

# Synthesis and Characterization of some Transition Metals Complexes with new Ligand Azo Imidazole Derivative

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## ABSTRACT

AZO complexes synthesized by binding metal ions as Mn(II), Co(II), Ni(II) and Cu(II), with the ligand (L) [ethyl 4-((E)-(5-oxo-1-(((1E,2E)-3-phenylallylidene)amino)-2-thioxo-2,5-dihydro-1H-imidazol-4-yl)diazanyl)benzoate] was prepared from three steps, first step preparation Schiff base product of condensation reaction by reaction Thiosemicarbazide with cinnamaldehyde, second step preparation the Imidazole ring close by reaction ligand 1 with chloroethylacetate, finally preparation Azo ligand (Diazonium compound) by reaction ligand 2 with benzocaine. Characterized the ligands and complexes in their solid state using the elemental micro analysis (C.H.N.S), flame atomic absorption. UV-vis spectroscopy, FT-IR, magnetic susceptibility measurements, Mass spectrum, 1H-NMR spectra and electrical molar conductivity. These techniques produced comparable results with those obtained for the solid complexes. From the data of all techniques, octahedral geometry was proposed for Mn(II), Co(II), Ni(II) and Cu(II) complexes

**KEYWORDS:** Schiff base derivative; 2-thioxoimidazole derivative; cinnamaldehyde; Azo complexes.

## الخلاصة

تم تخليق معقدات الازو عن طريق ربط أيونات المعادن مثل المنغنيز الثنائي، الكوبالت الثنائي، النيكل الثنائي والنحاس الثنائي مع ليكند:

ethyl4-((E)-(5-oxo-1-(((1E,2E)-3-phenylallylidene)amino)-2-thioxo-2,5-dihydro-1H-)]  
[imidazol-4-yl)diazanyl)benzoate

تم تحضيره في ثلاثة خطوات: الخطوة الأولى تحضير قاعدة شيف لتفاعل التكثيف لمركب الثايوسمي كاربازيد مع مركب السينام الديهايد لينتج ليكند 1. الخطوة الثانية تحضير غلق حلقة الازو لتفاعل ناتج عن تفاعل ليكند 1 مع مركب كلورواثيل استيت لينتج ليكند 2. الخطوة الثالثة تحضير مركب الازوت (الدايزانيوم) ناتج من تفاعل ليكند 2 مع مركب البنزوكاين لينتج ليكند 3 (مركب الازو). تم تشخيص الليكند والمعدن في حالتها الصلبة بواسطة الامتصاص الذري اللهيبي، والتحليل الجزيئي للعناصر، مطيافية الأشعة فوق البنفسجية - المرئية، الأشعة تحت الحمراء، الرنين النووي المغناطيسي، قياسات الحساسية المغناطيسية، طيف الكتلة والتوصيل المولاري الكهربائي. نتجت هذه التقنيات نتائج مماثلة لتلك التي تم الحصول عليها للمعدن الصلبة من بيانات جميع التقنيات حيث تم اقتراح الشكل الهندسي هو ثماني السطوح لجميع المعقدات المنغنيز، الكوبالت، النيكل والنحاس.

## INTRODUCTION

Condensation reactions involving primary amines and ketones or aldehydes produced under particular circumstances produce Schiff bases. The Schiff bases is one of the most important organic compound and more interesting by the researchers due to contain (-C=N-)group [1]. The imidazole ring, which may be found in a range of natural products and pharmaceuticals, is one of the most frequent five-membered, nitrogen-containing heterocyclic scaffolds. Furthermore, imidazole-based heterocyclic compounds[2], play a crucial role in

biochemistry, have been playing a key role in the treatment of a different of illnesses, and innovative pharmaceutical derivatives are being developed with encouragement all over the world [3-4]. Because of the peculiar structural characteristic of imidazole scaffold with a beneficial electron-rich property showing a wide variety of biological activities, it is desired for imidazole groups to bind with diverse receptors and enzymes in biological systems through various weak interactions. An imidazole-containing medications have been widely used to

