Supporting Information for

Highly Efficient Asymmetric Synthesis of Quaternary Stereocenter-Containing Indolizidine and Quinolizidine Alkaloids Using Aldehydes, Nitroalkenes, and Unactivated Cyclic Ketimines

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Table of Contents

1. General Experimental Methods:

NMR spectra were all recorded on a Bruke (400M Hz) spectrometer. Nuclear magnetic resonance (NMR) spectra are recorded in parts per million from internal TMS on the δ scale. Data for ^{1}H NMR are reported as follows: chemical shift (δppm), multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet), integration, coupling constant (Hz) and assignment. Data for ^{13}C NMR are reported in terms of chemical shift and no special nomenclature is used for equivalent carbons. Flash column chromatography was performed using silica gel (300–400 mesh). Analytical thin–layer chromatography was performed using glass plates pre-coated with 0.25 mm 300–400 mesh silica gel impregnated with a fluorescent indicator (254 nm). Thin layer chromatography plates were visualized by exposure to ultraviolet light. Commercial reagents and solvents were used as received. Catalyst ***S-1*** was purchased from Aldrich and used as received. The nitroalkenes **2**[1] and ketimines **3**[2] were synthesized using known literature procedures. Enantioselectivities were determined by high-performance liquid chromatography (HPLC) with a Jasco uv-2075 plus intelligent uv/ivs detector (λ = 254 nm) and a Phenomenex Lux5u Amylose-2 column or Daicel OD-H column. Optical rotations were measured in CH_3CH_2OH on a Jasco P-1030 polarimeter. Dichloromethane was fractionally distilled.

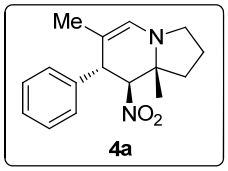
2. Procedure for the synthesis of indolizidine and quinolizidine Alkaloids:

Aldehyade **1** (0.6 mmol) was added to a solution of nitroalkene **2** (0.20 mmol), *p*-nitrophenol (0.010 mmol) and catalyst ***S-1*** (0.010 mmol) in CH_2Cl_2 (0.20 mL) at 0 ^{o}C. The resulted mixture was stirred until the nitroalkene was consumed (most examples needed only 5 hours, except **4n** needed 12 hours, **4j**, **4k** and **4q** needed 18 hours respectively), then the ketimine **3** (0.30 mmol) and DIPEA (0.20 mmol) were added subsequently and further stirred for another 5 hours at 0 ^{o}C. The solvent was removed under vacuum. The resulted crude product was purified by flash chromatography on a short silica gel directly to afford the desired products **4a**-**y**.

Procedure for gram-scale one-pot stereoselective synthesis of indolizidine 4a:

Aldehyade **1a** (18.0 mmol) was added to a solution of nitroalkene **2a** (6.0 mmol), *p*-nitrophenol (0.30 mmol) and catalyst ***S-1*** (0.060 mmol) in CH_2Cl_2 (6.0 mL) at 0 ^{o}C. The resulted mixture was stirred for 96 hours at 0 ^{o}C, then the ketimine **3a** (9.0 mmol) and DIPEA (6.0 mmol) were added subsequently and further stirred for another 5 hours at 0 ^{o}C. The solvent was removed under vacuum. The resulted crude product was purified by flash chromatography on a short silica gel directly to afford 1.391g (85% yield, >20:1, 96% ee) of the desired product **4a**.

3. Characterization data of indolizidine and quinolizidine derivatives



4a

(7*R*,8*S*,8a*R*)-6, 8a-dimethyl-8-nitro-7-phenyl-1,2,3,7,8,8a-hexahydroindolizine (4a)

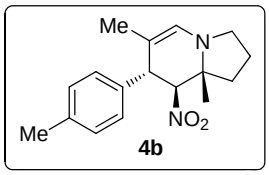
Purified by FC (PE:EtOAc = 50:1). 91% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1);flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 9.8 min, τ_{major} = 11.7 min, 98% ee.

$[\alpha]_D^{20}$ = - 60.5 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.30 – 7.20 (m, 3H), 7.16 (d, *J* = 6.8 Hz, 2H), 6.00 (s, 1H), 4.38 – 4.35 (d, *J* = 11.2Hz, 1H), 3.94 – 3.91 (d, *J* = 11.2Hz, 1H), 3.45 – 3.40 (m, 1H), 3.05 – 2.99 (m, 1H), 2.18 – 2.10 (m, 1H), 2.00 – 1.87 (m, 2H), 1.82 – 1.77 (m, 1H), 1.36 (s, 3H), 1.17 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 139.0, 128.6（2C）, 127.4（2C）, 105.6, 96.6, 61.0, 51.4, 46.4, 37.8, 23.3, 19.3, 17.7.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{16}H_{22}N_2O_2 \bullet H^+$：273.1598; found：273.1605.



4b

(7*R*,8*S*,8a*R*)-6,8a-dimethyl-8-nitro-7-(p-tolyl)-1,2,3,7,8,8a-hexahydroindolizine (4b)

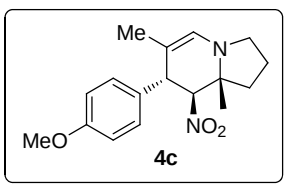
Purified by FC (PE:EtOAc = 50:1). 84% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 8.8 min, τ_{major} = 11.5 min, 98% ee.

$[\alpha]_D^{20}$ = - 60.5 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.09 – 7.03 (m, 4H), 5.98 (s, 1H), 4.34 (d, *J* = 11.2Hz, 1H), 3.88 (d, *J* = 11.2Hz, 1H), 3.44 – 3.39 (m, 1H), 3.04 – 2.97 (m, 1H), 2.29 (s, 3H), 2.17 – 2.09 (m, 1H), 1.93 – 1.86 (m, 2H), 1.81 – 1.76 (m, 1H), 1.36 (s, 3H), 1.16 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 136.9, 135.8, 129.3, 128.4, 127.3, 105.8, 96.7, 61.0, 51.4, 46.0, 37.7, 23.3, 21.0, 19.3, 17.7.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{17}H_{22}N_2O_2 \bullet H^+$：287.1754; found：287.1749.



4c

(7*R*,8*S*,8a*R*)-7-(4-methoxyphenyl)-6,8a-dimethyl-8-nitro-1,2,3,7,8,8a-hexahydroindolizine

(4c)

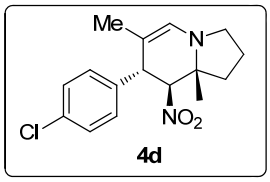
Purified by FC (PE:EtOAc = 40:1). 52% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 14.4 min, τ_{major} = 27.8 min, 98% ee.

$[\alpha]_D^{20}$ = - 61.7 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.08 (d, *J* = 8.8 Hz, 2H), 6.81 (d, *J* = 8.8 Hz, 2H), 5.91 (s, 1H), 4.32 (d, *J* = 11.2 Hz, 1H), 3.87 (d, *J* = 11.2 Hz, 1H), 3.77 (s, 3H), 3.44 – 3.40 (m, 1H), 3.03 – 2.97 (m, 1H), 2.17 – 2.09 (m, 1H), 1.94 – 1.87 (m, 2H), 1.81 – 1.76 (m, 1H), 1.36 (s, 3H), 1.16 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 158.8, 130.7, 129.6, 127.3, 114.0, 106.0, 96.8, 61.0, 55.2, 51.4, 45.6, 37.7, 23.3, 19.3, 17.7.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{17}H_{22}N_2O_3 \cdot H^+$：303.1703; found：303.1701.



4d

(7*R*,8*S*,8a*R*)-7-(4-chlorophenyl)-6,8a-dimethyl-8-nitro-1,2,3,7,8,8a-hexahydroindolizine (4d)

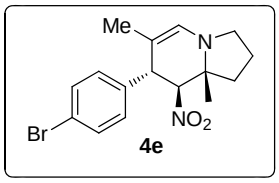
Purified by FC (PE:EtOAc = 50:1). 70% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 9.4 min, τ_{major} = 14.4 min, 98% ee.

$[\alpha]_D^{20}$ = - 81.2 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.27 – 7.25 (m, 2H), 7.12 – 7.10 (m, 2H), 6.00 (s, 1H), 4.29 (d, *J* = 11.2 Hz, 1H), 3.91 (d, *J* = 11.2 Hz, 1H), 3.45 – 3.40 (m, 1H), 3.05 – 2.99 (m, 1H), 2.17 – 2.10 (m, 1H), 1.96 – 1.88 (m, 2H), 1.83 – 1.77 (m, 1H), 1.35 (s, 3H), 1.15 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 137.7, 133.1, 130.0, 128.9, 127.8, 104.6, 96.6, 61.1, 51.3, 45.8, 37.8, 23.3, 19.2, 17.6.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{16}H_{19}ClN_2O_2 \cdot H^+$：307.1208; found：307.1193.



4e

(7*R*,8*S*,8a*R*)-7-(4-bromophenyl)-6,8a-dimethyl-8-nitro-1,2,3,7,8,8a-hexahydroindolizine (4e)

Purified by FC (PE:EtOAc = 50:1). 84% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 10.1 min, τ_{major} = 17.7 min, 98% ee.

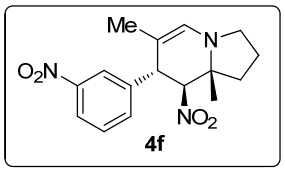
$[\alpha]_D^{20}$ = - 76.5 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.41 (d, *J* = 8.4 Hz, 2H), 7.05 (d, *J* = 8.4 Hz, 2H), 6.00 (s, 1H), 4.29 (d,

J = 11.2 Hz, 1H), 3.90 (d, *J* = 11.2 Hz, 1H), 3.44 – 3.40 (m, 1H), 3.05 – 2.99 (m, 1H), 2.17 – 2.08 (m, 1H), 1.94 – 1.87 (m, 2H), 1.82 – 1.77 (m, 1H), 1.34 (s, 3H), 1.14 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 138.2, 131.8, 130.3, 127.7, 121.2, 104.4, 96.4, 61.0, 51.3, 45.8, 37.8, 23.2, 19.2, 17.6.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{16}H_{19}BrN_2O_2 \bullet H^+$：351.0703; found：351.0699.



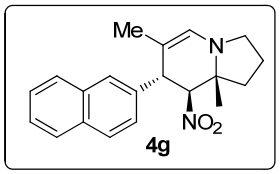
4f

(7*R*,8*S*,8a*R*)-6,8a-dimethyl-8-nitro-7-(3-nitrophenyl)-1,2,3,7,8,8a-hexahydroindolizine (4f)
Purified by FC (PE:EtOAc = 30:1). 89% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 16.2 min, τ_{major} = 18.5 min, 97% ee.
$[\alpha]_D^{20}$ = - 67.9 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 – 8.10 (m, 1H), 8.05 (s, 1H), 7.56 (d, *J* = 8.0 Hz, 1H), 7.48 (t, *J* = 8.0 Hz, 1H), 6.07 (s, 1H), 4.35 (d, *J* = 11.2 Hz, 1H), 4.09 (d, *J* = 11.2 Hz, 1H), 3.49 – 3.44 (m, 1H), 3.14 – 3.07 (m, 1H), 2.20 – 2.12 (m, 1H), 1.99 – 1.91 (m, 2H), 1.86 – 1.80 (m, 1H), 1.35 (s, 3H), 1.16 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 148.5, 141.7, 135.3(bs), 129.6, 128.4, 123.4, 122.6, 102.7, 96.3, 61.0, 51.1, 46.0, 37.8, 23.2, 19.1, 17.6.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{16}H_{19}N_3O_4 \bullet H^+$：318.1448; found：318.1446.



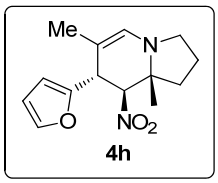
4g

(7*R*,8*S*,8a*R*)-6,8a-dimethyl-7-(naphthalen-2-yl)-8-nitro-1,2,3,7,8,8a-hexahydroindolizine (4g)
Purified by FC (PE:EtOAc = 40:1). 57% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 13.1min, τ_{major} = 16.4 min, 98% ee.
$[\alpha]_D^{20}$ = - 55.5 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.80 – 7.76 (m, 3H), 7.67 (s, 1H), 7.48 – 7.42 (m, 2H), 7.26 – 7.23 (m, 1H), 6.05 (s, 1H), 4.48 (d, *J* = 11.2 Hz, 1H), 4.10 (d, *J* = 11.2 Hz, 1H), 3.47 – 3.43 (m, 1H), 3.10 – 3.03 (m, 1H), 2.20 – 2.12 (m, 1H), 1.96 – 1.88 (m, 2H), 1.84 – 1.78 (m, 1H), 1.37 (s, 3H), 1.21 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 136.4, 133.4, 132.8, 128.6(2C), 127.7, 127.6 (2C), 126.1, 125.8(2C), 105.4, 96.4, 61.1, 51.4, 46.5, 37.8, 23.3, 19.3, 17.8.

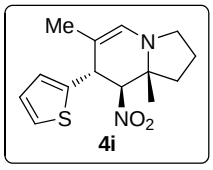
HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{20}H_{22}N_2O_2 \cdot H^+$：323.1754; found：323.1751.



4h

(7*R*,8*S*,8a*R*)-7-(furan-2-yl)-6,8a-dimethyl-8-nitro-1,2,3,7,8,8a-hexahydroindolizine (4h)
Purified by FC (PE:EtOAc = 40:1). 84% yield, yellow oil. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 9.1 min, τ_{major} = 12.2 min, 98% ee.
$[\alpha]_D^{20}$ = - 116.6 (c = 1.0, CH_3CH_2OH).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.33 (d, *J* = 1.2 Hz, 1H), 6.27 – 6.26 (m, 1H), 6.22 (d, *J* = 2.4 Hz, 1H), 5.97 (s, 1H), 4.57(d, *J* = 11.2 Hz, 1H), 4.13 (d, *J* = 11.6 Hz, 1H), 3.42 – 3.37 (m, 1H), 3.07 – 3.01 (m, 1H), 2.22 – 2.14 (m, 1H), 1.94 – 1.87 (m, 2H), 1.84 – 1.78 (m, 1H), 1.42 (s, 3H), 1.12 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 151.5, 142.2, 127.0, 110.1, 109.1, 103.2, 92.4, 60.8, 51.3, 40.0, 37.9, 23.3, 19.2, 17.4.

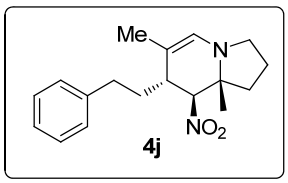
HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{14}H_{18}N_2O_3 \cdot H^+$：263.1390; found：263.1395.



4i

(7*R*,8*S*,8a*R*)-6,8a-dimethyl-8-nitro-7-(thiophen-2-yl)-1,2,3,7,8,8a-hexahydroindolizine (4i)
Purified by FC (PE:EtOAc = 40:1). 79% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 10.2 min, τ_{major} = 14.0 min, 98% ee.
$[\alpha]_D^{20}$ = - 65.5 (c = 1.0, CH_3CH_2OH).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.18 (d, *J* = 4.4 Hz, 1H), 6.92 – 6.88 (m, 2H), 5.95 (s, 1H), 4.41 (d, *J* = 11.2 Hz, 1H), 4.29 (d, *J* = 11.2 Hz, 1H), 3.43 – 3.38 (m, 1H), 3.07 – 3.01 (m, 1H), 2.20 – 2.12 (m, 1H), 1.96 – 1.87 (m, 2H), 1.82 – 1.77 (m, 1H), 1.47 (s, 3H), 1.14 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 142.4, 127.1, 127.0, 126.5, 124.6, 104.8, 96.8, 61.1, 51.2, 41.8, 37.8, 23.3, 19.1, 17.4.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{14}H_{18}N_2O_2S \cdot H^+$：279.1162; found：279.1156.



4j

(7*R*,8*S*,8a*R*)-6,8a-dimethyl-8-nitro-7-phenethyl-1,2,3,7,8,8a-hexahydroindolizine (4j)

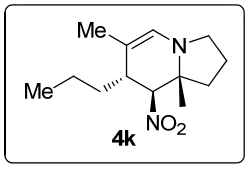
Purified by FC (PE:EtOAc = 50:1). 78% yield, yellow solid. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 99/1); flow rate 1.0 mL/min; λ = 254 nm; τ_{minor} = 4.8 min, τ_{major} = 5.9 min, 99% ee.

$[\alpha]_D^{20}$ = - 36.5 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.28 – 7.24 (m, 2H), 7.19 – 7.12 (m, 3H), 5.94 (s, 1H), 4.30 (d, *J* = 11.6Hz, 1H), 3.38 – 3.33 (m, 1H), 3.00 – 2.90 (m, 2H), 2.56 – 2.48 (m, 1H), 2.42 – 2.35 (m, 1H), 2.15 – 2.04 (m, 1H), 1.96 – 1.85 (m, 3H), 1.82 – 1.77 (m, 1H), 1.73 (s, 3H), 1.70 – 1.61 (m, 1H), 1.07 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 141.9, 128.4, 128.2, 127.9, 125.9, 105.0, 92.4, 60.9, 51.2, 38.0, 37.8, 30.1, 29.6, 23.2, 19.1, 17.3.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{18}H_{24}N_2O_2 \cdot H^+$：301.1911; found：301.1906.



4k

(7*R*,8*S*,8a*R*)-6,8a-dimethyl-8-nitro-7-propyl-1,2,3,7,8,8a-hexahydroindolizine (4k)

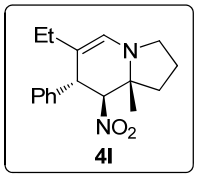
Purified by FC (PE:EtOAc = 100:1). 78% yield, yellow oil. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 5.2 min, τ_{major} = 5.7 min, 98% ee.

$[\alpha]_D^{20}$ = - 16.6 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 5.85 (s, 1H), 4.19 (d, *J* = 11.6 Hz, 1H), 3.34 – 3.30 (m, 1H), 2.90 – 2.84 (m, 2H), 2.13 – 2.05 (m, 1H), 1.91 – 1.82 (m, 2H), 1.79 – 1.73 (m, 1H), 1.63 (s, 3H), 1.58 – 1.52 (m, 1H), 1.38 – 1.31 (m, 1H), 1.24 – 1.18 (m, 1H), 1.14 – 1.09 (m, 1H), 1.05 (s, 3H), 0.88 (t, *J* = 7.2 Hz, 3H)

^{13}C NMR (100 MHz, $CDCl_3$) δ 127.4, 106.1, 92.7, 60.9, 51.3, 38.0, 37.8, 29.8, 23.2, 19.2, 17.2, 17.0, 14.5.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{13}H_{22}N_2O_2 \cdot H^+$：239.1754; found：239.1749.



4l

(7*R*,8*S*,8a*R*)-6-ethyl-8a-methyl-8-nitro-7-phenyl-1,2,3,7,8,8a-hexahydroindolizine (4l)

Purified by FC (PE:EtOAc = 50:1). 86% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 8.4 min, τ_{major} = 9.2 min, 99% ee.

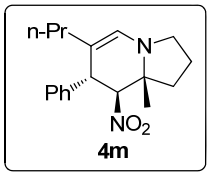
$[\alpha]_D^{20}$ = - 1.6 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.29 – 7.16 (m, 5H), 6.02 (s, 1H), 4.36 (d, *J* = 11.2 Hz, 1H), 4.02 (d, *J*

= 11.2 Hz, 1H), 3.45 – 3.41 (m, 1H), 3.06 – 2.99 (m, 1H), 2.17 – 2.09 (m, 1H), 1.96 – 1.88 (m, 2H), 1.82 – 1.69 (m, 2H), 1.68 – 1.58 (m, 1H), 1.17 (s, 3H), 0.88 (t, *J* = 7.2 Hz, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 138.9, 128.5(2C), 127.3, 126.2, 111.9, 96.9, 60.9, 51.3, 44.7, 37.7, 24.8, 23.3, 19.0, 13.4.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{17}H_{22}N_2O_2 \cdot H^+$：287.1754; found：287.1751.



4m

(7*R*,8*S*,8a*R*)-8a-methyl-8-nitro-7-phenyl-6-propyl-1,2,3,7,8,8a-hexahydroindolizine (4m)

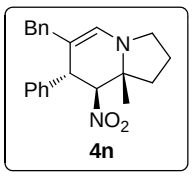
Purified by FC (PE:EtOAc = 50:1). 84% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 7.9 min, τ_{major} = 8.6 min, >99% ee.

$[\alpha]_D^{20}$ = - 1.8 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.30 – 7.22 (m, 3H), 7.17 (d, *J* = 6.4 Hz, 2H), 6.03 (s, 1H), 4.34 (d, *J* = 11.2 Hz, 1H), 3.97 (d, *J* = 11.2 Hz, 1H), 3.46 – 3.42 (m, 1H), 3.06 – 3.00 (m, 1H), 2.17 – 2.09 (m, 1H), 1.95 – 1.88 (m, 2H), 1.82 – 1.77 (m, 1H), 1.68 – 1.57 (m, 2H), 1.34 – 1.30 (m, 1H), 1.26 – 1.21 (m, 1H), 1.18 (s, 3H), 0.77 (t, *J* = 7.6 Hz, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 138.9, 128.5(2C), 127.3, 127.2, 109.7, 97.0, 60.9, 51.3, 44.7, 37.7, 33.7, 23.3, 21.4, 19.1, 13.4.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{18}H_{24}N_2O_2 \cdot H^+$：301.1911; found：301.1908.



4n

(7*R*,8*S*,8a*R*)-6-benzyl-8a-methyl-8-nitro-7-phenyl-1,2,3,7,8,8a-hexahydroindolizine (4n)

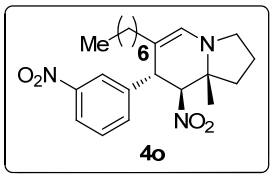
Purified by FC (PE:EtOAc = 50:1). 73% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 11.0 min, τ_{major} = 12.5 min, >99% ee.

$[\alpha]_D^{20}$ = 53.3 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.25 – 7.14 (m, 6H), 7.07 (br, 2H), 6.92 (d, *J* = 7.2Hz, 2H), 6.14 (s, 1H), 4.49 (d, *J* = 11.6 Hz, 1H), 3.74 (d, *J* = 11.2 Hz, 1H), 3.46 (m, 1H), 3.10 – 3.05 (m, 2H), 2.83 – 2.79 (m, 1H), 2.16 – 2.08 (m, 1H), 1.95 – 1.88 (m, 2H), 1.80 – 1.76 (m, 1H), 1.08 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 140.4, 138.5, 128.6(2C), 128.3, 128.1, 127.5(2C), 125.9, 109.3, 96.9, 61.0, 51.1, 44.5, 38.3, 37.7, 23.3, 19.3.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{22}H_{24}N_2O_2 \cdot H^+$：349.1911; found：349.1905.



4o

(7*R*,8*S*,8a*R*)-6-heptyl-8a-methyl-8-nitro-7-(3-nitrophenyl)-1,2,3,7,8,8a-hexahydroindolizine (4o)

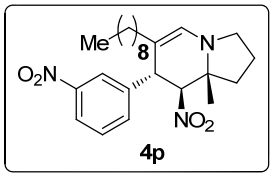
Purified by FC (PE:EtOAc = 50:1). 97% yield, yellow oil. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 99/1); flow rate 1.0 mL/min; λ = 254 nm; τ_{minor} = 5.7 min, τ_{major} = 6.6 min, 99% ee.

$[\alpha]_D^{20}$ = 12.0 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 – 8.10 (m, 1H), 8.06 (s, 1H), 7.56 (d, *J* = 8.0 Hz, 1H), 7.47 (t, *J* = 8.0 Hz, 1H), 6.09 (s, 1H), 4.32 (d, *J* = 11.2 Hz, 1H), 4.14 (d, *J* = 11.2 Hz, 1H), 3.49 – 3.45 (m, 1H), 3.14 – 3.08 (m, 1H), 2.19 – 2.11 (m, 1H), 1.99 – 1.91 (m, 2H), 1.85 – 1.80 (m, 1H), 1.75 – 1.64 (m, 1H), 1.59 – 1.53 (m, 1H), 1.26 – 1.15 (m, 12H), 1.08 – 1.06 (m, 1H), 0.85 (t, *J* = 7.2 Hz, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 148.5, 141.7, 135.2 (bs), 129.5, 128.1, 122.9, 122.6, 107.3, 96.6, 61.0, 51.1, 44.4, 37.8, 31.6, 31.6, 28.9, 28.8, 28.4, 23.2, 22.5, 18.9, 14.0.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{22}H_{31}N_3O_4 \bullet H^+$：402.2387; found：402.2388.



4p

(7*R*,8*S*,8a*R*)-8a-methyl-8-nitro-7-(3-nitrophenyl)-6-nonyl-1,2,3,7,8,8a-hexahydroindolizine (4p)

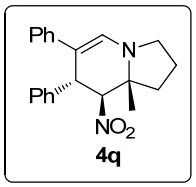
Purified by FC (PE:EtOAc = 50:1). 98% yield, yellow oil. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 14.5 min, τ_{major} = 18.4 min, 99% ee.

$[\alpha]_D^{20}$ = 9.4 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 – 8.10 (m, 1H), 8.05 (s, 1H), 7.56 (d, *J* = 7.6 Hz, 1H), 7.47 (t, *J* =7.6 Hz, 1H), 6.09 (s, 1H), 4.32 (d, *J* = 11.2 Hz, 1H), 4.14 (d, *J* = 11.2 Hz, 1H), 3.49 – 3.45 (m, 1H), 3.14 – 3.08 (m, 1H), 2.19 – 2.11 (m, 1H), 1.99 – 1.91 (m, 2H), 1.85 – 1.80 (m, 1Hz), 1.74 – 1.67 (m, 1H), 1.61 – 1.53 (m, 1H), 1.28 – 1.15 (m, 16H), 1.09 – 1.04 (m, 1H), 0.87 (t, *J* = 6.8 Hz, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 148.6, 141.7, 135.8 (bs), 129.6, 128.1, 123.2, 122.6, 107.4, 96.7, 61.0, 51.1, 44.5, 37.8, 31.8, 31.6, 29.4, 29.3, 29.2, 28.8, 28.4, 23.2, 22.6, 18.9, 14.0.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{24}H_{35}N_3O_4 \bullet H^+$：430.2700; found：430.2693.



4q

(7*R*,8*S*,8a*R*)-8a-methyl-8-nitro-6,7-diphenyl-1,2,3,7,8,8a-hexahydroindolizine (4q)

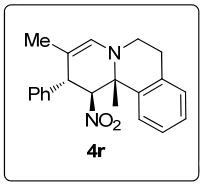
Purified by FC (PE:EtOAc = 50:1). 57% yield, yellow solid. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{major} = 14.2 min, τ_{minor} = 16.3 min, 90% ee.

$[\alpha]_D^{20}$ = - 71.7 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.16 – 7.05 (m, 9H), 6.91 – 6.87 (m, 1H), 6.74 (s, 1H), 4.61 (d, *J* = 10.8Hz, 1H), 4.46 (d, *J* = 10.8Hz, 1H), 3.59 – 3.54 (m, 1H), 3.34 – 3.28 (m, 1H), 2.16 – 2.08 (m, 1H), 2.06 – 1.97 (m, 2H), 1.89 – 1.84 (m, 1H), 1.24 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 139.6, 139.2, 130.3, 128.6, 128.4, 127.9, 127.1, 125.4, 124.3, 108.5, 98.2, 61.1, 50.4, 44.0, 37.8, 22.9, 18.3.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{21}H_{22}N_2O_2 \cdot H^+$：335.1754; found：335.1745.



4r

(1*R*,2*S*,11b*S*)-3,11b-dimethyl-1-nitro-2-phenyl-2,6,7,11b-tetrahydro-1H-pyrido[2,1-*a*]isoquinoline (4r)

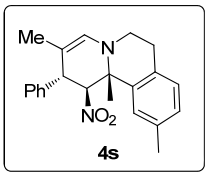
Purified by FC (PE:EtOAc = 50:1). 41% yield, yellow solid. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{major} = 11.2 min, τ_{minor} = 13.0 min, 99% ee.

$[\alpha]_D^{20}$ = - 276.8 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.27 – 7.12 (m, 8H), 7.00 (m, 1H), 5.95 (s, 1H), 4.62 (d, *J* = 10.8 Hz, 1H), 4.02 (d, *J* = 10.8 Hz, 1H), 3.38 – 3.32 (m, 1H), 3.20 – 3.14 (m, 1H), 3.03 – 2.96 (m, 1H), 2.86 – 2.80 (m, 1H), 1.71 (s, 3H), 1.42 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 138.5, 137.3, 134.6, 131.3, 129.2, 128.7, 128.3, 127.6, 127.2, 126.6, 126.0, 106.3, 96.8, 59.2, 48.8, 46.4, 30.8, 18.8, 17.6.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{21}H_{22}N_2O_2 \cdot H^+$：335.1754; found：335.1737.

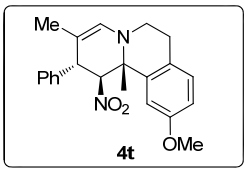


4s

(1*R*,2*S*,11b*S*)-3,10,11b-trimethyl-1-nitro-2-phenyl-2,6,7,11b-tetrahydro-1H-pyrido[2,1-*a*]isoquinoline (4s)

Purified by FC (PE:EtOAc = 50:1). 44% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 10.4 min, τ_{major} = 13.7 min, >99% ee.
$[\alpha]_D^{20}$ = - 289.8 (c = 1.0, CH_3CH_2OH).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.25 – 7.19 (m, 3H), 7.13 – 7.11 (m, 2H), 7.01 – 6.96 (m, 2H), 6.77 (s, 1H), 5.95 (s, 1H), 4.62 (d, *J* = 10.4 Hz, 1H), 4.00 (d, *J* = 10.8 Hz, 1H), 3.37 – 3.31 (m, 1H), 3.19 – 3.13 (m, 1H), 2.99 – 2.92 (m, 1H), 2.80 – 2.74 (m, 1H), 2.22 (s, 3H), 1.69 (s, 3H), 1.42 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 138.6, 137.1, 136.0, 131.5, 131.4, 129.0, 128.6, 128.3, 128.1, 127.5, 126.4, 106.0, 96.8, 59.0, 48.7, 46.6, 30.3, 21.2, 19.0, 17.6.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{22}H_{24}N_2O_2 \cdot H^+$：349.1911; found：349.1897.

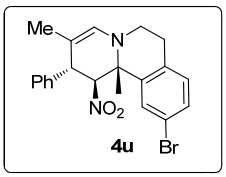


4t

(1*R*,2*S*,11b*S*)-10-methoxy-3,11b-dimethyl-1-nitro-2-phenyl-2,6,7,11b-tetrahydro-1H-pyrido[2,1-*a*]isoquinoline (4t)

Purified by FC (PE:EtOAc = 50:1). 42% yield, yellow solid. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{major} = 11.4 min, τ_{minor} = 13.7 min , 98% ee.
$[\alpha]_D^{20}$ = - 325.9 (c = 1.0, CH_3CH_2OH).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.26 – 7.20 (m, 3H), 7.12 (d, *J* = 6.8 Hz, 2H), 7.02 (d, *J* = 8.4 Hz, 1H), 6.74 (d, *J* = 8.4 Hz, 1H), 6.52 (s, 1H), 5.95 (s, 1H), 4.63 (d, *J* = 10.8 Hz, 1H), 4.00 (d, *J* = 10.8 Hz, 1H), 3.68 (s, 3H), 3.36 – 3.30 (m, 1H), 3.17 – 3.12 (m, 1H), 2.95 – 2.89 (m, 1H), 2.78 – 2.72 (m, 1H), 1.70 (s, 3H), 1.42 (s, 3H).
^{13}C NMR (100 MHz, $CDCl_3$) δ 158.0, 138.5, 138.2, 131.4, 130.0, 128.7, 128.3, 127.5, 126.6, 113.9, 110.8, 106.1, 96.8, 59.2, 55.2, 48.8, 46.6, 29.8, 18.9, 17.6.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{22}H_{24}N_2O_3 \cdot H^+$：365.1860; found：365.1850.



