



## Preparation, PhysicoChemical Studies and Antimicrobial Evaluation of 2-(2-[(2-hydroxyphenyl)ethanimidoyl]Phenyl-ethanimidoyl)phenol of Novel Transition metal Chelates

M. M. El-ajaily\*, H. A. Abdullah\*\*, A. A. Aljanga\*\*\*, A. A. Maihub\*\*\*\* and E. E. Saad\*\*

\* Deptt. of Chemistry, Faculty of Science, Benghazi University, Benghazi, Libya.

\*\* Deptt. of Chemistry, Faculty of Science, Sebha University, Sebha, Libya.

\*\*\* Deptt. of Zoology, Faculty of Science, Sebha University, Sebha, Libya.

\*\*\*\* Deptt. of chemistry, Faculty of Science, Tripoli University, Tripoli, Libya.

Email ID: [melajaliy@yahoo.com](mailto:melajaliy@yahoo.com)

(Received 12 June, 2013; Accepted 28 June, 2014; Published 19 Sept, 2014)

**ABSTRACT:** A series of chelates of Zr(IV), La(III) and Ce(IV) with a bidentate ligand, 2-(2-[(2-hydroxyphenyl)ethanimidoyl]phenylethanimidoyl)phenol were prepared and characterized on the basis of elemental analysis, molar conductivity, mass spectra, infrared, <sup>1</sup>HNMR, TGA and UV-Visible spectra. Their molar conductance values indicated that all the chelates are non-electrolyte. From elemental analysis data, the chelates have been found to be 1:1 (metal: ligand). According to these data, an octahedral geometry was suggested for the chelates. The ligand and its metal chelates were screened for their physiological activities against some pathogenic antimicrobial activity two Gram-negative (*Escherichia coli*, *Salmonella kentucky*), two Gram-positive (*Lactobacillus fermentum*, *Streptococcus faecalis*) bacterial strains and fungi unicellular (*Fusarium solani*) and filamentous fungi (*Aspergillus niger*). The activity data showed that the metal chelates have antibacterial and antifungal activity more than the parent Schiff base ligand against one or more bacterial or fungi species.

**Keywords:** Transition metal ions; Schiff base, Metal chelates, Spectral characterization, antimicrobial evaluation.

### INTRODUCTION

Schiff bases and their coordination compounds have gained importance recently because of their applications as models in biological, biochemical, analytical, antimicrobial system, anticancer, antibacterial and antifungal activities<sup>1</sup>. Schiff bases of o-phenylenediamine and its complexes have a variety of applications including biological, clinical and analytical science<sup>2</sup>. El-ajaily et al<sup>3 & 4</sup> studied many Schiff base complexes which showed a good antibacterial activity. The Fe(III) Schiff base complexes which formed from the reaction of o-phenylenediamine with salicylaldehyde and isatin or 2-hydroxynaphthaldehyde or acetyl acetone were synthesized. The complexes were characterized by elemental analyses, molar conductance measurements, magnetic susceptibility, IR, UV-Vis spectra and thermal analysis. The elemental analysis data confirmed the stoichiometry of the type [ML(H<sub>2</sub>O)<sub>2</sub>](OAc)<sub>2</sub>. The complexes were found to be electrolytic in nature on the basis of the values of the molar conductivity. From the spectral data, an octahedral geometry has been proposed for all the complexes. The complexes have been screened for their antibacterial and antifungal activity<sup>5</sup>. A synthesized Schiff base from 2-amino, 4-methyl phenol with o-hydroxyacetophenone (AMPOHA) and its Ti(IV) and Zr(IV) complexes were characterized on the basis of elemental analysis, molar conductivity, infrared, electronic, <sup>1</sup>HNMR and X-ray diffraction studies. The molar conductivity measurements revealed that the complexes are non-electrolytic in nature. The analytical data indicated that the metal to ligand ratio in the complexes was 1:1. The IR spectral data suggested that the ligand behaves as tridentate ONO donor system and X-ray diffraction studies indicated monoclinic crystal system for these complexes. The Schiff base and its metal complexes have been evaluated for their antimicrobial activity using disc diffusion

methods. The reaction was carried out by both conventional and microwave methods. This Schiff base ligand system coordinated with the metal ions in a tetradentate manner through the oxygen and azomethine nitrogen. The metal chelates formed with this Schiff base ligand may be used as precursors for the synthesis of compounds. Some of which they may exhibit interesting physical and chemical properties and potentially useful biological activities<sup>6</sup>.

This investigation aims to synthesize and characterize some new Schiff base chelates of Zr(IV), La(III) and Ce(IV) ions. Also all the synthesized compounds were screened for their antimicrobial activity against some pathogenic bacteria and fungi species.

## MATERIAL AND METHODS

All chemicals and solvents used for the investigation of Schiff base and its chelates were of Merck or Aldrich. They include; 2-hydroxyacetophenone, 1,2-diaminobenzene,  $ZrOCl_2 \cdot 8H_2O$ ,  $La(NO_3)_3 \cdot 6H_2O$ ,  $Ce(SO_4)_2 \cdot 4H_2O$ ,  $C_2H_5OH$ ,  $CHCl_3$ , DMF, DMSO and  $NH_4OH$ . Distilled water was used through the experimental and was used without further purification. The synthesized Schiff base and its chelates were subjected to CHN elemental analyses using Perkin-Elmer 2400 elemental analyzer, infrared spectra were obtained by KBr disc technique by using IFS-25DPUSR\IR spectrometer (Bruker) in the range of 4000-400  $cm^{-1}$ , proton nuclear magnetic resonance spectrum of the Schiff base was recorded on Varian Gemini 200-200MHz spectrometer using TMS as internal standard and  $D^6$  DMSO as a solvent. The electronic spectra of the Schiff base and its chelates were measured in DMSO solvent using a Perkin-Elmer-Lambda  $\beta$ -spectrophotometer. The mass spectra were carried out by using Shimadzu QP-2010 Plus. The molar conductivity of the chelates was measured in DMSO solvent using digital conductivity meter CMD 650, at Chemistry Department, Sebha University, Sebha, Libya. The thermogravimetric analysis of Ce(IV) chelate was achieved using Shimadzu thermal analyzer (Japan). The weight loss of the sample was measured from ambient temperature upto 1000<sup>o</sup>C in a rate of 10<sup>o</sup>C per minute. The CHN elemental analyses, thermogravimetric analysis and spectroscopic analyses were done at Micro analytical center, Cairo University, Giza, Egypt.

**Preparation of the Schiff Base (SB):** The Schiff base (SB) of 2-(2-[(2-hydroxyphenyl)ethanimidoyl]Phenyl-ethanimidoyl)phenol (**1**) was synthesized by drop wise addition of hot ethanolic solution (25 mL) of 0.02 mole; (2.72 g) of 2-hydroxyacetophenone with the same amount of the solvent of 0.01 mole; (1.08 g) of 1,2-diaminobenzene. The reaction mixture was refluxed for two hours, and then the product was filtered, washed several times with hot ethanol and dried in a desiccator over calcium chloride. The yield of the pale lemon-yellow precipitate was 71.40% and its melting point is in the range of 73-75<sup>o</sup>C.

**Preparation of the Schiff base chelates:** The metal chelates under investigation were synthesized by drop wise addition of an ethanolic solution of the metal salts (0.01 mole; 3.22, 4.33 and 4.04 g) of  $ZrOCl_2 \cdot 8H_2O$ ,  $La(NO_3)_3 \cdot 6H_2O$  and  $Ce(SO_4)_2 \cdot 4H_2O$ . If the chelates did not isolate, few drops of ammonia solution were added to adjust the pH = 7-8. The reaction mixtures were refluxed for four hours, and then they collected and washed several times with hot ethanol until the filtrates become clear. The chelates were dried in desiccators over anhydrous calcium chloride. The yield of the chelates were ranged from 66.79-84.79 % and their melting points are above 300 <sup>o</sup>C.

**Antimicrobial activity assay:** Antimicrobial activity of the tested samples was determined using a modified Kirby-Bauer disc diffusion method<sup>7</sup> The *in vitro* antibacterial screening of the ligand and its chelates was undertaken against the two Gram-negative bacterial strains (*Escherichia coli* & *Salmonella Kentucky*), two Gram-positive bacterial strains (*Lactobacillus fermentum* & *Streptococcus faecalis*) by disc diffusion method using Mueller Hinton agar as medium and ciprofloxacin as control. In a typical procedure, molten nutrient agar kept at 45<sup>o</sup>C was then poured into Petri dishes and each of the compounds was dissolved in DMSO solvent and solutions of the concentrations (5, 50 and 500  $\mu g/mL$ ) were prepared separately. The plates were incubated for 18 h at 37<sup>o</sup>C. The diameter of the zones of inhibition for all the tested compounds was measured and the results were compared with the standard drug ciprofloxacin of the same concentration as that of the test compound under identical conditions.<sup>8</sup> The antifungal activity of

the tested compounds was evaluated against the *Aspergillus niger* and *Fusarium solani*, by cup-plate method cultured on Sabourard Dextrose Agar medium adapting similar procedure described above. The plates were incubated at 37 °C for 40 hours. The diameters of the zone of inhibition for all the tested compounds were measured and the results were compared with the standard drug miconazole as that of the tested compound under identical conditions. Inhibition was recorded by measuring the diameter of the inhibitory zone after the period of incubation.<sup>9</sup>

The diameter of the inhibition zone (mm) was measured and the activity index was also calculated by using the following Eq.:

$$\% \text{ Activity index} = C / T \times 100 \text{ ----- Eq.}^{10}.$$

Where; AI% = Activity Index  
 C = diameter of zone inhibition of micro- organisms in check  
 T = diameter of the disc (The zone of inhibition was measured after 18-20 hrs.)  
 Ciprofloxacin (5µg/mL) and Miconazole (25µg/mL) was used as positive standard.

