

# Structure and Fungicidal Activity of Some Diorganotin(IV) with 2-Thioacetic-5-Phenyl-1,3,4-Oxadiazole and Benzamidophenylalanine

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**ABSTRACT:** New diorganotin(IV) complexes of the type  $Ph_2SnL_2$ ,  $Bu_2SnL_2$  and  $Me_2SnL_2$  of the ligands 2-thioacetic-5-phenyl-1,3,4-oxadiazole ( $HL^1$ ) and benzamidophenylalanine ( $HL^2$ ). Have been synthesized and characterized by physico-chemical techniques such as (elemental analysis, and electrolytic conductance) and spectral UV-Visible, IR and  $^1H$ ,  $^{13}C$  and  $^{119}Sn$  NMR). Monomer structures for the complexes, bidentate and octahedral geometry was proposed for the prepared complexes. Preliminary in vitro tests for fungicidal activity show that all prepared compounds display good fungicidal activity to *Gibberella*, *Cercospora arachidicola*, *Physalospora pircicola* and *Fusarium oxysporum*. Moreover, the  $Ph_2Sn(L^2)_2$  shows a higher fungicide activity than diorganotin carboxylate.

**KEY WORDS:** Diorganotin(IV), Carboxylate, Spectral studies, Fungicidal activity.

## INTRODUCTION

The interest in organotin compounds in general and organotin carboxylates in particular continues to grow because of their biological activity and its being introduced there compounds as potential antineoplastic and antituberculosis agents [1-3], PVC stabilizers [4-6] and anti-tumour drugs [7] as well a polymer catalysts [8].

Vast studies have been focused on organotin carboxylates and many of them have been characterized recently either by single crystal structure determination or by different spectroscopic techniques [9].

This paper describe the preparation and characterization

of 2-thioacetic-5-phenyl-1,3,4-oxadiazole ( $HL^1$ ) and its complexes, diphenyltin(IV) bis(2-thioacetic-5-phenyl-1,3,4-oxadiazole)  $Ph_2Sn(L^1)_2$ , dibutyltin(IV) bis(2-thioacetic-5-phenyl-1,3,4-oxadiazole)  $Bu_2Sn(L^1)_2$  and dimethyltin(IV) bis(2-thioacetic-5-phenyl-1,3,4-oxadiazole)  $Me_2Sn(L^1)_2$ . In addition the preparation and characterization of benzamidophenylalanine ( $HL^2$ ) and its complexes, diphenyltin(IV) bis(benzamidophenylalanine)  $Ph_2Sn(L^2)_2$ , dibutyltin(IV) bis(benzamidophenylalanine)  $Bu_2Sn(L^2)_2$  and Dimethyltin(IV) bis(benzamidophenylalanine)  $Me_2Sn(L^2)_2$ .

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1021-9986/11/2/67

6/\$/2.60

## EXPERIMENTAL SECTION

### Synthesis of 2-thioacetic -5-phenyl-1,3,4-oxadiazole(HL<sup>1</sup>)

A mixture of ethyl benzoate (0.1 mole) and hydrazine hydrate (0.1 mole) was refluxed for two hours, ethanol (50 mL) was added and refluxed for 5 h. The precipitate benzoyl hydrazine which separated on cooling was filtered and washed with cold methanol. To a solution of benzoyl hydrazine (0.02 mole) in ethanol 100mL at 0°C were added carbon disulfide (0.04mol) and potassium hydroxide (0.02 mole) the mixture was refluxed for 7 h. The solvent was evaporated and the residue dissolved in water and acidified with dilute hydrochloric acid. The precipitate was filtered and crystallized from (ethanol) to give 2-mercapto-5-phenyl 1,3,4-oxadiazole which react with one equivalent of chloroacetic acid in presence of KOH as a basic media to give 2-thioacetic-5-phenyl-1,3,4-oxadiazole.the final product was recrystallized from methanol (yield 82%).

### Synthesis of benzamidophenylalanine(HL<sup>2</sup>)

One gram of phenylalanine was dissolved in (25mL) of 5% NaOH solution in a conical flask. To this mixture benzoyl chloride (2.25mL) was added in a five portions in (0.49 mL increments) and shaken vigorously until all the chloride has reacted. Acidified with diluted hydrochloric acid and the crude product was washed with cold ether. Finally, the desired product was recrystallized from ethanol.

