Supporting Information

Photoinduced synthesis of trisubstituted allylic molecules via migratory allylation of olefins

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1 Materials and methods

All reagents and solvents were purchased from certified chemical vendors and used without prior purification.

^{1}H NMR (500 MHz) and ^{13}C NMR (126 MHz) spectra were recorded on a Bruker Avance 500 MHz NMR spectrometer with $CDCl_3$ or DMSO-d_6 as solvent. The yield was determined by ^{1}H NMR using the 1,3,5-trimethoxybenzene as the internal standard. The spectra were calibrated by using residual undeuterated solvents (for ^{1}H NMR) and deuterated solvents (for ^{13}C NMR) as internal references: undeuterated chloroform (δ_H=7.26 ppm) and $CDCl_3$ (δ_C=77.16 ppm); undeuterated DMSO-d_6 (δ_H=2.50 ppm) and DMSO-d_6 (δ_C=39.52 ppm). Chemical shifts (δ) were expressed in ppm and coupling constants (J) in Hertz (Hz). The data is being reported as s = singlet, d = doublet, dd = double doblet, t = triplet, dt = double triplet, ddt = double double triplet, tt = triple triplet, q = quatriplet, qd = quadruple doblet, quint = quintuplet and m = multiplet or unresolved. Integration of the signals is presented as the number of hydrogen atoms. High-resolution mass spectra (HRMS) were recorded on an Agilent MSD-Trap-XCT or Q-Tof micro mass spectrometer. Electron ionization mass spectrometry (EIMS) was recorded on a Thermo Fisher Scientific Q-Exactive-GC. Kessil lamps were purchased from Tansoole, with precise wavelengths (390 nm). Ultraviolet-visible absorption experiments were performed using a UV-8000S(T) spectrophotometer.

Thin layer chromatography (TLC) was performed using a petroleum ether/ethyl acetate (EtOAc) solvent system as mobile phase and using MilliporeSigma glass TLC plates (silica gel 60 coated with F_{254}, 250 μm) and spots were visualized using UV light (254 nm). SiliaFlash® P60 silica gel (particle size: 40-63 μm, pore size: 60 Å) was used for flash column chromatography and petroleum ether/EtOAc solvent system was used as mobile phase.

2 Setup for photocatalytic reactions

The reaction setup is depicted in **Figure S1**. The reaction setup consists of 4 commercially available Kessil lamps which were purchased from Tansoole, with precise wavelengths (390 nm), cooling of the setup was performed by two commercially available fans to keep the temperature around 30 °C. Magnetic stirring was performed at 500 rpm.

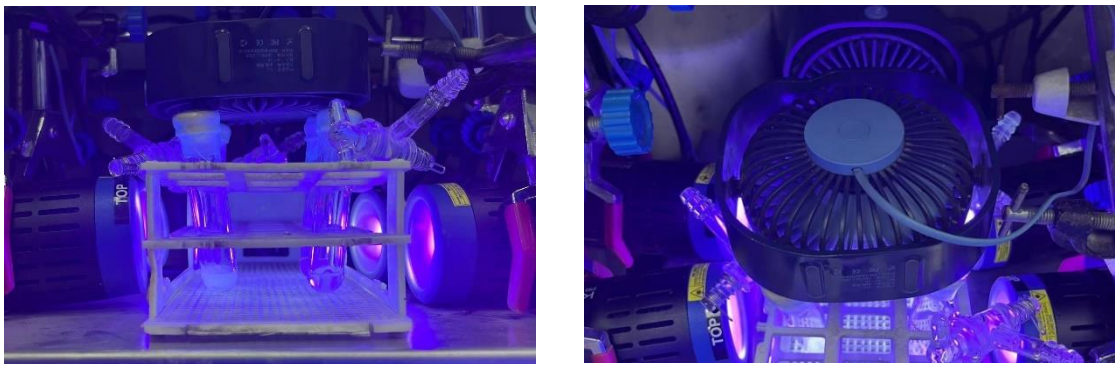


Figure S1. Photochemical set-up for regular-scale reactions.

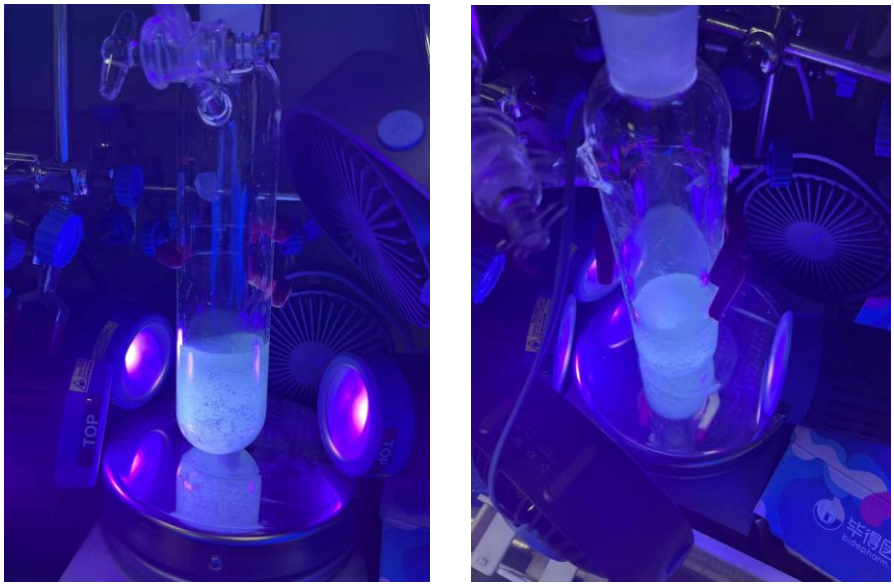
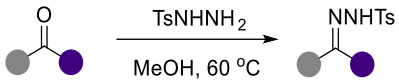


Figure S2. Photochemical set-up for large-scale reactions.

3 Preparation of starting materials

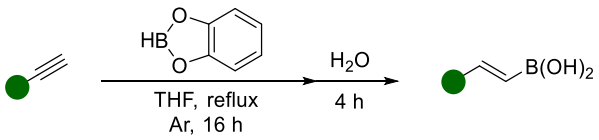
3.1. Synthesis of *N*-tosylhydrazones



General procedure A

N-tosylhydrazones were prepared according a reported procedure.[1] To a stirred solution of tosylhydrazide (5.0 mmol) in MeOH (10 mL) at 60 °C, ketone (1 equiv.) was added dropwise (or portionwise if solid). The reaction was completed within 0.5-3 h. After that, the solvent was removed directly under reduced pressure, and further purified by recrystallization or *via* silica gel chromatography (hexane:EtOAc, 2:1).

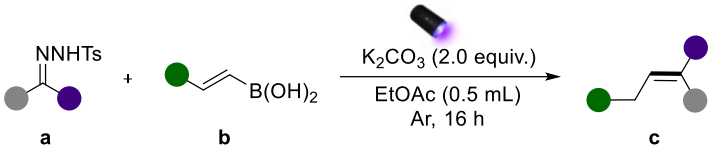
3.2. Synthesis of alkenyl boronic acids



General procedure B

Alkenyl boronic acids were prepared according a reported procedure.[2] The corresponding alkyne (2.0 mmol, 1.0 equiv) was added to catecholborane (1 M in THF, 3 mL, 1.5 equiv.) and the mixture was refluxed under argon atmosphere for 16 h. The solvent was evaporated and then H_2O (5 mL) was added. The suspension was vigorously stirred for 4 h at room temperature, and further purified by recrystallization or *via* silica gel chromatography to give the desired alkenyl boronic acids. (hexane:EtOAc, 1:1).

4. Experimental procedures

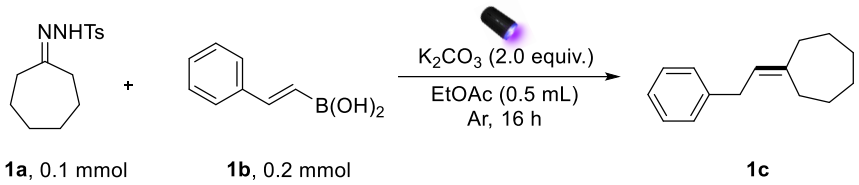


General procedure C

A dry 5 mL Schlenk tube containing a stirring bar was charged with 0.1 mmol of *N*-tosylhydrazone (1.0 equiv.), 0.3 mmol of K_2CO_3 (2.0 equiv.) and alkenyl boronic acids (2.0 equiv.), successively. After purging the flask three times under vacuum and three times under argon, it was charged with EtOAc (0.5 mL), The reaction was kept for 16 h under 40 W 390 nm Kessil lamps reaction setup (the progress can be monitored *via* TLC). Then, the mixture was filtered through a short path of silica gel with CH_2Cl_2 as an eluent. After removal of the solvent under vacuum, the residue was purified by column chromatography on silica gel (using hexane/EtOAc as eluent) to obtain the corresponding products. In addition, for products whose *E/Z* ratio can be obtained by ^{1}H NMR.

5. Optimization details for the reaction conditions

5.1 Control experiments

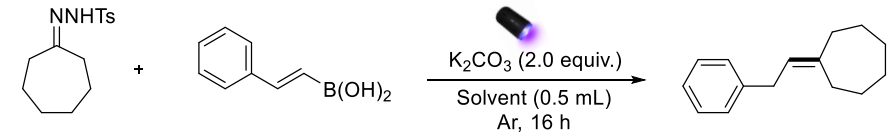


1a, 0.1 mmol **1b**, 0.2 mmol **1c**

Entry	Controlled parameter	Yield [%][b]
1[a]	Standard conditions[a]	89
2	without light	0
3	without base	0
4	without light and heating to 70 °C	0

[a]Standard conditions: **1a** (0.1 mmol, 1.0 equiv.), **1b** (0.2 mmol, 2.0 equiv.), K_2CO_3 (0.2 mmol, 2.0 equiv.), EtOAc (0.5 mL), irradiation with 40 W Kessil lamps (390 nm) at room temperature (around 30 °C) with cooling fan under argon atmosphere for 16 h. [b]Yields were determined by ^{1}H NMR using 1,3,5-trimethoxybenzene as the internal standard.

5.2 Screening of solvents[a]

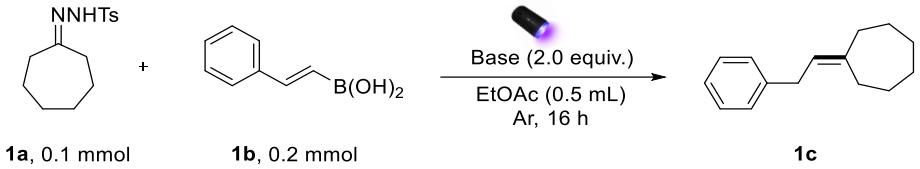


1a, 0.1 mmol **1b**, 0.2 mmol **1c**

Entry	Solvents	Yield [%][b]
1	1,4-dioxane	67
2	THF	18
3	ACN	20
4	DCM	24
5	toluene	60
6	EtOAc	89
7	MeOH	trace
8	EtOH	trace

[a]Standard conditions: **1a** (0.1 mmol, 1.0 equiv.), **1b** (0.2 mmol, 2.0 equiv.), K_2CO_3 (0.2 mmol, 2.0 equiv.), solvent (0.5 mL), irradiation with 40 W Kessil lamps (390 nm) at room temperature (around 30 °C) with cooling fan under argon atmosphere for 16 h. [b]Yields were determined by ^{1}H NMR using 1,3,5-trimethoxybenzene as the internal standard.

5.3 Screening of bases[a]

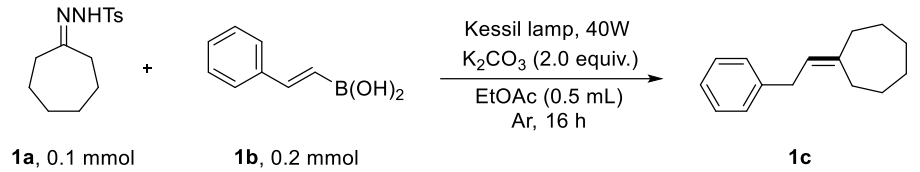


1a, 0.1 mmol **1b**, 0.2 mmol **1c**

Entry	Base	Yield [%][b]
1	K_2CO_3	89
2	K_3PO_4	42
3	Cs_2CO_3	62
4	DBU	24

[a]Standard conditions: **1a** (0.1 mmol, 1.0 equiv.), **1b** (0.2 mmol, 2.0 equiv.), base (0.2 mmol, 2.0 equiv.), 1,4-dioxane (0.5 mL), irradiation with 40 W Kessil lamps (390 nm) at room temperature (around 30 °C) with cooling fan under argon atmosphere for 16 h. [b]Yields were determined by ^{1}H NMR using 1,3,5-trimethoxybenzene as the internal standard.

5.4 Screening of light[a]

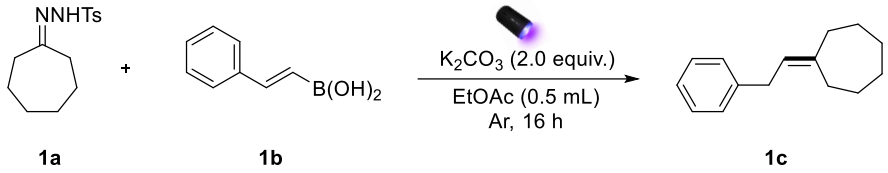


1a, 0.1 mmol **1b**, 0.2 mmol **1c**

Entry	Light	Yield [%][b]
1	390 nm	89
2[c]	400 nm	53
3[c]	405 nm	25
4	427 nm	trace
5	456 nm	trace

[a]Standard conditions: **1a** (0.1 mmol, 1.0 equiv.), **1b** (0.2 mmol, 2.0 equiv.), K_2CO_3 (0.2 mmol, 2.0 equiv.), EtOAc (0.5 mL), irradiation with 40 W Kessil lamps at room temperature (around 30 °C) with cooling fan under argon atmosphere for 16 h. [b]Yields were determined by ^{1}H NMR using 1,3,5-trimethoxybenzene as the internal standard. [c]10 W blue LED.

5.5 Screening of the ratio between the reagents[a]



1a **1b** **1c**

Entry	1a	1b	Yield [%][a]
1	1.0 equiv.	1.5 equiv.	56
2	1.0 equiv.	2.0 equiv.	89
3	1.0 equiv.	3.0 equiv.	74
4[c]	2.0 equiv.	1.0 equiv.	52

[a]Standard conditions: **1a** (0.1 mmol, 1.0 equiv.), **1b**, K_2CO_3 (0.2 mmol, 2.0 equiv.), EtOAc (0.5 mL), irradiation with 40 W Kessil lamps (390 nm) at room temperature (around 30 °C) with cooling fan under argon atmosphere for 16 h. [b]Yields were determined by ^{1}H NMR using 1,3,5-trimethoxybenzene as the internal standard. [c]**1a** (0.2 mmol, 2.0 equiv.), **1b**, K_2CO_3 (0.2 mmol, 2.0 equiv.).

6 Mechanistic studies

6.1 Time course experiments

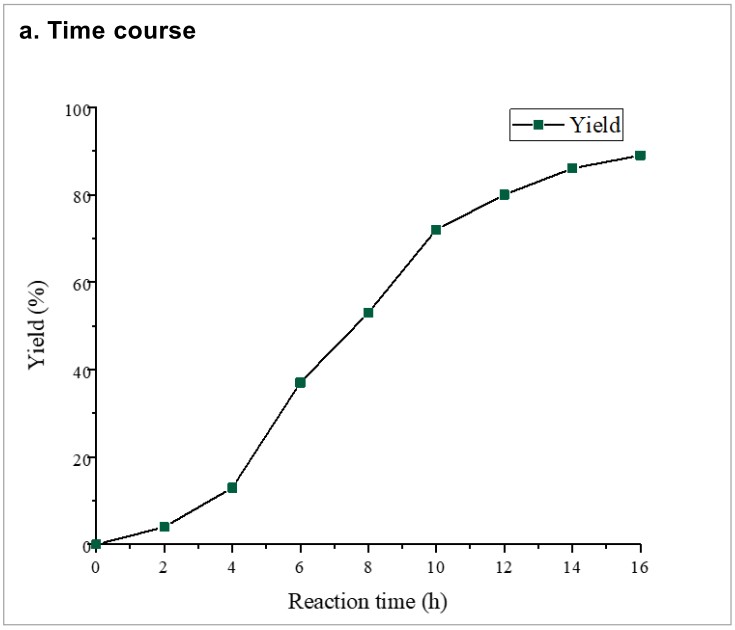
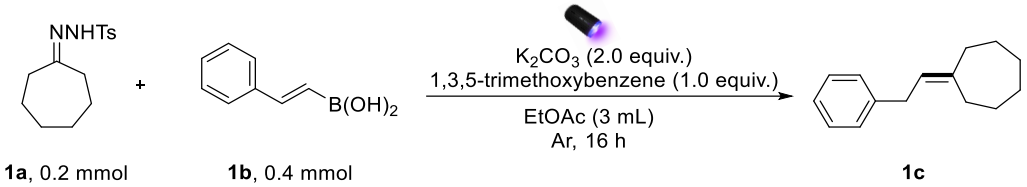


Figure S3. Time course study. Standard conditions: **1a** (0.1 mmol, 1.0 equiv.), **1b** (0.2 mmol, 2.0 equiv.), K_2CO_3 (0.2 mmol, 2.0 equiv.), EtOAc (0.5 mL), irradiation with 40 W Kessil lamps (390 nm) at room temperature (around 30 °C) with cooling fan under argon atmosphere. Yields were determined by ^1H NMR using 1,3,5-trimethoxybenzene as the internal standard.

6.2 On-off experiments



1a, 0.2 mmol **1b**, 0.4 mmol **1c**

Procedure: *N*-tosylhydrazone **1a** (0.2 mmol, 56.1 mg), **1b** (2.0 equiv., 0.4 mmol, 59.2 mg), K_2CO_3 (2.0 equiv., 0.4 mmol, 55.3 mg), and internal standard (1,3,5-trimethoxybenzene, 2.0 mmol, 33.6 mg) were added into a dry 5 mL Schlenk tube equipped with a stirring bar. The tube was evacuated and back filled with Ar for three times, followed by the addition of EtOAc (3 mL) via syringe. Then the reaction mixture was irradiated by a 390 nm Kessil lamps (40 W) at room temperature. An aliquot of the reaction mixture was then taken at the indicated times and the yields were determined by ^{1}H NMR spectroscopy.

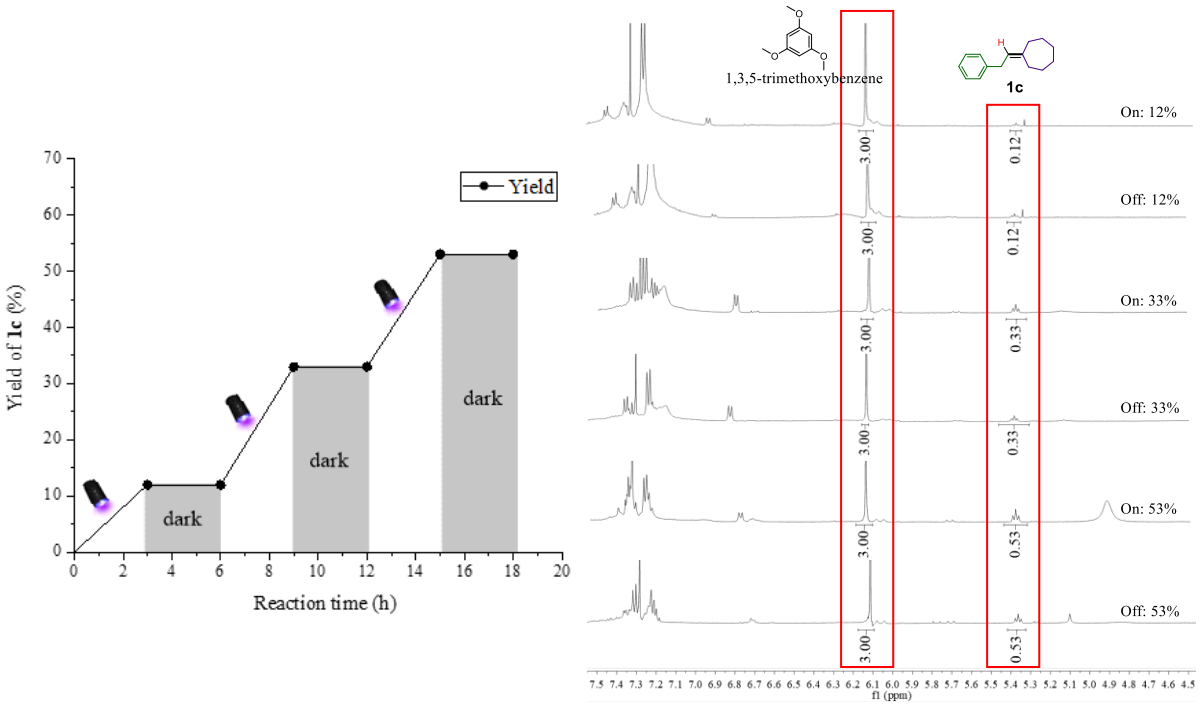


Figure S4. The comparison of On-off experiment yields determined by hydrogen spectrometry.

6.3 Ultraviolet visible absorption experiments

Ultraviolet-visible absorption experiments were performed using a UV-8000S(T) spectrophotometer. In each experiment, the varying samples were combined in the solvent EtOAc in screw-top 1.0 cm quartz cuvettes.

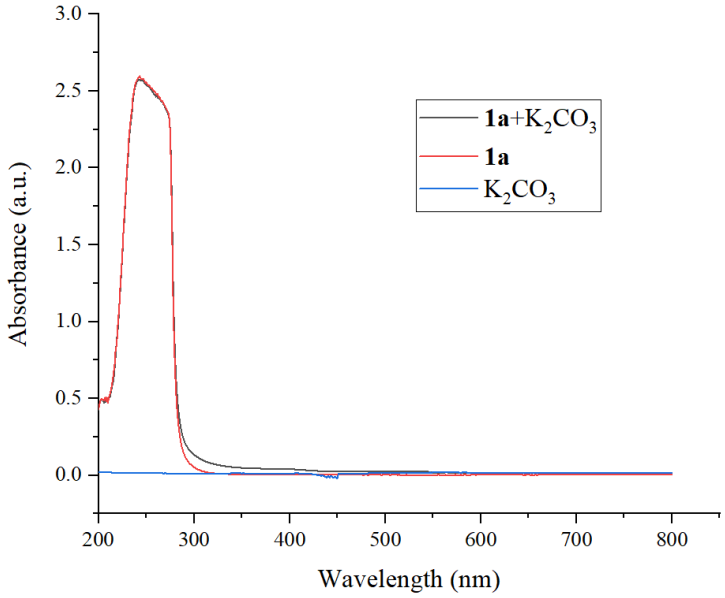


Figure S5. UV/vis absorption spectra of individual reaction components and a combination thereof. Ultraviolet-visible absorption experiments:**1a** (8.0×10^{-4} M) in EtOAc, K_2CO_3 (16.0×10^{-4} M) in EtOAc, **1a** (8.0×10^{-4} M) + **1b** (16.0×10^{-4} M) in EtOAc.

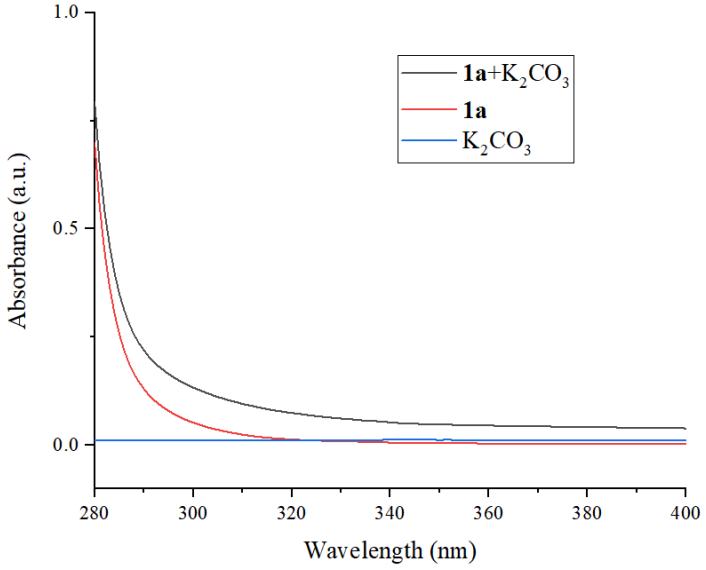
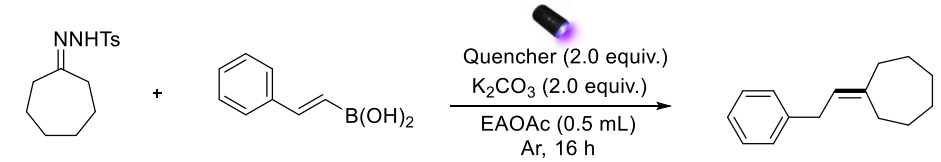


Figure S6. UV/vis absorption spectra of individual reaction components and a combination thereof. Ultraviolet-visible absorption experiments:**1a** (8.0×10^{-4} M) in EtOAc, K_2CO_3 (16.0×10^{-4} M) in EtOAc, **1a** (8.0×10^{-4} M) + **1b** (16.0×10^{-4} M) in EtOAc. This picture is a partial enlargement of **Figure S5**.

6.4 Radical quenching experiments

The reaction was operated under standard conditions with extra 2,2,6,6-tetramethyl-1-piperinedinyloxy (TEMPO) and Butylated Hydroxytoluene (BHT). The reaction also showed limited reactivity under air rather than under Ar.

Quenching experiments[a]:

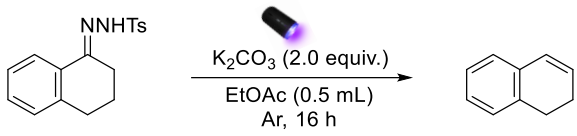


1a, 0.1 mmol **1b**, 0.2 mmol **1c**

Entry	Quencher (5.0 equiv)	**Yield [%]**[b]
1	TEMPO	70
2	BHT	78

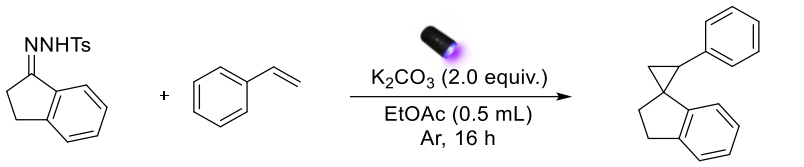
[a]Standard conditions: **1a** (0.1 mmol, 1.0 equiv.), **1b** (0.2 mmol, 2.0 equiv.), K_2CO_3 (0.2 mmol, 2.0 equiv.), EtOAc (0.5 mL), irradiation with 40 W Kessil lamps (390 nm) at room temperature (around 30 °C) with cooling fan under argon atmosphere for 16 h. [b]Yields were determined by ^{1}H NMR using 1,3,5-trimethoxybenzene as the internal standard.

6.5 Carbene trapping experiments



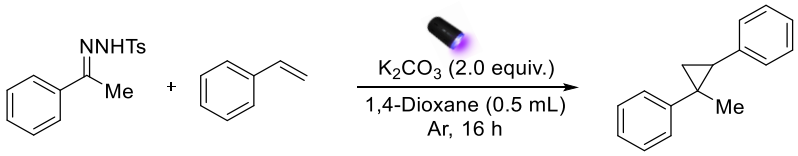
14a, 0.1 mmol **14d**, 18%

A dry 5 mL Schlenk tube containing a stirring bar was charged with 0.1 mmol of *N*-tosylhydrazone **14a** (0.1 mmol) and 0.2 mmol of K_2CO_3 (2.0 equiv.), successively. After purging the flask three times under vacuum and three times under argon, it was charged with EtOAc (0.5 mL), The reaction was kept for 16 h under 40 W 390 nm Kessil lamps reaction setup (the progress can be monitored *via* TLC). Then, the mixture was filtered through a short path of silica gel with CH_2Cl_2 as an eluent. After removal of the solvent under vacuum, the residue was purified by column chromatography on silica gel (hexane) to obtain **14d**.



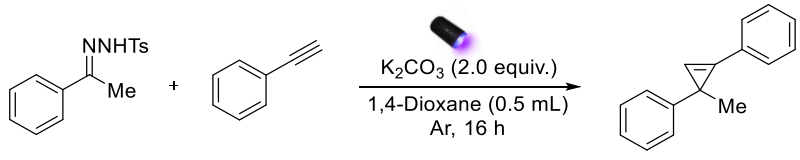
13a 5 equiv. **13d**, 43%, d.r. = 1:1.3

A dry 5 mL Schlenk tube containing a stirring bar was charged with 0.1 mmol of *N*-tosylhydrazone **13a** (0.1 mmol), 0.2 mmol of K_2CO_3 (2.0 equiv.) and Styrene (5.0 equiv.), successively. After purging the flask three times under vacuum and three times under argon, it was charged with EtOAc (0.5 mL), The reaction was kept for 16 h under 40 W 390 nm Kessil lamps reaction setup (the progress can be monitored *via* TLC). Then, the mixture was filtered through a short path of silica gel with CH_2Cl_2 as an eluent. After removal of the solvent under vacuum, the residue was purified by column chromatography on silica gel (hexane) to obtain **13d**.



25a 5 equiv. **25d**, 30%, d.r. = 1:1.2

A dry 5 mL Schlenk tube containing a stirring bar was charged with 0.1 mmol of *N*-tosylhydrazone **25a** (0.1 mmol), 0.2 mmol of K_2CO_3 (2.0 equiv.) and Styrene (5.0 equiv.), successively. After purging the flask three times under vacuum and three times under argon, it was charged with 1,4-Dioxane (0.5 mL), The reaction was kept for 16 h under 40 W 390 nm Kessil lamps reaction setup (the progress can be monitored *via* TLC). Then, the mixture was filtered through a short path of silica gel with CH_2Cl_2 as an eluent. After removal of the solvent under vacuum, the residue was purified by column chromatography on silica gel (hexane) to obtain **25d**.



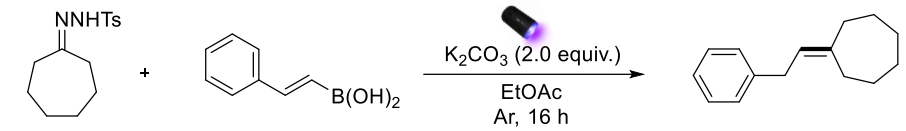
25a 5 equiv. **25e**, 11%

A dry 5 mL Schlenk tube containing a stirring bar was charged with 0.1 mmol of *N*-tosylhydrazone

25a (0.1 mmol), 0.2 mmol of K_2CO_3 (2.0 equiv.) and Phenylacetylene (5.0 equiv.), successively. After purging the flask three times under vacuum and three times under argon, it was charged with 1,4-Dioxane (0.5 mL), The reaction was kept for 16 h under 40 W 390 nm Kessil lamps reaction setup (the progress can be monitored *via* TLC). Then, the mixture was filtered through a short path of silica gel with CH_2Cl_2 as an eluent. After removal of the solvent under vacuum, the residue was purified by column chromatography on silica gel (hexane) to obtain **25e**.

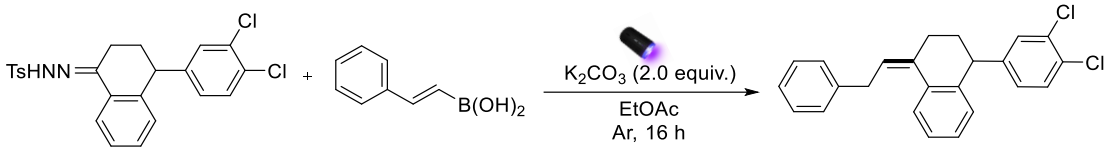
7 The Application of the Reaction

7.1 Gram-scale synthesis of alkenes



1a, 5 mmol **1b**, 2 equiv. **1c**, 1.34 g, 68%

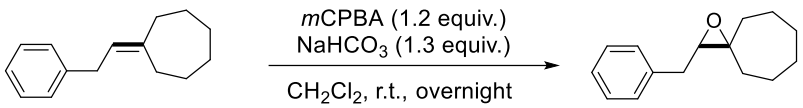
A dry 250 mL Schlenk tube containing a stirring bar was charged with 5 mmol of *N*-tosylhydrazone **1a** (1.40 g), **1b** (10 mmol, 2 equiv., 1.48 g), K_2CO_3 (10 mmol, 2 equiv., 1.38 g), After purging the flask for three times under vacuum and three times under argon, it was charged with EtOAc (25 mL). The reaction was kept for 16 h under 40 W 390 nm Kessil lamp reaction setup (the progress can be monitored via TLC). Then, the resulting mixture underwent a workup using distilled water and was extracted three times with ethyl acetate. The combined organic layers were dried over anhydrous Na_2SO_4, filtered and concentrated in vacuo. Products were purified *via* column chromatography with hexane as solvents.



43a, 5 mmol **1b**, 2 equiv. **43c**, 1.19 g
63%, *Z*/*E* = 37/63

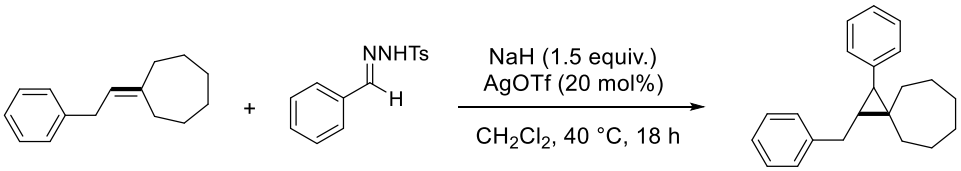
A dry 250 mL Schlenk tube containing a stirring bar was charged with 5 mmol of *N*-tosylhydrazone **43a** (2.29 g), **1b** (10 mmol, 2 equiv., 1.48 g), K_2CO_3 (10 mmol, 2 equiv., 1.38 g), After purging the flask for three times under vacuum and three times under argon, it was charged with EtOAc (25 mL). The reaction was kept for 16 h under 40 W 390 nm Kessil lamp reaction setup (the progress can be monitored via TLC). Then, the resulting mixture underwent a workup using distilled water and was extracted three times with ethyl acetate. The combined organic layers were dried over anhydrous Na_2SO_4, filtered and concentrated in vacuo. Products were purified *via* column chromatography with hexane as solvents.

7.2 Further transformation of compound 1c



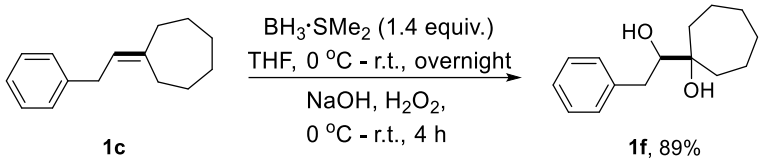
1c **1d**, 82%

Procedure D: The title compound was prepared following a literature protocol.[3] To an oven-dried flask equipped with a stir bar were added **1c** (200 mg, 1.0 mmol, 1.0 euqiv.), *m*CPBA (207 mg, 1.2 mmol, 1.2 equiv.), $NaHCO_3$ (109 mg, 1.3 mmol, 1.3 equiv.), and CH_2Cl_2 (5.0 mL). The reaction mixture was stired for overnight at room tempreture. When the reaction was completed, 5% aq solution of sodium thiosulfate $Na_2O_3S_2$ was added to the mixture at 0 °C. After stiring 5 min, the reaction mixture was slowly warmed to room temperature and stirring was continued for 15 min. The mixture was washed with saturated sodium chloride solution and extracted with CH_2Cl_2. The organic layer was dried over anhydrous sodium sulfate and concentrated in vacuo. After removal of the solvent under vacuum, the filtrate was concentrated in vacuo and purified by column chromatography on silica gel (using petroleum ether/EtOAc = 20/1, v/v, as eluent) to obtain **1d** (177.4 mg, 82%) as a colorless oil.



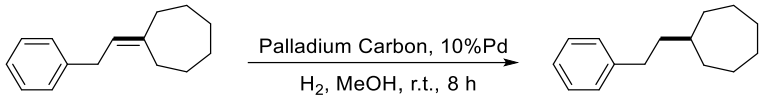
1c **1e**, 42%

Procedure E: The title compound was prepared following a literature protocol.[4] To an oven-dried screw-cap reaction tube equipped with a Teflon-coated magnetic stir bar were added **1c** (60.0 mg, 0.3 mmol, 1.5 euqiv.), *N*-tosylhydrazone (54.8 mg, 0.2 mmol, 1.0 equiv.), NaH (15.0 mg, 0.3 mmol, 1.5 equiv., 60 wt% dispersion in mineral oil) and AgOTf (10.3 mg, 20 mol%). Then, CH_2Cl_2 (2.0 mL) were added and the vial was sealed. The reaction mixture was transferred to preheated oil bath at 60 °C for 24 h. The mixture was cooled to room temperature and filtered through a short path of silica gel with CH_2Cl_2 as an eluent. After removal of the solvent under vacuum, the residue was purified by column chromatography on silica gel (using hexane) to obtain **1e** (24.0 mg, 42%) as a colorless oil.



1c **1f**, 89%

Procedure F: The title compound was prepared following a literature protocol.[5] To a stirred solution of **1c** (300 mg, 1.5 mmol) in dry THF (6.7 mL), cooled to 0 °C, was added a $BH_3 \cdot SMe_2$ solution (1.1 mL, 2.1 mmol, 1.4 equiv.). The mixture was kept at 0 °C for 3 h and then room temp. overnight. A mixture of NaOH (3 N, 0.47 mL) and H_2O_2 (30%, 0.47 mL) was added and was stirred for 4 h. The mixture was diluted with Et_2O, washed with brine, and dried, and the solvents were evaporated. Column chromatography (using petroleum ether/EtOAc = 8/1, v/v, as eluent), to obtain **1f** (312.4 mg, 89%) as a colorless oil.

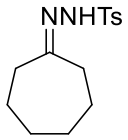


1c **1g**, 96%

Procedure G: To a stirred solution of **1c** (50 mg, 0.25 mmol) in MeOH (3 mL), was added Pd/C (6.3 mg, 10% Pd). After purging the flask for three times under vacuum and three times under H_2, it was charged with a hydrogen balloon. The reaction was kept for 8 h (the progress can be monitored *via* TLC). Then, the resulting mixture filter with diatomaceous earth and concentrated in vacuo. Products were purified *via* column chromatography with hexane as solvents, to obtain **1g** (47.1 mg, 93%) as a colorless oil.

8 Characterization data of products and synthesized substrates

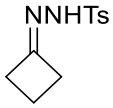
8.1 Characterization data of synthesized *N*-tosylhydrazones



***N*'-cycloheptylidene-4-methylbenzenesulfonohydrazide (1a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **1a** was obtained as a white solid (1.27 g, isolated yield: 90%). **1a** was known in the published literature.[6]

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.85 (s, 1H), 7.72 (d, J = 8.2 Hz, 2H), 7.37 (d, J = 8.0 Hz, 2H), 2.37 (s, 3H), 2.35 – 2.30 (m, 2H), 2.25 – 2.323 (m, 2H), 1.59 – 1.57 (m, 2H), 1.45 (d, J = 8.1 Hz, 6H).

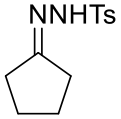
ESI HRMS: calcd. for $C_{14}H_{20}N_2O_2S$ $[M+H]^+$: 281.1318, found: 281.1327.



***N*'-cyclobutylidene-4-methylbenzenesulfonohydrazide (2a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **2a** was obtained as a white solid (1.04 g, isolated yield: 87%). **2a** was known in the published literature.[7]

^{1}H NMR (500 MHz, $CDCl_3$) δ 10.22 (s, 1H), 7.70 (d, J = 8.3 Hz, 2H), 7.44 – 7.22 (m, 2H), 2.79 (t, J = 8.1 Hz, 2H), 2.74 (t, J = 8.0 Hz, 2H), 2.37 (s, 3H), 1.86 – 1.79 (m, 2H).

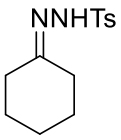
ESI HRMS: calcd. for $C_{11}H_{14}N_2O_2S$ $[M+H]^+$: 239.0849, found: 239.0862.



***N*'-cyclopentylidene-4-methylbenzenesulfonohydrazide (3a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **3a** was obtained as a white solid (1.15 g, isolated yield: 91%). **3a** was known in the published literature.[6]

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.96 (s, 1H), 7.90 – 7.60 (m, 2H), 7.38 (d, J = 8.0 Hz, 2H), 2.37 (s, 3H), 2.22 (t, J = 7.4 Hz, 2H), 2.17 (t, J = 7.2 Hz, 2H), 1.68 (q, J = 6.9 Hz, 2H), 1.60 (q, J = 7.2 Hz, 2H).

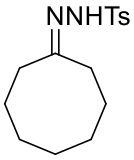
ESI HRMS: calcd. for $C_{12}H_{16}N_2O_2S$ $[M+H]^+$: 253.1005, found: 253.1013.



***N*'-cyclohexylidene-4-methylbenzenesulfonohydrazide (4a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **4a** was obtained as a white solid (1.26 g, isolated yield: 94%). **4a** was known in the published literature.[6]

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.84 (d, J = 8.4 Hz, 2H), 7.30 (d, J = 8.0 Hz, 2H), 2.42 (s, 3H), 2.23 – 2.19 (m, 4H), 1.69 – 1.51 (m, 6H).

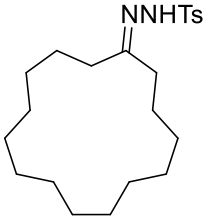
ESI HRMS: calcd. for $C_{13}H_{18}N_2O_2S$ $[M+H]^+$: 267.1162, found: 267.1175.



***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (5a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **5a** was obtained as a white solid (1.33 g, isolated yield: 90%). **5a** was known in the published literature.[8]

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.99 (s, 1H), 7.84 – 7.63 (m, 2H), 7.37 (d, J = 8.0 Hz, 2H), 2.37 (s, 3H), 2.35 – 2.28 (m, 2H), 2.20 – 2.09 (m, 2H), 1.69 – 1.61 (m, 2H), 1.61 – 1.53 (m, 2H), 1.40 – 1.36 (m, 4H), 1.25 – 1.22 (m, 2H).

ESI HRMS: calcd. for $C_{15}H_{22}N_2O_2S$ $[M+H]^+$: 295.1475, found: 295.1488.

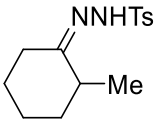


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (6a)**:

Prepared according to the synthesis of hydrazone. Following the general procedure, was obtained as a white solid (1.51 g, isolated yield: 77%). **6a** was known in the published literature.[7]

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.01 (s, 1H), 7.69 (d, J = 8.3 Hz, 2H), 7.36 (d, J = 8.0 Hz, 2H), 2.36 (s, 3H), 2.18 (t, J = 7.4 Hz, 2H), 2.06 (t, J = 7.3 Hz, 2H), 1.39 – 1.36 (m, 4H), 1.29 – 1.14 (m, 20H).

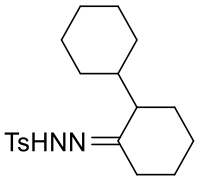
ESI HRMS: calcd. for $C_{22}H_{36}N_2O_2S$ $[M+Na]^+$: 415.2390, found: 415.2397.



4-methyl-*N*'-(2-methylcyclohexylidene)benzenesulfonohydrazide (7a): Prepared according to the synthesis of hydrazone. Following the general procedure, **7a** was obtained as a white solid (1.23 g, isolated yield: 88%). **7a** was known in the published literature.[6]

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.84 (d, J = 8.3 Hz, 2H), 7.84 (s, 1H), 7.29 (d, J = 8.0 Hz, 2H), 2.42 (s, 3H), 2.57 – 2.51 (m, 1H), 2.28 – 2.13 (m, 1H), 1.85 – 1.71 (m, 4H), 1.53 – 1.31 (m, 2H), 1.28 – 1.10 (m, 1H), 1.01 (dd, J = 6.5, 2.7 Hz, 3H).

ESI HRMS: calcd. for $C_{14}H_{20}N_2O_2S$ $[M+H]^+$: 281.1318, found: 281.1331.

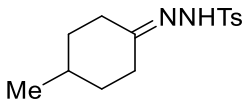


***N*'-([1,1'-bi(cyclohexan)]-2-ylidene)-4-methylbenzenesulfonohydrazide (8a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **8a** was obtained as a white solid (1.39 g, isolated yield: 80%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.83 (d, J = 8.3 Hz, 2H), 7.28 (d, J = 8.0 Hz, 2H), 2.41 (s, 3H), 2.26 – 2.21 (m, 1H), 2.03 – 1.94 (m, 2H), 1.78 – 1.64 (m, 4H), 1.63 – 1.52 (m, 4H), 1.50 – 1.43 (m, 3H), 1.23 – 1.12 (m, 1H), 1.10 – 0.97 (m, 3H), 0.82 – 0.68 (m, 1H), 0.66 – 0.52 (m, 1H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 164.3, 143.9, 135.5, 129.4, 128.4, 49.7, 36.1, 31.3, 30.0, 28.3, 26.5, 26.3, 25.3, 22.3, 21.7.

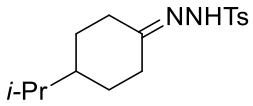
ESI HRMS: calcd. for $C_{19}H_{28}N_2O_2S$ $[M+H]^+$: 349.1944, found: 349.1957.



4-methyl-*N*'-(4-methylcyclohexylidene)benzenesulfonohydrazide (9a): Prepared according to the synthesis of hydrazone. Following the general procedure, **9a** was obtained as a white solid (1.25 g, isolated yield: 89%). **9a** was known in the published literature.[9]

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.10 (s, 1H), 7.71 (d, J = 8.0 Hz, 2H), 7.37 (d, J = 8.0 Hz, 2H), 2.73 (d, J = 14.6 Hz, 1H), 2.37 (s, 3H), 2.18 – 2.00 (m, 2H), 1.85 – 1.76 (m, 1H), 1.75 – 1.71 (m, 2H), 1.61 – 1.57 (m, 1H), 1.06 – 0.91 (m, 2H), 0.86 (d, J = 6.5 Hz, 3H).

ESI HRMS: calcd. for $C_{14}H_{20}N_2O_2S$ $[M+Na]^+$: 303.1138, found: 303.1145.

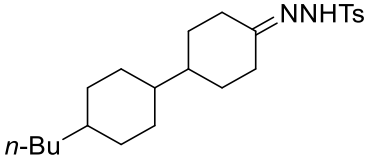


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (10a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **10a** was obtained as a white solid (1.33 g, isolated yield: 86%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.09 (s, 1H), 7.71 (d, J = 8.3 Hz, 2H), 7.37 (d, J = 8.0 Hz, 2H), 2.81 – 2.77 (m, 1H), 2.37 (s, 3H), 2.18 – 2.15 (m, 1H), 2.03 (td, J = 13.4, 4.8 Hz, 1H), 1.78 – 1.72 (m, 3H), 1.48 – 1.38 (m, 1H), 1.30 – 1.24 (m, 1H), 1.07 – 0.96 (m, 2H), 0.82 (dd, J = 6.8, 2.3 Hz, 6H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 162.4, 143.0, 136.4, 129.3, 127.5, 42.1, 34.1, 31.5, 29.4, 28.3, 26.8, 21.0, 19.8, 19.7.

ESI HRMS: calcd. for $C_{16}H_{24}N_2O_2S$ $[M+H]^+$: 309.1631, found: 309.1647.

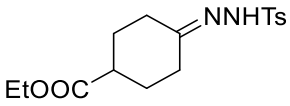


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (11a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **11a** was obtained as a white solid (1.71 g, isolated yield: 85%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.08 (s, 1H), 7.69 (d, J = 8.1 Hz, 2H), 7.36 (d, J = 8.0 Hz, 2H), 2.84 – 2.66 (m, 1H), 2.35 (s, 3H), 2.14 (d, J = 13.8 Hz, 1H), 2.06 – 1.92 (m, 1H), 1.74 (dd, J = 12.6, 3.8 Hz, 2H), 1.70 – 1.66 (m, 2H), 1.65 – 1.56 (m, 2H), 1.26 – 1.18 (m, 5H), 1.10 (q, J = 6.7 Hz, 3H), 1.01 (tt, J = 12.1, 4.6 Hz, 3H), 0.95 – 0.86 (m, 2H), 0.86 – 0.74 (m, 6H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 162.6, 143.1, 136.5, 129.4, 127.6, 41.9, 41.4, 37.3, 36.7, 34.2, 33.1, 29.7, 29.6, 29.6, 28.7, 28.6, 26.9, 22.5, 21.1, 14.1.

ESI HRMS: calcd. for $C_{23}H_{36}N_2O_2S$ $[M+Na]^+$: 427.2390, found: 427.2396.

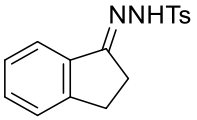


***N'*-cyclooctylidene-4-methylbenzenesulfonohydrazide (12a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **12a** was obtained as a yellow solid (1.40 g, isolated yield: 77%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.82 (d, J = 8.0 Hz, 2H), 7.30 (d, J = 8.0 Hz, 2H), 4.11 (q, J = 7.1 Hz, 2H), 2.72 – 2.56 (m, 1H), 2.55 – 2.46 (m, 2H), 2.42 (s, 3H), 2.21 – 2.11 (m, 1H), 2.04 – 1.94 (m, 3H), 1.78 – 1.64 (m, 2H), 1.23 (t, J = 7.1 Hz, 3H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 174.1, 160.7, 143.1, 136.4, 129.4, 127.5, 60.0, 40.5, 32.8, 28.7, 27.4, 25.6, 21.1, 14.1.

ESI HRMS: calcd. for $C_{16}H_{22}N_2O_4S$ $[M+H]^+$: 339.1373, found: 339.1380.

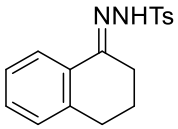


***N'*-(2,3-dihydro-1H-inden-1-ylidene)-4-methylbenzenesulfonohydrazide (13a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **13a** was obtained as a white solid (1.17 g, isolated yield: 78%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.93 (s, 2H), 7.70 (d, J = 7.7 Hz, 1H), 7.34 – 7.29 (m, 3H), 7.28 – 7.20 (m, 2H), 3.08 – 2.99 (m, 2H), 2.73 – 2.63 (m, 2H), 2.40 (s, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 161.9, 147.9, 143.6, 136.6, 135.0, 130.4, 129.1, 127.6, 126.5, 124.9, 121.7, 27.9, 26.2, 21.1.

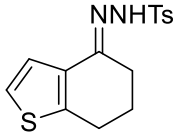
ESI HRMS: calcd. for $C_{16}H_{16}N_2O_2S$ $[M+Na]^+$: 323.0825, found: 323.0834.



***N'*-(3,4-Dihydronaphthalen-1(2*H*)-ylidene)-4-methylbenzenesulfonohydrazide (14a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, was obtained as a white solid (1.42 g, isolated yield: 90%). **14a** was known in the published literature.[10]

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.98 (dd, J=7.8, 1.2 Hz, 1 H), 7.94–7.91 (m, 2 H), 7.76 (s, 1 H), 7.32 (d, J=8.0 Hz, 2 H), 7.24 (dd, J=7.4, 1.5 Hz, 1 H), 7.20 (td, J=7.7, 1.4 Hz, 1 H), 7.09 (d, J=7.4 Hz, 1 H), 2.73–2.70 (m, 2 H), 2.47 (t, J=6.6 Hz, 2 H), 2.41 (s, 3 H), 1.92–1.86 (m, 2 H).

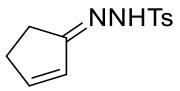
ESI HRMS: calcd. for $C_{17}H_{18}N_2O_2S$ $[M+H]^+$: 315.1162, found: 315.1187.



***N'*-(6,7-dihydrobenzo[b]thiophen-4(5*H*)-ylidene)-4-methylbenzenesulfonohydrazide (15a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, was obtained as a white solid (1.24g, isolated yield: 77%). **15a** was known in the published literature.[11]

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.90 (d, J = 8.3 Hz, 2H), 7.51 (s, 1H), 7.32 (dd, J = 8.3, 6.7 Hz, 3H), 7.01 (d, J = 5.3 Hz, 1H), 2.81 (t, J = 6.1 Hz, 2H), 2.43 (s, 2H), 2.41 (s, 3H), 2.05 – 1.93 (m, 2H).

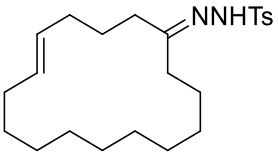
ESI HRMS: calcd. for $C_{15}H_{16}N_2O_2S_2$ $[M+H]^+$: 321.0726, found: 321.0741.



***N*'-(cyclopent-2-en-1-ylidene)-4-methylbenzenesulfonohydrazide (16a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **16a** was obtained as a white solid (1.09 g, isolated yield: 82%). **16a** was known in the published literature.[12]

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.03 (s, 1H), 7.73 (d, J = 8.2 Hz, 2H), 7.38 (d, J = 8.0 Hz, 2H), 6.78 – 6.66 (m, 1H), 6.13 (dd, J = 5.7, 2.1 Hz, 1H), 2.56 – 2.51 (m, 2H), 2.48 – 2.44 (m, 2H), 2.37 (s, 3H).

ESI HRMS: calcd. for $C_{12}H_{14}N_2O_2S$ $[M+Na]^+$: 273.0668, found: 273.0679.

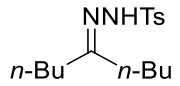


***N*'-(cyclohexadec-3-en-1-ylidene)-4-methylbenzenesulfonohydrazide (17a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **17a** was obtained as a white solid (1.68 g, isolated yield: 83%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.03 (d, J = 9.3 Hz, 1H), 7.70 (dd, J = 8.3, 2.3 Hz, 2H), 7.36 (dd, J = 8.1, 6.2 Hz, 2H), 5.39 – 5.27 (m, 1H), 5.26 – 5.19 (m, 1H), 2.36 (d, J = 3.1 Hz, 3H), 2.20 – 2.10 (m, 2H), 2.09 – 2.03 (m, 2H), 1.98 – 1.95 (m, 2H), 1.91 – 1.84 (m, 2H), 1.46 – 1.37 (m, 2H), 1.37 – 1.28 (m, 4H), 1.27 – 1.11 (m, 12H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 161.8, 143.4, 136.8, 131.6, 130.8, 129.7, 127.9, 35.6, 34.8, 31.8, 31.4, 29.2, 28.3, 28.2, 27.3, 27.1, 26.6, 26.6, 26.0, 25.5, 25.4, 23.0, 21.5.

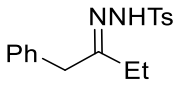
ESI HRMS: calcd. for $C_{23}H_{36}N_2O_2S$ $[M+H]^+$: 405.2570, found: 405.2578.



4-methyl-*N*'-(nonan-5-ylidene)benzenesulfonohydrazide (18a): Prepared according to the synthesis of hydrazone. Following the general procedure, **18a** was obtained as a white solid (1.32 g, isolated yield: 85%). **18a** was known in the published literature.[13]

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.00 (s, 1H), 7.70 (d, J = 8.3 Hz, 2H), 7.37 (d, J = 8.0 Hz, 2H), 2.37 (s, 3H), 2.19 – 2.12 (m, 2H), 2.06 (t, J = 7.3 Hz, 2H), 1.39 – 1.19 (m, 6H), 1.13 – 1.00 (m, 2H), 0.86 (t, J = 7.2 Hz, 3H), 0.75 (t, J = 7.3 Hz, 3H).

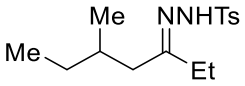
ESI HRMS: calcd. for $C_{16}H_{26}N_2O_2S$ $[M+Na]^+$: 311.1788, found: 311.1769.



***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (19a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **19a** was obtained as a white solid (1.23 g, isolated yield: 78%, E/Z = 56/44). **19a** was known in the published literature.[7]

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.36 (s, 0.5H), 10.12 (s, 0.5H), 7.72 – 7.69 (m, 1H), 7.52 – 7.45 (m, 1H), 7.43 – 7.38 (m, 2H), 7.30 – 7.27 (m, 1H), 7.20 – 7.17 (m, 2H), 7.12 – 7.09 (m, 1H), 6.99 – 6.93 (m, 1H), 3.60 (s, 1H), 3.39 (s, 1H), 2.40 (d, J = 9.6 Hz, 3H), 2.11 (q, J = 7.8 Hz, 1.1H), 1.99 (q, J = 7.3 Hz, 0.9H), 0.90 – 0.78 (m, 3H).

ESI HRMS: calcd. for $C_{17}H_{20}N_2O_2S$ [M+Na]$^+$: 339.1138, found: 339.1146.

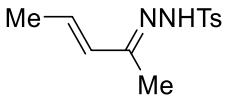


4-methyl-*N*'-(5-methylheptan-3-ylidene)benzenesulfonohydrazide (20a): Prepared according to the synthesis of hydrazone. Following the general procedure, **20a** was obtained as a white solid (1.26 g, isolated yield: 85%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.01 (s, 1H), 7.80 – 7.65 (m, 2H), 7.45 – 7.28 (m, 2H), 2.35 (s, 3H), 2.22 – 2.10 (m, 2H), 2.05 (dd, J = 14.5, 6.5 Hz, 1H), 1.85 (dd, J = 14.4, 7.7 Hz, 1H), 1.60 – 1.47 (m, 1H), 1.14 – 1.09 (m, 1H), 0.93 (t, J = 7.6 Hz, 3H), 0.87 – 0.82 (m, 1H), 0.70 (t, J = 7.4 Hz, 3H), 0.62 (d, J = 6.6 Hz, 3H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 161.8, 142.8, 136.4, 129.1, 127.5, 42.3, 36.2, 31.2, 31.1, 29.4, 28.9, 28.5, 22.6, 21.0, 18.7, 18.7, 11.2, 11.0, 10.5, 9.4.

ESI HRMS: calcd. for $C_{16}H_{24}N_2O_2S$ [M+H]$^+$: 297.1631, found: 297.1635.

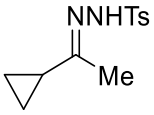


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (21a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **21a** was obtained as a white solid (983 mg, isolated yield: 78%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.26 (s, 1H), 7.72 (d, J = 8.3 Hz, 2H), 7.39 – 7.36 (m, 2H), 6.15 – 6.08 (m, 1H), 5.94 (dd, J = 16.0, 1.6 Hz, 1H), 2.36 (s, 3H), 1.85 (s, 3H), 1.74 (dd, J = 6.6, 1.6 Hz, 3H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 154.5, 143.3, 136.3, 132.2, 131.4, 129.5, 127.6, 21.1, 18.1, 12.3.

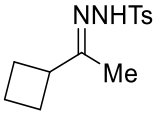
ESI HRMS: calcd. for $C_{12}H_{16}N_2O_2S$ [M+Na]$^+$: 275.0830, found: 275.0842.



***N'*-(1-cyclopropylethylidene)-4-methylbenzenesulfonohydrazide (22a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **22a** was obtained as a white solid (1.19 g, isolated yield: 91%). **22a** was known in the published literature.[15]

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.87 (s, 1H), 7.80 – 7.67 (m, 2H), 7.46 – 7.32 (m, 2H), 2.38 (s, 3H), 1.59 (s, 3H), 1.51 – 1.46 (m, 1H), 0.69 – 0.60 (m, 2H), 0.61 – 0.51 (m, 2H).

ESI HRMS: calcd. for $C_{12}H_{16}N_2O_2S$ [M+H]$^+$: 253.1005, found: 253.1012.

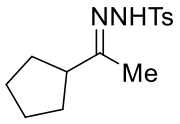


***N'*-(1-cyclobutylethylidene)-4-methylbenzenesulfonohydrazide (23a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **23a** was obtained as a white solid (1.09 mg, isolated yield: 82%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.92 (s, 1H), 7.73 (d, J = 8.2 Hz, 2H), 7.37 (d, J = 8.0 Hz, 2H), 2.96 (p, J = 8.4 Hz, 1H), 2.36 (s, 3H), 2.01 – 1.86 (m, 4H), 1.86 – 1.75 (m, 1H), 1.69 (s, 3H), 1.65 – 1.54 (m, 1H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 161.0, 143.7, 136.9, 129.8, 128.2, 42.1, 25.5, 21.6, 17.8, 14.9.

ESI HRMS: calcd. for $C_{13}H_{18}N_2O_2S$ [M+Na]$^+$: 289.0981, found: 289.0965.

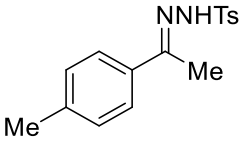


***N'*-(1-cyclopentylethylidene)-4-methylbenzenesulfonohydrazide (24a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **24a** was obtained as a white solid (1.20 g, isolated yield: 86%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.87 (s, 1H), 7.71 (d, J = 8.3 Hz, 2H), 7.37 (d, J = 8.0 Hz, 2H), 2.59 – 2.51 (m, 1H), 2.37 (s, 3H), 1.74 (s, 3H), 1.65 – 1.57 (m, 2H), 1.51 – 1.39 (m, 6H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 161.1, 143.0, 136.3, 129.2, 127.6, 47.2, 29.2, 24.8, 21.0, 15.3.

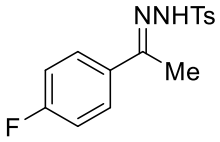
ESI HRMS: calcd. for $C_{14}H_{20}N_2O_2S$ $[M+H]^+$: 281.1318, found: 281.1331.



4-methyl-*N*'-(1-(*p*-tolyl)ethylidene)benzenesulfonohydrazide (26a): Prepared according to the synthesis of hydrazone. Following the general procedure, **26a** was obtained as a white solid (1.27 g, isolated yield: 84%). **26a** was known in the published literature.[16]

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.92 (d, J = 8.5 Hz, 2H), 7.91 (s, 1H), 7.54 (d, J = 8.5 Hz, 2H), 7.31 (d, J = 8.5 Hz, 2H), 7.14 (d, J = 8.5 Hz, 2H), 2.41 (s, 3H), 2.34 (s, 3H), 2.13 (s, 3H).

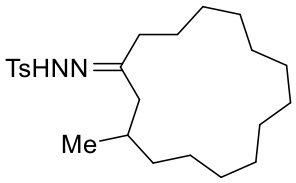
ESI HRMS: calcd. for $C_{16}H_{18}N_2O_2S$ $[M+H]^+$: 303.1162, found: 303.1155.



***N'*-(1-(4-fluorophenyl)ethylidene)-4-methylbenzenesulfonohydrazide (27a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **27a** was obtained as a white solid (1.31 g, isolated yield: 80%). **27a** was known in the published literature.[16]

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.91 (d, J = 8.3 Hz, 2H), 7.72 (s, 1H), 7.66 – 7.58 (m, 2H), 7.33 (d, J = 8.1 Hz, 2H), 7.05 – 6.98 (m, 2H), 2.42 (s, 3H), 2.14 (s, 3H).

ESI HRMS: calcd. for $C_{15}H_{15}FN_2O_2S$ $[M+H]^+$: 329.0911, found: 329.0924.

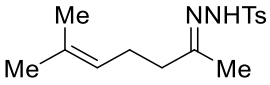


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (39a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **39a** was obtained as a white solid (1.64 g, isolated yield: 81%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.03 (s, 1H), 7.69 (d, J = 8.3 Hz, 2H), 7.35 (dd, J = 8.5, 0.8 Hz, 2H), 2.35 (s, 3H), 2.25 – 2.03 (m, 3H), 1.71 – 1.60 (m, 2H), 1.41 – 1.35 (m, 2H), 1.28 – 1.20 (m, 18H), 1.14 – 1.01 (m, 2H), 0.80 (d, J = 6.5 Hz, 0.6H), 0.55 (d, J = 6.4 Hz, 2.4H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 160.7, 136.4, 129.3, 127.5, 43.3, 35.0, 28.4, 28.3, 27.1, 26.3, 26.2, 26.2, 26.0, 25.9, 25.8, 25.7, 24.4, 23.1, 21.1, 19.6.

ESI HRMS: calcd. for $C_{23}H_{38}N_2O_2S$ $[M+H]^+$: 407.2727, found: 407.2736.

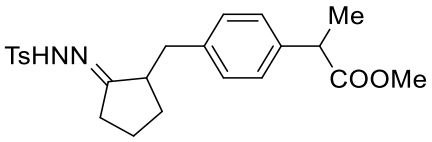


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (40a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **40a** was obtained as a white solid (1.12 g, isolated yield: 76%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.94 (s, 1H), 7.72 (d, J = 8.1 Hz, 2H), 7.37 (d, J = 8.0 Hz, 2H), 5.18 – 4.65 (m, 1H), 2.36 (s, 3H), 2.11 – 2.00 (m, 4H), 1.76 – 1.75 (m, 3H), 1.64 – 1.40 (m, 6H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 157.4, 142.0, 135.4, 130.2, 128.4, 128.3, 126.6, 126.5, 122.3, 37.0, 24.4, 23.2, 20.1, 16.5, 15.5.

ESI HRMS: calcd. for $C_{15}H_{22}N_2O_2S$ $[M+Na]^+$: 317.1294, found: 317.1398.

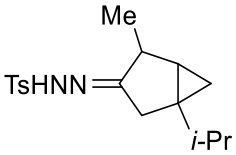


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (41a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, was obtained as a white solid (1.82 g, isolated yield: 85%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.98 (s, 1H), 7.75 (d, J = 8.2 Hz, 2H), 7.39 (d, J = 8.0 Hz, 2H), 7.07 (dd, J = 8.2, 2.1 Hz, 2H), 6.98 (dd, J = 7.9, 1.5 Hz, 2H), 3.72 (q, J = 7.1 Hz, 1H), 3.56 (d, J = 1.3 Hz, 3H), 2.82 (dd, J = 13.7, 4.7 Hz, 1H), 2.62 – 2.56 (m, 1H), 2.40 – 2.39 (m, 1H), 2.38 (s, 3H), 2.36 – 2.27 (m, 1H), 2.17 – 2.10 (m, 1H), 1.73 – 1.59 (m, 2H), 1.55 – 1.42 (m, 1H), 1.34 (dd, J = 7.1, 0.8 Hz, 3H), 1.22 – 1.14 (m, 1H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 174.5, 168.5, 143.2, 138.8, 138.1, 136.5, 129.4, 129.3, 127.6, 127.1, 51.8, 45.5, 44.1, 44.0, 36.6, 30.3, 28.9, 21.9, 21.1, 18.7, 18.6.

ESI HRMS: calcd. for $C_{23}H_{28}N_2O_4S$ $[M+K]^+$: 467.1407, found: 467.1413.

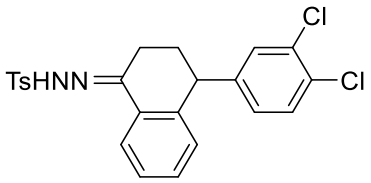


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (42a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **42a** was obtained as a white solid (1.26 g, isolated yield: 79%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.94 (s, 1H), 7.76 – 7.63 (m, 2H), 7.37 (d, J = 8.5 Hz, 2H), 2.82 – 2.77 (m, 0.5H), 2.47 – 2.43 (m, 1H), 2.36 (s, 3H), 2.35 – 2.19 (m, 1H), 1.42 – 1.37 (m, 0.5H), 1.34 – 1.28 (m, 0.5H), 1.26 – 1.22 (m, 0.5H), 1.00 – 0.83 (m, 10H), 0.55 – 0.52 (m, 0.4H), 0.38 – 0.35 (m, 0.6H), 0.21 (dd, J = 5.0, 4.0 Hz, 0.4H), 0.38 (dd, J = 5.1, 4.0 Hz, 0.6H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 168.8, 166.4, 143.0, 136.4, 136.2, 129.3, 129.2, 127.5, 127.3, 42.2, 32.4, 31.5, 31.4, 31.3, 30.2, 29.6, 25.8, 25.3, 21.2, 21.0, 19.9, 19.8, 19.6, 16.7, 14.6, 12.7.

ESI HRMS: calcd. for $C_{17}H_{24}N_2O_2S$ $[M+Na]^+$: 343.1451, found: 343.1456.

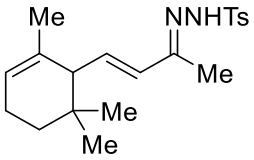


***N*'-(4-(3,4-dichlorophenyl)-3,4-dihydronaphthalen-1(2H)-ylidene)-4-methylbenzenesulfonohydrazide (43a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **43a** was obtained as a white solid (2.04 g, isolated yield: 89%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.07 (dd, J = 7.3, 2.0 Hz, 1H), 7.95 – 7.90 (m, 3H) 7.14 – 7.30 (m, 3H), 7.29 – 7.26 (m, 1H). 7.24 – 7.22 (m, 1H), 7.11 (d, J = 2.1 Hz, 1H), 6.84 (dd, J = 7.2, 1.9 Hz, 1H), 6.80 (dd, J = 8.3, 2.1 Hz, 1H), 4.05 (dd, J = 7.2, 4.3 Hz, 1H), 2.51 – 2.44 (m, 1H), 2.43 (s, 3H), 2.41 – 2.34 (m, 1H), 2.23 – 2.16 (m, 1H), 2.08 – 2.02 (m, 1H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 145.0, 143.4, 140.6, 136.2, 131.8, 131.1, 130.6, 130.3, 129.7, 129.5, 129.1, 128.8, 128.6, 127.6, 127.0, 124.3, 42.7, 28.6, 23.3, 21.0.

ESI HRMS: calcd. for $C_{23}H_{20}Cl_2N_2O_2S$ $[M+H]^+$: 459.0695, found: 459.0702.

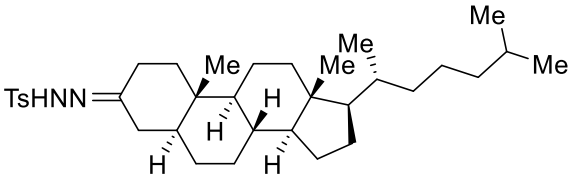


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (44a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **44a** was obtained as a white solid (936 mg, isolated yield: 72%, *E*/*Z* = 90/10).

^{1}H NMR (500 MHz, DMSO-d_6) δ 10.33 (s, 1H), 7.73 (d, J = 8.2 Hz, 1.8H), 7.48 (d, J = 8.2 Hz, 0.2H), 7.38 (d, J = 8.0 Hz, 1.8H), 7.11 (d, J = 8.0 Hz, 0.2H), 6.05 – 5.89 (m, 1H), 5.90 (d, J = 7.9 Hz, 1H), 5.40 (t, J = 3.7 Hz, 1H), 2.37 (s, 3H), 2.28 – 2.16 (m, 1H), 2.07 – 1.93 (m, 2H), 1.87 (s, J = 6.2 Hz, 3H), 1.62 – 1.52 (m, 1H), 1.48 (d, J = 1.9 Hz, 2H), 1.43 – 1.37 (m, 1H), 1.15 – 1.10 (m, 1H), 1.01 – 0.83 (m, 3H), 0.83 – 0.64 (m, 3H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 155.0, 143.8, 138.0, 136.7, 133.4, 131.7, 129.9, 128.0, 121.8, 54.2, 32.4, 31.4, 27.9, 27.1, 23.1, 21.5, 12.9.

ESI HRMS: calcd. for $C_{20}H_{28}N_2O_2S$ $[M+Na]^+$: 383.1769, found: 383.1782.

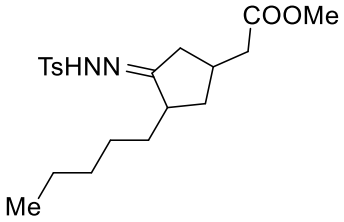


***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (45a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **45a** was obtained as a white solid (1.83 g, isolated yield: 66%, *E*/*Z* = 87/13).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.83 (d, J = 8.0 Hz, 1.7H), 7.80 (d, J = 8.3 Hz, 0.3H), 7.36 (d, J = 8.0 Hz, 0.3H), 7.30 (d, J = 8.1 Hz, 1.7H), 2.45 (s, 0.4H), 2.42 (s, 2.6H), 2.37 – 2.25 (m, 1H), 2.23 – 2.08 (m, 1H), 2.07 – 1.90 (m, 2H), 1.90 – 1.74 (m, 3H), 1.74 – 1.58 (m, 2H), 1.57 – 1.40 (m, 3H), 1.40 – 1.26 (m, 6H), 1.25 – 1.17 (m, 2H), 1.17 – 1.04 (m, 6H), 1.04 – 0.90 (m, 4H), 0.90 – 0.78 (m, 12H), 0.64 (s, 4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 163.1, 144.0, 135.6, 129.6, 128.4, 128.2, 56.5, 56.4, 54.0, 53.9, 46.5, 45.5, 42.7, 40.0, 39.6, 38.4, 37.6, 37.5, 36.3, 36.1, 35.9, 35.5, 31.8, 31.1, 30.1, 29.0, 28.6, 28.4, 28.2, 24.3, 24.0, 23.3, 23.0, 22.7, 21.8, 21.4, 21.3, 18.8, 12.2, 11.6, 11.4.

ESI HRMS: calcd. for $C_{34}H_{54}N_2O_2S$ $[M+H]^+$: 555.3979, found: 555.3983.



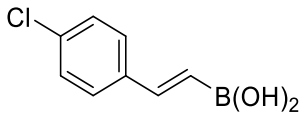
***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (46a)**: Prepared according to the synthesis of hydrazone. Following the general procedure, **46a** was obtained as a white solid (1.64 g, isolated yield: 83%).

^{1}H NMR (500 MHz, DMSO-d_6) δ 9.91 (s, 1H), 7.71 (d, J = 8.2 Hz, 2H), 7.36 (d, J = 8.0 Hz, 2H), 3.57 (s, 3H), 2.43 (dd, J = 15.4, 5.0 Hz, 1H), 2.36 (s, 3H), 2.26 – 2.10 (m, 2H), 2.02 (p, J = 6.7, 6.2 Hz, 1H), 1.99 – 1.82 (m, 2H), 1.42 – 1.33 (m, 1H), 1.33 – 1.19 (m, 3H), 1.17 – 1.08 (m, 3H), 1.03 (q, J = 8.2, 7.7 Hz, 2H), 0.88 (dt, J = 15.7, 4.9 Hz, 1H), 0.81 (t, J = 7.2 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 172.4, 168.1, 142.9, 136.3, 129.2, 129.1, 127.5, 51.2, 48.6, 37.5, 31.6, 29.4, 28.2, 27.6, 25.0, 22.0, 20.9, 13.9, 13.9.

ESI HRMS: calcd. for $C_{20}H_{30}N_2O_4S$ $[M+H]^+$: 395.1999, found: 395.1994.

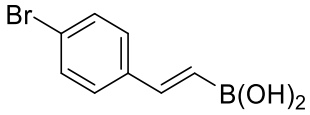
8.2 Characterization data of synthesized alkenyl boronic acids



(4-chlorostyryl)boronic acid (28b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, **28b** was obtained as a white solid (273 mg, isolated yield: 75%). **28b** was known in the published literature.[16]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.85 (s, 2H), 7.54 – 7.36 (m, 4H), 7.23 (d, J = 18.3 Hz, 1H), 6.13 (d, J = 18.3 Hz, 1H).

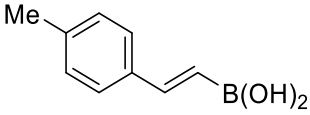
^{13}C NMR (126 MHz, DMSO-d_6) δ 144.3, 136.6, 132.8, 128.7, 128.3.



(4-chlorostyryl)boronic acid (29b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, **29b** was obtained as a white solid (316 mg, isolated yield: 70%).**29b** was known in the published literature.[16]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.84 (s, 2H), 7.55 (d, J = 8.4 Hz, 2H), 7.42 (d, J = 8.5 Hz, 2H), 7.20 (d, J = 18.4 Hz, 1H), 6.14 (d, J = 18.4 Hz, 1H).

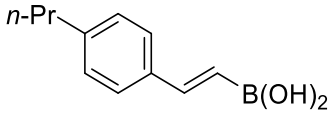
^{13}C NMR (126 MHz, DMSO-d_6) δ 144.4, 136.9, 131.6, 128.6, 121.4.



(4-methylstyryl)boronic acid (30b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, **30b** was obtained as a white solid (259 mg, isolated yield: 80%). **30b** was known in the published literature.[16]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.76 (s, 2H), 7.37 (d, J = 8.0 Hz, 2H), 7.24 (d, J = 18.3 Hz, 1H), 7.16 (d, J = 7.9 Hz, 2H), 6.08 (d, J = 18.4 Hz, 1H), 2.28 (s, 3H).

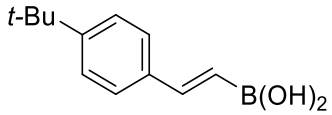
^{13}C NMR (126 MHz, DMSO-d_6) δ 145.8, 137.9, 135.0, 129.3, 126.6, 20.9.



(4-propylstyryl)boronic acid (31b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, was obtained as a white solid (312 mg, isolated yield: 82%). **31b** was known in the published literature.[16]

^{1}H NMR (500 MHz, DMSO-d_6) δ 8.81 (s, 1H), 7.76 (s, 1H), 7.38 (d, J = 8.1 Hz, 2H), 7.24 (s, 1H), 7.21 – 7.13 (m, 2H), 6.06 (d, J = 18.4 Hz, 1H), 2.57 – 2.51 (m, 2H), 1.63 – 1.51 (m, 2H), 0.88 (t, J = 7.3 Hz, 3H).

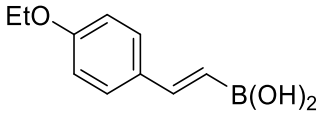
^{13}C NMR (126 MHz, DMSO-d_6) δ 145.8, 137.9, 135.0, 129.3, 126.6, 20.9.



(4-(tert-butyl)styryl)boronic acid (32b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, was obtained as a white solid (322 mg, isolated yield: 79%). **32b** was known in the published literature.[17]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.77 (s, 2H), 7.42 – 7.36 (m, 4H), 7.23 (d, J = 18.3 Hz, 1H), 6.07 (d, J = 18.4 Hz, 1H), 1.26 (s, 9H).

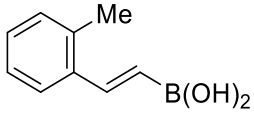
^{13}C NMR (126 MHz, DMSO-d_6) δ 151.1, 145.7, 135.0, 126.4, 125.5, 34.4, 31.1.



(4-ethoxystyryl)boronic acid (33b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, was obtained as a white solid (284 mg, isolated yield: 74%). **33b** was known in the published literature.[17]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.69 (s, 2H), 7.43 – 7.38 (m, 2H), 7.21 (d, J = 18.4 Hz, 1H), 6.93 – 6.88 (m, 2H), 5.96 (d, J = 18.4 Hz, 1H), 4.02 (q, J = 7.0 Hz, 2H), 1.32 (t, J = 7.0 Hz, 3H).

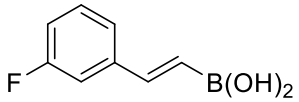
^{13}C NMR (126 MHz, DMSO-d_6) δ 159.3, 146.0, 130.7, 128.4, 115.0, 63.5, 15.1.



(2-methylstyryl)boronic acid (34b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, was obtained as a white solid (253 mg, isolated yield: 78%). **34b** was known in the published literature.[17]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.83 (s, 2H), 7.58 – 7.45 (m, 2H), 7.23 – 7.18 (m, 1H), 7.18 (s, 2H), 6.01 (d, J = 18.3 Hz, 1H), 2.34 (s, 3H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 143.4, 136.7, 135.5, 130.4, 128.1, 126.2, 125.1, 19.3.

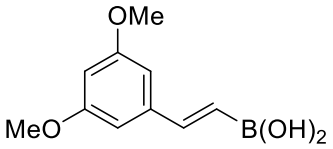


(3-fluorostyryl)boronic acid (35b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, was obtained as a white solid (205 mg, isolated yield: 62%, *E/Z* = 82/18). **35b** was known in the published literature.[16]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.87 (s, 2H), 7. 43 – 7.38 (m, 1.4H), 7.34 – 7.26 (m, 1.6H), 7.22 (dd, J = 18.2, 9.3 Hz, 1H), 7.16 – 7.04 (m, 1H), 6.35 (d, J = 18.1 Hz, 0.2H), 6.17 (d, J = 18.3 Hz, 0.8H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 163.8, 161.3, 144.3 (d, $J_{C\text{-}F}$ = 2.6 Hz), 140.3 (d, $J_{C\text{-}F}$ = 7.3 Hz), 130.6 (d, $J_{C\text{-}F}$ = 8.6 Hz), 122.8 (d, $J_{C\text{-}F}$ = 3.1 Hz), 115.1, 114.9, 112.8, 112.6.

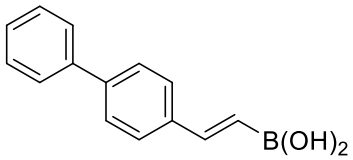
^{19}F NMR (282 MHz, $CDCl_3$) δ = −72.26.



(3,5-dimethoxystyryl)boronic acid (36b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, was obtained as a white solid (300 mg, isolated yield: 72%). **36b** was known in the published literature.[17]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.80 (s, 2H), 7.18 (d, J = 18.3 Hz, 1H), 6.62 (d, J = 2.3 Hz, 2H), 6.45 (t, J = 2.2 Hz, 1H), 6.11 (d, J = 18.3 Hz, 1H), 3.75 (s, 6H).

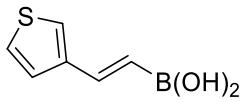
^{13}C NMR (126 MHz, DMSO-d_6) δ 160.7, 145.8, 139.8, 104.5, 100.6, 55.2.



(2-([1,1'-biphenyl]-4-yl)vinyl)boronic acid (37b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, was obtained as a white solid (309 mg, isolated yield: 69%). **37b** was known in the published literature.[17]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.84 (s, 2H), 7.70 – 7.63 (m, 4H), 7.57 (d, J = 8.3 Hz, 2H), 7.47 (dd, J = 8.3, 7.1 Hz, 2H), 7.40 – 7.33 (m, 1H), 7.30 (d, J = 18.4 Hz, 1H), 6.17 (d, J = 18.4 Hz, 1H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 145.4, 140.1, 139.7, 136.8, 129.1, 127.7, 127.3, 127.0, 126.6.

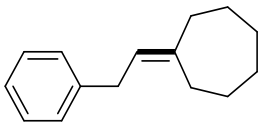


(2-(thiophen-3-yl)vinyl)boronic acid (38b): Prepared according to the synthesis of alkenyl boronic acids. Following the general procedure, was obtained as a white solid (245 mg, isolated yield: 80%). **38b** was known in the published literature.[17]

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.74 (s, 2H), 7.60 – 7.47 (m, 2H), 7.32 (dd, J = 5.0, 1.2 Hz, 1H), 7.24 (d, J = 18.3 Hz, 1H), 5.89 (d, J = 18.4 Hz, 1H).

^{13}C NMR (126 MHz, DMSO-d_6) δ 141.8, 140.4, 127.4, 125.5, 124.9.

8.3 Characterization data of synthesized substrates

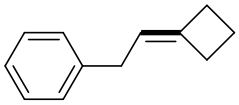


(2-phenylethylidene)cycloheptane (1c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (17.2 mg, isolated yield: 86%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.29 (t, J = 7.6 Hz, 2H), 7.22 – 7.15 (m, 3H), 5.35 (tt, J = 7.3, 1.4 Hz, 1H), 3.35 (d, J = 7.3 Hz, 2H), 2.40 – 2.32 (m, 2H), 2.25 (t, J = 6.0 Hz, 2H), 1.66 – 1.58 (m, 4H), 1.56 – 1.48 (m, 4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 143.7, 143.3, 129.8, 129.8, 127.1, 124.9, 39.3, 35.4, 31.5, 31.4, 30.8, 30.6, 28.6.

EI HRMS: calcd. for $C_{15}H_{20}$: 200.1565, found: 200.1568.

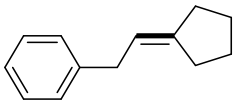


(2-cyclobutylideneethyl)benzene (2c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (5.7 mg, isolated yield: 36%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.30 – 7.28 (m, 2H), 7.22 – 7.17 (m, 3H), 5.28 – 5.23 (m, 1H), 3.22 (d, J = 7.4 Hz, 2H), 2.76 – 2.65 (m, 4H), 1.99 (q, J = 8.0 Hz, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 141.8, 141.3, 128.5, 128.5, 125.9, 119.1, 34.4, 31.0, 29.4, 17.1.

EI HRMS: calcd. for $C_{12}H_{14}$: 158.1096, found: 158.1099.

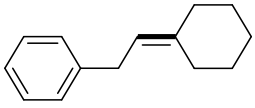


(2-cyclobutylideneethyl)benzene (3c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (8.3 mg, isolated yield: 48%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.31 – 7.26 (m, 2H), 7.22 – 7.15 (m, 3H), 5.48 – 5.43 (m, 1H), 3.33 (d, J = 7.3 Hz, 2H), 2.32 – 2.26 (m, 4H), 1.75 – 1.67 (m, 2H), 1.66 – 1.60 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 144.6, 142.0, 128.5, 128.4, 125.8, 118.8, 36.1, 33.8, 29.0, 26.6, 26.5.

EI HRMS: calcd. for $C_{13}H_{16}$: 172.1252, found: 172.1256.