## RESULTS AND DISCUSSION

Schiff base ligand (1) (Figure1) which obtained by the reaction of 2-hydroxyacetophenone and 1,2-diaminobenzene in 2:1[Carbonyl: amine] ratio.:

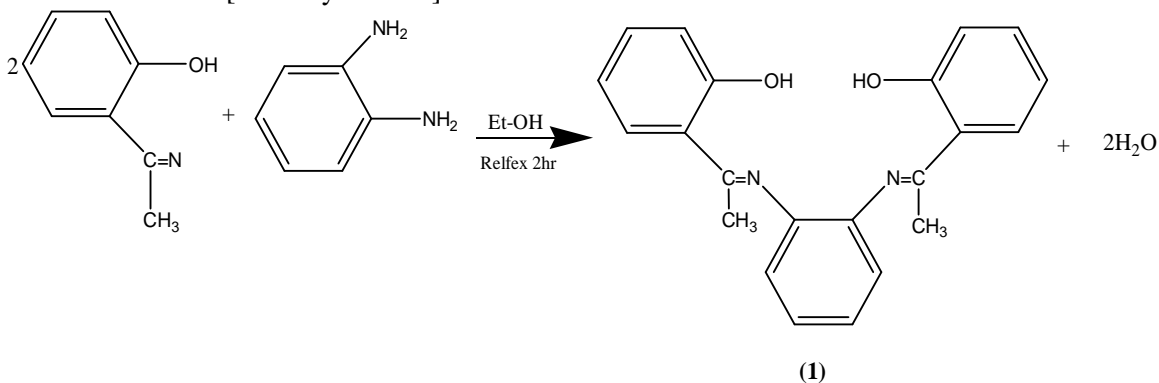


Figure 1: Structure of 2-(2-[(2-hydroxyphenyl) ethanimidoyl]Phenylethanimidoyl)phenol (1)

**1. Microanalysis and molar conductivity:** It is found that the found data are in a good agreement with those theoretical ones (Table-1) and the obtained analytical analysis data indicate the formation of 1:1 [M: L] ratio. The molar conductance values of the synthesized chelates were determined by using 10<sup>-3</sup>M concentration in DMSO solvent, and their values (Table 1) are In the range of 1.65 - 3.68 Ω<sup>-1</sup> cm<sup>2</sup> Mol<sup>-1</sup>. These values suggest the existence of non-electrolyte nature.<sup>11</sup>

Table 1: CHN analytical data and other physical properties of Schiff base and its chelates.

Comp.	Chemical Formulae	Mol. Wt.	Colour	M.P. (°C)	Yield %	Elemental analysis%			Am.
						Calc.	Found		
						C	H	N	
SB	C <sub>22</sub> H <sub>20</sub> N <sub>2</sub> O <sub>2</sub>	344.41	Dark Brown	73-75	71.40	77.10 <b>(76.70)</b>	5.63 <b>(5.85)</b>	8.19 <b>(8.23)</b>	-
[Zr(L)(OH) <sub>2</sub> ].H <sub>2</sub> O	C <sub>22</sub> H <sub>24</sub> N <sub>2</sub> O <sub>6</sub> Zr	503.76	Dark yellow	>300	84.79	41.42 <b>(41.22)</b>	6.95 <b>(7.05)</b>	4.03 <b>(4.00)</b>	1.77
[La(L)(OH)(OH <sub>2</sub> )].4H <sub>2</sub> O	C <sub>22</sub> H <sub>29</sub> N <sub>2</sub> O <sub>8</sub> La	588.49	Light Lime	>300	80.41	37.80 <b>(37.63)</b>	6.74 <b>(6.82)</b>	3.67 <b>(3.80)</b>	3.28
[Ce(L)(OH) <sub>2</sub> ].3H <sub>2</sub> O	C <sub>22</sub> H <sub>26</sub> N <sub>2</sub> O <sub>7</sub> Ce	570.57	Light Brick-red	>300	66.71	37.94 <b>(37.96)</b>	6.64 <b>(6.67)</b>	3.69 <b>(3.32)</b>	1.65

Am = Ω<sup>-1</sup> cm<sup>2</sup> Mol<sup>-1</sup> and M. Wt = Molecular weight.

**2. Thermogravimetric analysis of Ce(IV) chelate:** The thermogravimetric analysis was performed to assist in predicting the molecular structures of the chelates and the weight losses were measured from the ambient temperature up to 1000 °C using a heating rate of 10 °C / min<sup>12</sup>. Thermogravimetric curve of the Ce(IV) chelate of the formula [Ce(L)(OH)<sub>2</sub>].3H<sub>2</sub>O exhibits a weigh loss of 10.30% at temperature of 50-420 °C range analogous to the presence of three hydrated water molecules, this value is in a good agreement with the calculated one (9.78%). At temperature of 420-750°C range, the chelate loss two -OH groups and the Schiff base decomposes as carbonate or oxalate. The metal oxide (CeO<sub>2</sub>) appeared at the end of the curve at stable state above 750 °C and has weight loss of 52.95% which is in consistent with the calculated value (52.00%). The following steps show the decomposition of the chelate.

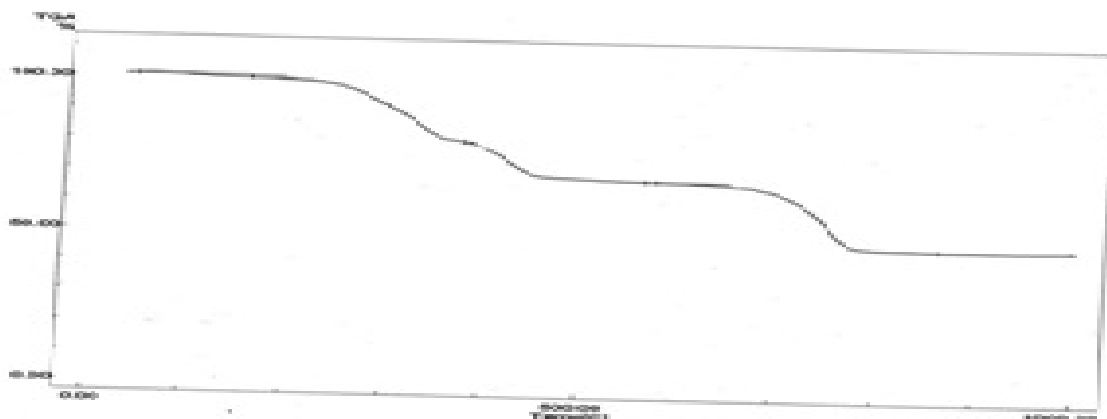
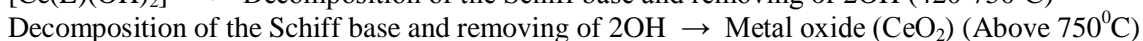
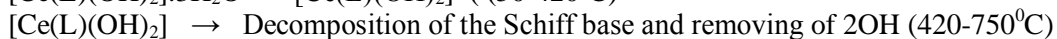


Figure 2: Thermogravimetric analysis data of the Ce(IV).

**3. Infrared spectra:** The significant IR bands of the Schiff base and its Zr(IV), La(III) and Ce(IV) chelates are presented in table 2 and figures (3-5). The IR spectra of the chelates show bands in the range of 1614-1643cm<sup>-1</sup> which could be assigned to -C=N vibration, this range is changed in comparison with its position in the 2-(2-[(2-hydroxyphenyl)ethanimidoyl]Phenyl-ethanimidoyl)phenol (1611cm<sup>-1</sup>) indicating its involvement in coordination<sup>13</sup>. The same spectra exhibit broad bands in the range of 3189- 3583 cm<sup>-1</sup> assigned to the existence of water molecules.<sup>14</sup> New bands observed at 609 - 650cm<sup>-1</sup> and 433-484cm<sup>-1</sup> which are not present in the 2-(2-[(2-hydroxyphenyl)ethanimidoyl]Phenylethanimidoyl)phenol spectrum can be attributed to ν(M-O) and ν(M-N) vibrations, respectively<sup>15</sup>. The spectral data of the [Zr(L)(H<sub>2</sub>O)<sub>2</sub>].H<sub>2</sub>O chelate display a band at 1028 cm<sup>-1</sup> assigned to ν(M=O) vibration suggesting the involvement of this group in complexation<sup>16</sup>.

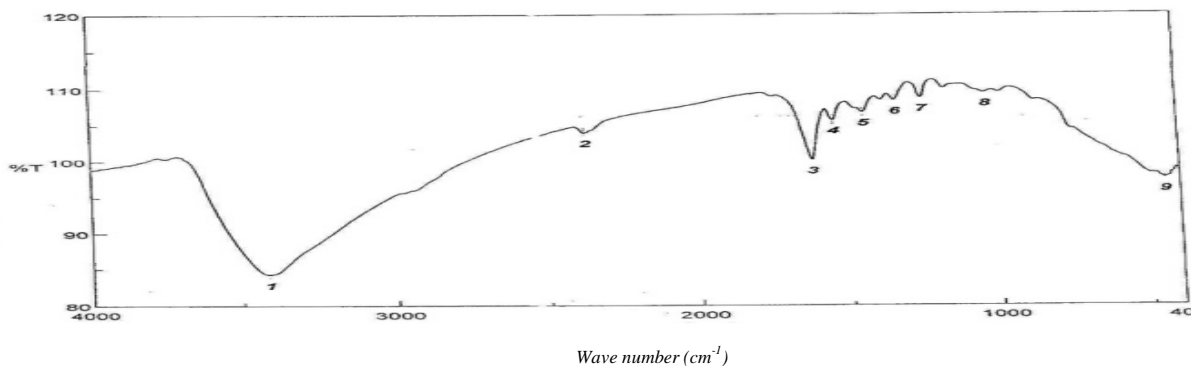


Figure 3: Infrared spectrum of Zr(IV) chelate.