### Preparation of Complexes

Complexes were synthesized by dissolving the free ligand (2 mmol) in hot toluene and adding the diorganotin oxide (1 mmol) to the solution. The solution was refluxed for 6 hours with magnetic stirrer and then cooled and filtered. The filtrate was reduced under vacuum to a small volume and solid was precipitated by the added of petroleum ether, dried at 60 °C and recrystallized from Ethanol.

### Instrumentation

Elemental C, H and N analysis were carried out on a Fison EA 1108 analyzer, the FTIR spectra in the range (4000-370) cm<sup>-1</sup> cut were recorded as potassium bromide discs using a Perkin-Elmer spectrophotometer GX, molar conductance measurements were made in anhydrous DMF at 25 °C using Inolop-Cond Level 1 WTW, atomic absorption measurements of the prepared

complexes were obtained using Shimadzu 680cc-flame.The <sup>1</sup>H, <sup>13</sup>C and <sup>119</sup>Sn nuclear magnetic resonance spectra were recorded on a jeol 400 MHz spectrometer, relative to the internal standard TetraMethylSilane (TMS). Melting points were determined in open capillary tubes using an electrothermal 9300 digital melting point apparatus.

## RESULTS AND DISCUSSION

Table 1 shows the physical data for the ligand and the prepared complexes. The purity of the ligand and its complexes were checked by TLC using silica gel-G as adsorbent. The conductance of these complexes has been recorded in DMF at room temperature in the range 9-19 ohm<sup>-1</sup> cm<sup>2</sup> mol<sup>-1</sup>, suggesting their non-electrolytic nature. The data of CHNS and Tin analysis were obtained using flame atomic absorption technique. The calculated values were in a good agreement with the experimental values.

### Infra-Red Spectroscopy

The FTIR spectrum for HL<sup>1</sup>, shows a characteristic stretching absorption bands at 3468 cm<sup>-1</sup>, 1633 cm<sup>-1</sup>, 1315 cm<sup>-1</sup>,1606 cm<sup>-1</sup>, 1187 cm<sup>-1</sup> and 1066 cm<sup>-1</sup> assigned to hydroxyl group,  $\nu(\text{COO})$  asym.,  $\nu(\text{COO})$  sym., C=N of the oxadiazole ring, asymmetrical C-O-C, symmetrical C-O-C stretching respectively.

The FTIR spectrum of the HL<sup>2</sup>, shows a characteristic stretching absorption bands at 3688 cm<sup>-1</sup>, 3326 cm<sup>-1</sup>, 1623 cm<sup>-1</sup> and 1344 cm<sup>-1</sup> assigned to  $\nu(\text{OH})$ ,  $\nu(\text{N-H})$ ,  $\nu(\text{COO})$  asym. And  $\nu(\text{COO})$  sym. group respectively.

The COO stretching vibrations are important to predict the bonding mode of the ligand. According to *Lebl et al.* [10] the values of  $\Delta\nu$  [ $\Delta\nu = \nu$  asym. (COO) -  $\nu$  sym.(COO)] can be divided into 3 groups ; (a) In compounds where  $\Delta\nu(\text{COO}) > 350$  cm<sup>-1</sup>, the carboxylate group binds in a monodentate fashion. However, other very weak intra- and intermolecular interactions can not be excluded. (b) When  $\Delta\nu$  (COO) < 200 cm<sup>-1</sup>, the carboxylate groups of these compounds can be considered to be bidentate. (c) In compounds where  $\Delta\nu$  (COO) > 200 cm<sup>-1</sup> and < 350 cm<sup>-1</sup> an intermediate state between monodentate and bidentate (anisobidentate) occurs. It has also been suggested that the  $\Delta\nu(\text{COO})$  value in the chelating mode is less than the  $\Delta\nu(\text{COO})$  in a bridging mode [11].

