



## Synthesis, characterization of tridentate Schiff base metal chelates and their antimicrobial activity

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### ABSTRACT

Biological active tridentate Schiff base ligand derived from 2-amino, 4-methyl phenol with indole 3-aldehyde and their Ti(IV), Zr(IV), Cd(II) and Hg(II) metal chelates have been synthesized and characterized on the basis of IR, Elemental analysis, Electronic spectral data, Magnetic measurement, Conductance, <sup>1</sup>H NMR and X-ray diffraction method. The Schiff base and its metal complexes have been tested for their antibacterial as well as antifungal activity using disc diffusion methods and the results discussed.

**Keywords:** Schiff base, Metal chelates, Spectral studies, Antimicrobial activity.

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### INTRODUCTION

Indole derivatives and their metal complexes have received much attention due to their wide application in physiology [1] and pharmacology [2]. They constitute an important class of compounds possessing antibacterial [3], anticonvulsant [4] and antihypertensive activity. These observation led to the conception that Schiff bases of indole 3-aldehyde would possess potential antimicrobial properties. It was therefore, proposed to investigate the ligation properties of Schiff base derived from 2-amino, 4-methyl phenol with Indole 3-aldehyde and their Ti(IV), Zr(IV), Cd(II) and Hg(II) metal ion complexes.

### EXPERIMENTAL SECTION

All chemicals and solvents are used AR grade. All the metals were used as their chloride salts. Infrared and UV spectra were recorded on Perkin Elmer spectrum -100 and UV-Vis spectrophotometer 119. Conductance of the metal complexes was determined in DMF on conductivity meter Equip-tronics model No-EQ665. Melting points were recorded by open capillary method and are uncorrected. <sup>1</sup>H-NMR spectra of Schiff base and its metal complexes recorded on Bruker 300MHz spectrometer in DMSO-d<sub>6</sub>. Elemental analysis was carried out on instrument Eager-300 analyser. Magnetic measurements were done on solid complexes using Guoy method. Powder XRD patterns of the complexes are recorded on Philips Analytical XRD B.V. at CFC Shivaji University, Kolhapur. In our studies we have conducted some microbial activity to see whether chelating agent (Schiff base) and metal chelates have any action on microbial activities. Bacteria species like Escherichia coli, Bacillus subtilis, Aspergillus flavus and Aspergillus niger.

### Synthesis of Schiff base

The Schiff base were prepared by condensing of 2-amino, 4-methyl phenol and Indole 3-aldehyde in ethanol by refluxing on water bath for two to three hours in presence of 2-3 drops of glacial acetic acid. The reaction mixture were poured in crushed ice, where by brown precipitate was obtained. It was suction filtered, washed with distilled water and ethanol.

Then dried in vacuum dessicator. The pure Schiff base was recrystallized from ethanol. The purity of the Schiff base was checked by TLC. Further the structure of Schiff base was confirmed by subjecting them to IR, NMR and Elemental analysis.

### Synthesis of Metal complexes

To warm ethanolic solution of AMPIA (0.02M) and ethanolic solution of the corresponding metal chloride (0.01M) in the ratio 1:2 were added drop wise with constant string. The  $P^H$  of the solution was adjusted in the range of 6.8 to 7.1 by adding alcoholic ammonia. The reaction mixture was refluxed for four to five hours on water bath. The reddish brown solid complex precipitated at the end was digested, separated by filtration washed with warm alcohol. The complex is dried in vacuum at room temperature and stored in glass bottle.

## RESULTS AND DISCUSSION

All the metal complexes were intense coloured, they were insoluble in common organic solvents and soluble in DMF, DMSO. All the results elemental analysis are given in the table. For the estimation of chloride by Volhard's method is used [5], metal ion percentage is determined by standard method [6]. Presence of lattice water molecules present in the metal complexes was recorded by keeping the known weight of complex in previously weighed crucible in oven at 110°C for one hour. The difference in weight gives the amount of lattice water in the complex. All compounds give satisfactory elemental analysis. Values are in close agreement with the values calculated for expected molecular formulae assigned to these complexes, suggesting 1:2 for Ti(IV), Zr(IV) complexes and 1:1 ratio for Cd(II), Hg(II) complexes. The molar conductance of all these complexes in DMF reveals their electrolytic nature [7-9] except Hg(II) complex. All the metal complexes are diamagnetic in nature.

