Electronic Supporting Information for

Efficient Hydrosilylation of Carbonyls by Zinc Alkyl Complexes Supported by Flexible Amidophosphino-Chalcogenide Ligands

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1. X-ray crystallographic studies of complexes 2a and 2b:

Single crystals of air-sensitive zinc complexes **2a** and **2b** were grown from the saturated THF/Toluene solvent at room temperature. For compounds **2a** and **2b**, a crystal of suitable dimensions was mounted on a CryoLoop (Hampton Research Corp.) with a layer of light mineral oil at 293(2) K and all measurements were made using an Agilent Supernova XCalibur Eos CCD detector with graphite monochromatic Mo (0.71073 Å) radiation for **2a** and **2b** were used. Crystal data and structure refinement parameters are summarized in Table S1. The structures were solved by direct methods (SIR92)[1,2] and refined on F^2 by full-matrix least-squares methods using SHELXL-97.[3] Non-hydrogen atoms were anisotropically refined. H atoms were included in the refinement in calculated positions riding on their carrier atoms. The ORTEP-3 program was used to draw molecules. Crystallographic data (excluding structure factors) for the structures reported in this article have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. CCDC 2408530 (**2a**), 2408531 (**2b**).

Table S1. Crystal refinement parameters for the compounds **2a**, and **2b**.

Crystal Parameters	**2a**	**2b**
CCDC No	2408530	2408531
Empirical formula	$C_{40}H_{54}N_4O_2P_2Se_2Zn_2$	$C_{40}H_{54}N_4O_2P_2S_2Zn_2$
Formula weight	973.51	879.67
T (K)	273(2) K	273(2) K
λ (Å)	0.71073	0.71073
Crystal system	Monoclinic	Monoclinic
Space group	$P\ 2_1/c$	$P\ 2_1/c$
a(Å)	12.3838(5)	12.2096(7)
b (Å)	11.5142(4)	11.4911(8)
c(Å)	15.4241(6)	15.4394(10)
α (°)	90	90
β (°)	106.6260 (10)	107.070(2)
γ (°)	90	90
V(Å^3)	2107.37(14)	2070.7(2)
Z	2	2

Density calc (g cm^{-3})	1.534	1.411
μ (mm^{-1})	2.980	1.375
F(000)	992	920
Theta range for data collection	2.242 to 27.121	2.487 to 27.146
Limiting indices	-15<=h<=15, -14<=k<=13, -19<=l<=19	-14<=h<=15, -14<=k<=14, -19<=l<=19
Reflections collected / unique	41324 / 4650 [R(int) = 0.0745]	46643 / 4584 [R(int) = 0.0717]
Completeness to theta	100.0 %	99.9 %
Refinement method	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2
Data / restraints / parameters	4650 / 0 / 236	4584 / 0 / 236
Goodness-of-fit on F^2	1.023	1.093
Final R indices [I>2sigma(I)]	R_1 = 0.0327, wR_2 = 0.0607	R_1 = 0.0372, wR_2 = 0.0948
R indices (all data)	R_1 = 0.0615, wR_2 = 0.0688	R_1 = 0.0502, wR_2 = 0.1005

2. ^{1}H, $^{13}C\{^{1}H\}$, and $^{31}P\{^{1}H\}$ NMR Spectra of 2a and 2b.

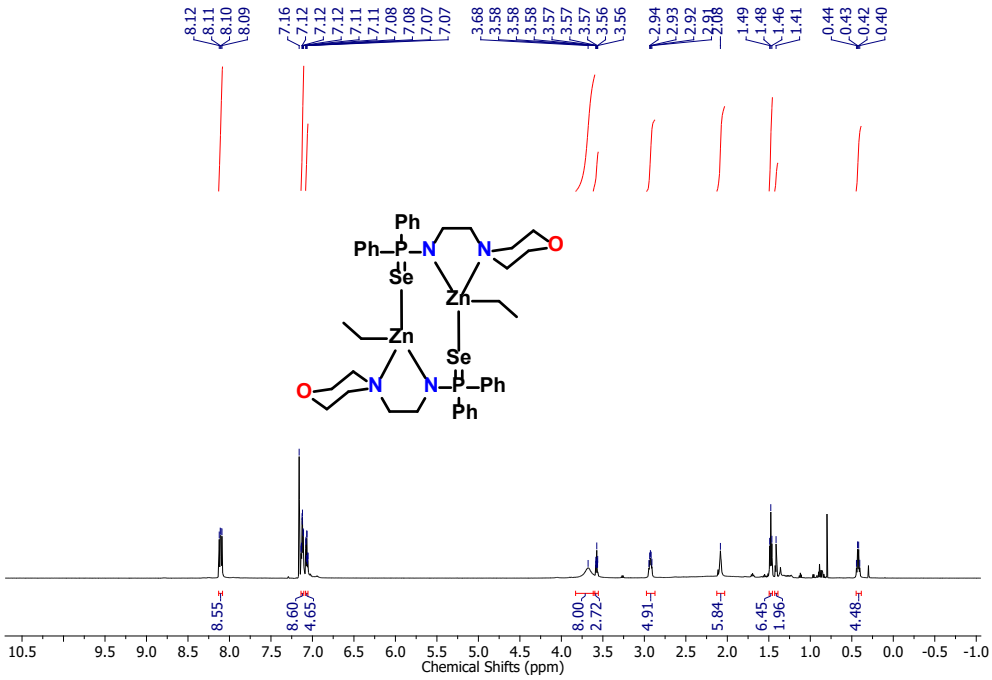


Figure S1: ^{1}H NMR spectrum (600 MHz, 25 °C, C_6D_6) of **2a**.

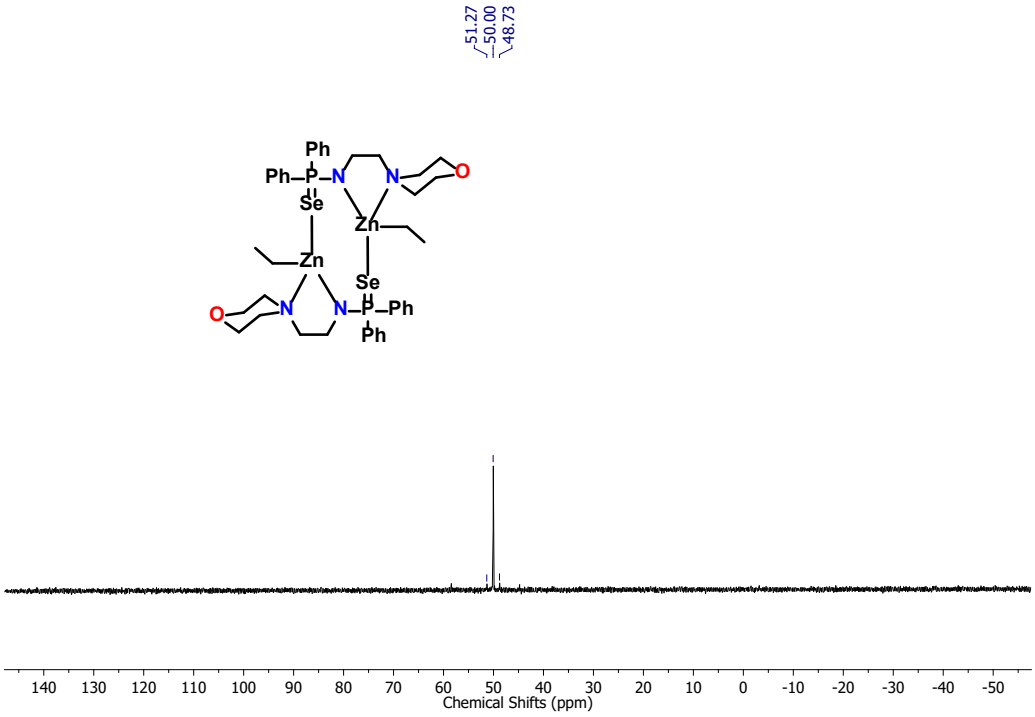


Figure S2: $^{31}P\{^{1}H\}$ NMR spectrum (243 MHz, 25 °C, C_6D_6) of **2a**.

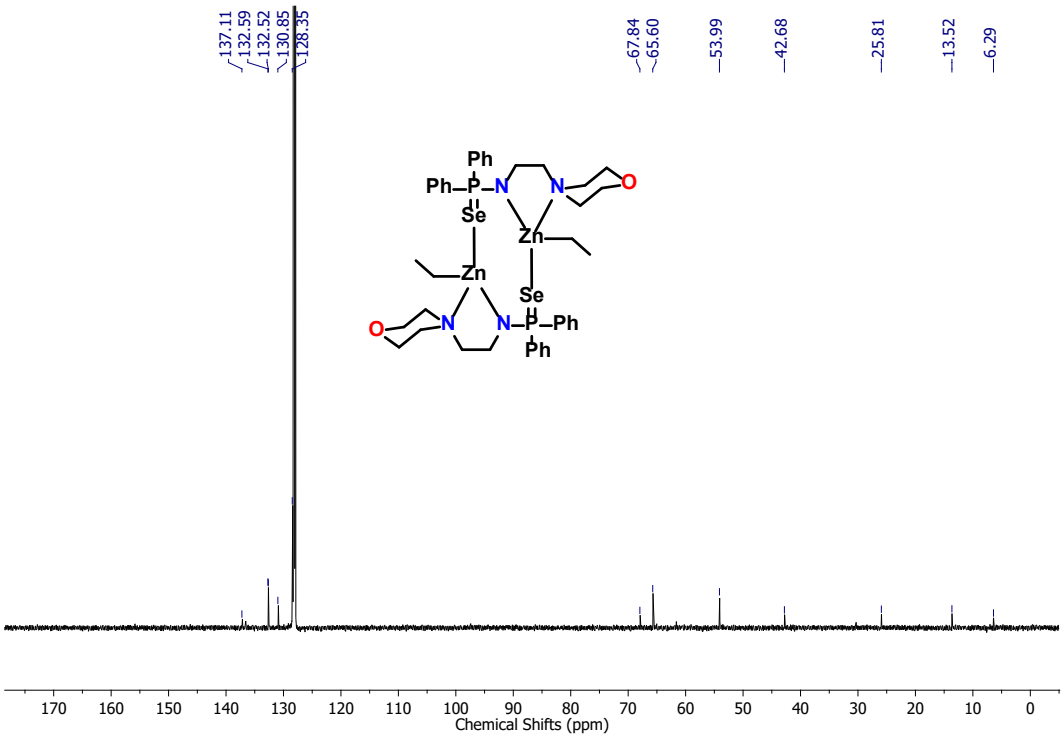


Figure S3: ^{13}C {^{1}H} NMR spectrum (150 MHz, 25 °C, C_6D_6) of **2a**.

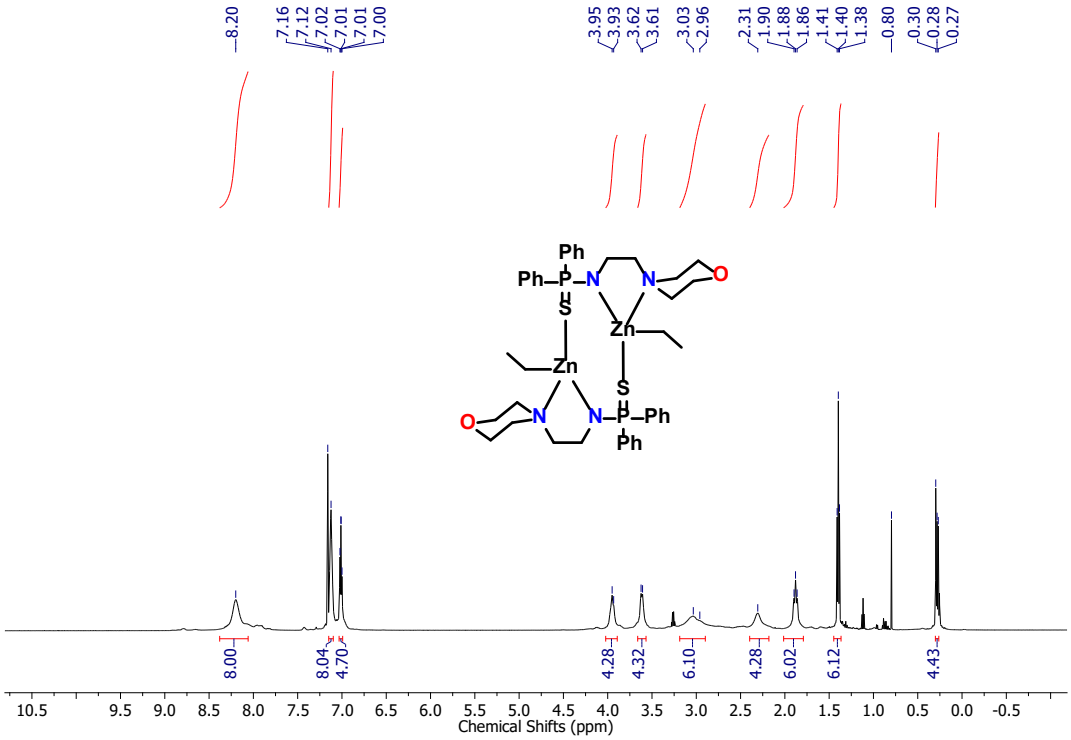


Figure S4: ^{1}H NMR spectrum (600 MHz, 25 °C, C_6D_6) of **2b**.

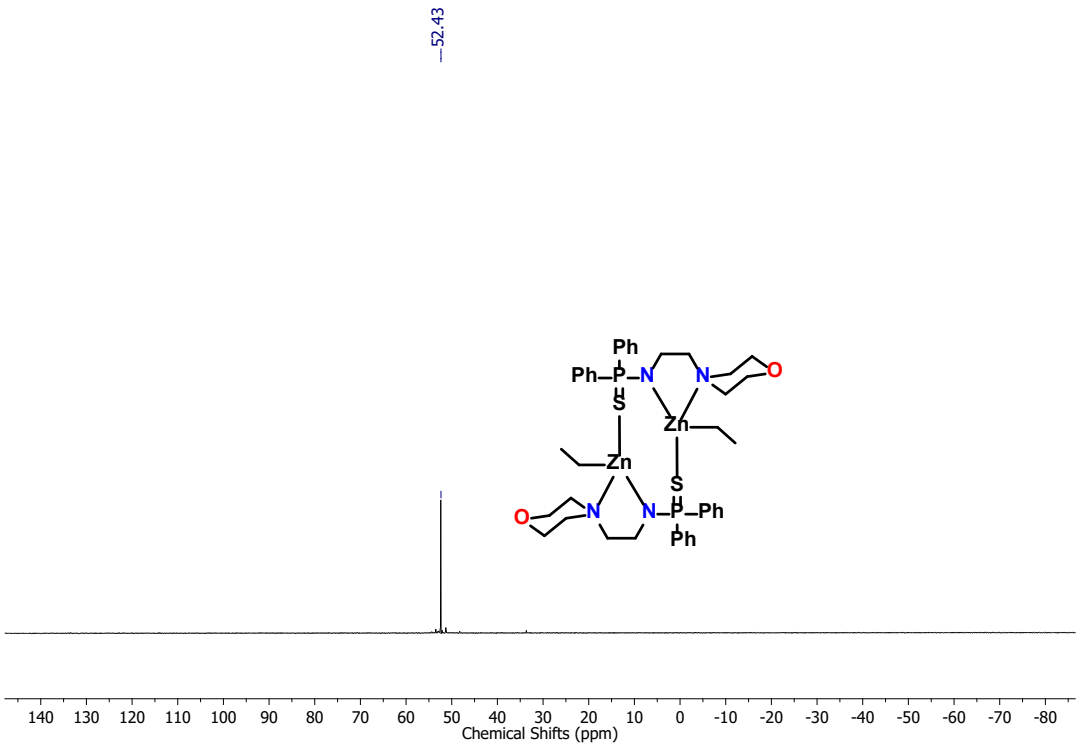


Figure S5: $^{31}P\{^{1}H\}$ NMR spectrum (243 MHz, 25 °C, C_6D_6) of **2b**.

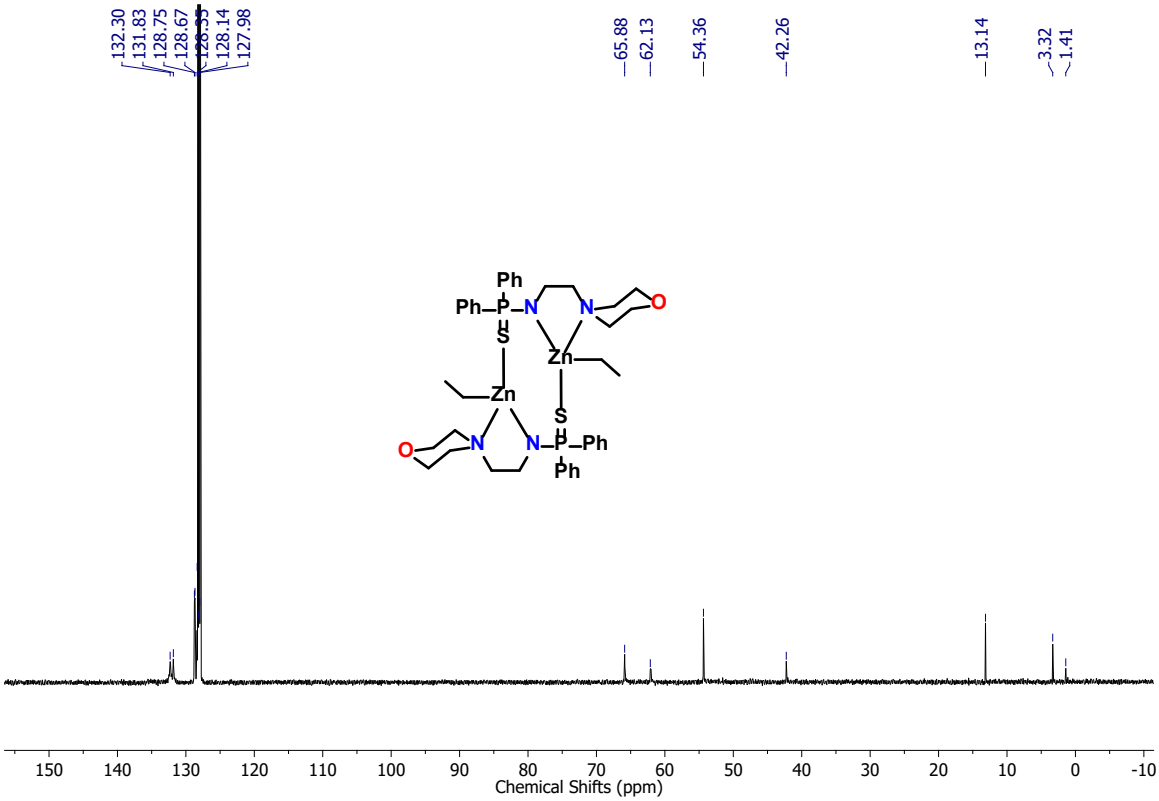


Figure S6: $^{13}C\{^{1}H\}$ NMR spectrum (150 MHz, 25 °C, C_6D_6) of **2b**.

3. NMR spectra for the mono-hydrosilylation of benzaldehyde (control reaction).

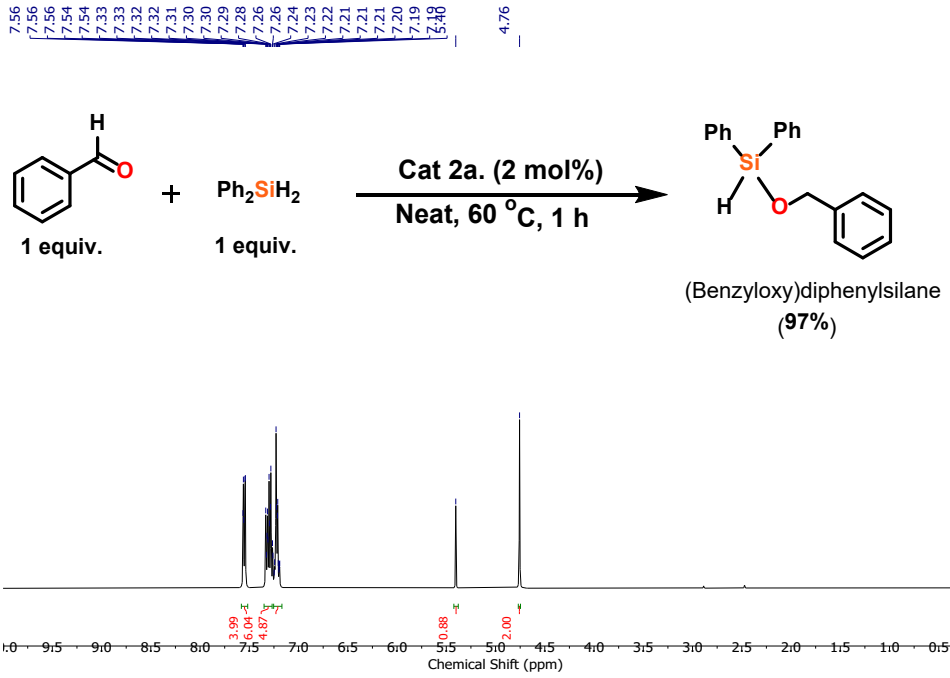


Figure S7: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of (Benzyloxy)diphenylsilane.

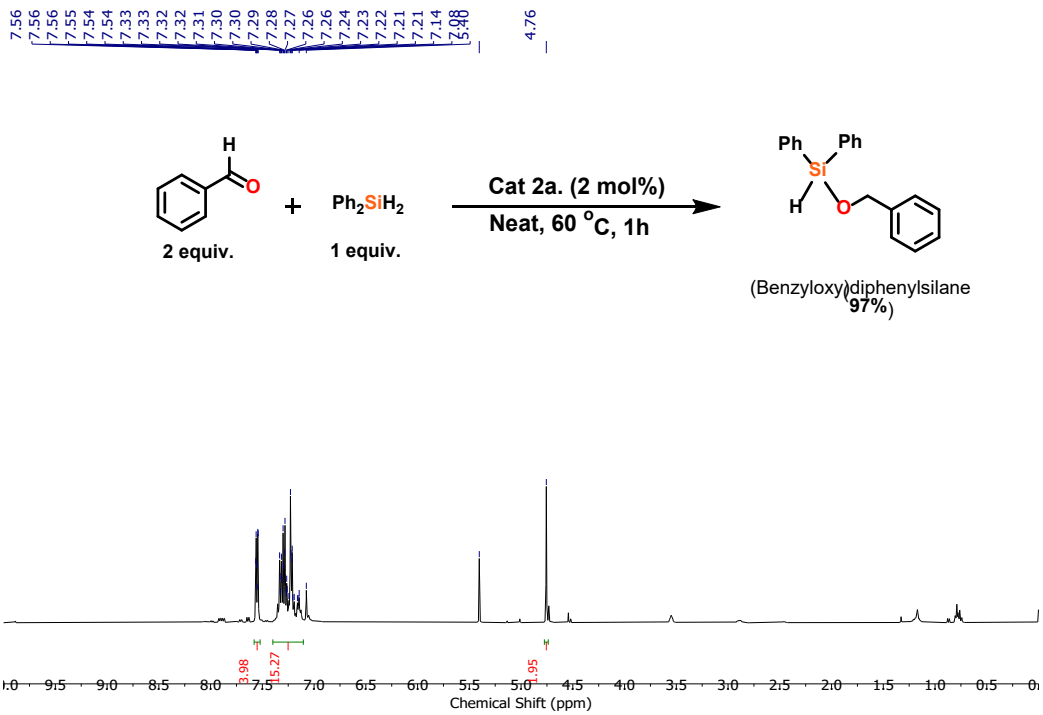
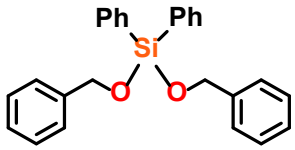


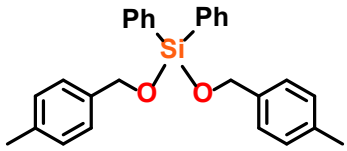
Figure S8: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of (Benzyloxy)diphenylsilane.

4. NMR data for the hydrosilylation of aldehydes.



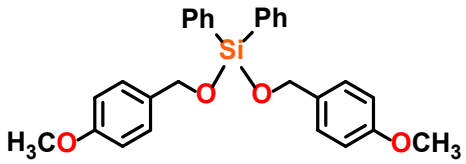
bis(benzyloxy)diphenylsilane (3a)[4]:

Following the procedure, the product was isolated with column chromatography using eluent hexane/ethylacetate 5:1 (R_f = 0.5). Isolated Yield: 97 mg (98 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.66 – 7.62 (m, 4H, Ar*H*), 7.32 (dd, *J* = 5.1, 3.5 Hz, 3H, ArH), 7.29 (d, *J* = 7.3 Hz, 3H, Ar*H*), 7.22 (dd, *J* = 6.5, 5.2 Hz, 8H, Ar*H*), 7.15 (d, *J* = 6.5 Hz, 2H, Ar*H*), 4.74 (s, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 140.4 (C_{Ar}), 135.1 (C_{Ar}), 132.4 (C_{Ar}), 131.8 (C_{Ar}), 130.6 (C_{Ar}), 128.6 (C_{Ar}), 128.5 (C_{Ar}), 128.1 (C_{Ar}), 127.3 (C_{Ar}), 126.6 (C_{Ar}), 65.0 (CH$_2$) ppm.



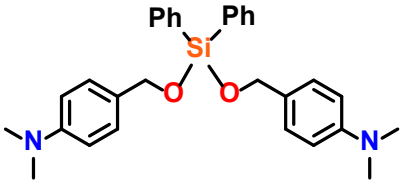
bis((4-methylbenzyl)oxy)diphenylsilane (3b)[4]:

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.52). Isolated Yield: 102 mg (97 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.66 – 7.61 (m, 4H, Ar*H*), 7.33 (d, *J* = 7.4 Hz, 2H, Ar*H*), 7.31 – 7.25 (m, 4H, Ar*H*), 7.12 (d, *J* = 8.4 Hz, 4H, Ar*H*), 7.03 (d, *J* = 7.9 Hz, 4H, Ar*H*), 4.70 (s, 4H, CH_2), 2.24 (s, 6H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 137.5 (C_{Ar}), 136.9 (C_{Ar}), 135.1 (C_{Ar}), 132.6 (C_{Ar}), 131.9 (C_{Ar}), 130.5 (C_{Ar}), 129.0 (C_{Ar}), 128.1 (C_{Ar}), 128.0 (C_{Ar}), 126.8 (C_{Ar}), 65.0 (CH$_2$), 21.3 (CH$_3$) ppm.



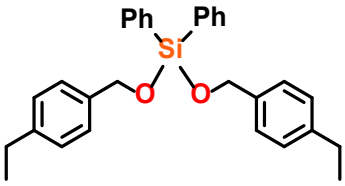
bis((4-methoxybenzyl)oxy)diphenylsilane (3c)[4]:

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.7). Isolated Yield: 110 mg (97 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.65 – 7.59 (m, 4H, Ar*H*), 7.33 (d, *J* = 7.3 Hz, 2H, Ar*H*), 7.28 (dt, *J* = 8.2, 3.7 Hz, 4H, Ar*H*), 7.13 (dd, *J* = 8.2, 1.5 Hz, 4H, Ar*H*), 6.78 – 6.72 (m, 4H, Ar*H*), 4.65 (s, 4H, CH_2), 3.70 (s, 6H, OCH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 158.9 (C_{Ar}), 135.1 (C_{Ar}), 132.7 (C_{Ar}), 130.5 (C_{Ar}), 128.3 (C_{Ar}), 128.0 (C_{Ar}), 113.8 (C_{Ar}), 64.8 (CH_2), 55.4 (OCH_3) ppm.



4,4'-(((diphenylsilanediyl)bis(oxy))bis(methylene))bis(N,N-dimethylaniline) (3d)[4]:

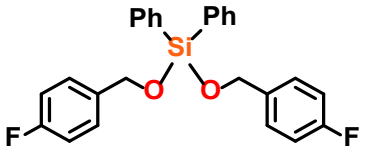
Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 3:2 (R_f = 0.8). Isolated Yield: 99 mg (97 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.63 (dd, *J* = 7.9 Hz, 1.4 Hz, 4H, Ar*H*), 7.32 – 7.21 (m, 6H, Ar*H*), 7.10 (d, *J* = 8.4 Hz, 4H, Ar*H*), 6.59 (d, *J* = 8.6 Hz, 4H, Ar*H*), 4.64 (s, 4H, CH_2), 2.81 (s, 12H, N(CH_3)$_2$) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 150.1 (C_{Ar}), 135.5 (C_{Ar}), 133.1 (C_{Ar}), 130.3 (C_{Ar}), 128.7 (C_{Ar}), 128.6 (C_{Ar}), 128.3 (C_{Ar}), 127.9 (C_{Ar}), 112.6 (C_{Ar}), 65.0 (CH_2), 40.8 (N(CH_3)$_2$) ppm.



bis((4-ethylbenzyl)oxy)diphenylsilane (3e):

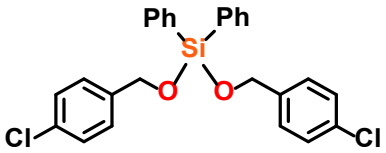
Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.6). Isolated Yield: 108 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.67 – 7.59 (m, 4H, Ar*H*), 7.33 – 7.22 (m, 6H, Ar*H*), 7.13 (d, *J* = 8.1 Hz, 4H, Ar*H*), 7.03 (d, *J* = 8.1 Hz, 4H, Ar*H*), 4.69 (s, 4H, CH_2), 2.52 (q, *J* = 7.6 Hz, 4H, CH_2), 1.11 (t, *J* = 7.6 Hz, 6H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 143.3 (C_{Ar}), 137.7 (C_{Ar}), 135.1

(C_{Ar}), 132.6 (C_{Ar}), 130.5 (C_{Ar}), 128.0 (C_{Ar}), 127.9 (C_{Ar}), 126.9 (C_{Ar}), 65.0 (CH_2), 28.7 (CH_2), 15.8 (CH_3) ppm.



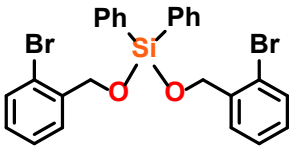
bis((4-fluorobenzyl)oxy)diphenylsilane (3f)[4]:

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.8). Isolated Yield: 102 mg (95 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.60 (dd, J = 7.9 Hz, 1.4 Hz, 4H, ArH), 7.38 – 7.34 (m, 2H, ArH), 7.29 (dd, J = 11.2, 4.4 Hz, 4H, ArH), 7.16 (dd, J = 8.6 Hz, 5.6 Hz, 4H, ArH), 6.93 – 6.84 (m, J = 8.7 Hz, 4H, ArH), 4.68 (s, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 163.4 (C_{Ar}), 161.0 (C_{Ar}), 136.1 (C_{Ar}), 136.0 (C_{Ar}), 135.1 (C_{Ar}), 132.2 (C_{Ar}), 130.7 (C_{Ar}), 128.4 (C_{Ar}), 128.2 (C_{Ar}), 128.1 (C_{Ar}), 115.3 (C_{Ar}), 115.1 (C_{Ar}), 64.5 (CH_2) ppm.



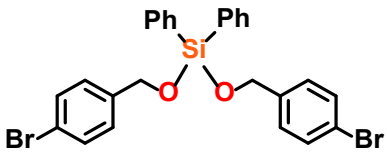
bis((4-chlorobenzyl)oxy)diphenylsilane (3g)[4]:

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.7). Isolated Yield: 111 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.64 – 7.57 (m, 4H, ArH), 7.37-7.27 (m, 6H, ArH), 7.19 – 7.10 (m, 6H, ArH), 4.68 (s, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 138.8 (C_{Ar}), 135.0 (C_{Ar}), 133.1 (C_{Ar}), 132.0 (C_{Ar}), 130.8 (C_{Ar}), 128.5 (C_{Ar}), 128.2 (C_{Ar}), 128.0 (C_{Ar}), 64.5 (CH_2) ppm.



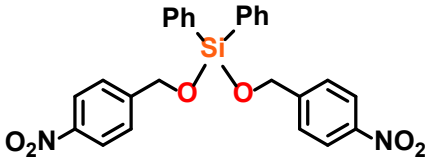
bis((2-bromobenzyl)oxy)diphenylsilane (3h)[4]:

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.6). Isolated Yield: 132 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.84 – 7.77 (m, 4H, Ar*H*), 7.68 – 7.64 (m, 2H, Ar*H*), 7.51 – 7.47 (m, 4H, Ar*H*), 7.44 (ddd, *J* = 8.4, 2.4, 1.1 Hz, 4H, Ar*H*), 7.33 (td, *J* = 7.6, 1.1 Hz, 2H, Ar*H*), 7.13 (ddd, *J* = 7.9, 4.6, 1.3 Hz, 2H, Ar*H*), 4.94 (s, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 139.3 (C_{Ar}), 135.1 (C_{Ar}), 132.3 (C_{Ar}), 132.0 (C_{Ar}), 130.8 (C_{Ar}), 128.6 (C_{Ar}), 128.2 (C_{Ar}), 128.1 (C_{Ar}), 127.5 (C_{Ar}), 121.6 (C_{Ar}), 64.9 (CH_2) ppm.



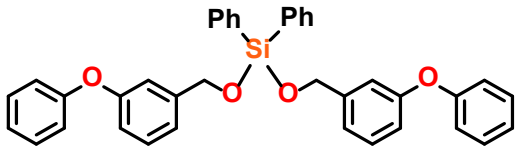
bis((4-bromobenzyl)oxy)diphenylsilane (3i)[4]:

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.61). Isolated Yield: 132 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.75 – 7.70 (m, 4H, Ar*H*), 7.49 – 7.40 (m, 10H, Ar*H*), 7.18 (d, *J* = 8.4 Hz, 4H, Ar*H*), 4.78 (s, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 139.3 (C_{Ar}), 135.0 (C_{Ar}), 131.9 (C_{Ar}), 131.5 (C_{Ar}), 130.8 (C_{Ar}), 128.3 (C_{Ar}), 128.1 (C_{Ar}), 121.2 (C_{Ar}), 64.5 (CH$_2$) ppm.



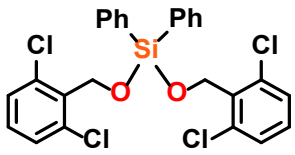
bis((4-nitrobenzyl)oxy)diphenylsilane (3j)[4]:

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 7:3 (R_f = 0.9). Isolated Yield: 115 mg (95 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 8.04 (ddd, *J* = 11.5 Hz, 8.0 Hz, 1.1 Hz, 4H, Ar*H*), 7.80 – 7.75 (m, 4H, Ar*H*), 7.68 (td, *J* = 7.8, 1.2 Hz, 2H, Ar*H*), 7.51 – 7.40 (m, 8H, Ar*H*), 5.30 (s, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 146.5 (C_{Ar}), 136.9 (C_{Ar}), 134.9 (C_{Ar}), 134.1 (C_{Ar}), 131.3 (C_{Ar}), 131.1 (C_{Ar}), 128.4 (C_{Ar}), 127.9 (C_{Ar}), 127.8 (C_{Ar}), 124.8 (C_{Ar}), 62.3 (CH$_2$) ppm.



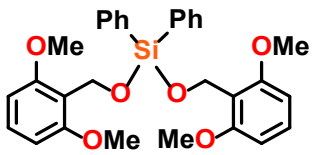
bis((3-phenoxybenzyl)oxy)diphenylsilane (3k):

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 4:1 (R_f = 0.5). Isolated Yield: 134 mg (93 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.73 – 7.68 (m, 4H, Ar*H*), 7.50 – 7.45 (m, 2H, Ar*H*), 7.41 – 7.33 (m, 8H, Ar*H*), 7.29 (dd, *J* = 15.2, 7.4 Hz, 2H, Ar*H*), 7.17 – 7.10 (m, 2H, Ar*H*), 7.10 – 7.02 (m, 8H, Ar*H*), 6.96 – 6.91 (m, 2H, Ar*H*), 4.82 (s, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 157.5 (C_{Ar}), 157.2 (C_{Ar}), 142.5 (C_{Ar}), 135.0 (C_{Ar}), 132.1 (C_{Ar}), 130.6 (C_{Ar}), 129.8 (C_{Ar}), 129.7 (C_{Ar}), 128.1 (C_{Ar}), 123.4 (C_{Ar}), 121.2 (C_{Ar}), 119.2 (C_{Ar}), 117.6 (C_{Ar}), 116.7 (C_{Ar}), 64.6 (CH$_2$) ppm.



bis((2,6-dichlorobenzyl)oxy)diphenylsilane (3l):

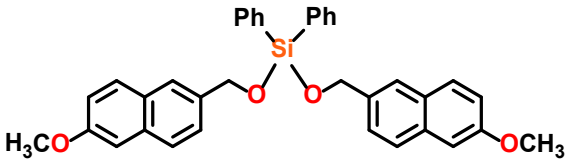
Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 4:1 (R_f = 0.5). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.66 – 7.63 (m, 2H, Ar*H*), 7.57 – 7.55 (m, 2H, Ar*H*), 7.33 – 7.23 (m, 8H, Ar*H*), 7.16 (d, *J* = 8.0 Hz, 3H, Ar*H*), 7.04 – 7.01 (m, 1H, Ar*H*), 5.01 (d, *J* = 13.2 Hz, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 136.7 (d, *J* = 4.0 Hz, C_{Ar}), 135.2 (d, *J* = 7.0 Hz, C_{Ar}), 134.8 (C_{Ar}), 133.8 (C_{Ar}), 132.3 (C_{Ar}), 130.5 (d, *J* = 3.1 Hz, C_{Ar}), 129.8 (d, *J* = 19.3 Hz, C_{Ar}), 128.4 (t, *J* = 5.2 Hz, C_{Ar}), 128.1 (C_{Ar}), 127.8 (C_{Ar}), 61.9 (CH$_2$), 60.5 (CH$_2$) ppm.



bis((2,6-dimethoxybenzyl)oxy)diphenylsilane (3m):

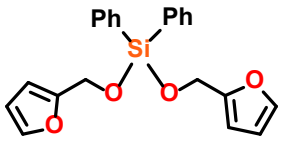
Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 7:3 (R_f = 0.7). Isolated Yield: 119 mg (93 %). ^{1}H NMR (400 MHz, 25 °C,

$CDCl_3$) δ_H 7.77 (d, *J* = 6.9 Hz, 2H, Ar*H*), 7.67 (d, *J* = 6.8 Hz, 2H, Ar*H*), 7.42 (dd, *J* = 17.4, 10.1 Hz, 8H, Ar*H*), 6.50 (d, *J* = 8.2 Hz, 2H, Ar*H*), 6.42 (s, 2H, Ar*H*), 4.89 (s, 4H, CH_2), 3.82 (s, 6H, OCH_3), 3.72 (s, 6H, OCH_3) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 160.1 (C_{Ar}), 157.6 (C_{Ar}), 135.1 (C_{Ar}), 134.8 (C_{Ar}), 134.3 (C_{Ar}), 133.3 (C_{Ar}), 130.3 (C_{Ar}), 130.2 (C_{Ar}), 129.4 (C_{Ar}), 128.6 (C_{Ar}), 128.0 (C_{Ar}), 127.8 (C_{Ar}), 121.5 (C_{Ar}), 103.8 (C_{Ar}), 98.2 (C_{Ar}), 98.11 (C_{Ar}), 62.1 (CH_2), 60.3 (CH_2), 55.4 (OCH_3), 55.1 (OCH_3) ppm.



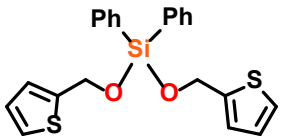
bis((6-methoxynaphthalen-2-yl)methoxy)diphenylsilane (3n):

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 4:1 (R_f = 0.4). Isolated Yield: 131 mg (95 %). ^1H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.92 (d, *J* = 6.6 Hz, 4H, Ar*H*), 7.71 (t, *J* = 10.3 Hz, 4H, ArH), 7.56 – 7.47 (m, 8H, Ar*H*), 7.35 (d, *J* = 7.1 Hz, 1H, Ar*H*), 7.30 – 7.15 (m, 6H, Ar*H*), 5.06 (s, 4H, CH_2), 3.96 (s, 6H, OCH_3) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 157.6 (C_{Ar}), 135.5 (C_{Ar}), 135.2 (C_{Ar}), 133.9 (C_{Ar}), 132.6 (C_{Ar}), 130.6 (C_{Ar}), 129.5 (C_{Ar}), 129.1 (C_{Ar}), 128.8 (C_{Ar}), 128.3 (C_{Ar}), 128.1 (C_{Ar}), 126.9 (C_{Ar}), 125.8 (C_{Ar}), 125.4 (C_{Ar}), 125.3 (C_{Ar}), 118.8 (C_{Ar}), 105.7 (C_{Ar}), 65.4 (CH_2), 55.3 (OCH_3) ppm.



