Electronic Supplementary Material (ESI) for Organic & Biomolecular Chemistry.

I_2/CHP Mediated [1+1+1+1] Cyclization of Aromaticisocyanide with Amine to Construct 1,3-Diazetidine-2,4-diimine Derivatives

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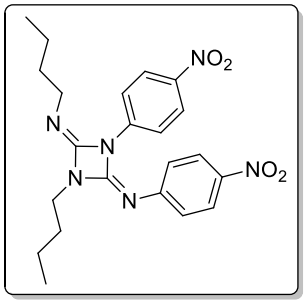
Experimental Section

General experimental: All chemicals were commercially available. Progresses of reactions were monitored by Thin Layer Chromatography while purification was performed using silicagel column chromatography. IR spectra were recorded on a Bruker spectrophotometer. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker Avance 300/75 or 400/100 MHz spectrometer using $CDCl_3$, as solvent and TMS as internal standard. High resolution mass spectra were obtained using GCT-TOF instrument with CI source. X-ray diffraction data were recorded on a Rigaku Mercury CCD area detector with graphite monochromated Mo Kα radiation.

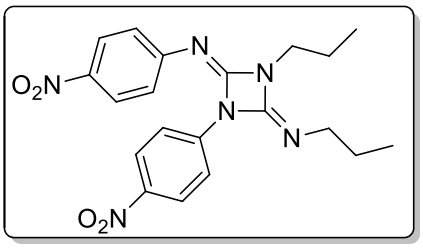
General procedures for reactions

I_2 (0.1 mmol), and amine (0.5 mmol) were added to test tube. MTBE (3.0 mL), Cumene Hydroperoxide (CHP) (1 mmol), isocyanides (0.6 mmol) were added via syringe. The test tube was closed and the reaction mixture was stirred at 55 ^{o}C. When the reactions were completed(monitored by TLC), the mixture was cooled to room temperature, washed with 10% $Na_2S_2O_3$ Solution(3*15 mL) and extracted with Ethyl acetate(3*15 mL). The combined organic layers were dried over Na_2SO_4. Removal of solvent followed by flash column chromatographic purification afforded products using petroleum and Ethyl acetate.



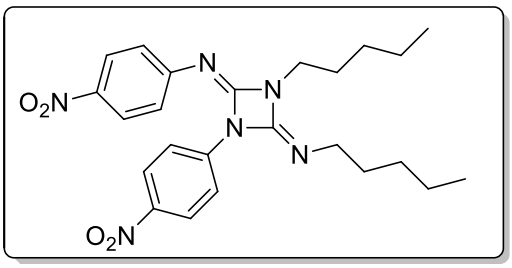
***N*-(1-butyl-4-(butylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3aa)**

Light yellow solid (0.0701 g, 64%), IR (ν cm^{-1}): 1694(-N=C-). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.27 - 8.19 (m, 4H, -ArH), 8.04 (d, J = 7.2 Hz, 2H, -ArH), 7.21 (d, J = 8.8 Hz, 2H, -ArH), 3.49 (t, J = 6.8 Hz, 2H, -CH_2), 3.10 - 3.01 (t, J = 8.0 Hz, 2H, -CH_2), 1.72 - 1.64 (m, 2H, -CH_2), 1.53 - 1.39 (m, 4H, -CH_2), 1.15 (dd, J = 14.9, 7.4 Hz, 2H, -CH_2), 0.99 (t, J = 7.3 Hz, 3H, -CH_3), 0.78 (t, J = 7.3 Hz, 3H, -CH_3). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.79, 147.07, 144.10, 142.83, 142.14, 141.88, 125.04, 124.92, 122.48, 116.98, 46.74, 45.66, 33.47, 31.23, 20.35, 19.61, 13.85, 13.55. HRMS (ESI) m/z: Found: 461.1905. Calcd for $C_{22}H_{26}N_6NaO_4^+$: $(M+Na)^+$ 461.1908.



4-nitro-*N*-(1-(4-nitrophenyl)-3-propyl-4-(propylimino)-1,3-diazetidin-2-ylidene)aniline (3ba)

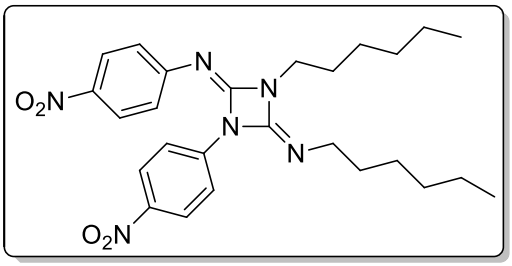
Light yellow solid (0.0729 g, 71%), IR (ν cm^{-1}): 1689(-N=C-). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.31 - 8.18 (m, 4H, -ArH), 8.05 (d, J = 7.2 Hz, 2H, -ArH), 7.20 (d, J = 8.8 Hz, 2H, -ArH), 3.46 (t, J = 6.8 Hz, 2H, -CH_2), 3.03 (t, J = 7.4 Hz, 2H, -CH_2), 1.77 - 1.67 (m, 2H, -CH_2), 1.54 - 1.42 (m, 2H, -CH_2), 1.03 (t, J = 7.3 Hz, 3H, -CH_3), 0.78 (t, J = 7.4 Hz, 3H, -CH_3). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.80, 147.07, 144.12, 142.85, 142.15, 141.94, 125.06, 124.93, 122.45, 116.99, 48.75, 47.37, 24.66, 22.69, 11.80, 10.90. HRMS (ESI) m/z: Found: 433.1594. Calcd for $C_{20}H_{22}N_6NaO_4^+$: $(M+Na)^+$ 433.1595.



4-nitro-*N*-(1-(4-nitrophenyl)-3-pentyl-4-(pentylimino)-1,3-diazetidin-2-ylidene)aniline (3ca)

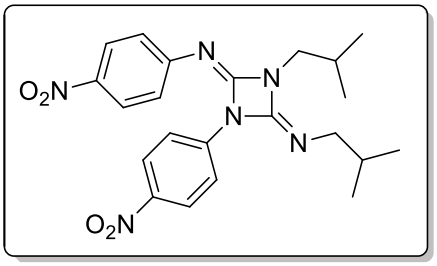
Light yellow solid (0.1038 g, 89%), IR (ν cm^{-1}): 1697(-N=C-). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.22 (dd, J = 12.3, 9.1 Hz, 4H, -ArH), 8.04 (d, J = 6.9 Hz, 2H, -ArH), 7.21 (d, J = 8.8 Hz, 2H, -ArH), 3.49 (t, J = 6.9 Hz, 2H, -CH_2), 3.14 - 2.99 (m, 2H, -CH_2), 1.75 - 1.66 (m, 2H, -CH_2), 1.50 - 1.35 (m, 6H, -CH_2), 1.22 - 1.05 (m, 4H, -CH_2), 0.95 (t, J = 7.0 Hz, 3H, -CH_3), 0.82 (t, J = 7.1 Hz, 3H, -CH_3). ^{13}C NMR (100

MHz, $CDCl_3$) δ 150.81, 147.01, 144.07, 142.76, 142.15, 141.81, 124.99, 124.85, 122.46, 116.94, 47.04, 45.89, 31.10, 29.41, 28.80, 28.42, 22.42, 22.04, 14.07, 13.75. HRMS (ESI) m/z: Found: 467.2405. Calcd for $C_{24}H_{31}N_6O_4^+$: $(M+H)^+$ 467.2401.



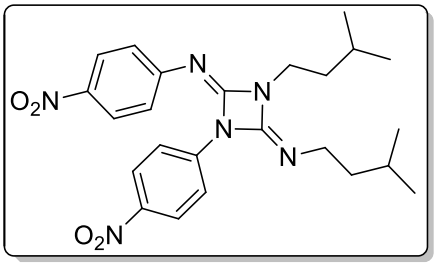
***N*-(-1-hexyl-4-(hexylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3da)**

Light yellow solid (0.0754 g, 61%), IR (ν cm^{-1}): 1680(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 8.23 (t, J = 8.5 Hz, 4H, -ArH), 8.04 (d, J = 7.6 Hz, 2H, -ArH), 7.20 (d, J = 8.8 Hz, 2H, -ArH), 3.48 (t, J = 6.9 Hz, 2H, $-CH_2$), 3.10 - 2.98 (m, 2H, $-CH_2$), 1.73 - 1.64 (m, 2H, $-CH_2$), 1.48 - 1.39 (m, 4H, $-CH_2$), 1.39 - 1.31 (m, 4H, $-CH_2$), 1.20 (dd, J = 13.4, 6.7 Hz, 2H, $-CH_2$), 1.14 - 1.05 (m, 4H, $-CH_2$), 0.92 (t, J = 6.7 Hz, 3H, $-CH_3$), 0.82 (t, J = 7.2 Hz, 3H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.80, 147.05, 144.11, 142.83, 142.14, 141.85, 125.04, 124.93, 122.47, 116.98, 47.08, 45.92, 31.55, 31.37, 31.08, 29.05, 26.90, 25.97, 22.63, 22.31, 14.07, 13.84. HRMS (ESI) m/z: Found: 517.2532. Calcd for $C_{26}H_{34}N_6NaO_4^+$: $(M+Na)^+$ 517.2534.



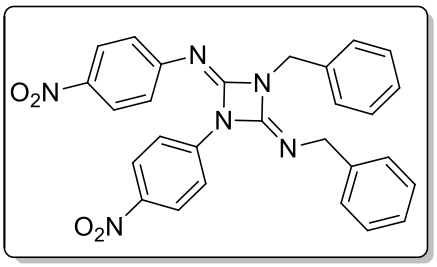
***N*-(1-isobutyl-4-(isobutylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3ea)**

Light yellow solid (0.0756 g, 69%), IR (ν cm^{-1}): 1715(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 8.22 (t, J = 8.4 Hz, 4H, -ArH), 8.06 (s, 2H, -ArH), 7.18 (d, J = 8.7 Hz, 2H, -ArH), 3.30 (d, J = 6.3 Hz, 2H, $-CH_2$), 2.92 (d, J = 7.2 Hz, 2H, $-CH_2$), 2.00 - 1.89 (m, 1H, -CH), 1.75 - 1.64 (m, 1H, -CH), 1.02 (d, J = 6.6 Hz, 6H, $-CH_3$), 0.79 (d, J = 6.7 Hz, 6H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.79, 147.25, 144.02, 142.82, 142.37, 141.81, 124.99, 124.90, 122.66, 116.97, 54.65, 52.99, 30.11, 29.71, 20.37, 19.86. HRMS (ESI) m/z: Found: 439.2089. Calcd for $C_{22}H_{27}N_6O_4^+$: $(M+H)^+$ 439.2088.



