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A Review On Organotin(Iv) Thiosemicarbazone Complexes, Synthesis, Characterization And Biological Activity

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A B S T R A C T

Organotin(IV) complexes recently have been receiving great attention due to their stability with a unique structure, physical and chemical properties. There are many applications, the organotin(IV) can be used as catalysts, antifouling agents, UV- and heat stabilizers, anticorrosion, anticancer and antimicrobial activities. This review summarizes the synthesis methods, characterization and biological activities of organotin(IV) thiosemicarbazones derivatives with their activities as anticancer and antimicrobial agents.

1. INTRODUCTION

Initially, chemistry of thiosemicarbazone derivatives was discovered in the 1960s and show an important class of NNS containing donor ligands with formula ($R^1R^2C^2{=}N^3{-}N^2(H){-}C^1(=S)N^1R^3R^4$). These heterocyclic compounds are able to bind readily with transition and non-transition metals. This leads to different structural binding which contribute in the development of the coordination chemistry. Last decades, thiosemicarbazones and their organotin(IV) complexes play a role in the pharmacological industry as antiviral, antimicrobial, antitumor, antioxidant, and also show interest in agricultural applications[1-5].

Synthesis of Organotin(IV) Thiosemicarbazone Complexes:

Organotin(IV) complexes can obtain using organotin halides in non-aqueous medium such as methanol, ethanol, acetone, benzene, and n-hexane due to these compounds have a hydrolysable nature in aqueous solutions[6].

Affan *et al.* (2011) synthesized three organotin(VI) complexes of the type $MeSnCl_2$, $PhSnCl_2$ and Ph_2SnCl with ligand 2-benzoylpyridine-*N*(4)-cyclohyxylthiosemicarb- azone (HBPCT). The prepared ligand and its complexes were characterized using ^1H NMR, UV-Vis, FTIR, XRD, CHN analyses and molar conductivity. These characterizations approved that the ligand coordinated through thiolate-S, azomethine-N and pyridine ring-N to the Sn(VI) to form octahedral geometry Figure 1 [7].

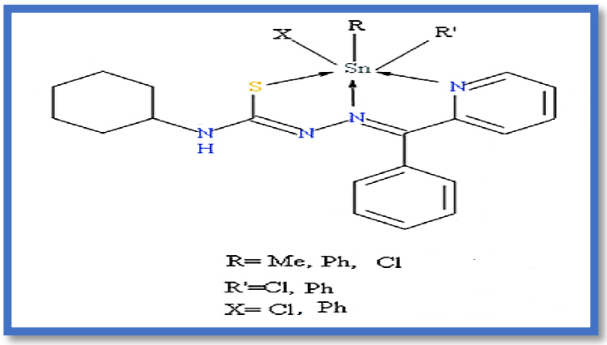

Fig. 1: Diorganotin(IV)chloride with 2-benzoylpyridine-*N*(4)-cyclohyxylthiosemicarbazone (HBPCT) complexes

Also, Salam *et al.* (2012) studied the preparation of 2-hydroxyacetophenone-2-methylphenylthiosemicarbazone (H_2dampt) from the reaction between 2-methylphenylisothiocyanate with hydrazine hydrate and 2-hydroxyacetophenone in absolute methanol. FTIR spectroscopy, molar conductivity and (^1H, ^{13}C, and ^{119}Sn) NMR suggested that H_2dampt connected to the tin(IV) in dinegative tridentate to form a five-member ring chelate [8].

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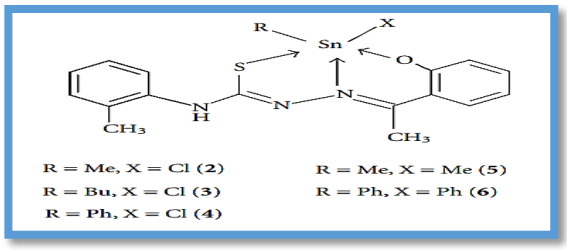

Fig.2: Organotin (IV)-2-2-hydroxyacetophenone-2-methylphenylthiosemicarbazone complexes

Six organotin(IV) complexes were prepared of the type $RSnCl(L_1)$, $RSnCl(L_2)$ (when R= alkyl or phenyl) of new thiosemicarbazone derivatives.

Ligands have been formed by reaction both 2,3-dihydroxybenzaldehyde and 2-hydroxy-5-methylbenzaldehyde separately with 4-methylthiosemicarbazide in ethanol to produce 2,3-dihydroxybenzaldehyde -N(4)-methylthiosemicarbazone (H_2DDTM) and 2-hydroxy-5-methylbenzaldehyde-N(4)-methyl thiosemicarbazone (H_2DMMT) respectively. Spectroscopy studies, elemental analysis and X-ray crystallography assumed that these ligands connected to tin(IV) as dinegative tridentate chelating ligands ONS, thus, the coordination number of tin(IV) is five as shown in Figure 3 [9].

