ISSN: 0975-8585

Research Journal of Pharmaceutical, Biological and Chemical Sciences

Synthesis, Spectroscopic Studies and Antimicrobial Activity of Mono and Binuclear Metal Complexes of New Azine Ligand Derived From 4, 6-Diacetylresorcinol.

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ABSTRACT

New azine ligand H_2L was synthesized by condensation of 4,6-diacetylresorcinol dihydrazone (DARDH) and isatin (IS) with molar ratio (1:2). Reaction of the ligand with metal acetate of copper (II), cobalt (II), nickel (II) and zinc (II) in molar ratios 1:1 and/or 1:2 (L:M) produced mono and binuclear metal complexes. The structure of the prepared ligand and its complexes were proved by elemental analysis, FTIR, electronic, mass, ^{1}HNMR and ESR spectral analysis as well as magnetic moments and conductance measurements. The data of spectroscopic, magnetic susceptibility and conductivity proved that the H_2L ligand acts as dibasic [tri, tetra and hexadentate] ligands. The coordination sites with all the metal ions are phenolic oxygen, azomethine nitrogen and isatinic oxygen. The complexes exhibited different geometrical structures such as: square planar, tetrahedral, and octahedral arrangement around the central metal ions. The ligand and some metal complexes showed anti microbiological activity.

Key word: 4,6-diacetylresorcinol, 4,6-diacetylresorcinol-dihydrazone, isatin and microbiological activity.

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INTRODUCTION

Recently, many authors have been used 4,6-diacetylresorcinol (DAR) as a starting point for the generation of multidentate S/N/O or O/N/O symmetrical chelating agents by condensation with thiosemicarbazide or semicarbazone to yield the corresponding bis (thiosemicarbazide) H_4L^2 or bis (semicarbazone) H_4L^2 ligands, respectively [1]. They have been reacted H_4L^1 and H_4L^2 with $M(NO_3)_2.6H_2O$ [M=Co (II) or Ni (II)] giving dimeric complexes with H_4L^1 and binuclear complexes with H_4L^2, revealing the tendency of S to form bridges. The bimetallic complexes derived from tridentate or tetradentate or hexadentate Schiff bases or/and hydrazones of 4,6-diacetylresorcinol of some di- and tri-valent transition metals have been synthesized and characterized by many authors using different spectroscopic methods [2-10]. The biological activity of those ligands and their complexes were studied and reported in literatures [11, 12] and revealed that they were used as model system for variety of biological reactions such as oxygen transport [13], photocatalytic water splitting [14] and used ad chemo-optical sensor for assessment of α-amylase activity in human-saliva [15]. Mixed-ligand complexes including 4,6-diacetylresorcinol as a primary ligand have been synthesized and characterized [16-18] because of their potential applications in the microelectronic industry, chemical vapor deposition of metals and as drugs [19, 20]. Isatin has been used for designing potential bioactive agents and its derivatives were reported to possess a broad spectrum of antiviral activities [21]. Ligands containing isatin moiety with molar ratio 1:1 and 1:2 (L:M) are known to possess a wide range of pharmacological properties that include anticonvulsant [22], antibacterial, antifungal [23] and anti-HIV [24] activities. In the present work we have synthesized new azine ligand derived from 4,6-diacetylresorcinol and reacted with transition metal ions : zinc (II), cobalt (II) , copper (II) and nickel(II) in molar ratio 1:1 and 1:2 (L:M) and characterized using Elemental Analysis and different spectroscopic techniques as well their biological activity were evaluated.

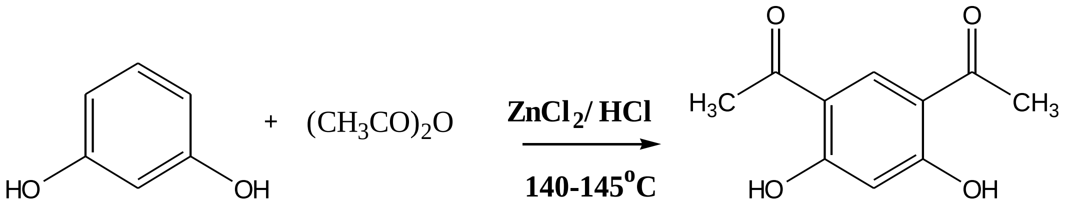
EXPERIMENTA PART

Materials

Resorcinol, hydrazine monohydrate, isatin, cobalt(II) acetate tetrahydrate, cadmium(II) acetate, zinc(II) acetate dihydrate, dimethylsulfoxid (Fluka, Switzerland); copper(II) acetate 1-hydrate (BDH, England); nickel (II) acetate tetrahydrate, acetic anhydride, chloroform, absolute ethanol (Riedel-de Haen, Germany); zinc(II) chloride extra pure, nitric acid 60% extra pure (Scharlan, Spain); acetic acid (glacial) (SURECHEM); acetone A.R. 99-85% (HAY-MAN, U.K.); (EDTA) (Analyticals), paraffin oil light B.P. (Pacegrove, U. K.);

Synthesis of 4,6-diacetylresorcinol (DAR)

4,6-diacetylresorcinol (I) was prepared by addition of resorcinol (2.00 gm, 18.16 mmol) to solidified $ZnCl_2$ (2.00 gm, 14.70 mmol) and (2.80 ml) of acetic anhydride, $(AcO)_2O$. The reaction mixture was heated to reflux at (140-145˚C) for (45 min) with continuous stirring, on a paraffin-oil bath, during which the color of the solution changed to dark red. After cooling add 8.00 ml (1:1 HCl: Water), with stirring. Reddish brown product formed which was left to coagulates, then filtered off and washed with water, then recrystallized from acetic acid. The yield was (0.67 gm, 45 %), and the melting point (178˚C) [25].



