Supporting Information for:

Organocatalytic Enantioselective Allylic Etherification of Morita-Baylis-Hillman Carbonates and Silanols

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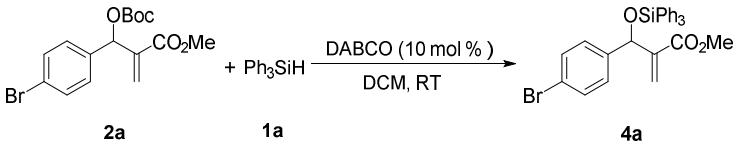
Contents

1. Control experiments [a]

Table S1

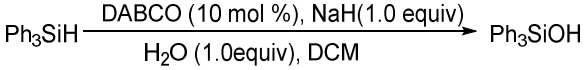


2a **1a** **4a**

entry	base	additive	atmosphere	time	yield (%)[b]
1	---	---	air	7 h	trace
2	NaH	---	air	7 h	18
3	NaH	---	N_2	7 h	20
4	NaH	---	O_2	7 h	15
5[c]	NaH	H_2O	air	1 h	40
6[d]	NaH	H_2O	air	2 h	50
7	NaOH	---	air	2 h	<5

[a] Unless otherwise noted, the reaction conditions were: **2a** (0.1 mmol), **1a** (2.5 equiv), DABCO (10 mol %), base (1.0 equiv), DCM (1.0 mL) at room temperature. [b] Isolated yield. [c] NaH (1.0 equiv) and H_2O (1.0 equiv) were used. [d] NaH (2.0 equiv) and H_2O (2.0 equiv) were used.

Scheme S1 Ph_3SiH transformed into Ph_3SiOH



1a **3a**

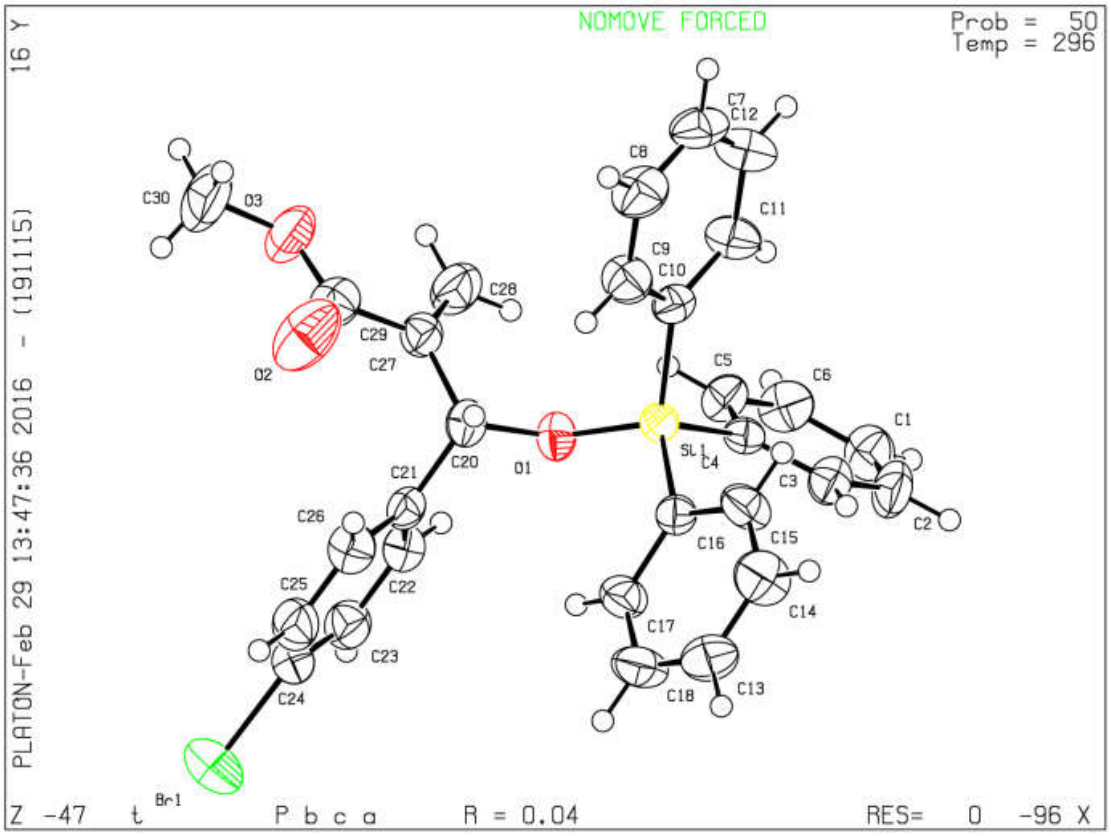
7% yield

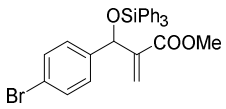
When Ph_3SiH was used as the nucleophile, the reaction was conducted under the catalyst of DABCO (10 mol %) in DCM at room temperature, affording trace of the product **4a** (Table S1, entry 1). Then we conducted a series of control experiments to investigate the oxygen sources of **4a**. First, in order to remove the influence of O_2, the reaction was conducted under the atmosphere of air, N_2, or O_2 (Table S1, entries 2-4), giving the product of **4a** in 15-20% yields. So oxygen gas wasn't the oxygen source of the product **4a**. Then H_2O was added as the additive, which shorted the reaction time and improved the yield from 18% to 40% (Table S1, entries 2 vs 5). When NaOH was used to replace NaH and H_2O, trace of **4a** was obtained (Table S1, entry 7). Thus, H_2O

is the oxygen source of the product **4a**. We assumed that Ph_3SiH transformed into Ph_3SiOH. In order to verify our conjecture, the substrate of **2a** was removed, and 7% yield of Ph_3SiOH (**3a**) was obtained (Scheme S1).

2. X-Ray crystallographic analysis of **4a**.

A single crystal of **4a** was obtained by recrystallized from hexane and DCM. The thermal ellipsoid was drawn at the 50% probability level.





4a

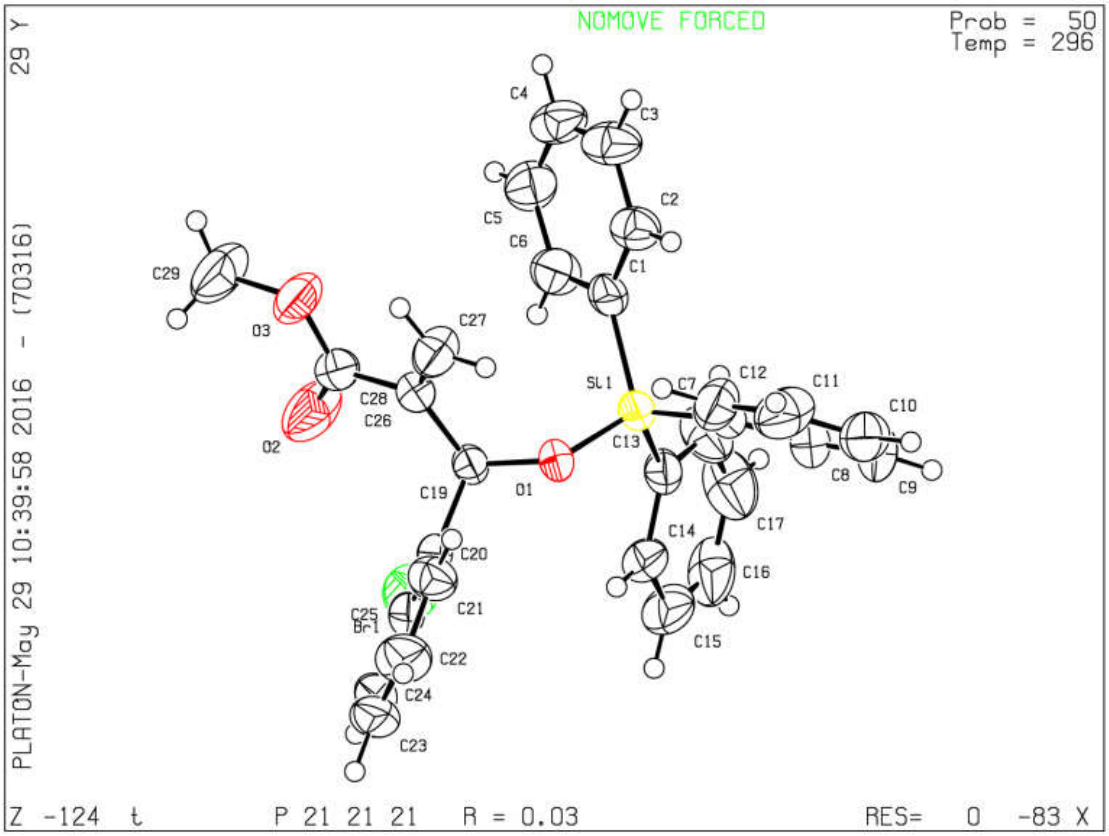
Figure S1 X-ray crystal structure **4a**.

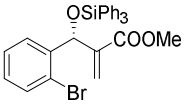
Table S2 Crystal data and structure refinement for **4a**.

Formula	$C_{29}H_{25}BrO_3Si$
Formula weight	529.49
Temperature (K)	296(2)
Crystal system	orthorhombic
Wavelength (Å)	0.71073
Space group	P b a c
a (Å)	19.289(4)
b (Å)	11.081(2)
c (Å)	23.943(5)
α (º)	90.00
β (º)	90.00
γ (º)	90.00
Volume ($Å^3$)	5117.7(18)
Z	8
Calculate density (Mg/m^3)	1.374
Absorption coefficient (mm^{-1})	1.681
F000	2176.0
Crystal size (mm)	0.35×0.25×0.15
Theta range for data collection (º)	2.11 to 25.00
Reflections collected	38908
Independent reflections	4500
Data/restraints/parameters	4500/0/307
Goodness-of-fit on F^2	1.085
Final R indices	$R_1 = 0.0403$ $wR_2 = 0.1003$
R indices (all data)	$R_1 = 0.0749$ $wR_2 = 0.1350$

3. X-Ray crystallographic analysis of **7ka**.

A single crystal of **7ka** was obtained by recrystallized from hexane and DCM. The thermal ellipsoid was drawn at the 50% probability level.





7ka

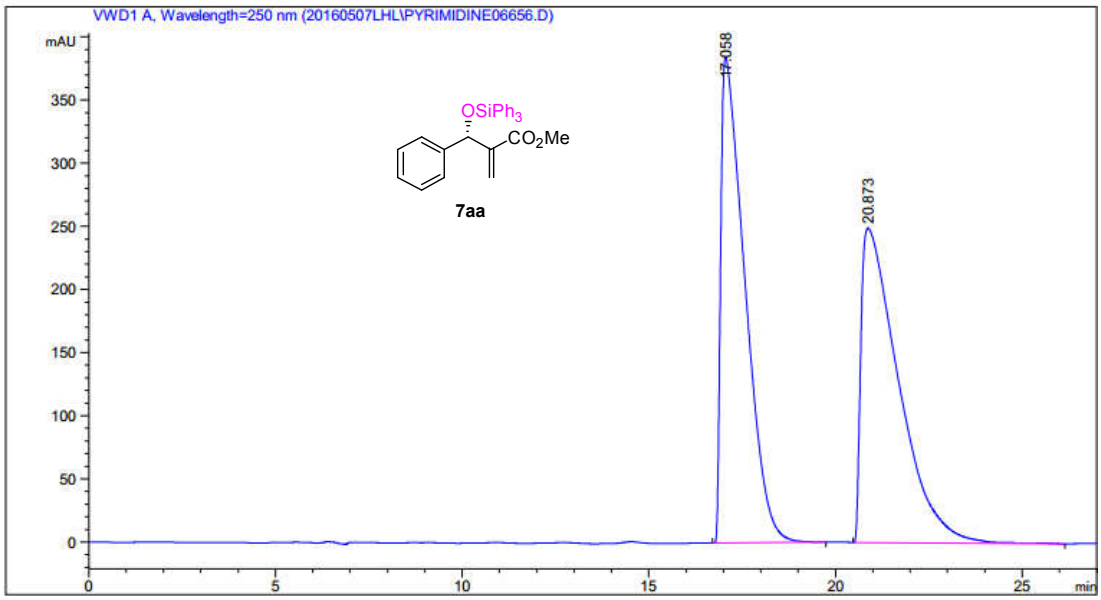
Figure S2 X-ray crystal structure **7ka**.

Table S3 Crystal data and structure refinement for **7ka**.

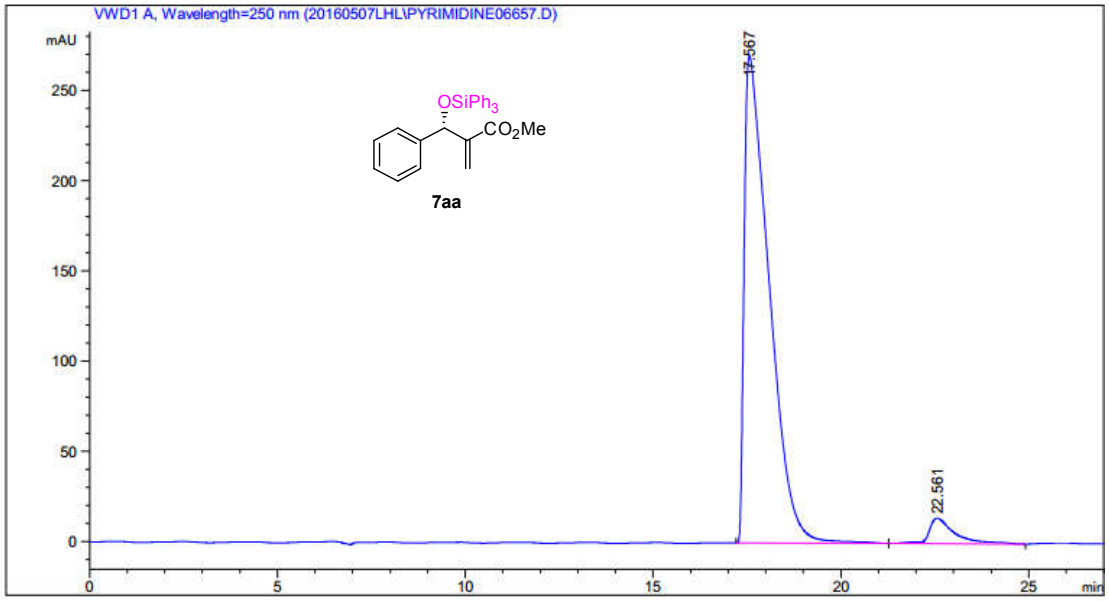
Formula	$C_{29}H_{25}BrO_3Si$
Formula weight	529.49
Temperature	296(2)
Crystal system	orthorhombic
Wavelength (Å)	0.71073
a(Å)	10.545(2)
b(Å)	12.431(3)

c(Å)	19.567(4)
α(°)	90.00
β(°)	90.00
γ(°)	90.00
Volume ($Å^3$)	2565.1(9)
Z	4
Calculate density(Mg/m^3)	1.371
Absorption coefficient(mm^{-1})	1.677
F000	1088
Crystal size	0.35×0.30×0.25
Theta range for data collection	1.94-25.00
Reflections collected	18234
Independent reflections	4510
Data/restraints/parameters	4510/0/307
Goodness-of-fit on F^2	1.033
Final R indices	$R_1 = 0.0311$ $wR_2 = 0.0645$
R indices (all data)	$R_1 = 0.0446$ $wR_2 = 0.0695$
Flack parameter	0.006(7)
Space group	$P2_12_12_1$

4. Copies of HPLC spectra for racemic and enantiomerically enriched compounds.



Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	17.058	BB	0.7012	1.72167e4	384.74515	49.7032
2	20.873	BB	1.0276	1.74223e4	248.94460	50.2968



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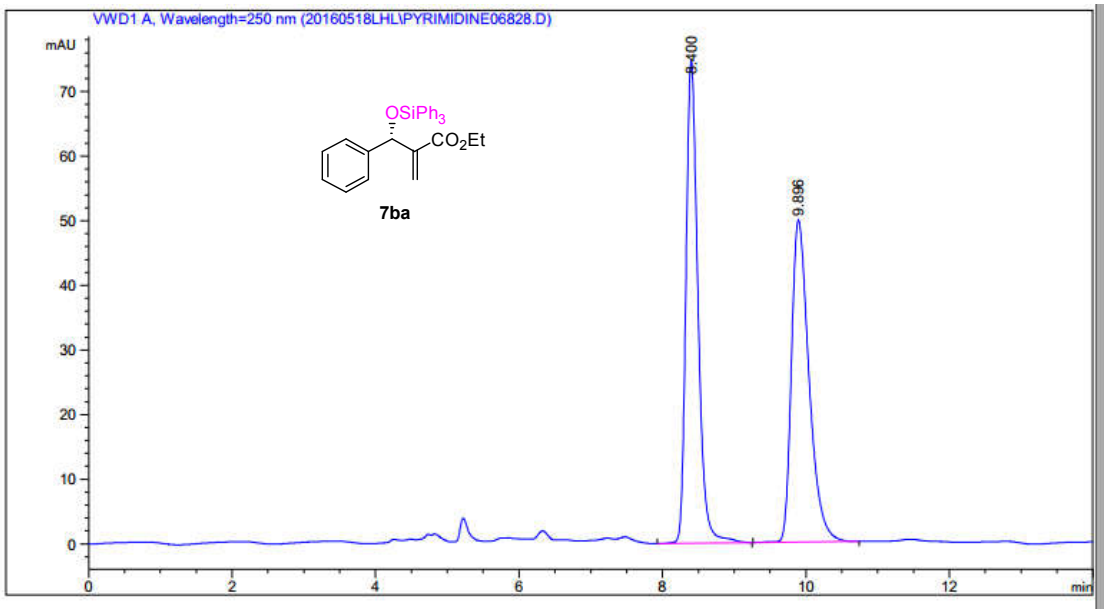
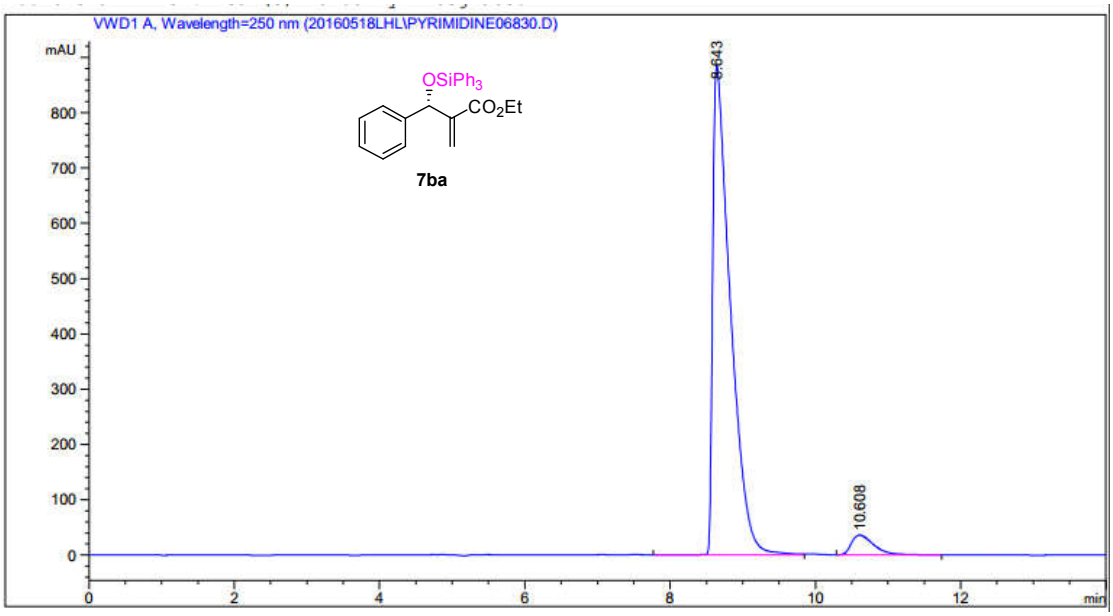
1	17.567	BB	0.6386	1.23881e4	270.02661	94.8558
2	22.561	BB	0.6735	671.83466	14.14605	5.1442



Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	8.400	BB	0.1773	865.31866	74.73851	50.5115
2	9.896	BB	0.2626	847.79456	49.83195	49.4885



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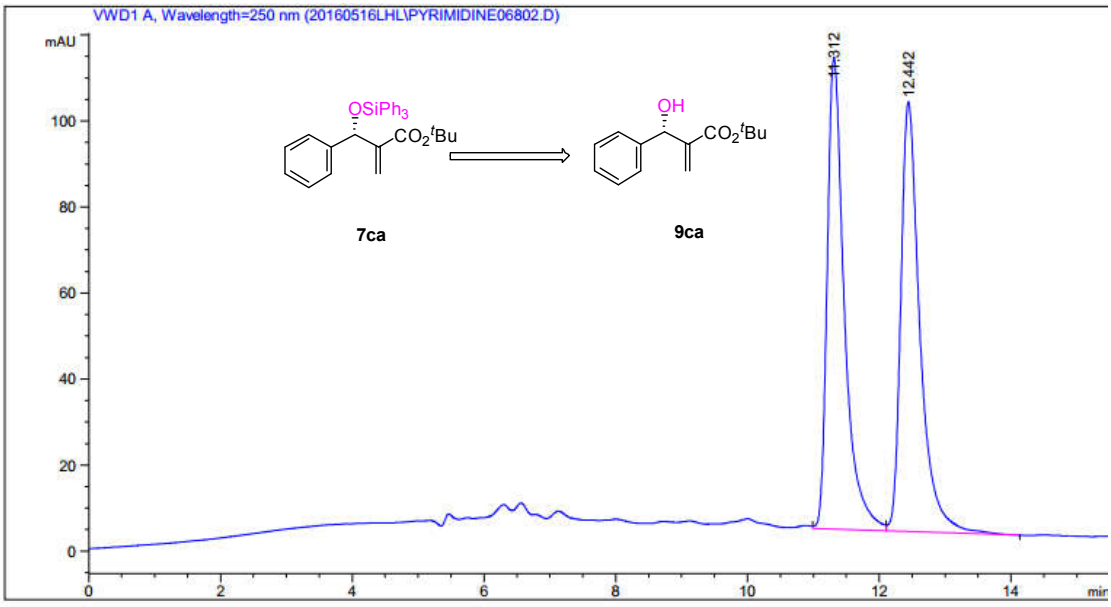
2	10.608	VB	0.3391	798.46753	36.16347	5.0704



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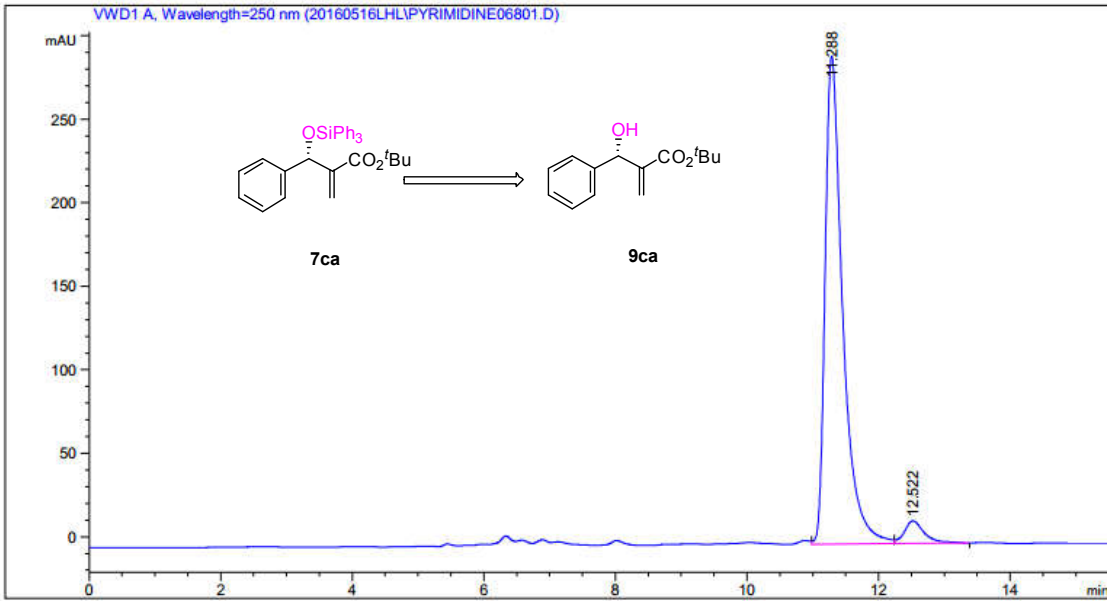
1	11.312	VV	0.2694	1985.70154	109.69669	49.6342
2	12.442	VB	0.3008	2014.97046	99.89694	50.3658



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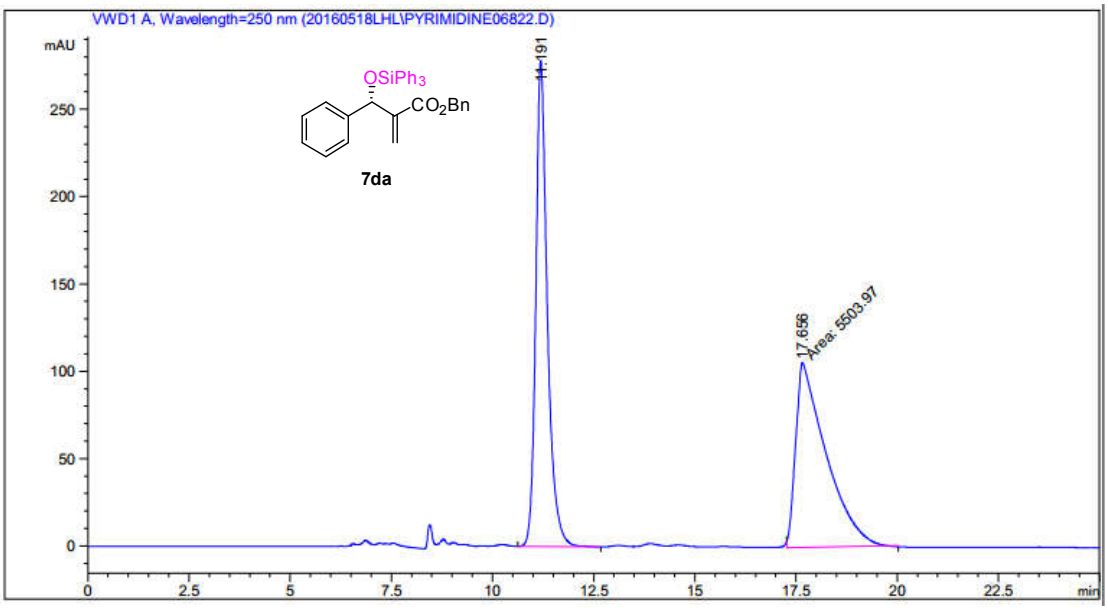
1	11.288	VV	0.2780	5429.13672	292.04575	95.0004
2	12.522	VB	0.3122	285.72122	13.51119	4.9996



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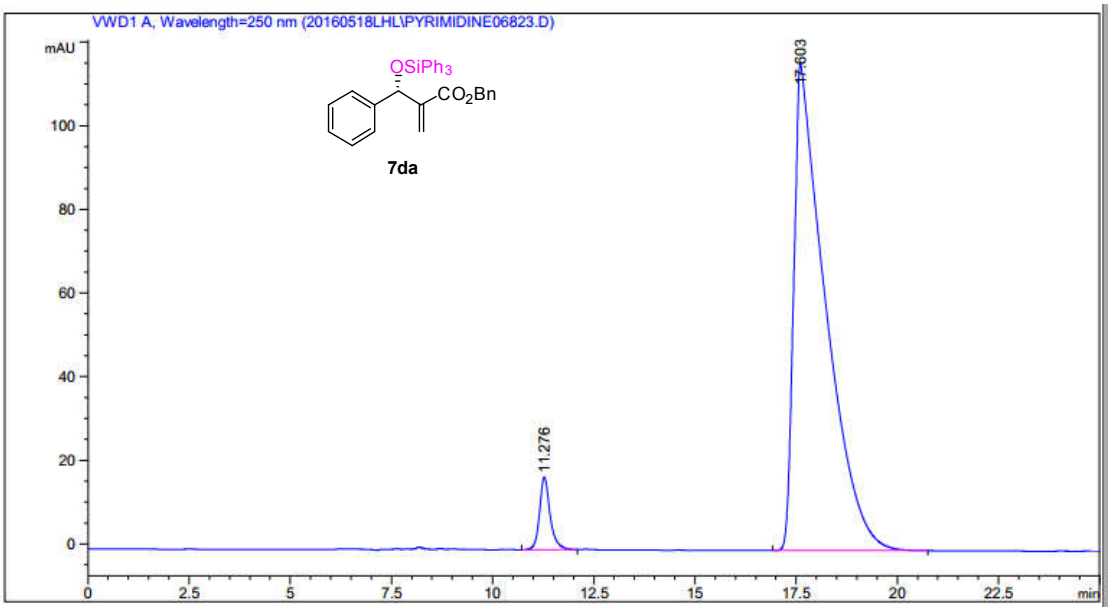
1	11.191	BB	0.2923	5511.33057	278.12177	50.0334
2	17.656	MM	0.8674	5503.97363	105.75124	49.9666



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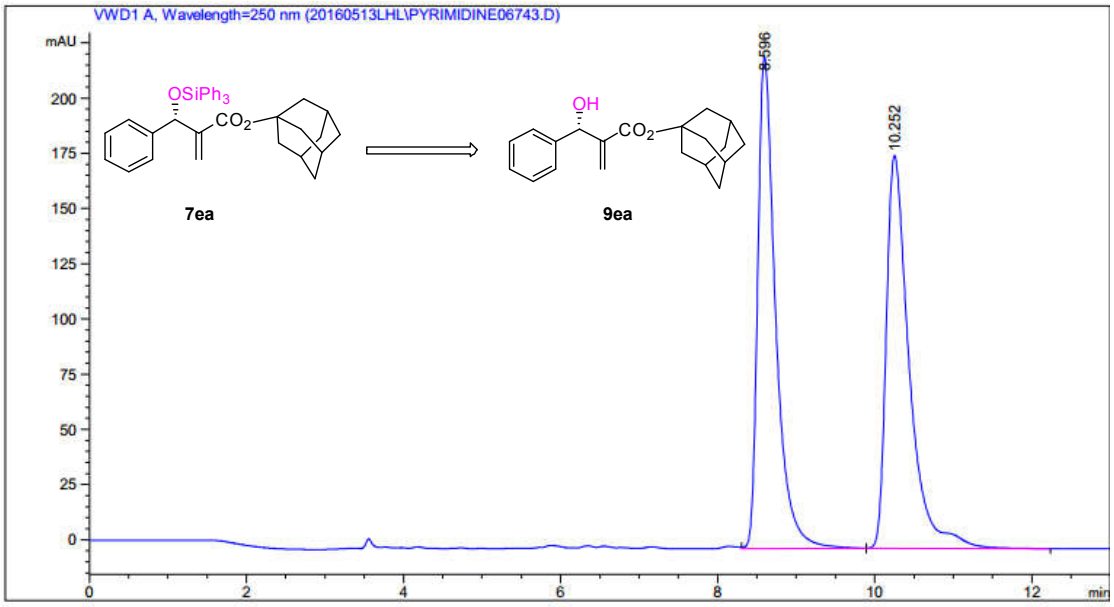
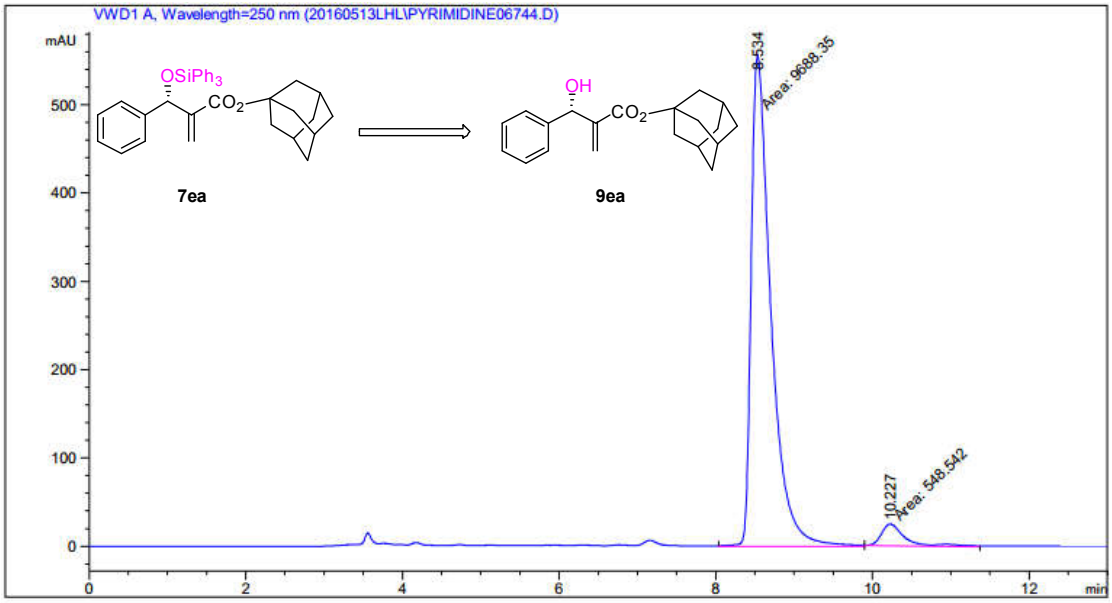
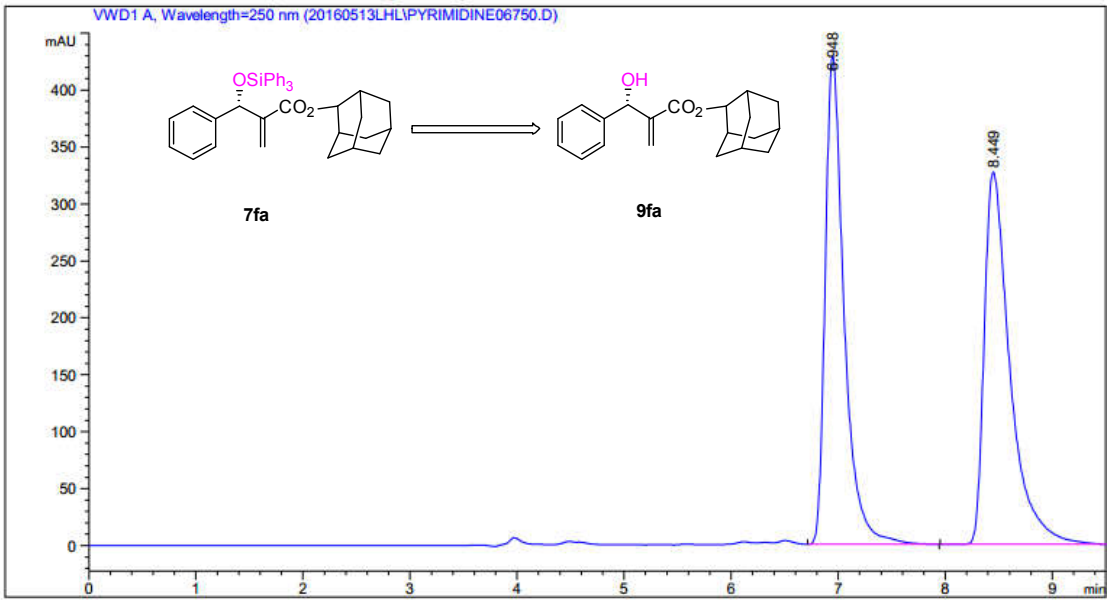
1	11.276	BB	0.2703	316.47406	17.41009	4.7755
2	17.603	BB	0.7010	6310.51611	116.39234	95.2245



Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	8.596	VB	0.2370	3523.81348	222.69391	50.0297
2	10.252	MM	0.3314	3519.63232	176.99677	49.9703



Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	8.534	MM	0.2909	9688.35059	555.16437	94.6415
2	10.227	MM	0.3677	548.54187	24.86687	5.3585



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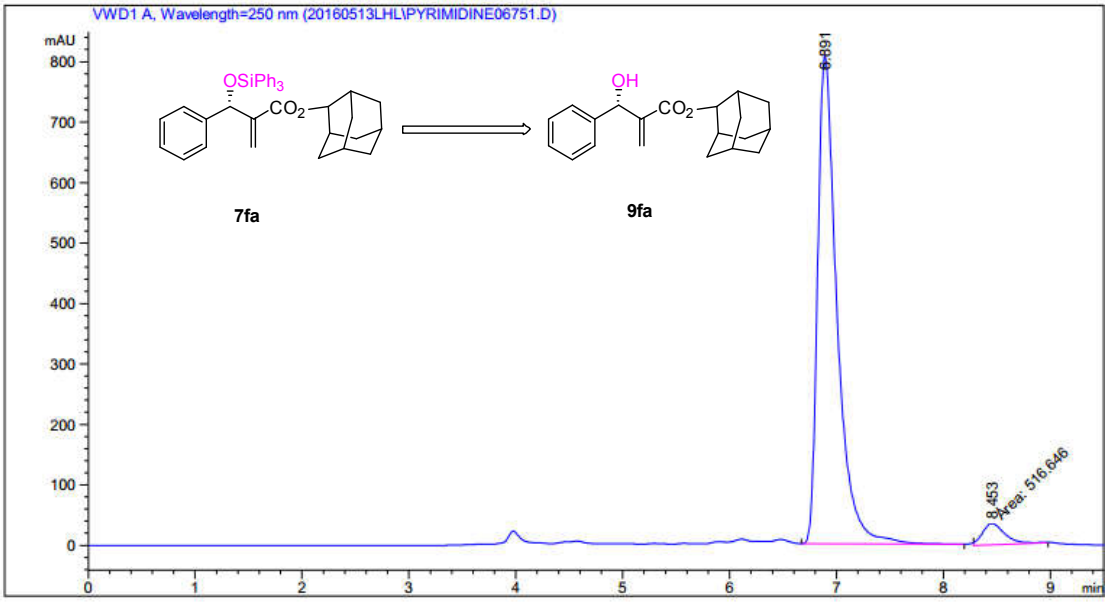
1	6.948	BB	0.1842	5264.30029	428.18542	49.0924
2	8.449	BB	0.2486	5458.93994	327.10687	50.9076



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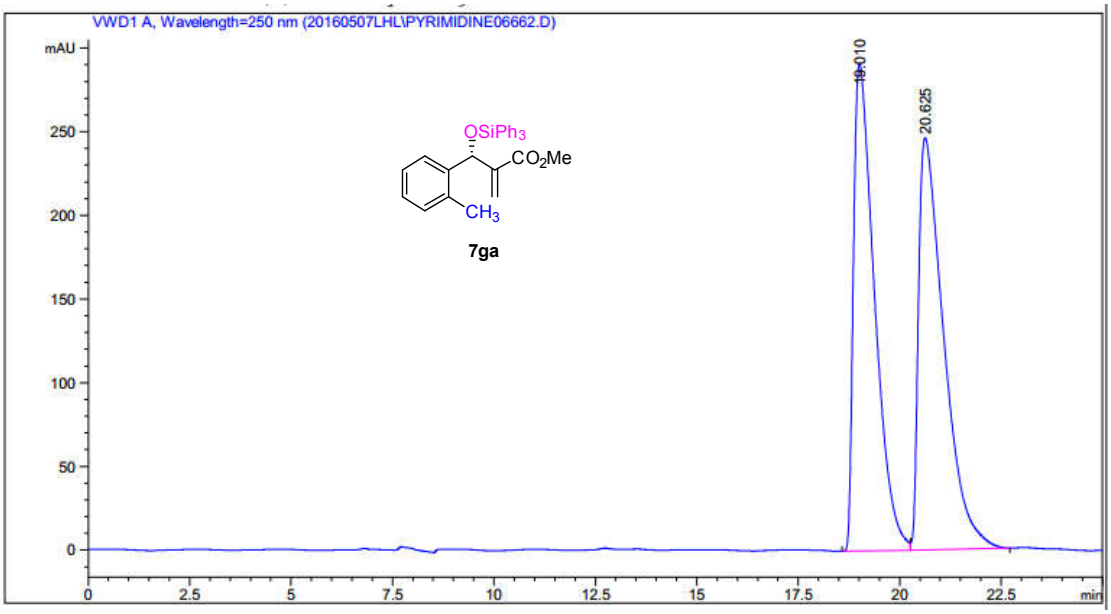
1	6.891	BB	0.1878	1.01709e4	806.84467	95.1659
2	8.453	MM	0.2433	516.64551	35.38952	4.8341



