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Synthesis, Characterization and Antimicrobial Screening of Some Organotin Complexes Derived from Salicyl Hydrazide Schiff Bases

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A series of new organotin(IV) compounds [Me₂SnL], [Et₂SnL], [Bu₂SnL] and [Ph₂SnL], where L = 2-hydroxy-N-(1-(4-hydroxy-6-methyl-2-oxo-2H-pyran-3-yl)ethylidene)benzohydrazide has been synthesized by refluxing methyl salicylate hydrazide hydrate with dehydroacetic acid (DHA) with the corresponding organotin(IV) chlorides. All the compounds were characterized by elemental analyses, FT-IR, ^{1}H NMR, ^{13}C NMR, ^{119}Sn NMR and mass spectrometry. The synthesized complexes found to have wide therapeutic potential.

Keywords: Organotin complexes, Dehydroacetic acid, Antimicrobial activity.

INTRODUCTION

Studying the experimental strategies to elevate out chemical transformations in the subject of organotin chemistry. The biochemical exercise of organotin(IV) compounds is affected prominently through the shape of the molecule and the coordination number of the tin atom [1]. Further, organotin(IV) complexes owe their functionalities and usefulness towards the distinct properties because of the presence of different moieties within the molecule [2]. Among these, organotin(IV) complexes of Schiff base exhibits anti-inflammatory [3], anti-bacterial [4,5] anticancer [6-8] antimalarial, anticonvulsant and antitumor activities [9,10].

However, Schiff bases possesses nitrogen or oxygen as electron-donating atoms will allow either as multidentate or a bridging building block in structural assemblies. Furthermore, Schiff bases derived from dehydroacetic acid (DHA), (3-acetyl-4-hydroxy-6-methyl-2*H*-pyran-2-one), have been paid consideration because of their invigorating adaptable applications in different areas like catalysis, biotechnology, logical and therapeutic science [11]. It is widely used as fungicide, herbicide and as a preservative, which has powerful antimicrobial effect against bacteria, yeast and particularly moulds [12-14]. It is well known that the biological activity of organotin is related

to the type of alkyl groups attached to the tin atom. Present work outlines the series of organotin halides Schiff bases prepared by using (3-acetyl-4-hydroxy-6-methyl-2*H*-pyran-2-one), hydrazine and methyl salicylate (**Scheme-I**). All the organotin complexes have been characterized by FT-IR, NMR (¹H, ¹³C and ¹¹⁹Sn), mass spectrometry and also evaluated for their antimicrobial activity.

EXPERIMENTAL

Organotin halides *viz*. dimethyltin dichloride, diethyltin dichloride, dibutyltindichloride, diphenyltindichloride were procured from Sigma-Aldrich, whereas hydrazine hydrate, triethylamine and 3-acetyl-4-hydroxy-6-methyl-2*H*-pyran-2-one (DHA) were obtained from Merck. Purity was checked through sharp melting points/TLC. Solvents were dried and purified according to standard procedures [15] and the moisture was excluded from the glass apparatus using CaCl₂ drying tubes. Methanol was dried using molecular sieves and further by distillation. Melting points were determined by using the open glass capillaries and were uncorrected. FT-IR spectra was recorded on a Perkin-Elmer System 2000 spectrophotometer in KBr pellet in 4000-400 cm⁻¹ range at room temperature. ¹H, ¹³C and ¹¹⁹Sn NMR spectra were recorded on Bruker 500 MHz NMR spectrophotometer. ¹H & ¹³C chemical shifts were determined

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Scheme-I: Synthesis of hydrazide ligand

comparative to internal TMS whereas in 119 Sn NMR tetraphenyltin(IV) was used as an external reference. Mass spectra (m/z) was made by using GC-MS from SAIF LAB, Chandigarh, India.