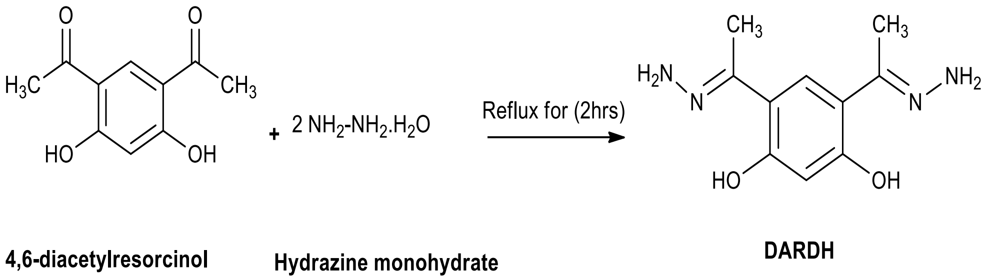
Resorcinol **acetic anhydride**

(I)

4,6-diacetylresorcinol

Synthesis of (H_2L) ligand derived from 4,6-diacetylresorcinol-dihydrazone (DARDH) and isatin (Is) with the molar ratio (1:2)

1- Synthesis of (DARDH)



The DARDH was prepared from an ethanolic solution of 4,6-diacetyl resorcinol (DAR) (1.49 gm, 10 mmol) in 100 ml absolute ethanol, and hydrazine monohydrate ($N_2H_4.H_2O$) (1.00 ml, 20 mmol) in 50 ml absolute ethanol in 250 ml round flask which were heated to reflux for (2 hrs.). In the beginning of the reaction, the color was buff, then turns to yellow at the end of the reaction. Yellow solid was obtained on hot. After complete the reaction, the mixture was left to cool. The yellow product was separated by filtration, washed with absolute ethanol and then air- dried. The yield was (1.77 gm., 80 %), its melting point > 300°C.

2- Synthesis of H_2L ligand

The H2L, ligand was prepared from an ethanolic solution of DARDH (2.22 gm, 10 mmol) in 100 ml absolute ethanol and isatin (Is) (2.94 gm., 20 mmol) in 100 ml absolute ethanol in 250 ml round flask which were heated to reflux for (3 hrs.). In the beginning of the reaction, the color was orange, and then turns to dark red at the end of the reaction. Dark red solid was obtained on hot. After complete the reaction, the mixture was left to cool. The dark red product was separated by filtration, washed with small portion of absolute ethanol and hot acetone and then air-dried. The yield was (3.60 gm. 75 %), its melting point > 300°C.

3- Synthesis of the complexes of H_2L ligand

An ethanolic or methanolic solution of metal acetate was added gradually with stirring to the H_2L ligand with the molar ratio 1:1 and 1:2 (L:M). The reaction was stirred using magnetic stirrer for 30 min. The mixture was heated to reflux for certain period for each complex as shown in Table (1). Most of the complexes were formed during the reflux. The products were isolated by filtration and washed with several portions of ethanol or methanol, then air-dried and dried in the oven. A complete description of the reactions for each of the obtained complexes was given in the Table (1).

Biological Study

Solutions of the ligands and their complexes were prepared with $5x10^{-4}$ M concentration in DMSO and were sterilized through microbiological filters. Equipped with filter paper discs of 7 mm diameter nozzle and then placed in a clean Petri dish and then in the futility of (autoclave) Device sterilization. Agar has been liquefaction in the glass tube (pipe) and left to cool until 45°C and then inoculated by adding 1 ml of farm modern bacteria and then pour the contents of the tube in a sterile Petri dish and leave to cool into to be a heart. In the case of the use of fungi the agar has been liquefaction and then poured into a straw in a sterile Petri dish and then solidifies to the field plate swab of cotton by fungi of the survey.

Added 0.05 ml prepared solution, the record (ligands or complexes) carefully by sterile pipit to CD absorbent paper then put through a sterile clamp in the center of the Petri dish with a simple click on the disk surface of the agar.

Petri dishes were putted in the autoclave at 30 °C temperature for 24 hours and then examined the Petri dishes and their impact on the area of bacterial growth and the results were recorded in the Tables [26, 27].

Physical measurements

Melting point

All melting points reported for the compounds are measured on a Stuart scientific melting point [SMP1] apparatus.

FT-IR spectra

The FT-IR spectra (4000-200 cm^{-1}) of the compounds were measured as Cesium Iodide discs using NICOET NEXUS 870.

UV-Visible spectra

The UV-Visible spectra of the investigated compounds were obtained using [Jasco V570, UV-Vis-Nir spectrophotometer] with one centimeter quartz cell.

^{1}H-NMR spectrum

The ^{1}H-NMR spectral measurements were carried out on 600 MHz-Bruker model 2000 DRX600 prob 5 m.m. BBO BB-1H.

Mass spectra

Mass spectra were carried out on GC-MS, Shimadzu, QP-5050A, with the detector gain (1.10 KV) and sampling rate (0.50 Sec.), at the temperature range 40-350°C.

Electronic spin resonance (ESR) spectrum

ESR spectra were recorded on the Bruker ELXSYS 500E. X-band with auto detection for peak without needs any calibration.

Magnetic measurements

Magnetic susceptibilities were measured by Gouy method at room temperature using Magnetic susceptibility balance Johnson matthey, catalytic system division York way, Royston, Hertfordshire.

Molar conductance measurements

The molar conductance was measured by HQ 14d (µs/cm), at 25°C, 5 x 10^{-4} M in DMSO.

Elemental analysis

Elemental analysis for C, H, N was performed by elemental analysis *vario ELIII*, and the metal cations were determined volumetrically by EDTA [28-30].

