

Supporting Information

A general ppm-level Pd-catalysed asymmetric diarylalkyne hydrosilylation to access structurally diverse Si-stereogenic vinylsilanes

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Table of Contents

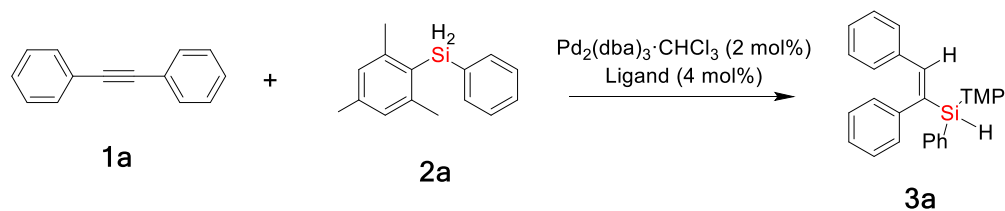
1 General Information	3
1.1 Evaluation of reaction parameters	4
2 Experimental Section	6
2.1 Preparation of substrates	6
2.2 General procedure for the synthesis of 3	9
2.3 General procedure for the synthesis of 9	9
2.3.1 Characterization of Si-stereogenic vinylsilanes 3 and 9	10
2.3.2 Synthetic applications of Si-stereogenic monohydrosilanes	31
2.4 Gram-scale synthesis of 3a	33
2.5 The effect of ligand on the configuration of product	33
2.6 Synthesis of chiral poly(vinylsilane) 5 bearing Si-stereogenic center	34
2.6.3 Preparation of polyalkyne 4	34
2.6.4 Synthesis of chiral poly(vinylsilane) 5 via Pd-catalyzed hydrosilylation	34
2.6.5 CD spectra of poly(vinylsilane) 5	35
2.6.6 SEM images of polyalkyne 4 and poly(vinylsilane) 5	36
2.6.7 TG analysis of polyalkyne 4 and poly(vinylsilane) 5	36
2.7 Kinetic studies for the Pd-catalyzed hydrosilylation of 1a and 2a	37
2.8 Experiments for non-linear effect	38
2.9 DFT calculations for the ECD spectrum of chiral product 3a	40
2.10 X-Ray structure of 9e	41
3 NMR Spectra	43
4 HPLC	87

1 General Information

Unless specifically stated, all reagents were commercially obtained and where appropriate, purified prior to use. Dichloromethane (DCM), toluene, were freshly distilled from CaH₂. Ether (Et₂O), tetrahydrofuran (THF), 1,4-dioxane and Cyclohexane were dried and distilled from metal sodium and benzophenone. Alcohol solvents were dried and distilled from metal magnesium. Other commercially available reagents and solvents were used directly without purification. Reactions were monitored by thin layer chromatography (TLC) using silica gel plates. Flash column chromatography was performed over silica (200 - 300 mesh). NMR spectra were recorded on a Bruker 400 MHz or 500 MHz (400 MHz for ¹H; 100 MHz for ¹³C, 500 MHz for ¹⁹F, 400MHz). The chemical shifts (δ , ppm) were quoted in parts per million (ppm) referenced to TMS (0.00 ppm for ¹H NMR) and CDCl₃ (77.16 ppm for ¹³C NMR) The following abbreviations were used to explain multiplicities: s = singlet, d = doublet, dd = doublets of doublet, t = triplet, q = quartet, m = multiplets. Coupling constants, J, were reported in Hertz unit (Hz). High resolution mass spectra (HRMS) of the products were obtained on a Bruker Daltonics micro TOF-spectrometer. HPLC analyses were carried out with an Agilent 1260 infinity, Waters AcQuity HPLC or Waters AcQuity UPLC using a chiralcel AD-H column, a chiralcel MD column, a chiralcel OX column, a chiralcel AS-H column and a chiral Phenomenex column.

1.1 Evaluation of reaction parameters

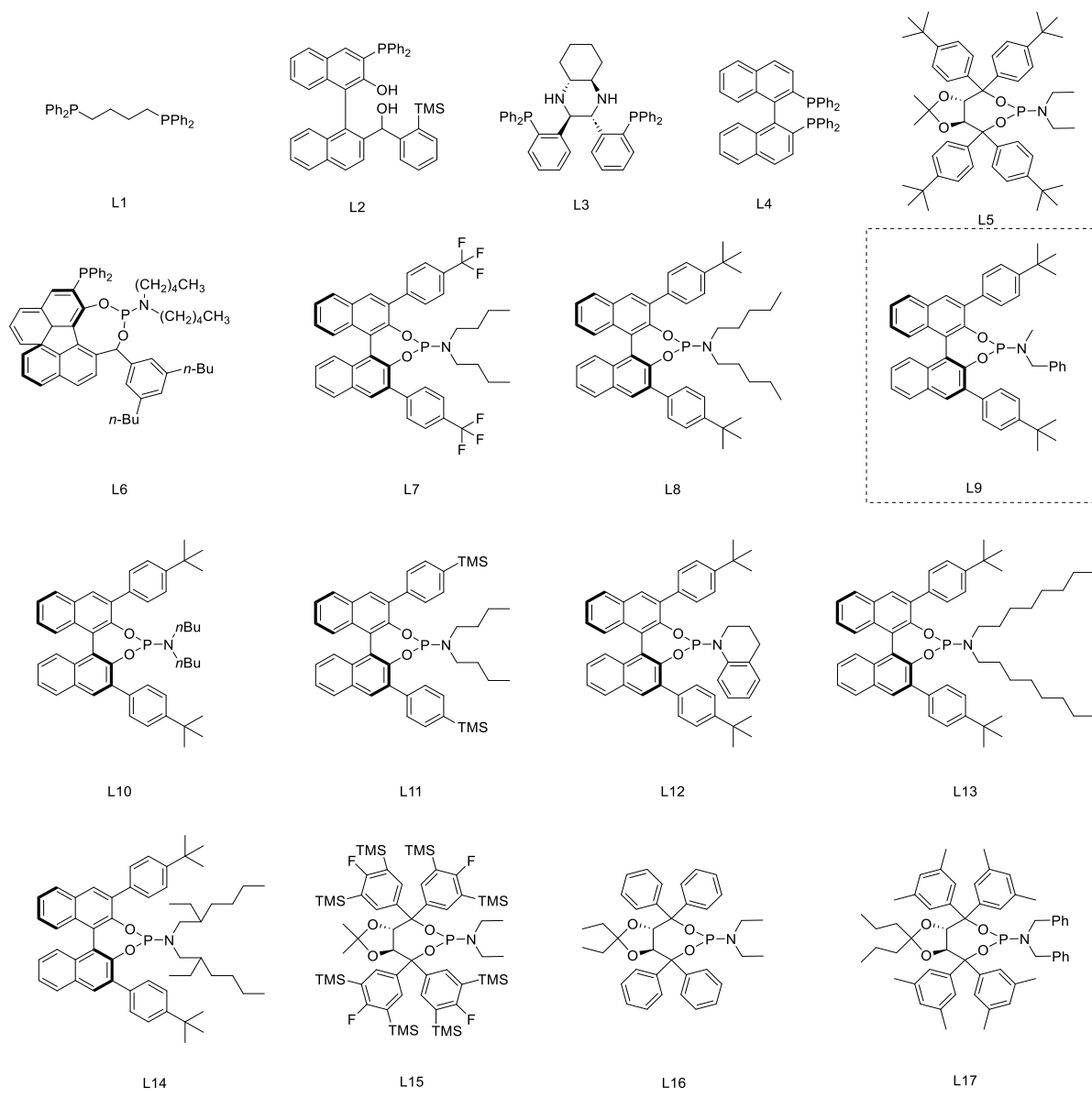
Table S1. Screening of the chiral phosphine ligands



Entry	Ligand	Solvent	T (°C)	Yield of 3a (%) ^c	ee of 3 (%) ^d
1	L1	DCM	rt	93	0
2	L2	DCM	rt	NR	
3	L3	DCM	rt	10	10
4	L4	DCM	rt	NR	
5	L5	DCM	rt	30	0
6	L6	cyclohexane	0	NR	
7	L7	cyclohexane	0	85	82
8	L8	cyclohexane	0	72	87
9	L9	cyclohexane	0	90	92
10	L10	cyclohexane	0	75	72
11	L11	cyclohexane	0	65	85
12	L12	cyclohexane	0	90	85
13	L13	cyclohexane	0	89	83
14	L14	cyclohexane	0	88	87
15	L15	DCM	0	40	23
16	L16	DCM	0	23	8
17	L14	DCM	0	Trace	-
18	L14	DCM	rt	76	90
19	L14	THF	rt	67	89

^a Unless otherwise noted, reactions were conducted under N_2 on 0.2 mmol scale: **1a** (0.2 mmol), **2a** (0.2 mmol), $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (2 mol%), ligand (8 mol%), DCM (1 mL). ^b Determined by ^1H NMR using dibromomethane as an internal standard. ^c Determined by GC-MS.

^d Determined by chiral HPLC.

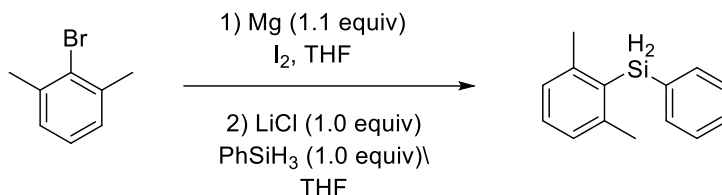


Chiral P-ligands L1-L16

2 Experimental Section

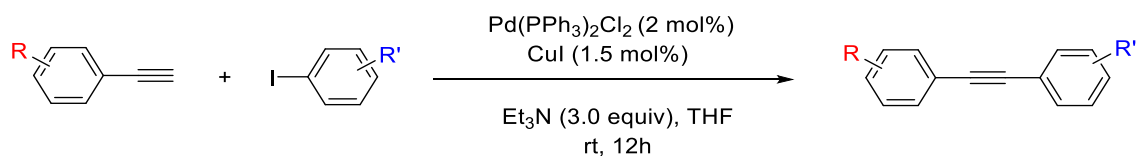
2.1 Preparation of substrates

A): General procedure for the preparation of (2,6-dimethylphenyl)(phenyl)silane from 2-bromo-1,3-dimethylbenzene



To a dried 2-neck round bottom flask equipped with a water-cooled condenser was added magnesium turnings (534.8 mg, 22.0 mmol, 1.1 equiv.), and three pieces of iodine partials, and THF (20 mL) under argon. 2-bromo-1,3-dimethylbenzene (4.5 g, 20.0 mmol, 1.0 equiv.) was added slowly over the course of 15 min to the refluxing mixture of THF and magnesium turnings. Following that, the mixture was refluxed for an additional hour. The resulting Grignard reagent was cooled to 25 °C for the following procedure. To a suspension of LiCl (847.8 mg, 20.0 mmol) in 20.0 mL of THF was added the Grignard reagent, followed by the addition of phenylsilane (2.2 g, 20.0 mmol), at room temperature under argon. After the reaction mixture was stirred in an oil bath maintained at 50 °C for 6 h, the reaction was quenched by the addition of an aqueous solution of NH₄Cl (10.0 mL) at room temperature. The resulting mixture was filtered through Celite and washed with diethyl ether (20 mL x 3). The organic phase was dried over Na₂SO₄ and concentrated in vacuum to give the crude product, which was purified by chromatography on silica gel eluting with hexane to afford the title compound (2.29 g, 54%) as colorless oil.

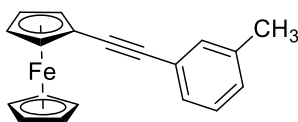
B): General procedure for the preparation of diaryl alkynes



Pd(PPh₃)₂Cl₂ (0.04 mmol, 28.04 mg, 2 mol%) catalyst was added to the

reaction tube, and CuI (0.03 mmol, 5.7 mg, 1.5 mol%) and aryl iodide (2.2 mmol, 1.1 equiv) was then added, triethylamine and tetrahydrofuran was added as solvent, the deoxidation operation was performed for 2 minutes, and triethylamine (6 mmol, 3 equiv) and aryl acetylene (2 mmol, 1 equiv) as reactants were added to the mixtures. The reaction was stirred at room temperature for 12 h. After the completion of this reaction, the solvent was removed by a rotary evaporator and the mixtures were separated by silica gel column to get the desired product.

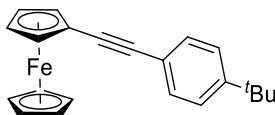
C) Characterization of ferrocenyl arylacetylene 8
cyclopenta-2,4-dien-1-yl(2-(*m*-tolylethynyl)cyclopenta-2,4-dien-1-yl)iron



8a

Reddish brown viscous oil (49.2 mg, 82% yield) purified by column chromatography (PE/EA= 100:1). ^1H NMR (400 MHz, CDCl_3): δ 7.25 – 7.03 (m, 4H), 4.42 (s, 2H), 4.17 (s, 7H), 2.27 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 138.1, 132.1, 128.7, 128.6, 128.3, 123.8, 88.0, 86.0, 71.5, 70.1, 68.9, 65.5, 21.4. HRMS (APCI-TOF) m/z : [M] calcd for $\text{C}_{19}\text{H}_{16}\text{Fe}$, 300.0601; found 300.0602. mp 116 - 118 °C.

(2-((4-(*tert*-butyl)phenyl)ethynyl)cyclopenta-2,4-dien-1-yl)(cyclopenta-2,4-dien-1-yl)iron

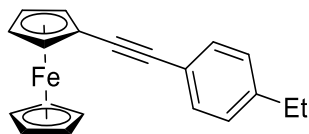


8b

Reddish brown viscous solid (470 mg, 69% yield) purified by column chromatography (PE/EA= 100:1). ^1H NMR (400 MHz, CDCl_3): δ 7.35 (d, J = 7.9 Hz, 2H), 7.27 (d, J = 8.0 Hz, 2H), 4.42 (s, 2H), 4.16 (s, 7H), 1.25 (s, 9H). ^{13}C NMR (100 MHz, CDCl_3) δ 151.0, 131.2, 125.4, 121.0, 87.6, 85.9, 71.5, 70.1, 68.8, 65.7, 34.9, 31.3. HRMS (APCI-TOF) m/z : [M] calcd for $\text{C}_{22}\text{H}_{22}\text{Fe}$, 342.1071; found 342.1063.

mp 149 - 151 °C.

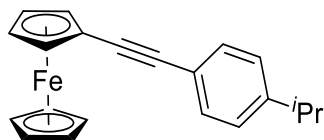
cyclopenta-2,4-dien-1-yl(2-((4-ethylphenyl)ethynyl)cyclopenta-2,4-dien-1-yl)iron



8c

Reddish brown viscous solid (310 mg, 49% yield) purified by column chromatography (PE/EA= 100:1). ¹H NMR (400 MHz, CDCl₃): δ 7.40 (d, *J* = 7.9 Hz, 2H), 7.14 (d, *J* = 8.0 Hz, 2H), 4.49 (s, 2H), 4.23 (s, 7H), 2.63 (q, *J* = 8.1 Hz, 2H), 1.23 (t, *J* = 7.9 Hz, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 144.2, 131.5, 127.97, 121.2, 87.6, 86.0, 71.5, 70.1, 68.8, 65.7, 28.9, 15.5. HRMS (APCI-TOF) *m/z*: [M] calcd for C₂₀H₁₈Fe, 314.0758; found 314.0759. mp 82 - 86 °C.

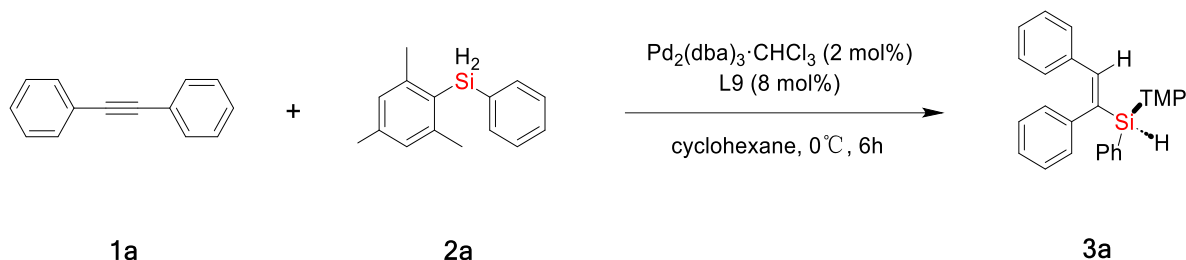
cyclopenta-2,4-dien-1-yl(2-((4-isopropylphenyl)ethynyl)cyclopenta-2,4-dien-1-yl)iron



8d

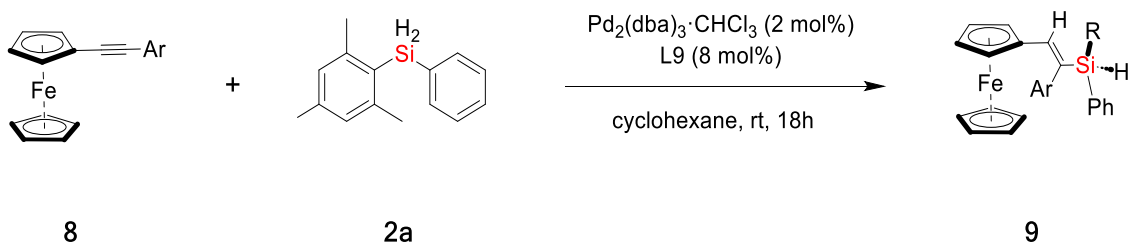
Reddish brown viscous solid (510 mg, 78% yield) purified by column chromatography (PE/EA= 100:1). ¹H NMR (400 MHz, CDCl₃): δ 7.41 (d, *J* = 8.2 Hz, 2H), 7.17 (d, *J* = 8.2 Hz, 2H), 4.49 (s, 2H), 4.24 – 4.21 (m, 7H), 2.90 (dt, *J* = 13.7, 6.9 Hz, 1H), 1.24 (d, *J* = 7 Hz, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 148.8, 131.5, 126.6, 121.4, 87.6, 86.00, 71.5, 70.1, 68.8, 65.7, 34.2, 24.0. HRMS (APCI-TOF) *m/z*: [M] calcd for C₂₁H₂₀Fe, 328.0914; found 328.0921. mp 108- 110 °C.

2.2 General procedure for the synthesis of 3



In a flame dried Schlenk tube, $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (4.1 mg, 0.004 mmol, 2 mol%), **L9** (11.2 mg, 0.016 mmol, 8 mol%) in cyclohexane (1 mL, 0.2 M) was stirred at room temperature for 30 min under nitrogen atmosphere. Then diaryl alkyne **1a** (0.2 mmol, 1 equiv), dihydrosilanes were added sequentially to the reaction mixture, and the reaction tube was cooled at 0 °C and then stirred for 6 h. After completion of the reaction, the mixture was passed through a short celite pad using DCM as a solvent. The mixture was then concentrated in vacuo and purified by column chromatography using PE and EA (70:1) to give the desired product **3a** in good yields.

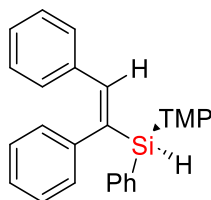
2.3 General procedure for the synthesis of 9



In a flame dried Schlenk tube, $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (4.1 mg, 0.004 mmol, 2 mol%), **L9** (11.2 mg, 0.016 mmol, 8 mol%) in cyclohexane (1 mL, 0.2 M) was stirred at room temperature for 30 min under nitrogen atmosphere. Then ferrocenyl arylacetylene **8** (0.2 mmol, 1 equiv), dihydrosilanes were added sequentially to the reaction mixture, and the reaction tube was cooled at r.t and then stirred for 18 h. After completion of the reaction, the mixture was passed through a short celite pad using DCM as a solvent. The mixture was then concentrated in vacuo and purified by column chromatography using PE and EA (70:1) to give the desired product **9** in good yields.

2.3.1 Characterization of Si-stereogenic vinylsilanes **3** and **9**

(*S,E*)-(1,2-diphenylvinyl)(mesityl)(phenyl)silane

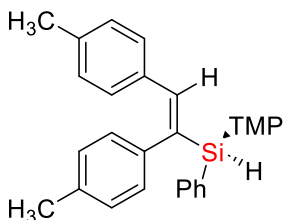


3a

3a was synthesized following the general procedure C. Yellow liquid (69.3 mg, 90% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 58.1$ ($c = 1.04$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.42 (d, $J = 6.8$ Hz, 2H), 7.17 - 7.11 (m, 3H), 7.06 - 6.95 (m, 5H), 6.91 - 6.90 (m, 6H), 6.70 (s, 2H), 5.49 (s, 1H), 2.21 (s, 6H), 2.12 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.7, 142.2, 140.4, 139.9, 137.3, 135.8, 134.1, 129.7, 129.7, 129.5, 128.9, 128.8, 128.19, 128.16, 128.1, 128.0, 127.5, 126.7, 126.3, 24.6, 21.4. HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{29}\text{H}_{28}\text{Si}$, 455.2156; found 455.2162;

HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4 mL/min, 254 nm, 92% ee). $t_R = 7.83$ min (minor), $t_R = 10.33$ min (major).

(*S, E*)-(1,2-*di-p*-tolylvinyl)(mesityl)(phenyl)silane



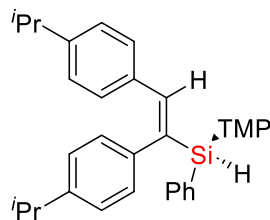
3b

3b was synthesized following the general procedure C. Yellow liquid (69.3 mg, 80% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 64.6$ ($c = 1.075$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.50 (d, $J = 1.5$ Hz, 1H), 7.50 (d, $J = 1.5$ Hz, 1H), 7.34 - 7.16 (m, 3H), 7.00 - 6.95 (, 4H), 6.89 - 6.84 (m, 5H), 6.81 (s, 2H), 5.49 (s, 1H), 2.29 (s, 6H) 2.60 (s, 3H), 2.26 (s, 3H), 2.20 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.7, 142.0, 139.8, 139.3, 139.1, 137.3, 135.9, 135.7, 134.7, 134.4, 129.6,

129.5, 129.4, 128.9, 128.7, 128.0, 127.9, 127.1, 24.6, 21.4, 21.3, 21.3. HRMS (APCI-TOF) m/z : $[M + Na]^+$ calcd for $C_{31}H_{32}Si$, 455.2156; found 455.2162;

HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4 mL/min, 254 nm, 92% ee). $t_R = 7.83$ min (minor), $t_R = 10.33$ min (major).

(S, E)-(1,2-bis(4-isopropylphenyl)vinyl)(mesityl)(phenyl)silane

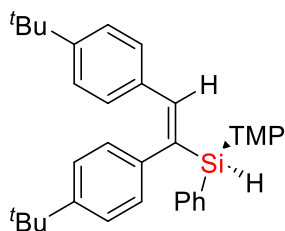


3c

3c was synthesized following the general procedure C. Clear viscous liquid (71.8 mg, 74% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 38.0$ ($c = 1.47$, $CHCl_3$). 1H NMR (400 MHz, $CDCl_3$): δ 7.55 (d, $J = 1.6$ Hz, 1H), 7.53 (d, $J = 1.6$ Hz, 1H), 7.37 - 7.31 (m, 3H), 7.12 - 7.05 (m, 4H), 7.00 - 6.94 (m, 4H), 6.90 (s, 1H), 6.87 (s, 2H), 5.54 (s, 1H), 2.91 (dt, $J = 29.1, 6.4$ Hz, 2H), 2.33 (s, 6H), 2.32 (s, 3H), 1.25 (d, $J = 6.8$ Hz, 6H), 1.19 (d, $J = 7.0$ Hz, 6H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 148.3, 146.8, 145.8, 141.9, 139.8, 139.7, 139.2, 135.9, 135.0, 134.6, 129.8, 129.3, 128.9, 128.0, 127.9, 127.1, 126.9, 126.1, 33.93, 33.86, 24.19, 24.16, 24.2, 23.97, 23.95, 21.4. HRMS (APCI-TOF) m/z : $[M + K]^+$ calcd for $C_{35}H_{40}Si$, 527.2531; found 527.2530;

HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4mL/min, 254 nm, 94% ee). $t_R = 5.13$ min (minor), $t_R = 7.50$ min (major).

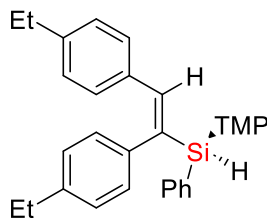
(S, E)-(1,2-bis(4-(tert-butyl)phenyl)vinyl)(mesityl)(phenyl)silane



3d

3d was synthesized following the general procedure C. Yellow liquid (70.0 mg, 62% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 6.8$ ($c = 0.85$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.44 (d, $J = 1.5$ Hz, 1H), 7.42 (d, $J = 1.6$ Hz, 1H), 7.27 - 7.15 (m, 5H), 7.03 (d, $J = 8.2$ Hz, 2H), 6.97 (d, $J = 8.4$ Hz, 2H), 6.86 (d, $J = 8.2$ Hz, 2H), 6.80 (s, 1H), 6.76 (s, 2H), 5.44 (s, 1H), 2.22 (s, 6H), 2.20 (s, 3H), 1.22 (s, 9H), 1.15 (s, 9H). $^{13}\text{CNMR}$ (100 MHz, CDCl_3) δ 150.6, 149.1, 145.8, 141.7, 139.8, 139.3, 139.2, 135.9, 134.63, 134.62, 129.5, 129.3, 128.9, 128.0, 127.5, 127.1, 125.7, 125.0, 34.7, 34.58, 31.55, 31.3, 24.6, 21.4. HRMS (APCI-TOF) m/z : $[\text{M} + \text{K}]^+$ calcd for $\text{C}_{37}\text{H}_{44}\text{Si}$, 555.2844; found 555.2825; HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4mL/min, 254 nm, 91% ee). $t_R = 6.70$ min (minor), $t_R = 8.84$ min (major).

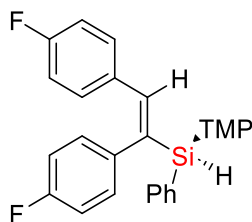
(S, E)-(1,2-bis(4-ethylphenyl)vinyl)(mesityl)(phenyl)silane



3e

3e was synthesized following the general procedure C. Clear viscous liquid (49.6 mg, 54% yield). purified by column chromatography (PE/EA= 80:1) $[\alpha]_D^{20} = 42.1$ ($c = 0.28$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.57 (d, $J = 1.6$ Hz, 1H), 7.55 (d, $J = 1.6$ Hz, 1H), 7.36 - 7.33 (m, 3H), 7.08 (s, 4H), 6.96 (s, 4H), 6.92 (s, 1H), 6.88 (s, 2H), 5.57 (s, 1H), 2.63 (q, $J = 7.6$ Hz, 2H), 2.56 (q, $J = 7.6$ Hz, 2H), 2.36 (s, 6H) 2.32 (s, 3H), 1.26 (t, $J = 7.6$ Hz, 3H), 1.20 (t, $J = 7.6$ Hz, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.8, 143.7, 142.1, 142.0, 139.8, 139.5, 139.2, 135.9, 135.0, 134.6, 129.8, 129.3, 128.9, 128.3, 128.01, 127.97, 127.6, 127.2, 28.67, 28.65, 24.6, 21.4, 15.6, 15.4. HRMS (APCI-TOF) m/z : $[\text{M} + \text{K}]^+$ calcd for $\text{C}_{33}\text{H}_{36}\text{Si}$, 499.2218; found 499.2213; HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.6mL/min, 254 nm, 91% ee). $t_R = 5.95$ min (minor), $t_R = 6.57$ min (major).

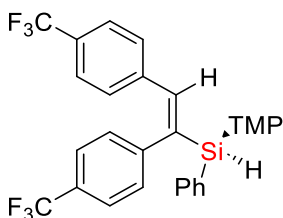
(S, E)-(1,2-bis(4-fluorophenyl)vinyl)(mesityl)(phenyl)silane



3f

3f was synthesized following the general procedure C. Yellow liquid (86.4 mg, 97% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_{\text{D}}^{20} = 36.3$ ($c = 0.65$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.43 (d, $J = 1.6$ Hz, 1H), 7.42 (d, $J = 1.2$ Hz, 1H) 7.29 - 7.20 (m, 3H), 6.97-6.93 (m, 2H), 6.86-6.80 (m, 5H), 6.76 (s, 2H), 6.70 (dd, $J = 8.8$ Hz, 2H), 5.47 (s, 1H), 2.24 (s, 6H), 2.19 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 162.0 (d, $J_{\text{C-F}} = 247$ Hz), 161.7 (d, $J_{\text{C-F}} = 244$ Hz), 145.66, 141.37, 140.18, 139.11 (d, $J_{\text{C-F}} = 1$ Hz), 137.69 (d, $J_{\text{C-F}} = 3$ Hz), 135.84, 133.72 (d, $J_{\text{C-F}} = 4$ Hz), 133.29, 133.25, 131.4 (d, $J_{\text{C-F}} = 8$ Hz), 131.29, 129.71, 129.64 (d, $J_{\text{C-F}} = 2$ Hz), 129.62, 129.01, 128.17, 126.33, 115.9 (d, $J_{\text{C-F}} = 21$ Hz), 115.1 (d, $J_{\text{C-F}} = 21$ Hz), 24.60, 21.35. $^{19}\text{F NMR}$ (500 MHz, CDCl_3) δ -113.42, -116.09. HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{29}\text{H}_{26}\text{F}_2\text{Si}$, 463.1664; found 463.1644; HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4 mL/min, 254 nm, 91% ee). $t_{\text{R}} = 6.86$ min (minor), $t_{\text{R}} = 7.69$ min (major).

(S, E)-1,2-bis(4-(trifluoromethyl)phenyl)vinyl(mesityl)(phenyl)silane



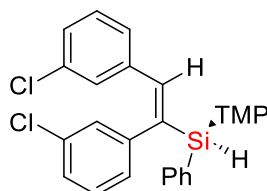
3g

3g was synthesized following the general procedure C. Yellow liquid (102.6 mg, 95% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_{\text{D}}^{20} = 46.1$ ($c = 0.43$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.45 (d, $J = 1.2$ Hz, 1H), 7.44 (d, $J = 2.0$ Hz, 1H) 7.39 (d, $J = 8.2$ Hz, 2H), 7.29 - 7.22 (m, 5H), 7.09 (d, $J = 8.0$ Hz, 2H), 6.95 (m, 3H), 6.79 (s, 2H), 5.49 (s, 1H), 2.26 (s, 6H), 2.21 (s, 3H). $^{13}\text{C NMR}$ (100 MHz,

CDCl₃) δ 145.7, 145.6, 142.7, 141.5, 140.6, 140.2, 135.9, 132.9, 130.0, 129.8, 129.2, 128.3 (d, J_{C-F} = 2 Hz), 126.8 (q, J_{C-F} = 270 Hz), 125.5 (q, J_{C-F} = 270 Hz), 125.9 (q, J_{C-F} = 4 Hz), 125.7, 125.2 (q, J_{C-F} = 4 Hz), 24.6, 21.4. ¹⁹F NMR (500 MHz, CDCl₃) δ -62.32, -62.64. HRMS (APCI-TOF) m/z: [M + K]⁺ calcd for C₃₁H₂₆F₆Si, 579.1340; found 579.1323

HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.6mL/min, 254 nm, 91% ee). t_R = 9.65 min (minor), t_R = 11.60 min (major).

(S, E)-(1,2-bis(3-chlorophenyl)vinyl)(mesityl)(phenyl)silane

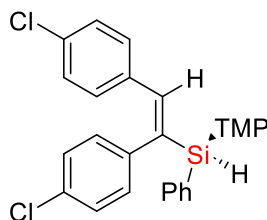


3h

3h was synthesized following the general procedure C. Yellow liquid (79.3 mg, 84% yield). purified by column chromatography (PE/EA= 80:1). [α]_D²⁰ = 34.6 (c = 0.925, CHCl₃). ¹H NMR (400 MHz, CDCl₃): δ 7.45 (d, J = 1.6 Hz, 1H), 7.43 (d, J = 1.6 Hz, 1H) 7.38 (d, J = 8.2 Hz, 2H), 7.30 - 7.22 (m, 5H), 7.09 (d, J = 8.0 Hz, 2H), 6.95 - 6.93 (m, 3H), 6.78 (s, 2H), 5.49 (s, 1H), 2.25 (s, 6H), 2.20 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 145.7, 145.6, 142.7, 141.5, 140.6, 140.2, 135.9, 132.9, 130.0, 129.7, 129.2, 128.34, 128.32, 125.98, 125.95, 125.9, 125.87, 125.6, 125.24, 125.20, 125.2, 125.1, 24.6, 21.4. HRMS (APCI-TOF) m/z: [M +]⁺ calcd for C₂₉H₂₆Cl₂Si, 472.1175; found 472.1800;

HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4mL/min, 254 nm, 92% ee). t_R = 9.76 min (minor), t_R = 12.74 min (major).

(S, E)-(1,2-bis(4-chlorophenyl)vinyl)(mesityl)(phenyl)silane

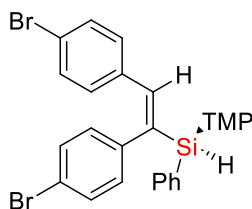


3i

3i was synthesized following the general procedure C. Yellow liquid (84.1 mg, 89% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 37.2$ ($c = 1.65$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.43 (d, $J = 1.6$ Hz, 1H), 7.42 (d, $J = 2.0$ Hz, 1H), 7.29 - 7.22 (m, 3H), 7.10 (d, $J = 8.2$ Hz, 2H), 6.99 (d, $J = 8.2$ Hz, 2H), 6.92 (d, $J = 8.2$ Hz, 2H), 6.82 - 6.80 (m, 3H), 6.77 (s, 2H), 5.46 (s, 1H), 2.24 (s, 6H), 2.20 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.6, 141.3, 140.28, 140.25, 140.2, 135.9, 135.4, 133.4, 132.4, 130.9, 129.7, 129.4, 129.2, 129.1, 128.4, 128.2, 126.1, 26.4, 21.4. HRMS (APCI-TOF) m/z : $[\text{M} + \text{K}]^+$ calcd for $\text{C}_{29}\text{H}_{26}\text{Cl}_2\text{Si}$, 511.0812; found 511.0813;

HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4mL/min, 254 nm, 91% ee). $t_R = 8.59$ min (minor), $t_R = 10.36$ min (major).

(S, E)-1,2-bis(4-bromophenyl)vinyl(mesityl)(phenyl)silane

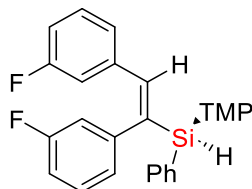


3j

3j was synthesized following the general procedure C. Yellow liquid (44.8 mg, 42% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 12.4$ ($c = 0.78$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.44 (d, $J = 1.6$ Hz, 1H), 7.42 (d, $J = 1.6$ Hz, 1H) 7.30 - 7.23 (m, 5H), 7.17 - 7.15 (m, 2H), 6.87 (d, $J = 8.2$ Hz, 2H), 6.79 (m, 3H), 6.74 (d, $J = 8.2$ Hz, 2H), 5.45 (s, 1H), 2.25 (s, 6H), 2.21 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.6, 141.3, 140.7, 140.4, 140.3, 135.8, 133.3, 132.2, 131.3, 131.1, 129.8, 129.7, 129.1, 128.2, 126.0, 121.7, 120.5, 24.6, 21.4. HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{29}\text{H}_{26}\text{Br}_2\text{Si}$, 583.0063; found 583.0060.

HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4mL/min, 254 nm, 90% ee). $t_R = 7.85$ min (minor), $t_R = 9.81$ min (major).

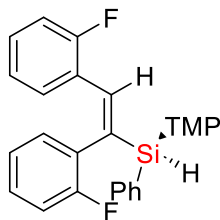
(S, E)-(1,2-bis(3-fluorophenyl)vinyl)(mesityl)(phenyl)silane



3k

3k was synthesized following the general procedure C. Yellow liquid (79.2 mg, 90% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 46.1$ ($c = 0.53$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.44 (d, $J = 1.6$ Hz, 1H), 7.42 (d, $J = 1.6$ Hz, 1H), 7.29 - 7.21 (m, 3H), 7.11 - 7.06 (m, 1H) 7.00 - 6.95 (m, 1H) 6.83 (s, 1H), 6.79 - 6.65 (m, 7H), 6.56 (d, $J = 12.2$ Hz, 1H), 5.48 (s, 1H), 2.25 (s, 6H), 2.20 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 163.2 (d, $J_{\text{C-F}} = 245$ Hz), 162.5 (d, $J_{\text{C-F}} = 244$ Hz) 145.7, 144.0 (d, $J_{\text{C-F}} = 7$ Hz), 141.4 (d, $J_{\text{C-F}} = 2$ Hz), 141.2 (d, $J_{\text{C-F}} = 2$ Hz), 140.3, 139.1 (d, $J_{\text{C-F}} = 7$ Hz), 135.8, 133.3, 130.4 (d, $J_{\text{C-F}} = 9$ Hz), 129.8, 129.6 (d, $J_{\text{C-F}} = 8$ Hz), 129.1, 128.2, 125.9, 125.4 (d, $J_{\text{C-F}} = 2$ Hz), 123.7 (d, $J_{\text{C-F}} = 3$ Hz), 116.1 (d, $J_{\text{C-F}} = 22$ Hz), 114.7 (d, $J_{\text{C-F}} = 44$ Hz), 114.7 (d, $J_{\text{C-F}} = 2$ Hz), 114.5, 113.5 (d, $J_{\text{C-F}} = 21$ Hz), 24.6, 21.4. $^{19}\text{F NMR}$ (500 MHz, CDCl_3) δ -112.58, -113.29. HRMS (APCI-TOF) m/z : $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{29}\text{H}_{26}\text{F}_2\text{Si}$, 463.1664; found 463.1125; HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.6mL/min, 254 nm, 88% ee). $t_R = 7.67$ min (minor), $t_R = 9.86$ min (major).

(S, E)-(1,2-bis(2-fluorophenyl)vinyl)(mesityl)(phenyl)silane

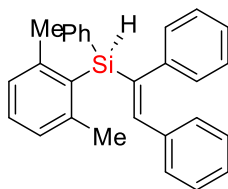


3l

3l was synthesized following the general procedure C. Yellow liquid (86.2 mg,

98% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 60.4$ ($c = 0.75$, CHCl_3) $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.47 (d, $J = 1.6\text{Hz}$, 1H), 7.46 (d, $J = 1.6\text{Hz}$, 1H), 7.25 - 7.18 (m, 4H), 7.00 - 6.98 (m, 3H), 6.89 - 6.74 (m, 6H), 6.68 (dd, $J = 8.2\text{Hz}$, 1H), 5.54 (s, 1H), 2.28 (s, 6H), 2.19 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 160.5 (d, $J_{\text{C-F}} = 247\text{ Hz}$), 158.9 (d, $J_{\text{C-F}} = 247\text{ Hz}$), 145.8, 140.2, 137.2 (d, $J_{\text{C-F}} = 4\text{ Hz}$), 135.8, 133.6, 130.2 (d, $J_{\text{C-F}} = 4\text{ Hz}$), 129.6, 129.5 (d, $J_{\text{C-F}} = 3\text{ Hz}$), 129.3 (d, $J_{\text{C-F}} = 8\text{ Hz}$), 128.9, 128.4 (d, $J_{\text{C-F}} = 7\text{ Hz}$), 128.1, 126.2, 125.6 (d, $J_{\text{C-F}} = 13\text{ Hz}$), 124.3 (d, $J_{\text{C-F}} = 3\text{ Hz}$), 123.5 (d, $J_{\text{C-F}} = 4\text{ Hz}$), 115.7 (d, $J_{\text{C-F}} = 47\text{ Hz}$), 115.5 (d, $J_{\text{C-F}} = 47\text{ Hz}$), 24.6, 21.3. $^{19}\text{F NMR}$ (500 MHz, CDCl_3) δ -113.31, -115.55 (m). HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{29}\text{H}_{26}\text{F}_2\text{Si}$, 463.1664; found 463.1648; HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.6mL/min, 254 nm, 92% ee). $t_R = 10.07\text{ min}$ (minor), $t_R = 13.53\text{ min}$ (major).

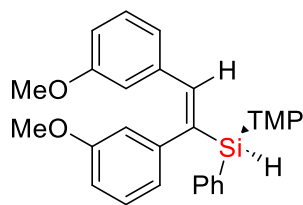
(S, E)-(2,6-dimethylphenyl)(1,2-diphenylvinyl)(phenyl)silane



3m

3m was synthesized following the general procedure C. Yellow liquid (69.4 mg, 89% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 43.6$ ($c = 1.59$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.46 (d, $J = 7.6\text{ Hz}$, 2H), 7.27 - 7.23 (m, 3H), 7.15-7.10 (m, 3H), 7.08 - 7.00 (m, 6H), 6.93 - 6.88 (m, 5H), 5.52 (dd, $J = 8.2\text{ Hz}$, 1H), 2.29 (s, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.7, 142.3, 142.1, 140.1, 137.3, 135.9, 133.9, 130.4, 130.2, 129.7, 129.5, 128.8, 128.12, 128.07, 127.9, 127.5, 126.4, 24.8. HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{28}\text{H}_{26}\text{Si}$, 413.1696, found 413.1695; HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.4mL/min, 254 nm, 86% ee). $t_R = 9.07\text{ min}$ (minor), $t_R = 10.23\text{ min}$ (major).

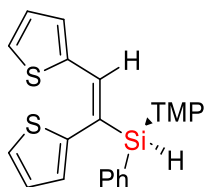
(S, E)-(1,2-bis(3-methoxyphenyl)vinyl)(mesityl)(phenyl)silane



3n

3n was synthesized following the general procedure C. Yellow liquid (82.7 mg, 89% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 38.3$ ($c = 1.6$, CHCl_3). $^1\text{H NMR}$ δ 7.48 (d, $J = 7.0$ Hz, 2H), 7.28 - 7.23 (m, 3H), 7.06 (dd, $J = 8$ Hz, 1H), 6.95 (dd, $J = 8$ Hz, 1H), 6.85 (s, 1H), 6.77(s, 2H), 6.67 - 6.55 (m, 5H), 6.48 (s, 1H), 5.48 (s, 1H), 3.50(s, 3H), 3.39 (s, 3H), 2.27 (s, 6H), 2.20 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 159.9, 159.1, 145.7, 143.7, 141.9, 140.5, 140.0, 138.5, 135.9, 134.0, 129.8, 129.0, 128.9, 128.1, 126.7, 122.6, 120.5, 114.3, 113.8, 113.0, 112.4, 55.1, 54.9, 24.6, 21.3. HRMS (APCI-TOF) m/z : $[\text{M} + \text{K}]^+$ calcd for $\text{C}_{31}\text{H}_{32}\text{O}_2\text{Si}$, 489.1647; found 489.1632; HPLC: Chiralpak MD column (hexanes: isopropanol = 98: 2, 0.5mL/min, 254 nm, 80% ee). $t_R = 7.79$ min (minor), $t_R = 8.71$ min (major).

(S, E)-1,2-di(thiophen-2-yl)vinyl(mesityl)(phenyl)silane



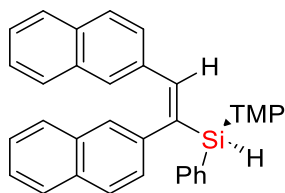
3o

3o was synthesized following the general procedure C. Yellow liquid (75.4mg, 88% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 35.6$ ($c = 1.0$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.48 (d, $J = 1.6$ Hz, 1H), 7.46 (d, $J = 1.6$ Hz, 1H) 7.29 - 7.20 (m, 4H), 7.10 (s, 1H), 7.03 (d, $J = 6.0$ Hz, 1H), 6.92 - 6.90 (m, 1H), 6.84 (d, $J = 5.2$ Hz, 1H), 6.76 (m, 3H), 6.70 (d, $J = 4.8$ Hz, 1H), 5.45 (s, 1H), 2.25 (s, 6H), 2.20 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 144.8, 141.0, 139.4, 139.1, 137.2, 134.8, 132.5, 129.5, 129.3, 128.7, 127.9, 127.23, 127.16, 126.8, 125.2, 125.13, 125.12, 124.8, 23.6, 20.4. HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{25}\text{H}_{24}\text{S}_2\text{Si}$,

439.0981; found 439.0982;

HPLC: Chiralpak MD column (hexanes: isopropanol = 98:2, 0.6mL/min, 254 nm, 87% ee). t_R = 16.08 min (minor), t_R = 18.00 min (major).

(S, E)-(1,2-di(naphthalen-2-yl)vinyl)(mesityl)(phenyl)silane

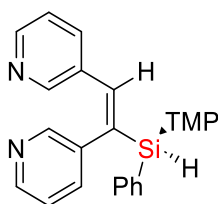


3p

3p was synthesized following the general procedure C. Yellow liquid (75.4 mg, 75% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 42.3$ ($c = 0.74$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.63 - 7.47 (m, 7H), 7.44 - 7.42 (m, 2H) 7.28 - 7.14 (m, 9H), 7.12 - 7.10(m, 1H) 6.93 - 6.90 (m, 1H) 7.45 (d, $J = 6.0$ Hz, 2H), 5.61(s, 1H), 2.28 (s, 6H), 2.17 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.7, 142.6, 140.4, 140.1, 139.9, 136.0, 135.1, 134.1, 133.9, 133.3, 132.7, 132.2, 129.6, 129.5, 129.0, 128.3, 128.2, 128.1, 128.1, 127.8, 127.6, 127.4, 127.3, 127.1, 126.8, 126.6, 126.2, 126.1, 125.9, 125.6, 24.7, 21.4. HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{37}\text{H}_{32}\text{Si}$, 527.2165; found 527.2156;

HPLC: Chiralpak MD column (hexanes: isopropanol = 99.8: 0.2, 0.5mL/min, 254nm, 84%), t_R = 10.69 (minor), t_R = 13.36 min (major).

(S, E)-3,3'-(1-(mesityl(phenyl)silyl)ethene-1,2-diyl)dipyridine

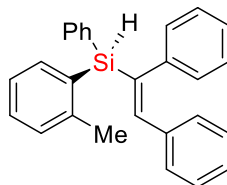


3q

3q was synthesized following the general procedure C. Clear viscous liquid (30.4 mg, 38% yield) purified by column chromatography (PE/EA= 80:1). $[\alpha]_D^{20} = 21.5$ ($c = 0.2$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 8.35 (dd, $J = 1.6$ Hz, 1H), 8.28

(dd, $J = 1.6$ Hz, 1H), 8.24 (d, $J = 2.8$ Hz, 1H), 8.19 (d, $J = 1.6$ Hz, 1H), 7.49 (d, $J = 1.6$ Hz, 1H), 7.47 (d, $J = 1.6$ Hz, 1H), 7.34 - 7.27 (m, 4H), 7.20 (s, 1H), 7.10 - 7.07(m, 2H), 6.97 (s, 1H), 6.80 (s, 2H), 5.52 (s, 1H), 2.27 (s, 6H), 2.22 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 150.7, 148.6, 148.5, 147.8, 145.6, 140.6, 140.3, 140.2, 136.1, 135.9, 135.7, 132.5, 130.0, 129.2, 128.4, 125.2, 123.8, 123.1, 24.6, 21.4. HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{27}\text{H}_{26}\text{N}_2\text{Si}$, 429.1757; found 429.1755; HPLC: Chiralpak MD column (hexanes: isopropanol = 80: 20, 0.5 mL/min, 254 nm, 87% ee). $t_{\text{R}} = 5.20$ (minor), $t_{\text{R}} = 6.06$ min (major).

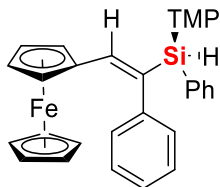
(*R,E*)-(1,2-diphenylvinyl)(phenyl)(*o*-tolyl)silane



3r

3r was synthesized following the general procedure C. Yellow liquid (64.7 mg, 86% yield). purified by column chromatography (PE/EA= 80:1). $[\alpha]_{\text{D}}^{20} = -26.8$ ($c = 0.25$, CHCl_3). ^1H NMR (400 MHz, CDCl_3): δ 7.50 (d, $J = 6.4$ Hz, 2H), 7.34 - 7.22 (m, 5H), 7.13 - 7.09 (m, 3H), 7.07 – 6.98 (m, 7H), 6.91 – 6.88 (m, 2H), 6.80 (s, 1H). 5.29 (s, 1H), 2.37 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 144.9, 142.9, 142.1, 139.9, 137.1, 137.1, 136.1, 133.1, 132.0, 130.3, 129.9, 129.8, 128.8, 128.2, 128.1, 128.1, 127.6, 126.3, 125.2, 22.8. HRMS (APCI-TOF) m/z : $[\text{M} + \text{Na}]^+$ calcd for $\text{C}_{27}\text{H}_{24}\text{Si}$, 399.1539, found 399.1541; HPLC: Chiralpak MD column (hexanes: isopropanol = 99.9:0.1, 0.5 mL/min, 254 nm, 34% ee). $t_{\text{R}} = 7.976$ min (minor), $t_{\text{R}} = 9.499$ min (major).

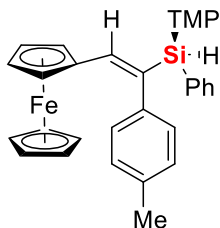
cyclopenta-2,4-dien-1-yl(3-((*E*)-2-((*S*)-mesityl(phenyl)silyl)-2-phenylvinyl)cyclopenta-2,4-dien-1-yl)iron



9a

9a was synthesized following the general procedure D. Reddish brown viscous oil (77mg, 75% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -34.33$ ($c = 0.3$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.46 (d, $J = 6.1$ Hz, 2H), 7.28 – 7.16 (m, 5H), 7.10 (d, $J = 7.2$ Hz, 1H), 7.05 – 7.03 (m, 2H), 6.76 (s, 2H), 6.64 (s, 1H), 5.40 (s, 1H), 3.95 (s, 7H), 3.69 (d, $J = 10.1$ Hz, 2H), 2.24 (s, 6H), 2.20 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.6, 143.2, 141.3, 139.8, 135.7, 135.6, 134.6, 129.3, 128.8, 128.5, 128.2, 128.0, 127.0, 126.1, 81.4, 70.0, 69.9, 69.3, 69.2, 24.7, 21.4. HRMS (APCI-TOF) m/z : $[M]$ calcd for $\text{C}_{33}\text{H}_{32}\text{FeSi}$, 512.1623; found 512.1642; HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 97% ee). $t_R = 29.45$ (minor), $t_R = 34.08$ min (major).

cyclopenta-2,4-dien-1-yl(3-((*E*)-2-((*S*)-mesityl(phenyl)silyl)-2-(*p*-tolyl)vinyl)cyclopenta-2,4-dien-1-yl)iron

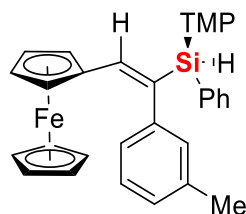


9b

9b was synthesized following the general procedure D. Reddish brown viscous oil (91.89mg, 87% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -28.16$ ($c = 1.14$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.47 (d, $J = 7.8$ Hz, 2H), 7.29 – 7.22 (m, 3H), 6.99 – 6.92 (m, 4H), 6.78 (s, 2H), 6.61 (s, 1H), 5.38 (s, 1H), 3.94 (s, 7H), 3.72 (d, $J = 9.5$ Hz, 2H), 2.24 (s, 6H), 2.21 (s, 3H), 2.19 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.6, 141.2, 140.0, 139.7, 135.8, 135.6, 135.5, 134.8, 129.3,

128.8, 128.0, 127.2, 81.5, 70.0, 69.9, 69.2, 69.1, 24.7, 21.4. HRMS (APCI-TOF) m/z : [M] calcd for $C_{34}H_{34}FeSi$, 526.1779; found 526.1771; HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 91% ee). t_R = 11.035 (minor), t_R = 12.60 min (major).

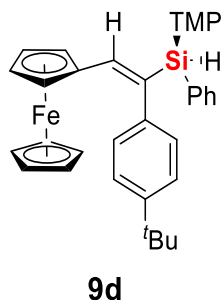
cyclopenta-2,4-dien-1-yl(3-((*E*)-2-((*S*)-mesityl(phenyl)silyl)-2-(*m*-tolyl)vinyl)cyclopenta-2,4-dien-1-yl)iron



9c

9c was synthesized following the general procedure D. Reddish brown viscous oil (102.9 mg, 98% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -10.31$ ($c = 0.5625$, $CHCl_3$). 1H NMR (400 MHz, $CDCl_3$): δ 7.45 (d, $J = 7.8$ Hz, 2H), 7.26 – 7.19 (m, 3H), 7.08 – 7.03 (m, 1H), 6.91 – 6.89 (m, 1H), 6.86 – 6.84 (m, 2H), 6.76 (s, 2H), 6.61 (s, 1H), 5.38 (s, 1H), 3.98 – 3.97 (m, 7H), 3.73 (d, $J = 11.8$ Hz), 2.24 (m, 12H). ^{13}C NMR (100 MHz, $CDCl_3$) δ 144.6, 142.0, 140.1, 138.7, 136.9, 134.7, 134.7, 133.7, 128.3, 127.81, 127.77, 127.4, 127.0, 126.2, 125.8, 124.2, 80.4, 69.0, 68.9, 68.2, 68.2, 23.7, 20.7, 20.4. HRMS (APCI-TOF) m/z : [M] calcd for $C_{34}H_{34}FeSi$, 526.1779; found 526.1777; HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 90% ee). t_R = 18.52 (minor), t_R = 20.05 min (major).

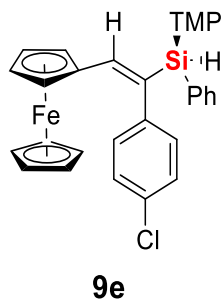
(3-((*E*)-2-(4-(tert-butyl)phenyl)-2-((*S*)-mesityl(phenyl)silyl)vinyl)cyclopenta-2,4-dien-1-yl)(cyclopenta-2,4-dien-1-yl)iron



9d was synthesized following the general procedure D. Reddish brown viscous oil (106.4 mg, 94% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -26.9$ ($c = 1.955$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.45 (d, $J = 7.9$ Hz, 2H), 7.25 – 7.16 (m, 5H), 6.97 (d, $J = 8.4$ Hz, 2H), 6.75 (s, 2H), 6.60 (s, 1H), 5.38 (s, 1H), 3.94 (s, 7H), 3.73 (d, $J = 14.2$ Hz, 2H), 2.23 (s, 6H), 2.19 (s, 3H), 1.21 (s, 9H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 148.9, 145.6, 141.2, 139.9, 139.7, 135.7, 135.5, 134.9, 129.2, 128.8, 128.0, 127.7, 127.4, 125.3, 81.6, 70.0, 69.9, 69.2, 69.1, 34.5, 31.6, 24.7, 21.4. HRMS (APCI-TOF) m/z : $[M]$ calcd for $\text{C}_{37}\text{H}_{40}\text{FeSi}$, 568.2249; found 568.2232;

HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 96% ee). $t_R = 20.411$ (major), $t_R = 26.030$ min (minor).

(3-((*E*)-2-(4-chlorophenyl)-2-((*S*)-mesityl(phenyl)silyl)vinyl)cyclopenta-2,4-dien-1-yl)(cyclopenta-2,4-dien-1-yl)iron

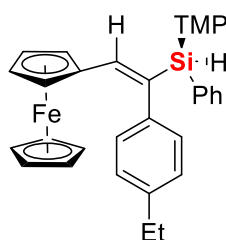


9e was synthesized following the general procedure D. Reddish brown viscous oil (80.5 mg, 74% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -34.62$ ($c = 0.6875$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.45 (d, $J = 7.8$ Hz, 2H), 7.29 – 7.21 (m, 3H), 7.16 – 7.14 (m, 2H), 6.98 – 6.96 (m, 2H), 6.77 (s, 2H), 6.65 (s, 1H), 5.37 (s, 1H), 4.00 (s, 2H), 3.95 (s, 5H), 3.71 (d, $J = 10$ Hz, 2H), 2.23 (s, 6H),

2.20 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 145.5, 142.0, 141.7, 140.0, 135.7, 134.2, 131.8, 129.6, 129.5, 128.9, 128.8, 128.1, 126.6, 81.0, 70.0, 69.9, 69.5, 69.4, 69.2, 24.7, 21.4. HRMS (APCI-TOF) m/z : [M] calcd for $\text{C}_{33}\text{H}_{31}\text{ClFeSi}$, 568.2249; found 568.2232;

HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 96% ee). t_{R} = 34.896 (major), t_{R} = 41.956 min (minor).

cyclopenta-2,4-dien-1-yl(3-((*E*)-2-(4-ethylphenyl)-2-((*S*)-mesityl(phenyl)silyl)vinyl)cyclopenta-2,4-dien-1-yl)iron

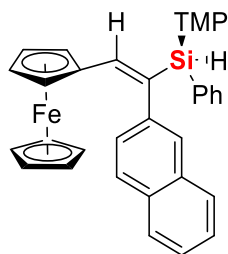


9f

9f was synthesized following the general procedure D. Reddish brown viscous oil (95.7 mg, 89% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_{\text{D}}^{20}$ = -29.41 (c = 1.5575, CHCl_3). ^1H NMR (400 MHz, CDCl_3): δ 7.43 (d, J = 5.8 Hz, 2H), 7.19 – 7.12 (m, 3H), 7.00 – 6.93 (m, 4H), 6.71 (s, 2H), 6.60 (s, 1H), 5.39 (s, 1H), 3.90 (s, 7H), 3.69 (d, J = 13.5 Hz, 2H), 2.47 (q, J = 15.1, 7.6 Hz, 2H), 2.22 (s, 6H), 2.21 (s, 3H), 1.08 (t, J = 7.6 Hz, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 145.5, 141.8, 141.2, 140.2, 139.6, 135.7, 135.5, 134.7, 129.2, 128.8, 128.0, 128.0, 127.2, 81.4, 70.0, 69.8, 69.2, 69.1, 69.1, 28.6, 24.7, 21.4, 15.6. HRMS (APCI-TOF) m/z : [M] calcd for $\text{C}_{35}\text{H}_{36}\text{FeSi}$, 540.1936; found 540.1933;

HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 96% ee). t_{R} = 24.455 (major), t_{R} = 31.760 min (minor).

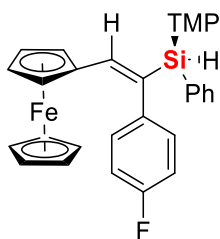
cyclopenta-2,4-dien-1-yl(3-((*E*)-2-((*S*)-mesityl(phenyl)silyl)-2-(naphthalen-2-yl)vinyl)cyclopenta-2,4-dien-1-yl)iron



9g

9g was synthesized following the general procedure D. Reddish brown viscous oil (102.3 mg, 91% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -32$ ($c = 0.075$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.71 – 7.61 (m, 3H), 7.52 – 7.48 (m, 3H), 7.33 – 7.31 (m, 2H), 7.00 – 6.93 (m, 4H), 6.77 (s, 2H), 6.72 (s, 1H) 5.48 (s, 1H), 3.95 (s, 5H), 3.90 (s, 2H), 3.69 (d, $J = 14.8$ Hz, 2H), 2.26 (s, 6H), 2.20 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.6, 141.8, 140.9, 139.9, 135.8, 135.3, 134.6, 133.9, 132.1, 129.4, 128.9, 128.1, 128.03, 127.96, 127.8, 127.6, 127.1, 126.2, 125.9, 125.4, 81.4, 70.2, 70.1, 69.4, 69.3, 69.2, 24.8, 21.4. HRMS (APCI-TOF) m/z : $[\text{M}]$ calcd for $\text{C}_{37}\text{H}_{34}\text{FeSi}$, 562.1779; found 562.1767; HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 210 nm, 93% ee). $t_R = 43.734$ (minor), $t_R = 48.368$ min (major).

cyclopenta-2,4-dien-1-yl(3-((*E*)-2-(4-fluorophenyl)-2-((*S*)-mesityl(phenyl)silyl)vinyl)cyclopenta-2,4-dien-1-yl)iron



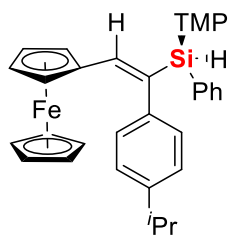
9h

9h was synthesized following the general procedure D. Reddish brown viscous oil (99.6 mg, 94% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -16.51$ ($c = 1.29$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.45 (d, $J = 7.7$ Hz, 2H), 7.26 – 7.19 (m, 3H), 6.99 – 6.95 (m, 2H), 6.88 – 6.83 (m, 2H), 6.75 (s, 2H), 6.66 (s, 1H), 5.38 (s, 1H), 3.97 – 3.94 (m, 7H), 3.69 (d, $J = 13.5$ Hz, 2H), 2.23 (s, 6H), 2.18 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 162.7(d, $J_{\text{C-F}} = 243$ Hz), 145.5, 142.0, 139.9,

138.9 (d, J_{C-F} = 3 Hz), 135.7, 134.4 (d, J_{C-F} = 9 Hz), 129.4, 128.9, 128.1, 126.8, 115.6, 115.4, 81.1, 70.0, 69.9, 69.4, 69.3, 69.2, 24.7, 21.4. ^{19}F NMR (400 MHz, CDCl_3) δ -116.9 (m). HRMS (APCI-TOF) m/z : [M] calcd for $\text{C}_{33}\text{H}_{31}\text{FFeSi}$, 530.1528; found 530.1525;

HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 92% ee). t_R = 33.536 (major), t_R = 40.253 min (minor).

cyclopenta-2,4-dien-1-yl(3-((*E*)-2-(4-isopropylphenyl)-2-((*S*)-mesityl(phenyl)silyl)vinyl)cyclopenta-2,4-dien-1-yl)iron

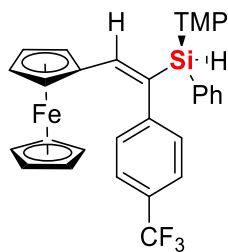


9i

9i was synthesized following the general procedure D. Reddish brown viscous oil (103.0 mg, 93% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20}$ = -15.92 (c = 0.76, CHCl_3). ^1H NMR (400 MHz, CDCl_3): δ 7.44 (d, J = 7.8 Hz, 2H), 7.26 – 7.19 (m, 3H), 7.05 – 7.02 (m, 2H), 6.97 – 6.95 (m, 2H), 6.76 (s, 2H), 6.61 (s, 1H), 5.38 (s, 1H), 3.94 (s, 7H), 3.72 (d, J = 13.2 Hz, 2H), 2.77 (dt, J = 13.8, 6.9 Hz, 1H), 2.23 (s, 6H), 2.20 (s, 3H), 1.14 (d, J = 7.0 Hz, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 146.6, 145.6, 141.2, 140.3, 139.7, 135.7, 135.6, 134.9, 129.2, 128.8, 128.0, 127.3, 126.5, 81.5, 70.0, 69.9, 69.2, 69.1, 33.8, 24.7, 24.2, 21.4. HRMS (APCI-TOF) m/z : [M] calcd for $\text{C}_{36}\text{H}_{38}\text{FeSi}$, 554.2092; found 554.2082;

HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 210 nm, 95% ee). t_R = 19.722 (major), t_R = 24.578 min (minor).

