Angewandte Chemie

Eine Zeitschrift der Gesellschaft Deutscher Chemiker

Supporting Information

69451 Weinheim, Germany

Supporting Information

Multiple C-H Activations to Construct Biological Active Molecules in a Completely Organohalogen-free and Organometal-free Process

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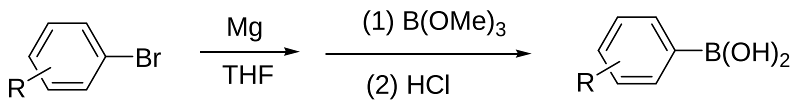
General. All the reactions were carried out under oxygen atmosphere. EtCOOH and arenes were purchased as analytical pure and used without further purification. $Pd(OAc)_2$ was purchased from Acros Chemicals and $Cu(OTf)_2$ was purchased from Aldrich. ^{1}H NMR (300 MHz) and ^{13}C NMR (75 MHz) were registered on Varian 300 M spectrometers with $CDCl_3$ as solvent and tetramethylsilane (TMS) as internal standard. Chemical shifts were reported in units (ppm) by assigning TMS resonance in the ^{1}H spectrum as 0.00 ppm and $CDCl_3$ resonance in the ^{13}C spectrum as 77.0 ppm. All coupling constants (*J* values) were reported in Hertz (Hz). Column chromatography was performed on silica gel 200-300 mesh. IR, X-ray, GC, MS, and HRMS were performed by the State-authorized Analytical Center in Peking University.

General procedures for cross-coupling of aceto-tetrahydroquinoline derivatives with arenes:

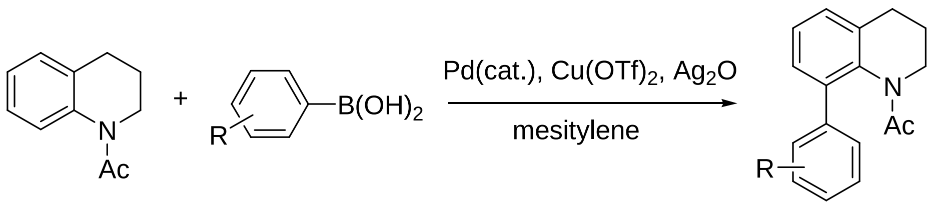
All the reactions were performed on 0.3 mmol scale. 6.7 mg $Pd(OAc)_2$ (0.03 mmol), indicated amount of $Cu(OTf)_2$ and 52.5 mg (0.3 mmol) *N*-acetyl-1, 2, 3, 4-tetrahydro-quinoline **1** were weighed in the air and added together into an oven-dried 25 mL Schlenck tube. The septumsealed tube was evacuated and refilled with O_2 three times. EtCOOH (1.5 mL) and arene (1 mL) were added and the mixture was stirred at 120 ℃ under 1 atm of oxygen (ballon pressure) until the substrate was completed consumed. After cooling down, the mixture was diluted with 80 mL CH_2Cl_2. The organic phase was washed with water (30 mL×2), saturated Na_2CO_3 (30 mL) and dried over $MgSO_4$. The solvent was removed and the residue was applied to flash column chromatography eluting with ethyl acetate/ petroleum ether.

Assignment of each isomers of the cross-coupling products:

The standard products of each isomer listed in Table 2 were prepared by coupling of *N*-acetyl-1, 2, 3, 4-tetrahydro- quinoline **1a** with the corresponding aryl boronic acids according to our previously reported method (equation 2).[1] The boronic acids were prepared according to literature procedure starting from the corresponding aryl bomide (equation 1).

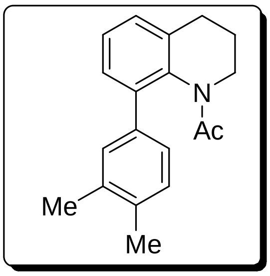


(1)

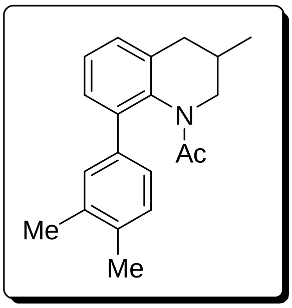


(2)

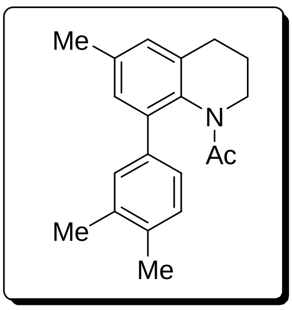
All the products listed in Table 2 were confirmed by comparing with these independently synthesized standard products. The ratio of isomers was based on GC-MS and GC analysis of crude reaction mixture. Assignment of the NMR data and GC peak of isomers were also based on these standard products. The products 3ad/3ad'-3ah/3ah' are quite similar in ^{1}H NMR. However, ^{13}C NMR data can be assigned.



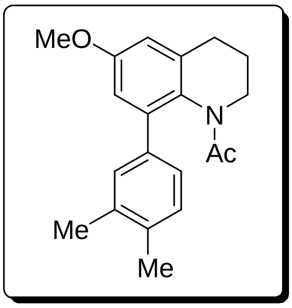
1-(8-(3,4-dimethylphenyl)-3,4-dihydroquinolin-1(2H)-yl)-ethanone (**3aa**). Follow the general procedures and 0.2 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL *o*-xylene and 1.5 mL EtCOOH, yielded 65.3 mg product in 3 h as a single isomer, 78%. 6 eq. *o*-xylene is sufficient enough to guarantee complete conversion in 7 h. *A 10 mmol scale reaction was also performed to demonstrate its preparative value. 1 equiv $Cu(OTf)_2$ was used and resulted in complete conversion in 3 h, 73%.* ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.30-7.05 (m, 6 H), 4.81-4.71 (m, 1 H), 3.10-3.01 (m, 1 H), 2.75-2.67 (m, 1 H), 2.52-2.36 (m, 1 H), 2.27-2.26 (m, 7 H), 1.77-1.73 (m, 1 H), 1.45 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 137.80, 137.56, 137.32, 136.80, 136.35, 135.64, 129.98, 129.15, 128.42, 126.54, 126.16, 125.35, 41.37, 26.64, 24.15, 21.72, 19.73, 19.22. MS ($C_{19}H_{21}NO$): 279 (M^+). HRMS: Anal. Calcd. 279.16231, Found: 279.16189. IR (cm^{-1}): ν 2940, 1657, 1374.



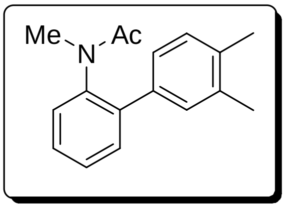
1-(8-(3,4-dimethylphenyl)-3,4-dihydro-3-methylquinolin-1(2H)-yl)ethanone (**3ba**). Follow the general procedures and 1.0 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL *o*-xylene and 1.5 mL EtCOOH, yielded 62.4 mg product in 5 h as a single isomer, 71%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.37-7.07 (m, 6 H), 5.03-4.97 (m, 0.3 H), 4.30-4.23 (m, 0.6 H), 3.36-3.20 (m, 0.5 H), 2.93-2.85 (m, 0.2 H), 2.71-2.60 (m, 0.6 H), 2.56-2.43 (m, 1 H), 2.35-2.20 (m, 6 H), 1.49 (s, 3 H), 1.24 (d, 2 H, *J* = 6.6 Hz), 0.98 (d, 1 H, *J* = 6.6 Hz). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.37, 169.85, 138.24, 137.72, 137.19, 137.04, 136.94, 136.85, 136.66, 135.85, 135.75, 134.85, 130.20, 130.10, 129.39, 129.27, 128.62, 128.54, 127.82, 126.64, 126.55, 125.96, 125.61, 125.47, 49.51, 49.05, 36.04, 34.56, 33.19, 29.41, 21.91, 21.85, 20.55, 20.29, 19.87, 19.37. MS ($C_{20}H_{23}NO$): 293 (M^+). HRMS: Anal. Calcd. 293.17796, Found: 293.17713. IR (cm^{-1}): ν 2958, 1659, 1372.



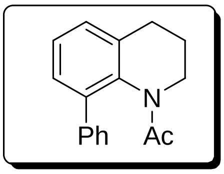
1-(8-(3,4-dimethylphenyl)-3,4-dihydro-6-methylquinolin-1(2H)-yl)ethanone (**3ca**). Follow the general procedures and 0.2 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL *o*-xylene and 1.5 mL EtCOOH, yielded 75.6 mg product in 5 h as a single isomer, 86%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.15-6.98 (m, 5 H), 4.80-4.70 (m, 1 H), 3.08-2.99 (m, 1 H), 2.71-2.63 (m, 1 H), 2.51-2.40 (m, 1 H), 2.37 (s, 3 H), 2.34-2.26 (m, 7 H), 1.79-1.70 (m, 1 H), 1.45 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.28, 137.72, 137.40, 136.91, 136.59, 136.35, 135.71, 134.96, 130.08, 129.27, 125.47, 41.54, 26.72, 24.24, 21.83, 20.97, 19.85, 19.34. MS ($C_{20}H_{23}NO$): 293 (M^+). HRMS: Anal. Calcd. 293.17796, Found: 293.17768. IR (cm^{-1}): ν 2942, 1655, 1375.



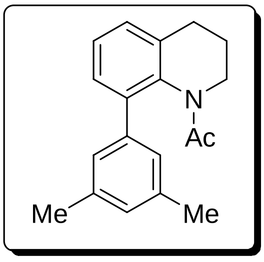
1-(8-(3,4-dimethylphenyl)-3,4-dihydro-6-methoxyquinolin-1(2H)-yl)ethanone (**3da**). Follow the general procedures and 1.0 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL *o*-xylene and 1.5 mL EtCOOH, yielded 59.3 mg product in 7 h as a single isomer, 64%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.26-7.05 (m, 3 H), 6.83 (d, 1 H, *J* = 2.7 Hz), 6.73 (d, 1 H, *J* = 2.7 Hz), 4.80-4.71 (m, 1 H), 3.81 (s, 3 H), 3.08-2.99 (m, 1 H), 2.71-2.64 (m, 1 H), 2.51-2.41 (m, 1 H), 2.36-2.43 (m, 7 H), 1.77-1.72 (m, 1 H), 1.49(s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.47, 157.94, 139.36, 138.76, 137.00, 136.46, 136.04, 130.58, 129.24, 125.46, 113.23, 111.95, 55.37, 41.56, 27.14, 24.09, 21.77, 19.88, 19.38. MS ($C_{20}H_{23}NO_2$): 309 (M^+). HRMS: Anal. Calcd. 309.17288, Found: 309.17383. IR (cm^{-1}): ν 2934, 1654, 1378.



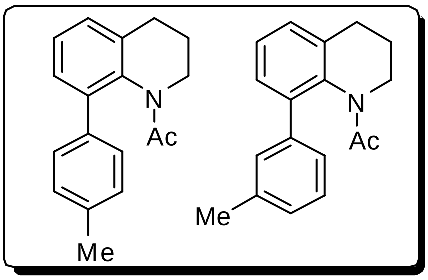
N-(3',4'-dimethylbiphenyl-2-yl)-N-methylacetamide (3ea). Follow the general procedures and 1.0 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL *o*-xylene and 1.5 mL EtCOOH, yielded 12.1 mg product in 12 h as a single isomer, 16%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.40-7.35 (m, 3 H), 7.22-7.14 (m, 2 H), 7.05-7.02 (m, 2 H), 3.02 (s, 3 H), 2.28 (s, 3 H), 1.77 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.69, 141.84, 139.80, 136.78, 136.13, 131.34, 129.89, 129.53, 128.40, 128.29, 128.26, 125.58, 37.00, 22.31, 19.88, 19.43. MS ($C_{17}H_{19}NO$): 253 (M^+). HRMS: Anal. Calcd. 253.14666, Found: 253.14660. IR (cm^{-1}): ν 2919, 1661, 1376.



1-(8-phenyl-3,4-dihydroquinolin-1(2H)-yl)ethanone (**3ab**). Follow the general procedures and 1 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL PhH and 1.5 mL EtCOOH, yielded 49.7 mg product in 8 h, 66%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.41-7.15 (m, 8 H), 4.82-4.72 (m, 1 H), 3.07-2.99 (m, 1 H), 2.75-2.67 (m, 1 H), 2.52-2.41 (m, 1 H), 2.32-2.28 (m, 1 H), 1.78-1.69 (m, 1 H) 1.48 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 169.6, 138.7, 137.66, 137.23, 137.17, 128.6, 127.9, 127.0, 126.5, 126.5, 41.2, 26.4, 23.9, 21.4. MS ($C_{17}H_{17}NO$): 251 (M^+). IR (cm^{-1}): ν 2946, 1657, 1375.



1-(8-(3,5-dimethylphenyl)-3,4-dihydroquinolin-1(2H)-yl)ethan one (**3ac**). Follow the general procedures and 1 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL *m*-xylene and 1.5 mL EtCOOH, yielded 38.5 mg product in 7 h as a single isomer, 46%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.31-7.15 (m, 3 H), 6.95 (s, 2 H), 4.81-4.72 (m, 1 H), 3.10-3.02 (m, 1 H), 2.75-2.69 (m, 1 H), 2.54-2.43 (m, 1 H), 2.33 (s, 6 H), 1.81-1.72 (m, 1 H), 1.51 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.09, 138.92, 138.25, 138.01, 137.88, 137.53, 129.07, 128.71, 126.64, 126.43, 125.94, 41.56, 26.83, 24.31, 21.87, 21.35. MS ($C_{19}H_{21}NO$): 279 (M^+). IR (cm^{-1}): ν 2946, 1658, 1374.

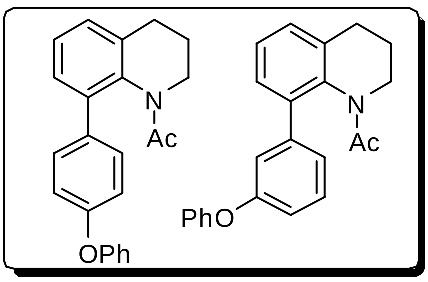


1-(8-p-tolyl-3,4-dihydroquinolin-1(2H)-yl)ethanone (**3ad**) and **1-(8-m-tolyl-3,4-dihydroquinolin-1(2H)-yl)ethanone** (**3ad′**). Follow the general procedures and 0.1 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL toluene and 1.5 mL EtCOOH, yielded 62.0 mg product in 12 h as 1.1:1 isomers (determined by GC), 78%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.25-7.04 (m, 7 H), 4.77-4.70 (m, 1 H), 3.01-2.69 (m, 1 H), 2.67-2.64 (m, 1 H), 2.47-2.43(m, 1 H), 2.30-2.29(d, 3 H), 2.25-2.23 (m, 1 H), 1.71-1.64 (m, 1 H) 1.40-1.39 (d, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 169.66, 169.58, 138.61, 138.01, 137.62, 137.58, 137.31, 137.19, 137.16, 136.73, 135.70, 129.29, 128.49, 128.42, 128.32, 128.18, 127.80, 127.67, 126.42, 126.41, 126.28, 126.14, 124.90, 41.20, 26.43, 26.41, 23.94, 21.45, 21.15, 20.72. MS ($C_{18}H_{19}NO$): 265 (M^+). IR (cm^{-1}): ν 2942, 1657, 1373.

Assignment of ^{13}C NMR peaks:

3ad: 169.66, 137.58, 137.19, 136.73, 135.70, 129.29, 128.18, 127.67, 126.42, 126.14, 41.20, 26.41, 23.94, 21.45, 20.72.

3ad': 169.58, 138.61, 138.01, 137.62, 137.31, 137.16, 128.49, 128.42, 128.32, 127.80, 126.41, 126.28, 124.90, 41.20, 26.43, 23.94, 21.45, 21.15.

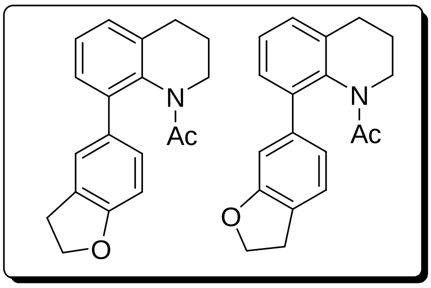


1-(8-(4-phenoxyphenyl)-3,4-dihydroquinolin-1(2H)-yl)ethanone (**3ae**) and **1-(8-(3-phenoxyphenyl)-3,4-dihydroquinolin-1(2H)-yl)ethanone** (**3ae′**). Follow the general procedures and 0.2 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1 mL PhOPh and 1.5 mL EtCOOH, yielded 71.0 mg product in 4 h as 2.5:1 isomers (determined by GC), 69%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.42-6.94 (m, 12 H), 4.82-4.73 (m, 1 H), 3.06-2.98 (m, 1 H), 2.77-2.68 (m, 1 H), 2.54-2.43 (m, 1 H), 2.34-2.58 (m, 1 H), 1.78-1.69 (m, 1 H) 1.53-1.46 (m, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.03, 157.68, 156.87, 156.52, 156.47, 140.59, 138.01, 137.59, 137.52, 136.99, 136.90, 133.66, 130.27, 129.70, 129.54, 128.47, 126.97, 126.81, 126.75, 126.59, 123.51, 123.40, 122.76, 119.22, 118.77, 118.05, 117.66, 41.51, 41.41, 26.68,24.17, 21.77. MS ($C_{23}H_{21}NO_2$): 343 (M^+). HRMS: Anal. Calcd. 343.15723, Found: 343.15694 (standard product 3ae), 343.15666 (standard product 3ae'). IR (cm^{-1}): ν 2950, 1654, 1376.

