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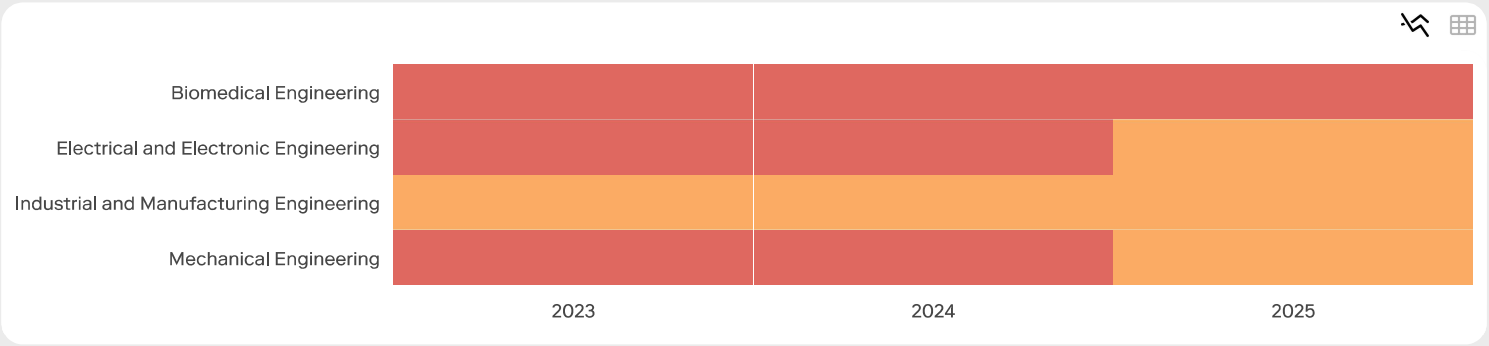

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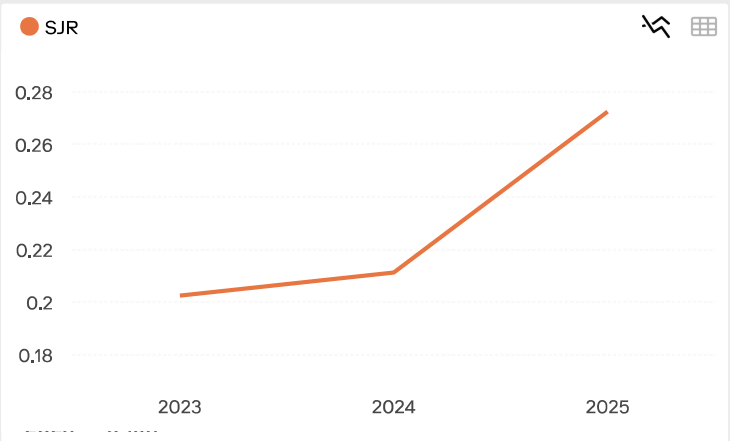

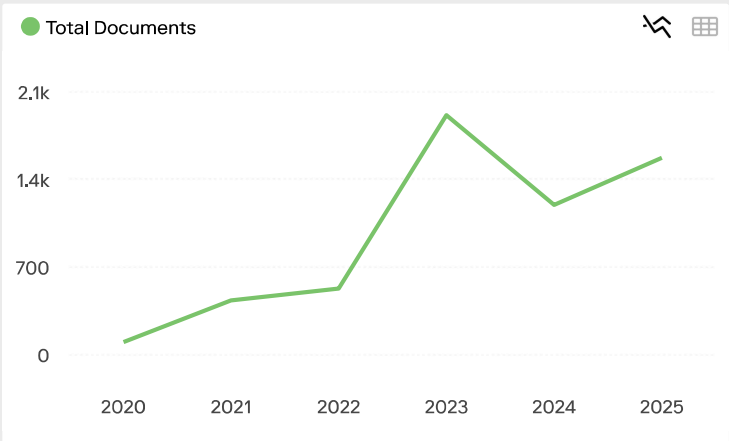

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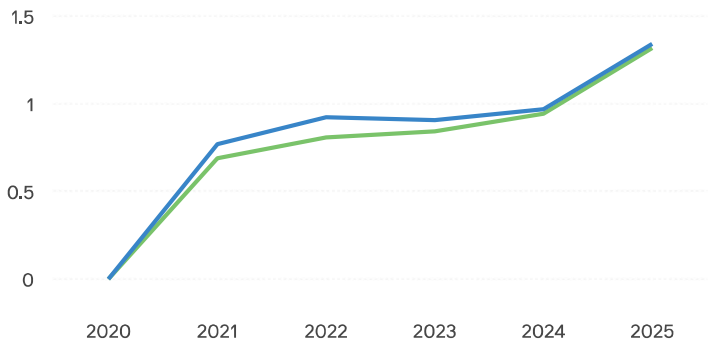

