

# Synthesis and characterization some of metals(II) complexes with 2-(6-methoxy naphthalene-2-yl) propionic acid (2-hydroxy benzylidene) hydrazone ligand

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## ABSTRACT

New Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Ca(II) hydrazone complexes which have the molar ratio 1:2 metal to ligand of the general formula  $[M(L)_2]$  where L=2-(6-methoxy naphthalene-2-yl) propionic acid (2-hydroxy benzylidene) hydrazone. All the synthesized complexes have been characterized by elemental analysis, molar conductance, UV-Visible, magnetic susceptibility, atomic absorption, infrared spectra, <sup>13</sup>C, <sup>1</sup>H,NMR spectroscopy and X-ray powder diffraction. These measurements indicate that the molecular structure for these complexes have an octahedral geometry.

**Keywords:** Naproxen, metal complexes, X-ray powder diffraction.

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## 1. INTRODUCTION

A large number of hydrazone of naproxen and their complexes have been studied due to their important properties<sup>[1]</sup>. The high affinity for chelation of the hydrazone towards metal ions is utilized in preparing there solid complexes<sup>[2]</sup>.

These compounds have a wide range of biological application such as anticonvulsant, antimicrobial, antimycobacterial, anticancer, antiplatelet, antimalarial, antimycobacterial, anthelmintic, antidiabetic and trypanocidal activities<sup>[3]</sup>. Metal(II) complexes of naproxen derivatives has a better therapeutic than a naproxen itself and has improved physicochemical characteristics, therefore these complexes is more effective in therapy and less ulcerogenic than either naproxen alone<sup>[4]</sup>. The majority complexes usually act as polydentate N and O donors with formation of mononuclear and polynuclear complexes<sup>[5]</sup>. The aim of the research is the synthesis and characterization of new cobalt(II), nickel(II), Copper(II), Zinc(II), Cadmium(II) and Calcium(II) coordination complexes with hydrazone ligand derived from naproxen.

## 2. EXPERIMENTAL

### A. Materials and Measurements

All the chemical were of reagent grade used as supplied without further purification and used throughout this investigation from Merck, B.D.H. and Fluke.

## B. Analytical and physical measurements

Melting point and decomposition temperature were determined using STRUART-SMPID melting point apparatus. The elemental contents were determined using spectral methods by atomic absorption spectrophotometer type NOVAA 350 Scientific Equipment. Elemental analysis was carried out on CHN analyzer type vector, model EA 3000 V.3.0 single Euro. The molar conductance measurement was carried out in solution ( $10^{-3}$  M) at  $25^{\circ}\text{C}$ , using BC 3020 professional Bench top conductivity, infrared spectra of ligand and its complexes were recorded using Shimadzu FTIR as KBr pellets at the range ( $400\text{-}4000\text{ cm}^{-1}$ ). UV-visible spectra measurements were recorded using Shimadzu 160 spectrophotometer at  $10^{-3}$  M, ligand and complexes in DMF solvent at  $25^{\circ}\text{C}$  using 1cm quartz cell at the range ( $200\text{-}900\text{ nm}$ ). Magnetic susceptibility measurement was made by Faraday method at  $25^{\circ}\text{C}$  using Magnetic susceptibility balance of Johnson Matthey Catalytic System Division, England. NMR spectra were recorded on Bruker Shield 300 MHz using deuterated DMSO- $d_6$  as a solvent. X-ray powder diffraction data for complex 6 was measured at General Company for Geological Survey and Mining - Baghdad by using Shimadzu X-ray diffraction 7000 model 2009 and the crystal data for complexes were analyzed by using Match program Version 1.6 C.

