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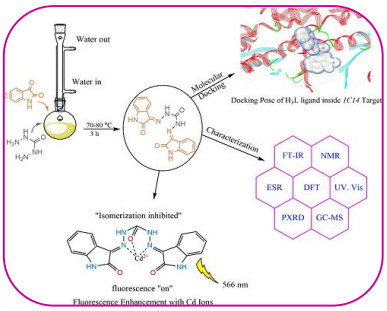
STUDIES OF SCHIFF BASE LIGANDS, ITS COMPLEXES WITH MERCURY (II) AND THEIR APPLICATIONS- A REVIEW

Vijay Kumar Chaudhary[1] , Anil Kumar[2] and D.K.Agarwal[2]
[1]Department of Chemistry, D.V.College,Orai, Jalaun, (U.P.)
[2]Department of Chemistry, Bipin Bihari College, Jhansi, (U.P.)

ABSTRACT:



A variety of Schiff bases have been investigated. Mercury(II) with Schiff base ligands formed as supramolecular frameworks or multifunctional coordination polymers coordination geometry. Mercury (II) are also formed asymmetrical as well as nitrogen, sulpher and oxygen as donor Schiff-base complexes. Sometime mercury (II) are formed supramolecular complexes by non covalent linkages. The Schiff-base complexes are ability as biologically active substances, dyes, liquid crystals, luminophores and polymer stabilizers. The applications these Schiff-base complexes such as antimicrobial, antitumor, antidepressants, nematocide, antiphlogogistic and other medicinal significants. Another applications in the paper industry and as preservative, fluorescent lamps, sensors and batteries.The Schiff-base are used to simultaneous detection and removal of mercury (II) ions from water.

KEYWORDS- *Schiff base, supramolecular complexes, mercury(II), detection and removal.*

INTRODUCTION

The heavy and transition metal ions are very attractive because of their extremely toxic effect on the environment and biological systems. Among of these, mercury one of the most poisonous and toxic pollutants that are serious to health problems. The mercury and its salts find wide applications in industrial processes and products as such paints, electrical equipments and batteries. The mercury has different forms in environment such as Hg^0, Hg^{2+}, and CH_3Hg^+. The mercuric ion is neurotoxin as methyl mercury. The methyl mercury easily enter the environment and food chain of aquatic organisms and environment with mining, transportation and processes of mercury ores, dumping of industrial wastes into rivers and lakes, combustion of fossil fuels, pulp and paper industry, wood pulping, oil refining, solid waste burning, coal and gold mining, rubber processing, fertilizer industries, the use of mercury compounds as seed dressings in agriculture.[1,2,3] The inorganic mercury may be change into organic mercury like methyl mercury, which are more toxic than inorganic mercury. Methyl mercury triggers several serious problems for humans such as allergic reactions, brain and neurological damage. For determination and identification of mercury compounds are generally used optical chemical sensors (optodes). Essential steps in fabrication of optical sensor is immobilization of ionophores on transparent membranes. The ionophore immobilized are physical entrapment, sol-gel, multilayered films or chemical bonding methods[4.]

The combination of inorganic mercury with natural component in soils, sediments or water is forming the complexes,specially amino or sulfide groups, which are change the toxicity of respective ion[5].

The ability to Schiff bases and their complexes as biologically active substances, liquid crystals, dyes, polymers and luminophores and their applications in antimicrobial, antitumor, antidepressants, nematocide, antiphlogogistic, pharmaceuticals, optical communication and optical device.The Schiff base complexes are also the ability to reversibly bind oxygen in epoxidation reactions, biological properties, photochromic properties and catalytic role in hydrogenations. The complexes of stable d^{10} electronic configuration have a lot of attention in the field of inorganic chemistry, biochemistry and environmental chemistry[6].The lability of d^{10} metal complexes, the coordination bonds is reversible, metal ions and ligands to rearrange during supramolecular assembly is formed thermodynamically most stable structure. In this ways mercury(II) can be coordinated different type of frameworks, which may be using a different type of organic/inorganic ligands[7]. The schiff base are also "salen-type" ligands, its formed mononuclear as well as multinuclear complexes with nitrogen and oxygen atoms as donor. Mercury was formed several adduct metal chelates. Mercury(II) have been reported various complexes of ligand species, which are dinuclear, trinuclear, tetranuclear and polynuclear, sometime its also formed mix metals complexes[8].

Mercury(II) and its compounds are very important in chemistry and its related disciplines. Mercury(II) compounds are diversity in coordination geometries and frameworks. Its formed halides and pseudohalide complexes with varied structures and interesting molecular properties using multidentate N-donor Schiff bases[3]. The pseudohalides are developed variety of bridging unit, which attached with organic blockers may affords different monomeric, dimeric, polymeric and suprameric metal-organic frameworks. The other pseudohalides like azide and thiocyanate in combination with N-donor Schiff bases at different denticities[9].