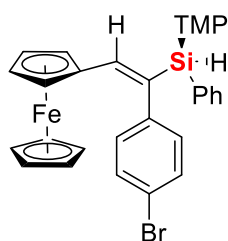
cyclopenta-2,4-dien-1-yl(3-((*E*)-2-((*S*)-mesityl(phenyl)silyl)-2-(4-(trifluoromethyl)phenyl)vinyl)cyclopenta-2,4-dien-1-yl)iron



9j

9j was synthesized following the general procedure D. Reddish brown viscous oil (109 mg, 94% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_{\text{D}}^{20} = -27.11$ ($c = 2.11$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.47 – 7.41 (m, 4H), 7.28 – 7.20 (m, 3H), 7.13 (d, $J = 8.2$ Hz, 2H), 6.76 (s, 2H), 6.69 (s, 1H), 5.38 (s, 1H), 3.98 (s, 2H), 3.95 (s, 5H), 3.67 (d, $J = 14.5$ Hz, 2H), 2.23 (s, 6H), 2.19 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 147.4, 145.6, 142.4, 140.1, 135.7, 134.1, 134.0, 129.6, 129.0, 128.6, 128.2, 126.4, 124.5 (q, $J_{\text{C-F}} = 271$ Hz), 125.5 (q, $J_{\text{C-F}} = 4$ Hz), 80.8, 70.1, 69.9, 69.6, 69.6, 69.3, 24.7, 21.4. $^{19}\text{F NMR}$ (400 MHz, CDCl_3) δ -61.9 (m). HRMS (APCI-TOF) m/z : [M] calcd for $\text{C}_{34}\text{H}_{31}\text{F}_3\text{FeSi}$, 580.1497; found 580.1491; HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 92% ee). $t_{\text{R}} = 14.122$ (major), $t_{\text{R}} = 17.064$ min (minor).

(3-((*E*)-2-(4-bromophenyl)-2-((*S*)-mesityl(phenyl)silyl)vinyl)cyclopenta-2,4-dien-1-yl)(cyclopenta-2,4-dien-1-yl)iron

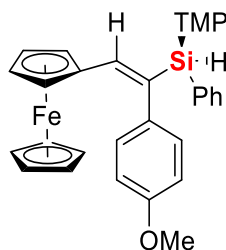


9k

9k was synthesized following the general procedure D. Reddish brown viscous oil (85.0 mg, 72% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_{\text{D}}^{20} = -29.17$ ($c = 2.3075$, CHCl_3). $^1\text{H NMR}$ (400 MHz, CDCl_3): δ 7.45 (d, $J = 7.7$ Hz, 2H), 7.30 – 7.21 (m, 5H), 6.91 (d, $J = 8.4$ Hz, 2H), 6.76 (s, 2H), 6.65 (s, 1H), 5.37 (s, 1H), 3.98 (s, 2H), 3.94 (s, 5H), 3.71 (d, $J = 10.6$ Hz, 2H), 2.23 (s, 6H), 2.19 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 145.5, 142.2, 142.0, 140.0, 135.7, 134.1, 131.7, 130.0,

129.5, 128.9, 128.1, 126.6, 120.0, 81.0, 70.1, 69.9, 69.5, 69.4, 69.2, 24.7, 21.4. HRMS (APCI-TOF) m/z: [M] calcd for C₃₃H₃₁BrFeSi, 590.0728; found 590.0719; HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 210 nm, 96% ee). t_R = 33.680 (major), t_R = 40.114 min (minor).

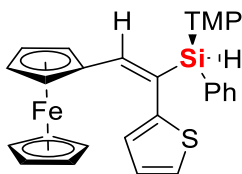
cyclopenta-2,4-dien-1-yl(3-((*E*)-2-((*S*)-mesityl(phenyl)silyl)-2-(4-methoxyphenyl)vinyl)cyclopenta-2,4-dien-1-yl)iron



9I

9I was synthesized following the general procedure D. Reddish brown viscous oil (100.8 mg, 93% yield) purified by column chromatography (PE/EA= 100:1). [α]_D²⁰ = -8.18 (c = 1.5975, CHCl₃). ¹H NMR (400 MHz, CDCl₃): δ 7.46 (d, *J* = 7.5 Hz, 2H), 7.26 – 7.19 (m, 3H), 6.97 – 6.95 (m, 2H), 6.75 – 6.71 (m, 4H), 6.62 (s, 1H), 5.38 (s, 1H), 3.98 – 3.94 (m, 7H), 3.73 (d, *J* = 10.5 Hz, 2H), 3.65 (s, 3H), 2.24 (s, 6H), 2.19 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 158.0, 145.5, 141.4, 139.7, 135.7, 135.2, 135.1, 134.7, 129.3, 129.2, 128.8, 128.0, 127.2, 114.0, 81.5, 70.0, 69.9, 69.2, 69.2, 55.2, 24.7, 21.4. HRMS (APCI-TOF) m/z: [M] calcd for C₃₄H₃₄OFeSi, 542.1728; found 542.1725; HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.5 mL/min, 211 nm, 94% ee). t_R = 25.037 (major), t_R = 42.816 min (minor).

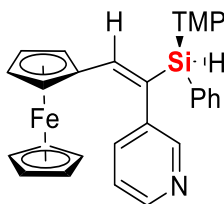
cyclopenta-2,4-dien-1-yl(3-((*E*)-2-((*R*)-mesityl(phenyl)silyl)-2-(thiophen-2-yl)vinyl)cyclopenta-2,4-dien-1-yl)iron



9m

9m was synthesized following the general procedure D. Reddish brown viscous oil (95.3 mg, 92% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -58.86$ ($c = 0.88$, CHCl_3). ^1H NMR (400 MHz, CDCl_3): δ 7.48 (d, $J = 7.5$ Hz, 2H), 7.27 – 7.20 (m, 3H), 7.04 (d, $J = 5.3$ Hz, 1H), 6.80 (t, $J = 3.9$ Hz, 1H), 6.75 (s, 2H), 6.63 (d, $J = 3.8$ Hz, 1H), 5.41 (s, 1H), 4.02 (s, 2H), 3.95 (s, 5H), 3.88 (d, $J = 8.8$ Hz, 2H), 2.26 (s, 6H), 2.18 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 158.0, 145.5, 141.4, 139.7, 135.7, 135.2, 135.1, 134.7, 129.3, 129.2, 128.8, 128.0, 127.2, 114.0, 81.5, 70.0, 69.9, 69.2, 69.2, 55.2, 24.7, 21.4. HRMS (APCI-TOF) m/z : $[\text{M}]$ calcd for $\text{C}_{31}\text{H}_{30}\text{FeSSi}$, 518.1187; found 518.1180; HPLC: Chiralpak MD column (hexanes: isopropanol = 98.1:1.9, 0.9 mL/min, 211 nm, 90% ee). $t_R = 18.015$ (major), $t_R = 35.574$ min (minor).

cyclopenta-2,4-dien-1-yl(3-((*E*)-2-((*S*)-mesityl(phenyl)silyl)-2-(pyridin-3-yl)vinyl)cyclopenta-2,4-dien-1-yl)iron



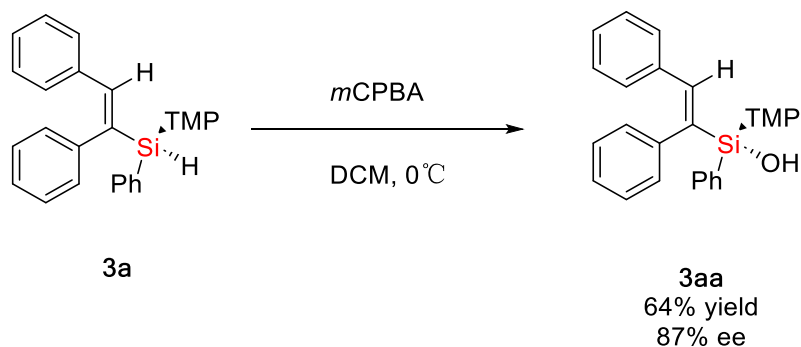
9n

9n was synthesized following the general procedure D. Reddish brown viscous oil (66.7 mg, 65% yield) purified by column chromatography (PE/EA= 100:1). $[\alpha]_D^{20} = -15.54$ ($c = 1.1775$, CHCl_3). ^1H NMR (400 MHz, CDCl_3): δ 8.32 (d, $J = 8.4$ Hz, 2H), 7.48 (d, $J = 6.6$ Hz, 2H), 7.32 – 7.26 (m, 4H), 7.09 (t, $J = 12.9$ Hz, 1H), 6.77 (s, 2H), 6.75 (s, 1H), 5.40 (s, 1H), 4.01 (s, 2H), 3.98 (s, 5H), 3.68 (d, $J = 13.4$ Hz, 2H), 2.24 (s, 6H), 2.21 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3) δ 149.1, 147.2, 145.5, 143.4, 140.1, 135.7, 133.7, 131.2, 129.6, 129.0, 128.2, 126.1, 123.4, 80.7, 70.0, 69.8, 69.6,

69.6, 69.4, 69.2, 24.7, 21.3. HRMS (APCI-TOF) m/z: [M] calcd for C₃₂H₃₁FeNSi, 514.1648; found 514.1648;

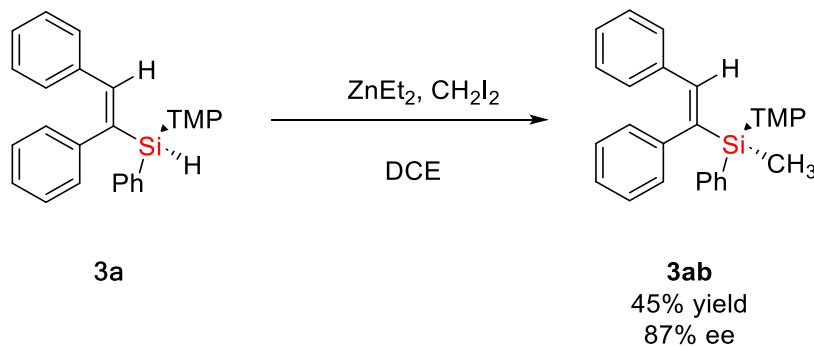
HPLC: Chiralpak MD column (hexanes: isopropanol = 95:5, 0.5 mL/min, 211 nm, 90% ee). t_R = 5.042 (major), t_R = 5.470 min (minor).

2.3.2 Synthetic applications of Si-stereogenic monohydrosilane **3a**



To a flame dried 25 mL Schlenk tube, a solution of **3a** (40.4 mg, 0.1 mmol, 1M) in DCM (1 mL) was cooled to 0 °C, and *m*CPBA (34.4mg, 0.2 mmol) was added to the solution under N₂ atmosphere. After the reaction was completed, the resulting solution was concentrated and purified by preparative thin-layer chromatography using petroleum ether as the eluent to afford colorless liquid **3aa** (26.88mg, 64% yield, 87% ee). $[\alpha]_{\text{D}}^{20} = 24.65$ ($c = 0.215$, CHCl₃). ¹H NMR (400 MHz, CDCl₃) δ 7.55 (d, $J = 8.0$ Hz, 2H), 7.30 – 7.22 (m, 3H), 7.15 – 7.07 (m, 3H), 7.03 - 6.99 (m, 5H), 6.90 – 6.86 (m, 3H), 6.76 (s, 2H), 2.24 (s, 6H), 2.21 (s, 3H), 1.44 (brs, 1H). ¹³C NMR (100 MHz, CDCl₃) δ 145.6, 143.2, 141.6, 141.2, 140.0, 138.1, 137.2, 135.0, 129.9, 129.8, 129.4, 128.9, 128.4, 128.3, 128.2, 128.0, 127.6, 126.4, 25.1, 21.3. HRMS (ESI m/z Calcd for C₂₉H₂₈OSi [M+K]⁺: 459.1541 found: 459.1531.

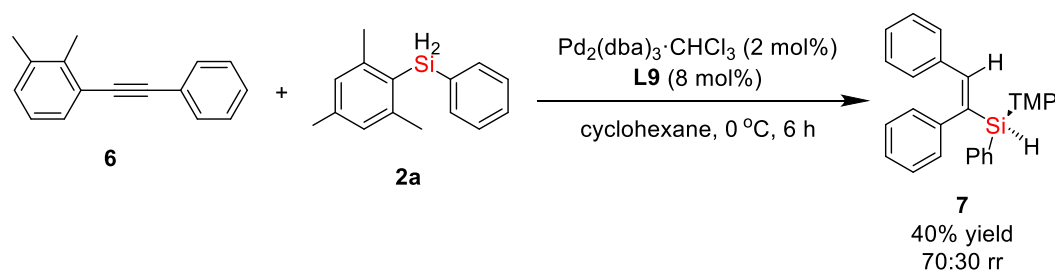
HPLC: Chiralpa AD-H column (hexanes: isopropanol = 99.7:0.3, 0.8 mL/min, 254 nm, 87% ee). $t_{\text{R}} = 6.659$ min (major), $t_{\text{R}} = 4.951$ min (minor)



To a flame dried 25 mL Schlenk tube, a solution of **3a** (40.4 mg, 0.1 mmol, 1M) in DCE (2 ml) was cooled to 0 °C, and Et₂Zn (0.8 mL, 0.8 mmol) was added to the

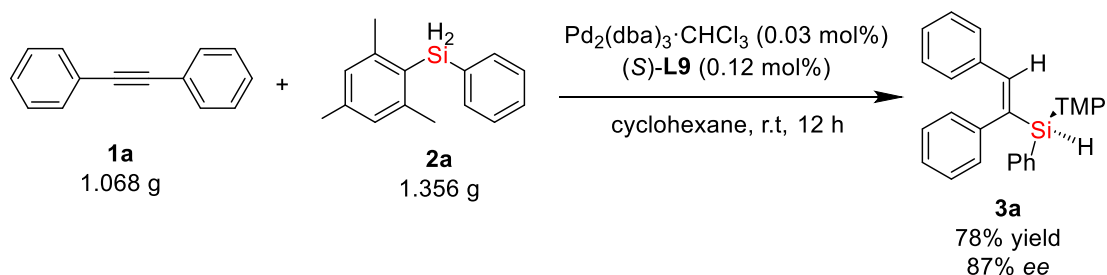
solution under N₂ atmosphere. After that, CH₂l₂ (428.5 mg, 1.6 mmol) was added dropwise. The reaction was kept at 0 °C for 20 min and warmed to room temperature for 36 h. After the reaction was completed, the reaction mixture was cooled to 0 °C and the saturated aqueous solution of NH₄Cl was added. The aqueous phase was then extracted by DCM. The organic layer was washed with brine and dried over anhydrous Na₂SO₄. The resulting solution was concentrated and purified by preparative thin-layer chromatography using petroleum ether as the eluent to afford colorless liquid **3ab** (mg, 45% yield, 87% ee). [α]_D²⁰ = 14.3 (c = 0.14, CHCl₃). ¹H NMR (400 MHz, CDCl₃) δ 7.50 (d, *J* = 7.9 Hz, 2H), 7.31 – 7.22 (m, 3H), 7.16 – 6.93 (m, 7H), 6.93 – 6.88 (m, 3H), 6.80 (s, 1H), 6.78 (s, 2H), 2.23 – 2.18 (m, 9H), 0.48 (s, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 145.8, 144.4, 142.3, 140.5, 140.0, 139.4, 137.7, 135.9, 135.3, 129.9, 129.7, 129.5, 129.3, 129.1, 128.9, 128.8, 128.7, 128.1, 128.0, 127.2, 126.0, 25.7, 21.2, 1.5. HRMS (ESI *m/z* Calcd for C₃₀H₃₀Si [M +Na]⁺: 441.2031 found: 441.2009.

HPLC: Chiralpa MD column (hexanes: isopropanol = 99.9:0.1, 0.5 mL/min, 254 nm, 87% ee). *t_R* = 10.350 min (minor), *t_R* = 11.103 min (major).



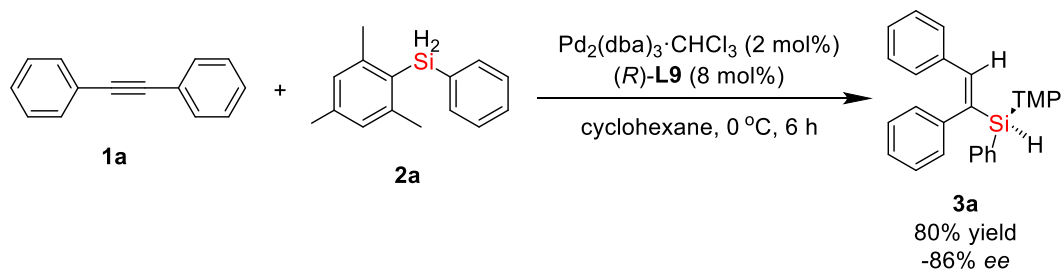
$\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (4.1 mg, 2 mol%), **L9** (11.2 mg, 8 mol%) and diarylacetylene **6** (2 mmol) and **2a** (2 mmol) were added to a 10 mL flask, and 1 mL cyclohexane was added to above mixtures under a nitrogen atmosphere, then the reaction was stirred at 0 °C for 6 hours. After the reaction is complete, the mixture is passed through a short celite pad with DCM as the solvent. Then the mixture was concentrated in vacuum and purified by flash column chromatography (PE/EA = 70:1) to obtain the desired product **7** (40% yield, 70: 30 *rr*).

2.4 Gram-scale synthesis of 3a



$\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (2.0 mg, 0.03 mol%), **L9** (6 mg, 0.12 mol%), diphenylacetylene **1a** (1.068 g, 6 mmol), and **2a** (1.356 g, 6 mmol) were added to a 100 mL flask, and 10 mL cyclohexane was added to above mixtures under a nitrogen atmosphere, then the reaction was stirred at room temperature for 12 hours. After the reaction is complete, the mixture is passed through a short celite pad with DCM as the solvent. Then the mixture was concentrated in vacuum and purified by flash column chromatography (PE/EA = 70:1) to obtain the desired product **3a** (1.89 g, 78% yield, 87% ee).

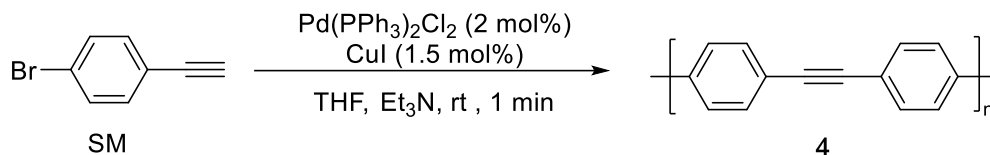
2.5 The effect of P-ligand on the configuration of product



In a flame dried Schlenk tube, $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (4.1 mg, 0.004 mmol, 2 mol%), (*R*)-**L9** (11.2 mg, 0.016 mmol, 8 mol%) in cyclohexane (1 mL, 0.2 M) was stirred at room temperature for 30 min under nitrogen atmosphere. Then **1a** (35.6mg, 0.2 mmol, 1 equiv), **2a** (45.2mg, 0.2 mmol, 1 equiv) were added sequentially to the reaction mixture, and the reaction tube was cooled at rt and then stirred for 6 h. After completion of the reaction, the mixture was passed through a short celite pad using DCM as a solvent. The mixture was then concentrated in vacuo and purified by column chromatography using PE and EA (70:1) to give the desired product **3a** (80% yield, 86% ee) in good yields.

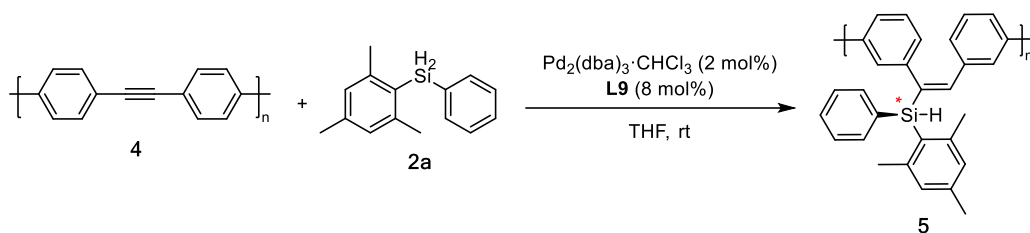
2.6 Synthesis of chiral poly(vinylsilane) **5** bearing Si-stereogenic center

2.6.3 Preparation of Polyalkyne **4**



2 mol% palladium catalyst was added to the reaction tube, and 1.5 mol% cuprous iodide and iodide were then added, triethylamine or tetrahydrofuran was added as solvent, the deoxidation operation was performed for 2 minutes, and 3 equiv triethylamine and acetylene as reactants were added, stirring at room temperature for 1 min. The reaction needs to be precipitated with methanol, pumped and filtered, and dried in a vacuum drying oven for 10 hours.

2.6.4 Synthesis of chiral poly(vinylsilane) **5** via Pd-catalyzed hydrosilylation



In a flame dried Schlenk tube, Pd₂(dba)₃·CHCl₃ (4.1 mg, 0.004 mmol, 2 mol%), L9 (11.2 mg, 0.016 mmol, 8 mol%) in THF (1 mL, 0.2 M) was stirred at room temperature for 30 min under nitrogen atmosphere. Then polyalkyne **4** (0.1 mmol, 0.5 equiv.), dihydrosilane **2a** were added sequentially to the reaction mixture, and the reaction tube was cooled at 0 °C and then stirred for 6 h. After completion of the reaction, the mixture was passed through a short celite pad using DCM as a solvent. The reaction needs to be precipitated with methanol, pumped and filtered, and dried in a vacuum drying oven for 10 hours.

2.6.5 CD spectra of poly(vinylsilane) **5**

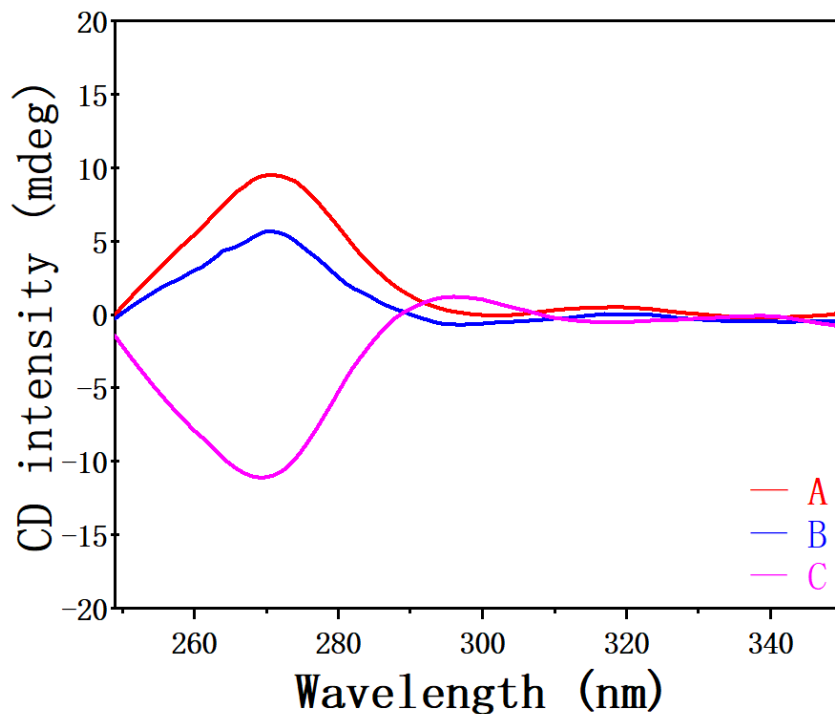
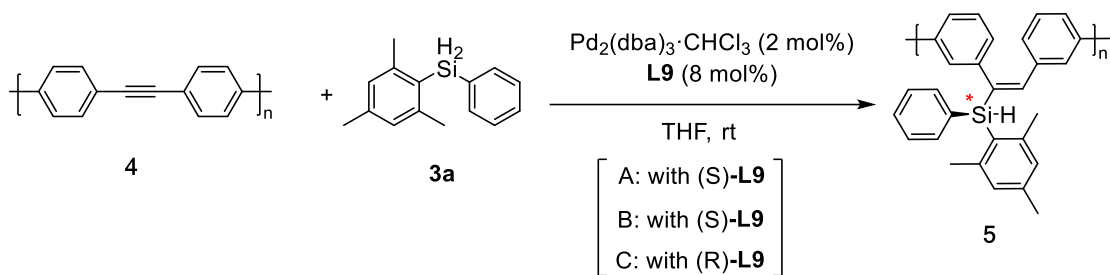


Figure S1. Circular dichroism spectroscopy analysis intensity spectra of (A) a diluted THF solution of the chiral polymer **5** (1 g/L in THF); (B) a diluted THF solution of the chiral polymer **5** (0.6 g/L in THF); (C) a diluted THF solution of the chiral polymer **5** (1 g/L in THF).

2.6.6 SEM images of polyalkyne 4 and poly(vinylsilane) 5

Polyalkyne (4)

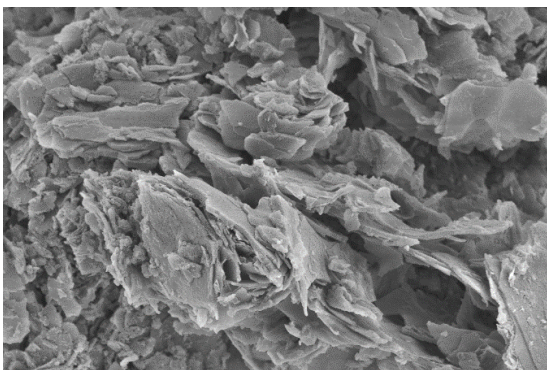


Figure S2. SEM images of 4

Chiral Poly(vinylsilane) (5)

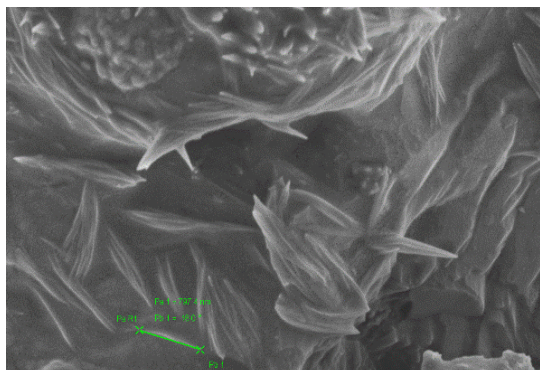


Figure S3. SEM images of 5.

2.6.7 TG analysis of polyalkyne 4 and poly(vinylsilane) 5

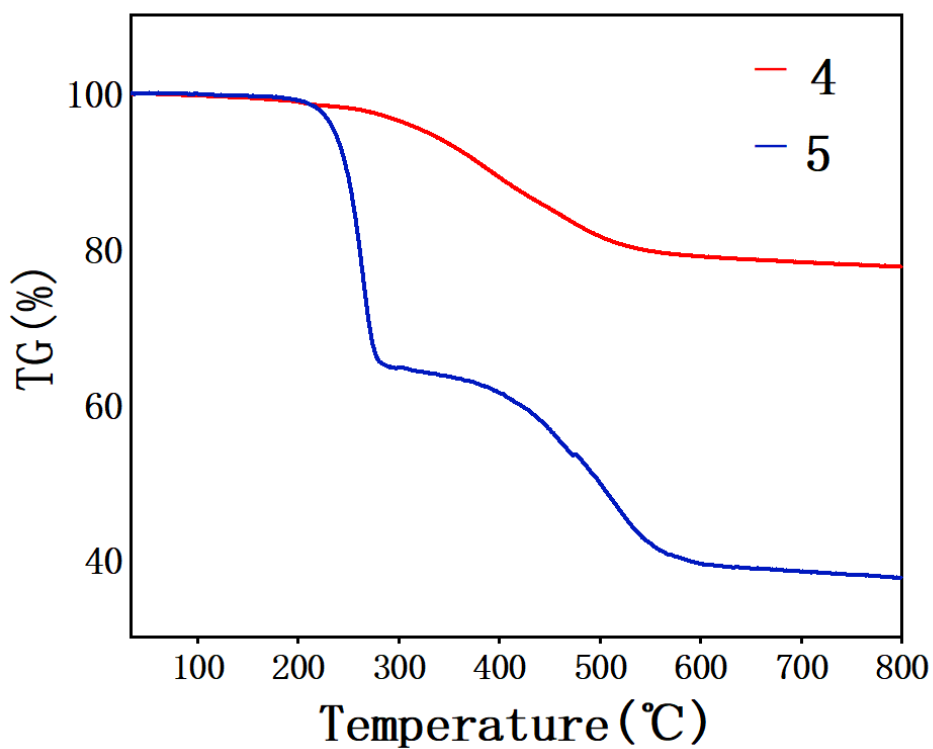
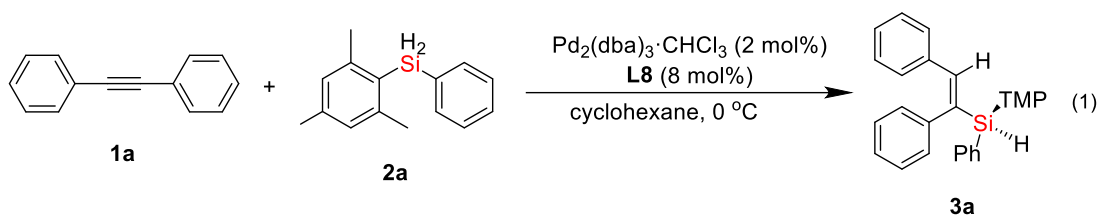
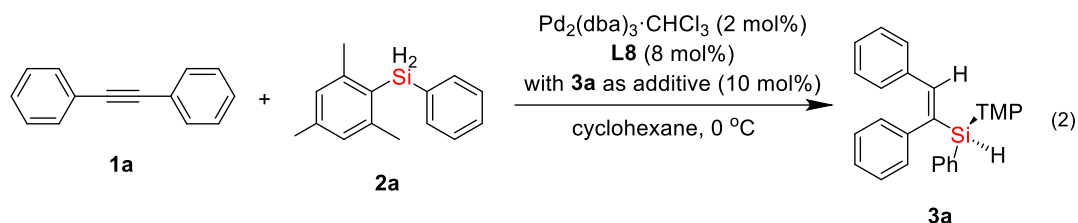


Figure S4. TGA (solid line) curves of 4 and 5 with a heating rate of 10 °C min⁻¹ under N₂.

2.7 Kinetic studies for the Pd-catalyzed hydrosilylation of **1a** and **2a**



In a flame dried Schlenk tube, $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (4.1 mg, 0.004 mmol, 2 mol%), **L8** (11.2 mg, 0.016 mmol, 8 mol%) in cyclohexane (1 mL, 0.2 M) was stirred at room temperature for 30 min under nitrogen atmosphere. Then diaryl alkyne **1a** (0.2 mmol, 1 equiv), **2a** (0.2 mmol, 1 equiv) were added sequentially to the reaction mixture, and the reaction tube was cooled at 0 °C. Then, the yield is measured by GC.



In a flame dried Schlenk tube, $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (4.1 mg, 0.004 mmol, 2 mol%), **L8** (11.2 mg, 0.016 mmol, 8 mol%) in cyclohexane (1 mL, 0.2 M) was stirred at room temperature for 30 min under nitrogen atmosphere. Then **3a** (0.02 mmol, 10 mol%), **1a** (0.2 mmol, 1 equiv), **2a** (0.2 mmol, 1 equiv) were added sequentially to the reaction mixture, and the reaction tube was cooled at 0 °C. Then, the yield is measured by GC (The data for additive **3a** has been deducted).

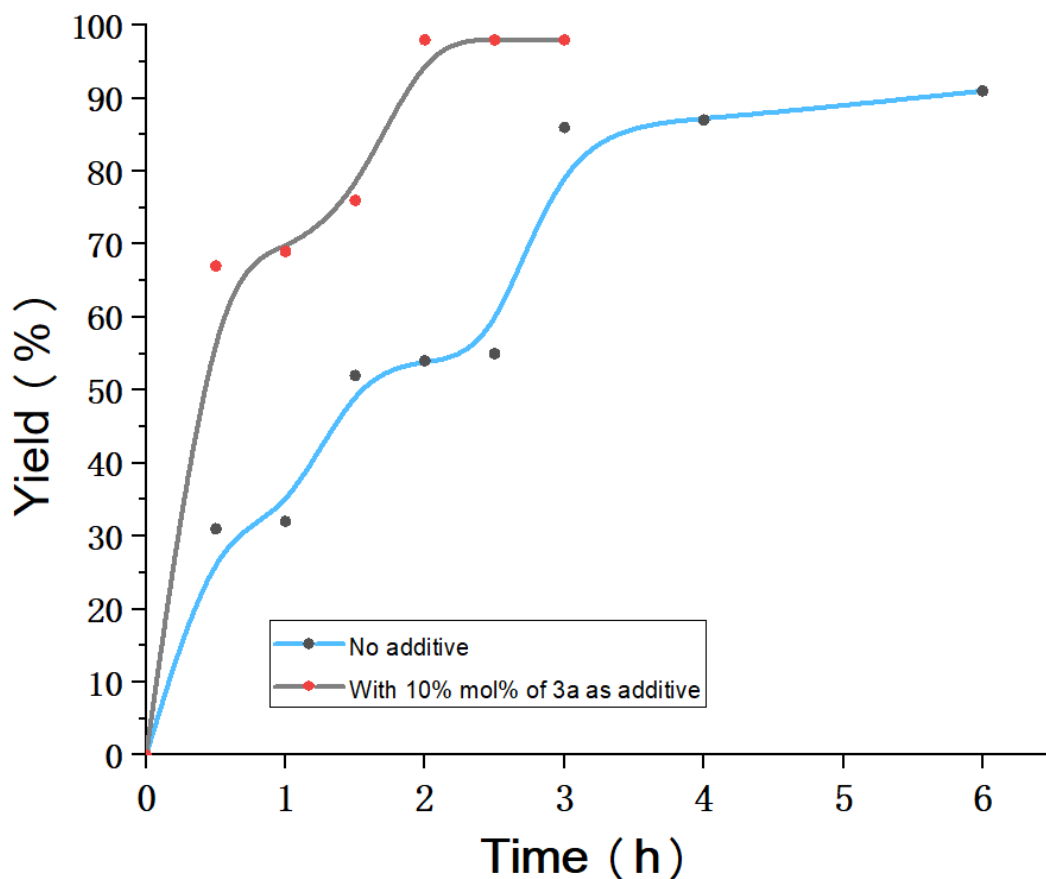
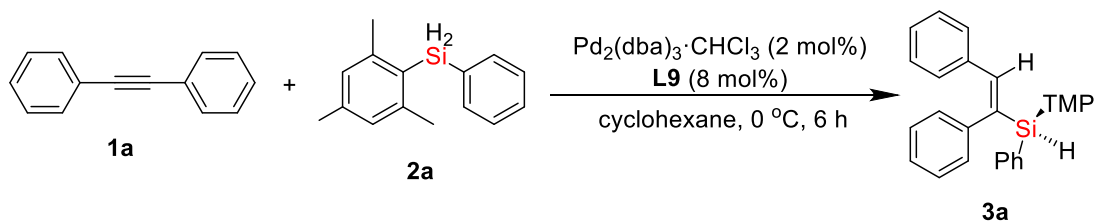


Figure S5. Kinetic studies for the Pd-catalyzed hydrosilylation of **1a** with **2a** with or without **3a** (10 mol%) as additive.

2.8 Experiments for non-linear effect



In a flame dried Schlenk tube, $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (4.1 mg, 0.004 mmol, 2 mol%), **L9** (set 0%, 20%, 40%, 60%, 80%, 100%) in cyclohexane (1 mL, 0.2 M) was stirred at room temperature for 30 min under nitrogen atmosphere. Then diphenylacetylene **1a** (35.6mg, 0.2 mmol, 1 equiv.), **2a** (45.2mg, 0.2 mmol, 1 equiv) were added sequentially to the reaction mixture, and the reaction tube was cooled at room temperature and then stirred for 6 h. After completion of the reaction, the mixture

was passed through a short celite pad using DCM as a solvent. The mixture was then concentrated in vacuo and purified by column chromatography using PE and EA (70:1).

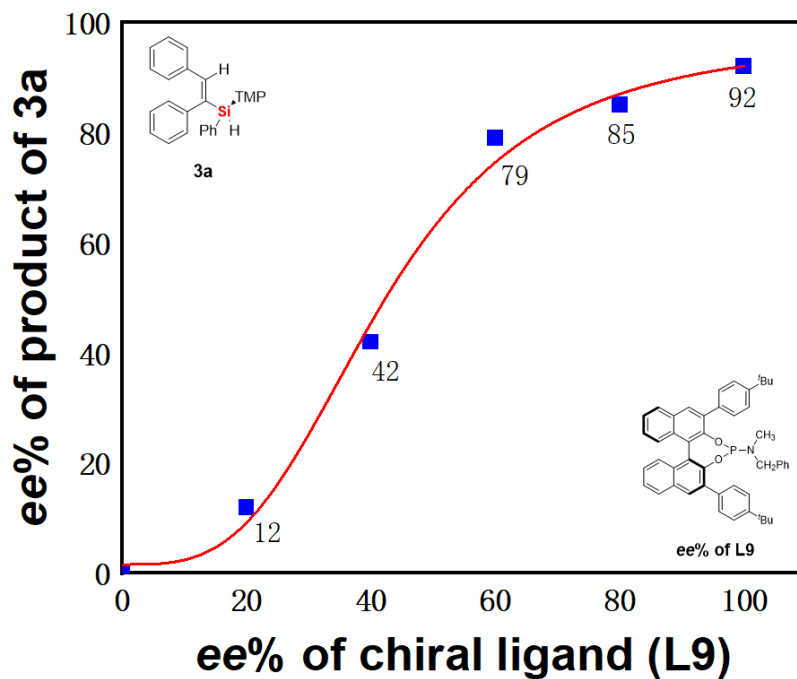
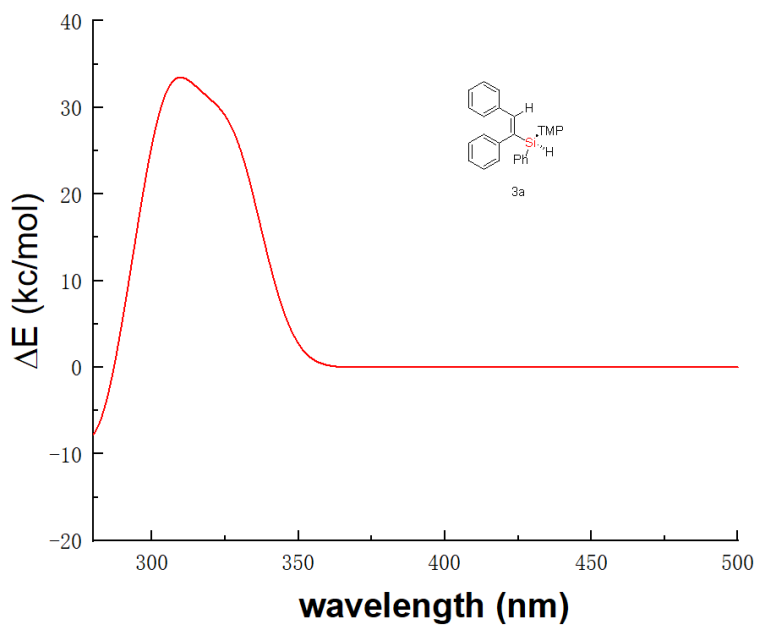
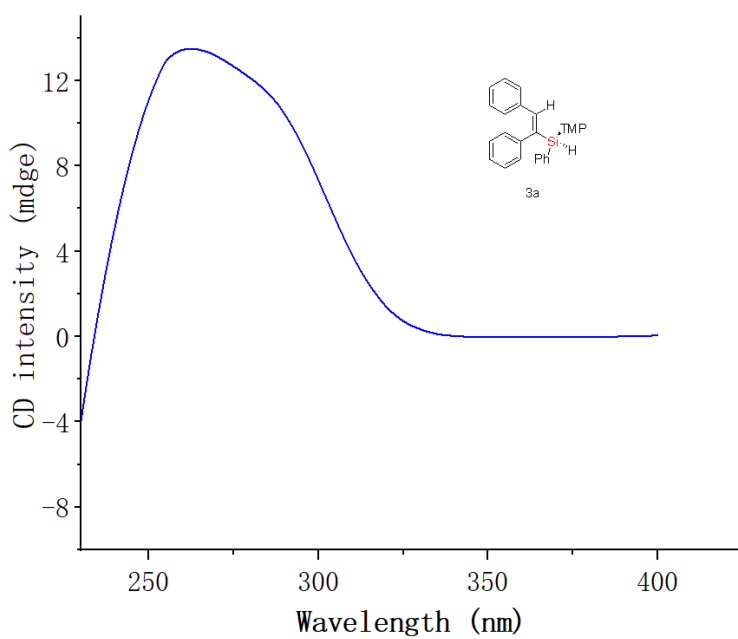


Figure S6. The study of possible NLE in the Pd-catalyzed hydrosilylation

2.9 DFT calculations for the ECD spectrum of chiral product 3a.



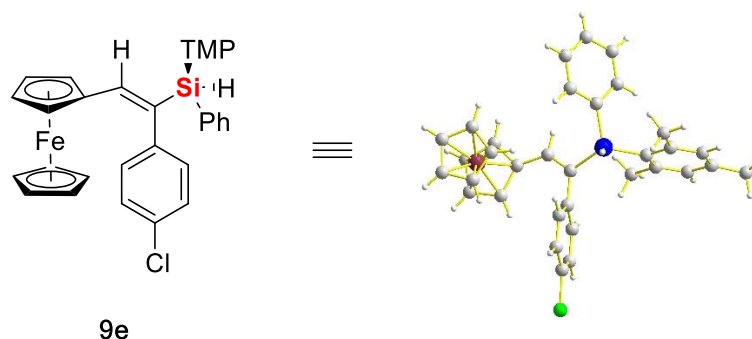
(a) Calculated result



(b) Experimental result

2.10 X-Ray Structure of 9e

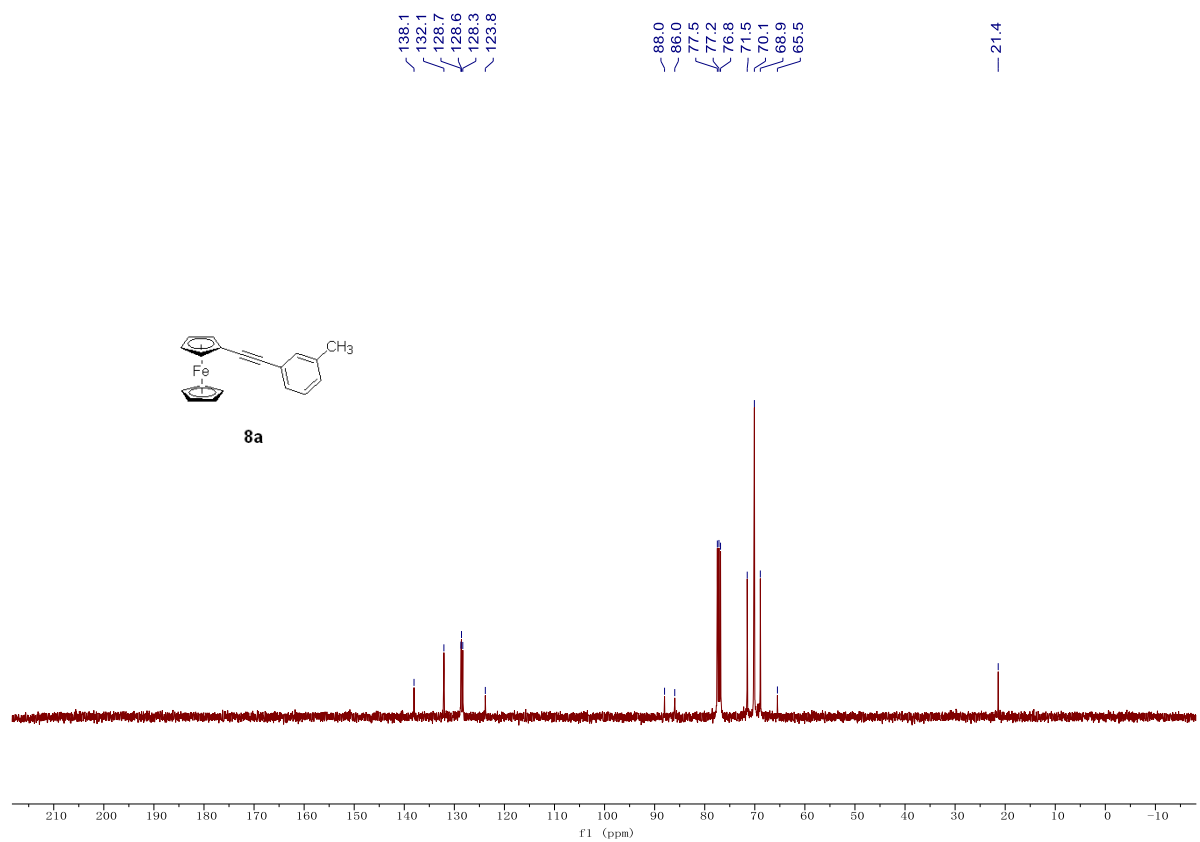
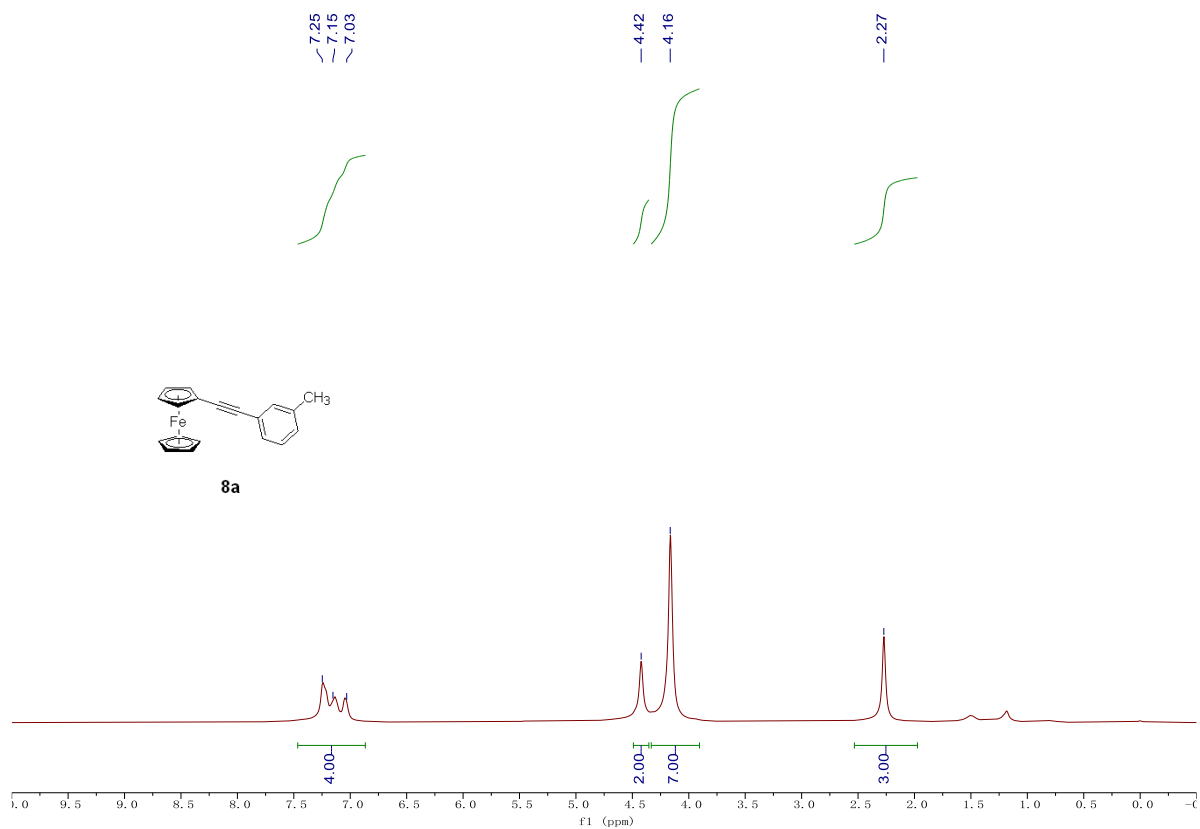
Single crystals of **9e** were obtained by recrystallization from THF. The molecular structure and X-ray diffraction data/refinement of **9e** were shown below. (CCDC:2380481)

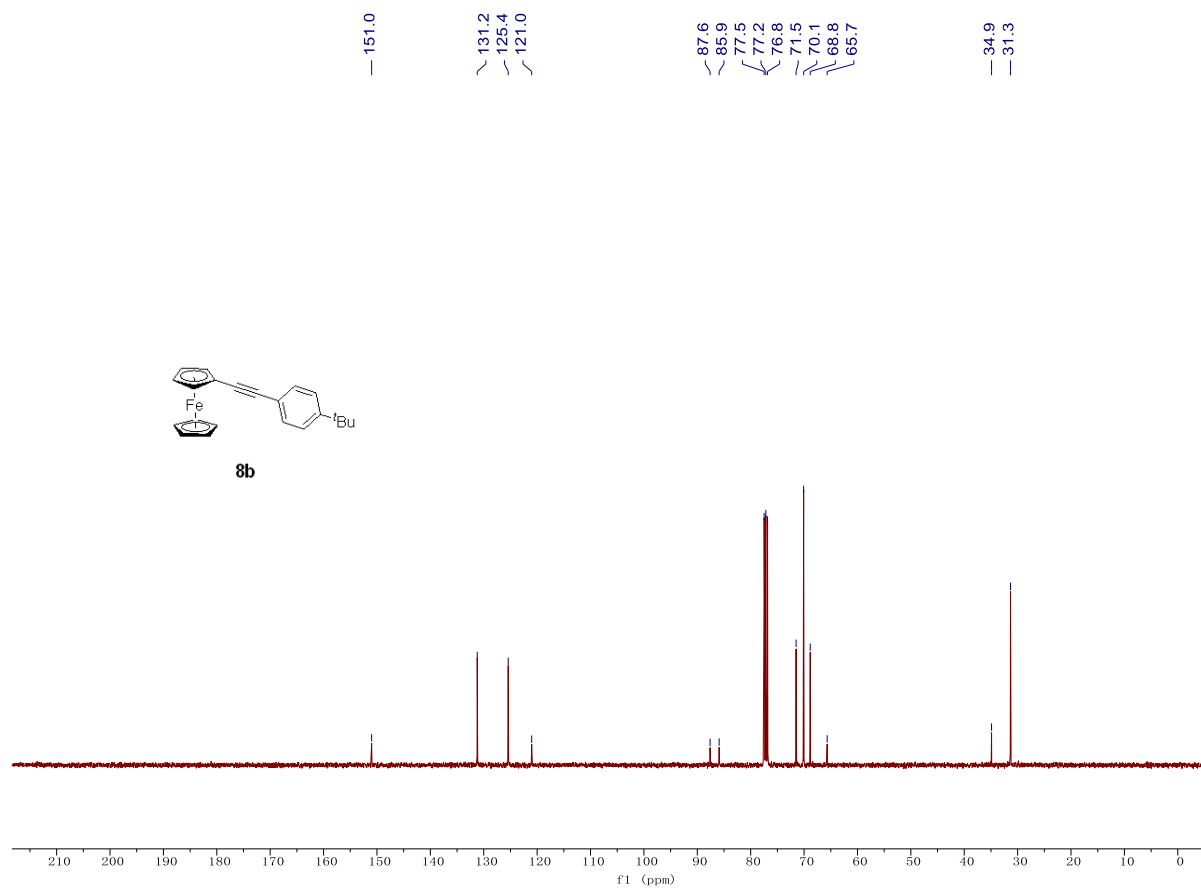
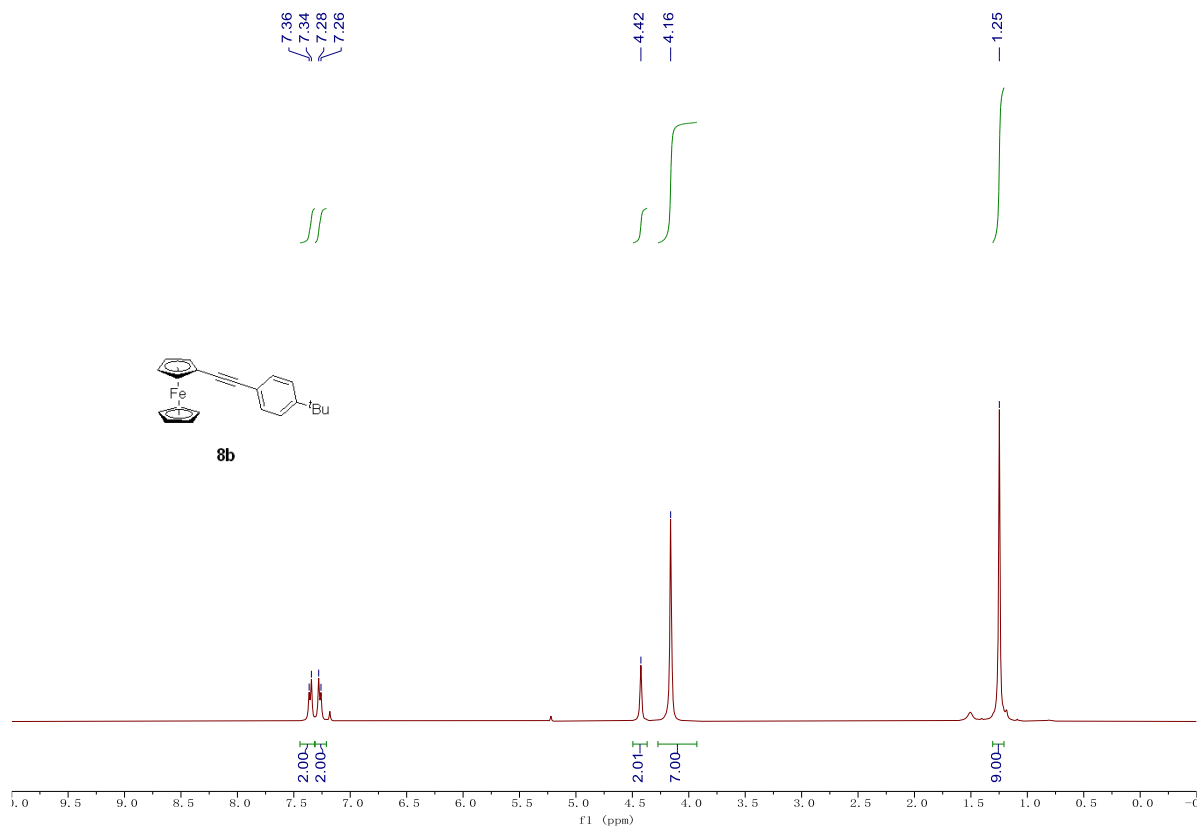


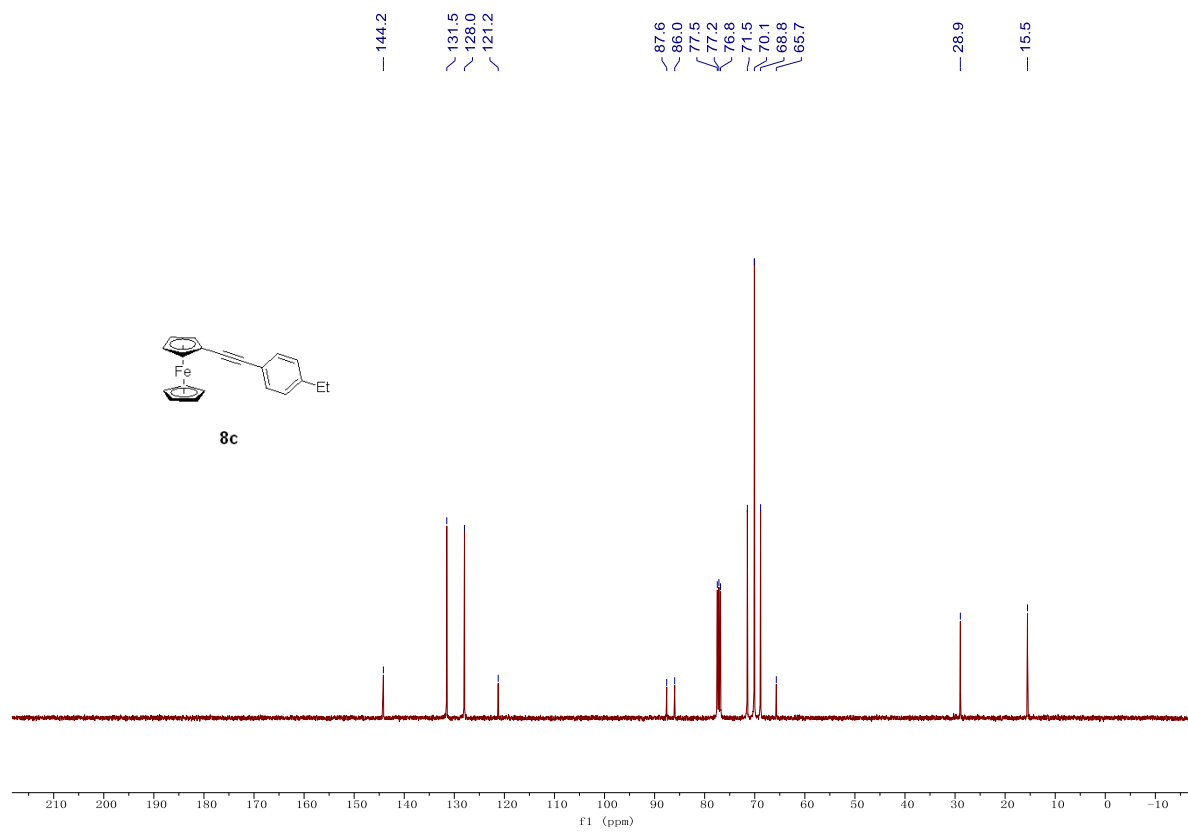
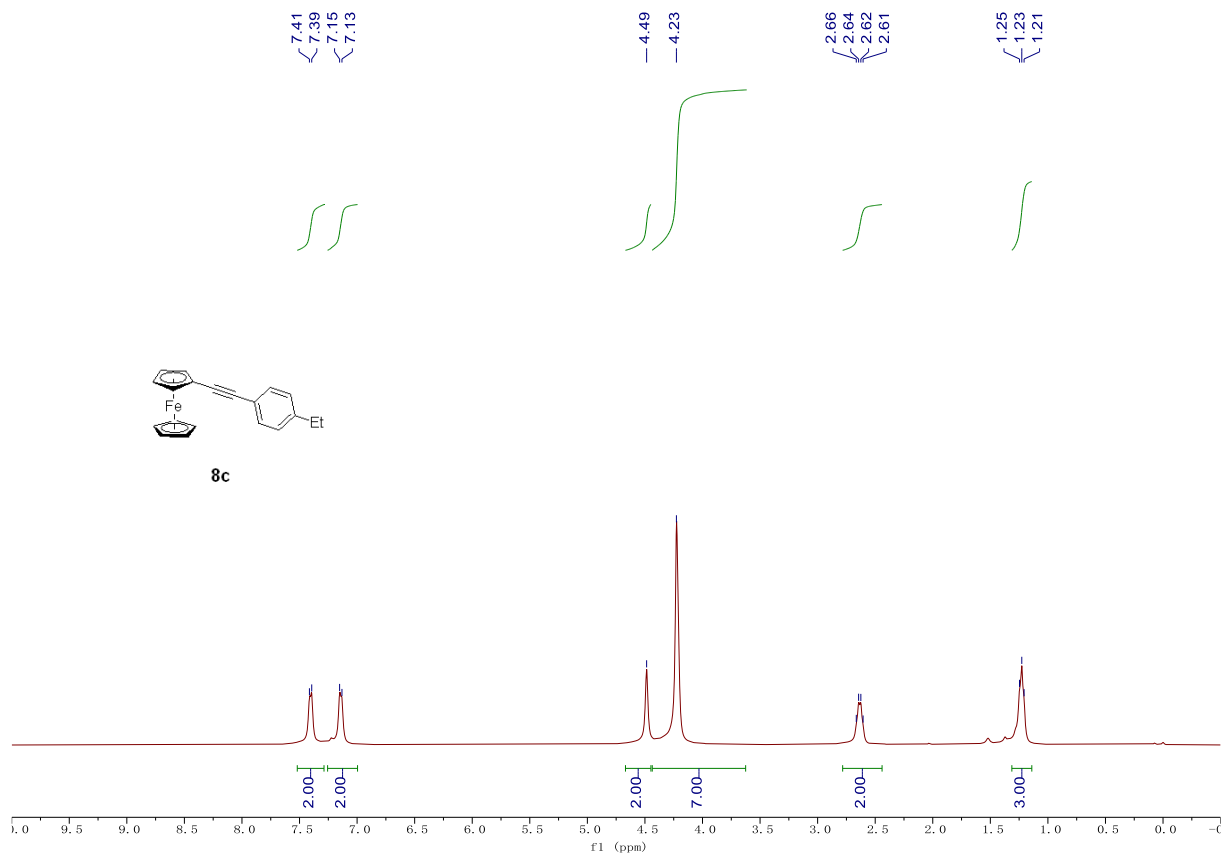
Empirical formula	C ₃₃ H ₃₁ ClFeSi
Formula weight	546.97
Temperature/K	170.00
Crystal system	orthorhombic
Space group	P2 ₁ 2 ₁ 2 ₁
a/Å	7.7294(4)
b/Å	9.1256(4)
c/Å	38.7823(17)
α/°	90
β/°	90
γ/°	90
Volume/Å ³	2735.5(2)
Z	4
ρ _{calc} /cm ³	1.328
μ/mm ⁻¹	4.016
F(000)	1144.0
Crystal size/mm ³	0.07 × 0.06 × 0.04
Radiation	GaKα (λ = 1.34139)