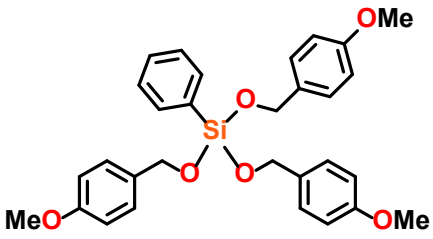
bis(furan-2-ylmethoxy)diphenylsilane (3o):

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.4). Isolated Yield: 90 mg (96 %). ^1H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.61 – 7.55 (m, 4H, Ar*H*), 7.30 (dd, *J* = 5.2, 3.6 Hz, 2H, Ar*H*), 7.28 – 7.22 (m, 6H, Ar*H*), 6.18 (dd, *J* = 3.1, 1.9 Hz, 2H, ArH), 6.09 (d, *J* = 2.9 Hz, 2H, Ar*H*), 4.64 (s, 4H, CH_2) ppm. $^{13}C\{^1H\}$ NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 153.5 (C_{Ar}), 142.4 (C_{Ar}), 135.1 (C_{Ar}), 132.1 (C_{Ar}), 130.5 (C_{Ar}), 127.9 (C_{Ar}), 110.3 (C_{Ar}), 108.1 (C_{Ar}), 57.8 (CH_2) ppm.



diphenylbis(thiophen-2-ylmethoxy)silane (3p):

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.25). Isolated Yield: 97 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.61 (dd, *J* = 7.9 Hz, 1.4 Hz, 4H, Ar*H*), 7.31 (dd, *J* = 5.1 Hz, 3.7 Hz, 2H, Ar*H*), 7.26 (dd, *J* = 11.2 Hz, 4.4 Hz, 4H, Ar*H*), 7.11 (dd, *J* = 5.1 Hz, 1.2 Hz, 2H, Ar*H*), 6.80 (dd, J = 5.0 Hz, 3.5 Hz, 2H, Ar*H*), 6.77 – 6.72 (m, 2H, Ar*H*), 4.85 (s, 4H, CH_2) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 143.7 (C_{Ar}), 135.1 (C_{Ar}), 131.9 (C_{Ar}), 130.7 (C_{Ar}), 128.0 (C_{Ar}), 126.6 (C_{Ar}), 125.2 (C_{Ar}), 124.9 (C_{Ar}), 60.4 (CH_2) ppm.



tris((4-methoxybenzyl)oxy)(phenyl)silane (3q):

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 5:1 (R_f = 0.61). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.60 – 7.56 (m, 2H, Ar*H*), 7.33 (m, 1H, Ar*H*), 7.27 ((dd, *J* = 11.2, 3.8 Hz, 2H, Ar*H*), 7.15 – 7.10 (m, 6H, Ar*H*), 6.77 – 6.72 (m, 6H, Ar*H*), 4.67 (s, 6H, CH_2), 3.69 (d, *J* = 1.2 Hz, 9H, OCH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 159.0 (C_{Ar}), 135.1 (C_{Ar}), 132.4 (C_{Ar}), 128.5 (C_{Ar}), 128.0 (C_{Ar}), 114.0 (C_{Ar}), 113.7 (C_{Ar}), 64.8 (C_{Ar}), 55.3 (C_{Ar}) ppm.

5. Spectral data for hydrosilylation of aldehydes:

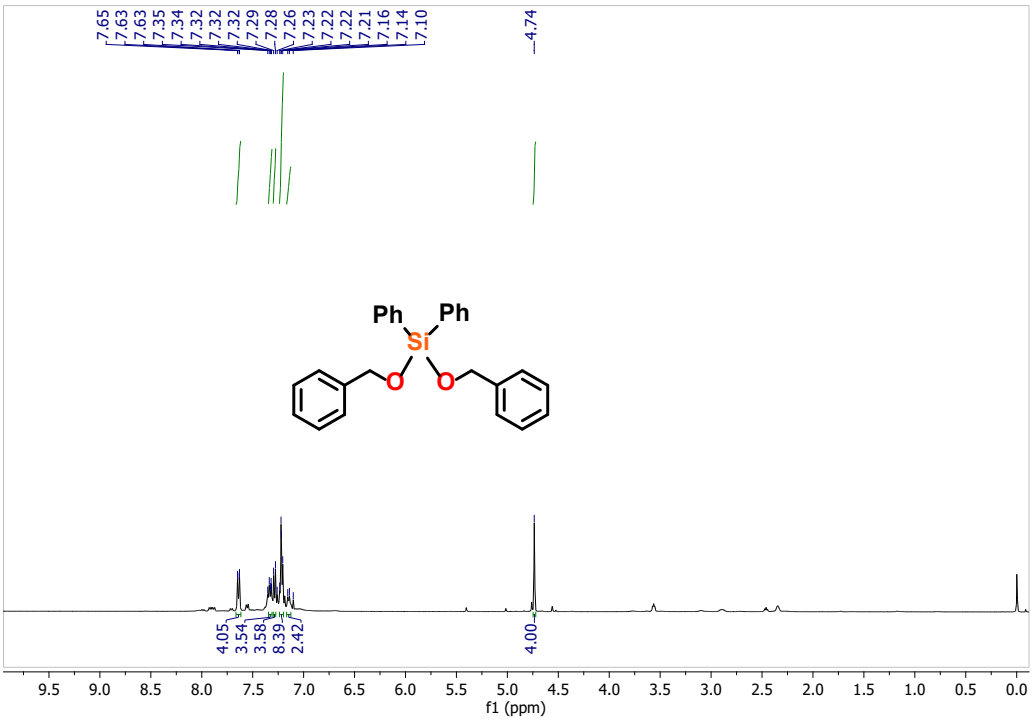


Figure S9: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3a**.

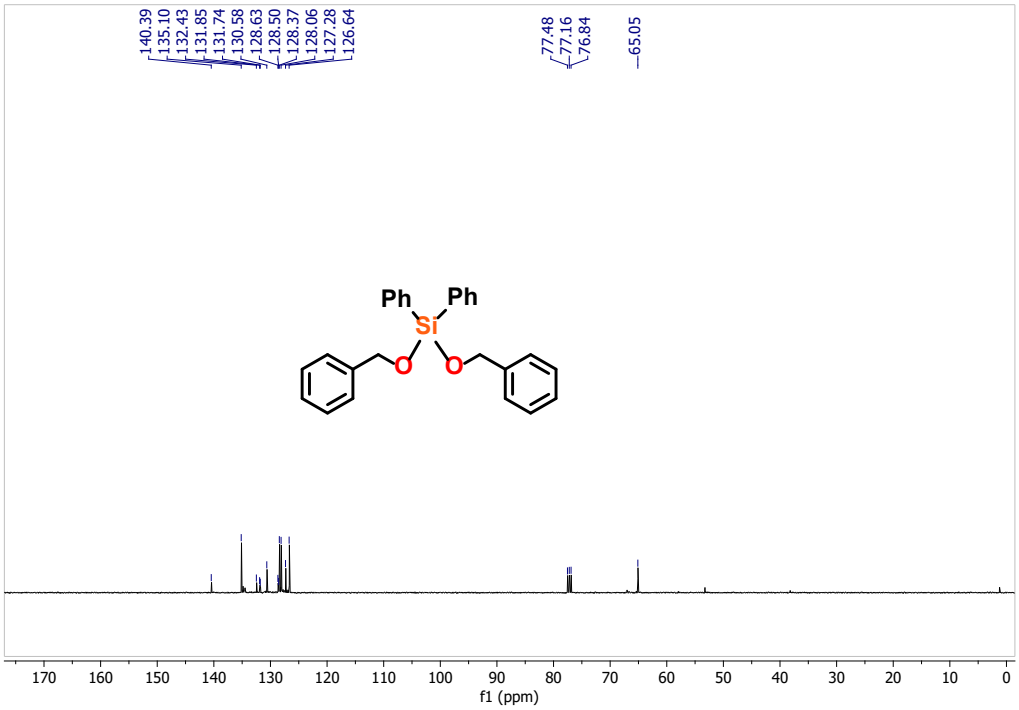


Figure S10: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3a**.

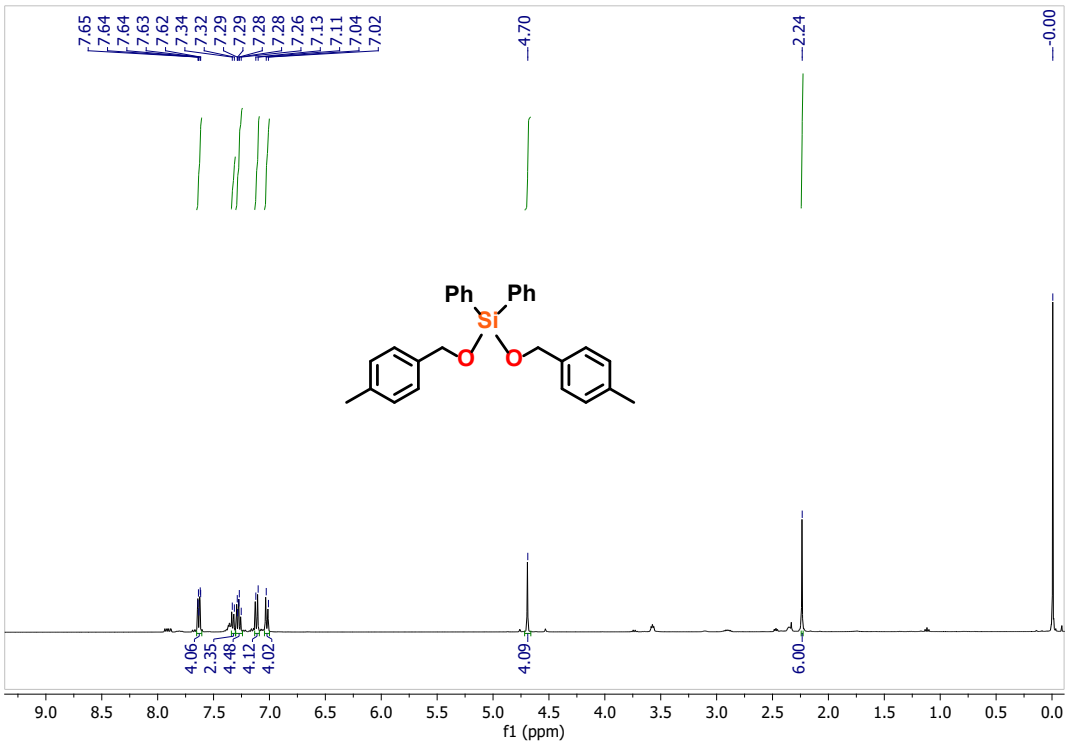


Figure S11: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3b**.

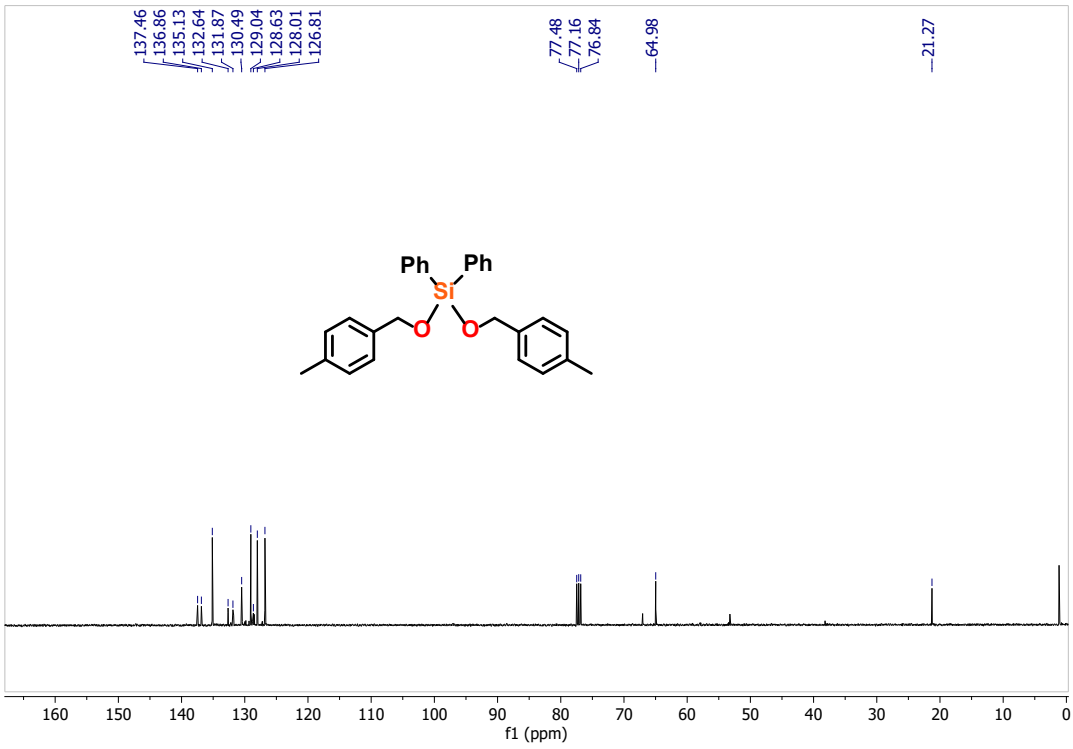


Figure S12: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3b**.

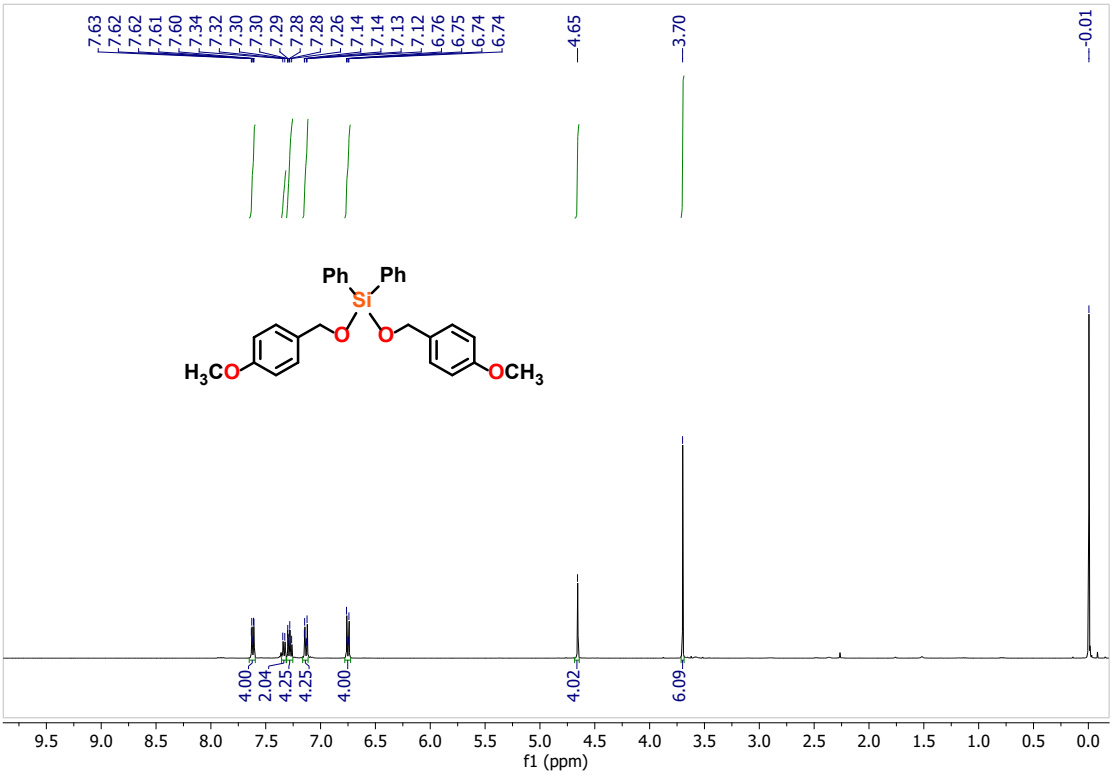


Figure S13: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3c**.

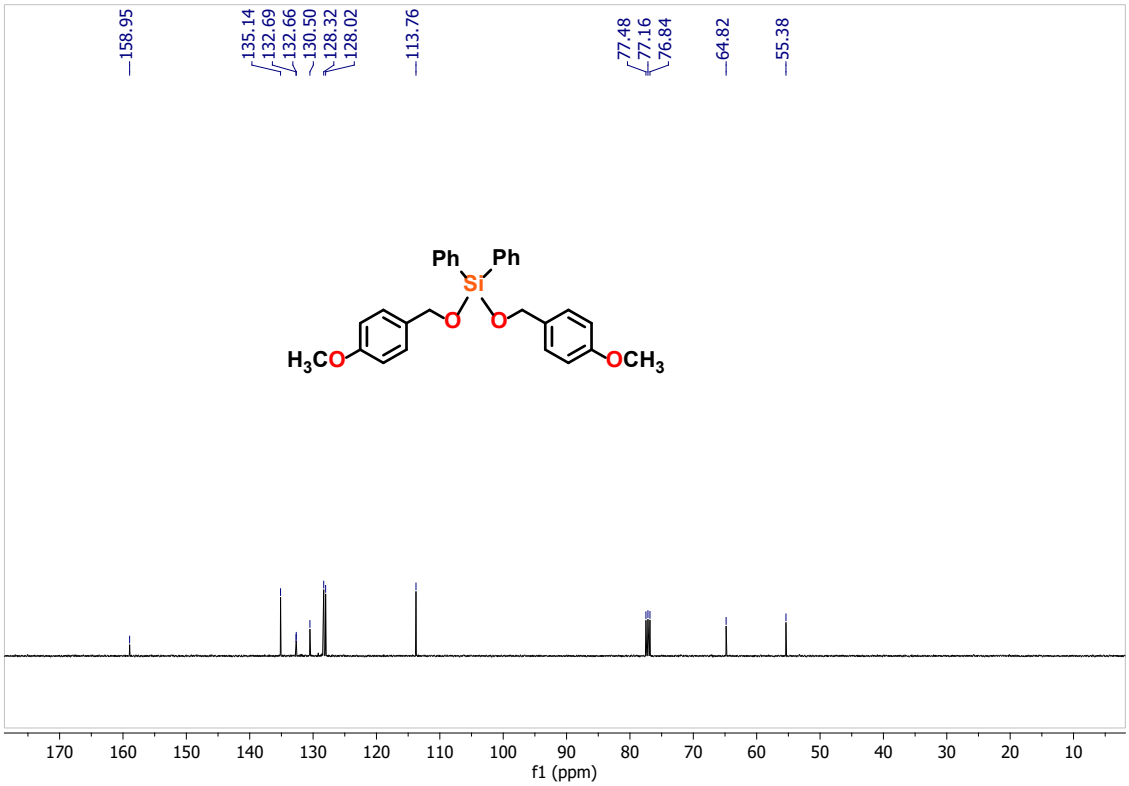


Figure S14: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3c**.

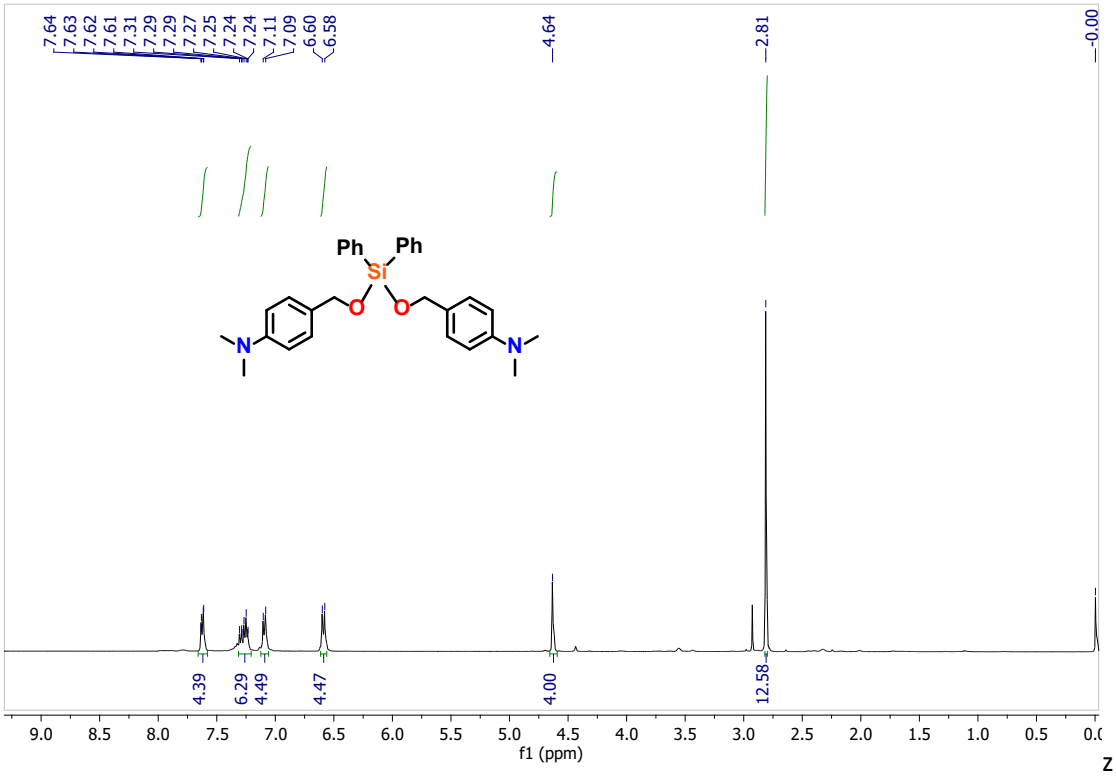


Figure S15: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3d**.

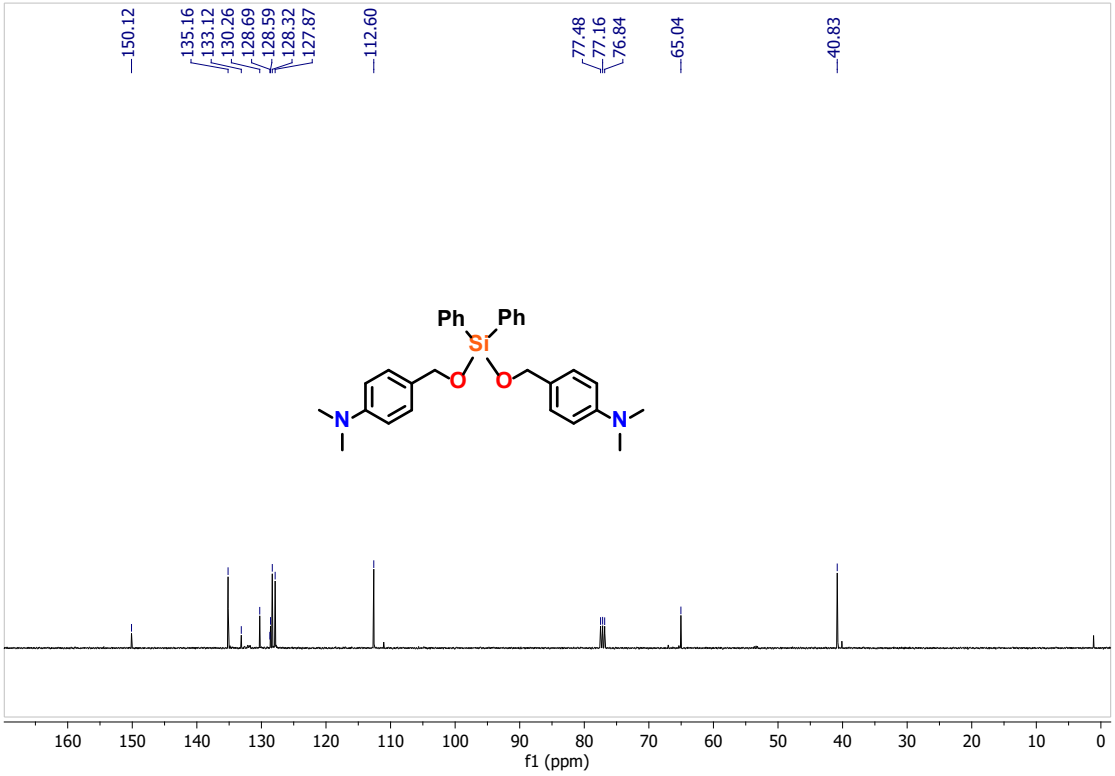


Figure S16: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3d**.

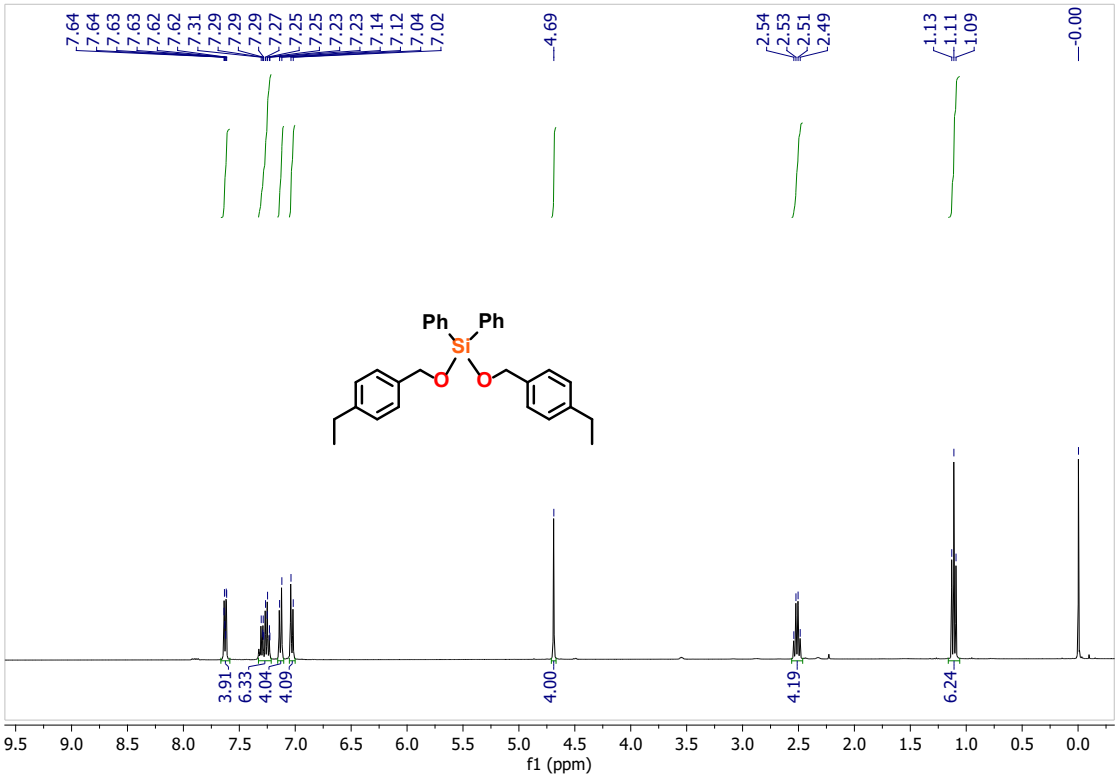


Figure S17: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3e**.

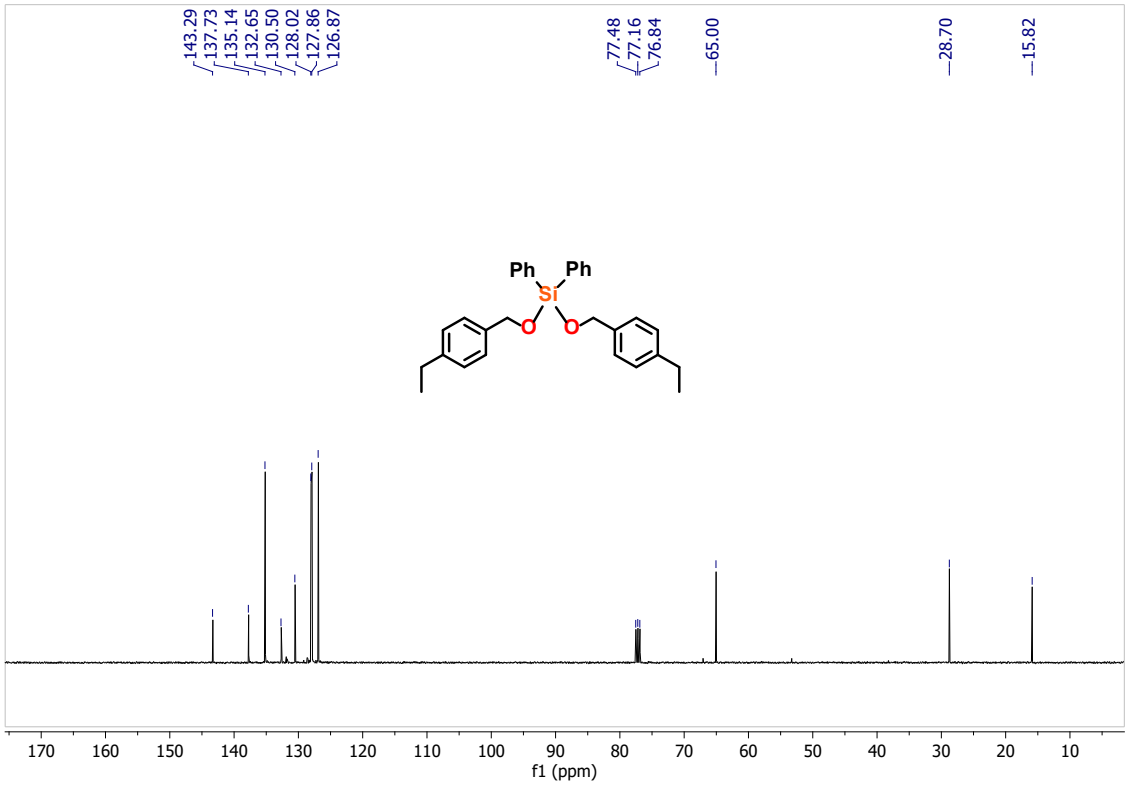


Figure S18: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3e**.

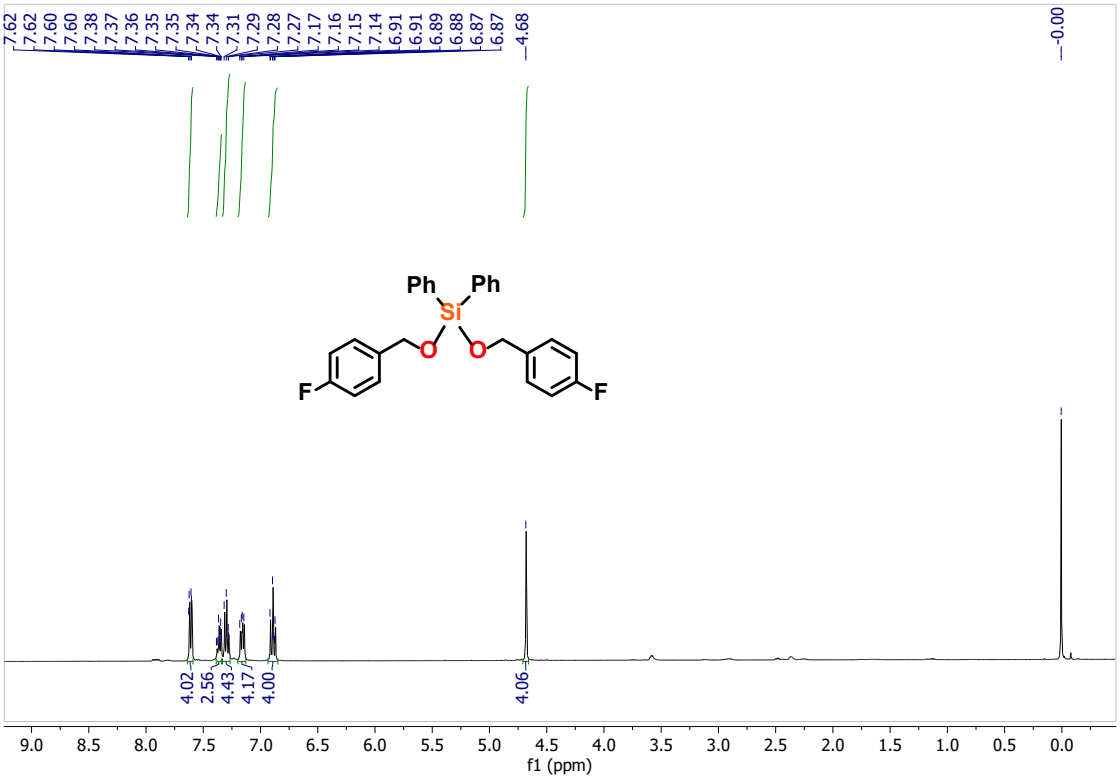


Figure S19: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3f**.

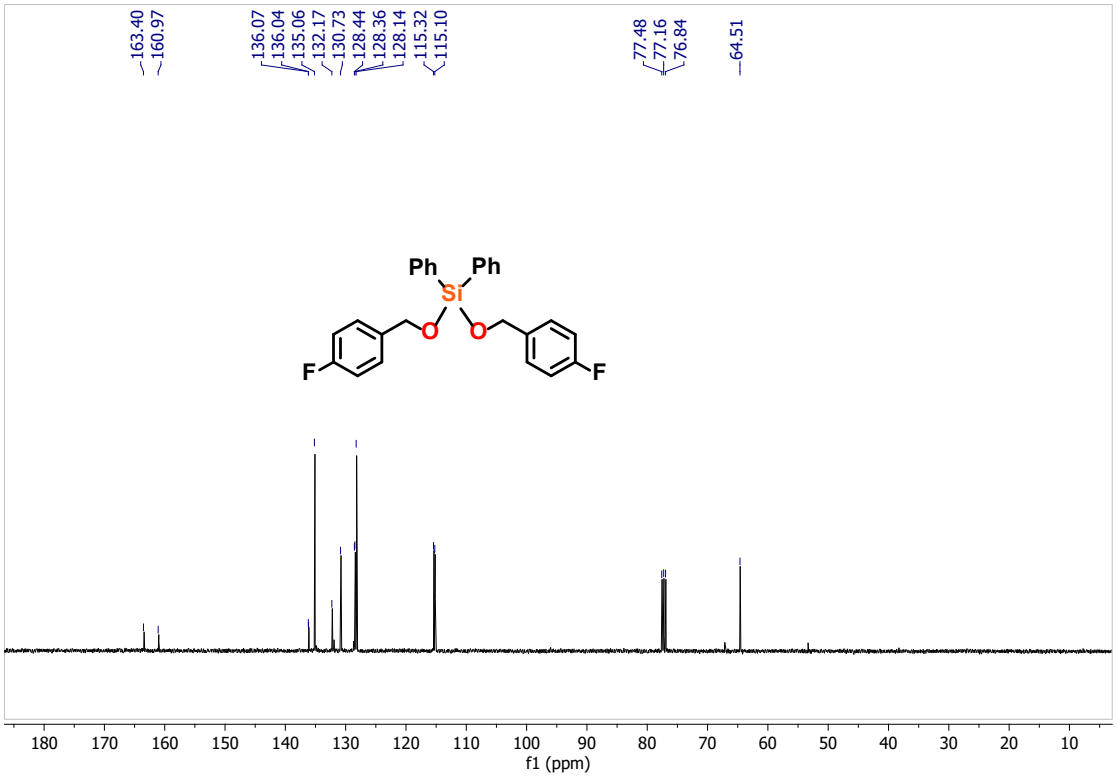


Figure S20: ^{13}C{^{1}H} NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3f**.

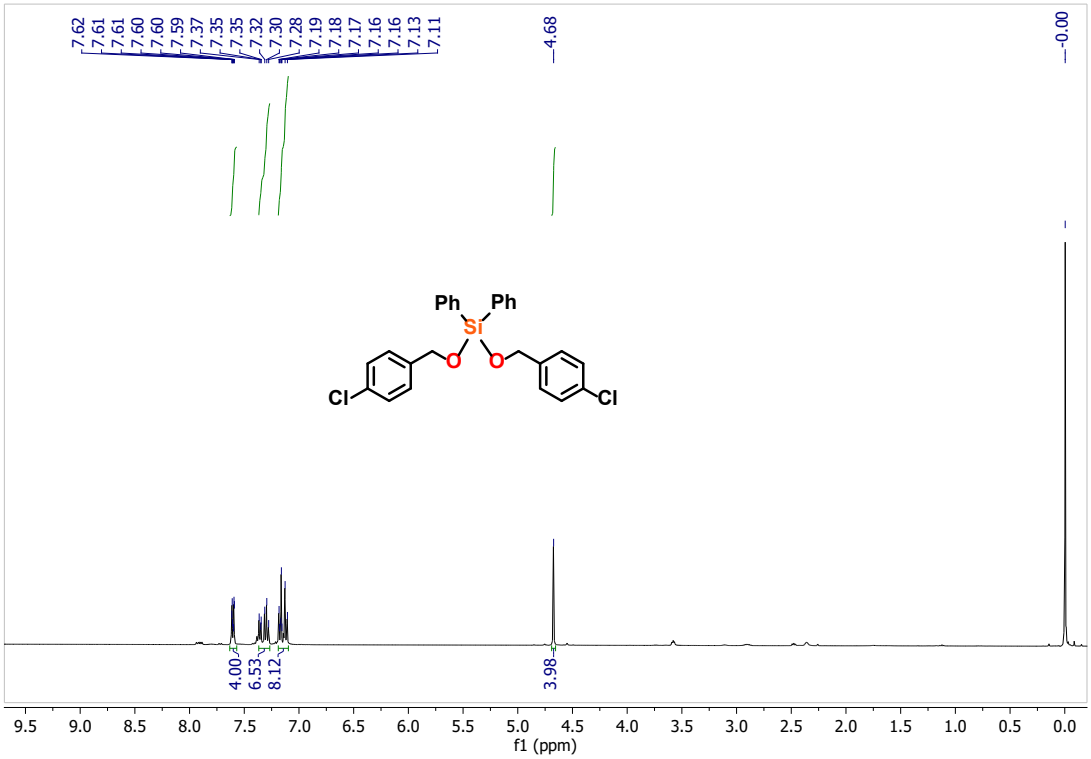


Figure S21: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3g**.

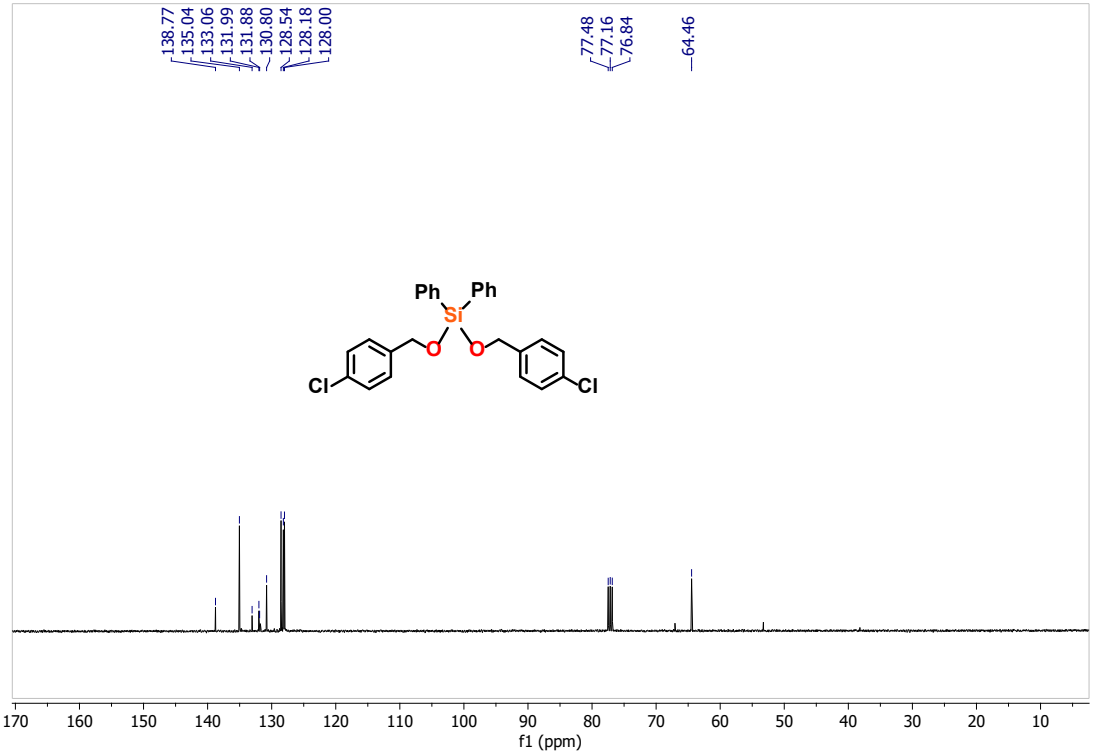


Figure S22: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3g**.

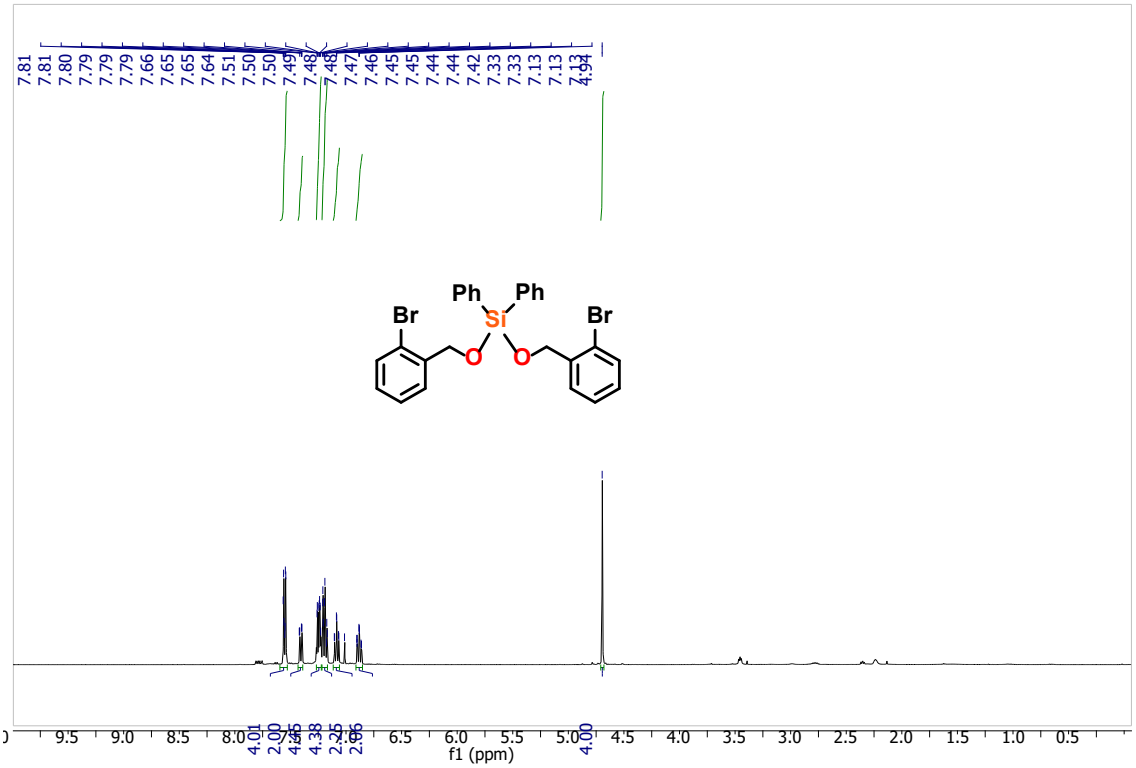


Figure S23: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3h**.