## 3. PREPARATION OF THE LIGANDS AND their COMPLEXES

### Synthesis of 2-(6-methoxy naphthalene-2-yl) propionic acid ethyl ester

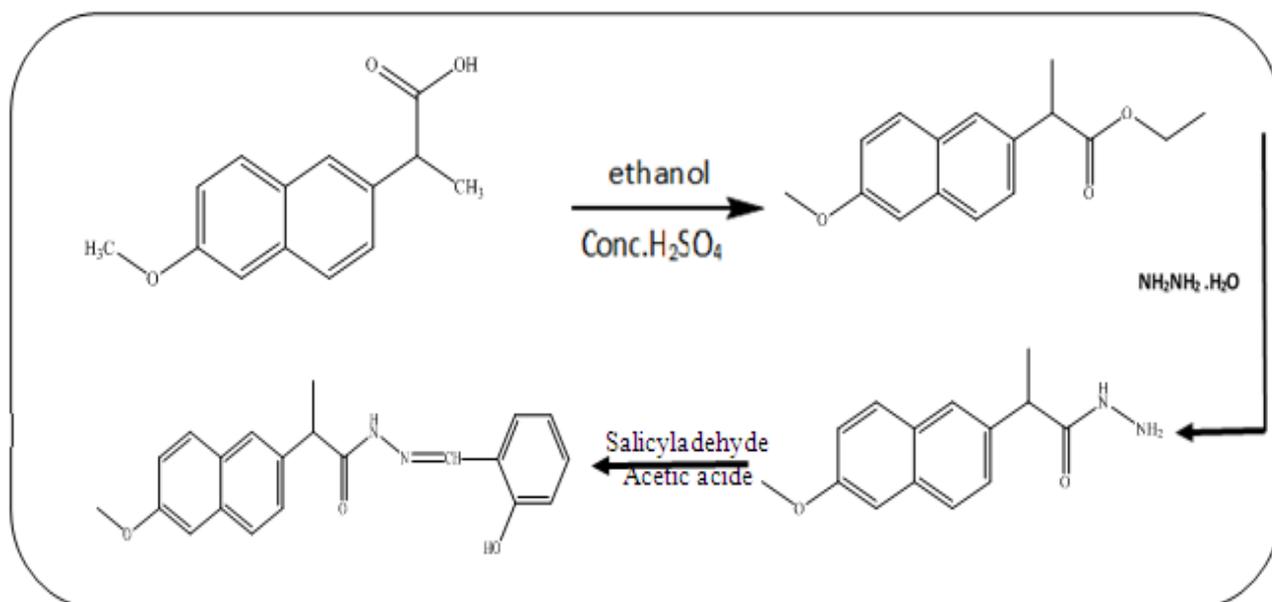
A mixture of 0.03 mol Naproxen (6.9 gm), 150 ml of absolute ethanol and 3 ml of concentrated sulfuric acid was refluxed for 6-8 hrs. The mixture was cooled to room temperature and neutralized with  $\text{NaHCO}_3$  solution. The precipitate that formed was collected and washed with water twice then recrystallized from ethanol to afford the ester as a white crystal<sup>[6]</sup> with m.p. =  $87\text{-}89^{\circ}\text{C}$ . The yield (79%), Anal. Calc. for  $\text{C}_{16}\text{H}_{18}\text{O}_3$ : C, 74.41; H, 6.97 Found: C, 74.22; H, 6.78 %).

### Synthesis of 2-(6-methoxy naphthalene-2-yl) propionic acid hydrazide

A mixture of 2-(6-methoxy naphthalene-2-yl) propionic acid ethyl ester (0.02 mole, 5.16 g) and an excess of hydrazine hydrate in 15 ml of ethanol was refluxed for 24 hrs. The solvent was removed and the precipitate that formed was recrystallized from ethanol to afford the hydrazide<sup>[7]</sup> as a yellow crystal with m.p. =  $135\text{-}139^{\circ}\text{C}$ , Anal. Calc.  $\text{C}_{14}\text{H}_{16}\text{N}_2\text{O}_2$ : C, 68.85; H, 6.55; N, 11.47, Found: C, 68.66; H, 6.43; N, 11.34%.