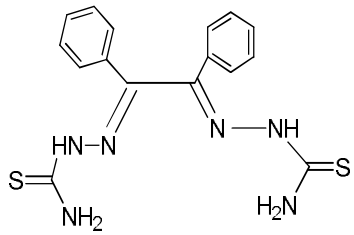
The polymer chemistry also studies of inorganic-organic hybrid coordination compounds recent few years, which have attractive flexibility in building up comprehensive structures. The mercury(II) have reported strongly asymmetric Hg-Halide-Hg bridges coordination polymers. Organic aromatic ligands are exhibits interesting self-assembly of organic-inorganic materials. It is not only act as hydrogen bonding acceptors or donors, but also represented in $\pi - \pi$ stacking interactions to formed supramolecular structures with metals[10].

SCHIFF BASE AND THEIR COMPOUNDS WITH MERCURY:

We knew that mercury is very toxic element to environment. The detection and identification of inorganic and organic mercury species from high performance liquid chromatography or gas chromatography coupled with adequate detection systems are used.To determination of different complex species of the metals in environmental samples like natural waters is analyzed by electro analytical methods. For mercurydetermination here a method developed with a chemical modified electrode (CME) based on the selective response of mercury species in voltammetric measurement process. The macroligands thiosemicarbazone, benzylbisthiosemicarbazone (BTC) has ability to used as modifier of a carbonpaste electrode to perform metal determination. This ligand react with mercury(II) ions. The ligand is insoluble in water, but it is light soluble in alcohols and mineral oil and high soluble in dimethylsulfoxide and N,N′-dimethylformamide[5].

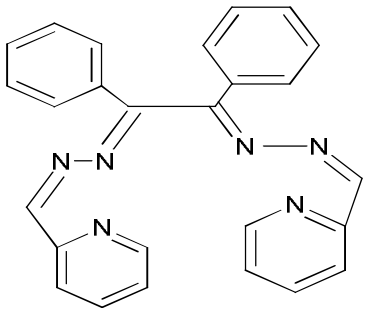
The benzylbisthiosemicarbazone (BTC) was synthesized from benzyl and thiosemicarbazide[11,12].

FIG :1

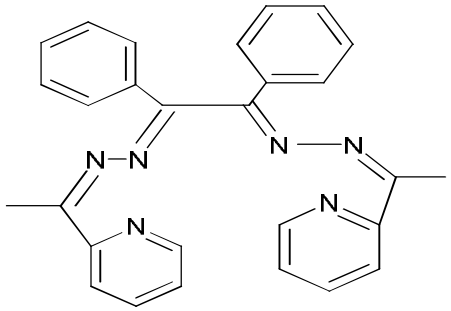


STRUCTURE OF BENZYLBISTHIOSEMICARBAZONE(BTC)

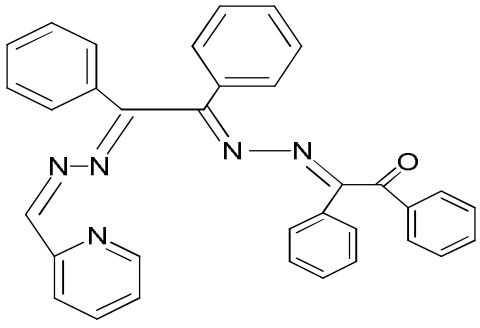
The mercury(II) is easily reacted with a bendy coordination environment, thus these complexes are can be vary from linear to octahedral geometries or even severe distorted polyhedral coordination geometries. Schiff base ligands are significant ability to formed stable complexes with metal ions and formation of coordination polymers with fascinating geometries[13,14]. The azine-based Schiff base ligands (L_1,L_2,L_3) [Fig:2] reaction with HgX_2 (X= Cl, Br, I) in 1:10 molar ratio, formation of solid state architecture of series of Hg(II) containing coordination compounds, which are $[Hg(L_1)(\mu\text{-}Cl)_2Hg_3Cl_6]_n$,$([Hg(L_1)(\mu\text{-}Br)_2HgBr_2]$, $[Hg(L_3)Br_2]$, $[Hg(L_1)I_2]$, $[Hg(L_2)Cl_2].CH_3OH$, $[Hg(L_2)(\mu\text{-}Br)HgBr_3]_2$ .The ligands benzilbis((pyridine-2-yl)methylidenehydrazone) (L_1) benzilbis((acetylpyridin-2-yl)methylidenehydrazone) (L_2), (L_3) and their compounds with mercury(II) were prepared following a reported method described elsewhere[15].



L1



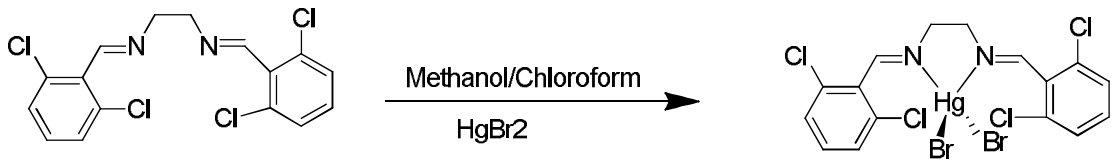
L2



L3

Fig:2- Molecular digrams of L1, L2 and L3.

