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Synthesis, Characterization and Biological Applications of Organotin(IV) Derivatives of 2-(2-Fluoro-4-biphenyl)propanoic Acid

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A series of organotin(IV) derivatives with the general formula $R_{4-n}SnL_n$ (where n = 1 or 2, R = Me, Et, *n*-Bu, Ph and L = 2-(2-fluoro-4-biphenyl)propanoate) have been synthesized and characterized by infrared, multinuclear NMR (^{1}H, ^{13}C, ^{119}Sn), ^{119m}Sn Mössbauer spectroscopy and mass spectrometry. Screening tests of these compounds showed that they are highly active against various bacteria and fungi.

Introduction

For the last three decades, a wide range of established and potential applications of tin and organotin compounds based on their structural and biological properties have been well documented in the literature[1–4]. Among organotin compounds, organotin carboxylates have received more attention due to their structural and biological importance, especially antitumor and anticancer activities[5–10]. With this and our continuing interest in the synthesis, characterization and crystal structures of organotin carboxylates[11–20] in mind, we have prepared new organotin derivatives of 2-(2-fluoro-4-biphenyl)propanoic acid, commonly known as flurbiprofen. Flurbiprofen is a non-steroidal, anti-inflammatory drug (NSAID) commonly marketed as Ansaid (frequently used in daily life)[21,22].

Experimental

Instruments

IR spectra were recorded on a Hitachi 270-50 spectrometer (Japan) in KBr/CsBr disks. ^{1}H, ^{13}C and ^{119}Sn NMR spectra were recorded on a Brucker 250 ARX (Germany). Mass spectra were recorded on MAT 8500 Finnigan equipment. ^{119m}Sn Mössbauer spectra were obtained with a constant acceleration microprocessor

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controlled spectrometer (Cryophysics Ltd., Oxford U.K.). A barium stannate source was used at room temperature and samples were packed in Perspex disks and cooled to -193 °C. Isomer data are relative to SnO_2.

Synthesis

Since most organotin precursors and their carboxylate derivatives are air and moisture sensitive, all glassware used was completely dried at 140 °C. All reactions were carried out under argon in dried solvent[23]. The chemicals were of analytical grade and used without further purification.

Flurbiprofen, 2.44 g (10 mmol), and R_2SnO (5 mmol), R_3SnOH (10 mmol) or $(R_3Sn)_2O$ (5 mmol) were refluxed in 100 mL of toluene in a 250 mL flask equipped with a Dean-Stark funnel. The mixtures were refluxed for 4-6 h, and the binary azeotrope toluene/water was distilled off down to 75% of the initial solvent volume. The remaining solution was evaporated under vacuum. The solid residue formed was recrystallized from appropriate solvents (yield 70-85%). Physical data are reported in Table 1.

Table 1. Physical data of organotin(IV) 2-(2-fluoro-4-biphenyl) propanoate.

No.	Compound	M. Formula (M. Weight)	M. P. (°C)	Yield (%)	% C Calctd.(Exp.)	% H Calctd.(Exp.)
I	Me_3SnL	$C_{18}H_{21}O_2FSn$ (407)	127-9	75	53.07 (52.9)	5.16 (5.19)
II	Bu_3SnL	$C_{27}H_{39}O_2FSn$ (533)	89-90	67	60.79 (60.85)	7.32 (7.40)
III	Ph_3SnL	$C_{33}H_{27}O_2FSn$ (593)	135-7	80	66.78 (66.9)	4.55 (4.48)
IV	Me_2SnL_2	$C_{32}H_{30}O_4F_2Sn$ (635)	130-2	86	60.47 (60.37)	4.72 (4.66)
V	Et_2SnL_2	$C_{34}H_{34}O_4F_2Sn$ (663)	154-5	82	61.54 (61.73)	5.13 (5.22)
VI	Bu_2SnL_2	$C_{38}H_{42}O_4F_2Sn$ (719)	81-82	71	63.42 (62.8)	5.84 (5.96)
VII	Ph_2SnL_2	$C_{42}H_{34}O_4F_2Sn$ (759)	126-7	68	66.4 (66.52)	4.48 (4.39)

