مجلة كلية التربية ـ جامعة سرت
المجلد (1) العدد(3) يناير 2023

Mixed Ligand Complexes: Synthesis and Characterization Studies

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ABSTRACT

The synthesis and structural characterization of mixed ligand complexes of Schiff base derived from the condensation of 2-hydroxyacetophenone and 2,4-dinitrophenylhydrazine (HL1) and 2-aminophenol(L2) are reported. The ligands and their mixed ligand complexes were characterized on the bases of their IR, ^{1}HNMR, electronic, elemental analyses, magnetic moment and molar conductance measureents. The mixed ligand complexes under investigation were formed in the 1:1:1 [M:L1:L2] ratio as found from the elemental analysis data and found to have the formulae $[L1ML2(H_2O)_2]$, where M = Mn(II), Co(II) and Ni(II) ions and $[L1FeL2(ONO_2)(H_2O)]$, Meanwhile, the 1:1:2[M:L1:L2] ratio is for $[L1Cr(L2)_2]$ complex, The molar conductance results revealed that the complexes are non-electrolytes. IR spectra showed that the ligands are coordinated to the metal ions in a bidentate manner with donor sites. Magnetic moments are used to confirm the coordinating capacity of the ligands and the geometrical structure of their complexes are found to be an octahedral.

Keywords: Schiff base, mixed ligand complexes and physiochemical characterization.

متراكبات الليجند المختلطة:دراسات تخليقية وتشخيصية

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الملخص:

لقد تم تخليق وتشخيص تركيبي لمتراكبات ليجند المختلطة لقاعدة **شف مشتقة من تكثيف 2**-هيدروكسي أسيتوفينون و 2،4- ثنائي نيتروفينيل هيدرازين(HL1) مع 2-أمينوفينول. حيث شخصت الليجندات ومتراكباتها باستخدام تحاليل الاشعة تحت الحمراء، فوق البنفسجية، الرنين المغناطيسي النووي البروتوني، التحليل العنصري ،القياسات المغناطيسية والمولارية. تكونت المتراكبات بنسب 1:1:1(فلز:ليجند:ليجند) حسب النتائج العملية المتحصل عليها، وأخذت الصيغ الكيميائية **التالية**: $[L1ML2(H_2O)_2]$ حيث M تمثل Mn(II), Co(II) and Ni(II) و $[L1FeL2(ONO_2)(H_2O)]$. بينما النسبة 1:1:2(فلز:ليجند:ليجند) لمتراكب $[L1Cr(L2)_2]$. أظهرت القياسات المولارية وجود ظاهرة غير اليكتروليتية للمتراكبات المحضرة. كما بينت نتائج الأشعة تحت الحمراء أن الليجندات تتناسق مع أيونات الفلزات لصورة ثنائية المنح. واستخدمت القياسات المغناطيسية لتأكيد الأشكال الهندسية للمتراكبات والتي هي ذات شكل ثماني السطوح.

الكلمات المفتاحية: *قاعدة شف، متراكبات ليجند المختلطة وتشخيص فيزيوكيميائي.*

INTRODUCTION

Schiff bases are organic compounds, considered to be a subclass of imines, which may be secondary aldimines or ketimines depending on the nature of the parent carbonyl compounds (El-ajaily *et. al*., 2018).(1) Mixed ligand complexes of some transition metals were synthesized from a Schiff base (L^1) obtained by the condensation reaction of oxamide and furfural(HL1) as primary ligand and 2,2′-bipyridine (L^2) as secondary ligand. The ligands and their mixed liand complexes were studied using various spectroscopic methods (Abd El-Halim *et. al*., 2017).(2) The hexacoordinated mixed ligands of Co(II), Ni(II), Cu(II) and Zn(II) complexes of the type $[MLX]Cl_2$ where X=1,10-phenonthroline and L represents Schiff base resulted from the reaction of thiophene-2-carboxaldehyde with *o*-phenylenediam- ine. All the compounds were characterized by elemental analysis, molar conductance, magnetic susceptibility, infrared, electronic absorption, proton magnetic resonance and mass spectral studies. An octahedral geometry has been proposed for all these complexes. (Vairalakshmi, *et.al*.,2018).(3) Four copper(II) mixed ligand complexes of the coumarin derivative (A1=7-hydroxy-10,11-dihydroin de no [5,4-c]chromen-6(9H)-one, A2=2-bromo-7-hydroxy-10,11-dihydroindeno[5,4-c]chromen-6(9H)-one, A3=7-hydroxy-4-methoxy-10,11-dihydroindeno[5,4-c]chromen-6(9H)-one, and A4=5-hydroxy-8,9-dihydrobenzo[f] in deno[5,4-c]chromen-4(7H)-one) and 1,10-Phenanthroline have been synthesized. The structural interpretations were confirmed from elemental analyses, magnetic susceptibility and mass spectra, as well as from IR spectral studies. From the analytical, spectroscopic and thermal data, the stoichiometry of the mentioned complexes was found to be 1:1:1 (coumarin ligand: copper metal:1,10-Phenanthroline) (Kharadi *et. al*.,2012).(4) The mixed ligand complexes using clioquinol (5-chloro-7-iodo-8- hydroxyquinoline) and 1,10-phenanthroline as ligands were synthesized and characterized (Kharadi *et. al*., 2010).(5) The mixed-ligand complexes of various metal(II) with 5-chloro-7-iodo-8-hydroxyquinoline(Clioquinol) and 5-(methoxymethyl-8-quinolinol) (MMQ) were prepared (Chauhan *et. al.,* 2010).(6) The structure of these complexes was investigated using several physicochemical tools, elemental analyses, thermal studies, magnetic moment and reflectance spectral studies revealed the existence of an octahedral geometry for all the complexes.This study aims to synthesize, characterization of some mixed ligand complexes with Schiff base as primary ligand(HL1)

