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METAL COMPLEXES OF SCHIFF BASE DERIVED FROM SALICYLALDEHYDE – A REVIEW

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ABSTRACT

Salicyaldehyde and their derivatives can be condensed with amines in 1:1 and 2:1 ratio to form bi, tri and tetra dentate NO and N2O2 Schiff base Ligands. They can contain additional donor atoms like oxygen, sulphur, nitrogen etc. which makes them Suitable chelating ligands to coordinate with Metal ions to form Schiff base complexes. Thus provides the possibility synthesis of large number of Schiff base complexes with divers' structural feature. The model system including those with bidentate, tridentate and tetra dentate Schiff base derived from Salicyaldehyde and their coordination chemistry are summarized in this review.

Key Words: Metal complexes, salicylaldehyde, Schiff Bases, Bidentate, Tridentate, Tetra dentate

INTRODUCTION

Salicylaldehyde

Salicylaldehyde is a key precursor to a variety chelating agent, some of which are commercially important, salicylaldehyde is a common highly functionalized arene that has often been exploited as a precursor to still other chemical.

Salicylaldehyde is converted to chelating ligands by condensation with amines. With ethylenediamine, it condenses to give the ligand salen. Hydroxylamine gives salicylaldoxime. Oxidation with hydrogen peroxide gives catechol (1,2-dihydroxybenzene) (Dakin reaction)[1]. {Dakin, 1923 #47}Condensation with diethyl malonate gives a derivative of the heterocycle coumarin [2] via an aldol condensation.

Schiff bases

Schiff bases are aldehyde- or ketone-like compounds in which the carbonyl group is replaced by an imine or azomethine group[3].Schiff bases are versatile ligands synthesized from the condensation of an amino compound with carbonyl compounds[4,5,6] and were first reported by Hugo Schiff in 1864. Formation of Schiff base generally takes place under acid or base catalysis or with heat. The common Schiff bases are crystalline solids, which are feebly basic but at least some form insoluble salts with strong acids[5].

Today, Schiff bases are used as intermediates for the synthesis of amino acids or as ligands for preparation of metal complexes having a series of different structures[5].

Schiff base metal complexes

Schiff bases are the most widely used organic compounds[7] for industrial purposes and also exhibit a broad range of biological activities.[3]

Schiff base compounds and their metal complexes are very important as catalysts in various biological systems, polymers, dyes and medicinal and pharmaceutical fields[4,8] they comprise miscellaneous therapeutically potent applications in the field of medicinal chemistry.[9] Their use in birth control, food packages and as an O_2 detector is also outlined[8] Schiff's bases chelates also used in quantitative analysis as an analytical chemical reagents and/or separation reagents have been also listed and discussed[6] and synthetic applications in the field of the organic and inorganic chemistry.[9]

They have been shown to exhibit a broad range of biological activities, including antifungal, antibacterial, antimalarial, antiproliferative, anti-inflammatory, antiviral, and antipyretic properties.[3,7]

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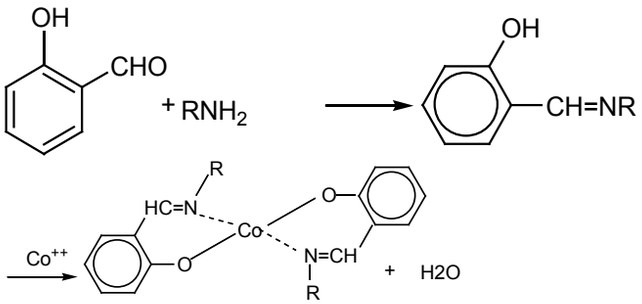
Schiff's base and their copper complexes possess remarkable properties as catalystsin various biological systems, polymers, dyes, antimicrobial activities, antifungal activities, antiviral activities insecticides, antitumor and cytotoxic activities, plant growth regulator, enzymatic activity and pharmaceutical fields.AvarietyofSchiff'sbase anditscomplexeshavebeenstudiedextensively.Severalmodelsystems, including those with bidentate, tridentate, tetradentate, multidentate Schiffbase ligands, and their coordination chemistry of copper attracts much attention because of its biological relevance and its own interesting coordination chemistry such as geometry, flexible redox property, and oxidation state.[4]

Gou and coworker used Salicylaldehyde base-Schiff as novel, easily available colorimetric and fluorescent double-sensor. The sensor exhibits highly selective and sensitive recognition toward Cu^{+2} in aqueous solution via a naked eye color change from colorless to yellow and toward Al^{+3} via a significant fluorescent enhancement in ethanol over a wide range of tested metal ions. This represents the first reported Salicylaldehyde Schiff-based sensor capable of detecting Cu^{+2} and Al^{+3} using two different modes[10].

DISCUSSION

Bidentate Schiff Base Metal complexes

New Schiff bases of salicylaldehyde and their Cobalt(II) derivatives Scheme (1) have been prepared and tested for their antitumor activities. Several of these, particularly the cobalt derivatives, have shown significant Inhibitory action against mouse cancers[11].



Scheme 1: Proposed structure of metal complexes

The complexes of Mn(II) and Ni(II) with Schiff base derived from salicylaldehyde and 2-amino benzoic acid have been prepared and characterized as a neutral complex with 1:1 metal to ligand ratio[12]Figure (1).

