



# SYNTHESIS AND CHARACTERIZATION AS WELL AS ANTIMICROBIAL STUDIES OF LANTHANIDE (III) CHLORIDE COMPLEXES OF p-HYDROXYBENZYLIDENE-2- AMINOTHIAZOLE AND p-HYDROXYBENZYLIDENE-2- AMINO-6-METHYLPYRIDINE

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## ABSTRACT

p-hydroxybenzylidene-2-aminothiazole (HBAT) and p-hydroxybenzylidene-2-amino-6-methylpyridine (HBAMP) complexes of Lanthanide (III) chloride with the general composition  $LnL_2.X_3$  [ $Ln = La$  (III),  $Gd$  (III) and  $Nd$  (III);  $L = HBAT$  and  $HBAMP$ ;  $X = Chloride$ ] have been prepared on the basis of analytical and molecular weight, infrared and electronic spectral data. Infrared studies of these complexes reveal that HBAT and HBAMP act as ONS and ONN tridentate ligand and coordinate through oxygen, nitrogen and sulphur. Coordination number of lanthanide (III) ion for  $La$  (III),  $Gd$  (III) and  $Nd$  (III) complexes were found to be six. Atomic absorption spectral data shows the percentage values of the complexes. All the complexes exhibited distorted octahedral geometry. Antimicrobial studies of these complexes against *E. coli*, *K. pneumoniae*, *Aspergillus niger* and *Aspergillus fumigatus* are also reported.

**Key words:** Lanthanide (III), IR, Electronic, AAS, HBAT and HBAMP.

## INTRODUCTION

In the past, a number of workers isolated solid complexes of lanthanide (III) with drugs. But literature survey shows that the solid lanthanide (III) complexes of p-hydroxybenzylidene-2-aminothiazole (HBAT) and p-hydroxy benzylidene-2-amino-6-methylpyridine (HBAMP) have not been reported. We have reported here the synthesis of lanthanide (III) complexes of p-hydroxybenzylidene-2-aminothiazole and p-hydroxy benzylidene-2-amino-6-methylpyridine and their characterization. Mannich bases HBAT and HBAMP (ligands) are widely studied subjects because of their applications in pharmaceutical and polymer chemistry<sup>1</sup>. Mannich base prepared from heterocycles bearing

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nitrogen, sulfur and thiazole moieties constitute, the core structure of a number of biologically interesting compounds.

## EXPERIMENTAL

All the lanthanide (III) chloride were obtained from M/s Indian Rare Earths Ltd., Kerala (India) and all other chemicals obtained from M/s Sigma Chemicals Company, E.Mark, U.S.A. were used as such.

### Preparation and analysis of complexes

#### Preparation of schiff bases

For the preparation of p-hydroxybenzylidene-2-imino-thiazole (SB<sub>1</sub>), 1.22 g 4-hydroxybenzaldehyde and 1.0 g 2-aminothiazole were dissolved in 20 mL ethanol and reflux for 6-7 hours over water bath. The obtained solution was allowed to cool at room temperature. The concentrated solution was cooled in refrigerator and obtained product was filtered, washed with ether and dried under reduced pressure over anhydrous calcium chloride.

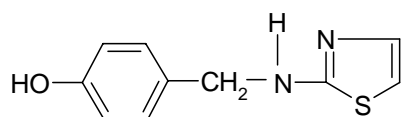
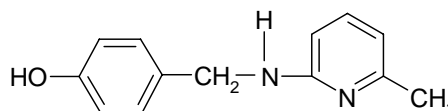
For the preparation of p-hydroxybenzylidene-2-amino-6-methyl pyridine (SB<sub>2</sub>), 1.22 gm 4-hydroxybenzaldehyde in ethanol was mixed with an ethanolic solution of 1.08 gm of 2-amino-6-methylpyridine (dissolved in 20 mL ethanol). Mixture was refluxed for 6-7 hours over a water bath. The obtained solution was allowed to cool at room temperature and the concentrated solution was cooled in refrigeration for 24 hours. The obtained product was filtrated, washed with acetone several times and followed by ether. It was recrystallized with absolute alcohol and dried under reduced pressure over anhydrous calcium chloride.

