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Organotin(IV) complexes with pyruvic acid phenylhydrazone (HPAPD): synthesis, spectral characterization, and in vitro antibacterial activity

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Organotin(IV) complexes with pyruvic acid phenylhydrazone (HPAPD): synthesis, spectral characterization, and *in vitro* antibacterial activity

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The reaction of pyruvic acid phenylhydrazone [HPAPD, (**1**)] with organotin(IV) chloride(s) leads to the formation of five new organotin(IV) complexes: [$MeSnCl_2$(PAPD)] (**2**), [$BuSnCl_2$(PAPD)] (**3**), [$PhSnCl_2$(PAPD)] (**4**), [Me_2SnCl(PAPD)] (**5**), and [Ph_2SnCl(PAPD)] (**6**). The ligand [HPAPD, (**1**)] and its organotin(IV) complexes (**2**–**6**) have been characterized by CHN analyses, molar conductivity, UV-Vis, FT-IR, ^1H, ^{13}C, and ^{119}Sn NMR spectral studies. Spectroscopic data suggested that HPAPD is coordinated to tin(IV) through the carboxylato-O and azomethine-N as a mononegative bidentate chelating agent; the coordination number of tin is five. Compound **1** and its organotin(IV) complexes (**2**–**6**) were assayed for *in vitro* antibacterial activity against *Staphylococcus aureus*, *Bacillus subtilis*, *Enterobacter aerogenes*, *Escherichia coli*, and *Salmonella typhi*. The screening results show that **2**–**6** have better antibacterial activity than **1** and that **6** exhibits significantly better activity than **2**–**5**.

Keywords: Pyruvic acid phenylhydrazone; Organotin(IV) complexes; Spectral analyses; Antibacterial activity

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1. Introduction

The chemistry of hydrazones plays an important role in coordination chemistry. The chemical properties of hydrazones have been the subject of studies for some time due to their chelating capability and pharmacological applications [1, 2].

Hydrazones and their metal complexes can also be used as analytical reagents, as a polymer-coating, ink, pigment, and fluorescent materials [3]. Organotin(IV)-hydrazone compounds are studied due to their agricultural and industrial applications [4, 5]. Studies have been reported on the synthesis, antitumor activities, and structural elucidations of various organotin(IV) derivatives of hydrazones [6, 7]. In addition to antitumor activities, organotin(IV) complexes with Schiff bases have interesting structural possibilities. Reaction of Schiff bases and the alkyltin in different solutions

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has resulted in different products, as recently reported for some diorganotin(IV) complexes of pyruvic acid isonicotinyl hydrazone and pyruvic acid salicylhydrazone [8]. In previous work, we reported the X-ray crystal structure of pyruvic acid phenylhydrazone (HPAPD) [9]. From the literature survey, studies on organotin(IV) complexes derived from pyruvic acid hydrazone ligands containing ON-donors are still lacking. We have synthesized a series of organotin(IV) complexes with the Schiff base, HPAPD. These complexes have been characterized by elemental analysis, molar conductivity, UV-Vis, FT-IR, ^{1}H, ^{13}C, and ^{119}Sn NMR spectral studies. The antibacterial activities of the studied compounds have been investigated against *Staphylococcus aureus*, *Bacillus subtilis*, *Enterobacter aerogenes*, *Escherichia coli*, and *Salmonella typhi*.

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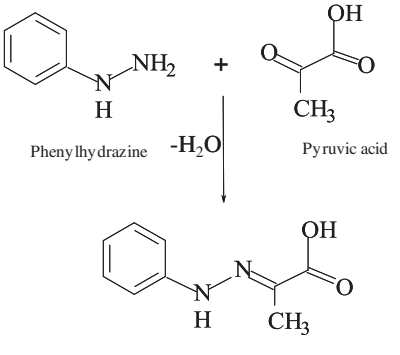
2. Experimental