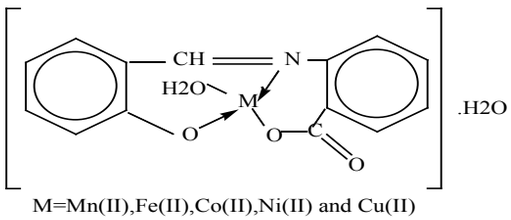
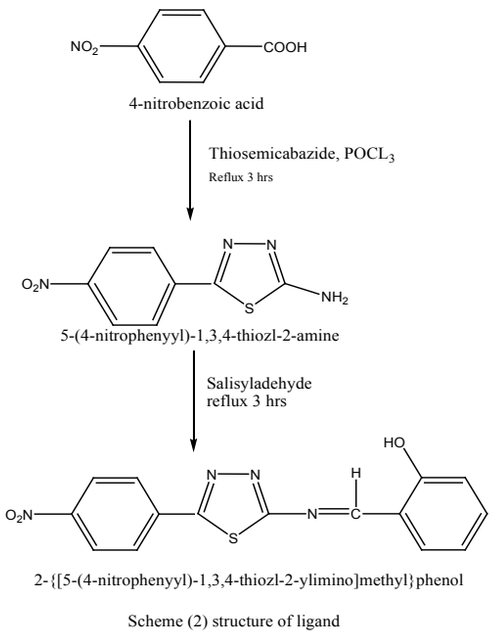


Figure 1: General proposed molecular structure of complexes

Yousif at el prepared and characterized a new metal complex derivatives of 2N-salicylidene-5-(*p*-nitro phenyl)-1,3,4-thiadiazole, HL Scheme (2) with the metal ions Vo(II), Co(II), Rh(III), Pd(II) and Au(III). The complexes obtained are monomeric with square planar geometry except VO(II) and Co complexes which existed as a square pyramidal and tetrahedral geometry respectively. The preliminary *in vitro* antibacterial screening activity revealed that complexes 1–5 Figure (2) showed moderate activity against tested bacterial strains and slightly higher compared to the ligand, HL[13].



Scheme (2) structure of ligand

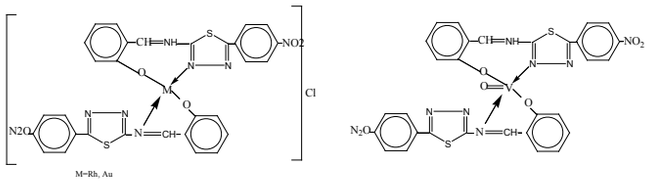
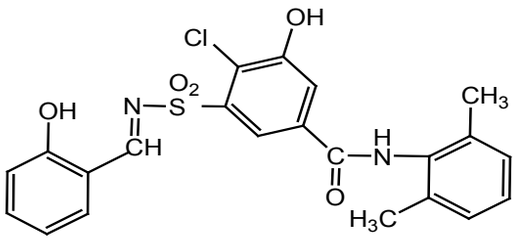


Figure 2: The structure of HL and the proposed structure of complexes 1-5, M=Metal

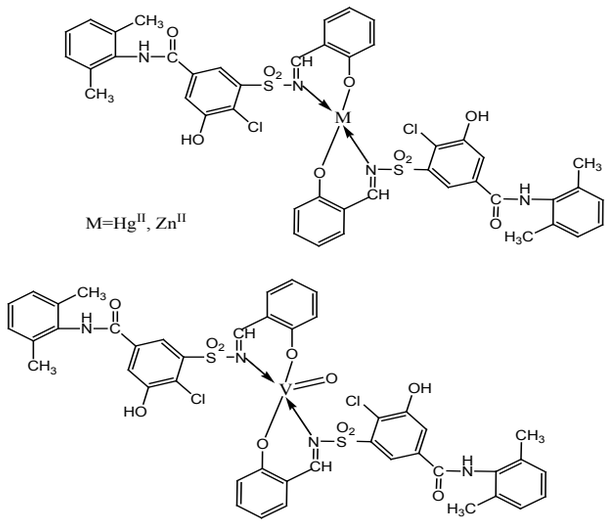
Malik and coworker reported the synthesis and characterization of metal complexes of Schiff base derived from xipamide, a diuretic drug. The bidentate ligand is derived from the inserted condensation of 5-aminosulfonyl-4-chloro-N-2,6-dimethylphenyl-2-hydroxybenzamide

(Xipamide) with salicylaldehyde in a 1:1 molar ratio Figure (3). Using this bidentate ligand, complexes of Hg(II), Zn(II), and VO(IV) with general formula ML_2have been synthesized Figure (4). All the complexes are nonelectrolytic in nature with 1:2 [M:L] ratio. Complexes of Hg(II) and Zn(II) have tetrahedral geometry via deprotonated phenolic oxygen and azomethine nitrogen atoms and VO(IV) complex have square pyramidal geometry. The pure drug, synthesized ligand, and metal complexes were screened for their antifungal activities against *Aspergillus niger* and *Aspergillus flavus*. The ligand and its Hg(II) and VO(IV) complexes were screened for their diuretic activity too.The complexes are found to have higher biological activities as compared to the respective ligand and the parent drug[14].



Xipamide salicylalde

Figure 3: Structure of ligand



Structure of VOL_2

Figure 4: Structure of metal complexes

New complexes of Schiff base ligand (derived from cefotaxime with salicylaldehyde with transition metals) prepared Figure (5) and characterized as nonelectrolyte complexes $[ML_2(H_2O)_2]$ with an octahedral geometry for Co(II), Ni(II), and Zn(II) complexes while a tetragonal geometry for Cu(II) complex Figure (6). All complexes were tested for *invitro* antibacterial activity against some pathogenic bacterial strains, namely *Escherichia coli, Klebsiella pneumoniae, Pseudomonasaeruginosa, Bacillus subtilis,* and *Staphylococcus aureus*. The results show that the metal complexes possess superior antibacterial activity than the Schiff base which could be used for the development of novel antimicrobial materials[15].

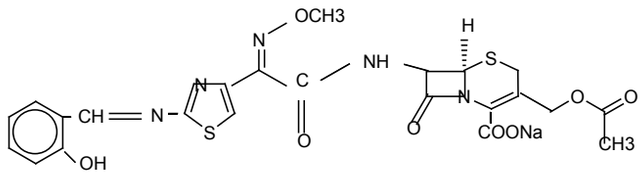


Figure 5: Chemical structure of the schiffbase

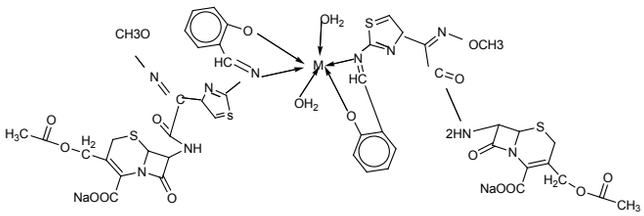


Figure 6: Proposed structure of metal (II) complexes

Silicon(IV) complexes containing mixed ligands: Shiff-bases (AH) derived from 2- or 3- amino-pyridine with 2-hydroxy- or 3-methoxy- or 2-hydroxy-3-methoxy-benzaldehyde and benzaldehyde semicarbazone (BSCH), have been prepared. Benzaldehyde semicarbazone acts as bidentate chelating ligand, octahedral Complexes of the type $[Si(BSCH)_2(AH)]Cl_4$, $[Si(BSC)_2(A_nH)]Cl_2$ and $[Si(BSC)_2(A)]Cl$ (where n=2 or 3, A=deprotonated Schiff-base ligands , BSC= dep- rotonated semicarbazone) have been proposed in neutral and basic medium, respectively[16]Figure (7).

