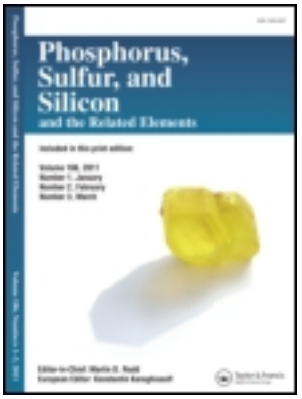

Phosphorus, Sulfur, and Silicon and the Related Elements

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Synthetic, Spectroscopic, and Biological Aspects of Triorganosilicon(IV) Complexes of Tridentate Schiff Bases

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SYNTHETIC, SPECTROSCOPIC, AND BIOLOGICAL ASPECTS OF TRIORGANOSILICON(IV) COMPLEXES OF TRIDENTATE SCHIFF BASES

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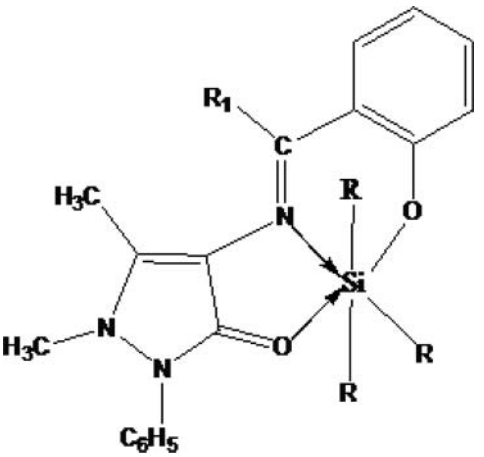
GRAPHICAL ABSTRACT



Abstract *Hexacoordinated organosilicon complexes of type $R_3Si(L)$ (R = ethyl, butyl, phenyl; HL = ligand, obtained by the condensation of 4-aminoantipyrine with 2-hydroxyacetophenone, 2-hydroxybenzophenone, 2-hydroxybenzaldehyde, and 2-hydroxynapthaldehyde) have been synthesized and characterized by elemental analysis, molar conductance, and spectroscopic studies (IR, ^1H, ^{13}C, and ^{29}Si NMR). The spectroscopic studies indicated that the ligands acted as tridentate coordinating through azomethine nitrogen, carbonyl oxygen, and oxygen of hydroxyl after deprotonation to the central silicon atom. The ligands and their organosilicon complexes have been evaluated for antimicrobial activities against fungi (Aspergillus niger and Candida albicans), and against Gram-positive bacteria (Staphylococcus aureus, Bacillus subtilis), and Gram-negative bacteria (Escherichia coli). The aim of the present work is to synthesize novel eco-friendly fungicides and bactericides and to study the effect of the biological activity of ligands on the complexation with organosilicon moiety.*

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Keywords Organosilicon; spectroscopic studies; 4-aminoantipyrine

INTRODUCTION

Tridentate and tetradentate Schiff bases ligands with donor centers like N, O, S, etc. showing broad biological activities are of particular interest not only for existing in the keto or enol form but also to form complexes with unusual coordination numbers. It is, however, noteworthy that the biological activity of Schiff bases was appreciably enhanced on complexation with organosilicon halide,[1–4] where the coordination number of silicon increases and the tetrahedral geometry changes to trigonal bipyramidal or octahedral geometries. In general, the biochemical activity of organosilicon compounds is greatly influenced by stereochemistry of the compound and the coordination number around silicon atom. Moreover, organosilicon compounds have wide range of biological activities including anticarcinogenic, antibacterial, antifungal, tuberculostatic, insecticidal, and acaricidal activities and also used in pharmaceutical and chemical industries for the synthesis of organic materials.[5–9]