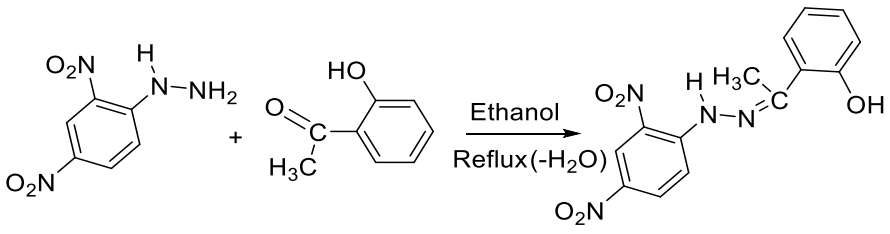
and 2-aminophenol(L2) with Cr(III),Mn(II), Fe(III), Co(II) and Ni(II) ions(Reiss *et. al*., 2021)[7]. A mononuclear octahedral mixed-ligand cobalt(II) complex $[Co(H_2L)(PhCOO)_2]$ (1) has been prepared by using H_2L(N,N0-dimethyl-N,N0-bi(2-hydroxy-3,5-di methyl benzyl)ethylenediamine) as a facially coordinating tetradentate ligand with a N_2O_2 donor center along with sodium benzoate as an ancillary ligand. Complex 1 has been characterized by a single-crystal X-ray diffraction study, as well as by other spectroscopic tools. The complex crystallizes in the monoclinic space group C2 with a = 31.73(3) Å, b= 7.868(3) Å, c = 19.131(15) Å, and β = 125.25(3)°. The single crystal X-ray diffraction study shows that in the mononuclear cobalt (II) complex $[Co(H_2L)(PhCOO)_2]$, the metal center adopts an octahedralenvironment (Mandal,*et. al.*,2022)[8].

CHEMICALS AND REAGENTS

All solvents and chemicals used in this investigation were of analar Grade (Aldrich, BDH). They include, 2-aminophenol, 2-hydroxyacetophenone, 2,4-dinitrophnylhy-drazine and some metal salts; $CrCl_3.6H_2O$, $Fe(NO_3)_3.6H_2O$, $CoCl_2.6H_2O$, $NiCl_2.6H_2O$, $MnCl_2.4H_2O$, DMF, glacial acetic acid, absolute ethanol and ether. These solvents were either spectroscopically pure.

Synthesis of the Schiff base

The Schiff base was synthesized by adding (6.807g, 0.05 mmole) of 2-hydroxyacetophenone dropwise to 2,4-dinitrophnylhydrazine (9.907 g, 0.05 mmol) in 50 mL of absolute ethanol. The reaction mixture was refluxed for 3 hours. Then the product was cooled at room temperature, filtered off and recrystallized from ethanol and dried under vacuum to get a yellow precipitate. The Schiff base formation is shown in **Scheme 1**.



Scheme 1: Synthesis Schiff base

Synthesis of mixed ligand complexes

To 30 mL absolute ethanol solution of the Schiff base (15.81g, 0.05mmol), 30mL absolute ethanol solution (13.322g, 0.1mmol) of $CrCl_3.6H_2O$, (9.896 g, 0.05 mmol) $MnCl_2.4H_2O$, $Fe(NO_3)_3.6H_2O$ (17.50g,0.05mmol), $CoCl_2.6H_2O$ (11.90g, 0.05 mmol) and $NiCl_2.6H_2O$ (11.885g, 0.05 mmol) were added, after 10 minutes 30 mLof the same solvent of 2-aminophenol (5.45g, 0.05 mmol) was added dropwise as a secondary ligand (L2) and the resulting mixtures were refluxed for about 3 hours. The obtained precipitates were filtered, washed with ethanol and dried under vacuum on anhydrous $CaCl_2$.

RESULTS AND DISCUSSION

Microanalysis and molar conductance measurements

The physical properties and elemental analysis data of the Schiff-base and mixed ligand complexes are summarized in Table 1, where, the results confirm the proposed composition. The synthesized mixed ligand complexes were formed in 1:1:2 or 1:1:1 (L1:M:L2) ratio. The obtained molar conductance values of the complexes in DMF solvent lie in the range of 09-18 ohm^{-1} cm^2 mol^{-1} indicating their non-electrolytic behavior (*Qamar et. al.*, 2019)).[9]

Table (1) Some physical properties and elemental amylases of Schiff base and mixed ligand complexex

Compound/ mixed ligand	Color	M.wt.	M.P.°C	**%**Calc**. (Found)**			Λ_m Ω^{-1} $cm^2.mol^{-1}$	μs (BM)
				C%	H%	N%		
Schiff base (HL1)	Orange	316.27	210	53.17 **(52.09)**	3.82 **(3.62)**	17.72 **(17.11)**	-	-
$[Cr(L1)(L2)_2]$	Light brown	546.86	221	53.52 **(52.93)**	3.97 **(4.68)**	14.40 **(14.04)**	17	3.78
$[Mn(L1)(L2)(H_2O)_2]$	Dark orange	514	219	46.70 **(47.11)**	4.12 **(4.64)**	13.62 **(13.95)**	13	5.65
$[Fe(L1)(L2)(ONO_2)(H_2O)]$	Brown	559.25	214	42.95 **(42.65)**	3.42 **(4.10)**	15.03 **(14.93)**	09	5.15
$[Co(L1)(L2)(H_2O)_2]$	Brown	518.35	217	36.34 **(36.07)**	4.08 **(3.59)**	13.51 **(14.22)**	18	3.87
$[Ni(L1)(L2)(H_2O)_2]$	Dark brown	518.11	225	46.36 **(46.71)**	4.09 **(5.30)**	13.52 **(13.45)**	11	2.81

Mass spectrum of Schiff base

The mass spectral data of the Schiff base ligand was demonstrated in Figure 1. Molecular ion showed peaks, which were in good agreement with the expected Values(Derebe *et. al.,* 2002).[10] The mass spectrum gives a base peak at 316 m/z.

