

Review

Advanced and Biomedical Applications of Schiff-Base Ligands and Their Metal Complexes: A Review

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Abstract: Because of their importance in a variety of interdisciplinary study domains, Schiff-base ligands have performed a significant role in the evolution of contemporary coordination chemistry. This almost-comprehensive review covers all the aspects and properties of complexes, starting from the Schiff-base ligands. Our work is centered on the eloquent advances that have been developed since 2015, with special consideration to recent developments. Schiff-base ligands and their complexes are adaptable compounds obtained from the condensation of two compounds: a carbonyl with an amino. The correspondent metal complexes have been shown to have antifungal, antibacterial, antioxidant, antiproliferative, and antiviral properties. This review begins with a short introduction to Schiff-base ligands and their metal complexes. It stands out in the recent advancements in the Schiff-base coordination chemistry domain and its future prospects as a potential bioactive core. Additionally, the review contains knowledge about the antioxidant, redox, and catalytic activities of the Schiff-base complexes, with important future applications in the obtaining of new compounds and materials.

Keywords: Schiff-base complexes; antimicrobial activity; antioxidant activity; redox activity; catalytic activity; siloxane



Citation: Soroceanu, A.; Bargan, A. Advanced and Biomedical Applications of Schiff-Base Ligands and Their Metal Complexes: A Review. *Crystals* **2022**, *12*, 1436. <https://doi.org/10.3390/cryst12101436>

Academic Editors: Yu-Chen Liu and Yu-Ze Chen

Received: 19 September 2022

Accepted: 10 October 2022

Published: 12 October 2022

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1. Introduction

Schiff-bases are a peculiar type of ligands possessing a diversity of donor atoms that exhibit remarkable coordination modes towards transition metals [1–3], with the existence of an azomethine linkage influencing biological activities [4–6]. An appreciable number of Schiff-bases starting from various amines have been investigated by different methods [7,8] and have been shown to have interesting applications in catalytic reactions, materials chemistry, and last but not least, in industry [9,10]. Due to the reason that the steric and electronic properties can be controlled by the amine/aldehydes basic, the salen type ligands, obtained after condensation of salicylaldehyde and the primary diamine, are stated as flexible ligands for coordination chemistry. This important type of ligands contains in their structure donor centers necessary for metal ions to project different geometries with other ligands [11]. Therefore, a numerous number of complexes were obtained by moving the metal ions in the salen-type ligand. These compounds have been considerably investigated in various domains of chemistry. The chelating activity of the tetradentate ligand with nitrogen-oxygen donor atoms gives it kinetic and thermodynamic stability, making it interesting for researchers. The presence of the nitrogen in the imine groups (C=N) in Schiff-bases and in their metallic complexes and their chelating properties are the reason for their many unique biological properties.

Metallo-salens compounds constitute relevant parameters in the progress of current inorganic biochemistry [12–14], catalysis [15–17], magnetism [18], medical imaging [19], and not long ago in sensors [20,21], nonlinear optical (NLO) devices [22], solar cells [23], and as building motifs [24] or building blocks [25].

These compounds are easy to synthesize and can be bonded with quasi all metal ions in order to form the appropriate complexes; azomethine nitrogen being responsible for coordination with metal ions through it [26]. Quite a large number of these types of metal complexes with different coordination geometry and flexible oxidation states have been studied in depth [27–34], some of them being representative of the progress of inorganic biochemistry and catalysis. Copper is a biologically essential component, which is why many chemicals need it to function [35]. The coordination chemistry of copper has aroused the interest of numerous scientists due to its well-informed biological characteristics. A very large number of copper complexes based on Schiff-bases have been successfully used as models in biological and supramolecular systems [36–42].

Beyond the last decades, there have been many scientific studies regarding applications, mainly in biology: the antimicrobial, redox, catalytic, and antioxidant activities. Therefore, a review accentuating the employment of the named ligands and their complexes is demanded. The significance of Schiff-base complexes in supramolecular chemistry, materials science and catalysis, coordination and separation processes, applications in biomedical fields, and the formation of new compounds with outstanding structures and properties, had been well studied and revised [43–49].