RESULTS AND DISCUSSION

Characterization of the synthesized azine ligand H_2L

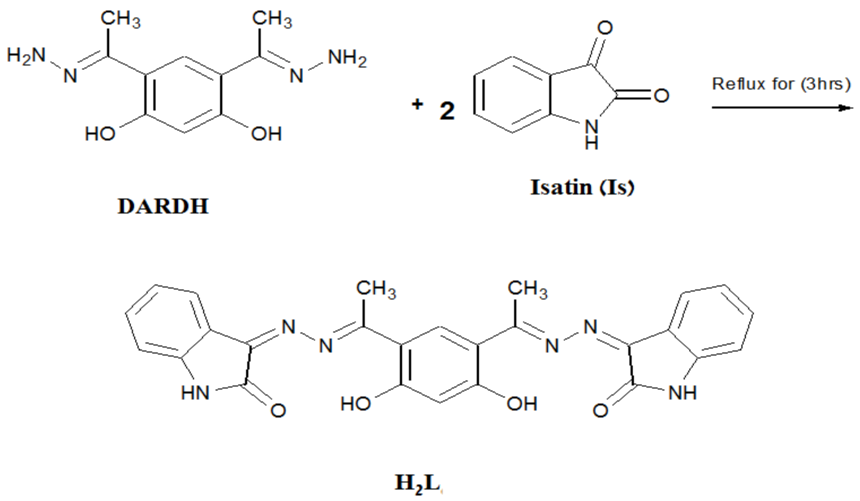
The ligand was characterized using the following techniques: elemental analyses for C, H and N, FTIR, ^{1}H-NMR, U.V. & visible and mass spectroscopic analysis.

IR spectrum figure (1) of the prepared azine ligand H_2L scheme (1) showed the following features: a broad band at 3446 cm^{-1} was assigned to ν(OH), strong band at 1622 cm^{-1} which was attributed to ν(C=N) of the azomethine group and a strong band at 1279 cm^{-1}, which was investigated to stretching vibration of the phenolic C-O bond; ν(C-O).

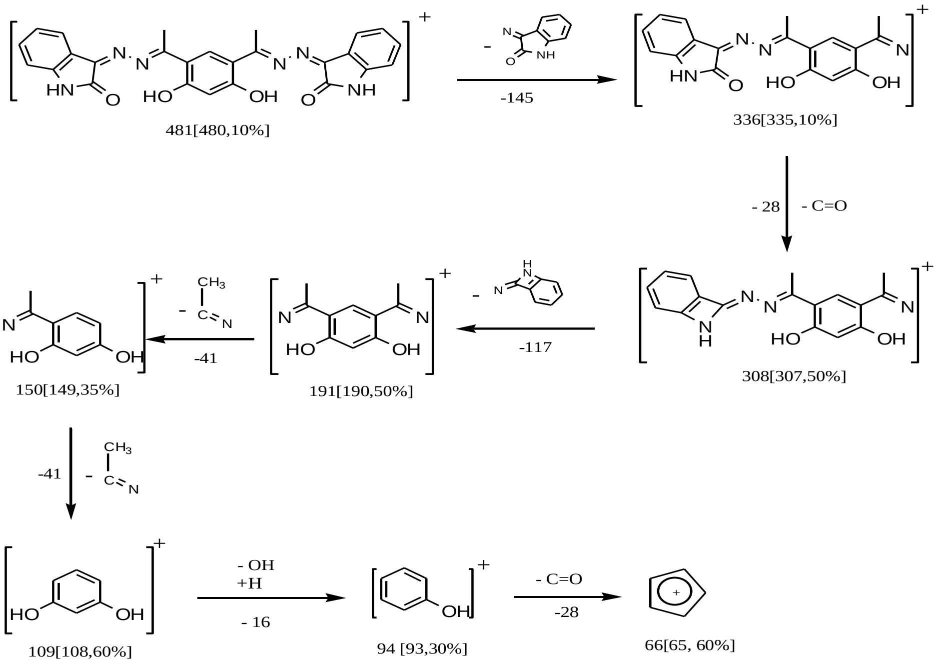
^{1}HNMR spectrum of the H_2L ligand dissolved in DMSO-d_6 solvent showed the following signals: 2.61, 2.55 ppm which were assigned to the protons of the methyl groups in two different environments [31]. The signals at 8.23, 6.52 ppm was assigned to the protons of aromatic groups. The protons of phenolic OH groups were shown at 13.65, 13.05 ppm. The signals at 11.03 and 10.90 ppm were assigned to the protons of NH group of the isatin moiety. The proton signals of the –OH phenolic groups and of –NH of the isatin moiety were disappeared by adding of D_2O, which indicated that these protons are acidic and the hydroxyl groups can coordinate to the metal ions.

Mass spectrum of the azine H_2L ligand showed the molecular ion peaks at 480 m/e which was coincident with its formula weights and confirmed the identity of its structures. The fragmentation pattern was shown in scheme (2)

The proposed structures after investigation of the obtained data are shown in the following Scheme (1). The results of the elemental analyses (Table 1) are in good agreement with the proposed formula.



