Research Article

Synthesis, Characterization and In Vitro Antibacterial Studies of Organotin(IV) Complexes with 2-Hydroxyacetophenone-2-methylphenylthiosemicarbazone (H$_2$dampt)

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Received 7 December 2011; Revised 31 January 2012; Accepted 14 February 2012

Academic Editor: Virtudes Moreno

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Five new organotin(IV) complexes of 2-hydroxyacetophenone-2-methylphenylthiosemicarbazone [H$_2$dampt, (1)] with formula [RSnCl$_{n-1}$, (dampt)] (where R = Me, n = 2 (2); R = Bu, n = 2 (3); R = Ph, n = 2 (4); R = Me$_2$, n = 1 (5); R = Ph$_2$, n = 1 (6)) have been synthesized by direct reaction of H$_2$dampt (1) with organotin(IV) chloride(s) in absolute methanol. The ligand (1) and its organotin(IV) complexes (2–6) were characterized by CHN analyses, molar conductivity, UV-Vis, FT-IR, $^{1}$H, $^{13}$C, and $^{119}$Sn NMR spectral studies. H$_2$dampt (1) is newly synthesized and has been structurally characterized by X-ray crystallography. Spectroscopic data suggested that H$_2$dampt (1) is coordinated to the tin(IV) atom through the thiolate-S, azomethine-N, and phenoxide-O atoms; the coordination number of tin is five. The in vitro antibacterial activity has been evaluated against Staphylococcus aureus, Enterobacter aerogenes, Escherichia coli, and Salmonella typhi. The screening results have shown that the organotin(IV) complexes (2–6) have better antibacterial activities and have potential as drugs. Furthermore, it has been shown that diphenyltin(IV) derivative (6) exhibits significantly better activity than the other organotin(IV) derivatives (2–5).

1. Introduction

Thiosemicarbazones and their metal complexes have received considerable attention in chemistry and biology, primarily because of their marked and various biological properties [1–3]. The pharmacological profiles of 2-formyl, 2-acetyl, and 2-benzoylpyridine thiosemicarbazones have been investigated [4]. Seena and Kurup [5] have synthesized and characterized dioxomolybdenum(IV) complexes with 2-hydroxyacetophenone-N(4)-cyclohexyl and N(4)-phenyl thiosemicarbazone which suggested that the Mo(IV) complex is pentacoordinated [5]. For the past few years, studies of the coordination chemistry of thiosemicarbazone involved complexes with transition metal ions [6–8]. Organotin(IV) complexes have been the subject of interest for some time because of their biomedical and commercial applications including in vitro and in vivo antitumor activity [9, 10]. Many organotin(IV) complexes have been found to be as effective as or even better than traditional anticancer drugs [11–14]. Organotin(IV) chelates with nitrogen, sulfur, and oxygen donor ligands have gained attention during the last few years [15]. The coordination chemistry of tin is extensive with various geometries and coordination numbers known for both inorganic and organometallic complexes [16, 17]. In our previous work, we have reported some new organotin(IV) complexes with heterocyclic-N(4)-cyclohexylthiosemicarbazone ligands [18, 19]. The results revealed that thiosemicarbazones derived from 2-benzoylpyridine and 2-acetylpyrazine and their tin (IV)/organotin(IV) complexes have been characterized by different spectroscopic techniques. From the literature survey, the studies on the organotin(IV) complexes derived from substituted thiosemicarbazone ligands con-taining ONS-donor atoms are still lacking.
To the best of our knowledge, there was no report on the organotin(IV) complexes of the 2-hydroxyacetophenone-2-methylphenylthiosemicarbazone. In this view, we have synthesized a series of organotin(IV) complexes with 2-hydroxyacetophenone-2-methylphenylthiosemicarbazone. These complexes have been characterized by elemental analysis, $^1$H, $^{13}$C, and $^{119}$Sn NMR spectroscopy. X-ray crystal structure of 2-hydroxyacetophenone-2-methylphenylthiosemicarbazone (1) is also described. Their biological activity data has also been reported.

