Synthesis and Characterization of Organotin(IV) Complexes Derived of 2-amino-5-nitrobenzoic Acid: In vitro Antibacterial Screening Activity

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Abstract: Problem statement: Tremendous studies have been carried out on organotin(IV) complexes derivatives of carboxylate anions. However, the synthesis and characterization as well as the in vitro antibacterial screening activity of organotin(IV) carboxylate derived of 2-amino-5-nitrobenzoic acid have not been carried out. Approach: Organotin(IV) carboxylate complexes derivative of 2-amino-5-nitrobenzoic acid, 2-NH₂-5-NO₂-C₆H₃COOH have been successfully synthesized. The acid and complexes obtained were characterized quantitatively using C, H, N and Sn elemental analysis as well as spectroscopic methods such as infrared (FTIR) and nuclear magnetic resonance (¹H, ¹³C, ¹H-¹³C HMQC and ¹¹⁹Sn NMR). Moreover, the complexes obtained were screened for their in vitro antibacterial screening activity. Results: Monomeric R₂Sn (2-NH₂-5-NO₂-C₆H₃COO)₂ (R = methyl 1, butyl 2) and dimeric [(Bu₂Sn(2-NH₂-5-NO₂-C₆H₃COO)₂)₂O]₃ as well as Ph₃Sn(2-NH₂-5-NO₂-C₆H₃COO)₄ are obtained in solid state. Results of the infrared spectroscopy on the acid and complexes showed that the coordination took place via oxygen atoms from the carboxylate anions. Based on the ¹¹⁹Sn NMR solution study, the tin atom of both complexes 1 and 2 exhibit six-coordination respectively and complex 3 exhibits five- and six-coordination whereas the tin atom of complex 4 exhibits five-coordination. Conclusion: Pure complexes derived of 2-amino-5-nitrobenzoic acid have been successfully obtained. Triphenyltin(IV) are found to possess better in vitro antibacterial screening activity on two gram-positive bacterial compared to the parent acid.

Key words: Organotin(IV) carboxylate, preparation, antibacterial activity

INTRODUCTION

Organotin(IV) complexes are extensively studied due to the applications in industrial as well as biocidal properties (Molloy et al., 1984; Willem et al., 1997; Gielen et al., 2000). Numerous studies on organotin(IV) complexes have been carried out in order to study its biological properties against bacterial, fungal and cancer cells line (Teoh et al., 1997; Novelli et al., 1999; Gielen et al., 2000; Crouse et al., 2004). Up to date, organotin(IV) complexes are still extensively studied due to its coordination geometries as well as structural diversity (monomer, dimeric, hexameric and oligomeric) (Zhang et al., 2005; Win et al., 2007a; 2008; Amini et al., 2009).

In this study, we are focus on synthesis and structural characterization of new organotin(IV) carboxylate complexes derived from 2-amino-5-nitrobenzoic acid. In addition, the in vitro antibacterial screening activities of the complexes obtained are carried out and the results are reported herein.

MATERIALS AND METHODS

General and instrumental: All the reagents, starting materials as well as the solvents were purchased commercially and used without any further purification. The melting points were determined in an open capillary and are uncorrected. Elemental C, H and N analyses were carried out on a Perkin-Elmer 2400 CHN Elemental Analyzer. Tin was determined gravimetrically by igniting a known quantity of each complex to SnO₂. Infrared spectra were recorded using a Perkin-Elmer System 2000 FTIR Spectrophotometer.
as a KBr disc in the frequency range of 4000-400 cm\(^{-1}\). The spectra for \(^1\)H, \(^13\)C HMQC and \(^{119}\)Sn NMR were recorded on a Bruker AC-P 400 MHz FTNMR Spectrometer and \(^{13}\)C NMR was recorded on a Bruker AC-P 300MHz FTNMR Spectrometer using deuterated d\(_6\)-DMSO as the solvent and tetramethylylsilane, TMS as the internal standard.

**In vitro antibacterial screening activity:** The synthesized complexes and parent acid were screened for their *in vitro* antibacterial activity against three gram-negative (*Escherichia coli*, *Pseudomonas aeruginosa* and *Klebsiella pneumonia*) and two gram-positive (*Bacillus subtilis* and *Staphylococcus aureus*) bacterial strains, by Inhibition Zone Method using agar well diffusion method. The seeded agar (nutrient agar medium) was prepared by cooling the molten agar to approximately 40°C and then adding bacterial inoculums containing approximately 10\(^2\)-10\(^3\) Colony Forming Units (CFU) mL\(^{-1}\). The bacterial inoculums were spread on the plate containing agar medium and even coverage was ensured before the agar solidified. The complexes were dissolved in DMSO to prepare 1.0 mg mL\(^{-1}\) concentration. By using a sterile metallic borer, the wells (6 mm in diameter) were dug and the standard drugs and complexes were introduced into the respective wells. The plates were incubated immediately at 37°C for 20-24 h. The activity was determined by measuring the diameter of the inhibition zone (in mm).