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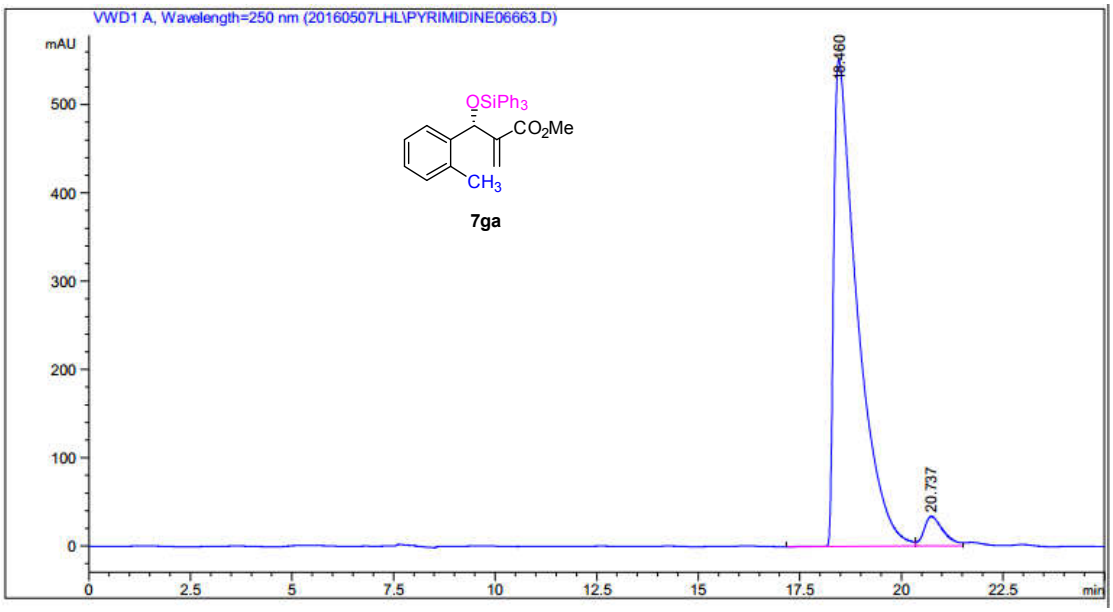
1	19.010	BV	0.5408	1.04839e4	291.22107	49.5974
2	20.625	VB	0.6516	1.06541e4	246.36525	50.4026



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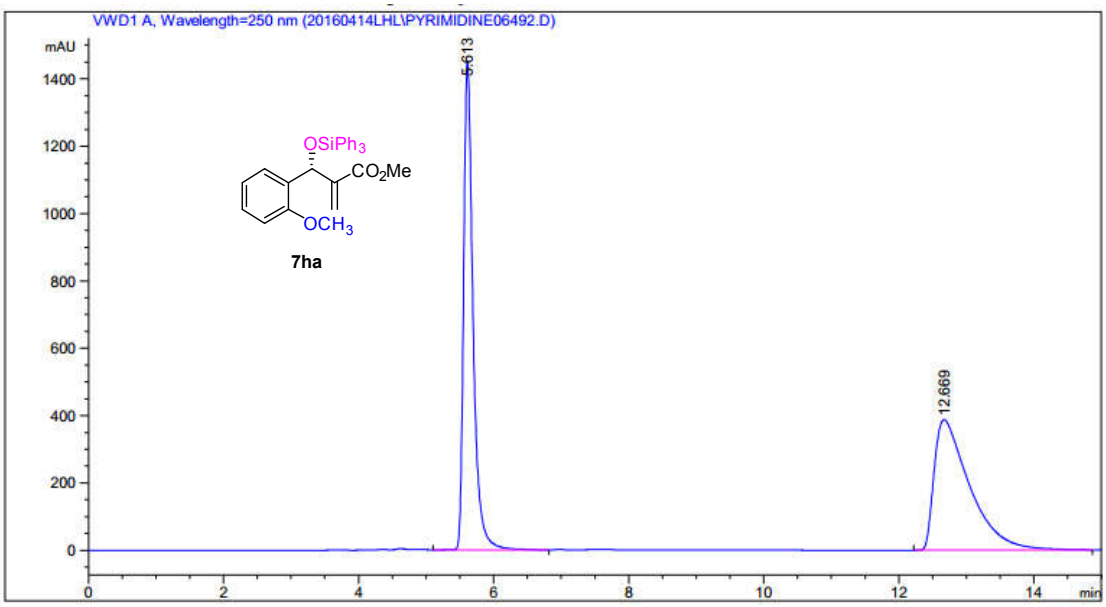
1	18.460	BV	0.5711	2.21918e4	551.84418	95.2062
2	20.737	VV	0.4952	1117.40186	33.52199	4.7938



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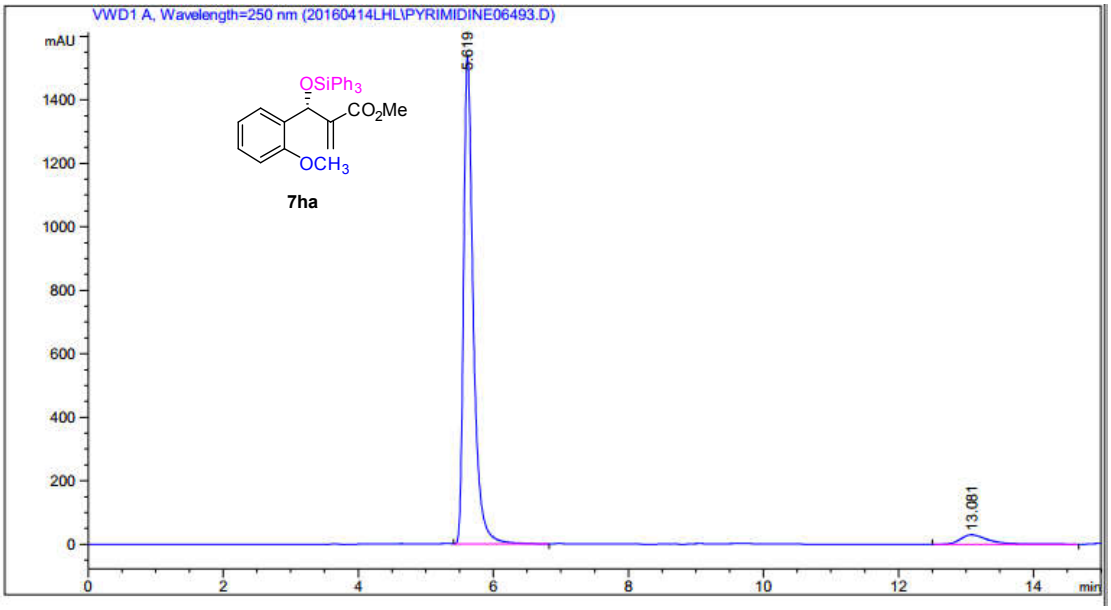
1	5.613	BV	0.1474	1.41571e4	1449.84131	49.8589
2	12.669	BV	0.5480	1.42372e4	387.52374	50.1411



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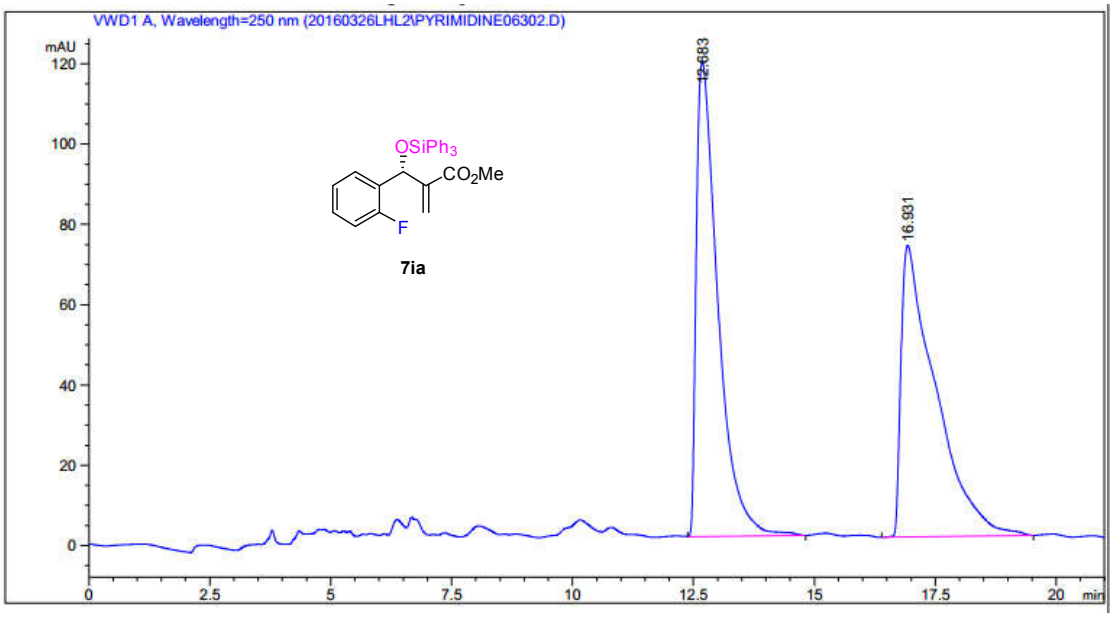
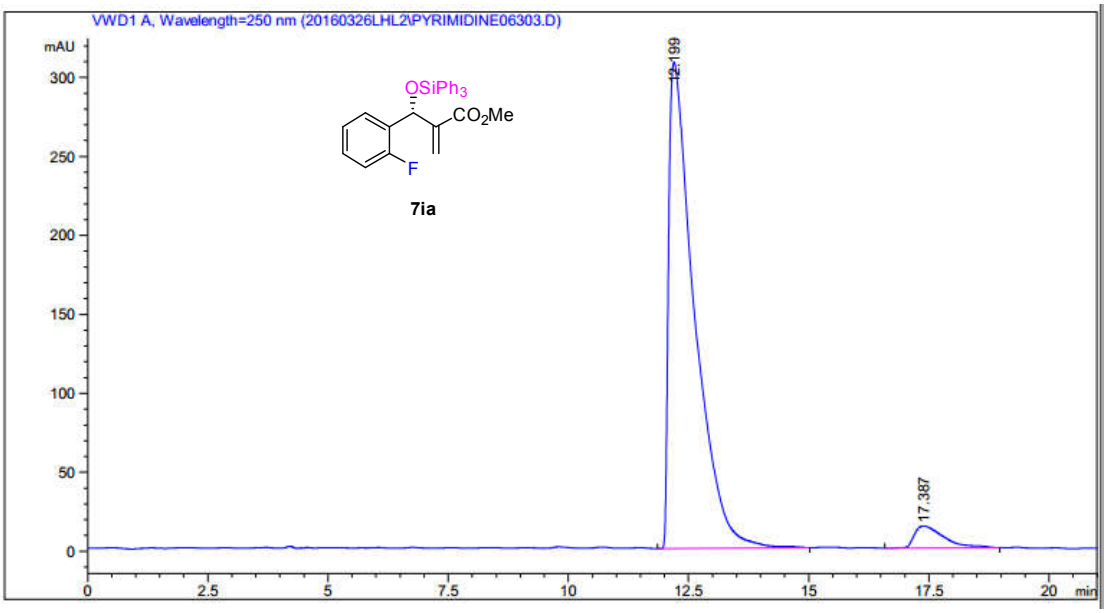
1	5.619	VV	0.1502	1.53688e4	1536.87708	94.8869
2	13.081	BB	0.4273	828.16620	29.43416	5.1131



7ia

Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	12.683	BB	0.4751	3672.64648	118.04672	50.3949
2	16.931	BB	0.6685	3615.08130	72.57122	49.6051



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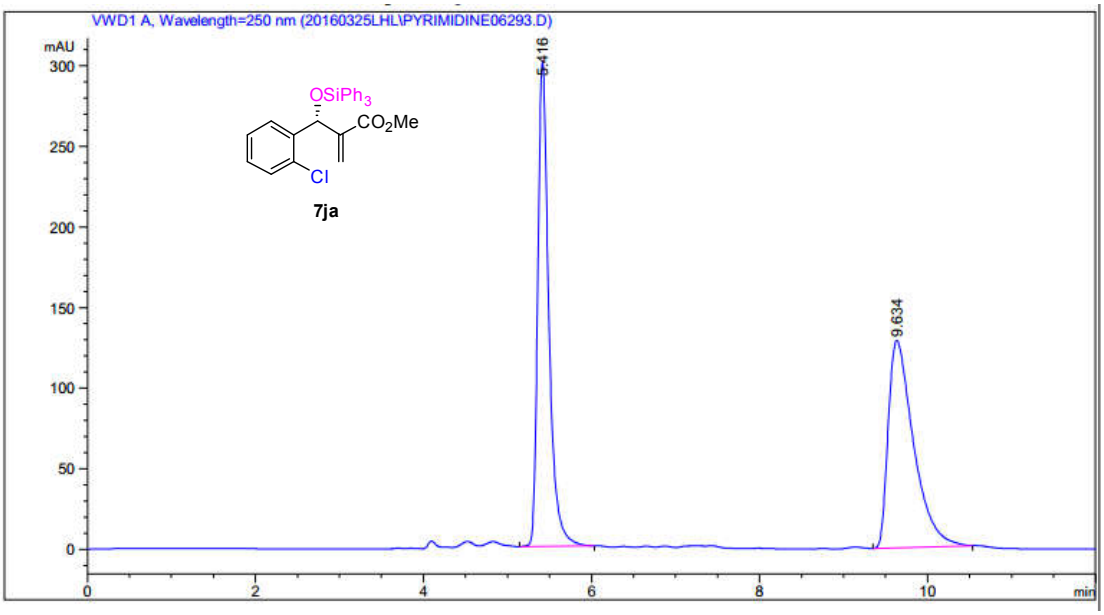
1	12.199	BB	0.5440	1.14761e4	307.90839	95.0238
2	17.387	BB	0.6476	600.98688	13.96851	4.9762



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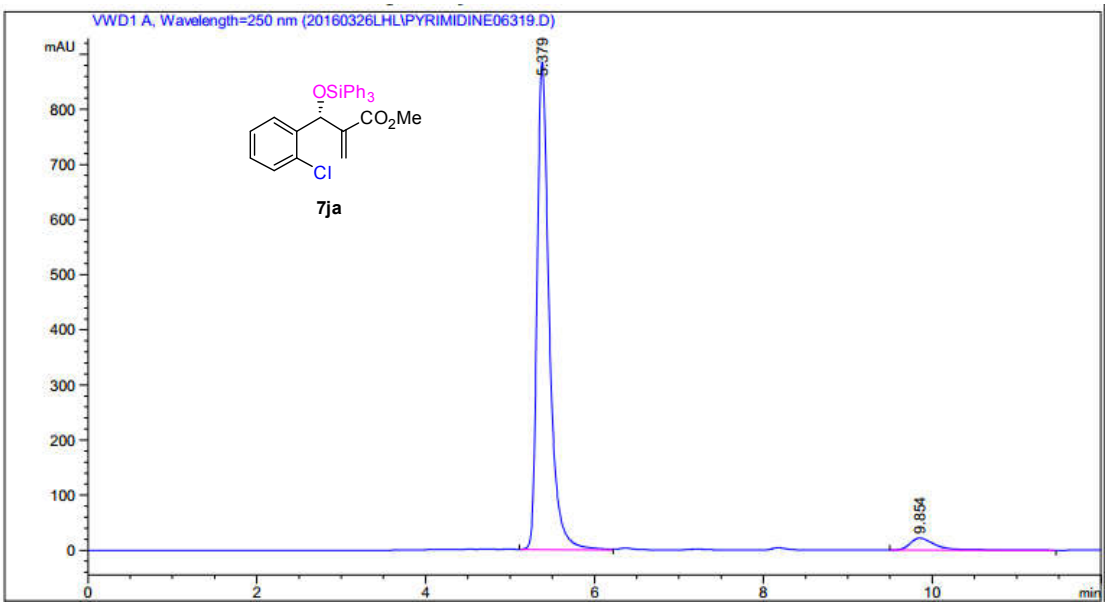
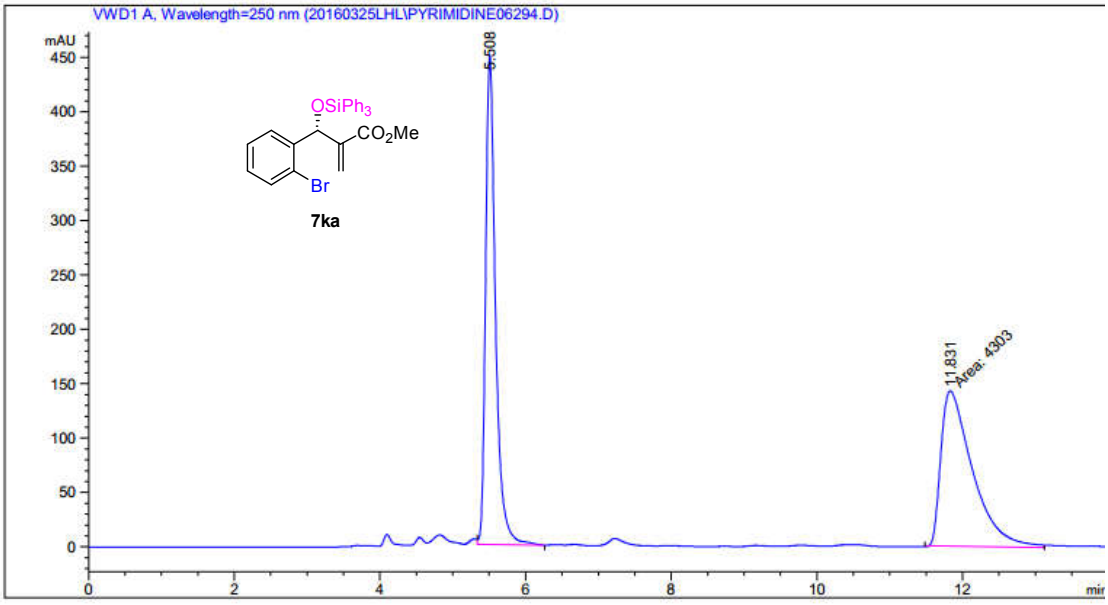
1	5.416	BB	0.1387	2748.16602	300.49734	50.5232
2	9.634	BB	0.3155	2691.24658	128.61560	49.4768



Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	5.379	BV	0.1504	8860.97070	884.61920	95.0280
2	9.854	VB	0.3148	463.61707	22.08590	4.9720



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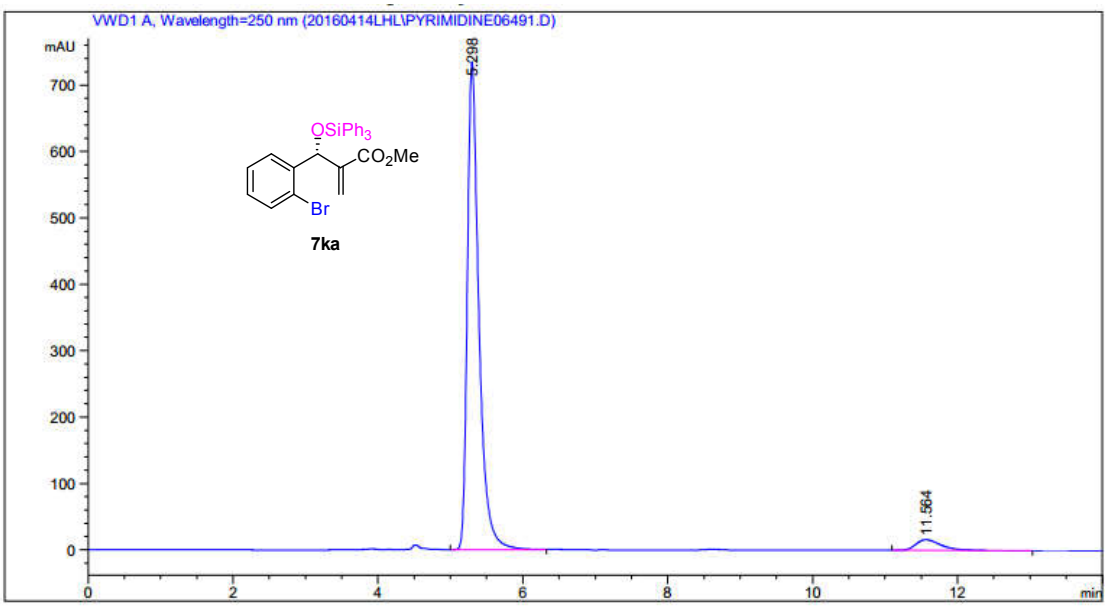
1	5.508	VB	0.1428	4261.39014	448.98669	49.7571
2	11.831	MM	0.5023	4302.99561	142.77823	50.2429



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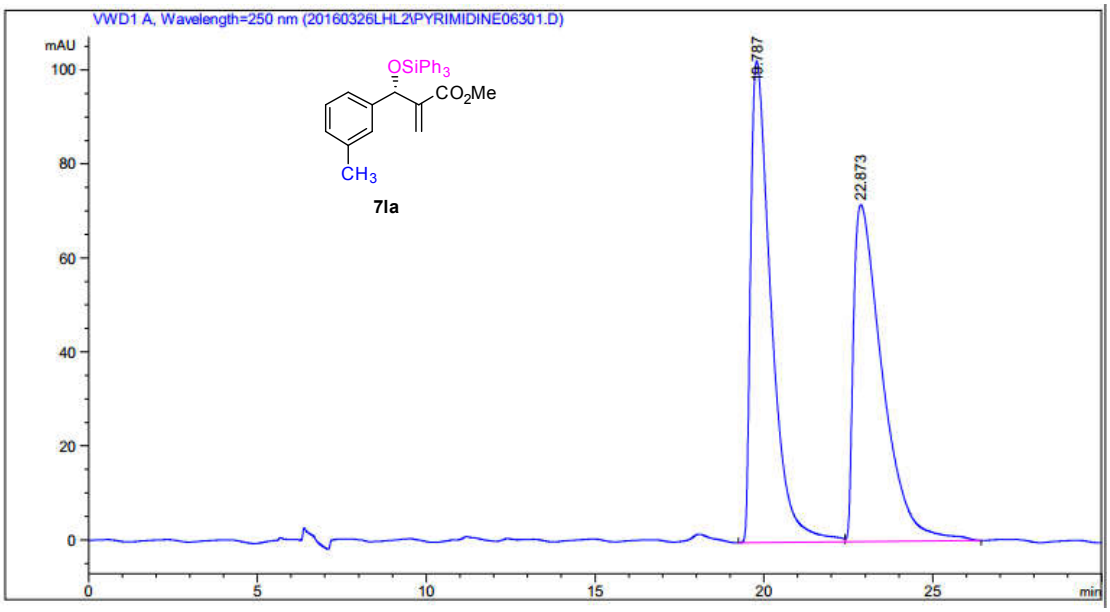
1	5.298	BV	0.1642	8135.57373	734.36688	95.0073
2	11.564	MM	0.4205	427.53326	16.94523	4.9927



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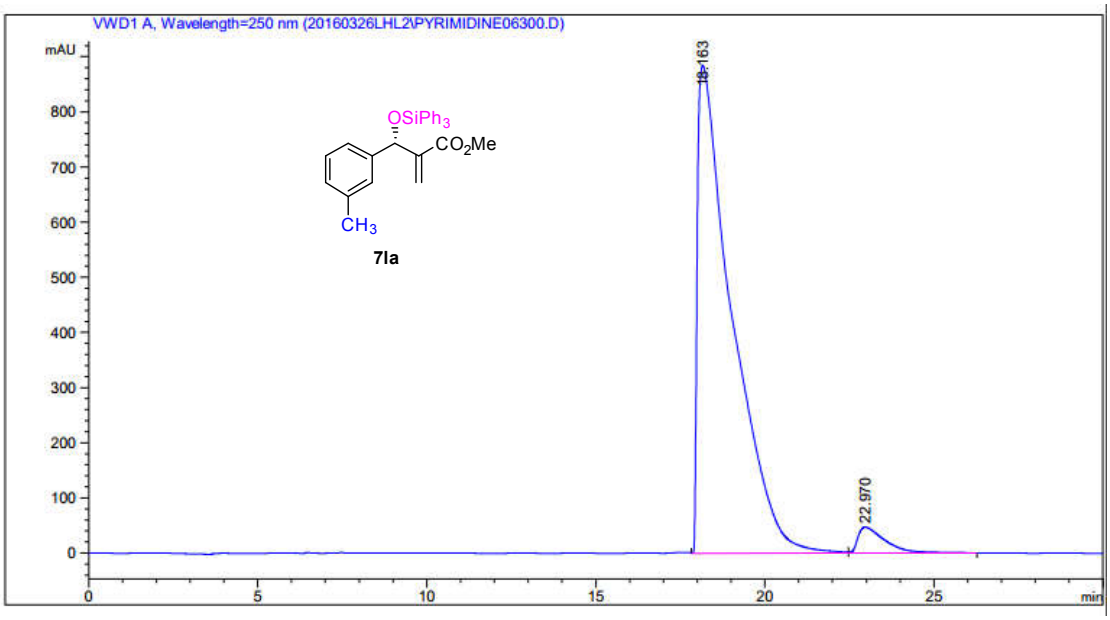
1	19.787	BV	0.6206	4315.40674	102.53961	50.3113
2	22.873	VB	0.8915	4262.00342	71.70576	49.6887



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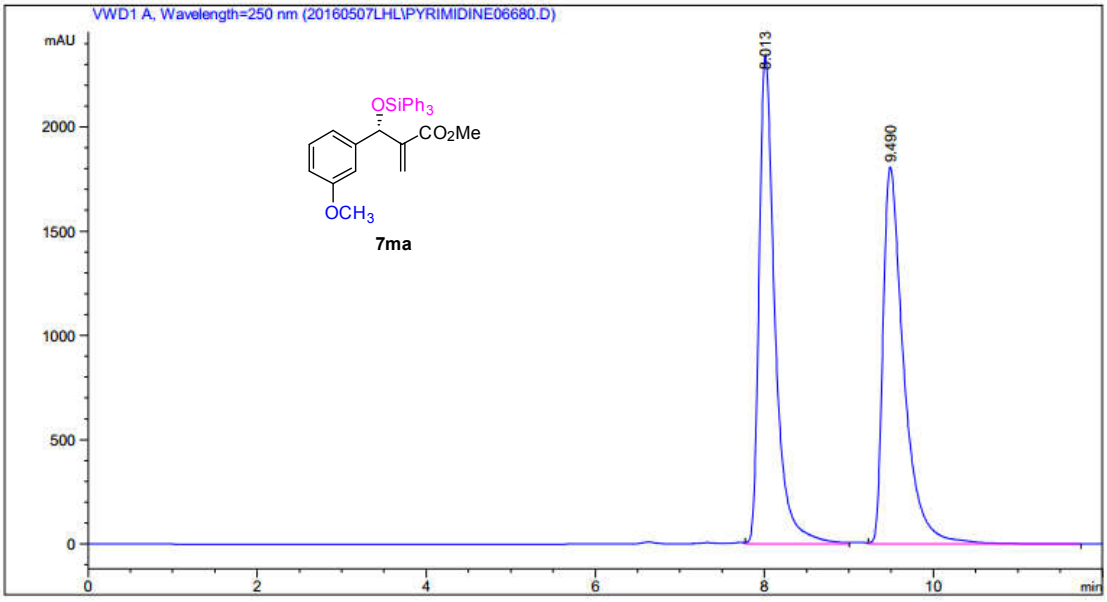
2	22.970	VB	0.8068	2639.28589	46.98482	4.0558



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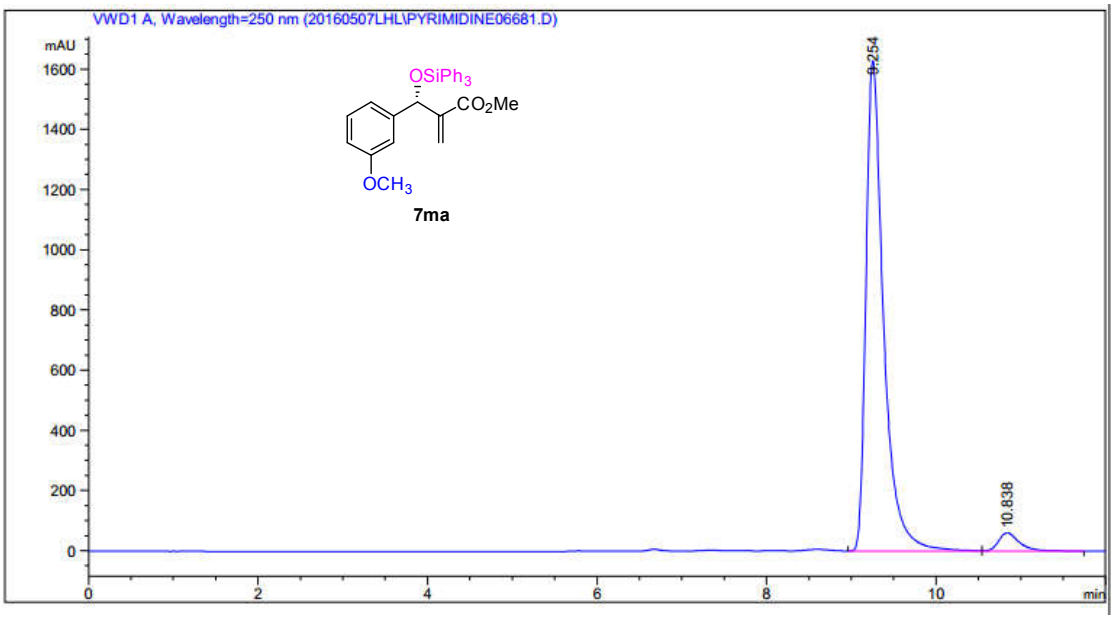
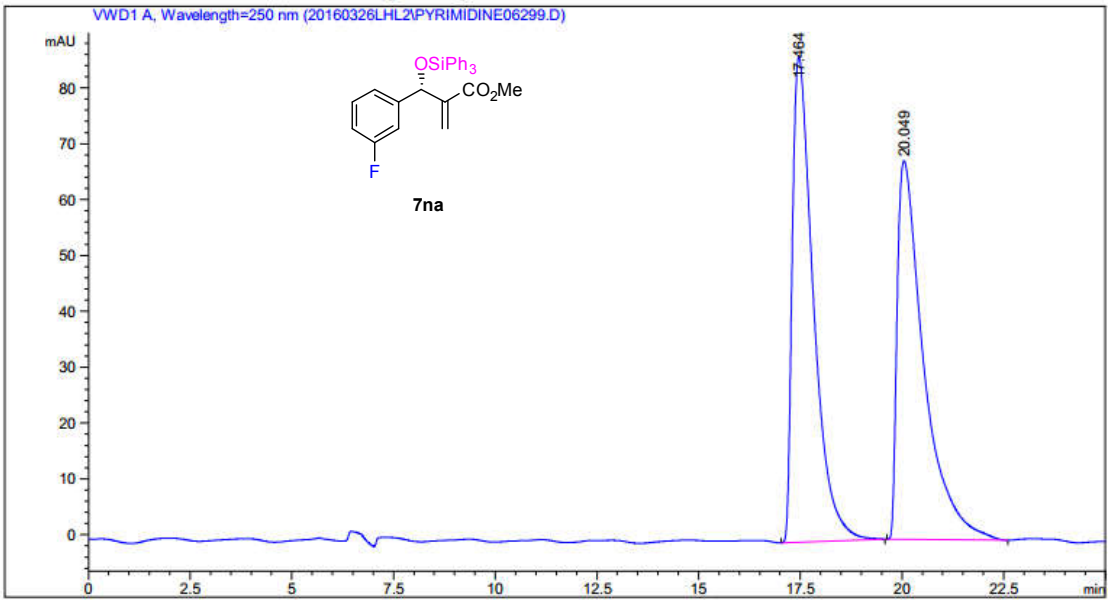
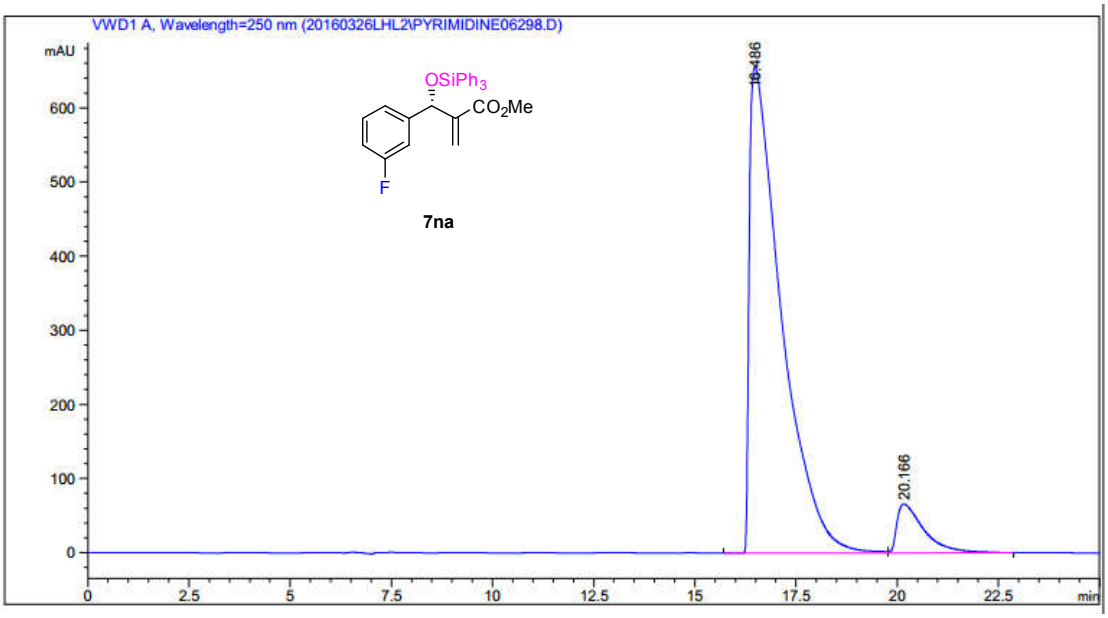
1	8.013	VV	0.1958	3.05074e4	2339.44751	49.7848
2	9.490	VB	0.2541	3.07711e4	1806.46960	50.2152



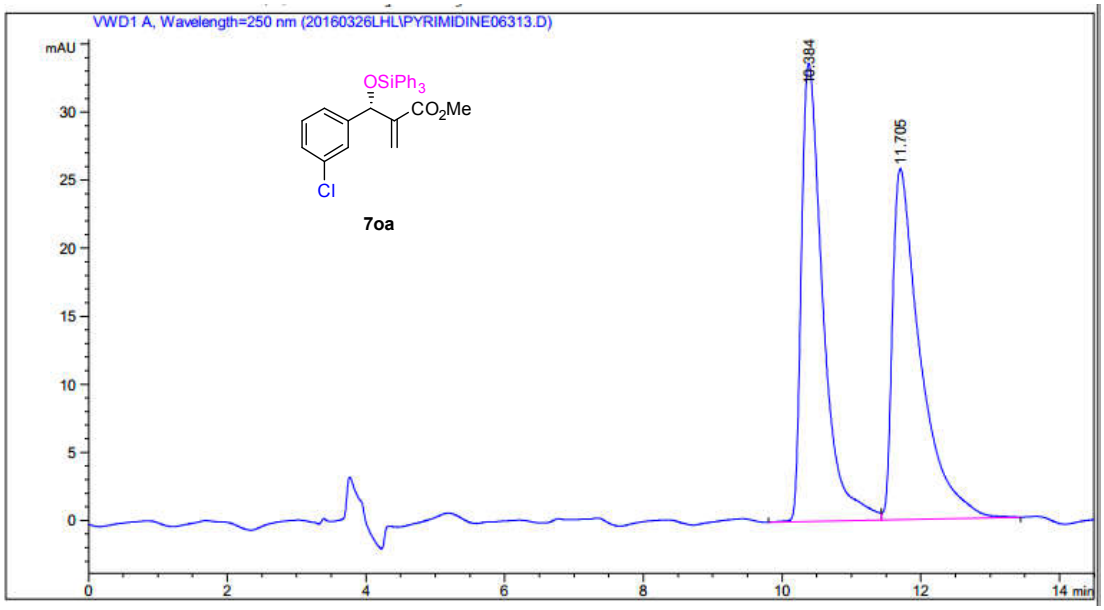
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	9.254	BV	0.2152	2.35246e4	1627.97400	95.7085
2	10.838	VB	0.2583	1054.83569	61.08167	4.2915



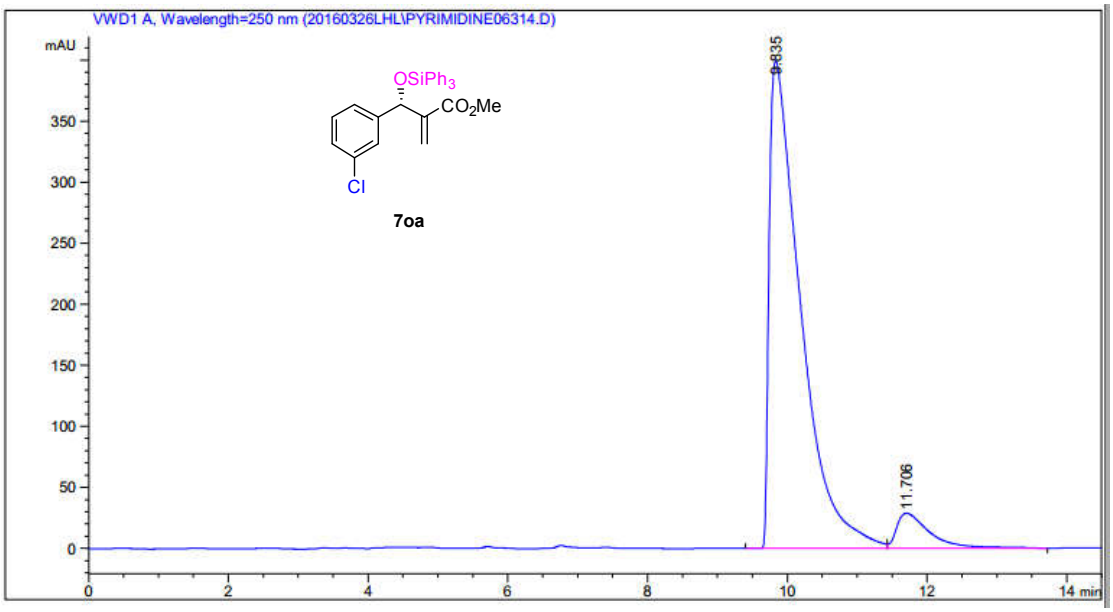
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	17.464	BB	0.5461	3135.74097	86.91165	50.5081
2	20.049	BB	0.6633	3072.65674	67.71957	49.4919



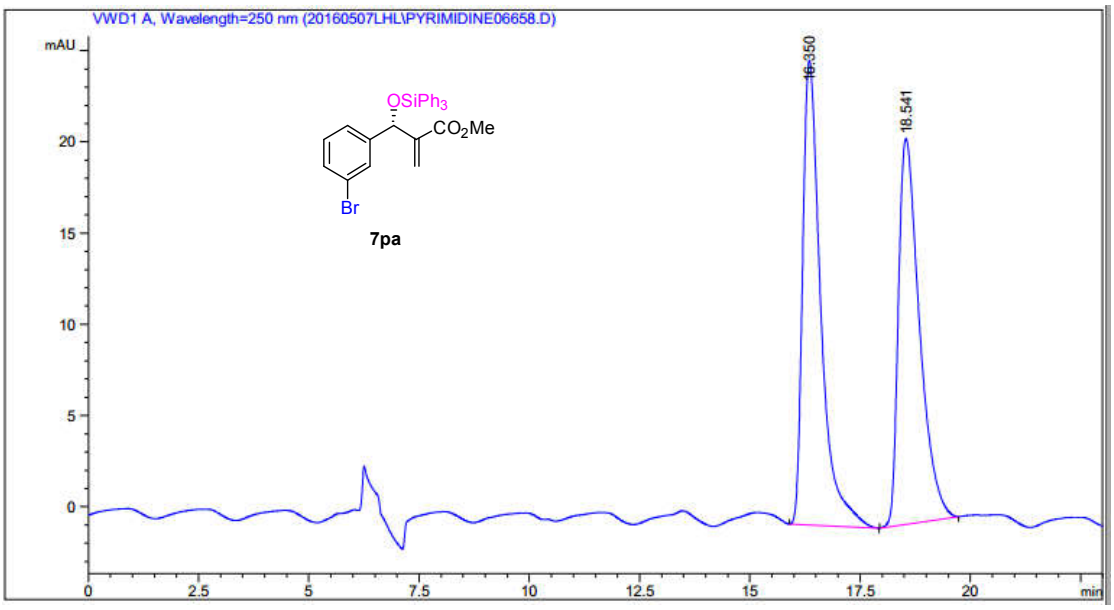
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	16.486	BV	0.7676	3.58875e4	656.25256	92.1124
2	20.166	VB	0.6991	3073.05811	65.99279	7.8876