2. Experimental

2.1. Materials and Methods

All reagents were purchased from Fluka, Aldrich, and JT Baker. All solvents were purified according to standard procedures [20]. UV-Vis spectra were recorded in CHCl$_3$ solution with a Perkin Elmer Lambda 4510 spectrometer. Infrared spectra were recorded on a Jasco FT-IR spectrometer in the range 4000–370 cm$^{-1}$ at room temperature. CHN analyses were obtained with a Flash EA 1112 series CHN elemental analyzer. Molar conductivity measurements were carried out with Jenway 4510 conductivity meter using DMF solvent mode.

2.2. Synthesis of 2-Hydroxyacetophenone-2-Methylphenylthiosemicarbazone (H$_2$dampt) (1).

The 2-methylphenylisothiocyanate (0.746 g, 5 mmol) and hydrazine hydrate (0.253 g, 5 mmol), each dissolved in 10 mL ethanol, were mixed with 1-2 drops of glacial acetic acid (Scheme 1). On cooling the solution to room temperature, light-yellow microcrystals were separated and washed with methanol. The microcrystals were recrystallized from methanol and dried in vacuo over silica gel. Yield: 0.43 g, 74%.

2.3. Synthesis of [MeSnCl(dampt)] (2).

H$_2$dampt (0.299 g, 1.0 mmol) was dissolved in absolute methanol (10 mL) in a Schlenk round bottom flask under a nitrogen atmosphere. Then, a methanolic solution of methyltin(IV) chloride (Scheme 2) was refuxed 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. The resulting reaction mixture was refuxed 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. The resulting reaction mixture was refuxed for 4 h (Scheme 2) and cooled to room temperature.
Scheme 1: Synthesis of 2-hydroxyacetophenone-2-methylphenylthiosemicarbazone (H₂dampt) ligand (1).

Scheme 2: Reaction scheme for the synthesis of organotin(IV) complexes (2–6).
respectively. After complexation, the UV-Vis spectra of the complexes (2–6) exhibited four absorption bands in the region at 262–268, 327–338, 367–382, and 384–414 nm, which is assigned to the HOMO/LUMO transition reaction of 2-hydroxyacetophenone and 2-methylphenylthiosemicarbazide in absolute methanol in 1:1 molar ratio. It has two tautomers within the structure, existing as either thione or thiol tautomer (Scheme 1). The present organotin(IV) complexes (2–6) were obtained by direct reaction of organotin(IV) chloride(s) and H₂dampt (1) in absolute methanol under N₂ atmosphere (Scheme 2). The physical properties and analytical data of H₂dampt (1) and its organotin(IV) complexes (2–6) are given in the experimental section. All complexes (2–6) were stable under N₂ atmosphere and soluble in CHCl₃, CH₂Cl₂, DMF, DMSO, and MeCN solvents except methanol, ethanol, hexane, pentane, THF, and ether. The molar conductance values of the complexes (2–6) are 9.1–3.1 Ω⁻¹ cm² mol⁻¹, respectively, indicate that the complexes behave as nonelectrolytes [22].

