

Synthesis and Physicochemical Studies of Some Mixed Schiff Bases Complexes

N. S. Al-barki*

Chemistry Department, Faculty of Science, Benghazi University, Benghazi, Libya

A. A. Maihub

Chemistry Department, Faculty of Science, Tripoli University, Tripoli, Libya

M. M. El-ajaily

Chemistry Department, Faculty of Science, Benghazi University, Benghazi, Libya

Taghreed H. Al-Noor

Chemistry Department, College of Education For Pure Science Ibn Al- Haitham, University of Baghdad

Abstract: Five mixed Schiff bases complexes of Co(II), Ni(II), Cu(II), Zn(II) and Fe(III) ions with two different Schiff bases, first Schiff base namely {2-((2-hydroxy-1-phenylethylidene)amino)-3-(4-hydroxyphenyl)propanoic acid} derived from the reaction of 2-hydroxyacetophenone and tyrosine as main ligand (HL1) and the second one namely {4-((2-(2,4-dinitrophenyl)hydrazono)methyl)-N,N-dimethylaniline} is formed from the condensation of 4-dimethylaminobenzaldehyde and 2,4-dinitrophenylhydrazine as a second ligand (HL2) were prepared. The Synthesized Schiff bases and their mixed Schiff base complexes were subjected to several physicochemical tools; in terms, CHN elemental analyses, molar conductivity, infrared, proton nuclear resonance, electronic, mass and electron paramagnetic resonance spectra. The CHN elemental analysis results showed the formation of the Schiff bases and their complexes in 1:1:1[M:L1:L2] ratio. The molar conductivity values revealed that Co(II), Ni(II), Cu(II) and Zn(II) mixed Schiff base complexes to be electrolytes due to the existence of sodium ion outside the complexes, but the iron(III) mixed Schiff base complex is neutral confirming the presence of non-electrolytic nature. The infrared spectral data of the mixed Schiff base complexes exhibited chelation behavior between the metal ions and the Schiff bases through the active groups presented in the Schiff bases such as nitrogen atom of the azomethine and oxygen atom of the hydroxyl group. The signals which obtained from the ¹HNMR spectra of the Schiff bases and their Zn(II) complex supported the chelation between the Zn(II) ion and the Schiff bases. Whereas, the UV spectral data of the Schiff bases displayed the $\pi \rightarrow \pi^*$ (Phenyl ring) and $n \rightarrow \pi^*$ (C=N and -OH) transitions and the UV-Vis spectral results of the mixed Schiff base complexes suggested the geometrical structure which is an octahedral geometry around the metal ions. The mass spectral fragmentations of the Schiff bases were studied and confirming the formation of the Schiff bases, also the mass spectral fragmentations of the Iron(III) mixed Schiff base complex was done. The electron paramagnetic resonance spectral results confirmed the data which are obtained from UV-Vis study.

Keywords: Schiff bases; Mixed Schiff base complexes; Physicochemical properties

1. Introduction

The Schiff base compounds formed from aromatic amines and aldehydes have wide variety of applications of inorganic, analytical and organic chemistry and are a field of coordination chemistry [1, 2]. Schiff base compounds and their complexes play an important applications in the area of food and dyes industry, agriculture, polymer, biological sciences as antibacterial and antifungal [3, 4]. Four Schiff base complexes of Co(II), Ni(II), Cu(II) and Cr(III) ions formed from 2,6-pyridinedicarboxaldehyde-thiosemicarbazone were prepared by conventional as well as microwave methods. This compound was investigated by using several techniques; such as, elemental analysis, molar conductance, magnetic susceptibility measurements, IR and Mass spectra. Analytical data displayed that all the complexes are in 1:1 (M:L) ratio with a coordination number of six [5]. Some mixed ligand chelates formed from di- and trivalent metal ions (Co(II), Ni(II), Cu(II) and Cr(III) ions) and Schiff base (L1) derived from the refluxing of 4-dimethylaminobenzaldehyde with 2-aminophenol as primary ligand, and 2-nitroaniline (L2) represents the secondary ligand were synthesized and investigated. The analytical data confirmed the existence of an octahedral structure for all mixed ligand chelates [6].

The main aim of this study is to synthesis and characterize the geometrical structures of the Schiff bases derived from the reaction of 2-hydroxyacetophenone and L-tyrosine as main ligand (HL1) in addition to other Schiff base formed from the reaction of 4-dimethylaminobenzaldehyde with 2,4-dinitrophenylhydrazine as secondary ligand (HL2) and their mixed Schiff base complexes with Co(II), Ni(II), Cu(II), Zn(II) and Fe(III) ions.

2. Materials and Methods

All chemicals used in this study are of pure grade (BDH or Aldrich). include; 2-hydroxyacetophenone, L-tyrosine, 4-dimethylaminobenzaldehyde, 2,4-dinitrophenyl-hydrazine, DMF, DMSO, NaOH, ethanol, methanol, $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$, $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$, $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, ZnCl_2 , $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and distilled water. The Schiff base compounds and their mixed Schiff base complexes were subjected to CHN elemental analysis using 2400-CHN elemental analyzer. The molar conductivity of the complexes was measured in DMF solvent using digital conductivity meter CMD-650, at chemistry department, Benghazi University, Benghazi, Libya. The infrared spectra were carried out applying KBr disc technique using IFS-25 DPUS/IR spectrometer. Proton nuclear magnetic resonance spectra of the Schiff bases and their Zn(II) mixed Schiff base complex were recorded on Varian Gemini200-200MHz spectrometer using TMS as internal standard and d^6 -DMSO as a solvent. The electronic spectra were measured in DMF solvent by using a Perkin-Elmer lambda-4 β spectrophotometer. The mass spectra also carried out by using Shimadzu QP-2010 Plus. The electron paramagnetic resonance spectrum was recorded by using EMX ESR spectrometer (Bruker) 1998Y. All previous tools and analysis were done at micro-analytical centre, Assiut University, Assiut, Egypt.