Results and Discussion

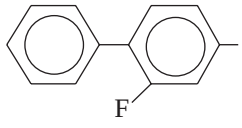
Preparation of Complexes

Triorganotin carboxylates were prepared by the reaction of ligand acid with an appropriate amount of $R_3SnOH/(R_3Sn)_2O$ in toluene in a 1:1/2:1 ratio, respectively.

$$CH_3CH(R')COOH + R_3SnOH \xrightarrow[4-6\ h\ reflux]{Toluene} R_3Sn(OCOCH(R')CH_3) + H_2O$$

$$2CH_3CH(R')COOH + (R_3Sn)_2O \xrightarrow[4-6\ h\ reflux]{Toluene} 2R_3Sn(OCOCH(R')CH_3) + H_2O$$

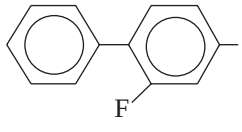
R = Me, n-Bu, Ph

R' = 

Diorganotin dicarboxylates were prepared by the condensation of ligand acid with diorganotin oxide in a 2:1 molar ratio.

$$2CH_3CH(R')COOH + R_2SnO \xrightarrow[4-6\ h\ reflux]{Toluene} R_2Sn(OCOCH(R')CH_3)_2 + H_2O$$

R = Me, Et, n-Bu

R' = 

Spectroscopy

Different instrumental techniques are used for the characterization of synthesized compounds. These include infrared, NMR, Mössbauer spectroscopy and mass spectrometry.

The main infrared spectral data are listed in Table 2. The assignments of ν(Sn−C) and ν(Sn−O) are consistent with the values reported in the literature[24]. An important feature of the infrared spectroscopy is the difference in $\Delta\nu$ between asymmetric (ν COO) and symmetric (ν COO) absorption frequencies. It is generally accepted that a value of $\Delta\nu$ less than 200 cm^{-1} indicates the bidentate nature of the ligand[24].

Table 2. Infrared data (cm^{-1}) of organotin(IV) 2-(2-fluoro-4-biphenyl)propanoate.

No.	ν $(COO)_{asym}$	ν $(COO)_{sym}$	$\Delta\nu$	ν (Sn-C)	ν (Sn-O)
LH	1690	1370	320	-	-
I	1596	1400	196	530	490
II	1588	1395	193	546	470
III	1600	1402	198	285	488
IV	1602	1405	157	525	483
V	1597	1403	194	528	470
VI	1594	1402	192	537	478
VII	1590	1400	190	283	493

The ^1H, ^{13}C and ^{119}Sn NMR data of the title compounds are given in Tables 3-5. All the proton NMR data are in good agreement with the expected structures. The proton resonances of the butyl and phenyl moieties of compounds II, III, VI and VII appear as complex patterns only the $^2J(^{119}Sn\text{-}^1H)$ coupling constant could be determined for compound II. The values of coupling constant ^2J $(^{119}Sn\text{-}^1H)$ for the case of methyl moiety for compound I and IV are in the range of a four-coordinated and five- or six- coordinated structure in solution, respectively[7].

To confirm the structure of the title compounds, ^{13}C and ^{119}Sn NMR data were also recorded for these compounds. Different resonances in ^{13}C NMR were assigned by comparison with values calculated from the incremental method[25].

Table 3. ^{1}H NMR data of organotin(IV) 2-(2-fluoro-4-biphenyl)propanoate^{a,b,c,d}.