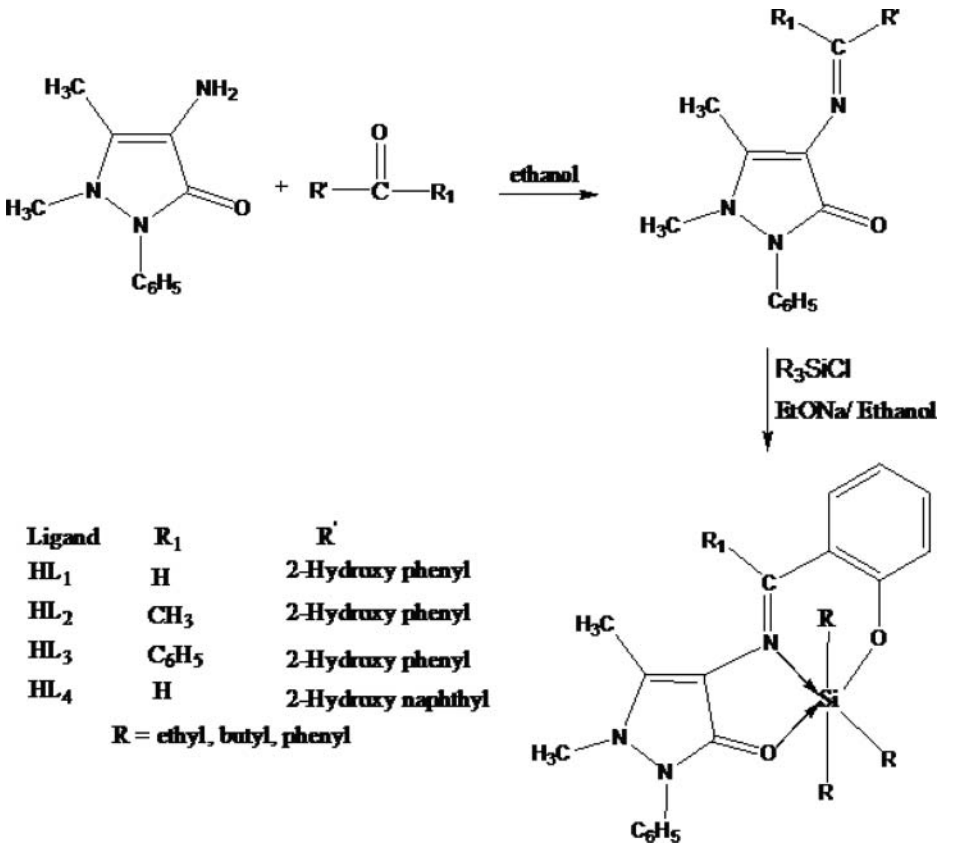
In continuation of our earlier research on the isolation of antimicrobial compounds,[10–13] an attempt has been made to synthesize complexes of Schiff base derived from 4-aminoantipyrine and aromatic aldehydes with triorganosilane and for studying the effect of complexation on the biocidal activity to explore the possibility of their use as potential biocides.

RESULTS AND DISCUSSION

Organosilicon (IV) complexes have been synthesized by refluxing chlorotriorganosilanes with sodium salts of Schiff base ligands in 1:1 molar ratio in dry ethanol (see Scheme 1). The isolated complexes were insoluble in most of the organic solvent except in $CDCl_3$, dimethyl sulfoxide (DMSO), and dimethylformamide (DMF). The molar conductance value of 10^{-3} M solution of these complexes was in the range of 8–15 Ω^{-1} cm^2 mol^{-1} in DMSO, indicating nonelectrolytic nature of complexes. The geometry around silicon atom in these complexes has been determined using various techniques Fourier transform infrared (FT-IR) and NMR (^{1}H, ^{13}C, and ^{29}Si) spectroscopic data.