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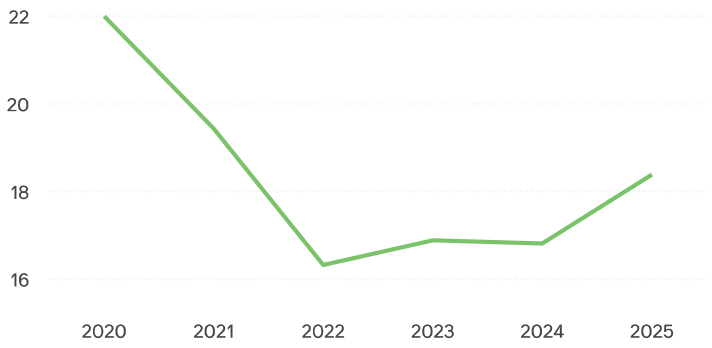

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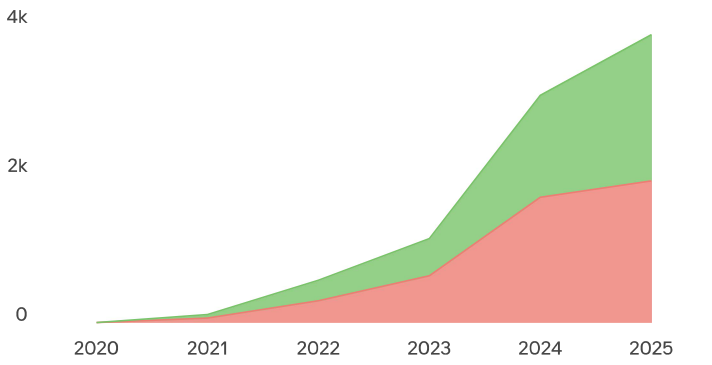

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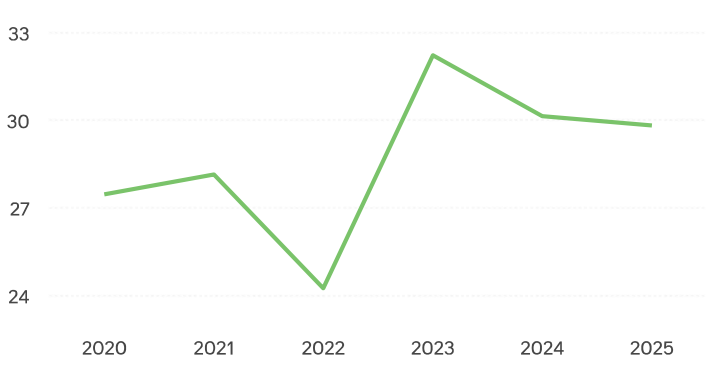

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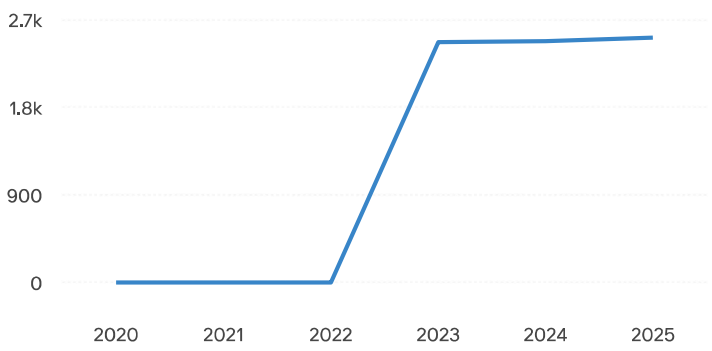

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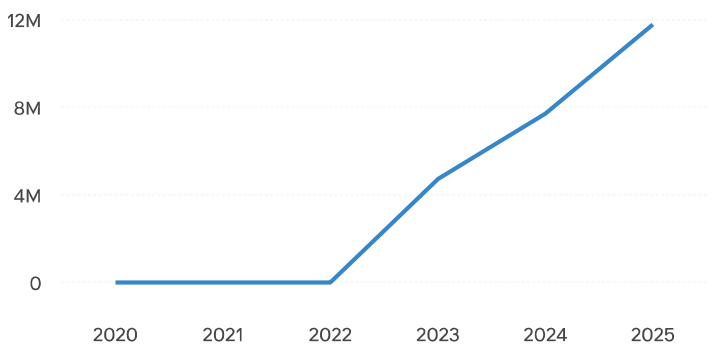

