the azo benzene compound is the simplest of these compounds. The groups associated with the bridging azo group are often arranged spatially, taking the position of the trans isomer at an angle of 120 ° [9]. Schiff base are characterized by many advantages, including relatively high stability. This stability is attributed to the aromatic characteristic of aliphatic basic [10]. Compounds containing Arielian compensators are distinguished by their high stability. They are often solid and have relative thermal stability. They are colored and most of the time their solubility in organic solvents depends on the nature of the groups associated with them. The reason for this is due to the increased stability by resonance [11] of the linked groups on both ends of a group. Azomethene. As for the liposomes derived from aliphatic compounds, they were distinguished by their liquid properties. It should be noted that the bases of lipids of ammonia are unstable interacting with each other to form different polymers [12]. Studies have shown that many of these rules are used as lycandes for the presence of the azo-methane group (C = N-) that behaves as a group of donor and receiver of electrons (accepter-Doner), in order to give them part of the non-ionic electron double charge of the nitrogen atom of the group referred to as the sama (σ -Donor)) Which leads to an increase in the electronic density on the atom or the metallic ion, which

in turn returns a part of this charge to the orbital (π^*) of the double insulin, so these ligands can also be called ($\pi^$ acid ligand) and this is the reason for the susceptibility of the likandes that include the azo group. Methane helps to form stable complexes at low oxidative stress. Thanks to its preparation, the German scientist Hugo Schiff was able to prepare the first organic compound in 1864, and this name was given to the organic compounds containing in their structures on the group Azomethene -CH = N-) as an effective group. And by joining together a group of carbonyl compounds (ketones or dihydrates) with elementary amines in contrasting solvents with the presence of a factor to withdraw water molecules resulting from the said reaction [13]. The succession of research and the proliferation of studies on the preparation of azo-isomethene reflects its importance. The reason for the effectiveness of azoisomethene is attributed to the nitrogen atom of both groups of isomethene, and these two groups helped to enter into harmony with the different metal ions.Based on that, the study of these organic compounds has gained a wide scope in many fields, and we will cite here a set of examples to illustrate some of the effects of this type of compounds. Where it acquired a wide area in terms of application, in the field of biology, the azo-base complexes were used as anti-carcinogens, due to their ability to link in harmony with the nitrogen atoms in the DNA bases to form a chelated ring, as the cobalt, copper and zinc ion complexes showed a positive charge with lycand (E) - (4- Ethylphenyl) Diazinyl) -2 - ((E) - (4 -)) - 4- Methoxyphenyl (Amino) methyl) Phenol inhibition in killing cancer cells towards three cancer lines, which is the MCF line. 7, prostate PC3, lung H2126 [14]. In the field of liquid crystals, the azobase compounds entered the pigments industry. Five dyes were prepared from the condensation of the 4pentyloxybenzyldehyde compound with five derivatives of the compound (4- aminoazobenzene). These compounds possess a liquid crystalline characteristic [15], and some of them have also been used in the industrial field as antioxidant materials [16] (Antioxidant), and it has been found that some of them have an important effect in the process of inhibition of corrosion [17] (Corrosion Inhibitors). In the field of analytical chemistry, it has been used in the manufacture of selective electrodes for many metal ions, an example of which is lycand 2,2 '- ((1 Z, 1'Z) - (2,2' -((Z) -diazene-1,2-diyl). bis (2,1-phenylene)) bis (azan-1-yl-1-ylidene) bis (methan-1-yl-1ylidene) di phenol[18]. This type of compound has been used for mineral analysis, instrumentation applications related to communications, optical computation [19], storage [20], and information processing [21]. Due to the importance of azo-containing Schiff base compounds and continuing interest in the syntheses of azo-based compounds, here the synthesis and characterization of a series of azo compounds bound together with the lip base are synthesized and characterized and these aggregates are collectively termed azo-Schiff [22,23].

Experimental

Materials and Measurements

All reagent and soluant were obtained from commercial sources were of highest purity. FTIRspectra of the prepared ligand and its solid-state complexes were recorded with a Shimadzu FTIR 8400S spectrophotometer, using KBr granules in the range 4000-400 cm⁻¹. Electronic spectra in (DMSO) were recorded on using a Shimadzu UV-Vis. (240) spectrophotometer. ¹HNMR spectra were performed in a BRUKER 500MHz spectrophotometer and with DMSO-d6 as the solvent and TMS as the internal standard. Elemental analyses were carried out by means of micro analytical unit of (ElementarAnalysen System (GMBH) elemental analyzer. Magnetic susceptibility measurement were carried out on abalance magnetic (Balance Magnetic Susceptibility Model(M.S.B Auto).

