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Synthesis, characterization and Antimicrobial Activity of Schiff base complexes of Sn(IV)

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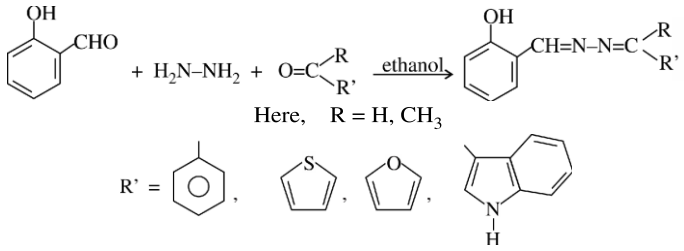
ABSTRACT

M(IV) complexes of Sn with Schiff base derived from mixed azines were synthesized, characterization by several techniques, including elemental analysis (C,H,N,S), molar conductance, measurements, electronic, IR and NMR spectral studies. A few representative complexes have also been screened for their bactericidal and fungicidal activity and found to be quite active in this respect against bacteria (Escherichia coli, staphylococcus, pseudomonas capacicola), Fungi (Fusarium oxysporum, Aspergillus flavus and Rhizoctonia Phaseoli). The metal complexes were found to be more potent as compared to the ligands.

Graphical Abstract:



Synthesis of the Schiff base ligand

Keywords: Organotin (IV) complexes, mixed azines, NOH donor, antibacterial and antifungal activity.

INTRODUCTION

The chemistry of organotin (IV) compounds derived from ligands containing nitrogen and oxygen donor [1] atom has attracted much attention in last few year [2],a fact that underlines the increasing number of publications dealing with this class of compounds [3], organotin [4] have become leading commercial [5] organometallics due to their use in polyvinyl stabilization and also as biocides [6]. Now they are widely [7] used as fungicides [8] and miticides as well as marine antifouling agents. Schiff base is a compound [9] that contain carbon nitrogen double bond C=N Schiff base is a good Lewis base and widely used as a ligand because the nitrogen atom in C = N is capable of donating [10] its loan pair of electron to form

coordinate covalent bond with a metal [11]. The aim of present study was to prepare, characterize and determine the antibacterial and antifungal activity of organotin (IV) Schiff base complexes. Schiff base derived from salicylaldehyde and hydrazine with aldehyde /ketones.

MATERIALS AND METHODS

All chemical and solvents were highest purity from Fluka. Tin was determined as tinoxide by decomposing the compound with fuming nitric and sulphuric acids and precipitating the form of tin hydroxide by the addition of ammonium hydroxide. C and H analysis were carried out on a Coleman 5612 analyzer. Nitrogen was estimated by Kjeldal's method. Sulphur was estimated as $BaSO_4$ by Messenger's method. Molecular weight determined by Rast camphor method. Infrared spectra were recorded on a Perkin Elmer, FT-IR- SP-2Spectrophotometer in KBrpellets. ^{1}H NMR spectra were recorded in DMSO – d_6 at 400 MHZ using tetramethylsilane (TMS) as an internal standard. ^{13}C NMR spectra were recorded on a 90 MHZ Joel spectrometer using dry DMSO as the solvent and TMS as an internal standard. ^{119}Sn NMR spectra were recorded using 90 MHZ Joel spectrometer operating 22.7 MHZ and TMT (tetramethyltin) standard are used.

Synthesis of the Schiff base: Synthesis of Schiff base of mixed azine derived from salicylaldehyde and hydrazine with aldehyde / ketones. A calculated amount of hydrazine is added with salicylaldehyde and benzaldehyde / acetophenone/thiophene-2 aldehyde / 2-furyl methyl ketone/ indole-3 carboxaldehyde / fur furaldehyde /2-acetylthiophene in1:1:1 molar ratio where ethanol is taken as the reaction medium and refluxed for 4-5hours. On cooling were further purified by recrystalization from acetone.

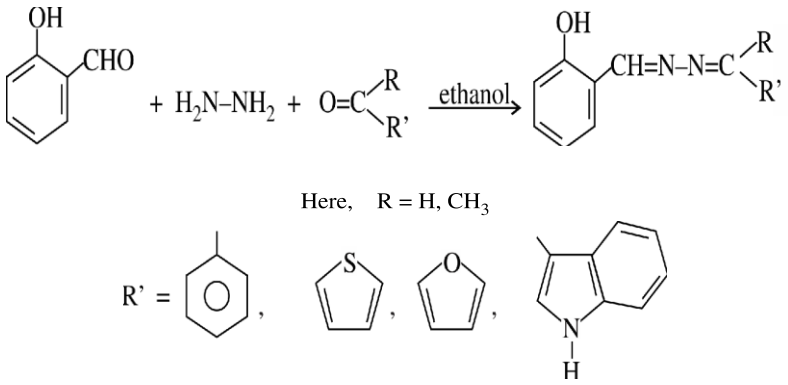


Fig. Synthesis of the Schiff base ligand

Synthesis of organotin (IV) complexes: The reaction of dibutyltin with mixed azines derived from salicylaldehyde and hydrazine with aldehyde / ketones in the medium of benzene in 1:2 molar ratio takes place of follows:

$$Bu_2SnO + 2N\overset{\frown}{O}H \xrightarrow[benzene]{1:2} Bu_2Sn(N\overset{\frown}{O})_2 + H_2O$$

where NOH represents the donor system of Schiff bases.

RESULTS AND DISCUSSION

All the newly synthesized complexes are colored solids and are soluble in DMSO, DMF and common organic solvents. The elemental analyses (C, H, N and S) and M.P. data of the Schiff base ligands (Table 1).

