DOI : 10.35124/bca.2019.19.2.4365

Biochem. Cell. Arch. Vol. 19, No. 2, pp. 4365-4377, 2019 | www.connectjournals.com/bca | *ISSN 0972-5075*

VARIOUS APPLICATIONS OF CARBAZONE DERIVATIVES AND THEIR METAL COMPLEXES : A REVIEW

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(Received 30 March 2019, Revised 17 June 2019, Accepted 2 July 2019)

ABSTRACT : Carbazone Derivatives (CD) (semicarbazone, semithiocarbasone) are produced by the condensation reaction between a aldehyde (or ketone) with a carbazide derivatives (semicarbazide, semithiocarbazide). CD and their metal complexes existent a wide range of implementation that stretch from their ply in the medicinal and pharmaceutical area because of their major significant pharmacological characteristic such as anti-fungal,anti-bacterial, anti-cancer, anti-human immunodeficiency virus, anti-inflammation, anti-neoplastic,inhibition corrosion, antioxidation, antiradical. This paper reviews the definition, importance and various applications of carbazone derivatives with transitional metal.

Key words : Carbazone derivatives, transitional metal.

INTRODUCTION

Carbon Derivative generally make as chelating ligands consist of the donor (imine groups) who react together with transition metal unoccupied (d-orbital) product complexes. These are multi-function ligands in togetheranionic and neutral shapes (Ljubijankic *et al*, 2016). The functional grouping for chelation is where X = Oxygen and Sulphur for (semicarbazones and thiosemicarbazones), respectively. In some of the complexes, the (CD) coordinate to the metal ion as a bi-dentate ligand bonding by the S or O and the (N, hydrazino group) (Srivastava *et al*, 2015; Xu *et al*, 2013) (Fig. 1).

Carbazone Derivatives complexes are known to exhibit interesting stereochemical, electrochemical, physicochemical, pharmacological properties and biological activities. The complexes can show bio-activities which are not found by the free ligands. Several transition metals complexes have various applications such as anti-viral agent, catalyst and chemical sensor (Saddam *et al*, 2017; Gajendra and Vidhi, 2016).

Carbazone Derivatives (CD) and their metal complexes have been used as a drug and possess a wide various of biological activities and they are as well a useful type for inorganic biological processes, as well applicable in physical sciences such as electro chemical sensor, Langmuir film and non-linear optical (NLO) (Adwav *et al*, 2018; Mourva *et al*, 2013). (CD) are exceptionally diversity multi-dentate ligands that coordinate to lots transition metals with literature discuss application, and their potential to coordinate transition metal ions in geometries and a different of oxidation states (Ragab *et al*, 2016; Ali *et al*, 2014). It often acts as high affinity bi-dentate, tri-dentate, or tetra-dentate chelating ligand for the composition of metal complexes because of various of donor groups of S, O and N (Wurood, 2018). Cu (ii), Ni (ii) and Co(ii) complexes of (CD) significantly increase the bio-logical activities such asanti-bacterial, anti-HIV, anti-fungal, anti-inflammatory and effective anti-proliferative agents in breastcancer, targeting RNR, which deserve fulfillment as anti-cancer drugs (Niharika and Sanjay, 2015; Salman *et al*, 2014). (CD) shows a great variety of biological activity against tumor, influenza, protozoa virus, leukemia, malaria, bacteria, neoplastic and anti-convulsant. They have as well been used in metal analysis, optical computing, optical storage, for device applications relative to telecommunications and optical information processing (Basima *et al*, 2016; Wurood *et al*, 2018). (CD) are known to have an activity of anti-viral, anti-infective and anti-neoplastic through binding to Cu or Fe in cells (Jungang *et al*, 2018).

Many organic compounds a specially those containing un saturated double bonds and triple bonds, electronegative atoms (sulfur, nitrogen and oxygen) and aromatic rings have proposed as productive organic inhibitors for mild-steel metallic corrosion (acid media) (Idouhli *et al*, 2018). The properties and structure of the inhibitor such as steric factor, active groups, molecular

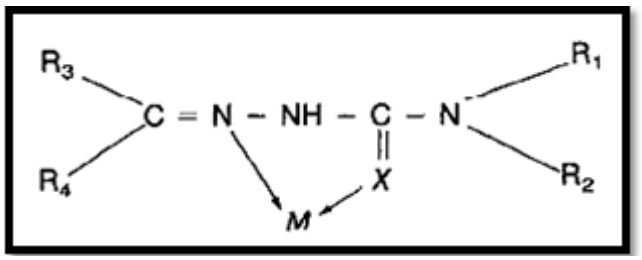
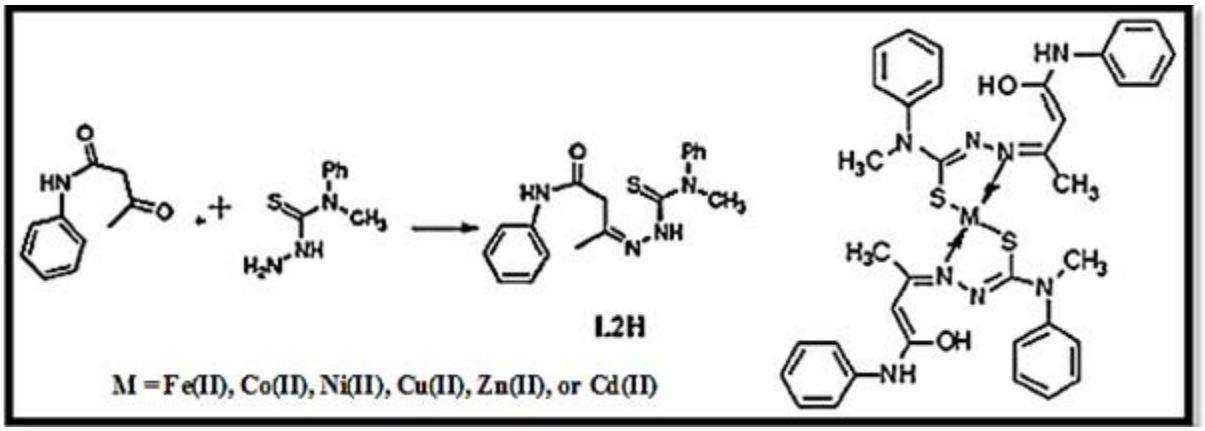


Fig. 1 : Structure of Carbazone Derivatives, where X = O, S.

micrbial and anti-fungal activities have been studied (Scheme 2).

Mei-Hsiu *et al* (2015) were synthesized a chain of derivatives [salicylaldehyde 2-aryl4-[(1H-benzo[d] imidazol-2-yl) methyl] semicarbazones] by reactions emicarbazides with different compound, such as salicylaldehyde, 5-chlorosalicylaldehyde and 4-methoxysalicylaldehyde. The semicarbazones derivatives



Scheme 1 : Synthesis of (L_2H) and its complexes.

size, molecular structure, molecular weight, aromatic, density of electron of the donor atoms and (p orbital) character of giving electrons effect on metal surface by the adsorption of corrosion inhibitor (Sam *et al*, 2017). (CD) molecule is rationally a good corrosion inhibitor because to the presence of N atom also an (imine group) (Awf, 2018). In the study of metabolism and in slow releases or long-acting drugs in nutrition and design of repository, (CD) complexes have become increasingly important in recent years (Su *et al*, 2018). Cu(II) and Fe(III) complexes of (thiosemicarbazone and semicarbazone) show higher anti-oxidant and anti-radical compared to the free ligands (Asha *et al*, 2018; Enass, 2016).

