REVIEW ARTICLE

Synthesis, Characterization of Schiff Bases and Biological Activities of Their Transition Metal Complexes-Review

D.T. Sakhare

U.G, P.G. & Research Centre, Department of Chemistry, Shivaji, Art's, Comm. & Science College, Kannad District, Aurangabad.431103, (M.S.) India.

ABSTRACT: Schiff bases are versatile ligands which are synthesized from the condensation of primary amines with carbonyl groups. These compounds are very important in medicinal and pharmaceutical fields because of their wide spectrum of biological activities. Most of them show biological activities such as antibacterial, antifungal as well as antitumor activity. Transition metal complexes derived from the Schiff base ligands with biological activity have been widely studied. This summarizes the synthesis and biological activities of Schiff bases and its transition complexes.

KEYWORDS: Schiff bases, metal complexes, antimicrobial activity, antitumor activity, nonlinear optical properties

https://doi.org/10.29294/IJASE.6.4.2020.1538-1544

1. INTRODUCTION

Schiff bases are the compounds carrying imine or azomethine (–C=N–) functional group. These are the condensation products of primary amines with carbonyl compounds and were first reported by Hugo Schiff [1–3]. Schiff bases form important classes of the most widely used organic compounds and have a wide variety of applications in many fields including analytical, biological, and inorganic chemistry. Schiff bases have gained importance in medicinal and pharmaceutical fields due to a broad spectrum of biological activities like anti-inflammatory [4–7], analgesic [5–8], antimicrobial [9, 10], anticonvulsant [11], antitubercular [12], anticancer [13, 14], antioxidant [15], anthelmintic [16], and so forth. The nitrogen atom of azomethine may be involved in the formation of a hydrogen bond with the active centers of cell constituents and interferes in normal cell processes [17, 18].

Apart from biological activities, Schiff bases are also used as catalysts, intermediates in organic synthesis, dyes, pigments, polymer stabilizers [3], and corrosion inhibitors [19]. Studies enlightened that metal complexes show greater biological activity than free organic compounds [20]. Augmentation of biological activity was reported by implementation of transition metals into Schiff bases [21]. Schiff bases played an influencing role in development of coordination chemistry and were involved as key point in the development of inorganic biochemistry and optical materials [22].

This concentrates on the synthesis and biological activity of Schiff bases and their complexes.

2. SCHIFF BASE

Hugo Schiff, one of the founders of modern chemistry, discovered the Schiff base [2]. The Schiff base is named after Hugo Schiff and is a compound with a functional group that contains a C-N double bond with the nitrogen connected to an aryl or alkyl group. Schiff bases in a broad sense have the general formula R1R2C=NR3, where R is an organic side chain. Schiff base is synonymous with azomethine and may also be referred to as imines [1].

3. SYNTHESIS OF SCHIFF BASE

A common method of synthesis of Schiff base is the nucleophilic attack of amines on electrophilic carbon of aldehydes or ketones. The end result of this reaction is a compound in which C=O double bond is replaced by a C=N double bond (Scheme 1).This type of compound is called an imine or a Schiff base.

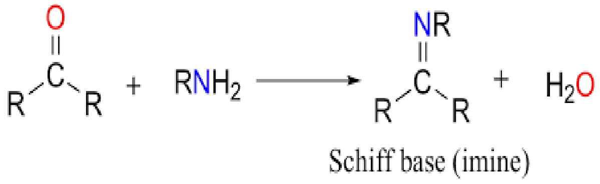

Fig.1: Synthesis of Schiff base

3.1. Synthesis of Novel Schiff's Base Ligands

Over the past 25 years, extensive chemistry has surrounded the use of Schiff base ligands in inorganic chemistry. For instance, N,N'–ethylene bis (salicylidenimine) (Salen) [23], which is nearly related