3.3. IR Spectra. The IR spectrum of free ligand (1) showed absorption bands at 3175 and 3000 cm⁻¹, which are due to the stretching vibrations of the OH and NH groups, respectively. The absorption bands at 1583, 1298, 943, and 861 cm⁻¹ are due to ν(C=N), ν(C–O), ν(N–N), and ν(C=S), respectively. Several significant changes with respect to the free ligand (1) bands on complexation suggest coordination through phenolic group, azomethine, and sulfur of the thiolic form of the ligand. The strong stretching band at 3375 cm⁻¹ that corresponds to the ν(OH) group in the spectrum of ligand (1) has disappeared in the spectra of complexes (2–6) due to the deprotonation, indicating coordination through the phenolic oxygen to tin(IV) atom. The free ligand (1) showed a band at 1298 cm⁻¹ which is due to ν(C–O). This band is shifted to lower wave numbers at 1240–1268 cm⁻¹ in the complexes (2–6), indicating the coordination of O⁻ to the tin(IV) atom [25]. The newly formed ν(C=N–N=C) band showed medium-to-strong absorption peaks in the range at 1592–1605 cm⁻¹ in the spectra of the complexes (2–6), indicating coordination of azomethine nitrogen to tin(IV) atom [26]. A sharp band at 943 cm⁻¹ is due to ν(N–N) for ligand (1) is shifted to higher frequencies at 1014–1039 cm⁻¹ in the spectra of organotin(IV) complexes (2–6). The increase in the frequency of this band in the spectra of complexes (2–6) due to an increase in the bond length again confirms coordination via the azomethine nitrogen atom [27]. The bands at 1371 and 861 cm⁻¹ in the free ligand (1) due to ν(C=S) stretching vibrations are shifted to lower frequencies at 1299–1307 cm⁻¹ and 821–838 cm⁻¹ in the spectra of the complexes (2–6), suggesting coordination through the thiolate sulfur with tin(IV) atom [28]. The IR bands observed in the range at 570–522 cm⁻¹ in the spectra of complexes (2–6) suggest the presence of Sn–O bonding in their structure. The ν(Sn–C) and ν(Sn–N) bands are tentatively assigned to absorptions in the regions 612–601 cm⁻¹ and 443–499 cm⁻¹, respectively. Based on the infrared spectra analyses of ligand (1) and its organotin(IV) complexes (2–6), it was suggested that ligand (1) was coordinated to the tin(IV) core through the phenoxide-O, azomethine-N, and thiolato-S atoms.

3.4. ¹H NMR Spectra. The ¹H NMR spectrum of free ligand (1) showed resonance signals at 10.82, 9.02, 7.31–7.25, 2.56, 2.29, and 1.19 ppm are due to OH, NH, phenyl ring protons, N=C–CH₃, CH₃, and SH, respectively. After complexation, the resonance signal of OH proton was absent in the spectra of the complexes (2–6), indicating deprotonation of the phenolic proton and supported the phenolic oxygen atom was coordinated with tin(IV) atom. The resonance signal of SH is not found in the spectra of complexes (2–6) which suggested the deprotonation of the SH proton and confirming that the ligand coordinated to the tin(IV) in the complex (2–6), one new absorption band appeared at 384–414 nm which is assigned to the ligand → metal charge transfer (LMCT) [24]. The shift of the λmax band from the ligand to the complex is supported by the coordination of ligand (1) to the tin(IV) ion.

3. Results and Discussion

3.1. Synthesis. 2-Hydroxyacetophenone-2-methylphenylthiosemicarbazone (H₂dampt) was synthesized by the condensation reaction of 2-hydroxyacetophenone and 2-methylphenylthiosemicarbazide in absolute methanol in 1:1 molar ratio. It has two tautomers within the structure, existing as either thione or thiol tautomer (Scheme 1). The present organotin(IV) complexes (2–6) were obtained by direct reaction of organotin(IV) chloride(s) and H₂dampt (1) in absolute methanol under N₂ atmosphere (Scheme 2). The physical properties and analytical data of H₂dampt (1) and its organotin(IV) complexes (2–6) are given in the experimental section. All complexes (2–6) were stable under N₂ atmosphere and soluble in CHCl₃, CH₂Cl₂, DMF, DMSO, and MeCN solvents except methanol, ethanol, hexane, pentane, THF, and ether. The molar conductance values of the complexes (2–6) are 9.1–3.1 Ω⁻¹ cm² mol⁻¹, respectively, indicate that the complexes behave as nonelectrolytes [22].