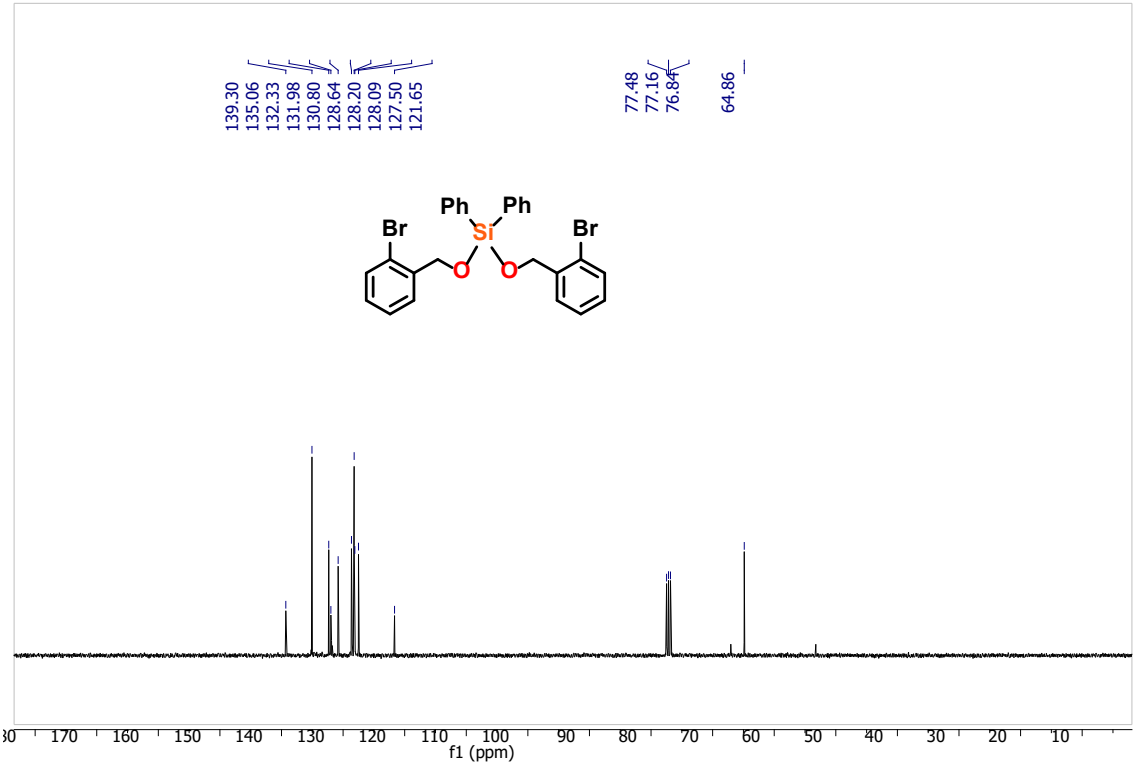


Figure S24: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3h**.

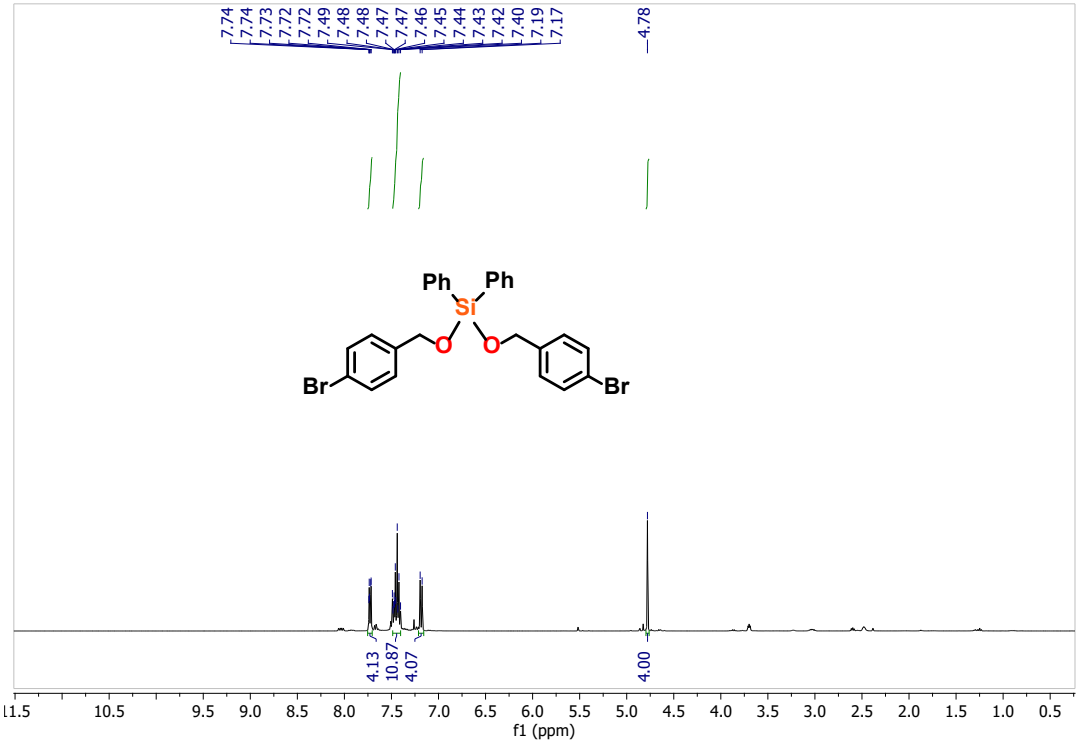


Figure S25: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3i**.

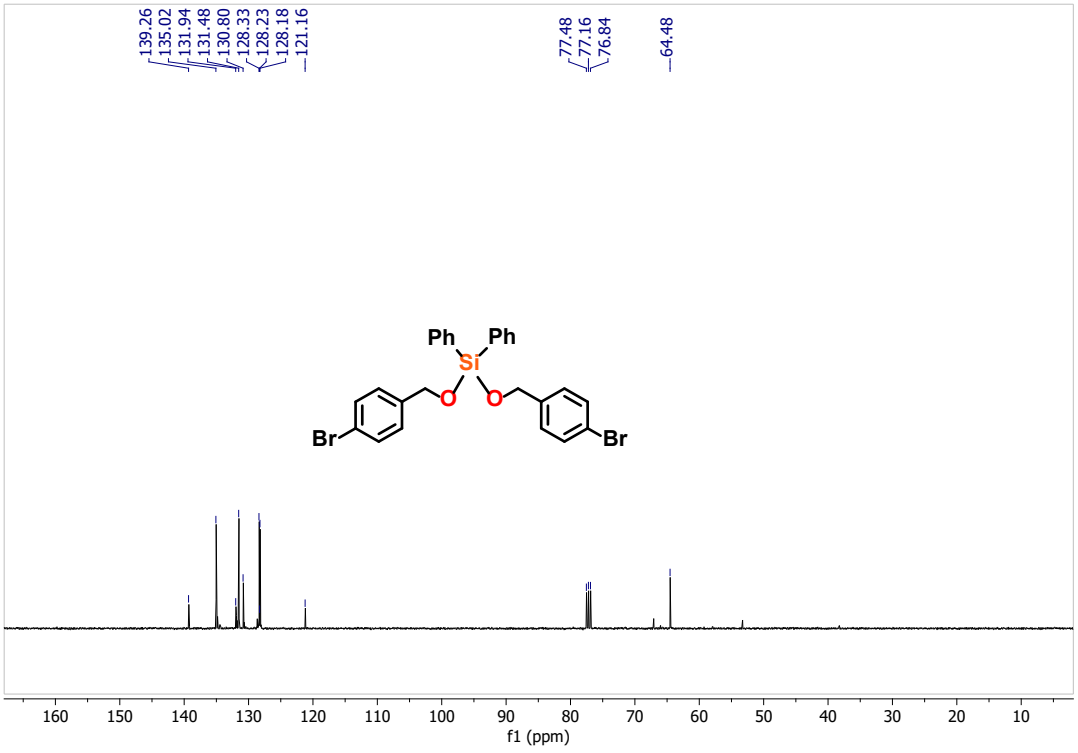


Figure S26: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3i**.

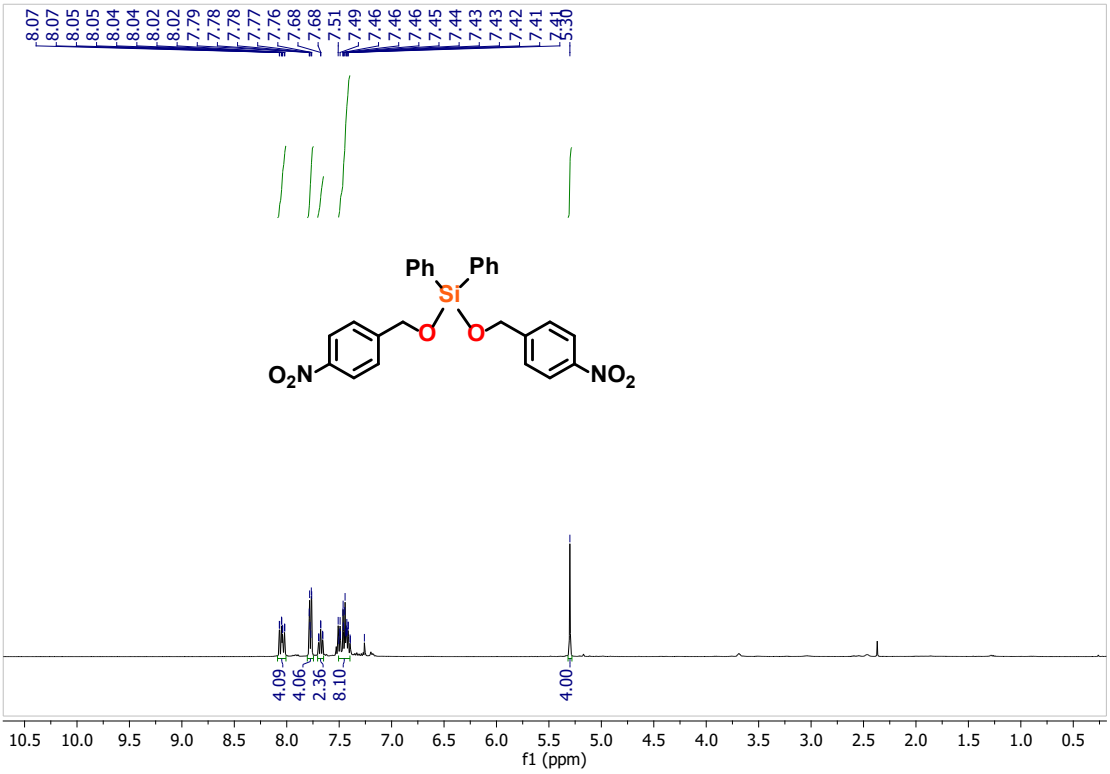


Figure S27: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3j**.

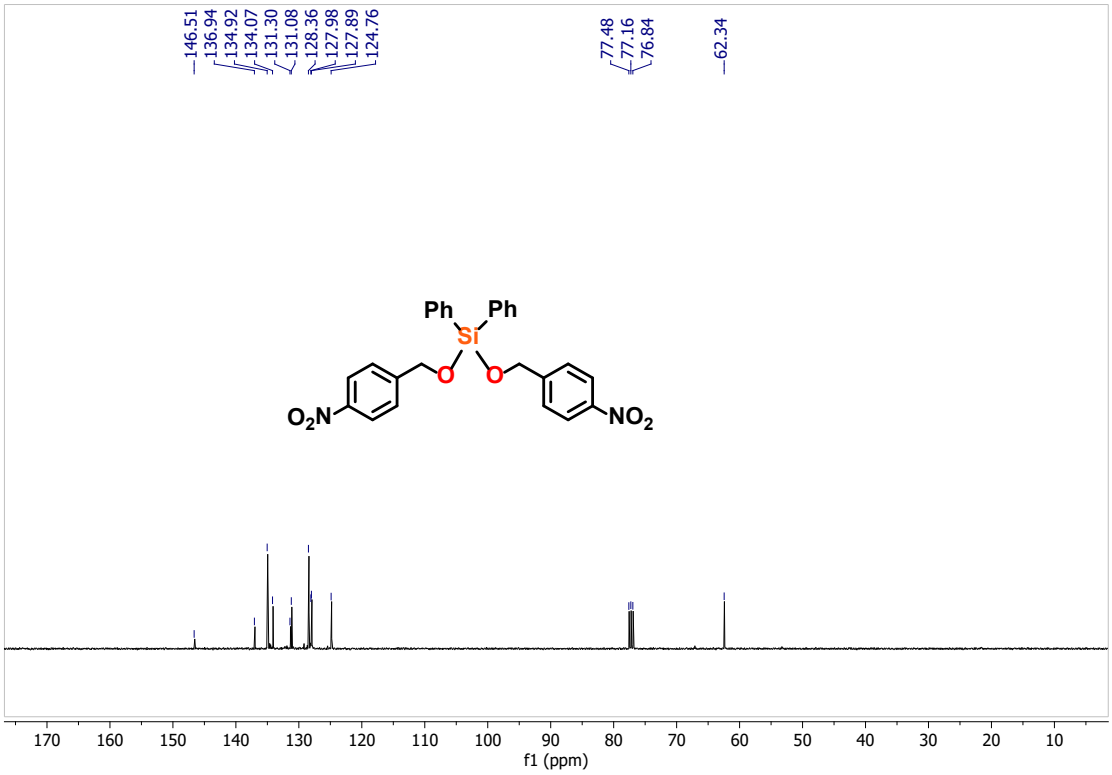


Figure S28: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3j**.

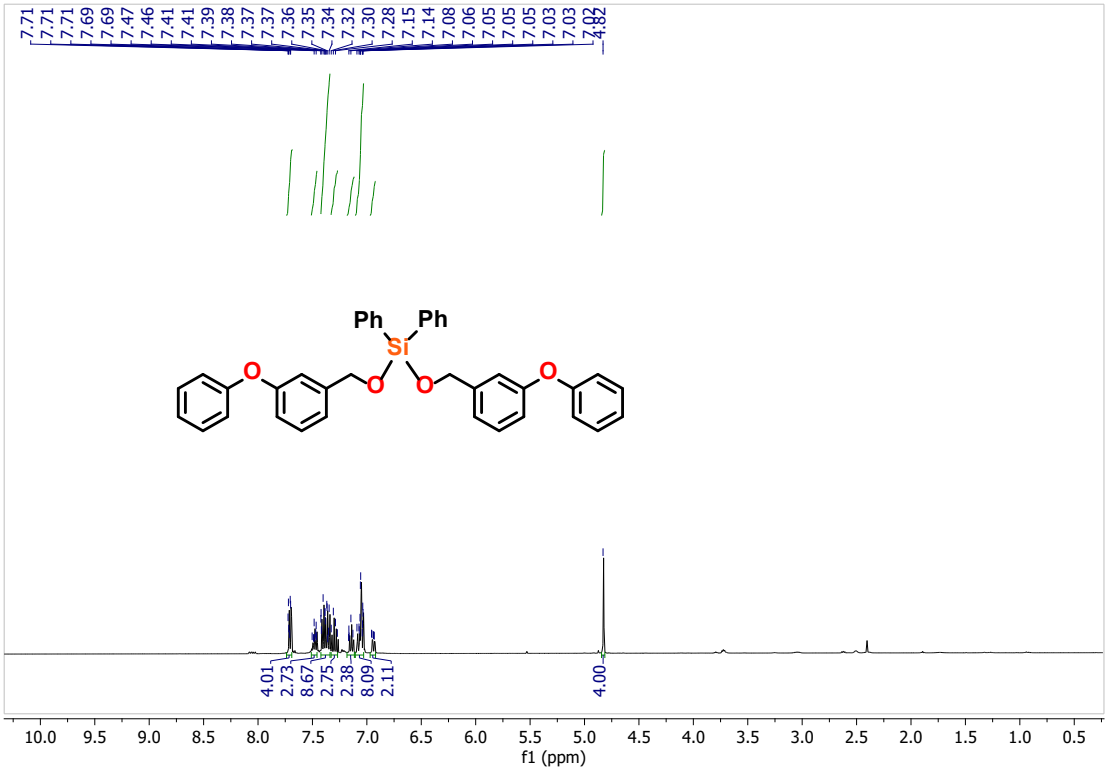


Figure S29: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3k**.

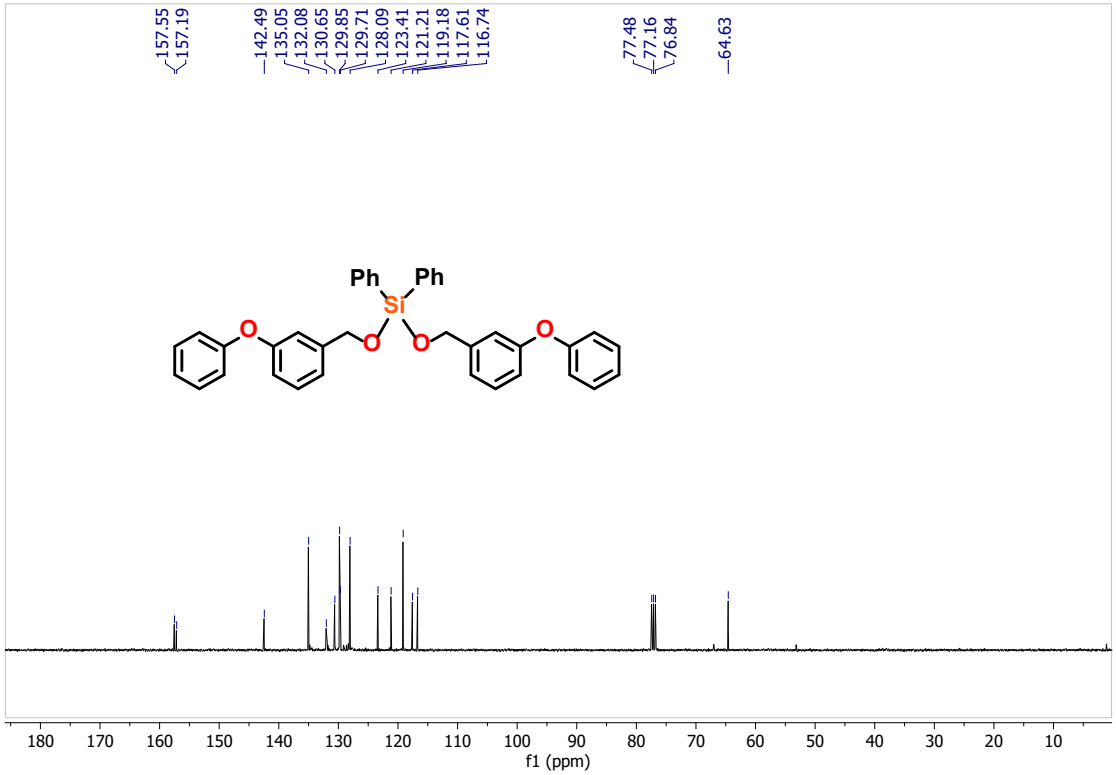


Figure S30: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3k**.

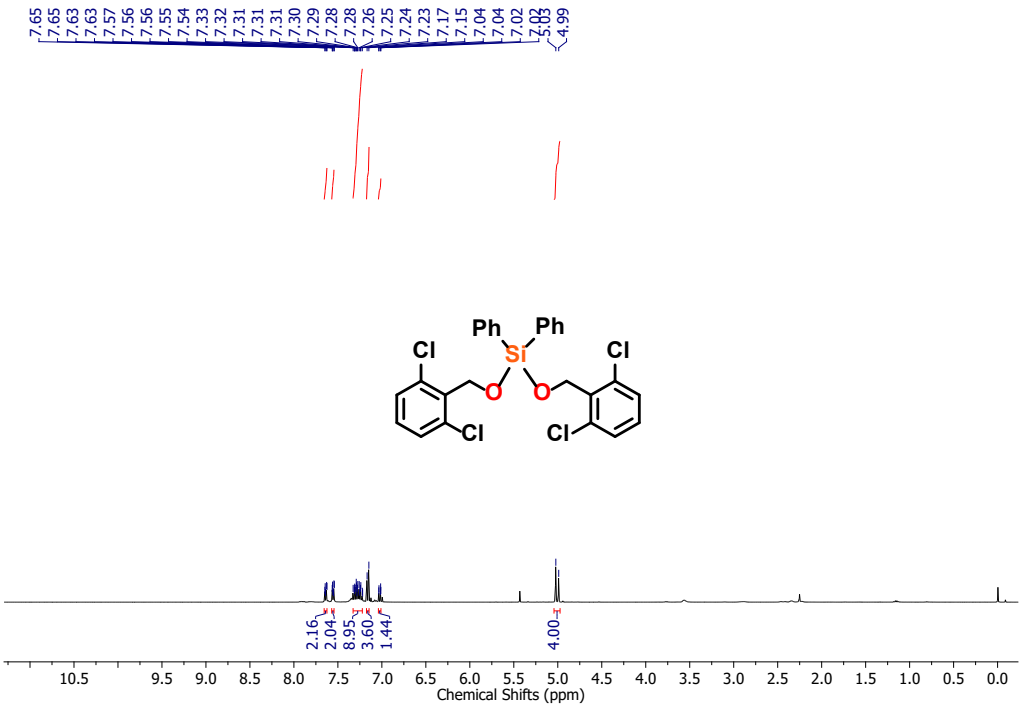


Figure S31: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3l**.

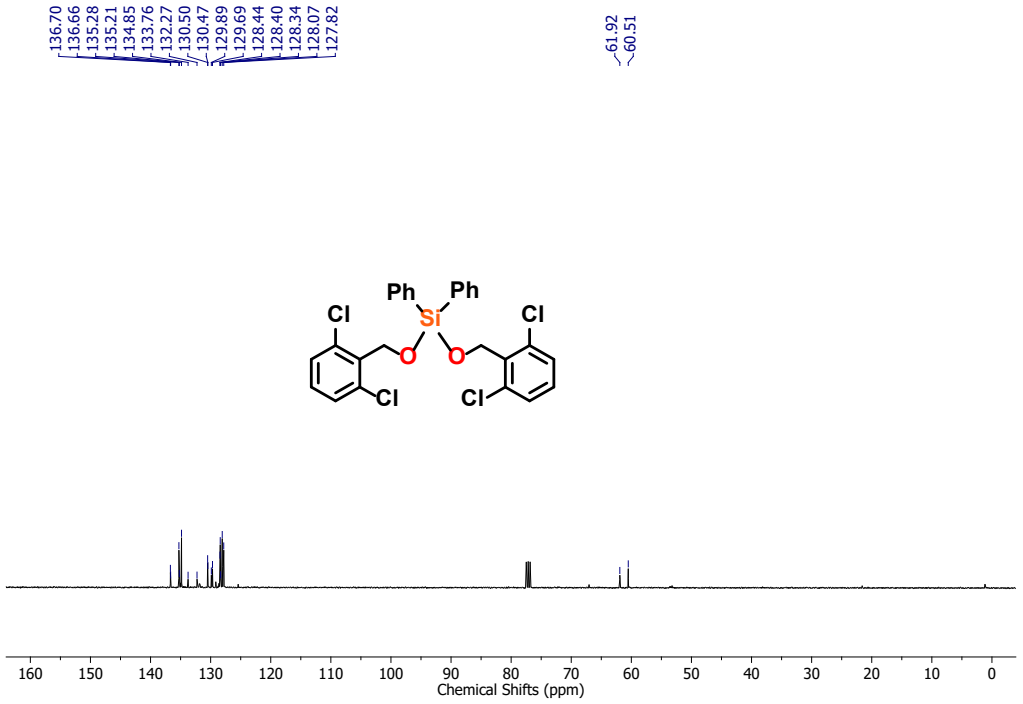


Figure S32: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3l**.

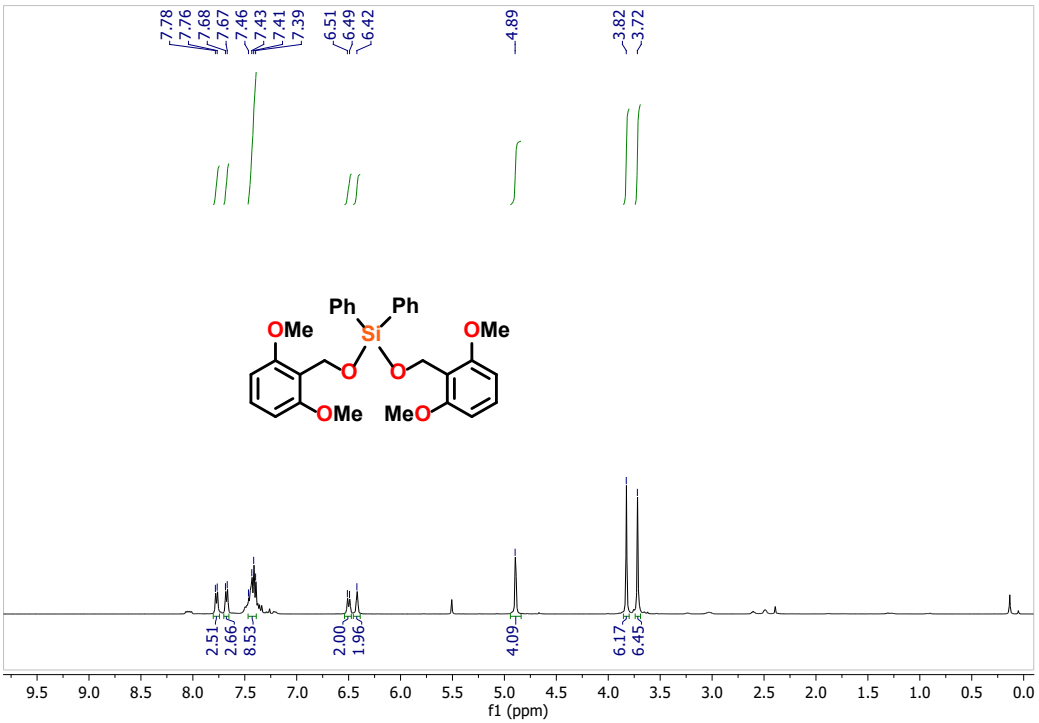


Figure S33: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3m**.

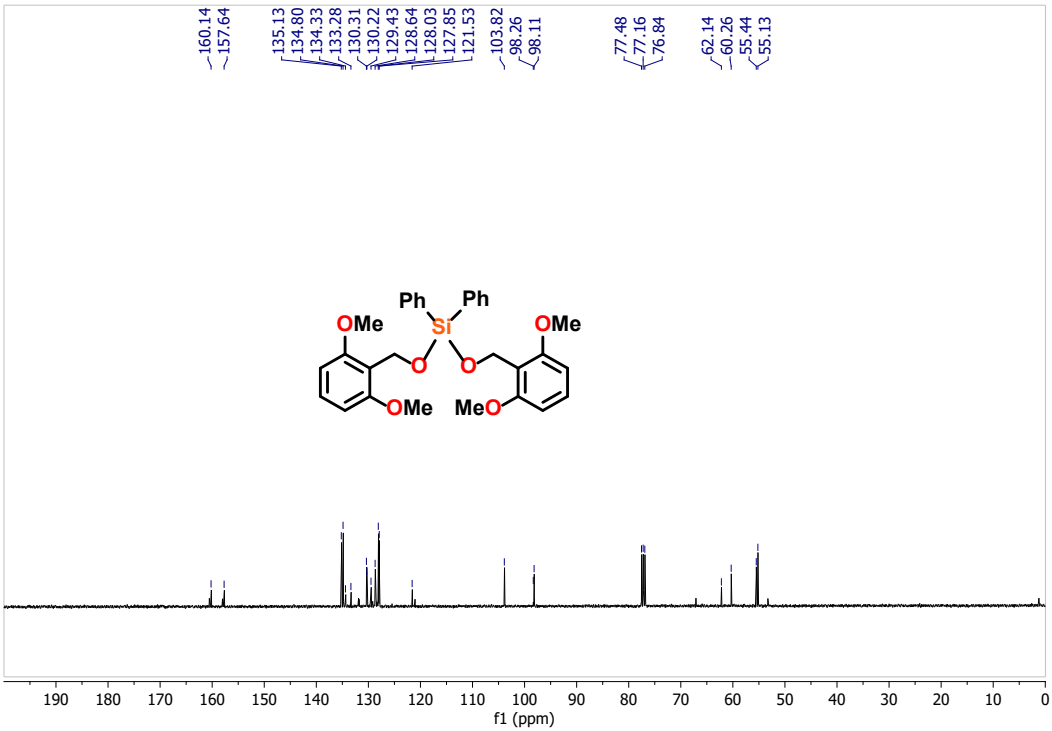


Figure S34: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3m**.

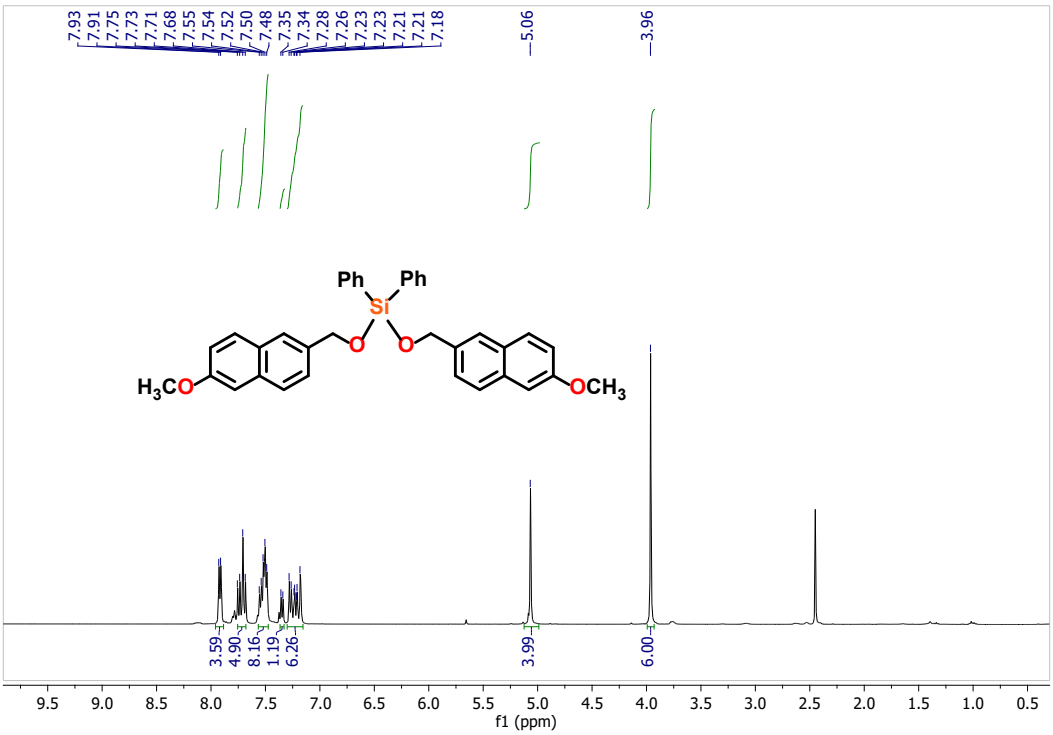


Figure S35: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3n**.

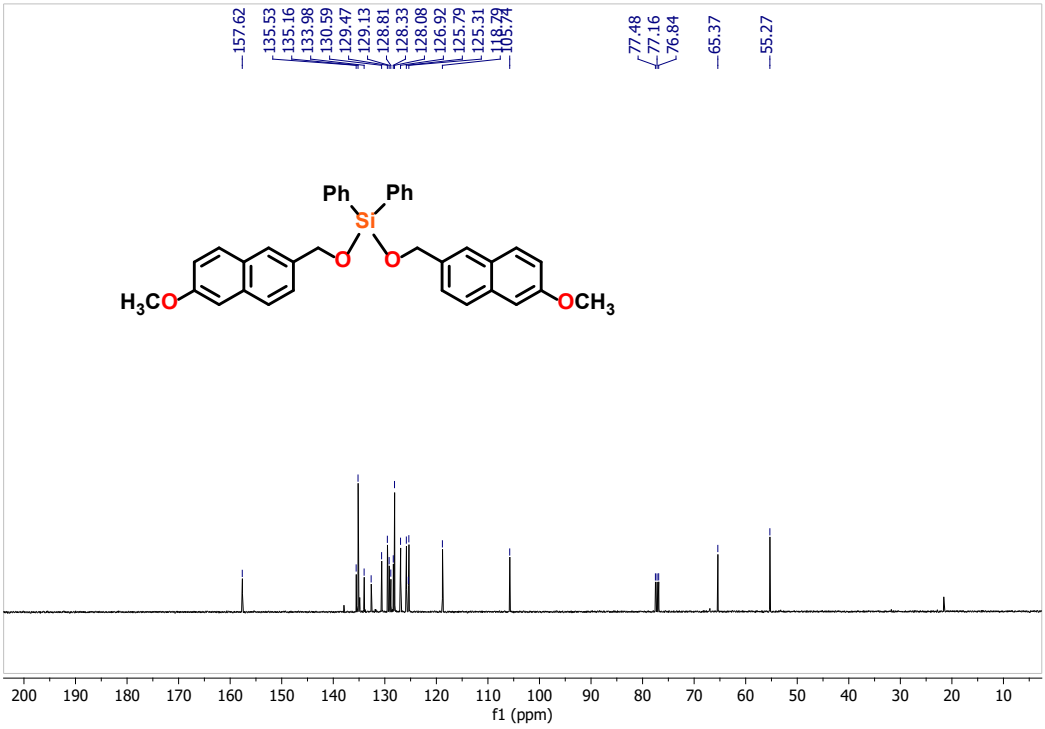


Figure S36: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3n**.

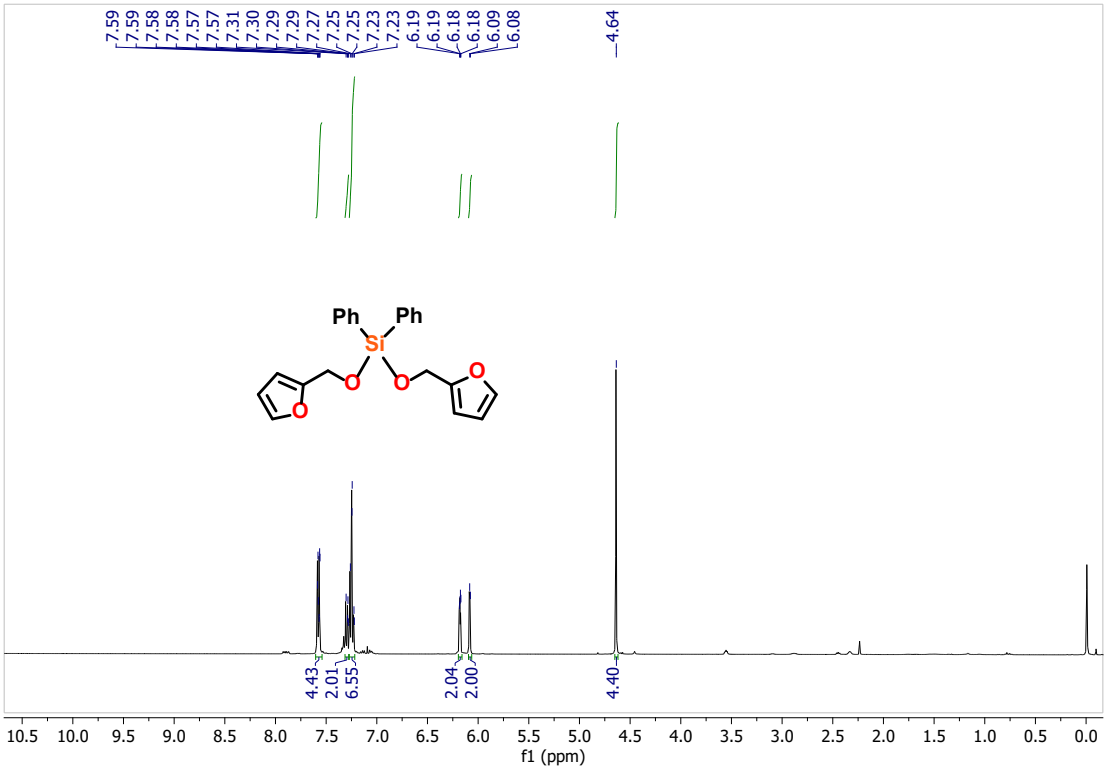


Figure S37: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3o**.

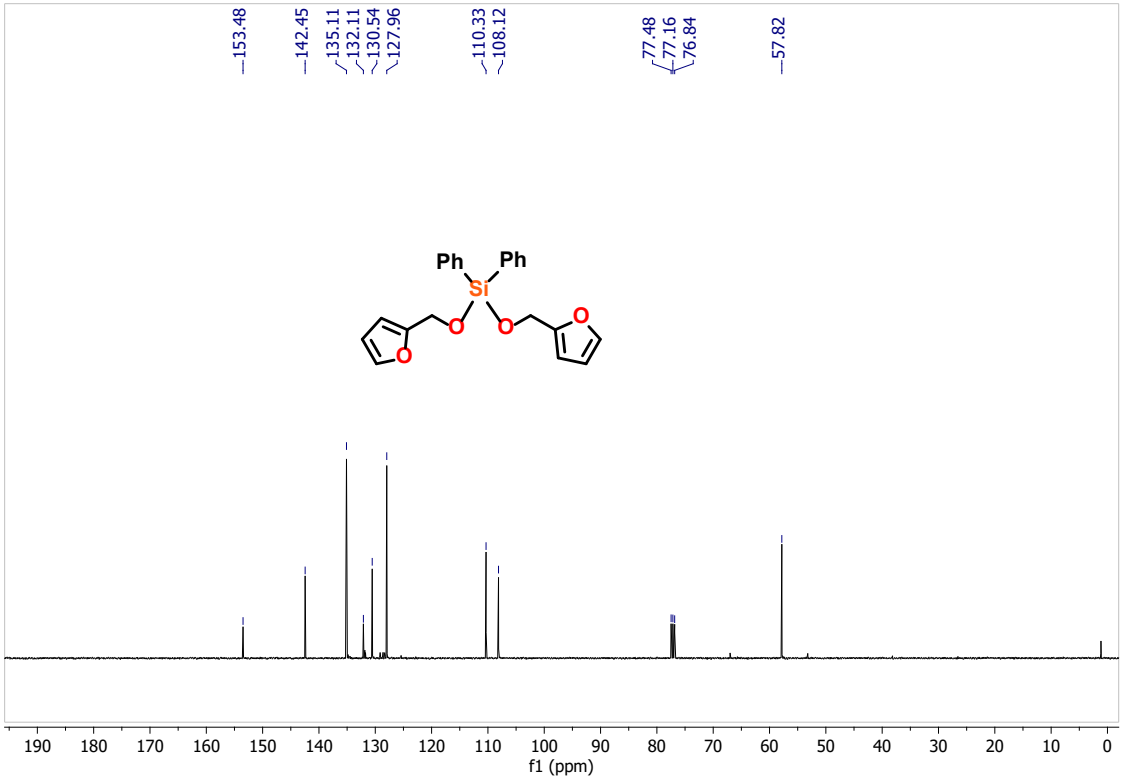


Figure S38: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3o**.

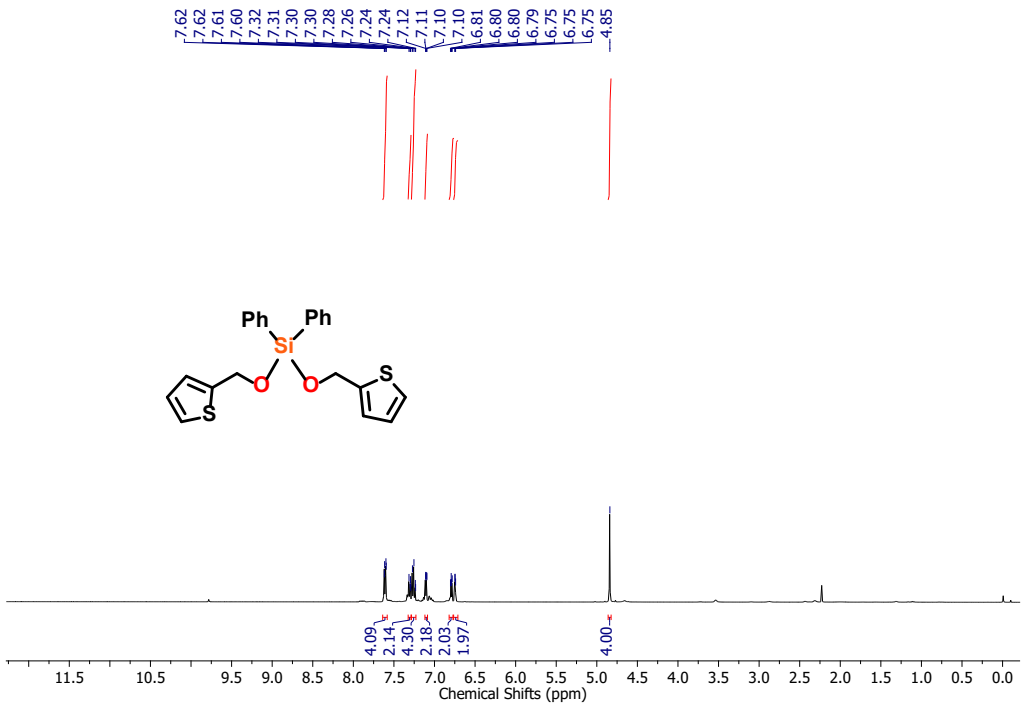


Figure S39: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3p**.