**Table-1: Analytical data and other physical properties of ligand and metal chelates.**

Ligand / Metal Chelates	Empirical Formula	Mol. Wt.	Colour	D.P <sup>o</sup> C	% of Yield	M:L ratio	Molar Cond. (Ohm <sup>-1</sup> cm <sup>2</sup> mol <sup>-1</sup> )	$\mu$ eff. B.M.	Elemental analysis % Cal (Obs)				
									C	H	N	M	Cl
AMPIA	C <sub>16</sub> H <sub>14</sub> N <sub>2</sub> O	250.31	Brown	203	60	-	-	-	76.70 (76.45)	5.59 (4.98)	11.18 (10.74)	-	-
[Ti(AMPIA) <sub>2</sub> ]Cl <sub>2</sub>	C <sub>32</sub> H <sub>28</sub> N <sub>4</sub> O <sub>2</sub> Cl <sub>2</sub> Ti	616.90	Reddish Brown	>300	58	1:2	126	Diamag.	62.24 (62.09)	4.21 (4.32)	9.07 (9.00)	7.76 (7.36)	11.50 (11.18)
[Zr(AMPIA) <sub>2</sub> ]Cl <sub>2</sub> ·H <sub>2</sub> O	C <sub>32</sub> H <sub>28</sub> N <sub>4</sub> O <sub>3</sub> Cl <sub>2</sub> Zr	678.22	Brown red	>300	55	1:2	122	Diamag.	56.61 (56.72)	4.12 (4.05)	8.25 (7.97)	13.44 (13.18)	10.46 (10.28)
[Cd(AMPIA)H <sub>2</sub> O]Cl·H <sub>2</sub> O	C <sub>16</sub> H <sub>17</sub> N <sub>2</sub> O <sub>3</sub> ClCd	432.90	Grey Brown	>300	55	1:1	62	Diamag.	44.35 (44.16)	3.92 (4.32)	6.46 (6.36)	25.96 (25.76)	8.20 (7.98)
[Hg(AMPIA)Cl]H <sub>2</sub> O	C <sub>16</sub> H <sub>15</sub> N <sub>2</sub> O <sub>2</sub> ClHg	503.09	Dark grey brown	210	53	1:1	12	Diamag.	38.16 (38.03)	2.98 (2.76)	5.56 (5.47)	39.87 (39.91)	7.05 (7.12)

Electronic spectra of the metal complexes show the transition is shifted towards higher or lower frequencies which confirm the coordination of the ligand to the metal ions. Also, electronic spectra of all complexes display adsorption band at 26629cm<sup>-1</sup>, 29498cm<sup>-1</sup>, 28571cm<sup>-1</sup> and 28328cm<sup>-1</sup> assigned ligand to metal charge transfer band [10, 11].

The IR spectra of the free ligand AMPIA exhibit a band in the region 3450cm<sup>-1</sup>. The intense band has disappeared in the complexes, indicating its involvement in the band formation process [12]. This has been further indicated by appearance of phenolic  $\nu$ (C-O) at 1259cm<sup>-1</sup> in ligand. This band is shifted towards lower frequency by 15-20cm<sup>-1</sup> in the spectra of complex indicates, that phenoxide oxygen has coordinated to the metal ion [13].

Besides, stretching frequency vibration of  $\nu$ (NH) at 3185cm<sup>-1</sup> in free ligand which on complexation shifted to lower wave number at 3171 to 3178 cm<sup>-1</sup> in all metal complexes which indicates -NH group involved in metal bonding.

Further, the band observed at  $1635\text{cm}^{-1}$  due to  $\nu(\text{C}=\text{N})$  azomethine in ligand and shifted in the complex towards lower frequency by  $10\text{-}20\text{cm}^{-1}$  attributed to involvement in coordinate bond [14].