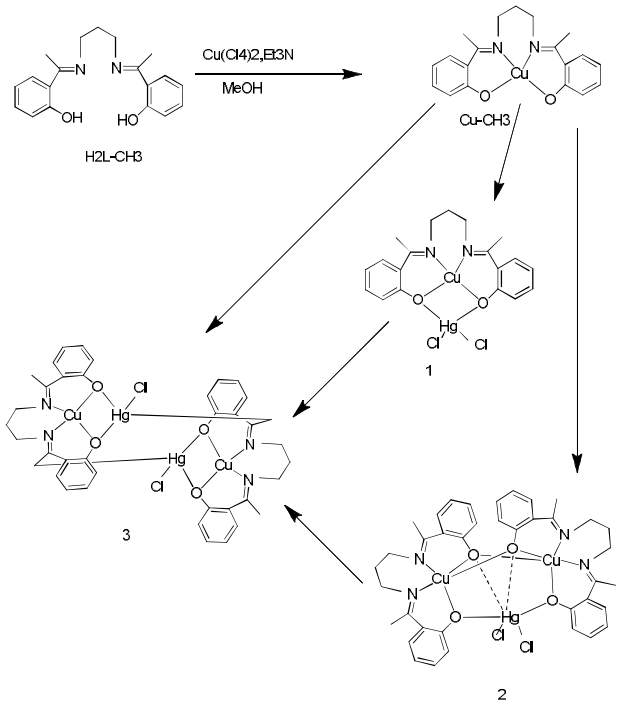
The mercury(II) formed symmetric bidentate Schiff base ligands crystal complexes. The bidentate Schiff base ligand (2,6-Cl-ba)$_2$en, having the systematic name [N,N′-bis(2,6-dichlobenzylidene)ethane-1,2-diamine] was prepared by the condensation of 2,6-dichlorobenzaldehyde with ethane-1,2-diamine following method described[16]. The bidentate Schiff base ligand mixed with $HgBr_2$ and formed micro crystals of complex [Hg{(2,6-Cl-ba)$_2$en}Br_2] by following method described[17].(Scheme-1).



Scheme 1. Chemical structures of Schiff base ligand and its mercury(II) complex.

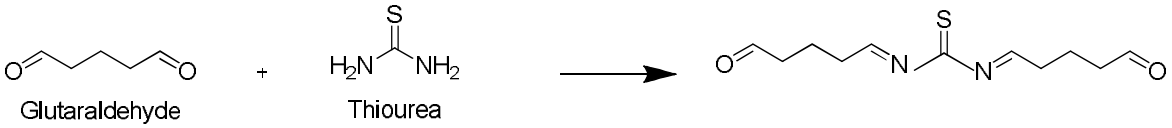
The dinegative anionic form of "salen types" of Schiff base acts as a tetradentate chelating ligand with two nitrogen and two oxygen donor atoms and form mononuclear complex[18]. This type of mononuclear complexes of such ligands with dipositive first row of transition elements can easily be used as a "complex as ligand"for the synthesis of heterometallic complexes in which the coordinating modes of these di-Schiff bases are penta- or hexadentate with the oxygen and nitrogen atoms as donor centers. Mercury is easily known to form an adduct with many metal chelates. The 14 heteronuclear complexes of Hg(II) have been reported with various "complex as ligand" species, which of them 10 are copper(II) chelates[19].

The di-Schiff base H_2L-CH_3=N,N'-bis(α-methylsalicylidene)-1,3-propanediamine was prepared with 1,3-propanediamine and 2-hydroxyacetophenone and their complexes by the reported methods[19,20]. The complexes are dinuclear [(CuL-CH_3)$HgCl_2$], (1), the trinuclear complex [(CuL-CH_3)$_2$$HgCl_2$] (2) and the tetranuclear cyclic organomercurial complex [(CuL-CH_2-HgCl)$_2$](3) are synthesized by the reported methods[8].(Scheme-2).



Scheme 2: Synthetic Route to Complexes 1-3

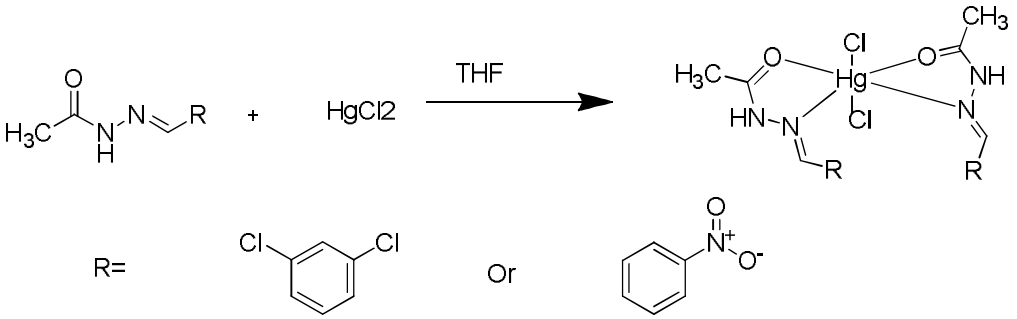
The magnetic resins is substance that may be used to removal of some metals from aqueous solutions. Chitosan is a major component of crustacean shells, which is natural biopolymers. Its ability to absorbed several metal ions by different mechanism and condition. The magnetic chitosan resin is prepared with chitosan and Schiff base by reported method[1].Schiff base prepared with glutaraldehyde and thiourea by reported method[21,22].(Scheme-3).



