



Synthesis, Structural Characterization and Anti-bacterial Activity of some Novel Schiff base Ligands and their Vanadium (IV) Metal Complexes

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ABSTRACT

A novel Schiff base ligands like OVPTH (*p*-Toulic Hydrazide and *o*-Vanilin Schiff base), andOHAPPTH (*p*-Toulic Hydrazide and *o*-Hydroxy acetophenone Schiff base) and its vanadium metal complexes have been synthesized and characterized by various physio chemical methods viz. FT-IR, ¹H-NMR, UV-Visible, ESR, VSM, Molar conductance, XRD and micro analytical data and found to be anti bacterial activity.

Key words: Schiffbase, antibacterial activity, characterization, vanadium (IV), OVPTH, OHAPPTH.

INTRODUCTION

A large number of Schiff bases and their metal complexes have been found to possess important biological and catalytic activity¹⁻³. These complexes are used as catalysts for water photolysis or reduction of oxygen at a modified carbon cathode. Schiff base have also been used for analytical purposes in the determination of metal ions and some Schiff base derivatives have been used in the extraction of metal solvents. The applications of such complexes depend to a large extent on their molecular structure. Schiff base ligands have an affinity for transition metals such as Cu, Mn, Co and Fe. Some of these complexes

have been studied⁴⁻⁵ in great deal for their various structures, steric effects and their coordination chemistry.

The main aim of the present paper is to prepare a new series of metal complexes of VO (IV), with Schiff base ligands derived from *p*-Toluic hydrazide and different aldehydes and ketones. These complexes were characterized by elemental analysis, IR, NMR, UV, ESR spectroscopy, TG-DTA, Powder XRD, VSM and Conductivity measurements to determine the mode of bonding, geometry and biological activities of the metal complexes⁶⁻⁸ were also studied. These complexes show square pyramidal or trigonal bipyramidal geometry.

EXPERIMENTAL

Materials and Methods

IR spectra were recorded with a Perkin-Elmer IR-598 spectrometer ($4000-400\text{ cm}^{-1}$) using KBr pellets. The ^1H NMR spectra of the ligands and their metal complexes are recorded on an AV-400 M-HZ NMR spectrometer in IICT, Hyderabad in $\text{DMSO-}D_6$ and CDCl_3 solvents at room temperature. The absorbances are measured on Elico SL UV-Visible spectrophotometer. The ESR spectra for all VO (IV) complexes were recorded at room temperature and at liquid nitrogen temperature (LNT) on a Bruker ESP 300E spectrometer in IIT Mumbai. The ESR spectrum obtained from the instrument is a plot of the first derivative of the absorption curve as a function of the magnetic field which was calibrated with an NMR gauss meter and the exact frequency was determined using DPPH radical as a field maker. Magnetic susceptibility data was recorded on an EG and G-155 magneto meter. Thermo gravimetric analyses of the metal complexes were carried out by using the Perkin Elmer System in thermal analysis center: STIC KOCHIN.

Synthesis of Schiff base ligands

Synthesis of *p*-toluic hydrazide and *o*-Vanilin Schiff base (OVPTH)

Mixture of *p*-Toluic hydrazide (0.01 moles) and *o*-Vanilin (0.01 moles) were dissolved in 50 ml methanol and added few drops of Tri ethyl amine. The whole mixture was transferred in to 250 ml round bottom flask. The mixture was refluxed about one and half hour on water bath. When the reaction mixture was allowed to cool light yellow crystals were obtained. The compound was recrystallized from methanol. The % of yield was 85% and melting point of the compound was $180-182^\circ\text{C}$.

Synthesis of *p*-Toluic hydrazide and *o*-Hydroxy Acetophenone Schiff base (OHAPPTH)

Mixture of *p*-Toluic hydrazide (1.50g) and *o*-Hydroxy Acetophenone (1.36mL) were dissolved in 30 ml methanol and added a pinch of NaOH. The whole mixture was transferred in to 100 ml round bottom flask. The mixture was refluxed about 2 hours on water bath. When the reaction mixture was allowed to cool light brown needles were obtained. The compound was recrystallized from water. The

%of yield was 83% and melting point of the compound was $188-190^\circ\text{C}$.

