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A Review on Synthesis and Versatile Applications of Some Selected Schiff Bases with Their Transition Metal Complexes

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Schiff base ligands and their transition metal complexes have covered a large field in chemistry that is being noticed. Schiff base ligands are viewed as special ligands since they are easily prepared by condensation reaction between aldehyde derivatives and amines. However they have simple preparation method; Their preparation technique is modest and also their versatile applications make them unique compounds in coordination chemistry, analytical chemistry, catalysis, pharmaceutical chemistry, etc. The biological activities (*e.g.* antimicrobial, antitumour, antioxidant, anti-inflammatory, anticancer, antifungal, analgesic etc.) of Schiff bases and their complexes have been widely studied. This review is ornately pronounced the synthesis and versatile applications of some Schiff bases with their transition metal complexes.

Keywords: Schiff bases, Metal complexes, Coordination chemistry, Biological activity.

Introduction

Schiff bases are those compounds which have the functional group namely imine or azomethine (–C=N–) [1]. These are formed when a carbonyl compound is condensed with a primary amine [2-4]. This type of compound was first synthesized by German scientist Hugo Schiff in 1864 and hence named after him as Schiff base [5-7]. The general structure of Schiff bases is CR'R''=RN, where R ≠ H [8]. The Schiff bases which are synthesized from aromatic aldehydes are relatively more stable than those from aliphatic aldehydes that are unstable and readily polymerizable [9].

Schiff bases act as impressive chelating agents because of containing nitrogen, oxygen or sulphur [10]. Generally these compounds act as bi-, tri-, tetra- or multi- dentate ligands [11-13]. Schiff bases are called versatile ligands as they coordinate with various metal ions in several oxidation states and geometries. Schiff bases are found to produce complexes with most of the transition metals and with lanthanides.

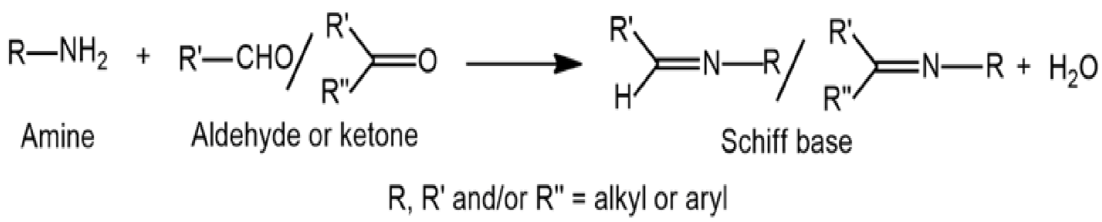

Fig. Formation of Schiff base.

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The Schiff bases and their metal complexes exhibit various biological activities *viz.,* antibacterial, antifungal, antileukemic, anti-inflammatory, analgesic, antioxidant, anticancer, antitumour, acetylcholinesterase inhibition, urease inhibition etc. [14,15]. They also exhibit clinical, biochemical and pharmacological properties [16]. Schiff bases have tremendous importance as catalyst, dyes, pigments, agrochemicals, corrosion inhibitors, polymeric stabilizers etc. [17].

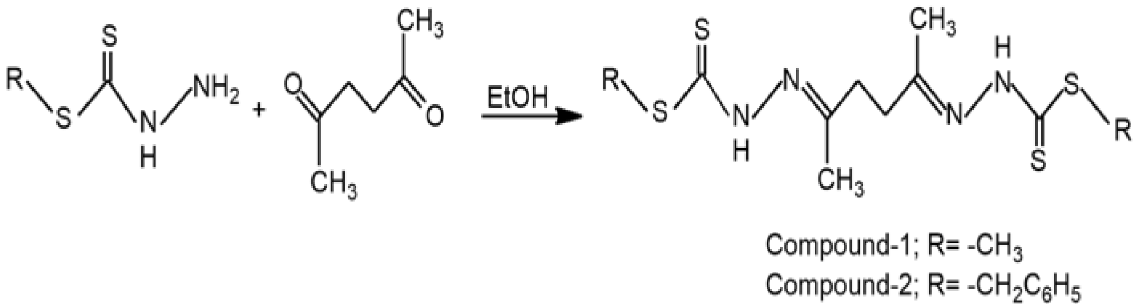
Literature Review

Synthesis of Schiff bases and its metal complexes with applications

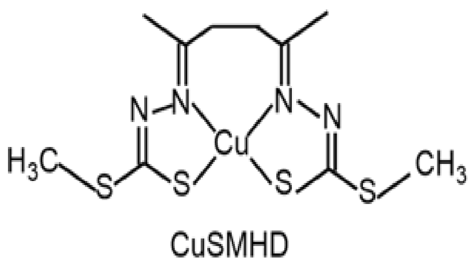
May Lee Low [18] has studied the synthesis of two new Schiff bases (Scheme 1) followed by complexation (with Cu) (Structure-1 & Structure 2), electrochemistry, antibacterial activity and cytotoxic assay. The Schiff bases and their complexes have been characterized by using several physicochemical and spectroscopic (Infrared (IR), Electron Paramagnetic Resonance (EPR) techniques. The crystal structure of the complexes was identified by single crystal X-ray diffraction method. The synthesized compounds were screened for antibacterial activity against ten strains of Gram-negative and Gram-positive bacteria which showed positive result. Both the Schiff base ligands and their Cu complexes showed potential cytotoxic activity against human breast adenocarcinoma cancer cell lines MDA-MB-231 and MCF-7 where the activity of the complexes was better than the ligands.

Hui Liu [19] synthesized a Schiff base (Scheme 2) and three transition metal complexes of the base (Structure-3).The synthesized products were characterized by elemental analysis and spectroscopic techniques (IR, UV-Vis, NMR and mass). The structures of these complexes were elucidated by single crystal X-ray diffraction method. Some selective properties of these complexes (linear optical properties, third order nonlinear optical properties (NLO), optical limiting) were investigated to get idea about the products.

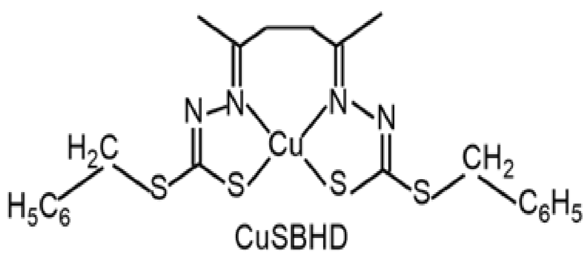
Preparation, characterization and antimicrobial activity of a new Schiff base (Scheme 3) and its Vanadium (V) complex (Structure 4) were investigated by X. W. Zhu [20]. The products have been characterized by several spectroscopic techniques including UV-Vis, IR and ^{1}H NMR. By using single crystal X-ray diffraction method the crystal structure of the complex has also been identified. From the data, it was found that the complex occupies centrosymmetric dinuclear structure with two V-atoms. The Schiff base and their complexes showed potential activities against both Gram positive (*Bacillus subtilis* and *Staphylococcus aureus*) and Gram negative (*Escherichia coli* and *Pseudomonas fluorescence*) bacteria. The complex showed better antibacterial activity than the ligand against all four bacteria. For *B. subtilis*, the activity of the complex was even better than the standard drug Penicillin G (MIC value, 1.2μg mL^{-1}).



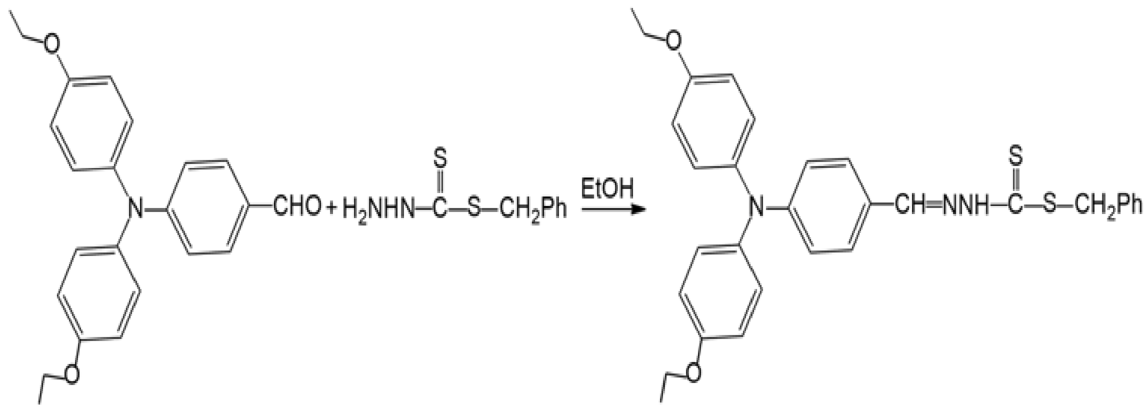
Scheme 1



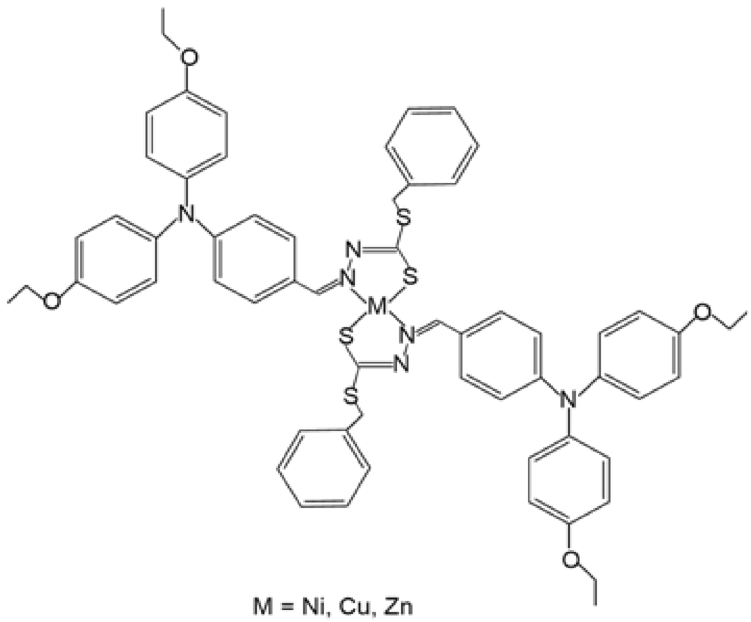
Structure 1



Structure 2



Scheme 2



M = Ni, Cu, Zn

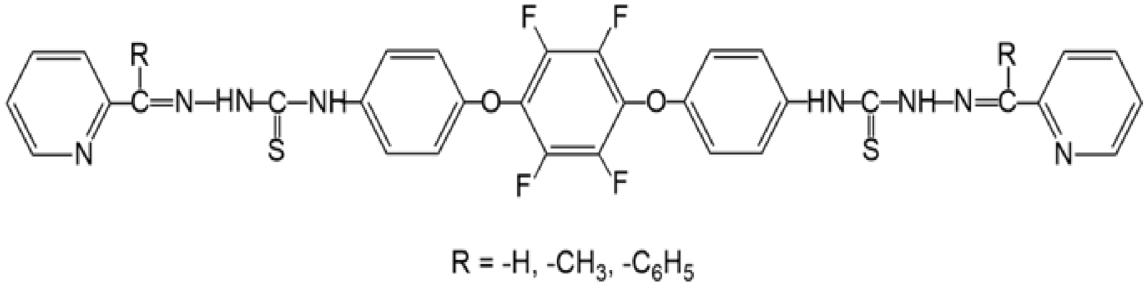
Structure 3

Pakhontsu [21] synthesized N, N'-[4,4'-(Perfluoro-1,4-phenylene) bis (oxy) bis (4,1-phenylene)] bis [2-(pyridin-2-ylmethylidene)-hydrazinecarbothioamide] (Structure 5). The author also prepared methyl and phenyl derivatives (Structure 5) of the compound and their complexes with Cu and Ni (Structure 6). The prepared complexes have biological functions and inhibit the development of the myeloid human leukemia HL-60 cancer cells.