treat a range of diseases, including antibiotic infections, in recent years. [5-6], antifungal [7-8], anti-inflammatory [9-10], antiviral [11-12], anti-parasitic [13-14], anticancer [10-15], antihistamine [16-17] and enzyme inhibition [15-18]. As demonstrated in Table 1, Imidazole and its derivatives are employed in a variety of medical applications. Many Azo Imidazole derivatives, thiosemicarbazone ligand molecules, and their complexes have been widely employed and applied in biological, analytical, and pharmacological activities. Many Azo Imidazole compounds and their significant heterocyclic complexes have been studied for their antibacterial, antifungal, anti-inflammatory, and analgesic properties. [10]. These compounds variously used in the spectrophotometric measurement of metal ions. As a result, several of those reagents produce vibrant colors when they react with transition metal ions. [19] delivering probes that can also coordinate with rare earth ions, creating metal complexes with unique geometries [20]. In the present paper, the preparation of Imidazole derivative [ethyl 4-((E)-(5-oxo-1-(((1E,2E)-3-phenylallylidene)amino)-2-thioxo-2,5-dihydro-1H-imidazol-4-yl)diazenyl)benzoate] (L) was used as a ligand that provides two sites "potential donor" to produce some transition metal ions. (L) ligand and its complexes were completely characterized. We study Synthesis and Characterization of some Transition Metals Complexes with new ligand Azo imidazole derivative.

## MATERIALS AND METHOD

All the used chemicals were analytical annular and were highly pure.

### The Physical Measurements and Analysis

The melting points were measured using Gallen Kamp melting point instrument and were uncorrected. While, FT-IR spectrum was measured using Bruker spectrophotometer at the range (4000-400  $\text{cm}^{-1}$ ). UV-vis. spectrum recorded on (Shimadzu1900) spectrophotometer at room temperature. A 500 MHz Agilent Technologies spectrometer was used to record  $^1\text{H}$  NMR spectra in DMSO- $d_6$ , using a tetramethylsilane (TMS) as an internal standard. Furthermore, all these measurements collected at a concentration of  $10^{-3}\text{M}$  of the complex in absolute ethanol as a solvent. The elemental

(C.H.N.S) analysis was performed using an Eager 300 for EA1112 analyzer. The atomic absorption of the prepared complexes was measured using "Shimadzu Atomic Absorption 680 Flame" spectrophotometer. The mass spectrum of the ligand was obtained on an Electron-Impact (EI) mass spectroscopy. The mass spectrum of the metal complexes was obtained on an MSDCHEM. The conductivity measurements recorded on Philips pw-Digital Conductometer. All these measurements recorded in "DMSO" as a solvent using the concentration of ( $10^{-3}\text{M}$ ) at  $25^\circ\text{C}$ . The measurements of magnetic susceptibility collected at  $25^\circ\text{C}$  in solid state using Faraday's method at "Bruker BM6" instrument.

### Steps of preparation azo compound (L)

#### Step 1: Synthesis of Schiff base ligand 1

the solution of Thiosemicarbazide (1mmole) in Ethanol as a solvent (25 mL), (E)-3-phenylprop-2-enal (1mmole) was added with two drops of glacial acetic acid was added drop by drop at room temperature with constant stirring. After heating the mixture was refluxed for 8h, the mixture was poured onto ice water. It was filtered off, washed with ethanol and diethyl ether then dried to give compound 1. The yield of yellow product was (85 %), and the melting point was equal to ( $97-100^\circ\text{C}$ ) [21].