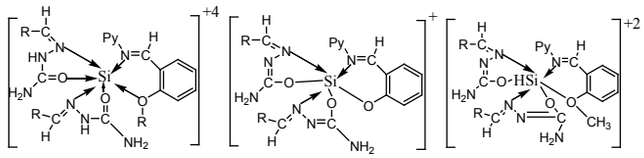
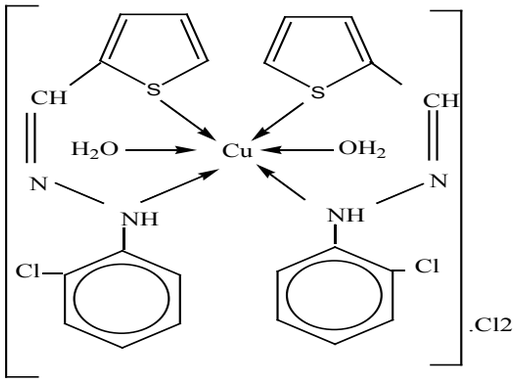
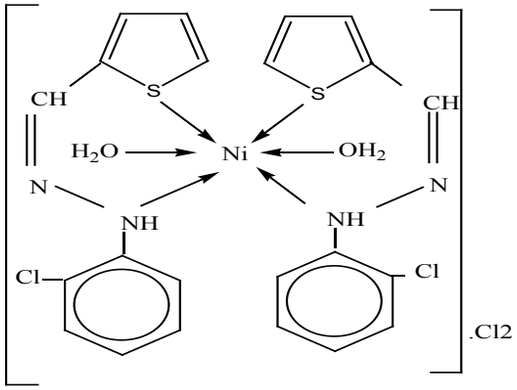


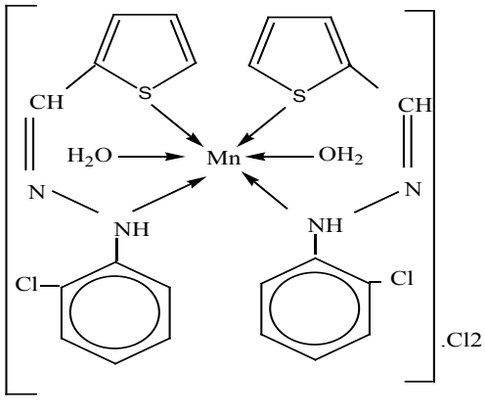
Figure 7: Proposed structure of the complexes

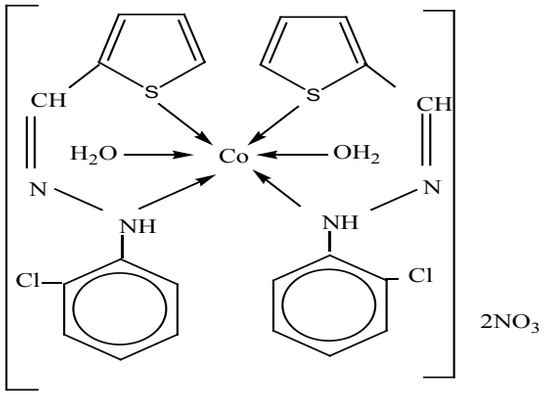
Figure 7: Proposed structure of the complexes

Metal chelates, $[M(HL)_2(H_2O)_2]X_2$ (where M= Mn(II), Co(II), Cu(II), Ni(II) or Zn(II), X= NO_3–or Cl– and HL= Schiff base moiety), have been prepared and characterized as 1:2 (metal-ligand). The coordination to the central metal atom have been through the nitrogen of the 2-chlorophenyl hydrazine (–Ph–NH-) group and the sulfur atom of the thiophene ring forming distorted octahedral complexes Figure (8). The Schiff base and its metal chelates have been screened for their in vitro antibacterial activity against four bacteria, gram-positive (*Staphylococcus aureus*) and gram-negative (*Escherichia coli*) and two strains of fungus (*Aspergillus flavus* and *Candida albicans*). The metal chelates were shown to possess more antibacterial activity than the free Schiff-base chelate [17].









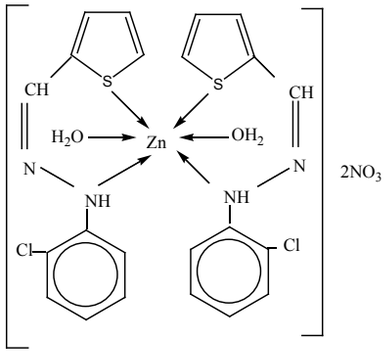


Figure 8: Proposed structure {M(HL)2(H2O)}X2 (where M= Mn(II), Ni(II), Cu(II) or Zn(II);X=(NO3 or Cl) schiff base complex

Robin R. and coworkers prepared and characterized Schiff bases complexes derived from sulfanilamides or aminobenzothiazoles Figure (9) with $Pd(OAc)_2$ as complexes of the type PdL_2Scheme (3,4). Palladium complexes and Schiff bases have been investigated as antifungal agents against *Aspergillus niger* and *Aspergillus flavus*[18].