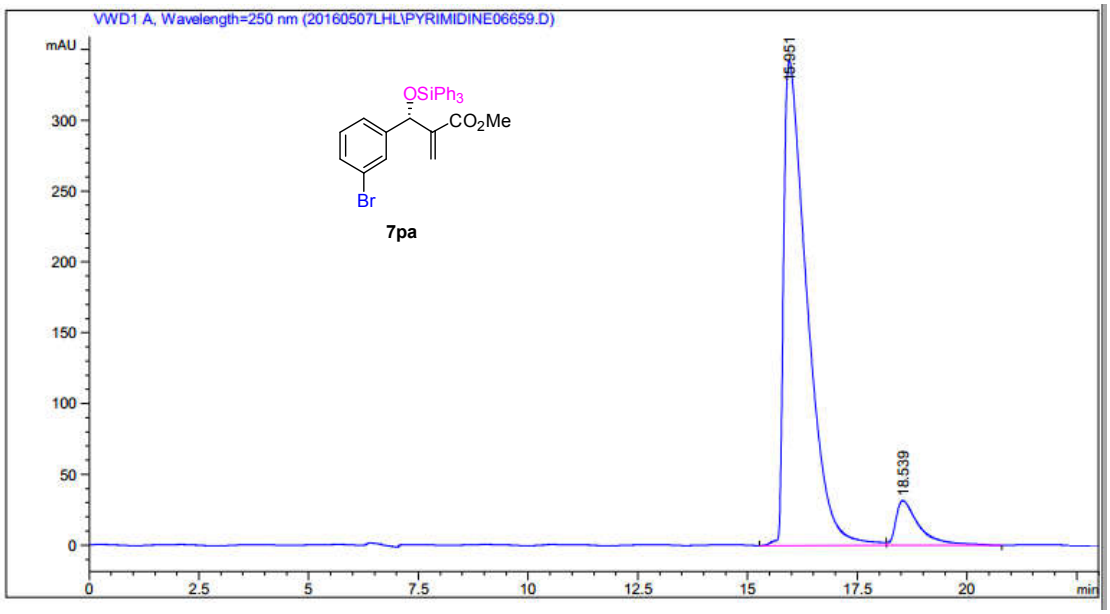
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	10.384	BV	0.3275	726.21338	33.65357	50.2358
2	11.705	VB	0.4027	719.39685	25.77947	49.7642



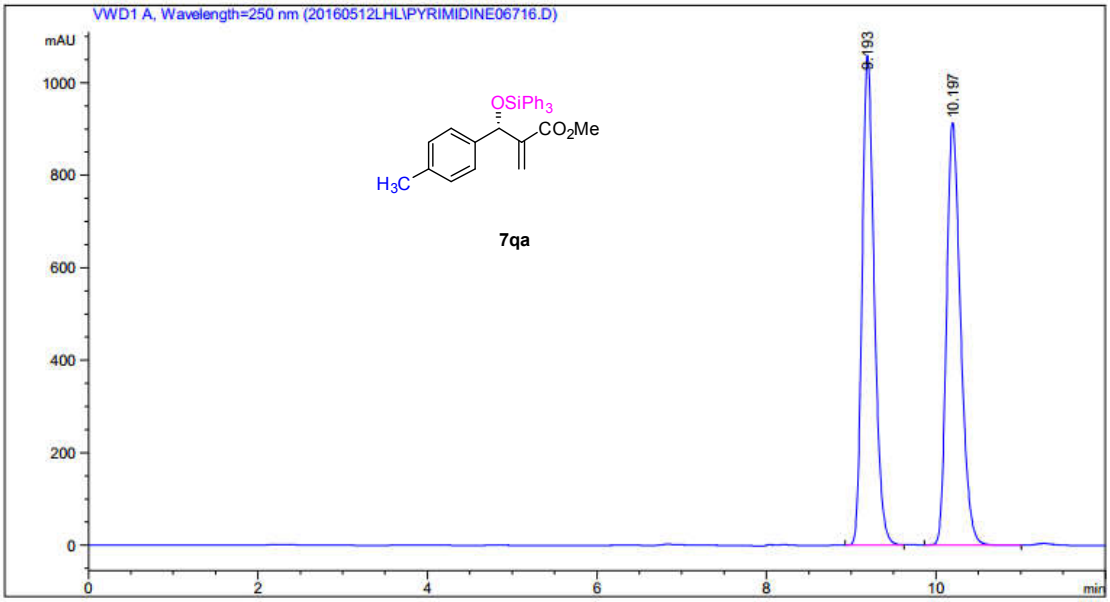
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	9.835	BV	0.4535	1.24372e4	399.43970	92.7811
2	11.706	VB	0.4969	967.68219	28.79776	7.2189



Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	16.350	MM	0.4767	728.31909	25.46144	50.2882
2	18.541	MM	0.5643	719.97144	21.26294	49.7118

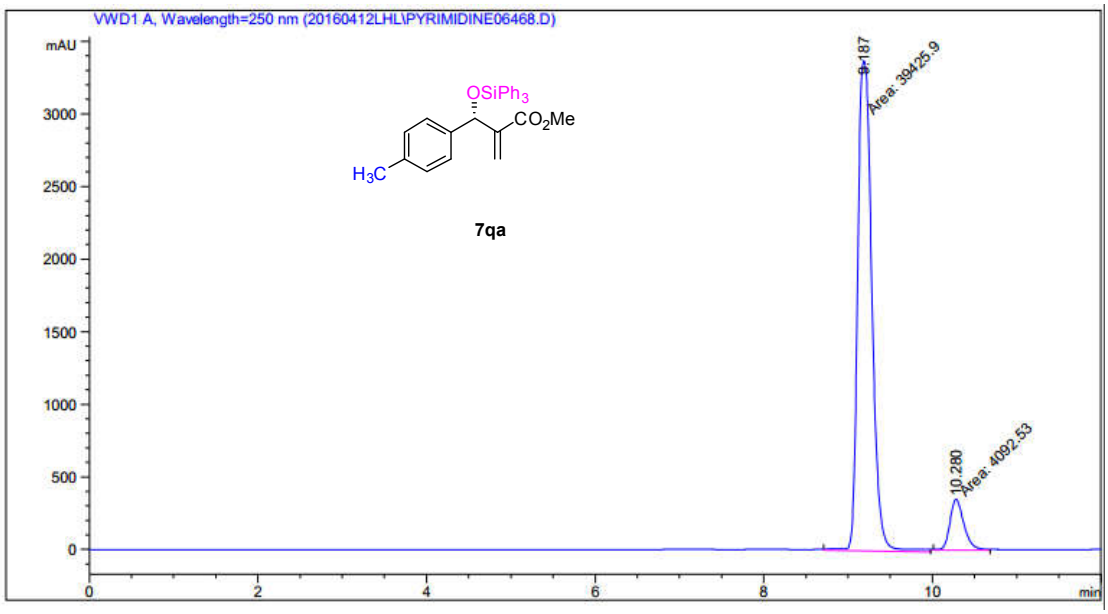


Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	15.951	BV	0.5496	1.26699e4	342.42725	91.4260
2	18.539	VB	0.5432	1188.19324	31.72512	8.5740



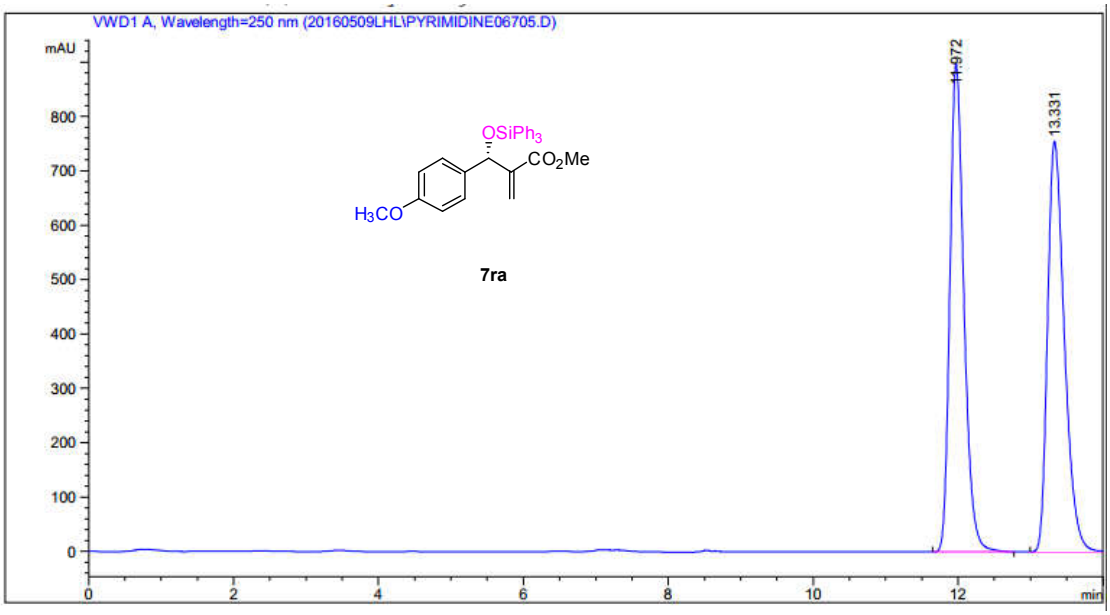
7qa

Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	9.193	VV	0.1519	1.04673e4	1057.66626	49.8654
2	10.197	BB	0.1766	1.05238e4	913.42200	50.1346



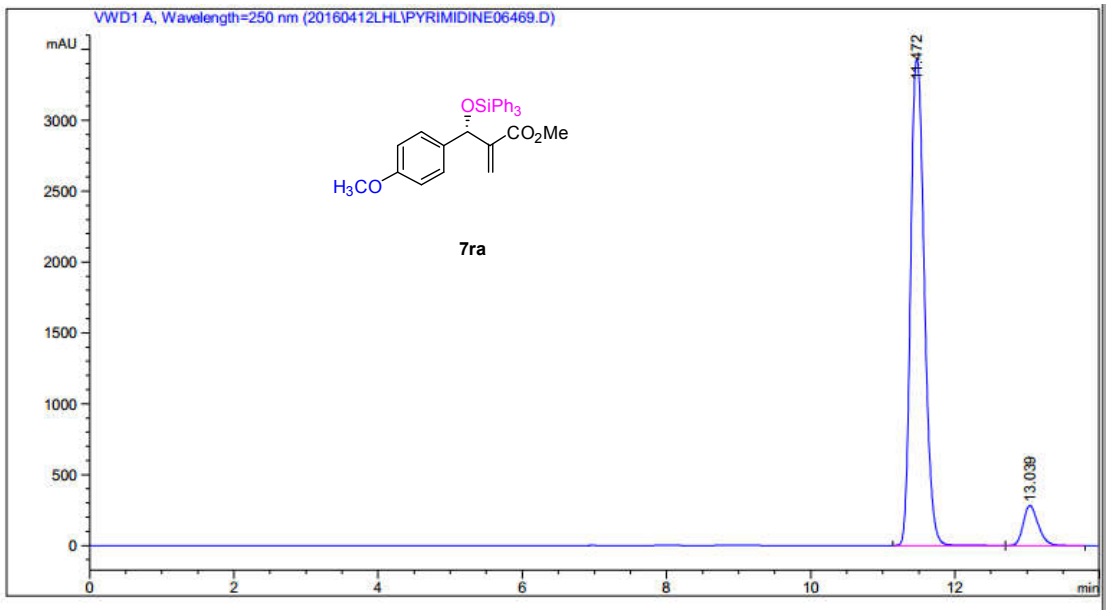
7qa

Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	9.187	MM	0.1947	3.94259e4	3375.13135	90.5959
2	10.280	MM	0.1946	4092.52661	350.55713	9.4041



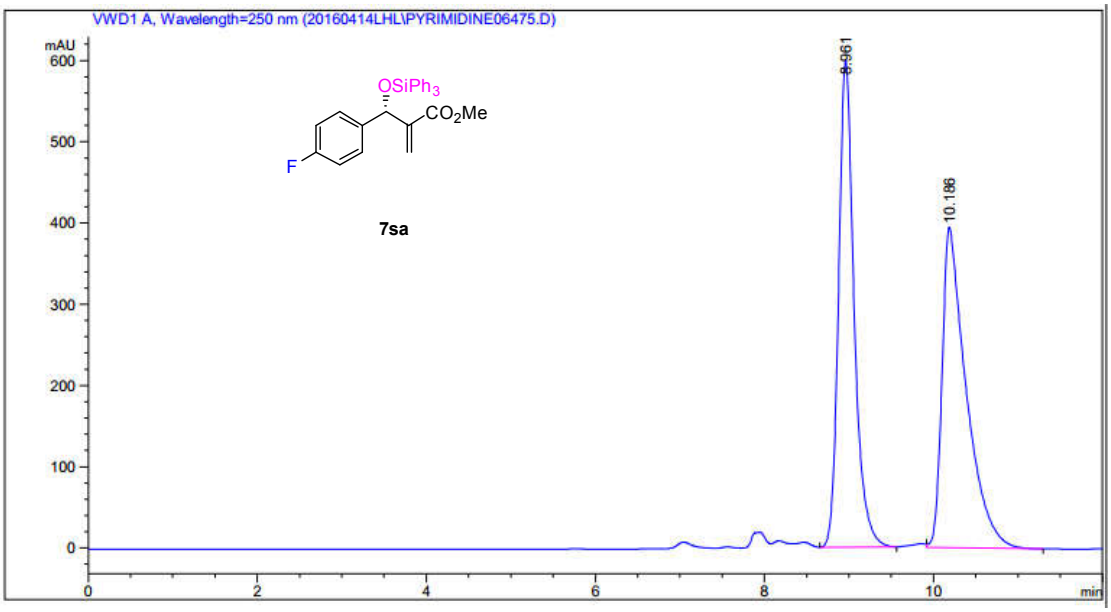
7ra

Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	11.972	BV	0.2049	1.20702e4	897.69958	49.8592
2	13.331	VB	0.2488	1.21384e4	754.67615	50.1408

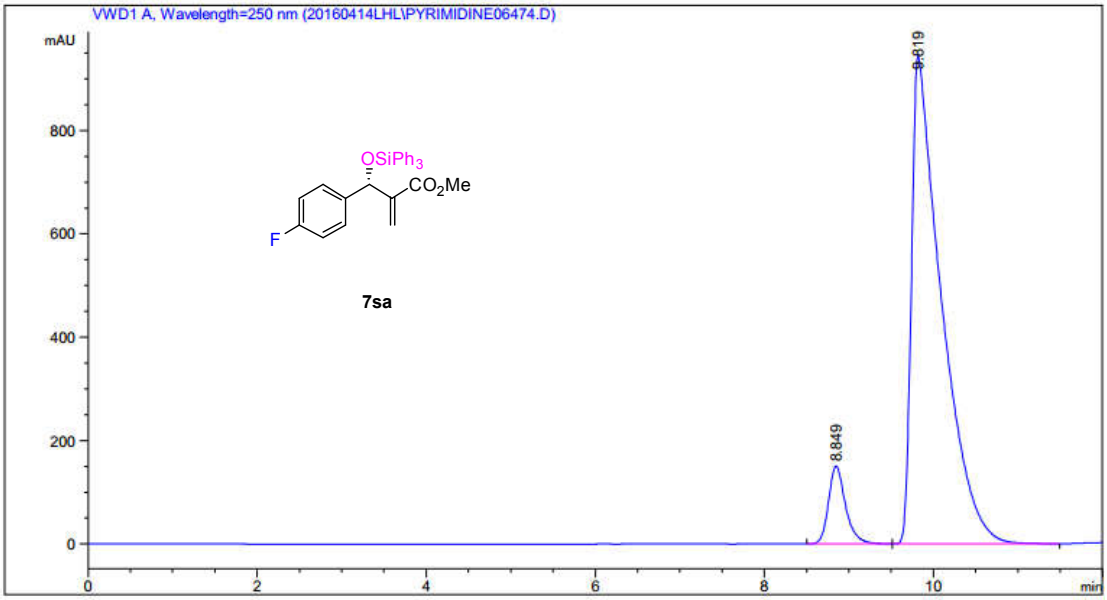


7ra

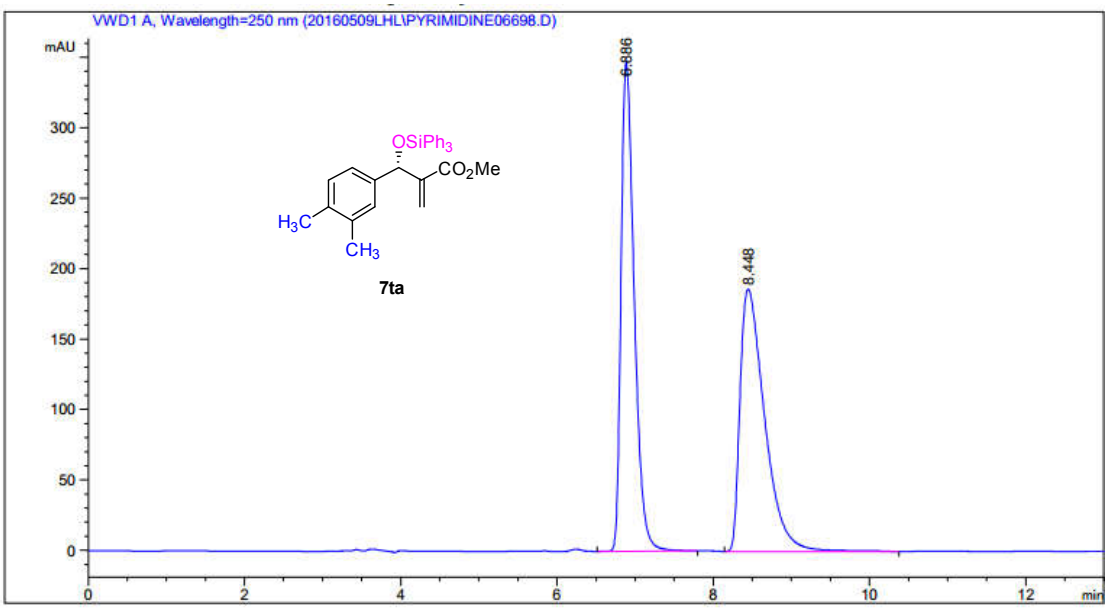
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	11.472	VB	0.2043	4.51086e4	3431.19409	91.8042
2	13.039	BB	0.2210	4027.06128	281.16498	8.1958



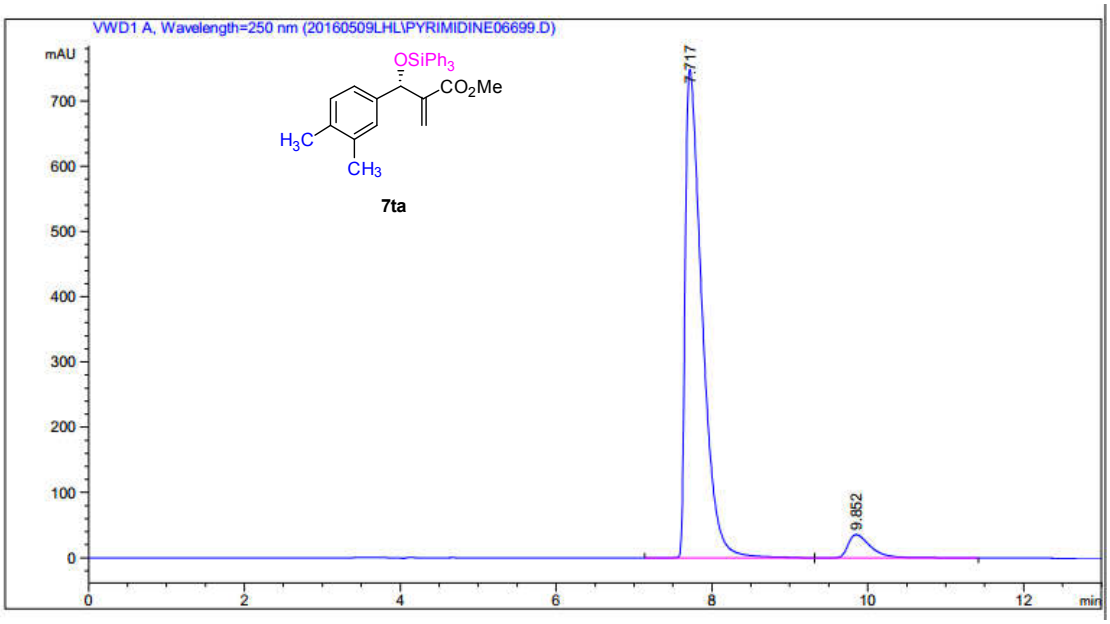
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	8.961	BB	0.1923	7624.82227	598.59155	49.4240
2	10.186	VB	0.2775	7802.53027	394.27710	50.5760



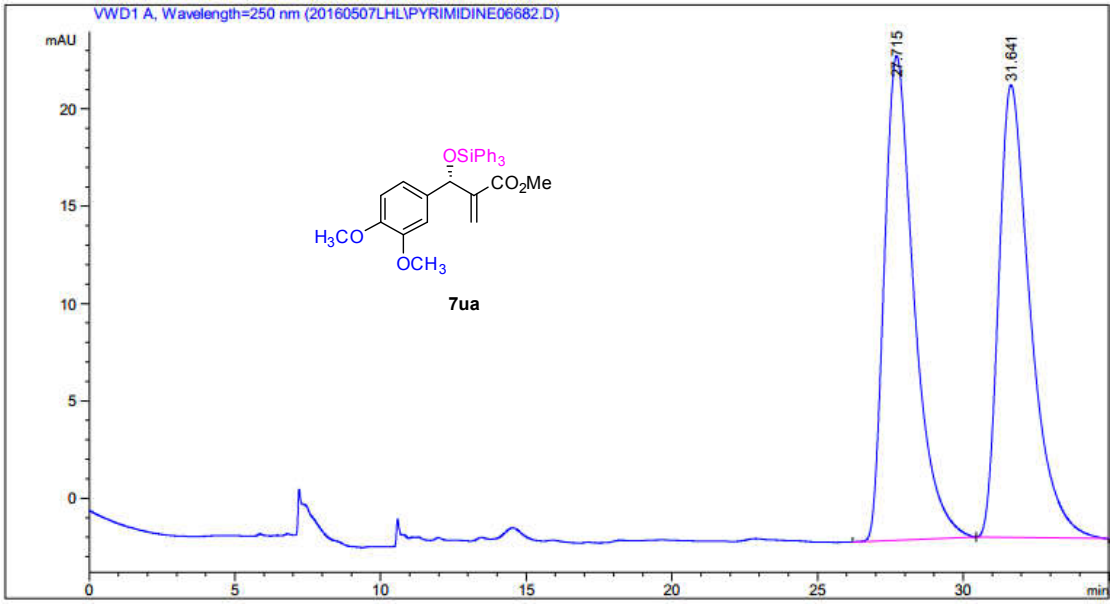
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	8.849	BB	0.2112	2103.70239	150.40987	8.3233
2	9.819	BV	0.3358	2.31712e4	944.29541	91.6767



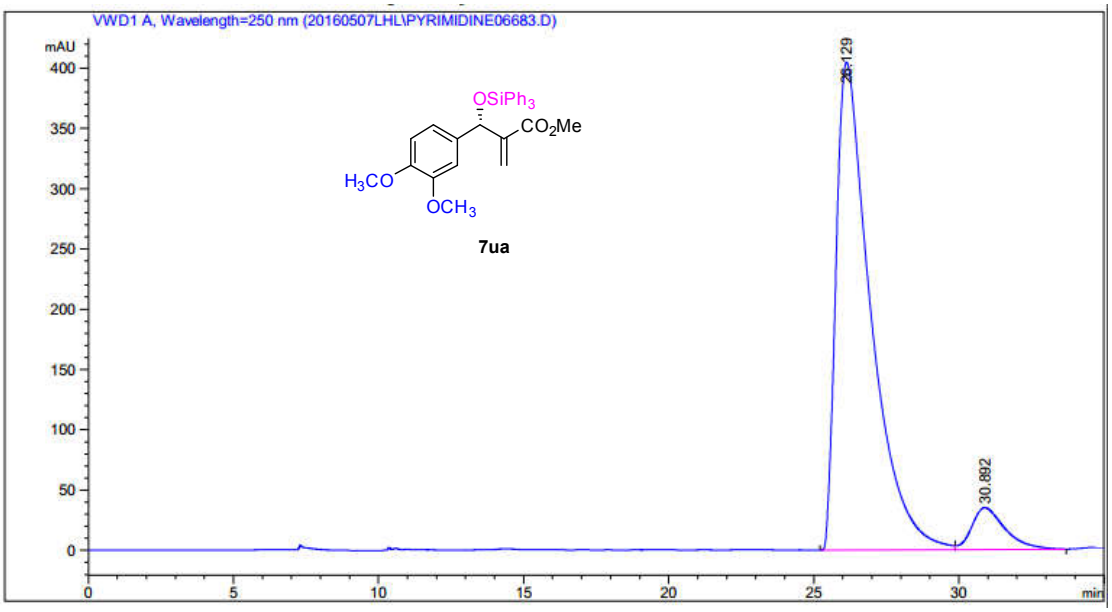
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	6.886	BB	0.1805	4066.60840	346.86542	49.9086
2	8.448	BB	0.3343	4081.50879	186.23712	50.0914



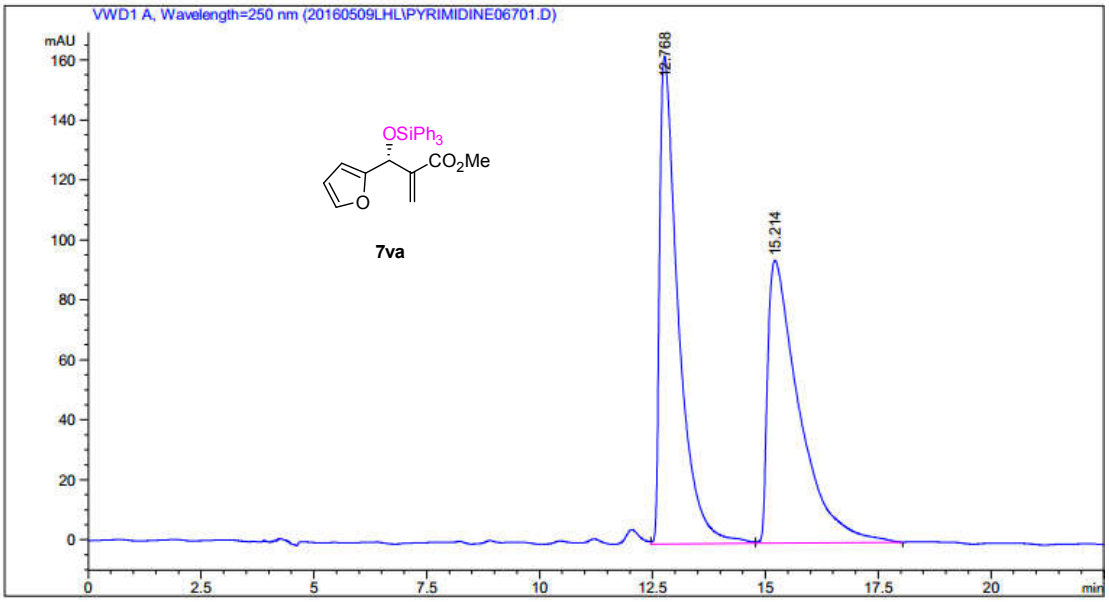
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	7.717	BB	0.2265	1.12556e4	748.08954	93.8115
2	9.852	BB	0.3103	742.50446	35.81837	6.1885



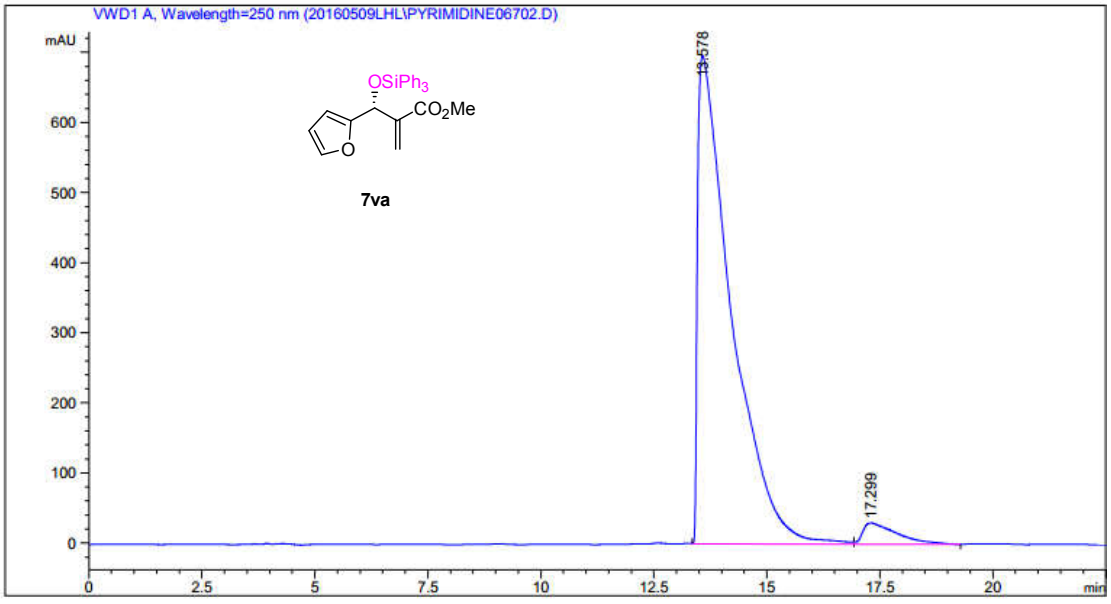
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	27.715	BB	1.0511	1767.05713	24.88709	50.0829
2	31.641	BB	1.1197	1761.20825	23.24776	49.9171



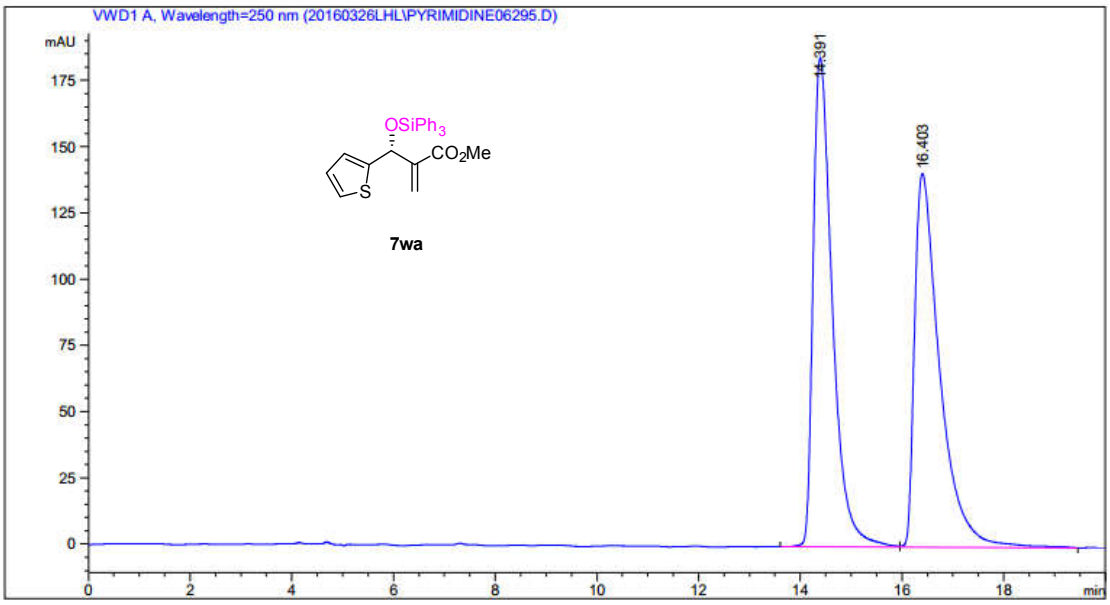
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	26.129	BV	1.1878	3.36073e4	404.45731	92.4430
2	30.892	VB	1.1780	2747.33618	34.60909	7.5570



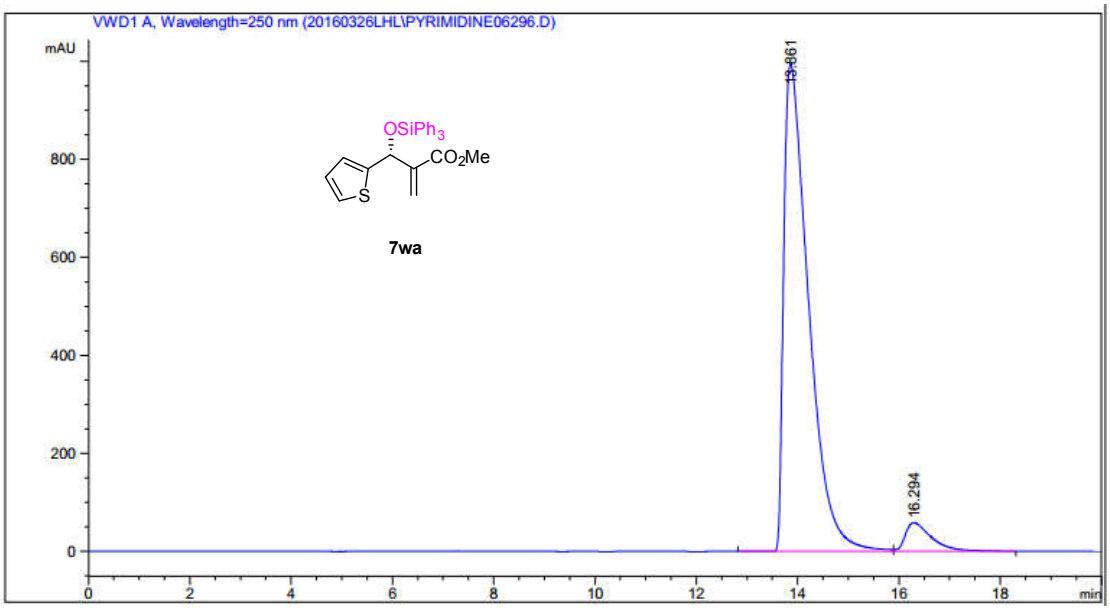
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	12.768	VV	0.4227	4714.61182	162.59259	50.7302
2	15.214	VB	0.7053	4578.89160	94.22749	49.2698



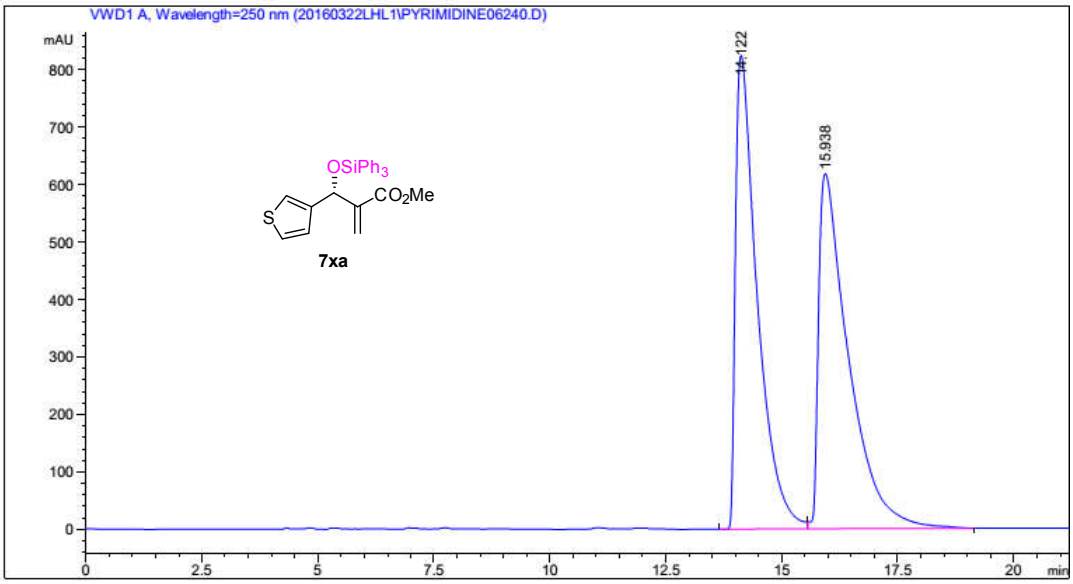
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	13.578	VV	0.7102	3.60462e4	696.31445	95.7804
2	17.299	VB	0.7179	1588.01416	30.29060	4.2196



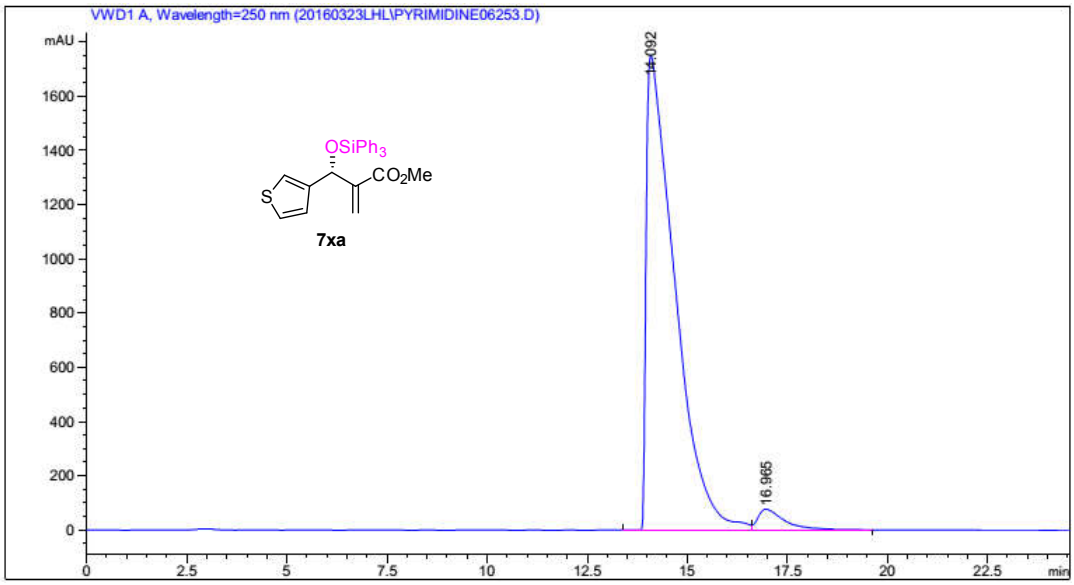
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	14.391	BV	0.4012	4852.22656	184.66325	50.0041
2	16.403	VB	0.5091	4851.43213	141.03835	49.9959



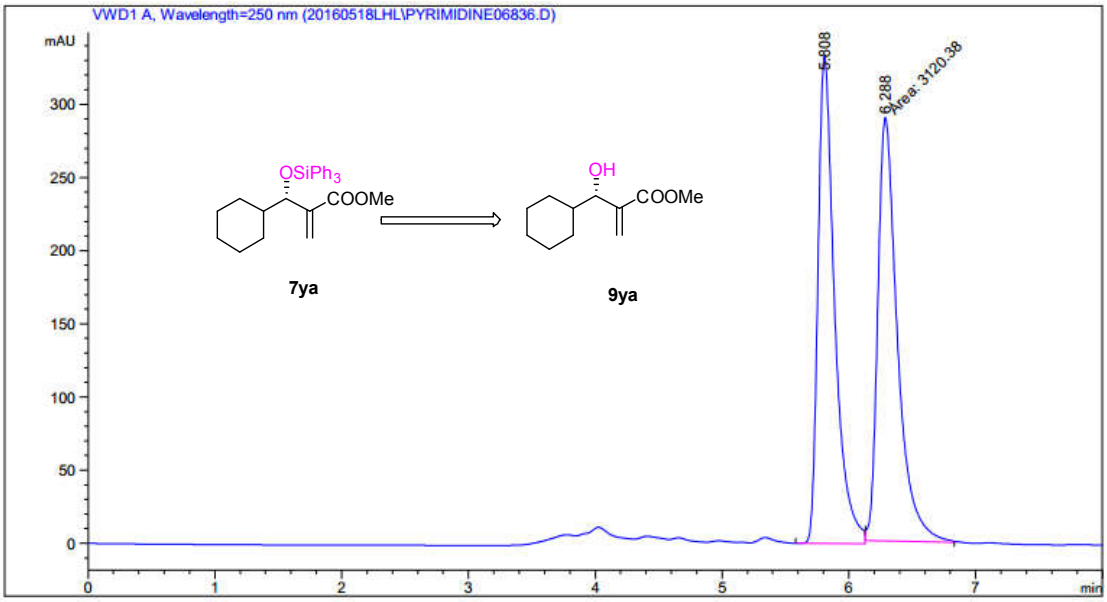
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	13.861	BV	0.5126	3.38122e4	995.82605	94.0780
2	16.294	VB	0.5421	2128.40674	58.54181	5.9220