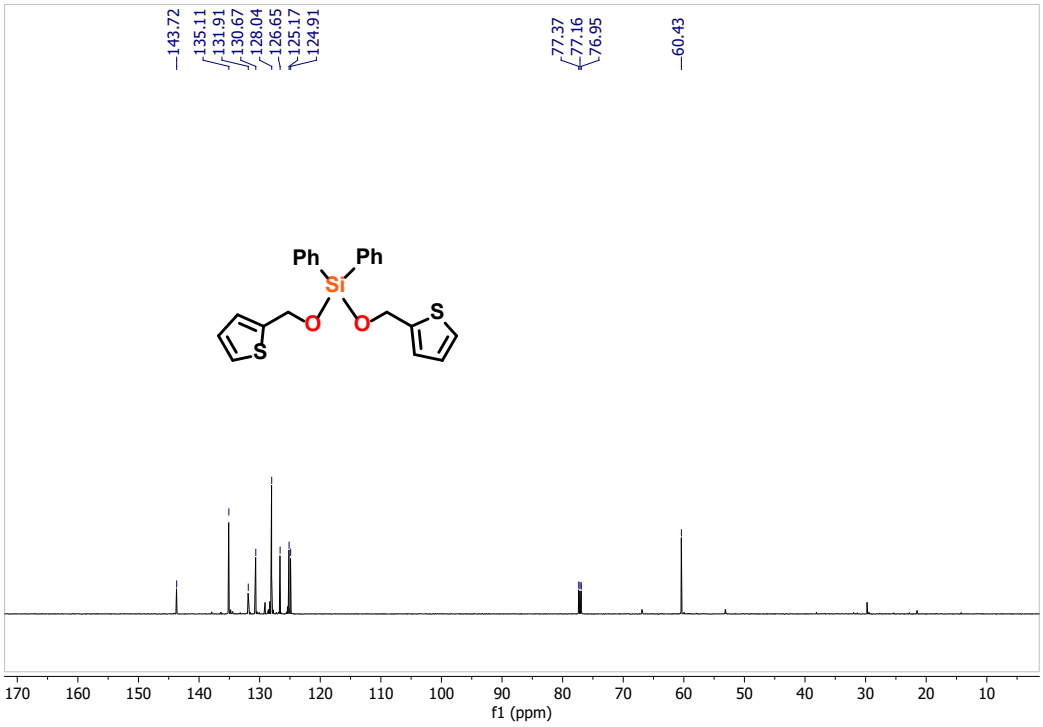


Figure S40: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3p**.

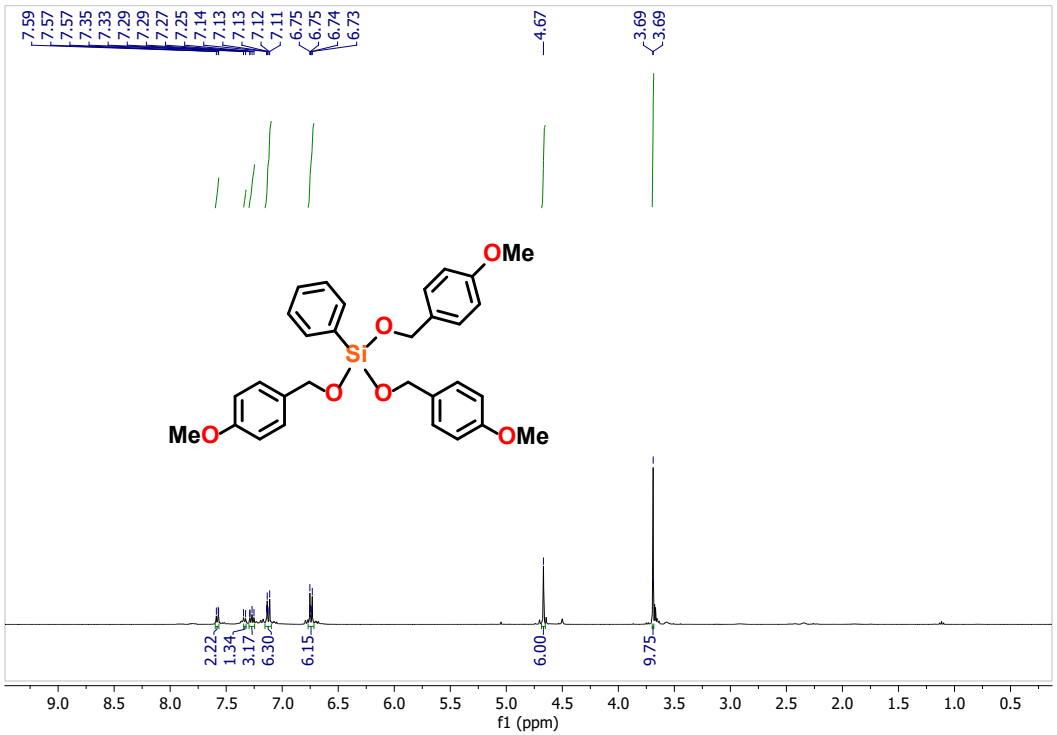


Figure S41: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **3q**.

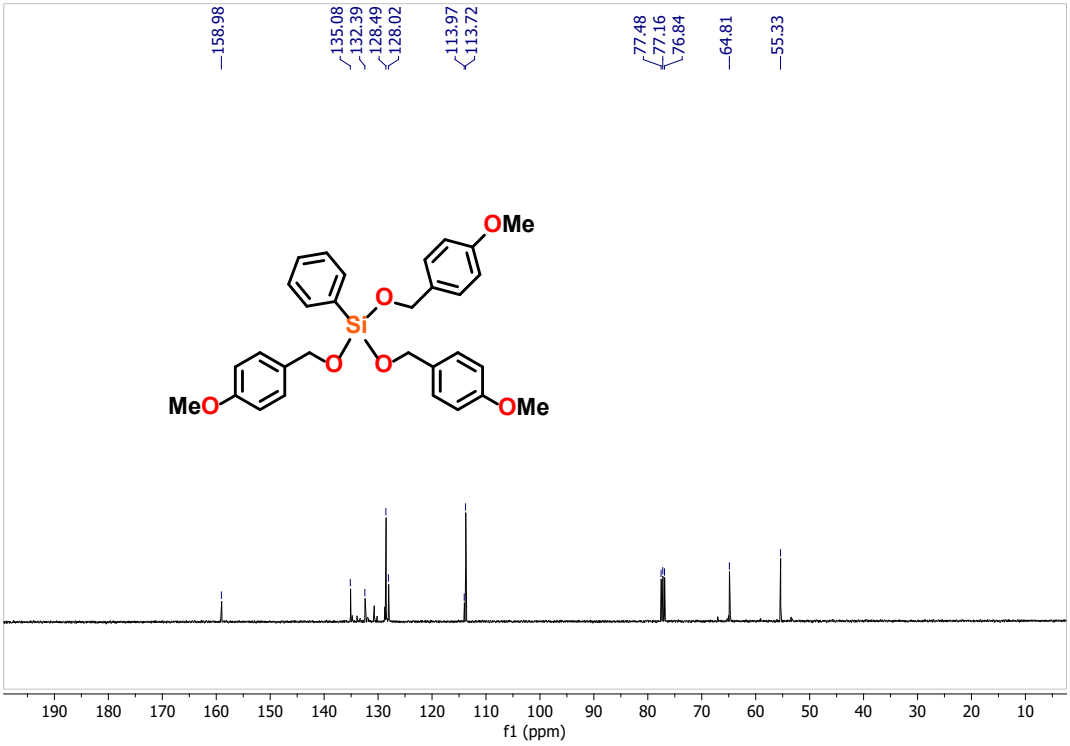
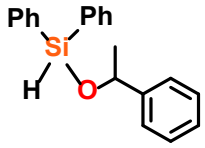


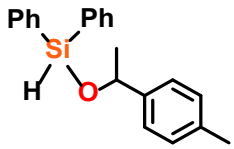
Figure S42: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **3q**.

6. NMR data for the hydrosilylation of ketones.



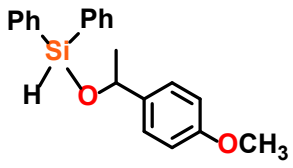
diphenyl(1-phenylethoxy)silane (4a).[5]

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 19:1 (R_f = 0.5). Isolated Yield: 73 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.73 – 7.62 (m, 4H, Ar*H*), 7.53 – 7.35 (m, 10H, Ar*H*), 7.33 – 7.29 (m, 1H, Ar*H*), 5.49 (s, 1H, Si-*H*), 5.07 (q, *J* = 6.4 Hz, 1H, C*H*), 1.58 (d, *J* = 6.4 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 145.4 (C_{Ar}), 134.8 (d, *J* = 4.0 Hz, C_{Ar}), 134.3 (C_{Ar}), 134.1 (C_{Ar}), 130.4 (d, *J* = 6.5 Hz, C_{Ar}), 128.3 (C_{Ar}), 128.1 (d, *J* = 6.8 Hz, C_{Ar}), 127.2 (C_{Ar}), 125.7 (C_{Ar}), 72.8 (*C*H), 26.4 (*C*H_3) ppm.



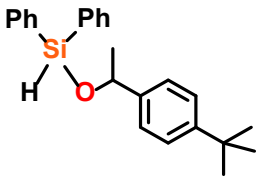
diphenyl(1-(p-tolyl)ethoxy)silane (4b).[5]

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 19:1 (R_f = 0.4). Isolated Yield: 77 mg (97 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.51 (dd, *J* = 20.2, 1.5 Hz, 4H, Ar*H*), 7.35 – 7.25 (m, 6H, Ar*H*), 7.14 (d, *J* = 5.3 Hz, 2H, Ar*H*), 7.04 (d, *J* = 5.1 Hz, 2H, Ar*H*), 5.32 (s, 1H, Si*H*), 4.89 (s, 1H, C*H*), 2.25 (s, 3H, CH_3), 1.41 (s, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 142.4 (C_{Ar}), 136.8 (C_{Ar}), 134.8 (d, *J* = 3.8 Hz, C_{Ar}), 134.4 (C_{Ar}), 130.4 (d, *J* = 7.4 Hz, C_{Ar}), 129.0 (C_{Ar}), 128.1 (d, *J* = 7.1 Hz, C_{Ar}), 125.6 (C_{Ar}), 72.7 (*C*H), 26.4 (*C*H_3), 21.3 (*C*H_3) ppm.



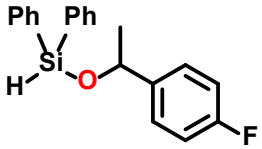
(1-(4-methoxyphenyl)ethoxy)diphenylsilane (4c).[5]

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.5). Isolated Yield: 81 mg (97 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.49 (d, J = 20.6 Hz, 4H, Ar*H*), 7.26 (dd, J = 12.2, 4.2 Hz, 6H, Ar*H*), 7.14 (s, 2H, Ar*H*), 6.73 (s, 2H, Ar*H*), 5.29 (s, 1H, Si*H*), 4.86 (s, 1H, C*H*), 3.68 (s, 3H, OCH_3), 1.39 (s, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 158.9 (C_{Ar}), 137.5 (C_{Ar}), 134.8 (d, J = 4.0 Hz, C_{Ar}), 134.4 (C_{Ar}), 130.4 (d, J = 7.3 Hz, C_{Ar}), 128.1 (d, J = 7.1 Hz, C_{Ar}), 126.9 (C_{Ar}), 113.7 (C_{Ar}), 72.5 (*C*H), 55.3 (O*C*H$_3$), 26.3 (*C*H$_3$) ppm.



(1-(4-(tert-butyl)phenyl)ethoxy)diphenylsilane (4d).[5]

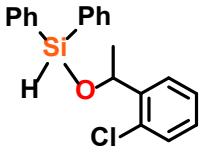
Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.4). Isolated Yield: 86 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.51 (dd, J = 21.6, 6.8 Hz, 4H, Ar*H*), 7.28 (dd, J = 17.3, 7.2 Hz, 6H, Ar*H*), 7.15 (d, J = 8.3 Hz, 2H, Ar*H*), 6.99 (d, J = 6.8 Hz, 2H, Ar*H*), 5.33 (s, 1H, Si*H*), 4.93 – 4.86 (m, 1H, C*H*), 1.42 (d, J = 6.3 Hz, 3H, CH_3), 0.82 (d, J = 6.6 Hz, 9H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 142.6 (C_{Ar}), 140.7 (C_{Ar}), 135.22 (C_{Ar}), 134.8 (d, J = 3.2 Hz, C_{Ar}), 130.4 (d, J = 6.9 Hz, C_{Ar}), 129.1 (C_{Ar}), 128.1 (d, J = 7.2 Hz, C_{Ar}), 127.7 (C_{Ar}), 125.5 (C_{Ar}), 72.7 (*C*H), 45.3 (C_{tBu}), 30.4 (*C*H$_3$), 26.3 (*C*H$_3$), 22.5 (*C*H$_3$) ppm.



(1-(4-fluorophenyl)ethoxy)diphenylsilane (4e).[5]

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.45). Isolated Yield: 76 mg (95 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.54 – 7.52 (m, 2H, Ar*H*), 7.48 – 7.46 (m, 2H, Ar*H*), 7.37 – 7.29 (m, 4H, Ar*H*), 7.28 – 7.12 (m, 4H, Ar*H*), 6.91 – 6.86 (m, 2H, Ar*H*), 5.32 (s, 1H, Si*H*), 4.91 – 4.86 (d, J = 6.67 Hz, 1H, C*H*), 1.41-1.39 (d, J = 8 Hz, 3H, CH_3), ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 163.3

(C_{Ar}), 160.8 (C_{Ar}), 141.1 (C_{Ar}), 141.1 (C_{Ar}), 134.8 (C_{Ar}), 134.8 (C_{Ar}), 134.4 (C_{Ar}), 134.2 (C_{Ar}), 134.0 (C_{Ar}), 130.5 (C_{Ar}), 130.5 (C_{Ar}), 128.2 (C_{Ar}), 128.1 (C_{Ar}), 128.1 (C_{Ar}), 127.3 (C_{Ar}), 127.3 (C_{Ar}), 115.2(C_{Ar}), 115.0 (C_{Ar}), 72.2 (CH), 26.4 (CH$_3$).



(1-(2-chlorophenyl)ethoxy)diphenylsilane (4f).[6]

Following the procedure, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.7). Isolated Yield: 81 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, CDCl$_3$) δ_H 7.77 (dd, J = 8.3, 1.4 Hz, 1H, ArH), 7.72 – 7.60 (m, 4H, ArH), 7.43 (dddd, J = 14.4, 9.5, 7.6, 3.5 Hz, 6H, ArH), 7.31 (d, J = 7.6 Hz, 2H, ArH), 7.22 (dd, J = 7.6, 1.2 Hz, 1H, ArH), 5.50 (s, 1H, SiH), 5.46 (q, J = 6.3 Hz, 1H, CH), 1.54 (d, J = 6.3 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, CDCl$_3$) δ_C 143.0 (C_{Ar}), 135.1 (C_{Ar}), 134.8 (d, J = 6.5 Hz, C_{Ar}), 134.1 (C_{Ar}), 133.8 (C_{Ar}), 131.2 (C_{Ar}), 130.5 (d, J = 6.9 Hz, C_{Ar}), 129.2 (C_{Ar}), 128.4 – 127.8 (C_{Ar}), 127.2 (d, J = 7.3 Hz, C_{Ar}), 69.5 (CH), 24.9 (CH$_3$) ppm.

7. ^{1}H and ^{13}C{^{1}H} NMR Spectra for the mono-hydrosilylated ketones:

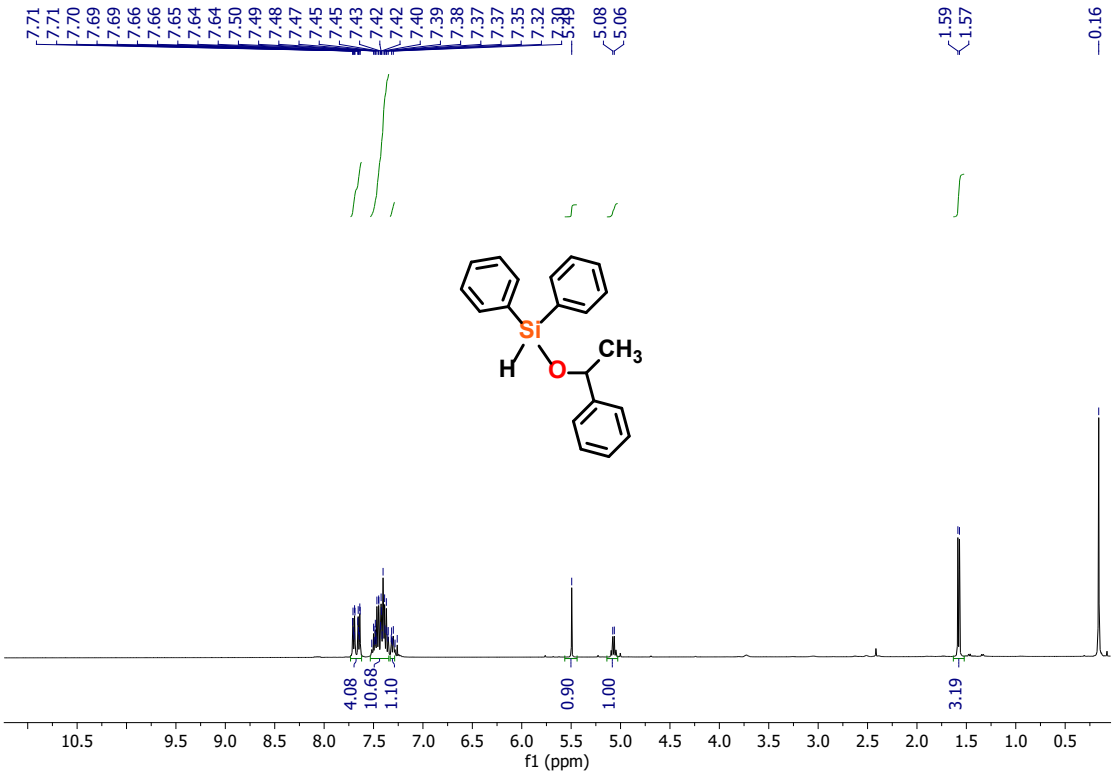


Figure FS43: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **4a**.

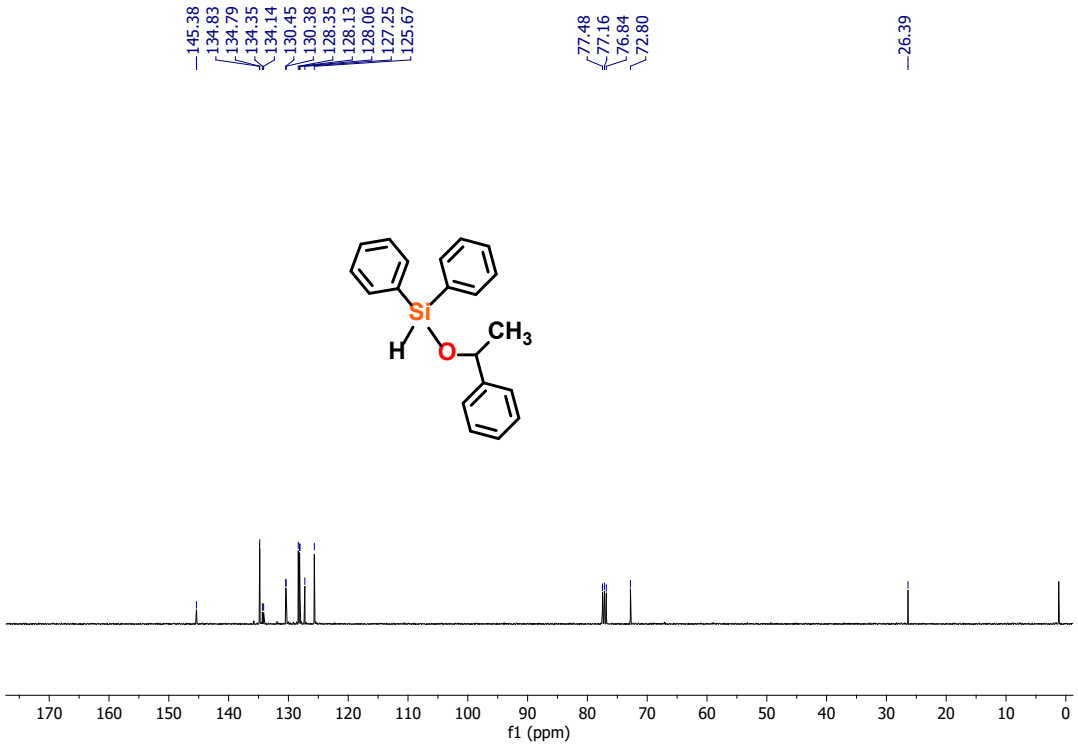


Figure FS44: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **4a**.

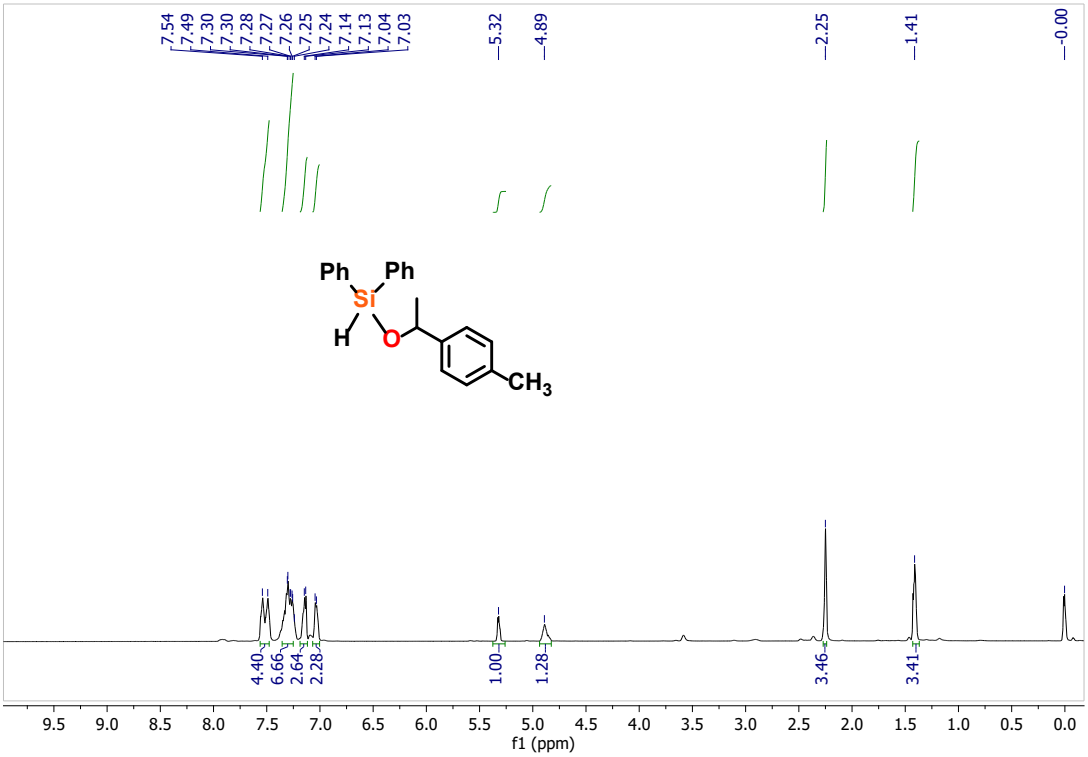


Figure S45: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **4b**.

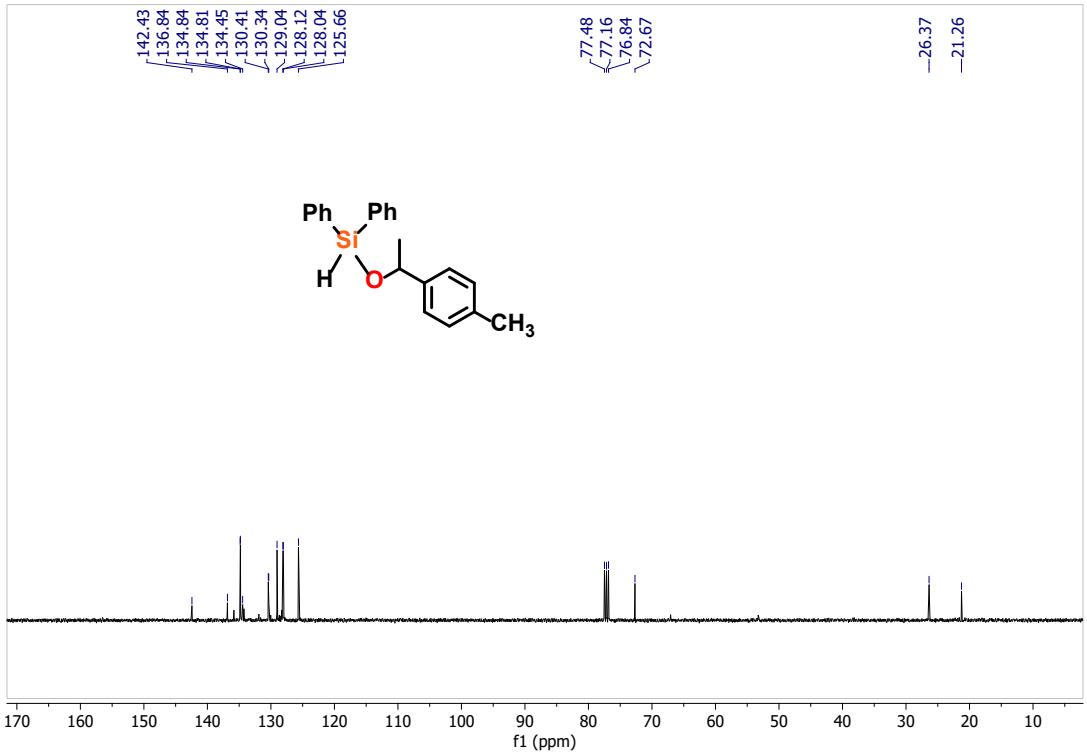


Figure S46: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **4b**.

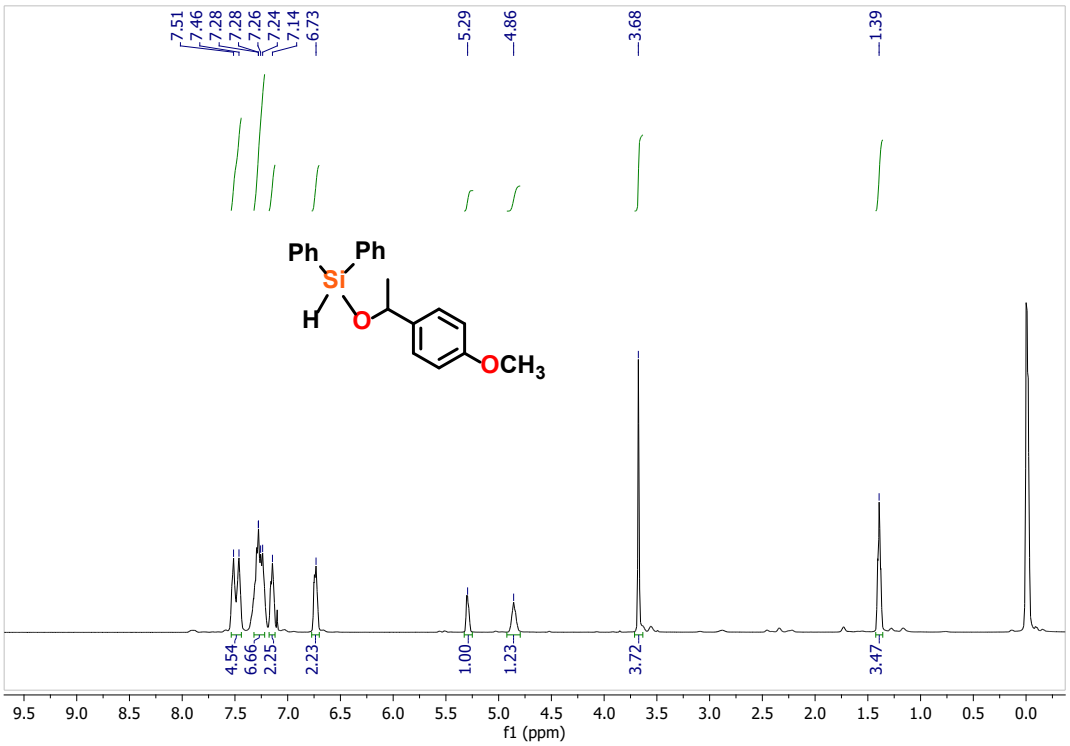


Figure S47: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **4c**.

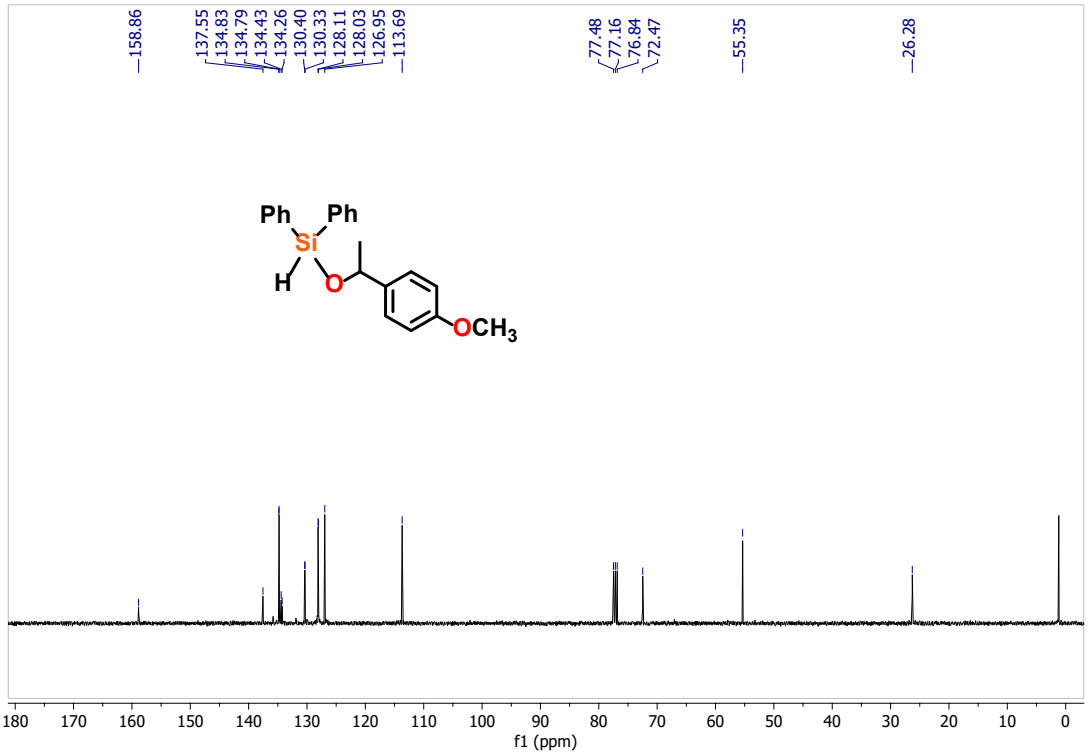


Figure S48: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **4c**.

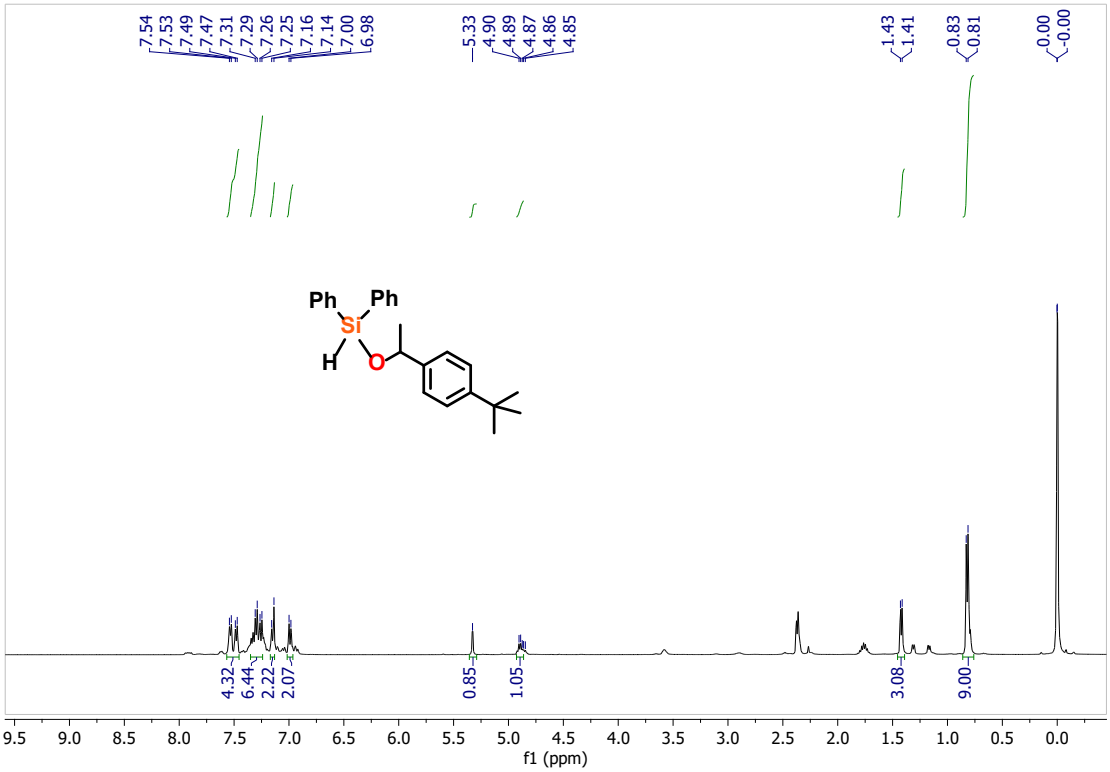


Figure S49: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **4d**.

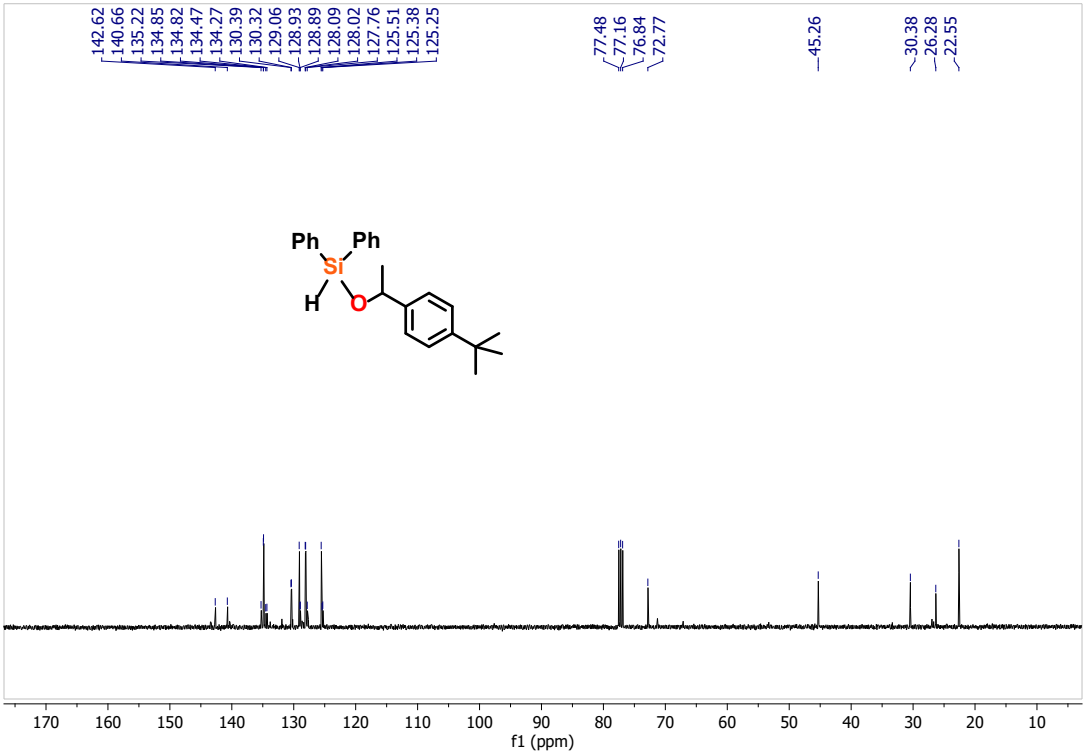


Figure S50: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **4d**.

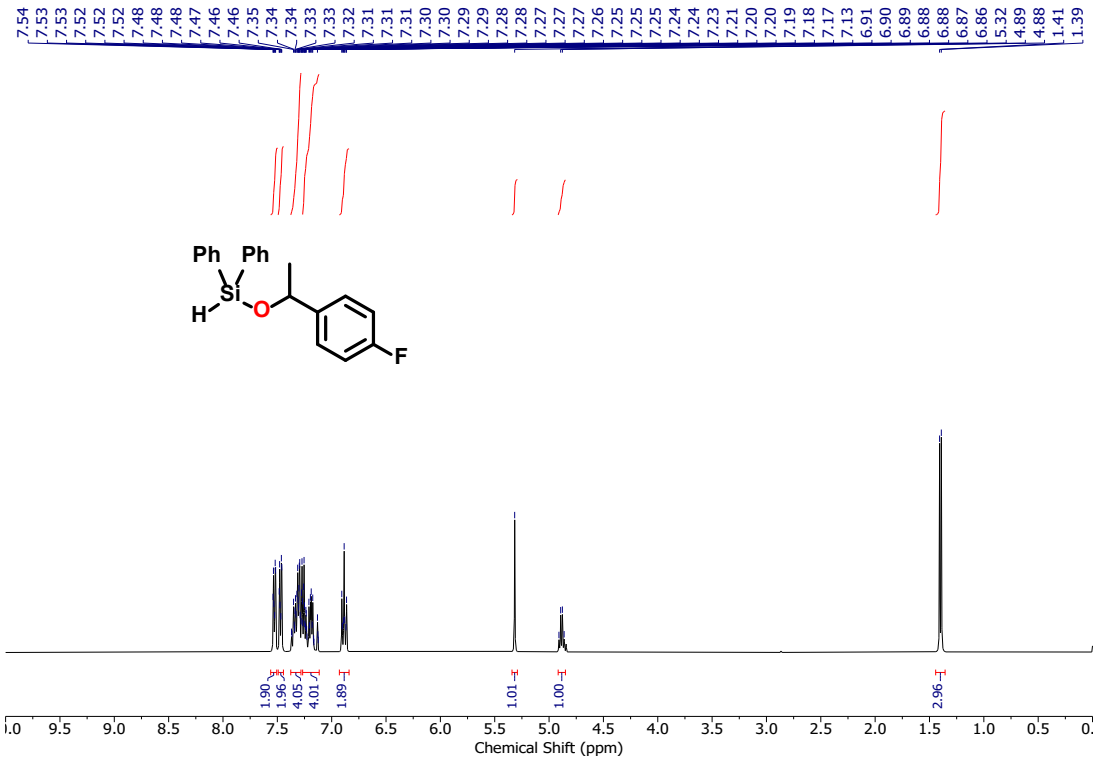


Figure S51: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **4e.**

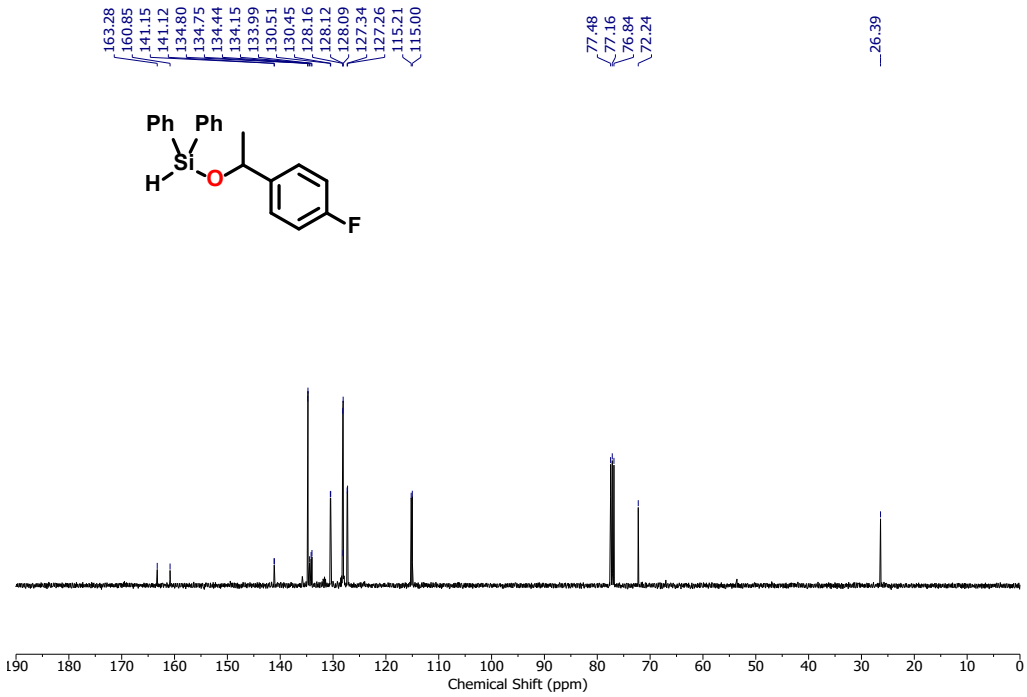


Figure S52: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **4e.**

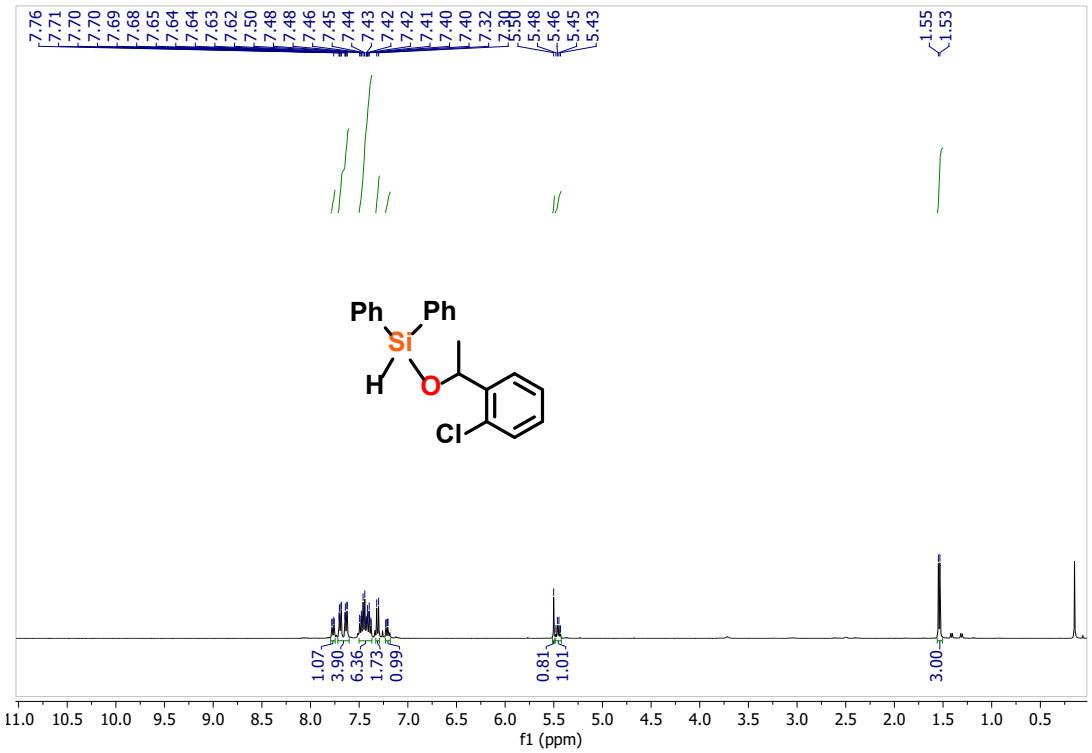


Figure S53: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **4f**.