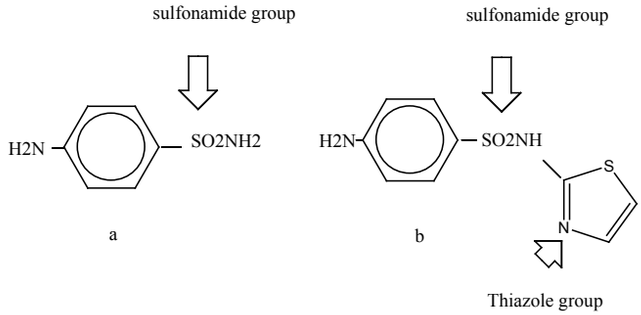
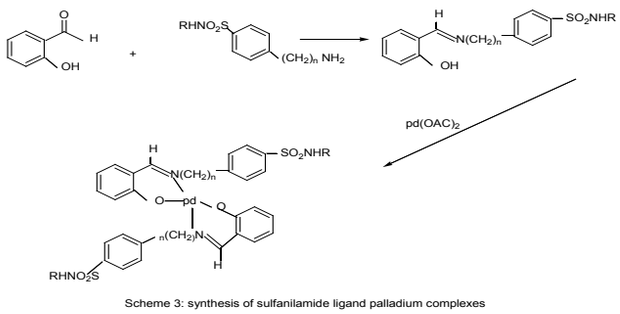
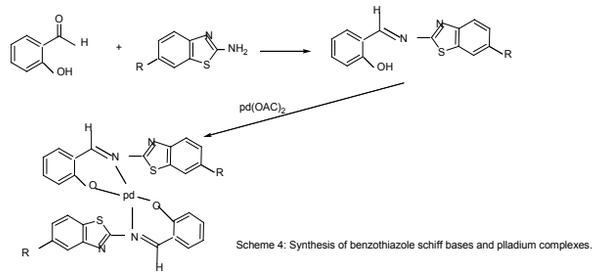


Figure 9: (a) sulfamilamide and (b) sulfathaizole



Scheme 3: synthesis of sulfanilamide ligand palladium complexes



Scheme 4: Synthesis of benzothiazole schiff bases and plladium complexes.

Cobalt(II), nickel(II) and copper(II) with Schiff bases derived from (2-aminobenzothiazole,6-nitro-2aminobenzothiazole,4,6-dibromo-2-aminobenzothiazole) and 4-N dimethyl bezaldehyde to give ligands (La, Lan and Ladb) were prepared in 1:2 ratio (metal: ligand) Figure (10). These measurements indicated that the ligands coordinate with metal(II) ion in a bidentate manner through the nitrogen atoms in ligands with general formula [ML2Cl2] Where M=Co (II), Ni (II) and Cu (II), (L=La, Lan, or Ladb) Figure (11)., Octahedral structures were suggested for metal complexes[19].

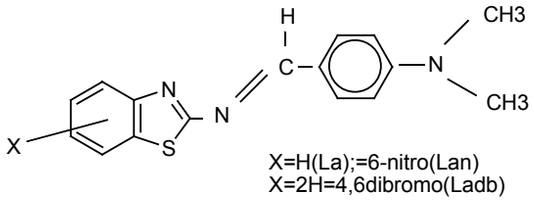


Figure 10: The structure of ligands

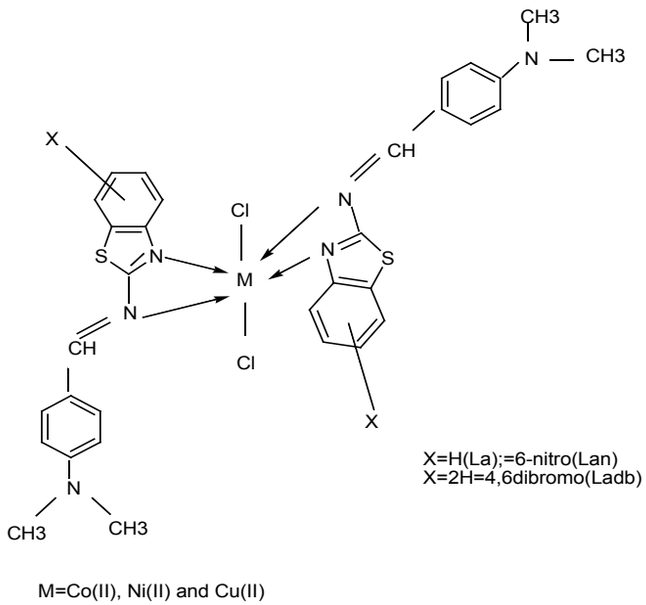
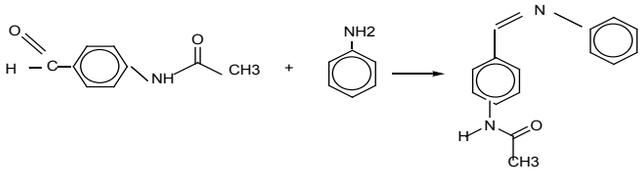


Figure 11: The suggested structure for the complexes

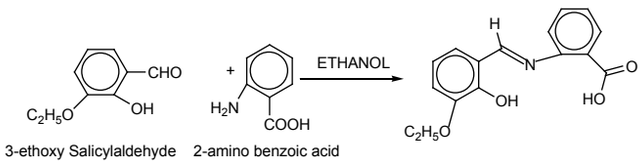
T. Mahmud reported preparation of Schiff bases (L-Lysine-Salicyaldehyde, DL-2,3-Diaminopropion-Salicyaldehyde and 4-acetylamido benzylidene aniline) in basic media (using 2M NaOH) Scheme (5). A copper(II) complex with 2,2′-bipyridine and L-lysine was synthesized and characterized as a monomer with distorted square planar geometry, or a distorted octahedron if the long axial coordination from the perchlorate ions is admitted. The asymmetric unit contains two formula units of [Cu (L-Lysine) (2,2′-bipyridine)] with two perchlorate ions and one water molecule[5].