2.1. *Materials and methods*

All reagents were purchased from Fluka, Aldrich, and JT Baker. Solvents were purified according to standard procedures [10]. UV-Vis spectra were recorded in $CHCl_3$ solution with a Perkin Elmer Lambda 25 UV-Vis spectrophotometer. Infrared (IR) spectra were recorded as KBr discs using a Perkin Elmer Spectrum GX Fourier-Transform spectrometer from 4000 to 370 cm^{-1} at room temperature. ^{1}H, ^{13}C, and ^{119}Sn NMR spectra were recorded on a JEOL 500 MHz-NMR spectrophotometer with chemical shifts given in ppm relative to $SiMe_4$ and $SnMe_4$ in $CDCl_3$ solvent. CHN analyses were obtained with a Flash EA 1112 series CHN elemental analyzer. Molar conductivity measurements were carried out with a Jenway 4510 conductivity meter using DMF.

2.2. *Synthesis of HPAPD (1)*

Pyruvic acid (0.440 g, 5 mmol) was dissolved in 10 mL absolute ethanol with constant stirring. An ethanolic solution of phenylhydrazine (0.540 g, 5 mmol) was then added to the solution dropwise. The resulting reaction mixture was refluxed for 4 h (scheme 1). On cooling the solution to room temperature, a light-orange powder separated, which



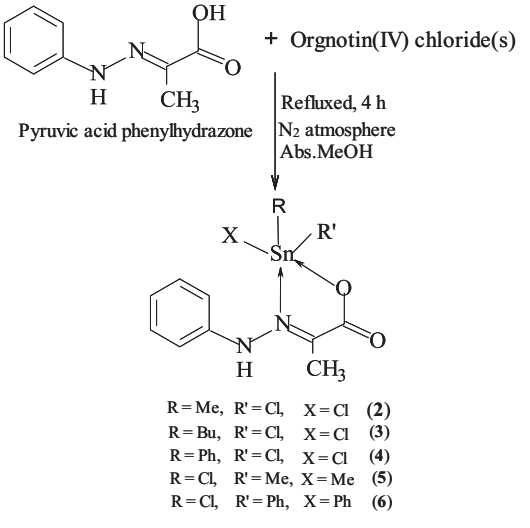
Scheme 1. Synthesis of pyruvic acid phenylhydrazone [HPAPD] (**1**).

was filtered and washed with ethanol. Light-orange microcrystals were recrystallized from ethanol and dried *in vacuo* over silica gel. Yield: 0.724 g, 73%: m.p.: 187–189°C: UV-Vis ($CHCl_3$) λ_{max} (nm): 298, 338: FT-IR (KBr disc, cm^{-1}) ν_{max}: 3333 (br, OH), 3285 (s, NH), 1709 (m, C=O), 1595 (m, C=N), 991 (m, N–N). ^1H NMR ($CDCl_3$, ppm) δ: 11.56 (s, 1H, COOH), 9.43 (s, 1H, N–H), 7.28–7.25 (m, 5H, phenyl ring), 2.13 (s, 3H, N=C–CH_3). ^{13}C NMR ($CDCl_3$, ppm) δ: 165.71 (COOH), 143.10 (C=N), 132.24–122.17 (phenyl ring), 9.04 (CH_3). Anal. Calcd for $C_9H_{10}N_2O_2$ (%): C, 60.66; H, 5.65; N, 15.72. Found (%): C, 60.61; H, 5.59; N, 15.68.