### Synthesis of 2-(6-methoxy naphthalene-2-yl) propionic acid (2-hydroxy benzylidene) hydrazone (L)

A mixture of equimolar ratio from 2-(6-methoxy naphthalene-2-yl) propionic acid hydrazide (0.01 mole, 2.44 gm) and Salicylaldehyde (0.01 mole, 1.22 gm) with few drops of glacial acetic acid in 20 ml of absolute ethanol was refluxed for 7 hrs. The mixture was concentrated to half and the precipitate that formed was collected and recrystallized from ethanol to afford the hydrazone (L)<sup>[8]</sup> with m.p. =  $165\text{-}167^{\circ}\text{C}$ . Anal. Calc.  $\text{C}_{21}\text{H}_{20}\text{N}_2\text{O}_3$ : C, 72.41; H, 5.74; N, 8.04; Found: C, 72.17; H, 5.43; N, 7.77 %.



Scheme (1): synthesis of the Ligand (L)

### General synthetic method of the complexes

A solution of (0.005mole) metal(II) chloride or Nitrate and ethanolic solution of Ligand (L) (0.01mole, 3.48gm) was refluxed for 5hrs. On cooling, the colored solid complexes were precipitated then filtered off, washed with ethanol three times then with diethyl ether and dried in vacuum<sup>[9]</sup>.

**Table 1 : Weight of metal salts that used in the prepared complexes**

Metal salt	Wt(g) metal salt	metal salt	Wt(g) metal salt
CoCl <sub>2</sub> .6H <sub>2</sub> O	1.19	CuCl <sub>2</sub> .2H <sub>2</sub> O	0.85
Co(NO <sub>3</sub> ) <sub>2</sub> . 6H <sub>2</sub> O	1.45	ZnCl <sub>2</sub>	0.68
NiCl <sub>2</sub> . 6H <sub>2</sub> O	1.18	CdCl <sub>2</sub>	0.91
Ni(NO <sub>3</sub> ) <sub>2</sub> .6H <sub>2</sub> O	1.45	CaCl <sub>2</sub>	0.55

### 4. RESULTS AND DISCUSSION

The synthesized complexes were solids and stable in air at room temperature. They were insoluble in water, but soluble in organic solvents such as dimethyl formamide. The molar conductance values are too low. This decrease in conductance indicates that there is no dissociation of the complexes in DMF, and confirming the non-electrolytic nature of these complexes<sup>[10]</sup>. The analytical and physical data of complexes are given in (table2).

**Table 2 : Characterization, analytical, molar conductance data of the complexes**

NO.	Formula	Molecular Weight	Colour	Yield	m.p (°C)	$\Omega$ Ohm <sup>-1</sup> . cm <sup>2</sup> . mol <sup>-1</sup>	calculate ( Found)%			
							%C	%H	%N	%M
1	[Co(L) <sub>2</sub> ]	753	gray	67	266-268	12	66.93 (66.54)	5.04 (4.67)	7.43 (7.11)	7.83 (7.54)
2	[Co(L) <sub>2</sub> ]	735	Brownish Yellow	79	246-248	17	66.93 (66.49)	5.04 (5.31)	7.43 (7.30)	7.83 (7.63)
3	[Ni(L) <sub>2</sub> ]	752.6	Brown	73	>300	22	66.96 (66.73)	5.04 (4.74)	7.44 (7.12)	7.78 (7.58)
4	[Ni(L) <sub>2</sub> ]	752.6	Green	75	>300	18	66.96 (66.67)	5.04 (5.28)	7.44 (7.09)	7.78 (7.49)
5	[Cu(L) <sub>2</sub> ]	757.5	gray	81	268*	32	66.53 (66.23)	5.01 (5.39)	7.39 (7.14)	8.38 (8.49)
6	[Zn(L) <sub>2</sub> ]	759	White Yellowish	74	286-288	27	66.37 (66.18)	5.00 (5.43)	7.37 (7.12)	8.60 (8.34)
7	[Cd(L) <sub>2</sub> ]	806	Light Yellow	80	>300	14	62.53 (62.11)	4.71 (4.54)	6.94 (6.67)	13.89 (13.67)
8	[Ca(L) <sub>2</sub> ]	734	White	69	288*	23	68.66 (68.43)	5.17 (5.45)	7.62 (7.43)	5.44 (5.21)