No.	Aromatic Protons	CH	CH_3	Sn−R
I	7.23-7.58 (m, 8H)	3.79 (q, 1H)	1.56 (d, 3H)	0.58 (R = Me) (s, 9H, ^{2}J[59.6])
II	7.20-7.56	3.81 (q, 1H)	1.58 (d, 3H)	1.58 (R = Bu) (q, 6H, ^{2}J[54.5]) 1.31-1.38 (m, 12H) 0.92 (t, 9H)
III	7.12-7.55 (m, 8H)	3.93 (q, 1H)	1.61 (d, 3H)	7.6-7.74 (R = Ph) (m, 15H)
IV	7.22-7.56 (m, 16H)	3.87 (q, 2H)	1.58 (d, 6H)	1.01 (R = Me) (s, 6H, ^{2}J[73.5])
V	7.22-7.61 (m, 16H)	3.91 (q, 2H)	1.58 (d, 6H)	1.69 (R = Et) (q, 4H, ^{2}J[71.0]) 1.3 (t, 6H)
VI	(m, 16H) 7.21-7.59	3.76 (q, 2H)	1.56 (d, 6H)	1.58 (R = Bu) (q, 4H) 1.30-1.4 (m, 8H) 0.84 (t, 6H)
VII	7.12-7.59 (m, 16H)	3.83 (q, 2H)	1.64 (d, 6H)	7.64-7.8 (R = Ph) (m, 10H)

^{a}In $CDCl_3$ at 25^{o}C (40%) ^{b}Chemical shift (δ) in ppm ^{c}^{2}J[^{119}Sn, ^{1}H] in Hz

^{d}Multiplicity is given as s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet

Table 4. ^{13}C NMR data of triorganotin(IV) 2-(2-fluoro-4-biphenyl)Propanoate^{a,b}.

Carbon	L H	I Me_3SnL	II Bu_3SnL	III Ph_3SnL	IV Me_2SnL_2	V Et_2SnL_2	VI Bu_2SnL_2	VII Ph_2SnL_2
1	180.3	179.8	179.5	180.1	183.6	184.8	184.7	182.8
2	45.2	46.3	46.3	45.9	45.3	45.5	45.5	45.3
3	18.4	19.6	19.4	19.1	19.1	18.2	19.1	18.5
4	141.3 141.4(7)	143.8 144.0(7)	144.0 144.1(8)	142.9 143.1(7)	142.4 142.1(8)	142.4 142.5(8)	143.4 142.5(7)	143.4 143.6(7)
5	115.6 116.0(24)	115.4 115.8(23)	115.4 115.8(24)	115.0 115.4(24)	115.5 115.9(24)	115.5 115.9(24)	115.5 115.8(23)	115.6 116(24)
6	158.1 162(248)	158.1 162(248)	158.1 162(248)	157.6 161(248)	158.2 162(248)	158.4 162(249)	158.1 162(248)	158.1 162(248)
7	128.5 128.7(14)	127.6 127.8(14)	127.5 127.7(14)	127.2 127.4(13)	128.5 128.7(14)	128.1 128.3(14)	128.1 128.3(14)	128.4 128.7(13)
8	131.3(4)	131.0(4)	130.9(4)	130.5(4)	131.3(4)	131.2(4)	131.1(4)	131.3(4)
9	124.1(4)	124.0(3)	124.0(3)	123.6(3)	124.0(3)	124.0(3)	124.1(3)	124.2(3)
10	135.8(1)	136.2(1)	136.2(1)	135.7(1)	136.0(1)	136.0(1)	136.0(1)	136.0(1)
11,15	129.4(3)	129.4(3)	129.4(3)	129.5(2)	129.4(3)	129.4(3)	129.4(3)	129.4(3)
12,14	129.9	128.8	128.8	128.4	128.9(3)	128.9(3)	128.9(3)	128.9(3)
13	128.1	127.9	127.9	127.5	128.1	128.1	128.0	128.1
α	-	8.83[n.o.]	16.9[355]	136[640]	4.8[514]	19.3[566, 540]	25.5[570, 552]	141.4 [n.o.]
β	-	-	28.2	136.0[49]	-	9.2[37]	27.0[34]	130.8[62]
γ	-	-	27.4	128.9[63]	-	-	26.6[97]	129.5[96]
δ	-	-	14.1	130.2[13]	-	-	13.9[n.o.]	[n.o.]