Scheme:3- Preparation of Schiff base

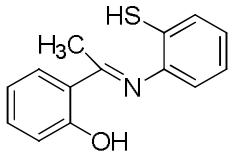
Mercury has formed stable complexes with ligands as mercuric and mercurous form, such as Se(II)[23,24] and Gold(I)[25] because mercury is behave as soft Lewis base[26]. The coordination chemistry of Hg(II) differ from other transition metals because its large size and d^{10} configuration. The field of bioinorganic chemistry of Schiff base complexes have play interested role in biologically important. The two Schiff base(L_4,L_5) synthesized with acetohydrazides R-CO-NH-NH_2 and 2,4-dichlorobenzylidene, 3-nitrobenzylidene respectively .(Figure: 3) These Schiff base have reacted with mercuric chloride to formed complexes by reported method[27].(Scheme-4)

Fig:3 Structure of Schiff base ligands L4 and L5.



Scheme:4- Schematic representation of metal complex formation.

Optical chemical sensors (optodes) are simple and selective tools for used to determination of heavy metal ions. It also used in combination with cheap spectrophotometric or spectroflorurometric techniques to provide simple and fast determination methods[28,29,30]. Optical sensors with ionophores play an important role. Schiff base ligands have been used to as ionophores in formation of membrane sensors due to ability of form stable complexes with heavy metal ions. Schiff base ligand 2-[(2-sulfanylphenyl)ethanimidoyl]phenol (L_6) is covalently toothless on an agarose membrane to used an as effective ionophore with N, S and O donor atoms for formation of a chemical optical sensor, which used as spectrophotometric identification of Hg(II) ions in aqueous solutions[4].



2-[(2-sulfanylphenyl)ethanimidoyl]phenol

L6

Fig: 4 Structure of Schiff bas ligand L_6.

The complexation of Hg(II) with L_6 in methanolic solution was determinate by spectrophotometric titration. Hg(II) has great affinity for sulfur coordination and this way various types of identification systems used in mercury optical sensors, which is based on sulfur containing ligands for better sensitivity and selectivity[31,32,33]. (Fig: 5)

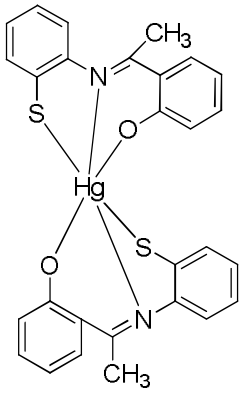
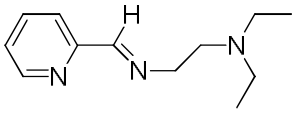
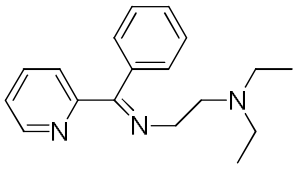


Fig: 5: Complexation of Hg(II) with L_6 in an octahedral geometry.

Another way Schiff bases are very important and interesting ligands, because its are easily prepare, structural varieties, varied denticities and slight electronic effects[34,9,35,36]. When halides are present as terminal or bridging units, its indicate molecule must be form different frameworks, crystalline networks and different non-covalent forces[37-42]. For example two tridentate Schiff bases (N,N-diethyl,N'-(pyridine-2-yl)formyllidene)ethane-1,2-diamine (L_7) and (N,N-diethyl,N'-(pyridine-2-yl)-benzylidene)ethane-1,2-diamine (L_8), (Fig: 6) which are capable to isolate HgX_2 compounds with different geometries and molecular properties. The Schiff bases L_7, L_8 and their complexes with $HgBr_2$were prepared following the reported methods described elsewhere[35,36].(Scheme: 5).