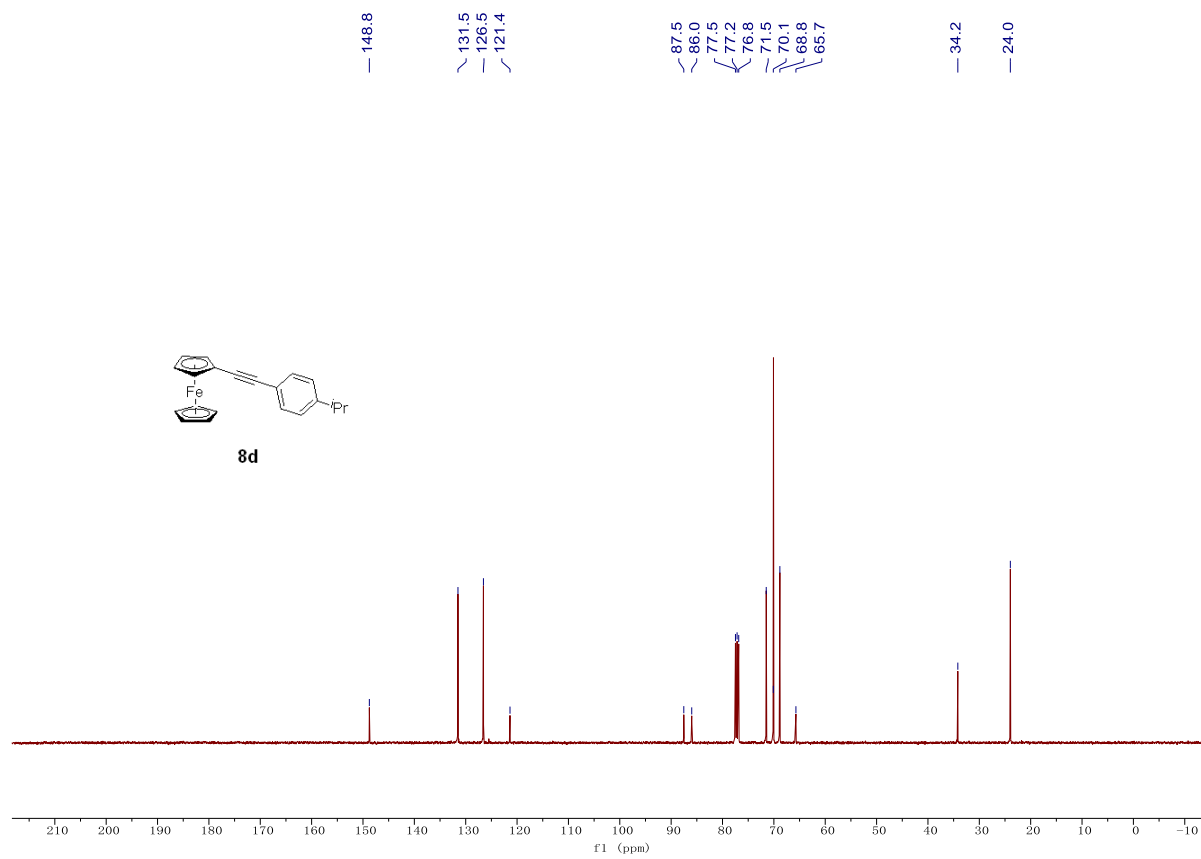
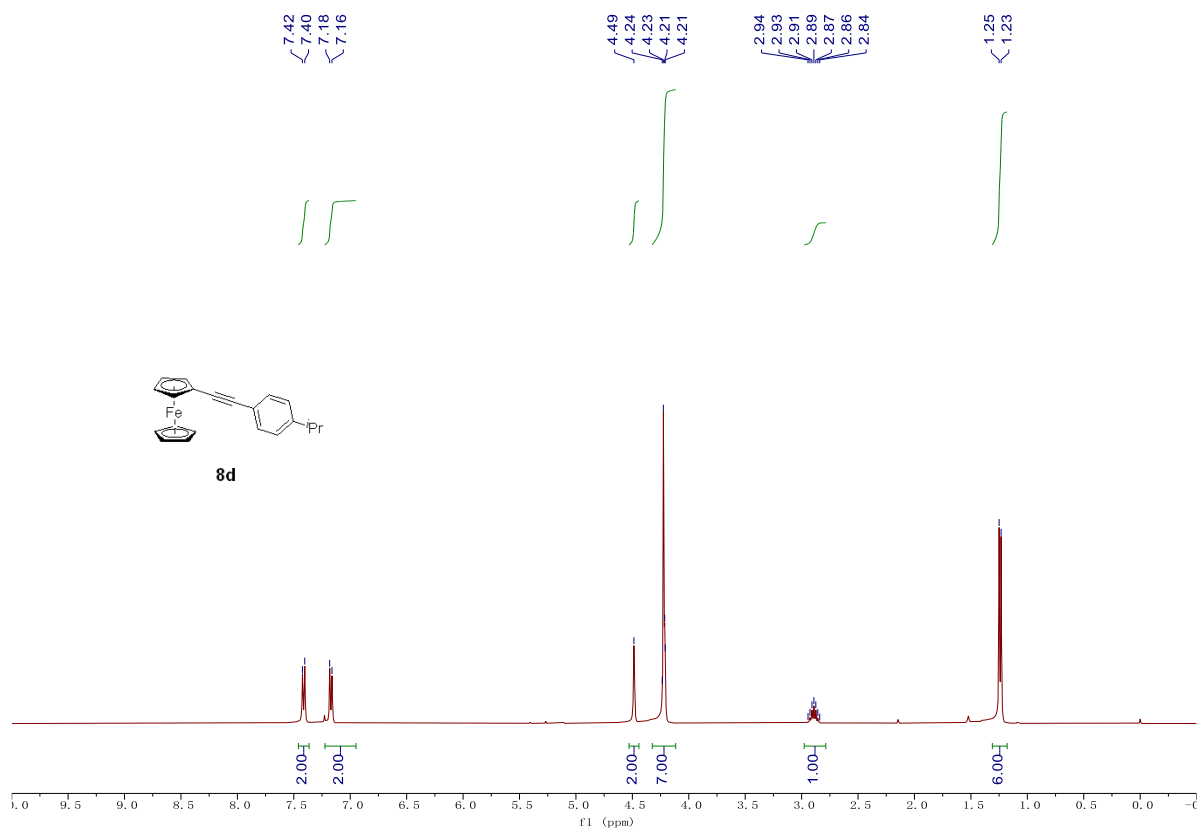
2 θ range for data collection/ $^{\circ}$	7.934 to 114.42
Index ranges	$-9 \leq h \leq 9, -11 \leq k \leq 11, -48 \leq l \leq 48$
Reflections collected	54369
Independent reflections	5609 [$R_{\text{int}} = 0.0824, R_{\text{sigma}} = 0.0700$]
Data/restraints/parameters	5609/0/329
Goodness-of-fit on F^2	1.044
Final R indexes [$I \geq 2\sigma(I)$]	$R_1 = 0.0305, wR_2 = 0.0703$
Final R indexes [all data]	$R_1 = 0.0350, wR_2 = 0.0717$
Largest diff. peak/hole / $e \text{ \AA}^{-3}$	0.24/-0.26
Flack parameter	0.010(5)

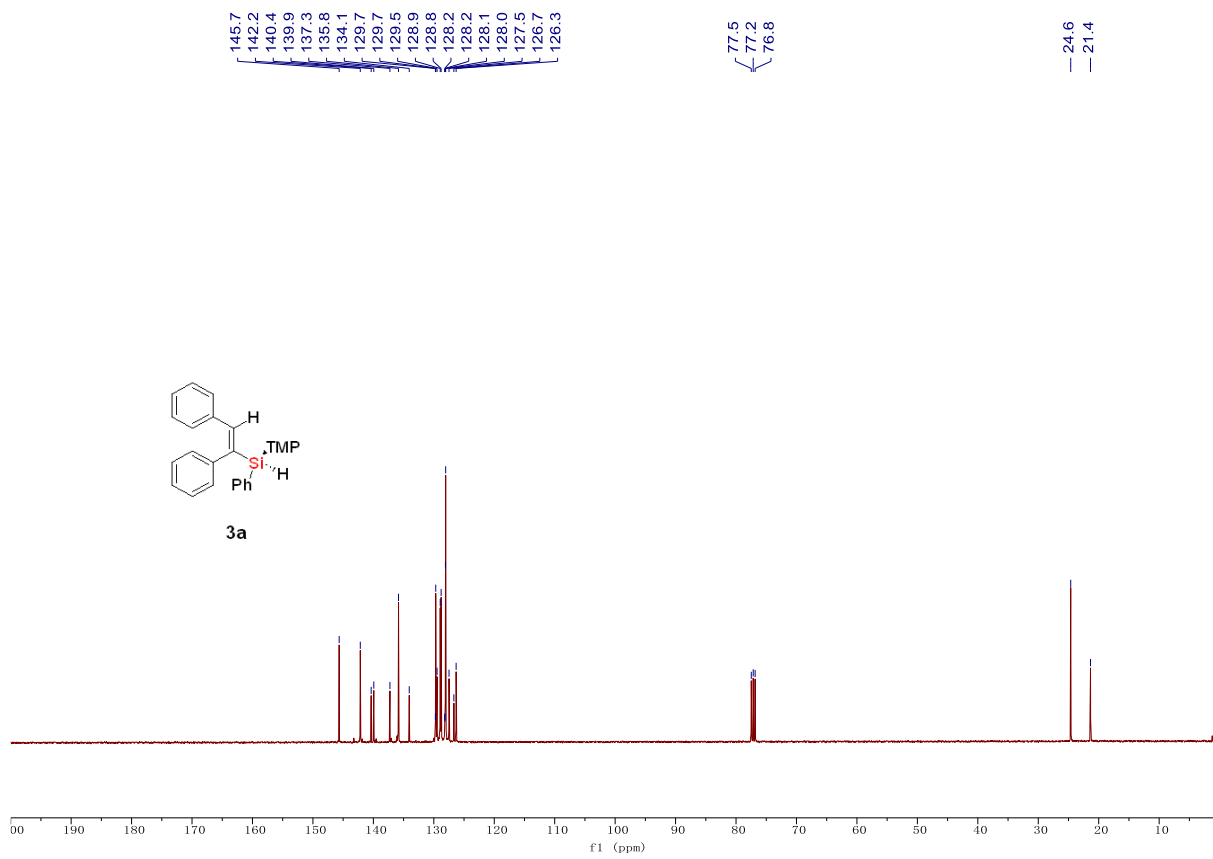
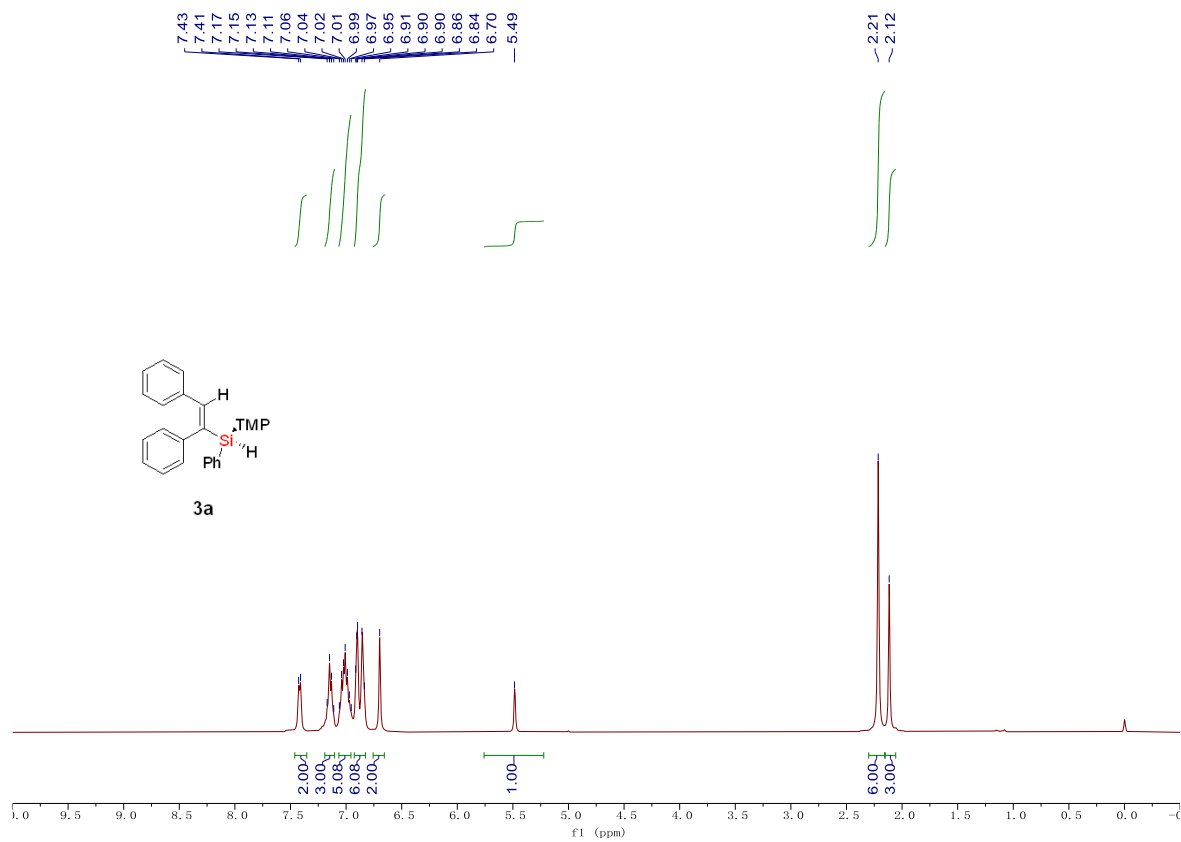
3 NMR Spectra

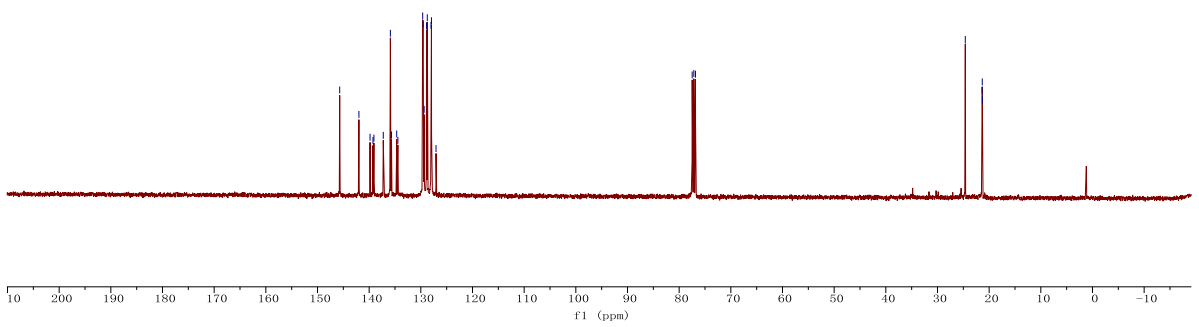
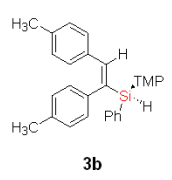
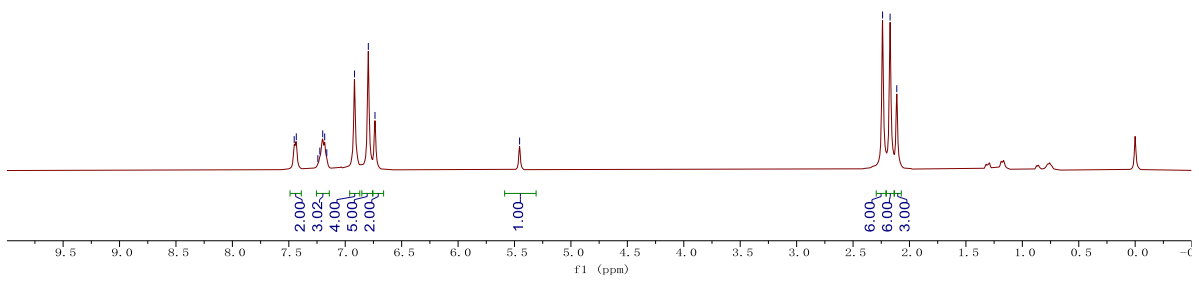
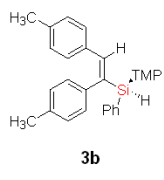
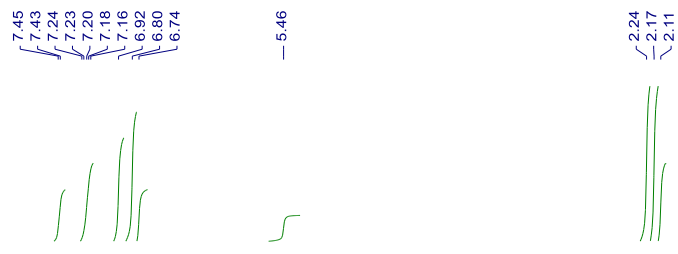


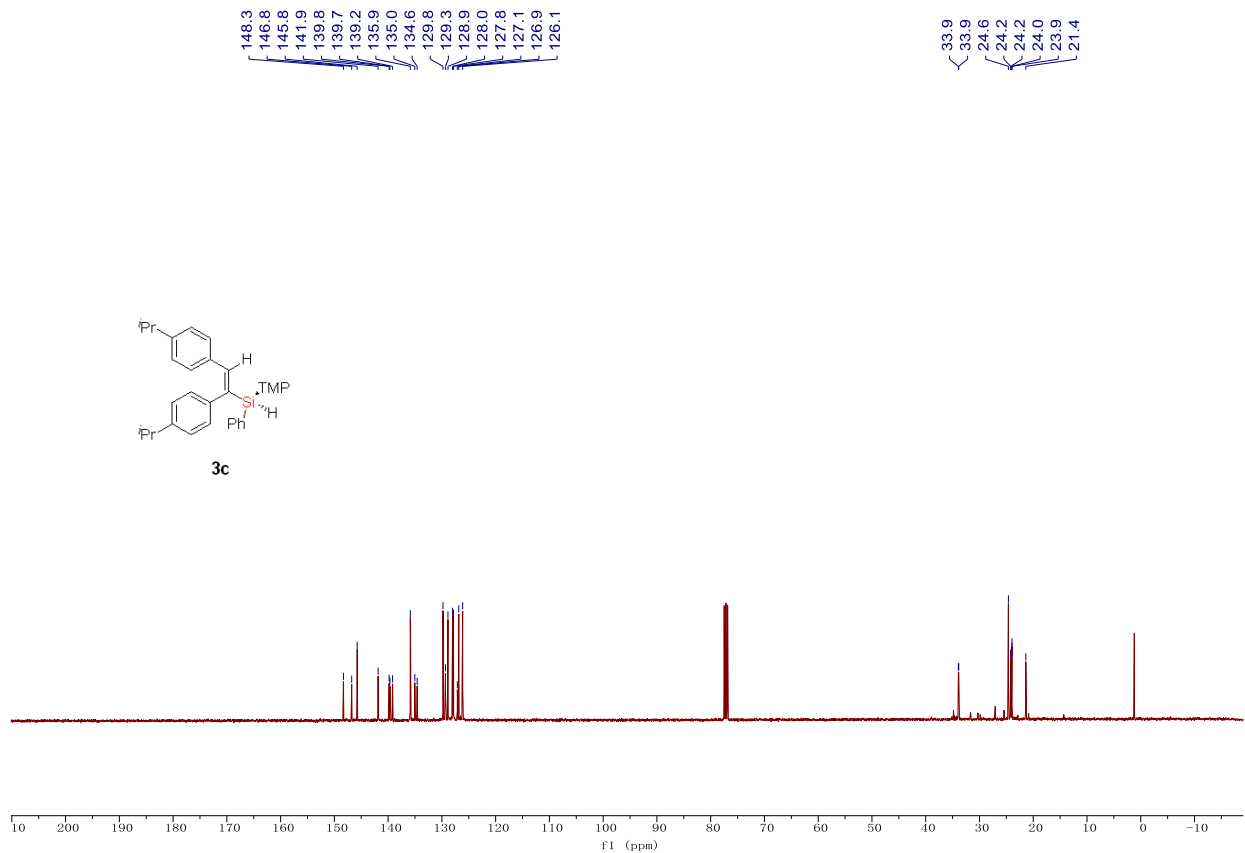
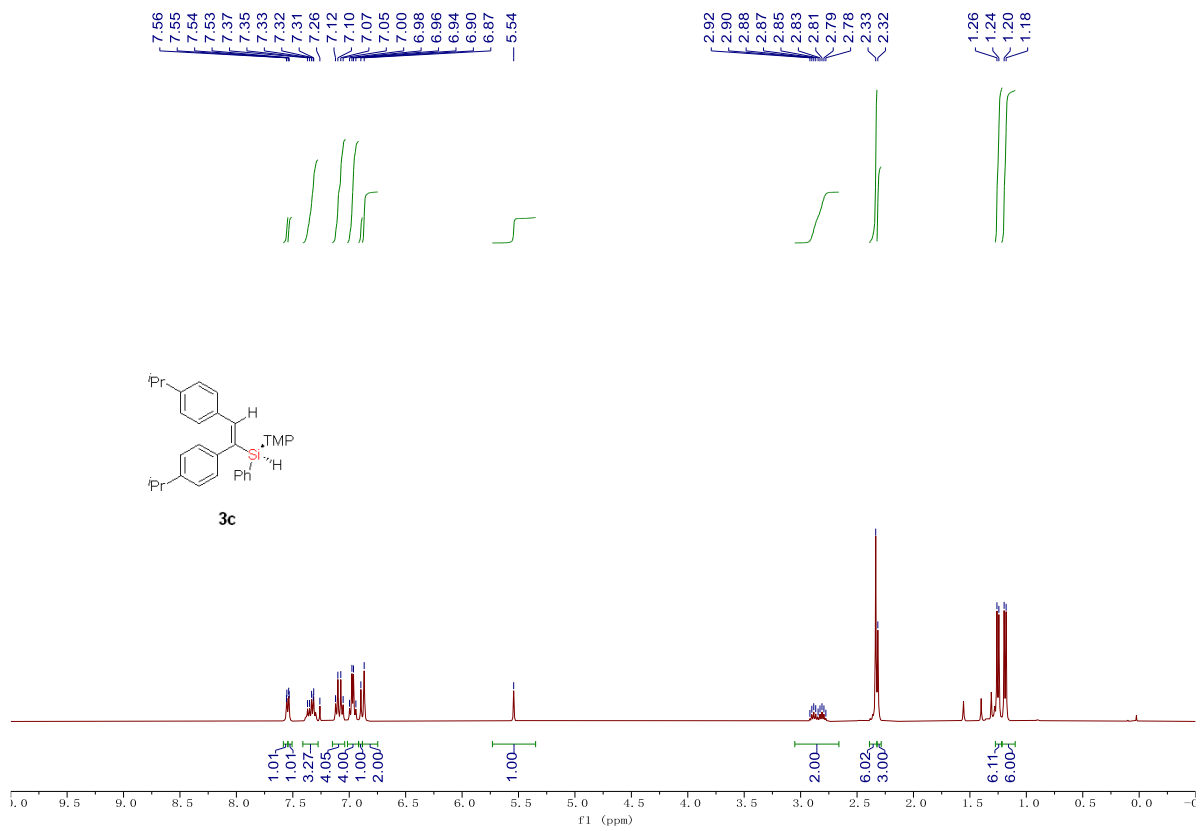


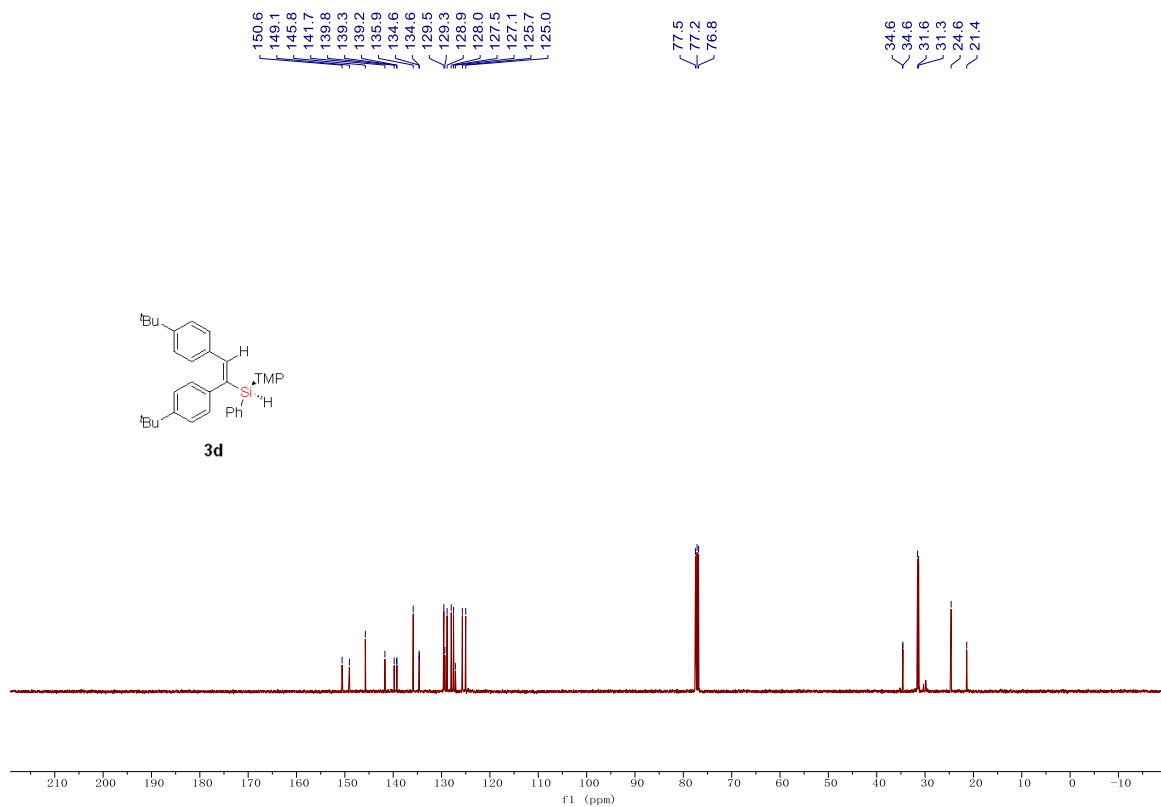
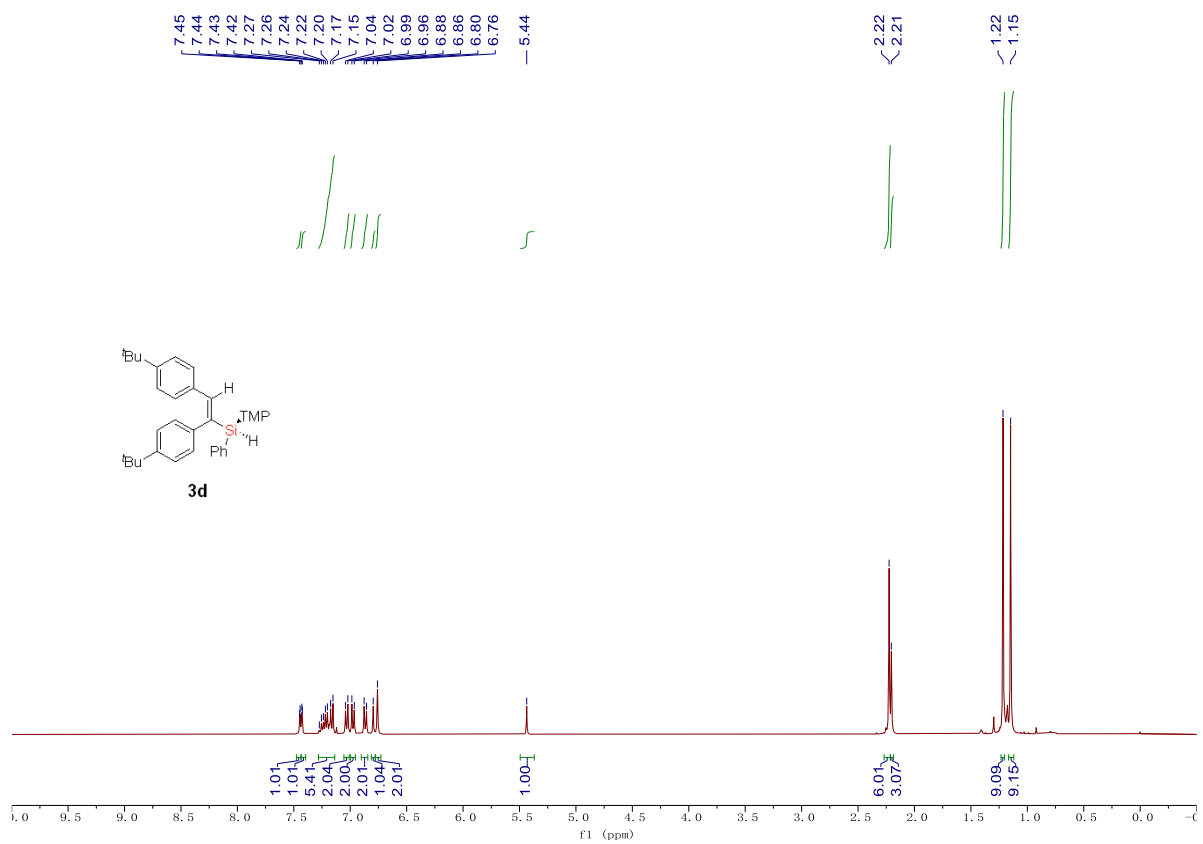


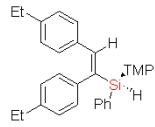
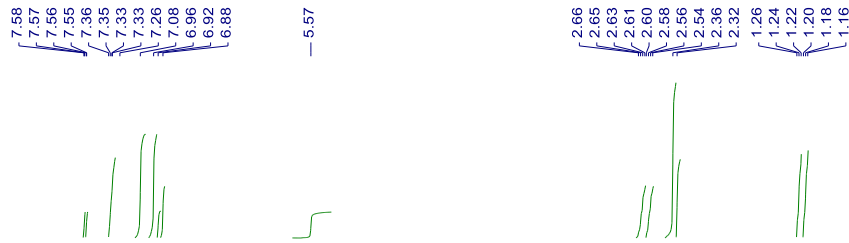




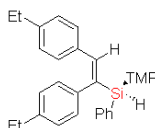
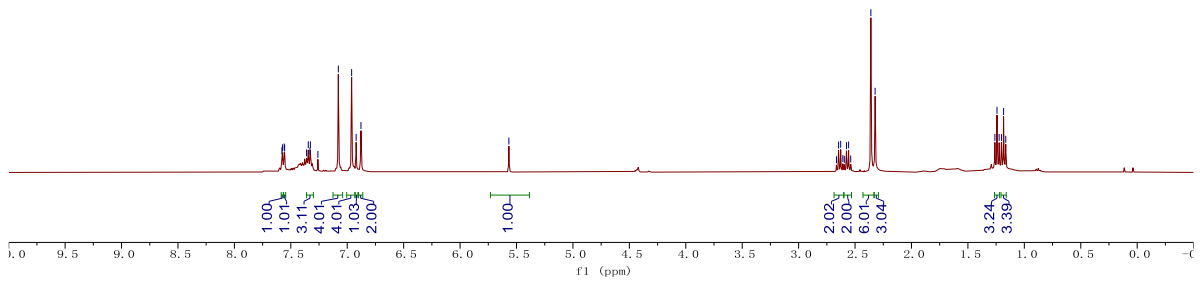




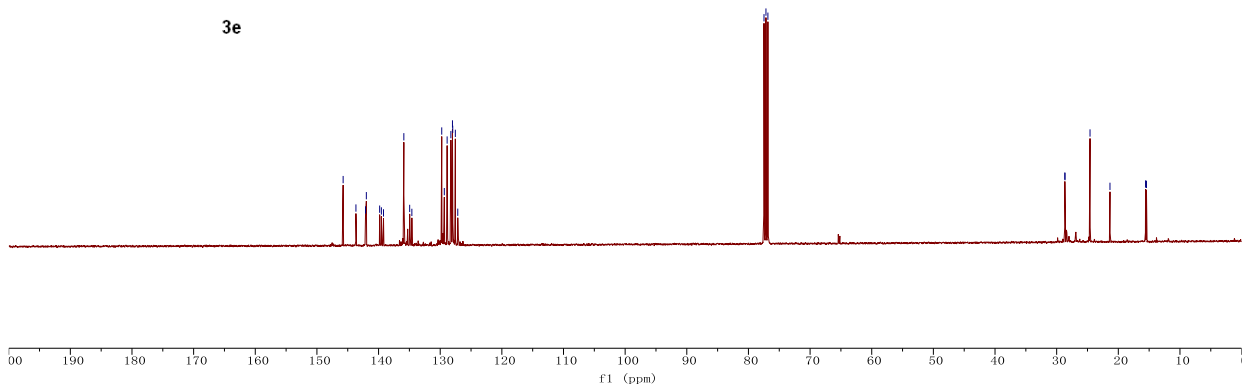


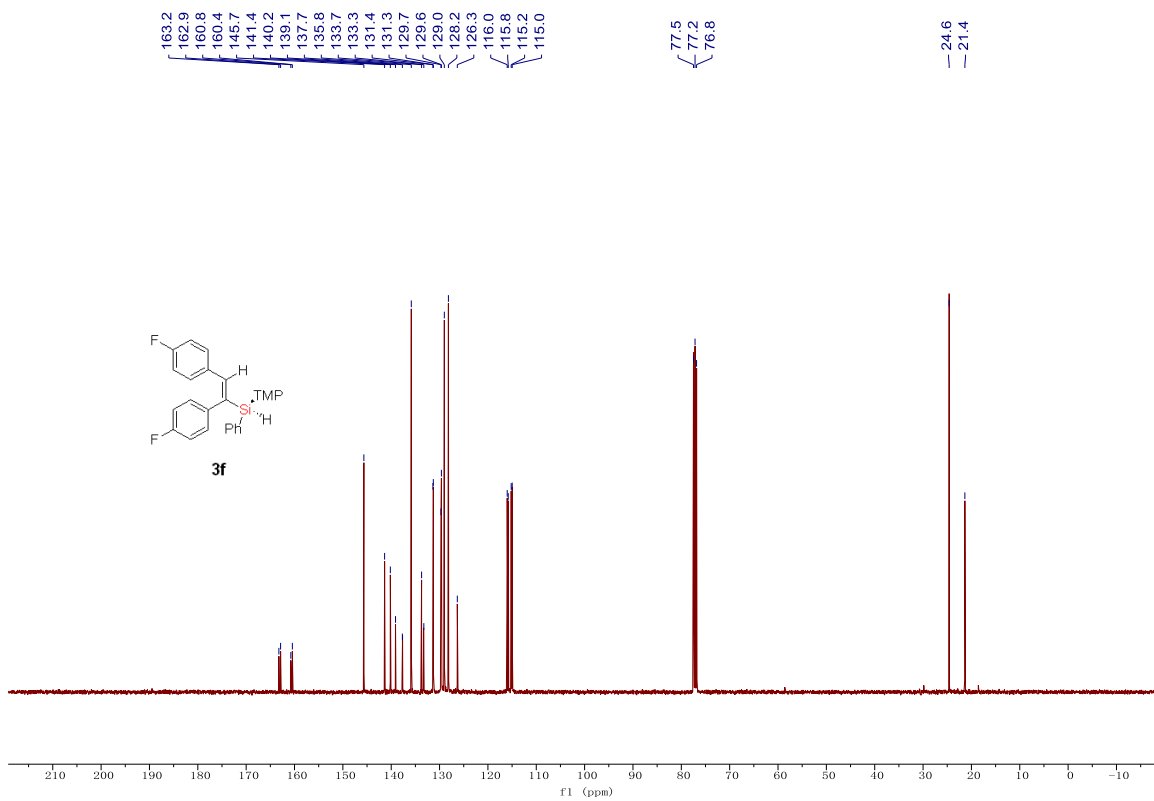
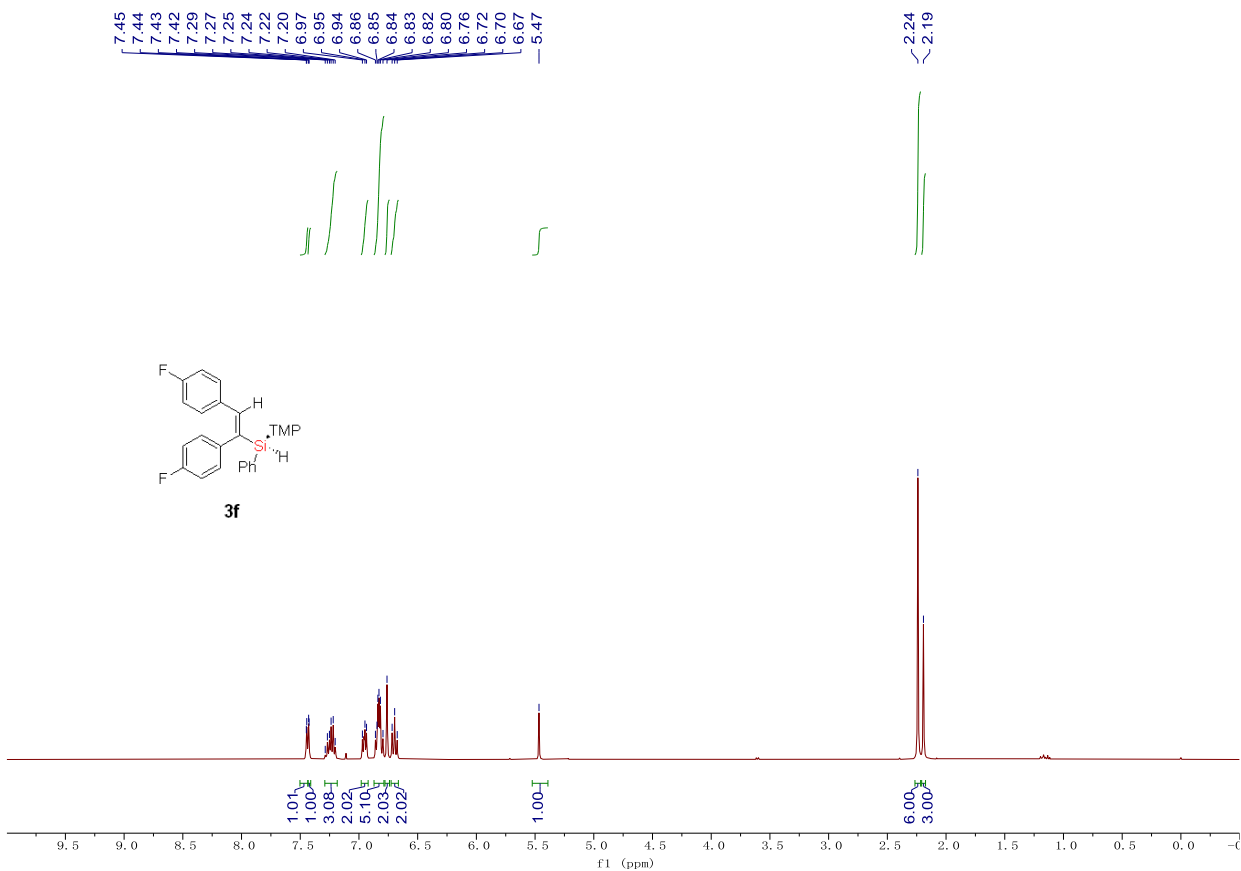


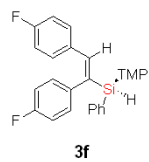
3e



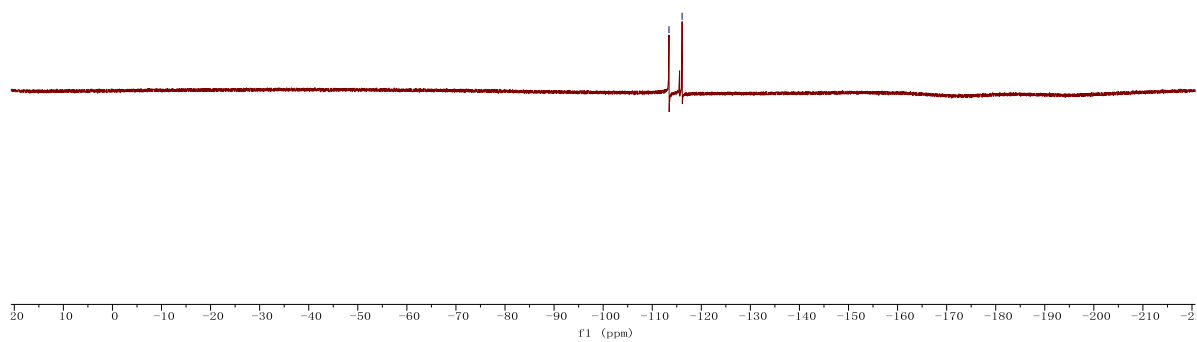
3e

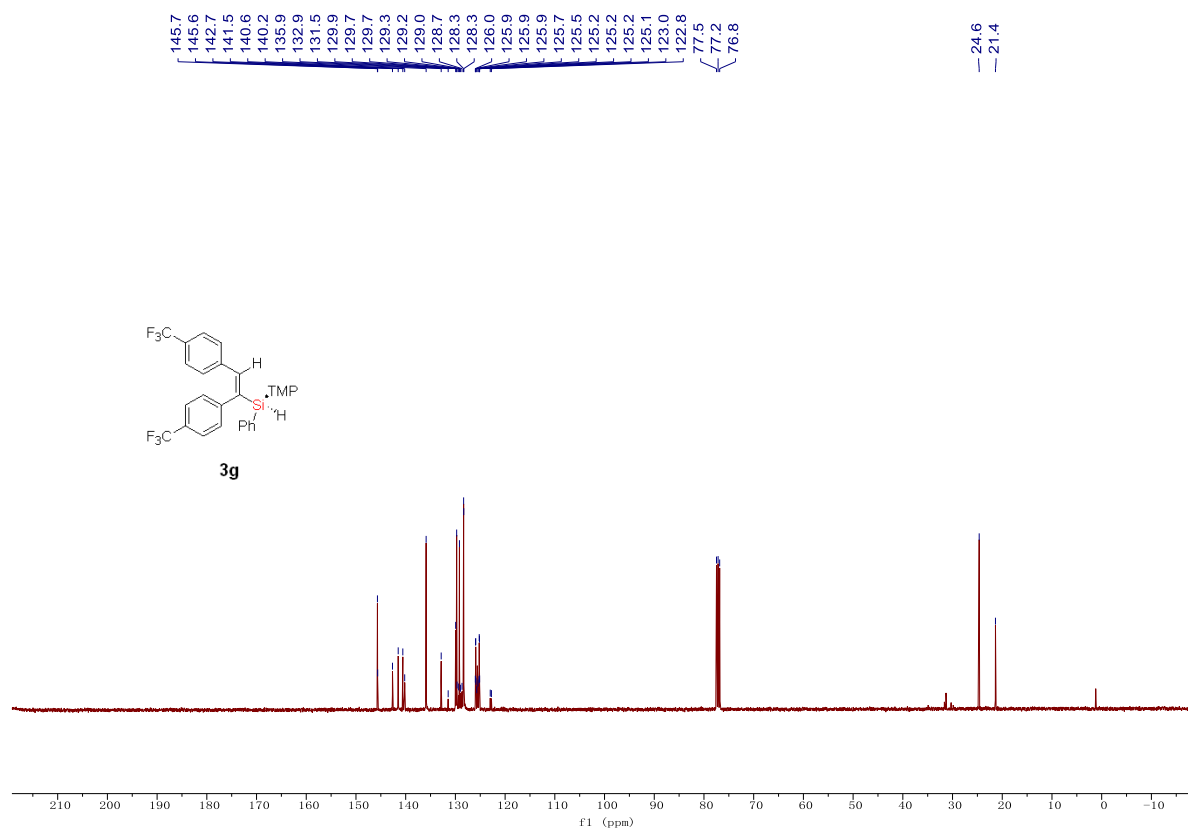
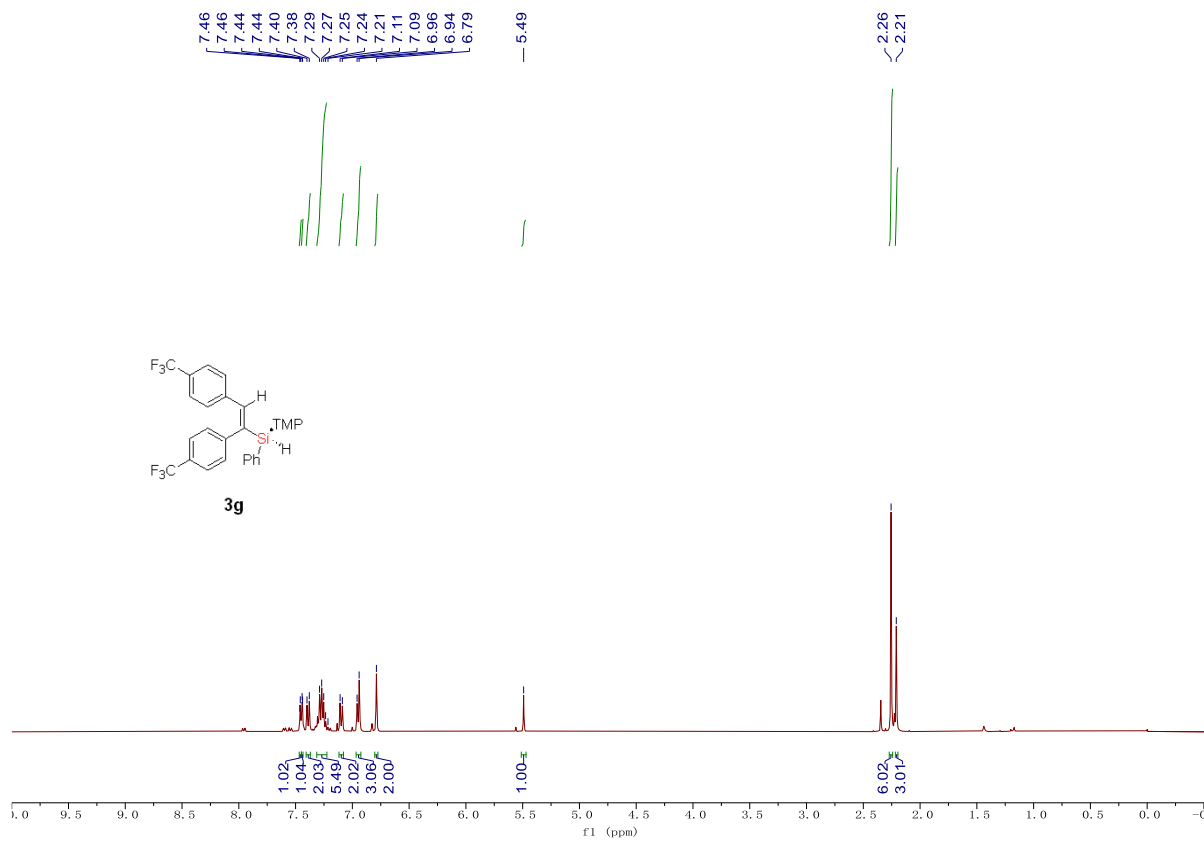


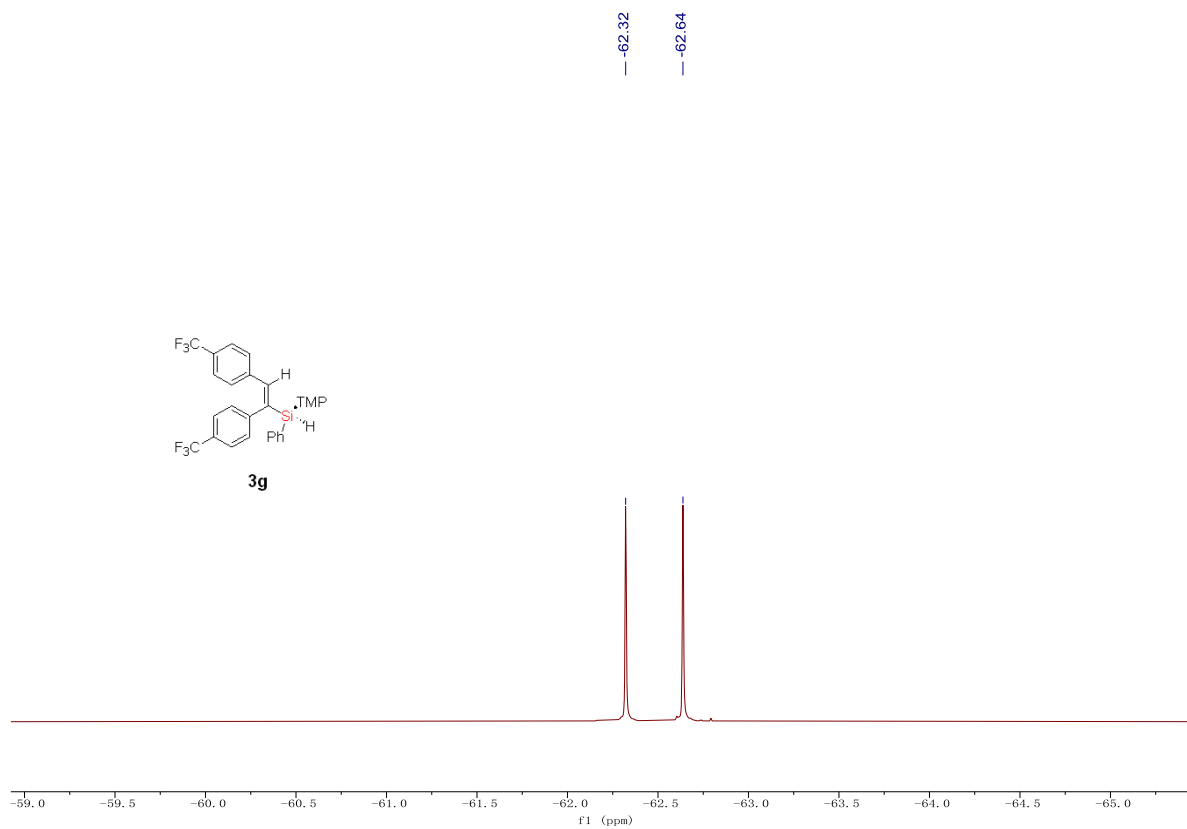


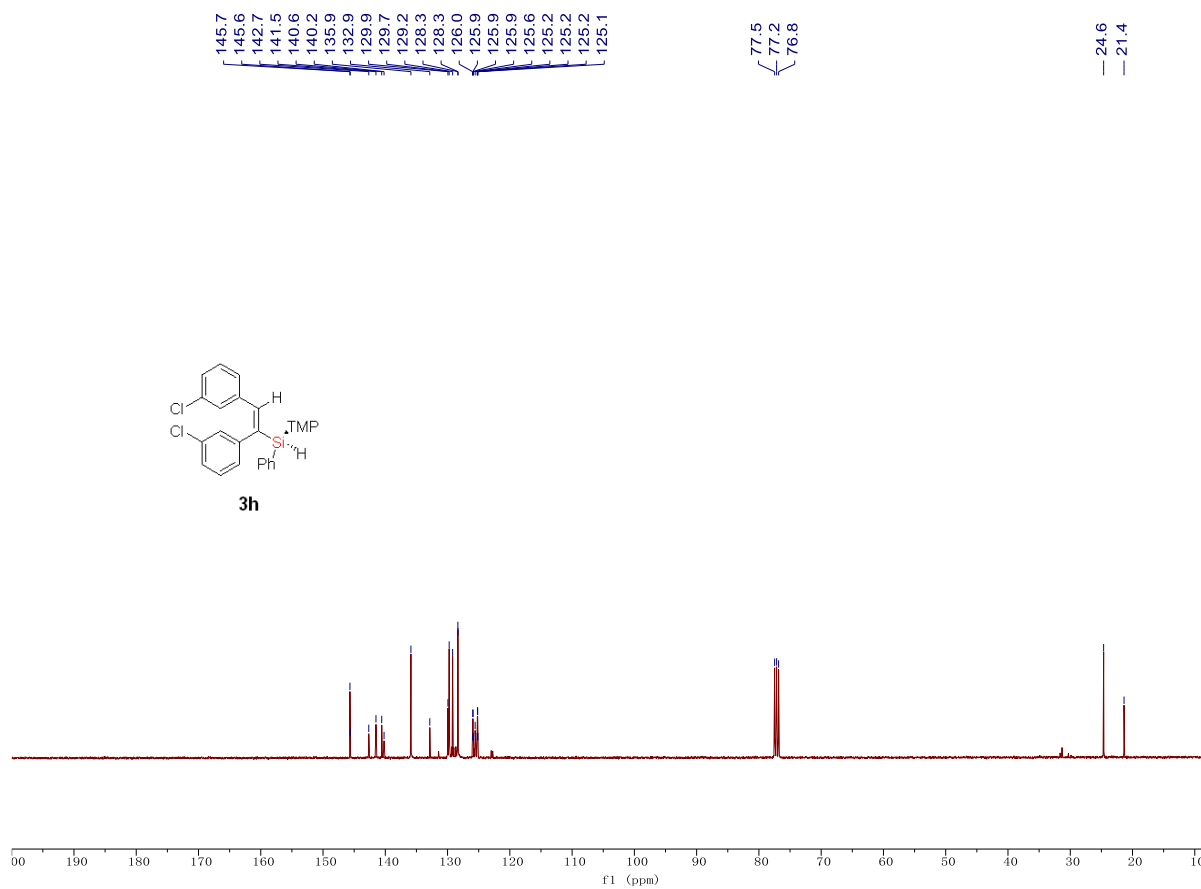
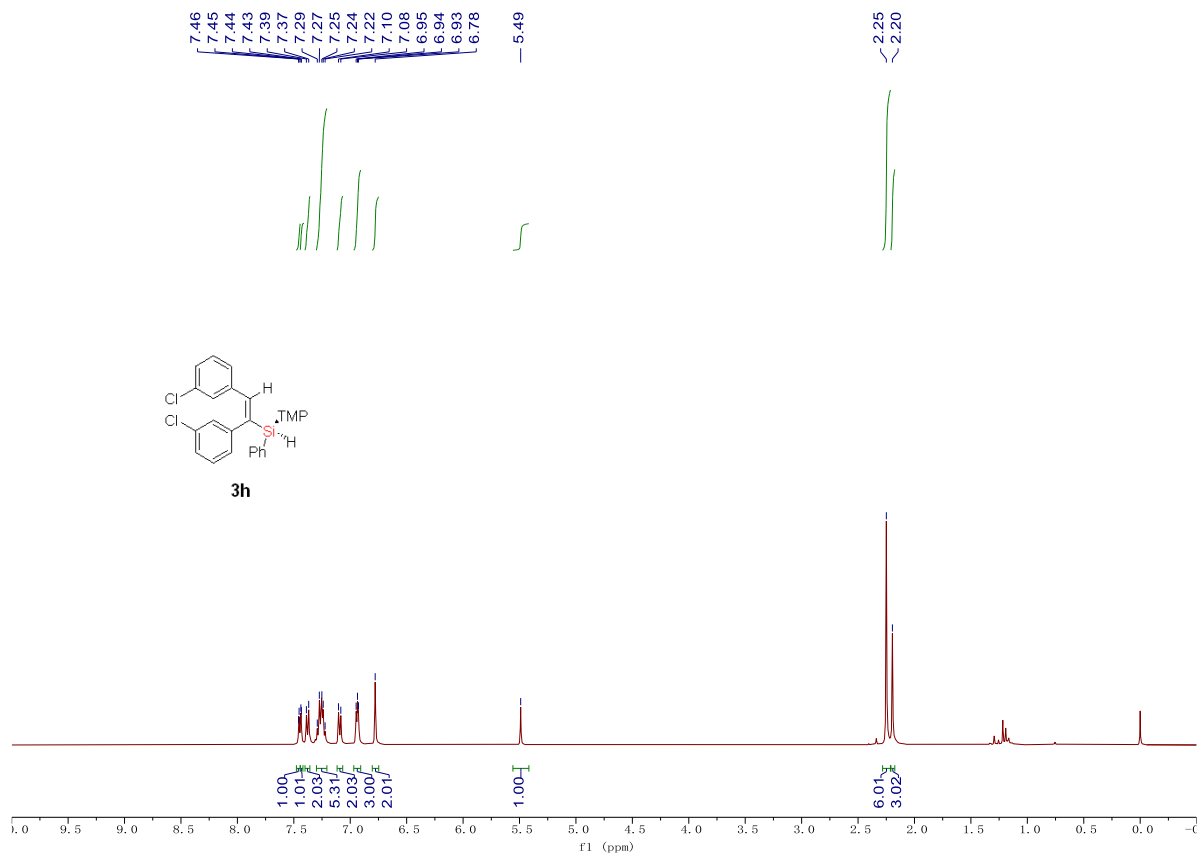