Assignment of ^{13}C NMR peaks:

3ae: 170.03, 156.87, 156.47, 138.01, 137.52, 136.90, 133.66, 129.70, 129.54, 128.47, 126.81, 126.59, 123.51, 119.22, 118.77, 41.51, 26.68, 24.17, 21.77.

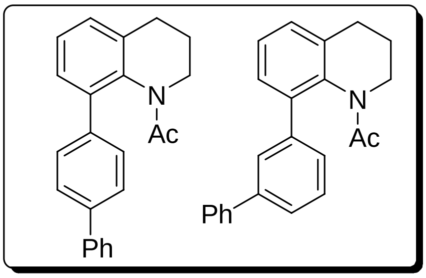
3ae': 170.03, 157.68, 156.52, 140.59, 138.01, 137.59, 136.99, 130.27, 129.70, 128.47, 126.97, 126.75, 123.40, 122.76, 119.22, 118.05, 117.66, 41.41, 26.68, 24.17, 21.77.



1-(8-(2,3-dihydrobenzofuran-5-yl)-3,4-dihydro-quinolin-1(2H)-yl)ethanone (**3af**) and **1-(8-(2,3-dihydrobenzofuran-6-yl)-3,4-dihydroquinolin-1(2H)-yl)ethanone** (**3af′**). Follow the general procedures and 0.2 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 0.4 mL dihydrobenzofuran and 2.0 mL EtCOOH, yielded 60.7 mg product (purified on neutral aluminium oxide) in 7 h as 9.8:1 isomers (determined by GC. Indeed, Other two isomes were also observed in a <0.5:<0.5:1:9.8 ratio. The major isomer was confirmed by comparing with standard product, the minor isomers were not determined due to the difficulty of synthesizing their standard products), 69%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.29-6.82 (m, 5 H), 6.81-6.77 (m, 1 H), 4.78-4.71 (m, 1 H), 4.57 (t, 2 H, J = 8.7 Hz), 3.27-3.15 (m, 2 H), 2.75-2.68 (m, 1 H), 2.53-2.42 (m, 1 H), 2.34-2.26 (m, 1 H), 1.79-1.71 (m, 1 H), 1.51 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.05, 159.47, 137.85, 137.56, 137.26, 131.16, 128.49, 128.41, 128.11, 127.52, 126.60, 126.47, 125.97, 124.52, 109.58, 71.17, 41.41, 29.46, 26.63, 24.12, 21.74. MS ($C_{19}H_{19}NO_2$): 293 (M^+). HRMS: Anal. Calcd. 293.14158, Found: 293.14139 (standard product 3af). IR (cm^{-1}): ν 2944, 1655, 1375.

Assignment of ^{13}C NMR peaks:

3af: 170.05, 159.47, 137.85, 137.56, 137.26, 131.16, 128.41, 128.11, 127.52, 126.60, 125.97, 124.52, 109.58, 71.17, 41.41, 29.46, 26.63, 24.12, 21.74.

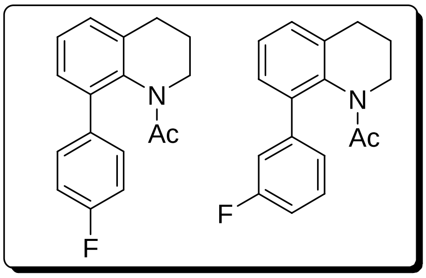


1-(8-(biphenyl-4-yl)-3,4-dihydroquinolin-1(2H)-yl)ethanone (**3ag**) and **1-(8-(biphenyl-3-yl)-3,4-dihydroquinolin-1(2H)-yl)ethanone** (**3ag′**). Follow the general procedures and 0.2 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate and 5 eq. biphenyl in 2.0 mL EtCOOH, yielded 42.2 mg product in 13 h as 1.0:1 isomers (determined by GC), 43%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.65-7.17 (m, 12 H), 4.85-4.76 (m, 1 H), 3.13-3.05 (m, 1 H), 2.77-2.70 (m, 1 H), 2.55-2.44 (m, 1 H), 2.35-2.31 (m, 1 H), 1.80-1.74 (m, 1 H), 1.50(s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.08, 170.02, 141.87, 140.80, 140.19, 140.06, 139.38, 138.10, 137.88, 137.57, 137.40, 137.08, 129.32, 128.68, 128.62, 128.53, 127.53, 127.31, 127.09, 126.96, 126.85, 126.83, 126.80, 126.26, 41.62, 41.57, 26.77, 26.72, 24.26, 24.21, 21.85, 21.81. MS ($C_{23}H_{21}NO$): 327 (M^+). HRMS: Anal. Calcd. 327.16231, Found: 327.16286 (standard product 3ag’, for 3ag, see ref. 1). IR (cm^{-1}): ν 2947, 1655, 1374.

Assignment of ^{13}C NMR peaks:

3ag: 170.08, 140.19, 140.06, 137.88, 137.57, 137.08, 128.68, 128.53, 127.53, 127.31, 126.85, 126.83, 126.80, 41.57, 26.72, 24.21, 21.85, .

3ag’: 170.02, 141.87, 140.80, 139.38, 138.10, 137.57, 137.40, 129.32, 128.68, 128.62, 127.53, 127.09, 126.96, 126.85, 126.26, 41.62, 26.77, 24.26, 21.81

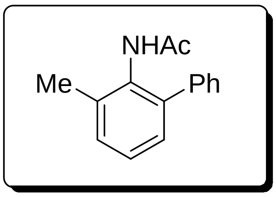


1-(8-(4-fluorophenyl)-3,4-dihydroquinolin-1(2H)-yl)ethanone (**3ah**) and **1-(8-(4-fluorophenyl)-3,4-dihydroquinolin-1(2H)-yl)ethanone (3ah′).** Follow the general procedures and 0.2 eq $Pd(OAc)_2$, 1.0 eq. $Cu(OTf)_2$ were used. Starting from 0.3 mmol substrate in 1 mL PhF and 1.5 mL EtCOOH, yielded 38.7 mg product in 6 h as 2.3:1 isomers (determined by ^{1}H NMR, for they had the same retention time on GC), 48%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.39-7.04 (m, 7 H), 4.83-4.73 (m, 1 H), 3.08-2.99 (m, 1 H), 2.79-2.71 (m, 1 H), 2.56-2.45(m, 1 H), 2.37-2.29(d, 1 H), 1.83-1.74 (m, 1 H), 1.51-1.46 (m, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.07, 163.82, 160.53, 138.17, 137.64, 136.58, 135.05, 130.54, 130.44, 129.99, 129.88, 128.57, 127.38, 126.97, 123.96, 116.13, 115.85, 115.39, 115.39, 115.10, 114.53, 114.25, 41.66, 26.76, 24.24, 21.78. MS ($C_{17}H_{16}FNO$): 269 (M^+). IR (cm^{-1}): ν 2942, 1657, 1374.

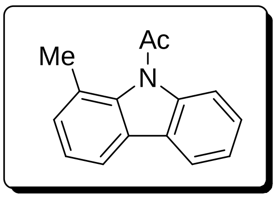
Assignment of ^{13}C NMR peaks:

3ah: 170.07, 160.53, 138.18, 136.58, 135.05, 129.99, 129.88, 128.54, 127.36, 116.13, 115.85, 41.66, 26.76, 24.24, 21.78.

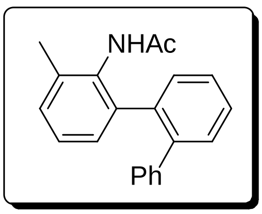
3ah': 170.07, 163.82, 160.53, 138.17, 137.64, 130.54, 130.44, 128.57, 127.38, 126.97, 123.96, 115.39, 115.39, 115.10, 114.53, 114.25, 41.66, 26.76, 24.24, 21.78.



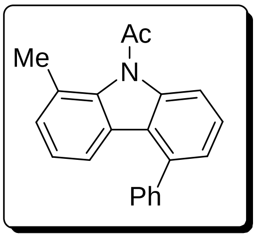
N-(3-methylbiphenyl-2-yl)acetamide (**3fb**). Follow the general procedures and 1 eq. $Cu(OTf)_2$ was used. Starting from 0.3 mmol substrate in 1.0 mL PhH and 1.5 mL EtCOOH, yielded 45.0 mg product in 8 h, 66%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.42-7.14 (m, 8 H), 6.74 (s, 1 H),2.30 (s, 3 H), 1.96 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 169.36, 139.59, 139.52, 136.77, 132.57, 130.76, 128.79, 128.47, 128.16, 127.82, 127.36, 127.30, 22.94, 18.57, 18.55. MS ($C_{15}H_{15}NO$): 225 (M^+). HRMS: Anal. Calcd. 225.11536, Found: 225.11535. IR (cm^{-1}): ν 3252, 1655, 1523.



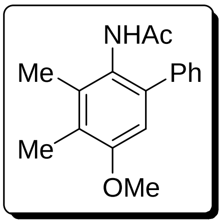
1-(1-methyl-9H-carbazol-9-yl)ethanone (**8a**). According to literature procedures using 20 mol% $Pd(OAc)_2$. Starting from 0.2 mmol substrate yielded 40.1 mg product in 12 h, 90%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.94-7.89 (m, 2 H), 7.81-7.78 (m, 1 H), 7.45-7.27 (m, 4 H), 2.67 (s, 3 H), 2.47 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.74, 139.64, 138.90, 130.03, 127.67, 126.97, 126.57, 126.23, 124.06, 123.25, 119.97, 117.24, 114.30, 26.96, 21.25. MS ($C_{15}H_{13}NO$): 223 (M^+). HRMS: Anal. Calcd. 223.09971, Found: 223.09964. IR (cm^{-1}): ν 2961, 1702, 1320.



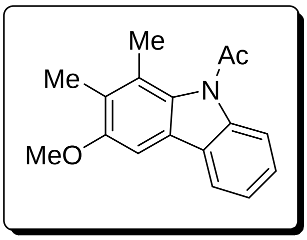
***N*-Acetyl-2-methyl-6-(2-phenylphenyl)-aniline** (**7**). Follow the general procedures and 0.2 eq $Pd(OAc)_2$, 1.0 eq. $Cu(OTf)_2$ were used. Starting from 0.2 mmol substrate in 0.6 mL PhH and 1.5 mL EtCOOH, yielded 23.0 mg product in 6 h, 38%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.46-7.32 (m, 5 H), 7.25-7.10 (m, 7 H), 6.16 (s, 1 H), 2.04 (s, 3 H), 1.81 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 168.14, 141.14, 140.17, 139.21, 137.94, 136.30, 132.57, 131.22, 130.03, 129.86, 129.59, 128.94, 128.82, 128.29, 127.99, 127.56, 127.23, 127.08, 126.99, 22.96, 18.46 . MS ($C_{21}H_{19}NO$): 301 (M^+). IR (cm^{-1}): ν 3252, 1659, 1523.



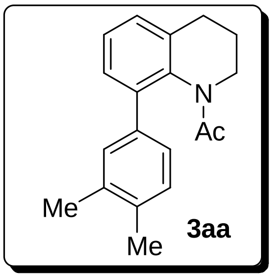
1-(1-methyl-5-phenyl-9H-carbazol-9-yl)ethanone (8b). According to literature procedures using 20 mol% $Pd(OAc)_2$. Starting from 0.2 mmol substrate yielded 52.0 mg product in 12 h, 89%. Since the starting material (**7**) contains some unseperatable **3fb**, this product is always contaminated by some carbazole **8a**. Selected data: ^{1}H NMR ($CDCl_3$, 300 MHz): δ 8.00-7.98 (m, 1 H), 7-51-7.42 (m, 5 H), 7.21-7.18 (m, 2 H), 7.04-7.00 (m, 3 H), 2.68 (s, 3 H), 2.46 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 171.33, 140.47, 140.17, 139.20, 137.58, 129.59, 129.08, 128.50, 127.74, 127.45, 127.01, 126.52, 125.50, 125.03, 123.51, 119.81, 113.10, 27.03, 20.98. MS ($C_{21}H_{17}NO$): 299 (M^+). HRMS: Anal. Calcd. 299.13101, Found: 299.13035. IR (cm^{-1}): ν 2961, 1702, 1320.



N-(5-methoxy-3,4-dimethylbiphenyl-2-yl)acetamide (3gb). Follow the general procedures and 0.2 eq $Pd(OAc)_2$, 1.0 eq. $Cu(OTf)_2$ were used. Starting from 0.3 mmol substrate in 1 mL PhH and 1.5 mL EtCOOH, yielded 33.1 mg product in 12 h, 41%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 7.39-7.30 (m, 6 H), 6.69 (s, 1 H), 3.80 (s, 1 H), 2.20 (s, 3 H), 2.17 (s, 3 H), 1.95 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 170.15, 156.45, 140.28, 137.82, 136.54, 129.01, 128.78, 128.44, 128.20, 127.33, 127.20, 125.37, 109.56, 55.65, 22.86, 15.18, 12.32. MS ($C_{17}H_{19}NO_2$): 269 (M^+). IR (cm^{-1}): ν 3252, 1655, 1464.