IR Spectra

The infrared spectra of the compounds were recorded using KBr pellets in order to find the coordination sites of ligand on complexation with chlorotriorganosilane on the basis of shifting in the frequency of various groups and/or from the lowering in the intensities of the absorptions. The broad band centered at 3350 cm^{-1} in spectra of the free ligands are due to the hydroxyl group ν(OH). The broadness of the band suggested the presence of hydrogen bonding between the azomethine nitrogen and the OH group. This band was completely absent in the spectra of the complexes indicating the coordination of the ligand to the silicon atom through the hydroxyl oxygen. A sharp high-intensity band appeared at 1645 $\pm$ 10 cm^{-1}. It was due to the azomethine group ν(C=N) in ligands and was shifted to a lower frequency as compared to the free ligand by 10–15 cm^{-1} which



Scheme 1

indicated the donation of the lone pair of the azomethine nitrogen and inferred that the ligands coordinate to the silicon atom through the azomethine nitrogen. In the pyrine ring of free ligands, sharp bands appeared at 1680 cm^{-1} due to the ν(C═O). It was shifted by 20–30 cm^{-1} toward lower frequencies on complexation with the silicon atom indicating the participation of the carbonyl oxygen atom in coordination. The formation of the resulting complexes were further supported by appearance of new bands at 624 ± 10 cm^{-1} and 410 ± 15 cm^{-1} due to ν(SiO) and ν(SiN). The appearance of medium to strong intensity bands around 1470–1410 cm^{-1}, 1150–1075 cm^{-1}, 850–785 cm^{-1}, and 670–630 cm^{-1} have been assigned to aromatic ring.

NMR Spectra

The ^{1}H, ^{13}C, and ^{29}Si NMR spectra of the ligands and their corresponding organosilicon(IV) complexes were recorded in $CDCl_3$ using tetramethylsilane (TMS) as the internal standard. The chemical shift (ppm) and coupling constant (Hz) value are given in Table 1. The ^{1}H NMR spectra of the free ligand showed a singlet at 15.2 ppm due to an OH proton which completely disappeared in the complexes indicating the coordination of the hydroxyl oxygen to the silicon atom on complexation after deprotonation of this group. The sharp signal at 10.8 ppm in the ligand due to the azomethine proton was shifted downfield in the

Table 1 ^{1}H NMR spectroscopic chemical shifts δ and coupling constants J (Hz) of ligands and organosilicon(IV) complexes