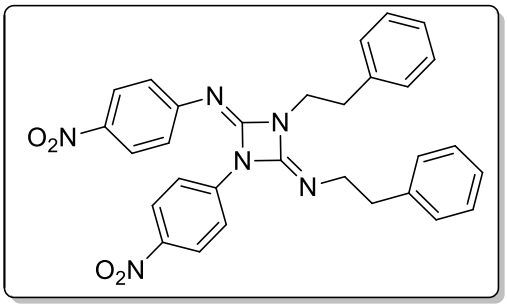
***N*-(-1-isopentyl-4-(isopentylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3fa)**

Light yellow solid (0.0781 g, 67%), IR (ν cm^{-1}): 1699(-N=C-). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.23 (t, J = 9.3 Hz, 4H, -ArH), 8.04 (d, J = 6.3 Hz, 2H, -ArH), 7.21 (d, J = 8.7 Hz, 2H, -ArH), 3.50 (t, J = 7.2 Hz, 2H, $-CH_2$), 3.14 – 3.00 (m, 2H, $-CH_2$), 1.78 (tt, J = 13.1, 6.5 Hz, 1H, -CH), 1.59 (dd, J = 14.0, 6.8 Hz, 2H, $-CH_2$), 1.40 – 1.28 (m, 3H, $-CH_2$ + -CH), 0.97 (d, J = 6.6 Hz, 6H, $-CH_3$), 0.71 (d, J = 5.9 Hz, 6H, $-CH_3$). ^{13}C NMR (101 MHz, $CDCl_3$) δ 150.77, 146.96, 144.12, 142.86, 142.06, 141.80, 125.01, 124.92, 122.50, 116.97, 45.22, 44.41, 40.30, 37.61, 25.91, 25.72, 22.51, 22.15. HRMS (ESI) m/z: Found: 489.2224. Calcd for $C_{24}H_{30}N_6NaO_4^+$: $(M+Na)^+$ 489.2221.



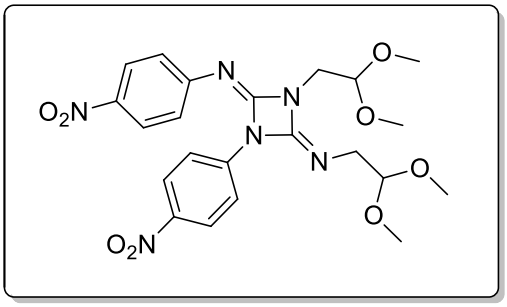
***N*-(-1-benzyl-4-(benzylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3ga)**

Light yellow solid (0.1089 g, 86%), IR (ν cm^{-1}): 1698(-N=C-). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.21 (d, J = 8.3 Hz, 2H, -ArH), 8.11 (d, J = 7.3 Hz, 4H, -ArH), 7.37 – 7.23 (m, 8H, -ArH), 7.09 (d, J = 7.2 Hz, 2H, -ArH), 6.99 (s, 2H, -ArH), 4.67 (s, 2H, $-CH_2$), 4.35 (s, 2H, $-CH_2$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.16, 146.28, 144.23, 143.07, 141.92, 138.73, 135.19, 129.19, 128.64, 128.48, 127.28, 126.96, 126.35, 124.98, 124.94, 122.58, 117.19, 50.60, 49.35. HRMS (ESI) m/z: Found: 529.1586. Calcd for $C_{28}H_{22}N_6NaO_4^+$: $(M+Na)^+$ 529.1595.



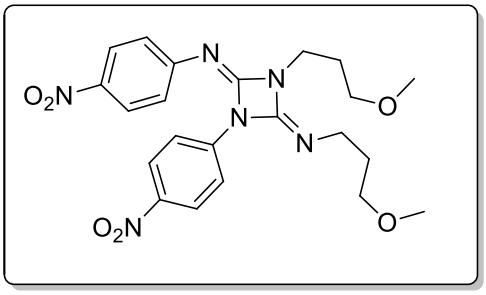
4-nitro-*N*-(1-(4-nitrophenyl)-3-phenethyl-4-(phenethylimino)-1,3-diazetidin-2-ylidene)aniline (3ha)

Light yellow solid (0.0962 g, 72%), IR (ν cm^{-1}): 1700(-N=C-). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.19 (d, J = 7.7 Hz, 4H, -ArH), 7.97 (s, 2H, -ArH), 7.27 (dd, J = 30.0, 8.1 Hz, 8H, -ArH), 7.03 (d, J = 6.9 Hz, 2H, -ArH), 6.86 (s, 2H, -ArH), 3.80 – 3.55 (m, 2H, $-CH_2$), 3.33 – 3.11 (m, 2H, $-CH_2$), 3.06- 2.88 (m, 2H, $-CH_2$), 2.70 – 2.46 (m, 2H, $-CH_2$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.46, 146.34, 144.14, 142.94, 141.90, 139.41, 136.55, 129.04, 128.89, 128.69, 128.54, 127.29, 126.58, 124.96, 124.92, 122.59, 117.05, 48.64, 47.10, 37.85, 35.73. HRMS (ESI) m/z: Found: 535.2087. Calcd for $C_{30}H_{27}N_6O_4^+$: $(M+H)^+$ 535.2088.



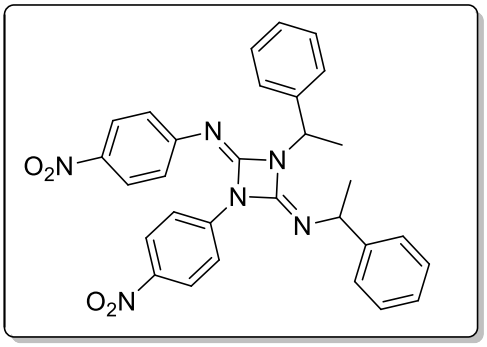
***N*-(-1-(2,2-dimethoxyethyl)-4-((2,2-dimethoxyethyl)imino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3ia)**

Light yellow solid (0.0942 g, 75%), IR (ν cm^{-1}): 1716(-N=C-). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.22 (t, J = 9.4 Hz, 4H, -ArH), 8.05 (d, J = 6.5 Hz, 2H, -ArH), 7.20 (d, J = 8.8 Hz, 2H, -ArH), 4.62 (t, J = 5.2 Hz, 1H, -CH), 4.31 – 4.21 (m, 1H, -CH), 3.67 (d, J = 5.1 Hz, 2H, $-CH_2$), 3.48 (s, 6H, $-CH_3$), 3.29 (s, 6H, $-CH_3$), 3.22 (d, J = 5.0 Hz, 2H, $-CH_2$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.47, 146.18, 144.13, 143.38, 142.94, 141.94, 124.97, 124.84, 122.63, 117.17, 104.41, 102.80, 55.54, 54.64, 49.49, 47.42. HRMS (ESI) m/z: Found: 525.1696. Calcd for $C_{22}H_{26}N_6NaO_8^+$: $(M+Na)^+$ 525.1704.



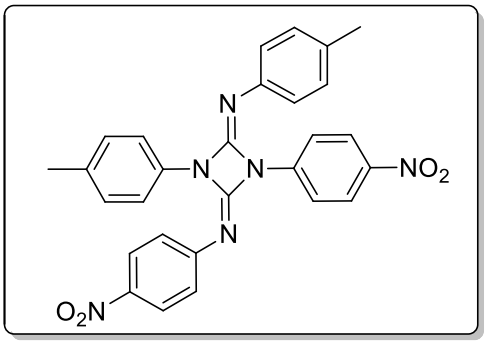
***N*-(-1-(3-methoxypropyl)-4-((3-methoxypropyl)imino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3ja)**

Light yellow solid (0.0671 g, 57%), IR (ν cm^{-1}): 1699(-N=C-). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.22 (t, J = 8.6 Hz, 4H, -ArH), 8.04 (d, J = 7.2 Hz, 2H, -ArH), 7.21 (d, J = 8.8 Hz, 2H, -ArH), 3.62 (t, J = 6.5 Hz, 2H, $-CH_2$), 3.55 (t, J = 5.9 Hz, 2H, $-CH_2$), 3.37 (s, 3H, $-CH_3$), 3.30 – 3.22 (m, 4H, $-CH_2$), 3.21 (s, 3H, $-CH_3$), 2.02 – 1.90 (m, 2H, $-CH_2$), 1.66 (dt, J = 12.4, 6.1 Hz, 2H, $-CH_2$). ^{13}C NMR (101 MHz, $CDCl_3$) δ 150.68, 146.92, 144.12, 142.80, 142.26, 142.15, 126.32, 125.01, 124.90, 122.52, 116.94, 113.32, 69.82, 68.99, 58.67, 58.61, 43.51, 43.14, 31.30, 29.29. HRMS (ESI) m/z: Found: 493.1804. Calcd for $C_{22}H_{26}N_6NaO_6^+$: $(M+Na)^+$ 493.1806.



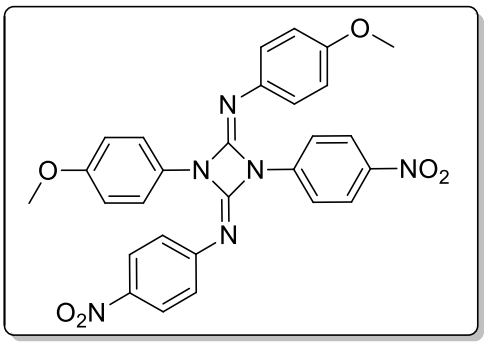
4-nitro-*N*-(1-(4-nitrophenyl)-3-(1-phenylethyl)-4-((1-phenylethyl)imino)-1,3-diazetidin-2-ylidene)aniline (3na)

Light yellow solid (0.0722 g, 54%), IR (ν cm^{-1}): 1688(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 8.20 (d, J = 30.3 Hz, 4H, -ArH), 8.06 (d, J = 8.6 Hz, 2H, -ArH), 7.35 – 7.23 (m, 8H, -ArH), 7.04 (s, 4H, -ArH), 4.72 – 4.60 (m, 1H, -CH), 4.48 – 4.26 (m, 1H, -CH), 1.60 (d, J = 4.2 Hz, 3H, $-CH_3$), 1.46 (d, J = 6.0 Hz, 3H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 150.68, 146.18, 144.53, 143.95, 142.05, 140.53, 128.91, 128.49, 128.08, 127.11, 126.13, 125.75, 124.95, 124.85, 122.09, 117.63, 57.33, 53.51, 26.38, 17.47. HRMS (ESI) m/z: Found: 535.2085. Calcd for $C_{30}H_{27}N_6O_4^+$: $(M+H)^+$ 535.2088.



