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Synthesis, Characterization, and Antibacterial Studies of Oxovanadium(IV) Complexes with Thiazole-Derived Schiff Bases

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SYNTHESIS, CHARACTERIZATION, AND ANTIBACTERIAL STUDIES OF OXOVANADIUM(IV) COMPLEXES WITH THIAZOLE-DERIVED SCHIFF BASES

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Oxovanadium(IV) complexes of Schiff bases, derived from 2-amino-4-phenyl thiazole/substituted 2-amino-4-phenyl thiazoles and thiophene-2-aldehyde have been synthesized and characterized on the basis of elemental analysis, molar conductance measurements, magnetic susceptibility data, and UV-visible, and IR spectral studies. All the complexes are monomeric possessing a 1:2 (metal:ligand) stoichiometry. On the basis of these data, a square pyramidal geometry has been assigned for the complexes. A few complexes have been subjected to thermal decomposition studies. The ligands and their metal complexes have been screened for their antibacterial activities.

Supplemental materials are available for this article. Go to the publisher's online edition of Phosphorus, Sulfur, and Silicon and the Related Elements to view the free supplemental file.

Keywords 2-Amino-4-phenylthiazole; antibacterial activity; oxovanadium(IV); square pyramidal geometry; thermal decomposition; thiophene-2-aldehyde

INTRODUCTION

Transition metal complexes of Schiff bases have received considerable attention, mainly due to their preparative accessibility, structural diversity, and wide range of applications in various fields.^{1–3} Recently, metal complexes of ligands containing nitrogen and other donor atoms have attained much interest due to their remarkable biological activities.^{4,5} Among these ligands, thiazole and its derivatives have been well studied owing to their significant antifungal, antibacterial, and anticancer activities.^{6–9} Recognition of thiazole and its derivatives in vitamin B and coenzyme carboxylase has generated increasing interest in their structural and functional properties and has created an exciting topic for research.^{10,11}

Among the transition elements, vanadium is versatile in forming complexes with relevant biological properties and possesses a number of stable and accessible oxidation states. It has been observed that there is a series of nitrogenase metalloenzymes that have vanadium at their active sites.^{12,13} Apart from this, insulinomimetic potency of vanadium(IV)

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Table I Analytical data and other details of the ligands

Compound/ molecular formula	Color	Yield (%)	Analytical data calculated (found)			
			C	H	N	S
L ¹	Yellow	76	62.22	3.72	10.36	23.71
C ₁₄ H ₁₀ N ₂ S ₂			(62.35)	(3.60)	(10.27)	(23.60)
L ²	Brown	78	58.74	3.52	9.78	22.40
C ₁₄ H ₁₀ N ₂ S ₂ O			(58.68)	(3.59)	(9.85)	(22.31)
L ³	Yellowish brown	76	60.00	4.02	9.36	21.35
C ₁₅ H ₁₂ N ₂ S ₂ O			(60.09)	(4.08)	(9.42)	(21.42)
L ⁴	Yellowish brown	75	53.35	2.87	20.35	20.35
C ₁₄ H ₉ N ₃ S ₂ O ₂			(53.22)	(2.75)	(20.38)	(20.43)
L ⁵	Yellow	77	55.15	2.97	9.18	21.03
C ₁₄ H ₉ N ₂ S ₂ Cl			(55.24)	(2.88)	(9.29)	(21.09)
L ⁶	Yellowish brown	74	48.13	2.59	8.01	18.35
C ₁₄ H ₉ N ₂ S ₂ Br			(48.02)	(2.65)	(8.10)	(18.25)
L ⁷	Yellow	78	63.37	4.25	9.85	22.56
C ₁₅ H ₁₂ N ₂ S ₂			(63.42)	(4.16)	(9.76)	(22.45)

compounds has stimulated further investigation into vanadium complexes.^{14,15} However the chemistry of oxovanadium complexes with Schiff bases derived from heterocyclic amines and heterocyclic aldehydes has received less attention so far, and most parts of the biochemistry of oxovanadium complexes remain obscure. In this article, we describe the synthesis, spectroscopic characterization, thermal decomposition, and antibacterial studies of some oxovanadium(IV) complexes of Schiff bases formed by the condensation of 2-amino-4-phenylthiazole or its derivatives with thiophene-2-aldehyde.