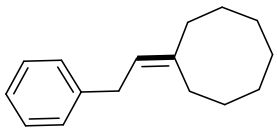


(2-cyclohexylideneethyl)benzene (4c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (14.0 mg, isolated yield: 75%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.30 – 7.27 (m, 2H), 7.20 – 7.18 (m, 3H), 5.28 (tt, J = 7.5, 1.3 Hz, 1H), 3.37 (d, J = 7.4 Hz, 2H), 2.27 – 2.25 (m, 2H), 2.14 – 2.12 (m, 2H), 1.58 – 1.56 (m, 6H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.1, 140.8, 128.5, 128.5, 125.8, 119.9, 37.3, 33.6, 28.9, 28.8, 28.0, 27.1.

EI HRMS: calcd. for $C_{14}H_{18}$: 186.1409, found: 186.1413.

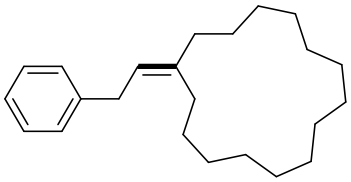


(2-phenylethylidene)cyclooctane (5c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (13.7 mg, isolated yield: 64%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.35 – 7.26 (m, 2H), 7.25 – 7.13 (m, 3H), 5.37 (t, J = 7.3 Hz, 1H), 3.38 (d, J = 7.2 Hz, 2H), 2.39 – 2.26 (m, 2H), 2.25 – 2.14 (m, 2H), 1.73 – 1.61 (m, 4H), 1.56 – 1.48 (m, 6H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.0, 128.4, 128.3, 125.7, 123.7, 37.7, 34.2, 29.2, 27.3, 27.2, 26.3, 26.1.

EI HRMS: calcd. for $C_{16}H_{22}$: 214.1722, found: 214.1729.

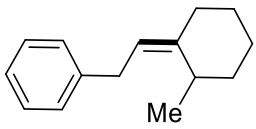


(2-phenylethylidene)cyclopentadecane (6c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (16.8 mg, isolated yield: 54%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.29 – 7.17 (m, 2H), 7.24 – 7.11 (m, 3H), 5.33 (t, J = 7.3 Hz, 1H), 3.37 (d, J = 7.3 Hz, 2H), 2.12 (t, J = 7.7 Hz, 2H), 2.04 (t, J = 7.7 Hz, 2H), 1.52 – 1.43 (m, 4H), 1.41 – 1.30 (m, 20H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.1, 141.4, 128.5, 128. 5, 125.8, 123.5, 37.7, 34.2, 30.2, 28.0, 27.8, 27.5, 27.4, 26.9, 26.9, 26.7, 26.7, 26.6, 26.6.

EI HRMS: calcd. for $C_{23}H_{36}$: 312.2817, found: 312.2813.

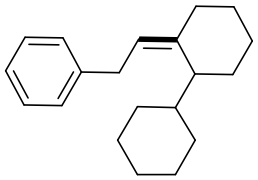


(2-(2-methylcyclohexylidene)ethyl)benzene (7c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (9.8 mg, isolated yield: 49%, E/Z = 70/30).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.38 – 7.26 (m, 2H), 7.21 – 7.16 (m, 3H), 5.35 – 5.16 (m, 1H), 3.39 (d, J = 7.4 Hz, 2H), 3.01 (t, J = 6.1 Hz, 0.3H), 2.62 (dt, J = 13.6, 4.7 Hz, 0.7H), 2.37 – 2.26 (m, 0.3H), 2.14 (d, J = 6.5 Hz, 0.7H), 2.05 – 1.83 (m, 1H), 1.83 – 1.63 (m, 3H), 1.60 (t, J = 2.0 Hz, 0.3H), 1.53 – 1.45 (m, 0.7H), 1.44 – 1.14 (m, 2H), 1.10 (d, J = 7.2 Hz, 1H), 1.04 (d, J = 6.7 Hz, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 144.7, 144.1, 142.2, 142.0, 128.4, 128.3, 126.0, 125.6, 119.7, 117.2, 38.6, 36.79, 33.4, 33.3, 33.2, 32.6, 30.2, 28.6, 28.3, 28.2, 25.5, 21.0, 18.7, 18.2.

EI HRMS: calcd. for $C_{15}H_{20}$: 200.1565, found: 200.1568.

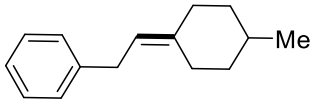


2-(2-phenylethylidene)-1,1'-bi(cyclohexane) (8c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (12.4 mg, isolated yield: 46%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.36 – 7.26 (m, 2H), 7.23 – 7.11 (m, 3H), 5.32 (td, J = 7.4, 1.4 Hz, 0.2H), 5.24 (td, J = 7.4, 1.4 Hz, 0.8H), 3.55 – 3.18 (m, 2H), 2.54 – 2.31 (m, 1H), 2.04 – 1.85 (m, 3H), 1.79 – 1.68 (m, 4H), 1.65 – 1.61 (m, 2H), 1.56 – 1.07 (m, 8H), 1.04 – 0.69 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.6, 142.3, 128.5, 128.4, 125.7, 120.7, 51.4, 35.7, 33.5, 32.1, 31.1, 29.6, 28.3, 26.8, 26.8, 25.7, 22.2.

EI HRMS: calcd. for $C_{20}H_{28}$: 268.2191, found: 268.2196.

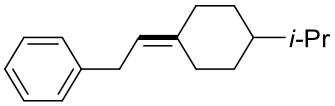


(2-(4-methylcyclohexylidene)ethyl)benzene (9c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (14.2 mg, isolated yield: 71%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.31 – 7.28 (m, 2H), 7.23 – 7.12 (m, 3H), 5.28 (t, J = 7.5 Hz, 1H), 3.43 – 3.27 (m, 2H), 2.73 – 2.66 (m, 1H), 2.29 – 1.99 (m, 2H), 1.91 – 1.70 (m, 3H), 1.64 – 1.55 (m, 1H), 1.11 – 0.94 (m, 2H), 0.92 (d, J = 6.6 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.1, 140.3, 128.5, 125.8, 120.0, 36.9, 36.6, 36.2, 33.7, 33.1, 28.2, 22.3.

EI HRMS: calcd. for $C_{15}H_{20}$: 200.1565, found: 200.1569.

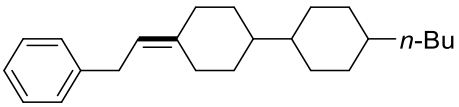


(2-(4-isopropylcyclohexylidene)ethyl)benzene (10c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (9.2 mg, isolated yield: 40%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.29 – 7.26 (m, 2H), 7.19 – 7.16 (m, 3H), 5.26 (t, J = 7.5 Hz, 1H), 3.41 – 3.30 (m, 2H), 2.78 – 2.71 (m, 1H), 2.23 (dd, J = 13.3, 3.0 Hz, 1H), 2.10 – 2.00 (m, 1H), 1.86 – 1.73 (m, 3H), 1.48 – 1.43 (m, 1H), 1.28 – 1.22 (m, 1H), 1.11 – 0.99 (m, 2H), 0.87 (dd, J = 6.8, 1.3 Hz, 6H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.1, 140.7, 128.5, 125.8, 119.7, 44.5, 36.8, 33.7, 32.7, 31.6, 30.9, 28.4, 20.1, 20.1.

EI HRMS: calcd. for $C_{17}H_{24}$: 228.1878, found: 228.1884.

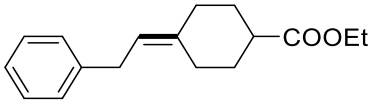


4-butyl-4'-(2-phenylethylidene)-1,1'-bi(cyclohexane) (11c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (19.8 mg, isolated yield: 61%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.39 – 7.26 (m, 2H), 7.21 – 7.16 (m, 3H), 5.27 (t, *J* = 7.5 Hz, 1H), 3.37 (d, *J* = 7.5 Hz, 2H), 2.87 – 2.63 (m, 1H), 2.34 – 2.15 (m, 1H), 2.06 (td, *J* = 13.0, 3.9 Hz, 1H), 1.89 – 1.69 (m, 7H), 1.32 – 1.23 (m,, 6H), 1.18 – 0.97 (m, 7H), 0.92 – 0.83 (m, 5H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.0, 140.7, 128.4, 128.3, 125.7, 119.5, 43.6, 43.0, 37.9, 37.2, 36.8, 33.7, 33.5, 31.8, 31.1, 30.2, 30.2, 29.3, 28.4, 23.1, 14.2.

EI HRMS: calcd. for $C_{24}H_{36}$: 324.2817, found: 324.2819.

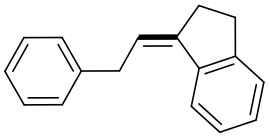


ethyl 4-(2-phenylethylidene)cyclohexane-1-carboxylate (12c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 30:1) to give the title compound as a colorless oil (15.2 mg, isolated yield: 59%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.32 – 7.26 (m, 2H), 7.22 – 7.13 (m, 3H), 5.33 (t, *J* = 7.5 Hz, 1H), 4.13 (q, *J* = 7.1 Hz, 2H), 3.40 – 3.31 (m, 2H), 2.79 – 2.63 (m, 1H), 2.49 (tt, *J* = 11.2, 3.7 Hz, 1H), 2.31 – 2.26 (m, 1H), 2.14 – 2.08 (m, 1H), 2.05 – 1.98 (m, 2H), 1.96 – 1.87 (m, 1H), 1.59 – 1.50 (m, 2H), 1.26 (t, *J* = 7.1 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 175.7, 141.7, 138.4, 128.5, 128.5, 125.9, 121.3, 60.4, 43.4, 35.5, 33.6, 30.6, 29.9, 27.2, 14.4.

EI HRMS: calcd. for $C_{17}H_{22}O_2$: 258.1620, found: 258.1626.

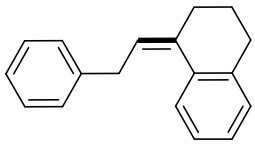


1-(2-phenylethylidene)-2,3-dihydro-1*H*-indene (13c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (13.4 mg, isolated yield: 61%, *E/Z* = 90/10).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.52 – 7.43 (m, 1H), 7.37 – 7.33 (m, 2H), 7.33 – 7.30 (m, 2H), 7.30 – 7.18 (m, 4H), 6.17 (tt, *J* = 7.5, 2.7 Hz, 0.9H), 5.79 (tt, *J* = 7.5, 2.7 Hz, 0.1H), 3.87 (d, *J* = 7.4 Hz, 0.2H), 3.60 (d, *J* = 7.4 Hz, 1.8H), 3.13 – 2.97 (m, 2H), 2.90 – 2.87 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 146.2, 143.2, 141.5, 141.2, 128.6, 128.5, 127.7, 126.5, 126.1, 125.4, 120.1, 117.8, 35.8, 30.2, 28.1.

EI HRMS: calcd. for $C_{17}H_{16}$: 220.1252, found: 220.1258.



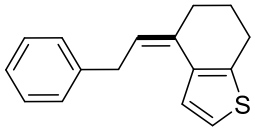
1-(2-phenylethylidene)-1,2,3,4-tetrahydronaphthalene (14c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (15.2 mg, isolated yield: 65%, *E/Z* = 69/31).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.66 – 7.62 (m, 0.7H), 7.45 – 7.42 (m, 0.6H), 7.39 – 7.35 (m, 2.3H), 7.32 – 7.24 (m, 2.4H), 7.24 – 7.12 (m, 3H), 6.25 (t, *J* = 7.4 Hz, 0.7H), 5.68 (t, *J* = 7.4 Hz, 0.3H), 3.77 (d, *J* =

7.4 Hz, 0.7H), 3.62 (d, *J* = 7.4 Hz, 1.3H), 2.91 (t, *J* = 6.7 Hz, 0.6H), 2.86 (t, *J* = 6.2 Hz, 1.4H), 2.67 (t, *J* = 6.5 Hz, 1.4H), 2.53 (t, *J* = 6.5 Hz, 0.6H), 2.03 – 1.90 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 141.9, 141.3, 138.9, 137.6, 137.1, 136.2, 135.8, 135.2, 129.0, 128.8, 128.6, 128.5, 128.0, 127.2, 126.8, 126.1, 126.1, 125.2, 124.4, 123.9, 122.9, 35.7, 34.6, 34.5, 30.7, 29.8, 26.8, 24.6, 23.4.

EI HRMS: calcd. for $C_{18}H_{18}$: 234.1409, found: 234.1415.

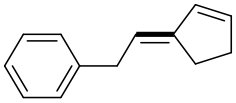


1-(2-phenylethylidene)-1,2,3,4-tetrahydronaphthalene (15c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (19.4 mg, isolated yield: 81%, *E/Z* = 92/8).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.33 – 7.28 (m, 2H), 7.24 – 7.20 (m, 3H), 7.17 – 7.16 (m, 1H), 7.04 – 7.03 (m, 1H), 5.97 (t, *J* = 7.5 Hz, 0.9H), 5.48 (t, *J* = 7.5 Hz, 0.1H), 3.74 (d, *J* = 7.5 Hz, 0.2H), 3.56 (d, *J* = 7.5 Hz, 1.8H), 3.00 – 2.93 (m, 0.2H), 2.89 – 2.86 (m, 1.8H), 2.58 (t, *J* = 6.2 Hz, 1.8H), 2.47 (t, *J* = 6.2 Hz, 0.2H), 2.01 – 1.95 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 141.2, 137.2, 136.3, 132.3, 128.5, 125.9, 123.3, 122.0, 120.7, 33.8, 25.5, 25.4, 23.9.

EI HRMS: calcd. for $C_{16}H_{16}S$: 240.0973, found: 240.0979.

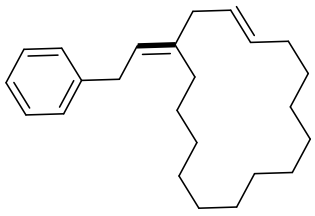


(2-(cyclopent-2-en-1-ylidene)ethyl)benzene (16c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (13.1 mg, isolated yield: 77%, *E/Z* = 73/27).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.32 – 7.27 (m, 2H), 7.26 – 7.13 (m, 3H), 6.54 – 6.51 (m, 0.3H), 6.18 – 6.14 (m, 1H), 6.07 – 6.04 (m, 0.7H), 5.52 (td, *J* = 7.9, 0.7H), 5.34 (t, *J* = 7.9 Hz, 0.3H), 3.48 (d, *J* = 7.8 Hz, 0.5H), 3.41 (d, *J* = 7.5 Hz, 1.5H), 2.57 – 2.51 (m, 4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 148.3, 141.9, 141.6, 139.2, 137.0, 134.6, 130.0, 128.5, 128.5, 128.5, 125.9, 117.8, 116.3, 36.1, 35.8, 32.1, 31.6, 29.4, 26.2.

EI HRMS: calcd. for $C_{13}H_{14}$: 170.1096, found: 170.1098.

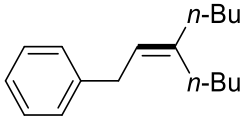


(2-(cyclopent-2-en-1-ylidene)ethyl)benzene (17c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (25.3 mg, isolated yield: 78%, *E/Z* > 99/1).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.40 – 7.26 (m, 2H), 7.20 – 7.15 (m, 3H), 5.45 – 5.39 (m, 1H), 7.35 – 5.31 (m, 2H), 3.38 (d, *J* = 7.3 Hz, 2H), 2.17 – 2.00 (m, 8H), 1.58 – 1.38 (m, 6H), 1.36 – 1.28 (m, 12H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 141.9, 141.2, 131.2, 130.9, 130.1, 129.6, 128.3, 125.7, 122.4, 37.1, 36.9, 36.8, 35.7, 34.0, 32.6, 32.2, 31.9, 31.7, 30.0, 29.7, 28.9, 28.6, 28.2, 28.0, 27.7, 27.4, 26.8, 26.8, 26.7, 26.5, 26.0, 25.6, 25.4.

EI HRMS: calcd. for $C_{24}H_{36}$: 324.2817, found: 324.2813.

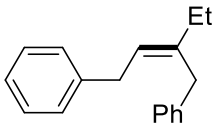


(2-(cyclopent-2-en-1-ylidene)ethyl)benzene (18c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (7.4 mg, isolated yield: 32%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.31 – 7.28 (m, 2H), 7.23 – 7.15 (m, 3H), 5.32 (t, J = 7.3 Hz, 1H), 3.39 (d, J = 7.3 Hz, 2H), 2.14 – 2.11 (m, 2H), 2.06 – 2.03 (m, 2H), 1.44 – 1.37 (m, 4H), 1.36 – 1.29 (m, 4H), 0.95 – 0.89 (m, 6H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.2, 141.1, 128.5, 128.4, 125.8, 122.9, 36.8, 34.1, 31.0, 30.6, 30.1, 23.1, 22.7, 14.2, 14.2.

EI HRMS: calcd. for $C_{17}H_{26}$: 230.2035, found: 230.2042.

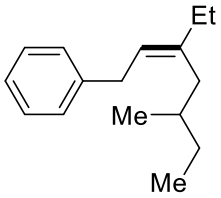


(2-ethylbut-2-ene-1,4-diyl)dibenzene (19c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (12.3 mg, isolated yield: 52%, E/Z = 63/37).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.35 – 7.28 (m, 4H), 7.25 – 7.18 (m, 6H), 5.56 (t, J = 7.3 Hz, 0.6H), 5.42 (t, J = 7.4 Hz, 0.4H), 3.54 (s, 1.2H), 3.53 (d, J = 7.4 Hz, 1.2H), 3.44 (d, J = 7.4 Hz, 0.8H), 3.40 (s, 0.8H), 2.11 (q, J = 7.6 Hz, 0.8H), 2.03 (q, J = 7.4 Hz, 1.2H), 1.02 (t, J = 7.6 Hz, 1.8H), 1.02 (t, J = 7.6 Hz, 1.2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 141.7, 141.6, 140.5, 140.3, 129.1, 128.7, 128.5, 128.5, 128.5, 128.4, 124.9, 123.6, 43.4, 36.4, 34.4, 34.1, 29.5, 22.8, 13.2, 12.8.

EI HRMS: calcd. for $C_{18}H_{20}$: 236.1565, found: 236.1573.

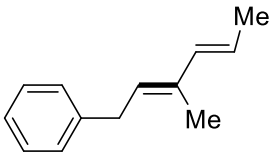


(2-(cyclopent-2-en-1-ylidene)ethyl)benzene (20c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (9.3 mg, isolated yield: 43%, E/Z = 80/20).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.32 – 7.27 (m, 2H), 7.24 – 7.16 (m, 3H), 5.39 (t, J = 7.2 Hz, 0.2H), 5.27 (t, J = 7.3 Hz, 0.8H), 3.40 (d, J = 7.1 Hz, 2H), 2.20 – 2.00 (m, 3H), 1.83 – 1.71 (m, 1H), 1.58 – 1.50 (m, 1H), 1.42 – 1.34 (m, 1H), 1.17 – 1.09 (m, 1H), 1.02 (td, J = 7.5, 2.5 Hz, 3H), 0.93 – 0.88 (m, 3H), 0.86 – 0.84 (m, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.2, 142.1, 141.3, 141.3, 128.6, 128.5, 128.5, 128. 5, 126.1, 125.8, 124.0, 123.0, 44.4, 37.5, 34.2, 34.0, 33.5, 32.7, 29.9, 29.8, 29.7, 23.0, 19.3, 13.6, 13.1, 11.7.

EI HRMS: calcd. for $C_{16}H_{24}$: 216.1878, found: 216.1882.

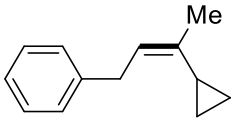


3-methylhexa-2,4-dien-1-yl)benzene (21c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (10.3 mg, isolated yield: 60%, *E/Z* = 67/33).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.35 – 7.29 (m, 2H), 7.24 – 7.20 (m, 3H), 6.61 (dd, *J* = 15.4, 1.5 Hz, 0.3H), 6.14 (dd, *J* = 15.5, 1.5 Hz, 0.7H), 5.84 – 5.80 (m, 0.3H), 5.72 – 5.65 (m, 0.7H), 5.57 (t, *J* = 7.6 Hz, 0.7H), 5.43 (t, *J* = 7.6 Hz, 0.3H), 3.52 (d, *J* = 7.6 Hz, 0.6H), 3.51 (d, *J* = 7.6 Hz, 1.4H), 1.88 – 1.86 (m, 3H), 1.83 – 1.78 (m, 2.6H), 1.73 – 1.68 (m, 0.4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 141.4, 136.0, 134.6, 128.6, 128.5, 128.4, 128.3, 126.5, 126.3, 126.0, 123.1, 34.5, 33.7, 20.9, 18.9, 18.4, 12.7.

EI HRMS: calcd. for $C_{13}H_{16}$: 172.1252, found: 172.1258.

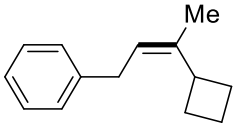


(3-cyclopropylbut-2-en-1-yl)benzene (22c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (14.5 mg, isolated yield: 84%, *E/Z* = 77/23).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.33 – 7.27 (m, 2H), 7.25 – 7.16 (m, 3H), 5.48 – 5.36 (m, 1H), 3.53 (d, *J* = 7.4 Hz, 0.4H), 3.38 (d, *J* = 7.4 Hz, 1.6H), 1.85 – 1.73 (m, 0.2H), 1.62 (d, *J* = 1.2 Hz, 2.3H), 1.48 (d, *J* = 1.4 Hz, 0.7H), 1.46 – 1.36 (m, 0.8H), 0.70 – 0.64 (m, 0.4H), 0.62 – 0.54 (m, 2H), 0.49 – 0.46 (m, 1.6H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 143.3, 143.2, 138.2, 137.3, 129.8, 129.8, 127.1, 127.1, 125.7, 123.0, 35.6, 35.2, 20.4, 20.2, 15.5, 13.8, 5.8, 5.6.

EI HRMS: calcd. for $C_{13}H_{16}$: 172.1252, found: 172.1256.

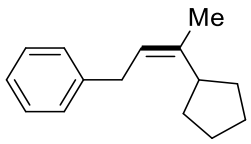


(3-cyclobutylbut-2-en-1-yl)benzene (23c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (11.7 mg, isolated yield: 63%, *E/Z* = 85/15).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.34 – 7.26 (m, 2H), 7.20 – 7.16 (m, 3H), 5.28 (tt, *J* = 7.2, 1.5 Hz, 1H), 3.52 – 3.45 (m, 0.2H), 3.37 (d, *J* = 7.4 Hz, 2H), 2.92 – 2.86 (m, 0.8H), 2.08 – 1.99 (m, 2H), 1.96 – 1.87 (m, 2H), 1.87 – 1.81 (m, 0.6H), 1.78 (d, *J* = 1.4 Hz, 0.6H), 1.73 – 1.66 (m, 1.4H), 1.66 – 1.63 (m, 2.4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.1, 139.8, 128.5, 125.8, 123.1, 120.5, 43.5, 37.3, 34.2, 34.0, 27.4, 20.4, 19.1, 17.9, 14.1.

EI HRMS: calcd. for $C_{14}H_{18}$: 186.1409, found: 186.1414.

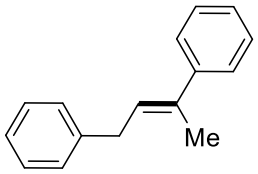


(3-cyclopentylbut-2-en-1-yl)benzene (24c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (15.0 mg, isolated yield: 75%, *E/Z* = 87/13).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.32 – 7.27 (m, 2H), 7.20 – 7.17 (m, 3H), 5.40 (td, *J* = 7.3, 1.3 Hz, 0.8H), 5.33 (td, *J* = 7.3, 1.3 Hz, 0.2H), 3.39 (dd, *J* = 13.8, 7.3 Hz, 2H), 3.04 – 3.00 (m, 0.2H), 2.52 – 2.38 (m, 0.8H), 1.78 – 1.74 (m, 2H), 1.71 (s, 3H), 1.69 – 1.61 (m, 2H), 1.60 – 1.52 (m, 2H), 1.45 – 1.39 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.1, 139.5, 128.5, 128.5, 125.8, 121.3, 49.1, 34.3, 31.2, 25.4, 14.7.

EI HRMS: calcd. for $C_{15}H_{20}$: 200.1565, found: 200.1573.

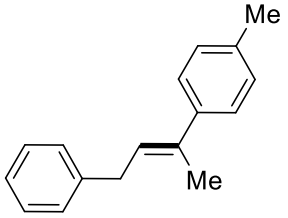


but-2-ene-1,3-diyldibenzene (25c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (9.6 mg, isolated yield: 46%, *E/Z* = 90/10).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.43 – 7.38 (m, 2H), 7.31 – 7.28 (m, 4H), 7.25 – 7.19 (m, 4H), 6.40 (td, *J* = 7.4, 1.5 Hz, 0.1H), 5.97 (td, *J* = 7.4, 1.5 Hz, 0.9H), 3.56 (d, *J* = 7.4 Hz, 2H), 2.14 (s, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 143.8, 141.2, 135.3, 135.4, 128.6, 128.6, 128.3, 127.4, 127.2, 126.9, 126.6, 126.3, 126.1, 125.9, 42.7, 35.1, 21.4, 16.1.

EI HRMS: calcd. for $C_{16}H_{16}$: 208.1252, found: 208.1255.

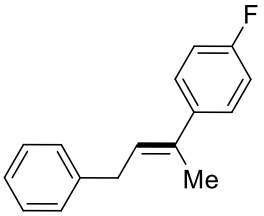


1-methyl-4-(4-phenylbut-2-en-2-yl)benzene (26c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (8.9 mg, isolated yield: 40%, *E/Z* = 98/2).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.37 – 7.34 (m, 3H), 7.33 – 7.32 (m, 1H), 7.30 – 7.27 (m, 2H), 7.24 (s, 1H), 7.17 – 7.14 (m, 2H), 6.04 – 5.94 (m, 0.98H), 5.69 – 5.66 (m, 0.02H), 3.60 (d, *J* = 7.3 Hz, 2H), 2.37 (s, 3H), 2.17 (q, *J* = 1.0 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.7, 141.3, 140.9, 136.5, 135.8, 135.6, 135.6, 129.3, 129.0, 128.6, 128.6, 128.5, 128.4, 128.0, 127.3, 127.1, 126.3, 126.0, 126.0, 125.7, 42.3, 35.1, 21.4, 21.2, 21.2, 18.6, 16.1.

EI HRMS: calcd. for $C_{17}H_{18}$: 222.1409, found: 222.1413.



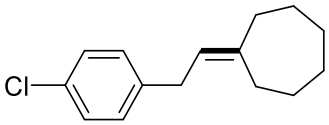
1-fluoro-4-(4-phenylbut-2-en-2-yl)benzene (27c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (11.3 mg, isolated yield: 50%, *E/Z* > 99/1).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.39 – 7.35 (m, 2H), 7.33 – 7.29 (m, 2H), 7.25 – 7.19 (m, 3H), 7.02 – 6.96 (m, 2H), 5.91 (td, J = 7.4, 1.4 Hz, 1H), 3.56 (d, J = 7.3 Hz, 2H), 2.13 (q, J = 1.0 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 162.0 (d, $J_{C\text{-}F}$ = 245.2 Hz), 141.1, 139.8 (d, $J_{C\text{-}F}$ = 3.4 Hz), 134.8, 128.7, 128.6, 127.4 (d, $J_{C\text{-}F}$ = 7.8 Hz), 126.8, 126.8, 126.2, 115.1 (d, $J_{C\text{-}F}$ = 21.2 Hz), 35.1, 16.3.

^{19}F NMR (282 MHz, $CDCl_3$) δ = −116.38.

EI HRMS: calcd. for $C_{16}H_{15}F$: 226.1158, found: 226.1164.

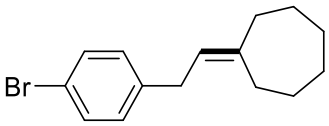


(2-(4-chlorophenyl)ethylidene)cycloheptane (28c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (16.8 mg, isolated yield: 72%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.40 – 7.23 (m, 2H), 7.14 – 7.11 (m, 2H), 5.30 (tt, J = 7.3, 1.4 Hz, 1H), 3.31 (d, J = 7.3 Hz, 2H), 2.48 – 2.08 (m, 4H), 1.66 – 1.51 (m, 8H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 143.0, 140.5, 131.5, 129.8, 128.5, 123.0, 38.0, 33.4, 30.2, 30.1, 29.5, 29.3, 27.3.

EI HRMS: calcd. for $C_{15}H_{19}Cl$: 234.1175, found: 234.1178.

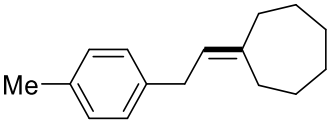


(2-(4-chlorophenyl)ethylidene)cycloheptane (29c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (18.9 mg, isolated yield: 68%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.43 – 7.35 (m, 2H), 7.11 – 7.03 (m, 2H), 5.30 – 5.27 (m, 1H), 3.28 (d, J = 7.2 Hz, 2H), 2.34 – 2.29 (m, 2H), 2.24 (t, J = 5.9 Hz, 2H), 1.61 – 1.58 (m, 4H), 1.54 – 1.51 (m, 4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 144.4, 142.3, 132.8, 131.5, 129.8, 124.1, 120.8, 39.2, 34.7, 31.5, 31.3, 30.7, 30.5, 28.5.

EI HRMS: calcd. for $C_{15}H_{19}Br$: 278.0670, found: 278.0676.



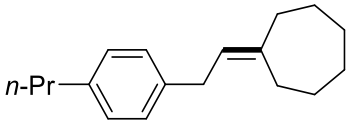
(2-(4-chlorophenyl)ethylidene)cycloheptane (30c): Prepared according to the general procedure C for

the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (13.9 mg, isolated yield: 65%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.09 (s, 4H), 5.33 (t, J = 7.3 Hz, 1H), 3.31 (d, J = 7.2 Hz, 2H), 2.39 – 2.33 (m, 2H), 2.32 (s, 3H), 2.28 – 2.20 (m, 2H), 1.66 – 1.57 (m, 4H), 1.57 – 1.52 (m, 4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.3, 140.1, 136.2, 130.0, 128.6, 125.9, 125.9, 122.8, 37.8, 31.7, 30.2, 30.0, 29.5, 29.3, 27.0, 19.5.

EI HRMS: calcd. for $C_{16}H_{22}$: 214.1722, found: 214.1728.

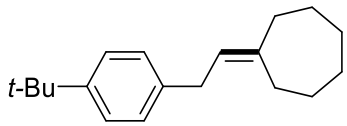


(2-(4-chlorophenyl)ethylidene)cycloheptane (31c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (21.1 mg, isolated yield: 87%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.12 – 7.09 (m, 4H), 5.39 – 5.28 (m, 1H), 3.31 (d, J = 7.2 Hz, 2H), 2.59 – 2.52 (m, 2H), 2.39 – 2.31 (m, 2H), 2.30 – 2.20 (m, 2H), 1.68 – 1.58 (m, 6H), 1.55 – 1.51 (m, 4H), 0.95 (t, J = 7.4 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.1, 140.1, 139.2, 128.6, 128.3, 123.9, 38.0, 37.8, 33.7, 30.2, 30.1, 29.5, 29.3, 27.3, 24.8, 14.0.

EI HRMS: calcd. for $C_{18}H_{26}$: 242.2035, found: 242.2042.

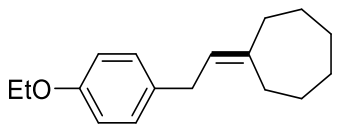


(2-(4-(*tert*-butyl)phenyl)ethylidene)cycloheptane (32c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (16.6 mg, isolated yield: 65%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.38 – 7.33 (m, 2H), 7.18 (d, J = 8.1 Hz, 2H), 5.46 – 5.31 (m, 1H), 3.36 (d, J = 7.3 Hz, 2H), 2.39 (t, J = 6.0 Hz, 2H), 2.29 (t, J = 6.1 Hz, 2H), 1.70 – 1.62 (m, 4H), 1.60 – 1.53 (m, 4H), 1.35 (s, 9H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 148.6, 142.1, 138.9, 128.1, 125.4, 123.8, 38.0, 34.5, 33.5, 31.6, 30.2, 30.1, 29.5, 29.3, 27.3.

EI HRMS: calcd. for $C_{19}H_{28}$: 256.2191, found: 256.2197.

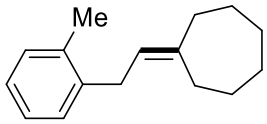


(2-(4-(*tert*-butyl)phenyl)ethylidene)cycloheptane (33c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (13.7 mg, isolated yield: 56%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.10 (d, J = 8.6 Hz, 2H), 6.86 – 6.80 (m, 2H), 5.32 (tt, J = 7.2, 1.3 Hz, 1H), 4.01 (q, J = 7.0 Hz, 2H), 3.28 (d, J = 7.2 Hz, 2H), 2.36 – 2.32 (m, 2H), 2.27 – 2.22 (m, 2H), 1.65 – 1.58 (m, 4H), 1.55 – 1.52 (m, 4H), 1.41 (t, J = 7.0 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 157.2, 142.0, 134.0, 129.3, 124.1, 114.6, 63.6, 38.0, 33.1, 30.2, 30.1, 29.5, 29.3, 27.3, 15.1.

EI HRMS: calcd. for $C_{17}H_{24}O$: 244.1827, found: 244.1834.

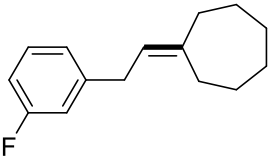


(2-(4-chlorophenyl)ethylidene)cycloheptane (34c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (7.1 mg, isolated yield: 33%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.20 – 7.12 (m, 3H), 7.12 – 7.08 (m, 1H), 5.25 (t, J = 7.0 Hz, 1H), 3.29 (d, J = 7.0 Hz, 2H), 2.37 – 2.33 (m, 2H), 2.30 (s, 3H), 2.26 – 2.21 (m, 2H), 1.67 – 1.61 (m, 2H), 1.60 – 1.57 (m, 2H), 1.55 – 1.49 (m, 4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.3, 140.1, 136.2, 130.0, 128.6, 125.9, 125.9, 122.8, 37.8, 31.7, 30.2, 30.0, 29.5, 29.3, 27.0, 19.5.

EI HRMS: calcd. for $C_{16}H_{22}$: 214.1722, found: 214.1725.



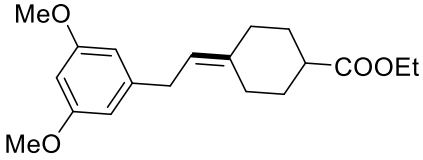
(2-(3-fluorophenyl)ethylidene)cycloheptane (35c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (16.8 mg, isolated yield: 77%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.25 – 7.21 (m, 1H), 6.97 (d, J = 7.6 Hz, 1H), 6.94 – 6.80 (m, 2H), 5.32 (tt, J = 7.3, 1.4 Hz, 1H), 3.34 (d, J = 7.3 Hz, 2H), 2.39 – 2.21 (m, 4H), 1.70 – 1.58 (m, 4H), 1.55 – 1.52 (m, 4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 163.10 (d, $J_{C\text{-}F}$ = 245.1 Hz), 144.65 (d, $J_{C\text{-}F}$ = 6.9 Hz), 143.18, 129.76 (d, $J_{C\text{-}F}$ = 8.3 Hz), 124.08 (d, $J_{C\text{-}F}$ = 2.7 Hz), 122.67, 115.26 (d, $J_{C\text{-}F}$ = 21.0 Hz), 112.62 (d, $J_{C\text{-}F}$ = 21.1 Hz), 37.95, 33.74, 30.15 (d, $J_{C\text{-}F}$ = 20.4 Hz), 29.37 (d, $J_{C\text{-}F}$ = 23.9 Hz), 27.25.

^{19}F NMR (282 MHz, $CDCl_3$) δ = −115.14.

EI HRMS: calcd. for $C_{15}H_{19}F$: 218.1471, found: 218.1476.

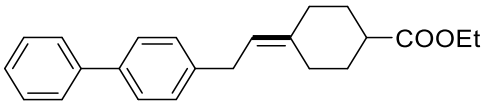


ethyl 4-(2-(3,5-dimethoxyphenyl)ethylidene)cyclohexane-1-carboxylate (36c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 10:1) to give the title compound as a colorless oil (14.6 mg, isolated yield: 46%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 6.34 (d, J = 2.3 Hz, 2H), 6.30 (t, J = 2.3 Hz, 1H), 5.31 (t, J = 7.6 Hz, 1H), 4.13 (q, J = 7.1 Hz, 2H), 3.77 (s, 6H), 3.29 (t, J = 6.7 Hz, 2H), 2.69 (dt, J = 13.8, 3.7 Hz, 1H), 2.48 (tt, J = 11.1, 3.7 Hz, 1H), 2.28 (dt, J = 13.6, 3.7 Hz, 1H), 2.10 (td, J = 13.0, 3.5 Hz, 1H), 2.03 – 1.98 (m, 2H), 1.91 (td, J = 13.0, 3.8 Hz, 1H), 1.61 – 1.52 (m, 2H), 1.25 (t, J = 7.1 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 175.6, 160.9, 144.1, 138.6, 120.9, 106.5, 97.9, 60.4, 55.4, 43.4, 35.5, 33.8, 30.7, 29.9, 27.2, 14.4.

EI HRMS: calcd. for $C_{19}H_{26}O_4$: 318.1831, found: 318.1835.

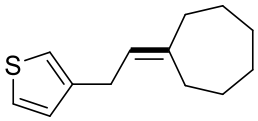


methyl 4-(2-([1,1'-biphenyl]-4-yl)ethylidene)cyclohexane-1-carboxylate (37c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 15:1) to give the title compound as a colorless oil (16.7 mg, isolated yield: 50%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.60 – 7.57 (m, 2H), 7.54 – 7.51 (m, 2H), 7.45 – 7.42 (m, 2H), 7.35 – 7.31 (m, 1H), 7.25 (d, J = 8.1 Hz, 2H), 5.37 (t, J = 7.5 Hz, 1H), 4.15 (q, J = 7.1 Hz, 2H), 3.45 – 3.36 (m, 2H), 2.75 (dt, J = 13.9, 3.7 Hz, 1H), 2.51 (tt, J = 11.1, 3.7 Hz, 1H), 2.36 – 2.27 (m, 1H), 2.13 (td, J = 12.9, 3.3 Hz, 1H), 2.07 – 2.00 (m, 2H), 1.95 (td, J = 13.0, 3.9 Hz, 1H), 1.65 – 1.60 (m, 2H), 1.27 (t, J = 7.1 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 175.7, 141.2, 140.8, 139.0, 138.5, 128.9, 127.3, 127.2, 121.1, 60.4, 43.4, 35.6, 33.3, 30.6, 29.9, 27.2, 14.4.

EI HRMS: calcd. for $C_{23}H_{26}O_2$: 334.1933, found: 334.1938.

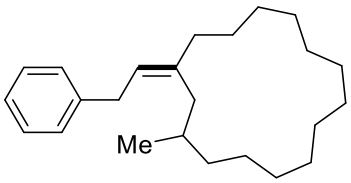


3-(2-cycloheptylideneethyl)thiophene (38c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (8.9 mg, isolated yield: 43%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.25 – 7.24 (m, 1H), 6.95 – 6.93 (m, 2H), 5.37 (tt, J = 7.3, 1.4 Hz, 1H), 3.33 (d, J = 7.2 Hz, 2H), 2.36 – 2.29 (m, 2H), 2.28 – 2.21 (m, 2H), 1.61 – 1.58 (m, 4H), 1.55 – 1.50 (m, 4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.5, 142.3, 128.5, 125.4, 122.9, 120.1, 37.9, 30.1, 30.1, 29.5, 29.3, 28.7, 27.3.

EI HRMS: calcd. for $C_{13}H_{18}S$: 206.1129, found: 206.1124.

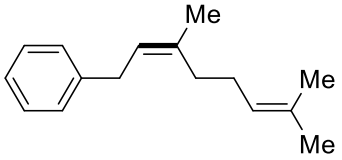


1-methyl-3-(2-phenylethylidene)cyclopentadecane (39c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (12.7 mg, isolated yield: 39%, E/Z = 77/23).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.32 – 7.27 (m, 2H), 7.23 – 7.15 (m, 3H), 5.41 (t, J = 7.3 Hz, 0.2H), 5.30 (t, J = 7.3 Hz, 0.8H), 3.45 – 3.33 (m, 2H), 2.27 – 2.16 (m, 1H), 2.04 – 1.94 (m, 1H), 1.70 – 1.62 (m, 1H), 1.47 – 1.27 (m, 22H), 1.24 – 1.16 (m, 2H), 0.87 (d, J = 6.3 Hz, 1H), 0.82 (d, J = 6.3 Hz, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.2, 142.1, 140.2, 140.1, 128.5, 128.4, 125.8, 124.9, 124.4, 45.5, 37.4, 36.7, 36.2, 35.9, 34.3, 34.2, 30.3, 29.6, 29.4, 28.2, 27.9, 27.5, 27.3, 27.2, 27.1, 27.0, 26.9, 26.9, 26.8, 26.8, 26.7, 26.7, 26.6, 26.6, 26.5, 25.5, 25.4, 20.5, 20.3.

EI HRMS: calcd. for $C_{24}H_{38}$: 326.2974, found: 326.2978.

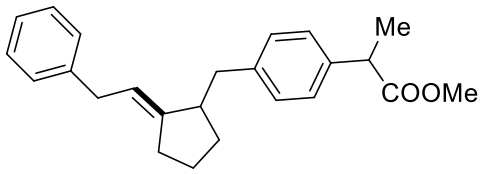


(3,7-dimethylocta-2,6-dien-1-yl)benzene (40c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (13.9 mg, isolated yield: 65%, *E/Z* = 66/34).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.31 – 7.28 (m, 2H), 7.21 – 7.18 (m, 3H), 5.39 – 5.33 (m,1H), 5.20 – 5.15 (m, 0.3H), 5.15 – 5.11 (m, 0.7H), 3.38 (dd, *J* = 7.5, 3.2 Hz, 2H), 2.21 – 2.04 (m, 4H), 1.77 (d, *J* = 1.3 Hz, 1H), 1.73 (d, *J* = 1.3 Hz, 2H), 1.71 – 1.70 (m, 3H), 1.64 (d, *J* = 1.3 Hz, 1H), 1.62 (d, *J* = 1.2 Hz, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 141.8, 136.2, 131.8, 131.5, 125.7, 125.7, 124.3, 124.2, 123.9, 123.0, 39.7, 34.2, 34.1, 32.0, 26.6, 25.8, 23.5, 17.7, 17.7, 16.1.

EI HRMS: calcd. for $C_{16}H_{22}$: 214.1722, found: 214.1725.

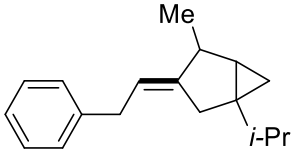


methyl-2-(4-((2-(2-phenylethylidene)cyclopentyl)methyl)phenyl)propanoate (41c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 30:1) to give the title compound as a colorless oil (17.4 mg, isolated yield: 50%, *E/Z* = 76/24).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.41 – 7.29 (m, 1.7H), 7.26 (s, 0.3H), 7.25 – 7.21 (m, 4H), 7.21 – 7.18 (m, 1H), 7.16 (d, *J* = 8.1 Hz, 2H), 5.49 – 5.44 (m, 0.8H), 5.38 (p, *J* = 1.7 Hz, 0.2H), 3.79 – 3.71 (m, 1H), 3.71 – 3.68 (m, 3H), 3.41 (d, *J* = 6.3 Hz, 2H), 2.96 (dd, *J* = 13.5, 5.1 Hz, 0.8H), 2.85 (dd, *J* = 13.6, 5.3 Hz, 0.2H), 2.57 – 2.40 (m, 2H), 2.40 – 2.28 (m, 1H), 2.26 – 2.23 (m, 0.5H), 1.93 – 1.87 (m, 0.5H), 1.86 – 1.67 (m, 2H), 1.66 – 1.56 (m, 1H), 1.55 – 1.51 (m, 3H), 1.41 – 1.29 (m, 1H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 175.3, 147.2, 147.1, 143.7, 141.7, 140.5, 140.2, 139.1, 138.1, 138.0, 129.5, 129.4, 129.2, 128.5, 128.4, 128.4, 127.4, 127.3, 125.9, 125.6, 120.1, 119.2, 52.1, 46.2, 45.1, 42.3, 40.6, 40.4, 37.6, 35.9, 35.4, 34.9, 33.0, 32.7, 32.6, 31.5, 29.6, 24.0, 23.6, 23.6, 18.7.

EI HRMS: calcd. for $C_{24}H_{28}O_2$: 348.2089, found: 348.2096.



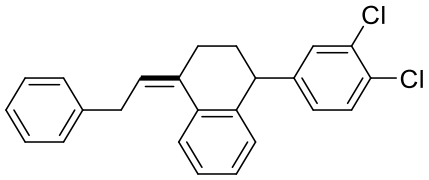
1-isopropyl-4-methyl-3-(2-phenylethylidene)bicyclo[3.1.0]hexane (42c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (19.7 mg, isolated yield: 82%, *E/Z* = 55/45).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.30 – 7.26 (m, 2H), 7.22 – 7.12 (m, 3H), 5.44 – 5.30 (m, 0.6H), 5.29 – 5.20 (m, 0.4H), 3.51 – 3.33 (m, 1.2H), 3.32 – 3.20 (m, 0.8H), 2.63 – 2.43 (m, 1H), 2.39 – 2.23 (m, 1H), 1.51 – 1.34 (m, 1H), 1.23 (d, *J* = 6.7 Hz, 1H), 1.20 (dt, *J* = 8.4, 4.3 Hz, 0.5H), 1.13 (dt, *J* = 8.1, 4.2 Hz, 0.5H), 1.07 (d, *J* = 7.1 Hz, 1H), 1.04 – 0.96 (m, 4H), 0.95 (d, *J* = 6.4 Hz, 1H), 0.94 – 0.90 (m, 2H), 0.85

(d, *J* = 6.9 Hz, 1H), 0.41 – 0.29 (m, 0.6H), 0.24 – 0.21 (m, 0.4H), 0.14 (t, 4.2 Hz, 0.6H), 0.02 (t, *J* = 4.2 Hz, 0.4H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 147.3, 145.4, 145.2, 141.7, 128.3, 128.3, 128.3, 128.2, 125.7, 122.5, 121.8, 120.9, 43.4, 39.6, 39.2, 37.9, 35.5, 35.4, 34.5, 32.9, 32.2, 30.6, 30.3, 29.5, 27.9, 27.6, 23.1, 20.3, 20.0, 19.9, 19.8, 18.0, 15.7, 15.3, 12.5, 11.1.

EI HRMS: calcd. for $C_{18}H_{24}$: 240.1878, found: 240.1161.

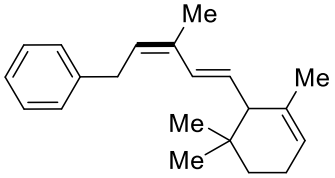


1-(3,4-dichlorophenyl)-4-(2-phenylethylidene)-1,2,3,4-tetrahydronaphthalene (43c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (27.6 mg, isolated yield: 73%, *E/Z* = 62/38).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.67 (dd, *J* = 7.9, 1.3 Hz, 0.4H), 7.47 (dd, *J* = 7.6, 1.5 Hz, 0.6H), 7.42 – 7.39 (m, 0.4H), 7.37 – 7.34 (m, 1.6H), 7.34 – 7.31 (m, 1H), 7.31 – 7.29 (m, 0.3H), 7.27 – 7.26 (m, 0.7H), 7.26 – 7.20 (m, 4H), 7.19 – 7.11 (m, 1H), 6.98 (dd, *J* = 8.3, 2.1 Hz, 0.6H), 6.94 (dd, *J* = 8.3, 2.2 Hz, 0.4H), 6.90 – 6.88 (m, 0.6H), 6.89 – 6.86 (m, 0.4H). 6.31 – 6.26 (m, 0.4H), 5.72 – 5.67 (m, 0.6H), 4.18 – 4.13 (m, 1H), 3.78 (t, *J* = 6.5 Hz, 1H), 3.57 (d, *J* = 7.4 Hz, 1H), 2.65 – 2.48 (m, 2H), 2.37 – 2.31 (m, 0.6H), 2.24 – 2.18 (m, 0.4H), 2.05 – 1.94 (m, 1H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 147.2, 146.0, 141.6, 140.9, 139.9, 138.1, 136.6, 136.4, 135.9, 134.5, 132.5, 130.6, 130.5, 130.4, 130.2, 129.7, 129.5, 128.7, 128.5, 128.5, 128.2, 128.2, 127.6, 127.2, 127.1, 126.3, 126.2, 126.1, 125.0, 124.2, 124.0, 45.7, 45.3, 35.6, 34.5, 34.3, 33.3, 31.8, 23.9.

EI HRMS: calcd. for $C_{24}H_{20}Cl_2$: 378.0942, found: 378.0948.

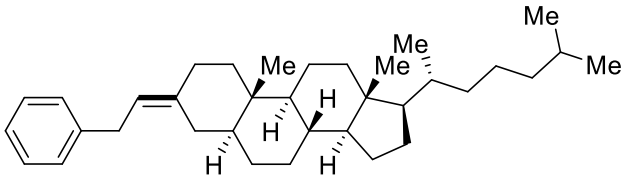


3-methyl-5-(2,6,6-trimethylcyclohex-2-en-1-yl)penta-2,4-dien-1-yl)benzene (44c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (10.4 mg, isolated yield: 37%, *E/Z* = 81/19).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.34 – 7.27 (m, 2H), 7.24 – 7.14 (m, 3H), 6.53 (d, *J* = 15.4 Hz, 0.2H), 6.05 (d, *J* = 15.5 Hz, 0.8H), 5.66 – 5.51 (m, 1H), 5.47 – 5.37 (m, 2H), 3.53 (d, *J* = 7.7 Hz, 0.4H), 3.50 (d, *J* = 7.5 Hz, 1.6H), 2.22 (d, *J* = 9.5 Hz, 0.2H), 2.14 (d, *J* = 9.5 Hz, 0.8H), 2.02 (dd, *J* = 3.6, 1.9 Hz, 2H), 1.86 (d, *J* = 1.2 Hz, 0.7H), 1.85 (d, *J* = 1.2 Hz, 2.2H), 1.62 (d, *J* = 1.9 Hz, 0.6H), 1.60 (d, *J* = 1.9 Hz, 2.4H), 1.51 – 1.41 (m, 1H), 1.24 – 1.14 (m, 1H), 0.92 (d, *J* = 10.9 Hz, 3H), 0.84 (d, *J* = 9.6 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 141.5, 141.4, 136.0, 134.8, 134.5, 133.1, 132.5, 129.2, 128.8, 128.6, 128.6, 128.5, 126.8, 126.0, 126.0, 121.0, 120.8, 55.3, 54.8, 34.7, 33.8, 32.5, 31.9, 27.8, 27.2, 23.3, 23.2, 23.2, 21.0, 12.9.

EI HRMS: calcd. for $C_{21}H_{28}$: 280.2191, found: 280.2188.

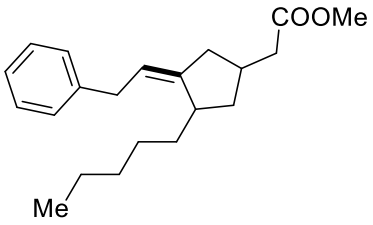


(8*R*,13*R*)-8,13-dimethyl-17-((*R*)-6-methylheptan-2-yl)-3-(2-phenylethylidene)hexadecahydro-1*H*-cyclopenta[a]phenanthrene (45c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (29.4 mg, isolated yield: 62%, *E/Z* > 99/1).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.30 – 7.27 (m, 2H), 7.22 – 7.14 (m, 3H), 5.25 (tt, *J* = 7.3, 2.3 Hz, 1H), 3.45 – 3.24 (m, 2H), 2.65 – 2.53 (m, 1H), 2.40 – 2.19 (m, 1H), 2.13 – 2.03 (m, 1H), 1.99 – 1.95 (m, 1H), 1.84 – 1.75 (m, 3H), 1.70 – 1.63 (m, 1H), 1.59 – 1.55 (m, 1H), 1.54 – 1.48 (m, 2H), 1.42 – 1.29 (m, 6H), 1.28 – 1.18 (m, 3H), 1.17 – 1.06 (m, 6H), 1.06 – 0.95 (m, 4H), 0.93 – 0.89 (m, 6H), 0.87 (dd, *J* = 6.6, 2.3 Hz, 7H), 0.67 (s, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.2, 140.6, 140.5, 128.5, 125.8, 119.6, 119.4, 56.7, 56.4, 54.6, 54.6, 48.5, 47.7, 42.8, 40.2, 39.7, 36.7, 36.7, 36.3, 36.0, 35.7, 33.6, 32.8, 32.2, 32.2, 31.6, 29.3, 29.0, 28.4, 28.2, 24.6, 24.4, 24.0, 2300, 22.7, 21.3, 21.3, 18.8, 12.3, 12.0, 12.0.

EI HRMS: calcd. for $C_{35}H_{54}$: 474.4226, found: 474.4231.

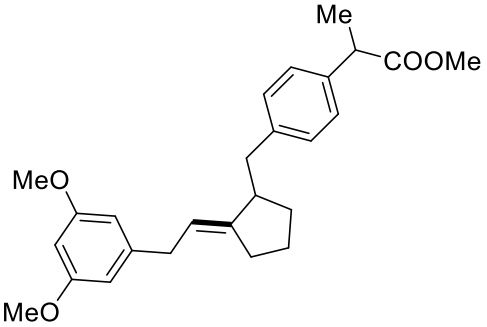


methyl-2-(3-pentyl-4-(2-phenylethylidene)cyclopentyl)acetate (46c): Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 30:1) to give the title compound as a colorless oil (13.2 mg, isolated yield: 42%, *E/Z* > 99/1).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.37 – 7.26 (m, 2H), 7.19 – 7.26 (m, 3H), 5.40 – 5.37 (m, 1H), 3.68 (s, 3H), 3.35 (d, *J* = 7.3 Hz, 2H), 2.44 (dd, *J* = 14.4, 5.1 Hz, 2H), 2.35 – 2.19 (m, 2H), 2.18 – 2.11 (m, 1H), 2.08 – 2.06 (m, 1H), 2.03 – 1.92 (m, 1H), 1.49 – 1.38 (m, 2H), 1.34 – 1.20 (m, 7H), 0.87 (t, *J* = 7.0 Hz, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 175.1, 147.7, 143.0, 129.8, 129.7, 127.2, 121.2, 52.9, 51.5, 42.0, 40.6, 37.0, 33.7, 31.5, 28.9, 27.6, 24.1, 15.5.

EI HRMS: calcd. for $C_{21}H_{30}O_2$: 314.2246, found: 314.2240.



methyl-2-(4-((2-(2-(3,5-dimethoxyphenyl)ethylidene)cyclopentyl)methyl)phenyl)propanoate (47c):

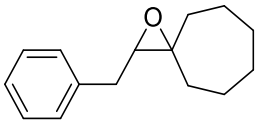
Prepared according to the general procedure C for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 30:1) to give the title compound as a colorless oil (18.7 mg, isolated yield: 46%, *Z/E* = 85/15).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.23 – 7.16 (m, 2H), 7.16 – 7.08 (m, 2H), 6.35 (dd, *J* = 16.7, 2.2 Hz, 2H), 6.30 (dt, *J* = 10.7, 2.3 Hz, 1H), 5.43 – 5.39 (m, 1H), 3.78 (d, *J* = 8.9 Hz, 6H), 3.71 – 3.68 (m, 1H), 3.66 – 3.64 (m, 3H), 3.28 (dd, *J* = 22.4, 7.7 Hz, 1.7H), 3.12 (dd, *J* = 15.8, 6.4 Hz, 0.3H), 2.93 (dd, *J* = 13.5, 4.9 Hz, 1H), 2.80 – 2.77 (m, 0.4H), 2.61 – 2.58 (m, 0.7H), 2.52 – 2.16 (m, 3H), 1.83 – 1.64 (m, 2H), 1.51 – 1.45 (m, 3H), 1.35 – 1.19 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 175.2, 160.8, 147.5, 144.1, 140.4, 140.1, 138.1, 137.8, 129.4, 129.3, 127.3, 127.2, 119.6, 118.6, 106.4, 106.4, 97.6, 55.3, 52.0, 46.1, 45.0, 42.2, 40.5, 40.3, 36.0, 35.5, 32.9, 32.6, 31.4, 29.5, 23.8, 23.5, 18.6.

EI HRMS: calcd. for $C_{26}H_{32}O_4$: 408.2301, found: 408.2307.

8.4 Characterization data of further transformation products

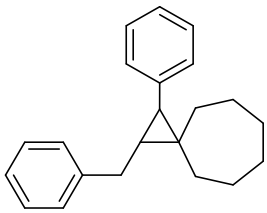


2-benzyl-1-oxaspiro[2.6]nonane (1d): Prepared according to the general procedure D for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 20:1) to give the title compound as a colorless oil (177.4 mg, isolated yield: 82%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.35 – 7.30 (m, 2H), 7.30 – 7.21 (m, 3H), 2.97 (t, *J* = 6.2 Hz, 1H), 2.94 – 2.80 (m, 2H), 1.96 – 1.89 (m, 1H), 1.84 – 1.79 (m, 1H), 1.75 – 1.66 (m, 5H), 1.64 – 1.49 (m, 5H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 138.4, 128.8, 128.7, 126.5, 65.5, 64.6, 37.6, 35.3, 31.7, 29.4, 29.3, 24.8, 24.6.

EI HRMS: calcd. for $C_{15}H_{20}O$: 216.1514, found: 216.1519.

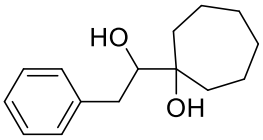


1-benzyl-2-phenylspiro[2.6]nonane (1e): Prepared according to the general procedure E for the alkenylation. Following workup, the product was purified by column chromatography (hexanes) to give the title compound as a colorless oil (24.0 mg, isolated yield: 42%, d.r. = 1:1.1).

^{1}H NMR (500 MHz, DMSO-d_6) δ 7.65 – 7.54 (m, 0.5H), 7.43 – 7.34 (m, 0.5H), 7.30 – 7.26 (m, 4.5H), 7.22 – 7.13 (m, 3.5H), 7.11 – 7.08 (m, 1H), 2.93 – 2.88 (m, 1H), 2.70 (dd, *J* = 15.0, 8.2 Hz, 0.5H), 2.47 (dd, *J* = 15.0, 8.2 Hz, 0.5H), 1.94 (d, *J* = 9.1 Hz, 0.5H), 1.80 (d, *J* = 6.1 Hz, 0.5H), 1.74 – 1.69 (m, 1H), 1.64 – 1.60 (m, 2H), 1.57 – 1.39 (m, 6H), 1.39 – 1.29 (m, 2H), 1.24 – 1.17 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 142.5, 142.3, 140.2, 138.5, 130.9, 129.2, 128.9, 128.8, 128.7, 128.7, 128.6, 128.6, 128.5, 128.3, 128.1, 127.7, 126.9, 126.1, 125.8, 42.0, 36.9, 35.0, 34.3, 34.1, 32.6, 32.5, 31.4, 30.8, 29.8, 29.5, 29.2, 29.1, 28.4, 28.4, 28.2, 26.8, 26.3, 25.8, 25.7.

EI HRMS: calcd. for $C_{22}H_{26}$: 290.2035, found: 290.2038.

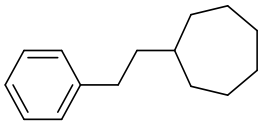


1-(1-hydroxy-2-phenylethyl)cycloheptan-1-ol (1f): Prepared according to the general procedure E for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 8:1) to give the title compound as a colorless oil (312.4 mg, isolated yield: 89%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.34 – 7.31 (m, 2H), 7.26 – 7.18 (m, 3H), 3.69 (ddd, J = 9.7, 4.6, 3.4 Hz, 1H), 2.84 (dd, J = 13.6, 3.4 Hz, 1H), 2.60 (dd, J = 13.6, 9.6 Hz, 1H), 1.88 – 1.79 (m, 2H), 1.77 – 1.72 (m, 2H), 1.69 – 1.60 (m, 2H), 1.58 – 1.55 (m, 2H), 1.52 – 1.47 (m, 2H), 1.44 – 1.35 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 139.5, 129.5, 128.7, 126.4, 77.7, 44.6, 40.5, 30.7, 29.2, 28.5, 28.5, 27.4, 27.1.

EI HRMS: calcd. for $C_{15}H_{22}O_2$: 234.1620, found: 234.1624.

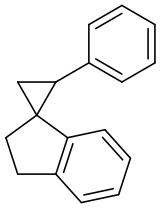


phenethylcycloheptane (1g): Prepared according to the general procedure E for the alkenylation. Following workup, the product was purified by column chromatography (hexanes/ethyl acetate 30:1) to give the title compound as a colorless oil (47.1 mg, isolated yield: 96%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.33 – 7.30 (m, 2H), 7.23 – 7.20 (m, 3H), 2.74 – 2.59 (m, 2H), 1.83 – 1.77 (m, 2H), 1.73 – 1.70 (m, 2H), 1.64 – 1.48 (m, 8H), 1.49 – 1.46 (m, 1H), 1.32 – 1.25 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 143.4, 128.5, 128.4, 125.6, 40.3, 39.0, 34.7, 34.0, 28.7, 26.6.

EI HRMS: calcd. for $C_{15}H_{22}$: 202.1722, found: 202.1728.

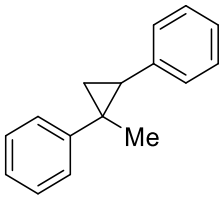


2-phenyl-2',3'-dihydrospiro[cyclopropane-1,1'-indene] (13d): **13d** was purified by column chromatography (hexanes) to give the title compound as a colorless oil (9.5 mg, isolated yield: 43%, d.r. = 1:1.3).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.32 (t, J = 7.6 Hz, 1H), 7.26 – 7.14 (m, 5H), 7.14 – 7.08 (m, 1H), 7.01 (t, J = 7.4 Hz, 0.5H), 6.87 (d, J = 8.4 Hz, 0.6H), 6.78 (t, J = 7.5 Hz, 0.5H), 5.98 (d, J = 7.6 Hz, 0.4H), 3.19 – 3.12 (m, 0.4H), 3.08 – 2.86 (m, 1.6H), 2.59 – 2.56 (m, 0.4H), 2.48 – 2.34 (m, 1H), 2.00 – 1.94 (m, 0.4H), 2.00 – 1.94 (m, 0.6H), 1.87 – 1.81 (m, 0.6H), 1.55 – 1.42 (m, 2H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 148.3, 145.0, 144.3, 143.8, 139.4, 138.4, 130.2, 128.3, 128.2, 128.0, 126.7, 126.1, 126.1, 125.6, 125.4, 124.4, 124.0, 121.4, 118.6, 37.4, 35.4, 34.2, 33.3, 33.2, 31.0, 30.7, 29.2, 20.7, 18.9.

EI HRMS: calcd. for $C_{17}H_{16}$: 220.1252, found: 220.1257.

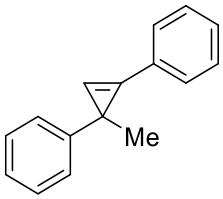


(1-methylcyclopropane-1,2-diyl)dibenzene (25d): **25d** was purified by column chromatography (hexanes) to give the title compound as a colorless oil (6.3 mg, isolated yield: 30%, d.r. = 1:1.2).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.40 – 7.27 (m, 4H), 7.25 – 7.20 (m, 1H), 7.17 – 7.09 (m, 1H), 7.10 – 6.94 (m, 3H), 6.78 – 6.70 (m, 1H), 2.41 (dd, J = 8.8, 6.4 Hz, 0.5H), 2.22 (dd, J = 8.6, 5.9 Hz, 0.5H), 1.54 (s, 1.4H), 1.50 (t, J = 5.6 Hz, 0.5H), 1.46 (t, J = 5.1 Hz, 0.5H), 1.28 – 1.23 (m, 1H), 1.12 (s, 1.6H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 148.0, 142.4, 140.0, 139.2, 130.0, 129.3, 128.5, 128.2, 128.0, 127.6, 127.6, 127.0, 126.1, 126.0, 125.9, 125.2, 31.5, 31.3, 29.8, 27.1, 21.1, 19.8, 18.8.

EI HRMS: calcd. for $C_{16}H_{16}$: 208.1252, found: 208.1255.



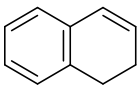
Methylcycloprop-2-ene-1,2-diyl)dibenzene (25e)

25e was purified by column chromatography (hexanes) to give the title compound as a colorless oil (2.3 mg, isolated yield: 11%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.52 – 7.50 (m, 2H), 7.39 – 7.36 (m, 2H), 7.34 – 7.27 (m, 6H), 7.16 – 7.12 (m, 1H), 1.78 (s, 3H).

^{13}C NMR (126 MHz, $CDCl_3$) δ 148.9, 129.5, 129.1, 128.8, 128.0, 127.9, 126.2, 125.1, 124.2, 108.7, 25.2, 23.7.

EI HRMS: calcd. for $C_{16}H_{14}$: 206.1096, found: 206.1088.



1,2-Dihydronaphthalene (14d)

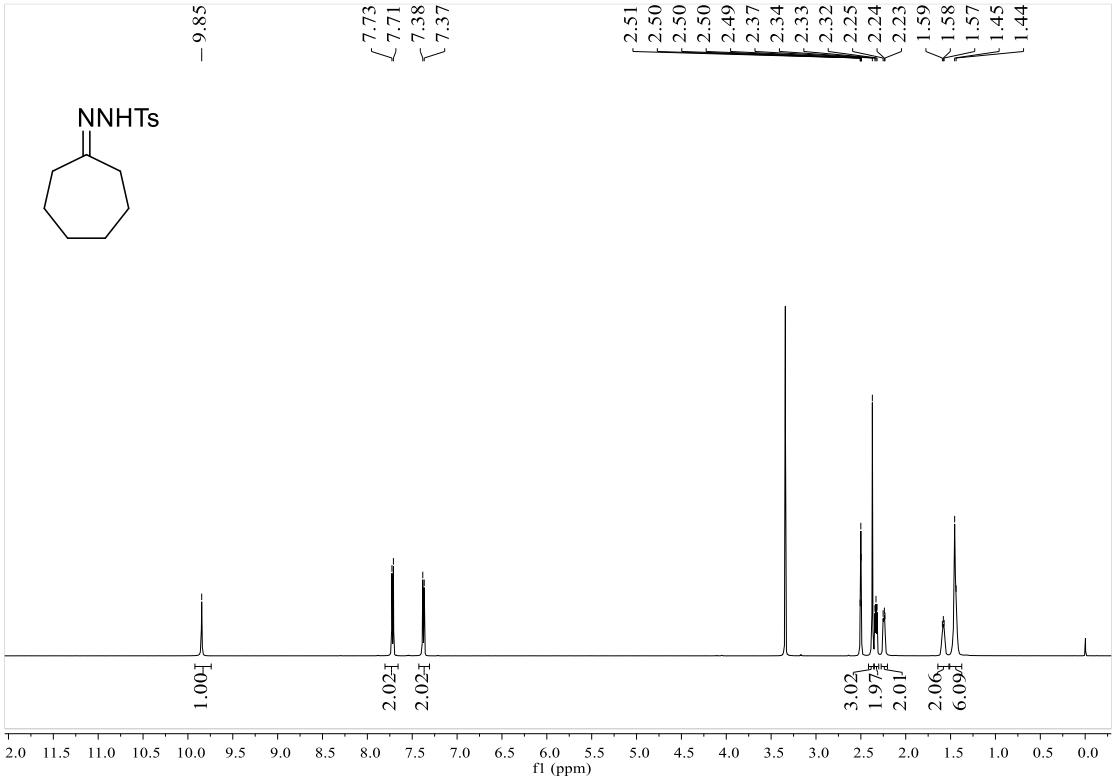
14d was purified by column chromatography (hexanes) to give the title compound as a colorless oil (2.3 mg, isolated yield: 18%). **26b** was known in the published literature.[18]

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.21 – 7.08 (m, 3H), 7.07 – 7.01 (m, 1H), 6.48 (dt, J = 9.6, 1.8 Hz, 1H), 6.07 – 6.02 (m, 1H), 2.82 (t, J = 8.2 Hz, 2H), 2.36 – 2.31 (m, 2H).

9 NMR spectra of products and synthesized substrates

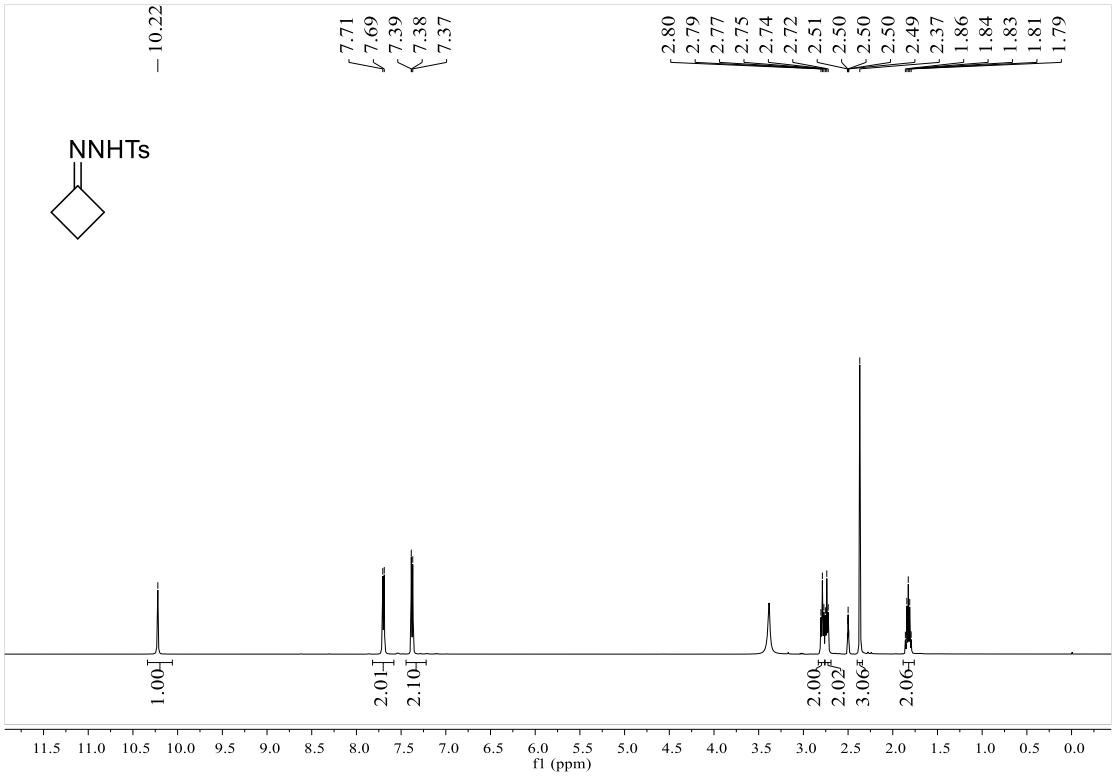
9.1 NMR spectra of synthesized *N*-tosylhydrazones

_N'_-cycloheptylidene-4-methylbenzenesulfonohydrazide (1a):



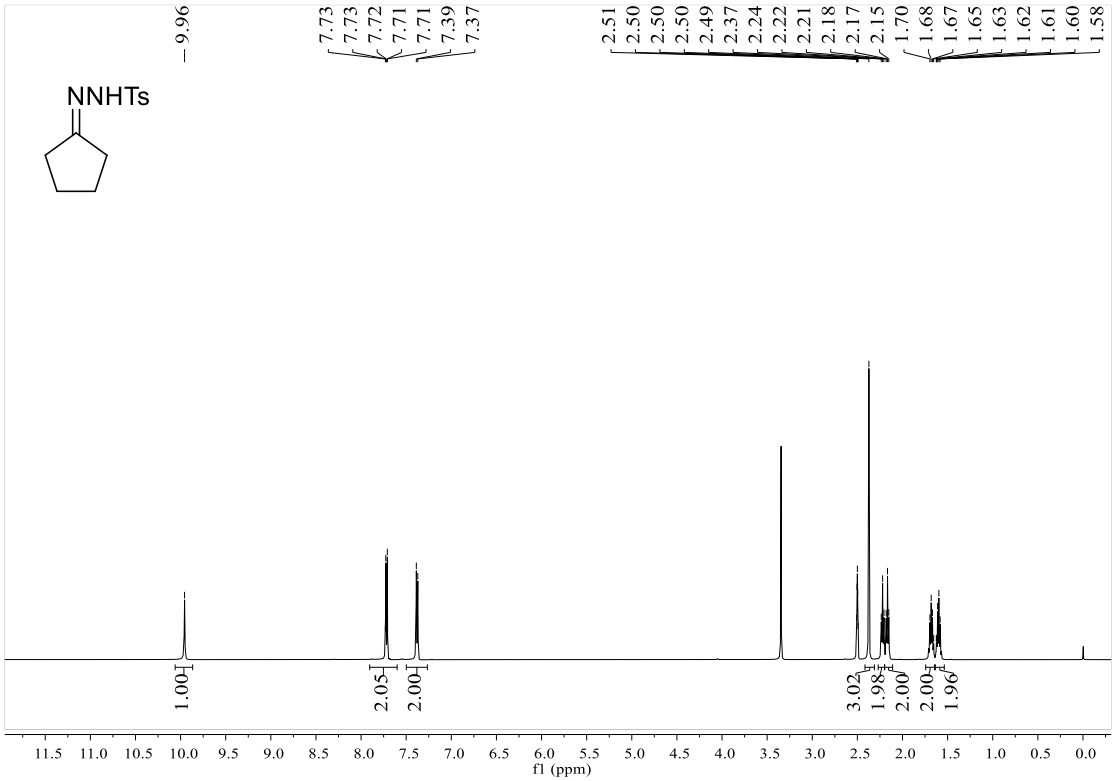
^{1}H NMR spectrum in DMSO-d_6.

***N*'-cyclobutylidene-4-methylbenzenesulfonohydrazide (2a)**:



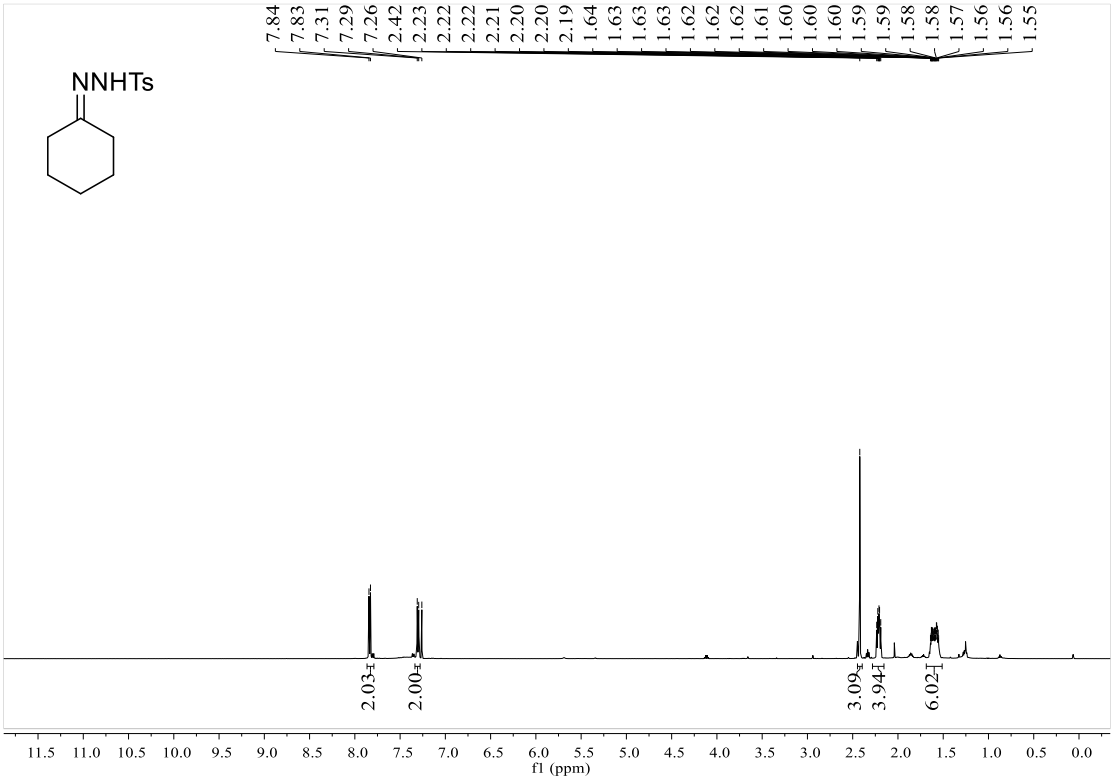
^{1}H NMR spectrum in DMSO-d_6.

***N*'-cyclopentylidene-4-methylbenzenesulfonohydrazide (3a)**:



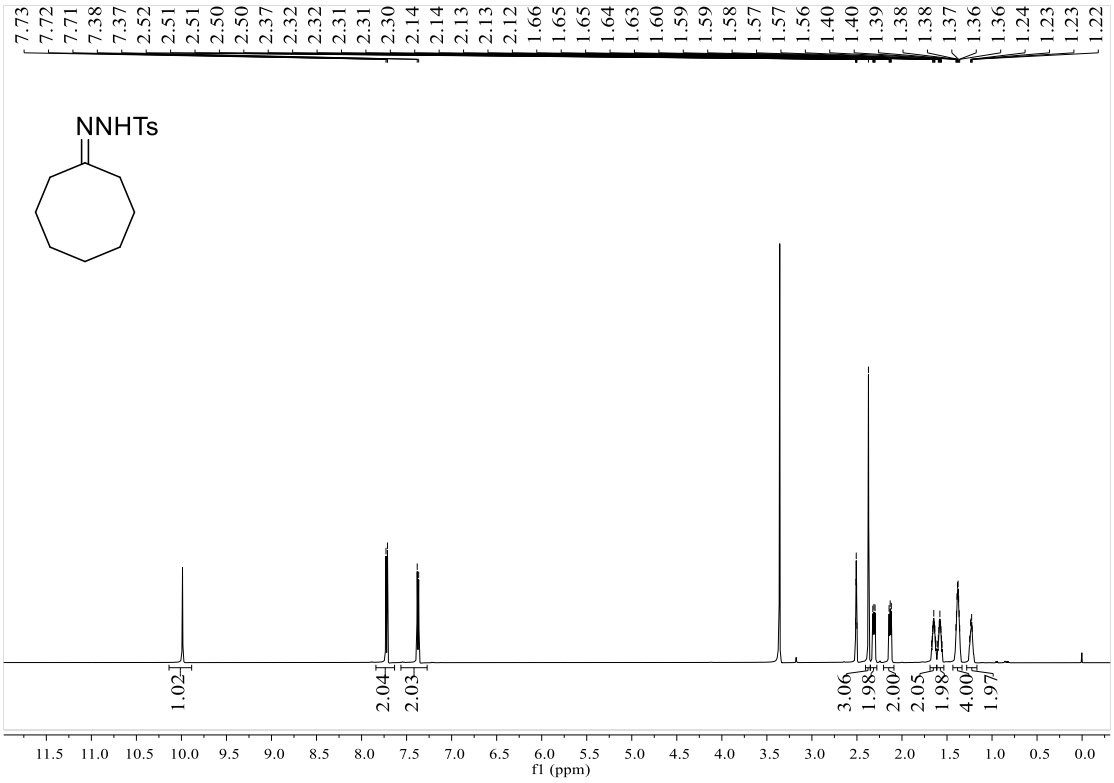
^{1}H NMR spectrum in DMSO-d_6.

***N*'-cyclohexylidene-4-methylbenzenesulfonohydrazide (4a)**:



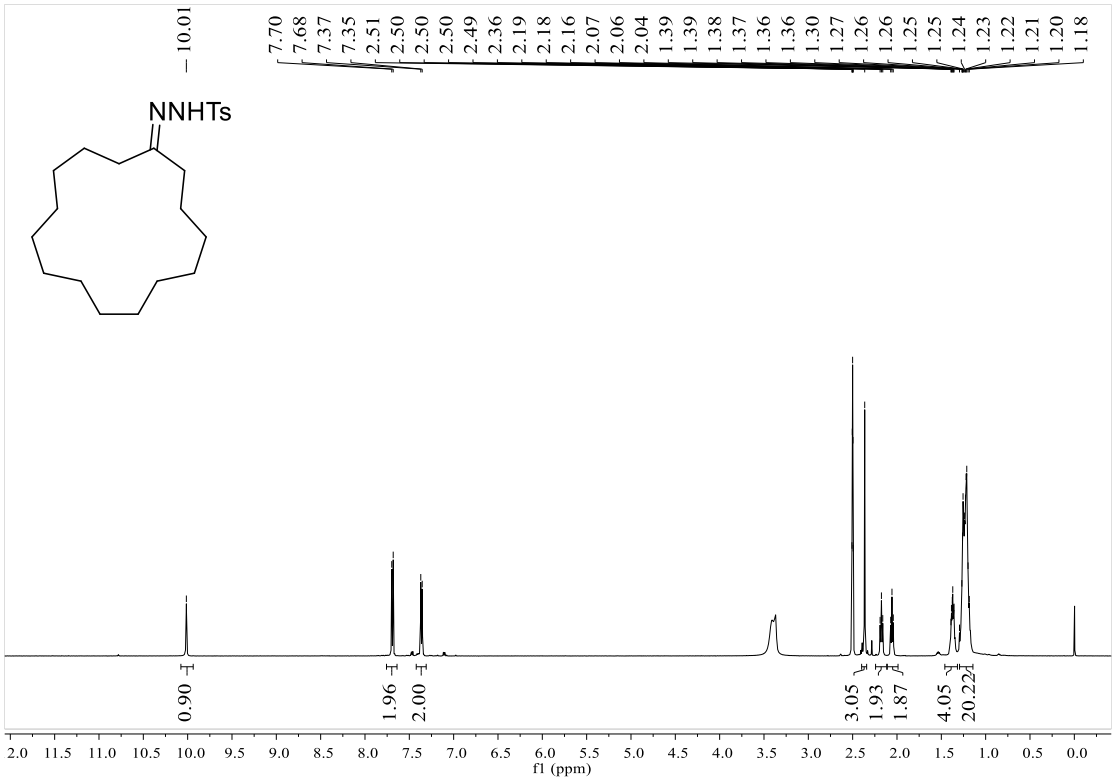
^{1}H NMR spectrum in $CDCl_3$.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (5a)**:



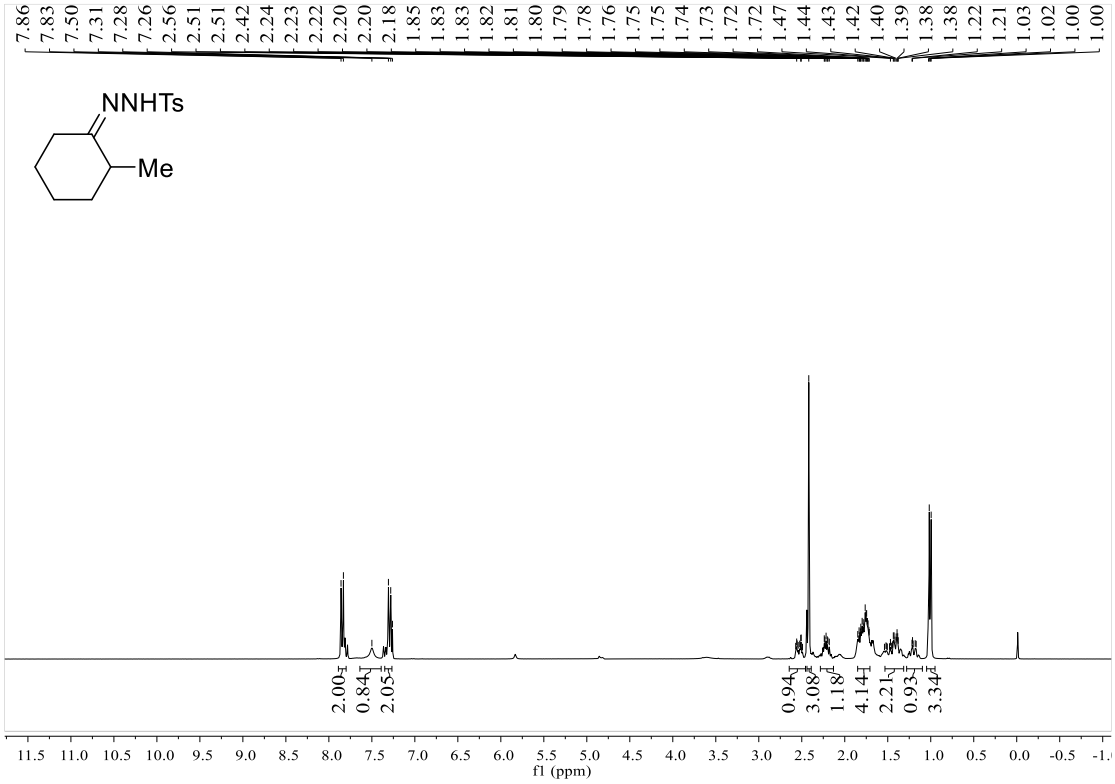
^{1}H NMR spectrum in DMSO-d_6.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (6a)**:



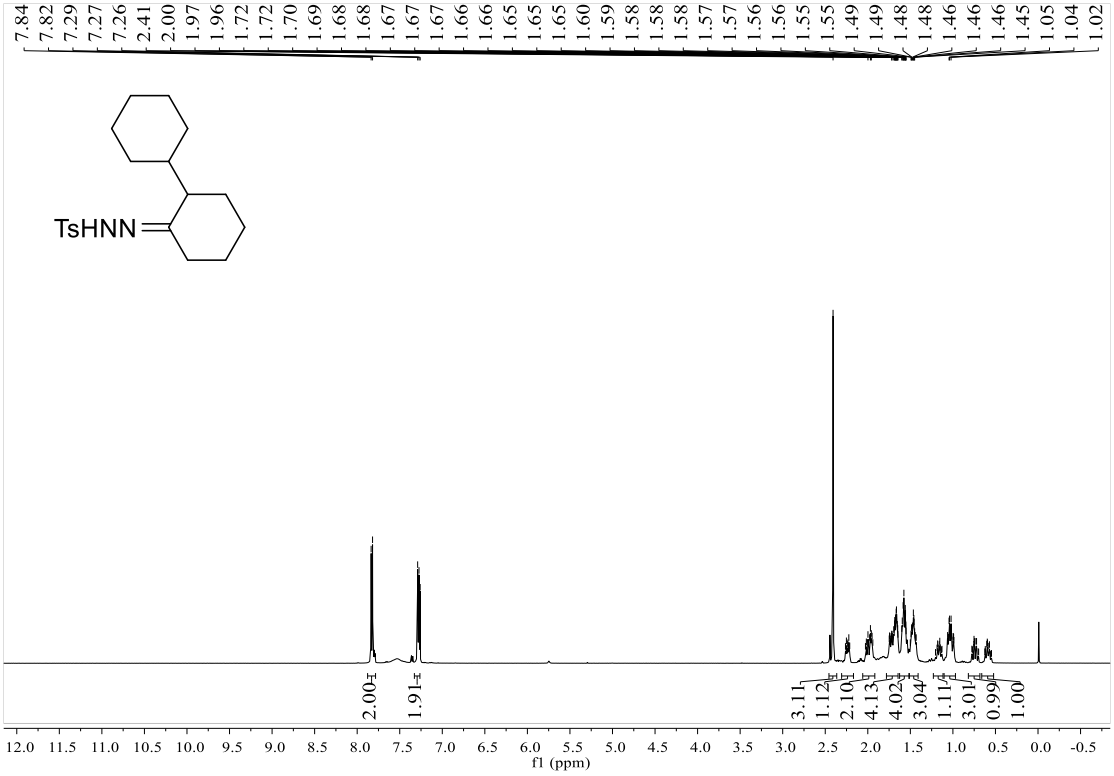
^{1}H NMR spectrum in DMSO-d_6.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (7a)**:

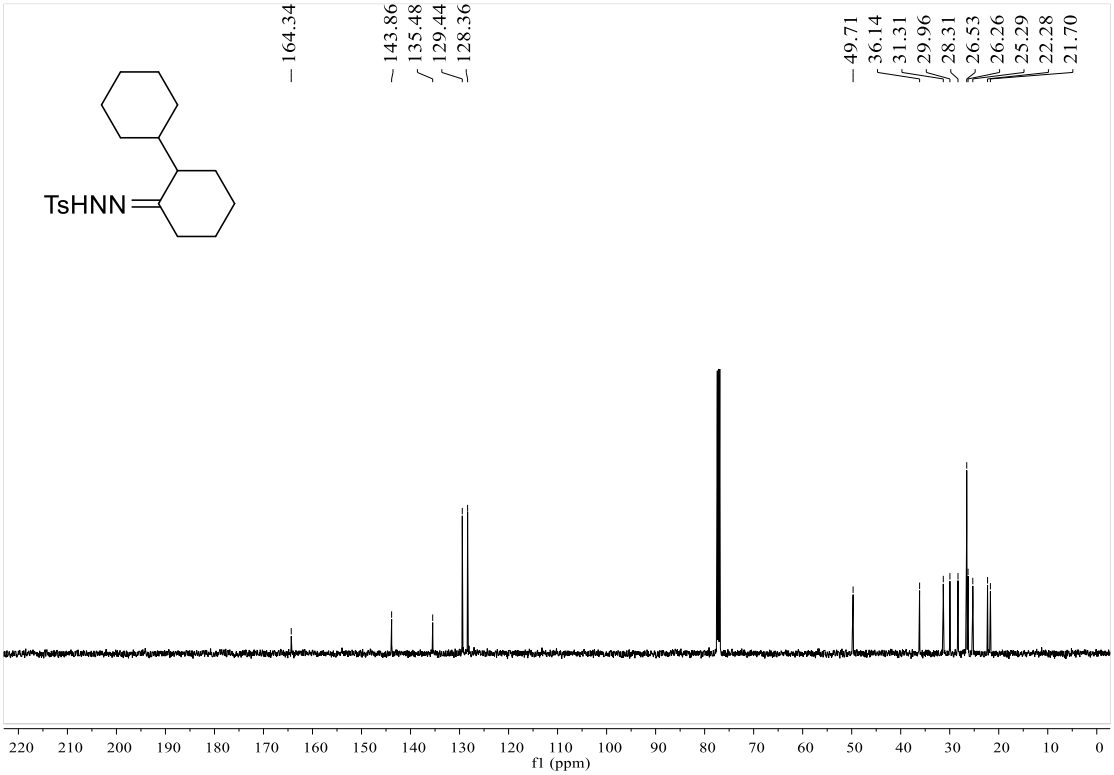


^{1}H NMR spectrum in DMSO-d_6.