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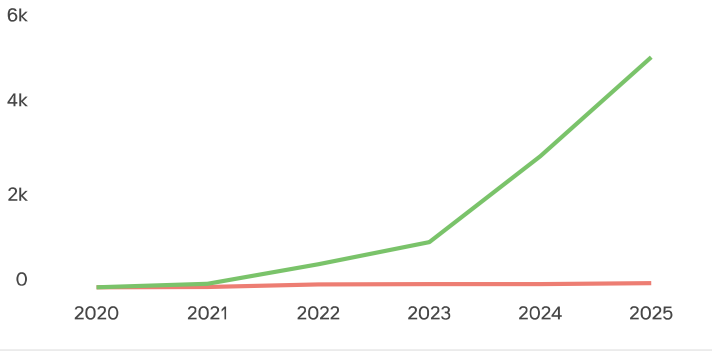

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The Potentiality of Vanadium Complexes as Antibacterial Agents †

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Abstract

Metal ions and ligand binding are crucial in various biological processes, and their rational design can be used to develop novel therapeutic drugs and diagnostic tools. Metal atoms are soluble in biological fluids due to their ability to easily lose electrons and form positively charged ions. Because of their electron deficiency, they can interact with electron-rich biomolecules like proteins and DNA, and potentially participate in catalytic mechanisms or stabilize their tertiary or quaternary structures. Antibacterial resistance has become a major global concern and requires novel strategies to combat resistance mechanisms in infectious microbes.

Keywords: Schiff base; vanadium complexes; biochemistry; antibacterial activity; mechanism of action

1. Introduction

Microorganisms plays an important role in the spoilage and decomposition of organic substances. To prevent the transmission of infections and diseases, regularization of microbial populations is needed [1]. Due to increasing microbial resistance, old antibiotics are becoming less effective, causing numerous deaths [2]. Metal ions play a fundamental role in biology, because they function as both pharmaceuticals and diagnostic agents [3]. Variable coordination modes, redox activity, and reactivity towards organic substances are the unique characteristics of metals that help in the regulation of their reactivity under normal conditions. Advancements in coordination chemistry have established metal complexes with optimistic pharmacological potential as either drugs or pro-drugs. Metal complexes have emerged as highly attractive options in pharmaceutical chemistry due to their potential to enhance the therapeutic efficacy [4]. Studies have established transition metal complexes as therapeutic agents for the treatment of human diseases [5]. The transition metals Fe, Mn, and Cu, involved in numerous biological functions including electron transfer and catalysis as well as structural roles, are found in the active sites of enzymes and proteins. Their complexes exhibit antidiabetic, anti-infective, and anti-inflammatory activities [6]. The synthesis of metal-based drugs with antimicrobial properties, a growing research field in inorganic, pharmaceutical, and medicinal chemistry, is thus essential for their use in pharmaceutical and drug delivery systems.

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2. Schiff Bases

Schiff bases are chemical compounds formed by the condensation of an aldehyde or ketone with primary amines. It is an analogue of aldehyde and ketone, having an azomethine or imine in place of a C=O group (Figure 1).

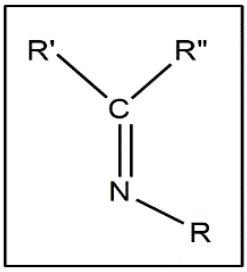

Figure 1. General structure of Schiff base.