A new band appeared at  $3410\text{cm}^{-1}$  due to the presence of coordinated water molecule in Cd(II) complex [15]. The appearance of new bands in the spectra of Ti(IV), Zr(IV), Cd(II) and Hg(II) metal ion complexes at  $525\text{-}545\text{cm}^{-1}$  and  $620\text{-}670\text{cm}^{-1}$  due to new bonding i.e.  $\nu(\text{M-N})$  [16, 17] and  $\nu(\text{M-O})$  [18, 19].

**Table-2: Infrared spectral data of ligand (ACBAI) and their Ti(IV), Zr(IV) and Cd(II) metal chelates.**

Compound	$\nu(\text{H}_2\text{O})$	$\nu(\text{OH})$	$\nu(\text{C-O})$	$\nu(\text{NH})$	$\nu(\text{N=H})$	$\nu(\text{M-N})$	$\nu(\text{M-O})$
AMPIA	-	3450	1259	3185	1635	-	-
[Ti(AMPIA) <sub>2</sub> ]Cl <sub>2</sub>	-	-	1237	3178	1615	529	640
[Zr(AMPIA) <sub>2</sub> ]Cl <sub>2</sub> .H <sub>2</sub> O	-	-	1240	3174	1620	525	645
[Cd(AMPIA)H <sub>2</sub> O]Cl.H <sub>2</sub> O	3410	-	1243	3176	1624	545	670
[Hg(AMPIA)Cl]H <sub>2</sub> O	-	-	1242	3171	1623	538	620

The <sup>1</sup>H-NMR spectrum of the ligand AMPIA exhibits signals at  $\delta$  2.31 (s, 3H) for methyl proton,  $\delta$  6.80-8.27 (m, 8H) due to Ar-H,  $\delta$  8.96 ppm (s, 1H) for azomethine proton [20],  $\delta$  10.1 ppm (s, 1H) NH of Indole ring [21] and  $\delta$  12.10 (s, 1H) due to Ar-OH. In Ti(IV), Zr(IV), Cd(II) and Hg(II) complexes phenolic OH peak at  $\delta$  12.10 ppm is disappeared due to attributed deprotonation and involvement in bonding [22]. A signal corresponding to azomethine in free ligand is shifted to downfield regions may be attributed to corresponding effect of the nitrogen atoms which results on deshielding of protons attached to it. All the metal complexes signal corresponding to NH of indole ring is shifted to down field at  $\delta$  10.20,  $\delta$  10.25,  $\delta$  10.28 and  $\delta$  10.29 respectively. A new peak observed at  $\delta$  2.0 ppm due to coordinated water molecule in only Cd(II) complex [23].

X-ray diffraction study of Ti(IV), Zr(IV), Cd(II) and Hg(II) metal chelates were scanned in the range  $2\theta = 10$  to  $90^\circ$ . With the help of X-ray diffraction technique it is possible as certain the spatial arrangement of the structural units substance in crystalline state and employed in investigating the interior of crystal.

The major reflexes were measured and corresponding 'd' values were obtained, independent indexing, calculation of miller indices and evaluation of lattice parameters a, b, c and angles  $\alpha$ ,  $\beta$ ,  $\gamma$  were done by power X programme. Data has been summarized in the following tables.

### X-ray analysis

#### [Ti (AMPIA)<sub>2</sub>] Cl<sub>2</sub>

Crystal system: Monoclinic

Lattice Type: P

Radiation: Cu

Wavelength:  $1.540598 \text{ \AA}$

Lattice Parameter: a= 4.9168 b= 4.9168 c= 5.4089  $\text{ \AA}$

Lattice Parameter: Alpha= 90 Beta= 90 Gamma=120 $^\circ$

2Theta Start= 10 2Theta End= 89.98

**Table No. 3: Powder XRD data of [Ti (AMPIA)<sub>2</sub>] Cl<sub>2</sub> Complex**

h	k	l	2 $\theta$	2 $\theta$	d	d	Intensity
			(Exp.)	(Calc.)	(Exp.)	(Calc.)	
0	0	1	16.292	16.375	5.43631	5.40890	62.04
0	0	1	16.464	16.375	5.37996	5.40890	64.14
-1	1	0	20.867	20.845	4.25355	4.25807	67.87
0	2	0	42.415	42.422	2.12937	2.12904	89.27
-2	0	1	45.794	45.763	1.97980	1.98109	83.81
1	1	2	50.118	50.104	1.81868	1.81913	84.19
0	0	3	50.623	50.585	1.80170	1.80297	85.99
0	0	2	54.818	54.835	1.67334	1.67286	83.54
0	1	3	55.377	55.286	1.65777	1.66027	83.15
-2	3	0	57.158	57.191	1.61026	1.60940	85.13