Synthesis of Schiff base Metal Complexes

Synthesis of OVPTH Schiff base-VO (IV) metal complex

The reagent *p*-Toluic hydrazide and *o*-Vanilin Schiff base (OVPTH) was dissolved in 40ml of methanol and VO (IV) metal ion dissolved in 10ml of distilled water. This mixture was stirred for 6 hours. A colored precipitate of complex was obtained with good yield. The product were washed several times with hot water and cold methanol to free from unreacted metal salts and ligand respectively and finally with ether and dried in vacuo over calcium chloride dessicator.

Synthesis of OHAPPTH Schiff base-VO (IV) metal complex

This metal complex were prepared by adding required amount of Schiff base (2g (0.0074moles)) in 50ml of methanol to the VO(IV) metal ion (0.0074moles) in distilled water in the presence of Sodium acetate and stirred about 10hours. Green precipitates of metal complex were obtained with good yield. The product were washed several times with hot water and cold methanol to free from unreacted metal salts and ligand respectively and finally with ether and dried in vacuo over calcium chloride dessicator.

RESULTS AND DISCUSSION

IR spectral studies

Interpretation of OVPTH ligand and its Vanadium complex

The infrared (IR) spectrum of the compound (OVPTH ligand and OVPTH-VO(IV) complex) is shown in figures 1 & 2. The Infrared spectrum of the ligand is compared with the spectrum of its vanadium complex. The important and specific IR spectral frequencies along with their assignment are given in Table 1.

A strong band exhibited at 1645 cm^{-1} in the IR spectrum of the ligand has been assigned to the (C=N) stretching vibration of the azomethine group. On complexation this band is shifted to 1600 cm^{-1} for OVPTH-VO(IV) complex. Bands appeared at 3215 and 1372 cm^{-1} due to the

stretching and bending vibrations of phenolic OH respectively.⁹⁻¹¹ These bands are disappeared in spectra of complexes indicating the deprotonation of phenolic OH. The IR spectrum of the ligand has shown a band in the region 1450-1550 cm^{-1} due to the C=C stretching vibrations. A broad signal is noticed at 3330 cm^{-1} in the IR spectrum of the ligand due to N-H stretching vibration. On complexation this band shifted to 3310 cm^{-1} for VO(IV) complex. The IR spectrum of the ligand has shown a sharp

band at 1711 cm^{-1} due to C=O stretching vibration. These results indicate the formation of complex.

Interpretation of OHAPPTH ligand and its Vanadium complex

The Infrared spectrum of the OHAPPTH ligand is compared with the spectra of its VO(IV) complex. A strong band exhibited at 1654 cm^{-1} , 3191 cm^{-1} which was assigned to C=N and Phenolic OH respectively. These bands are disappeared in

Table 1: Important IR Spectral bands of OVPTH and OHAPPTH ligands and its VO (IV) metal complexes

Compound	OH Water	OH Phenolic	C=N	N-H	C=O	M-O	M-N
OVPTH	-	3215	1645	3330	1711	-	-
OVPTH-VO (IV)	3542	-	1600	3310	1680	582	486
OHAPPTH	-	3192	1654	3320	1704	-	-
OHAPPTH-VO (IV)	3415	-	1597	3331	1682	633	429

Table 2: Important ¹H NMR Spectral data of OVPTH and OHAPPTH ligands and its VO (IV) metal complexes

Compound	H-C=N	CH ₃ -C=N	Ar-H	CH ₃	OCH ₃	Ar-OH	N-H
OVPTH	8.69		6.80-7.86	2.4	3.81	12.04	11.07
OVPTH-VO (IV)	8.95		6.92-7.93	2.53	3.83	-	11.16
OHAPPTH	-	2.57	6.94-7.89	2.41	-	13.51	11.25
OHAPPTH-VO (IV)	-	2.63	6.93-8.15	2.49	-	-	11.36