Preparation of Azo-Azomethine ligands (FBATYP): The Azo-Schiff ligand (FBATYP) was prepared by two steps.

Preparation of Schiff Base

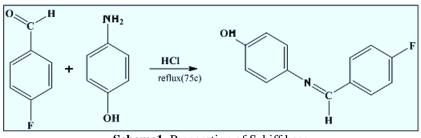
Preparation of the compound 4 - ((4-fluorobenzylidene) amino) phenol was prepare by using the condensation method from the reaction of 4 – flouro benzaldehyde (0.01 mol) (2ml) and (0.01 mol, 1.24gm) of para-amino phenol in equimolar (1:1) mole ratio in absolute ethanol followed by adding few drops of (HCl) acid to the reaction mixture and refluxed for (45h). The product was recrystallized from hot ethanol and dried.m.p (78-81°C). Scheme 1.

Preparation of Schiff base by(Microwave)[24]

A quantity of para-aminophenol (0.01 mol) was placed in a jar and ground well, then (2ml) of parafluorobenzyldehyde was added and mixed well with a very small amount of polar solvent (absolute ethanol) and then

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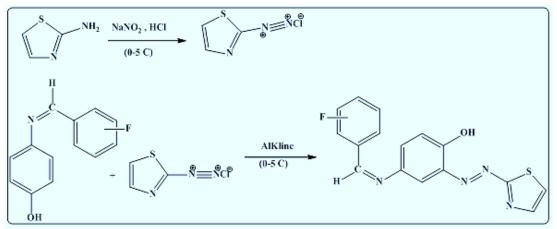
adding a few drops of glacial acetic acid. Then we mix the materials is placed inside the microwave apparatus and after a few periods (7-15 minutes) the reaction was completed and the product was collected and it was recrystallized with hot ethanol, and then the product was filtered and left for a period to dry and be collected. And then measuring the physical properties of the product from the melting point, product quantity, and color. When compared to the product of the classical condensation method for preparing schiff bases, it was found that the product ratio is high compared to the first method.



Scheme1. Preparation of Schiff base

Preparation of Azo-Schiff Ligand

The hetrocyclic azo Schiff ligand was prepared by coupling reaction of diazonium salt of 2-amino thiazole by dissolving (0.01 mol, 1.001g) in (3ml) of concentrated hydrochloric acid and (30ml) of distilled water at temperature (0-5 °C), followed by adding (10ml),(0.01mol, 0,69gm) from sodium nitrite.diazonium salt was added dropwise to Schiff base (0.01mol, 0.7gm) dissolved in (150ml) of ethanol and (10ml) of sodium hydroxide solution (10%) with continuous sitirring with control of temperature not increasing. The mixture was allowed to stand over night, the greenish-brown precipitate was filtered and washed with distilled water several times to ensure that the solution was free of the sodium chloride salt resulting from the reaction. The solid was dried and recrystallized with hot absolute ethanol, and then its melting point was measured ($60-63\circ$ C).



Scheme2.Preparation of Azo Schiff ligand

Preparation of theChelate Complexes

Chelate complexes were prepared by dissolving the ligand (FBATYP) in 30 mL ethanol (0.002 mol) with (0.001 mol) the mineral salts of [MnCL₂.2H₂O, FeSO₄,CoCl₂.6H₂O, NiCl₂.6H₂O,CuCl₂.2H₂O,ZnCl₂, PdCL₂,CdCL₂.H₂O,HgCL₂]in molar ratio (1: 2) [metal: ligand], Except for the palladium complex PdCl₂, which has a molar ratio of (1: 1). These mineral salts were dissolved in 30 ml of distilled waterwith stirring for 30 minutes, a colored precipitate was formed at room temperature, and the resulting solids were then filtered and washed several times with distilled water. The chelate complexes were air-dried, recrystallized from ethanol, and dried in an oven at 50 $^{\circ}$ C for several hours.