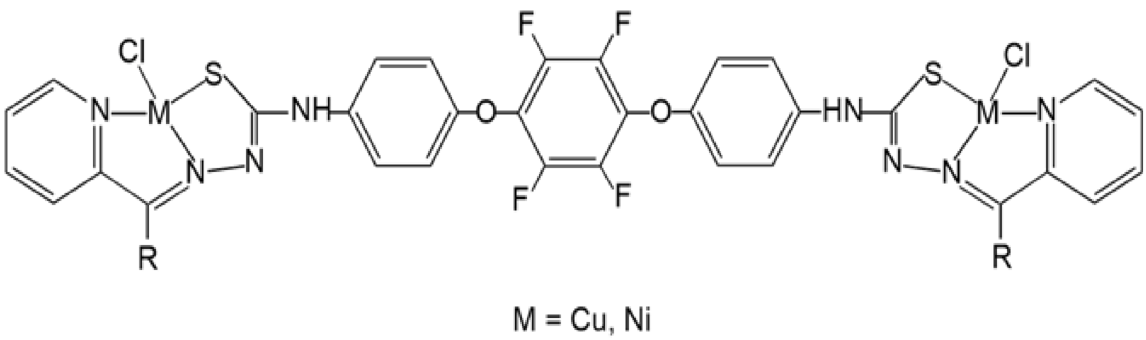
Zangrando [22] prepared a Schiff base ligand (HL) from S-benzyldithiocarbazate (SBDTC) and 3-hydroxy acetophenone in absolute ethanol (Scheme 4) and some of its metal complexes (Structure 7) .The characteristics of Schiff base ligands were determined by various methods (physical as well as chemical analysis). The properties of Schiff bases and their metal complexes were investigated under analgesic and anti-inflammatory activity. All of the synthesized compounds showed promising anti-inflammatory activity whereas analgesic activity was only limited to the complex CuL_2.

Hoq [23] reported the synthesis of eight new 2,2'-bipyridyl-5,5'-dialdehyde Schiff bases from amines containing O, S, N and F atoms (Scheme 5). The properties of the formed products were evaluated by different spectroscopic methods (IR, ^{1}H-NMR, ^{13}C-NMR and mass).

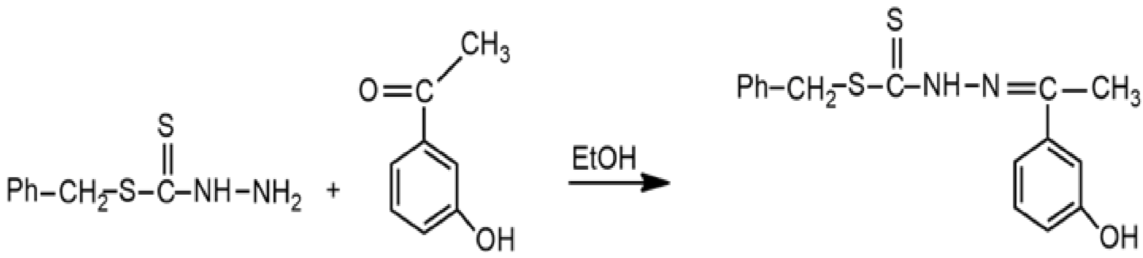
Jambol [24] used pyruvic acid and amines to synthesize Schiff bases where N and S act as donor atom. The author synthesized four complexes like thiocarbohydrazide, 2-methyl-3-thiosemicarbazide, S-benzyldithiocarbazate and S-n-octyldithiocarbazate (Scheme 6). First two Schiff bases (Scheme 14 & 15) were found as cyclic Schiff bases.



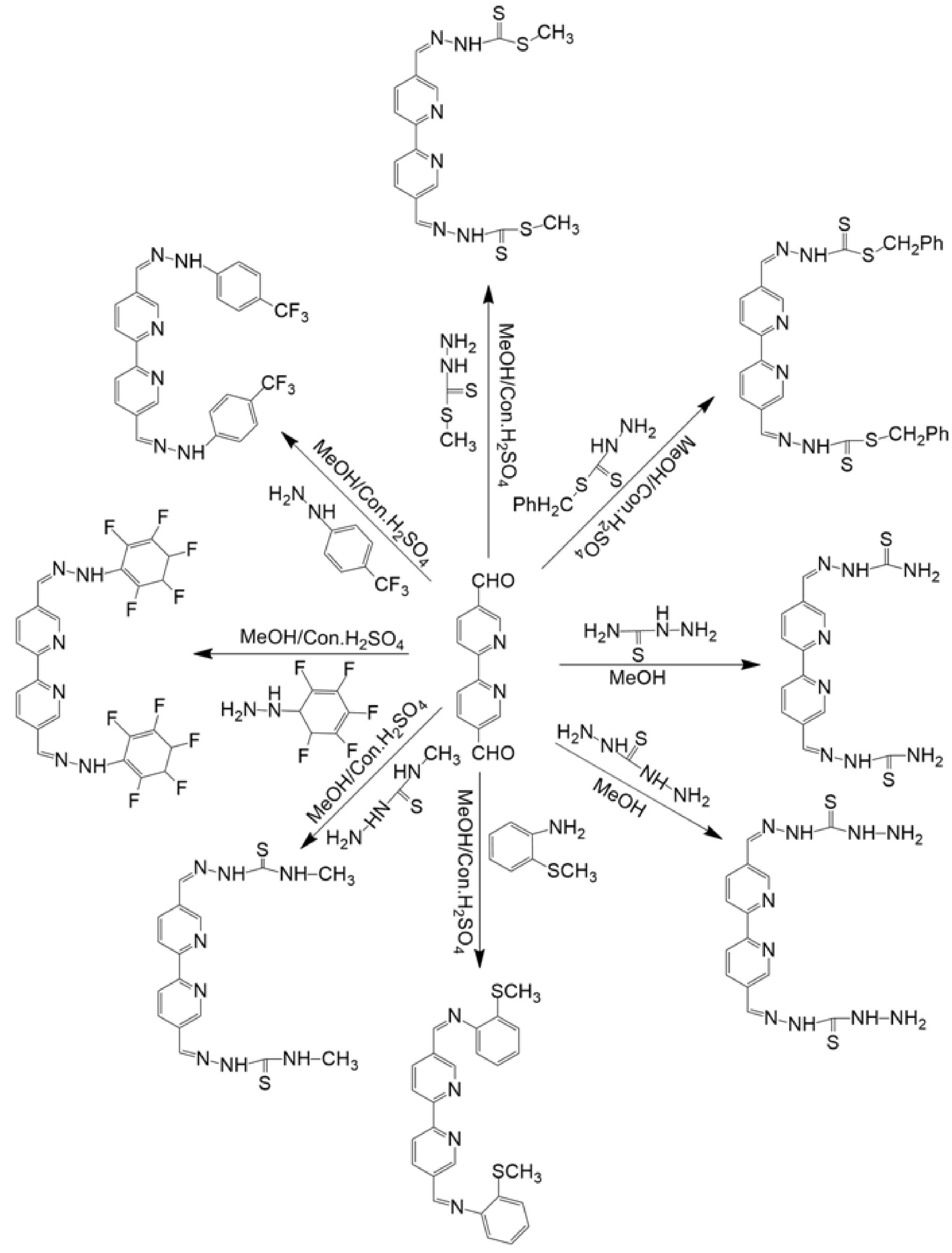
Structure 5



Structure 6



Scheme 4

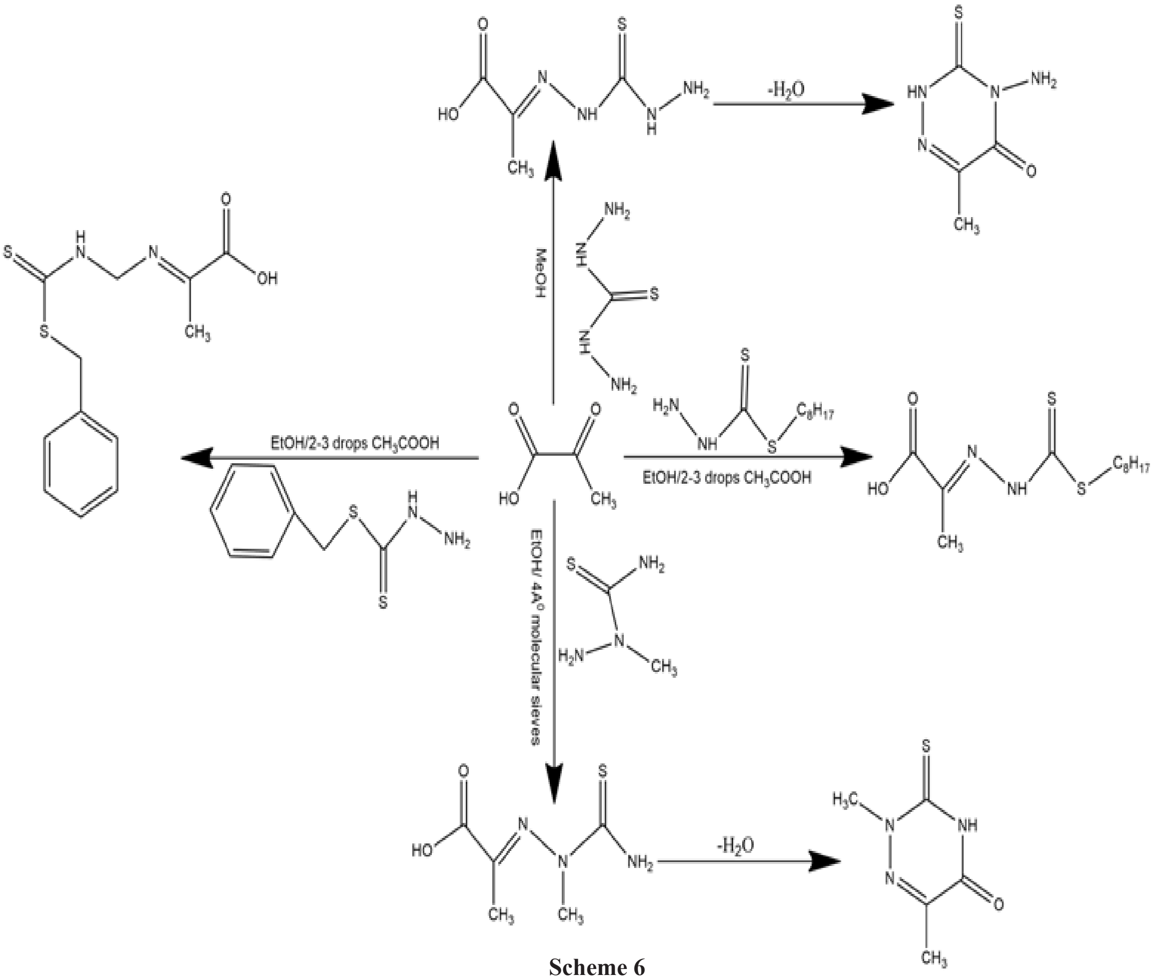


Scheme 5

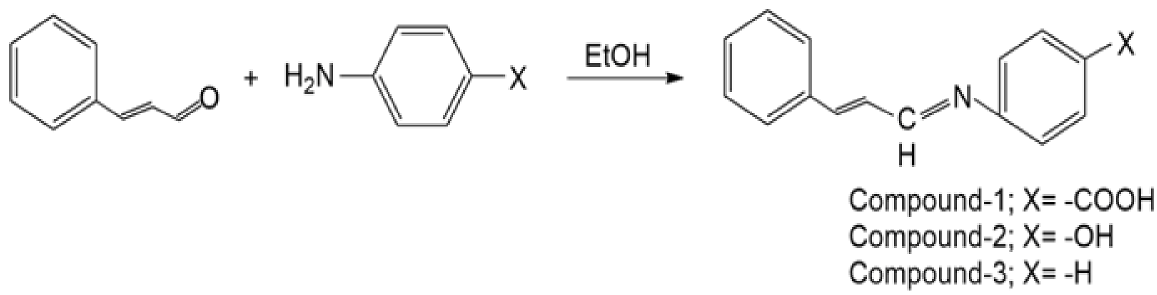
Six new Schiff bases were synthesized by S. Chigurupati [25] (Scheme7 & 8). The Schiff bases were screened for in vitro acetylcholinesterase (AChE) inhibition and antioxidant activities. Compound – 1 and 4 were showed better result as compared to other compounds in both AChE enzyme inhibition and antioxidant assays.