1-(3-methoxy-1,2-dimethyl-9H-carbazol-9-yl)ethanone (8c). According to literature procedures using 20 mol% $Pd(OAc)_2$. Starting from 0.2 mmol substrate yielded 48.6 mg product in 12 h, 91%. ^{1}H NMR ($CDCl_3$, 300 MHz): δ 8.02-7.99 (m, 1 H), 7.86-7.83 (m, 1 H), 7.38-7.25 (m, 3 H), 3.93 (s, 3 H), 2.55 (s, 3 H), 2.34 (s, 3 H), 2.29 (s, 3 H). ^{13}C NMR ($CDCl_3$, 75 MHz): δ 171.38, 155.28, 140.42, 133.98, 126.96, 126.57, 125.96, 125.67, 123.32, 119.24, 114.93, 98.50, 55.88, 26.53, 18.56, 12.49. MS ($C_{12}H_{17}NO_2$): 267 (M^+). HRMS: Anal. Calcd. 267.12593, Found: 267.12493. IR (cm^{-1}): ν 2961, 1702, 1320.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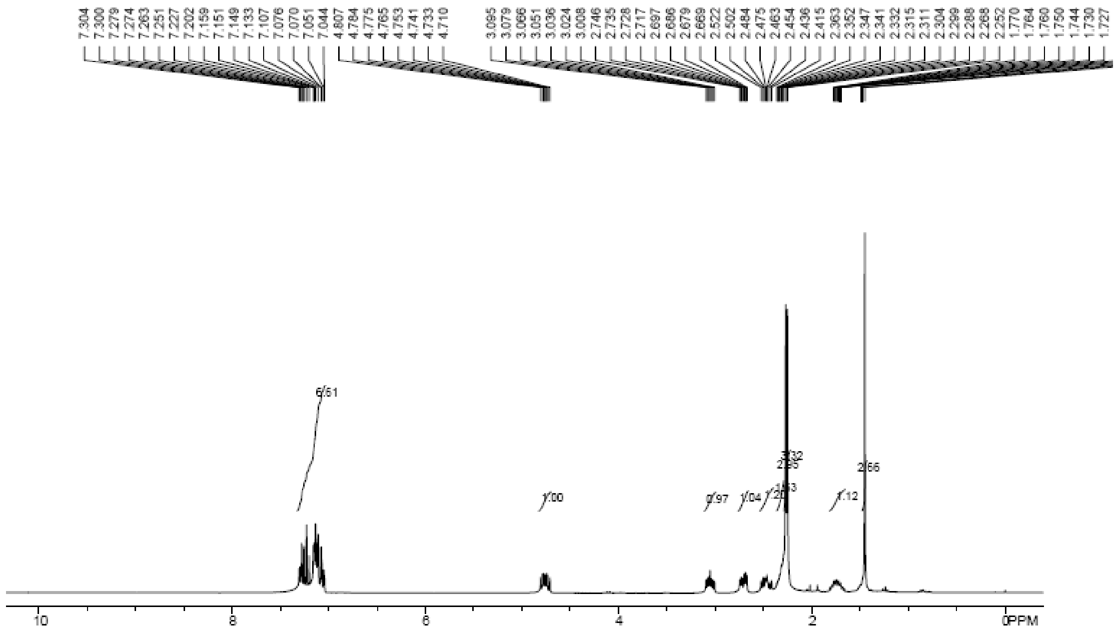
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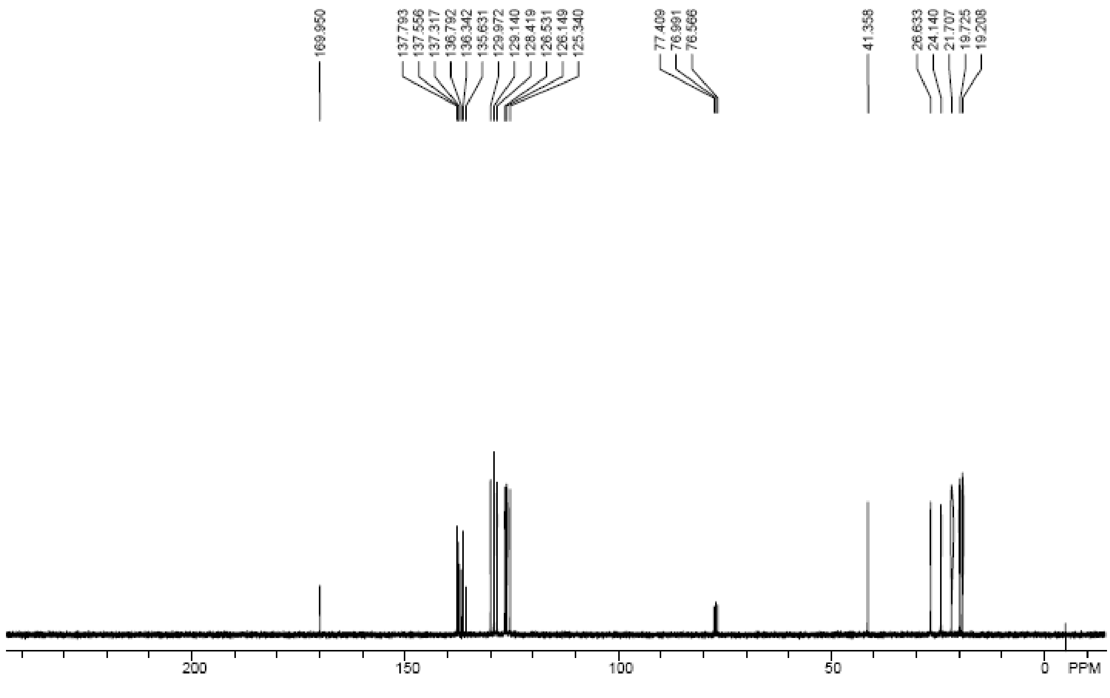
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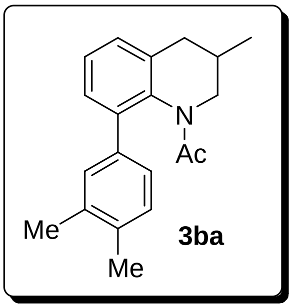
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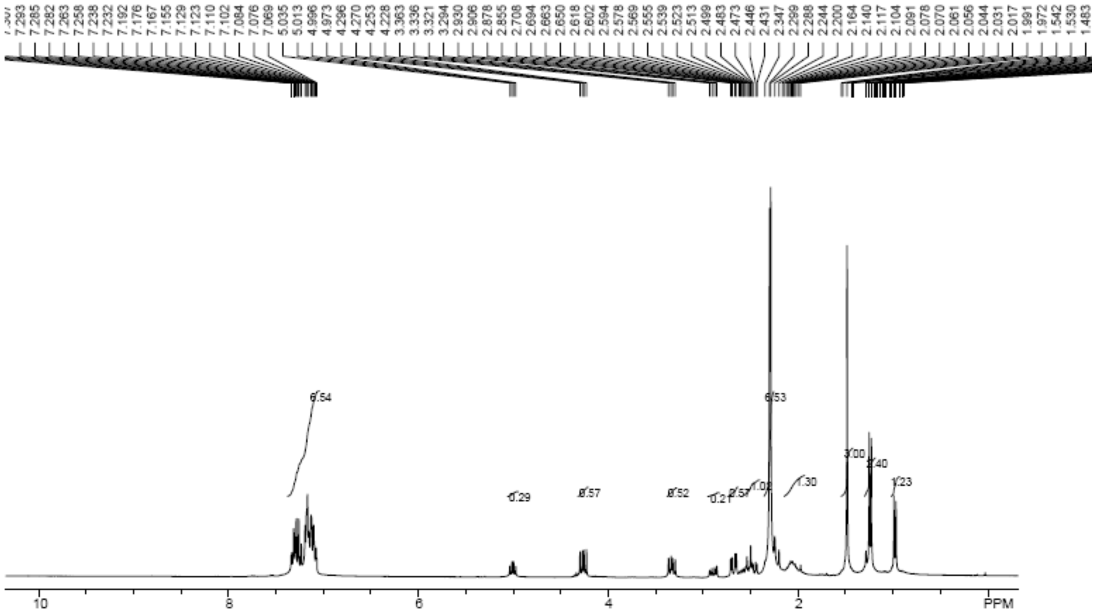
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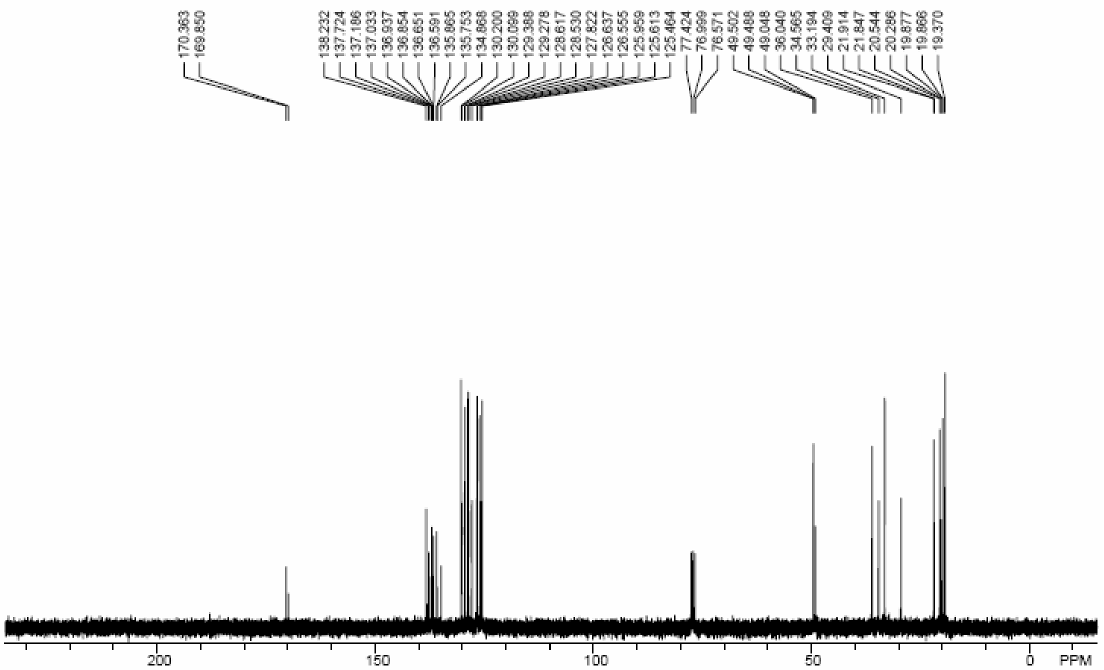
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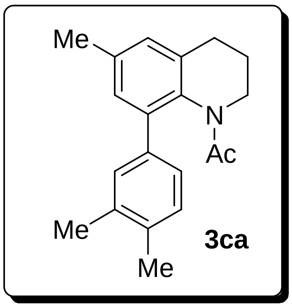
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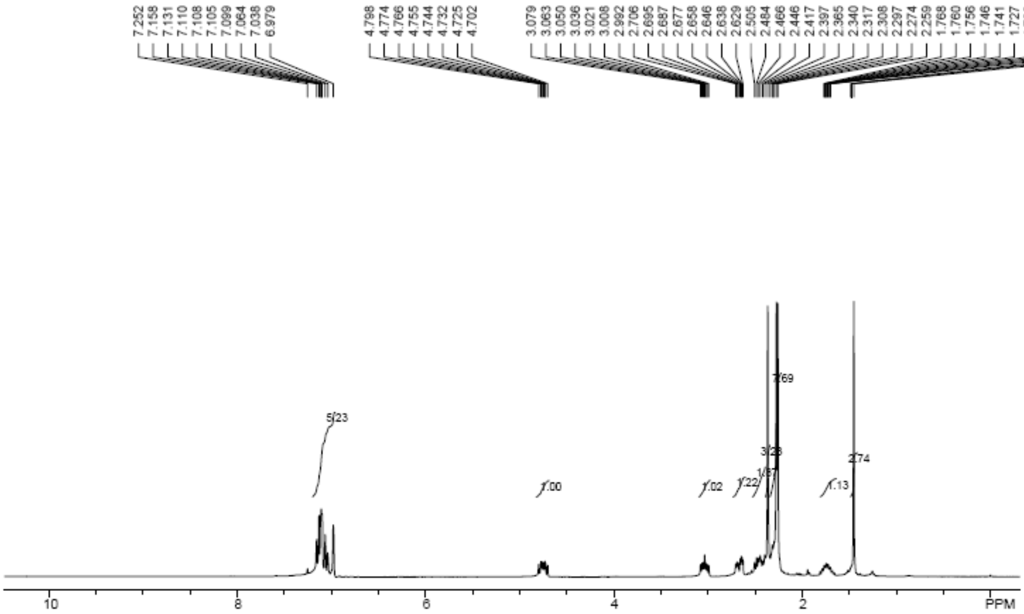
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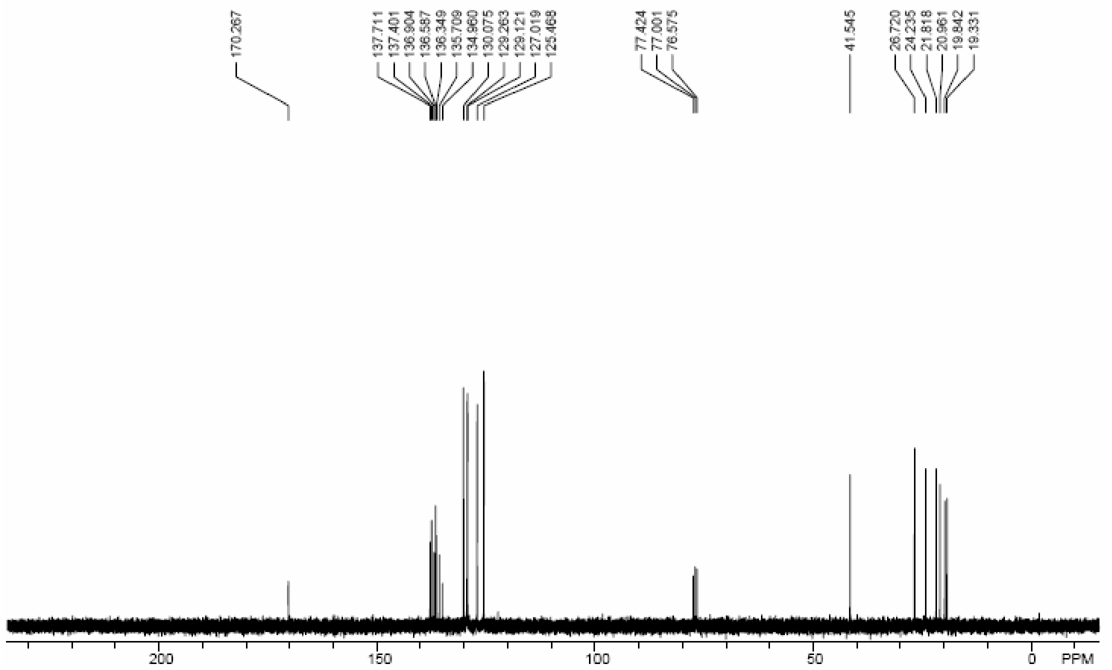
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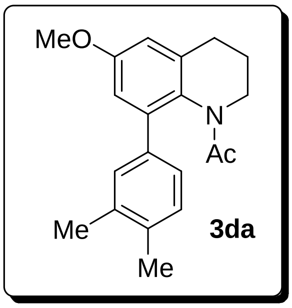
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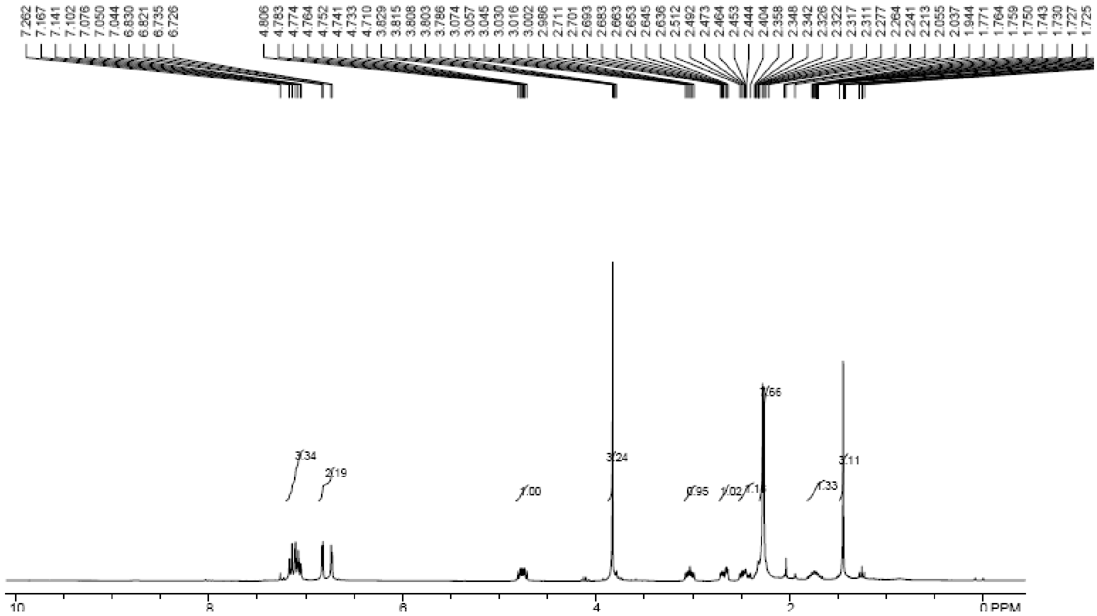
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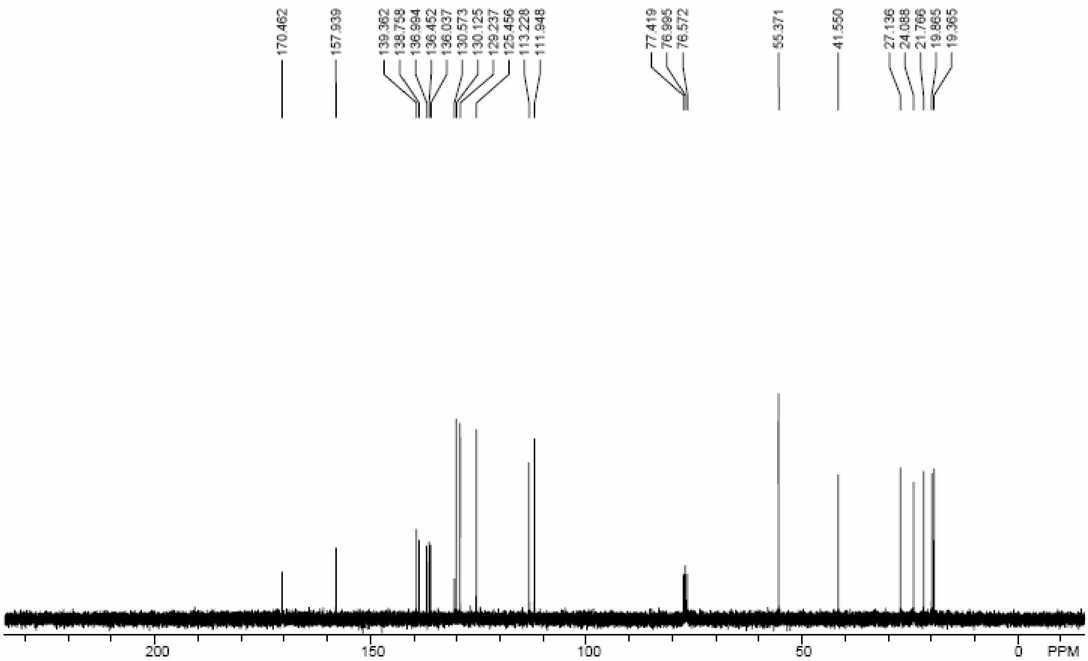
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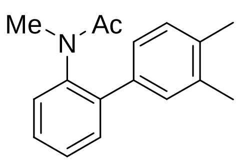

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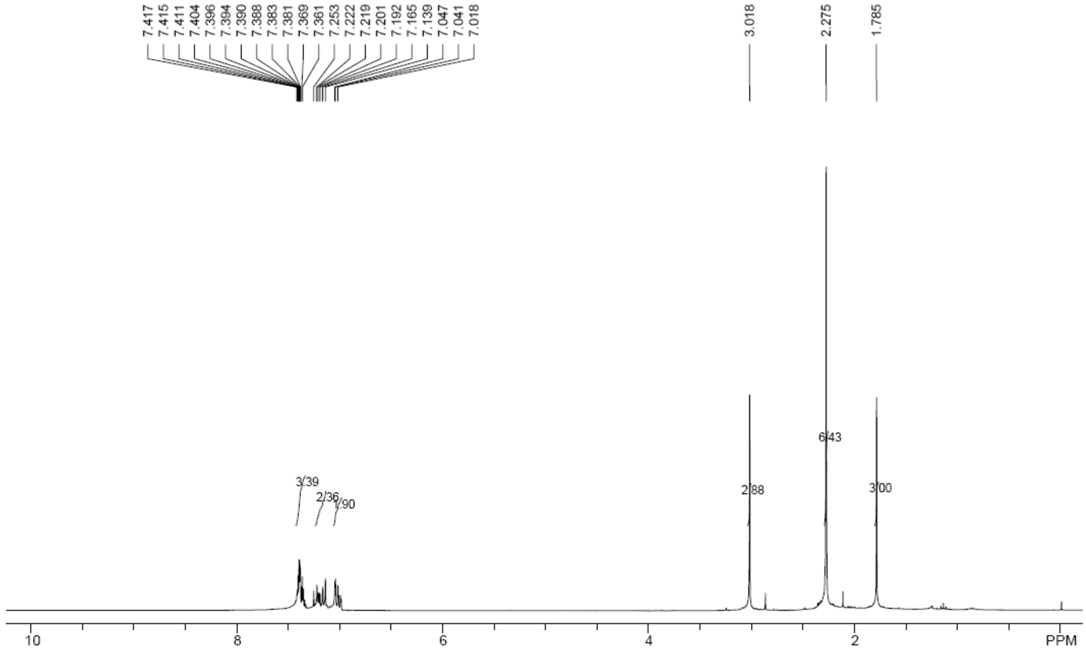