They coordinate with metals to form stable metal complexes [7]. Schiff base ligands have been widely explored in coordination chemistry due to their ease of synthesis, accessibility, and favourable electronic characteristics. Schiff base coordination chemistry has garnered substantial attention because of its crucial applications in analytical chemistry, organic synthesis, metal refining, electroplating, metallurgy, and photography [8]. The advancement of bioinorganic chemistry and modern coordination chemistry is greatly influenced by Schiff bases. The formation of Schiff bases occurs more readily with aldehydes than with ketones because of differences in carbonyl reactivity. These ligands are readily prepared and demonstrate good chelating characteristics. Schiff bases can regulate the performance of metal ions in different catalytic reactions by stabilizing them in various oxidation states [9]. They play a vital role in medicine and drug discovery due to their diverse pharmacological properties, including antioxidant, anticancer, antitubercular, antibacterial, analgesic, inflammatory, and cardiovascular effects. Apart from these biological properties, Schiff bases are extensively used as catalysts, corrosion inhibitors, dyes, and polymer stabilizers [10]. Bi- or tridentate Schiff base ligands form stable complexes with transition metals, which are used in drug discovery. Transition metal complexes are used in treating microbial infections due to their higher biological activity as compared to ligands. Ommenya et al. synthesized complexes of Mn(II), Ni(II), Co(II), Cu(II), and Zn(II) using the Schiff base 4-chloro-2-{(E)-[(4-fluorophenyl)imino]methyl}phenol, which was prepared by the condensation of 4-fluoroaniline and 5-chlorosalicylaldehyde. These complexes were investigated for their antibacterial activity against *B. subtilis*, *S. aureus*, *P. aeruginosa*, and *E. coli* [11]. Similarly, Anacona et al. reported Mn(II), Co(II), Ni(II), Cu(II), and Zn(II) complexes of a ceftriaxone-based Schiff base ligand, and found that the Zn(II) complex exhibited significant antibacterial activity against *S. aureus* (MIC = 0.0048 µmol/mL) and *E. coli* (MIC = 0.0024 µmol/mL) compared to the free ligand [12]. Other than antibacterial activity, Saritha and Metilda synthesized Mn(II), Co(II), Ni(II), Cu(II), and Zn(II) complexes of 2-[5-hydroxy-1,7-bis (4-hydroxy-3-methoxyphenyl)-hepta-1,4,6-triene-3-ylidene] hydrazine carboxamide [HMHC] ligand and these complexes exhibited anti-inflammatory, antibacterial, and antifungal activities. The anti-inflammatory findings demonstrated that the Zn(II) complex showed higher inhibition (92.2%) than the ligand. The enhanced activity of the Zn(II) complex may be due to its ability to be easily reduced and oxidized. The antimicrobial assay demonstrated that all the complexes exhibited moderate activity against the tested strain. The Zn(II) complex exhibited superior antifungal and antibacterial activity due to its higher lipid solubility, making it a promising candidate for in vivo drug development [13]. In contrast, Anita et al. synthesized complexes of Mn(II) and Co(II) of (Z)-2-(2-methyl-1-phenylpropylidene)hydrazine-1-carbothioamide

ligand and reported antibacterial activity against *S. aureus* as well as antifungal activity towards *C. albicans* and *A. fumigatus*. The results showed that the complexes had increased activity compared to the ligand [14]. In addition, Gavali et al. also reported mixed ligand metal complexes (Ni(II), Co(II), Cu(II), Zn(II), and Cd(II)) of terephthalaldehyde bis(thiosemicarbazone) (TPTSC) as primary ligand and alanine as co-ligand. These complexes displayed antibacterial activity against *S. aureus*, *K. pneumoniae*, *S. typhi*, *E. Faecalis*, and *E. coli* and antifungal activity against *A. niger* and *A. flavus* [15]. Other than antibacterial activity, Deswal et al. reported Co(II), Ni(II), Cu(II), and Zn(II) complexes with Schiff base ligands and evaluated their antidiabetic potential using in vitro tests on α-amylase and α-glucosidase enzymes. The complexes exhibited strong activity, with IC_{50} values 1.28 ± 0.05 and 0.58 ± 0.06 μmol/mL [16]. In addition, Xiao et al. reported that Co(II) complexes derived from two Schiff base ligands (ON donors), formed by the condensation of p-nitrophenylamine with 3,5-dichlorosalicylaldehyde or 3,5-bromosalicylaldehyde, exhibited better antitumor activity against human cancer cell lines (SK-MEL-30, HT-144, and A-431) as compared to the free ligands [17]. Manimalathi et al. also reported Mn(II), Fe(II), Co(II), Ni(II), Cu(II), and Zn(II) complexes of a hydroxybenzylideneaminocyclohexyliminomethyl-4,6-dibromophenol Schiff base ligand, and these complexes were tested for their antimicrobial activity against *S. aureus* and *E. coli* and antifungal activity against *A. niger* and *C. albicans*. The anticancer potential of the Zn(II) complex was also evaluated on MCF-7 breast cancer cell line ($IC_{50} = 86.68 \pm 0.72$ μg/mL) [18]. Furthermore, Yadav et al. reported Co(II), Cu(II), and Zn(II) complexes with tetraaza macrocyclic Schiff base ligand. These complexes were tested for their anticancer activity against a human liver cancer cell line (Hep-G_2 cells) as well as antibacterial efficacy against tested bacterial strains [19]. Similarly, Raju et al. synthesized Ni (II), Cu(II), Co(II), and Zn(II) complexes of [4-(2-oxo-1,2-diphenylethylidene) amino] benzenesulphonamide and the complexes exhibited anticancer activity against a breast cancer cell line [20].