4u

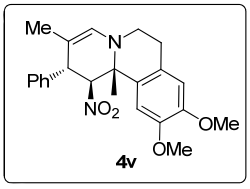
(1*R*,2*S*,11b*S*)-10-bromo-3,11b-dimethyl-1-nitro-2-phenyl-2,6,7,11b-tetrahydro-1H-pyrido[2,1-*a*]isoquinoline (4u)

Purified by FC (PE:EtOAc = 50:1). 58% yield, yellow solid. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 99/1);flow rate 0.7 mL/min; λ = 254 nm; τ_{major} = 7.8 min, τ_{minor} = 8.6 min, 98% ee.
$[\alpha]_D^{20}$ = - 268.8 (c = 1.0, CH_3CH_2OH).
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.29 – 7.21 (m, 4H), 7.11 – 7.07 (m, 3H), 6.99 (d, *J* = 8.4 Hz, 1H), 5.95

(s, 1H), 4.64 (d, *J* = 10.4 Hz, 1H), 3.98 (d, *J* = 10.4 Hz, 1H), 3.36 – 3.30 (m, 1H), 3.20 – 3.15 (m, 1H), 2.98 – 2.90 (m, 1H), 2.80 – 2.74 (m, 1H), 1.68 (s, 3H), 1.43 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 139.5, 138.2, 133.6, 131.2, 130.7, 130.4, 129.0, 128.7, 128.3, 127.6, 120.1, 106.9, 96.4, 58.6, 48.5, 46.2, 30.1, 19.3, 17.6.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{21}H_{21}BrN_2O_2 \bullet H^+$：413.0859; found：413.0850.



4v

(1*R*,2*S*,11b*S*)-9,10-dimethoxy-3,11b-dimethyl-1-nitro-2-phenyl-2,6,7,11b-tetrahydro-1H-pyrido[2,1-*a*]isoquinoline (4v)

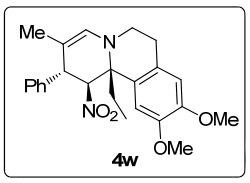
Purified by FC (PE:EtOAc = 20:1). 67% yield, yellow oil. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 95/5); flow rate 0.7 mL/min; λ = 254 nm; τ_{major} = 11.4 min, τ_{minor} = 13.5 min, 98% ee.

$[\alpha]_D^{20}$ = - 209.9 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.27 – 7.20 (m, 3H), 7.12 (d, *J* = 6.4 Hz, 2H), 6.58 (s, 1H), 6.46 (s, 1H), 5.95 (s, 1H), 4.62 (d, *J* = 10.8 Hz, 1H), 4.00 (d, *J* = 10.8 Hz, 1H), 3.84 (s, 3H), 3.74 (s, 3H), 3.38 – 3.32 (m, 1H), 3.16 – 3.10 (m, 1H), 2.95 – 2.88 (m, 1H), 2.78 – 2.72 (m, 1H), 1.70 (s, 3H), 1.43 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 148.0, 147.3, 138.4, 131.6, 129.2, 128.7, 128.3, 127.6, 126.9, 111.4, 109.0, 106.4, 97.1, 58.9, 55.8, 55.7, 48.9, 46.3, 30.2, 18.9, 17.6.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{23}H_{26}N_2O_4 \bullet H^+$：395.1965; found：395.1959.



4w

(1*R*,2*S*,11b*S*)-11b-ethyl-9,10-dimethoxy-3-methyl-1-nitro-2-phenyl-2,6,7,11b-tetrahydro-1H-pyrido[2,1-*a*]isoquinoline (4w)

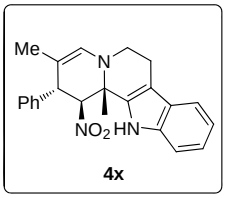
Purified by FC (PE:EtOAc = 20:1). 30% yield, yellow solid. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 99/1);flow rate 0.7 mL/min; λ = 254 nm; τ_{major} = 18.2 min, τ_{minor} = 25.2 min, >99% ee.

$[\alpha]_D^{20}$ = - 177.1 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.28 – 7.19 (m, 3H), 7.13 – 7.11 (m, 2H), 6.59 (s, 1H), 6.48 (s, 1H), 5.97 (s, 1H), 4.54 (d, *J* = 11.2 Hz, 1H), 3.98 (d, *J* = 11.6 Hz, 1H), 3.84 (s, 3H), 3.75 (s, 3H), 3.40 – 3.34 (m, 1H), 3.19 – 3.13 (m, 1H), 2.89 – 2.74 (m, 2H), 2.23 (q, *J* = 7.2 Hz, 2H), 1.42 (s, 3H), 0.66 (t, *J* = 7.2 Hz, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 148.0, 147.5, 138.4, 131.1, 128.7(2C), 128.5, 127.6, 126.4, 111.3, 109.0, 108.6, 97.2, 62.6, 55.9, 55.7, 48.4, 47.7, 30.0, 23.0, 17.5, 8.3.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{24}H_{28}N_2O_4 \cdot H^+$：409.2122; found：409.2125.



4x

(1*R*,2*S*,12b*R*)-3,12b-dimethyl-1-nitro-2-phenyl-1,2,6,7,12,12b-hexahydroindolo[2,3-*a*]quinolizine (4x)

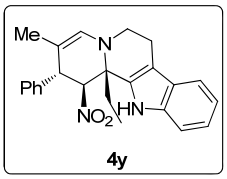
Purified by FC (PE:EtOAc = 20:1). 83% yield, yellow solid. The ee was determined by chiral HPLC using a Phenomenex Lux 5u Amylose-2 column (hexane/*i*-PrOH = 99/1); flow rate 0.7 mL/min; λ = 254 nm; τ_{minor} = 12.9 min, τ_{major} = 15.9 min, 98% ee.

$[\alpha]_D^{20}$ = - 128.4 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (s, 1H), 7.49 (d, *J* = 7.6 Hz, 1H), 7.30 – 7.24 (m, 3H), 7.21 – 7.06 (m, 5H), 5.99 (s, 1H), 4.85 (d, *J* = 11.2 Hz, 1H), 3.96 (d, *J* = 11.2 Hz, 1H), 3.47 – 3.41 (m, 1H), 3.19 – 3.13 (m, 1H), 2.92 – 2.80 (m, 2H), 1.73 (s, 3H), 1.44 (s, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 137.8, 136.5, 133.6, 131.7, 128.9, 128.3, 127.8, 126.1, 122.4, 119.6, 118.4, 111.3, 109.6, 108.3, 95.6, 57.0, 48.9, 46.7, 21.9, 18.1, 17.7.

HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{23}H_{23}N_3O_2 \cdot H^+$：374.1863; found：374.1859.



4y

(1*R*,2*S*,12b*R*)-12b-ethyl-3-methyl-1-nitro-2-phenyl-1,2,6,7,12,12b-hexahydroindolo[2,3-*a*]quinolizine (4y)

Purified by FC (PE:EtOAc = 20:1). 70% yield, yellow solid. The ee was determined by chiral HPLC using a Daicel OD-H column (hexane/*i*-PrOH = 95/5);flow rate 0.7 mL/min; λ = 254 nm; τ_{major} = 7.0 min, τ_{minor} = 15.4 min, >99% ee.

$[\alpha]_D^{20}$ = - 41.4 (c = 1.0, CH_3CH_2OH).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.67 (s, 1H), 7.50 (d, *J* = 7.6 Hz, 1H), 7.31 – 7.06 (m, 8H), 6.00 (s, 1H), 4.81 (d, *J* = 11.2 Hz, 1H), 3.95 (d, *J* = 11.2 Hz, 1H), 3.50 – 3.45 (m, 1H), 3.16 – 3.10 (m, 1H), 2.94 – 2.80 (m, 2H), 2.28 – 2.16 (m, 2H), 1.44 (s, 3H), 0.55 (t, *J* = 7.2 Hz, 3H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 137.7, 136.6, 131.3, 131.2, 128.9, 128.3, 127.8, 126.0, 122.4, 119.5, 118.3, 112.2, 111.9, 111.3, 95.1, 60.6, 48.4, 47.3, 22.9, 21.6, 17.6, 7.9.

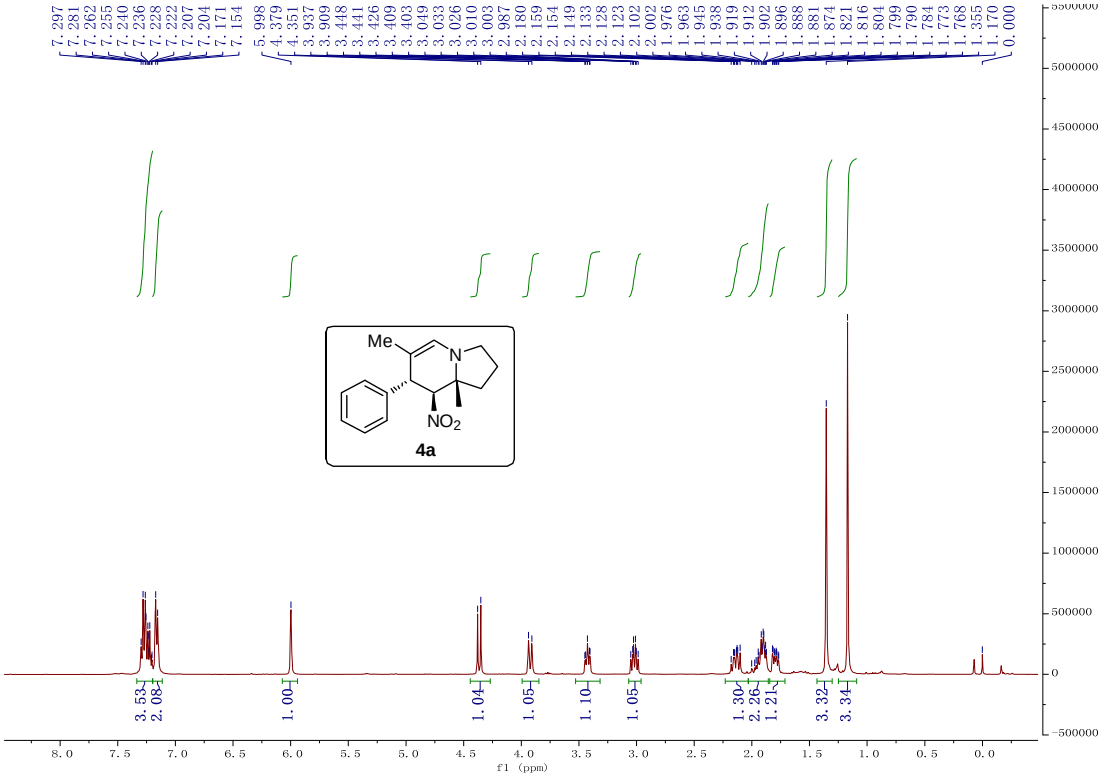
HRMS-ESI (*m/z*): $[M + H]^+$ calcd for $C_{24}H_{25}N_3O_2 \cdot H^+$：388.2020; found：388.2005.

4. Reference:

1. (a) Organic Syntheses, Coll. *Vol. 1,* p.413 (**1941**); *Vol. 9,* p.66 (**1929**). (b) Duursma, A.; Minnaard, A. J.; Feringa, B. L. *Tetrahedron* **2002**, *58*, 5773.

2. (a) Xie, J.-H.; Yan, P.-C.; Zhang, Q.-Q.; Yuan, K.-X.; Zhou, Q.-L. *ACS Catal.* **2012**, *2*, 561. (b) Roszkowski, P.; Wojtasiewicz, K.; Leniewski, A.; Maurin, J. K.; Lis, T.; Czarnocki, Z. *J. Mol. Catal. A Chem.* **2005**, *232*, 143.

5. ^{1}H and ^{13}C NMR Spectra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7.240
7.236
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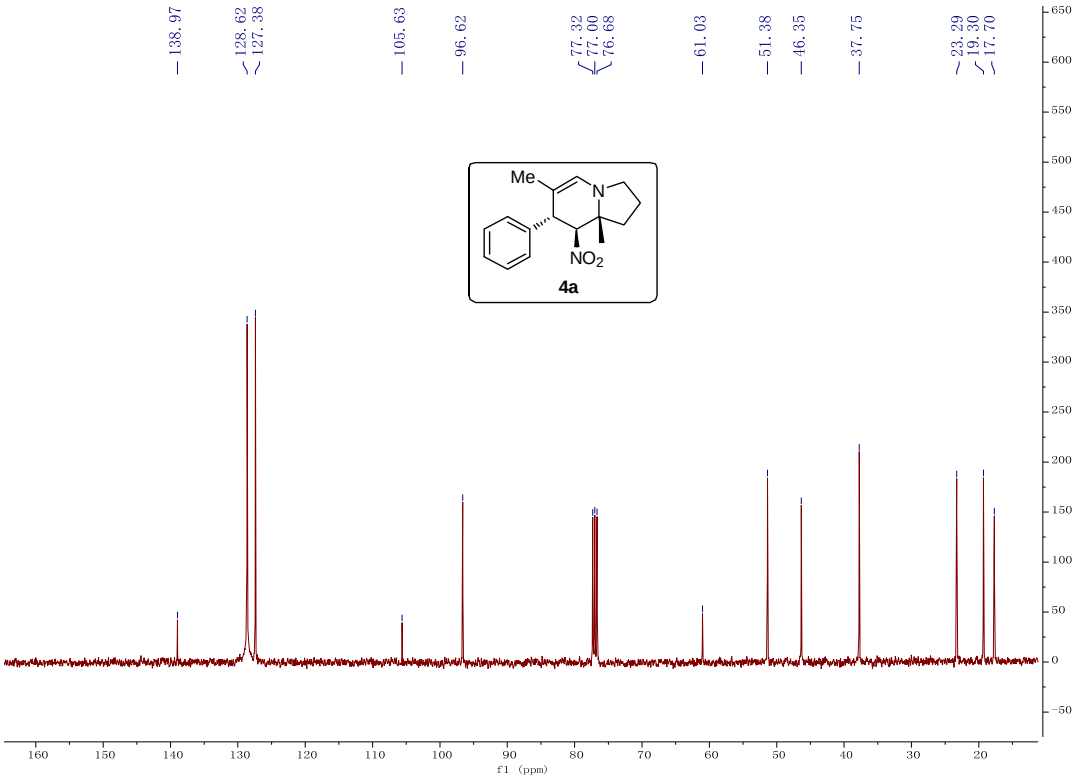
7.222
7.207
7.204
7.171
7.154
5.998
4.379
4.351
3.937
3.909
3.448
3.441
3.426
3.409
3.403
3.049
3.033
3.026
3.010
3.003
2.987
2.180
2.159
2.154
2.149
2.133
2.128
2.123
2.102
2.002
1.976
1.963
1.945
1.938
1.919
1.912
1.902
1.896
1.888
1.881
1.874
1.821
1.816
1.804
1.799
1.790
1.784
1.773
1.768
1.355
1.170
0.000
Me
N
NO2
4a
3.53
2.08
1.00
1.04
1.05
1.10
1.05
1.30
2.26
1.21
3.32
3.34
f1 (ppm)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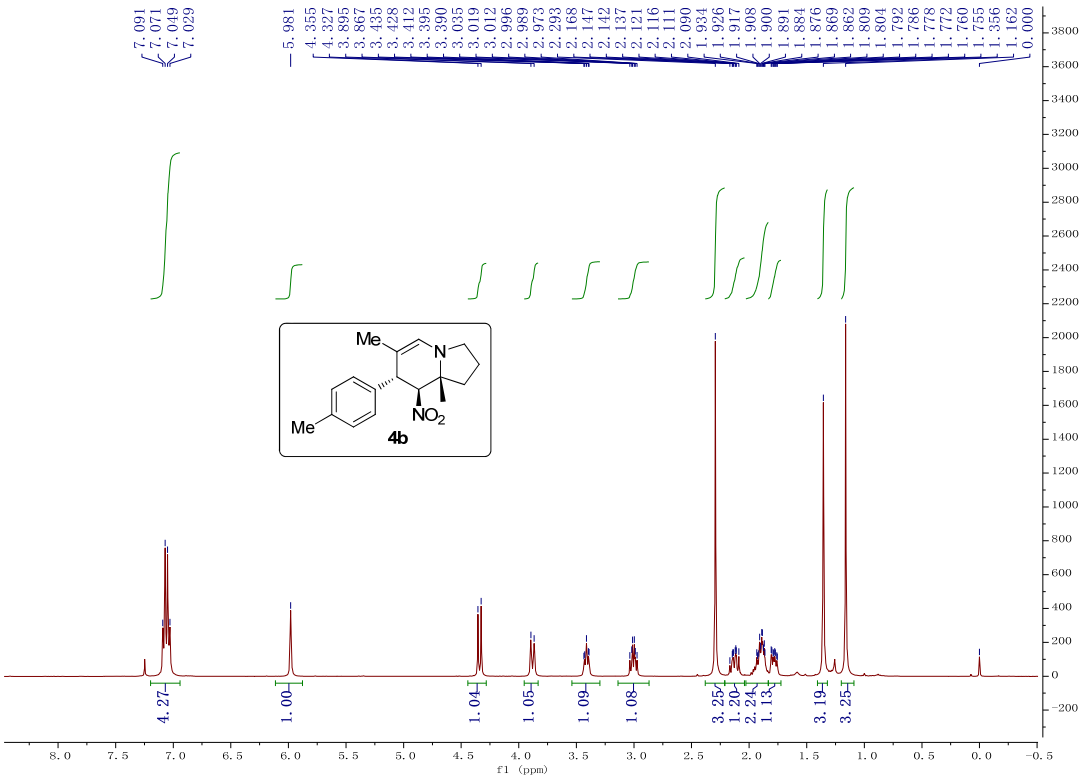
128.62
127.38
105.63
96.62
77.32
77.00
76.68
61.03
51.38
46.35
37.75
23.29
19.30
17.70
Me
N
NO2
4a
f1 (ppm)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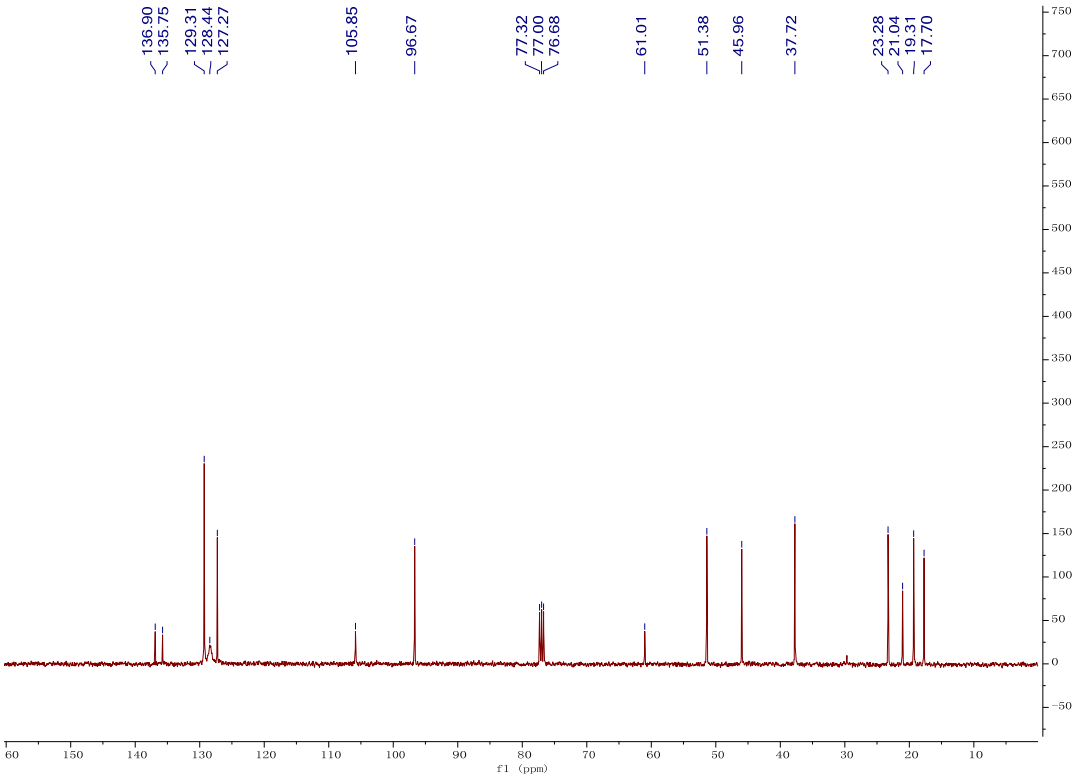
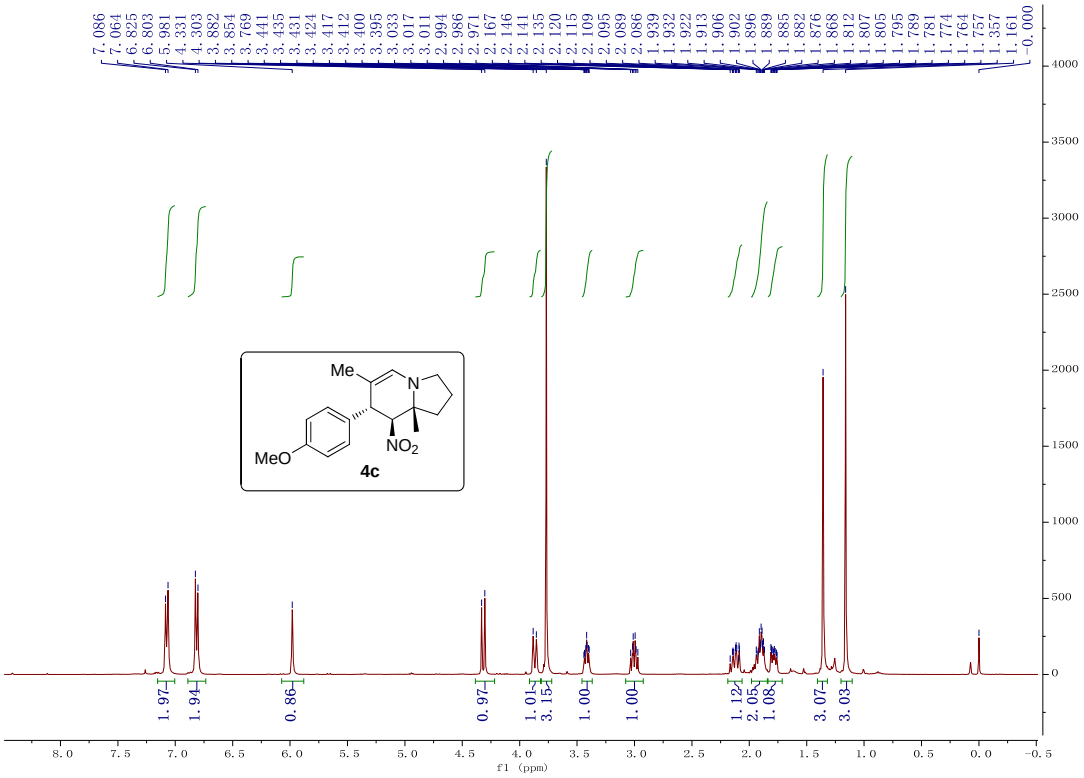
NO2
Me
4b
f1 (ppm)


