Dibutyltin(IV) Complex of 2-[(*E*)-(4-hydroxy-3-{(*E*)-[(quinolin-2-yl)imino] methyl} phenyl) diazenyl]benzoic Acid: Synthesis, Spectroscopy and *in vitro* Antifungal Activity

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ABSTRACT:*Dibutyltin(IV) complex of 2-[(E)-(4-hydroxy-3-{(E)-[(quinolin-2-yl)imino] methyl} phenyl) diazenyl]benzoic acid was synthesized by refluxing 2-[(E)-(4-hydroxy-3-{(E)-[(quinolin-2-yl) imino] methyl} phenyl) diazenyl]benzoic acid with dibutyltin(IV) oxide in hot toluene. The complex was characterized by elemental analysis in combination with UV-Visible, IR, ^{1}H, ^{13}C, ^{119}Sn NMR spectroscopy, and Mass spectrometry technique. The carboxylate ligand acts as a chelating bidentate mode of coordination with the tin atom in the complex. The complex exhibited a cyclic dimeric structure in the solution state where the tin centers adopt 6- coordinate octahedral geometry. The in vitro antifungal property of the ligand and the complex was observed and compared with the reference drug, Amphotericin-B.*

KEYWORDS: *Carboxylate; NMR spectroscopy; Coordination geometry; Antifungal activity.*

INTRODUCTION

Organotin (IV) carboxylates have received particular interest owing to their significant structural diversity and important biological activity, such as pesticides, wood preservatives, antibacterial, antifungal, antitumor agents, and anti-diabetic properties, etc. [1-10]. Thus, the synthesis of organotin(IV) carboxylates has been a research interest. Literature also implied that the biological activities of organotin(IV) carboxylates depend greatly on their structures, which are significantly related to the carboxylic acid ligands and organic substituents bonded to the tin center [11–14]. Previously, we had reported molecular structure and topological studies of diorganotin (IV) complexes (dimethyl, dibutyl, and diphenyl-) of azo-dicarboxylic acid ligand and their anti-diabetic activity in the solid state [8]. The complexes exhibited cyclic tetra-nuclear (dimethyl and dibutyl-) and cyclic dimeric (diphenyl-) structures in In view of the interesting molecular structures and biological properties exhibited by diorganotin (IV) complexes of carboxylic acid ligands, my current interest focuses mainly on the synthesis, characterization, and *in vitro* antifungal activity of dibutyltin (IV) complex of 2-[(*E*)-(4-hydroxy-3-{(*E*)-[(quinolin-2-yl)imino] methyl} phenyl) diazenyl]benzoic acid. The complex was characterized by, UV, IR, multinuclear (^{1}H, ^{13}C, and ^{119}Sn NMR) spectroscopy and Mass spectrometry techniques. The *in vitro* antifungal study of the ligand and the complex was carried out with reference to the standard drug, Amphotericin-B, and reported in this work.

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EXPERIMENTAL SECTION

Materials and methods

Dibutyltin(IV) dioxide, *ortho*-amino benzoic acid, salicylaldehyde, and 2-aminoquinolin were obtained from MERCK and used without further purification. The solvents were dried and purified according to standard procedures [15]. Perkin Elmer 2400 series II instrument was used for the analysis of Carbon, hydrogen, and nitrogen. UV-Visible spectra of the ligand and the complex were carried out on a UV-1800 Shimadzu spectrophotometer in DMF solution from 200 to 800 nm range. IR spectra were obtained from Shimadzu FT-IR-8400S spectrophotometer from 4000 to 400 cm^{-1} range using KBr disks. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker AMX 400 spectrometer measuring at 400.13 and 100.62 MHz frequency, respectively while for ^{119}Sn NMR spectra, Jeol JNM-ECZ600R/S1 spectrometer operating at 600.17 MHz frequency was used. For ^{1}H and ^{13}C NMR spectroscopy, tetramethylsilane was used as a reference while tetramethyltin was employed as a reference for ^{119}Sn NMR spectroscopy.