2.3. *Synthesis of [MeSnCl$_2$(PAPD)] (2)*

HPAPD (0.356 g, 2.0 mmol) was dissolved in absolute methanol (10 mL) under nitrogen in a Schlenk round bottom flask. Then, 10 mL methanolic solution of methyltin(IV) trichloride (0.48 g, 2.0 mmol) was added dropwise. The resulting reaction mixture was refluxed for 4 h (scheme 2) and cooled to room temperature. The microcrystals were filtered off, washed with a small amount of cold methanol, and dried *in vacuo* over silica gel. Yield: 0.62 g, 74%: m.p.: 224–226°C. Molar conductance (DMF) $\Omega^{-1}\,cm^2\,mol^{-1}$: 7.5: UV-Vis ($CHCl_3$) λ_{max} (nm): 312, 368, 411: FT-IR (KBr disc, cm^{-1}) ν_{max}: 3331 (s, NH), 1603 [m, $\nu_{asy}(COO^-)$], 1370 [m, $\nu_{sym}(COO^-)$], 1584 (m, C=N), 1024 (m, N–N), 602 (w, Sn–C), 550 (w, Sn–O), 436 (w, Sn–N). ^1H NMR ($CDCl_3$, ppm, $^2J[^{119}Sn, ^1H]$) δ: 9.76 (s, 1 H, N–H), 7.29–7.18 (m, 5H, phenyl ring), 2.70 (s, 3H, N=C–CH_3), 1.20 (s, 3H, Sn–CH_3), [75.3 Hz]. ^{13}C NMR ($CDCl_3$, ppm) δ: 173.25 (COO^-), 150.08 (C=N), 132.03–114.11 (phenyl ring), 19.69 (CH_3), 9.21 (Sn–CH_3). ^{119}Sn NMR ($CDCl_3$, ppm) δ: −170.06. Anal. Calcd for $C_{10}H_{12}N_2O_2Cl_2Sn$ (%): C, 31.45; H, 3.16; N, 7.33. Found (%): C, 31.40; H, 3.12; N, 7.10.

The other complexes (**3**–**6**) were synthesized using a similar procedure to organotin(IV) complex (**2**) using appropriate organotin(IV) chloride(s) (scheme 2).



Scheme 2. The reaction scheme for the synthesis of organotin(IV) complexes (**2**–**6**).

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2.4. *Synthesis of [BuSnCl$_2$(PAPD)] (3)*

Yield: 0.71 g, 77%: m.p.: 218–220°C. Molar conductance (DMF) $\Omega^{-1}\,cm^2\,mol^{-1}$: 5.8: UV-Vis ($CHCl_3$) λ_{max} (nm): 300, 354, 402: FT-IR (KBr disc, cm^{-1}) ν_{max}: 3286 (s, NH), 1611 [m, $\nu_{asy}(COO^-)$], 1375 [m, $\nu_{sym}(COO^-)$], 1581 (m, C=N), 1072 (m, N–N), 600 (w, Sn–C), 557 (w, Sn–O), 463 (w, Sn–N). ^{1}H NMR ($CDCl_3$, ppm) δ: 9.65 (s, 1H, N–H), 7.29–7.24 (m, 5H, phenyl ring), 2.55 (s, 3 H, N=C–CH_3), 2.01–2.00 (t, 2H, Sn–CH_2–CH_2–CH_2–CH_3), 1.67–1.61 (m, 2H Sn–CH_2–CH_2–CH_2–CH_3), 1.50–1.28 (m, 2H, Sn–CH_2–CH_2–CH_2–CH_3), 0.99–0.91 (t, 3H, Sn–CH_2–CH_2–CH_2–CH_3). ^{13}C NMR ($CDCl_3$, ppm) δ: 172.42 (COO^-), 151.23 (C=N), 132.78–114.87 (phenyl ring), 27.08 (Sn–CH_2–CH_2–CH_2–CH_3), 24.69 (Sn–CH_2–CH_2–CH_2–CH_3), 22.65 (Sn–CH_2–CH_2–CH_2–CH_3), 20.75 (Sn–CH_2–CH_2–CH_2–CH_3), 15.95 (CH_3). ^{119}Sn NMR ($CDCl_3$, ppm) δ: −174.5. Anal. Calcd for $C_{13}H_{18}N_2O_2Cl_2Sn$ (%): C, 36.83; H, 4.28; N, 6.60. Found (%): C, 36.75; H, 4.25; N, 6.57.

