

## Synthesis, spectroscopic and biological studies of some metal complexes with orthoamino hydrazo benzene

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Received 9, August, 2009

Accepted 30, May, 2010

### Abstract:

Ortho amino hydrazobenzene (L) has been prepared from the reaction of ortho amino phenyl thiol with phenyl hyrazan in mole ratio(1:1). It has been characterized by elemental analysis (C, H, N), IR, UV-Vis. The complexes of the bivalent ions (Co, Ni, Cu, Zn, Pd, Cd, Hg and Pb) and the trivalent (Cr) have been prepared and characterized too. The structural have been established by elemental analysis(C,H,N), IR , UV – Vis spectra , conductivity measurements , atomic absorption and magnetic susceptibility . The complexes showed characteristic behaviour of octahedral geometry around the metal ion and the( N,N) ligand coordinated in bidentate modeexcept with pd showed square planer.  $\alpha$  , $\kappa$ f ,  $\epsilon_{\max}$  for the complexes were estimated too .  $\beta$  for Co – complex was calculated . The study of biological activity of the ligand (L) and its complexes showed various activity toward Streptococcus pyogenes Staphylococcus aureus , Pseudomonas aeruginosa , E. coli and Candida albicans .

**Key words:** synthesis, hydrazobenzene, complexes, biological study, transition metals .

### Introduction:

The metal chelates with ligands are of biologically important and have been played an important role in the development of new coordination chemistry ; complexes containing chelating ligands , which show promising biological activity have been studied in this work , investigating the synthesis and characterization of many metal ions complexes that contain a range of ancillary ligands such as [1,2,3] macrocyclic ligands incorporating benzimidazol , Leucine – o – acetic acid, oxadiazole derivatives, [4,5,6,7,8] triazol derivatives, dioxadiazol and ditriazol , ortho amino phenyl thio derivatives , bis oxadiazol [9] derivatives. The chemistry of hydrazides has been intensively investigated in recent years, the reasons are manifold; First is the

coordinating ability of these compounds to chelate metal ions and [9-14] demonstrates tuberculostatic antibacterial and antifungal activities. Here we reported the synthesis of various metal ions complexes of orthoamino hydrazobenzene and their bactericidal and fungicidal activities.

### Materials and Methods:

–**Chemicals;** All reagents used were Analar or chemically pure grade by British Drug Houses (BDH), Merk and Fluka

- **Materials :** ortho amino phenylthiol ( $C_6H_7NS$ ), phenyl hyrazen ( $C_6H_8N_2$ ), Cobalt chloride hexahydrate ( $CoCl_2.6H_2O$ ), Nickel chloride hexahydrate ( $NiCl_2.6H_2O$ ), Paladium chloride ( $PdCl_2$ ), Cadimum chloride dihydrate ( $CdCl_2.2H_2O$ ), Mercury chloride ( $HgCl_2$ ), Copper Sulphate

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pentahydrate ( $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ ) Zinc sulphate heptahydrate ( $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ ), Lead nitrate  $\text{Pb}(\text{NO}_3)_2$ , Chromium trichloride hexahydrate ( $\text{CrCl}_3 \cdot 6\text{H}_2\text{O}$ ), Ethanol 99% ( $\text{CH}_3\text{CH}_2\text{OH}$ ), Dimethyl formamide 99.5% (DMF), Dimethyl sulphoxide 99.5% (DMSO), Carbon tetra chloride 99.5% ( $\text{CCl}_4$ ), Chloroform 99% ( $\text{CHCl}_3$ ), Toluene 99% ( $\text{C}_7\text{H}_8$ ).

#### b – Instruments ;

- Elemental analysis for the new ligand (L) and complexes were determined by calibration type: Linear Regression Euro EA Elemental Analysis were made in Al – mustansiriyah University and Ministry of Science and Technology .
- Melting points were determined by Gallen – Kamp apparatus .
- IR spectra were recorded as KBr discs in the range  $(4000 - 400)\text{cm}^{-1}$  using Shimadzu – FTIR .
- UV –Visible spectra were recorded by Shimadzu –UV–Vis –160A Ultra violet spectrophotometer at  $25\text{ }^\circ\text{C}$ , using 1 cm quartz cell and examined at the range of  $(200 - 1100)\text{ nm}$  at  $10^{-3}\text{ M}$  in DMSO - Atomic Absorption (A.A) technique have been measured using a Shimadzu AA 680 G atomic absorption spectro photometer .
- Molar conductivity of the complexes were measured on PW 9526 digital conductivity in DMSO at  $10^{-3}\text{ M}$  - Magnetic susceptibility were recorded by Magnetic susceptibility balance , model , MsB – MK1 .and made in AL – Nahrain University .

#### Preparation

##### Synthesis of ortho amino hydrazobenzene( L )

Orthoaminophenyl thiol (0.125 gm, 0.001 mol) and phenyl hydrazen (0.10 gm , 0.001 mol ) in ethanol (25  $\text{cm}^3$ ) was stirred under reflux for 4 hours . The precipitate was filtered and recrystallized from hot absolute ethanol , fine orange yellow crystals were obtained , m.p  $78\text{ }^\circ\text{C}$ , yield 91% and it is soluble in most organic solvents as indicated in Table(1) .

