

## RESEARCH PAPER

### Synthesis and characterization of Ni(II) complexes with ester semicarbazone

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#### ABSTRACT

General composition of Ni(II) complexes [ML<sub>2</sub>X<sub>2</sub>], [ML<sub>2</sub>X]X were prepared with semicarbazones (L<sup>1</sup>, L<sup>2</sup>, L<sup>3</sup> and L<sup>4</sup>). These complexes were characterized by elemental analysis, molar conductances measurements, Magnetic moments IR, electronic spectra, and EPR spectral studies. All are the nonelectrolyte in nature therefore these complexes may formulated [M(L)<sub>2</sub>X<sub>2</sub>]. All the complexes are of high-spin and show octahedral geometry.

**Key Words :** Acetoacetic ester semicarbazone, Isopropyl semicarbazone, 6-methyl Pyran-2-one-4 hydroxy 3 diacarboxylic acid, Semicarbazone, Biological activity

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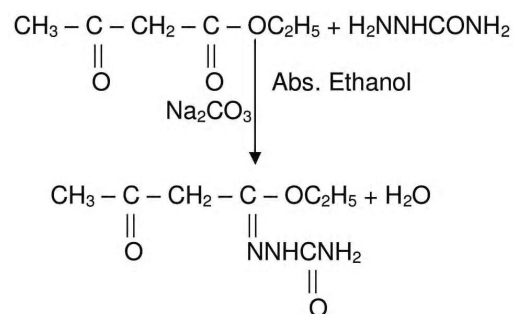
**T**he biological and medicinal properties of these ligands and their derivatives have gained much interest. Semicarbazones and their 3d-metal complexes have been found to exhibit anti-fungal[1], anti-bacterial[2], anti-viral[3], anti-tubercular[4] and anti carcinogenic activities [5]. The anti-fungal activity of these compounds is due to the presence of toxophyrically important N-C=O moiety[6]. Semicarbazides and their Schiff bases also display anti-tumour [7-8] activity. It is expected that ligands will also show variability in structure and bonding in its transition metal complexes. It has been reported that semicarbazide and its complexes with 3d-metal ions show *in vitro* and *in vivo* anti-tumour activity[9].

isopropyl ester, methyl ester of 6-methyl Pyran-2-one-4 hydroxy 3 diacarboxylic acid, sodium acetate and different metallic salts.

#### Preparation of ligands :

##### Preparation of Acetoacetic ester semicarbazone (AESCL<sup>1</sup>):

Aqueous solution of semicarbazide hydrochloride (.01 mol, 1.12 g) and acetoacetic ester (0.01 mol. of 1.83 ml) were



#### RESEARCH METHODOLOGY

A.R. Grade chemical and fluka reagents were used in the present study. The solvents were purified before use by processing. Semicarbazide hydrochloride, acetoacetic ester,

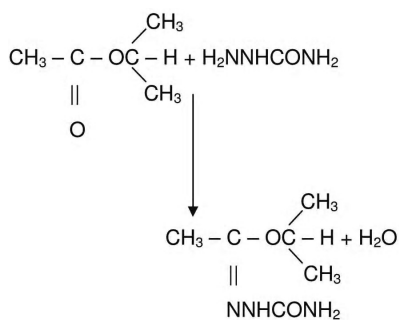
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mixed in the presence of sodium acetate (.01 mol, 1.36 g). This solution was stirred with the help of mechanical stirrer for an hour. On cooling a white product was formed, filtered, washed with cold ethanol and dried under vacuum over  $P_4O_{10}$  (mt.  $180^\circ C$  yield 68%).

#### Preparation of isopropyl ester semicarbazone (IESC)(L<sup>2</sup>):

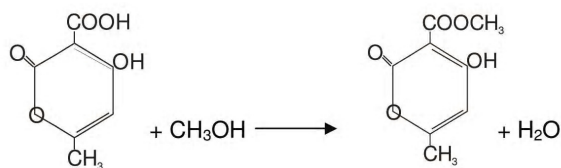
IESC was prepared by the condensation of isopropyl acetate (0.01 mol, 0.849 ml) with semicarbazide (0.01 mol, 1.12 g) in absolute ethanol. It was purified and recrystallized from the same solvent and dried under vacuum over  $P_4O_{10}$ . The purity was checked by T.L.C. (mp  $185^\circ C$ , yield 80%).