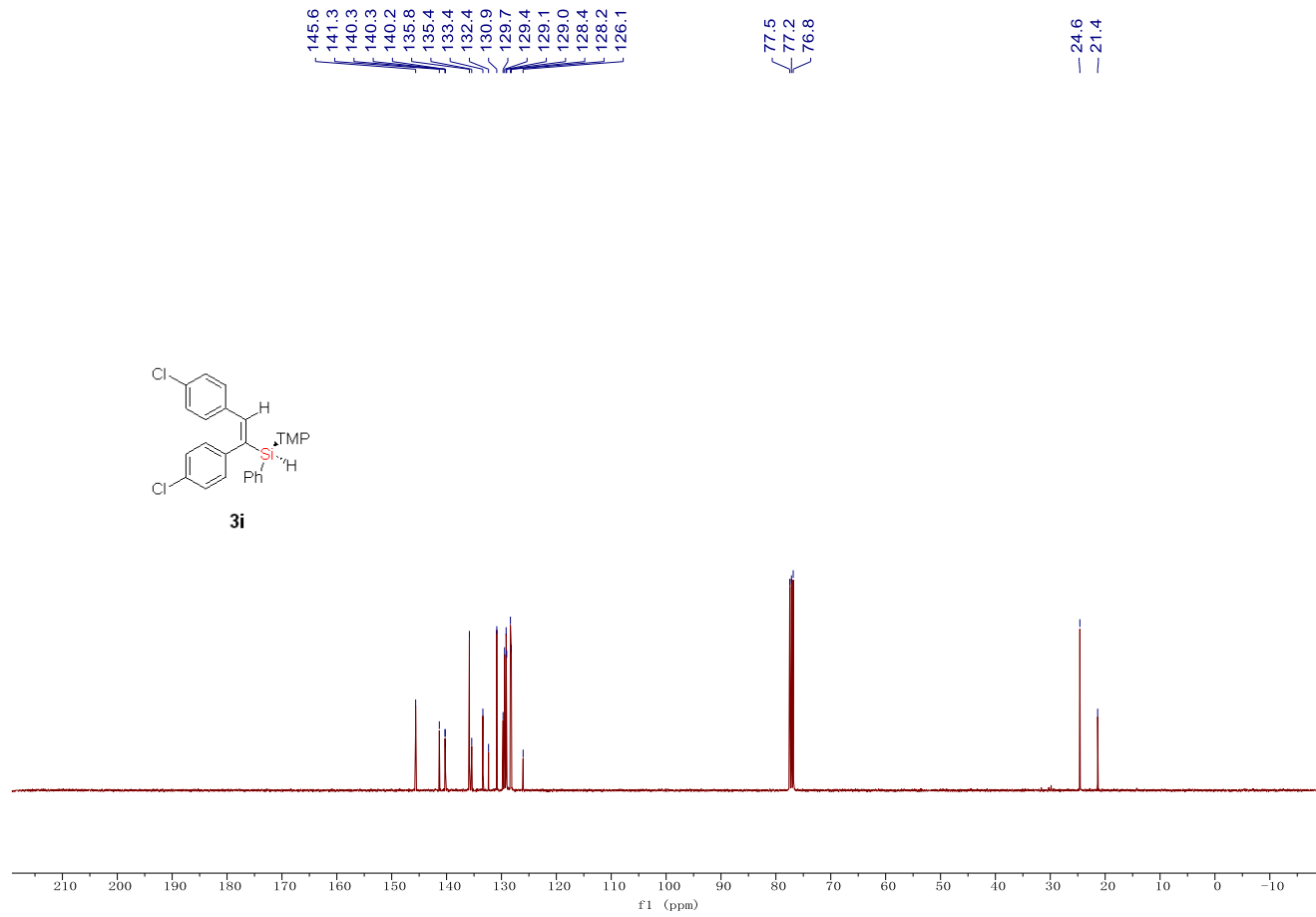
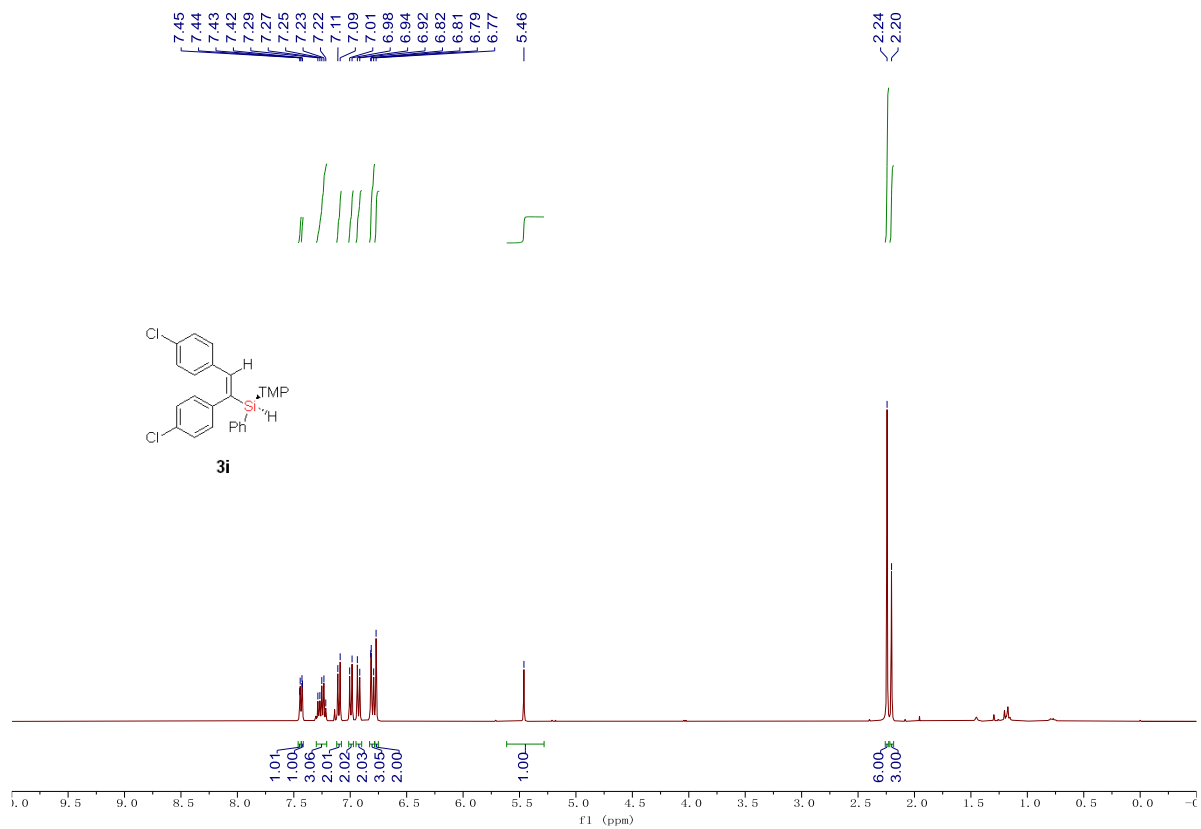
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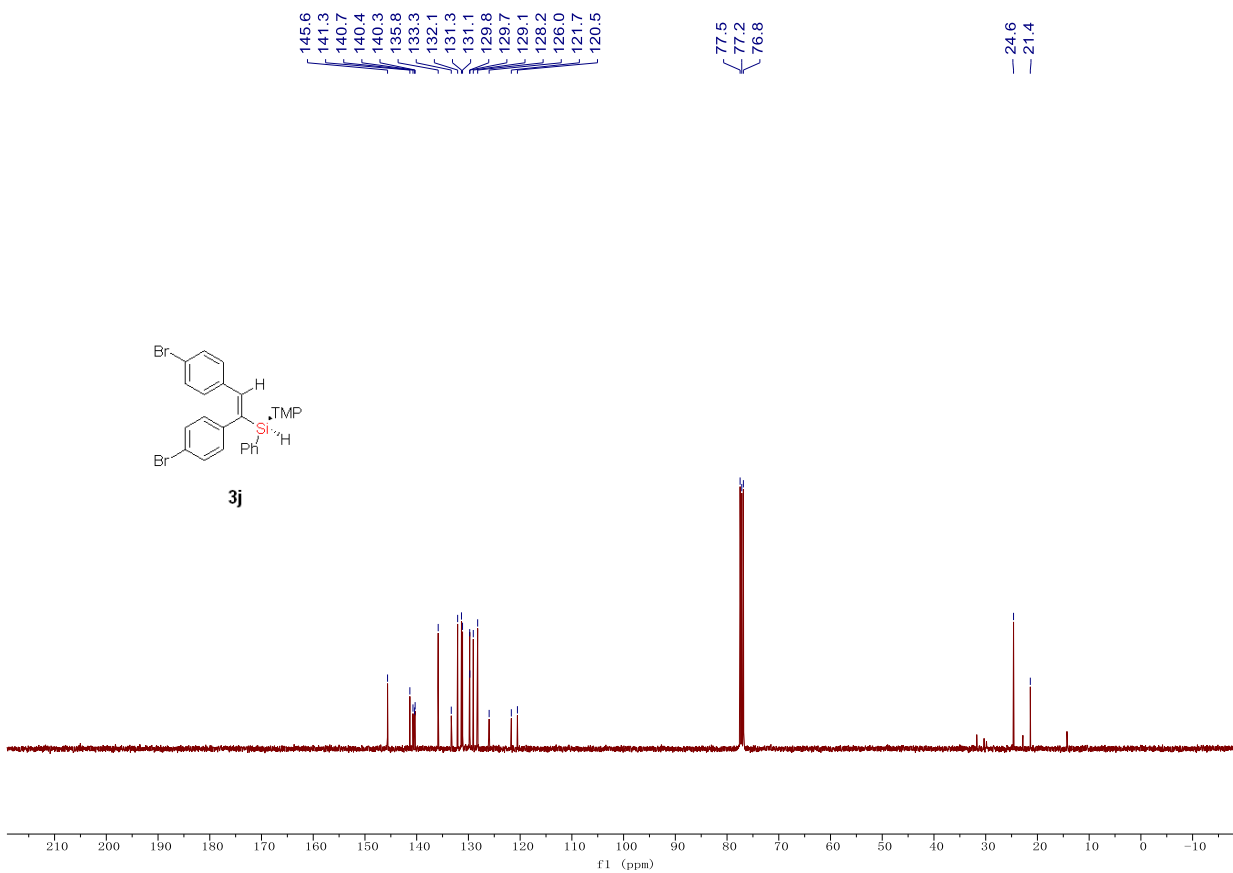
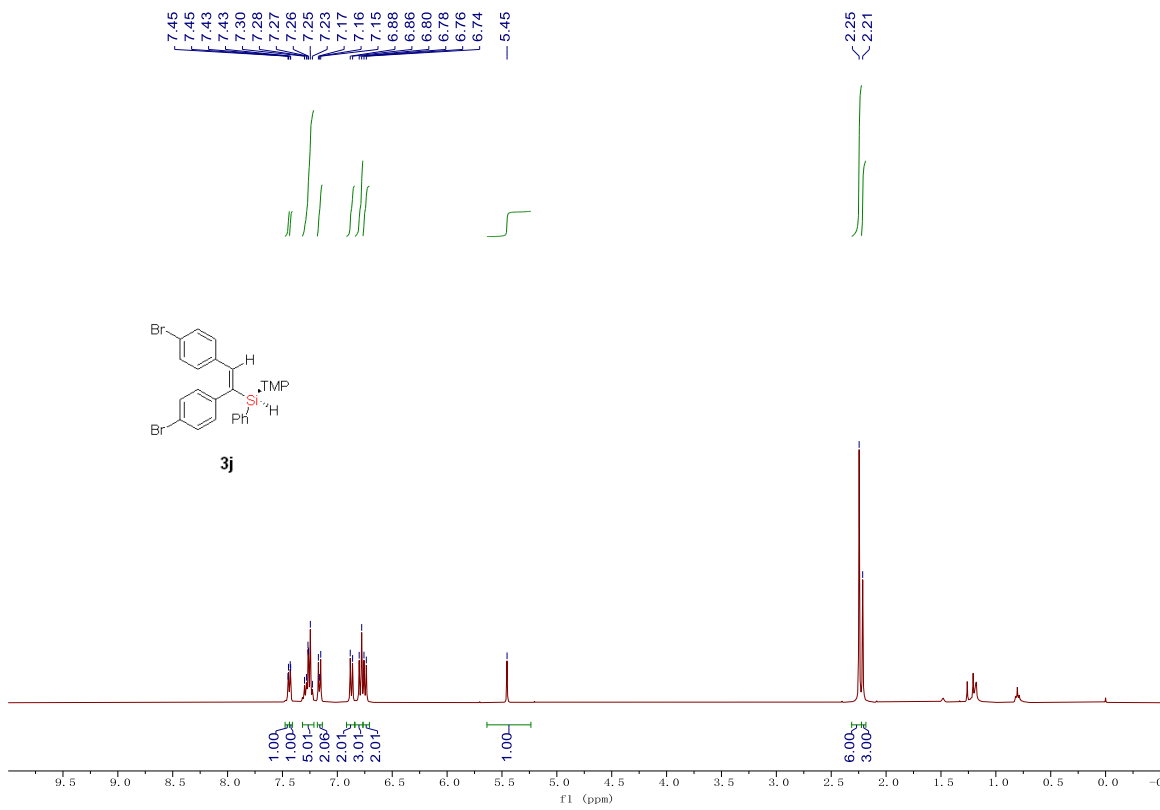


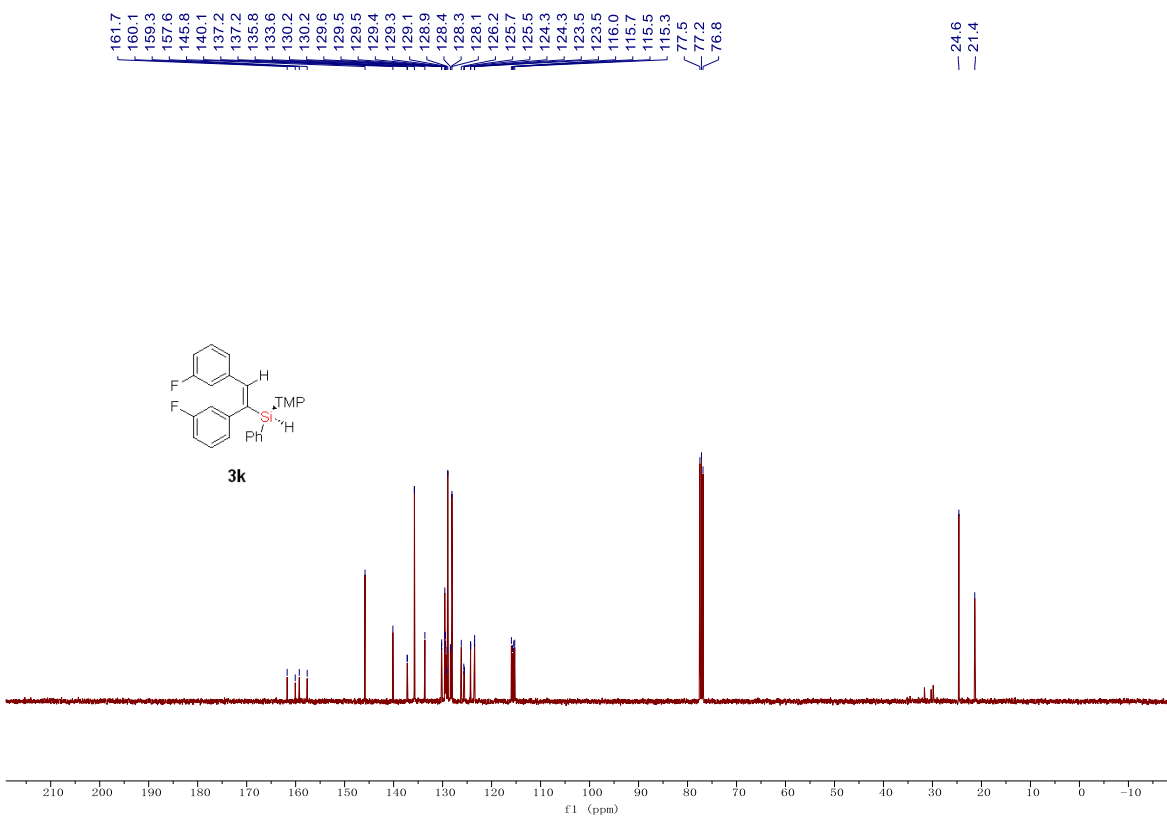
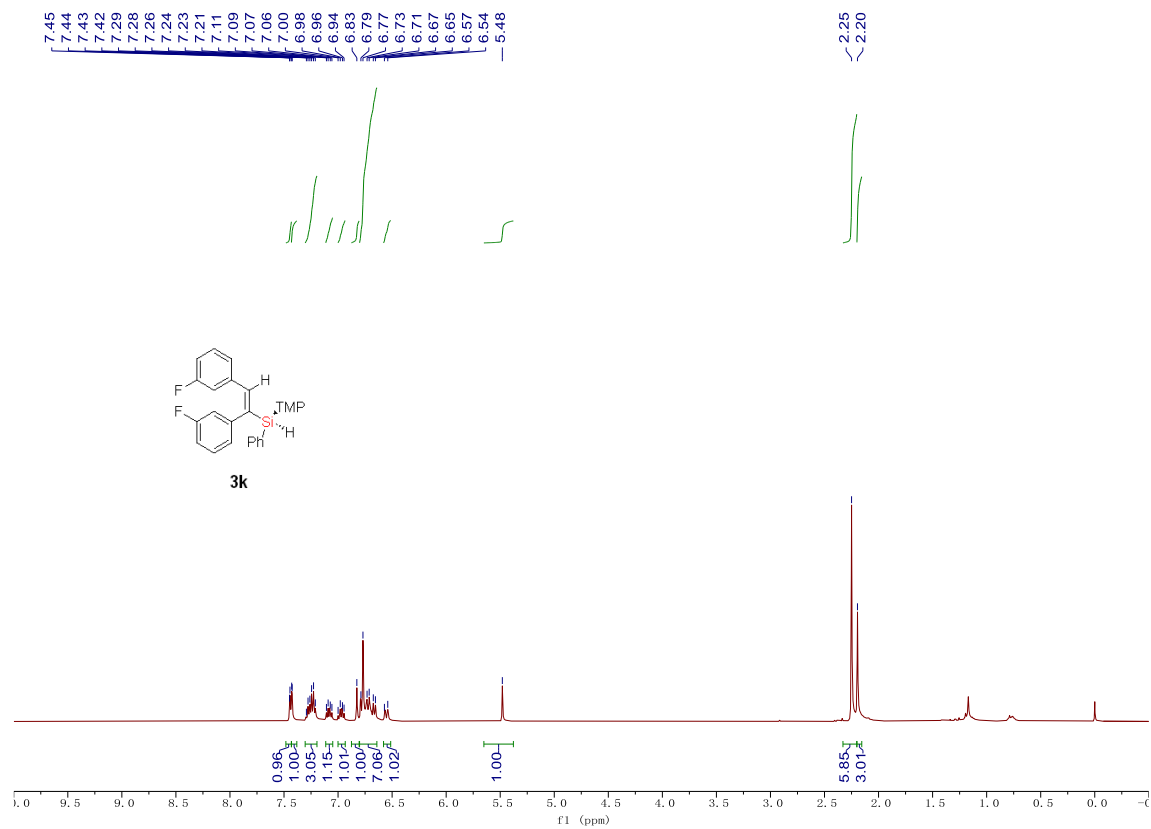


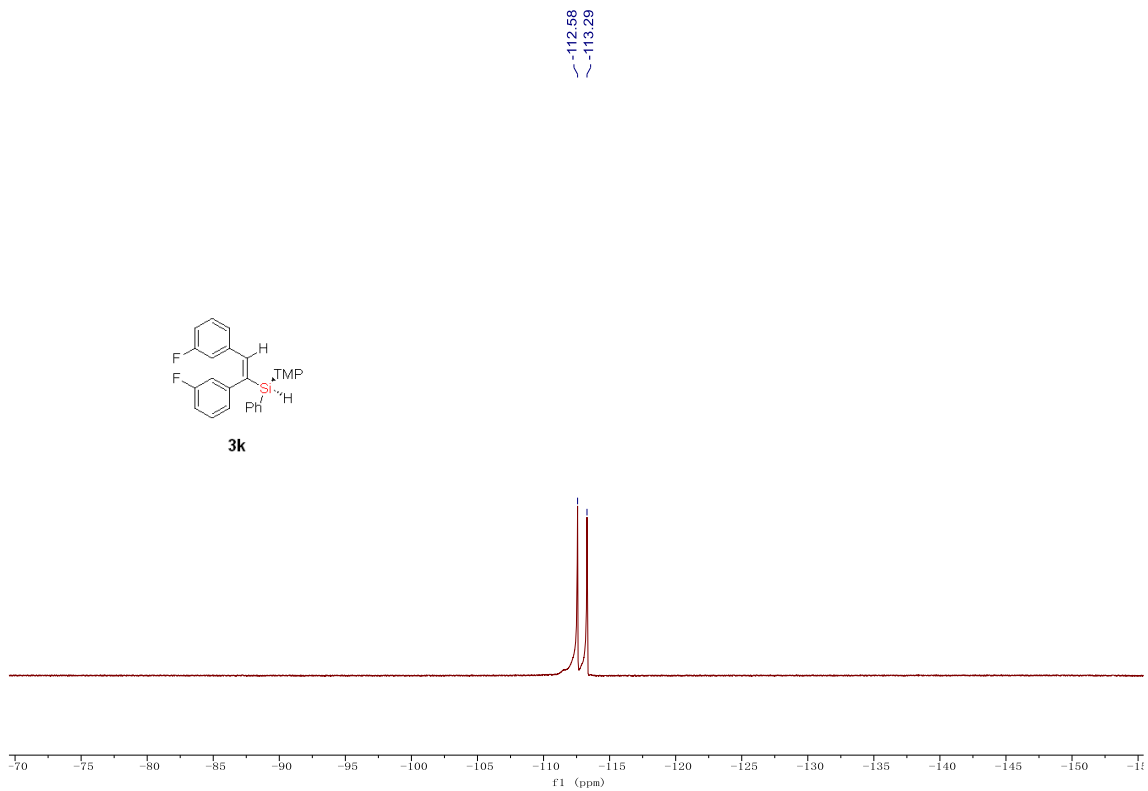
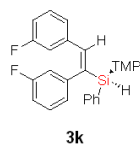


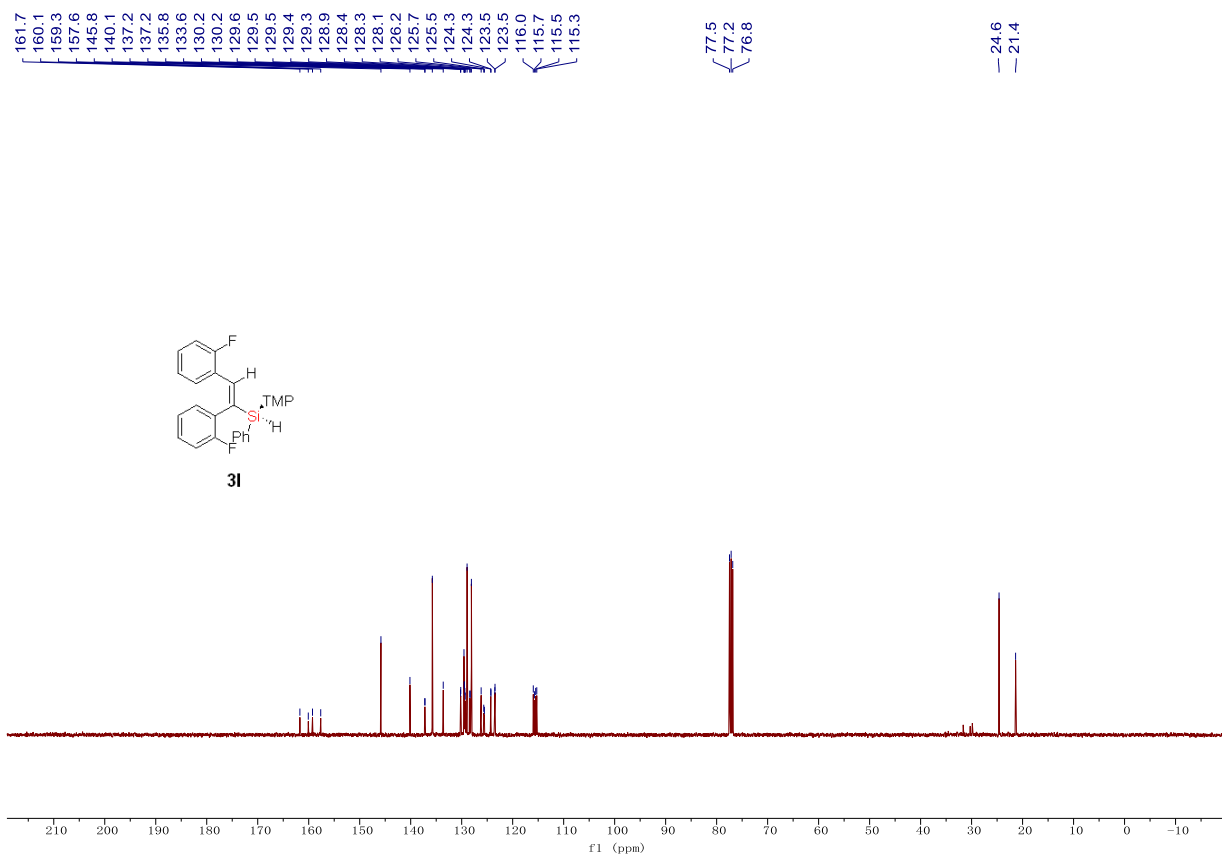
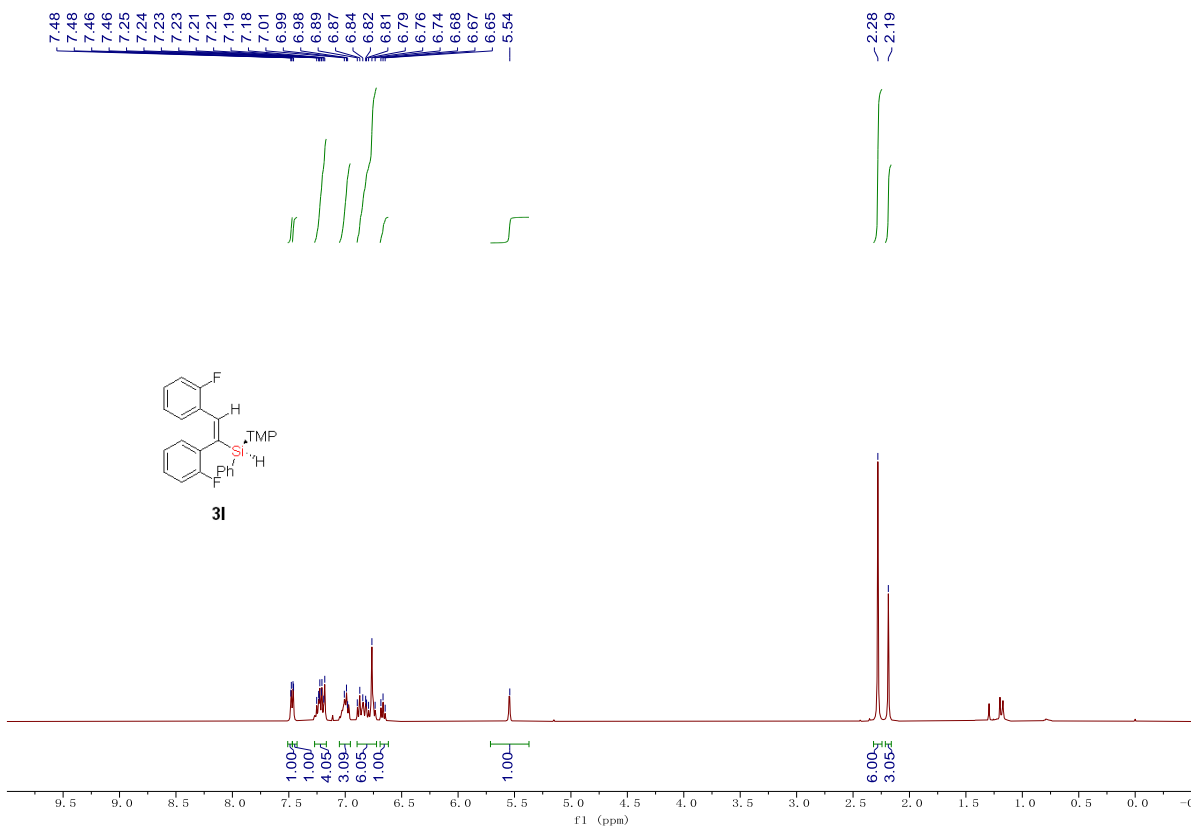




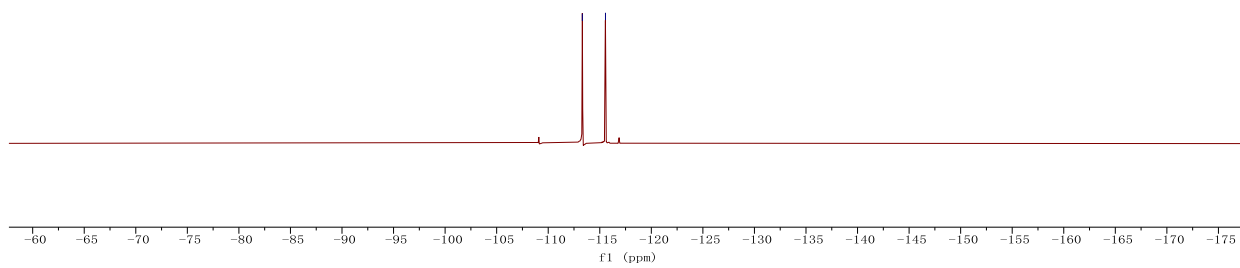
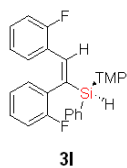






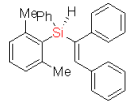


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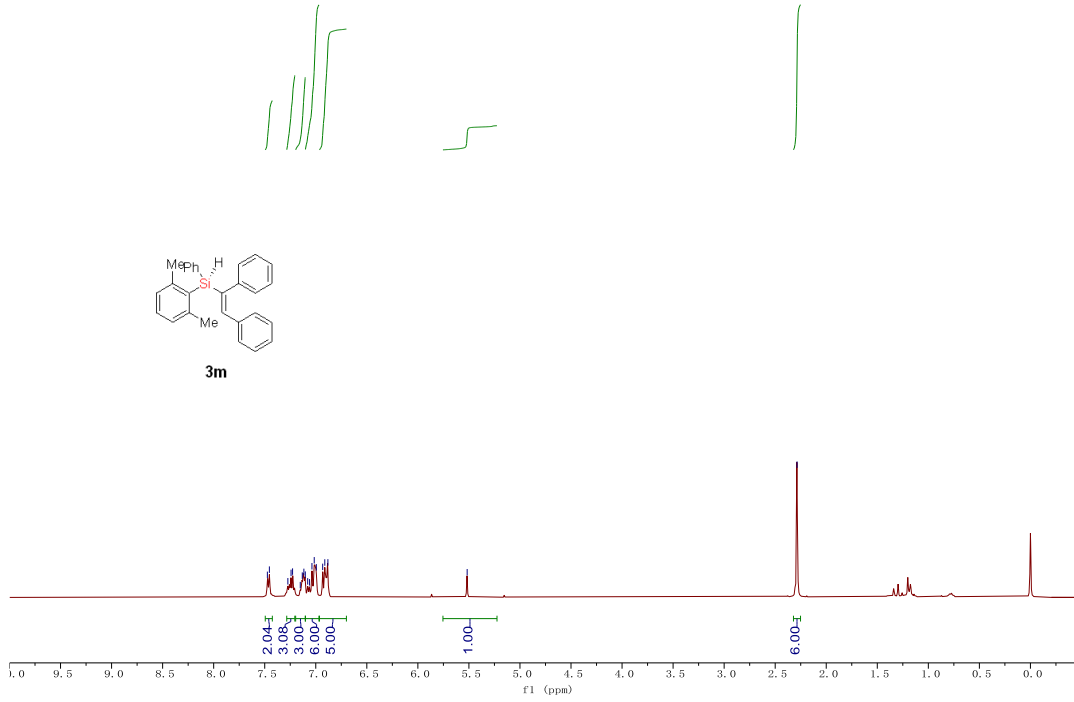


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— 5.52

— 2.29



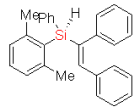
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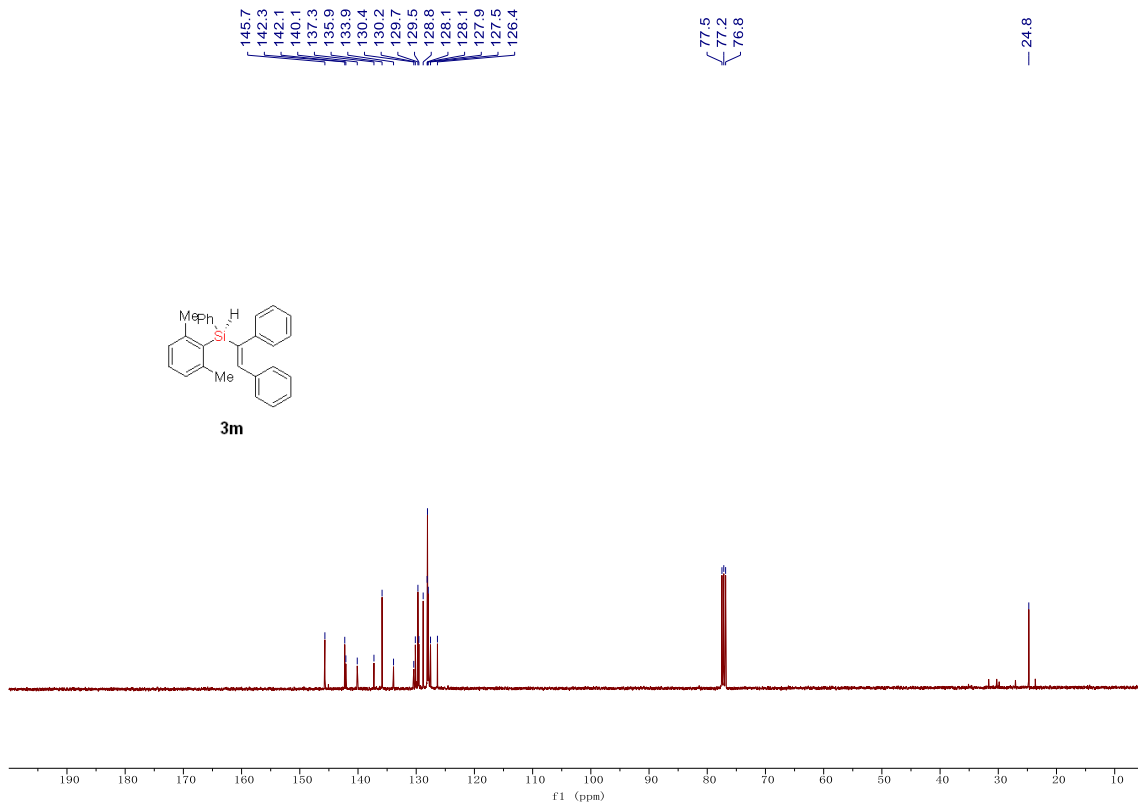
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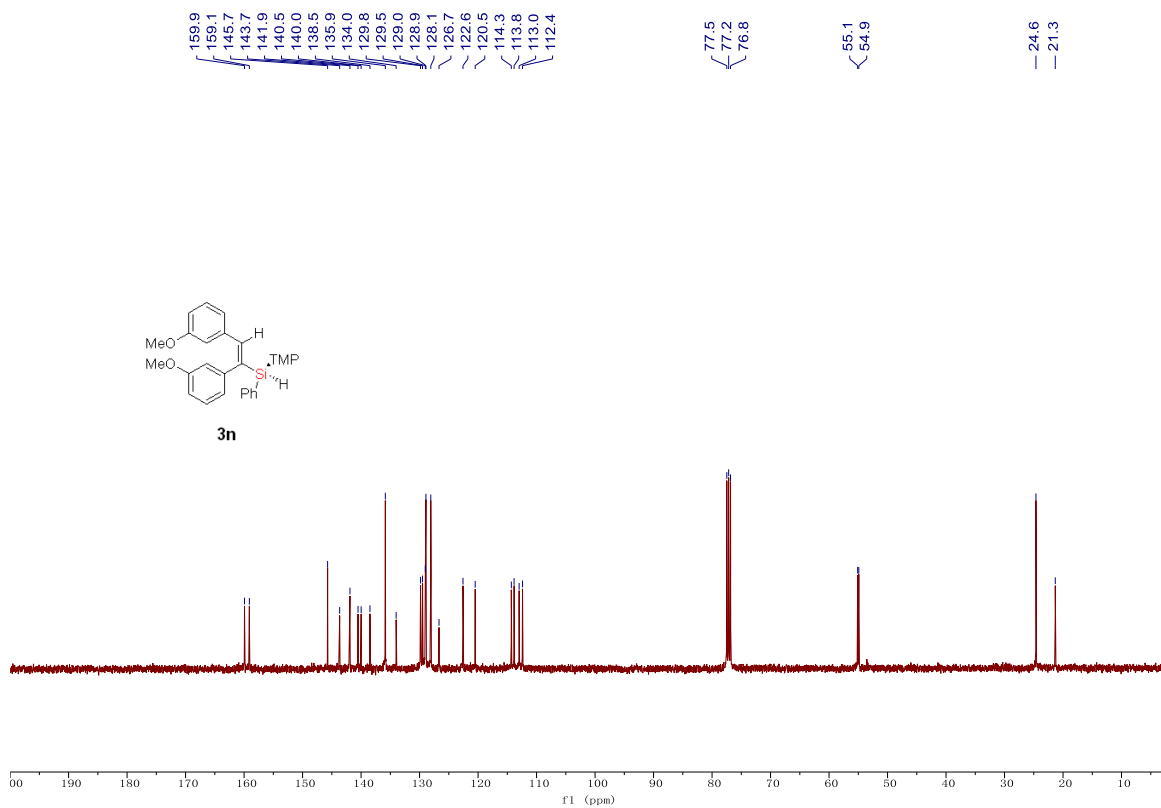
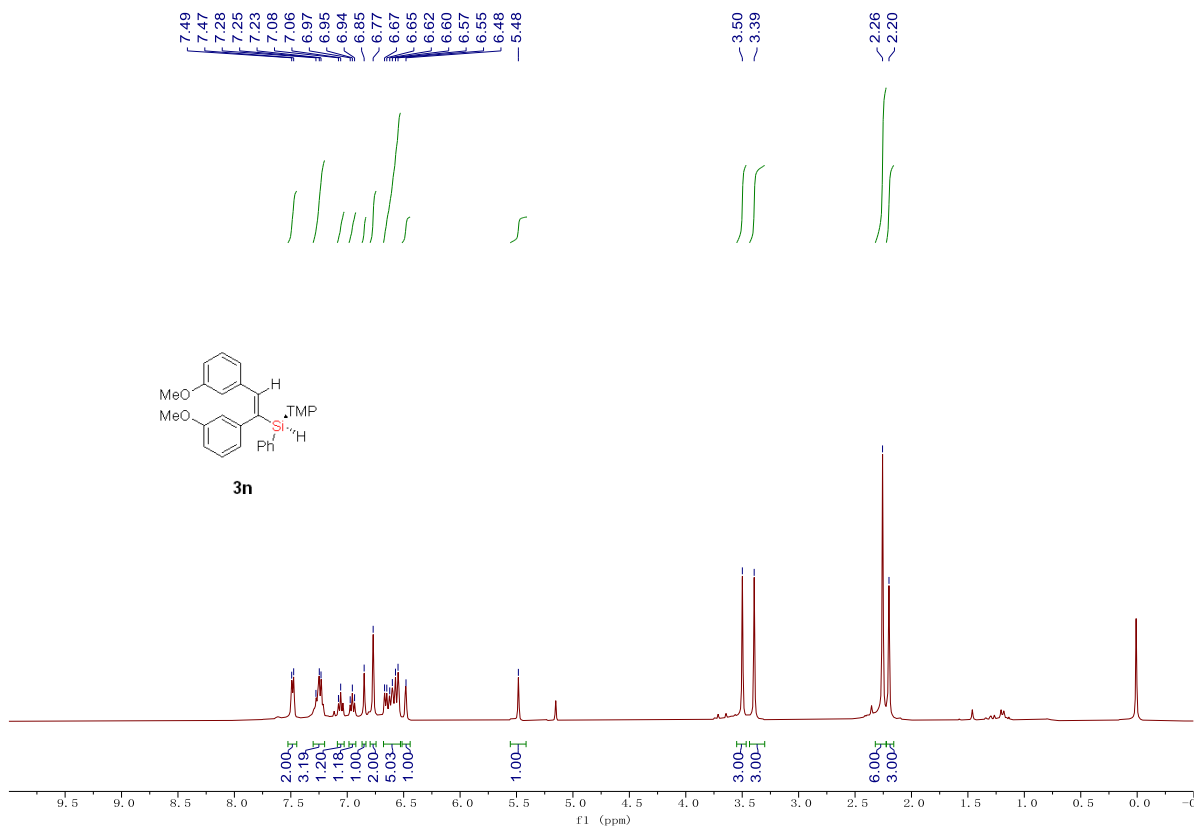
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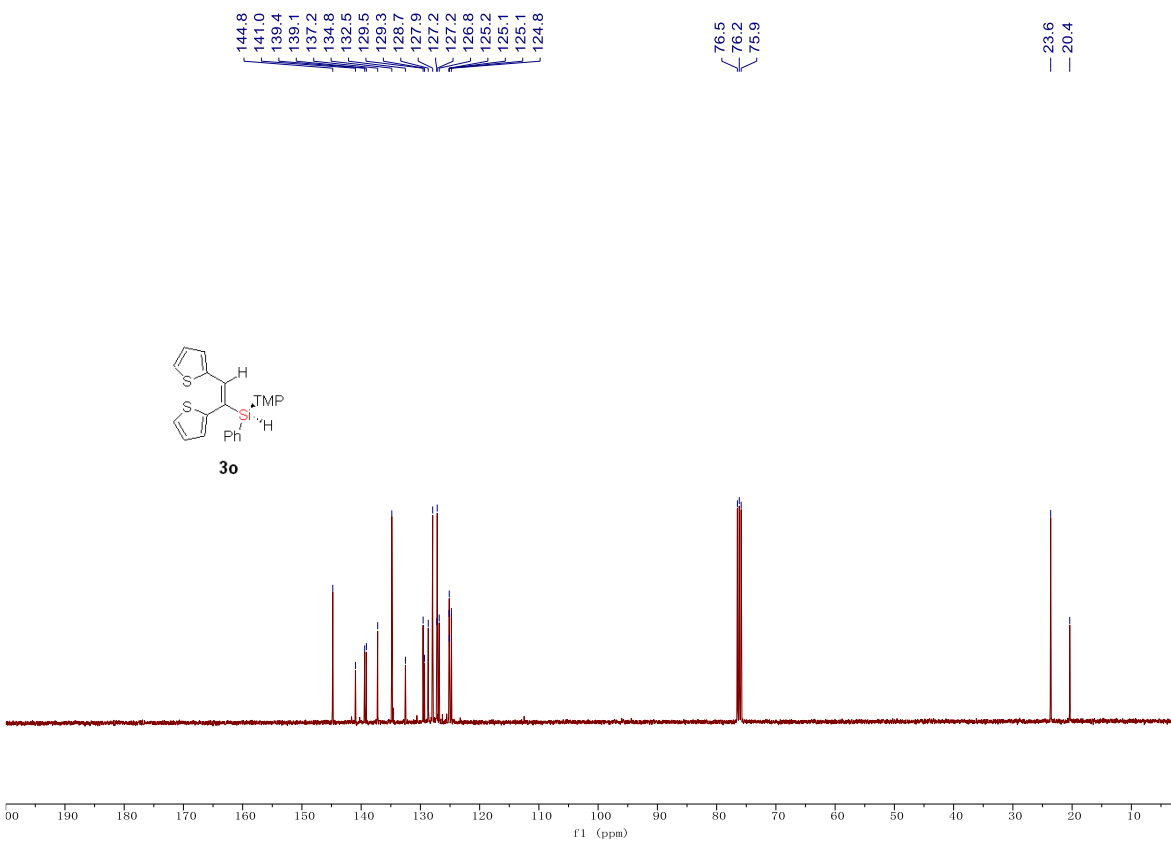
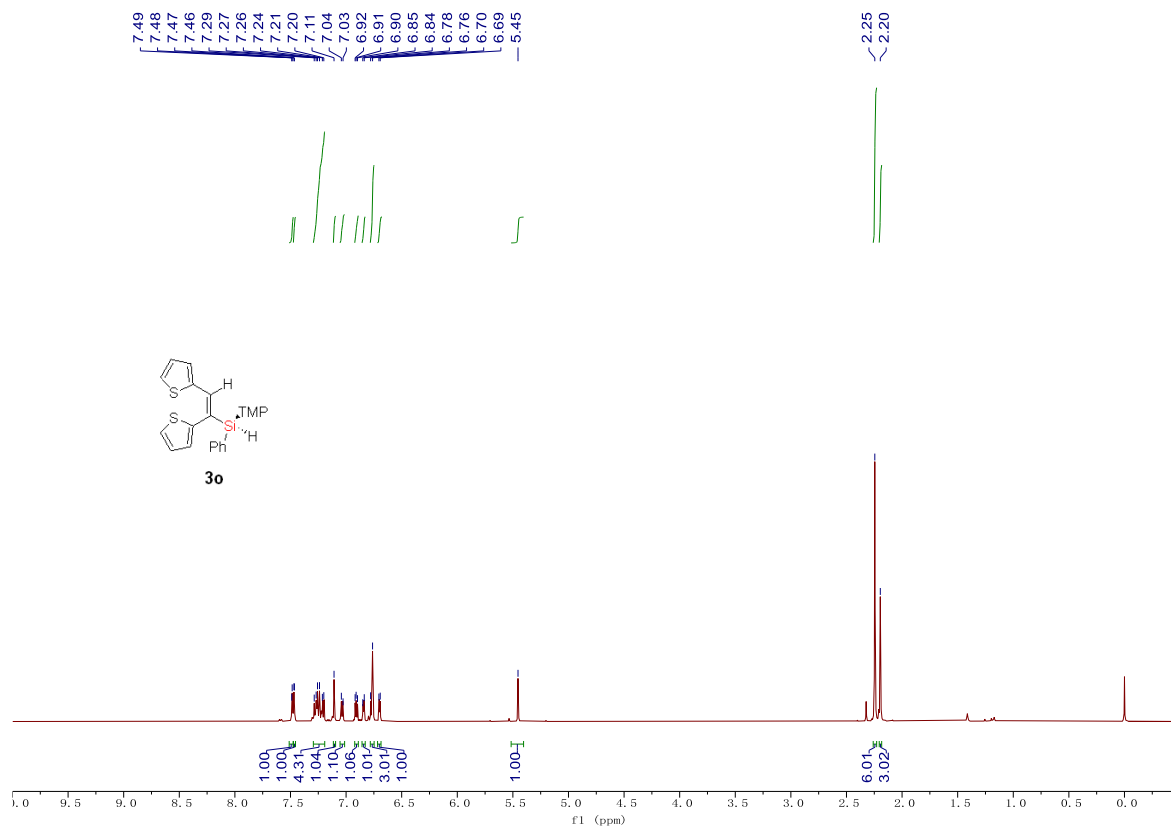
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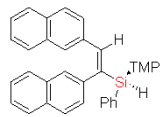
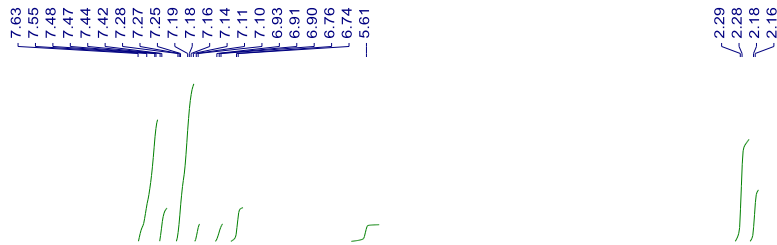


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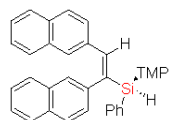
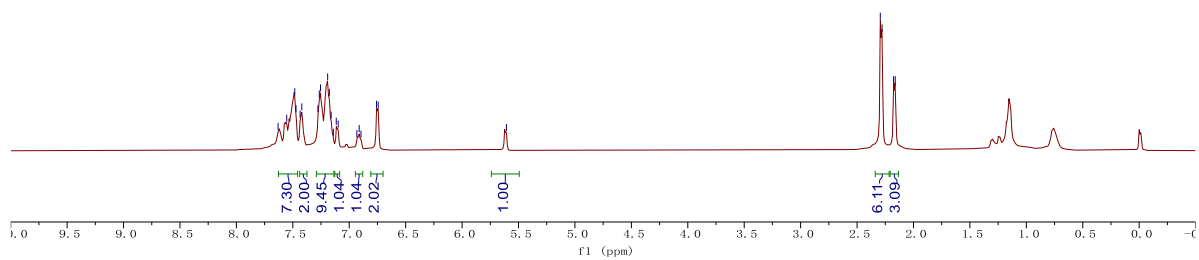




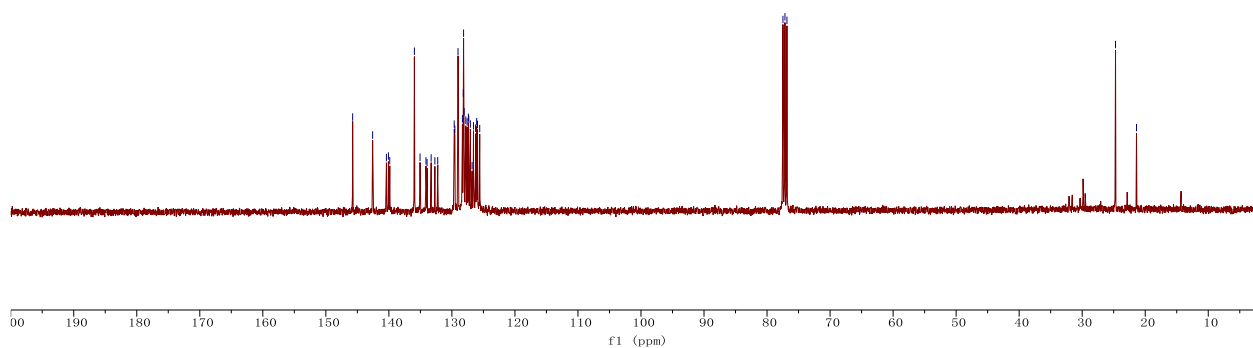


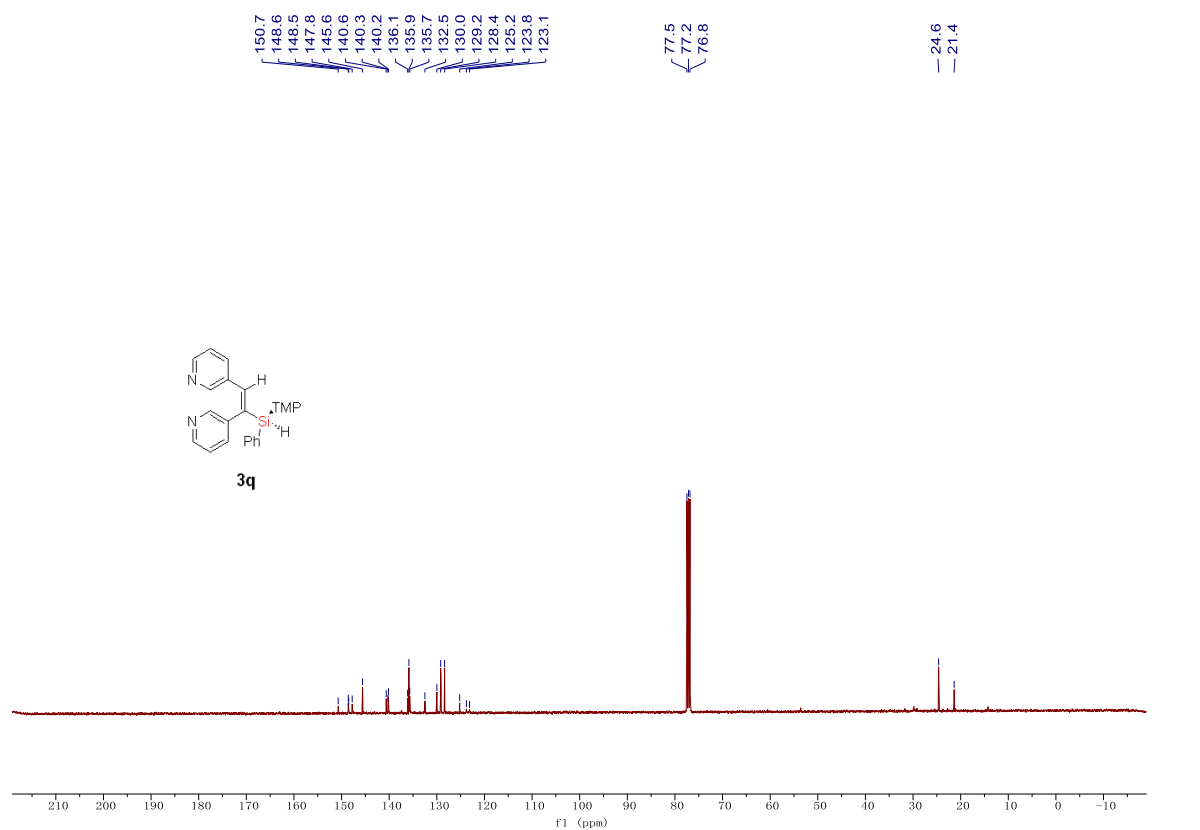
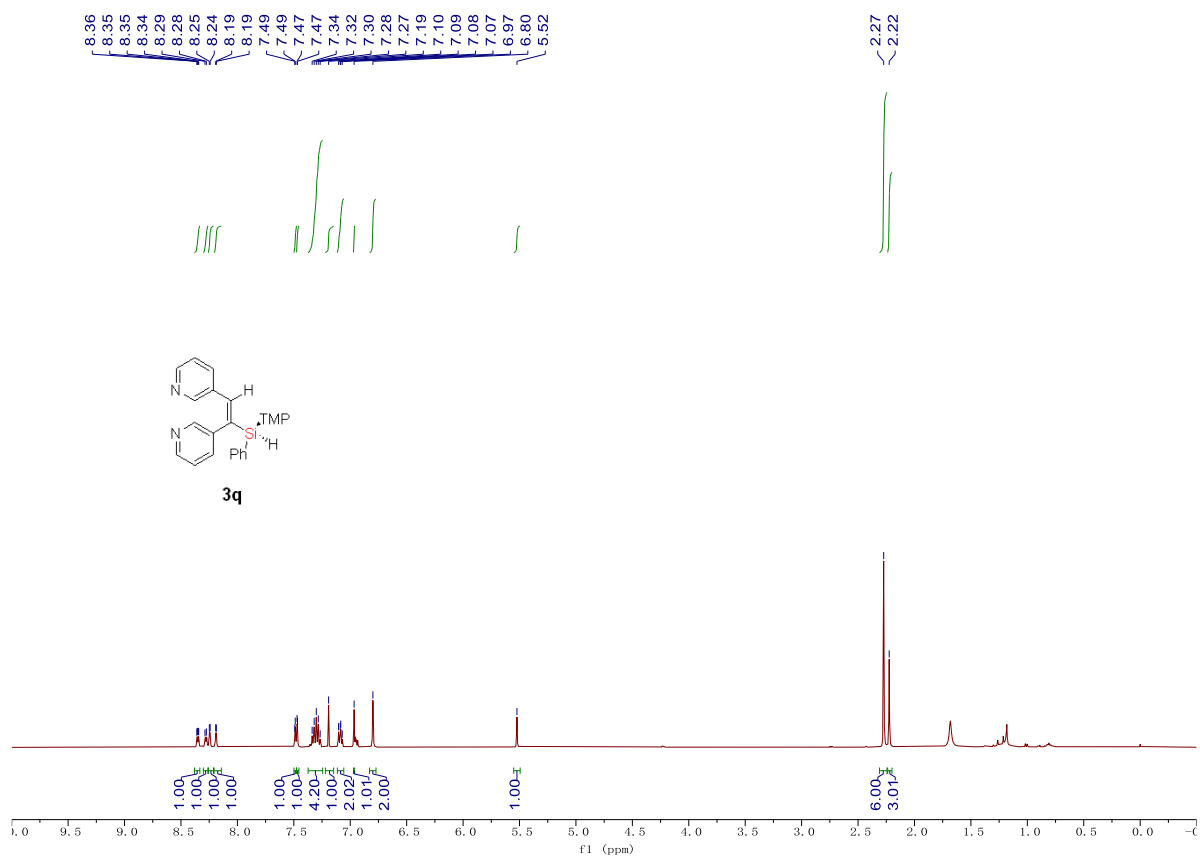