#### Preparation of mannich bases

For the preparation of p-hydroxybenzylidene-2-aminothiazole (MB<sub>1</sub>) derived from p-hydroxybenzylidene-2-imino-thiazole (SB<sub>1</sub>) (2.04 g) was stirred with 20 mL of methanol. The product is then cooled to 0°C and sodium borohydride (1.0 g) was added over a period of 1 hour in 3 to 4 installments. Slowly the temperature was raised to room temperature. A dark brown solution resulted and the solvent was slowly evaporated. A solid coloured powder was obtained. It was then washed with ethanol and dried in air, a deep brown coloured crystals was obtained. Mass spectra of the ligand exhibits m/z values: 205, 189, 107, 122, 113 and 99 assignable to C<sub>10</sub>H<sub>10</sub>N<sub>2</sub>OS, C<sub>10</sub>H<sub>10</sub>N<sub>2</sub>OS, C<sub>7</sub>H<sub>7</sub>O, C<sub>7</sub>H<sub>8</sub>NO, C<sub>4</sub>H<sub>5</sub>N<sub>2</sub>S and C<sub>3</sub>H<sub>3</sub>N<sub>2</sub>S molecular ion.

For the preparation of p-hydroxybenzylidene-2-amino-6-methylpyridine (MB<sub>2</sub>) derived from p-hydroxybenzylidene-2-imino-6-methylpyridine (SB<sub>2</sub>): p-hydroxy

benzylidene-2-imino-6-methylpyridine (2.12 g) was stirred with 20 mL of methanol. The product was then cooled to 0°C and then sodium borohydride (1.0 g) was added to it, in a period of one hour. Slowly the temperature was raised to room temperature. A light brown coloured solution was resulted and then solvent was slowly evaporated. A solid chocolate coloured powder is obtained. It was then washed with alcohol and dried in air. The mass spectra of the ligand exhibits m/z values 214, 198, 176, 121, 107 and 93 assignable to C<sub>13</sub>H<sub>14</sub>N<sub>2</sub>O, C<sub>13</sub>H<sub>14</sub>N<sub>2</sub>, C<sub>12</sub>H<sub>12</sub>N<sub>2</sub>O, C<sub>7</sub>H<sub>9</sub>N<sub>2</sub>, C<sub>6</sub>H<sub>7</sub>N<sub>2</sub> and C<sub>6</sub>H<sub>7</sub>N molecular ion.

(MB<sub>1</sub>)(MB<sub>2</sub>)

### Preparation of metal complexes

The metal complexes of Lanthanide (III) chloride were prepared by refluxing the alcoholic solution of HBAT and HBAMP with Lanthanide (III) chloride for 3-4 hours (M : L ratio 1 : 2). The precipitated complex was filtered and washed with ethanol, ether and dried in air.

### Physical measurements

The Lanthanide metal ions were estimated by standard method<sup>2</sup>. The percentage of metal of Lanthanide (III) in complexes were determined by AAS method from Dayalbagh Educational Institute, Agra. FTIR spectra of the ligand and their lanthanide (III) complexes were recorded on (KBr matrix) Perkin Elmer 842 grating, IR spectrometer in the range 4000-200 cm<sup>-1</sup>. FAB Mass spectra of ligands were determined on Jeal, JMS-D-300 double focusing spectrometer at R.S.I.C., C.D.R.I., Lucknow. The electronic spectrum of the complexes in (DMF/Ethanol) solution were recorded on UV 5704SS (Double Beam UV-Vis Spectrophotometer) in the range of 380-1100 nm at the Department of Chemistry, I.B.S., Khandari, Agra. The <sup>1</sup>H NMR spectra of the ligand was recorded in DMSO at R.S.I.C., C.D.R.I., Lucknow.

## RESULTS AND DISCUSSION

The analytical data of the present complexes (Table 1) indicate that all the six complexes show 1 : 2 (metal : ligand) ratio. The analytical data, percentage of metal of lanthanide (III) HBAT and HBAMP complexes are represented in Table 1. All these complexes are soluble in ethanol, methanol and DMSO.