^{a}In $CDCl_3$ at 298 K (40%); ^{b}chemical shift (δ) in ppm;^{n}J[$^{117/119}$Sn-^{13}C] and ^{n}J(^{13}C, ^{19}F) in Hz., n.o. = not observed.

Table 5. ^{119}Sn NMR data of organotin(IV) 2-(2-fluoro-4-biphenyl) propanoate[a,b].

No.	Compound	Chemical Shift	No.	Compound	Chemical Shift
I	Me_3SnL	139	V	Et_2SnL_2	– 144.6
II	Bu_3SnL	114.5	VI	Bu_2SnL_2	223
III	Ph_3SnL	– 95.1	VII	Ph_2SnL_2	140.8
IV	Me_2SnL_2	115.6	-	-	-

[a]In $CDCl_3$ at 298 K (40%). [b]chemical shift (δ) in ppm

In the trimethyl and tributyltin(IV) derivatives, the values of $^1J(^{119}Sn\text{-}^{13}C)$ are 400 and 355 Hz, respectively, which are typical of the pseudo-tetrahedral arrangement of R_3SnO configuration with the four-coordinating tin(IV) atom[26,27]. As expected, the ^1J value for triphenyltin derivatives is higher than that of the alkyl substituted[7,26,27]. In case the of diorganotin dicarboxylates, the geometry around tin could not be defined with certainty due to the fluxional behavior of the carboxylate oxygens in their coordination with the tin atom; however, earlier reports suggest a geometry in between penta- and hexa-coordination[28−30].

Table 6. Mössbauer data of organotin(IV) 2-(2-fluoro-4-biphenyl) propanoate.

No.	Compound	IS	QS	QS/IS
I	Me_3SnL	1.29	3.56	2.75
V	Et_2SnL_2	1.45	3.53	2.43
VI	Bu_2SnL_2	1.38	3.33	2.41

^{119}Sn NMR parameters are very useful for the determination of the coordination number of tin, its molecular geometry and stereochemistry. It is reported that in alkyltin carboxylates, the range for four-coordinate tin is about +200 to -60 ppm, five-coordinate tin from -90 to -190 ppm and six-coordinate tin from -200 to -400 ppm[29,30]. A single resonance at +139 ppm and +114.5 ppm for trimethyltin and tributyltin derivatives, respectively, is compatible with a tetrahedral geometry in solution[31]. In the triphenyltin derivative, the ^{119}Sn NMR chemical shift at -95.1 ppm is within the expected range reported for other triphenyltin carboxylates, e.g., Ph_3Sn(N-acetyl-L-phenylalaninato) -114.8 ppm and Ph_3Sn(N-acetyl-L-phenylalanylglycinato) -99.4 ppm, which correspond to the pseudo-tetrahedral configuration of the Ph_3SnO group[32].

The ^{119m}Sn Mössbauer data for three representative compounds are given in Table 5. Mössbauer spectral data are characterized by a single doublet, revealing the occurrence of only one type of tin atom in solid state. The tin atom of compound I (Q.S. = 3.56 mm/s) is obviously five-coordinate in solid state, whereas the QS/IS ratio greater than 2.1 suggests trans-octahedral geometry for compounds V and VI[33].

The main mass spectral data of the title compounds are in good agreement with the expected structure, and generally they have almost the same fragmentation pattern as reported in earlier reports[34−36]. Possible fragmentation patterns for both tri- and diorganotin compounds are given in Schemes 1 and 2, while the data are reported in Tables 7 and 8.

Table 7. Fragmentation pattern of triorganotin(IV) 2-(2-fluoro-4-biphenyl)propanoate.