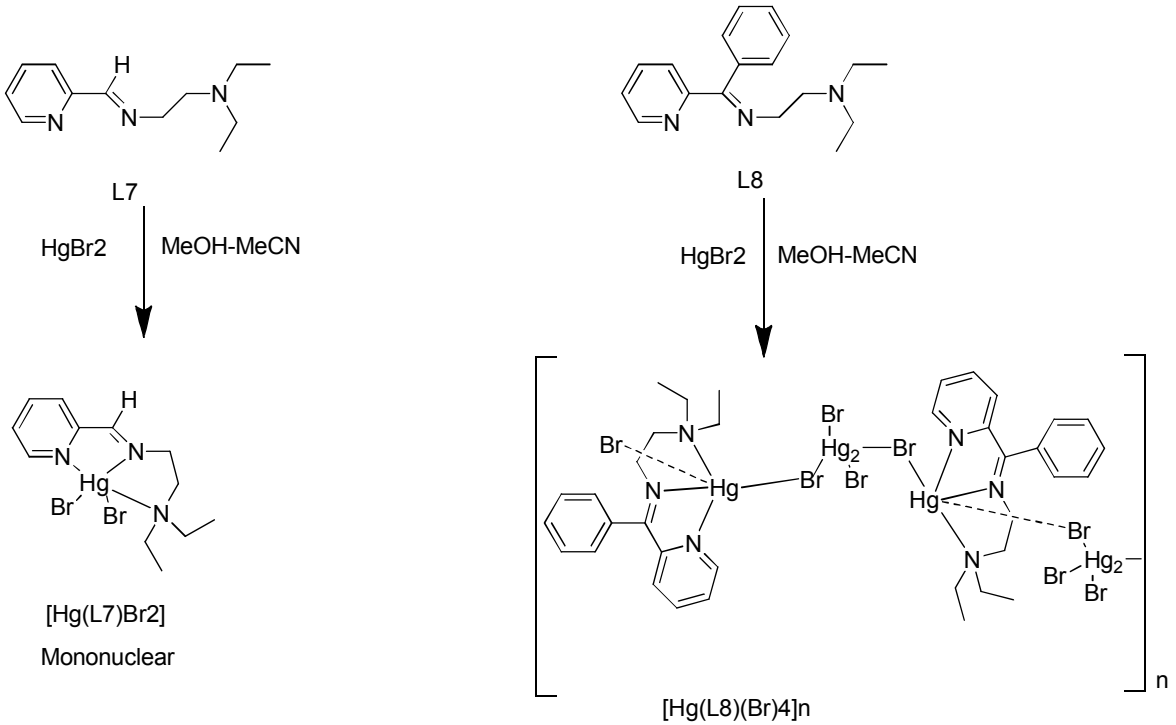


L7



L8

Fig: 6 The structure of Schiff base ligands L_7 and L_8



Scheme: 5- Different reaction behaviors of L_7 and L_8 in presence of $HgBr_2$.

The transition and non-transition metal ions are binding with Schiff bases of different denticities by pseudohalides like azide and thiocynate with N donor atoms, which are formed tetra and pentacoordinated complexes[43].Schiff base ligands are interesting their coordination behavior in tetradentate [N,N'-(bis-(pyridine-2-yl)benzylidene)-1,2-ethanediamine] (L_9) and a hexadentate [N-((1-pyridin-2-yl)formylidene)-N'-[2-(4-{2-[((1-pyridin-2- yl)formylidene)amino]ethyl}piperazin-1-yl)ethyl]amine] (L_{10}). (Fig:7) These ligands are binds with mercury(II) and give rise to different coordination numbers and molecular and crystalline aggregates, its show different coordination geometries[44,45,46].The crystallographically studies its charecterised a tetracoordinated dinuclear compound $[Hg(L_9)(SCN)_4]$ and two pentacoordinated dinuclear compounds $[Hg(L_{10})(N_3)_4]$ and $[Hg(L_{10})(SCN)_4]$. The Schiff bases L_9 and L_{10} were prepared following the reported methods[47,48]. The complexes $[Hg(L_9)(SCN)_4]$, $[Hg(L_{10})(N_3)_4]$ and $[Hg(L_{10})(N_3)_4]$ were prepared following the reported methods described elsewhere[9].

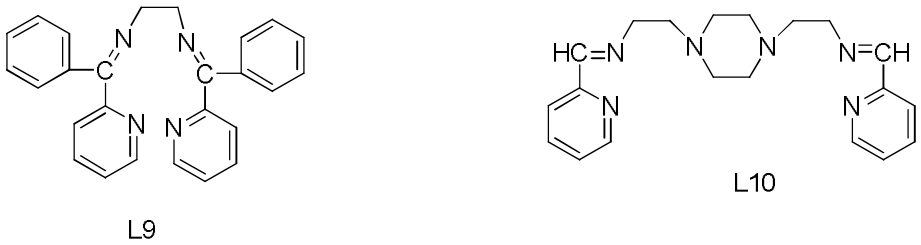
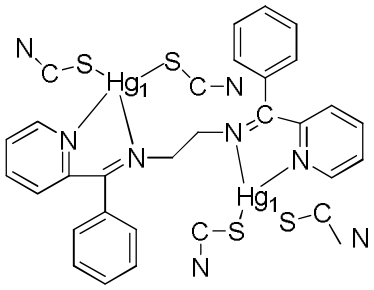


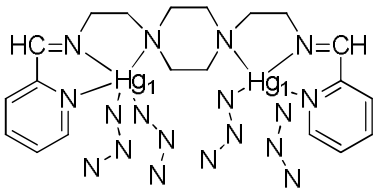
Fig: 7Structure of Schiff bases L_9 and L_{10}

$2Hg(II)\ salt + L_9/L_{10} + 4X^{-} \xrightarrow{-MeOH} [Hg_2(L_9/L_{10})(X)_4]$

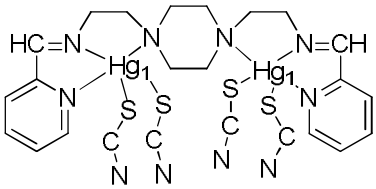
Where X= NCS^-, N_3^-



[Hg2(L9)(SCN)4]