ResultsandDiscussion

The ligand is greenish brown crystal which is soluble in common organic solvents. The reaction of the ligand with metal ions mentioned above gives vary in color depending of metal ion. All complexes are quiet air-stable,insoluble in water, but its soluble in solvents such as DMF, DMSO, acetone and methanol. The elemental analysis and metal contents data are show In all cases (1:2) (metal : ligand) except for the palladium complex it is (1:1) solid complexes are isolated correspond well with the general formula $[M(L)_2]$.H₂O where M= Mn(II), Fe (II),Co (II),Ni (II),Cu (II),Zn (II),Cd (II) and Hg (II) except Pd (II) was formula [ML].H₂O that is agreement with the stoichimertric ratio found using molar ratio methods. The conductivity values of the chelates complexes support no electrolytic nature of the metal complexes attempts suggest the structure of the isolated complexes come from full investigation using the following studied. Some physical and chemical properties for ligand and its complexes are listed in table 1.

No	Compound	Colour	M.PC	Yield	Formula	Found (calc.) %			
				%		С	Н	Ν	М
1	L=(FBATYP)	Black	60-63	77	C ₁₆ H ₁₁ N ₄ SOF	58.61	3.21	17.01	
						(58.89)	(3.37)	(17.17)	
2	[MnL ₂].H ₂ O	Red	310 Dec.	69	$[(C_{23}H_{16}N_4O_2)_2Mn]$	53.04	2.91	15.39	7.38
						(53.11)	(3.04)	(15.49)	(7.59)
3	[FeL ₂].H ₂ O	Brown	310 Dec.	74	$[(C_{23}H_{16}N_4O_2)_2Fe]$	52.98	2.88	15.29	7.43
						(52.03)	(3.03)	(15.47)	(7.71)
4	[CoL ₂].H ₂ O	Brown	310 Dec.	83	$[(C_{23}H_{16}N_4O_2)_2Co]$	52.68	2.98	15.22	8.04
						(52.82)	(3.02)	(15.40)	(8.10)
5	[NiL ₂].H ₂ O	Black	310 Dec.	79	[(C ₂₃ H ₁₆ N ₄ O ₂) ₂ Ni]	52.71	2.92	15.24	7.97
						(52.84)	(3.02)	(15.41)	(8.07)
6	[CuL ₂].H ₂ O	Dark brown	156	87	$[(C_{23}H_{16}N_4O_2)_2Cu]$	52.31	2.85	15.27	8.49
						(52.49)	(3.00)	(15.31)	(8.68)
7	[ZnL ₂].H ₂ O	Dark brown	138-140	66	$[(C_{23}H_{16}N_4O_2)_2Zn]$	52.22	2.79	15.09	8.63
						(52.35)	(2.99)	(15.27)	(8.91)
8	[Pd (L)Cl].H ₂ O	Dark brown	310 Dec.	65	$[(C_{23}H_{16}N_4O_2)PdCl]$	39.45	2.00	11.33	21.32
						(39.59)	(2.06)	(11.54)	(21.94)
9	[CdL ₂].H ₂ O	Reddish brown	310 Dec.	76	$[(C_{23}H_{16}N_4O_2)_2Cd]$	49.05	2.59	14.19	14.10
						(49.20)	(2.81)	(14.35)	(14.40)
10	[HgL ₂].H ₂ O	green	310 Dec.	64	$[(C_{23}H_{16}N_4O_2)_2Hg]$	44.09	2.21	12.67	22.96
						(44.20)	(2.53)	(12.89)	(23.09)

Table1.Physical properties and analysis of ligand and its complexes

FTIRSpectra

The most important IR absorption bands corresponding to the ligand and its complexes are presented in table 2. The band in the region 3498cm^{-1} due to stretching mode of (O-H) in the spectrum of ligand. This band has disappeard and new ones have appeard in the complexes spectra wiche due to (H₂O) in the range (3400 - 3437) cm⁻¹ in addition to the bands that appeared in the region (800 - 900) cm⁻¹[25]. The observed band in the range (1400-1415) cm⁻¹ is due to the v(N=N) group of the free ligand [26]. These bands are shifted to a higher or lower frequency during the complexes formation indicating their participation in coordination with the metal ions. The sharp band in the range (1600-1620) cm⁻¹ is due to the frequency (C=N) Schiff base groupand ($1504 - 1506 \text{ cm}^{-1}$) due to the (C=N) azomithine group in thiazole ring. Some bands in the range of 470- 480 and 530-567 cm–1 are not present in the free ligand spectrum assigned to v (M–O) and v (M–N) vibrations [27]. appearance In all complexes.