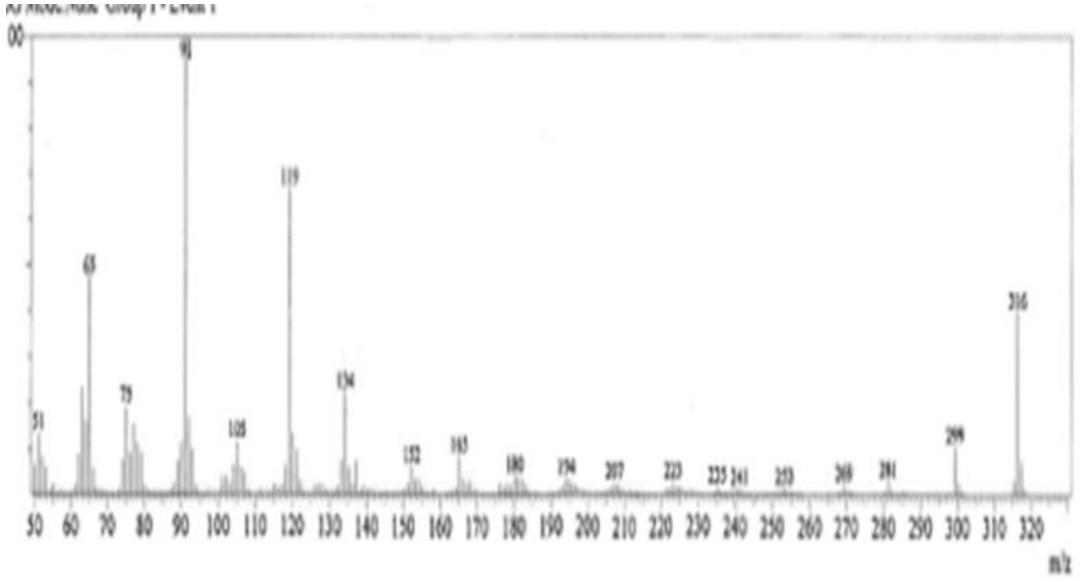


Figure. 1: Mass spectrum of Schiff base

^{1}HNMR spectrum of Schiff base (HL1)

The Schiff base(HL1) shows five signals (figure 2) at 11.09, 10.72, 6.88-8.90, 3.321 and 2.499 ppm, downfield of TMS, assignable to the protons of OH (phenyl ring), NH, phenyl ring, DMSO and, CH_3, respectively(Shaker *et. al*., 2010, Shaker, *et. al*., 2016., Shaker *et. al*., 2015)).[11-13]

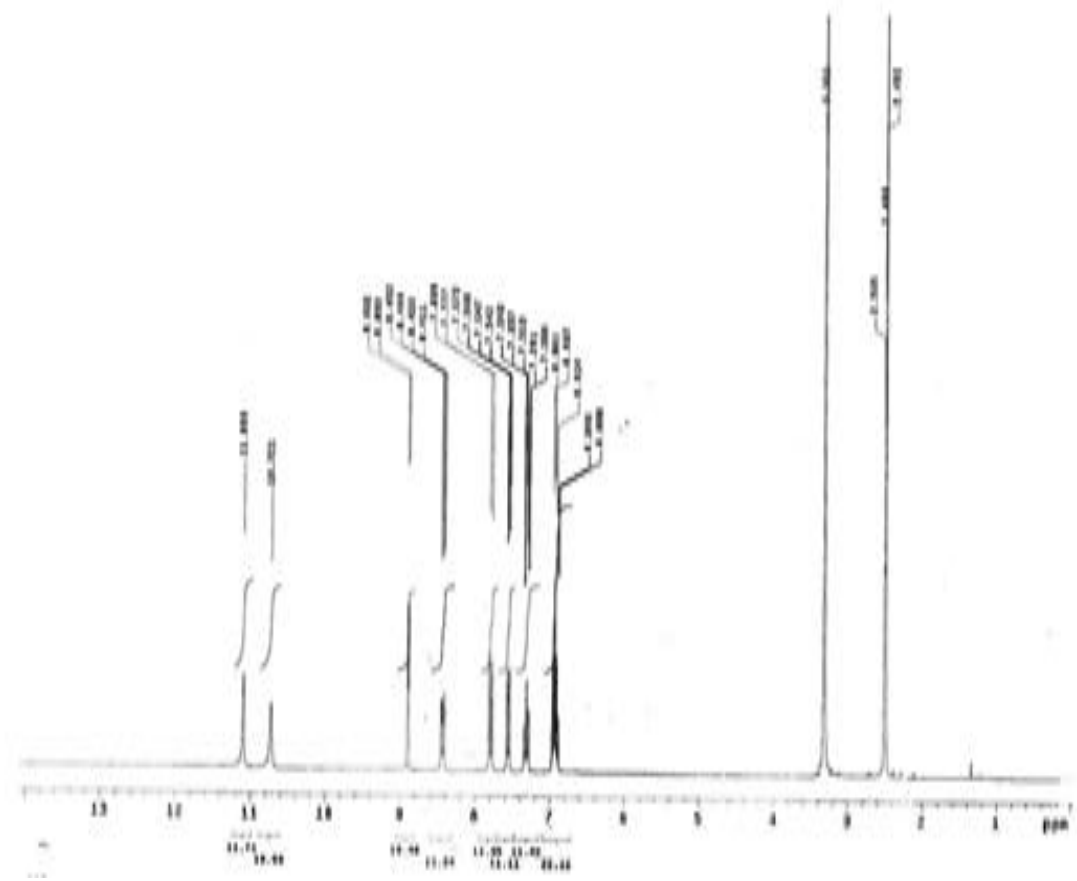


Figure 2. ^{1}H NMR spectrum of the Schiff base (HL1)