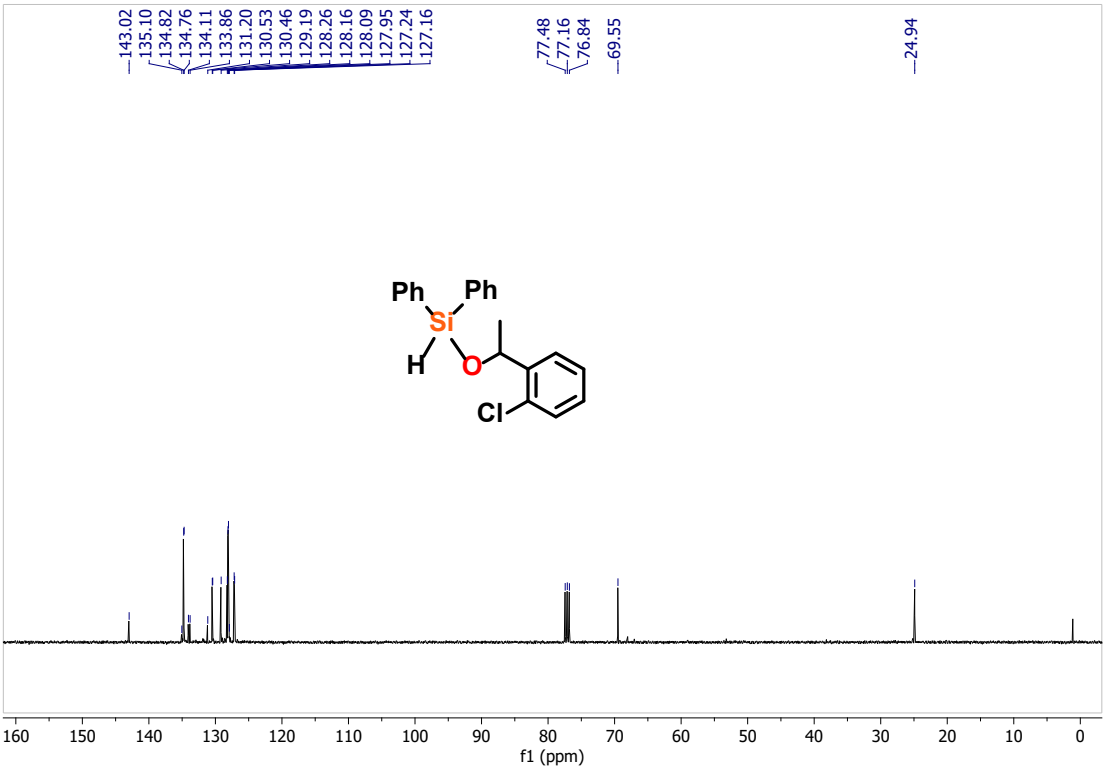
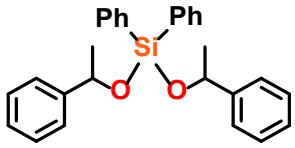


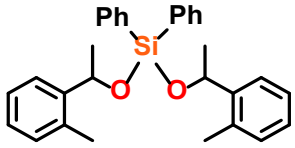
Figure S54: ^{13}C{^{1}H} NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **4f**.

8. NMR data for the hydrosilylation of ketones.



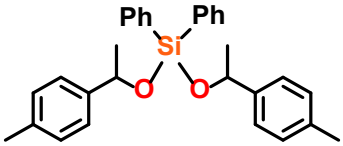
diphenylbis(1-phenylethoxy)silane (5a).

Following the procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.6). Isolated Yield: 100 mg (95 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.64 – 7.44 (m, 4H, Ar*H*), 7.35 – 7.11 (m, 16H, Ar*H*), 4.92-4.84 (m, 2H, C*H*), 1.33-1.31 (d, J = 8 Hz, 3H, CH_3), 1.19-1.18 (d, J = 4 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 146.1 (C_{Ar}), 145.9 (C_{Ar}), 135.2 (C_{Ar}), 135.2 (C_{Ar}), 135.2(C_{Ar}), 133.8 (C_{Ar}), 133.6 (C_{Ar}), 133.1 (C_{Ar}), 130.3 (C_{Ar}), 130.3 (C_{Ar}), 130.2, 128.2 (C_{Ar}), 128.2 (C_{Ar}), 127.9 (C_{Ar}), 127.8 (C_{Ar}), 127.7 (C_{Ar}), 127.0 (C_{Ar}), 126.9 (C_{Ar}), 125.6 (C_{Ar}), 125.4 (C_{Ar}), 71.4 (*C*H), 71.4 (*C*H), 26.9 (*C*H$_3$), 26.7 (*C*H$_3$) ppm.



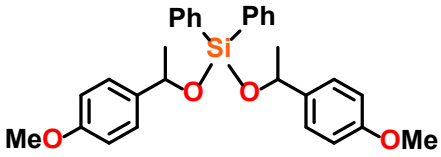
diphenylbis(1-(o-tolyl)ethoxy)silane (5b).

Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.5). Isolated Yield: 108 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.69 (dd, J = 6.7, 0.9 Hz, 1H, Ar*H*), 7.63 (dd, J = 6.7, 0.8 Hz, 2H, Ar*H*), 7.56 – 7.51 (m, 1H, Ar*H*), 7.35 (ddd, J = 15.6, 10.7, 4.5 Hz, 6H, Ar*H*), 7.22 – 7.17 (m, 2H, Ar*H*), 7.12 – 7.07 (m, 4H, Ar*H*), 7.04 (t, J = 8.2 Hz, 2H, Ar*H*), 4.98 – 4.88 (m, 2H, Ar*H*), 2.31 (s, 6H, Ar*H*), 1.37 (d, J = 6.4 Hz, 3H, Ar*H*), 1.26 (d, J = 6.3 Hz, 3H, Ar*H*) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 143.1 (d, J = 12.4 Hz, C_{Ar}), 136.4 (d, J = 12.2 Hz, C_{Ar}), 135.2 (t, J = 2.7 Hz, C_{Ar}), 134.8 (d, J = 3.9 Hz, C_{Ar}), 133.7 (C_{Ar}), 130.4 (d, J = 7.3 Hz, C_{Ar}), 130.2 (C_{Ar}), 129.0 (C_{Ar}), 128.9 (d, J = 5.9 Hz, C_{Ar}), 128.1 (d, J = 7.1 Hz, C_{Ar}), 127.8 (t, J = 8.4 Hz, C_{Ar}), 125.7 (C_{Ar}), 125.4 (d, J = 11.7 Hz, C_{Ar}), 71.2 (d, J = 2.3 Hz, C_{Ar}), 27.0 (C_{Ar}), 21.2 (C_{Ar}) ppm.



diphenylbis(1-(p-tolyl)ethoxy)silane (5c).

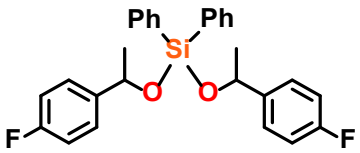
Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.4). Isolated Yield: 108 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.71 (dd, J = 7.9, 1.4 Hz, 1H, Ar*H*), 7.65 (dd, J = 7.9, 1.4 Hz, 2H, Ar*H*), 7.55 (dd, J = 8.0, 1.3 Hz, 1H, Ar*H*), 7.44 – 7.31 (m, 5H, Ar*H*), 7.29 – 7.25 (m, 2H, Ar*H*), 7.20 (s, 2H, Ar*H*), 7.13 – 7.03 (m, 5H, Ar*H*), 5.01 – 4.88 (m, 2H, C*H*), 2.34 (s, 6H, CH_3), 1.39 (d, J = 6.4 Hz, 3H, CH_3), 1.27 (d, J = 6.4 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 143.1 (d, J = 12.2 Hz, C_{Ar}), 136.4 (d, J = 12.2 Hz, C_{Ar}), 135.2 (d, J = 3.3 Hz, C_{Ar}), 133.7 (C_{Ar}), 130.2 (C_{Ar}), 129.2 (C_{Ar}), 128.9 (d, J = 5.9 Hz, C_{Ar}), 128.4 (C_{Ar}), 127.8 (C_{Ar}), 125.4 (d, J = 11.7 Hz, C_{Ar}), 71.2 (*C*H), 26.9 (*C*H$_3$), 26.7 (*C*H$_3$), 21.2 (*C*H$_3$) ppm.



bis(1-(4-methoxyphenyl)ethoxy)diphenylsilane (5d).

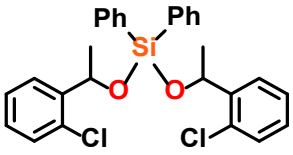
Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.7). Isolated Yield: 117 mg (97 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.85 (d, J = 8.7 Hz, 1H, Ar*H*), 7.54 (d, J = 7.0 Hz, 2H, Ar*H*), 7.30 (d, J = 8.0 Hz, 2H, Ar*H*), 7.24 (dd, J = 14.1, 6.9 Hz, 3H, Ar*H*), 7.13 (d, J = 8.5 Hz, 2H, Ar*H*), 7.04 (d, J = 8.4 Hz, 2H, Ar*H*), 6.84 (d, J = 8.7 Hz, 2H, Ar*H*), 6.70 (dd, J = 18.0, 8.4 Hz, 4H, Ar*H*), 4.83 (dd, J = 13.8, 6.5 Hz, 2H, C*H*), 3.76 (s, 3H, OCH_3), 3.69 (s, 3H, OCH_3), 1.29 (d, J = 6.3 Hz, 3H, CH_3), 1.18 (d, J = 6.3 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 158.6 ($C_{C=O}$), 158.5 ($C_{C=O}$), 138.3 (C_{Ar}), 138.2 (C_{Ar}), 135.2 (C_{Ar}), 135.1 (C_{Ar}), 133.7 (C_{Ar}), 130.7 (C_{Ar}), 130.2 (C_{Ar}), 130.1

(C_{Ar}), 128.6 (C_{Ar}), 127.8 (C_{Ar}), 127.8 (C_{Ar}), 127.6 (C_{Ar}), 126.7 (C_{Ar}), 126.5 (C_{Ar}), 113.7 (C_{Ar}), 113.5 (C_{Ar}), 70.9 (*C*H), 70.8 (*C*H), 55.5 (O*C*H$_3$), 55.3 (O*C*H$_3$), 26.9 (*C*H$_3$), 26.6 (*C*H$_3$) ppm.



bis(1-(4-fluorophenyl)ethoxy)diphenylsilane (5e).

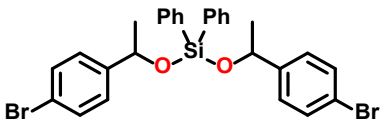
Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.45). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.58 (dd, *J* = 8.0, 1.4 Hz, 1H, Ar*H*), 7.54 (dd, *J* = 8.0, 1.3 Hz, 2H, Ar*H*), 7.41 (dd, *J* = 8.0, 1.3 Hz, 1H, Ar*H*), 7.35 – 7.31 (m, 2H, Ar*H*), 7.30 – 7.23 (m, 4H, Ar*H*), 7.18 – 7.13 (m, 3H, Ar*H*), 7.06 – 7.02 (m, 2H, Ar*H*), 6.89 – 6.84 (m, 2H, Ar*H*), 6.82 – 6.77 (m, 2H, Ar*H*), 4.83 (dd, *J* = 27.0, 6.4 Hz, 2H, C*H*), 1.27 (d, *J* = 6.4 Hz, 3H, C*H*$_3$), 1.17 (d, *J* = 6.4 Hz, 3H, C*H*$_3$) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 162.8 (d, *J* = 12.4 Hz, C_{Ar}), 161.1 (d, *J* = 12.0 Hz, C_{Ar}), 141.8 – 141.6 (C_{Ar}), 135.1 (t, *J* = 7.5 Hz, C_{Ar}), 134.8 (d, *J* = 7.5 Hz, C_{Ar}), 133.4 (C_{Ar}), 133.2 (C_{Ar}), 132.8 (C_{Ar}), 130.4 (d, *J* = 17.1 Hz, C_{Ar}), 129.2 (C_{Ar}), 128.4 (C_{Ar}), 128.0 (dd, *J* = 27.9, 8.6 Hz, C_{Ar}), 127.8 (C_{Ar}), 127.1 (d, *J* = 7.7 Hz, C_{Ar}), 26.9 (C_{Ar}), 26.7 (C_{Ar}) ppm.



bis(1-(2-chlorophenyl)ethoxy)diphenylsilane (5f).

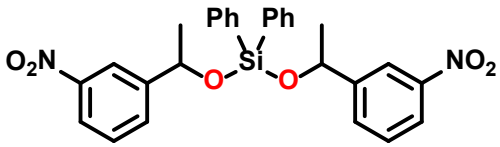
Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.5). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.69 – 7.62 (m, 2H, Ar*H*), 7.61 – 7.58 (m, 2H, Ar*H*), 7.46 – 7.42 (m, 1H, Ar*H*), 7.35 – 7.24 (m, 6H, Ar*H*), 7.18 – 7.09 (m, 4H, Ar*H*), 7.07 – 7.03 (m, 1H, Ar*H*), 7.01 – 6.93 (m, 2H, Ar*H*), 5.27 (dq, *J* = 33.6, 6.2 Hz, 2H, C*H*), 1.27 (d, *J* = 6.3 Hz, 3H, C*H*$_3$), 1.16 (d, *J* = 6.3 Hz, 3H, C*H*$_3$) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 143.4 (d, *J* = 29.7 Hz, C_{Ar}), 135.1 (C_{Ar}), 134.9 (C_{Ar}), 132.9 (C_{Ar}), 130.7 (C_{Ar}),

130.5 (C_{Ar}), 130.3 (C_{Ar}), 129.0 (d, J = 8.6 Hz, C_{Ar}), 128.1 (C_{Ar}), 127.9 (C_{Ar}), 127.8 (C_{Ar}), 127.2 (C_{Ar}), 127.1 (C_{Ar}), 126.7 (C_{Ar}), 68.0 (CH), 25.1 (CH$_3$) ppm.



bis(1-(4-bromophenyl)ethoxy)diphenylsilane (5g)

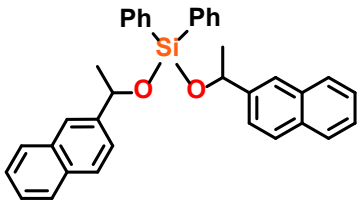
Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.6). Isolated Yield: 137 mg (95 %). ^{1}H NMR (400 MHz, 25 °C, CDCl$_3$) δ_H 7.59 – 7.54 (m, 3H, ArH), 7.44 – 7.37 (m, 1H, ArH), 7.36 – 7.17 (m, 10H, ArH), 7.08 – 7.06 (m, 2H, ArH), 6.95 – 6.93 (m, 2H, ArH), 4.86 – 4.81 (q, J = 6.67 Hz, 1H, CH), 4.78 – 4.73 (m, J = 6.67 Hz, 1H, CH), 1.26-1.24 (d, J = 8 Hz, 3H, CH_3), 1.18 –1.16 (d, J = 8 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, CDCl$_3$) δ_C 145.0 (C_{Ar}), 144.8 (C_{Ar}), 135.1 (C_{Ar}), 135.1(C_{Ar}), 135.0 (C_{Ar}), 133.0 (C_{Ar}), 131.4 (C_{Ar}), 131.2 (C_{Ar}), 130.5 (C_{Ar}), 130.4 (C_{Ar}), 128.0 (C_{Ar}), 127.9 (C_{Ar}), 127.3 (C_{Ar}), 127.0 (C_{Ar}), 120.8 (C_{Ar}), 120.7 (C_{Ar}), 70.9 (CH), 70.7 (CH), 26.9 (CH$_3$), 26.6 (CH$_3$) ppm.



bis(1-(3-nitrophenyl)ethoxy)diphenylsilane (5h)

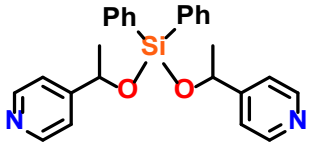
Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 7:3 (R_f = 0.8). Isolated Yield: 118 mg (92 %). ^{1}H NMR (400 MHz, 25 °C, CDCl$_3$) δ_H 8.08 – 8.07 (m, 1H, ArH), 8.00 – 7.97 (m, 1H, ArH), 7.90 – 7.89 (m, 1H, ArH), 7.85 – 7.83 (m, 1H, ArH), 7.62 – 7.57 (m, 3H, ArH), 7.53 – 7.50 (m, 1H, ArH), 7.45 – 7.42 (m, 10H, ArH), 7.39 – 7.34 (m, 3H, ArH), 7.33-7.29 (m, 4H, ArH), 7.24-7.17 (m, 2H, ArH), 5.00 – 4.96 (q, J = 5.33 Hz, 1H, CH), 4.94 – 4.89 (m, J = 6.67 Hz, 1H, CH), 1.32-1.31 (d, J = 4 Hz, 3H, CH_3), 1.25 –1.23 (d, J = 8 Hz, 6H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, CDCl$_3$) δ_C 148.1(C_{Ar}), 147.8 (C_{Ar}), 135.0 (C_{Ar}), 134.9 (C_{Ar}), 133.9 (C_{Ar}), 132.1 (C_{Ar}), 131.9 (C_{Ar}), 131.8 (C_{Ar}), 131.7 (C_{Ar}), 131.3 (C_{Ar}), 130.9 (C_{Ar}), 130.7 (C_{Ar}), 130.0 (C_{Ar}), 129.3 (C_{Ar}), 129.1 (C_{Ar}), 128.6 (C_{Ar}),

128.2 (C_{Ar}), 128.0 (C_{Ar}), 127.5 (C_{Ar}), 123.3 (C_{Ar}), 122.2 (C_{Ar}), 122.0 (C_{Ar}), 120.6 (C_{Ar}), 120.1 (C_{Ar}), 70.7(*C*H), 70.4 (*C*H), 26.8 (*C*H_3), 26.4 (*C*H_3).



bis(1-(naphthalen-2-yl)ethoxy)diphenylsilane (5i).

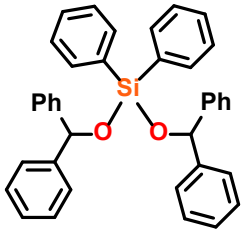
Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.4). Isolated Yield: 124 mg (95 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.67 – 7.61 (m, 6H, Ar*H*), 7.56 (s, 1H, Ar*H*), 7.52 – 7.39 (m, 4H, Ar*H*), 7.35 – 7.23 (m, 10H, Ar*H*), 7.15 – 7.01 (m, 2H, Ar*H*), 5.11 – 5.02 (m, 1H, C*H*), 4.98 (m, 1H, C*H*), 1.32 (d, J = 3.6 Hz, 3H, CH_3), 1.23 (s, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 143.3 (d, J = 17.4 Hz, C_{Ar}), 135.2 (d, J = 8.5 Hz, C_{Ar}), 133.6 (C_{Ar}), 133.3 (d, J = 11.0 Hz, C_{Ar}), 132.8 (d, J = 9.4 Hz, C_{Ar}), 130.3 (d, J = 12.7 Hz, C_{Ar}), 127.8 (C_{Ar}), 125.9 (d, J = 14.5 Hz, C_{Ar}), 125.6 (d, J = 10.5 Hz, C_{Ar}), 124.0 (C_{Ar}), 71.5 (d, J = 18.8 Hz, *C*H), 26.8 (*C*H_3), 26.6 (*C*H_3) ppm.



diphenylbis(1-(pyridin-4-yl)ethoxy)silane (5j).

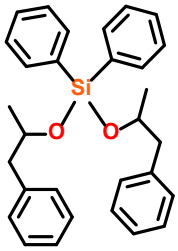
Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 7:3 (R_f = 0.9). Isolated Yield: 102 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 8.38 (dd, J = 31.6, 5.7 Hz, 4H, Ar*H*), 7.63 – 7.55 (m, 3H, Ar*H*), 7.49 – 7.43 (m, 1H, Ar*H*), 7.36 (dd, J = 14.6, 7.2 Hz, 3H, Ar*H*), 7.29 (dd, J = 12.5, 5.0 Hz, 3H, Ar*H*), 7.24 – 7.17 (m, 1H, Ar*H*), 7.13 (d, J = 5.8 Hz, 1H, Ar*H*), 7.00 (d, J = 5.7 Hz, 2H, Ar*H*), 4.84 (dd, J = 29.4, 6.4 Hz, 2H, C*H*), 1.29 (d, J = 6.5 Hz, 3H, CH_3), 1.18 (d, J = 6.4 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 154.3 (C_{Ar}), 149.8 (C_{Ar}), 134.9 (C_{Ar}), 132.1 (C_{Ar}), 131.8 (C_{Ar}), 130.7 (C_{Ar}),

128.6 (C_{Ar}), 128.4 (C_{Ar}) 127.8 (C_{Ar}), 120.4 (C_{Ar}), 120.2 (C_{Ar}), 70.1 (CH), 26.3 (CH$_3$), 26.1 (CH$_3$) ppm.



bis(benzhydryloxy)diphenylsilane (5k):

Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.5). Isolated Yield: 131 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, CDCl$_3$) δ_H 7.41 (d, J = 7.6 Hz, 4H, ArH), 7.23 (d, J = 7.3 Hz, 2H, ArH), 7.12 (t, J = 7.6 Hz, 4H, ArH), 7.05 (ddd, J = 17.7, 14.0, 7.2 Hz, 20H, ArH), 5.69 (s, 2H, CH) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, CDCl$_3$) δ_C 144.2 (C_{Ar}), 135.2 (C_{Ar}), 132.8 (C_{Ar}), 130.3 (C_{Ar}), 128.2 (C_{Ar}), 127.7 (C_{Ar}), 127.1 (C_{Ar}), 126.5 (C_{Ar}), 77.1 (CH) ppm.



Diphenylbis((1-phenylpropan-2-yl)oxy)silane **(5l):**

Following procedure **9**, the product was isolated with column chromatography using eluent hexane/ ethylacetate 9:1 (R_f = 0.54). Isolated Yield: 142 mg (92 %). ^{1}H NMR (400 MHz, 25 °C, CDCl$_3$) δ_H 7.50-7.48 (m, 1H, ArH), 7.43-7.38 (m, 3H, ArH), 7.31-7.26 (m, 2H, ArH), 7.24-7.18 (m, 5H, ArH), 6.90-6.87 (m, 4H, ArH), 6.67-6.65 (m, 4H, CH), 4.07-3.98 (m, 2H, ArH), 2.73-2.68 (m, 2H, CH_2), 2.55-2.51 (m, 2H, CH_2), 1.03-1.02 (d, J = 4 Hz, 3H, CH_3), 1.00-0.99 (d, J = 4 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, CDCl$_3$) δ_C 158.1 (C_{Ar}), 135.2 (C_{Ar}), 130.7 (C_{Ar}), 130.6 (C_{Ar}), 127.7 (C_{Ar}), 113.6 (C_{Ar}), 113.6 (C_{Ar}), 70.7 (CH), 70.6 (CH), 55.3 (CH$_3$), 55.3 (CH$_3$), 45.2 (CH$_2$), 45.2 (CH$_2$), 23.1 (CH$_3$), 23.0 (CH$_3$) ppm.

9. NMR Spectra for Ketones.

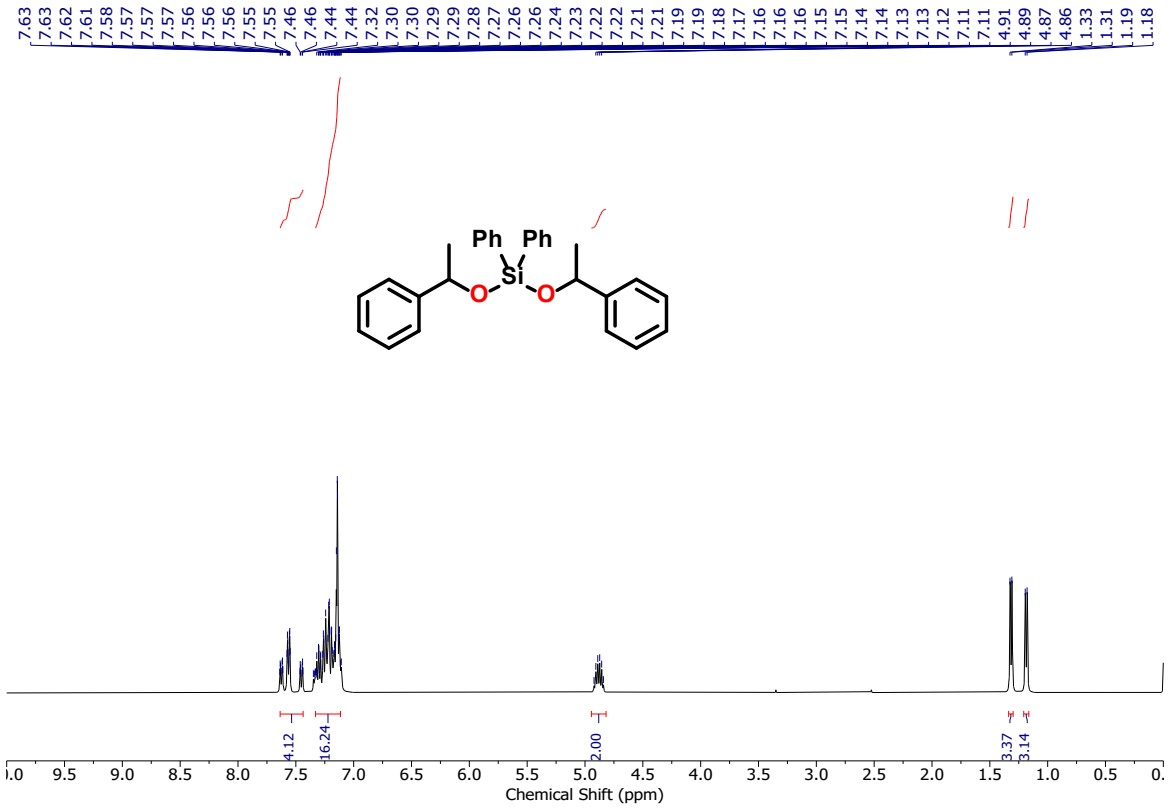


Figure S55: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5a.**

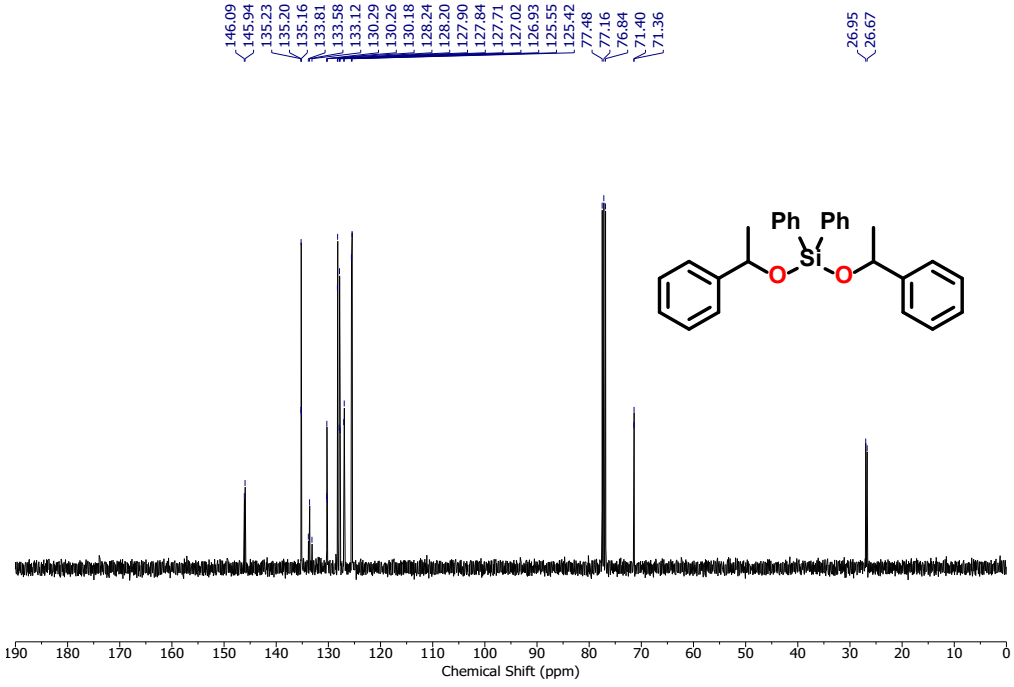


Figure S56: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5a**.

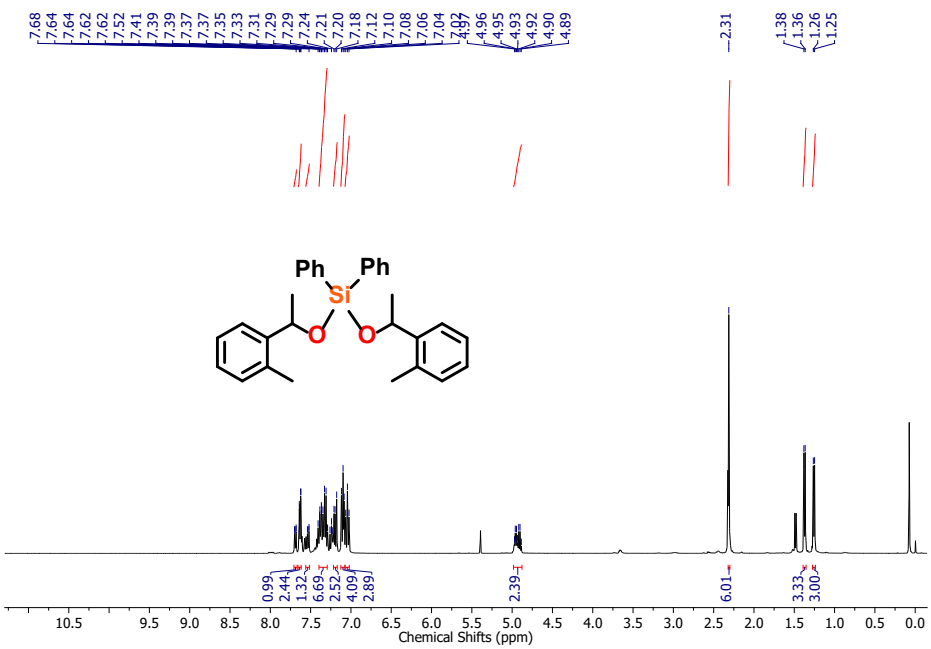


Figure S57: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5b**.

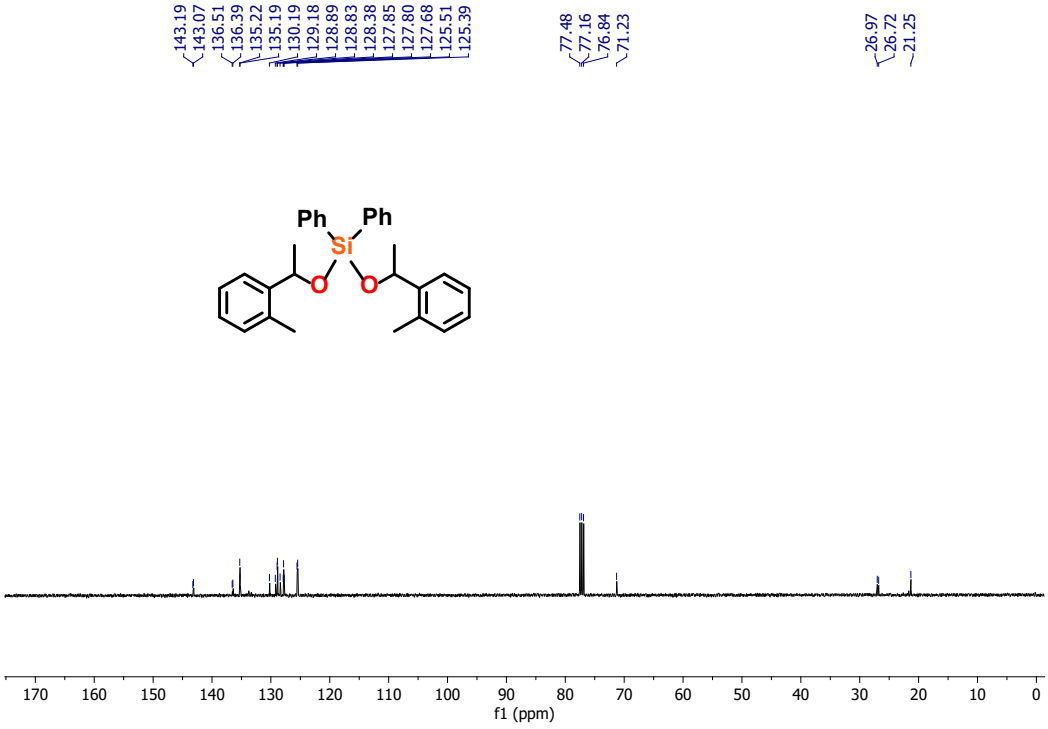


Figure S58: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5b**.

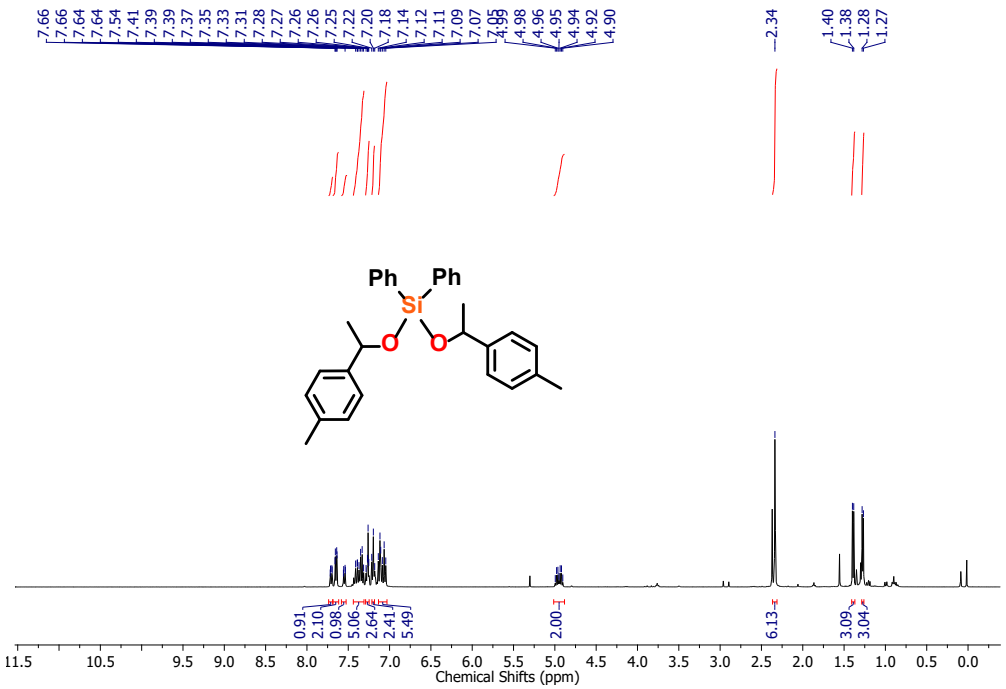


Figure S59: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5c**.

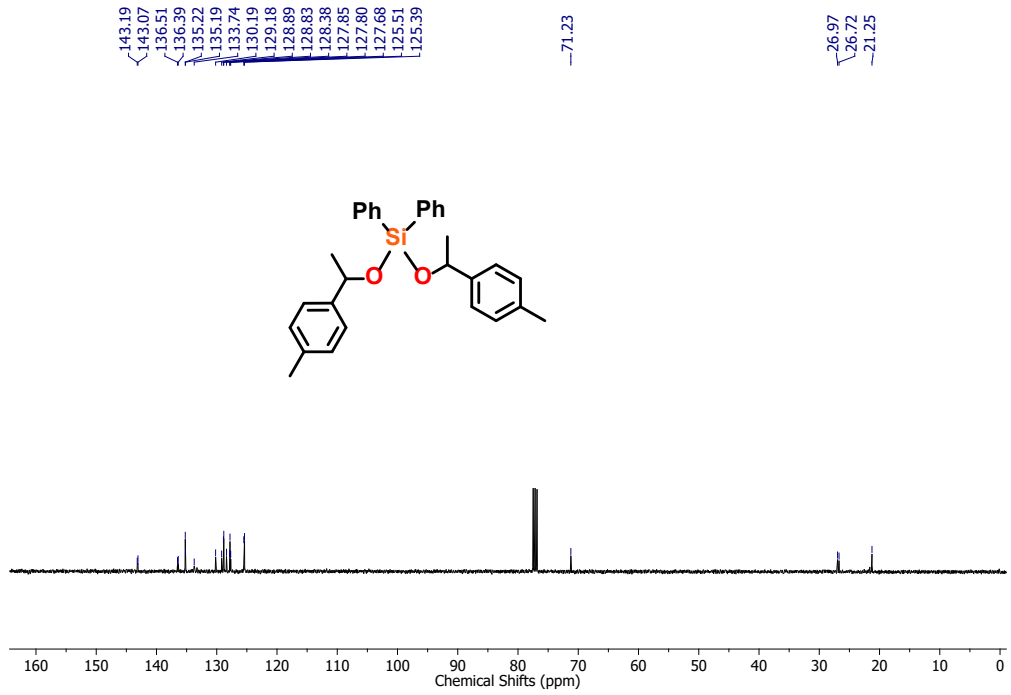


Figure S60: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5c**.