Table 3: ESR spectral data of OVPTH-VO (IV) and OHAPPTH-VO (IV) complexes

Parameters	OVPTH-VO	OHAPPTH-VO
$g_{ }$	1.95243	1.93126
g_{\perp}	1.98831	1.98612
g_{ave}	1.97635	1.96783
G	4.07626	4.95244
$A_{ }^*$	0.01907	0.01882
A_{\perp}^*	0.00664	0.00667
A_{ave}^*	0.01078	0.01072
d-d	17535	17622
KQ%	0.623	0.635
K_{\perp}	0.749	0.729
P^*	0.0196	0.0197
α^2	0.637	0.539

spectra of complexes indicating the deprotonation of phenolic OH. This is further confirmed by the appearance of new bands in the region of 509 and 648 cm^{-1} , which are assigned to the stretching frequencies of M-N and M-O of the metal ligand bands for VO (IV) complex. On complexation this band shifted to 3331 for VO(IV) complex. The IR spectrum of the ligand has shown a sharp band at 1704 cm^{-1} due to C=O stretching vibration. On complexation this band shifted to 1682 cm^{-1} for VO (IV) complex. These results indicate the formation of complex.

NMR Spectral Studies

Interpretation of NMR spectra of OVPTH ligand and its Vanadium complex

Figs 3 and 4 are shows the NMR spectra of the OVPTH ligand and its VO(IV) complex. Table

Table 4: Thermal Analytical Data of the OVPTH-VO (IV) and OHAPPTH-VO (IV) vanadium metal complexes

Complex X=H ₂ O	Molecular Weight in grams	Weight of the complex taken in mgs	Stage	Temperature range in °C	Probable assignment	Mass loss (%)	Total mass loss (%)
VOL ₂ X	671.586	7.00	1	70.43 to 120.29	Loss of 2H ₂ O molecules	5.360	84.53
			2	251.56 to 570.81	Loss of 2L molecules	79.17	
			3	Above 789.39	Corresponds to VO	15.07	
L=C ₁₆ H ₁₆ O ₃ N ₂	637.570	7.20	1	65.32 to 107.91	Loss of 2H ₂ O molecules	5.646	83.86
			2	280.15 to 583.69	Loss of 2L molecules	78.213	
			3	Above 612.59	Corresponds to VO	15.92	

Table 5: Analytical data of the OVPTH and OHAPPTH ligands and its Vanadium complexes

Molecular Formula X=H ₂ O	Molecular Weight	Colour	Yield in %	Melting Point in °C	Elemental analysis							
					Carbon %		Hydrogen %		Nitrogen %		Metal %	
					Calc.	Found	Calc.	Found	Calc.	Found	Calc.	Found
L=C ₁₆ H ₁₆ O ₃ N ₂ (OVPTH)	285.33	Light yellow	85	180-182	67.29	66.78	5.60	5.35	9.81	9.52	-	-
[VO L ₂]2X(OVPTH-VO)	671.586	orange	69	298-312	57.17	56.49	5.06	4.67	8.39	8.09	7.58	7.03
L=C ₁₆ H ₁₆ O ₂ N ₂ (OHAPPTH)	268.315	Light brown	85	188-190	71.55	71.21	5.96	5.42	10.43	10.02	-	-
[VO L ₂]2X(OHAPPTH-VO)	637.57	Green	72	302-312	60.23	59.84	5.33	4.99	8.78	7.99	7.99	7.15