Compound	OH	CH=N	H-aromatic	C-CH_3, N-CH_3	Si-R
HL_1	12.98	9.71	7.57–7.46(m, 3H), 7.42–7.38(m, 2H), 7.35(d, J = 8.2, 1H), 7.25(d, J = 8.1, 1H), 6.99(dd, J = 7.8, 2H)	2.40, 3.20	–
HL_2	12.41	–	7.78–7.76(m, 3H), 7.74–7.52(m, 2H), 7.39(d, J = 7.7, 1H), 7.21(d, J = 7.1, 1H), 7.11(dd, J = 9.6, 2H)	2.41, 3.22	–
HL_3	12.98	–	7.45–7.42(m, 3H), 7.35–7.29(m, 2H), 7.28(d, J = 12.0, 1H), 7.19(d, J = 8.1, 1H), 7.02(dd, J = 7.5, 2H)	2.40, 3.20	–
HL_4	15.22	10.83	8.26(d, J = 1H), 7.78–7.70(m, 2H), 7.51–7.43(m, 3H),7.17(d, J = 1H), 7.40–7.25(m, 4H)	2.38, 3.11	–
$Et_3Si(L_1)$	–	11.54	7.97–7.96(m, 3H), 7.89–7.72(m, 2H), 7.47(d, J = 7.9, 1H), 7.34(d, J = 8.2, 1H), 7.01(dd, J = 7.6, 2H)	2.43, 3.21	0.86(t, 9H), 1.14(q, 6H)
$Bu_3Si(L_1)$	–	12.18	7.99–7.94(m, 3H), 7.93–7.79(m, 2H), 7.54(d, J = 8.4, 1H), 7.33(d, J = 7.9, 1H), 7.24(dd, J = 8.2, 2H)	2.41, 3.22	0.85(t, 9H),1.07–0.93 (m, 18H)
$Ph_3Si(L_1)$	–	11.69	8.17–8.10(m, 3H), 7.92–7.77(m, 2H), 7.64(d, J = 8.9, 1H), 7.51(d, J = 8.8, 1H), 7.30(dd, J = 8.1, 2H)	2.45, 3.25	7.74–7.66(m, 15H)
$Et_3Si(L_2)$	–	–	7.98–7.93(m, 3H), 7.81–7.66(m, 2H), 7.59(d, J = 12.3, 1H), 7.33(d, J = 8.4, 1H), 7.12(dd, J = 8.8, 2H)	2.42, 3.21	0.80(t, 9H),1.10 (q, 6H)
$Bu_3Si(L_2)$	–	–	7.89–7.86(m, 3H), 7.84–7.65(m, 2H), 7.51(d, J = 9.9, 1H), 7.42(d, J = 10.2, 1H), 7.21(dd, J = 10.8, 2H)	2.43, 3.20	0.82(t, 9H),1.08–1.22 (m, 18H)
$Ph_3Si(L_2)$	–	–	8.08–8.06(m, 3H), 7.94–7.71(m, 2H), 7.69(d, J = 11.7, 1 H), 7.58(d, J = 8.4. 1H), 7.38(dd, J = 9.9, 2H)	2.40, 3.21	7.65–7.57(m, 15H)
$Et_3Si(L_3)$	–	–	7.66–7.63(m, 3H), 7.57–7.38(m, 2H), 7.35(d, J = 8.8, 1H), 7.21(d, J = 8.5, 1H), 7.09(dd, J = 8.3, 2H)	2.46, 3.19	0.85(t, 9H),1.15 (q, 6H)
$Bu_3Si(L_3)$	–	–	7.89–7.82(m, 3H), 7.79–7.58(m, 2H), 7.49(d, J = 7.5, 1H), 7.55(d, J = 7.9, 1H), 7.37(dd, J = 8.9, 2H)	2.45, 3.21	0.80(t, 9H),0.97–1.23 (m, 18H)
$Ph_3Si(L_3)$	–	–	8.05–8.02(m, 3H), 7.95–7.74(m, 4H), 7.48(d, J = 8.6, 1H), 7.36(d, J = 7.6, 1H), 7.21(dd, J = 6.9, 2H)	2.43, 3.20	7.55–7.49(m, 15H)
$Et_3Si(L_4)$	–	–	8.33(d, J = 11.1, 1H), 7.86–7.65(m, 2H), 7.63–7.56(m, 3H), 7.19(d, J = 11.1, 1H), 7.37–7.22(m, 4H)	2.45, 3.19	0.81(t, 9H),1.12 (q, 6H)
$Bu_3Si(L_4)$	–	–	7.81(d, J = 8.4, 1H), 7.76–7.67(m, 2H), 7.76–7.54 (m, 3H), 7.17(d, J = 8.4, 1H), 7.42–7.33(m, 4H)	2.45, 3.10	0.96(t, 9H),1.32–1.19 (m, 18H)
$Ph_3Si(L_4)$	–	–	7.63(d, J = 11.4, 1H), 7.46–7.36(m, 5H),6.96–6.94(m, 4H), 6.82 (d, J = 11.4, 1H)	2.47, 3.08	7.34–7.23(m, 15H)

spectra of complexes indicating the involvement of nitrogen of azomethine group in coordination. Aromatic protons of ligands appeared as multiplets in the range 8.26–6.74 ppm. The singlets at 2.38 and 3.11 ppm due to the methyl protons of C-CH_3 and N-CH_3 groups, respectively, remain unchanged on complexation. Furthermore, the formation of complexes was supported by appearance of new signals at 0.83–1.55, 0.80–1.97, and 7.34–7.23 ppm in the spectra of the complexes due to the ethyl, butyl, and phenyl proton attached to the silicon atom. Integrated proton ratios were well in agreement with the proposed structure.