Table 1: Physical data for preparation ligands and its complexes.

| compound   | Color | %Yield | M.P, °C | Found(Calcd.)%   |                |                  |                  |
|--|-------|--------|---------|------------------|----------------|------------------|------------------|
|  |       |        |         | C                | H              | N                | Sn               |
| HL <sup>1</sup>                                  | White | 84     | 162-164 | 50.81<br>(50.84) | 3.43<br>(3.41) | 11.79<br>(11.86) | -                |
| HL <sup>2</sup>                                  | White | 90     | 165-166 | 71.32<br>(71.36) | 5.56<br>(5.61) | 5.33<br>(5.20)   | -                |
| Ph <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | White | 78     | 110-111 | 50.97<br>(51.70) | 3.76<br>(3.25) | 7.51<br>(7.54)   | 8.44<br>(8.63)   |
| Bu <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | White | 69     | 145-147 | 47.54<br>(47.81) | 2.34<br>(4.59) | 7.10<br>(7.96)   | 10.01<br>(9.12)  |
| Me <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | White | 84     | 138-139 | 42.76<br>(42.67) | 2.27<br>(3.26) | 9.53<br>(9.05)   | 10.44<br>(10.36) |
| Ph <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | White | 80     | 110-112 | 64.98<br>(65.28) | 4.78<br>(4.73) | 3.76<br>(3.46)   | 14.56<br>(14.66) |
| Bu <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | White | 77     | 133-135 | 62.24<br>(62.43) | 6.12<br>(6.03) | 4.01<br>(3.64)   | 15.33<br>(15.43) |
| Me <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | White | 84     | 101-103 | 60.12<br>(59.58) | 4.93<br>(5.00) | 4.11<br>(4.09)   | 17.12<br>(17.32) |

Table 2: Characteristic absorption bands of ligands and its complexes (cm<sup>-1</sup>).

| compound   | $\nu(\text{O-H})$ | $\nu(\text{COO})$ asym | $\nu(\text{COO})$ sym | $\nu(\text{Sn-C})$ | $\nu(\text{Sn-O})$ | UV ( $\lambda$ ) |
|--|-------------------|------------------------|-----------------------|--------------------|--------------------|------------------|
| HL <sup>1</sup>                                  | 3468              | 1633                   | 1351                  | -                  | -                  | 205,276          |
| HL <sup>2</sup>                                  | 3688              | 1623                   | 1344                  | -                  | -                  | 221,265          |
| Ph <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | -                 | 1590                   | 1341                  | 570                | 455                | 234,285          |
| Bu <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | -                 | 1596                   | 1341                  | 554                | 466                | 236,289          |
| Me <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | -                 | 1594                   | 1345                  | 564                | 430                | 224,284          |
| Ph <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | -                 | 1532                   | 1336                  | 568                | 445                | 225,266          |
| Bu <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | -                 | 1535                   | 1335                  | 553                | 467                | 226,278          |
| Me <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | -                 | 1540                   | 1337                  | 556                | 451                | 231,269          |

From the preceding discussion it is proposed that in the investigated compounds, have chelating-type carboxylates. The disappearance of the hydrogen from hydroxyl group on complexation indicate the complexation is through the oxygen atom. The bands for  $\nu(\text{Sn-C})$  and  $\nu(\text{Sn-O})$  are assigned in the range of (553-570) and (430-467) cm<sup>-1</sup> respectively. The IR data of the complexes are shown in Table 2. The Table lists the stretching frequency ( $\nu$ ) for some of the characteristics groups exhibited by the ligands and its complexes.

The presence of only one Sn-C band indicates that the two R groups are in trans axial position [12]. According to group theoretical predictions the trans- SnO<sub>4</sub>C<sub>2</sub> system should exhibit one Sn-O and one Sn-C vibration and the cis isomer two Sn-C and four Sn-O stretching vibrations in the IR spectra [13].

The appearance of one Sn-C and one Sn-O stretching vibration in the IR spectra of the ligand di-alkyletin(IV) complexes suggests that two R groups are in trans configuration. The UV spectrum of the ligand exhibit a bands attributable to the intra-ligand  $\pi \rightarrow \pi^*$  transition. These bands shift to longer wavelengths in the spectra of tin complexes.