Preparation of ligand precursor, {2-[(E)-(3-formyl-4-hydroxyphenyl) diazenyl] benzoic acid}

The ligand precursor was prepared by a diazo coupling reaction of *ortho*-amino benzoic acid (5 g, 36.45 mmol) and salicylaldehyde (4.45 g, 36.45 mmol) in an alkaline medium at 0-5°C [16]. It was recrystallized from toluene to obtain a pure product. Yield: 55%; m.p.: 177–179°C. Anal. Calcd for $C_{14}H_{10}N_2O_4$: C, 62.22; H, 3.70; N, 10.37%. Found: C, 61.85; H, 3.74; N, 10.35%.

Preparation of {2-[(E)-(4-hydroxy-3-{(E)-[(quinolin-2-yl)imino] methyl} phenyl) diazenyl]benzoic acid} (H_2L)

0.5g (1.85 mmol) ligand precursor was dissolved in hot toluene (50 mL). An alcoholic solution of 2-aminoquinoline (0.26g, 1.85 mmol) was added dropwise to hot toluene with constant stirring. The reaction mixture was refluxed for 3 hours and then filtered. The red solid product was dried and recrystallized with anhydrous methanol. Yield: 58%, m.p.:158-162°C. Anal. Calcd. for $C_{23}H_{16}N_4O_3$: C, 69.69; H, 4.07; N, 14.13%. Found: C, 69.97; H, 4.01; N, 14.19%. UV-Visible (DMF) λ_{max}(nm): 256, 370. IR (cm^{-1}): 1670 ν_{asy} (COO). ^{1}H-NMR(DMSO-d_6, 400 MH_Z) δ_H: 10.13 [CH=N], (7.90-7.91) [d, 1H, (A) H3, *J*=2.4 Hz], (7.82-7.85) [m, 2H, (A) H4 and H5], (7.54-7.52) [d, 1H, (A) H6, *J*= 7.2 Hz], (7.44-7.46) [d, 1H, (B) H6, *J*= 8 Hz], (7.37-7.41) [t, 1H, (C) H3, *J*=7.2 Hz], (7.26-7.34) [m, 4H, (D) H5, H6, H7 and H8], (6.93-7.00) [m, 2H, (B) H3 and H2], 6.60- 6.62 [d,1H, (C) H2, *J*=8.8 Hz] ppm. ^{13}C NMR ($CDCl_3$, 100 MHz) δ_C, Ligand skeleton: 190.27 [CO_2], 164.52 [(B) C-4], 157.33 [C=N], 150.42 [(B) C-1], 144.38 [(A) C-1], 131.23 [(B) C-3], 129.79 [(B) C-6], 129.55 [(B) C-4], 129.00 [(B) C-5], 127.73 [(A) C-2], 124.23 [(A) C-6], 122.65 [(D) C-6], 121.96 [(D) C-5], 118.80 [(B) C-2], 117.99 [(A) C-3], 112.79 [(A) C-5] ppm.

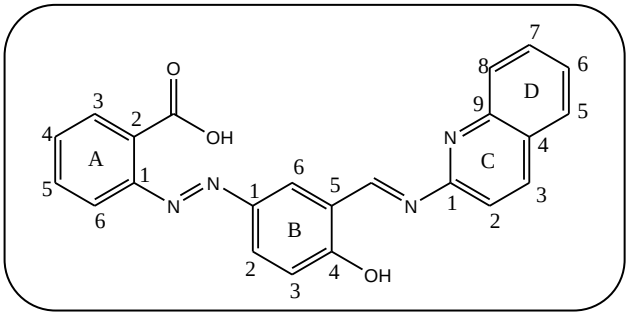
Synthesis of dibutyltin(IV) complex, $[Bu_2SnL]_2$