The ^{13}C NMR spectra of the ligands and their corresponding organosilicon (IV) complexes were recorded in $CDCl_3$ with few drops of DMSO-d_6 and the chemical shift values are given in Table 2. In the ^{13}C NMR spectra of the ligands, signal appeared due to the azomethine carbon at 161.9 ppm was shifted downfield by 8–15 ppm on complexation showed the involvement of this group in the coordination. The signal at 160.3 ppm due to carbonyl carbon was shifted downfield by 5–10 ppm, confirming the involvement of carbonyl oxygen in coordination with silicon atom. Signal due to carbon of ethyl group attached to silicon appeared at 9.2–9.5 ppm whereas signal of carbon due to butyl group attached to silicon atom appeared at 12.5–12.9, 13.3–14.0, 21.1–21.5, and 25.9–26.3 ppm, respectively, while the carbon atom of the phenyl group attached to the silicon moiety appeared at 146.9–136.9 ppm. Signals due to aromatic carbon atoms of the ligands appeared in the range 156.9–109.1 ppm.

The ^{29}Si NMR spectra reflect the coordination number of the nucleus in the complexes. In order to confirm the geometry of the complexes, ^{29}Si NMR spectra were recorded. The spectra showed one sharp singlet in each complex indicating the formation of a single species. The value of chemical shift in ^{29}Si NMR spectra depends upon the nature of R group attached to silicon atom[12] which is higher for alkyl group than aryl group because of if R is phenyl, shielding of the ^{29}Si nucleus is observed because the delocalized system of phenyl group which allows for $d\pi$-$p\pi$ interactions; thus the chemical shift is lowered by 12–15 ppm in these complexes as compared to those which have greater σ donation capacity, i.e., when R = alkyl group. This is well in agreement with the present isolated organosilicon complexes, as the sharp singlet at -95 to -99 ppm was observed if R = ethyl or butyl and -102 to -110 ppm if R = phenyl.

Antimicrobial Activity

The ligands and their corresponding complexes have been evaluated for in vitro antibacterial activity against Gram-positive bacteria *Bacillus subtilis* and *Staphylococcus aureus* and Gram-negative bacteria *Escherichia coli* and in vitro antifungal activity against *Candida albicans* and *Aspergillus niger* fungi. The conventional bactericides tetracycline, chloramphenicol, kanamycin, cefazoline sodium, and cefotaxime and fungicides cycloheximide, carbendazim, and fluconazole were used as standards for comparing the activity of compounds. Minimum inhibitory concentrations (MICs) were determined by means of twofold serial dilution technique which is given in Table S1 (online Supplementary Materials). Organosilicon (IV) complexes exhibited better antibacterial and antifungal properties than those of the corresponding ligands. The complexes $Ph_3Si(L_1)$ and $Ph_3Si(L_3)$ were most active antibacterial having MIC value of 3.12 μg mL^{-1} against *E. coli* and complexes $Ph_3Si(L_2)$ and $Ph_3Si(L_4)$ having MIC value of 3.12 μg mL^{-1} against fungi *A. niger* while complexes $Bu_3Si(L_1)$, $Bu_3Si(L_2)$, $Bu_3Si(L_3)$, and $Bu_3Si(L_4)$ were more potent against *E. coli* and *A. niger* having MIC value in range of 6.25–25 μg mL^{-1}. It may be concluded that Si-Ph complexes are more potent than Si-Bu complexes. This is in agreement with our earlier results,[8] that if a bulkier substituent R is attached to the silicon atom the lipophilicity

Table 2 ^{13}C NMR Spectroscopic chemical shifts δ of ligands and their organosilicon(IV) complexes