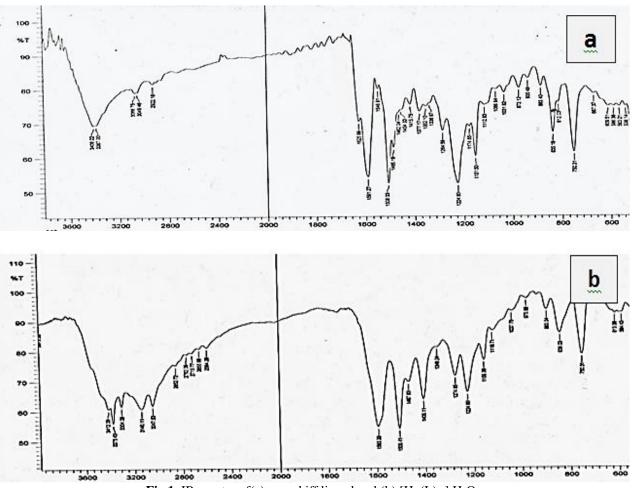


Fig.1. IR spectra of(a)azo-schiff ligand and (b) $[Hg(L)_2]$.H₂O

Compound	v(OH)	$\nu(N=N)$	υ(C=N)	v(C=N)		
_	H ₂ O	Azo	Schiff	Thiazole	υ(M-O)	υ(M-N)
C ₁₆ H ₁₁ N ₄ SOF	3408	1415	1625	1591		
	phenol					
$[Mn(L)_2].H_2O$	3417	1404	1624	1508	516	442
$[Fe(L)_2]. H_2O$	3400	1427	1598	1504	531	447
$[Co(L)_2]. H_2O$	3450	1404	1602	1502	524	447
$[Ni(L)_2], H_2O$	3400	1406	1598	1504	514	449
$[Cu(L)_2]$ H ₂ O	3400	1404	1593	1504	513	418
$[Zn(L)_2] H_2O$	3414	1417	1597	1504	501	414
[Pd(L)Cl]H2O	3437	1404	1598	1504	551	441
$[Cd(L)_2] H_2O$	3400	1404	1598	1506	516	453
$[Hg(L)_2] H_2O$	3412	1406	1593	1506	514	449

Table 2. Some IR frequencies in (cm⁻¹) of the ligand and its metal complexes

Electronic Absorption Spectra

The electronic absorption spectrum of free ligand shows bands at (255 nm) and (415 nm) which alue to $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ respectively [28]. the electronic spectra of metal complexes shows as fallow. The electronic spectra of the ligand complexes show two intense transition peaks < 420 nm which are assigned to ligand-centered transitions, along with an intense peak at longer wavelength (419-585) nm that can be referred to as the combination of MLCT and ILCT [29] as shown in Table.3.

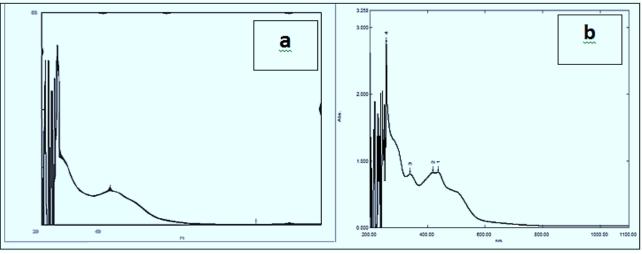
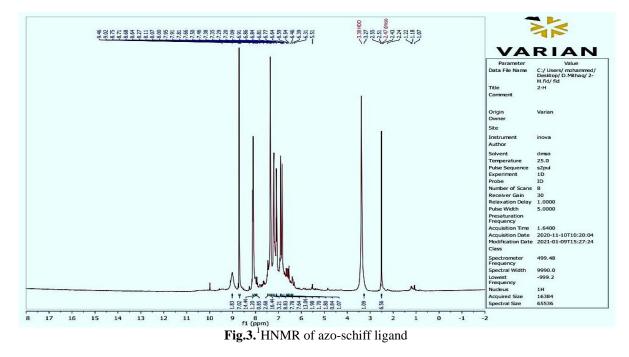