#### Nuclear magnetic resonance

The data of proton NMR of the 2-thioacetic-5-phenyl-1,3,4-oxadiazole (HL<sup>1</sup>) and its complexes displayed good solubility in DMSO. The proton nuclear magnetic resonance spectral data gave additional support for the composition of the complexes. The observed changes are evidences of complexation had happened because the chemical shift of a compound is heavily depended on

Table 3:  $^1\text{H}$ NMR spectral data ( $\delta$ ,ppm) of the ligand and complexes.

| compound   | O-H  | C-H aromatic | C-(2)H aliphatic |
|--|------|--------------|------------------|
| HL <sup>1</sup>                                  | 9.58 |              | 3.88             |
| HL <sup>2</sup>                                  | 9.11 | 7.41-7.85    | 3.92             |
| Ph <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> |      | 7.66-7.94    | 3.32             |
| Bu <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> |      | 7.57-7.95    | 3.33             |
| Me <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> |      | 7.61-7.93    | 3.34             |
| Ph <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | -    | 7.40-7.81    | 3.81             |
| Bu <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | -    | 7.37-7.81    | 3.87             |
| Me <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | -    | 7.36-7.83    | 3.88             |

Table 4:  $^{13}\text{C}$ NMR spectral data ( $\delta$ ,ppm) of the ligands and complexes.

| compound   | C=O acid | C-H aromatic  | C-H <sub>2</sub> aliphatic | $^1\text{H-Sn}$ | $^{119}\text{Sn}$ |
|--|----------|---------------|----------------------------|-----------------|-------------------|
| HL <sup>1</sup>                                  | 168.15   | 123.12-131.87 | 39.69                      |                 | -                 |
| HL <sup>2</sup>                                  | 179.64   | 124.41-130.65 | 41.44                      |                 | -                 |
| Ph <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | 170.21   | 122.13-131.88 | 39.93                      | 124             | -425.64           |
| Bu <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | 170.34   | 122.26-31.85  | 39.91                      | 110             | -422.25           |
| Me <sub>2</sub> Sn(L <sup>1</sup> ) <sub>2</sub> | 170.91   | 122.24-131.83 | 39.65                      | 104             | -432.64           |
| Ph <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | 175.54   | 126.13-133.55 | 42.07                      | 109             | -445.22           |
| Bu <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | 174.18   | 124.43-131.65 | 42.11                      | 124             | -433.22           |
| Me <sub>2</sub> Sn(L <sup>2</sup> ) <sub>2</sub> | 174.10   | 127.52-132.87 | 42.34                      | 103             | -431.65           |

its electronic environment. The spectra also exhibit a singlet -OH peaks at 9.58 ppm due to hydroxyl group.

Ligand (HL<sup>2</sup>) give a single resonance near  $\delta$  8.47 ppm attributable to the N-H proton. The spectra also exhibit a singlet -OH peaks at 9.11 ppm due to hydroxyl group. The hydroxyl resonances is absent in the spectra of the complexes indicating deprotonation and coordination of Tin to the oxygen. There is a small upfield shift of the aromatic protons resonances of the ligand upon chelation with the diorganotin(IV) moiety [14]. The complexes Ph<sub>2</sub>SnL<sub>2</sub>, Bu<sub>2</sub>SnL<sub>2</sub> and Me<sub>2</sub>SnL<sub>2</sub> Show additional signals. The methyltin (Sn-CH<sub>3</sub>) accure at 1.35, 1.33 and 1.31 ppm as on the sharp singlet at integrates for the protons accompanied by satellites due to the  $^1\text{H}$ - $^{119}\text{Sn}$  coupling that corresponds to the hydrogen atom of the methyl protons of the Me-Sn for the Me<sub>2</sub>SnL<sub>2</sub>. In dibutyltin(IV) complex the butyl protons appears as a multiple and a triplet in the range 1.45-0.89 ppm due -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>CH<sub>3</sub> group. The aromatic protons in Ph-Sn appears in the range 7.23-7.31 ppm [14].

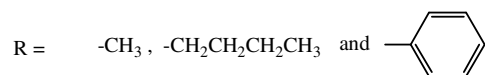
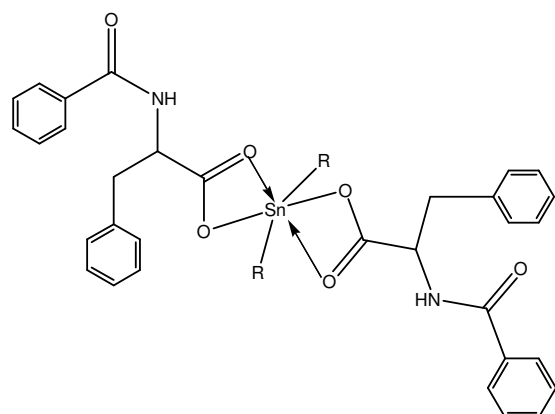
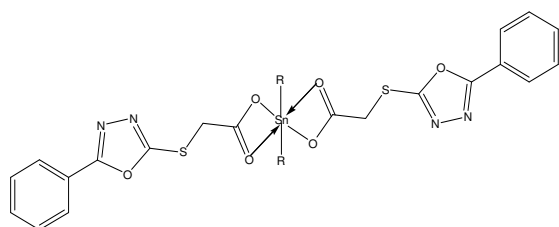
Table 4 shows the most relevant  $^{13}\text{C}$  and  $^{119}\text{Sn}$ NMR data, their spectra were recorded in [ $^2\text{H}_6$ ] DMSO. The C=O resonance group of the complexes where shifted downfield compared with the position in the free ligand which appeared. It is most likely that shift is due to the decrease of electron density at carbon atoms when oxygen is bonded to metal ion [14].