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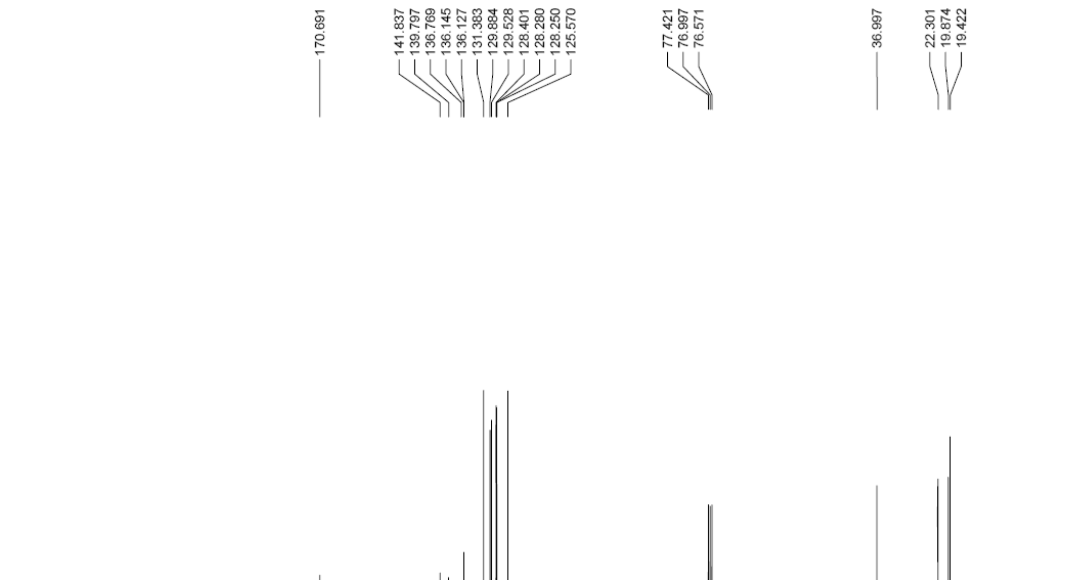

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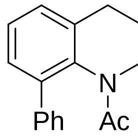

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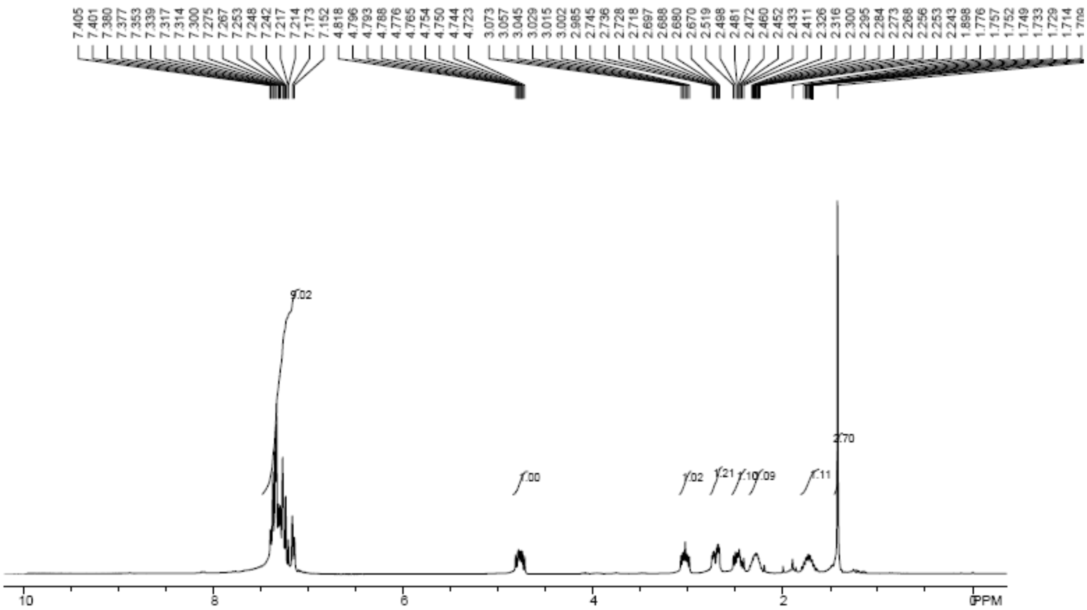

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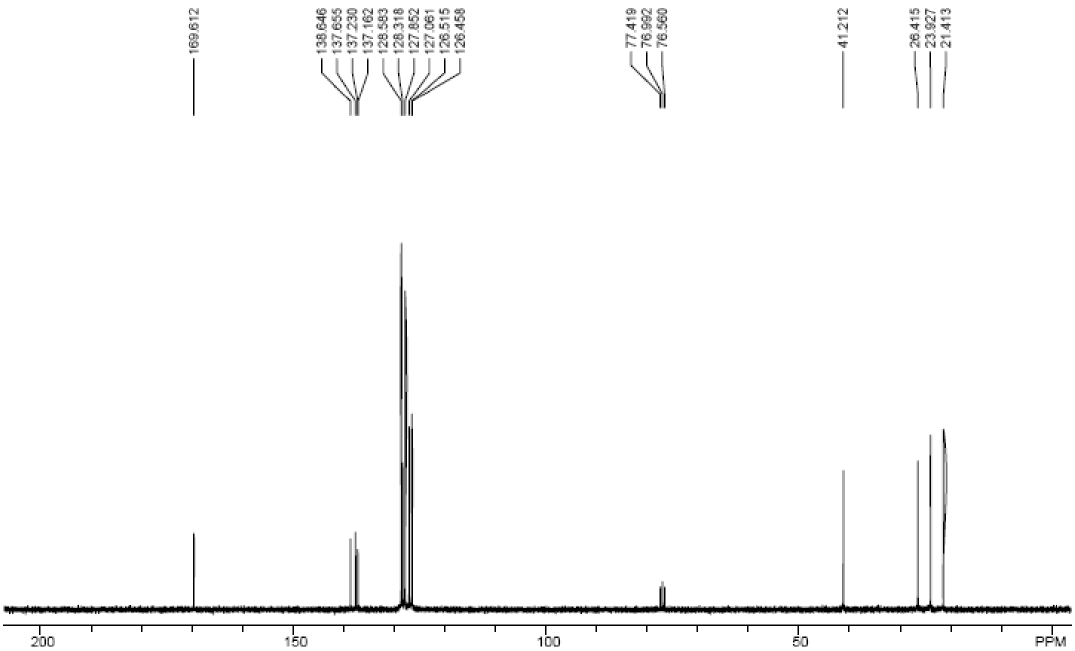

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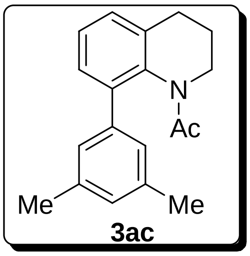
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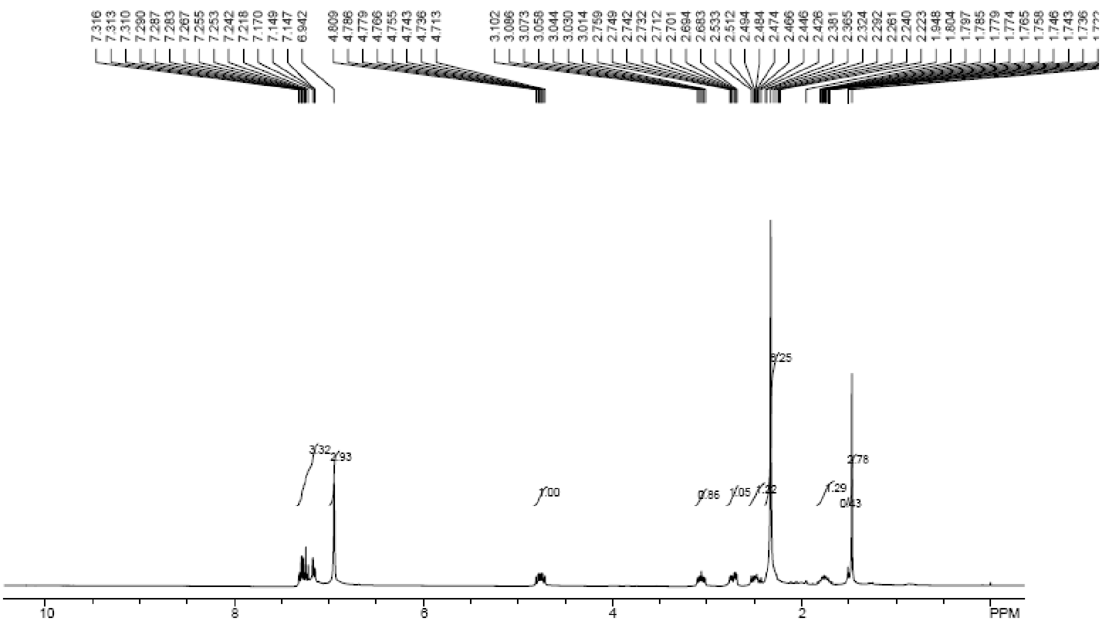
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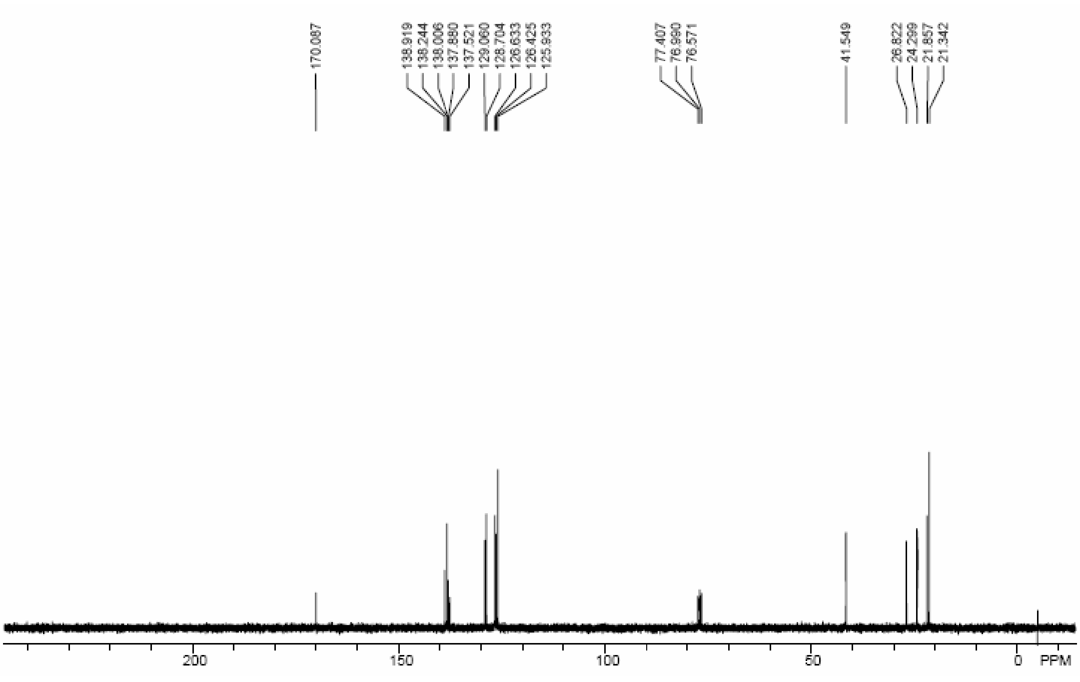
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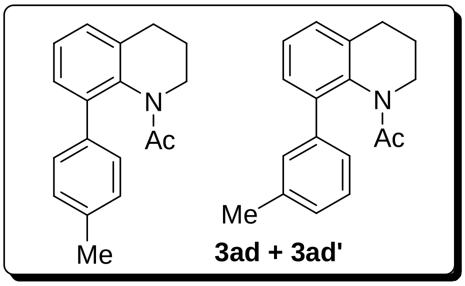
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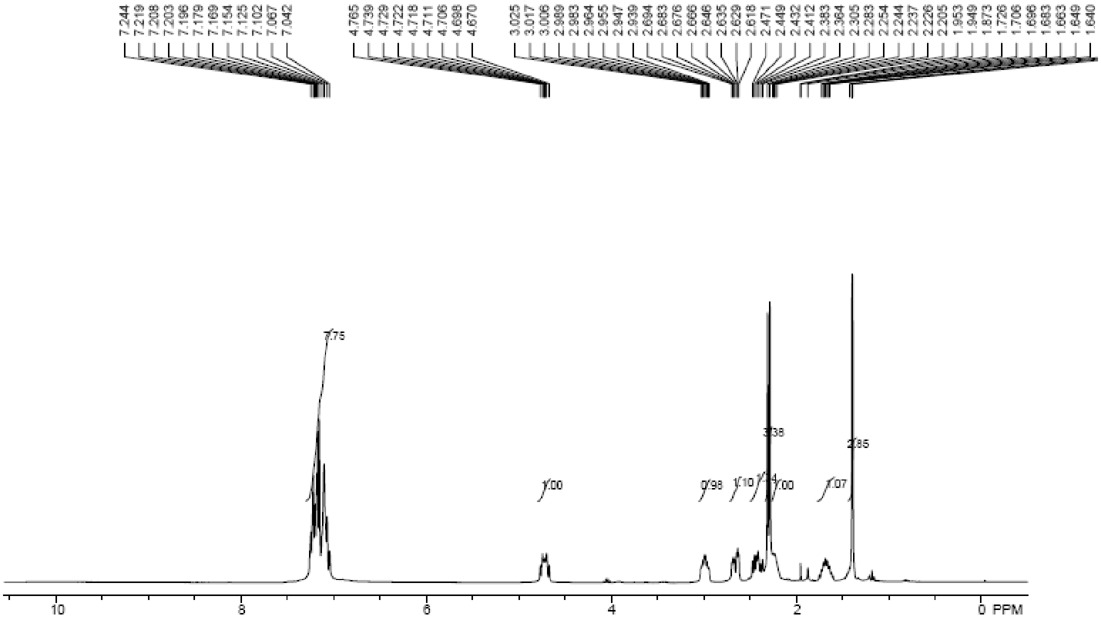
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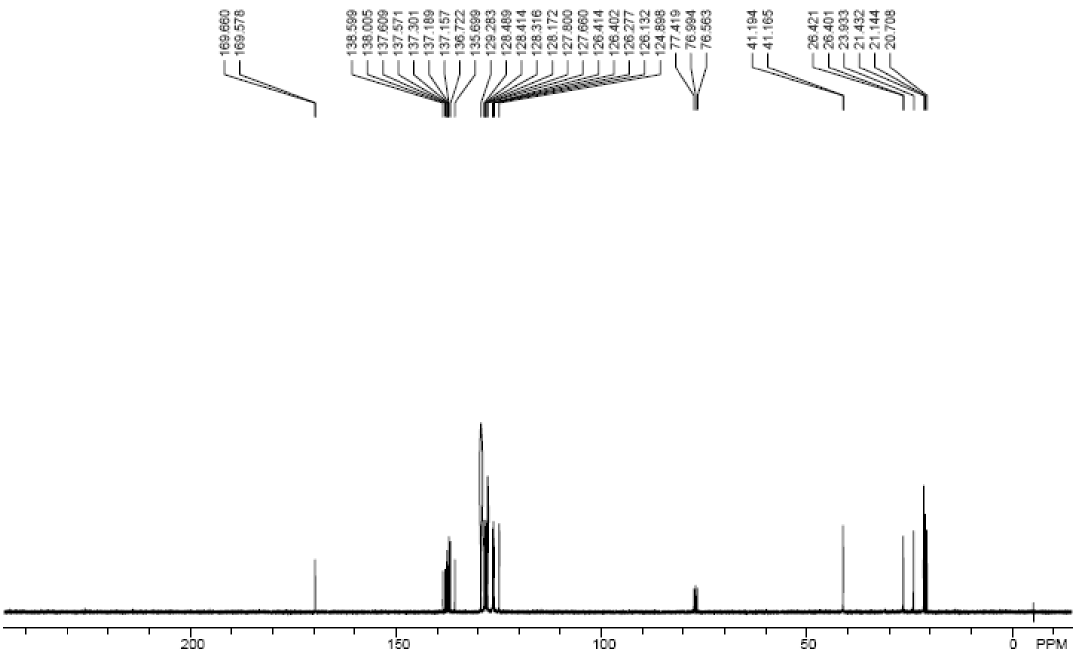
0 PPM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126.414
126.402
126.277
126.132
124.898
77.419
76.994
76.563
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26.421
26.401
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20.708
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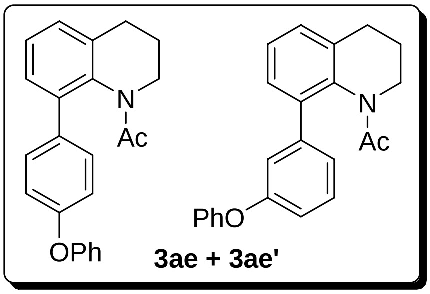
100
50
0 PPM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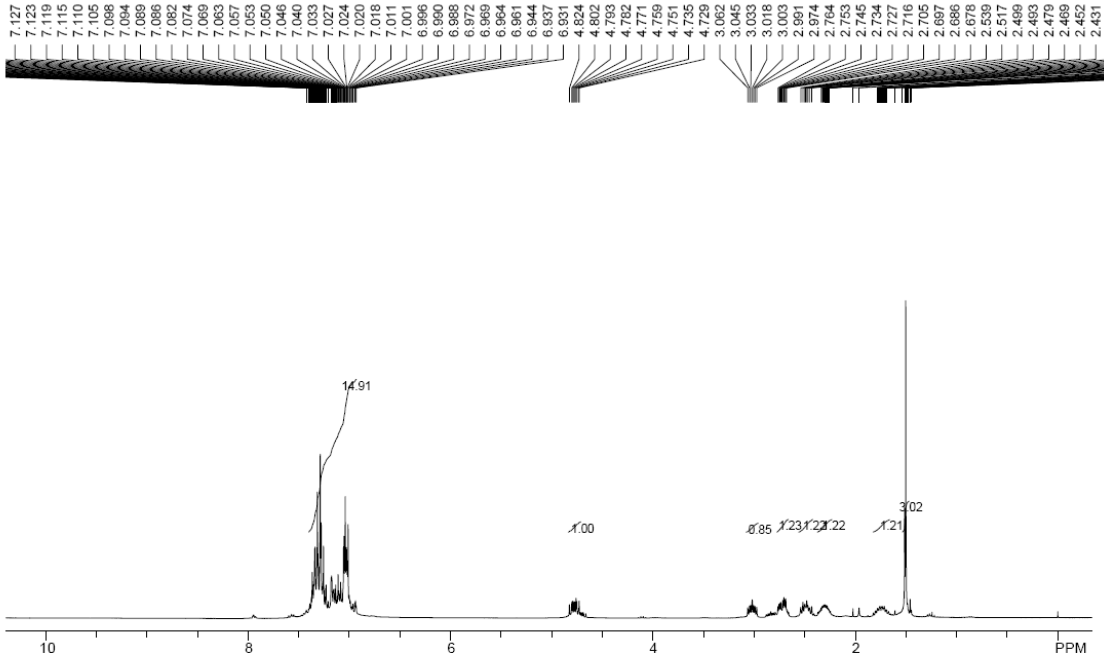
Ac
OPh
N
Ac
PhO