Table 6: X-ray Diffraction data of OHAPPTH-VO (IV) complex

S.No.	d expt	d Calc	2 θ expt	Calc	h k l
1.	7.323	7.329	12.06248	12.073	2 1 1
2.	14.185	14.198	6.23856	6.244	4 2 2
3.	14.516	14.529	6.09709	6.103	4 2 2
4.	15.607	15.621	5.67325	5.678	4 3 2
5.	16.899	16.915	5.24221	5.247	4 4 1
6.	18.037	18.054	4.91407	4.919	5 3 2
7.	19.493	19.511	4.55014	4.554	5 3 3
8.	20.038	20.057	4.42757	4.432	6 3 1
9.	20.256	20.275	4.38047	4.384	6 3 1
10.	21.781	21.801	4.0771	4.081	5 5 2
11.	22.856	22.877	3.88772	3.891	5 5 3
12.	23.089	23.110	3.84904	3.853	6 4 3
13.	23.599	23.621	3.76691	3.770	6 5 1
14.	23.881	23.903	3.72321	3.727	6 2 1
15.	24.558	24.580	3.62203	3.625	6 5 3
16.	25.826	25.850	3.44702	3.450	6 6 2
17.	27.036	27.061	3.29536	3.298	7 5 3
18.	29.028	29.055	3.07361	3.076	7 6 3
19.	30.498	30.526	2.92877	2.931	8 6 2
20.	31.368	31.397	2.84945	2.852	7 6 5
21.	32.314	32.344	2.76815	2.771	8 6 3
22.	36.617	36.652	2.45212	2.454	8 7 6

2 contains the important chemical shift values along with their assignments. The signals appeared at 8.69, 2.41, 3.81 ppm was assigned to azomethine (H-C=N) proton, methyl protons and methoxy protons respectively. A multiplet is observed in the region 6.8-7.86 due to the aromatic protons. A singlet appeared at 12.04 ppm is attributed to the hydroxyl proton. The singlet appeared at 11.07 ppm due to N-H proton of ligand.

In the ^1H NMR spectrum of the OVPTH-VO (IV) complex shows the signals at 8.95, 2.53 and 6.92-7.91 for azomethine proton, methyl protons and aromatic protons respectively¹²⁻¹⁵. The singlet appeared at 12.04 ppm due to phenolic hydroxyl proton is absent in the NMR spectrum of VO complex indicating the deprotonation of hydroxyl group and the involvement of that oxygen in coordination.

Table 7: Antibacterial activities of ligands and their transitions metal complexes (Zone of inhibition in mm)

Compound	<i>E. Coli</i>	<i>B. Subtilis</i>
OVPTH ligand	7	6
OVPTH-VO (IV)	11	8
OHAPPTH ligand	6	6
OHAPPTH-VO (IV)	12	9
Streptomycin	22	24

Interpretation of NMR spectra of OHAPPTH ligand and its Vanadium complex

The ^1H NMR spectrum of the OHAPPTH ligand shows signals at 2.57, 2.41 and 6.94-7.89 for methyl protons of azomethine group, methyl protons and aromatic protons of phenyl ring respectively. A singlet appeared at 13.51 ppm is attributed to the hydroxyl proton attached to the phenyl ring in the ligand. The singlet appeared at 11.25 ppm due to N-H proton of ligand.

In the ^1H NMR spectrum of the OHAPPTH-VO complex signals are noticed at 2.63, 2.49 and

6.93-8.15 for methyl protons of azomethine group, methyl protons and aromatic protons of phenyl ring respectively. The singlet appeared at 13.51 ppm due to phenolic hydroxyl proton is absent in the NMR spectrum of VO complex indicating the deprotonation of hydroxyl group and the involvement of that oxygen in coordination. A signal observed at 11.25 ppm due to the N-H proton in the ligand is shifted to 11.36 ppm for the VO complex.

UV Spectra

The UV-Visible spectra of the ligands and its metal complexes were recorded in DMF. The electronic spectra of the aqueous solutions of V^{+4} ions are compared with the corresponding ligands nature. The transitions of these ligands occurred at 303nm, 291nm respectively for OVPTH and OHAPPTH. But on complexation with vanadium metal ion new bands appeared at 410 nm and 405 nm respectively corresponding to the transitional charge transfer from the different ligands to the vanadium metal ion. Bands occurred in the region of 403-410 nm for all complexes are assigned to charge transfer transition (L!M). Based on the results square pyramidal or trigonal bipyramidal structure is proposed for VO (IV) complexes.