Scheme 5: Preparation of 4-acetoamidobenzylideneaniline schiff base

Tridentate Schiff Base Metal complexes

Mounika and coworkers reported a new Schiff base, 3-ethoxy salicylidene amino benzoic acid (ETSAN) Scheme (6), has been synthesized from 3-ethoxy salicylaldehyde and 2-amino benzoic acid. The ligand act as neutral and tridentate coordinating through nitrogen atom of the azomethine and oxygen atoms of hydroxyl group of the 3-ethoxy salicylaldehyde beside the hydroxyl group of the carboxyl group of the 2-amino benzoic acid respectively. All complexes are non-electrolytes and show 1:1 metal: ligand ratio with octahedral geometry Figure (12). Biological studies of these complexes reveal that they show better activity when compared to that of the ligand[20].



Scheme 6: Preparation of Ligand 3-ethoxy salicylidene amino benzoic acid

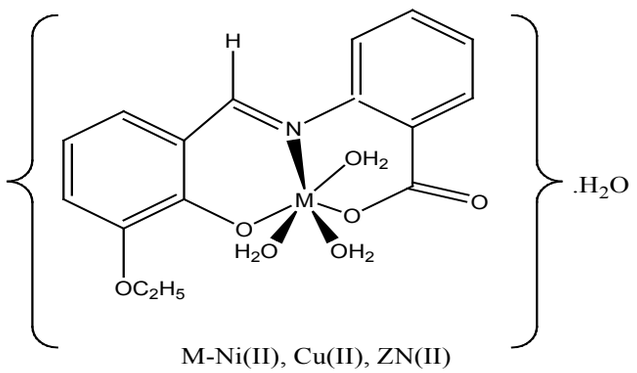
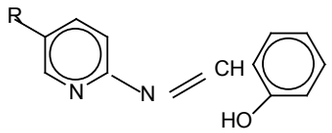


Figure 12: Proposed structure of metal complexes

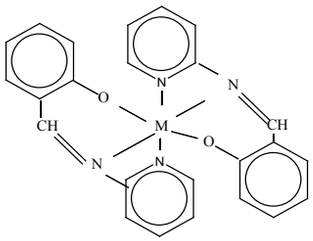
Figure 12: Proposed structure of metal complexes

Novel transition metal [Co(II), Cu(II), Ni(II) and Zn(II)] complexes of substituted pyridine Schiff-bases Figure (13) have been prepared and characterized with general formula [M(L)2] where [M=Co(II), Cu(II), Ni(II) and Zn(II) and HL=HL[1], HL[2], HL[3]and HL[4]] and an octahedral geometry. The synthesized Schiff-bases act as deprotonated tridentate with Co(II), Ni(II) and Zn(II) ions Figure (14). The Schiff bases and their complexes have been screened for antibacterial activity against the strains such as *Escherichia coli, Staphylococcus aureus,* and *Pseudomonas aeruginosa.* The complexed Schiff bases have shown to be more antibacterial against one more bacterial species as compared to uncomplexed Schiff-bases[21].



HL1:R=OH
HL2:R=NO2
HL3:R=OCH3
HL4:R=Br

Figure 13: Structure of schiff base



R=OH HL1
R=NO2 HL2
R=OCH3 HL3
R=Br HL4
M= Co(II), Ni(II), Cu(II) or Zn(II)

Figure 14: Proposed structure of the Metal(II) Complexes

Z. F. Dawood and M. W. Ibrahim reported Preparation and characterization of new cobalt (II) complexes with mixed ligands including salicylaldehyde thiosemicarbazone-STH_2 and carboxylic acid-AH_2 {salicylic acid-SH_2 or anthranilic acid-AnH_2 or phthalic acid-PH_2} Figure (15) as $[Co_2(AH_2)_2(STH_2)_2(NO_3)_2](NO_3)_2$ or $[Co_2(AH_2)_2(STH_2)_2Xn]$ in neutral medium whereas, in basic medium as $[Co_2(AH)_2(STH)_2]$ {where X = CO_3^{2-} or CH_3CO^{2-} , n = 2 or 4} with octahedral geometries forming dimer (binuclear complexes)[22]Figure (16).

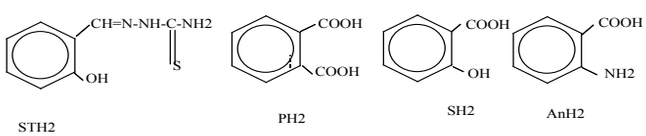


Figure 15: Structures of ligands

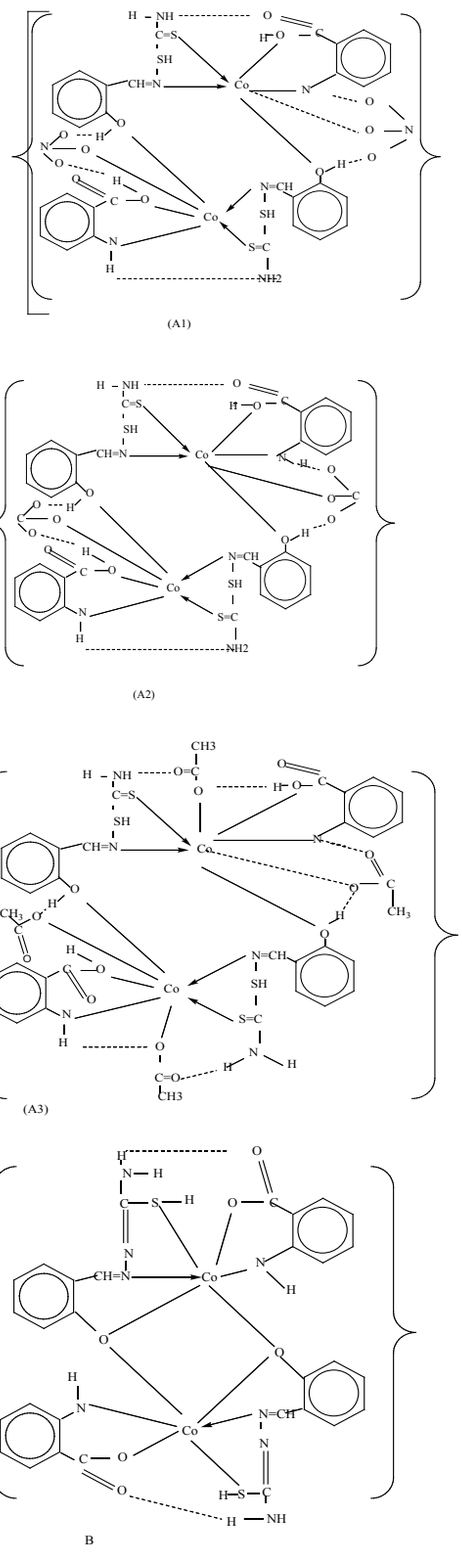
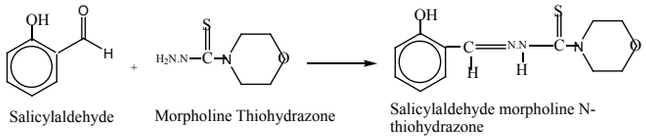