##### Synthesis of the metal complexes

One mole of ethanolic solution of metal salts was added to two moles of the ligand (L) except with Cr – complex the mole ratio was (1:1) [ $\text{CrCl}_3 \cdot 6\text{H}_2\text{O}$  (0.26 gm.1.00 mmol) ,  $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$  (0.24gm.1.00 mmol),  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$  (0.24 gm.1.00 mmol),  $\text{Cu SO}_4 \cdot 5\text{H}_2\text{O}$  (0.24 gm.1.00 mmol),  $\text{ZnSO}_4 \cdot 7\text{ H}_2\text{O}$ ( 0.28 gm.1.00 mmol),  $\text{PdCl}_2$ (0.17gm.1.00 mmol),  $\text{CdCl}_2 \cdot 2\text{ H}_2\text{O}$  (0.22gm.1.00 mmol),  $\text{HgCl}_2$ (0.27 gm.1.00 mmol) and  $\text{Pb}(\text{NO}_3)_2$ (0.33 gm.1.00 mmol)] was added to (0.19gm.1.00 mmol) in  $\text{Cr}^{+3}$  complex and (0.398gm.2.00 mmol) in the cases of  $\text{Co}^{+2}$  ,  $\text{Ni}^{+2}$  ,  $\text{Cu}^{+2}$  ,  $\text{Zn}^{+2}$  ,  $\text{Pd}^{+2}$  ,  $\text{Cd}^{+2}$  ,  $\text{Hg}^{+2}$  ,and  $\text{Pb}^{+2}$  of the ligand (L). At room temperature the mixture was stirred for 30 minutes.

The product was filtered and washed with distilled water and dried under vacuum. The Color, melting point, yield, metal analysis and solubility of the ligand and its complexes are given in ( Table I ) .

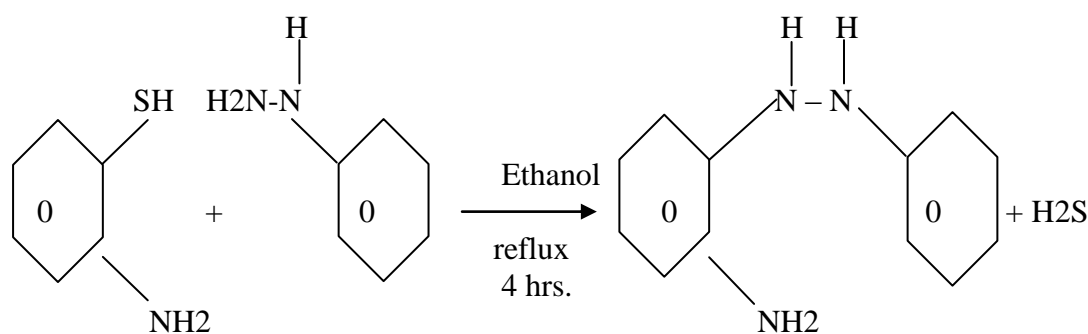
**Table (1): Color, melting point, yield, metal analysis and solubility for the ligand (L) and its complexes**

NO	Compound	no.of mole and gm	Color	m. p. C° or (dec.)	Yield %	(Metal analysis Found calculated )				Solubility
						C %	H %	N %	M %	
1	(L)C12 H13 N3	1.00-2.00 0.19-0.39	orange yellow	78 C°	91	72.350 (72.361)	6.500 (6.532)	21.103 (21.105)	—	EtOH, CCl4, DMF, CHCl3, DMSO, C7H8
2	L – Cr Cl3 .H2O	1.00 0.26	green	212(dec.)	83	37.399 (38.348)	3.091 (3.994)	10.989 (11.185)	13.82 (13.84)	DMF, DMSO
3	L2 – Co Cl2	1.00 0.24	violet	250(dec.)	72	54.491 (54.545)	4.810 (4.924)	14.999 (15.909)	11.20 (11.17)	DMF, DMSO
4	L2 – Ni Cl2	1.00 0.24	green	202(dec.)	87	54.532 (54.576)	4.812 (4.927)	14.999 (15.918)	11.10 (11.12)	DMF, DMSO
5	L2 – Cu SO4	1.00 0.24	blue	180 C°	76	51.523 (51.659)	4.231 (4.663)	15.130 (15.918)	11.36 (11.39)	DMF, DMSO
6	L2 – Zn SO4	1.00 0.28	Light yellow	164 C°	63	—	—	—	11.63 (11.67)	DMF, DMSO
7	L2 – Pd Cl2	1.00 0.17	brown	196(dec.)	81	50.015 (50.053)	4.500 (4.518)	14.499 (14.598)	18.46 (18.49)	DMF, DMSO
8	L2 – Cd Cl2	1.00 0.22	Light yellow	184 C°	78	—	—	—	19.34 (19.33)	DMF, DMSO
9	L2 – Hg Cl2	1.00 0.27	violet	200 C°	93	42.918 (43.017)	3.012 (3.883)	12.231 (12.546)	29.93 (29.94)	DMF, DMSO
10	L2 – Pb (NO3)2	1.00 0.33	light yellow	190 C°	88	—	—	—	28.39 (28.40)	DMF, DMSO

dec. = Decomposition

**Results and Discussion:**  
**Synthesis and characterization of the ligand orthoamino hydrazobenzene (L).**

The ligand was obtained by the reaction of one mole of orthoaminophenyl thiol and one mole with slightly excess of phenyl hydrazine;



Elemental analysis (C, H, N) confirmed the purity of the ligand (L) with the formula  $C_{12}H_{13}N_3$ . The infrared spectrum of the ligand (L) in the solid state does not contain the  $\nu$  (S – H) which appears in the starting material orthoaminophenyl thiol at (2600–2500)  $cm^{-1}$  region. This indicates

the displacement of SH in orthoaminophenyl thiol by means of –NH – NH –. Bands in the 3379  $cm^{-1}$ , [15] 1583  $cm^{-1}$  and 1301  $cm^{-1}$  are diagnostic of the primary aromatic amine. Bands at 3299  $cm^{-1}$  are due to [16]  $\nu$  (N – H) groups.