0.5g (1.263 mmol) ligand (H_2L) was suspended in hot toluene (50 mL). To this hot suspension, dibutyltin(IV) oxide (0.314g, 1.263 mmol) was added as a solid with stirring. The reaction mixture was refluxed for 5 hours. The solution was then filtered in the hot condition and the filtrate was dried using a rotary evaporator. The solid mass obtained was recrystallized from chloroform when a red crystalline compound was obtained. Yield: 42%, m.p.:98-102°C Anal. Calcd. for $C_{69}H_{86}N_8O_6Sn_2$: C, 60.90; H, 6.37; N, 8.23%. Found: C, 60.71; H, 6.31; N, 8.16 %. UV-visible (DMF) λ_{max}(nm): 258, 376, 460. IR (KBr, cm^{-1}): 1613 ν_{asy}(COO), 460 ν(Sn-O), 600 ν(Sn-C). ^{1}H-NMR($CDCl_3$, 400 MH_Z) δ_H: 9.80 [CH=N], (8.04-8.08) [m, 2H, (A) H3 and H4], (7.88-7.89) [d, 1H, (A) H5, *J*=5.2 Hz], (7.79-7.81) [d, 1H, (A) H6, *J*=8.8 Hz], (7.65-7.67) [d, 1H, (B) H6, *J*=8 Hz], (7.41-7.55) [m, 5H, (C) H3, (D) H5, H6, H7 and H8], (7.23-7.27) [m, 2H, (B) H3 and H2], (6.70-6.73) [d, 1H, (C) H2, *J*=8.8 Hz]; tin-butyl skeleton: 1.78 [brm, 8H, H-α and H-β], 1.25 [brm, 4H, H- γ], 0.84 [brm, 6H, H-δ] ppm. ^{13}C NMR ($CDCl_3$, 100 MHz) δ_C, Ligand skeleton: 196.32 [CO_2], 170.235 [(B) C-4], 155.95 [C=N], 149.520 [(B) C-1], 141.20 [(A) C-1], 140.38 [(C) C-3], 131.19 [(B) C-3], 130.12 [(B) C-6], 127.75 [(B) C-4], 127.63 [(B) C-5], 123.83 [(A) C-2], 121.87 [(A) C-6], 121.23 [(D) C-6], 117.27 [(A) C-3], 113.00 [(A) C-5]; tin-butyl skeleton: 31.00, 27.29, 26.87, 26.45, 26.06, 13.61 ppm.^{119}Sn NMR ($CDCl_3$, 600 MHz): -285, -466 ppm. EI-MS: MW calculated for $C_{69}H_{86}N_8O_6Sn_2$, (1360); found: 1357.25; (m/z): 1357.25 $[C_{69}H_{86}N_8O_6Sn_2]^+$; other prominent peaks: 923.74, 907.77, 881.75, 523.24, 507.27, 360.32, 182.19, 145.07.

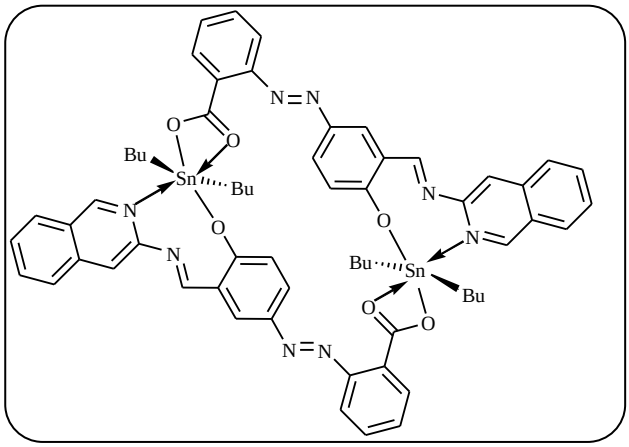
Antifungal assay

The antifungal activities of the ligand and the compound were carried out by the agar well diffusion method