4-methyl-*N*-(1-(4-nitrophenyl)-4-((4-nitrophenyl)imino)-3-(*p*-tolyl)-1,3-diazetidin-2-ylidene)aniline (3oa)

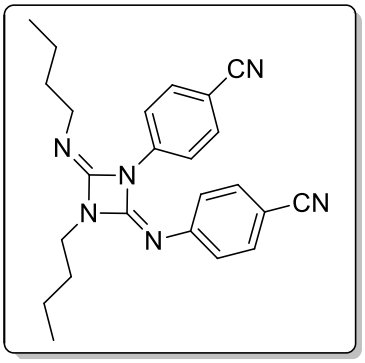
Light yellow solid (0.1122 g, 84%), IR (ν cm^{-1}): 1680(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 8.24 (d, J = 8.0 Hz, 2H, -ArH), 8.08 (s, 2H, -ArH), 7.92 (d, J = 8.7 Hz, 2H, -ArH), 6.93 (d, J = 8.7 Hz, 2H, -ArH), 6.85 (d, J = 7.9 Hz, 2H, -ArH), 6.71 (d, J = 7.8 Hz, 4H, -ArH), 6.59 (s, 2H, -ArH), 2.19 (s, 3H, $-CH_3$), 2.14 (s, 3H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 149.67, 144.81, 143.94, 143.40, 141.50, 141.40, 139.76, 134.16, 131.62, 129.65, 129.27, 129.14, 125.37, 124.97, 124.34, 122.62, 121.84, 117.83, 20.99, 20.80. HRMS (ESI) m/z: Found: 529.1590. Calcd for $C_{28}H_{22}N_6NaO_4^+$: $(M+Na)^+$ 529.1595.



4-methoxy-*N*-(1-(4-methoxyphenyl)-3-(4-nitrophenyl)-4-((4-nitrophenyl)imino)-1,3-diazetidin-2-ylidene)aniline (3pa)

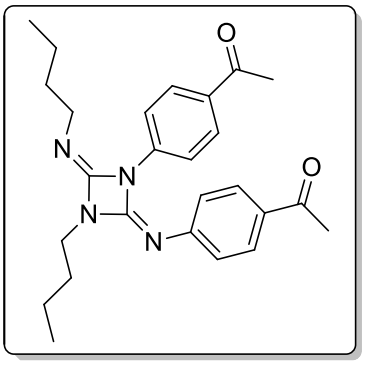
Light yellow solid (0.1010 g, 75%), IR (ν cm^{-1}): 1692(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 8.26 (d, J = 7.6 Hz, 2H, -ArH), 8.13 (s, 2H, -ArH), 7.93 (d, J = 8.4 Hz, 2H, -ArH), 6.93 (d, J = 8.3 Hz, 2H, -ArH), 6.77 (d, J = 8.2 Hz, 2H, -ArH), 6.64 (s, 2H, -ArH), 6.59 (d, J = 8.2 Hz, 2H，-ArH), 6.43 (d, J = 6.8 Hz, 2H, -ArH), 3.68 (s, 3H, $-CH_3$), 3.63 (s, 3H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 158.91, 156.66, 149.73, 145.16, 143.91, 143.26, 141.63, 141.55, 135.29, 127.28, 126.96, 125.00, 124.30, 123.15, 122.57, 117.45, 113.91, 113.82, 55.42, 55.39. HRMS (ESI) m/z: Found: 561.1491. Calcd for

$C_{28}H_{22}N_6NaO_6^+$: $(M+Na)^+$ 561.1493.



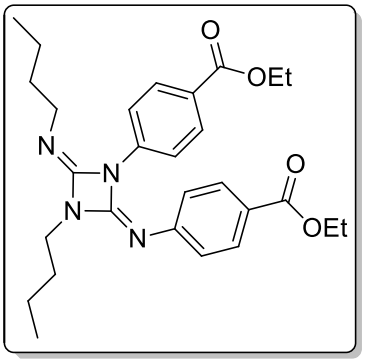
4-(3-butyl-2-(butylimino)-4-((4-cyanophenyl)imino)-1,3-diazetidin-1-yl)benzonitrile (3ab)

Light yellow solid (0.0807 g, 81%), IR (ν cm^{-1}): 2226(-CN), 1686(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 7.98 (s, 2H, -ArH), 7.61 (t, J = 7.9 Hz, 4H, -ArH), 7.15 (d, J = 8.2 Hz, 2H, -ArH), 3.46 (t, J = 6.7 Hz, 2H, $-CH_2$), 3.01 (t, J = 7.3 Hz, 2H, $-CH_2$), 1.66 (dd, J = 14.2, 7.1 Hz, 2H, $-CH_2$), 1.49 – 1.36 (m, 4H, $-CH_2$), 1.13 (dq, J = 14.5, 7.2 Hz, 2H, $-CH_2$), 0.97 (t, J = 7.3 Hz, 3H, $-CH_3$), 0.77 (t, J = 7.3 Hz, 3H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 148.91, 147.04, 142.27, 140.49, 133.25, 133.06, 122.83, 119.04, 118.93, 117.43, 107.39, 106.26, 46.71, 45.59, 33.50, 31.13, 20.34, 19.59, 13.84, 13.52. HRMS (ESI) m/z: Found: 399.2291. Calcd for $C_{24}H_{27}N_6^+$: $(M+H)^+$ 399.2292.



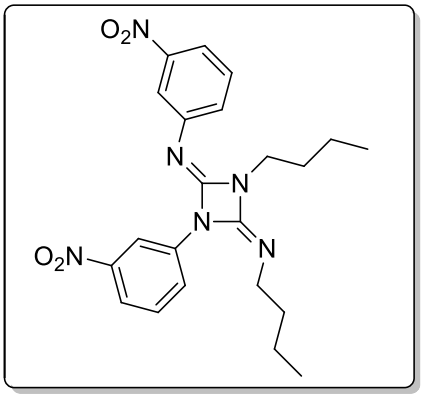
1-(4-(1-(4-acetylphenyl)-3-butyl-4-(butylimino)-1,3-diazetidin-2-ylidene)amino)phenyl)ethan one (3ac)

Light yellow solid (0.0325 g, 30%), IR (ν cm^{-1}): 1739(-C=O), 1677(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 7.94 (s, 6H, -ArH), 7.14 (d, J = 7.8 Hz, 2H, -ArH), 3.47 (t, J = 6.6 Hz, 2H, $-CH_2$), 3.01 (s, 2H, $-CH_2$), 2.60 (s, 3H, $-CH_3$), 2.57 (s, 3H, $-CH_3$), 1.67 (dd, J = 14.1, 7.1 Hz, 2H, $-CH_2$), 1.49 – 1.39 (m, 4H, $-CH_2$), 1.13 (dd, J = 13.6, 6.9 Hz, 2H, $-CH_2$), 0.97 (t, J = 7.1 Hz, 3H, $-CH_3$), 0.76 (t, J = 7.1 Hz, 3H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 196.62, 196.50, 148.99, 146.42, 142.65, 140.57, 132.47, 131.45, 129.19, 129.02, 121.60, 116.36, 46.23, 45.02, 33.09, 30.61, 26.00, 25.95, 19.87, 19.14, 13.38, 13.08. HRMS (ESI) m/z: Found: 455.2418. Calcd for $C_{26}H_{32}N_4NaO_2^+$: $(M+Na)^+$ 455.2417.



Ethyl 4-(3-butyl-2-(butylimino)-4-((4-(ethoxycarbonyl)phenyl)imino)-1,3-diazetidin-1-yl) benzoate (3ad)

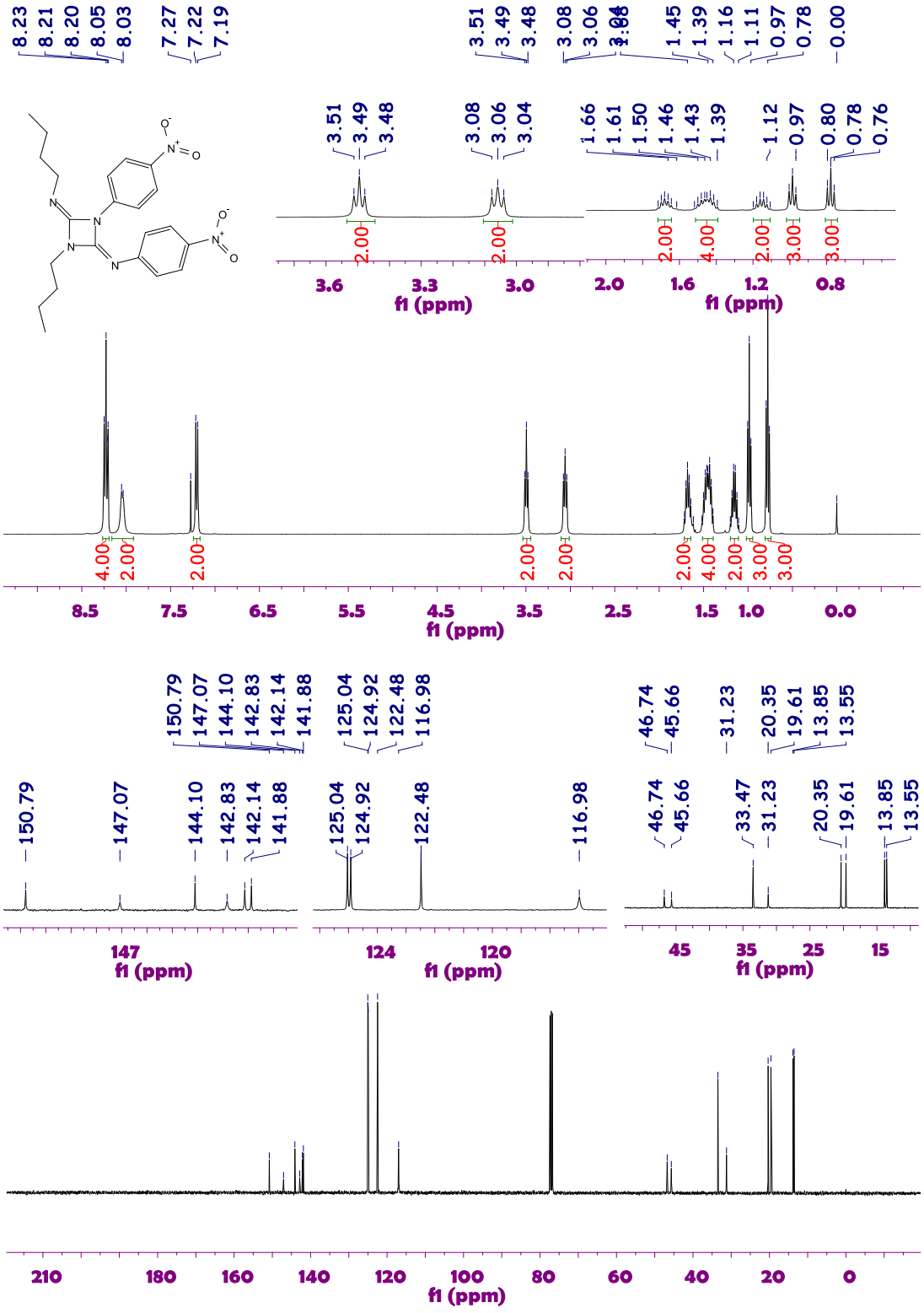
Light yellow solid (0.0603 g, 49%), IR (ν cm^{-1}): 1711(-C=O), 1681(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 8.07 – 7.92 (m, 6H, -ArH), 7.12 (d, J = 8.1 Hz, 2H, -ArH), 4.39 – 4.34 (m, 4H, $-CH_2$), 3.46 (t, J = 6.7 Hz, 2H, $-CH_2$), 3.00 (s, 2H, $-CH_2$), 1.69 – 1.62 (m, 2H, $-CH_2$), 1.49 – 1.44 (m, 2H, $-CH_2$), 1.42 – 1.37 (m, 8H， $-CH_2$ + $-CH_3$), 1.13 (dd, J = 13.2, 6.0 Hz, 2H, $-CH_2$), 0.97 (t, J = 7.9 Hz, 3H, $-CH_3$), 0.76 (t, J = 7.2 Hz, 3H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 165.88, 165.80, 148.74, 146.43, 142.86, 140.38, 130.23, 130.04, 125.42, 124.41, 121.41, 116.24, 60.38, 60.30, 46.19, 44.99, 33.10, 30.57, 19.86, 19.14, 13.87, 13.86, 13.37, 13.09. HRMS (ESI) m/z: Found: 515.2632. Calcd for $C_{28}H_{36}N_4NaO_4^+$: $(M+Na)^+$ 515.2629.