2.1. Synthesis of Schiff base compounds

2.1.1. Synthesis of Schiff base (HL1)

The Schiff base {HL1 = 2-((2-hydroxy-1-phenylethylidene)amino)-3-(4-hydroxyphenyl)propanoic acid} was synthesized as follows: NaOH (0.01 mol; 0.4gm) was dissolved in methanol (30 cm^3) and amino acid (L-tyrosine) (0.01 mol; 1.81gm) was added to it. The mixture was stirred magnetically at room temperature. When the mixture becomes homogeneous, 2-hydroxyacetophenone (1.36 gm) was added. After 2 minutes, the mixture was evaporated to 20% of its original volume and 1 cm^3 of acetic acid was added immediately. After 2 hours, yellow crystals formed. The obtained product was filtered, washed, dried and recrystallized from hot methanol to give pure crystals in excellent yield of 90.32%

2.1.2. Synthesis of Schiff base (HL2)

The Schiff base {HL2 = 4-(2-(2,4-dinitrophenyl)hydrazono)methyl)-N,N-dimethylaniline} was synthesized by dissolving 4-dimethylaminobenzaldehyde (0.01 mol; 1.49 gm) and same moles of 2,4-dinitrophenylhydrazine in ethanolic solution (30 cm^3). The mixture was heated and stirred magnetically for 2 hrs. Black crystals were formed. The obtained product was filtered, washed, dried and recrystallized from hot methanol to give pure crystals in excellent yield 92.70%. The resulting products of the ligands HL1 and HL2 were subsequently used for complex formation.

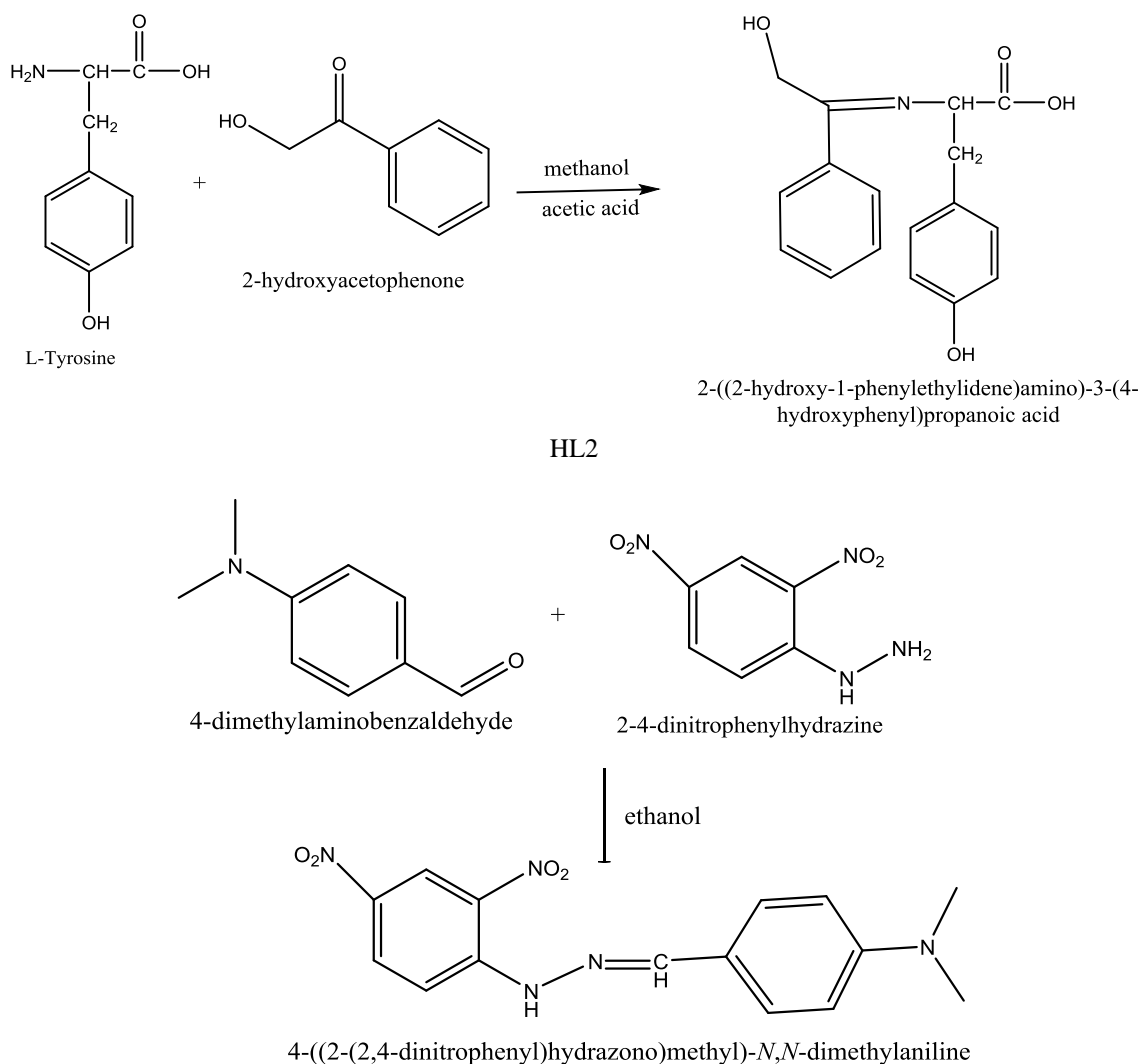
2.1.3. Synthesis of Mixed Schiff Base Complexes

The following mixed Schiff base complexes of Schiff bases under investigation with Co(II), Ni(II), Cu(II), Zn(II) and Fe(III) ions were synthesized by adding 0.01 mole, 3.29 gm of the Schiff base (HL1) and 0.01 mole; 3.30 gm of the Schiff base (HL2) in 25 cm^3 of the absolute ethanol to 0.01mole; 2.38, 2.37, 1.70, 1.36 and 2.70 gm of the desired metal salts, respectively. Few drops of sodium hydroxide solution were added slowly to adjusted the P^{H} value at 8 until the complexes isolated, and then the mixtures were stirred and refluxed for three hours. The formed precipitates were filtered and washed several time with hot ethanol until the filtrates become clear. The obtained mixed Schiff base complexes were dried in desecrator under calcium chloride.