Much literature is available on the later elements of the first transition series, and complexes of vanadium have been, as of yet, not explored very much. Vanadium, an important transition element, with its unique properties, has drawn considerable interest from biologists, chemists, toxicologists, biochemists, and pharmacologists. In the III, IV, and V oxidation states, they readily bind with S, N, and O [21]. Previous studies have demonstrated that vanadium complexes possess a broad spectrum of possible diagnostic and medicinal uses due to their low toxicity. One of the most important impetuses for medical applications of vanadium complexes has been the ability to boost insulin activity in the pathophysiological condition of diabetes mellitus in humans [22]. In 1992, the discovery of the insulin-enhancing properties of oxovanadium(IV) [23] and oxovanadate(V) (2001) [24] complexes in vivo, along with the 'benchmark' complex bis(maltolato)oxovanadium(IV) (BMOV), drove the exploration of vanadium-based complexes as potential treatments for type II diabetes. Studies have reported vanadium complexes with a wide range of medicinal and diagnostic application with low toxicity and as an insulin-enhancing agent essential for humans. In addition to their antidiabetic properties, vanadium complexes have been found to exhibit various medicinal properties like antibacterial, anticancer, tissue regeneration, and anti-inflammatory effects, etc., [25–28]. This review focuses on the biological applications of vanadium complexes as antibacterial agents.

3. Vanadium as an Enzyme Switch

The ability of vanadium to activate enzymes is enhanced due to its complexation with the ligand. The anionic form of vanadium $(VO_4)^{-3}$ has been reported to stimulate glucose-6-phosphate dehydrogenase in mammalian cells [29]. Vanadate compounds are also known to energize the tyrosine kinases $p59^{fyn}$ and $p56^{lck}$ [30,31]. The activation of protein kinase B (PKB/AKT) and extracellular-signal-regulated kinases (ERKs), crucial for regulating

antilipolytic processes and the cell cycle, have been reported for vanadium complexes (Figure 2). Vanadium has shown the ability to inhibit the enzymes acid phosphatase (AP) and alkaline phosphatase (ALP), involved in the mineralization of bone [32,33], and glucose-6-phosphatase (G6P) [34,35]. The literature has established the regulatory role of vanadium complexes in a variety of biological processes [36].

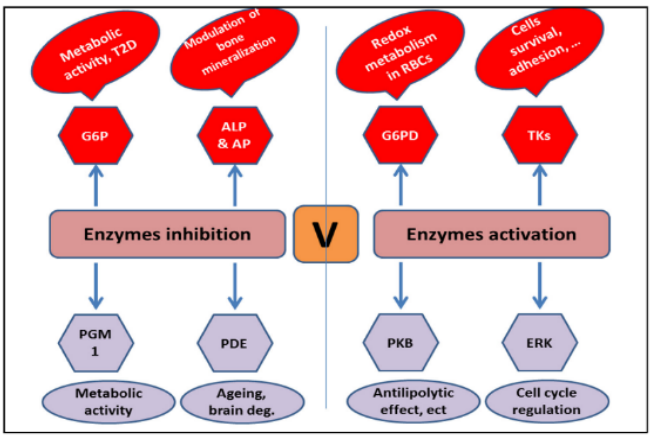

Figure 2. Various enzyme targets of vanadium complexes [26].

4. Antibacterial Activity of Vanadium Complexes

Pawar et al. reported the synthesis of oxovanadium (IV) complexes with macrocyclic Schiff base ligands. These complexes have been found to inhibit the bacterial strains *B. licheniformis*, *S. aureus*, *M. luteus*, and *E. coli* [37]. Rosu et al. synthesized a VO(II) complex using a 3-formyl-6-methyl-chromone and 4-amino-2,3-dimethyl-1-phenyl-3-pyrazolin-5-one functionalized Schiff base ligand. These complexes have been reported to exhibit antibacterial activity against *S. aureus*, *E. coli*, *K. pneumoniae*, and *P. aeruginosa* [38].