Figure 16: Proposed structures of the Complexes(A1,A2,A3) in neutral medium,(B) Complexes in basic medium

Saxena synthesized and characterized metal complexes of Ti (III), V (III), VO (IV), CO (II) and MN (III) with salicylaldehyde and thiohydrazones ligand Scheme (7) and proposed an octahedral geometry for all the synthesized complexes[23]Figure (17).



Scheme 7: Preparation of Salicylaldehyde morpholine N-thiohydrazone

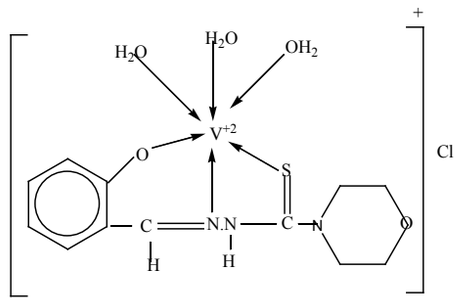
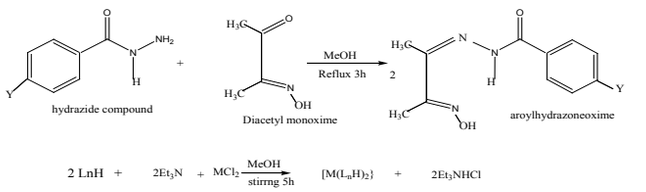


Figure 17: Salicylaldehyde morpholine N-thiohydrazone Vanadium (III) Chloride

Mohammed M. Al-Ne'aimi *at el* have been prepared Mononuclear complexes of two novel ligands {Diacetylmonoxime-4- nitrobenzoylhydrazone (L_1H_2), Diacetylmonoxime-4- hydroxybenzoylhydrazone (L_2H_2)} Scheme (8) with [Co(II), Ni(II), Cu(II), Zn(II) and Cd(II)] as $[M(L_nH)_2]$ where (n =1,2) in the presence of Et_3N. The metal complexes $[M(L_nH)_2]$ are proposed to be six-coordinated with a N_4O_2 donor environment through the oxime nitrogen, the imine nitrogen and the enolic oxygen atoms while the phenolic hydroxyl and oxime hydroxyl groups of aroylhydrazonemonoxime moiety do not participate in coordination[24].



Scheme 8: Preparation of ligands and metal complexes

Dinuclear complexes from salicylaldehyde and 2-aminophenol with Cu (II), Ni (II) and Co (II) Figure (18) were obtained by new synthetic route and characterized. Low temperature intramolecular ferromagnetism was exhibited by homodinuclear complex while the heterodinuclear complexes showed antiferromagnetic coupling[25].

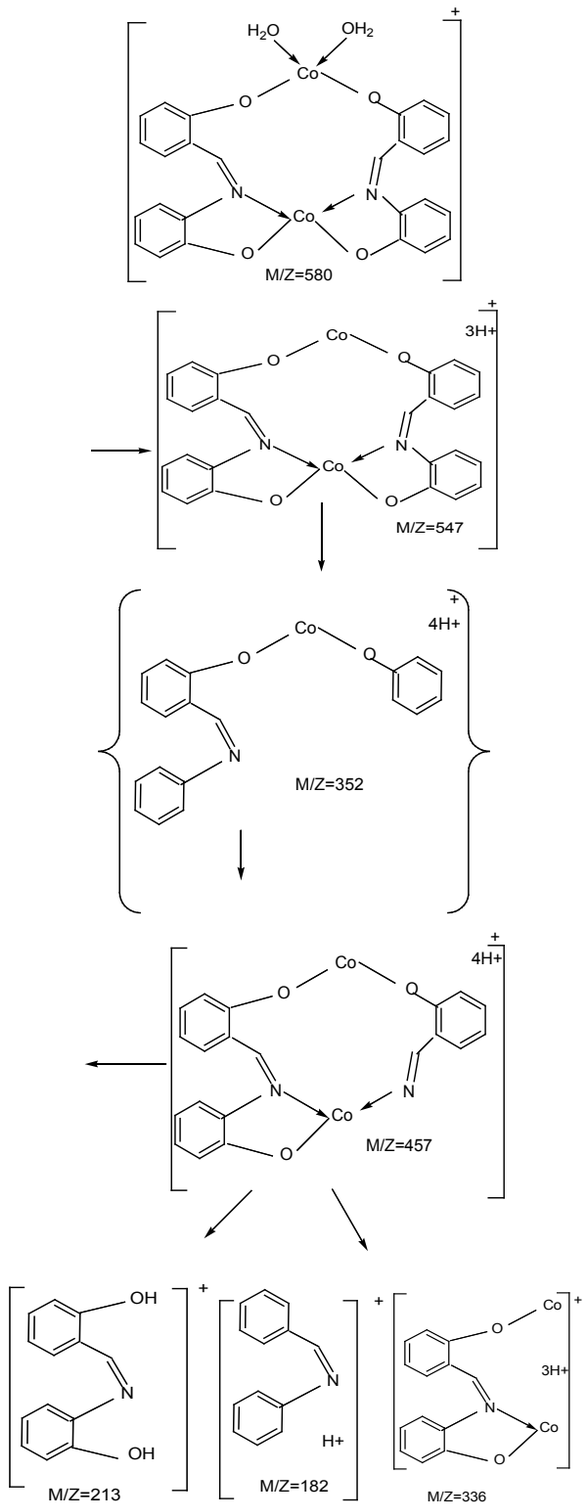
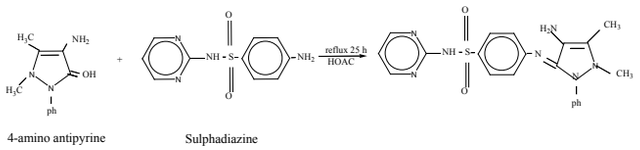