3. Results and Discussion

The refluxing of 2-hydroxyacetophenone with L-tyrosine and 4-dimethylaminobenzaldehyde with 2,4-dinitrophenylhydrazine gives two Schiff bases HL1 and HL2 respectively as presented in Scheme 1:

Scheme-1. Structure of Schiff bases HL1 and HL2



3.1. Physical Properties, Microanalyses and Molar Conductivity

The Schiff bases and their mixed Schiff base complexes were synthesized in powder form with high melting points (Table 1). The Schiff bases are soluble in alcohols, DMF or DMSO solvents, whereas, the mixed Schiff base complexes are not soluble in alcohols, but they are partially soluble in DMF and DMSO. All compounds are stable in air. The CHN elemental analysis results of the Schiff bases and their mixed Schiff base complexes agree with the proposed compositions. This means that the Schiff bases were formed (Fig. 1) and their mixed Schiff base complexes were also formed in 1:1:1[M:L1:L2] ratio. The obtained molar conductivity values in DMF reveal the existence of electrolytic nature for Co(II), Ni(II), Cu(II) and Zn(II) mixed Schiff base complexes, because due to the presence of sodium ion outside the coordination sphere, whereas, the molar conductivity values for [Fe(L1)(L2)(H₂O)].2H₂O complexes exhibit non-electrolytic nature in this solvent., this means that there is no negative or positive ions outside the iron(III) complex [7].

Table-1. Some physical properties and CHN elemental analyses of Schiff bases and their mixed Schiff base complexes

Schiff bases /Mixed Schiff base complexes	M.wt	Colour	Yield%	(C% Calc. Exp.)	(H% Calc. Exp.)	(N% Calc. Exp.)	M.P °C	Am* $\Omega^{-1} \text{cm}^2 \text{mol}^{-1}$ In DMF
C ₁₈ H ₂₁ NO ₅ . (HL1)	331	Pale -yellow	90.32	68.22 69.01	5.69 6.20	4.68 5.45	112	-
C ₁₅ H ₁₆ N ₄ O ₅ (HL2)	330	Black	92.70	54.50 53.63	4.85 4.60	21.21 21.53	138	-
Na[Co(L1)(L2)(H ₂ O)]	758.9	Black	77.34	52.18 53.10	4.74 5.67	11.06 11.98	>250	95
Na[Ni(L1)(L2)(H ₂ O)]	768.6	Black	80.86	51.72 53.05	4.68 5.25	10.93 12.15	>250	89
Na[Cu(L1)(L2)(H ₂ O)]	763.5	Black	77.87	51.86 52.34	4.71 5.26	11.09 11.34	>250	85
Na[Zn(L1)(L2)(H ₂ O)]	765.4	Black	67.45	51.73 52.96	4.70 5.57	10.97 12.59	>250	83
[Fe(L1)(L2)(H ₂ O)].2H ₂ O	770	Black	75.54	51.42 52.25	5.19 5.39	10.90 12.70	>250	28

Am*= Molar Conductivity

3.2. Infrared Spectra of Schiff Bases and Their Mixed Schiff Base Complexes

The infrared spectra techniques have very important role in the determining of the positions of the groups that belong to the chemical formula of the compounds, these groups help in the investigation of the chemical structures of the compounds [8]. The infrared spectra for the present Schiff bases and their mixed Schiff base complexes were taken in the 400–4000 cm^{-1} range and helped to indicate regions of absorption due to the respective vibrations. The tentative IR bands assignments for the Schiff bases and their complexes are given in table 2. The infrared spectral data of the mixed Schiff base complexes exhibit bands in the range of 3428-3433 cm^{-1} corresponding to water molecules [9]. The infrared spectra of free Schiff bases HL1 and HL2 show two bands at 1609 and 1607 cm^{-1} , these bands are changed on complexation formation (Table 2) confirming the coordination of azomethine group through nitrogen atom [10]. The disappearance of COOH band of the free Schiff base HL1 in the spectra of the mixed Schiff base complexes indicates the involvement of COOH group through the oxygen atom in bonding with the metal ions [11]. In addition to the above, the IR spectra of mixed Schiff base complexes display bands related to $-\text{NO}_2$ group in the range of 1350 -1550 cm^{-1} . The slight deviation in these bands compared to the free Schiff base HL2 (1508 cm^{-1}) may be due to the effect of complexation. This deviation supports the involvement of nitro group (ortho-position) in bonding with the metal ions through oxygen atom [12]. The same spectra exhibit bands at 600–650 and 497-502 cm^{-1} range which are not exist in the free Schiff bases HL1 and HL2 due to (M-O) and (M-N) vibrations, respectively [13]. These bands confirm the formation of the mixed Schiff base complexes with these two Schiff bases. The spectral results of the mixed Schiff base complexes of HL1 and HL2 exhibit no change in NH group in comparison with its position in the spectrum of the free Schiff base HL2 suggesting that this group does not inter in chelation with the metal ions [14].