3ae + 3ae'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7.053
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7.046
7.040
7.033
7.027
7.024
7.020
7.018
7.011
7.001
6.996
6.990
6.988
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6.969
6.964
6.961
6.944
6.937
6.931
4.824
4.802
4.793
4.782
4.771
4.759
4.751
4.735
4.729
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3.045
3.033
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3.003
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2.753
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2.727
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2.705
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2.517
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2.493
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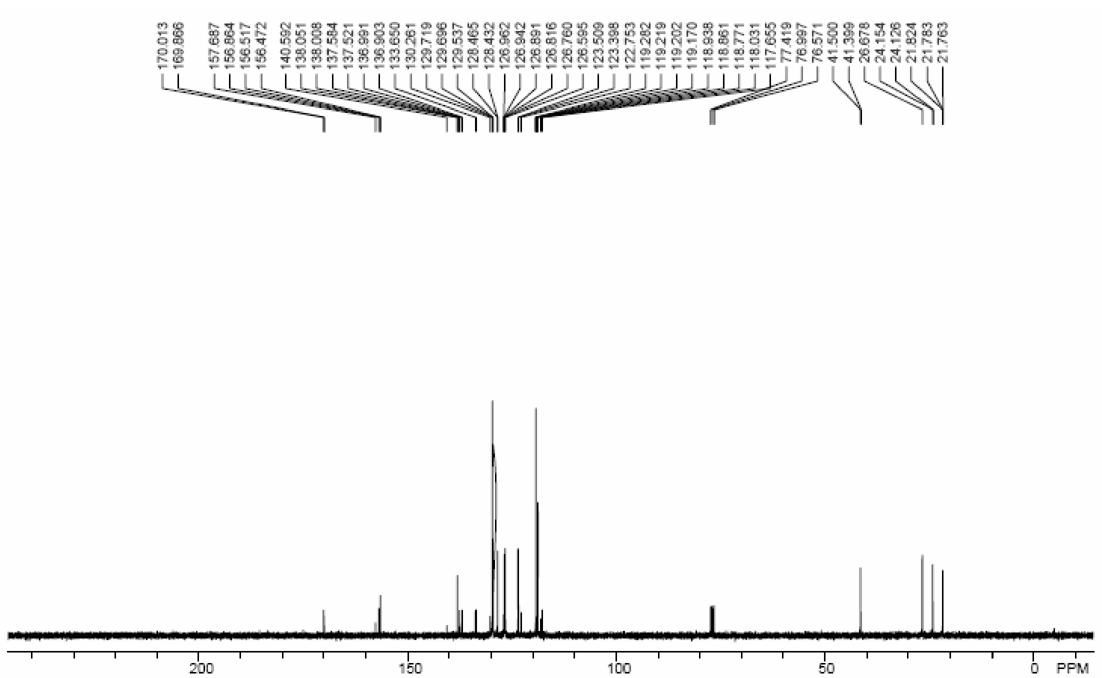
PPM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117.655
77.419
76.997
76.571
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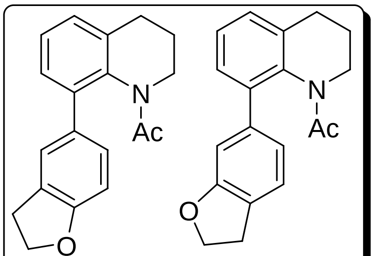
PPM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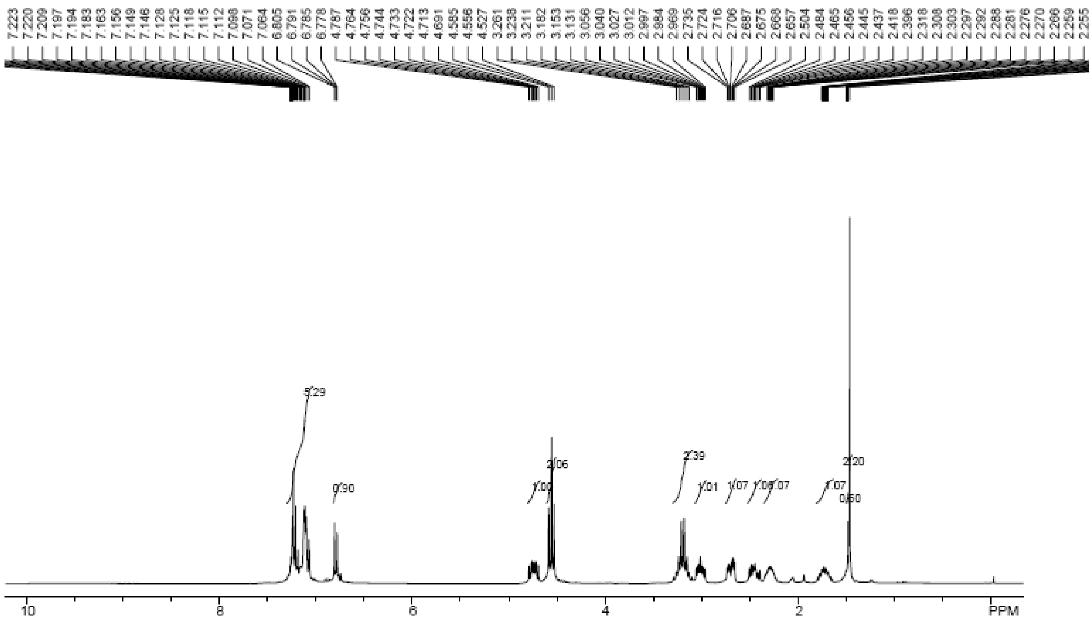
Ac
O
N
Ac
O

3af + 3af'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0.90
1.00
2.06
2.39
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1.07
1.06
1.07
1.07
2.20
0.60
10
8
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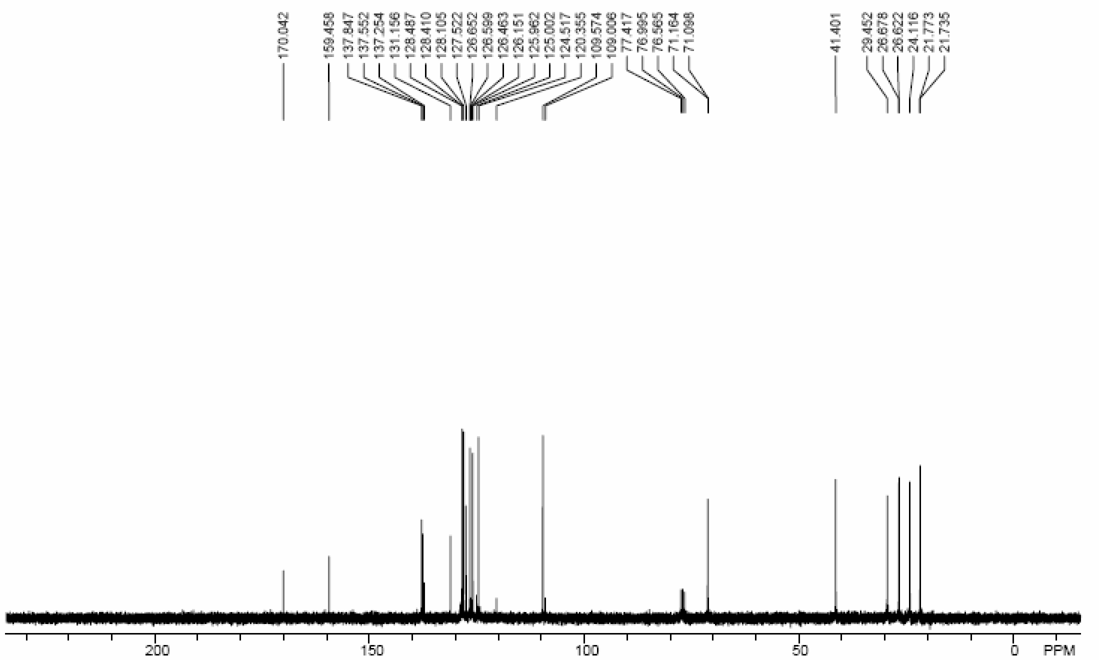
4
2
PPM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41.401
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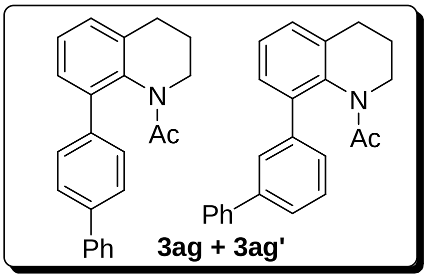
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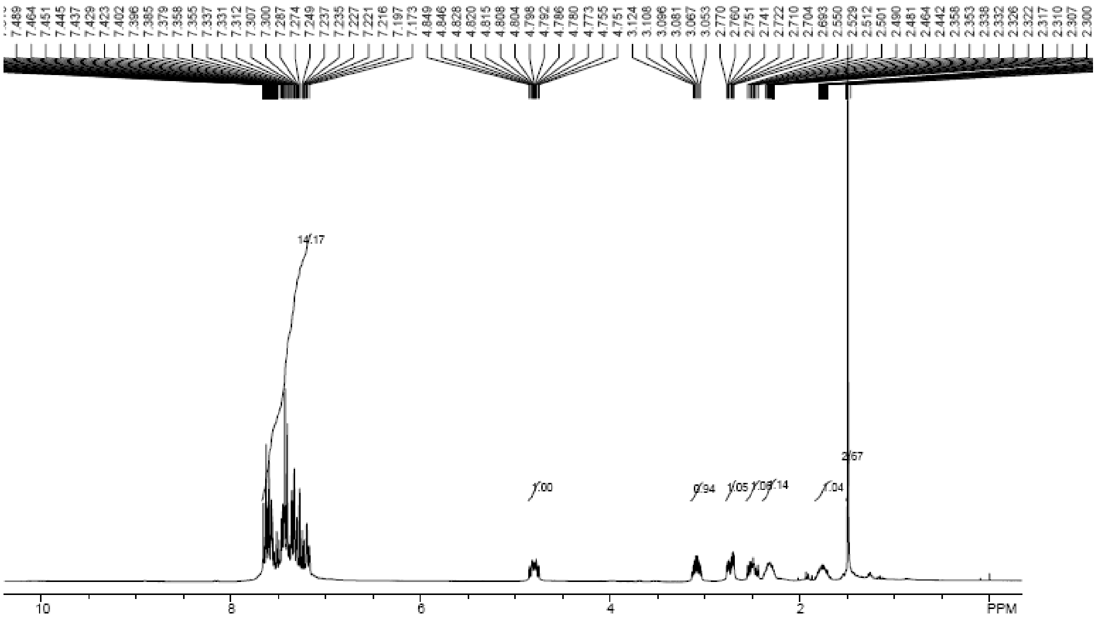
Ac
Ph
N
Ac
Ph
3ag + 3ag'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7.312
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7.249
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7.235
7.227
7.221
7.216
7.197
7.173
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4.846
4.828
4.820
4.815
4.808
4.804
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4.792
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4.751
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3.108
3.086
3.081
3.067
3.053
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2.704
2.693
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2.501
2.490
2.481
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2.353
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2.322
2.317
2.310
2.307
2.300
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4
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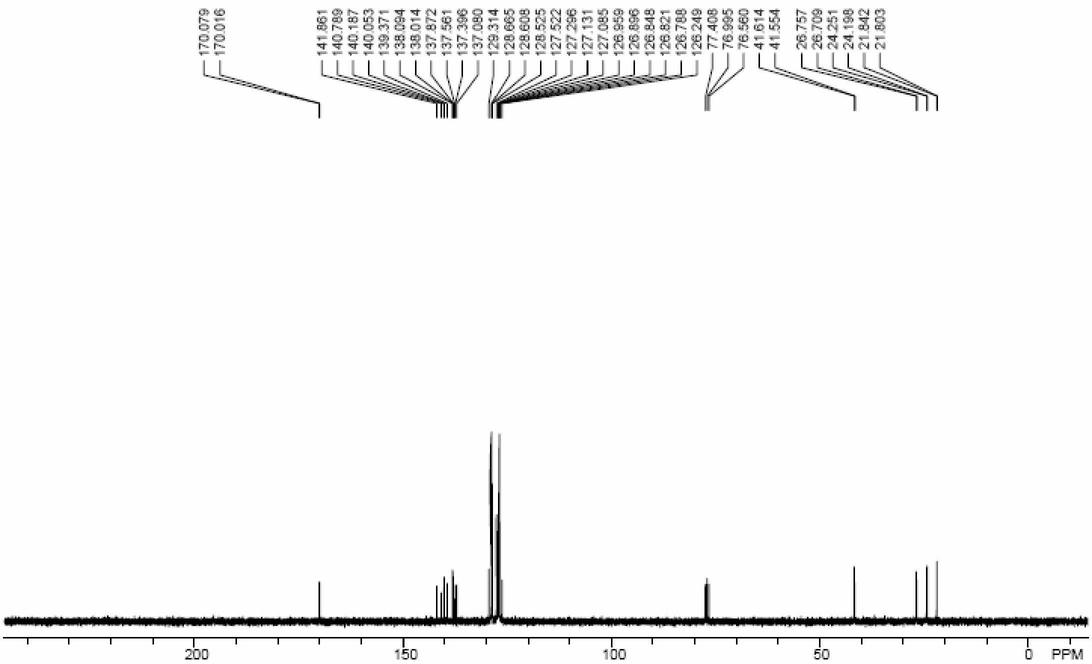
PPM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127.131
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126.821
126.788
126.249
77.408
76.995
76.560
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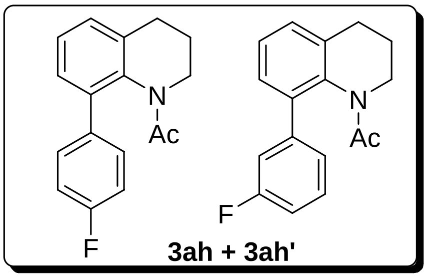
PPM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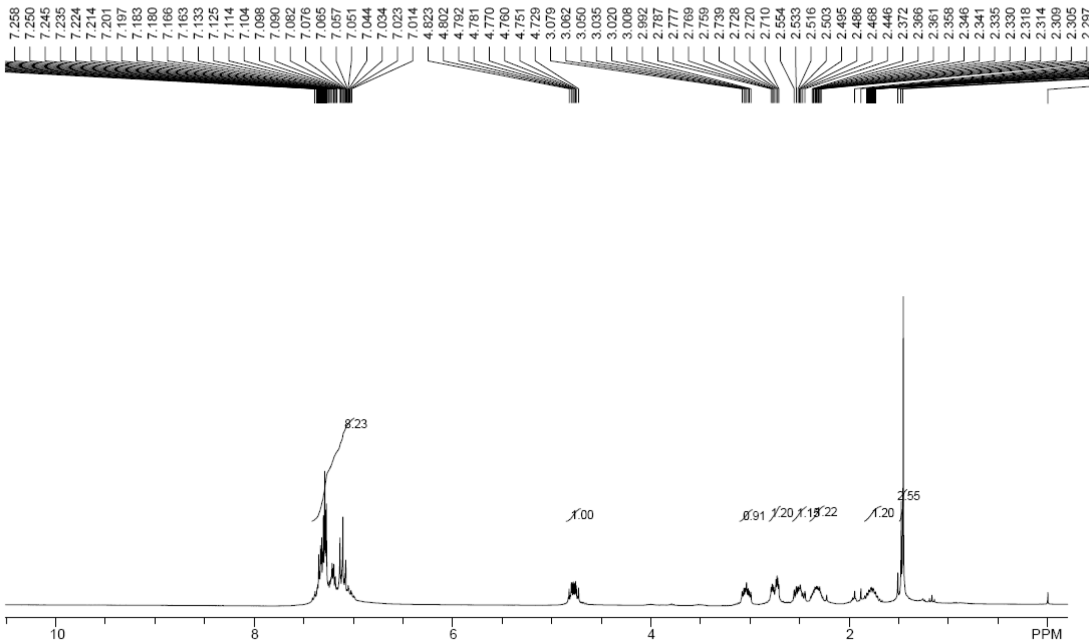
Ac
F
N
Ac
F