Compound	C=N	C=O	N-CH_3,C-CH_3	C-pyrine	C-Aromatic	Si-R
HL_1	161.3	158.9	35.8, 10.3	156.1, 148.1	134.1, 133.4, 131.9, 128.9, 124.5, 123.7, 120.4, 119.8, 116.7, 110.3	–
HL_2	160.9	159.1	35.1, 10.4	156.1, 147.2	134.6, 133.9, 133.0,128.5,124.4, 123.5, 120.7, 119.7, 116.5, 110.7	–
HL_3	162.3	160.9	35.0, 10.8	155.9, 148.0	134.7, 133.9, 132.5, 128.2, 125.1, 123.4, 120.6, 119.6, 116.4, 110.5	–
HL_4	161.9	160.3	35.4, 10.1	156.5, 148.9	134.1, 133.5, 132.6, 129.2, 128.7, 127.7, 127.4, 127.2, 124.4, 123.2, 120.3, 119.3, 116.1, 110.4	–
$Et_3Si(L_1)$	170.8	165.8	35.2, 10.6	155.6, 148.0	134.7, 133.0, 132.3, 127.9, 124.9, 123.9, 121.7, 119.1, 116.9, 110.1	20.7, 9.7
$Bu_3Si(L_1)$	170.3	166.2	35.1, 10.5	155.9, 148.7	133.1, 132.4, 131.9, 128.9, 123.9, 122.8. 120.1, 118.2, 117.1, 109.9	25.0, 20.5, 12.3, 11.4
$Ph_3Si(L_1)$	170.5	165.9	35.2, 10.4	156.2, 149.1	133.8, 132.9, 131.0, 128.0, 123.7, 122.0, 120.7, 118.9, 117.5, 109.1	146.0, 141.8, 137.5, 135.6
$Et_3Si(L_2)$	175.0	166.1	35.1, 10.3	155.1, 148.9	134.1, 132.0, 131.8, 128.9, 123.9, 122.7, 120.9, 119.1, 116. 1,110.2	21.8, 9.2
$Bu_3Si(L_2)$	175.1	165.7	35.4, 10.8	156.6, 147.8	133.9, 132.9, 131.1, 127.7, 123.0. 122.7, 121.8, 120.1, 116.8, 110.7	26.5, 21.4, 13.7, 11.9
$Ph_3Si(L_2)$	175.5	165.9	35.2, 11.0	156.9, 148.9	134.9, 133.4, 132.9, 130.5, 127.9, 124.7, 121.1, 120.9, 116.2, 110.7	145.5, 140.2, 136.5, 135.9
$Et_3Si(L_3)$	173.5	166.7	35.4, 11.5	156.7, 148.9	134.8, 133.9, 132.1, 127.7, 124.9, 123.0, 120.1, 118.9, 115.1, 111.8	21.5, 10.2
$Bu_3Si(L_3)$	173.9	166.1	35.0, 10.9	155.9, 147.8	134.0, 133.9, 132.6, 128.9, 123.7 121.4, 120.0, 119.1, 115.9, 111.4	26.7, 21.8, 14.5, 12.1
$Ph_3Si(L_3)$	173.1	166.8	35.8, 10.6	157.1, 149.1	134.9, 133.5, 132.0, 129.8, 125.7, 123.2, 120.8, 118.6, 117.3, 112.1	147.6, 142.1, 136.1, 135.0
$Et_3Si(L_4)$	172.0	167.6	35.3, 10.8	157.0, 148.1	134.9, 133.0, 132.1, 129.7, 128.9, 127.9, 127.5, 127.0, 124.4, 123.9, 121.8, 120.4, 117.2, 111.3	9.8, 20.1
$Bu_3Si(L_4)$	171.9	166.9	35.7, 10.5	156.9, 149.7	135.8, 134.1, 132.0, 129.8, 128.0, 127.8, 127.5, 127.2, 125.0, 123.9, 121.7, 119.9, 117.2, 112.1	23.3, 22.1, 14.9, 12.4
$Ph_3Si(L_4)$	176.5	165.3	35.6, 11.1	157.6, 149.3	138.4, 135.5, 135.0, 134.0, 131.3, 129.5, 128.9, 128.3, 127.8, 127.5, 126.9, 125.9, 120.9, 119.7	148.6, 142.3, 137.9, 135.1