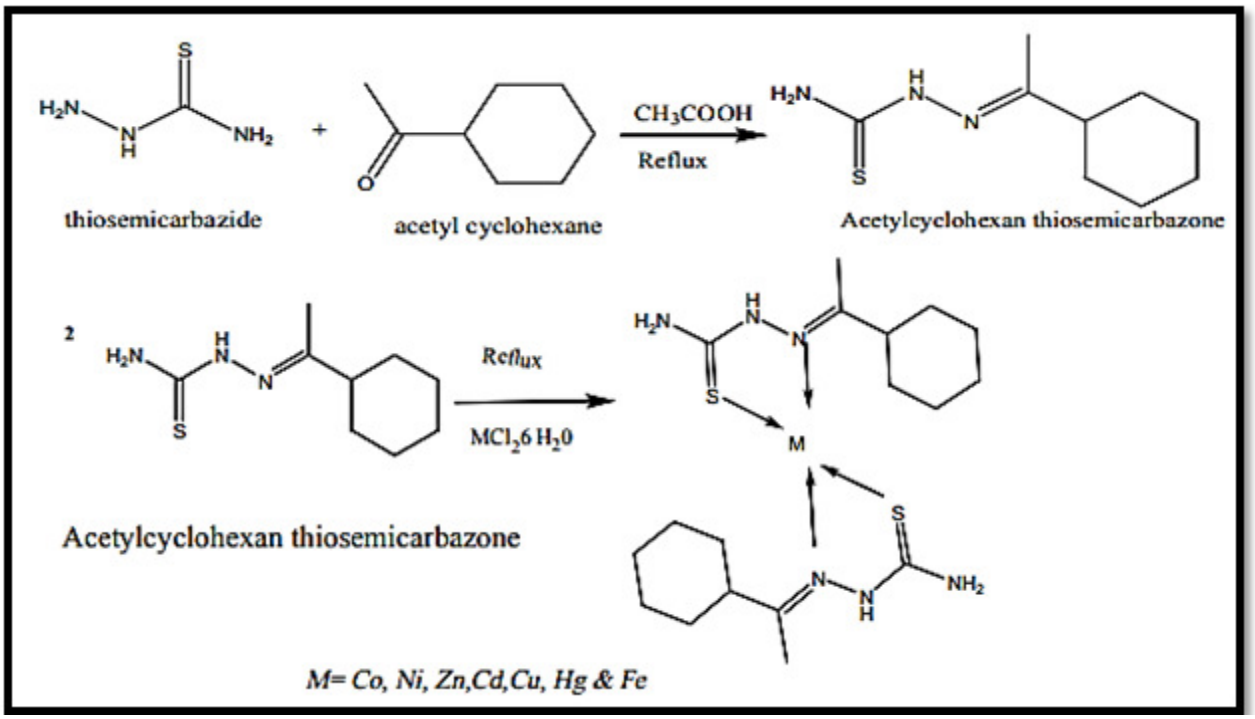
Priya *et al* (2015) were studied cytotoxic activities of ligand (L_2H), where (L_2H) is [acetoacetanilideN(4)-methyl (phenyl) thiosemicarbazone]and its various metal complexes. Anti-tumour studies of Cu-complex were performed by Daltons Lymphoma. Management of Cu-complex at various concentrations (1, 5 and10 $mg.kg^{-1}$) led to inhibition of tumor growth and increased survival rate in mice carrying Ascites tumors (Scheme 1).

Kalapala *et al* (2015) were synthesized of some new acetylhexanthiosemicarbazone [L] by condensation of cyclohexylmethylketone with thiosemicarbazide. And their metal complexes were synthesized by reaction[L] and metal ions [M = Cu(ii),Co(ii), Ni(II) and Fe(III)] and identification by C.H.N.S, FTIR, Mass spectrum. Anti-co-ordination with Ni (II) ion to productnew Ni (II) complexes (Scheme 3). The ligandsand its complexes were characterized by FTIR, ^{1}HNMR and X-Ray.

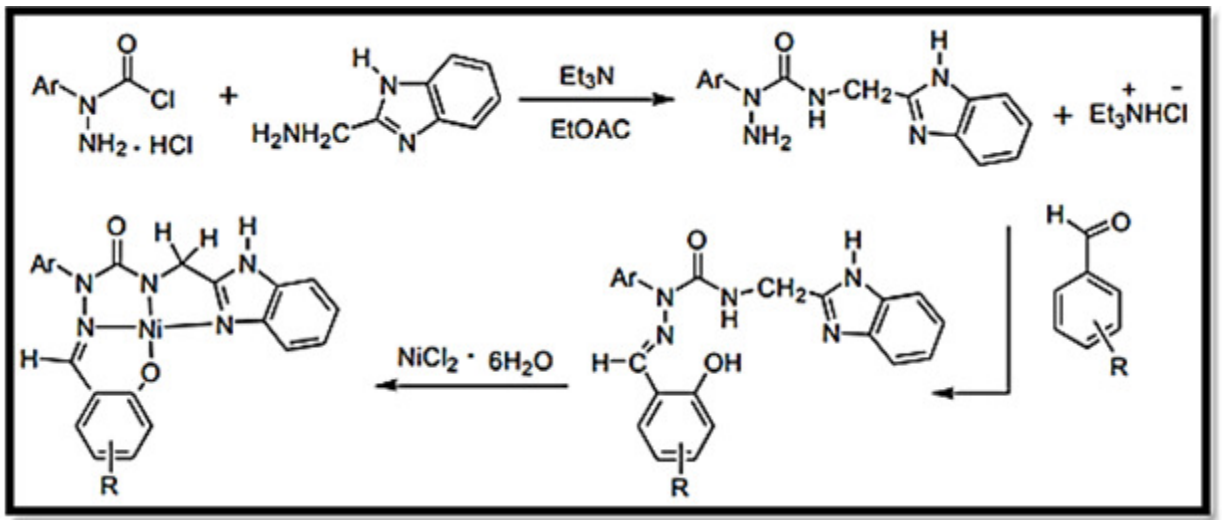
Singh *et al* (2015) were prepared a new chain of metal complexes [$M(L)_2$] (where M = Sn(ii), Pb(ii) and ligand (HL) =(semicarbazone, thiosemicarbazone or phenylthiosemicarbazone) and characterized by C%, H%, N%, S% conductance, UV–Vis, FTIR and ^{1}H-,^{13}C-NMR. Elemental analysis of the metal complexes proposed (1:2) (M–L) stoichiometry. The electronic spectra refer to a distorted tetrahedral geometry for all complexes. The bio-logical action of ligand and its complex was studied in 4 types of bacteria and 4 strains of fungus.

The anti-microbial action of the complexes was appear higher than the free ligand (Fig. 2).

Neha *et al* (2015) were prepared semicarbazone and thiosemicarbazone ligands by reaction acetylacetone, benzoylacetone and gly-oxal with semicarbazidehydrochloride and thiosemicarbazide in ethanol as solvent. A series of complexes Tin (II) for these ligands was prepared by dry (THF) as a reaction medium in (1:1) molar ratio. By elemental analysis, molar conductance and UV-Vis, FT-IR, ^{1}H-^{13}CNMR spectral studies, Sn (II) complexes were diagnosed. The ligands and their complexes have been examined for anti-bacterial and anti-fungal and have been appear to be very activity in this regard. Sn(II) complexes appear to be more activity than free ligands (Fig. 3).



Scheme 2 : Synthesis of [L] and its complexes.



Scheme 3 : Complexes of Ni (II),Ar = C_6H_5 (a), p-$CH_3C_6H_4$ (b), p-$CH_3OC_6H_4$(c), R = H,5-Cl, 4-OCH.

Sushil and Atnafu (2015) were synthesized VO (IV & V) complexes from semicarbazones and thiosemicarbazones (HL_3, H_2L^4, L_1 and HL_2) derived from pyrrole-2-carbaldehyde (HL_3, H_2L_4) and furan-2-carbaldehyde (L_1, HL_2). The complexes were identification by A.A, UV-Vis, FTIR, ^{1}HNMR spectra, magnetic measurement, mass spectra and conductivity. The complexes except $[VO(L_2)(OH)_2]$. H_2O are para-magnetic and square pyramidal geometry. Ligands and their complexes have demonstrated significant biological activity as anti-bacterial agents also anti-fungal properties (Fig. 4).

Elena *et al* (2015) were synthesized ligand [1-phenyl-3-methyl-4-benzoyl-5-pyrazolone-4-ethyl-thiosemicarbazone] (HL) and its complexes Cu(II), V(V) and Ni(II). The (HL) has been characterized by C.H.N.S, FT-IR, ^{1}HNMR and ^{13}CNMR spectroscopy. The complexes have been characterized by magnetic measurements, conductance, FT-R, UV-Vis and ^{1}HNMR spectral studies, X-Ray diffraction. The anti-bacterial activity of the complexes against four type of becterial and the anti-fungal activity for the metal complexes were higher than for free ligand. On the spread of HL-60 cells, the effect of free ligand and its complexes has been tested (Scheme 4).