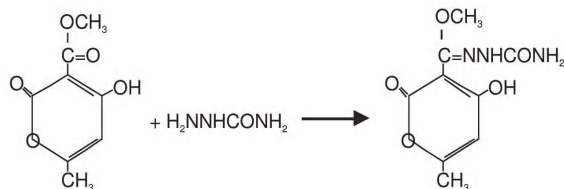
#### Preparation of semicarbazone of 6-methyl Pyran-2-one-4 hydroxy 3 dicarboxylic acid (L<sup>3</sup>):

The starting materials 6-methyl Pyran-2-one-4 hydroxy 3 dicarboxylic acid and semicarbazide are fluka reagents. The ligands was prepared in two steps :

**Step 1 :** 6-methyl Pyran-2-one-4 hydroxy 3 dicarboxylic acid (0.01 mol, 1.70 g) was refluxed with methanol (.01 mol, 0.25 ml) in the presence of 10% sulphuric acid. The ester of 6-methyl Pyran-2-one-4 hydroxy 3 dicarboxylic acid formed.



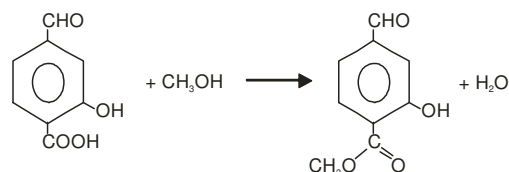
**Step 2 :** The methyl ester of 6-methyl Pyran-2-one-4 hydroxy 3 dicarboxylic acid (.01 mol, 1.84 g) and semicarbazide (.01mol, 1.12 g) were refluxed in ethanol for 1½hr. On cooling yellow colored compound was formed which was filtered, washed with ethanol, dried, recrystallized and under vacuum over  $P_4O_{10}$  (mp  $190^\circ C$  and yield 70%).



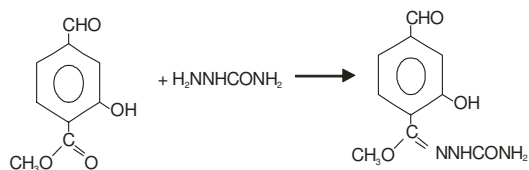
#### Preparation of semicarbazone of 3-formyl methyl salicylate (L<sup>4</sup>):

The ligand was prepared in two steps :

**Step 1 :** The 3-formyl salicylic acid (.01, 1.66g) was refluxed with methanol in the presence of 10%  $\text{H}_2\text{SO}_4$  for 2 hr a light yellow coloured precipitate of 3-formyl methyl salicylate was formed.



**Step 2 :** The 3-formyl methyl salicylate (.01mol 1.80g) and semicarbazide (.01mol 1.12g) in 15 ml of ethanol were refluxed for two hours . After cooling, white solid was separated out. It was filtered, washed with water and dried under vacuum over  $P_4O_{10}$ .



#### Characterization of ligands:

##### Elemental analysis:

The ligands were found to have the composition as shown in Table 1.

Mass spectra [10-14] : Electron impact mass spectra of the ligands were recorded on TOF MS ES+ Mass spectrometer from University of Delhi.

Mass spectrum is a presentation of the masses of the positively charged fragments (including the molecular ion) versus their relative concentrations. The more intense peak in the spectrum, called the base peak, is assigned a value of 100% and the intensities of other peaks, including the molecular ion peak are reported as percentages of the base peak. The molecular ion peak may some times the base peak. The molecular ion peaks is usually the peak of highest mass number for the isotopic peaks. The intensities of the isotope peaks relative to parent peak can lead to the determination of a molecular formula. The measured isotope peaks are usually slightly higher than the calculated contribution because of incomplete resolution, bimolecular couisions or a contribution from the incident peak of ion impurity of the proposed ligands.

##### Mass spectrum of the ligands :

The mass spectrum of ligand L<sup>1</sup> shows the molecular ion peak ( $M^+$ ) at  $m/z = 203$  amu. The different ions gives the peaks

of different mass like 160, 146, 101, 75, 60 and 16. The intensities of these give the idea of stability of ligands

The mass spectrum of Ligand L<sup>2</sup> shows that molecular ion peak (M<sup>+</sup>) at m/z=175 amu. The other peaks at 160, 148, 101, 75, 60 and 16 amu. Corresponds to various fragments show the stability of ligand.