The cell data and crystal parameters of Ti(IV) complex is given in the table indicates that the complex have monoclinic crystal<sup>24</sup>.

**[Zr(AMPIA)<sub>2</sub>]Cl<sub>2</sub>.H<sub>2</sub>O**

Crystal system: Monoclinic

Lattice Type: P

Radiation: Cu

Wavelength: 1.540598 Å<sup>0</sup>

Lattice Parameter: a= 4.9168 b= 4.9168 c= 5.4089 Å<sup>0</sup>

Lattice Parameter: Alpha= 90 Beta= 90 Gama=120<sup>0</sup>

2Theta Start= 10 2Theta End= 89.98

**Table No. 4: Powder XRD data of [Zr(AMPIA)<sub>2</sub>]Cl<sub>2</sub>.H<sub>2</sub>O Complex**

h	k	l	2θ (Exp.)	2θ (Calc.)	d (Exp.)	d (Calc.)	Intensity (Exp.)
-1	1	1	26.616	26.622	3.34637	3.34573	54.45
0	0	2	33.028	33.097	2.70998	2.70445	46.56
-1	1	2	39.495	39.440	3.27983	2.28291	54.63
-1	2	1	40.240	40.263	2.23932	2.23807	48.00
0	2	0	42.426	42.422	2.12885	2.12904	52.78
-2	0	1	45.741	45.763	1.98199	1.98109	52.04
0	2	2	54.882	54.835	1.67153	1.67286	48.12
0	1	3	55.323	55.286	1.65925	1.66027	44.16
-2	3	0	57.198	57.191	1.60922	1.60940	40.08
-2	1	3	63.957	63.987	1.45450	1.45388	40.21
-3	1	2	67.679	67.692	1.38327	1.38303	41.02
-2	0	3	68.125	68.092	1.37530	1.37589	36.28
0	0	4	69.419	69.452	1.35279	1.35222	39.03
-1	0	4	73.377	73.409	1.28928	1.28880	40.20

Cell data and crystal lattice parameters of Zr(IV) complex attributed to monoclinic crystal system<sup>25</sup>.

**[Cd(AMPIA)H<sub>2</sub>O]Cl.H<sub>2</sub>O**

Crystal system: Orthorhombic

Lattice Type: P

Radiation: Cu

Wavelength: 1.540598 Å<sup>0</sup>

Lattice Parameter: a= 4.9168 b= 4.9168 c= 5.4089 Å<sup>0</sup>

Lattice Parameter: Alpha= 90 Beta= 90 Gama=90<sup>0</sup>

2Theta Start= 10 2Theta End= 89.98

**Table No. 5: Powder XRD data of [Cd(AMPIA)H<sub>2</sub>O]Cl.H<sub>2</sub>O Complex**

h	k	l	2θ (Exp.)	2θ (Calc.)	d (Exp.)	d (Calc.)	Intensity (Exp.)
0	1	0	17.993	18.027	4.92591	4.91680	63.00
1	1	0	25.596	25.601	3.47742	3.47670	51.40
1	1	1	30.581	30.542	2.92101	2.92464	49.01
0	2	0	36.497	36.520	2.45991	2.45840	51.31
2	2	0	52.644	52.606	1.73721	1.73835	41.45
1	0	3	54.159	54.137	1.69213	1.69275	41.06
2	2	1	55.481	55.478	1.65490	1.65498	39.02
0	3	1	58.842	58.826	1.56811	1.56851	36.13
1	3	1	62.057	62.060	1.49439	1.49431	28.17
2	3	0	68.820	68.787	1.36309	1.36367	35.00
0	0	4	69.481	69.452	1.35173	1.35222	33.03
2	2	3	76.021	75.982	1.25088	1.25142	40.02

Cell data and crystal lattice parameters of Cd(II) complex indicates that complex have orthorhombic crystal system<sup>26</sup>.