Synthesis of 2-hydroxy-*N*-(1-(4-hydroxy-6-methyl-2oxo-2H-pyran-3-yl)ethylidene)benzohydrazide (L): An ethanolic solution of methyl salicylate (38.44 mmol) was slowly added to the solution of hydrazine hydrate (38.44 mmol) in ethanol in 1: 1 ratio with constant stirring at room temperature for 1 h and left for overnight. The obtained white crystals were recrystallized using absolute ethanol. This resultant hydrazide (1.31 mmol) was treated with dehydroacetic acid (1.31 mmol) in dry methanol in 1: 1 ratio. Further, the mixture was refluxed for 4 h and yellow crystalline solid was obtained after cooling, purity of synthesized compound was checked by TLC and melting point (**Scheme-I**). Yield 83%, m.p.: 170-175 °C. Anal. calcd. (found) % for $C_{15}H_{14}N_2O_5$ (m.w. = 302.09): C, 59.60 (59.63); H, 4.67 (4.71); N, 9.27 (9.29); O, 26.46 (26.40). GC-MS (m/z) calcd. (found) 302.09 (302). FT-IR (KBr, v_{max} , cm⁻¹): 3061 (N-H); 3252 (O-H); 1709 (C=O); 1666 (C=N); 1067 (N-N) and 1153 (C-O-C). 1 H NMR (δ ppm): H-4: 14 (s, 1H, OH), H-5: 6.2 (s, 1H, phenyl), H-7: 1.3 (s, 3H, CH₃) H-9: 0.9 (s, 3H, CH₃) H-11: 5.7 (s, 1H, NH), H-15: 13.0 (s, 1H, OH), H-16-19: 6.91-7.2 (m, 4H, phenyl). 13 C NMR (δ ppm): C-1: 162; C-2: 103; C-3: 177; C-5: 102; C-6: 163; C-7: 21; C-8: 155; C-9: 11; C-12; 163; C-13: 119; C-14: 159; C-16: 116; C-17: 130; C-18: 128; C-19: 120.

Synthesis of organotin(IV) complexes

Synthesis of dimethyltin(IV) Schiff base complex: A 250 mL two-necked flask containing 30 mL benzene, equipped with a reflux condenser was charged with 2-hydroxy-*N*-(1-(4-hydroxy-6-methyl-2oxo-2*H*-pyran-3-yl)ethylidene)benzo-hydrazide (1.65 mmol) and triethylamine (3.3 mmol) along with a magnetic bar in 1:2 ratio. To a solution of triethylammonium salt of ligand, dimethyltin(IV) dichloride (1.65 mmol) in dry methanol was added dropwise in the flask with continuous

 R_2 SnCl₂

(21*E*)-2-hydroxy-*N*'-(1-(4-hydroxy-6-methyl-2-oxo-2*H*-pyran-3-yl)ethylidene)benzohydrazide

stirring at room temperature. When the solution turned yellow, then it was stirred and refluxed for 5 h at it's refluxed temperature. The white precipitates of Et₃NHCl formed during the reaction and was removed by filtration. The filtrate was concentrated using rotary evaporator yielding yellow solid and collected after recrystallization. Purity was checked by TLC and melting point was measured in open capillary (Scheme-II). Yield 73%, m.p.: 168-170 °C. Anal. calcd. (found) % for C₁₇H₁₈N₂O₅Sn (m.w. = 450.02): C, 45.47 (45.43); H, 4.04 (4.02); N, 6.24 (6.22); O, 17.81 (17.80); Sn, 26.44 (26.41). GC-MS (*m/z*) calcd. (found) 450.02 (450.01). FT-IR (KBr, v_{max} , cm⁻¹): 1669 (C=O); 1645 (C=N); 1072 (N-N); 1150 (C-O-C); 578 (Sn-O); 475 (Sn-N). ¹H NMR (DMSO) (δ ppm): H-5: 6.3 (s, 1H, phenyl) H-7: 1.5 (s, 3H, CH₃) H-9: 1.9 (s, 3H, CH₃) H-11: 6.0 $(s, 1H, NH) H-16-19: 6-7.7 (m, 4H, phenyl), H-\alpha: 0.82 (s, 6H, phenyl)$ 2CH₃), ¹³C NMR (DMSO) (δ ppm): C-1: 164; C-2: 108; C-3: 178; C-5: 98; C-6: 167; C-7: 22 (CH₃); C-8: 167.7 (C=N); C-9: 47 (CH₃); C-12: 162.73; C-13: 117; C-14: 159; C-16: 117; C-17: 133; C-18: 119; C-19: 128; C-α; 2.4. 119 Sn NMR (DMSO) $(\delta ppm) = -266.$