Kavita *et al* (2016) have been synthesized some new

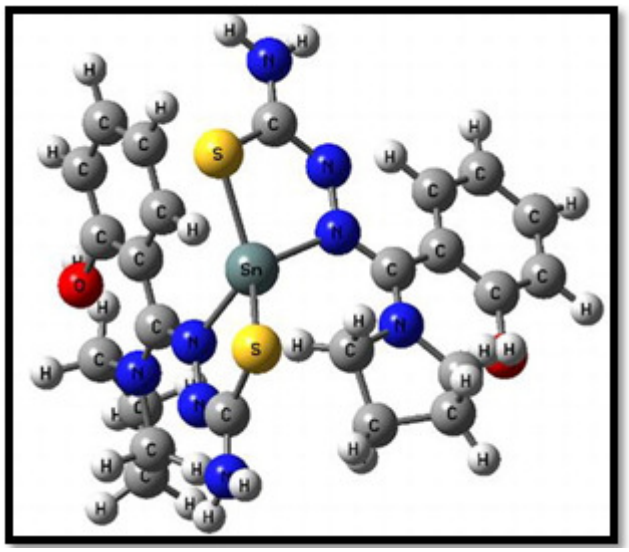


Fig. 2 : Structure of Sn(II) complex.

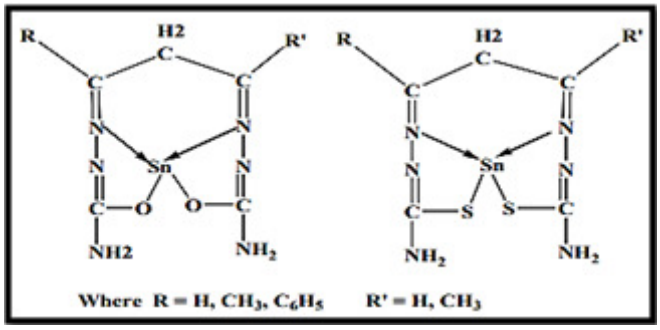


Fig. 3 : Structure of Sn (II) complexes.

Neha and Varshney (2016) were prepared series of new Ti(IV) complexes from semicarbazone and thiosemicarbazone ligands. These ligands have been prepared by reaction carbonyl compounds with thiosemicarbazide and semicarbazide hydrochloride in ethanol as solvent. The complexes have been characterized on the basis of C.H.N.S, ^{1}H, ^{13}C-NMR, UV-Vis, FT-IR spectral studies. Ligands and its complexes were examined for anti-bacterial and anti-fungal activity and were appear to be very action in this regard. Titanium (IV) complexes show more active than free ligands (Fig. 6).

Kpomah *et al* (2016) were synthesized complexes of ligand by condensation of acetaldehyde with thiosemicarbazide in the presence of $FeCl_3.6H_2O$ and $CuSO_4.5H_2O$. The products were investigation by molar conductance, FTIR and UV-Vis study. Based on spectral data, general formulas were proposed: $Cu(ADTSC)_2SO_4.H_2O$ and $Fe(ADTSC)_2Cl_2.3H_2O$ and octahedral geometry of the complex. The ligand and its complexes were examined for their anti-bacterial activity against two types of bacteria. The results of these studies showed that metal complexes are more active than free ligand (Fig. 7).

Sanjay *et al* (2016) were synthesized complexes of general formula $[M(L)_2]X_2$, where, M = Copper(II) and

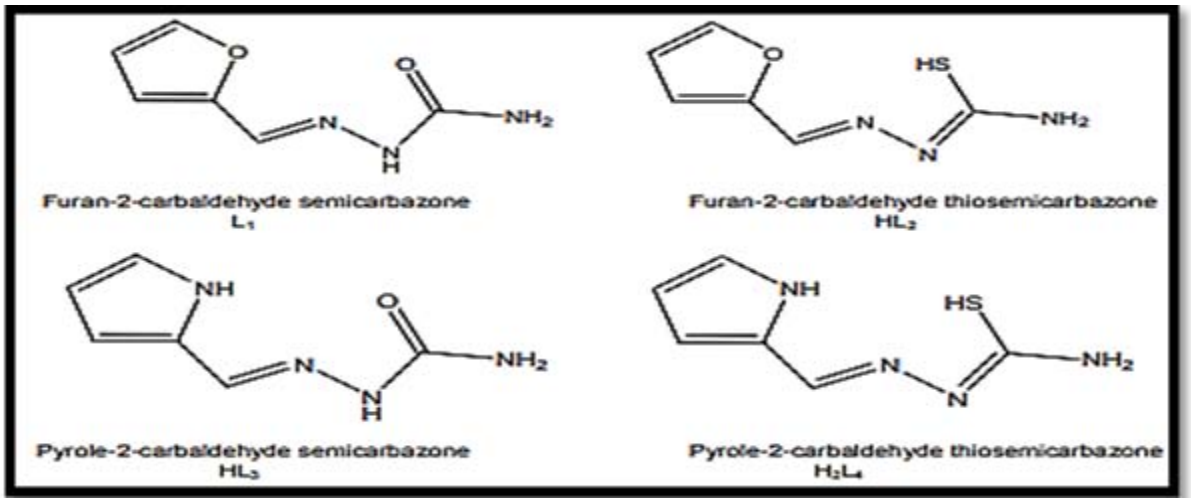
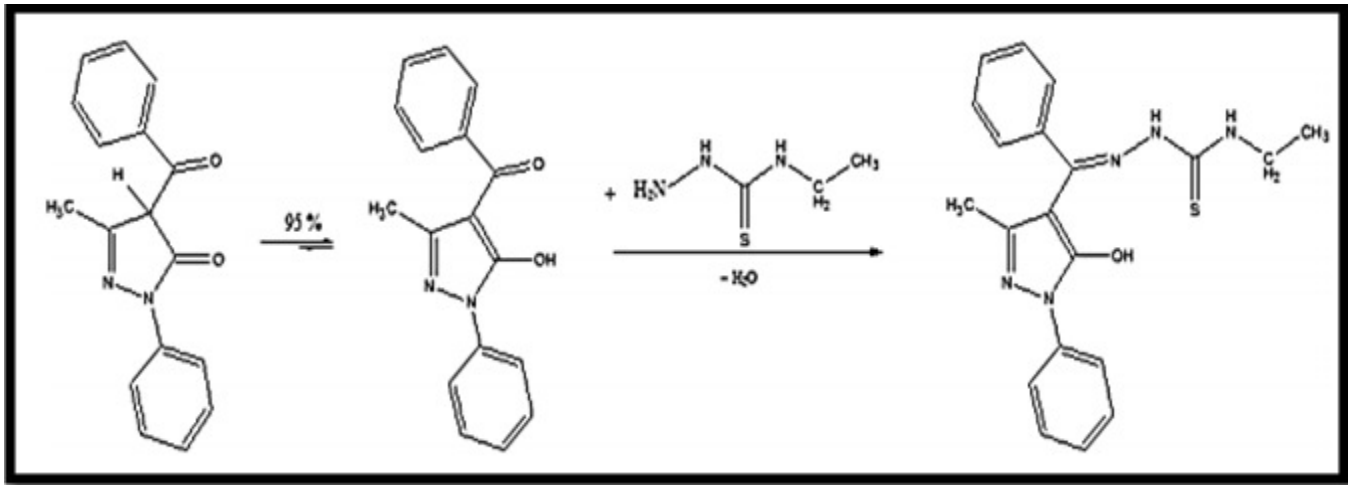


Fig. 4 : Structure of ligands (L^1 to H_2L^4).

Pd (II) complexes with biologically active nitrogen donor ligands. The ligands used in these studies are semicarbazones and thiosemicarbazones and prepared by the reaction of diketones with thiosemicarbazides and semicarbazide hydrochloride. The geometry and coordination pattern of complexes were confirmed by elemental analysis, FT-IR and ^{1}H,^{13}C-NMR spectral studies (Fig. 5).

$X = NO_3$, Cl) and $[M(L)_2]X_3$, where M = iron(III) and X = NO_3,Cl) by the reaction semicarbazone (L_a)/ thiosemicarbazone (L_b) derived from (2-acetylbenzofuran) with metal salts. The ligands and its complexes were investigation by ^{1}HNMR, C.H.N.S, magnetic measurement, conductivity, FTIR, A.A and UV-Vis studies. Based on the results of the measurements, it was found that the complexes have a tetrahedral geometry. The metal complexes appear higher anti-



Scheme 4 : Synthesis of ligand (HL).