**Preparation of sodium salt and dimethyltin(IV) oxide, Me\(_2\)SnO:** Dimethyltin(IV) dichloride, Me\(_2\)SnCl\(_2\) was dissolve in distilled water and stirred for overnight. Colorless solution was obtained. Ammonia solution (60\%) was added into the colorless solution and finally fine white precipitate was obtained and filtered. The precipitate was dried in oven (60°C) for a day until dry qualitative. Then, 2-amino-5-nitrobenzoic acid dissolved in ethanol (30 mL) added to the dibutyltin(IV) oxide (20 mL) and heated for an hour until clear solution was obtained. After two weeks, yellow crystals (1.60 g, 404\%) was added into the colorless solution and finally a yellowish precipitate was obtained. Then, 2-amino-5-nitrobenzoic acid dissolved in DMSO to prepare 1.0 mg mL\(^{-1}\) and acid (0.363 g, 2 mmole) in ethanol (50 mL) for an hour. A clear yellow transparent solution was separated by filtration and kept in a bottle. After four days, fine yellow solids (0.45 g, 88.0\% yield) were collected. Melting point: >300°C (decomposed). Analysis for C\(_{16}\)H\(_{16}\)N\(_2\)O\(_4\)Sn: C, 37.66; H, 3.01; N, 11.04; Sn, 23.26\%.

Complex 2 was obtained by heating under reflux a 1:2 molar mixture of dibutyltin(IV) oxide (0.75 g, 3 mmole) and acid (1.09 g, 6 mmole) in methanol (50 mL) for 4 h. After two weeks, yellow crystals (1.60 g, 90.0\% yield) were collected. Melting point: 209.3-209.7°C. Analysis for C\(_{22}\)H\(_{28}\)N\(_2\)O\(_4\)Sn: C, 44.42; H, 4.19; N, 9.38; Sn, 19.91\%.

Complex 3 was prepared from a 1:1 molar mixture of dibutyltin(IV) oxide (0.50 g, 2 mmole) and 2-amino-5-nitrobenzoic acid (0.36 g, 2 mmole) in ethanol (50 mL). Dibutyltin(IV) oxide was first dissolved in ethanol (20 mL) and heated for an hour until clear solution was obtained. Then, 2-amino-5-nitrobenzoic acid dissolved in ethanol (30 mL) added to the dibutyltin(IV) oxide solution. The resulting mixture was heated under reflux

\(\text{Bis(2-amino-5-nitrobenzoato)dibutyltin(IV), Bu}_2\text{Sn(2-NH}_2\text{-5-NO}_2\text{-C}_4\text{H}_4\text{COO}_2\text{)}\) (2)

\(\text{Bis(2-amino-5-nitrobenzoato)tetrabutyldistannoxane (IV) dimer, [([Bu}_2\text{Sn(2-NH}_2\text{-5-NO}_2\text{-C}_4\text{H}_4\text{COO}_2\text{)}])_2O}_2\) (3)
for 2 h. A clear yellow transparent solution was isolated by filtration and kept in a bottle. After four days, yellow solids (0.61 g, 73.0% yield) were collected. Melting point: 240.7-241.5°C. Analysis for C$_6$H$_2$N$_2$O$_4$Sn: C, 42.74; H, 5.79; Sn, 27.98%. Calculated for C$_6$H$_2$N$_2$O$_4$Sn: C, 42.66; H, 5.49; N, 6.46; Sn, 28.12%. FTIR as KBr disc (cm$^{-1}$): ν(NH)$_2$ 3457, 3344, 3314; ν(C-H) aromatic 3059, ν(C-H) aromatic saturated 2956, 2926, 2870; ν(COO)$_{as}$ 1622, ν(COO)$_{s}$ 1310, ν(NO$_2$) 1537, ν(Sn-O-Sn) 3314, ν(Sn-C) 531, ν(Sn-O) 391. $^1$H-NMR (ppm) (d$_6$-DMSO): δ: benzene protons 6.92 (d, 9.3 Hz, 4H); 8.12 (dd, 2.4 Hz, 9.2 Hz, 4H); 8.72 (s, 4H); butyl, CH$_2$ 0.84 (t, 7.3 Hz, 12H); 0.90 (t, 7.3 Hz, 12H); CH$_2$ 1.28-1.43 (m, 32H); CH$_3$ 1.64-1.80 (m, 15H). $^{13}$C-NMR (ppm) (d$_6$-DMSO): δ: benzene carbons 112.27, 116.50, 124.89, 129.47, 135.56, 156.49; butyl carbons 13.71, 13.91, 26.09, 26.77, 27.02, 27.29, 29.90; COO 172.11. $^{119}$Sn-NMR (ppm) (d$_6$-DMSO): δ: -173.87, -213.71.