using 20 mL of sterile nutrient agar (Hi-Media) and potato-dextrose agar (Hi-Media) and sabouraud dextrose agar (Hi-Media) for testing the antifungal activity [17]. The test cultures were swabbed on top of the solidified media and allowed to dry for 10 minutes. Sterile 6 mm diameter cork borers were pierced in the agar. The ligand and the compound were diluted in 3 mg/mL in DMSO and diluted again. The dilutions of the concentration of the compound were deposited 20µL on the inoculated well and left for 10 minutes at room temperature for diffusion. DMSO was used for negative control while Amphotericin-B served as a positive control. The plates were inoculated with fungi at 30ºC for 48 hours. The experiment was repeated twice and the diameter of the inhibition zone (mm) around the well was measured. The microbial susceptibility was determined by the minimum inhibitory concentration detection method [18]. The minimum inhibitory concentration (MIC) of the ligand and the compound were determined by serial dilution against the micro-organisms. The minimum concentration is defined as the concentration at which no visible growth of microorganisms was observed and is expressed in µg/mL. Four fungal species viz. *A. flavus, A. fumigates, A.niger, and C. albicans* were employed for the test.



Scheme 1: {2-[(E)-(4-hydroxy-3-{(E)-[(quinolin-2-yl)imino] methyl} phenyl) diazenyl]benzoic acid} (H_2L).



Scheme 2: Synthesis of dibutyltin(IV) complex, $[Bu_2SnL]_2$.

RESULT AND DISCUSSION

Synthesis

2-[(*E*)-(4-hydroxy-3-{(*E*)-[(quinolin-2-yl)imino] methyl} phenyl) diazenyl]benzoic acid (H_2L) was prepared by the condensation of 2-[(*E*)-(3-formyl-4-hydroxyphenyl) diazenyl] benzoic acid with 2-aminoquinoline in hot toluene. Dibutyltin(IV) complex of 2-[(*E*)-(4-hydroxy-3-{(*E*)-[(quinolin-2-yl)imino] methyl} phenyl) diazenyl]benzoic acid was synthesized following 1:1 metal-ligand ratio. Physical properties and analytical data were included in the experimental section whereas the schematic diagram of the ligand and the complex were represented by scheme 1 and scheme 2.

UV-visible spectroscopy

The UV-visible spectra of the ligand and the compound were recorded in DMF solution (10^{-5} M) at room temperature shown in "Fig. 1". The electronic spectrum of the ligand showed an absorption band at 256 and 370 nm which may be due to π-π* transition of the aromatic ring and n-π* transition of imine (C=N) respectively [19]. UV-Visible spectra of the compound exhibited three absorptions at 258, 376, and 460 nm, respectively. After coordination, π-π* transition of the aromatic ring and n-π* transition of mine were shifted slightly to a higher wavelength in the complex [20]. The new band observed at 460 nm may be assigned to ligand-to-metal charge transfer [20]. The shifting of λ_{max} value from the ligand to the dibutyltin (IV) complex is a clear indication of coordination between the ligand and the tin atom.

IR spectroscopy

IR spectroscopic data for the ligand and the complexwere mentioned in the Experimental section. IR absorption bands for the carboxylate vibrations ν_{asy}(COO) of the ligand were observed at 1670 cm^{-1}. But in the complex, this band shifted to 1613 cm^{-1} which indicates the carboxylate coordination to the tin [21]. The mode of coordination can be predicted by the difference between the asymmetric and symmetric frequencies of the carboxylate group. The $\Delta\nu(\nu_{asy}COO\text{-}\nu_{sym}COO)$ less than 200 cm^{-1} indicates bidentate bridging or bidentate chelating coordination mood and for monodentate mode, the value is more than

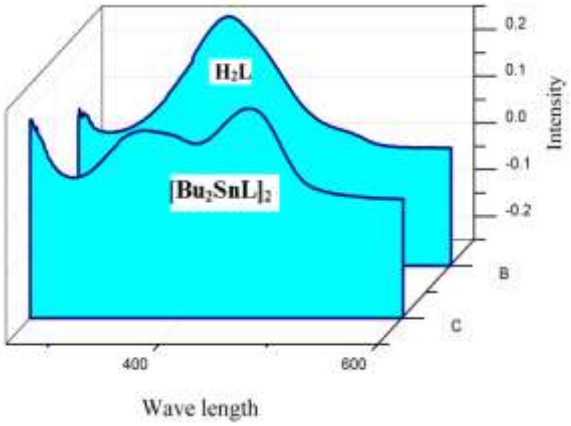


Fig. 1: UV-Visible spectra of H_2L and $[Bu_2SnL]_2$.