3.2. UV-Visible Spectra. The UV-Vis spectra of ligand (1) and its organotin(IV) complexes (2–6) were carried out in CHCl₃ (1 × 10⁻⁴ mol L⁻¹) at room temperature. The free ligand (1) exhibited three absorption bands at 262, 318, and 359 nm assigned to the HOMO/LUMO transition of phenolic group, azomethine, and thiolate function, respectively [23]. After complexation, the UV-Vis spectra of the complexes (2–6) exhibited four absorption bands in the region at 262–268, 327–338, 367–382, and 384–414 nm, respectively. In the electronic spectra of the complexes (2–6), the intraligand transition is shifted to higher wavelength as a result of coordination. In the spectra of organotin(IV) complexes (2–6), one new absorption band appeared at 384–414 nm which is assigned to the ligand → metal charge transfer (LMCT) [24]. The shift of the λmax band from the ligand to the complex is supported by the coordination of ligand (1) to the tin(IV) ion.
thiolate form. The azomethine proton (N=C–CH₃) signal appears at 2.56 ppm in the free ligand (1) which is shifted to high frequency at 2.98–2.62 ppm in the complexes (2–6), supporting the coordination of azomethine nitrogen to the central tin(IV) atom. The resonance signals for the protons of phenyl moiety of the ligand (1) were observed at 7.31–7.25 ppm, which is shifted to low frequency at 7.30–6.94 ppm in the complexes (2–6). This is due to the electron withdrawal tendency from the aromatic ring owing to coordination with tin(IV). The methyl group attached to the tin(IV) atom in complexes 2 and 5 gave a singlet at 1.09 and 0.98 ppm with 2[J¹¹⁹Sn,¹H] coupling constant value equal to 74.4 and 77.5 Hz, respectively, supporting the five-coordinate environment around tin(IV) [29]. The three butyl groups attached to the tin(IV) moiety in the organotin(IV) complex 3 gave four resonance signals, namely, 2.28–2.15 ppm (triplet, Sn–CH₂–CH₂–CH₂–CH₃), 2.14–1.73 ppm (multiplet, Sn–CH₂–CH₂–CH₂–CH₃), 1.24–1.22 ppm (multiplet, Sn–CH₂–CH₂–CH₂–CH₃), and 0.99–0.86 ppm (triplet, Sn–CH₂–CH₂–CH₂–CH₃). ¹H NMR information also supported the IR data of the complexes (2–6).

3.5. ¹³C NMR Spectra. The ¹³C–¹H NMR spectrum of free ligand (1) showed the resonance signals at 185.20, 165.32, 145.30–136.21, and 10.45 ppm which were shifted to high frequency at 179.99–181.10 ppm in all the complexes (2–6) compared to ligand (1), indicating participation of the N=C=S group in coordination to tin(IV) atom. The chemical shifts of carbon in C=N and CH₃ in the free ligand (1) were observed at 165.32 and 10.45 ppm which were shifted to high frequency at 168.36–178.45 and 16.44–18.70 ppm, respectively, in the complexes (2–6). This results supported the azomethine-N is coordinated to the tin(IV) atom [30]. After complexation, the δ value of carbon atoms in the aromatic ring did not have much change in the complexes (2–6) as compared to the free ligand. Besides, the butyl group attached to the organotin(IV) moiety in complex 3 gave four resonance signals at 23.78, 26.31, 24.18, and 20.11 ppm. In the ¹³C–¹H NMR spectra of the organotin(IV) complexes 2 and 5, a sharp singlet resonance signal appeared at 12.80 ppm [(Sn–CH₃)] and 14.97 ppm [(Sn–CH₃)] respectively [31]. In organotin(IV) compounds, the ¹¹⁹Sn, ¹³C value is an important parameter to assess the coordination number of the Sn atom. The calculated coupling constants for dimethyltin(IV) (4) and diphenyltin(IV) (5) compounds were found to be 557 and 546 Hz, which described the penta-coordinate environment about the Sn atom in these compounds [32]. All these statements are also supported by the ¹H NMR spectra analyses.