The biological activity of the metal complexes is higher than that of their ligands. The complexes of the Schiff-bases are of great consideration due to their stability, electron donating capacity, optical nonlinearity, catalytic, photochromic, and biological activity. These practical activities are all based on the coordination of Schiff-bases with the metal ions. An interesting class of Schiff-base complexes is that obtained starting from amino acids [50]. Amino acids are dynamically implicated in a part of biological processes and they possess the $-NH_2$ and $-COOH$ coordinating sites, which can be bonded with aldehydes/ketones for synthesize Schiff-bases which are easily coordinated with the metal ions (Figure 1).

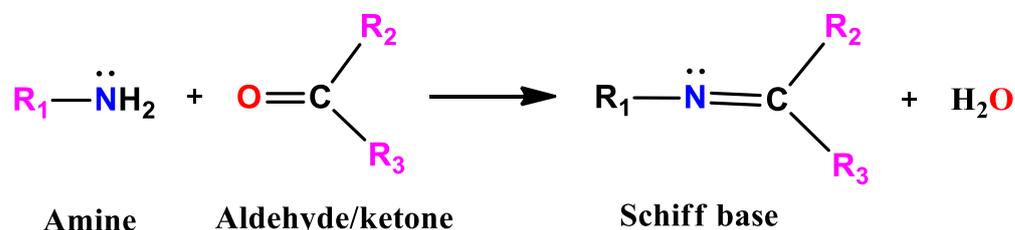


Figure 1. The obtaining reaction for the Schiff-bases.

The majority of the ligands derived from the amino acids and their complexes with the appropriate metal present distinct activities as drugs. An important study focused on the analysis of complexes based on Schiff-bases of amino acid derivatives from the last five years was made by Ghanghas et al. [51]. The complexes made with these ligands have high thermal stability and antibacterial activity, making them suitable for medical applications. The different types of the metal, ion, and ligand, the surrounding of the complex, coordination sites, hydrophilicity, lipophilicity, and the presence of co-ligands, together with the concentration, all affect the antibacterial activity of these compounds [52–54]. Inclusion of polar and lipophilic substituents increases the antibacterial action. Heterocyclic ligands with multifunctionality, which can interact with nucleoside bases or specific biological metal ions, are good candidates for bactericides [55–57]. The heterocyclic ligands interfere with functional groups (enzymatic type) to get access to high coordination numbers. In a recent study, Ghanghas et al. presented the history of the evolution of different types of investigations used in order to improve the metal complexes of the Schiff-bases' biological activity, thus being of real help in projecting a new class of drugs starting from the named compounds. The research regarding the antimicrobial activity of the synthesized compounds has been considerably studied because there is a relevant issue to exploring the linking properties of the complexes with a large variety of metal ions [51].

Researchers have concentrated their efforts in recent years on producing and investigating a new category of ligands and their complexes, having tetramethyldisiloxane spacers between the complexing groups (from 320 to 3249 structures in the Cambridge Crystallographic Data Centre (CCDC) database). These structures are organized as a single crystal (<https://www.ccdc.cam.ac.uk/structures/Search?Compound=tetramethyldisiloxane&DatabaseToSearch=Published>, accessed on 1 October 2022). The tetramethyldisiloxane spacers are well known as flexible and hydrophobic, and these properties are of real interest. Thus, research activities focused primarily on the production of such ligands and their metal complexes of relevance for catalysis, biological activity, materials science, and nanoscience, reporting a vast number of such structures in the CCDC crystallographic database [58–64]. The scientists have obtained and investigated around 259,536 Schiff-base ligands and their complexes, organized as a single crystal (the structures are presented in the CCDC Cambridge base) and more others in different forms.

In this review, advanced and biomedical applications (antimicrobial, antioxidant, redox, and catalytic activities) of novel Schiff-bases and the metal complexes starting from their discovery from 2015 to present are highlighted.

2. Some Aspects of the Biological Significance of Schiff-Base Complexes

The subject—the metal complexes starting from Schiff-bases has attracted the attention of researchers because of its biological activity, with the main goal of discovering straight and active therapeutic agents for the cure of various bacterial diseases.