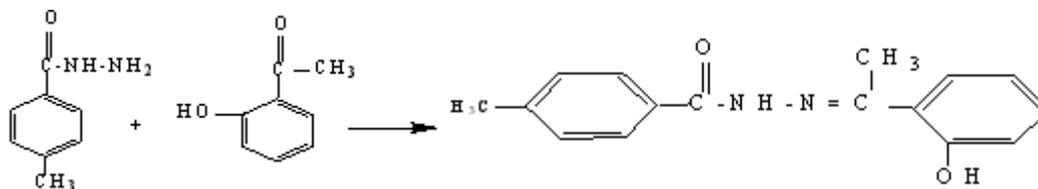
ESR Spectral studies

The X-band ESR spectra of OVPTH-VO(IV) and OHAPPTH-VO(IV) complexes at room temperature shows normal eight lines isotropic

features revealing hyperfine splitting of the ^{51}V nucleus $I = 7/2$. The absence of the super hyperfine splitting in this spectrum at room temperature and the trend of g values calculated from the spectrum ($g_{||} < g_{\perp} < 2$) show that the unpaired electron (d^1) is in the d_{xy} orbital localized on the metal. The spin Hamiltonian, orbital reduction and bonding parameters of OVPTH-V(IV) and OHAPPTH-VO(IV) complexes are presented in Table 3. Figs 5. Show the ESR spectrum of the OHAPPTH-VO (IV) complex. The values are typical of the spectra displayed by trigonal bipyramidal or square pyramidal VO (IV) complex with one unpaired electron in an orbital of mostly d_{xy} character. For the present OVPTH-VO(IV) and OHAPPTH-VO(IV) complexes G values are greater than four it is 4.07626 and 4.95244 respectively, which suggest that there are no interactions between Vanadium-Vanadium centers in DMF medium. The α^2 values of these complexes are 0.637 and 0.539 respectively. It indicates that the complex have covalent character. This shows an appreciable covalency in the inplane σ bonding. Spin Hamiltonian Para meters A_{\perp}^* and $A_{||}^*$ values are obtained from the spectrum (Figure.5) by taking the seventh parts of the magnetic induction in the parallel and perpendicular range from $m = +7/2$ to $-7/2$ respectively. $A_{\perp}^* > A_{||}^*$ is the general trend for trigonal bipyramidal or square pyramidal. The ESR parameters g_{\perp} , $g_{||}$, A_{\perp}^* and $A_{||}^*$ of the complexes and the energies of d-d transitions are used to evaluate the orbital reduction parameters (K_{\perp} , $K_{||}$),



Scheme 1: Synthesis of *p*-toluic hydrazide and *o*-Vanillin Schiff base



Scheme 2: Synthesis of *p*-Toluic hydrazide and *o*-Hydroxy Acetophenone Schiff base

the bonding parameters (α^2), the dipolar interaction (P). The trend of K_A , K_{V+} values of OVPTH-VO(IV) and OHAPPTH-VO(IV) complexes are $K_I < K_{II}$ indicates the presence of out of plane p-bonding. Girdano and Bereman suggested the identification¹⁶ of bonding groups from the values of dipolar term P. The reduction of P values from the free ion value (0.036cm^{-1}) might be attributable to the strong covalent bonding. The value of P obtained for the present complexes are 0.0196 and 0.0199 respectively. The shape of ESR lines, ESR data together with the electronic spectral data suggest trigonal bipyramidal or square pyramidal geometry for OVPTH-VO (IV) and OHAPPTH- VO (IV) complexes.

Magnetic susceptibility

The magnetic properties of Vanadium complexes help to know the geometry of them. The magnetic properties of these Vanadium complexes are 1.76 B.M. and 1.75 B.M. respectively¹⁷⁻¹⁸ for OVPTH-VO(IV) and OHAPPTH- VO (IV). This result clearly indicates that the vanadium complexes of these ligands show square pyramidal or trigonal bipyramidal geometry.