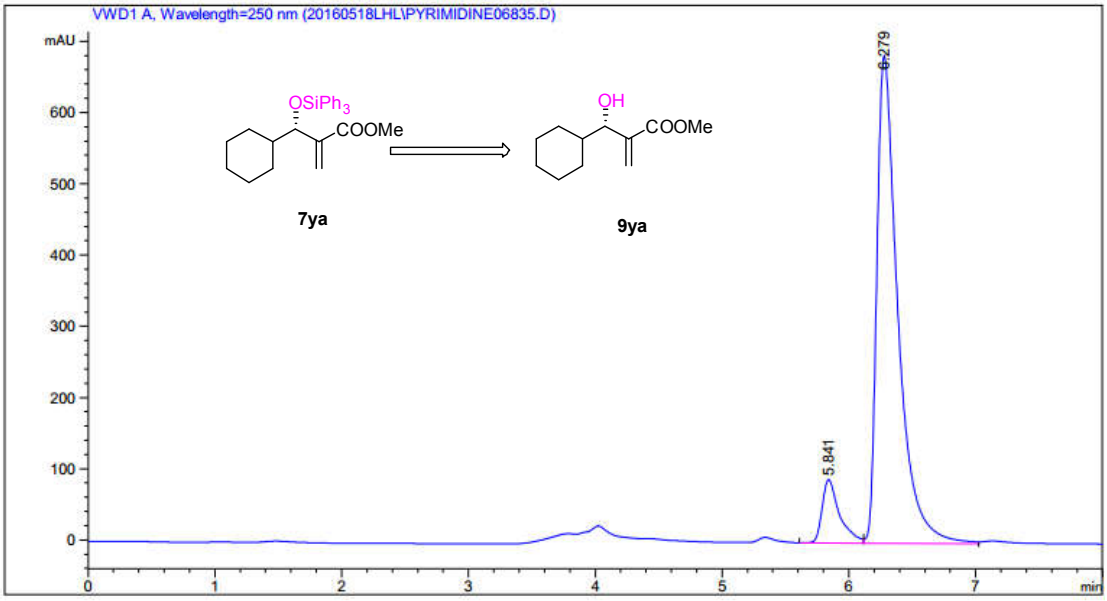
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	14.122	BV	0.4798	2.69244e4	824.87463	49.1602
2	15.938	VB	0.6361	2.78443e4	618.25940	50.8398



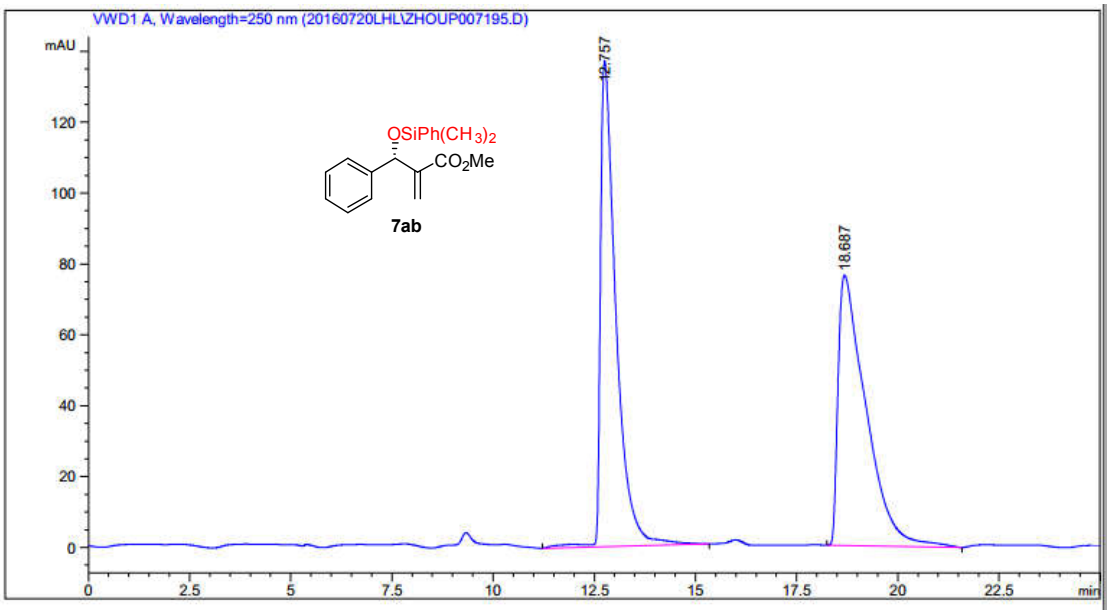
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	14.092	BV	0.7691	8.69784e4	1750.09937	96.0428
2	16.965	VB	0.6850	3583.72827	76.47387	3.9572



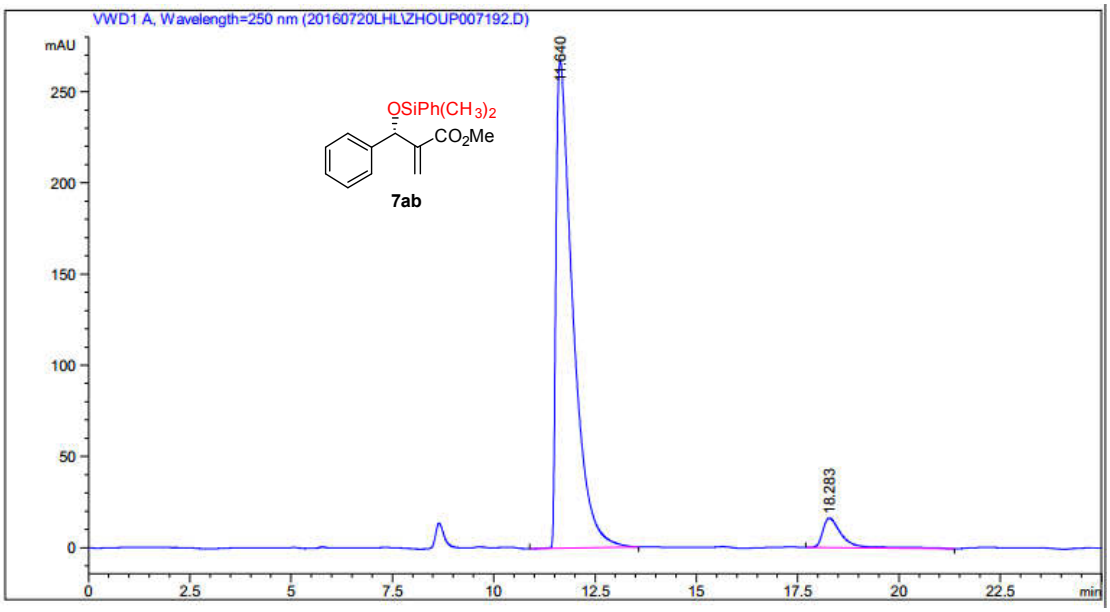
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	5.808	BV	0.1361	3052.74707	332.89465	49.4522
2	6.288	MM	0.1798	3120.37939	289.28937	50.5478



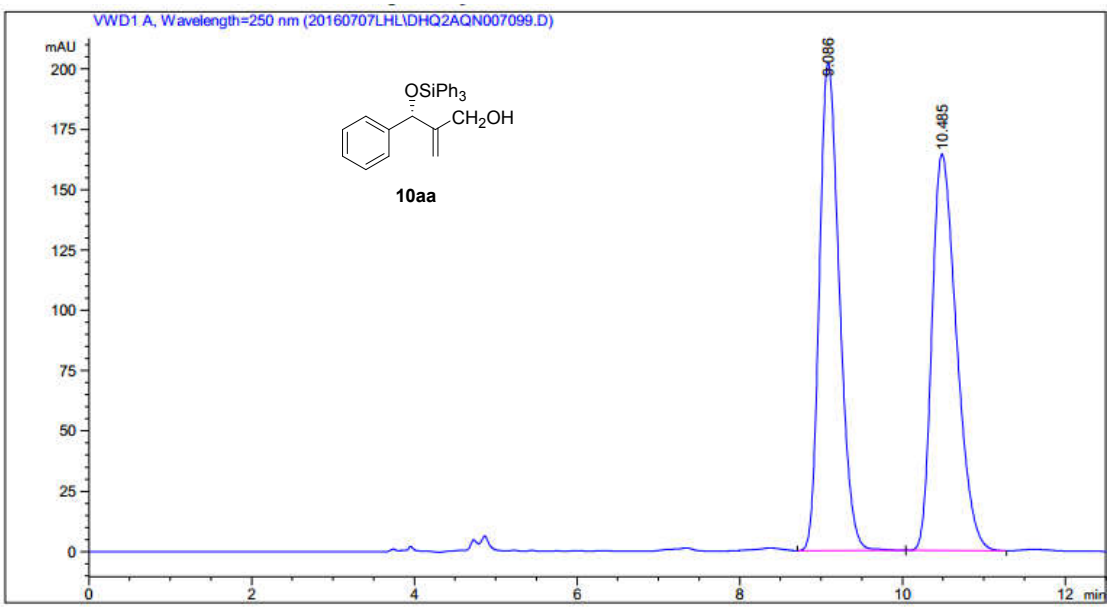
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	5.841	BV	0.1387	845.24249	88.81093	9.6696
2	6.279	VV	0.1710	7895.98926	684.33173	90.3304



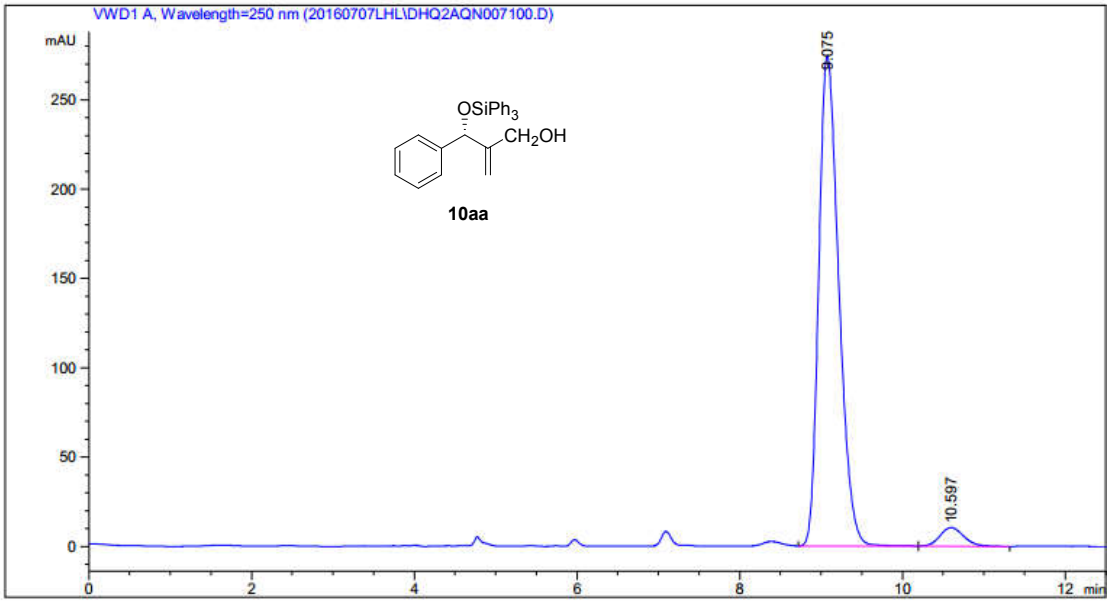
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	12.757	BB	0.3945	3677.64893	137.11285	50.2409
2	18.687	BB	0.6730	3642.38013	76.16355	49.7591



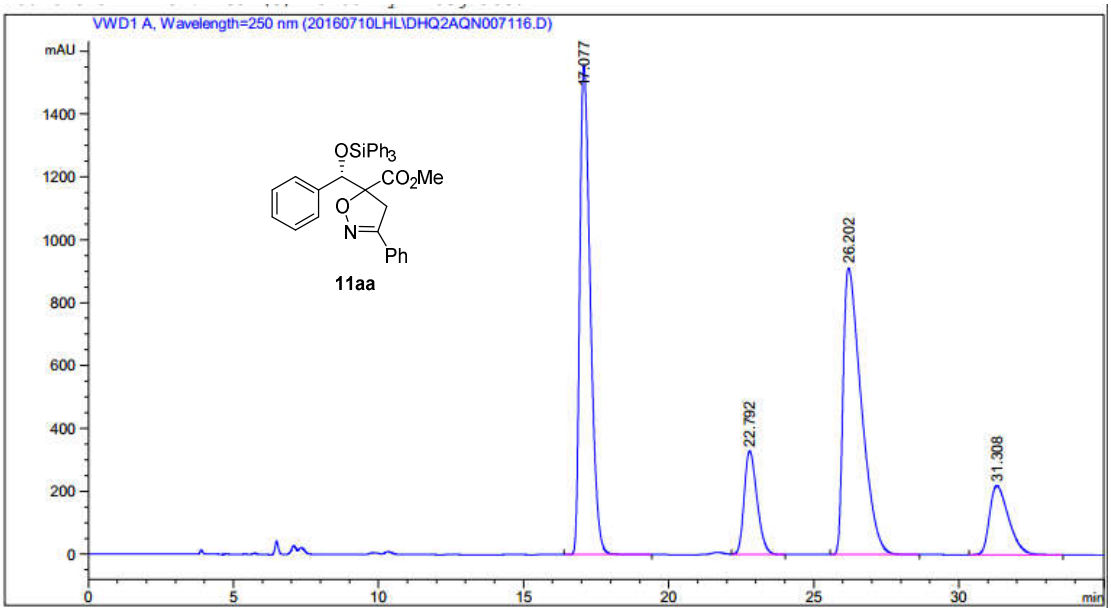
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	11.640	BB	0.4203	7726.89795	267.25018	93.2903
2	18.283	BB	0.5075	555.73975	16.15867	6.7097



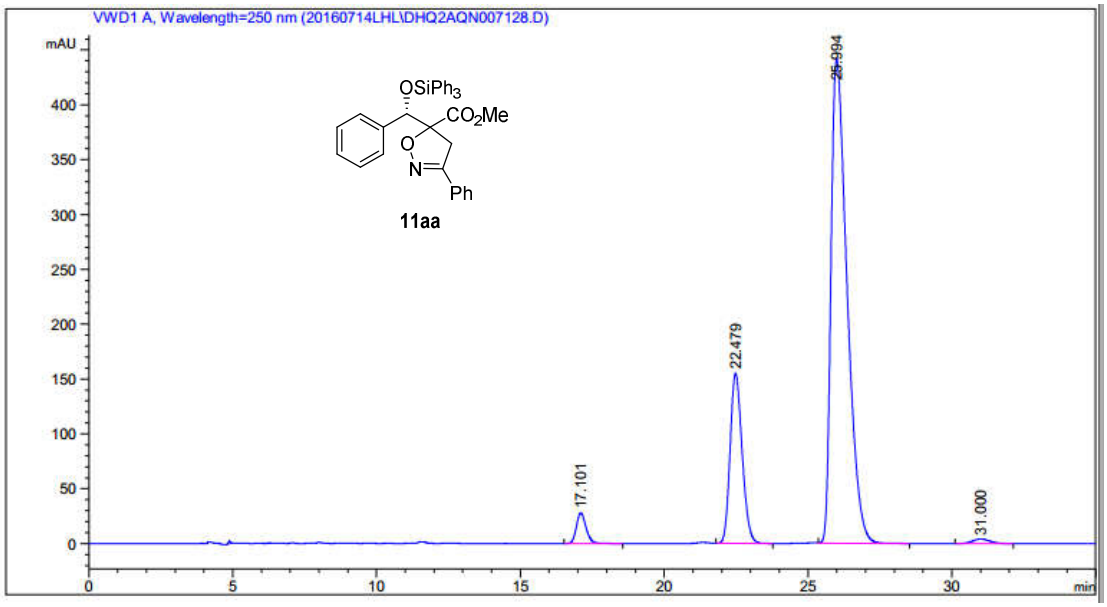
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	9.086	VB	0.2628	3446.42993	202.35806	50.0020
2	10.485	BB	0.3249	3446.15161	164.25748	49.9980



Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	9.075	VV	0.2652	4696.66602	274.43857	95.6178
2	10.597	VB	0.3183	215.24884	10.47913	4.3822



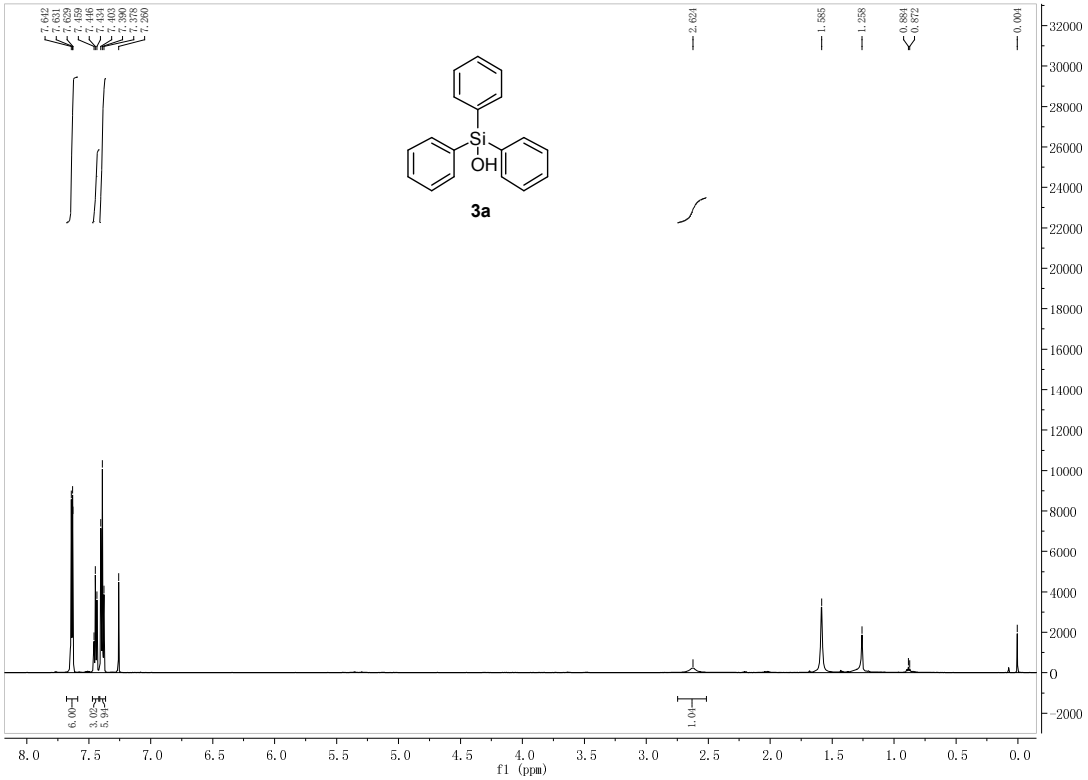
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	17.077	BB	0.3894	3.91311e4	1556.31104	39.5461
2	22.792	VB	0.4779	1.01734e4	329.76599	10.2813
3	26.202	BB	0.6543	3.96553e4	912.04364	40.0758
4	31.308	BB	0.6974	9990.79590	219.34000	10.0967



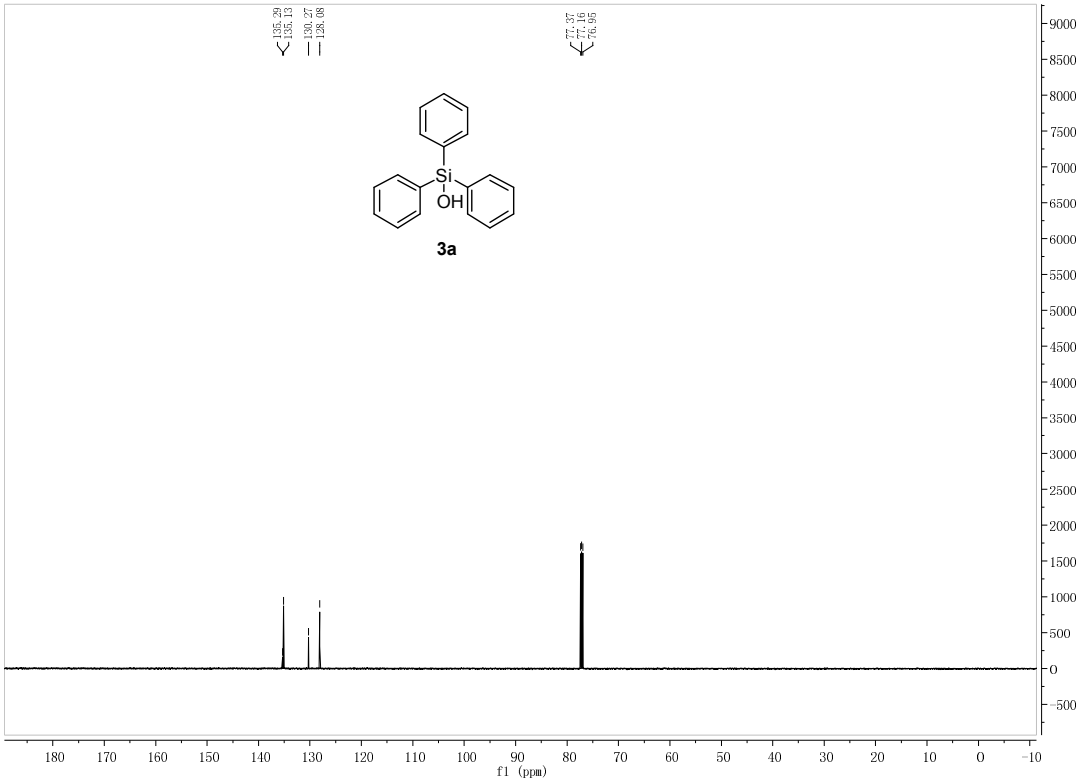
Peak #	RetTime [min]	Type	Width [min]	Area [mAU*s]	Height [mAU]	Area %
1	17.101	BB	0.3603	647.99408	27.84282	2.8926
2	22.479	BB	0.4497	4505.53320	155.04117	20.1122
3	25.994	VB	0.5926	1.70774e4	441.45065	76.2314
4	31.000	BB	0.6120	171.11867	4.13806	0.7639

5. Copies of NMR spectra for the 3a, 4a, 5e, 5f, 7, 9, 10aa and 11aa.

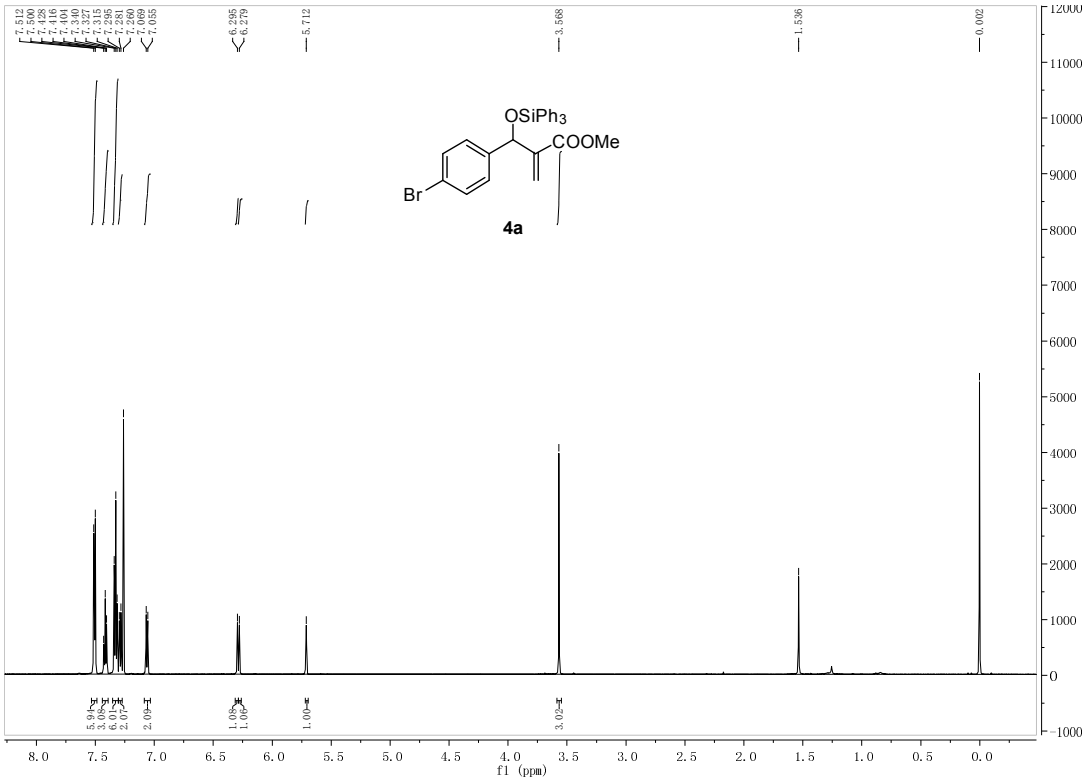
^{1}H-NMR for 3a



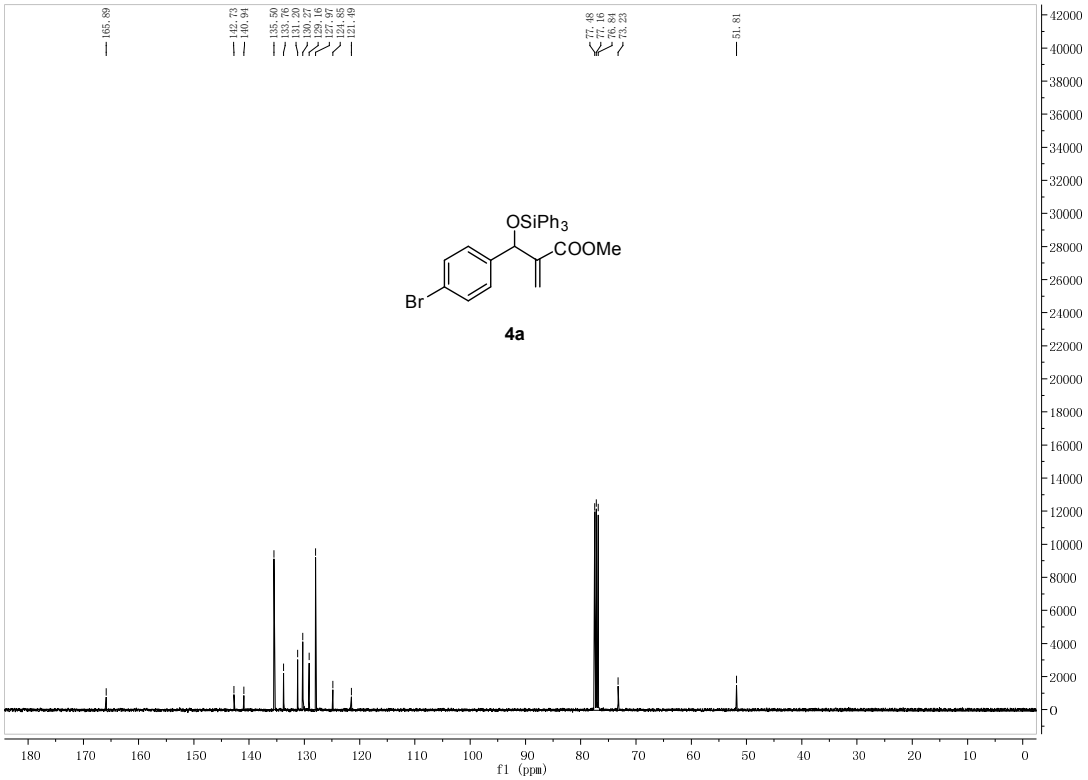
^{13}C-NMR for 3a



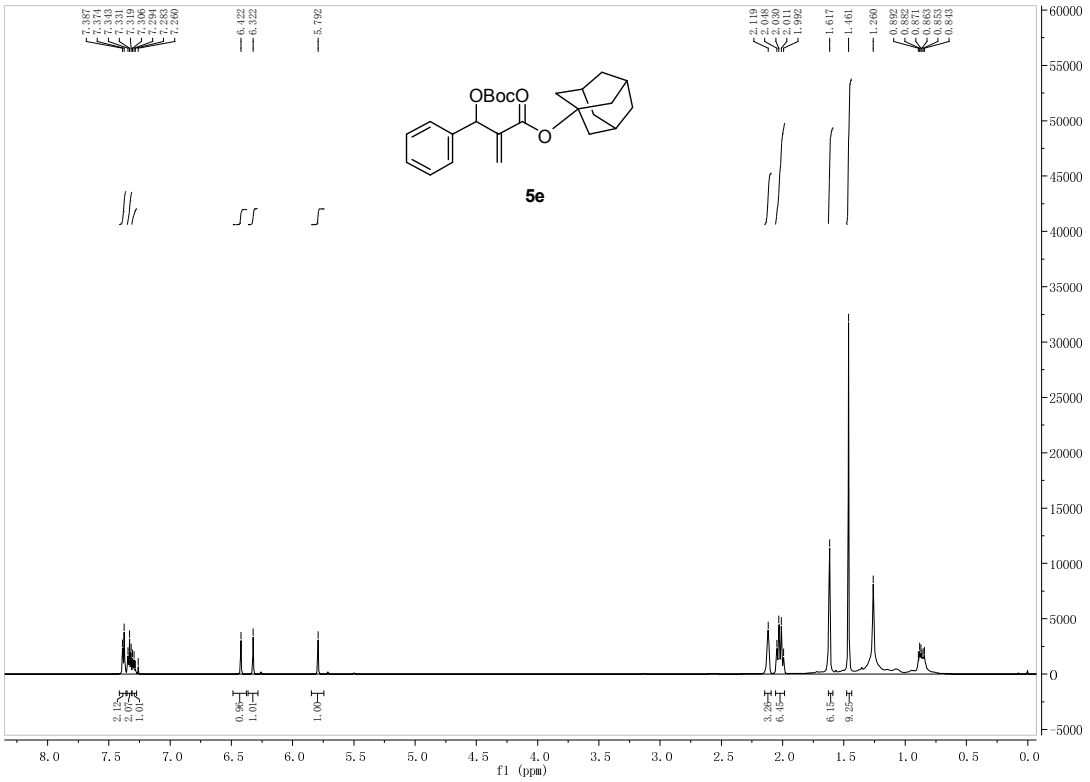
^{1}H-NMR for 4a



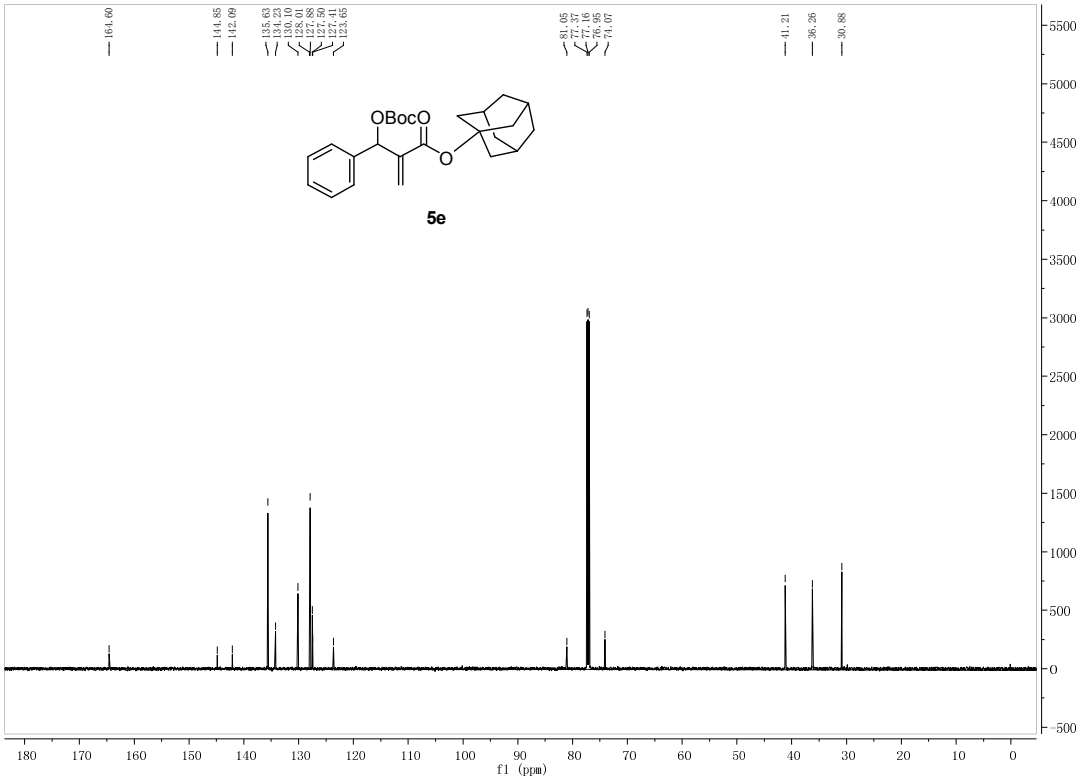
^{13}C-NMR for 4a



^{1}H-NMR for 5e



^{13}C-NMR for 5e



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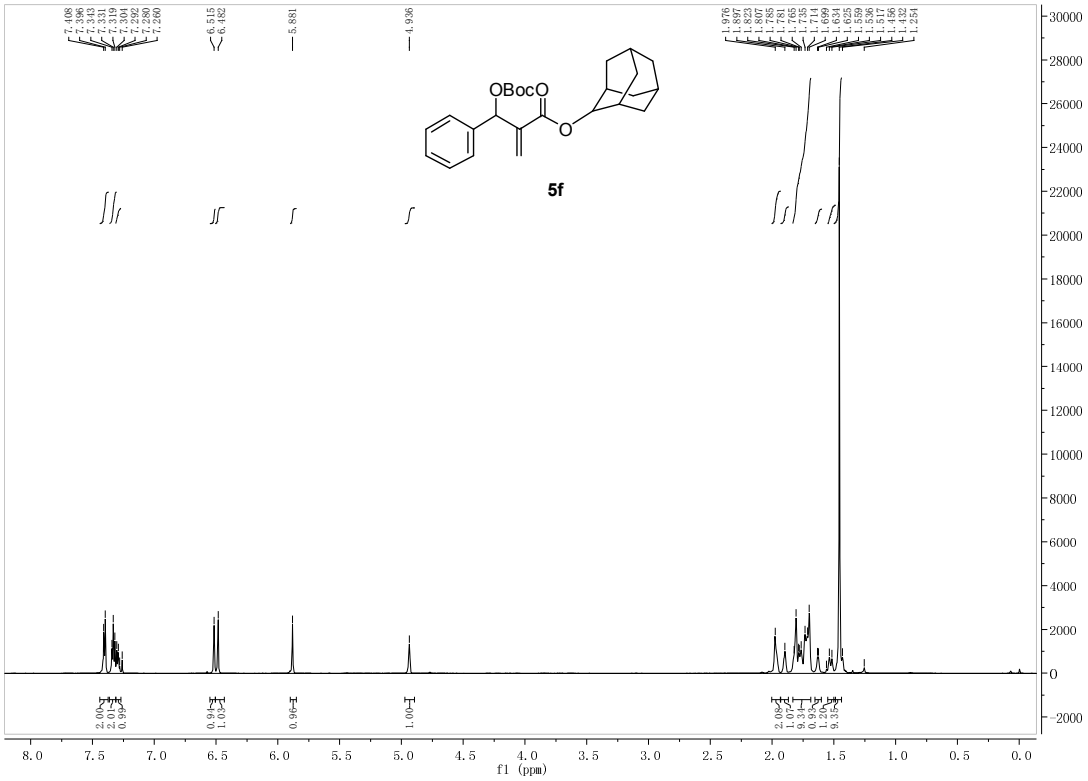

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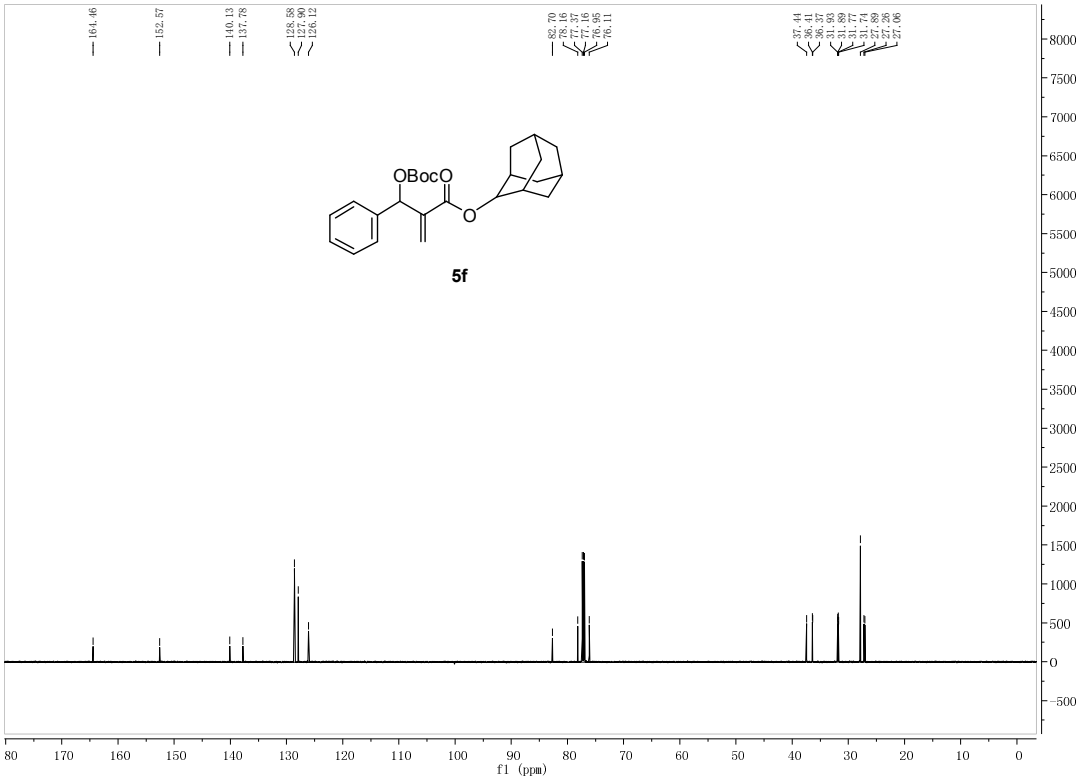

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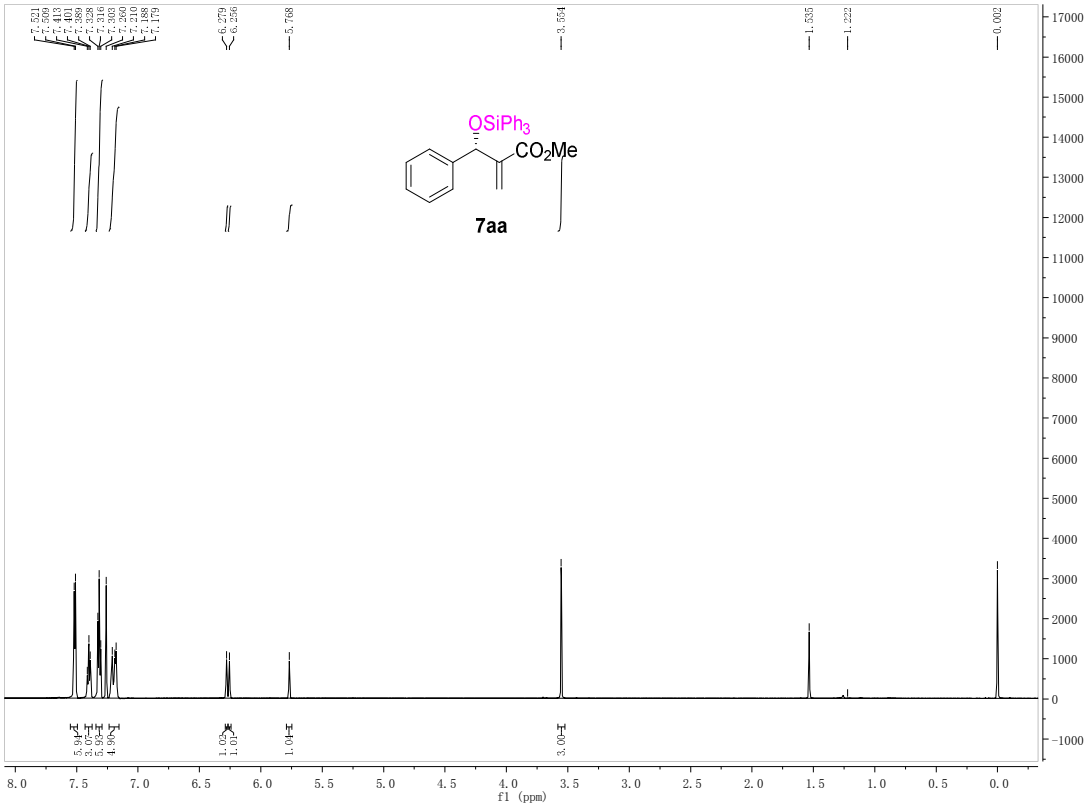