Table 1: Physical properties and analytical data of metal complexes of MB<sub>1</sub> and MB<sub>2</sub>

S. No.	Ligand/Complexes	Yield (%)	M.P. (°C)	Formul a weight	Colour	Percentage of Elements(Found/Calculated)					
						C	H	N	S	M	Cl
1	C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS	88	150	206	Dark brown	57.92/8.25	4.80/4.85	17.53/13.59	15.50/15.53	-	-
1.1	La (C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS) <sub>2</sub> .Cl <sub>3</sub>	82	215-217	656	Dark brown	36.56/36.58	3.00/3.04	8.52/8.53	9.70/9.75	21.08/21.18	15.95/16.00
1.2	Nd (C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS) <sub>2</sub> .Cl <sub>3</sub>	82	180-183	661	Dark brown	32.15/36.30	3.00/3.02	8.40/8.47	9.68/9.68	21.68/21.78	15.88/15.88
1.3	Gd (C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS) <sub>2</sub> .Cl <sub>3</sub>	81	210-212	674	Brown	35.45/35.60	2.91/2.96	8.20/8.30	9.48/9.49	23.14/23.29	15.60/15.57
2	C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O	87	190	214	Light brown	72.80/72.98	6.50/6.54	13.56/13.08	-	-	-
2.1	La (C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O) <sub>2</sub> .Cl <sub>3</sub>	79	245-248	672	Light Brown	46.30/46.42	4.06/4.16	8.23/8.33	-	20.50/20.68	15.45/15.62
2.2	Nd (C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O) <sub>2</sub> .Cl <sub>3</sub>	80	225-228	677	Light Brown	46.00/46.08	4.11/4.13	8.17/8.27	-	21.17/21.27	15.49/15.50
2.3	Gd (C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O) <sub>2</sub> .Cl <sub>3</sub>	80	250-252	690	Yellow	45.11/45.21	4.00/4.05	8.06/8.11	-	22.60/22.75	15.20/15.21

The  $^1\text{H}$  NMR spectra of the Mannich bases ( $\text{MB}_1$  &  $\text{MB}_2$ ) were recorded in DMSO, assigned as a solvent peak in the spectrum at  $\delta$  3.41 ppm. A doublet may be due to heterocyclic proton (2H) present in thiazole ring was observed in the spectrum at  $\delta$  7.0 ppm<sup>3</sup> in  $\text{MB}_1$  while a triplet at  $\delta$  7.2 ppm equivalent was observed may be due to 3H proton present in pyridine ring in  $\text{MB}_2$ . A singlet was observed at  $\delta$  5.3 - 5.7 ppm equivalent to 1H, may be assigned as -OH proton on para position while a doublet at  $\delta$  6.4 - 6.68 ppm equivalent to 4H was present in the aromatic benzene ring<sup>4</sup> in the Mannich bases. A singlet was observed at  $\delta$  2.56 ppm equivalent to 2H of - $\text{CH}_2$  group attached with the benzene ring<sup>5</sup>. A singlet at  $\delta$  5.56 - 5.58 ppm equivalent to the 1H proton of secondary -NH group was present in these Mannich bases. These data are in good agreement with other spectrometric results.

### IR Spectra

Infrared spectral data and their tentative assignments are shown in Table 2 and 3. The IR spectra of  $\text{SB}_1$  and  $\text{SB}_2$  show the bands at  $\sim 1605\text{-}1610\text{ cm}^{-1}$  due to azomethine<sup>6</sup> ( $\nu > \text{C}=\text{N}$ ) which were disappeared in the spectrum of  $\text{MB}_1$  and  $\text{MB}_2$  and new bands was observed in the region  $\sim 3358\text{-}3365\text{ cm}^{-1}$  due to secondary amino group. In the IR spectra of all the Lanthanide (III) complexes, a broad band at  $\sim 3452\text{-}3520\text{ cm}^{-1}$  due to  $\nu_{(\text{-OH})}$  disappeared thus indicating the deprotonation of phenolic OH. The bands observed at  $\sim 1230\text{-}1480\text{ cm}^{-1}$  in the ligands is due to  $\nu_{(\text{-C-O-})}$  phenolic mode. These bands are shifted to higher frequency region ( $\sim 1242\text{-}1251\text{ cm}^{-1}$ ) for the complexes of  $\text{MB}_1$ . The characteristic IR bands in the spectrum of the  $\text{MB}_1$  is observed at  $\sim 1513\text{ cm}^{-1}$  ( $\nu_{(\text{C}=\text{N cyclic})}$ ),  $\sim 1352\text{ cm}^{-1}$  ( $\nu_{(\text{C-N cyclic})}$ ) and  $\sim 830\text{ cm}^{-1}$  ( $\nu_{(\text{C-S-C})}$ ) of the thiazole moiety.