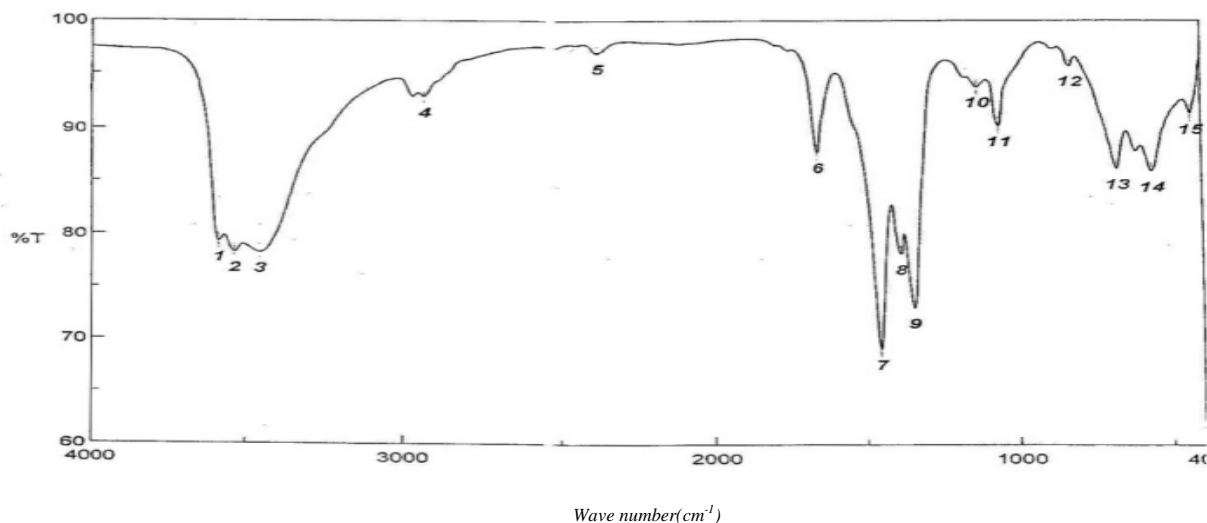


Figure 4: Infrared spectrum of La(III)-chelate.

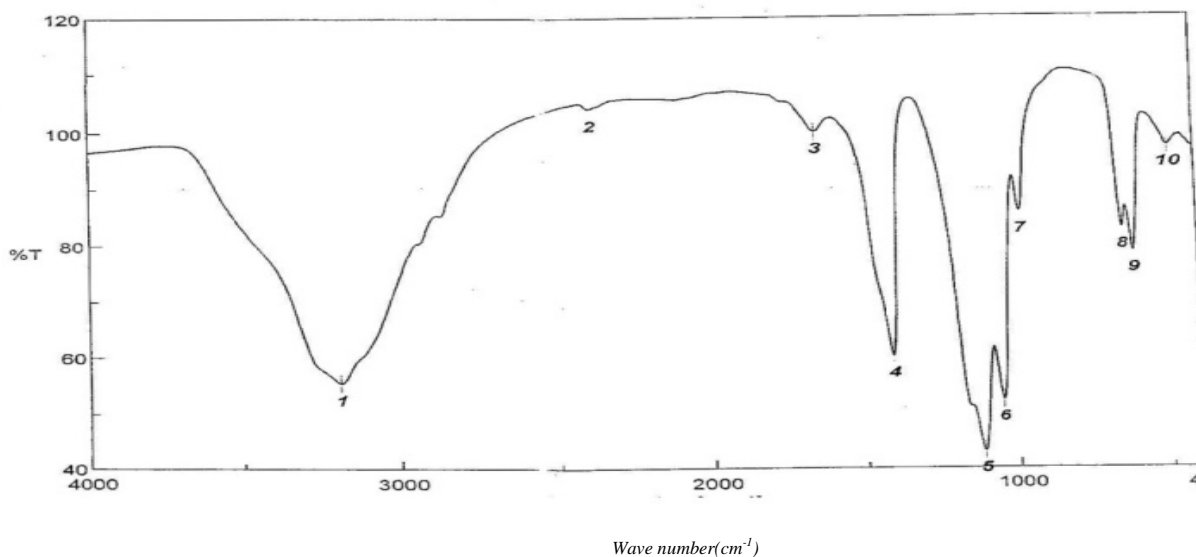


Figure 5: Infrared spectrum of Ce(IV)-chelate.

**4. Electronic spectra of the chelates:** The electronic spectral data of 2-(2-[(2-hydroxyphenyl) ethanimidoyl]Phenylethanimidoyl) phenol and its chelates were recorded in DMSO solvent and their assignments are shown in Table 2 and Figures (6-9). The infrared spectral results of the free Schiff base show two bands at 280 nm ( $35714\text{cm}^{-1}$ ) and 322nm ( $31055\text{cm}^{-1}$ ) characteristic of  $\pi\rightarrow\pi^*$  (Phenyl rings) and  $n\rightarrow\pi^*$  ( $-\text{C}=\text{N}$ ) transitions, respectively.<sup>17</sup> In the metal chelates, these bands are shifted to a longer wavelength with increasing the intensity. This shift attributed to the donation of lone pair of electrons of nitrogen of Schiff base to metal ion. The electronic absorption spectrum of Zr(IV) chelate in DMSO solvent showed one band at 323 nm ( $30959\text{cm}^{-1}$ ) assigned to the charge transfer transition which indicates an octahedral geometry of this chelate.<sup>18</sup> The La (III) and Ce(IV) chelates display the characteristic bands due to f-f transition. Thus;  $\text{La}^{3+}$  and  $\text{Ce}^{4+}$  have no transitions in this region because the f-f bands are sharp and line like. This is because of the effective shielding of the 4f orbital by the 5s, 5p octet and consequently minimum ligand field perturbation of the electronic energy levels in lanthanides and observed f-f transitions in 500–700 nm region<sup>19</sup>. The tentative assignments are given in Table 2.

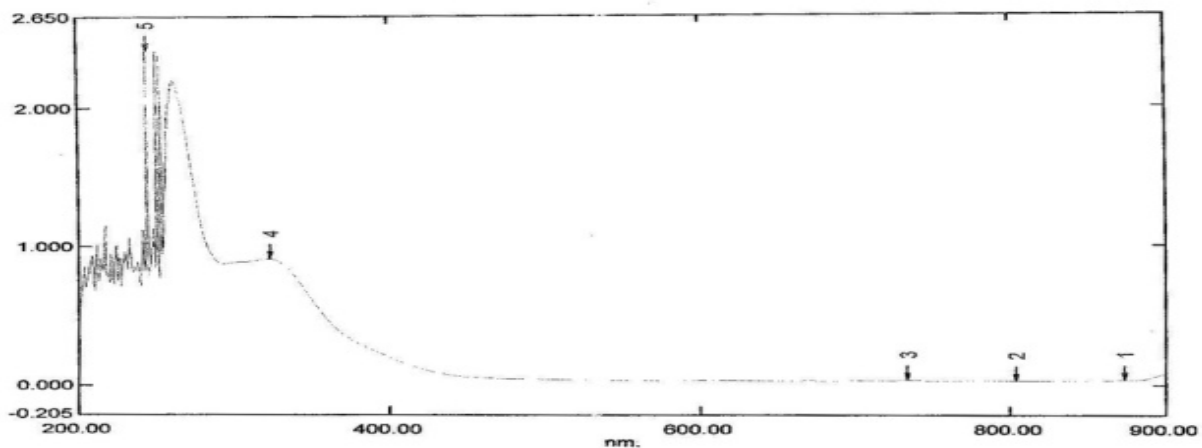


Figure 6: Ultraviolet spectrum of 2-(2-[(2-hydroxyphenyl) ethanimidoyl]- Phenylethanimidoyl) phenol.

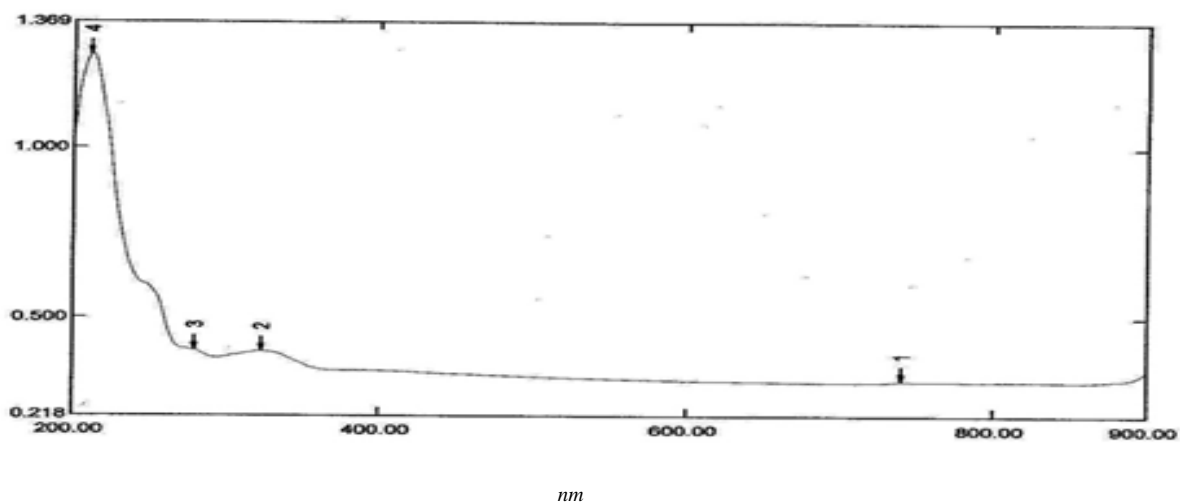


Figure 7: Electronic spectrum of Zr(IV) chelate.