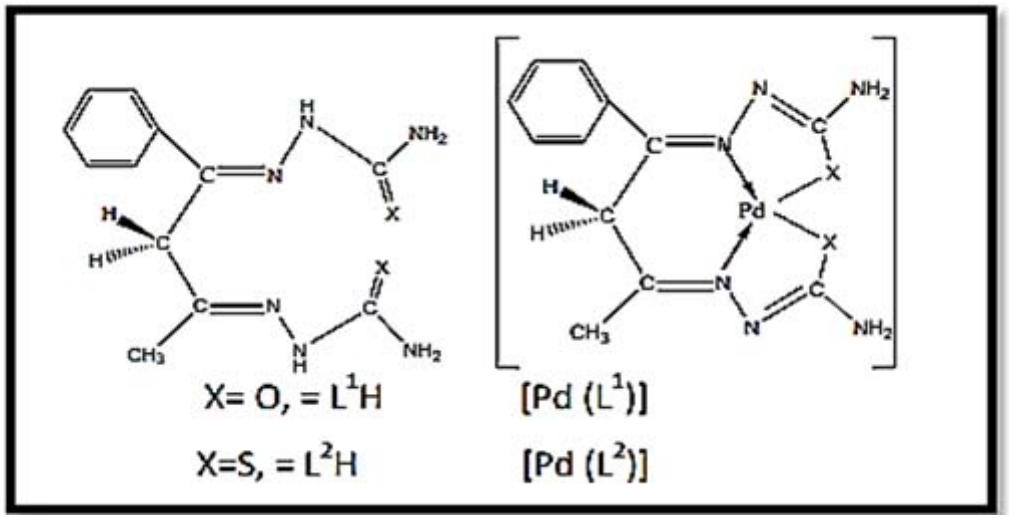


Fig. 5 : Ligands and its Pd (II) complexes.

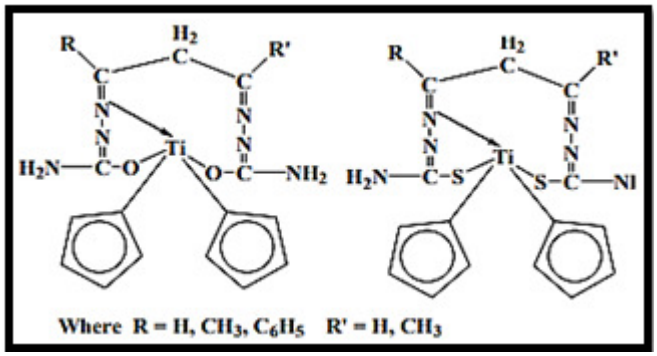


Fig. 6 : Structure of Ti (IV) complexes.

oxidants, anti-bacterial activities and a radical scavenging potency compared to Ligand (Scheme 5).

Shirisha (2017) was synthesized a novel ligand [benzylmethyl-4-methyl-3-thiosemicarbazone] (BMM) derived from benzylmethylketone and 4-methylthiosemicarbazide and investigation by C.H.N.S, FT-IR, ^{1}H,^{13}C-NMR and physical measurements. Ligand showed an average antibacterial activity on three types of bacteria compared with standard medicine, ciprofloxacin (Scheme 6).

Shobha (2017) was synthesized2,4-Dihydroxy cinnamaldehydethiosemicarbazide. Complex of 2,4-dihydroxy cinnamaldehydethiosemicarbazide with transition metals, Vanadium (v) has been synthesized in methanol medium. The ligand and its complexes was investigation by M.P, C.H.N.S, anti-microbial activity and absorption spectra. The stability constant, change in free energy and dissociation constant of V (v) has been determined by Job's variation and mole ratio method indicate that the (M:L) is (2:1). Anti-micrbial activity and anti-fungal activity have been studied (Fig. 8).

Shirode *et al* (2017) were study mixed-ligand complexes of metals ion with O, N and S donor ligands. Co(II)) complexes of formula $Co(L^1L^2)Cl_2$ Where L^1 = salicylaldehydesemicarbazone and L^2 = salicylaldehydeoxime, o-hydroxyacetophenoneoxime, benzaldehyde semicarbazone, anthranilicacid, acetone semicarbazone and acetophenonesemicarbazone have

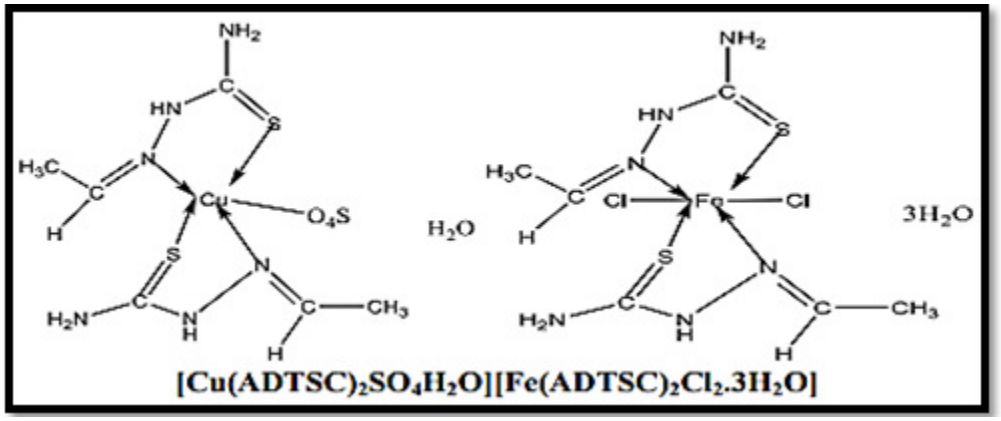
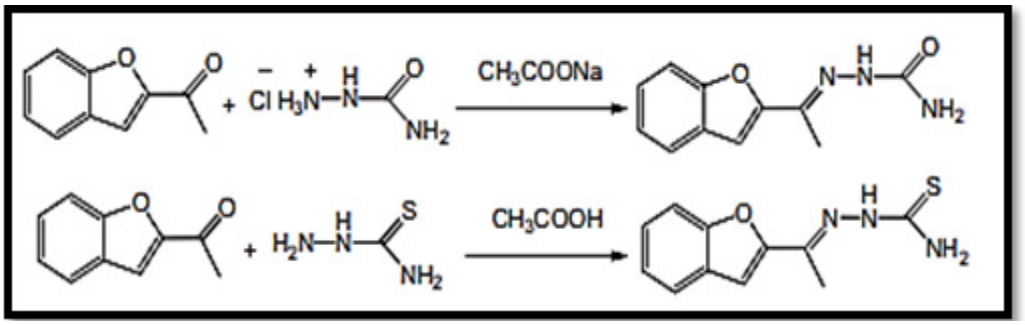
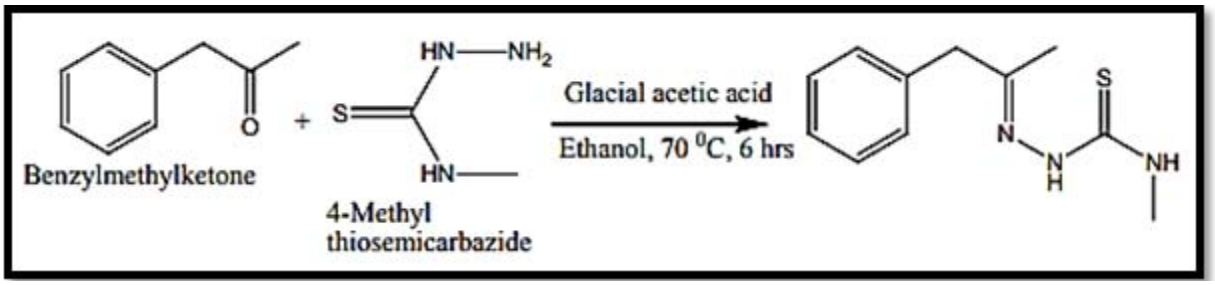


Fig. 7 : Metal complexes of acetaldehyde thiosemicarbazone.



Scheme 5 : Synthesis of ligand (La) and ligand (Lb).



Scheme 6 : Synthesis of ligand (BMM).

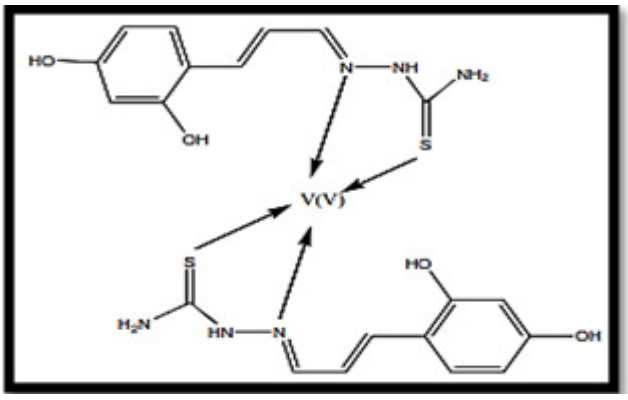


Fig. 8 : Structure of V (V) complex.

been synthesized and investigation on the basis of FTIR and UV-Vis spectral analysis, magnetic measurement, conductivity measurement, TGA, anti-microbial activities. Based on the results of the measurements, complexes shows electrolytic nature, octahedral geometry and having more anti-microbial activity than the free ligands (Fig. 9).