RESULTS AND DISCUSSION

Structure of the Ligands

The structure of the ligands was established by UV, IR, and ¹H NMR spectral data. Analytical data and other details of ligands obtained are given in Table I. The electronic spectra of the ligands exhibited two absorption bands at ~290 and ~330 nm, which are attributed to $\pi-\pi^*$ and $n-\pi^*$ transitions of the C=N chromophore, respectively.¹⁶

The infrared spectral data of the ligands are presented in Table II. The free ligands showed absorption bands in the range ~1650 cm⁻¹, characteristic of the azomethine $\nu(\text{C}=\text{N})$ group.¹⁷ The band due to the thiazole ring $\nu(\text{C}=\text{N})$ was observed in the range 1600–1610 cm⁻¹, and the vibrational characteristics of the thiazole ring have been found at ~2600 cm⁻¹.^{18,19} The C–S–C group vibrations of the thiazole ring in the free ligands occur in the range 701–708 cm⁻¹.²⁰ In the spectra of the ligands, vibrational characteristics of the substituted thiophene ring occur around 3100, 1590, and 1405 cm⁻¹, and bands due to $\nu(\text{C}=\text{S}-\text{C})$ of the thiophene moiety occur in the range 830–850 cm⁻¹.^{21,22}

The ¹H NMR spectra of the ligands recorded in DMSO-d₆ gave added support to the conclusions drawn on the basis of UV and IR spectral data. The spectral assignments are detailed in Table III. The singlet observed at 7.25–7.39 ppm in the spectra of the ligands has been assigned to the azomethine proton.²³ The thiazole ring C–H resonances are observed at 7.20–7.23 ppm as a singlet.²⁴ The four hydrogen atoms of the phenyl ring give two doublets, one at 8.17–8.02 ppm and another at 8.04–7.87 ppm, except for L₁. The phenyl

Table II Important IR spectral bands of the ligands and complexes

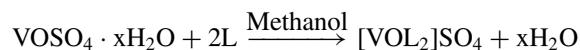
Compound	$\nu(\text{C}=\text{N})$	Thiazole $\nu(\text{C}=\text{N})$	Thiazole $\nu(\text{C}-\text{S}-\text{C})$	Thiophene $\nu(\text{C}-\text{S}-\text{C})$	$\nu(\text{V}=\text{O})$	$\nu(\text{M}-\text{N})$
L ¹	1645	1612	708	850	–	–
L ²	1649	1605	704	838	–	–
L ³	1648	1610	706	834	–	–
L ⁴	1645	1605	704	841	–	–
L ⁵	1648	1602	703	831	–	–
L ⁶	1650	1601	704	831	–	–
L ⁷	1655	1611	701	840	–	–
[VO(L ¹) ₂]SO ₄	1630	1600	706	852	969	405
[VO(L ²) ₂]SO ₄	1629	1590	707	836	972	408
[VO(L ³) ₂]SO ₄	1630	1595	704	838	973	407
[VO(L ⁴) ₂]SO ₄	1625	1594	705	844	969	411
[VO(L ⁵) ₂]SO ₄	1635	1597	707	834	973	410
[VO(L ⁶) ₂]SO ₄	1630	1595	708	834	972	409
[VO(L ⁷) ₂]SO ₄	1638	1590	704	842	970	412

protons of L₁ resonate as a singlet at 8.1 ppm, and the thionyl protons resonate as multiplets in the range 4.21–5.49 ppm.²⁵

On the basis of the above spectral data, the structure shown in Figure 1 has been assigned to the free ligands.

Structure of Metal Complexes

The reaction of oxovanadium(IV) sulfate with the Schiff bases (L¹–L⁷) in the molar ratio 1:2 gave the respective complexes. The formation of the complexes can be represented as follows.