Cai [26] has synthesized two novel Schiff bases (Scheme-9) and their Cu complexes (Structure-8 & 9). The two copper complexes have the same activities against some microorganisms like *B. subtilis, E. coli* and *P. putida*. As for *S. aureus,* complex II is stronger than complex I.

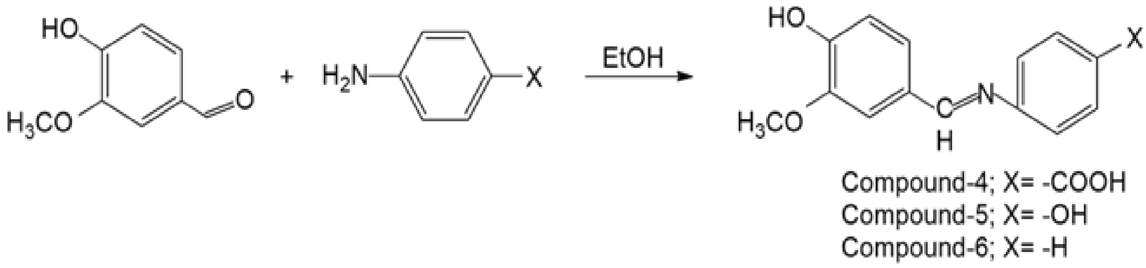
Fan [27] has synthesized an asymmetric Schiff base ligands from L-lysine, salicylaldehyde and 2-hydroxy-1-naphthaldehyde (Scheme 10). He also synthesized three complexes of rare-earth ion with La, Sm and Ho (Structure 10) and characterized them. These Schiff bases as well as their complexes showed anti-bacterial activities against *Escherichia coli*, *Staphylococcus aureus* and *Bacillus subtilis* bacterial species. Moreover, these complexes possess higher activities than Schiff base. Among these complexes, Ho^{3+} complex shows more reactivity against the test microorganism.



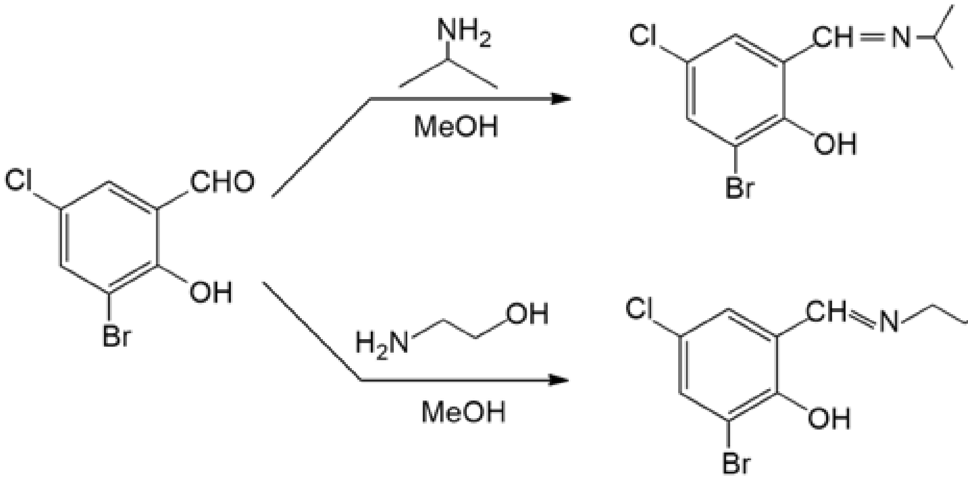
Scheme 6



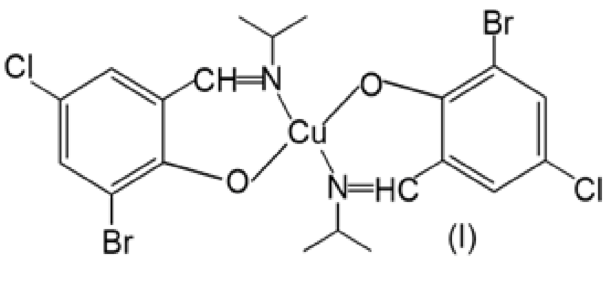
Scheme 7



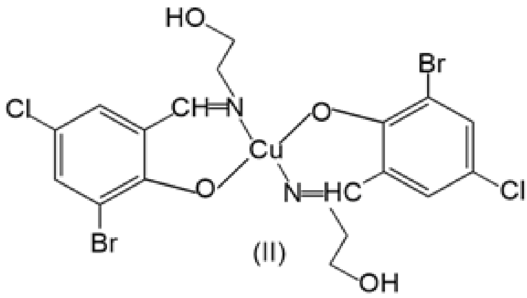
Scheme 8



Scheme 9



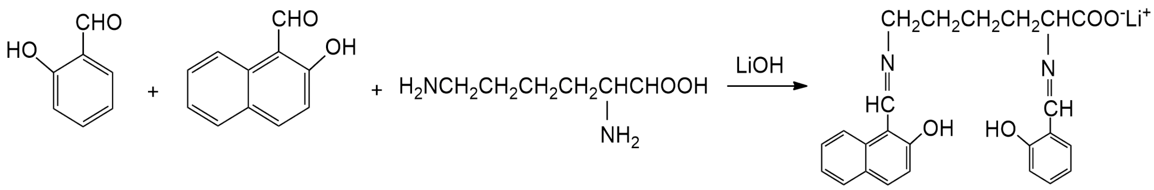
Structure 8



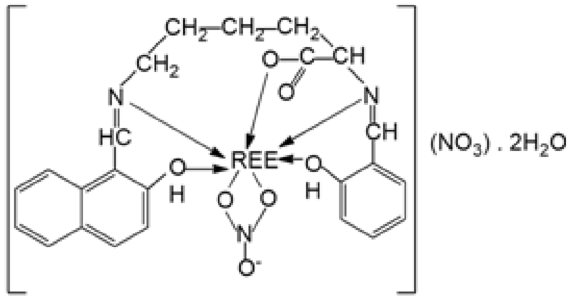
Structure 9

Guo [28] synthesized two new Schiff base namely 1-(cyclopentyliminomethyl)napthalen-2-ol and 4-nitro-2-(cyclopentyliminomethyl) phenol (Scheme 11 & 12). These bases were synthesized from cyclopentylamine with the reaction of 2-hydroxy-1-naphthaldehydeand 5-nitrosalicylaldehyde, respectively in methanol. Two new complexes of Schiff bases were also synthesized by using Zn metal (Structure 11 & 12) and complexes were characterized by different methods like single crystal X-ray diffraction, FT-IR and elemental analysis. The Schiff bases and their complexes showed stronger antibacterial activity against *Escherichia coli* and *Pseudomonas fluorescens* but weaker antibacterial activity against *Bacillus subtilis* and *Staphylococcus aureus* than Penicillin. The zinc complexes showed relatively stronger activities against all the bacteria than the Schiff base ligands.

S. Ilhan [29] indicated the synthesis of a new Schiff base from 1,6-dibromohexane, K_2CO_3, salicylaldehyde and 2,6-diaminopyridine (Scheme 13).The author also synthesized six Schiff base complexes (Structure 13 – 18) from the Schiff base. The properties of both Schiff base and the complexes were analyzed by different analytical (elemental analysis, conductivity measurements) and spectroscopic methods (NMR, UV-visible, Mass spectroscopy). The analytical data indicated that all of the complexes are diamagnetic in nature and only the Cu(II) complex has binuclear structure.