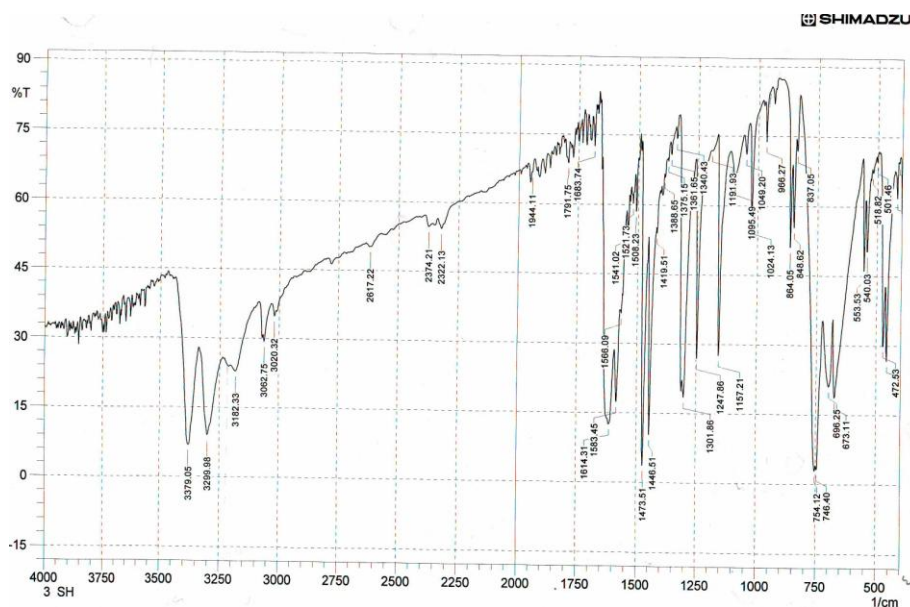


Fig. (1): Infrared spectrum of the ligand (L)

The infrared spectra of all complexes showed, the multi bands in the range (3284, 3247, 3211)  $cm^{-1}$  and split or broader band at (1608–1593)  $cm^{-1}$  and 1311  $cm^{-1}$  with the formation of a new

absorption bands for the coordination bands (M–N) in the region (540–515)  $cm^{-1}$ . Suggesting the coordination [17,18] through nitrogen atom.

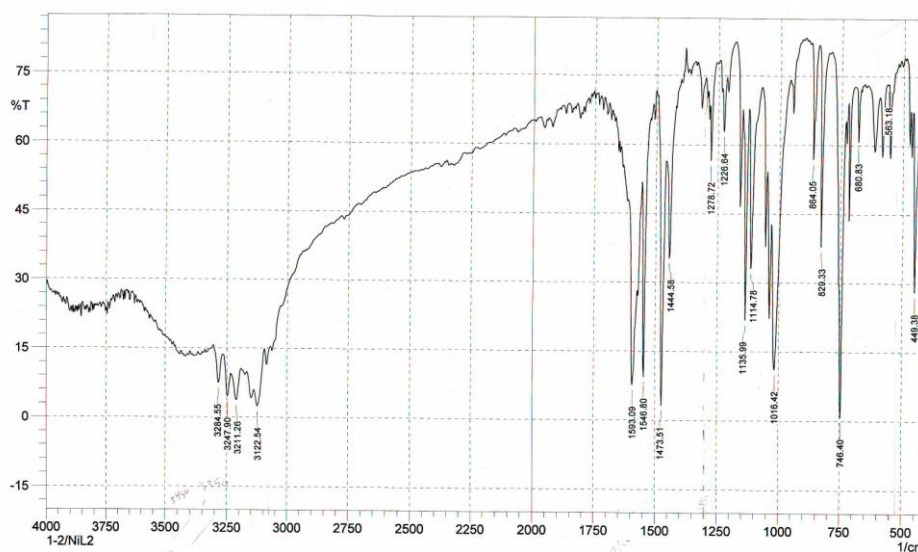


Fig. (2): Infrared spectrum of  $L_2.NiCl_2$

The bands characteristic of coordinated water and (M–O) are seen in Cr complex in  $827\text{ cm}^{-1}$  and [19]  $510\text{ cm}^{-1}$ . The Cu, Zn complexes showed bands at  $1030\text{ cm}^{-1}$ ,  $975\text{ cm}^{-1}$  and  $698\text{ cm}^{-1}$  are due to coordinate sulphato[20]. The

Pb complex spectrum exhibits band at  $688\text{ cm}^{-1}$  and  $925\text{ cm}^{-1}$  due to coordinate nitrate ion[21,22]. (Table 2) showed the characteristic stretching vibrational frequencies( $\text{cm}^{-1}$ ) located in the FT–IR of the ligand and its complexes.