***N*'-([1,1'-bi(cyclohexan)]-2-ylidene)-4-methylbenzenesulfonohydrazide (8a)**:

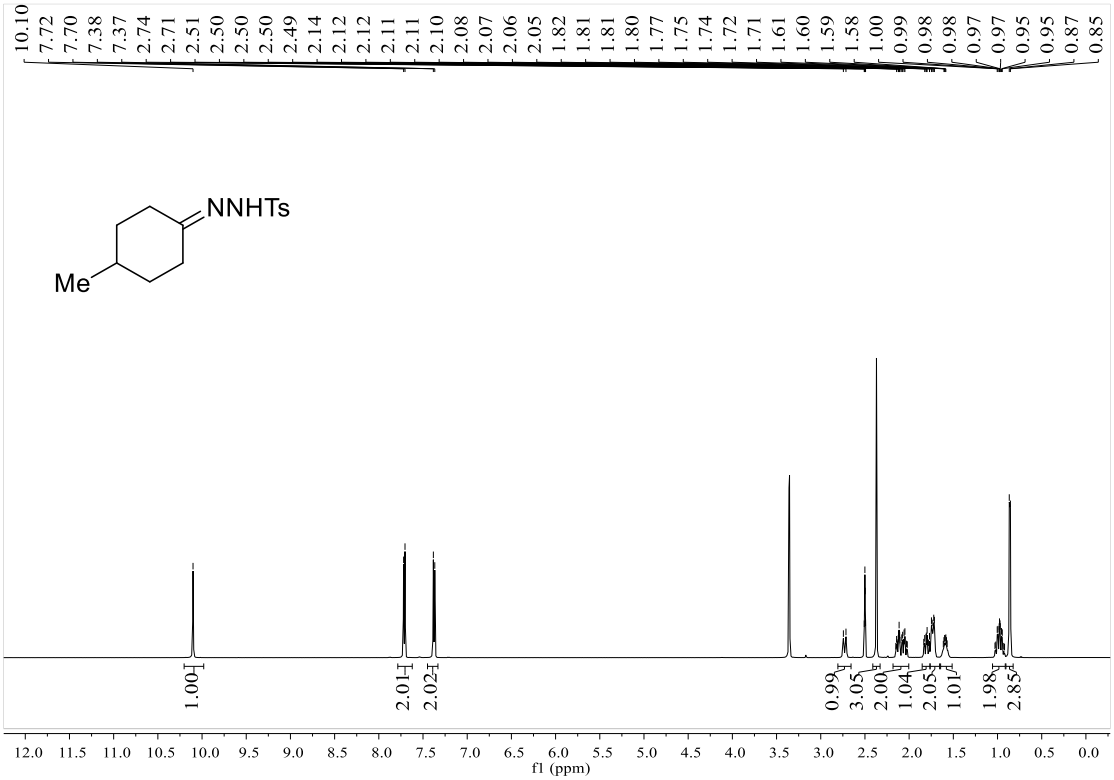


^{1}H NMR spectrum in $CDCl_3$.



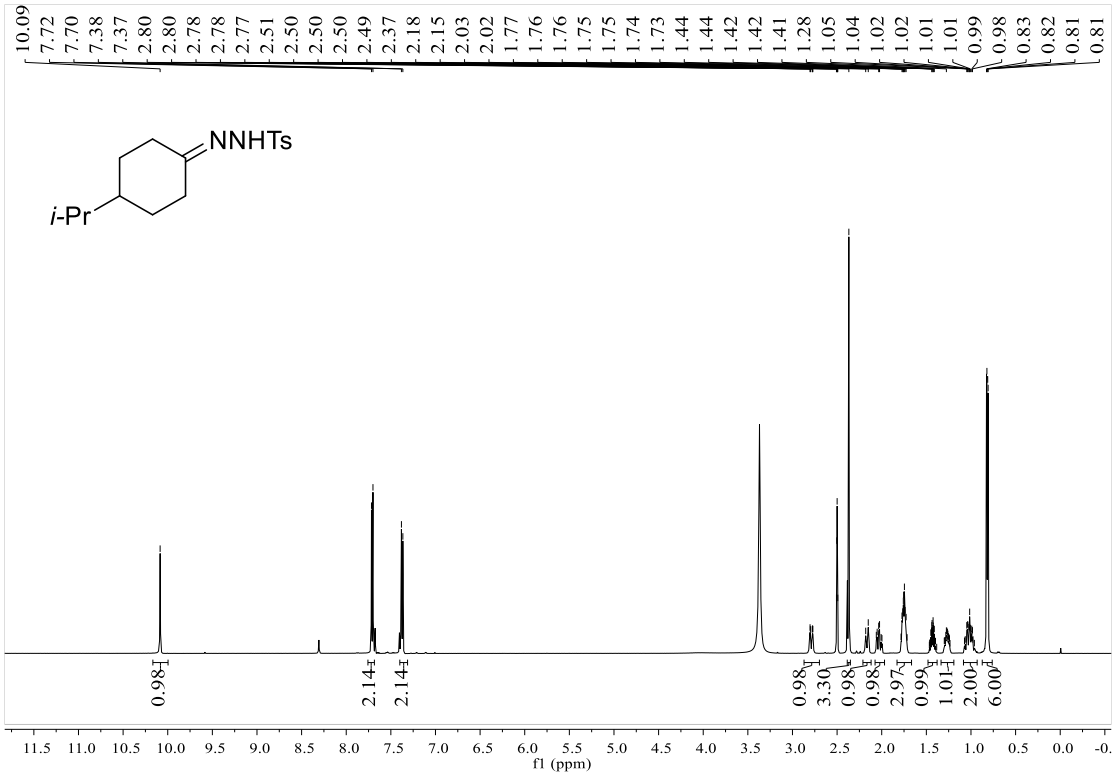
^{13}C NMR spectrum in $CDCl_3$.

4-methyl-*N*'-(4-methylcyclohexylidene)benzenesulfonohydrazide (9a):

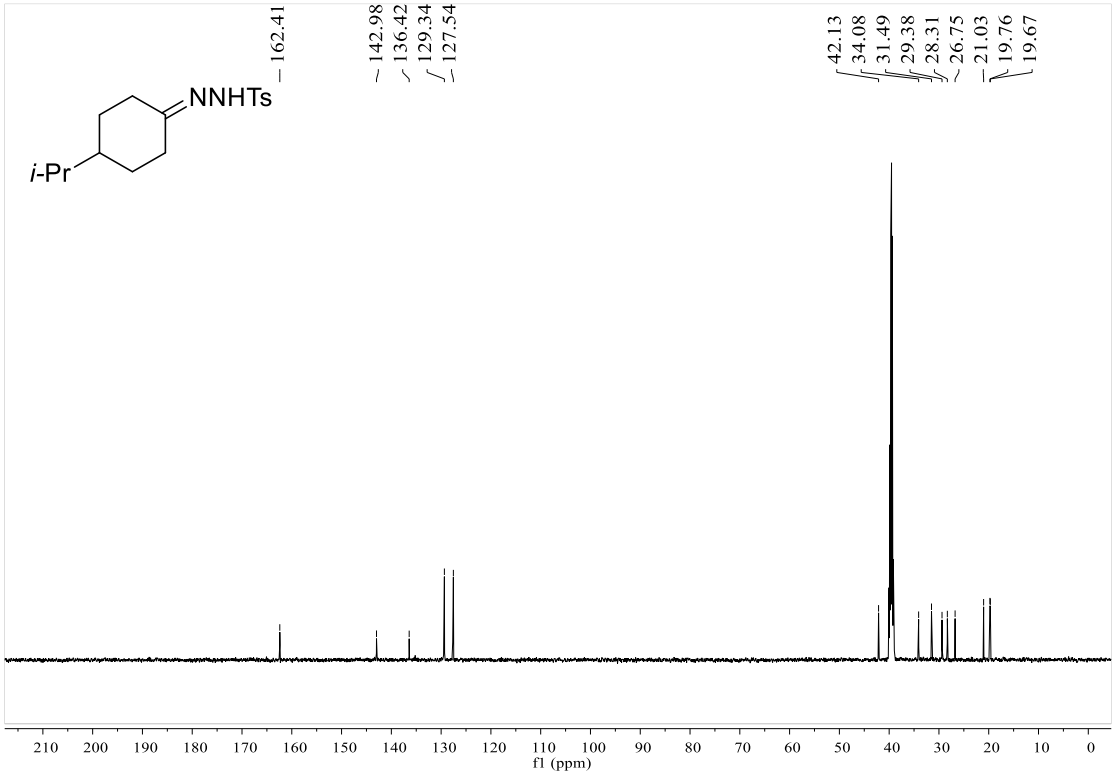


^{1}H NMR spectrum in DMSO-d_{6}.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (10a)**:

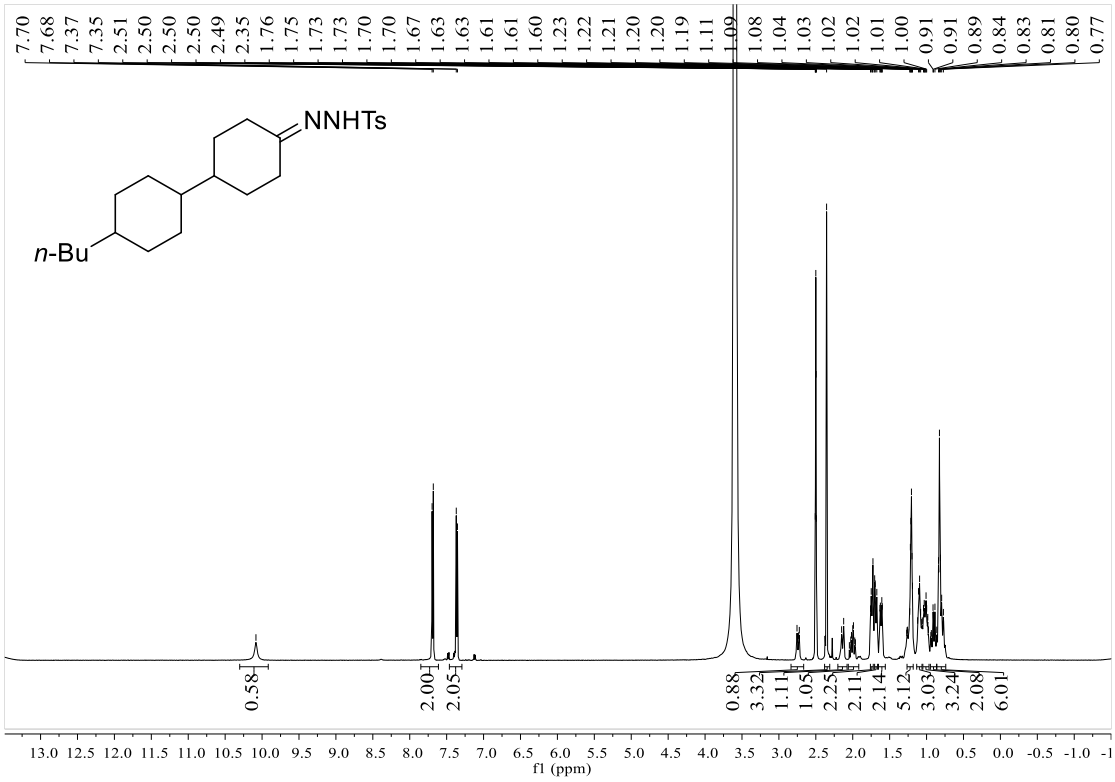


^{1}H NMR spectrum in DMSO-d_6.

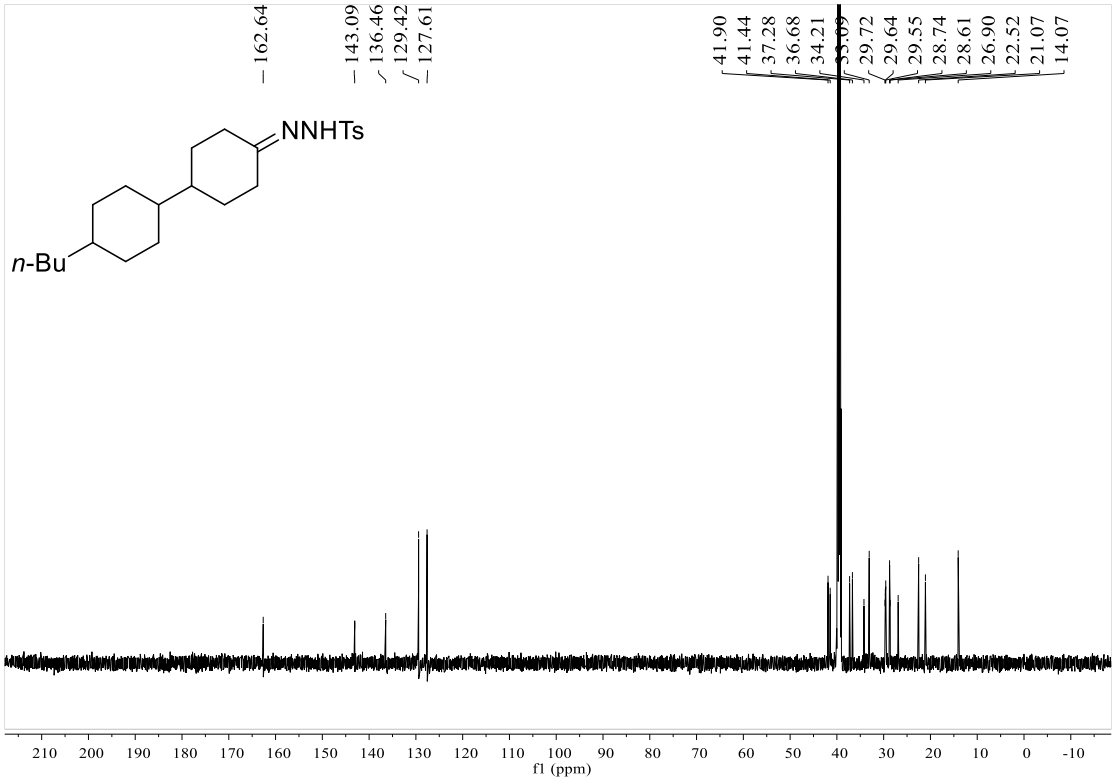


^{13}C NMR spectrum in DMSO-d_6.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (11a)**:

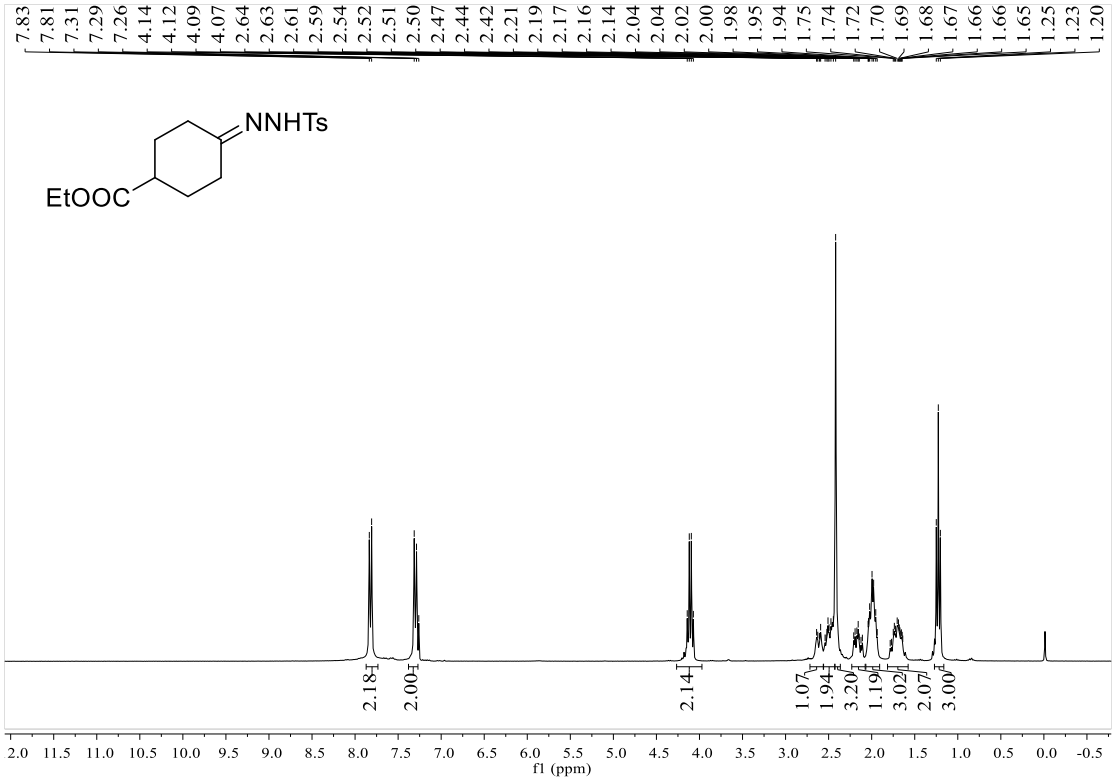


^{1}H NMR spectrum in DMSO-d_6.

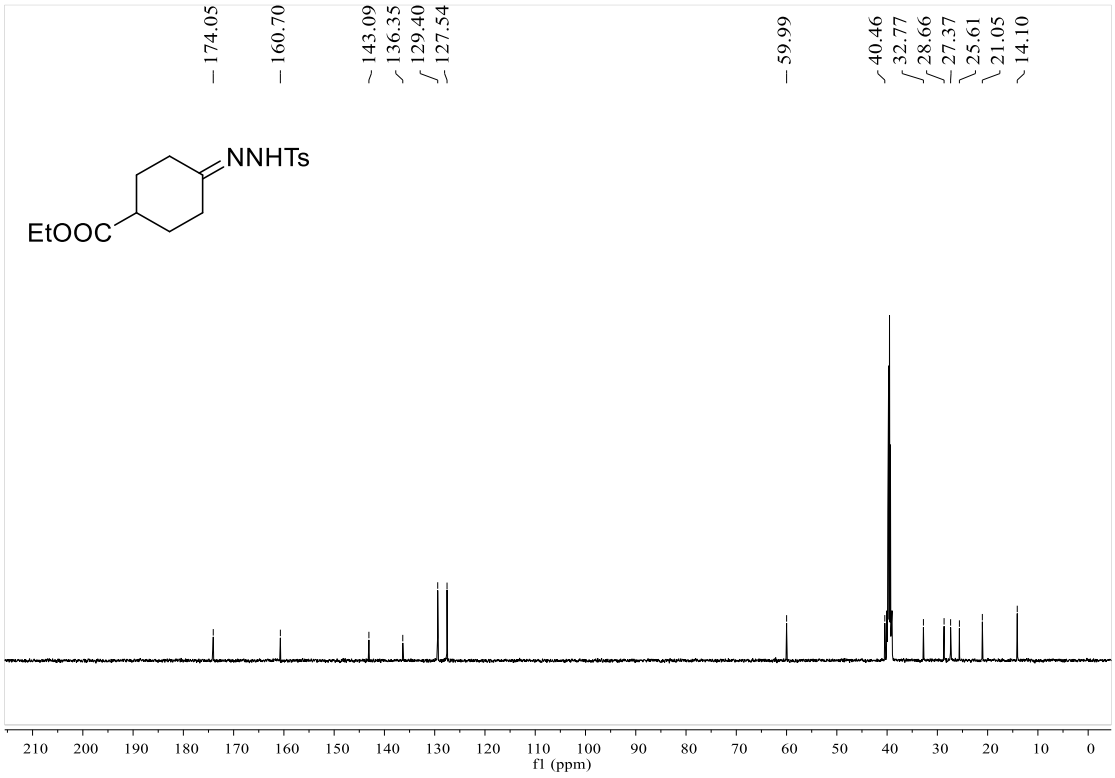


^{13}C NMR spectrum in DMSO-d_6.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (12a)**:

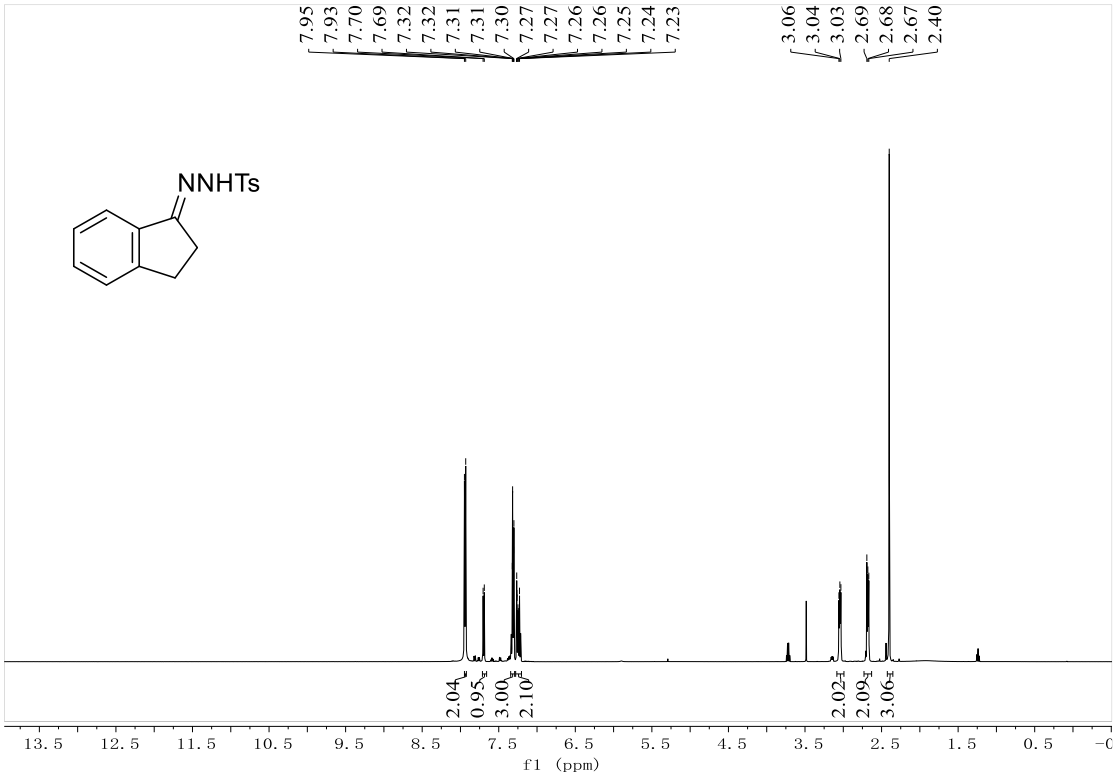


^{1}H NMR spectrum in $CDCl_3$.

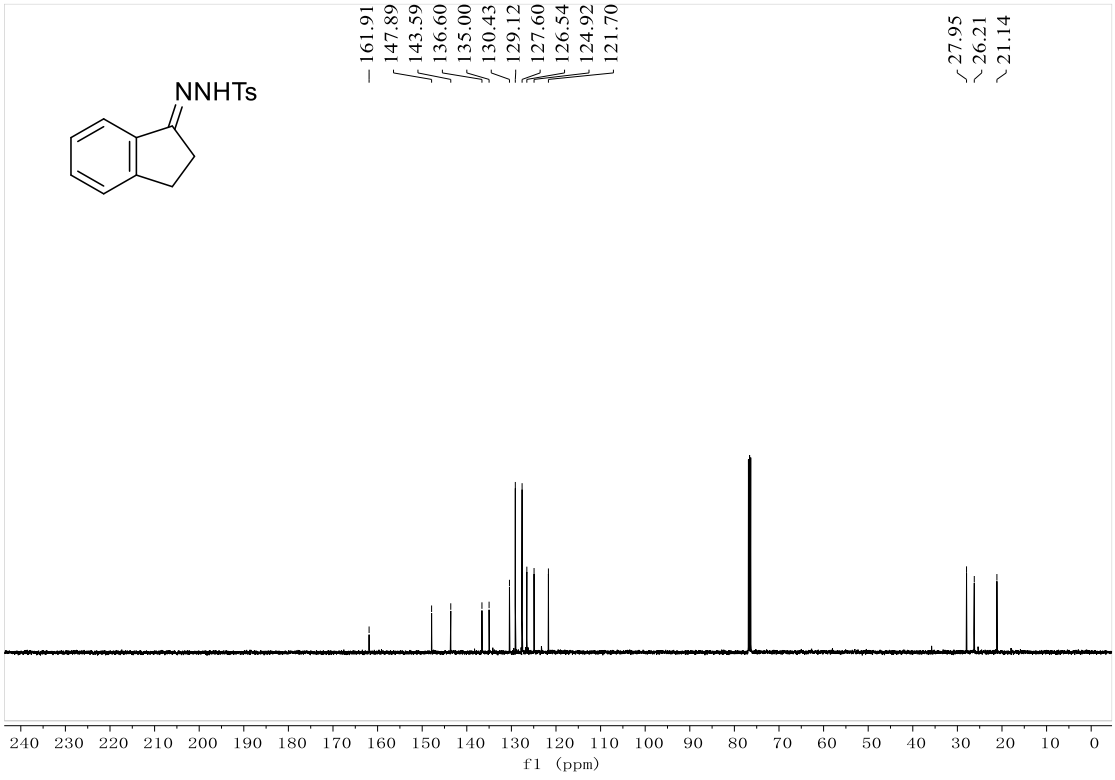


^{13}C NMR spectrum in DMSO-d_6.

***N*'-(2,3-dihydro-1*H*-inden-1-ylidene)-4-methylbenzenesulfonohydrazide (13a)**:

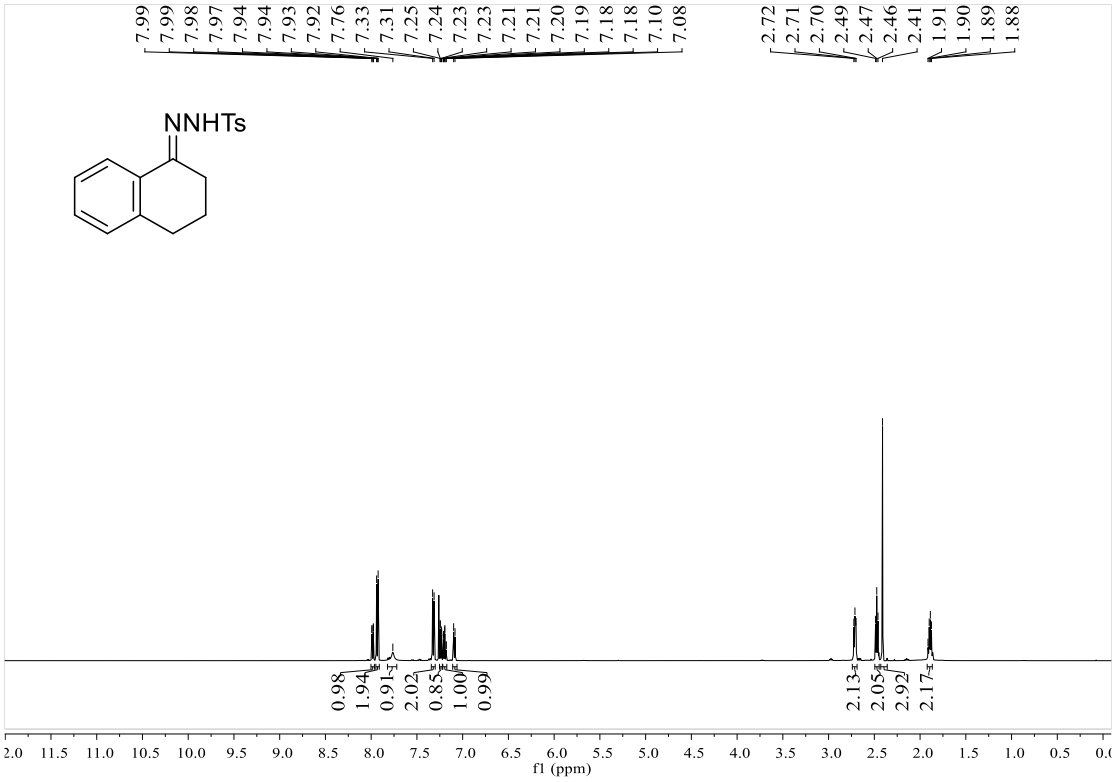


^{1}H NMR spectrum in $CDCl_3$.



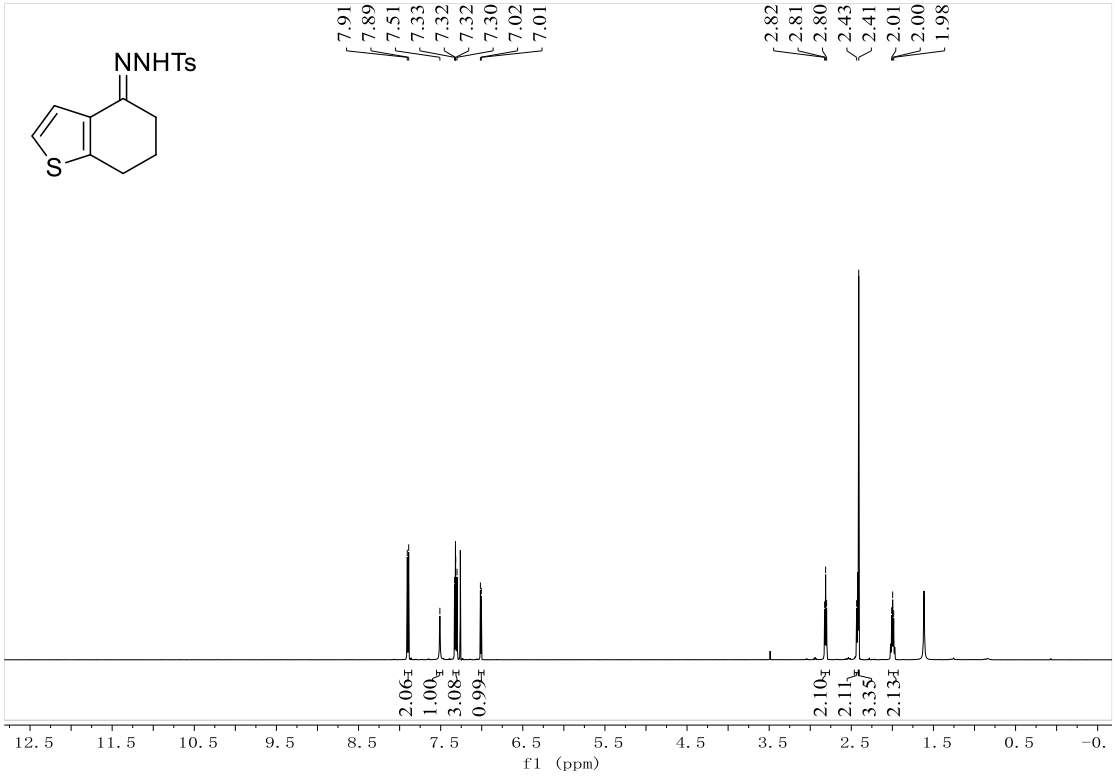
^{13}C NMR spectrum in $CDCl_3$.

***N'*-(3,4-Dihydronaphthalen-1(2*H*)-ylidene)-4-methylbenzenesulfonohydrazide (14a)**:



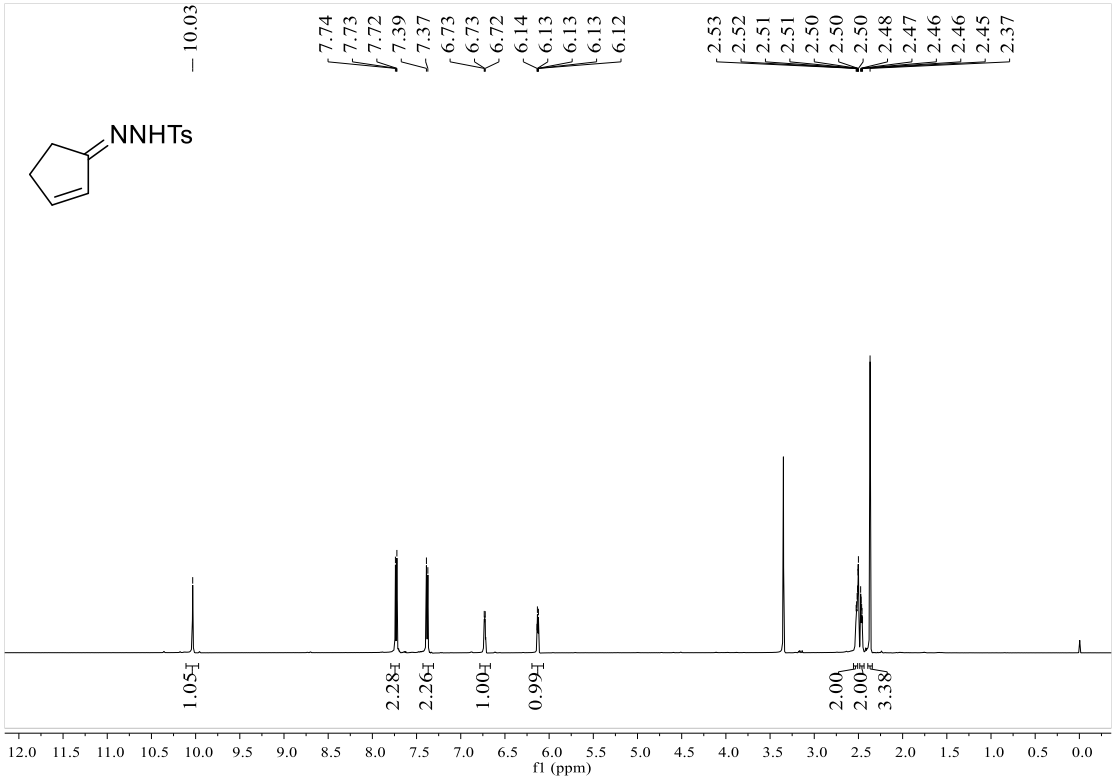
^{1}H NMR spectrum in DMSO-d_6.

***N'*-(6,7-dihydrobenzo[b]thiophen-4(5*H*)-ylidene)-4-methylbenzenesulfonohydrazide (15a)**:



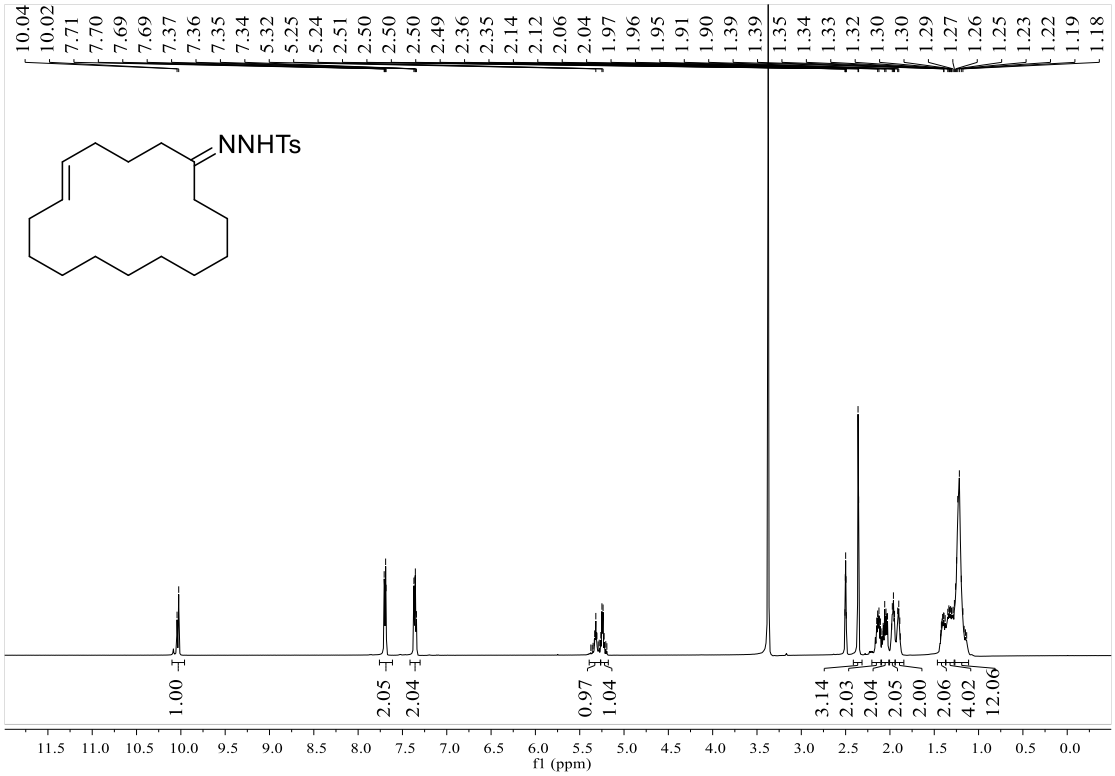
^{1}H NMR spectrum in DMSO-d_6.

***N*'-(cyclopent-2-en-1-ylidene)-4-methylbenzenesulfonohydrazide (16a)**:

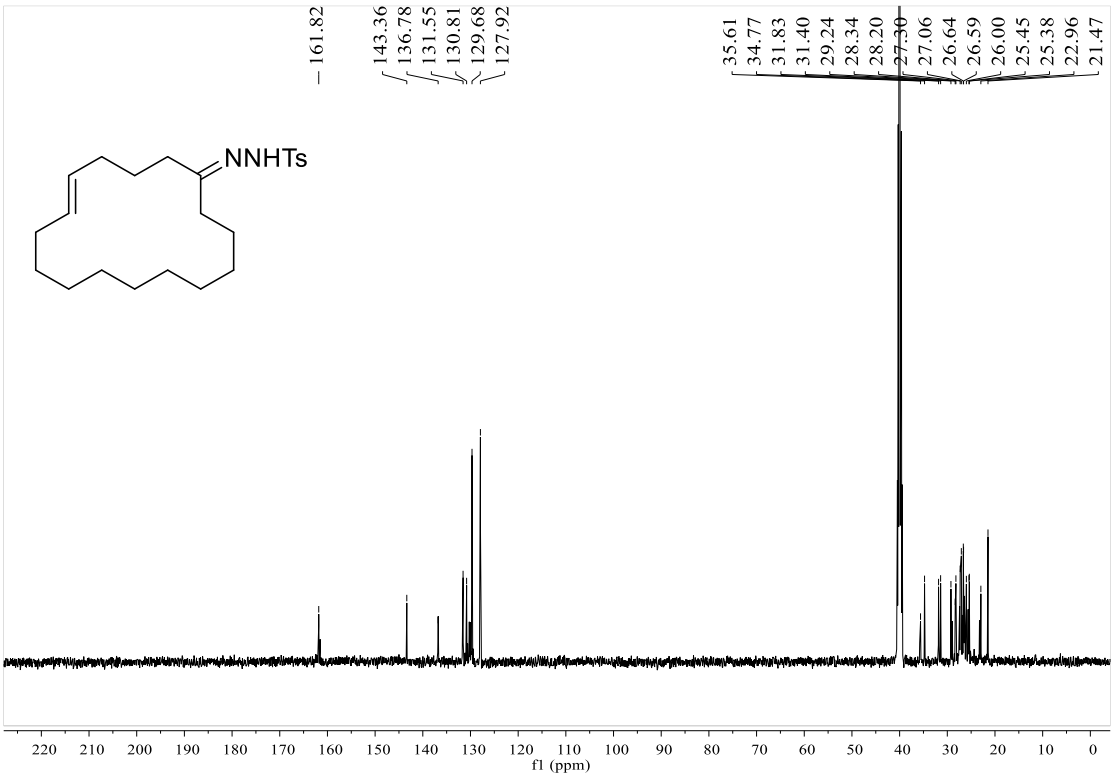


^{1}H NMR spectrum in DMSO-d_6.

***N*'-(cyclohexadec-3-en-1-ylidene)-4-methylbenzenesulfonohydrazide (17a)**:

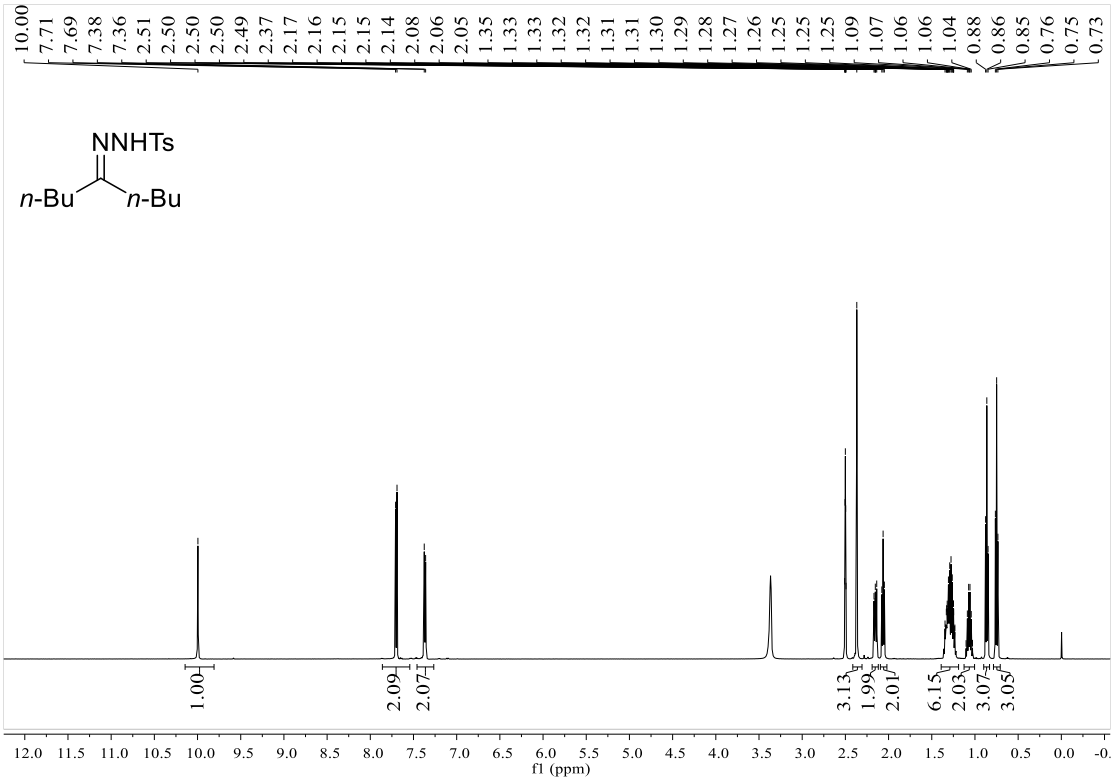


^{1}H NMR spectrum in DMSO-d_6.



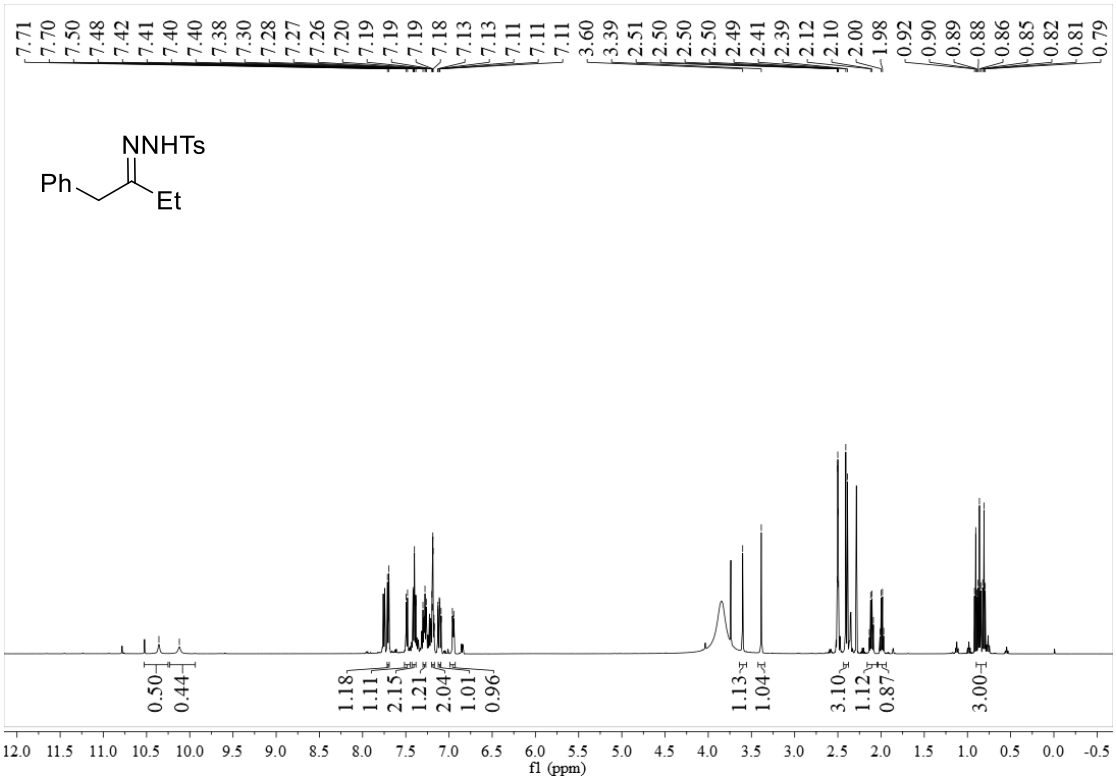
^{13}C NMR spectrum in DMSO-d_6.

4-methyl-*N*'-(nonan-5-ylidene)benzenesulfonohydrazide (18a):



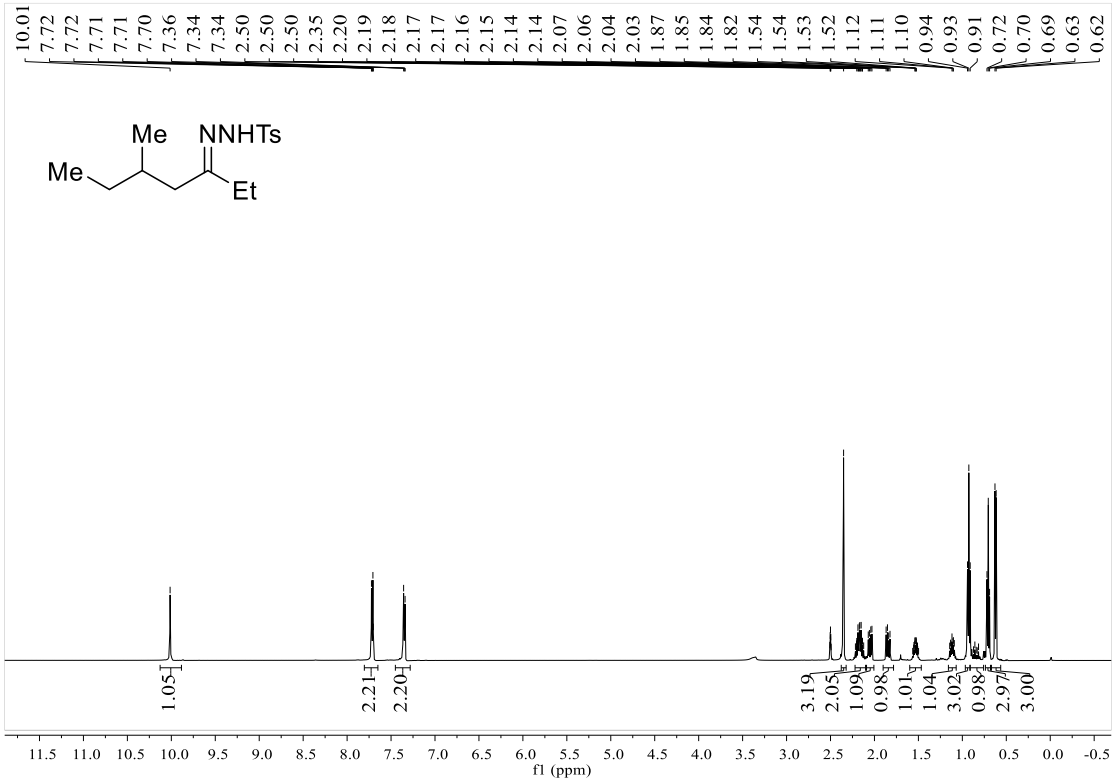
^{1}H NMR spectrum in DMSO-d_6.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (19a)**:

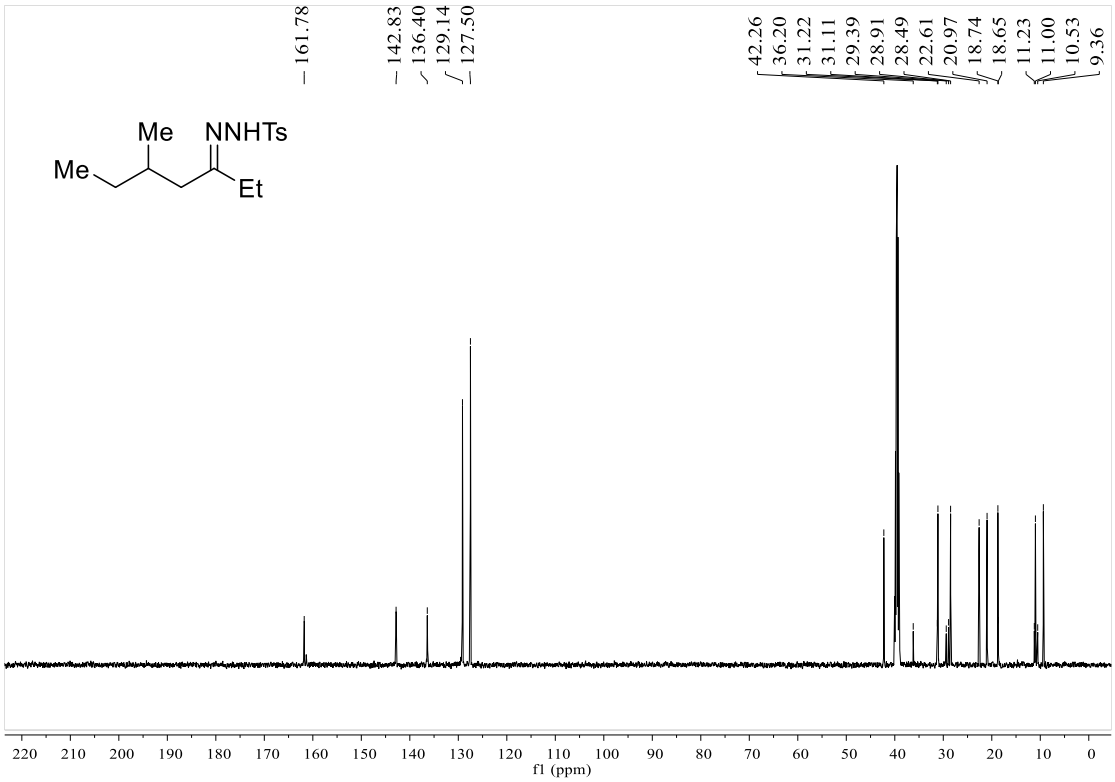


^{1}H NMR spectrum in DMSO-d_6.

4-methyl-*N*'-(5-methylheptan-3-ylidene)benzenesulfonohydrazide (20a):

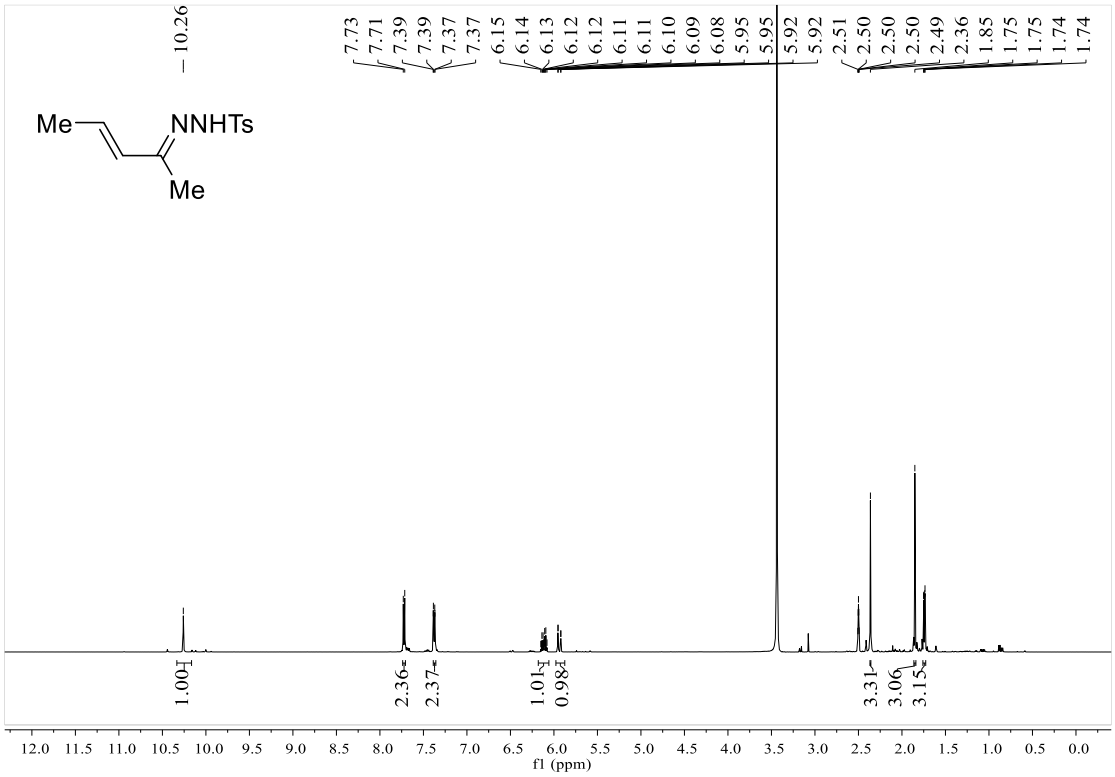


^{1}H NMR spectrum in DMSO-d_6.

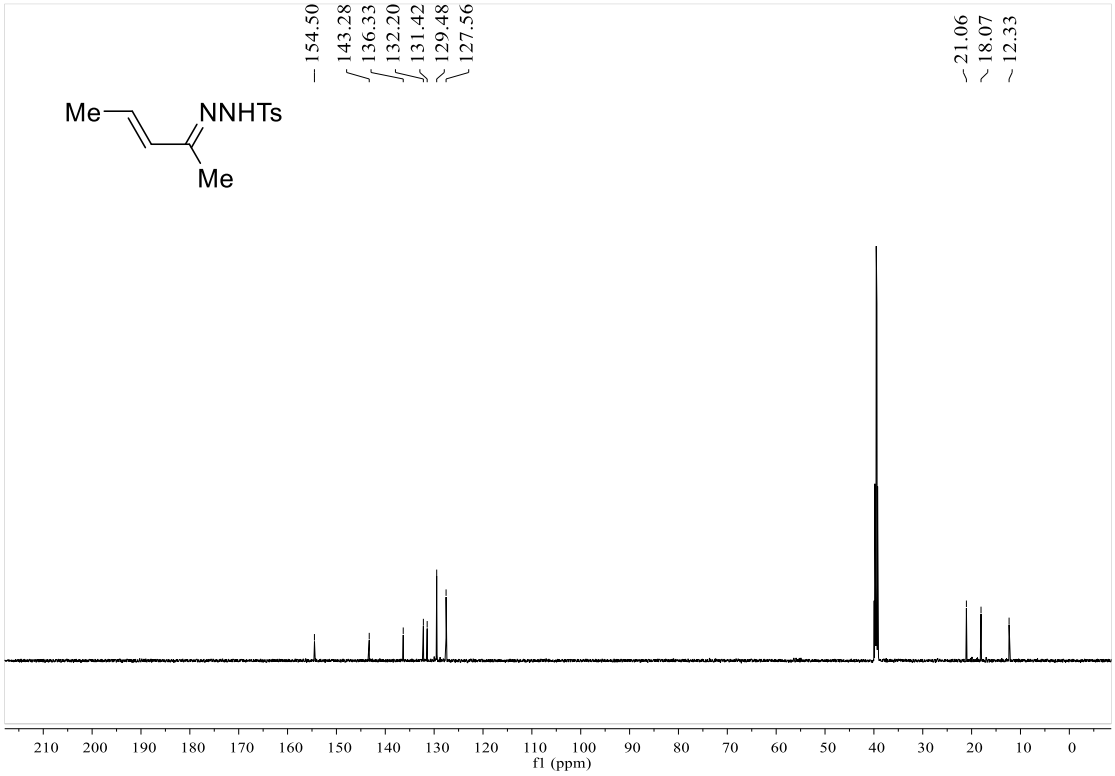


^{13}C NMR spectrum in DMSO-d_6.

***N'*-cyclooctylidene-4-methylbenzenesulfonohydrazide (21a)**:

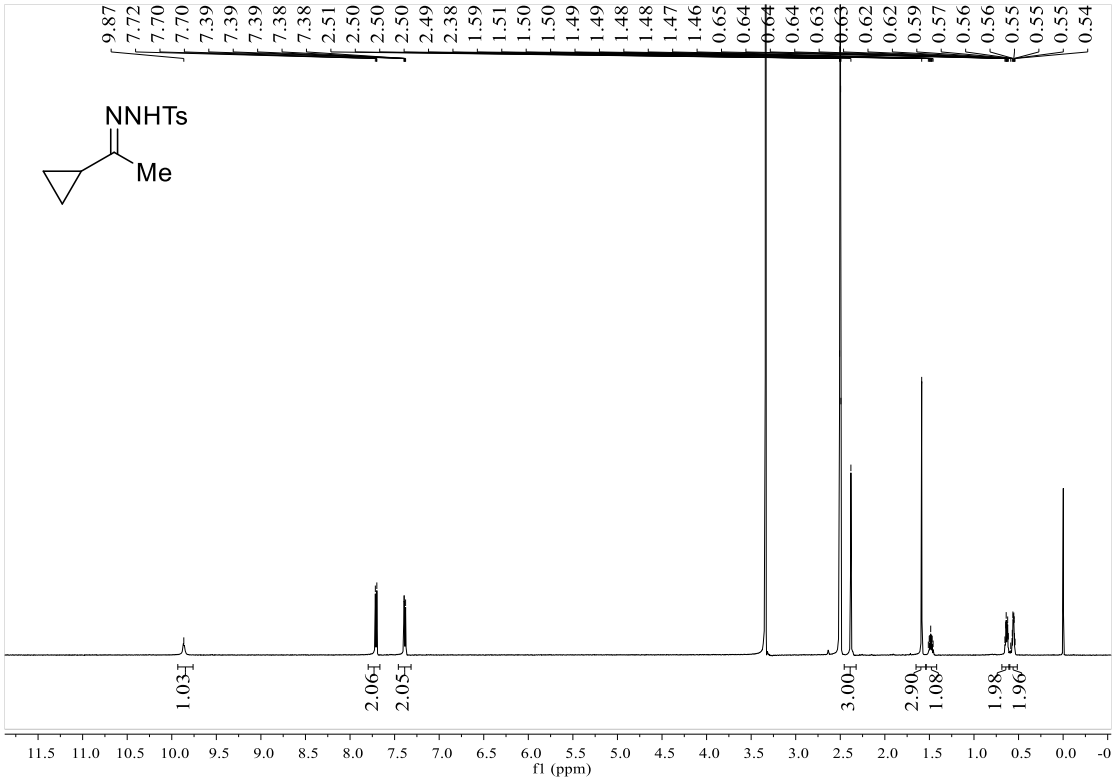


^{1}H NMR spectrum in DMSO-d_6.



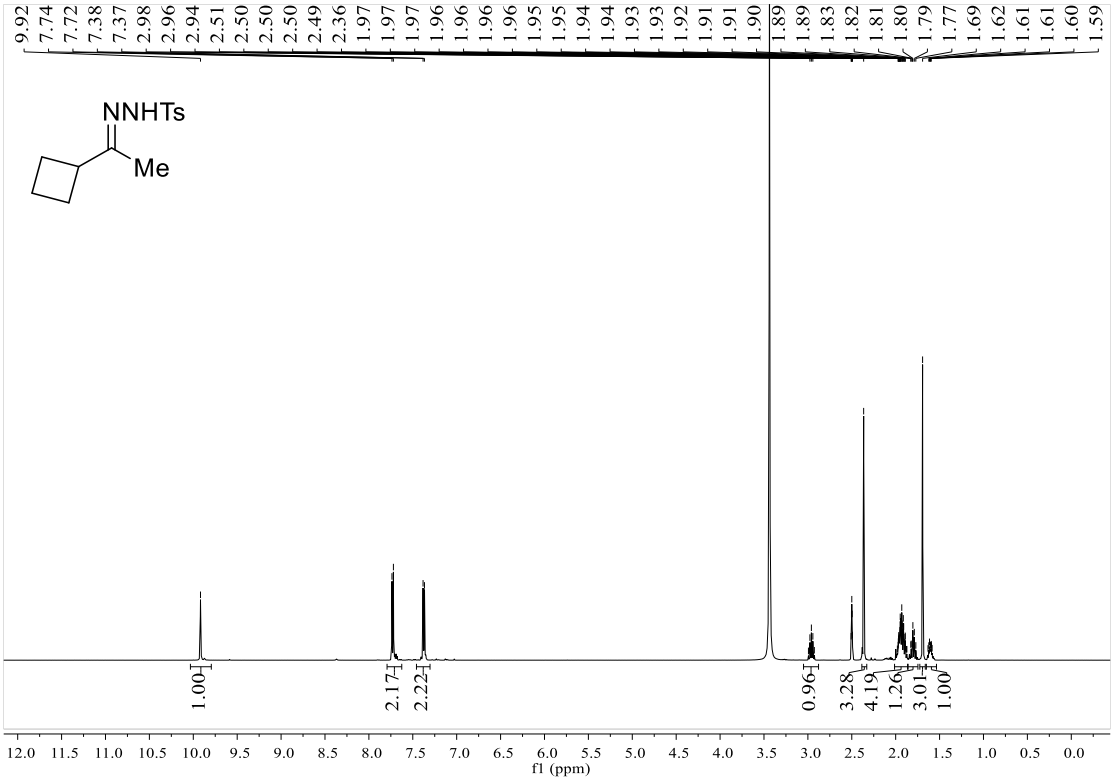
^{13}C NMR spectrum in DMSO-d_6.

***N′*-(1-cyclopropylethylidene)-4-methylbenzenesulfonohydrazide (22a)**:

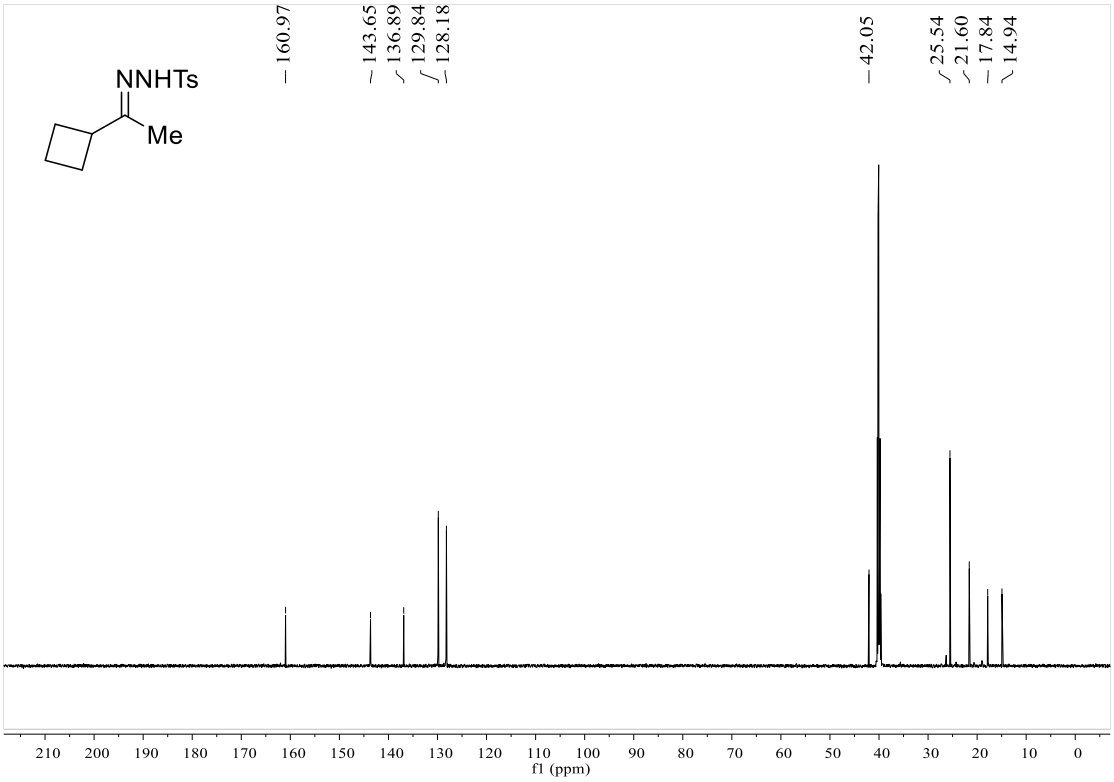


^{1}H NMR spectrum in DMSO-d_6.

***N′*-(1-cyclobutylethylidene)-4-methylbenzenesulfonohydrazide (23a)**:

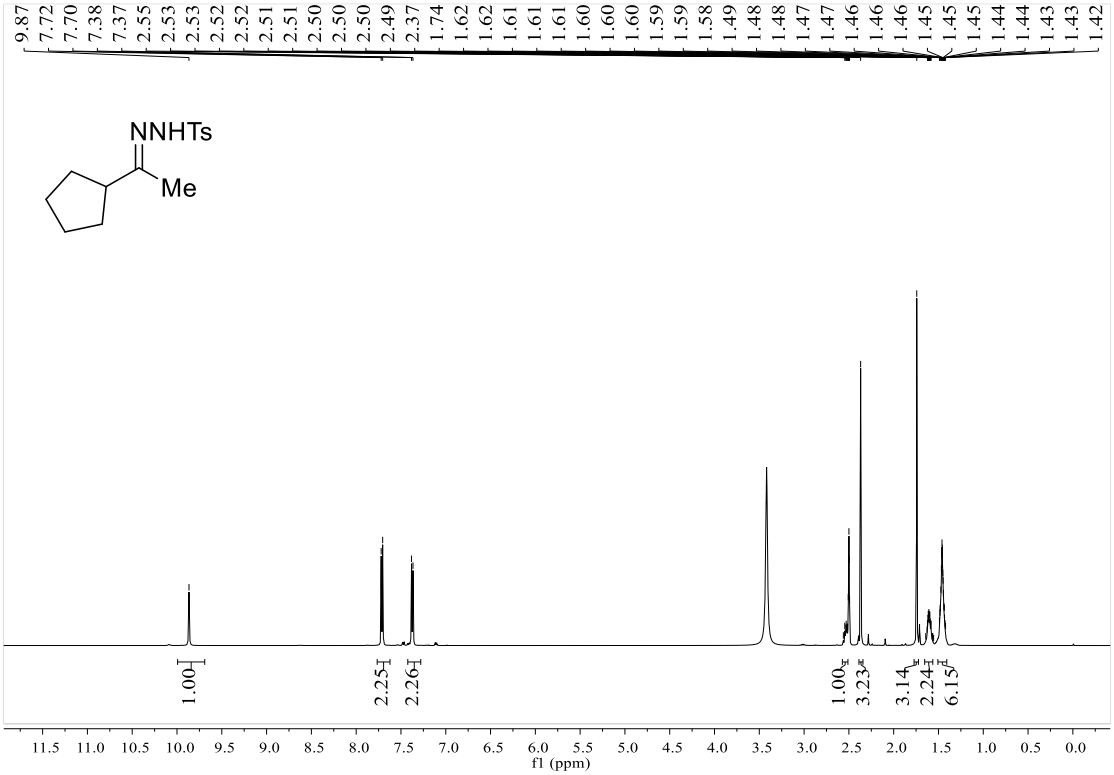


^{1}H NMR spectrum in DMSO-d_6.

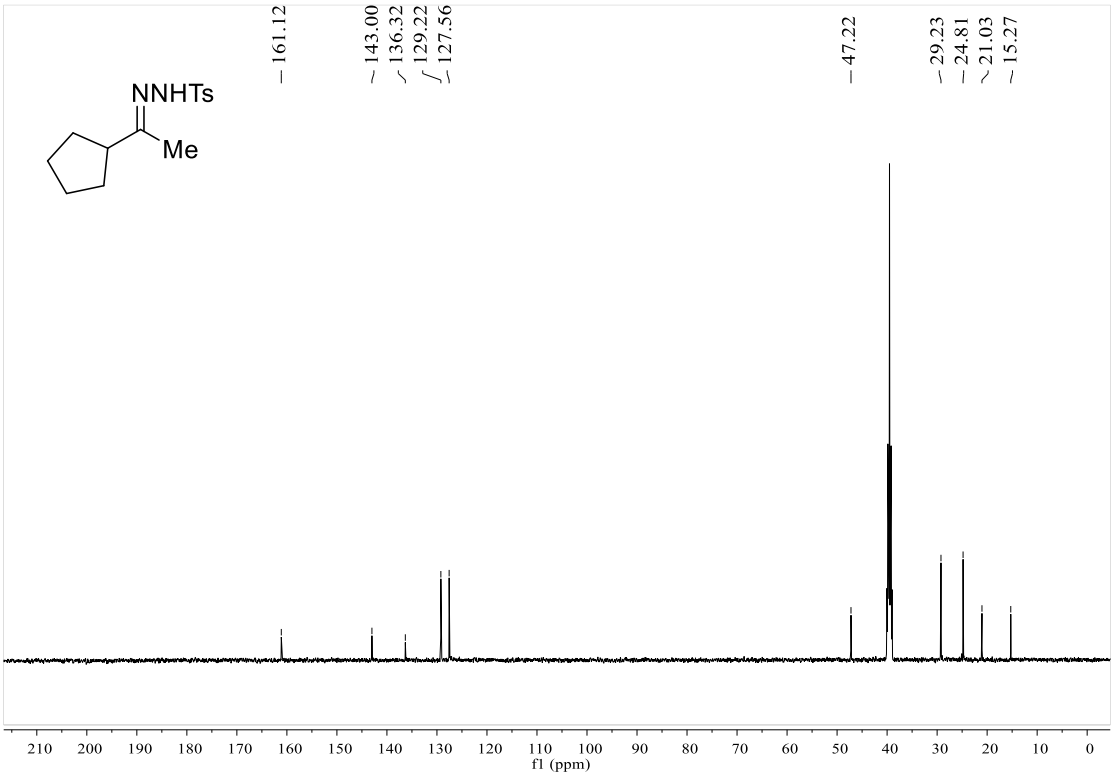


^{13}C NMR spectrum in DMSO-d_6.

***N′*-(1-cyclopentylethylidene)-4-methylbenzenesulfonohydrazide (24a)**:

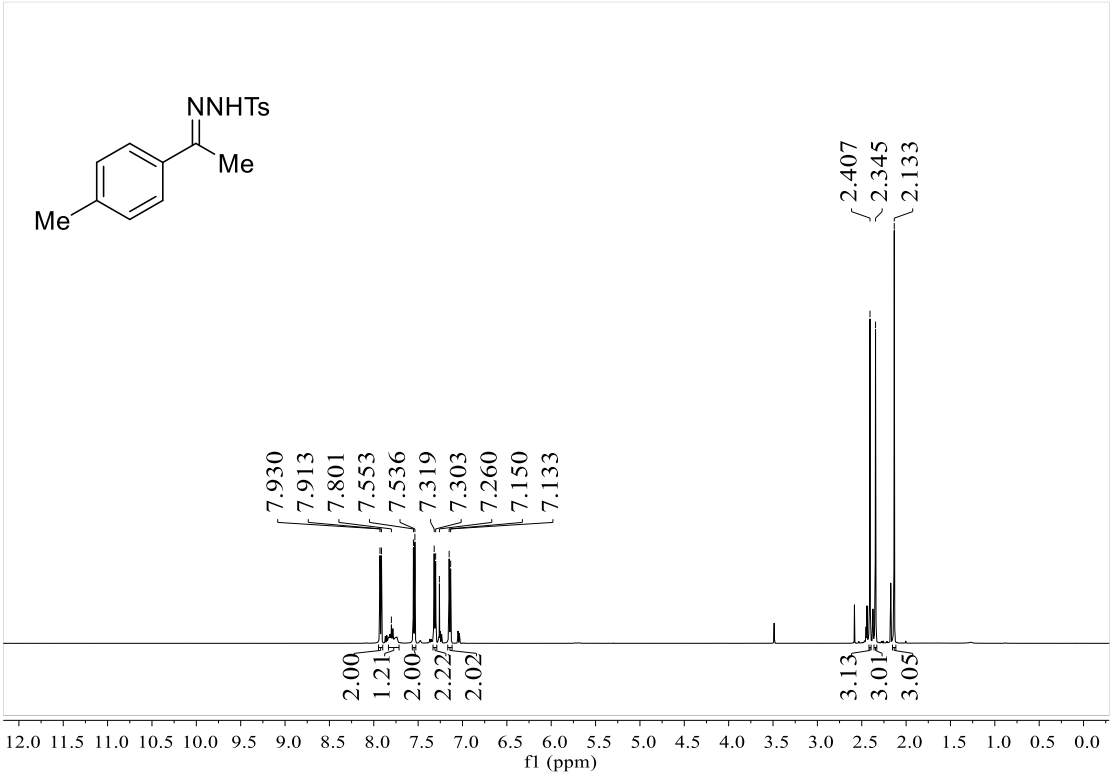


^{1}H NMR spectrum in DMSO-d_6.



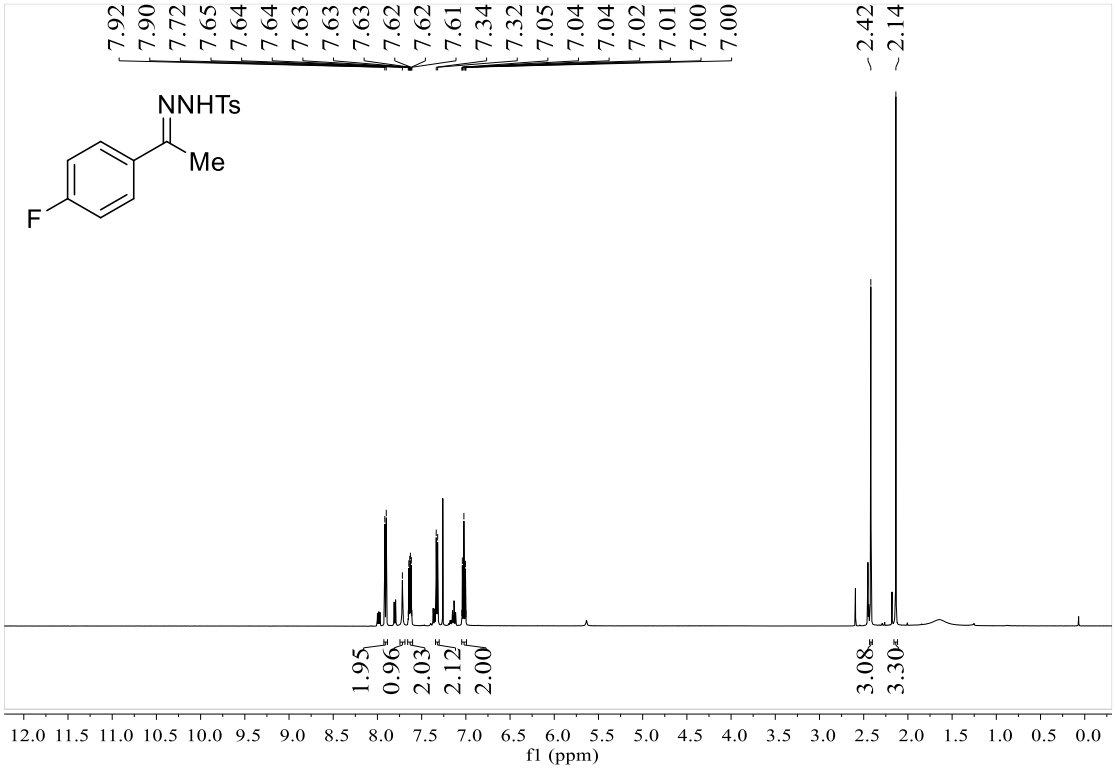
^{13}C NMR spectrum in DMSO-d_6.

4-methyl-*N*'-(1-(*p*-tolyl)ethylidene)benzenesulfonohydrazide (26a):



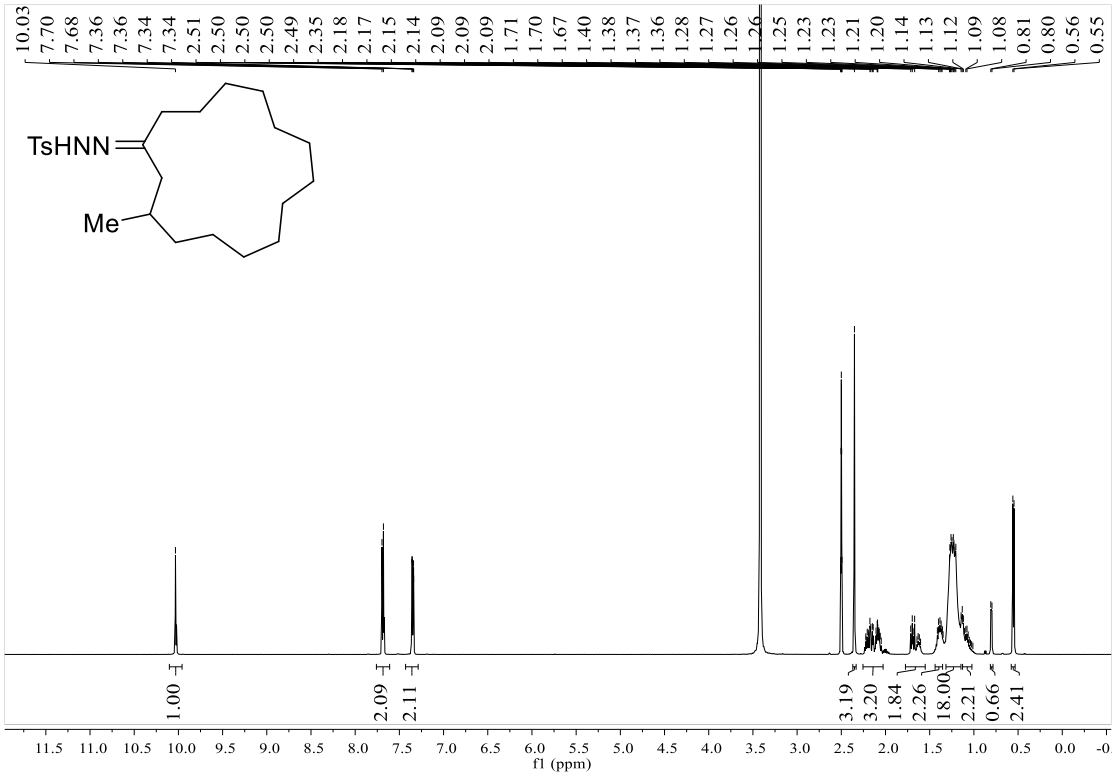
^{1}H NMR spectrum in $CDCl_3$.