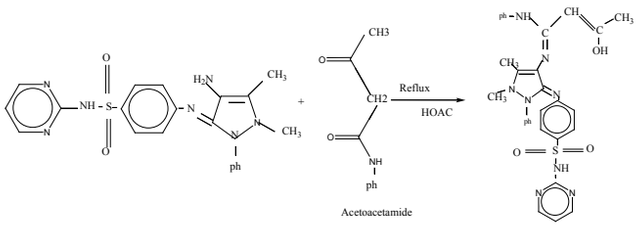
Figure 18: The Metal Complex

New Schiff base ligand derived from 4-Amino antipyrine, sulphadiazine and acetoacetanilide were prepared Scheme (9) and reacted with metal salts in 1:1 ratio (metal: ligand). The complexes have the general formula $[MLCl.2H_2O]$ where M = Co(II), Ni(II) and Cu(II) Figure

(19). The ligand coordinate with the metal (II) ion in neutral tridentate manner through the azomethane nitrogen atoms, and oxygen group of the acetoacetanilide forming octahedral complexes[26].

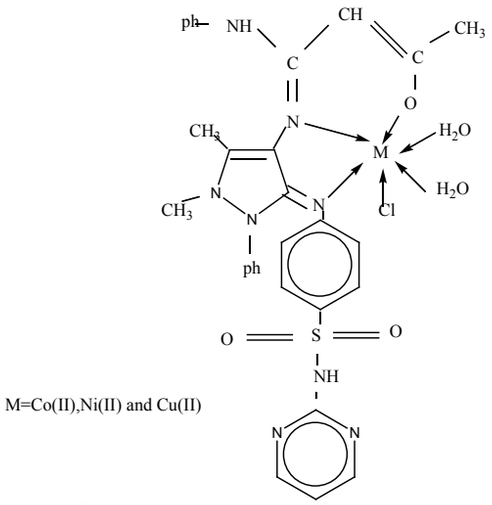


4-amino antipyrine Sulphadiazine



Acetoacetamide

Scheme 9: Preparation of the Ligand



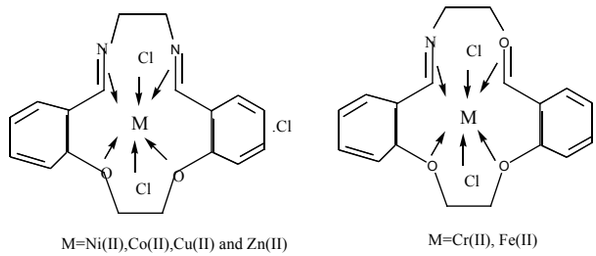
M=Co(II),Ni(II) and Cu(II)

Figure 19:The proposed structural formula of the metal chelate complexes

Figure 19: The proposed structural formula of the metal chelate complexes

Tetra dentate Schiff Base Metal complexes

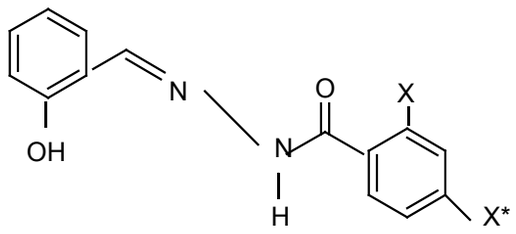
Mostafa M. and coworker synthesized macro-cyclic Schiff base ligand resulted from thecondensation of {bis-aldehyde and ethylenediamine was prepared (7, 8, 15, 16, 17, 18-hexahydrodibenzo (a, g) (14) annulene)} (L) and its complexes were synthesized and characterized as 1:1 [ML] complex with octahedral structure for the all complexes via (N_2O_2) group act as a tetradentate ligand and two chlorides as monodentate ligands Figure (20). They reported using of neutral chelating ligand as selective reagent to determine iron (III) in different types of natural water within recovery test and described that the metal complexes are more potent/ antibacterial than the parent Schiff base ligand against one or more bacterial species[27].



M=Ni(II),Co(II),Cu(II) and Zn(II) M=Cr(II), Fe(II)

Figure 20: Structural formula of metal complexes

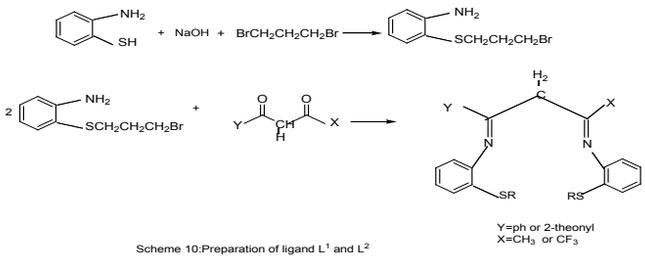
Salicylaldehyde 2-chlorobenzoyl hydrazone (H_2LASS-Bio-466), salicylaldehyde 4-chlorobenzoyl hydrazone (H_2LASSBio-1064) Figure (21) and their complexes $[Zn(LASSBio466)H_2O]_2$ and [Zn(HLASSBio-1064)Cl] were evaluated in animal models of peripheral and central nociception, and acute inflammation. All studied compounds significantly inhibited acetic acid-induced writhing response. All compounds showed levels of inhibition of zymosan-induced peritonitis comparable or superior to indomethacin, indicating an expressive anti-inflammatory profile[28].