Fig.2.Uv-Vis spectra of : (a) the ligand (FBATYP) and (b)[Hg(L)₂].H₂O

¹H.NMR Spectra

¹H-NMR spectra of azo-azomethine ligand was performed in deuterated dimethyl sulfoxide solutions with tetramethylsilane as an internal standard. spectra showed a peak at 2.5 ppm which was due to DMSO solvent [30]. The spectrum showed the singlate singal at (9.46 ppm) was assigned to (OH) the proton of phenolic [31]. The multiplate range signals at (6.84-7.35ppm) were attributed to the oromatic protonsof the [32]. The signals at (8.11ppm and 8.71ppm) which were due to of ethylene group in thiazole ring. The singlate signal showed at (9.04ppm) was attributed to the proton of azomethine group (HC=N) [33].



Mass Spectroscopy

The fragmentation pattern of the azo-azomithine ligand in (Fig.4) which showed fragments elearly corresponding to the calculated formula $[C_{16}H_{11}N_4SOF]^+$. The mass spectrum showed fragment at m/z=326which correspond to the exact molecular weight of the synthesized azo Schiff base. The fragments wereappeared at (m/z=299.3) and(m/z=264.3) due to $[C_{16}H_{11}N_2OSF]^+$ and $[C_{16}H_{10}N_2S]^+$ [34].

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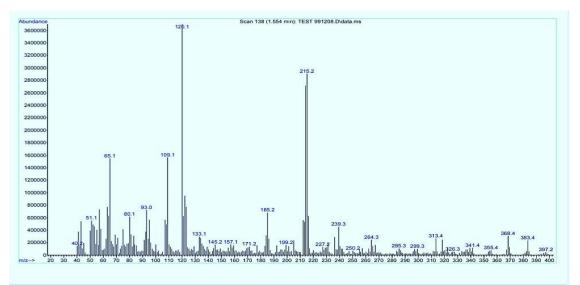
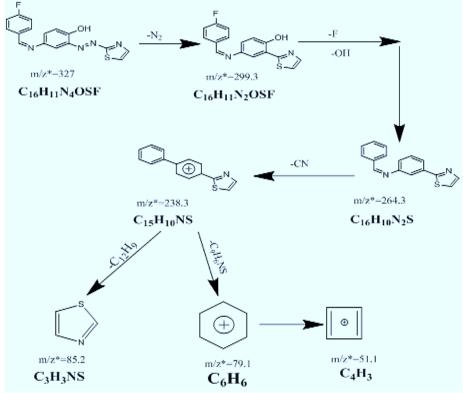


Fig. 4. Mass spectra of azo-schiff ligand

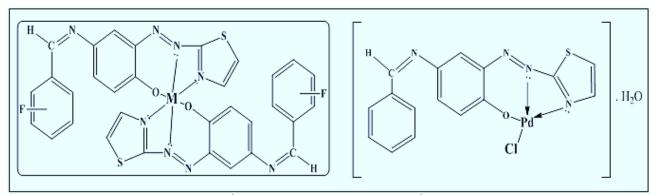


Scheme3.Suggested mass fractionation pathways for ligand(FBATYP)

Magnetic Properties and Molar Conductivity Measurements

Themagnetic moment values measured at room temperature. The magnetic moments value of Mn(II), Fe(II), Co(II), Ni(II), Cu(II) complexes (5.4 B.M), (4.3 B.M), (4.72 B.M), (2.92 B.M), (1.73 B.M). suggest a high-spin octahedral configuration [35,36]. The complexes of Zn(II), Pd(II), Cd(II) and Hg(II) its diamagnetic moments [37]. Molar conductance datd of the complexes were measured in the solvent DMSO (10^{-4} M) were found to be non-electrolytic nature[38]. According to these result the structural formulas of these ligand and its complexes may be proposed in (Fig.5)

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 M^{+2} =Mn,Fe,Co,Ni,Cu,Zn,Cd,Hg[Pd^{+2}] Fig.5.The proposed structural formula of prepared chelate complexes