### Infrared Spectra

The infrared spectra of the free ligand showed a strong band at 1648cm<sup>-1</sup> assignable to (C=O) of the amide group<sup>[11]</sup>. Also, showed a band at the region 1607cm<sup>-1</sup> due to the azomethine group, and these bands in complexes were shifted to lower frequency indicating the coordination of oxygen atom of carbonyl and Nitrogen atom of Azomethine to the

metal. The IR spectrum of the Ligand showed a band at 3210 cm<sup>-1</sup> for (N-H) and another bands at 2954 cm<sup>-1</sup> due to C-H aliphatic groups and band at 3053 cm<sup>-1</sup> which is attributed to the aromatic (C-H) group. Also the spectrum of Ligand showed a band at 1029 cm<sup>-1</sup> due to N-N group which is shifted towards lower frequency on complexation<sup>[12,13]</sup>. In addition, the spectra of the Ligand showed a band at 3464 cm<sup>-1</sup> which is attributed to (O-H) and this band was disappeared in the spectra of the complexes because of the deprotonation that take takes place and because of the coordination of Oxygen with metal. Moreover, the band that observed at 1268 cm<sup>-1</sup> which is belonging to (C-O) stretching vibration undergoes a slight shifting toward low frequency at the region of 1206-1253 cm<sup>-1</sup> in comparison with free ligand bands.

The new bands at the range of 410-475 cm<sup>-1</sup> in the spectra of complexes are assigned to stretching frequencies of  $\nu$  (M-N) and the bands at the range of 471-595 cm<sup>-1</sup> are assigned to stretching frequencies of  $\nu$  (M-O)<sup>[14,15]</sup>. The band which is located at 1381 cm<sup>-1</sup> and 1384 cm<sup>-1</sup> in nitrate complexes (2,4) were attributed to ionic nitrate group<sup>[16]</sup>. The spectral data of the synthesized ligand and its complexes are given in (table3) .

**Table 3: Selected I.R. bands of the ligands and its complexes (cm)<sup>-1</sup>**

Compound No.	$\nu$ (O-H)	$\nu$ (N-H)	$\nu$ (C=N)	$\nu$ (N-N)	$\nu$ (C=O)	$\nu$ (C-O)	$\nu$ (M-N)	$\nu$ (M-O)
L	3464	3210	1607	1029	1648	1268	--	--
1	--	3205	1515	1007	1607	1253	416	563
2	--	3206	1521	1006	1616	1235	420	569
3	--	3211	1541	1010	1612	1206	475	595
4	--	3215	1539	1002	1595	1250	474	586
5	--	3214	1519	1009	1615	1228	471	578
6	--	3212	1548	960	1610	1209	410	592
7	--	3208	1516	1005	1616	1233	420	593
8	--	3207	1568	1000	1608	1211	474	560

### Electronic Spectra and Magnetic Measurement

The UV spectrum of Ligand(L) showed intense bands at 33222 cm<sup>-1</sup>, 28901 cm<sup>-1</sup> assigned to  $\pi \rightarrow \pi^*$  and  $n \rightarrow \pi^*$  transitions respectively. The electronic spectrum of Co(II) complexes shows absorption bands at (13315, 13404) cm<sup>-1</sup> due to  $\nu_1$  and (16260, 15576) cm<sup>-1</sup> due to  $\nu_2$  and (24449, 20618) cm<sup>-1</sup> due to  $\nu_3$ , which are assigned to following transition:  ${}^4T_{1g}(F) \rightarrow {}^4T_{2g}(F) (\nu_1)$ ,  ${}^4T_{1g}(F) \rightarrow {}^4A_{2g}(F) (\nu_2)$ ,  ${}^4T_{1g}(F) \rightarrow {}^4T_{1g}(P) (\nu_3)$ <sup>[17]</sup>.

and these values agreed with high spin octahedral configuration. The magnetic moment values for these complexes are (4.69, 4.76) B.M supports an octahedral around Co(II) ion<sup>[18]</sup>.