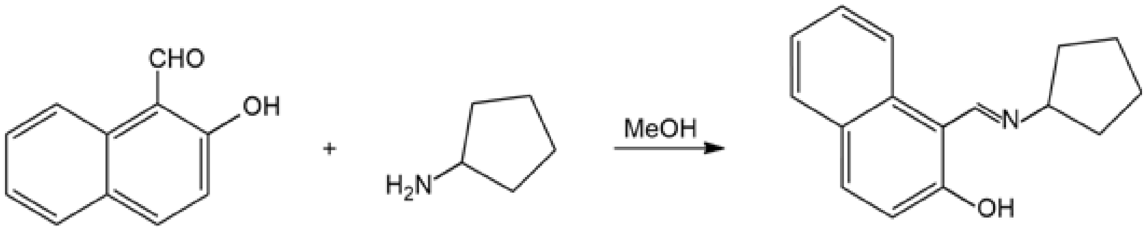


Scheme-10

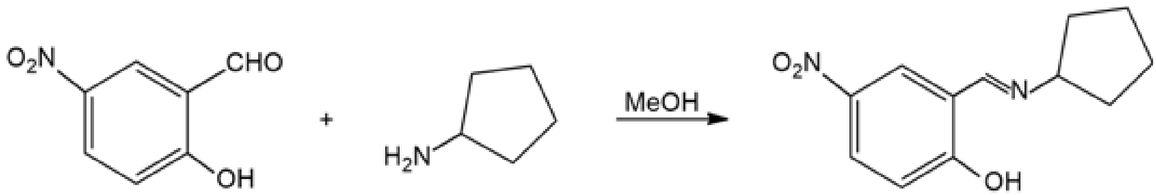


REE = La, Sm, Ho

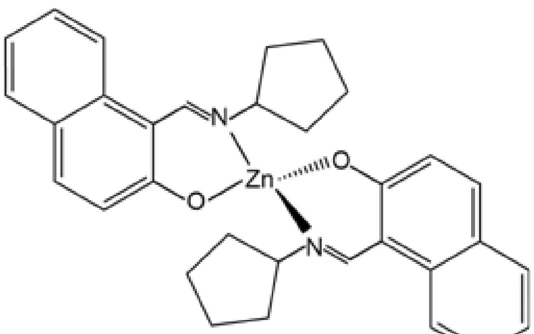
Structure-10



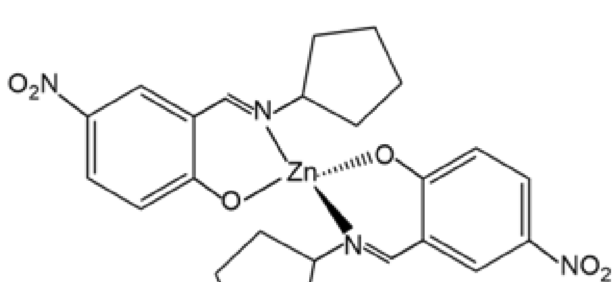
Scheme 11



Scheme 12

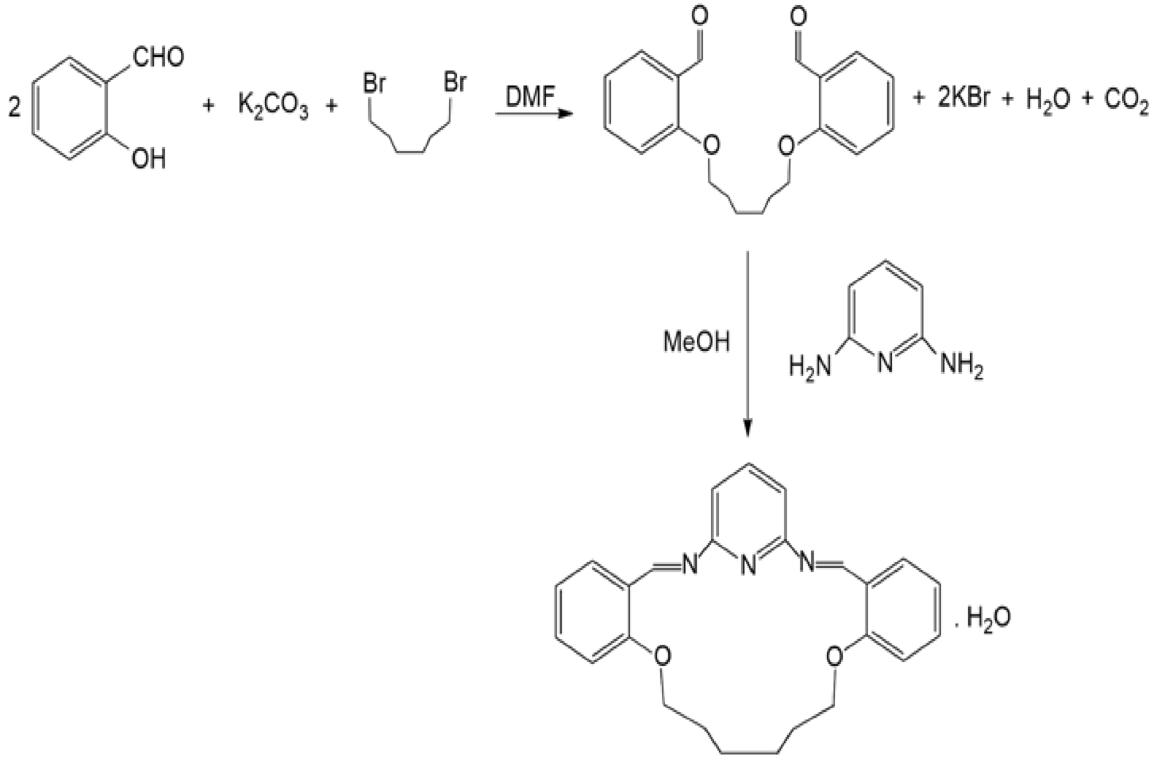


Structure 11

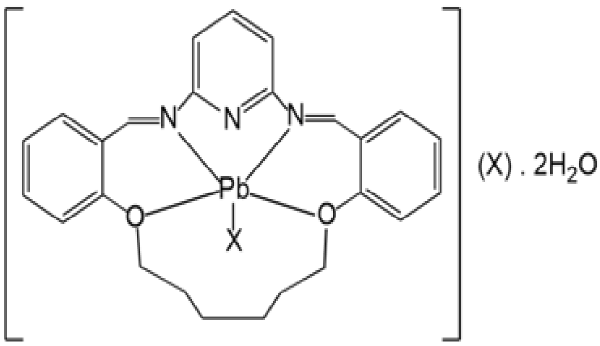


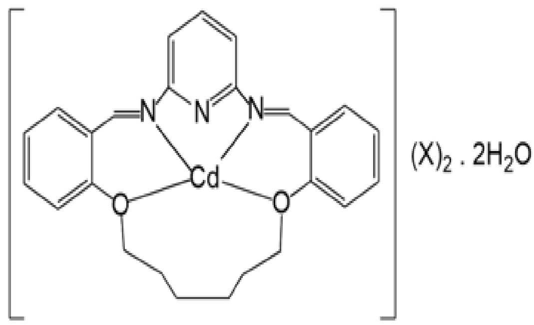
Structure 12

Khalaji [30] showed the synthesis of a Hg^{2+} complex (Structure 19) with a Schiff base ligand (Scheme 14) that produced from the reaction of N,N'-bis[(E)-2-benzyl-idenepropylidene]ethane-1,2-diamine with $HgBr_2$. The synthesized products was characterized by elemental analysis and the crystal structure of the complex was investigated by single crystal X-ray crystallography. From the analytical data, the complex was found to have distorted tetrahedral geometry and non-classical intra- and intermolecular hydrogen bonds were also found in the complex structure.



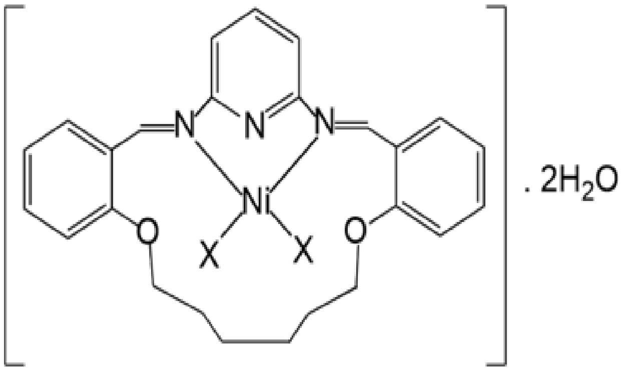
Scheme 13



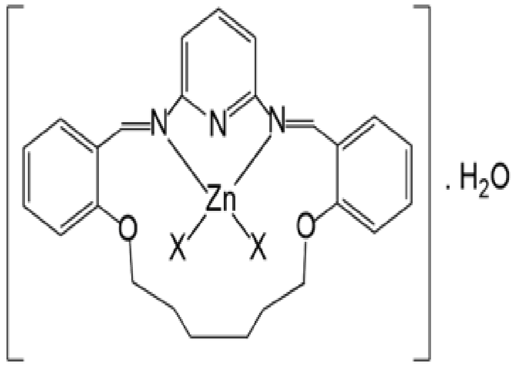


Structure 13

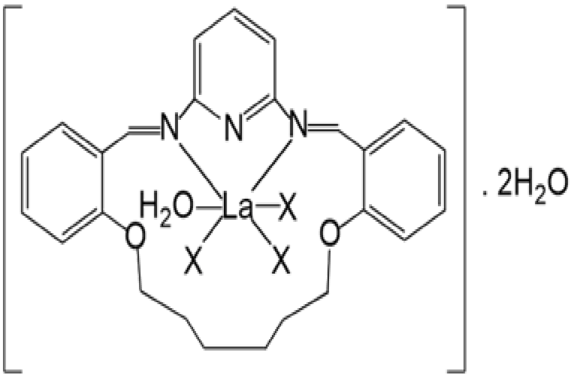
Structure 14



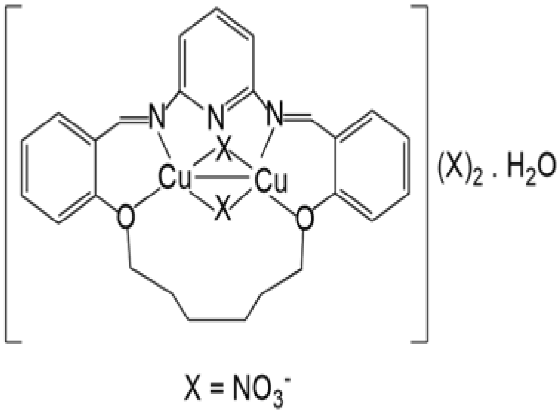
Structure 15



Structure 16



Structure 17



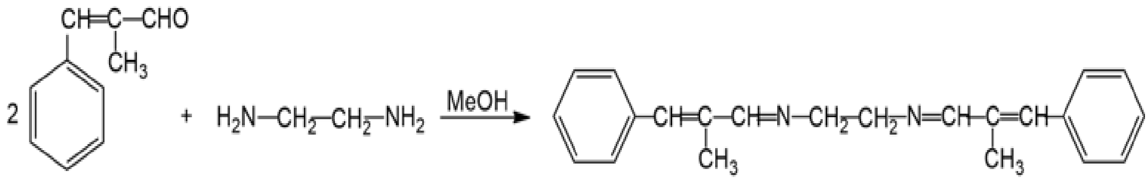
Structure 18

Jone Kirubavathy [31] showed a reaction to synthesis a tridentate Schiff base (Scheme 15) and its Co (III) complex (Structure 20) from salicylaldehyde, ethylene diamine and $CoCl_2$. $6H_2O$ in ethanol. The products were fully characterized by elemental and spectroscopic analysis. The complex exposed evident activity against *S. aureus*, *S. paratyphi* and *A. niger* and considerable activity against *B. subtilis* and *K. pneumonia* microbial species. Concentration of Co(III) influenced its anti-microbial activity and found that its activity increased with the increase of concentration.

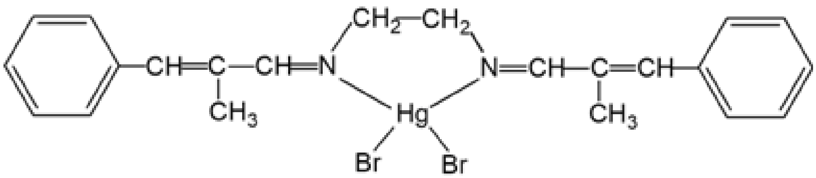
Liu [32] synthesized three Schiff base ligands 2-[1-(2-emthylamino-ethylimino)ethyl]phenol (I), N,N'-ethylene-bis(2-hydroxyacetophenonylidene-imine) (II) and 2-(1-iminoethyl)phenol (III) (Scheme-16) and two metal complexes with $Cu^{2+}$and Ni^{2+} (Structure 21 & 22). The products were fully characterized by elemental analysis, FT-IR spectroscopy and single crystal X-ray diffraction. In copper complex, ligand I and ligand II are coordinated with two Cu(II) atom whereas in nickel complex, two ligands (III) are coordinated with a Ni(II) atom.

Rezaeivala [33] synthesized two Schiff bases from 2-[2-(2-formylphenoxy)ethoxy]-benzaldehyde and/or 2-[2-(3-formylphenoxy) propoxy]benzaldehyde and ethanol amine (Scheme 17) and the complexes of Cu^{2+}, Ni^{2+} and Co^{2+} with the bases (Structure 23). The metal complexes were fully characterized through microanalysis, single crystal X-ray diffraction, IR and mass spectroscopy. From the analytical data, it was found that metal atoms coordinated with both nitrogen and oxygen atoms of the ligands.

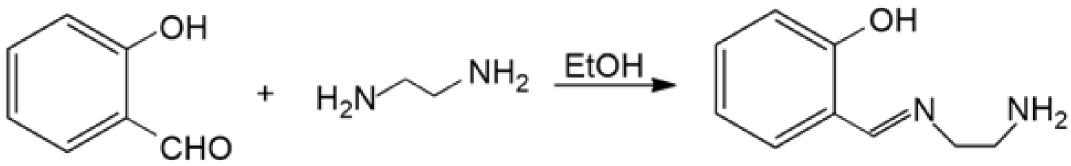
Shi [34] showed the synthesis pathway of the Schiff bases 4-chloro-2-[(2-piperazin-1-ylethylimino)methyl]phenol and 4-chloro-2-[(3-diethylaminopropylimino)methyl]-phenol (Scheme 18) and their Zn(II) complexes (Structure 24 & 25). The Schiff bases were synthesized from 5-chlorosalicylaldehyde, N-(2-aminoethyl) piperazine and N,N-diethylpropane-1,3-diamine. The formed complexes showed the inhibitory activities against urease enzyme. Both complexes showed better activities than the Schiff bases and $ZnBr_2$ used in the complex preparation.