f1 (ppm)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6.825
6.803
5.981
4.331
4.303
3.882
3.854
3.769
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3.435
3.431
3.424
3.417
3.412
3.400
3.395
3.033
3.017
3.011
2.994
2.986
2.971
2.167
2.146
2.141
2.135
2.120
2.115
2.109
2.095
2.089
2.086
1.939
1.932
1.922
1.913
1.906
1.902
1.896
1.889
1.885
1.882
1.876
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1.795
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1.764
1.757
1.357
1.161
0.000
Me
N
NO2
MeO
4c
1.97
1.94
0.86
0.97
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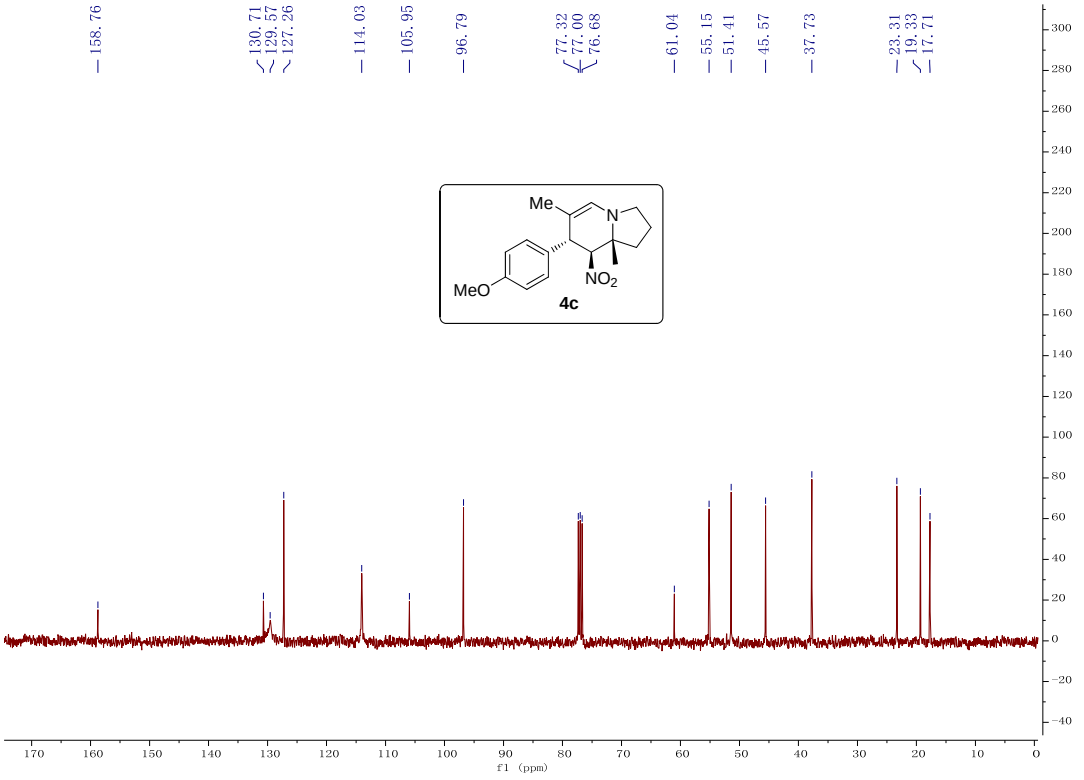
3.15
1.00
1.00
1.12
2.05
1.08
3.07
3.03
f1 (ppm)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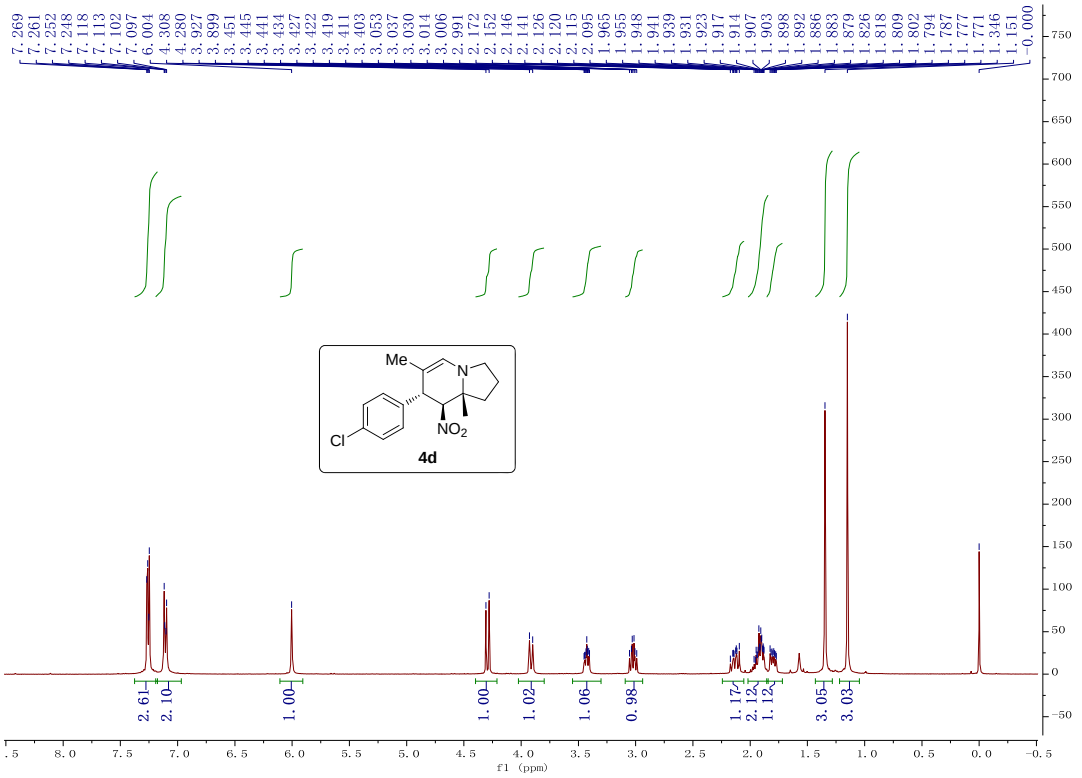
127.26
114.03
105.95
96.79
77.32
77.00
76.68
61.04
55.15
51.41
45.57
37.73
23.31
19.33
17.71
Me
N
NO2
MeO
4c
f1 (ppm)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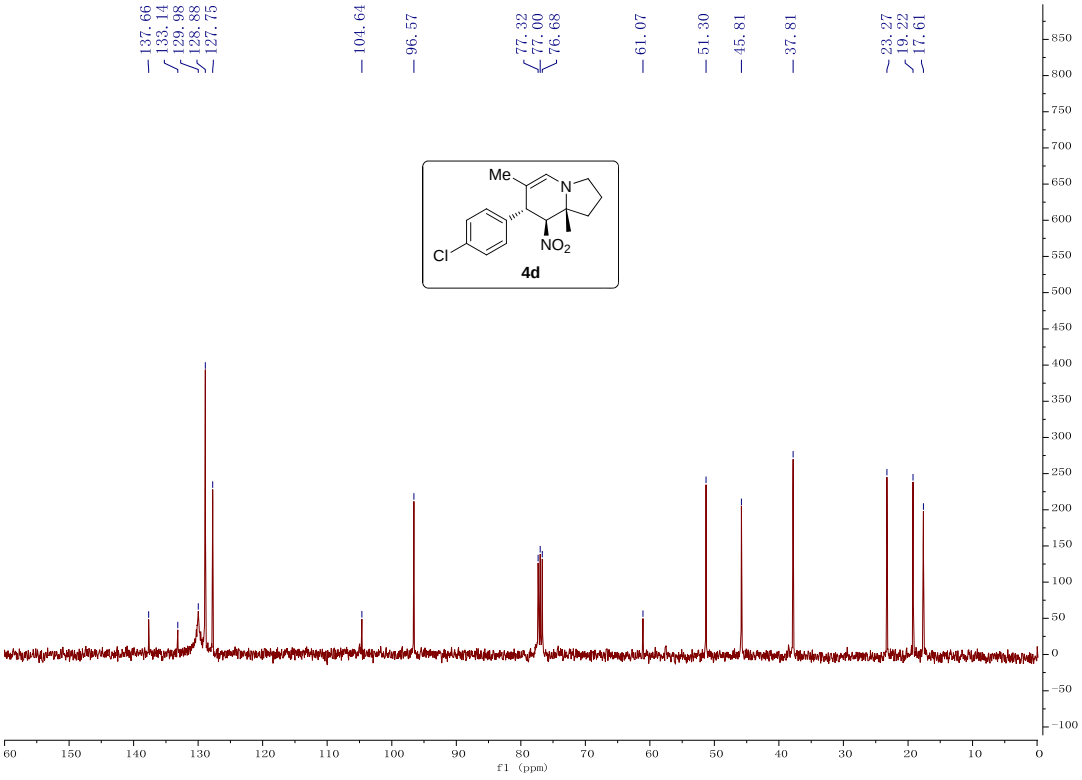
Cl
NO2
4d
f1 (ppm)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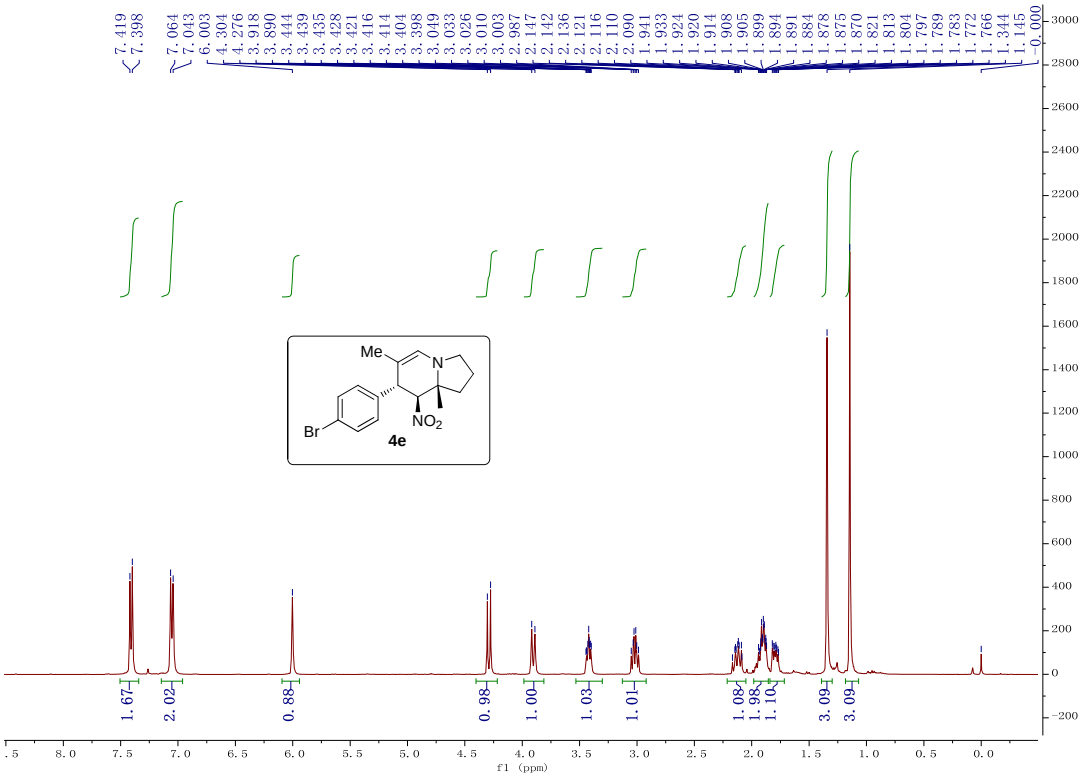
Cl
NO2
4d
f1 (ppm)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3.918
3.890
3.444
3.439
3.435
3.428
3.421
3.416
3.414
3.404
3.398
3.049
3.033
3.026
3.010
3.003
2.987
2.147
2.142
2.136
2.121
2.116
2.110
2.090
1.941
1.933
1.924
1.920
1.914
1.908
1.905
1.899
1.894
1.891
1.884
1.878
1.875
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1.821
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1.766
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0.000
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NO2
Br
4e
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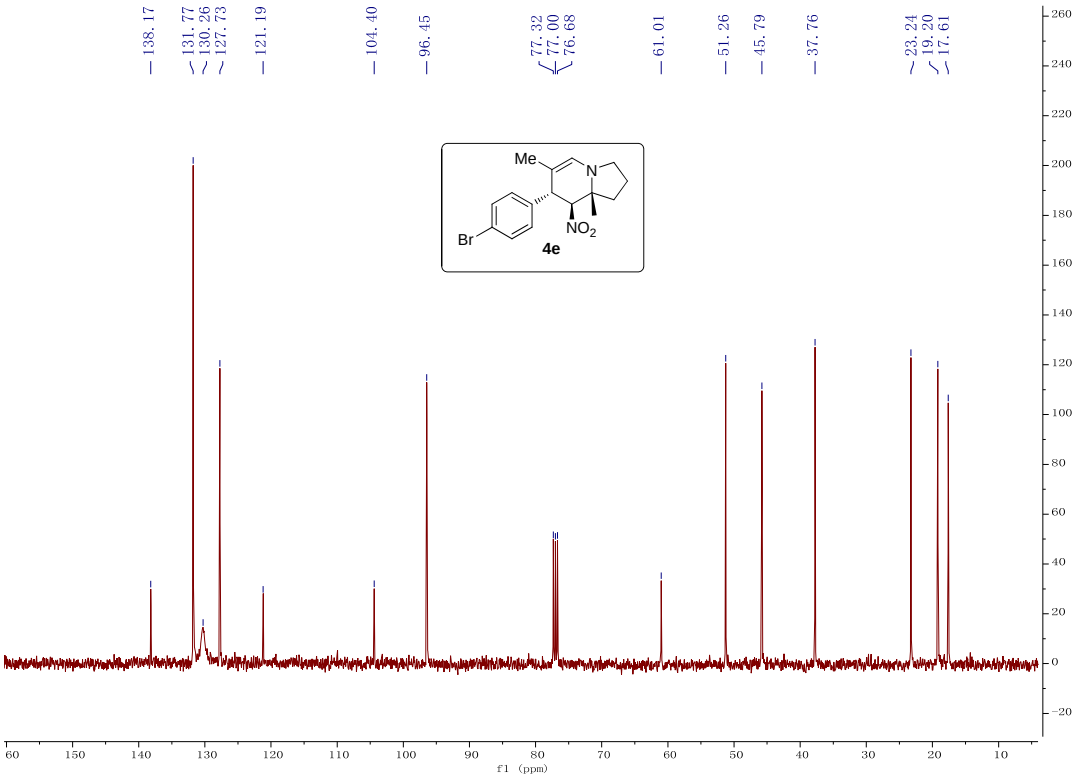
3.09
f1 (ppm)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77.32
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76.68
61.01
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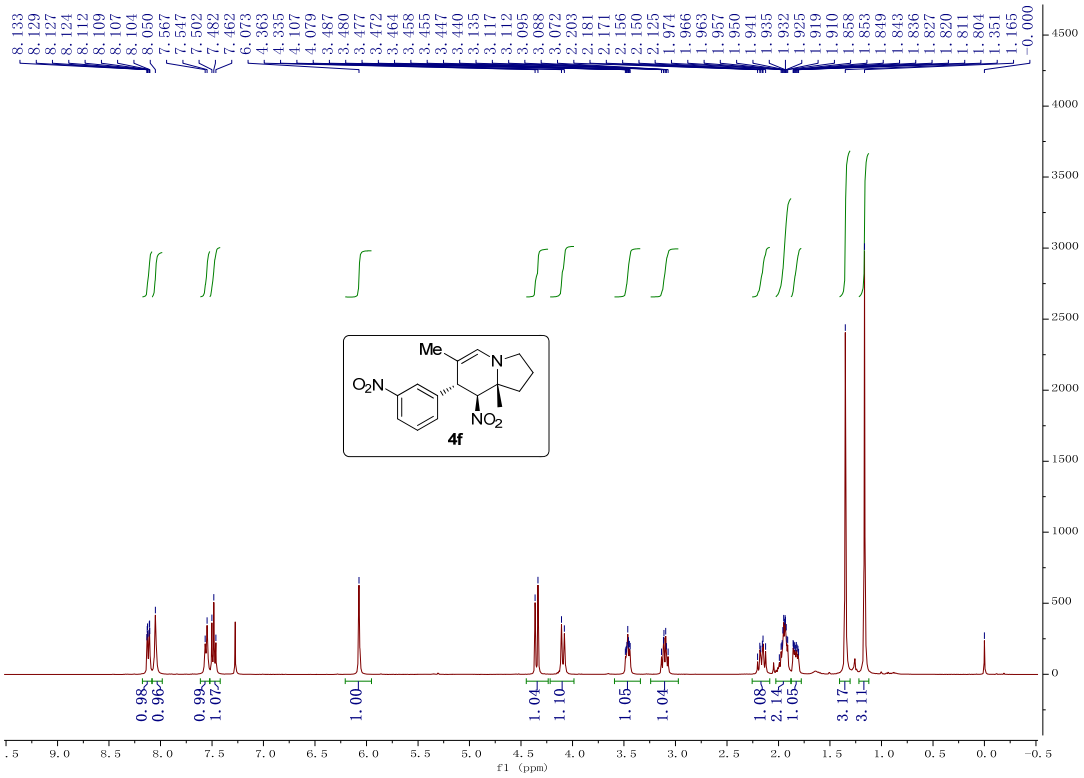
45.79
37.76
23.24
19.20
17.61
Me
N
NO2
Br
4e
f1 (ppm)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8.127
8.124
8.112
8.109
8.107
8.104
8.050
7.567
7.547
7.502
7.482
7.462
6.073
4.363
4.335
4.107
4.079
3.487
3.480
3.477
3.472
3.464
3.458
3.455
3.447
3.440
3.135
3.117
3.112
3.095
3.088
3.072
2.203
2.181
2.171
2.156
2.150
2.125
1.974
1.966
1.963
1.957
1.950
1.941
1.935
1.932
1.925
1.919
1.910
1.858
1.853
1.849
1.843
1.836
1.827
1.820
1.811
1.804
1.351
1.165
0.000
Me
N
O2N
NO2
4f
0.98
0.96
0.99
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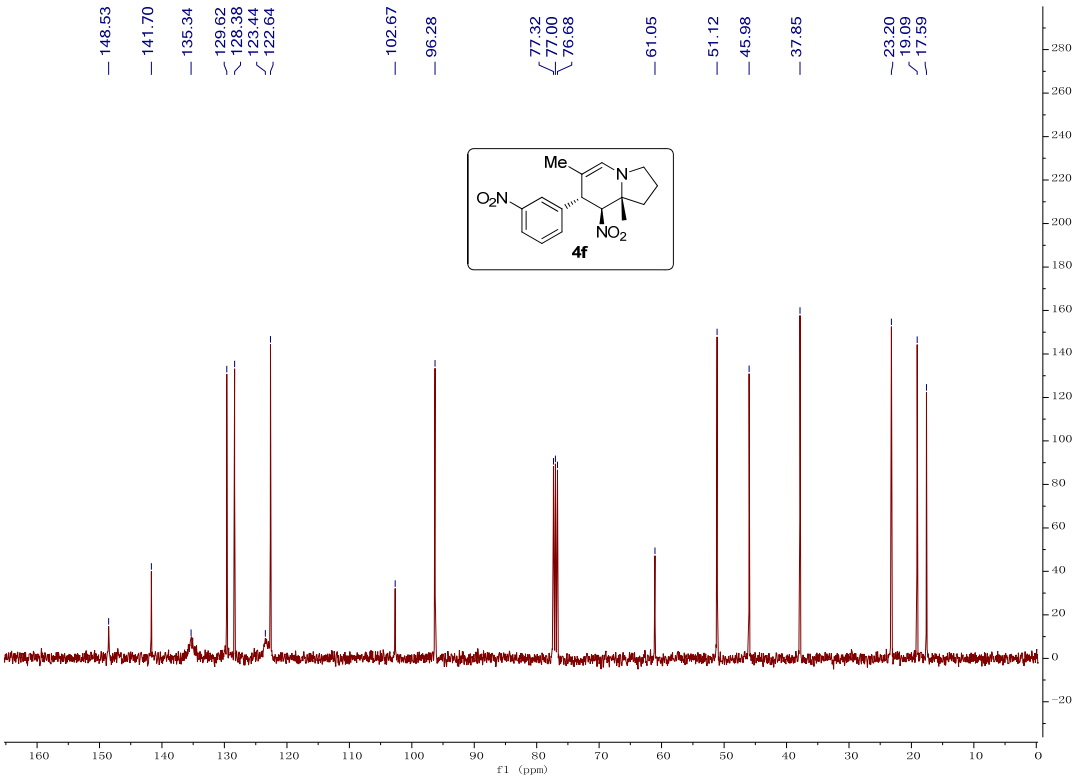
1.10
1.05
1.04
1.08
2.14
1.05
3.17
3.11
f1 (ppm)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77.00
76.68
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37.85
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19.09
17.59
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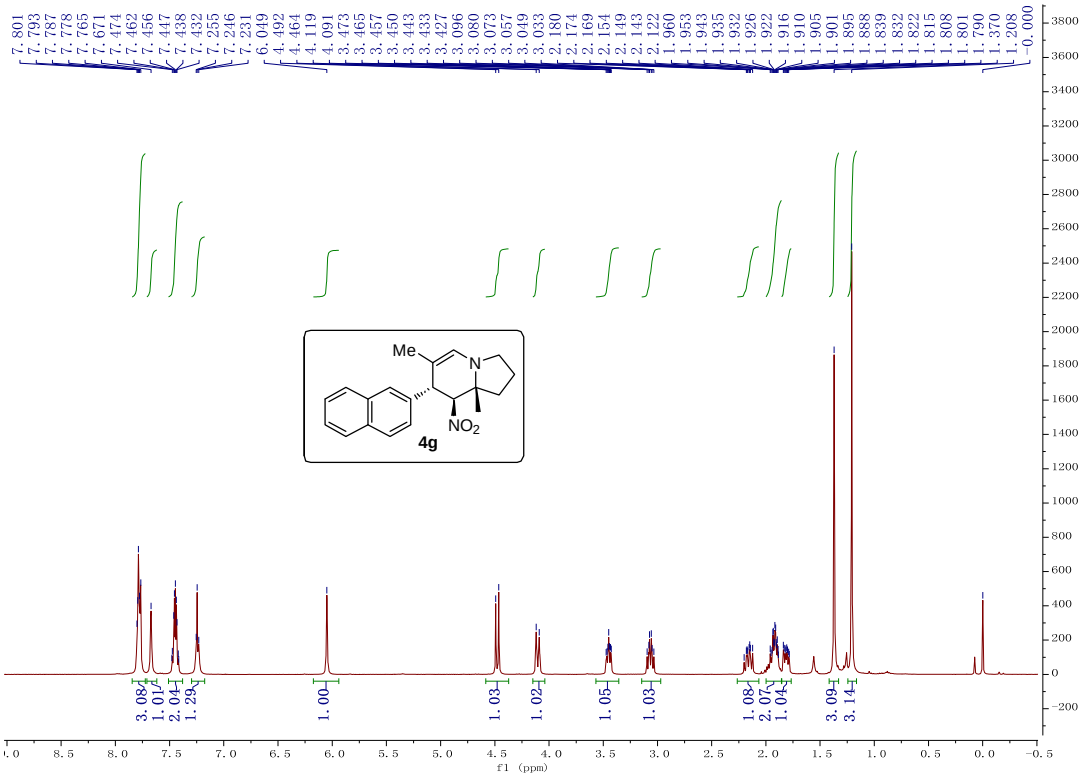
O2N
NO2
4f
f1 (ppm)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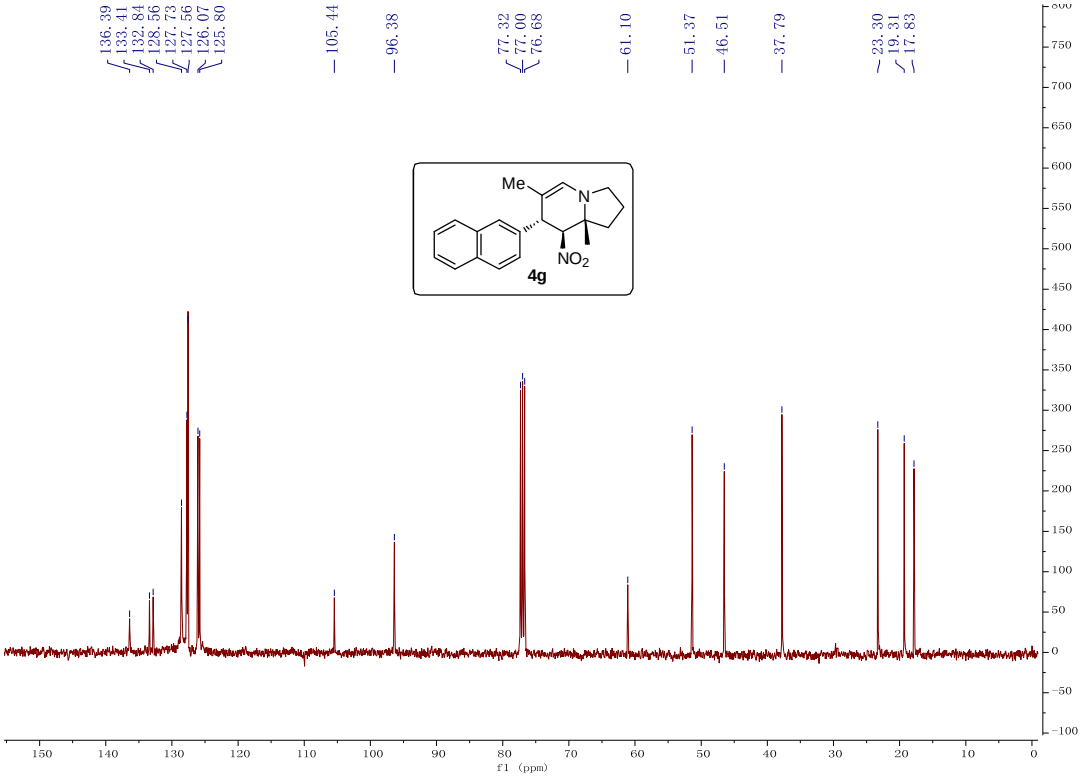
4g
f1 (ppm)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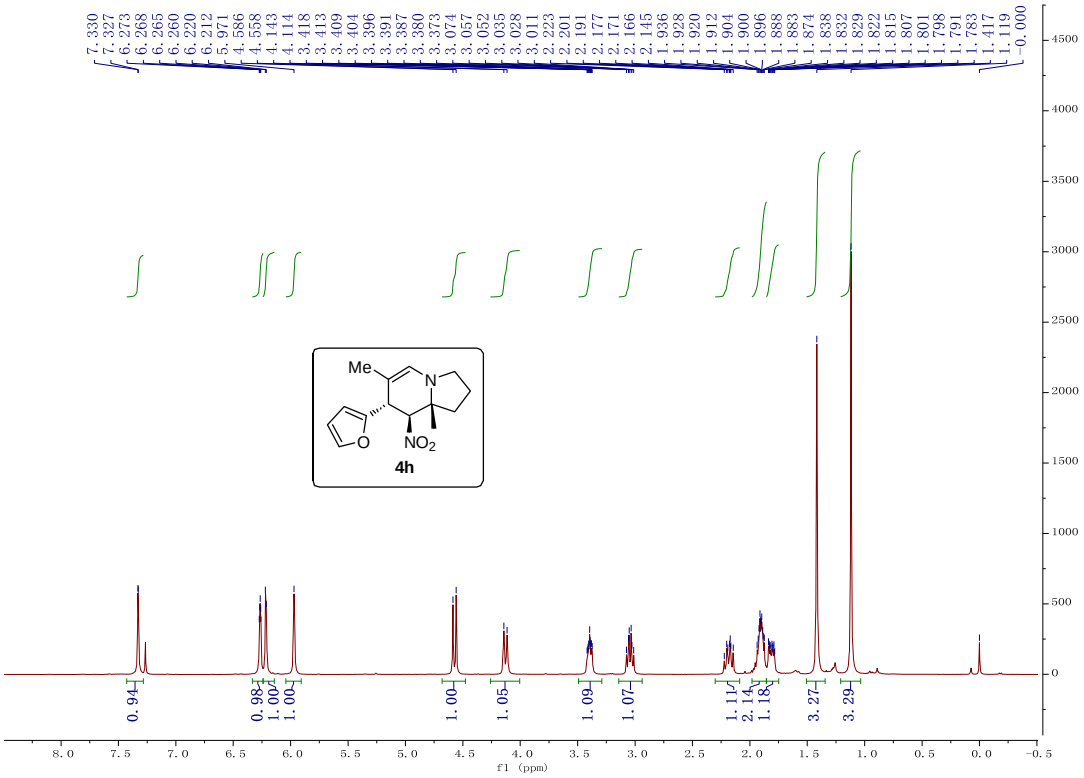
N
NO2
4g
f1 (ppm)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4.114
3.418
3.413
3.409
3.404
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3.391
3.387
3.380
3.373
3.074
3.057
3.052
3.035
3.028
3.011
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2.201
2.191
2.177
2.171
2.166
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1.928
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1.888
1.883
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1.829
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O
NO2
4h
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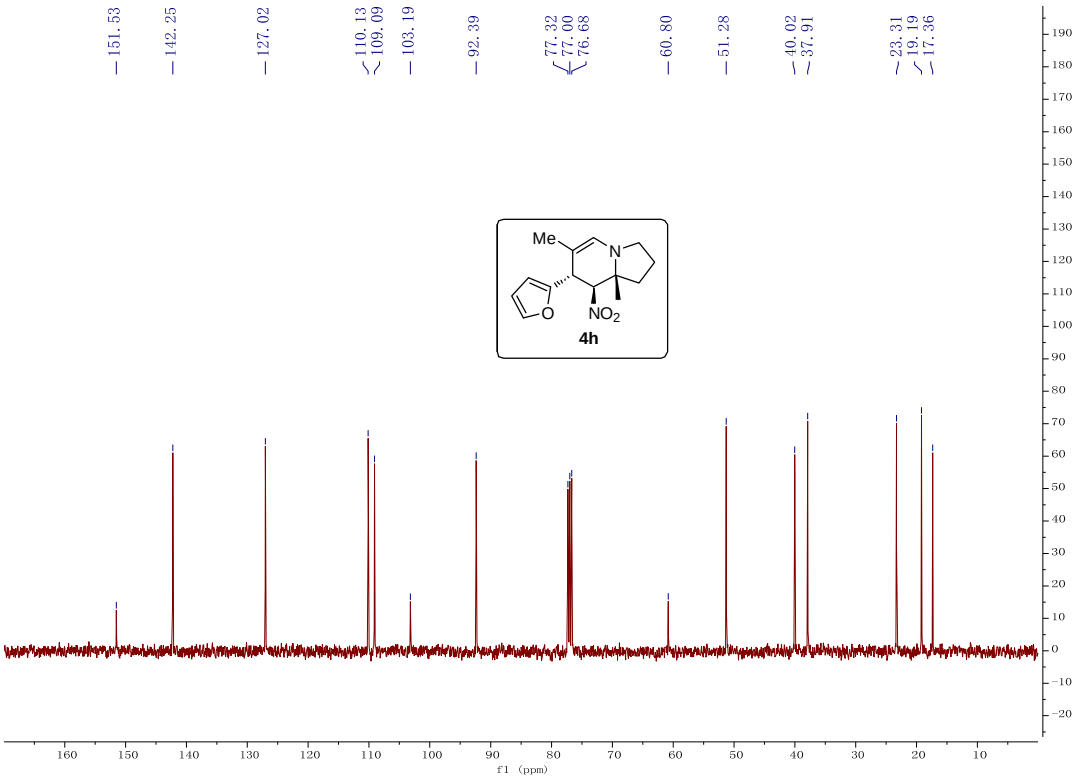
3.29
f1 (ppm)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37.91
23.31
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17.36
Me
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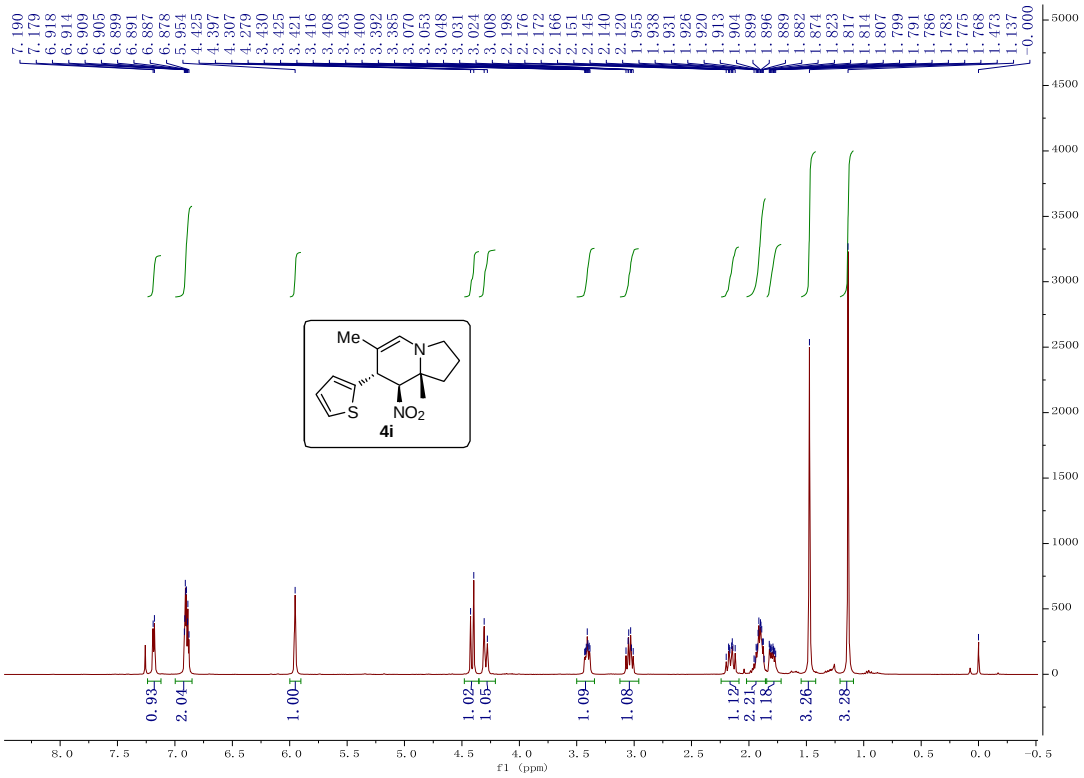
O
NO2
4h
f1 (ppm)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S
NO2
4i
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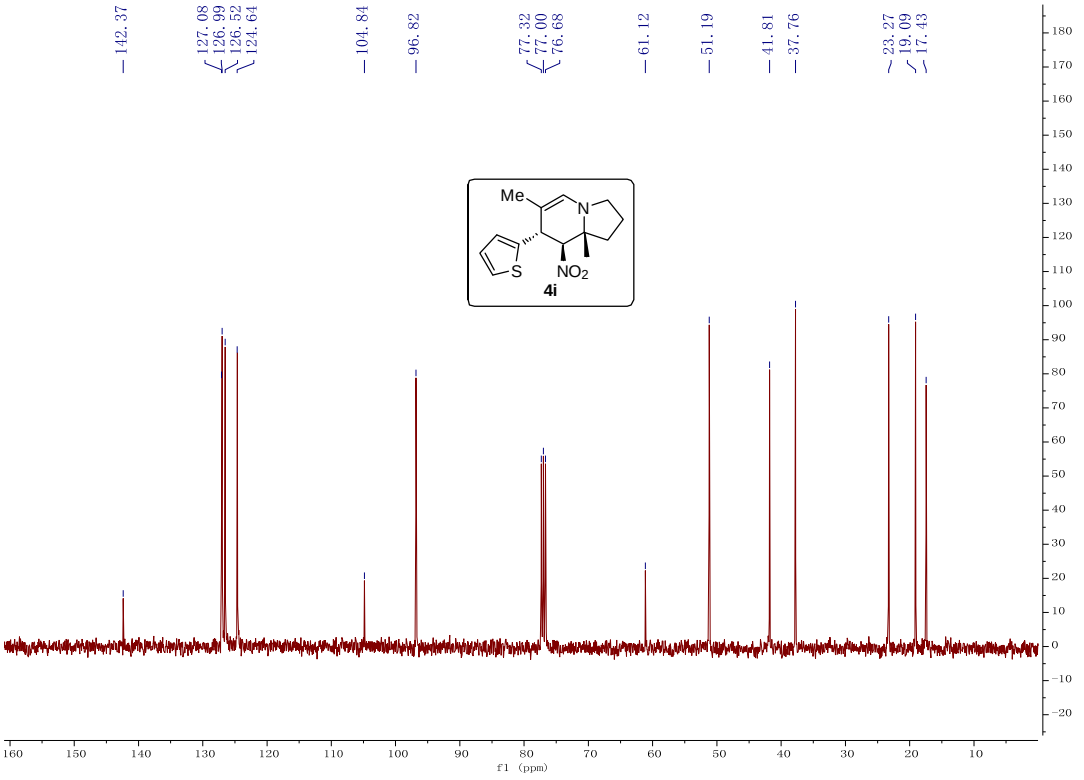
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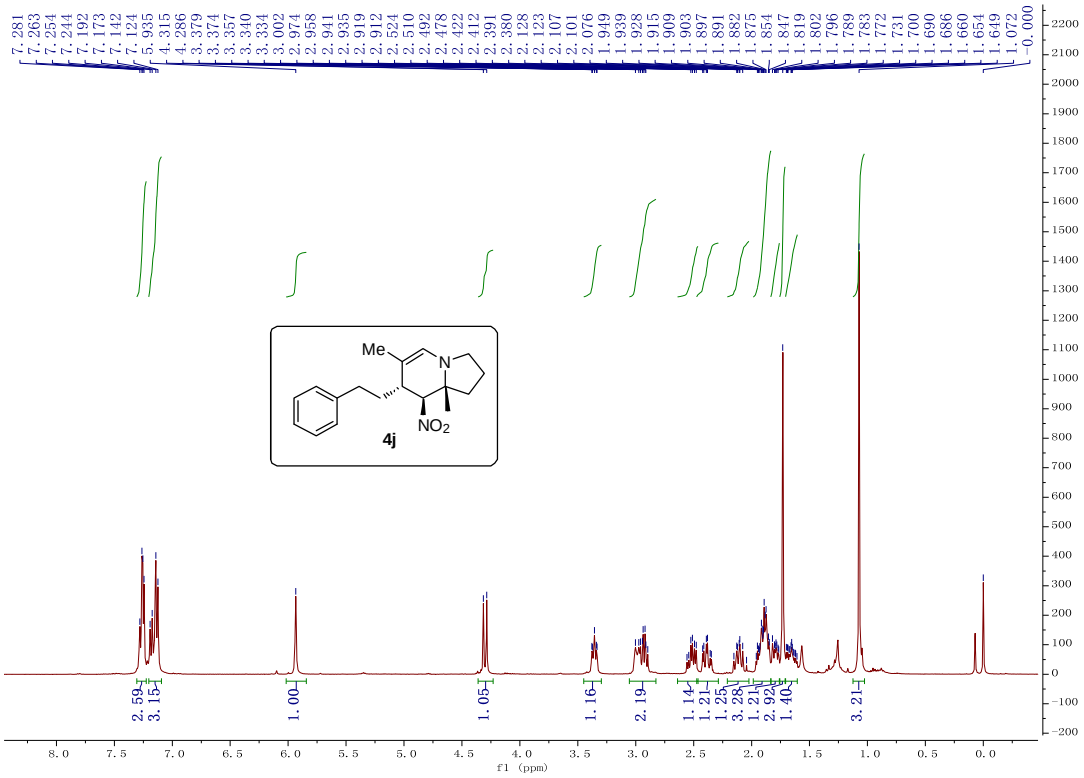
S
NO2
4i
f1 (ppm)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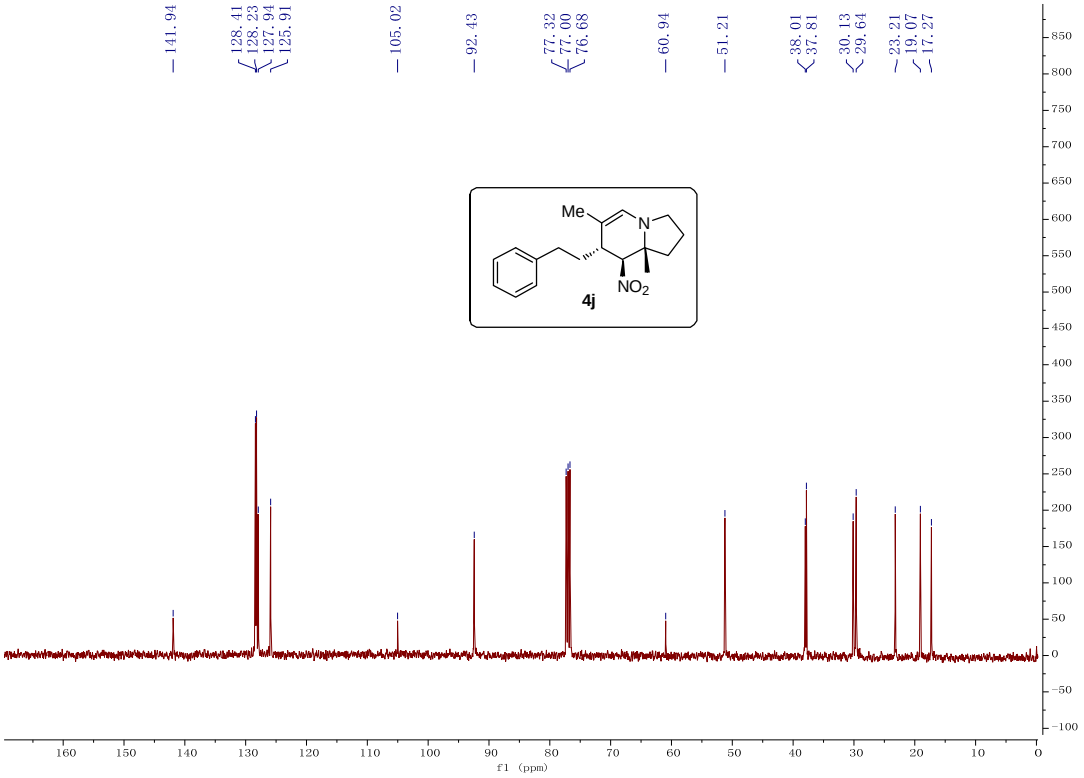
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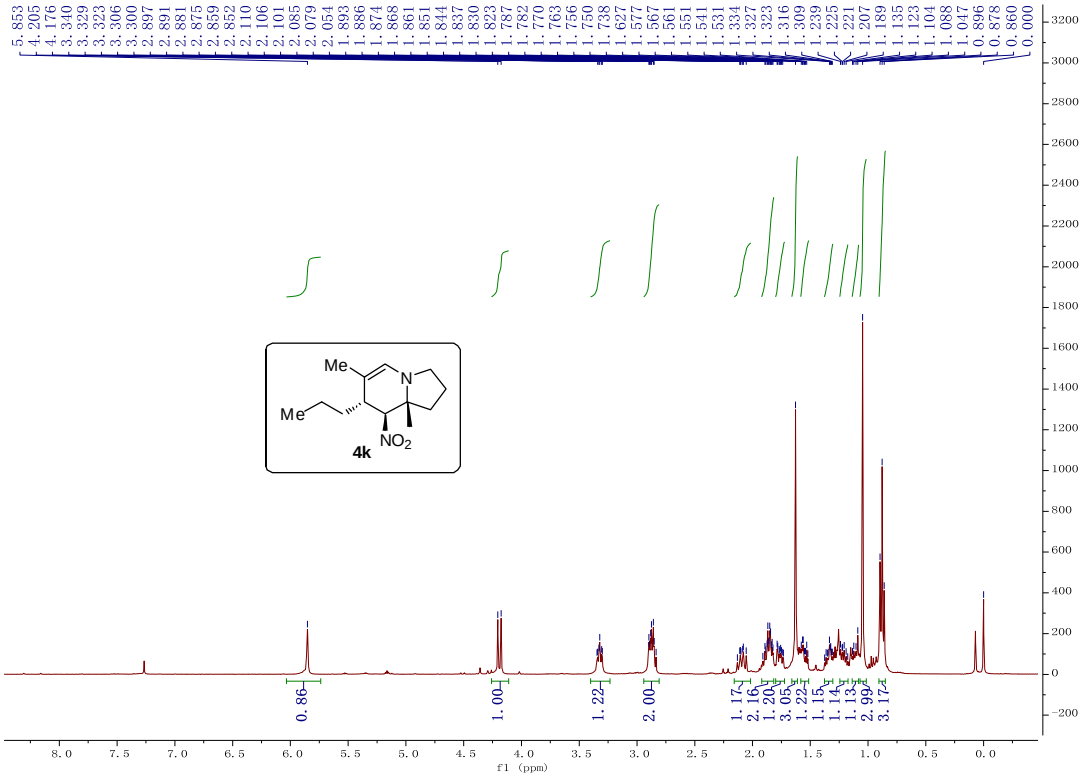
NO2
4j
f1 (ppm)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4k
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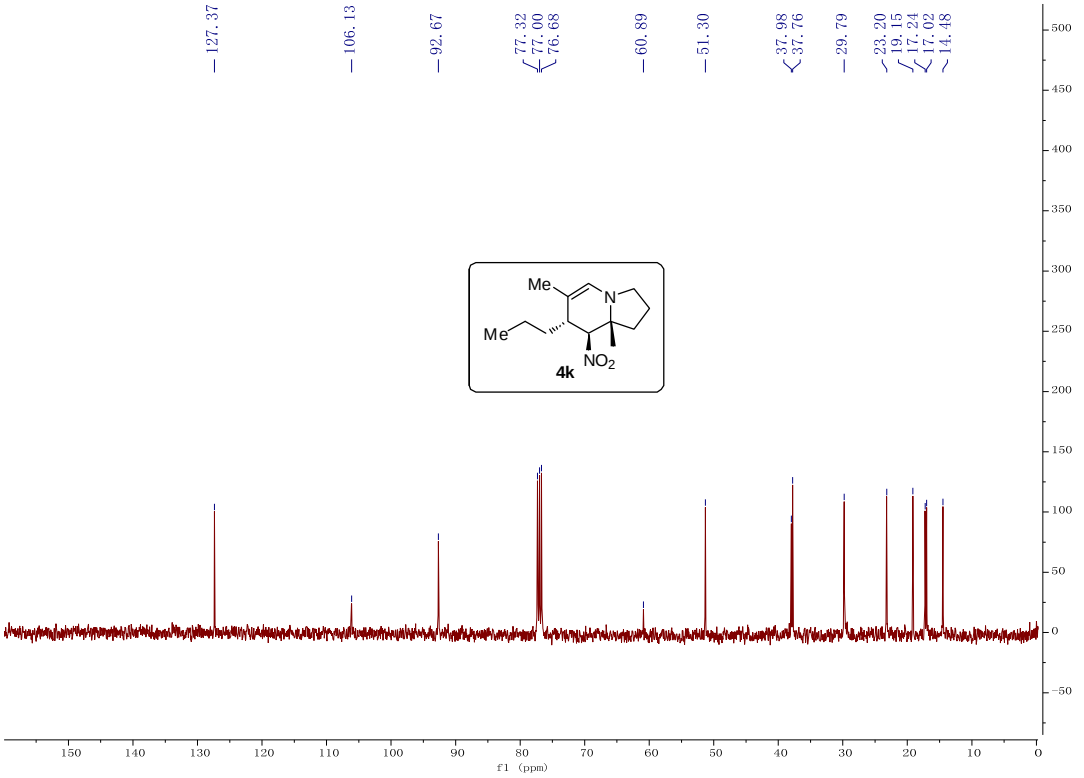
