CHAPTER II

Organotin Carboxylates

IIA. Introduction

During the recent years interest in the field of organotin carboxylates is growing in a very rapid rate. Apart from the theoretical and structural interests involved in these investigations, the compounds containing -OCOR groups bonded to tin have acquired tremendous importance in industry and agriculture. Many of these group of compounds have already found important uses and new applications are likely to emerge in the near future.

Organotin carboxylates which may be either monomeric or polymeric are of the three general types, viz., $R_3SnOCOR'$; $R_2Sn(OCOR')_2$ and $RSn(OCOR')_3$ where R and R' may be same or different groups. Though tin tetracarboxylates, $Sn(OCOR)_4$ are not organotin compounds in the strict sense of the term, they are sometimes included in the discussion of organotin carboxylates for the sake of comparison. Many discussions with varying degree of details are available on these compounds (34-36, 38) and as such only the more important aspects will be treated here.

IIB. Preparation

A variety of methods have been applied for the preparation of organotin carboxylates. These are generally prepared by the reaction between organotin oxides or hydroxides and carboxylic acids or their anhydrides (1-10):

$$R_3SnOSnR_3 + 2R'COOH \longrightarrow 2R_3SnOCOR' + H_2O$$

$$R_3SnOSnR_3 + (R'CO)_2O \longrightarrow 2R_3SnOCOR'$$

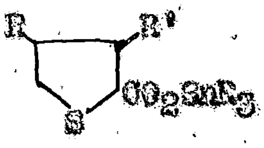
$$R_2SnO + 2R'COOH \longrightarrow R_2Sn(OCOR')_2 + H_2O$$

The water produced in these reactions is removed usually by azeotropic distillation or alternatively by refluxing at higher temperatures (11).

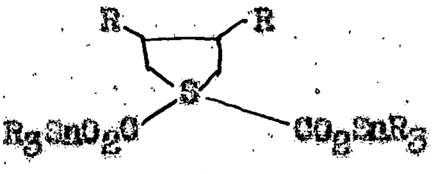
$$Bu_3SnOSnBu_3 + 2o\text{-}OH\ C_6H_4COOH \xrightarrow{\quad} 100\text{-}110^\circ C$$

$$2Bu_3Sn\ OCO\ C_6H_4OH\text{-}o + H_2O$$

Recently a number of tin thiophene carboxylates (15) of the type I and II have been prepared from the thiophene carboxylic acids and R_3SnOH or $(R_3Sn)_2O$.



I



II

R = Ph, H;
R' = Bu, Me
Cyclo C_6H_{11} etc.

Organotin carboxylates have also been prepared by the reaction of the corresponding organotin halides with the alkali metal or silver salts of carboxylic acids either by stirring the reactants in mixtures of organic/aqueous medium at room temperature or by refluxing the mixture (9,12-14). This method, represented as

$$R_3SnX + R'COOH \longrightarrow R_3SnOCOR' + MX$$

M = Na, K, Ag; X = halogen

is frequently used for its simplicity.

The ability of carboxylic acids to cleave metal carbon bonds is the basis of yet another method for the preparation of organotin carboxylates (17,18).

$$R_4Sn + R'COOH \longrightarrow R_3SnOCOR' + RH$$

The cleavage of organic groups depends on the acid strength, nature of groups R and R' and also on temperature (19-21). Vinyl groups are cleaved more readily than normal alkyl groups but less readily than phenyl groups. $Pb(OAc)_4$ has also been used for acylation of R_3SnH, R_2SnH_2, $(R_3Sn)_2$, R_4Sn and $(R_3Sn)_2O$ (16).

Kuivila (22) showed that the reaction of organotin hydrides with carboxylic acids produces carboxylates according to following equations:

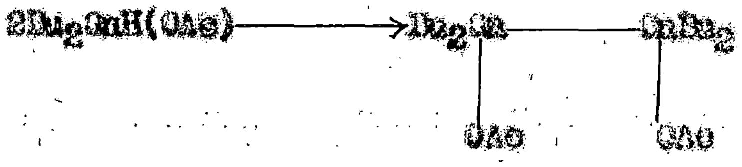
$$R_3SnH + R'COOH \longrightarrow R_3SnOCOR' + H_2$$

$$R_2SnH_2 + 2R'COOH \longrightarrow R_2Sn(OCOR')_2 + 2H_2$$

$$R_2Sn(OCOR')_2 + R_2SnH_2 \longrightarrow 2R_2SnH(OCOR')$$

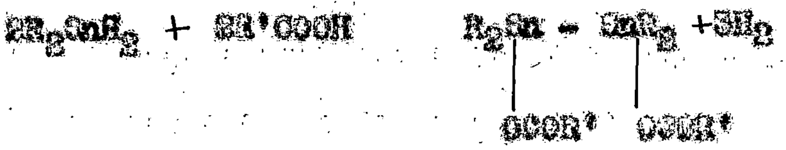
With di n-butyl tin dihydride, the intermediate hydride carboxylate decomposes to tetra-n-butyl 1,2 diacetate.