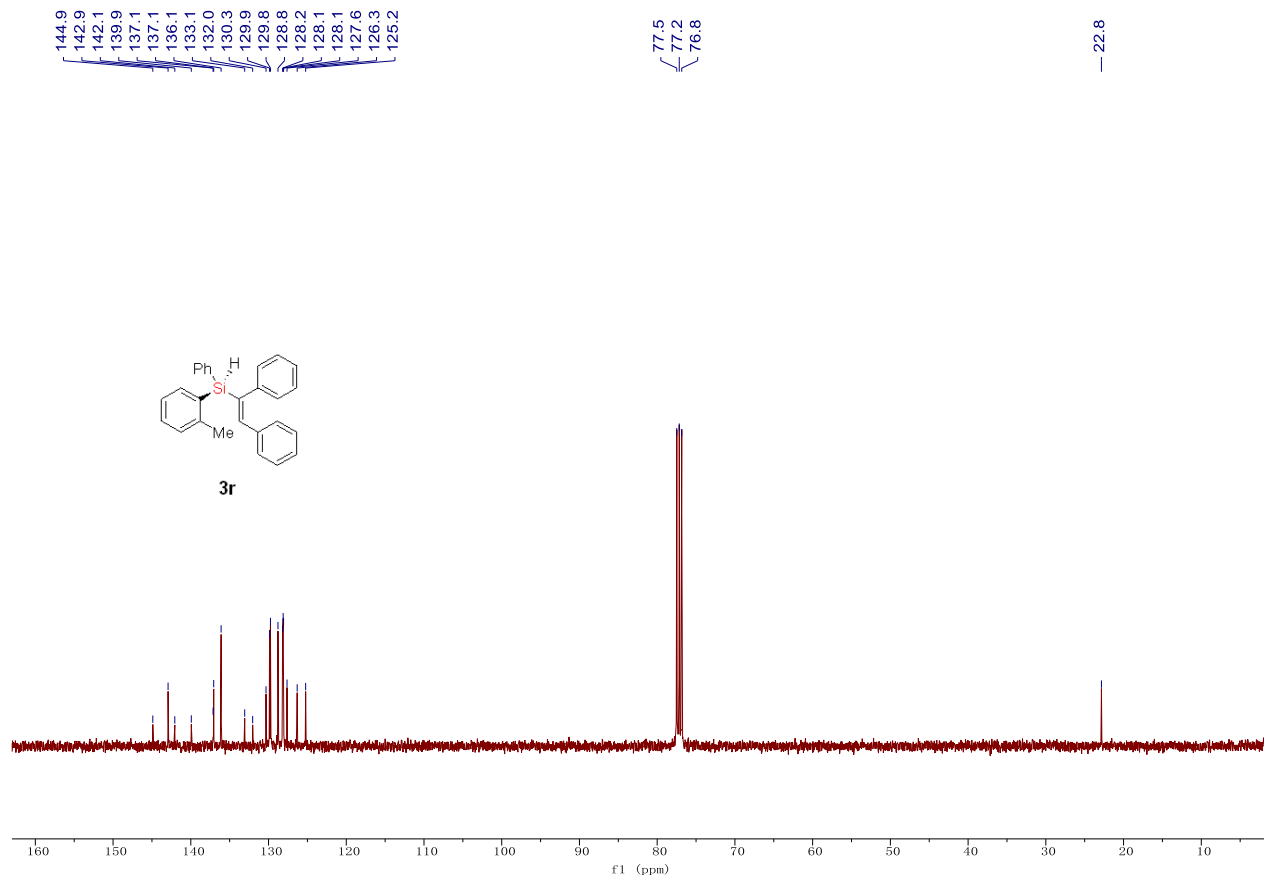
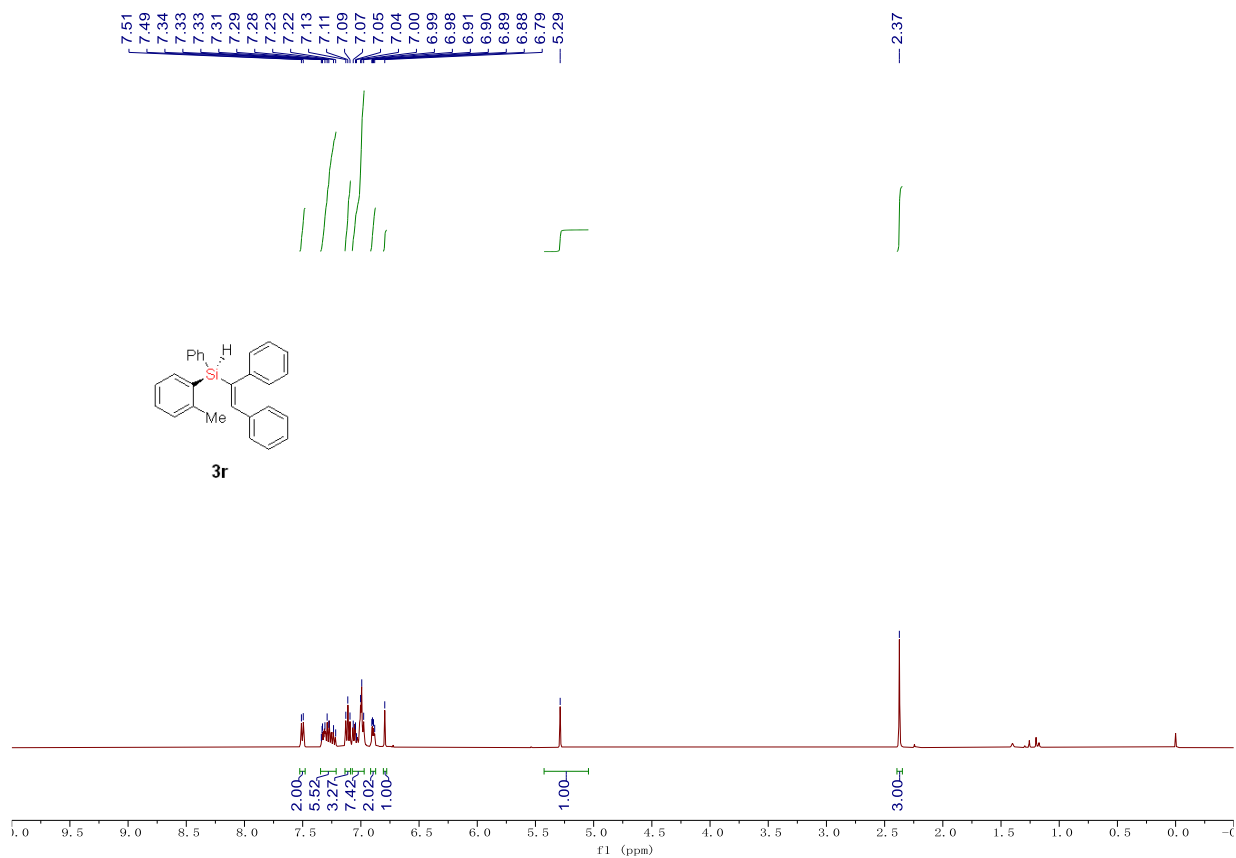
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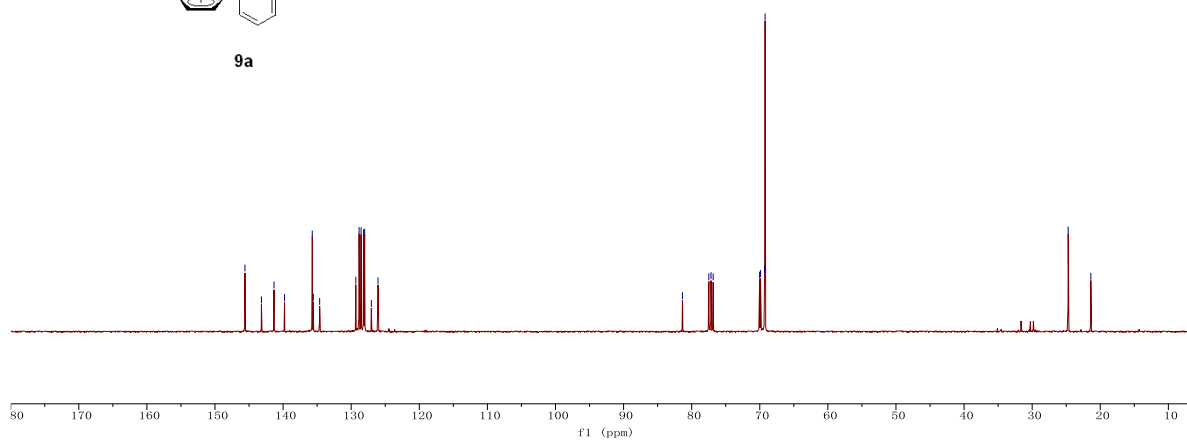
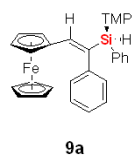
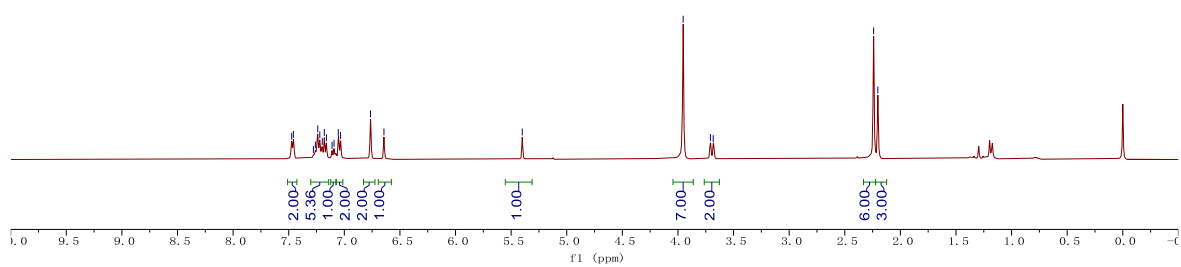
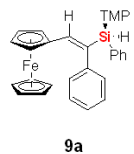
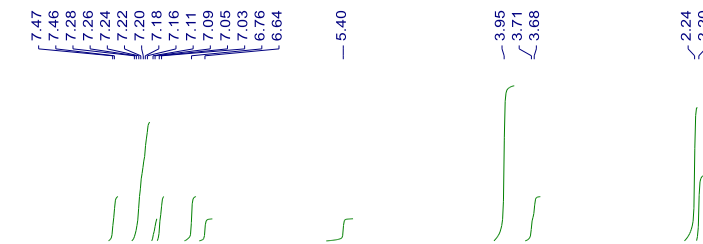


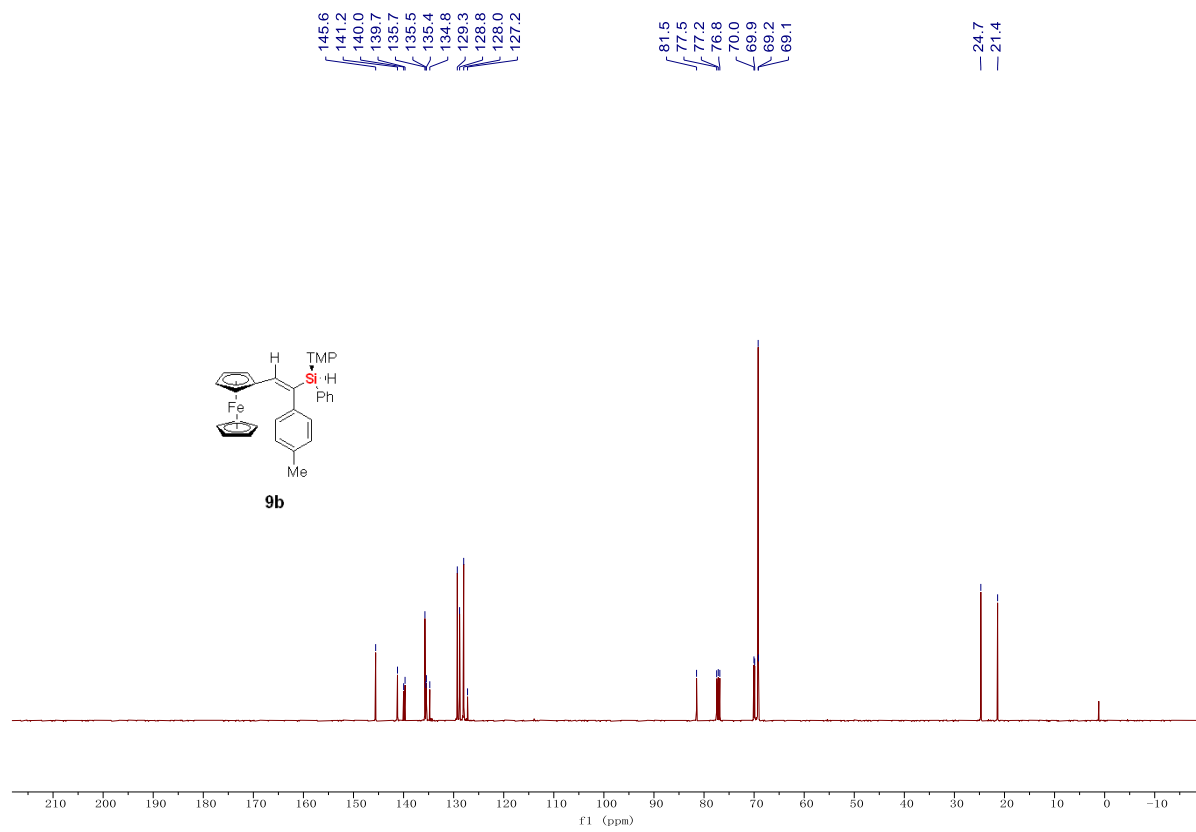
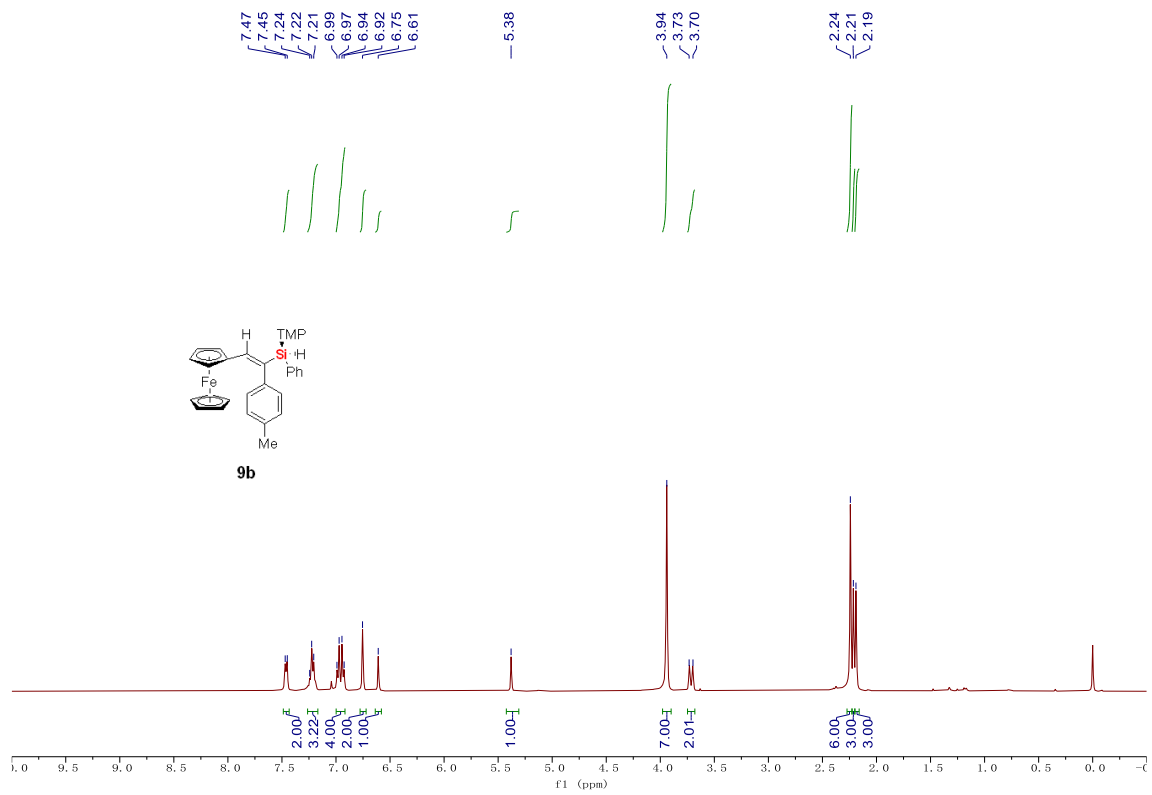
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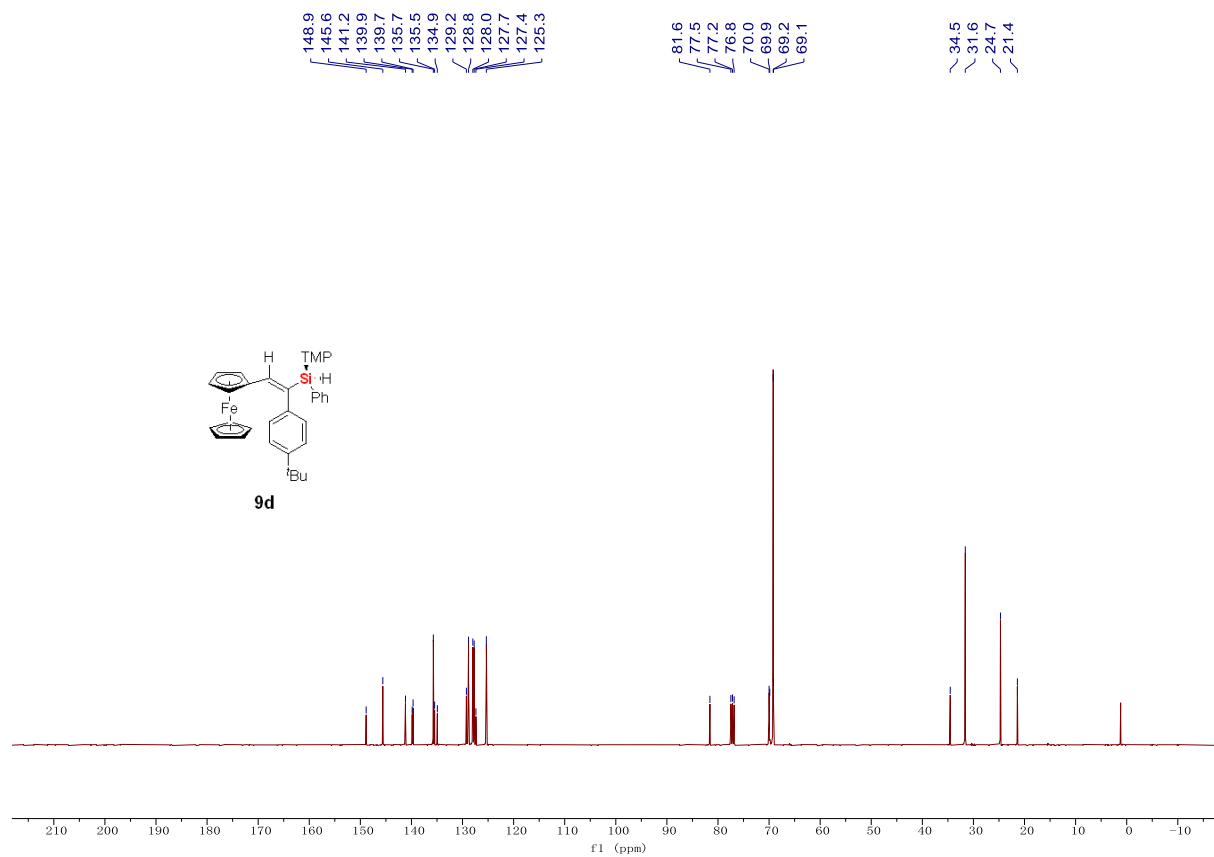
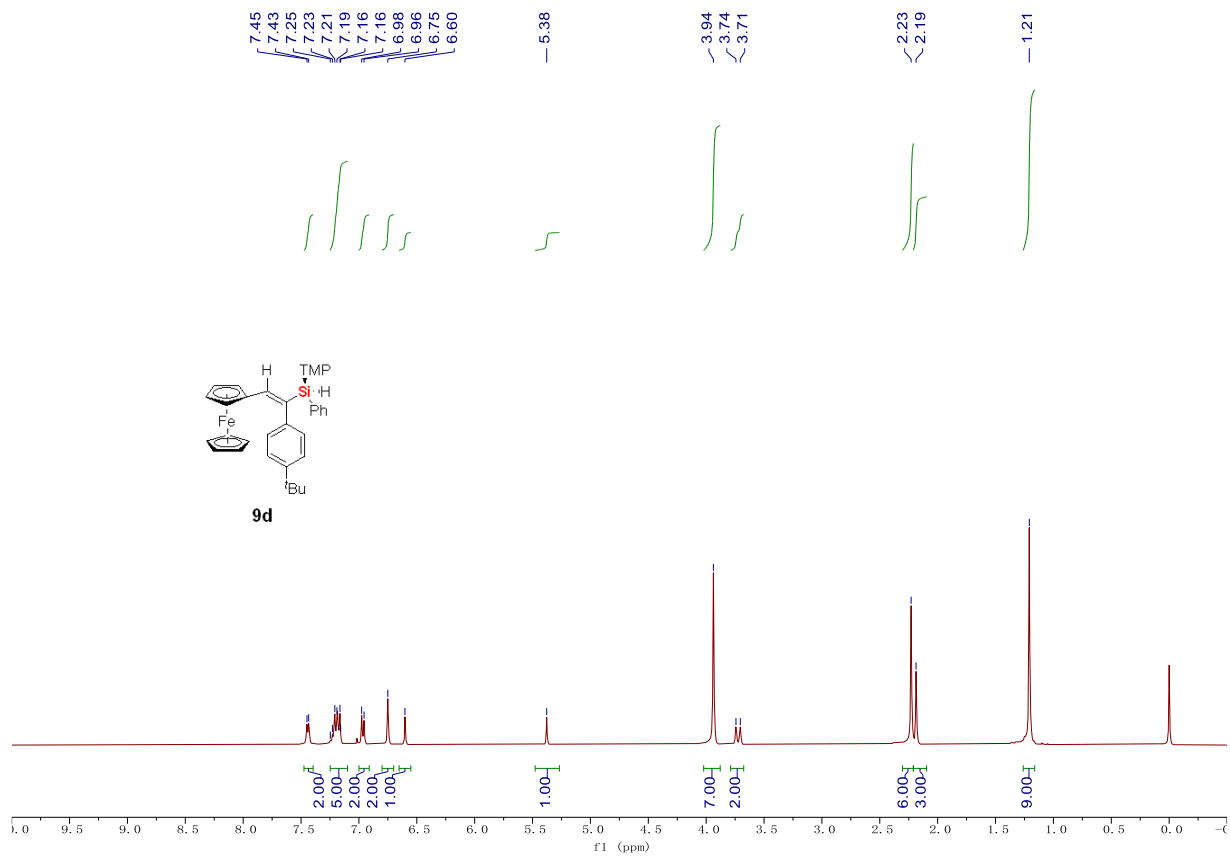


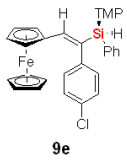
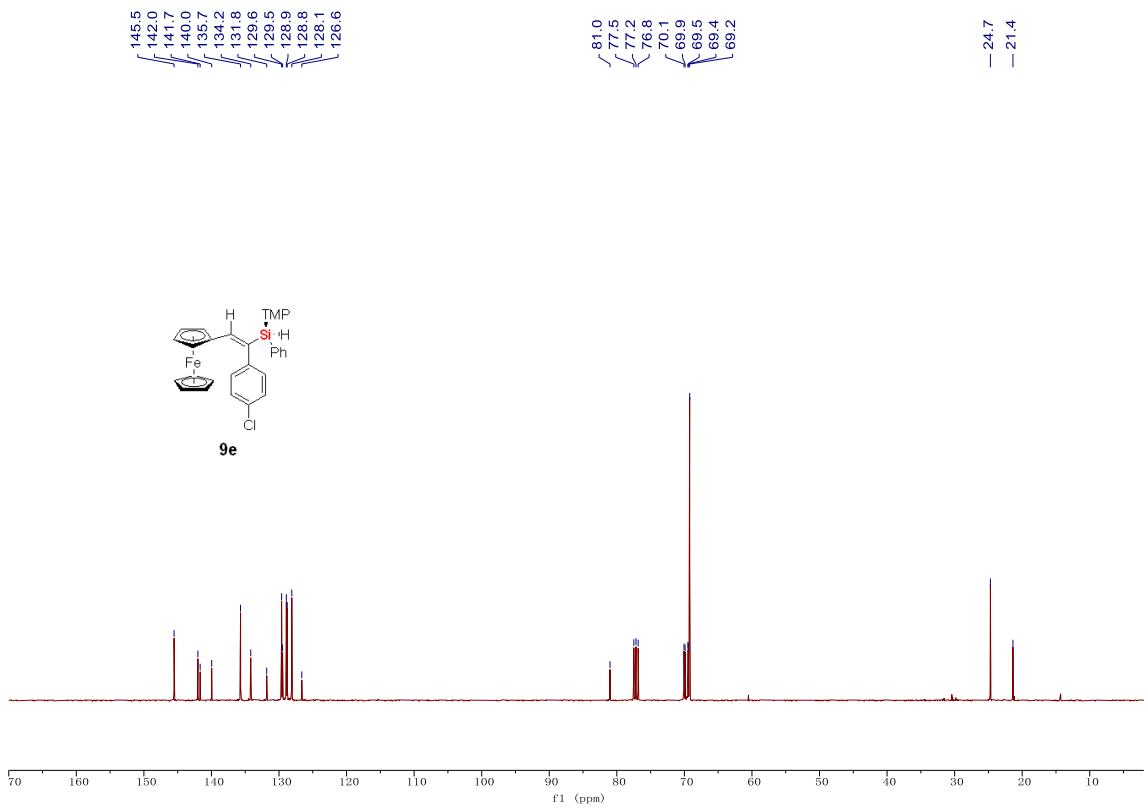
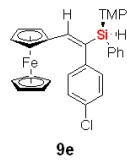
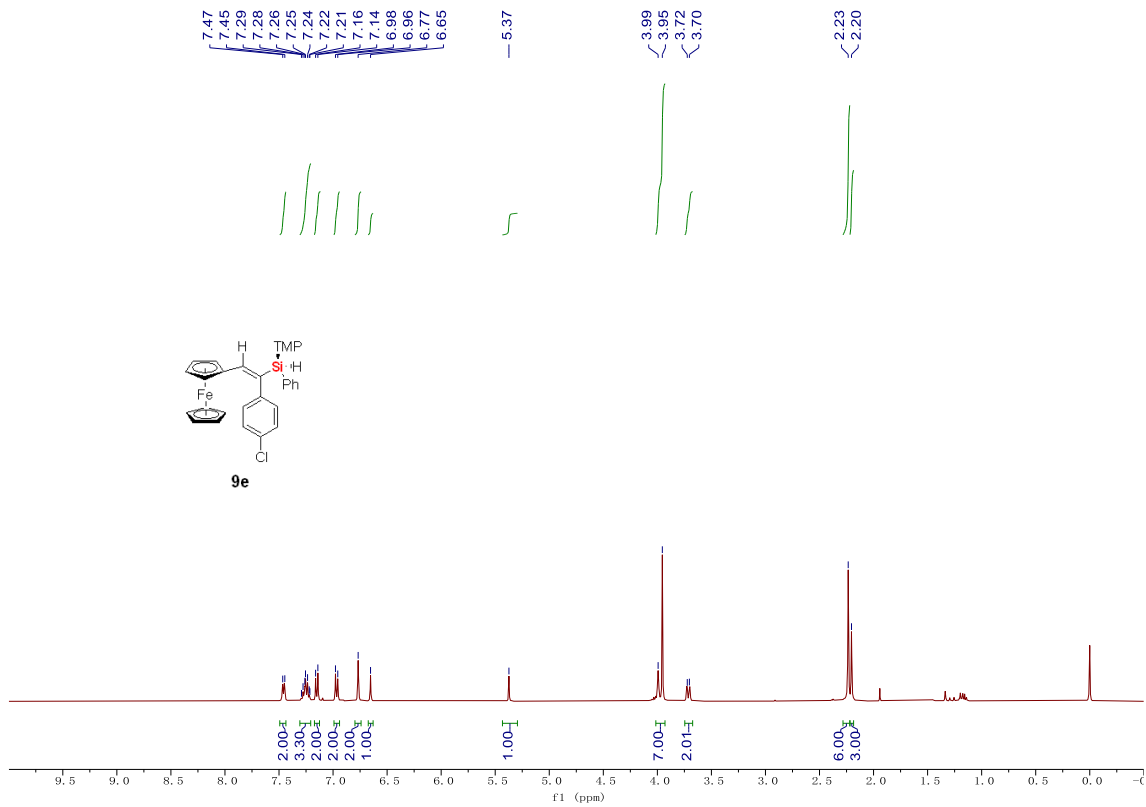


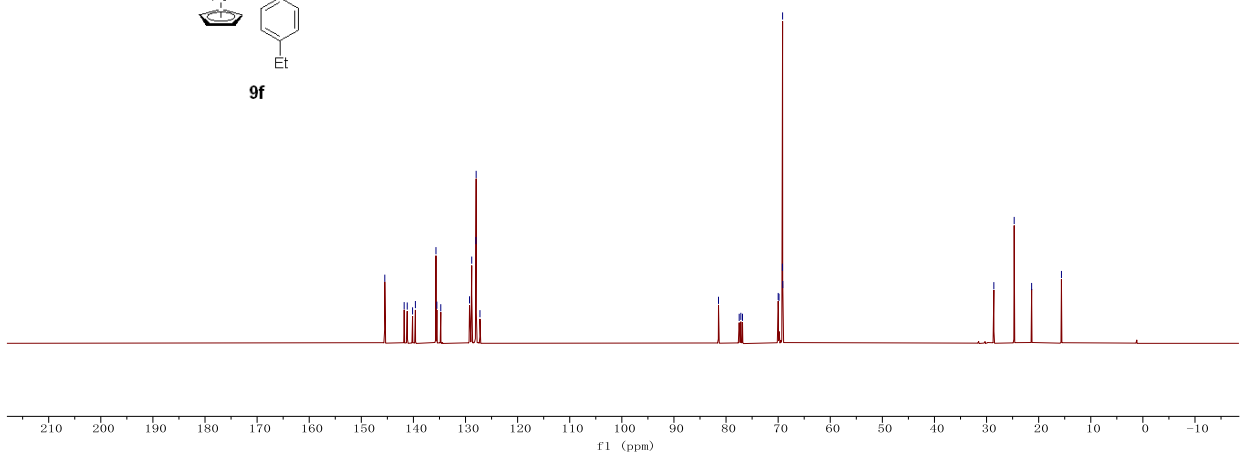
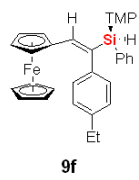
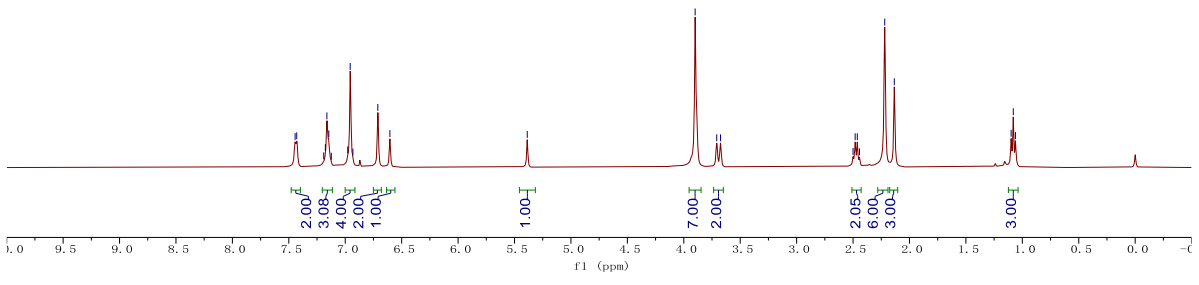
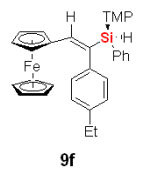
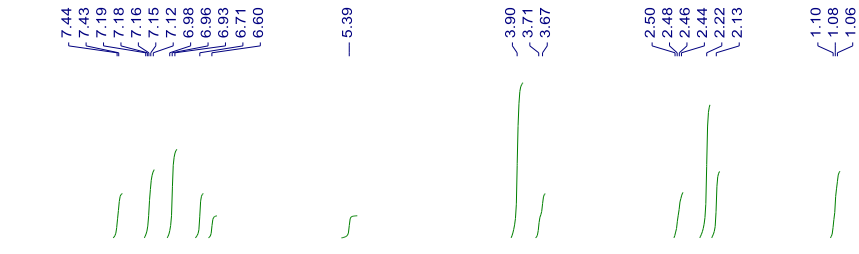


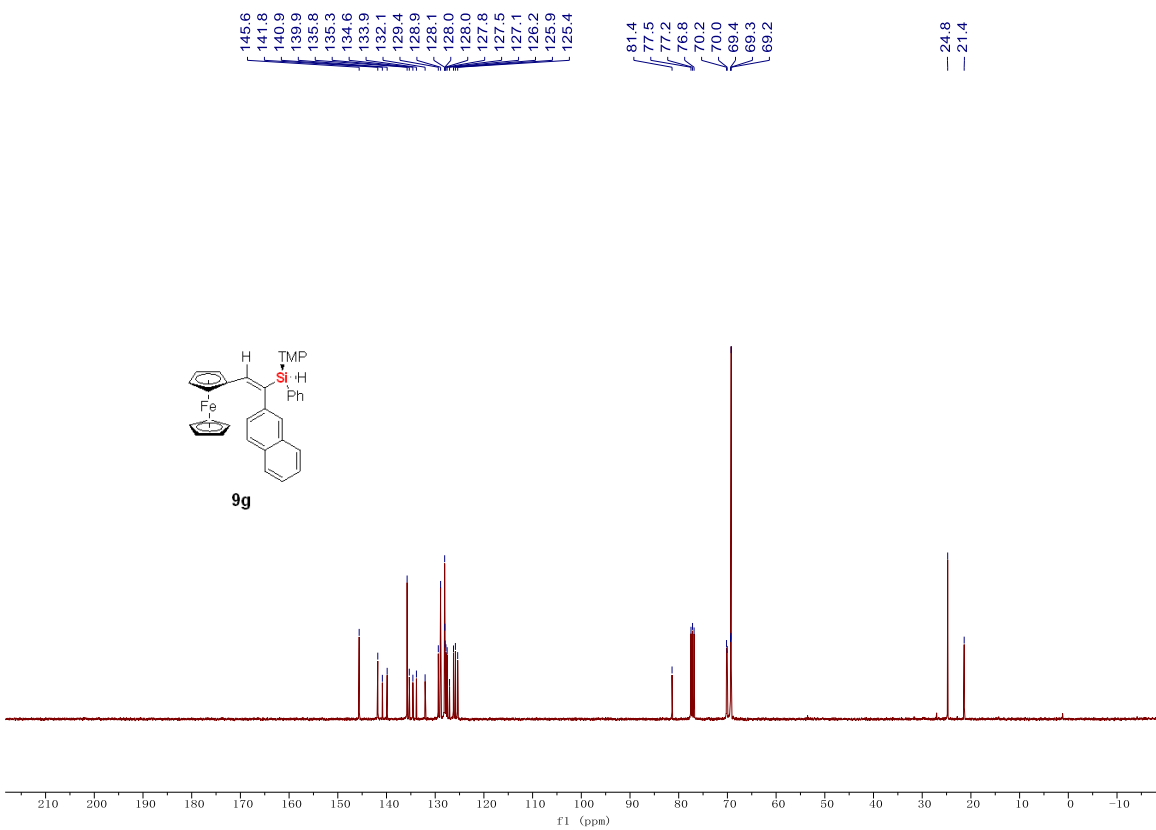
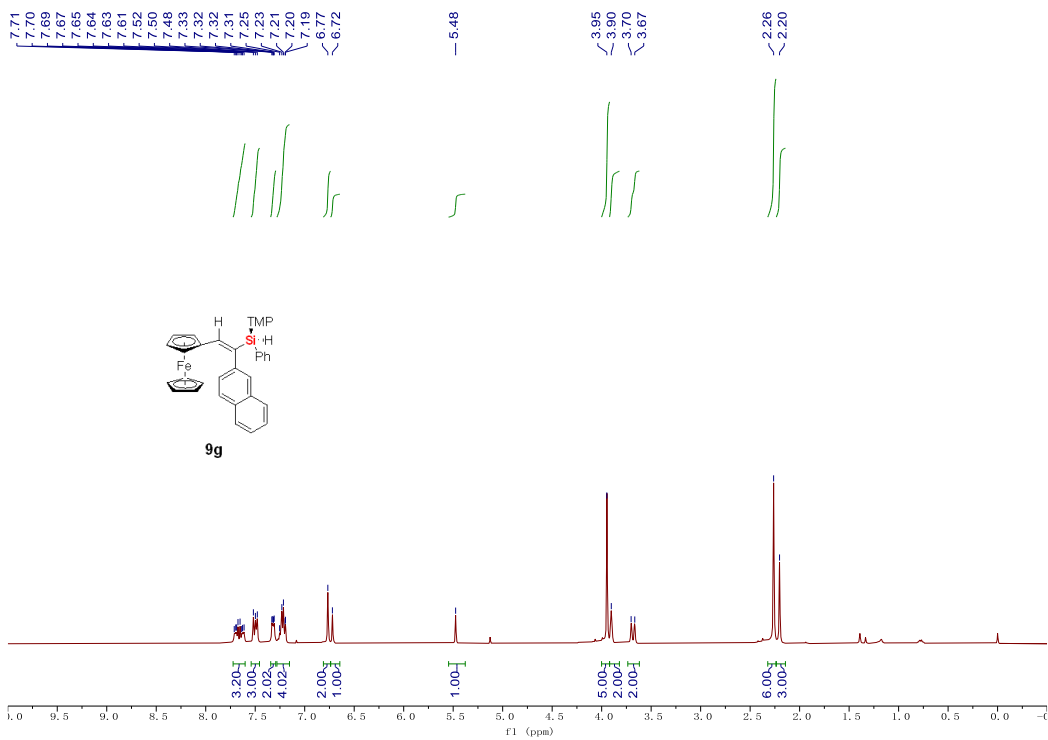


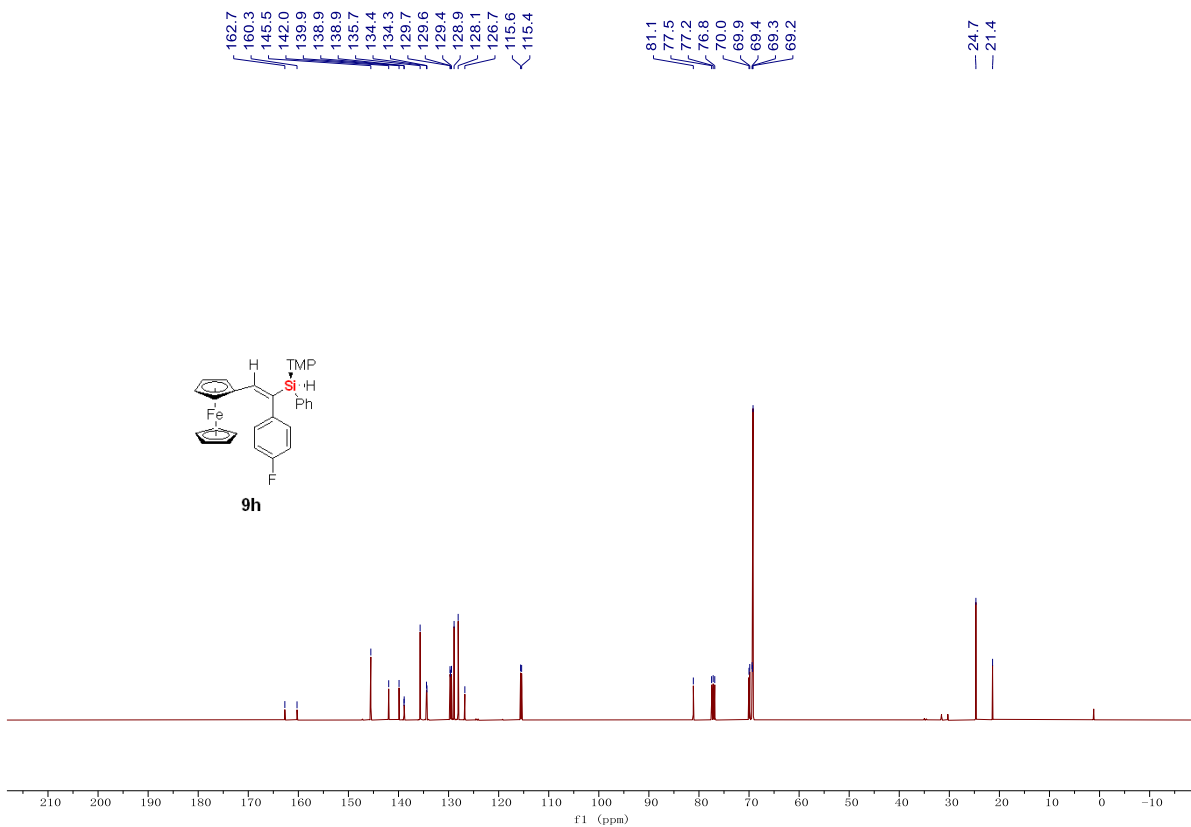
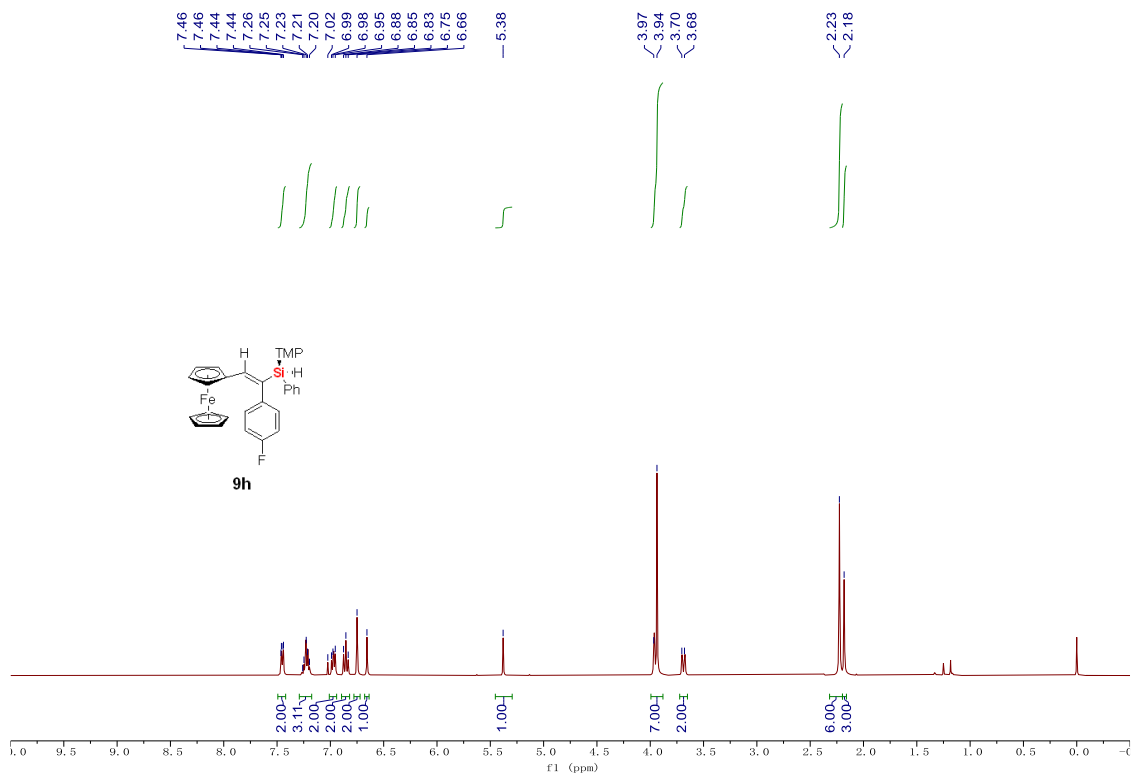




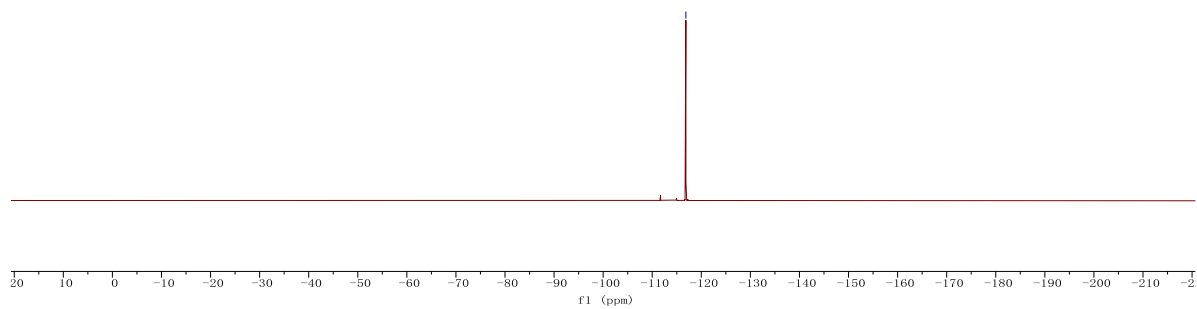
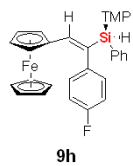


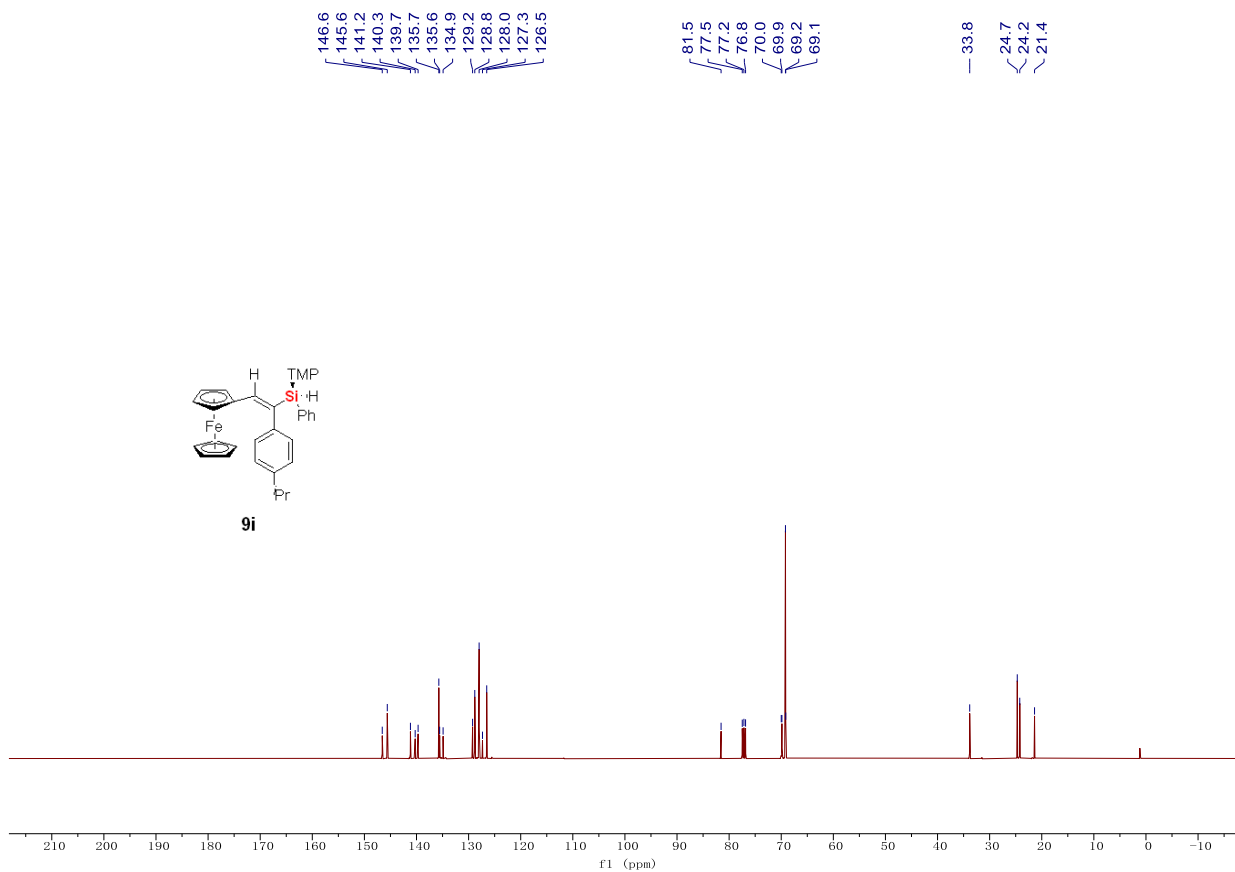
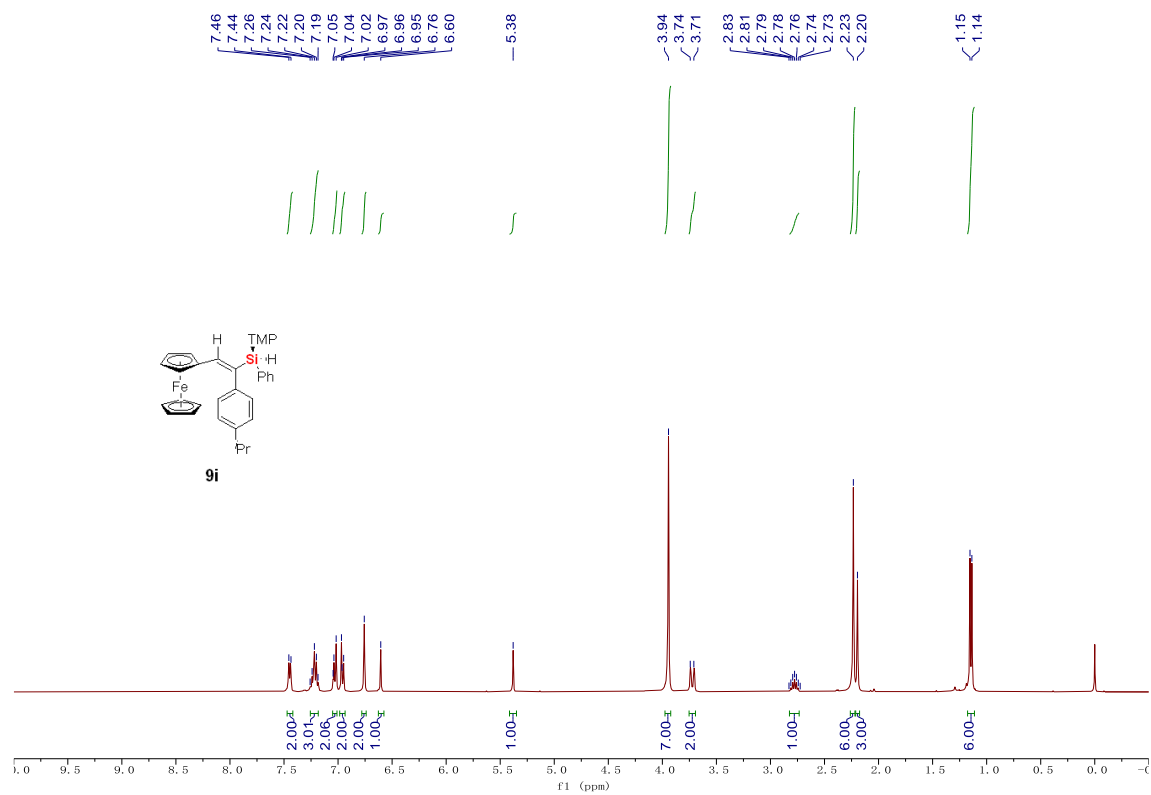


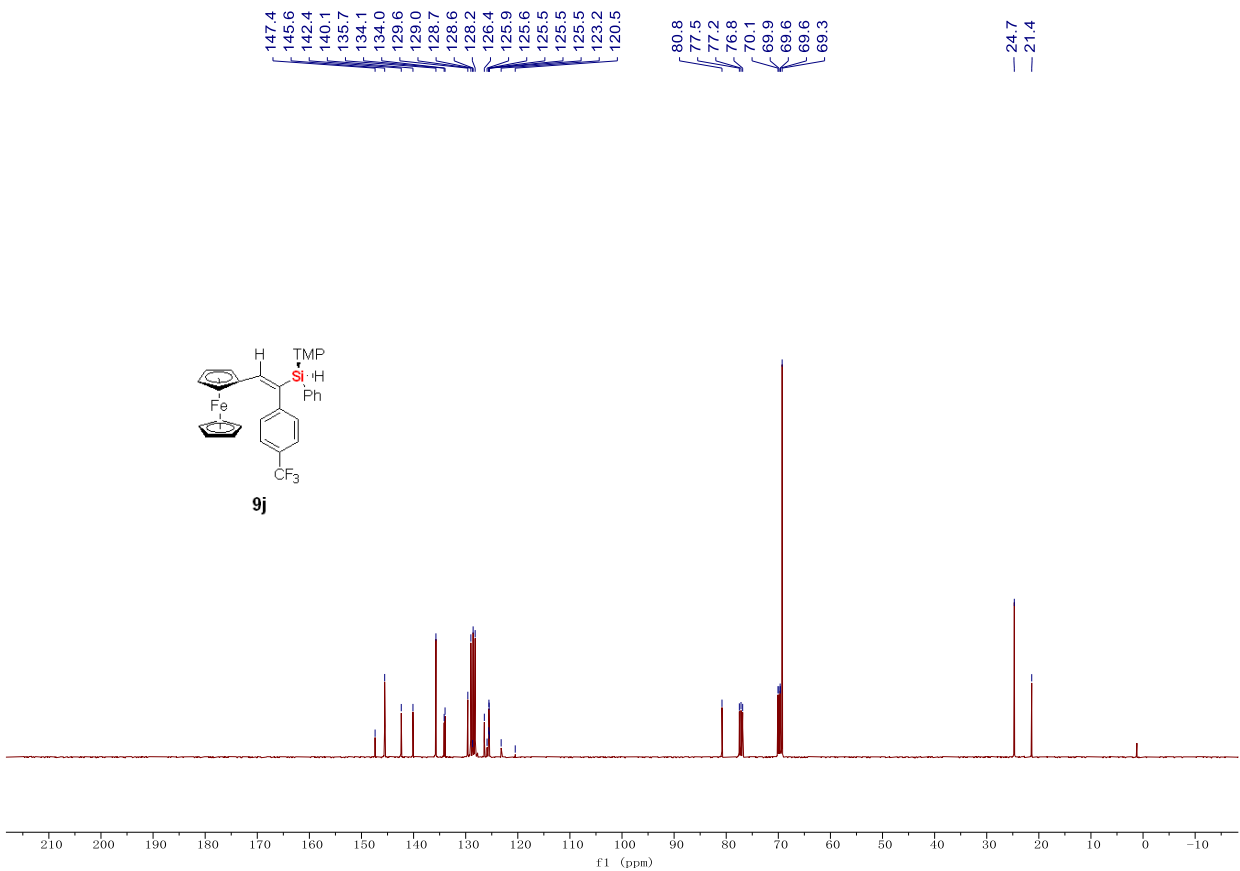
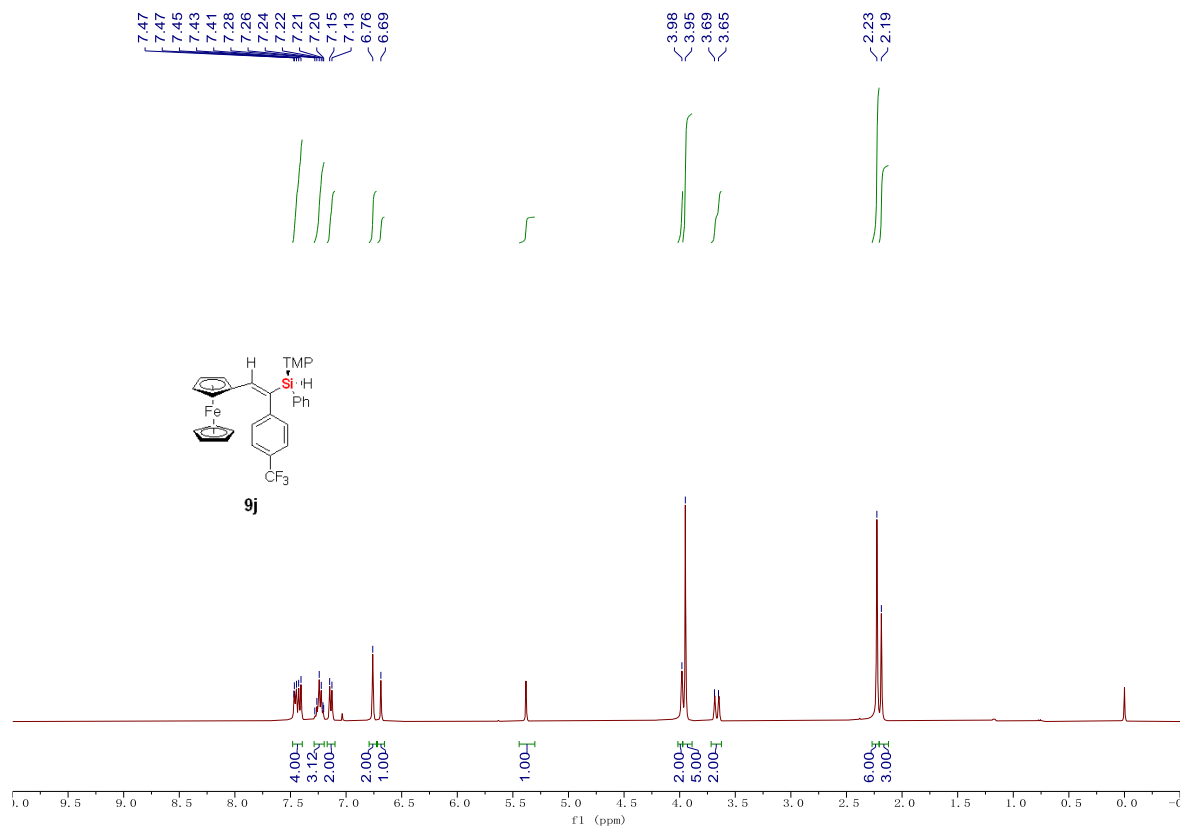




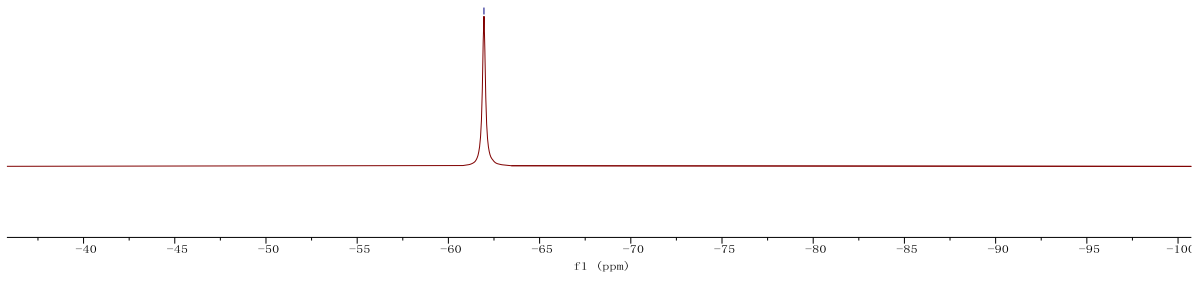
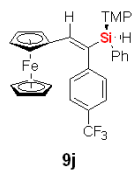
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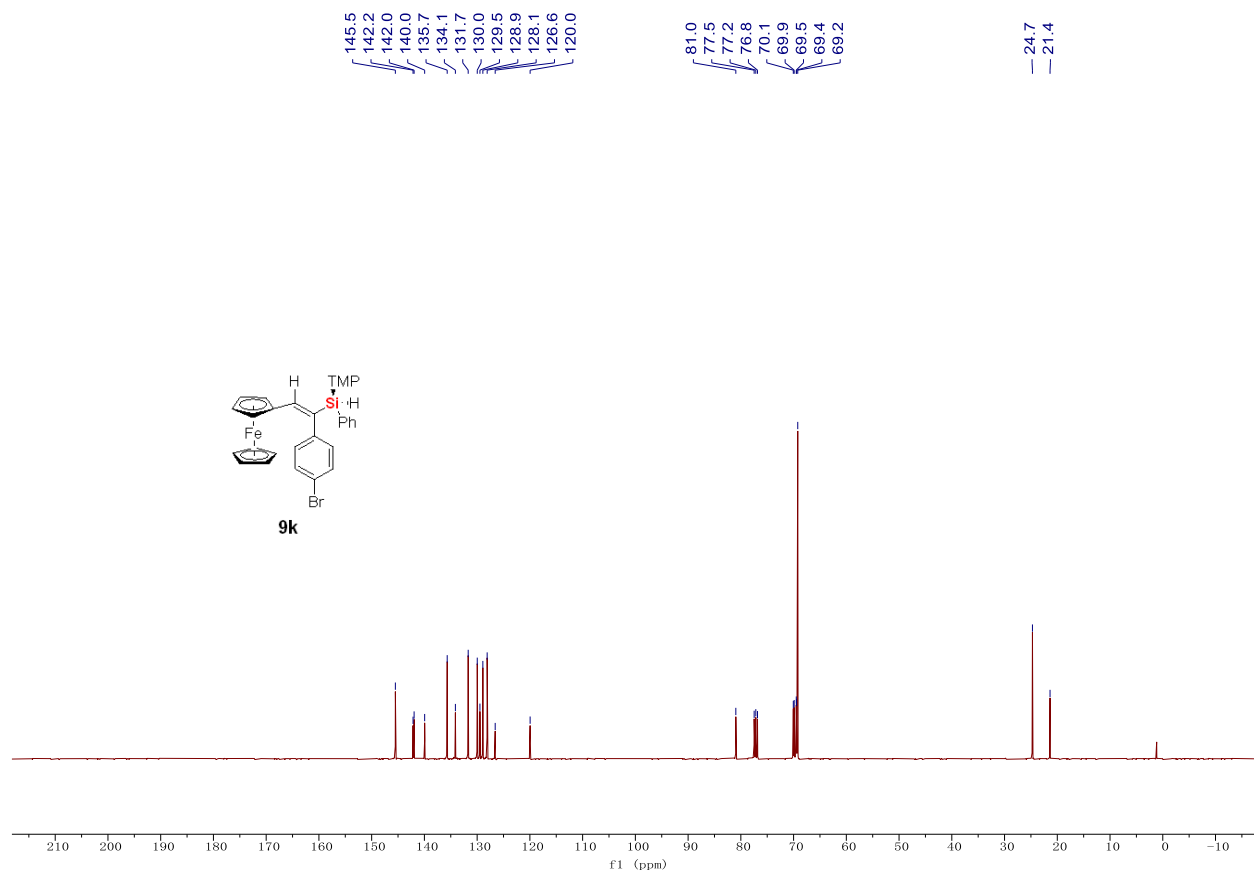
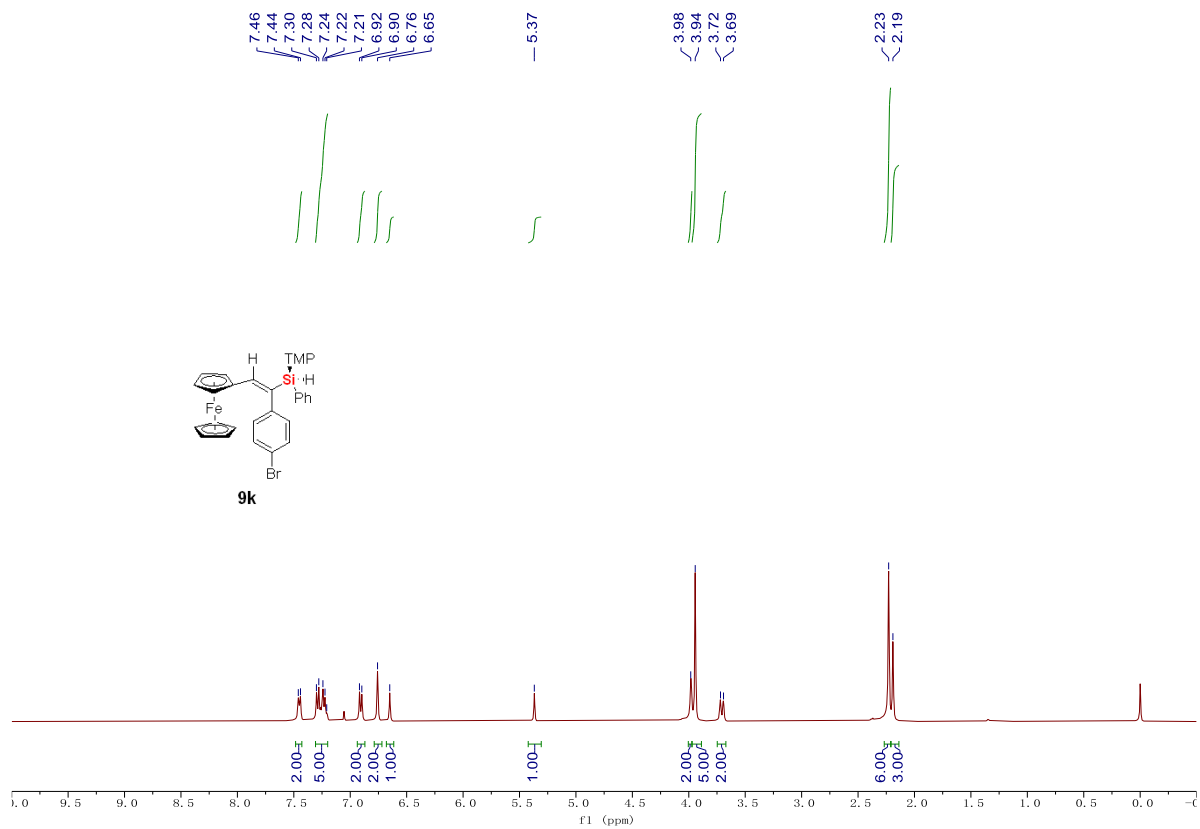