Research into biological and inorganic chemistry has been of particular concern to Schiff-base metal complexes, as it has been observed that a lot of the complexes can be used as models for biologically important species. Therefore, we report them hereunder.

2.1. Antimicrobial Activity (Antibacterial and Antifungal)

Over the recent few years, from 2015 to present, the Schiff-base metal complexes have earned much attention due to the biological properties of them. A large number of studies have been published on their use in biological applications [65,66]. Schiff-bases have been found as potentially effective antibacterial agents. The metal complexes of the Schiff-bases have much better antibacterial activity than their free ligands [67–72].

The recently published literature [73] emphasizes the notable potential for antimicrobial activity and progress in the field of other types of interesting topoisomerase complexes. It was demonstrated that the Cu(II)-picolinic acid complex is a significant delayer of gel electrophoresis [74]. The thiosemicarbazone derivative of copper(II) has good activity in the killing of *S. aureus*, *S. typhimurium* and *K. pneumoniae* after 6 h of incubation [75].

The antibacterial activity of a special class of complexes of transition metals bonded through coordination bonds in the N_2O_2 mode was investigated, with the Schiff-bases of the salen-type starting from the, 1,3-bis(3-propyl)tetramethyldisiloxane (AP0)—a diamine having a siloxane spacer commercially available, with various salicylic aldehyde derivatives [76,77]. All the metal complexes studied were assessed for antifungal (in vitro with three types of fungi species (*Aspergillus niger*, *Penicillium frequentans* and *Alternaria alternata*) and antibacterial activity (with two types of bacteria—Gram-negative (*P. aeruginosa*) and Gram-positive bacteria (*Bacillus*)). The results of the antimicrobial activity tests showed a higher efficiency, closer to that of the reference compounds (in this case—Caspofungin and Kanamycin), in the case of the azomethines originating from substituted salicylaldehyde [77]. The ligands derived from 5-chlorosalicylaldehyde and its metal complexes have been shown to present the highest potential for biological applications (this could be caused by the presence of chlorine in the 5 position). The results of the antifungal and antibacterial activity measurements recommended some of the synthesized compounds as possible antimicrobial agents [59,76].

In another study, Zaltariov et al. [60] obtained and studied metal complexes starting from silicon-containing ligands (starting from a new amine-trimethylsilyl-propyl-*p*-aminobenzoate). The Schiff-bases behave as bidentate (NO), tridentate (N_2O), or tetradentate (N_2O_2) ligands,

and they have a large diversity of interesting characteristics with applications in various domains like biological, analytical, or industrial applications. Many of these ligands and complexes have antibacterial, antifungal, antiviral, and antitumor properties. For projecting different types of ligands, it is really important to choose the appropriate carbonylic and aminic precursors. Homometallic and heterometallic complexes with trimethylsilyl groups in structure have demonstrated an amphiphilic character and they can self-assemble in solution as a function of solvent polarity. These special properties enhance the catalytic activity for the complexes in various substrates and affect the behavior in solution. The authors synthesized Cu(II) and Zn(II) complexes with two bidentate Schiff-bases having trimethylsilyl units. The ligands were prepared by the condensation reaction of a novel trimethylsilyl-propyl-*p*-aminobenzoate with *o*-vanillin and salicylaldehyde. Some of the complexes can be used as possible candidates for cellular imaging because of their absorbance in the visible region and because of their green emission. Complexes with different types of metals were estimated against various fungi species and some types of bacteria, and the analysis showed that they have a higher biological activity than the standard compounds Kanamycin and Caspofungin [78].

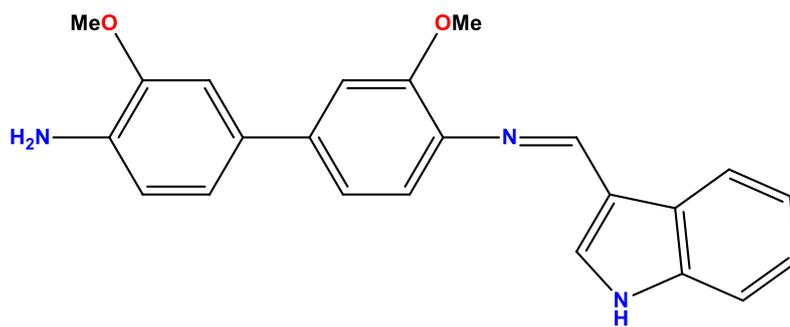
Literature studies indicate that Schiff-bases having antibacterial activity are obtained from indole [79,80], pyridine [81–83], isatin [84,85], hydrazide [86,87], benzimidazole [88,89], thiazolidiones [90,91], thiazole [92], thiosemi-carbazone [93,94], lysine/curcumin [95,96], and siloxane [76] (Figure 2).