3.6. ¹¹⁹Sn NMR Spectra. ¹¹⁹Sn NMR spectra can be used as an indicator of the coordination number of the tin atom. ¹¹⁹Sn NMR of all the complexes (2–6) shows only one resonance signals in the range of −149.60 to −185.32 ppm. ¹¹⁹Sn NMR values are characteristic for the five-coordinated tin atom observed in the organotin(IV) complexes (2–6) [33–36].

3.7. X-Ray Crystallography Diffraction Analysis. The molecular structure of the ligand (1) with atom numbering scheme is depicted in Figure 1. The main crystal parameters are reported in Table 1. Selected bond lengths and bond angles are given in Table 2. The compound crystallizes into monoclinic crystal system with a space group of P2₁/c. In the title substituted thiosemicarbazone, C₁₆H₁₇N₃OS, the hydroxy- and methyl-substituted benzene rings form dihedral angles of 9.62 (12) and 55.69 (6)°, respectively, with the central CN₃S chromophore (r.m.s. deviation = 0.0117 Å) in (C₁₆H₁₇N₃OS) (Figure 1) and the OH– and Me-benzene rings are twisted as seen in the respective dihedral angles of 9.62 (12) and 55.69 (6)°. The almost coplanarity of the central atoms is ascribed to the formation of an intramolecular hydroxyl–O–H···N-imine hydrogen bond (Table 3). The N1–N2 bond length (1.375 Å) is closer to single bond length (1.45 Å) than to double bond length (1.25 Å) [37]. The C9–S1 bond distance (1.694 Å) is close to that expected of a C=S double bond (1.60 Å) [37] and the C7–N1 bond length (1.295 Å) is nearly the same as that of the

| Compound | H₂dampt (1) |
|----------|-------------|
| Empirical formula | C₁₆H₁₇N₃OS |
| Formula weight | 299.39 |
| Temperature (K) | 100 (2) |
| Wavelength (Å) | 0.71073 |
| Crystal system | Monoclinic |
| Space group | P2₁/c |
| Unit cell dimensions | |
| a (Å) | 14.6966(8) |
| b (Å) | 7.3586(4) |
| c (Å) | 14.0926(8) |
| α (°) | 90.00 |
| β (°) | 94.358(5) |
| γ (°) | 90.00 |
| Volume (Å³) | 1519.66(15) |
| Z | 4 |
| Calculated density (mg/m3) | 1.309 |
| Radiation type λ (Å) | Mo Kα |
| F (000) | 632 |
| Crystal size (mm) | 0.30 × 0.1 × 0.05 |
| Crystal colour | Light-yellow |
| Scan range θ (°) | 2.8–29.3 |
| Absorption coefficient (μ) (mm⁻¹) | 0.225 |
| Max. and min. transm | 1.00 and 0.419 |
| Goodness of fit on F² | 0.995 |
| Data/restrains/ parameters | 3375/3/201 |
| Final R indices [I > 2σ(I)] | R₁ = 0.0599, wR₂ = 0.1324 |
| R indices (all data) | R₁ = 0.110, wR₂ = 0.1738 |
Table 2: Selected bond lengths (Å) and bond angles (°) of ligand [H$_2$dampt] (1).

| Bond lengths (Å)       |          | Bond angles (°)           |          |
|------------------------|----------|---------------------------|----------|
| S1–C9                  | 1.694 (3)| C9–N2–N1                  | 120.6 (2)|
| N1–C7                  | 1.295 (4)| N3–C9–N2                  | 113.2 (2)|
| N2–C9                  | 1.352 (4)| N2–C9–S1                  | 122.4 (2)|
| C7–C8                  | 1.500 (4)| O1–C1–C2                  | 116.8 (3)|
|                         |          | 123.2 (3)                 |          |