Synthesis of diethyltin(IV) Schiff base complex: The compound was synthesized by adopting above said using following precursors quantities: 2-hydroxy-N-(1-(4-hydroxy-6-methyl-2-oxo-2*H*-pyran-3-yl)ethylidene)benzohydrazide (0.66 mmol), diethyltindichloride (0.66) and triethylamine (1.32 mmol) in 1:1:2 ratio. Yield 70%, m.p.: 182-185 °C, Anal. calcd. (found) % for $C_{19}H_{22}N_2O_5Sn$ (*m.w.* = 477.1): C, 47.83 (47.81); H, 4.65 (4.62); N, 5.87 (5.83); O, 16.77 (16.75); Sn, 24.88 (24.89). GC-MS (m/z) calcd. (found) 478.06 (478). FT-IR (KBr, v_{max} , cm⁻¹): 1667 (C=O); 1643 (C=N); 1076 (N-N); 1155 (C-O-C); 573 (Sn-O); 450 (Sn-N). 1 H NMR (DMSO) (δ ppm): H-5: 6.01 (s, 1H, phenyl) H-7: 1.6 (s, 3H, CH₃) H-9: 2.1 (s, 3H, CH₃) H-11: 5.9 (s, 1H, NH) H-16-19: 6.2-7.9 (m, 4H, phenyl), H-α: 1.54 (q, 4H, 2CH₂), H-β: 1.28 (t, 6H, 2CH₃), ¹³C (DMSO) (δ ppm): C-1: 164; C-2: 107; C-3: 178; C-5: 99; C-6: 167; C-7: 21 (CH₃); C-8: 167.7 (C=N); C-9: 43 (CH₃); C-12: 161.73; C-13: 117; C-14: 159; C-16: 117; C-17: 133; C-

Scheme-II: Synthesis of organotin complexes

18: 119; C-19: 128; C-α; 13.7, C-β; 9.3 ¹¹⁹Sn NMR: (DMSO) (δ ppm) = -329.

Synthesis of dibutyltin(IV) Schiff base complex: The compound was prepared by using same procedure adopted for the synthesis of dimethyl derivative, using following precursors quantities: 2-hydroxy-N-(1-(4-hydroxy-6-methyl-2-oxo-2*H*-pyran-3-yl)-ethylidene)benzohydrazide (1.32 mmol), dibutyltin dichloride (1.32 mmol) and triethylamine (2.64 mmol) in 1:1:2 ratio. The product was recrystallized. Purity was checked by TLC and melting point. Yield 80%, m.p.: 220-224 °C; Anal. calcd. (found) % for $C_{23}H_{30}N_2O_5Sn$ (*m.w.* = 534.12): C, 51.81 (51.80); H, 5.67 (5.63); N, 5.25 (5.22); O, 15.00 (15.01); Sn, 22.26 (22.23). GC-MS (*m/z*) calcd. (found) 534.12 (534). FT-IR (KBr, v_{max} , cm⁻¹): 1667 (C=O); 1640 (C=N); 1076 (N-N); 1156 (C-O-C); 569 (Sn-O); 456 (Sn-N). ¹H NMR (DMSO) (δ ppm): H-5: 6.9 (s, 1H, phenyl) H-7: 1.42 (s, 3H, CH₃) H-9: 2.1 (s, 3H, CH₃) H-11: 5.8 (s, 1H, NH), H-16-19: 6.1-7.2 (m, 4H, phenyl) $H\alpha$ -1.4 (m, 4H, 2CH₂), $H\beta$ -1.4 (m, 4H, 2CH₂), H γ -1.25 (m, 4H, 2CH₂), H δ -0.7 (t, 6H, 2CH₃). ¹³C NMR (DMSO) (δ ppm): C-1: 163; C-2: 106; C-3: 178; C-5: 98; C-6: 167; C-7: 21(CH₃); C-8: 167.7 (C=N); C-9: 40 (CH₃); C-12: 163.73; C-13: 117; C-14: 159; C-16: 116; C-17: 133; C-18: 119; C-19: 128; C- α ; 13, C- β ; 39, C- γ ; 27, C- δ -25. ¹¹⁹Sn NMR: (DMSO) (δ ppm) = -325.

Synthesis of diphenyltin(IV) Schiff base complex: The compound was prepared by using above mentioned using following precursors quantities: 2-hydroxy-N-(1-(4-hydroxy-6-methyl-2-oxo-2*H*-pyran-3-yl)ethylidene)benzohydrazide (1.32 mmol), diphenyltin dichloride (1.32 mmol) and triethylamine (2.64 mmol) in 1:1:2 ratio. The product was recrystallized. Purity was checked by TLC and melting point. Yield 80%, Anal. calcd. (found) % for $C_{27}H_{22}N_2O_5Sn$ (*m.w.* = 573.18): C, 56.58 (56.57); H, 3.87 (3.85); N, 4.89 (4.90); O, 13.96 (13.95); Sn, 20.71 (20.71). GC-MS (*m/z*) calcd. (found) 574.06 (574). FT-IR (KBr, v_{max} , cm⁻¹): 1667 (C=O); 1640 (C=N); 1074 (N-N); 1154 (C-O-C), 570 (Sn-O), 476 (Sn-N). 1 H NMR (DMSO) (δ ppm): H-5: 6.4 (s, 1H, phenyl) H-7: 1.7 (s, 3H, CH₃) H-9: 2.0 (s, 3H, CH₃) H-11: 5.9 (s, 1H, NH) H-16-19: 6-7.9 (m, 4H, phenyl), 6.40-7.75 (m, overlapping of protons in aromatic ring and protons of phenyl ring attached to the tin(IV) atom). ¹³C NMR (DMSO) (δ ppm): C-1: 164; C-2: 108; C-3: 178; C-5: 98; C-6: 167; C-7: 22 (CH₃); C-8: 168.7 (C=N); C-9: 45 (CH₃); C-12: 162.73; C-13: 117; C-14: 159; C-16: 117; C-17: 133; C-18: 119; C-19: 128; C- α ; 128, C- β ; 137, C- γ ; 128, C- δ -128. ¹¹⁹Sn NMR: (DMSO) (δ ppm) = -222.