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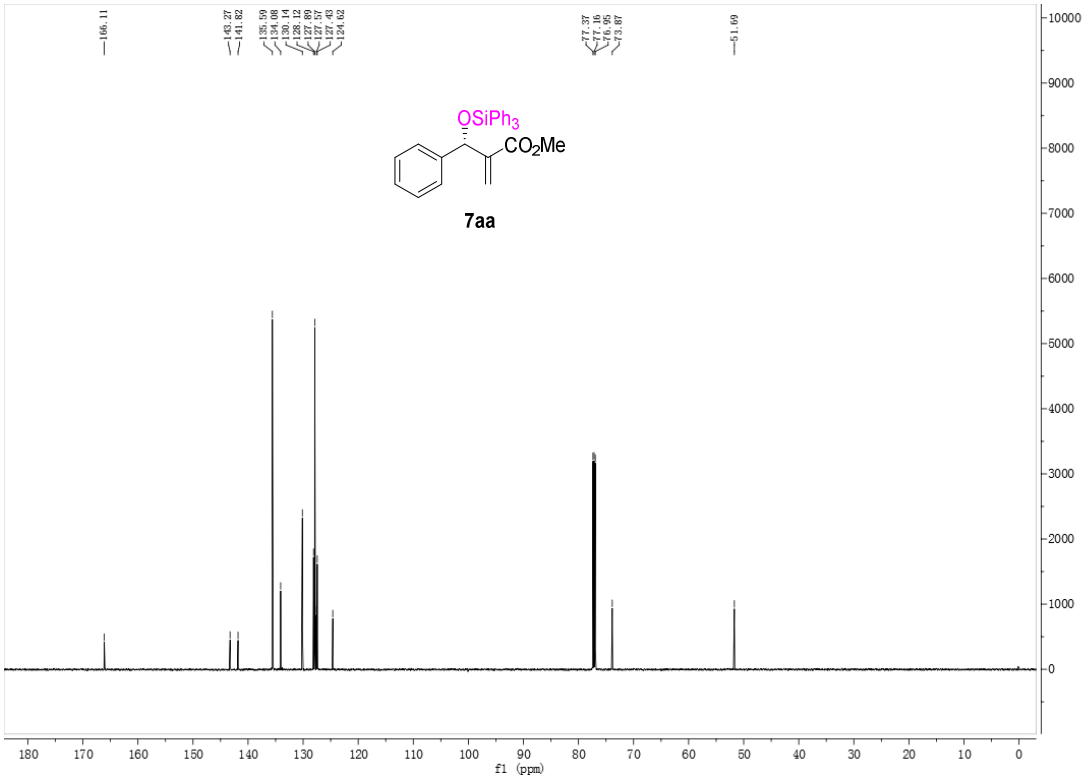

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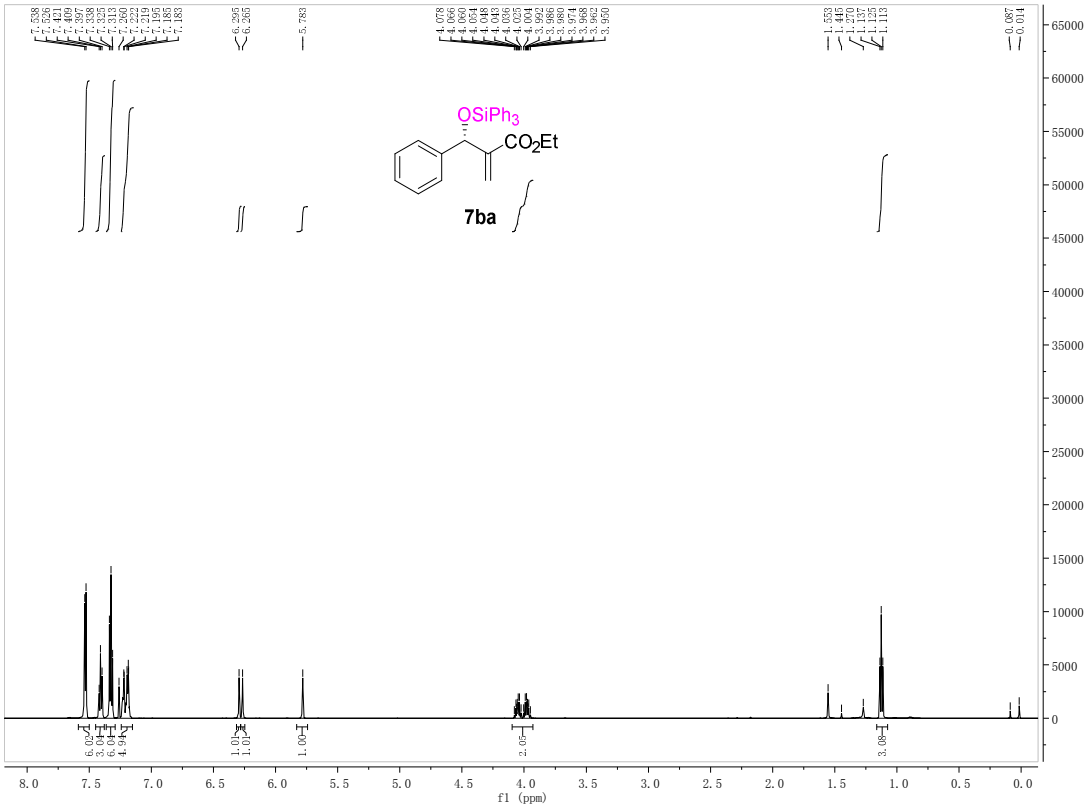
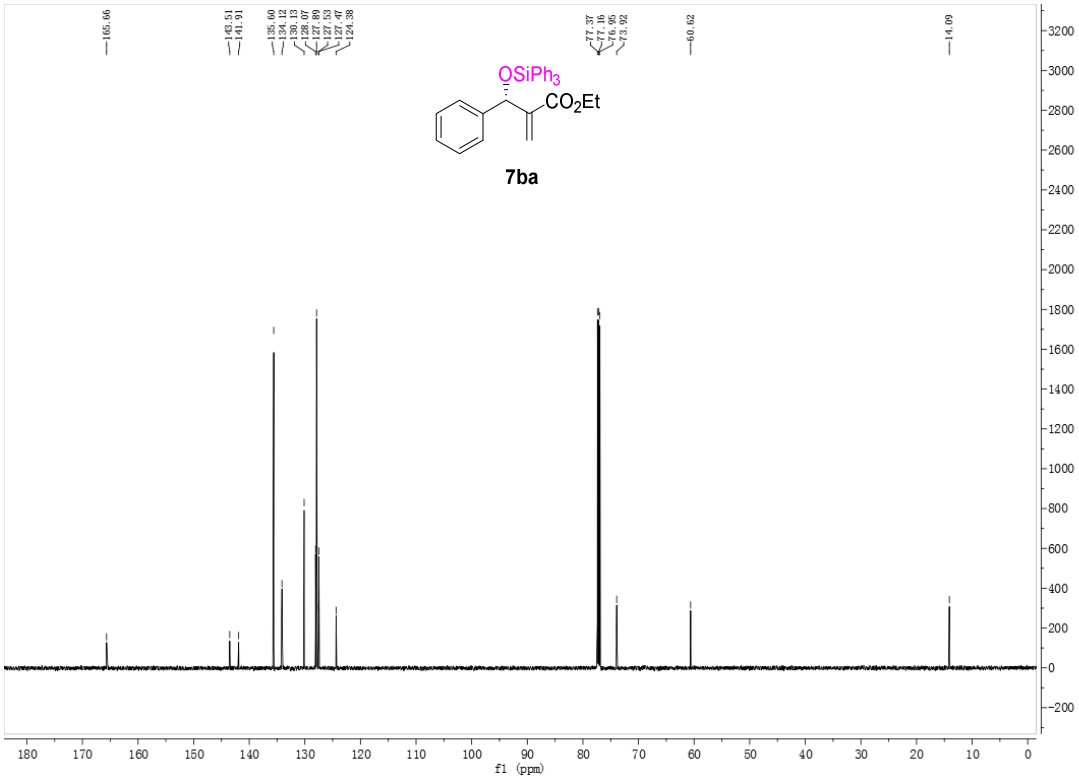
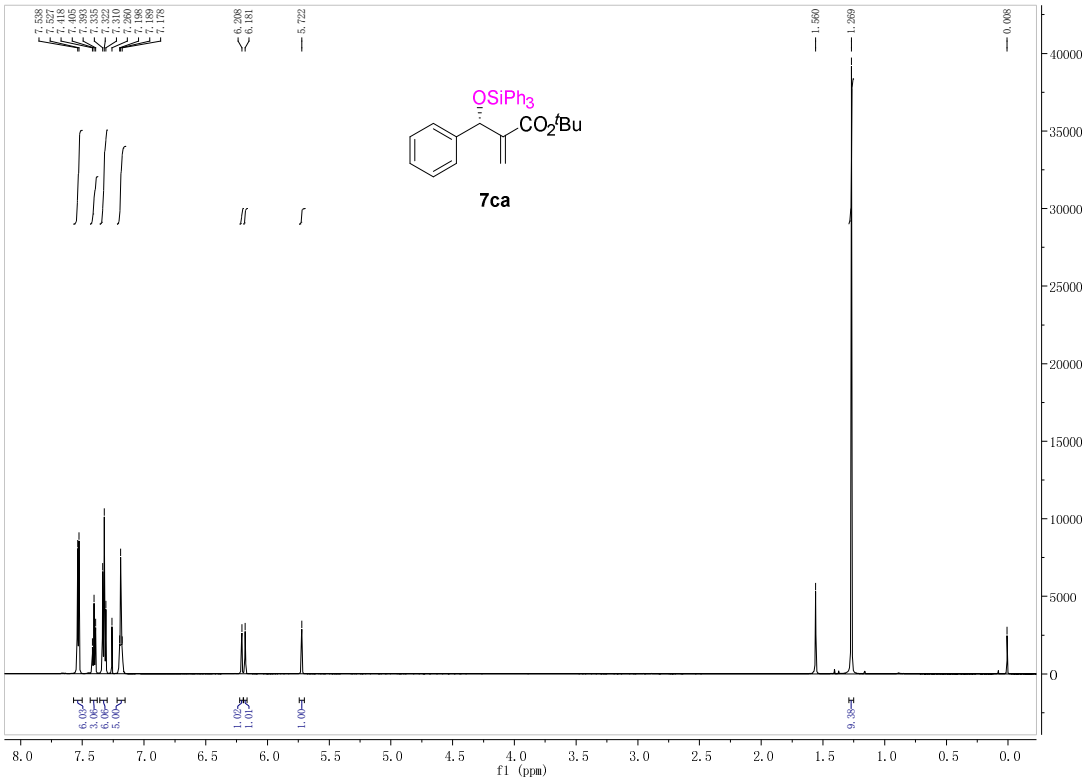


^{13}C-NMR for 7ba



^{1}H-NMR for 7ca



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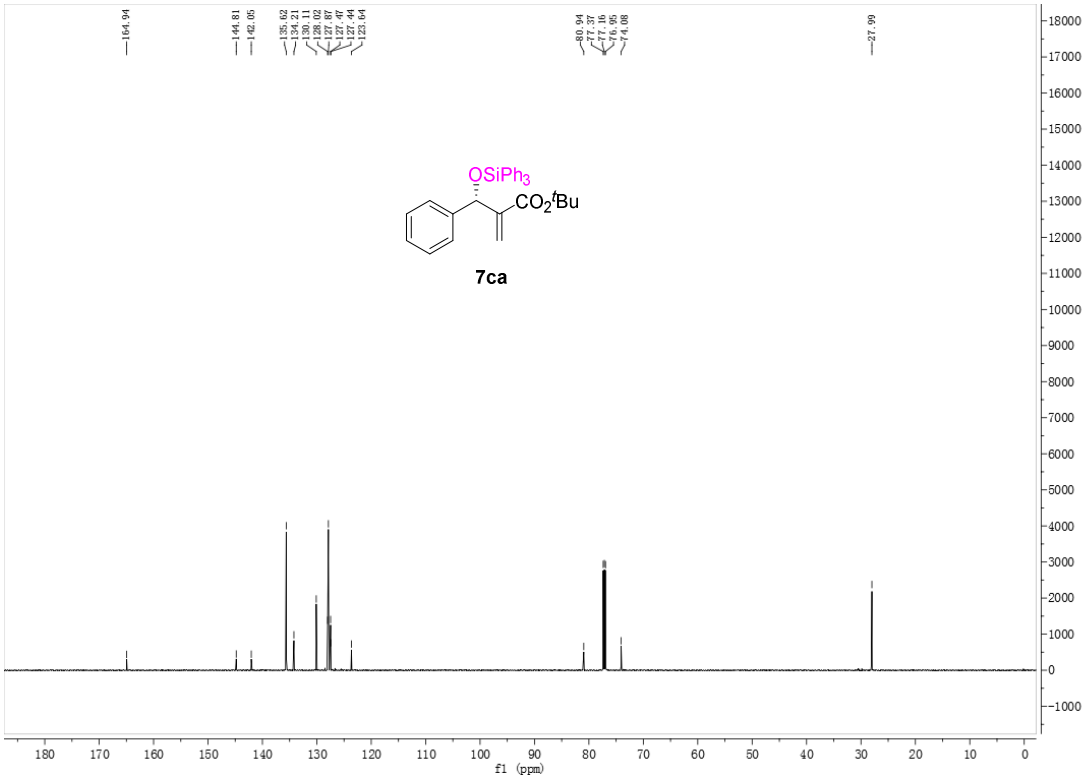

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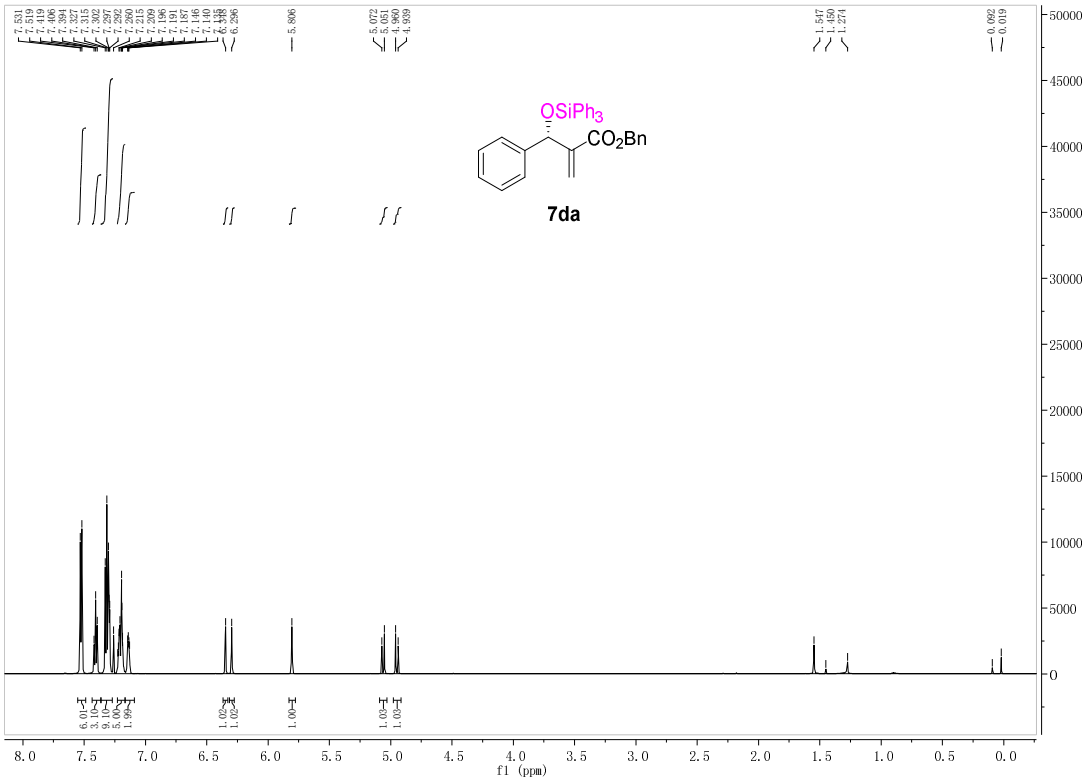

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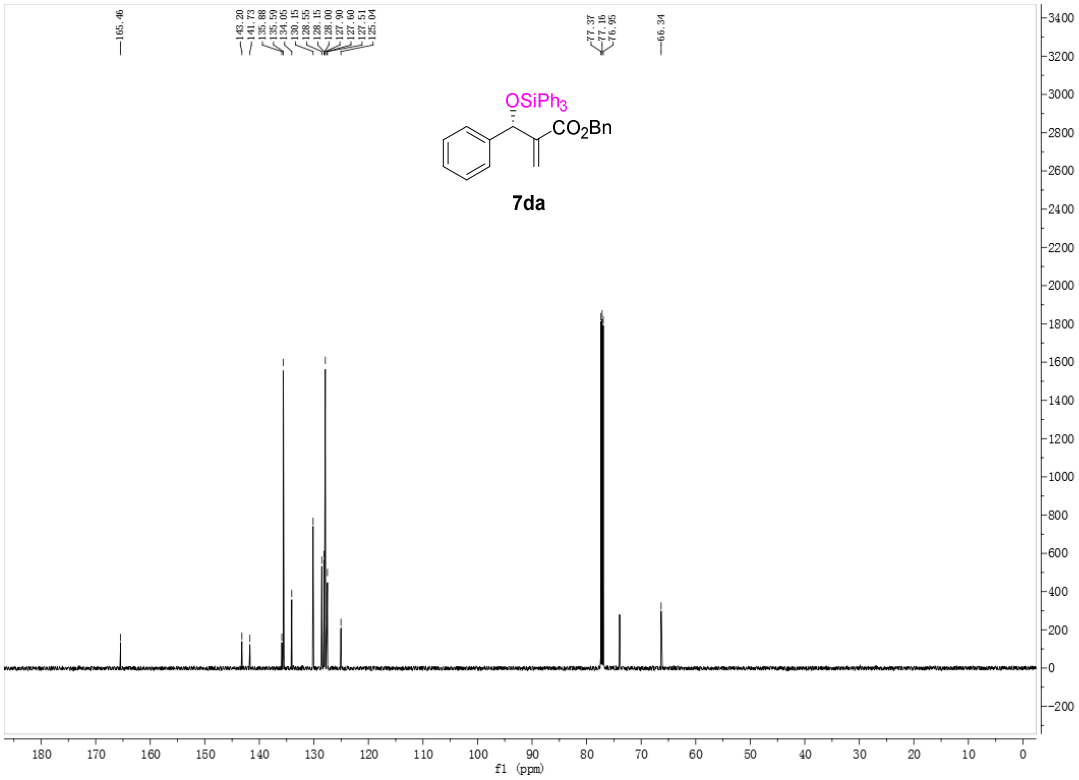

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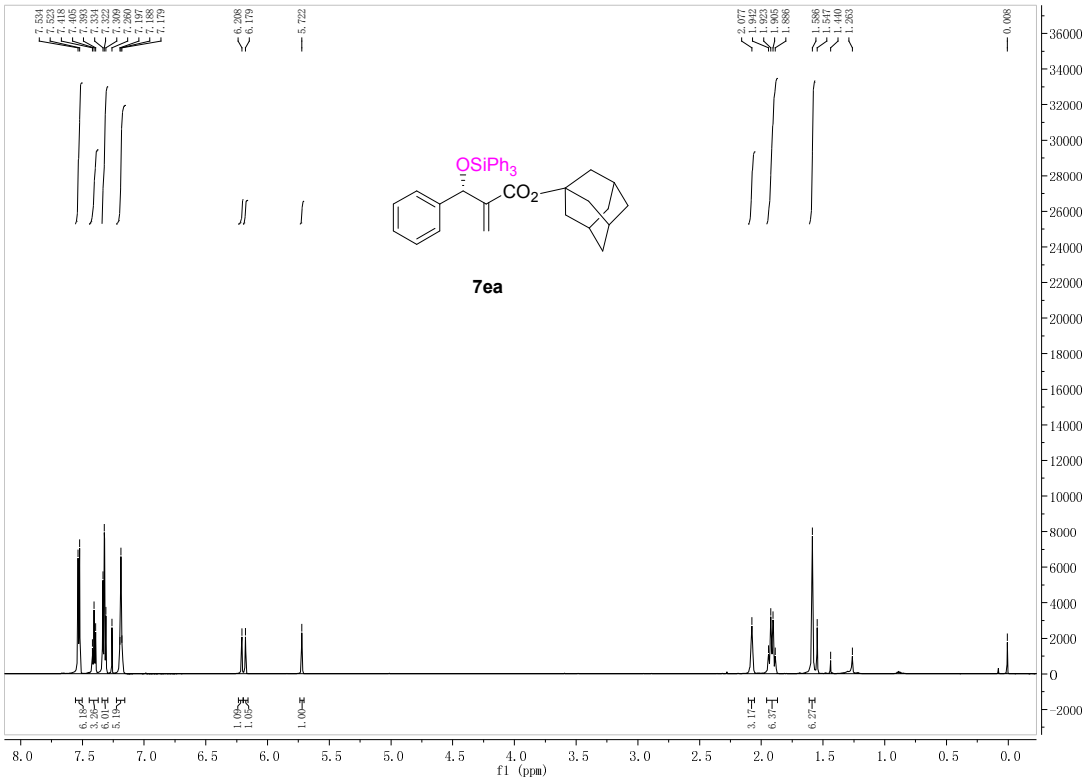

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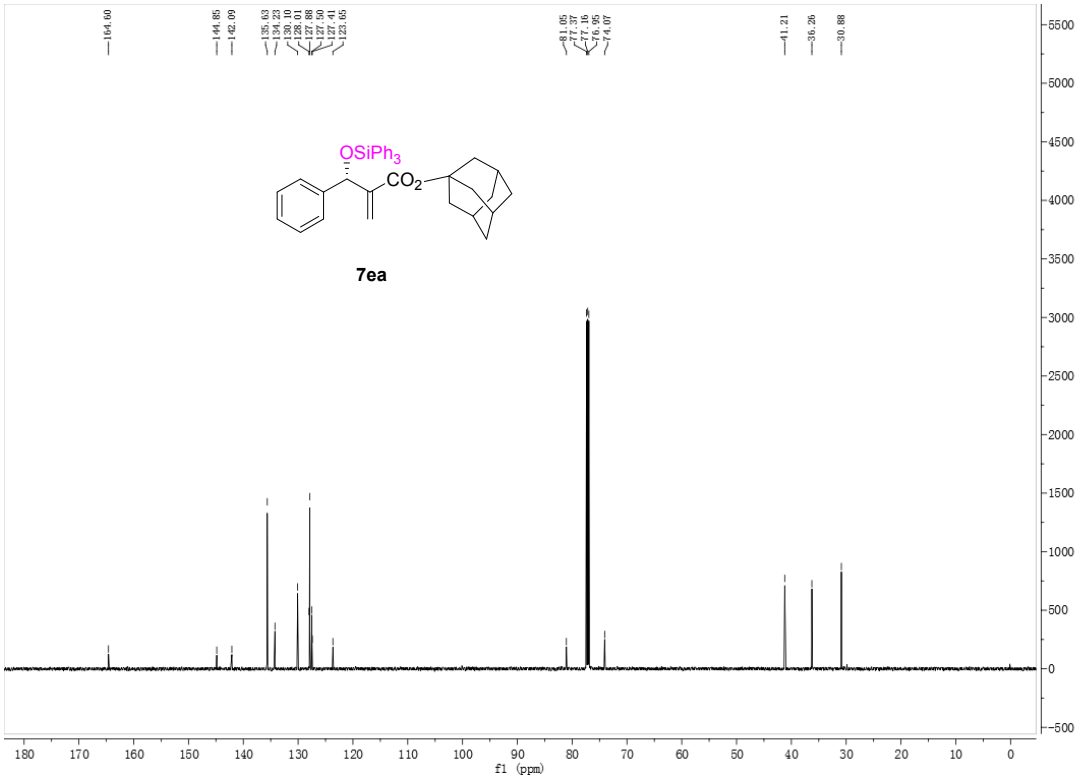

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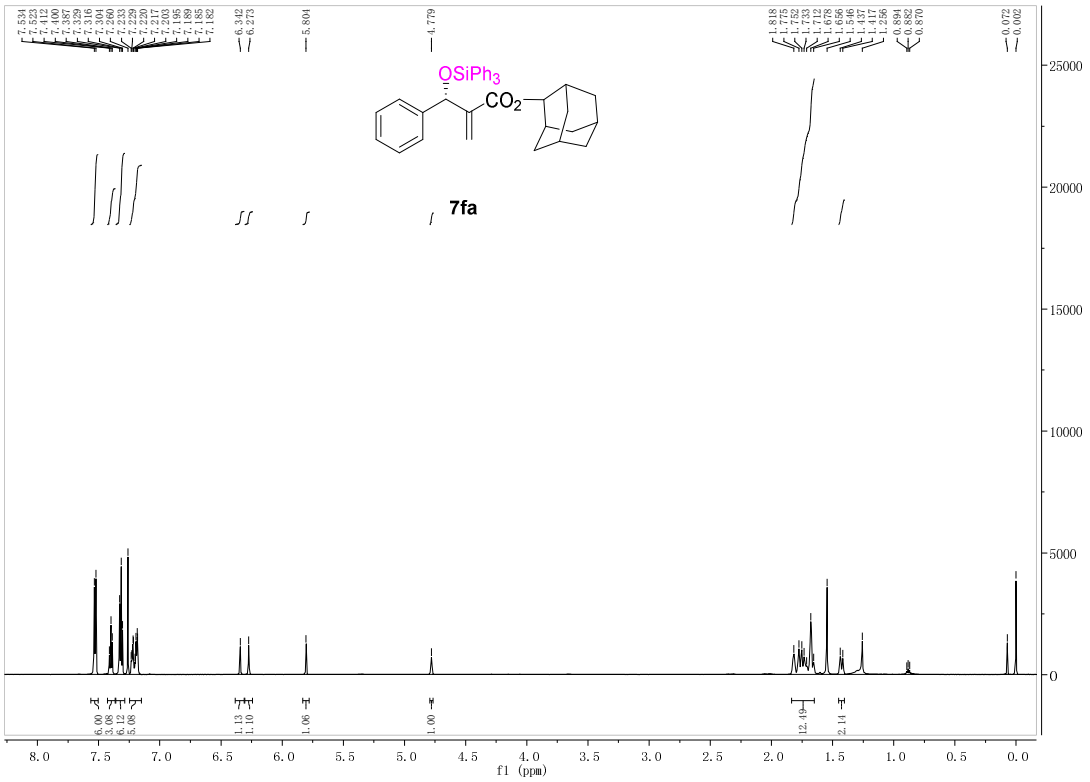

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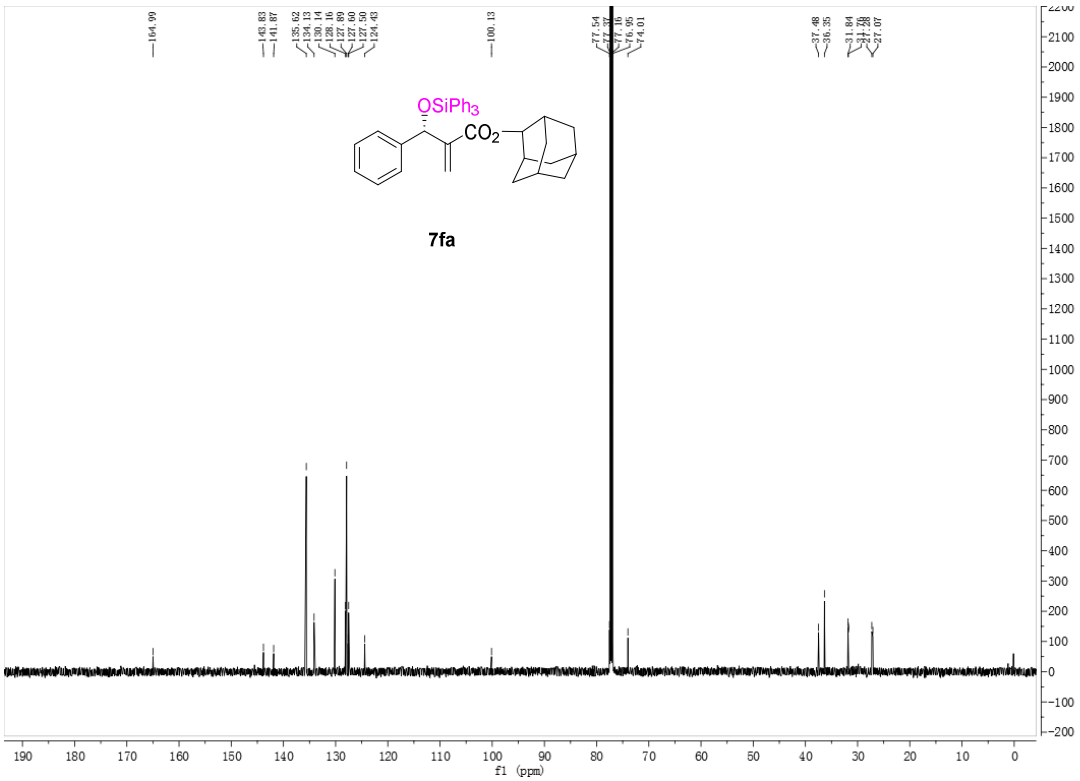
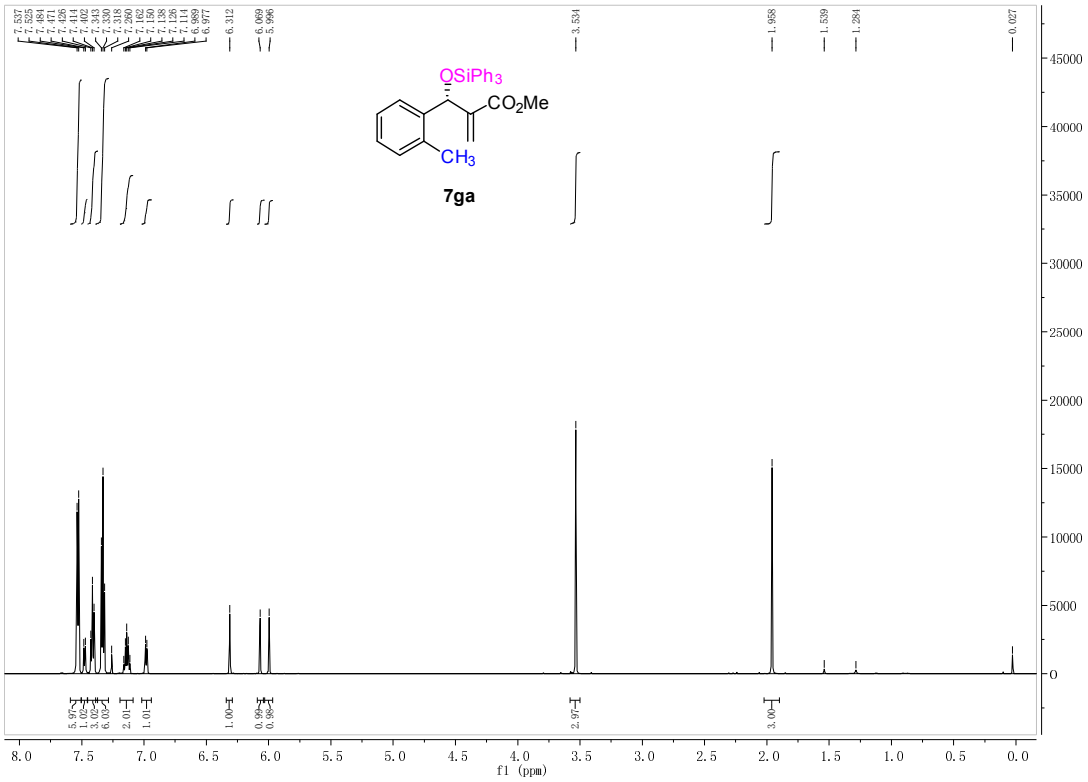
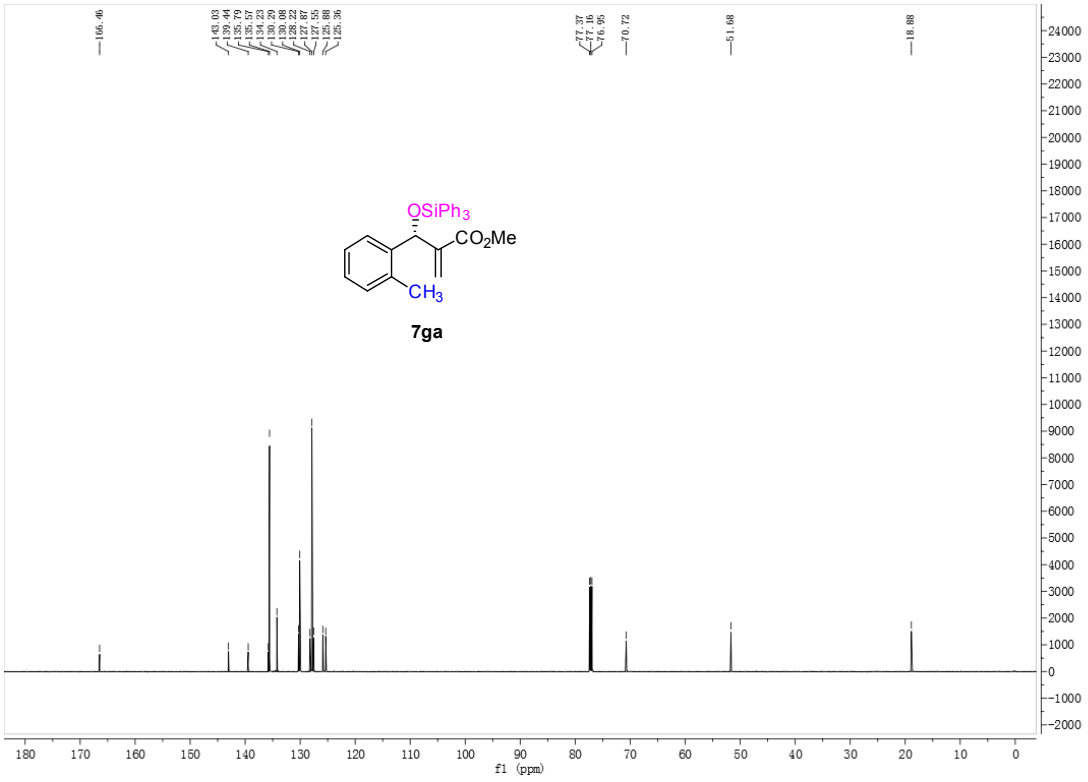


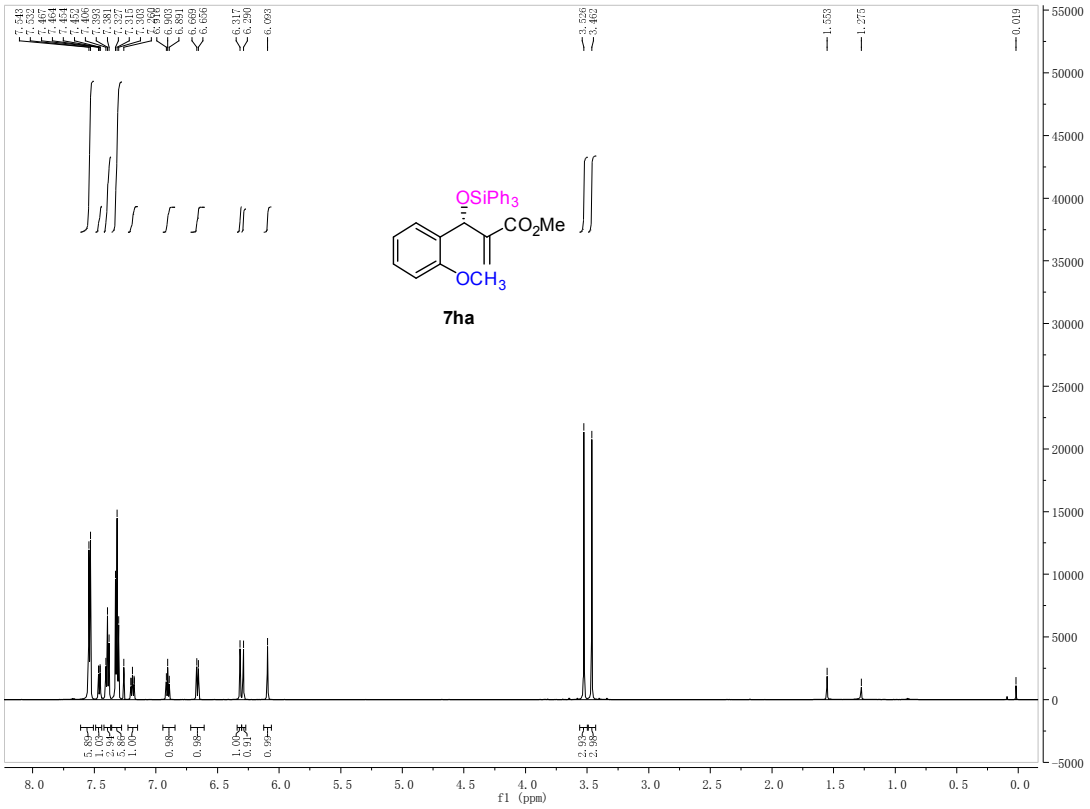
^{1}H-NMR for 7ga



^{13}C-NMR for 7ga



^{1}H-NMR for 7ha



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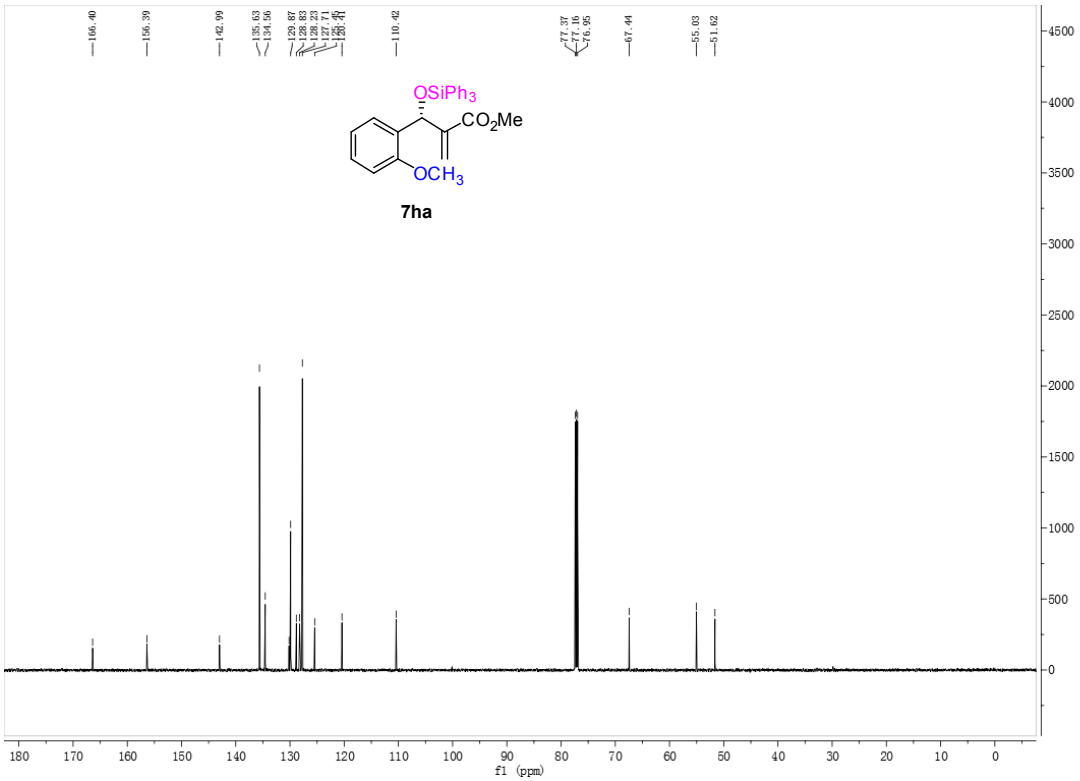
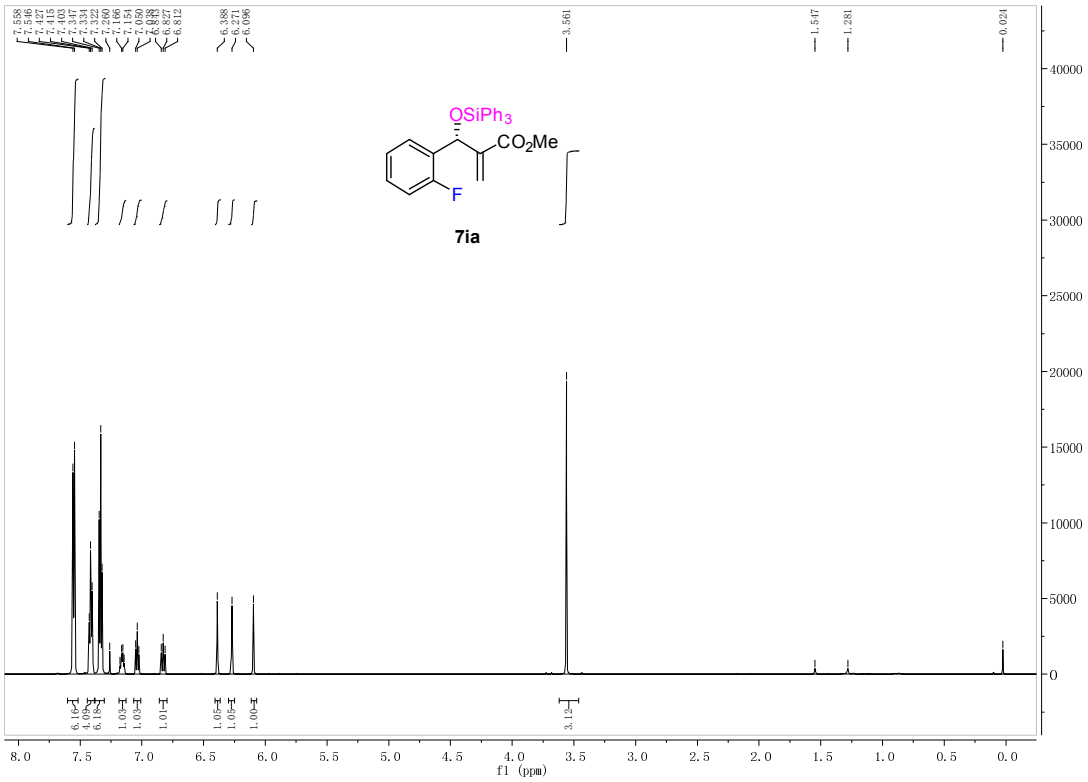