Table 1. Elemental analyses, color, state and M.P. (in °C)

S.No.	Ligand	Colorand state	M.P (°C)	Elemental analysis (%)			
				C Found (calcd.)	H Found (calcd.)	N Found (calcd.)	S Found (calcd.)
1	$C_{14}H_{12}N_2O$ (L1 H)	Lemon yellow Shiny crystal	152	74.82 (74.98)	5.24 (5.39)	12.36 (12.49)	-
2	$C_{15}H_{14}N_2O$ (L2 H)	Yellow Shiny crystal	163	75.66 (75.76)	5.82 (5.92)	11.66 (11.76)	-
3	$C_{12}H_{10}N_2OS$ (L3 H)	Greenish brown Shiny crystal	166	62.44 (62.59)	4.24 (4.36)	12.04 (12.16)	13.76 (13.92)
4	$C_{13}H_{12}N_2O_2$ (L4 H)	Reddish brown Solid	181	68.32 (68.41)	5.22 (530)	12.10 (12.27)	-

Spectral Studies: The I.R spectra of tin derivatives showed two sharp bands at ~1635 $cm^{-1}$and 1600 cm^{-1} in all these complexes, compared to one at ~ 1620 cm^{-1} in ligands may be attributed to *v* (C=N) group. The band at 1635 cm^{-1} in the tin complexes indicates the coordination of the azomethine nitrogen to the tin atom whereas the other one (~ 1600 cm^{-1}) is due to uncoordinated azomethine group. The phenolic *v*(C-O) and *v*(N-N) respectively at ~1260 and ~970 cm^{-1} are also shifted to higher frequency region as a result of complex formation.

^{1}H NMR spectra of the ligands shows the OH proton signal at δ ~ 12.60 ppm. However, in the complexes, this signal disappear which shows the chelation thought the deprotonated alcoholic oxygen. One additional signal appears in the spectra of the complexes in the butyl protons. The ligands show a complex multiplet signal in the region δ 7.75-7.10 ppm for the aromatic protons and it remains almost at the same position in the spectra of metal complexes. The proton signal of the azomethine moiety (-HC=N-) is observed at δ 8.80 ppm in the spectra of the ligands and it gets shifted downfield in the spectra of tin complexes, which may be due to coordination of azomethine nitrogen to tin atom.

Antibacterial activity: All the synthesized ligands and tin complexes were screened in vitro applying paper disc plate method technique for antibacterial activity against gram positive and gram negative bacteria {Escherichia Coli (-),Pseudomonas capacicola (-) and Staphylococcus aureus (+)} at the both 500 ppm and 1000 ppm concentration. Streptomycin was used for as a reference compound for antibacterial activities. The antibacterial studies suggest that the Schiff base are biologically active and their tin complexes showed significantly enhanced antibacterial activity against microbial strains in comparison to the free ligands. It has been observed that the tin complexes showed increased zone of inhibition the bacterial strain (table 2) as compared to ligands.

Table 2. Antibacterial screening data of the ligands and their Tin (IV) complexes inhibition zone (mm)after 24 h (conc.in ppm)

Compounds	Diameter of inhibition zone (mm)					
	E.Coli(–)		S. aureus (+)		P. capacicola (–)	
	500 ppm	1000 ppm	500 ppm	1000ppm	500ppm	1000ppm
L1H	3	5	4	8	3	4
$Bu_2Sn(L1)_2$	7	11	8	12	6	8
L2H	4	7	7	8	4	5
$Bu_2Sn(L2)_2$	8	12	9	13	7	9
L3H	5	8	8	9	4	7

$Bu_2Sn(L3)_2$	9	14	9	15	7	10
L4 H	3	7	6	8	4	6
$Bu_2Sn\ (L4)_2$	7	13	7	14	5	8
Streptomycin	15	18	15	17	10	14

Antifungal activity: The antifungal activities of the newly synthesized compounds were evaluated by the radial growth method. In fungicidal screening, theorganism used are fusarium oxysporum, Aspergillus flavus and Rhizoctonia Phaseoli. The antifungal activity of Schiff base ligands and its Sn (IV) complexes was evaluated by measuring the zone of growth inhibition against test fungal organism. The medium were used potato, dextrose and agar medium (composition: potato slices -200g, dextrose-20g, agar-agar-15g and distilled water-1000 mL).The compounds were directly mixed with the medium in different concentration (Table 3).

Table 3. Antifungal screening data of the ligands and their Tin (IV) complexes (percent inhibition after 96 h at 25±2°C)

Compounds	**Diameter of inhibition zone (mm)**					
	Organism F. oxysporum		**Organism A.Flavus**		**Organism R.Phaseoli**	
	50 ppm	**100 ppm**	**50 ppm**	**100ppm**	**50ppm**	**100ppm**
L1H	24	35	28	39	26	36
$Bu_2Sn(L1)_2$	40	51	45	52	40	48
L2H	25	36	27	37	24	36
$Bu_2Sn(L2)_2$	41	52	42	56	40	46
L3H	31	40	33	42	31	39
$Bu_2Sn(L3)_2$	44	55	48	63	48	56

APPLICATIONS

The formation of the coordination compounds can be used as the prospective antibiotic agents against some known pathogenic organism and can be used as marketed drugs.

CONCLUSIONS

The newly synthesized Schiff base and Schiff base tin (IV) complexes were found to be highly active against some of the antibacterial and antifungal species. The activity is significantly increased on coordination.

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