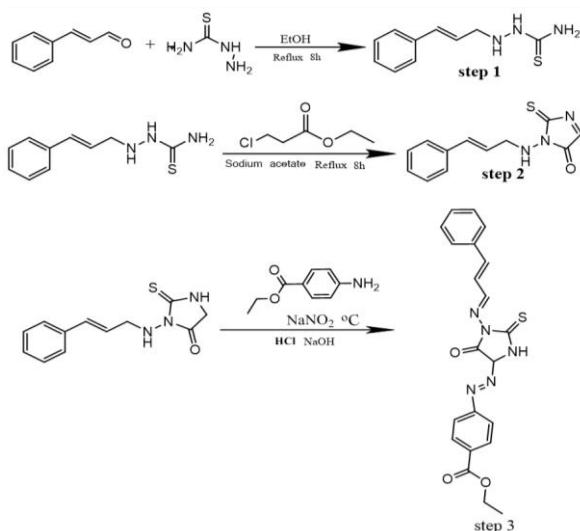
#### Step 2: Synthesis of Imidazole ligand 2

A mixture of compound 1 (1mmol) and ethyl chloroacetate (1mmol) was heated under reflux for 8 hours in ethanol solution (30 mL) containing anhydrous sodium acetate (1mmol). To get the product compound 2, the reaction mixture was cooled, and the resultant precipitate was filtered out and recrystallized from ethanol. The yield of yellow product was (85%), and the melting point was equal to ( $92-94^\circ\text{C}$ ) [22].

#### Step 3: (Synthesis of AZO ligand 3) (L)

(Ethyl 4-aminobenzoate (1mmol) in (10 mL) water and hydrochloric acid conc. (2.14 mL, 60.98 mmol) mixture with stirring, a clear solution was prepared). The temperature was maintained at ( $0-4^\circ\text{C}$ ). The aqueous solution of sodium nitrite  $\text{NaNO}_2$  (1mmol) dissolved in (5 mL) water; was drop wise slowly added to solution, while keeping the temperature below  $5^\circ\text{C}$  using ice bath, the mixture stirring for 1 h,

The pH was adjusted to 6-7 solution (1). The compound 2 (1mmol) was dissolved in 40% NaOH 10 mmol aqueous solution, cooled by ice bath to (0-4) °C solution (2). Gradually the last solution was mixed under cooling (solution1), the resulted mixture was stirred at (0-4) °C continually for 1h; the resulted precipitate was then filtered using acidification, washed with cold H<sub>2</sub>O for several times and dried using diethyl ether, the compound 3 (a brown color solid precipitate) was obtained with (85%) Yield and melting point(90-95 °C)[23].



**Scheme 1.** Steps preparation and synthesis of (L) ethyl 4-((E)-(5-oxo-1-(((1E,2E)-3-phenylallylidene)amino)-2-thioxo-2,5-dihydro-1H-imidazol-4-yl)diazenyl)benzoate.

**Table 1.** Chemical Formula for azo ligand and azo complexes.

Compound No.	Compound	Chemical Formula
L	C <sub>21</sub> H <sub>19</sub> N <sub>5</sub> O <sub>3</sub> S	C <sub>21</sub> H <sub>19</sub> N <sub>5</sub> O <sub>3</sub> S
1	[MnL(H <sub>2</sub> O)Cl <sub>2</sub> ]H <sub>2</sub> O	C <sub>21</sub> H <sub>21</sub> C <sub>12</sub> MnN <sub>5</sub> O <sub>4</sub> S
2	[CoL(H <sub>2</sub> O)Cl <sub>2</sub> ]H <sub>2</sub> O	C <sub>21</sub> H <sub>21</sub> C <sub>12</sub> CoN <sub>5</sub> O <sub>4</sub> S
3	[NiL(H <sub>2</sub> O)Cl <sub>2</sub> ]H <sub>2</sub> O	C <sub>21</sub> H <sub>21</sub> C <sub>12</sub> NiN <sub>5</sub> O <sub>4</sub> S
4	[CuL(H <sub>2</sub> O)Cl <sub>2</sub> ]H <sub>2</sub> O	C <sub>21</sub> H <sub>21</sub> C <sub>12</sub> CuN <sub>5</sub> O <sub>4</sub> S

### Synthesis of Complexes (1-4)

Ethanol solution of each following metal ion salts MnCl<sub>2</sub>.4H<sub>2</sub>O, CoCl<sub>2</sub>.6H<sub>2</sub>O, NiCl<sub>2</sub>.6H<sub>2</sub>O, and CuCl<sub>2</sub>.2H<sub>2</sub>O] with quantities of (1mmol) (0.198, 0.238, 0.237, 0.170) g was respectively added to ethanol solution (1mmole, 0.264 g) of (L) with continuous stirring. The mixture then heated with reflux for two hrs. Over this time, the formed precipitate was filtered, washed with water, recrystallized from the ethanol, and then dried using the oven at 50°C for 1 h. The physical properties of the synthesized compounds are shown in Table 2.