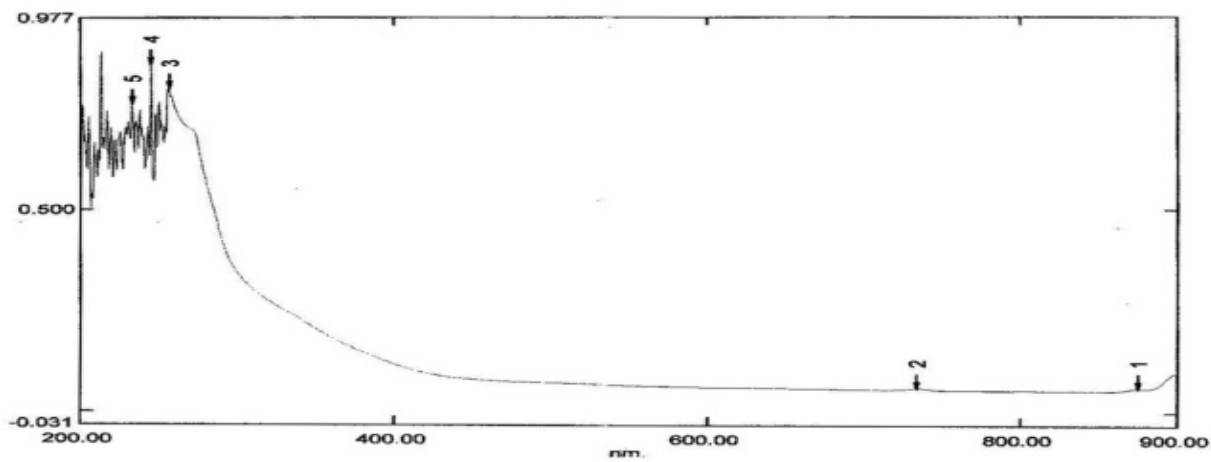


Figure 8: Electronic spectrum of La(III) chelate.

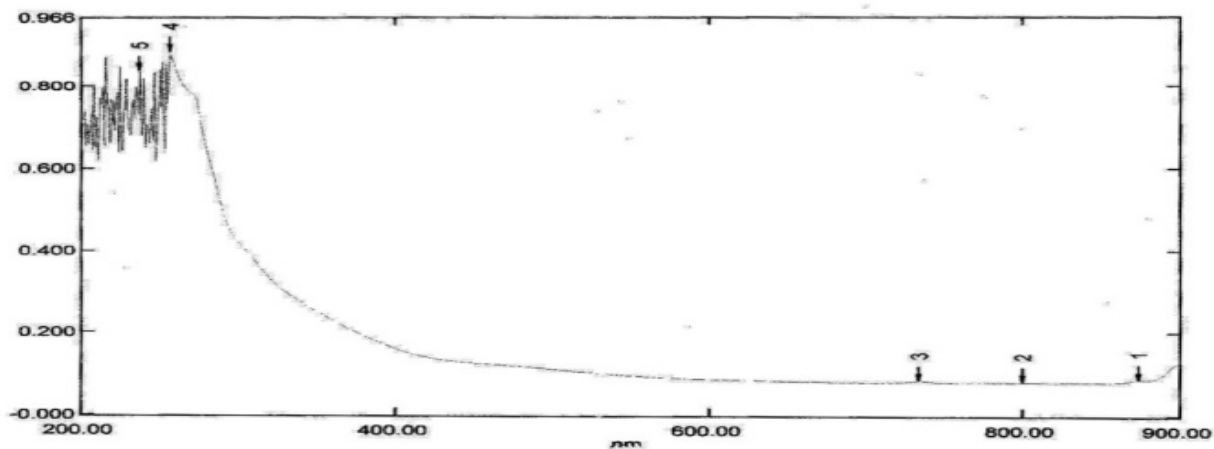


Figure 9: Electronic spectrum of Ce(IV) chelate.

Table 2: Infrared band ( $\text{cm}^{-1}$ ) and electronic spectral assignments (nm,  $\text{cm}^{-1}$ ) of Schiff base (SB) and its chelates.

Schiff base/ Chelate	$\nu(\text{OH})$	$\nu(\text{C}=\text{N})$	$\nu(\text{CH}_3)$	$\nu(\text{CH})$	$\nu(\text{M}-\text{O})$	$\nu(\text{M}-\text{N})$	nm( $\text{cm}^{-1}$ )
SB	3450	1611	1360	746	-	-	280(35714), 322(31055)
[Zr(L) OH <sub>2</sub> ] <sub>2</sub> H <sub>2</sub> O	3415	1614	1328	753	626	454	323(30959)
[La(L)(OH)(OH <sub>2</sub> )] 4H <sub>2</sub> O	3583	1643	1340	821	650	433	-
[Ce(L)(OH) <sub>2</sub> ] 3H <sub>2</sub> O	3189	1631	1405	767	609	484	-

5. <sup>1</sup>HNMR spectra of the Zr(IV) chelates: The <sup>1</sup>HNMR spectrum of free Schiff base and its Zr(IV) chelate are recorded in d<sub>6</sub> DMSO solvent at room temperature. The <sup>1</sup>HNMR spectral data of the 2-(2-[(2-hydroxyphenyl)ethanimidoyl]Phenyl-ethanimidoyl)phenol show singlet at 7.768 ppm, which is due to protons bonded to azomethine group. The methyl group signal appeared at 3.307ppm. The signals in the range of 6.903-7.412 ppm are due to phenyl protons. Phenolic proton shows singlet at 14.971 ppm, attributing to the -OH group<sup>20</sup>. In the Zr(IV) chelate, the phenolic protons were absent, confirming subsequent involvement of deprotonated hydroxyls group in chelation to the metal ions. Whereas; the saturated alkyl protons of the Schiff base were equally observed at signals of 2.592 (3H) and 2.638 (3H) ppm<sup>21</sup>.

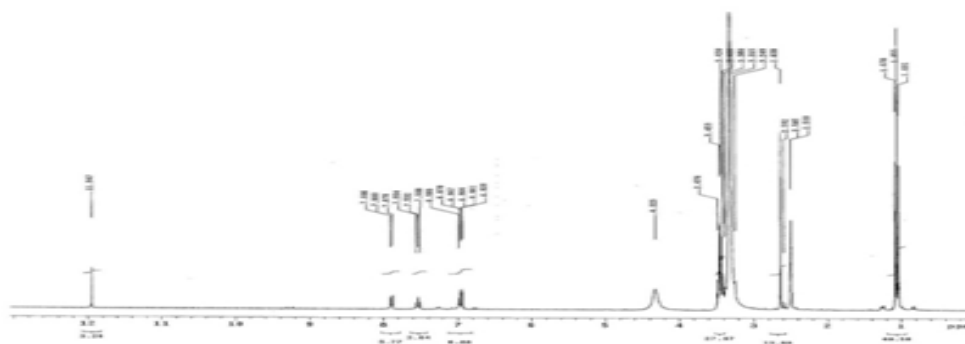
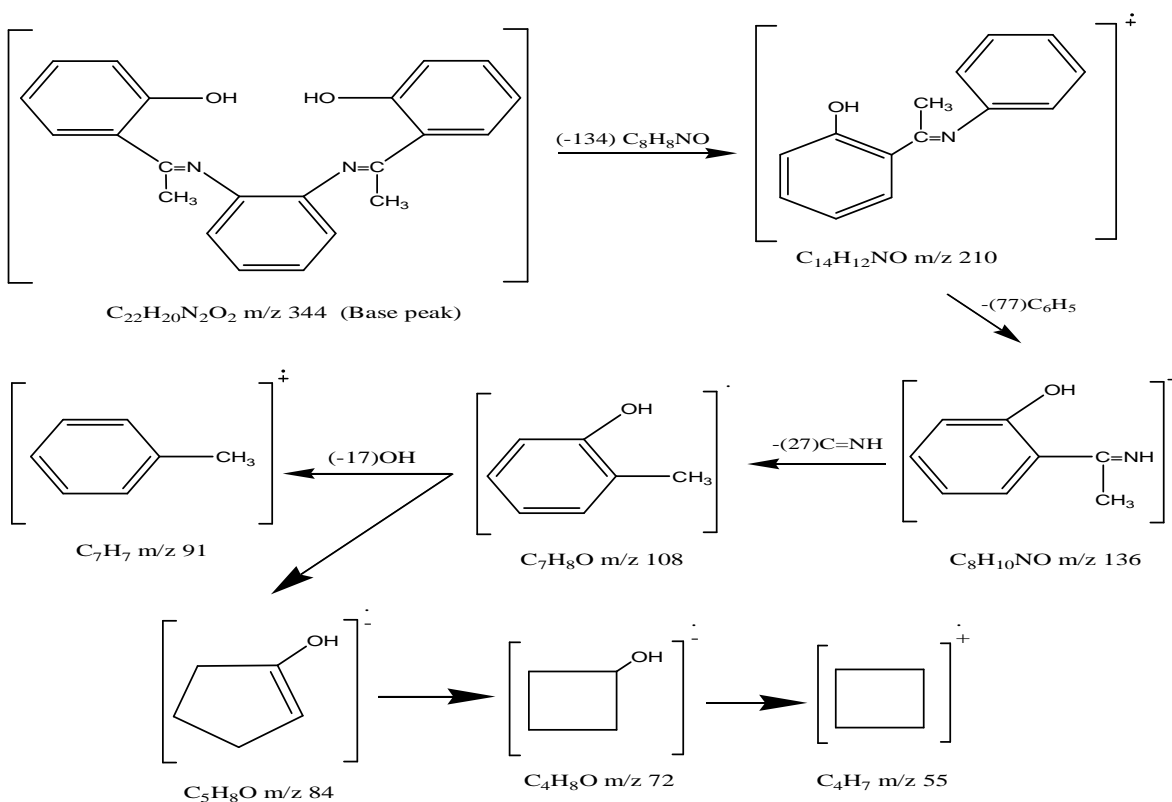


Figure 10: Proton nuclear magnetic resonance spectrum of Zr(IV) chelate.