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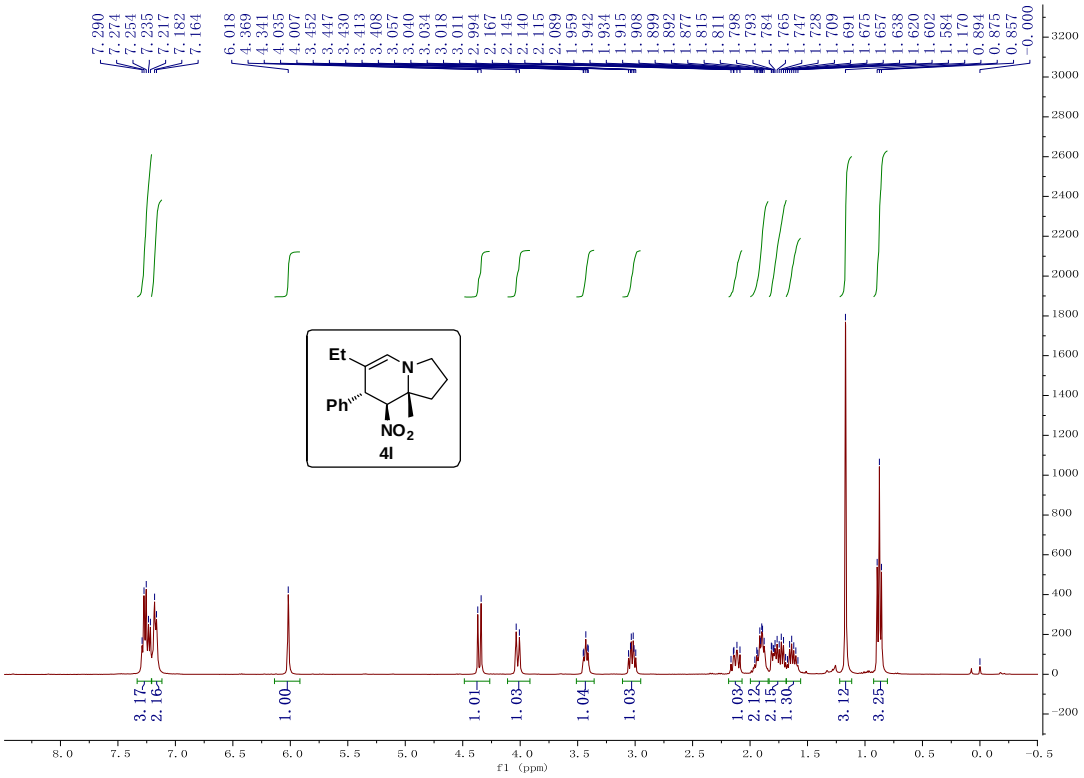
Me
NO2
4k
f1 (ppm)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4.035
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3.447
3.430
3.413
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3.040
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3.018
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1.620
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1.584
1.170
0.894
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0.857
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Et
N
Ph
NO2
4l
3.17
2.16
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1.04
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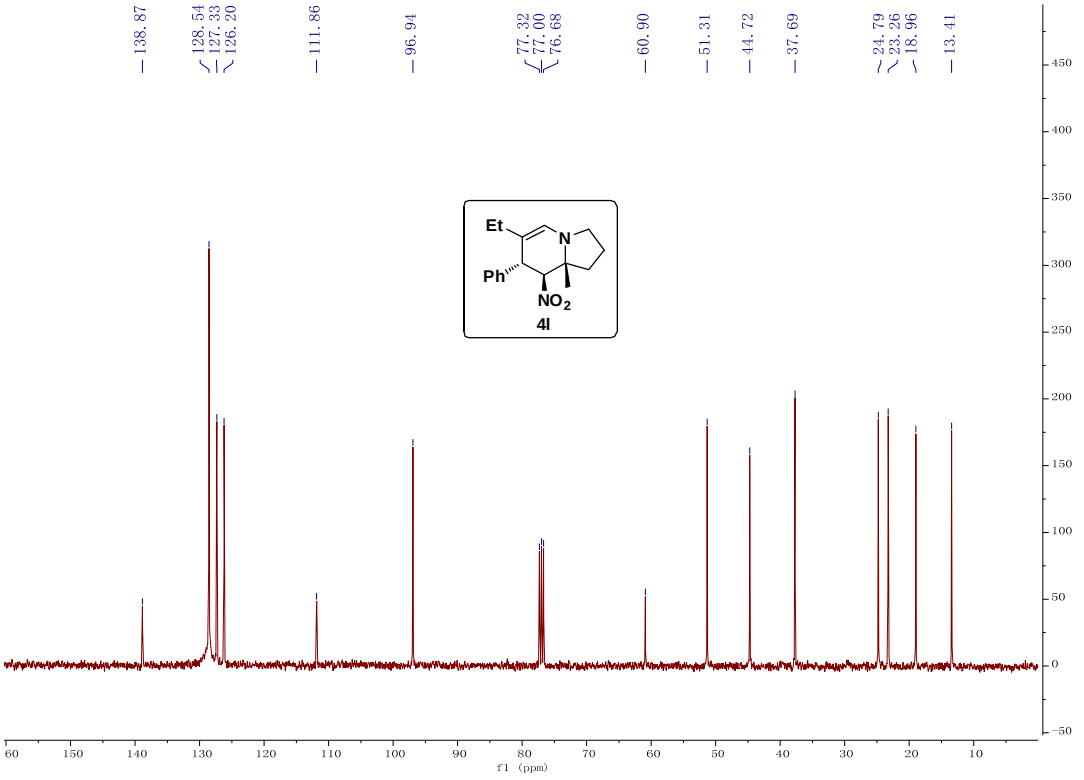
1.30
3.12
3.25
f1 (ppm)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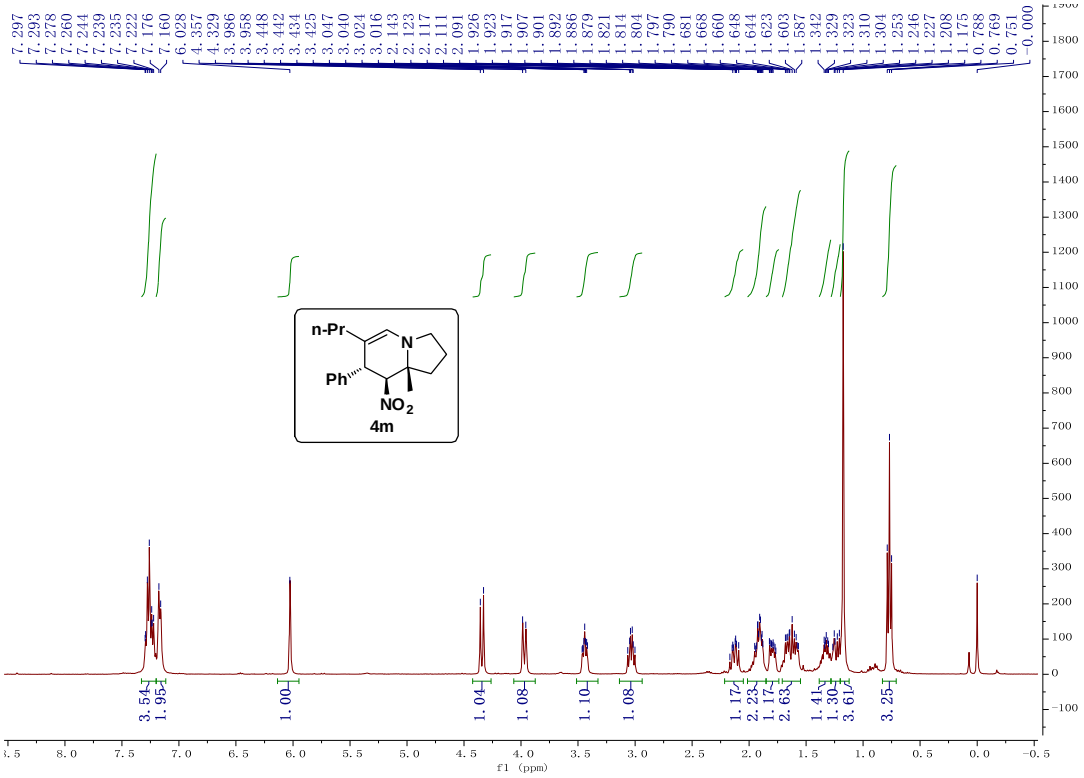
24.79
23.26
18.96
13.41
Et
N
Ph
NO2
4l
f1 (ppm)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3.958
3.448
3.442
3.434
3.425
3.047
3.040
3.024
3.016
2.143
2.123
2.117
2.111
2.091
1.926
1.923
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1.907
1.901
1.892
1.886
1.879
1.821
1.814
1.804
1.797
1.790
1.681
1.668
1.660
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1.644
1.623
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1.587
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1.329
1.323
1.310
1.304
1.253
1.246
1.227
1.208
1.175
0.788
0.769
0.751
-0.000
n-Pr
N
Ph
NO2
4m
3.54
1.95
1.00
1.04
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1.17
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1.30
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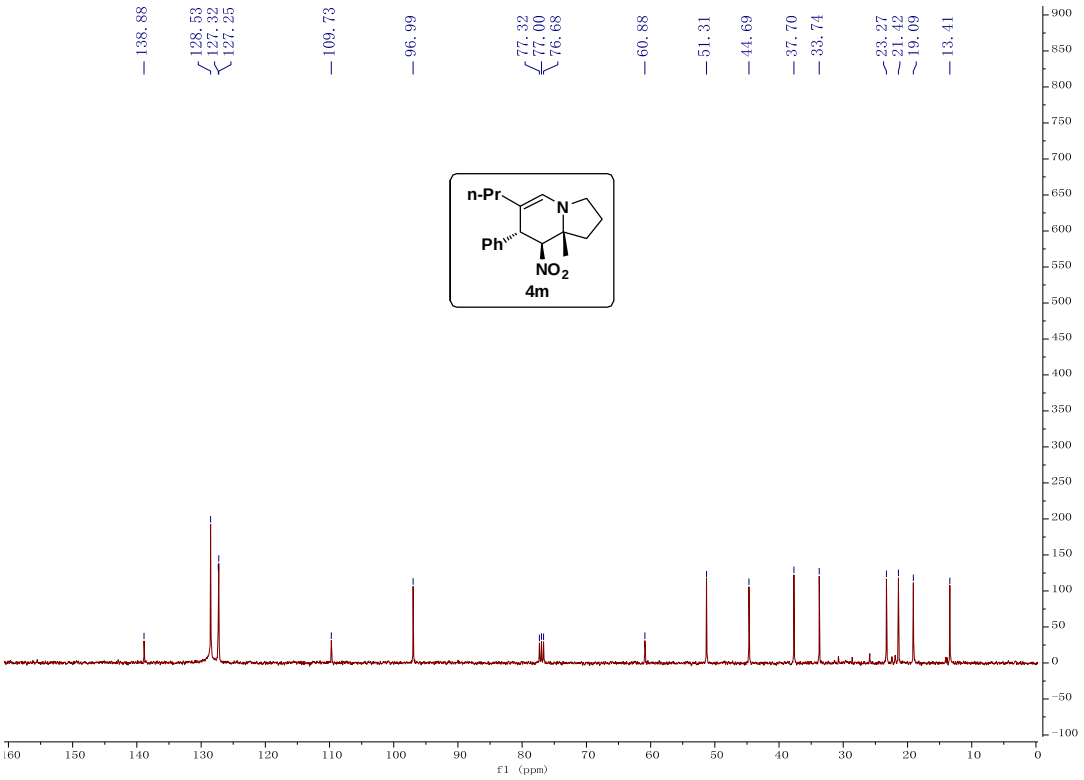
3.25
f1 (ppm)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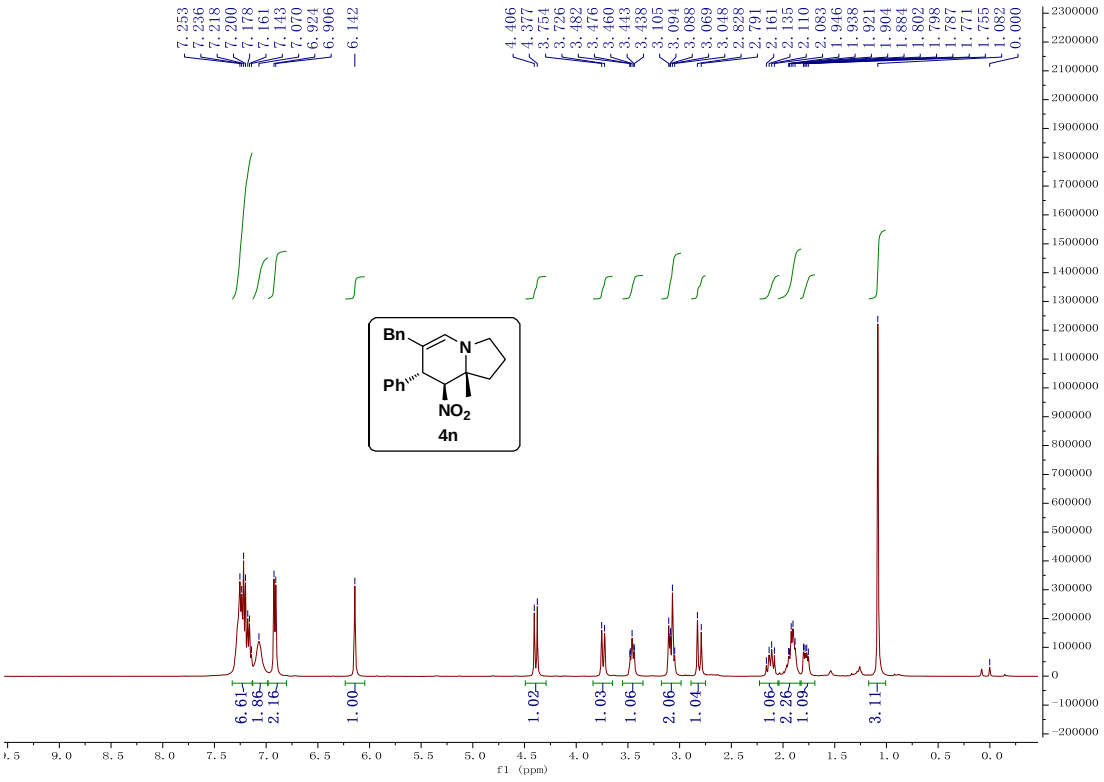
13.41
n-Pr
N
Ph
NO2
4m
f1 (ppm)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4.377
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3.726
3.482
3.476
3.460
3.443
3.438
3.105
3.094
3.088
3.069
3.048
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2.083
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1.938
1.921
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1.787
1.771
1.755
1.082
0.000
Bn
Ph
NO2
4n
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1.06
2.06
1.04
1.06
2.26
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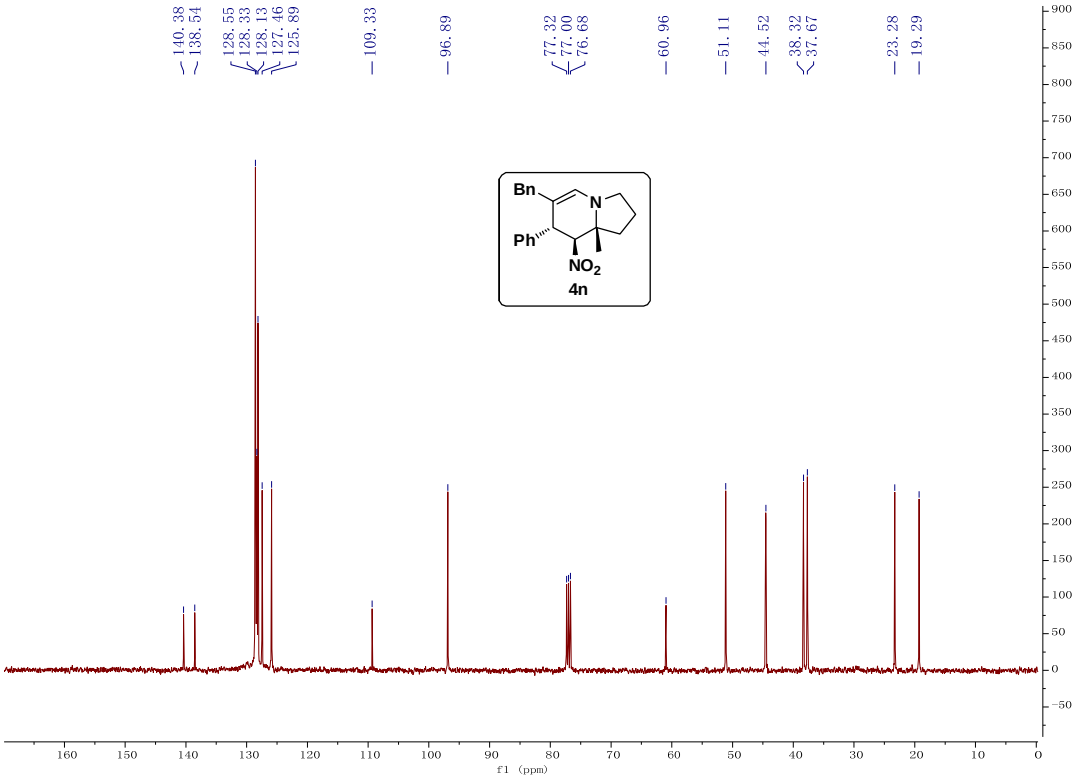
f1 (ppm)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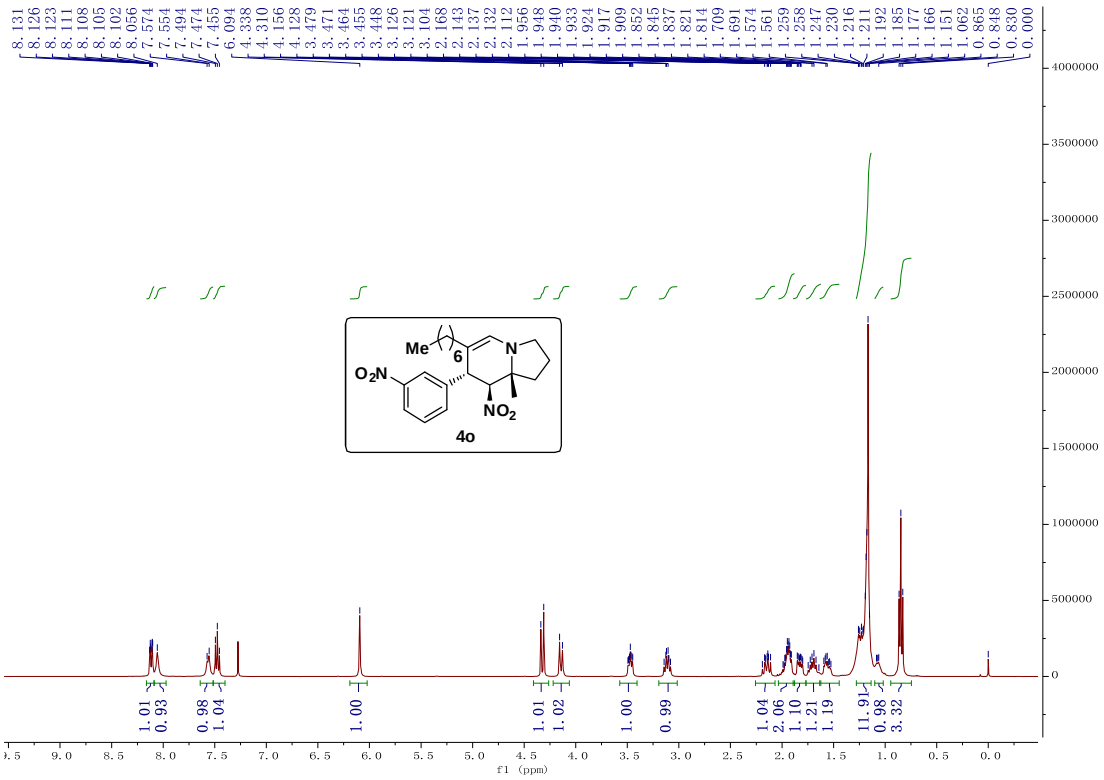
44.52
38.32
37.67
23.28
19.29
Bn
Ph
NO2
4n
f1 (ppm)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8.056
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7.494
7.474
7.455
6.094
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4.310
4.156
4.128
3.479
3.471
3.464
3.455
3.448
3.126
3.121
3.104
2.168
2.143
2.137
2.132
2.112
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1.948
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1.917
1.909
1.852
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1.837
1.821
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1.258
1.247
1.230
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1.211
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Me
6
N
O2N
NO2
4o
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0.93
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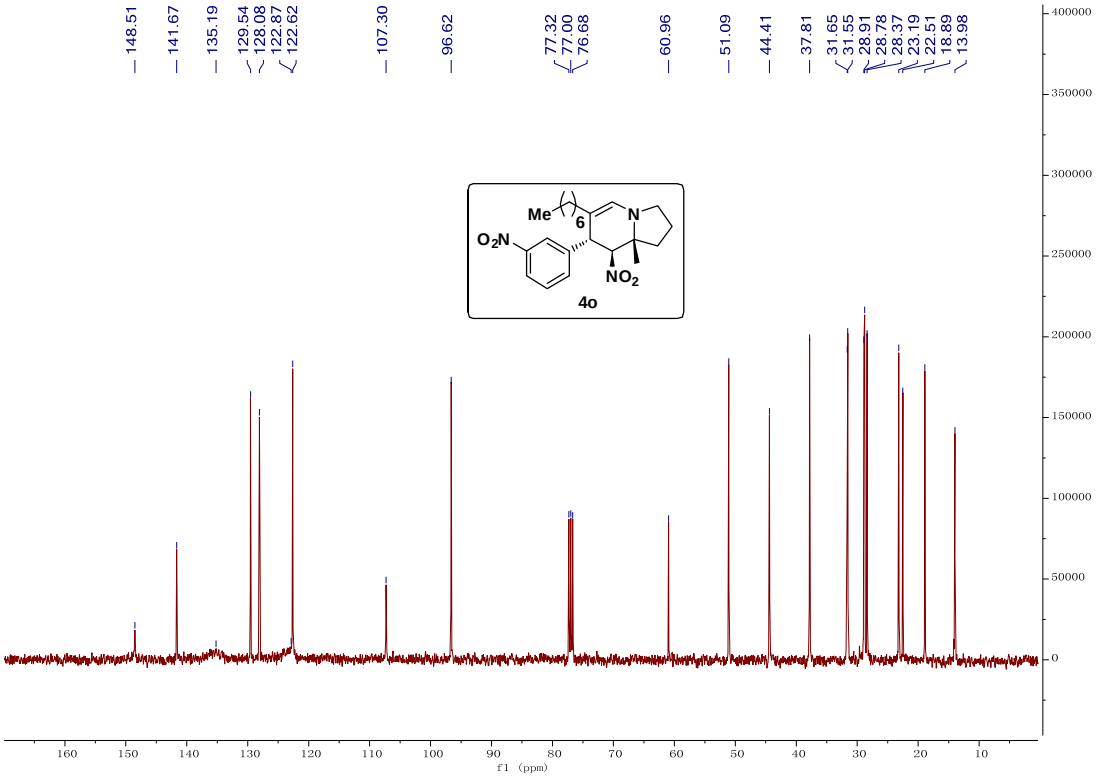
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51.09
44.41
37.81
31.65
31.55
28.91
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22.51
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Me
6
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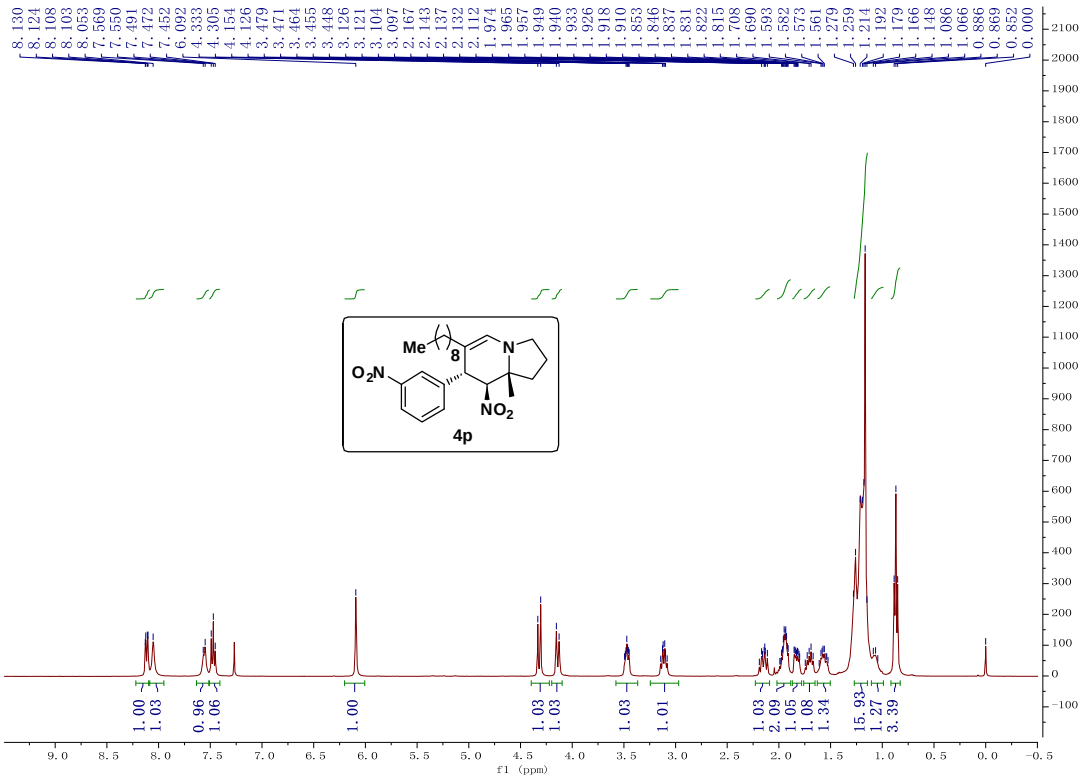
O2N
NO2
4o
f1 (ppm)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3.455
3.448
3.126
3.121
3.104
3.097
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2.143
2.137
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1.086
1.066
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0.869
0.852
0.000
Me
8
N
O2N
NO2
4p
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1.03
0.96
1.06
1.00
1.03
1.03
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15.93
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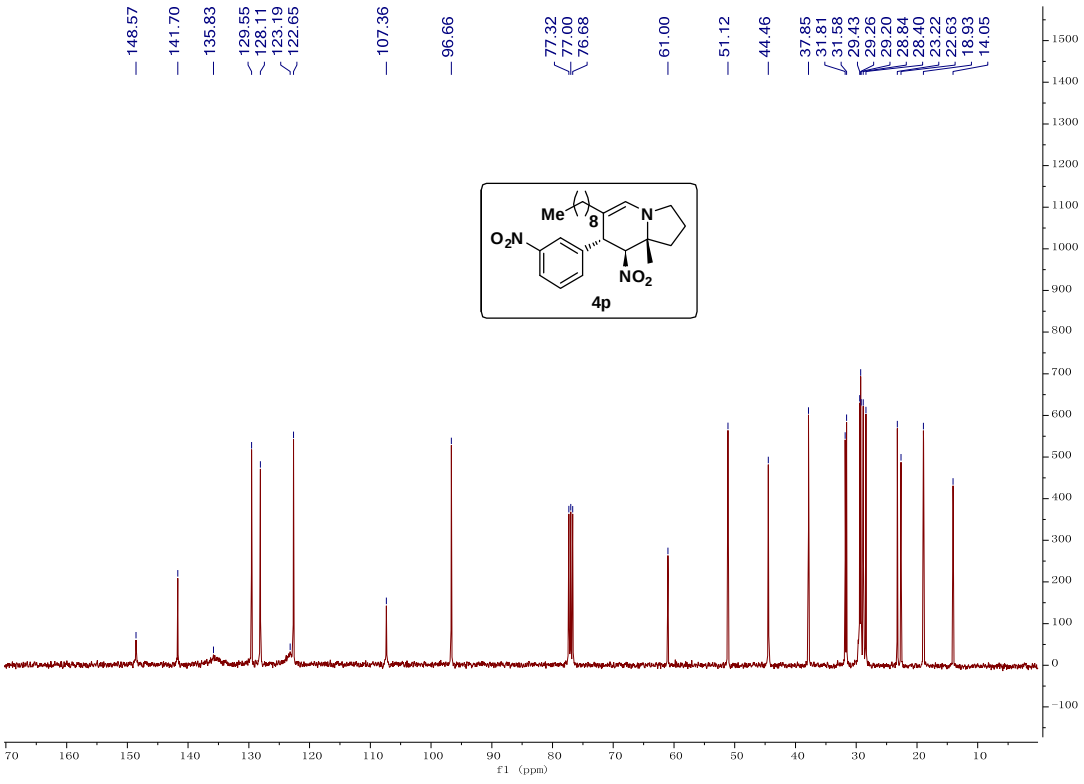
3.39
f1 (ppm)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37.85
31.81
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29.43
29.26
29.20
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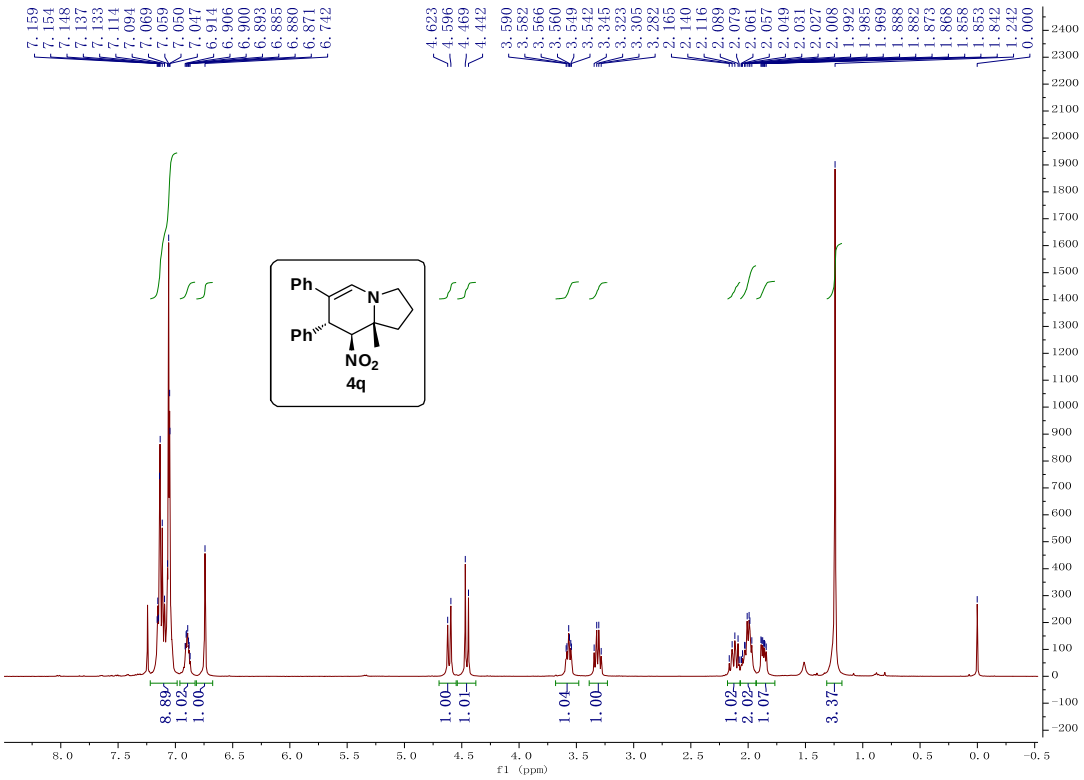
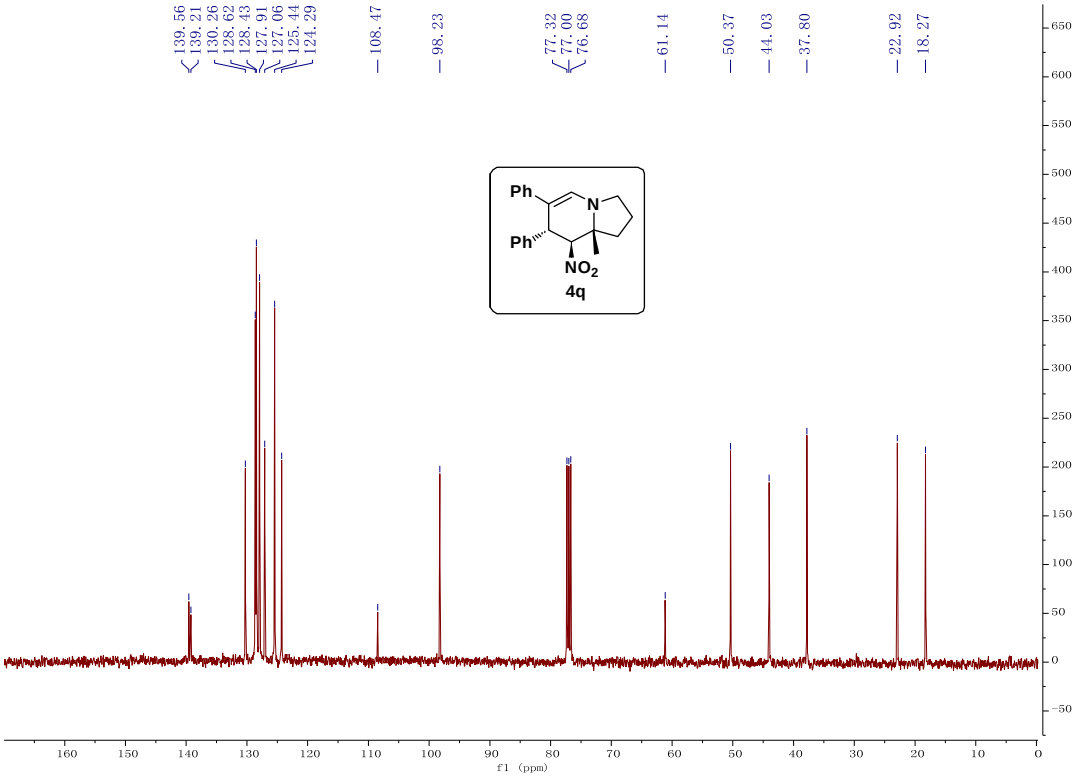
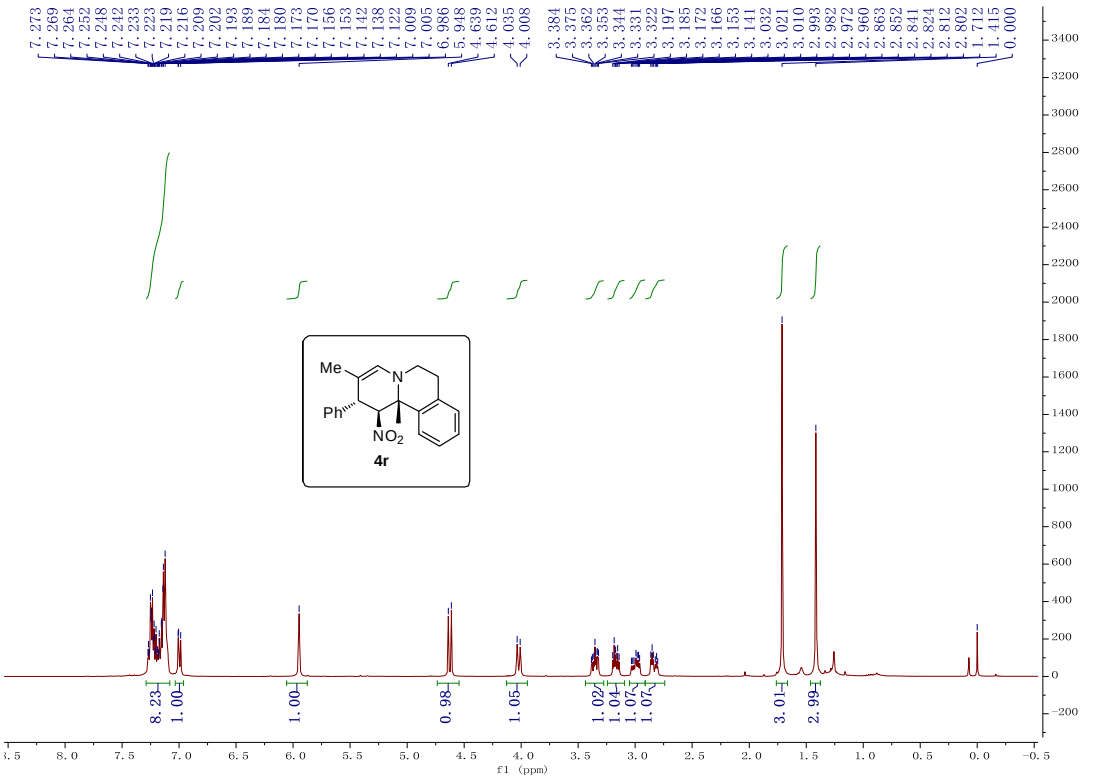
8
N
O2N
NO2
4p
f1 (ppm)