**Table III** ¹H NMR data of the ligands

Compound	¹ H NMR (DMSO-d ₆) (ppm)
L ¹	7.20 (s, 1H, C–H thiazole), 7.33 (s, H, HC=N), 8.10 (s, 5H, aromatic), 4.26–5.41 (m, thionyl)
L ²	7.25(s, 1H, C–H thiazole), 7.28(s, HC=N), 8.05 (d, 2H, aromatic), 7.91 (d, 2H, aromatic), 4.28–5.46 (m, thionyl), 9.82 (s, H, OH)
L ³	7.26 (s, 1H, C–H thiazole), 7.25 (s, HC=N), 8.02 (d, 2H, aromatic), 7.87 (d, 2H, aromatic), 4.21–5.43 (m, thionyl), 3.45 (s, 3H, OCH ₃)
L ⁴	7.23 (s, 1H, C–H thiazole), 7.39 (s, H, HC=N), 8.17 (d, 2H, aromatic), 8.04 (d, 2H, aromatic), 4.26–5.44 (thionyl)
L ⁵	7.21 (s, 1H, C–H thiazole), 7.31 (s, H, HC=N), 8.06 (d, 2H, aromatic), 8.00 (d, 2H, aromatic), 4.26–5.45 (m, thionyl)
L ⁶	7.22 (s, 1H, C–H thiazole), 7.32 (s, H, HC=N), 8.00 (d, 2H, aromatic), 8.02 (d, 2H, aromatic), 4.27–5.44 (m, thionyl)
L ⁷	7.25 (s, 1H, C–H thiazole), 7.30 (s, H, HC=N), 8.07 (d, 2H, aromatic), 7.96 (d, 2H, aromatic), 4.22–5.48 (m, thionyl), 2.46 (s, 3H, CH ₃)

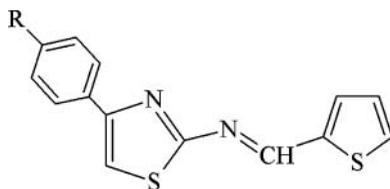


Figure 1 Structure of the ligand (For L^1 -R=H, L^2 -R=OH, L^3 -R=OCH₃, L^4 -R=NO₂, L^5 -R=Cl, L^6 -R=Br, L^7 -R=CH₃)

The complexes formed are sparingly soluble in common organic solvents, whereas they possess good solubility in DMSO. All the complexes decompose above 250°C. Formulations of these complexes have been done on the basis of elemental analysis, molar conductance, magnetic susceptibility measurements, and various spectral data. Analytical data and other details of the complexes obtained are listed in Table IV. The analytical data of the complexes are in accordance with the general formula $[\text{VOL}_2]\text{SO}_4$, where $L = L^1$ - L^7 . Moreover, the molar conductance values of the complexes measured in DMSO were obtained in the range 53.2–58.1 $\Omega^{-1} \text{cm}^2 \text{mol}^{-1}$ (Table IV). This adequately confirmed the electrolytic nature of the Complexes.²⁶

In order to determine the coordination sites involved in complexation, the infrared spectra of the complexes were compared with those of the free ligands. In all the complexes, the absorption bands characteristic of the azomethine $\nu(\text{C}=\text{N})$ group have been shifted to lower frequencies by 15–20 cm^{-1} , indicating coordination of azomethine nitrogen with the metal ion.²⁷ The band due to $\nu(\text{C}=\text{N})$ of the thiazole ring has been displaced to a lower frequency, by about 10–15 cm^{-1} in the complexes, indicating coordination by the thiazole ring nitrogen.¹⁸ In addition, the frequency at $\sim 2600 \text{cm}^{-1}$ attributed to the thiazole ring is shifted to $\sim 2500 \text{cm}^{-1}$ in metal complexes, which gives added support to the coordination of the endocyclic nitrogen.¹⁹ In the spectra of the metal complexes, the bands due to substituted thiophene ring remained almost unaffected, indicating the nonparticipation of the heterocyclic ring sulfur atom of the thiophene ring during coordination. Moreover, the bands due to $\nu(\text{C}-\text{S}-\text{C})$ of the thiophene moiety that persist in the complexes confirmed the noncoordination of thiophene ring sulfur.²² In addition to these observations, absence of $\nu(\text{M}-\text{S})$ bands in the far IR spectra of the metal complexes agreed with the non-involvement of sulfur atoms in chelation.