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

Complexes	ν OH (H ₂ O)	ν RC=N HL1, (HL2)	ν C-O	ν M-O	ν M-N	nm (cm ⁻¹)
C ₁₈ H ₂₁ NO ₅ (L1)	3422	1609	1158	–	–	258 (38759) 270 (37037)
C ₁₅ H ₁₆ N ₄ O ₅ (L2)		1607)(–	–	–	265 (37735) 290 (34482)
Na[Co(L1)(L2)(H ₂ O)]	3432	1609, (1513)	1045	633	499	439 (22676) 734 (13624)
Na[Ni(L1)(L2)(H ₂ O)]	3425	1609, (1512)	1043	634	497	439 (22779) 734 (13624)
Na[Cu(L1)(L2)(H ₂ O)]	3433	1607, (1509)	1071	628	502	441 (22676) 733 (13643)
Na[Zn(L1)(L2)(H ₂ O)]	3428	1608, (1512)	1073	632	528	445 (22472) 733 (13643)
[Fe(L1)(L2)(H ₂ O)].2H ₂ O	3425	1609, (1513)	1046	633	497	441 (22676) 733 (13643)

R=CH₃ for HL1, (R=H for HL2)

3.3. ¹HNMR Spectra of Schiff bases and their Zn(II) mixed Schiff Base Comple

The resonance of protons has been assigned on the basis of their integration and multiplicity pattern. ¹HNMR spectra of the Schiff base compounds (HL1 and HL2) are shown in figures (1, 2). The ¹HNMR spectrum of Schiff base (HL1) shows signals in the range of 6.60-7.00 ppm indicating the existence of phenyl protons. Whereas, the

signals at 7.04 and 3.50 ppm due to the existence of -OH and -COOH groups in the Schiff base HL1. The DMSO solvent and methyl group appeared at 2.00 and 2.51 ppm, respectively. The ^1H NMR spectrum of Schiff base (HL2) reveals several signals in the region of 2.15-11.5 ppm. The NH signal which is present in this Schiff base is appeared at 11.50 ppm. The HC=N signal was observed at 8.86 ppm [15]. The signals in the range of 6.78-7.61 ppm are assigned to the presence of phenyl protons of the Schiff base (HL2). The ^1H NMR spectrum of the diamagnetic Zn(II)-L1L2 complex displays the disappearance of signals due to the OH/COOH protons indicating their replacement by the metal ion during chelation formation. The azomethine proton signal of HL2 is shifted slightly to low field during the chelation process confirming the bonding of this group with Zn(II) ion through azomethine nitrogen. The integrated intensities of all the signals agree well with the figure (3) of the complex.

Fig-1. ^1H NMR spectrum of Schiff base HL1

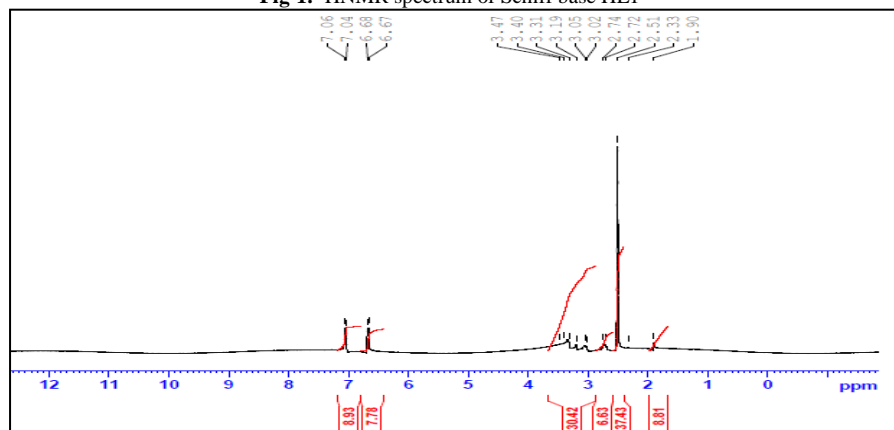


Fig-2. ^1H NMR spectrum of Schiff base HL2

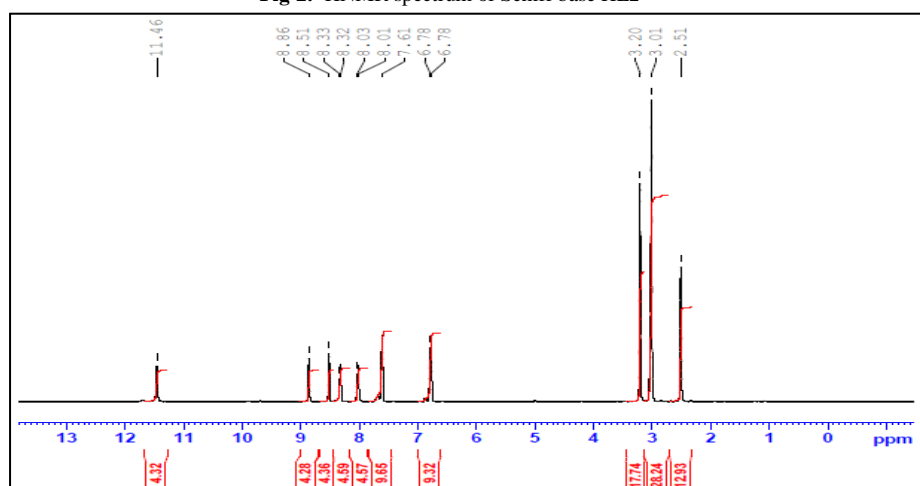
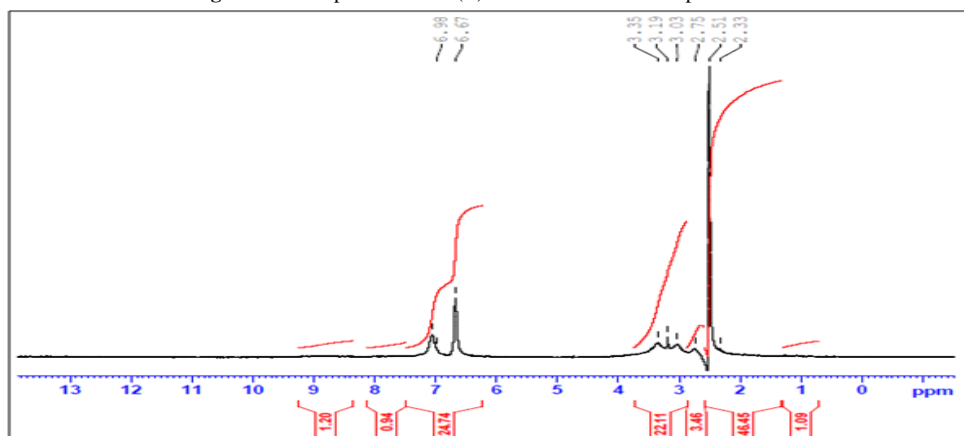