Fragment	I Me_3SnL (m/z)	Intensity (%)	II Bu_3SnL (m/z)	Intensity (%)	III Ph_3SnL (m/z)	Intensity (%)
$[R_3SnOCOR']^+$	408	(n.o.)	534	(n.o.)	594	(n.o.)
$[R_2SnOCOR']^+$	393	37	477	2	517	21
$[RsnOCOR']^+$	378	18	420	8	440	13
$[R_2SnR']^+$	349	(n.o.)	433	18	473	(n.o.)
$[RSnR']^+$	334	4	376	(n.o.)	396	(n.o.)
$[R_3Sn]^+$	165	27	291	(n.o.)	351	100
$[R_2Sn]^+$	150	12	234	4	274	9
$[RSn]^+$	135	37	177	20	197	14
$[Sn/SnH]^+$	121	4	121	2	121	2
$[R'COOH]^+$	244	23	244	26	244	21
$[R']^+$	199	100	199	100	199	11
$[C_6H_5]^+$	77	8	77	3	77	6
$[R]^+$	15	(n.o.)	57	2	-	-

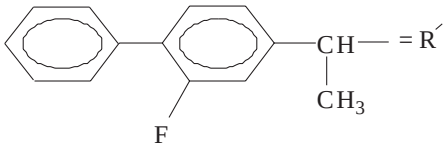
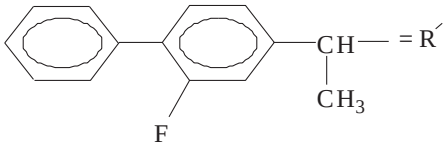
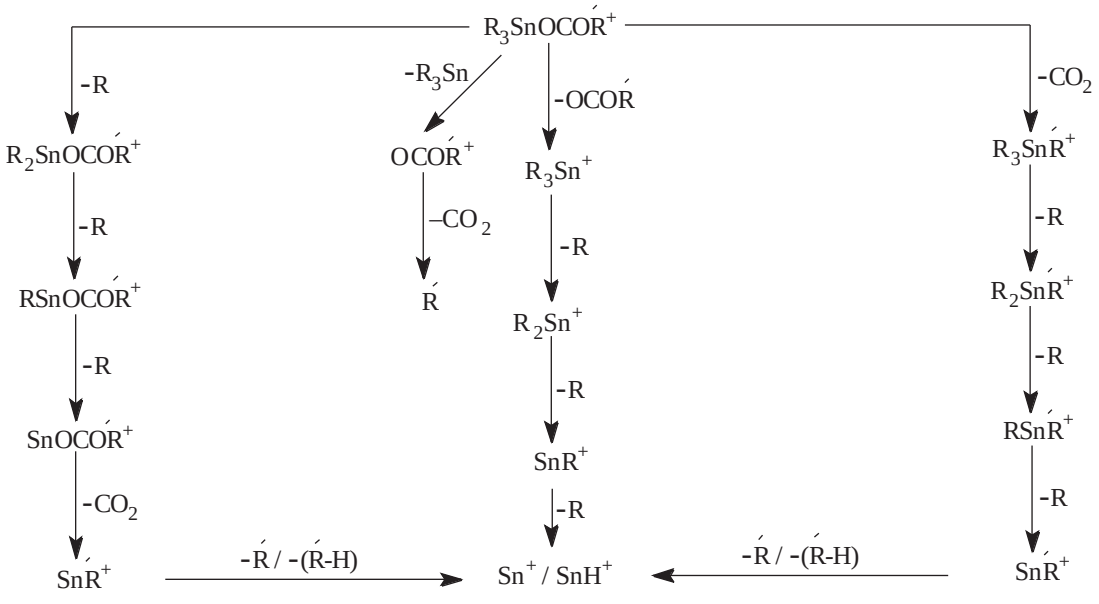