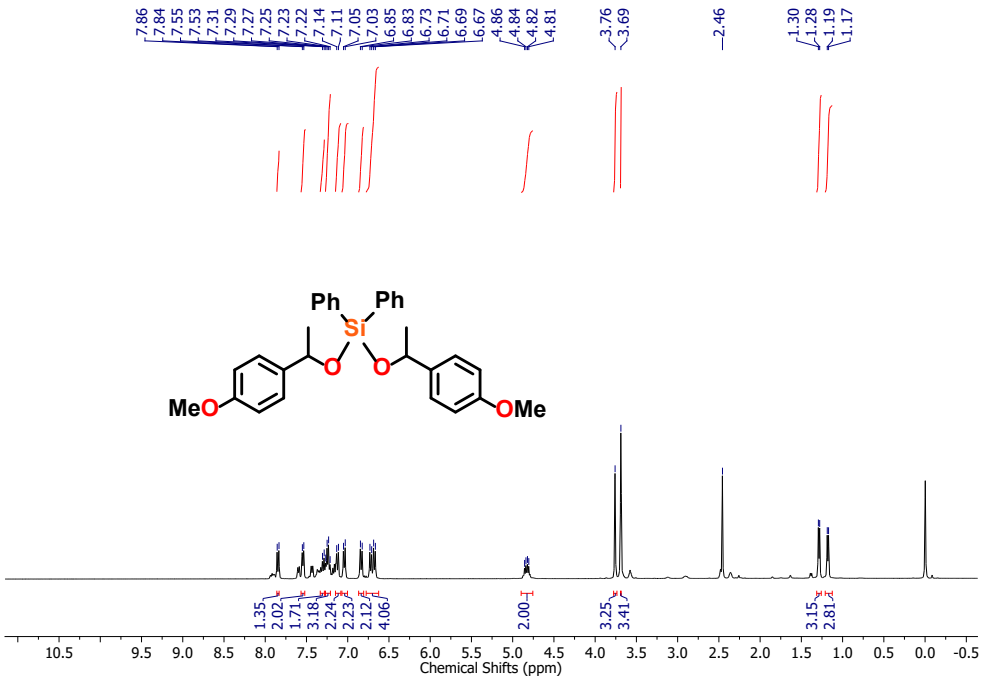


Figure S61: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5d**.

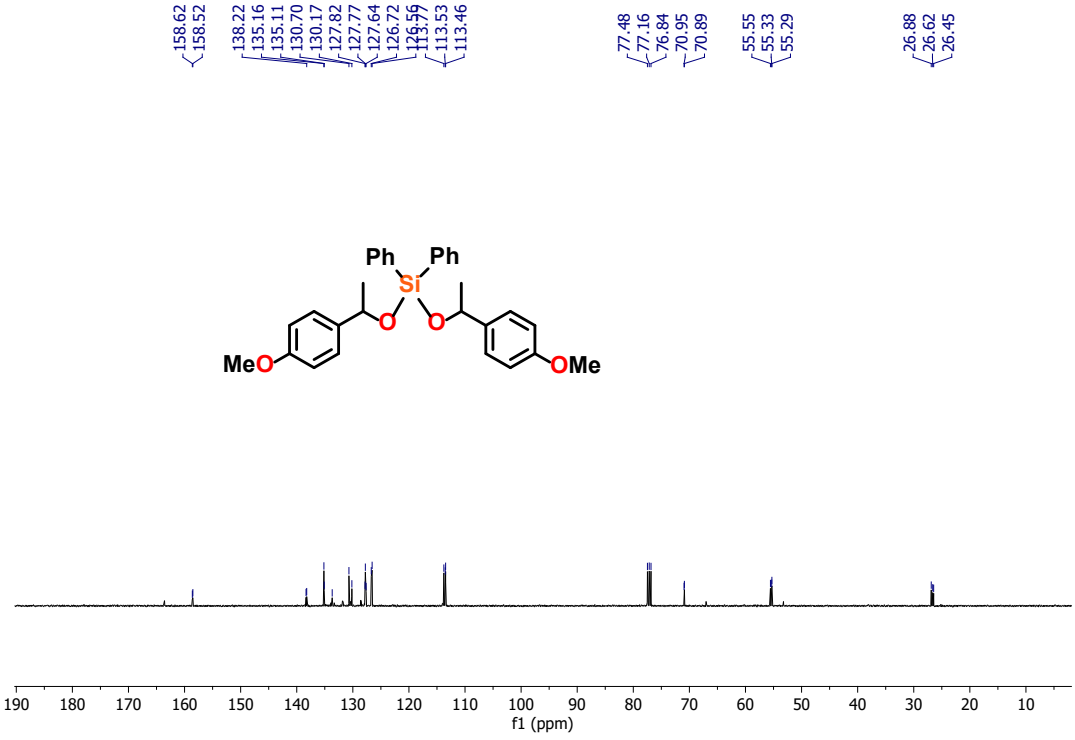


Figure S62: ^{13}C{^{1}H} NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5d**.

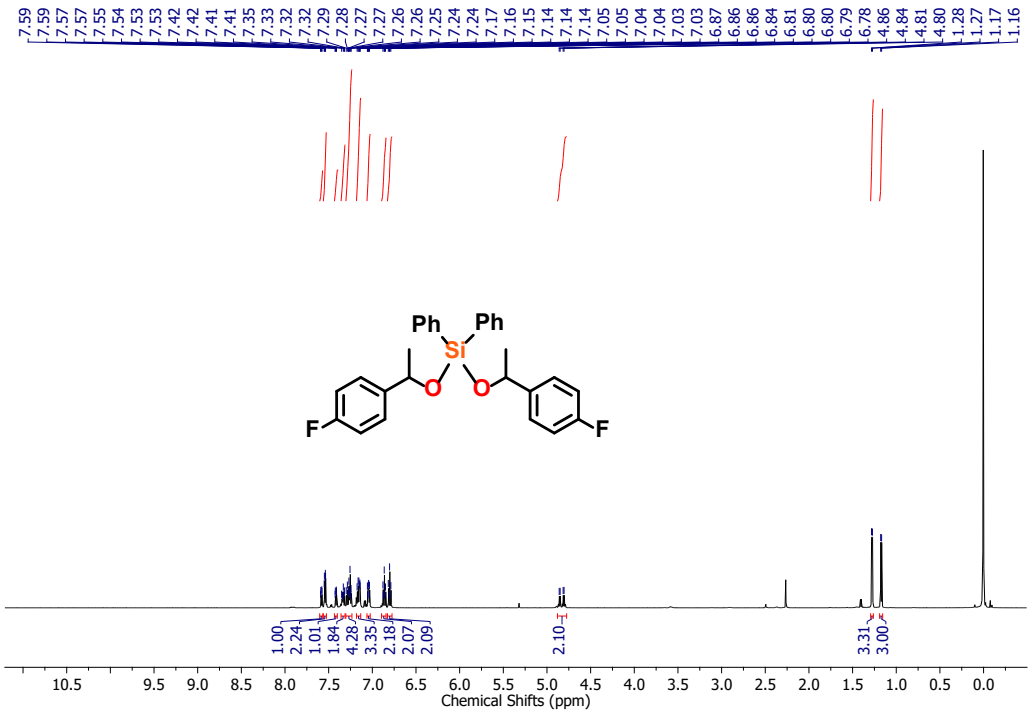


Figure S63: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5e**.

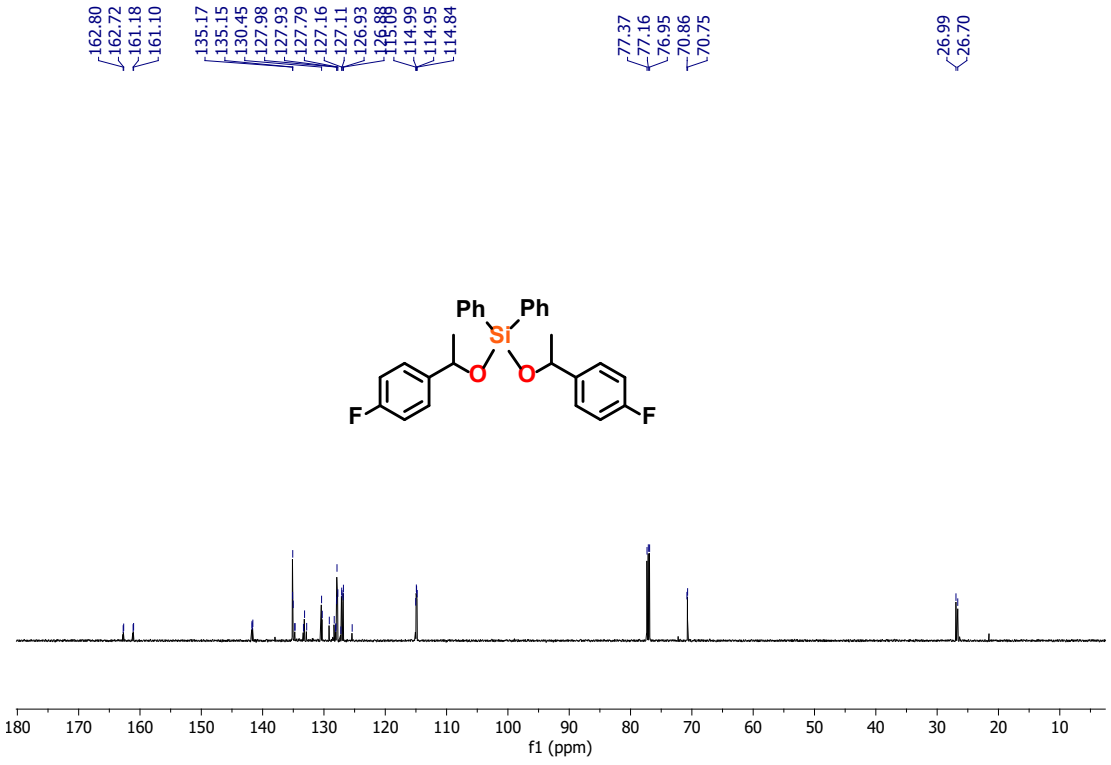


Figure S64: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5e**.

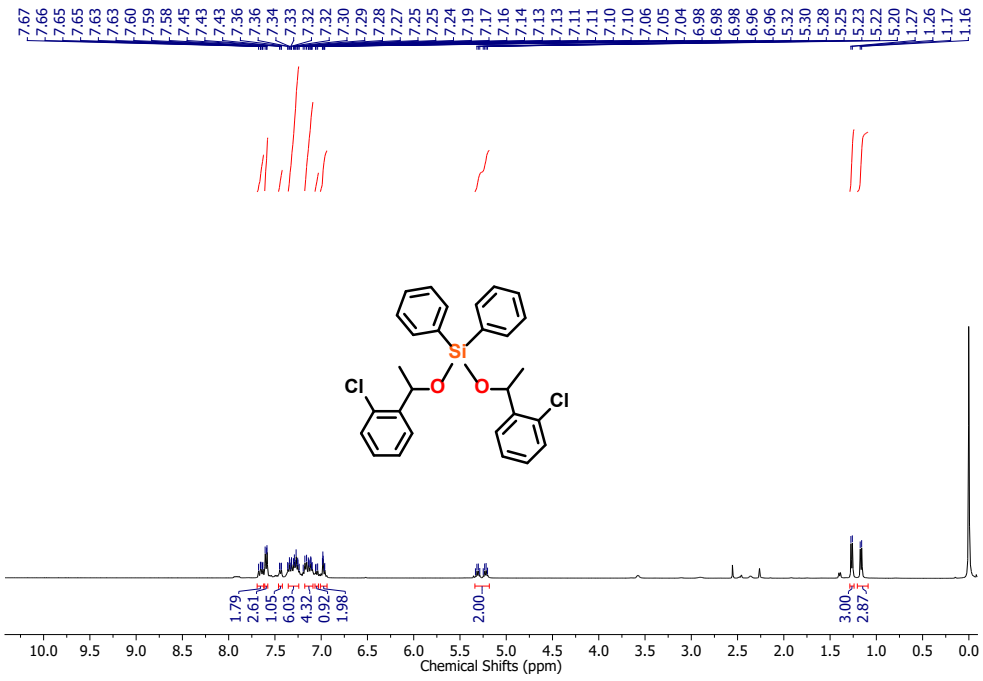


Figure S65: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5f**.

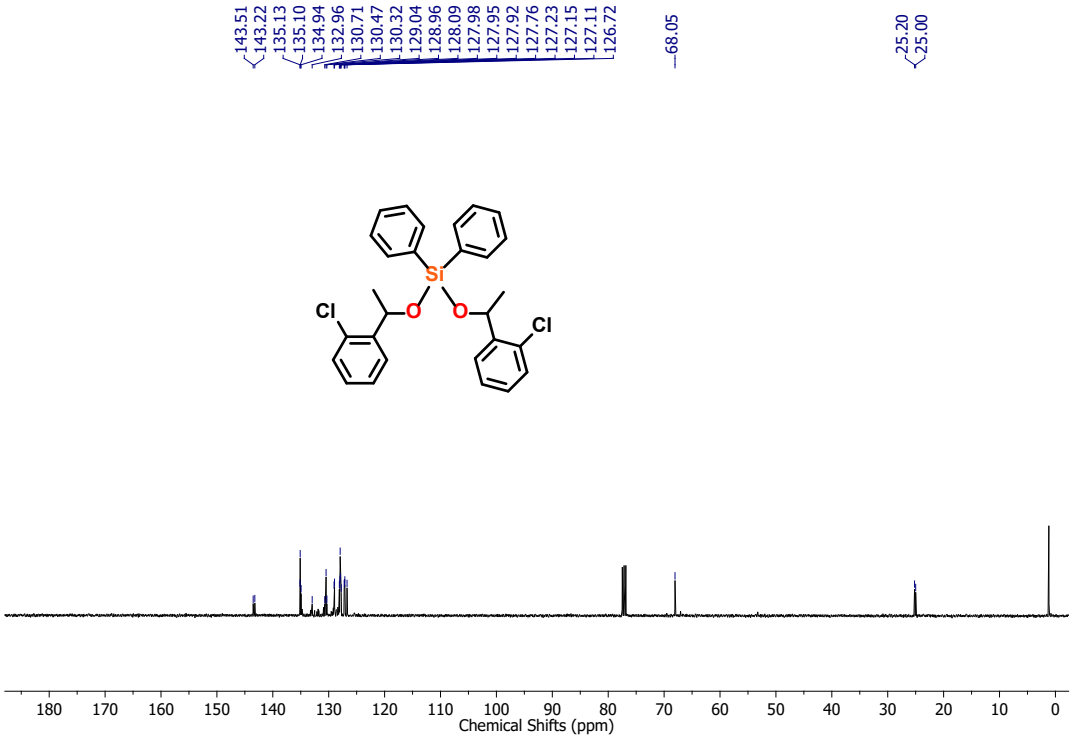


Figure S66: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5f**.

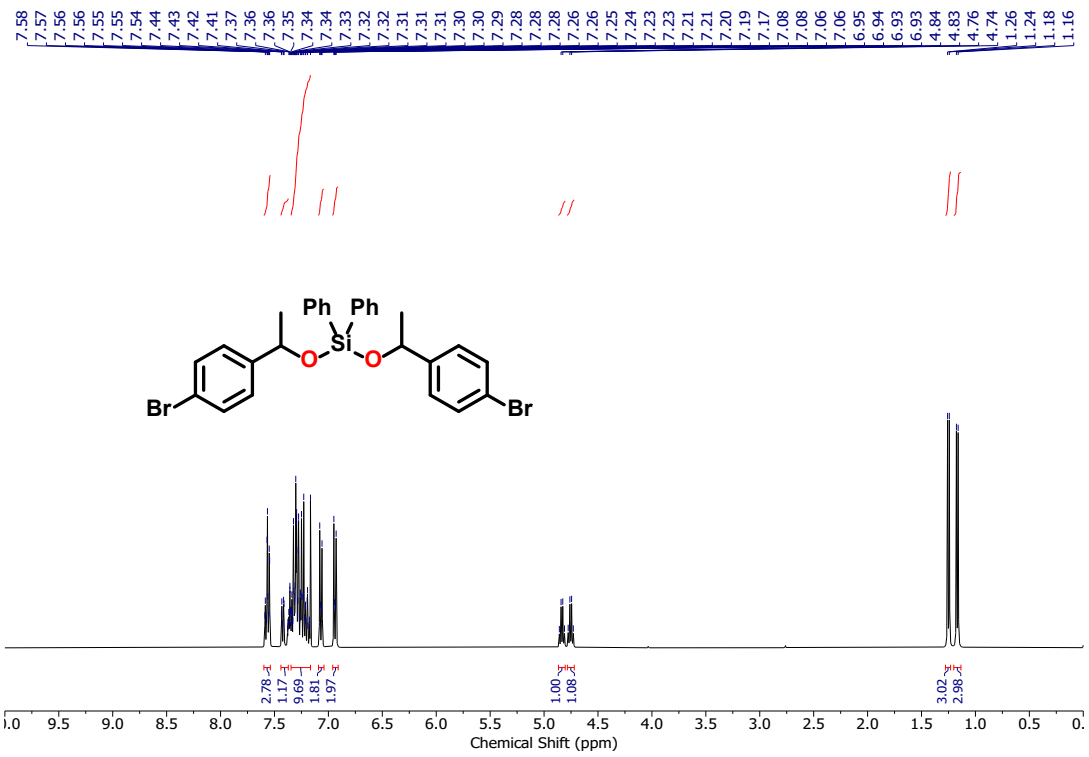


Figure S67: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5g.**

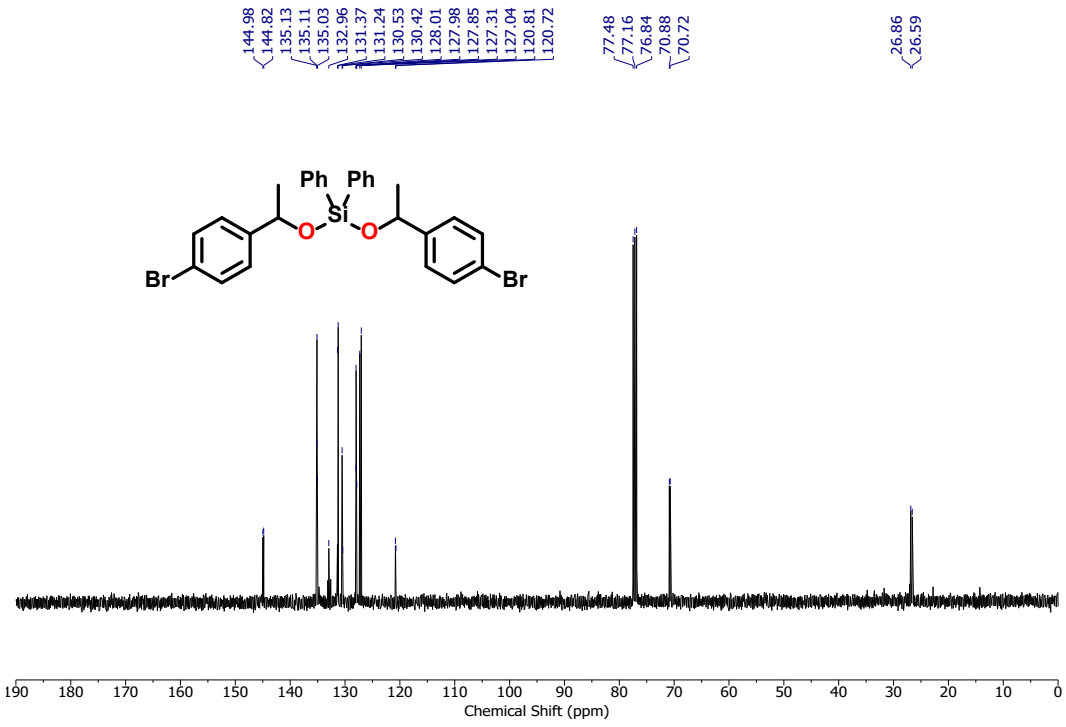


Figure S68: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5g.**

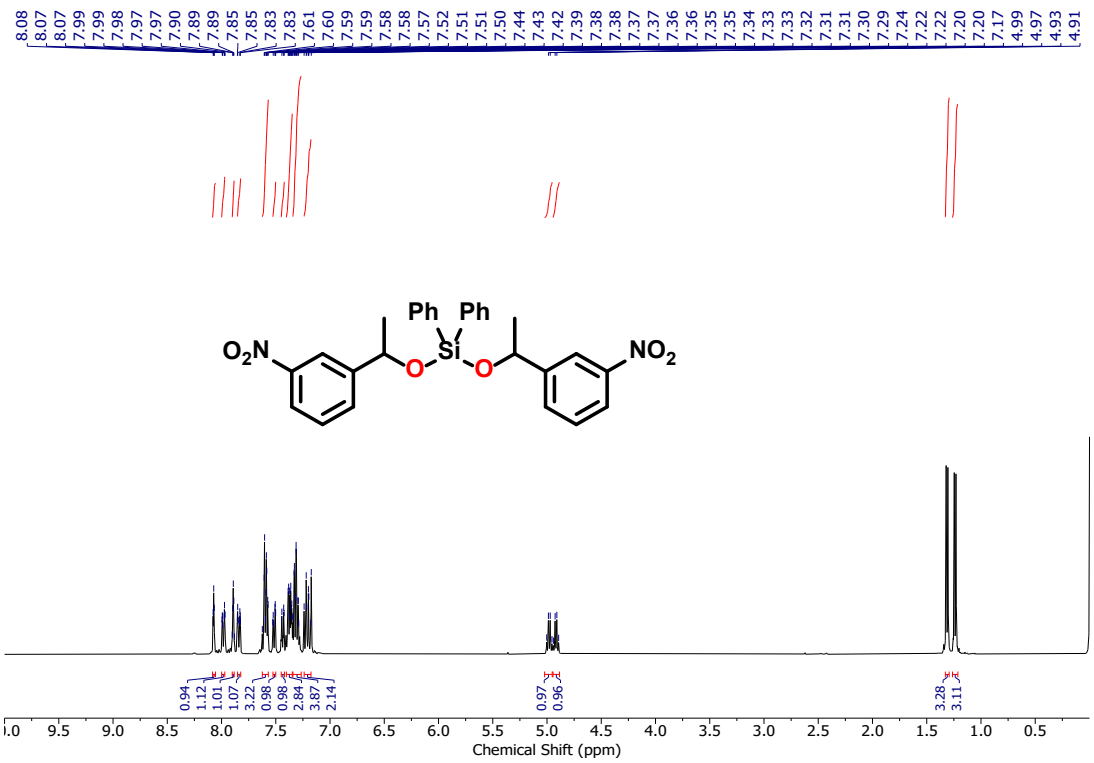


Figure S69: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5h.**

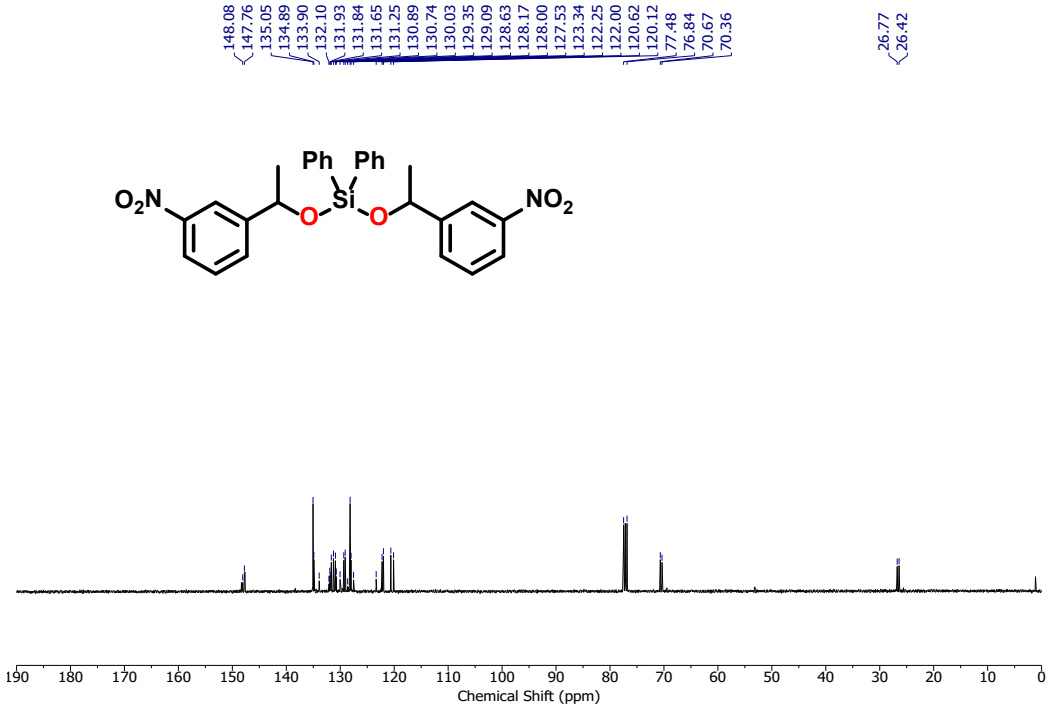


Figure S70: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5h.**

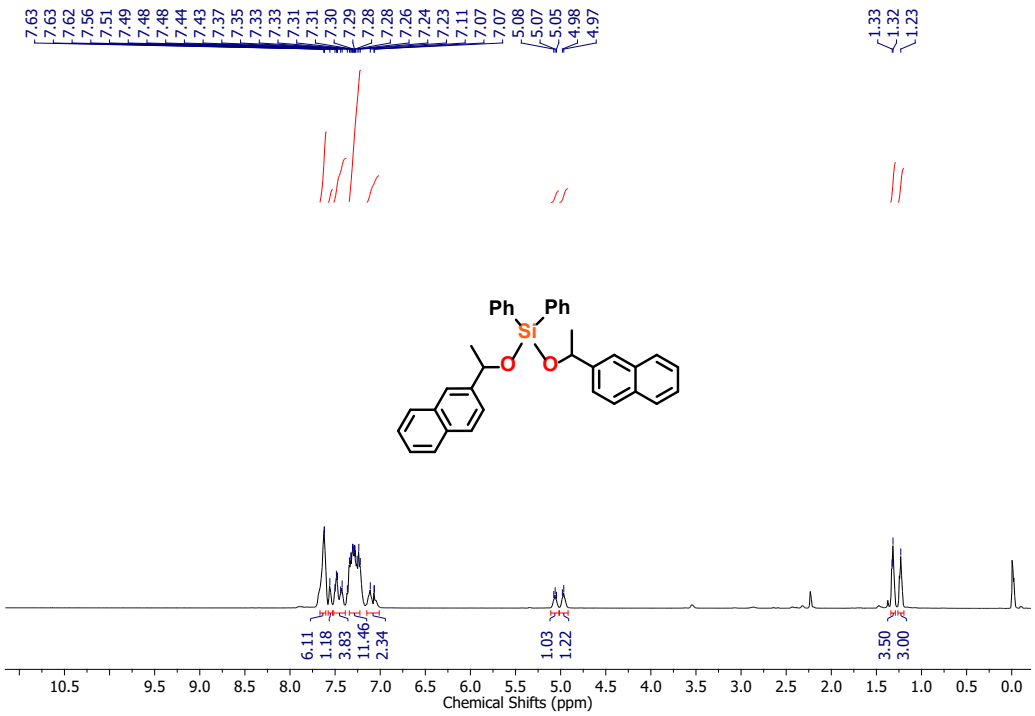


Figure S71: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5i**.

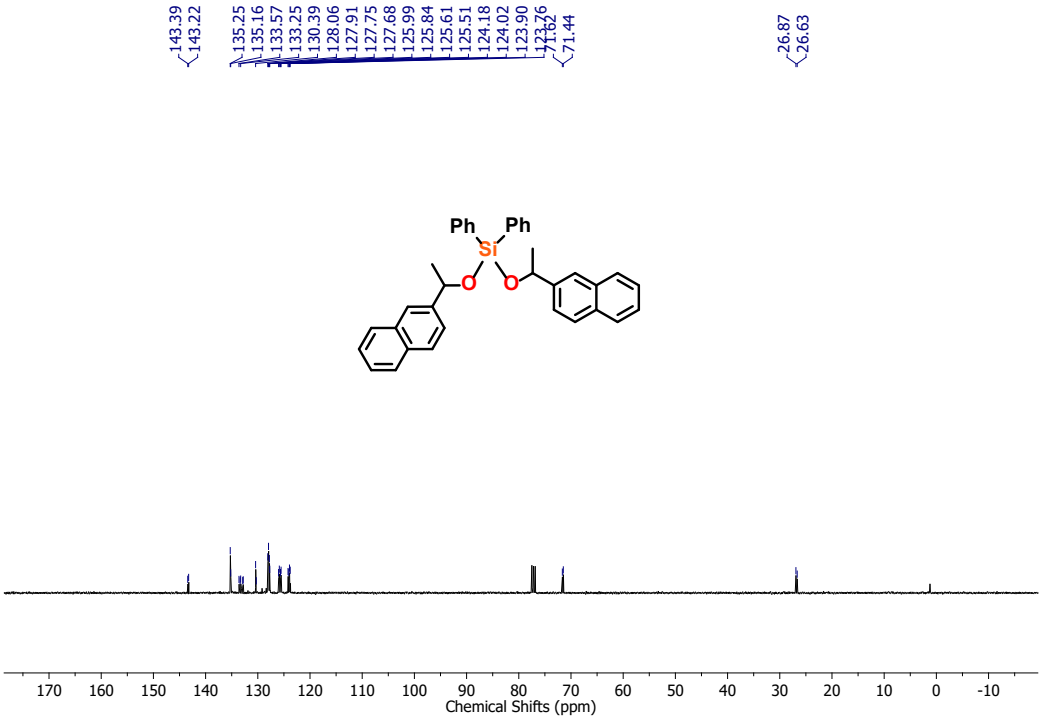


Figure S72: ^{13}C{^{1}H} NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5i**.

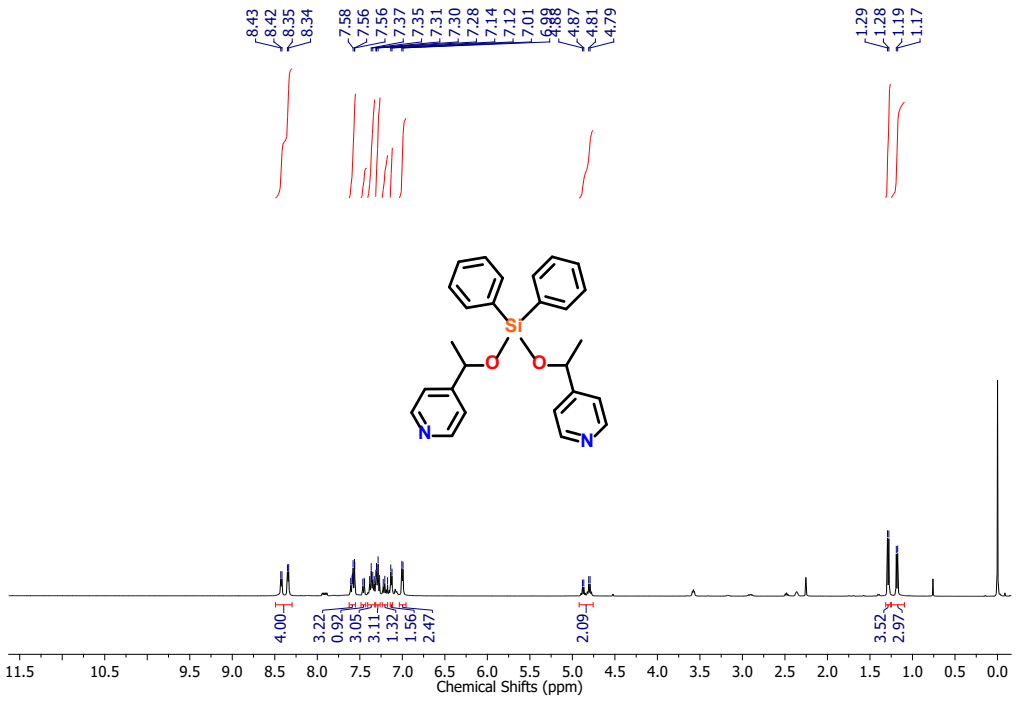


Figure S73: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5j**.

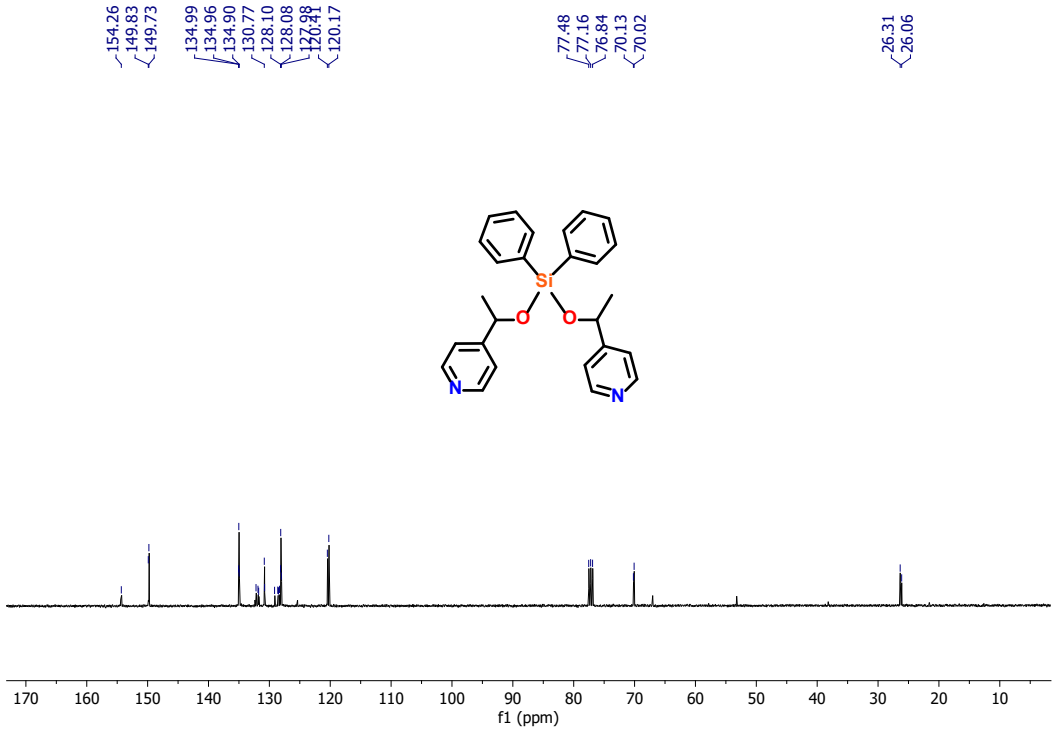


Figure S74: ^{13}C{^{1}H} NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5j**.

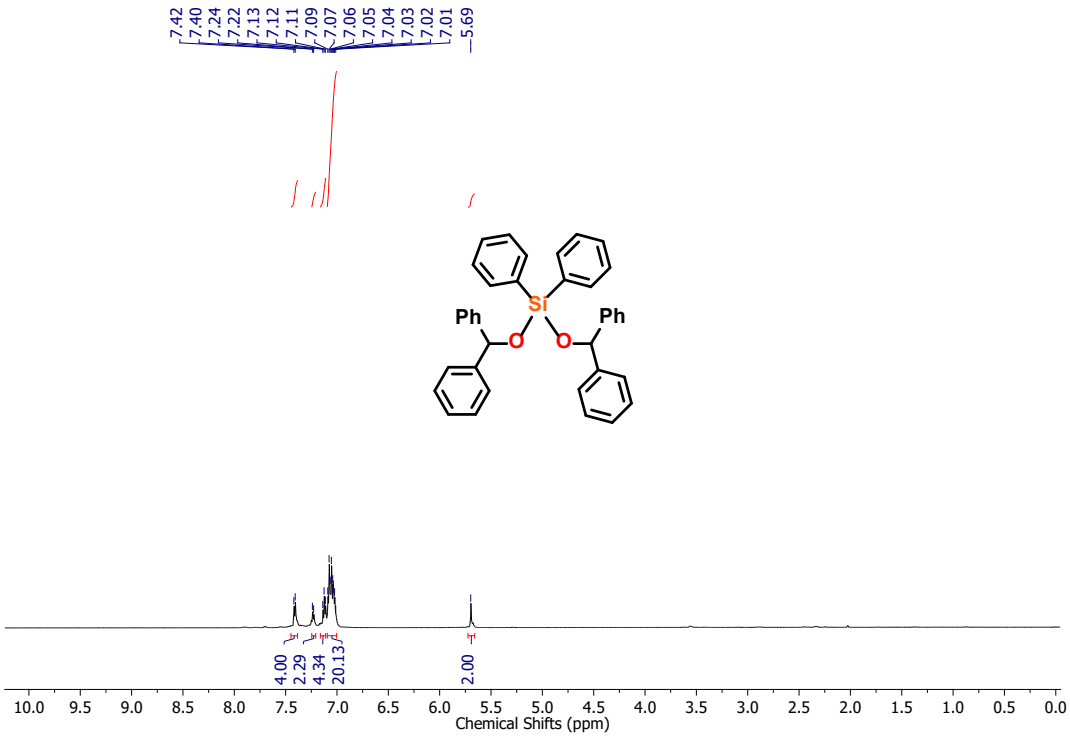


Figure S75: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5k**.

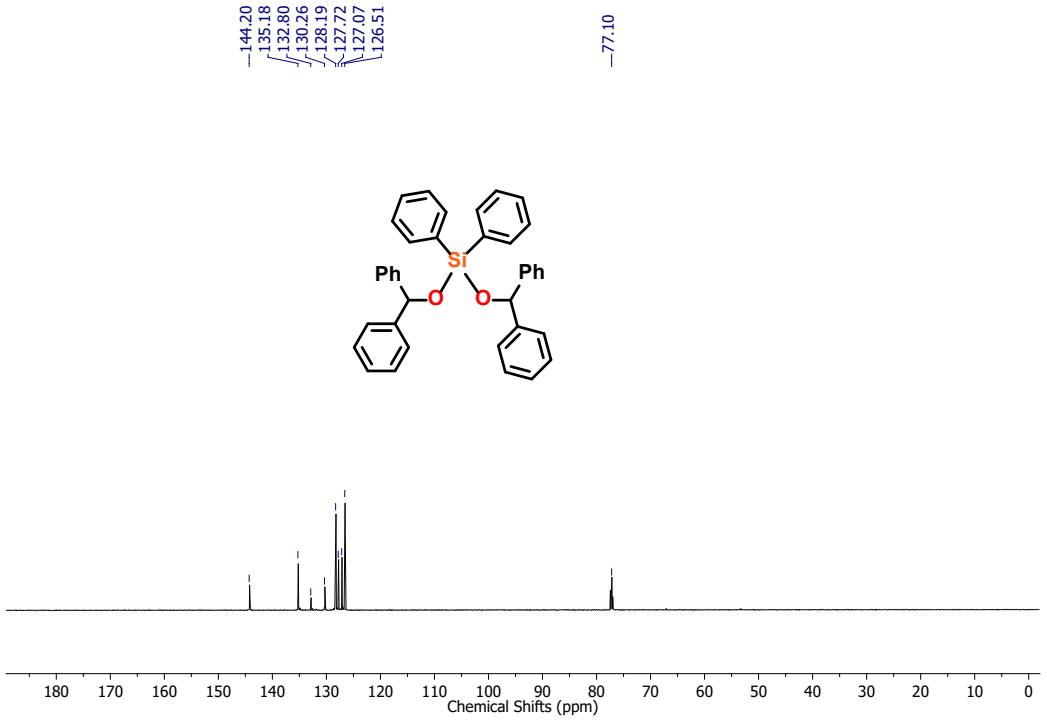


Figure S76: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5k**.

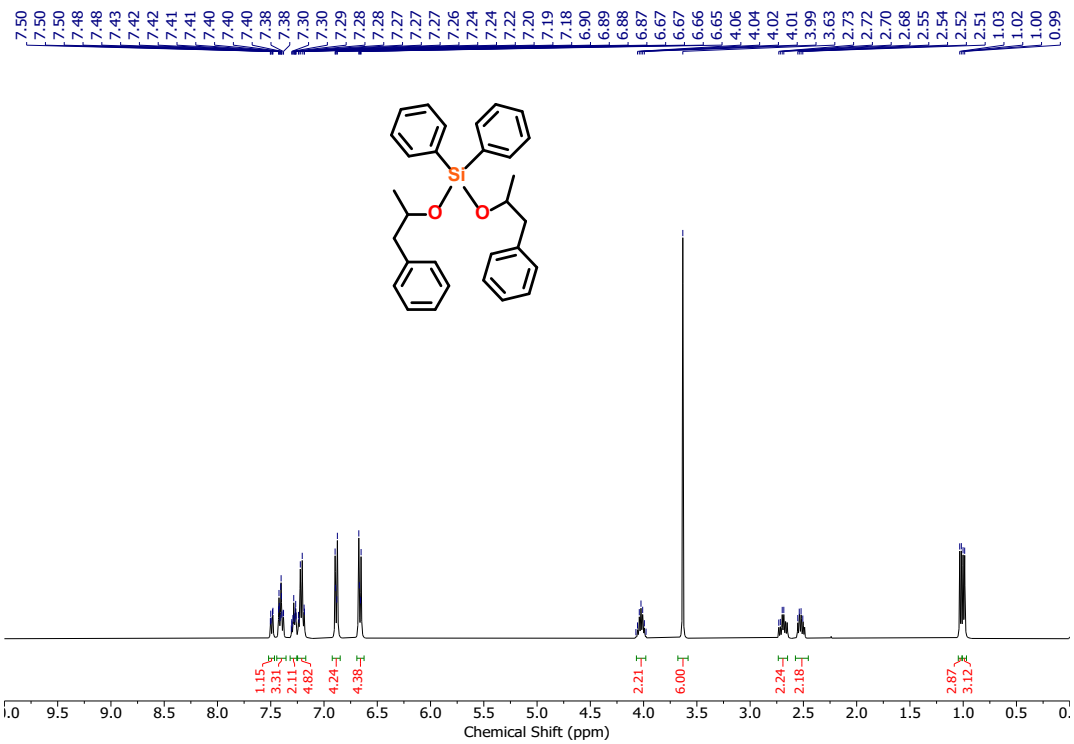


Figure S77: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **5l**.