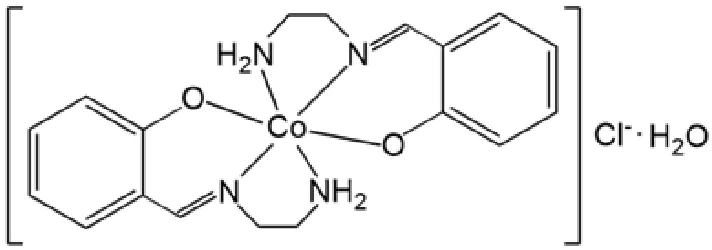
Scheme 14



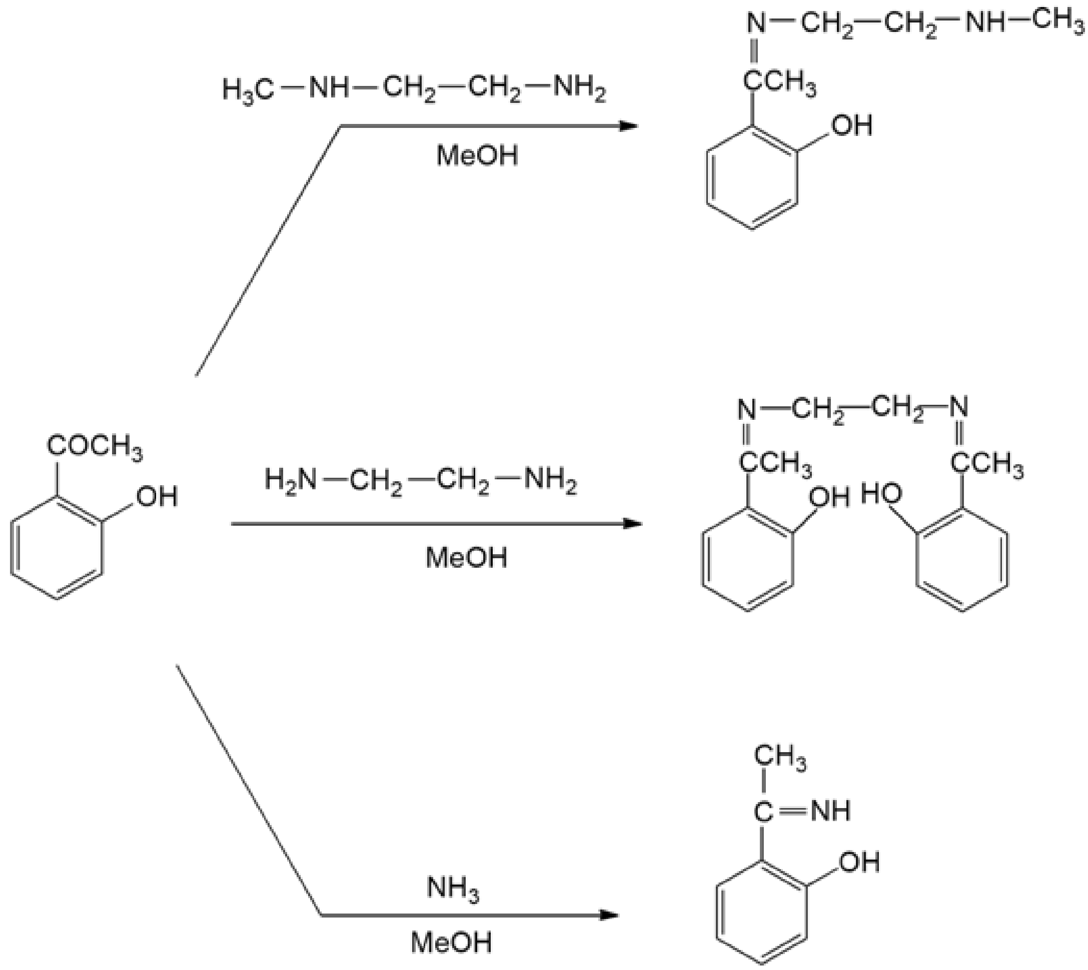
Structure 19



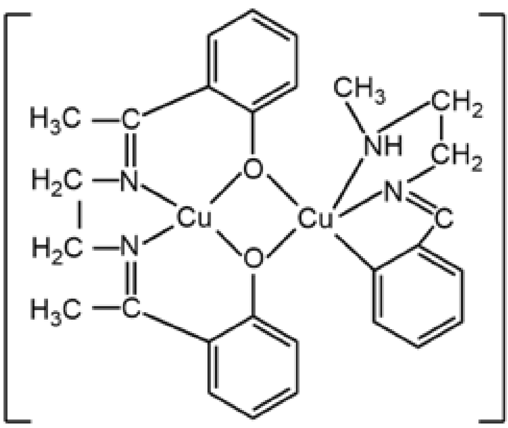
Scheme 15

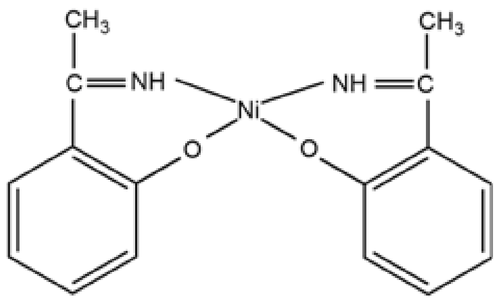


Structure 20

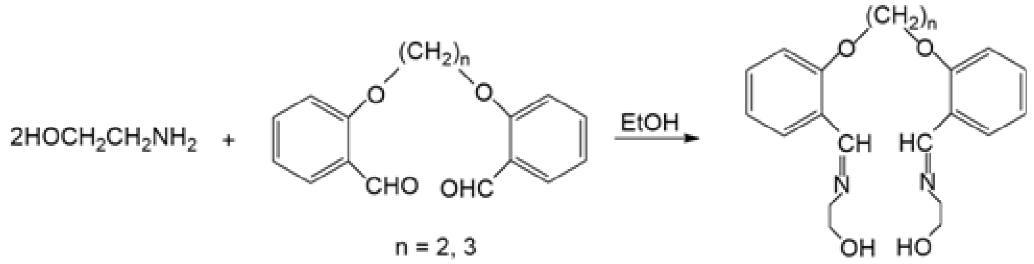


Scheme 16

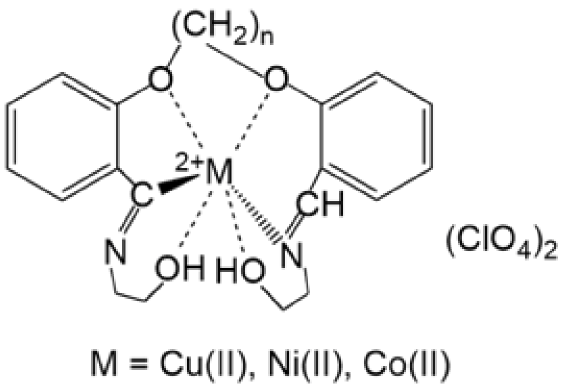




Structure 21 **Structure 22**



Scheme 17



Structure 23

A novel Schiff base N,N'-bis-(furaldehyde) diethylenetriamine (Scheme 19) and its Mn (II) complex (Structure 26) were prepared and characterized by Wang [35]. The characterization techniques used here are IR spectroscopy and X-ray crystallographic analysis. The crystal was found as orthorhombic and the coordination geometry was trigonal-bipyramidic.

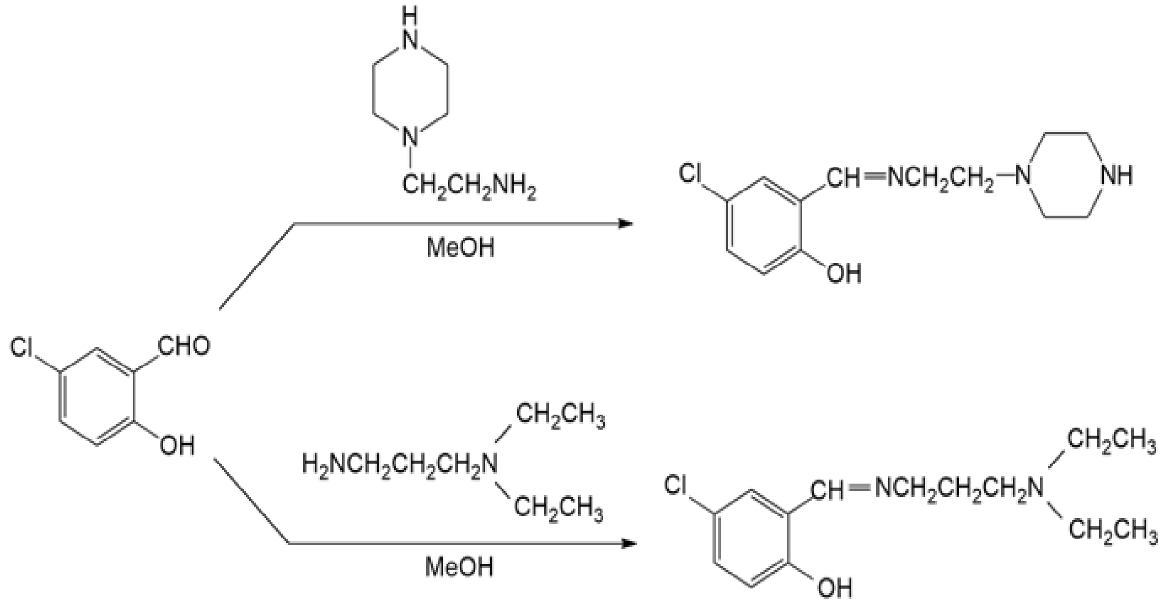
John P. Grundhoefer [36] has reported the synthesis and characterization of a Schiff base namely 1,1′-(((2-hydroxypropane-1,3-diyl)bis(azanylylidene))bis(methanylylidene)) bis(naphthalen-2-ol) (Scheme-20) derived from 2-hydroxynaphthaldehyde and 1,3-diamino-2-propanol and its two mono nuclear Cu(II) and Ni(II) complexes (Structure 27 & 28). Two complexes had 4- coordinated square planar structure. Phenolic oxygen and imine nitrogen have lone paired electron. 'These atoms are coordinated with the central metal atom, but the central pendant hydroxyl group did not participate in the coordinate bond formation.'

Mohammad Habibi [37] has reported the synthesis and structure elucidation of a Schiff base (Scheme 21) derived from 2-pyridinecarboxaldehyde and 4-nitro-o-phenylenediamine and its Ni (II) complex (Structure-29). From this study, it was identified that the Schiff base is tridentate and the complex is square planar.