***N'*-(1-(4-fluorophenyl)ethylidene)-4-methylbenzenesulfonohydrazide(27a)**:

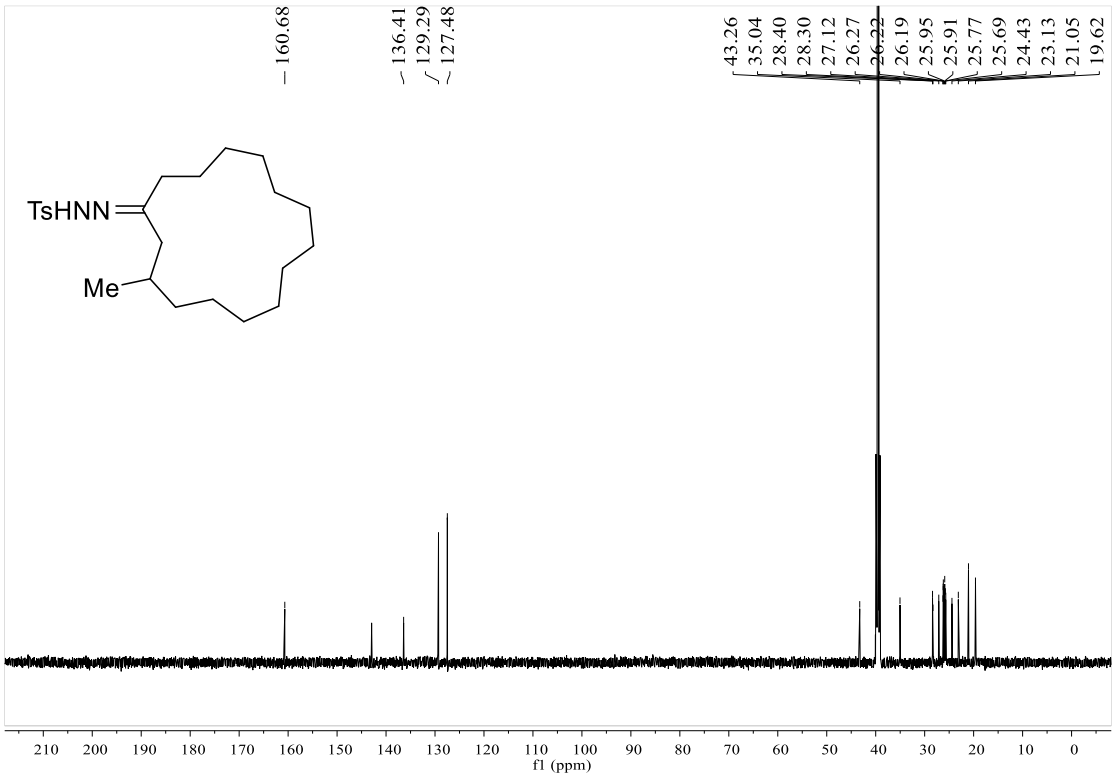


^{1}H NMR spectrum in $CDCl_3$.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (39a)**:

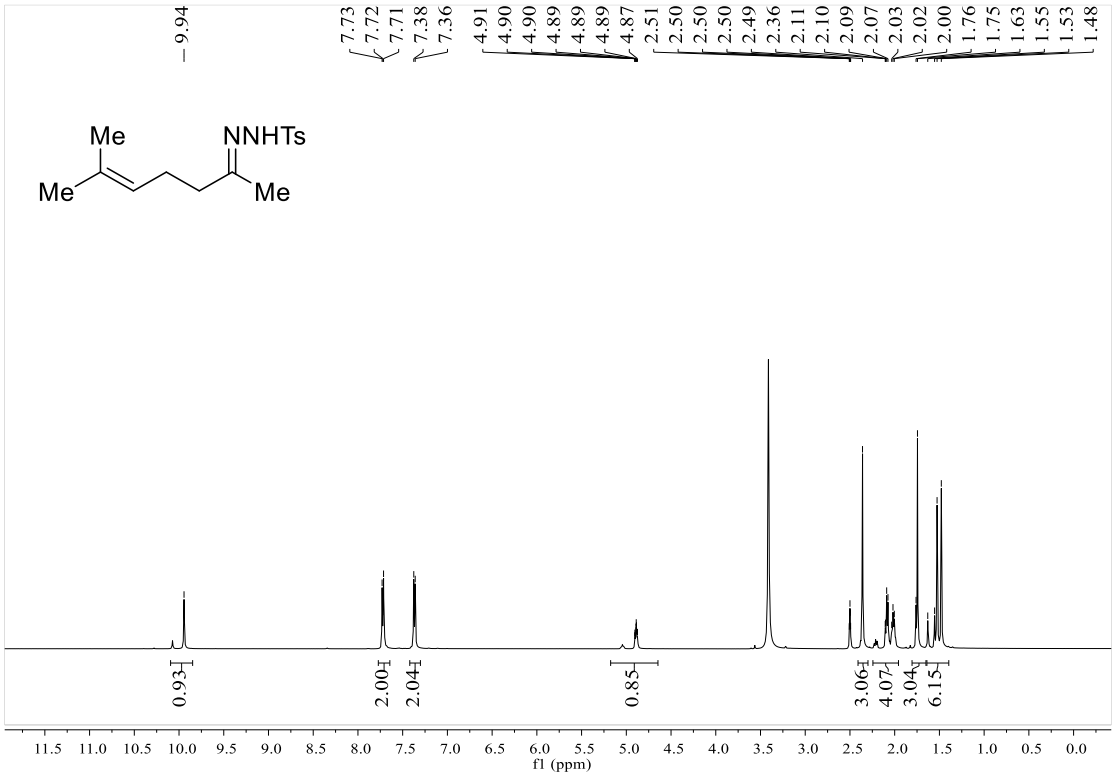


^{1}H NMR spectrum in DMSO-d_6.

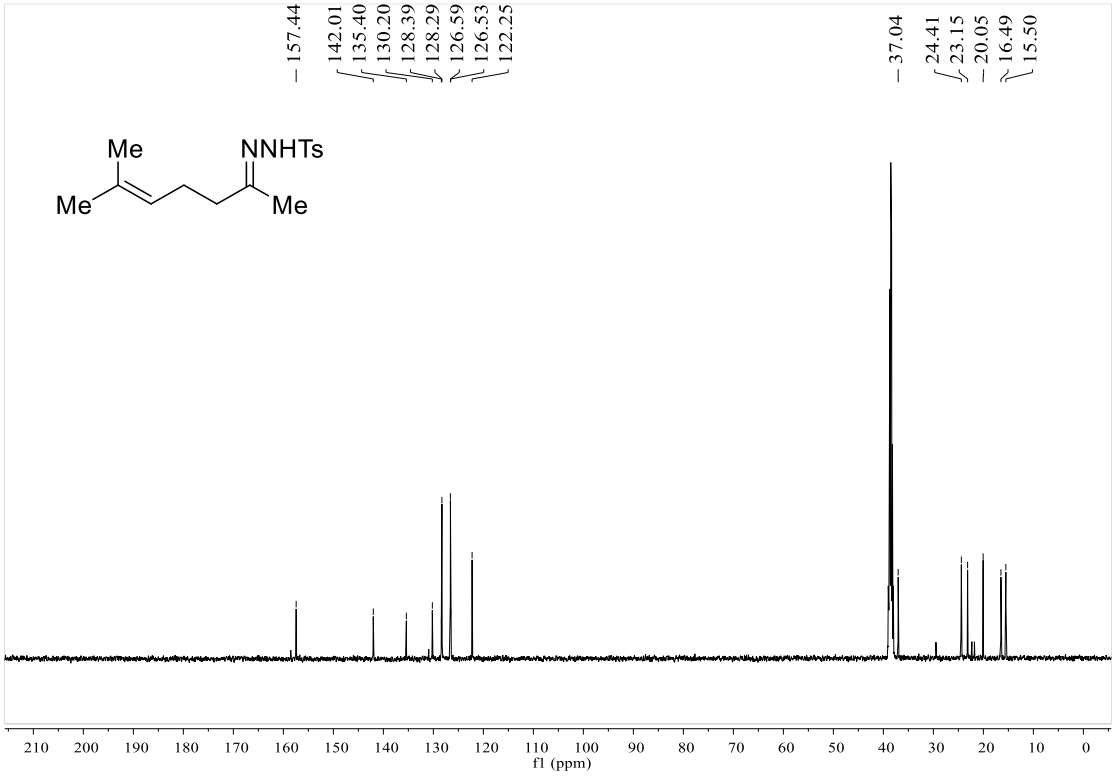


^{13}C NMR spectrum in DMSO-d_6.

***N'*-cyclooctylidene-4-methylbenzenesulfonohydrazide (40a)**:

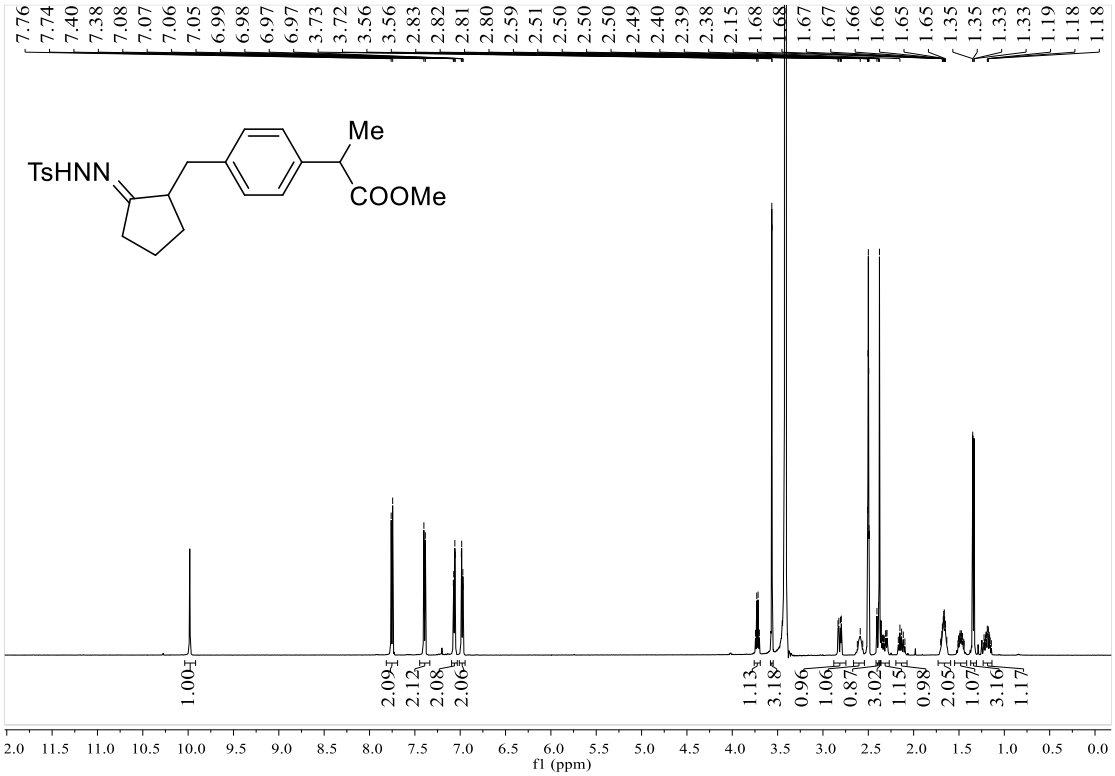


^{1}H NMR spectrum in DMSO-d_6.

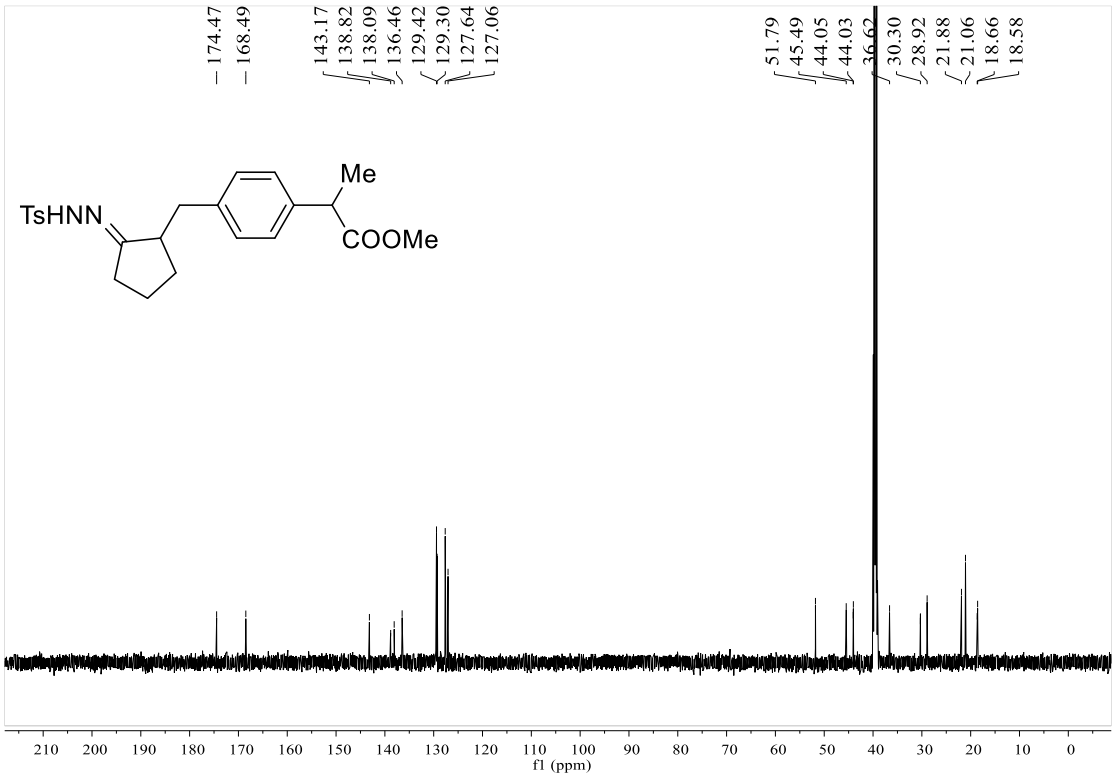


^{13}C NMR spectrum in DMSO-d_6.

***N*'-cyclooctylidene-4-methylbenzenesulfonohydrazide (41a)**:

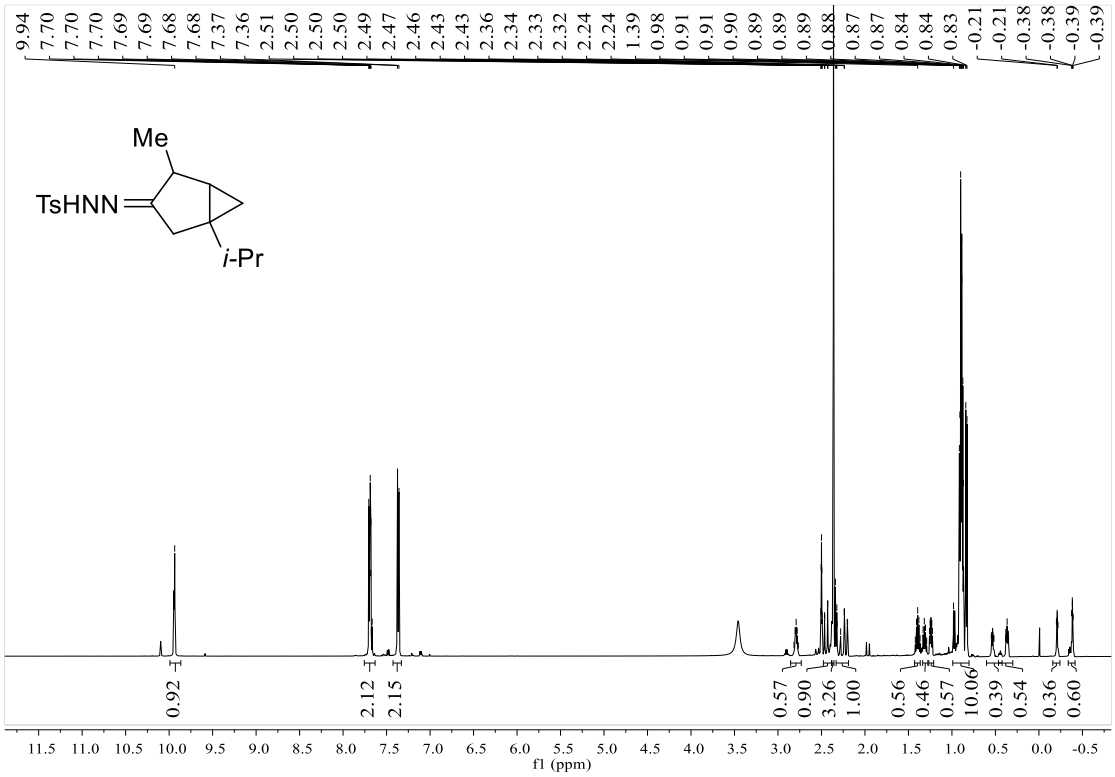


^{1}H NMR spectrum in DMSO-d_6.

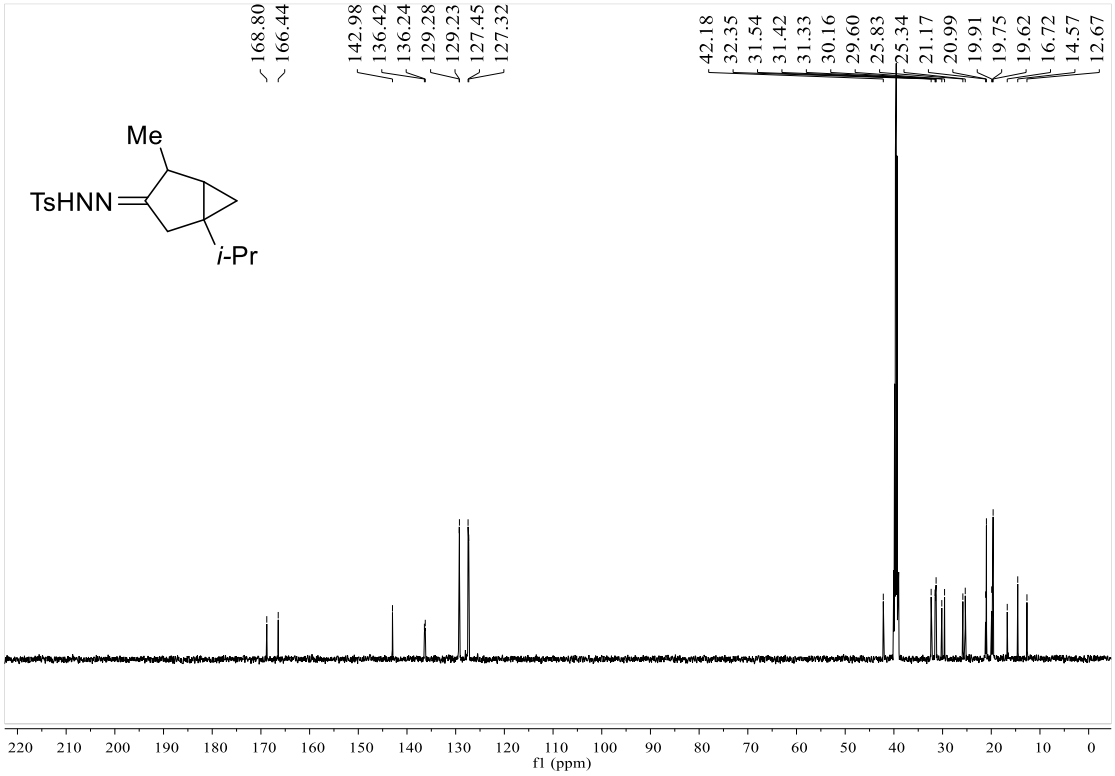


^{13}C NMR spectrum in DMSO-d_6.

***N'*-cyclooctylidene-4-methylbenzenesulfonohydrazide (42a)**:

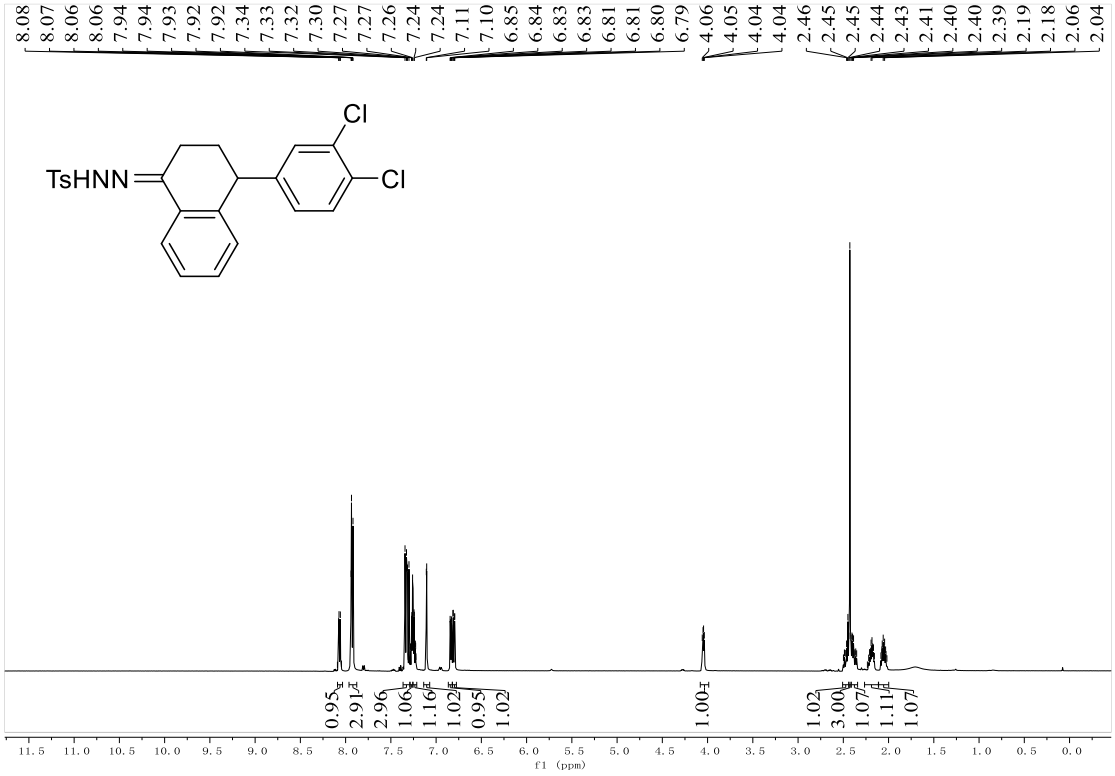


^{1}H NMR spectrum in DMSO-d_6.

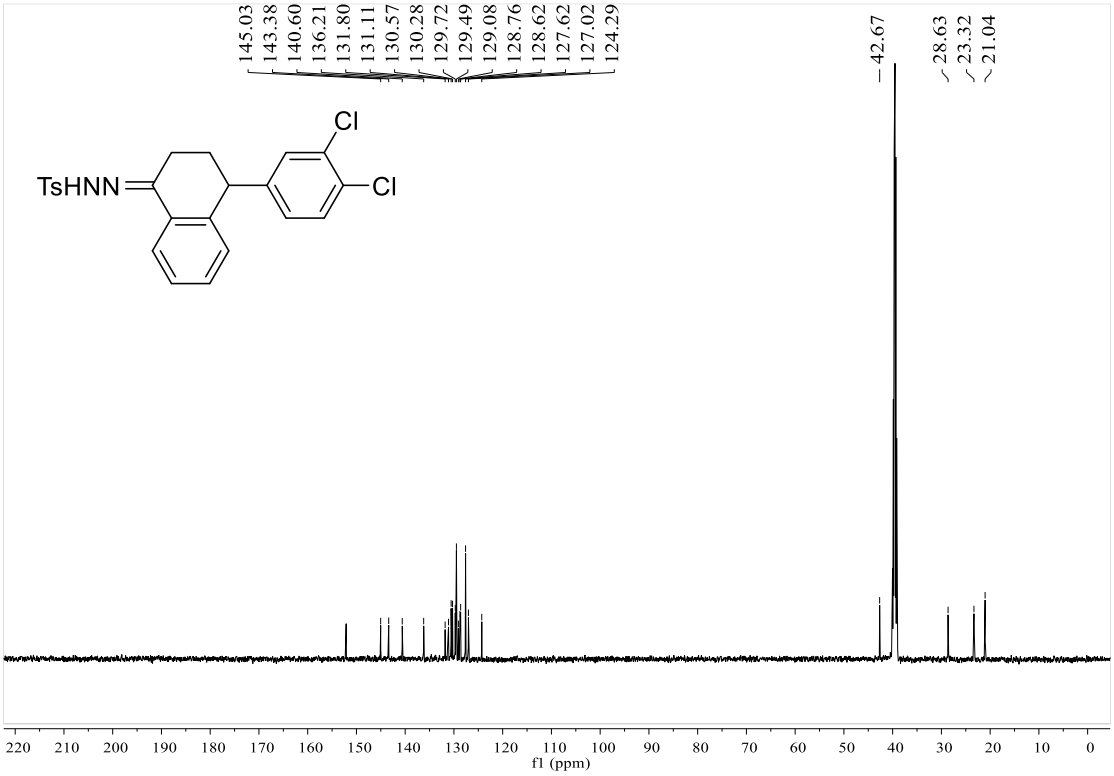


^{13}C NMR spectrum in DMSO-d_6.

***N*'-(4-(3,4-dichlorophenyl)-3,4-dihydronaphthalen-1(2*H*)-ylidene)-4-methylbenzenesulfonohydrazide (43a)**:

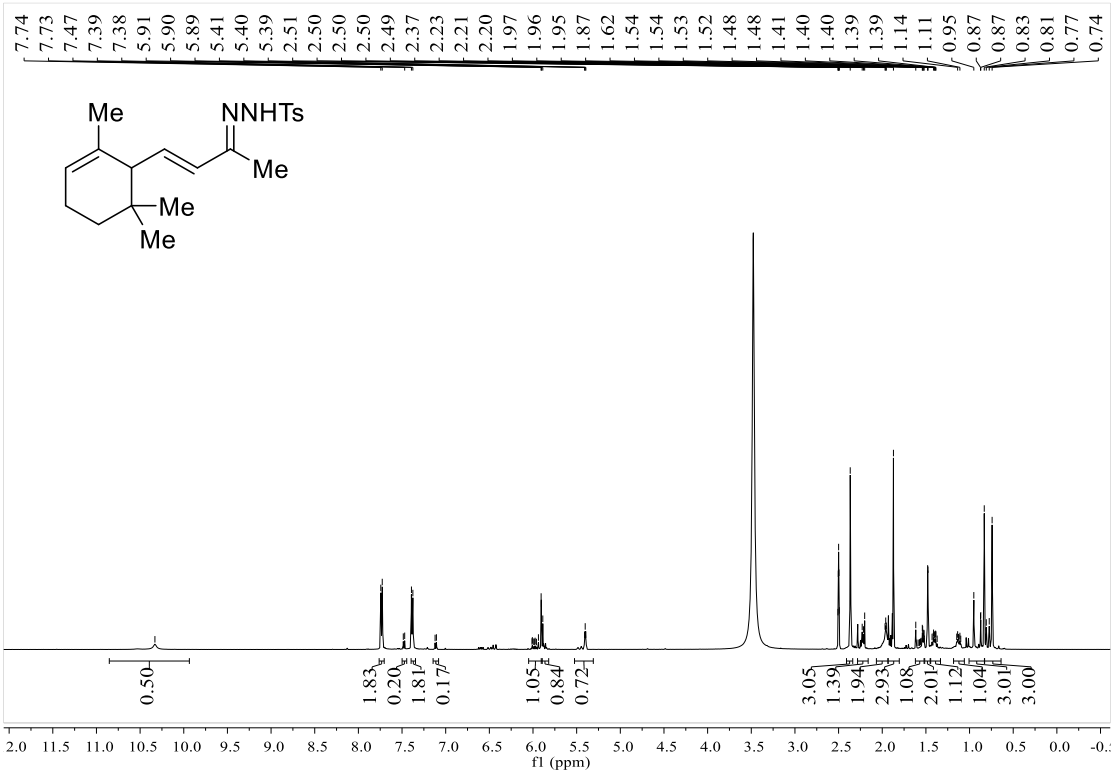


^{1}H NMR spectrum in $CDCl_3$.

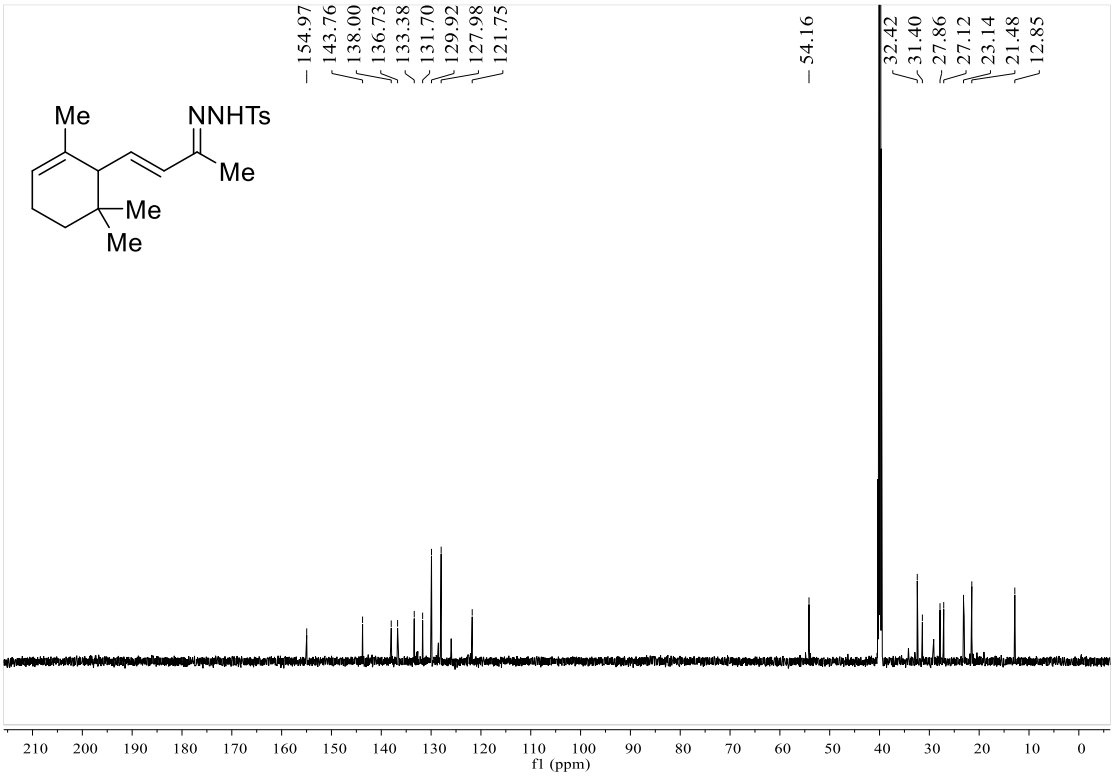


^{13}C NMR spectrum in DMSO-d_6.

***N'*-cyclooctylidene-4-methylbenzenesulfonohydrazide (44a)**:

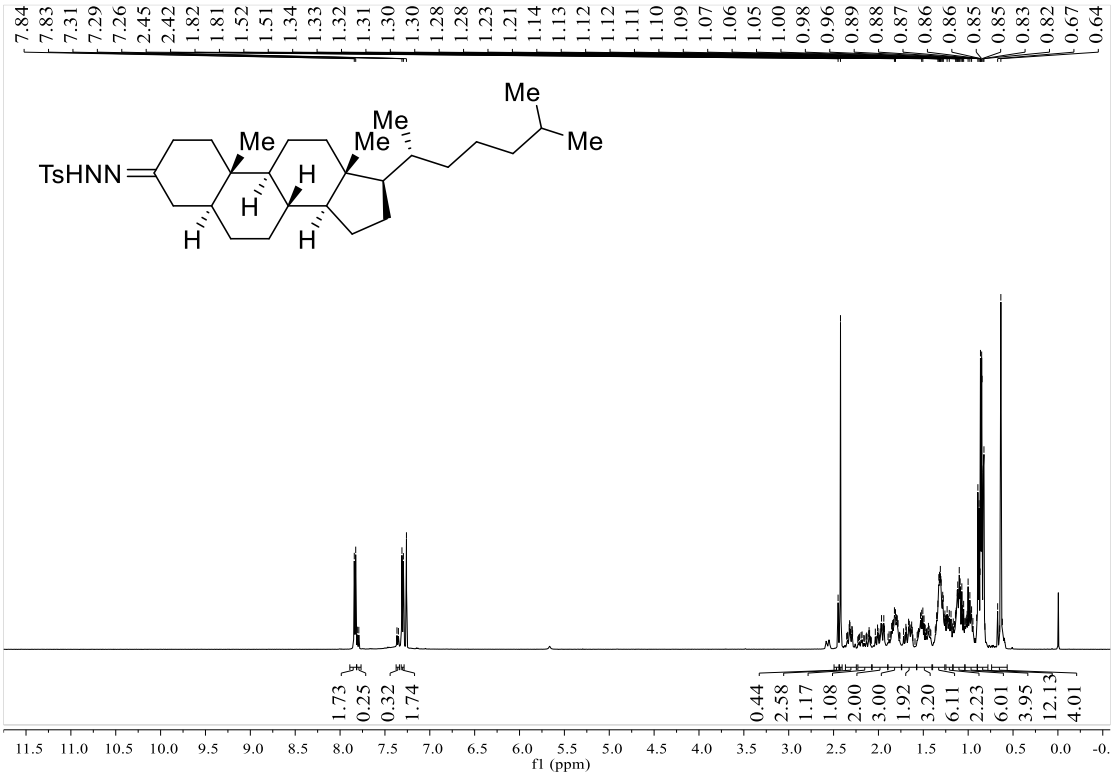


^{1}H NMR spectrum in DMSO-d_6.

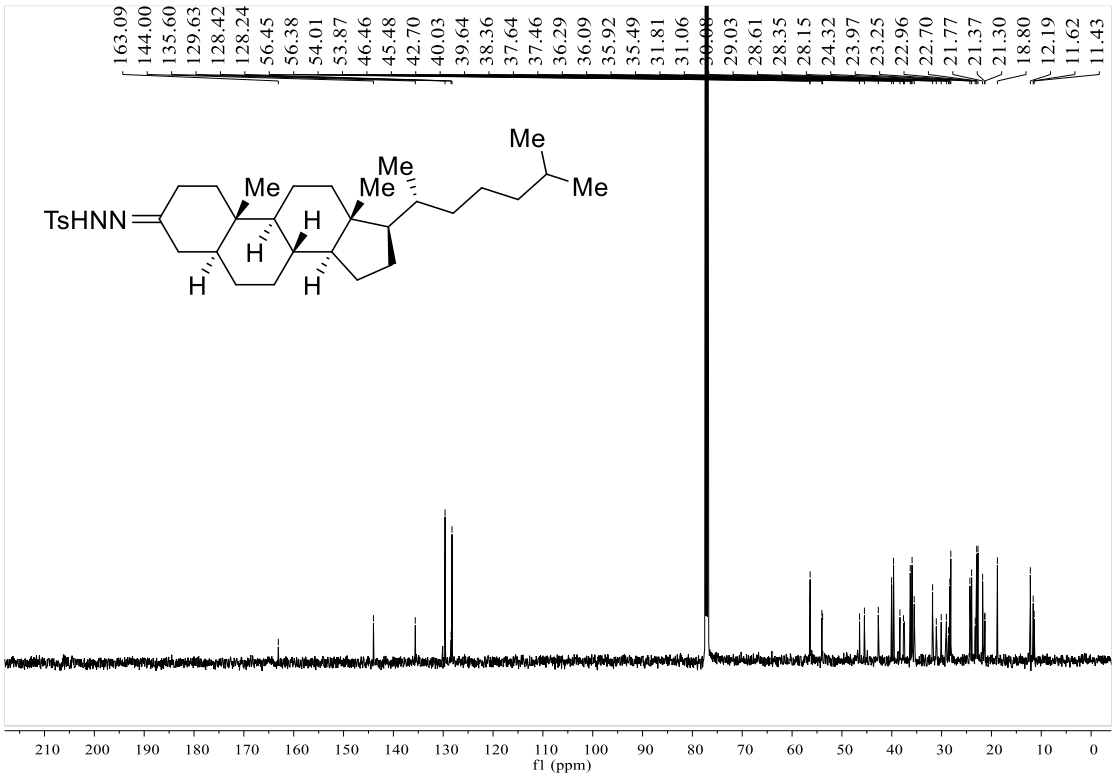


^{13}C NMR spectrum in DMSO-d_6.

***N'*-cyclooctylidene-4-methylbenzenesulfonohydrazide (45a)**:

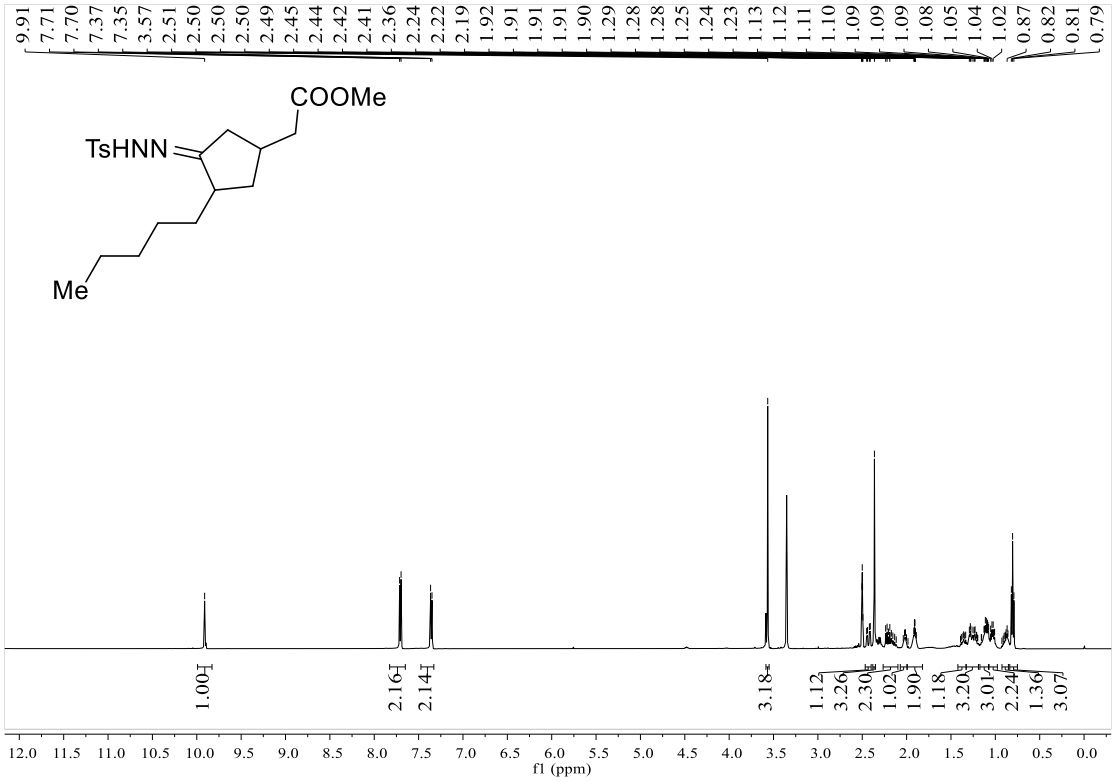


^{1}H NMR spectrum in $CDCl_3$.

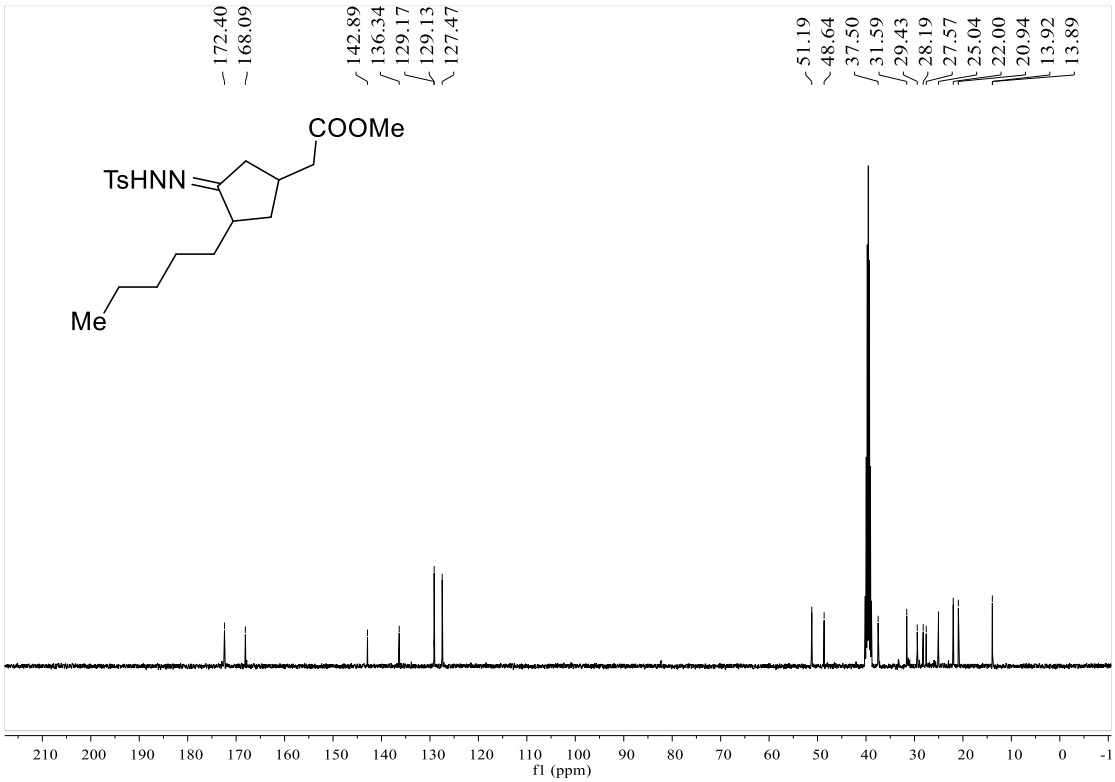


^{13}C NMR spectrum in $CDCl_3$.

***N'*-cyclooctylidene-4-methylbenzenesulfonohydrazide (46a)**:



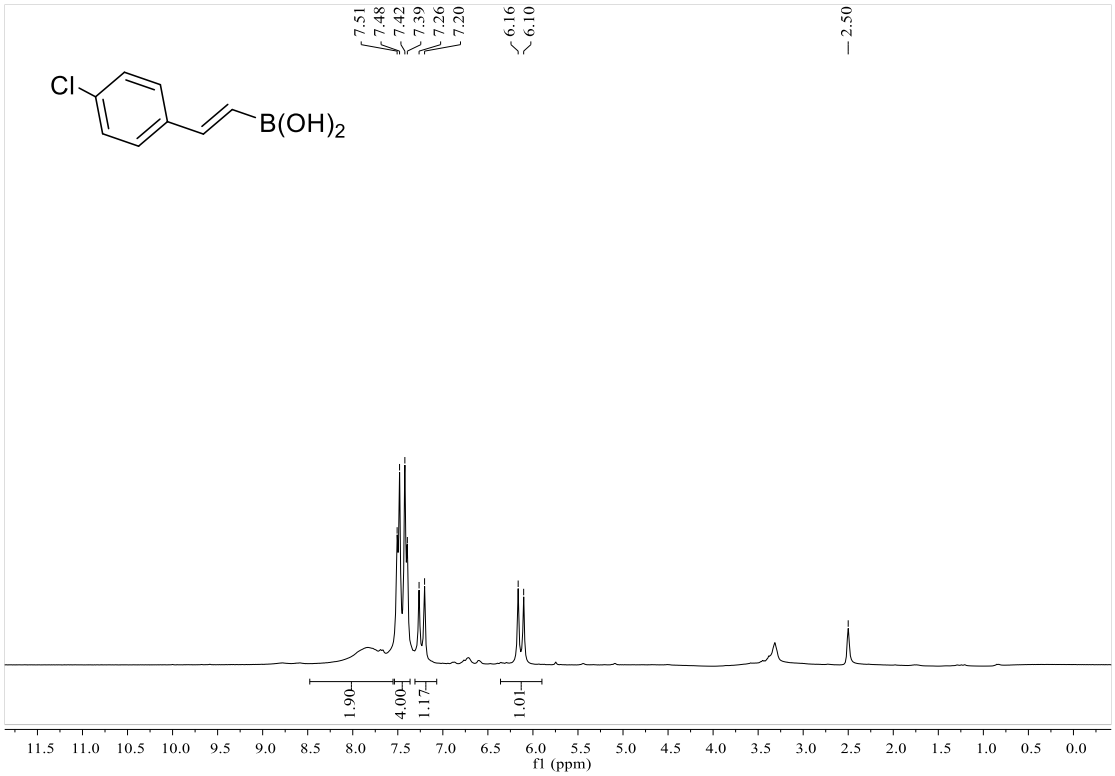
^{1}H NMR spectrum in DMSO-d_6.



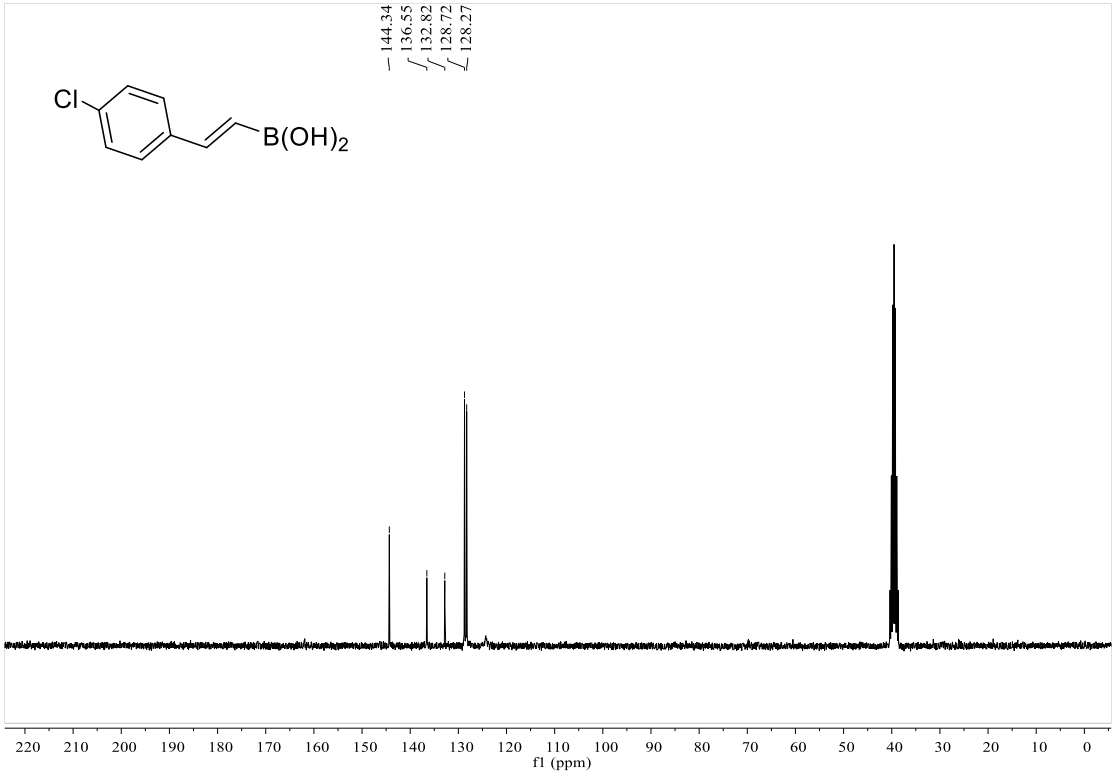
^{13}C NMR spectrum in DMSO-d_6.

9.2 NMR spectra of synthesized alkenyl boronic acids

(4-chlorostyryl)boronic acid (28b):

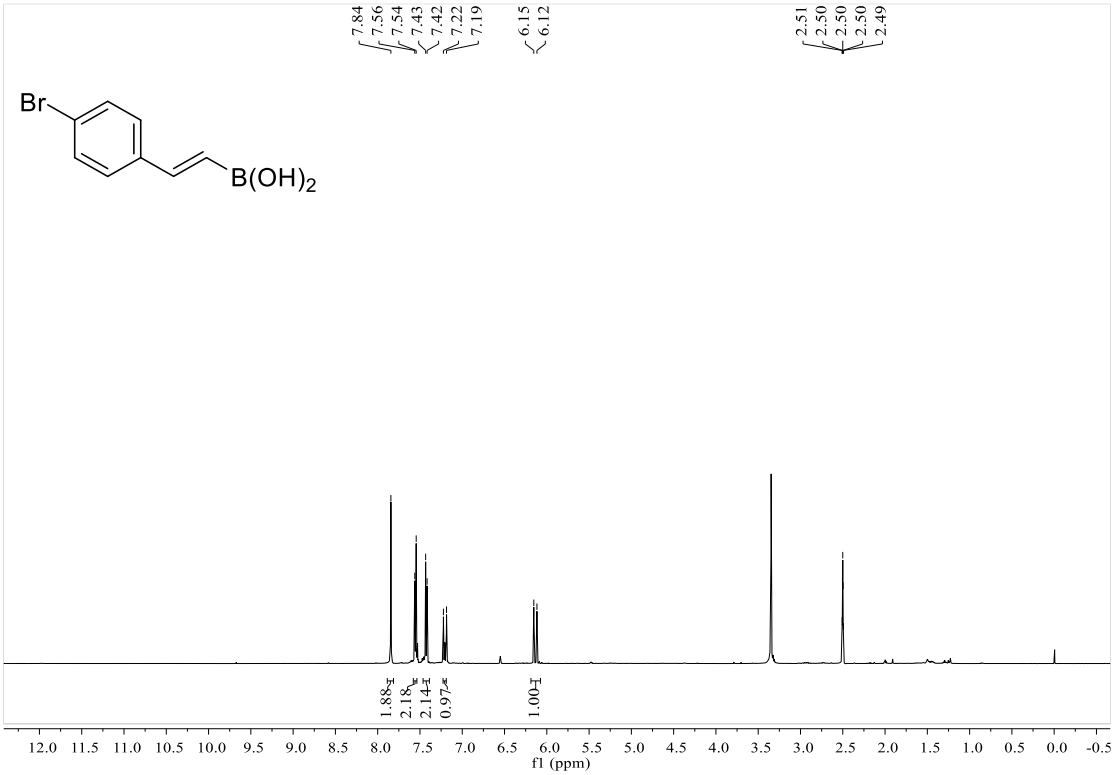


^{1}H NMR spectrum in DMSO-d_6.

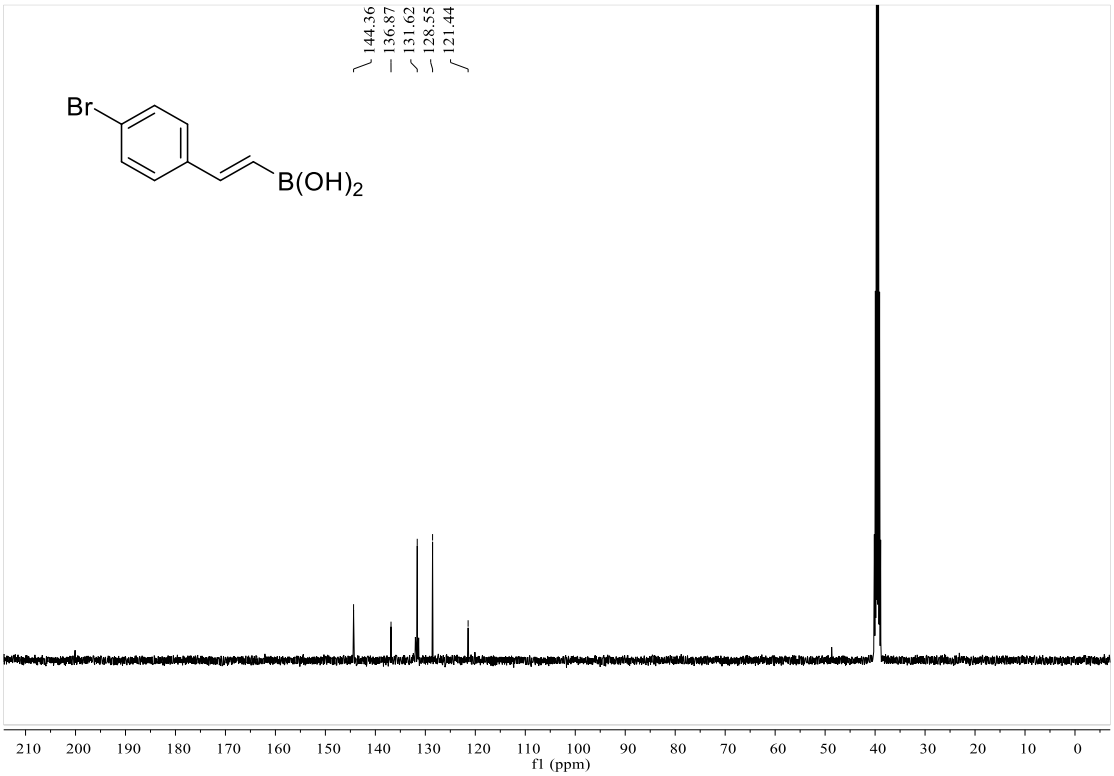


^{13}C NMR spectrum in DMSO-d_6.

(4-chlorostyryl)boronic acid (29b):

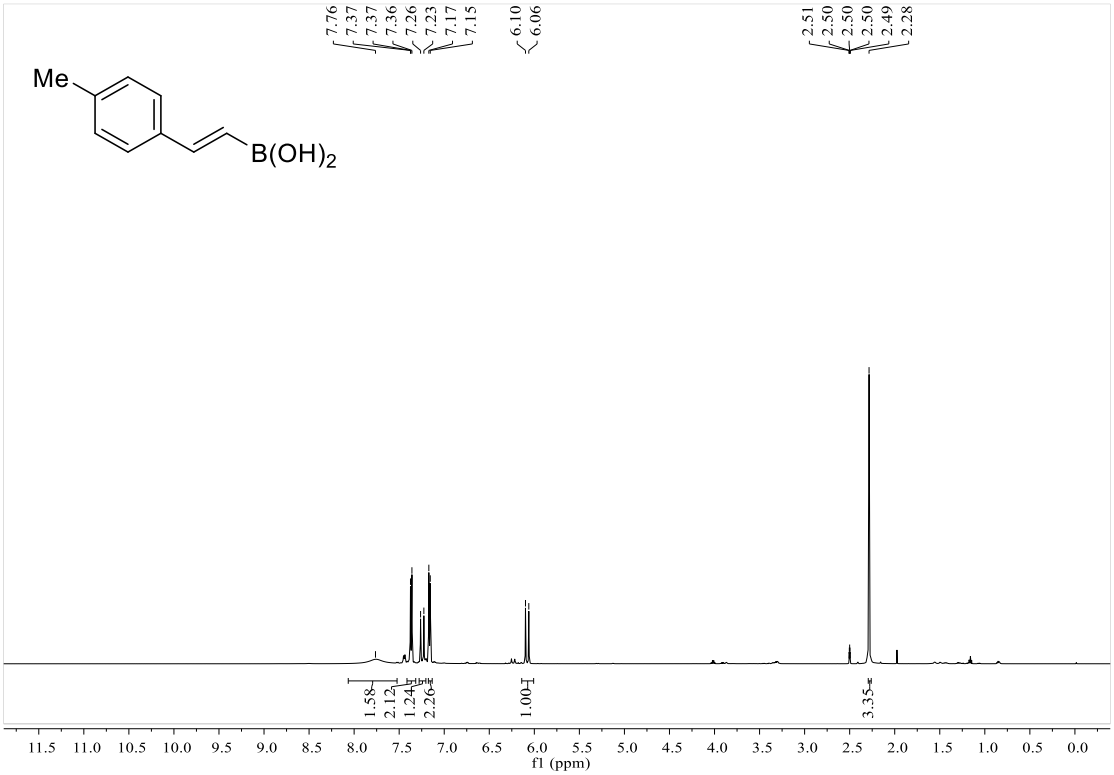


^{1}H NMR spectrum in DMSO-d_6.

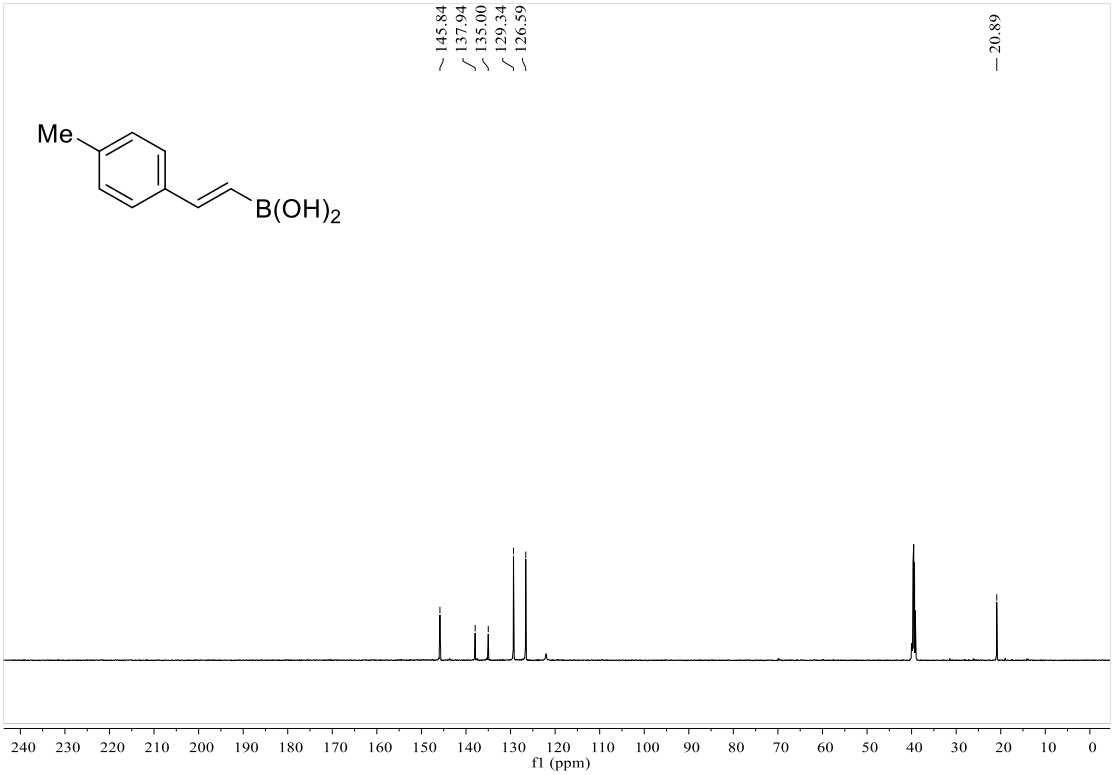


^{13}C NMR spectrum in DMSO-d_6.

(4-methylstyryl)boronic acid (30b):

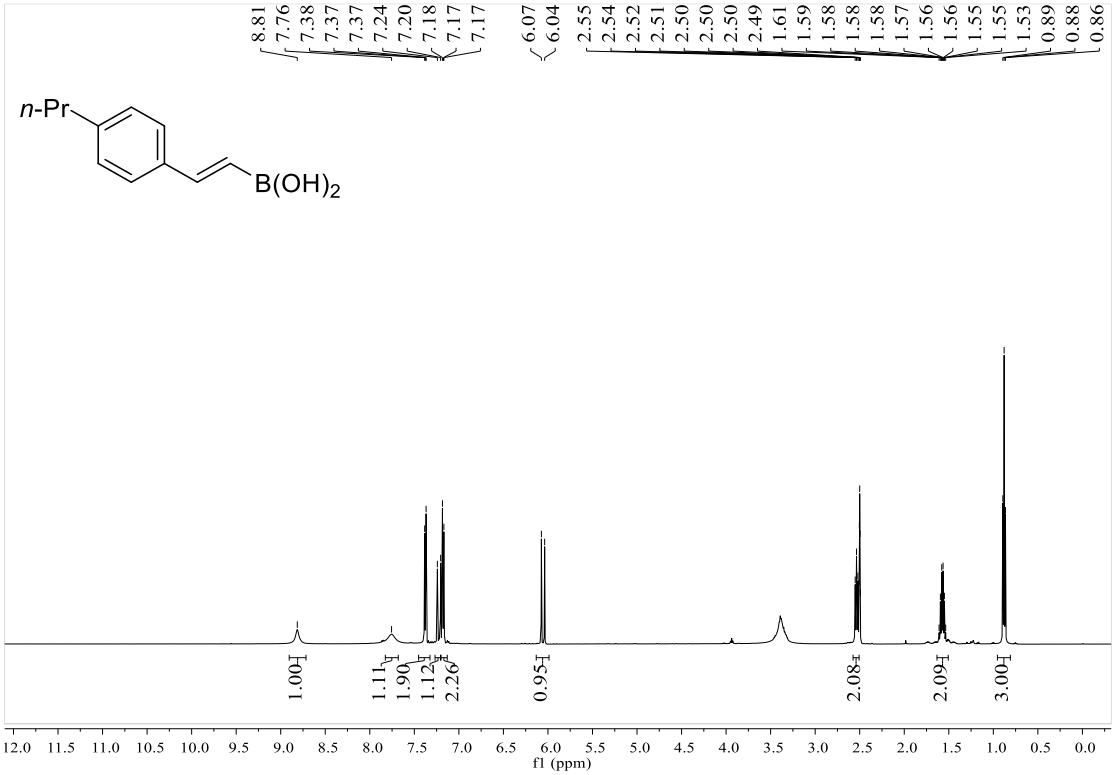


^{1}H NMR spectrum in DMSO-d_6.

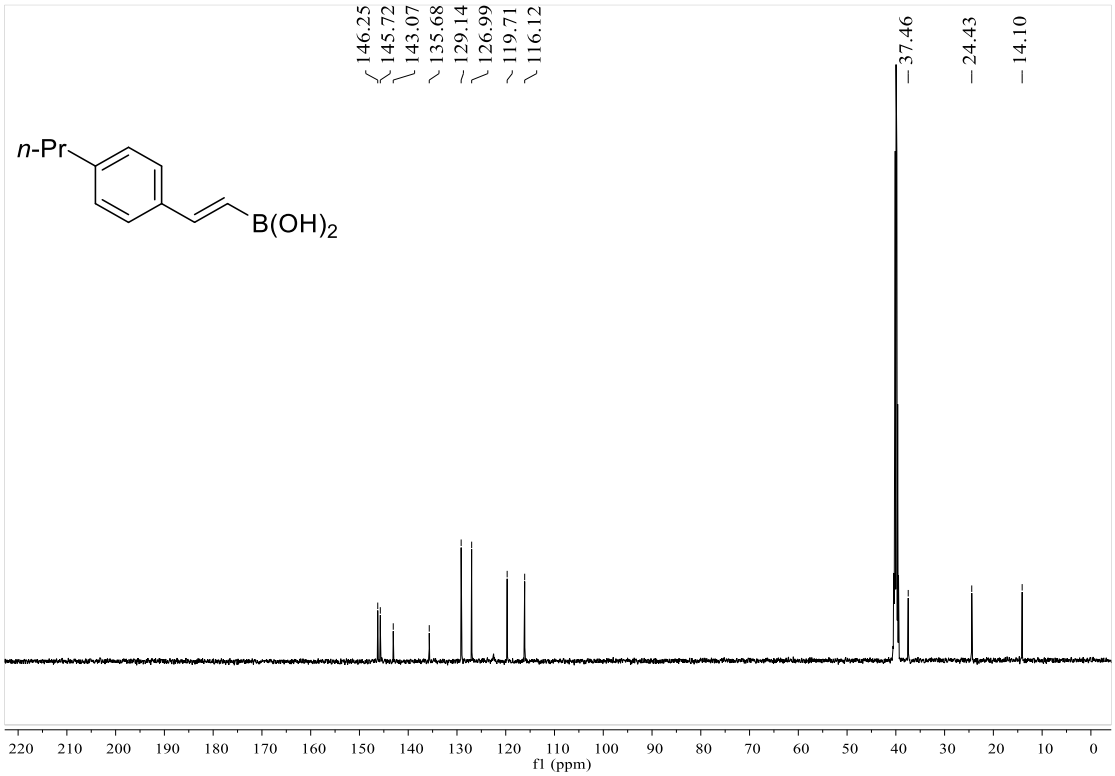


^{13}C NMR spectrum in DMSO-d_6.

(4-propylstyryl)boronic acid (31b):

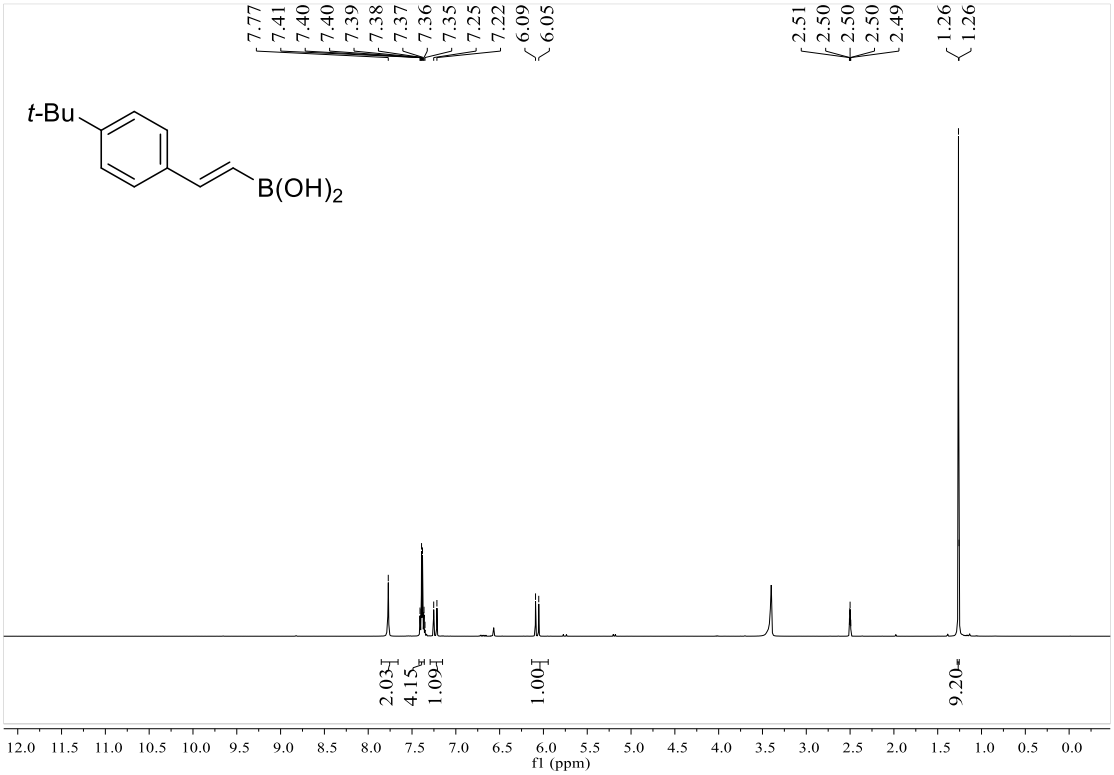


^{1}H NMR spectrum in DMSO-d_6.

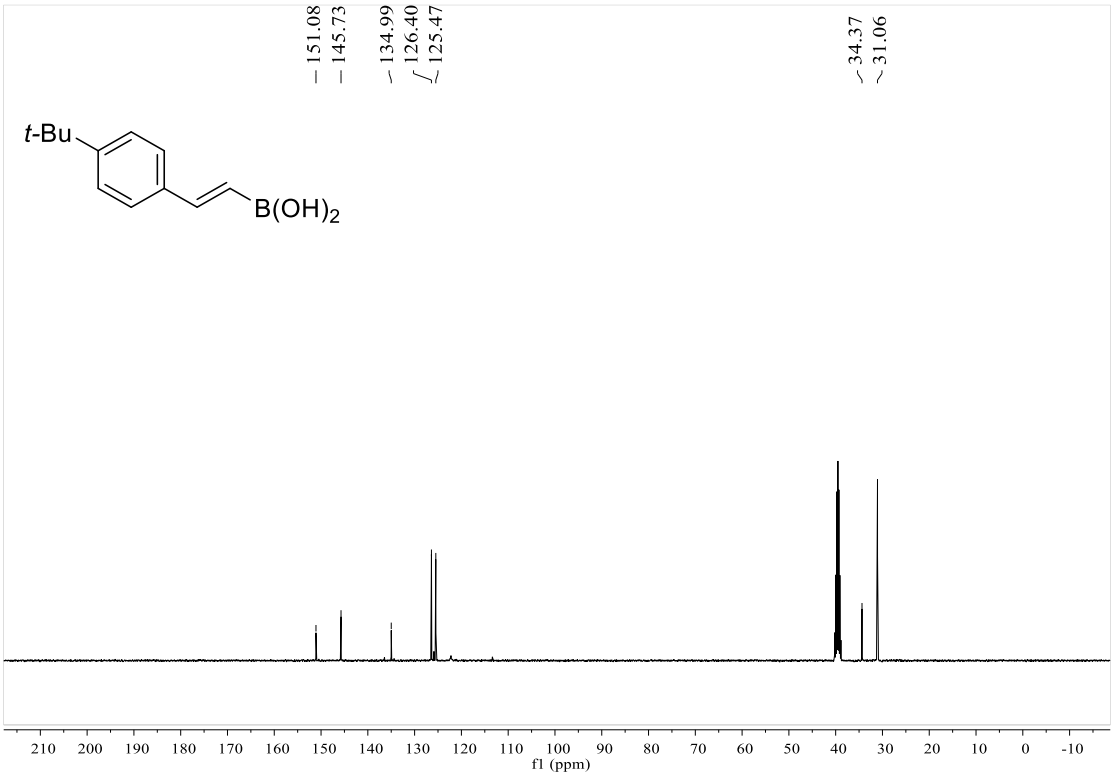


^{13}C NMR spectrum in DMSO-d_6.

(4-(tert-butyl)styryl)boronic acid (32b):

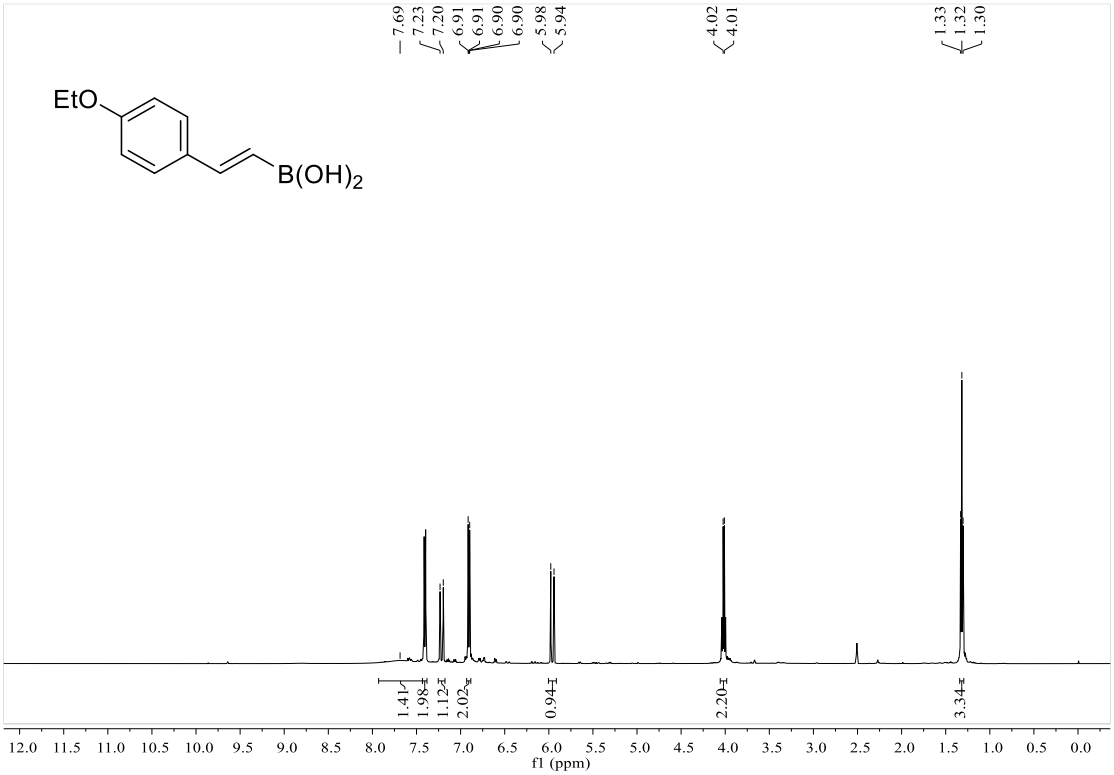


^{1}H NMR spectrum in DMSO-d_6.

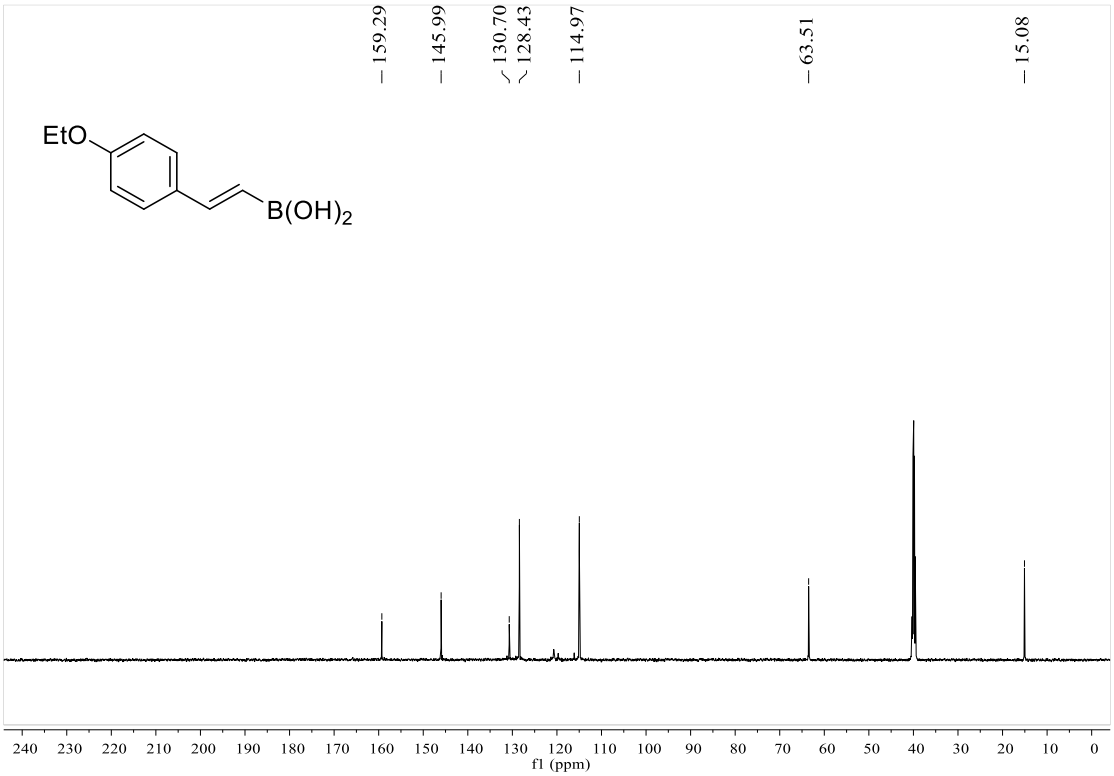


^{13}C NMR spectrum in DMSO-d_6.

(4-ethoxystyryl)boronic acid (33b):

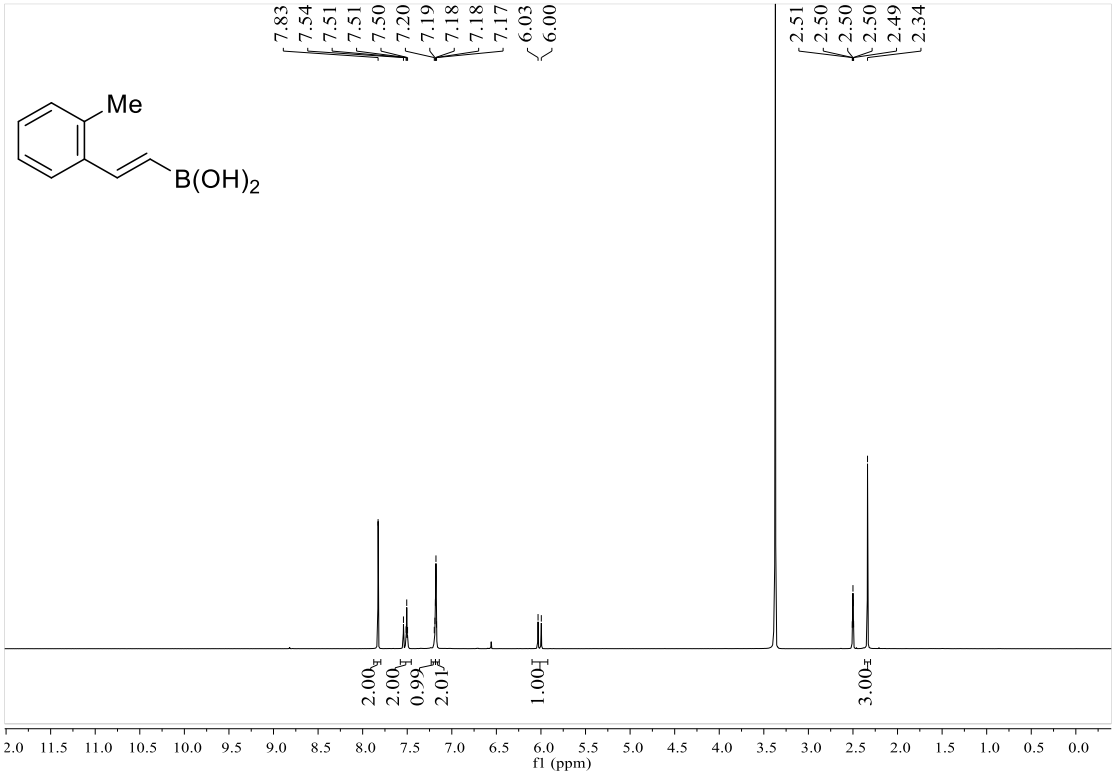


^{1}H NMR spectrum in DMSO-d_6.

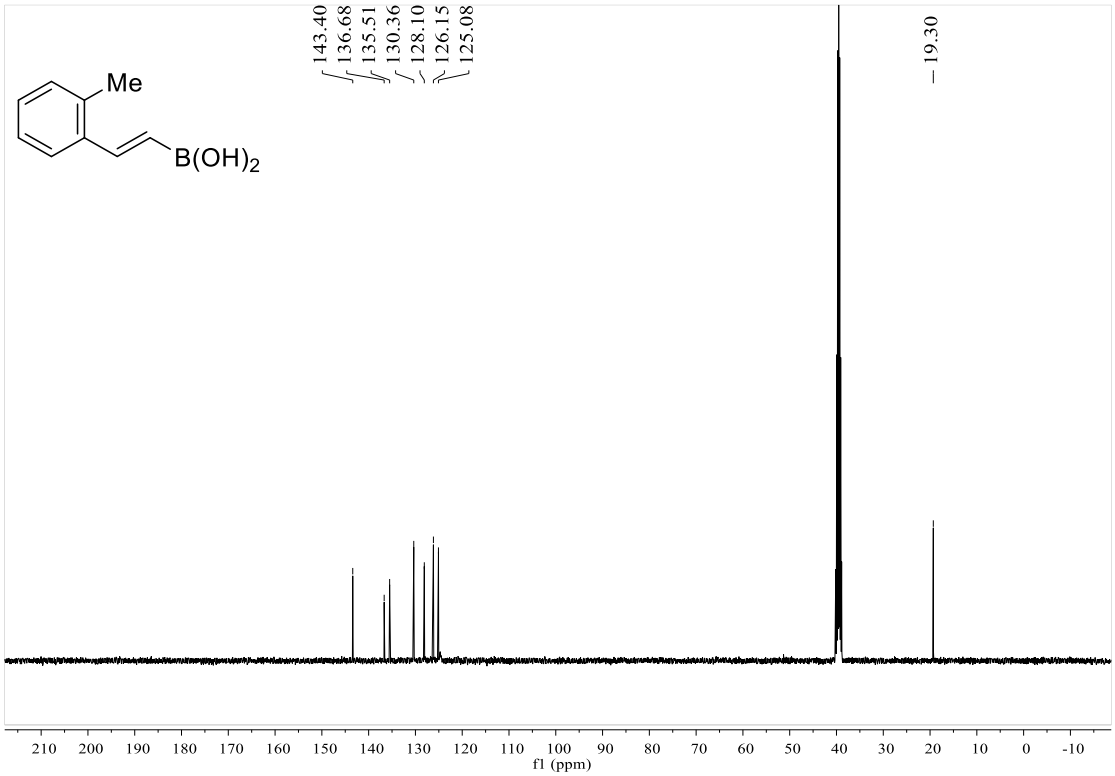


^{13}C NMR spectrum in DMSO-d_6.

(2-methylstyryl)boronic acid (34b):

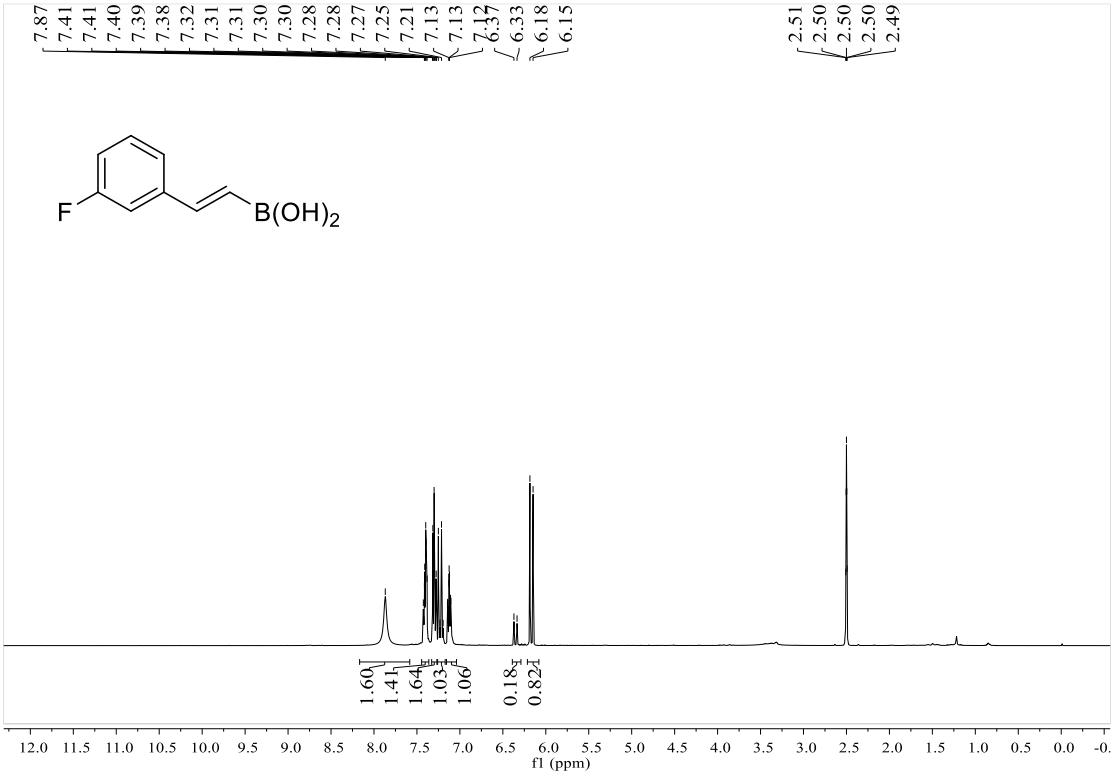


^{1}H NMR spectrum in DMSO-d_6.

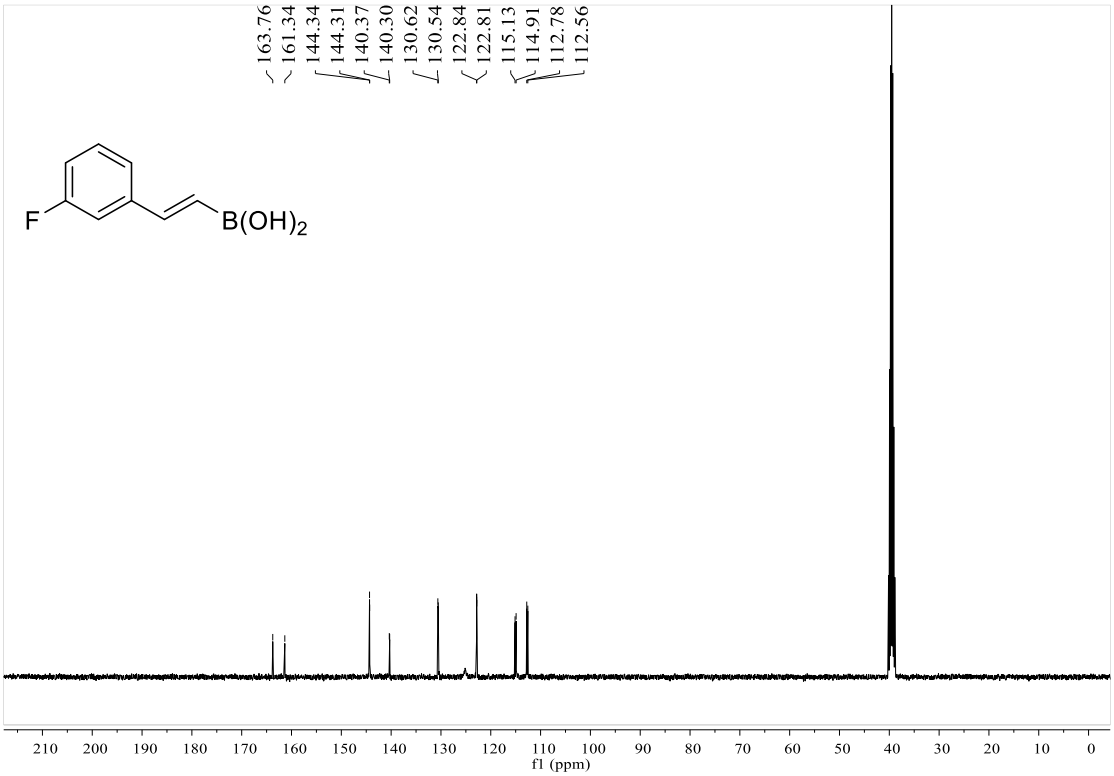


^{13}C NMR spectrum in DMSO-d_6.