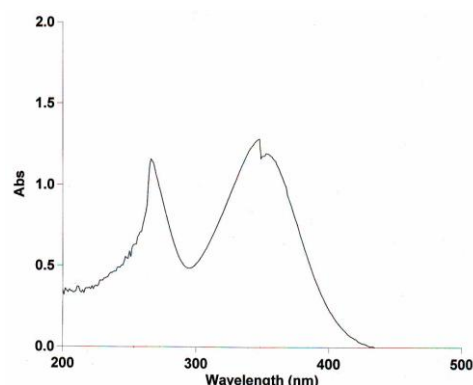
**Table (2): characteristic stretching vibrational frequencies ( $\text{cm}^{-1}$ ) located in the FT – IR of the ligand (L) and its complexes**

NO	Compound	$\nu(\text{N-H})\text{cm}^{-1}$	$\nu(\text{C-H})\text{cm}^{-1}$ Ar.	$\nu(\text{N-N})\text{cm}^{-1}$	$\nu(\text{M-N})\text{cm}^{-1}$	$\nu(\text{M-X})\text{cm}^{-1}$	$\nu(\text{M-O}) + \nu(\text{OH}_2)\text{aq. cm}^{-1}$	$\nu\text{ SO}_4\text{-2} + \nu\text{ NO}_3\text{-cm}^{-1}$
1	(L) C12 H13 N3	3379(asym) m 3299(sym) m 1614 s, 1583m 1301 s, 1247 s	3182 m 3062 m 837 (di-sub) 754 (mon . sub)	1446 s	—	—	—	—
2	L – Cr Cl3 .H2O	3380 (asym) br 3310 (sym) br 1608 s, 1575m 1311s, 1260 w	3170 w 3060 s	1446 m 1455 (sh)	520 m	420 w	550 m 827m	—
3	L2 – Co Cl2	3417 (asym) br 3379 (sym) br 1618 s, 1580 m 1300(sh), 1305m 1245m	3110 w 3050 w	1446m	520 m	405 w	—	—
4	L2 – Ni Cl2	3284 (asym)m ,3247,3211(sym)m 1610(sh), 1593 m 1315 m, 1278 m	3122 m 3050 m	1444 m	520 s	415 m	—	—
5	L2 – Cu SO4	3377(asym) br 3299(sym)br 1610s, 1580 m 1305 m, 1260 (sh)	3150 m 3060 m	1447 m	530 m	415 (sh)	—	1030m 975 m 695m
6	L2 – Zn SO4	3386,3355(asym)s 3292 (sym) m 1616 s, 1575 m 1310 s, 1249 s	3182 m 3062 m	1446 m 1440 m	525 m	410 w	—	1026 m 975 m 698 m
7	L2 – Pd Cl2	3440 (asym) br 3350 (sym)br 1608s, 1550 s 1303 m, 1270 m	3170 m 3049 m	1450 m	520 m	405 w	—	—
8	L2 – Cd Cl2	3434 (asym) br 3382 (sym) br 1604 s, 1583 m 1307 s, 1250 m	3150 m 3056m	1450 m	538 m	400 w	—	—
9	L2 – Hg Cl2	3450 (asym) br 3355 (sym) br 1606s, 1560 m 1303 m, 1270 (sh)	3130 w 3050 w	1455 m	515 m	410 w	—	—
10	L2 – Pb (NO3)2	3386 (asym) br 3346 (sym) br 1616 s, 1560 m 1305 s, 1288 m	3120 s 3060 w	1448 m 1455 (sh)	540 w	405 w	—	925 s 688 s

Ar. = aromatic, Ali = aliphatic, asym = asymmetric, sym = symmetric,  
sh = shoulder br = broad, m = medium, s = strong, w = weak

**The UV – Visible spectra of the ligand ( L ) and complexes**

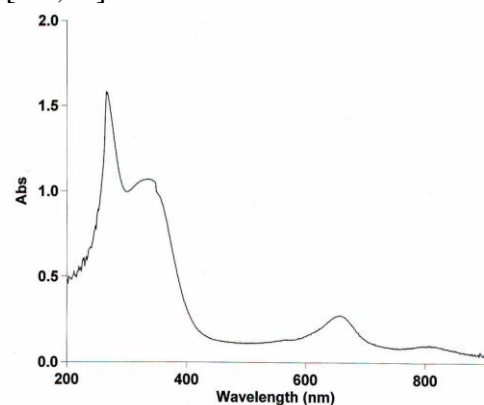
The UV – Visible spectrum of the ligand (L) in DMSO solution exhibited strong absorption bands at(266 nm, 37593 cm<sup>-1</sup>) and (348 nm, 28735cm<sup>-1</sup>), (353nm, 28328cm<sup>-1</sup>). This may attributed to the  $\pi - \pi^*$  and  $n - \pi^*$  transition[23].



**Fig. (3): UV – Visible spectrum of the ligand (L)**

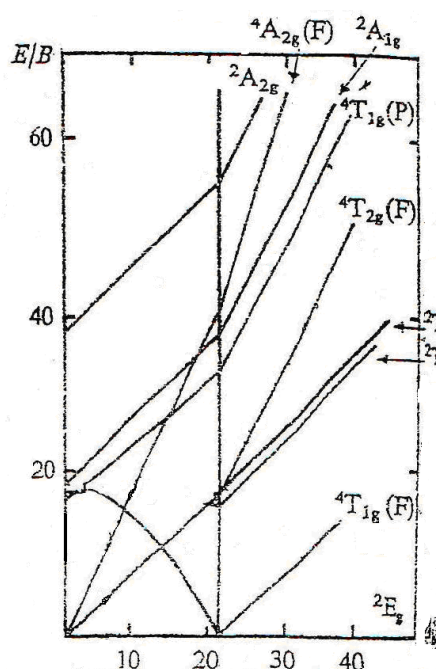
The UV – Visible spectrum of Cr<sup>+3</sup> complex showed one band in the region (580 nm, 17241 cm<sup>-1</sup>) is due to  ${}^4A_{2g} \longrightarrow {}^4T_{2g}$  [24].