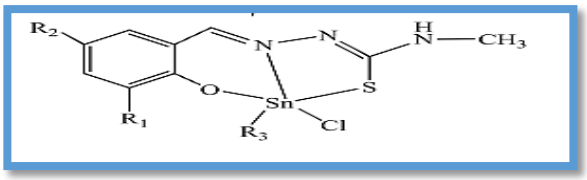

Fig.3: Organotin (IV)- 2,3-dihydroxybenzaldehyde-N(4)-methylthiosemicarbazone and 2-hydroxy-5-methylbenzaldehyde -N(4)- methylthiosemicarbazone complexes

Organotin(IV) thiosemicarbazone complexes can be synthesized by the condensation reaction between pyruvic acid thiosemicarbazide H_2pt and R_2SnCl_2, where R= Me, Ph. The structural formula of these compounds was confirmed by X-ray analysis, NMR, elemental analysis and IR. Theses characterizations suggested that the tridentate and octahedral geometry were for the prepared complexes Figure 4 [10].

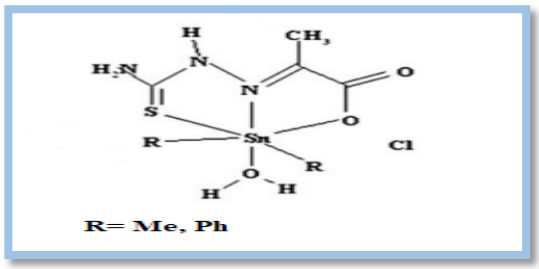

Fig. 4: Diorganotin(IV) pyruvic acid thiosemicarbazone complexes

On 2015 Rosenani *et al.* synthesized 5-bromo-2-hydroxybenzaldehyde-4,4-dimethylthiosemicarbazone (H_2L) by reaction of 5-bromo-2-hydroxybenzaldehyde with 4,4-dimethylthiosemicarbazide in an ethanolic solution. A new mono-organotin(IV) compounds have been formed with the ligand H_2L and characterized by CHN-analysis, UV-Vis., FTIR, and (^{1}H, ^{13}C, ^{119}Sn) NMR spectroscopic studies. The ligand is coordinated to tin(IV) *via* ONS-donor atoms and NMR studies confirmed pentadentate coordination in all complexes as shown in Figure 5 [11].

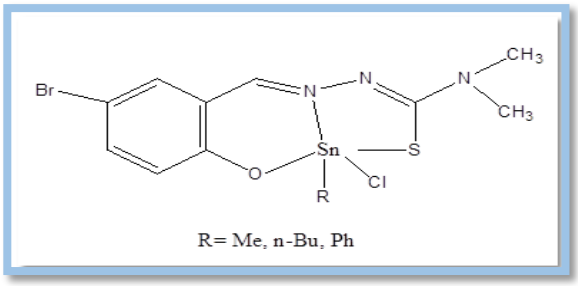

Fig. 5: Mono-organotin(IV)- 5-bromo-2-hydroxybenzaldehyde-4,4- dimethylthiosemicarbazone complexes

Huedo *et al.* (2018) reported a new structural form of diorganotin(IV) compounds by preparing new complexes of ligand diacetyl-2-thiosemicarbazone-3-(3-hydroxy-2-naphthohydrazone). The ligand was synthesized by the condensation of diacetyl-2-thiosemicarbazone and 3-hydroxy-2-naphthohydrazide in absolute ethanol with drops of concentrated HCl. Tetradentate ligand, octahedral geometry was suggested of the prepared compounds Figure 6.[12]

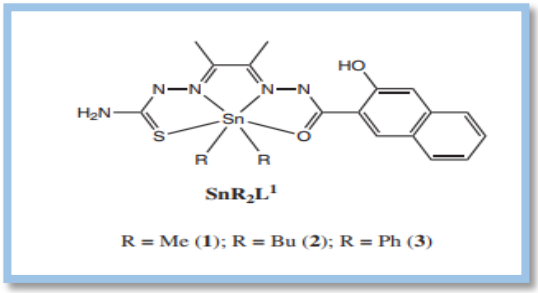

Fig. 6: Diorganotin(IV) complexes of diacetyl-2-thiosemicarbazone-3-(3-hydroxy-2-naphthohydrazone)