Infrared spectra

Infrared spectra FT-IR spectrum of the Schiff bases Figs.(3-8), showed bands due to υ(OH), υ(NH) and υ(C=N) respectively which absorbed at 3393 cm^{-1}, 3297 cm^{-1} and 1608 cm^{-1}(Ommenya *et. al*., 2020, Khalifa *et. al*., 2016).[14,15] The IR spectra of mixed ligand complexes: a strong sharp absorption band at 1608 cm^{-1} in spectrum of the Schiff base may be assigned to the υ(C=N) stretching, in spectra complexes, this band is shifted the range between 1612-1620 cm^{-1}(Hamil *et. al*., 2011, Al-Nuzal *et. al*., 2016).[16,17] upon complexation with metal, which may be attributed to the coordination of the imine nitrogen to the metal center(Sumrra *et. al*., 2014)[18]. The Schiff base shows band at 3393 cm^{-1} due to υ(OH) stretching from phenolic group which disappears in the spectra complexes indicating the deprotonation of the Schiff base upon complexation, shifted the range between 3300-3404 cm^{-1}(Abou Melha *et. al*., 2018, Benerjee and Chattopadhyay, 2019).[19,20] Also, new absorption bands in the 437-455 cm^{-1} region are considered to be due to metal-nitrogen υ(M-N) vibrations whilst those accruing around 456- 507cm^{-1} are through to a wise from metal-oxygen (M-O) vibration(Sabik *et. al*., 2012).[21] The spectrum of Fe(III) mixed ligand complex exhibits a band at 1422cm^{-1} which is not exist in the free Schiff base, this may be due to the appearance of nitrate ion group that bonded to Fe(III) ion through nitrogen atom(Ahmad *et. al*., 2017)[22].

Table 2: Infrared spectral data(cm^{-1}) and electronic spectral data (cm^{-1}) of the Schiff base and mixed ligand complexes

Ligand/mixed ligand complexes	IR spectra (cm^{-1})					UV-Vis (cm^{-1})
	νOH	**νNH**	**νC=N**	**νM-O**	**νM-N**	
$C_{14}H_{12}N_4O_5$ (HL1)	3393	3297	1608	---	---	$\pi\rightarrow\pi^*$, $n\rightarrow\pi^*$
[Cr(L1)(L2)$_2$]	3303	3249	1616	456	437	$^4A_{2g}(F)\rightarrow {}^4T_{2g}(F)$ $^4A_{2g}(F)\rightarrow {}^4T_{1g}(F)$
[Mn(L1)(L2)(H_2O)$_2$]	3404	3300	1612	501	455	$^6A_{1g}\rightarrow {}^4T_{1g}(P)$, CT
[Fe(L1)(L2)(ONO_2)(H_2O)]	3300	3100	1615	507	453	$^3A_{2g}(F)\rightarrow {}^3T_{1g}(F)$, $^3A_{2g}(F)\rightarrow {}^3T_{1g}(P)$, CT
[Co(L1)(L2)(H_2O)$_2$]	3300	3193	1628	501	453	$^4A_{2g}(F)\rightarrow {}^4T_{1g}(F)$, $^4A_2g(F)\rightarrow {}^4T_1g(P)$
[Ni(L1)(L2)(H_2O)$_2$]	3300	3103	1618	501	453	$^3A_{2g}(F) \rightarrow {}^3T_{1g}(F)$, $^3A_{2g}(F) \rightarrow {}^3T_{1g}(p)$