Scheme (1) Structure of H_2L



Scheme (2) fragmentation of H_2L ligand dissolved in DMSO-d_6 solvent

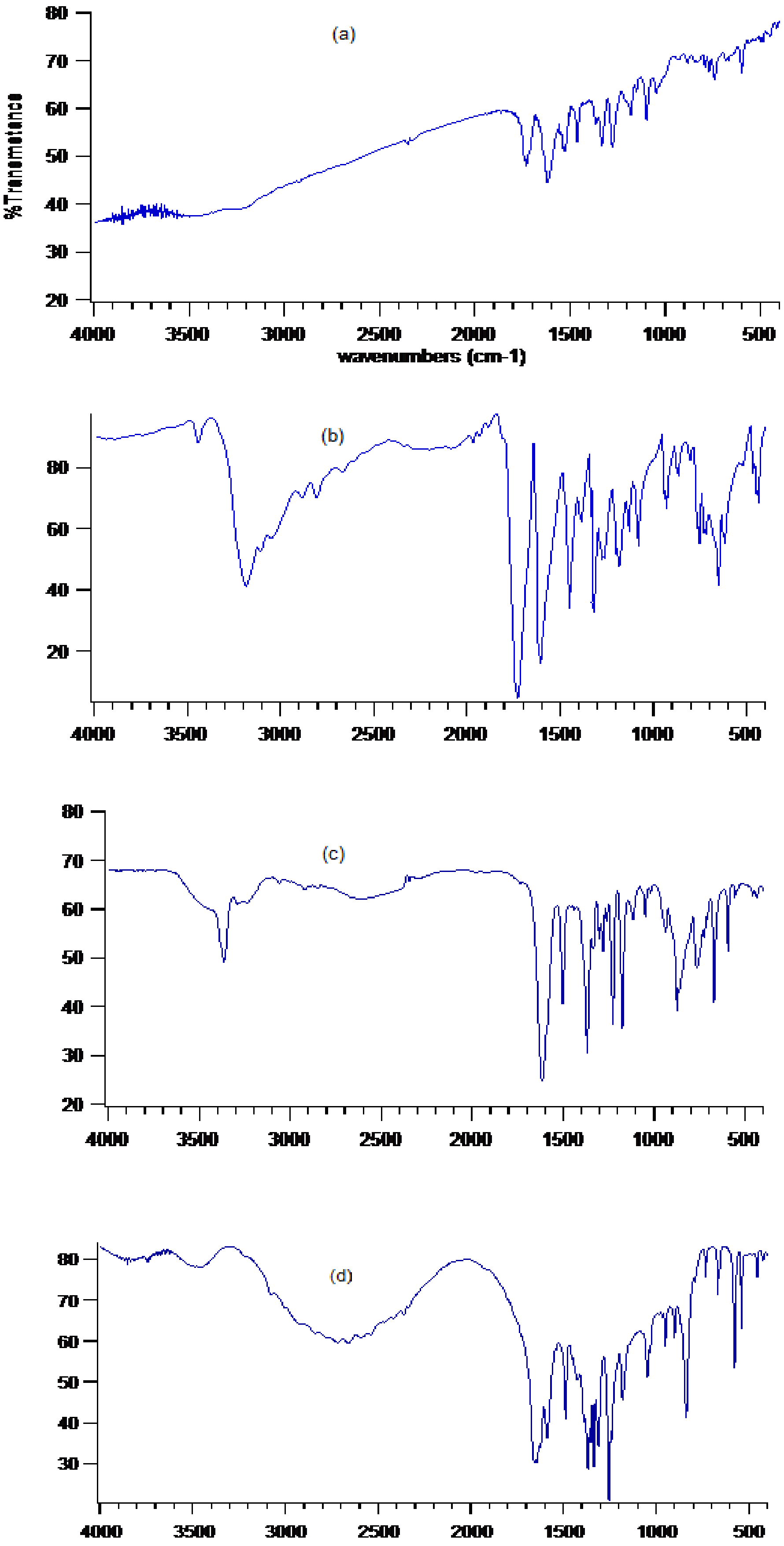


Figure 1. FT-IR spectra of: (a) H2L ligand, (b) isatin (IS), (c) DARDH, (d) 4,6-diacetylresorcinol (DAR)

	Ligand/ complexes	Period of reaction (hrs)	Molecular formula	M. Wt. (Yield %)	Color	M. p. °C a	Elemental Analyses Calc./(Found)%			
							C	H	N	M
	H_2L		$C_{26}H_{20}N_6O_4$	480.48 (75)	Deep red	2300	64.99 (65.12)	4.20 (4.19)	17.49 (16.1)	……
(1)	**$[Co_2(L)_2]$**	5	$C_{52}H_{36}N_{12}O_8Co_2$	1074.78 (84)	Deep brown	250	58.11 (57.46)	3.38 (4.06)	15.64 (14.91)	10.97 (10.09)
(2)	**$[Ni_2(L)_2]$**	5	$C_{52}H_{36}N_{12}O_8Ni_2$	1074.31 (49)	Deep brown	300	58.14 (58.3)	3.38 (3.47)	15.65 (16.02)	10.93 (10.18)
(3)	**$[Cu_2(L)_2]$**	5	$C_{52}H_{36}N_{12}O_8Cu_2$	1084.00 (82)	Black	300	57.62 (57.72)	3.35 (3.30)	15.51 (15.52)	11.72 (11.98)
(4)	**$[Zn(L)(CH_3COO)]$**	8	$C_{29}H_{22}N_6O_6Zn$	602,09 (87)	Deep red	300	55.69 (55.04)	3.67 (3.54)	13.92 (14.27)	10.83 (10.01)
(5)	**$[Co_2(L)(CH_3COO)_2(H_2O)_4]$**	8	$C_{30}H_{32}N_6O_{12}Co_2$	786.47 (77)	Deep brown	250	45.81 (45.59)	4.10 (4.49)	10.69 (10.78)	14.99 (15.03)
(6)	**$[Ni_2(L)(CH_3COO)_2(H_2O)_4]$**	5	$C_{30}H_{32}N_6O_{12}Ni_2$	784.08 (55)	Deep brown	300	45.84 (50.3)	4.10 (3.86)	10.69 (14.05)	14.93 (18.18)
(7)	**$[Cu_2 (L)(CH_3COO)_2]$**	5	$C_{26}H_{22}N_6O_6Cu_2$	641.58 (82)	Black	300	48.67 (51.21)	3.46 (3.75)	13.10 (12.26)	19.81 (18.06)
(8)	**$[Zn_2(L)(CH_3COO)_2]$**	8	$C_{30}H_{24}N_6O_8Zn_2$	727.33 (73)	Deep red	250	49.54 (49.19)	3.33 (3.11)	11.55 (11.81)	17.98 (17.59)

a All complexes were changed their colors at the cited temperature

Table 1 Physical and analytical data of the, H_2L ligand and its transition metal complexes with molar ratio 1:1 and 1:2 (L:M)

Characterization of the transition metal complexes of H_2L ligand

The azine, H_2L, ligand was prepared by the condensation of (DARDH) and isatin in the molar ratio 1:2. The H_2L ligand was reacted with Co(II), Ni(II), Cu(II) and Zn(II) ions with molar ratio 1:1 and 1:2 (L:M) to produce the corresponding metal complexes (1-8) as shown in Table (1). The solid metal complexes structures were characterized using elemental and IR, UV-visible, ESR spectral analyses as well as molar conductance and magnetic moment measurements. The coordination of the H_2L_c ligand with transition metal ions can take place via the phenolic oxygen atom of the DAR moiety and azomethine nitrogen atom, $(CH_3)C=N$, as well the oxygen of the isatin guinone form. The ligand H_2L, behaves as mono or dibasic and has at least one set of ONO donor sites; to react with all metal ions in the molar ratio 1:1, two sites of ONO donor sites; of the ligand molecule and the H_2L ligand behaves as di- or tribasic to react with all metal ions in the molar ratio, 1:2 (L:M). The elemental analyses Table (1) in agreement with proposed structures.