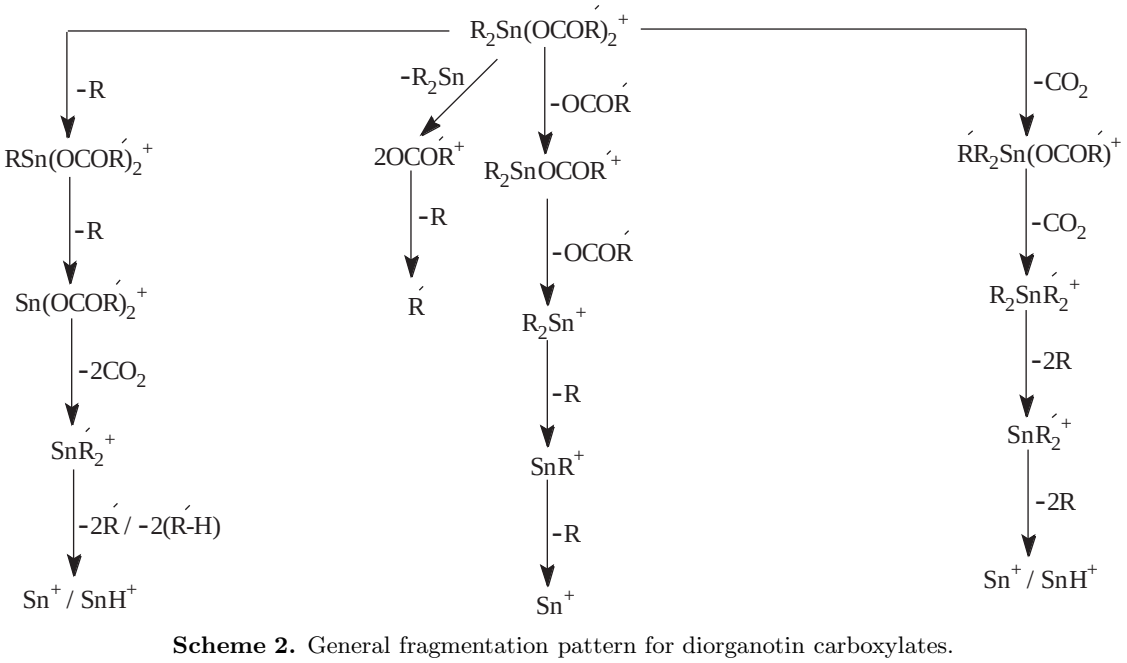
Table 8. Fragmentation Pattern of Diorganotin(IV) bis(2-(2-fluoro-4- biphenyl) propanoate.

Fragment	IV Me_2SnL_2 (m/z)	Itensity (%)	V Et_2SnL_2 (m/z)	Itensity (%)	VI Bu_2SnL_2 (m/z)	Itensity (%)	VII Ph_2SnL_2 (m/z)	Itensity (%)
$[R_2Sn(OCOR')_2]^+$	636	(n.o.)	664	n.o.	720	2	760	(n.o.)
$[RSn(OCOR')_2]^+$	621	(n.o.)	635	12	663	9	683	2
$[Sn(OCOR')_2]^+$	606	(n.o.)	606	5	606	2	606	4
$[R_2SnOCOR']^+$	393	37	421	5	477	100	517	(n.o.)
$[R_2SnR']^+$	349	(n.o.)	377	4	433	(n.o.)	473	2
$[RSnR']^+$	334	(n.o.)	348	2	376	4	396	7
$[SnR']^+$	319	3	319	2	319	5	319	2
$[R_2Sn]^+$	150	5	178	23	234	8	274	12
$[RSn]^+$	135	5	149	4	177	23	197	(n.o.)
$[Sn/SnH]^+$	121	2	121	2	121	3	121	11
$[R'COOH]^+$	244	37	244	62	244	23	244	38
$[R']^+$	199	100	199	100	199	78	199	100
$[R]^+$	15	n.o.	29	n.o.	57	18	77	9





Scheme 1. General fragmentation pattern for triorganotin carboxylates.



Scheme 2. General fragmentation pattern for diorganotin carboxylates.