The UV-Visible spectrum of Co<sup>+2</sup> complex (fig4) showed three bands in the region(800nm ,12500cm<sup>-1</sup>) due to  ${}^4T_{1g} \longrightarrow {}^2E_g$ , (660 nm, 15151 cm<sup>-1</sup>) due to  ${}^4T_{1g} \longrightarrow {}^4A_{2g}$  and ( 520 nm , 19230cm<sup>-1</sup>)due to  ${}^4T_{1g}(F) \longrightarrow {}^4T_{1g}(p)$  [ 25,26 ] .



**Fig. (4) : UV – Visible spectrum of L2 . Co Cl2**

The ratio of 19230 cm<sup>-1</sup> to 12500 is 1.54 which fits with Tanaba–Sugano d<sup>7</sup> curve (fig 5) for E / B = 26 and  $\Delta_0 / B = 16$  .  $\Delta_0 = 12500 \text{ cm}^{-1}$  B complex = 12500 / 16 = 781 cm<sup>-1</sup> B for free Co<sup>+2</sup> = 971 cm<sup>-1</sup>  $\beta$ ( nephelavetic effect ) = B complex / B Co<sup>+2</sup> = 781 / 971 = 0.804 Which indicates of d – electron delocalization on the ligand hence a significant covalent character in the complex [27].



**Fig. (5): Tanabe- Sugano diagram of the Co+2 ion in octahedral complexes.**

The UV – Visible spectrum of Ni<sup>+2</sup> showed one band at (840 nm, 11904 cm<sup>-1</sup>) which is due to  ${}^3A_{2g} \longrightarrow {}^3T_{1g}$ . Cu<sup>+2</sup> complex appeared band at (670 nm, 14925 cm<sup>-1</sup>) is due to  ${}^2E_g \longrightarrow {}^2T_{2g}$ . The brown palladium complex exhibited a strong band at (405nm,24691cm<sup>-1</sup>)due to  ${}^1B_{1g} \longleftarrow {}^1A_{1g}$  [28] transition.

The UV – Visible spectra of Zn<sup>+2</sup>, Cd<sup>+2</sup>, Hg<sup>+2</sup> and Pb<sup>+2</sup> complexes showed shifted bands compared with free ligand (L) are due to charge transfer. (Table 3)

The molar conductance of all complexes in DMSO were found to be low which suggested coordination of anion to the metal [22].

The  $\mu_{\text{eff}}$  value of Cr, Co, Ni, Cu complexes are within the range (3.81,

3.83, 2.84, 1.73) [29,30] respectively expected for spin – free octahedral structures. Electronic spectra, Conductance in (DMSO), Magnetic moment (B.M) of the ligand and its complexes are given in Table (3).

**Table (3) Electronic spectra (DMSO), Conductance in DMSO, Magnetic moment (B.M) of the ligand(L) and its complexes**

NO	Compound	$\lambda$ (nm) $\text{cm}^{-1}$	Assignment Bands	$\Lambda$ s. $\text{cm}^{-1}$ DMSO ( $10^{-3}$ M)	$\mu_{\text{eff}}$ (B.M)
1	(L) C12 H13 N3	37593 ( 266 ) 28735 ( 348 ) 28328 ( 353 )	$\pi - \pi^*$ $n - \pi^*$	—	—
2	L – Cr Cl3 .H2O	17241 ( 580 )	$4 A_{2g} \longrightarrow 4 T_{2g}$	6.35	3.81
3	L2 – Co Cl2	19230 ( 520 ) 15151 ( 660 ) 12500 ( 800 )	$4 T_{1g}(F) \longrightarrow 2 T_{1g}(P)$ $4 T_{1g} \longrightarrow 4 A_{2g}$ $4 T_{1g} \longrightarrow 4 E_g$	9.34	3.83
4	L2 – Ni Cl2	11904 ( 840 )	$3 A_{2g} \longrightarrow 3 T_{1g}$	10.92	2.84
5	L2 – Cu SO4	14925 ( 670 )	$2 E_g \longrightarrow 2 T_{2g}$	11.20	1.73
6	L2 – Zn SO4	28735 ( 348 )	Charge – Transfer	8.56	—
7	L2 – Pd Cl2	24691 ( 405 )	$1 B_{1g} \longleftarrow 1 A_{1g}$	9.21	—
8	L2 – Cd Cl2	27932 ( 358 )	Charge - Transfer	7.73	—
9	L2 – Hg Cl2	28571 ( 350 )	Charge - Transfer	10.34	—
10	L2 – Pb ( NO3 )2	25188 ( 397 )	Charge - Transfer	11.35	—