Another diorganotin (IV) complexes were synthesized by Singh *et al.* (2016) by the condensation of (2-hydroxyphenyl)(pyrrolidine-1-yl)methanone with phenylthiosemicarbazide in ethanol solution as a reaction medium. The prepared ligand 2-hydroxyphenyl (pyrrolidine-1-yl)methanone phenylthiosemicarbazone and its diorganotin(IV) complexes were characterized by elemental analysis, molar conductivity, molecular weight determination and spectral studies. These may assume that the ligand connected through-OSN leading to the formation of a penta-coordination around Sn(IV) in these complexes Figure 7.[13]

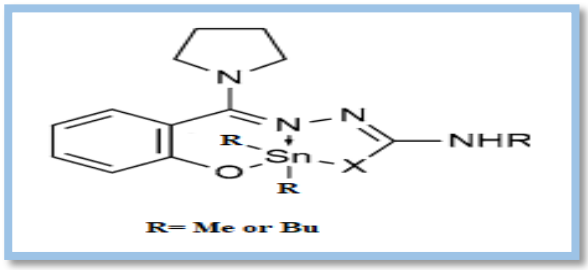

Fig. 7: Diorganotin(IV)-2-hydroxyphenyl(pyrrolidine-1-yl)methanone phenylthiosemicarbazone complexes

Mendes *et al.* (2008) reported the synthesis, characterization, antimicrobial and cytotoxic activity of n-butyltin(IV) trichloride with 2-pyridineformamide-derived thiosemic- arbazones. Octahedral geometry was proposed for the prepared complexes Figure 8. [14]

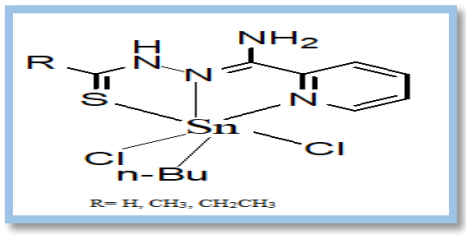

Fig. 8: *n*-Butyltin(IV) trichloride-2-pyridineformamide-derived thiosemicarbazones comprlexes

While, Singh *et al.* prepared tridentate ligand ONS 2-hydroxyacetophenone thiosemicarbazone to form a 5-coordinated complexes with diorgano tin(IV) chloride in a trigonal bipyramidal geometry Figure 9.[15] There is no meaning of this sentence, the authors need to re-write this sentence to be more consistent.

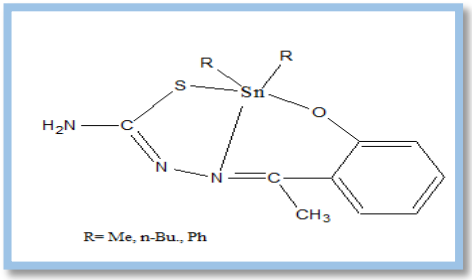

Fig.9: Diorganotin(IV) 2-hydroxyacetophenone thiosemicarbazone complexes

Khan *et al.* (2016) have been formed a new ligand (5-bromo-2-hydroxybenzaldehyde-N(4)-methylthiosemica-rbazide) using the condensation of 5-bromo-2-hydroxybenzaldehyde and 4-methyl-3-thiosemicarbazide in ethanolic solution. Elemental analysis, FTIR, electronic and ^{1}H, ^{13}C NMR spectroscopy showed that the newly synthesized ligand connected through ONS to Sn(IV) to form pentacoordinated geometry Figure 10. [16] Anti-tumor results showed that these compounds could be promising and active against A549, MCF-7 and HCT-116 cancer cell lines.

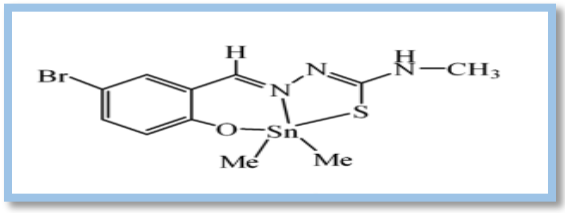

Fig. 10: Dimethyltin(IV)-5-bromo-2-hydroxybenzaldehyde-N(4)-methylthiosemicarbazone complex

Anticancer Activity:

The extraordinary achievements of cisplatin and carboplatin (Figure 11) *in vitro* proliferative activity and *in vivo* experiments have been encouraging the researchers to investigate a new non-platinum complexes. This is to use as anticancer therapies with optimistic results, and with low or no side effects [6,17]. The feature of the metal ion and the organic ligand are versatile and the ability of the metal ions to lose electrons to be cationic in the biological systems are preferred. These positively charged species interact and coordinate with the opposite charged molecules and rich of electrons e.g. DNA and proteins to become soluble in the biological systems. [18-20]