Table 2 Vibrational frequencies (cm^{-1}) of H_2L ligand and its transition metal complexes with its assignments

Table 2

	Ligand/complex	ν(OH)	ν(NH)	ν(MeC=N)	ν(C=N)	ν(C=O)	ν(M-O)	ν(M-N)
	H_2L	3446 m,br	3212 m,br	1622 vs	1532 s			
1	$[Co_2(L)_2]$	3435 m,br	3206 m,br	1619 vs	1529 s	1725 s	558 w	334 w
2	$[Ni_2(L)_2]$	3485 m,br	3200 m,br	1612 vs	1541 s	1729 s	565 w	333 w
3	$[Cu_2(L)_2]$	3429 m,br	3207 m,br	1619 vs	1515 s	1734 s	568 w	334 w
4	$[Zn(L)(CH_3COO)]$	3438 m,br	3212 m,br	1619 vs	1529 s	1733 s	562 w	330 w
5	$[Co_2(L)(CH_3COO)_2(H_2O)_4]$	3445m,br	3210 m,br	1616 vs	1526 s	1728 s	492 w	329 w
6	$[Ni_2(L)(CH_3COO)_2 (H_2O)_4]$	3437 m,br	3205 m,br	1618 vs	1531 s	1733 s	492 w	352 w
7	$[Cu_2 (L)(H_2O)_2]$	3435 m,br	3208 m,br	1619 vs	1530 s	1731 s	566 w	334 w
8	$[Zn_2(L)(CH_3COO)_2]$	3416 m,br	3208 m,br	1619 vs	1528 s	1718 s	530 w	333 w

IR spectra of the metal complexes:

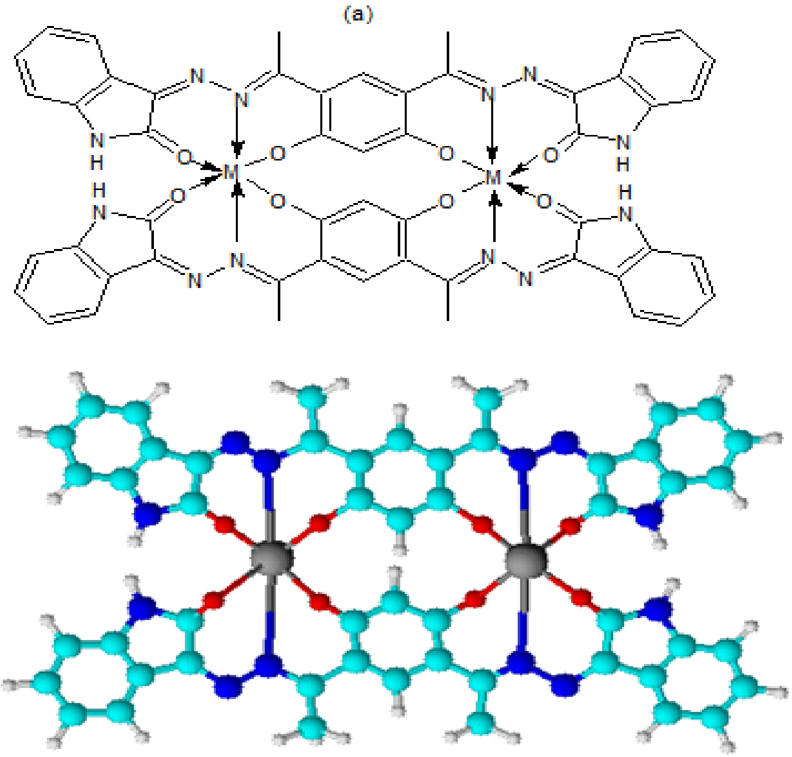
The IR spectral data of the H_2L ligand, and its transition metal complexes (1-4) shown in Table (2) revealed the following characteristic absorption bands for NH, C=N, and phenolic-OH groups as well M-N and M-O. Comparison of the IR spectra of the metal complexes with the free, H_2L, ligand revealed the following evidences which confirmed the formation of the complexes. The first evidence is the shifted band observed around 3485-3429 cm^{-1} in all the complexes (1-4) with molar ratio (1:1) was assigned to v (OH) group after deprotonation. The second evidence is the shift of the observed strong band in the ligand spectrum at 1622 cm^{-1} of the azomethine group v(C=N) to 1619-1612 cm^{-1} in all the complexes indicating the participation of azomethine nitrogen atom of the H_2L, ligand in complex formation. The new bands located in all the complexes at 568-558 cm^{-1} and 334-330 cm^{-1} ranges were assigned to v(M-O) and v(M-N) non-ligand bands which confirmed and supported the above interpretation of the complex formation in the molar ratio 1:1.