## RESULTS AND DISCUSSION

### Characterization of ligand (L) along its complexes achieved by

#### A) The Elemental Micro Analysis

physical and analytical data of ligand (L) and its metal complexes are shown in Table 2. These data are; consistent with the calculated values. The suggested molecular formula also supported by the subsequent spectra and the magnetic moment.

**Table 2.** Ligand (L) and its Metal Complexes Physical Data.

CompoundNo.	Color	M. p. °C	M.Wt g/mol	M:L inEtOH	%Elemental Analysis Found (Calc.)						Metal M%Found (Calc.)
					C	H	N	O	S	Cl	
(L)	Brown	90-95	421	-	59.84 (58.45)	4.54; (3.03)	16.62 (12.21)	11.39 (11.21)	6.61 (5.12)	-	-
1	Pale Brown	300d*	584.5	1:1	42.07 (43.07)	3.95 (4.81)	14.48 (12.14)	10.91 (11.21)	6.62 (5.51)	11.62 (12.51)	11.67 (10.67)
2	DarkGreen	300d*	588.2	1:1	41.86 (44.90)	3.94 (4.79)	14.40 (13.02)	10.87 (11.21)	6.59 (5.44)	10.59 (12.44)	10.06 (10.01)
3	Yellow	300d*	588.4	1:1	43.01 (43.90)	4.69 (4.42)	13.28 (12.60)	11.39 (11.21)	6.89 (5.77)	11.89 (12.77)	9.02 (12.25)
4	Green	300d*	589.1	1:1	41.90 (40.90)	3.48 (4.40)	13.09 (12.44)	11.39 (11.21)	6.86 (5.68)	11.86 (12.68)	10.66 (9.04)

d\* = decomposition

#### B) <sup>1</sup>H-NMR Spectra

In Figure 1, H-NMR spectrum of AZO compound (L), in DMSO-d<sub>6</sub> showed peak at 1.35 ppm which could be related to Methyl groups that appeared at range 1.26-2.5 ppm[24].

Aromatic ring appeared as multiple peaks at range (6.5-8.5) ppm. Peak attributed to (-SH-C-) appeared at 3.89 ppm which that noticed at range (3-3.6) ppm. The peak appeared as doublet for imidazole ring are observed at (4.3) ppm and

could be attributed to (CH-CH), and (N-CH) which generally appeared at range (4-4.9). (CH=CH) appeared at 5.97 ppm which generally appeared at range (4.5-6.5). Peaks appeared at 11.96 ppm, which attributed to N-H, generally appeared at range (10.5-12) ppm. To confirm the chemical structure and to check on the purity of compounds, the NMR technique is required approach in the synthetic chemistry. The spectrum revealed peaks at 2.51 and 3.35 ppm related to the DMSO-d<sub>6</sub> solvent and the traces of water molecules in the solvent, respectively [24].

### C) Infrared Spectroscopic Study

Figure 2 and 3, FT-IR spectra provided good information about complex behavior of the ligand with varied metal ions. The IR spectrum of (L), show an absorption band at 3234, 3036-3057, 1710, 1647, 1447-1518 and 1367 cm<sup>-1</sup> corresponding to the NH, C-H and C=O ester, C=O imidazole, N=N and C=S groups respectively. The IR spectrum of compounds (1-4), show an absorption band at 3035-3046, 1702-1707, 1673-1678, 1604-1608, 1446-1518 and 1365-1367 cm<sup>-1</sup> corresponding to the C-H aromatic, C=O ester, C=O imidazole, C=C aromatic, N=N and C=S groups respectively.