^{1}H-NMR for 7ia



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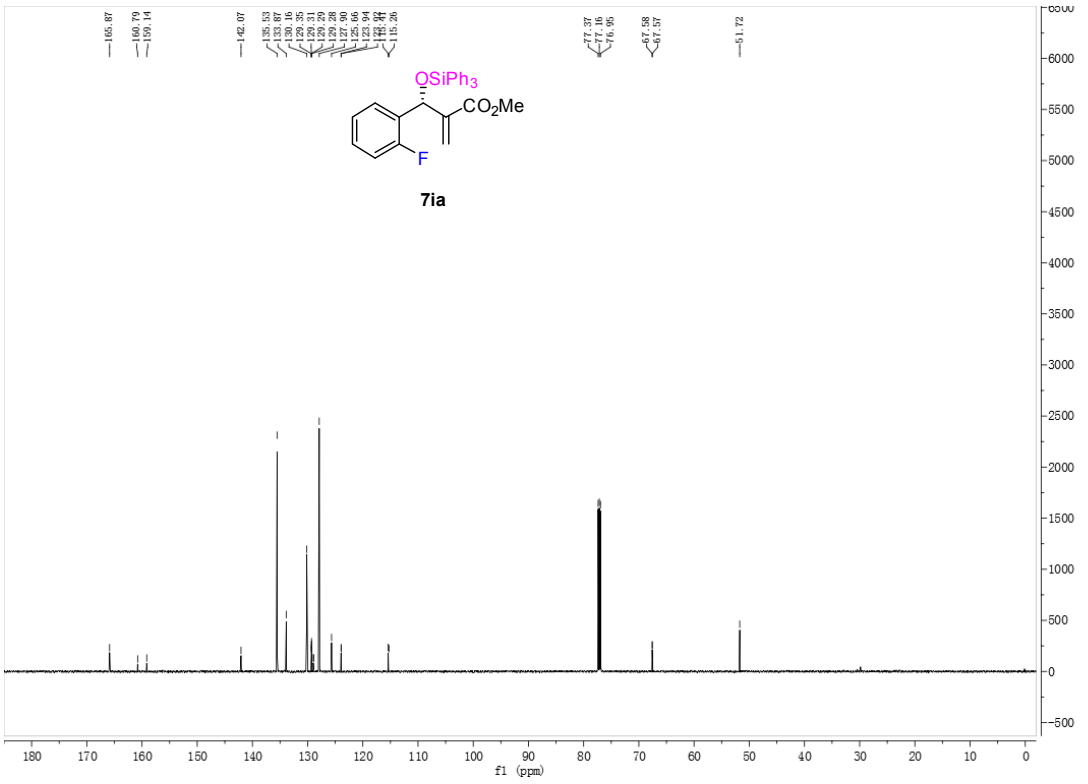
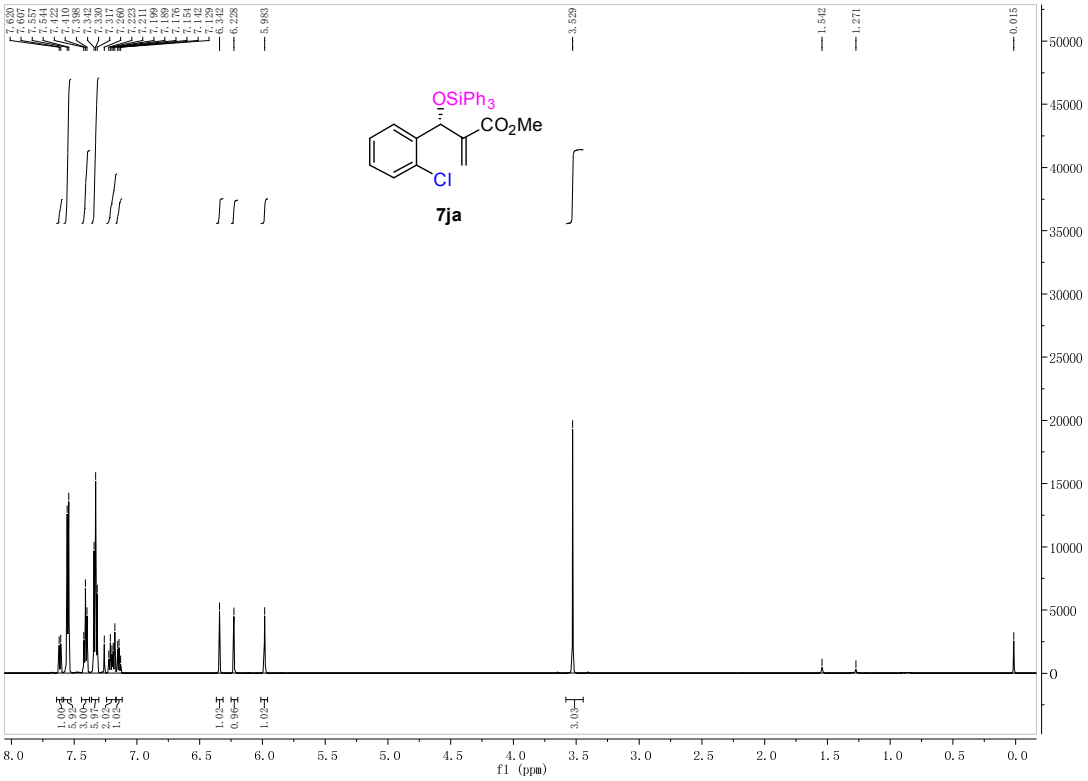


^{1}H-NMR for 7ja



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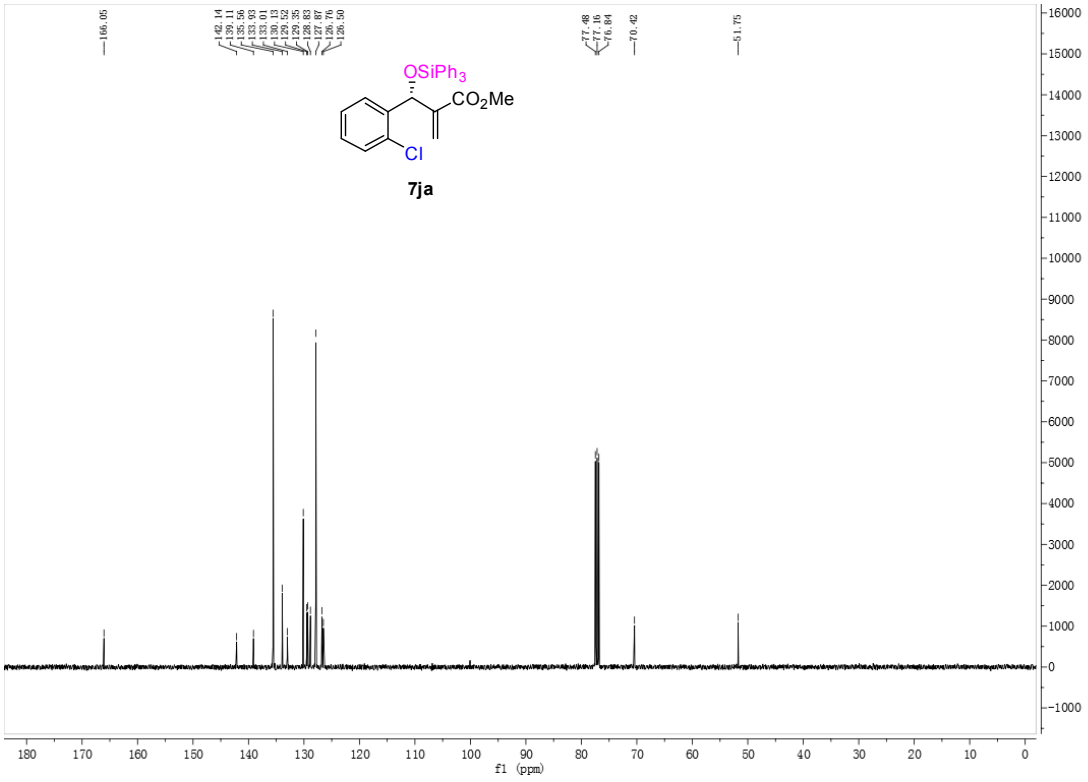
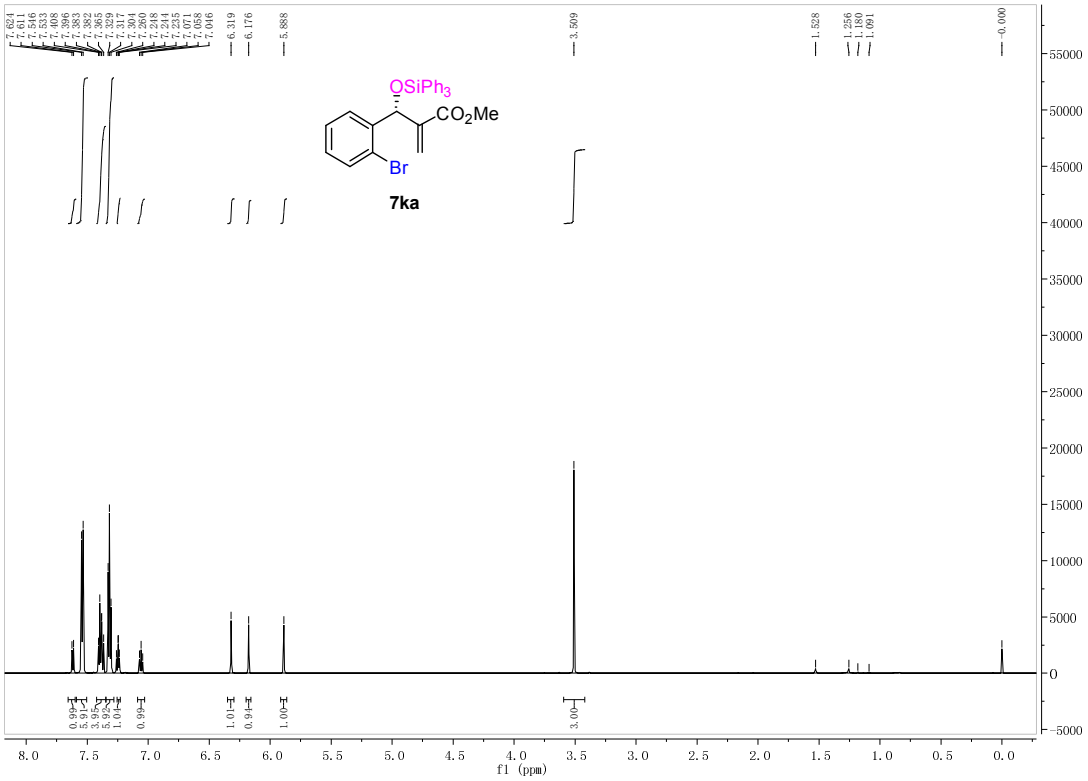


^{1}H-NMR for 7ka



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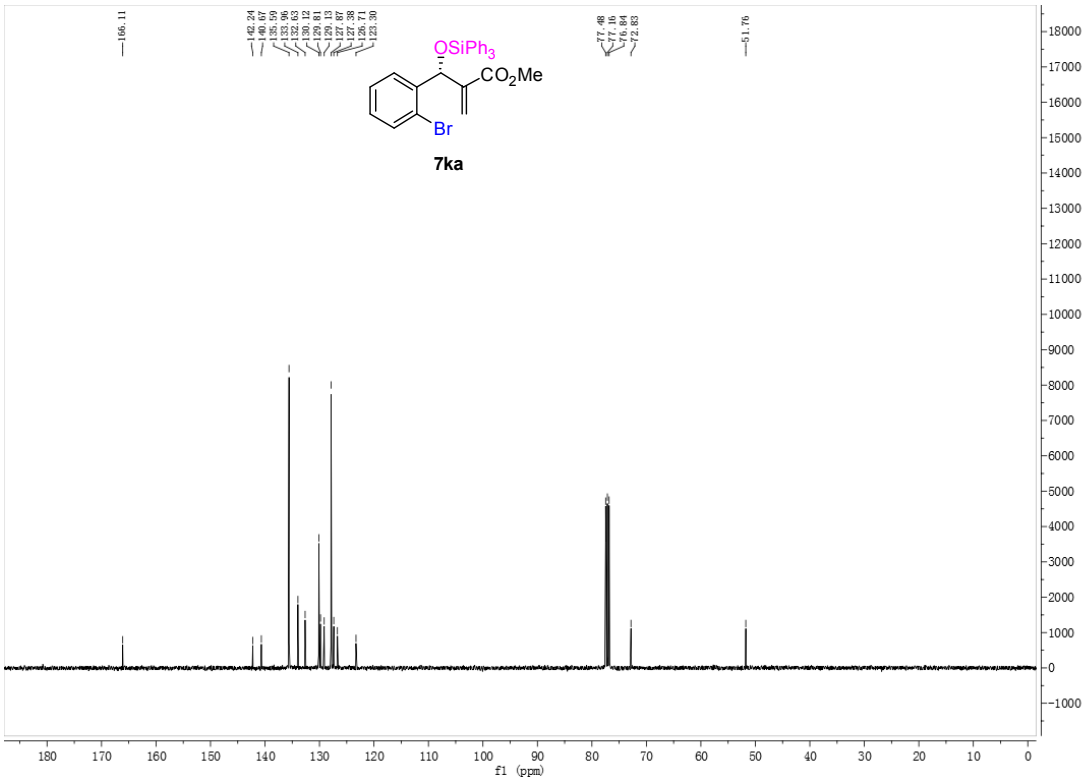

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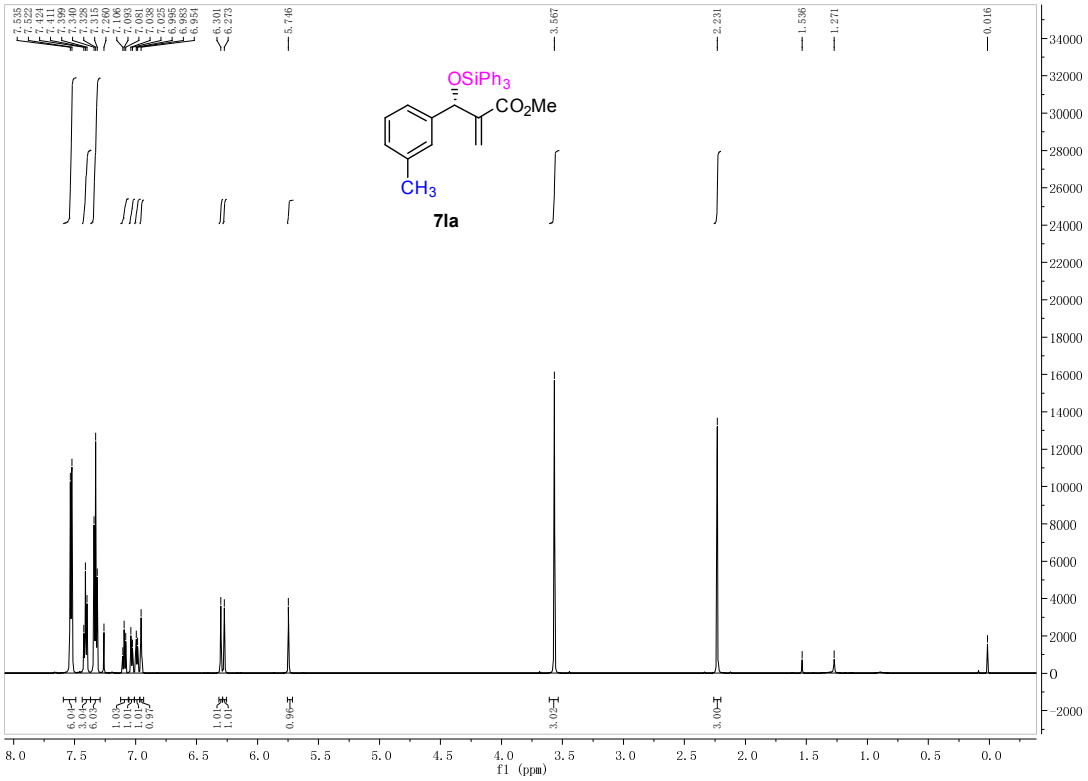

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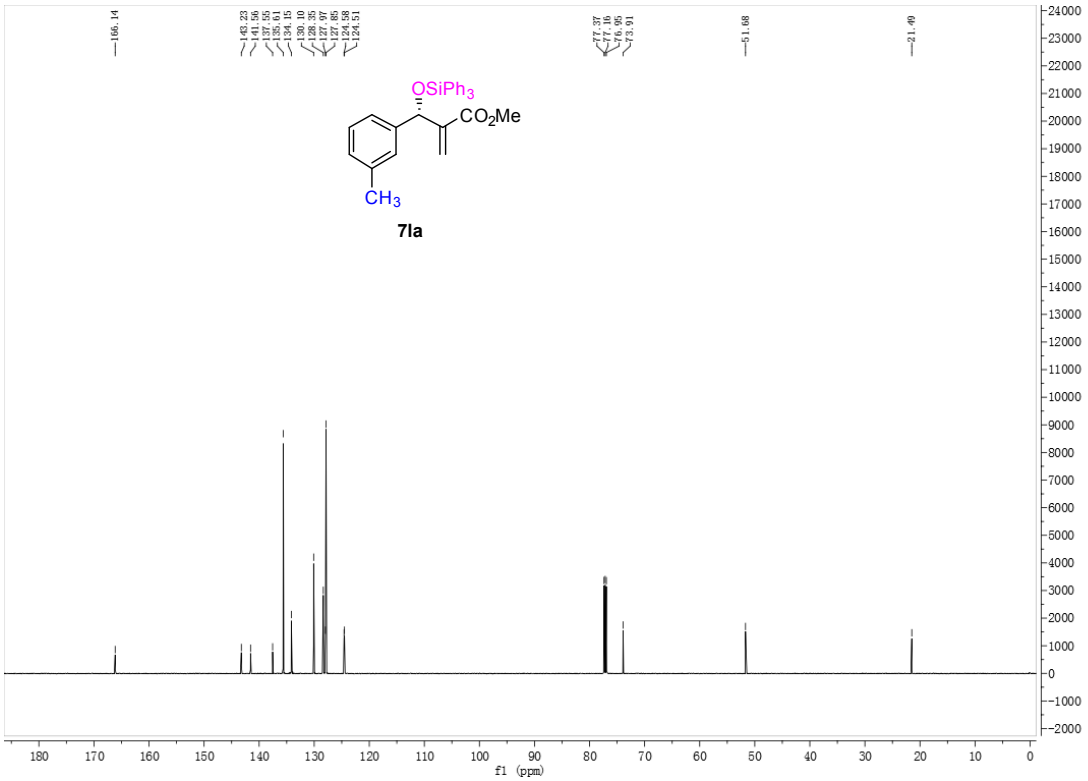
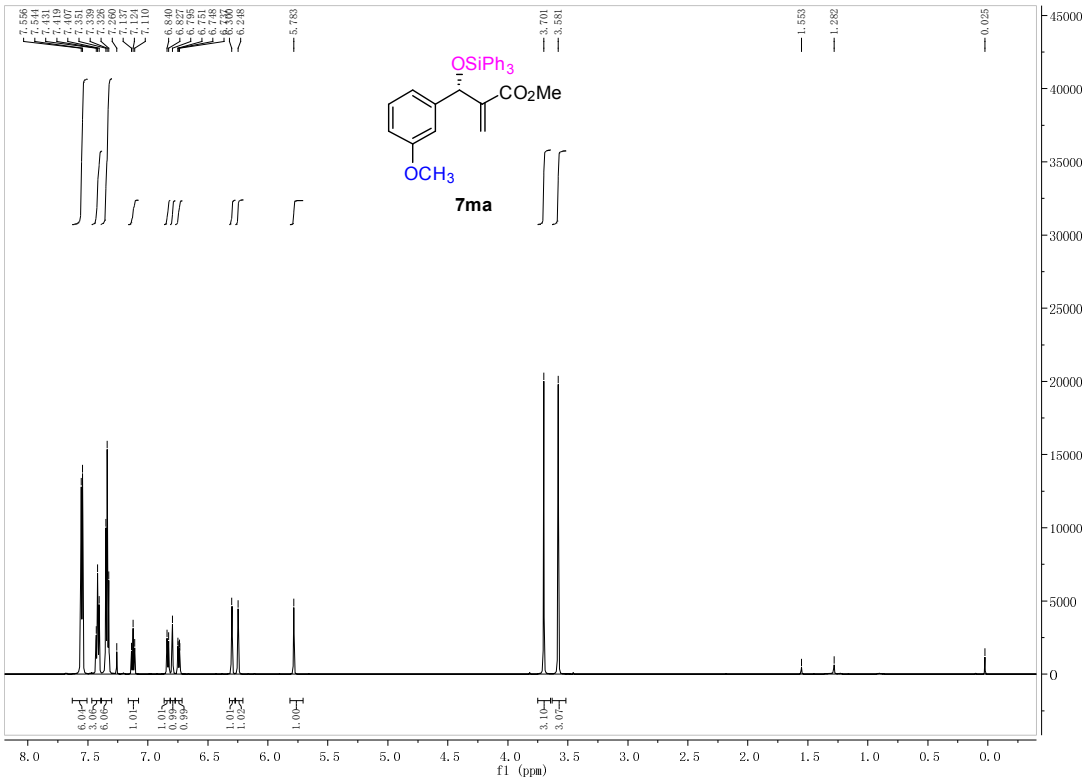


^{1}H-NMR for 7ma



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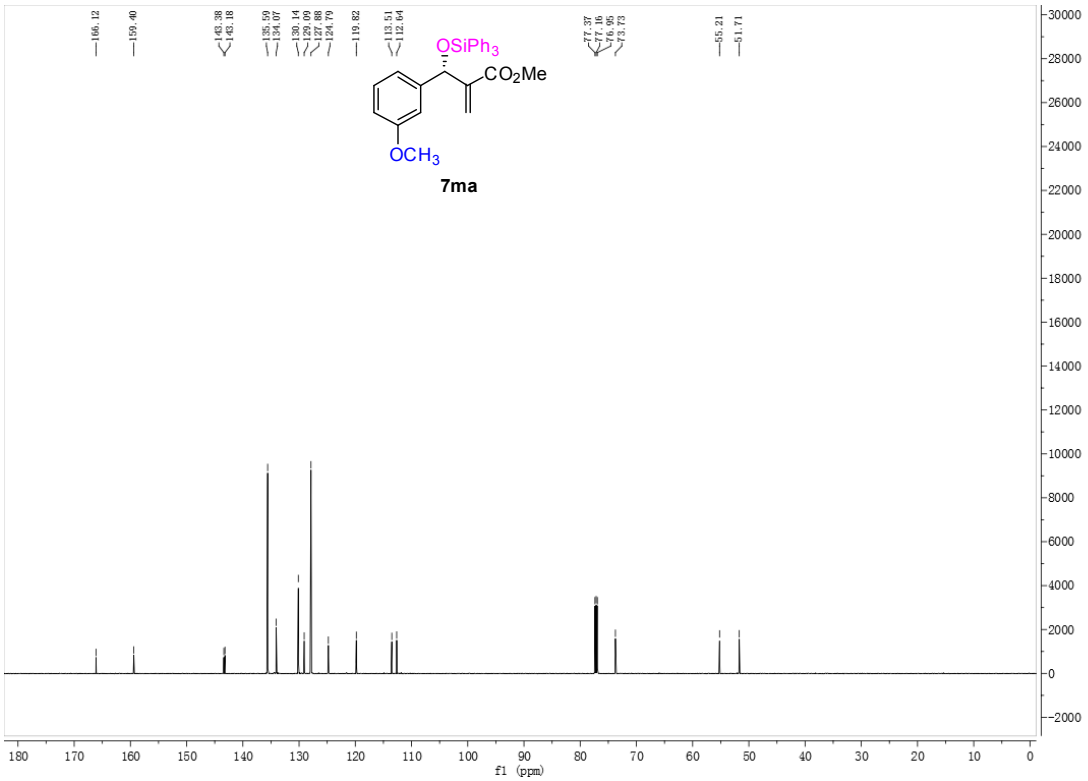

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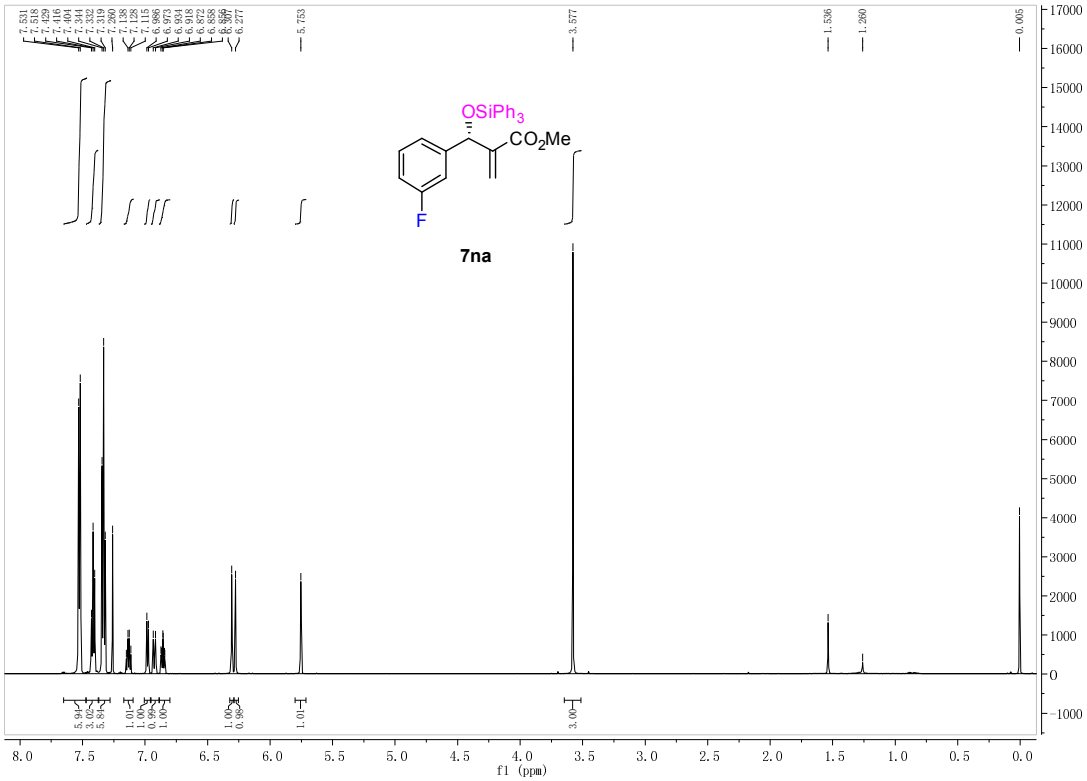

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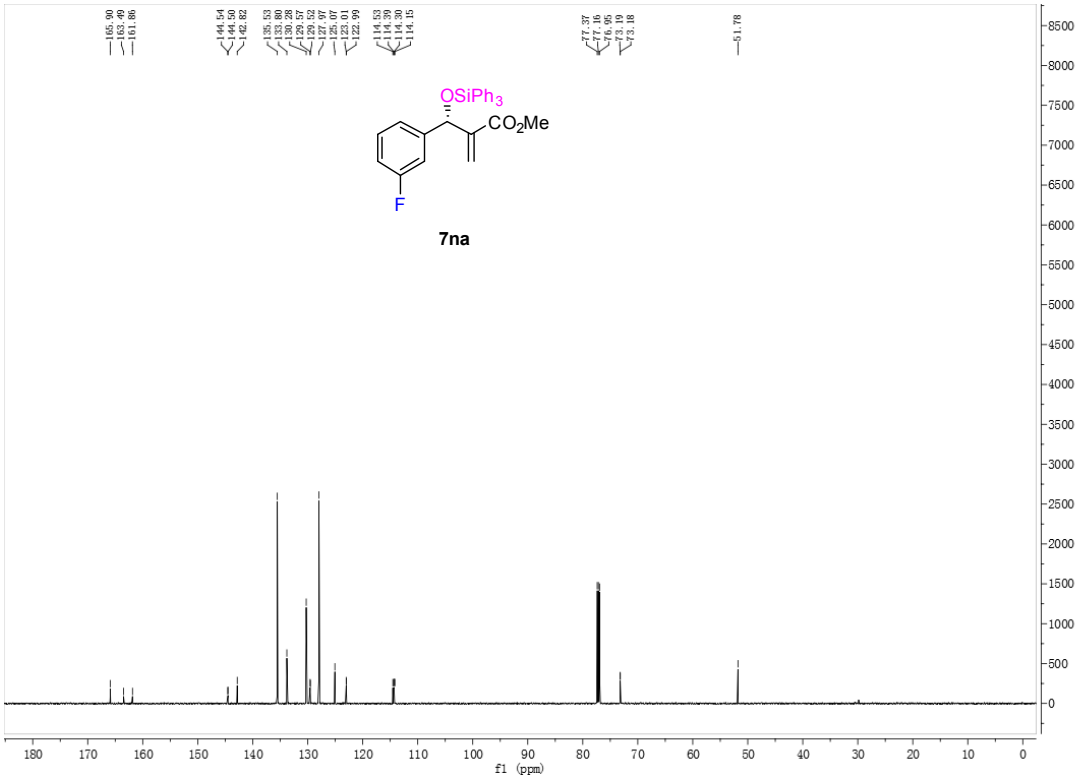
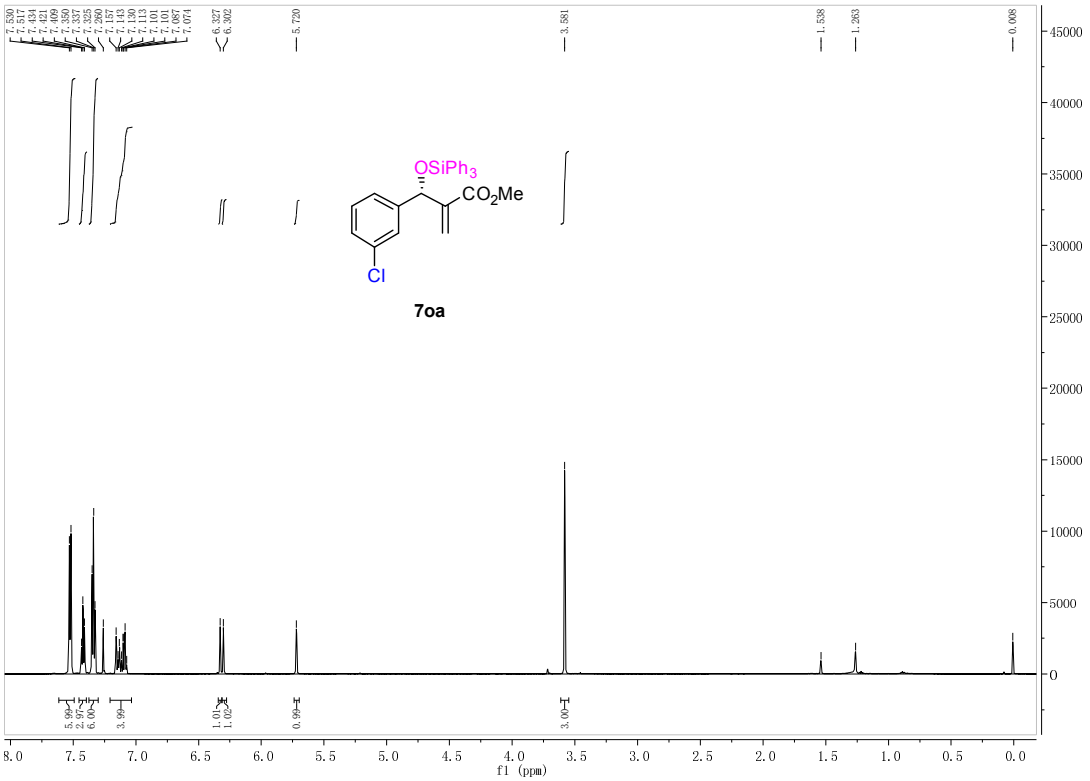


^{1}H-NMR for 7oa



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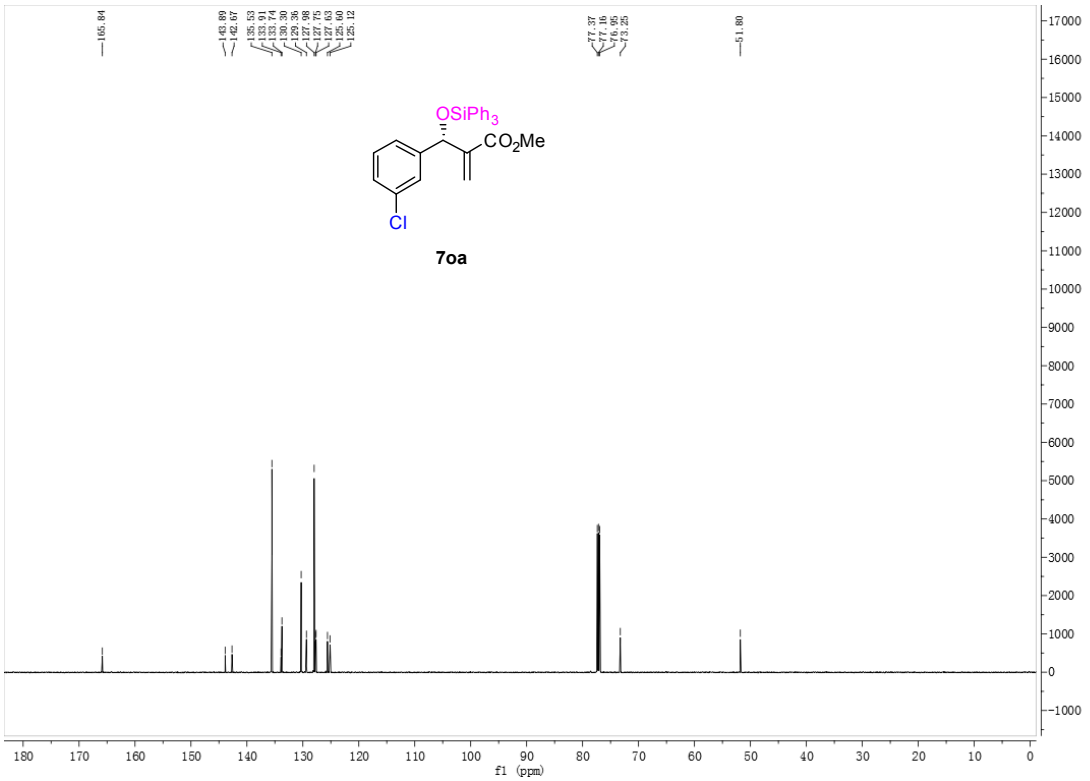
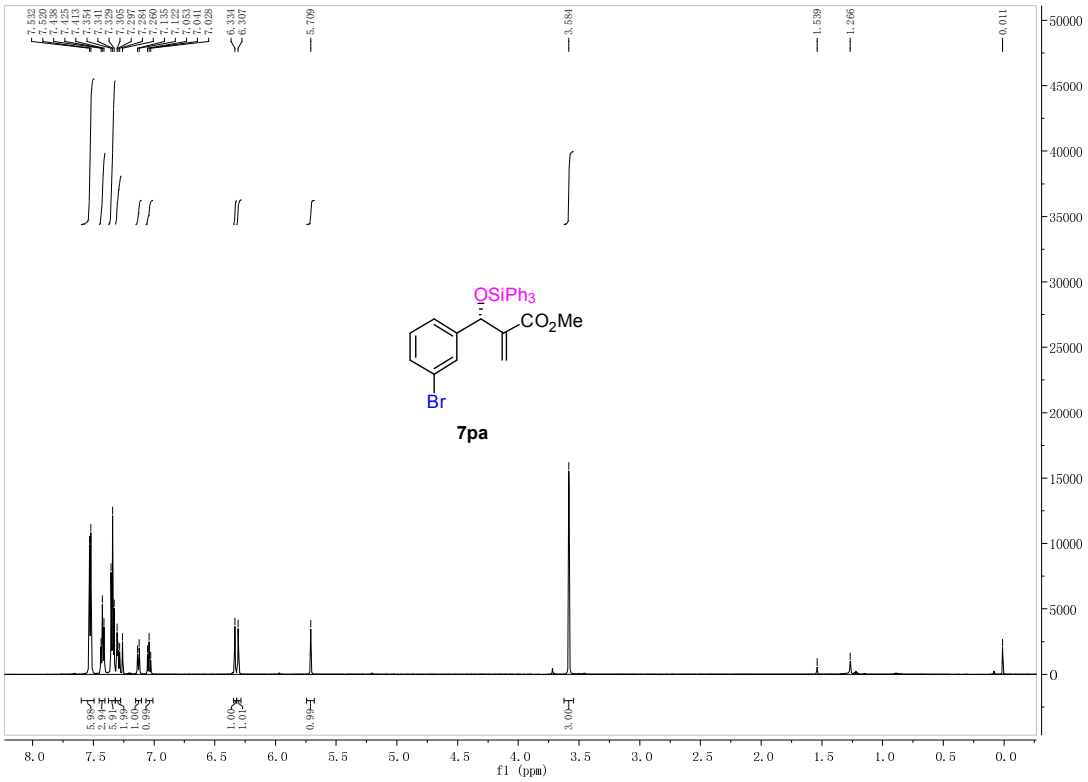
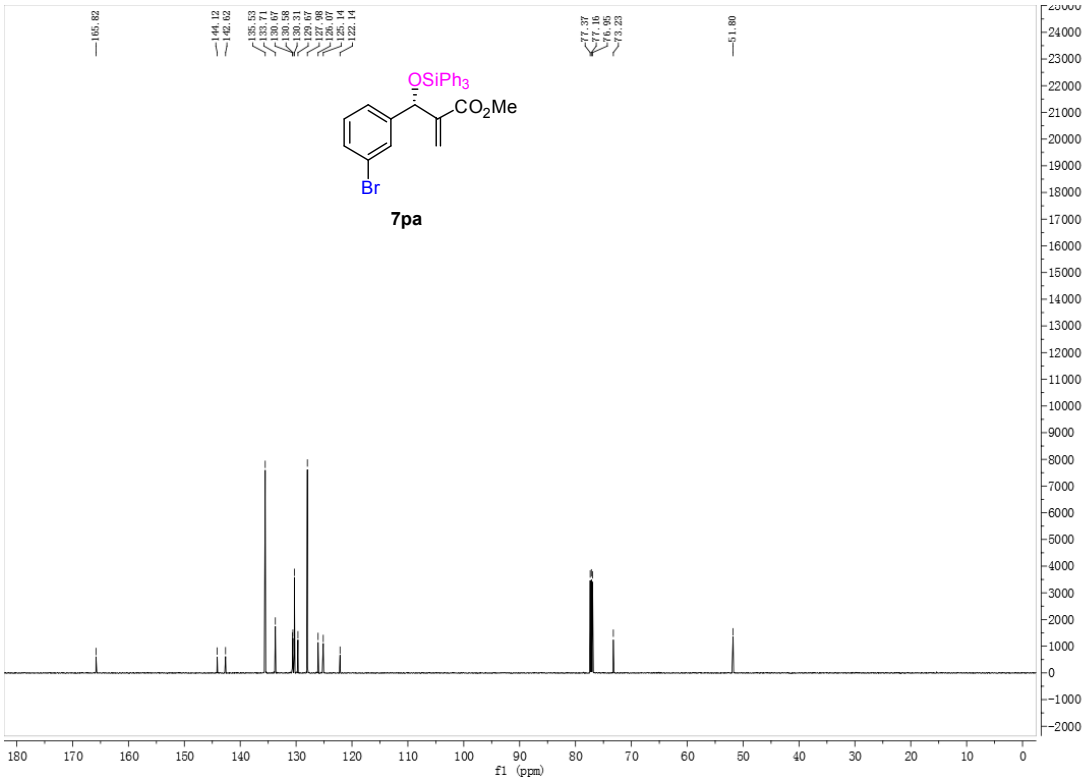