Further study of the literature has shown an important increase in systemic fungal infections with life-threatening effects [97]. Numerous studies show that *Candida* (*albicans* and non-*albicans*) and *Aspergillus* (*Asp.*) species are responsible for the most severe fungal infections [98–102]. Thus, the progress of new antifungal species with decreased resistance and bigger efficacy is a priority [103,104]. A number of lengthy and laborious investigations have been carried out, and some Schiff ligands have been found to be bright antifungal agents [105,106]. The researchers also highlighted the existence of different groups such as methoxy, halogen, and naphthyl, which enhance the ligand's fungicidal activity [44,107]. While very disseminated, new literature clearly accentuates the remarkable potential of antifungal drug research in the metal complex domain [108–111].

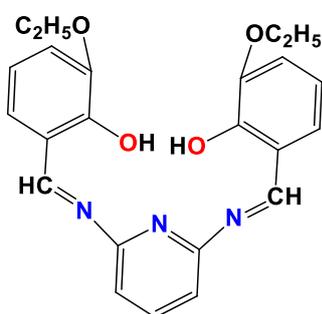
In another study, ligands, Schiff-bases type and their Cr(III), Fe(III), Mn(II), Cu(II), Zn(II), Ni(II), and Cd(II) metals, mononuclear chelate complexes have been obtained from 4-((1-5-acetyl-2,4-dihydroxyphenyl)ethylidene) amino)-1,5-dimethyl-2-phenyl-1H-pyrazol-3(2H)-one ligand, a tridentate ligand, were synthesized and used for in vitro determinations to establish their antimicrobial activity as compared with Gram-negative and Gram-positive bacterial and fungal organisms. In this study the MOE 2008 was used for heading the screen potential of drugs using molecular docking through the protein sites of the novel coronavirus, and the research was constructed to molecular docking free from the validation through MD simulations [112].

Some of the compounds were analyzed for their application for in vitro antimicrobial activities against four bacterial strains (*S. aureus*, *B. subtilis*, *P. aeruginosa*, *E. coli*) and also two fungal strains (*A. niger*, *C. albicans*) using the method of serial dilution, and the results have shown that the metal(II) complexes are not better than free Schiff-base ligands because they are more harmful (Figure 3).

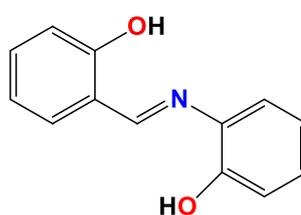
The complex $[\text{Cu}(\text{L}_2)(\text{CH}_3\text{COO})\cdot\text{H}_2\text{O}]$ was discovered to have antifungal activity against *Candida albicans* that was comparable to conventional medicaments. The molecular docking of the ligand H_2L_2 and its Cu(II) complex using the *C. albicans* sterol 14- α demethylase enzyme implies hydrophobic binding. The research “*in silico*” brings out that the named compounds can be employed as drugs active in orally derived forms [113].