$$2Bu_2SnH_2 + 2CH_3COOH \longrightarrow 2Bu_2SnH(OAc) + 2H_2$$



$$2Bu_2SnH(OAc) \longrightarrow \underset{OAc}{Bu_2Sn} - \underset{OAc}{SnBu_2}$$

Using similar methods 1,2 dicarboxylates have also been prepared (23-25).



$$2R_2SnH_2 + 2R'COOH \qquad \underset{OCOR'}{R_2Sn} - \underset{OCOR'}{SnR_2} + 3H_2$$

The nature of the products sometimes depends on the carboxylic acid. Action of benzoyl per-oxide on di n-butyl tin dihydride also produces the 1,2, dibenzoate (26).

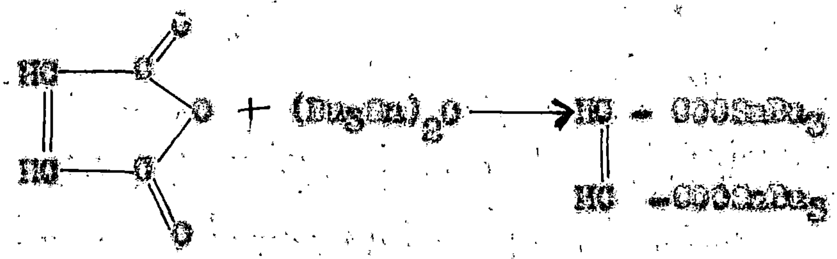
Halo carboxylate derivatives of organotin compounds are most conveniently prepared by heating equimolecular mixture of the dihalide and the dicarboxylate in an inert solvent (27, 28)

$$Bu_2SnBr_2 + Bu_2Sn(OAc)_2 \longrightarrow 2Bu_2SnBr(OAc)$$

These may be prepared by the following reaction also (29,30):

$$Bu_2SnCl_2 + KOOCR' \longrightarrow Bu_2SnCl(OOCR') + KCl$$

Anhydride of an unsaturated acid e.g., maleic anhydride forms disubstituted organotin esters when reacted with hexabutyl distannoxane (31)



A novel method of preparation of trialkyl tin acetate by electrochemical method using R_4Sn (where R = Me, Et, Pr, Bu) and Hg(I) acetate have been described by Tagliavini and his co-workers (32).

Organotin carboxylates may also be prepared from the carboxylic esters by the following reactions (66,67)

$$(R_3Sn)_2O + R'COOEt \longrightarrow R_3SnOOCR' + R_3SnOEt$$

$$R_3SnOH + R'COOEt \longrightarrow R_3SnOOCR' + EtOH$$

Tricarboxylates derivatives of the type $RSn(OOCR')_3$ are usually prepared from the corresponding trichloride by the action of silver salts of carboxylic acids (33).

III. Physical properties of organotin carboxylates

In organotin carboxylates the Sn-O bond is essentially covalent but undergoes polar reactions depending on the solvents and the attaching groups. This is why the carboxylates with small organic groups are more soluble in alcohol, ether etc than in water (35). Many of the carboxylates have low melting points indicating these to be covalent compounds.

The polymeric stannoic acids are colourless, infusible, few of them are soluble in chloroform, carbon tetrachloride and reasonably stable to hydrolysis.

Some of the physical properties of a few representative carboxylates are given in table I (34-37)

Table 1

Compound	Type	B.P. (°C/mm)	M.P. (°C)
Trimethyl tin acetate	$R_3SnOCOR'$	-	196.5-197.5
Triphenyl tin formate	"	-	203-205
Tricyclohexyl tin acetate	"	-	62-63
Tripropyltin trifluoro acetate	"	89-90/1	-
Dibutyl tin diacetate	$R_2Sn(OCOR')_2$	142-145/10	-
Diphenyl tin diacetate	"	-	116-117
Diethyl chlorotin acetate	$R_2SnX(OCOR')$	-	94
Dibutyl bromotin acetate	"	-	67-68.5

Contd...