In all the metal complexes, the band occurring in the region 969–973 cm^{-1} has been assigned to the $\nu(\text{V}=\text{O})$ vibration, and this value is in the range observed for monomeric oxovanadium(IV) complexes.²⁸ The presence of the ionic sulfate group in the complexes has been confirmed by the appearance of bands at $\sim 1100 \text{cm}^{-1}$ and $\sim 616 \text{cm}^{-1}$ in the spectra of the complexes.²¹

The nonligand broad bands with medium intensities in the range 405–412 cm^{-1} observed in the far IR spectra of the complexes could be tentatively assigned for $\nu(\text{M}-\text{N})$ stretching vibrations.²⁹ Thus it is clear that the ligand is coordinated to the metal ion in a bidentate fashion through the azomethine nitrogen and thiazole ring nitrogen.

Electronic Spectra and Magnetic Moment

The assignments for the electronic spectrum of $\text{VOSO}_4 \cdot \text{XH}_2\text{O}$ had been made using extended the Huckel molecular orbital treatment.³⁰ The band in the region

Table IV Analytical data and other details of the complexes

Compound/ molecular formula	Color	Yield%	Analytical data calculated (found)						Molar conductance $\Omega^{-1}\text{cm}^2\text{mol}^{-1}$	Magnetic moment (BM)
			C	H	N	S	V			
$[\text{VO}(\text{L})_2]\text{SO}_4$	Green	65	47.80(47.72)	2.84(2.89)	7.96(7.85)	22.79(22.88)	7.24(7.12)	53.2	1.73	
$[\text{VO}(\text{L}^2)_2]\text{SO}_4$	Brown	67	45.72(45.60)	3.01(3.13)	7.61(7.69)	21.79(21.89)	6.92(6.85)	57.6	1.75	
$[\text{VO}(\text{L}^3)_2]\text{SO}_4$	Green	56	47.19(47.30)	3.43(3.55)	7.36(7.44)	19.17(19.29)	6.67(6.55)	58.1	1.74	
$[\text{VO}(\text{L}^4)_2]\text{SO}_4$	Reddish brown	63	42.39(42.47)	2.54(2.45)	10.59(10.68)	20.21(20.28)	6.42(6.35)	56.6	1.76	
$[\text{VO}(\text{L}^5)_2]\text{SO}_4$	Brown	60	43.52(43.65)	2.60(2.51)	7.25(7.29)	20.74(20.88)	6.59(6.68)	56.5	1.73	
$[\text{VO}(\text{L}^6)_2]\text{SO}_4$	Green	61	39.03(39.00)	2.33(2.42)	6.50(6.42)	18.60(18.72)	5.91(5.83)	56.2	1.73	
$[\text{VO}(\text{L}^7)_2]\text{SO}_4$	Greenish yellow	65	49.25(49.13)	3.58(3.45)	7.65(7.53)	21.91(21.99)	6.96(6.84)	55.0	1.74	

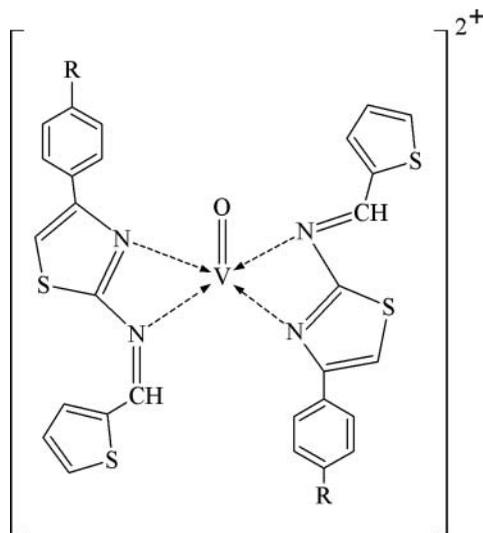


Figure 2 Structure of the complexes ($R = -H, -OH, -OCH_3, -NO_2, -Cl, -Br, -CH_3$).