2.5. *Synthesis of [PhSnCl$_2$(PAPD)] (4)*

Yield: 0.75 g, 78%: m.p.: 212–214°C. Molar conductance (DMF) $\Omega^{-1}\,cm^2\,mol^{-1}$: 8.6: UV-Vis ($CHCl_3$) λ_{max} (nm): 299, 352, 406: FT-IR (KBr disc, cm^{-1}) ν_{max}: 3284 (s, NH), 1605 [m, $\nu_{asy}(COO^-)$], 1367 [m, $\nu_{sym}(COO^-)$], 1580 (m, C=N), 1022 (m, N–N), 614 (w, Sn–C), 579 (w, Sn–O), 448 (w, Sn–N). ^{1}H NMR ($CDCl_3$, ppm) δ: 9.62 (s, 1H, N–H), 7.29–7.21 (m, 10H, phenyl ring), 2.52 (s, 3H, N=C–CH_3). ^{13}C NMR ($CDCl_3$, ppm) δ: 175.21 (COO^-), 154.56 (C=N), 132.70–115.60 (phenyl ring), 16.45 (CH_3). ^{119}Sn NMR ($CDCl_3$, ppm) δ: −178.3. Anal. Calcd for $C_{15}H_{14}N_2O_2Cl_2Sn$ (%): C, 40.58; H, 3.17; N, 6.31. Found (%): C, 40.52; H, 3.15; N, 6.25.

2.6. *Synthesis of [Me$_2$SnCl(PAPD)] (5)*

Yield: 0.62 g, 74%: m.p.: 208–210°C. Molar conductance (DMF) $\Omega^{-1}\,cm^2\,mol^{-1}$: 6.5: UV-Vis ($CHCl_3$) λ_{max} (nm): 301, 347, 412: FT-IR (KBr disc, cm^{-1}) ν_{max}: 3288 (s, NH), 1604 [m, $\nu_{asy}(COO^-)$], 1376 [m, $\nu_{sym}(COO^-)$], 1579 (m, C=N), 1076 (m, N–N), 620 (w, Sn–C), 536 (w, Sn–O), 462 (w, Sn–N). ^{1}H NMR ($CDCl_3$, ppm, $^2J[^{119}Sn, ^1H]$) δ: 9.66 (s, 1H, N–H), 7.35–7.16 (m, 5H, phenyl ring), 2.65 (s, 3H, N=C–CH_3), 1.06 (s, 6H, Sn–CH_3), [79.5 Hz]. ^{13}C NMR ($CDCl_3$, ppm) δ: 171.88 (COO^-), 150.50 (C=N), 132.75–118.22 (phenyl ring) 14.76 (CH_3), 9.15 (Sn–CH_3). ^{119}Sn NMR ($CDCl_3$, ppm) δ: −179.2. Anal. Calcd for $C_{11}H_{15}N_2O_2ClSn$ (%): C, 36.55; H, 4.18; N, 7.75. Found (%): C, 36.51; H, 4.13; N, 7.70.

2.7. *Synthesis of [Ph$_2$SnCl(PAPD)] (6)*

Yield: 0.92 g, 74%: m.p.: 218–220°C. Molar conductance (DMF) $\Omega^{-1}\,cm^2\,mol^{-1}$: 10.2: UV-Vis ($CHCl_3$) λ_{max} (nm): 314, 342, 408: FT-IR (KBr disc, cm^{-1}) ν_{max}: 3286 (s, NH), 1602 [m, $\nu_{asy}(COO^-)$], 1371 [m, $\nu_{sym}(COO^-)$], 1584 (m, C=N), 1023 (m, N–N), 602 (w, Sn–C), 557 (w, Sn–O), 449 (w, Sn–N). ^{1}H NMR ($CDCl_3$, ppm) δ: 9.70 (s, 1H, N–H), 7.26–7.21 (m, 10H, phenyl ring), 2.55 (s, 3H, N=C–CH_3). ^{13}C NMR ($CDCl_3$, ppm) δ: 174.11 (COO^-), 148.03 (C=N), 132.29–114.01 (phenyl ring), 16.93 (CH_3). ^{119}Sn NMR

($CDCl_3$, ppm) δ: −175.1. Anal. Calcd for $C_{27}H_{24}N_2O_2ClSn$ (%): C, 57.63; H, 4.30; N, 4.80. Found (%): C, 57.59; H, 4.26; N, 4.77.