Table 1 (Contd.)

Compound	Type	B.P.(°C/mm)	M.P.(°C)
Butyltin triacetate	$RSn(OCOR')_3$	117-119/1	-
Ethyl tin tribenzoate	"	171-173/1	-

III. Chemical properties of organotin carboxylates

The most widely studied reactions of organotin carboxylates are decarboxylation and disproportionation. An interesting cyanomethyl derivative (yield 50%) is formed when triphenyltin cyano acetate is heated under vacuum (68)

$$(C_6H_5)_3SnOOCCH_2CN \xrightarrow{\Delta} (C_6H_5)_3SnCH_2CN + CO_2$$

In the aliphatic series this type of conversion is less pronounced, the cyanomethyl compound is produced in low yield.

Trialkyl tin carboxylates of unsaturated acids give tetra substituted organotin compounds after decarboxylation (69-70)

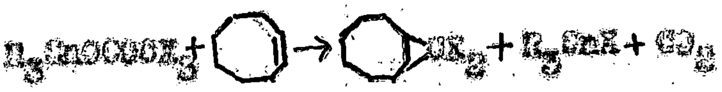
$$R_3SnOOCC\equiv CR' \longrightarrow R_3SnC\equiv CR' + CO_2$$

$$[R' = C_6H_5 \text{ or } SnR_3]$$

Thermal decomposition of triorganotin formate yields the corresponding hydride and this reaction is sometimes used for the preparation of hydrides (71).

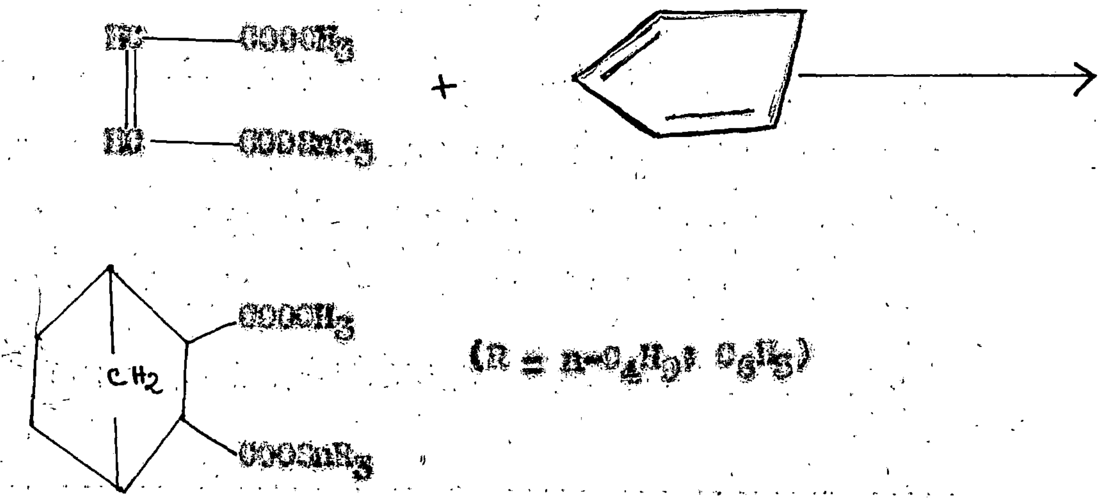
$$R_3SnOOCH \longrightarrow R_3SnH + CO_2 \qquad \left[R = n\text{-}C_3H_7,\ n\text{-}C_4H_9 \right]$$

Seyferth et al (72) have used successfully the reaction of triorganotin carboxylates of halogen substituted carboxylic acids with cyclooctene as a carbene transfer reaction, although the reaction mechanism is not yet established



($R = CH_3$, C_6H_5; $X = Cl$, Br)

Diels-Alder type reactions have been carried out with organotin carboxylates and dienes (80):



($R = n\text{-}C_4H_9$; C_6H_5)

With thiols the following equilibrium is established and the reaction can be driven from left to right removing the organic acid from the mixture when it is volatile (73).

$$R_3SnOCOR' + R''SH \rightleftharpoons R_3SnSR'' + R'COOH$$

Action of alkoxy derivatives of metals and metalloids on organotin carboxylates produces metallostannoxanes (74,75) as shown below:

$$Ph_3SnOAc + Ph_2Si(OEt)_2 \longrightarrow Ph_2Si(OSnPh_3)_2 + 2EtOAc$$

$$\begin{tikzcd} Bu_3SnOAc + Ti(OBu)_4 \arrow[r] & Bu_3SnOTi(OBu)_3 \arrow[d, "(HOCH_2CH_2)_3N"] \\ & Bu_3SnOTi(OCH_2CH_2)_3N \end{tikzcd}$$

Diorganotin dicarboxylates undergo redistribution with di-organotin dihalides, di-alkoxides and dihydrides to produce mixed carboxylates of the type $R_2SnX(OCOR')$, where X = Cl, Br, I, OCH_3, H (56).