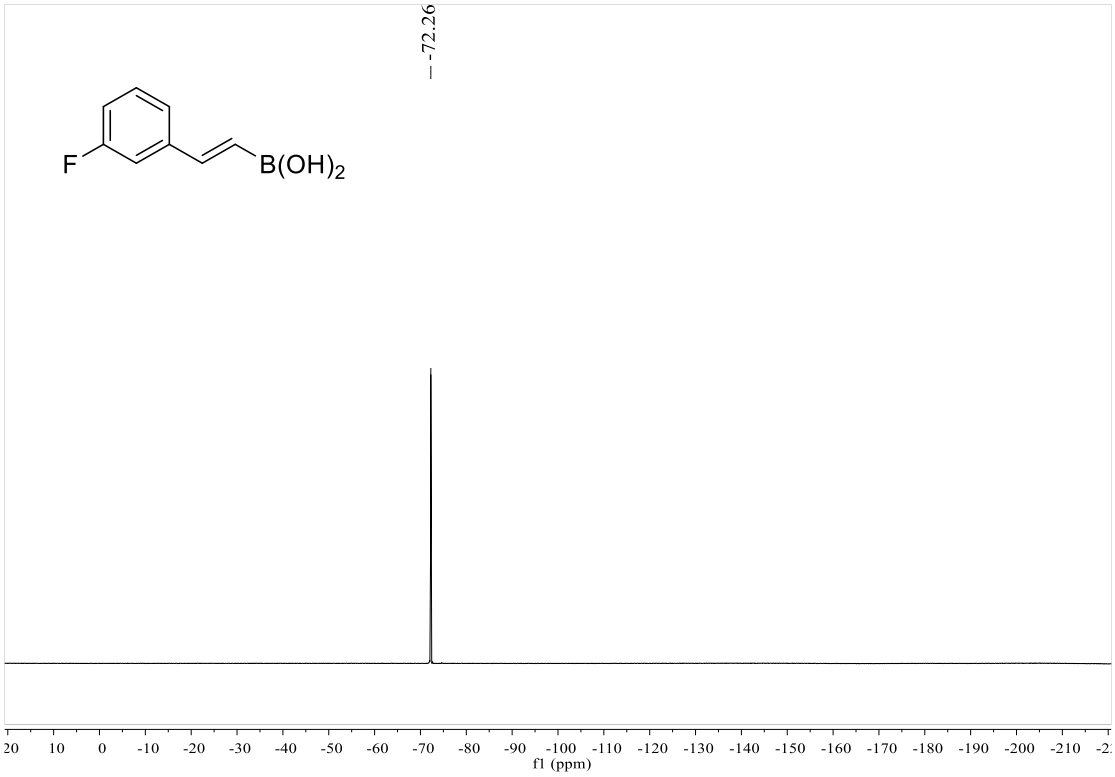
(3-fluorostyryl)boronic acid (35b):



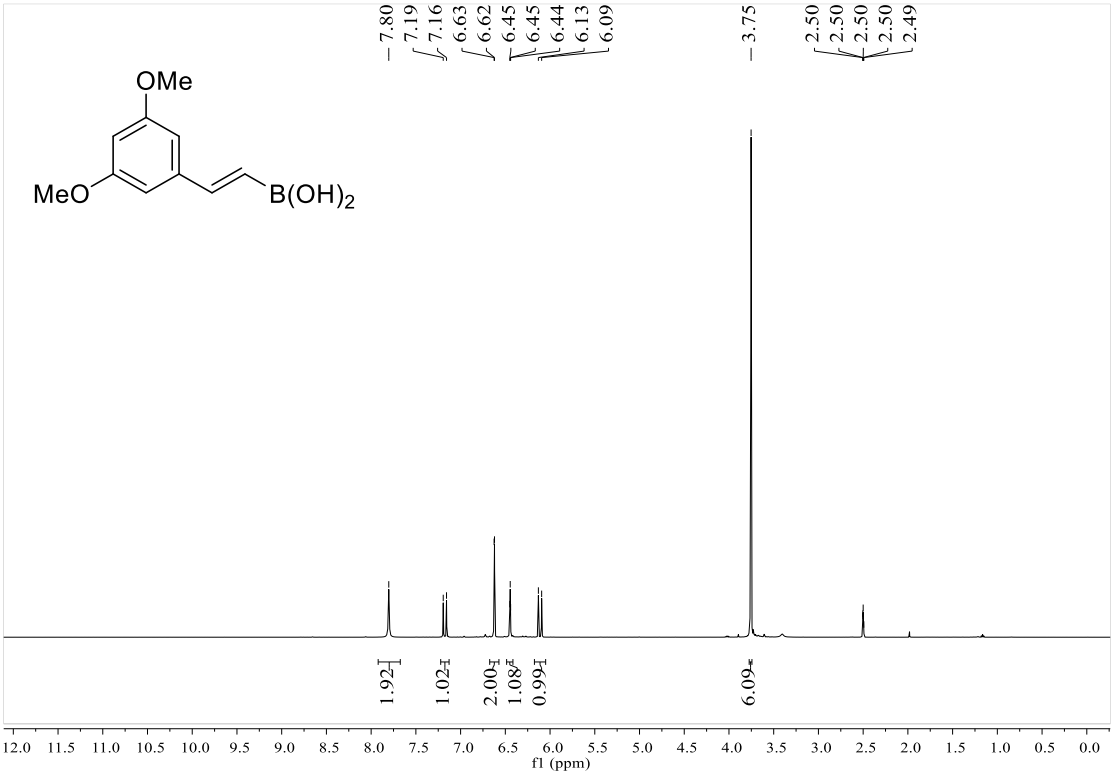
^{1}H NMR spectrum in DMSO-d_6.



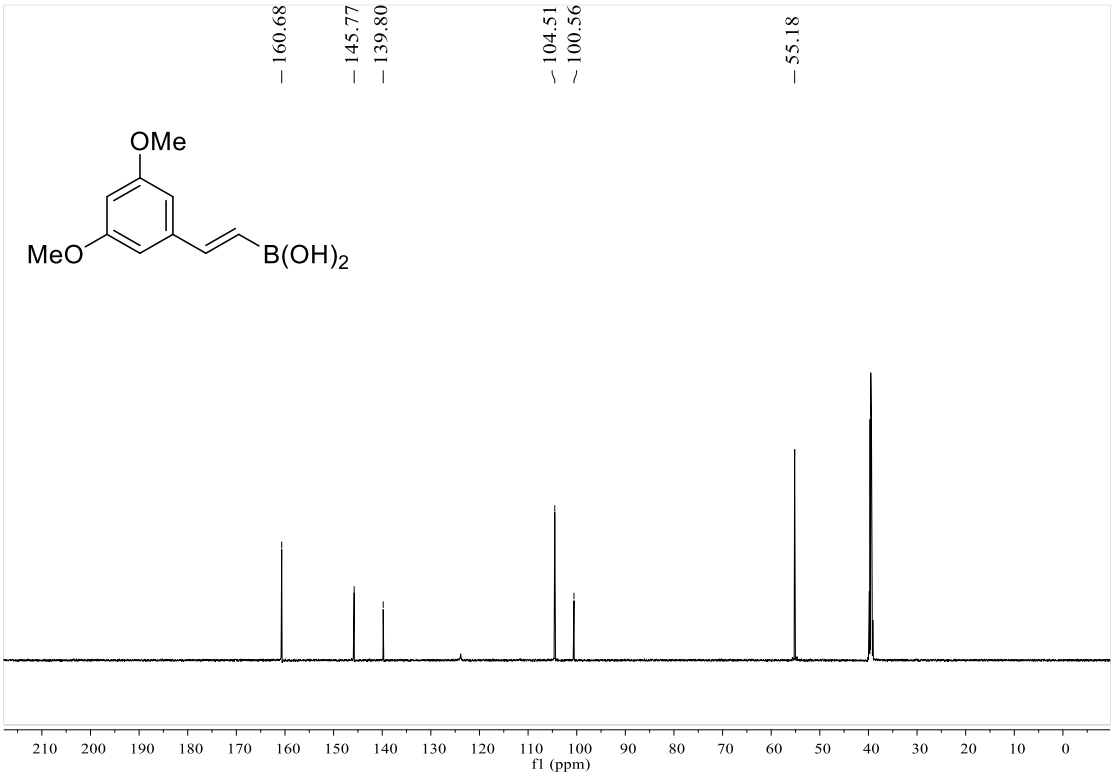
^{19}F NMR spectrum in DMSO-d_6.



(3,5-dimethoxystyryl)boronic acid (36b):

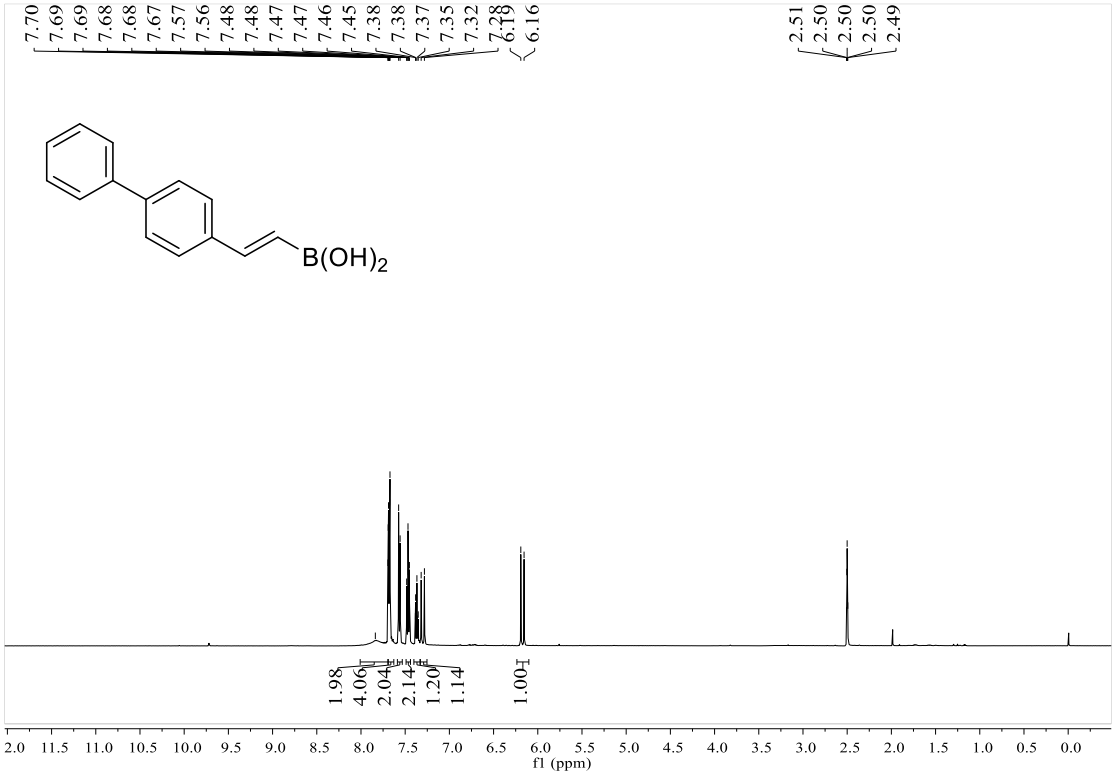


^{1}H NMR spectrum in DMSO-d_6.

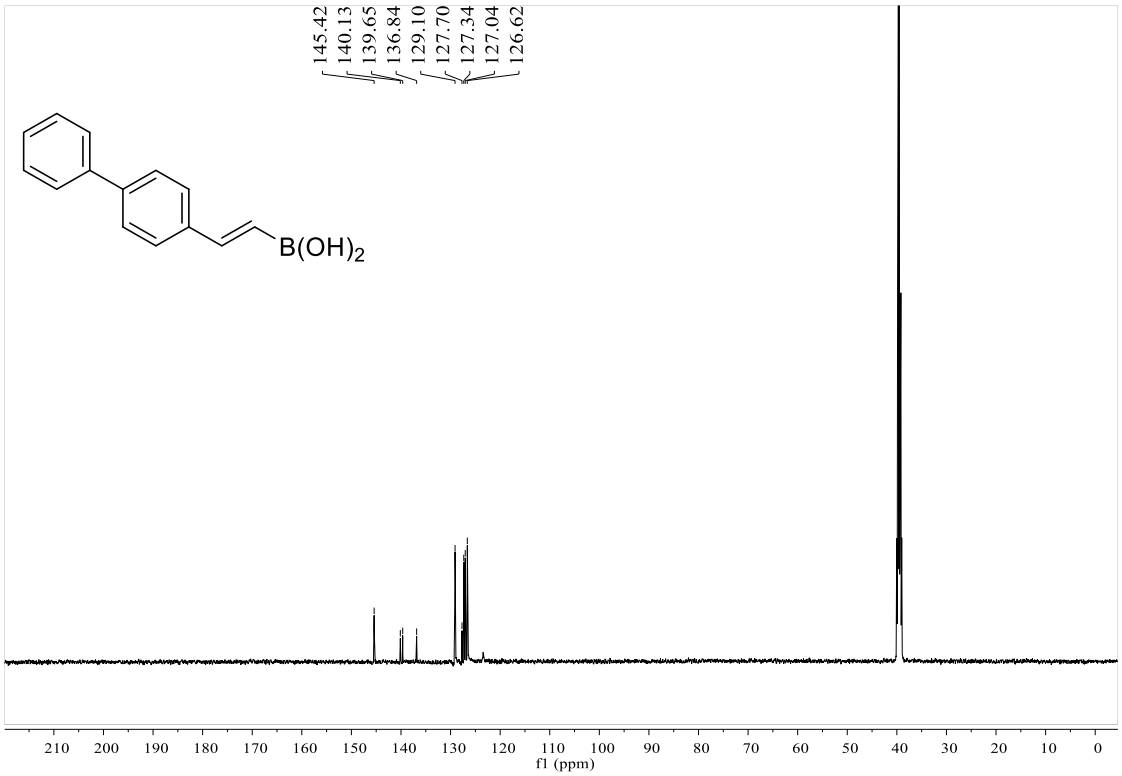


^{13}C NMR spectrum in DMSO-d_6.

(2-([1,1'-biphenyl]-4-yl)vinyl)boronic acid (37b):

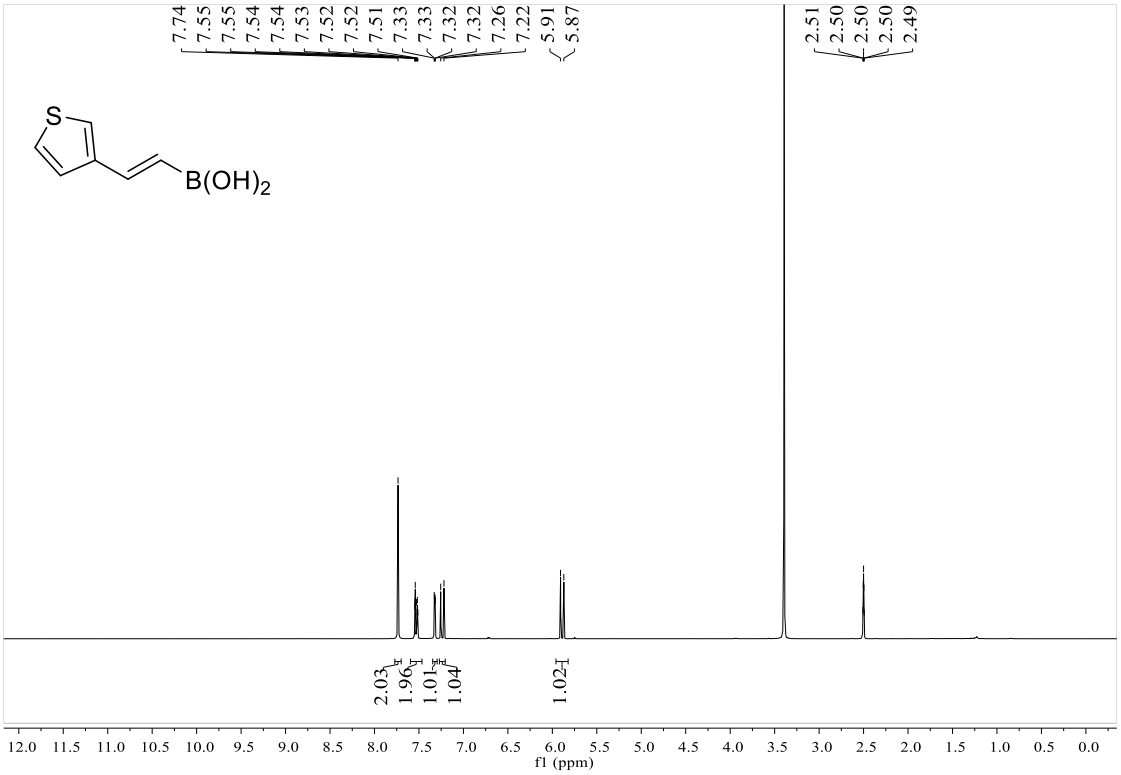


^{1}H NMR spectrum in DMSO-d_6.

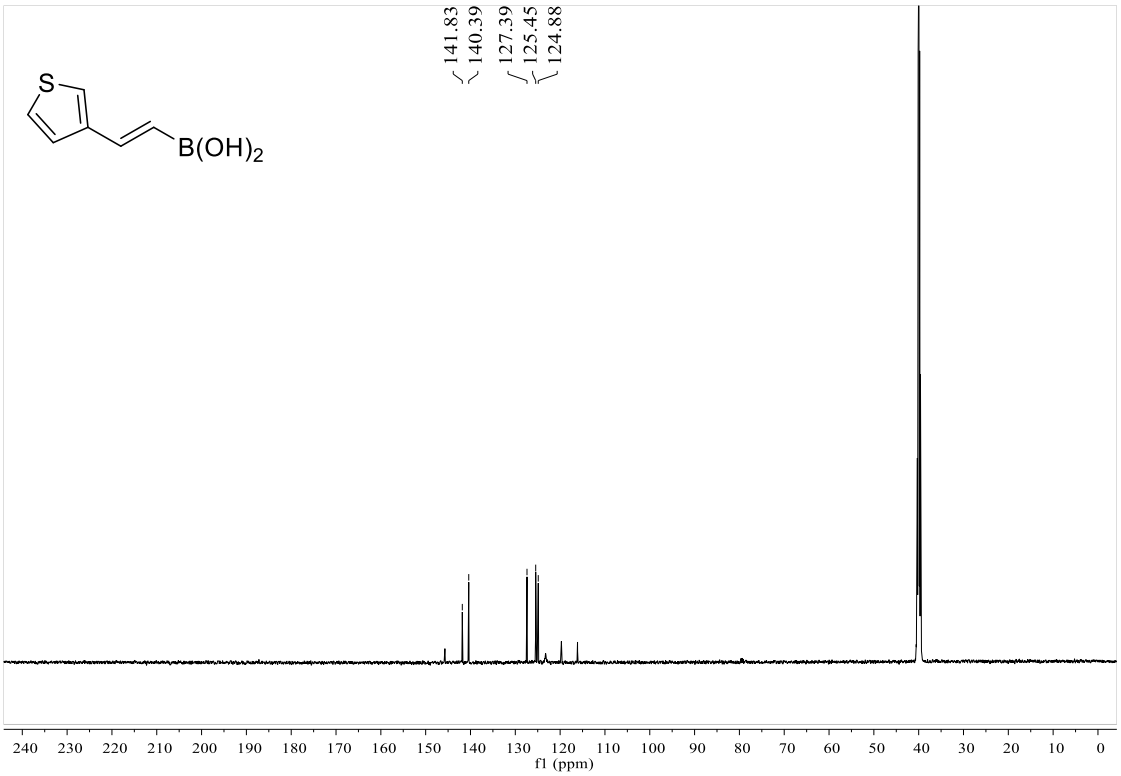


^{13}C NMR spectrum in DMSO-d_6.

(2-(thiophen-3-yl)vinyl)boronic acid (38b):



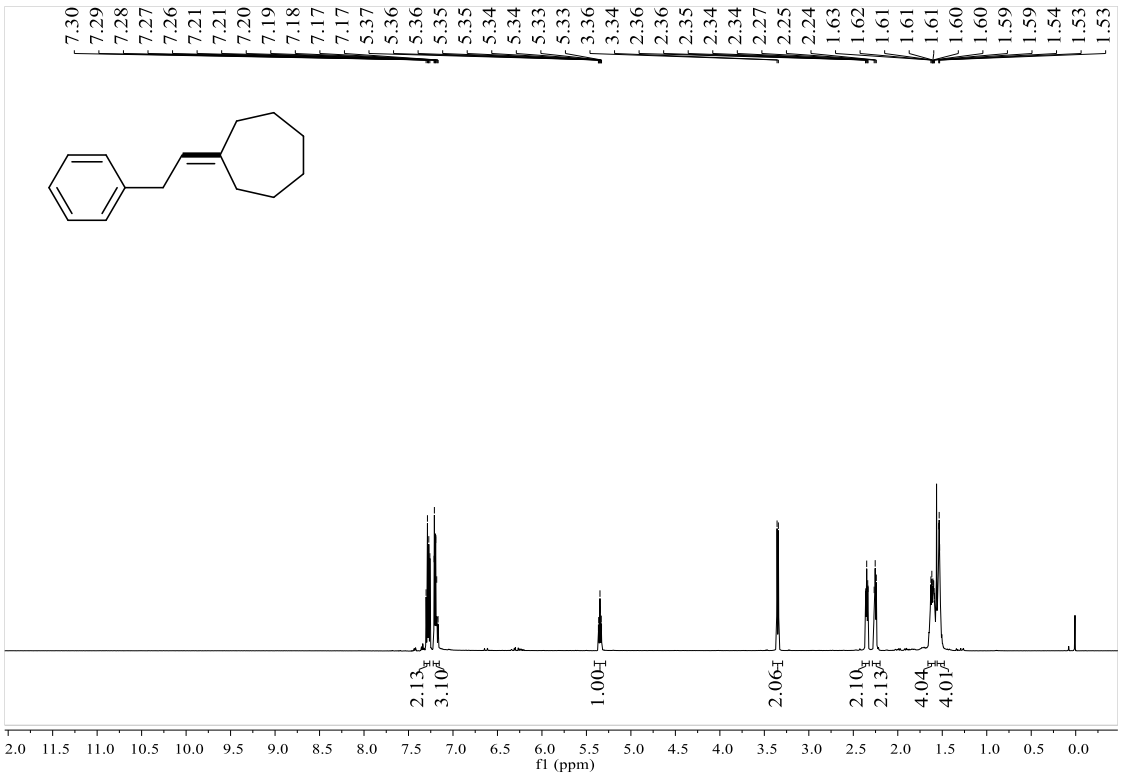
^{1}H NMR spectrum in DMSO-d_6.



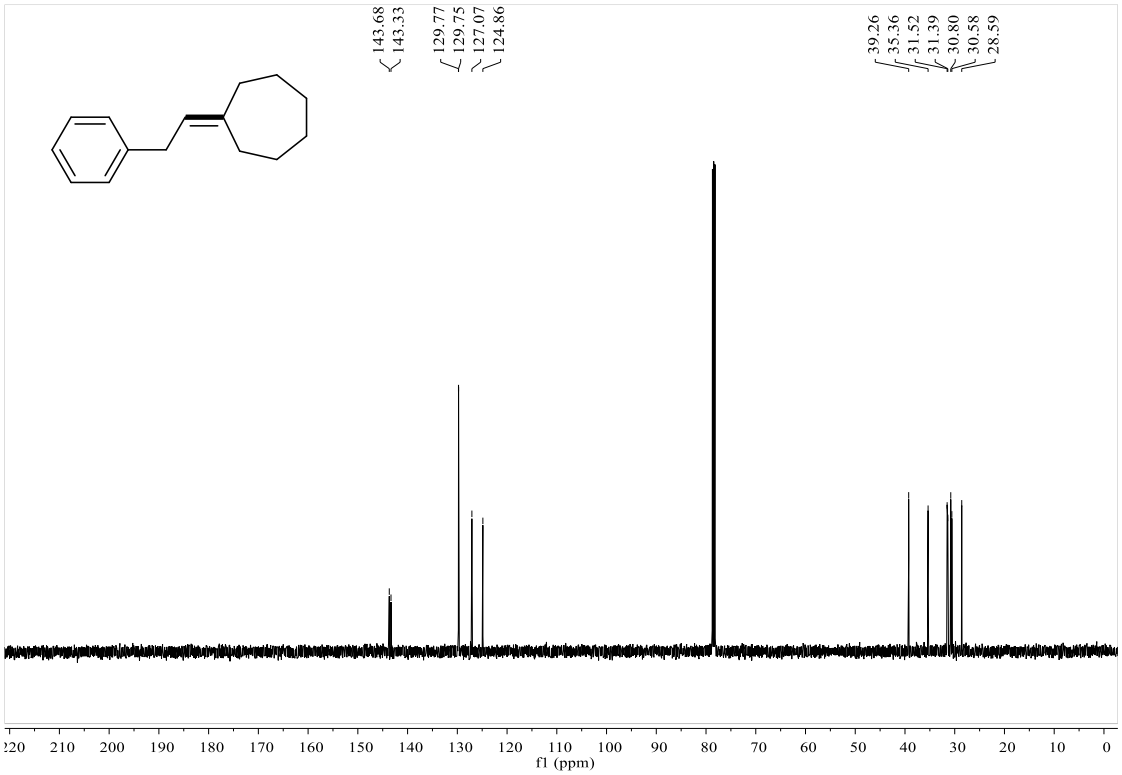
^{13}C NMR spectrum in DMSO-d_6.

9.3 NMR spectra of synthesized synthesized substrates

(2-methylcyclopropane-1,1,2-triyl)tribenzene (1c):

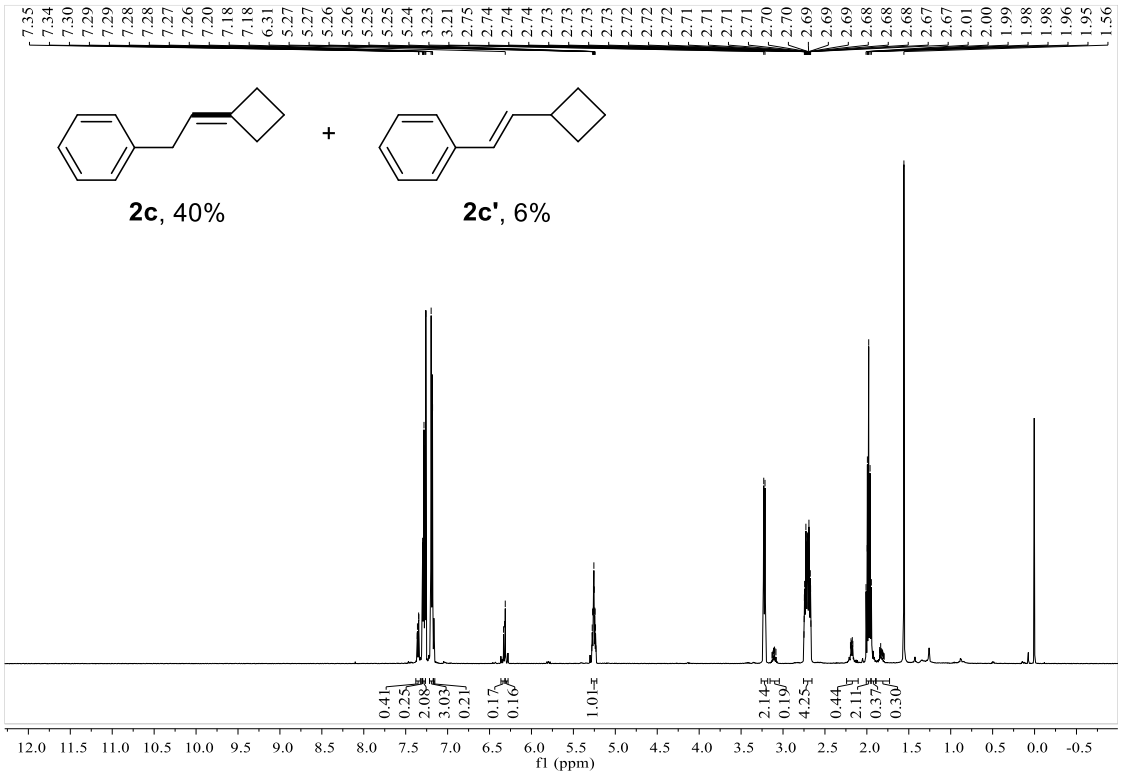


^{1}H NMR spectrum in $CDCl_3$.

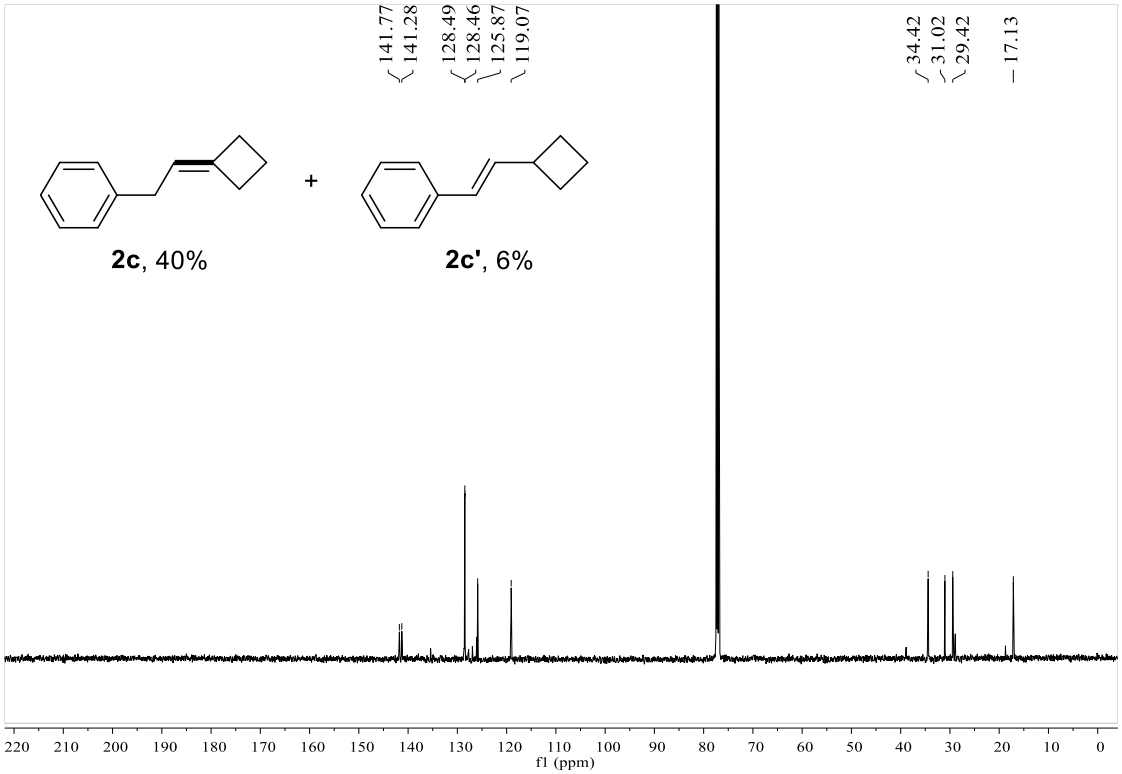


^{13}C NMR spectrum in $CDCl_3$.

(2-cyclobutylideneethyl)benzene (2c):

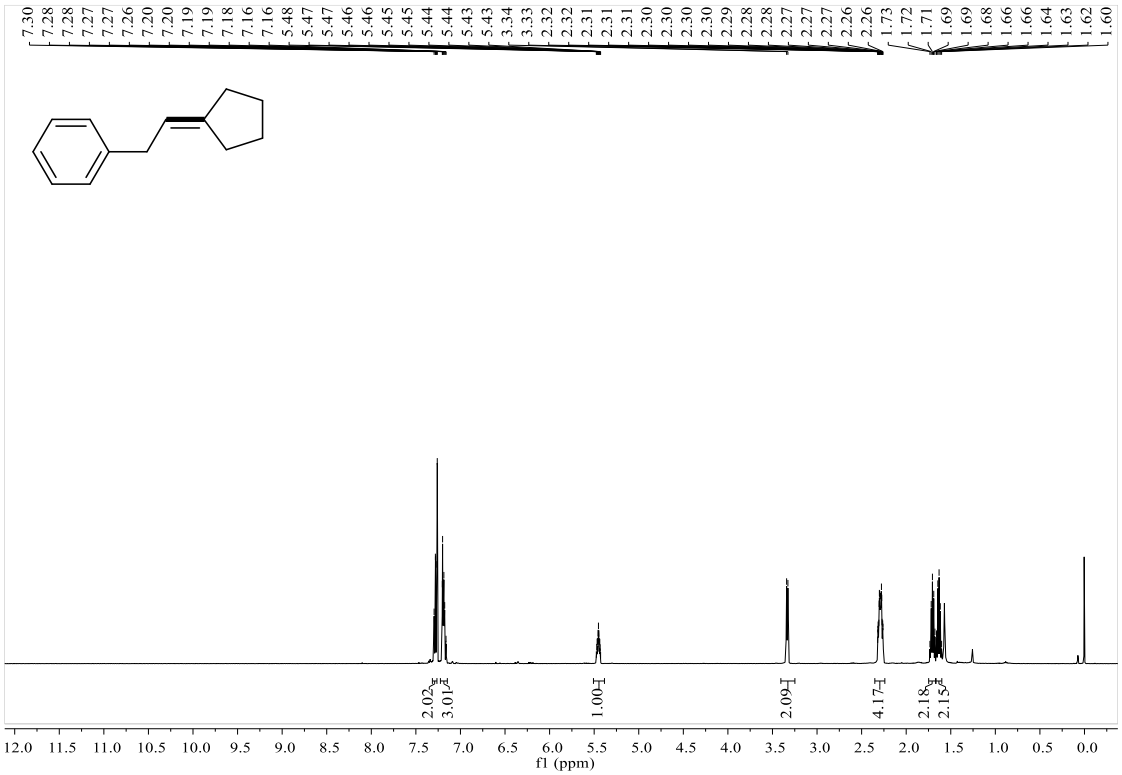


^{1}H NMR spectrum in $CDCl_3$.

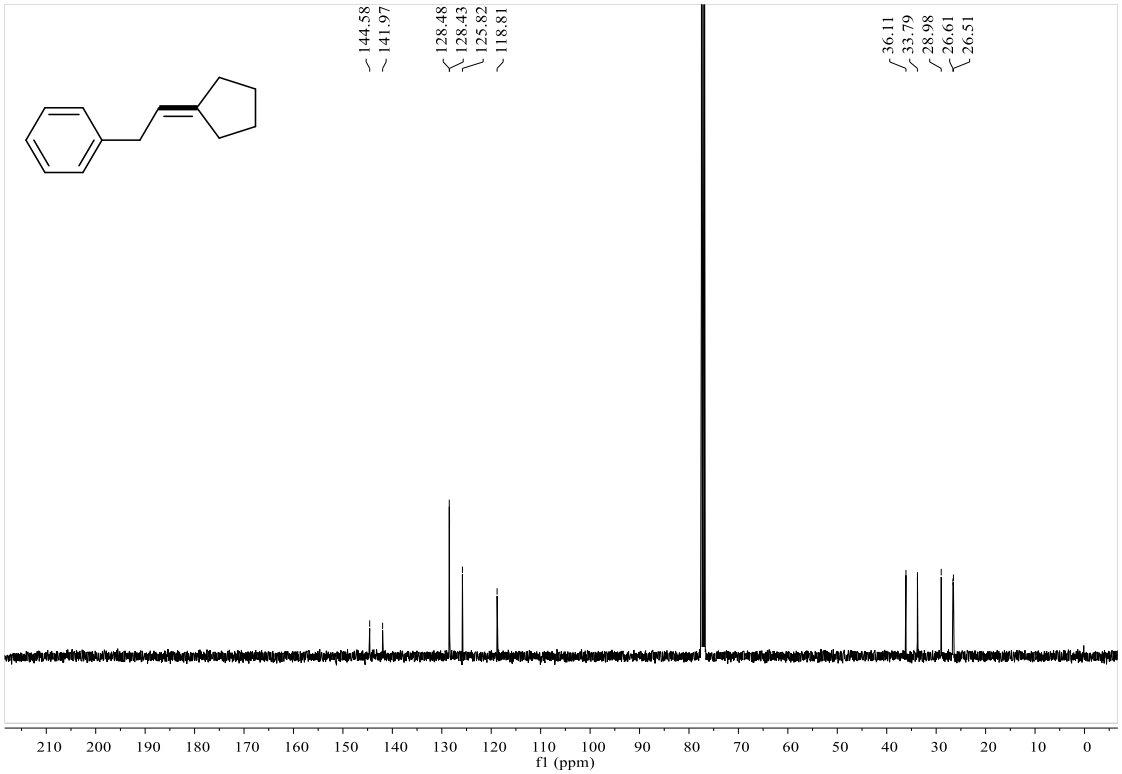


^{13}C NMR spectrum in $CDCl_3$.

(2-cyclopentylideneethyl)benzene (3c):

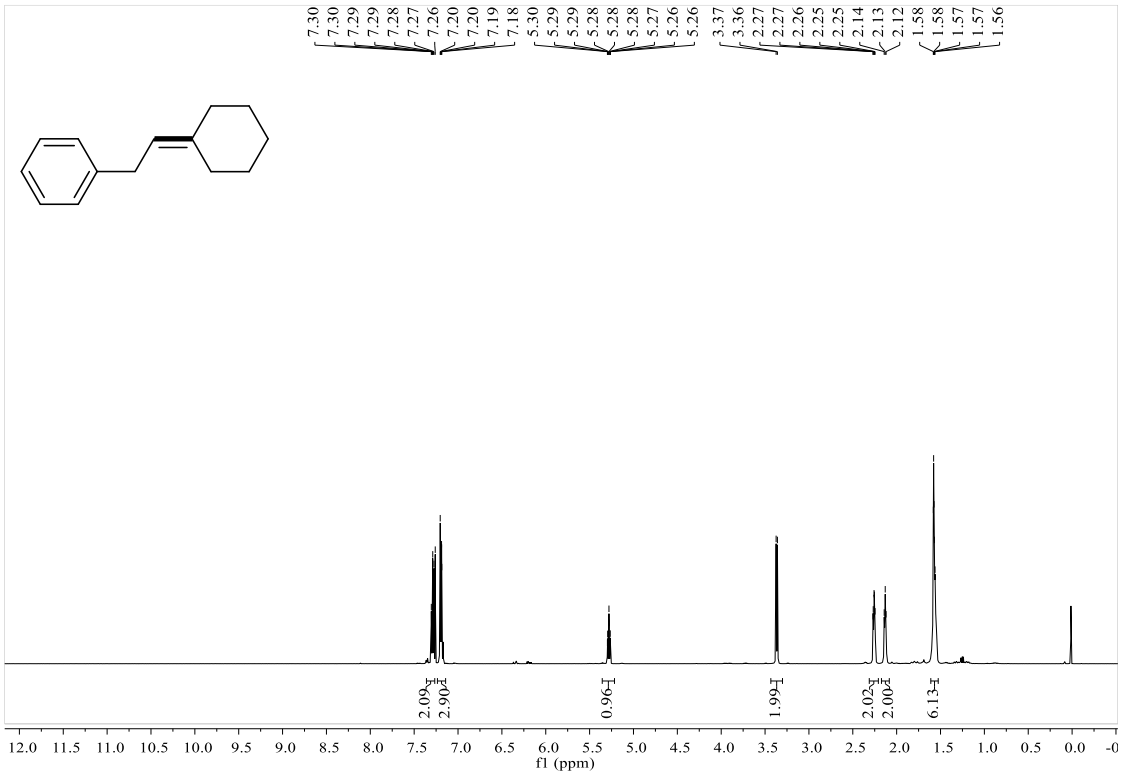


^{1}H NMR spectrum in $CDCl_3$.

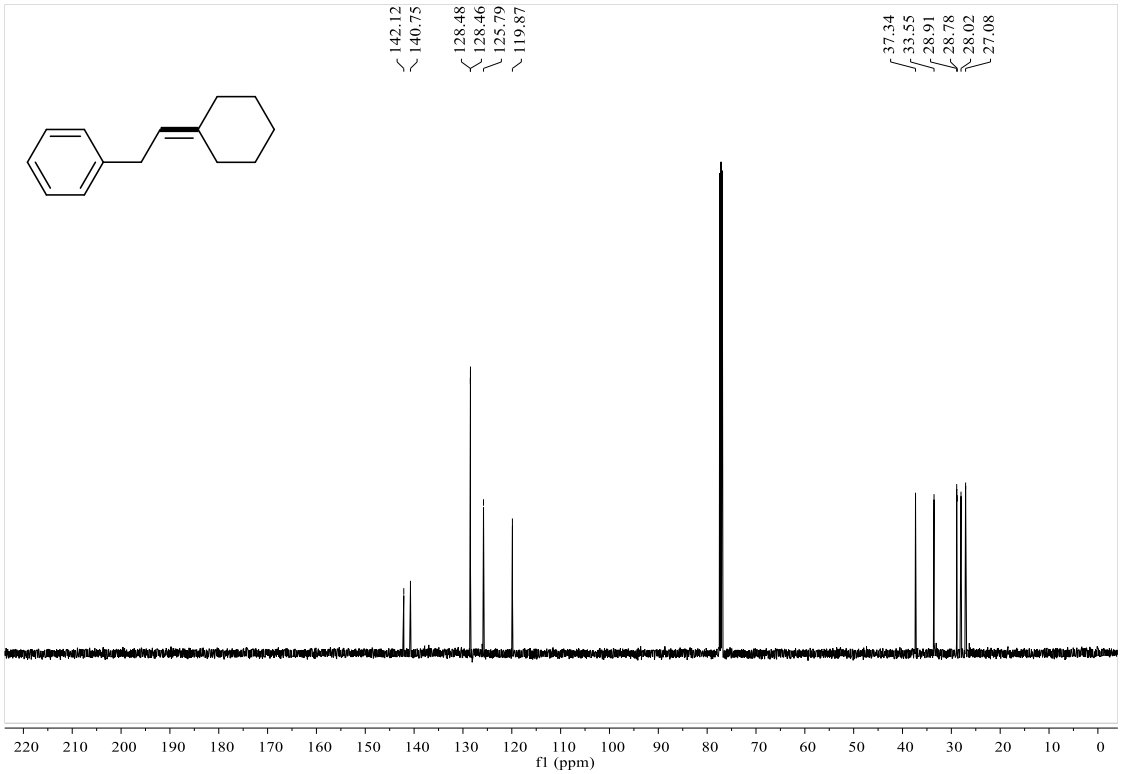


^{13}C NMR spectrum in $CDCl_3$.

(2-cyclohexylideneethyl)benzene (4c):

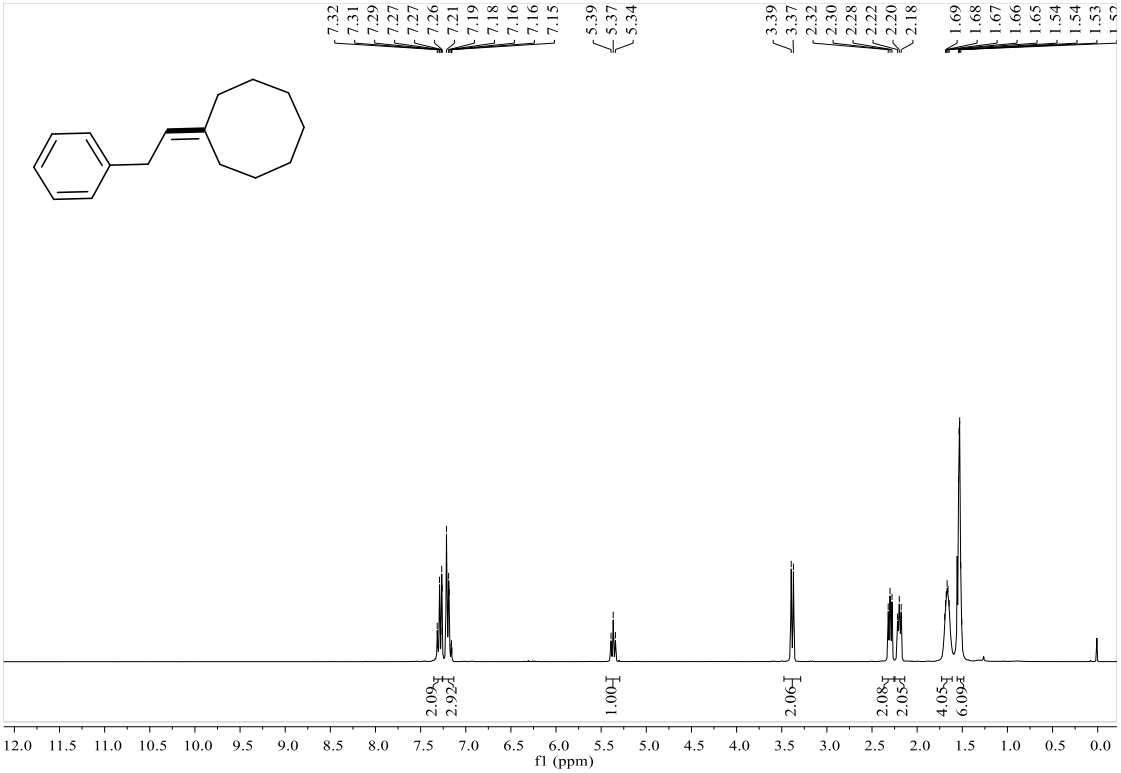


^{1}H NMR spectrum in $CDCl_3$.

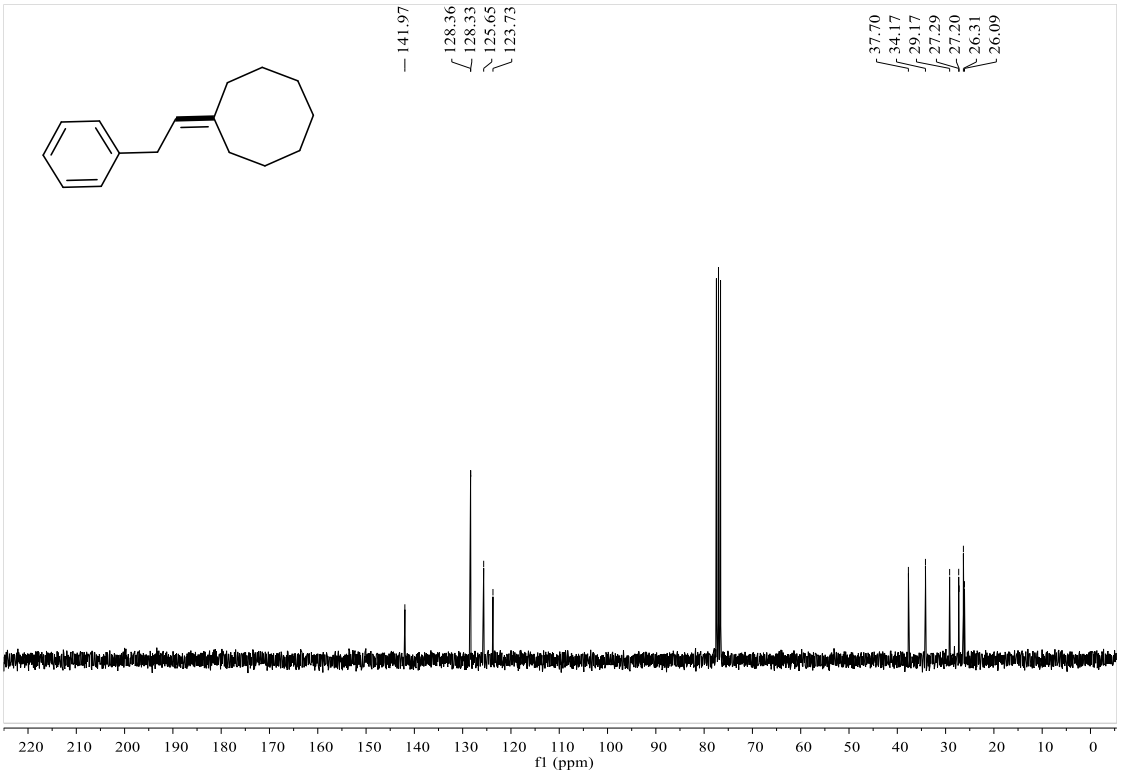


^{13}C NMR spectrum in $CDCl_3$.

(2-phenylethylidene)cyclooctane (5c):

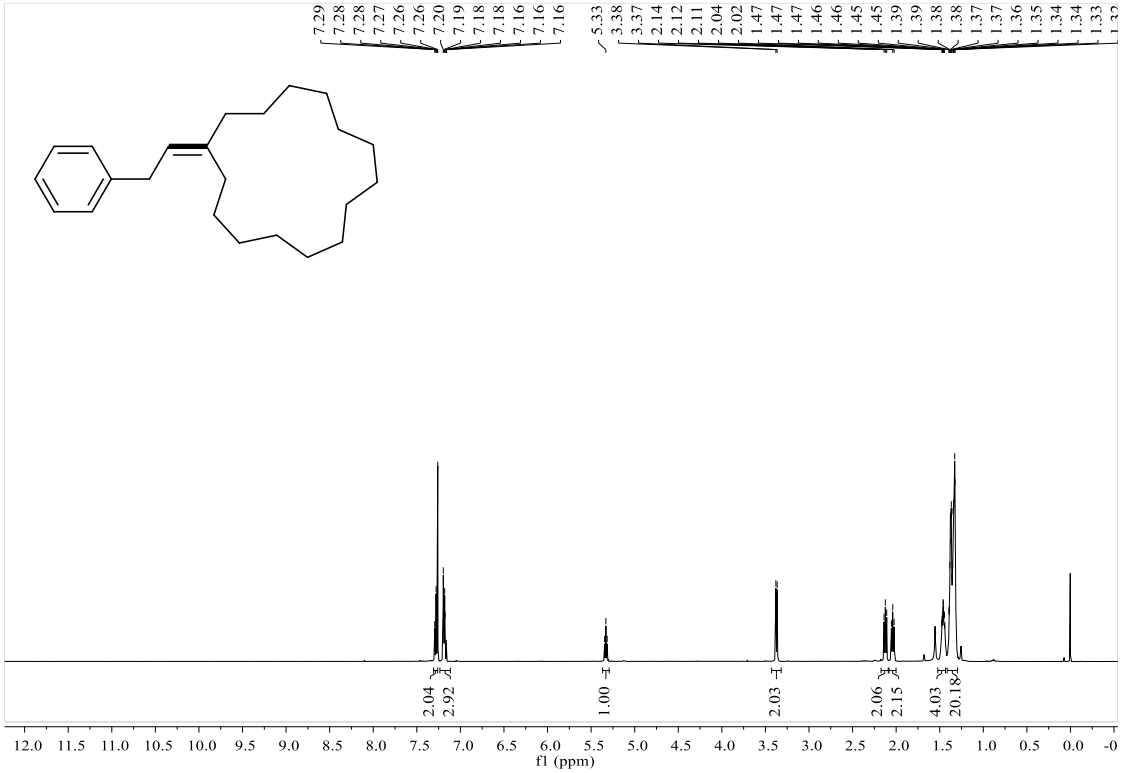


^{1}H NMR spectrum in $CDCl_3$.

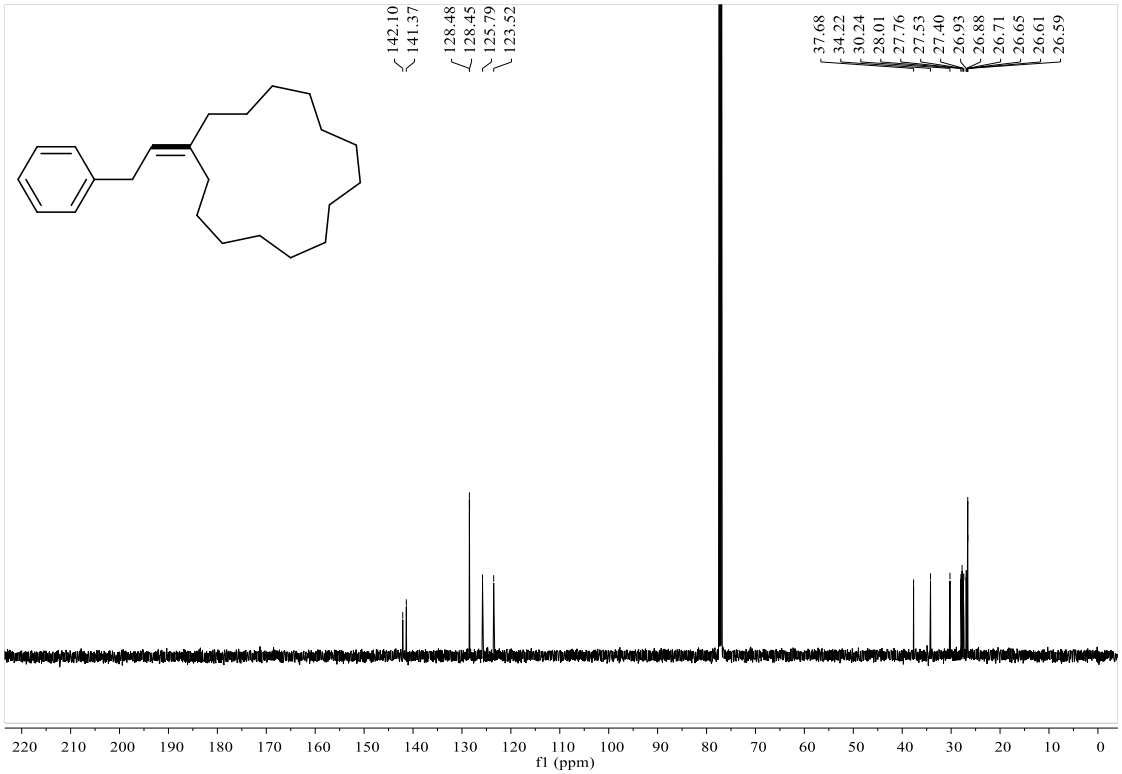


^{13}C NMR spectrum in $CDCl_3$.

(2-phenylethylidene)cyclopentadecane (6c):

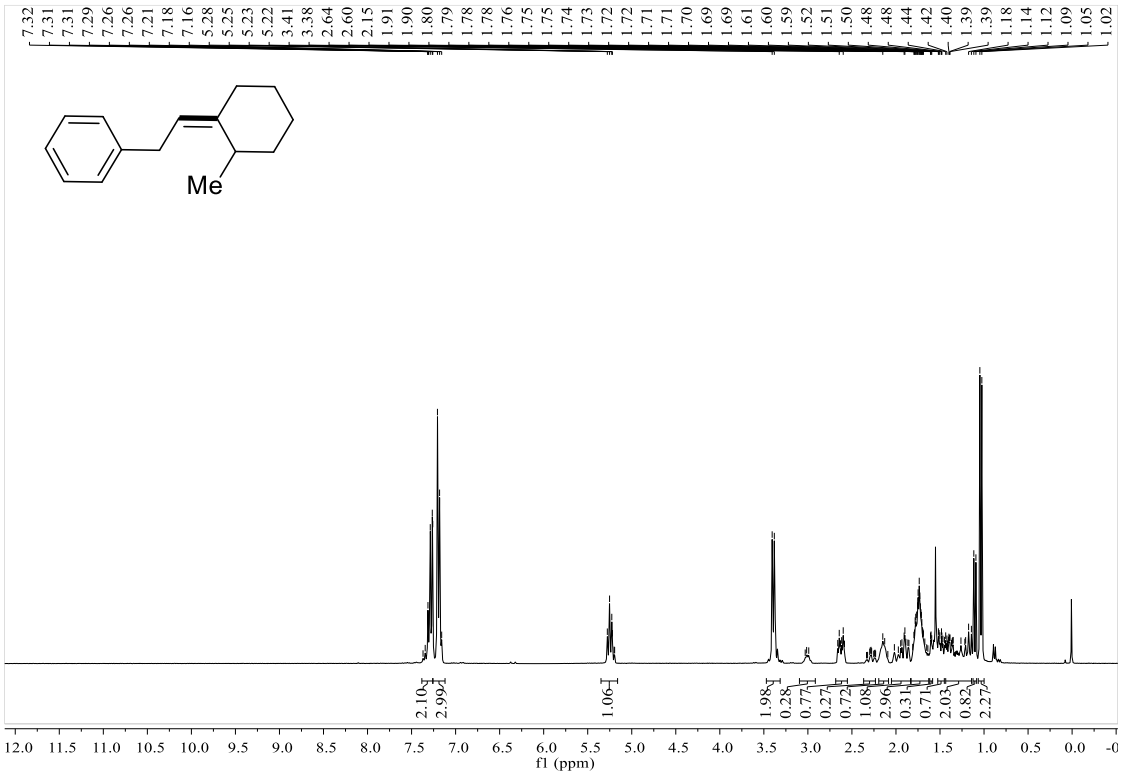


^{1}H NMR spectrum in $CDCl_3$.

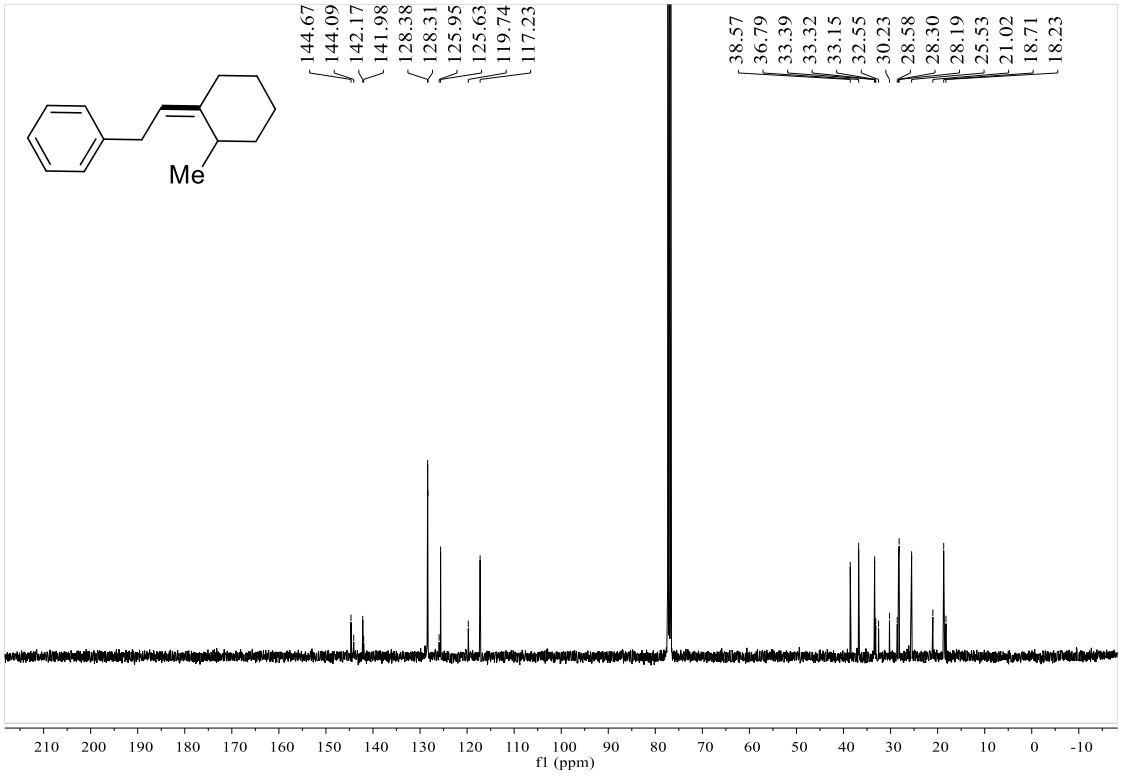


^{13}C NMR spectrum in $CDCl_3$.

(2-(2-methylcyclohexylidene)ethyl)benzene (7c):

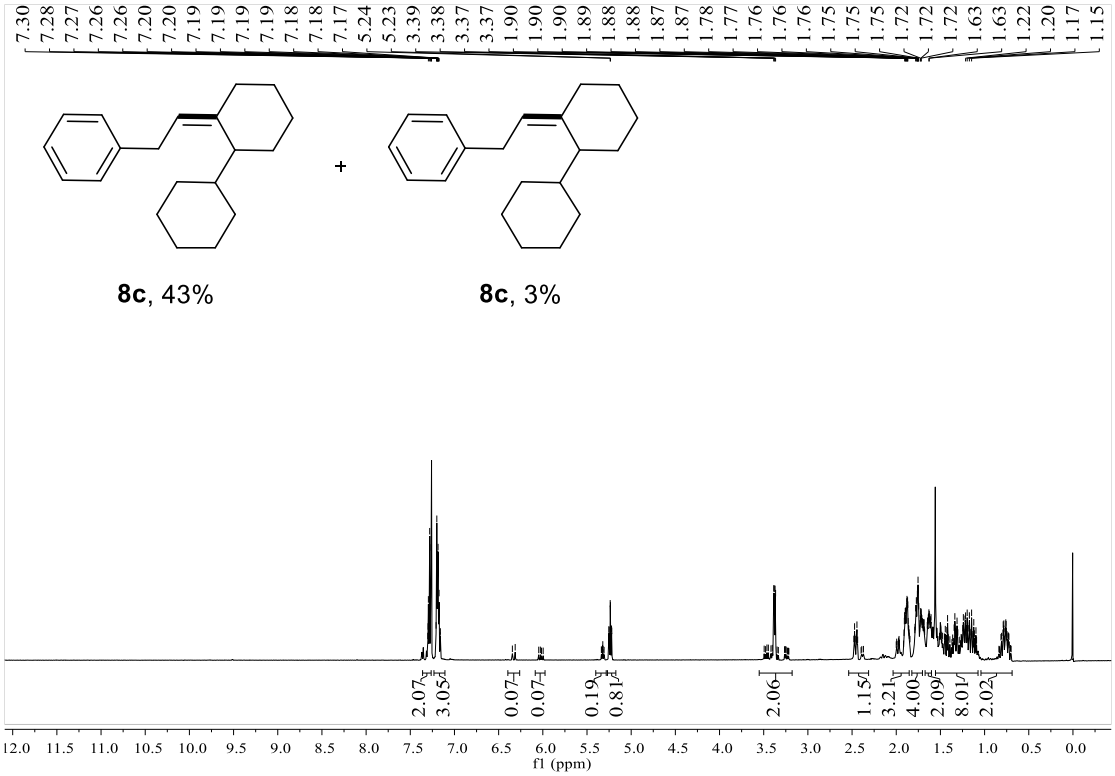


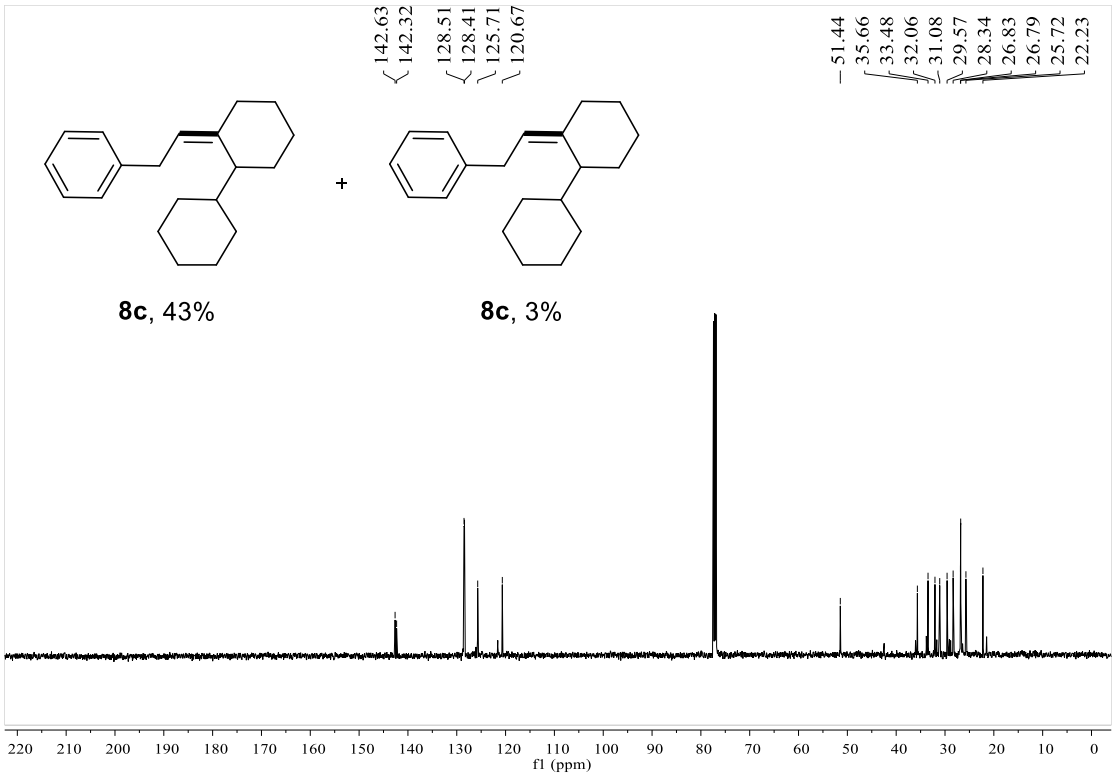
^{1}H NMR spectrum in $CDCl_3$.



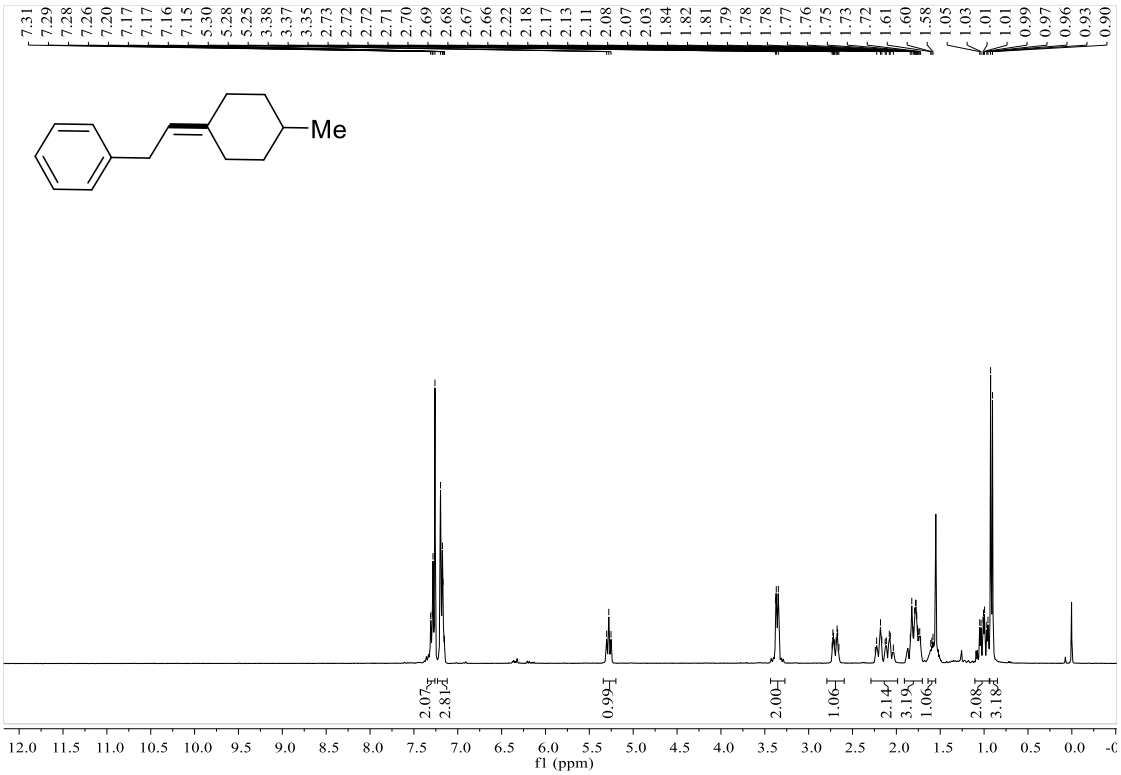
^{13}C NMR spectrum in $CDCl_3$.

2-(2-phenylethylidene)-1,1'-bi(cyclohexane) (8c):

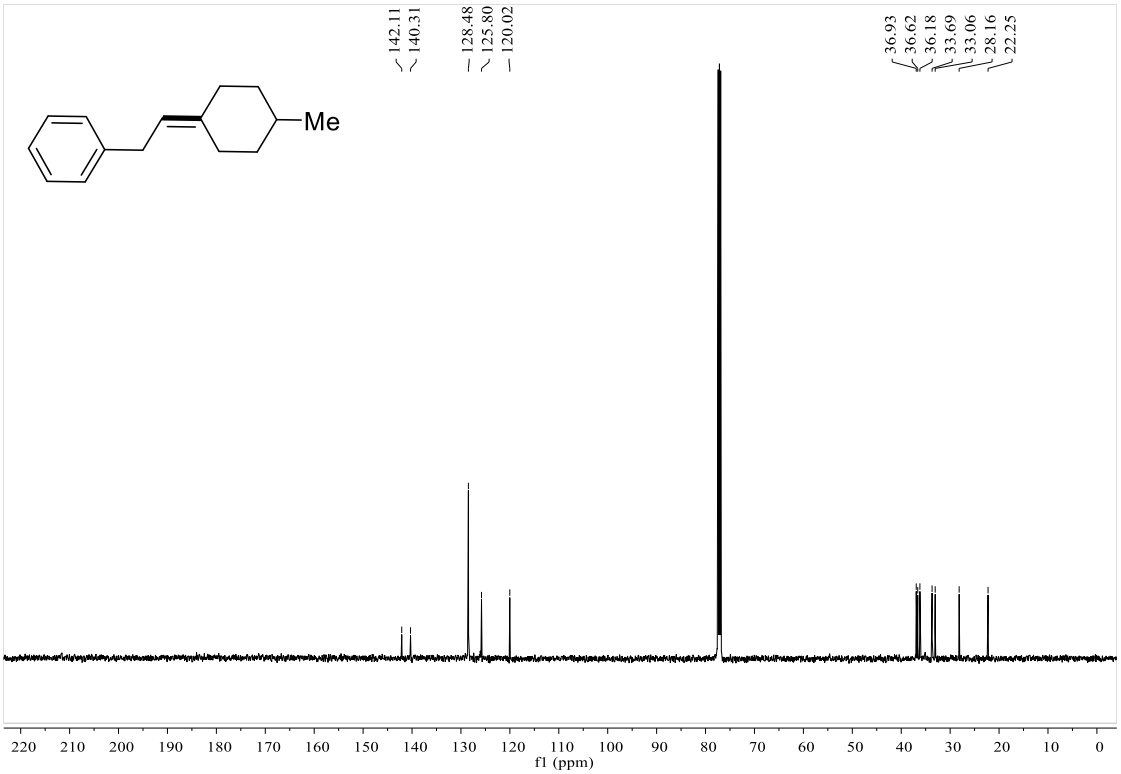




(2-(4-methylcyclohexylidene)ethyl)benzene (9c):

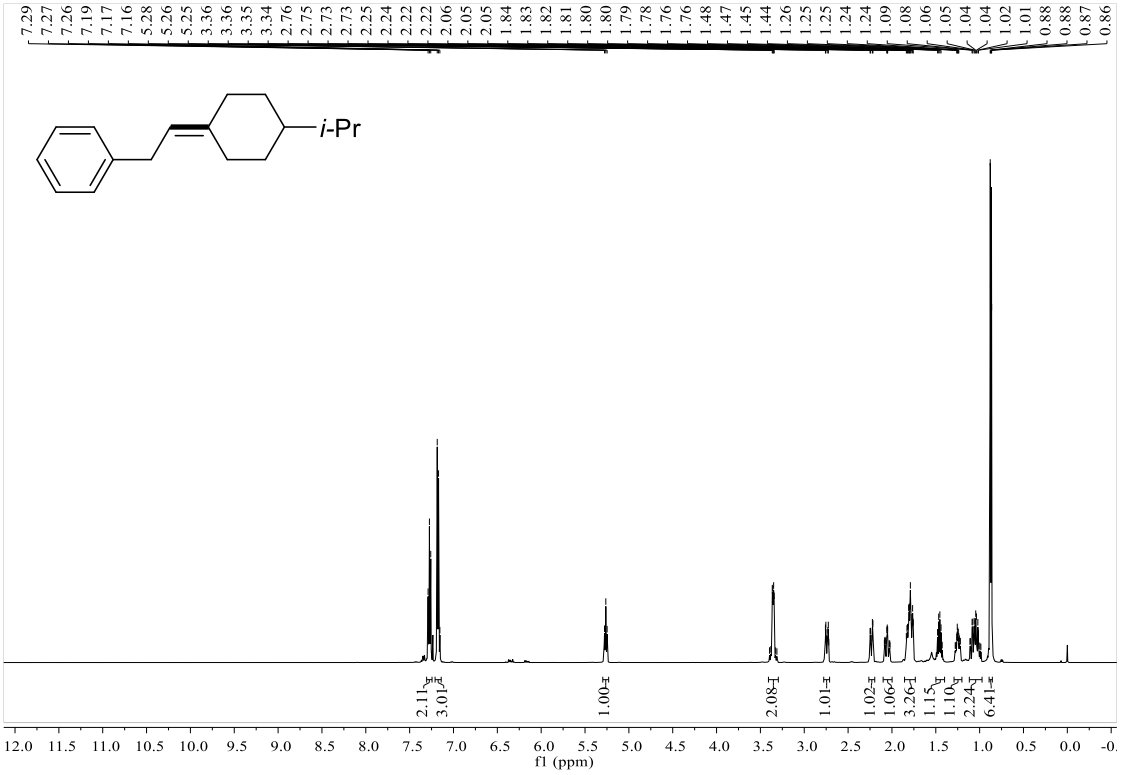


^{1}H NMR spectrum in $CDCl_3$.

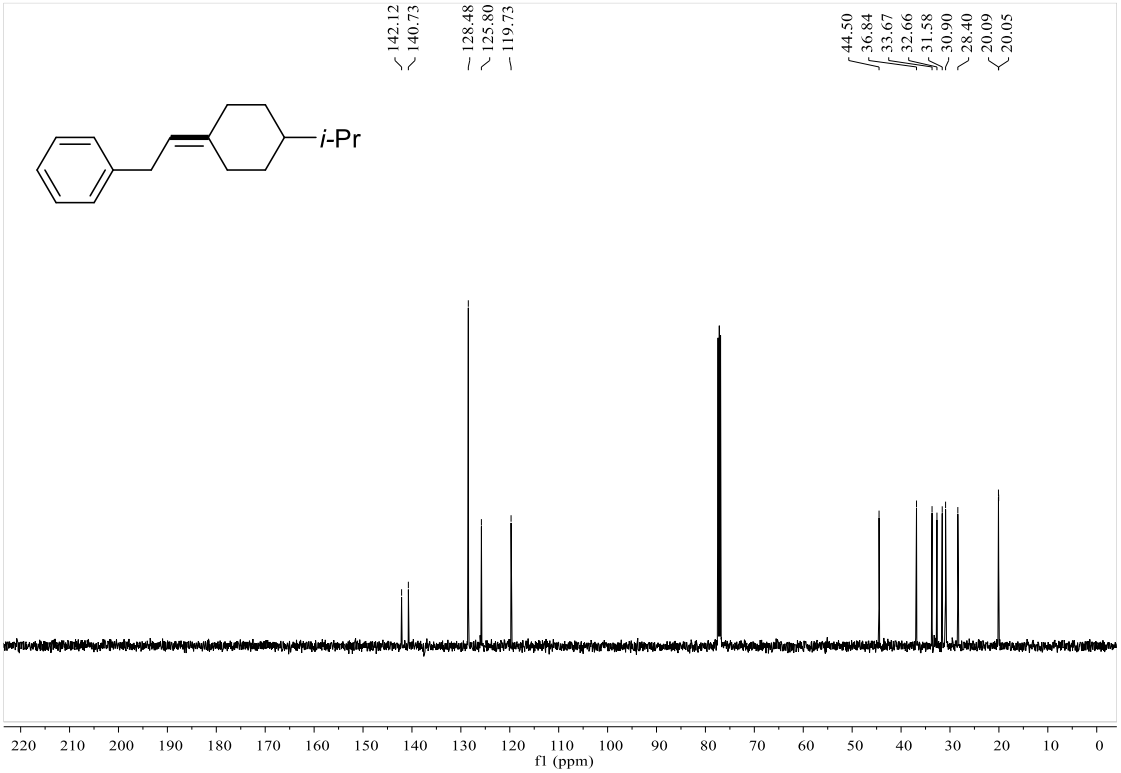


^{13}C NMR spectrum in $CDCl_3$.

(2-(4-isopropylcyclohexylidene)ethyl)benzene (10c):

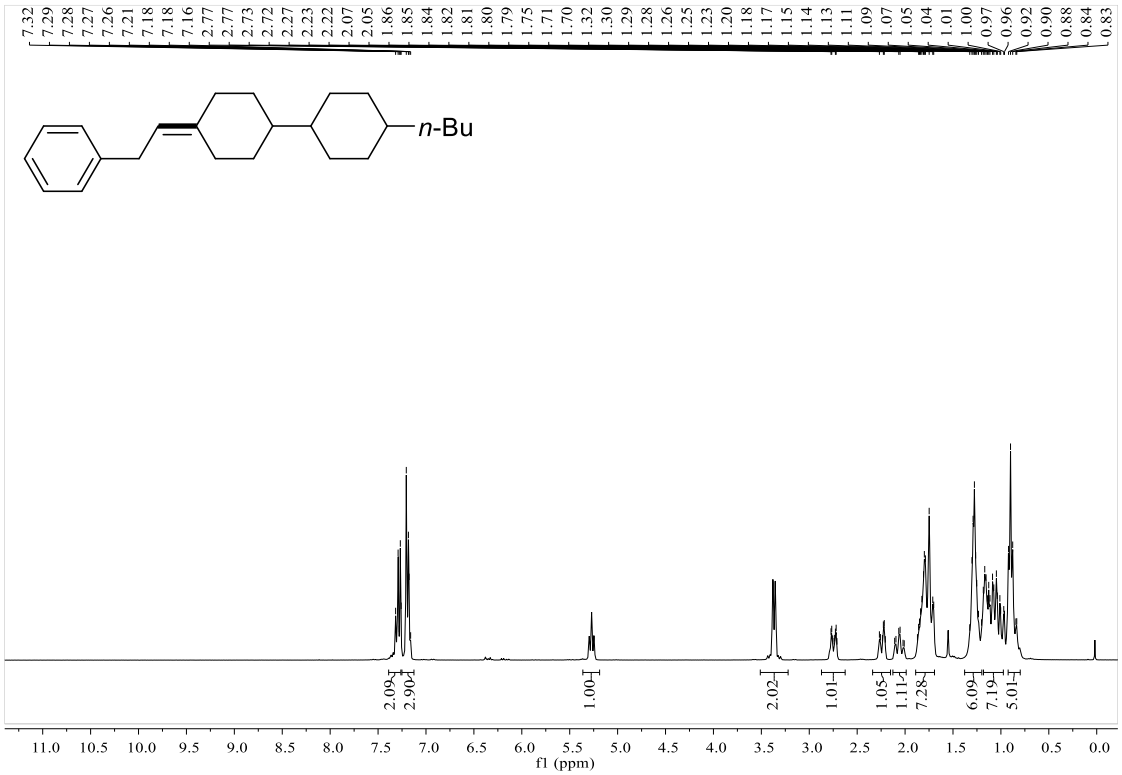


^{1}H NMR spectrum in $CDCl_3$.

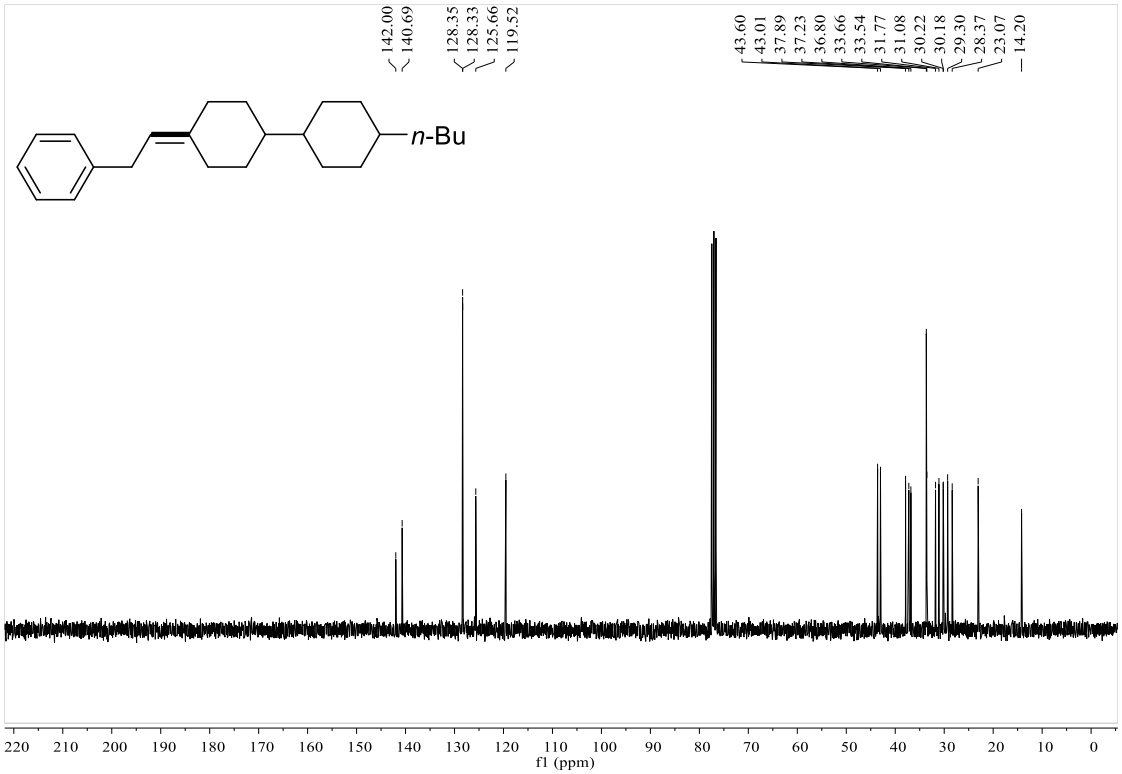


^{13}C NMR spectrum in $CDCl_3$.

4-butyl-4'-(2-phenylethylidene)-1,1'-bi(cyclohexane) (11c):

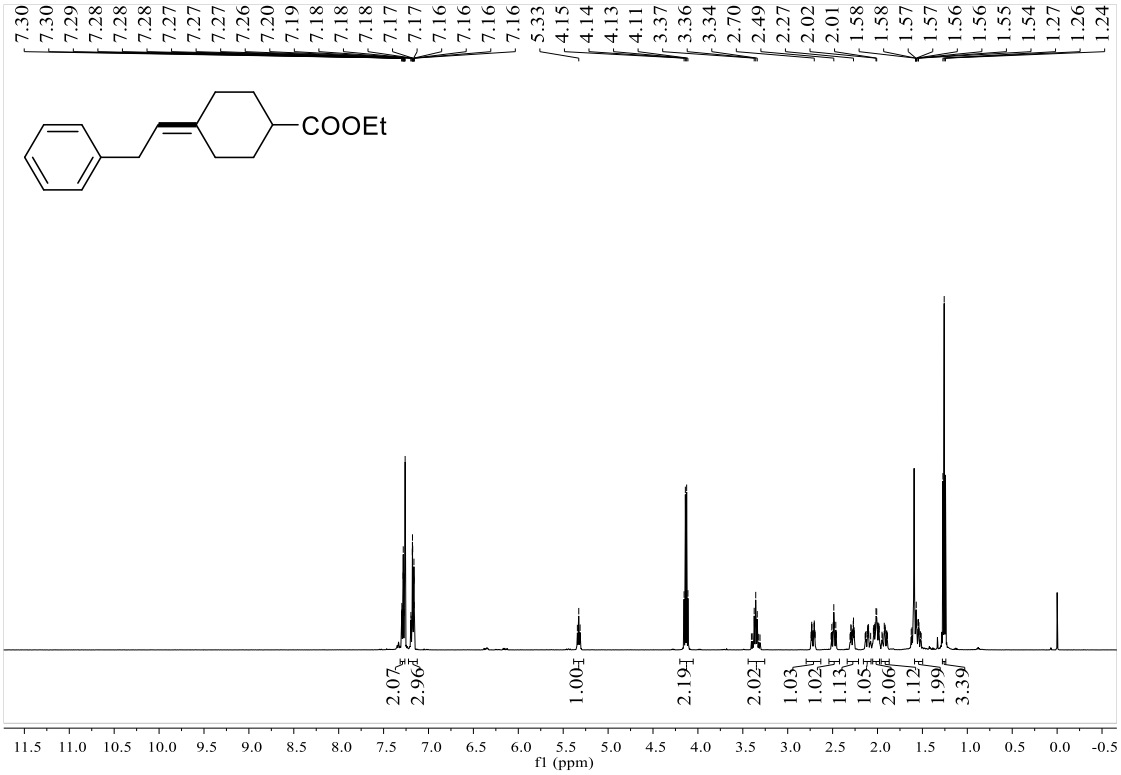


^{1}H NMR spectrum in $CDCl_3$.