**6. Mass spectra of Schiff base and its chelates:** The electron impact mass spectrum of Schiff base is recorded and investigated at 70 eV of electron energy. The mass spectrum of the Schiff base under investigation is shown in schemes (1-4) and figures (11-14). The mass spectrum of Schiff base shows base peak at  $m/z = 344$  due to the original molecular weight of the free Schiff base. The mass spectrum of Schiff base exhibits peaks at  $m/z$  values; 210 and 136 suggesting different fragmentations and the same spectrum displays peaks at values;  $m/z = 108, 91, 84$  and  $72$  attributed to loss of different atoms (scheme 1). Whereas, the same spectrum shows a peak at  $m/z = 71$  is assigned to  $C_5H_{10}$ . The last peak at  $m/z = 55$  is attributed to  $C_4H_7^+$  ion. The mass spectrum of the  $[Zr(L)(H_2O)_2].H_2O$  chelate exhibits a characteristic molecular ion ( $M^+$ ) peak at  $m/z = 504$ , which is corresponding to the molecular weight of the chelate. The present mass spectrum shows multiple peaks representing successive degradation of the chelate molecule by the formation of different peaks. The peaks have been observed at  $m/z$  values; 450, 118 and 77 suggesting the proper fragmentations (scheme 2). A peak at  $m/z = 64$  is analogous to  $C_5H_4$  ion. The mass spectrum of  $[La(L)(OH)(OH_2)].4H_2O$  chelate has been recorded. The spectrum of the La(III) chelate exhibits the molecular ion ( $M^+$ ) peak at  $m/z = 588$  due to its original molecular weight. The peak at  $m/z = 498$  analogous to the composition of  $[La(L)(OH)]$ . The same spectrum shows a peak at  $m/z = 209$  analogous to the loss of  $C_{14}H_{11}NO^+$  molecule from chemical formula of the chelate, the final peaks at  $m/z = 90$  and  $77$  are assigned to the appearance of  $C_7H_6$  and  $C_6H_5$ , respectively. The final peak at  $m/z = 55$  is analogous to  $C_4H_6$  ion. For  $[Ce(L)(OH)_2].3H_2O$  chelate, the spectrum exhibits a peak at  $m/z = 571$  due to the original molecular weight of the chelate. The peak at  $m/z = 515$  corresponds to  $[Ce(L)(OH)_2]$  ion and other peak at  $m/z = 133$  attributed to  $C_8H_8NO$  ion. The present spectrum displays several peaks at  $m/z = 103, 89$  and  $64$  suggesting existence of different ion species after the successive fragmentation of different groups (see scheme 4). The following fragmentations illustrate the formation of the Schiff base and the chelates in 1:1 [M:L] ratio.



**Scheme 1: Mass spectral fragmentation of 2-(2-[(2-hydroxyphenyl) ethanimidoyl]Phenyl-ethanimidoyl)phenol**



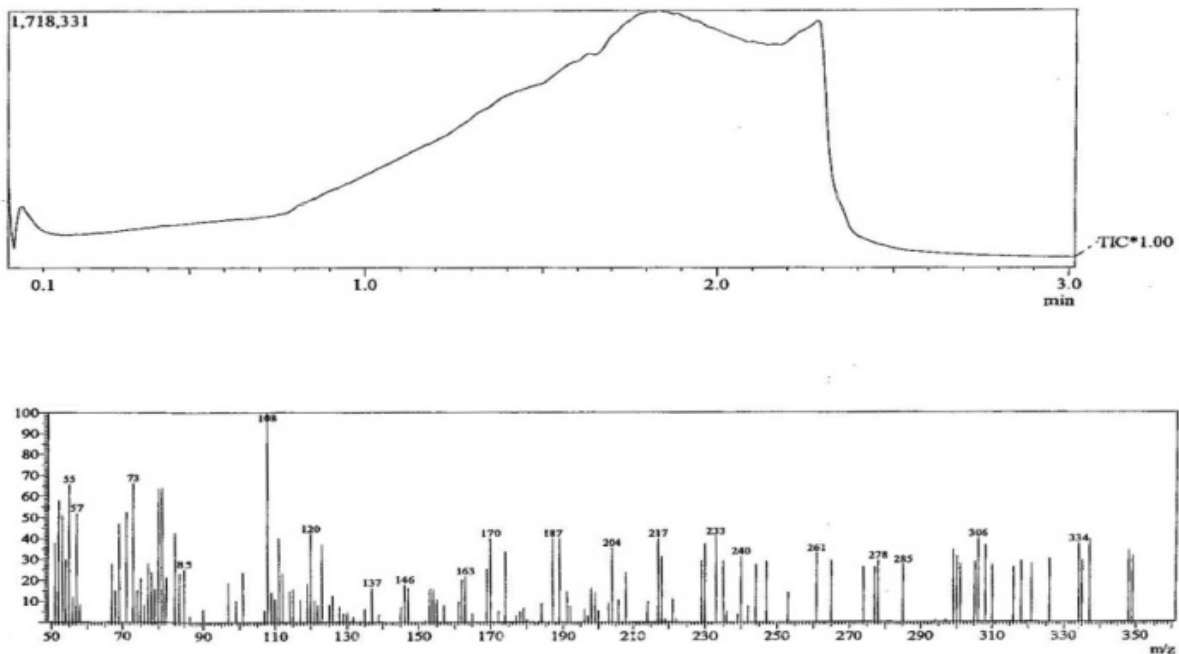
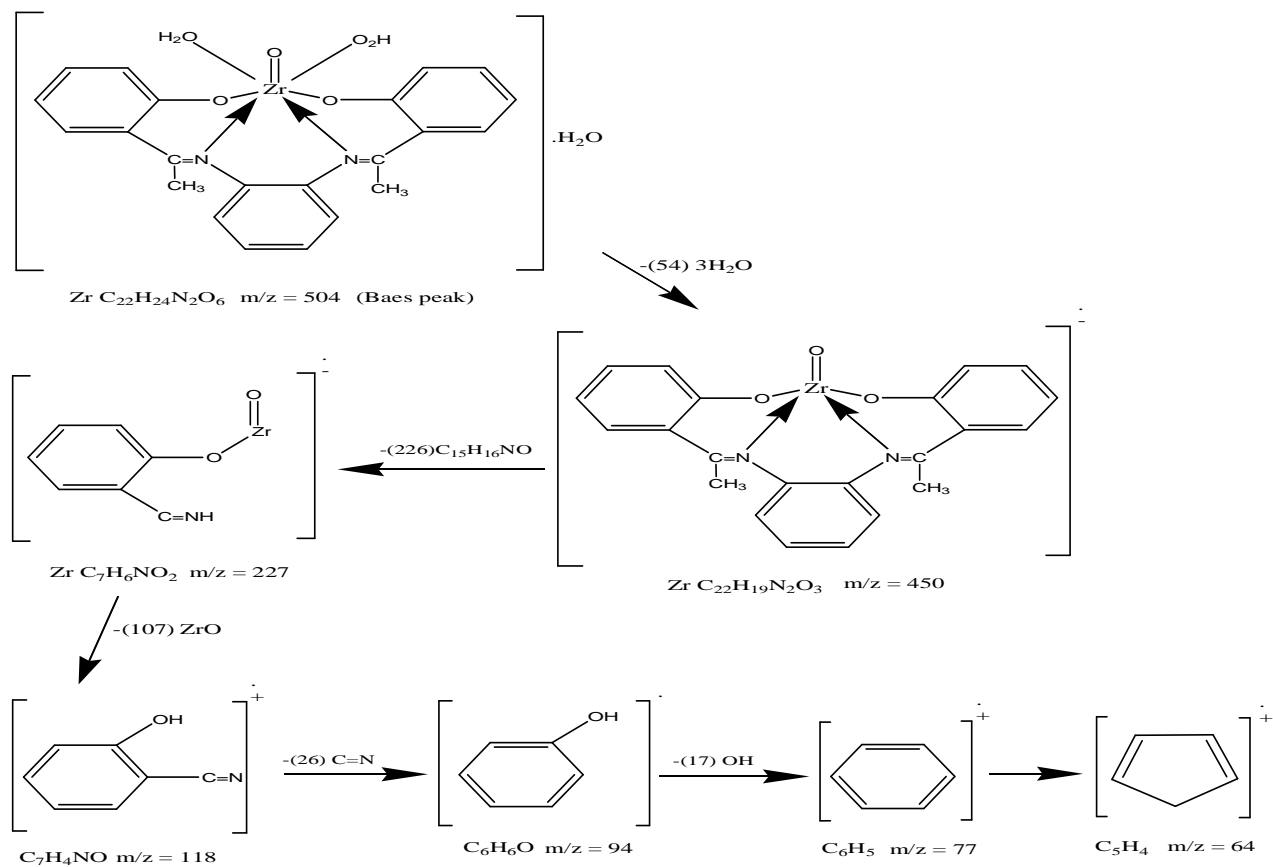


Figure 11: Mass spectrum of 2-(2-[(2-hydroxyphenyl) ethanimidoyl]Phenylethanimidoyl)phenol.