B. M = Bohr Magnetron

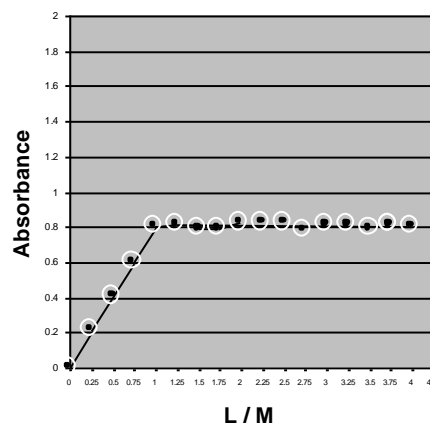
### Study of $\text{Cr}^{+3}$ , $\text{Co}^{+2}$ complexes formation in solution

The complexes of the ligand (L) with selected metal ions ( $\text{Cr}^{+3}$ ,  $\text{Co}^{+2}$ ) were studied in solution using ethanol as solvent, in order to determine (M:L) ratio in the prepared complexes , following molar [31] ratio methode . A series of solutions were prepared having a constant concentration (C)  $10^{-3}$  M of the hydrated metal salts and the

ligand (L). The (M:L) ratio was determined from the relation ship between the absorption of the observed light and the mole ratio (M:L) found to be (1:1) for ( $\text{Cr}^{+3}$ ) complex while (1:2) for ( $\text{Co}^{+2}$ ) complex. fig 6,7. The result of complexes formation in solution are shown in (Table4,5).  $V_m$  = volume of metal in ml(M)  $V_l$  = volume of ligand in ml(L)

**Table 4: cotinuos variation slop for Cr<sup>+3</sup> ion λ ( 392 nm )**

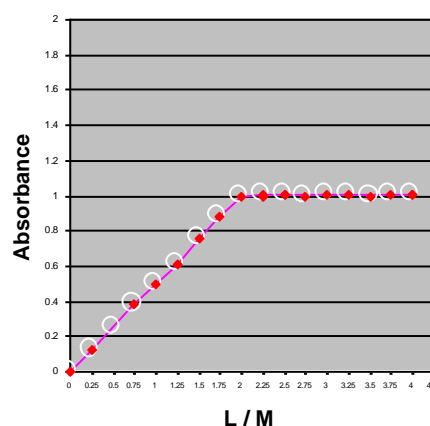
L – Cr Cl3 .H2O		
V M	VL	Abs
1 ml	0.25	0.22
1	0.50	0.41
1	0.75	0.60
1	1	0.80
1	1.25	0.81
1	1.50	0.79
1	1.75	0.79
1	2	0.82
1	2.25	0.83
1	2.50	0.82
1	2.75	0.78
1	3	0.81
1	3.25	0.81
1	3.50	0.79
1	3.75	0.81
1	4	0.80



**Fig (6): continuous variation slop for Cr<sup>+3</sup> ion λ ( 392 nm )**

**Table 5: continuous variation slop for Co<sup>+2</sup> ion λ ( 348 nm )**

L2 – Co Cl2		
V M	VL	Abs
1 ml	0.25	0.12
1	0.50	0.25
1	0.75	0.38
1	1	0.50
1	1.25	0.61
1	1.50	0.76
1	1.75	0.88
1	2	1.00
1	2.25	1.01
1	2.50	1.01
1	2.75	0.99
1	3	1.01
1	3.25	1.01
1	3.50	0.99
1	3.75	1.01
1	4	1.01



**Fig (7): continuous variation slop for Co<sup>+2</sup> ion λ ( 348 nm )**

The stability constant of the (1:1) [metal : ligand] (eq.1) or (1:2) [metal : ligand] (eq.2) complex was evaluated using the following equations :-

$$K_f = 1 - \alpha / \alpha^2 c \dots (1)$$

$$K_f = 1 - \alpha / 4\alpha^3 c^2 \dots (2)$$

$$\alpha = A_m - A_s / A_m \dots (3)$$

(α) is the degree of the dissociation, (c) is the concentration of the complex (10<sup>-3</sup> M . (A<sub>m</sub>) and (A<sub>s</sub>) are the absorbance of the partially and fully formed complex respectively Table

(6). The absorbance of the solution were measured at (λ<sub>max</sub>) of the maximum absorption .The molar absorptivity (ε<sub>max</sub>) (eq.4) has been calculated using equation; A=ε<sub>max</sub>.b.c ..... (4)

(A) is the average of three measurement of the absorption containing the same amount of metal ion and three fold excess of ligand , (b) is the path way of the quartz cell usually equal 1 cm .

**Table ( 6 ):A<sub>s</sub> , A<sub>m</sub> , K<sub>f</sub> , ε<sub>max</sub> and λ<sub>max</sub> of the Cr<sup>+3</sup> and Co<sup>+2</sup> complexes**

NO	Compound	A <sub>s</sub>	A <sub>m</sub>	α	Formation constant (K <sub>f</sub> )	Molar Absorptivity ε <sub>max</sub> L . mol <sup>-1</sup> .cm <sup>-1</sup>	λ <sub>max</sub> (nm)
1-	Cr – complex	0.80	0.83	0.0361	7.378x10 <sup>5</sup>	3122	392
2-	Co – complex	0.50	1.01	0.504	9.680x10 <sup>5</sup>	2111	348



The atomic absorption analysis was used to confirm the molar ratio calculation of [metal:ligand] (M:L) for the Synthesis of complex as well. The results showed a ratio M:L(1:2) for all complexes except for Cr complex which was (1:1)Table ( I ) .