The Nickel (II) complexes exhibit electronic spectrum bands at (12195, 13071) cm<sup>-1</sup>, (14104, 17825) cm<sup>-1</sup> and (26041, 26737) cm<sup>-1</sup> respectively and assigned to transitions  ${}^3A_{2g}(F) \rightarrow {}^3T_{2g}(F) (\nu_1)$ ,  ${}^3A_{2g}(F) \rightarrow {}^3T_{1g}(F) (\nu_2)$ ,  ${}^3A_{2g}(F) \rightarrow {}^3T_{1g}(P) (\nu_3)$ .

The observed electronic spectrum suggests that these complexes as octahedral geometry around the Nickel(II) ion.

The octahedral geometry of Ni(II) ion in the complexes is confirmed with the measured magnetic moment values (2.89, 3.06) B.M<sup>[19]</sup>.

The electronic spectrum of Cu(II) complexes shows a broad band at 20576 cm<sup>-1</sup>, this band may arise from the  ${}^2E_g \rightarrow {}^2T_{2g}$  in octahedral field. the magnetic moment value of Cu(II) complex (2.07) B.M which may suggest an distorted octahedral structure<sup>[20]</sup>.

The complexes of Zn(II), Cd(II) and Ca(II) are diamagnetic and didn't show any d-d transition. These peaks are assigned to charge transfer transitions and suggest an octahedral structure around the metal ions<sup>[21-23]</sup>. The summary of Electronic transitions and Magnetic Moment for Ligand and Its Metal Complexes are given in (table4).

**Table (4): Electronic transition and Magnetic Moment for Ligand and Its Metal Complexes**

No.	Compound	Band cm <sup>-1</sup>	Assignment	M <sub>eff</sub> B.M
	C <sub>21</sub> H <sub>20</sub> N <sub>2</sub> O <sub>3</sub> (L)	33222 28901	$\pi \rightarrow \pi^*$ $n \rightarrow \pi^*$	--

1	[Co(L) <sub>2</sub> ]	13315	<sup>4</sup> T <sub>1g</sub> (F) → <sup>4</sup> T <sub>2g</sub> (F)	4.69
		16260	<sup>4</sup> T <sub>1g</sub> (F) → <sup>4</sup> A <sub>2g</sub> (F)	
		24449	<sup>4</sup> T <sub>1g</sub> (F) → <sup>4</sup> T <sub>1g</sub> (P)	
2	[Co(L) <sub>2</sub> ]	13404	<sup>4</sup> T <sub>1g</sub> (F) → <sup>4</sup> T <sub>2g</sub> (F)	4.76
		15576	<sup>4</sup> T <sub>1g</sub> (F) → <sup>4</sup> A <sub>2g</sub> (F)	
		20618	<sup>4</sup> T <sub>1g</sub> (F) → <sup>4</sup> T <sub>1g</sub> (P)	
3	[Ni(L) <sub>2</sub> ]	12195	<sup>3</sup> A <sub>2g</sub> (F) → <sup>3</sup> T <sub>2g</sub> (F)	2.89
		14104	<sup>3</sup> A <sub>2g</sub> (F) → <sup>3</sup> T <sub>1g</sub> (F)	
		26041	<sup>3</sup> A <sub>2g</sub> (F) → <sup>3</sup> T <sub>1g</sub> (P)	
4	[Ni(L) <sub>2</sub> ]	13071	<sup>3</sup> A <sub>2g</sub> (F) → <sup>3</sup> T <sub>2g</sub> (F)	3.06
		17825	<sup>3</sup> A <sub>2g</sub> (F) → <sup>3</sup> T <sub>1g</sub> (F)	
		26737	<sup>3</sup> A <sub>2g</sub> (F) → <sup>3</sup> T <sub>1g</sub> (P)	
5	[Cu(L) <sub>2</sub> ]	20576	<sup>2</sup> E <sub>g</sub> → <sup>2</sup> T <sub>2g</sub>	2.07
6	[Zn(L) <sub>2</sub> ]	29411	C.T	--
7	[Cd(L) <sub>2</sub> ]	24691	C.T	--
8	[Ca(L) <sub>2</sub> ]	26595	C.T	--