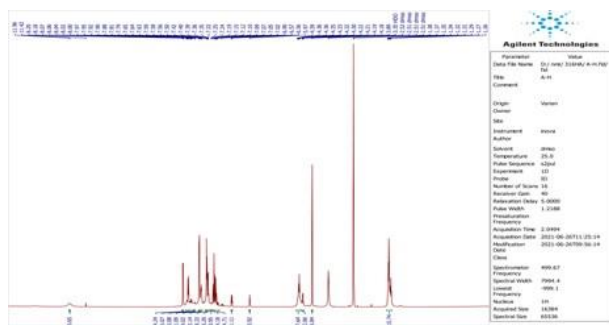


Figure 1. The <sup>1</sup>H-NMR Spectrum of the Ligand (L).

Moreover, these observations indicated by ( $\nu_{M-O}$ ,  $\nu_{M-N}$  and  $\nu_{M-Cl}$ ) appearance at (599-618) cm<sup>-1</sup>, (500-506) cm<sup>-1</sup> and (282-286) cm<sup>-1</sup> respectively [25].

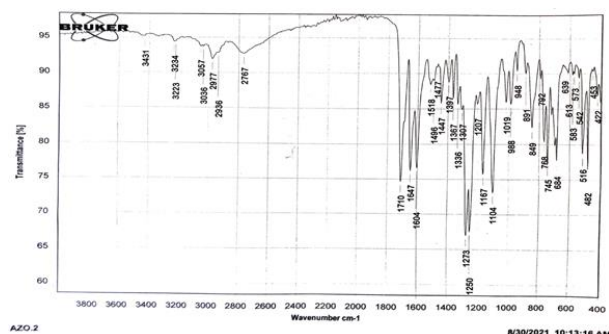


Figure 2. The FT-IR spectrum of ethyl 4-((E)-(5-oxo-1-(((1E,2E)-3-phenylallylidene)amino)-2-thioxo-2,5-dihydro-1H-imidazol-4-yl)diazenyl)benzoate (L).

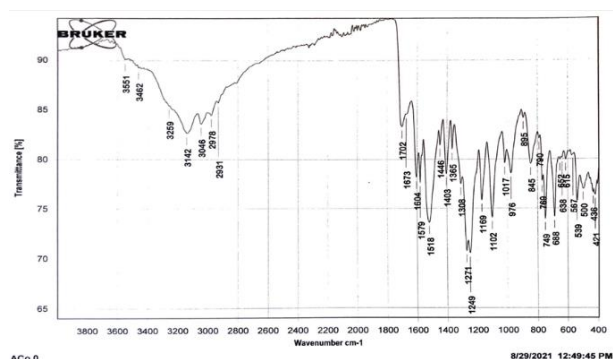


Figure 3. FT-IR Spectra of [Co(L)(H<sub>2</sub>O)Cl<sub>2</sub>].

### D) Mass spectrum of L and complexes

The electrospray (+) mass spectrum of L, as shown in Figures 4 and 5, shows no peak may assign to the ion around m/z=421 amu Figure 4. However, Peaks seen at m/z= 244, 218, 203, 164, 115, 114 and 46 amu assigned to [C<sub>12</sub>H<sub>10</sub>N<sub>3</sub>OS]<sup>+</sup>, [C<sub>11</sub>H<sub>11</sub>N<sub>2</sub>O<sub>3</sub>]<sup>+</sup>, [C<sub>10</sub>H<sub>10</sub>N<sub>3</sub>S]<sup>+</sup>, [C<sub>9</sub>H<sub>9</sub>NO<sub>2</sub>]<sup>+</sup>, C<sub>9</sub>H<sub>10</sub>, [C<sub>3</sub>H<sub>2</sub>N<sub>2</sub>OS]<sup>+</sup> and [CH<sub>2</sub>O<sub>2</sub>]<sup>+</sup> respectively [23].

Table 3. Ligand (L) and its Metal Complexes Characteristic Stretching Vibration Frequencies (cm<sup>-1</sup>) Located in FTIR.