The position of the former two bands remains unaltered in all the complexes ruling out the possibility of coordination of thiazole ring nitrogen. The vibration modes due to  $\nu_{(\text{-M-S})}$  coordination were observed at  $300\text{-}365\text{ cm}^{-1}$  in all the complexes confirming the participation of thiazole ring S-atom in complexation. The strong absorption band due to pyridine ring occurring at  $\sim 1498\text{ cm}^{-1}$  ( $\nu_{(\text{C-N-C})}$  pyridine) has shifted to higher frequency region by  $\sim 10\text{-}25\text{ cm}^{-1}$  in the complexes showing the participation of nitrogen atom of pyridine in the complexation. This is also supported by the appearance of bond in the far infrared region  $\sim 500\text{-}520\text{ cm}^{-1}$  due to  $\nu_{(\text{M-N})}$  vibration. And the above discussion indicates that both the ligands ( $\text{MB}_1$  and  $\text{MB}_2$ ) are tridentate in nature with ONS and ONN donor system having distorted octahedral environment around metal ion.

**Table 2: Infra-Red frequencies (cm<sup>-1</sup>) of MB<sub>1</sub> and its metal complexes**

S. No.	Ligand/Complexes	Ligand modes					Coordination modes			
		$\nu(\text{OH})$	$\nu(\text{CH}_2\text{-NH})$	$\nu(\text{C-O})$ Phenolic	$\nu(\text{C=N})$ cyclic	$\nu(\text{C-N})$ cyclic	$\nu(\text{C-S-C})$ thiazole	$\nu(\text{M-O})$	$\nu(\text{M-N})$	$\nu(\text{M-S})$
1	C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS	3450	3355	1243	1512	1350	833	-	-	-
1.1	La(C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS) <sub>2</sub> Cl <sub>3</sub>	-	3376	1247	1512	1349	836	525	465	341
1.2	Nd(C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS) <sub>2</sub> Cl <sub>3</sub>	-	3382	1248	1521	1356	838	544	472	360
1.3	Gd(C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS) <sub>2</sub> Cl <sub>3</sub>	-	3378	1245	1513	1352	838	514	480	328

**Table 3: Infra-Red frequencies (cm<sup>-1</sup>) of MB<sub>2</sub> and its metal complexes**

S. No.	Ligand/Complexes	Ligand modes				Coordination modes		
		$\nu(\text{OH})$	$\nu(\text{CH}_2\text{-NH})$	$\nu(\text{C-O})$ Phenolic	$\nu(\text{C-N-C})$ pyridine	$\nu(\text{M-O})$	$\nu(\text{M-N})$	$\nu(\text{M-N})$ pyridine
2	C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O	3518	3353	1480	1502	-	-	-
2.1	La(C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O) <sub>2</sub> Cl <sub>3</sub>	-	3384	1470	1510	532	483	507
2.2	Nd(C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O) <sub>2</sub> Cl <sub>3</sub>	-	3377	1467	1512	538	484	518
2.3	Gd(C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O) <sub>2</sub> Cl <sub>3</sub>	-	3382	1457	1514	542	482	513

### Electronic spectra

The electronic spectral data of the representative complexes are given in Table 4. The electronic spectra of the Nd (III) complexes have been attributed to the transition from ground levels 4I<sub>9/2</sub> to the excited J-levels of 4f<sup>n</sup> configuration<sup>7</sup>. The electronic spectra of f-f transition for Nd (III) complexes and their assignments are given in Table 4. The nephelauxetic ratio  $\beta$  ( $\nu_{\text{complex}}/\nu_{\text{aquo}}$ ), covalency factor ( $b^{1/2}$ ) and Sinha's parameter  $\delta\%$  suggest covalency in the metal-ligand bonding<sup>8</sup>.