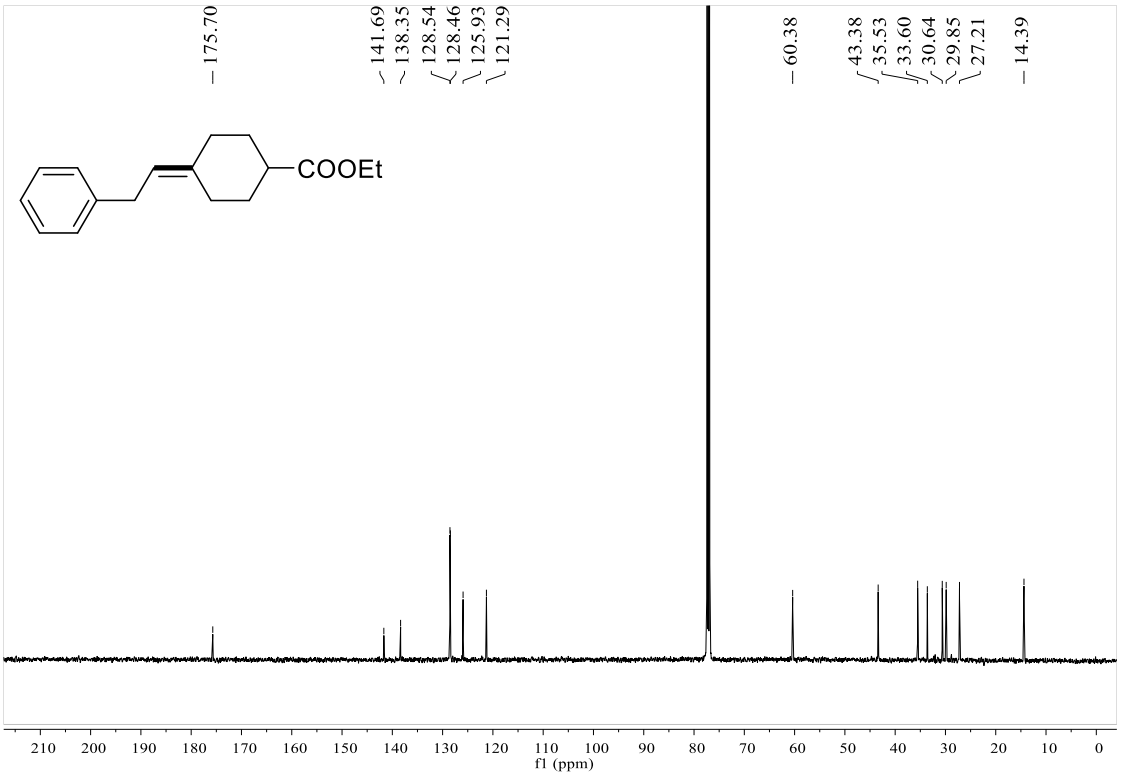


^{13}C NMR spectrum in $CDCl_3$.

ethyl 4-(2-phenylethylidene)cyclohexane-1-carboxylate (12c):

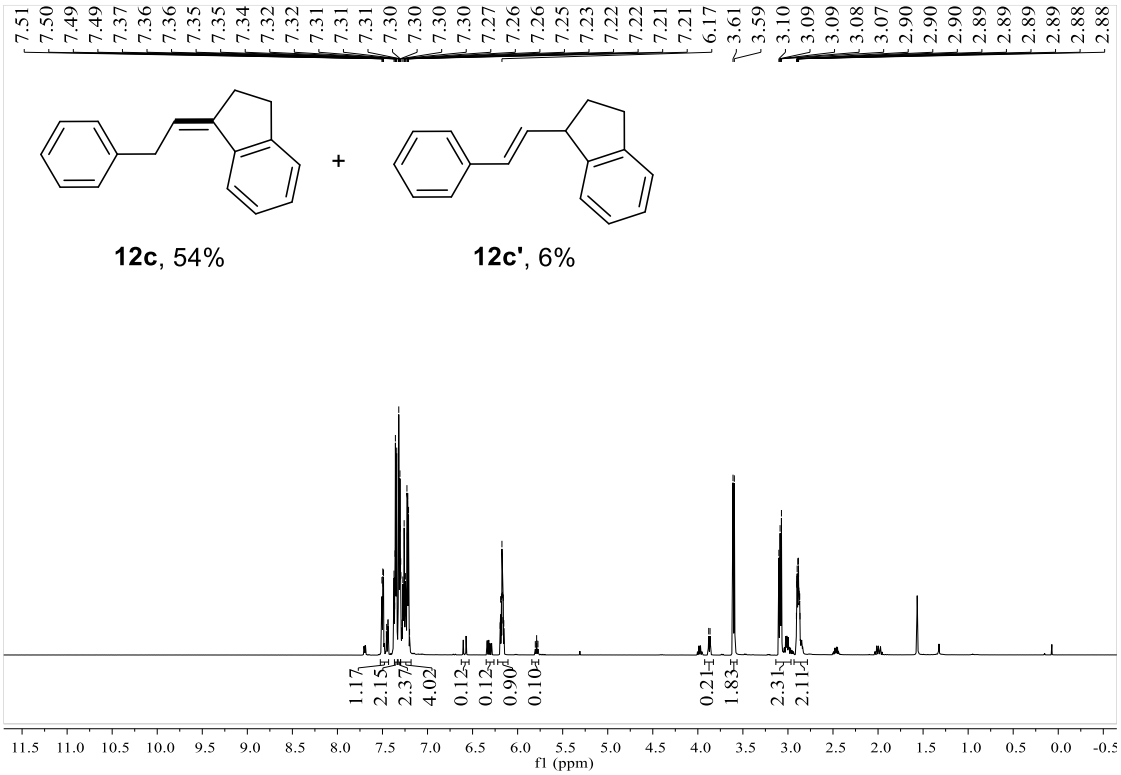


^{1}H NMR spectrum in $CDCl_3$.

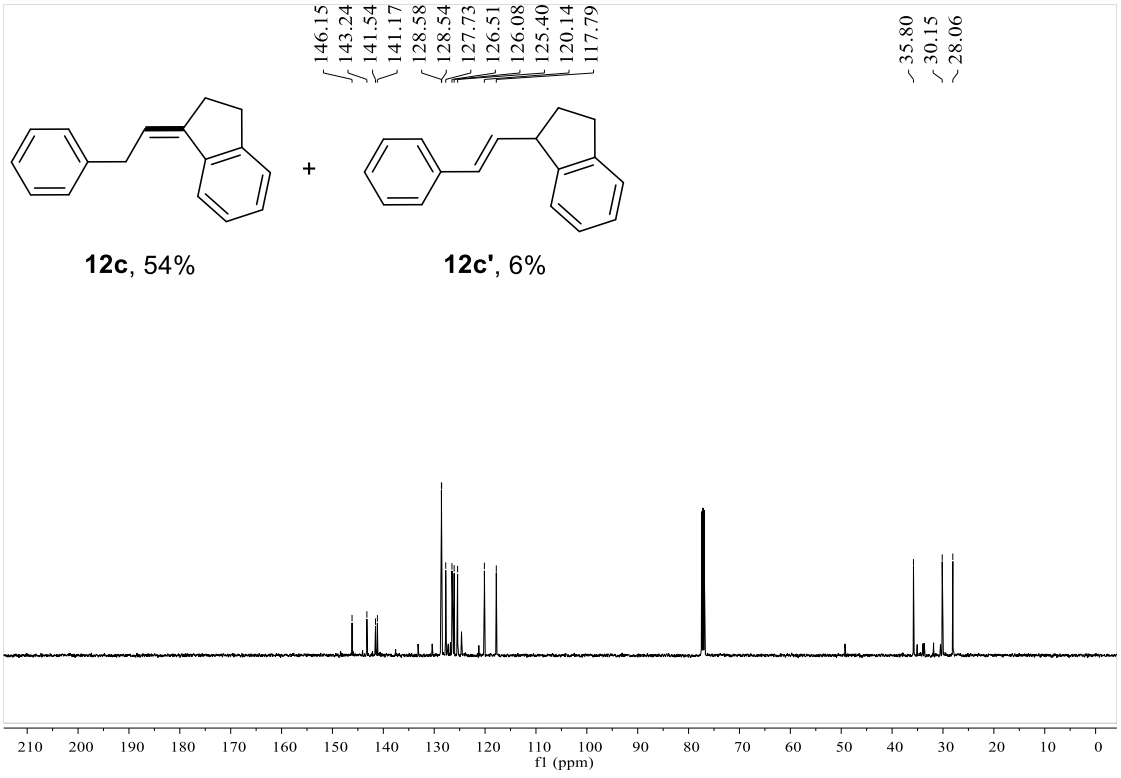


^{13}C NMR spectrum in $CDCl_3$.

1-(2-phenylethylidene)-2,3-dihydro-1H-indene (13c):

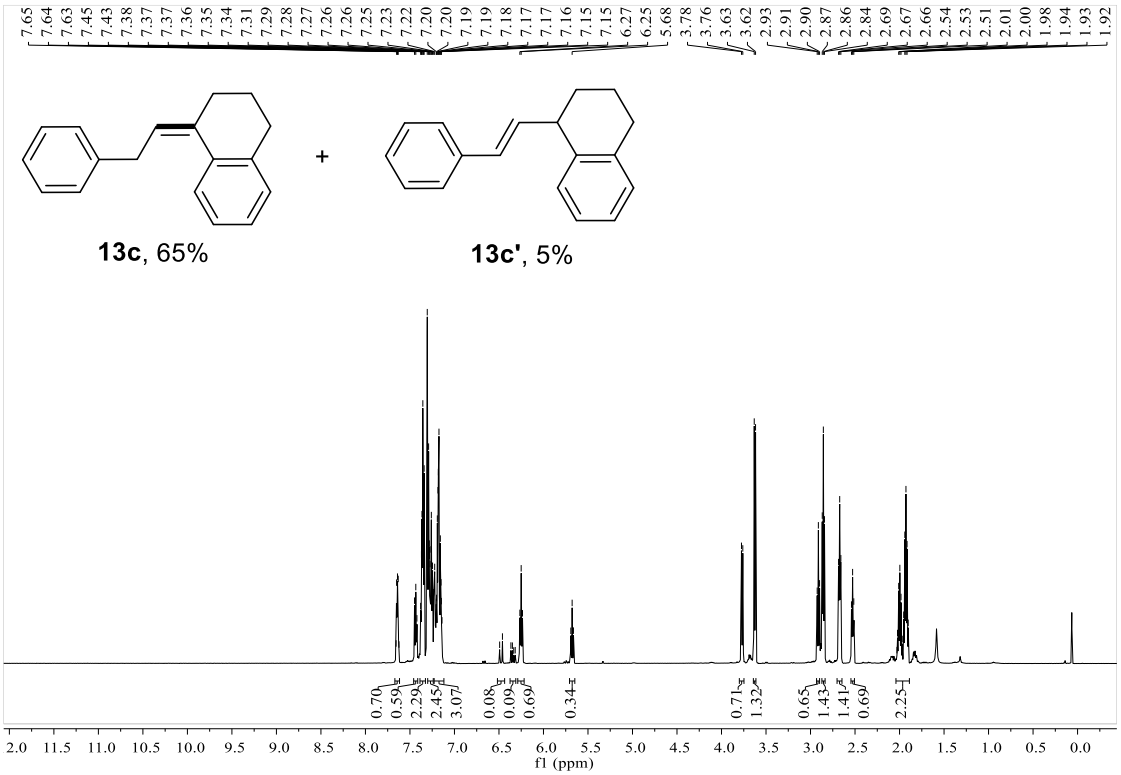


^{1}H NMR spectrum in $CDCl_3$.

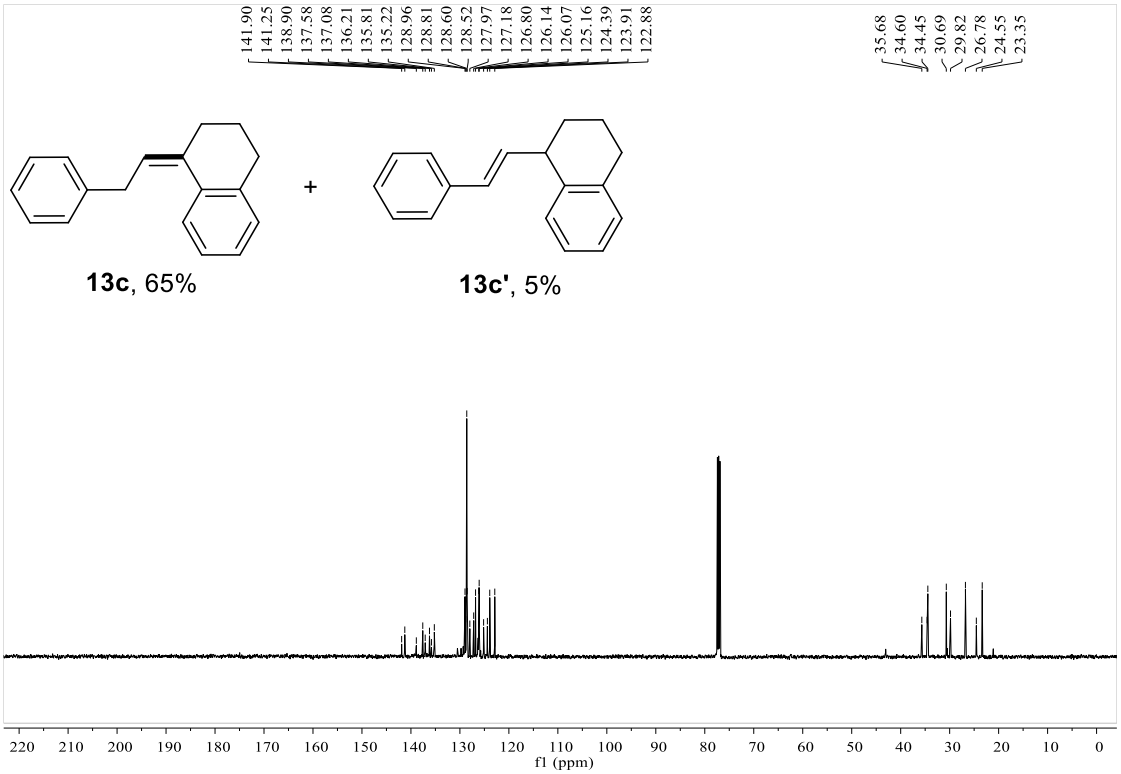


^{13}C NMR spectrum in $CDCl_3$.

1-(2-phenylethylidene)-1,2,3,4-tetrahydronaphthalene (14c):

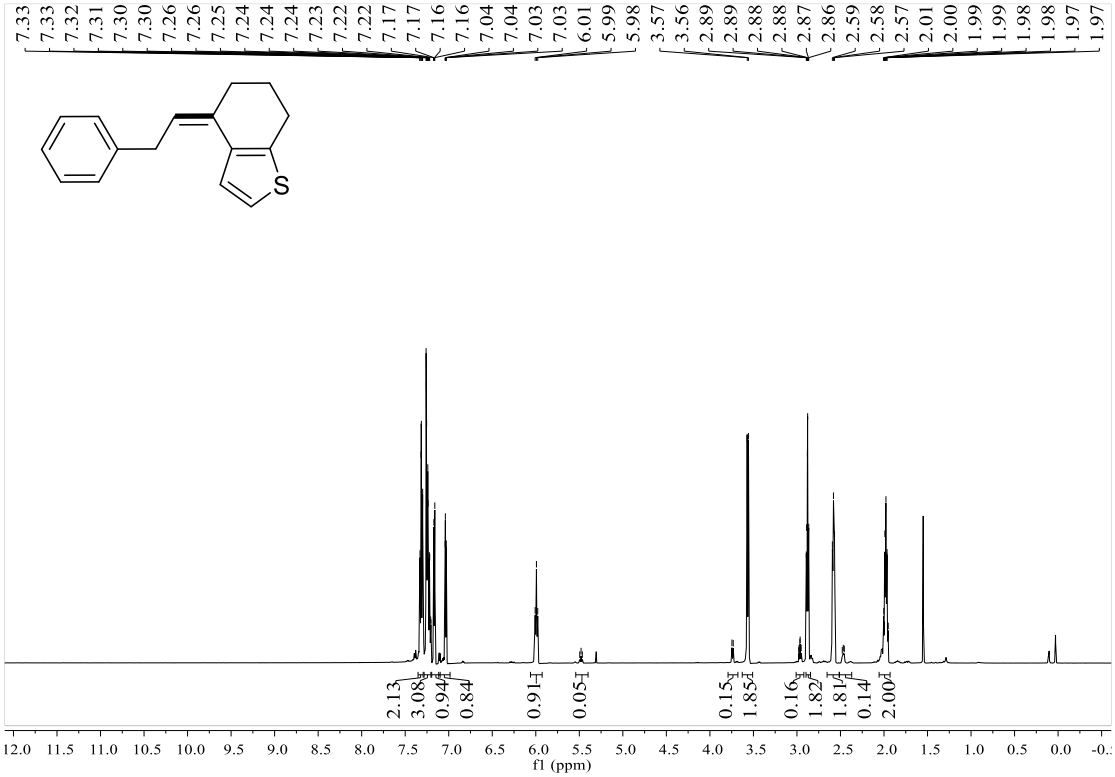


^{1}H NMR spectrum in $CDCl_3$.

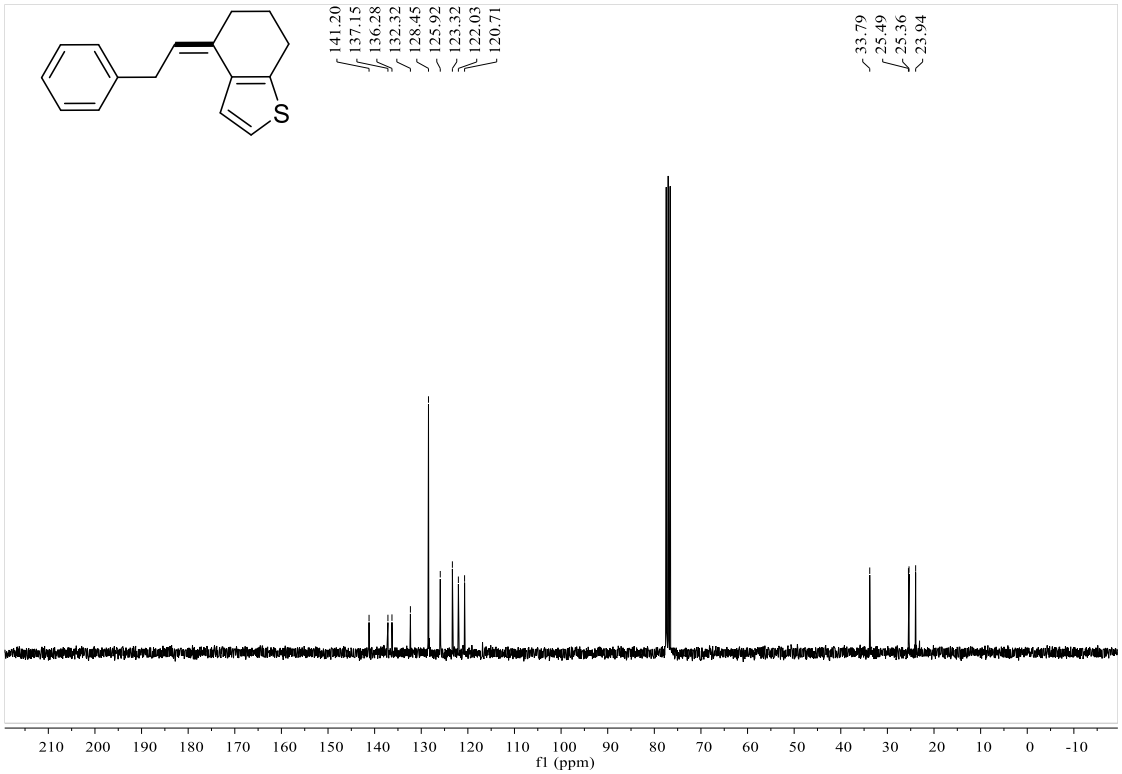


^{13}C NMR spectrum in $CDCl_3$.

1-(2-phenylethylidene)-1,2,3,4-tetrahydronaphthalene (15c):

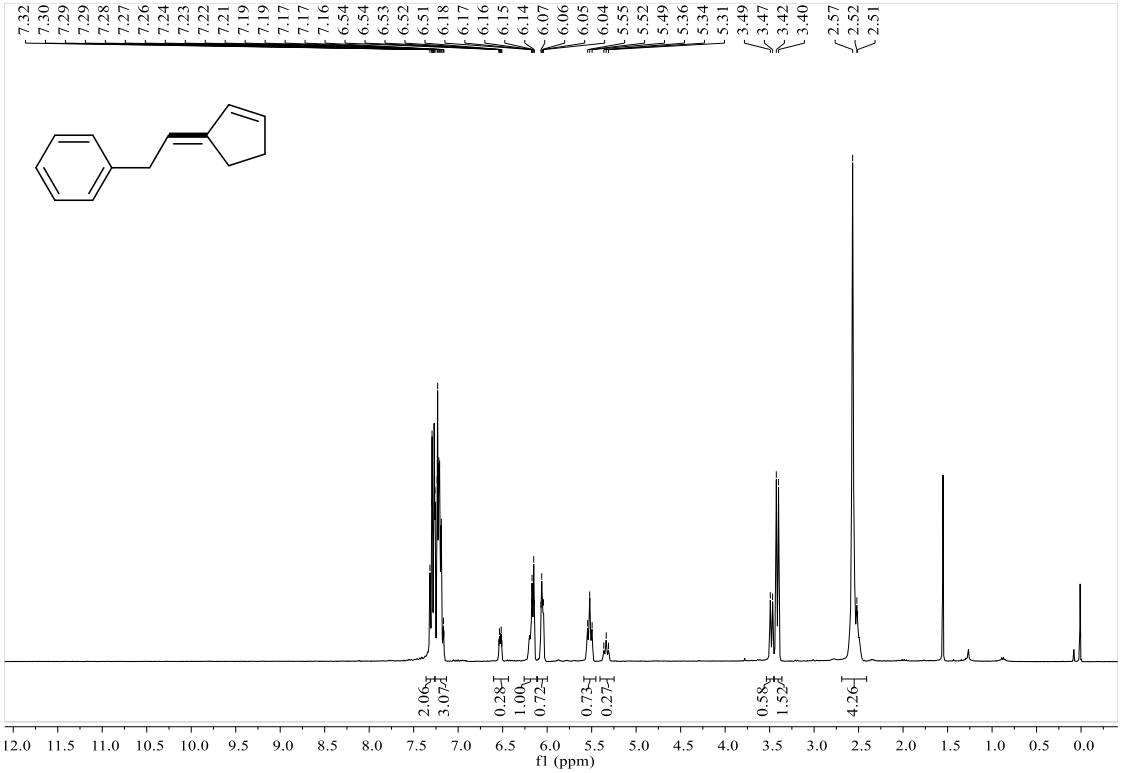


^{1}H NMR spectrum in $CDCl_3$.

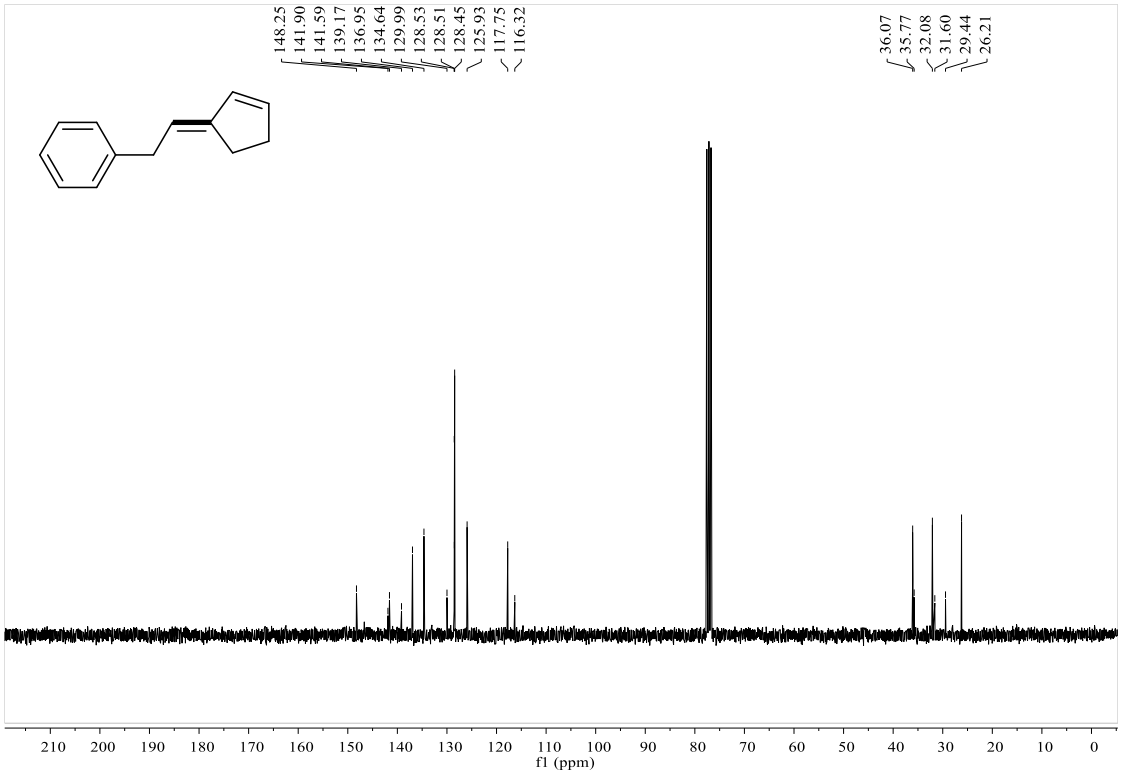


^{13}C NMR spectrum in $CDCl_3$.

(2-(cyclopent-2-en-1-ylidene)ethyl)benzene (16c):

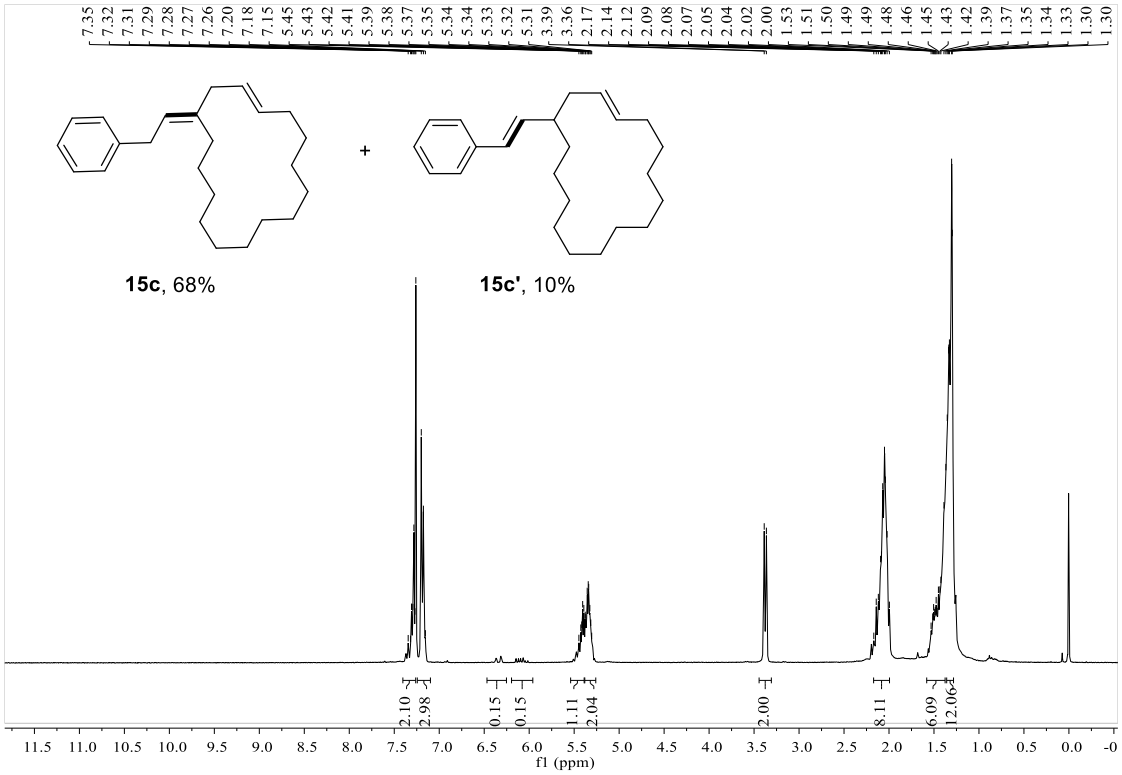


^{1}H NMR spectrum in $CDCl_3$.

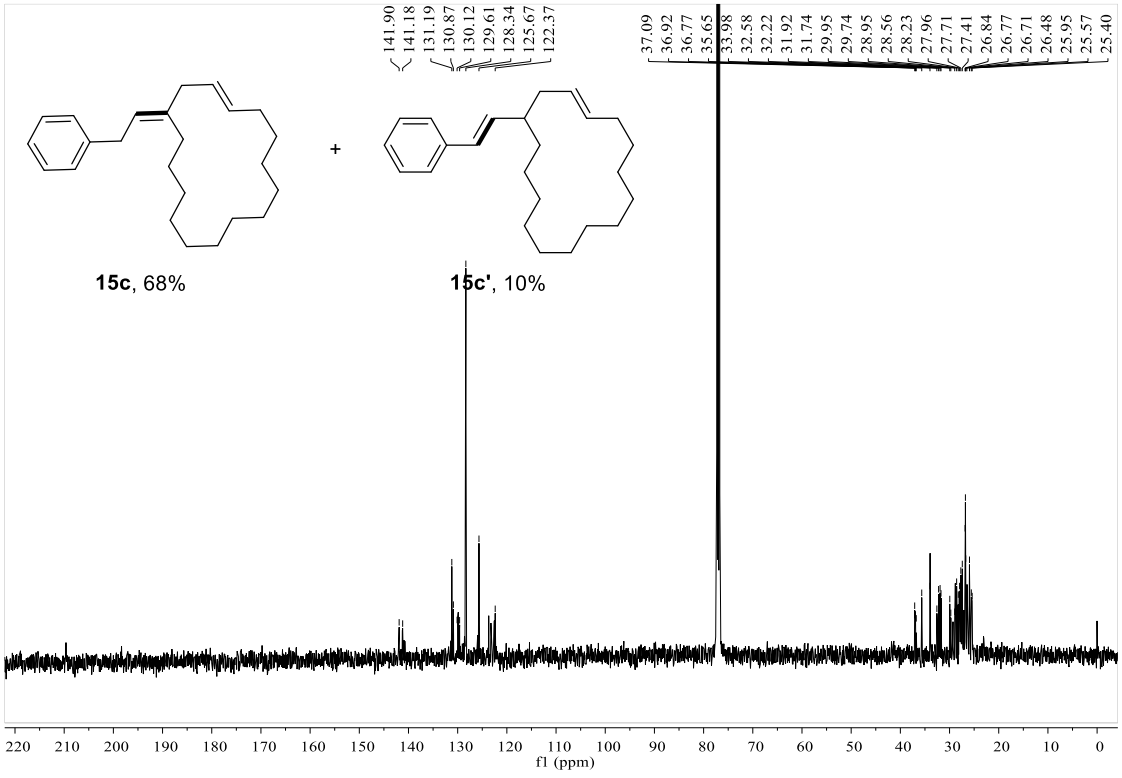


^{13}C NMR spectrum in $CDCl_3$.

(2-(cyclopent-2-en-1-ylidene)ethyl)benzene (17c):

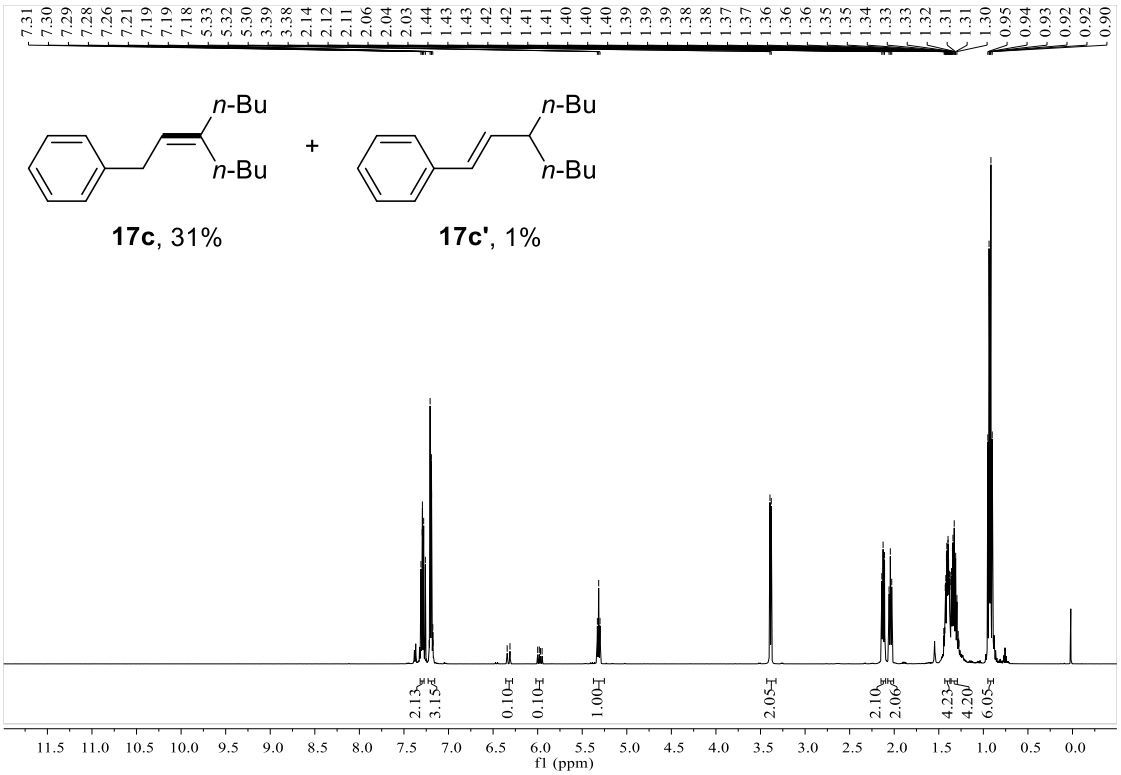


^{1}H NMR spectrum in $CDCl_3$.

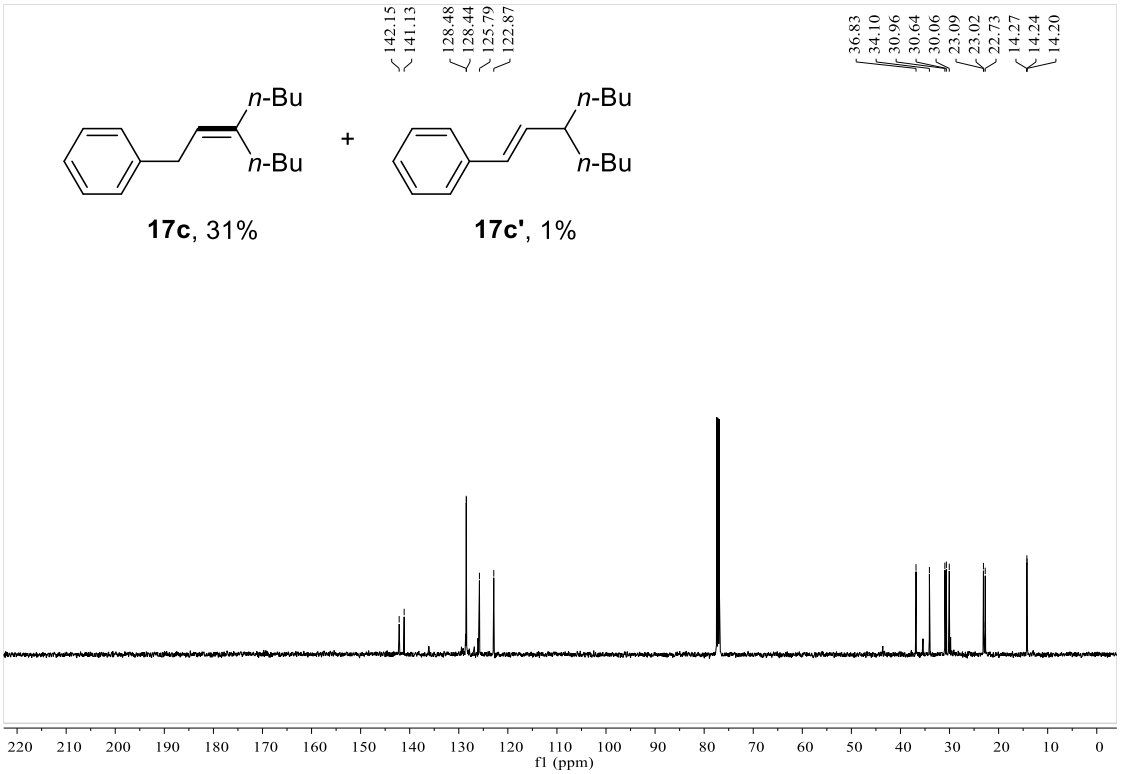


^{13}C NMR spectrum in $CDCl_3$.

(2-(cyclopent-2-en-1-ylidene)ethyl)benzene (18c):

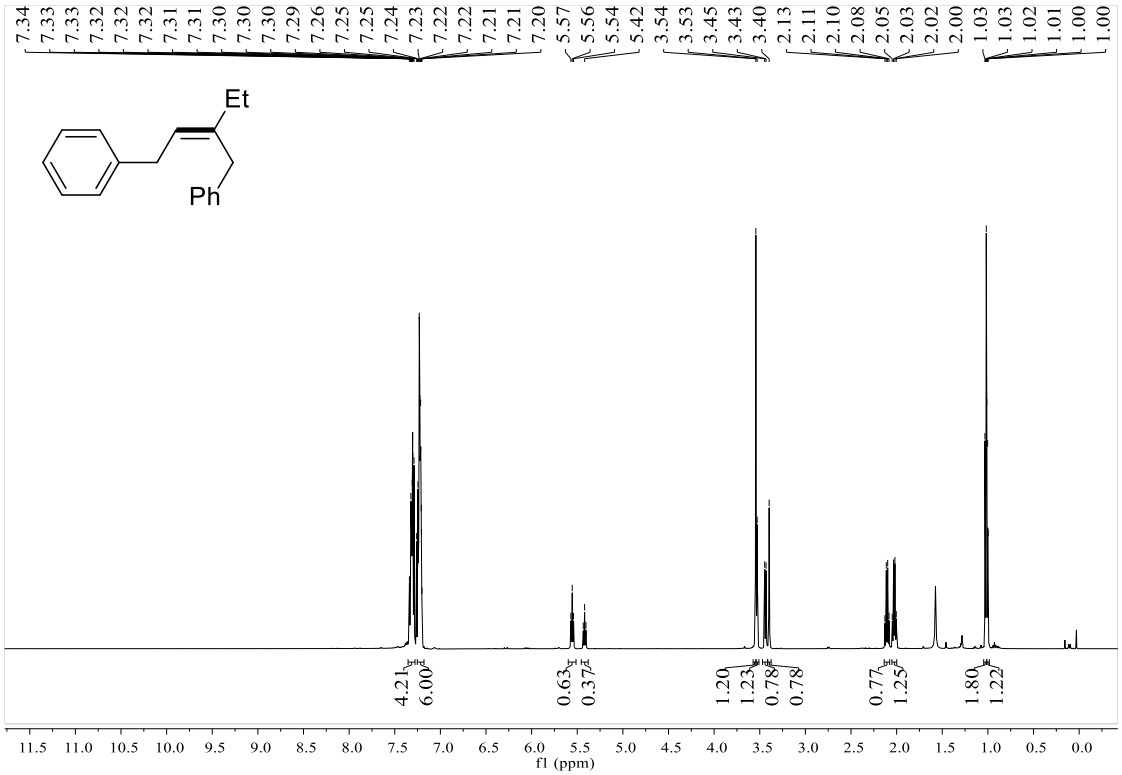


^{1}H NMR spectrum in $CDCl_3$.

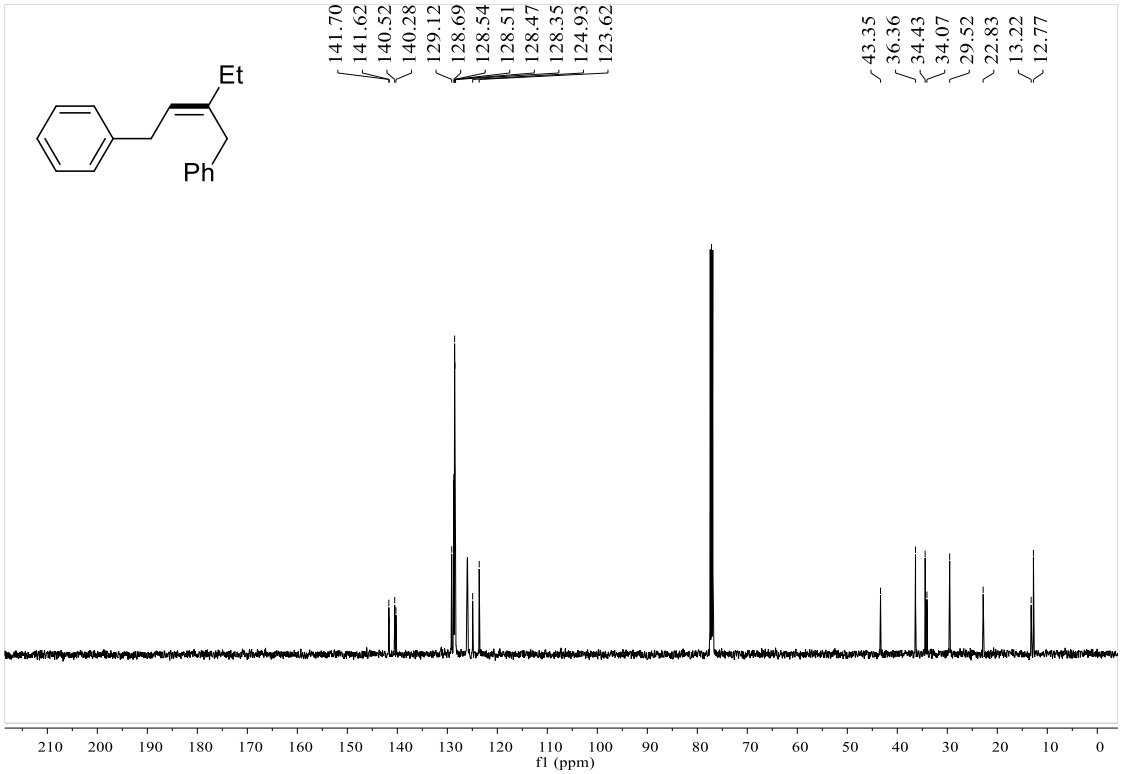


^{13}C NMR spectrum in $CDCl_3$.

(2-ethylbut-2-ene-1,4-diyl)dibenzene (19c):

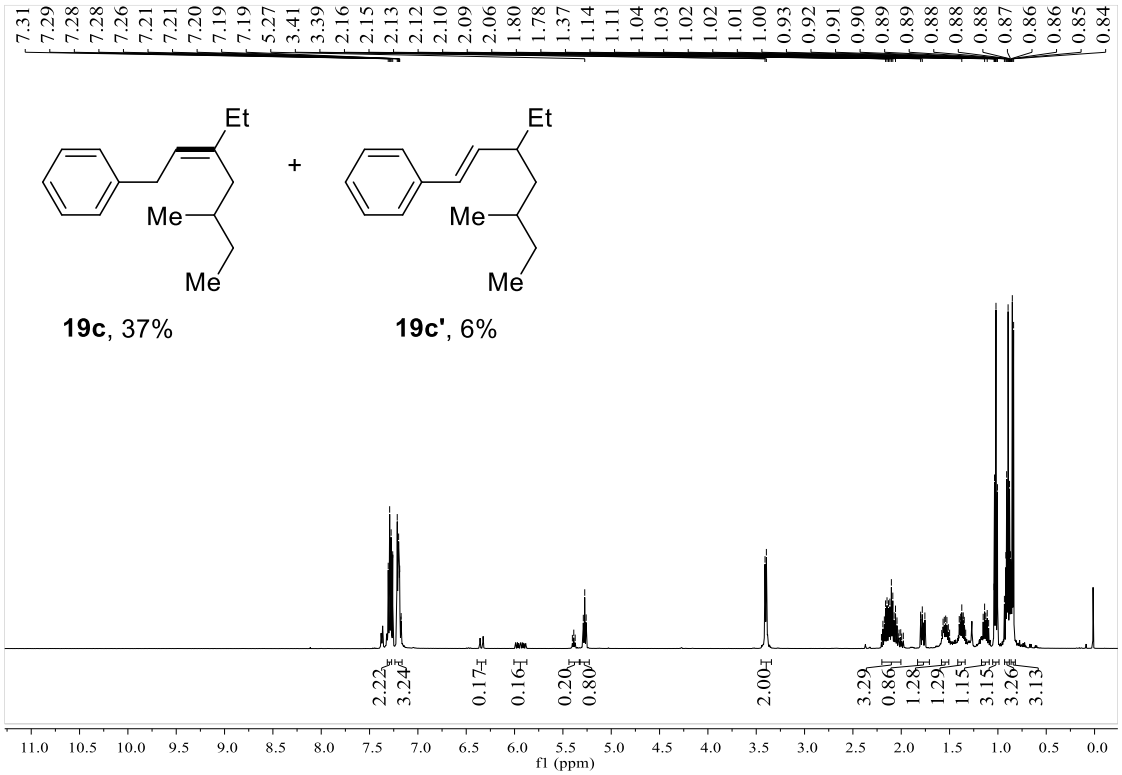


^{1}H NMR spectrum in $CDCl_3$.

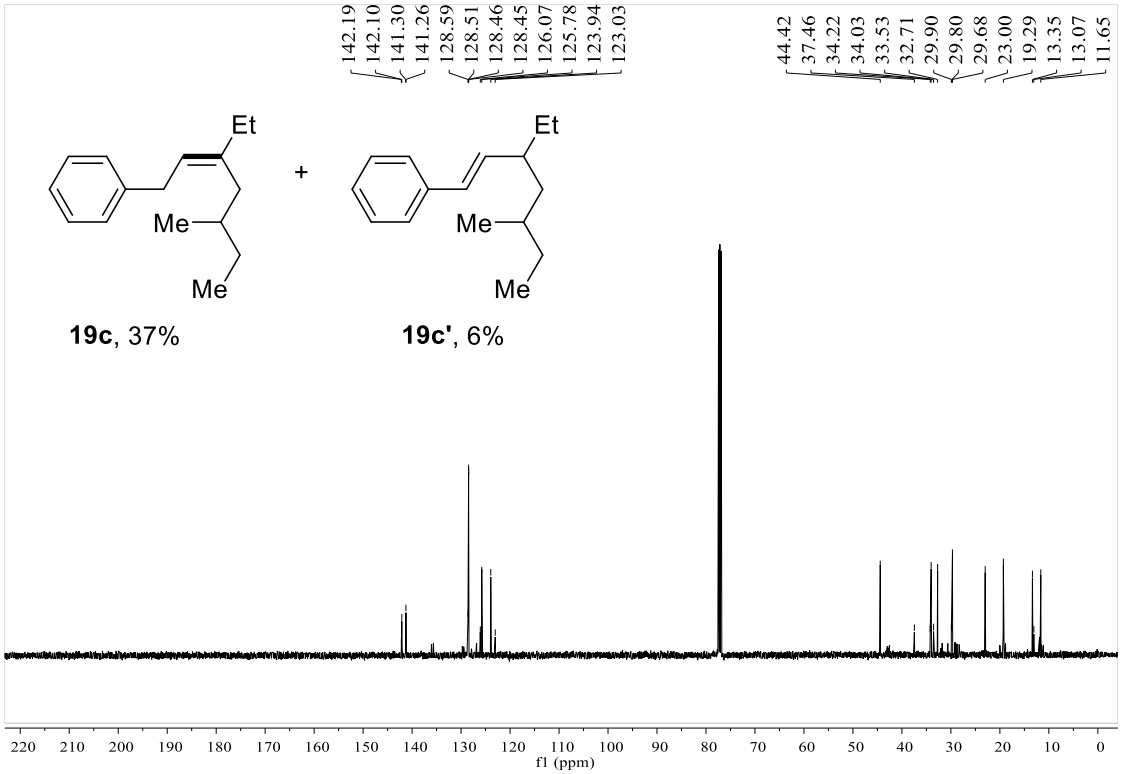


^{13}C NMR spectrum in $CDCl_3$.

(2-(cyclopent-2-en-1-ylidene)ethyl)benzene (20c):

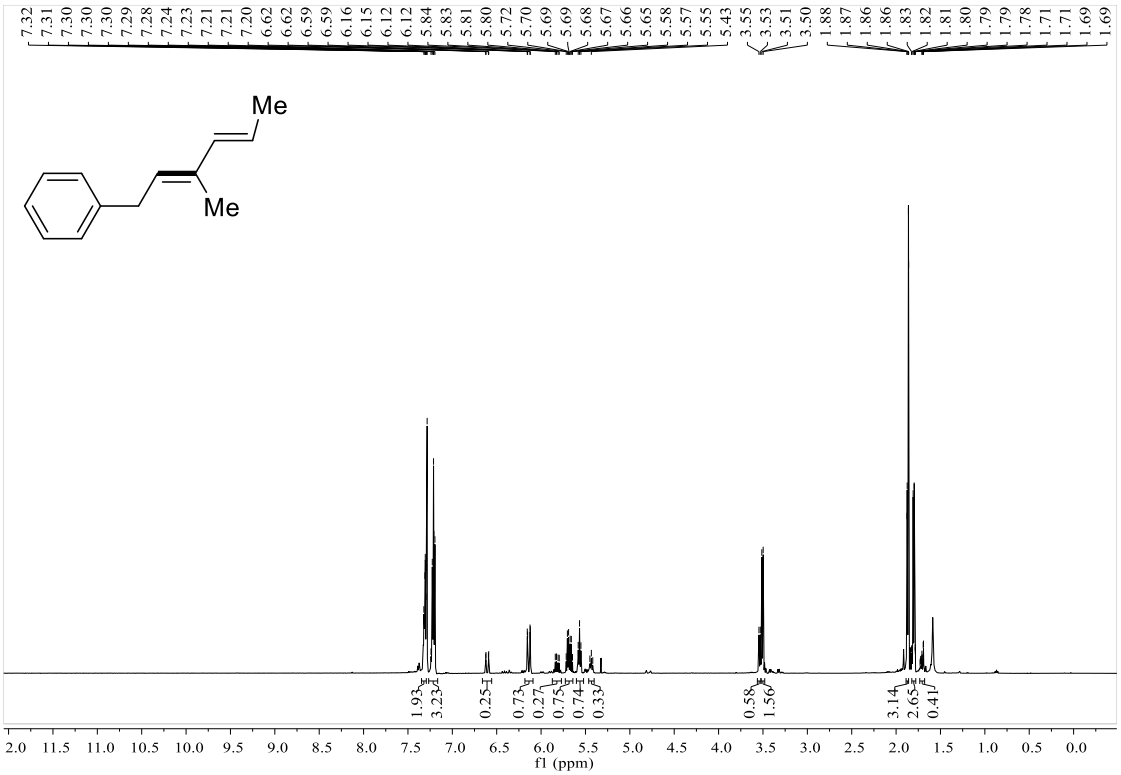


^{1}H NMR spectrum in $CDCl_3$.

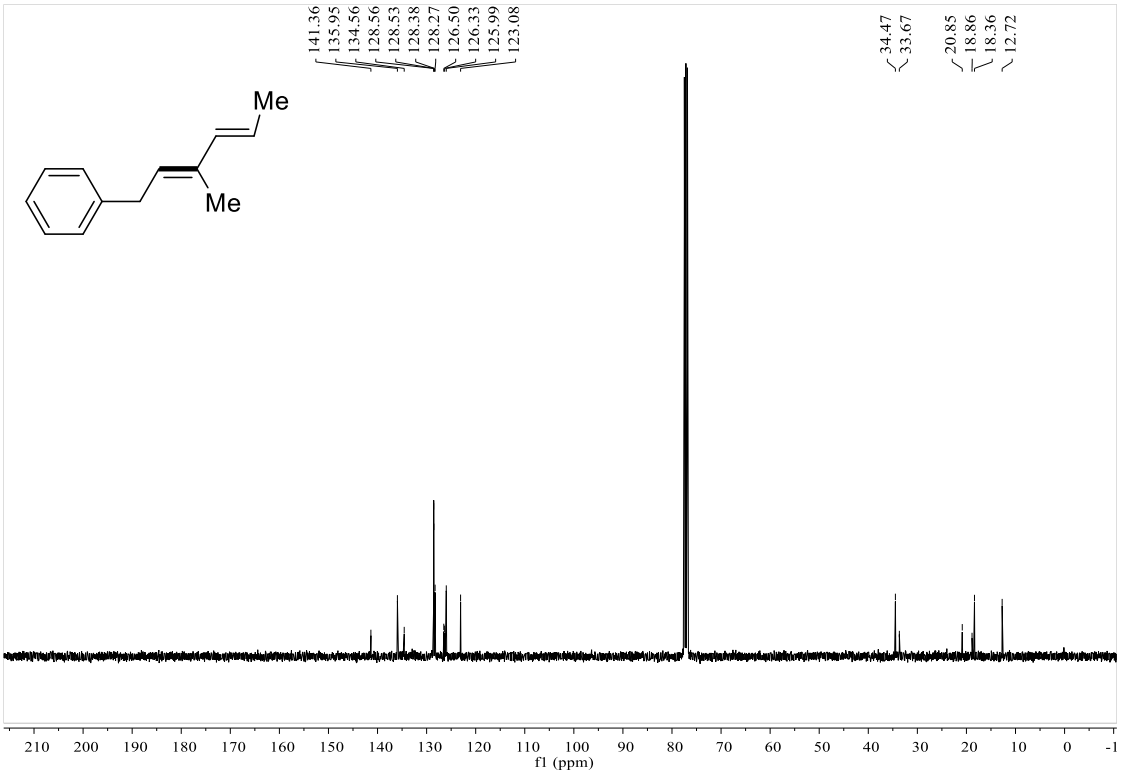


^{13}C NMR spectrum in $CDCl_3$.

3-methylhexa-2,4-dien-1-yl)benzene (21c):

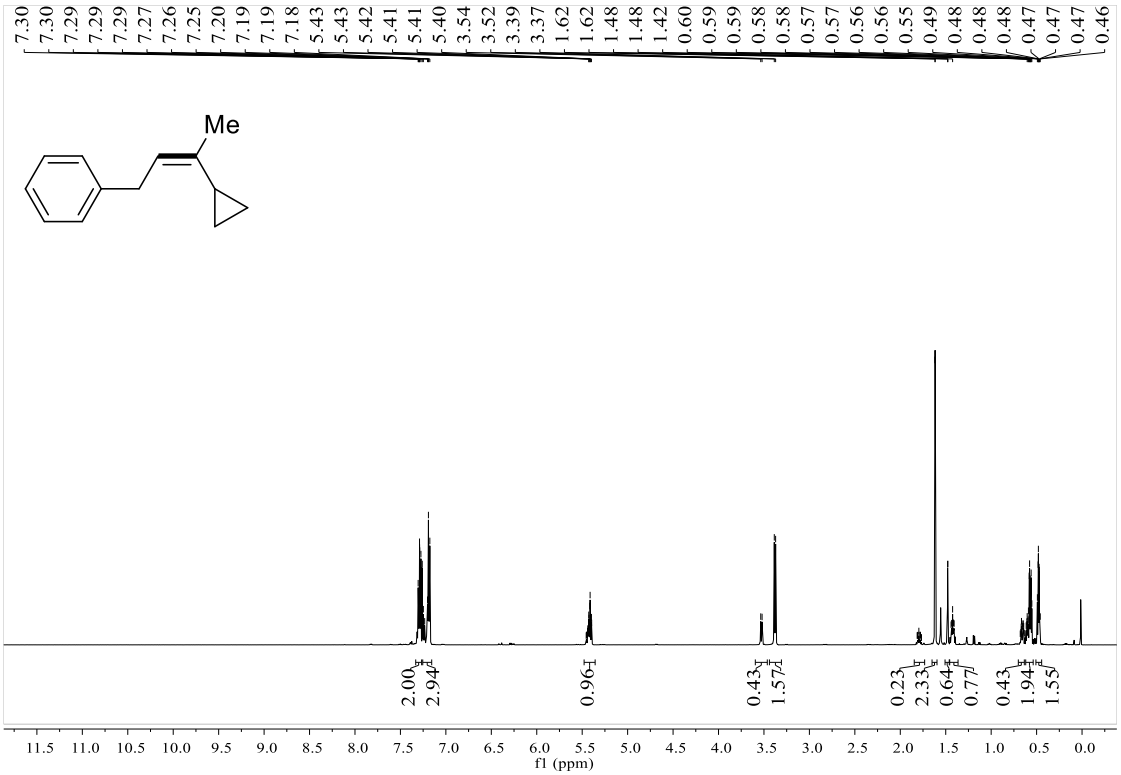


^{1}H NMR spectrum in $CDCl_3$.

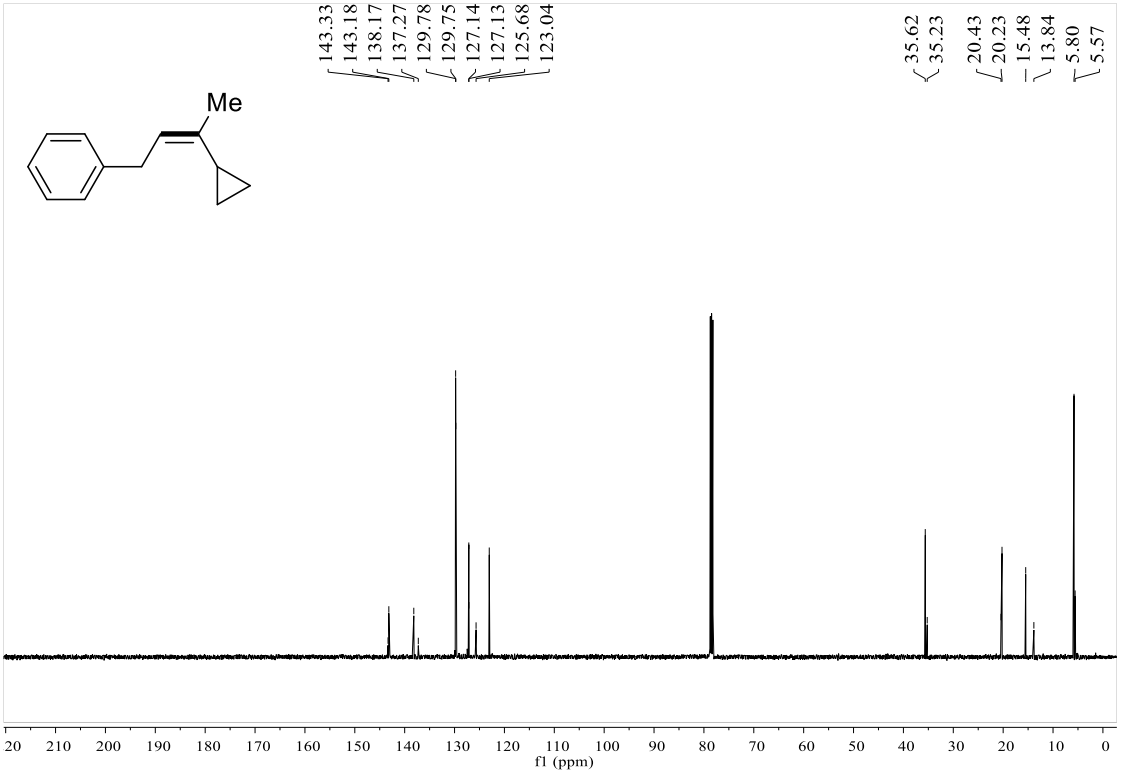


^{13}C NMR spectrum in $CDCl_3$.

(3-cyclopropylbut-2-en-1-yl)benzene (22c):

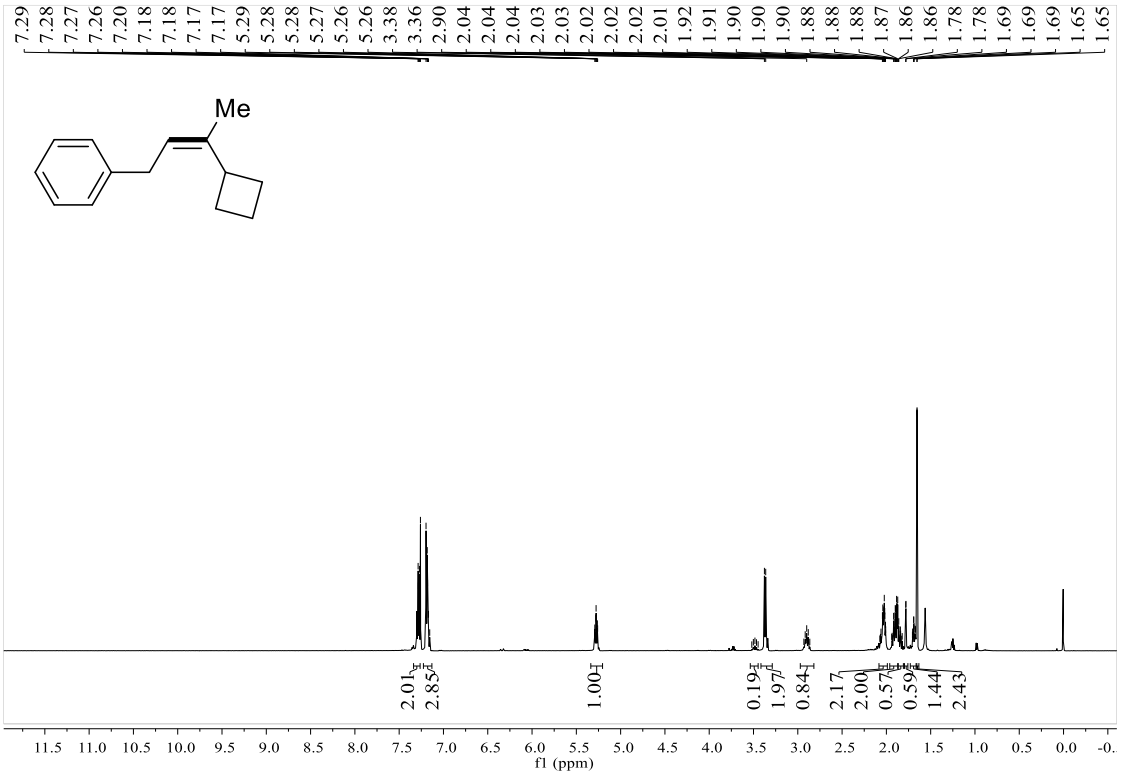


^{1}H NMR spectrum in $CDCl_3$.

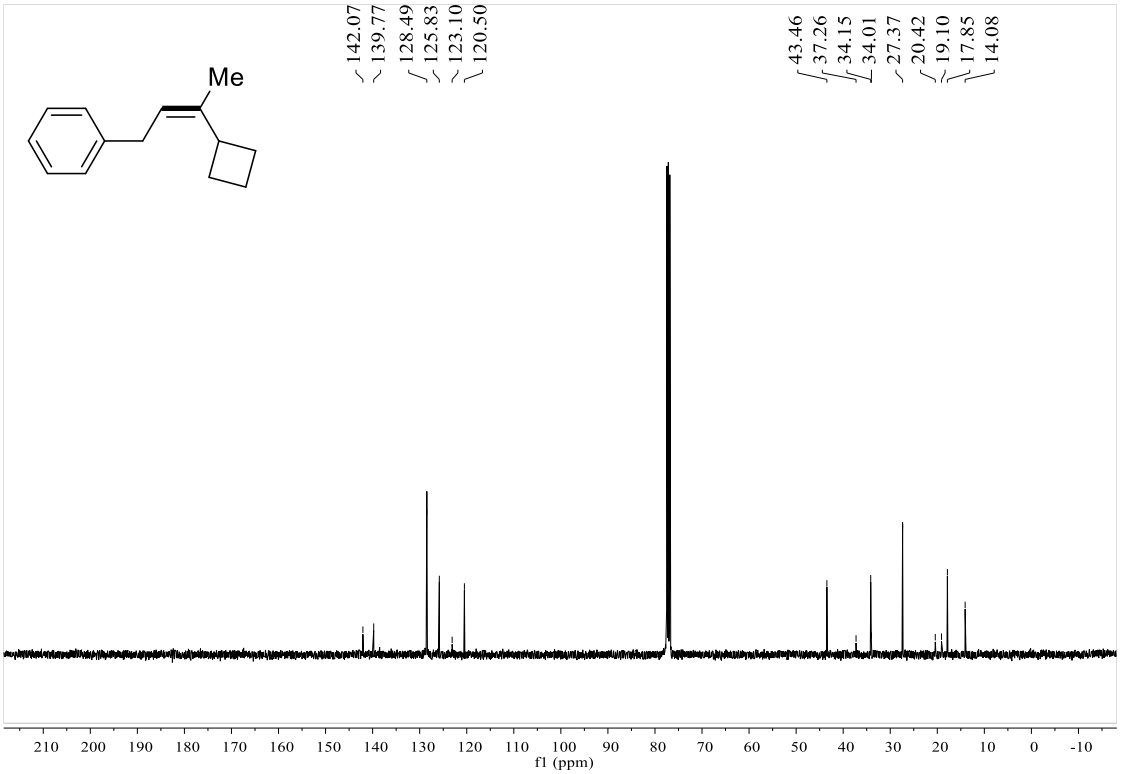


^{13}C NMR spectrum in $CDCl_3$.

(3-cyclobutylbut-2-en-1-yl)benzene (23c):

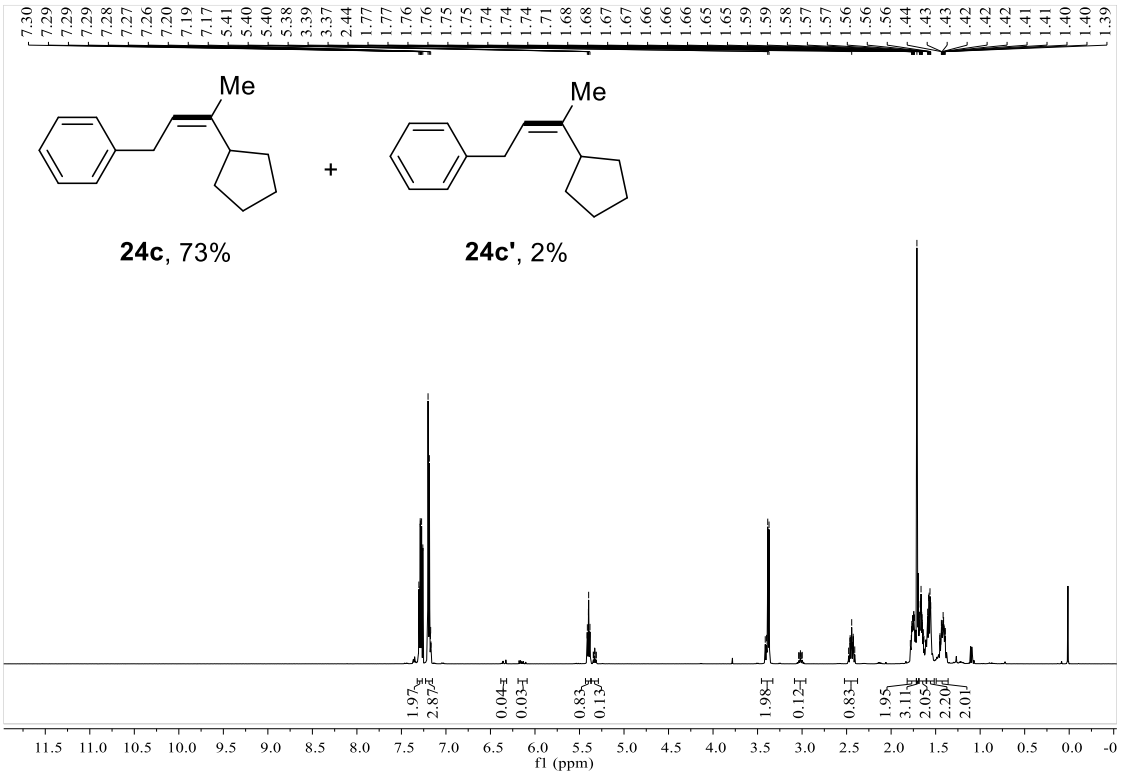


^{1}H NMR spectrum in $CDCl_3$.

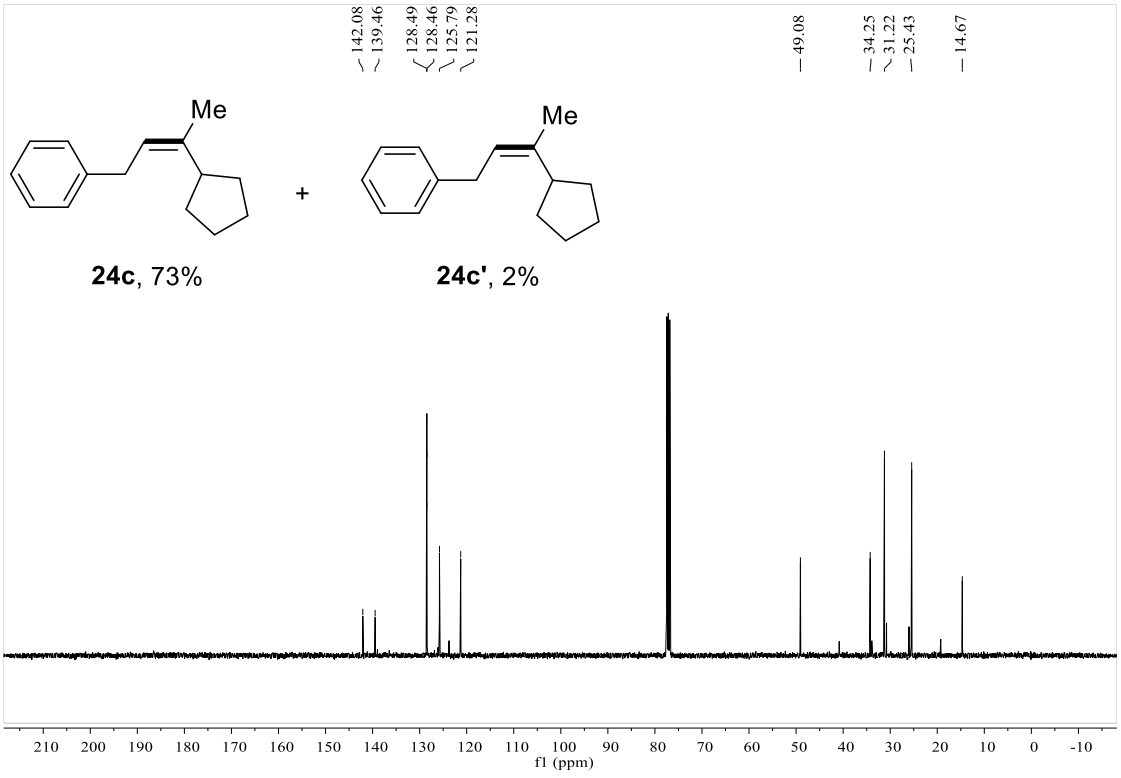


^{13}C NMR spectrum in $CDCl_3$.

(3-cyclopentylbut-2-en-1-yl)benzene (24c):

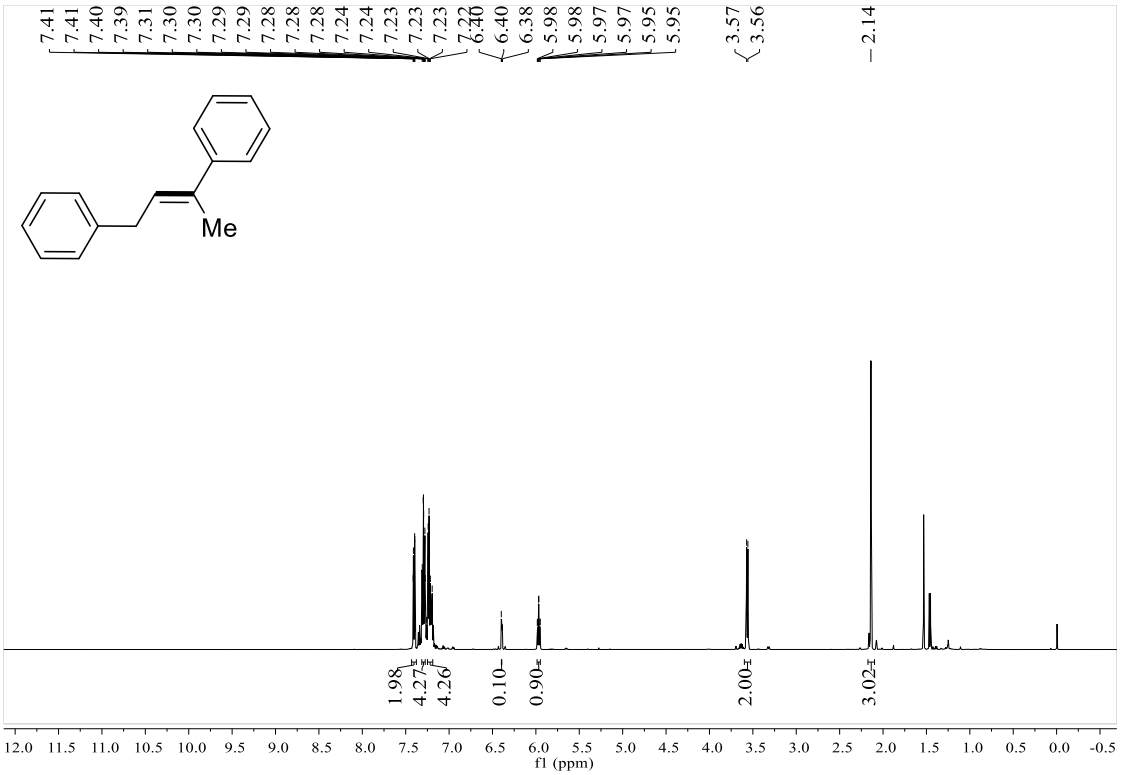


^{1}H NMR spectrum in $CDCl_3$.

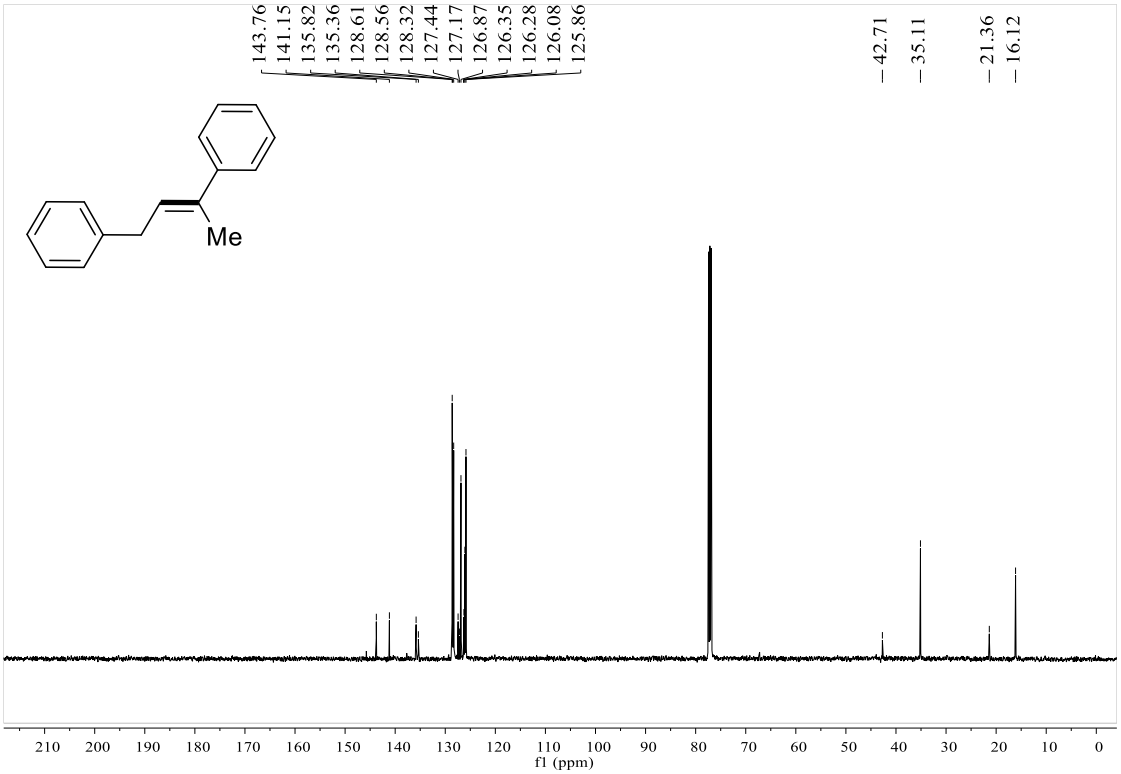


^{13}C NMR spectrum in $CDCl_3$.

but-2-ene-1,3-diyldibenzene (25c):

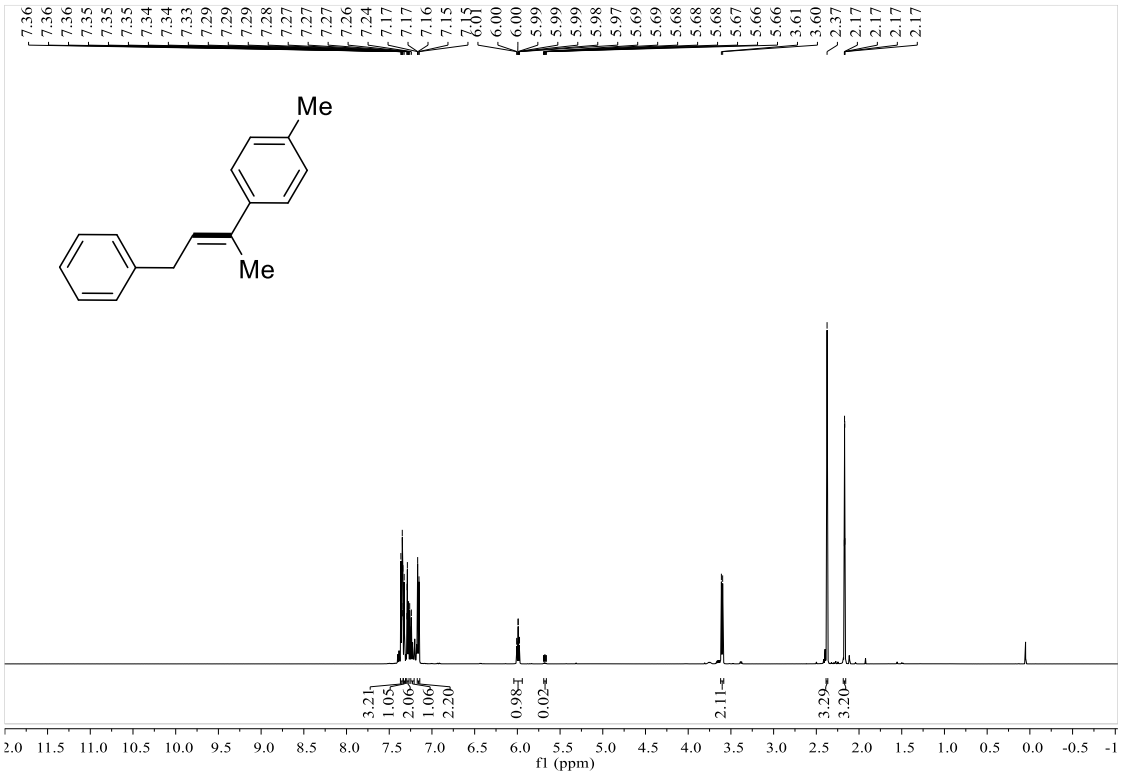


^{1}H NMR spectrum in $CDCl_3$.

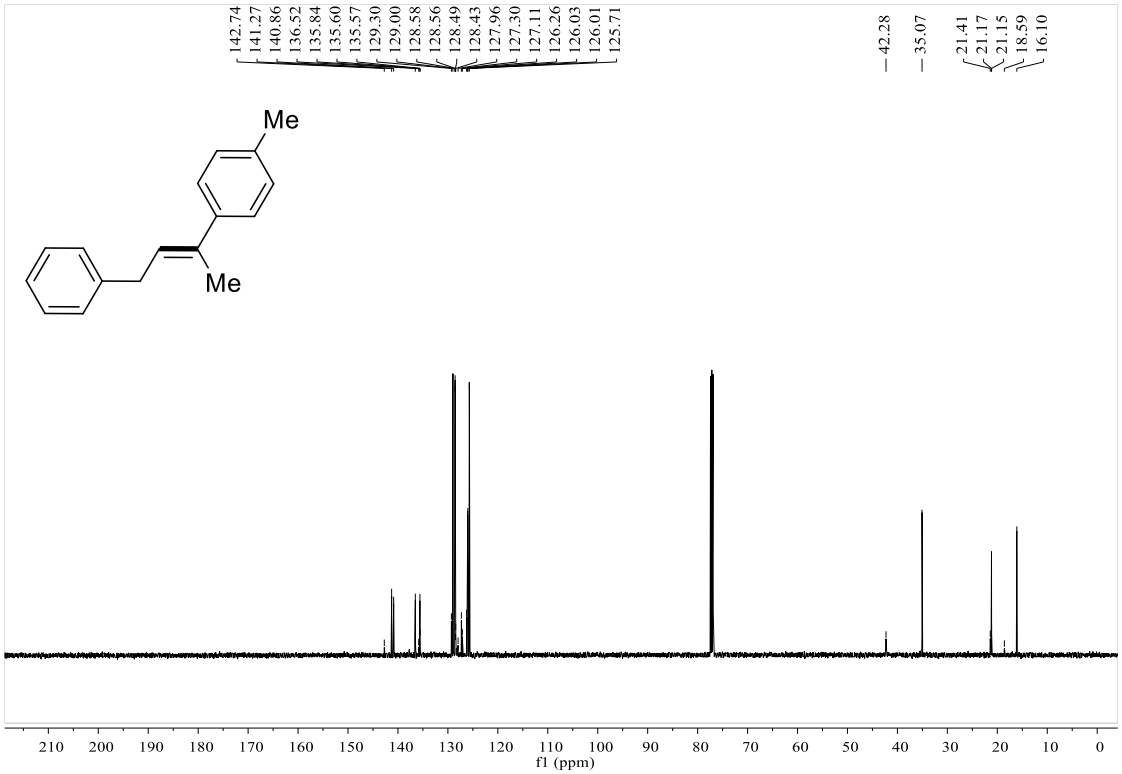


^{13}C NMR spectrum in $CDCl_3$.

methyl-4-(4-phenylbut-2-en-2-yl)benzene (26c):

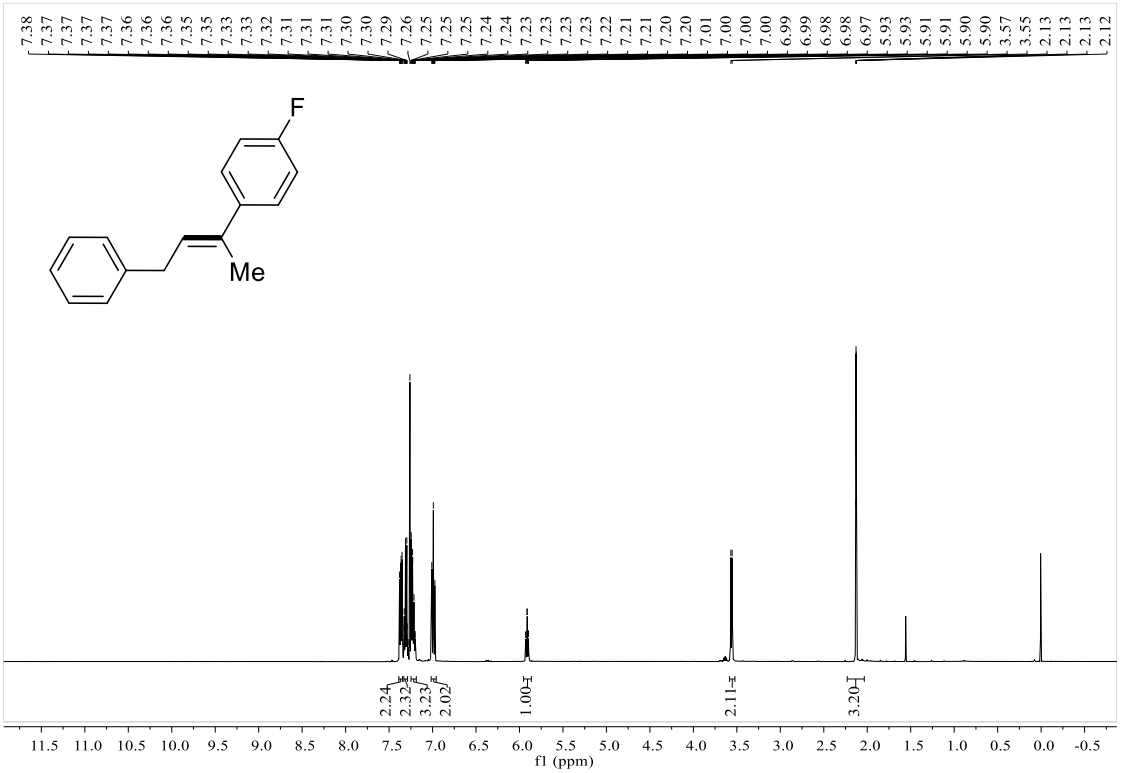


^{1}H NMR spectrum in $CDCl_3$.