Molar conductivity

The molar conductance data of complexes which was carried out in DMF at 10^{-3} . The molar conductance values are 5.10 and 6.20 $\text{ohm}^{-1}\text{cm}^2\text{mol}^{-1}$ respectively for VPTH-VO(IV) and OHAPPTH- VO(IV) and RAPPTH- VO(IV). These

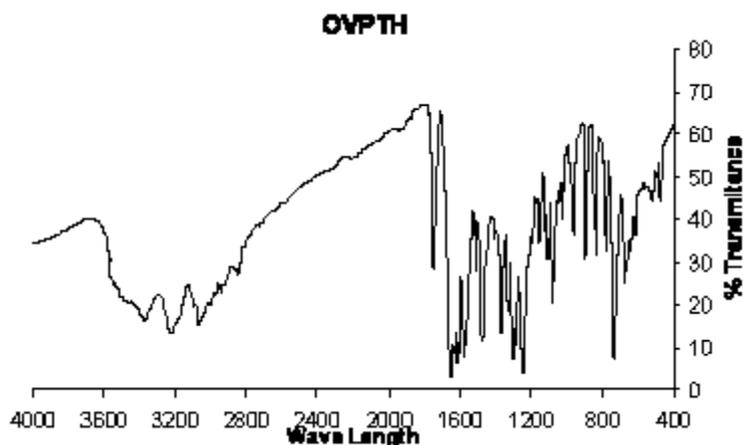


Fig. 1: IR Spectrum of OVPTH Ligand

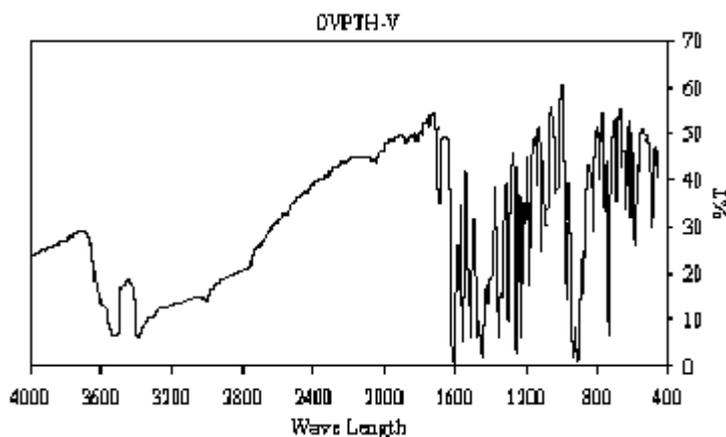


Fig. 2: IR Spectrum of OVPTH-V Complex

values suggest non-electrolytic nature of the present complexes.

Thermo gravimetric studies

The Thermo gravimetric studies of all the complexes ¹⁹⁻²¹ were carried out in air at a heating rate of 10°C per minute. The thermo analytical data

is summarized in Table.4. The thermal decomposition of the complexes proceeds in three stages. The OVPTH- VO (IV) and OHAPPTH- VO(IV) complexes are thermally stable up to 70.43 and 65.32 °C respectively. The first stage of decomposition corresponding to endothermic dehydration of complexes by the loss of two water