A series of oxovanadium (IV)-salicylidene-o-aminothiophenol complexes containing heteroligands have been reported to exhibit good antibacterial activity against *E. coli*, *S. typhi*, and *S. marcescens* using the disc diffusion method (Figure 3a–d). The results of this study show that the complexes exhibit higher activity (40–60% inhibition) compared to the corresponding uncoordinated ligands (10–25%) [39].

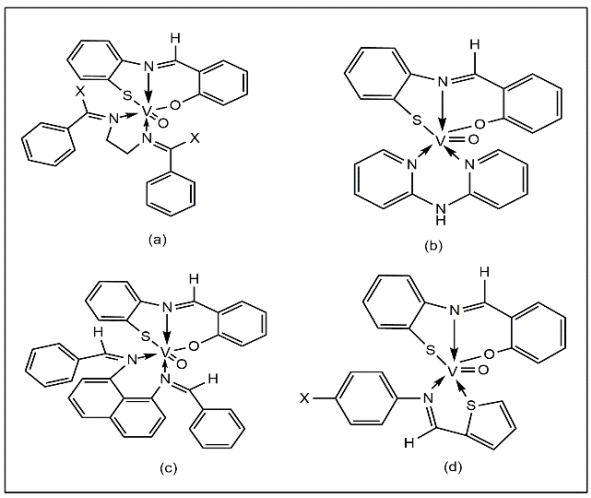

Figure 3. Oxovanadium heteroligand complexes containing common ligand salicylidene-*o*-aminothiophenol [VO(L)($A^{1–6}$)], (**a**) A^1 =bis(benzylidene)ethylenediamine, A^2 = bis(acetophenone)ethylenediamine, (**b**) A^3 = 2,2′-bipyridylamine, (**c**) A^4 =bis(benzylidene)-1,8-diaminonaphthalene, (**d**) A^5 = thiophene-*o*-carboxaldeneaniline and A^6 = thiophene-*o*-carboxaldene-p-anisidine.

Good antibacterial activities against *B. subtilis*, *S. aureus*, *S. flexneri*, *E. coli*, *S. typhi*, and *P. aeruginosa* have also been reported by Chohan et al. for oxovanadium (IV) complexes derived from a 1,2,4-triazole Schiff base. The presence of the NO_2 group in the ligand has been suggested to be responsible for its antibacterial activity (Figure 4a). This study reported a minimum inhibitory concentration (MIC) of 8.523×10^{-4} M, establishing the higher activity of the complexes compared to their uncoordinated ligand [40]. Chohan and Sumrra synthesized an oxovanadium (IV) complex using a thienyl-derived triazole Schiff base ligand and the complex was found to be more efficient than the ligands in inhibiting bacterial growth (Figure 4b), with the lowest MIC value of 3.891×10^{-8} M against *B. subtilis* [41].

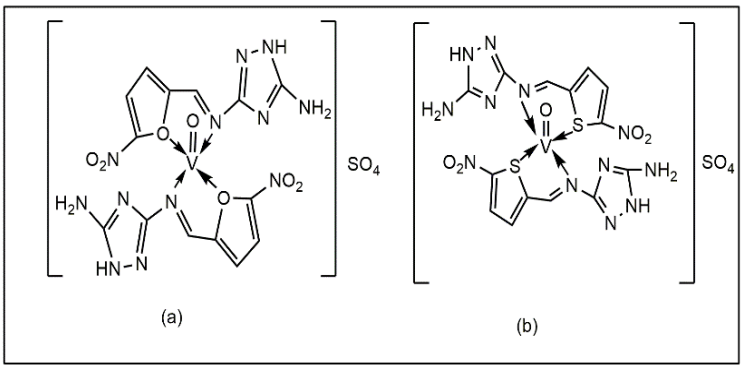

Figure 4. Vanadium complexes with (**a**) 1,2,4-triazole derived Schiff base, (**b**) thienyl derived triazole Schiff base.

Sheikhshoaie et al. reported that vanadium (V) complexes containing a dimalonitrile-based Schiff base (Figure 5a,b) exhibited antibacterial activity against *P. aeruginosa*, *E. coli*, and *K. pneumoniae*. The experimental data showed that the complexes exhibited superior efficacy compared to vanadium sulphate and ciprofloxacin against *L. monocytogenes*, *E. faecalis*, and *C. albicans*, with MIC values of 0.62, 1.25, and 2.5 mg/mL [42].