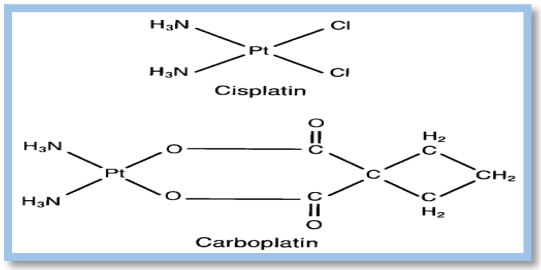

Fig.11: Structure of cisplatin and carboplatin complexes

Among these non-platinum compounds, organotin complexes have taken a significant attention as an active antitumor against different types of human cancer cell lines [21]. For this researchers have been synthesized organotin(IV) complexes and study their anticancer activity as drugs[22-28].

The biological activity of organotin(IV) compounds was discovered in 1929 [29]. I could not actually understand this sentence, what the authors want to say here? I kindly ask the authors to re-write this sentence to be understandable.

Thiosemicarbazones and their tin(IV) complexes the subject of this review attracted attentions due to their significant flexibility as a multi-dentate donor molecules through the sulfur as well as azomethane nitrogen atoms. Due to the ability of thiosemicarbazones to behave as N,S-multidentate ligands and the opportunity to modify the binding properties through the insertion of other atoms/molecules in the backbone structure *i.e.* (phenolic or pyridyl halves). This is characterized them as versatile ligands with the excellent

bending ability with most transition and non-transition elements such as Sn. These ligands are also have the flexibility and ability to bind either in neutral or in deprotonated forms [30, 31]

The higher activity of organotin(IV) complexes may be due to the number and type of R- groups [R= alkyl or aryl] that attached to the Sn(IV) core. Many studies show that tri-organotin complexes have higher activities compared with di and mono-organotin and this can increase their ability to bind with the DNA of tumors and damage them. [32-34]. Also, the high cytotoxic activity can be assigned to the bulky aryl groups attached to Sn-center compared to alkyl groups [27]. These bulky groups increase the lipophilicity nature of the complexes by catalyzes π-π interaction between the metal ions and the lipid membrane that surrounds the cancer cells.[35] Literatures describe the mechanism of apoptotic (programmed cell death) using organotin(IV) complexes. This is happened by the interaction between organotin(IV) moieties to cellular proteins and bind with DNA, leading to the death of pathogenic cells. The intracellular metabolism of the phospholipids of the endoplasmic reticulum is modified by tin(IV) complexes when they bind to the phosphodiester of DNA. [36]

Gielen in 1996 and 2003 reported the influence of R-groups of various organotin(IV) complexes as a metallodrugs against colon and breast cancer cell lines [37,38]

The structure of organotin(IV) compounds to be potent as antitumor drugs is distinguished by: (i) the availability of coordination positions around tin atom, (ii) the relative stability of Sn–S thione and (iii) slow hydrolytic decomposition of Sn—S bond besides lipophilic and hydrophilic properties of the complexes. [39,40]

Devi *et al.* reviewed different types of organotin(IV) complexes which they have therapeutic activities against different tumor cell lines depends on the reduction potential of these complexes. The review summarized that the number and type of R-groups attached to the tin, the type of ligands, geometry of the prepared complexes and the presence of heteroatom (OH, X, NH, …etc.) in the backbone of the complexes. This leads to assist to modulate the lipophilicity character to increase the absorption of the compounds. Also, preventing cell recognition and adhesion process and interfering in carbohydrate-protein interaction and helps in transport at the molecular levels all these are crucial in cancer growth. [41, 42].

Biological Activities:

Organotin(IV) have been investigated due to their structural features and potent biological activity [43]. Inorganic tin(IV) compounds can be considered non-poisonous or with very little toxicity towards mammals and organisms while organotin(IV) compounds reveal varying biological activities [44,45]. Antibiological drugs work by preventing bacteria from multiplying or killing them without harming the host.

The complexes of organotin(IV) with thiosemicarbazones derivatives are more efficient against all organisms than semicarbazones complexes, so as a result sulfur is more active as antimicrobial drugs than oxygen, as suggested by Tandon [46-47].

Currently, the inhibition activity of organotin(IV) thiosemicarbazones is studied and found that the geometry has no impact role on their biological activity. It has been observed whether the ligands coordinated around Sn(IV) in trigonal bipyramidal or octahedral geometry have good antimicrobial activity and degrade DNA[47-49].