Scheme 2: Mass spectral fragmentations of the Zr(IV)-chelate.

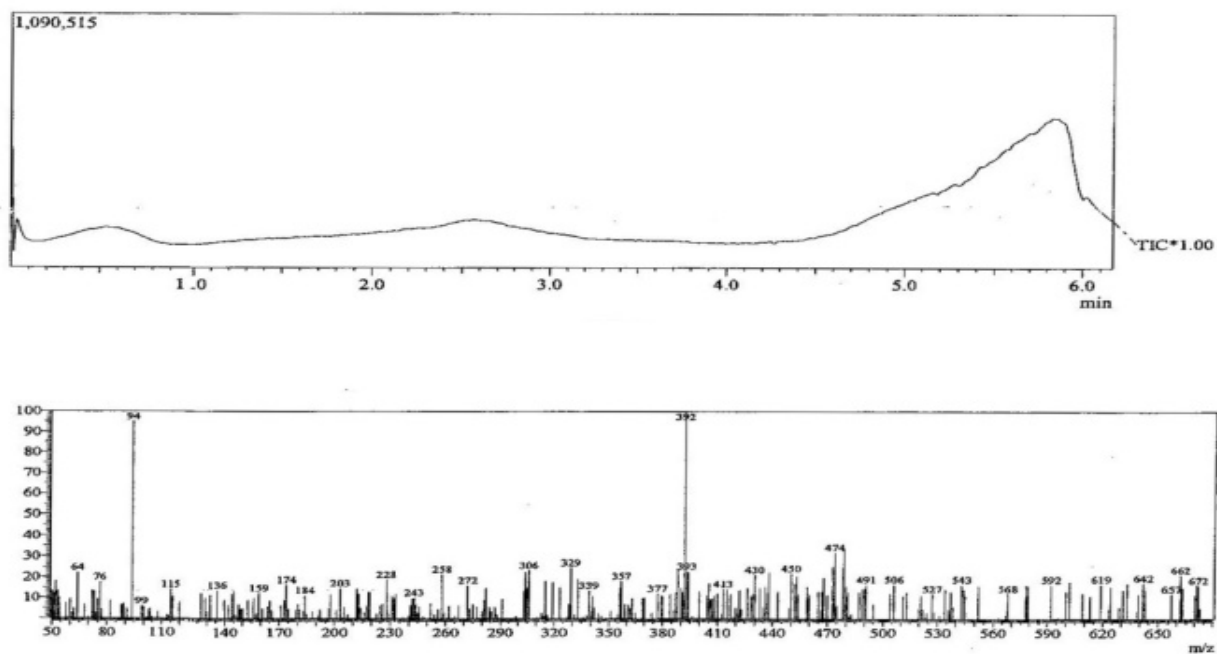
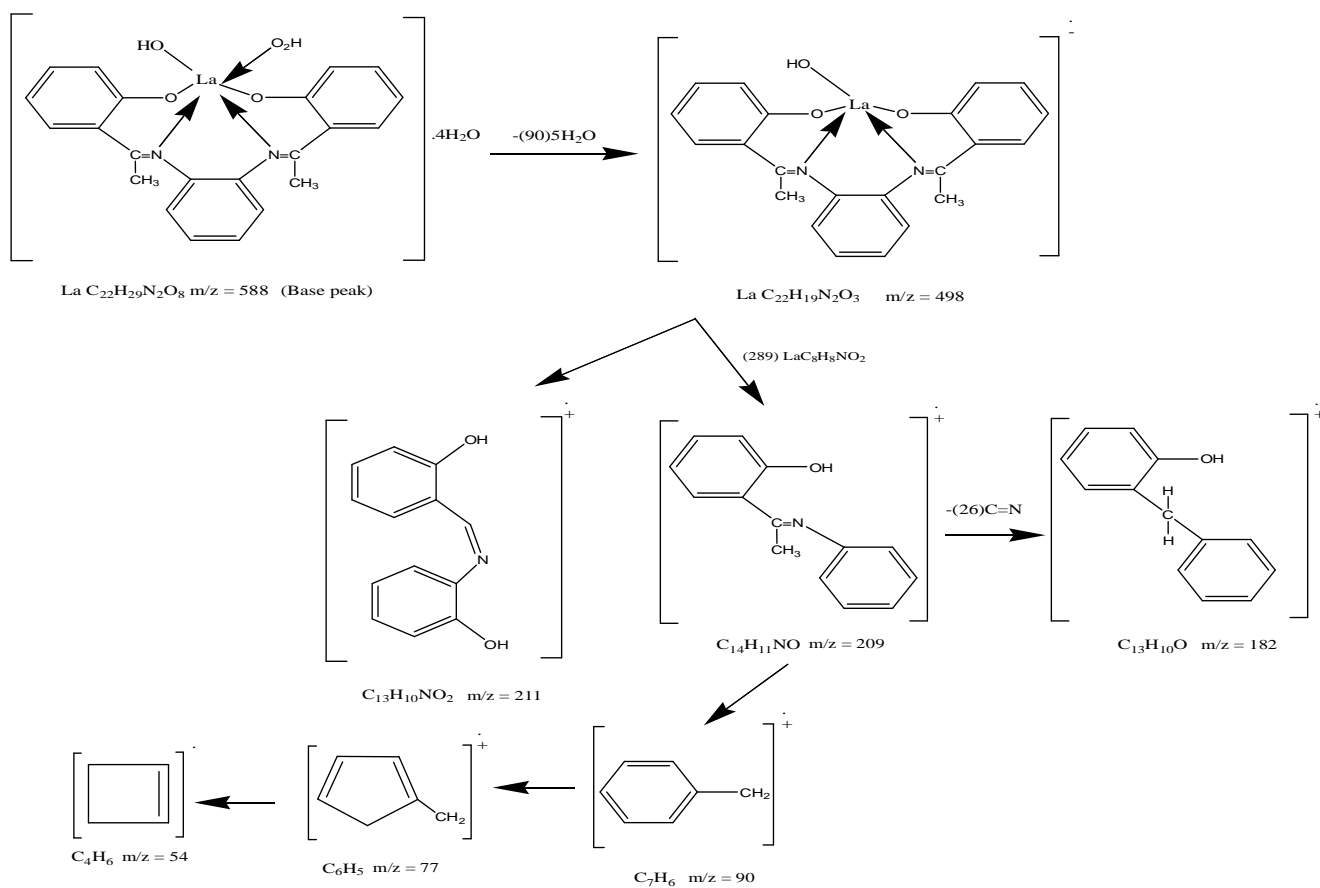


Figure 12: Mass spectrum of the Zr(IV)-chelate.



Scheme 3: Mass spectral fragmentations of the La(III) chelate.