### <sup>1</sup>H.NMR & <sup>13</sup>C.NMR Spectra

The <sup>1</sup>H.NMR spectra of the ligand showed a sharp singlet of δ 8.2ppm due to the proton azomethine group was shifted down field in the spectra of the complex [Ca(L)<sub>2</sub>] to δ 8.5ppm. This deshielding due to the coordination of azomethine nitrogen and Metal ion.

The spectrum of the Ligand showed singlet signal at δ 11.1ppm due to amide proton, and this signal remained unaltered in the complexation which indicates non participation of NH in coordination to metal ion.

The <sup>1</sup>H.NMR of ligand exhibit a signal at δ 11.8ppm due to the OH proton and this signal in the spectra of complex disappeared because of the deprotonation that takes place and to the coordination between oxygen with the metal ion.

The aliphatic protons showed signals at δ 1.5 - 4.7ppm and the aromatic protons showed signals at δ 6.9 - 7.7ppm in free ligand. These signals values for the complex which indicate non participation in the coordination<sup>[24,25]</sup>.

<sup>13</sup>C.NMR Spectra of the ligand showed signals at range 104-157ppm due to the aromatic ring, and also showed signals at range 18-55ppm due to the aliphatic carbon atoms<sup>[26]</sup>. The signals in the ligand at 169ppm and 178ppm refer to azomethine carbon and carbonyl carbon respectively and these signals in the complex altered in coordination<sup>[27,28]</sup>.

### X-ray Powder Diffraction Analysis

The XRD patterns indicate a crystalline nature for complexes. Indexing of the diffraction patterns was performed using High Score Plus software Match<sup>[29]</sup> program, and their Miller indices (hkl) along with observed and calculated 2θ angle, d values, and relative intensities. The unit cell parameters were calculated based on the indexed data. The Powder XRD patterns of the compounds are completely different from those of the starting materials, demonstrating the formation of coordination compounds. It was found that the structures of these complexes were orthorhombic. Moreover, using diffraction data, the mean crystallite sizes of the complexes were determined according to Scherrer equation ( $D = 0.9 \lambda / (\beta \cos \theta)$ ), where λ X-ray wavelength (1.5406 Å), θ is Bragg diffraction angle, and β is the full width at half maximum of the diffraction peak<sup>[30]</sup>. The crystal data and structure refinement for complex 6 are given in (table 5).