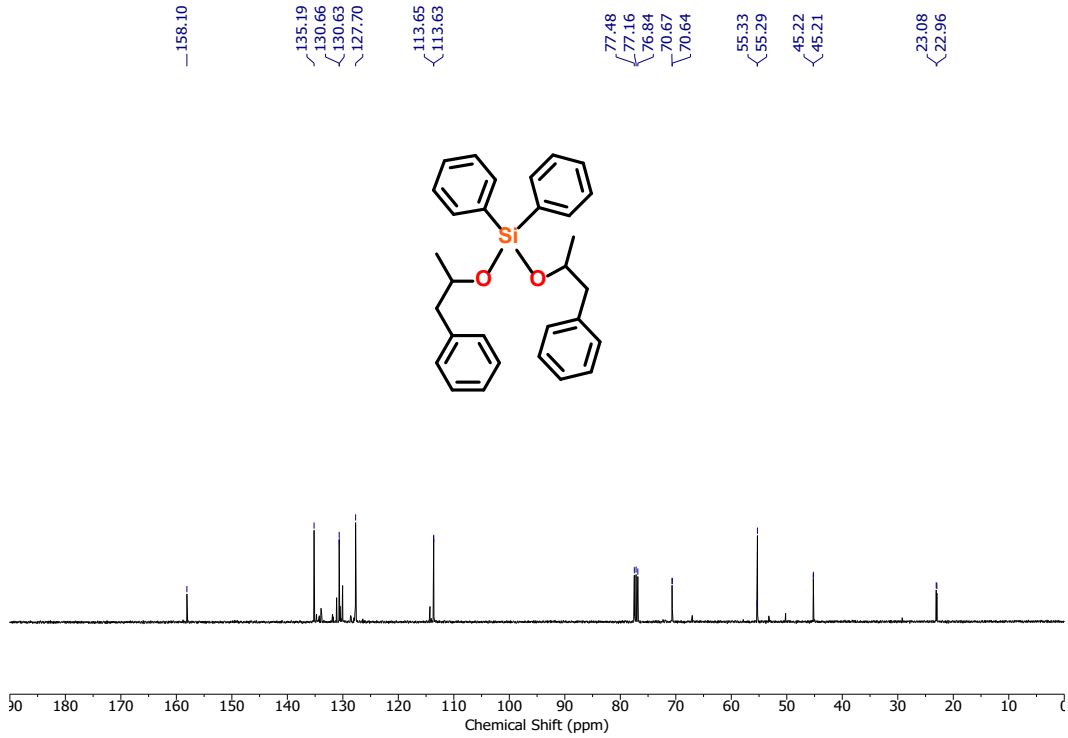
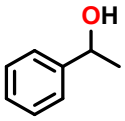


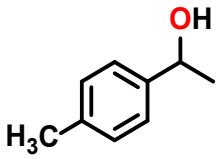
Figure S78: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **5l**.

10. NMR Data for dehydrosilylation products from ketones into alcohols.



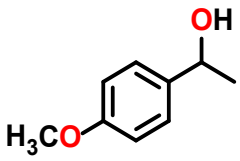
1-phenylethan-1-ol (6a).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 27 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.25 (dd, *J* = 3.3, 1.9 Hz, 5H, Ar*H*), 4.76 (q, *J* = 6.5 Hz, 1H, C*H*), 2.76 (br.s, 1H, O*H*), 1.38 (d, *J* = 6.5 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 145.9 (C_{Ar}), 134.5 (C_{Ar}), 128.6 (C_{Ar}), 127.8 (C_{Ar}), 127.5 (C_{Ar}), 125.5 (C_{Ar}), 70.4 (*C*H), 25.2 (*C*H_3) ppm.



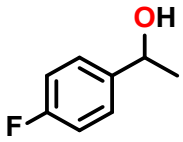
1-(p-tolyl)ethan-1-ol (6b).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 32 mg (98 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.27 (d, *J* = 8.3 Hz, 2H, Ar*H*), 7.16 (d, *J* = 7.9 Hz, 2H, Ar*H*), 4.87 (q, *J* = 6.4 Hz, 1H, C*H*), 2.35 (s, 3H, CH_3), 1.70 (br. s, 1H, O*H*), 1.49 (d, *J* = 6.5 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 142.9 (C_{Ar}), 137.3 (C_{Ar}), 129.3 (C_{Ar}), 125.5 (C_{Ar}), 70.4 (*C*H), 25.2 (*C*H_3), 21.2 (*C*H_3) ppm.



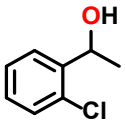
1-(4-methoxyphenyl)ethan-1-ol (6c).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 36 mg (98 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.33 – 7.26 (m, 2H, Ar*H*), 6.92 – 6.85 (m, 2H, Ar*H*), 4.85 (q, *J* = 6.4 Hz, 1H, C*H*), 3.80 (s, 3H, OCH_3), 1.87 (br. s, 1H, O*H*), 1.47 (d, *J* = 6.5 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 159.1 (C_{Ar}), 138.1 (C_{Ar}), 126.8 (C_{Ar}), 113.9 (C_{Ar}), 70.1 (*C*H), 55.4 (O*C*H$_3$), 25.1 (*C*H$_3$) ppm.



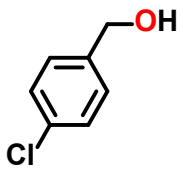
1-(4-fluorophenyl)ethan-1-ol (6d).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 30 mg (95 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.35 – 7.29 (m, 2H, Ar*H*), 7.05 – 6.99 (m, 2H, Ar*H*), 4.87 (q, *J* = 6.4 Hz, 1H, C*H*), 2.16 (br. s, 1H, O*H*), 1.46 (d, *J* = 6.4 Hz, 3H, CH_3) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 163.4 (C_{Ar}), 161.0 (C_{Ar}), 141.6 (d, *J* = 3.1 Hz, C_{Ar}), 127.2 (d, *J* = 8.1 Hz, C_{Ar}), 115.5 (C_{Ar}), 115.2 (C_{Ar}), 69.9 (C_{Ar}), 25.4 (C_{Ar}) ppm.



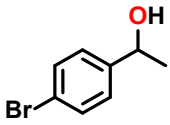
1-(2-chlorophenyl)ethan-1-ol (6e).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 35 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.53 (dd, *J* = 7.7, 1.7 Hz, 1H, Ar*H*), 7.30 – 7.21 (m, 2H, Ar*H*), 7.15 (td, *J* = 7.6, 1.7 Hz, 1H, Ar*H*), 5.23 (q, *J* = 6.4 Hz, 1H, C*H*), 2.35 (br. s, 1H, O*H*), 1.43 (d, *J* = 6.4 Hz, 3H, *CH*$_3$) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 143.2 (C_{Ar}), 131.7 (C_{Ar}), 129.5 (C_{Ar}), 128.5 (C_{Ar}), 127.3 (C_{Ar}), 126.5 (C_{Ar}), 67.0 (*C*H), 23.6 (*C*H$_3$) ppm.



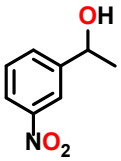
1-(4-chlorophenyl)ethan-1-ol (6f).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 43 mg (96 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.34 – 7.25 (m, 4H, Ar*H*), 4.64 (s, 2H, CH_2), 1.98 (br.s, 1H, O*H*) ppm. $^{13}C\{^{1}H\}$ NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 139.4 (C_{Ar}), 133.5 (C_{Ar}), 128.8 (C_{Ar}), 128.4 (C_{Ar}), 64.6 (CH_2) ppm.



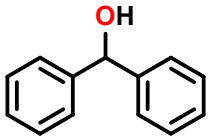
1-(4-bromophenyl)ethan-1-ol (6g).[7]

Following procedure **12**, the product was isolated. ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.46 – 7.42 (m, 2H, Ar*H*), 7.23 – 7.17 (m, 2H, Ar*H*), 4.80 (q, J = 6.5 Hz, 1H, C*H*), 2.52 (br. s, 1H, O*H*), 1.42 (d, J = 6.5 Hz, 3H, CH_3) ppm. $^{13}C\{^{1}H\}$ NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 144.8 (C_{Ar}), 131.6 (C_{Ar}), 127.2 (C_{Ar}), 121.2 (C_{Ar}), 69.7 (*C*H), 25.3 (*C*H_3) ppm.



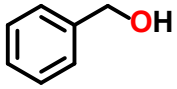
1-(3-nitrophenyl)ethan-1-ol (6h).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 41 mg (95 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.94 (s, 1H, Ar*H*), 7.86 – 7.82 (m, 1H, Ar*H*), 7.53 – 7.50 (m, 2H, Ar*H*), 5.03 (q, J = 6.4 Hz, 1H, C*H*), 1.93 (s, 1H, O*H*), 1.56 (s, 3H, CH_3) ppm. $^{13}C\{^{1}H\}$ NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 134.5 (C_{Ar}), 129.6 (C_{Ar}), 115.8 (C_{Ar}), 114.4 (C_{Ar}), 112.1 (C_{Ar}), 70.5 (*C*H), 25.1 (*C*H_3) ppm.



diphenylmethanol (6i).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 42 mg (92 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.41 – 7.30 (m, 8H, Ar*H*), 7.30 – 7.23 (m, 2H, Ar*H*), 5.84 (s, 1H, C*H*), 2.26 (br. s, 1H, O*H*) ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 143.9 (C_{Ar}), 128.6 (C_{Ar}), 127.7 (C_{Ar}), 126.7 (C_{Ar}), 76.4 (*C*H) ppm.



Benzyl alcohol (6j).[7]

Following procedure **12**, the product was isolated. Isolated Yield: 24 mg (92 %). ^{1}H NMR (400 MHz, 25 °C, $CDCl_3$) δ_H 7.40 – 7.27 (m, 5H, Ar*H*), 4.64 (s, 2H, CH_2), 2.62 (br. s, 1H, O*H*). ppm. ^{13}C{^{1}H} NMR (100 MHz, 25 °C, $CDCl_3$) δ_C 140.9 (C_{Ar}), 131.8 (C_{Ar}), 128.5 (C_{Ar}), 127.6 (C_{Ar}), 127.0 (C_{Ar}), 65.14 (*C*H_2) ppm.

11. Spectra for the alcohols.

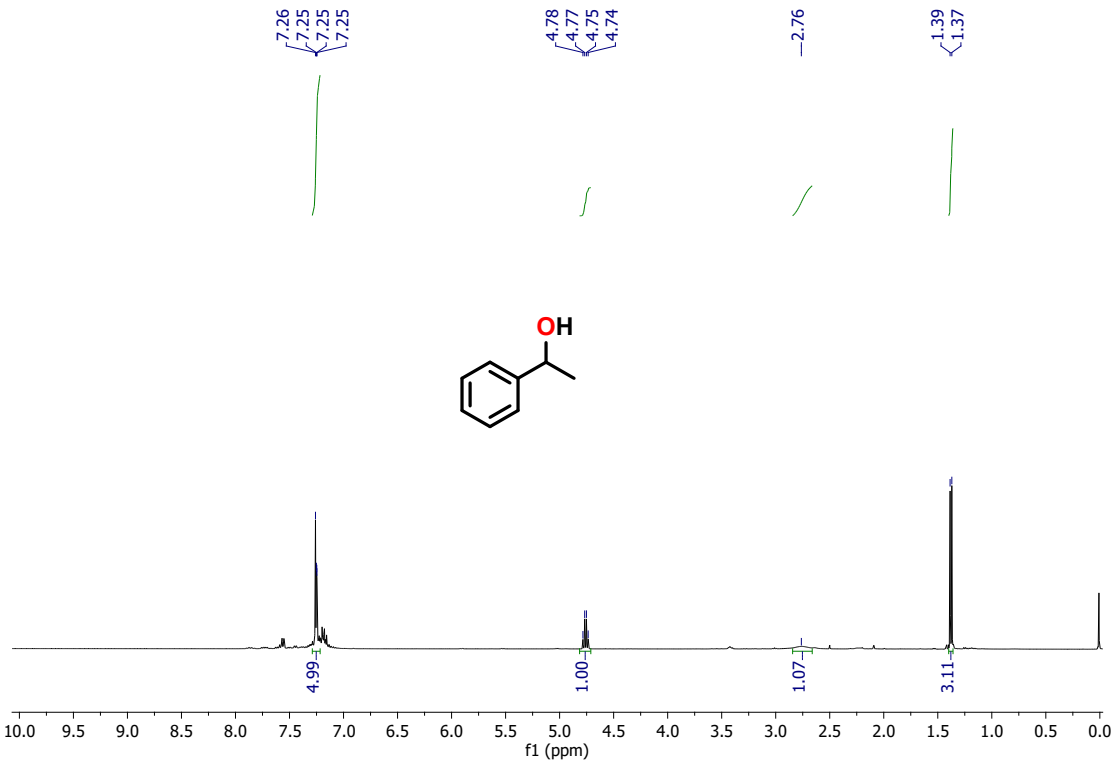


Figure S79: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6a**.

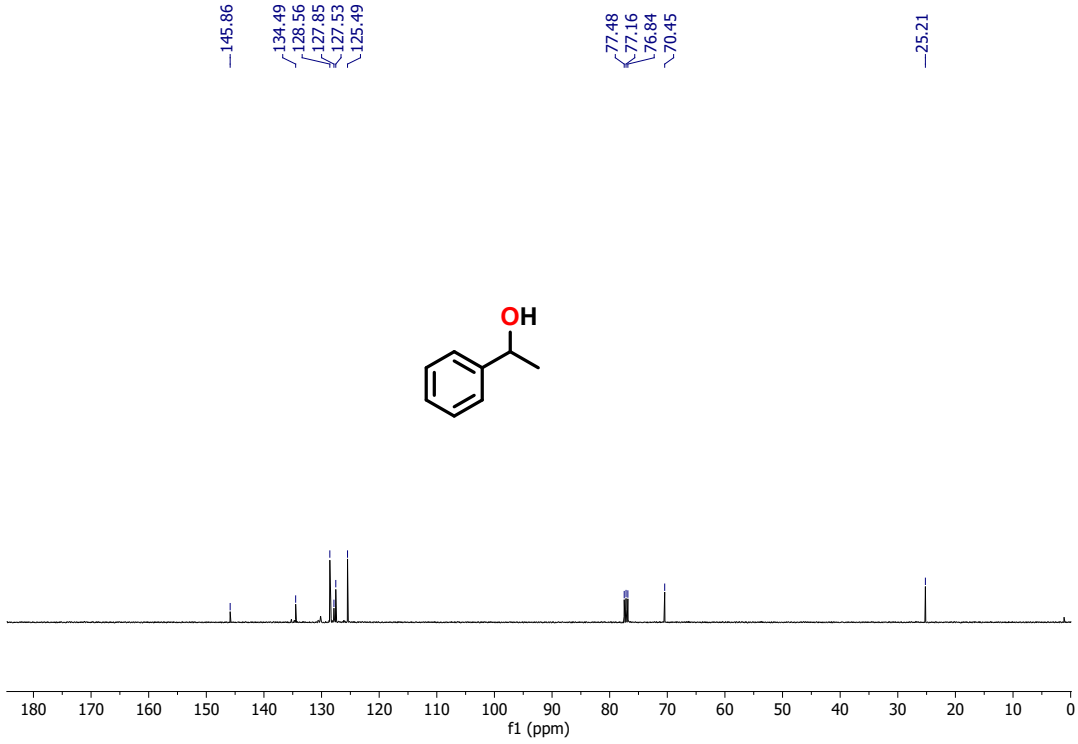


Figure S80: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6a**.

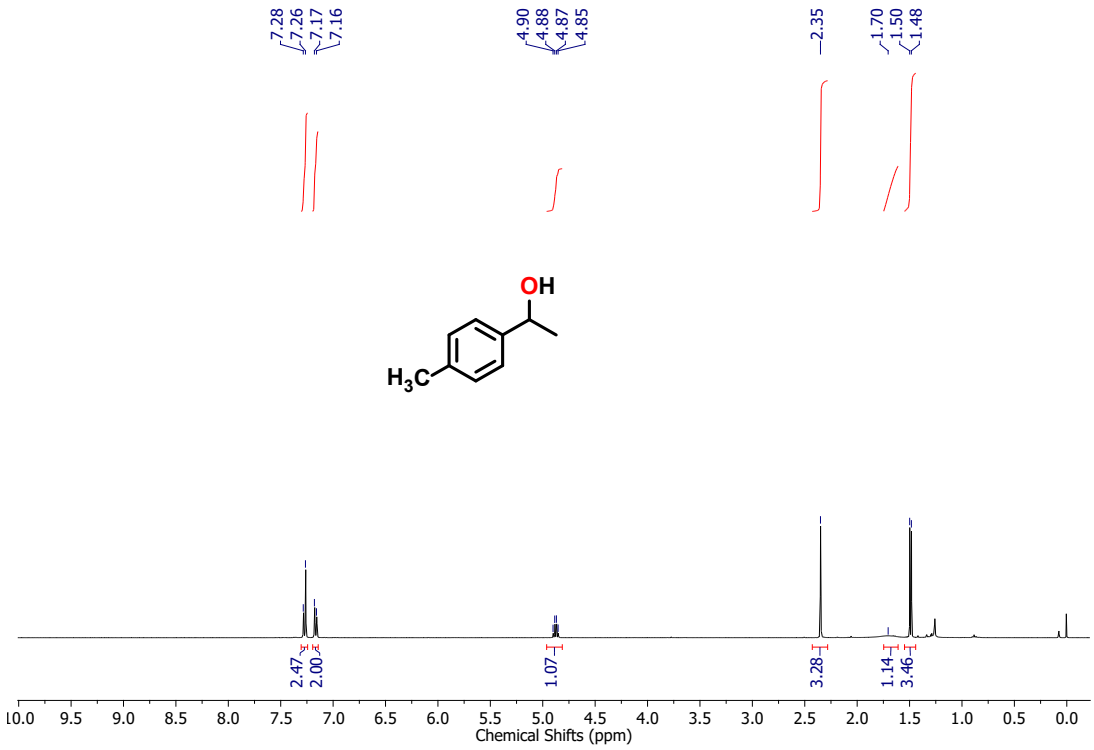


Figure S81: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6b**.

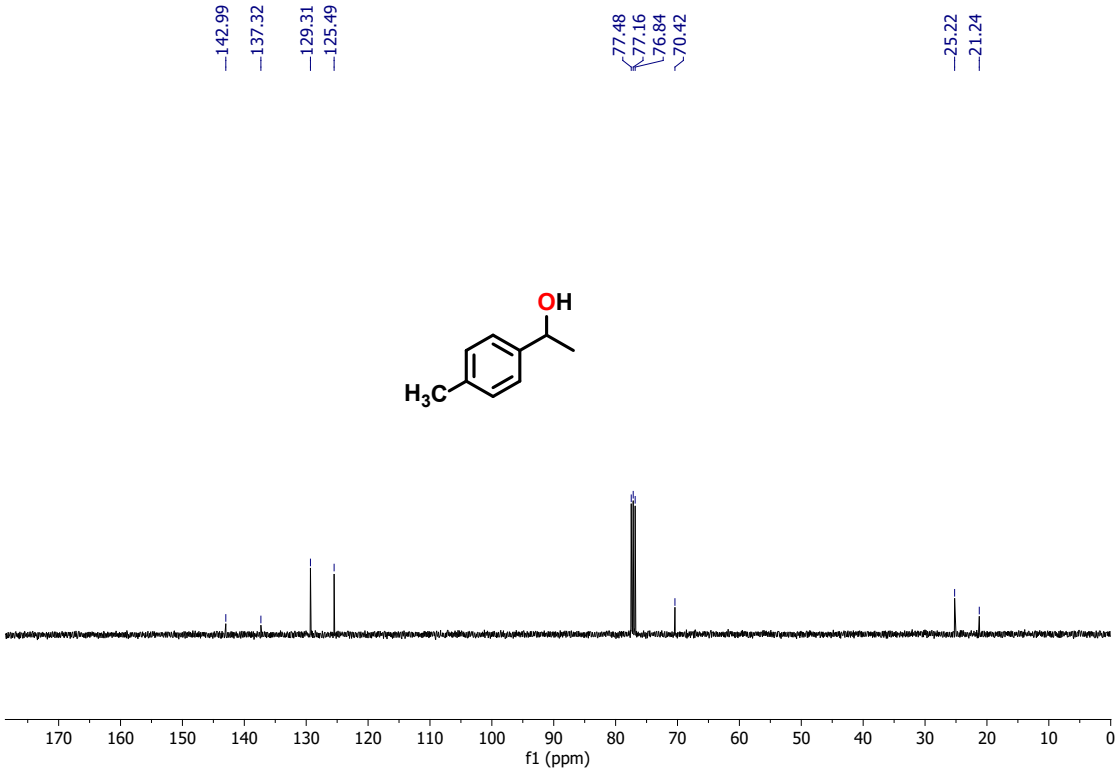


Figure S82: ^{13}C{^{1}H} NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6b**.

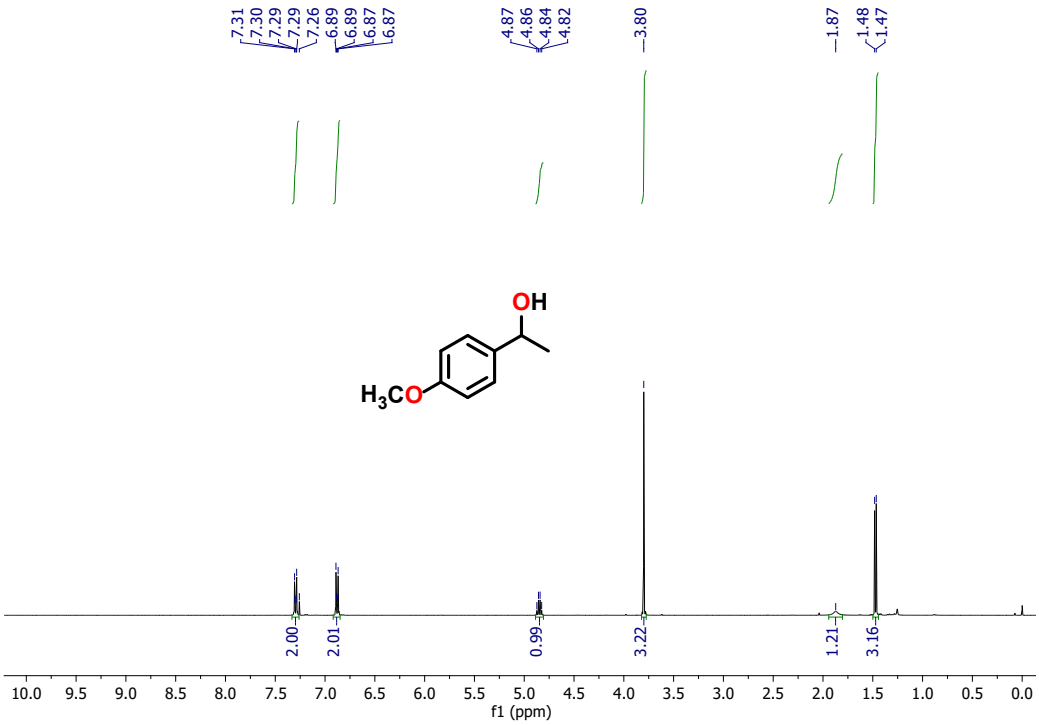


Figure S83: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6c**.

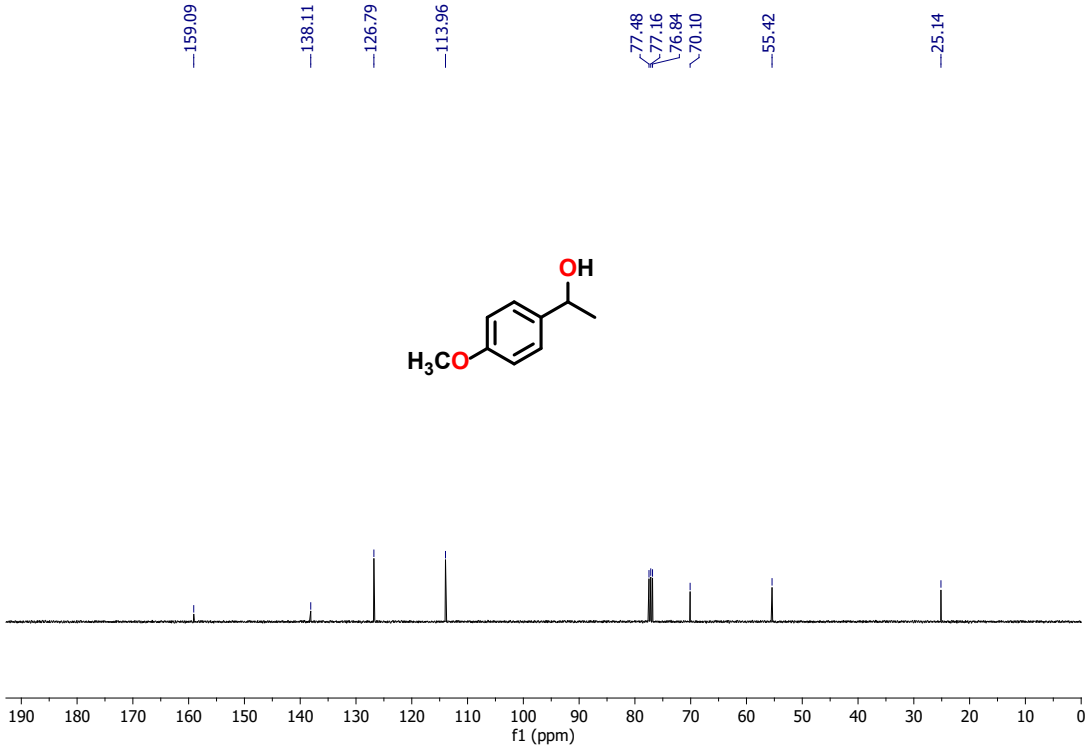


Figure S84: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6c**.

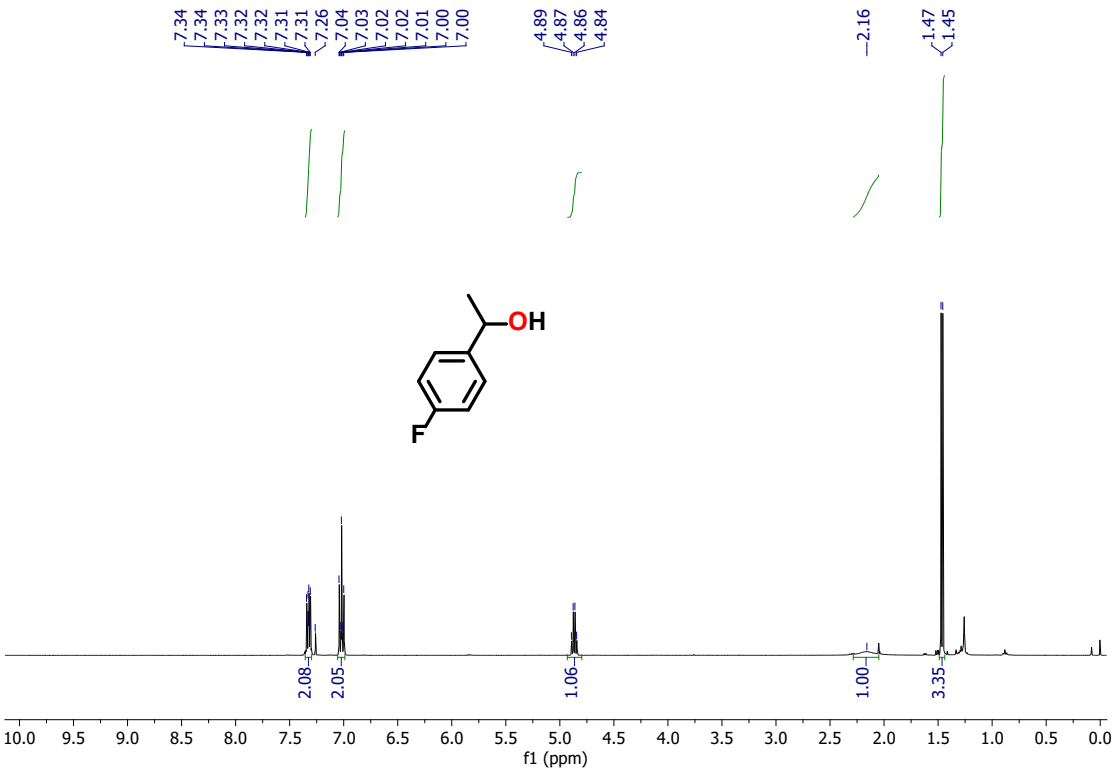


Figure S85: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6d**.

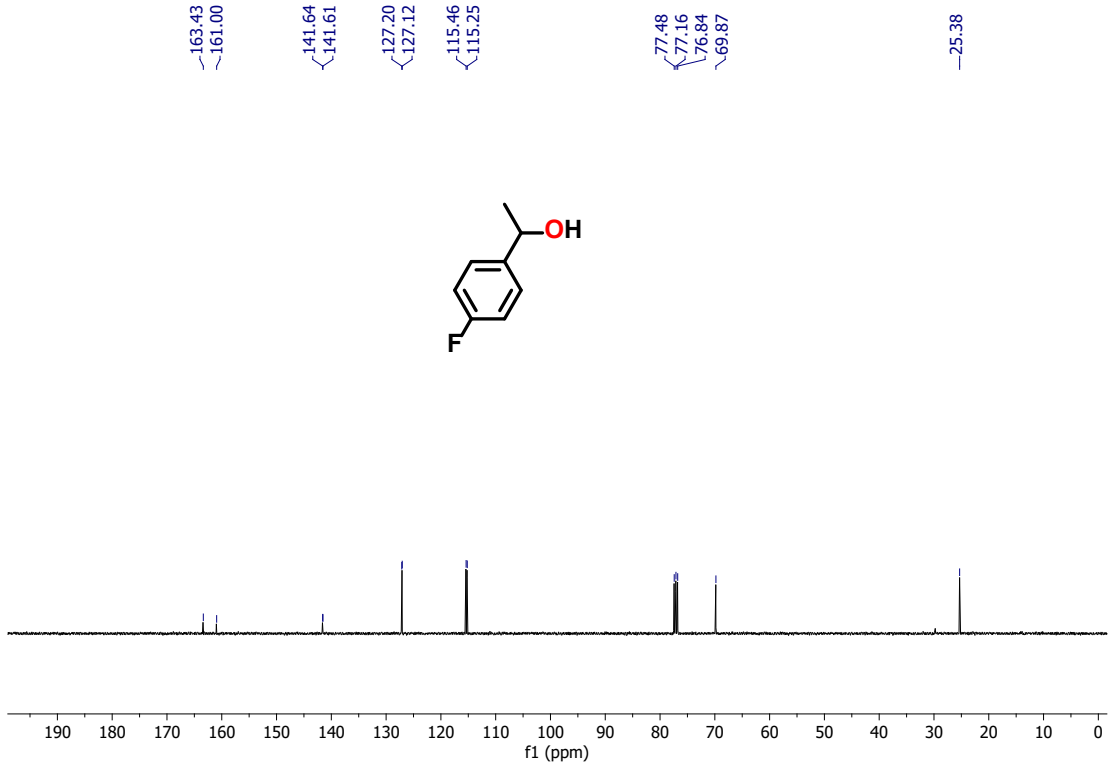


Figure S86: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6d**.

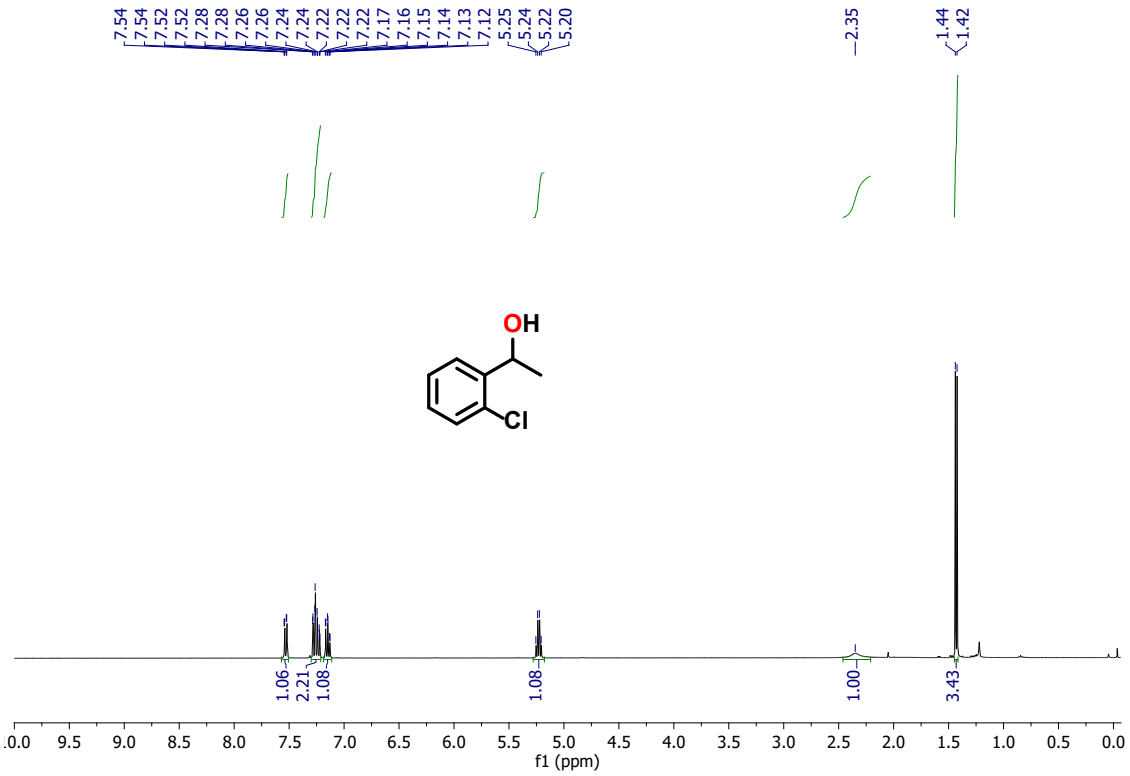


Figure S87: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6e**.

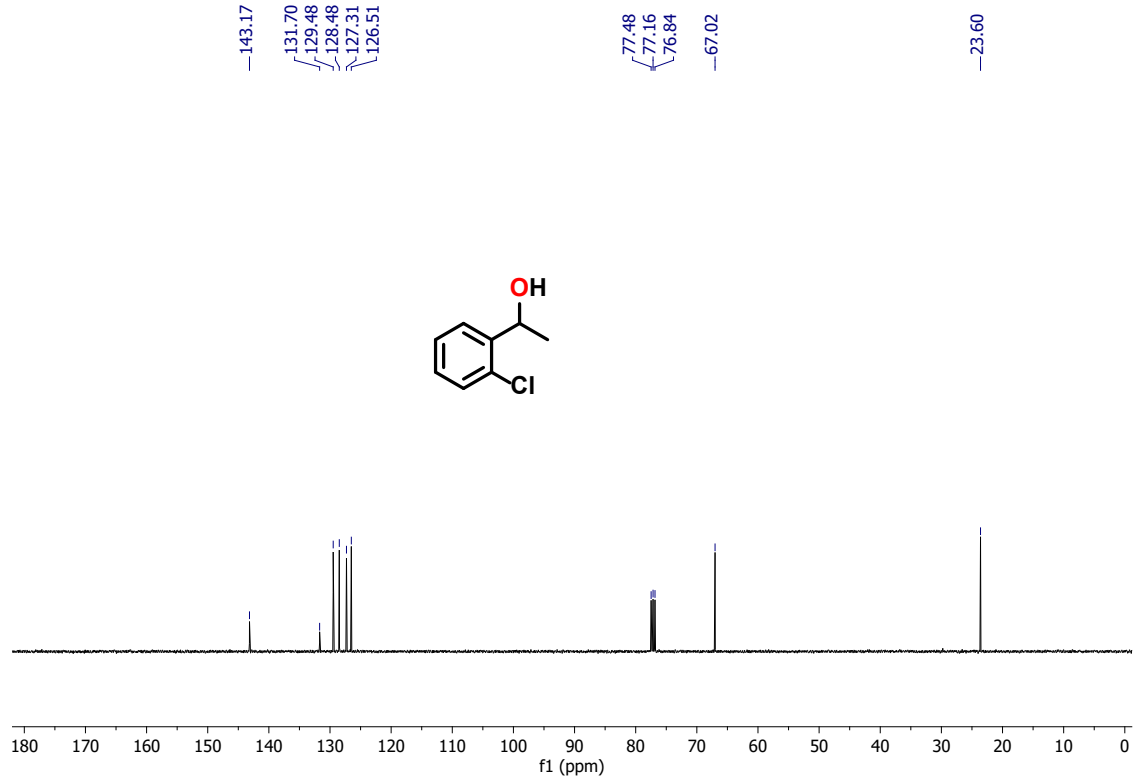


Figure S88: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6e**.

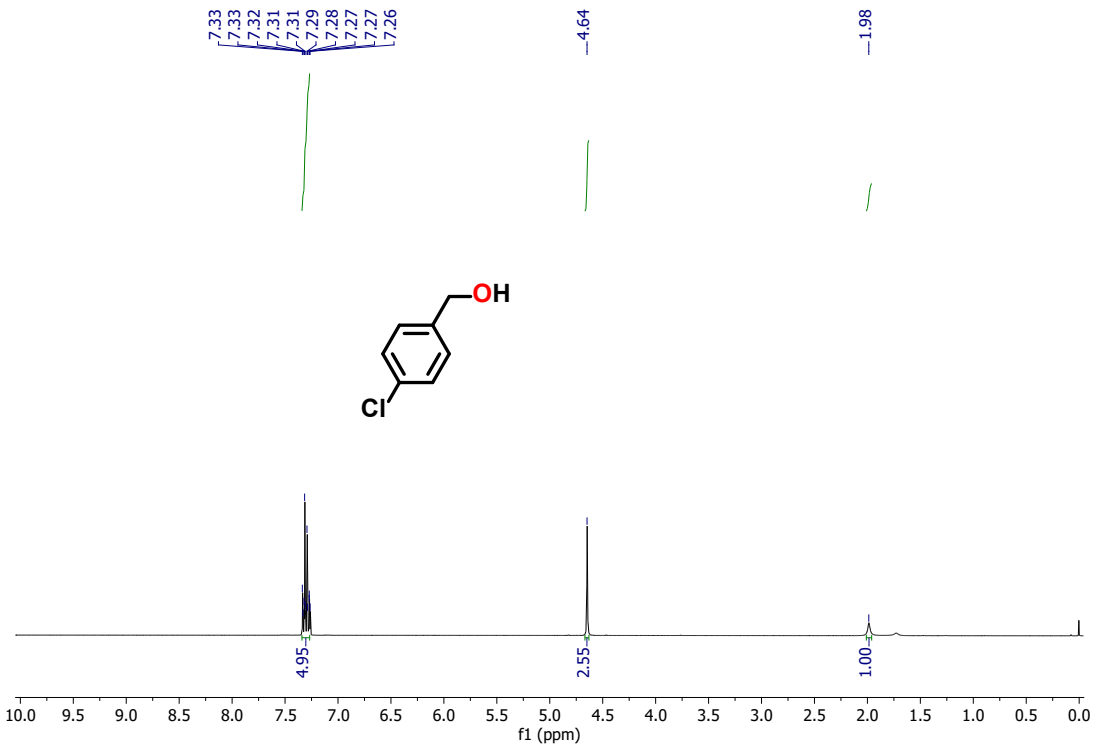


Figure S89: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6f**.

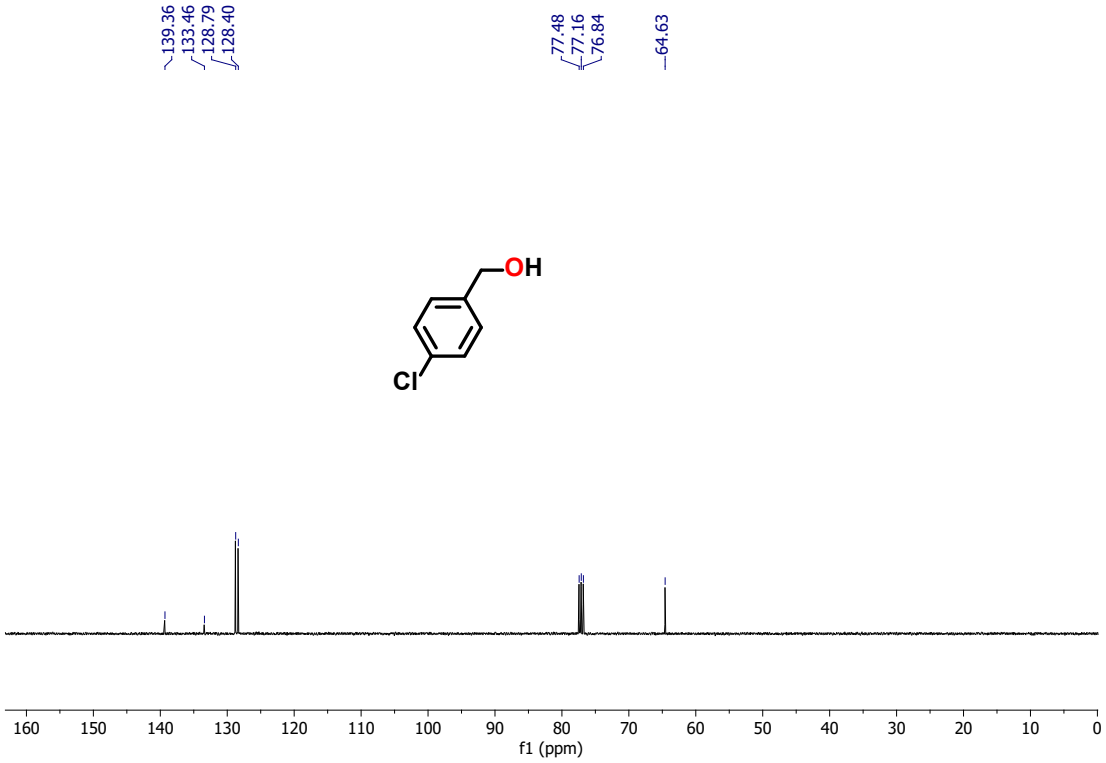


Figure S90: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6f**.