Compound No.	$\nu_{C=O}$ imidazole	$\nu_{C=N}$	$\nu_{N=N}$	M-O	M-N	M-Cl
(L)	1647	1604	1447	-	-	-
1	1673	1576	1446	599	506	286
2	1673	1579	1446	615	500	284
3	1670	1677	1446	618	506	284
4	1666	1579	1446	603	506	282

As shown in Table 3, the electrospray (+) mass spectrum of [Co(L)(H<sub>2</sub>O)Cl<sub>2</sub>]<sup>+</sup> Peak seen at m/z=588 Figure 5, successive fragmentation pattern with their relative abundance of [Co(L)(H<sub>2</sub>O)Cl<sub>2</sub>] are depicted in Scheme 3.

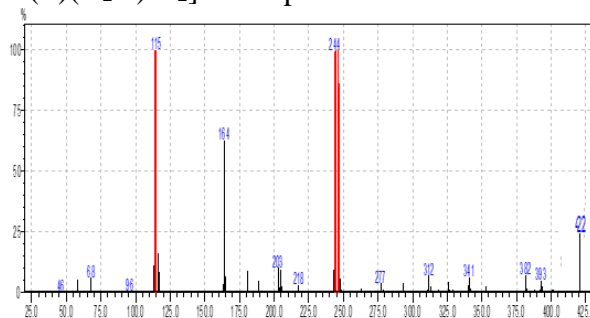
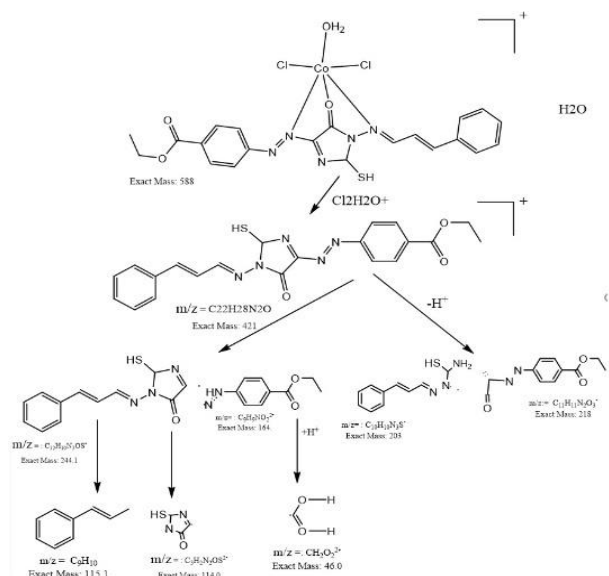


Figure 4. The electrospray (+) mass spectrum of L.





Figure 5. The electrospray (+) mass spectrum of  $[\text{Co}(\text{L})(\text{H}_2\text{O})\text{Cl}_2]$ .



Scheme 3. The electrospray (+) mass spectrum of  $[\text{Co}(\text{L})(\text{H}_2\text{O})\text{Cl}_2]$ .

### E) Studying of Electronic Spectra, Magnetic Moment, Conductance

In Figures 6 and 7, the UV spectrum of ligand (L) mostly shows three intense absorption peaks at  $37543\text{ cm}^{-1}$ ,  $34482\text{ cm}^{-1}$  and  $33456\text{ cm}^{-1}$  which belong to  $\pi \rightarrow \pi^*$ , and  $n \rightarrow \pi^*$  and C.T, respectively, (See Table 4, Figures 6 and 7) [26,27]. The ligand metal complexes in electronic spectra were measured in dimethyl form (DMSO) as a solvent at the range (200-800)nm.

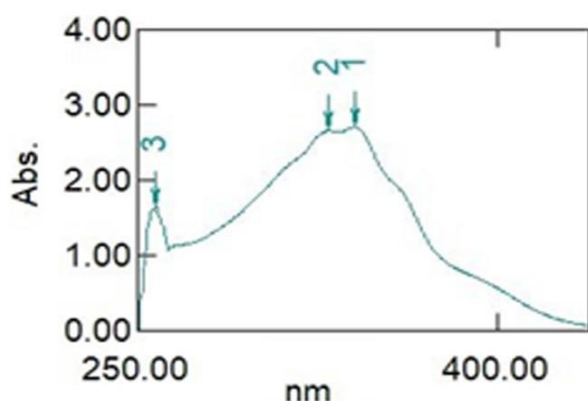


Figure 6. Electronic spectrum of (L) in DMSO solvent.