16800–17600 cm^{-1} is attributed to ${}^2B_2 \rightarrow {}^2B_1$, and the band at about 21200–21640 cm^{-1} can be assigned to the ${}^2B_2 \rightarrow {}^2A_1$ transition. These observations give adequate support to a square pyramidal geometry for the complexes. In the visible region, the oxovanadium complexes showed another absorption band in the range 17650–17680 cm^{-1} , presumably due to ligand–metal charge transfer.³¹

The room temperature magnetic moment values of the complexes fall in the range 1.73–1.76 B.M. These values correspond to one unpaired spin per vanadium atom, demonstrating the presence of vanadium(IV) in these complexes, and the values are characteristic of monomeric oxovanadium complexes.³²

Thus on the basis of elemental analyses, conductance and magnetic moment values, and IR and UV-visible spectral data, a square pyramidal geometry has been proposed for all the complexes (Figure 2). The difficulty of obtaining crystalline complexes in the proper symmetric forms rendered the single crystal X-ray crystallographic study unsuitable for the complexes.

Thermal Decomposition Studies

The complexes $[\text{VO}(\text{L}^1)_2]\text{SO}_4$ and $[\text{VO}(\text{L}^2)_2]\text{SO}_4$ were subjected to thermal decomposition studies in dynamic air. In both cases, a single stage decomposition pattern was observed in the temperature range 310–510°C with DTG peaks at 420°C and 435°C, respectively. The final mass loss of 87.36% (86.99% theoretical) for $[\text{VO}(\text{L}^1)_2]\text{SO}_4$ corresponds to the formation of V_2O_5 . In the case of $[\text{VO}(\text{L}^2)_2]\text{SO}_4$, the mass loss of 88.93% (87.68% theoretical) also indicates the formation of the residue left as V_2O_5 . The above mass loss also agreed well with the mass loss obtained by independent pyrolysis experiment. Examination of the mass loss data revealed that $[\text{VO}(\text{L}^1)_2]\text{SO}_4$ decomposed at a faster rate compared to $[\text{VO}(\text{L}^2)_2]\text{SO}_4$. Apart from providing valuable insight into the thermal stability of the vanadium complexes, this study also supported the formulation of the complexes.

Antibacterial Activity

The ligand and the metal complexes have been screened for their antibacterial activity and the results obtained are available in Table S1 (see the Supplemental Materials, available online). The results indicated that all the metal complexes are more potent bactericides than the ligands and less potent than the standard streptomycin.

EXPERIMENTAL

All the chemicals used were of analytical grade; purchased from Aldrich, Merck, Sisco (India), etc.; and were used without further purification. The elemental analysis data were obtained by using a Heraeus Carlo Erba 1180-CHN analyzer. Infrared spectra were recorded on a Shimadzu FT IR 8000 spectrophotometer, and proton NMR spectra were recorded in DMSO- d_6 on a JEOL GSX 400NB 400 MHz FT-NMR spectrometer. Vanadium was determined by the standard method.³⁵ Far IR spectra were recorded on a Polytec FIR 30 Fourier spectrometer using CsI discs. The electronic spectra of the complexes were recorded on a Hitachi 320 UV-visible spectrophotometer using DMSO as solvent. Molar conductance measurements in DMSO were conducted using 10^{-3} M solutions of the complexes at room temperature with a Systronic Conductivity Meter Type-304. Magnetic moment values of the complexes were measured at room temperature using a Magway MSB Mk1 susceptibility balance, and X-ray diffraction data were recorded on a Siemens D 5005 model spectrometer. Thermal decomposition studies were carried out using a thermobalance of the type Mettler Toledo thermogravimetric system.

Synthesis of the Ligands

2-Amino-4-phenylthiazole and its derivatives were prepared according to a reported method.³⁶ The ligand N-(2-thiophenylmethylene)-2-amino-4-phenyl thiazole (L^1) was prepared by refluxing an equimolar mixture of 2-amino-4-phenyl thiazole (1.76 g, 0.01 mol) and thiophene-2-aldehyde (1 mL, 0.01 mol) in ethanol (30 mL) for 5 h. The volume of the reaction mixture was reduced to one-third of its initial volume and kept aside for 24 h. To the mixture, an excess of diethyl ether was added to precipitate the Schiff base (L^1). The yellow precipitate that formed was filtered, washed with ether, recrystallized from ethanol, and dried in vacuum over P_4O_{10} (yield 76%).