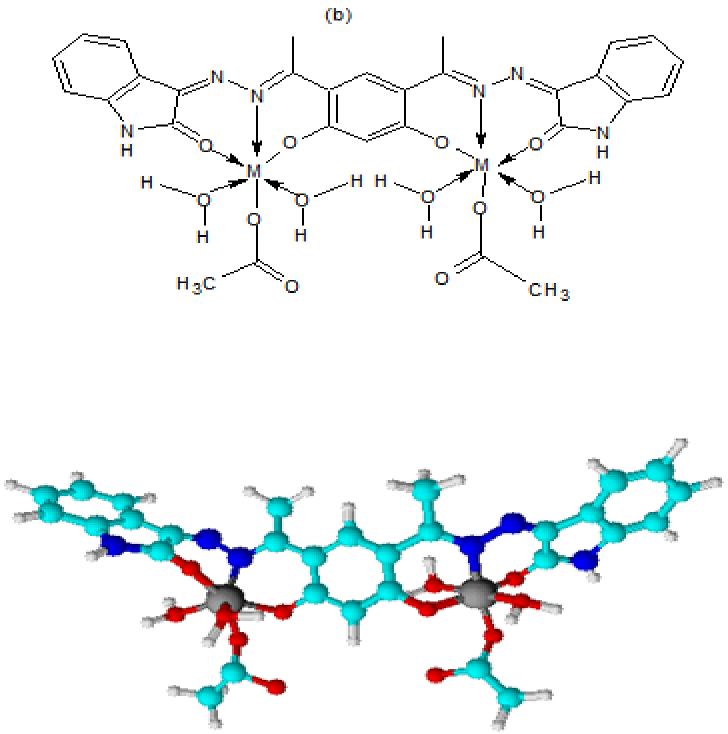
On the other hand the comparison of the IR spectral data of the metal complexes in the molar ratio 1:2 (L:M)) as shown in Table (2) for the complexes (5-8) with that of the free H_2L, ligand gave these features : The appearance of the strong band at 1622 cm^{-1} assigned to v($CH_3C=N$) azomethine group for the H_2L, free ligand, was shifted to 1619-1616 cm^{-1} in all the complexes(5-8).. The shifted bands observed around 3445-3416 cm^{-1} molecules in all complexes(5-8) were assigned to v (OH) The bands observed around 1733-1718 cm^{-1} were assigned to the v(C=O) of the isatin moiety which was observed at 1733 cm^{-1} in the free H_2L ligand which indicate that the ligand reacted throughout the isatinic oxygen atom. The new bands located in all the complexes at 566-492 cm^{-1} and 352-329 cm^{-1} ranges were assigned to v(M-O) and v(M-N) non-ligand bands which confirmed and supported the above interpretation of the complex formation.

Electronic, ESR spectra, Molar conductance and Magnetic moment measurements

The metal complexes of the H_2L, ligand, are insoluble in most common solvents except N,N-Dimethylformamide (DMF). Table (3) lists the electronic spectral data, the magnetic moments (μ_{eff}), of the metal ions in their solid complexes, as well, their molar conductance in solution.

The electronic spectra of the Co (II) complexes (1) and (5) showed red shift absorption band at 440 nm and 448 nm respectively, which assigned to the n-π* transition. The absorption band observed at 550 nm and 560 nm for Co (II)-complexes (1) and (5) respectively were assigned to $^4T_{1g}$ (P) $\leftarrow ^4T_{1g}$ transition for the six coordinate cobalt (II) complexes [31]. The measured magnetic moments (μ_{eff}) was 2.07 B.M. for complex (1) which was consistent with low spin octahedral geometrical arrangement and 1.9 B.M. for complex (5) which was consistent with low spin octahedral geometrical structure as shown in the Scheme (3). The molar conductance values were 1.33 and 2.29 ohm^{-1}cm^{2}mol^{-1} for (1) and (5) respectively, which indicate the neutral nature of the complexes.





Scheme (3): The structures of: (a) $[M(L)_2]$ complex (1), (2), (b) $[M(L)(CH_3COO)_2(H_2O)_4]$ complex (5), (6) [M= Co^{2+} or Ni^{2+}]

● **M,** ● **O,** ● **N,** ● **C,** ● **H**

The electronic spectra of the Ni (II) complexes (2) and (6), red shit absorption band at 452 nm for (2) and 442 nm, (6) complexes which was attributed to the n→π* transition. The band observed at 550 nm (2) and 554 nm (6); respectively, was suggested due to spin allowed transition to $^3T_{1g}(F)$ level for the octahedral geometrical arrangement around Ni^{+2} with the magnetic moment (μ_{eff}) = 2.1 and 1.78 BM respectively [31]. The molar conductances were 0.59 and 0.12 $ohm^{-1}cm^2mol^{-1}$ for (2) and (6); respectively which confirmed the non-electrolytic behavior of the two complexes. From the above data one can safely suggest the octahedral geometrical arrangement around Ni^{+2} with complexes (2) and (6) respectively as shown in Scheme (3).