Table3.Electronic sp	pectra, conductivity	and magnetic mome	ent of complexes

	Abs.	Transition	Cond.	μ_{eff}
Compounds	$nm(cm^{-1})$		$S.mol^{-1}.Cm^2$	(B.M)
Ligand=HL (FBATYP)	255	π→π *		
	415	n→π *		
	259	π→π *		
$[Mn(L)_2].H_2O$	420	ML CT	16.5	4.3
	265	π→π *		
$[Fe(L)_2].H_2O$	419	IL CT	5	4.72
	436	ML CT		
	265	$\pi \rightarrow \pi *$		
$[Co(L)_2].H_2O$	436	ML CT	13.6	2.92
	261	$\pi \rightarrow \pi *$		
$[Ni(L)_2].H_2O$	438	ML CT	15.9	1.73
	277	$\pi \rightarrow \pi *$		
$[Cu(L)_2].H_2O$	420	ML CT	10.8	Dia
	267	$\pi { ightarrow} \pi *$		
$[Zn(L)_2].H_2O$	437	ML CT	1.6	5.4
	271	$\pi \rightarrow \pi *$		
[Pd(L)Cl].H ₂ O	410	IL CT	2.4	Dia
	585	ML CT		
	289	π→π *		
$[Cd(L)_2].H_2O$	422	C.T	8.2	Dia
	257	π→π *		
$[Hg(L)_2].H_2O$	419	IL CT	12.4	
	437	ML CT		Dia

Pharmacological Evaluation[39]

Biological Studies: The organisms used in the present investigation included Gram-positive bacteria (*Staphylococcus aureus*) and Gram-negative bacteria (*Escherichia coli*). The results of the bactericidal screening of the synthesized compounds are recorded in Table 4. An influence of the central ion of the complexes in the antibacterial activity against the tested Gram-positive and Gram-negative organisms shows that the L has an enhanced activity compared to its complexes.

Different microbial species and strains have different degrees of susceptibility to different chemotherapeutic agents. Moreover, the susceptibility of a microorganism can change with time, even during therapy with a specific drug. Several tests have been used to indicate which chemotherapeutic agent is most likely to combat a specific

pathogen[40].

Principle Method Involved

Disk Diffusion Method: Probably the most widely used, although not necessarily the best, method of testing is the disk diffusion method also known as the Kirby-Bauer test. A Petri plate containing an agar medium is inoculated(seeded) amount of test organism. Next, filter paper disk impregnated with known concentrations of chemotherapeutic agents diffuse from the disk into the agar. The farther the agent diffuses from the disk, higher effective it is. If the chemotherapeutic agent is effective, a zone of inhibition forms around the disk after a standardized incubation[41].

Experimental (Microbes):All microorganisms were obtained. University of Kufa / College of Education for Girls - Life Sciences Laboratory.

Table4. Areas of inhibition resulting from the biological effect of the chelating complexes of ligand in concentration
(1000M) with the bacteria under study.

Compound	G(-)	G(+)
Bacteria	Escherichia coli	Sta.aureus
(FBATYP)	++	++
[Hg(FBATYP)]. H ₂ O	+++	+++
[Cd(FBATYP)]. H ₂ O	+++	++ +
[Zn(FBATYP)]. H ₂ O	+	++
[Co(FBATYP)]. H ₂ O	+	++
[Ni(FBATYP)]. H ₂ O	+	+
[Cu(FBATYP)]. H ₂ O	+	+
[Fe(FBATYP)]. H ₂ O	+	+
[Mn(FBATYP)]. H ₂ O	+	+
[Pd(FBATYP)]. H ₂ O	+	+
DMSO	_	_

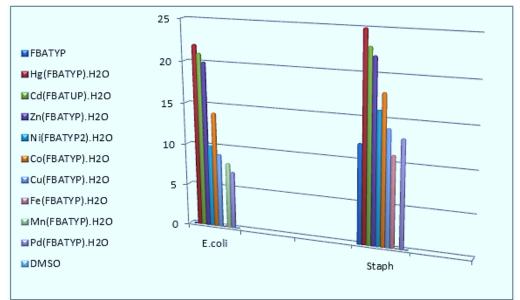


Fig.6. Biological efficacy and complexes of ligand with concentration (1000 M)