Fig-3. ^1H NMR spectrum of Zn(II) mixed Schiff base complex

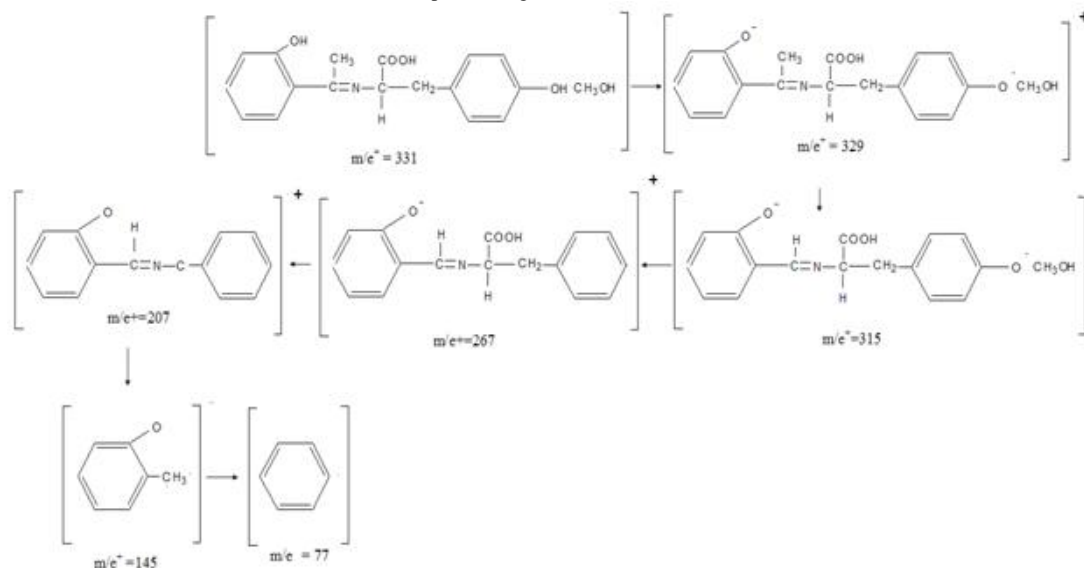


3.4. Mass Spectra of The Schiff Base and Fe(III) Mixed Schiff Base Complex

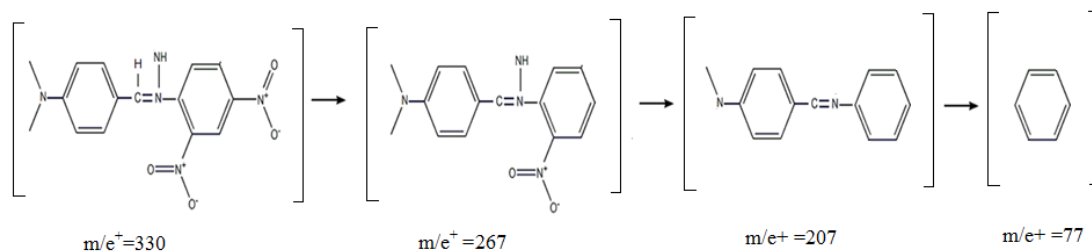
The peak at $m/e^+ = 329$ analogous to $\text{C}_{18}\text{H}_{19}\text{NO}_5^+$. Meanwhile, another peak at $m/e^+ = 315$ due to the formula of $\text{C}_{17}\text{H}_{17}\text{NO}_5^+$. The structure $\text{C}_{16}\text{H}_{13}\text{NO}_3^+$ is analogous to peak at $m/e^+ = 267$. The same spectrum shows a peak at $m/e^+ = 207$ corresponding to the formula of $\text{C}_{14}\text{H}_{10}\text{NO}^+$, the peak at $m/e^+ = 145$ is attributing to $\text{C}_{10}\text{H}_9\text{O}$. The loss of