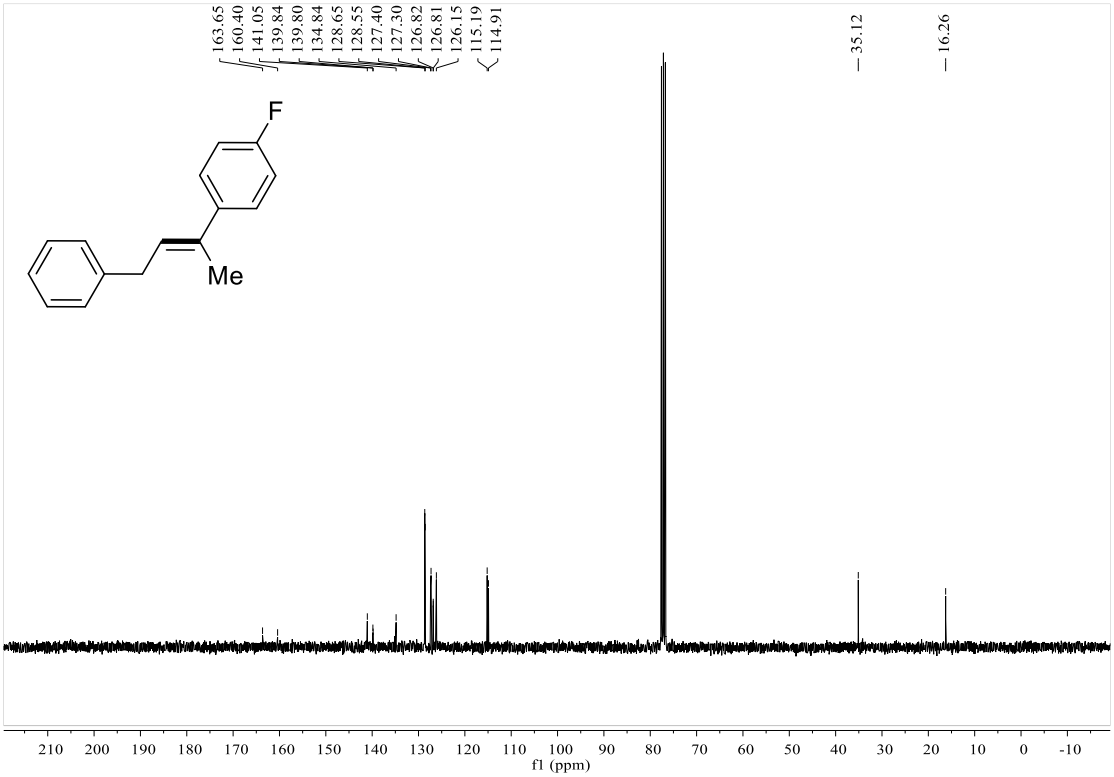


^{13}C NMR spectrum in $CDCl_3$.

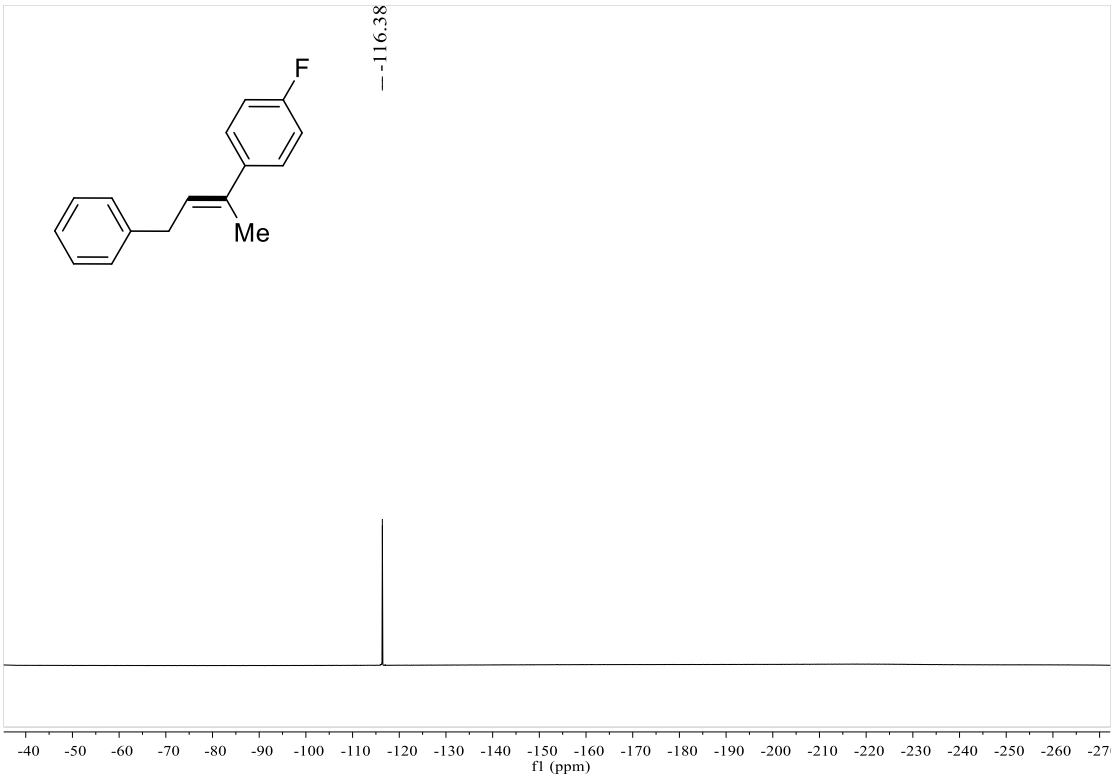
1-fluoro-4-(4-phenylbut-2-en-2-yl)benzene (27c):



^{1}H NMR spectrum in $CDCl_3$.

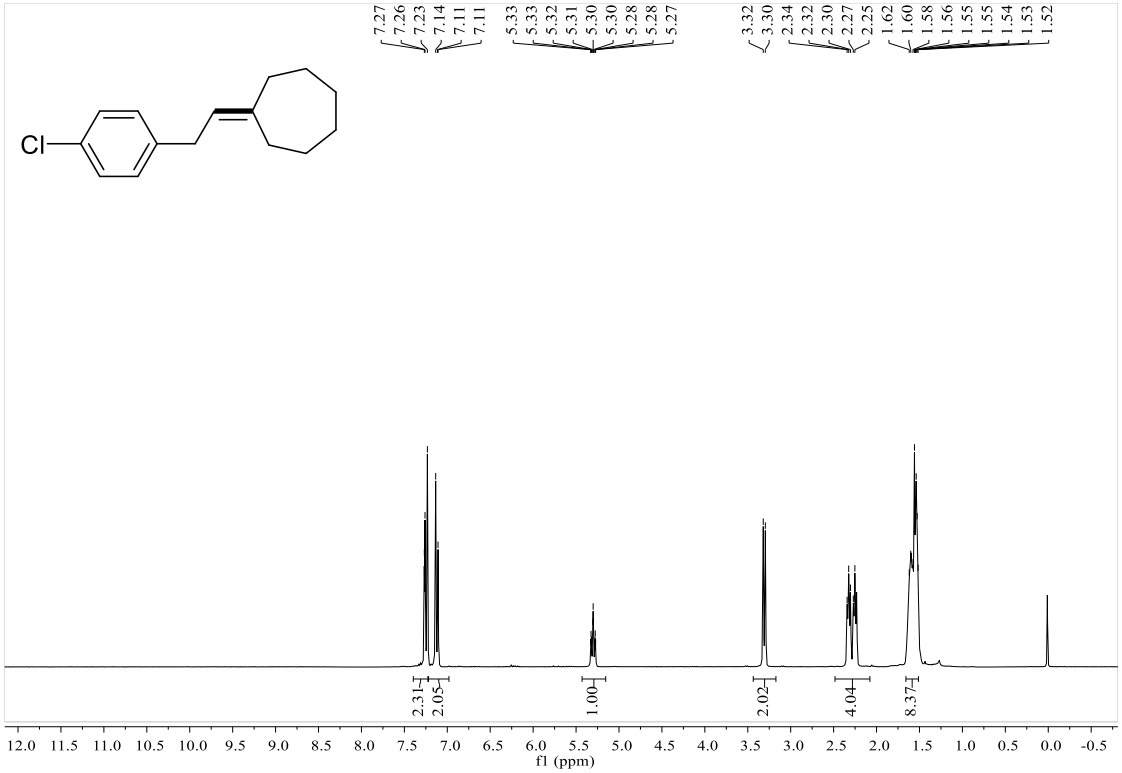


^{13}C NMR spectrum in $CDCl_3$.

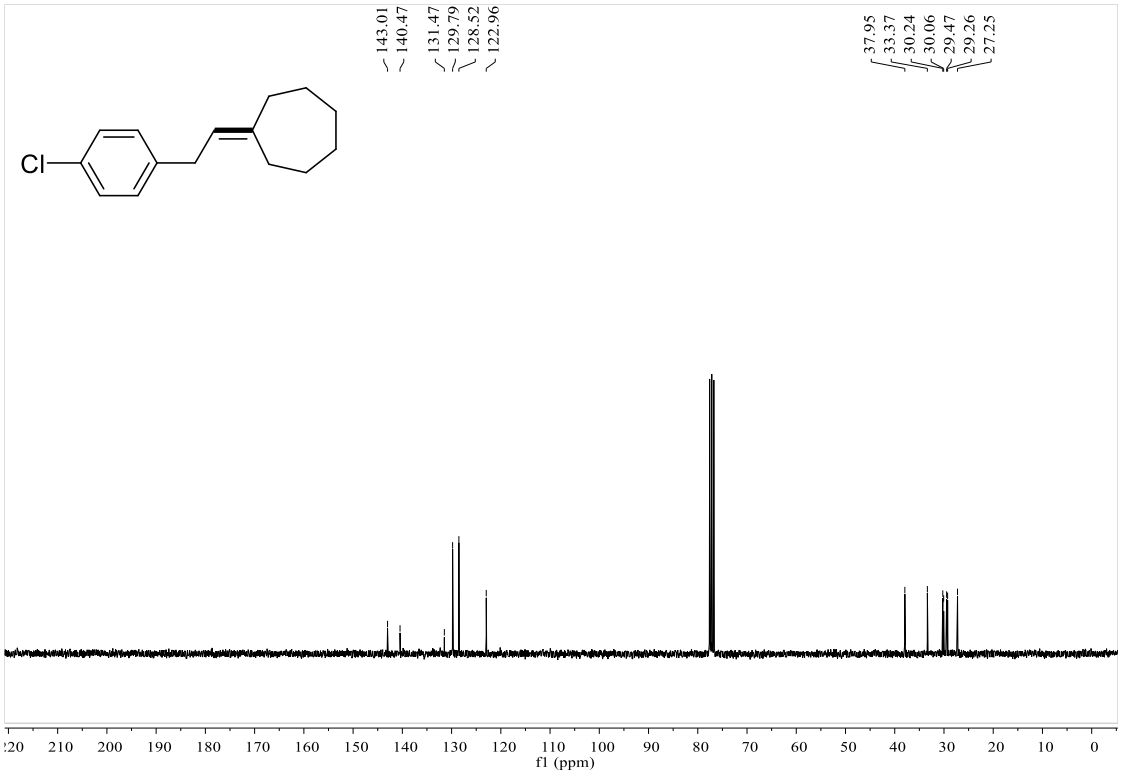


^{19}F NMR spectrum in $CDCl_3$.

(2-(4-chlorophenyl)ethylidene)cycloheptane (28c):

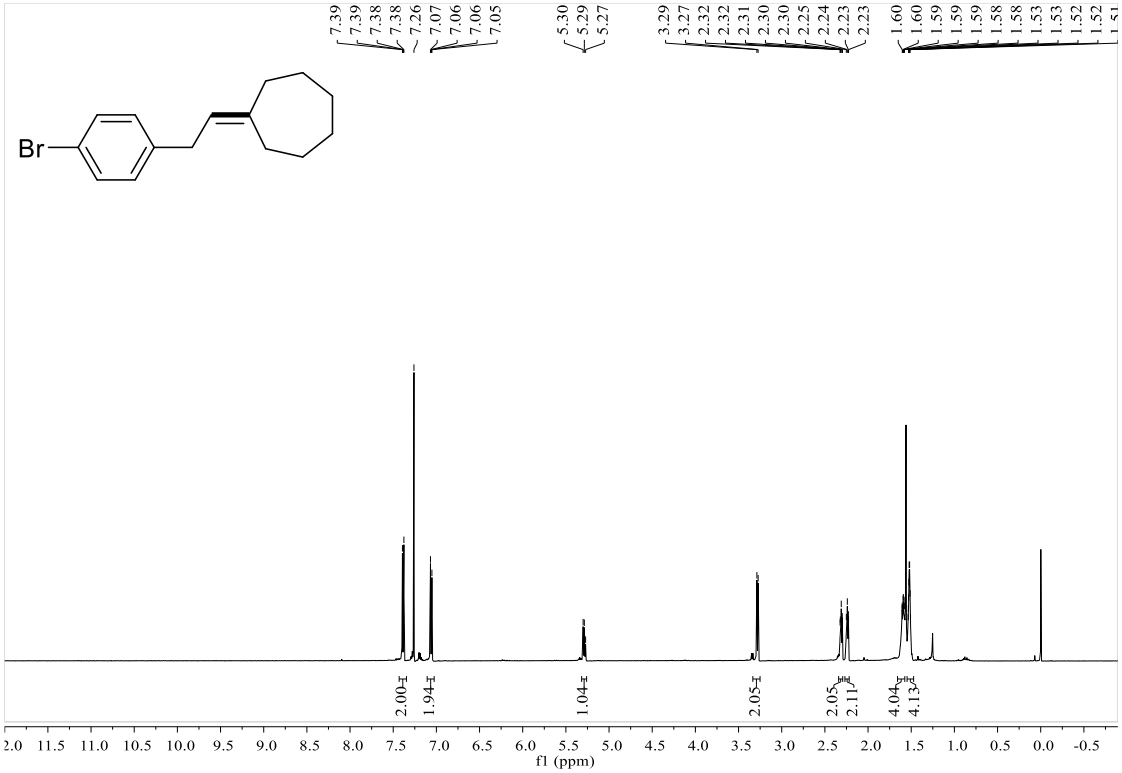


^{1}H NMR spectrum in $CDCl_3$.

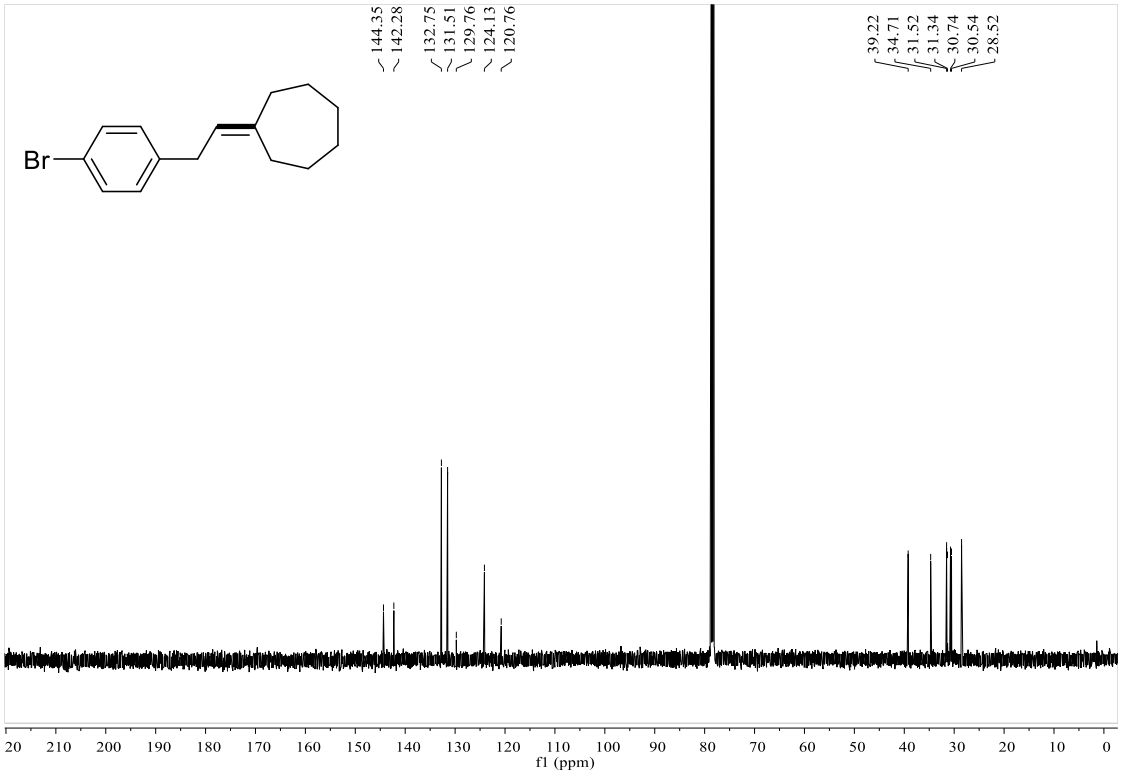


^{13}C NMR spectrum in $CDCl_3$.

(2-(4-chlorophenyl)ethylidene)cycloheptane (29c):

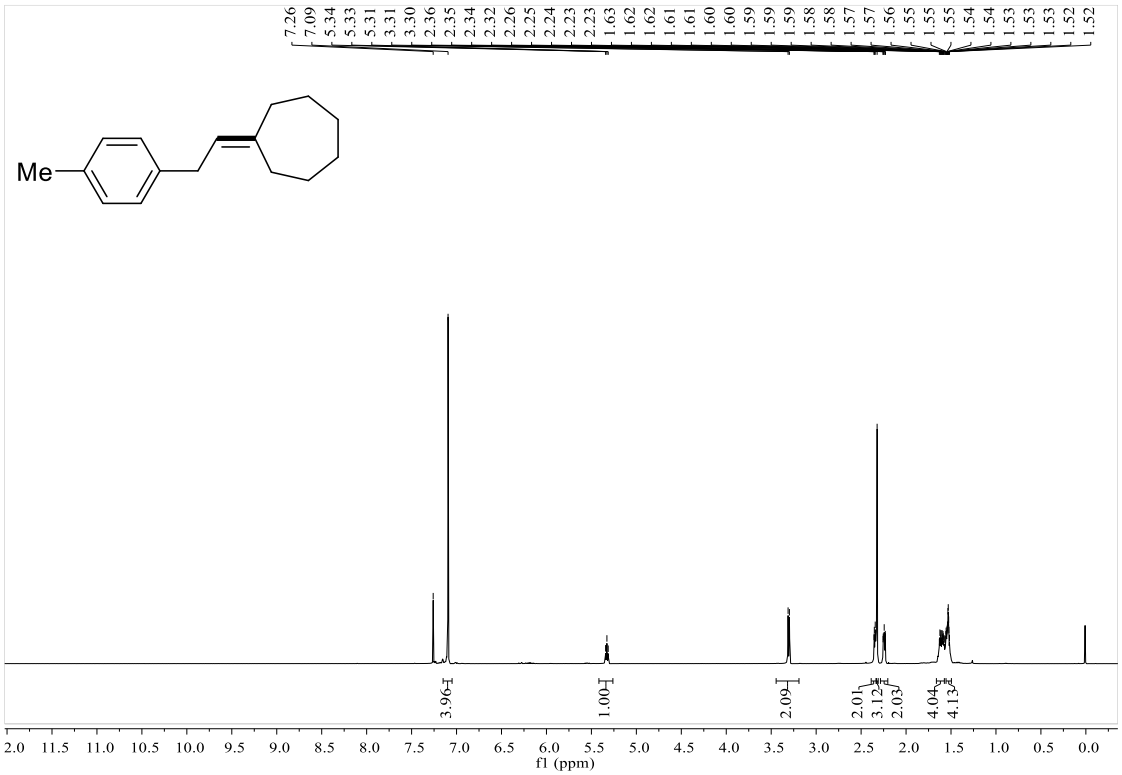


^{1}H NMR spectrum in $CDCl_3$.

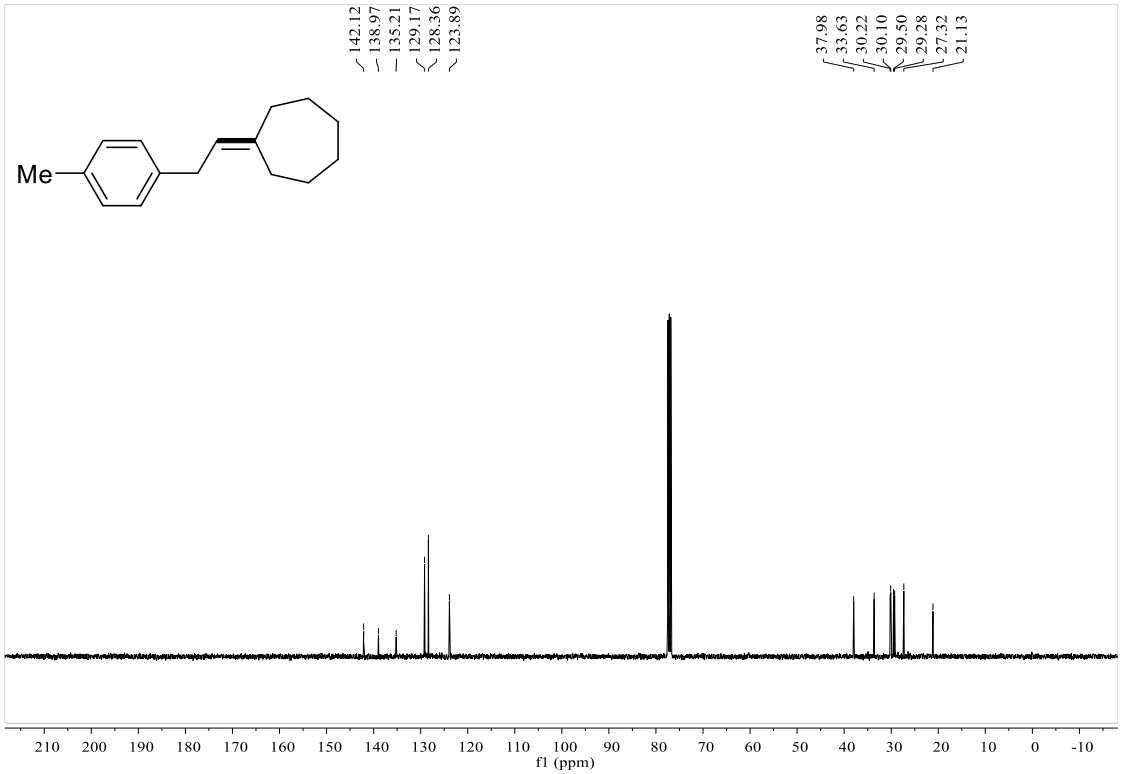


^{13}C NMR spectrum in $CDCl_3$.

(2-(4-chlorophenyl)ethylidene)cycloheptane (30c):

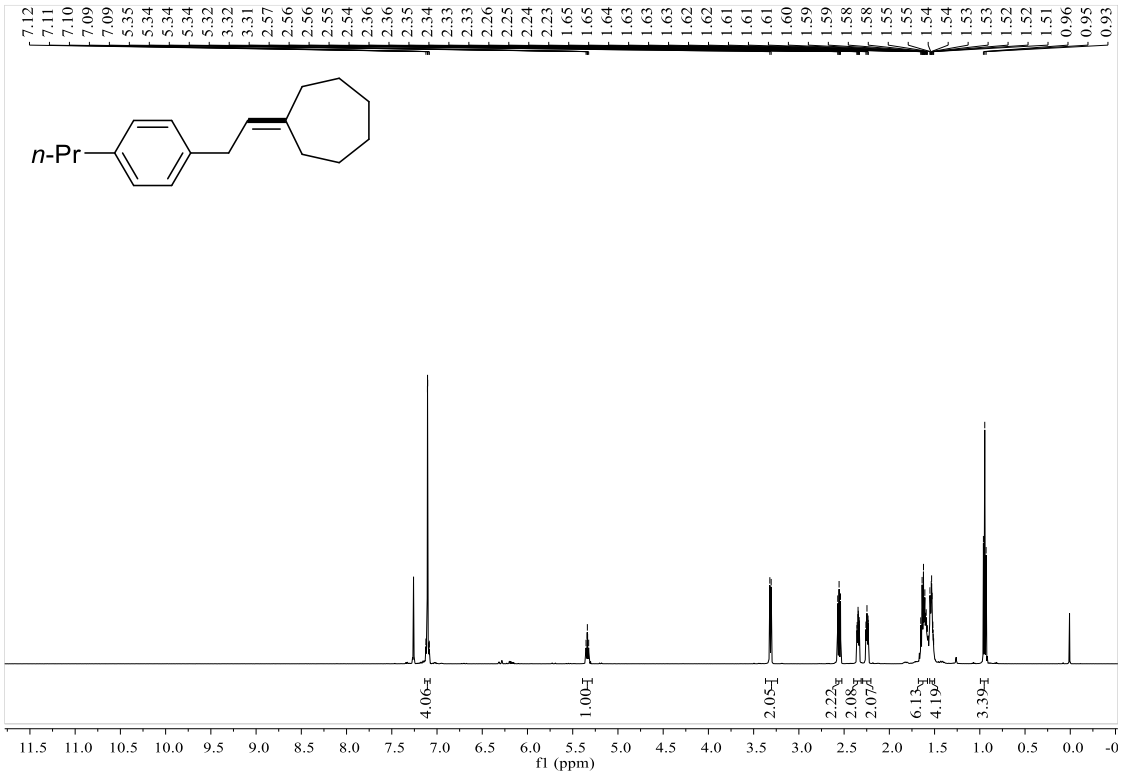


^{1}H NMR spectrum in $CDCl_3$.

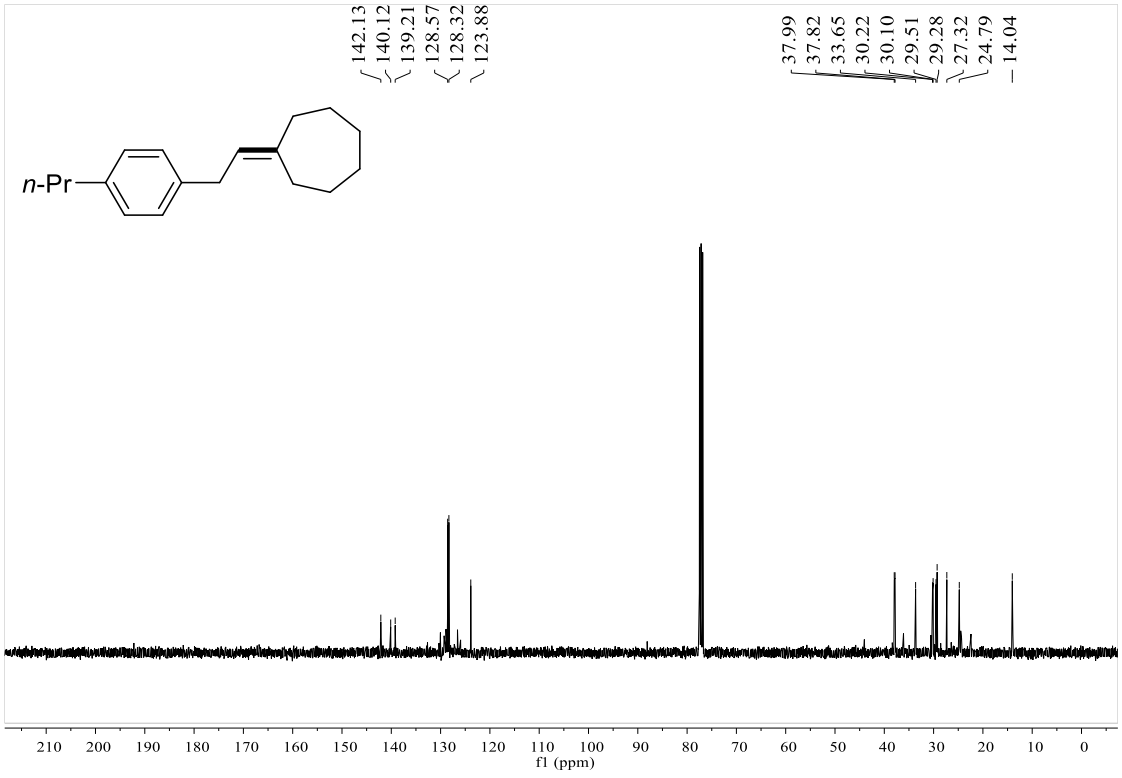


^{13}C NMR spectrum in $CDCl_3$.

(2-(4-chlorophenyl)ethylidene)cycloheptane (31c):

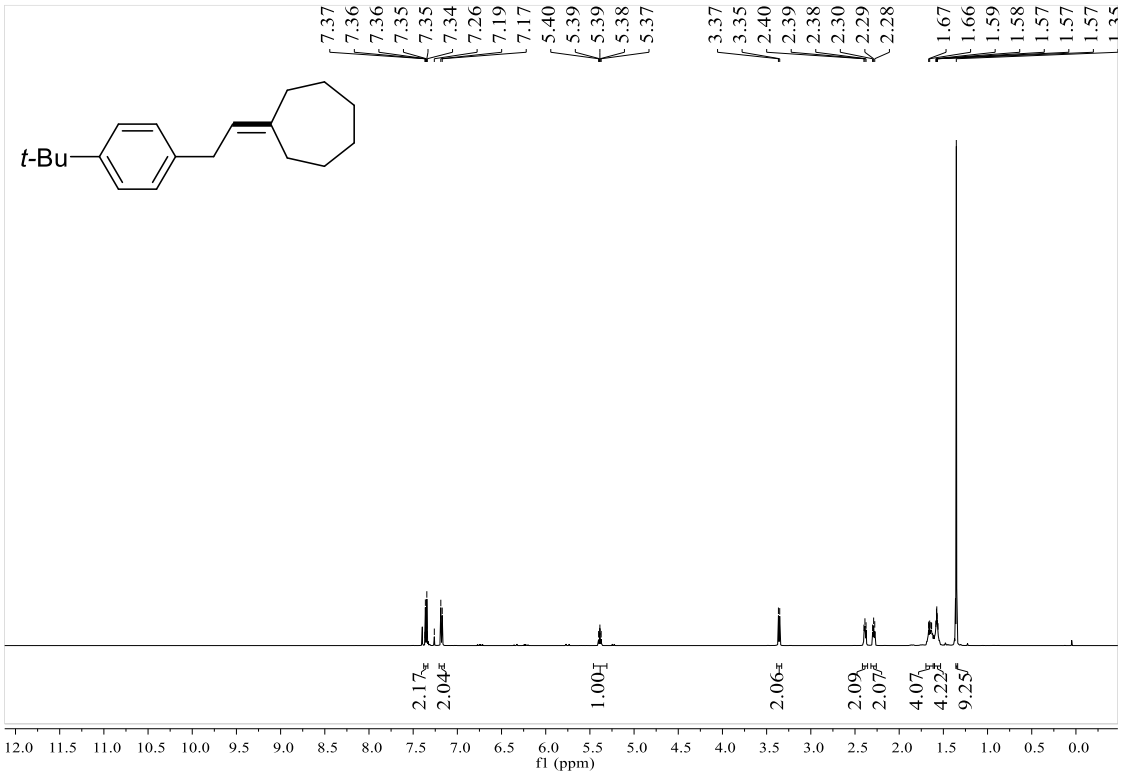


^{1}H NMR spectrum in $CDCl_3$.

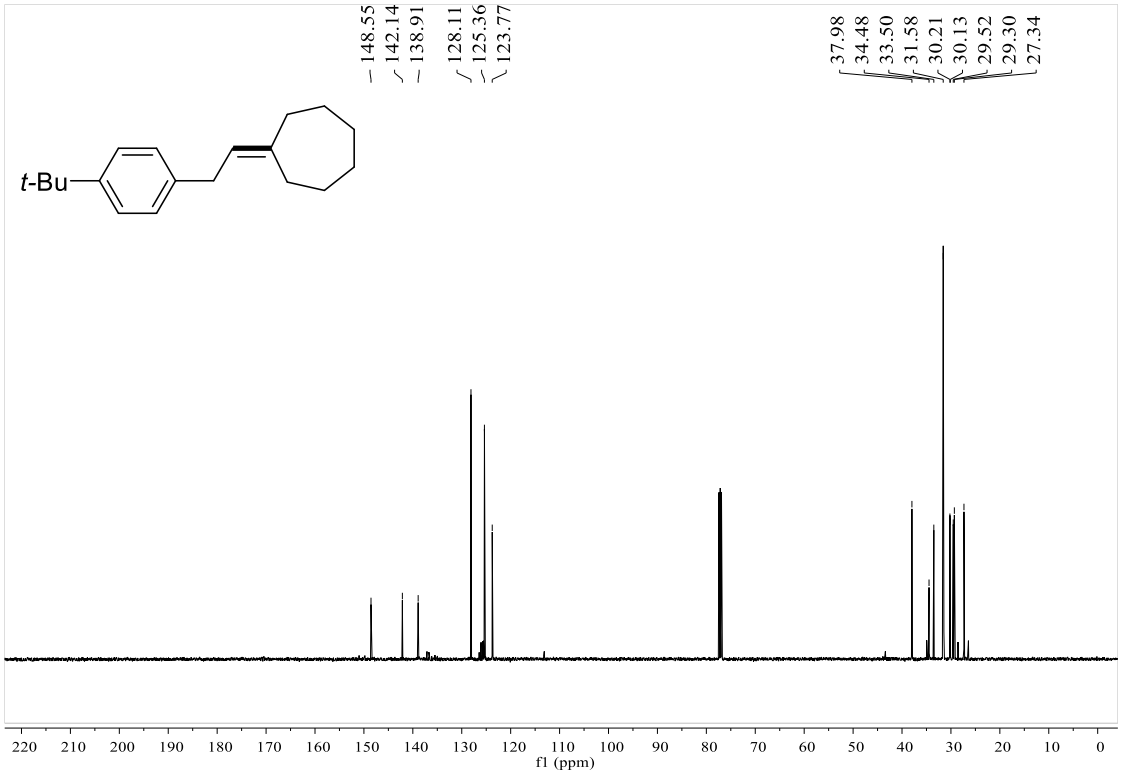


^{13}C NMR spectrum in $CDCl_3$.

(2-(4-(*tert*-butyl)phenyl)ethylidene)cycloheptane (32c):

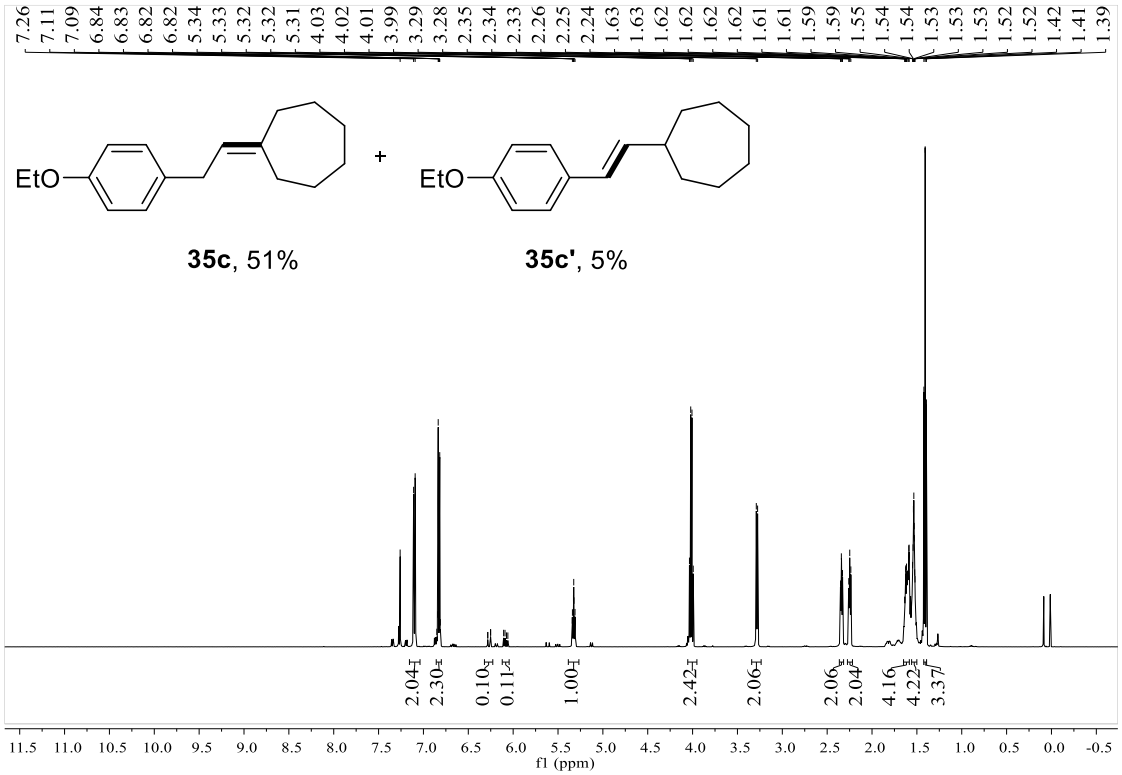


^{1}H NMR spectrum in $CDCl_3$.

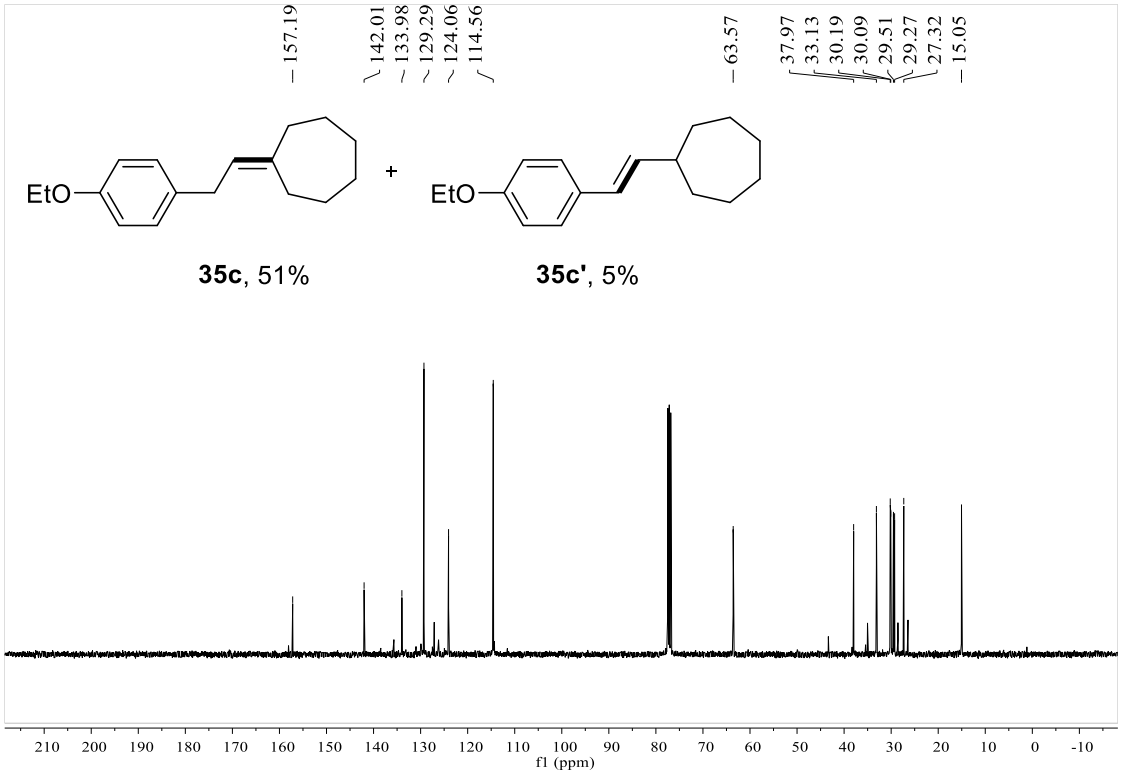


^{13}C NMR spectrum in $CDCl_3$.

(2-(4-(*tert*-butyl)phenyl)ethylidene)cycloheptane (33c):

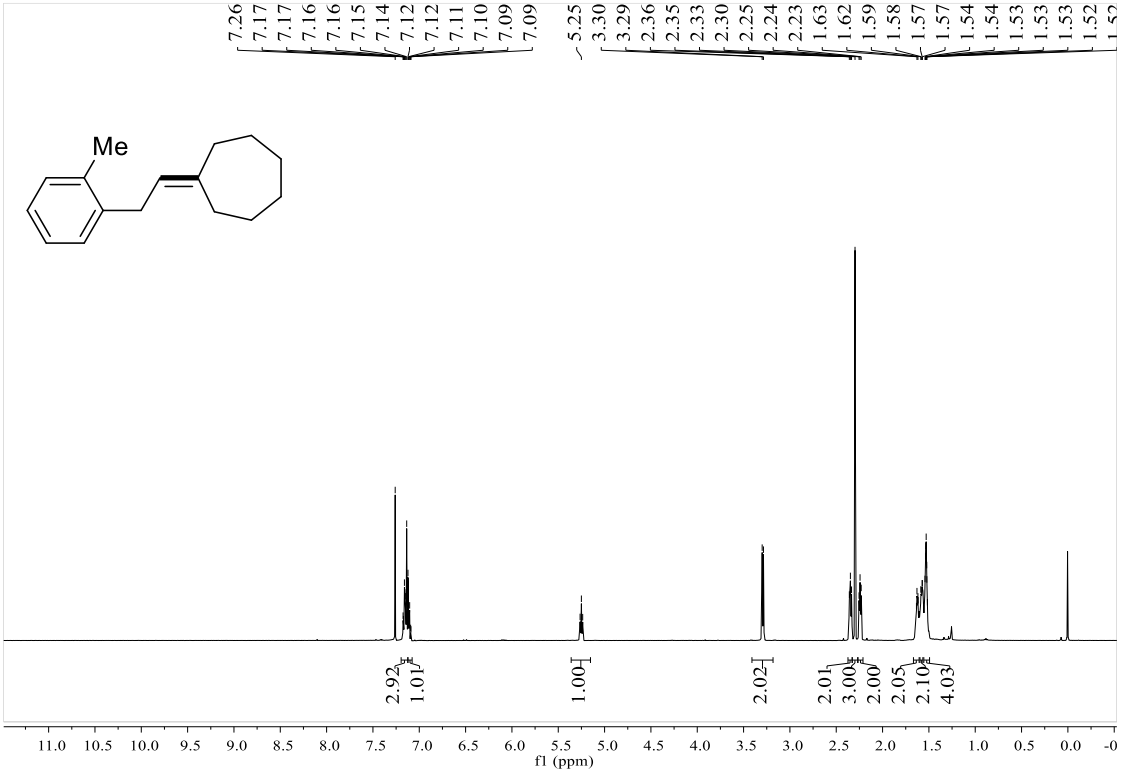


^{1}H NMR spectrum in $CDCl_3$.

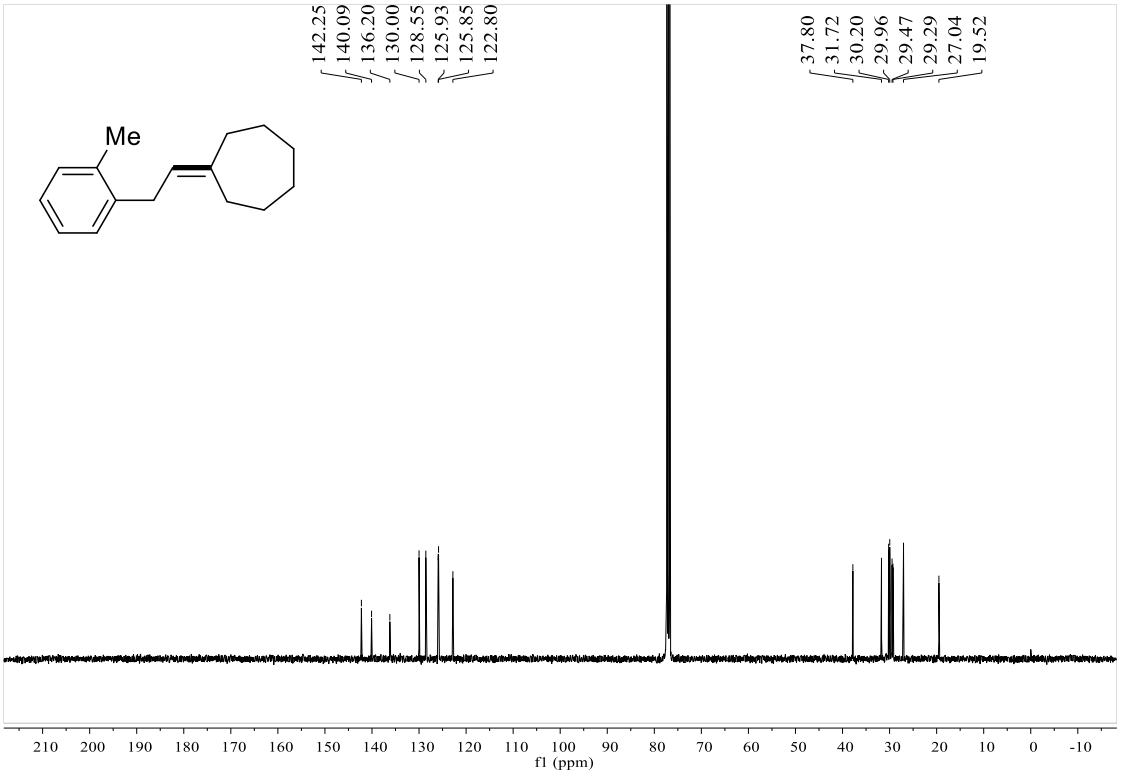


^{13}C NMR spectrum in $CDCl_3$.

(2-(4-chlorophenyl)ethylidene)cycloheptane (34c):

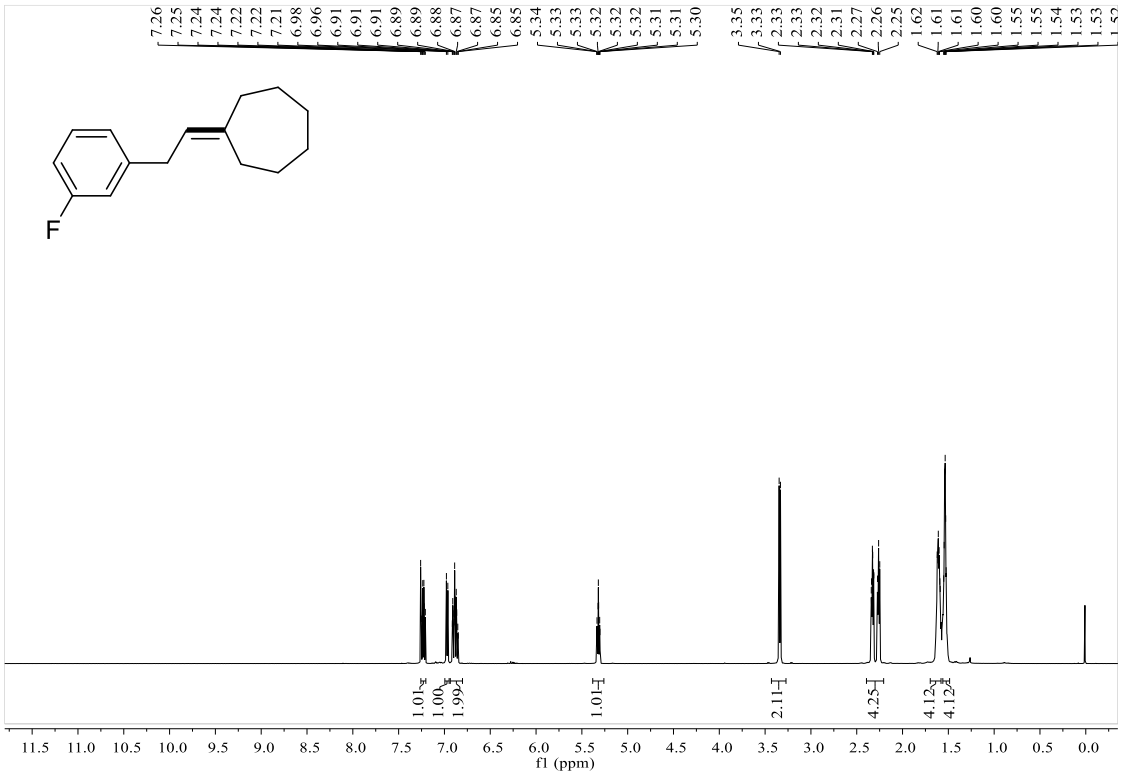


^{1}H NMR spectrum in $CDCl_3$.

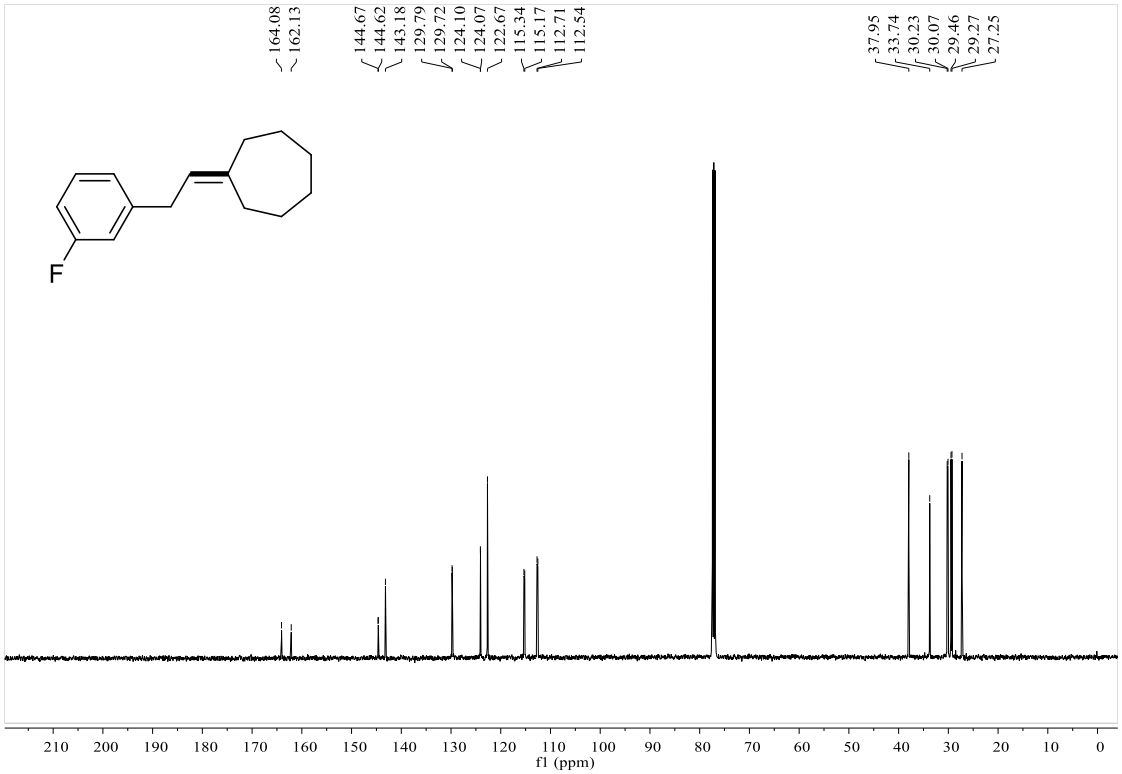


^{13}C NMR spectrum in $CDCl_3$.

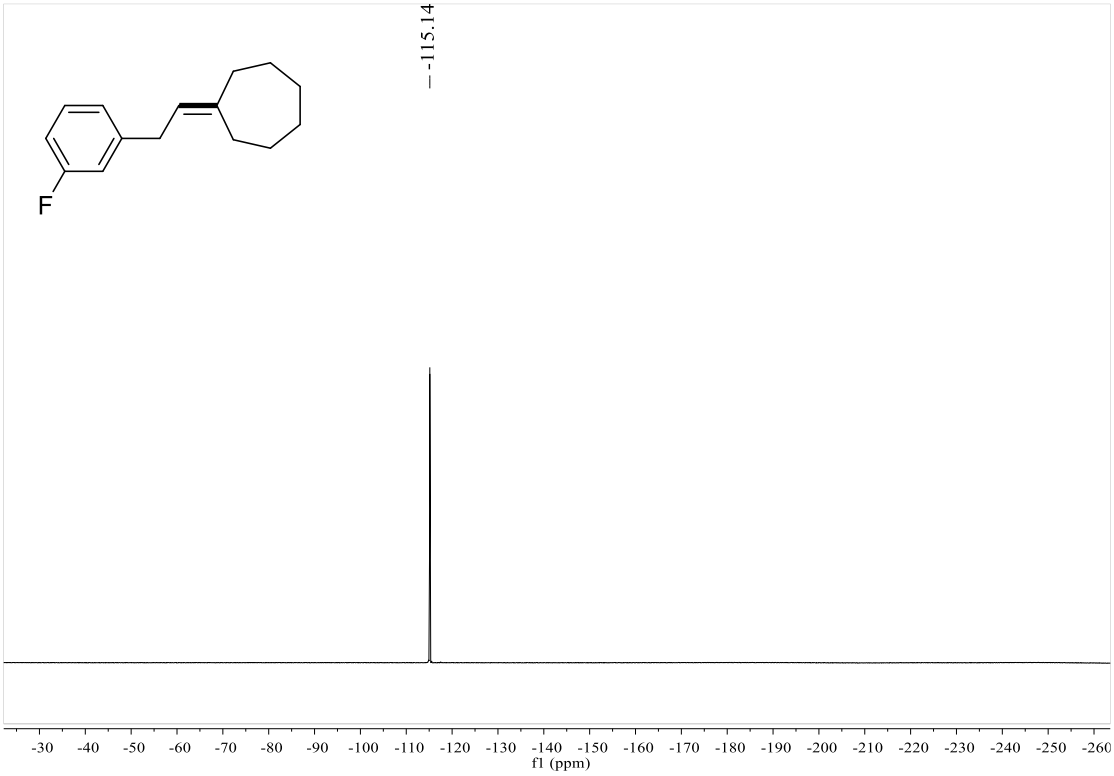
(2-(3-fluorophenyl)ethylidene)cycloheptane (35c):



^{1}H NMR spectrum in $CDCl_3$.

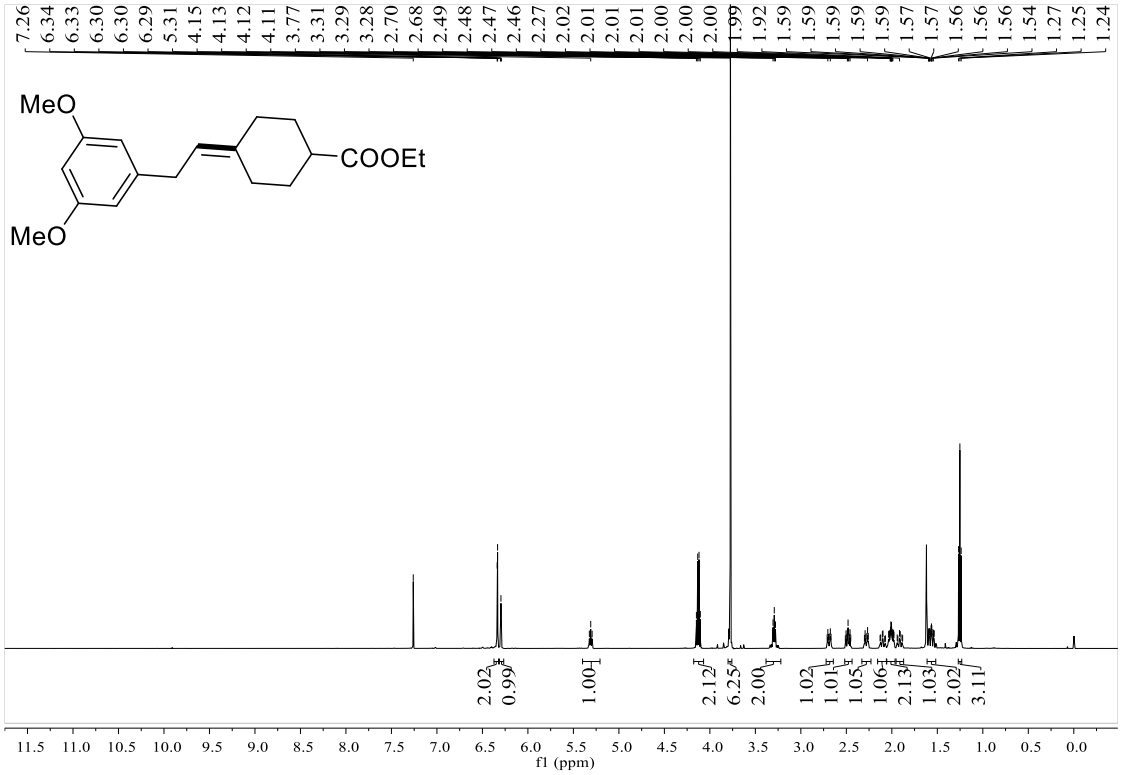


^{13}C NMR spectrum in $CDCl_3$.

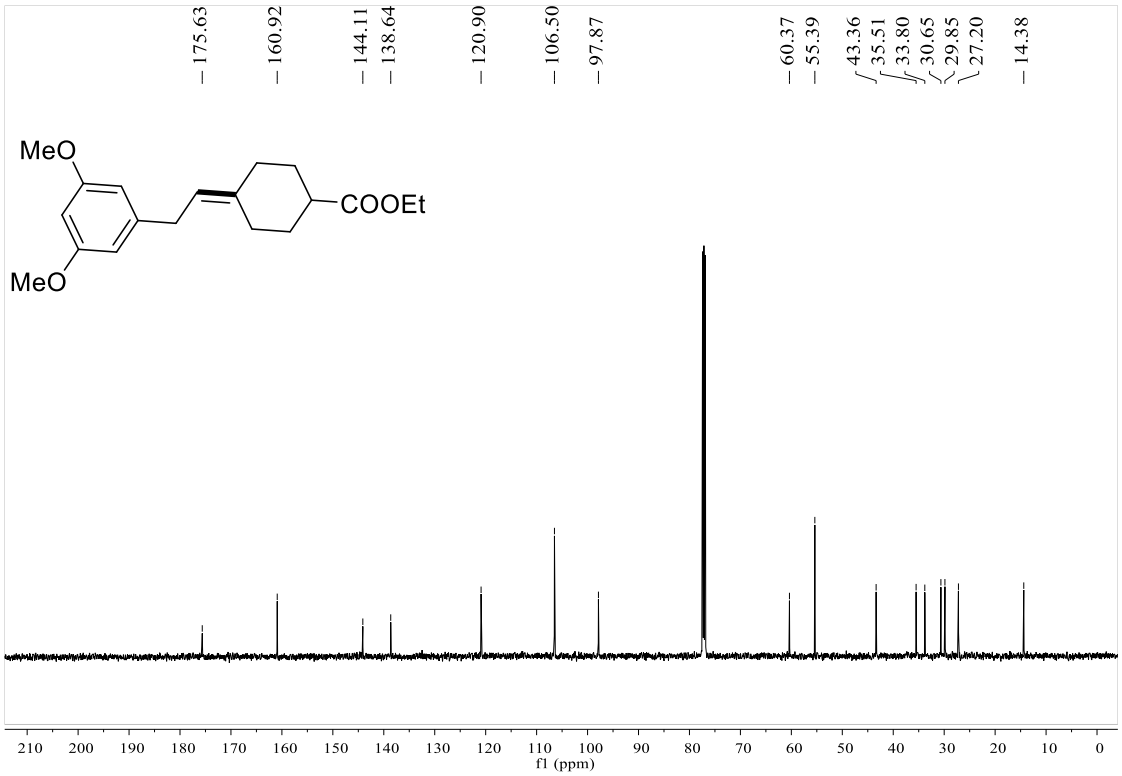


^{19}F NMR spectrum in $CDCl_3$.

ethyl 4-(2-(3,5-dimethoxyphenyl)ethylidene)cyclohexane-1-carboxylate (36c):

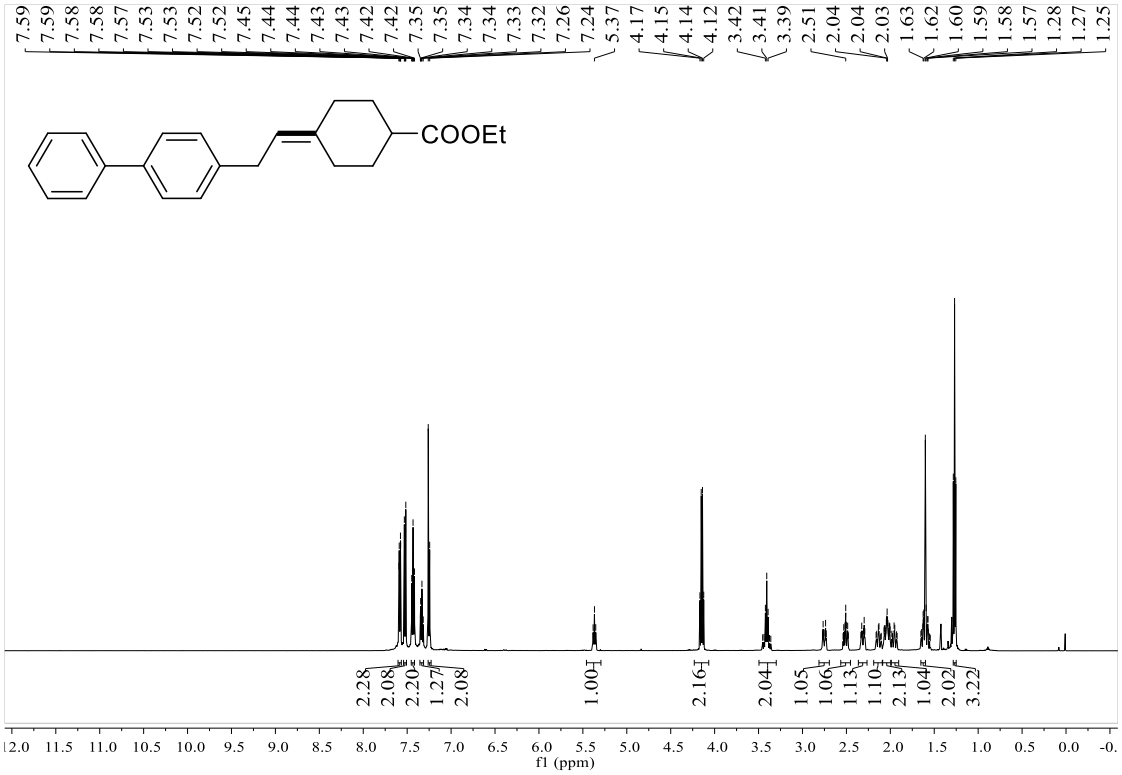


^{1}H NMR spectrum in $CDCl_3$.

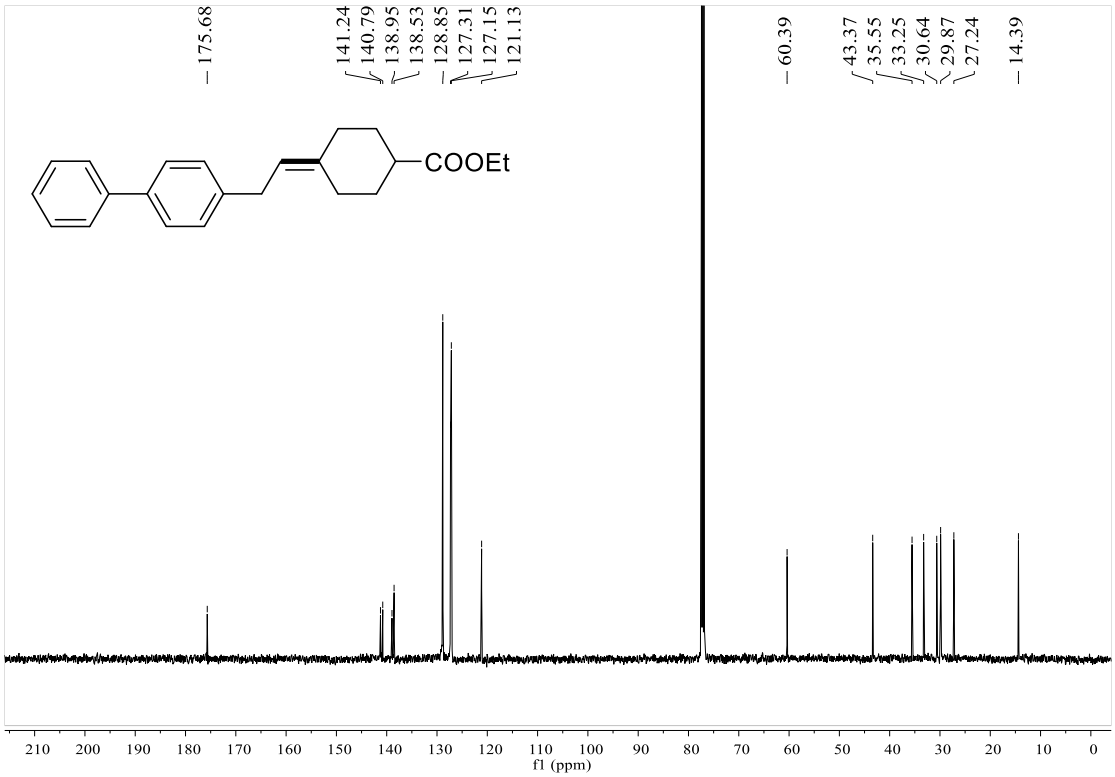


^{13}C NMR spectrum in $CDCl_3$.

methyl 4-(2-([1,1'-biphenyl]-4-yl)ethylidene)cyclohexane-1-carboxylate (37c):

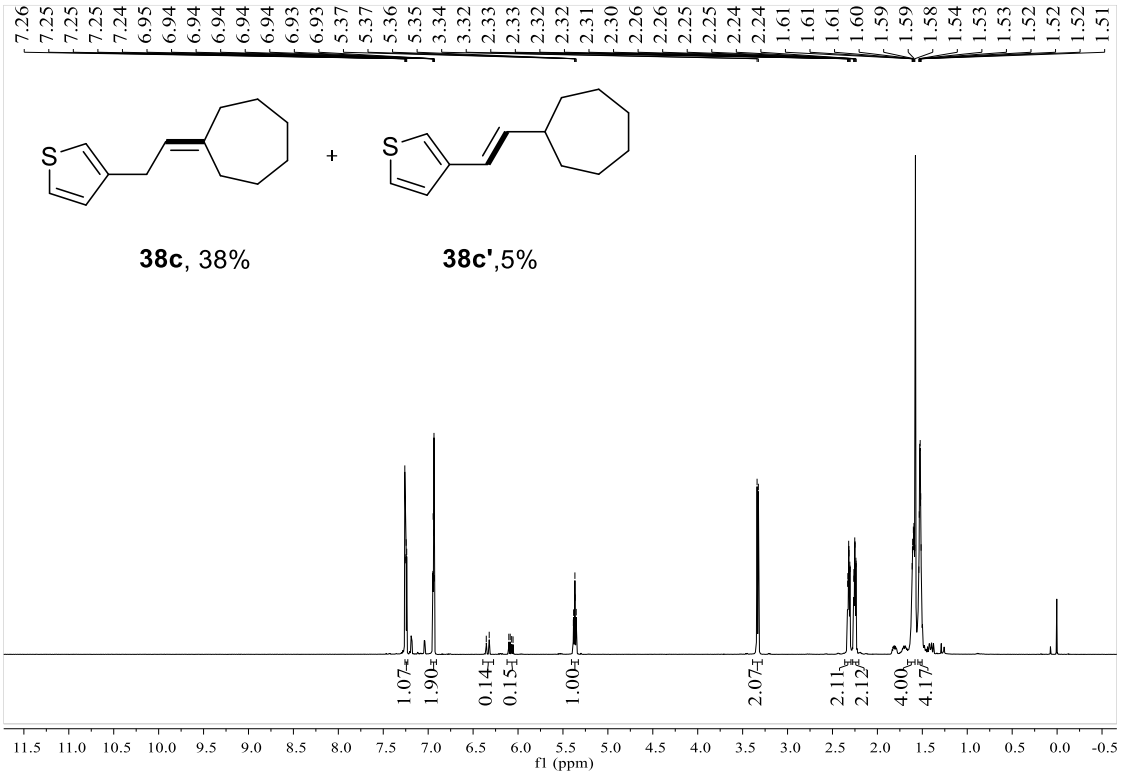


^{1}H NMR spectrum in $CDCl_3$.

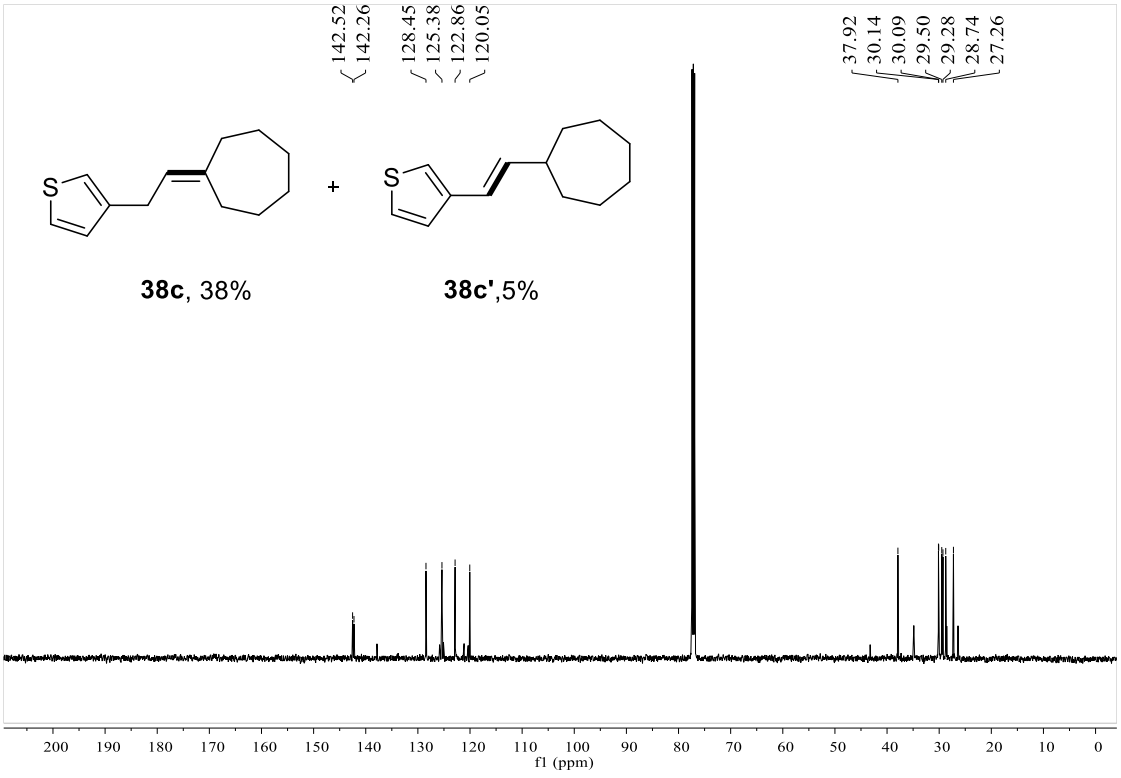


^{13}C NMR spectrum in $CDCl_3$.

3-(2-cycloheptylideneethyl)thiophene (38c):

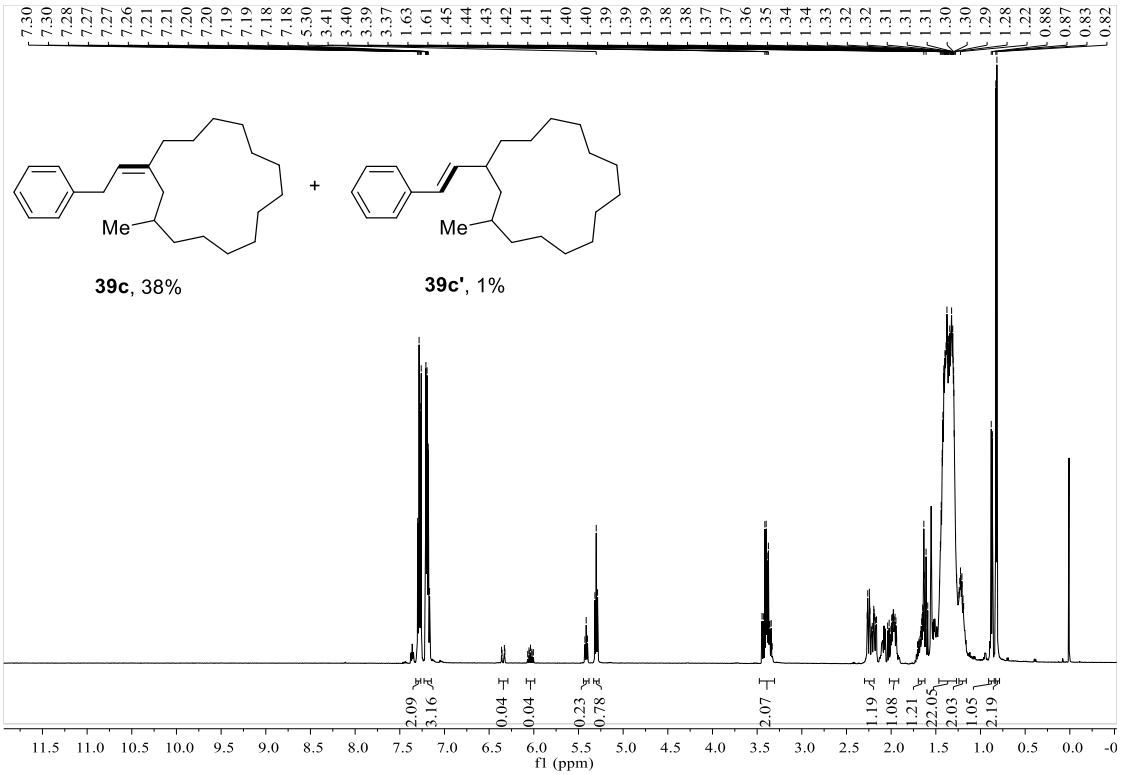


^{1}H NMR spectrum in $CDCl_3$.

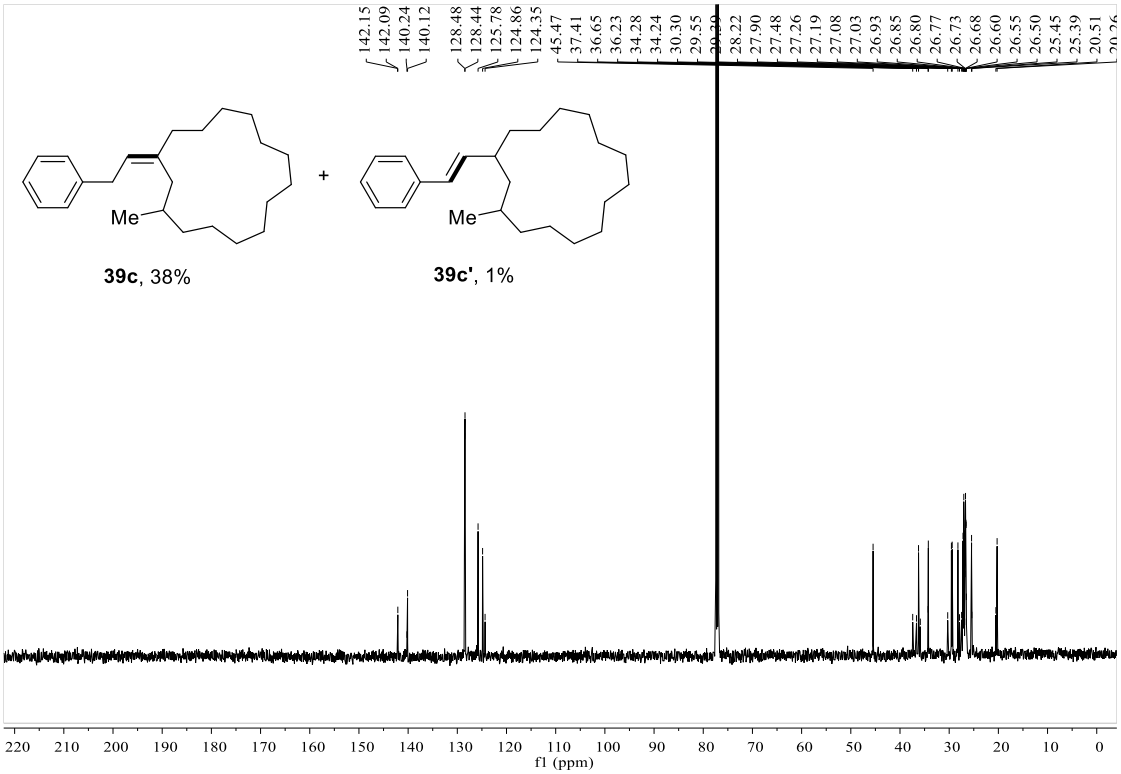


^{13}C NMR spectrum in $CDCl_3$.

1-methyl-3-(2-phenylethylidene)cyclopentadecane (39c):

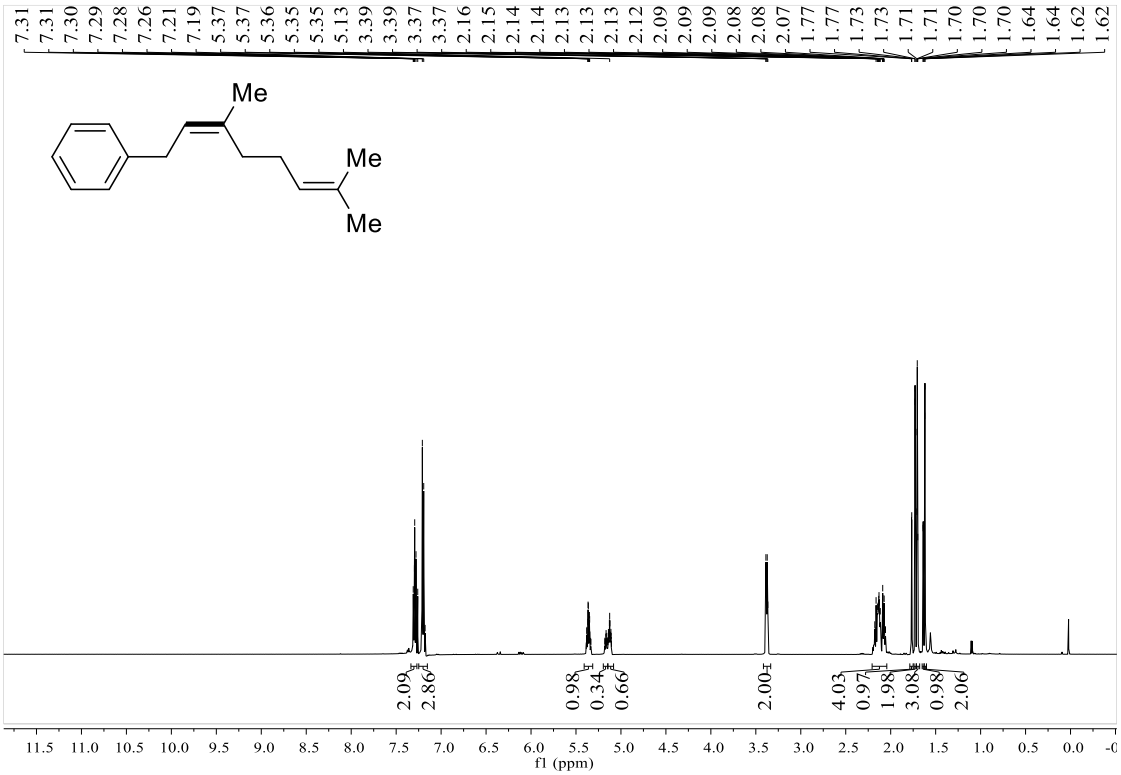


^{1}H NMR spectrum in $CDCl_3$.

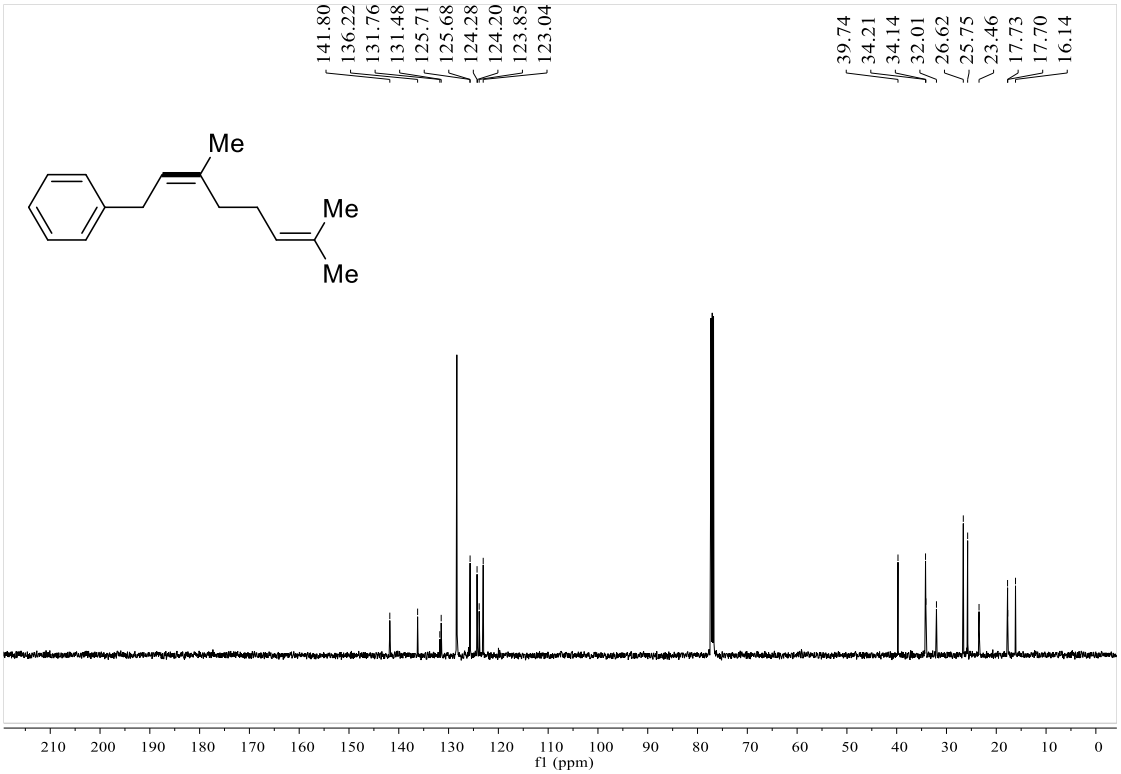


^{13}C NMR spectrum in $CDCl_3$.