Table 3: Hydrogen-bond geometry (Å, °)

| D-H···A                | D-H    | H···A         | D···A     | D-H···A       |
|------------------------|--------|--------------|-----------|--------------|
| O1–H1ο···N1            | 0.84 (1)| 1.81 (2)     | 2.551 (3) | 145 (3)      |
| N2–H2n···S1i           | 0.88 (1)| 2.51 (2)     | 3.323 (2) | 154 (3)      |
| N3–H3n···S1i           | 0.88 (1)| 2.49 (2)     | 3.286 (3) | 151 (2)      |
| C8–H8b···Cg1i          | 0.98   | 2.59         | 3.501 (3) | 155          |

Symmetry codes: –x + 1, y + 1/2, –z + 1/2.

Figures

Figure 1: The molecular structure of H$_2$dampt (1) showing the atom-labelling scheme and displacement ellipsoids at the 50% probability level.

Figure 2: A view of the helical supramolecular chain aligned along the b axis in (1). The N–H···S hydrogen bonds are shown as orange dashed lines. Further stabilization to the chain is provided by C–H···π and π–π interactions, shown as blue and purple dashed lines, respectively.

3.8. Antibacterial Activity. The synthesized ligand (1) and its organotin(IV) complexes (2–6) were tested against Escherichia coli, Staphylococcus aureus, Enterobacter aerogenes, and Salmonella typhi bacterial strains for their antibacterial activity using agar-well diffusion method and data are shown in Table 4 and Figure 3. The doxycycline was used as a reference drug. The results showed that the substituted thiosemicarbazone ligand (1) possessed moderate antibacterial activity. The antibacterial studies of the compounds (2–6) showed relatively better activity against...
Table 4: Antibacterial activity\(^{a,b}\) of the free ligand (1) and its organotin(IV) complexes 2–6 (inhibition zone in mm).

| Bacterium          | Clinical implication                                      | Zone of Inhibition (mm) |
|--------------------|-----------------------------------------------------------|-------------------------|
| Escherichia coli   | Infection of wounds, urinary tract, and dysentery         | (1) 24 (2) 12 (3) 20 (4) 21 (5) 28 (6) 29 R 34 |
| Staphylococcus aureus | Food poisoning, scaled skin syndrome, endocarditis        | (2) 14 (3) 22 (4) 24 (5) — (6) 29 R 30 36 |
| Enterobacter aerogenes | Lower respiratory tract infections, skin and soft-tissue infections | — — (4) 20 (5) 22 (6) 20 22 26 |
| Salmonella typhi   | Typhoid fever, localized infection                         | (1) 21 — (4) 14 (5) 23 (6) 23 R 27 31 |

\(^{a}\) *In vitro*, agar-well diffusion method, conc. 200 mg/mL of DMSO. \(^{b}\) Reference drug (R), doxycycline, dash indicate inactivity.

4. Conclusion

The ligand (1) and its organotin(IV) complexes (2–6) have been synthesized and fully characterized by different spectroscopic techniques. The ligand (H2dampt) exists in thione form in a solid state but it takes on a thiol form when it is in solution. All organotin(IV) complexes (2–6) of H2dampt were proposed to be five coordinated and the ligand binds to the central tin(IV) atom in dinegative tridentate form. Single crystal X-ray analysis of newly synthesized ligand (1) has been reported. The *in vitro* antibacterial activities of the synthesized complexes against the selected bacterial strains have been established. All compounds have been found biologically active, while the studies have confirmed that compounds 5 and 6 are more active and have the potency to be used as antibacterial agents. Trials to obtain single crystals suitable for structure determination by X-ray crystallography were in vain due to the amorphous nature of the complexes.

Acknowledgment

This work was financially supported by the Ministry of Science Technology and Innovation (MOSTI) under a Research Grant (no. 06-01-09-SF0046). The authors would like to thank Universiti Malaysia Sarawak (UNIMAS) for the facilities to carry out the research work.

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