In some cases carboxylate groups may be exchanged with other carboxylate groups as shown below (12,76)

$$AcOBu_2SnOSnBu_2OAc + 2Bu^tCOOH \longrightarrow (Bu^tCO)Bu_2SnOSnBu_2(COBu^t)$$

Oligomeric acetate is usually formed when a dialkyl tin diacetate and a dialkyltin dialkoxide are heated at 180° in water for 2 hrs (77-78)

$$(X+1)Bu_2Sn(OAc)_2 \quad (X+1)Bu_2Sn(OBu)_2 \longrightarrow$$

$$BuO(Bu_2SnO)_{2X+1}SnBu_2OAc + (2X+1)BuOAc$$

Oligomeric α-ω diacetoxy stannoxanes are also obtained by the reaction (63)

$$Bu_2Sn(OAc)_2 + H_2O \xrightarrow{CH_2N_2} AcO(Bu_2SnO)_xSnBu_2OAc + MeOAc$$

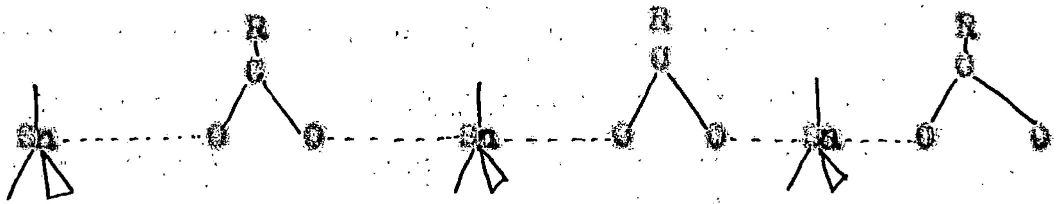
$$(X = 1,3,7,15)$$

Organotin tricarboxylates are easily hydrolysed in benzene solution to give polymeric organotin carboxylates (10)

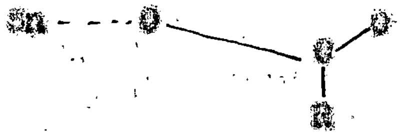
$$RSn(OCOR')_3 + H_2O \longrightarrow \left[RSn(O)OCOR'\right]_n + 2R'COOH$$

IIC. Structure of organotin complex

Structural aspects of organotin carboxylates have been recently reviewed by Okawara and Wada (33) and by Okawara and Ohara (35). The possibility of chelation or bridging through oxygen atoms of carboxylate group was pointed out by Beattie and Gilson (39) as an alternative to the previously postulated ionic nature of bonding by Freeman (40) and Okawara et al (41). Later spectroscopic studies on tin carboxylates in the solid as well as in the solution phase by different group of workers (9,13,42-45) have confirmed that trialkyl tin carboxylates are polymeric in the solid state with planar trialkyl tin groups with bridging carboxylato groups in between two tin atoms (IIIA) and are more like monomeric esters in non polar solvents (IIIB)



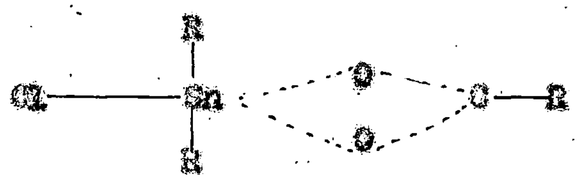
in solid phase (IIIA)



in solution phase (IIIB)

Thus, infrared spectra of trimethyl tin carboxylates in the solid state consist of two C-O stretching bands at about 1570 cm^{-1} and 1410 cm^{-1} indicating a symmetrical CO$_2$ group (46). Appearance of a single Sn-C stretching frequency in $Me_3SnOOCMe$ is consistent with a planar trimethyltin group. However, in carbon tetra chloride solution the carboxylate absorption bands are shifted to 1650 cm^{-1} and 1300 cm^{-1} respectively with the appearance of both ν_{as}(Sn-C) and ν_s(Sn-C) bands indicating the breakdown of polymeric structure (IIIA) into monomers with essentially tetrahedral tin atoms having ester like carboxylate groups bonded to it (IIIB). This conclusion is supported also by the works of Hester (23) and Tagliavini (47). Similar behaviour of trialkyl tin formates has been demonstrated by Okawara and Ohara (48-49) from spectroscopic studies and molecular weight in cyclohexane by cryoscopy.