#### Biological activity Study:

The biological activity of the prepared new ligand and its complexes were studied against selected types of microorganisms which include gram positive bacteria like Streptococcus pyogenes, Staphylococcus aureus and

gram Negative bacteria like E. coli, Pseudomonas aeruginosa and yeast like Candida albicans, in agar diffusion method, which is used (DMSO) as a solvent, and we are used these Antibiotics disc which include Cephalosporin, Amoxicillin and Nystatin as control. Agar diffusion method involves the exposure of the zone of inhibition toward the diffusion of microorganisms on agar plate. The plates were incubated for [24] hrs at (37C°), The zone of inhibition of bacterial growth around the disc was observed .Tables (7), (8), (9).

**Table (7) Effect of ligand and its complexes on gram positive bacteria**

Ligand or complex	Diameter of inhibition zone (mm)at concentration mg/ml		Diameter of inhibition zone (mm)at concentration 5mg/ml	
	<i>Streptococcus pyogenes</i>	<i>Staphylococcus aureus</i>	<i>Streptococcus pyogenes</i>	<i>Staphylococcus aureus</i>
Amoxicilin	27.4	27	29.2	28.6
(L) C12H13N3	17.8	17.6	18.2	17.8
L-CrCl3.H2O	14.2	14	15	14.2
L2-CoCl2	14	14	14.6	14.2
L2-NiCl2	16.2	15.4	17	15.6
L2-CuSO4	14.4	14	14.6	14.6
L2-ZnSO4	17	17	17.4	17
L2-PdCl2	13.8	14	14.2	14.6
L2-CdCl2	16.2	15.6	16.4	16
L2-HgCl2	17.8	17.6	18.2	17.8
L2-Pb(NO3)2	16.2	15.6	16.8	16

**Table (8)Effect of ligand and its complexes on gram negative bacteria**

Ligand or complex	Diameter of inhibition zone (mm)at concentration mg/ml		Diameter of inhibition zone (mm)at concentration 5mg/ml	
	<i>E. coli</i>	<i>Pseudomonas aeruginosa</i>	<i>E. coli</i>	<i>Pseudomonas aeruginosa</i>
Cephalosporin	26.4	25.8	28.6	27.2
(L) C12H13N3	16.6	16.4	17.2	17.8
L-CrCl3.H2O	12.6	12	13.4	14.2
L2-CoCl2	14.8	14	15.6	14.2
L2-NiCl2	16	15.8	16.4	15.6
L2-CuSO4	15	14.6	15.8	14.6
L2-ZnSO4	17.4	17	18.4	17
L2-PdCl2	14.8	14.6	15.6	14.6
L2-CdCl2	16	16	16.4	16
L2-HgCl2	18	17.8	18.2	17.8
L2-Pb(NO3)2	15	14.8	15.2	16

**Table (9) Effect of ligand and its complexes on *Candida albicans***

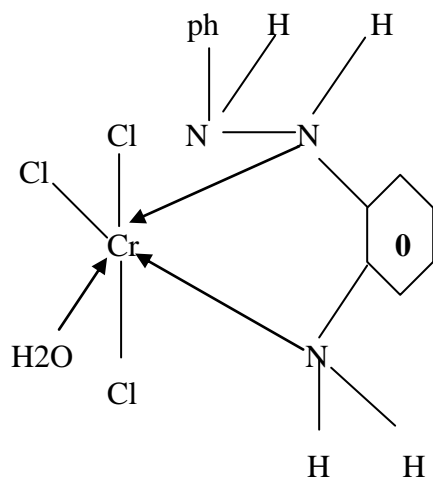
Ligand or complex	Diameter of inhibition zone (mm)at concentration mg/ml		Diameter of inhibition zone (mm)at concentration 5mg/ml	
Nystatin	15.6		18.4	
(L) C12H13N3	12.2		12.4	
L-CrCl3.H2O	7.6		8	
L2-CoCl2	9.4		9.6	
L2-NiCl2	8.2		8.2	
L2-CuSO4	11		11.2	
L2-ZnSO4	12		12.2	
L2-PdCl2	9		9.2	
L2-CdCl2	9.4		9.8	
L2-HgCl2	13.4		13.6	
L2-Pb(NO3)2	8		8.2	

**Conclusion:**

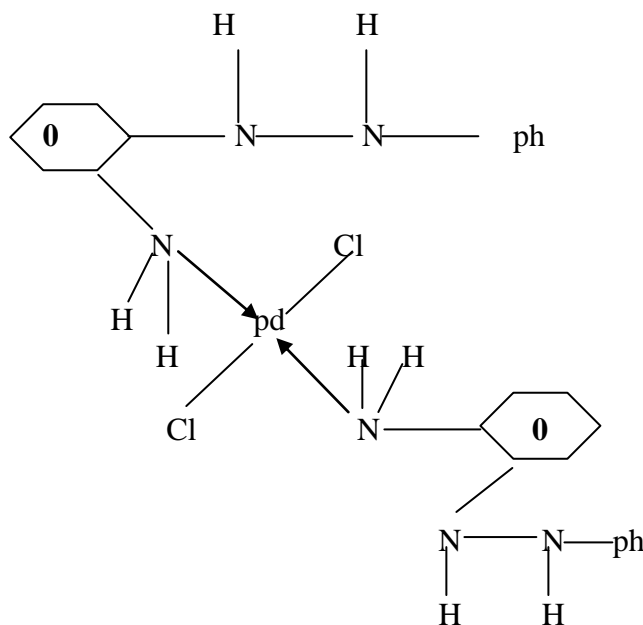
A series of complexes of  $Cr^{+3}$ ,  $Co^{+2}$ ,  $Ni^{+2}$ ,  $Cu^{+2}$ ,  $Zn^{+2}$ ,  $Pd^{+2}$ ,  $Cd^{+2}$ ,  $Hg^{+2}$ ,  $Pb^{+2}$ , with ortho amino hydrazo

benzene (L) have been prepared and characterized .