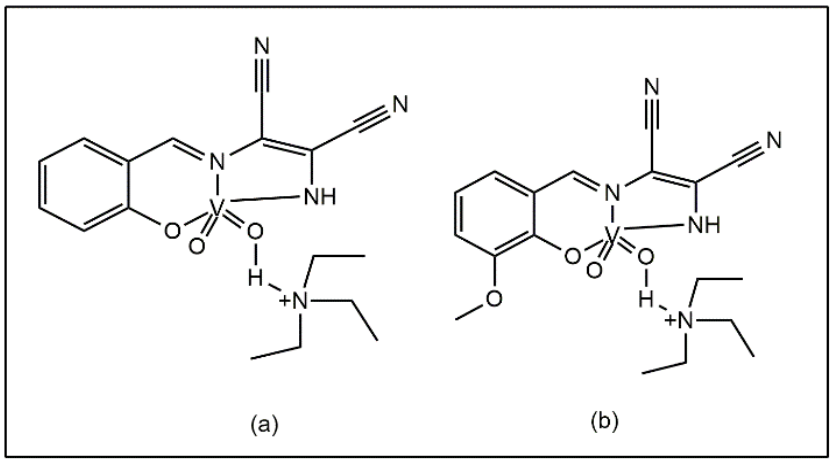

Figure 5. Vanadium complexes with dimalonitrile-based Schiff base: (**a**) triethylammonium 1,2-dicyano-2-(2-oxidobenzylidene)amino)vinyl)amide di-oxido vanadate, (**b**) triethylammonium 1,2-dicyano-2-(3-methoxy-2-oxidobenzylidene)amino) vinyl)amide di-oxido vanadate.

Machado et al. reported a vanadium stilbene complex (Figure 6) and the complex was tested against *T. cruzi* and *Leishmania* sp., which causes leishmaniasis, with an IC_{50} value of 3.51 μM against *L. amazonensis* [43].

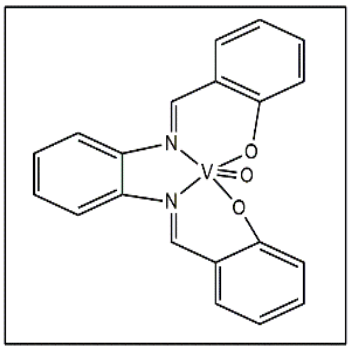

Figure 6. Vanadium stilbene complex.

Liu et al. also reported that mixed ligand vanadium complexes containing methanol and methoxy ligands, [VOL(OMe)(MeOH)] MeOH and salicylhydroxamic acid ([VOL(SHA)]·H_2O) (Figure 7a,b), showed antibacterial activity against *S. aureus*, *B. subtilis*, *K. pneumoniae*, *P. aeruginosa*, and *E. coli* [44].

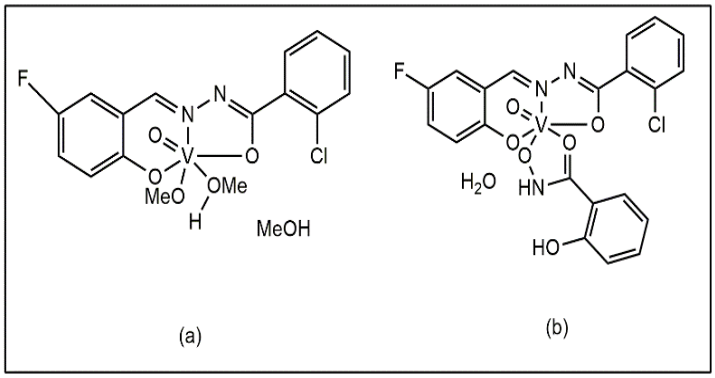

Figure 7. Vanadium Complexes of Fluoro- and Chloro-Substituted Benzohydrazone Ligands: (**a**) with methanol and methoxy ligands [VOL(OMe)(MeOH)] MeOH, (**b**) with salicylhydroxamic acid [VOL(SHA)]·H_2O.

