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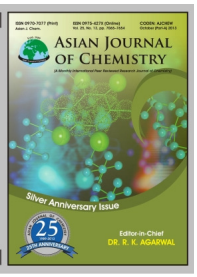

Synthesis, Structure Elucidation and Study of Antimicrobial Activity of Stannic(IV) Derivatives

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Six new organotin(IV) compounds with general formulae $R_2Sn(OL)_2$ and R_3SnOL where R = CH_3, *n*-C_4H_9, C_6H_5, OL = 4-oxypiperidine were synthesized by the reaction of R_2SnCl_2 and R_3SnCl with stoichiometric amount of 4-hydroxypiperidine in dry ethanol/methanol. These compounds have been characterized by using multinuclear (^1H, ^{13}C, ^{119}Sn) NMR and FT-IR spectroscopy. The spectroscopic results revealed that all the diorganotin(IV) compounds possess trigonal-bipyramidal geometry in solution and a tetrahedral environment around the tin atom in non-coordinating solvents, has been proposed for the triorganotin(IV) compounds. All the synthesized compounds were tested *in vitro* against a number of microorganisms to assess their biocidal activity. The biological activity of these compounds against various strains of bacteria and fungi has revealed promising activity in comparison with Amphotericin B and miconazole as reference drugs. LD_{50} values of some of the tested compounds showed cytotoxicity.

Key Words: Organotin, Biological activity, Organotin(IV) alkoxide.

INTRODUCTION

Organotin compounds are of current interest due to increase in their biological activities as well as industrial and agricultural applications[1,2]. There are now more organometallic compounds of tin in commercial use than any other element, covering a wide spectrum of biological and non-biological applications[3,4]. In general, the biochemical activity of organotin compounds is influenced remarkably by the coordination number of tin atoms [5]. Among organotin(IV) compounds, organotin(IV) carboxylates have been studied extensively for their antimicrobial, antiinflammatory and antituberculosis activities[5]. In view of well known application and biological activities of organotin derivatives we are continuously involved in synthesis and characterization of organotin(IV) carboxylates[6-8].

Recently, we have investigated organotin compounds containing alkoxide ligand 4-hydroxypiperidine with di- and triorganotin moieties and the result of this study are reported here in.

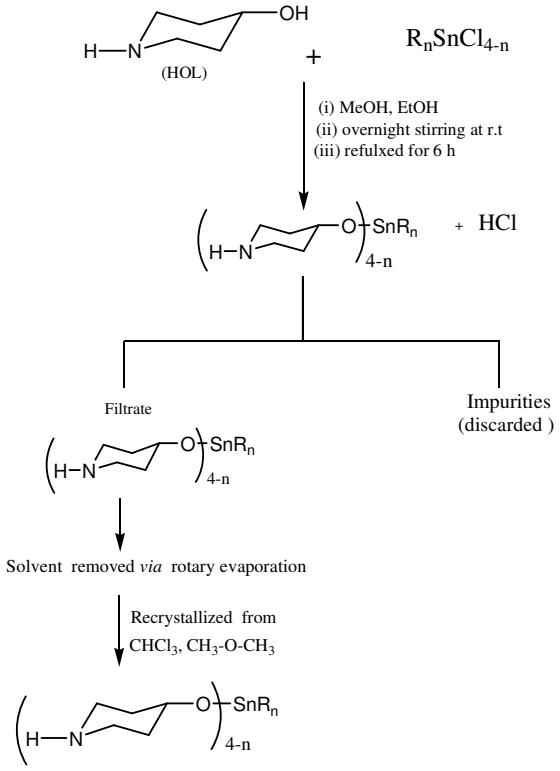
EXPERIMENTAL

Analytical grade organotin chlorides (dimethyltin dichloride, trimethyltin chloride, triphenyltin chloride, diphenyltin dichloride, tri-*n*-butyltin chloride, di-*n*-butyltin dichloride) were purchased from Aldrich. The ligand, 4-hydroxypiperidine was obtained from ACROS. Organic solvents used for synthesis like methanol, ethanol, toluene, chloroform, acetone and dimethyl sulfoxide were used of Merck, Germany and dried *in situ* using standard procedures[9-11].

All other chemicals were of analytical grade and used without further purification.