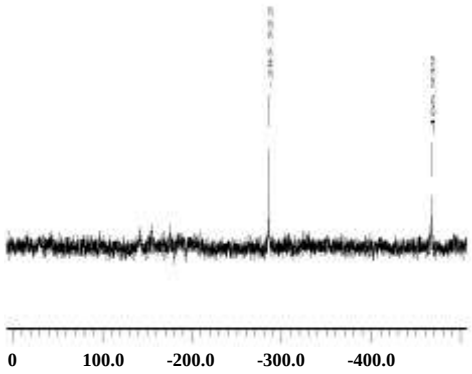


Fig. 2: ^{119}Sn -NMR spectrum of $[Bu_2SnL]_2$.

200 cm^{-1} [22]. In the complex, the value is less than 200 cm^{-1}, suggesting bidentate bridging or bidentate chelating carboxylate ligand [16]. IR absorption bands observed at 460 cm^{-1} and 600 cm^{-1} in the complex may be assigned to Sn-O and Sn-C stretching frequencies, respectively [6,10].

Multinuclear spectroscopy

The ^{1}H NMR spectrum of the ligand and the complex were carried out in DMSO-D_6 and $CDCl_3$ respectively. The signals for aromatic protons of the ligand were observed at 6.60-7.91 ppm while in the complex these peaks were observed at 6.70-8.08 ppm which is due to the attachment of the ligand to the tin atom. Signals for -COOH and –OH protons were not observed due to the solvent exchange reaction. Signals for tin-butyl protons of the complex appears at 0.84-1.78 ppm. In the ^{13}C NMR spectrum, the chemical shift for -COO carbon was observed at 190.27 ppm but after coordination, there was an increase in chemical shift value to 196.32 ppm which indicates that the coordination occurs through the carboxylic oxygen atoms [10, 23]. Also, the chemical shift value for the phenolic oxygen atom in the complex is higher as compared to that of the ligand which indicates that the phenolic oxygen atom also participates in the coordination with the tin atom [16]. Signals for tin-butyl carbons appear at 13.61-31.00 ppm in the complex indicating the presence of tin-butyl moiety in the complex. The chemical shift values of the ligand and the complex are fully consistent with the formulation of the compound. The coordination geometry around tin atoms in the complex was confirmed by analyzing ^{119}Sn NMR spectrum of the complex. Two resonance peaks were observed in the ^{119}Sn NMR spectrum of the dibutyltin(IV) complex at -285 and -466 ppm (shown in Fig. 2) confirming the presence of two tin centers in the complex [8, 24]. Moreover, the chemical shift value (-285 to -466 ppm) falls within the specified range of 6-coordinate octahedral geometry in the solution state [8, 24].

Mass spectrometry

The mass spectrometry technique was used to predict the geometry of the complex. The Electron Ionization (EI) technique was employed to determine the mass spectra of the complex. The molecular ion $[M]^+$ peak of the complex appears at (m/z) 1357.25. The molecular mass of the complex obtained from the mass spectrum confirmed the proposed 6-coordinate octahedral geometry of the complex.

Antifungal activity

The antifungal activities of the ligand and the dibutyltin(IV) complex were studied at (3.0 mg/mL) concentration and compared with the standard drug, Amphotericin-B. The minimum inhibitory concentration (MIC) of the ligand and the complex were also determined. The zone of inhibition of H_2L and the complex against various fungi are given in Table 1 along with the standard drug, Amphotericin-B while their antifungal activity has been presented in Fig. 3.

In vitro agar well-diffusion method, concentrations of compounds: 3 mg/mL in DMSO, reference drug, Amphotericin-B for antifungal test: 16 µg/mL in DMSO.

Table 1: Zone of inhibition (mm) of tested fungal species.