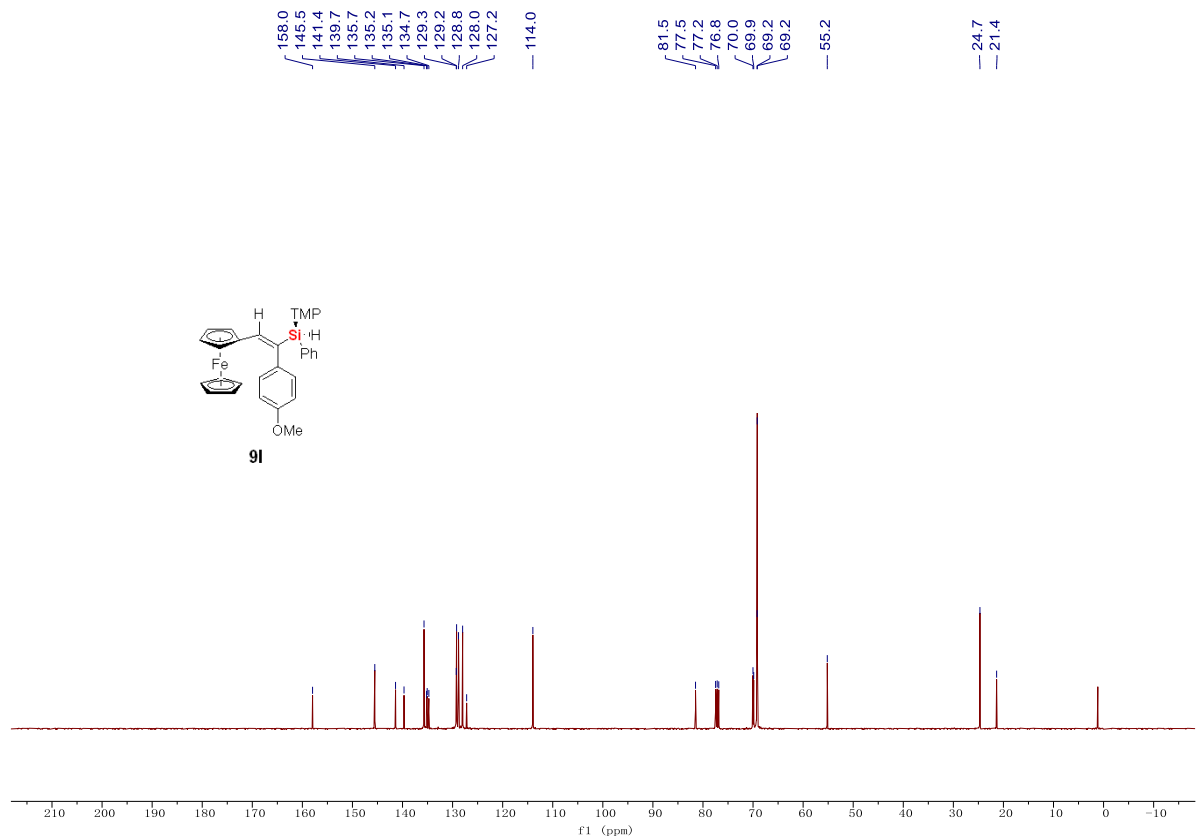
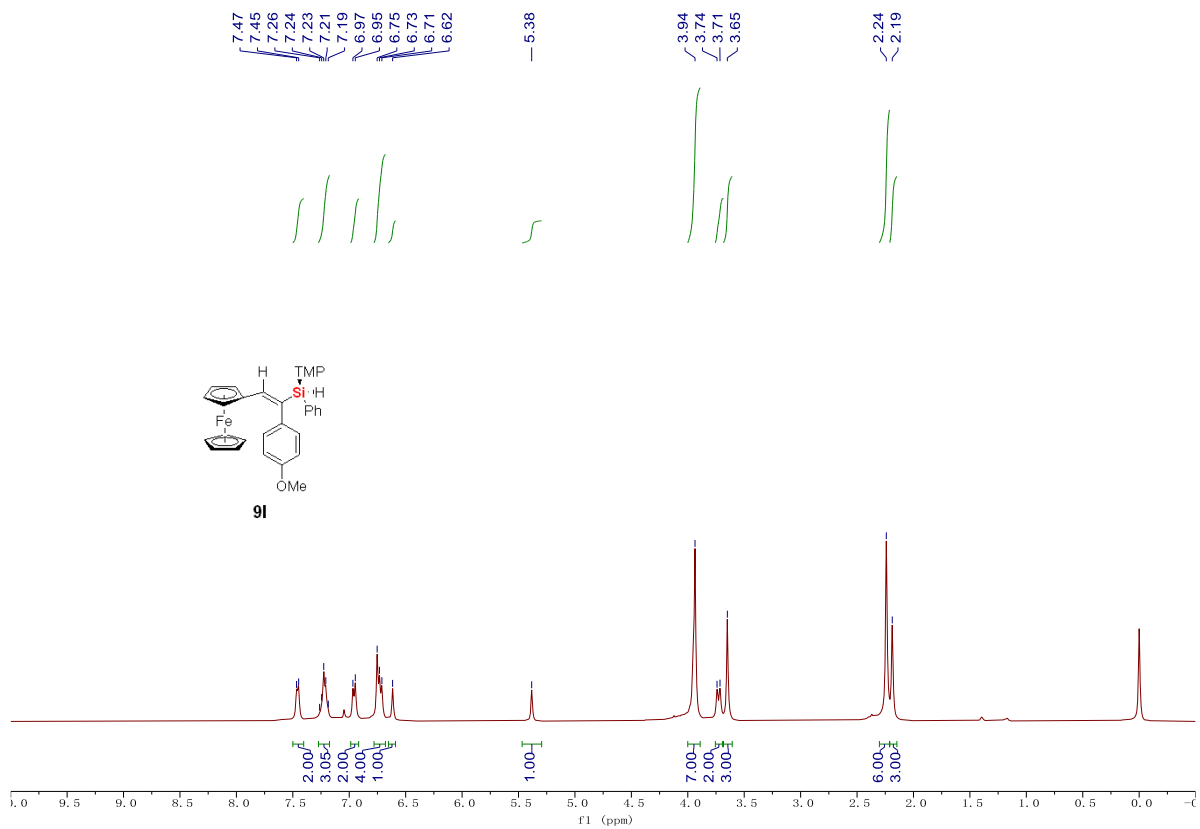


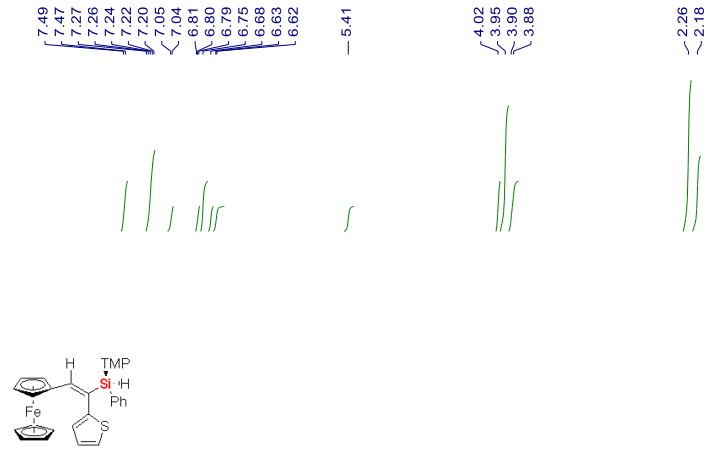


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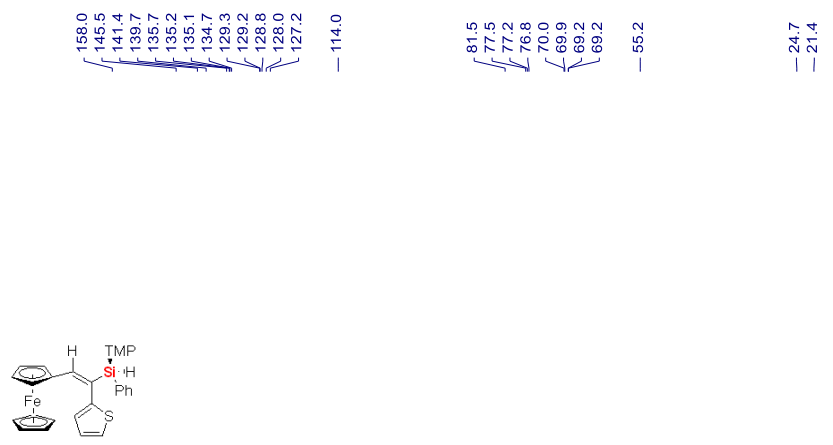
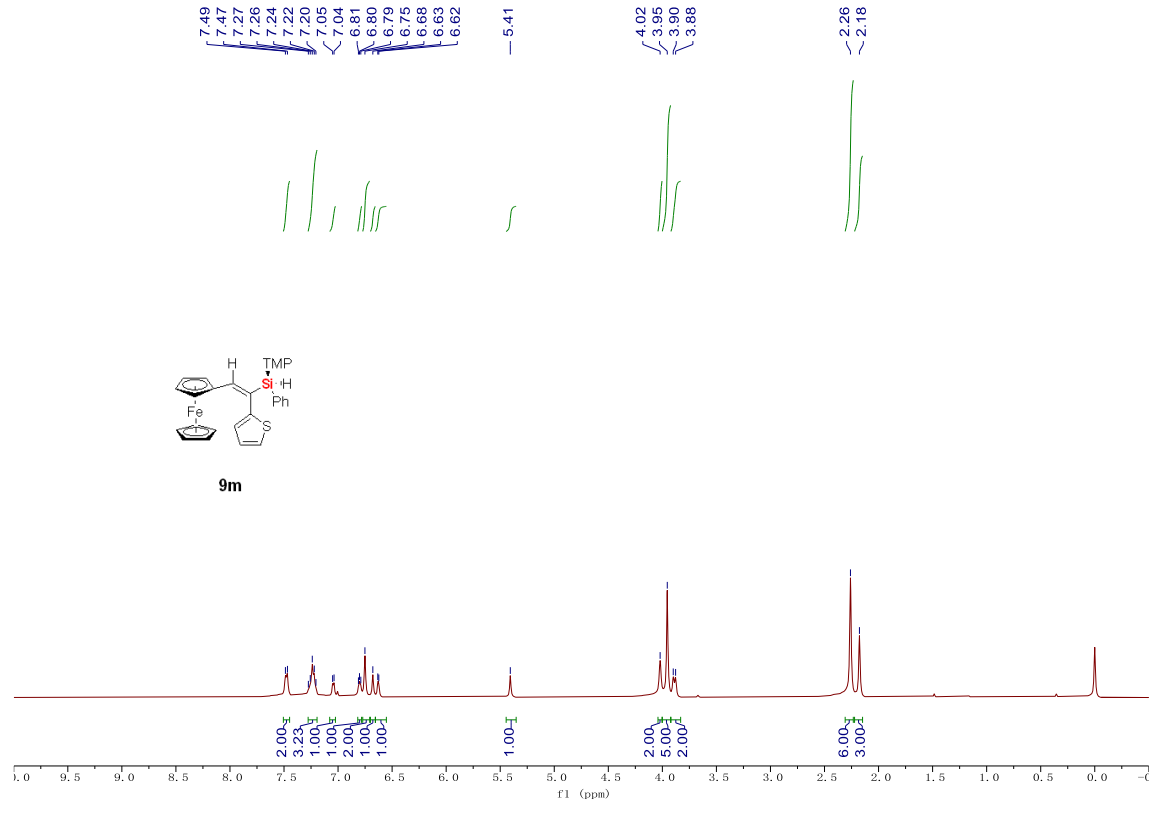




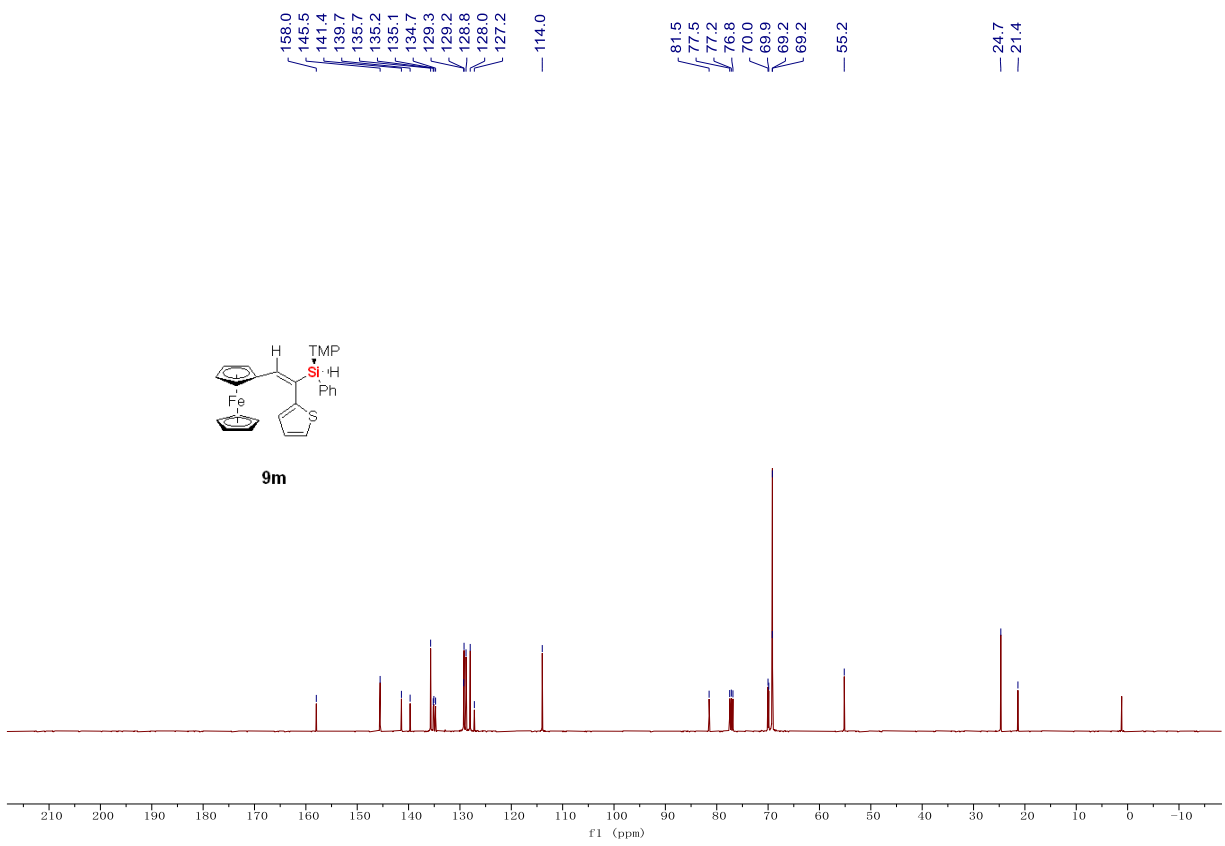


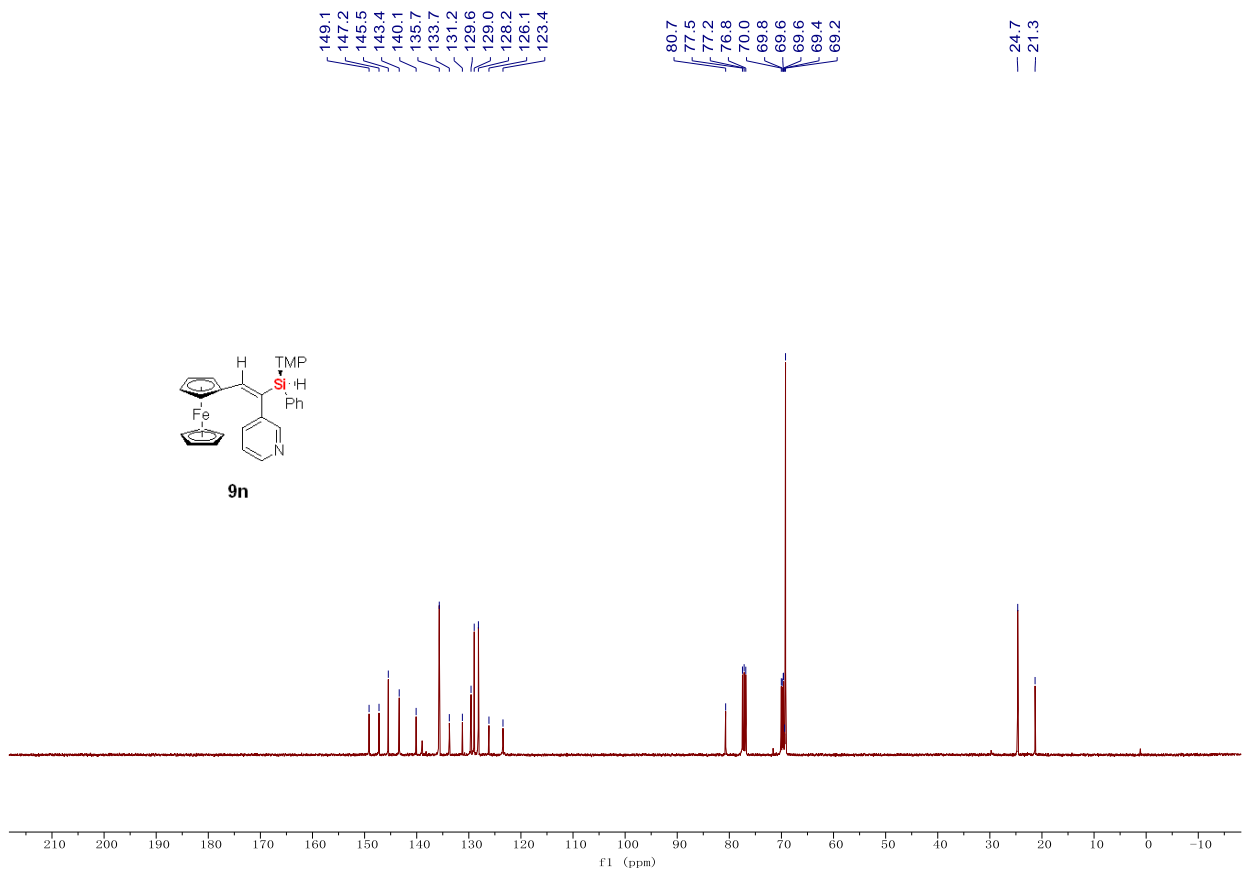
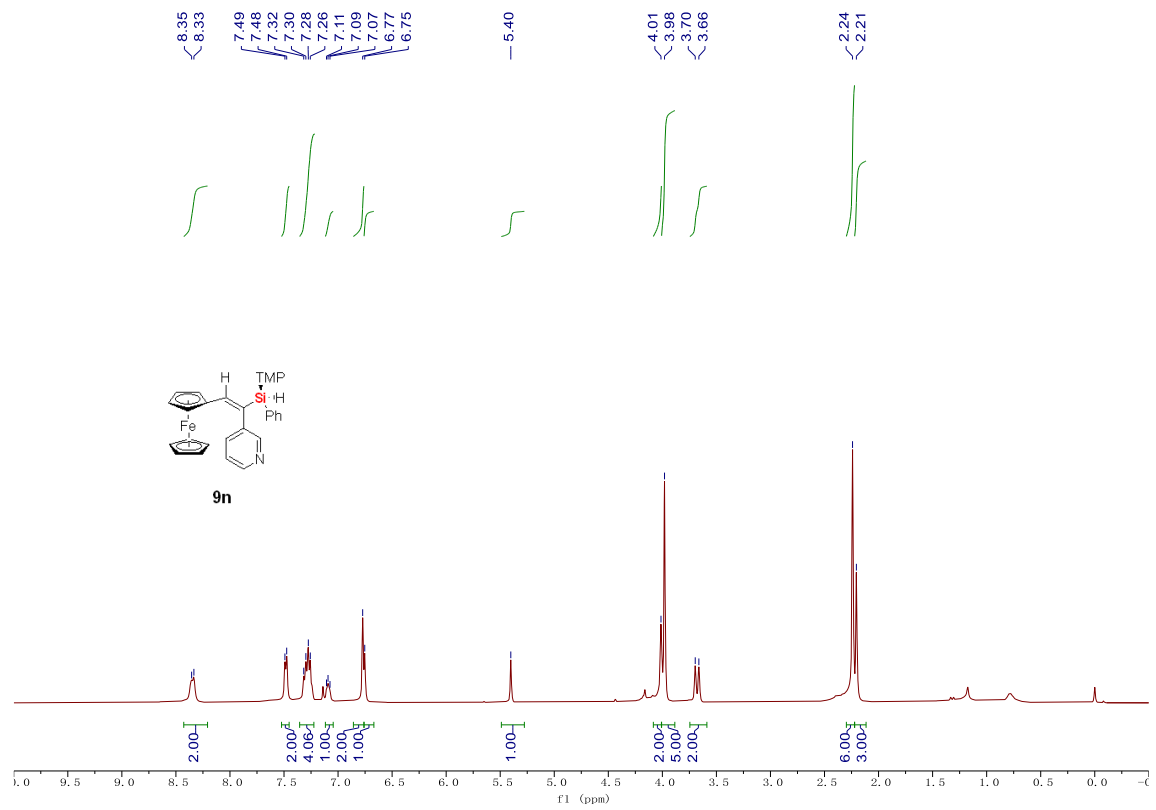


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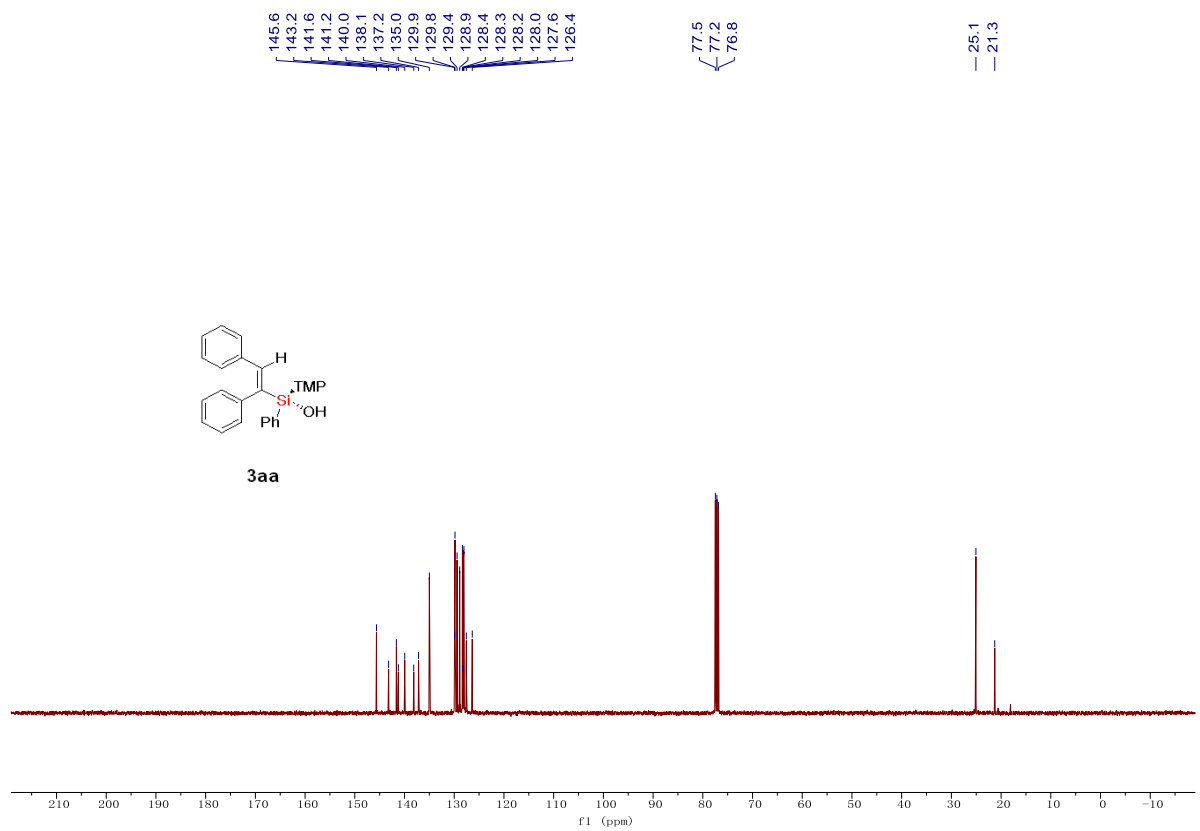
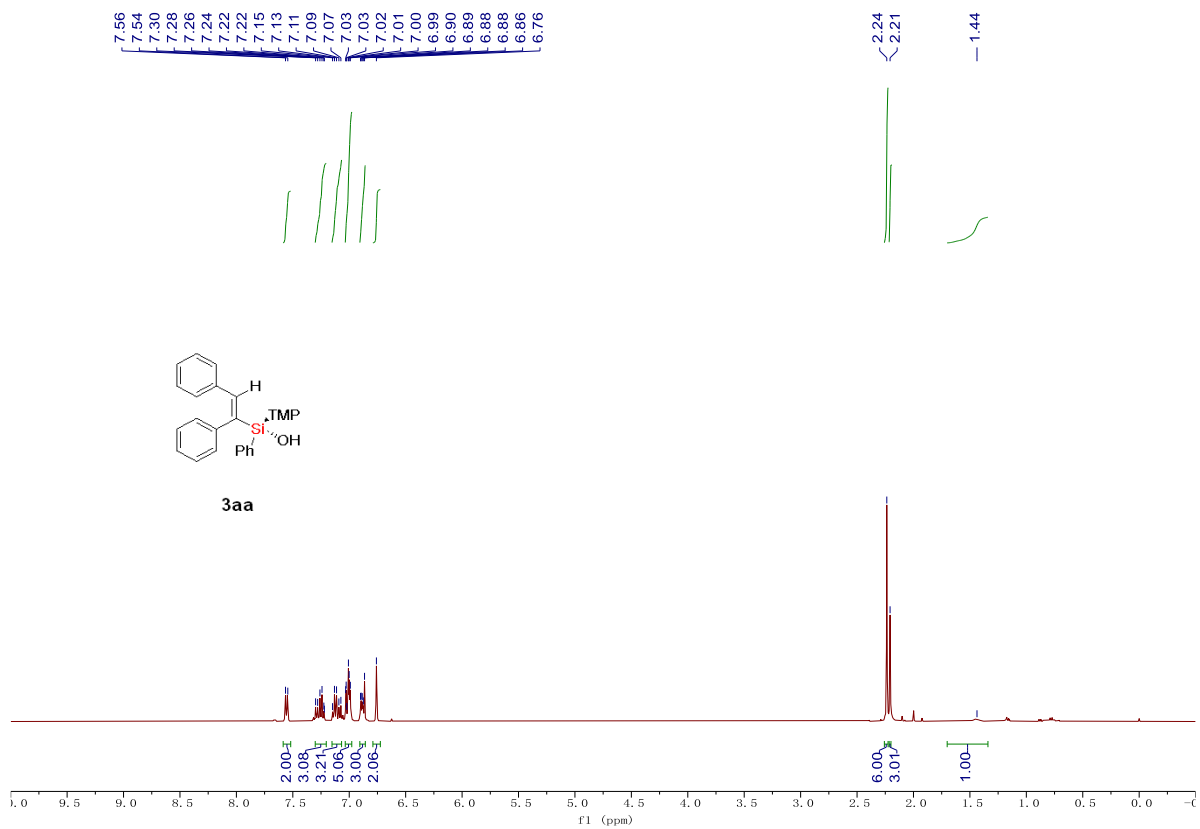


9m





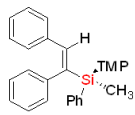
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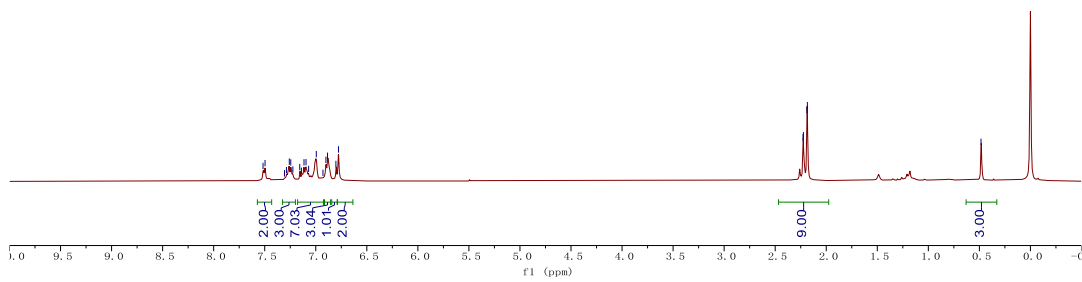
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3ab



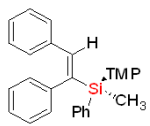
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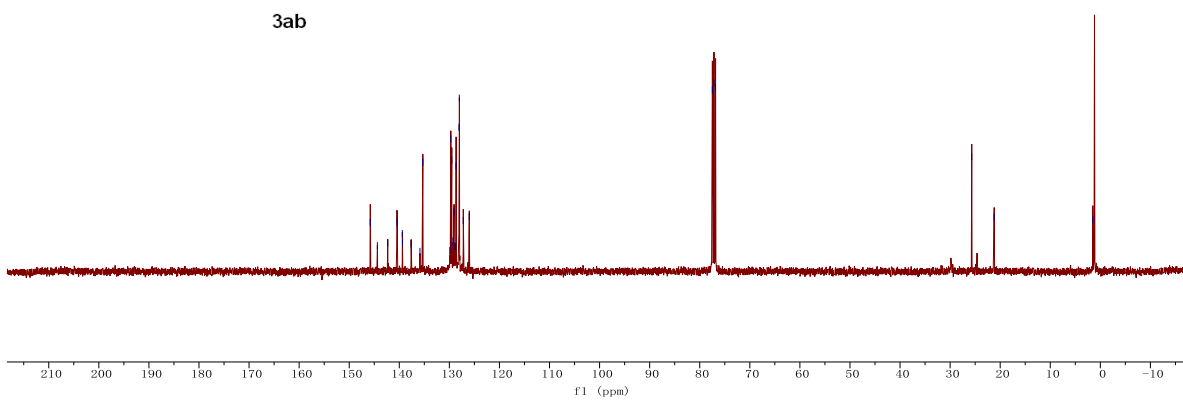
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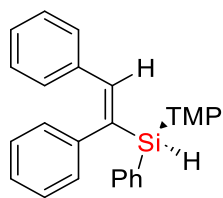
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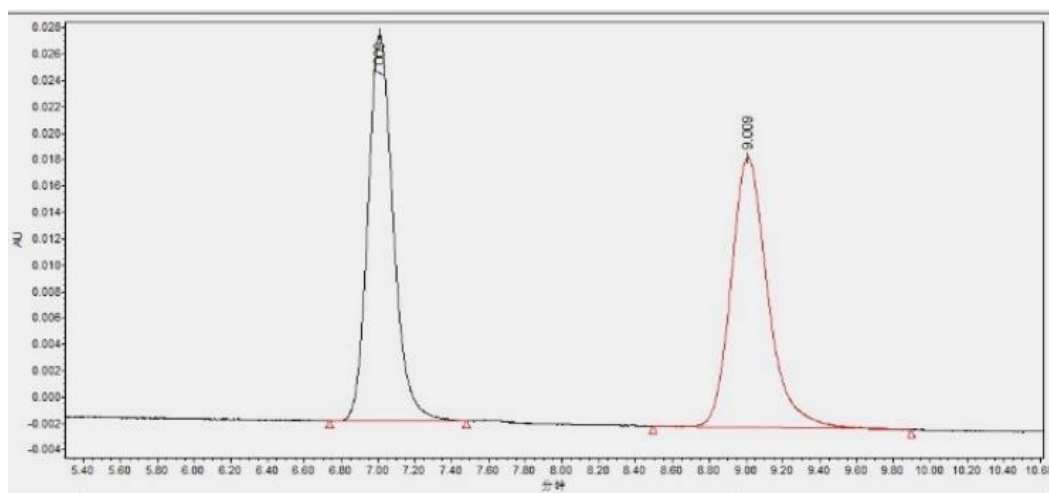
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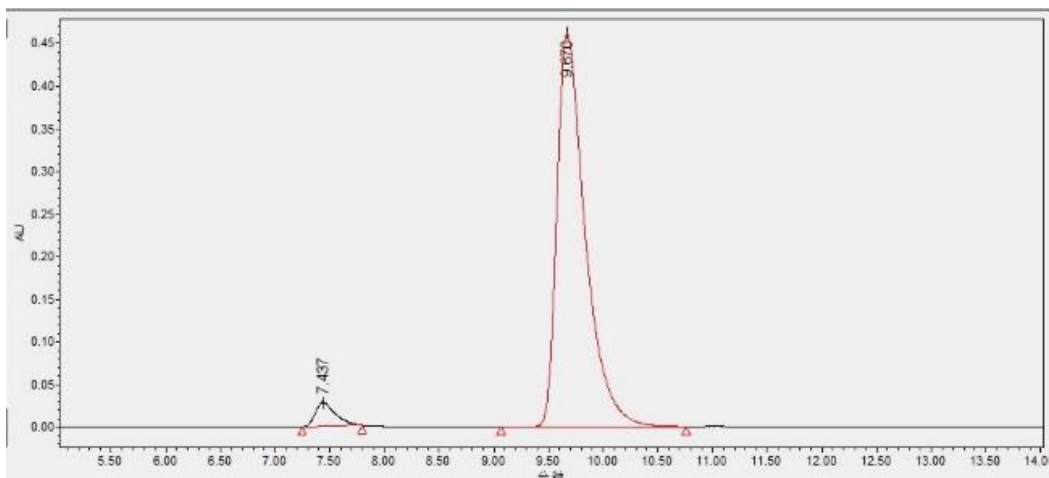
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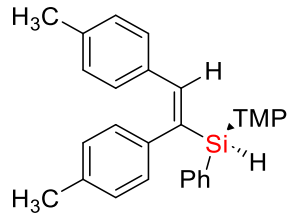
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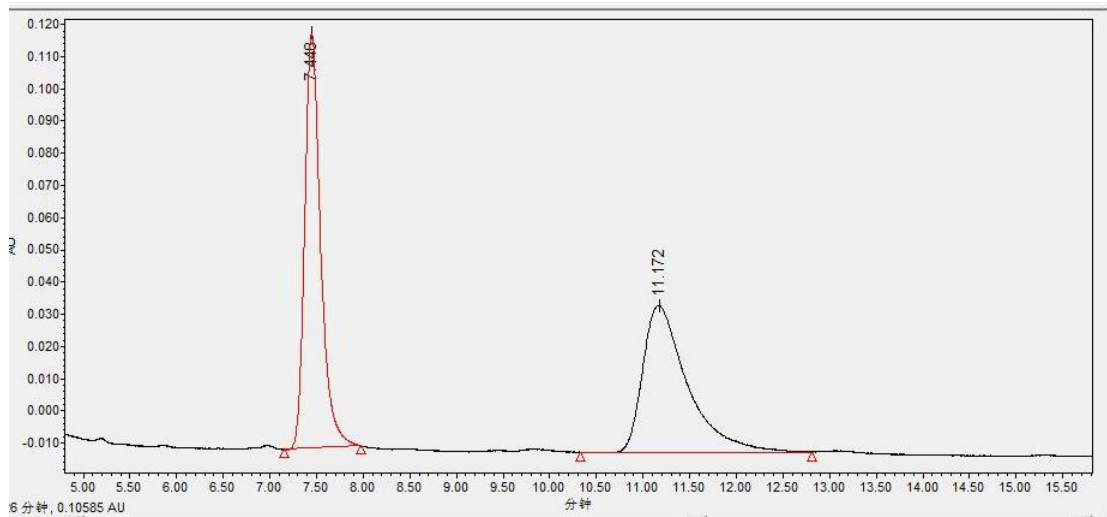
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2	9.009	284956	20478	50.38



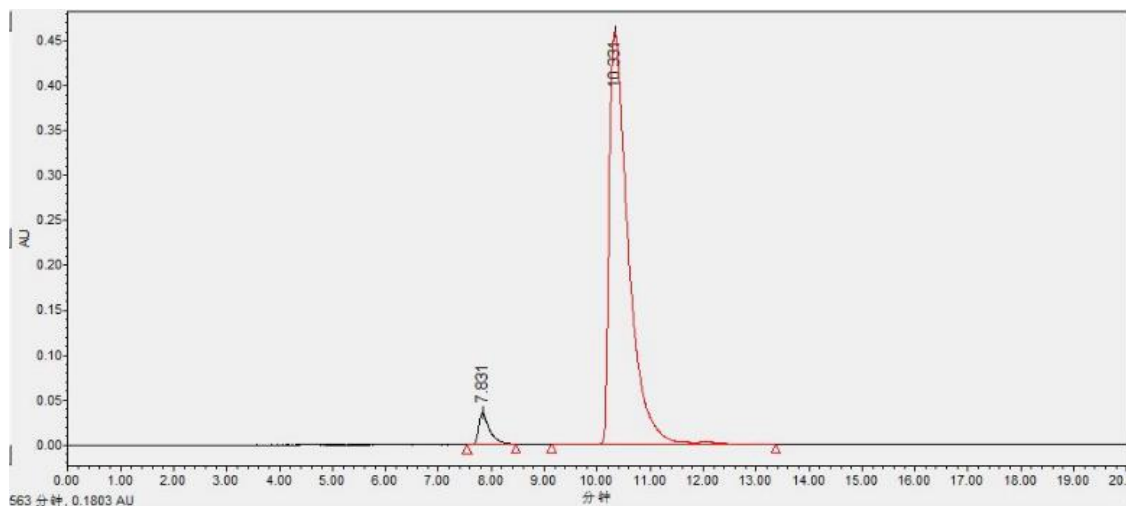
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2	9.670	8303133	461522	95.87



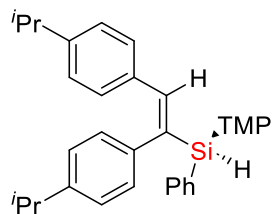
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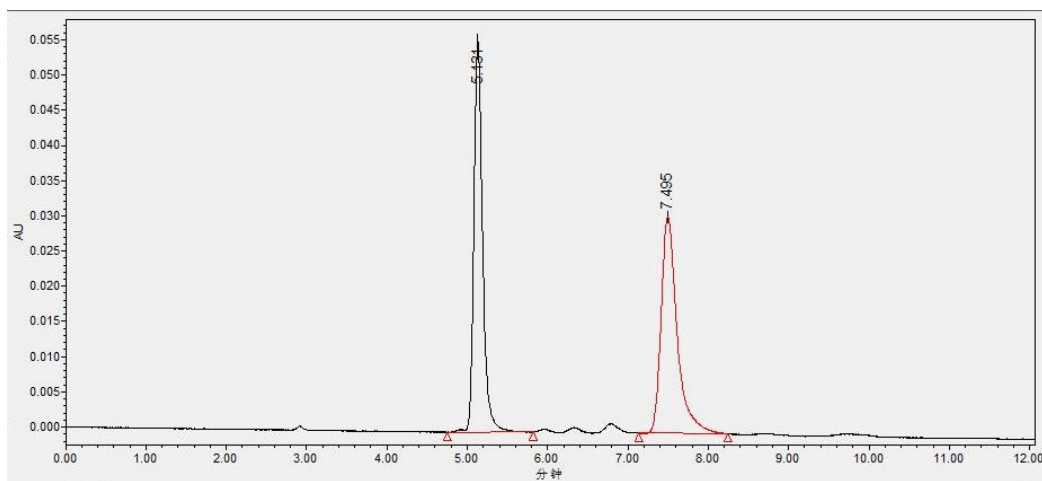
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2	11.172	1510493	45621	49.70



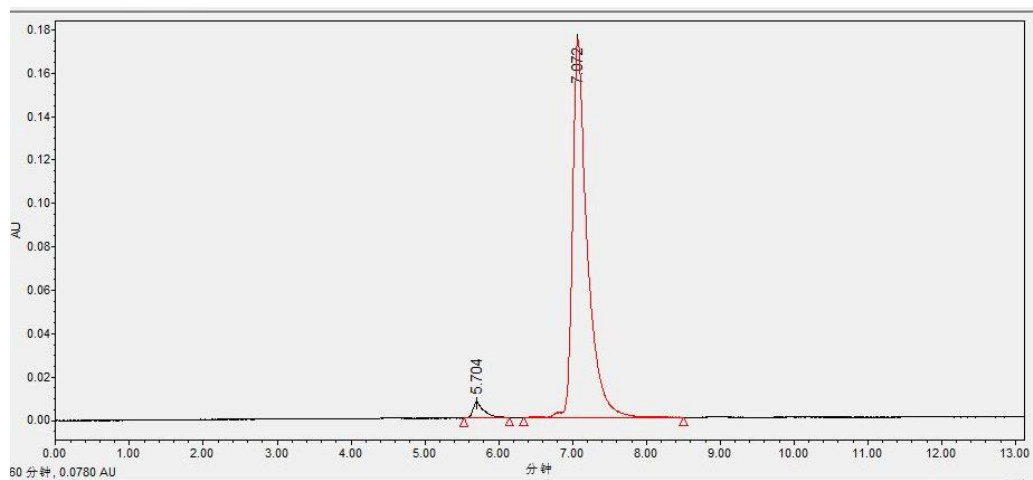
	Time/min	Area	Height	Area%
1	7.831	468851	35327	3.98
2	10.331	11316766	458559	96.02



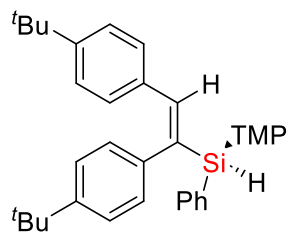
3c



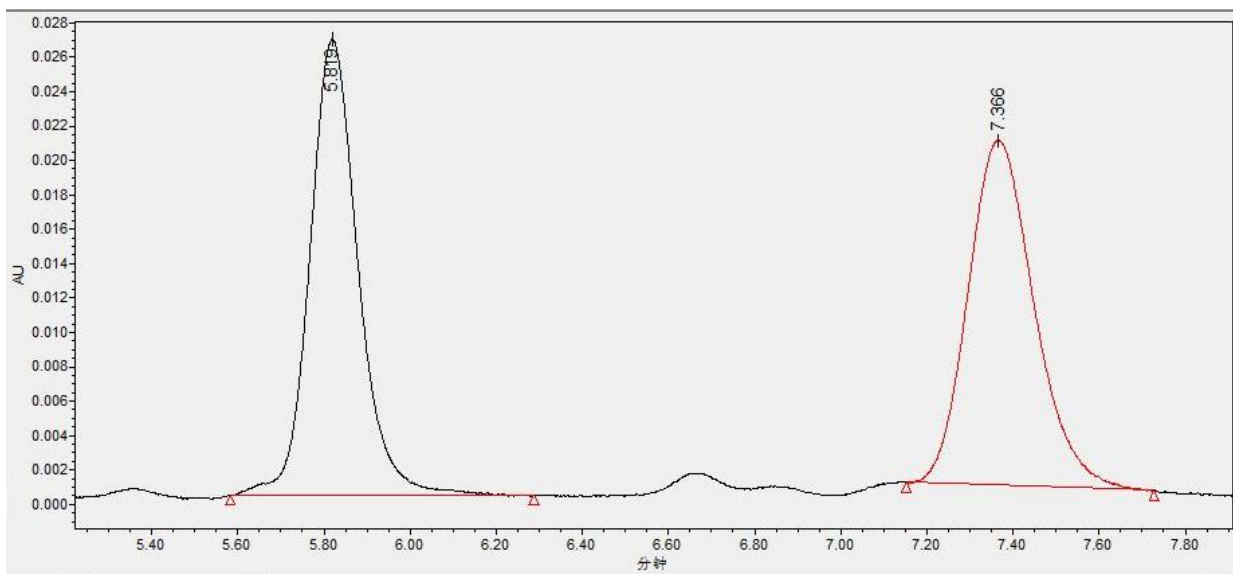
	Time/min	Area	Height	Area%
1	5.131	420822	55709	49.72
2	7.495	425646	30756	50.28



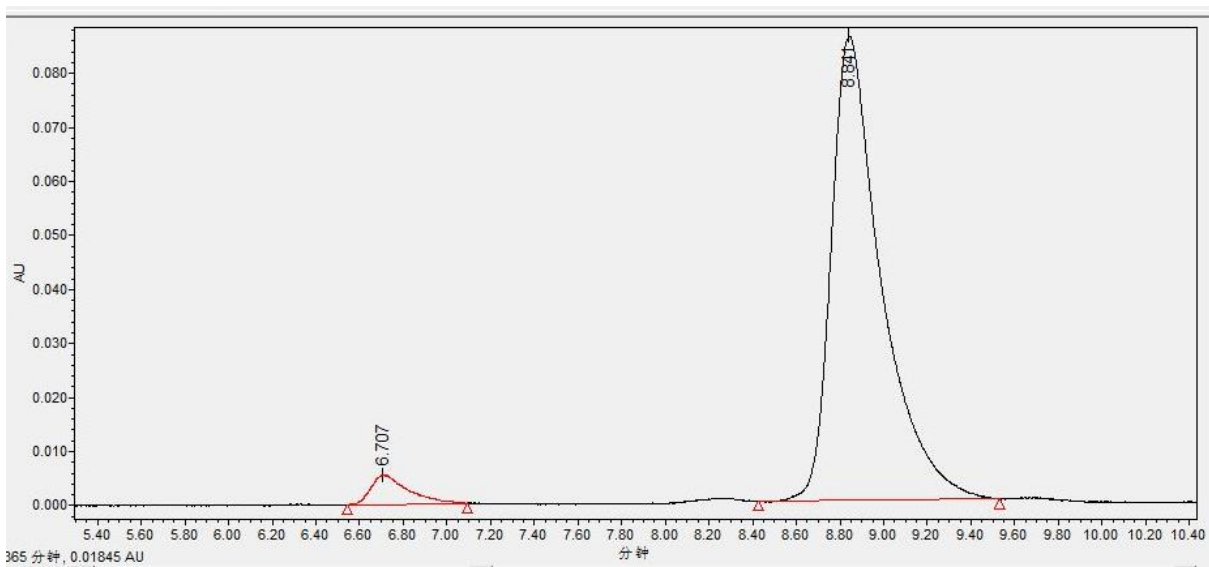
	Time/min	Area	Height	Area%
1	5.704	76575	7241	3.06
2	7.072	242866	45621	96.94



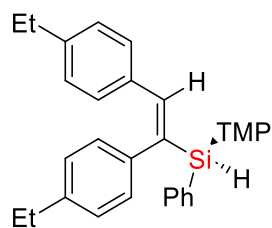
3d



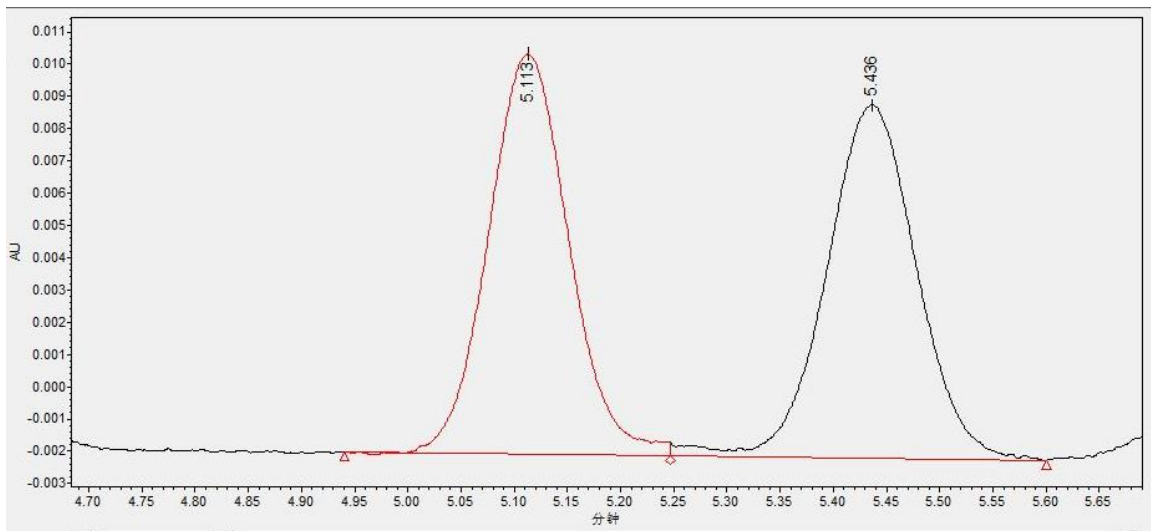
	Time/min	Area	Height	Area%
1	5.819	207023	26508	49.23
2	7.366	213488	20074	50.77



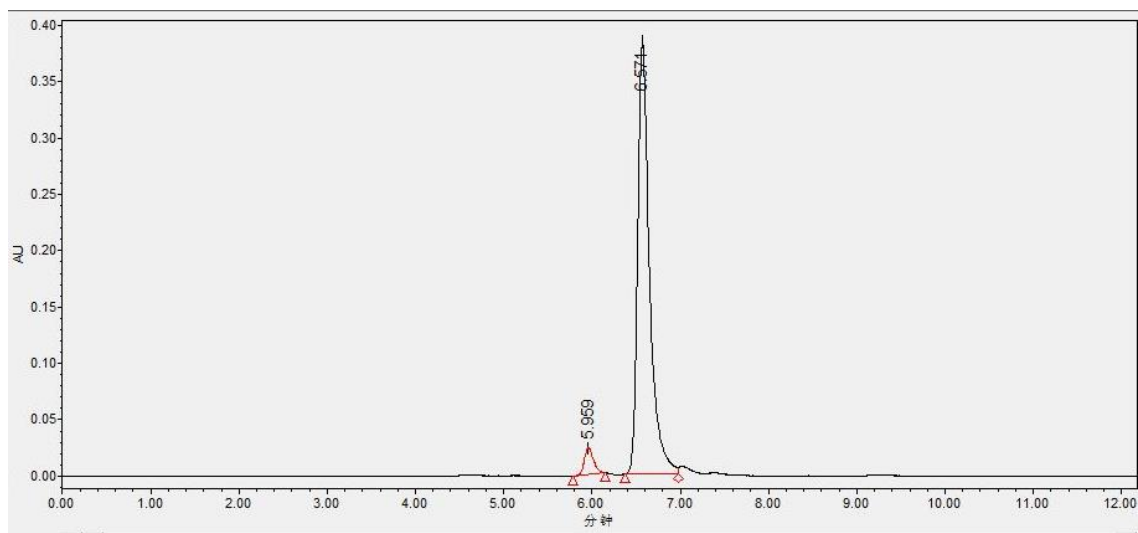
	Time/min	Area	Height	Area%
1	6.707	63702	5494	4.45
2	8.841	13682299	86000	95.55



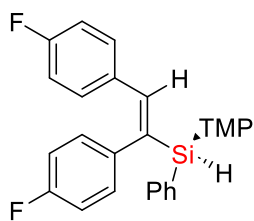
3e



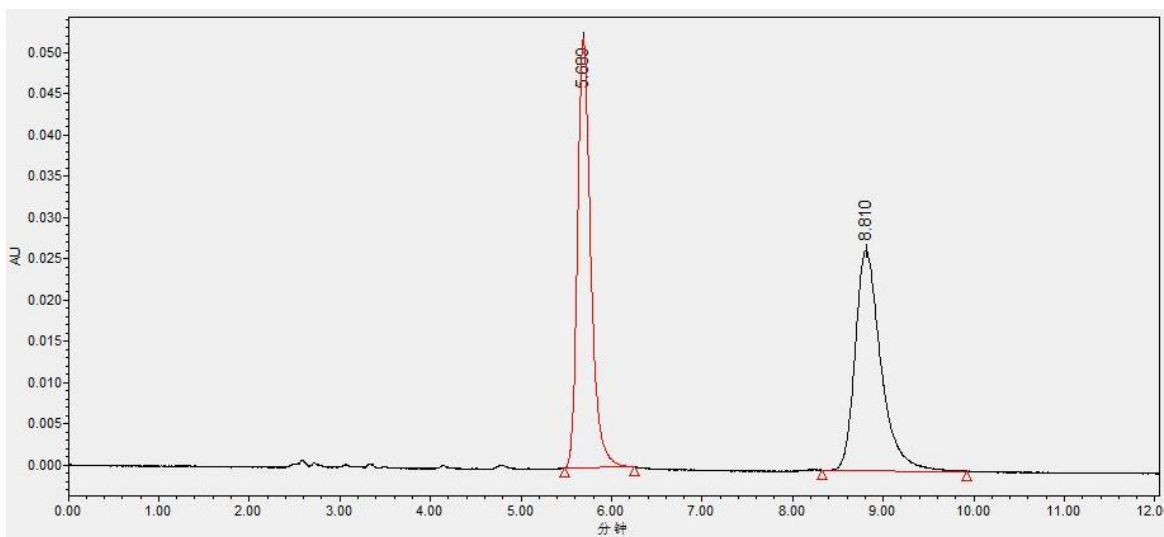
	Time/min	Area	Height	Area%
1	5.113	65342	12411	50.24
2	5.436	64708	10959	49.76



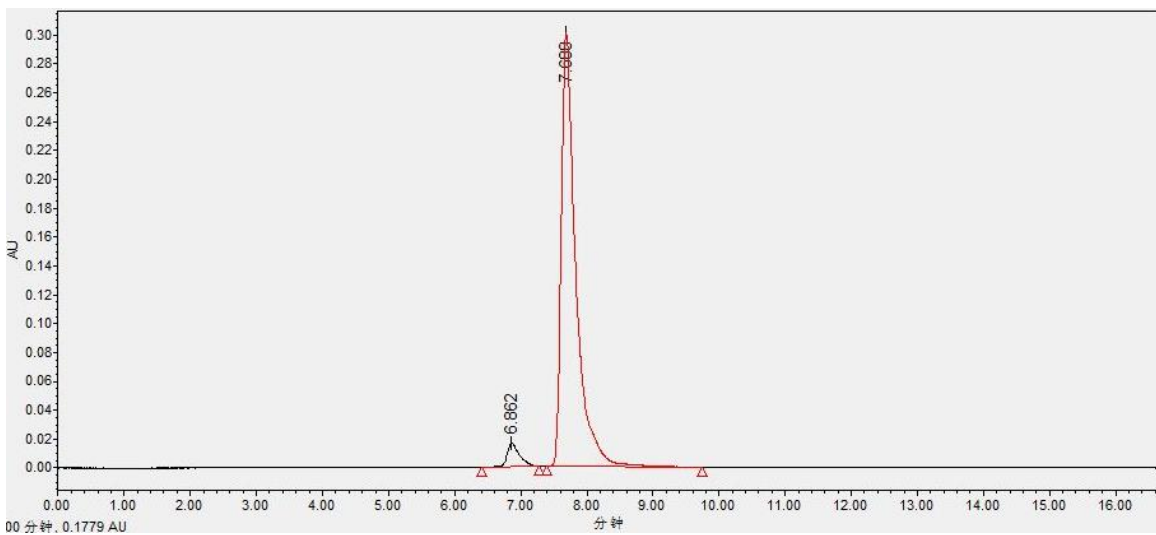
	Time/min	Area	Height	Area%
1	5.959	170546	23756	4.55
2	6.571	3578645	382945	95.45



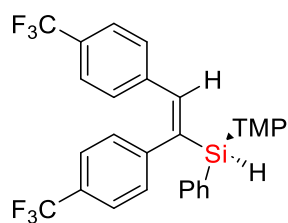
3f



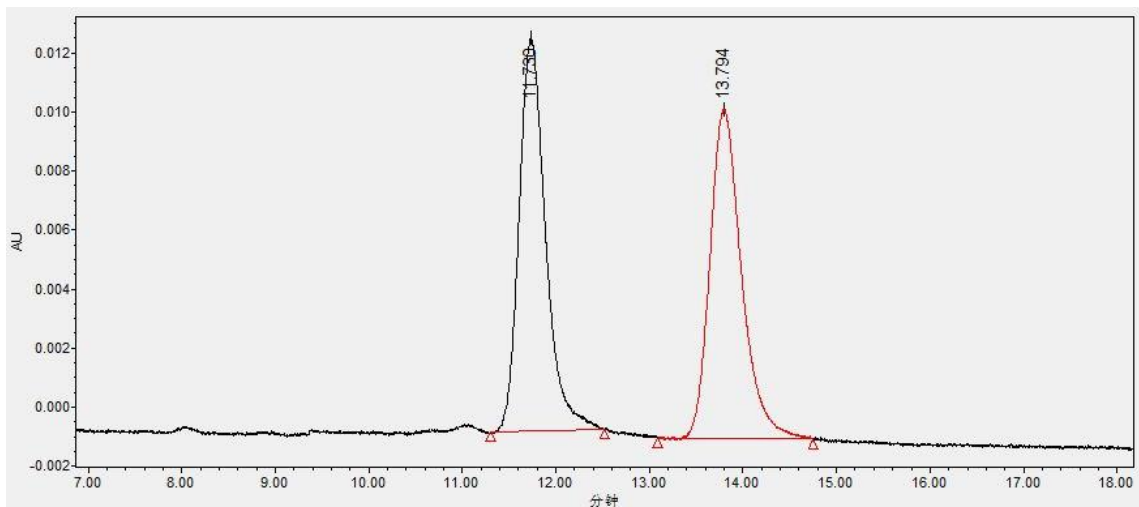
	Time/min	Area	Height	Area%
1	5.689	527907	51909	50.15
2	8.810	524796	26639	49.85



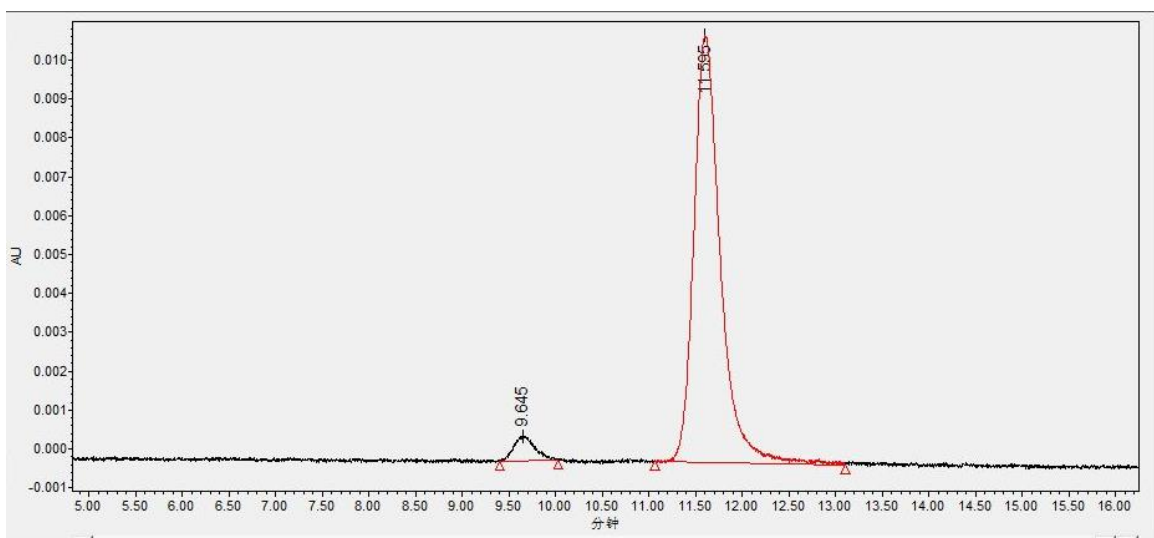
	Time/min	Area	Height	Area%
1	6.862	215398	16584	4.49
2	7.688	4579305	300398	95.51