and, hence, the bioactivity is increased. A marked enhancement of in vitro biocidal studies of ligands was exhibited in coordination with silicon atom against all microorganisms strains under tested identical experimental conditions. The increase in antimicrobial activity may be explained on the basis of fact that on chelation, the polarity of silicon atom is reduced due to overlap of ligand orbital and sharing of positive charge of silicon atom with donor groups. Further, it increases delocalization of chelate ring and increases the lipophilicity of complexes.[14] This increased lipophilicity enhances penetration of complexes there by disturbing the respiration process of cell and blocking the synthesis of proteins, which further restricts growth of organisms. The conventional fungicides and bactericides showed the inhibition at concentration less than 3.12 ppm.

EXPERIMENTAL

Analytical grade chlorotriorganosilane, 4-aminoantipyrine, 2-hydroxybenzaldehyde, 2-hydroxyacetophenone, 2-hydroxybenzophenone, and 2-hydroxynaphthaldehyde purchased from Aldrich were used as such. All the experimental work was carried out under the dry nitrogen using a vacuum line as chlorotriorganosilanes and their complexes were highly moisture sensitive. Solvents were purified according to the standard procedures. All the ligands and organosilicon (IV) complexes were analyzed for C, H, and N on Perkin Elmer 2400 elemental analyzer (Waltham, Massachusetts) while silicon was estimated gravimetrically as silicon dioxide (SiO_2). Molar conductance were measured with a conductivity bridge type Model-306 Systronics (Mumbai, India) in DMSO as solvent. IR spectra were recorded using Spectrum BX Series FT-IR spectrophotometer (Perkin Elmer, New Jersey) in range 400–4000 cm^{-1} as KBr pellets. Nuclear magnetic resonance spectra (^1H, ^{13}C, and ^{29}Si) were recorded on Bruker Avance II 300 MHz NMR Spectrometer (Bruker, Switzerland) and all chemical shifts were reported in ppm relative to TMS as an internal standard in $CDCl_3$.

Synthesis of Ligands

Schiff base ligands were prepared by refluxing the 4-aminoantipyrine (10 mmol) and 2-hydroxyacetophenone/2-hydroxynaphthaldehyde (10 mmol) in the dry ethanol in equimolar ratio for 2 h. Yellowish solids separated out at room temperature which were filtered and recrystallized in the same solvent. The purity of the compound was monitored through thin layer chromatography.

Synthesis of Organosilicon Complexes

Organosilicon complexes were prepared in inert atmosphere using dry nitrogen. Sodium salts of the ligands were prepared by stirring the respective ligand (10 mmol) and sodium metal (10 mmol) in 30 mL dry ethanol in equimolar ratio. A calculated amount of triorganosilicon(IV) chloride was slowly added to the sodium salt of the Schiff base ligand in 1:1 molar ratio and the resulting solution was refluxed for 3 h. The precipitate of NaCl separated out during the course of reaction was removed by filtration. The excess solvent was removed under vacuum to yield the desired compound which was repeatedly washed with dry hexane. The solid was recrystallized using a mixture of dry ethanol and dry hexane to ensure the purity of compound and finally dried under reduced pressure. The elemental analyses of these complexes are given in Table 3.