**[Hg(AMPIA)Cl]H<sub>2</sub>O**

Crystal system: Monoclinic

Lattice Type: P

Radiation: Cu

Wavelength: 1.540598 Å<sup>0</sup>

Lattice Parameter: a= 4.9168 b= 4.9168 c= 5.4089 Å<sup>0</sup>

Lattice Parameter: Alpha= 90 Beta= 90 Gama=90<sup>0</sup>

2Theta Start= 10 2Theta End= 89.98

**Table No. 6: Powder XRD data of [Hg(AMPIA)Cl]H<sub>2</sub>O Complex**

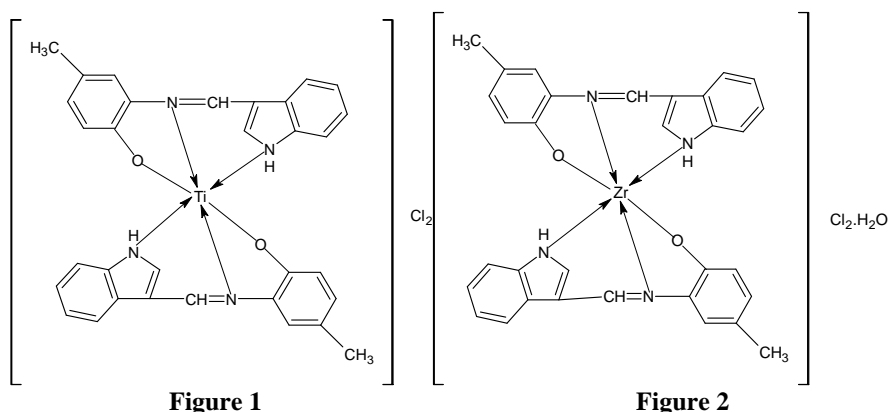
h	k	l	2θ (Exp.)	2θ (Calc.)	d (Exp.)	d (Calc.)	Intensity (Exp.)
0	0	1	16.364	16.375	5.41246	5.40890	56.97
-1	0	0	17.929	18.027	4.94345	4.91680	51.95
0	1	1	24.498	24.447	3.63078	3.63826	59.70
-1	1	0	25.676	25.601	3.46675	3.47670	50.56
-1	1	1	30.607	30.542	2.91851	2.92464	51.27
-1	1	2	42.262	42.305	2.13676	2.13466	86.16
2	0	2	50.196	50.104	1.81601	1.81913	49.09
0	0	3	50.538	50.585	1.80452	1.80297	44.20
-2	2	0	52.657	52.606	1.73678	1.73835	43.26
-1	2	2	53.993	53.679	1.70865	1.70610	41.07

Cell data and crystal lattice parameters of Hg(II) complex indicates that complex have monoclinic system<sup>27</sup>.

On the basis of the above observation it is, therefore, proposed that [Ti(IV) AMPIA], [Zr(IV) AMPIA] complexes shows octahedral geometry in which two ligands behaving as uninegative tridentate and [Cd(II) AMPIA], [Hg(II) AMPIA] complexes exhibits tetrahedral geometry with uninegative tridentate in nature.

The ligand (AMPIA) and their [Ti(IV) AMPIA], [Zr(IV) AMPIA], [Cd(II) AMPIA] and [Hg(II) AMPIA] complexes antimicrobial tests were performed by the standard disc diffusion method [28]. The antibacterial and antifungal activity in vitro against *Escherichia coli*, *Bacillus subtilis*, *Aspergillus flavus* and *Aspergillus niger*.