Melting points were determined in a capillary tube using electrothermal melting point apparatus model MPD Mitamura Ricken Kogyo (Japan). The infrared absorption spectra were recorded as KBr pallets on a Bio-Red Merlin (USA) spectrometer in the frequency range 4000-400 cm^{-1}. Multinuclear NMR (^1H, ^{13}C, ^{119}Sn) spectra were recorded on Bruker 300 MHz, FT-NMR Spectrometer using $CDCl_3$ as internal reference, (δ ^1H $CDCl_3$ = 7.27, δ ^{13}C $CDCl_3$ = 77.6 ppm). The elemental analysis was made on CHN-932 Elemental Analyzer Leco Corporation (USA).

General procedure for synthesis of organotin(IV) alkoxides: R_2SnOL_2 and R_3SnOL, complexes of organotin compounds were prepared by the reaction of ligand 4-hydroxypiperidine (1 g, 9.8 mmol) with organotin chlorides, R_2SnCl_2 (0.5 g, 4.9 mmol) and R_3SnCl (1 g, 9.8 mmol) in dry methanol (25 mL) in two-neck round bottom flask (250 mL) equipped with water condenser (**Scheme-I**). The reaction mixture was refluxed for 6 h and stirred over night at 273 K. The resulting product was obtained by evaporating the mixture under reduce pressure on rotary evaporator. The solid product thus obtained was recrystallized from chloroform.



Scheme-I: Synthesis of organotin derivatives

RESULTS AND DISCUSSION

Synthesis of organotin(IV) alkoxides: Organotin(IV) compounds of general formula $R_2Sn(OL)_2$ and R_3SnOL were prepared from the corresponding di- and triorganotin chlorides by refluxing the stoichiometric amount of ligand HOL in (1:2) and (1:1) ratios in dry methanol for 6 h. They are air stable and soluble in common organic solvents. To predict structures and other properties all compounds have been characterized by various analytical techniques IR, NMR (^{1}H, ^{13}C, ^{119}Sn) and were tested for their biological activity. The general **Scheme-I** depicted the procedure adopted for the synthesis of organotin(IV) alkoxides. The physical data are given in the Table-1.

Infrared spectroscopy: Vibrational data for synthesized compounds are given in Table-2. The coordination mode of the ligand, 4-hydroxypiperidine towards the di- and triorganotin(IV) moieties can be deduced by comparing infrared spectra of free ligand with synthesized compounds.

The explicit feature in the spectra of all complexes is absence of a band in the region 3602 cm^{-1}, which appears in the free ligand as the ν(O-H) vibration thus indicating metal-ligand bond formation through this site[12]. Diagnostically important IR bands are ν(O-H), ν(N-H) ν(Sn-C) and ν(Sn-O). The N-H stretching frequencies, 3500-3200 cm^{-1} in the complexes approximately remain the same in comparison with free ligand (Table-2). The disappearance of ν(O-H) stretching band and appearance of absorption band for the Sn-O bond in the range of 480-420 cm^{-1} reflect the formation of complexes[13]. The band Sn-C bond appears in the region 625-520 cm^{-1} [14]. These results suggest that the tin atom in both of the di- and triorganotin complexes adopt a tetrahedral geometry in solid state[15].

TABLE-2
INFRARED DATA (cm^{-1}) OF ORGANOTIN(IV) COMPLEXES WITH 4-HYDROXYPIPERIDINE

Compound. No.	ν(O-H)	ν(N-H)	ν(Sn-C)	ν(Sn-O)
LOH	3602	3292	–	–
$Me_2Sn(OL)_2$ (I)	–	3400	550	463
Me_3SnOL (II)	–	3416	559	428
n-$Bu_2Sn(OL)_2$ (III)	–	3460	567	480
n-Bu_3SnOL (IV)	–	3369	518	450
$Ph_2Sn(OL)_2$ (V)	–	3441	579	446
Ph_3SnOL (VI)	–	3416	523	466

^{1}H NMR spectroscopy: ^{1}H NMR spectra of all the compounds have been recorded on 300 MHz Bruker, FT-NMR spectrometer. The characteristic resonance peaks in the ^{1}H NMR spectra for complexes (**II, III, IV and VI**) and free ligand HOL are listed in the Table-3. The assignment is made on the basis of peak multiplicity, intensity pattern, integration, tin satellites and by comparing with literature values[16]. The integration of spectra showed good agreement with the composition of compounds. In the spectrum of free ligand, OH and NH protons resonate, respectively at 4.94 and 4.11 ppm as singlets, while the signals for equatorial and axial protons of the ligand moiety with their distinct multiplicities have been assigned according to the literature[16]. The analogous pattern of the signals, rather similar positions of the protons has been observed for the investigated tin compounds[17,18].