Vidhya and Rathika (2017) were prepared Co (II) complexes of general formula $[Co(L^4)_2Cl_2]$, where, L^4 = [2,3,4-trimethoxy benzaldehyde semicarbazone]. Complexes have been investigation on the basis FTIR, UV-Vis, mass spectra and ^{1}HNMR spectra. The data agree with the suggested structures and appear that the complexes were eventually dissolved into the corresponding ligand. The complexes were examined for anti-microbial activity by the disc diffusion technique. Activity data appear that semicarbaponic complexes are more activity than free ligand (Fig. 10).

Farhadi *et al* (2017) were synthesized Cu(II) semicarbazone complex of general formula $[Cu_2(HL)_2(PMo_{12}O_{40})(OCH_3)_2(Cl)(H_2O)]\cdot 8CH_3OH\cdot 4H_2O$, where ligand (HL) = [pyridine-2-carbaldehyde-semicarbazone]. The complex was investigation using FTIR, UV–Vis, TGA study and X-ray diffraction. Based on spectral data, Ligand acts bi-dentate. The photo

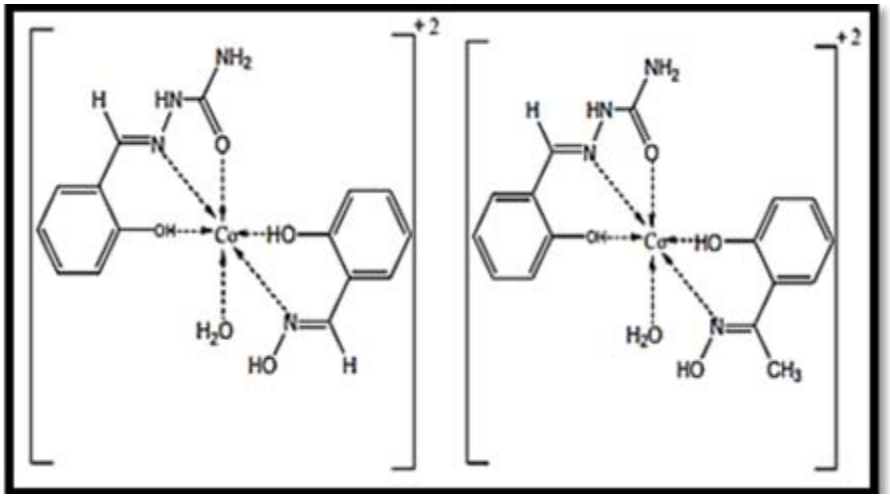


Fig. 9 : Structure of Co(II) complexes.

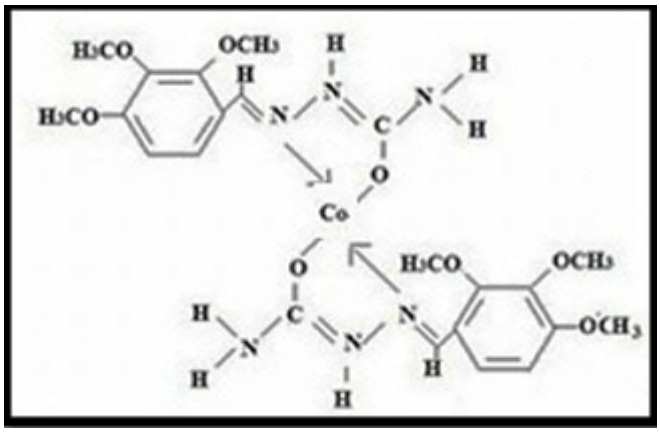


Fig. 10 : Proposed structure of $[Co(L^4)_2]$.

(RhB) (Fig. 11).

Shirode *et al* (2017) were prepared the Mixed-ligand complexes of formula $M(L^1L^2)Cl_2$, where, M = Cobalt(II), nickel(II) and copper(II), L^1 = O-hydroxyacetophenone oxime and L^2= Salicylaldehydesemicarbazone by reacting equimolar quantities of transition metal(II) chloride with two ligands L^1 and L^2. These synthesized complexes have been studied by of UV-Vis and FTIR, conductance, magnetic measurement, TGA and anti-microbial activities. Based on measurements results, complexes appear octahedral geometry, good anti-microbial activity than the free ligands (Fig. 12).

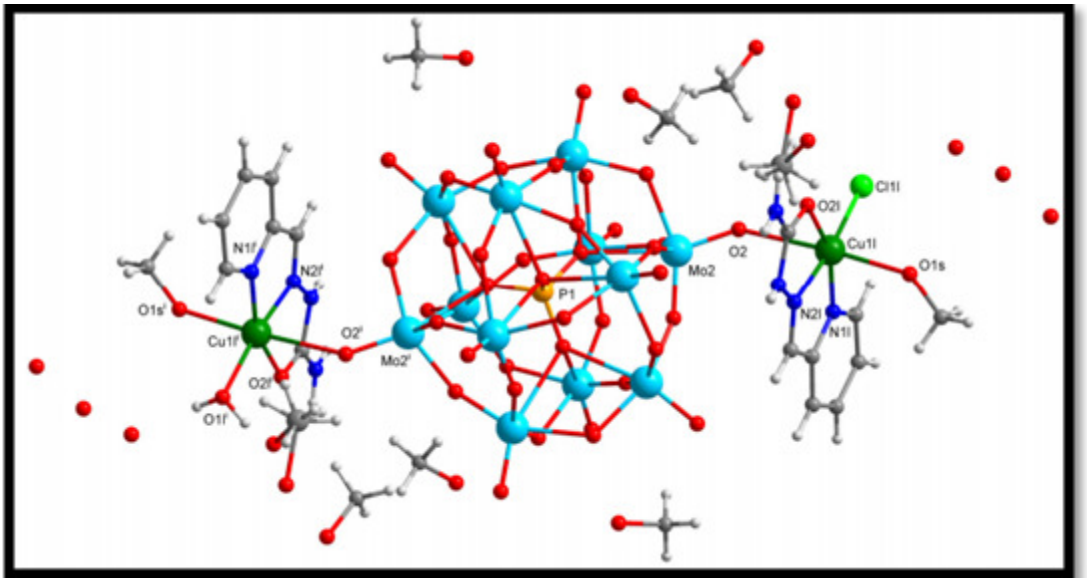


Fig. 11 : The structure of Cu(II) complex.

catalytic properties of the nanohoepide Cu-complex were characterized in detail. The data of photosynthetic experiments appear that they can be used as an effective and recoverable photo catalyst to completede-gradation of (cationic methylene blue) (MB) and Rhoda mineB

Masoumeh *et al* (2017) were synthesized semicarbazone or thiosemicarbazone complexes by reaction the alkylated isatin with semicarbazide or thiosemicarbazide and identification by FTIR, ^{1}H-^{13}CNMR and C.H.N.S. Cytotoxic activity of complexes

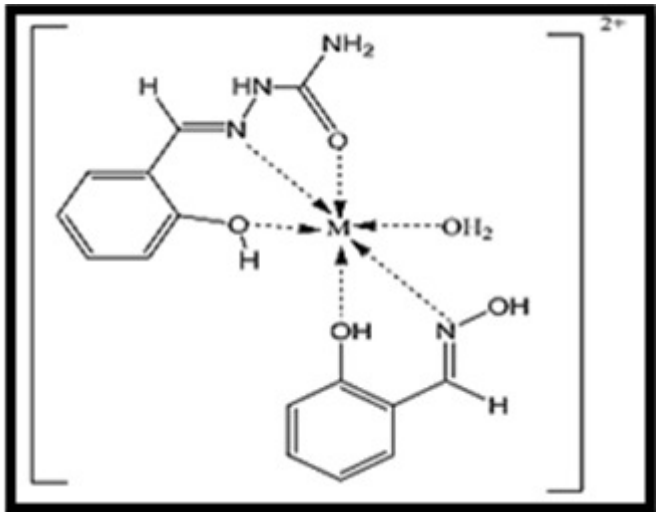


Fig. 12 : Structure of Co(II), Ni(II) and Co(II) complexes.