**Proposed structural formulae of the investigated metal complexes:**



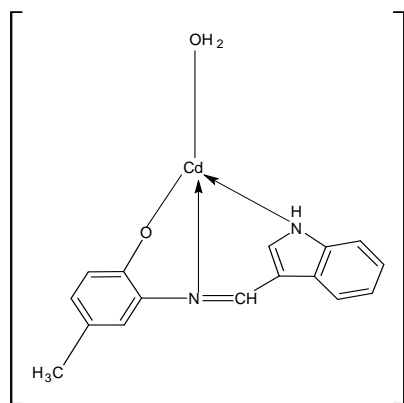


Figure 3

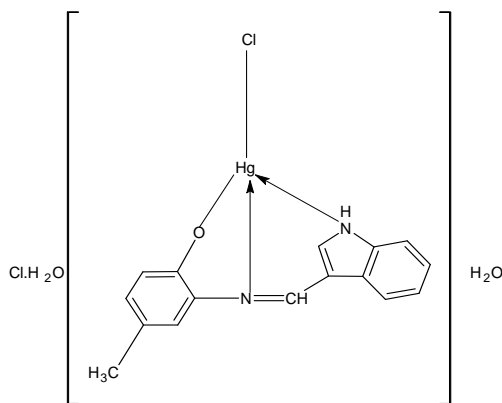


Figure 4

Table No.7: Result of antibacterial study of transition metal complexes

Sr. No.	Compounds	<i>Escherichia coli</i>	<i>Bacillus subtilis</i>
1	AMPIA	6 mm	3 mm
2	[Ti(AMPIA) <sub>2</sub> ] Cl <sub>2</sub>	-ve	-ve
3	[Zr(AMPIA) <sub>2</sub> ]Cl <sub>2</sub> .H <sub>2</sub> O	-ve	-ve
4	[Cd(AMPIA)H <sub>2</sub> O]Cl.H <sub>2</sub> O	15 mm	16 mm
5	[Hg(AMPIA)Cl]H <sub>2</sub> O	18 mm	26 mm
6	Streptomycin	24 mm	30 mm
7	Control (DMSO)	10 mm	14 mm

-ve –No antibacterial activity, zone of inhibition -----mm.

Table No.8: Result of antifungal study of transition metal complexes

Sr. No.	Compounds	<i>Aspergillus flavus</i>	<i>Aspergillus niger</i>
1	AMPIA	-ve	-ve
2	[Ti(AMPIA) <sub>2</sub> ] Cl <sub>2</sub>	-ve	-ve
3	[Zr(AMPIA) <sub>2</sub> ]Cl <sub>2</sub> .H <sub>2</sub> O	-ve	-ve
4	[Cd(AMPIA)H <sub>2</sub> O]Cl.H <sub>2</sub> O	15 mm	30 mm
5	[Hg(AMPIA)Cl]H <sub>2</sub> O	25 mm	26 mm
6	Fluconazole	15 mm	10 mm
7	Control (DMSO)	-ve	-ve

-ve –No antifungal activity, zone of inhibition -----mm.

Among four metal complexes [Cd(II) AMPIA] and [Hg(II) AMPIA] metal ion complexes produces more zone of inhibition towards *E. coli* and *B. subtilis* while [Ti(IV) AMPIA] and [Zr(IV) AMPIA] do not produce zone of inhibition or they have negative zone of inhibition.

The metal chelates of [Cd(II) AMPIA] and [Hg(II) AMPIA] acquires antifungal activities. Ti(IV), Zr(IV) metal chelates do not form zone of inhibition towards *Aspergillus flavus* and *Aspergillus niger* indicating negative antifungal activity.

### CONCLUSION

After conducting antibacterial and antifungal experiments towards as *Escherichia coli*, *Bacillus subtilis*, *Aspergillus flavus* and *Aspergillus niger* it indicates that [Cd(AMPIA)H<sub>2</sub>O]Cl.H<sub>2</sub>O and [Hg(AMPIA)Cl]H<sub>2</sub>O metal complexes produces more zone of inhibition than the Schiff base ligand. While [Ti(AMPIA)<sub>2</sub>] Cl<sub>2</sub> and [Zr(AMPIA)<sub>2</sub>]Cl<sub>2</sub>.H<sub>2</sub>O do not produce zone of inhibition or they have negative zone of inhibition.

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