The mass spectrum of Ligand L<sup>3</sup> shows the molecular ion peak (M<sup>+</sup>) at m/z=259 amu. The other different ions give peaks of 132, 101, 75, 60 and 16 amu. The intensities of peaks are in correspondance with the abundance of the ions.

The mass spectrum of Ligands L<sup>4</sup> shows the molecular ion peak (M<sup>+</sup>) at m/z=259 amu. The other different ions give the peaks of different mass numbers like 132, 101, 75, 60 and 16 amu. The intensities of these peaks give the idea of stability of ligand.

### I.R. Spectra[15-32] :

The important IR bands and their assignments are given in Table 2. In the present study in IR spectra of ligands the bands appeared at 1570-1592 cm<sup>-1</sup> and 1672-1695 cm<sup>-1</sup> due to n(C=N) and n(C=O) groups respectively.

L<sup>1</sup> ⇒ Nitrogen atom of n(C=N) group and oxygen of n(C=O) group ⇒ Bidentate

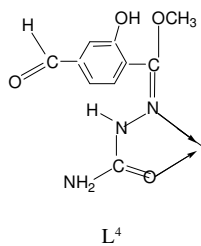
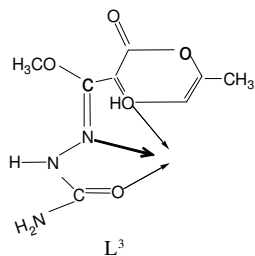
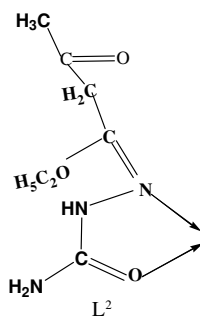
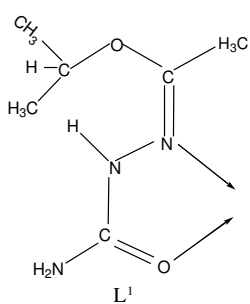
L<sup>2</sup> ⇒ Nitrogen atom n(C=N) group and oxygen of n(C=O) group Bidentate

L<sup>3</sup> ⇒ Nitrogen atom n(C=N) group and atom n(C=O) group and atom of n(C=O) phenolic oxygen group ⇒ Tridentate

L<sup>4</sup> ⇒ Nitrogen atom of n(C=N) group and oxygen atom of n(C=O) group ⇒ Bidentate

### Proposed structure of the ligands :

On the basis of elemental analysis, mass and IR spectra the following structure has been assigned for the ligands.



### Preparation of complexes :

All the complexes were prepared by the following method. A hot ethanolic solution of corresponding metal salt (0.05 mol) was mixed with hot ethanolic solution of the corresponding ligand (0.1 mol). The contents were refluxed for about 2-4 hr. On cooling the contents coloured complex was separated out. The complex was filtered washed with 50% ethanol and dried in a vacuum desiccator over P<sub>4</sub>O<sub>10</sub>.

## RESEARCH AND REMONSTRATION FINDINGS

On the basis of the elemental analysis (Table 2) the complexes have the composition M(L<sub>2</sub>) X<sub>2</sub> (where X=Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and CH<sub>3</sub>COO<sup>-</sup>). Molar conductance (Table 1) of the complexes with ligand L<sup>1</sup>, L<sup>2</sup>, L<sup>3</sup> and L<sup>4</sup> indicates the following nature :

Ligands L<sup>1</sup>, L<sup>2</sup> and L<sup>4</sup> act as bidentate manner coordinate through N and O atom. On the other hand L<sup>3</sup> acts as tridentate coordinate through N, two O atoms.

### Magnetic moment :

Ni(II) complexes either have octahedral or tetrahedral geometry and show magnetic moment between 2.92-3.01 B.M. corresponding to two unpaired electrons. The ground state of a regular octahedral complexes is <sup>3</sup>A<sub>2g</sub> and no singlet levels arising from d-configuration can cross it. The major contribution to the magnetic susceptibility of octahedral Ni(II) complex is given by the spin only term

$S(S+1) N\beta^2 g^2 / 3kT$ , where N=Avogadro number, β=Bohr magneton, g=2 and S=1.