three carbon atoms, two hydrogen atoms give a peak at $m/e^+ = 107$. The last peak at $m/e^+ = 77$ is attributing to C_6H_5 (Scheme 2, Fig.4). The mass spectrum of this Schiff base (HL2) reveals a peak at $m/e^+=330$ assigning to the original molecular weight of the Schiff base (Scheme 3, Fig.5). The peak at $m/e^+ =267$ analogous to $C_{15}H_{13}N_3O_2$. whereas, the same spectrum exhibits a peak at $m/e^+ =207$ which is attributed to the formula $C_{14}H_{11}N_2$. The last peak at $m/e^+ =77$ is corresponding to C_6H_5 . The mass spectrum fragmentation of the Fe(III) mixed Schiff base complex (Scheme 4, Fig.6) shows a molecular ion peak at $m/e^+ = 329$ which is corresponding to $[C_{15}H_{15}N_5O_4]^+$. Meanwhile, the peak at $m/e^+ = 315$ is analogous to $[C_{14}H_{13}N_5O_4]^+$. The peak at $m/e^+ = 268$ due to $[C_{14}H_{12}N_4O_2]^+$. Whereas, the peak at $m/e^+ = 207$ is attributed $[C_{13}H_9N_3]^+$ and the peak at $m/e^+ = 107$ is assigned to $[C_6H_7N_2]^+$. The final peak appeared at $m/e^+=77$ analogues to C_6H_5 , (Scheme 3, fig. 5)

Scheme-2. Mass spectral fragmentation of Schiff base (HL1)



Scheme-3. Mass spectral fragmentation of Schiff base (HL2)



Scheme-4. Mass spectral fragmentation of Fe(III) mixed Schiff base complex

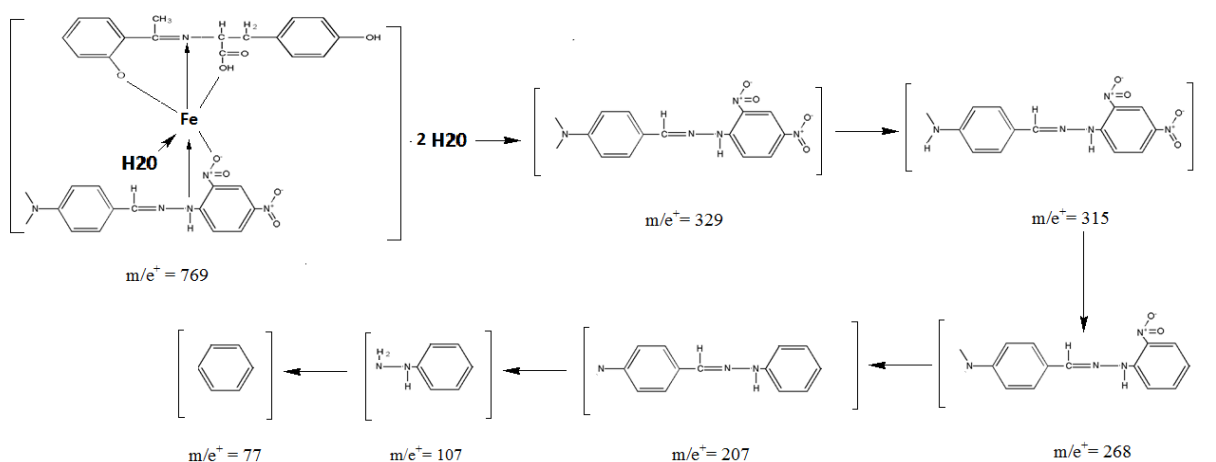


Fig-4. Mass spectrum of Schiff base (HL1)

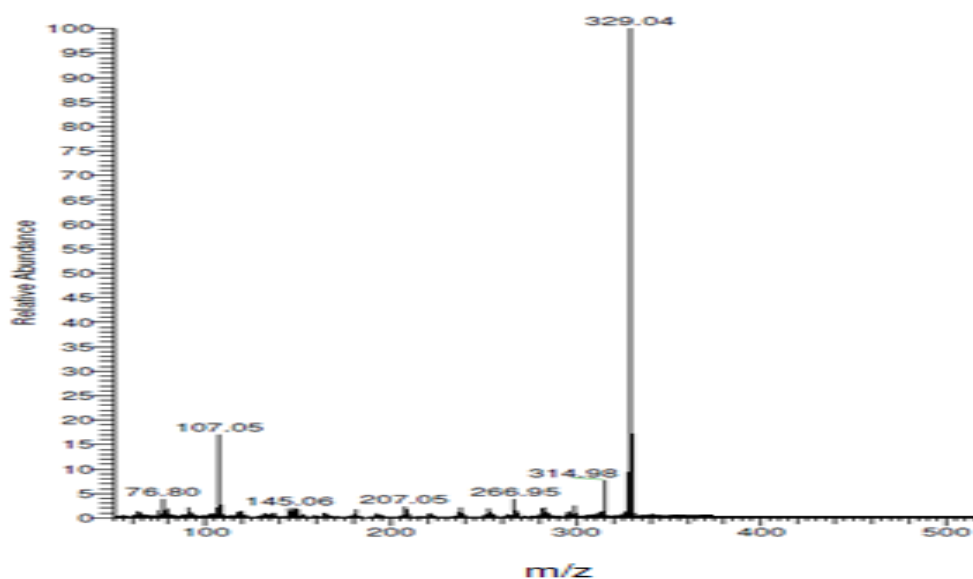


Fig-5. Mass spectrum of Schiff base (HL2)