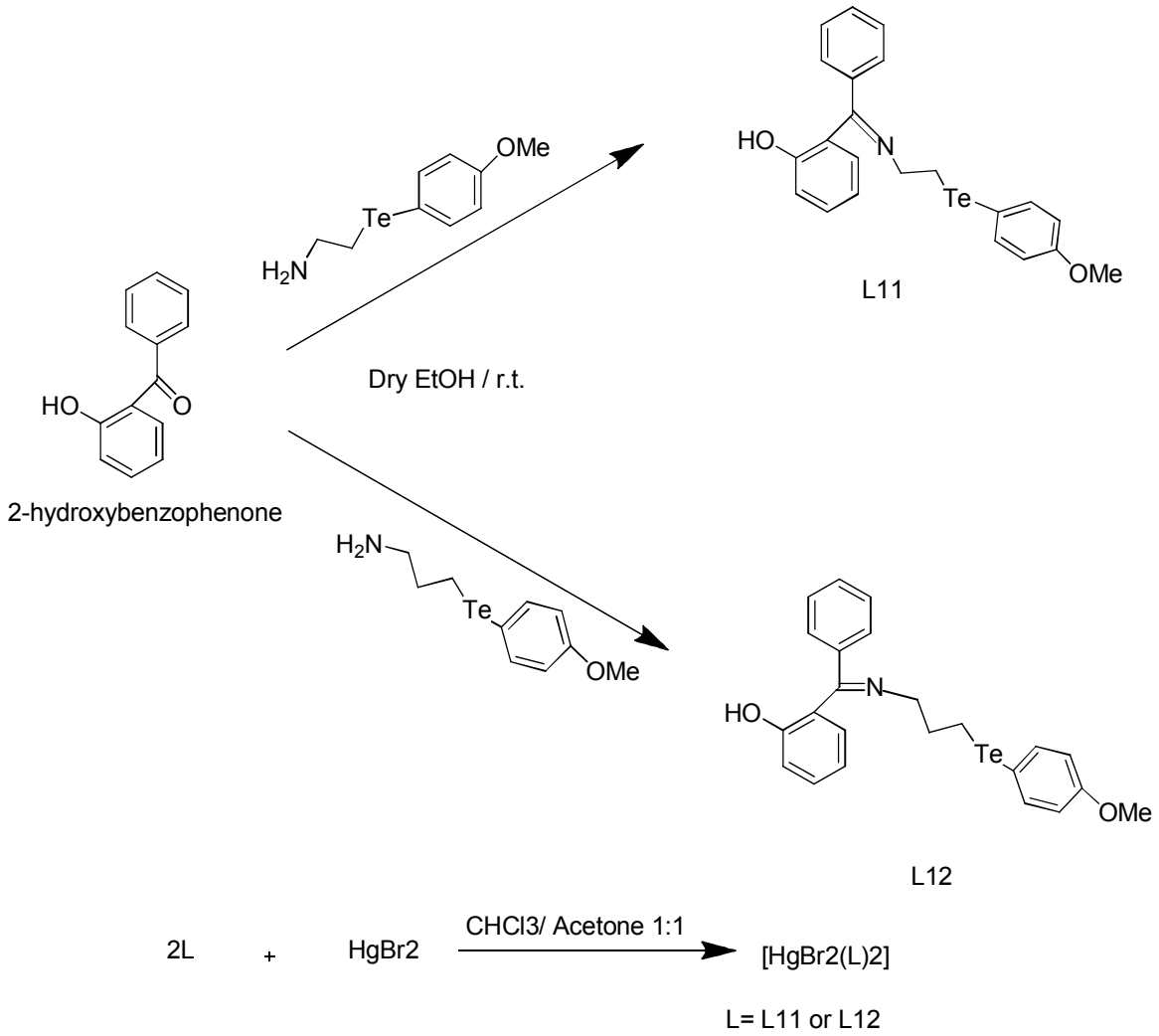
[Hg2(L10)(N3)4]



[Hg2(L10)(SCN)4]

Scheme: 6 Molecular structure of complexes.