3ah + 3ah'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7.098
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7.076
7.065
7.057
7.051
7.044
7.034
7.023
7.014
4.823
4.802
4.792
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3.062
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2.516
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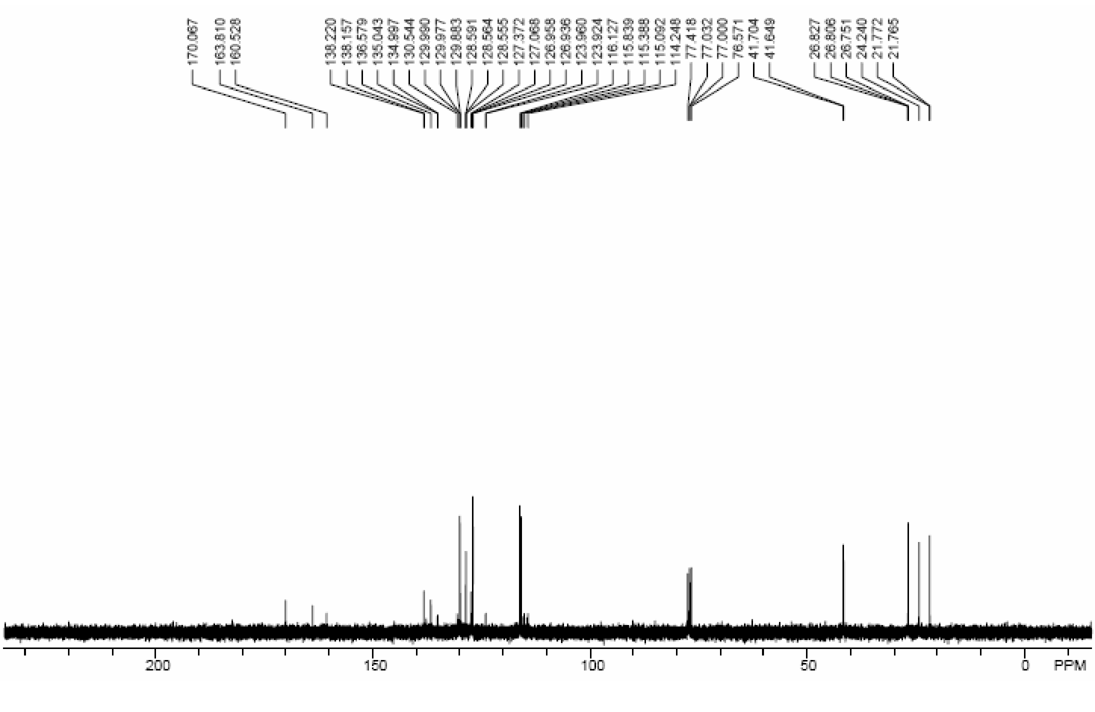
PPM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126.936
123.960
123.924
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115.388
115.092
114.248
77.418
77.032
77.000
76.571
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21.772
21.765
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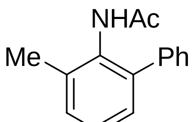
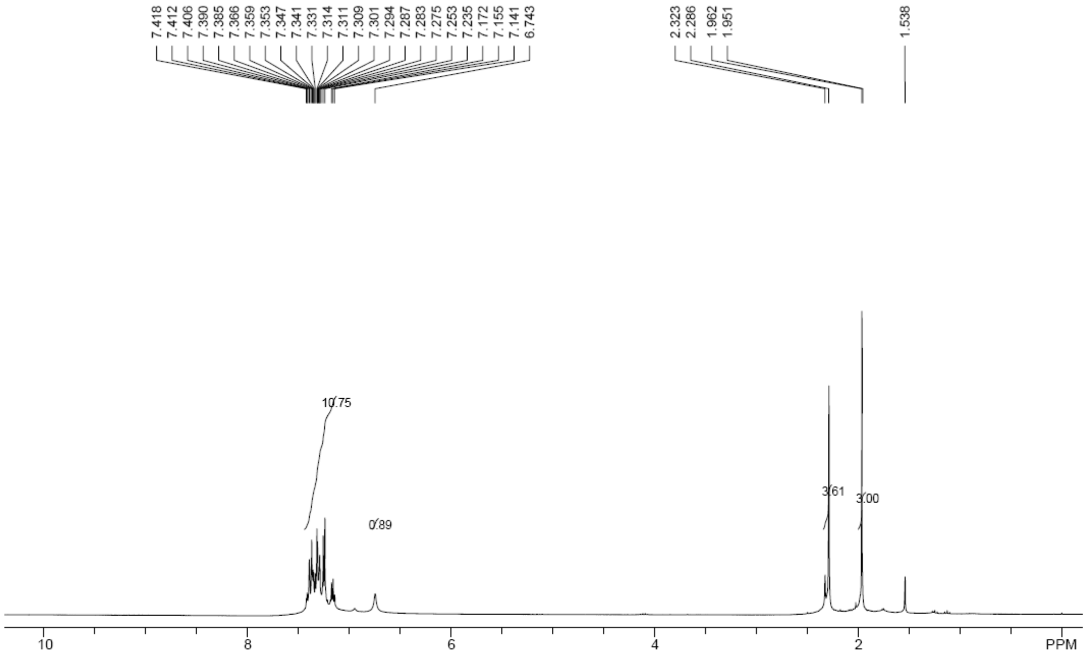
150
100
50
0
PPM


NHAc
Me
Ph

3fb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1.538
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3.00
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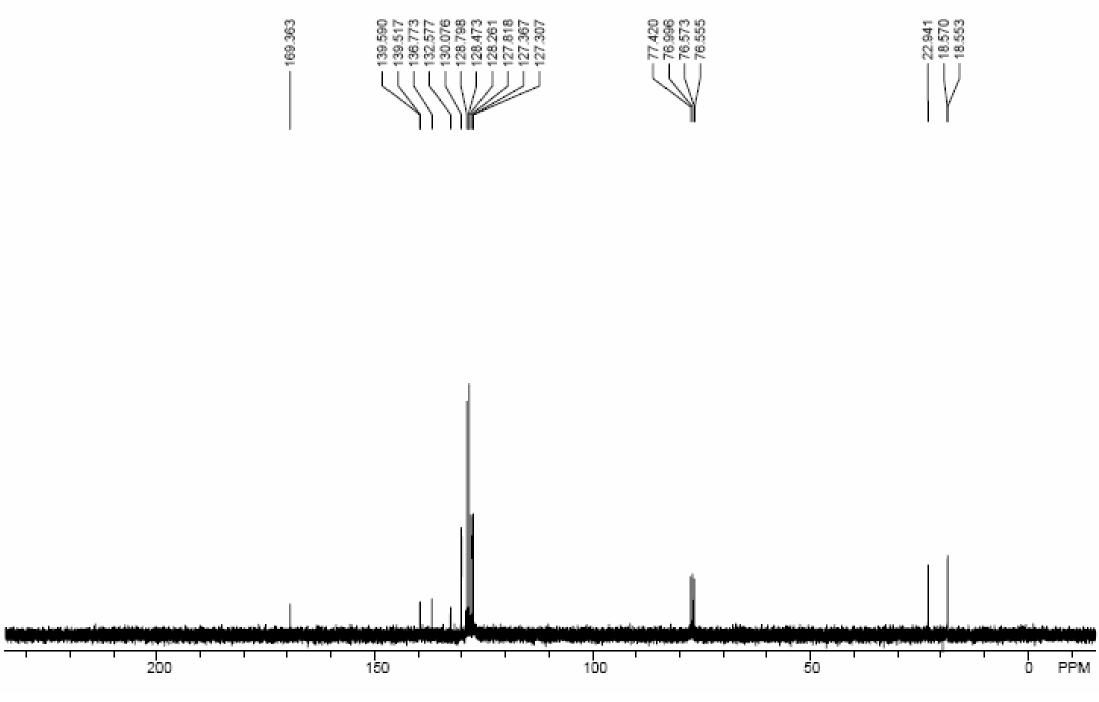
8
6
4
2
PPM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77.420 76.996 76.573 76.555
22.941 18.570 18.553
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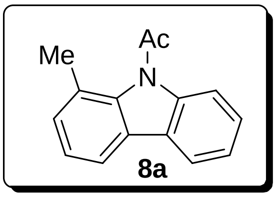
100
50
0
PPM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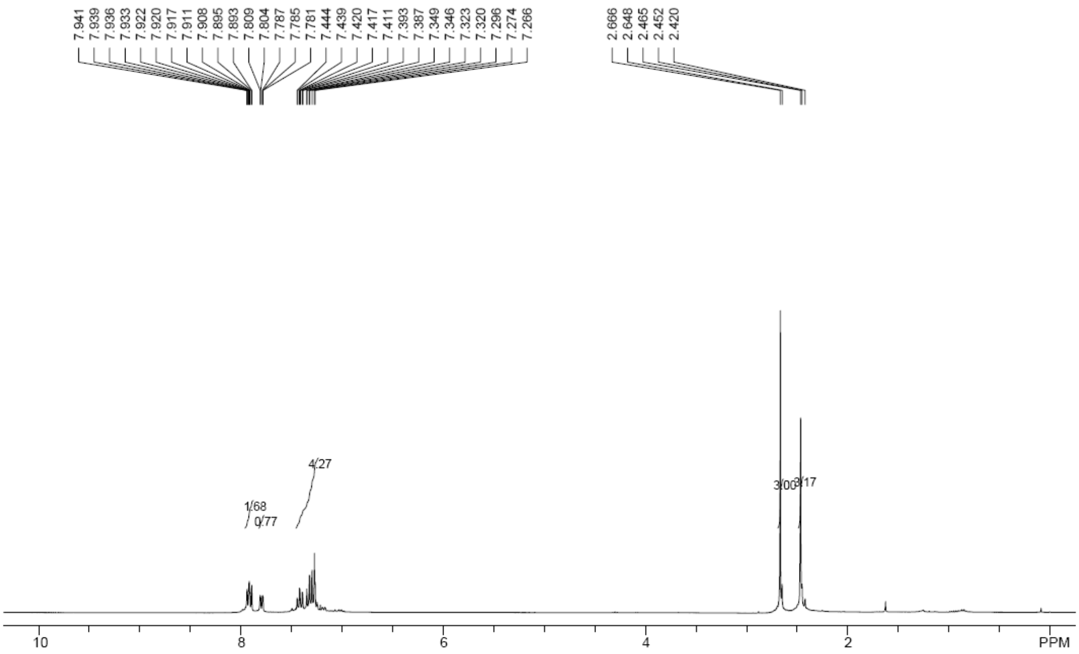
Me
N
8a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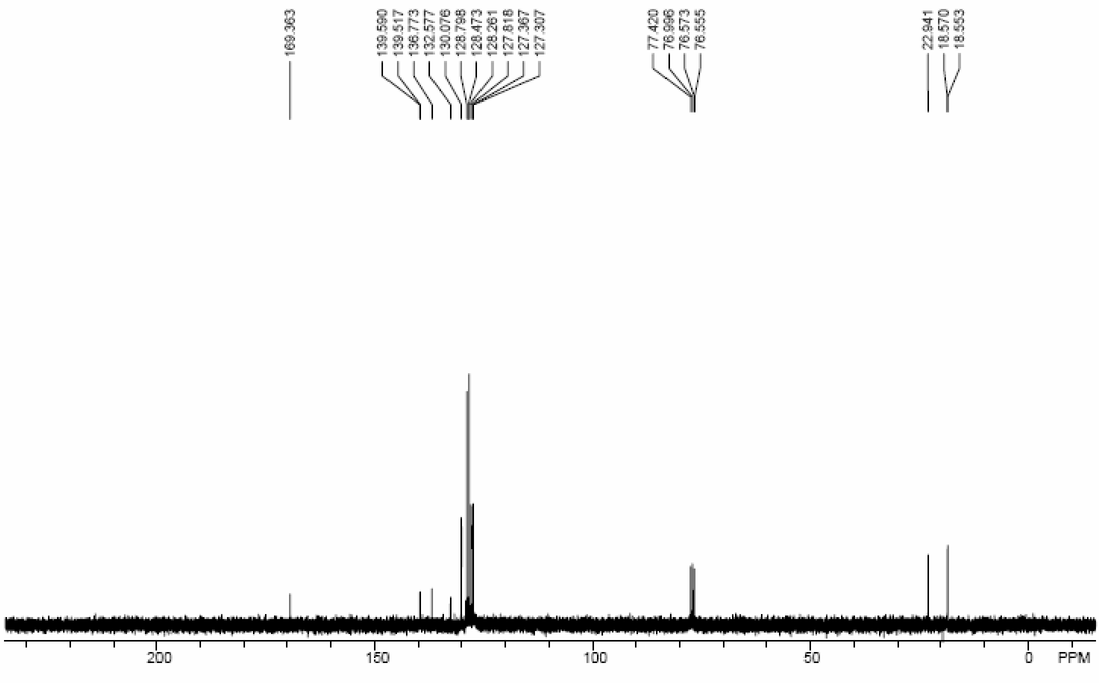
PPM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76.573
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18.553
200
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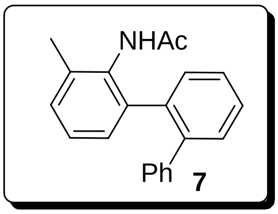
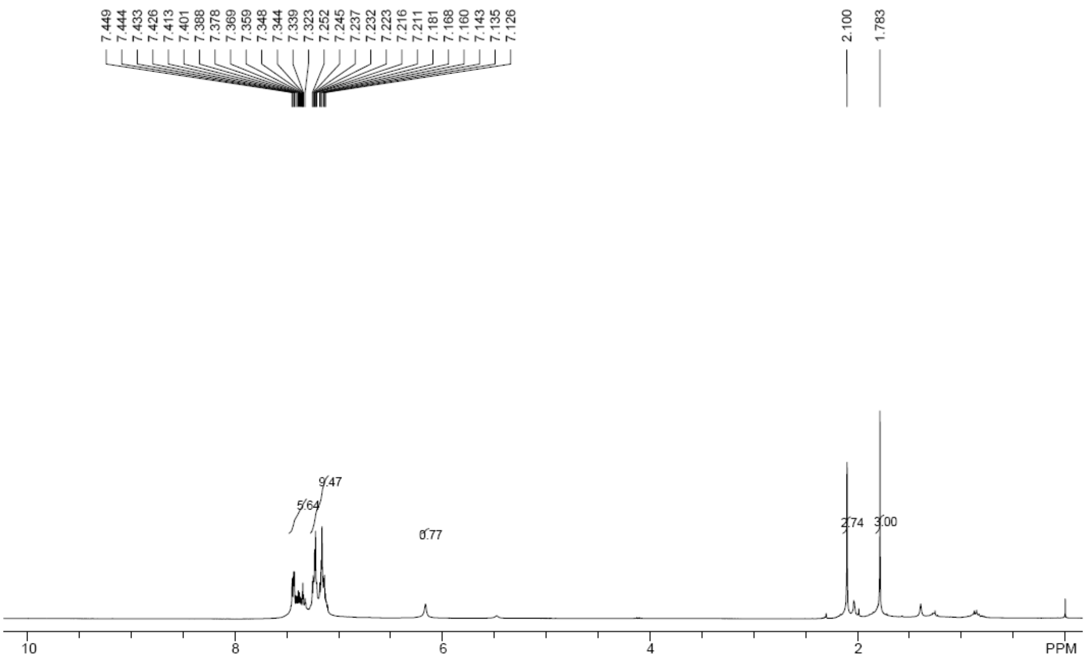
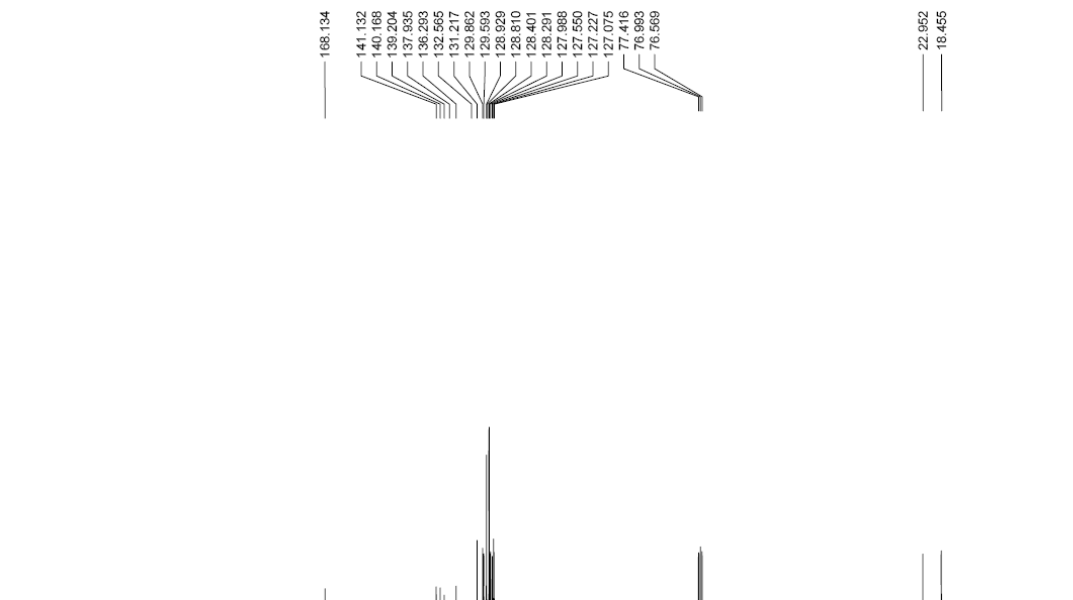
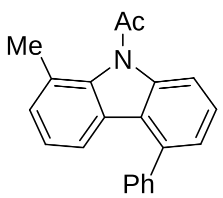
PPM