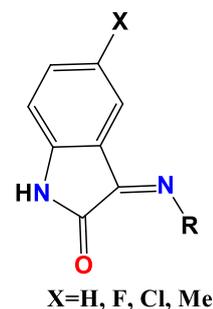
Indole (Vlad,2016)



Pyridine (Damoc,2020)

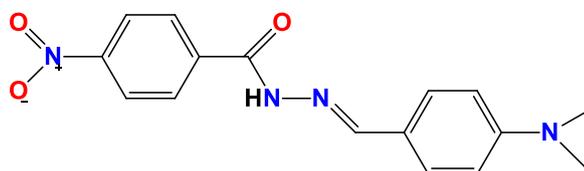


Pyridinehydrazone (Racles,2020)

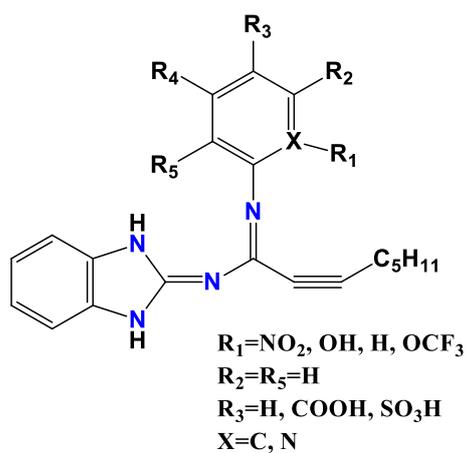


Isatins (Nath,2022)

X=H, F, Cl, Me

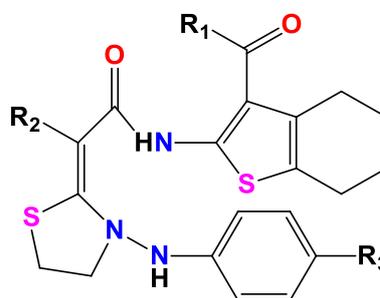


Hydrazone (Abu-Dief,2015)



Benzimidazole (Yousif,2017)

R₁=NO₂, OH, H, OCF₃
 R₂=R₅=H
 R₃=H, COOH, SO₃H
 X=C, N



Thiazole (Parveen,2020)

R₁=NH₂
 R₂=CN
 R₃=CH₃

Figure 2. Cont.