Ph
Ph
N
NO2
4q
f1 (ppm)


Ph
Ph
N
NO2
4q
f1 (ppm)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Ph
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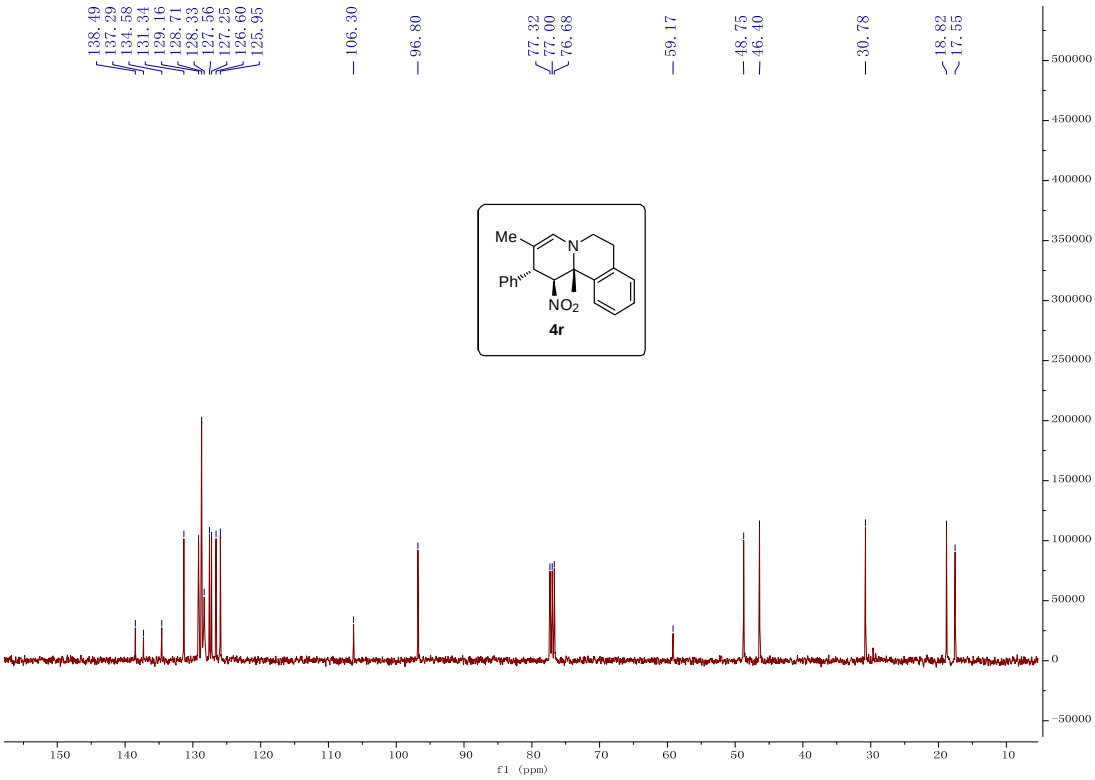
4r
f1 (ppm)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Ph
NO2
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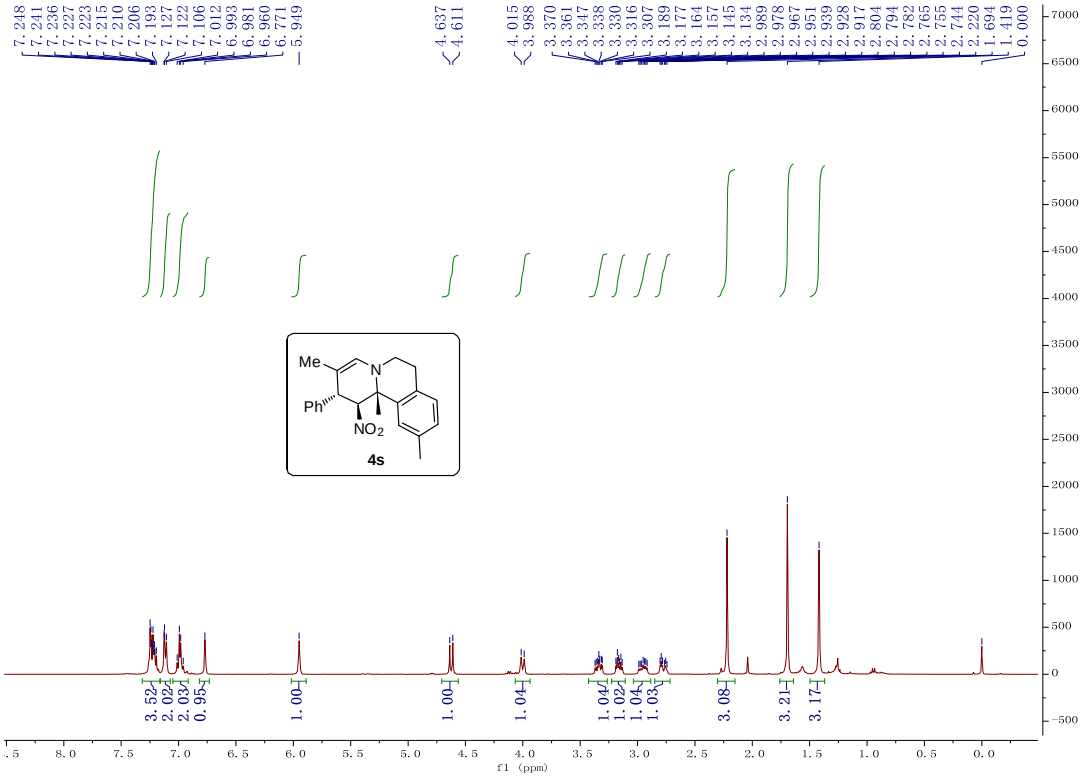
f1 (ppm)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4.015
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3.157
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1.419
0.000
Me
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Ph
NO2
4s
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2.03
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1.02
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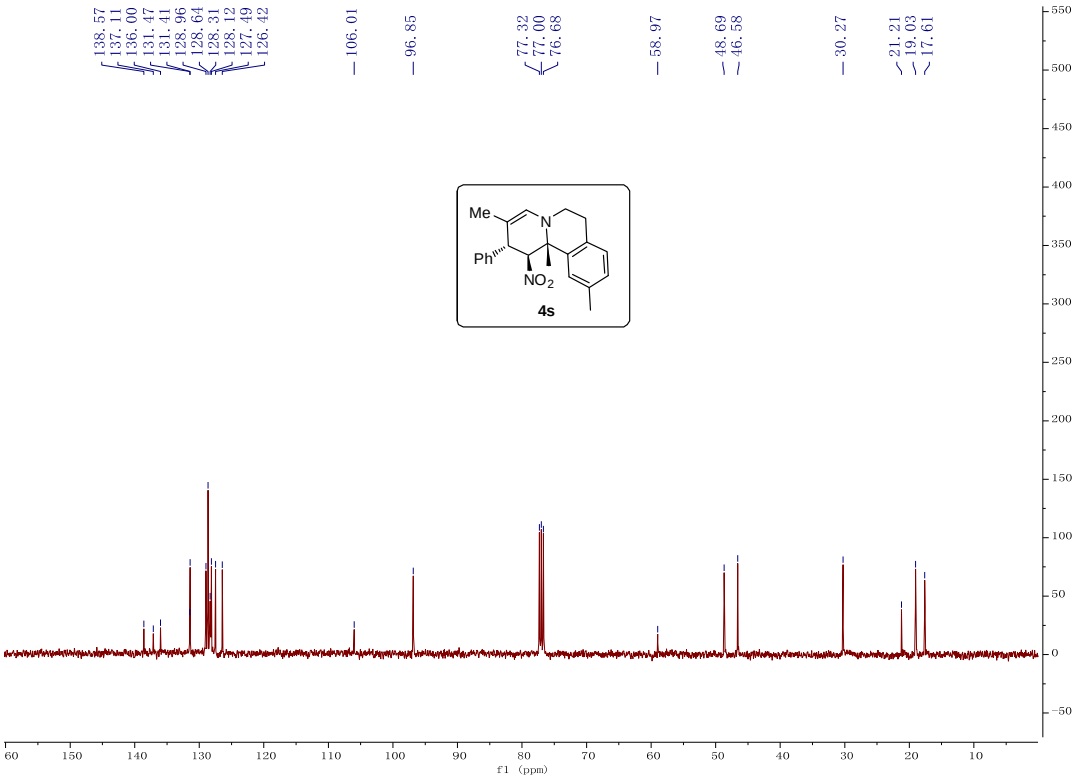
3.17
f1 (ppm)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127.49
126.42
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77.00
76.68
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30.27
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Me
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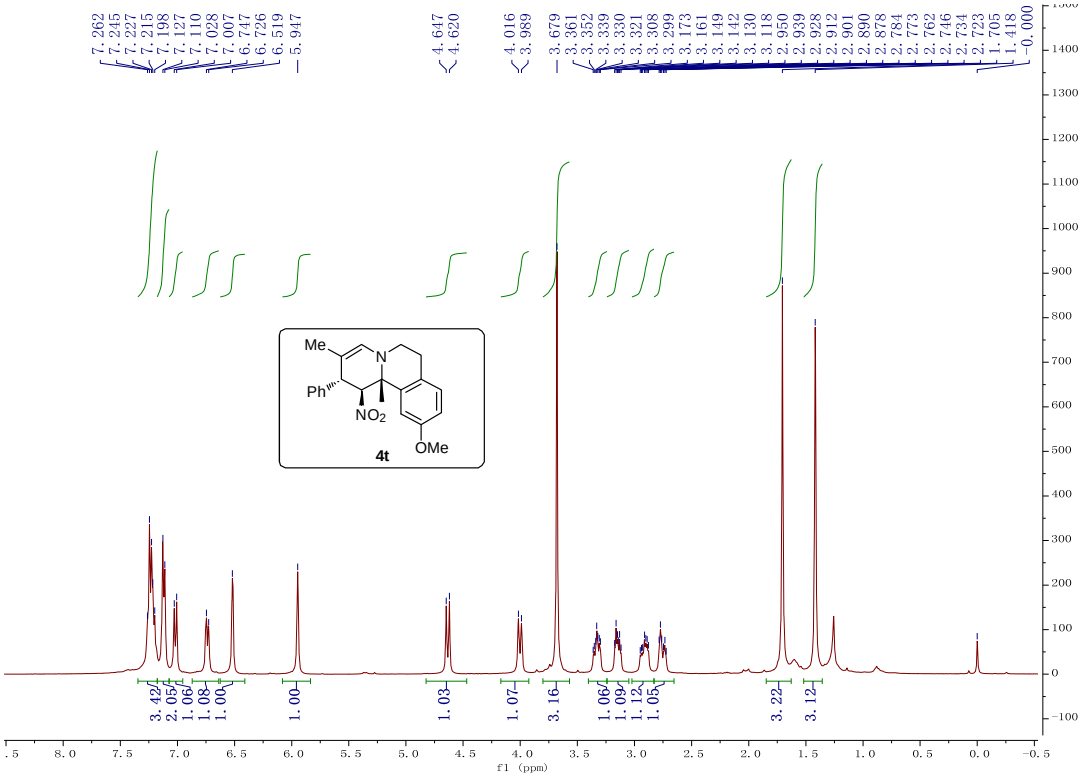
Ph
NO2
4s
f1 (ppm)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5.947
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0.000
Me
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Ph
NO2
OMe
4t
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2.05
1.06
1.08
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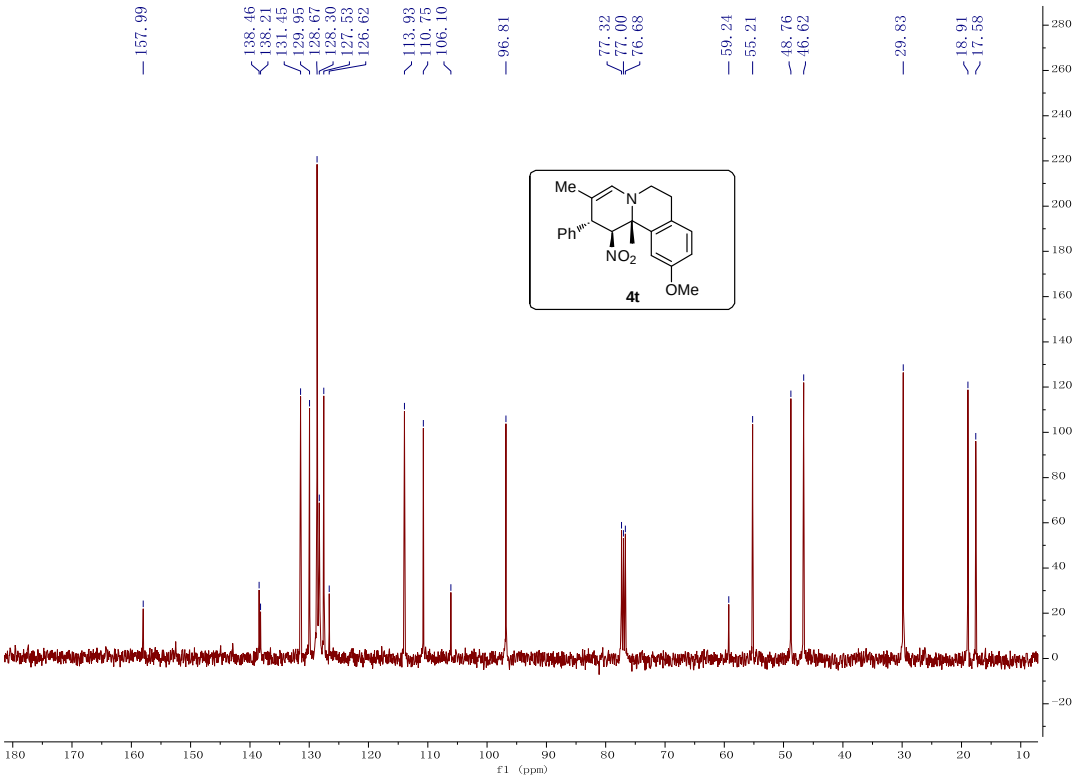
3.12
f1 (ppm)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77.00
76.68
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17.58
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N
Ph
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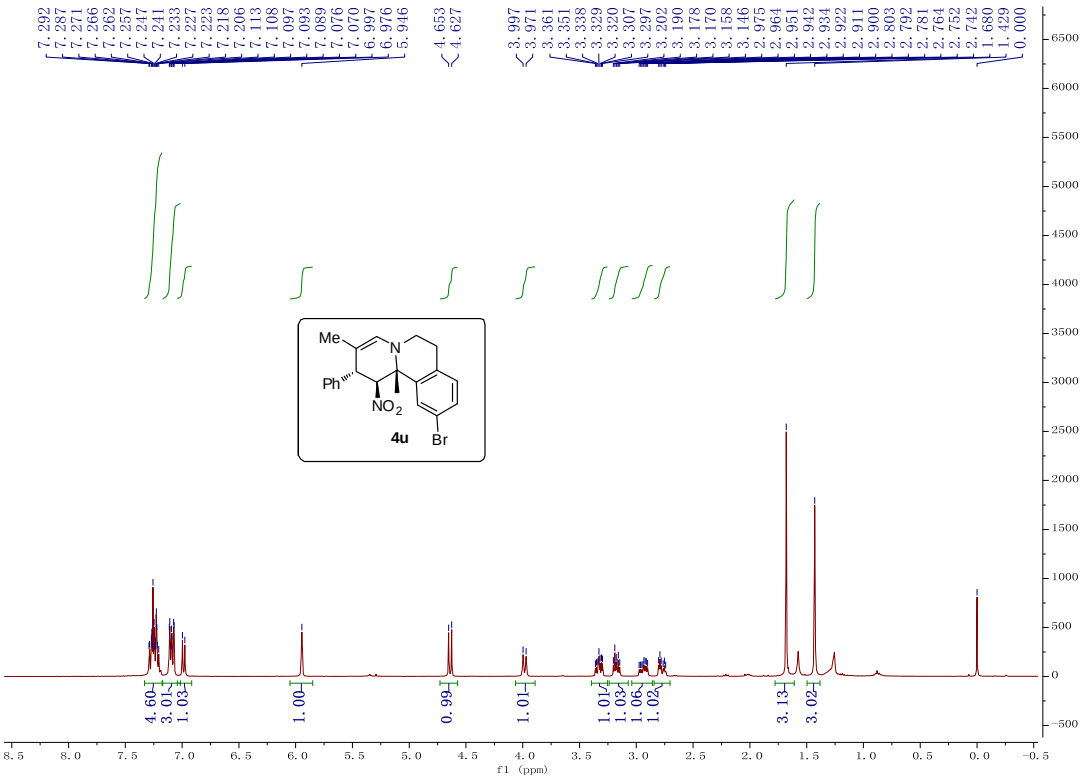
OMe
4t
f1 (ppm)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4.653 4.627
3.997 3.971 3.361 3.351 3.338 3.329 3.320 3.307 3.297 3.202 3.190 3.178 3.170 3.158 3.146 2.975 2.964 2.951 2.942 2.934 2.922 2.911 2.900 2.803 2.792 2.781 2.764 2.752 2.742 1.680 1.429 0.000
Me
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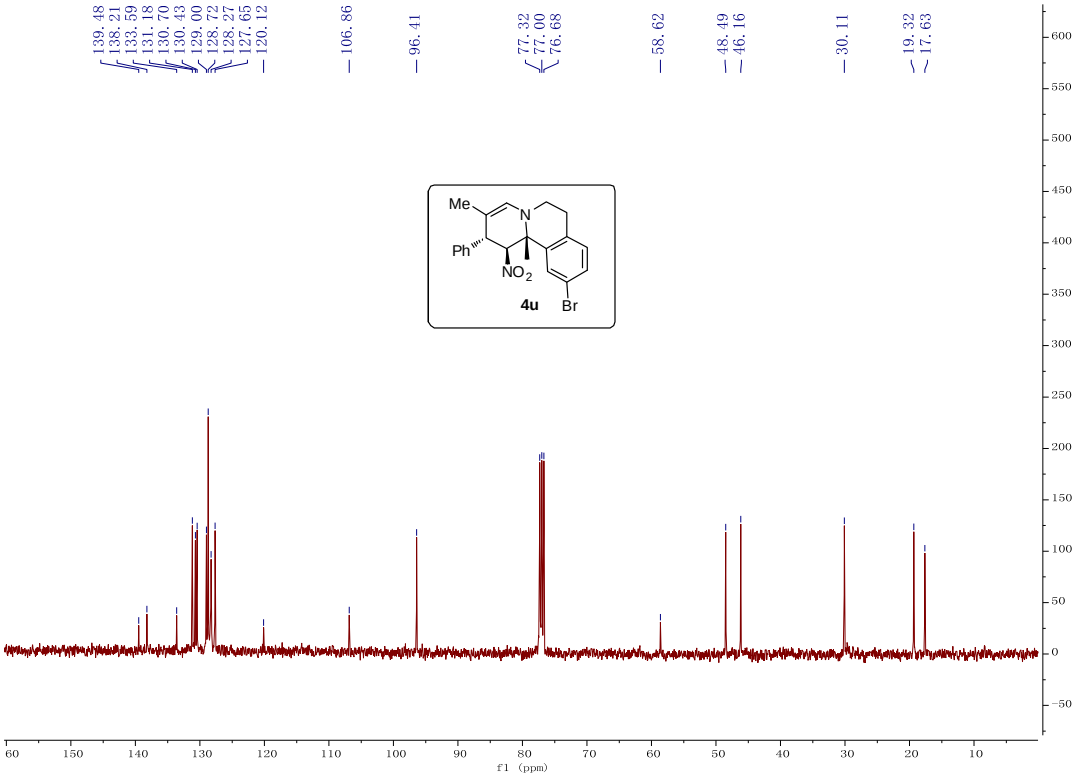
Ph
NO2
4u
Br
4.60 3.01 1.03 1.00 0.99 1.01 1.01 1.03 1.06 1.02 3.13 3.02
f1 (ppm)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106.86
96.41
77.32 77.00 76.68
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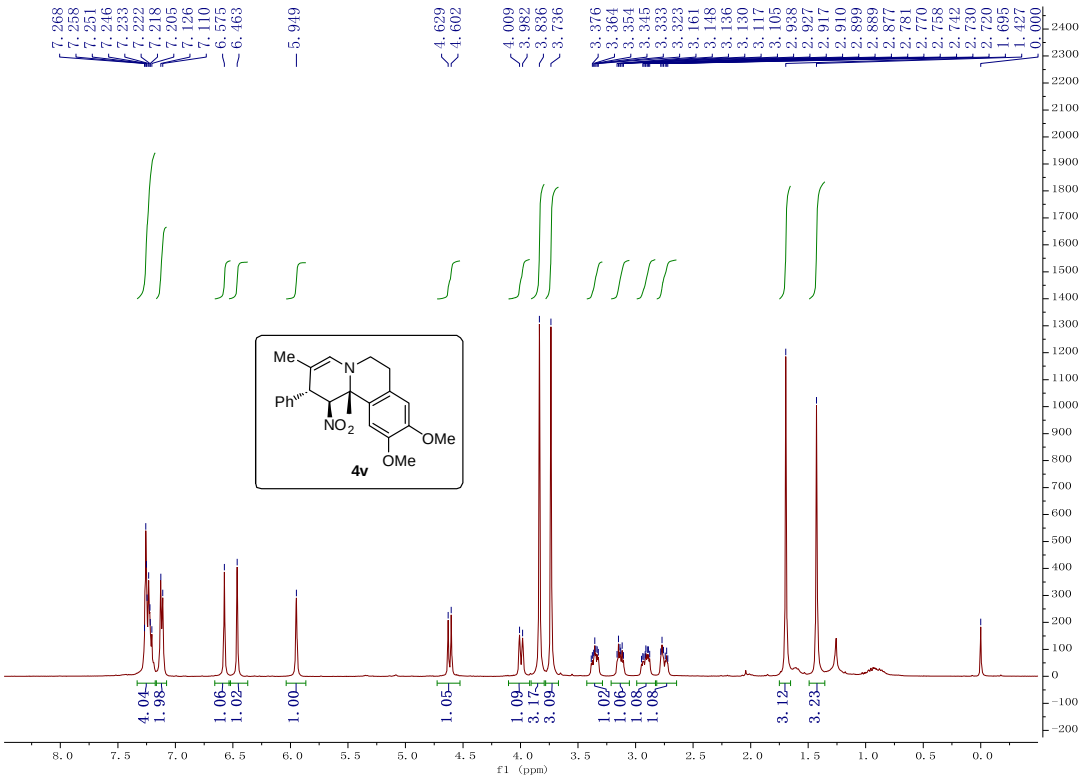
48.49 46.16
30.11
19.32 17.63
Me
N
Ph
NO2
4u
Br
f1 (ppm)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7.110
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4.629
4.602
4.009
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OMe
OMe
4v
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1.06
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1.08
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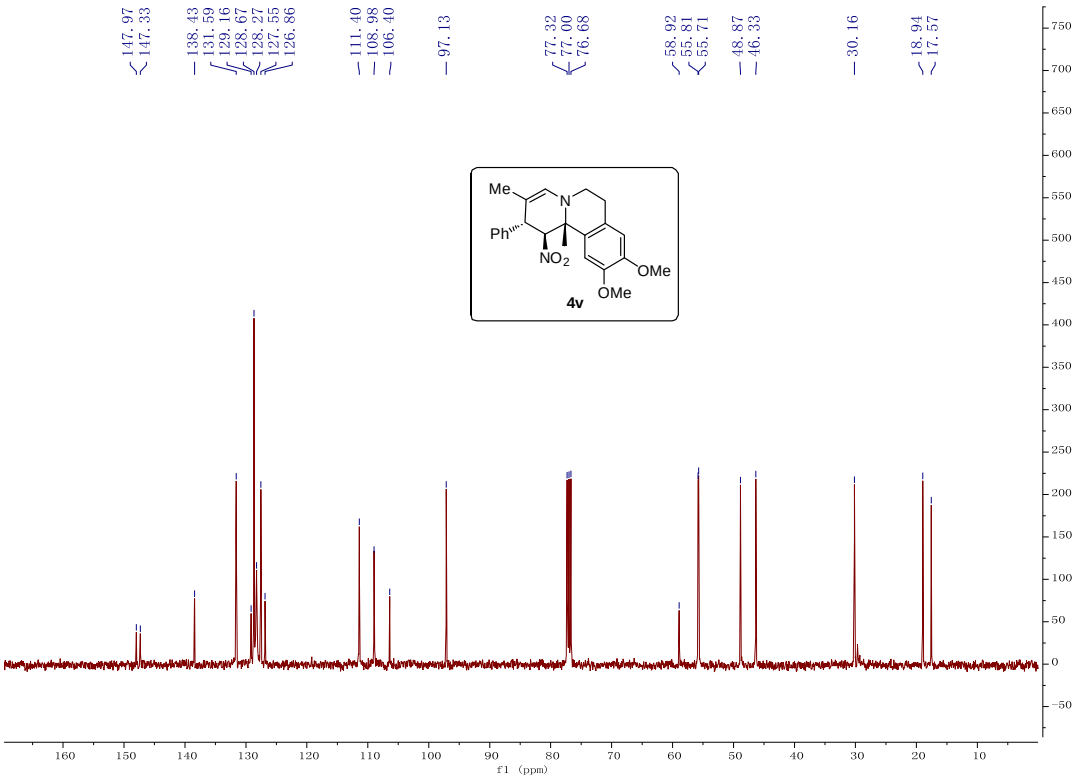
3.23
f1 (ppm)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77.00
76.68
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Ph
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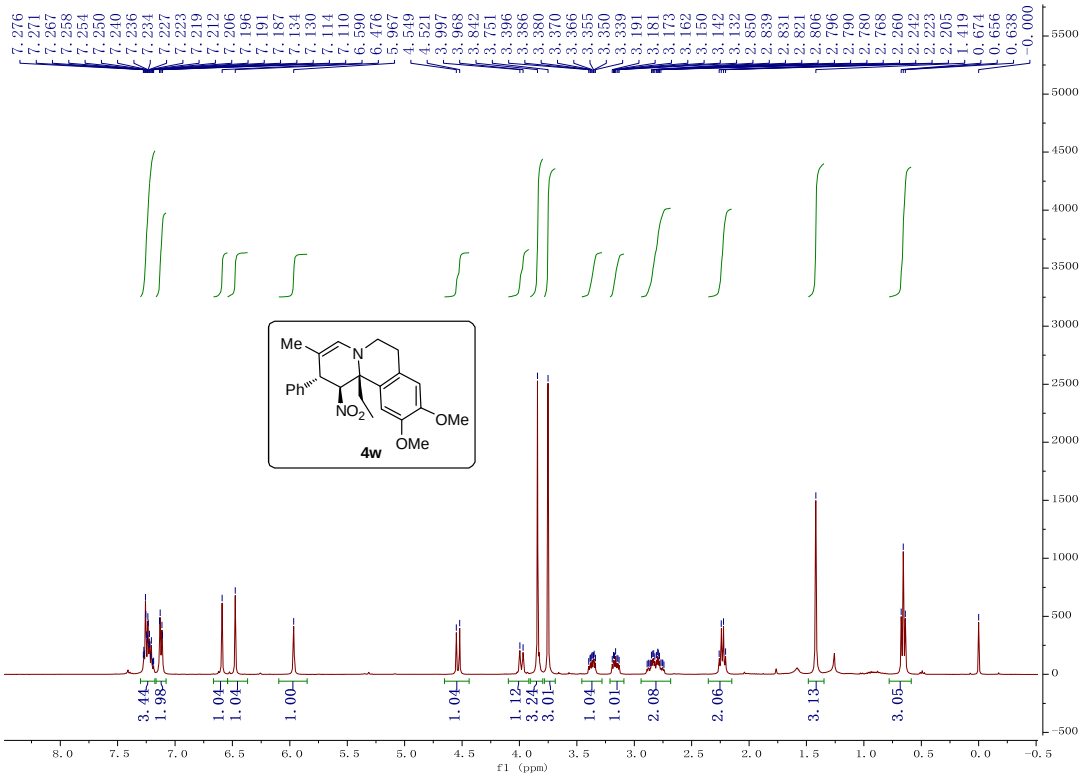
OMe
4v
f1 (ppm)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Ph
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OMe
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1.04
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2.06
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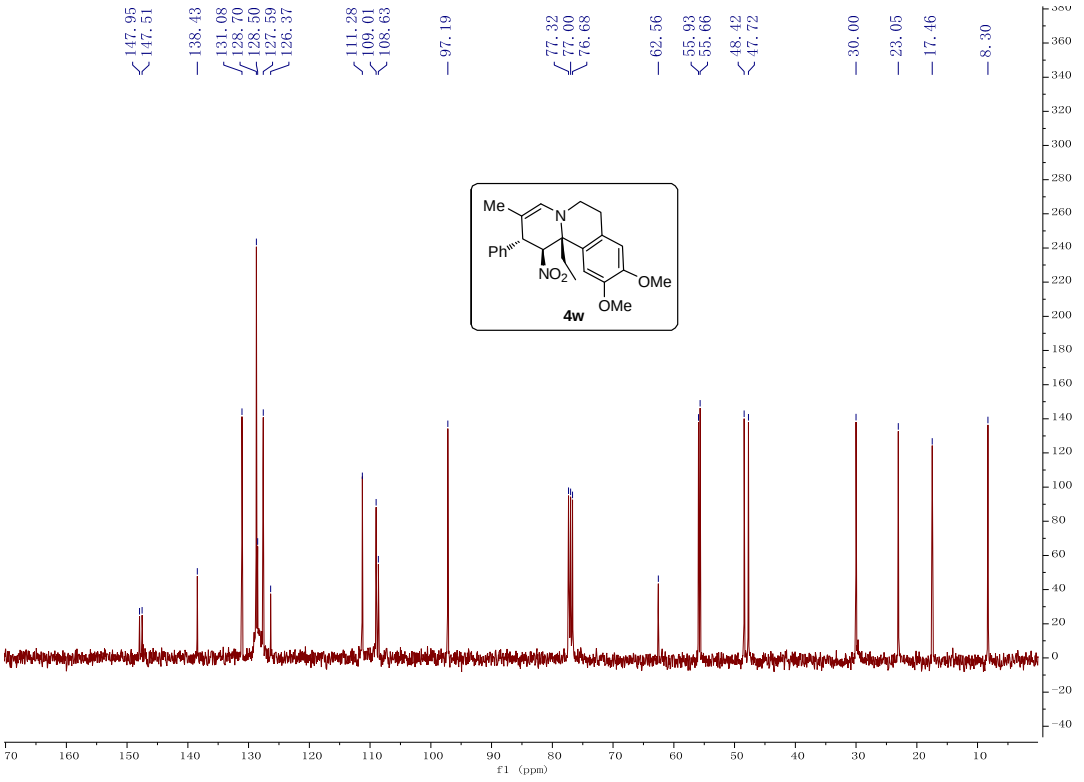
3.05
f1 (ppm)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76.68
62.56
55.93
55.66
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23.05
17.46
8.30
Me
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Ph
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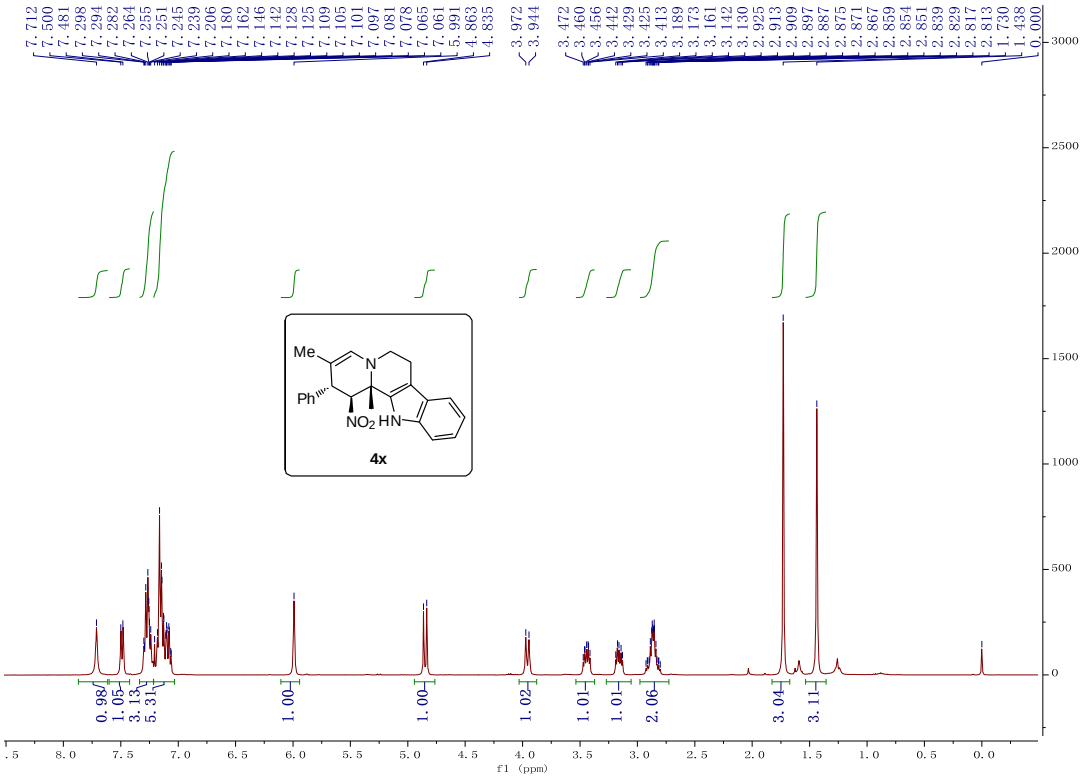
OMe
4w
f1 (ppm)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Ph
NO2 HN
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1.00
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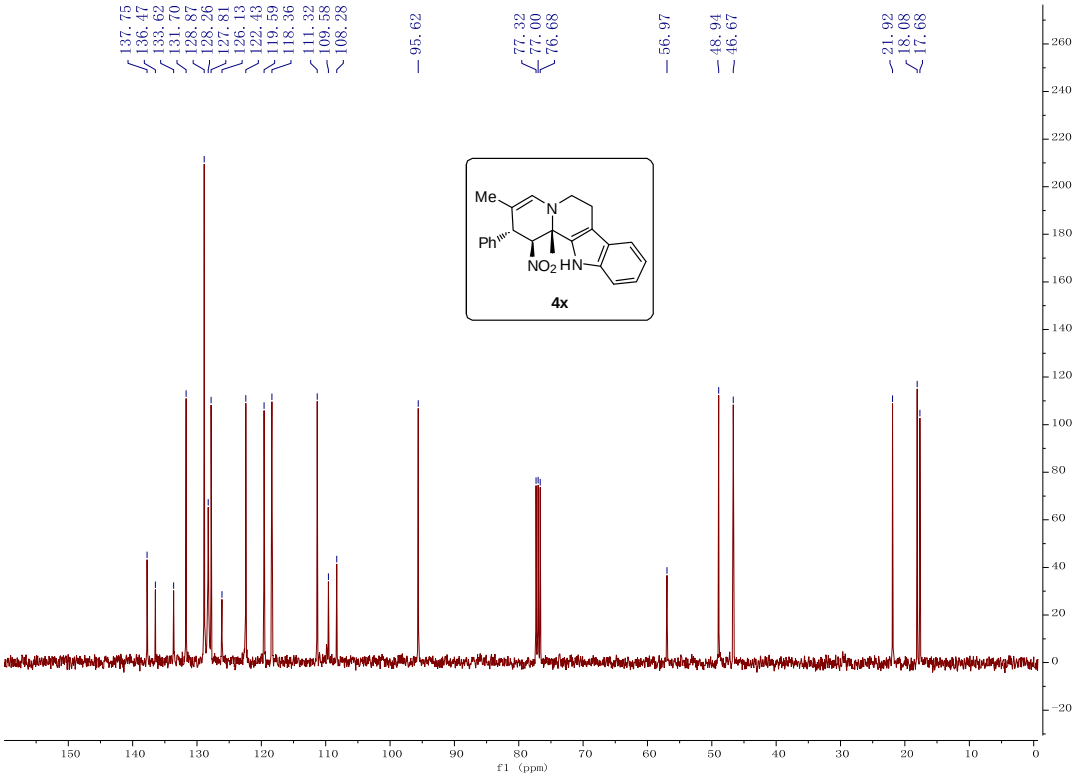
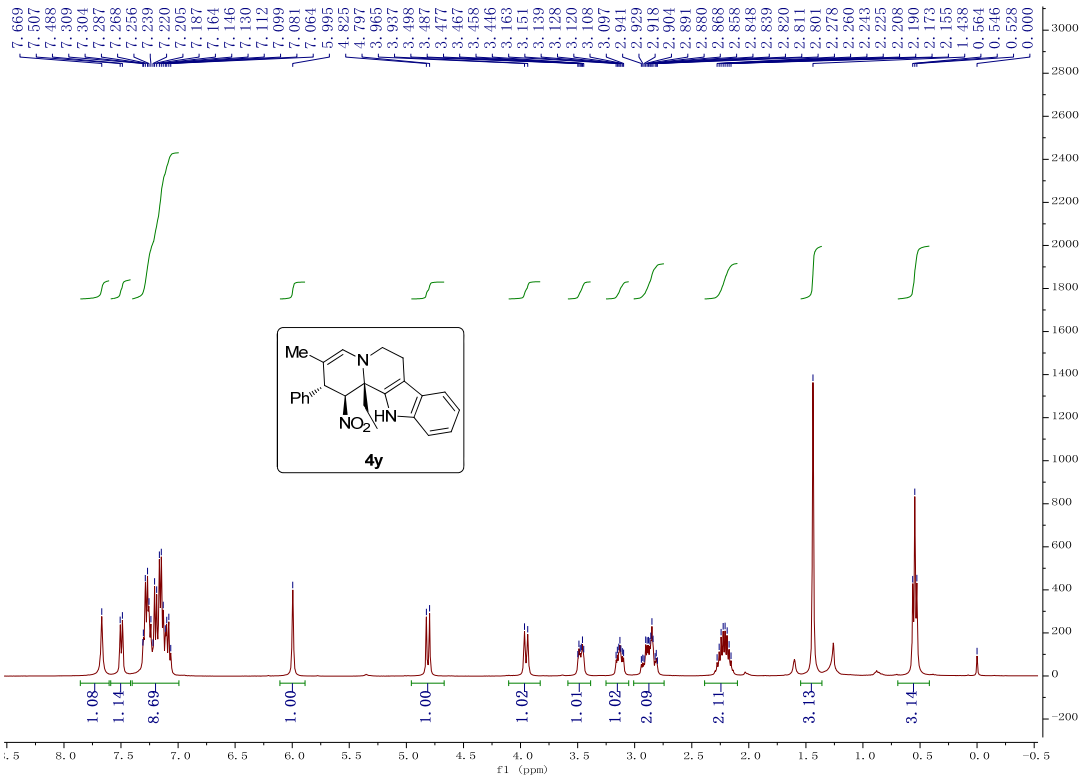
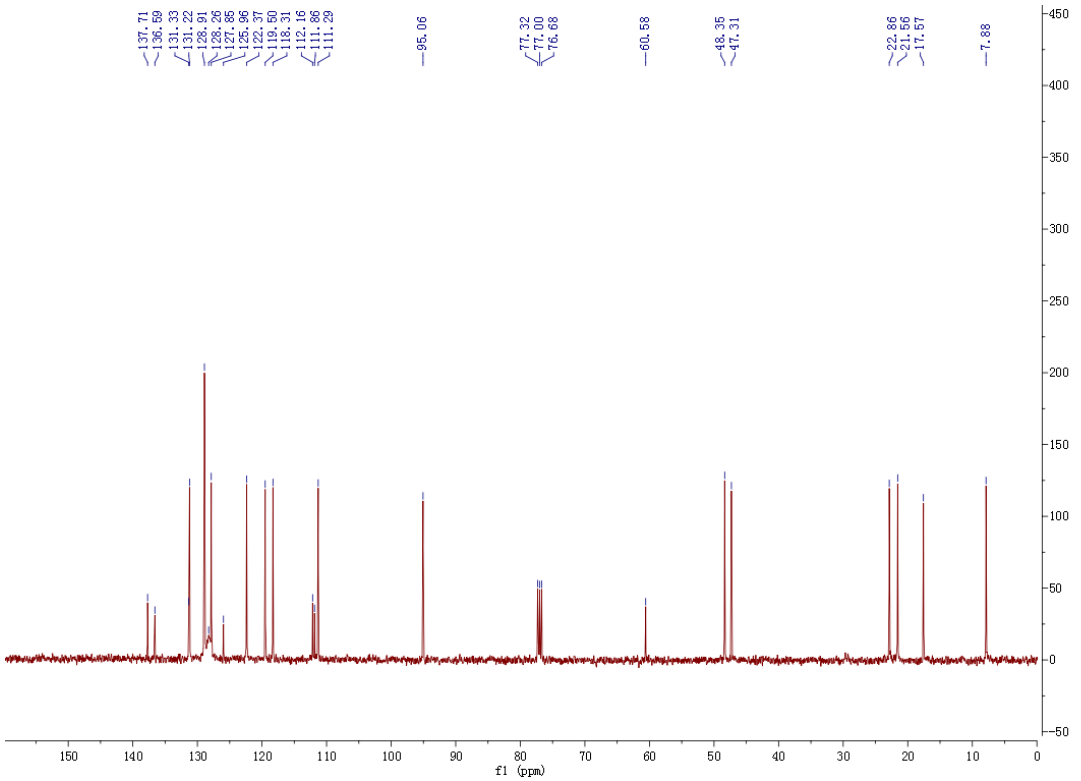
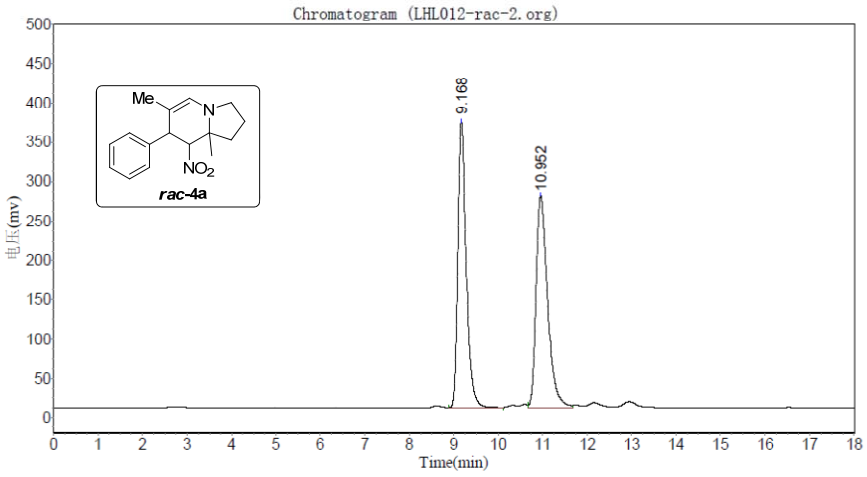
1.01
1.01
2.06
3.04
3.11
f1 (ppm)