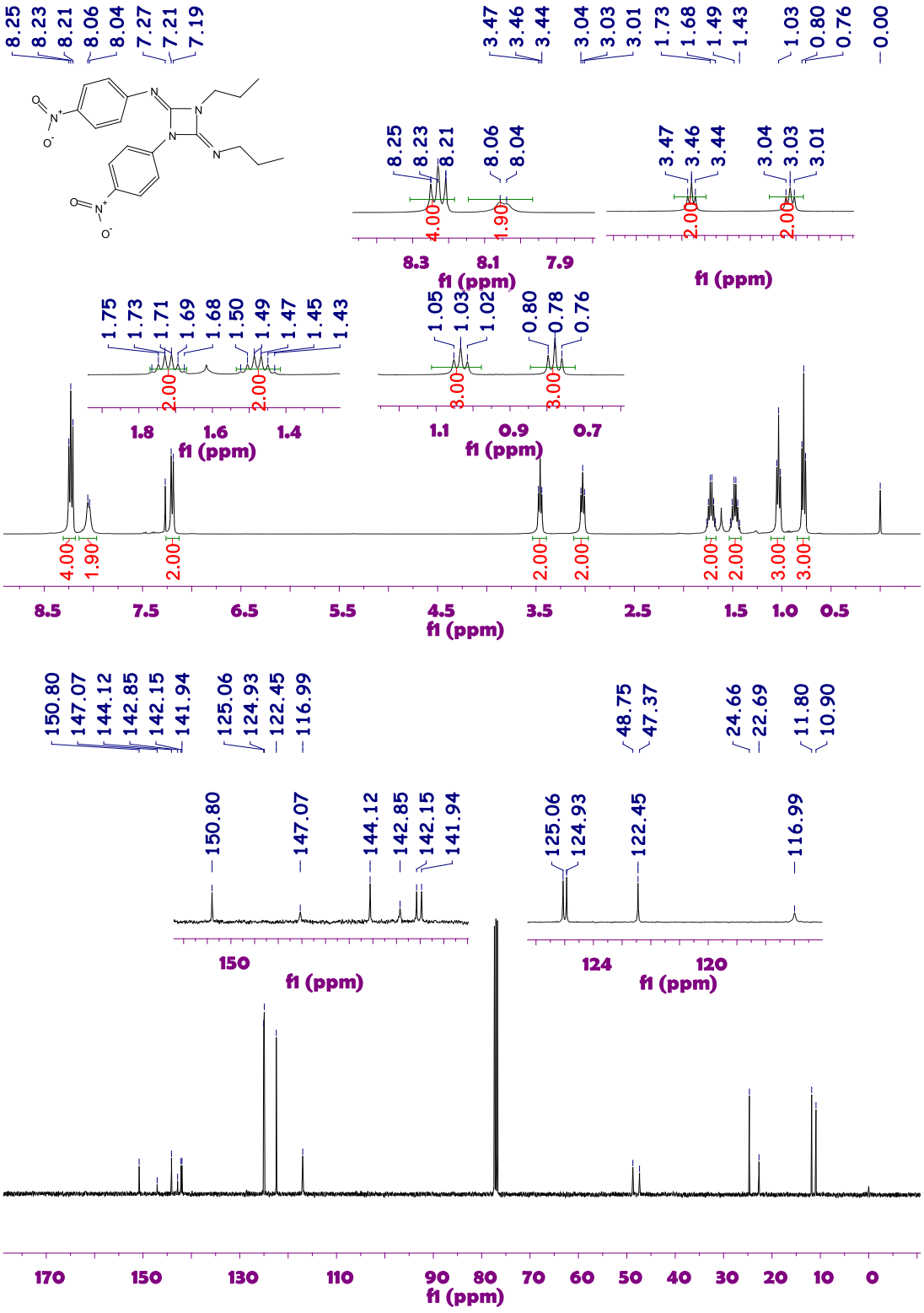
***N*-(1-butyl-4-(butylimino)-3-(3-nitrophenyl)-1,3-diazetidin-2-ylidene)-3-nitroaniline (3af)**

Light yellow solid (0.0669 g, 61%), IR (ν cm^{-1}): 1680(-N=C-). ^1H NMR (400 MHz, $CDCl_3$) δ 8.79 (s, 1H, -ArH), 8.24 (s, 1H, -ArH), 8.07 – 7.82 (m, 3H, -ArH), 7.59 – 7.37 (m, 3H，-ArH), 3.62 – 3.35 (m, 2H, $-CH_2$), 3.23 – 2.83 (m, 2H, $-CH_2$), 1.78 – 1.63 (m, 2H, $-CH_2$), 1.56 – 1.34 (m, 4H, $-CH_2$), 1.24 – 1.08 (m, 2H, $-CH_2$), 0.99 (t, J = 6.6 Hz, 3H, $-CH_3$), 0.87 – 0.67 (m, 3H, $-CH_3$). ^{13}C NMR (100 MHz, $CDCl_3$) δ 148.26, 147.97, 147.27, 145.40, 142.02, 137.34, 129.35, 129.17, 127.98, 122.56, 118.32, 117.52, 117.49, 116.41, 46.21, 45.08, 33.01, 30.62, 19.85, 19.13, 13.36, 13.02. HRMS (ESI) m/z: Found: 439.2095. Calcd for $C_{22}H_{27}N_6O_4^+$: $(M+H)^+$ 439.2088.

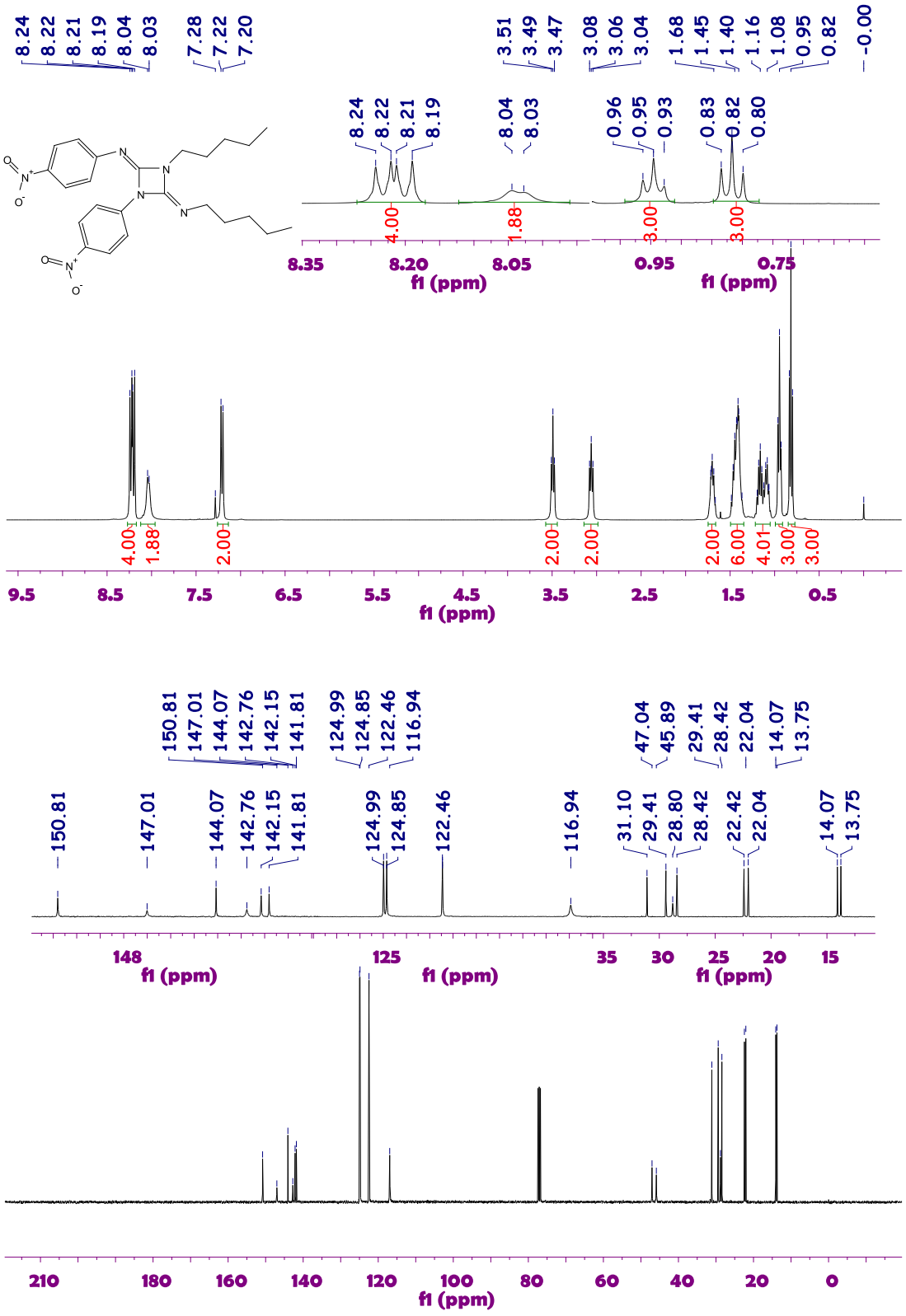
***N*-(1-butyl-4-(butylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3aa)**