The free ligands found to be inactive or lower activity than their complexes against the tested bacteria and fungi, which indicates that metalation increases the antimicrobial activity [30,39,44]. Also, studies show that the highest inhibition activity of complexes attributed to the type and size of R-groups attached to the Sn atom. The screening results showed that the activity of R-groups can be arranged from the higher to the lower activity on the microorganisms as follows: Ph > Bu > Me. It can be noted that the complexes with bulkier phenyl groups exhibited greater effectiveness compared to other alkyl groups against the tested microorganisms, due to the rise of lipophilicity character of the complexes. The increases in the lipophilicity nature of the coordinated metal ion resulting from reducing the polarity character of it in which enable them to penetrate through the lipoid layers of the microorganisms' membranes[13, 51-54].

Antimicrobial activity of some of organotin(IV) thiosemicarbazones complexes showed promising results as drugs and also found to be competing with reference drug such as ampicillin and miconazole that used for treatment of bacteria and fungi respectively.[51]

More than 3.8 billion years microscopic organisms have survived on the earth and characterized with the extreme variety in genetic and metabolic. They are consist about 50% of the living biomass, one component of the ecosphere in which play an essential role in the maintenance and sustainability of environments. These microorganisms adapt with various environmental challenges such as pressure. The disease-causing microorganisms are particularly vulnerable to man's selfishness for survival, who has sought to deprive them of their habitat by using antimicrobial agents. But these disease-causing microorganisms develop resistance against antimicrobial agents which consider a serious threat to infectious disease management globally [55-58]

Mechanism action of antimicrobial agents can be classified depends on the structure of microorganisms or on the function that they are affect, *i.e.* inhibition the synthesis of cellular wall, of ribosome function, of nucleic acid synthesis,

of folic acid metabolism or of cell membrane function. Thus, the mechanisms of inhibition depend on which pathways are inhibited by the antimicrobial drugs and whether the organisms can change those mechanisms. Resistance of antimicrobial drugs by microorganisms can be either intrinsic or acquired immunity. Because of the chemical nature of the drugs or the structure of the microbial membranes, microorganisms with intrinsic or natural resistance either do not have functional sites for drugs or have low penetrability to them. While, microorganisms with acquired resistance in which naturally susceptible gains ways of being not affected by various means [59-61].

Conclusion:

In conclusion, thiosemicarbazone derivatives are well known as Schiff base ligands exist either in neutral or anionic forms also characterized with bidentate, tridentate and polydentate ligands due to the presence of sulfur, nitrogen or even oxygen in some cases as donor atoms. Organotin(IV) thiosemicarbazone complexes are synthesized in different molar ratio by refluxing the calculated amount of the prepared thiosemicarbazone derivatives and organotin(IV) compounds in a suitable solvent.

Spectroscopic results show that the coordination of thiosemicarbazone derivatives through azomethine-N, thiolate-S, and O-in some cases forming either hexa- or penta-coordinate Sn(IV). Various applications of organotin(IV) complexes have been noted because of their ability to be diverse in coordination behaviors. The presence of R-groups on tin(IV) atom affects the anticancer and antimicrobial activities of the complexes. It has been noted that the existence of aryl group in the complexes plays a role in the biological activity owing to its ability to increase lipophilic character of Sn(IV) complexes and form stable structures with the biological molecules. There are several complexes containing thiosemicarbazones as ligands which can be effective and even better than traditional antitumor and antimicrobial drugs.

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إستعراض عن تحضير وتشخيص والفعالية البايولوجية لمعقدات القصديرالعضوي (IV) لمشتقات الثايوسمي كاربازون

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الخلاصــة:

حظيت معقدات القصدير العضوي (IV) في السنوات الأخيرة بأهتمام كبير نظرا لأستقرارها ببنية فريدة وخصائص كيميائية وفيزيائية. بعض الأمثلة لها تطبيقات واسعة ومختلفة مثل العوامل المحفزة والمضادة للأشعة فوق البنفسجية ومثبتات الحرارة ومقاومة التآكل وفعاليتها كعوامل مضادة للأمراض السرطانية والفعالية البايولوجية وغيرها من المجالات. هذا الملخص يستعرض طرق التحضير والتشخيص والفعالية البايولوجية لمعقدات القصدير العضوي (IV) مع مشتقات الثايوسيمي كاربازون ودراسة نشاطها البايولوجي.

الكلمات المفتاحية: القصدير العضوي (IV)، الثايوسيمي كاربازون, مضاد للسرطان, مثبتات, ثنائي القصدير العضوي (IV), ثلاثي القصدير العضوي (IV).