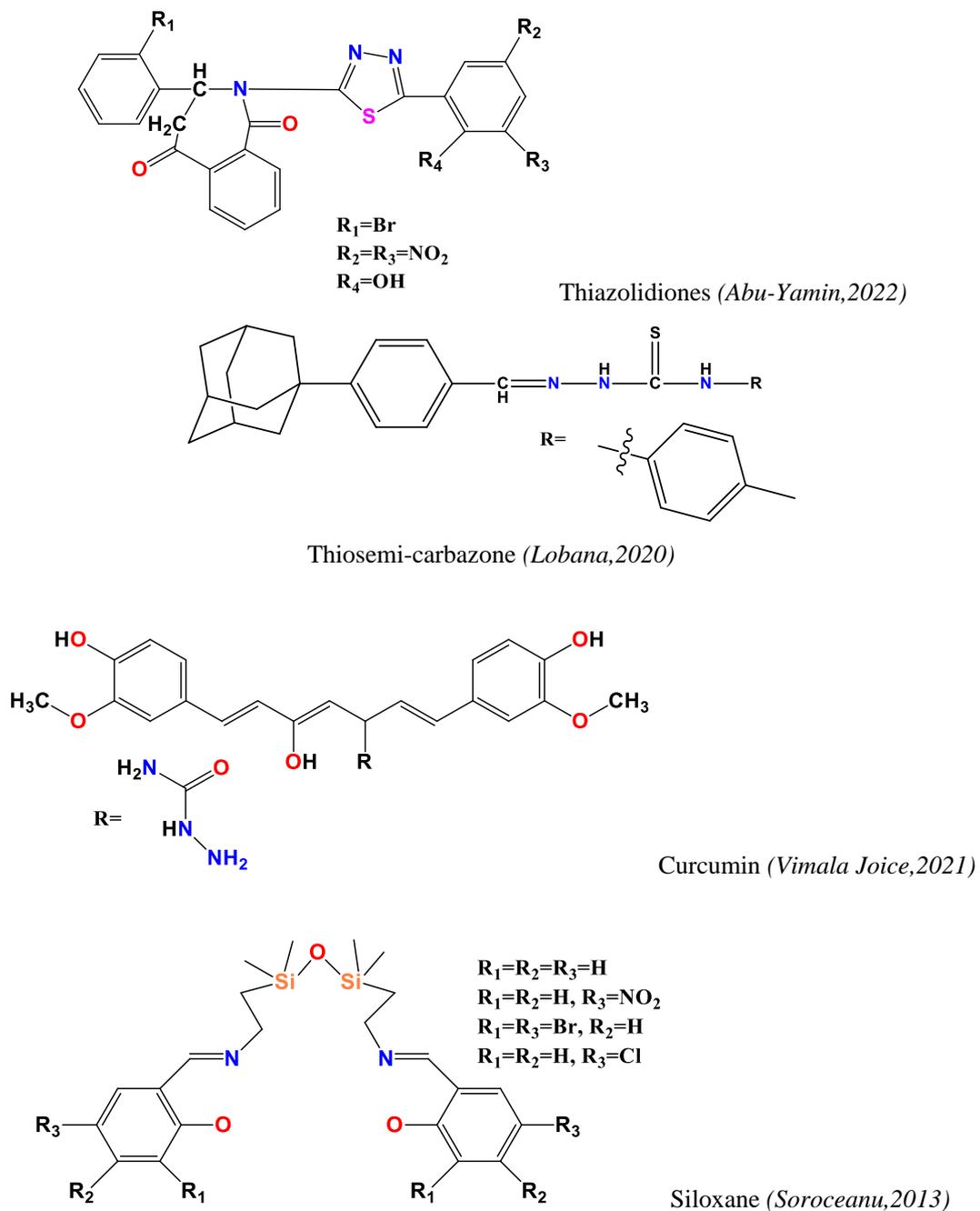


Figure 2. Representative Schiff-bases studied as antibacterial agents [58,61,63,64,66,68,70,72,74,75,95].

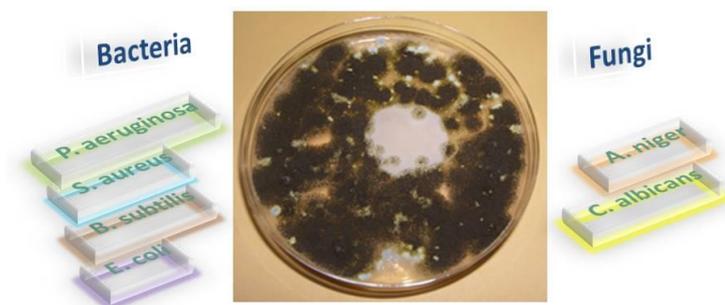


Figure 3. Biological activities of the studied compounds.

2.2. Antioxidant Activity

Much interest has been aroused in the identification of compounds with antioxidant capacity. Natural antioxidants are known to be the most expensive, which is why researchers have opted for the widespread use of synthetic antioxidants, this being more effective and cheaper. Thus, some metal complexes have been investigated in order to act as efficient traps of reactive oxygen species (ROS) behaving as antioxidants [43].

A series of Schiff-bases starting from diamine, sulfanilamide, hydroxyquinoline, thio-carbohydrazide, and benzohydrazide, with substituted ketone or aldehyde and their Co(II), Zn(II), Cu(II), Fe(II), Ni(II), Pd(II), Cd(II), and Ru(II), metal complexes were studied to determine their effect on the antioxidant activity. The compounds having methyl and nitro-substituents presented higher antioxidant activities than the ones with 4-hydroxy groups, conducting to an improvement in the antioxidant activity [114,115].

A study elaborated by Inan et al. [98] evidences the antioxidant potency by the L-ascorbic acid-standard method (DPPH). The complexes presented higher activity as compared with the ligand, this fact being due to the coordination binding of the metal ion with the organic ligand. [Cu(II)-(furfural-MAP)₂Cl₂] and [Ni(II)-(furfural-MAP)₂Cl₂] presented the higher antimicrobial activity, meanwhile [Zn(II)-(furfural-MAP)₂Cl₂] had a mild activity. The differences in the antioxidant activities of the complexes are due to their coordination sphere and redox characteristics [116].

Kizilkaya et al. [117] studies the antioxidant activities of synthesized Schiff-bases obtained using 2,2'-azino-bis(3-ethylbenzthiazoline-6-sulfonic acid) (ABTS) radical scavenging and 1,1-diphenyl-2-picryl-hydrazyl free radical (DPPH) scavenging and decreasing their activity. Due to the revealing of good antioxidant activity, the synthesized compounds (3, 5, and 7) have the potential to be used as synthetic antioxidant agents [117].