### Thermal Studies

From the TGA analysis it is divided into four stages based on the heating rate, the first stage is evaporation of water vapour<sup>9</sup> and chloride molecules between 90-130°C from the complex, the second stage shows 19% mass loss of remaining compounds between 130-317°C and third stage can be seen above 320-867°C. The fourth stage can

be seen between 867-1400°C, the oxide of neodymium chloride was remaining. The decomposition temperatures are approximately 130, 320, 867 and 1400°C for components 1, 2, 3 and 4 respectively. Thermal data are represented in Table 5.

**Table 4: Electronic spectral data (cm<sup>-1</sup>)**

Electronic Spectral Band (In Standard)	Electronic Spectral Band (In Complex)	J-Levels (Assignments)	(1-β)	β	b <sup>1/2</sup>	δ%	η
Nd(C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS) <sub>2</sub> Cl <sub>3</sub>							
19420	19250	<sup>4</sup> I <sub>9/2</sub> → <sup>2</sup> G <sub>9/2</sub>	0.00875	0.99124	0.04677	0.88273	0.00440
17390	17200	→ <sup>4</sup> G <sub>5/2</sub> , <sup>2</sup> G <sub>7/2</sub>	0.01092	0.98907	0.05224	1.10406	0.00551
13420	13200	→ <sup>2</sup> S <sub>3/2</sub> , <sup>4</sup> F <sub>7/2</sub>	0.01639	0.98360	0.06401	1.66632	0.00823
12500	12350	→ <sup>4</sup> F <sub>5/2</sub> , <sup>4</sup> H <sub>9/2</sub>	0.01200	0.98800	0.05477	1.21457	0.00601
Nd(C <sub>13</sub> H <sub>14</sub> N <sub>2</sub> O) <sub>2</sub> Cl <sub>3</sub>							
19607	19520	<sup>4</sup> I <sub>9/2</sub> → <sup>2</sup> G <sub>9/2</sub>	0.0045	0.9955	0.0335	0.4520	0.0022
17241	17177	→ <sup>4</sup> G <sub>5/2</sub> , <sup>2</sup> G <sub>7/2</sub>	0.0037	0.9963	0.0304	0.3713	0.0018
13513	13487	→ <sup>2</sup> S <sub>3/2</sub> , <sup>4</sup> F <sub>7/2</sub>	0.0019	0.9981	0.0217	0.1903	0.0009
12500	12420	→ <sup>4</sup> F <sub>5/2</sub> , <sup>4</sup> H <sub>9/2</sub>	0.0064	0.9936	0.0400	0.6441	0.0032

**Table 5: Thermal decomposition data of NCMB<sub>1</sub>**

Compound	Decom- position Temp. (°C)	Weight loss				Probable assignments
		Observed values		Calculated values		
		Weight loss	Residual mass	Weight loss	Residual mass	
Nd(C <sub>10</sub> H <sub>10</sub> N <sub>2</sub> OS) <sub>2</sub> Cl <sub>3</sub>	90-130	125.6	535.4	124.5	536.5	3Cl + H <sub>2</sub> O
	130-317	101.7	433.7	104	432.5	C <sub>6</sub> H <sub>6</sub> .CH <sub>2</sub> N
	320-867	86.7	347	85	347.5	C <sub>3</sub> H <sub>3</sub> NS
	867-1400	11	336	11.5	336	Nd <sub>2</sub> O <sub>3</sub>