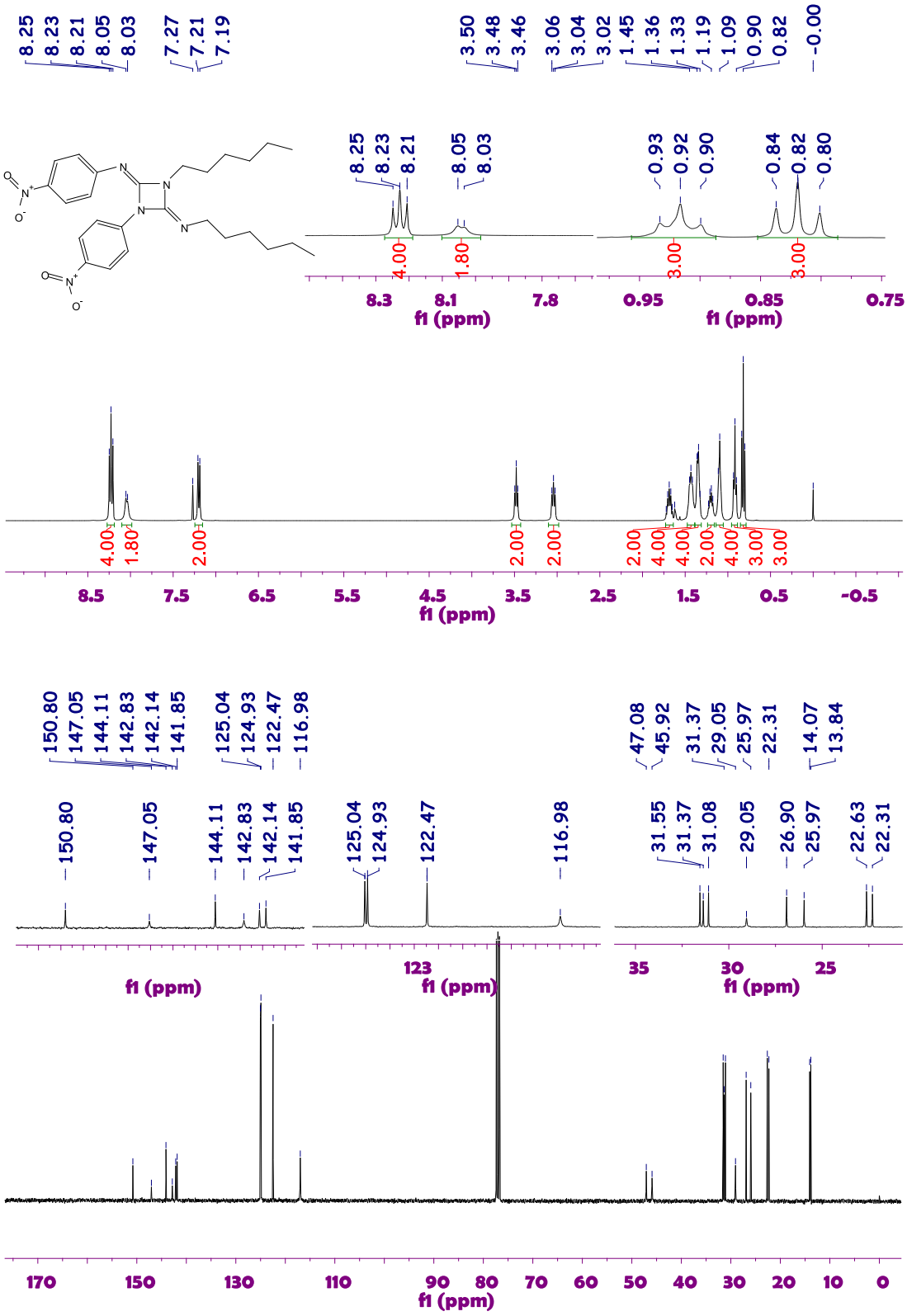
4-nitro-*N*-(1-(4-nitrophenyl)-3-propyl-4-(propylimino)-1,3-diazetidin-2-ylidene)aniline (3ba)



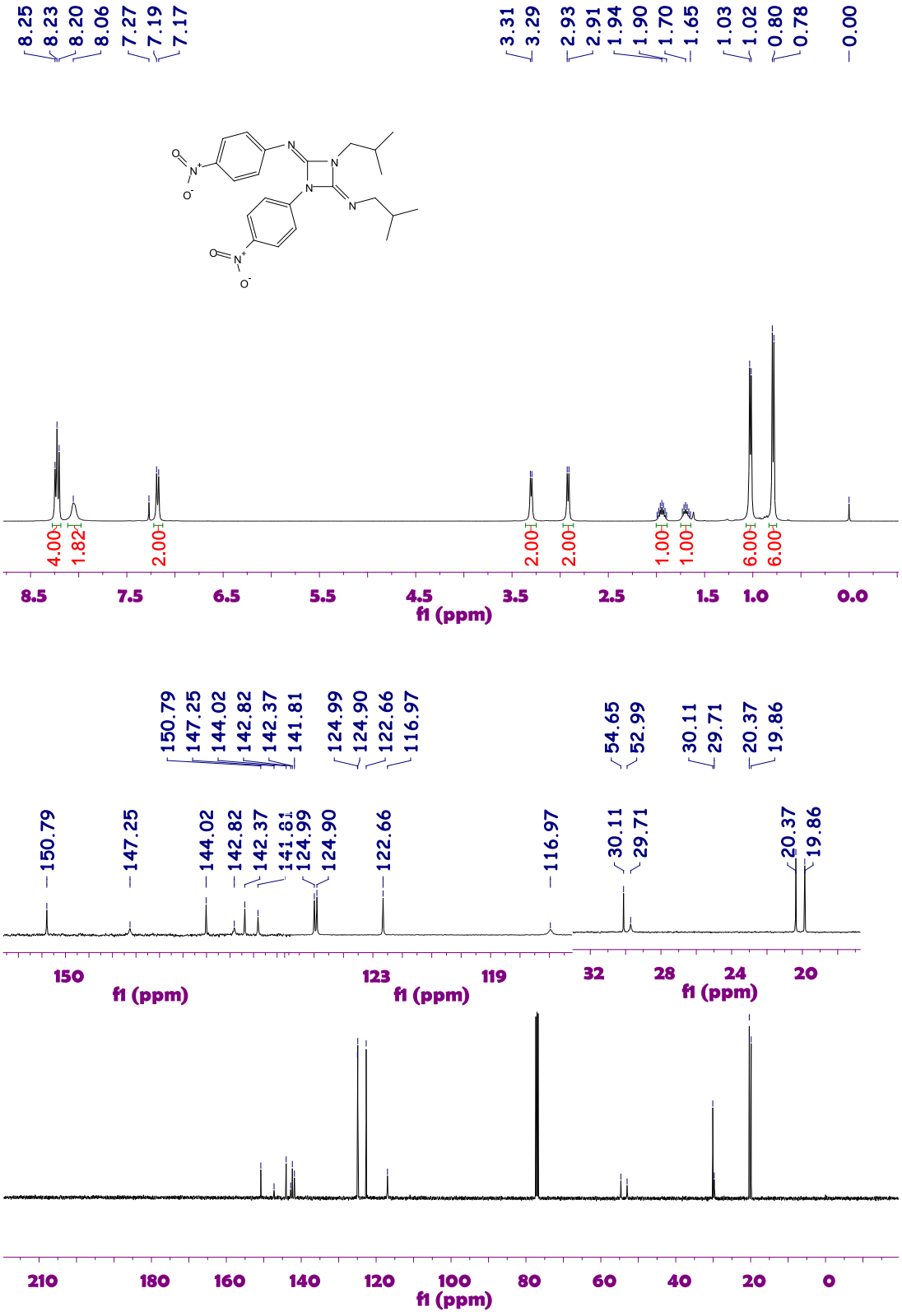
4-nitro-*N*-(1-(4-nitrophenyl)-3-pentyl-4-(pentylimino)-1,3-diazetidin-2-ylidene)aniline (3ca)