### IR spectra of complexes :

#### IR spectra of nitrate complex :

The IR spectra of nitrate complexes with ligands L<sup>1</sup>, L<sup>2</sup> and L<sup>4</sup> show bands in the region 1409-1416 cm<sup>-1</sup> (ν<sub>3</sub>) ν<sub>a</sub> (NO<sub>2</sub>), 1288-1306 (ν<sub>1</sub>), ν<sub>a</sub> (NO<sub>2</sub>) and 890-930 cm<sup>-1</sup> (ν<sub>2</sub>) ν (NO). This indicates that nitrate group coordinated to central metal as unidentate[33-35]. However, nitrate complex of ligand L<sup>3</sup> the position of at 1393 cm<sup>-1</sup> indicating that nitrate group is uncoordinated.

#### IR spectra of acetato complexes :

IR spectral bands of acetato complex, L<sup>1</sup>, L<sup>2</sup> and L<sup>4</sup> show bands in the region 1410-1475 cm<sup>-1</sup> (ν<sub>3</sub>) ν<sub>a</sub> (NO<sub>2</sub>), 1290-1310 (ν<sub>1</sub>), ν<sub>a</sub> (NO<sub>2</sub>) and 895-944 cm<sup>-1</sup> (ν<sub>2</sub>) ν (NO) corresponding to monodentate nature. On the other hand ligands shows IR bands corresponding to uncoordinated acetate group [36].

### Electronic spectra :

The electronic spectra of the complexes of Ni(II) metals with L<sup>1</sup>, L<sup>2</sup>, L<sup>3</sup> and L<sup>4</sup> show three bands in the range of 9037-11085 cm<sup>-1</sup>, 13023-14880 cm<sup>-1</sup>, 23164-25600 cm<sup>-1</sup> (Table 2). These

**Table 1 : Elemental analysis of ligands**

Sr. No.	Empirical formula	Colour	Yield (%)	M. Point (0°C)	Molecular weight	Elemental analysis observed (Calculated)			
						C	H	N	O
1.	C <sub>7</sub> H <sub>13</sub> N <sub>3</sub> O <sub>3</sub>	White	68	180	187	44.23 (44.91)	6.50 (6.95)	22.15 (22.45)	25.10 (25.66)
2.	C <sub>7</sub> H <sub>13</sub> N <sub>3</sub> O <sub>2</sub>	Green	70	210	159	45.00 (45.28)	8.20 (8.17)	25.90 (26.41)	20.00 (20.12)
3.	C <sub>9</sub> H <sub>11</sub> N <sub>3</sub> O <sub>5</sub>	White	62	197	241	44.12 (44.81)	4.20 (4.56)	17.25 (17.42)	33.00 (33.19)
4.	C <sub>10</sub> H <sub>11</sub> N <sub>3</sub> O <sub>4</sub>	Shiny Green	65	225	237	50.50 (50.63)	4.54 (4.64)	17.70 (17.72)	27.00 (27.00)