Table 3 Elemental analyses of ligands and their organosilicon(IV) complexes

Compound number	Ligand/complexes	Molecular formula	Molecularweight	Yield(%)	Analyses (%)			
					Cfound (calcd.)	Hfound (calcd.)	Nfound (calcd.)	Sifound (calcd.)
1	HL_1	$C_{18}H_{17}N_3O_2$	307.35	74	70.55 (70.34)	5.79 (5.58)	13.91 (13.67)	–
2	HL_2	$C_{19}H_{19}N_3O_2$	321.15	80	71.34 (71.01)	5.79 (5.96)	13.29 (13.03)	–
3	HL_3	$C_{24}H_{21}N_3O_2$	383.44	76	75.39 (75.18)	5.79 (5.52)	10.75 (10.96)	–
4	HL_4	$C_{22}H_{19}N_3O_2$	357.41	85	73.71 (73.93)	5.58 (5.36)	11.97 (11.76)	–
5	$Et_3Si(L_1)$	$C_{25}H_{34}N_3O_2Si$	436.64	65	68.98 (68.77)	8.07 (7.85)	9.91 (9.62)	6.67 (6.43)
6	$Bu_3Si(L_1)$	$C_{31}H_{46}N_3O_2Si$	520.80	70	71.76 (71.49)	8.78 (8.90)	8.32 (8.07)	6.70 (6.39)
7	$Ph_3Si(L_1)$	$C_{37}H_{34}N_3O_2Si$	580.77	72	76.35 (76.52)	6.14 (5.90)	7.01 (7.24)	4.61 (4.84)
8	$Et_3Si(L_2)$	$C_{26}H_{36}N_3O_2Si$	450.67	68	69.54 (69.29)	8.31 (8.05)	9.57 (9.32)	6.56 (6.23)
9	$Bu_3Si(L_2)$	$C_{32}H_{48}N_3O_2Si$	534.83	67	71.59 (71.86)	9.27 (9.05)	7.61 (7.86)	5.52 (5.25)
10	$Ph_3Si(L_2)$	$C_{38}H_{36}N_3O_2Si$	594.80	71	76.99 (76.73)	6.38 (6.10)	7.29 (7.06)	4.97 (4.72)
11	$Et_3Si(L_3)$	$C_{31}H_{38}N_3O_2Si$	512.74	65	72.79 (72.62)	7.19 (7.47)	8.43 (8.20)	5.69 (5.48)
12	$Bu_3Si(L_3)$	$C_{37}H_{50}N_3O_2Si$	596.90	68	74.23 (74.45)	8.65 (8.44)	7.29 (7.04)	4.92 (4.71)
13	$Ph_3Si(L_3)$	$C_{43}H_{38}N_3O_2Si$	656.87	66	78.87 (78.62)	5.67 (5.83)	6.68 (6.40)	4.56 (4.28)
14	$Et_3Si(L_4)$	$C_{29}H_{36}N_3O_2Si$	486.70	69	71.81 (71.57)	7.68 (7.46)	8.81 (8.63)	5.90 (5.77)
15	$Bu_3Si(L_4)$	$C_{35}H_{48}N_3O_2Si$	570.86	67	73.31 (73.64)	8.71 (8.48)	7.57 (7.36)	5.15 (4.92)
16	$Ph_3Si(L_4)$	$C_{41}H_{36}N_3O_2Si$	630.83	70	78.29 (78.06)	5.98 (5.75)	6.89 (6.66)	4.67 (4.45)

Antimicrobial Activity

Antimicrobial activities of ligands and their corresponding organosilicon complexes were carried out using a twofold serial dilution technique. The stock solution was prepared by dissolving the compounds in dry DMSO with a concentration of 100 μg mL^{-1}. For in vitro antimicrobial activity, these compounds were screened against Gram-positive bacteria (*S. aureus* and *B. subtilis*) and Gram-negative bacteria (*E. coli*) and fungi (*A. niger* and *C. albicans*). The incubation period of *A. niger* and *C. albicans* was 7 d at 25 $\pm$ 1 °C and 36 h at 37 $\pm$ 1 °C, respectively, whereas for bacteria it was 24 h at 37 $\pm$ 1 °C. The conventional bactericides tetracycline, chloramphenicol, kanamycin, cefazoline sodium, and cefotaxime and fungicides cycloheximide, carbendazim, and fluconazole were used as standards for comparing the activity of compounds.

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