H2LASSBIO-466; X=Cl and X*=H
H2LASSBIO-1064;X=H and X*=Cl

Figure 21: The structure of the Ligand

Two new tetra dentate ligands (L1 and L2) N,N'-bis{(o-aminophenylthio)bromo propyl} 1-phenyl butane 1,3-dilidene (L1), N, N'-bis (3- bromo propyl (phenylthio) imino)1,1,1-trifluoro -3-(2-theonyl) acetone (L2) were prepared Scheme (10) and reacted with M= Co(II), Ni(II), Cu(II) and Zn(II) chloride salts to give $[M(L)]Cl_2$ and $[Zn(L)Cl_2]$ Figure (22) as tetrahedral complexes via nitrogen atoms of the azomthine and two sulfur atoms while for zinc (II) complexes from the two sulfur atoms and two chloride ions[29].



Y=ph or 2-theonyl
X=CH_3 or CF_3

Scheme 10:Preparation of ligand L^1 and L^2

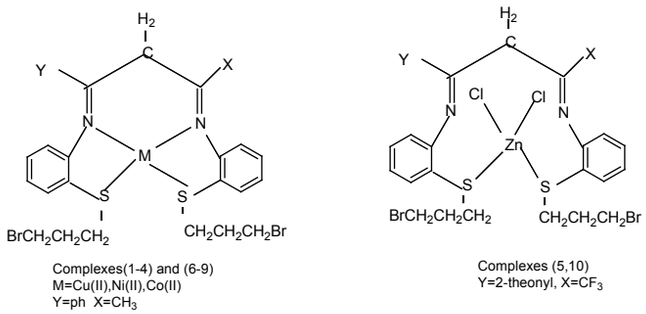


Figure 22: Suggested structures for prepared complexes

New Schiff base chelates of Cu(II), Co(II), Ni(II) and Zn(II) derived from benzil-2,4-dinitrophenylhydrazone with aniline have been synthesized Figure (23) and characterized to suggest tentative structures for the complexes[30].

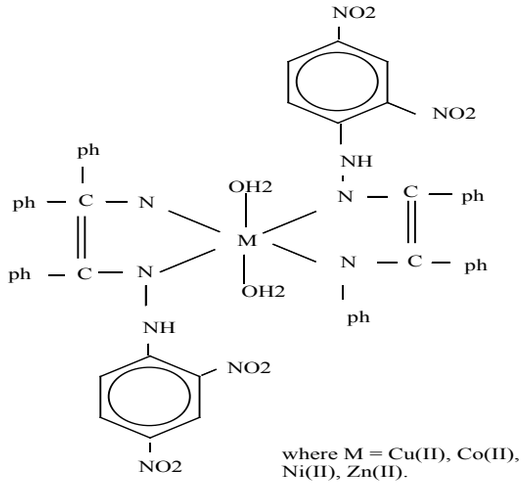


Figure 23: Proposed structure for the complexes

Three new metal complexes of Cr(III), Pb(II)) and TiO(IV) ions with a Schiff base derived from salicylaldehyde and urea Figure (24) have been investigated with 1:1 [M:L] ratio. The coordination behavior of the metal ions towards to the investigated Schiff base takes place through –C=N and –OH groups[31] Figure (25).

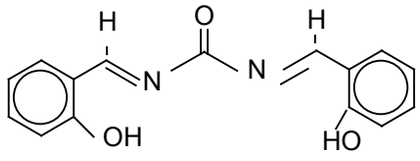


Figure 24: Ligand

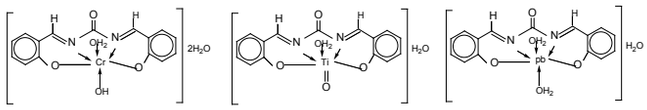


Figure 25: Proposed structures of Metal Complexs

Tetra dentate N_2O_2 type complexes of Co(II) have been synthesized by the condensation of o-phenylenediamine, salicylaldehyde and isatin/ naphthaldehyde/acetyl acetone and characterized as $[ML(H_2O)(OAc)]$ with octahedral geometry Figure (26). The metal complexes have been screened for their antibacterial and antifungal activity. DNA cleavage activities of Schiff bases and their metal complexes were monitored by agarose gel electrophoresis method in the presence of H_2O_2[32].

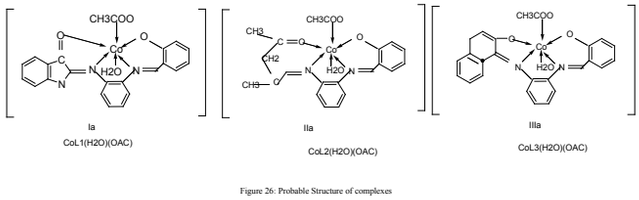


Figure 26: Probable Structure of complexes

Figure 26: Probable structure of complexes

N. Kumar at el reported preparation of Schiff bases (Salicyladehyde glycine, DL-2,3-Diaminopropion-Salicyaldehyde, benzylidene glycine and 4-acetylamido benzylidene aniline) in basic media (using 2M NaOH) figure (27) and described the applications of shiff bases and their copper(II) complexes as antimicrobial activities, antifungal activities, antiviral activities. Tetra dentate Schiff base and its metal complexes with Mn (II), Ni (II), Cu (II), and Zn (II) show miscellaneous effect on membrane in amylose production[33].

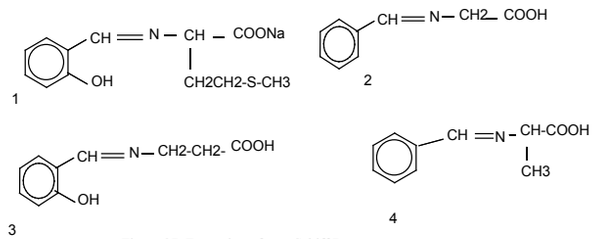


Figure 27: Formation of new Schiff Bases

CONCLUSION

Salicyaldehyde and their derivatives can be good chelating ligands when they condensed with amines in 1:1 and 2:1 ratio to form bi, tri and tetra dentate Schiff base ligands suitable to form complexes with Metal ions. These Metal-Schiff base complexes have been shown to exhibit a broad range of biological activities, including antifungal, antibacterial, antimalarial, anti-proliferative, anti-inflammatory, antiviral, and antipyretic properties.

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