(3,7-dimethylocta-2,6-dien-1-yl)benzene (40):

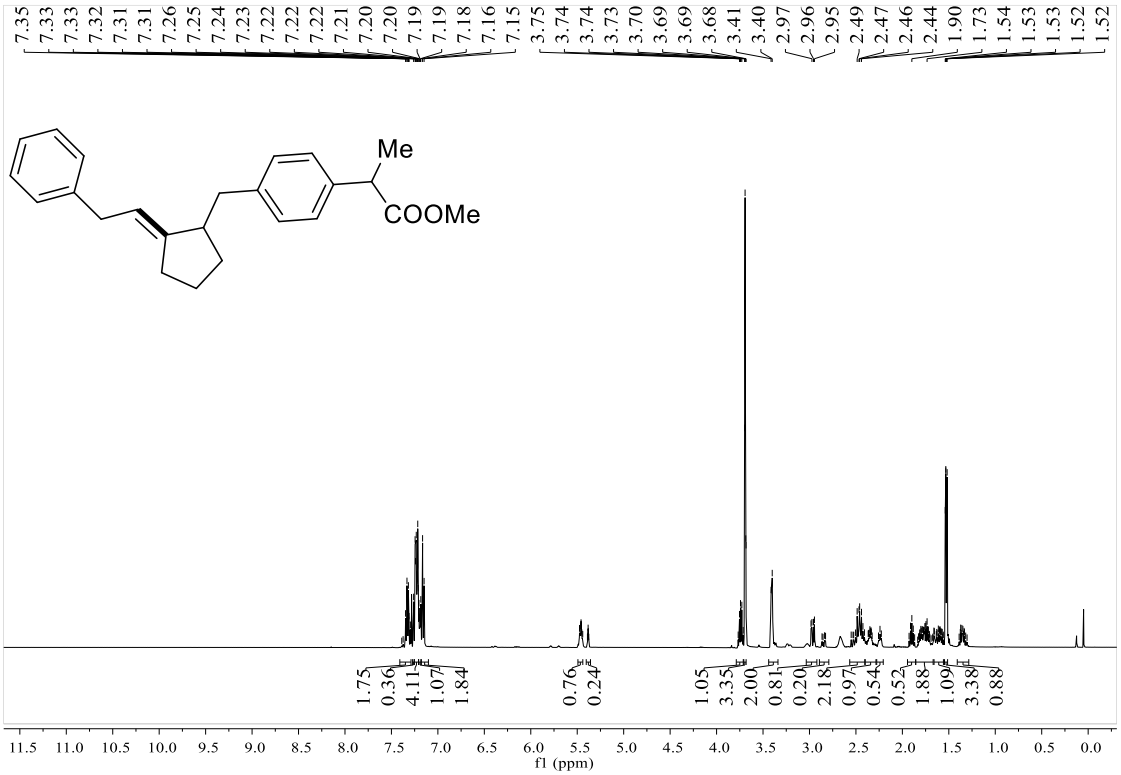


^{1}H NMR spectrum in $CDCl_3$.

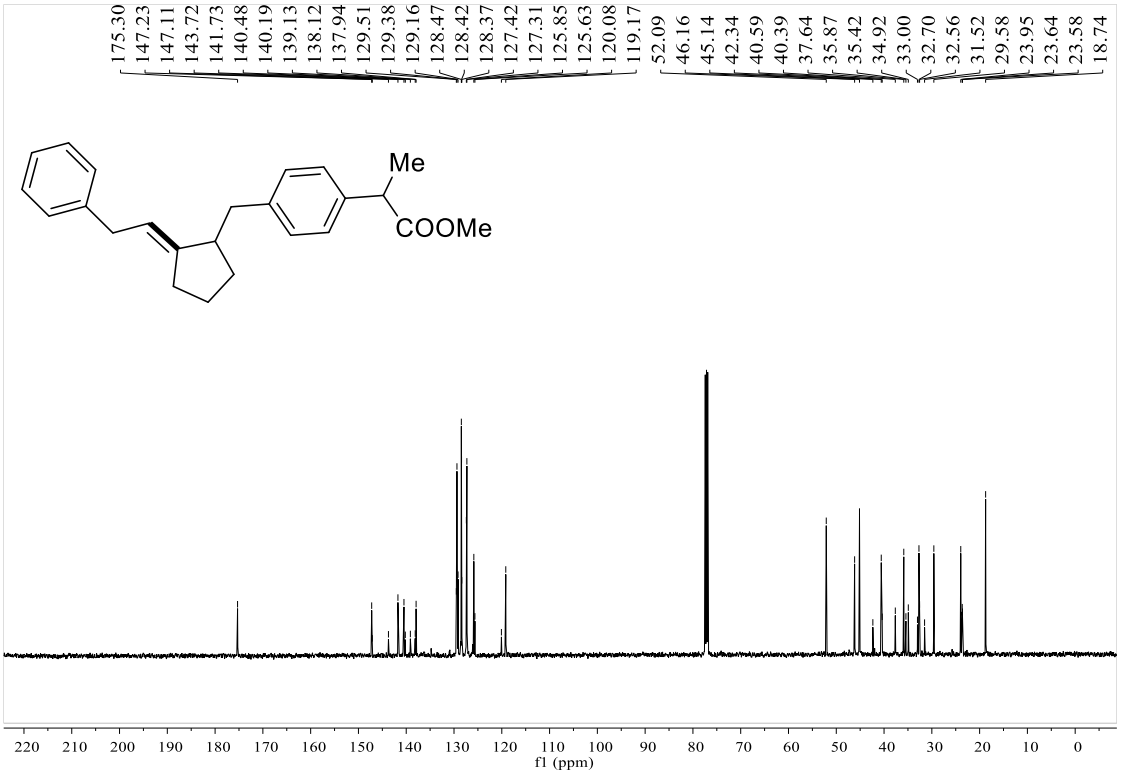


^{13}C NMR spectrum in $CDCl_3$.

methyl-2-(4-((2-(2-phenylethylidene)cyclopentyl)methyl)phenyl)propanoate (41c):

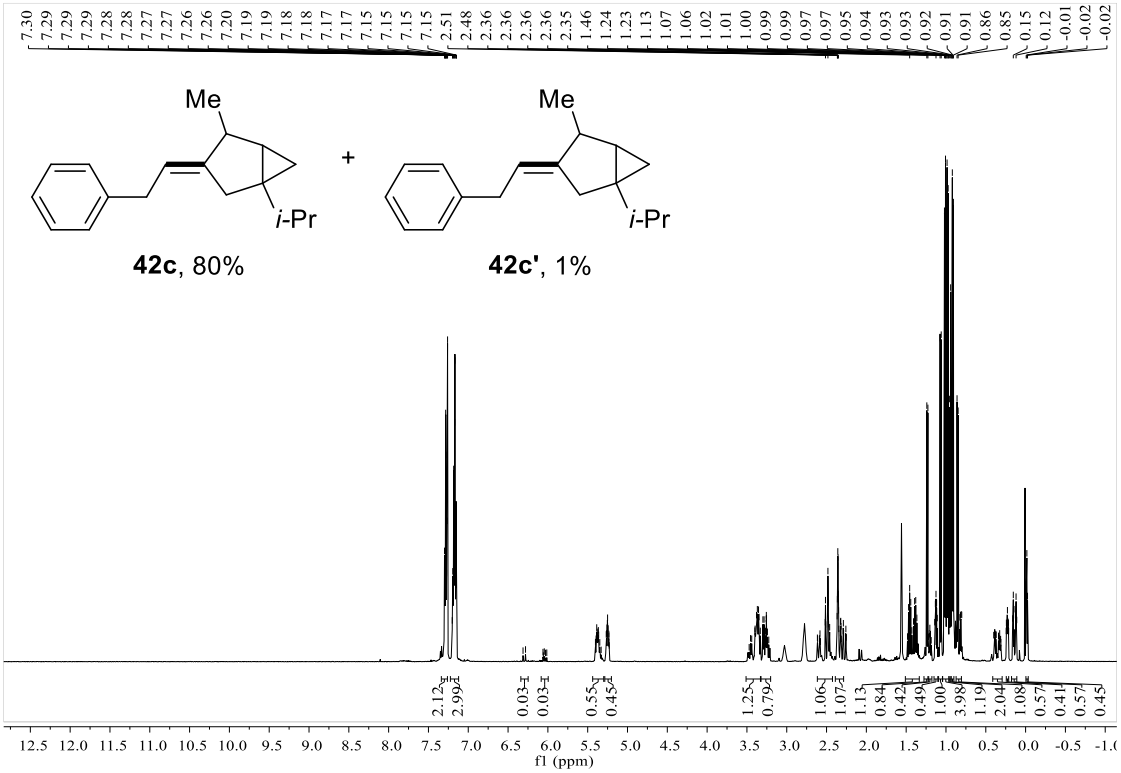


^{1}H NMR spectrum in $CDCl_3$.

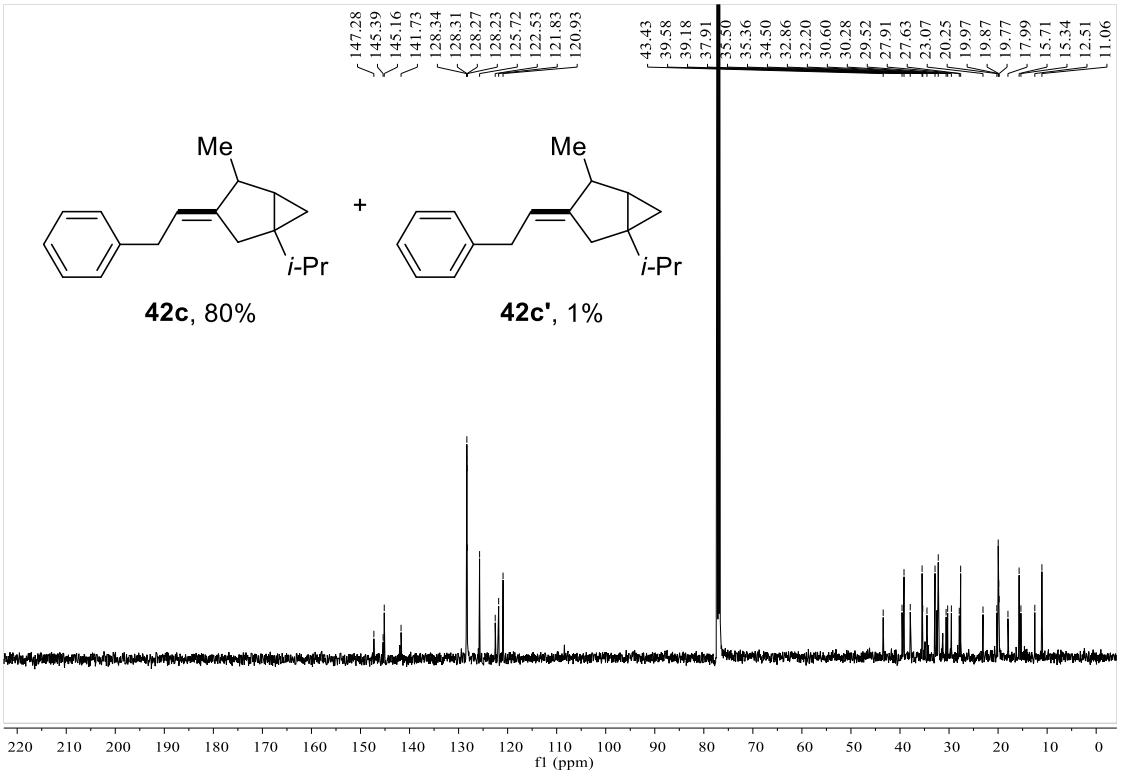


^{13}C NMR spectrum in $CDCl_3$.

1-isopropyl-4-methyl-3-(2-phenylethylidene)bicyclo[3.1.0]hexane (42c):

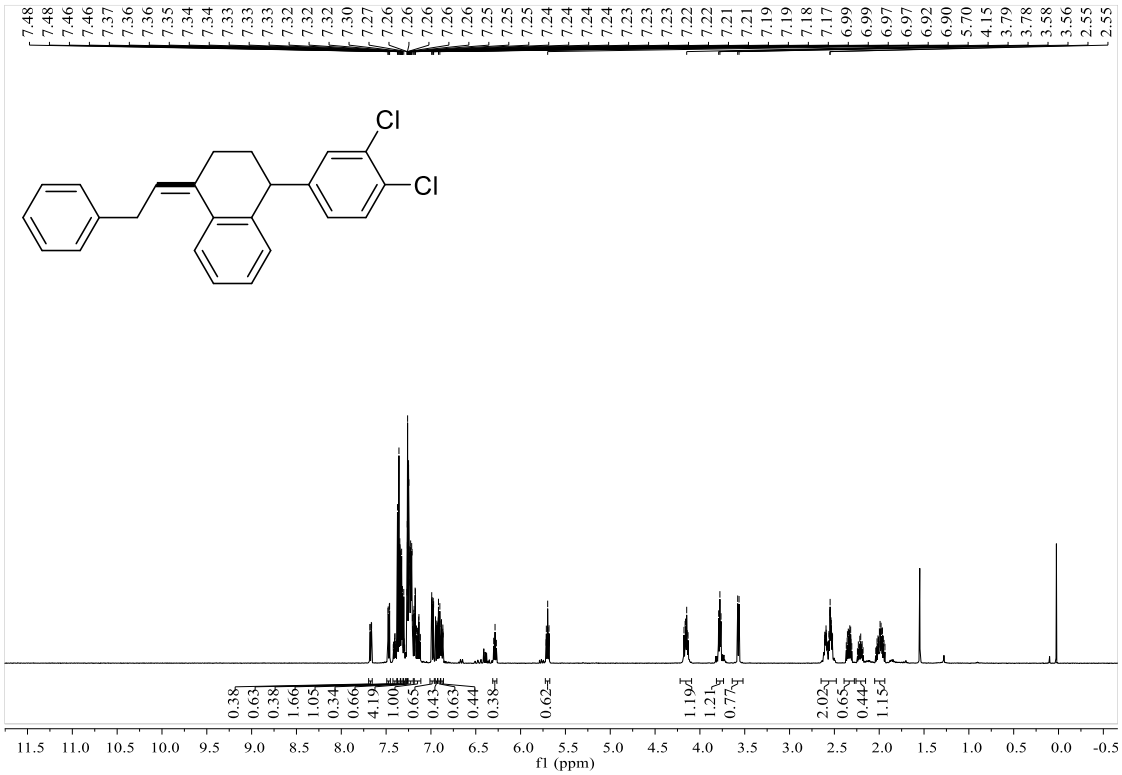


^{1}H NMR spectrum in $CDCl_3$.

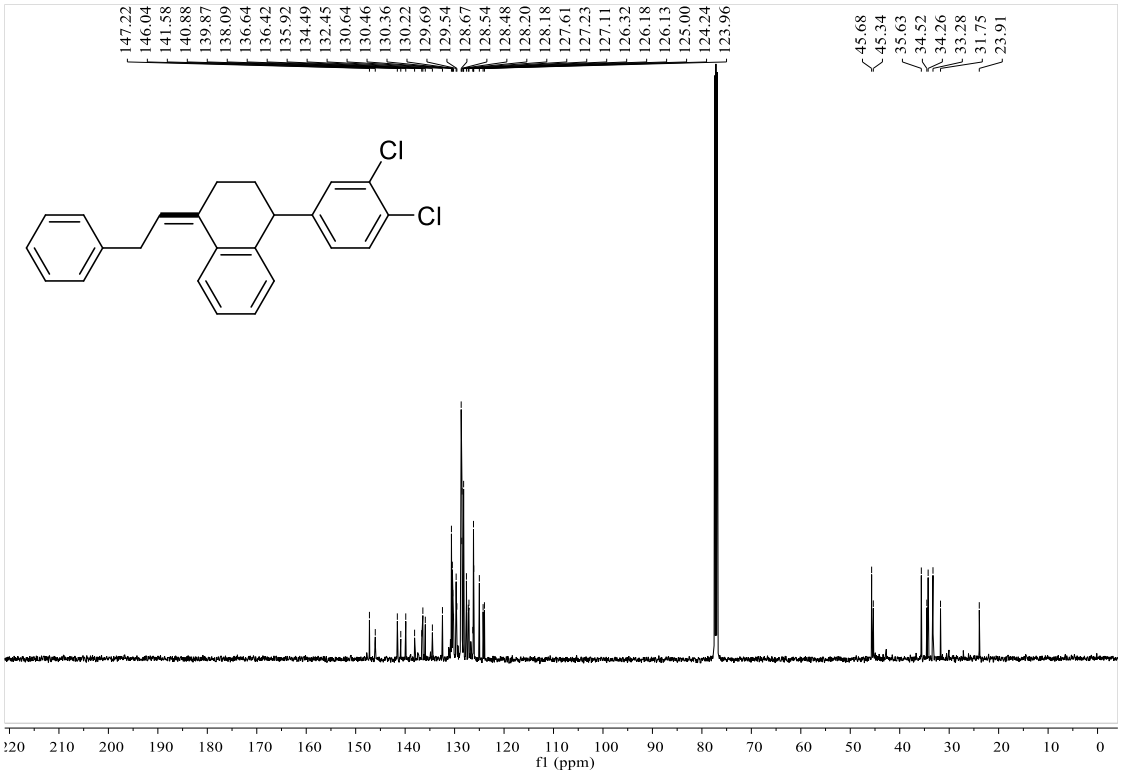


^{13}C NMR spectrum in $CDCl_3$.

1-(3,4-dichlorophenyl)-4-(2-phenylethylidene)-1,2,3,4-tetrahydronaphthalene (43c):

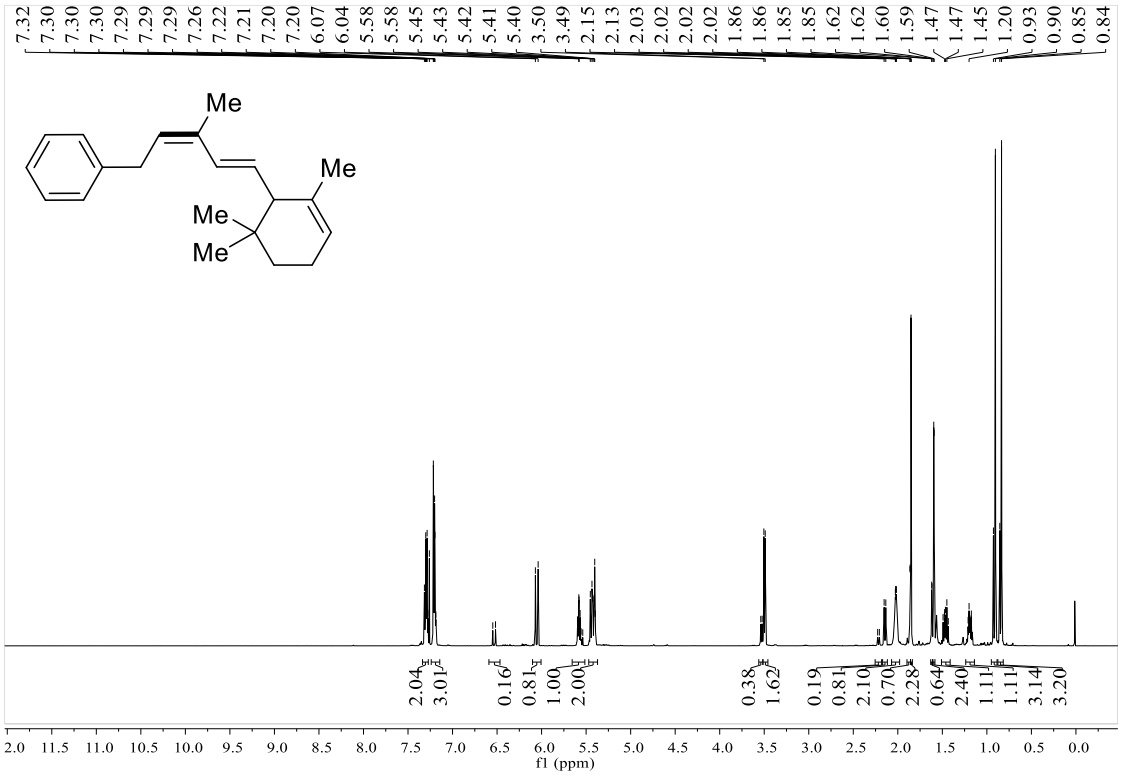


^{1}H NMR spectrum in $CDCl_3$.

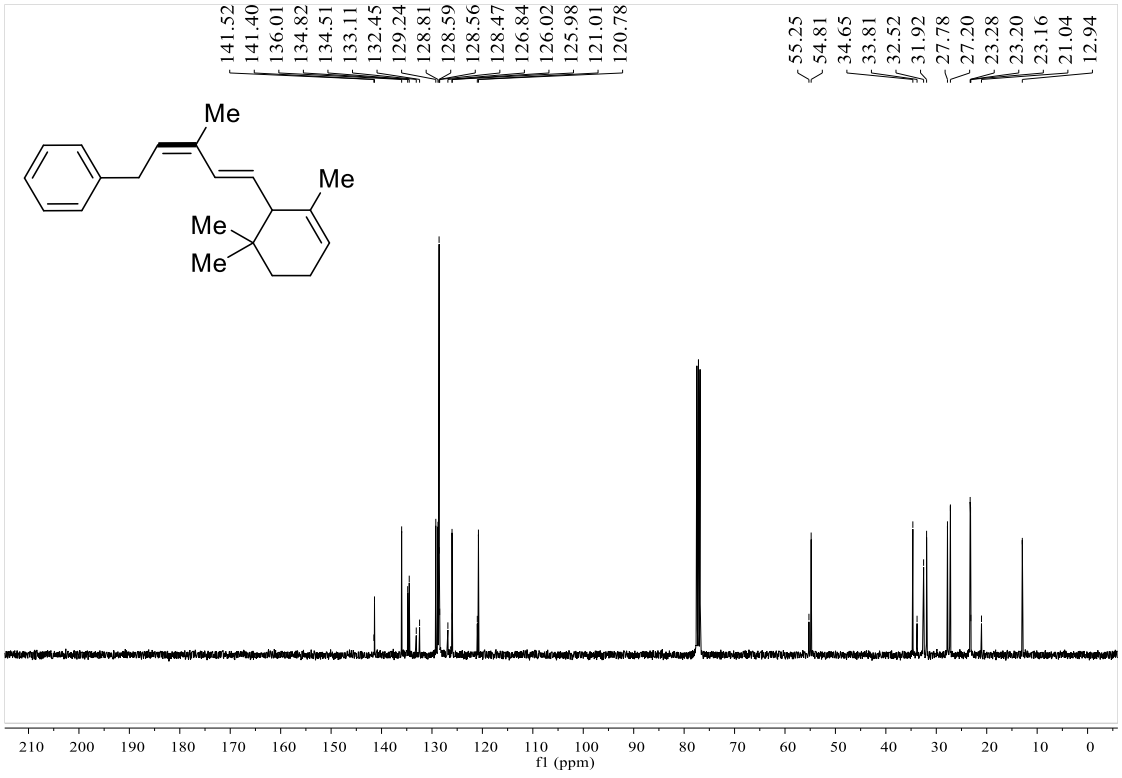


^{13}C NMR spectrum in $CDCl_3$.

3-methyl-5-(2,6,6-trimethylcyclohex-2-en-1-yl)penta-2,4-dien-1-yl)benzene (44c):

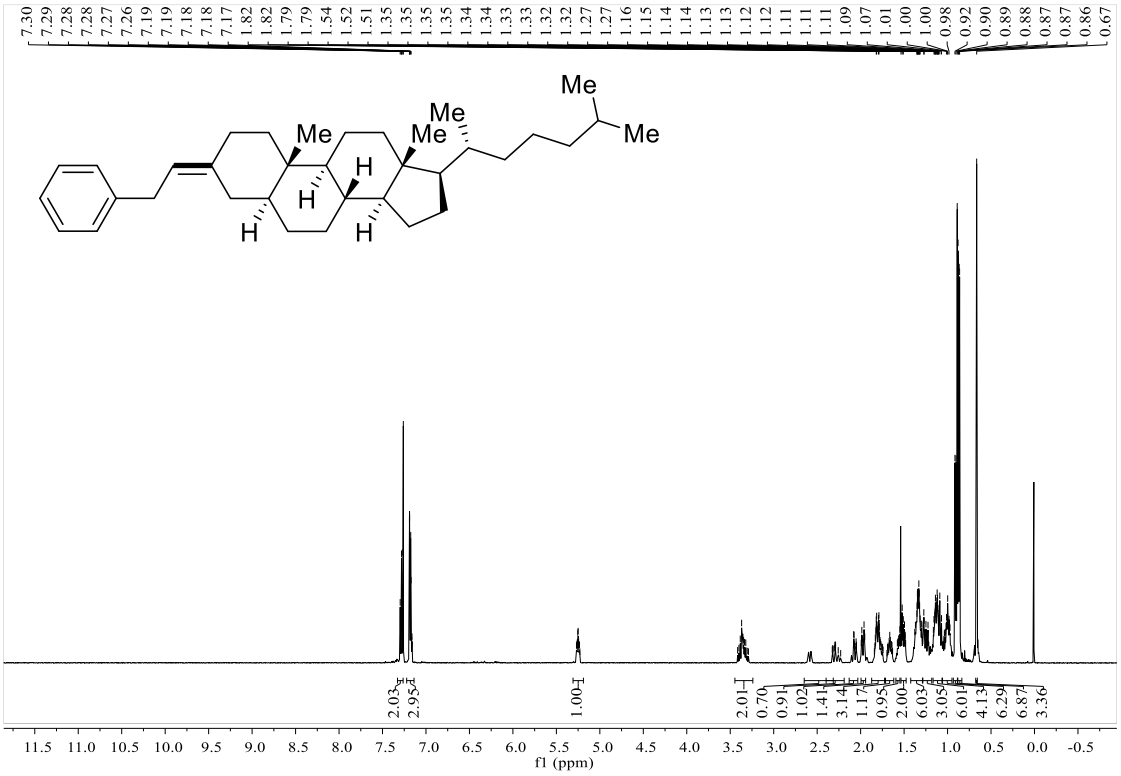


^{1}H NMR spectrum in $CDCl_3$.

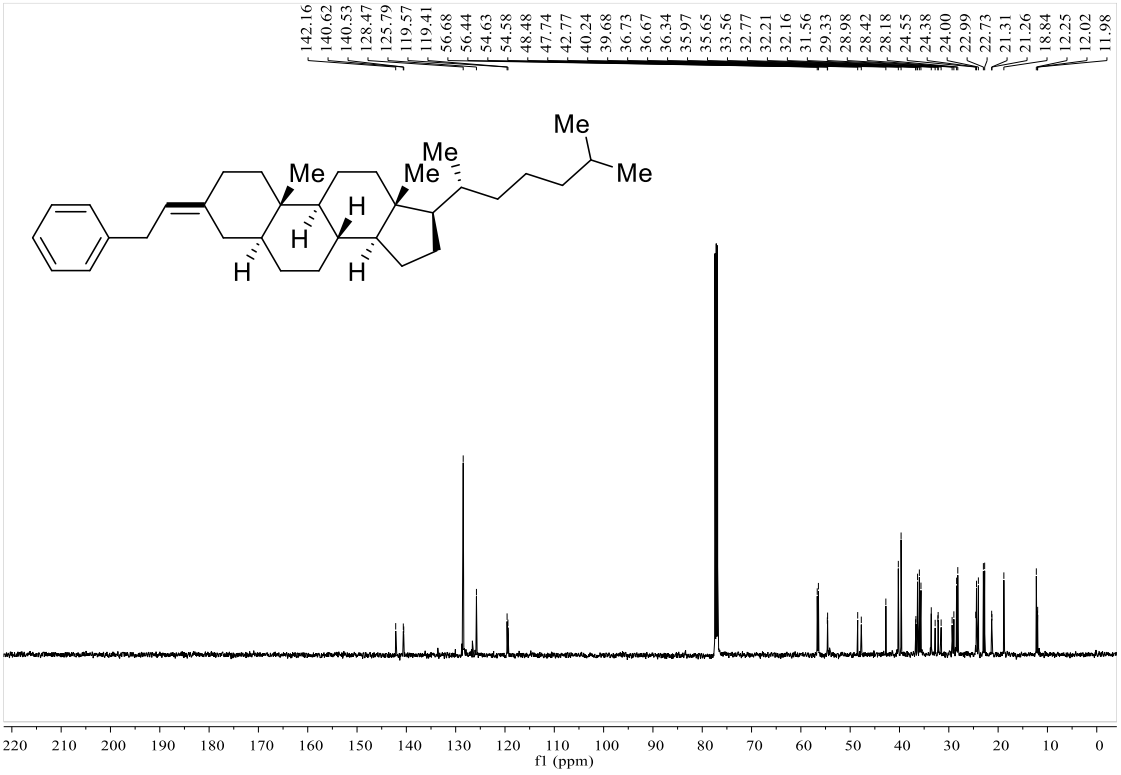


^{13}C NMR spectrum in $CDCl_3$.

(8R,13R)-8,13-dimethyl-17-((R)-6-methylheptan-2-yl)-3-(2-phenylethylidene)hexadecahydro-1H-cyclopenta[a]phenanthrene (45c):

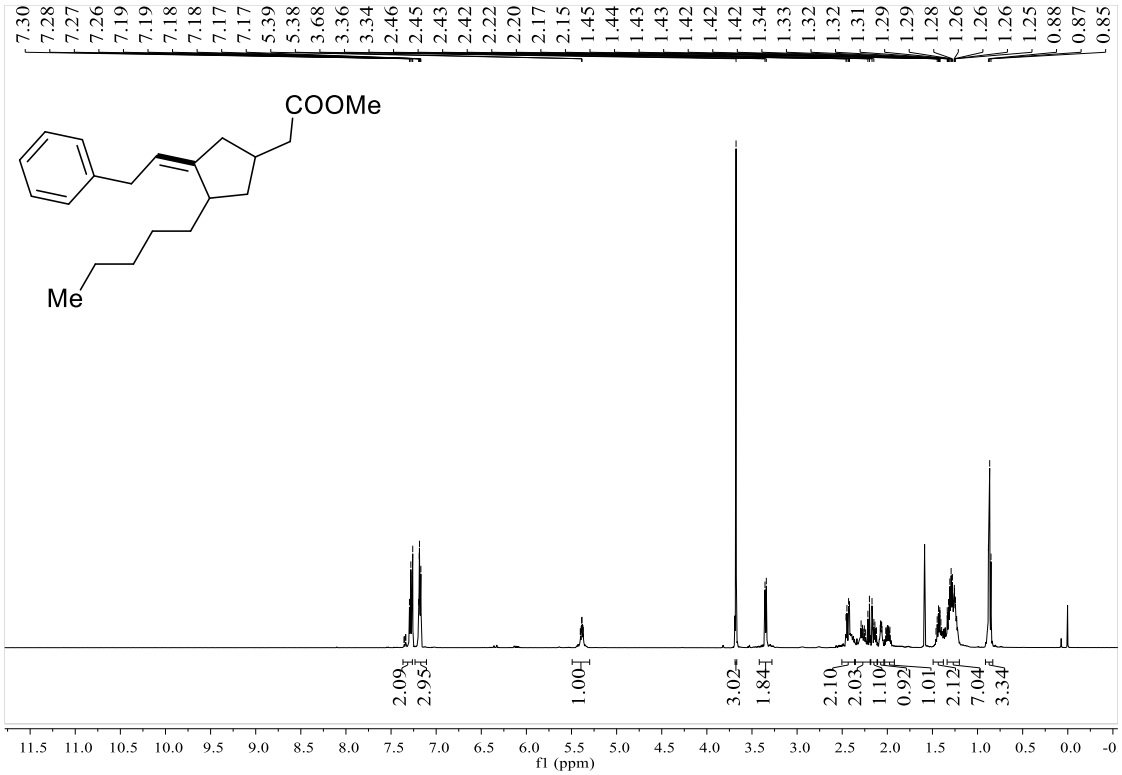


^{1}H NMR spectrum in $CDCl_3$.

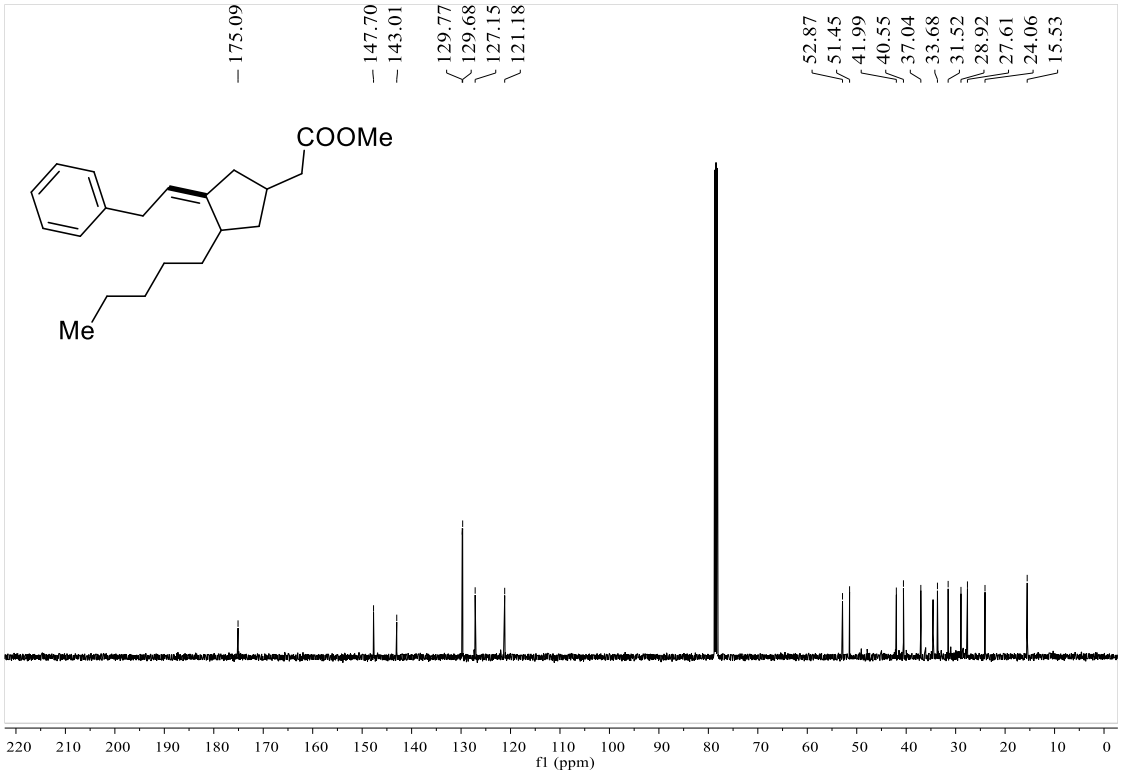


^{13}C NMR spectrum in $CDCl_3$.

methyl-2-(3-pentyl-4-(2-phenylethylidene)cyclopentyl)acetate (46c):

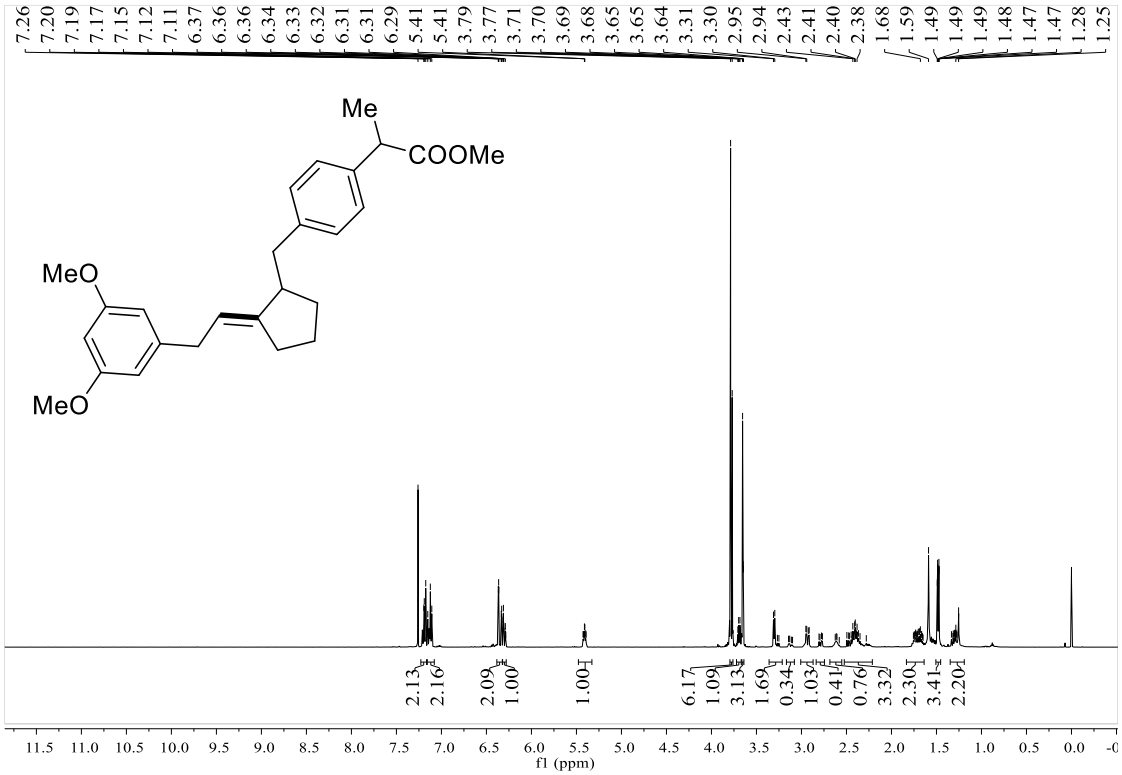


^{1}H NMR spectrum in $CDCl_3$.

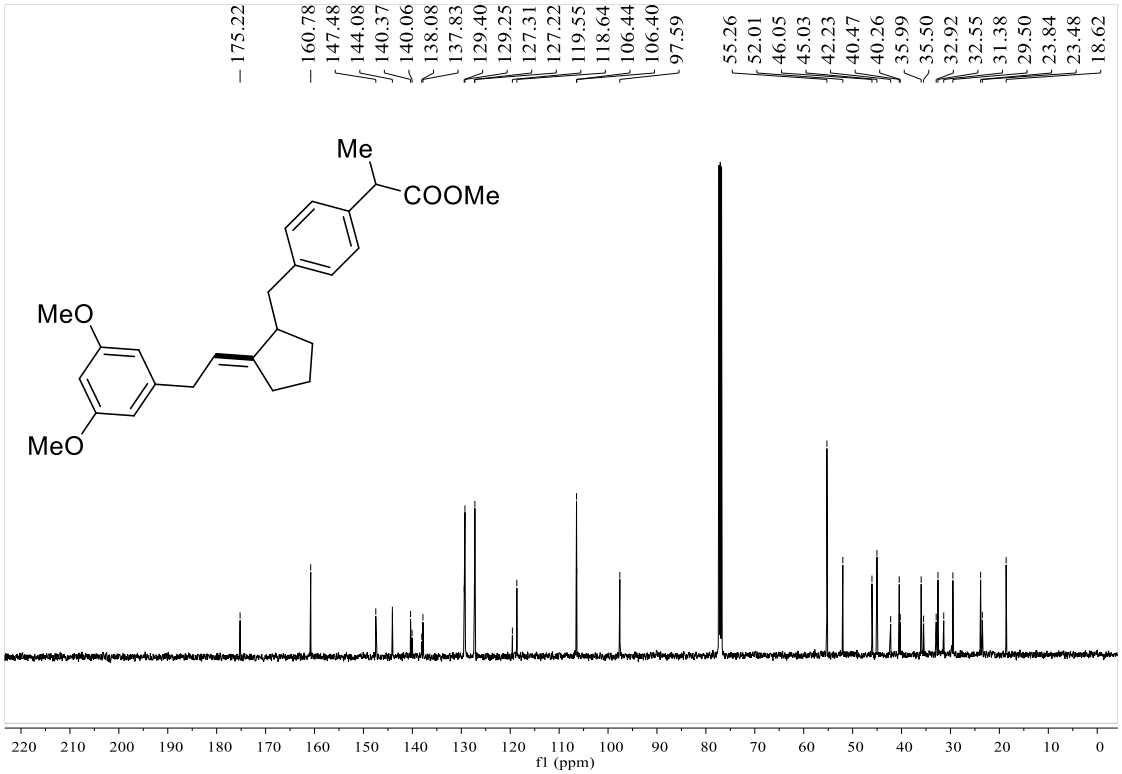


^{13}C NMR spectrum in $CDCl_3$.

methyl-2-(4-((2-(2-(3,5-dimethoxyphenyl)ethylidene)cyclopentyl)methyl)phenyl)propanoate (47c):



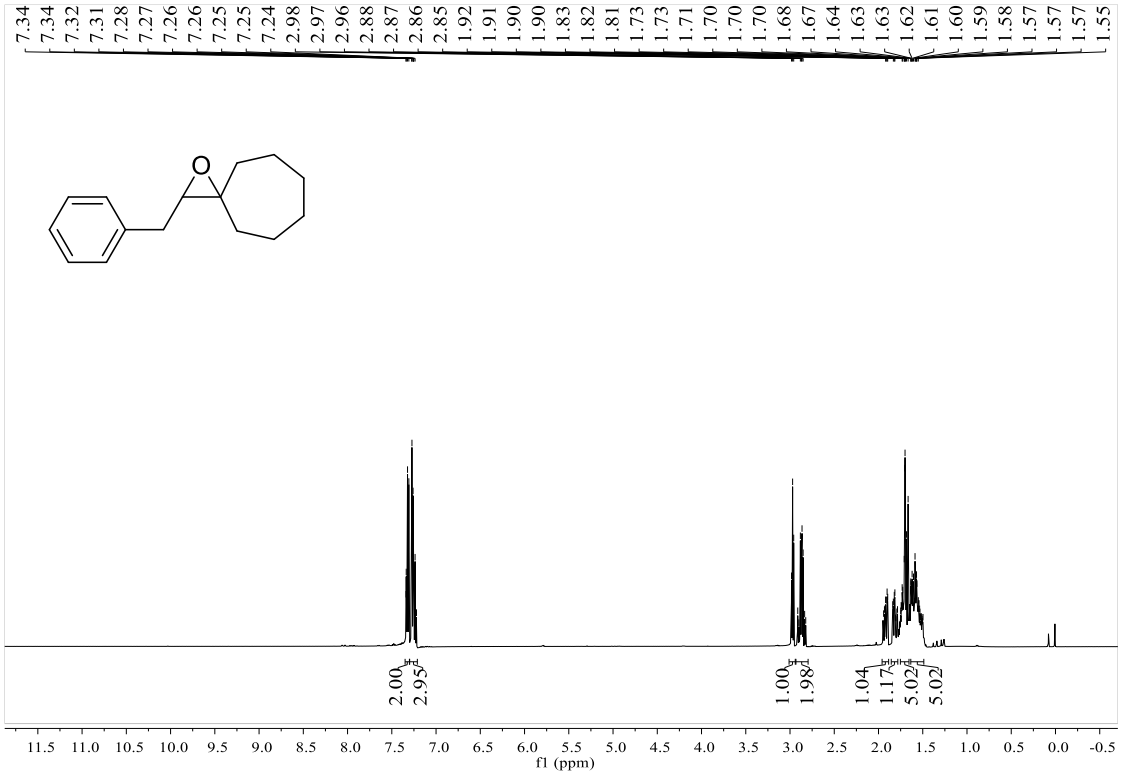
^{1}H NMR spectrum in $CDCl_3$.



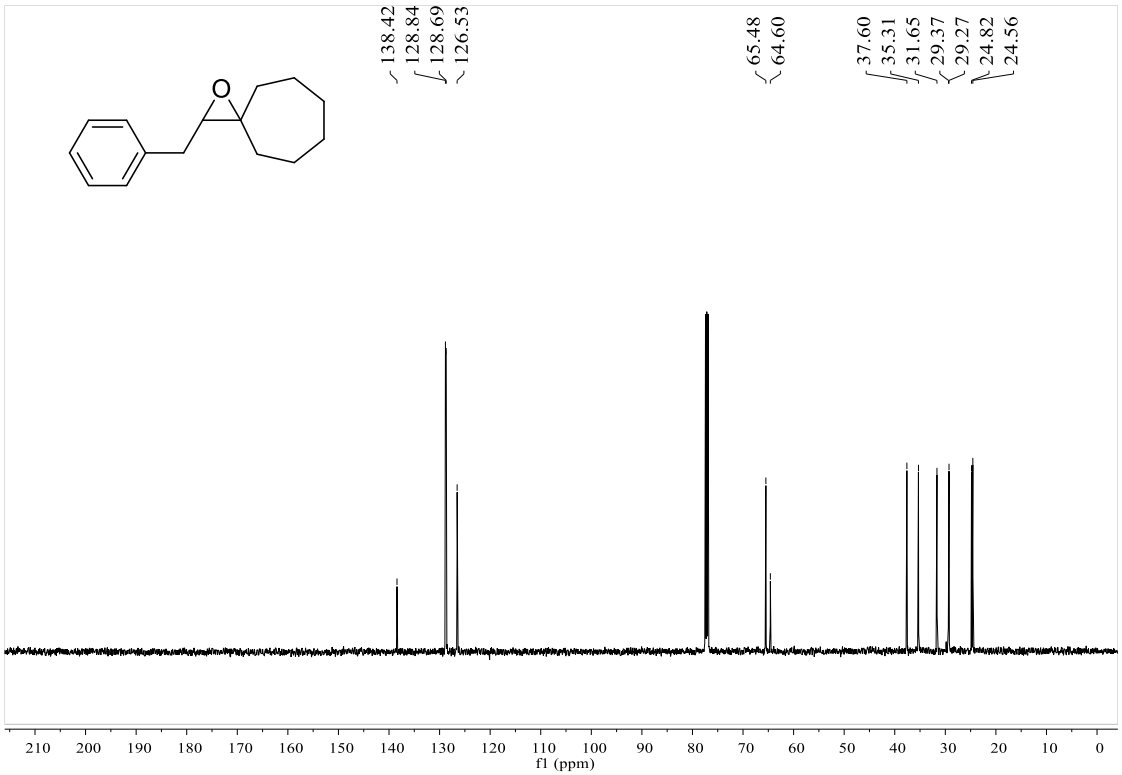
^{13}C NMR spectrum in $CDCl_3$.

9.4 NMR spectra of further transformation products

2-benzyl-1-oxaspiro[2.6]nonane (1d):

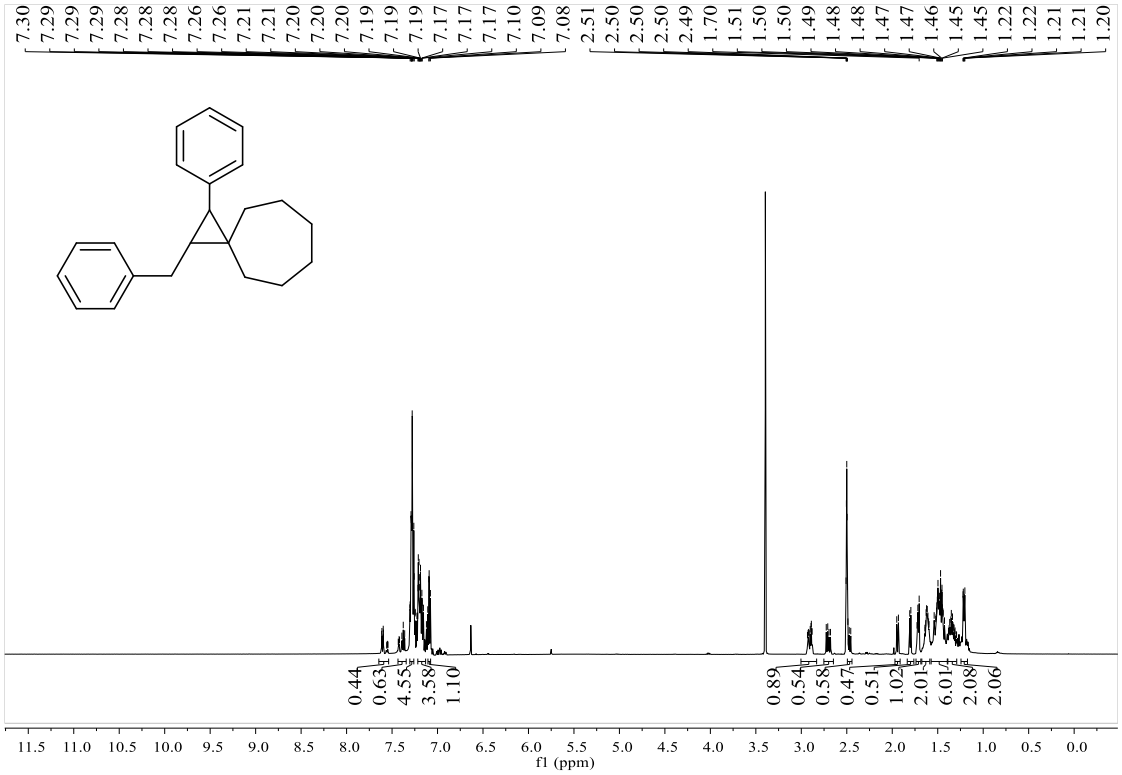


^{1}H NMR spectrum in $CDCl_3$.

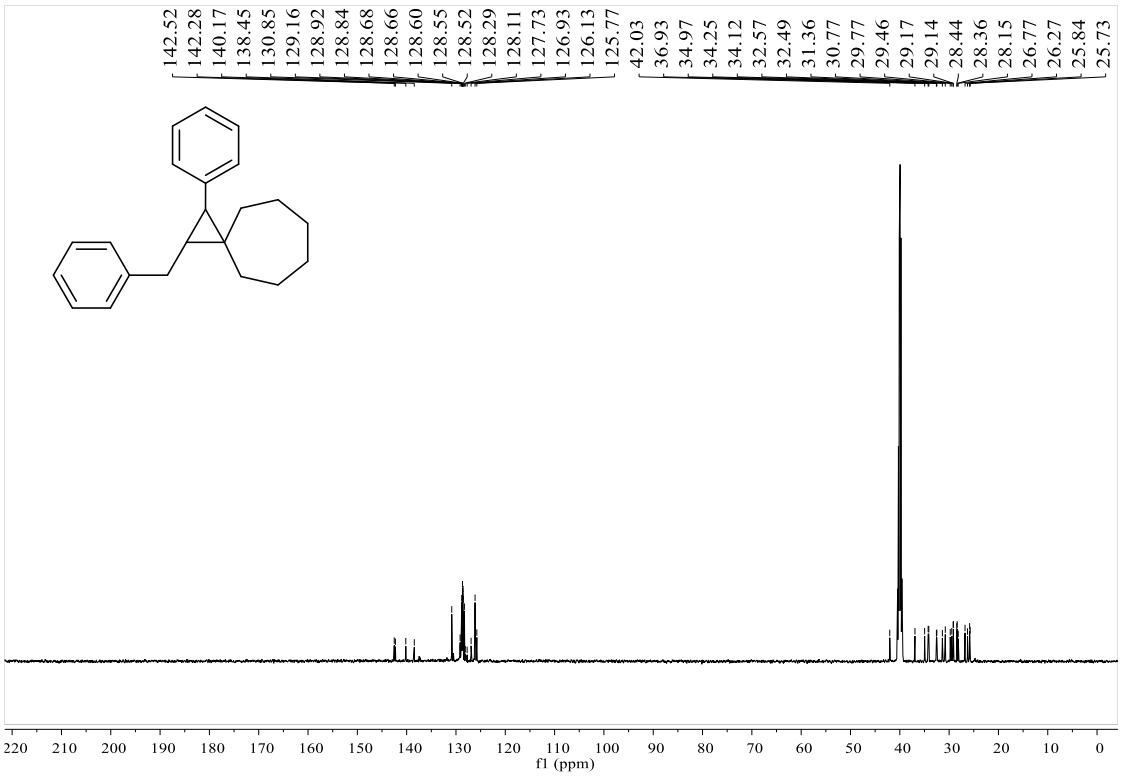


^{13}C NMR spectrum in $CDCl_3$.

1-benzyl-2-phenylspiro[2.6]nonane (1e):

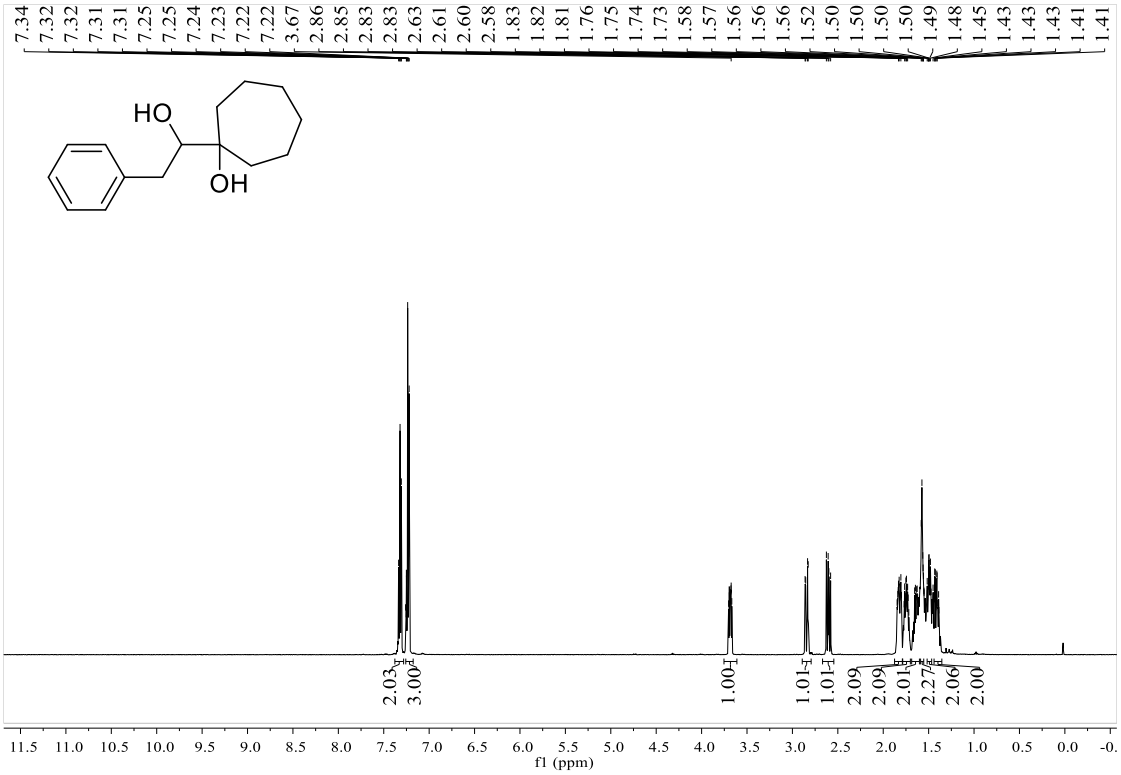


^{1}H NMR spectrum in DMSO-d_6.

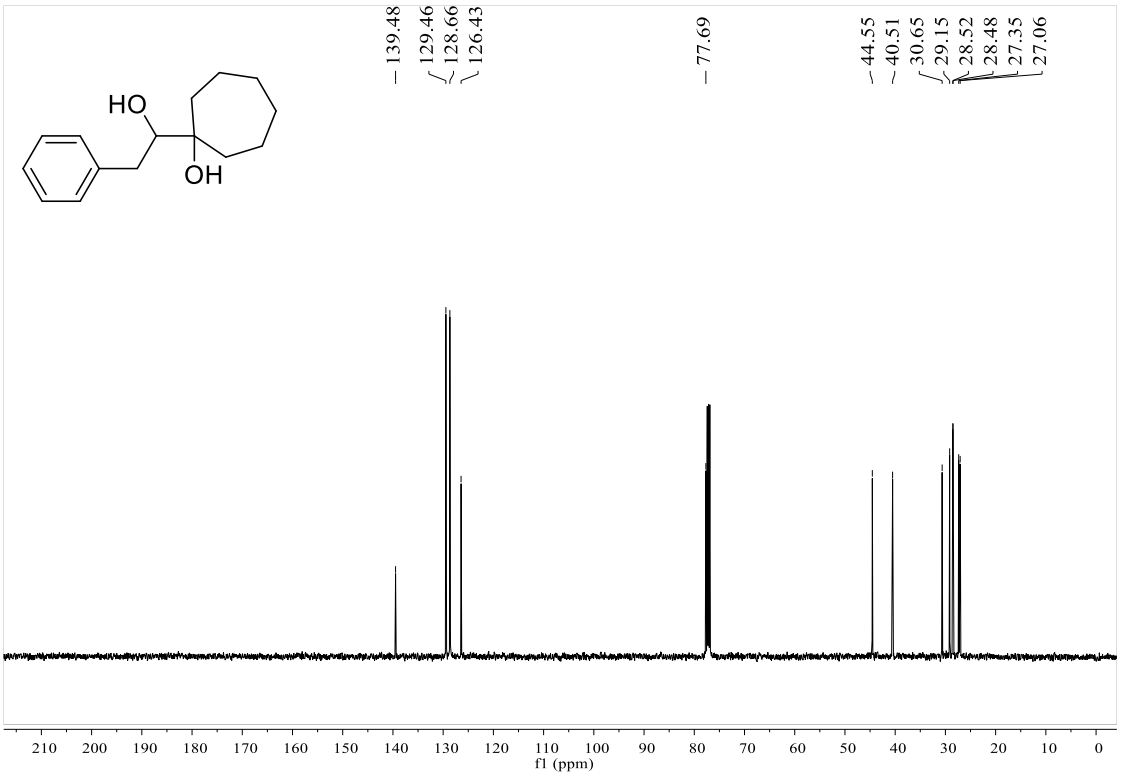


^{13}C NMR spectrum in DMSO-d_6.

1-(1-hydroxy-2-phenylethyl)cycloheptan-1-ol (1f):

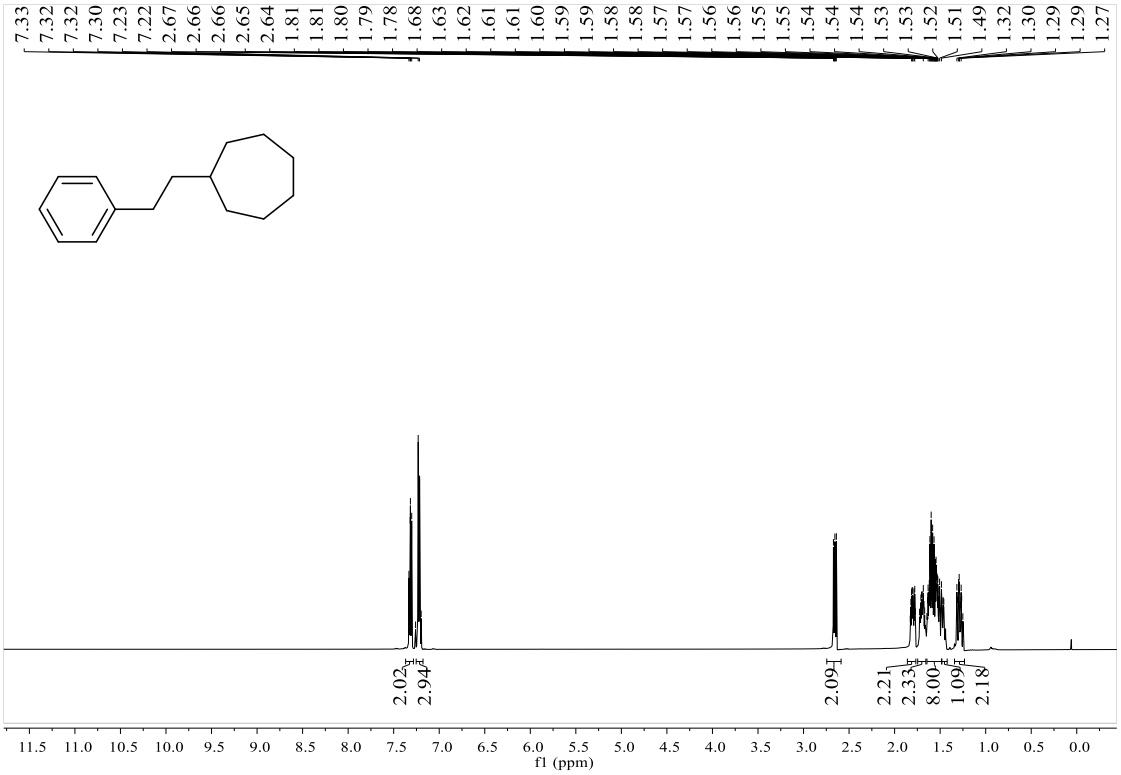


^{1}H NMR spectrum in $CDCl_3$.

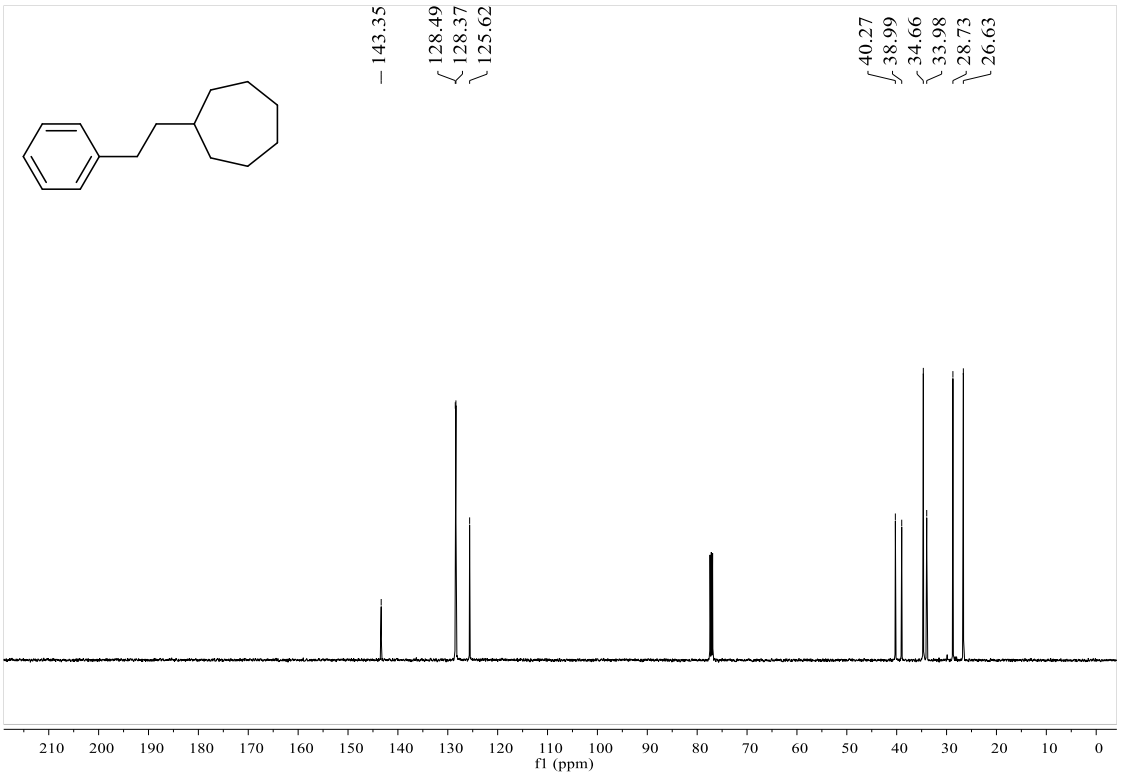


^{13}C NMR spectrum in $CDCl_3$.

phenethylcycloheptane (1g):

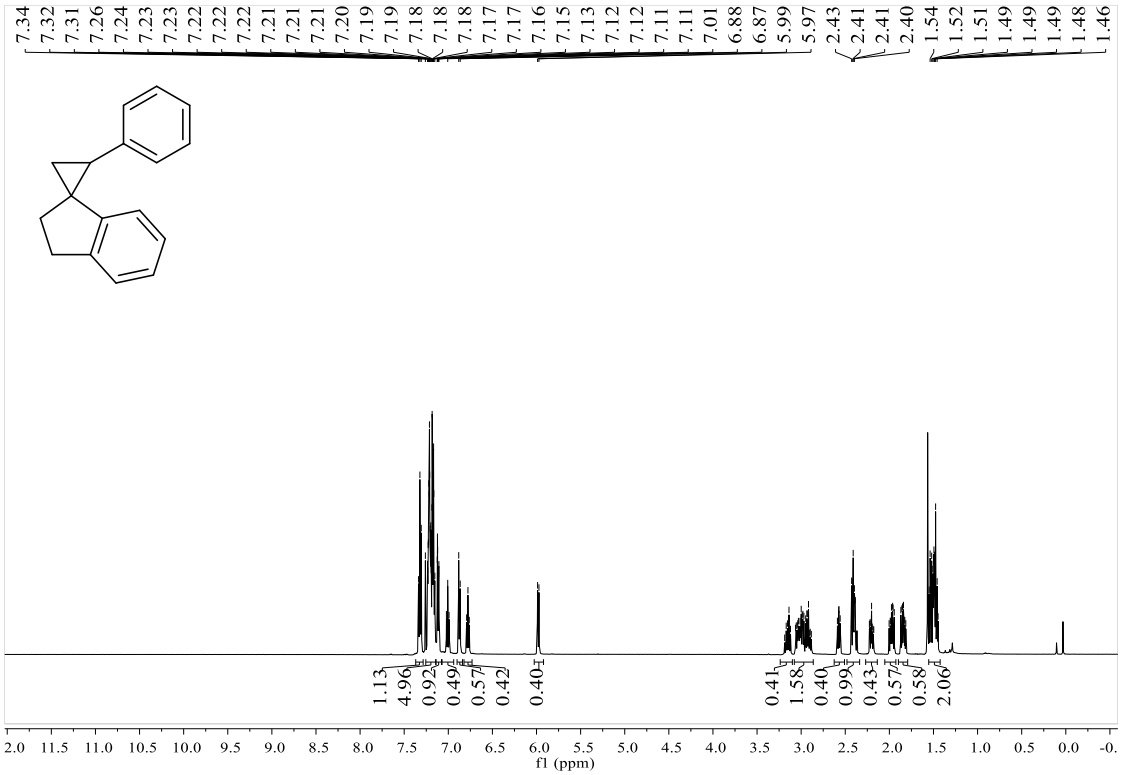


^{1}H NMR spectrum in $CDCl_3$.

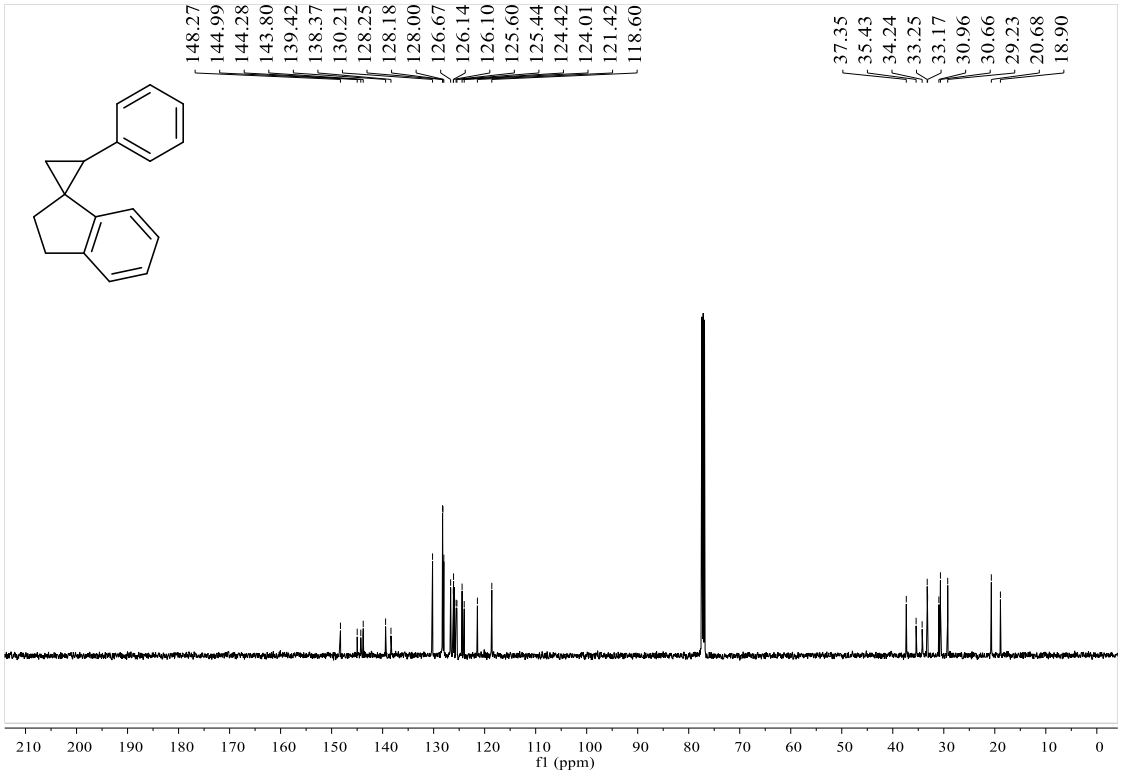


^{13}C NMR spectrum in $CDCl_3$.

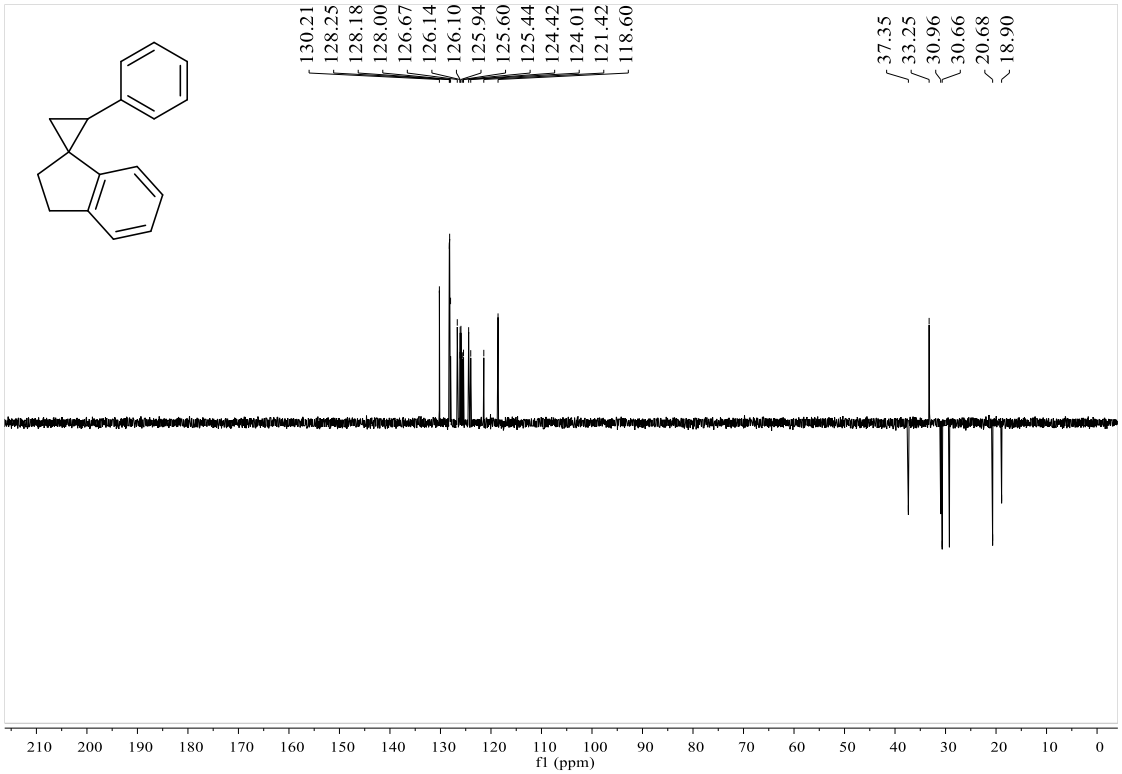
2-phenyl-2',3'-dihydrospiro[cyclopropane-1,1'-indene] (13d):



^{1}H NMR spectrum in $CDCl_3$.

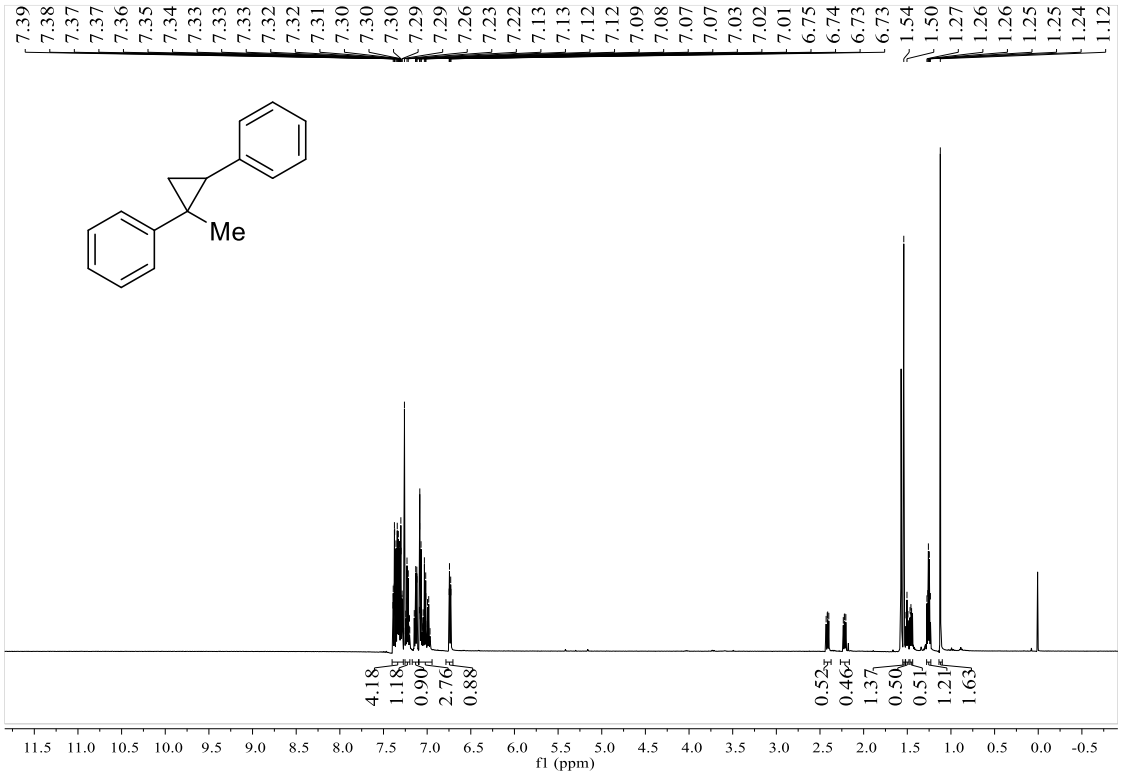


^{13}C NMR spectrum in $CDCl_3$.

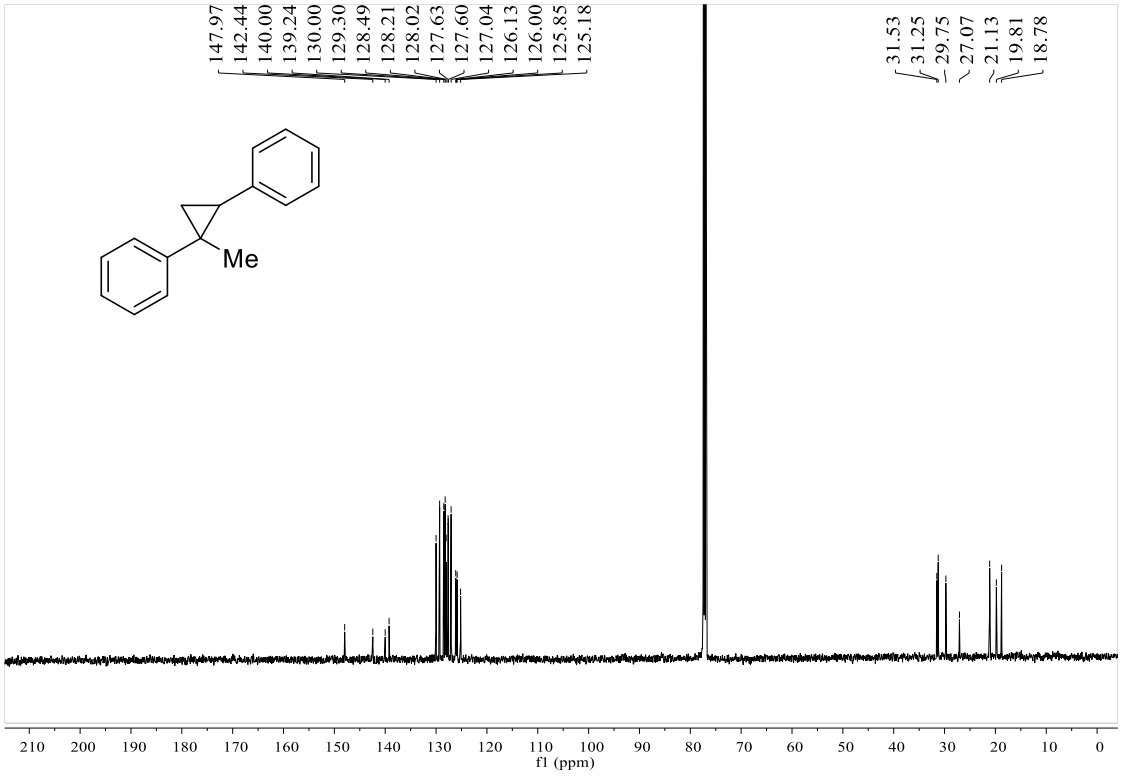


DEPT 135 spectrum in $CDCl_3$.

methylcyclopropane-1,2-diyl)dibenzene (25d):

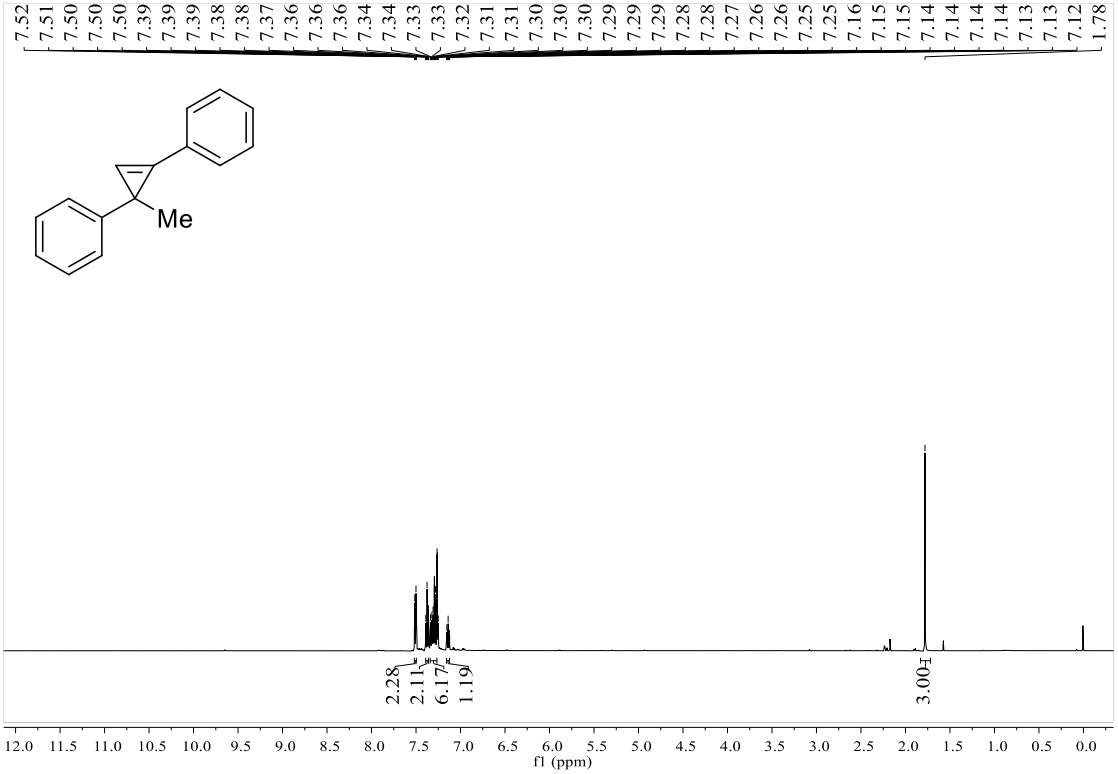


^{1}H NMR spectrum in $CDCl_3$.

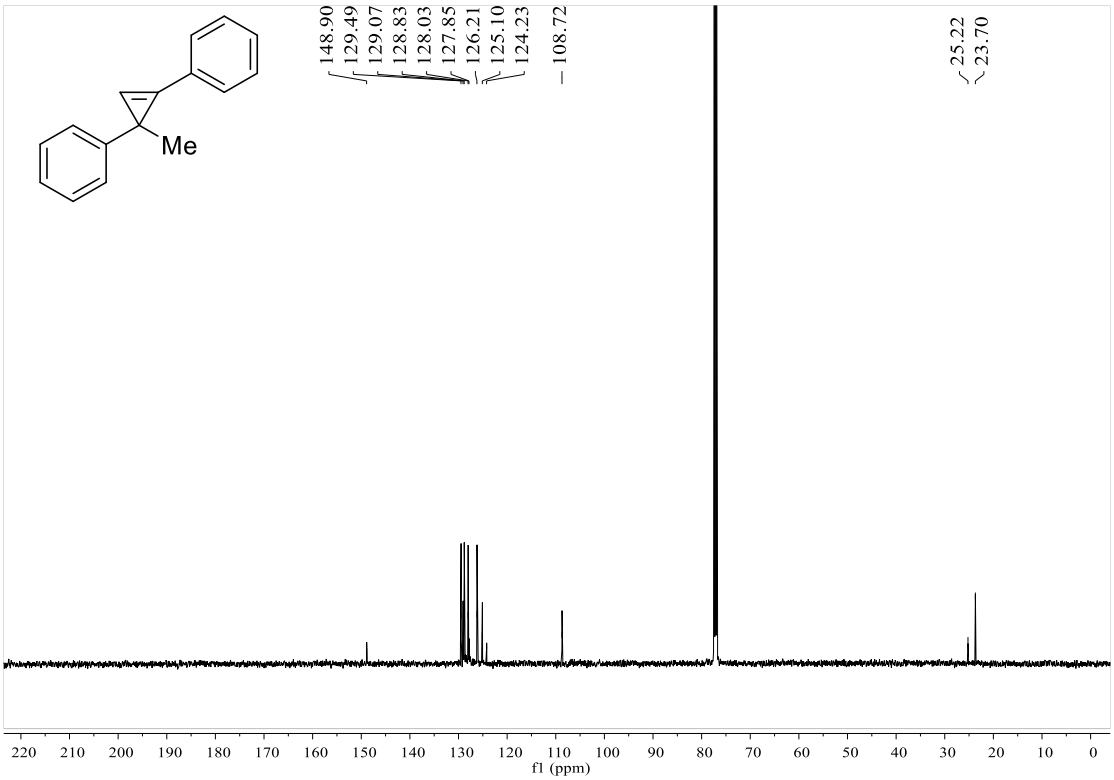


^{13}C NMR spectrum in $CDCl_3$.

Methylcycloprop-2-ene-1,2-diyl)dibenzene (25e)

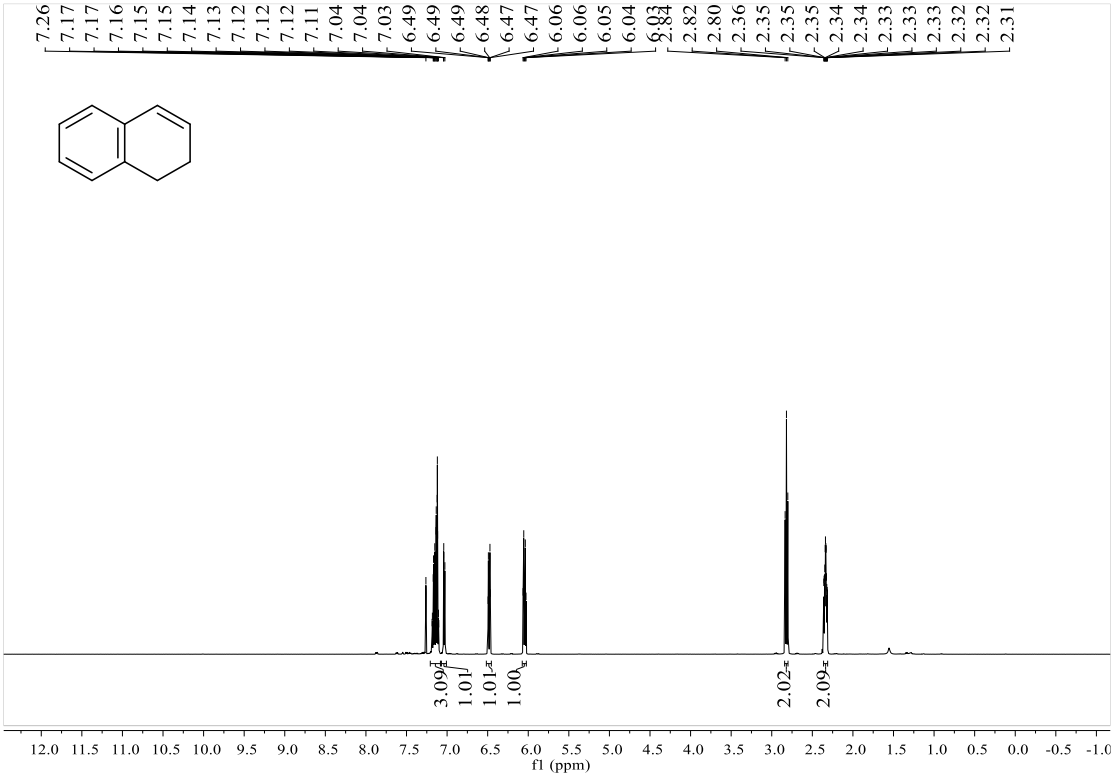


^{1}H NMR spectrum in $CDCl_3$.



^{13}C NMR spectrum in $CDCl_3$.

1,2-Dihydronaphthalene (14d):



^{1}H NMR spectrum in $CDCl_3$.

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