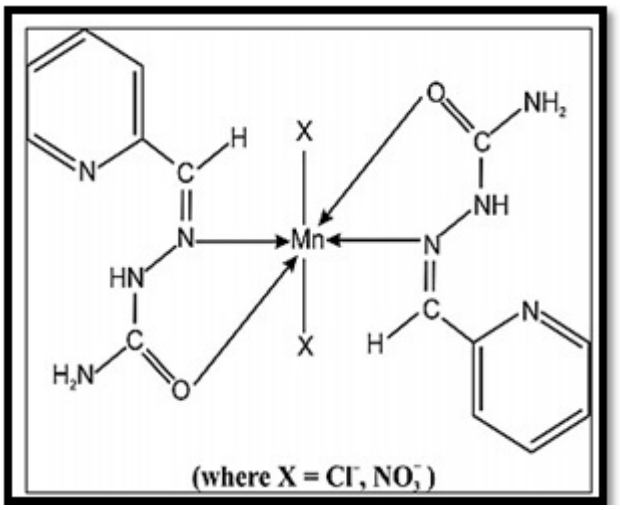
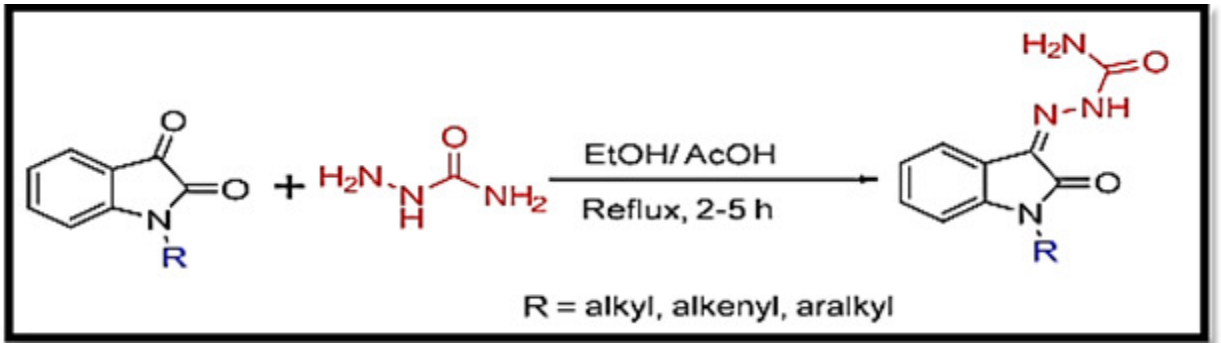
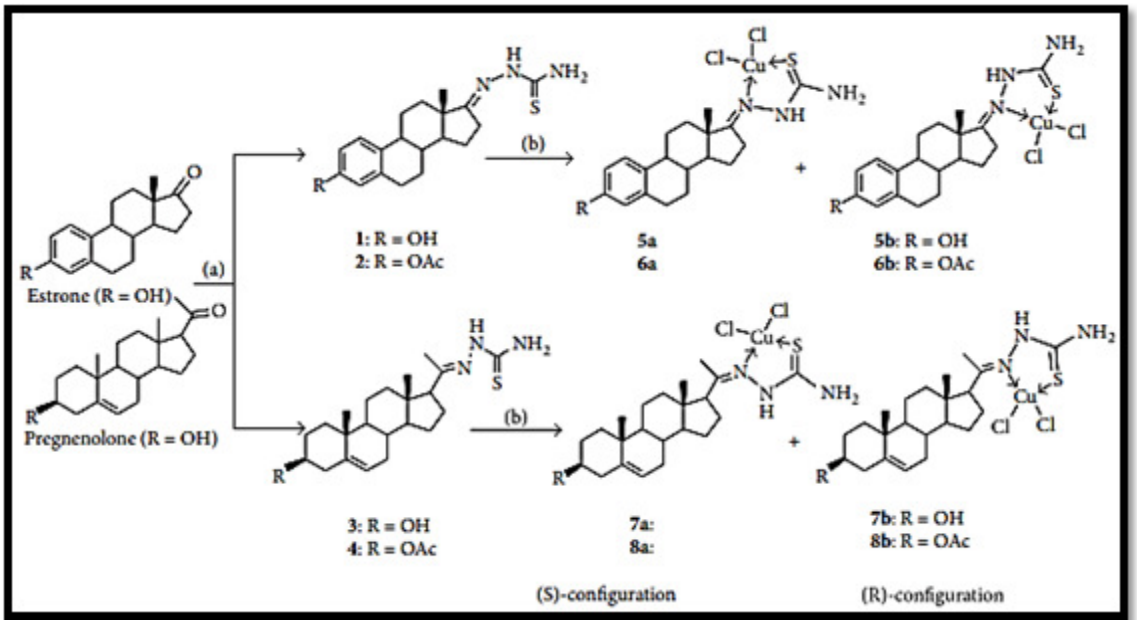


Fig. 13 : Mn(II) complexes with ligand L^1.



Scheme 7 : Synthesis of compounds.



Scheme 8: Synthesis of complexes, (a) thiosemicarbazide, acetic acid and ethanol; (b) $CuCl_2 \cdot 2H_2O$, $CH_3OH/CHCl_3$ = 1 : 1.

was studied by MTT [3-(4,5-dimethylthiazol-2-yl)-2, 5-diphenyltetrazolium bromide] against MCF-7iii and MDA-231iv breast cancer cell lines. Anti-microbial activities of complexes have been studied against various types of microorganisms, including (G^-) and (G^+) bacteria as well fungi by the micro-dilution method (Scheme 7).

Ruchi (2017) was synthesized and characterized Manganese (II) complexes with four ligands such as [2-formylpyridine semicarbazone] (L^1), [2-formylpyridine thiosemicarbazone] (L^2), [5-methyl-2-formylpyridine-semicarbazone] (L^3) and [5- methyl-2-formylpyridine-thiosemicarbazone] (L^4). The ligands and its complexes

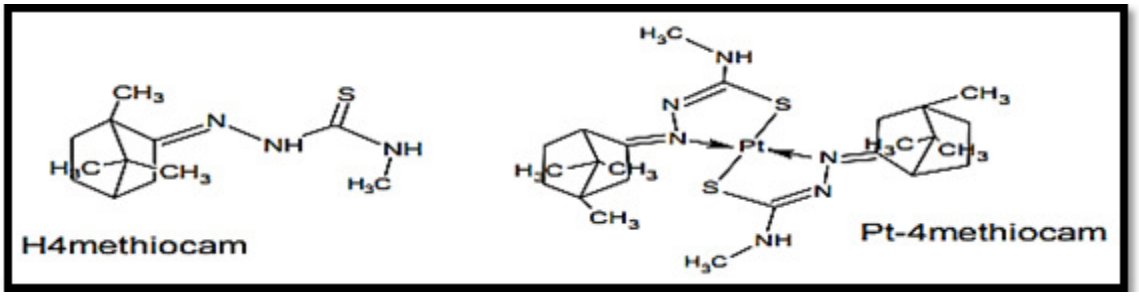
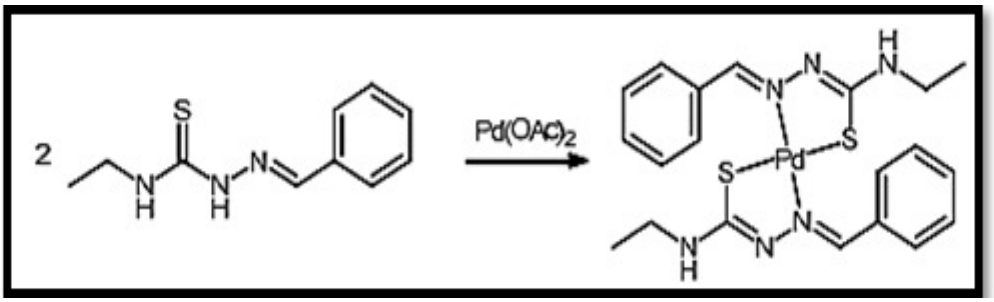
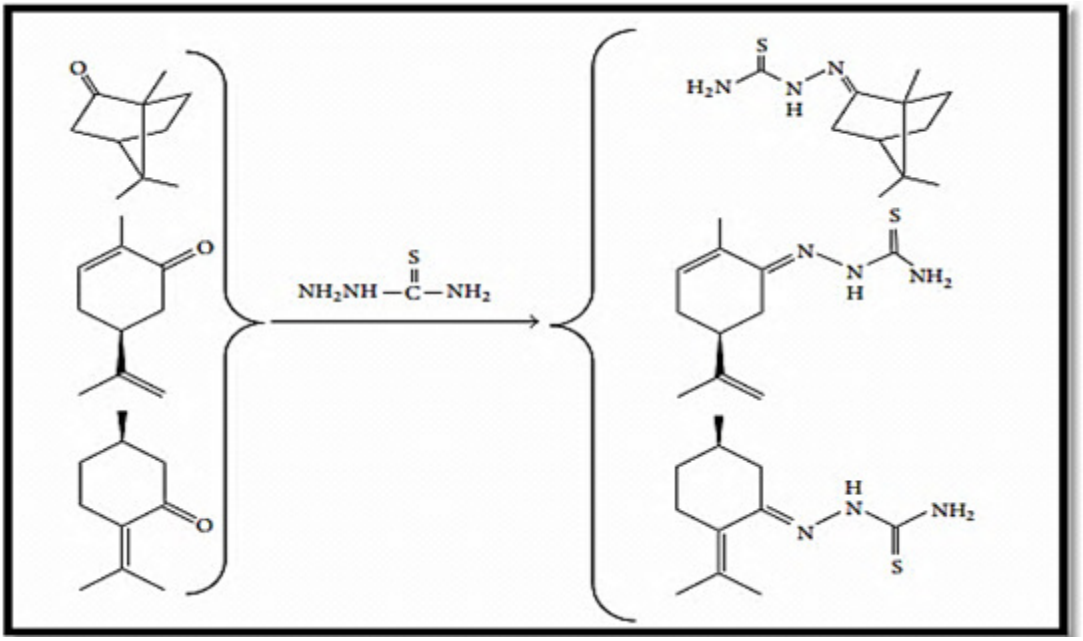