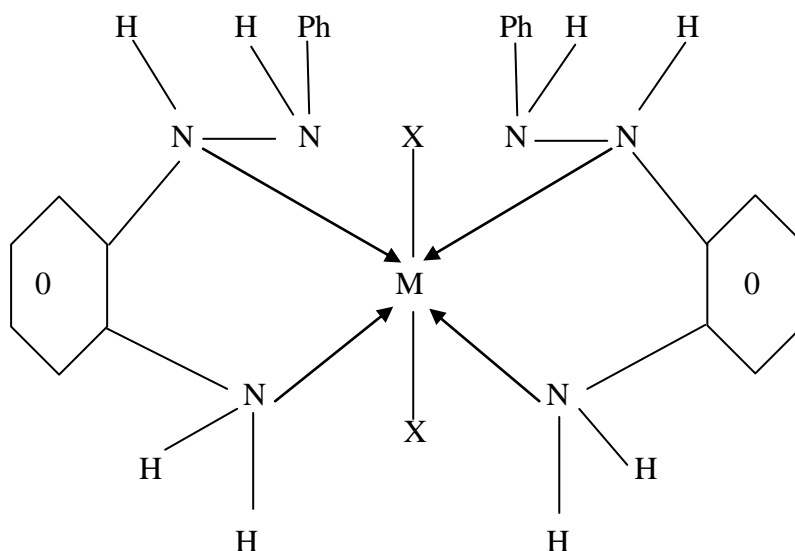
The bidentate ligand (L) (N,N) is binding metal ions forming octahedral structure as follow ;



$M^{+3} = Cr$

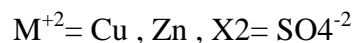
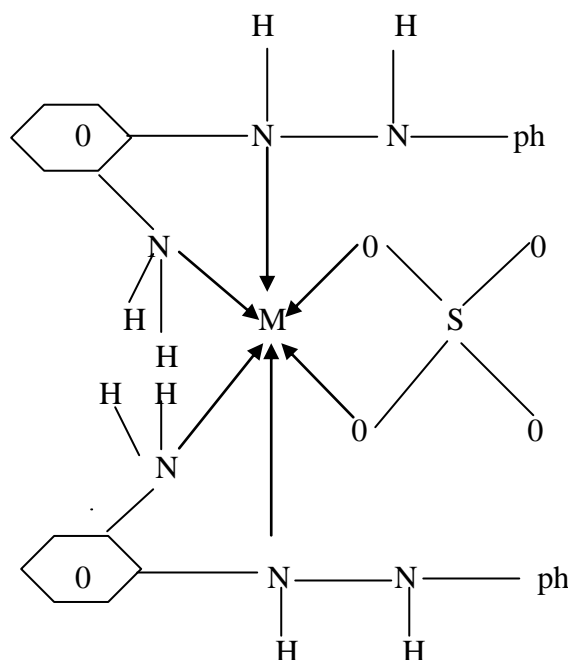


$M^{+2} = Cu, Zn, Pd$



$M^{+2} = Co, Ni, Cd, Hg, X = Cl^{-1}$

$M^{+2} = pb, X = NO3^{-1}$



Biological effects of new ligand and its complexes indicated that the new ligand than its complexes exhibited almost equal or more antibacterial activity against both gram positive and gram negative bacteria. Generally the results of the prepared complexes exhibited antimicrobial activity toward bacteria more than yeast candida albicans.

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## تحضير, دراسة طيفية وبايولوجية لبعض المعقدات الفلزية مع اورثوامينوهيدرازو بنزين

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### الخلاصة:

حضر الليكاند (L) اورثو امينو هيدرازو بنزين من تفاعل اورثو امينو فنيل ثايول مع فنيل هيدرازين وبنسبة 1:1 وشخص بواسطة التحليل الدقيق للعناصر (N, H,C), الأشعة تحت الحمراء والأشعة فوق البنفسجية - المرئية كما حضرت وشخصت معقدات أملاح بعض ايونات العناصر الثنائية التكافؤ ( Ni , Cu , Zn , Pd , Cd , Hg , Pb ) والثلاثي التكافؤ (Cr) . استخدمت تقنيات التحليل الدقيق للعناصر (N, H,C), طيف الأشعة تحت الحمراء , الأشعة فوق البنفسجية- المرئية , التوصيلية الكهربائية , الامتصاصية الذرية والخواص المغناطيسية لتحديد تركيب المعقدات واستنتاج من التحاليل ان المعقدات لها شكل ثنائي السطوح حول ايون الفلز مع الليكاند(L) ثنائي السن (N,N) مع pd مكونا " مربع مستوي . وقد تم حساب قيم  $\alpha$  ,  $k_f$  ,  $\epsilon_{max}$  للمعقدات وحساب  $\beta$  لمعقد الكوبلت كما تم دراسة الفعالية البيولوجية للليكاند ومعقداته وقد أظهرت النتائج امتلاكها فعالية متفاوتة اتجاه أنواع البكتريا Streptococcus pyogenes , Candida albicans E.coli , , Pseudomona aeruginosa , Staphylococcus aureus ,