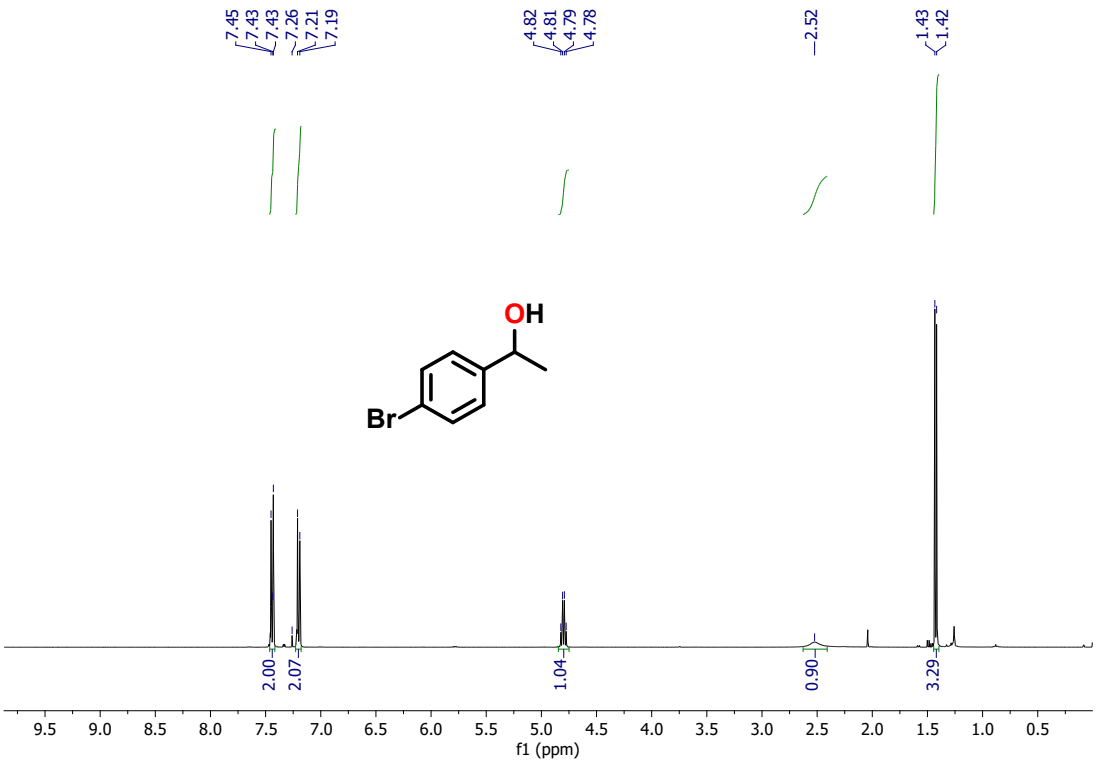


Figure S91: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6g**.

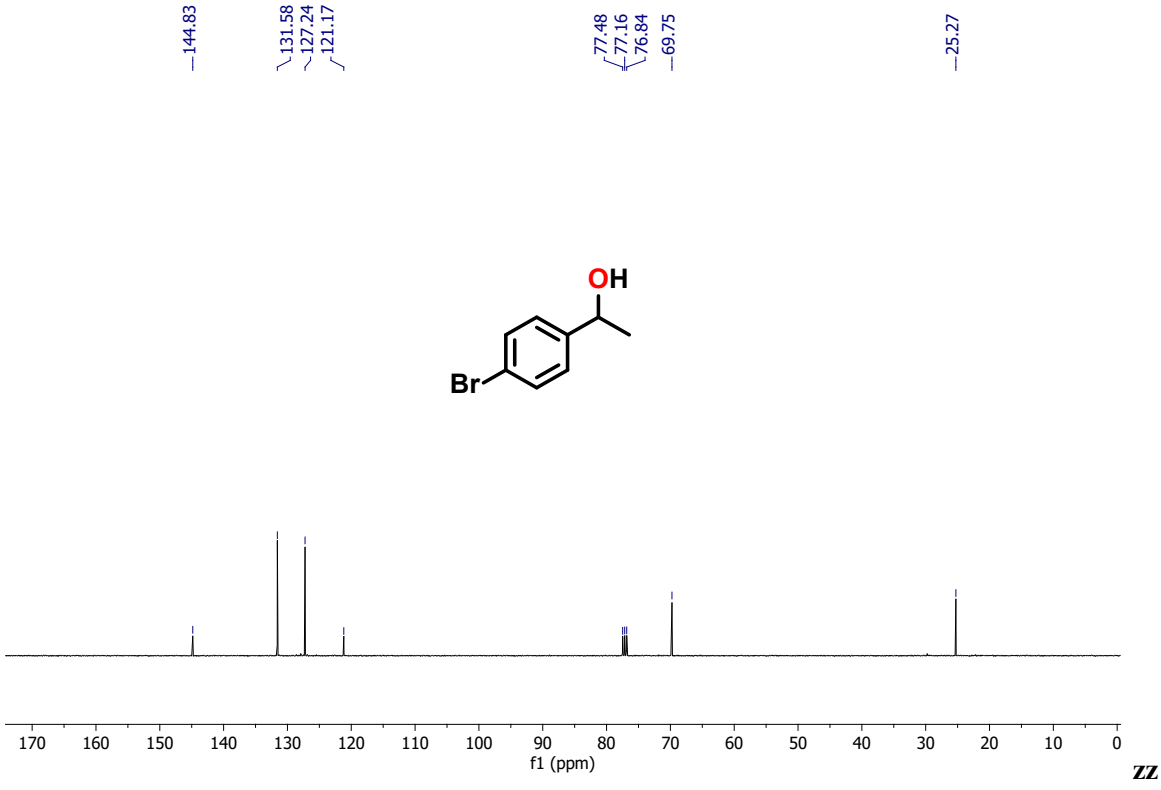


Figure S92: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6g**.

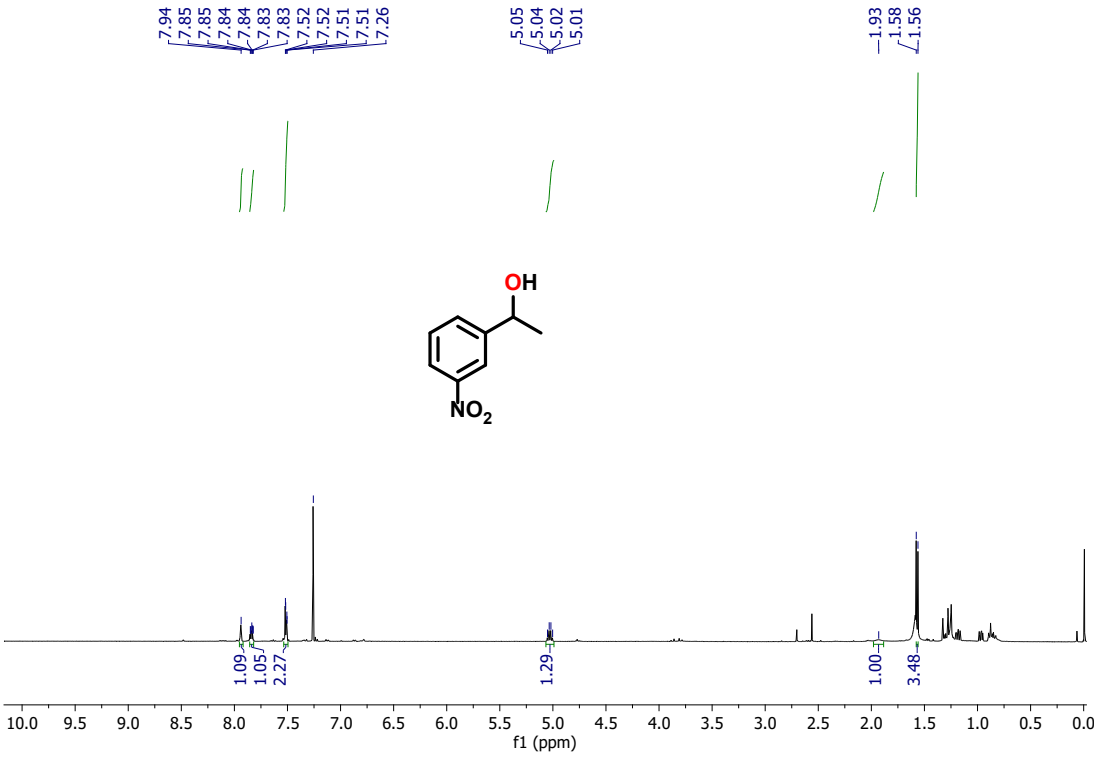


Figure S93: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6h**.

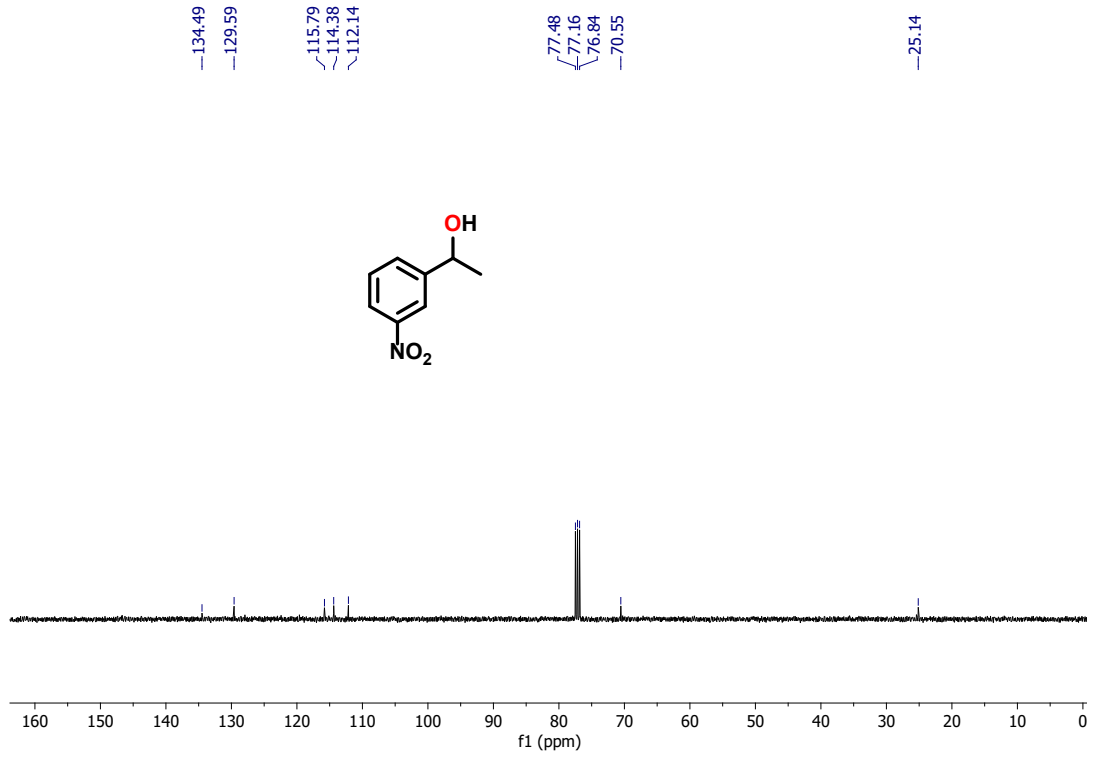


Figure S94: ^{13}C{^{1}H} NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6h**.

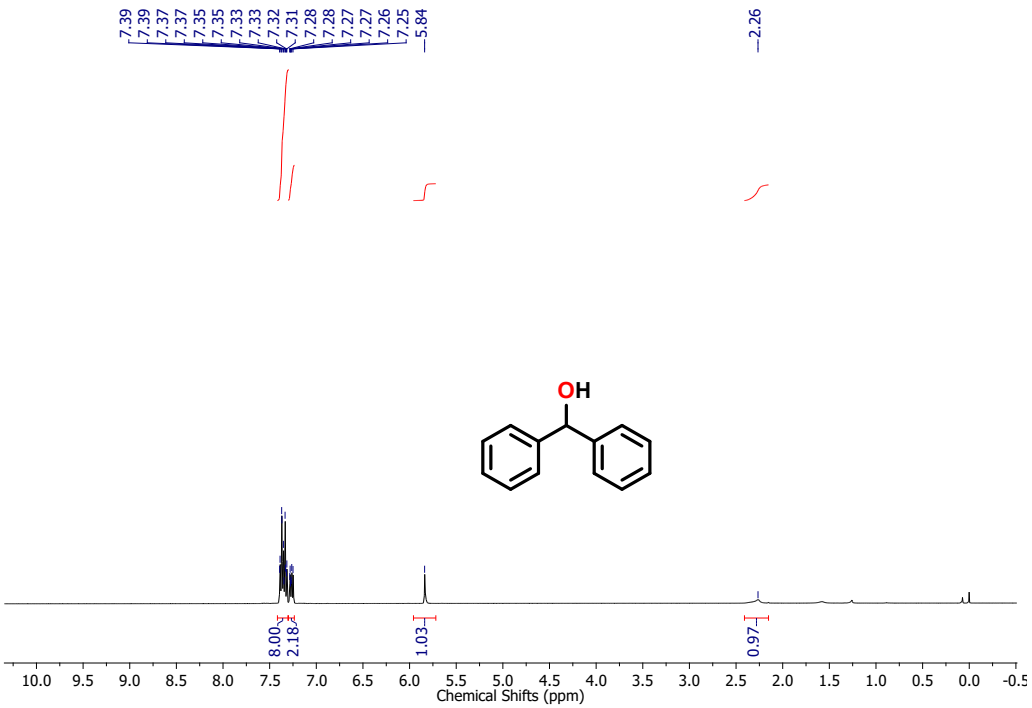


Figure S95: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6i**.

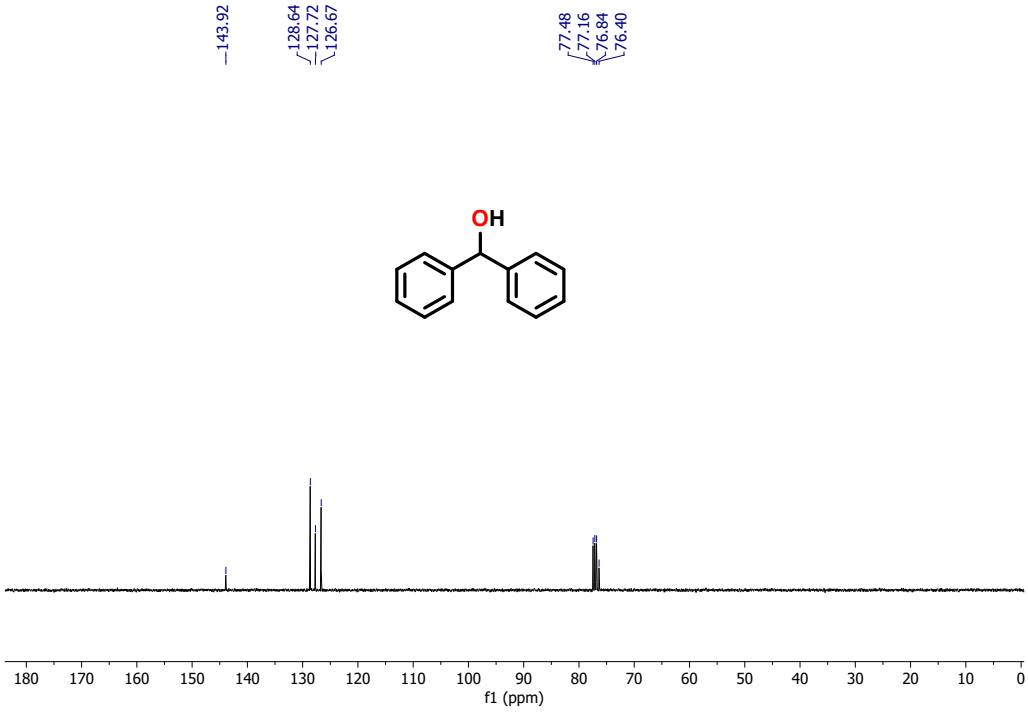


Figure S96: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6i**.

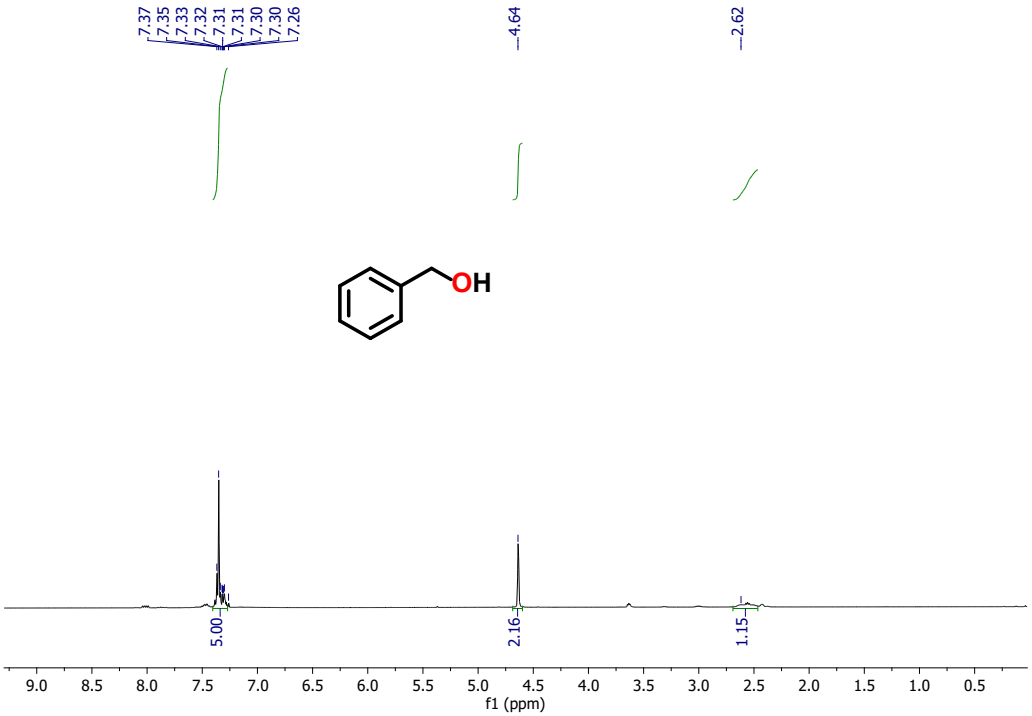


Figure S97: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **6j**.

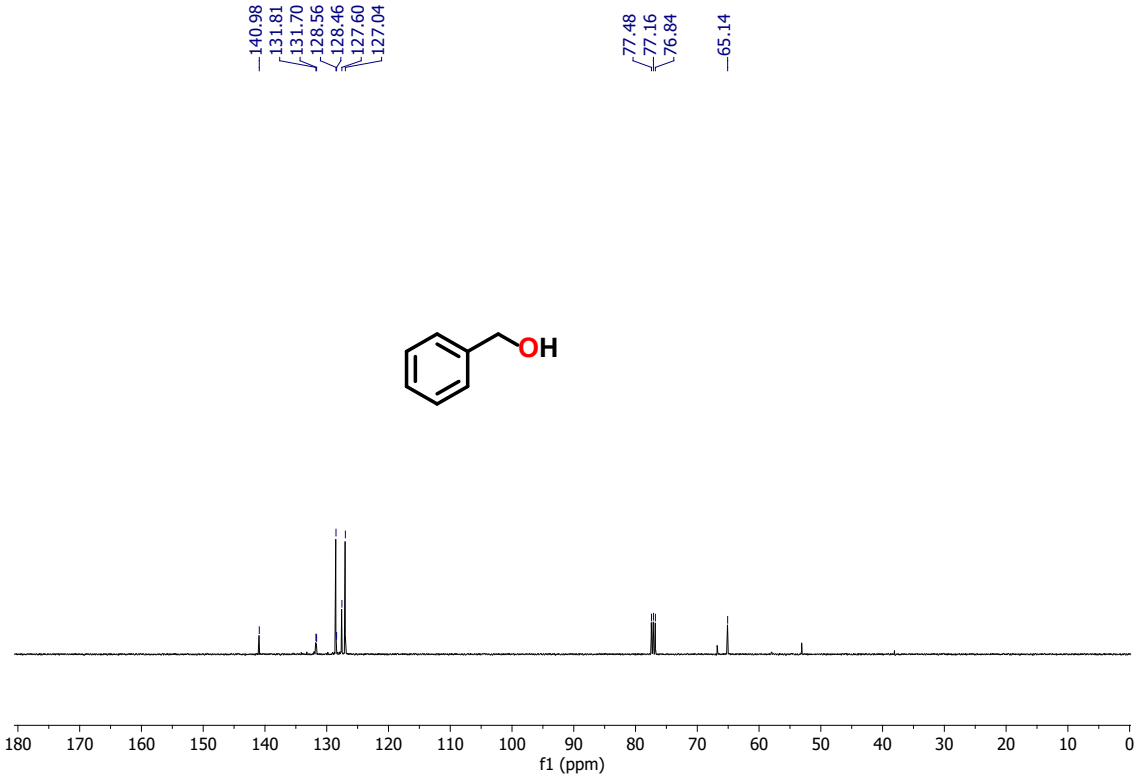


Figure S98: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **6j**.

12. Chemoselectivity

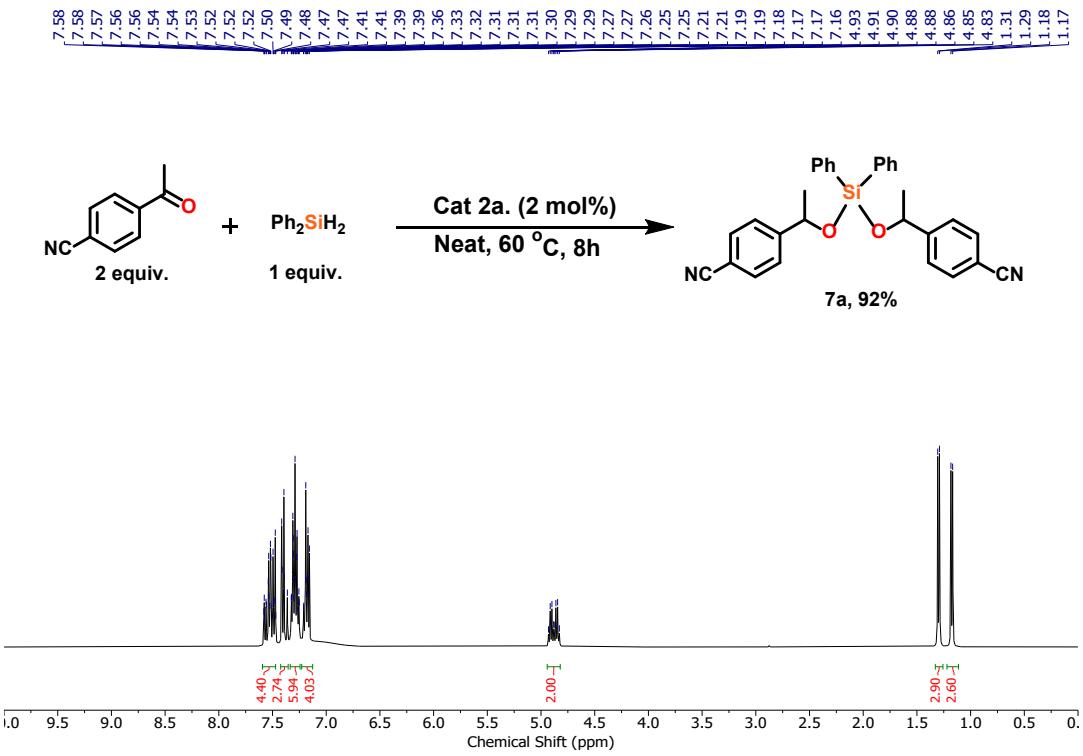


Figure S99: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **7a**.

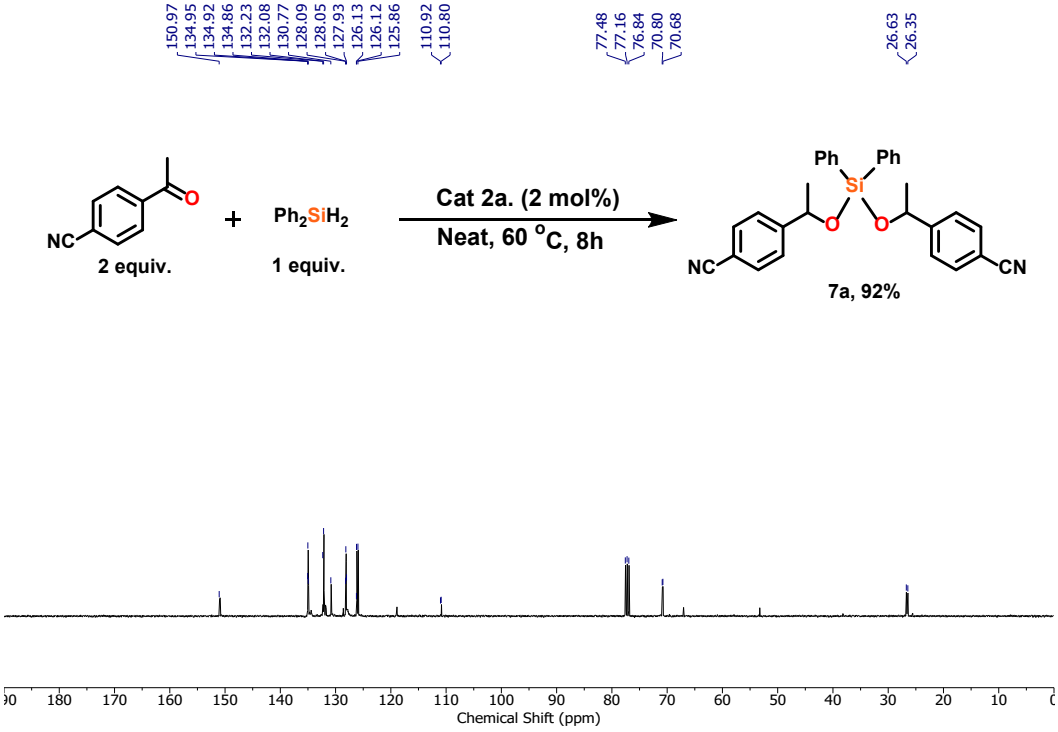


Figure S100: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **7a**.

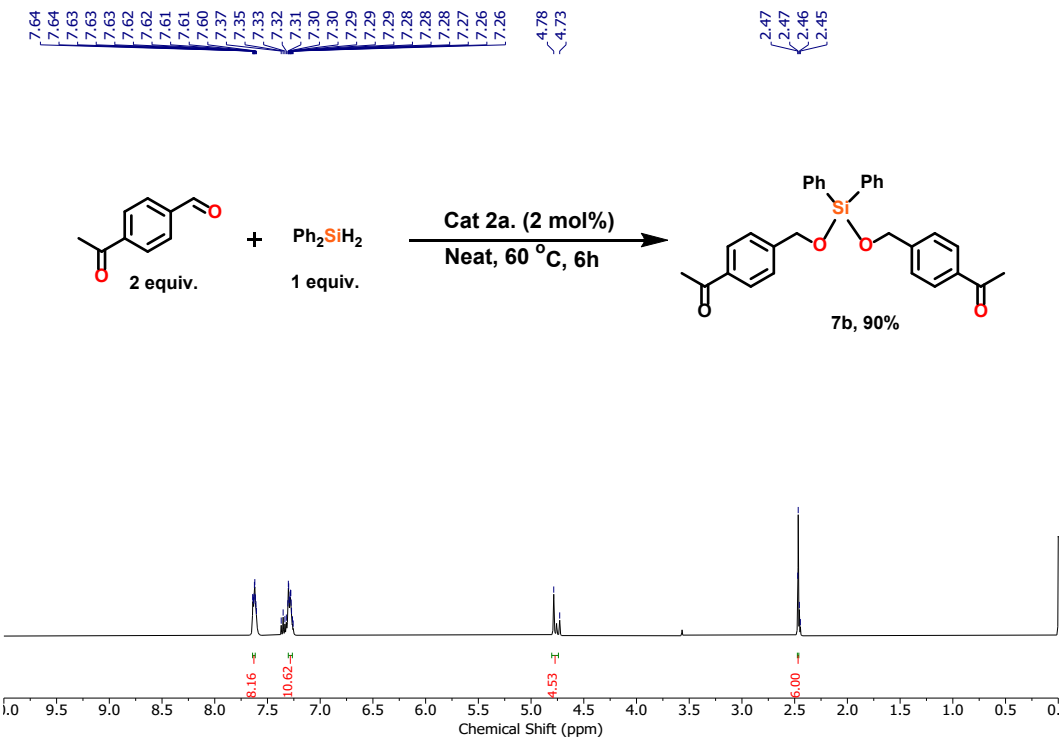


Figure S101: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **7b**.

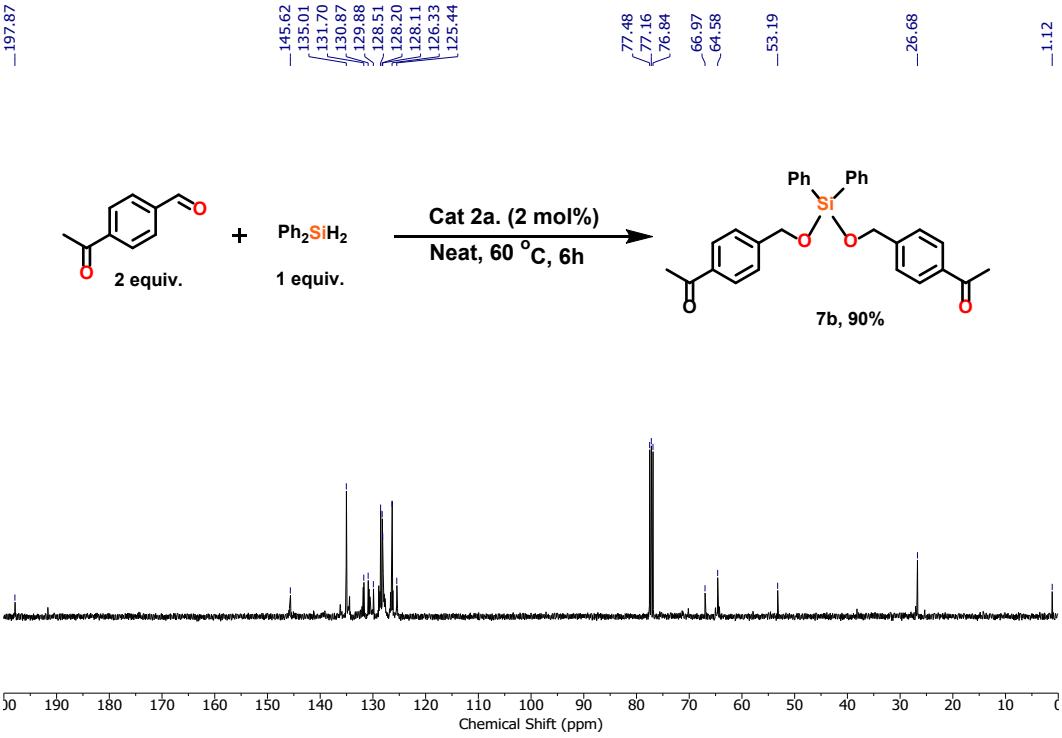


Figure S102: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **7b**.

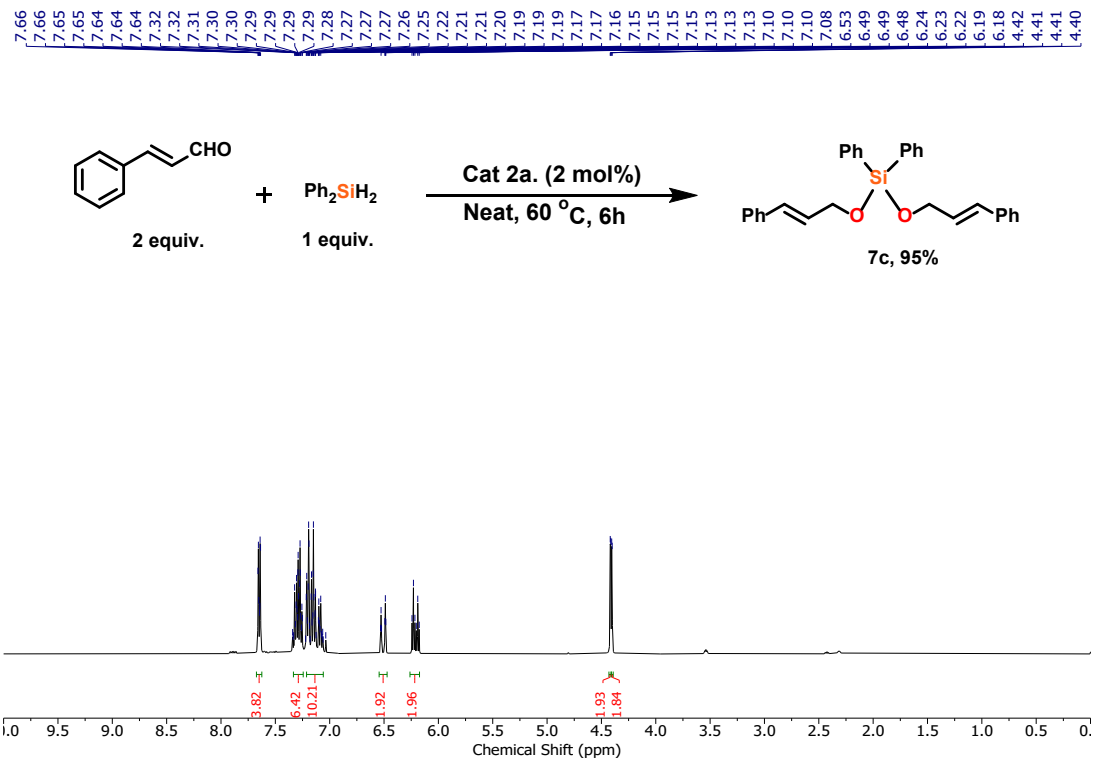


Figure S103: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of **7c**.

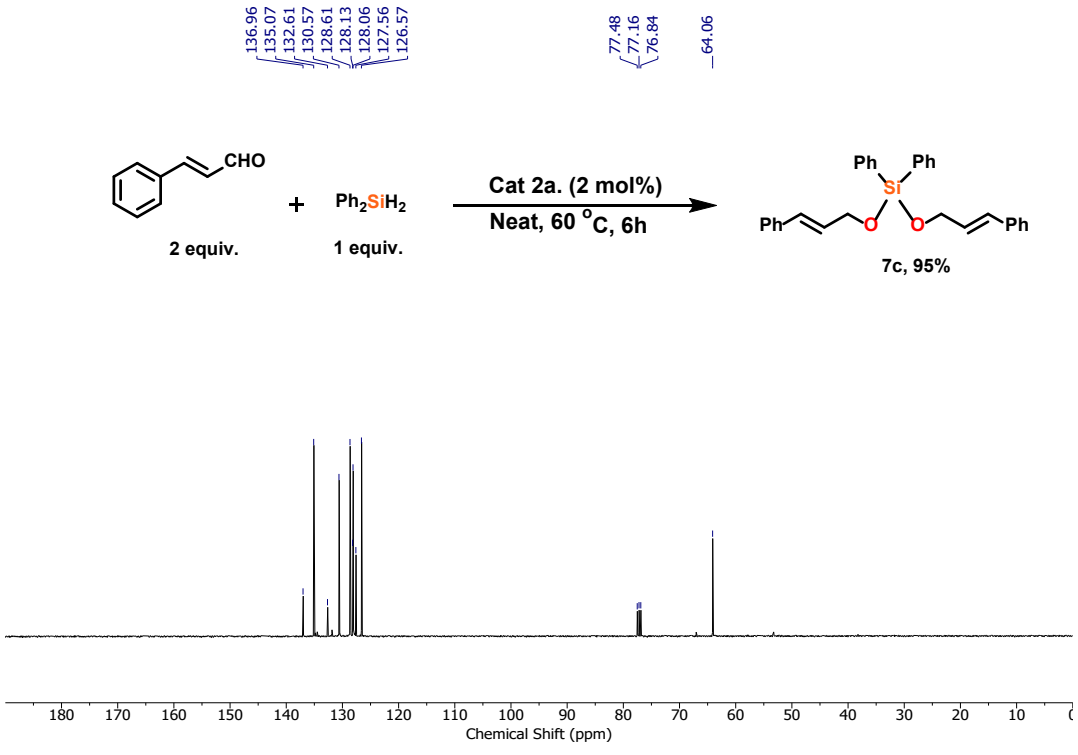


Figure S104: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of **7c**.

12. NMR study of Reaction Mechanism

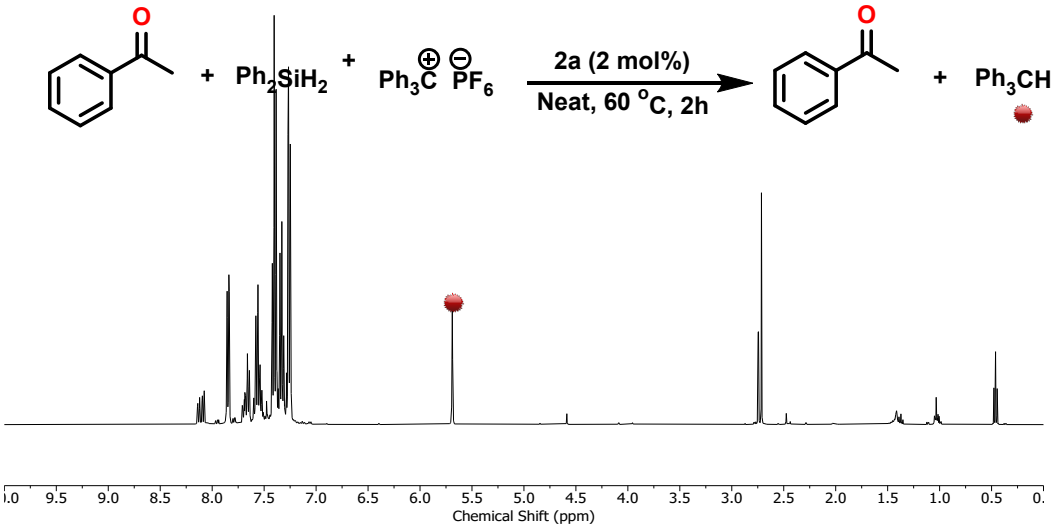


Figure S105: ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of the reaction mixture of **2a** and Ph_2SiH_2 in the presence of trityl hexafluorophosphate.

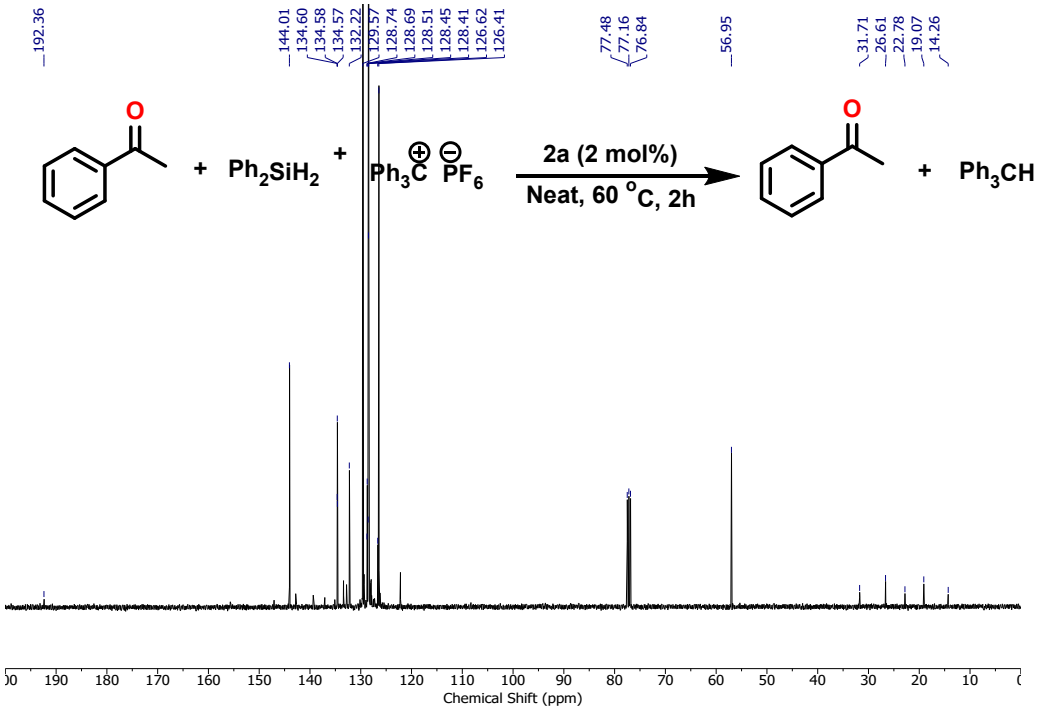


Figure S106: $^{13}C\{^{1}H\}$ NMR spectrum (100 MHz, 25 °C, $CDCl_3$) of the reaction mixture of **2a** and Ph_2SiH_2 in the presence of trityl hexafluorophosphate.

13. **References**.

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