The Schiff bases of other thiazole derivatives (L^2 – L^7) were prepared by adopting the same method.

For the synthesis of ligand L^2 , thiophene-2-aldehyde (1 mL, 0.01 mol) was dissolved in methanol (10 mL) and was added in small portions to a solution of 2-amino-4-(*p*-hydroxy)phenylthiazole (1.92 g, 0.01 mol) in hot methanol (50 mL). The solution was then refluxed on a water bath for 4 h. The brown colored product that was precipitated by the addition of diethyl ether was filtered, washed with methanol, and dried in vacuum. The ligand was further purified by recrystallization from ethanol (yield 78%).

The ligand L^3 was prepared by refluxing an equimolar mixture of 2-amino-4-(*p*-methoxy)phenylthiazole (2.06 g, 0.01 mol) and thiophene-2-aldehyde (1 mL, 0.01 mol) in ethanol (30 mL) for 4 h. The yellowish brown colored product obtained upon addition of diethyl ether was filtered, washed several times with ether, recrystallized from ethanol, and dried in vacuum over P_4O_{10} (yield 76%).

For the preparation of ligand L⁴, 2-amino-4-(*p*-nitro)phenylthiazole (2.21 g, 0.01 mol) was condensed with thiophene-2-aldehyde (1 mL, 0.01 mol) in 1:1 molar ratio in ethanol medium. The resulting mixture was refluxed on a water bath for 5 h. The solution changed to yellowish brown, and it was then concentrated to ca. 20 mL and allowed to cool. Addition of diethyl ether results in the precipitation of the ligand, which was filtered and dried. It was further purified by recrystallization from ethanol (yield 75%).

The ligand L⁵ was prepared by refluxing equimolar mixtures of 2-amino-4-(*p*-chloro)phenylthiazole (2.1 g, 0.01 mol) and thiophene-2-aldehyde (1 mL, 0.01 mol) in ethanol (30 mL) for 4 h. The yellow colored product precipitated by adding diethyl ether was filtered, washed several times with diethyl ether, recrystallized from ethanol, and dried in vacuum over P₄O₁₀ (yield 78%).

The ligand L⁶ has been obtained by refluxing together equimolar solutions of 2-amino-4-(*p*-bromo)phenylthiazole (2.1 g, 0.01 mol) and thiophene-2-aldehyde (1 mL, 0.01 mol) in ethanol (30 mL) for 4 h. The volume of the reaction mixture was reduced to one-third of its initial volume, and excess of diethyl ether was added to precipitate the product. The product that was obtained was filtered, washed with ether, purified by recrystallization from ethanol, and dried in vacuum (yield 74%).

Condensation of thiophene-2-aldehyde (1 mL, 0.01 mmol) with an 2-amino-4-(*p*-methyl)phenylthiazole (2.1 g, 0.01 mol) in 1:1 molar ratio for about 5 h yielded ligand L⁷. The yellow colored product that was obtained upon addition of ether was filtered, washed with ether, purified by recrystallization from methanol, and dried in vacuum over P₄O₁₀ (yield 74%).

Synthesis of Complexes

The metal complexes were synthesized according to the following general procedure. (To avoid repetition, only a general method is given in the case of the complexes.) Vanadyl sulfate (0.005 mol) dissolved in ethanol (20 mL) was added to the solution of the appropriate ligand (0.01 mol) dissolved in ethanol (25 mL). After adjusting the pH of the solution to 6.5–7.0, the mixture was refluxed for about 4–5 h on a water bath. The mixture was then allowed to cool, and the complex that separated was filtered, washed with ethanol, and dried in vacuum over P₄O₁₀.

Antibacterial Experiments

The ligands and their metal complexes were screened for their antibacterial activity against *Escherichia coli*, *Staphylococcus aureus*, *Klebsiella pneumoniae*, *Salmonella typhi*, and *Bacillus subtilis*, at a concentration of 300 μg/mL in DMSO by the agar diffusion method described in the Supplemental Materials.^{33,37}

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