2.8. *Antibacterial test*

The synthesized ligand (**1**) and **2**–**6** were screened *in vitro* for their antibacterial activities against *S. aureus* (ATCC 6538), *B. subtilis* (ATCC 6633), *E. aerogenes* (ATCC 13048), *E. coli* (ATCC 15224), and *S. typhi* (ATCC 10749) bacterial strains using the agar-well diffusion method [11]. Wells (size of well 6 mm in diameter) were dug in the media with the help of a sterile metallic borer with centers at least 24 mm. Eight-hour-old bacterial inoculums containing 10^4–10^6 colony forming units (CFU) mL^{-1} were spread on the surface of the nutrient agar using a sterile cotton swab. Recommended concentration of the test sample (2 mg mL^{-1} in DMSO) was introduced in the respective wells. Other wells supplemented with DMSO and reference antibacterial drug (doxycycline) served as negative and positive controls, respectively. All tests were performed in triplicate with full agreement between the results. The plates were incubated immediately at 37°C for 20 h. Activity was determined by measuring the diameter of zones showing complete inhibition (mm). Growth inhibition was calculated with reference to the positive control.

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3. Results and discussion

3.1. *Synthesis*

HPAPD was synthesized by the condensation of pyruvic acid and phenylhydrazine in absolute ethanol in 1 : 1 mole ratio (scheme 1). Complexes **2**–**6** were obtained by direct reaction of organotin(IV) chloride(s) and HPAPD in absolute methanol under N_2 (scheme 2). The physical properties and analytical data of **1**–**6** are given in Section 2. Compounds **2**–**6** were stable under N_2 and soluble in $CHCl_3$, CH_2Cl_2, DMF, DMSO, and MeCN, but insoluble in methanol, ethanol, hexane, pentane, THF, and ether. The structures of these compounds and coordination behavior of tin(IV) have been examined by different analytical techniques such as elemental analysis, molar conductivity, multinuclear NMR (^1H, ^{13}C, and ^{119}Sn) spectroscopy. The molar conductances of **2**–**6** are 5.8–10.2 $\Omega^{-1}cm^2mol^{-1}$, respectively, indicating non-electrolytes [12].

3.2. *UV-Vis spectra*

The UV-Vis spectra of **1**–**6** were carried out in a DMF solution (1×10^{-4} mol L^{-1}) at room temperature. The electronic spectrum of HPAPD showed absorptions at 298 and 338 nm assigned to $\pi \rightarrow \pi^*$ transition of aromatic ring and $n \rightarrow \pi^*$ transition of imine (>C=N), respectively [13]. After complexation, UV-Vis spectra of **2**–**6** exhibited three absorptions at 299–314, 342–368, and 402–412 nm, respectively. The $\pi \rightarrow \pi^*$ transition of aromatic ring and $n \rightarrow \pi^*$ transition of imine function are observed at higher wavelength in the complexes due to slight shift as a result of coordination. The new

band at 402–414 nm is assigned to the ligand→metal charge transfer [14]. The shift of the λ_{max} band from the ligand to the tin(IV) complex is a clear indication that coordination occurred between tin(IV) and **1**.