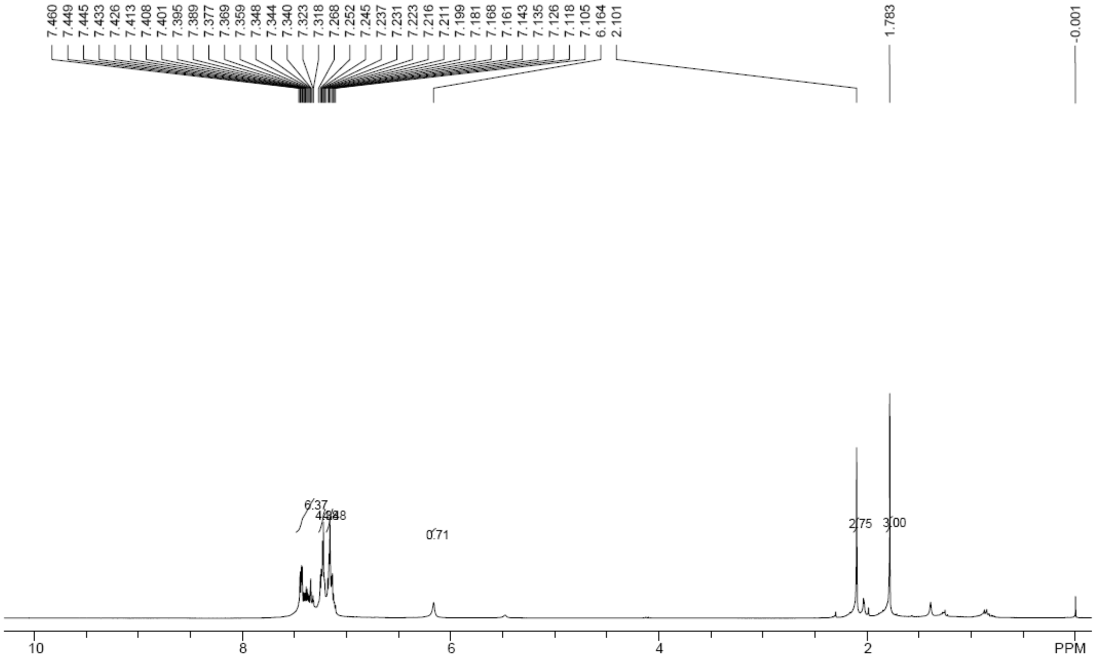
7






Ac
N
Me
Ph

8b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1.783
-0.001
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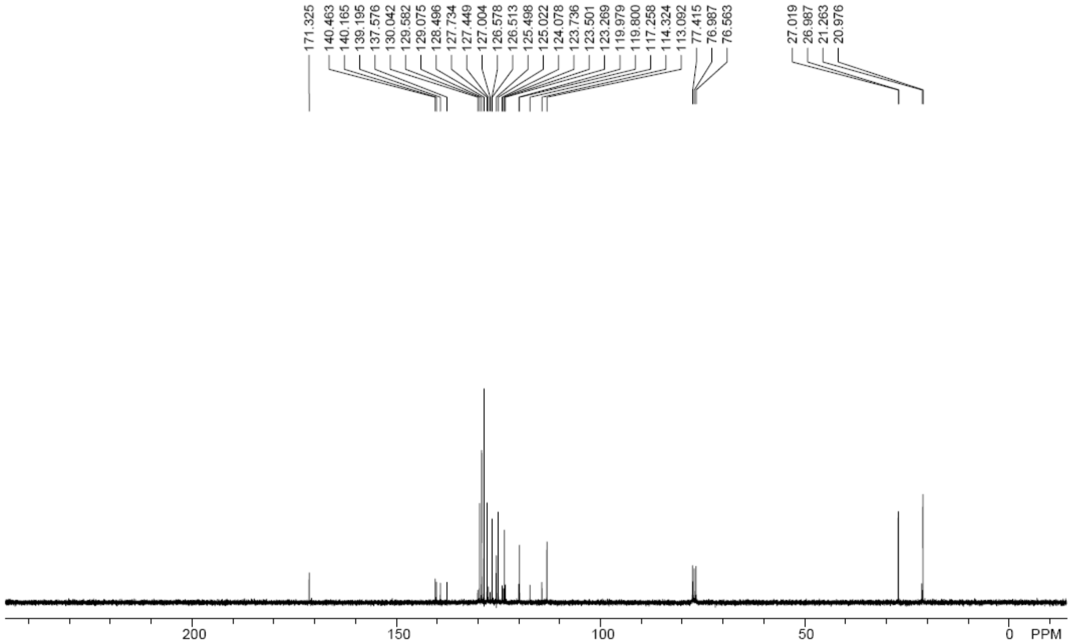
0.71
2.75
3.00
10
8
6
4
2
PPM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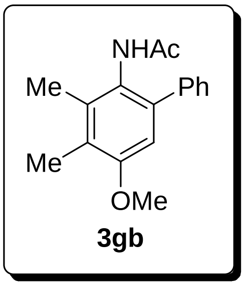
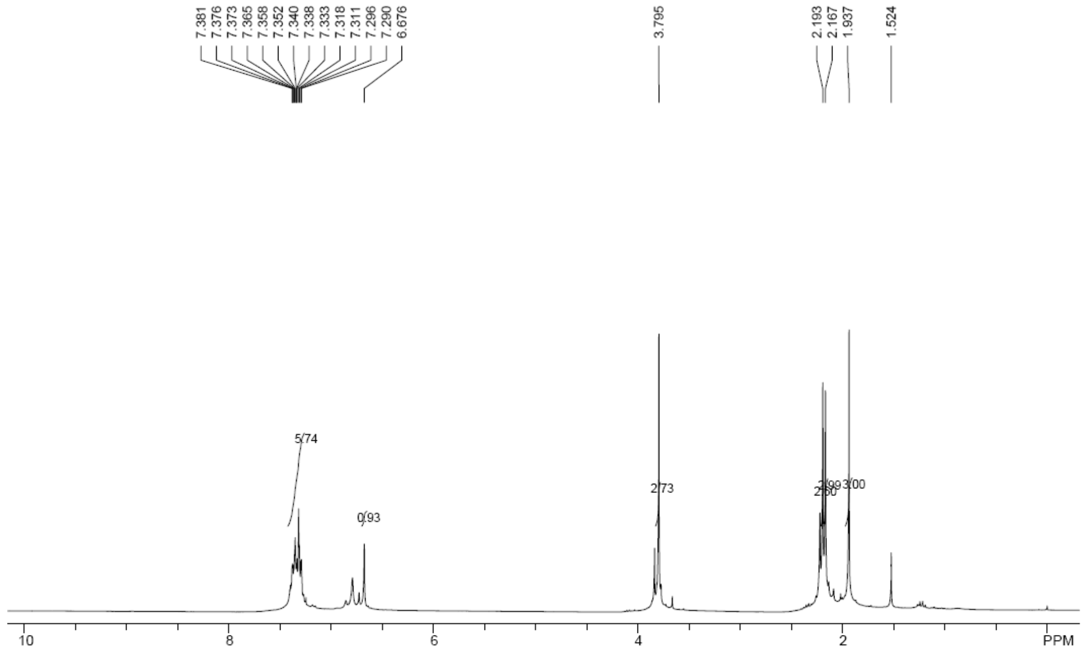
77.415 76.987 76.563
27.019 26.987 21.263 20.976
200
150
100
50
0
PPM


NHAc
Me
Ph
Me
OMe
3gb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8
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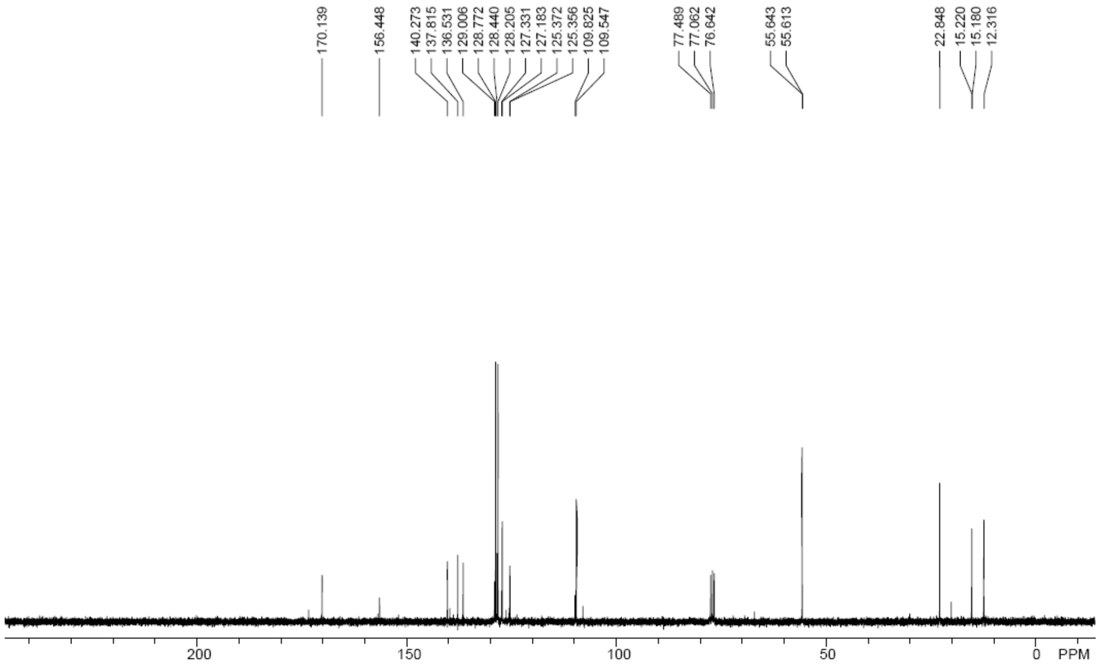
4
2
PPM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125.356
109.825
109.547
77.489
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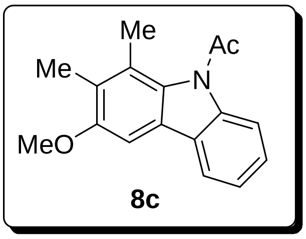
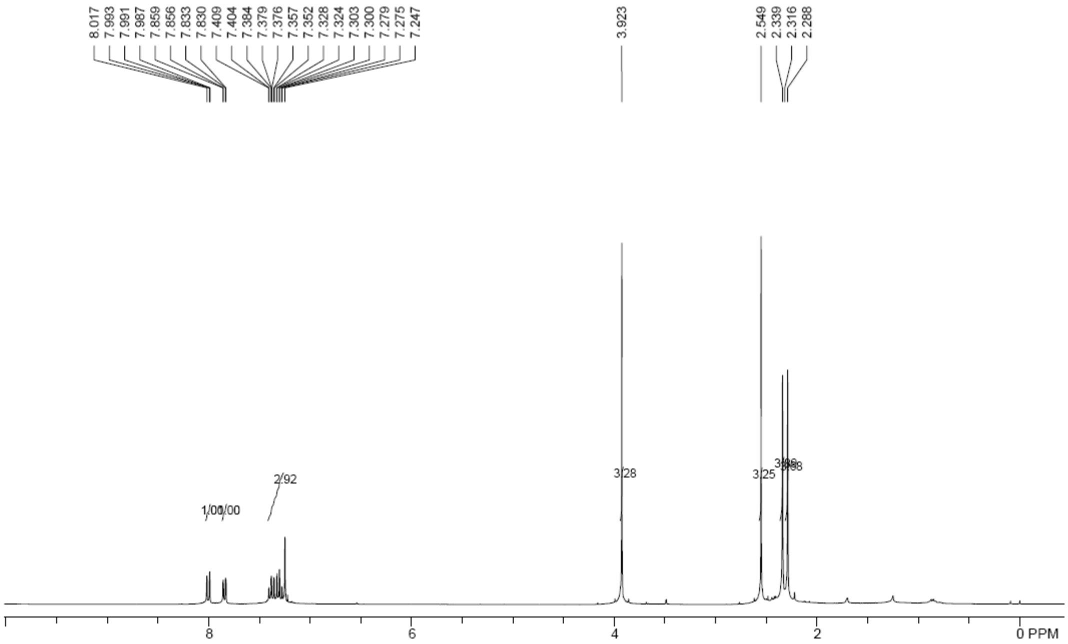
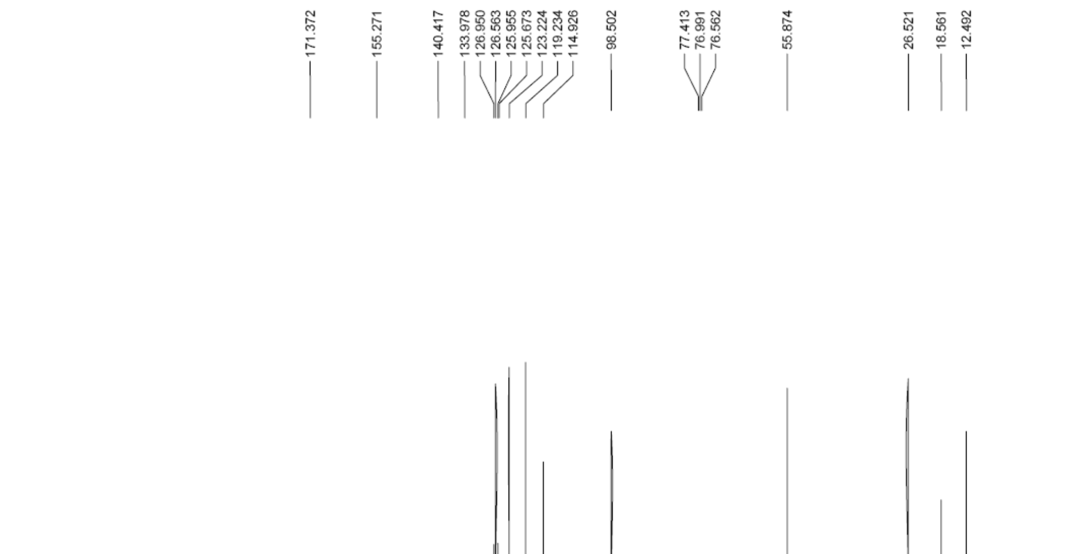
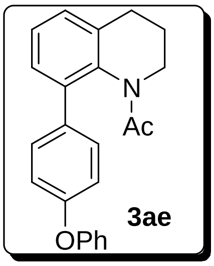
12.316
200
150
100
50
0
PPM