Conclusion

New compounds of Mn(ll),Fe(ll),Co(II), Ni(II), Cu(II), Zn(ll),Pd(ll), and Cd (II), Hg(ll) were synthesized with an Azo-Schiff base derived from the reaction of 2-aminothiazole with4-((4-florobenzylidene)amino)phenol. All complexes are insoluble in water but soluble in DMF and DMSO, acetone and methanol. The structure of the complexes based on mass UV and IR spectroscopy has been suggested, that the Azo-Schiff Ligand L base behaves as a tridentate ligand with metal ions through N,N, O atoms. The compound ligand and its mineral complexes were examined for their biological activity against Bacterial species, Gram-positive bacteria (Staphylococcus aureus), and Gram-negative bacteria (Escherichia coli). Where it became clear to us when studying the biological effects of these complexes that the two complexes of mercury and cadmium have a very high inhibitory activity compared to the rest of the complexes, and the reason for this is likely to the high toxicity of both mercury and cadmium, which leads to hindering the growth of the bacteria under study and eliminating them, while each of the complexes possesses Zinc, cobalt, nickel, and copper binary inhibitory activity is moderate and higher than that of binary iron, manganese and palladium complexes whose inhibitory efficacy against bacteria, microbes, and fungi is very weak because they do not have toxicity effectiveness.

References

- [1] DehnoKhalaji, A.; Mighani, H.; Kazemnejadi, M. (2017) Synthesis and characterization of Schiff-base polymer derived from 2, 5-dichloroaniline and 2-hydroxybenzaldehyde. *Iranian Chemical Communication*, 5: 237-241.
- [2] Birzan. R. A. C, Tecuceanu. L, Nica. V, & Enache. S; Rev. Chim (Bucharest), 6, 55, (2010).
- [3] Labd-Alredha R. AL-Rubaie, &R. Jameel Mhessn; E-Journal of Chemistry, 9(1), 465-470, (2012).
- [4] Nihad. I. T, Nashwan. O. T, & Marwa. N. El-Subeyhi; *International Journal of Organic chemistry*, 8,309-31,(2018).
- [5] Surbhi. V. U, Raksha V. Z, & Keyur D. B; An Scientific journal, 145, 1-15, (2020).
- [6] Jasmine. M. C, Tim. R. B, Alexander.D. H. K, Robert.P, David. M. P, & James. W. W; molecules, 25, 2383. (2020).
- [7] Widad. I. Y, Raheem T. M, & Hussein. K. A; Journal of Physics: Conf. Series 1294. (2019).
- [8] Radha. V. P, Mahalakshmi. D, Jonekirubavathy. S & Chitra. S; J. Serb. Chem. Soc, 85 (2), 215–225, (2020).
- [9] Hunger.K; "Industrial Dyes "; WILEY –VCH VerlagGmbH&Co.KGa A., p. 19, (2003).
- [10] El-Sherif, A. a, & Aljahdali, M. S; J. Coord. Chem, 66, 3423-3468. (2013).
- [11] Martin, K; Molecules, 22 (15-73), 1-15, (2017).
- [12] Zhang. D, Wu. Y, Huo. C, He. L, Lu. W, & Feng. Z; Emerging Materials Research, 1-6, (2019).
- [13] Constable. E. C; Metals and ligand reactivity. 308 (John Wiley and SonsInc., Publication, (2000).
- [14] Ispir. E, Ikiz. M, Inan. A, Sünbül.A. B, Tayhan. S. E, Bilgin. S,& Elmastaş. M; Journal of Molecular Structure, 1182, 63-71, (2019).
- [15] Abed Mosa. Z.S,& Al-Haijaj. A. J; Basrah Journal of Science, 33(2), P22-36, (2015).
- [16] Danish. S, Aamer. S, Fayaz. A. L, Pervaiz. A. C, Qamar. A, Mohamed. F. A, Ifzan Arshad. M, Mauricio. F. E, Mubashir. H, Hussain. R, Sung-Yum. S,&El-Seedi. H. R; *Molecules*, 24,1511. (2019).
- [17] Fouda.A. S, El-morsi. M. A, Gaber. M, & Fakeeh M; Int. J. Electrochem. Sci, 12, 8745-8760, (2017).
- [18] Ganjali. MR, Gholivand.M. B, Rahimi-Nasrabadi. M, Maddah. B, Salavati-Niasari. M,&Ahmadi. F;*American Scientific Publishers*, 4(4),356-363,(2006).
- [19] B. Kosar, C. Albayrak, Spectroscopic investigations and quantum chemical computational study of (E)-4methoxy-2-[(p-tolylimino)methyl]phenol, Spectrochim. *Acta, Part A* 78 (2011) 160-167.