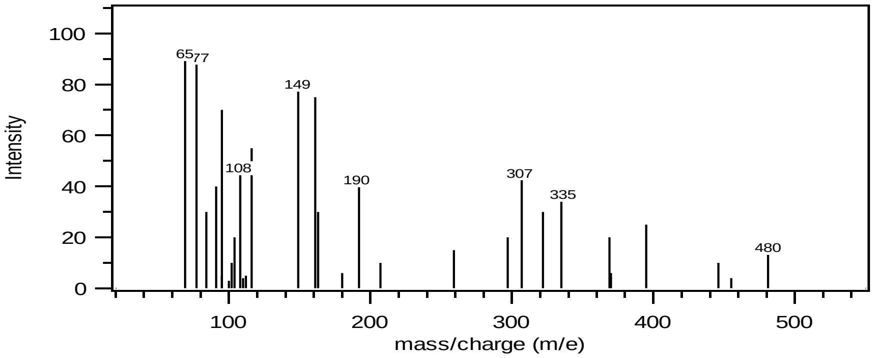
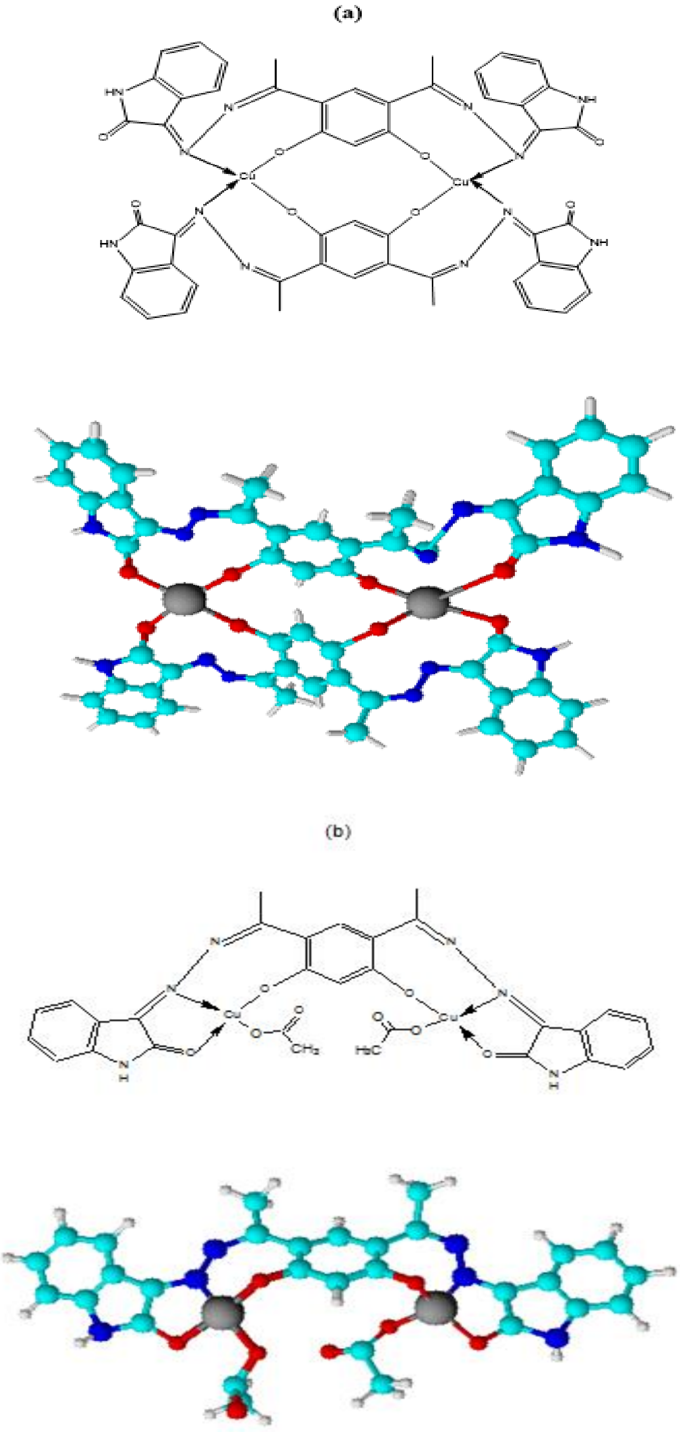


Figure 2 Mass spectrum of the H_2L ligand

The electronic spectra of the Cu (II) complexes (3) and (7) showed bathochromic shift absorption band at 434 nm and 551 nm for (3) and 430 nm, 560 nm for (7) complexes which were assigned for n→π* and d-d transition respectively. The measured value of the magnetic moment for Cu (II) complex (3) was 2.02 B.M. which consistent with the square planer geometric structure Scheme (4: a). The molar conductance of (3) was 3.10 $ohm^{-1}cm^2mol^{-1}$, which indicate the non-electrolytic complex. X-Band ESR spectrum of Cu (II) complex (3) was recorded in the solid state at 25 °C and shown in figure (3). The spectrum showed two bands with g = 2.0177 and 2.1834 which was consistent with the square planar geometry around the Cu (II) environment in the complex (3). On the other hand, the measured value of the magnetic moment (μ_{eff}), of the Cu(II) complex (7) was 1.68 B.M., which consistent with the square planar geometrical structure as shown in Scheme (4: b). The molar conductance of (7) was 5.74 ohm^{-1}cm^{2}mol^{-1}, which indicate a non-electrolytic complex.



Scheme (4): The structures of: a) $[Cu_2(L)_2$ complex (3),
b) $[Cu_2(L)(CH_3COO)_2]$ complex (7)

● Cu, ● O, ● N, ● C, ● H

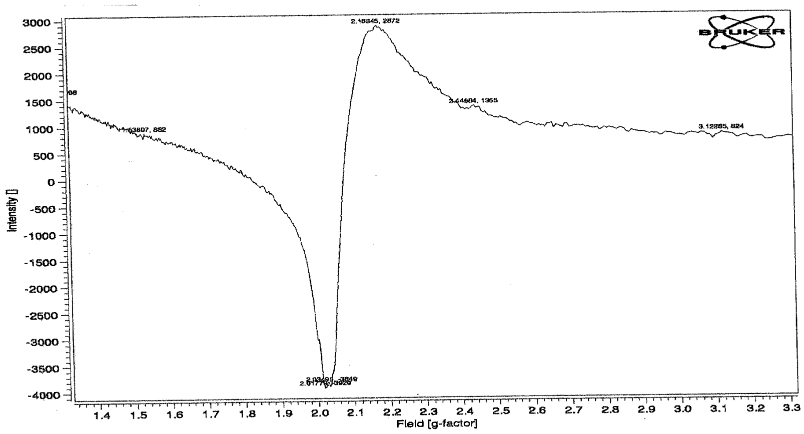
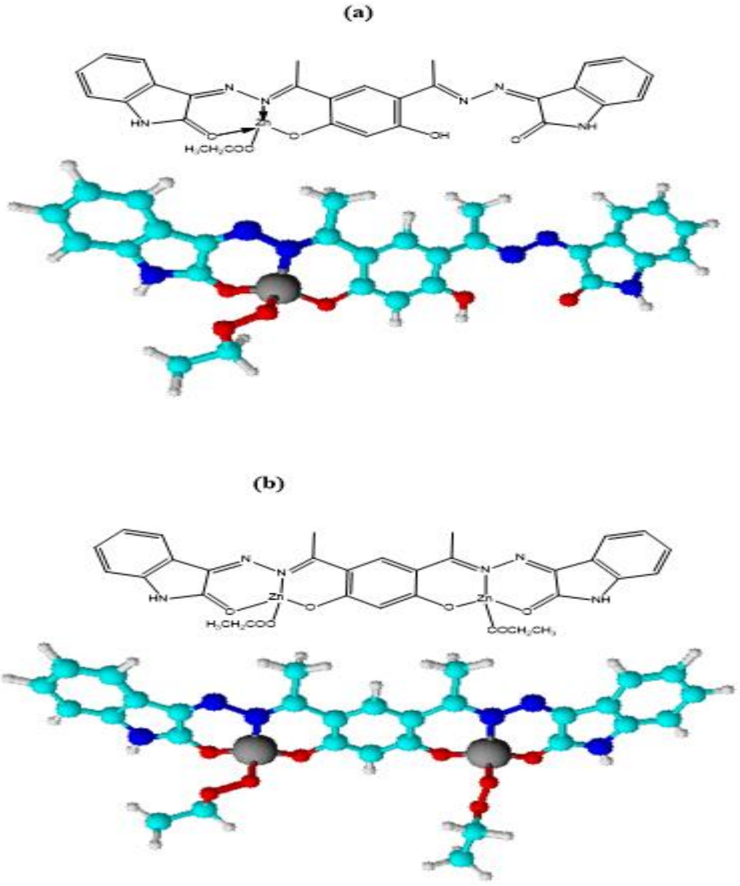