2-Amino-5-nitrobenzoatotriphenyltin(IV), Ph$_3$Sn(2-NH$_2$-5-NO$_2$-C$_6$H$_5$COO) (4)

Complex 4 was obtained by heating under reflux a 1:1 molar mixture of triphenyltin(IV) hydroxide (0.73 g, 2 mmole) and 2-amino-5-nitrobenzoic acid (0.36 g, 2 mmole) in methanol (60 mL) for an hour. A clear yellow transparent solution was separated by filtration and kept in a bottle. After six days, yellow crystals (0.51 g, 96.0% yield) were collected. Melting point: 208.5-208.9°C. Analysis for C$_{25}$H$_{20}$N$_2$O$_4$Sn: C, 56.41; H, 3.48; N, 5.23; Sn, 22.03%. Calculated for C$_{25}$H$_{20}$N$_2$O$_4$Sn: C, 56.53; H, 3.80; N, 5.27; Sn, 22.35%. FTIR as KBr disc (cm$^{-1}$): ν(NH)$_2$ 3442, 3328; ν(C-H) aromatic 3058, ν(COO)$_{as}$ 1618, ν(COO)$_{s}$ 1310, ν(NO$_2$) 1556, ν(Sn-O) 443. $^1$H-NMR (ppm) (d$_6$-DMSO): δ: phenyl protons 7.53-7.59 (m, 9H); 7.95-7.97 *(m, 6H); benzene 6.82 (d, 9.2 Hz, 1H); 8.07 (dd, 2.8 Hz, 9.2 Hz, 1H); 8.73 (d, 2.9 Hz, 1H); $^{13}$C-NMR (ppm) (d$_6$-DMSO): δ: phenyl carbons C$_{ipso}$ 142.87 (839.3 Hz), C$_{ortho}$ 136.11 (45.6 Hz), C$_{meta}$ 128.43, C$_{para}$ 129.03 (18.4 Hz); benzene 112.77, 115.75, 127.72, 127.97, 134.89, 155.92; COO 169.52. $^{119}$Sn-NMR (ppm) (d$_6$-DMSO): δ: -265.89.

2-Amino-5-nitrobenzoic acid: The parent acid, 2-amino-5-nitrobenzoic acid, 2-NH$_2$-5-NO$_2$-C$_6$H$_5$COOH was purchased from Acros Organics and used without any further purification. FTIR as KBr disc (cm$^{-1}$): selected data: ν(OH) 2892-2616, ν(COO)$_{as}$ 1685, ν(COO)$_{s}$ 1330. $^1$H-NMR (ppm) (d$_6$-DMSO): δ: benzene protons 6.96 (d, 9.3 Hz, 1H); 8.17 (dd, 2.8 Hz, 9.3 Hz, 1H); 8.68 (d, 2.8 Hz, 1H). $^{13}$C-NMR (ppm) (d$_6$-DMSO): δ: benzene carbons 109.31, 117.34, 129.54, 129.58, 135.87, 156.99; COO 168.95.

RESULT

Physical and elemental analysis: Elemental analysis C, H, N and Sn data obtained were in agreement with the predicted formula and complexes 1-4 gave a sharp melting point indicated the isolation of fairly pure complexes.

Structural and in vitro antibacterial screening activity: An outline of the proposed structure for complexes 1-4 are depicted in Fig. 1. The in vitro antibacterial screening activity of complexes 1-4 are given in Table 1.