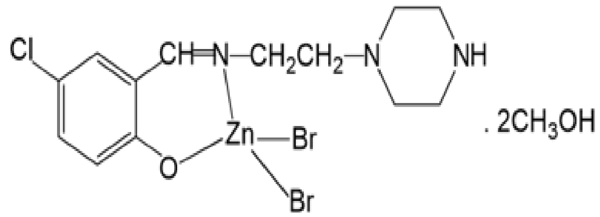
Kargar [38] has synthesized and characterized a new Pd(II) complex (Structure 30) obtained from bis(5-methoxysalicylidene)-4-methyl benzene-1,2-diamine (Structure-31). The crystal structure was elucidated from different analytical studies. The central metal atom coordinated with two N-atoms and two O-atoms.

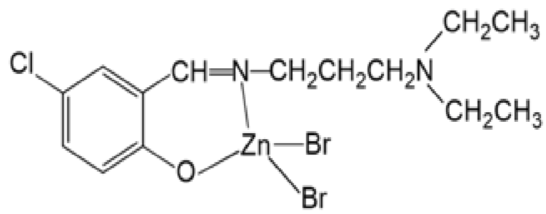
Synthesis and characterization of Schiff base [N[1]-(4-methoxy-benzylidene)-ethane-1,2-diamine] (Scheme 22) and its four Co(II), Cu(II), Ni(II) and Zn(II) metal complexes (Structure 32) were investigated [39] (Scheme 22). All the metal complexes synthesized here were found tetrahedral in nature.

M. Karunakar [40] has studied the synthesis and characterization of a Schiff base N-(4′-(5-(4″-octyloxyphenyl)-1,3,4-oxadiazol)-2-phenyl)- 4‴- octyloxy-salicylaldimine (Scheme 23) and its complexes with Lanthanide metals (Structure 33). The Schiff base was found to have mesogenic behavior from thermal and optical behavior studies, where the complexes were non-mesogenic.

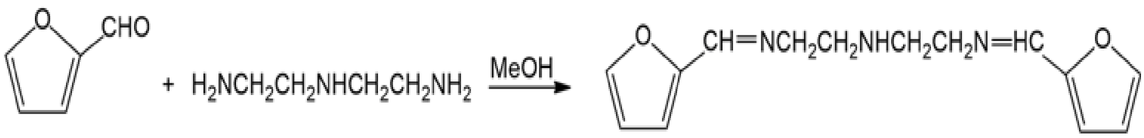


Scheme 18

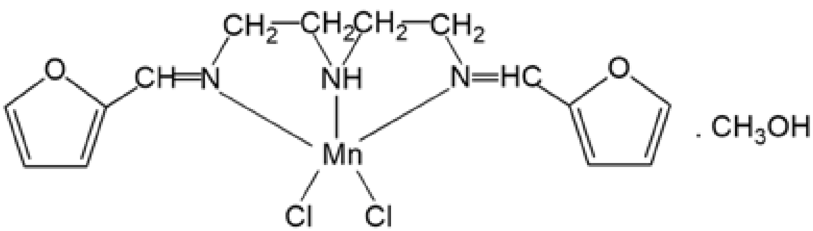




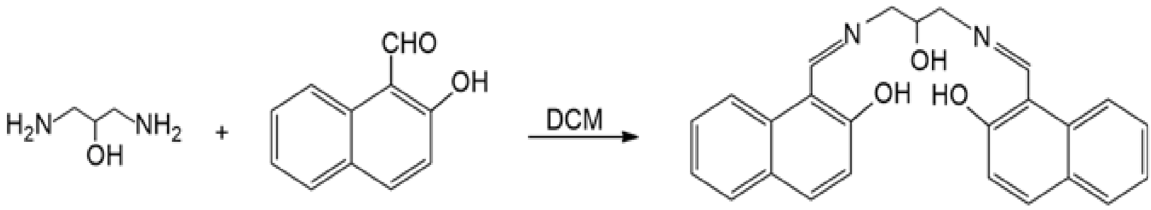
Structure-24 **Structure-25**



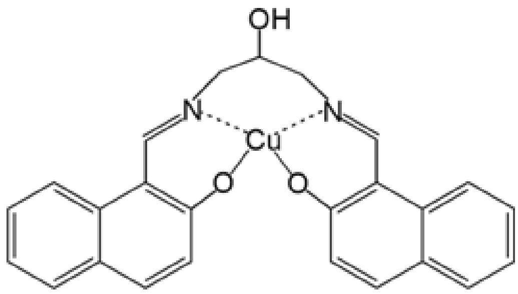
Scheme 19



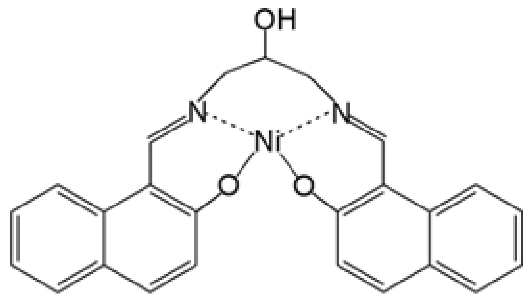
Structure 26



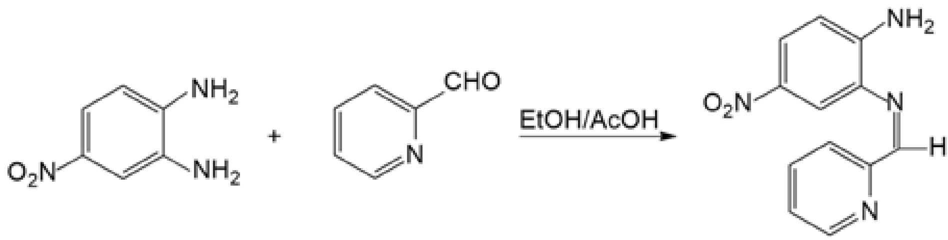
Scheme 20



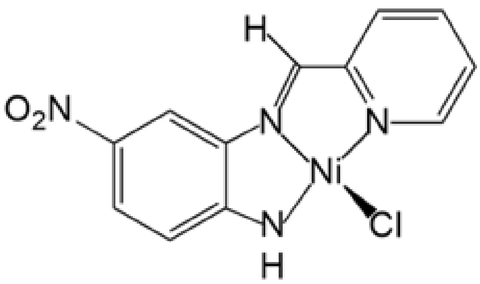
Structure 27



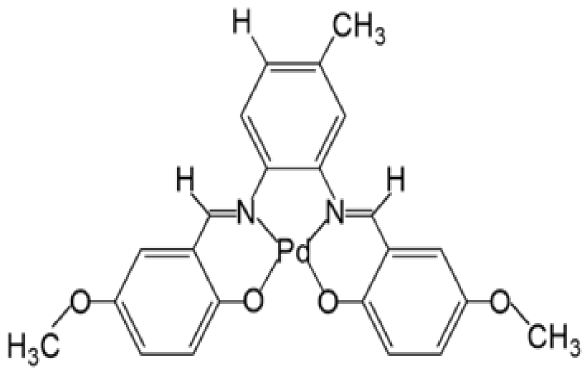
Structure 28



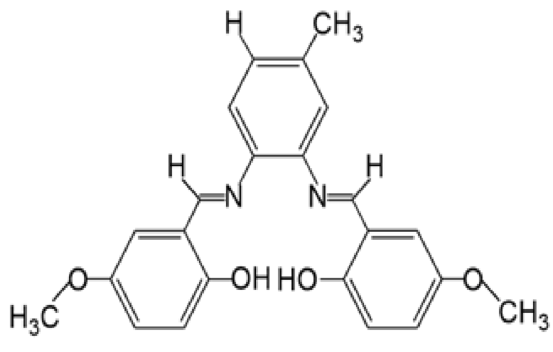
Scheme 21



Structure 29



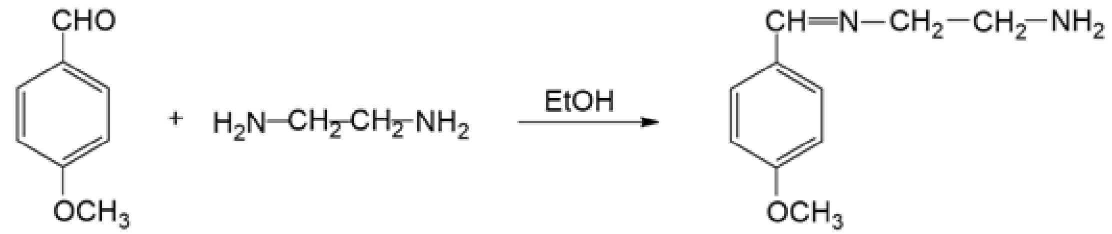
Structure 30



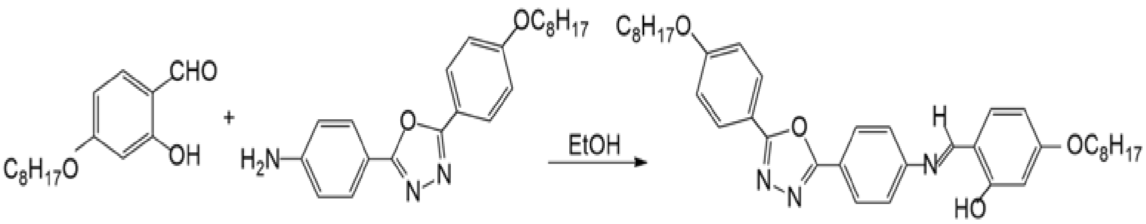
Structure 31

A. Jayamani [41] has synthesized a Schiff base ligand N,N′-bis(2-hydroxy-1-naphthyl)-1,4-bis(3-imino-propyl)piperazin (Scheme 24) and its two mononuclear copper(II) and nickel(II) complexes (Structure 34). The products were characterized by single crystal X-ray diffraction analysis. Both complexes showed impressive binding abilities to bovine serum albumin protein (BSA). The copper(II) complex displayed better interaction with calf thymus DNA (CT-DNA) than the nickel(II) complex.

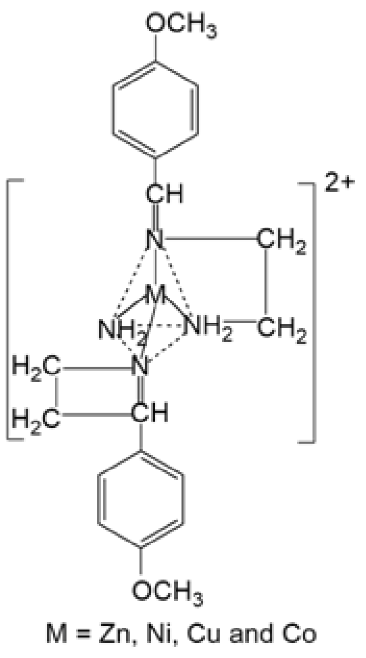
M. Shabbir [42] has studied the synthesis and characterization of four new Schiff bases based on ether (Scheme 45) and their copper(II) complexes (Structure 25). He has also studied the several biochemical activities of the synthesized products.