**Table 6: Lanthanum (III) chloride complex of MB<sub>1</sub>**

S. No.	Interplanar spacing (dÅ <sup>o</sup> )	Rel. Int.	sin <sup>2</sup> θ (Obs.)	sin <sup>2</sup> θ (Calc.)	hkl
1	26.39091	70.1	0.00085	0.00084	100
2	23.35719	90.8	0.00108	0.00109	110
3	22.18282	79.7	0.00120	0.00119	112
4	21.42934	65.4	0.00129	0.00121	111
5	19.71494	78.6	0.00152	0.00147	122
6	17.75024	77.4	0.00188	0.00182	123
7	16.29418	83.6	0.00223	0.00222	103
8	15.62379	100	0.00243	0.00241	201
9	14.67935	85	0.00275	0.00274	211
10	14.08525	97	0.00299	0.00291	202
11	13.48309	80.9	0.00326	0.00317	213
12	12.84675	96.6	0.00471	0.00419	310
13	12.2934	81.6	0.00392	0.00390	311
14	11.83782	75.6	0.00423	0.00421	312
15	11.3601	89.7	0.00459	0.00453	400

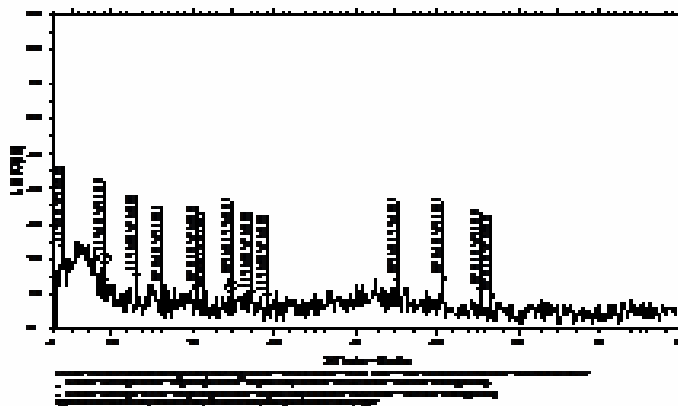
**X-Ray (Powder Diffraction) analysis**

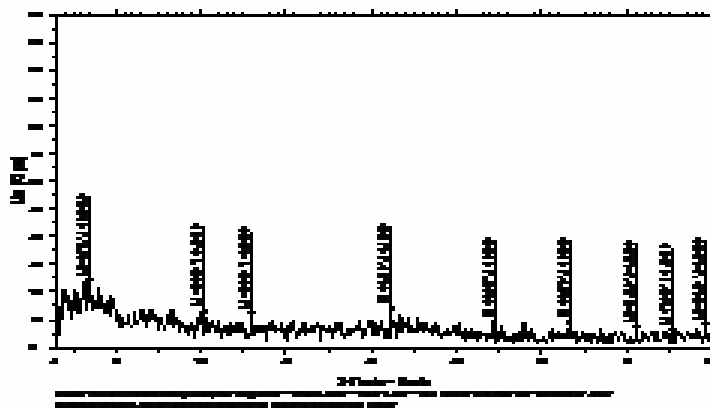
The data were collected on Bruker AXS D8 Advance Diffractometer using lanthanide (III)  $\alpha$  radiation over a range of 3-79° 2 $\theta$  and the values sin<sup>2</sup>θ (observed and calculated), interplanar spacings (d), relative intensities and hkl values of different lattice planes have been summarized in tables. In the X-ray powder diffraction pattern the following interesting points were noted<sup>10</sup>. X-ray powder diffraction pattern of the lanthanide (III) complexes remained the same indicating that the crystal structure did not change. However, a large decrease in the line intensity of the X-ray powder diffraction was noticed when the ligand was exchanged by La (III) ions. On the basis of electronic spectra studies and X-ray (Powder diffraction) analysis, it was concluded that the La (III) complexes of MB<sub>1</sub> and MB<sub>2</sub> have distorted octahedral geometry. (Table 6 and 7).