![Fig. 1: The proposed structure for complexes 1-4](image)

| Complexes | Bacillus subtilis | Escherichia coli | Klebsiella pneumonia | Pseudomonas aeruginosa | Staphylococcus aureus |
|-----------|-----------------|-----------------|---------------------|-----------------------|---------------------|
|            | Inhibition zone (mm) |          |                     |                       |                     |
| 1          | 10              | 9               | 12                  | 10                    | 12                  |
| 2          | 13              | 9               | -                   | -                     | -                   |
| 3          | 13              | 7               | -                   | -                     | 11                  |
| 4          | 18              | -               | 23                  | -                     | 19                  |
| Chloramphenicol | 29           | -               | 23                  | 34                    | 30                  |
| Doxycycline | 34              | 24              | 21                  | 40                    | 28                  |
| Rifampicin | 25              | 24              | 23                  | 29                    | 37                  |

*Agar well diffusion method (in vitro) = 1.0 mg mL$^{-1}$; Reference drug = Chloramphenicol, Doxycycline and Rifampicin

Table 1: In vitro antibacterial screening activity of parent acid and complexes 1-4
DISCUSSION

In this study, complexes 1-4 derived of 2-amino-5-nitrobenzoic acid have been obtained in solid state. Complexes 2 and 4 were obtained as single yellow crystals and the X-ray crystal structure of both complexes have been reported (Win et al., 2006; 2007b).

The $\nu$(O-H) bands which appeared in the range of 2892-2616 cm$^{-1}$ for the acid, were absent in the infrared spectra of salt and complexes 1-4 showed the deprotonation and coordination of the carboxylate anion. The infrared spectra of complexes 1-4 revealed that the $\nu$(COO)$_\text{as}$ was shifted to a lower wave length number compared to the parent acid which signify that the coordination took place via the oxygen atoms of the carboxylate anion. Complex 4 showed the $\nu$(COO)$_\text{as}$ and $\nu$(COO), are in the range of 1618-1626 and 1310-1315 cm$^{-1}$ respectively.

Generally, the $\Delta \nu = [\nu$(COO)$_\text{as}$ - $\nu$(COO)$_\text{s}$] value is used to determine the bonding properties of carboxylate anion to tin atom in organotin(IV) carboxylate complexes. Sandhu and Verma (1987) in their studies and reports have shown that the $\Delta \nu$ value of complexes greater by 65-90 cm$^{-1}$ than in their sodium salts indicates either asymmetric or monodentate bonding of the carboxylate group to tin(IV) atom. Complexes 1-3 showed that the $\Delta \nu$ is comparable to the sodium salt ($\Delta \nu = 303$ cm$^{-1}$) indicating bidentate bonding of the carboxylate group to tin(IV) atom. Moreover, for complexes derived from triphenyltin(IV) carboxylate, $\Delta \nu$ below 200 cm$^{-1}$ would be expected for bridging or chelating carboxylates, but greater than 200 cm$^{-1}$ for the monodentate bonding carboxylate anions (Yeap and Teoh, 2003). Hence, carboxylate anion in complex 4 would be expected to bond to the tin atom in monodentate manner since the $\Delta \nu$ above 200 cm$^{-1}$.

Based on the infrared spectroscopy study, both complexes 1 and 2 exhibit six-coordinated tin atom; complex 3 exhibits five- and six-coordinated whereas complex 4 exhibits four-coordinated tin atom.

The upfield regions of the $^1$H NMR spectra of the complexes 1-3 showed the signal of the methyl and butyl protons in the range of 0.93 and 0.84-1.80 ppm respectively. In addition, complex 1 showed $^3$J($^{119}$Sn-$^1$H) at 95.2 Hz and based on the application of the Lockhart-Manders equation, the C-Sn-C angle is 153.61° (Lockhart and Manders, 1986). Based on the $^3$J($^{119}$Sn-$^1$H) and C-Sn-C angle, the tin atom of complex 1 is believed to exist in distorted octahedral geometry and six-coordinated. In general, complex 3 is one of the distannoxane dimer types and should exhibited two unresolved sets of butyl signals, one of the butyl groups linked to the endo-cyclic tin atom and the other one linked to the exo-cyclic tin atom respectively (Danish et al., 1995). However, complex 3 only showed two unresolved sets of CH$_3$ signal at 0.84 and 0.90 ppm respectively and two set of methylene signals of butyl groups in the range of 1.28-1.43 and 1.64-1.80 ppm in the spectra. This may due to a very similar environment or overlapping of methylene signals multiplicity in the NMR spectra. For complex 4, the resonances appearing as two well separated sets of multiplets in the regions centering around δ= 7.55 ppm and 7.96 ppm ascribed to phenyl protons. At the low field arising from ortho and at higher field arising from meta and para phenyl protons respectively (Sau and Holmes, 1981).