In trimethyltin(IV) compound (**II**) CH_3 protons appeared as sharp singlet at 0.50 ppm with well resolved $^2J[^{119}Sn, ^1H]$ coupling constant 68 Hz, which propose tetrahedral geometry around tin in solution[17]. The *n*-butyltin (IV) derivatives (**III , IV**) show a clear triplet due to a methylenic group, which appears at 1.69 and 1.70 ppm, with well defined $^2J[^{119}Sn, ^1H]$ couplings constants of 82 and 63 Hz, respectively[19-21]. The $^2J[^{119}Sn, ^1H]$ couplings constants of 82 Hz suggest more than four coordination around tin in solution whereas the $^2J[^{119}Sn, ^1H]$ couplings constants of 63 Hz support tetrahedral geometry around tin in solution for compound (**IV**)[19-21]. The terminal methyl group of (**III**, **IV**) appears at 0.95 ppm as triplet showing $^3J[^1H, ^1H)$ coupling constants at 7.4 and 7.2 Hz, respectively. The methylenic protons (CH_2) of *n*-butyl derivatives (**III, IV**) show multiplets in the range of 1.40-1.50 ppm. In triphenyltin

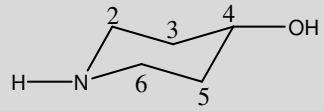
TABLE-1
PHYSICAL DATA OF ORGANOTIN(IV) COMPLEXES WITH 4-HYDROXYPIPERIDINE

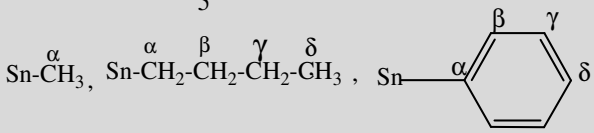
Compound No.	m.f.	m.p. (ºC)	Yield (%)	Elemental analysis calcd. (found) (%)		
				C	H	N
$Me_2Sn(OL)_2$ (**I**)	$C_{12}H_{26}N_2O_2Sn$	250	57	41.3 (40.9)	7.4 (5.9)	8.0 (7.8)
Me_3SnOL (**II**)	$C_8H_{19}NOSn$	56	49	37.0 (36.8)	7.1 (6.9)	5.3 (4.7)
n-$Bu_2Sn(OL)_2$ (**III**)	$C_{18}H_{38}N_2O_2Sn$	100	65	49.8 (49.1)	8.7 (7.9)	6.4 (5.7)
n-Bu_3SnOL (**IV**)	$C_{17}H_{31}NOSn$	64	64	53.1 (52.1)	8.7 (7.0)	3.6 (2.9)
$Ph_2Sn(OL)_2$ (**V**)	$C_{22}H_{20}N_2O_2Sn$	230	68	55.8 (55.5)	6.3 (5.9)	5.9 (5.5)
Ph_3SnOL (**VI**)	$C_{23}H_{25}NOSn$	98	70	61.3 (57.4)	5.5 (4.3)	3.1 (9.6)

TABLE-3
^{1}H NMR DATA OF ORGANOTIN(IV) COMPLEXES WITH 4-HYDROXYPIPERIDINE[a,b,c]

Proton No.	LOH	Me_3SnOL (**II**)	*n*-$Bu_2Sn(OL)_2$ (**III**)	*n*-Bu_3SnOL (**IV**)	Ph_3SnOL (**VI**)
O-H	4.94 (s)	–	–	–	–
N-H	4.11 (s)	4.11 (s)	4.11 (s)	4.11 (s)	3.74 (s)
4H	3.72 (m)	3.50 (m)	3.73 (m)	3.60 (m)	3.73 (m)
3H[e]	1.94 (m)	1.73 (m)	1.95 (m)	1.98 (m)	1.94 (m)
2H[e]	3.11 (m)	3.46 (m)	3.10 (m)	3.12 (m)	3.11 (m)
6H[e]	3.07 (m)	3.47 (m)	3.07 (m)	3.16 (m)	3.08 (m)
5H[e]	1.88 (m)	1.72 (m)	1.74 (m)	1.72 (m)	1.90 (m)
3H[a]	1.44 (m)	1.33 (m)	1.43 (m)	1.39 (m)	1.48 (m)
2H[a]	2.65 (m)	2.86 (m)	2.65 (m)	2.62 (m)	2.63 (m)
6H[a]	2.58 (m)	2.52 (m)	2.58 (m)	2.59 (m)	2.58 (m)
5H[a]	1.37 (m)	1.37(m)	1.36 (m)	1.36 (m)	1.37 (m)
α	–	0.50 (s) [68]	1.69 (t 6.2) [82]	1.70 (t 6.4) [63]	7.28 (m)
β	–	–	1.40-1.50 (m)	1.40-150 (m)	7.29 (m)
γ	–	–	1.40-1.50 (m)	1.40-1.50(m)	7.47 (m)
δ	–	–	0.95 (t 7.4)	0.95 (t 7.2)	7.69 (m)