**Table 2 : Elemental analysis of Ni(II) complexes**

Sr. No.	Complex	Colour	Yield (%)	M. Point (0°C)	Molecular weight	Elemental Analysis observed (Calculated)			
						C	H	N	M
1.	[Ni (L <sup>1</sup> ) <sub>2</sub> Cl <sub>2</sub> ] NiC <sub>14</sub> H <sub>26</sub> N <sub>6</sub> O <sub>4</sub> O <sub>2</sub> Cl <sub>2</sub>	Dark Green	72	282	502.69	33.30 (33.42)	5.10 (5.17)	16.53 (16.71)	11.51 (11.67)
2.	[Ni (L <sup>1</sup> ) <sub>2</sub> NO <sub>3</sub> ] <sub>2</sub> ] NiC <sub>14</sub> H <sub>26</sub> N <sub>8</sub> O <sub>10</sub> O <sub>2</sub>	Green	75	275	554.69	30.00 (30.28)	4.12 (4.32)	20.10 (20.19)	10.53 (10.58)
3.	[Ni (L <sup>1</sup> ) <sub>2</sub> CH <sub>3</sub> COO] <sub>2</sub> ] NiC <sub>18</sub> H <sub>30</sub> N <sub>6</sub> O <sub>8</sub> O <sub>2</sub>	Shiny	70	269	554.69	39.00 (39.22)	5.51 (5.81)	15.15 (15.25)	10.55 (10.65)
4.	[Ni (L <sup>2</sup> ) <sub>2</sub> Cl <sub>2</sub> ] NiC <sub>12</sub> H <sub>26</sub> N <sub>6</sub> O <sub>6</sub> O <sub>2</sub> Cl <sub>2</sub>	Dark Green	70	273	447.69	32.00 (32.16)	5.60 (5.80)	18.70 (18.76)	13.00 (13.10)
5.	[Ni (L <sup>2</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ] NiC <sub>12</sub> H <sub>26</sub> N <sub>8</sub> O <sub>12</sub> O <sub>2</sub>	Green	60	270	594.69	32.20 (32.28)	5.30 (5.38)	14.10 (14.12)	19.58 (19.86)
6.	[Ni (L <sup>2</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ] NiC <sub>16</sub> H <sub>32</sub> N <sub>6</sub> O <sub>10</sub> O <sub>2</sub>	Shiny Green	63	280	500.69	28.66 (28.76)	5.20 (5.19)	22.25 (22.36)	11.70 (11.72)
7.	[Ni (L <sup>3</sup> ) <sub>2</sub> Cl <sub>2</sub> ] NiC <sub>18</sub> H <sub>22</sub> N <sub>6</sub> O <sub>8</sub> O <sub>2</sub> Cl <sub>2</sub>	Dark Green	72	280	613.69	35.20 (35.19)	3.81 (3.91)	13.58 (13.68)	9.46 (9.56)
8.	[Ni (L <sup>3</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ] NiC <sub>18</sub> H <sub>22</sub> N <sub>8</sub> O <sub>10</sub> O <sub>2</sub> Cl <sub>2</sub>	Green	80	280	660.69	39.75 (39.95)	4.45 (4.54)	12.51 (12.71)	8.78 (8.88)
9.	[Ni (L <sup>3</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ] NiC <sub>22</sub> H <sub>28</sub> N <sub>6</sub> O <sub>12</sub> O <sub>2</sub>	Shiny Green	70	265	666.69	32.35 (32.39)	3.50 (3.59)	16.59 (16.79)	8.70 (8.80)
10.	[Ni (L <sup>4</sup> ) <sub>2</sub> Cl <sub>2</sub> ] NiC <sub>20</sub> H <sub>22</sub> N <sub>6</sub> O <sub>6</sub> O <sub>2</sub> Cl <sub>2</sub>	Dark Green	70	295	603.69	39.65 (39.75)	3.54 (3.64)	13.81 (13.91)	9.80 (9.72)
11.	[Ni (L <sup>4</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ] NiC <sub>20</sub> H <sub>22</sub> N <sub>8</sub> O <sub>12</sub> O <sub>2</sub>	Green	65	259	650.69	44.16 (44.26)	4.30 (4.30)	12.69 (12.90)	9.00 (9.01)
12.	[Ni (L <sup>4</sup> ) <sub>2</sub> CH <sub>3</sub> COO] <sub>2</sub> ] NiC <sub>24</sub> H <sub>28</sub> N <sub>6</sub> O <sub>10</sub> O <sub>2</sub>	Shiny Green	69	278	656.69	36.44 (36.54)	3.30 (3.35)	17.00 (17.05)	8.83 (8.93)

bands may be assigned to <sup>3</sup>A<sub>2g</sub> (F) → <sup>3</sup>T<sub>2g</sub> (F) (ν<sub>1</sub>), <sup>3</sup>A<sub>2g</sub> (F) → <sup>3</sup>T<sub>1g</sub> (F) (ν<sub>2</sub>) and <sup>3</sup>A<sub>2g</sub> (F) → <sup>3</sup>T<sub>1g</sub> (P) (ν<sub>3</sub>) transition respectively. The position of these bands corresponding to an octahedral geometry around the nickel ion.