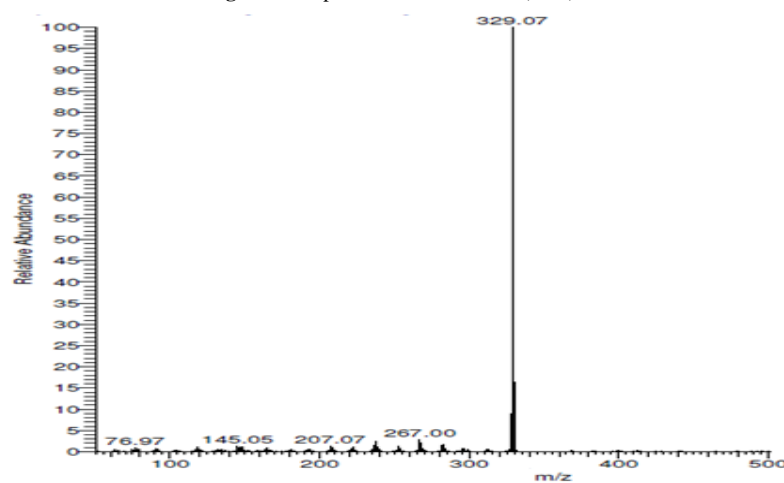
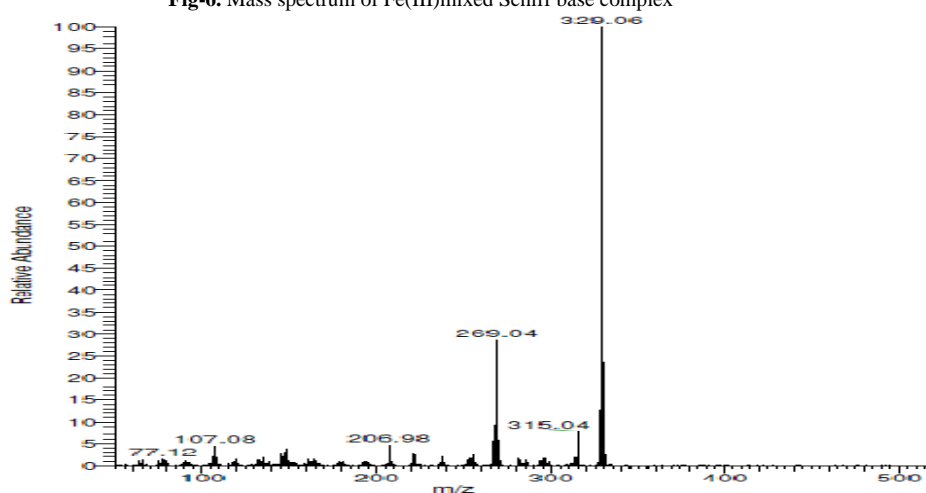


Fig-6. Mass spectrum of Fe(III) mixed Schiff base complex



3.5. UV/Visible Spectra and Magnetic Moments

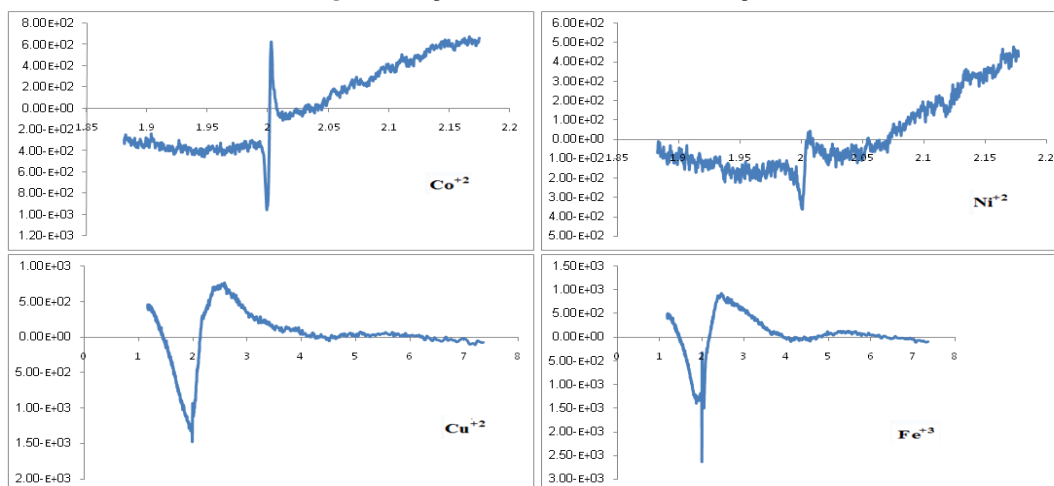
Information concerning the geometry of the complexes was obtained from the electronic spectra. UV/Visible spectra of the Schiff bases (HL1 and HL2) and their mixed Schiff base complexes were recorded at room temperature in DMF (10^{-3} M) and their data are recorded in Table 2, meanwhile, the spectrum of Schiff base (HL1) exhibits two bands at 265 nm (37736 cm^{-1}) and 290 nm (34483 cm^{-1}) corresponding to $\pi \rightarrow \pi^*$ (Phenyl ring) and $n \rightarrow \pi^*$ transitions, respectively. In the spectrum of the Schiff base (HL2), the band observed at 258 nm (38759 cm^{-1}) is attributed to $\pi \rightarrow \pi^*$ transition and the other band at 270 nm (37037 cm^{-1}) is due to $n \rightarrow \pi^*$ transition associated with the azomethine