(a) Chemical shifts (δ) in ppm, $^{2}J[^{119}Sn, ^{1}H]$ are listed in square brackets.

(b) 

(c) 

. (d) a = axial H, (e) equatorial H.

derivative (**VI**) a complex multiplet pattern is observed in the range 7.28-7.69 ppm due to phenyl groups of the complexes. The C-Sn-C bond angle has been calculated by using Lockhart's and Holeèek *et al* equations for compound (**II, IV, VI**) and are 128º, 134º and 115º, respectively. The bond angles calculated support the tetrahedral geometry around tin for triorganotin(IV) derivatives and five-coordinated geometry for diorganotin(IV) derivatives[16,17, 22].

^{13}C NMR spectroscopy: ^{13}C NMR chemical shift values obtained for free ligand (HOL) and respective di- and triorganotin(IV) derivatives are listed in the Table-4. The number of signals observed for ligand corresponds to the presence of expected magnetically non-equivalent carbon atoms[12]. In the spectrum of ligand carbon (4) resonates at 68.41 ppm, while carbons (2, 6) being magnetically equivalent were observed as a single speak at 44.40 ppm. The assignment of ^{13}C signal for carbons (3, 5) observed at 36.00 ppm. In the ^{13}C spectra of triphenyltin and tri-*n*-butyltin and methyltin derivatives resonance due to all carbon atoms are visible and appeared in expected regions as shown in the Table-4[19].

The signals of R groups attached to Sn atom, were assigned by comparing with literature values[24,25]. The magnitudes of $^{n}J[^{119}Sn, ^{13}C]$ couplings constant in compounds (**II, III, VI**) are also observed for trimethyl, triphenyl and tri-*n*-butyltin(IV) derivatives with the $^{1}J[^{119}Sn, ^{13}C]$, 532, 625 and 598 Hz, respectively. By using Holeèek equations[23] C-Sn-C bond angles were calculated fond 123º, 134º and 115º for (**II, III, VI**), respectively[23], which corresponds to tetrahedral geometry in $CDCl_3$ for triphenyl- (**VI**) and five coordinated environment around the tin atom for trimethyl- (**II**) and di-*n*-butyltin(IV) alkoxides in (DMSO).

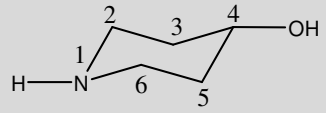
^{119}Sn NMR spectroscopy: ^{19}Sn NMR spectroscopy is most convenient technique used to study organotin(IV) derivatives in solution and in the solid state . The ^{119}Sn nucleus has natural abundance of 8.7 % and is 25 times more sensitive than ^{13}C nucleus. ^{119}Sn NMR spectra were recorded in $CDCl_3$ solution, chemicals shift data is presented in the Table-4. The δ(^{119}Sn) chemical shifts of compounds (**II, IV, VI**) are observed at -84, -48 and -32 ppm, reflecting tetrahedral geometries around the tin moiety in solution[27-29]. The δ(^{119}Sn) chemical shift of compound (**III**) is observed at -158 ppm, suggesting a five coordinated geometry around tin.