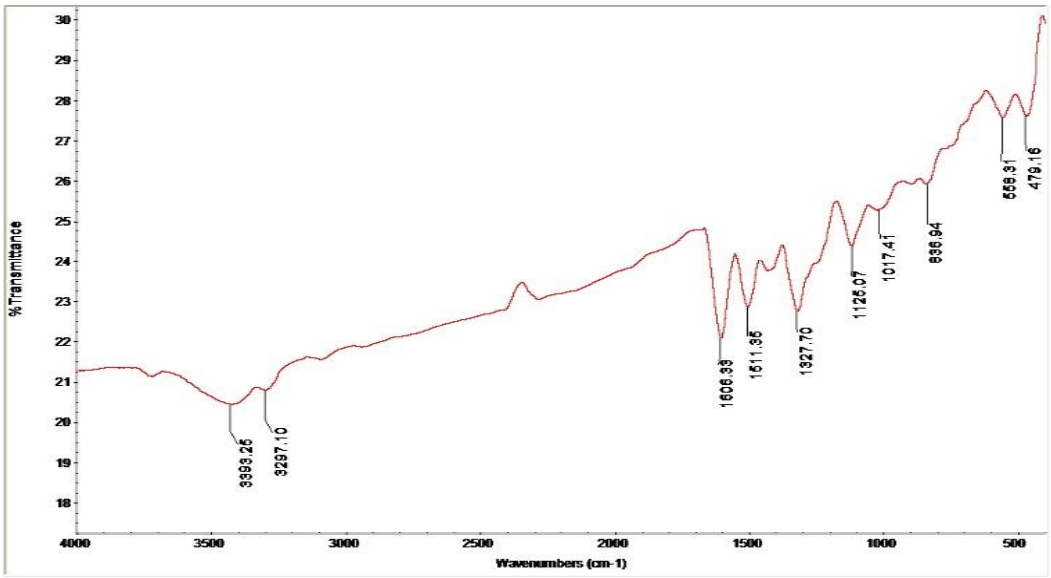


Figure.3: Infrared spectrum of Schiff base HL1

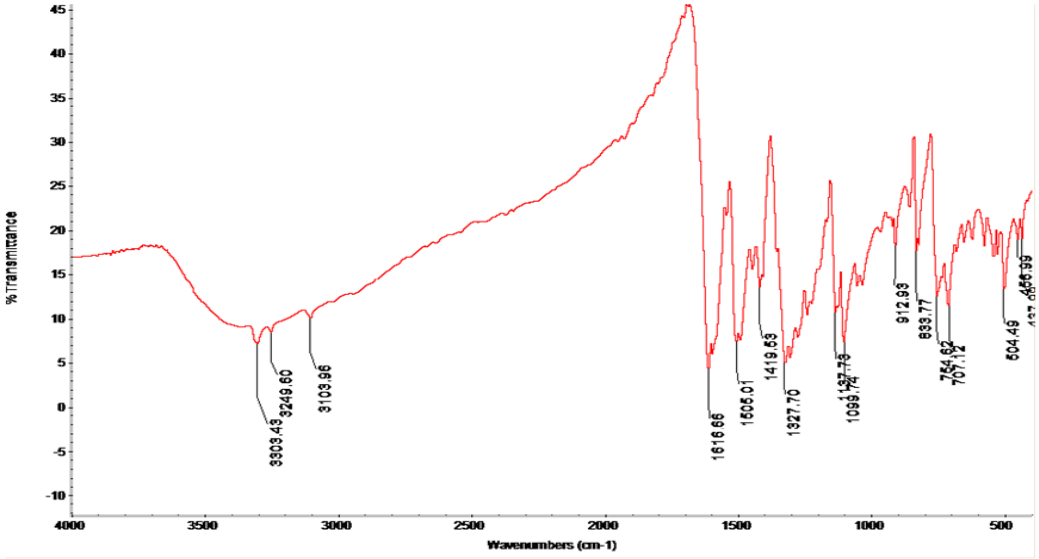


Figure.4: Infrared spectrum of [Cr(L1)(L2)$_2$] mixed ligand complex

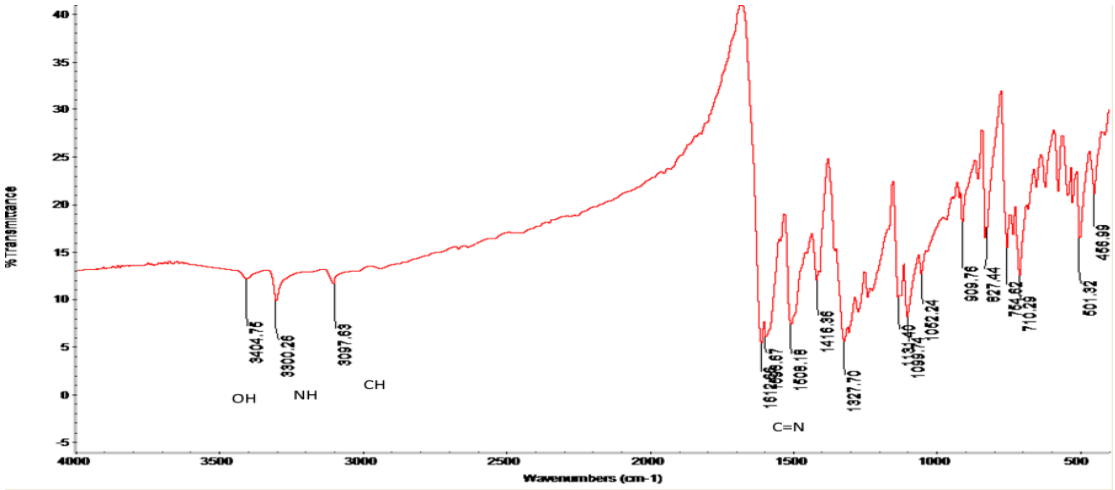


Figure.5: IR spectrum of [Mn(L1)(L2)(H_2O)$_2$] mixed ligand complex

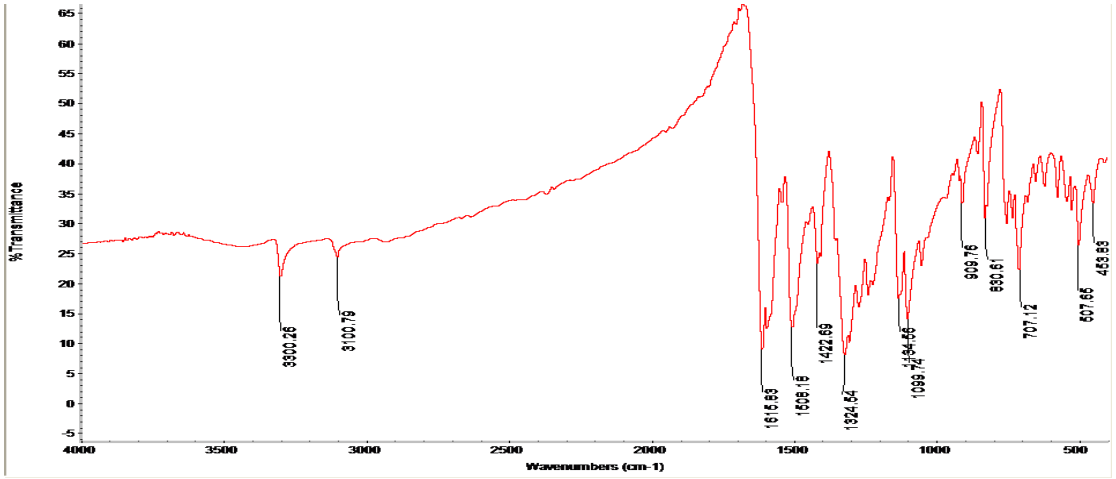


Figure.6: Infrared spectrum of [Fe(L1)(L2)(ONO_2)(H_2O)] mixed ligand complex

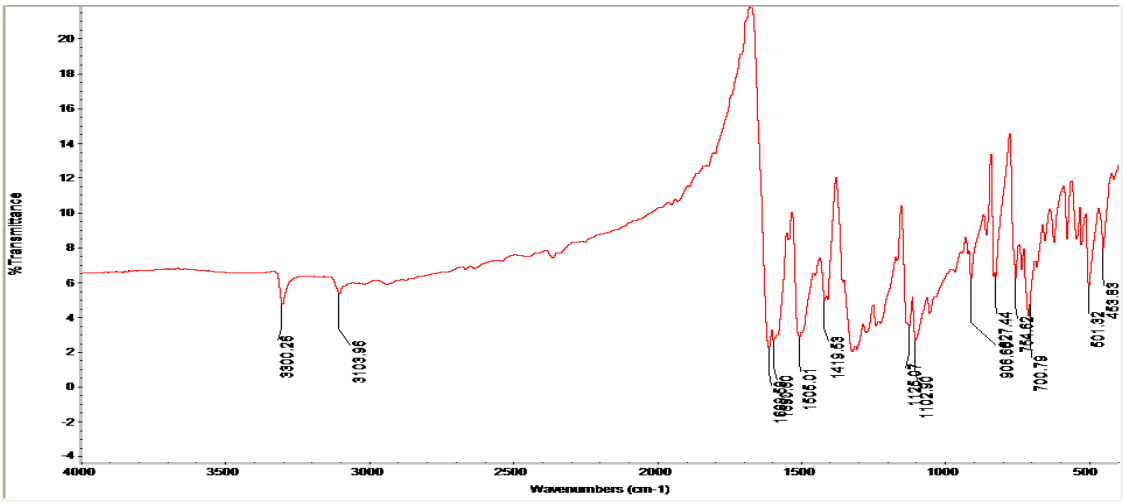


Figure. 7: IR spectrum of [Co(L1)(L2)(H_2O)$_2$] mixed ligand complex

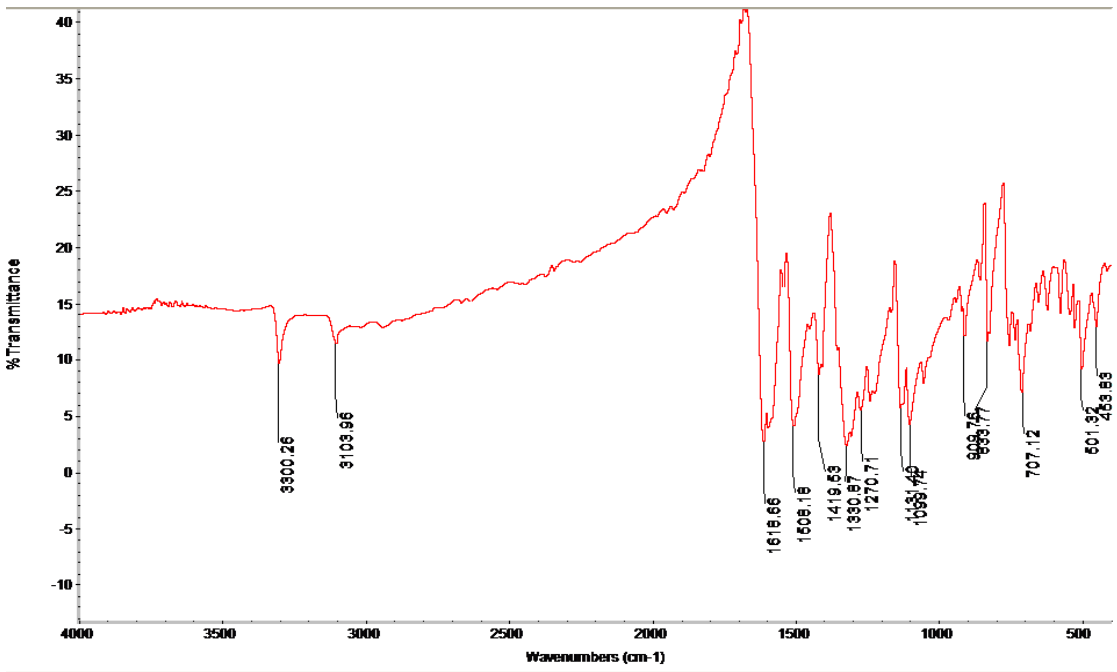


Figure.8: Infrared spectrum of [Ni(L1)(L2)(H_2O)$_2$] mixed ligand complex

Electronic spectra and magnetic moment

The electronic spectra of the Schiff base and its mixed ligand complexes are shown in figures (9-14) and their electronic spectral data (cm^{-1}) are listed in table 2. The electronic spectrum of the Schiff base shows bands at 44444 and 28169cm^{-1} attributed to $\pi\rightarrow\pi^*$ (phenyl ring) and $n\rightarrow\pi^*$ (C=N)(Sani *et. al*., 2015; Hamil *et. a*l., 2009).