Antimicrobial activity: in vitro antimicrobial activity of the organotin complexes against the selected bacterial strain (Staphylococcus aureus and Klebsiella pneumoniae) and fungal strain (Aspergillus niger and Trichophyton rubrum) was investigated by well plate diffusion method. The sterilized petri plates (150 mm in diameter) were used throughout the investigation [16]. All the materials were sterilized at 15 psi for 45 min in autoclave. The melted nutrient agar and potato dextrose agar used as medium for bacteria and fungi, respectively. After the solidification of pour plates bacteria and fungi under investigation were separately spread uniformly over the plates with the help of sterilized glass spreader. In each case, control plate

was also maintained with DMSO. The activity was expressed in the terms of the zone of inhibition in mm. The various concentrations of the complexes *viz.* 100, 250, 400 ppm were loaded in wells followed by incubation at 30 °C for 24 and 72 h to evaluate the effect of compounds on bacterial and fungal growth, respectively [17]. Commercial antifungal fluconazole and antibacterial neomycin were used as standard drugs. The experiment run in triplicate and the result was observed with deviations.

RESULTS AND DISCUSSION

IR spectra: Solid state infrared spectra of the ligand and their complexes have been recorded with KBr pellets in the range 4000-400 cm⁻¹. The principal absorptions of ν (O-H), v(C=O), v(N-H) and v(C=N) stretching vibrations of the ligand and complexes were examined by comparing the IR spectra of the free ligand with their corresponding diorganotin(IV) complexes, which illustrated the differences between them. In the IR spectrum of ligand (L) absorption bands were observed at 3252, 3061, 1709, 1666, 1067, 1153 cm⁻¹ due to the stretching vibrations of the O-H, N-H, C=O and C=N, N-N, C-O-C groups, respectively. The significant differences in the IR spectra of ligand and complexes were the disappearance of v(N-H) and ν (O-H) bands due to deprotonation and enolization of the ligand, thus suggested the coordination of phenolic oxygens to the diorganotin(IV) moiety [18]. Whereas C-O-C band remains the same in the complexes. Further, the v(C=N) band is shifted to lower frequency with respect to the ligand indicating the coordination of azomethine nitrogen to tin center [19]. Whereas, the band at 1067 cm⁻¹ due to v(N-N) is shifted to higher frequencies at 1076-1072 cm⁻¹ in the spectra of organotin(IV) complexes concluded that the increase in the frequency of this band is accredited to an increase in bond length and decrease in repulsion of the lone pairs of electrons present on the nitrogen atoms [20,21]. The IR bands observed in the regions of 578-569 and 475-450 cm⁻¹ indicated the presence of Sn-O and Sn-N bonds, respectively, which support the formation of diorganotin (IV) complexes [22].

NMR spectra: The ¹H NMR signals at 14 ppm and 5.7 ppm in the ligand are due to -OH and -NH, respectively [23]. In the diorgnaotin(IV) complexes, all these signals were disappeared due to subsequent deprotonation of hydroxyl group. Proton attached to the C5 carbon of DHA ring appeared as a sharp singlet in the range δ 6.0-6.9 ppm [24]. Similarly, methyl protons attached to C6 carbon appeared as a singlet at δ 1.4-1.7 ppm, which remained almost unchanged on complexation, whereas methyl protons attached to azomethine carbon (C9) appeared at δ 0.9 ppm in the ligand and shifted downfield in the complexes, suggesting the participation of azomethine nitrogen in bond formation [19]. In addition, the formation of complexes was supported by appearance of new signals at the range of δ 0.7-1.4, δ 1.2-1.54, δ 0.70-1.4 and δ 6.4-7.7 ppm due to methyl, ethyl, butyl and phenyl protons, respectively which are attached directly to the tin atom [25]. In the ¹³C NMR spectrum of the ligand, C-8 (H-C=N) and C-12 (CO-NH) were appeared at 155 and 163 ppm, respectively. The complexes showed a downfield shift of phenyl carbon resonances, especially C-2 due to the formation of Sn-O bond [26]. The ¹¹⁹Sn

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NMR resonances for dimethyl, diethyl, dibutyl and diphenyl complexes were observed at -266, -329, -325 and -222 ppm, respectively [27,28]. The ¹¹⁹Sn NMR chemical shifts for all the complexes demonstrated the presence of hexa-coordinate tin(IV) cores.