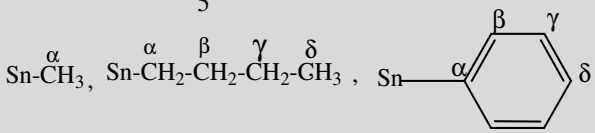
Antibacterial activity: The synthesized compounds were screened for antibacterial activity against *Escherichia coli, Bacillus subtilis, Shigella flexenari, Staphylococcus aureus, Pseudomonas aeruginosa and Salmonella typhi* bacterial strains, using the agar well diffusion method[30]. Imipenum was used as a standard drug and the wells (6 mm in diameter) were dug in the media with the help of a sterile metallic borer. Two to eight hours old bacterial inoculums containing approximately 10^4-10^6 colony-forming units (CFU)/mL were spread on the surface of a nutrient agar with the help of a sterile cotton swab. The recommended concentration of the test sample (200 mg/mL in DMSO) was introduced into the respective wells. Other wells supplemented with DMSO and reference antibacterial drug served as negative and positive controls, respectively. The plates were incubated immediately at 37 ºC for 20 h. The activity was determined by measuring the diameter of the inhibition zone (in mm), showing complete inhibition. Growth inhibition was calculated with reference to the positive control. The results of the antibacterial activity so obtained are collected in the Table-5.

TABLE-4
^{13}C NMR DATA OF ORGANOTIN(IV) COMPLEXES WITH 4-HYDROXYPIPERIDINE

Carbon No.	LOH	Me_3SnOL (**II**)	*n*-$Bu_2Sn(OL)_2$ (**III**)	*n*-Bu_3SnOL (**IV**)	Ph_3SnOL (**VI**)
4	68.4	65.6	68.2	68.9	68.6
3, 5	36.0	34.4	36.2	36.4	36.1
2, 6	44.40	42.8	44.4	49.4	44.4
α	–	3.2 [532]	25.8 [598]	23.7	140.5 [625]
β	–	–	26.94 [34]	37.9 [28]	136.2 [48]
γ	–	–	26.5 [85]	36.4	129.0 [29]
δ	–	–	13.7	14.0	128.1 [14]
δ^{119}(Sn)	–	-32.4	-158.3	- 48.4	-84.8

(a) Chemical shifts (δ) in ppm,^1J[^{119}Sn,^{13}C] are listed in square brackets.

(b) 

(c) 

The free ligand show non-significant activity against *Pseudomonas aeruginosa*. The investigated compounds (**III-VI**) show significant activity against *Eschericha coli*, *Bacillus subtilis, Salmonella typhi, Staphylococcus aureus* and gives low activity against *Shigella flexenari* and *Pseudomonas aeruginosa*. Compound (**II**) observed to have low activity against *Shigella flexenari* and non significant activity against *Staphylococcus aureus a*nd *Pseudomonas aerugiosa*, while compound (**I**) shows good activity against *Staphylococcus aureus, Pseudomonas aeruginosa* and low activity against *Shigella flexenaria* and *Salmonella thphi* and non-significant activity against *Eschericha coli and Bacilius subtilis.*

Antifungal activity: The synthesized organotin derivatives were also tested for antifungal activity against six different human, animal and plant pathogens, namely *Trichophyton longifusus, Candida albicans, Aspergillus flavus, Microsporum canis, Fusarium solani and Candida glaberata* by using the tube diffusion test[31]. Amphotericin B and miconazole were used as standard antifungal agents for the comparison test. Stock solutions of pure compounds (200 g/ mL) were prepared in sterile DMSO. Sabouraud dextrose agar was prepared by mixing Sabouraud (32.5 g), glucose agar (4 %) and agar-agar (4 g) in 500 mL of distilled water followed by dissolution at 90-95 ºC on a water bath. The media (4 mL) was dispensed into screw-capped tubes and autoclaved at 121ºC for 15 min. Test compounds (66.6 mL) were added from the stock solution to nonsolidified Sabouraud agar media (50 ºC). The tubes were then solidified at room temperature and inoculated with 4 mm diameter portion of inoculums derived from a 7-days old respective fungal culture. For nonmycelial growth, an agar surface streak was employed. The tubes were incubated at 27-29 ºC for 7-10 days and growth in the compound containing media was determined by measuring the linear growth (in mm) and growth inhibition with reference to the respective control.

The fungicidal screening data for the tested compounds are listed in Table-6. The free ligand 4-hydroxypiperidine shows moderate activity against *Trichophyton longifusus, Aspergillus flavus* and *Fusarium solani.* While compounds (**I, VI**) show significant activity against all the tested fungi. Compounds (**I, III, V**) show significant activity against *Trichphyton longifusus, Aspergillus flavus, Microsportum canis, Fusarium solani, Candida glabrata* and *Candida albicans*.