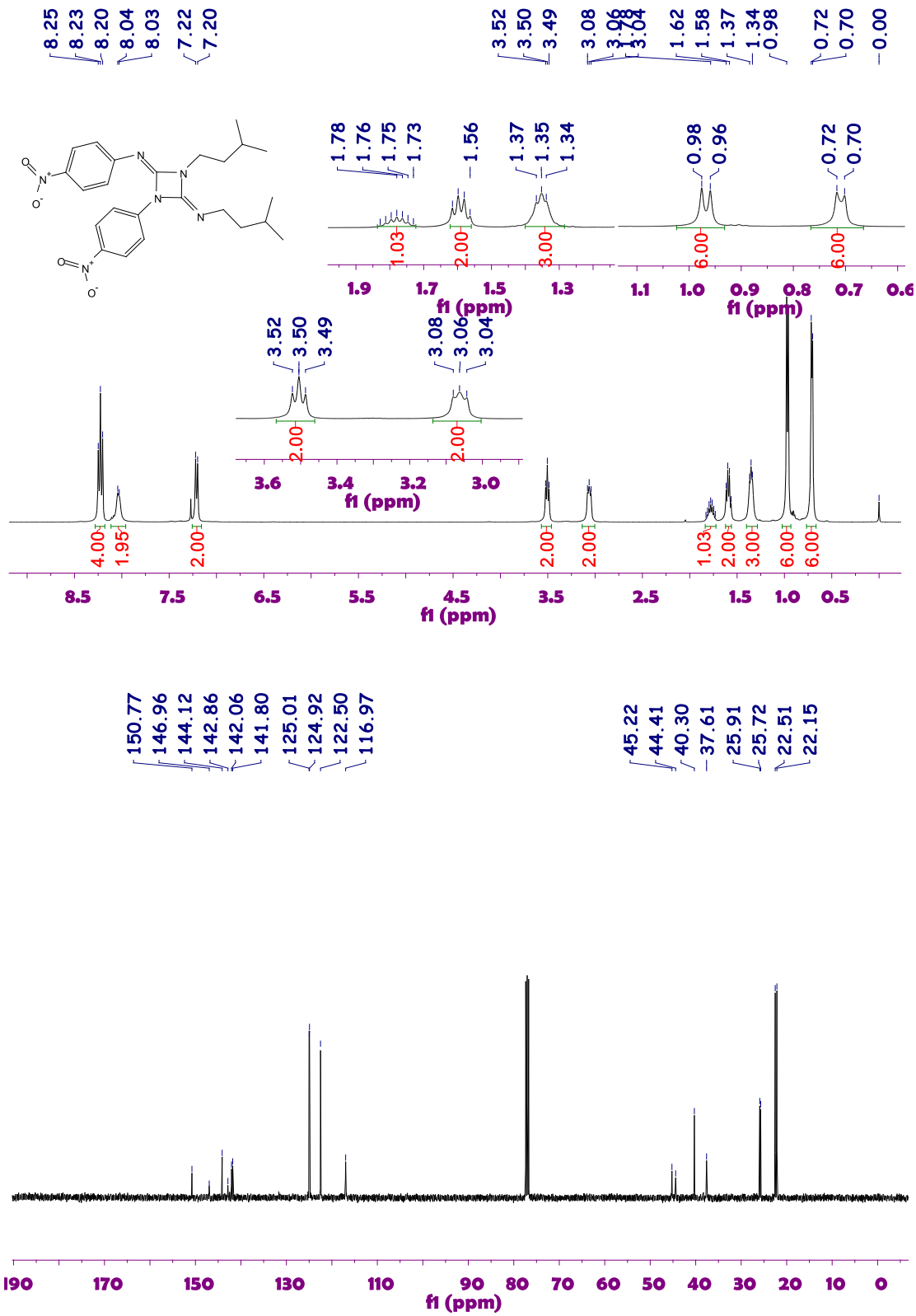
***N*-(-1-hexyl-4-(hexylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3da)**



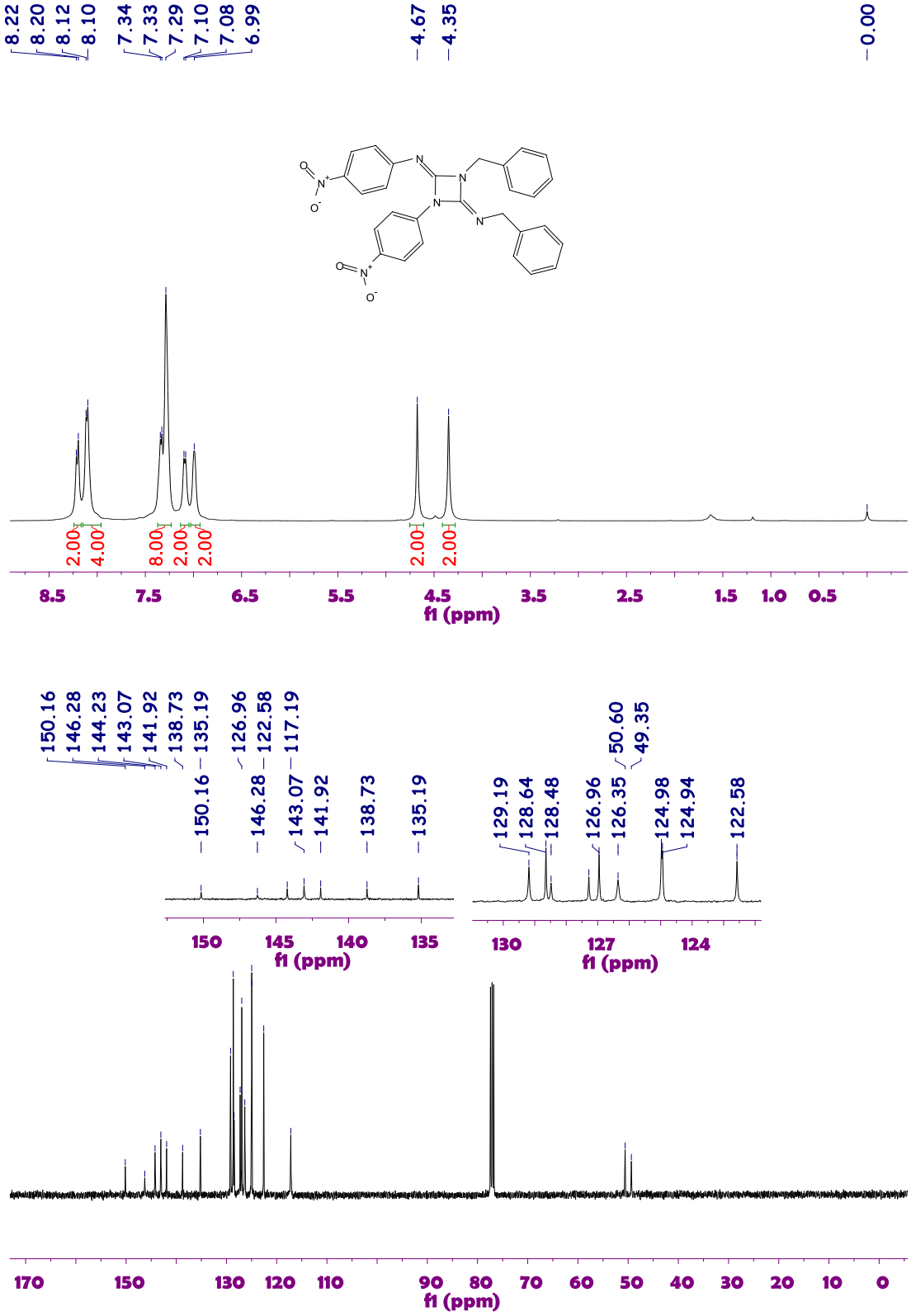
***N*-(1-isobutyl-4-(isobutylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3ea)**



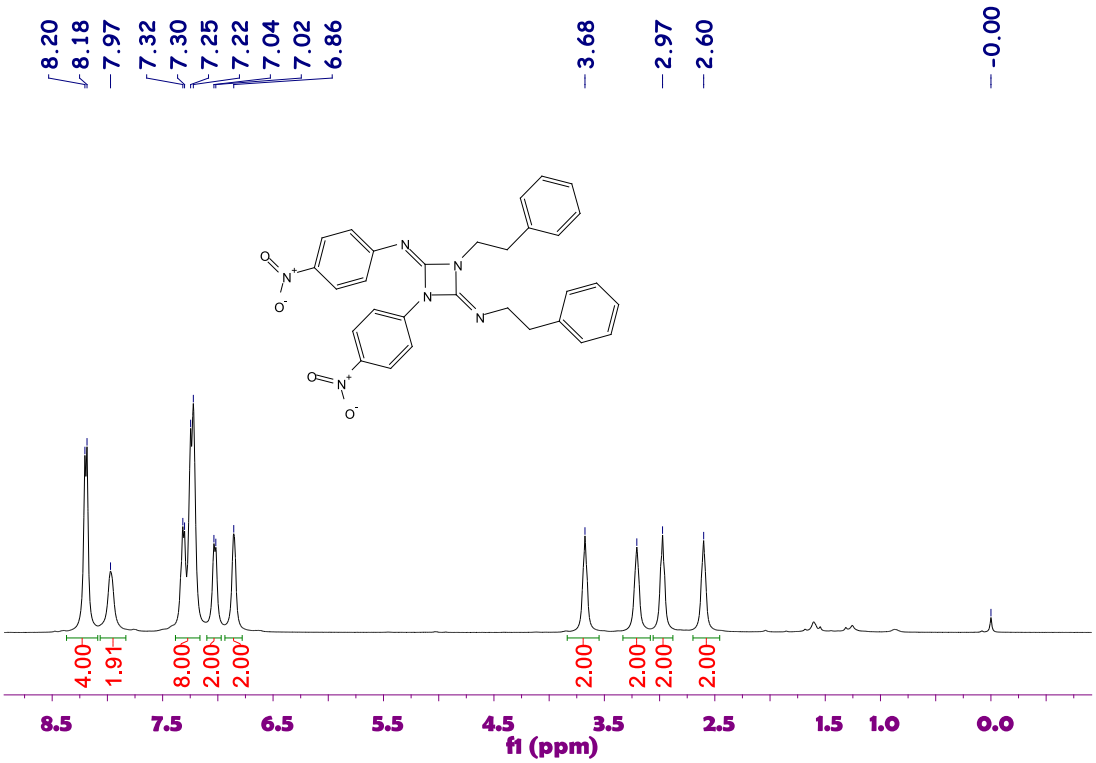
***N*-(-1-isopentyl-4-(isopentylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3fa)**