**Table 7: Lanthanum (III) chloride complex of MB<sub>2</sub>**

S. No.	Interplanar Spacing (dÅ <sup>o</sup> )	Rel. Int.	sin <sup>2</sup> θ (Obs.)	sin <sup>2</sup> θ (Calc.)	hkl
1	27.50023	60.7	0.00077	0.00078	110
2	25.24384	46.3	0.00092	0.00091	111
3	23.13189	87.8	0.00109	0.00108	112
4	21.5871	71.7	0.00126	0.00127	122
5	20.33243	77.8	0.00143	0.00144	123
6	18.44238	69.4	0.00173	0.00174	108
7	17.14533	88	0.00201	0.00202	211
8	16.22249	55.4	0.00225	0.00226	212
9	15.22544	73.9	0.00255	0.00256	222
10	14.83288	77.6	0.00268	0.00266	223
11	14.0508	100	0.00300	0.00301	225
12	13.08545	98.7	0.00345	0.00344	206
13	12.54623	75.8	0.00376	0.00377	300
14	11.797	66.2	0.00425	0.00424	301
15	11.342	92.5	0.00460	0.00461	311

**Fig. 1: X-ray Diffractograph of Lanthanum (III) Chloride of MB<sub>1</sub>**



**Fig. 2: X-ray Diffractograph of Lanthanum (III) Chloride of MB<sub>2</sub>**

### **Antibacterial activities**

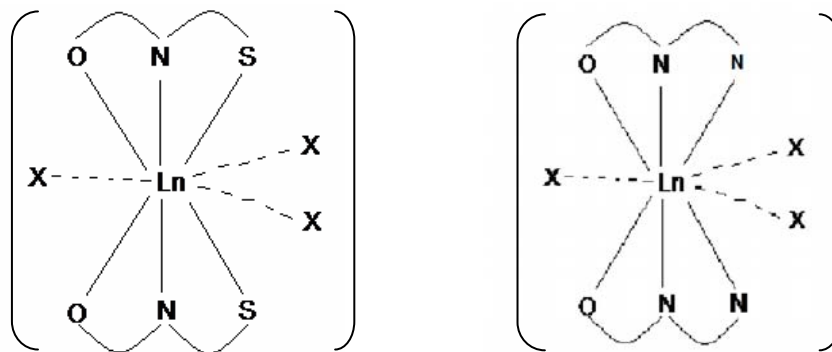
The antibacterial activities of ligands (MB<sub>1</sub> and MB<sub>2</sub>) and the complexes were assayed against the bacteria *E. coli* and *K. pneumoniae* using paper-dick method<sup>11</sup>. The results of antibacterial activities of the ligand (MB<sub>1</sub>) and complexes show that MB<sub>1</sub> has good antibacterial activity against *E. coli* and *K. pneumoniae* at all concentrations i.e. 250 ppm, 500 ppm and 750 ppm, but MB<sub>2</sub> was inactive against *E. coli* and *K. pneumoniae* at all concentrations i.e. 250 ppm, 500 ppm and 750 ppm. Ln (III) and Nd (III) complexes of MB<sub>1</sub> have good antibacterial activities against *E. coli* and *K. pneumoniae* at all the concentrations but Gd (III) is less active. La (III), Gd (III) and Nd (III) complexes of MB<sub>2</sub> have less active or inactive at all the concentrations against *E. coli* and *K. pneumoniae*.

### **Antifungal activities**

Form the present studies, it was observed that the ligands (MB<sub>1</sub> and MB<sub>2</sub>) and their metal complexes were inactive at all concentration levels against fungi *A. niger* and *A. fumigatus*.

### **Stereo chemistry**

Both the present complexes are non-ionic in nature and infra-red data reveal the tridentate nature (ONS, ONN) of the coordinating lignds, resulting in the coordination number six in these complexes. The tentative structure of the complexes may be represented as -



**Proposed structure of  $\text{LnL}_2 \cdot \text{X}_3$**   
 (where Ln = La, Gd and Nd; L = HBAT and HBAMP; X = chloride ions)

### ACKNOWLEDGMENT

The authors are thankful to Head, University Department of Chemistry, Dr. BRA University, Khandari, Agra, for providing laboratory facilities. The authors are also thankful to IIT Chennai, SAIF, Cochin and CDRI, Lucknow for various instrumental facilities.

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*Revised : 03.01.2012*

*Accepted : 07.01.2012*