^{1}H-NMR for 7pa



^{13}C-NMR for 7pa



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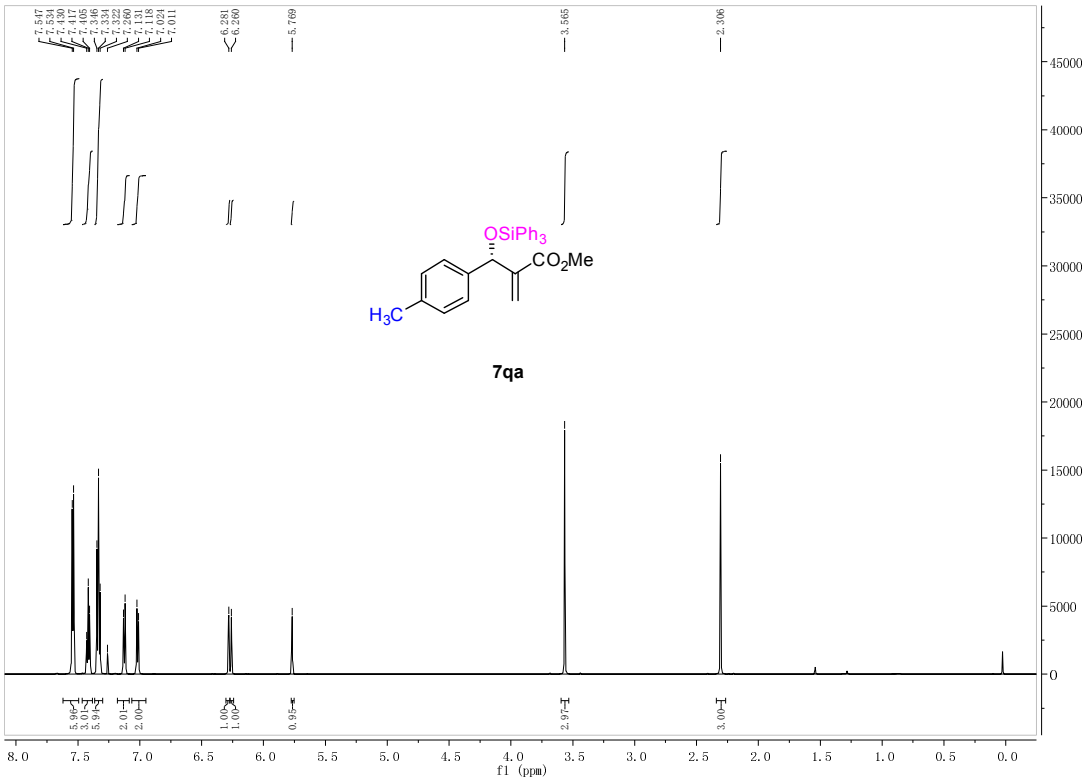

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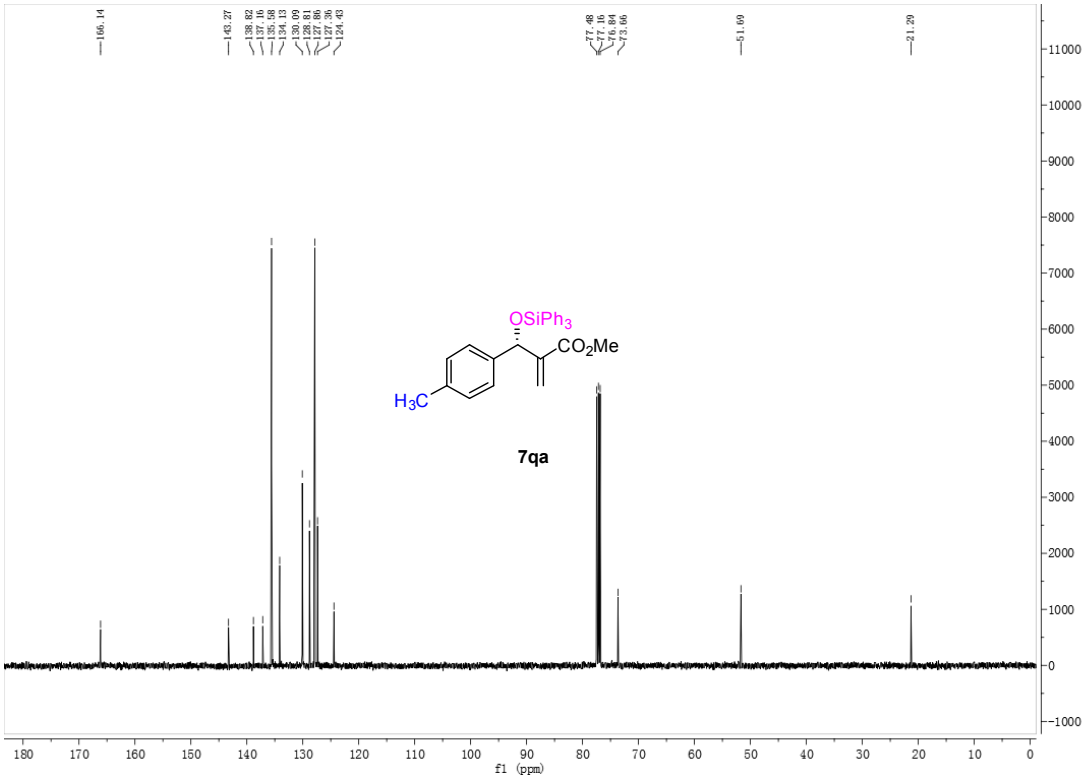
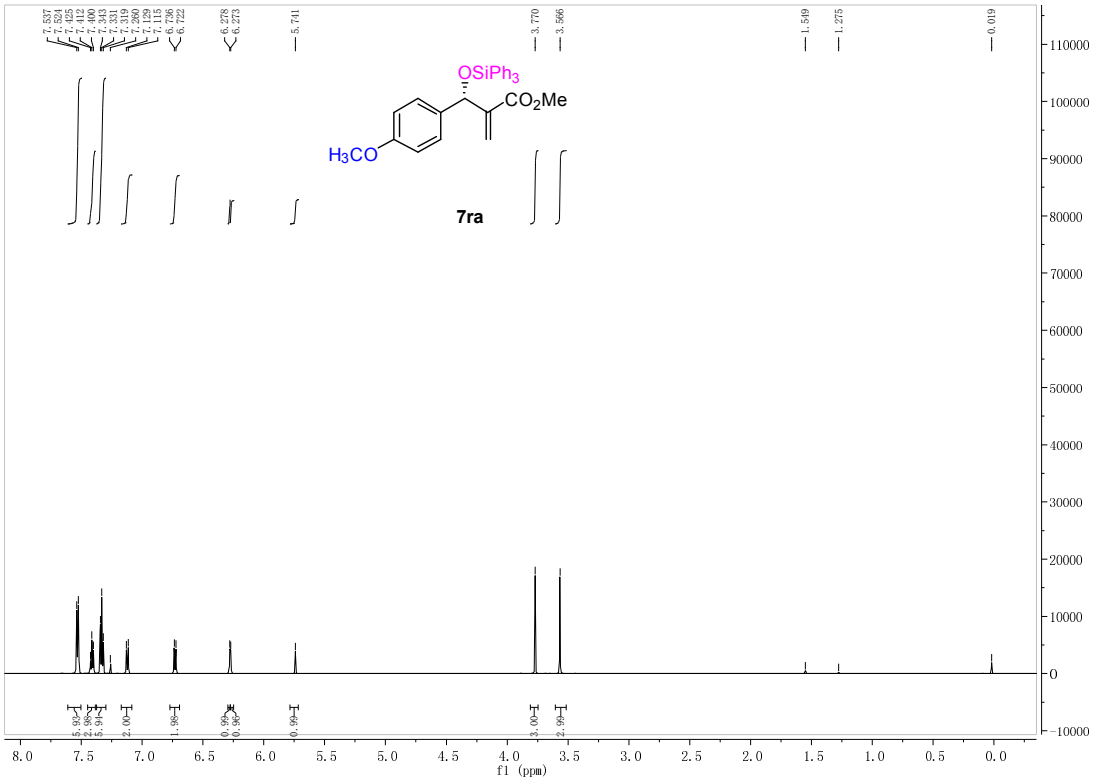


^{1}H-NMR for 7ra



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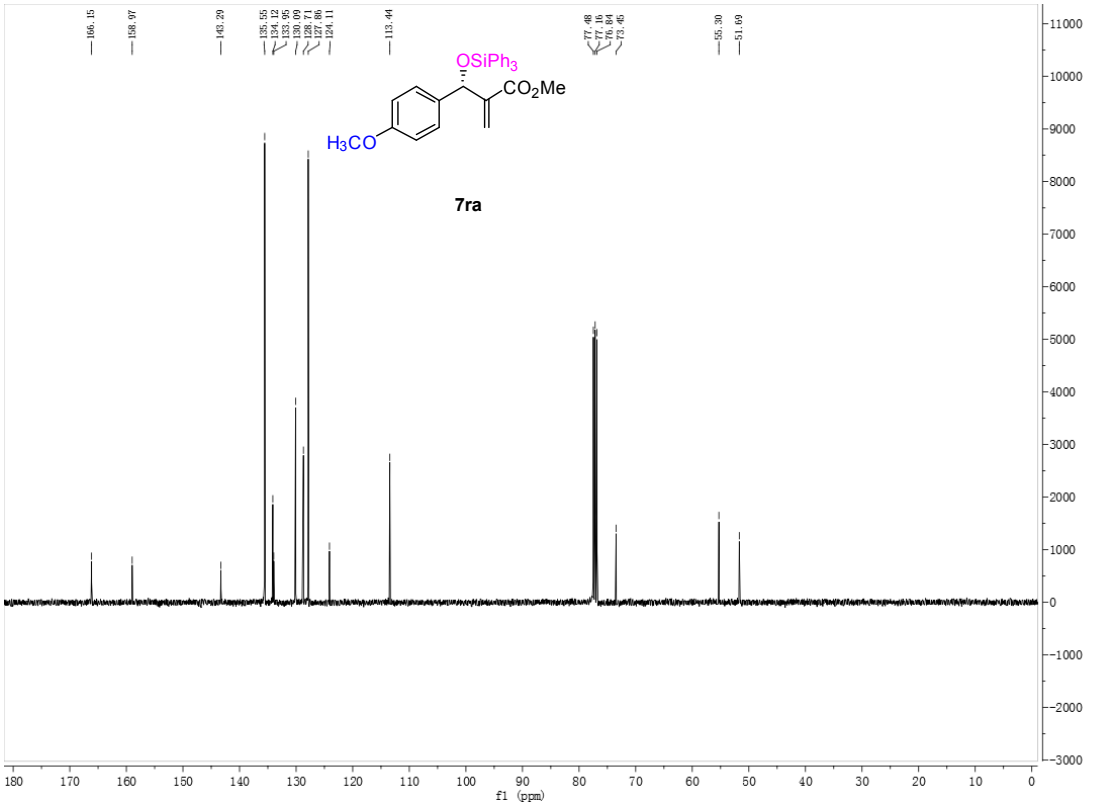
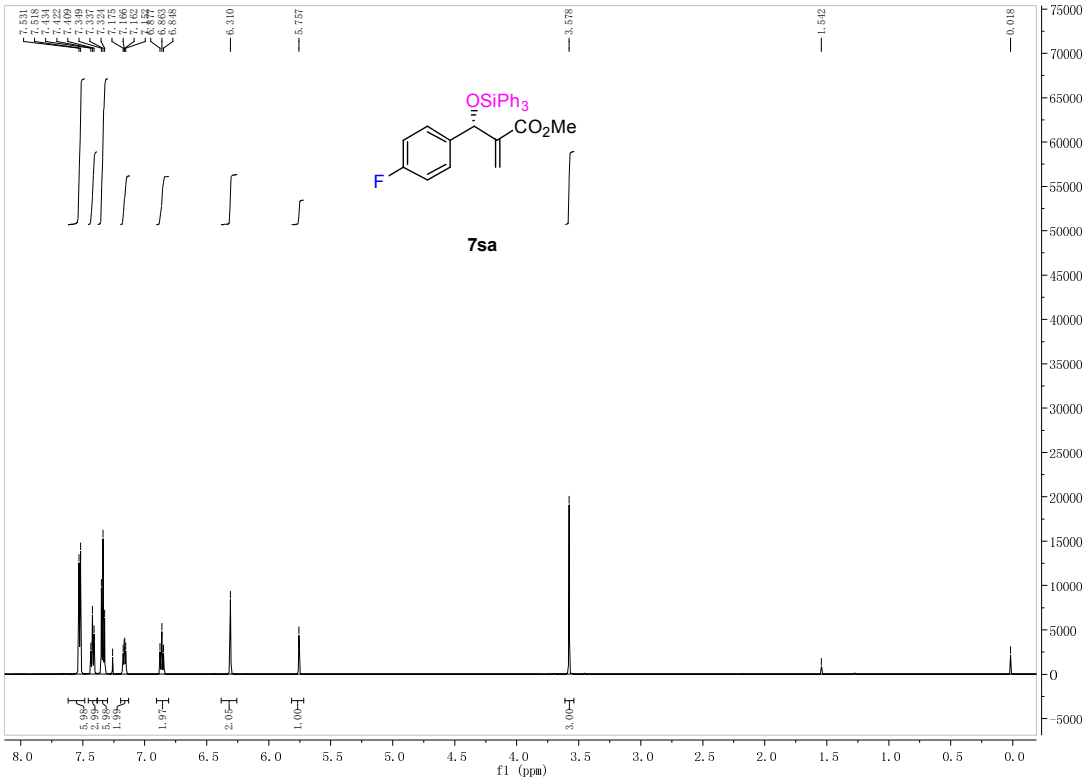


^{1}H-NMR for 7sa



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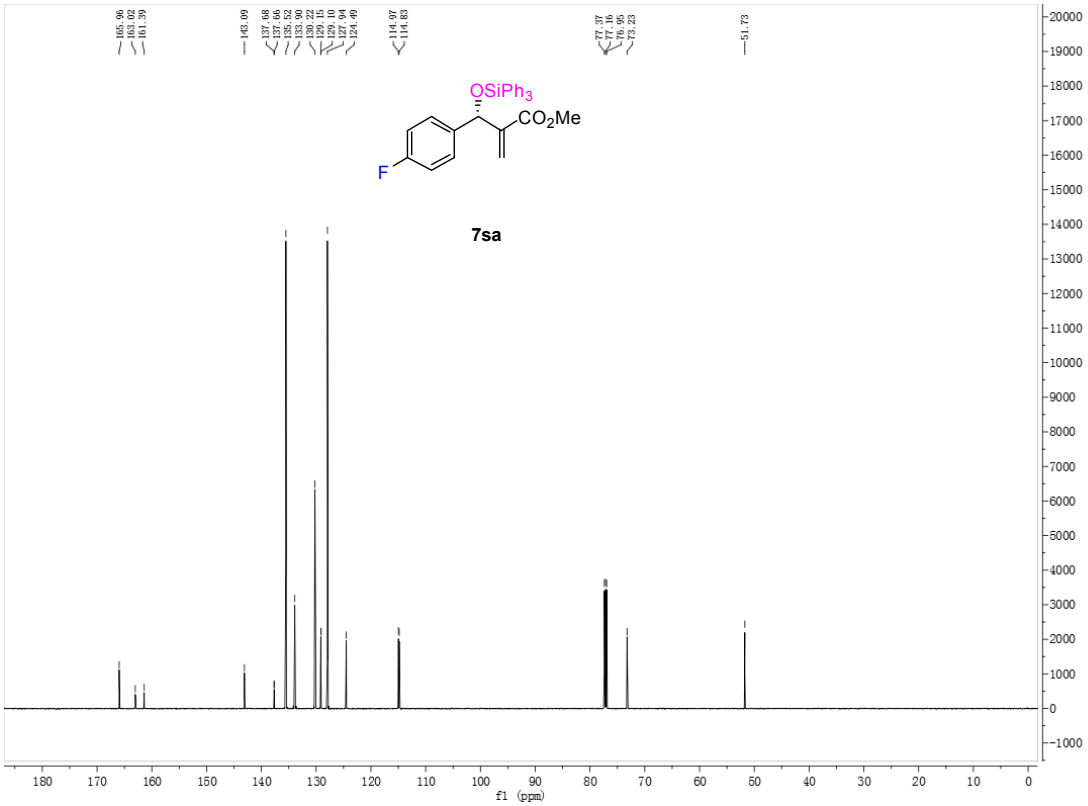
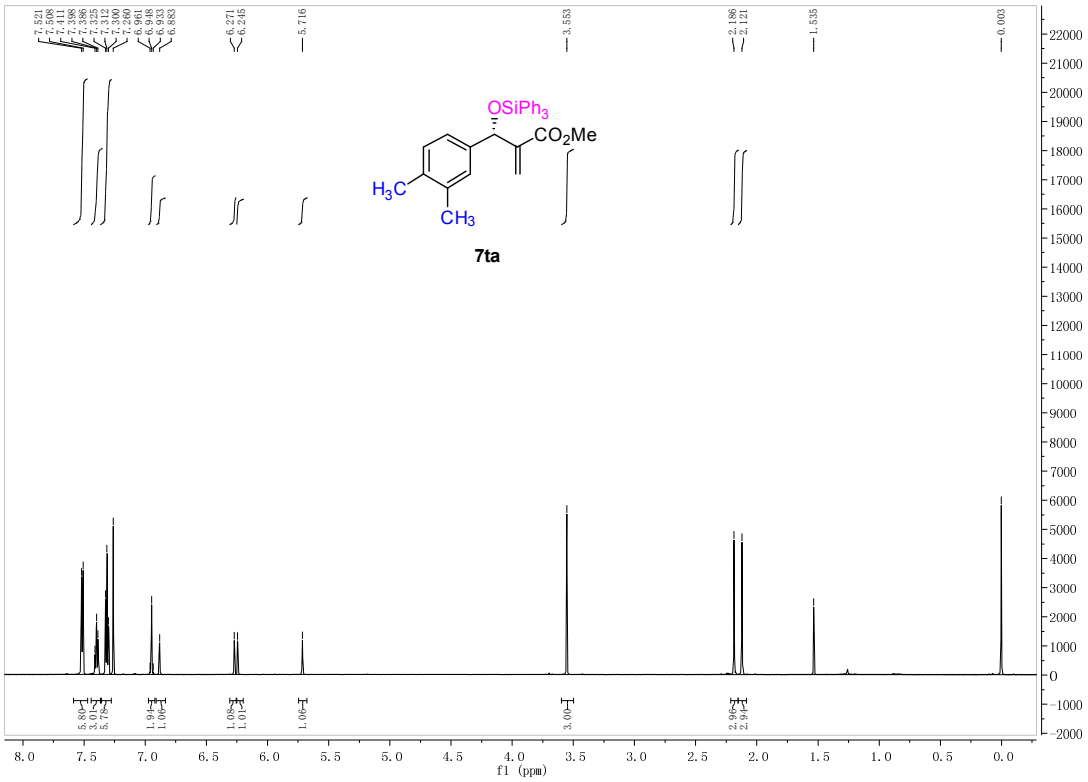


^{1}H-NMR for 7ta



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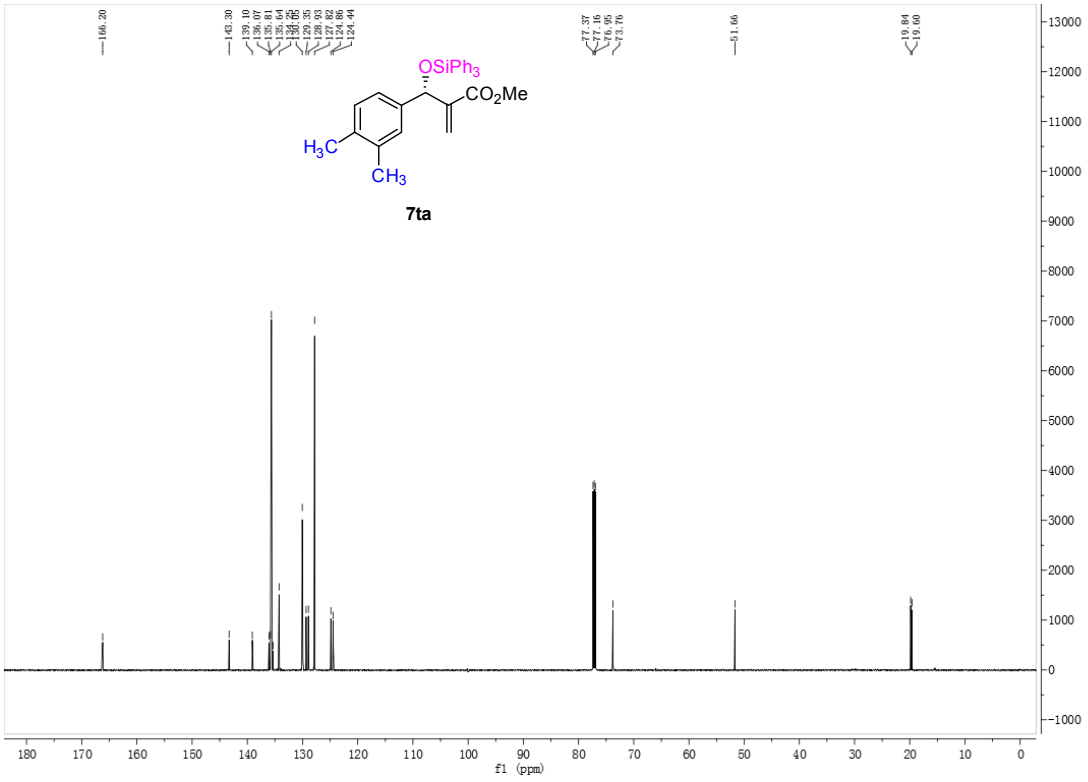

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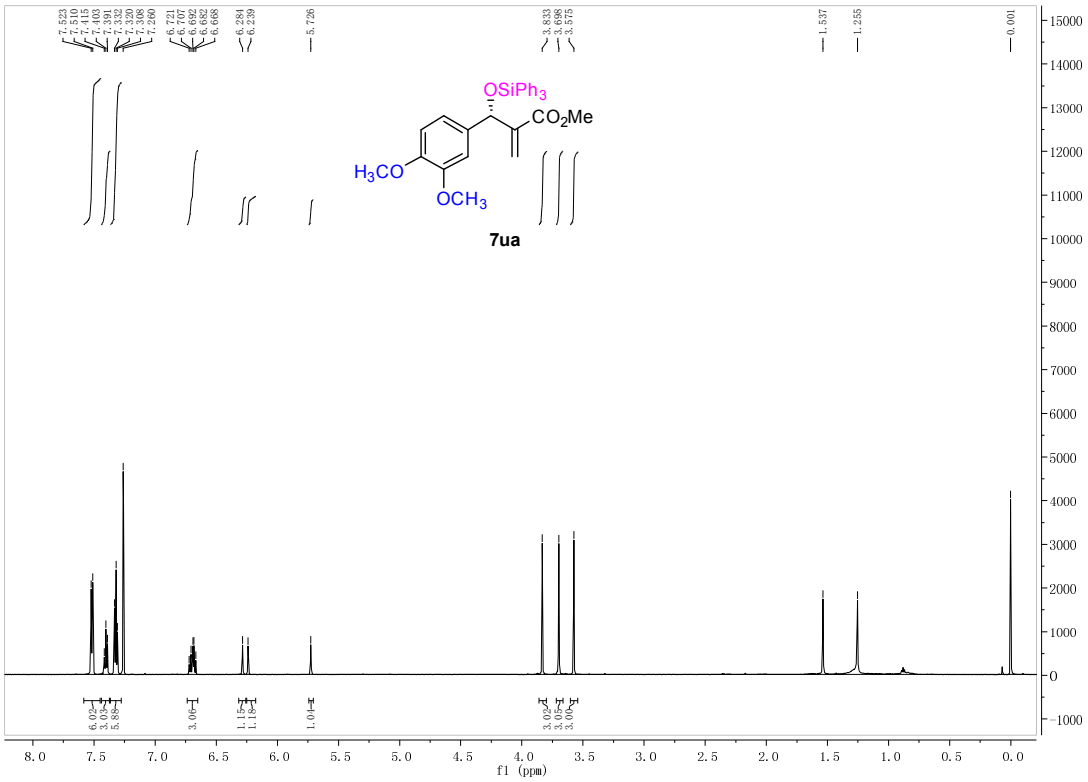

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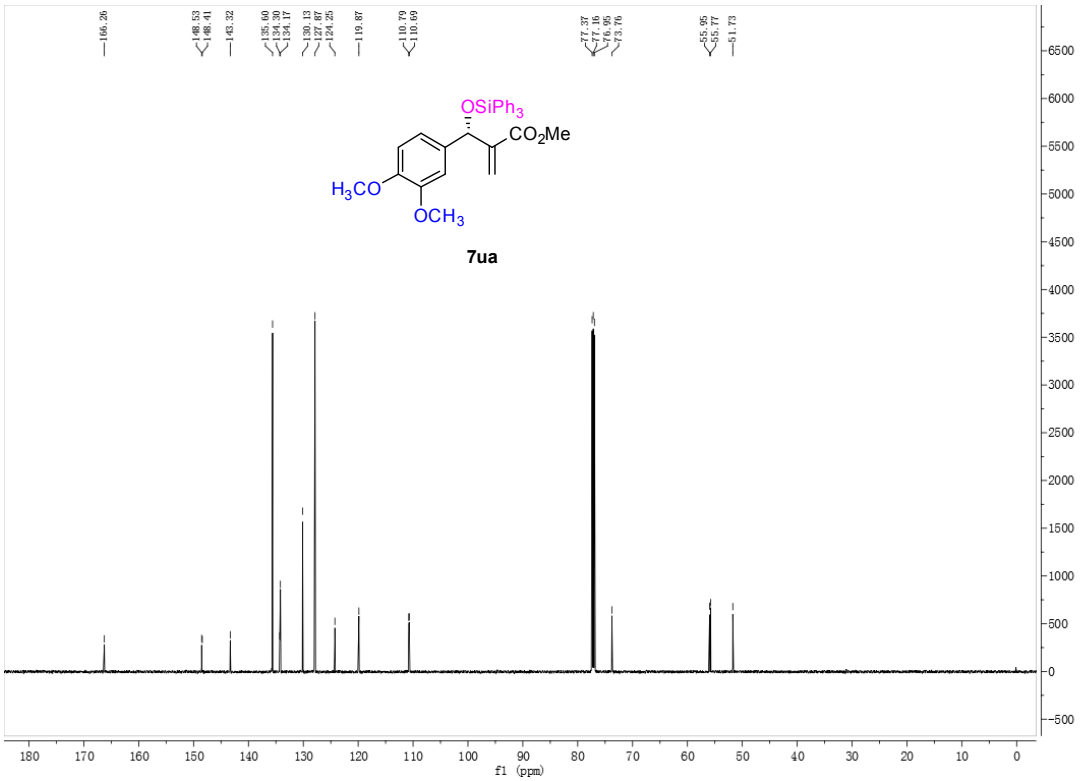

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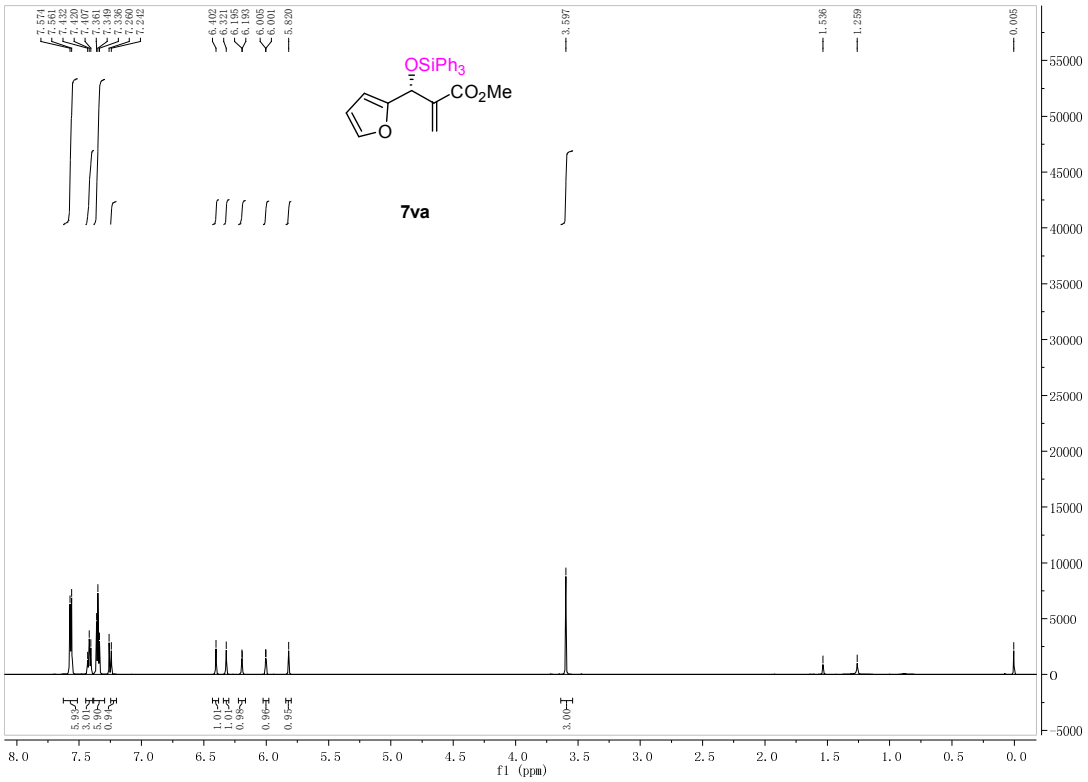

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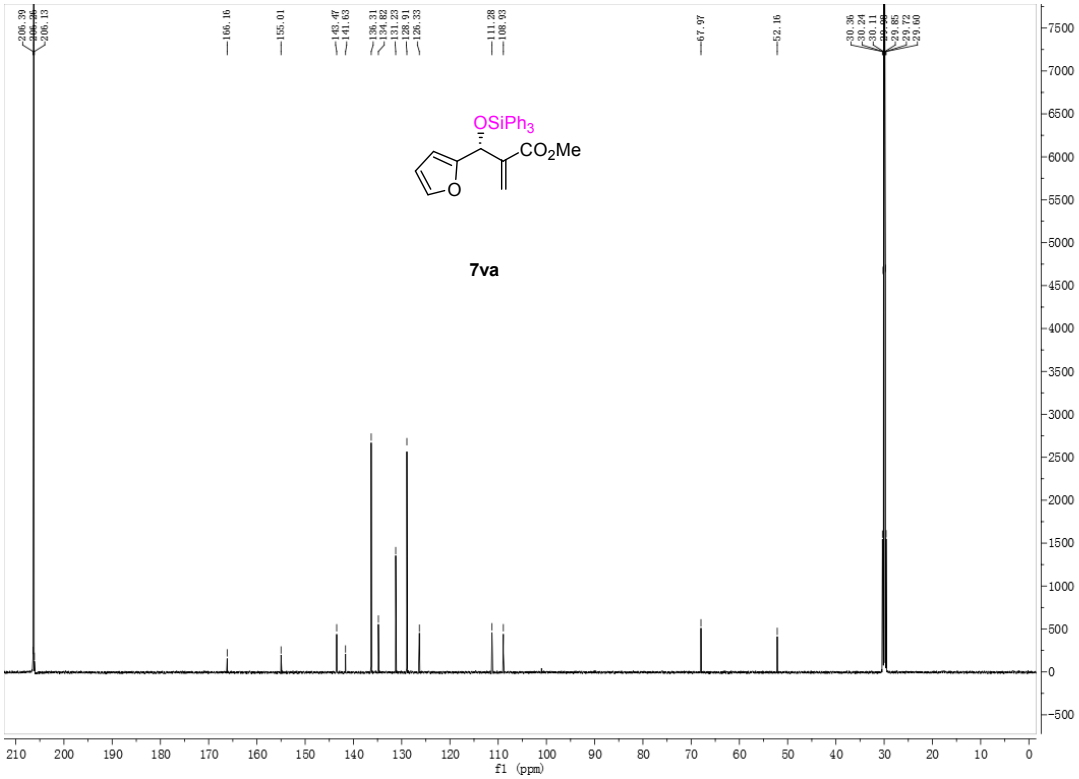
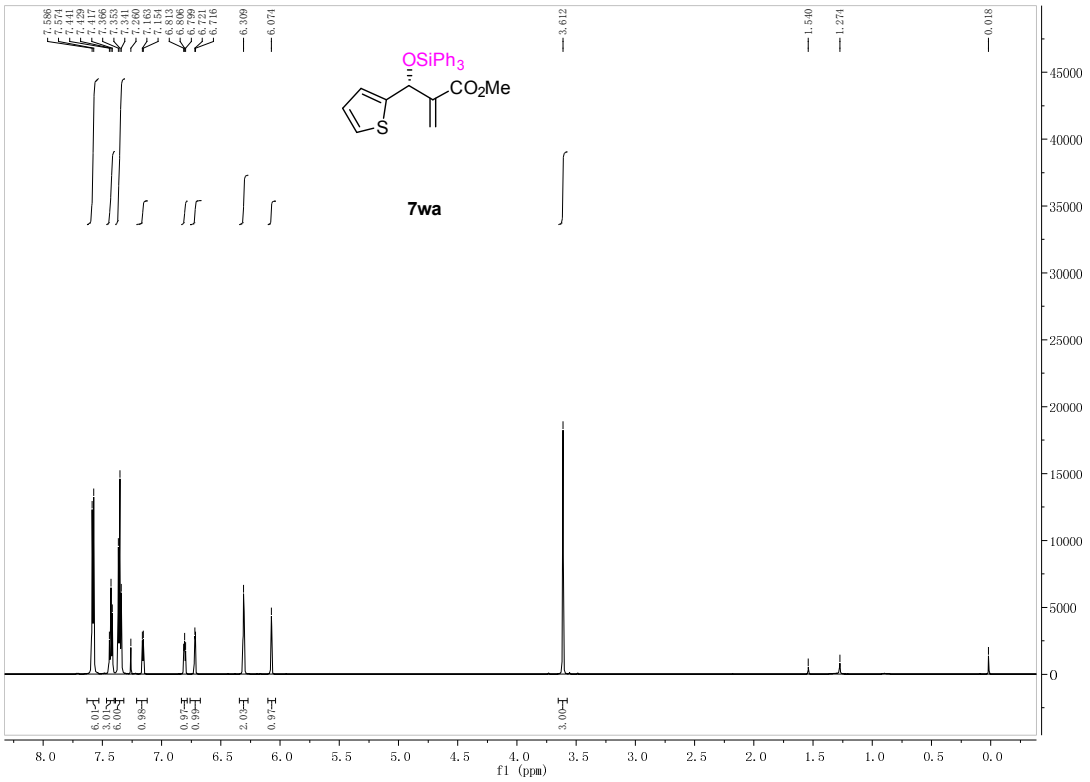


^{1}H-NMR for 7wa



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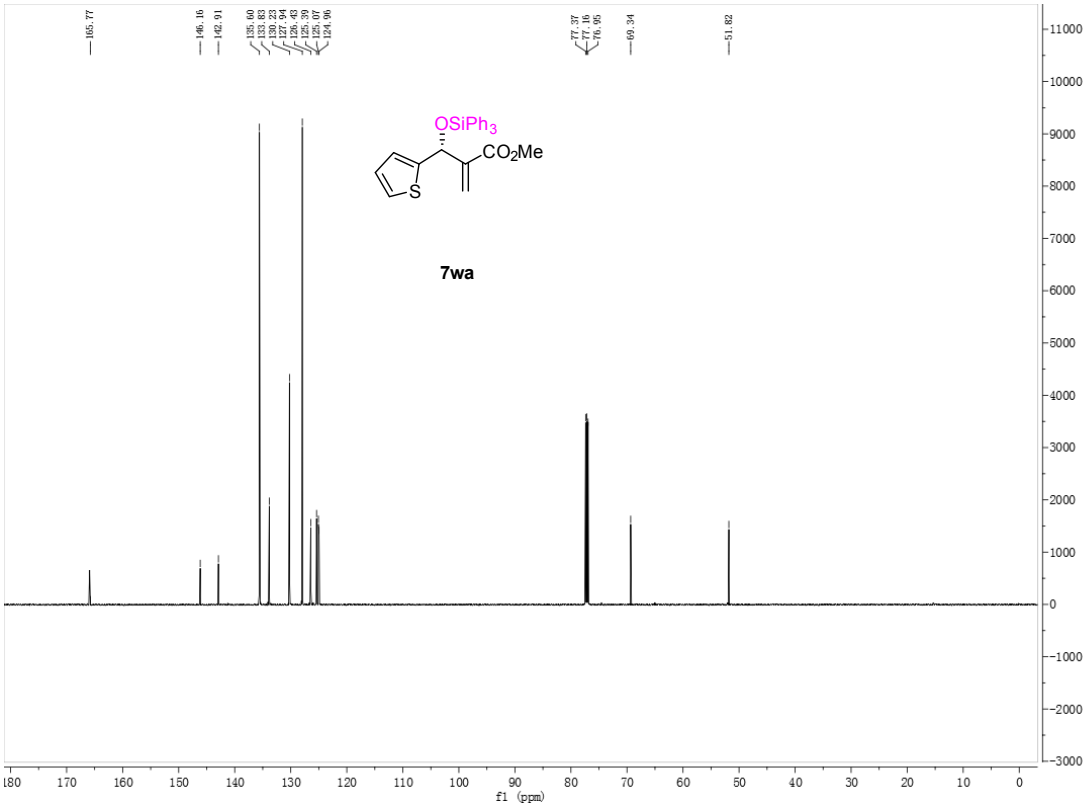
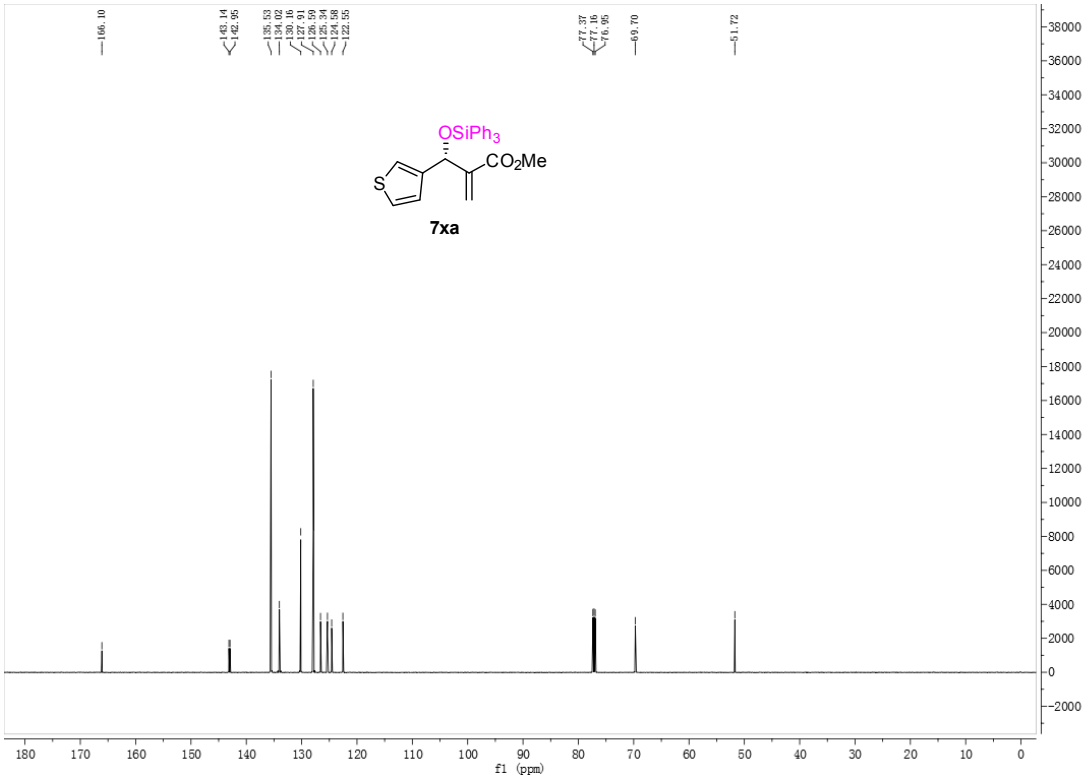
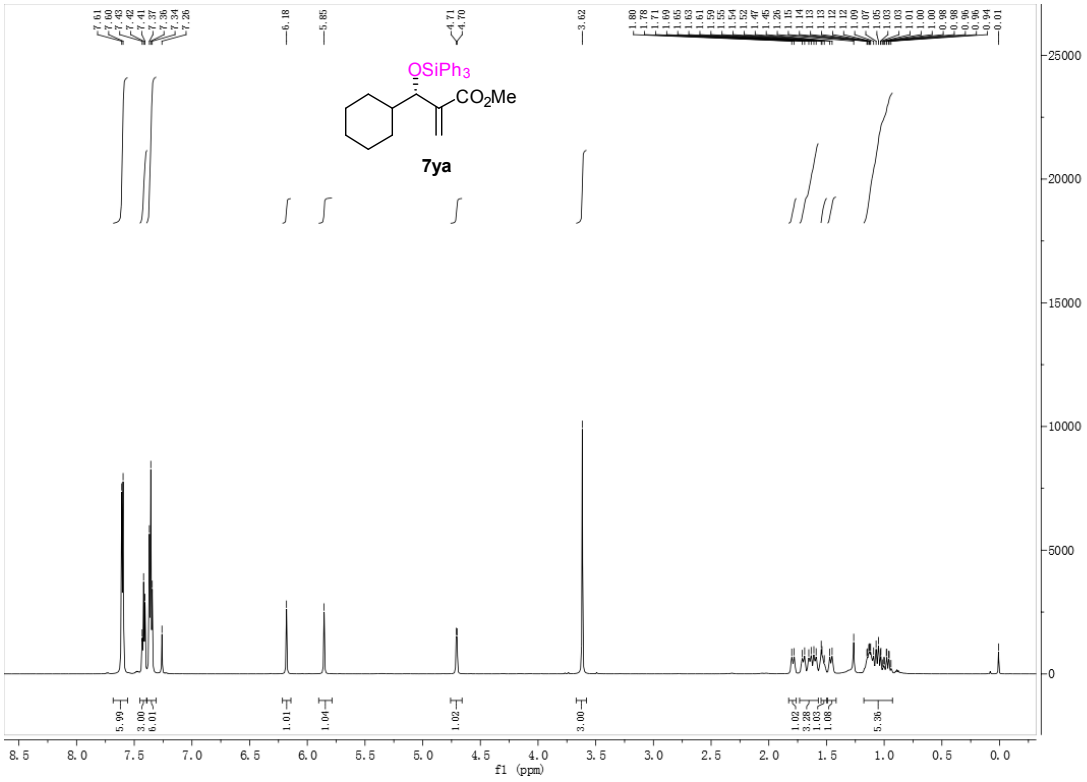