**Corresponding Author: sakharedhondiram@yahoo.com*

Received: 15.04.2020 **Accepted:** 21.05.2020 **Published on**: 29.05.2020

and similar novel ligand N,N'-bis(benzoylacetone) ethylenediamine, and some related derivatives have been the ligands of choice for several reasons. Firstly, the case of synthesis of these compounds has meant that bulk amounts can be synthesis both simply and cheaply; properties that become very important when industrial applications are being sought. Secondly, these ligands are multidentate, which means for most metals, several binding sites are occupied, leaving vacant sites for potential catalytic/enzymatic activity [23]. Furthermore, substitution at the aromatic ring can modify the electronic and steric properties of the resulting complexes, which can enable fine-tuning of properties with the ubiquitous nature of the ligands in transition metal chemistry, it comes as a bit of a surprise that main group metals and lanthanoids have only been studied to a limited extent. It is the aim of this work to study some Group 1 and 2 chemistry of these salen ligands and to prepare some "cage complexes" using some lanthanoid metals. Our plan will encompass several fronts [24].

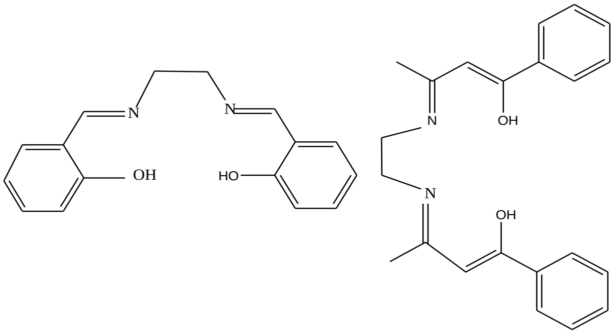

Fig.2.StructureofN,N'–ethylenebis (salicylidenimine) N,N'bis(benzoylacetone)Ethylenediamine respectively

Different R groups on the aromatic moieties will be employed to modify the steric and electronic requirements of the ligand; Different bridging groups (eg.: ethylene and propylene, but also other alcohol containing bridges) between the two nitrogen centers will be incorporated to study the effects of chelate size, rigidity and further coordination sites; Different metals (s block metals) will be used to elucidate the effect of metal size and coordination number requirements, Different coordinating solvents (Diethylether, THF, DME, TMEDA, PMDETA) will be used to modify the resulting nuclearity of the complexes, To expand some chemistry of lanthanoid "cage complexes" using, in particular, alcohol substituents on the bridging groups of the Schiff base ligands, for which we have recently isolated some exciting and novel complexes [25, 26]

3.2. Metal Complexes with Schiff's Base Ligands

Schiff bases offer a versatile and flexible series of ligands able to bind with various metal ions to give complexes with suitable properties for theoretical and/or practical applications. Since the publication of Schiff base complexes, a large number of poly-dentate Schiff's base compounds have been structurally characterized and extensively investigated [27]. Schiff's base ligands and their metal complexes have been extensively studied over past few decades.

Metal complexes containing synthetic macrocyclic ligands have attracted a great deal of attention because they can be used as models for more intricate biological macrocyclic systems like metalloporphyrins (hemoglobin, myoglobin, cytochromes, chlorophylls), corrins (vitamin B12) and antibiotics (valinomycin, nonactin). So it attracted the attention of both inorganic and bioinorganic chemists [28]. These discoveries have created supramolecular chemistry and its enormous diversity. Metal complexes of the Schiff bases are generally prepared by treating metal salts with Schiff's base ligands under suitable experimental conditions. However, for some catalytic application the Schiff base metal complexes are prepared in situ in the reaction system. For instance five synthetic routes that are commonly employed for the preparation of Schiff's base metal complexes and these are depicted in Figure 3 [29].

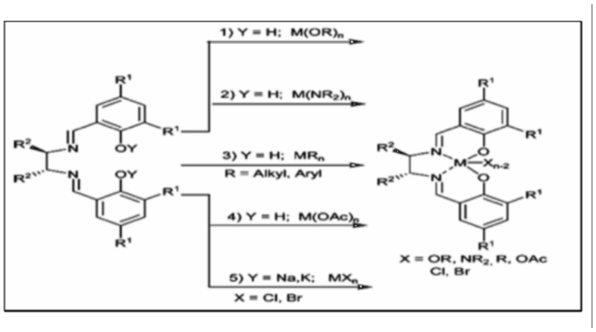

Fig.3. Preparation of Schiff base complexes [29]