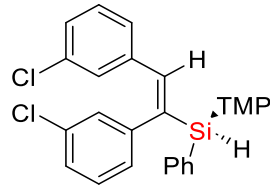
3g



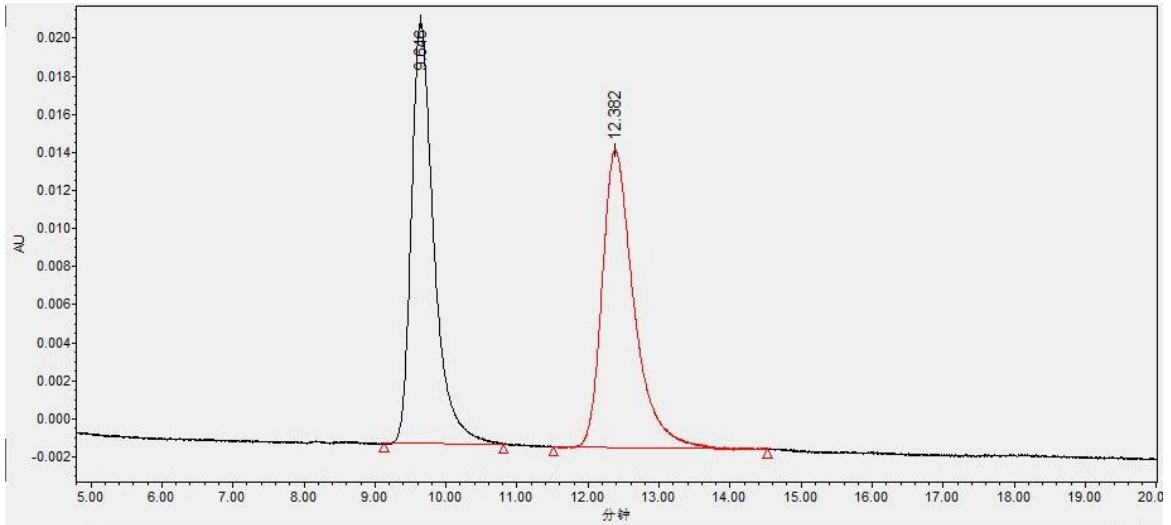
	Time/min	Area	Height	Area%
1	11.730	262328	13313	49.98
2	13.794	262512	11167	50.02



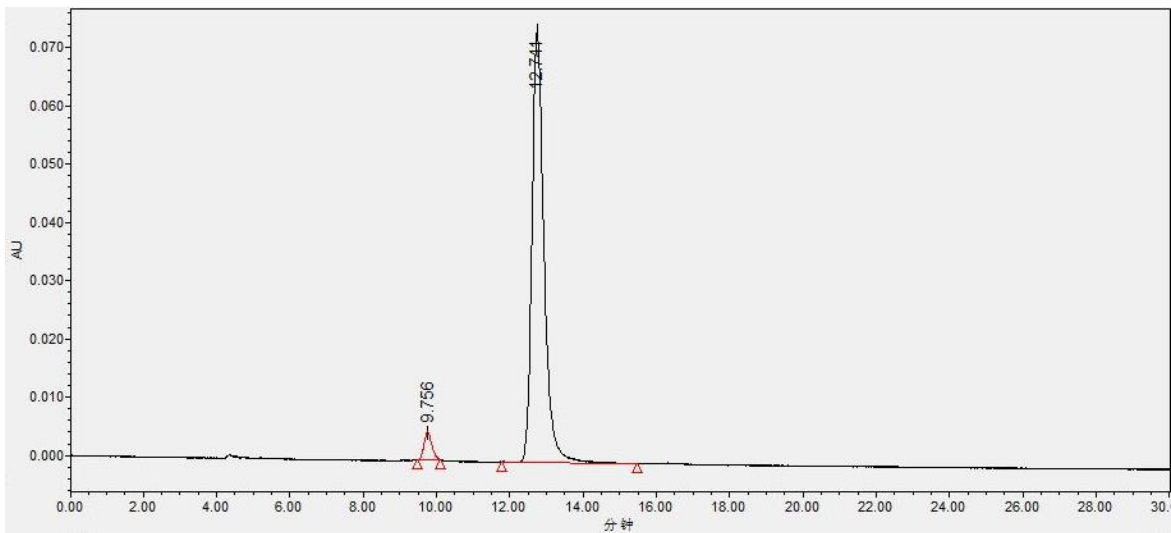
	Time/min	Area	Height	Area%
1	9.645	10034	649	4.41
2	11.595	217507	10976	95.59



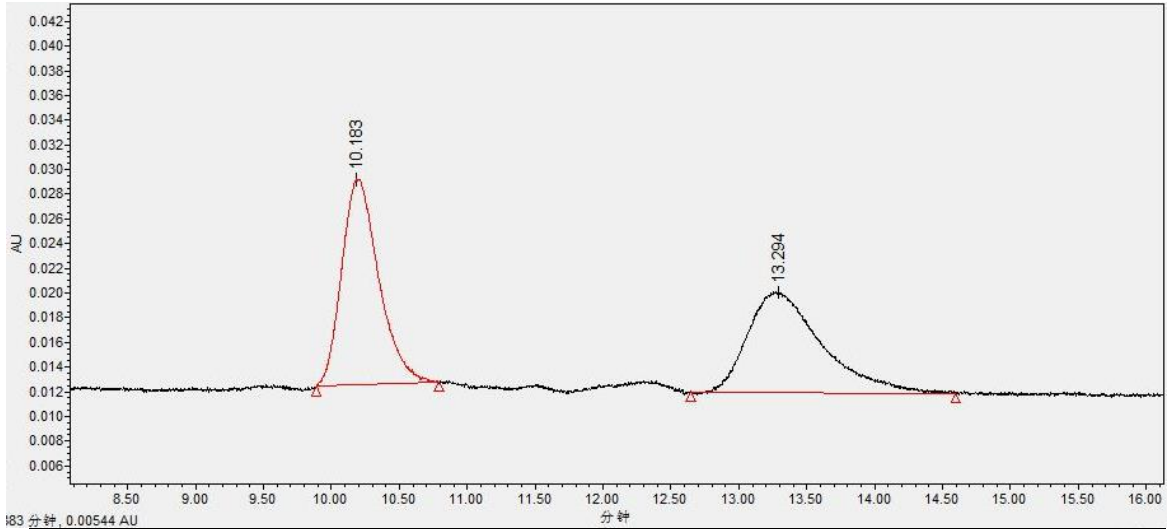
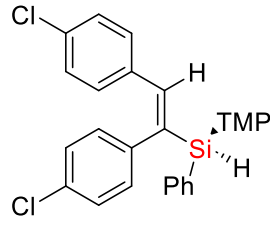
3h



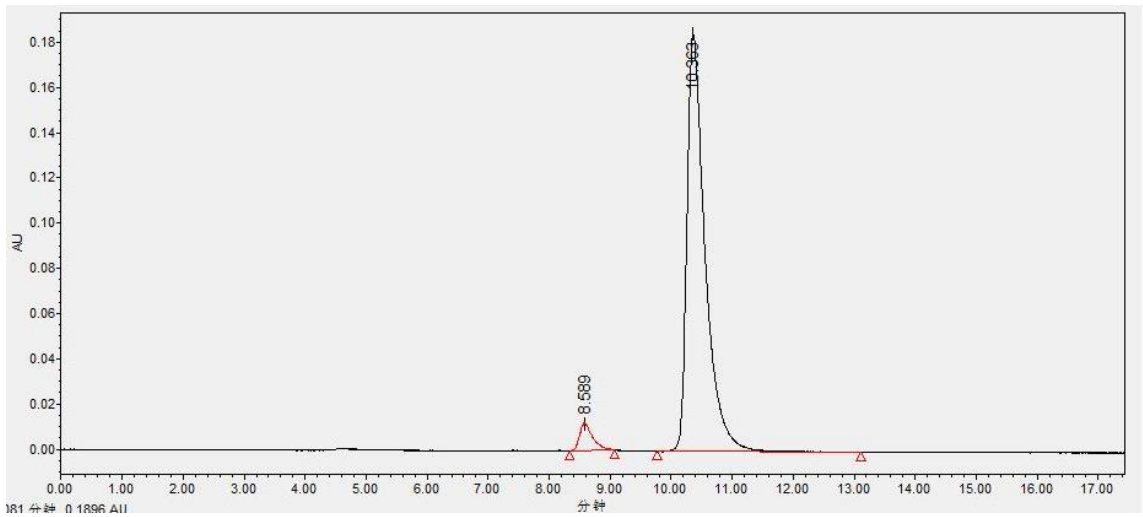
	Time/min	Area	Height	Area%
1	9.464	486739	22160	50.01
2	12.382	486580	15655	49.99



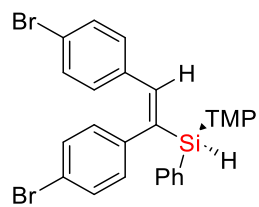
	Time/min	Area	Height	Area%
1	9.756	70144	4801	3.96
2	12.741	1702092	74040	96.04



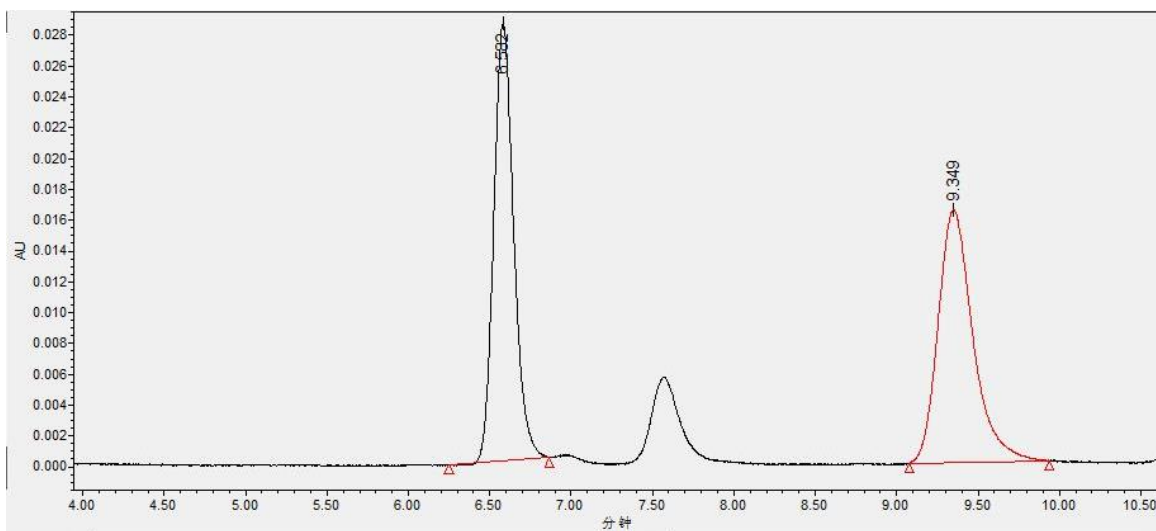
	Time/min	Area	Height	Area%
1	10.183	317200	16701	50.95
2	13.294	305313	8132	49.05



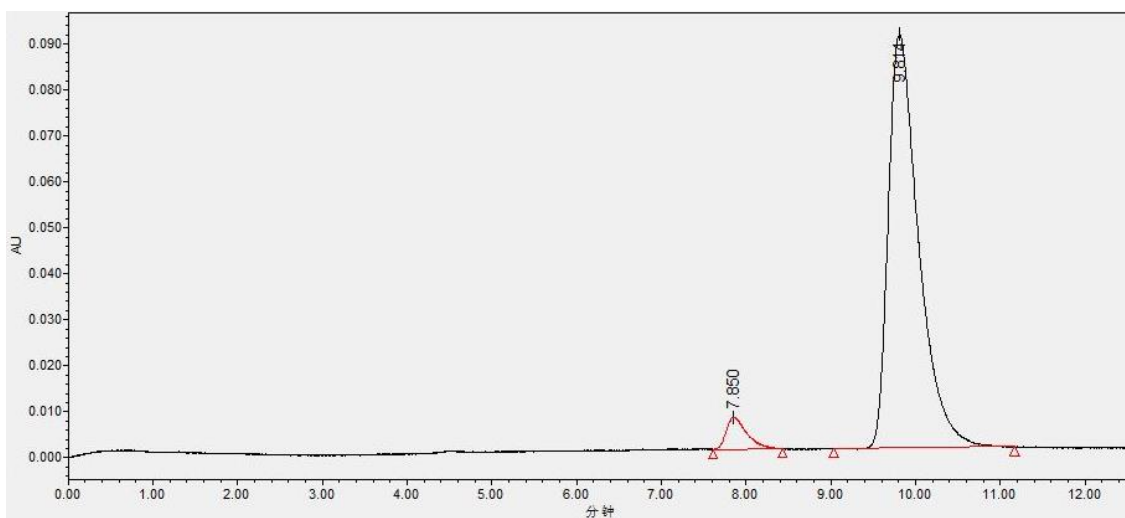
	Time/min	Area	Height	Area%
1	8.589	180187	12080	4.50
2	10.363	3824919	184641	95.50



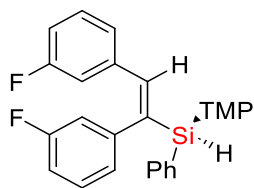
3j



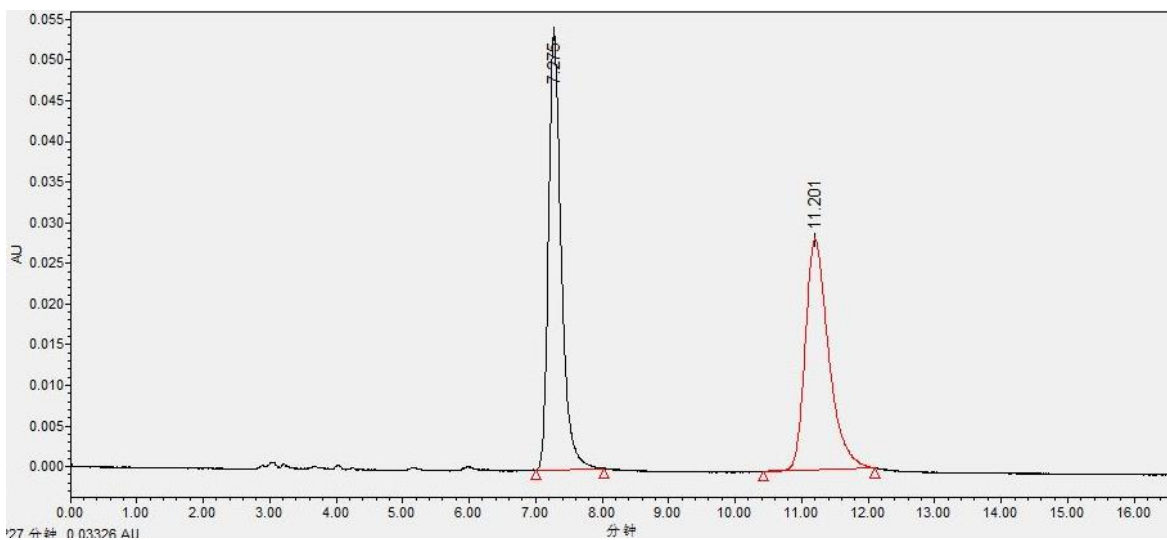
	Time/min	Area	Height	Area%
1	6.582	242142	28385	49.90
2	9.349	243130	16403	50.10



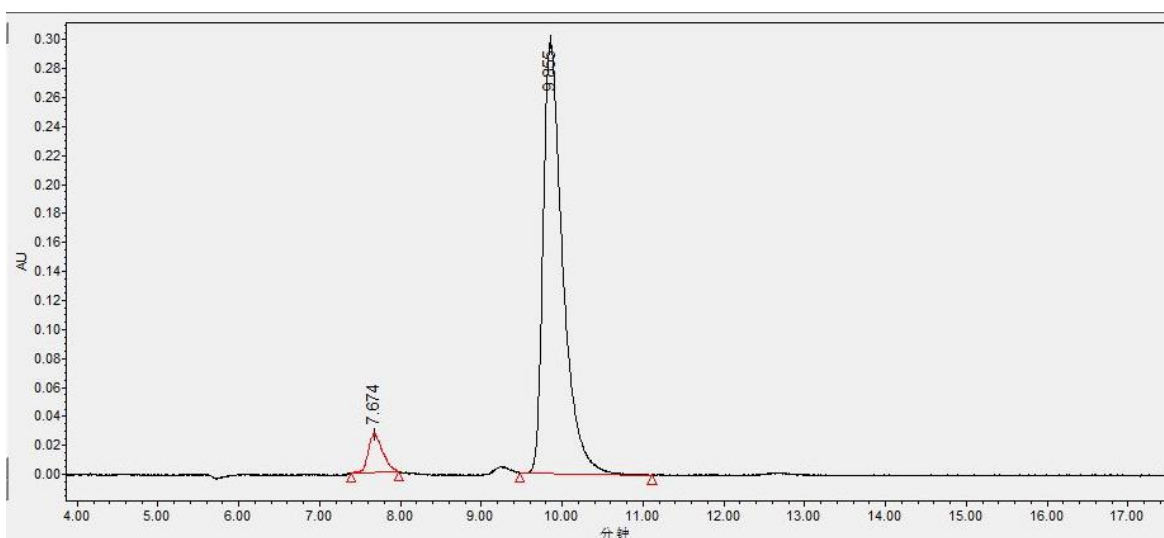
	Time/min	Area	Height	Area%
1	7.850	117167	6952	4.97
2	9.814	2240643	89998	95.03



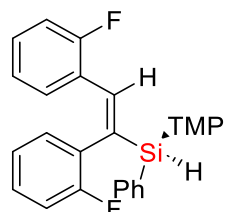
3k



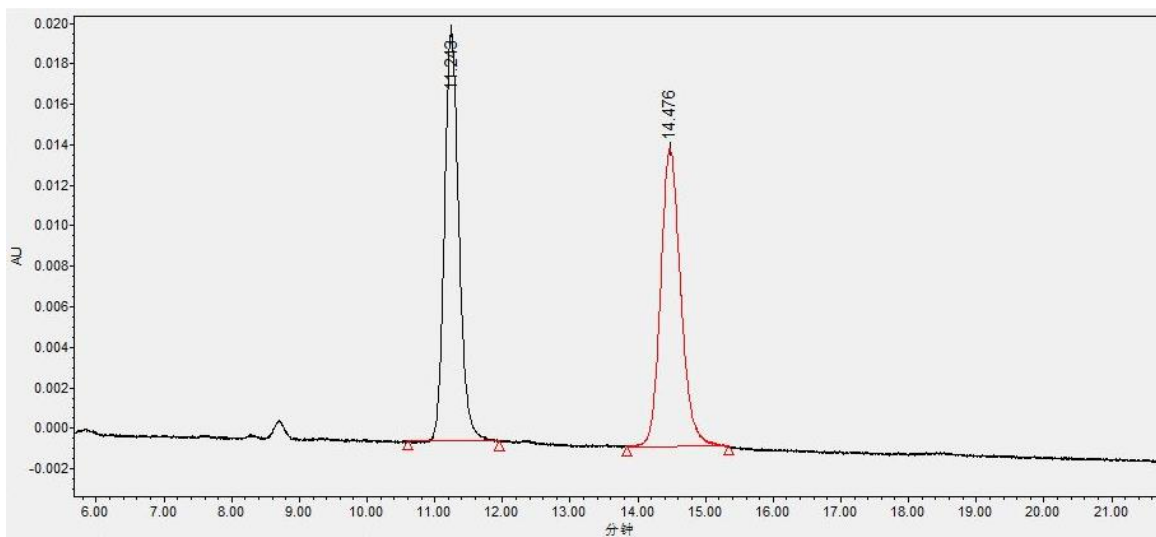
	Time/min	Area	Height	Area%
1	7.275	718808	53542	50.89
2	11.201	693734	28403	49.11



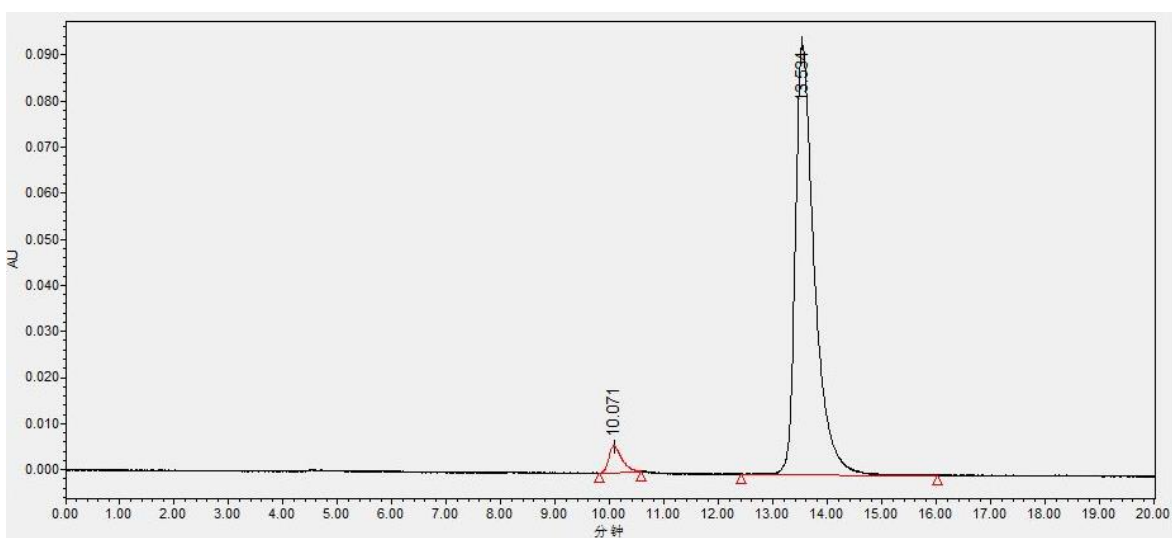
	Time/min	Area	Height	Area%
1	7.674	324505	26520	6.09
2	9.855	5008261	297423	93.91



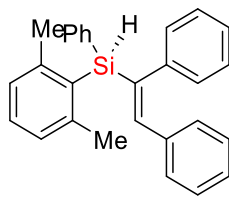
3I



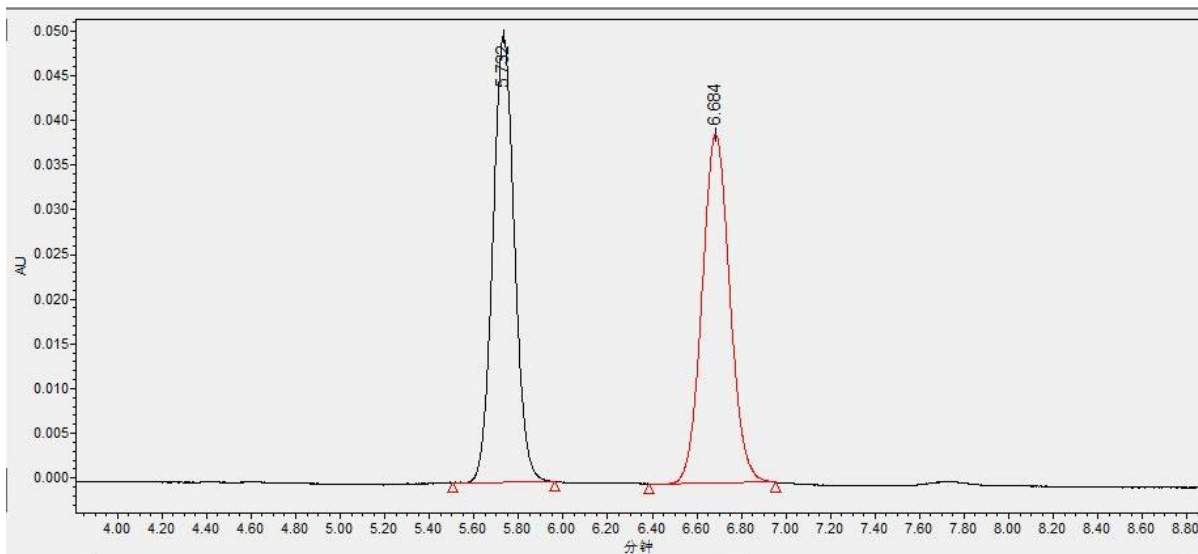
	Time/min	Area	Height	Area%
1	11.243	301741	20238	49.98
2	14.476	301978	14712	50.02



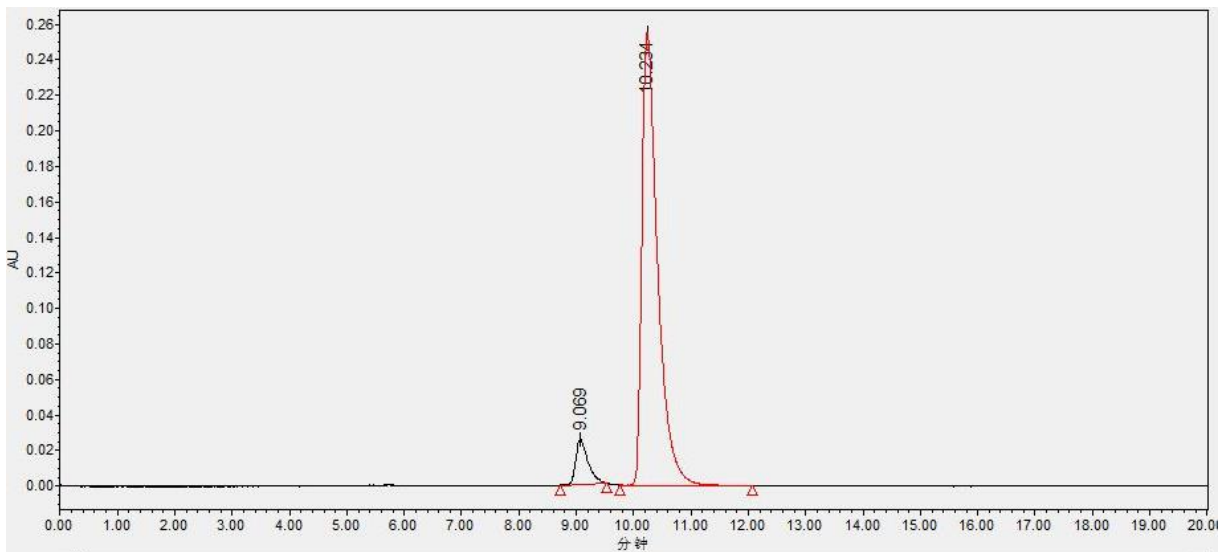
	Time/min	Area	Height	Area%
1	10.071	94473	5765	3.99
2	13.534	2274083	93517	96.01



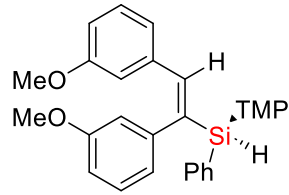
3m



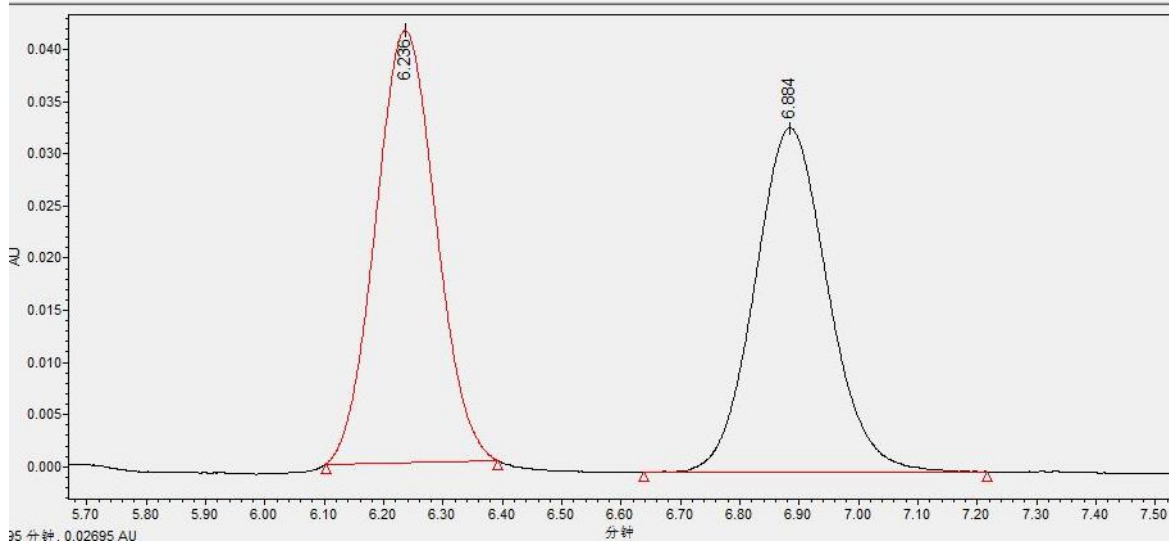
	Time/min	Area	Height	Area%
1	5.732	335533	49775	49.90
2	6.684	336879	38983	50.10



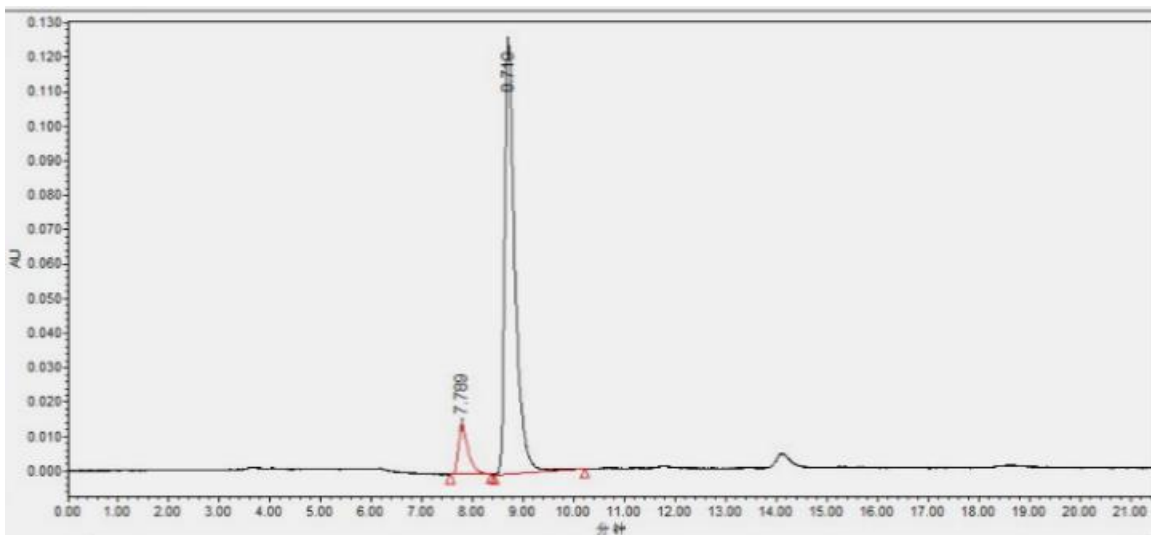
	Time/min	Area	Height	Area%
1	9.069	376365	25532	7.19
2	10.234	4859147	254651	92.81



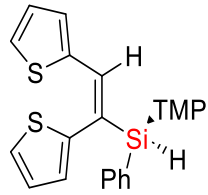
3n



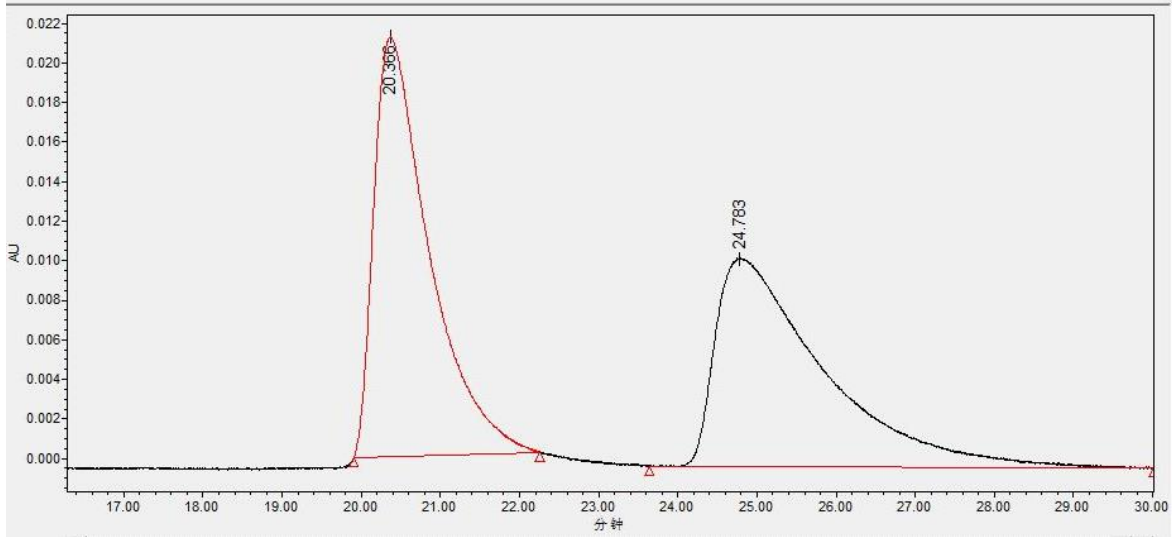
	Time/min	Area	Height	Area%
1	6.236	292916	41404	50.91
2	6.884	282412	33006	49.09



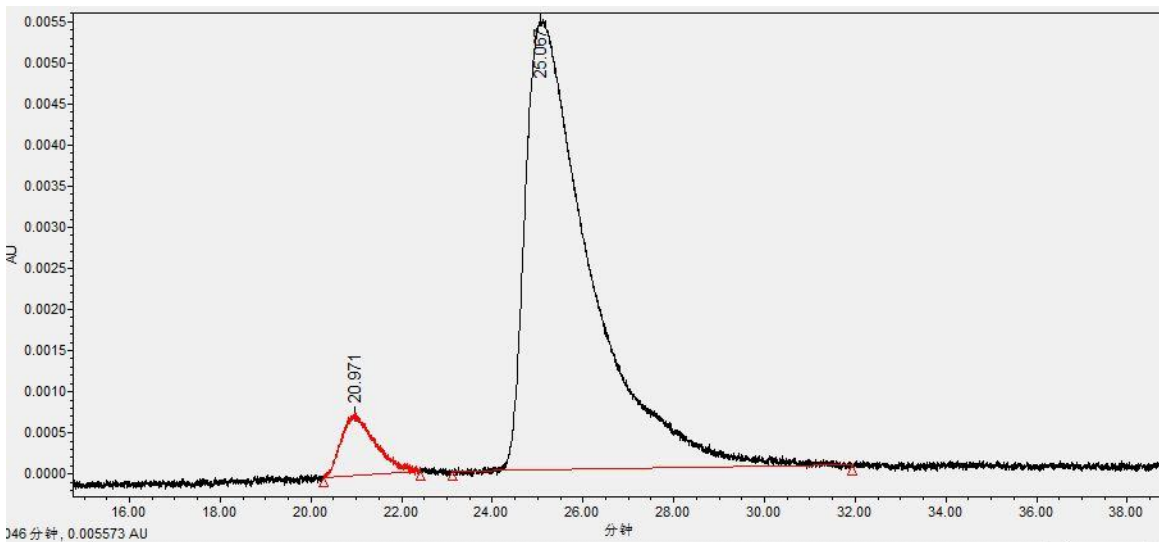
	Time/min	Area	Height	Area%
1	7.789	191605	14665	9.80
2	8.710	176316	124880	90.20



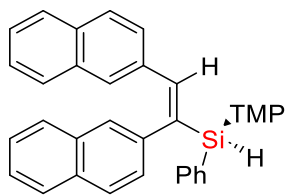
3o



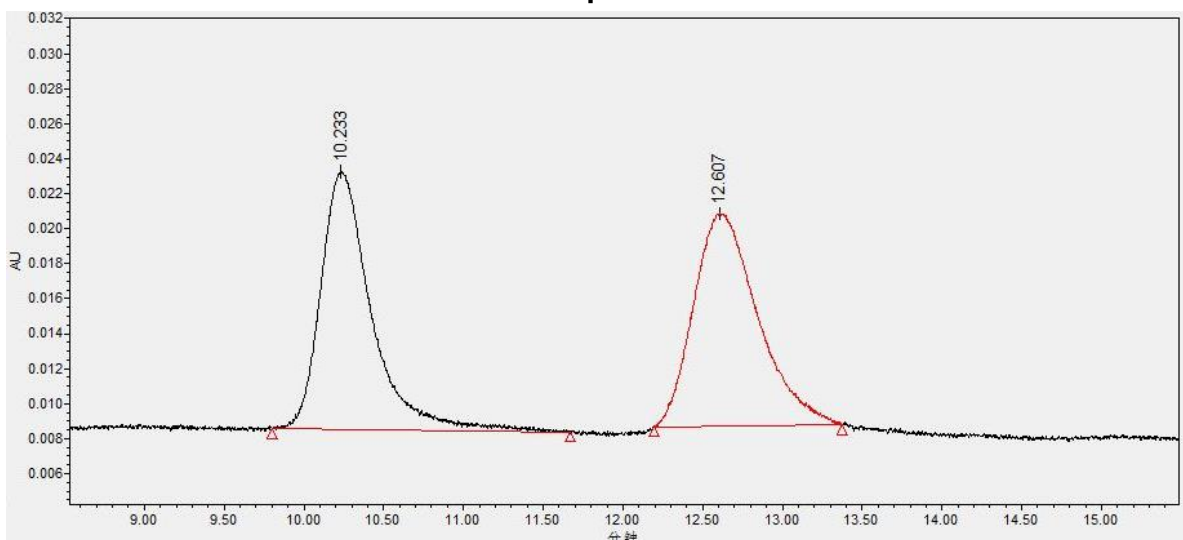
	Time/min	Area	Height	Area%
1	20.366	1028800	21238	50.86
2	24.783	994195	10534	49.14



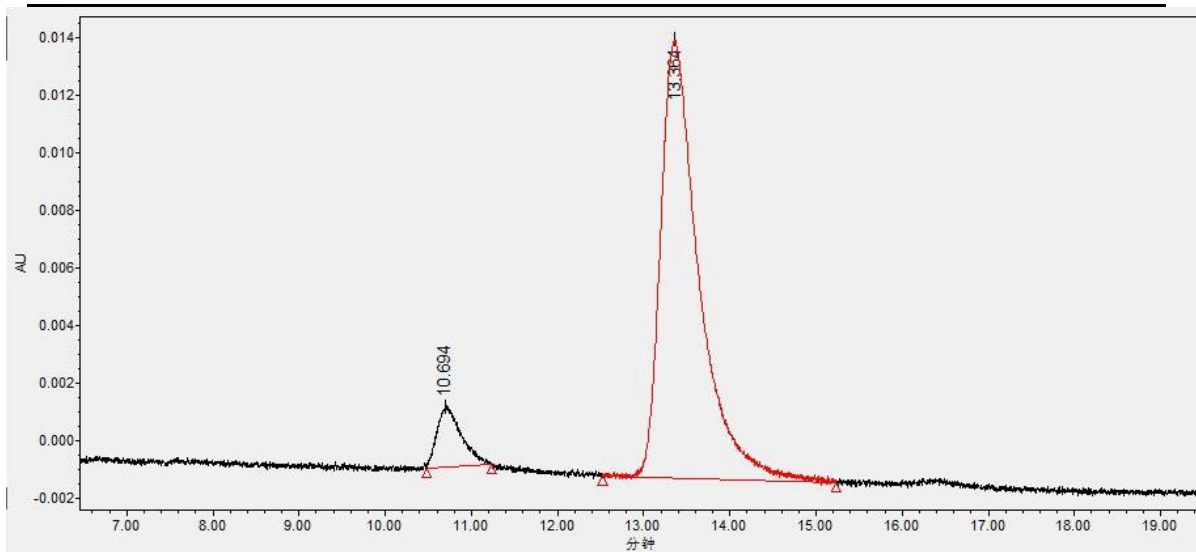
	Time/min	Area	Height	Area%
1	20.971	38047	759	6.55
2	25.067	543166	5478	93.45



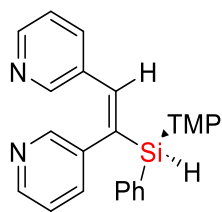
3p



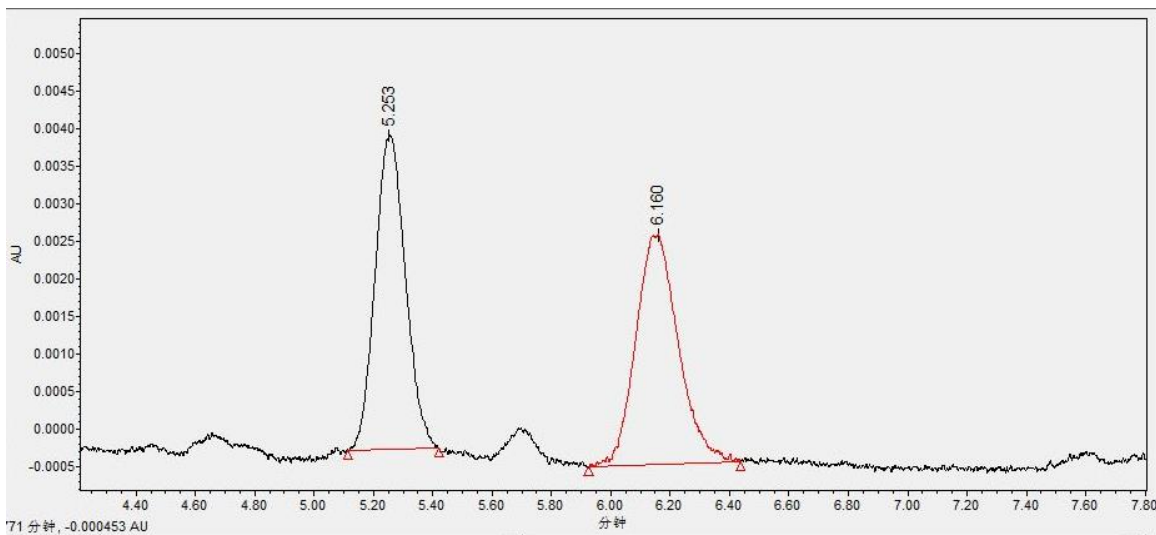
	Time/min	Area	Height	Area%
1	10.233	328446	14809	49.04
2	12.607	341350	12208	50.96



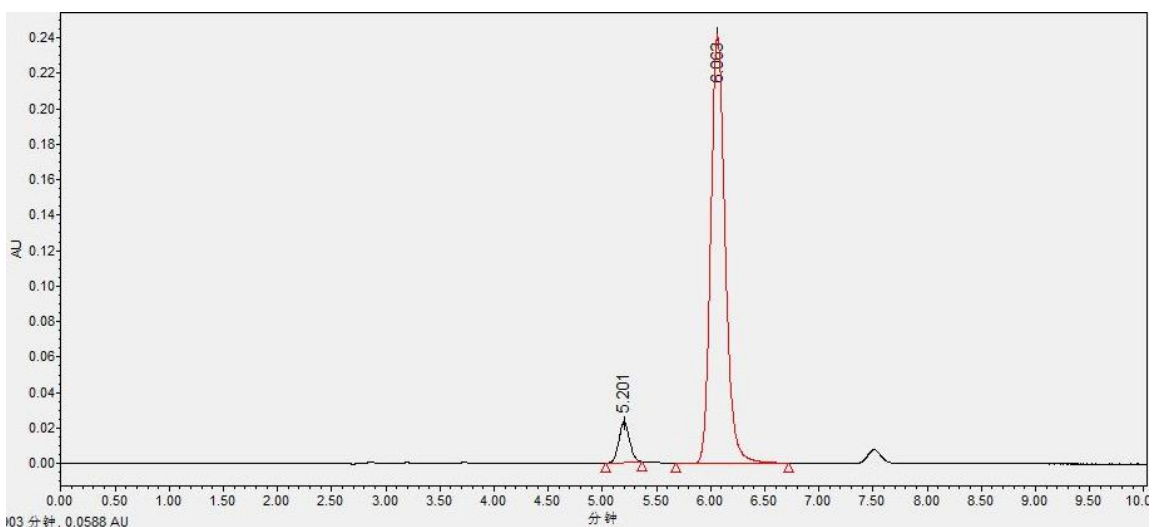
	Time/min	Area	Height	Area%
1	10.694	40506	2114	7.93
2	13.364	470327	15202	92.07



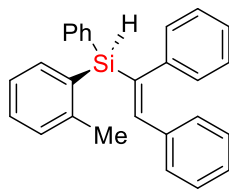
3q



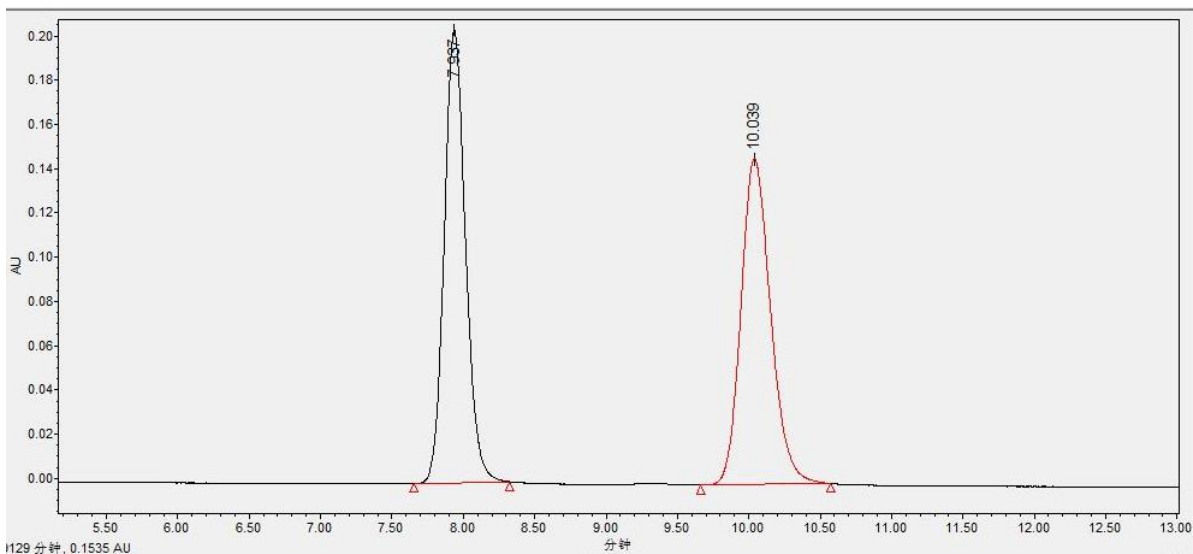
	Time/min	Area	Height	Area%
1	5.253	29786	4197	49.31
2	6.160	30619	3073	50.69



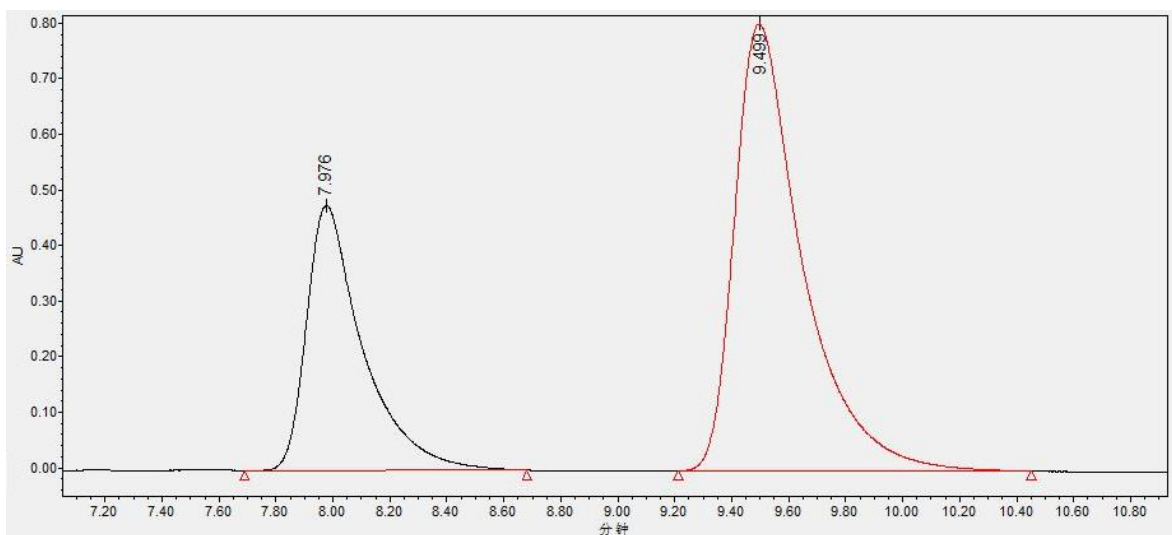
	Time/min	Area	Height	Area%
1	5.201	155300	22651	6.50
2	6.063	2234795	241941	93.50



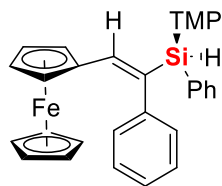
3r



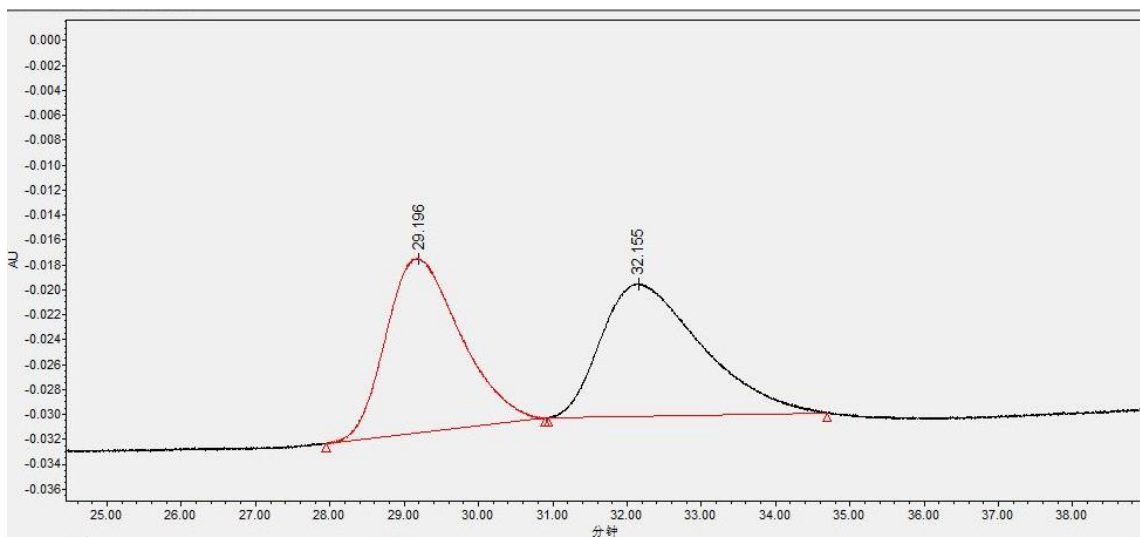
	Time/min	Area	Height	Area%
1	7.937	2121153	204569	49.95
2	10.039	2125697	147146	50.05



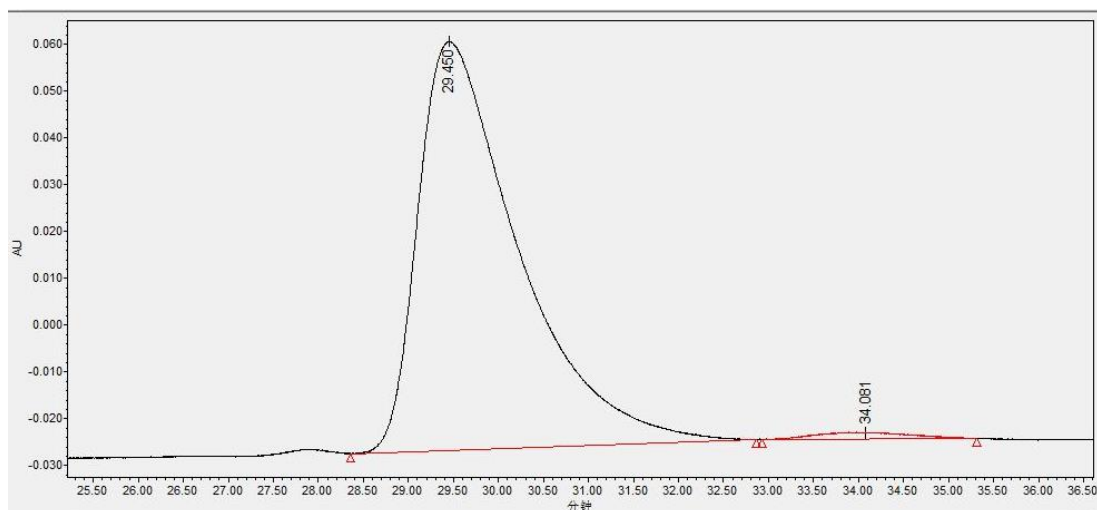
	Time/min	Area	Height	Area%
1	7.976	6541942	476958	32.92
2	9.499	13332400	804749	67.08



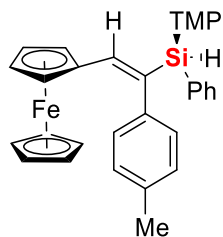
9a



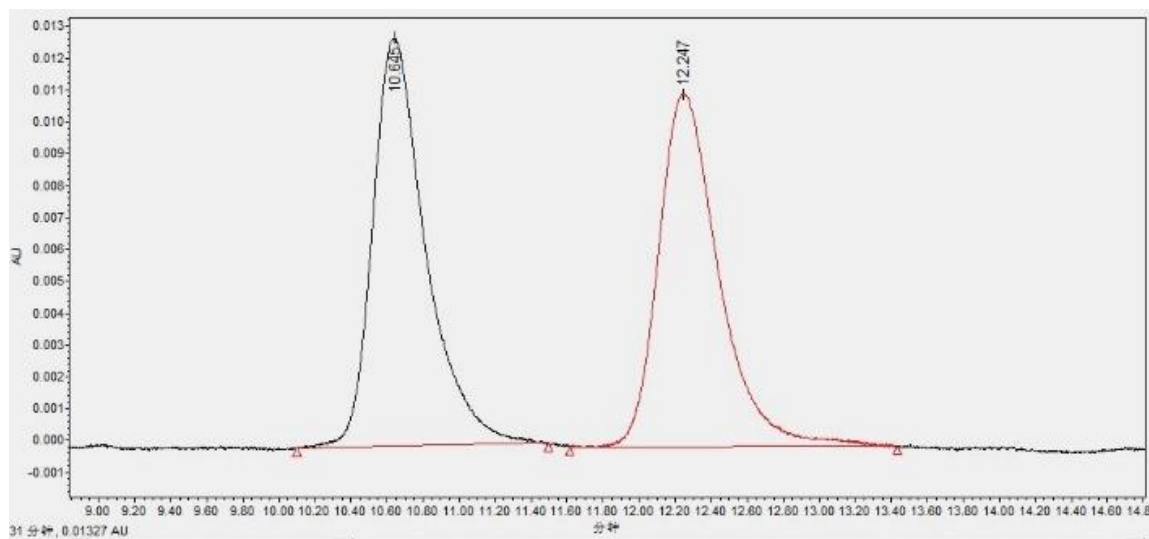
	Time/min	Area	Height	Area%
1	29.196	971007	14004	49.28
2	32.155	999438	10662	50.72



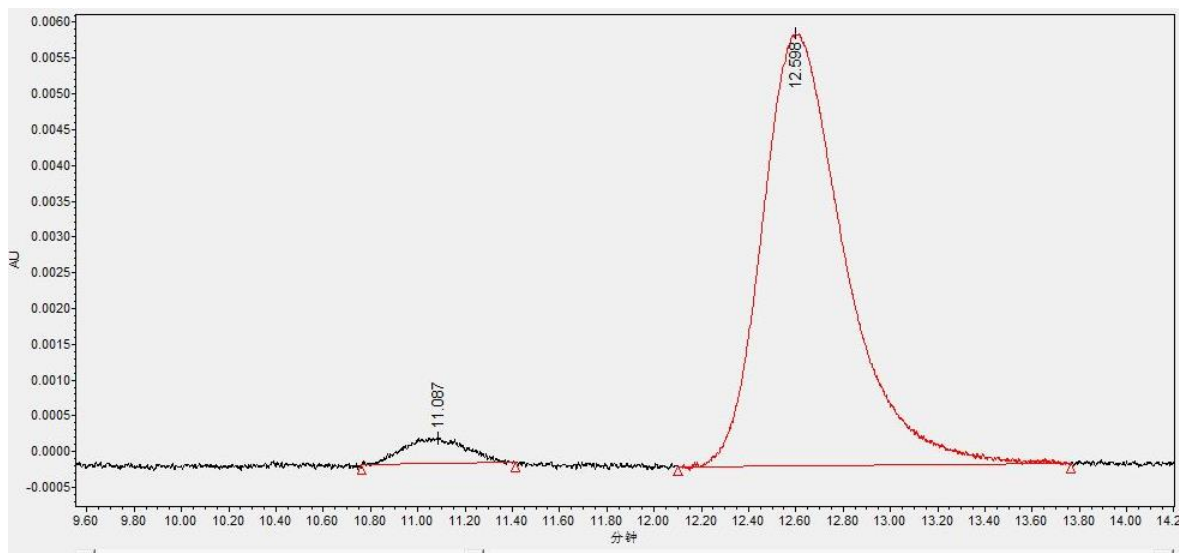
	Time/min	Area	Height	Area%
1	29.450	6750339	87265	1.52
2	34.081	104067	4170	98.48



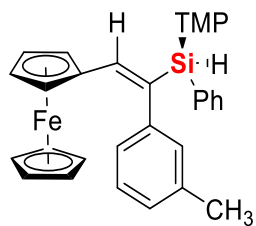
9b



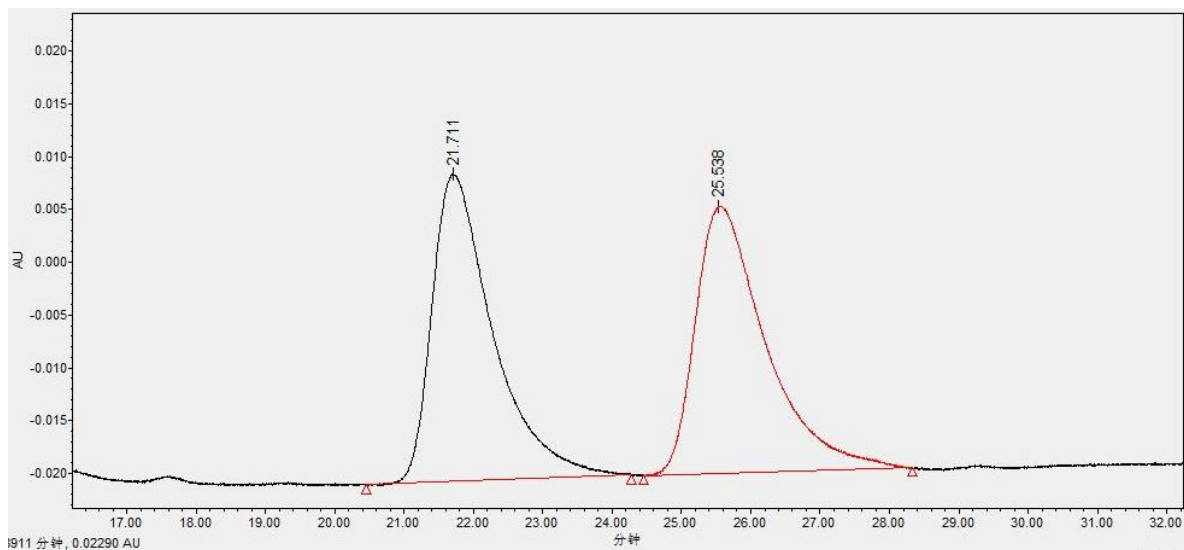
	Time/min	Area	Height	Area%
1	10.645	262454	12803	50.44
2	12.247	257900	11073	49.56