Compounds	*A. flavus*	*A. fumigatus*	*A. niger*	*C. albicans*
H_2L	06	08	08	10
$[Bu_2SnL]_2$	26	30	32	34
Amphotericin-B	30	32	36	40

Table 2: Minimum inhibition concentration (MIC) for tested fungi (µg/mL)

Sl. No.	Compounds	A. flavus	A. fumigatus	A. niger	C. albicans
1	H2L	195.25	225.75	200	219
2	[Bu2SnL]2	6.0	12.0	8.0	11
3	Amphotericin-B	0.5	1.0	0.5	1.0

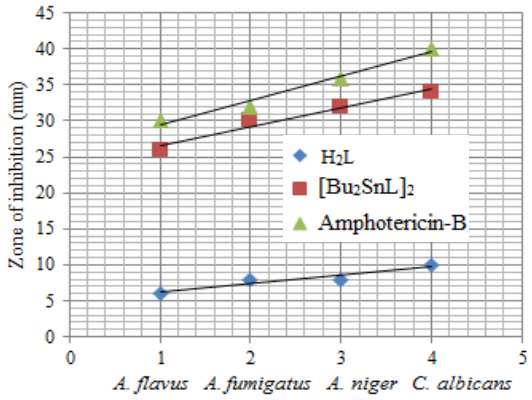


Fig. 3: Comparison of antifungal activity of the compounds with the standard drug.

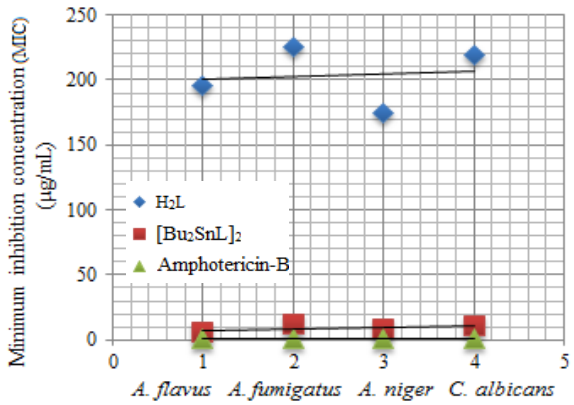


Fig. 4: Minimum inhibition concentrations (MICs) of the compounds and the standard drug.

It has been observed that the complex is an effective antifungal agent against all the screened fungal strains. The antifungal activity of the dibutyltin(IV) complex may be assigned due to the ability of the complex to inhibit various cellular enzymes which play important roles in different metabolic pathways for the microorganisms [25]. The minimum inhibitory concentrations of H_2L and the complex was determined against various fungi and compared with the standard reference drug. The MIC values are listed in Table 2 and are presented in Fig. 4.

In vitro agar well-diffusion method, concentrations: µg mL^{-1} in DMSO, reference drug, Amphotericin-B for the antifungal test.

From the MIC values, it can be concluded that the dibutyltin(IV) complex is a more effective antifungal agent than the ligand (H_2L) and is comparable to that of the standard drug, Amphotericin-B.

CONCLUSIONS

Dibutyltin(IV) complex of 2-[(*E*)-(4-hydroxy-3-{(*E*)-[(quinolin-2-yl)imino] methyl} phenyl) diazenyl]benzoic acid was synthesized by reacting 2-[(*E*)-(4-hydroxy-3-{(*E*)-[(quinolin-2-yl) imino] methyl} phenyl) diazenyl]benzoic acid with dibutyltin(IV) oxide using standard procedure. The complex was characterized by elemental analysis, UV, IR, ^{1}H, ^{13}C, ^{119}Sn NMR spectroscopy, and Mass spectrometry technique. IR spectra of the complexes indicated that the carboxylate oxygen atoms act as a chelating bi-dentate mode of coordination. ^{119}Sn NMR spectra of the complex confirmed the presence of two tin centers that adopt 6-coordinate octahedral geometry in solution state. EI-MS spectra of the complex are also in good agreement with the formulation of the cyclic dimeric structure of the complex. The antifungal activity of the complex is

higher than that of the ligand but comparable to that of the standard drug.

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