On the other hand, tricyclohexyltin acetate, trineophyltin formate, trineophyl tin acetate and tribenzyltin acetate have been suggested to be tetra-co-ordinated monomers probably due to steric hindrance arising from the bulky organic groups (50-52). Sams et al (13) have pointed out that branching at carbon atom generally prevent the formation of polymeric structure. Similarly, the trialkyl and aryl tin derivatives of halocarboxylic acids behave more like organic esters and thus possess monomeric structures (9,53-55). A monomeric penta co-ordinated structure (IV) has been proposed for dialkyl chlorotin formates and acetates by Okawara et al (56).



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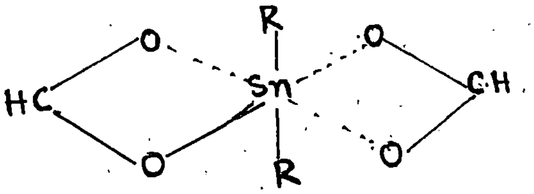
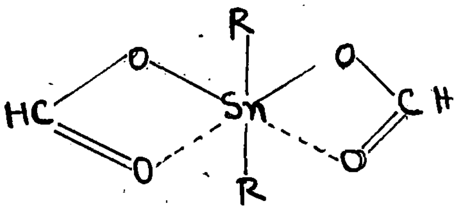
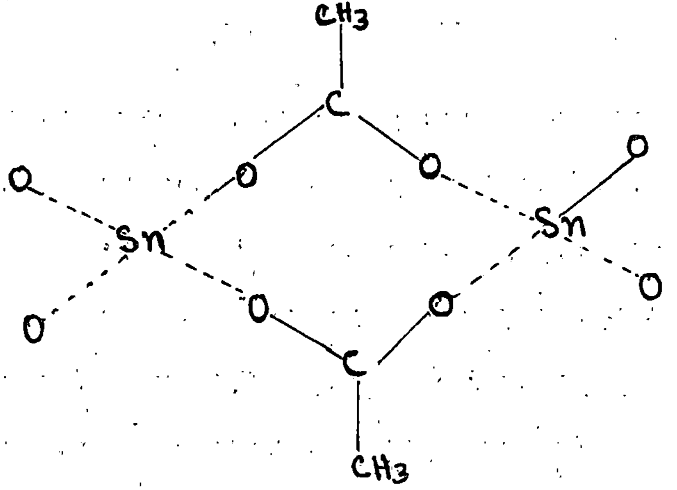
On the basis of infrared studies and molecular weights of diorganotin dicarboxylates (41,56,57,59) ionic structure with linear R_2Sn cation (V), octahedral chelated structure with linear R_2Sn group (VI) as well as polymeric structure with bridging carboxy groups (VII) have been proposed



V



VI

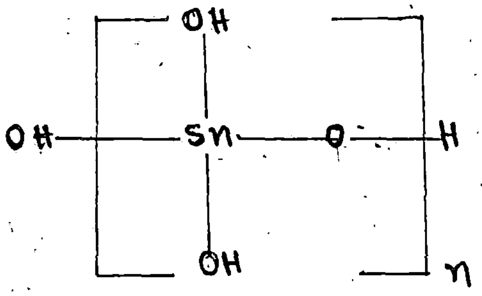


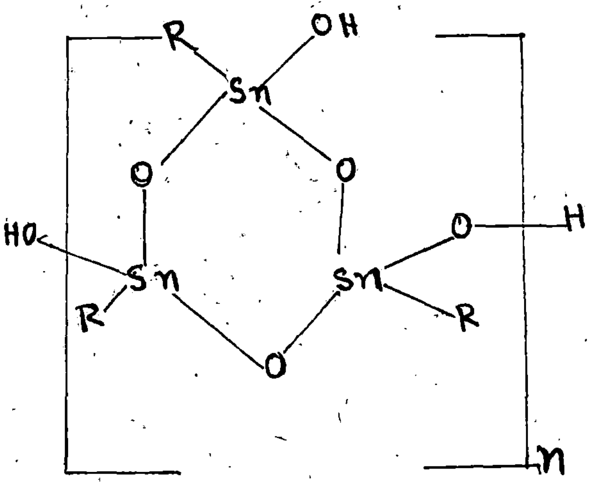
VIII

Allock and Timms (52) have studied the structure of tribenzyl and tricyclohexyltin acetate by X-Ray defraction method. This study confirms that the majority of organotin carboxylates have polymeric structure in the solid state. But with larger organic groups, the structure may become less polymeric and ultimately become monomeric.