3.3. *IR spectra*

HPAPD displayed absorptions at 3333 cm^{-1} and 3285 cm^{-1} attributed to the OH and NH, respectively. Other absorptions at 1709, 1595, and 991 cm^{-1} are due to ν(C=O), ν(C=N), and ν(N–N), respectively. The ν(C=O), ν(C=N), and ν(Sn–O) provide valuable information about the formation of tin(IV) complex and coordination mode of the ligand. Compound **1** showed a broad OH absorption at 3333 cm^{-1}, which is absent in the spectra of **2**–**6**, suggesting **1** is coordinated to tin(IV) through carboxylate oxygen after deprotonation. The ν(C=O) of COOH at 1709 cm^{-1} for **1** was absent in IR spectra of **2**–**6**. Two new absorptions were observed at 1611–1602 cm^{-1} and 1376–1367 cm^{-1}, attributed to $\nu_{asy}(COO^-)$ and $\nu_{sym}(COO^-)$, respectively, in spectra of **2**–**6**. The magnitude of $\Delta[\nu_{asy}(COO^-)-\nu_{sym}(COO^-)]$ is 238–228 cm^{-1}, indicating the presence of monodentate carboxylate [15]. A medium band at 1595 cm^{-1} of **1** for ν(C=N) shifts to lower wavenumber at 1584–1579 cm^{-1} in spectra of **2**–**6**, suggesting the coordination of the azomethine nitrogen to tin [16]. The ν(N–N) of **1** at 991 cm^{-1} shifts to higher frequencies at 1076–1022 cm^{-1} in **2**–**6**. This observation also suggests the coordination of azomethine nitrogen to Sn(IV). Absorptions at 620–600 cm^{-1} in spectra of **2**–**6** are assigned to ν(Sn–C). New absorptions at 579–536 cm^{-1} in **2**–**6** are assigned to ν(Sn–O), comparable with the literature [17]. Another new band at 462–436 cm^{-1} is characteristic of Sn–N absorption in **2**–**6**. IR spectra indicate the coordination of **1** to tin(IV) *via* carboxylate-O and azomethine-N.

3.4. *^{1}H, ^{13}C, and ^{119}Sn NMR spectra*

^{1}H NMR spectra of **1** and organotin(IV) complexes (**2**–**6**) were recorded in $CDCl_3$. Compound **1** showed resonances at 11.56, 9.43, 7.28–7.25, and 2.13 ppm due to OH, NH, aromatic-H, and N=C–CH_3, respectively. After complexation, the OH signal was absent in **2**–**6**, suggesting deprotonation and coordination. The NH signal shifts downfield at 9.76–9.62 ppm in **2**–**6**, deshielded by the adjacent nitrogen of (N=C–CH_3). The resonance of the phenyl protons in **2**–**6** appears as a multiplet at 7.29–7.16 ppm. The azomethine proton (N=C–CH_3) signal appears at 2.13 ppm in **1,** which is shifted downfield (2.70–2.52 ppm) in **2**–**6**. This downfield shift indicates that the azomethine nitrogen is coordinated to tin (IV). The methyl attached to organotin(IV) in **2** and **5** are sharp singlets at 1.20 and 1.06 ppm, respectively, with well-defined satellites. The $^2J[^{119}Sn, ^1H]$ values of **2**, 75.3 Hz and **5**, 79.5 Hz support the five-coordinate environment around tin in the solution [18, 19]. The butyl groups attached to tin(IV) in **3** gave four resonances, 2.01–2.00 ppm (triplet, Sn–CH_2–CH_2–CH_2–CH_3), 1.67–1.61 ppm (multiplet, Sn–CH_2–CH_2–CH_2–CH_3), 1.50–1.28 ppm (multiplet, Sn–CH_2–CH_2–CH_2–CH_3), and 0.99–0.91 ppm (triplet, Sn–CH_2–CH_2–CH_2–CH_3). ^{1}H NMR information supports the IR data of **2**–**6**.

In ^{13}C NMR spectra, the position of the phenyl carbon signals undergo minor variation in **2**–**6** compared to those observed in **1**. In **1**, the chemical shifts at 165.71, 143.10, 132.24–122.17, and 9.04 ppm are due to δ(COOH), δ(C=N), δ(phenyl ring), and

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$\delta(CH_3)$, respectively. In **2–6**, the position of the carboxylate carbon shifts to lower field at 175.21–171.88 ppm in comparison with **1**, indicating the participation of the carboxylate (COO^-) in coordination to tin(IV) [20]. The chemical shifts of carbon in C=N and CH_3 in **2–6** shift downfield at 154.56–148.03 and 19.69–14.76 ppm, respectively, supporting H_3C–C=N coordination to tin(IV). The carbon signals in phenyl rings in **2–6** are at 132.78–114.01 ppm, while the four resonances at 27.08, 24.69, 22.65, and 20.75 ppm are assigned to $Sn–CH_2CH_2CH_2CH_3$ of **3**. Methyl group(s) attached to tin(IV) in **2** and **5** are at 9.21 and 9.15 ppm, respectively.