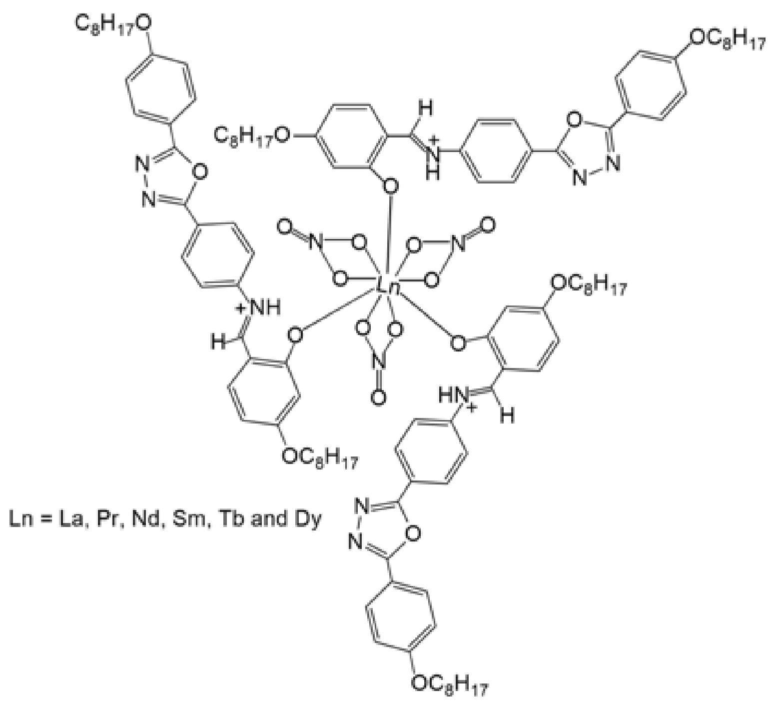
Scheme 22



Scheme 23



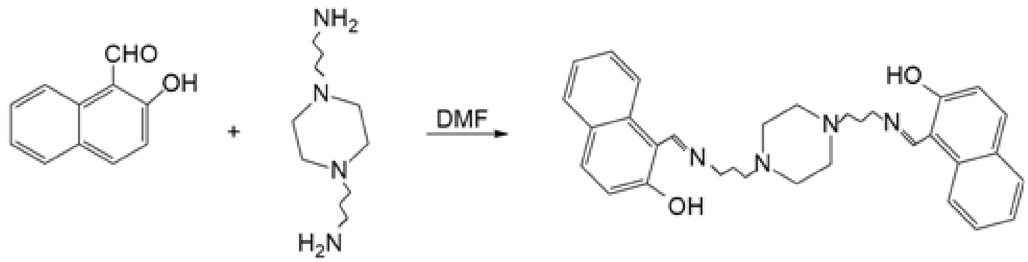
Structure 32



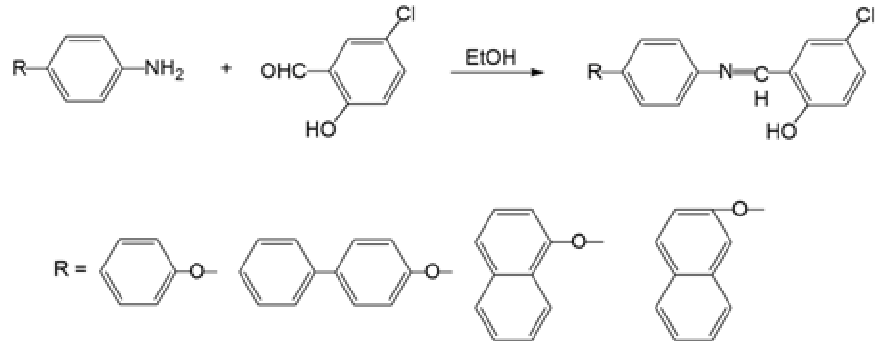
Structure 33

The copper(II) complexes showed higher activity than the ligands in brine shrimp cytotoxic assay, potato dise antitumor assay and DPPH free radical scavenging assay. But in DNA damage assay all ligands showed significant protection behavior whereas the complexes remained neutral.

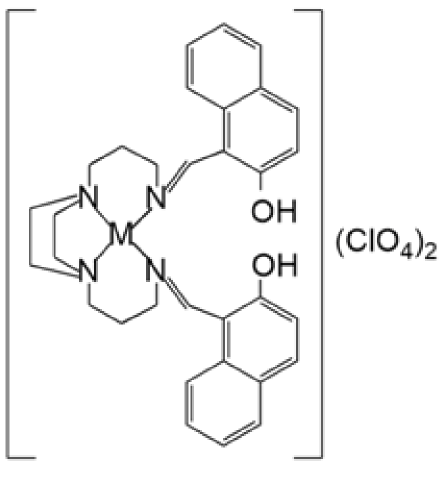
Synthesis and characterization of a Schiff base 2-((2-mercaptophenyl)imino)-1,2-diphenylethan-1-ol (Scheme 26) and its Co(II), Cd(II), La(III) and Gd(III) metal complexes (Structure 36 & 37) were investigated [43]. The heavy metal removal activity of the Schiff base was studied. The result indicated that the Schiff base synthesized here can remove heavy metals. The Cd(II) complex exhibit better cytotoxicity effect on Caco-2, Vero and MCF-7 cells than the other complexes and the ligand.



Scheme 24

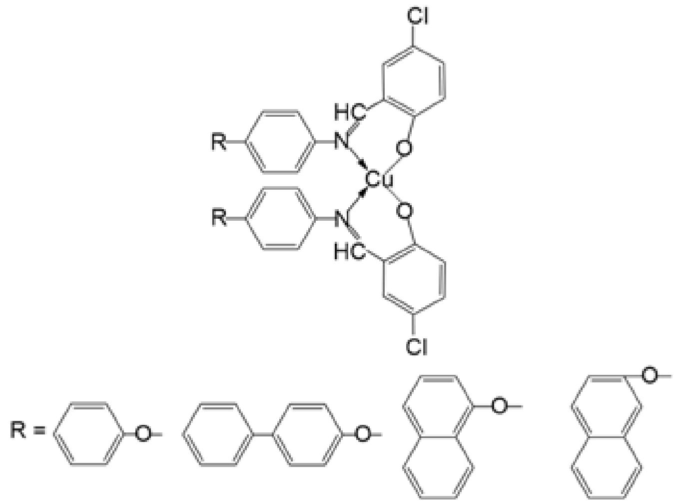


Scheme 25



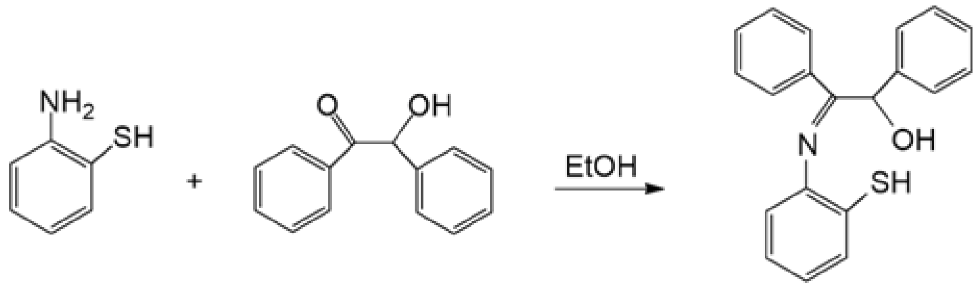
M = Cu, Ni

Structure 34

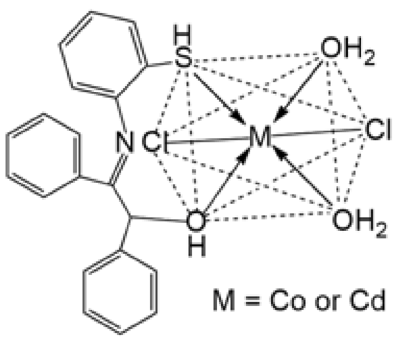


Structure 35

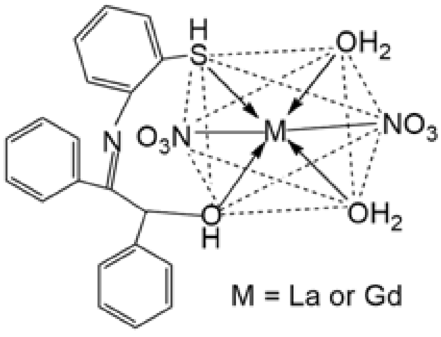
Tahmasebi [44] has investigated the synthesis and characterization of two schiff bases namely {(E)-[2-bromoethyl)imino]methyl}-2- naphthol and 2-{(E)-[2-chloroethyl)imino]methyl}-2-naphthol (Scheme 27) and their vanadium(IV) complexes (Structure 38 & 39). From the structural study, the geometry of the complexes was found as distorted square pyramidal. They also studied the catalytic activity of the complexes in alkene epoxidation reaction. The study showed that high epoxide yield was obtained in ratio 1:4 and 1:3 of cyclooctene/oxidant, by using tert-butyl hydroxide (TBHP) as oxidant.



Scheme 26



Structure 36



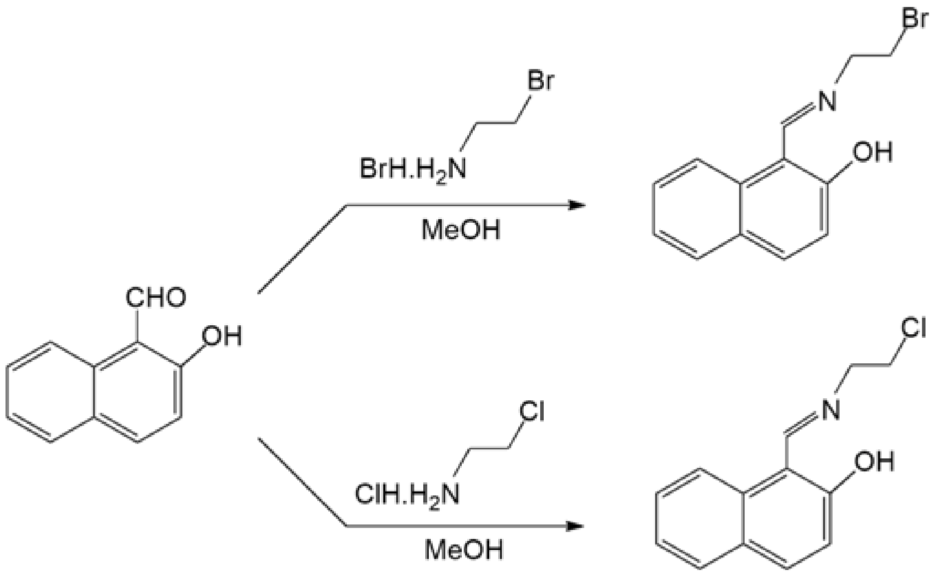
Structure 37

Kuddushi [45] has studied the complexation of Cu (II), Co (II), Mn (II), Fe(II), Ni (II) and V (II) (Structure 40) with Schiff base derived from 5- bromo 2-hydroxy benzaldehydeand and aniline (Scheme 28). The complexes and the Schiff base ligand showed very impressive antibacterial properties against the experimental bacteria (*E. coli* and *B. subtilis*).

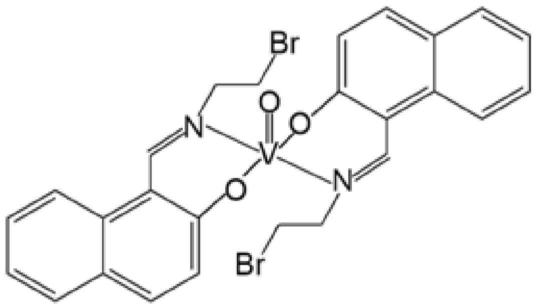
A novel Azo- Schiff base ligand (Scheme 29) and some of its metal complexes with Ni(II), Co(II), Pd(II) and Pt(IV) metal ions (Structure 41 – 43) were synthesized and characterized by W. Al Zoubi [46]. They also studied the activation thermodynamic parameters and the antibacterial activities. The products exhibited antibacterial activities against both gram positive (*B. subtilis* and *S. aureus*) and gram negative bacteria (*E. coli* and *P. aeruginosa*).