The structure of polymeric monoorgano stannoic acids (60) and their carboxylate derivatives (9,10,15) have recently been studied through elemental analysis, IR and Mossbauer spectroscopy. Herber and his co-workers (61) have shown that polymeric tin compounds are most likely to show a mossbauer effect at ambient temperature.

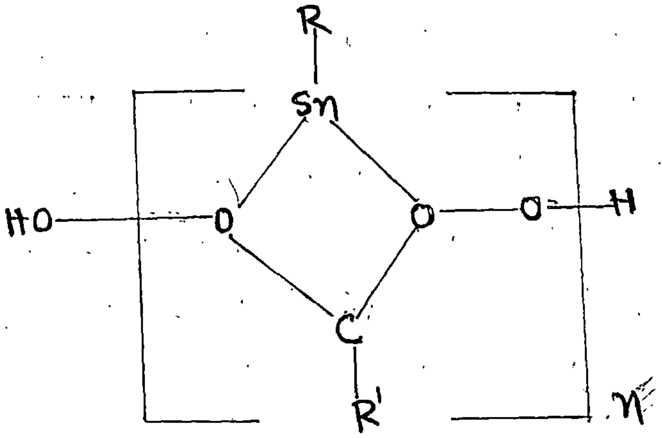
The stannoic acids are infusible powders for which a polymeric structure (VIII) have been proposed (62-63). For the partially dehydrated material structure IX has suggested by Platt and co-workers (64). In both the structures tin atom is tetra





VIII

IX



X

co-ordinated and is enclosed within the tetrahedra formed by the three oxygen atoms and the organic group R.

From the low values of quadrupole splitting (ΔE_Q = 1.29-1.83 mm/sec, table II) in organo stannoic acids Davies et al (60) have suggested a tetrahedral geometry at tin, though the possibility of association has not been completely excluded. The isomer shift values (δ = 0.40-0.78 mm/sec) are very small because of the very low 's' electron density at the tin atom resulting from electron attraction by the surrounding oxygens.

On the other hand, carboxylic acid derivatives $[RSn(O)OCOR']_n$ of the stannoic acids $[RSn(O)OH]_n$ showed larger quadrupole splitting values (ΔE_q = 2.00 - 2.64 mm/sec) (10,13) indicating a penta co-ordinated trigonal bipyramidal tin (X) which would be expected to show a quadrupole splitting of about 2.25 mm/sec (64).

Table II

Mossbauer parameters for organostannoic acid

Compound	δ mm/sec	ΔE_q mm/sec
$[MeSn(O)OH]_n$	0.40	1.20
$[EtSn(O)OH]_n$	0.76-0.70	1.61-1.70
$[BuSn(O)OH]_n$	0.65-0.70	1.52-1.71
$[C_8H_{17}Sn(O)OH]_n$	0.66	1.62
$[PhSn(O)OH]_n$	0.78	1.83

(ΔE_q = Quadrupole splitting, δ = isomer shift)

Table III

Mossbauer parameters for organometallic carboxylates

Compound	δ mm/sec	ΔE_q mm/sec
$PhSn(O)[OCO(CH_2)_8CH=CH_2]$	0.57	2.31
$PhSn(O)[OCO(CH_2)_{16}Me]$	0.56	2.32
$PhSn(O)[OCO(CH_3)_3]$	0.59	2.00
$PhSn(O)[OCOCCl_3]$	0.72	2.33

Contd..

Table III (Contd.)

Compound	δ mm/sec	ΔE_q mm/sec
$PhSn[(O)\ OCOCF_3]$	0.66	2.64
$PhSn[(O)\ OCOMe]$	0.70	2.26
$BuSn(O)OH$	0.65	1.71

(ΔE_q and δ have their usual significance)

On the basis of the preceding discussions the following generalisation may be made concerning the structure of organotin carboxylates:

(1) These compounds are generally polymeric in the solid state with the carboxy groups acting as the bridges.

(2) The polymeric structures are broken down in solution, the extent of splitting being determined by the concentration, nature of the solvent and the carboxylate.