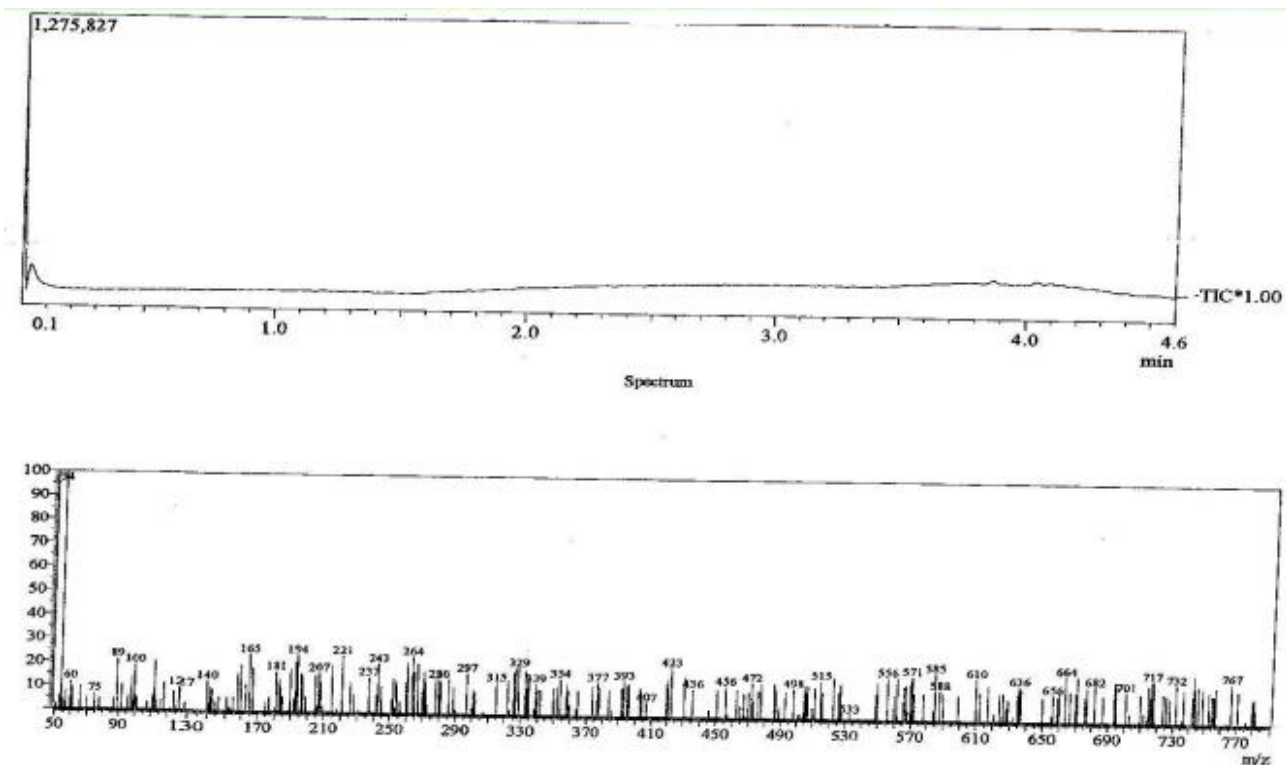
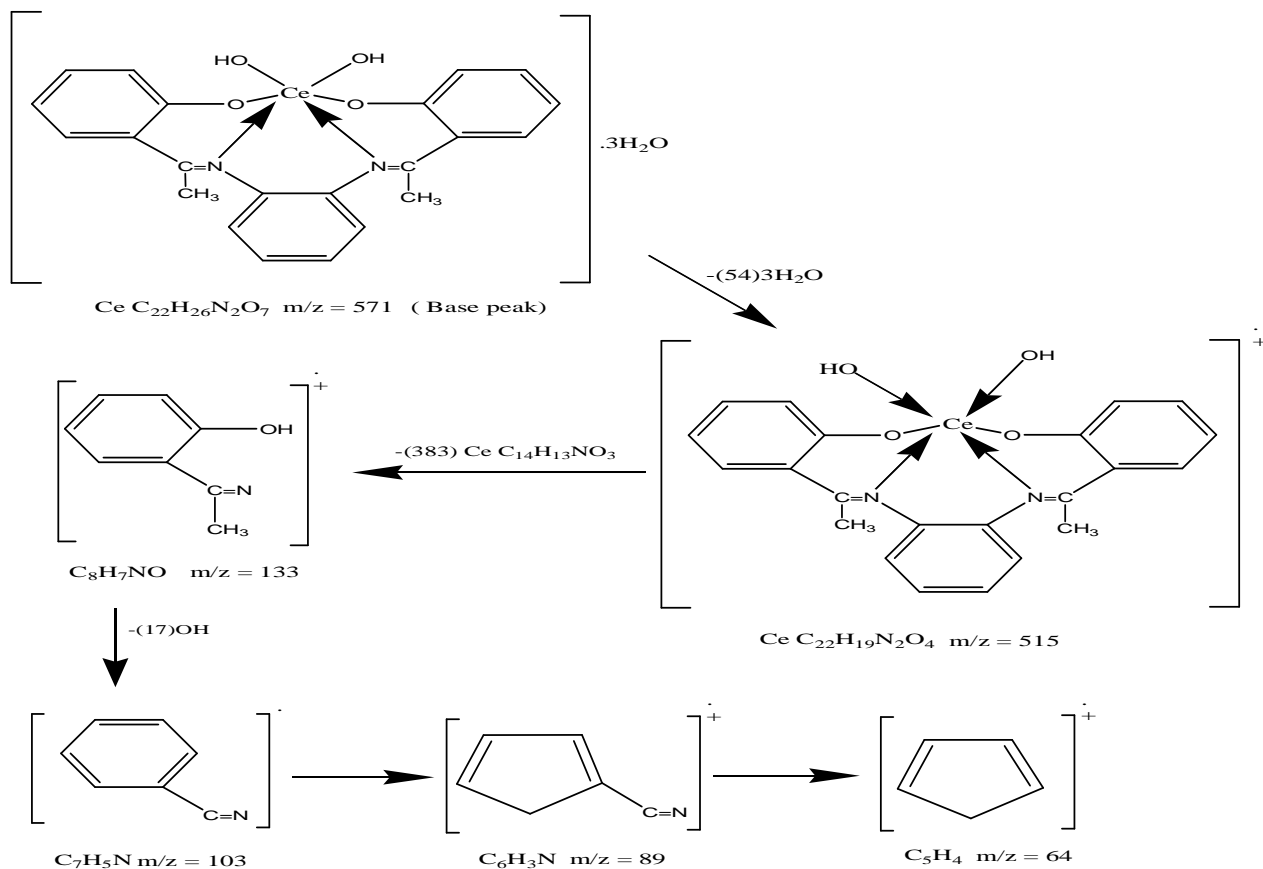


Figure 13: Mass spectrum of the La(III) chelate.



Scheme 4: Mass spectral fragmentations of the Ce(IV) chelate.

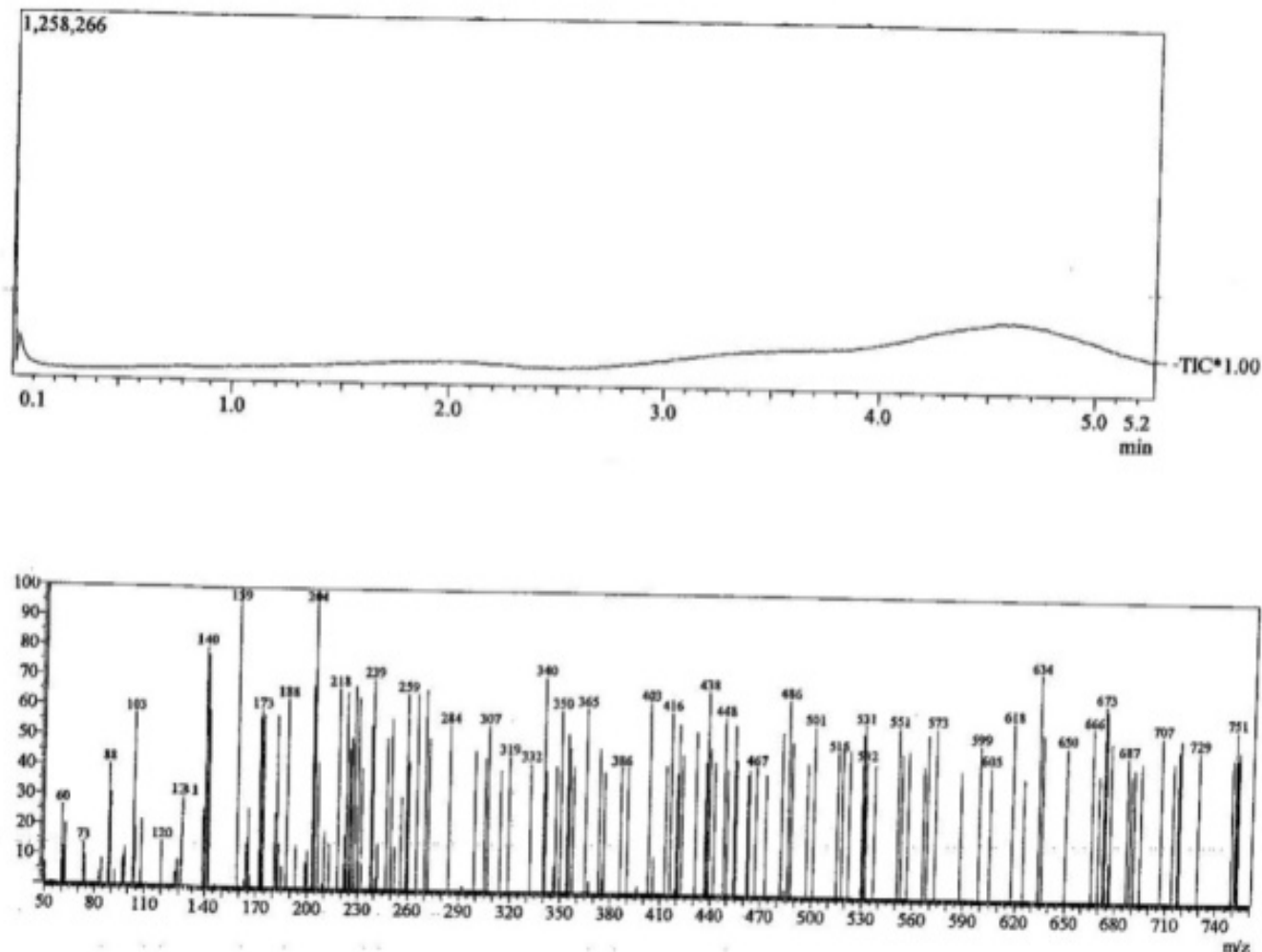


Figure 14: Mass spectrum of the Ce(IV) chelate.

7. **Antimicrobial Activity of the Schiff base and its chelates:** The *in vitro* antimicrobial screenings of the free Schiff base, metal precursors and its chelates were tested for their effect on certain bacteria and fungus (Tables 3-5). It is observed that the synthesized Schiff base and metal chelates exhibited particularly effective against the tested microorganisms. This observation can be assumed as selective activity. The compound 2C and 3C are effective on two different microorganisms: one of them is *Streptococcus faecalis* type Gram positive bacteria and the other one is *E. coli* type Gram negative bacteria. Antibacterial activity of the compound 2C is better than antibacterial activity of 3C. On the other hand, the synthesized metal- chelate has no any antifungal activity against unicellular fungi represented by *Fusarium solani* and filamentous fungi represented by *Aspergillus niger*. The antimicrobial activity of the metal chelates can be explained on the basis of overtone's concept<sup>22</sup>, and chelation theory. According to overtone's concept of cell permeability, the lipid membrane that surrounds the cell favors the passage of only lipid soluble materials due to which lip solubility has an important factor which controls the antimicrobial activity. On chelation, the polarity of the metal atom will be reduced to a greater extent due to the overlap of the ligand orbital and partial sharing of positive charge of metal atom with donor groups. Further, it increases the delocalization of  $\pi$  electrons over the whole chelate ring and enhances the lipophilicity which enhances the penetration of the chelates. This increased lipophilicity enhances the penetration of the chelates into lipid membrane and blocking the metal binding-sites on enzymes of microorganisms<sup>23 & 24</sup>.