137.75
136.47
133.62
131.70
128.87
128.26
127.81
126.13
122.43
119.59
118.36
111.32
109.58
108.28
95.62
77.32
77.00
76.68
56.97
48.94
46.67
21.92
18.08
17.68
Me
N
Ph
NO2 HN
4x
f1 (ppm)



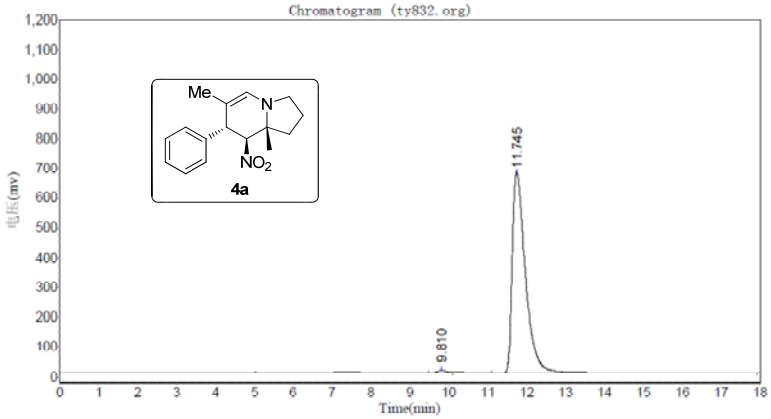


6. Chiral HPLC Traces



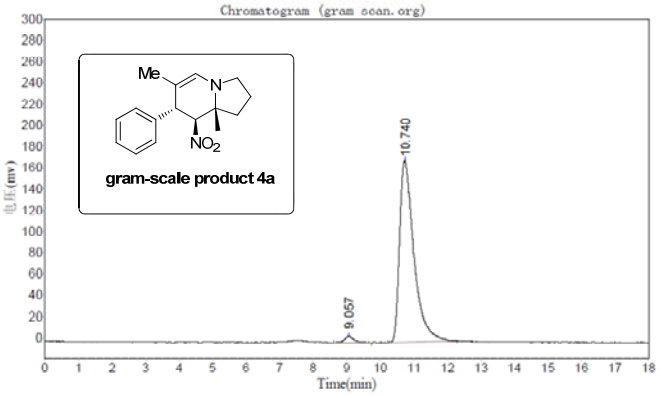
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		9.168	362405.594	4804997.000	49.8824
2		10.952	269225.219	4827648.000	50.1176
Total			631630.813	9632645.000	100.0000



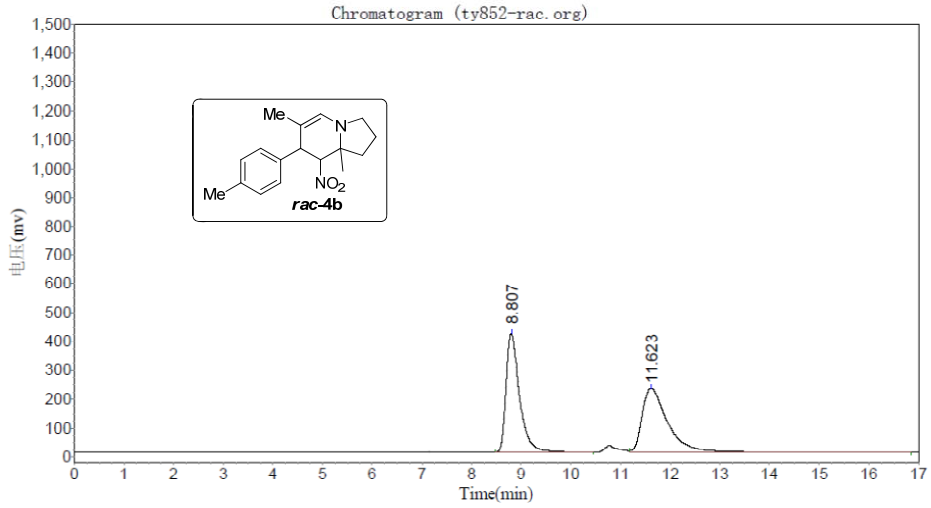
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		9.810	9633.085	145463.625	0.9087
2		11.745	675050.500	15862092.000	99.0913
Total			684683.585	16007555.625	100.0000



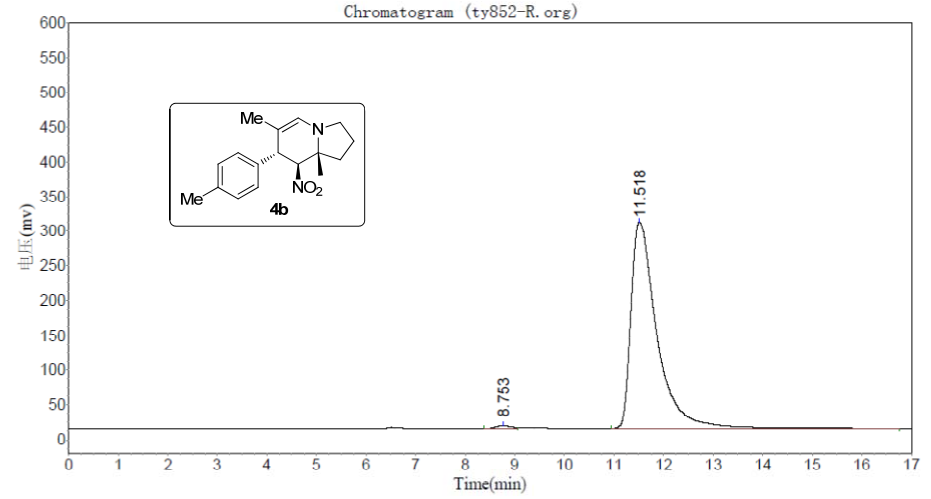
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		9.057	5898.614	112878.789	2.1445
2		10.740	171482.578	5150769.000	97.8555
Total			177381.192	5263647.789	100.0000



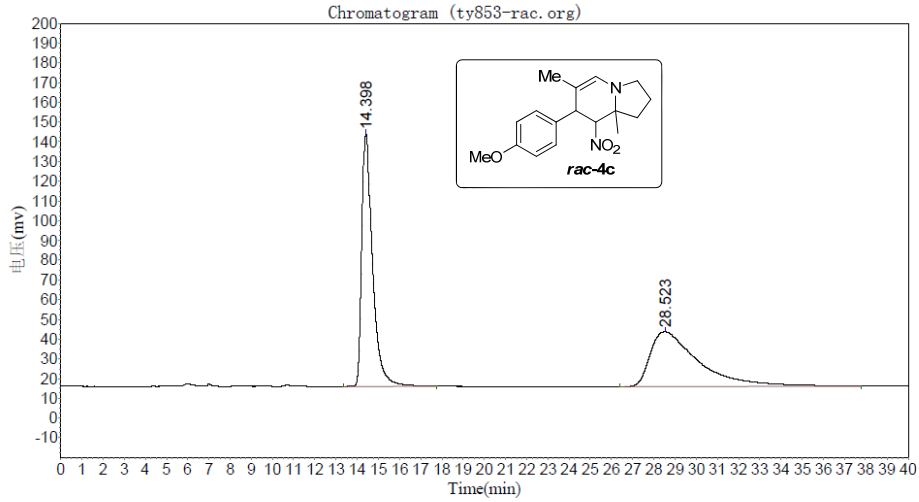
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		8.807	412721.719	7903534.500	49.2157
2		11.623	222501.828	8155442.500	50.7843
Total			635223.547	16058977.000	100.0000



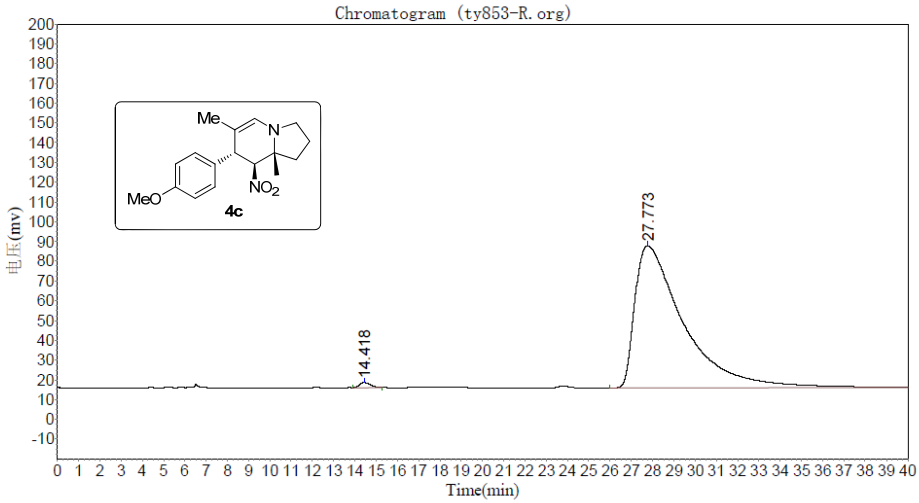
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		8.753	4083.870	81580.203	0.7512
2		11.518	297101.406	10778230.000	99.2488
Total			301185.276	10859810.203	100.0000



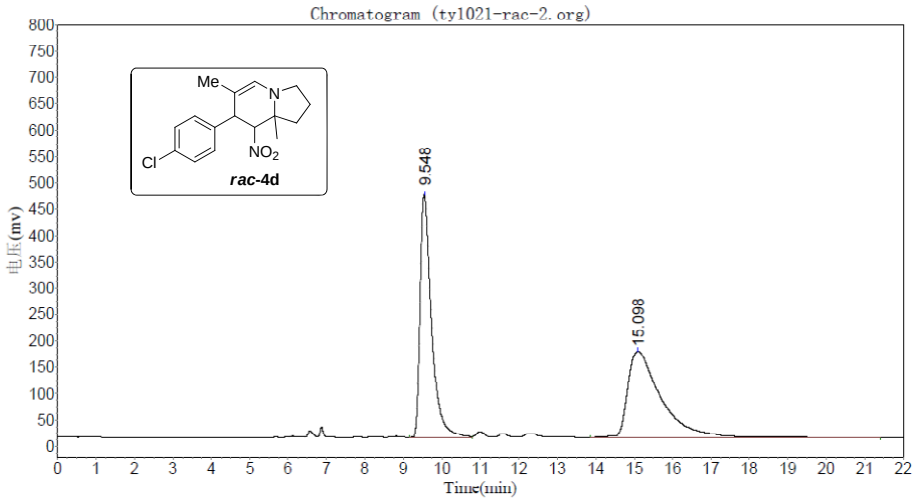
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		14.398	128056.875	4542154.000	50.3568
2		28.523	27799.648	4477794.000	49.6432
Total			155856.523	9019948.000	100.0000



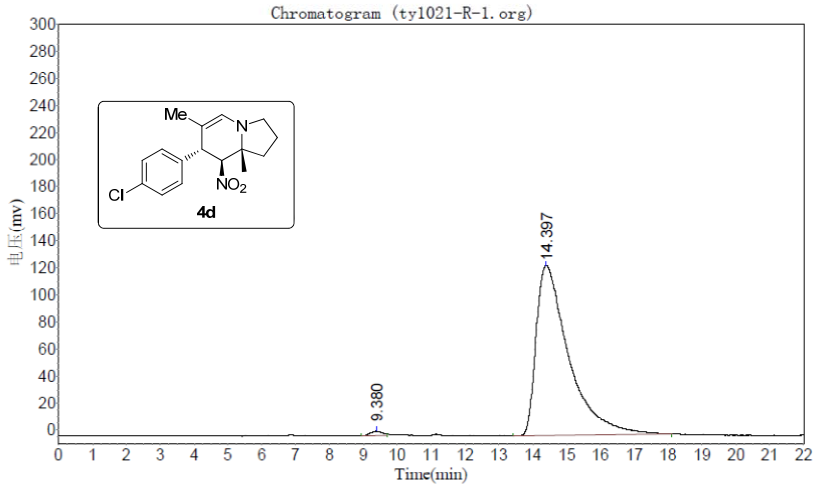
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		14.418	2711.739	89229.516	0.7704
2		27.773	72004.242	11492792.000	99.2296
Total			74715.981	11582021.516	100.0000



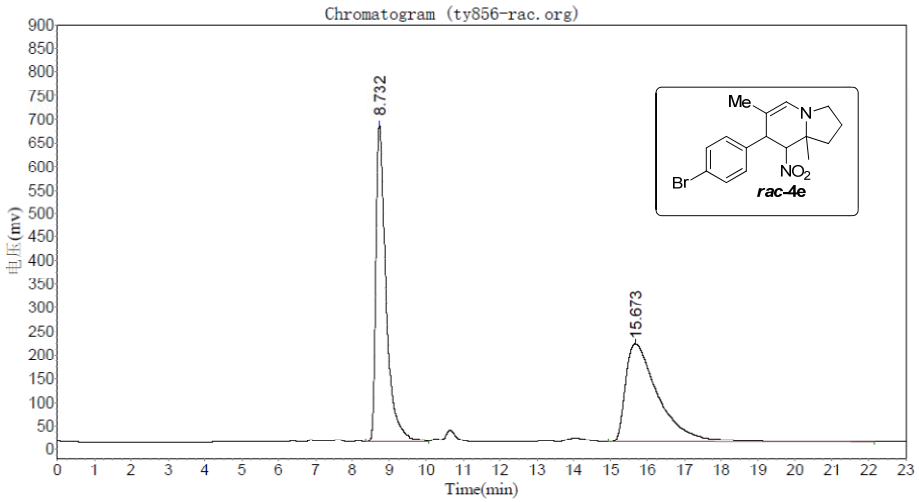
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		9.548	462397.406	9636925.000	49.1519
2		15.098	163623.203	9969470.000	50.8480
Total			626020.609	19606395.000	100.0000