[23-24] The magnetic moment value of the Cr(III) complex (3.78 B.M) and the spectrum of the same complex which exhibits two bands at 26667 cm^{-1} and 19802 cm^{-1} belong to $^4A_{2g}(F)\rightarrow{}^4T_{2g}(F)$ and $^4A_{2g}(F)\rightarrow{}^4T_{1g}(F)$ transition supporting the presence of an octahedral geometry(Adalikwu *et. al*., 2011).[25] Whereas, the spectrum of Mn(II) complex shows three bands,The first two of them at 18518 cm^{-1} and 32258 cm^{-1} which are due to $^6A_{1g}\rightarrow{}^4T_{1g}(P)$ transition and 35714 cm^{-1} assigned to CT transition for octahedral geometry with magnetic moment value of 5.65 B.M)(Al- Mukhtar *et. al*., 2017).[26] The magnetic moent of Fe(III) mixed ligand complex shows a magnetic moment value of 5.15 B.M confirming the existence of five odd electrons(d^5) and the electronic absorption spectrum of this complex exhibits three bands at 33333 cm^{-1}, 28571 cm^{-1} and 22471 cm^{-1} due to $^3A_{2g}(F)\rightarrow{}^3T_{1g}(F)$ and $^3A_{2g}(F)\rightarrow{}^3T_{1g}(P)$ transitions which suggest an octahedral geometry(Mane *et. al* 2011; Sonmez *et.al*.,2001).