Figure 3: ESR spectrum of the $[Cu_2(L)_2]$ complex (3)

The electronic spectra of the Zn (II) complexes (4) and (8) showed absorption band at 437 nm and 440 nm respectively, which was assigned for the n→π* transition, that is shifted from 430 nm for the free H_2L, ligand. The measured values of the magnetic moment (μ_{eff}), for these complexes were indicated the diamagnetic and there is no either electronic d-d transition or significant magnetic moment. Molar conductances of the complexes in DMF solution were 1.49 and 0.78 $ohm^{-1}cm^2mol^{-1}$; respectively, which indicate the neutral nature of the complexes. The expected structure for the complexes (4) and (8) is tetrahedral structure (scheme 5).



Scheme (5): The structures of: a) $[Zn(L)(CH_3COO)]$ complex (4), b) $[Zn_2(L)(CH_3COO)_2]$ complex (8)

● Zn, ● O, ● N, ● C, ● H

Table 3 Electronic absorption bands (nm), magnetic moments, and conductances of the metal complexes of H2L ligand

Ligand/complexes	Phenyl ring	$^1L\rightarrow{}^1A$	C=N	π→π*	n→π*		d-d transition		Magnetic moment (B.M.) µeff	Λ^b
H_2L	-0.7	292	-1.1	337	-1.1	430			1.8	9.12
1[$Co_2(L)_2$]	-0.97	290	-0.96	329	(0.9)2	446	-0.4	550	1.76	1.33
2[$Ni_2(L)_2$]	-0.5	290	-0.6	328	-0.58	452	-0.5	550	1.66	0.59
3 [$Cu_2(L)_2$]	-1.1	290	-1.25	331	-1.35	434	-0.28	551	Dia	3.1
4 [$Zn(L)(CH_3COO)$]	-1.97	293	-2.46	330	-2.6	437			1.8	1.49
5[$Co_2(L)(CH_3COO)_2(H_2O)_4$]	-0.47	291	-0.46	326	-0.4	450	-0.16	560	1.76	2.29
6[$Ni_2(L)(CH_3COO)_2$ $(H_2O)_4$]	-66	289	-0.7	328	-0.65	442	-0.48	554	1.66	0.12
7[Cu_2 $(L)(H_2O)_2$]	-1.01	290	-1.03	323	-0.98	430	-0.27	560	Dia	5.74
8[$Zn_2(L)(CH_3COO)_2$]	-0.5	290	-0.55	332	-0.53	440				0.78

Biological activity for H_2L ligand and its complexes

Evaluation of the biological activity for the ligand and its complexes (1-8) was tested on some of bacteria and a type of yeast (*Salmonella* group C, *Salmonella* group D, *Staphlycoccus aureus*, *E. coli.,* Candida (yeast)) showed that the results shown in the table (4) Ligand and its complexes were discouraging bacteria *Candida* (yeast) and *salmonella group C* between the very good (+ +) and good (+). On the other hand Cu complex (1) did not show an impact on the bacteria *Candida* (yeast), whereas bacteria *Candida* (yeast) is an excellent grown compared with control. The results showed that non affected of the bacteria *Salmonella group D*, *Staphlycoccus aureus* and *E. coli.* with either ligand and its complexes because they did not indicate any inhibition for the growth of microbial.

Table 4 Effects of the H_2L ligand and its complexes

Compound No.	*Candida (yeast)*	*Salmonella group D*	*Salmonella groupC*	*Staphlycoccus aureus*	*E. coli*
H_2L	+	-	++	-	-
1	-	-	++	-	-
2	++	-	++	-	-
3	++	-	++	-	-
4	+	-	++	-	-
5	+	-	+	-	-
6	+	-	+	-	-
7	+	-	+	-	-

CONCLUSION

The reaction of 4,6-diacetylresorcinol-dihydrazone (DARDH) and isatin (Is) with the molar ratio 1:2 forming azine H_2L ligand.the ligand acts as dibasic (tri, tetra and hexadentate) forms in its mono or/and binuclear metal complexes. The types of coordination sites and geometries of the ligand sits around the transition metal ions were determined using the spectral (IR, UV and ESR), magnetic moments and molar conduction measurements. The types of bonding sites are phenolic oxygen, azomethine nitrogen and isatinic oxygen. The complexes exhibited square planar, tetrahedral, and octahedral structures. The ligand and its complexes were evaluated against *Candida* (yeast) and *salmonella group C* and they showed discouraging them between very good (+ +) and good (+) results except for complex of $[Co_2(L)_2]$ did not show an impact on the bacteria *Candida* (yeast). Thabrew MI, Gove CD, Hughes RD, McFarlane IG, Williams R. J Ethnopharm 1995; 49: 69-76.

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