Evidence of the formation of the complexes is clearly displayed in the $^{13}$C NMR spectra. The $^{13}$C NMR spectra of complexes 1-4 showed the $\delta$(COO) signal shifted to the downfield region which is lower compared to that of the acid (168.95 ppm) indicating the carboxylate anion is bonded to tin atom upon complexation. Complex 1 showed a sharp signal at 12.25 ppm indicated the present of methyl groups in the SnMe$_2$ moiety. In the upfield region of $^{13}$C NMR spectra, complexes 2 and 3 showed the occurrence of CH$_3$ and CH$_2$ in the range of 13.71-14.42 and 26.09-29.90 ppm respectively (Danish et al., 1995; Holecek et al., 1986). In addition, complex 3 exhibited two sets of butyl signals in $^{13}$C NMR spectra. This attributed to the butyl groups linked to the exo- and endo-cyclic tin atom respectively. The $^{13}$C NMR spectra of complex 4 showed that the chemical shifts of the $\delta$(CH)$_{ipso}$ at 142.87 ppm indicative of a five-coordinated Sn atom (Holecek et al., 1983a; 1983b; Baul et al., 2001).

For diorganotin(IV) carboxylate complexes, the $\delta$(Sn) value for four-coordinated complexes fall in the range between +200 to -60 ppm; for five-coordinated complexes between -90 to -190 ppm and for six-coordinated complexes between -210 to -400 ppm (Holecek et al., 1986). Complexes 1 and 2 showed that the $\delta$(Sn) are -281.39 and -310.76 ppm respectively, indicated that the tin atom in complexes 1 and 2 are six-coordinated. Complex 3 showed two well separated resonances of $\delta$(Sn) at -173.87 and -213.71 ppm respectively. These two low- and high-field resonances respectively are attributed to the exo- and endo-cyclic tin atoms in complex 3 as observed in distannoxane dimer (Danish et al., 1995). As a result, complex 3 showed that the exo- and endo-cyclic tin atoms are five- and six-coordinated respectively (Danish et al., 1995; Holecek et al., 1986).
coordination number of tin in triphenyltin(IV) carboxylate could be determined by the studies of $^{119}$Sn-$^{13}$C coupling constant (Holecék et al., 1983a; Baul et al., 2001). Basically, the tin atom of triphenyltin(IV) compounds with higher $\delta$($^{119}$Sn) and $^{1}$J($^{119}$Sn-$^{13}$C) value lie in the range of -200 to -260 ppm and 750-850 Hz respectively are believed to exhibit five-coordinated and in trigonal bipyramid geometry of the substituents and ligand, Ph$_3$SnX•L and L is a monodentate ligand (Holecék et al., 1983b). The three phenyl groups lie in the equatorial positions with the substituent, X and the ligand, L lie in the axial positions to form trans-trigonal bipyramid geometry (Holecék et al., 1983b). Complex 4 showed that the $\delta$($^{119}$Sn) and $^{1}$J($^{119}$Sn-$^{13}$C) are -265.89 ppm and 839.3 Hz respectively indicated that the tin atom in complex 4 is five-coordinated and having trans-trigonal bipyramid geometry. This due to one $d_0$-DMSO molecule coordinated to the tin atom in complex 4 resulting the complex exhibited five-coordinated tin atom in solution state.

The in vitro antibacterial screening activity of parent acid and complexes 1-4 are given in Table 1. Inhibition zones with a diameter less than 10 mm are considered as weak; larger than 10 mm but less than 16 mm are considered as moderate and finally larger than 16 mm and above are active (Chohan et al., 2006). Complex 1 showed a significant result against all the tested bacterial strains even though the activities obtained were from weak to moderate activity. Meanwhile, complexes 2-4 were found to be significantly active against gram-positive bacterial strains. Against Bacillus subtilis and Staphylococcus aureus at 1.0 mg mL$^{-1}$, the inhibition zones obtained for complex 4 were 18 and 19 mm respectively indicating that the in vitro antibacterial activity was in the active mode. Moreover, the inhibition zone diameters of complexes 1-3 in the range of 9-13 mm indicated that their activities were weak. Hence, complex 4 was more active compared to diorganotin(IV) complexes derivatives. In this study, the tin atom moiety of complex 4 is five-coordinated and exists in trans-R$_{3}$SnO$_2$ geometry in solution form; hence causing it’s activity to be greater compared to complexes 1-3 (Danish et al., 1995; Baul et al., 2002). Although complex 4 showed significant in vitro antibacterial activity against gram-positive bacterial strains but the value obtained was lower compared to the reference drugs.

**CONCLUSION**

Complexes 1-4 have been successfully synthesized. The structural as well as the coordination number of tin moieties of complexes 1-4 have been successfully characterized quantitatively and qualitatively. Based on the in vitro antibacterial screening activity, complex 4 showed significant activity on Bacillus subtilis and Staphylococcus aureus compared to complexes 1-3 but lower compared to reference drugs.

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