[27,28] The electronic absorption spectrum of Co(II) complex(3.87B.M) shows two bands at 19230 cm^{-1} and 24390 cm^{-1} due to $^4A_{2g}(F)\rightarrow{}^4T_{1g}(F)$ and $^4A_{2}g(F)\rightarrow{}^4T_{1}g(P)$ transitions which suggest an octahedral geometry(Shyamala et al., 2010).[29] While, two absorption bands were observed for Ni(II) complex (2.81B.M) at 20408 and 26667 cm^{-1} corresponding to $^3A_{2g}(F)\rightarrow{}^3T_{1g}(F)$, $^3A_{2g}(F)\rightarrow{}^3T_{1g}(p)$ transitions, respectively(Pal *et. al*., 2011).[30]

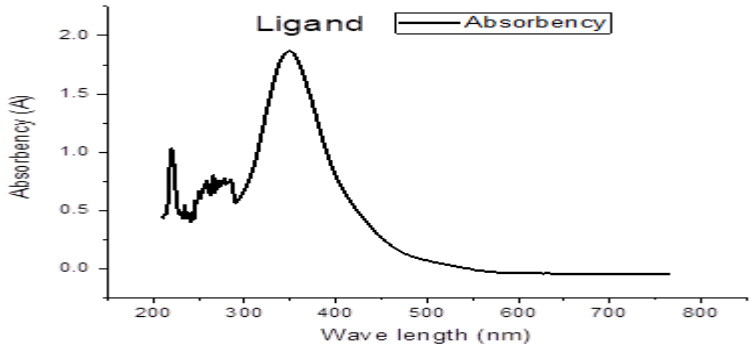


Figure.(9): Electronic spectrum of Schiff base (HL1)

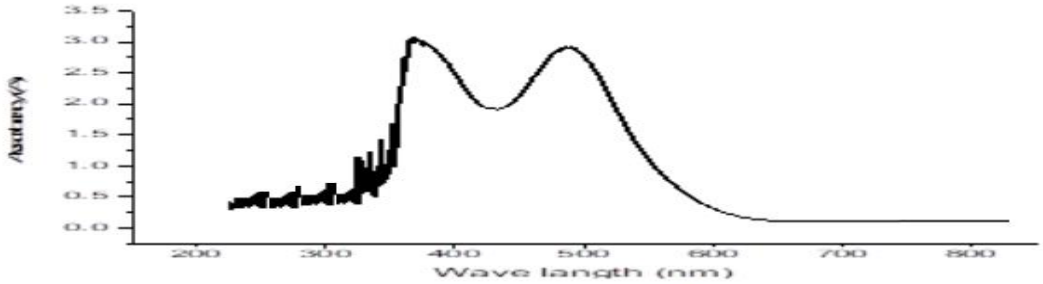


Figure.(10): Electronic spectrum of [Cr (L1)$(L2)_2$] mixed ligand complex

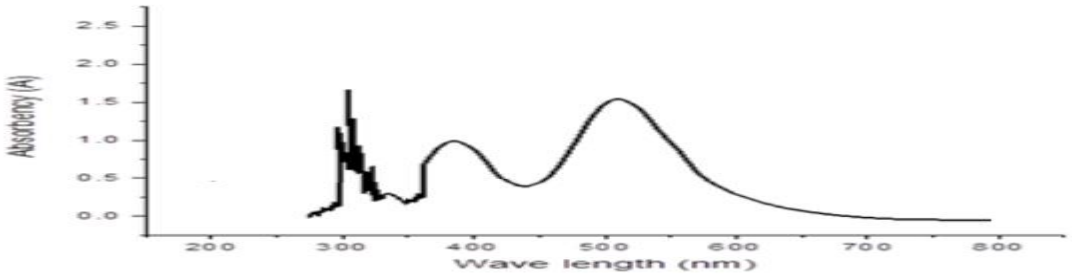


Figure.(11): Electronic spectrum of [Mn(L1)(L2)$(H_2O)_2$] mixed ligand complex

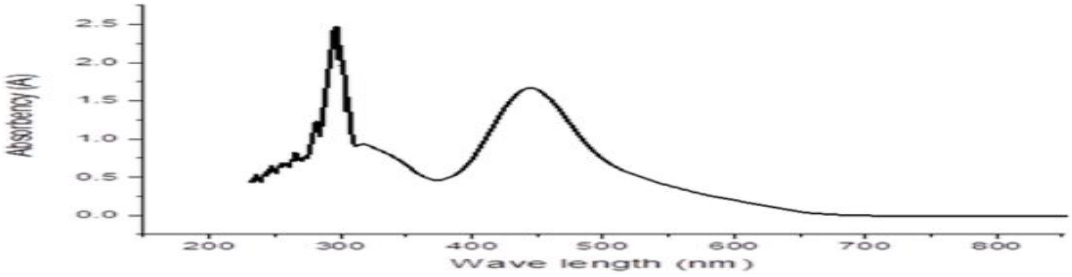


Figure.(12): Electronic spectrum of [Fe(L1)(L2)$(ONO_2)(H_2O)$] mixed ligand complex