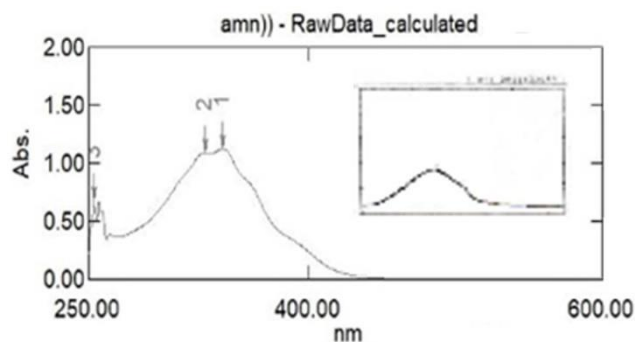


Figure 7. Electronic spectrum of  $[\text{Co}(\text{L})(\text{H}_2\text{O})\text{Cl}_2]$  in DMSO solvent.

**$[\text{MnL}(\text{H}_2\text{O})\text{Cl}_2]\text{H}_2\text{O}$  (1):** The electronic spectrum of Mn(II) complex showed absorption bands at  $(16891, 27173)\text{ cm}^{-1}$  due to the transitions  ${}^6\text{A}_{1g} \rightarrow {}^4\text{T}_{1g}(\text{G})$ ,  ${}^6\text{A}_{1g} \rightarrow {}^4\text{T}_{2g}(\text{D})$ , respectively, indicating octahedral geometry [27]. The measured magnetic moment  $\mu_{\text{eff}}$  of Mn(II) complex is (5.18 B.M) which corresponds to five unpaired electronic indicates high-spin octahedral environment [28], (Table 4). Measuring the conductivity in (DMSO) demonstrated that this complex has no conductivity and showed that this complex is [1:1] metal to ligand; Scheme3.

**$[\text{Co}(\text{H}_2\text{O})\text{Cl}_2]\text{H}_2\text{O}$  (2):** The electronic spectrum of Co(II) complex showed three peaks at  $(14858, 16528, \text{ and } 30303)\text{ cm}^{-1}$  assignable to  ${}^4\text{T}_{1g} \rightarrow {}^4\text{A}_{2g}(\text{F})$ ,  ${}^4\text{T}_{1g} \rightarrow {}^4\text{T}_{1g}(\text{P})$ , and C.T transitions, respectively[13], (See Table 4, Figure 5) for an octahedral geometry [28]. Moreover, the magnetic moment  $\mu_{\text{eff}}$  of this complex in the sold state (4.50 B.M) suggests a low-spin octahedral geometry[29]. Furthermore, measuring the conductivity in (DMSO) demonstrated that this complex has a no conductivity showed that this complex is [1:1] metal to ligand; Scheme3.

**$[\text{NiL}(\text{H}_2\text{O})\text{Cl}_2]\text{H}_2\text{O}$  (3):** The electronic spectra of Ni(II) complex appears at  $(32786)\text{ cm}^{-1}$  and this attributed to the charge transfer transition (C.T). In addition, the other peak which appears at  $(27397\text{ cm}^{-1})$  can be attributed to  $({}^3\text{T}_1 \rightarrow {}^1\text{T}_2)$  electronic transition, that suggests octahedral geometry [5] around Ni ion. The magnetic measurement  $\mu_{\text{eff}}$  of the solid complex (3.21 B.M) demonstrated two unpaired electrons for Ni(II) ion which also suggests a octahedral geometry for Ni(II) complex [30]. Measuring the no conductivity in (DMF) shows that this

complex is [1:1] metal to ligand, Table 4, Scheme3.