Fig. 3: Proton NMR Spectrum of OVPTH Ligand

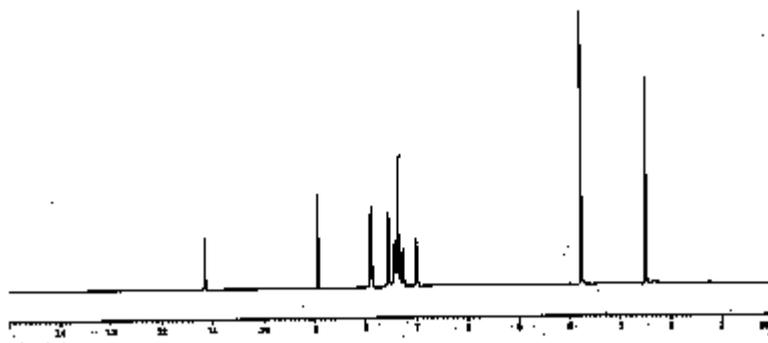


Fig. 4: Proton NMR Spectrum of OVPTH-V Complex

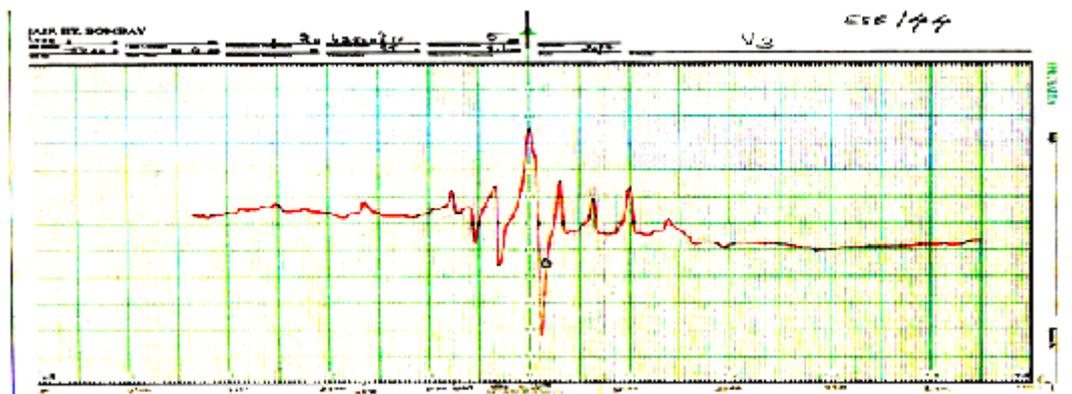


Fig. 5: ESR Spectrum of OHAPPTH-VO Complex

molecules occurs in the temperature range 70-120°C and 66-107°C respectively. The intermediates formed are stable up to 251 and 280°C. The second decomposition with exothermic peak by the loss of ligand moiety occurs in the temperature range 251-570 and 280-583°C. The solid residues above 789 and 612 °C were identified as OVPTH-VO(IV) and OHAPPTH-VO(IV) metal oxides respectively. In all the complexes, the final products are metal oxides.

The Powder XRD Studies

The powder X-ray diffraction data obtained for OVPTH-VO(IV) and OHAPPTH-VO(IV) complexes with diffractograms using DROL-2 powder diffractometer. Radiation filled by metal foil. The diffractograms (13 diffractions) reflects Fig.6. between 5-80 (2 θ) values for OVPTH-VO(IV) complex and (22 diffractions) for OHAPPTH-VO(IV). Where θ is Bragg's angle all the main peaks are indicated and calculated values of Miller indices (h k l)