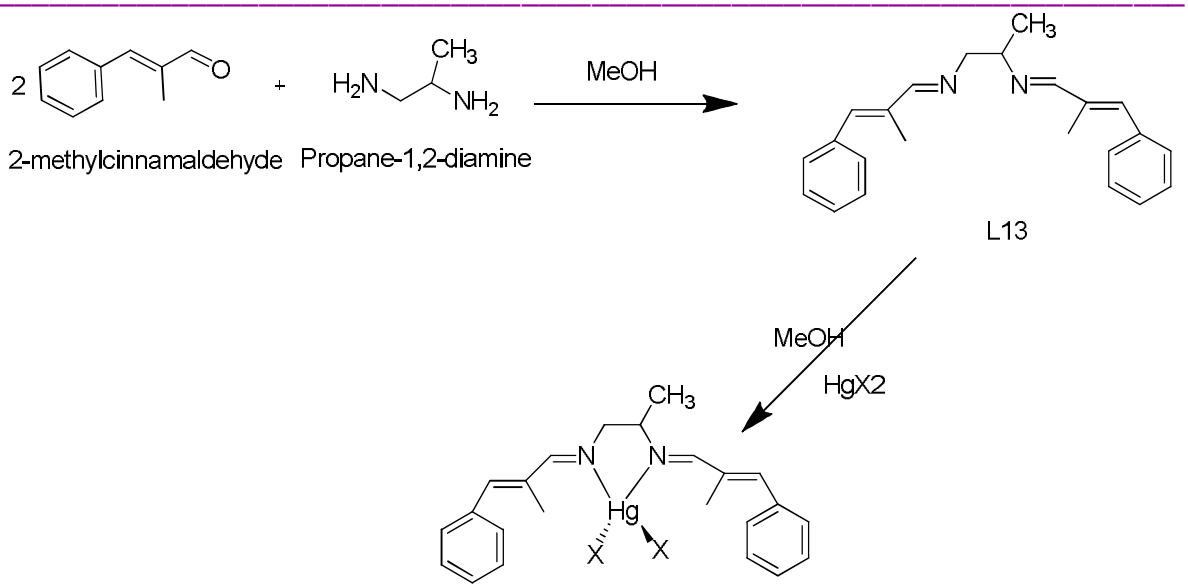
Schiff bases and related compounds continue to be interest for catalyst designing. Thiosemicarbazones [49,50] and salicylaldehyde derived chalcogenated Schiff bases [51,52] are known as capable ligands for the palladium-catalyzed Heck and Suzki reactions under aerobic condition. Here use of 2-hydroxyacetophenone derived Teluriated Schiff bases as ligands in Heck reaction. For examples of Schiff bases of 2-hydroxybenzophenone (HBP) containing chacogen functionalities ligands (L_{11}, L_{12}) are shown. (Scheme: 7) The ligands L_{11}, L_{12} are synthesized by further standarding general procedures [53-55]and their Hg(II) complexes are given in scheme -7.



Scheme: 7 Schiff bases and their complexes with mercury(II).

The ligands (L_{11}, L_{12}) and their complexes are stable but can be stored under ambient conditions up to six months. They are soluble in chloroform, DCM, MeCN, MeOH, EtOH and acetone but are sparingly soluble in hexane. The ligands L_{11}, L_{12} can behave as hemilabile ligands. Therefore lower values in MeCN may be due to partial substitution of weak Hg---N bond. These complexes are prepared the following methods described elsewhere[56].

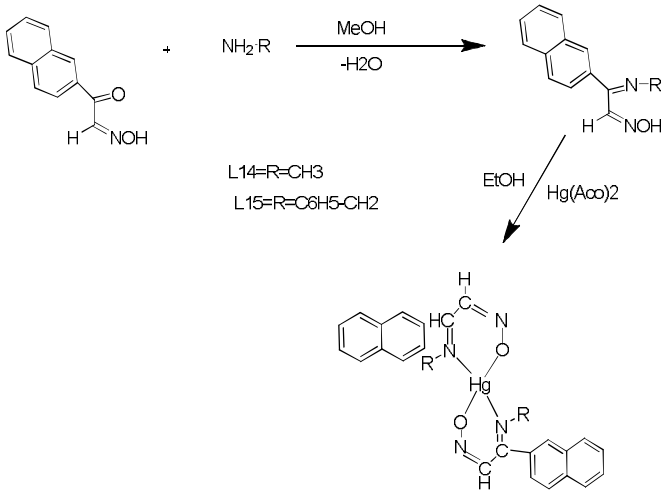
The 12 group metal complexes with polydentate ligands have been considerably investigated as latent glowing materials[57-59].The mercury(II) complexes are toxioc, which may effected the environment. Therefore the organisms are mobilization and hold of them in the environment.On other hand, here synthesized bidentate Schiff base complex with mercury(II). The asymmetric bidentate Schiff base ligand of N,N'-bis[α-methylcinamaldehydene]propane-1,2-diamine (L_{13}) and it complex with mercury(II), general formula of $[HgL_{13}X_2]$ (X= Cl^-, Br^-, I^-).Schiff base ligand (L_{13}) and it complex was synthesized by following methods described elsewhere.[6].(Scheme-8)



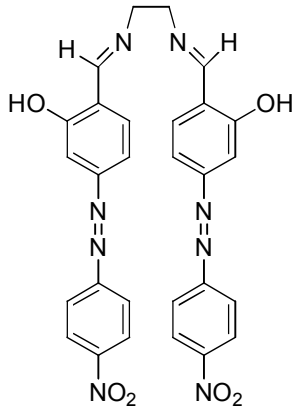
Scheme: 8 Schiff base ligand and their Hg(II) complex.

In series of mercury(II) and their Schiff bases complexes, another Schiff bases ligands methyliminoisonitroso-2-acetylnaphthalene (L_{14}) and benzyliminoisonitroso-2-acetylnaphthalene (L_{15}) and their Hg(II) complexes are tetrahedral geometries. Schiff bases and complexes are synthesized by following reported methods describe elsewhere[60].(Scheme-9)

A number of potential ion-carriers has been used in the creation of Hg(II) selective membrane electrodes. Here to be used some thio-substituted crown ethers as neutral carriers in membrane transport [61-63], in solid phase extraction [64] and in polyvinyl chloride (PVC) membrane electrode [65-67] studies of some transition and heavy metal ions. There are many commercial mercury(II) ion-selective electrodes reported in literature. A interesting synthesis of a PVC-based Schiff base sensor for Hg(II) ions. Schiff base ligand (L_{16}) bis[5-(4-nitrophenyl)azo salicylaldehyde)] (BNAS) was prepared by following method describe elsewhere[68].(Fig: 8). Which are used as an excellent neutral carrier in construction of mercury(II)-PVC membrane electrode.