Fig. 14 : Structures of the H4methiocam and its Pt complex.



Scheme 9 : Synthesis of complexes of pd(II).



Scheme 10 : Synthesis of the Monoterpenic Tiosemicarbazones.

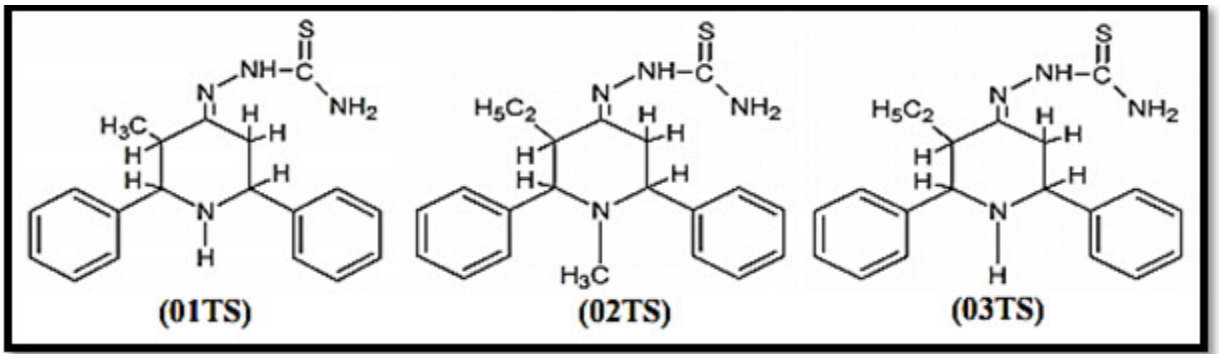
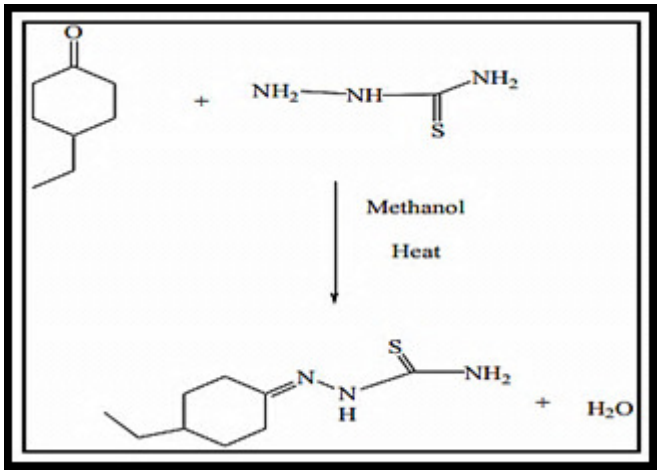


Fig. 15 : Structure of the inhibitors.



Scheme 11 : Synthesis of (ECHTSC).

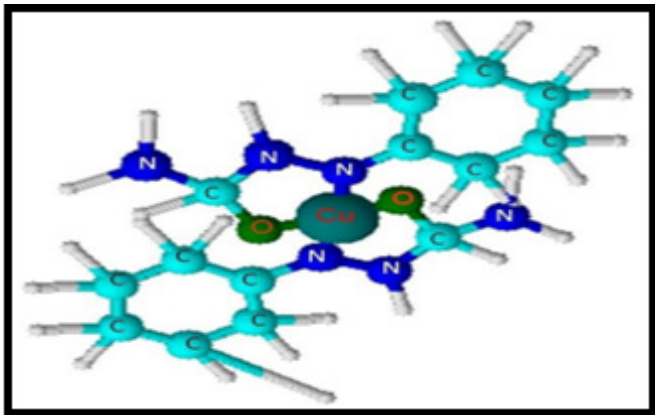


Fig. 16 : Structure of Cu (II)-complex.

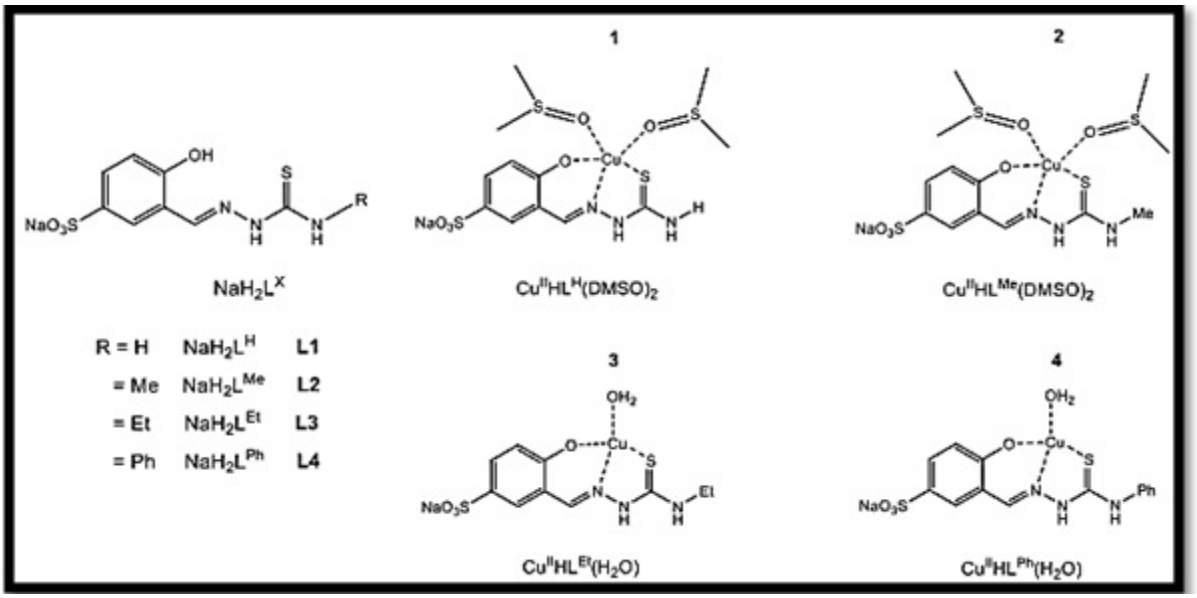


Fig. 17 : Structure ligands (L^{1}–$L^{4)}$and their Copper (II) complexes.

were investigation by C.H.N.S, ^{1}HNMR, FTIR and mass spectra. The general formula of Mn (II) complexes $[Mn(L)_2X_2]$, where X = NO_3^-Cl$^-$ based on FTIR and EPR spectral data, complexes octahedral geometry (Fig. 13).

Yanmin *et al* (2017) were synthesized some copper complexes by the reaction of (ketones with thiosemicarbazide or diazanyl pyridine) and then coordination of thiosemicarbazones or diazanyl pyridines with Copper (II). The complexes were investagation by FTIR, ^{1}HNMR and screened for their cytotoxicity against HeLa, Bel-7404 (human liver carcinoma) and 293T cell lines. Based on measurements result, Cu (II)-complexes show anti-proliferative activity against the tested cancer cells (Scheme 8).