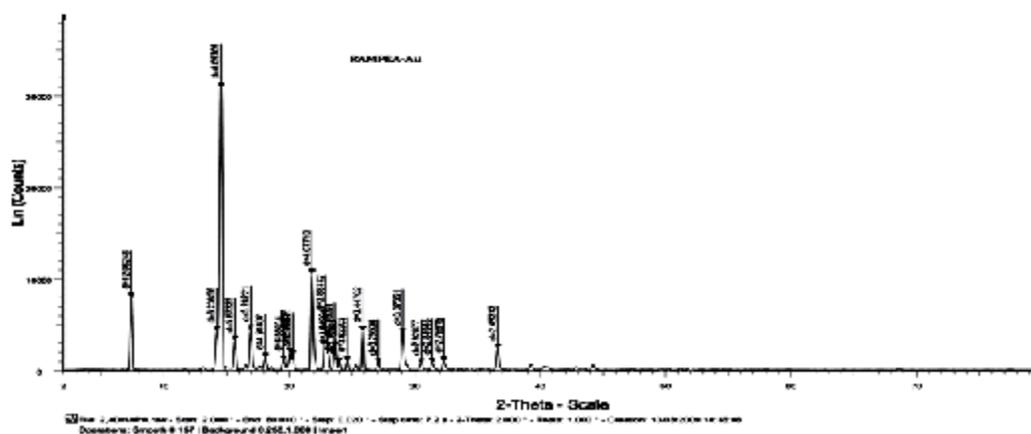


Fig. 6: XRD Spectrum of OHAPPTH-VO Complex

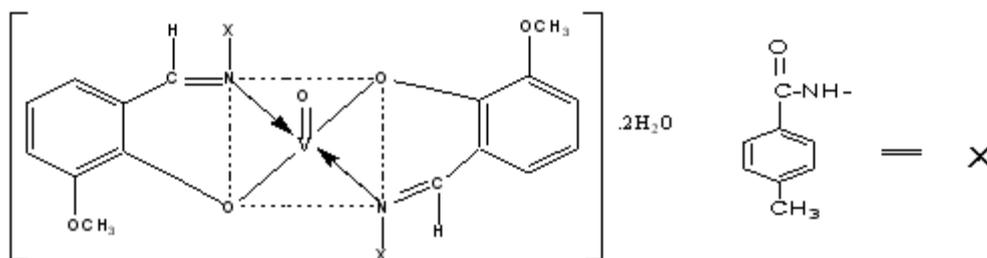


Fig. 7: VPTH-VO(IV) Complex

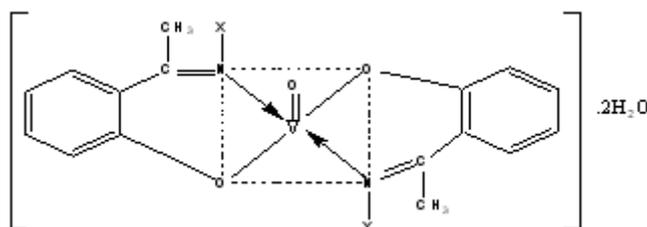


Fig. 8: OHAPPTH-VO(IV) Complex

l) along with observed d-specified and revealed intensities are specified in the Fig: 6. All the peaks have been indexed 2θ values compared in graph. Comparison values reveals that there is good agreement between values of 2θ and d-values. The powder x-ray diffraction²²⁻²³ data showed identical features with very poor crystallinity. The patterns are qualitative and dispersive in intensity for OVPTH-VO(IV) and OHAPPTH-VO(IV) complexes. The XRD patterns are used to explain qualitatively the degree of crystallinity. X-ray Diffraction data of OHAPPTH-VO(IV) complex are presented in Table.6.

Antibacterial Activity of OVPTH and OHAPPTH ligands and its vanadium (IV) Metal Complexes Paper disc diffusion technique

The sterilized (autoclaved at 120°C for 30 min) medium (40–50°C) was inoculated (1 ml/100 ml of medium) with the suspension (5×10^{-5} cfu/ml) of the microorganism (matched to McFarland barium sulphate standard) and poured into a petridish to give a depth of 3–4 mm. The paper impregnated with the test compounds (100 µg/ml in dimethyl formamide) was placed on the solidified medium. The plates were pre-incubated for 1 h at room temperature and incubated at 37°C for 24 and 48 h for antibacterial and antifungal activities, respectively. Streptomycin (100 µg/ml/disc) was used as standard for anti-bacterial activity. A comparison of antibacterial activity of the synthesized

compounds with that of standard drugs was effectively²⁴⁻²⁷ presented in Table 7. In this study, all the synthesized compounds were evaluated for their *in vitro* antibacterial activity against two bacterias namely *E.coli* and *B. subtiles*. Streptomycin was used as a reference drug. The antibacterial activity results indicate that most of the metal chelates exhibited good to moderate antibacterial activity when compared to streptomycin. Among the synthesized metal complexes OHAPPTH-VO (IV) showed high activity.

CONCLUSION

In the present research studies, our efforts are synthesized of some newly compounds. These synthesized compounds characterized by various physio- chemical and spectral analysis. By using above all spectral studies it is concluded that they behave as bidentates during complexation. The IR data of both Schiff base and its vanadium metal complexes show that the OVPTH and OHAPPTH Schiff bases are coordinate to the metal ion in bidentate manner with ON donor sites of azomethaine nitrogen and phenolic oxygen. The ESR spectras and data suggest trigonal bipyramidal or square pyramidal structures for OVPTH-VO(IV) and OHAPPTH-VO(IV) Complexes. On the basis of chelation theory, metal complexes have more biological activity than free ligands.

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