Another series of transition metal complexes of VO(IV), CO(III), Ni(II), and Cu(II) of a 2-ethoxy-6-(E)-[(prop-2-em-1-yl)imino]methylphenol Schiff base ligand was reported and the synthesized complexes displayed considerable anticancer and antioxidant activities [45]. Additionally, Askarian et al. reported vanadium complexes of *N*,*N*′-dipyridoxyl(1,2-propanediamine) Schiff bases and the complexes exhibited significant cytotoxicity against a human prostate cancer (PC-3) cell line [46]. Lewis et al. also reported mixed ligand vanadium complexes with a Schiff base and thiosemicarbazone ligands and the complexes exhibited antitumor effects against non-malignant colon myofibroblasts (CCD18-Co) and various colon cancer cell lines, including Caco-2, HTC-116, and HT-29 [47]. On the other hand, vanadium-based coordination compounds are widely studied due to their essential biological functions and their ability to mimic insulin [48]. Studies have shown that vanadium complexes are more effective than vanadium salts in reducing blood glucose levels, suggesting their strong potential as insulin-mimicking agents [49]. Patel et al. synthesized vanadium complexes with a Schiff base and imidazole as a co-ligand, including [VO(L1)(H_2O)]NO_3, [VO(L2)(H_2O)]NO_3, and [VO(L2)($ImH)_2$]SO_4·H_2O (L1 = Acetic acid (2-hydroxy-3-methoxy-benzylidene)-hydrazide, and L2 = 5-bromo-2-[(E)-(pyridine-2-ylhydrazono)methyl]phenol) and reported antidiabetic activity [50].

Vanadium complexes, along with their ligands, exhibit improved antibacterial activity. The increased activity of the complex over the ligand can be attributed to the chelation theory. Chelation leads to enhanced bactericidal properties in contrast to the ligand itself.

The biological activity of metal-based drugs is regulated by the formation of metal ion complexes, which depend on the ligand type and the metal's oxidation state [51]. Various target sites within an organism are influenced by metal chelates, including the following:

1. Tweedy's chelation theory suggests that when metal ions bind to ligands, the polarity of the metal atom is reduced as its positive charge is partially shared with donor groups, and the electrons become delocalized across the entire chelate structure. This enhances the chelates' lipophilicity, improving their ability to penetrate the lipid membranes of bacterial cells [52].
2. Metal complexes disrupt cell wall formation, which alters the cell's permeability, causing the lipid–protein structure to become disorganized and ultimately leading to the death of the cell.
3. The regular cellular functions are disrupted by metal complexes, causing the denaturation of one or more enzymes [53].

Mechanisms of the Antibacterial Activity of Vanadium

The antibacterial activity of vanadium complexes has not been established yet. A review of the literature suggests potential mechanisms involving the following:

1. Disruption of Na^+/K^+-ATPase activity [54–57].
2. ROS formation [58].
3. The uptake of substrates through the bacterial cell membrane, along with the activation of K^+ efflux from the cell [59].
4. The interaction with topoisomerase type-II (gyrase), which is essential for genome function and bacterial growth, using the ATP-binding site as a target for antibacterial drugs [60,61].
5. Interaction with DNA through intercalation [62].
6. The disruption of cytoskeleton interactions, leading to changes in bacterial cell morphology and inhibiting proper cell division [56].
7. Other non-specific mechanisms, emphasizing the improved bioavailability of vanadium complexes, facilitated by a specific ligand that enables the complex to pass through the hydrophobic, lipid-rich bacterial wall [63].

5. Conclusions

Metal ions are important for cellular processes and biological functions in microorganisms. Vanadium is a well-known transition metal, and its complexes have been extensively studied for their medicinal properties. Schiff bases are considered valuable ligands because they are easy to synthesize and readily form complexes, with a wide range of metal ions exhibiting different denticity. The bioactive azomethine core is responsible for the medicinal importance attributed to various Schiff bases. The diverse therapeutic applications of Schiff bases and their metal complexes have initiated a new era in medicinal science. Metal complexes demonstrate superior antibacterial efficacy as compared to their Schiff base ligands. Research on antimicrobial metallodrugs is crucial for combating antibiotic resistance. Still, the mechanisms of toxicity remain uncertain, and limited in vivo data hinder further development due to the limited number of bacterial targets. Future multidisciplinary research on vanadium complexes as potential antibacterial agents offers opportunities to explore biochemistry and improve solubility, bioavailability, and toxicity and focus on developing novel strategies for targeting toxic metals and nanostructured antimicrobials for better understanding metal complex behaviour in living organisms [64,65]. Further exploration of vanadium complexes for their antibacterial potential will help to develop new lines of drugs for targeting these resistant bacteria and resolve the challenge of drug resistance to some extent.

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