Devi and Pachwania [118] have obtained complexes containing new diorganotin(IV) and starting from ligands like R₂SnL (where R: methyl, butyl, phenyl, and ethyl) of Schiff-base ligands. The antioxidant activity of the complexes was evaluated using the measurement of DPPH (1,1-Diphenyl-2-picrylhydrazyl). The complex Ph₂SnL₃ possessed the highest antioxidant potential, having the smallest (2.95 μM) IC₅₀ value of all obtained compounds. The authors examined the biological profile of the compounds; then the complexes were screened for antimicrobial applications in vitro versus different fungal and bacterial strains using the method of serial dilution. Experiments showed that the Bu₂SnL₃ and Ph₂SnL₃ complexes were the most active antimicrobial agents [118].

In another research study, Devi and collaborators [113] obtained 16 new Ni(II), Cu(II), Co(II) and Zn(II) complexes starting from four Schiff-base ligands, synthesized through 4-(benzyloxy)-2-hydroxybenzaldehyde with various aminophenol derivatives condensation reaction. Some of the compounds were tested for their antioxidant properties in vitro applications and the obtained metal(II) complexes were found to have a significant potential and presented an important decolorizing the purple-colored solution of DPPH with a good efficiency as compared to free ligands and the Cu(II) complexes, which were the best, showed an IC₅₀ value in the range of 2.98 to 3.89 μM [113].

The antioxidant activities of macrocyclic Schiff-base ligands (N₄O₂), obtained by condensation of 2-hydroxybenzaldehyde or 2-hydroxy-3-methoxybenzaldehyde with polyamine and the copper(II) and cobalt(II) complexes, have been investigated [114,119]. The disk diffusion method and DPPH free radical scavenging were utilized to assess the characteristics (antibacterial and antioxidant) of the obtained compounds in vitro. The findings of this investigation clearly showed that all of the synthesized complexes have biological features that could aid in the prevention of disease progression and the development of innovative therapeutic medicines.

The new class of tetradentate Schiff-bases and the copper complexes of their having a tetramethyldisiloxane spacer between the coordination bounding groups were measured as additives in the obtaining of mass of *S. platensis* to determine their effect on the antioxidant activity of the 70 wt percent extract in ethanol based on spirulina biomass. The antioxidant activity determined from experiments in the presence of copper(II) complexes for *S. platensis*

biomass showed the potential to change in the direction of having increased activities in experiments [76]. The improved antioxidant activity of these compounds after coordination with different types of metals constitutes an important research direction.

2.3. Redox Activity of the Studied Schiff-Bases and Their Complexes with Different Metals

Cyclic voltammetry (CV) has become an electroanalytical method of great interest in many fields of chemistry. It is used to study redox processes, to understand reaction intermediates, and to obtain the stability of reaction products. The complexes presented complex redox behavior, including different oxidation/reduction processes not only of the central metal atom but also of the ligand. Electrochemical methods supply a remarkable manner for the research of the redox behavior for different types of metal complexes [120–122].

Tsantis et al. [123] identified the novel track to lanthanoid (Ln) multielectron redox transfer [124], presenting the redox activity of tetradentate N, N, O, O-Schiff-bases. The studies of reactivity and electrochemical ones made on K_3 mononuclear complexes [Ln(*bis*-Rsalophen)] demonstrated that the complexes oxidation potential can be adjusted through the changing of the substituent at the ligand and that the complexes can act as formal two-electron reducers.

Andrez et al. have shown that the redox activity of the ligands is influenced by the reversible storage of e^- and by the redox active ligands coordinated to cobalt ions and the important role played by the bounded alkali cations in these types of processes [125].

The determinations of cyclic voltammograms for the synthesized H_2L^x siloxane ligands and Cu(II) complexes showed that they have electroactivity. The irreversible electron and quasireversible transfer processes undergoes in both of the complexes with the central Cu atom and ligands. The CuL^3 , CuL^4 and CuL^1 , complexes are studied in the oxidation/reduction processes with a smallest potential value than CuL^2 complexes because of the substituents which are attached at the aromatic rings [76].