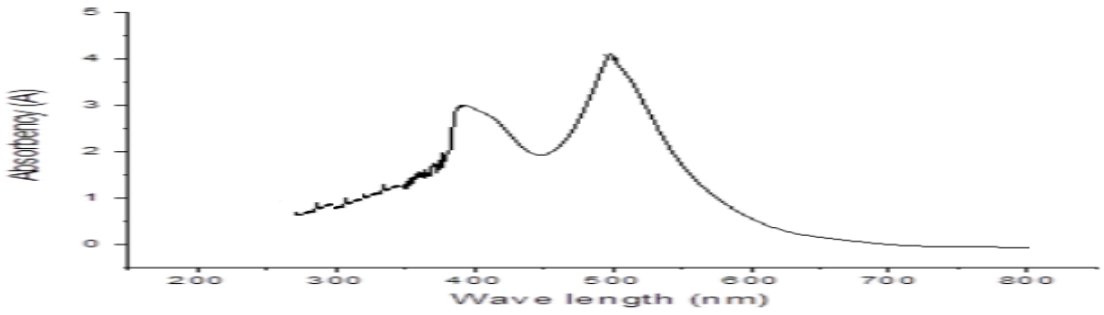


Figure.(13): Electronic spectrum of $[Co(L1)(L2)(H_2O)_2]$ mixed ligand complex

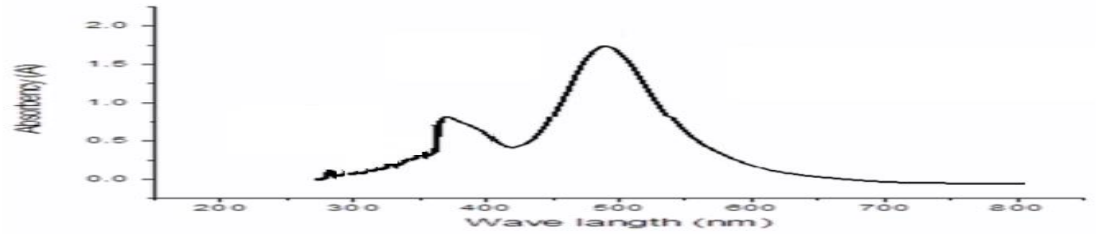
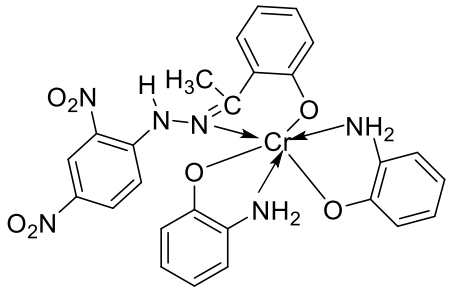


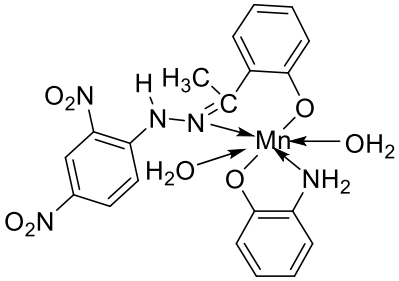
Figure.(14): Electronic spectrum of $[Ni(L1)(L2)(H_2O)_2]$ mixed ligand complex

Conclusion

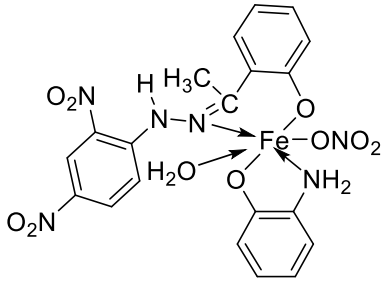
Based on the analytical data and spectroscopic techniques, the mixed ligand complexes were formed in two ratios, 1:1:2[M:L1:L2] as in Cr(III) complex and 1:1:1[ML1L2] as in the rest complexes. Also an octahedral structure was suggested according to electronic spectral data and magnetic mement values. The suggested structures were shown below:



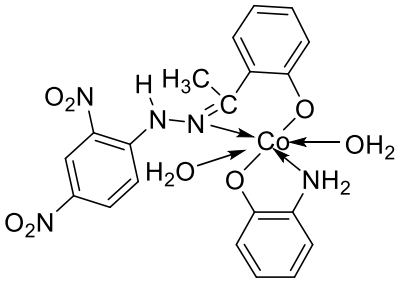
Structure of $[Cr(L1)(L2)_2]$ mixed ligand complex



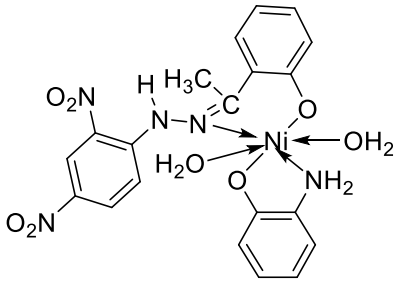
Structure of [Mn(L1)(L2)(H_2O)$_2$] mixed ligand complex



Structure of [Fe(L1)(L2)(ONO_2)(H_2O)] mixed ligand complex



Structure of [Co(L1)(L2)(H_2O)$_2$] mixed ligand complex



Structure of [Ni(L1(L2)(H_2O)$_2$] mixed ligand complex

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