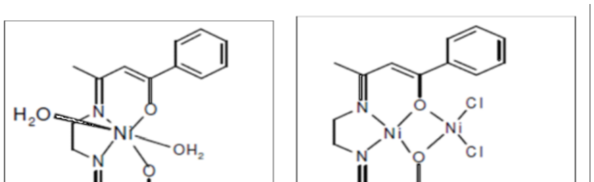

Fig.4. Two types of nikel complexes with the noble ligand (L), N,N'-bis (benzoylacetone) ethylenediamine [29]

4. CHARACTERIZATION METHODS

Among various physico-chemical techniques used in complexometric investigations elemental analysis, IR and electronic spectral methods (UV-Vis spectroscopy), Atomic absorption spectroscopy, Molar

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conductometric and susceptibility measurements being highly sensitive and informative are most popular.

4.1. Elemental Analysis

Novel Schiff's base ligands and their complexes are analyzed micro analytically for carbon (C), hydrogen (H), Sulfur(S) and nitrogen (N) contents to find out their empirical composition. That means Elemental analysis of a compound enables one to determine the empirical formula of the compound. The empirical formula is the formula for a compound that contains the smallest set of integer ratios for the elements in the compound that gives the correct elemental composition by mass. So physical characteristics and elemental analysis of C, H and N of the compounds are considered to determine the composition of complexes and the novel ligand. The results of C, H and N percentage are in accordance with the composition suggested for the most complexes [29, 30].

4.2. Infrared Spectra

IR spectra of the samples are usually recorded in Nujol or KBr medium in 200-4000 cm^{-1} range to identify coordination sites of ligands, formation and strength of metal-ligand bonds in the complexes, and to study the bonding vibrational modes of ligand in metal complexes. The IR spectra of the complexes and ligands have a diagnostic importance to determine the structure of the compounds. The solid state IR spectra of complexes compared with those of ligands indicated that the (C=N) stretching vibration band at region about 1500-17000 cm^{-1} is shifted to lower frequencies in most complexes as excepted. In contrast there are many complexes shifted to higher frequencies, which indicated that the ligands coordinated to the metal ions through nitrogen atom of the azomethine group. In general the observed IR bands of Schiff's bases and their complexes are in conformity with the previously reported results [31, 23, 32].

The presence of sharp band corresponding to the remaining hydroxyl group at 3400cm-1 but it is obscured by the presence of water molecules bands. This was appeared for the most complexes and a very broad band at about 3100-3500 cm^{-1} region, which is associated with coordinated or solvent water molecules. The other bands appeared at 1323-1427 cm^{-1} region assigned to the (C—O), which are shifted to a higher frequency after complexation with central metal ions, compared to the free ligands in which was noted at 1261-1315 cm^{-1}. In addition the two bands at 729-511 and 531-442 cm^{-1}, is attributed to the (M—O) and (M—N) respectively [26, 30].

Moreover new bands appeared in some complexes in the 220-290 cm^{-1} regions which is assigned to (M—Cl) vibration, which indicated the formation of (M —Cl) coordinated bond. The IR spectra of [Cu2L1Cl2] 8H2O complex exhibit a strong band at 1568 cm^{-1} which is assigned to the (C=N) stretching, because this band is shifted to lower frequency by 36cm 1compared to free ligand, indicating that the ligand coordinated to the metal ion through nitrogen atom of the azomethine group and probably dianionic form. The broad band around 3425 cm^{-1} indicating the presence of coordinated or lattice water in the complex. The spectrum reversals a weak band at 1399 cm^{-1} which is attributed to (C—O) vibration, again this band is shifted to higher value compared to the free ligand due to formation (C—O—M) bond. In addition three new bands in the regions 527,466 and 221 cm^{-1} were emerge, which are probably due to the formation of (Cu— O), (Cu—N) and (Cu—Cl) bond respectively [25]. The IR spectra results usually provide valuable information regarding the nature of functional group attached to the metal atom. In order to study the binding modes of the ligand to metal in complexes, the IR spectrum of the free ligand should be compared with the corresponding metal complexes. The electronic absorption spectra are often very helpful in the evaluation of results furnished by other methods of structural investigation. The electronic spectral measurements have been used by chemists for assisting the stereochemistry of meal ions in the complexes based on the position and number of transition peaks. Ultra violet spectrum recording frequencies of different d-d electronic transitions are called as d-d transition spectrum [23].