3. Schiff-Base Complexes as Catalysts

Among the many co-catalysts commonly used in many studies, the complexes starting from Schiff-base ligands with transition metals are a class of highly sought-after materials because of their ease of obtaining and the multitude of metal centers that can be embedded into the N_2O_2 coordination realm [50,126]. Their structure enables a wide range of substituents to be added. This chemical flexibility for the covalent stability of such catalysts on a support can be used. Exceptional studies have been published on this topic [127–129].

Various Schiff-base metal complexes have strong activity as catalysts and have been used in a variety of processes to improve the product selectivity and yield. The most relevant ways of synthesis and the thermogravimetric stability of the ligands have a significant influence on their applications in the catalysis field as metal complexes [130,131].

The Schiff-base complexes of the transition metal ion are effective catalysts in both homogeneous and heterogeneous processes, and their activity varies depending on metal ions used, ligands, and the coordination sites. The high catalytic activity of the metal complexes is due to the ability of Schiff-bases to settle down a wide range of metals at different oxidation states, allowing them to govern the ability and performance of metal ions in a wide range of important catalytic reactions [132].

Racles et al. effectively assessed the catalytic activity of Congo red (CR) in photodecomposition under natural light without the use of any extra oxidation agents or pH modifications. An important catalytic activity was determined for the Co-complex of CX and EBPpy, which presented a particular behavior: a discoloration efficiency of almost 82% after 80 min of sunlight exposure and 50% discoloration after 6 min [133].

The Schiff-base complexes of V, Mn, Fe, Co, Ni, Cu, and Zn ions from polymers, were utilized as catalysts for the peroxidation of several alkenes, including limonene, cyclohexene, styrene, trans-stilbene, verbenone, cis-stilbene, linear alkene, cyclooctene, α -methyl styrene, and α -pinene. The different Schiff-bases and various types of oxidants were explored. The data obtained for the complexes of polymer-supported first-row

transition metals for alkene peroxidation have been shown to be very interesting and significant to material and catalytic scientists because of their special properties as compared to unsupported catalysts and metals from the periodic table. Additionally, the authors presented challenges and opportunities for future study [134].

4. Conclusions

Because of their capacity to cast complexes with different types of metal ions and their pharmacological properties, the Schiff-bases are a relevant class of chemical molecules. The complexes have presented an increasing interest over the last few years because of their various applications in biological processes and possible uses in designing new interesting therapeutic agents. But still, it needs to explore the biological applications of the transition metal complexes, already synthesized, and to obtain new complexes with improved properties accordingly. Schiff-base complexes have been intensively studied in the antimicrobial domain, showing antibacterial activity against Gram-positive and Gram-negative bacteria, and also having antifungal potential. The most recent studies on Schiff-bases and their complexes as antibacterial have been reviewed herein. It is good to notice that antibacterial and antitumor activities are higher for the Schiff-base compounds complexed with metals. Therefore, the antimicrobial action of the Schiff-base compounds reviewed herein should be taken into account for increasing the development of novel metal complexes with enhanced antimicrobial capacities.

We conclude this study by emphasizing that it is expected that this short review presentation will be of real use for inorganic chemistry researchers who are working with Schiff-base ligands or who are just starting out in this interesting field. We would be honored if readers considered this short review useful and helpful for their future work, as we enjoyed editing it.

Author Contributions: Conceptualization, methodology, A.S. and A.B.; validation, A.S. and A.B.; resources, A.S. and A.B.; writing—original draft preparation, A.S. and A.B.; writing—review and editing, A.B. and A.B.; visualization, supervision, A.S. and A.B.; funding acquisition, A.S. and A.B. All authors have read and agreed to the published version of the manuscript.

Funding: This research was supported by a grant of the Ministry of Research, Innovation and Digitization, CNCS/CCCDI—UEFISCDI, project number PN-III-P1-1.1-PD-2021-0687 (DE-Comp), within PNCDI III (Contract 33/2022).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data are available upon request to the corresponding authors.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

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