- [20] F. Robert, A.D. Naik, F. Hidara, Engineering Solid-State Molecular Switches: N-Salicylidene NHeterocycle Derivatives, *Eur. J. Org. Chem.* 4 (2010) 621-637.
- [21] M.N. Chaur, D. Collado, Configurational and Constitutional Information Storage: Multiple Dynamics in Systems Based on Pyridyl and Acyl Hydrazones, J.M. Lehn, Chem. Eur. J. 17 (2011) 248-258.
- [22] Al-Noaimi M, Sunjuk M, El-khateeb M, Haddad SF, Haniyeh A, AlDamen M (2012) cis-trans Isomerism in mixed-ligand ruthenium (II) complexes containing bis (phosphine) and azoimine ligands. *Polyhedron*.42(1):66-73.
- [23] Bal, S. (2016) A Novel Azo-Schiff Base Ligand and Its Cobalt, Copper, Nickel Complexes: Synthesis, Characterization, Antimicrobial, Catalytic and Electrochemical Features. *Anadolu university journal of science and technology–Applied Sciences and Engineering*, 17(2): 315-326.
- [24] Suresh Patil, S. D. Jadhav, and U. P.Patil, "Natural Acid-Catalyzed Synthesis of Schiff Base under Solventfree Condition: As Agreen Approach", Archives of Applied Science Research, 4(2):1074-1078 (2012).
- [25] Suhad. K. A, Jassim. A. Al-Hilfi, Sawsan. K. Abbas, & Luma. M. Ahmed; *Indones. J. Chem*, 20(2), 404 412, (2020).
- [26] Rabab Mahdi Ubaid Mahmood., Synthesis, Spectral Investigation and Microbial Studying of Pyridine-Heterocyclic Compounds., *European Journal of Molecular & Clinical Medicine*, 2020, Volume 7, Issue 11, Pages 4444-4453.
- [27] Ibtihal. K.K, Fawzi. Y. W, & Ghusoon. J. A; J. Pharm. Sci. & Res. 11(1), 119-124, (2019).
- [28] Suhad. K. Abass, Jassim. A. A-Hilfi, Sawsan. K. Abbas, & luma M. Ahmed; *Indones. J. Chem*, 20(2),404-412,(2020).
- [29] Damian. C. O, Anthony. C. E, Boitumelo. M. S., Olujide. O., Eric Hosten; *Inorganicachimica Acta*, 450, 69-80, (2016).
- [30] Ommenya F. K, Nyawade. E. A, Andala. D. M, & Kinyua. J;*Hindawi Journal of Chemistry*, 8 pages, (2020).
- [31] Rasim. F. M,& Suheb. E. S; Oriental Journal OF Chemistry, 35, (4), 1360-1367, (2019).
- [32] Sadeek. S. A, El-Hamid. S.M. Abd, Rashid. N.G; Egypt. J. Chem. 63, 3, 939-951, (2020).
- [33] Khalid J. AL-Adilee, Hassein A. K. Kyhoiesh, J.Molecular Structure, 1137, (2017), 160-178.
- [34] Lee. K. L. K, & McCarthy. M. C; The journal of physical chemistry letters. (2019).
- [35] Sahar. S, Hoda. P, Naser. F, Mehran. D, & Fereshteh. M; Appl. Sci., 8 (85), 1-12, (2018).
- [36] N.S.Al-Barki, A.A.Maihub, M.M.Elajaily and T.H.Al-Noor; *AcademicJournal of Chemistry*, 1(3), p. 66-75, (2016).
- [37] S.A. Sadeek, A.W.H. EL-Shwiniy, W.A. Zordok, A.M. EL-Didamony, Spectrochim. Acta Part A 78 (2011) 854.
- [38] El-Ghamry. H. A, Gaber. M, & Farghaly. T. A; *Mini reviews in medicinal chemistry*, 19(13),1068-1079, (2019).
- [39] Rabab Mahdi Ubaid Mahmood., Synthesis, Characterization of Diazepine-Bicycles System and Study of their Bio-Behavior. *International Journal of Pharmaceutical Research*, 2021, Volume 13, Issue 1, Pages 4225-4233.
- [40] MM.Abd-elzaher, Shakdofa MME., Mousa HA. And Mostafa SA. Sop. Trans.On Appl. J. 1(1):42-52,(2014).
- [41] Swati, Manu Gupta, RomilaKarnawat, IK Sharma, PS Verma, Research Journal of Pharmaceutical, Biological and ChemicalSciences. Vol. 2, No. April – June, (2011),805-811.