Phan Thi *et al* (2018) were synthesized the new Pt(II)-complex by reaction Pt(II) ion with ligand [camphor 4-methylthiosemicarbazone] and investigation by ^{1}H-NMR and FTIR study. The DATA of the measurements showed a formula of $[Pt(C_{12}H_{20}N_3S)_2]$ and four coordinate. When studying anti-cancer activity, the Pt(II)-complex showed its ability to inhibit Hep-G2 and RD cancer cells with IC50 values of 7.74 and 7.61 ìg/mL. These datashowed the complex role of bio-medical applications (Fig. 14).

Ryan *et al* (2018) were synthesized derivatives thiosemicarbazones by reaction 4-ethyl-3-thiosemicarbazide with benzaldehyde. Coordination thiosemicarbazones with pd(II) characterized by X-ray diffraction. The bio-logical activity of the complexes were investigated against two types of fungi and two types of bacteria (Scheme 9).

Idouhli *et al* (2018) were studied the inhibitory effect of some Monoterpenicthiosemicarbazones on (steel

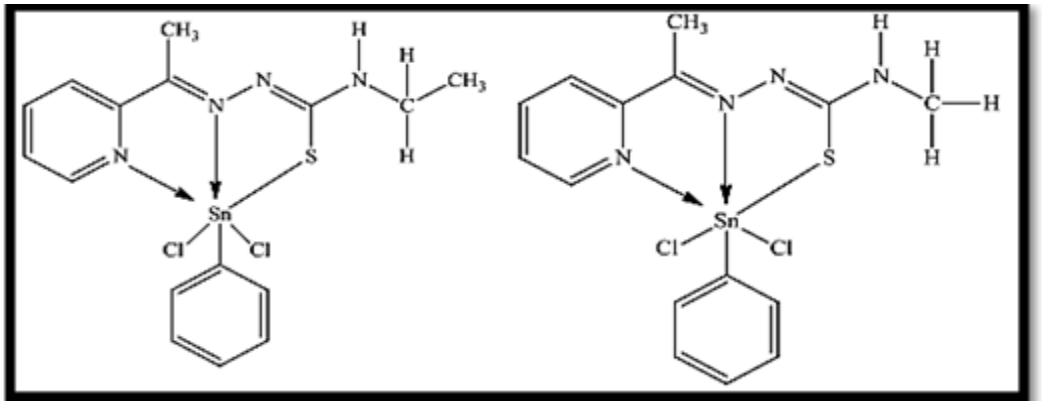
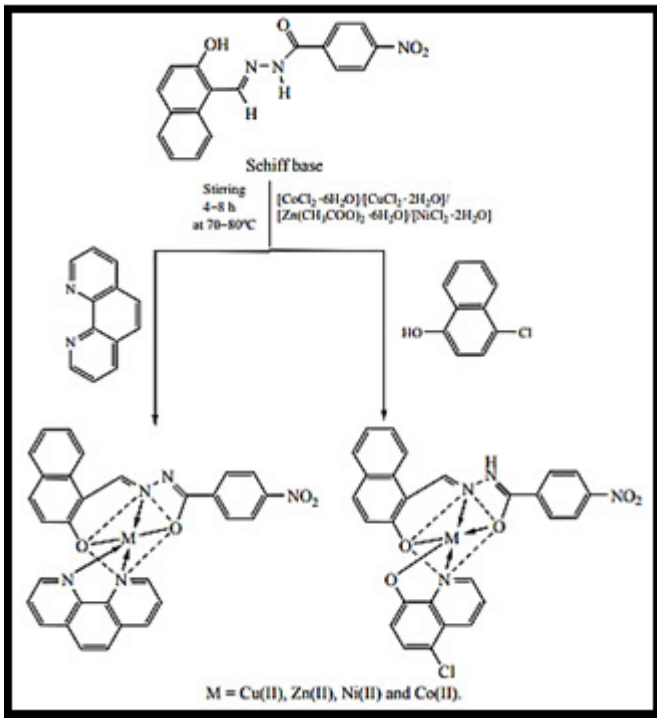


Fig. 18 : Structure of inhibitors.



Scheme 12 : Structure of [M(L)Phen] and [M(L)Oxine] complexes.

corrosion) in 1M HCl solution. Increased inhibition activity by increasing the inhibitor concentration and also with increasing temperature (293-323 K). The obtained results showed that the adsorption of inhibitor on the steel surface obeys Langmir's adsorption model and thermodynamic parameters such as enthalpy and activation energy were determined (Scheme 10).

Shanmuga *et al* (2018) were studied the acid corrosion inhibition technique of mild-steel in 1N sulfuric acid by a part of alkyl substituted 2,6-diphenyl piperidin-4-one with thiosemicarbazone by quantum chemical calculations, electrochemical AC impedance measurements, weight loss process and potentiodynamic polarization studies. Results show that substituted [γ-2,c-6-diphenylpiperidin-4-ones] with thiosemicarbazone act as perfect corrosion inhibitors and their inhibition efficiency increase with the addition of inhibitors. For all the three types of alkyl substituted 2,6-diphenyl pipieridin-4-one with thiosemicarbazone, the inhibition efficiency raised to increment in the inhibitor concentration and the performance of the three inhibitors are 01TS> 02TS> 03TS (Fig. 15).

Lakshmi *et al* (2018) were studies the corrosion inhibition of iron (Fe) metal in acidic medium (0.5N HCl) in the presence of synthesized4-ethylcyclohexanone thiosemicarbazone (ECHTSC) by the weight loss method. The protective layer of thiosemicarbazone on Fe surface was confirmed by scanning electron microscopy (SEM). The activity of inhibition, surface coverage and corrosion rate were evaluated at various concentrations of thiosemicarbazone in 0.5N hydrochloric acid (Scheme 11).

Dhanwe *et al* (2018) were synthesized copper (II) complexes with semicarbazone derivatives of [cyclopentanone(C_5), cyclohexanone(C_6), cycloheptanone(C_7) and cyclooctanone(C_8)] and investigated by M.P, UV-Vis, FTIR, ^{1}HNMR spectroscopy and Mass study. The molar conductance measurement of Cu-complexes appear nonelectrolytic nature. The Cu-complexes were appearing more activity against bacterial than free ligands. The cycloaactanon Cu-complex (CC_8) was used as a precursor for the prepare of nano-particles and was studies by XRD analysis and UV-Vis spectra (Fig. 16).

Ganga *et al* (2018) were synthesized mixed Ligand Complexes Derived from Semicarbazone Schiff Base and Heterocyclic Ligands [M(L)Phen] and [M(L)Oxine], where [M = Co(II), Ni(II), Cu(II), and Zn(II)], L = ((2-hydroxynaphthalen-1-yl)methylene)-4-nitrobenzohydrazide, Phen = 1,10-phenanthroline and Oxine = 8-hydroxy chloroquinoline and investgeted by FT-IR, UV-Vis, mass spectra and TG-DTA data. Morphology and crystallinity of the complex [Ni(L)Phen] were studied by scanning electron microscopy. Biological active of the ligand and its complexes has been studies

against four type of bacterial (Scheme 12).

Michal *et al* (2018) were used EPR spectroscopy to determine the properties of poly-poly crystalline Copper (II)-complexes containing sodium 5-sulfonate salicylaldehyde thiosemicarbazones possessing (H, CH_3, C_2H_5, or ph) substituent at the terminal -N. Mixing of pro-ligand solutions with Cu (II) ions resulted in the formation of (1:1)(M:L) in complex. The ligands and their complexes have been appearing moderate to strong antimicrobial activity (Fig. 17).

Nur *et al* (2018) were used thiosemicarbazone derivatives and its tin(IV) complexes in this study were 2-acetylpyridine 4-ethyl-3-thiosemicarbazone dichlorophenyltin (Sn(HAcETSc)PhenCl$_2$) and 2-acetylpyridine 4-methyl-3-thiosemicarbazone dichlorophenyltin (Sn(HAcMTSc) PhenCl$_2$). All these title complexes were investigated by FTIR and NMR. The efficiency of the synthesized complexes as a corrosion inhibitor of mild steel in (1.0 M)HCl was investigated by losing to weigh at different concentrations, 1, 2 and 3 mM and at a temperature range of 30–60°C. The thiosemicarbazone ligand and its tin(IV) complexes affirmed the hypothesis where the inhibitor efficiency tends to increase as the inhibitor concentration increases, indicating used as a corrosion inhibitor for mild steel (Fig. 18).

ACKNOWLEDGEMENT

The authors express their sincere appreciation to the Department of Chemistry for the continuous giving throughout the research period.

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