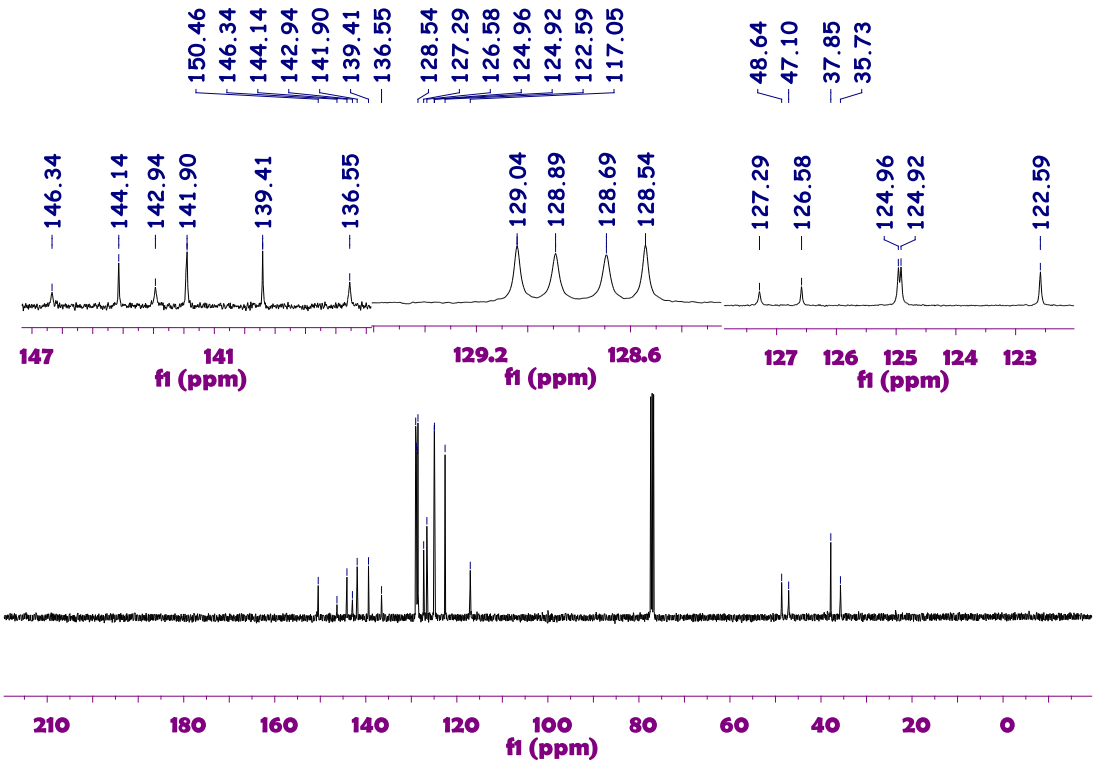


***N*-(-1-benzyl-4-(benzylimino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene)-4-nitroaniline (3ga)**

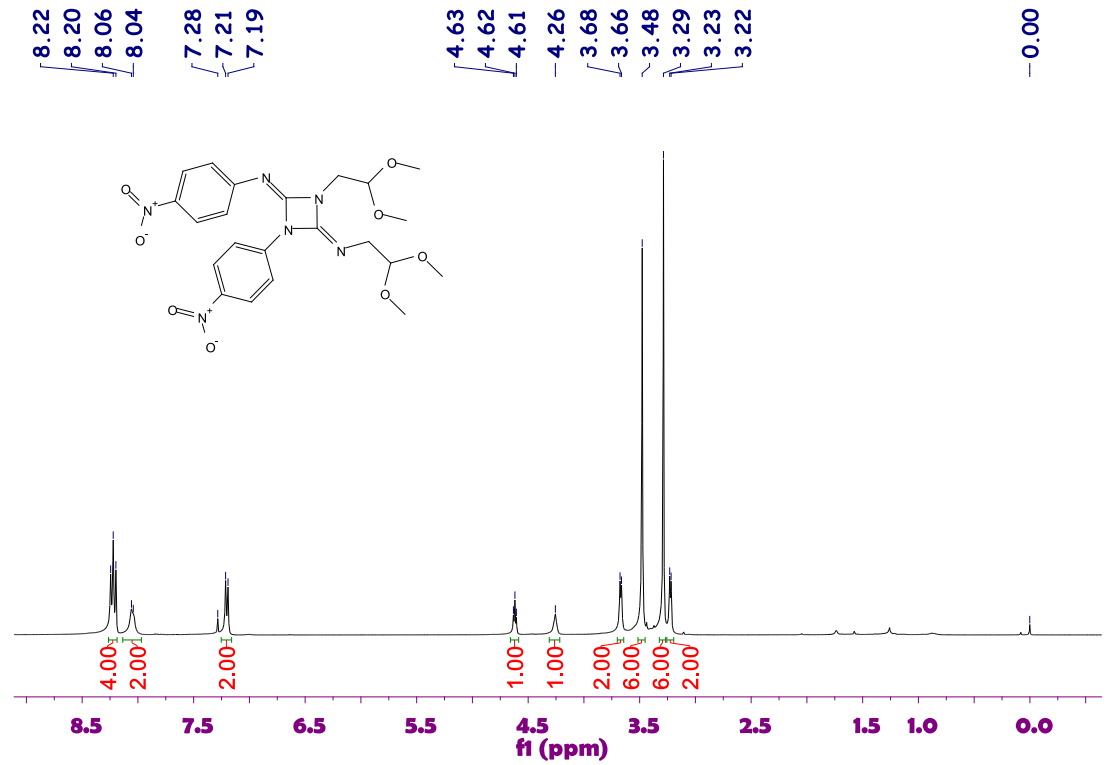


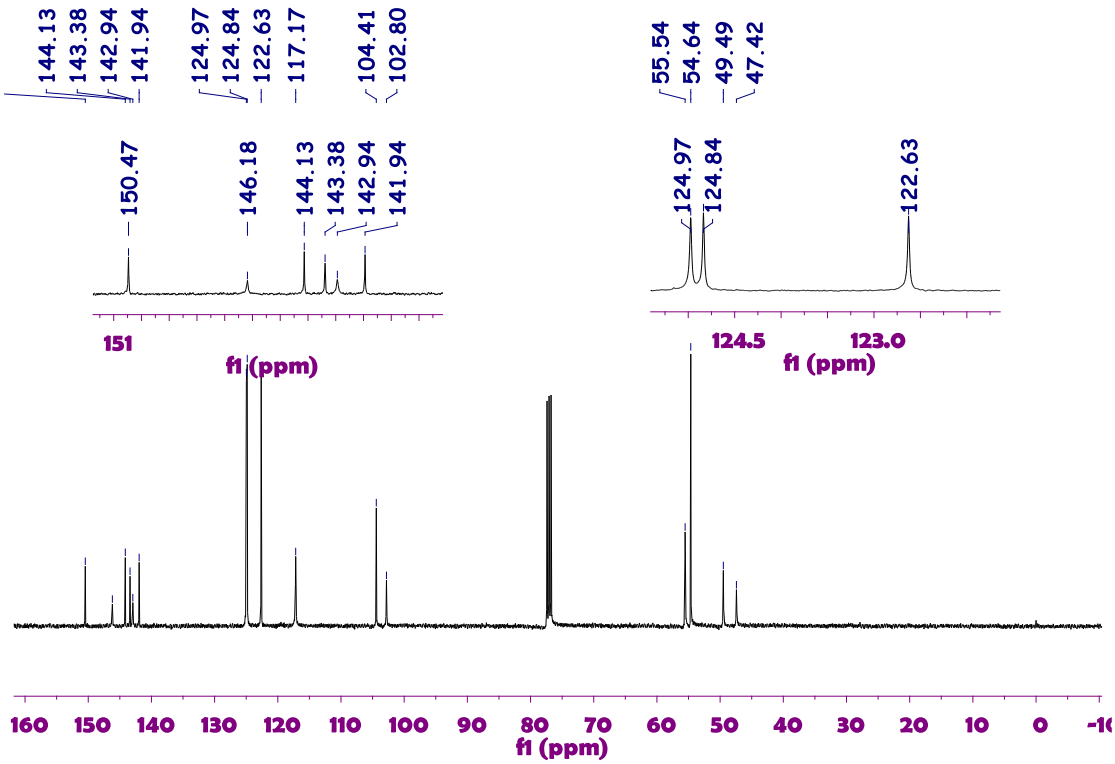
4-nitro-*N*-(1-(4-nitrophenyl)-3-phenethyl-4-(phenethylimino)-1,3-diazetidin-2-ylidene)aniline (3ha)