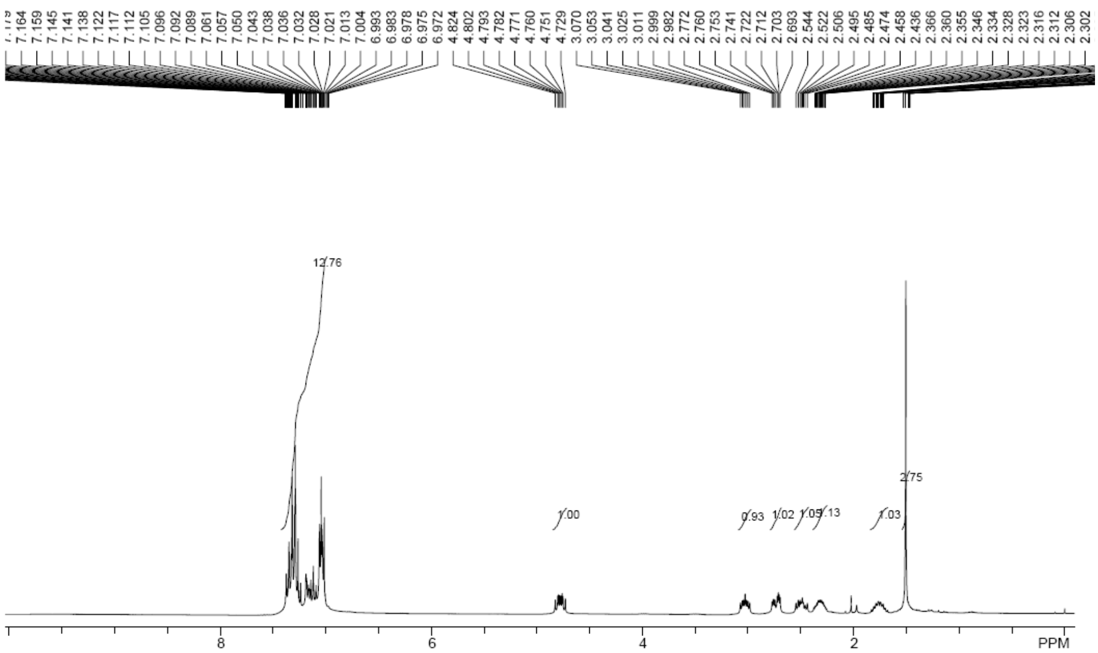

Me
Ac
Me
N
MeO
8c

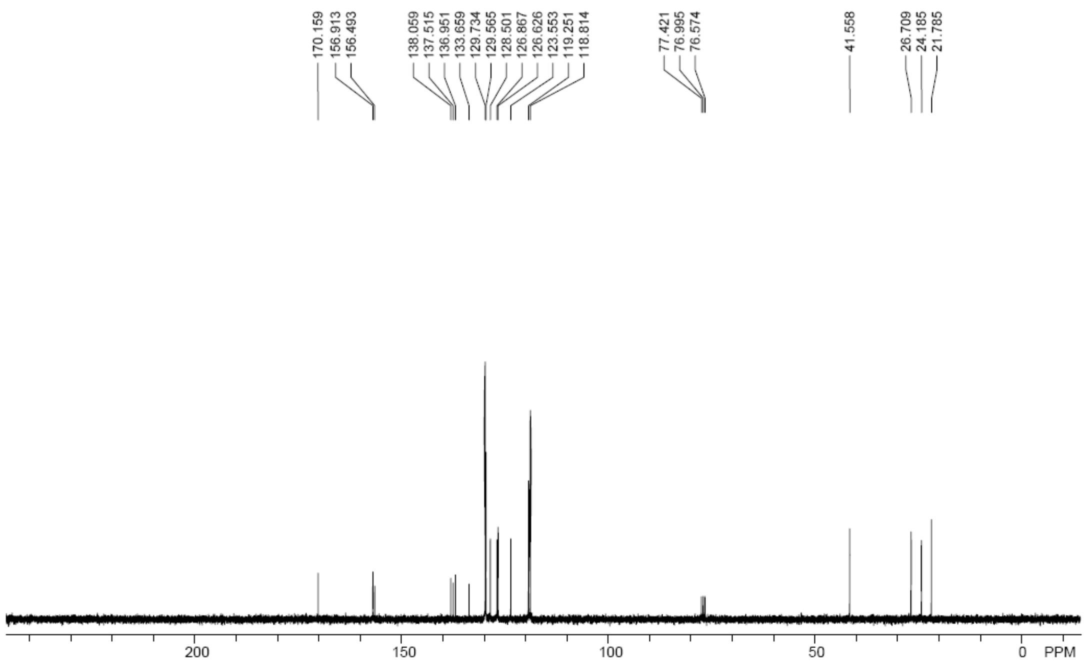


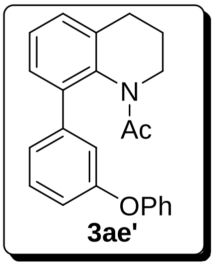


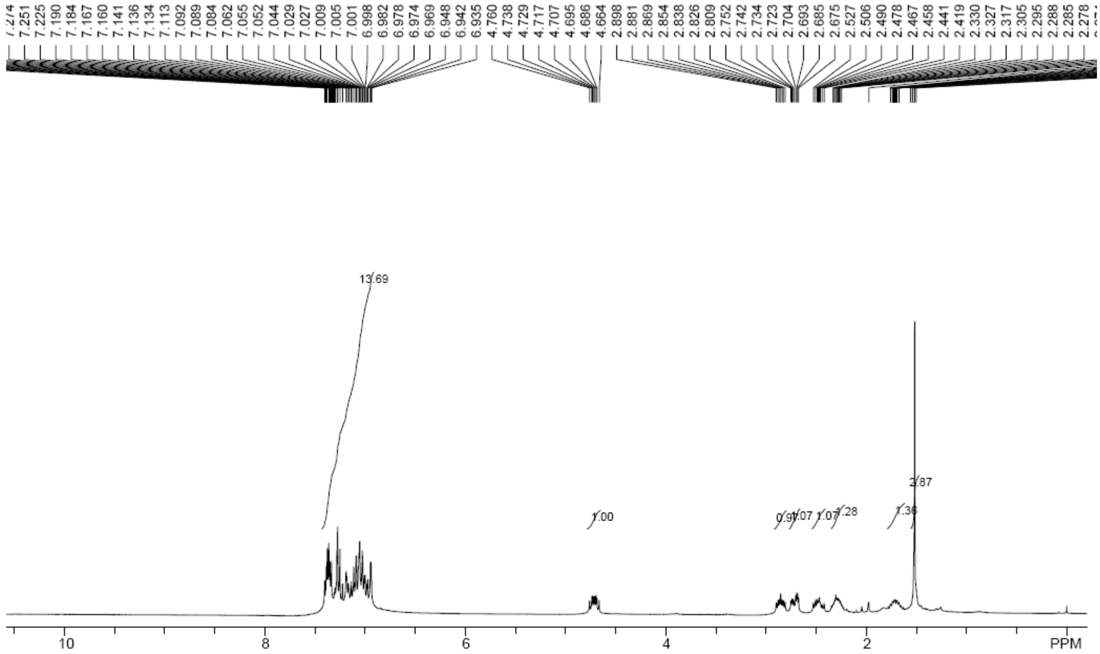
Note: the spectral data of standard products **3ae**, **3ae'**, **3af**, **3ag'** are given here, other isomers **3a**c, **3ad**, **3ad'**, **3ag**, **3ah**, **3ah'** see Ref 1.








N
Ac
OPh
3ae'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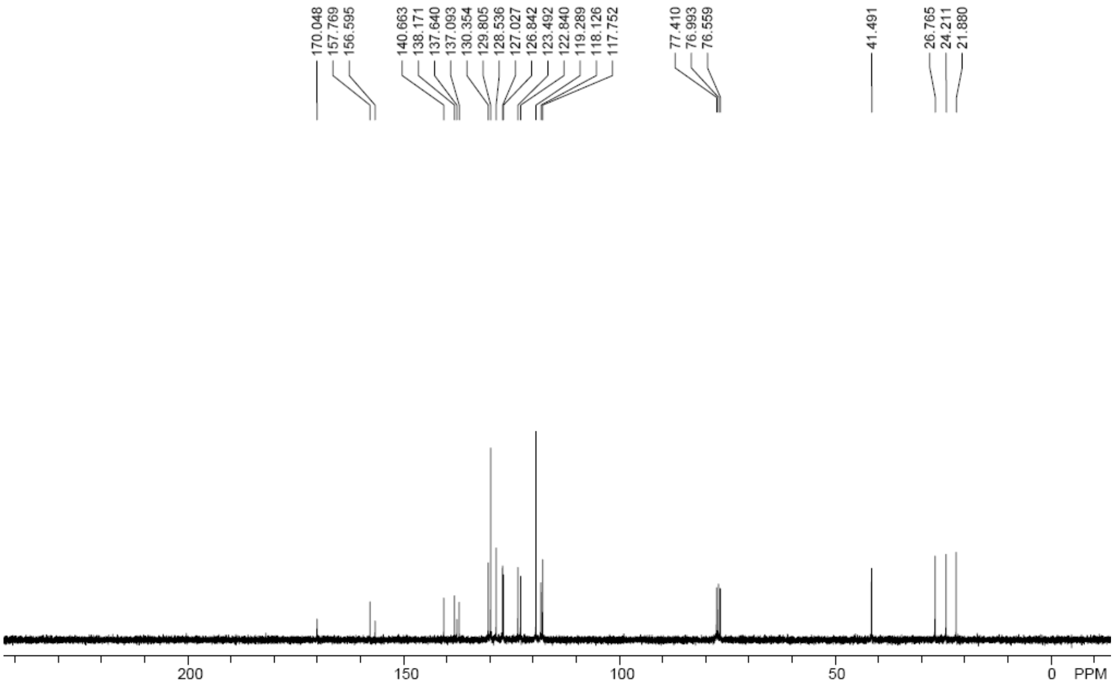
10
8
6
4
2
PPM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123.492
122.840
119.289
118.126
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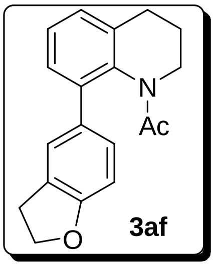
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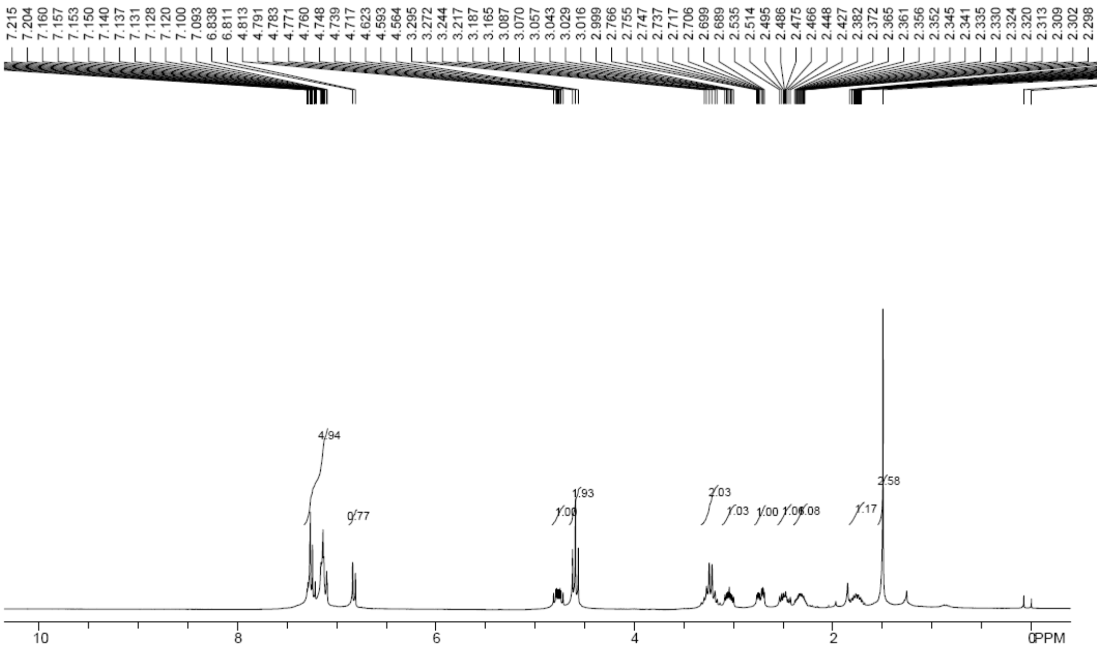
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6.838
6.811
4.813
4.791
4.783
4.771
4.760
4.748
4.739
4.717
4.623
4.593
4.564
3.295
3.272
3.244
3.217
3.187
3.165
3.087
3.070
3.057
3.043
3.029
3.016
2.999
2.766
2.755
2.747
2.737
2.717
2.706
2.699
2.689
2.535
2.514
2.495
2.486
2.475
2.466
2.448
2.427
2.382
2.372
2.365
2.361
2.356
2.352
2.345
2.341
2.335
2.330
2.324
2.320
2.313
2.309
2.302
2.298
4.94
0.77
1.00
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1.17
2.58
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8
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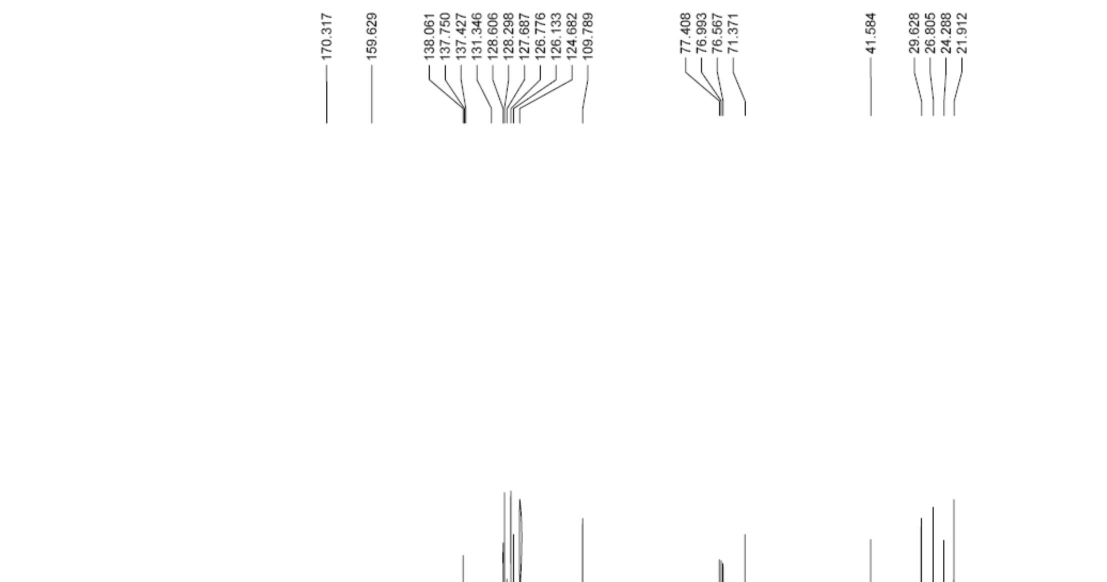
4
2
0PPM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77.408
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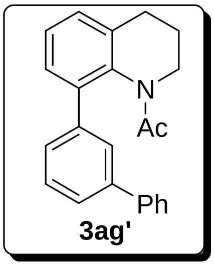
21.912
200
150
100
50
0
PPM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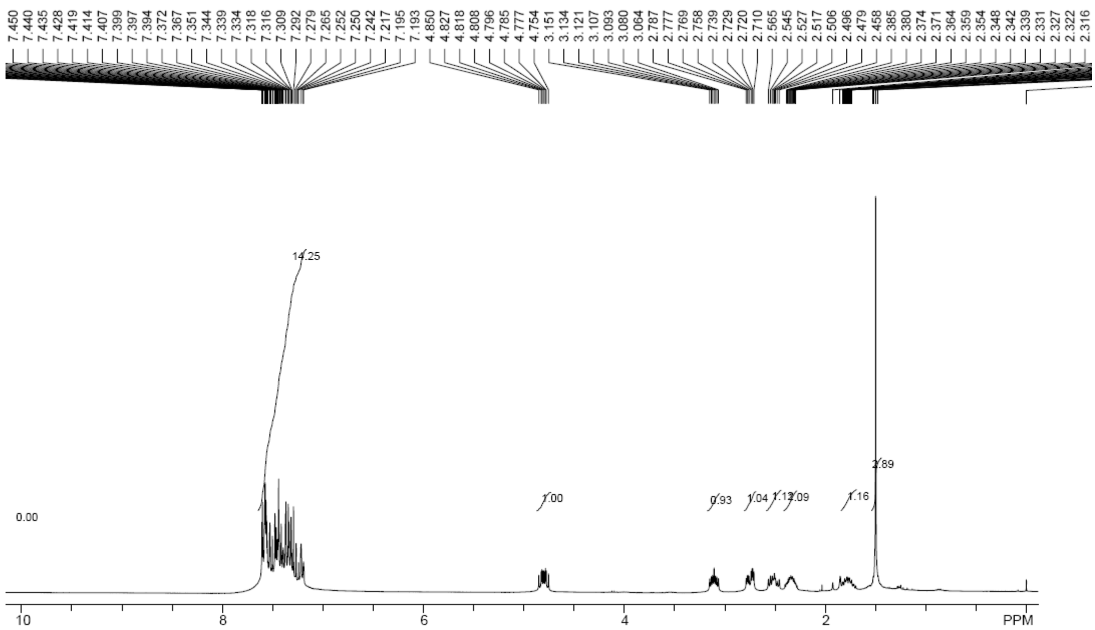
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14.25
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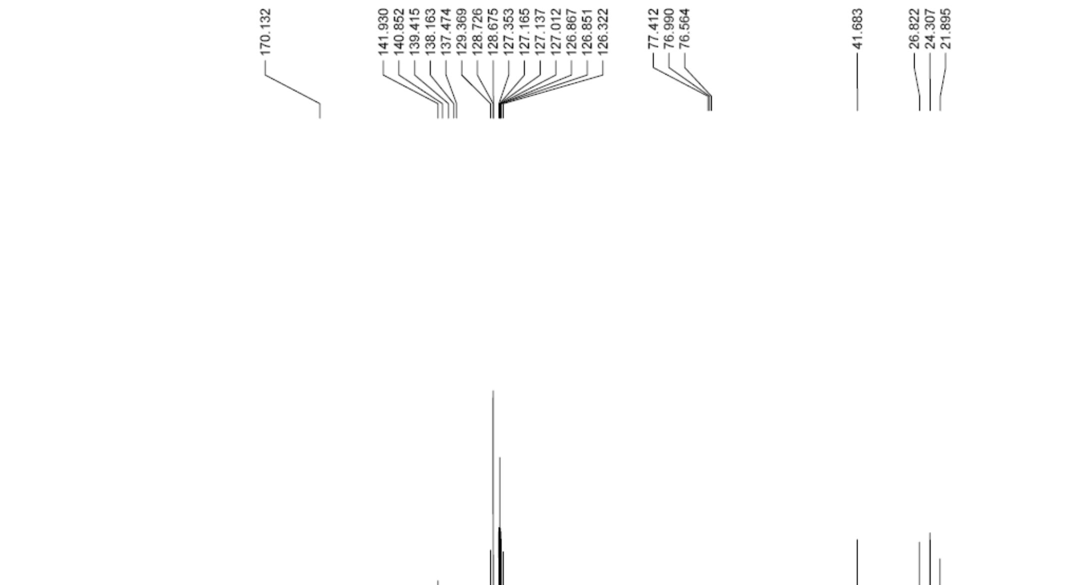
0.93
1.04
1.13
1.09
1.16
2.89
0.00
10
8
6
4
2
PPM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77.412 76.990 76.564
41.683
26.822 24.307 21.895
200
150
100
50
0
PPM

References:

(1) Shi, Z.; Li, B.; Wan, X.; Cheng, J.; Fang, Z.; Cao, B.; Qin, C.; Wang, Y. *Angew. Chem., Int. Ed.* **2007**, *46*, 5554.