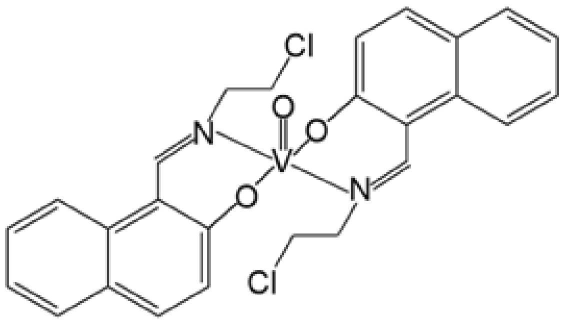
Three Schiff bases were synthesized from 2, 5-dihydroxyacetophenone (Scheme-30). The complexes of Cu^{2+}, Zn^{2+}, Cd^{2+} and Hg^{2+} with these bases (Structure 44 – 46) were also prepared. The resulted complexes were

characterized by using various physical and spectroscopic measurements. The antimicrobial activities of the products were investigated against one gram positive bacteria (*S. aureus*), three gram negative bacteria (*E. coli*, *K. pneumonia* and *P. vulgarius*) and one yeast (*C. albicans*). First (L_1) and second ligand (L_2) showed better activity than the third one (L_3). The metal complexes also exhibited the antimicrobial activities. This study was done [47].

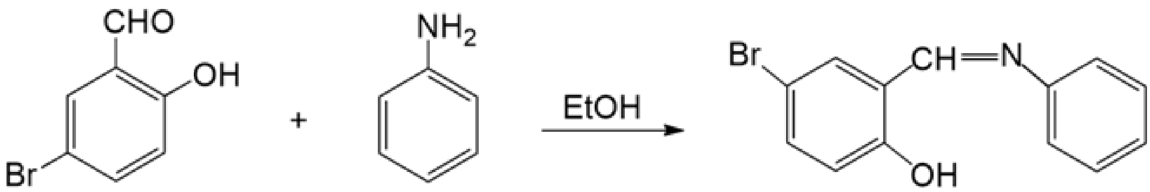


Scheme 27

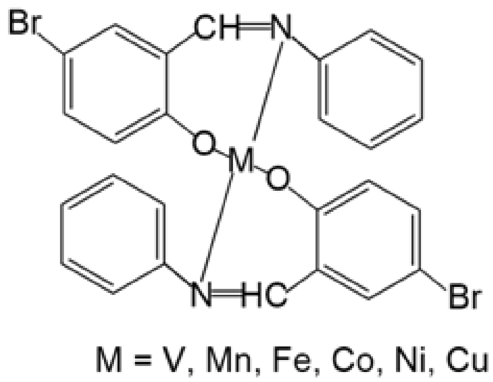




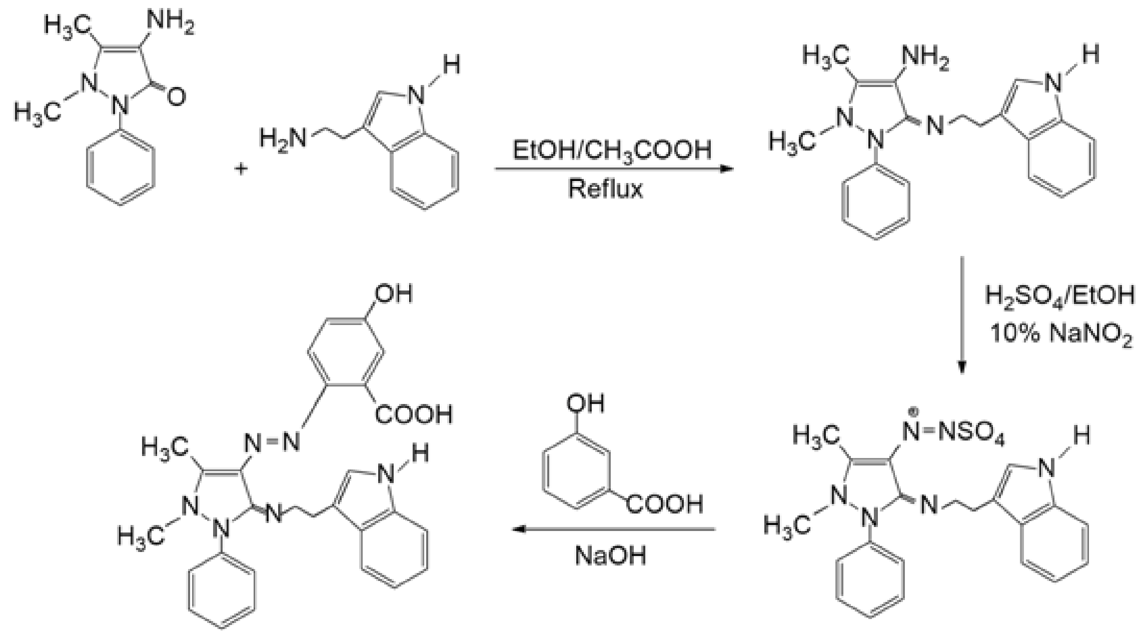
Structure 38 **Structure 39**



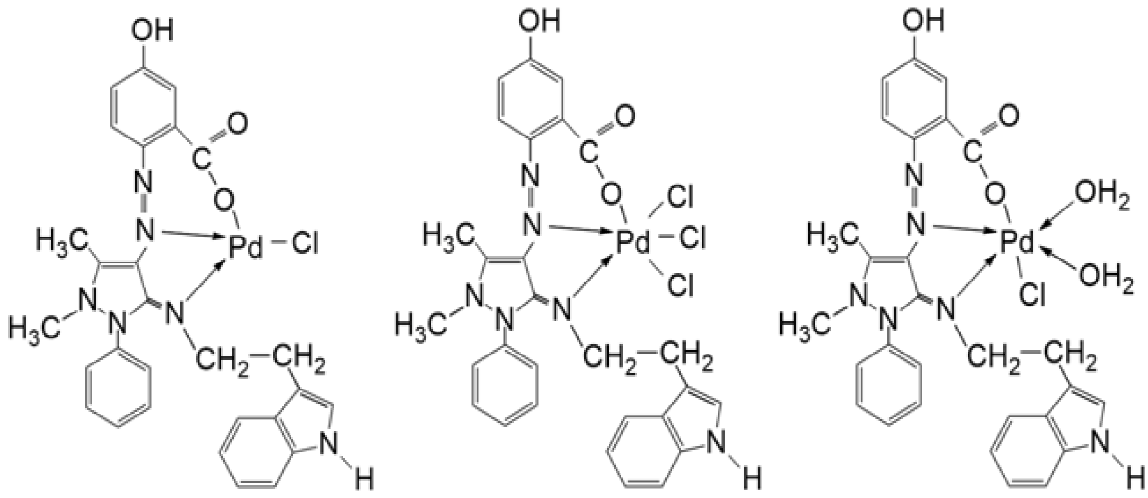
Scheme 28



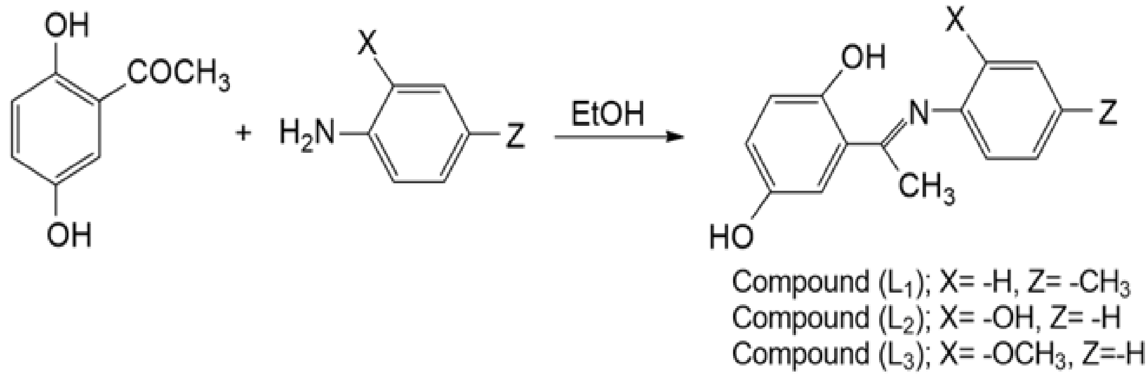
Structure 40



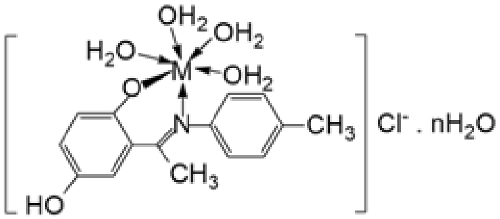
Scheme 29



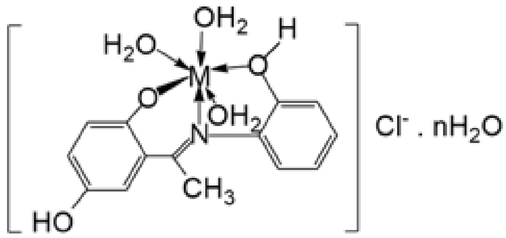
Structure 41 **Structure 42** **Structure-43**



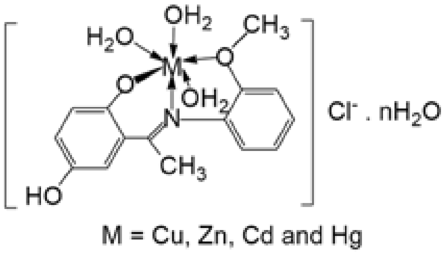
Scheme 30



Structure 44



Structure 45



Structure 46

Conclusion

Schiff bases perform as a significant class of chemical compounds since of their ability to form metal complexes, their pharmacological properties and industrial applications. However, the biological activities of these classes of compounds promote further research. On reviewing the literature, it is proved that Schiff base metal complexes exhibit better biological activity than the parent Schiff base ligands. On chelation, the polarity of the metal ions is reduced by delocalization of the π- electrons over the rings. Hence the lipophilic character of the complexes is increased. The penetration of the complexes into the microorganism's lipid membrane is increased by the increased lipophilicity which blocks the metal binding sites inside the enzymes of the microorganisms. Other factors including the nature of the donor atoms/metal ions/counter ions that neutralize the complex, the geometric structure of the complex, the total charge on the complex ion, solubility, conductivity and bond length between metal and ligand influence the increased activity of the Schiff base metal complexes. Schiff base compounds have been shown to be promising leads for the design of more competent antimicrobial agents. Advances in this field require analyses of the structure activity relationships of the Schiff bases as well as the mechanism of action of these compounds.

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مراجعة للتوليف والتطبيقات متعددة الاستخدامات لبعض قواعد الشيف المختارة مع مجمعاتها المعدنية الانتقالية

الخلاصة: غطت سندات قاعدة شيف ومجمعاتها المعدنية الانتقالية مجالًا كبيرًا في الكيمياء يتم ملاحظته. يُنظر إلى بروابط قاعدة شيف على أنها بروابط خاصة حيث يتم تحضيرها بسهولة عن طريق تفاعل التكثيف بين مشتقات الألدهيد والأمينات. ومع ذلك لديهم طريقة إعداد بسيطة. لكن تطبيقاتها متعددة الاستخدامات تجعلها مركبًا فريدًا في كيمياء التنسيق ، والكيمياء التحليلية ، والحفز الكيميائي ، والكيمياء الصيدلانية ، إلخ. تم دراستها على نطاق واسع. هذا الاستعراض واضح بشكل مزخرف في التوليف والتطبيقات متعددة الاستخدامات لبعض قواعد شيف مع مجمعاتها المعدنية الانتقالية.