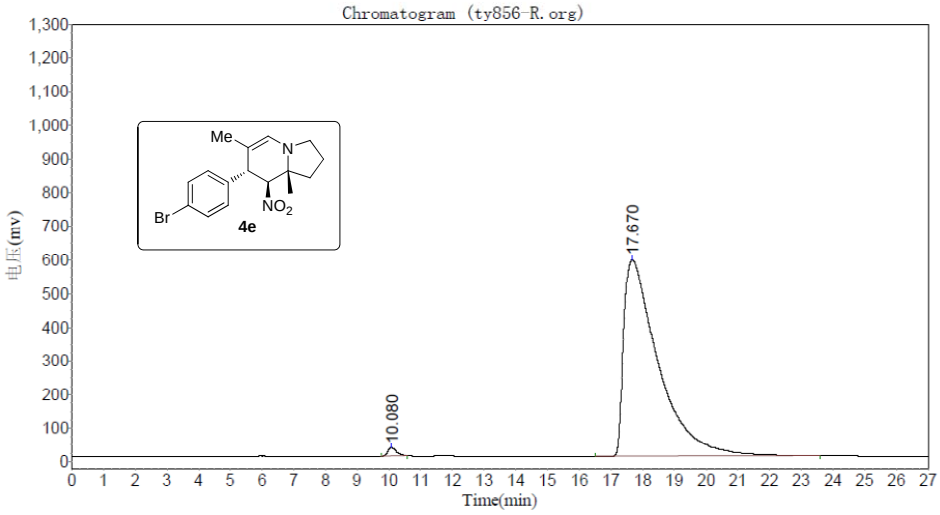
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		9.380	3316.521	79112.375	0.9297
2		14.397	125758.695	8430454.000	99.0703
Total			129075.217	8509566.375	100.0000



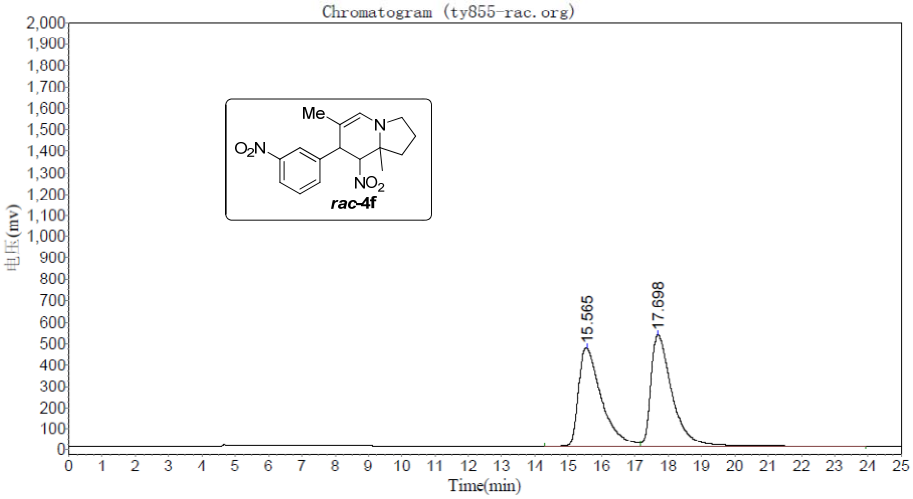
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		8.732	672309.688	13020911.000	49.9790
2		15.673	208022.719	13031859.000	50.0210
Total			880332.406	26052770.000	100.0000



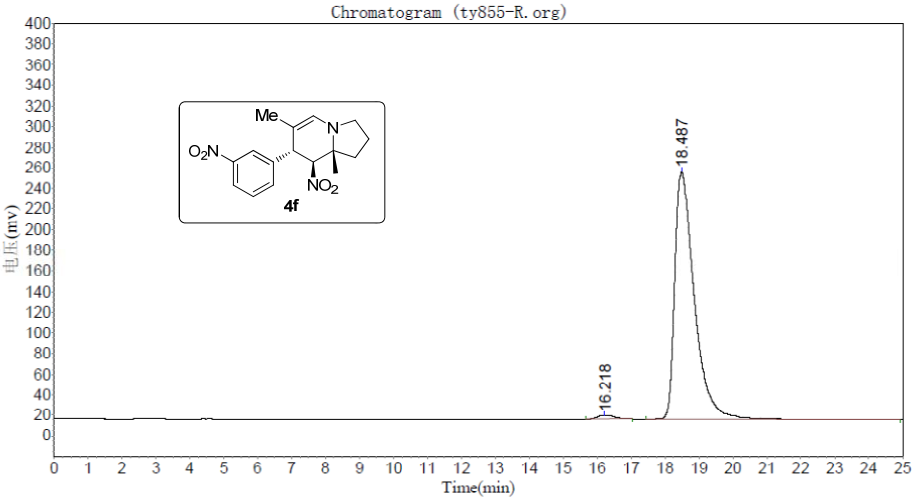
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		10.080	23536.977	456431.438	1.0169
2		17.670	584166.188	44430008.000	98.9831
Total			607703.164	44886439.438	100.0000



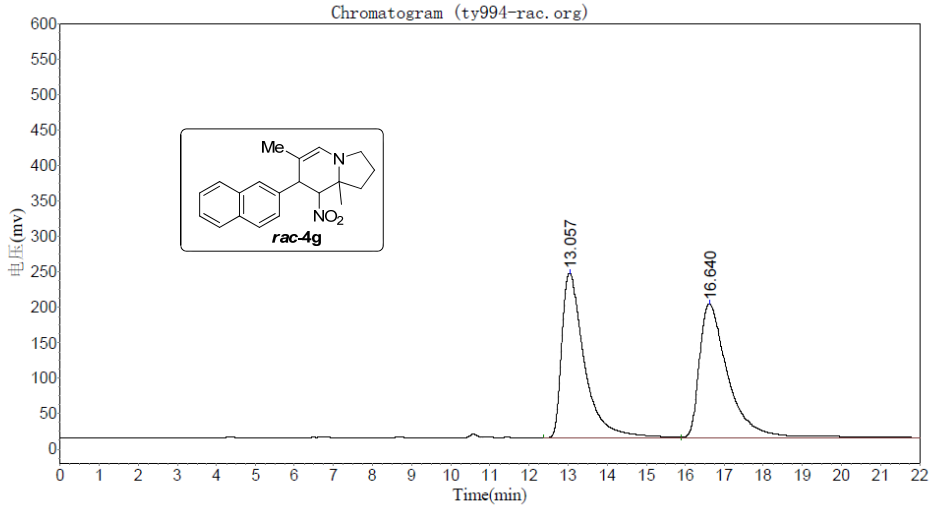
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		15.565	463370.438	22239074.000	48.7063
2		17.698	522724.094	23420450.000	51.2937
Total			986094.531	45659524.000	100.0000



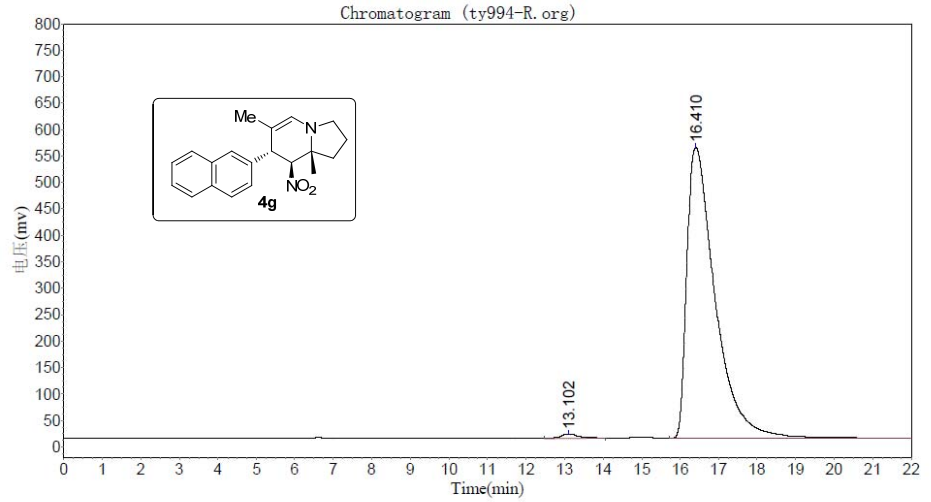
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		16.218	3491.727	129012.320	1.3235
2		18.487	240927.172	9618443.000	98.6765
Total			244418.899	9747455.320	100.0000



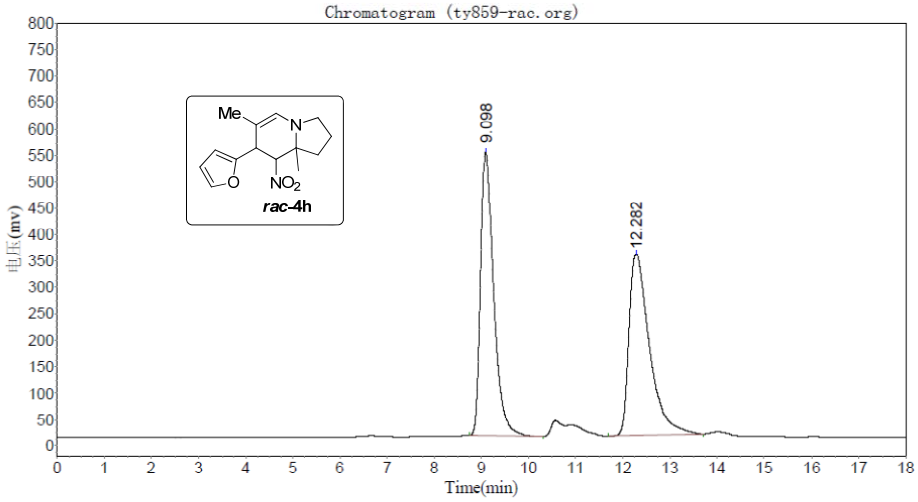
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		13.057	232158.625	9370117.000	49.5112
2		16.640	188826.344	9555133.000	50.4888
Total			420984.969	18925250.000	100.0000



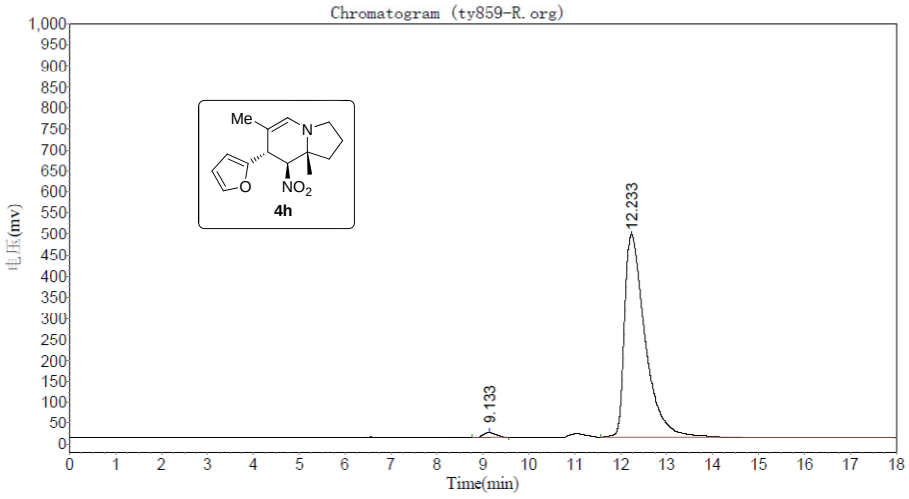
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		13.102	7527.441	271595.969	0.9546
2		16.410	550345.938	28178478.000	99.0454
Total			557873.378	28450073.969	100.0000



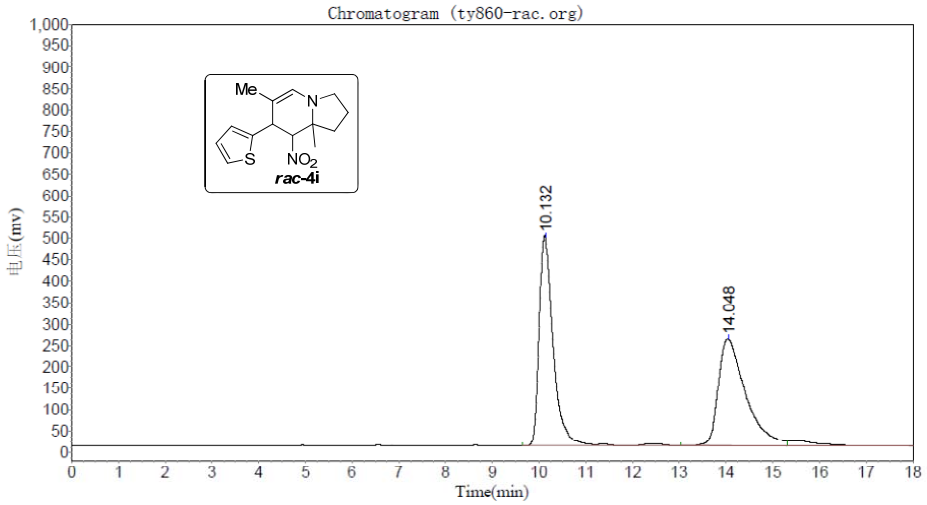
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		9.098	541810.875	10677284.000	49.4792
2		12.282	346727.969	10902055.000	50.5208
Total			888538.844	21579339.000	100.0000



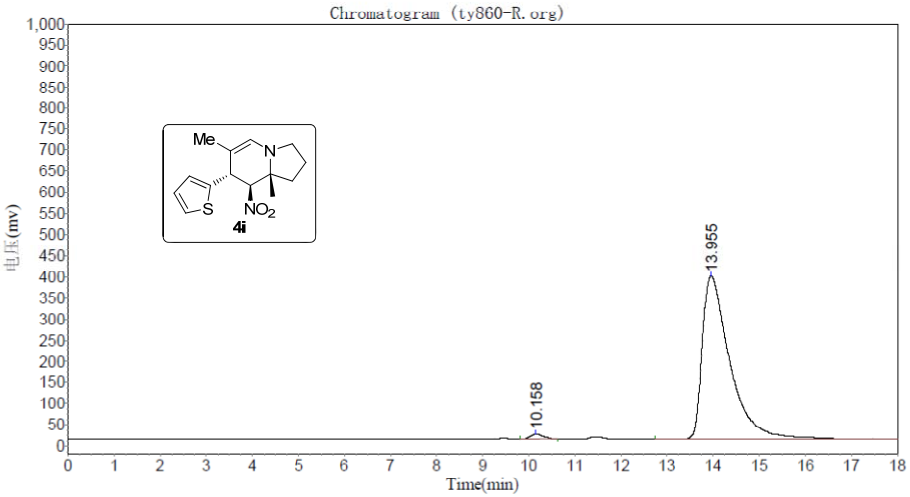
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		9.133	10362.652	185691.094	1.2312
2		12.233	479892.156	14896183.000	98.7688
Total			490254.809	15081874.094	100.0000



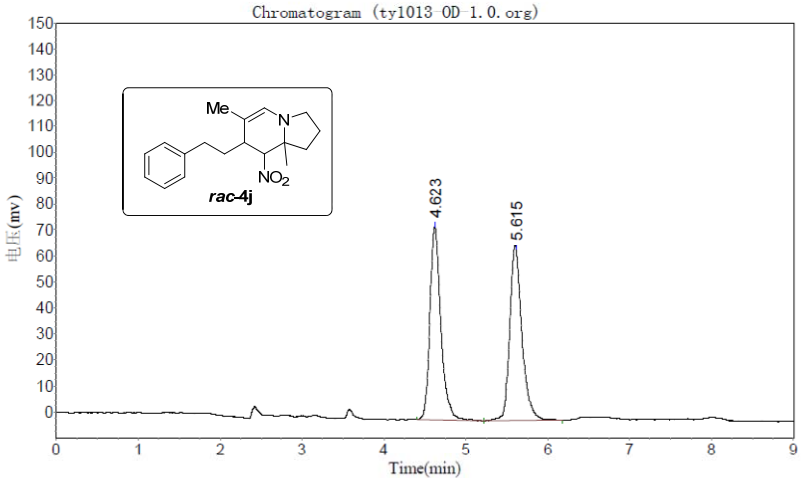
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		10.132	487952.156	10367446.000	50.8191
2		14.048	247747.375	10033235.000	49.1809
Total			735699.531	20400681.000	100.0000



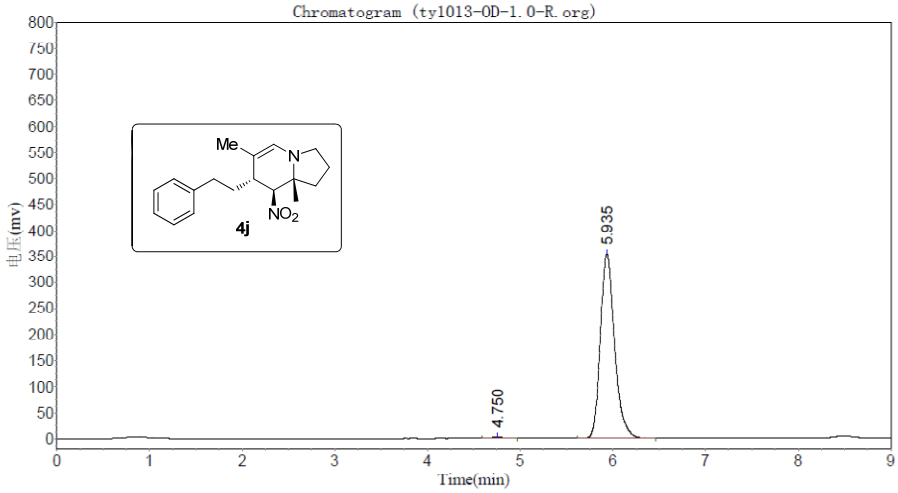
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		10.158	10585.622	196769.141	1.2097
2		13.955	385482.781	16069009.000	98.7903
Total			396068.403	16265778.141	100.0000



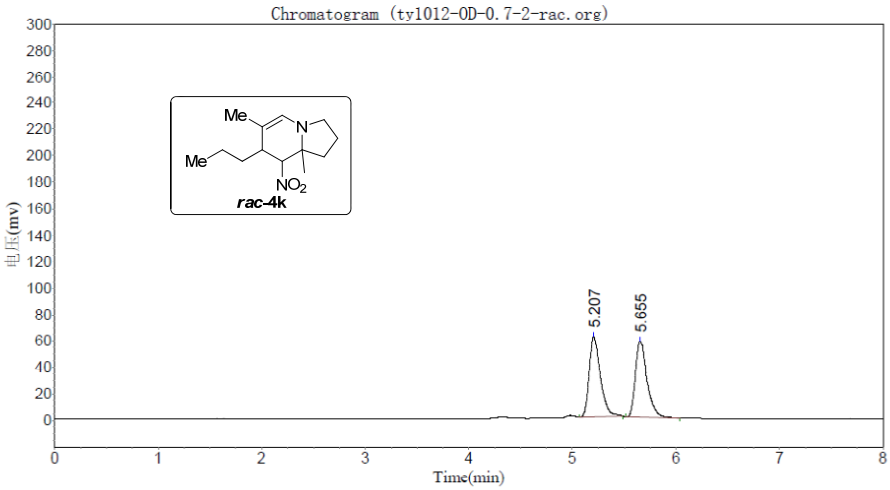
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		4.623	74431.141	692382.188	49.7485
2		5.615	66769.891	699381.625	50.2515
Total			141201.031	1391763.813	100.0000



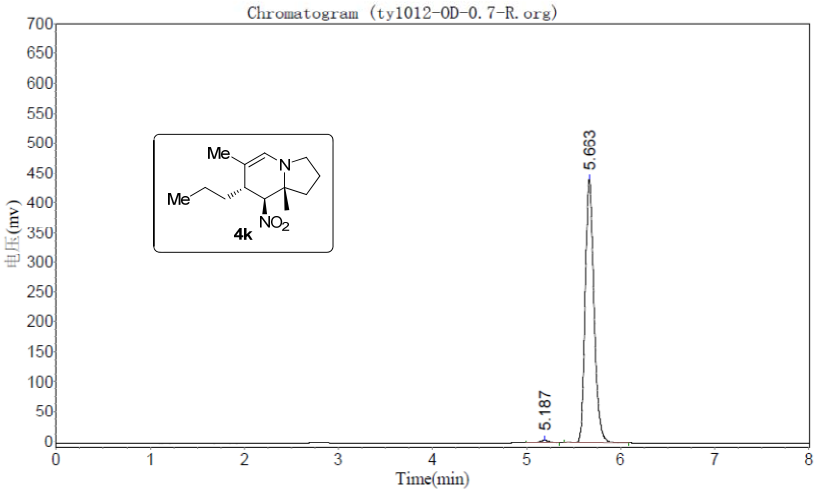
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		4.750	1881.145	21934.199	0.5825
2		5.935	353550.125	3743694.750	99.4175
Total			355431.270	3765628.949	100.0000



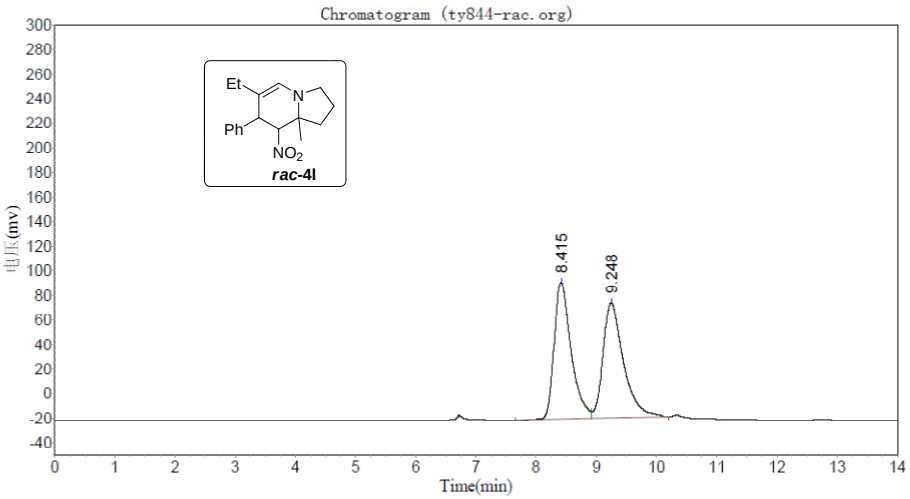
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		5.207	60431.836	443779.031	50.1151
2		5.655	57343.328	441740.688	49.8849
Total			117775.164	885519.719	100.0000



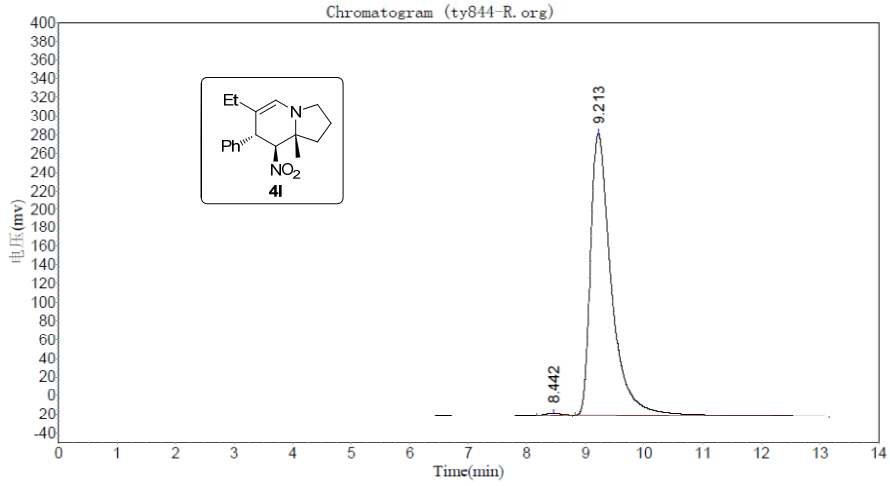
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		5.187	4999.486	34866.199	1.2098
2		5.663	441289.094	2847124.250	98.7902
Total			446288.580	2881990.449	100.0000



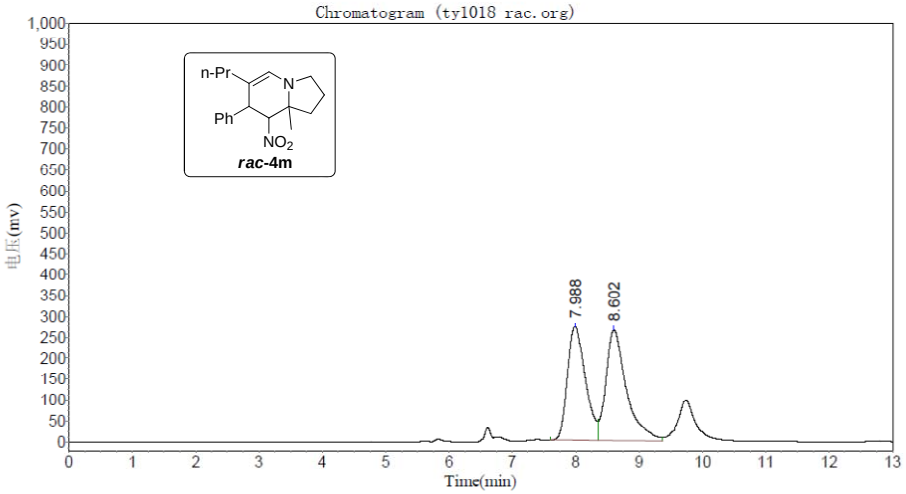
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		8.415	112333.125	2247537.750	48.5266
2		9.248	95821.875	2384016.250	51.4734
Total			208155.000	4631554.000	100.0000



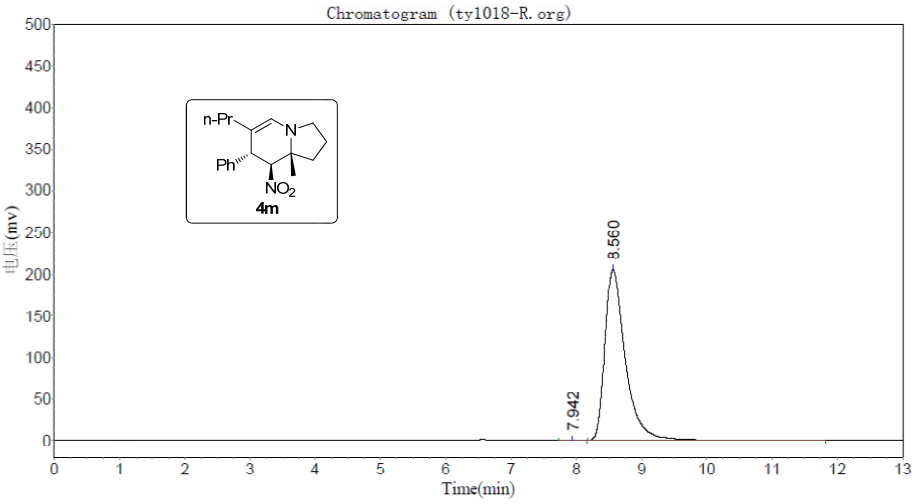
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		8.442	2185.354	40477.480	0.5411
2		9.213	302570.594	7440738.000	99.4589
Total			304755.948	7481215.480	100.0000



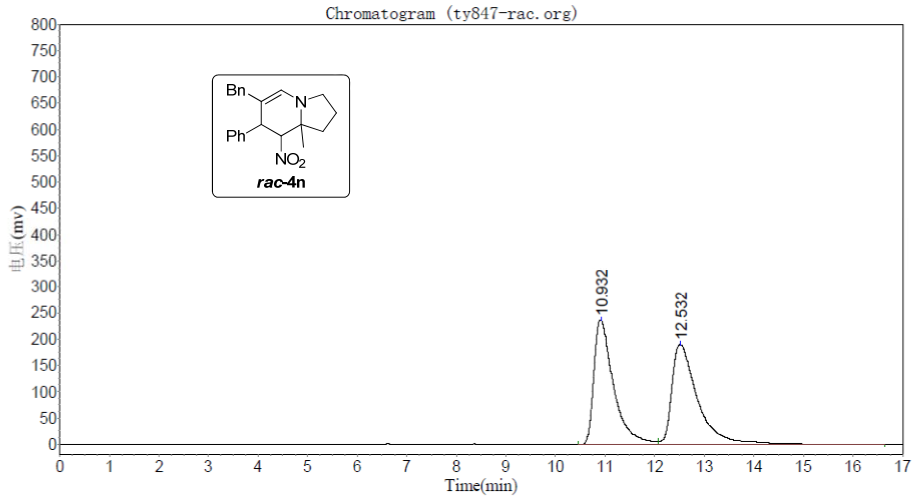
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		7.988	269632.531	5342756.500	47.3793
2		8.602	263170.781	5933805.000	52.6207
Total			532803.313	11276561.500	100.0000



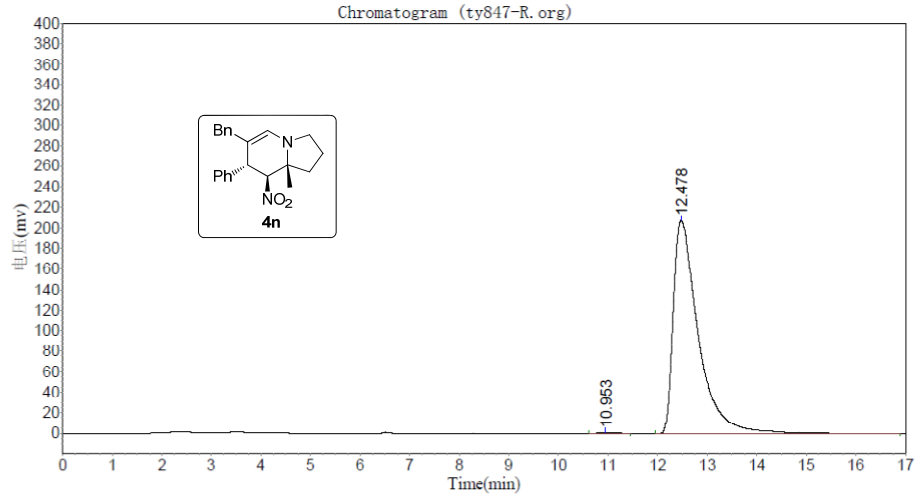
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		7.942	460.202	6498.000	0.1440
2		8.560	205533.047	4505688.000	99.8560
Total			205993.248	4512186.000	100.0000



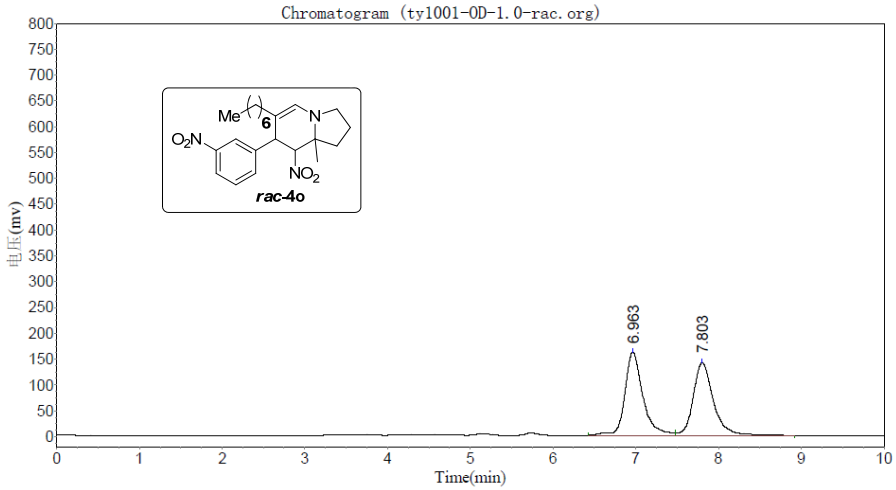
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		10.932	236126.359	6500001.500	48.6828
2		12.532	190201.172	6851737.500	51.3172
Total			426327.531	13351739.000	100.0000