chromosphere (HC=N). The bands observed at 439 nm (22779 cm^{-1}) and 734 nm (13624 cm^{-1}) in the spectrum of Na[Co(L1)(L2)(H₂O)] complex are assigned to ${}^4\text{T}_{1g} \rightarrow {}^4\text{A}_{2g}(\text{F})$ and ${}^4\text{T}_{1g}(\text{F}) \rightarrow {}^4\text{T}_{1g}(\text{P})$ transitions, respectively. Based on the intensity of the bands and the magnetic moment value (5.20 B.M) an octahedral structure was suggested for the complex [16]. The spectrum of Na[Ni(L1)(L2)(H₂O)] complex shows two bands at 439 nm (22779 cm^{-1}) and 734 nm (13624 cm^{-1}) corresponding to ${}^3\text{A}_{2g}(\text{F}) \rightarrow {}^3\text{T}_{2g}(\text{F})$ and ${}^3\text{A}_{2g}(\text{F}) \rightarrow {}^3\text{T}_{1g}(\text{P})$ transitions. The nature of the bands and the magnetic moment (4.33 B.M) support the presence of an octahedral geometry around Ni(II) ion [17]. The spectrum of Na[Cu(L1)(L2)(H₂O)] complex reveals two bands at 441 nm (22676 cm^{-1}) and 733 nm (13643 cm^{-1}) due to ${}^2\text{B}_{1g} \rightarrow {}^2\text{B}_{2g}$ and ${}^2\text{E}_g \rightarrow {}^2\text{T}_{2g}$ transitions. The magnetic moment value (2.10 B.M) and intensity of the bands support the existence of an octahedral geometry around Cu(II) ion [18]. The electronic spectral data of the Na[Zn(L1)(L2)(H₂O)] show two bands at 626 nm (15974 cm^{-1}) and 733 nm (13643 cm^{-1}) due to charge transfer transition and an octahedral geometry was suggested for this complex [19]. The electronic spectrum of the [Fe(L1)(L2)(H₂O)].2H₂O complex reveals two bands at 441 nm (22676 cm^{-1}) and 733 nm (13643 cm^{-1}). The first band is due to ${}^2\text{E}_g \rightarrow {}^2\text{T}_{2g}$ and the second band is due to d-d transition, the investigation of the bands and the obtained magnetic moment value (5.44 B.M) confirm the existence of an octahedral structure for the complex [20].

3.6. Electron Paramagnetic Resonance Spectra of Mixed Schiff Base Complexes L1L2

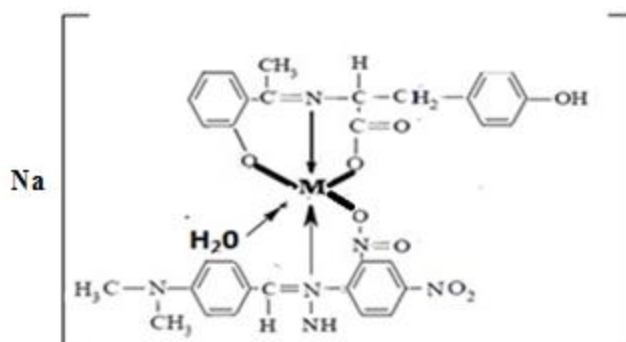
The electronic paramagnetic resonance spectral data of Co(II), Ni(II), Cu(II) and Fe(III) mixed Schiff base complexes under investigation exhibit g values of 2.019, 2.174, 2.122 and 2.183, respectively. These values are deviated from the ideal free electron (2.0023) indicating the formation of the covalent bonds between the metal ions and the Schiff bases HL1 and HL2. The obtained values confirm the existence of an octahedral structure for all the complexes [21]. All obtained g values support the data resulted from the electronic spectra.

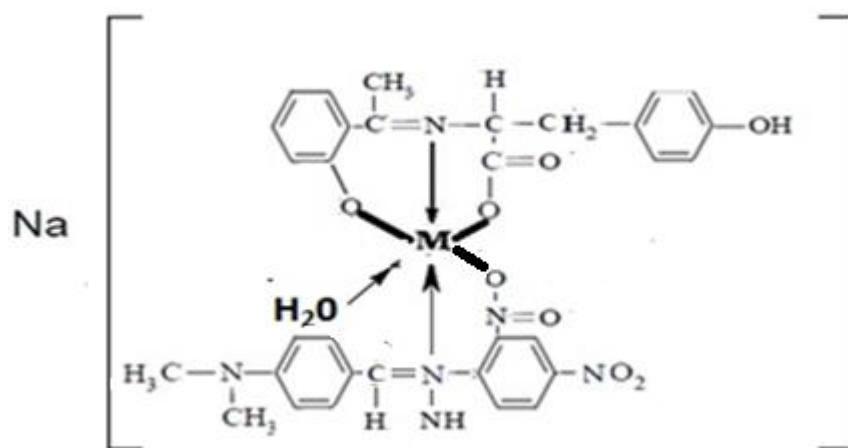
Fig-7. E.P.R spectra of the mixed Schiff base complexes L1L2



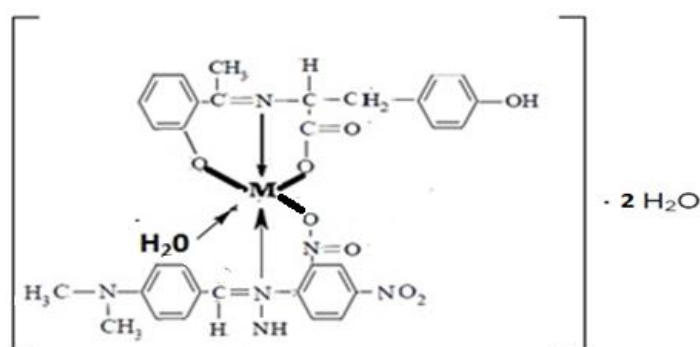
4. Conclusion

From the previous physicochemical data we can conclude that the mixed ligand complexes were found to have the general formulae Na[M(L1)(L2)(H₂O)], M= Co(II), Ni(II), Cu(II), Zn(II) and [Fe(L1)(L2)(H₂O)].2H₂O, respectively. The ligand HL1 behaves as a tridentate ligand via the nitrogen atom of the azomethine and both the oxygen (carboxyl and hydroxyl groups), while, the ligand HL2 behaves as a bidentate ligand via both the oxygen atom of the nitro and the nitrogen atom of the azomethine groups. Based on the obtained experimental data and literature indications, structural formulae to these complexes were assigned for all mixed Schiff base complexes as shown be





M= Co(II), Ni(II), Cu(II) and Zn(II) ions



M= Fe(III) ion

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