4.3. Molar Conductance

The molar conductance for the complexes measured in 10-3 M solution in DMF and chloroform as solvents at room temperature (29 - 31 ºC). The molar conductivity is applied to help in the investigation of the geometrical structures of the complexes. Some complexes showed a lower molar conductivity values in the range 0.03-44.37 Scm^2mol^{-1} which indicated their non-electrolytic nature. Other complexes found to be a higher electrolyte with the values more than $50cm^2mol^{-1}$, these results is demonstrated that the complexes have a binuclear nature [24].

The molar conductance has been demonstrated to be a very useful tool in the investigation of electrolytic nature and geometrical structure of inorganic compounds.

4.4. Magnetic Susceptibility Measurements

Magnetic susceptibility of the complexes will be measured and magnetic moments can be calculated. From the magnetic moment values in conjugation with molecular formulae, geometry (hybridization) of complexes will be determined. The gram magnetic susceptibility values, the magnetic moment in BM are calculated and presented as data. From the obtained result, M-L is concluded to be a high spin complex or M-L as a low spin complex [23]. Magnetic susceptibility measurements are used to determine the extent of electron pairing, the stereochemistry and metal-metal interactions in the complexes.

4.5. Electronic Spectra

Electronic spectra (UV-Vis region-spectroscopy) of the complexes are recorded in 250-1000cm-1 range to identify ligand field (d-d transitions) and charge transfer and to calculate ligand field parameters, viz. 10Dq(ligand field splitting energy), B and C (Racah inter electronic repulsion parameters), and β(Nephelauxetic ratio) [33]. The electronic absorption spectra and magnetic moment values are often very helpful in the evaluation of results provided by other methods of structural investigation. Information about geometry of the complexes around the Cu(II), Co(II) and Ni(II) ions was obtained from electronic spectra and from values of the magnetic moments. The electronic absorption spectra of the Schiff base ligands and its complexes were recorded at room temperature using (DMF) and (CHCl3) as solvents [34].

5. BIOLOGICAL ACTIVITIES OF SCHIFF BASE AND THEIR METAL TRANSTSION COMPLEXES

Schiff bases are generally bi- or tridentate ligands capable of forming very stable complexes with transition metals. In organic synthesis, Schiff base reactions are useful in making carbon-nitrogen bonds.

Hakan Arslan *et al*. synthesized five thiourea derivatives ligand and their Ni^{2+} and Cu^{2+} complexes. Those compounds were screened for their *in vitro* antibacterial activity using Gram-positive bacteria (two different standard strains of *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Enterococcus faecalis*, *Streptococcus pyogenes*, *Bacillus cereus*) and Gram negative bacteria (*Esherichia coli*, *Pseudomonas aeruginosa*, *Enterobacter cloacae*, *Proteus vulgaris*, *Enterobacter aerogenes*) [35].

Yu-Ye Yu *et al.* synthesized Mn(II), Co(II), Ni(II), Cu(II) and Cd(II) complexes with Schiff base ligand 2-[(4- methylphenylimino) methyl]-6-methoxyphenol, obtained by condensation of *o*-vanillin (2-hydroxy-3-methoxybenzaldehyde) with *p*-toluidine. The Schiff base ligand and its complexes have been tested *in vitro* to evaluate their antibacterial activity against bacteria, viz., *Escherichia coli*, *Staphylococcus aureus* and *Bacillus subtilis* [36].

Sohail Saeed *et al.*[37] prepared nickel(II) and copper(II) complexes of *N*-(alkyl (aryl) carbamothioyl)-4-nitrobenzamide and those complexes were screened for antibacterial activity against *S. aureus*, *S. epidermidis*, *E. faecalis*, *E. coli*, *E. cloacae* and *P. valgaris* by the broth microdilution procedure.