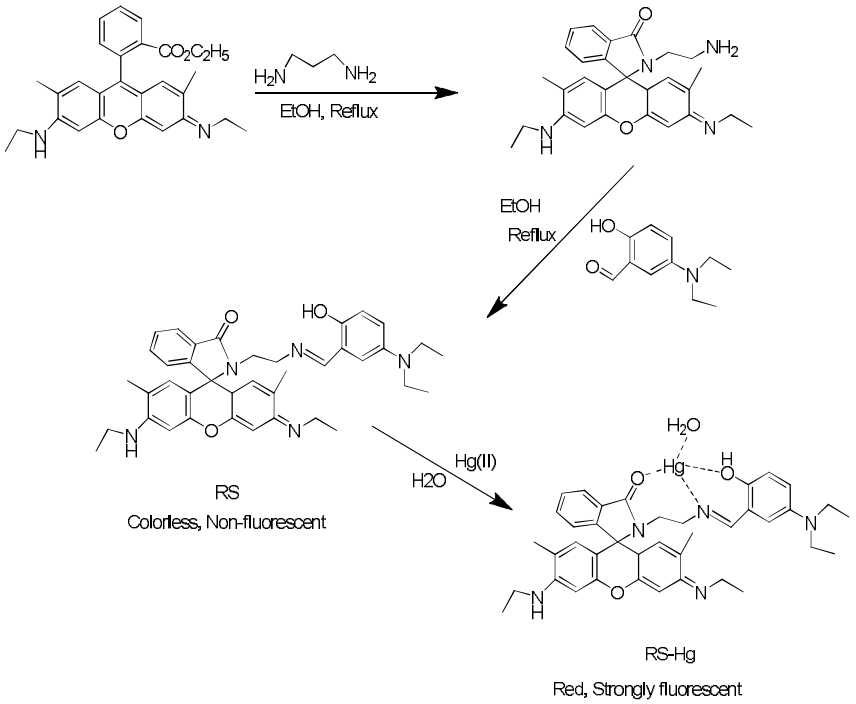
Scheme:9- The structure of ligands L_{14}, L_{15} and their Hg(II) complexes.



L16

Figure: 8 Structure of Schiff base ligand (L_{16}).

The rhodamine-ethylenediamine derived Schiff base (RS) is also used in determination of Hg(II) ions in aqueous solution, which is good binding framework for Hg(II) ions and attached 4-diethylaminosalicylaldehyde as additional binding groups, those are create ring-opening for complexation with Hg(II) ions. The aqueous solution of RS gave rise with Hg(II) ions as enhanced fluorescence as well as visual change from colorless to pink, indicating that Hg(II) did induce the ring-opening with RS. Schiff base was synthesized, first N-(rhodamine-6G)lactam-ethylenediamine then RS by following method described elsewhere[69]. (Scheme-10)



Scheme: 10 Synthetic rout of RS and their complexation for determination of Hg(II) ions.

APPLICATIONS OF SCHIFF BASES AND THEIR COMPLEXATION WITH MERCURY(II) :

We known that mercury is most prevalent toxic metal element in environment, which can easily passing through biological membranes such as skin, respiratory and gastrointestinal tissues. Whenever our body absorbed mercury, then it causes damage to the central nervous and endocrine system. A number of symptoms are observed mercuric exposure including digestive, kidney and especially neurological diseases. In such ways it must be necessary that mercury identify and find out. There are many analytical technique as well as equipments have been developed for identification and separation of mercury from environment or nature. The several Schiff bases have developed, which are showing photo physical properties for identification of metals. The many complexation of mercury(II) with Schiff bases are showing colorful, that can be easily identify and separated with help of UV/Vis or fluorosecence of a chromophore. The mercury and it salts find wide applications in diverse industrial process and products like paints, electronic equipments, batteries[70,71], sensor absorbents, [73] in paper industries as preservations, cleaning solutions, cosmetics, manometers, thermostate probes, thermoelectric devices,[74-78] amalgams with many many metals and seed dressings in agricultures[1].

Schiff bases and their complexes have ability in biologically, liquid crystals dyes, luminophore and polymer stabilizers, therefore its have a lot of applications such as antidepressants, antimicrobial, antitumor, antiphlogogistic, nematocide and medicinal agents[79,80]. Due to toxic nature of mercury its may generally used in medicine field, such as antifungal, antibacterial, anticancer and herbicidal[81,82].

RESULT AND DISCUSSION

Here we are studies that two main charecterestic, one is Schiff bases and other is their mercury complexes. The Schiff bases are very applicable to human being and environment. Its have a lot of applications such as antidepressants, antimicrobial, antitumor, antiphlogogistic etc. But Schiff complexes of mercury is very toxic to human being and environment. The chemistry of mercury and their complexes are very important. Its forms different diversity in coordination in geometries and frameworks. Mercury formed multidentate N-donor Schiff base complexes in different denticities. This types of complexes are very useful to identification and removal of mercury from environment.

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