^{119}Sn NMR chemical shifts of **2–6** were recorded in $CDCl_3$. ^{119}Sn NMR spectroscopy gives information to determine the coordination number around tin. The ^{119}Sn NMR of **2–6** shows only one resonance in the range −170.6 to −179.2 ppm, indicating five-coordinate organotin(IV) [21–25].

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3.5. *Antibacterial activity*

Compound **1** and its organotin(IV) complexes (**2–6**) were tested for antibacterial activity against *S. aureus*, *B. subtilis*, *E. aerogenes*, *E. coli*, and *S. typhi*. The agar well-diffusion method was used in these assays and each experiment was performed in triplicate. The zone of inhibition represents the mean value of three readings, as given in table 1, along with the values for [*n*-$BuSnCl_3$]. Doxycyclin antibiotic was used as a standard drug in these assays. Compound **1** was inactive against all bacteria. *n*-$BuSnCl_3$ shows almost the same antibacterial activity against all bacterial species with inhibition zones of 12.9–12.5 mm. Antibacterial data indicate that **2–6** have remarkable and significant activities toward all bacterial strains but lower activity than the reference drug. Compounds **5** and **6** have significant activities against all the five tested strains. Compounds **3** and **4** also have significant activities against all test strains except *E. aerogenes*. Some microorganisms have the ability to degrade or reduce the toxicity of particular tin complexes [26, 27]. Lipopolysaccharides form an outer lipid membrane and contribute to the complex antigenic specificity of Gram(−) cells [28]. The effect of various compounds may then vary among microorganisms due to difference in the structures of the cell. The antibacterial activities shown by these compounds indicate

Table 1. Results of antibacterial activity of the free HPAPD (**1**) and its organotin (IV) derivatives[a] (**2–6**) (inhibition zone in mm).

	Zone of inhibition (mm)				
Compound	*S. aureus*	*B. subtilis*	*E. aerogenes*	*E. coli*	*S. typhi*
HPAPD (**1**)	–	–	–	–	–
n-$BuSnCl_3$	12.9	12.8	12.6	12.5	12.6
2	21.1	11.4	21.7	13.2	14.8
3	22.4	24.1	–	23.7	23.3
4	24.9	23.8	–	21.1	23.5
5	25.2	24.7	22.1	22.3	21.8
6	32.3	26.5	24.8	23.8	22.7
Doxycyclin	38.8	31.5	30.4	35.2	29.7

[a]*In vitro*, agar well-diffusion method, concentration: 2 mg mL^{-1} in DMSO, reference drugs, Doxycycline: 2 mg mL^{-1} in DMSO: Dash (–) indicated inactivity.

that coupling of HPAPD with R/R_2Sn(IV) results in complexes with important biological properties. Organotin(IV) compounds (**2**–**6**) exhibit more inhibitory effects than the parent ligand or starting organotin(IV) chloride (n-$BuSnCl_3$). Among the organotin(IV) complexes (**2**–**6**), **6** is more active, attributed to the presence of bulky phenyl groups which facilitate binding to biological molecules through π–π interactions. This antibacterial result is similar to those previously reported for other tin compounds [29–33].

4. Conclusions

We have obtained a series of new organotin(IV) complexes by direct reactions of HPAPD with corresponding organotin(IV) chloride(s) in absolute methanol. The synthesized ligand (**1**) and **2**–**6** have been characterized by electronic, IR, and NMR spectral studies, showing that the monodeprotonated ligand is coordinated to tin(IV) through carboxylate-O and azomethine-N. The tin is from the spectroscopic characterization of five coordinate in **2**–**6**. Bioactivity studies showed that **2**–**6** are more potent antibacterial agents than **1**, with diphenyltin(IV) complex (**6**) more active than **2**–**5**.

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