**Table 4: Antibacterial activities of used materials, Schiff base and its chelates on growth of microorganisms.**

S.No.	Comp.	Diameter of inhibition zone (mm); Concentration in µg/ml											
		Gram negative organisms											
		<i>Escherichia coli</i>						<i>Salmonella kentucky</i>					
		% Activity index						% Activity index					
5 µg/ml	50 µg/ml	500 µg/ml	5 µg/ml	50 µg/ml	500 µg/ml	5 µg/ml	50 µg/ml	500 µg/ml	5 µg/ml	50 µg/ml	500 µg/ml		
1B	HAPH	0	0	0	0	0	0	0	0	0	0	0	
2B	OPD	0	0	11	0	0	31	0	0	17	0	0	43
3B	ZrOCl <sub>2</sub> . 8H <sub>2</sub> O	0	0	0	0	0	0	0	0	0	0	0	
4B	La(NO <sub>3</sub> ) <sub>3</sub> .6H <sub>2</sub> O	8	13	18	20	33	45	8	11	16	23	31	46
5B	Ce(SO <sub>4</sub> ) <sub>2</sub> .4H <sub>2</sub> O	8	10	13	23	29	37	0	0	8	0	0	20
1C	SB	0	0	0	0	0	0	0	0	0	0	0	0
2C	La(III) chelate	0	8	10	0	20	50	0	0	0	0	0	0
3C	Zr(IV) chelate	0	0	11	0	0	28	0	0	0	0	0	0
4C	Ce(IV) chelate	0	0	10	0	0	25	0	0	0	0	0	0
C	DMSO (Control)	0	0	0	0	0	0	0	0	0	0	0	0
CIP	Ciprofloxacin (Standard)	40	40	40	100	100	100	35	35	35	100	100	100

HAPH(*o*-hydroxyacetophenone), OPD(1,2-diaminobenzene).

$$\%I = C-T/C \times 100 \text{ ----- Eq}$$

**Table 5: Antibacterial activities of used materials, Schiff base and its chelates on growth of microorganisms.**

S.No.	Comp.	Diameter of inhibition zone (mm); Concentration in µg/ml											
		Gram positive organisms											
		<i>Lactobacillus fermentum</i>						<i>Streptococcus faecalis</i>					
		% Activity index						% Activity index					
5 µg/ml	50 µg/ml	500 µg/ml	5 µg/ml	50 µg/ml	500 µg/ml	5 µg/ml	50 µg/ml	500 µg/ml	5 µg/ml	50 µg/ml	500 µg/ml		
1B	HAPH	0	1	2	0	3	6	5	8	10	13	20	25
2B	OPD	0	0	0	0	0	0	0	0	22	0	0	55
3B	ZrOCl <sub>2</sub> . 8H <sub>2</sub> O	0	7	15	0	20	43	0	0	16	0	0	40
4B	La(NO <sub>3</sub> ) <sub>3</sub> .6H <sub>2</sub> O	0	0	0	0	0	0	0	0	16	0	0	40
5B	Ce(SO <sub>4</sub> ) <sub>2</sub> .4H <sub>2</sub> O	8	13	16	23	37	46	0	9	15	0	23	38
1C	SB	0	0	0	0	0	0	0	0	8	0	0	20
2C	La(III) chelate	0	0	0	0	0	0	0	0	0	0	0	0
3C	Zr(IV) chelate	0	0	15	0	0	43	0	0	0	0	0	0
4C	Ce(IV) chelate	0	0	0	0	0	0	0	0	0	0	0	0
C	DMSO (Control)	0	0	0	0	0	0	0	0	0	0	0	0
CIP	Ciprofloxacin (Standard)	35	35	35	100	100	100	40	40	40	100	100	100

HAPH(*o*-hydroxyacetophenone), OPD(1,2-diaminobenzene).

$$\%I = C-T/C \times 100 \text{ ----- Eq}$$

**Table 6: Antifungal activities of used materials, Schiff base and its chelates on growth of microorganisms.**

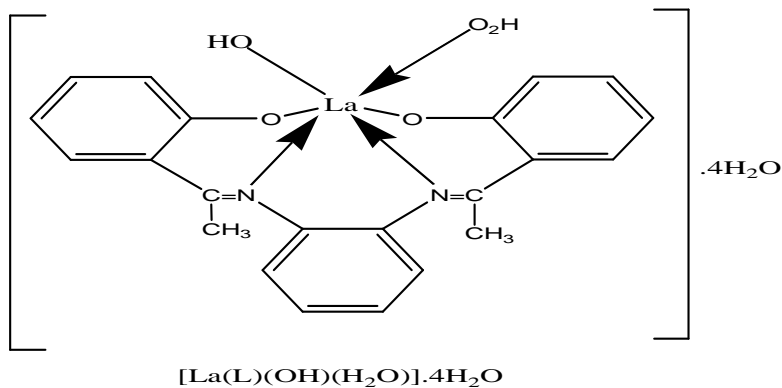
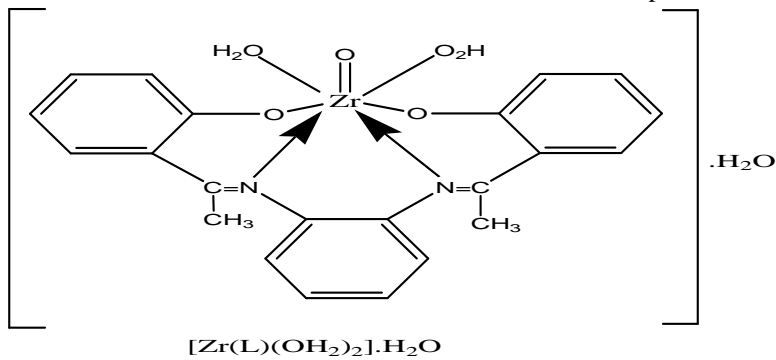
S.No.	Comp.	Diameter of inhibition zone (mm); Concentration in $\mu\text{g/ml}$											
		Fungi											
		<i>Aspergillus niger</i>						<i>Fusarium solani</i>					
		% Activity index						% Activity index					
		5 $\mu\text{g/ml}$	50 $\mu\text{g/ml}$	500 $\mu\text{g/ml}$	5 $\mu\text{g/ml}$	50 $\mu\text{g/ml}$	500 $\mu\text{g/ml}$	5 $\mu\text{g/ml}$	50 $\mu\text{g/ml}$	500 $\mu\text{g/ml}$	5 $\mu\text{g/ml}$	50 $\mu\text{g/ml}$	500 $\mu\text{g/ml}$
1B	HAPH	0	0	19	0	0	90	0	0	0	0	0	0
2B	OPD	0	0	11	0	0	52	0	0	11	0	0	85
3B	ZrOCl <sub>2</sub> . 8H <sub>2</sub> O	0	0	4	0	0	19	0	0	0	0	0	0
4B	La(NO <sub>3</sub> ) <sub>3</sub> .6H <sub>2</sub> O	0	0	3	0	0	14	0	0	0	0	0	0
5B	Ce(SO <sub>4</sub> ) <sub>2</sub> .4H <sub>2</sub> O	0	1	8	0	5	38	0	0	0	0	0	0
1C	SB	0	0	3	0	0	14	0	0	0	0	0	0
2C	La(III) chelate	0	0	0	0	0	0	0	0	0	0	0	0
3C	Zr(IV) chelate	0	0	0	0	0	0	0	0	0	0	0	0
4C	Ce(IV) chelate	0	0	0	0	0	0	0	0	0	0	0	0
C	DMSO (Control)	0	0	0	0	0	0	0	0	0	0	0	0
MC Z	Miconazole (Standard)	21	21	21	100	100	100	13	13	13	100	100	100

HAPH(*o*-hydroxyacetophenone), OPD(1,2-diaminobenzene).

$$\%I = C-T/C \times 100 \text{ ----- Eq}$$

### CONCLUSION

The Schiff base and its chelates were synthesized and characterized by several techniques which mentioned above, the obtained data confirmed that all chelates have an octahedral structure as shown below. In addition to the chemistry part of Schiff base and its chelates, antimicrobial activities of these compounds were also studied.



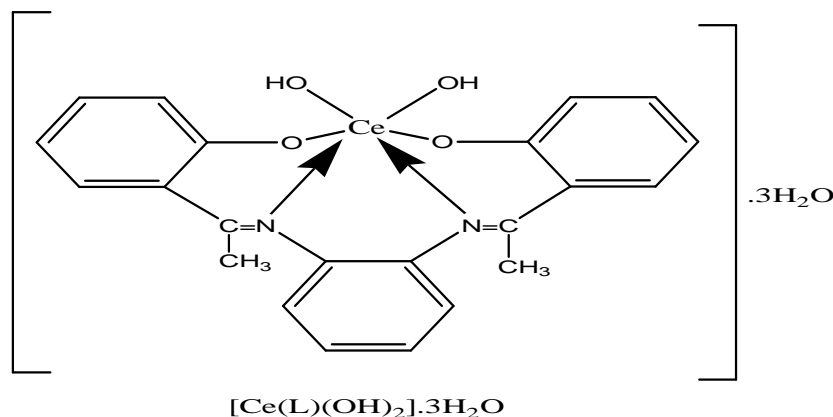


Figure 15: Geometrical structures of Schiff base chelates.

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