Cytotoxicity: For all the synthesized complexes, including ligand, LD_{50} values were measured and compared with standard drug etoposide to evaluate their toxicity. The cytotoxicity data have been collected by the brine-shrimp lethality bioassay method[31] and the results are summarized in the Table-7. It has been noted that free ligand has a positive lethality, while compounds (**V, VI**) show cytotoxicity at the highest dose only.

TABLE-5
ANTIBACTERIAL ACTIVITY DATA OF ORGANOTIN(IV) COMPLEXES WITH 4-HYDROXYPIPERIDINE[a,b,c]

Name of bacterium	Zone of inhabitation (nm)							Ref.
	HOL	$Me_2Sn(OL)_2$ (**I**)	Me_3SnOL (**II**)	*n*-$Bu_2Sn(OL)_2$ (**III**)	*n*-Bu_3SnOL (**IV**)	$Ph_2Sn(OL)_2$ (**V**)	Ph_3SnOL (**VI**)	
Escherishia coli	–	10	–	19	42	18	20	25
Bacillus subtilis	–	11	–	17	42	17	21	26
Shigella flexenari	–	12	14	10	12	13	12	24
Staphlococcus aureus	–	16	10	19	–	18	–	17
Pseudomona aeruginosa	11	16	10	13	14	11	–	17
Salmonella typhi	–	13	21	17	–	21	22	21

[a]–, Show no activity. [b]Std durg, imipenum. [c]Concentration: 100 µg/mL of DMSO.

TABLE-6
ANTIFUNGAL ACTIVITY DATA OF ORGANOTIN(IV) COMPLEXES WITH 4-HYDROXYPIPERIDINE[abc]

Name of fungus	Inhibition (%)								
	HOL	$Me_2Sn(OL)_2$ (**I**)	Me_3SnOL (**II**)	n-$Bu_2Sn(OL)_2$ (**III**)	n-Bu_3SnOL (**IV**)	$Ph_2Sn(OL)_2$ (**V**)	Ph_3SnOL (**VI**)	Std. Drug µg	MIC (µg/mL)
Trichophyton longifusus	50	–	–	–	20	80	80	Miconazole	70
Candida albicans	–	–	–	–	80	–	80	Miconazole	110.8
Aspergillus flarus	50	–	–	–	90	80	–	Amphotericin B	20
Microsporium canis	20	–	–	–	–	80	80	Miconazole	98.4
Fusarium solani	50	–	–	–	90	80	80	Miconazole	73.25
Candida glaberata	–	–	–	–	80	–	80	Miconazole	110.8

[a]MIC, minimum inhibitory concentration. [c]Incubation period = 7 days. [b]Concentration, 200µg/ mL of DMSO.

TABLE-7
CYTOTOXICITY DATA OF ORGANOTIN(IV) COMPLEXES WITH 4-HYDROXYPIPERIDINE[abc]

Compounds	LOH	$Me_2Sn(OL)_2$ (**I**)	Me_3SnOL (**II**)	n-$Bu_2Sn(OL)_2$ (**III**)	n-Bu_3SnOL (**IV**)	$Ph_2Sn(OL)_2$ (**V**)	Ph_3SnOL (**VI**)
LD_{50}	34.6983	65.0235	–	36.1322	0.9271	–	–

[a]MIC, minimum inhibitory concentration, [c]Incubation period = 7 days. [b]Concentration, 200 µg/ mL of DMSO.

Similarly, compounds (**I, III, IV**) show a positive lethality and compound (**II**) exhibits no cytotoxicity.

Conclusion

The elemental analyses results of all the synthesized compounds are comparable with calculated and found values of carbon and hydrogen atoms, which confirmed the formation and purity of compounds. Spectroscopic studies (FT-IR, multinuclear NMR) as well support the formation of products. Furthermore, results obtained by different spectroscopic techniques are consistent with the proposed structures, which probably lead to the conclusion that the synthesized compounds in non-coordinating solvent ($CDCl_3$) are monomeric with tetrahedral geometry and in the coordinating solvent (DMSO) are five coordinated.

Biological activity data collected for the investigated compounds show that triorganotin(IV) alkoxides have good activities compared to their diorganotin(IV) analogues. Particularly, tri-*n*-butyltin(IV) complex found to be more effective and potent against all the tested fungi and also showed more activity against few medically important bacteria.

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