(3) Branching at the α-carbon atom of the carboxy group usually prevents the formation of polymeric structure possibly by steric effect.

(4) Electronegative substituents like halogens etc. when attached to the carboxy group as in the halo-carboxylates, decreases the possibility of polymeric structure, probably by

diminishing the ability of the $>C = O$ group to co-ordinate with the tin atom by electron withdrawal from the oxygen atom through the inductive effect.

BIBLIOGRAPHY

1. Jen-Hsi Cho, Han-Sheng Hsu, Hsu-Yu Cheng, Cheng Lich Fen, Wen-Lich Huo and Chih-Hu, Chem.Abst. 65 (1966) 13752e

2. F.Dunn and T.Norris, Report No. 269, Australian Defence Scientific Service, Defence Standard Laboratories (1964)

3. H.Vilaren and J.C.Maire, Compt. Rend 262 (1966) 480

4. Japanese patent, Chem.Abstr. 65 (1966) 8209g

5. Japanese patent, Chem.Abstr. 65 (1966) 12240e

6. Japanese patent, Chem.Abstr. 69 (1968) 37186

7. Polish patent, Chem.Abstr. 68 (1968) 40775

8. U.S. patent, Chem.Abstr. 65 (1966) 12240b

9. B.F.E.Ford and J.R.Sams, J.Organometal.Chem. 31(1971)47

10. R.C.Poller J.N.R.Ruddick, B.Taylor and D.L.B.Toley, J.Organometal.Chem. 24(1970)341

11. French patent, Chem.Abstr. 62 (1965) 16306a

12. H.Frankel, D.Gertner, D.Wagner and A.Zilkha, J.Organometal.Chem. 9(1967)83

13. B.F.E.Ford, B.V.Liengme and J.R.Sams, J.Organometal.Chem. 19(1969)53

14. T.N.Srivastava, S.K.Tandon, J.Prakt.Chem. 311(6) (1969) 976

15. M & T International, B.V. Neth Appl,	Chem.Abstr. 81 (1974) 91752b
16. Udo Christen and W.P.Neumann,	J.Organometal.Chem. 39(1972)C58
17. H.C.Clark, R.J.O'Brien and A.L.Pickard,	J.Organometal.Chem. 4(1965)43
18. V.Peruzzo, G.Plazzogna and G.Tagliavini,	J.Organometal.Chem. 40(1972)121
19. R.H.Anderson,	Inorg.Chem. 1(1962)647
20. A.Henderson and A.K.Holliday,	J.Organometal.Chem. 4(1965)377
21. E.Wiberg and H.Behringer,	Z.Anorg.Allgem.Chem. (1964)290,329 Chem.Abstr. 61 (1964) 14163a
22. H.G.Kuivila,	Adv.Organometal.Chem. 1(1964)82
23. R.E.Hector,	J.Organometal.Chem. 23(1970)113
24. A.K.Sawyer and H.G.Kuivila,	J.Org.Chem. 27(1962)610
25. A.K.Sawyer and H.G.Kuivila,	J.Org.Chem. 27(1962)837
26. N.S.Vyazankin and V.T.Bychkov,	Zhur.Obshchei.Khim 35(1965)684 Chem.Abstr. 63(1965) 4320b
27. D.L.Alleston and A.G.Davies,	J.Chem.Soc. (1962)2050
28. A.K.Sawyer and H.G.Kuivila,	Chem. and Ind. (1961)260
29. A.S.Mufti and R.C.Poller,	J.Chem.Soc (C) (1967)1362; 1787

30. R.Okawara and E.G.Rochow,	J.Am.Chem.Soc. 82 (1960)3285
31. Z.M.Rzaev and D.A.Kochkin,	Dokl.Akad.Nauk (English translation) Consultants Bureau, Vol. 172, No. 103, January (1967).
32. G.Plazzogna, V.Peruzzo and G.Tagliavini,	J.Organometal.Chem. 16(1969)500
33. H.H.Anderson,	Inorg.Chem.3(1963)912
34. R.C.Poller,	The Chemistry of Organotin compounds, Logos, London (1970), Chapter 10
35. W.P.Neumann,	The Organic Chemistry of tin, Wiley, Chapter 17
36. R.Okawara and M.Ohara,	Organotin compounds, Volume II, Marcel Dekker (1971) p.253
37. R.K.Ingham, S.D.Rosenberg and H.Gilman,	Chem.Rev. 60 (1960) 490 and references therein
38. R.Okawara and M.Wada,	Adv.Organometal.Chem. 5(1967)137
39. I.R.Beattie and T.Gilson,	J.Chem.Soc. (1961)2585
40. J.P.Freeman,	J.Am.Chem.Soc. 80(1960)5954
41. R.Okawara, D.E.Webster and E.G.Rochow,	J.Am.Chem.Soc. 82(1960)3287
42. M.J.Janssen, J.G.A.Luijten and G.J.M.Vander Kerk,	Rec.Trav.Chim, Pays-Bas 82 (1963)90
43. R.A.Cummins and P.Dunn,	Australian J.Chem. 17(1964)185
44. B.F.E.Ford, B.V.Liengme and J.R.Sams,	J.Organometal.Chem. 19(1969)67