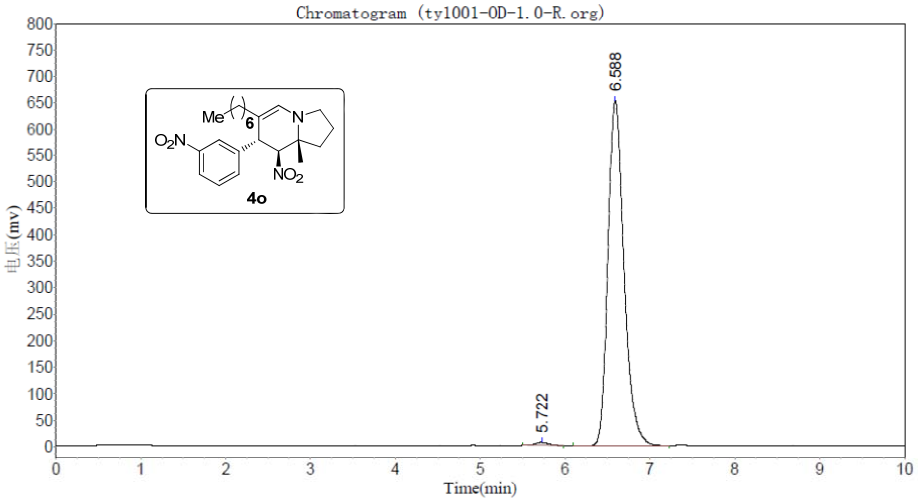
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		10.953	1288.208	29612.750	0.4069
2		12.478	207209.094	7247583.000	99.5931
Total			208497.302	7277195.750	100.0000



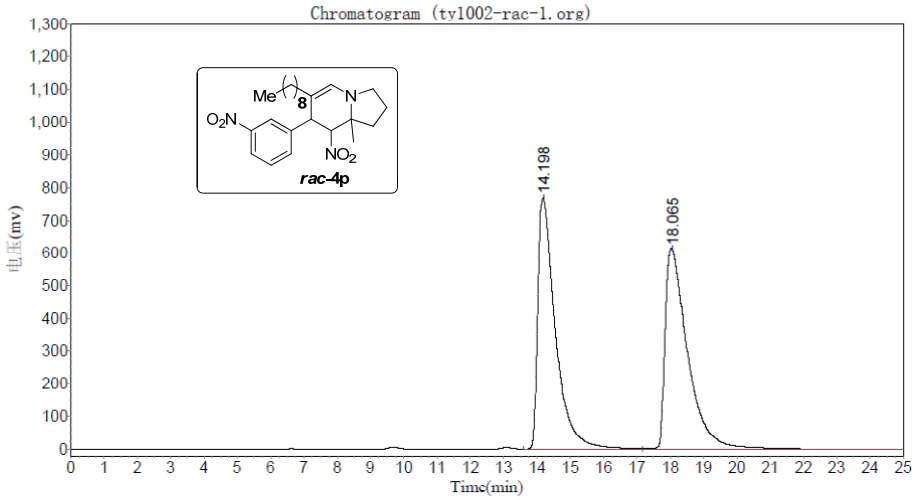
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		6.963	160852.578	2434481.750	50.5679
2		7.803	140751.594	2379800.250	49.4321
Total			301604.172	4814282.000	100.0000



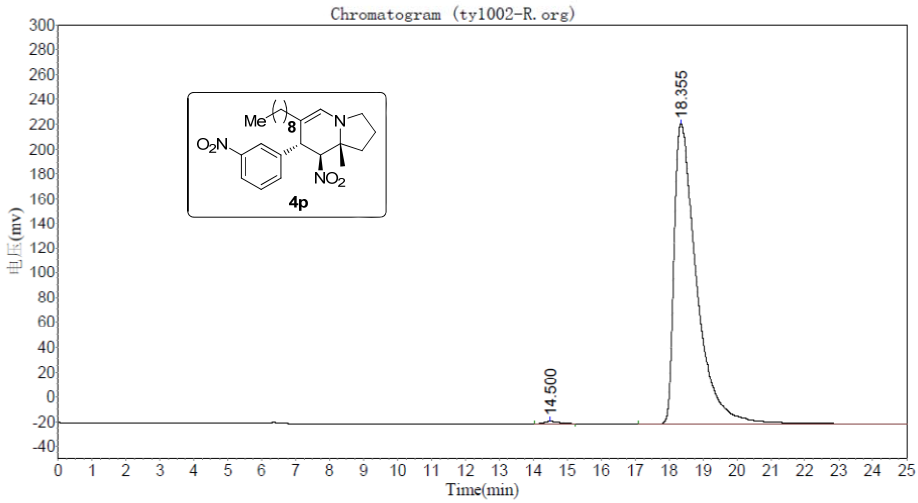
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		5.722	5828.419	62360.699	0.7287
2		6.588	652943.875	8495369.000	99.2713
Total			658772.294	8557729.699	100.0000



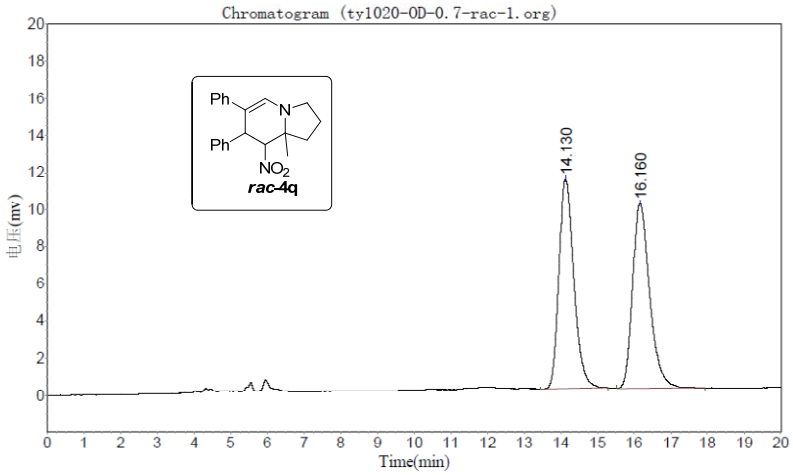
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		14.198	768563.813	28420868.000	49.5939
2		18.065	613087.000	28886258.000	50.4060
Total			1381650.813	57307126.000	100.0000



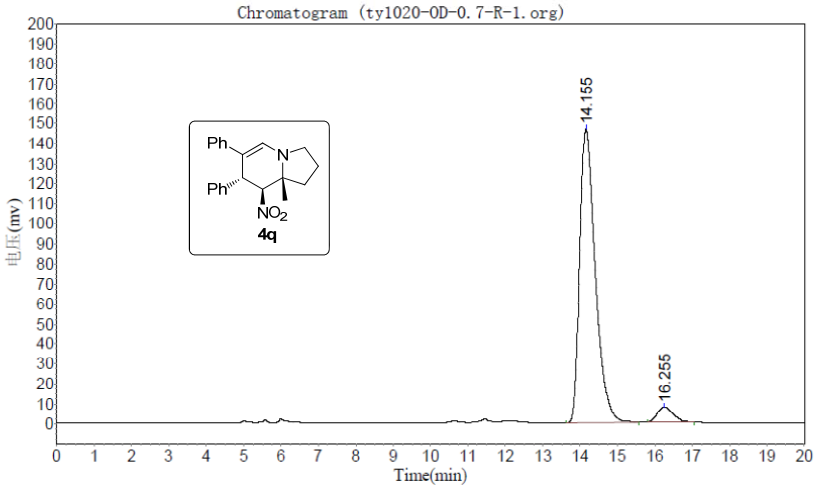
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		14.500	1931.230	60003.688	0.5365
2		18.355	242455.813	11124738.000	99.4635
Total			244387.043	11184741.688	100.0000



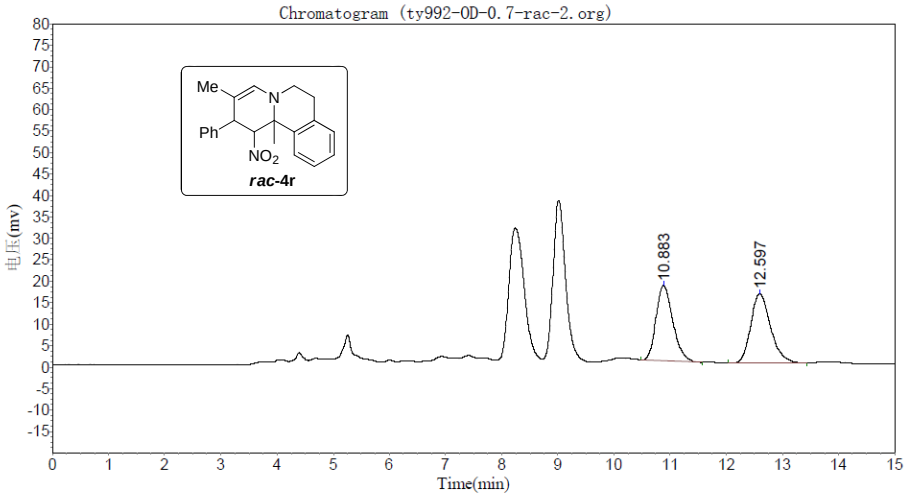
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		14.130	11284.771	316931.594	49.7254
2		16.160	9935.960	320431.406	50.2746
Total			21220.730	637363.000	100.0000



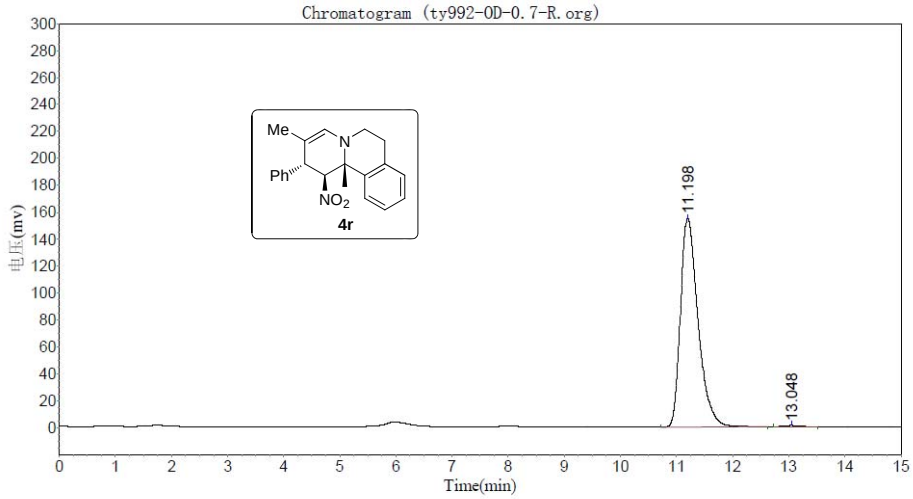
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		14.155	146630.109	4250598.500	94.8699
2		16.255	7656.704	229851.641	5.1301
Total			154286.813	4480450.141	100.0000



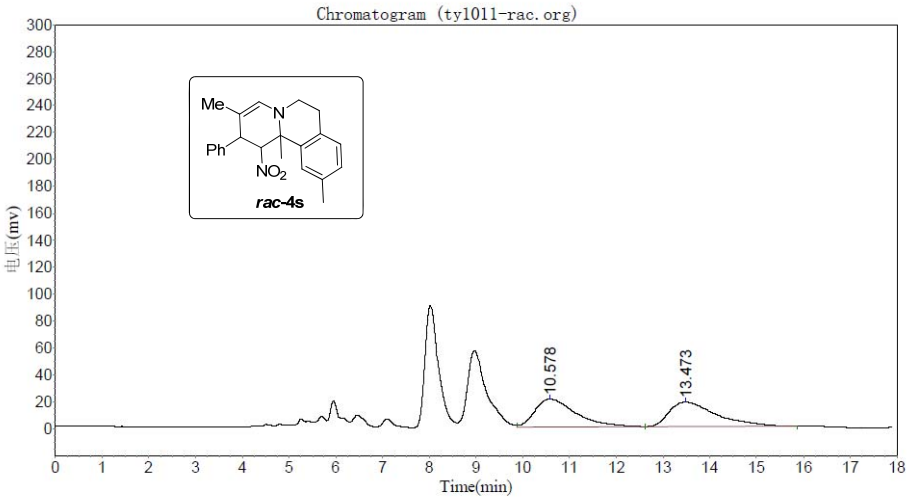
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		10.883	17555.656	366038.906	48.1662
2		12.597	16178.455	393910.125	51.8338
Total			33734.111	759949.031	100.0000



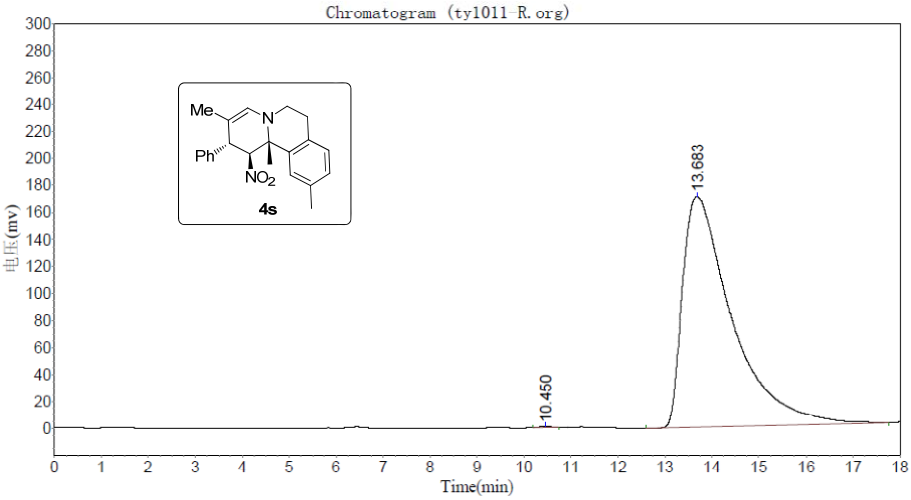
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		11.198	155041.031	3373907.500	99.3211
2		13.048	1028.454	23061.301	0.6789
Total			156069.485	3396968.801	100.0000



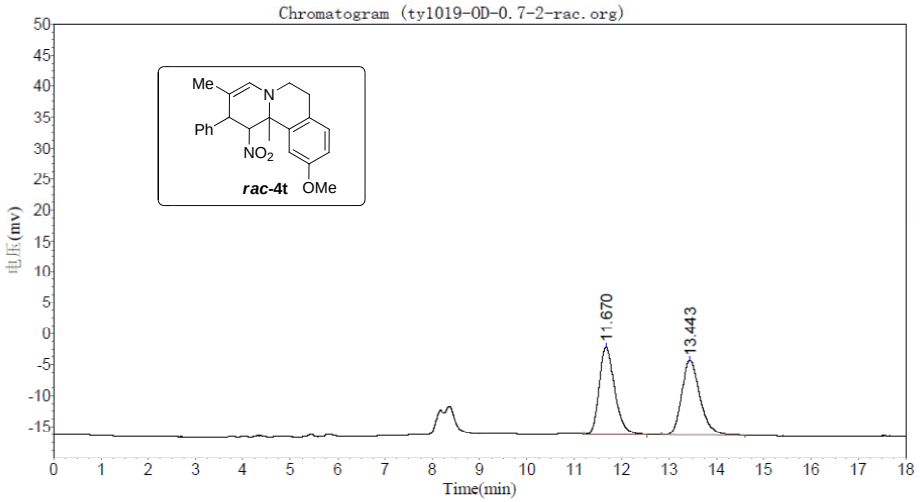
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		10.578	21037.031	1305122.875	50.0156
2		13.473	18318.514	1304309.250	49.9844
Total			39355.545	2609432.125	100.0000



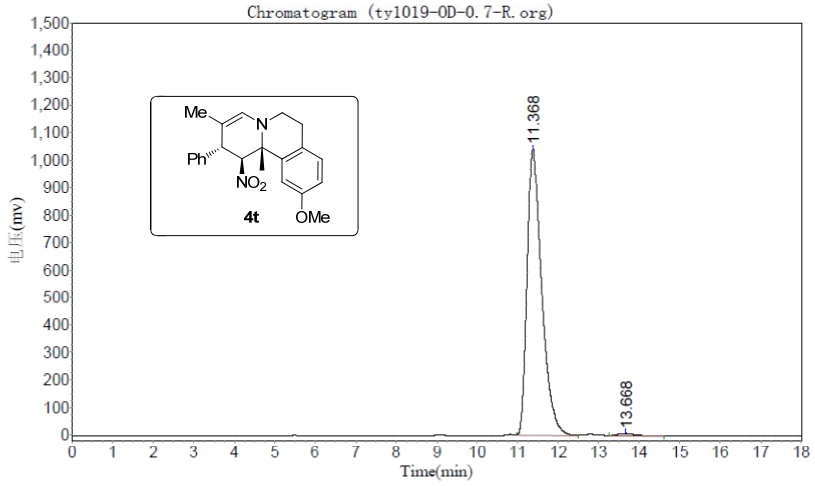
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		10.450	498.554	8938.400	0.0714
2		13.683	170905.172	12516122.000	99.9286
Total			171403.726	12525060.400	100.0000



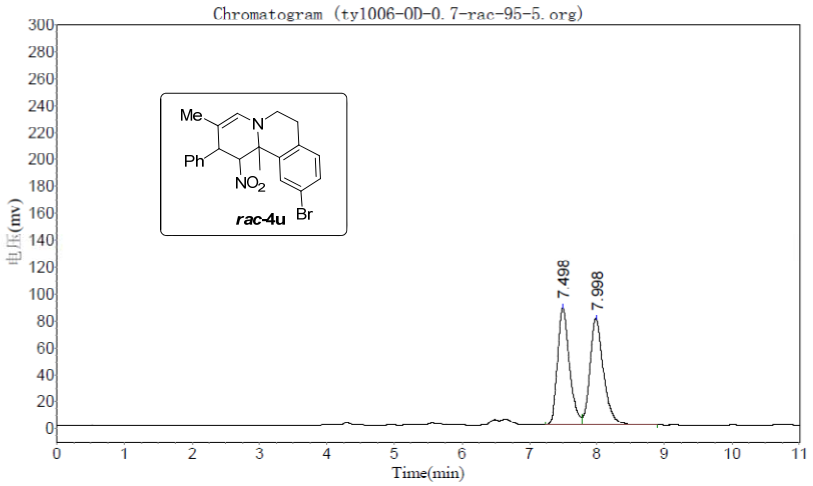
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		11.670	14137.702	317584.375	49.6751
2		13.443	11984.763	321739.219	50.3249
Total			26122.465	639323.594	100.0000



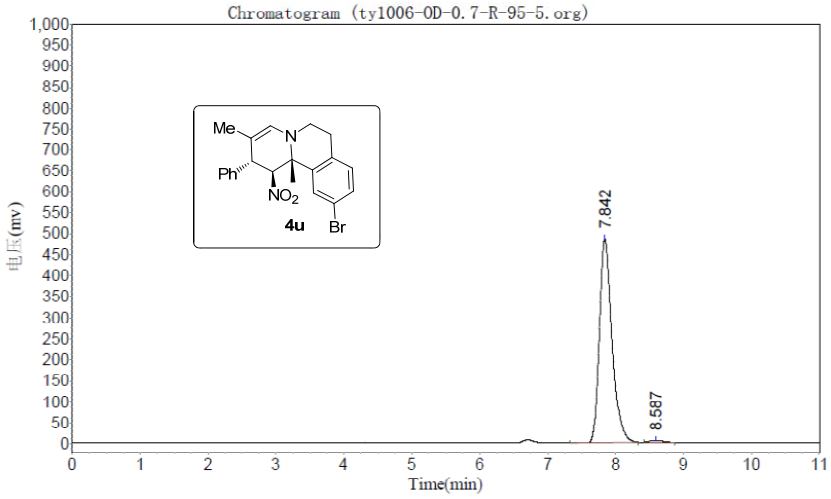
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		11.368	1041658.188	25976492.000	99.1135
2		13.668	8309.133	232346.031	0.8865
Total			1049967.320	26208838.031	100.0000



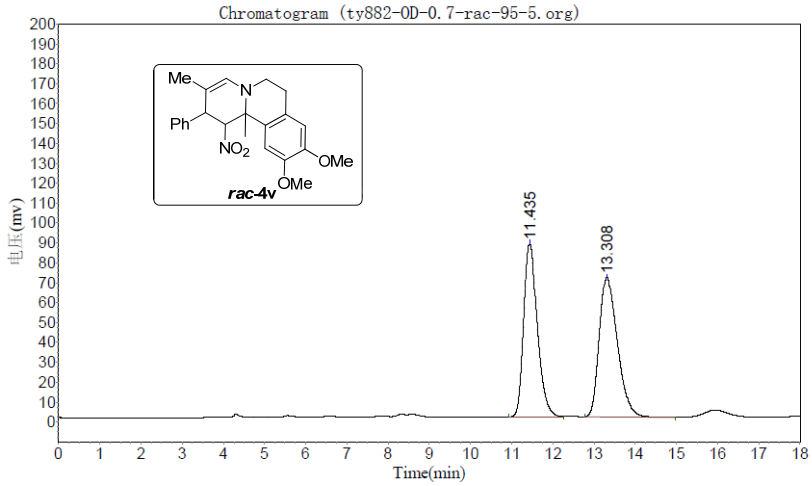
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		7.498	87444.945	1087278.750	49.1218
2		7.998	79271.406	1126154.250	50.8782
Total			166716.352	2213433.000	100.0000



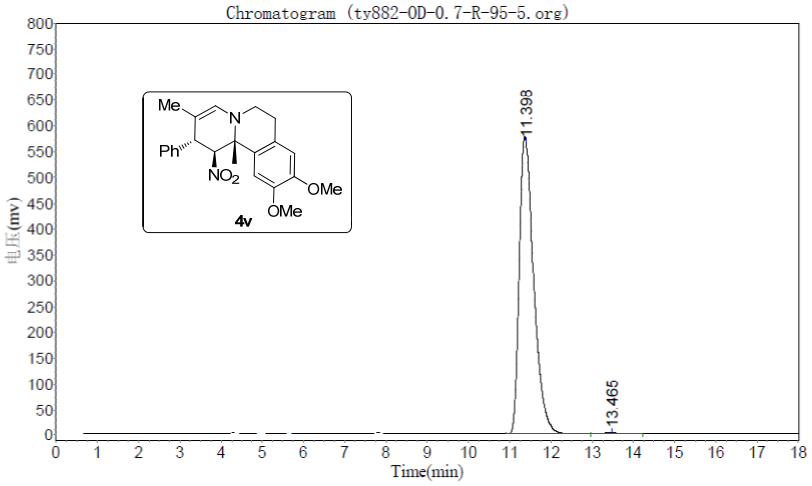
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		7.842	484896.188	6264710.000	98.9062
2		8.587	4766.614	69284.258	1.0938
Total			489662.801	6333994.258	100.0000



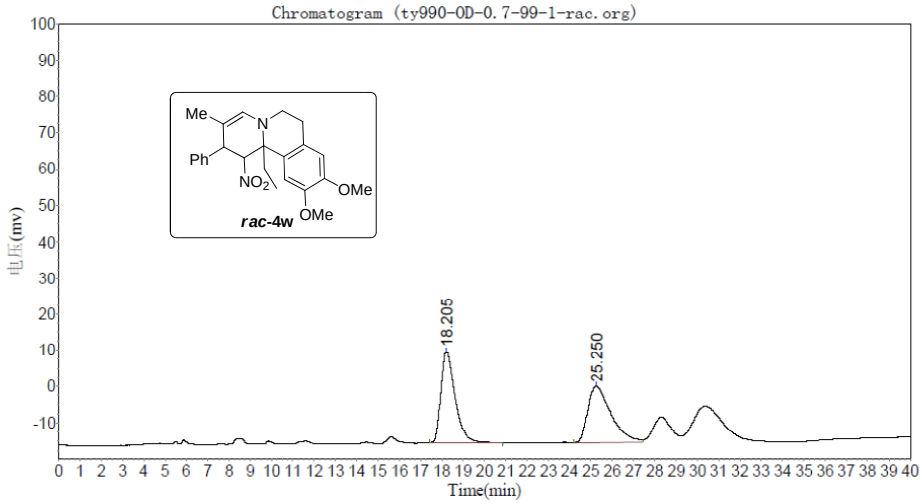
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		11.435	86900.891	2007935.375	48.1006
2		13.308	70100.750	2166513.500	51.8994
Total			157001.641	4174448.875	100.0000



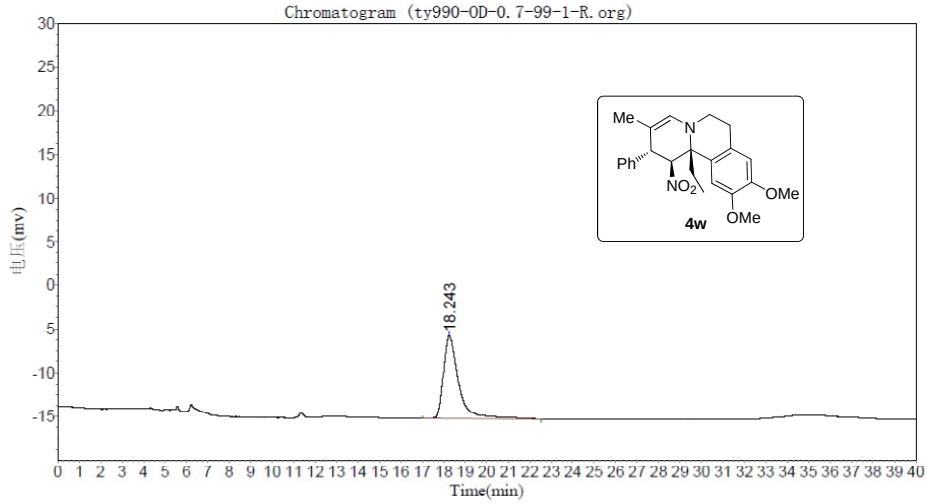
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		11.398	574262.875	14068463.000	98.9888
2		13.465	4519.881	143718.297	1.0112
Total			578782.756	14212181.297	100.0000



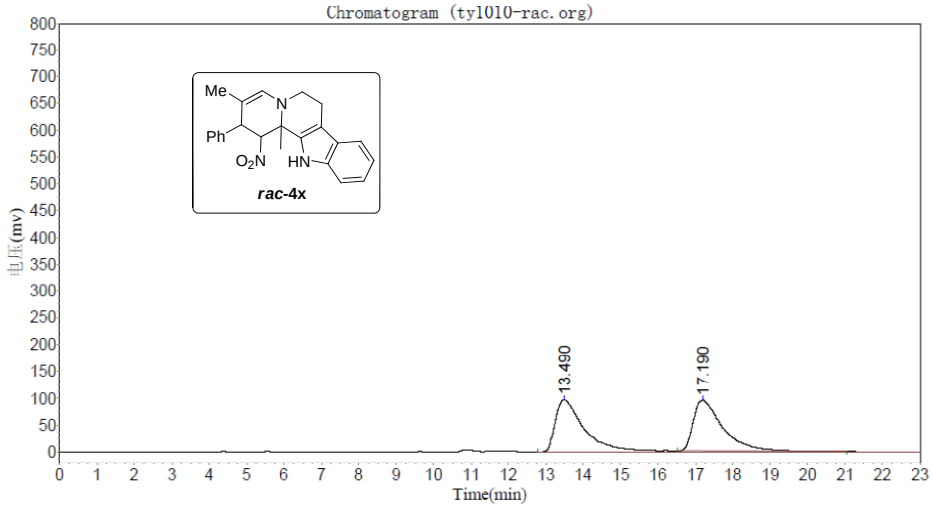
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		18.205	25046.809	1162490.875	49.8534
2		25.250	15440.978	1169328.875	50.1466
Total			40487.786	2331819.750	100.0000



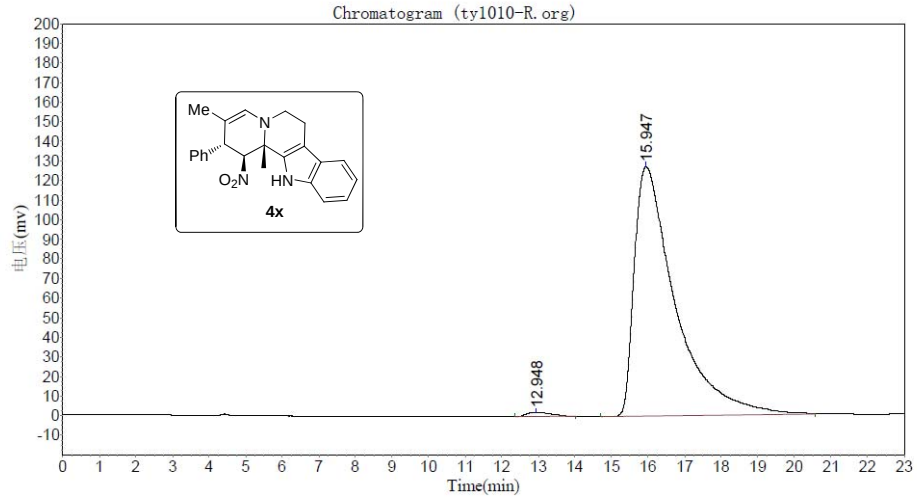
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		18.243	9372.722	469723.719	100.0000
Total			9372.722	469723.719	100.0000



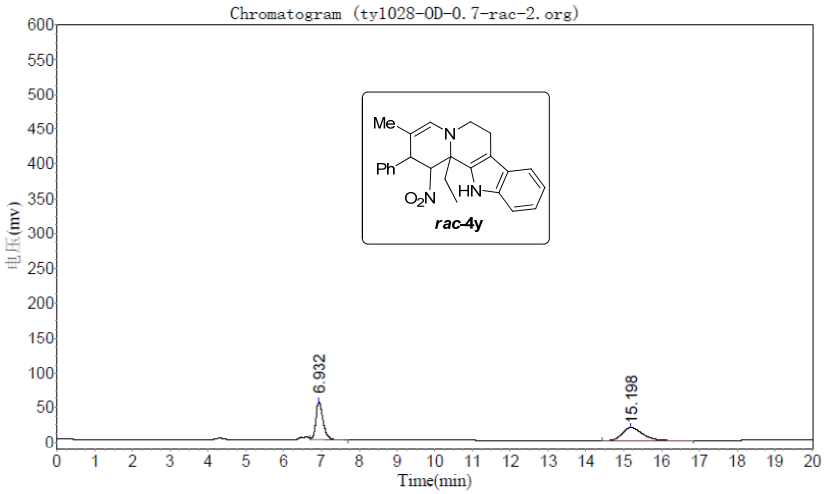
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		13.490	96966.898	5589868.500	51.9865
2		17.190	94429.500	5162672.500	48.0135
Total			191396.398	10752541.000	100.0000



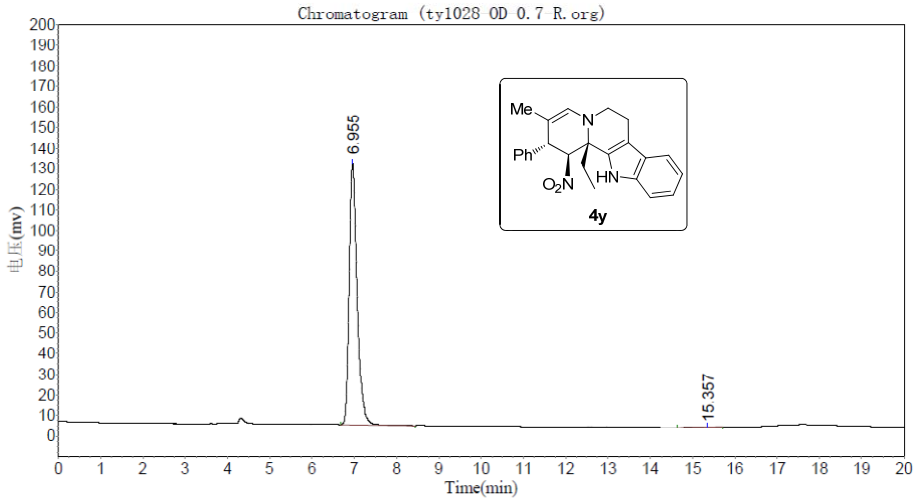
Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		12.948	1998.977	86626.305	0.8704
2		15.947	127278.969	9865458.000	99.1296
Total			129277.946	9952084.305	100.0000



Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		6.932	53499.801	710233.438	51.1194
2		15.198	17538.346	679129.563	48.8806
Total			71038.146	1389363.000	100.0000



Results

Peak No.	Peak ID	Ret Time	Height	Area	Conc.
1		6.955	127198.414	1668133.750	99.6046
2		15.357	230.039	6622.794	0.3954
Total			127428.453	1674756.544	100.0000

7. X-ray Crystallography of 4n

The single crystal of **4n** grown from a solution of petroleum: ethyl acetate = 5:1 was submitted to X-Ray crystallography