^{1}H-NMR for 7xa

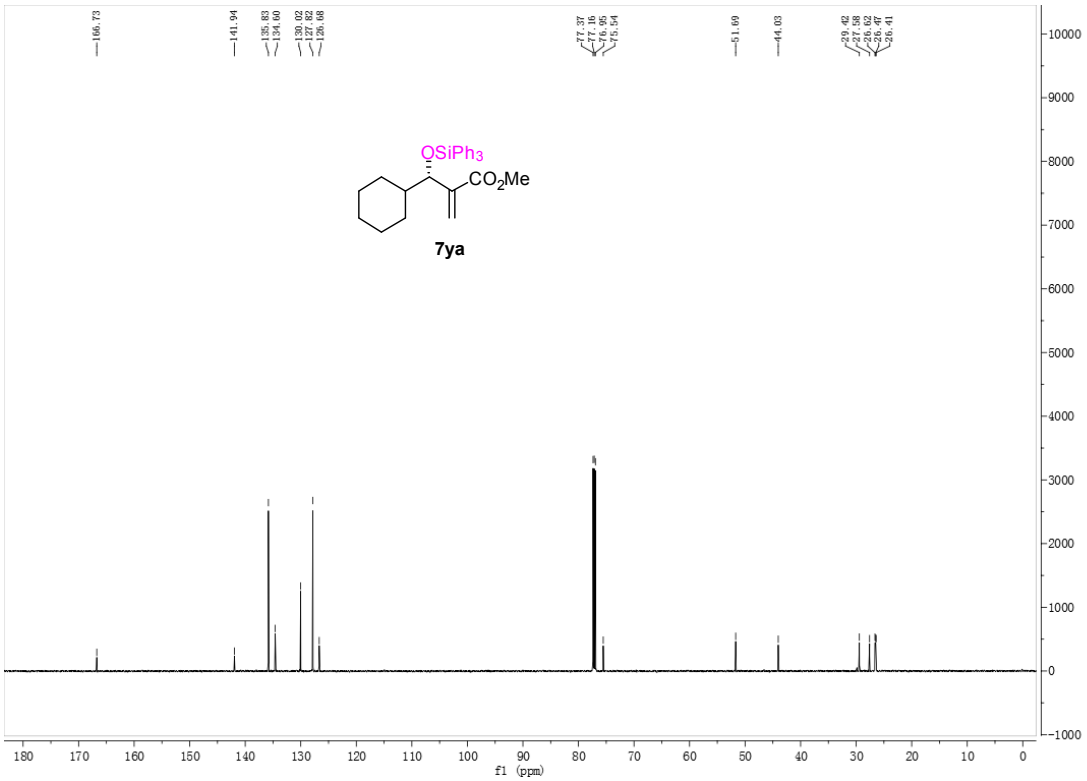
^{13}C-NMR for 7xa



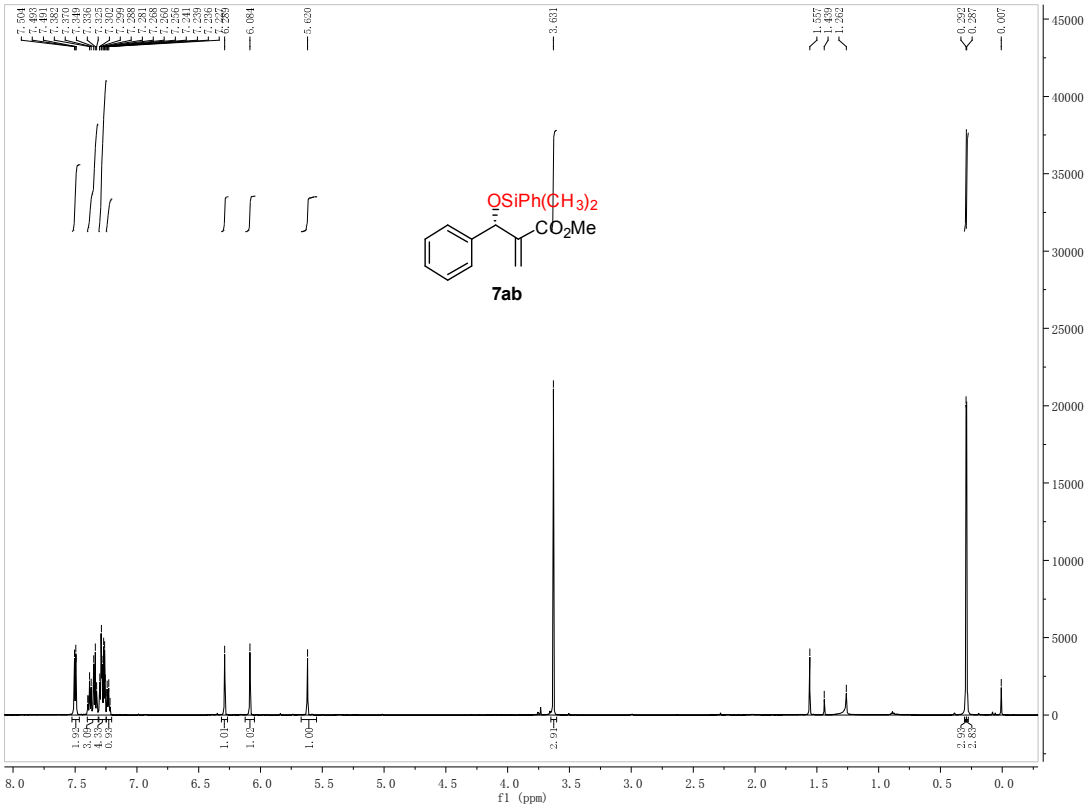
^{1}H-NMR for 7ya



^{13}C-NMR for 7ya



^{1}H-NMR for 7ab



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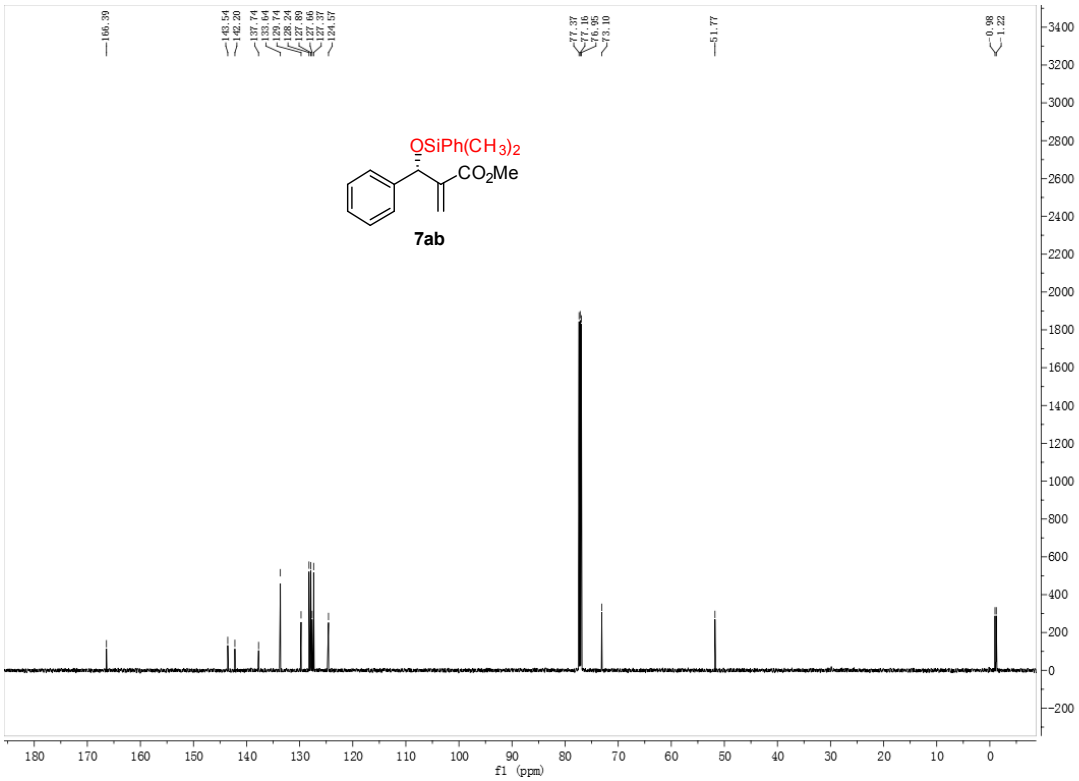
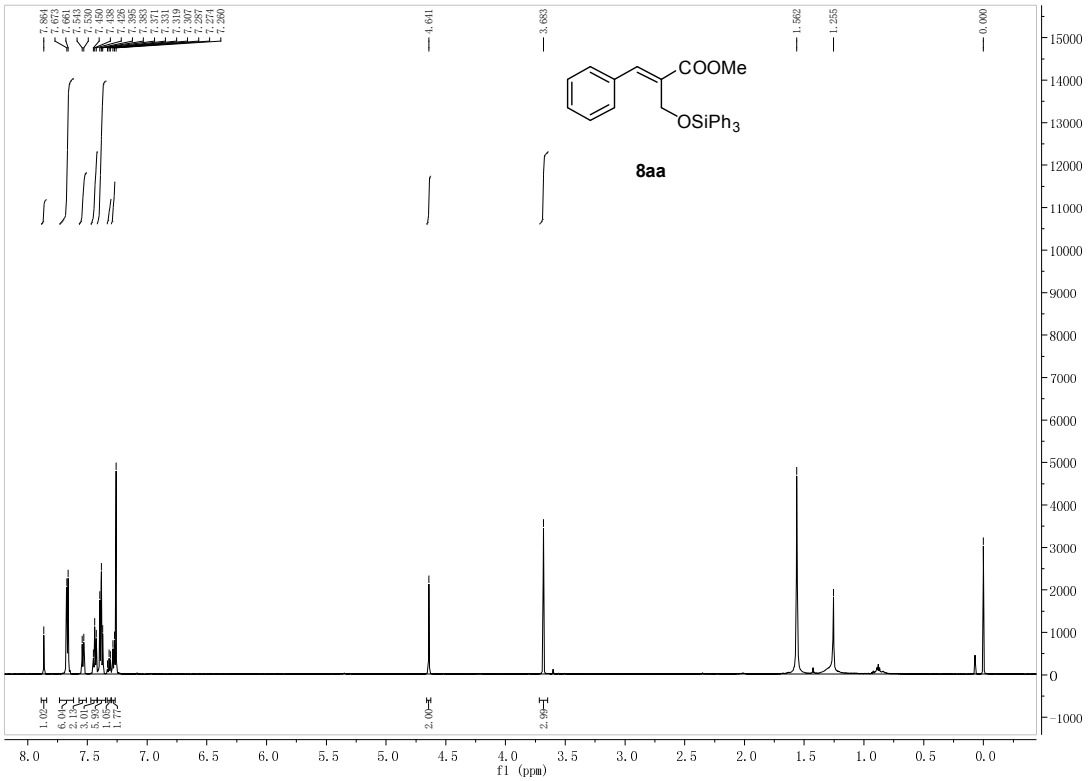


^{1}H-NMR for 8aa



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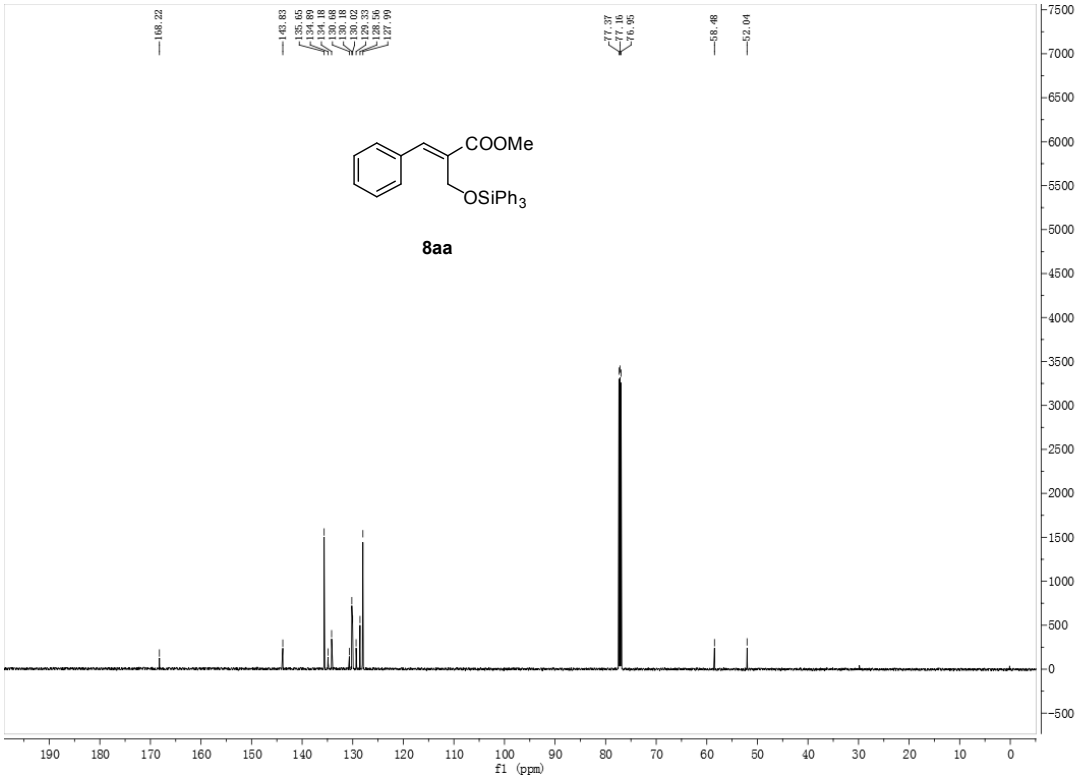
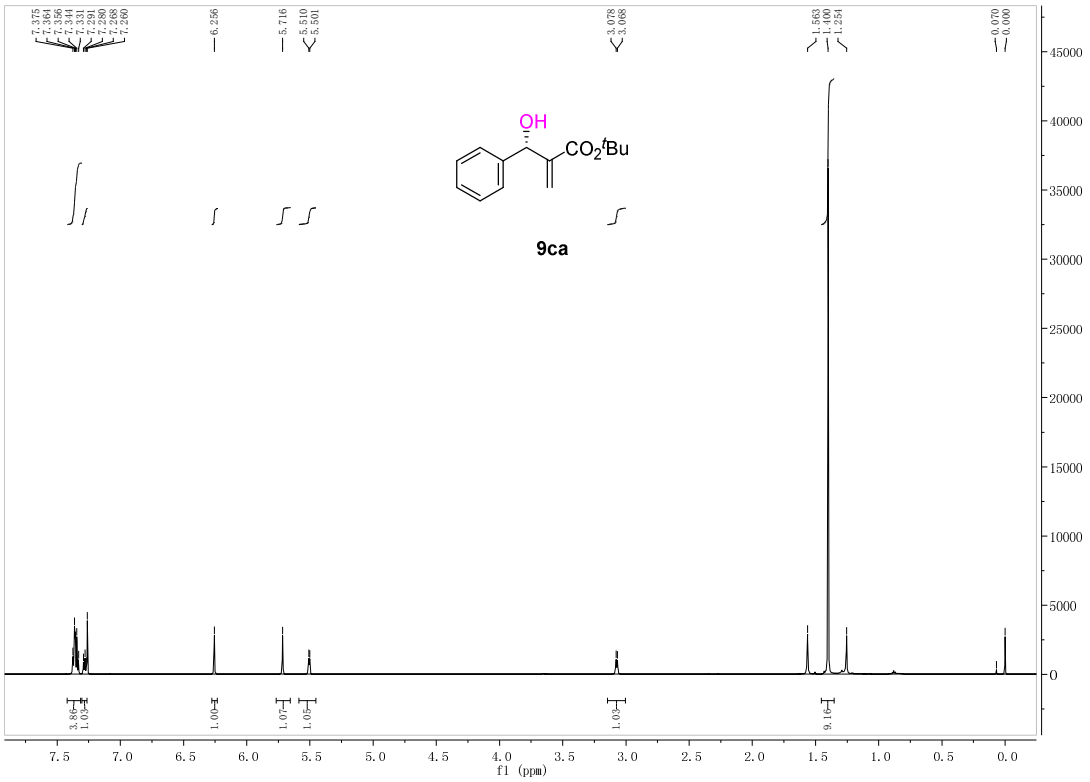
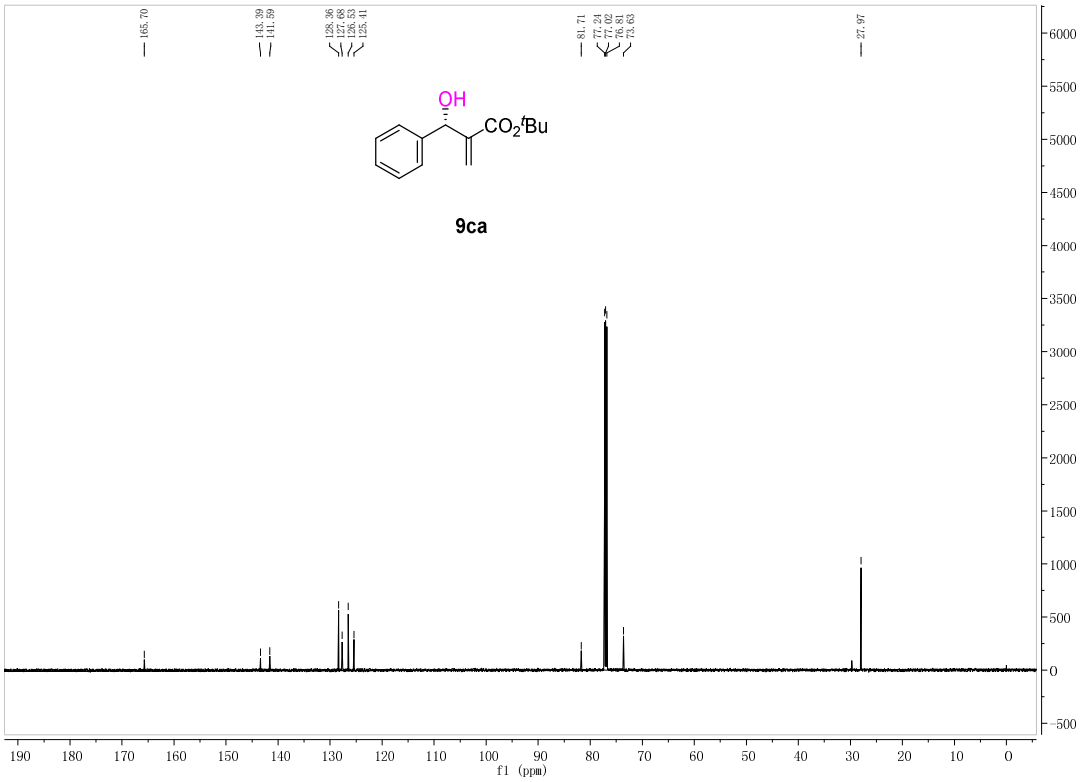


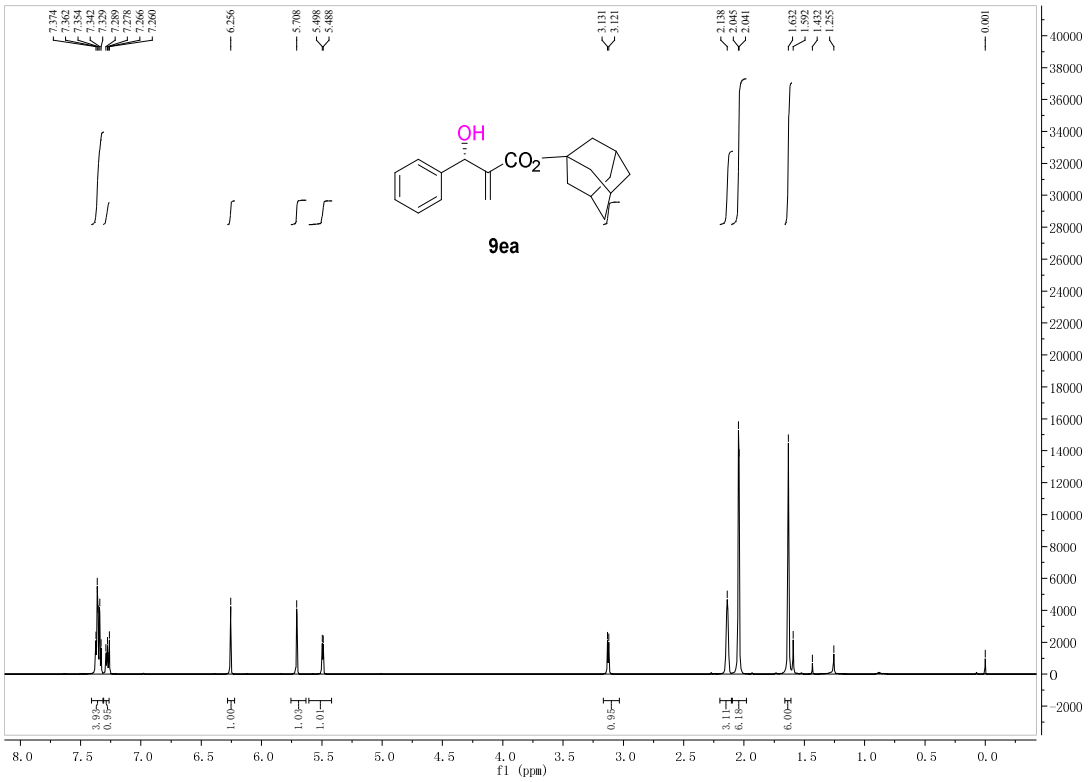
^{1}H-NMR for 9ca



^{13}C-NMR for 9ca



^{1}H-NMR for 9ea



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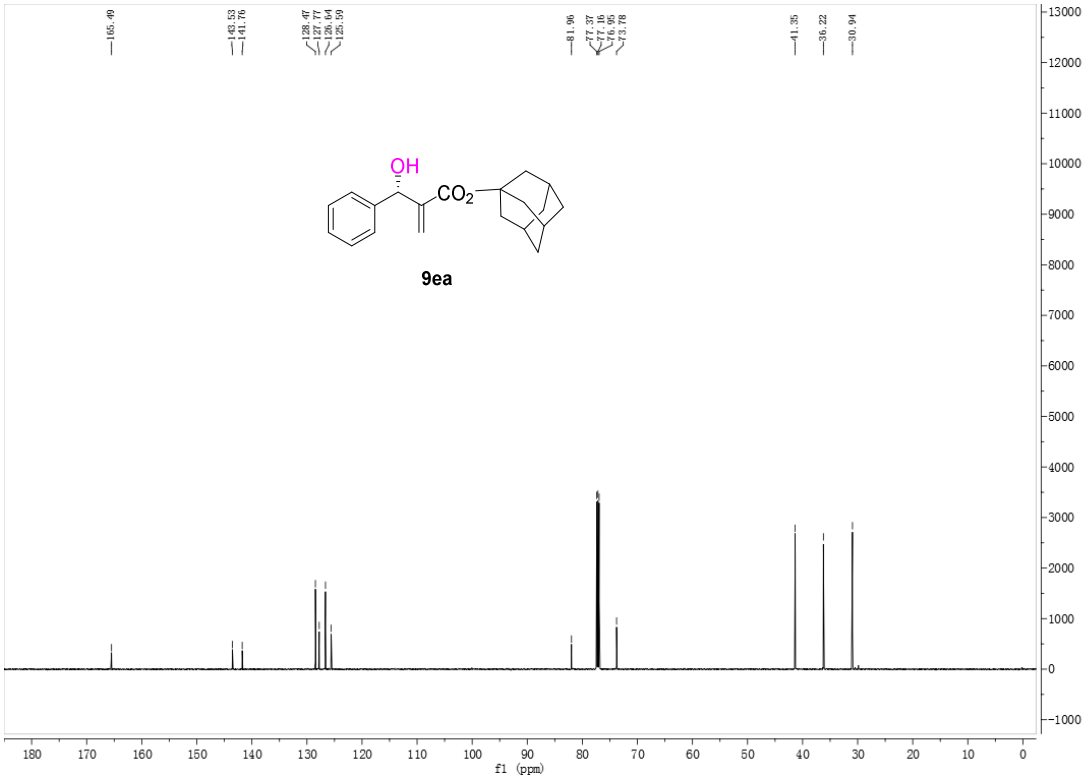
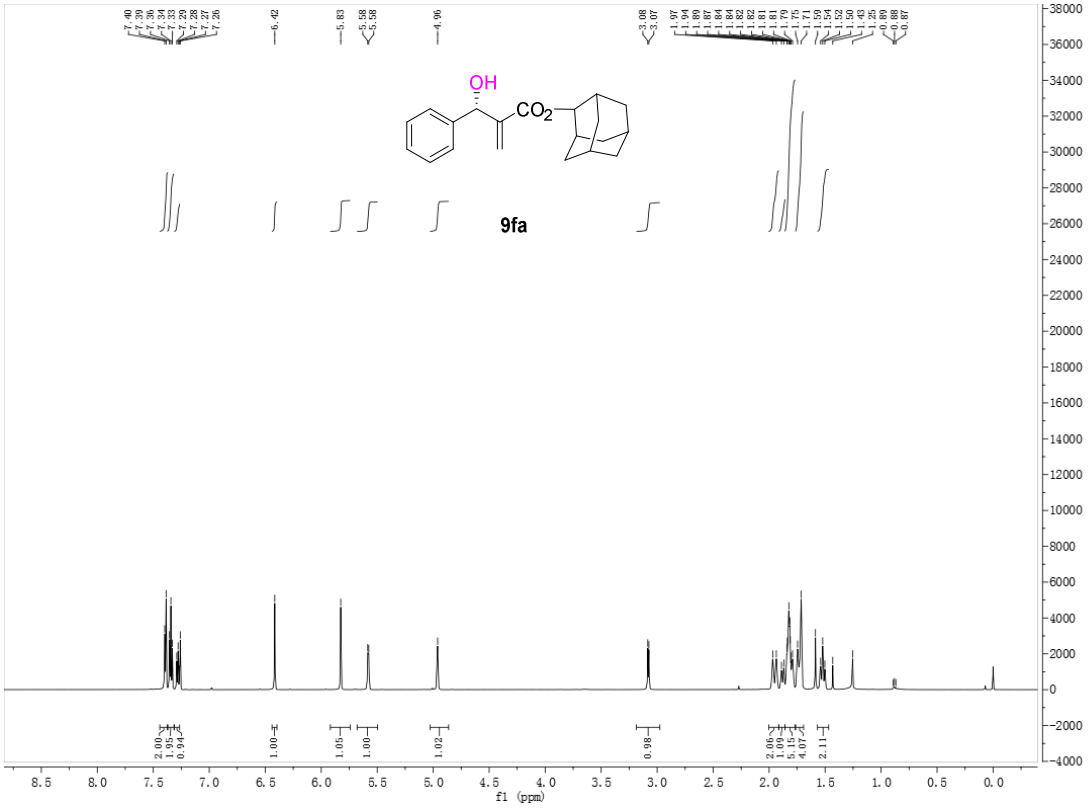
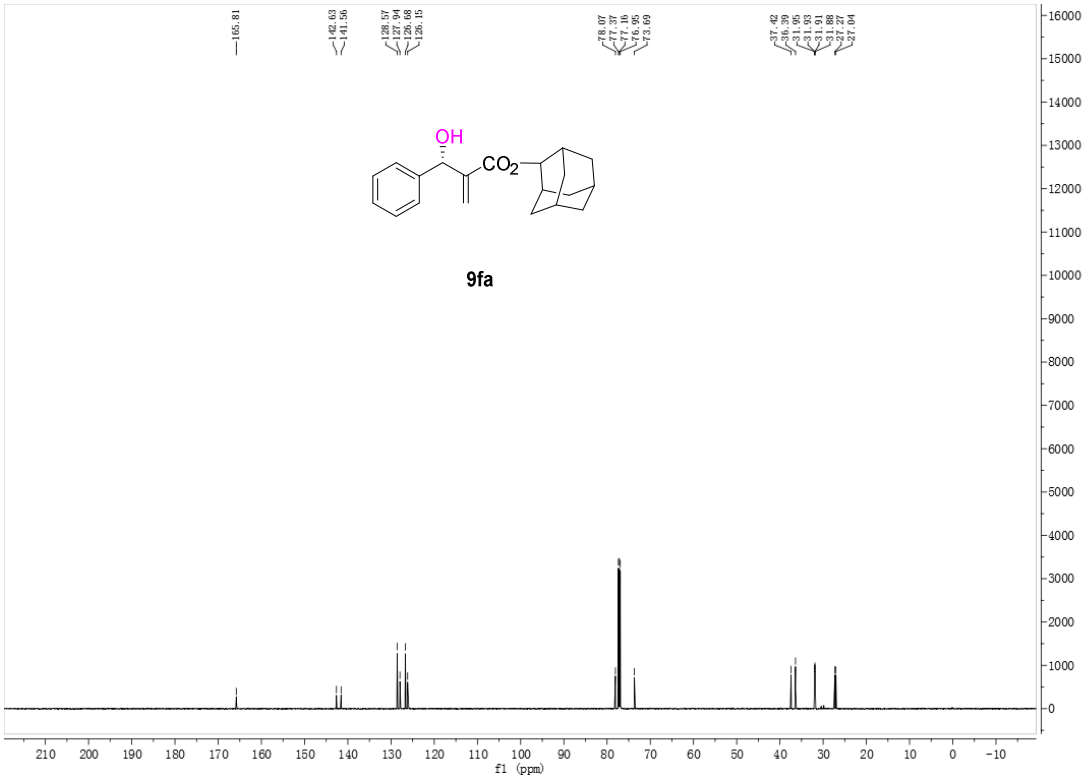


^{1}H-NMR for 9fa

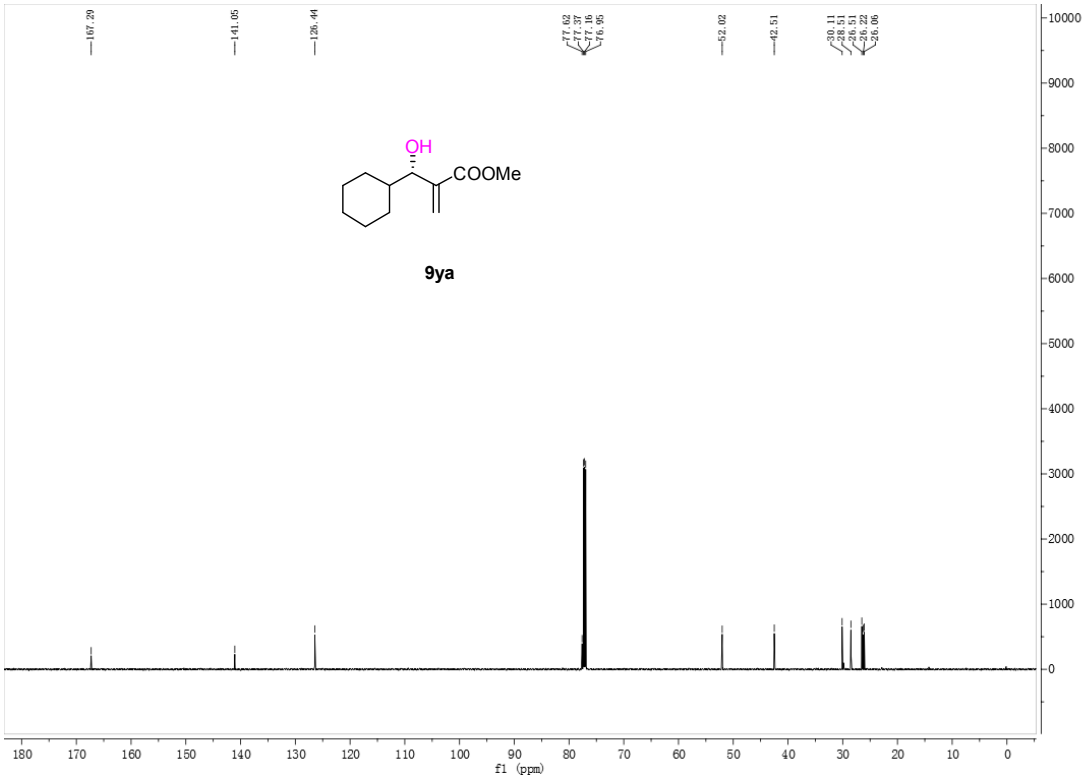


^{13}C-NMR for 9fa

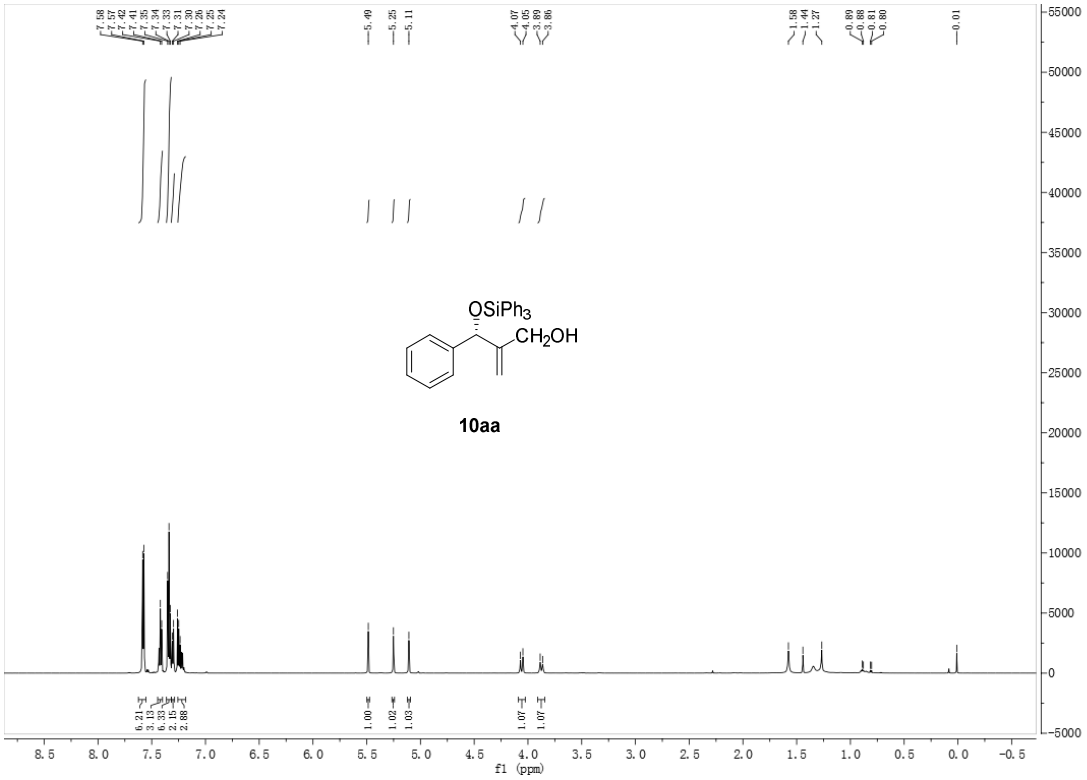


^{1}H-NMR for 9ya

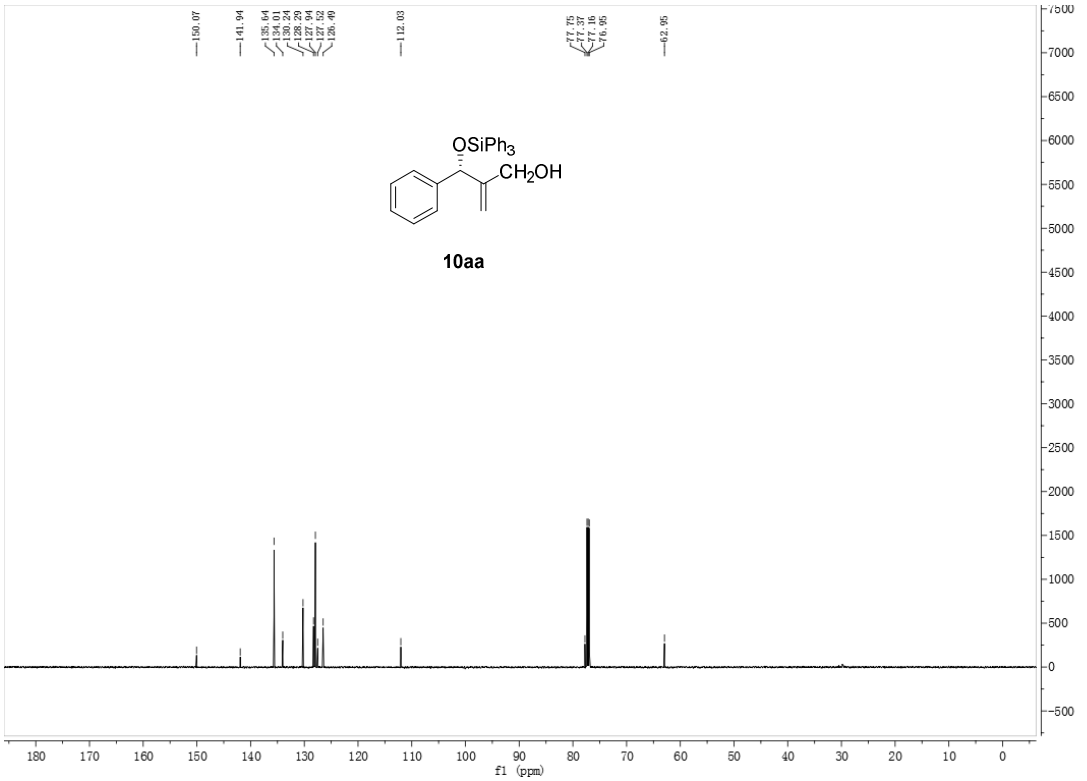
^{13}C-R for 9ya



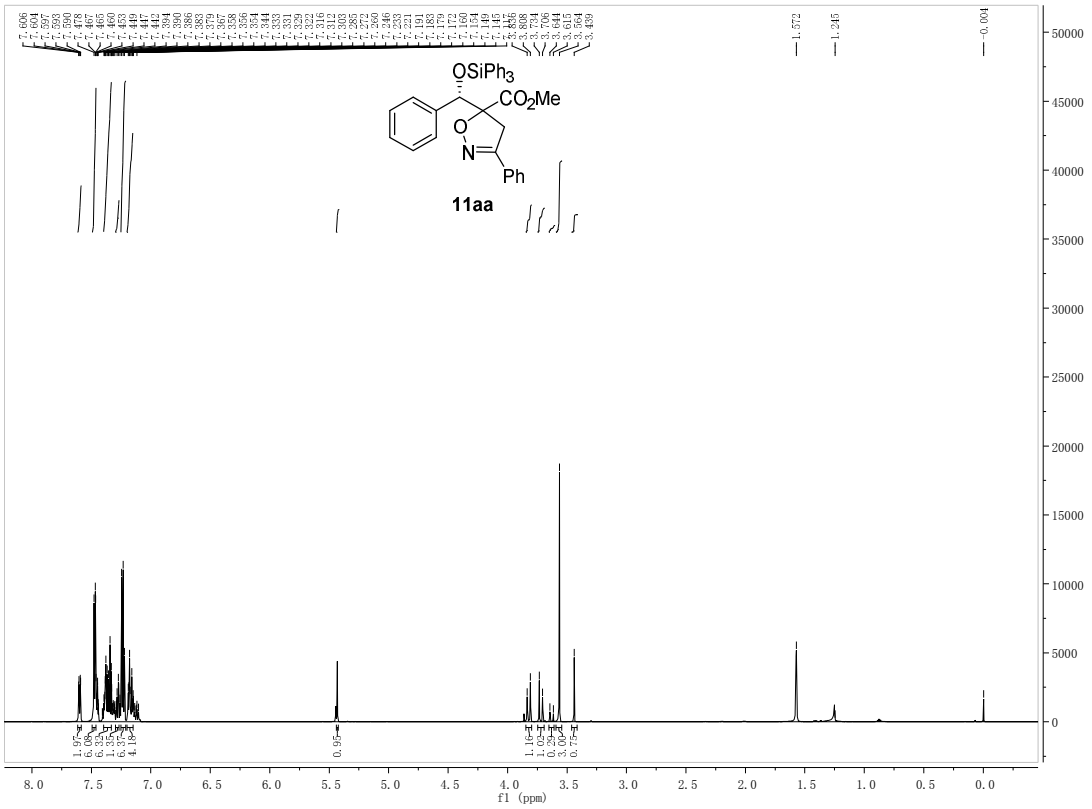
^{1}H-NMR for 10aa



^{13}C-NMR for 10aa



^{1}H-NMR for 11aa



^{13}C-NMR for 11aa