Biological Testing

Biological activity tests for the title compounds were carried out against various bacteria and fungi by the "agar well diffusion" method[37]. These results are given in Tables 9-11. These results show that tributyl and triphenyltin derivatives are highly active against various bacteria and fungi. Furthermore, the extent of activity decreases with a decrease in the number of R groups, which is in accordance with earlier reports[29,38].

Table 9. Antibacterial activity(Gram positive) of organotin(IV) 2-(2-fluoro-4-biphenyl)propanoate[a,b].

Bacterium	Compounds							
	LH	I	II	III	IV	V	VI	VII
Staphylococcus aureus	-	++	+++	+++	+	0	+	+
Staphylococcus epidermidis	-	+	++	+++	++	+	0	++
Streptococcus pyogenes	-	-	+++	++	+++	++	+	++
Corynebacterium species	0	++	+++	+++	++	-	+	-
Clostridium species	0	+	+++	+++	0	+	+	0
Peptococcus species	-	++	+++	+++	0	+	+	+
Streptococcus pneumonial	-	++	+++	+++	0	+	+	+
Streptococcus faecalis	-	+++	+++	+++	-	++	+	+
Listeria monocytogenes	-	++	++	+++	+	+++	++	+
Micrococci	-	++	+++	+++	-	++	+	++

[a]+++ = High activity, ++ = moderate activity, + = low activity, 0 = not tested, - = no activity.

[b]LH = 2-(2-fluoro-4-biphenyl)propanoic acid.

Table 10. Antibacterial activity (gram negative) of organotin(IV) 2-(2-fluoro-4-biphenyl)propanoate[a,b].

Bacterium	Compounds							
	LH	I	II	III	IV	V	VI	VII
Escherichia coli	-	++	+++	+++	++	-	0	++
Proteus mirabilis	-	++	++	++	++	+	+	+
Proteus vulgaris	-	-	+++	+++	+	+	++	++
Salmonella typhi	0	++	+++	++	++	+	+	0
Corynebacterium diphtheriaa	-	0	++	+++	+	++	+	++
Proteus aeruginosa	0	-	++	++	+	+	++	++
Aeromonas sobria	+	+	++	+++	+	+	+	0
Shigella boydii	-	-	++	++	+	0	-	+
Vibrio cholera	-	+	0	+++	0	+	+	+
Brucella species	-	+	+++	++	+	+	+	+

[a]+++ = High activity, ++ = moderate activity, + = low activity, 0 = not tested, - = no activity.

[b]LH = 2-(2-fluoro-4-biphenyl)propanoic acid.

Table 11. Antifungal activity of organotin(IV) 2-(2-fluoro-4-biphenyl)propanoate[a,b].

Fungus	Compounds							
	LH	I	II	III	IV	V	VI	VII
Candida albican	+	0	++	++	+	++	+	++
Penicillium notatum	+	++	++	+++	+	++	+	++
Duterium notatum	0	+	++	+++	+	+	++	++
Genicularia	+	+	++	++	0	++	++	+
Alternaria solani	0	+	+++	++	++	++	++	++
Fusarium solani	-	++	++	++	0	+	++	+
Epidermophyton floccosum	++	+	++	0	+	+	+	++
Candida tropicalis	-	+	++	++	++	++	+	+
Aspergillus niger	+	++	+++	+++	+	++	+	++
Ascomycetes	0	+	++	++	+	+	0	+
Microsporum canis	++	0	+++	+++	+	++	+	+

[a]+++ = High activity, ++ = moderate activity, + = low activity, 0 = not tested, - = no activity.

[b]LH = 2-(2-fluoro-4-biphenyl)propanoic acid.

Conclusion

It has been concluded that triorganotin carboxylates in solid state form a polymeric trigonalbipyramid structure having three R groups at equatorial and two oxygen atoms at axial positions. For diorganotin dicarboxylates, a distorted octahedral structure is proposed in solid form. In non-coordinating solvents, R_3SnL forms a four-coordination environment around tin, while R_2SnL_2 forms a penta- or hexa-coordination.

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