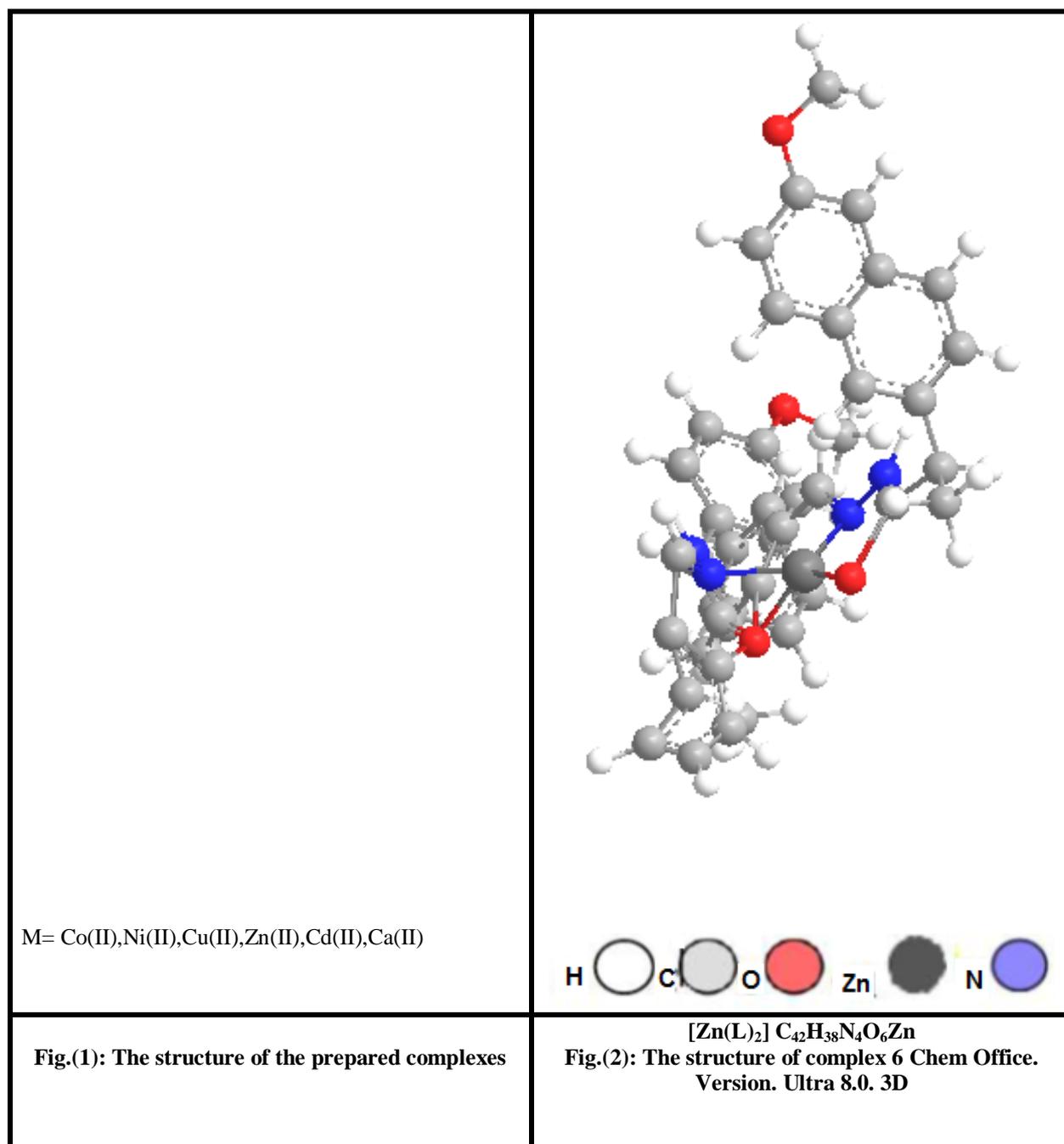
**Table 5: Crystal data and structure refinement for complex 6**

No	Complex	6
1	Molecular Formula	C <sub>42</sub> H <sub>38</sub> N <sub>4</sub> O <sub>6</sub> Zn
2	Molecular weight	759
3	Crystal system	Orthorhombic
4	Space group	P.4(63)
5	Unit cell parameters (Å)	a= 11.507, b= 10.340, c= 12.008

6	Cell Volume (A <sup>o</sup> )	697
7	Z	8
8	$\theta$ range , deg	2.03 – 38.35
9	Index ranges	$2 \leq h \leq 4$

### CONCLUSION

According to the results which are obtained from various physicochemical, spectral analysis as well as magnetic susceptibility and the measurement of XRD analysis, the synthesized complexes appear to have octahedral geometry, three coordination sites are occupied by tridentate ligand with O,N,O donor atoms sequence towards the metal ions.(Figure 1) is showing the structure of the prepared complexes generally and (Figure2) is showing the structure of complex  $[Zn(L)_2] C_{42}H_{38}N_4O_6Zn$ .



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