45. B.F.E.Ford and J.R.Sams,	J.Organometal.Chem. 21(1970)345
46. R.C.Poller,	J.Organometal.Chem. 3(1965)321
47. V.Peruzzo, G.Plazzogna and G.Tagliavini,	J.Organometal.Chem. 18(1969)84
48. R.Okawara and M.Ohara,	Bull.Chem.Soc.(Japan) 36(1963)624
49. R.Okawara and M.Ohara,	J.Organometal.Chem. 1(1964)360
50. W.T.Reichle,	Inorg.Chem. 5(1966)87
51. H.Zimmer, O.A.Homberg and M.Jayawant,	J.Org.Chem. 31(1966)3857
52. N.W.Alcock and R.E.Timms,	J.Chem.Soc. A (1968)1873, 1876
53. P.B.Simons and W.A.G.Graham,	J.Organometal.Chem. 10(1967)457
54. M.Vilarem and J.C.Maire,	Compt.Rend.Ser. 262 C (1966) 480
55. R.A.Cummins,	Australian J.Chem. 17(1964)594
56. H.Sato and R.Okawara,	Int.Symp.Mol.Struct. Spectry, Tokyo (Japan), September (1962)
57. Y.Maeda and R.Okawara,	J.Organometal.Chem. 10(1967)247
58. M.Wada, M.Shindo and R.Okawara,	J.Organometal.Chem. 1(1963)95
59. Y.Maeda, R.C.Dillard and R.Okawara,	Inorg.Nucl.Chem.Letters 2(1966)197
60. A.G.Davies, L.Smith and P.J.Smith,	J.Organometal.Chem. 39(1972)279

61. H.A.Stoeckler, H.Sano and R.H.Herber,	J.Chem.Phys 47 (1967)1182
62. J.G.A.Luijten,	Rec.Trav. Chim. Pays-Bas, 85(1966)873
63. H.Iwamoto, A.Shinagawa, H.Matsuda and S.Matsuda,	J.Chem.Soc. (Japan) Ind.Chem.Sect. 73(1970)2483
64. R.V.Parish and R.H.Platt,	Inorg.Chim.Acta, 4(1970)65
65. N.N.Zemlyanski, E.M.Panov, N.A.Slovokhotova, O.P.Samagina and K.A.Kocheshkov,	Proc.Acad.Sci. U.S.S.R. 149(1963)263
66. H.H.Anderson,	J.Org.Chem. 19(1954)1766
67. C.Biswas,	Proceedings of Indian Science Congress, Nagpur (1974)
68. J.G.M.Vander Kerk and J.G.A.Luijten,	J.Appl.Chem. 6(1956)93
69. J.G.A.Luijten and G.J.M. Vander Kerk,	Rec.Trav.Chim. Pays-Bas, 83(1964)295
70. L.K.Sunova,	Chem.Abstr. 69(1968)21975r
71. M.Ohara and R.Okawara,	J.Organometal.Chem. 3(1965)484
72. D.Seyferth F.M.Armbrecht Jr., B.Prokai and R.J.Cross,	J.Organometal.Chem. 6(1966)573
73. H.H.Anderson,	J.Org.Chem. 21(1956)869
74. C.Thies and J.B.Kinsinger,	Inorg.Chem. 3(1964)551
75. H.J.Cohen,	J.Organometal.Chem. (1967)177

76. A.J.Bloodworth, A.G.Davies and I.F.Graham,	J.Organometal.Chem. 13(1968)351
77. N.N.Zemlyanskii, E.M.Panov, O.P.Samagina and K.A.Kocheshkov,	J.Gen.Chem. U.S.S.R. 35(1965)1034
78. S.M.Shivukhin, E.D.Dudikova and E.M.Ter-Sarkisjan,	J.Gen.Chem. U.S.S.R. 32(1962)3010