Mass spectra: The molecular mass observed at m/z was 450, 477, 534 and 573 for dimethyl, diethyl, dibutyl and diphenyltin derivatives, respectively which has been found in full agreement with the proposed structure. The mass fragmentation pattern of the butyl derivative has been given in Fig. 1. Primary decomposition seems to be due to the loss of the H⁺, OH⁻ ion.

Antimicrobial assay: In the present study, the synthesized organotin(IV) complexes were investigated against the selected fungi viz. Aspergillus niger and Trichophyton rubrum and bacteria viz. Staphylococcus aureus and Klebsiella pneumoniae. It was observed that all the complexes quenched the multiplication of the selected fungal and bacterial strains with some deviations and this deviation can be subjected to the diversity in the ribosome and impenetrability of the cell membrane of microorganisms. This transformation can be elucidated employing Tweedy's chelation theory, which considers that the overlapping of orbitals of ligand reduced the polarity of tin ion which increases the lipophilicity character of complexes.

Further, increment in the lipophilicity character results in more penetration of Sn atom across the cell membrane of microorganism resulting the potent antimicrobial property of organotin complexes. Moreover, concentration is also one of the important factors to calculate the zone inhibition and it has been observed that the synthesized compounds showed maximum activity at 400 ppm. The experiment performed in triplet, the average mean value $\pm\,0.5$ to $\pm\,0.9$ standard deviation of three values is represented are data given in Table-1.

Conclusion

In this work, four new diorganotin(IV) derivatives of *N*-substituted, Schiff base 2-hydroxy-*N*-(1-(4-hydroxy-6-methyl-20x0-2*H*-pyran-3-yl)ethylidene)benzohydrazide were synthesized and characterized. It was concluded that the ligands act as bidentate forming distorted octahedral complexes with organotin chlorides. Furthermore, the present study strongly establishes that these organotin complexes are more effective antibacterial agents than the parent ligands.

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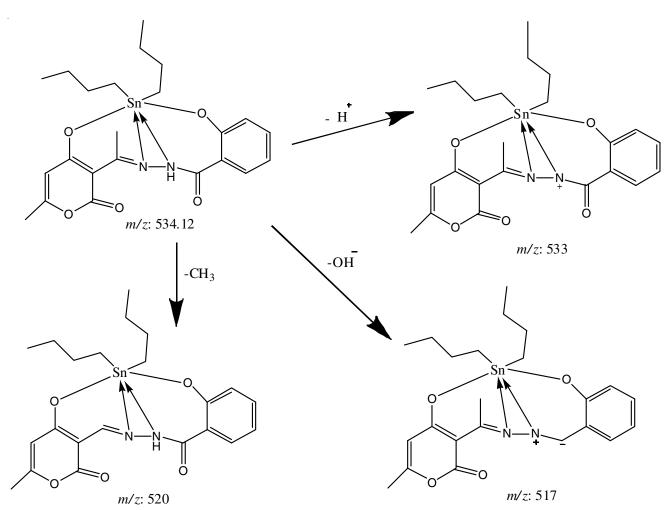


Fig. 1. Mass fragmentation pattern of C₂₃H₃₀N₂O₅Sn

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$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$		19 mm (Neomycin)			17 mm (Neomycin)			21 mm (Fluconazole)			19 mm (Fluconazole)		
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Standard 250 ppm 250 ppm 250 ppm 250 ppm		08± 0.6	11±0.8	15±0.7	12±0.6	15±0.6	18±0.8	11±0.6	13±0.6	16±0.6	12±0.7	14±0.5	17±0.6
Standard 22 (Magazzain) 24 (Flygorogale) 22 (Flygorogale)	Standard							**			• •		

21 (Neomycin)

TABLE-1

CONFLICT OF INTEREST

22 (Neomycin)

The authors declare that there is no conflict of interests regarding the publication of this article.

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24 (Fluconazole)

22 (Fluconazole)

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