Obaleye. *et al.* [38] Co(II), Ni(II) and Cu(II) complexes were synthesized with 2,5-diamino-1,3,4-thiadiazole ligand. The ligand and metal complexes were tested agianst bacteriaViz *Salmonella typhi, Shigella species, Echerichia coli, Klebsiella species, Staphylococcus aureus, Pseudomonas aeruginosa, Niesseria gonorrhoe.*

Elzahany *et al* [39] have synthesized some transition metal complexes with Schiff bases derived from 2-formylindole, salicylaldehyde and N-amino Rhodanine. The Schiff base ligands were characterized by elemental analysis, IR, Mass, ^1H NMR and electronic spectra. The free ligands and their metal complexes were also screened for antimicrobial activities against *Bacillus cerens, Escherichia coli, Pseudomonas aeruginosa, Staphylococcus aureus and Candida albicans*. The results indicated that the ligands do not have any activity, where as their complexes showed more activity against the same organisms under identical experimental conditions.

Complexes of Co(II),Cu(II),Ni(II),Mn(II) and Cr(III) with Schiff bases derived from 2,6-diacetylpyridine and 2-pyridine carboxaldehyde with 4-amino-2,3-dimethyl-1-phenyl- 3-pyrozolin-5-one show antibacterial and antifungal activities against *Escherichia coli, Staphylococcus aureus, Klebsiella pneumoniae, Mycobacterium Smegmatis, Pseudomonas aeruginosa, Enterococcus cloacae, Bacillus megaterium* and *Micrococcus leteus*. The results showed that L_1 ligand has a greater effect against *E. coli* than the other bacteria while it has no activity against *S. aureus*. Metal complexes have a greater effect than L_2 against almost all bacteria [40].

Natarajan and coworkers have reported the synthesis of neutral tetradentate chelate complexes of Cu(II), Ni(II), Co(II), Mn(II) and Zn(II) in EtOH using Schiff bases derived from acetoacetanilido-4-aminoantipyrine and 2-aminophenol/2-aminothiophenol. Mi- croanalytical data, magnetic susceptibility, IR., UV–vis., ^1H-NMR. and ESR spectral techniques were used to con-firm the structures of the chelates. The *in vitro* antim- icrobial activity of the investigated compounds was tested against the microorganisms such as *Salmonella typhi*, *Staphylococcus aureus*, *Klebsiella pneumoniae*, *Bacillus subtilis*, *Shigella flexneri*, *Pseudomonas aerugi- nosa*, *Aspergillus niger and Rhizoctonia bataicola*. Most of the metal chelates have higher antimicrobial activity than the free ligands [41].

Singh *et al* developed a new Schiff base 2-aminophenol-pyrrole - 2- carboxaldehyde and its Zn(II), Cd(II), Sn(II) and Pb(II) complexes. The bio-efficacy of the ligand and their complexes has been examined against the growth of bacteria *in vitro* to evaluate their antimicrobial potential. The ligand and complexes exhibited high activity against E. coli (100 ppm) and others show very effective against S. aureus (100 ppm) bacteria. The zinc complex showed better activity than other metal complexes for both microorganisms. [42]

Ibrahim *et al* derived metal complexes of novel Schiff base (HL) ligand, prepared via condensation of 4-aminoantipyrine and 2-aminophenol. The synthesized ligand, in comparison to its metal complexes is screened for its antibacterial activity against bacterial species, *Escherichia coli, Pseudomonas putida, Exiguobacterium acetylicum* and *Bacillus simplex*. The activity data show that the metal complexes to be more potent/antibacterial than the parent Shciff base ligand against one or more bacterial species. [43]

6. CONCLUSION

The chemistry of Schiff bases is a field that is being noticed. Schiff base ligands are considered privileged ligands because they are easily prepared by a simple condensation of an aldehyde derivatives and primary amines. These compounds and their metal complexes had a variety of applications including clinical, analytical, agrochemical industrial they also play important roles in catalysts and corrosion inhibitor. In this review, synthesis of the Schiff and its complexes and the biological activities has been summarized.

ACKNOWLEDGEMENT

Dr. D.T. Sakhare acknowledges the immense help received from the scholars whose articles are cited and included in references of this manuscript. The author is also grateful to authors / editors /publishers of all those articles, journals and books from where the literature for this article has been reviewed and discussed.

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