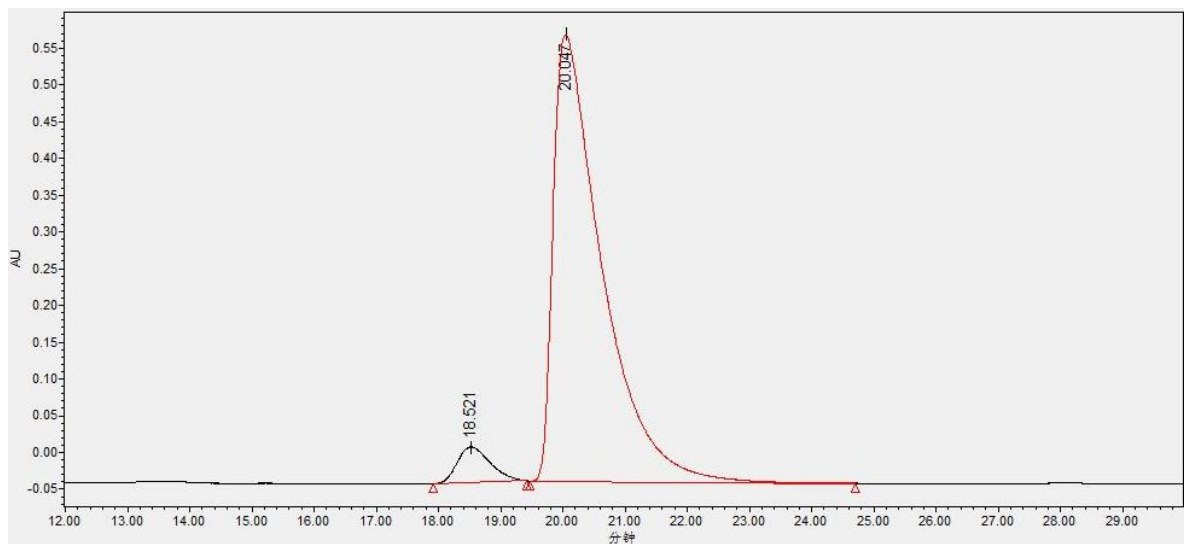
	Time/min	Area	Height	Area%
1	11.035	45845	2399	4.51
2	12.600	971111	40187	95.49



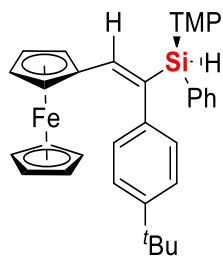
9c



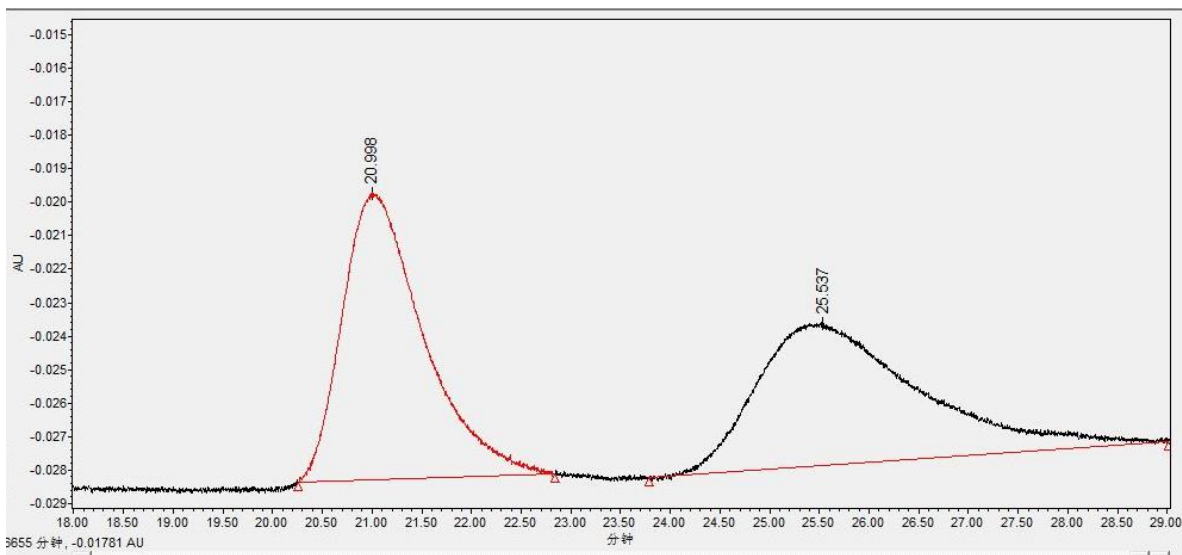
	Time/min	Area	Height	Area%
1	21.711	1785034	29197	49.99
2	25.538	1786105	25363	50.01



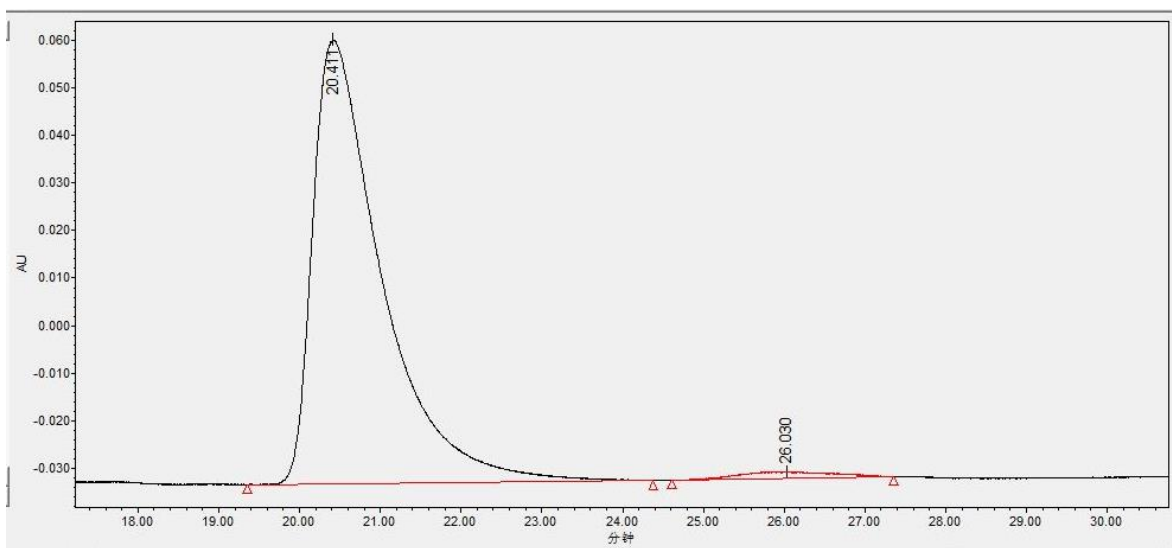
	Time/min	Area	Height	Area%
1	18.521	1781880	48488	5.12
2	20.047	33028250	607257	94.88



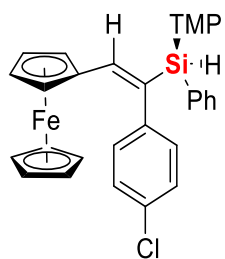
9d



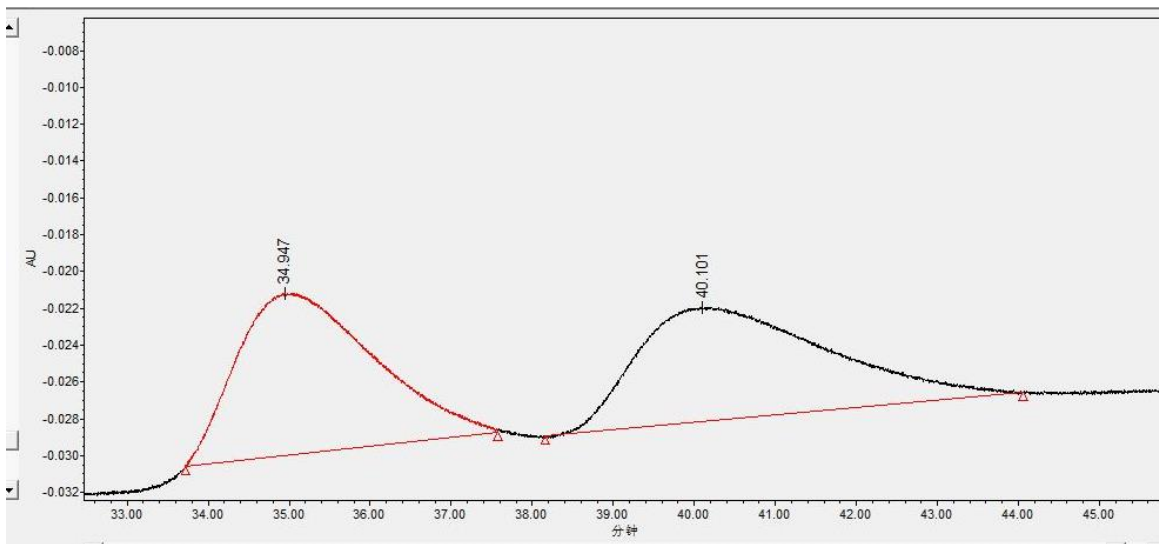
	Time/min	Area	Height	Area%
1	20.998	485373	8586	50.25
2	25.537	480465	4268	49.75



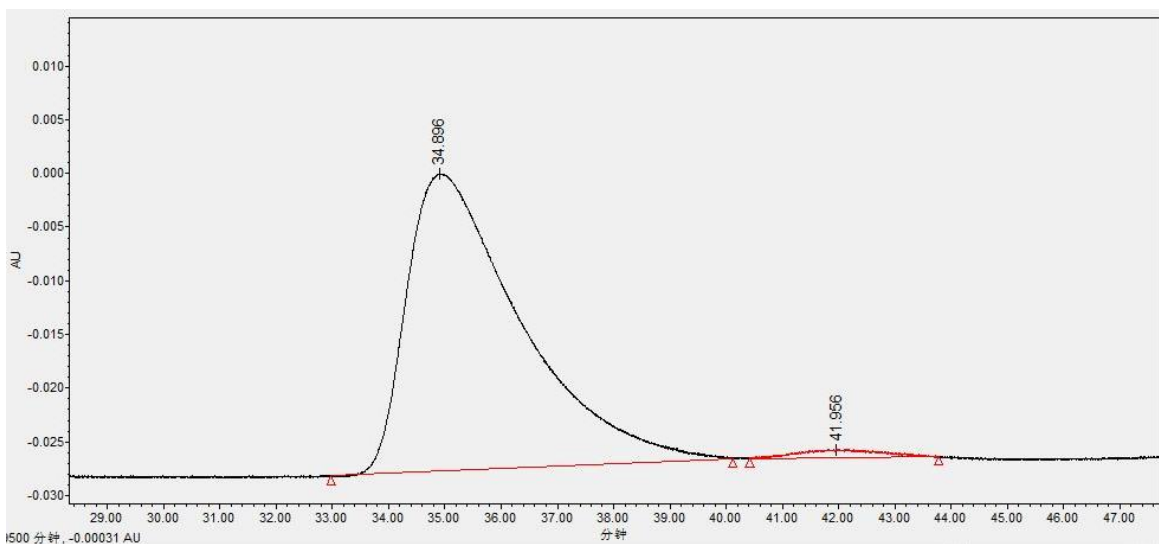
	Time/min	Area	Height	Area%
1	20.411	5522570	93154	98.02
2	26.030	111681	1409	1.98



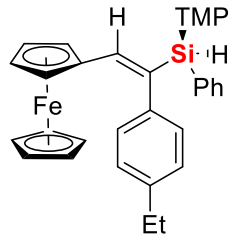
9e



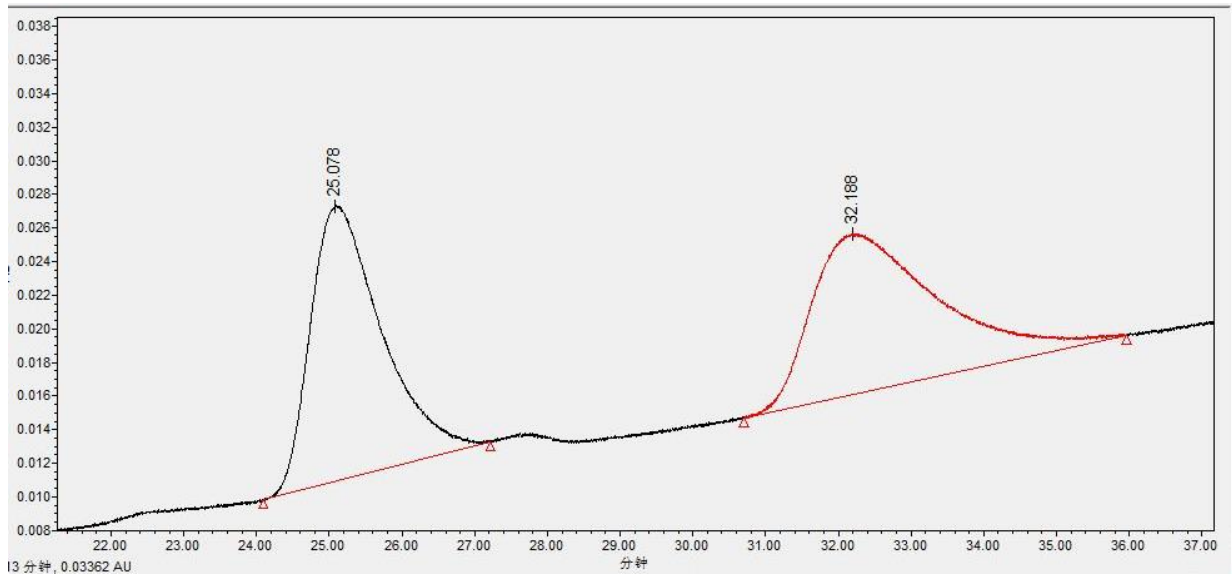
	Time/min	Area	Height	Area%
1	34.947	1029903	8809	50.58
2	40.101	1006181	6229	49.42



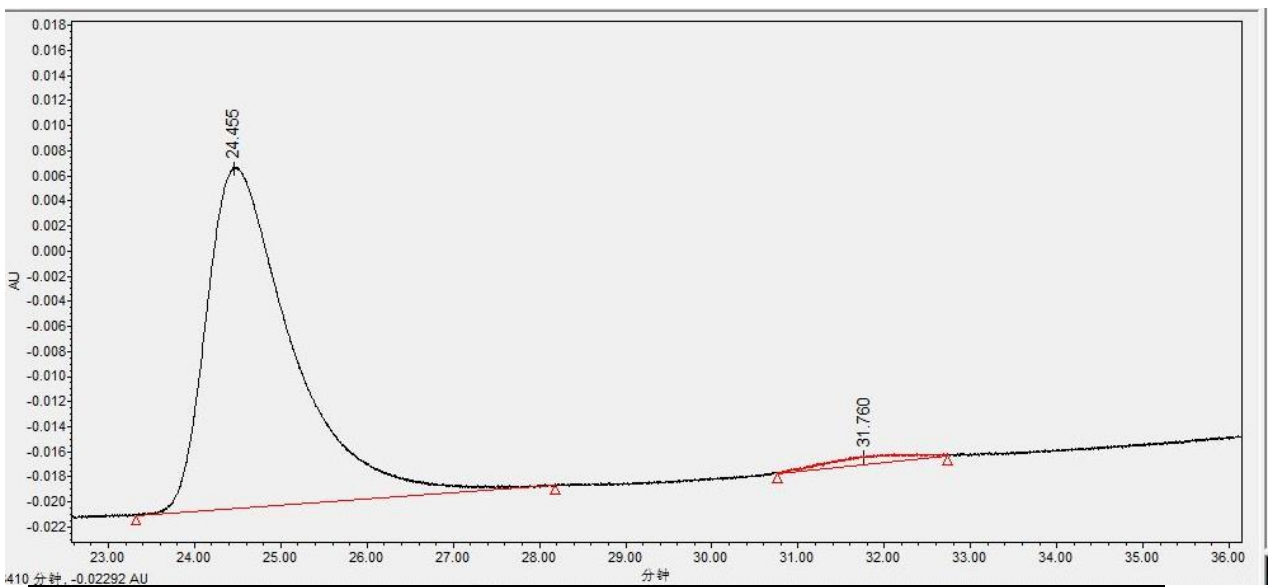
	Time/min	Area	Height	Area%
1	34.896	3897846	27758	98.14
2	41.956	74036	752	1.86



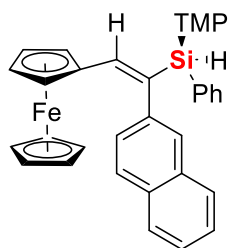
9f



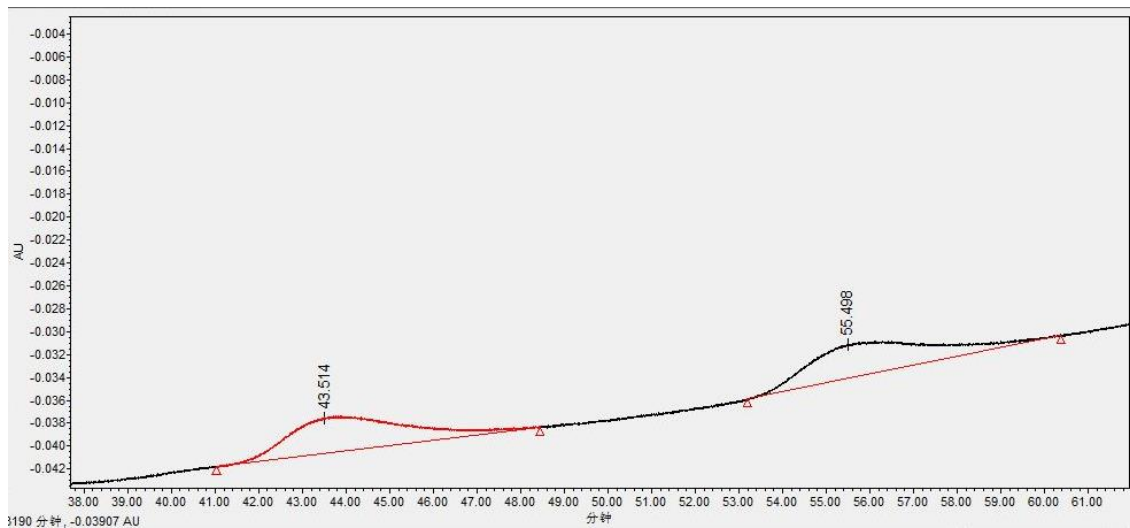
	Time/min	Area	Height	Area%
1	25.078	1110204	16432	49.72
2	32.188	1122869	9576	50.26



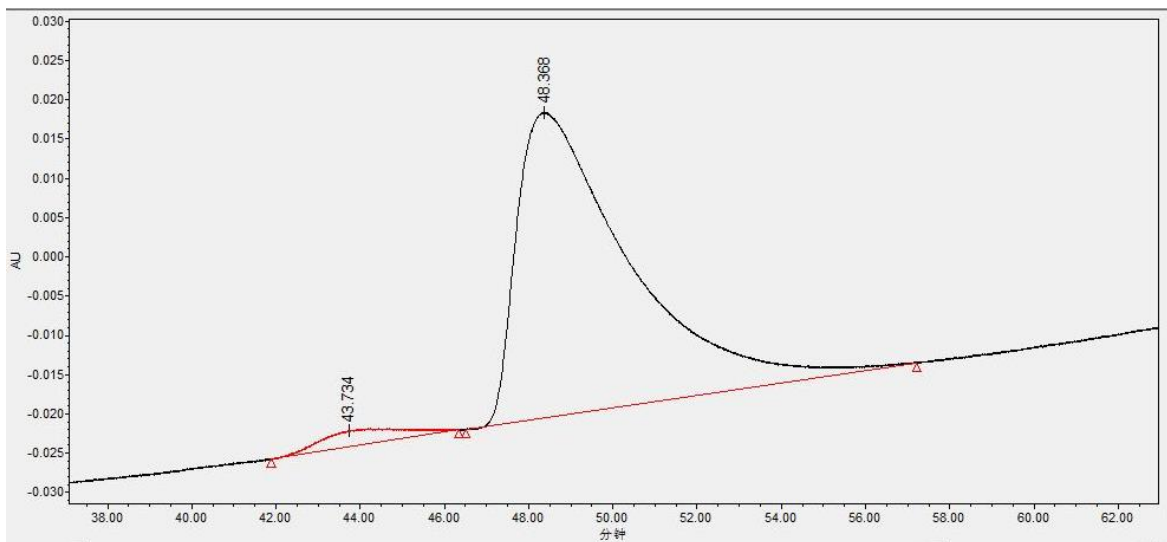
	Time/min	Area	Height	Area%
1	24.455	1863560	27145	97.86
2	31.760	40839	649	2.14



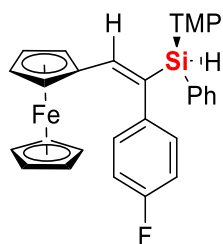
9g



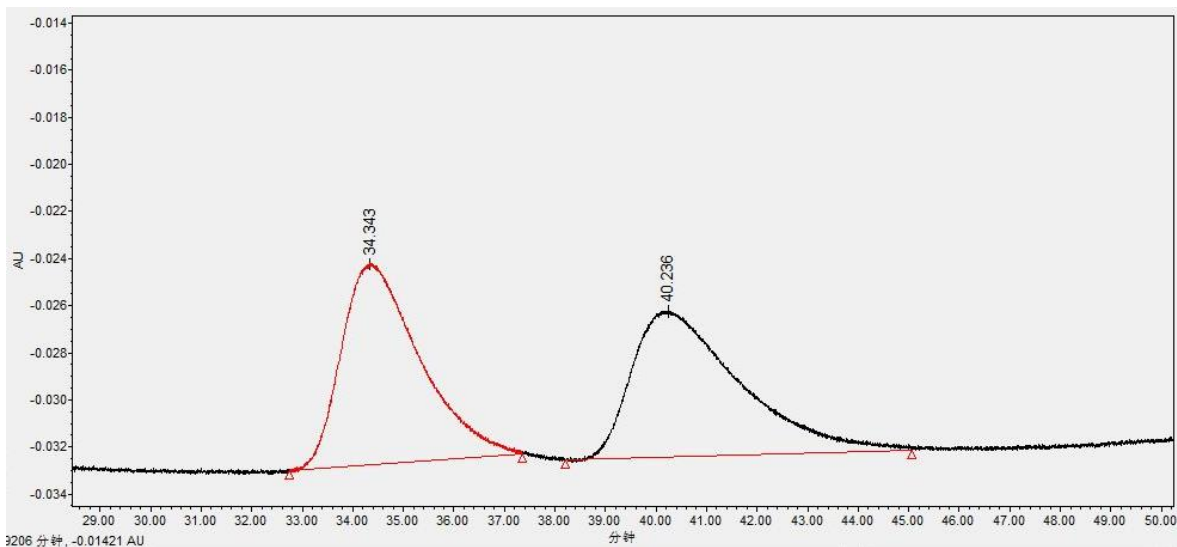
	Time/min	Area	Height	Area%
1	43.514	572265	3150	50.28
2	55.498	565979	3017	49.72



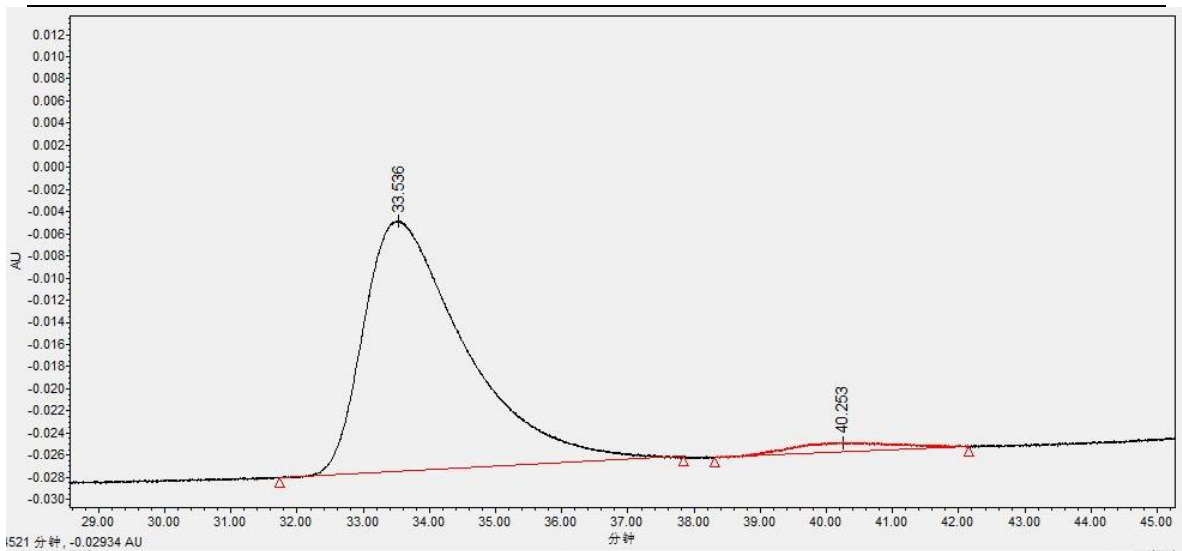
	Time/min	Area	Height	Area%
1	43.734	263976	2064	3.54
2	48.368	7195450	38967	96.46



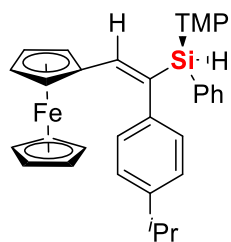
9h



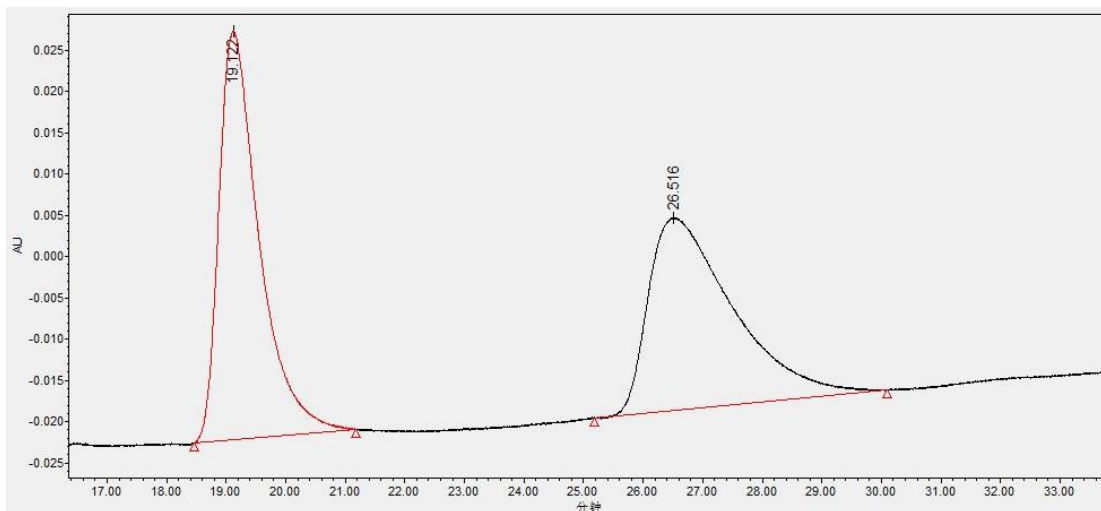
	Time/min	Area	Height	Area%
1	35.371	828081	8675	50.34
2	39.909	816930	6565	49.66



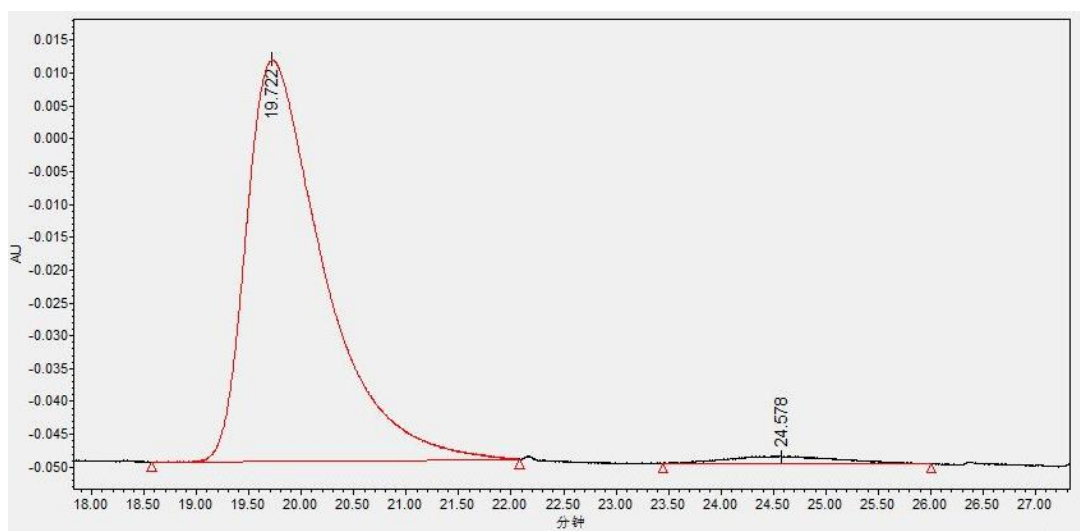
	Time/min	Area	Height	Area%
1	33.536	2418736	22668	96.04
2	40.253	99717	922	3.96



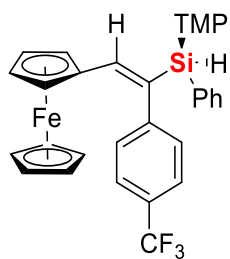
9i



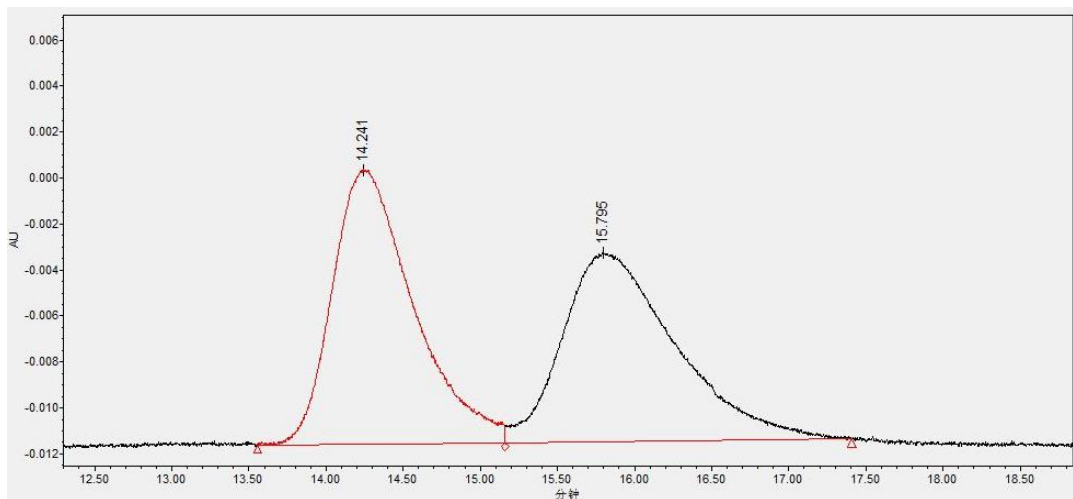
	Time/min	Area	Height	Area%
1	19.122	2321459	49441	50.40
2	26.516	2284722	23327	49.60



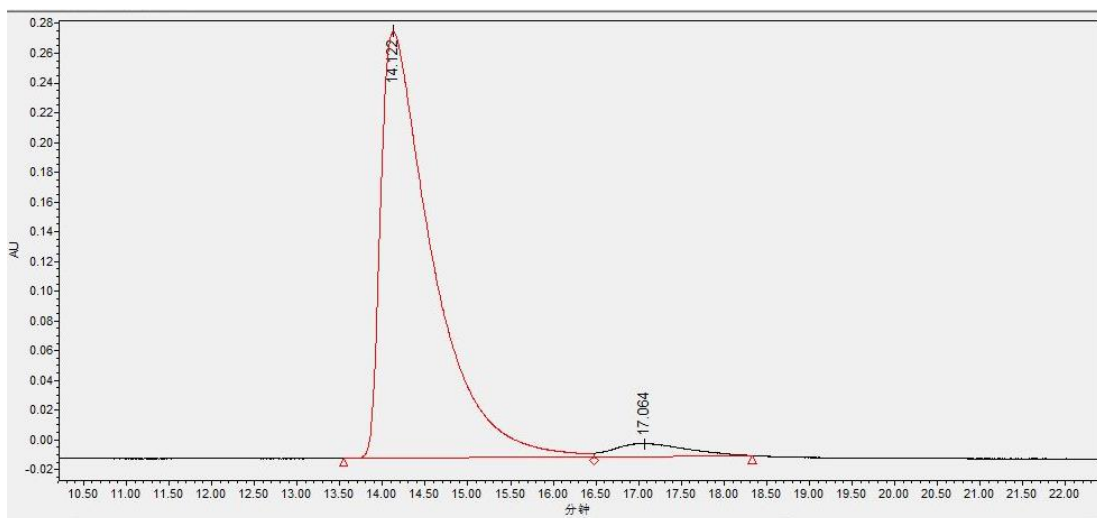
	Time/min	Area	Height	Area%
1	19.722	3131360	61087	97.55
2	24.578	78683	1111	2.45



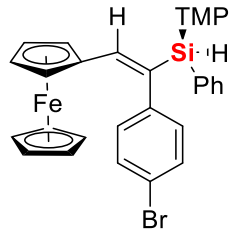
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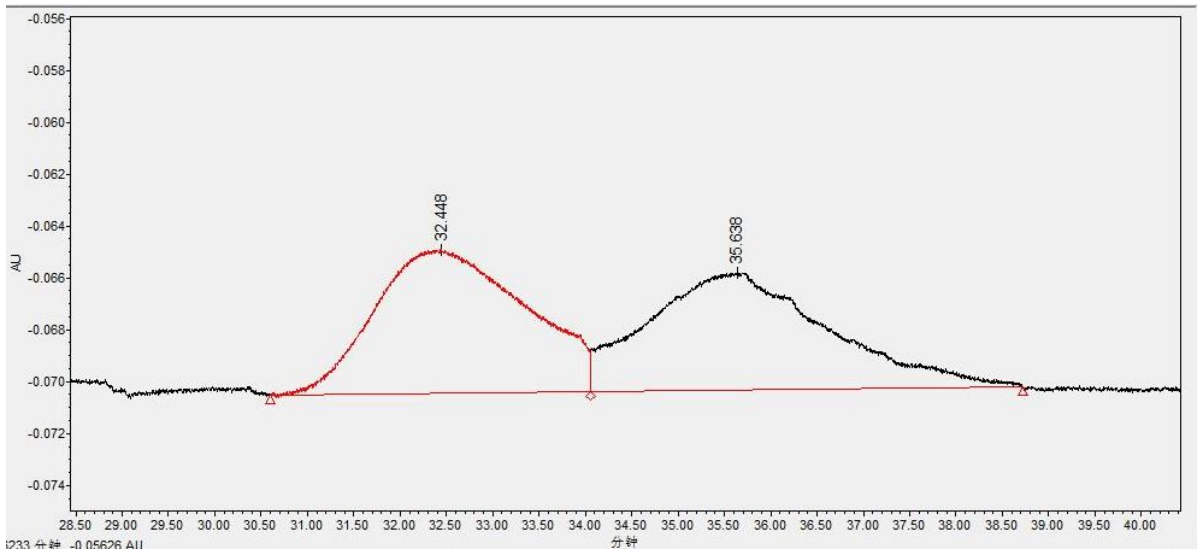
	Time/min	Area	Height	Area%
1	14.241	434400	11918	50.97
2	15.795	417907	8229	49.03



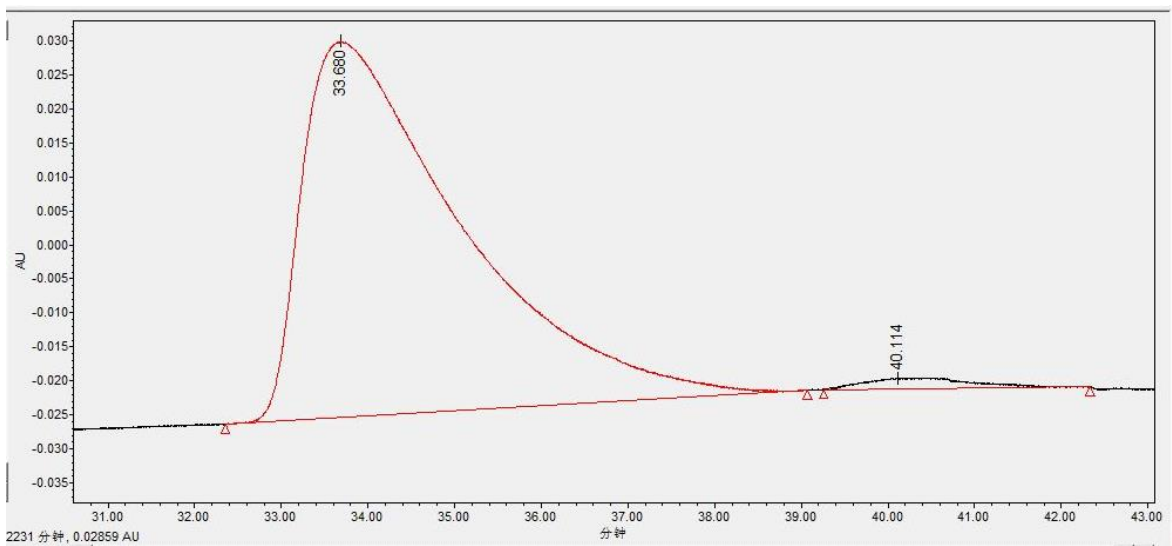
	Time/min	Area	Height	Area%
1	14.122	11983976	286494	95.96
2	17.064	504007	8652	4.04



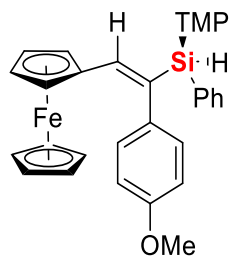
9k



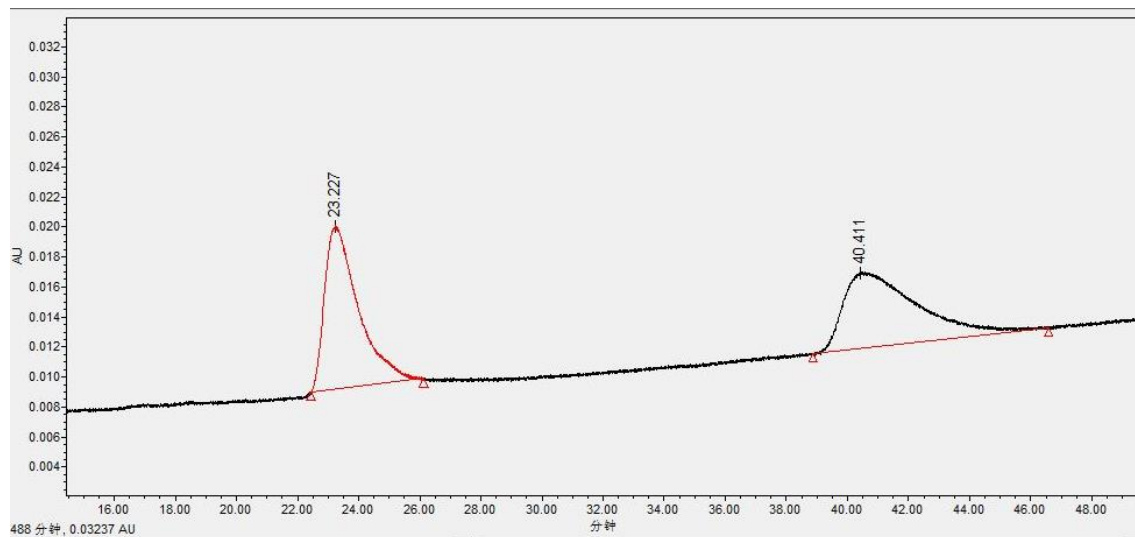
	Time/min	Area	Height	Area%
1	32.448	626729	5556	50.68
2	35.638	609939	4535	49.32



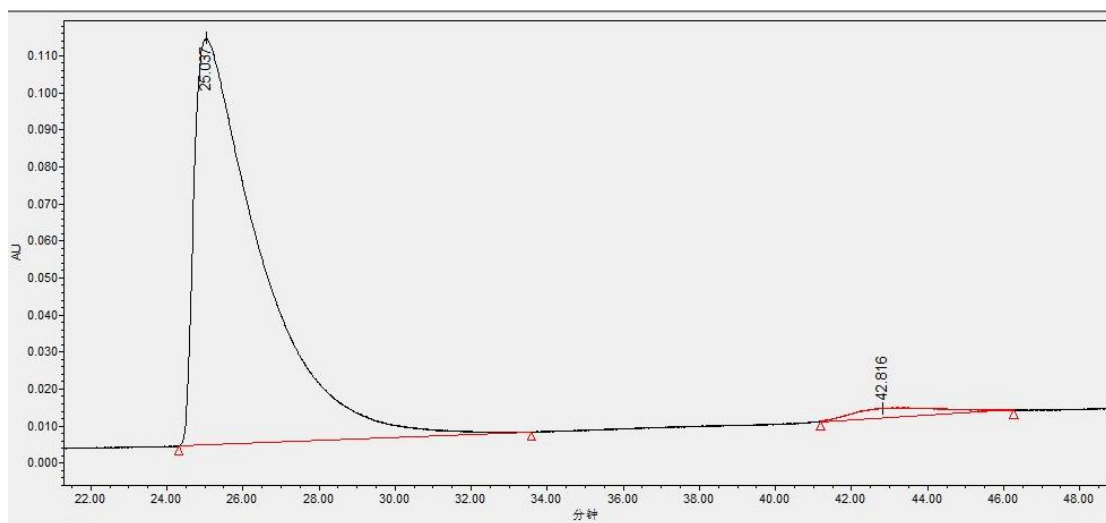
	Time/min	Area	Height	Area%
1	33.680	7094678	55297	98.14
2	40.114	134680	1569	1.86



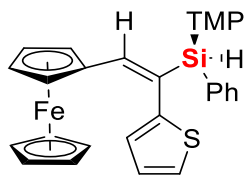
91



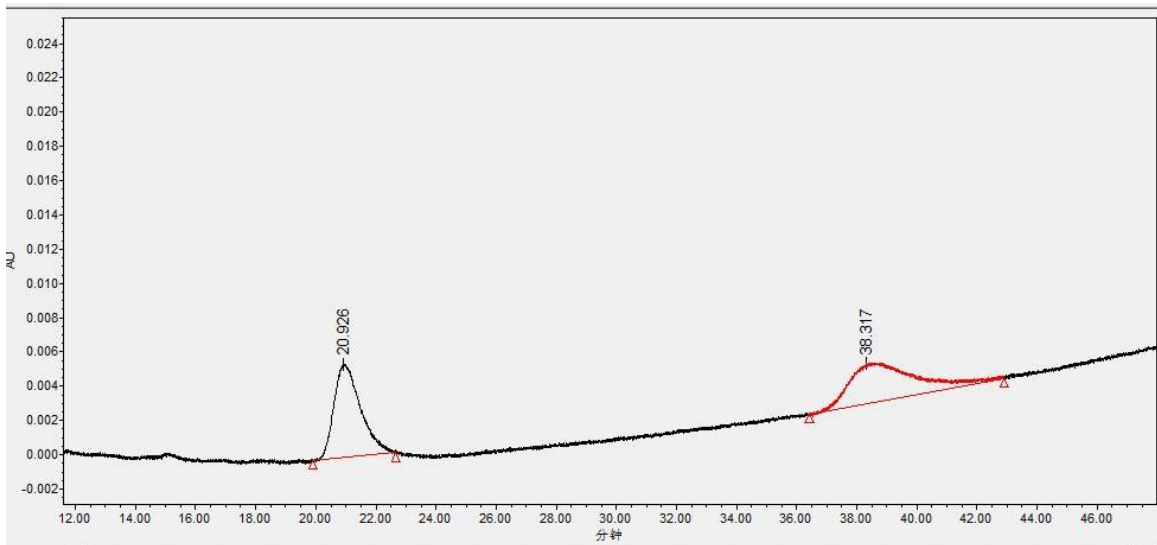
	Time/min	Area	Height	Area%
1	23.227	833675	10923	50.28
2	40.411	824391	5068	49.72



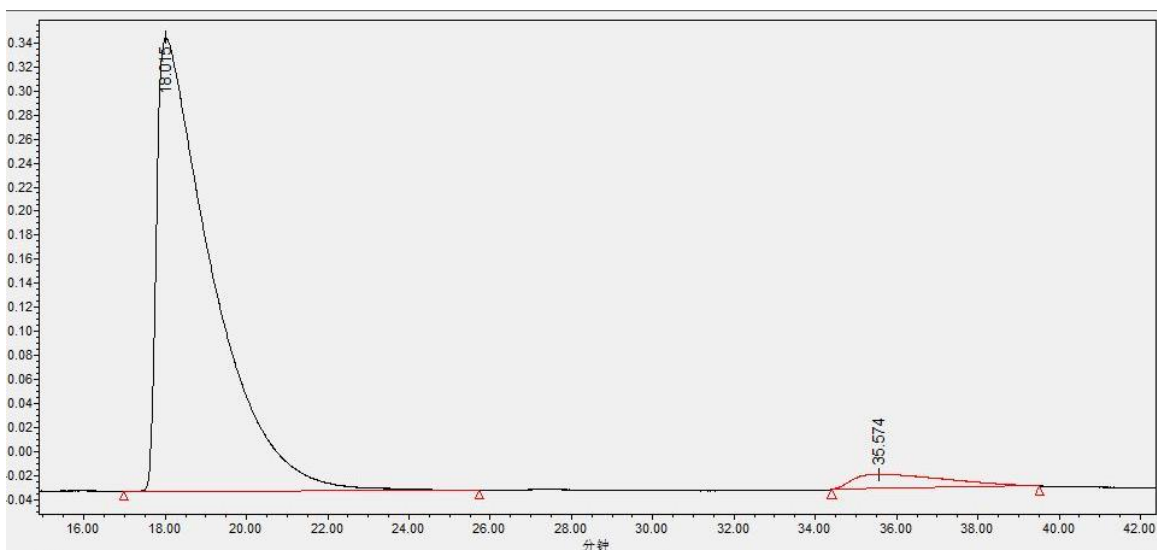
	Time/min	Area	Height	Area%
1	25.037	13313507	109808	97.01
2	42.816	409713	2609	2.99



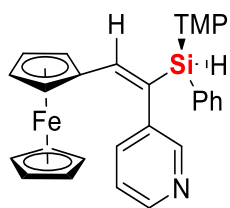
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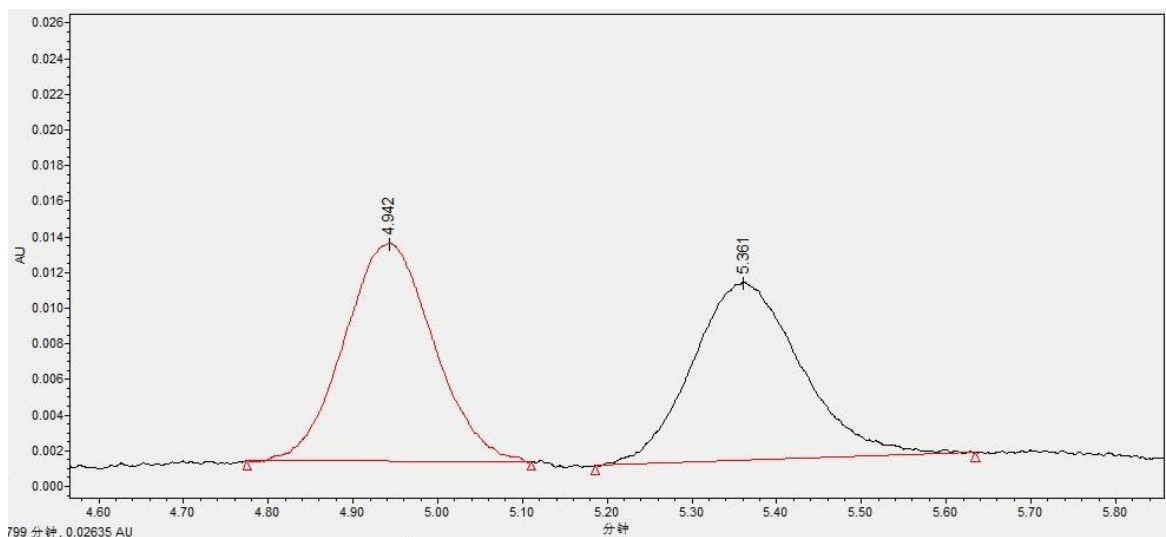
	Time/min	Area	Height	Area%
1	20.926	334691	5474	50.34
2	38.317	330129	2345	49.66



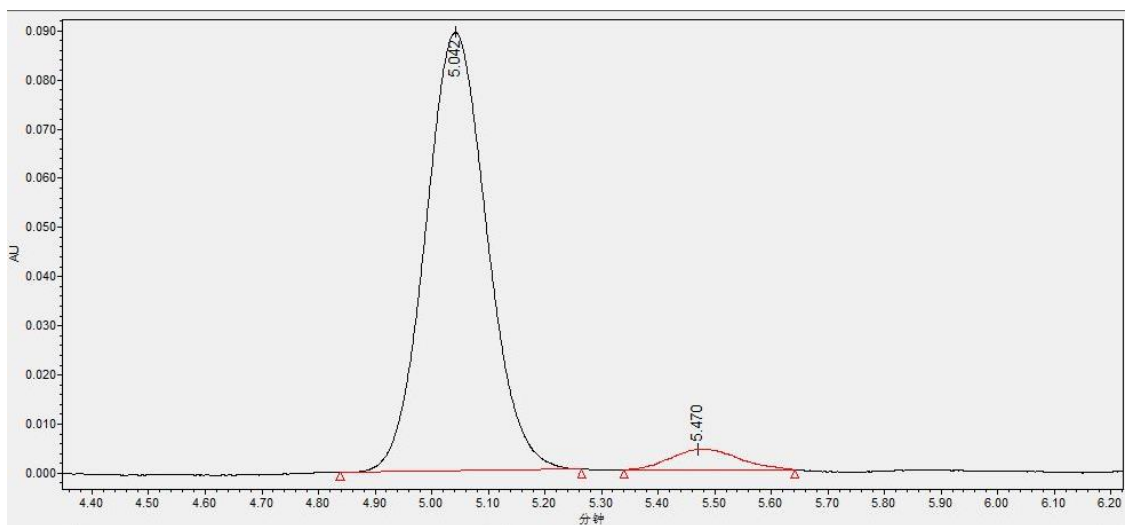
	Time/min	Area	Height	Area%
1	18.015	35604274	377271	94.87
2	35.574	1925584	12115	5.13



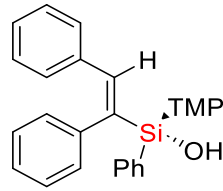
9n



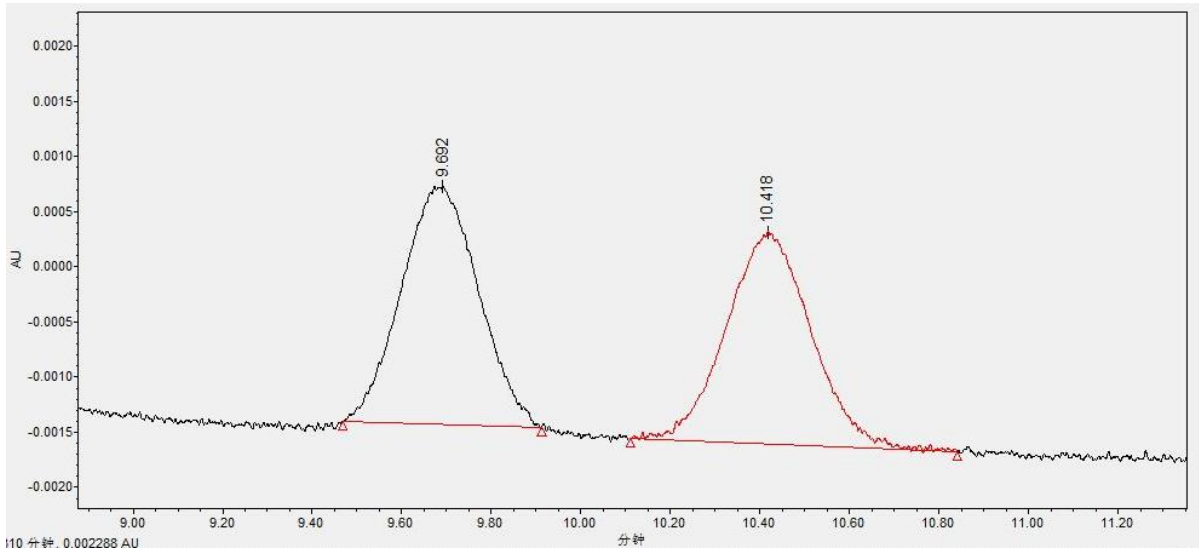
	Time/min	Area	Height	Area%
1	4.942	89095	12210	50.07
2	5.361	88857	9996	49.93



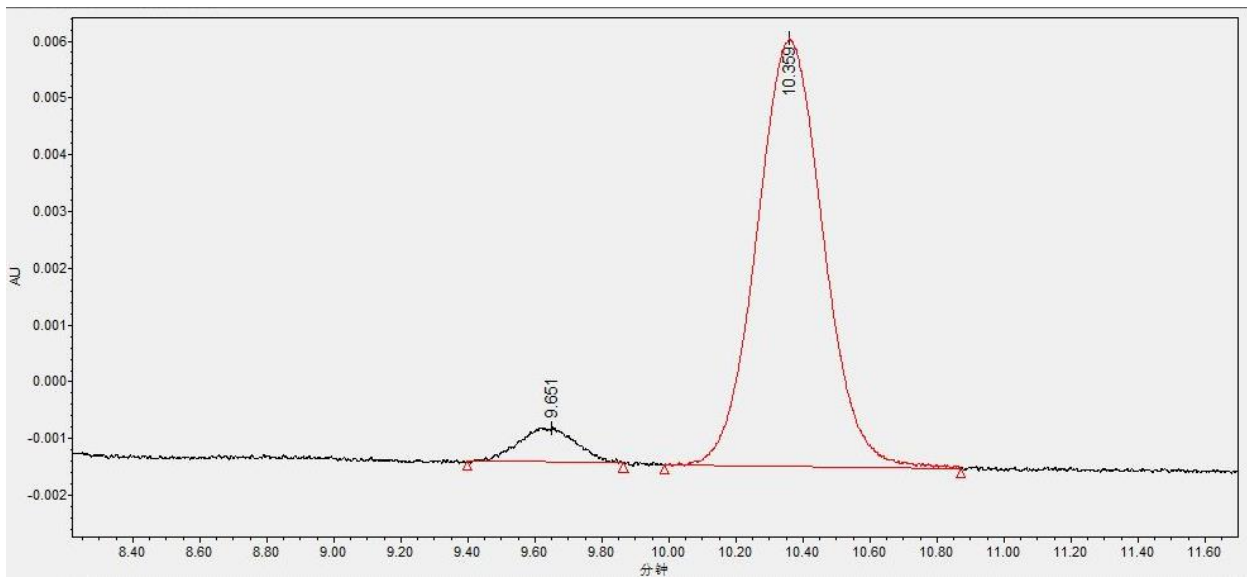
	Time/min	Area	Height	Area%
1	5.042	670829	89156	95.00
2	5.470	35305	4206	5.00



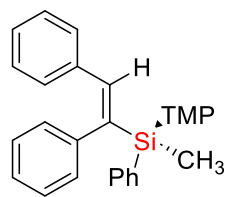
3aa



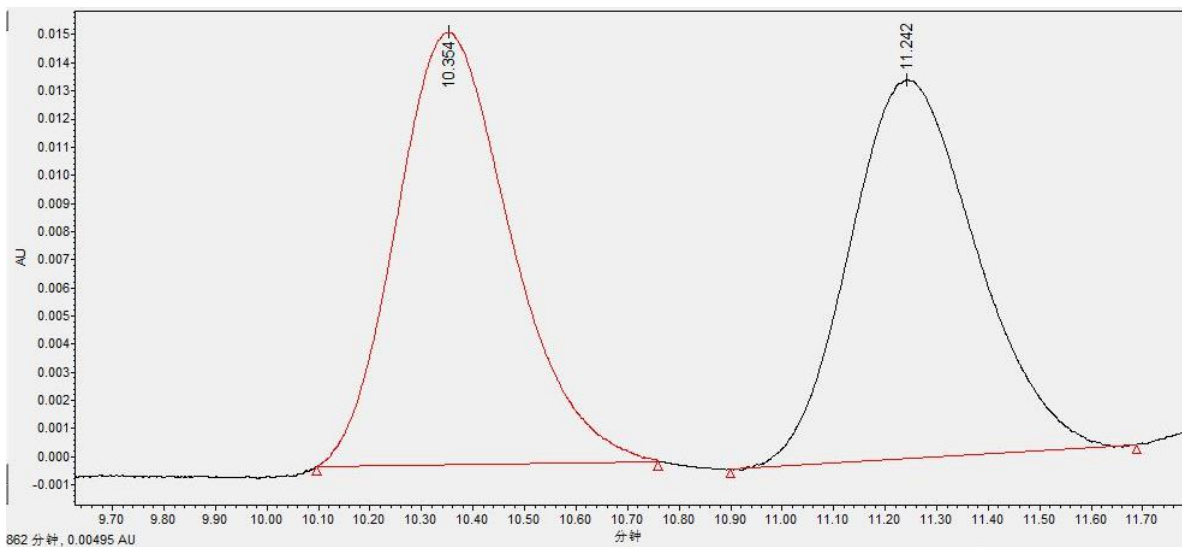
	Time/min	Area	Height	Area%
1	9.692	25935	2162	50.59
2	10.418	25331	1929	49.41



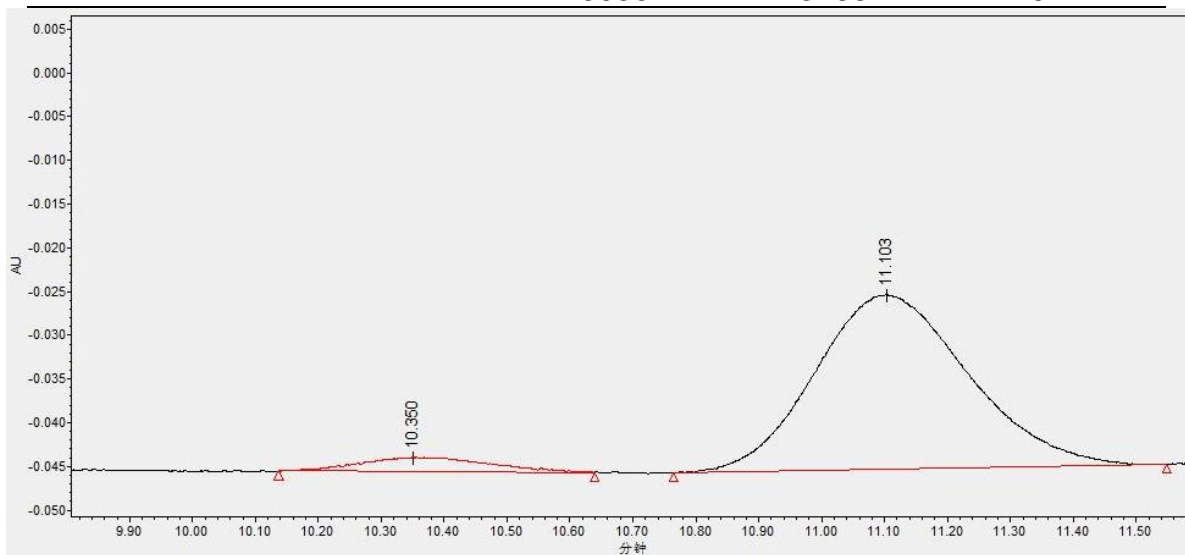
	Time/min	Area	Height	Area%
1	9.651	7171	609	6.49
2	10.359	103351	7537	93.51



3ab



	Time/min	Area	Height	Area%
1	10.354	236385	15343	50.79
2	11.242	229035	13468	49.21



	Time/min	Area	Height	Area%
1	10.350	23133	1627	6.44
2	11.103	336217	19914	93.56