This observation lends further evidence that the complexation occurred through the oxygen atoms of the carboxylate group.  $^{119}\text{Sn}$ NMR and  $^1\text{H-Sn}$  spectra for the complexes were recorded in [ $^2\text{H}_6$ ] DMSO. solution are listed in Table 4. The coupling constants observed are of the same order as those reported in the literature chelate tin(IV) derivatives [15].  $^{119}\text{Sn}$  resonance appear, as usual, at lower field region than in Bu<sub>2</sub>SnL<sub>2</sub> and Me<sub>2</sub>SnL<sub>2</sub> in spite of the greater electron withdrawing capability of the phenyl group. This resonance probably reflects the greater shielding ability of the phenyl group.

Table 5: Fungicidal activities of prepared compounds

| compound                             | <i>Gibberela</i> | <i>Cercospora arachidicola</i> | <i>Physolospora piricola</i> | <i>Fusarium oxysporum</i> |
|--------------------------------------|------------------|--------------------------------|------------------------------|---------------------------|
| $\text{Ph}_2\text{Sn}(\text{L}^1)_2$ | 23.4             | 35.4                           | 84.1                         | 55.4                      |
| $\text{Bu}_2\text{Sn}(\text{L}^1)_2$ | 21.3             | 33.4                           | 44.5                         | 18.4                      |
| $\text{Me}_2\text{Sn}(\text{L}^1)_2$ | 18.2             | 23.4                           | 38.1                         | 12.4                      |
| $\text{Ph}_2\text{Sn}(\text{L}^2)_2$ | 22.4             | 44.3                           | 83.6                         | 70.3                      |
| $\text{Bu}_2\text{Sn}(\text{L}^2)_2$ | 30.2             | 60.4                           | 80.1                         | 50.4                      |
| $\text{Me}_2\text{Sn}(\text{L}^2)_2$ | 20.4             | 43.4                           | 50.2                         | 30.5                      |

On the basis of the preceding discussion, the structure of the complexes suggested as follows:



### Biological activity

Preliminary *in vitro* tests for fungicidal activity of ligand and complexes have been carried out by the fungi growth inhibition method [16]. These compounds are dissolved in DMF at a concentration of 50 ppm. The data

are summarized in Table 5, and show that all compounds display certain activity to *Physolospora piricola* at a low concentration. Moreover, the  $\text{Ph}_2\text{SnL}_2$  are more active than the other diorganotin derivatives. In addition,  $\text{Ph}_2\text{Sn}(\text{L}^2)_2$  shows the highest inhibition percentage for *Physolospora piricola* (83.6%) *in vitro*.

### CONCLUSIONS

The ligands 2-thioacetic-5-phenyl-1,3,4-oxadiazole and benzamidophenylalanine acid was successfully synthesized. The ligand was treated with different diorganotin(IV) salts to afford the corresponding complexes. It may conclude that the ligand coordinated through carboxylate to the Tin atom leading to the formation of four membered ring chelate. Octahedral geometry was proposed for the prepared complexes. Biological activity data have shown that the reported complexes have a significant biological activity against *Gibberela*, *Cercospora arachidicola*, *Physolospora piricola* and *Fusarium oxysporum*.

### Acknowledgment

The authors acknowledge the Universiti Kebangsaan Malaysia, IIE, SRF and to the supporting staffs of School of Chemical Sciences and Food Technology, Faculty of Science and Technology, University Kebangsaan Malaysia and Department of Chemistry, College of Science, Al-Nahrain University for their encouragement.

Received : July 28, 2008 ; Accepted : Sep. 1, 2010

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