**Ligand field parameters :**

Various ligand field parameters have been calculated and are given in Table 3. The triplet terms <sup>3</sup>F and <sup>3</sup>P arising from <sup>3</sup>d<sub>g</sub> configuration of the free ion Ni(II) (which have an energy separation equal to 15B in terms of the Racah interelectronic

repulsion parameters) are presented as:

$${}^3A_{2g}(F) E = -12Dq \tag{1}$$

$${}^3T_{2g}(F) E = -2Dq \tag{2}$$

$${}^3T_{1g}(F) \text{ and } {}^3T_{1g}(P) : [6DqP - 16(Dq)^2] + [-6Dq - P] E + E^2 = 0 \tag{3}$$

where P is the energy of the <sup>3</sup>P state. From equation (1) and (2) it is seen that the energies of both <sup>3</sup>A<sub>2g</sub> (F) and <sup>3</sup>T<sub>2g</sub> (F) are linear functions of Dq. The difference in the energy between <sup>3</sup>T<sub>2g</sub> state and <sup>3</sup>A<sub>2g</sub> state in the complexes gives the direct value of 10 Dq.

**Table 3 : Magnetic moments and electronic spectral data of the complexes**

Complex	$\mu_{\text{eff}}$ (B.M.)	$\lambda_{\text{max}}$ (cm <sup>-1</sup> )
[Ni(L <sup>1</sup> ) <sub>2</sub> Cl <sub>2</sub> ]	2.92	11085, 14088, 25675
[Ni(L <sup>2</sup> ) <sub>2</sub> Cl <sub>2</sub> ]	2.99	9880, 12141, 24056
[Ni(L <sup>3</sup> ) <sub>2</sub> Cl <sub>2</sub> ]	2.99	9037, 14129, 24098
[Ni(L <sup>4</sup> ) <sub>2</sub> (Cl <sub>2</sub> ) <sub>2</sub> ]	2.96	9559, 14166, 24038
[Ni(L <sup>1</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ]	2.94	11235, 14049, 23164
[Ni(L <sup>2</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ]	2.95	9680, 14224, 24300
[Ni(L <sup>3</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ]	2.98	9678, 14388, 24570
[Ni(L <sup>4</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ]	3.01	9354, 13023, 24272
[Ni(L <sup>1</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ]	2.95	9766, 14880, 23984
[Ni(L <sup>2</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ]	2.98	9990, 14128, 24096
[Ni(L <sup>3</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ]	2.94	9870, 14577, 25600
[Ni(L <sup>4</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ]	2.97	9737, 13468, 24818

In the complexes, the value of Racah parameter B is found less than the value of free ion *i.e.* 1041 cm<sup>-1</sup>. The value of b has been calculated with the relation  $b = \frac{B_{\text{Complex}}}{B_{\text{Free ion}}}$ . The value of b for the nickel (II) complexes, under study, lie in the range 0.34-0.65 (Table 4).

**Table 4 : Ligand field parameters and ESR spectral data of Ni (II) complexes**

Complex	Dq (cm <sup>-1</sup> )	B (cm <sup>-1</sup> )	$\beta$	LFSE (kJmole <sup>-1</sup> )
[Ni(L <sup>1</sup> ) <sub>2</sub> Cl <sub>2</sub> ]	1118	407	0.39	160
[Ni(L <sup>2</sup> ) <sub>2</sub> Cl <sub>2</sub> ]	988	568	0.55	151
[Ni(L <sup>3</sup> ) <sub>2</sub> Cl <sub>2</sub> ]	933	681	0.65	134
[Ni(L <sup>4</sup> ) <sub>2</sub> Cl <sub>2</sub> ]	956	633	0.61	137
[Ni(L <sup>1</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ]	1123	359	0.34	161
[Ni(L <sup>2</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ]	968	632	0.61	139
[Ni(L <sup>3</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ]	967	663	0.64	138
[Ni(L <sup>4</sup> ) <sub>2</sub> (CH <sub>3</sub> COO) <sub>2</sub> ]	935	622	0.60	134
[Ni(L <sup>1</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ]	975	646	0.62	140
[Ni(L <sup>2</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ]	999	550	0.53	143
[Ni(L <sup>3</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ]	987	563	0.54	142
[Ni(L <sup>4</sup> ) <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ]	973	604	0.58	140

#### Proposed structure of the complexes :

On the basis of elemental analysis, magnetic moment, molar conductance, IR, electronic and EPR spectral studies an octahedral geometry has been assigned for the complexes with ligand L<sup>1</sup>, L<sup>2</sup>, L<sup>3</sup> and L<sup>4</sup>.

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