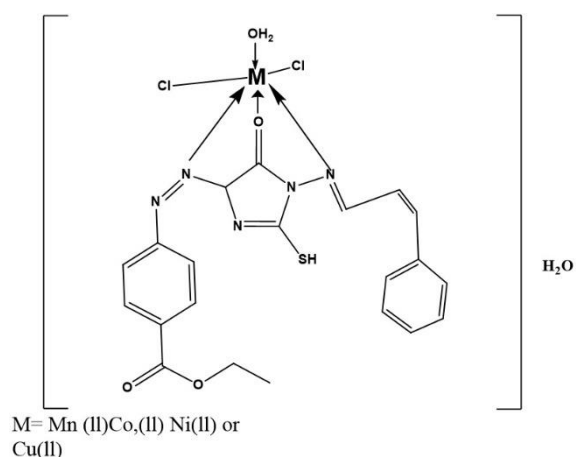
**Table 4.** Ligand (L) and its Metal Complexes (1-4)  
Electronic Spectra, Conductance in (DMF) and Magnetic Moment (B.M)

Compound No.	Bands $\text{cm}^{-1}$	Assignment	Molar Cond. $\text{ohm}^{-1} \cdot \text{cm}^2 \cdot \text{mol}^{-1}$	$\mu_{\text{eff}}$ B.M	Suggested Structure
(L)	37543 34482 33456	$\pi \rightarrow \pi^*$ $n \rightarrow \pi^*$ C.T	-	-	-
1	16891 27173	${}^6\text{A}_{1g} \rightarrow {}^4\text{T}_{1g(\text{g})}$ ${}^6\text{A}_{1g} \rightarrow {}^4\text{T}_{2g(\text{g})}$ C.T	19	5.18	Octahedral
2	14858 16528 30303	${}^4\text{T}_{1g} \rightarrow {}^4\text{A}_{2g(\text{f})}$ ${}^4\text{T}_{1g} \rightarrow {}^4\text{T}_{1g(\text{p})}$ C.T	16	4.50	Octahedral
3	27397 32786	${}^4\text{T}_{1g} \rightarrow {}^4\text{A}_{2g(\text{f})}$ ${}^4\text{T}_{1g} \rightarrow {}^4\text{T}_{1g(\text{p})}$ C.T	7	3.21	Octahedral
4	12658 32051	${}^4\text{T}_{1g} \rightarrow {}^4\text{A}_{2g(\text{f})}$ ${}^4\text{T}_{1g} \rightarrow {}^4\text{T}_{1g(\text{p})}$ C.T	6	1.87	Octahedral

**[CuL(H<sub>2</sub>O)Cl<sub>2</sub>]H<sub>2</sub>O (4):** The electronic spectra of Co(II) complex demonstrated a peak at (3205)  $\text{cm}^{-1}$  which refers to (C.T) transition, while the peak that appears at (12658)  $\text{cm}^{-1}$  is attributed to ( ${}^2\text{T} \rightarrow {}^2\text{E}$ ) electronic transition. However, these transitions indicate that the complex has a octahedral geometry. Moreover, the value of measuring the magnetic moment is (1.87 B.M), which corresponds to one unpaired electron. Furthermore, measuring the conductivity in (DMSO) demonstrated that the complex has no conductivity [1:1] metal to ligand, Table 4, Scheme3.

### Suggested Structure for Complexes (1-4)

According to the obtained results from the elemental and spectral analyses, besides measuring the magnetic moment and conductivity, the mentioned above compounds suggested structures can be explained as shown in Scheme 4.



**Scheme 4.** Suggestion of metal complexes.

## CONCLUSIONS

The current study suggested that the complexes of Mn(II) , Co(II) , Ni(II) , Cu(II) and ions with new imidazole azo ligand [ethyl 4-((E)-(5-oxo-1-(((1E,2E)-3-phenylallylidene)amino)-2-thioxo-2,5-dihydro-1H-imidazol-4-yl)diazenyl)benzoate] synthesis and characterization of all above complexes are octahedral and no conductivity. Future we can use these complexes in biological activity and pharmaceuticals.

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