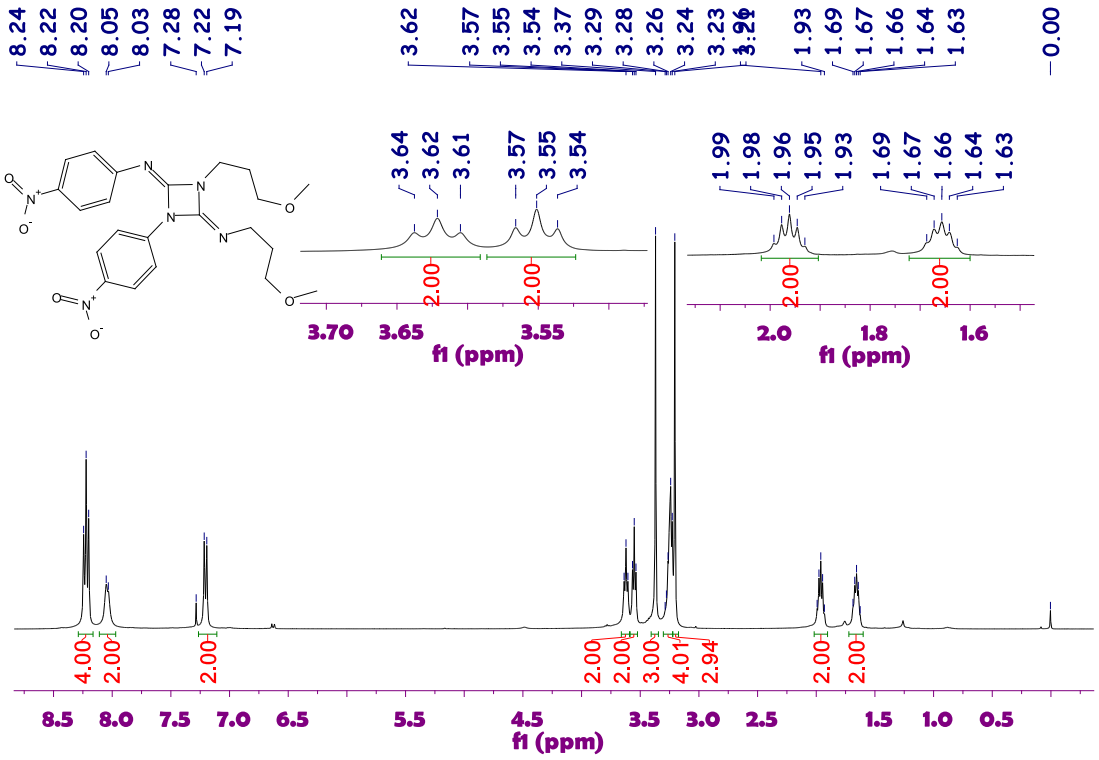


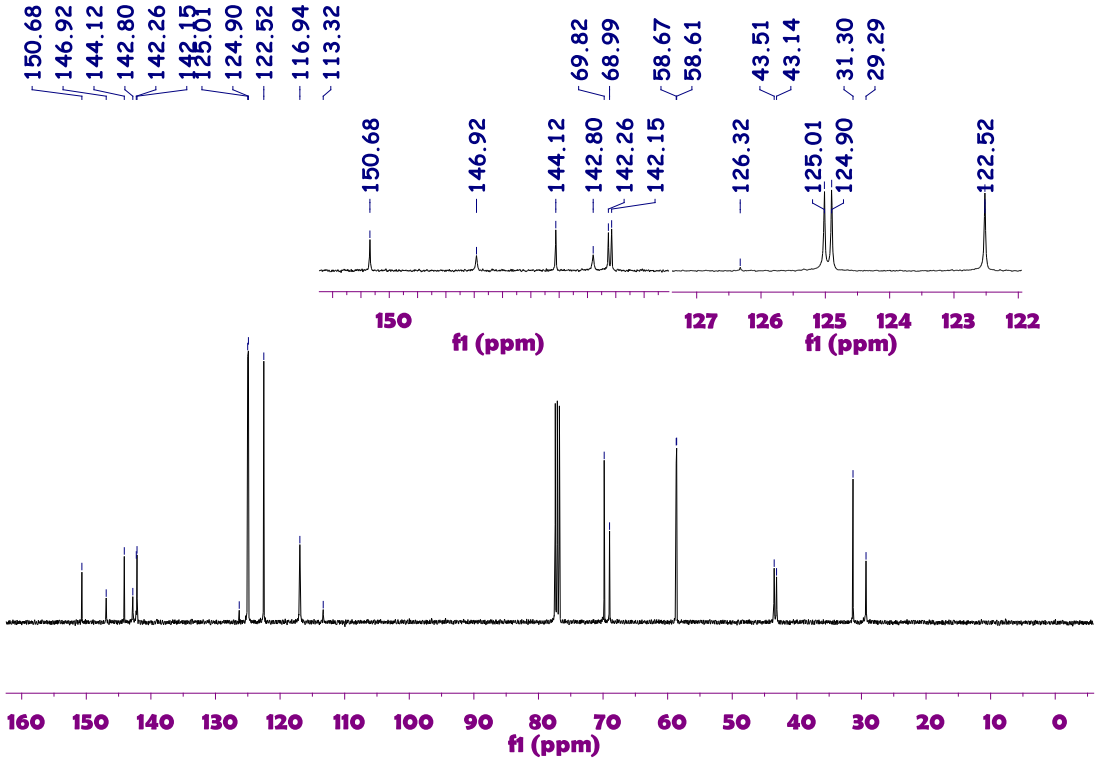
***N*-(-1-(2,2-dimethoxyethyl)-4-((2,2-dimethoxyethyl)imino)-3-(4-nitrophenyl)-1,3-diazetidin-2-yli dene)-4-nitroaniline (3ia)**



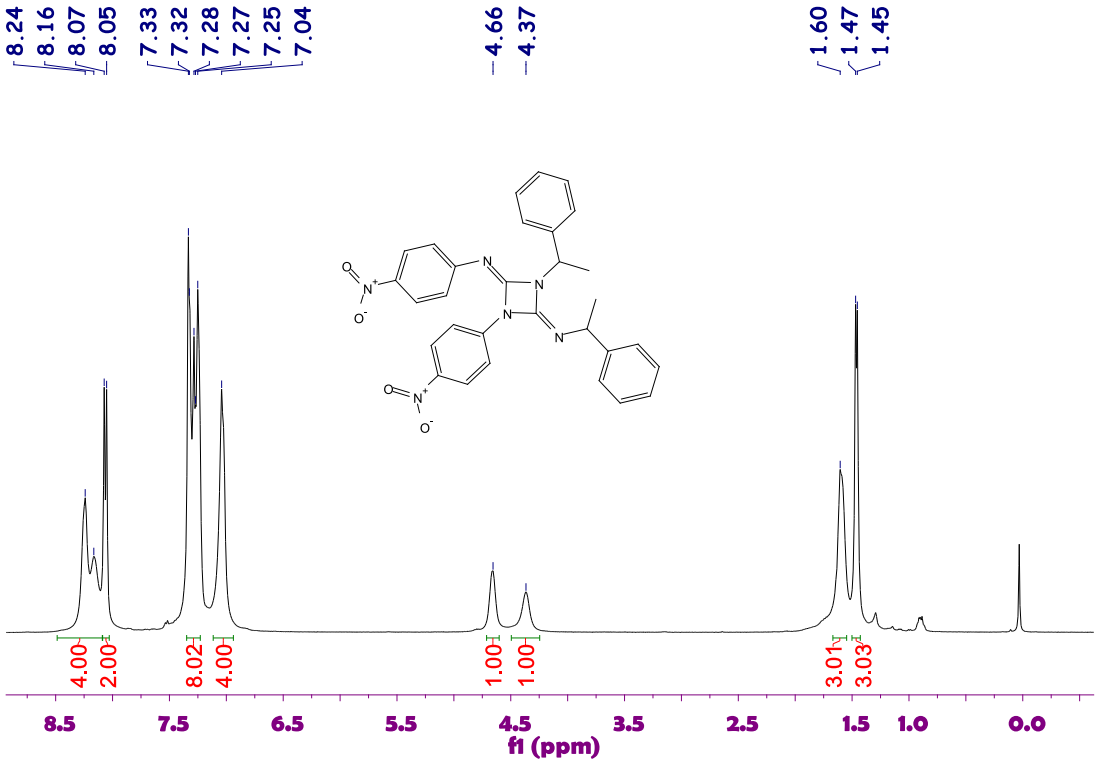


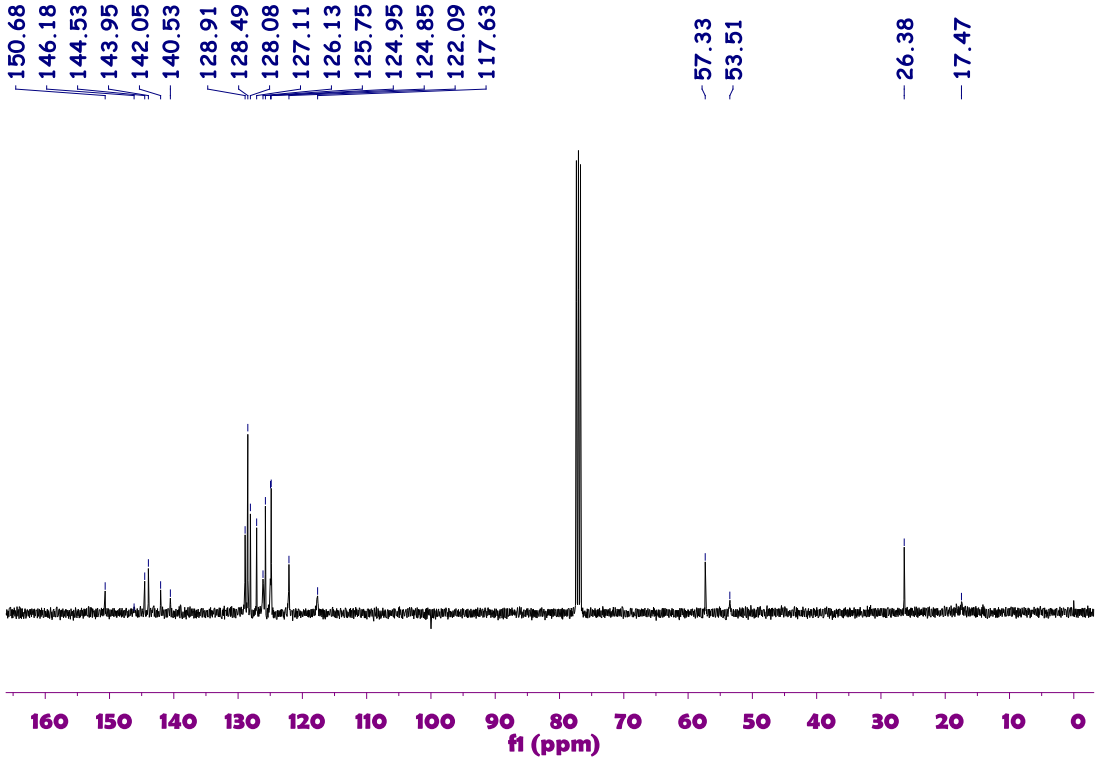
***N*-(1-(3-methoxypropyl)-4-((3-methoxypropyl)imino)-3-(4-nitrophenyl)-1,3-diazetidin-2-ylidene) -4-nitroaniline (3ja)**



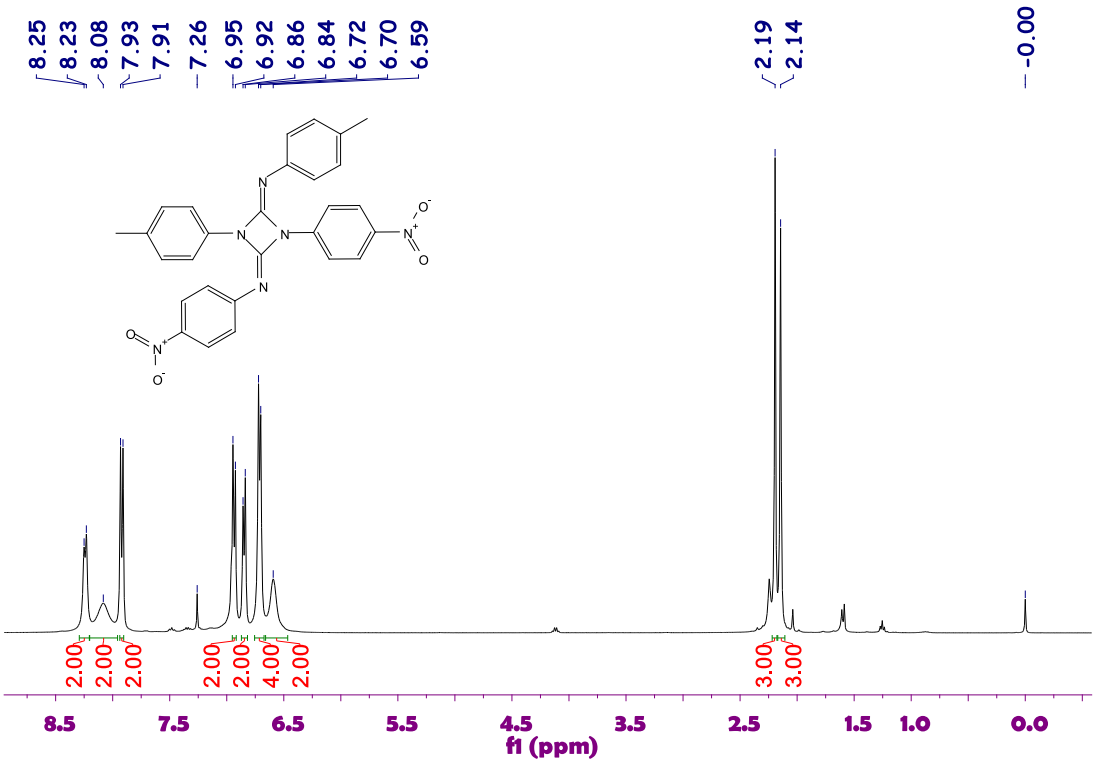


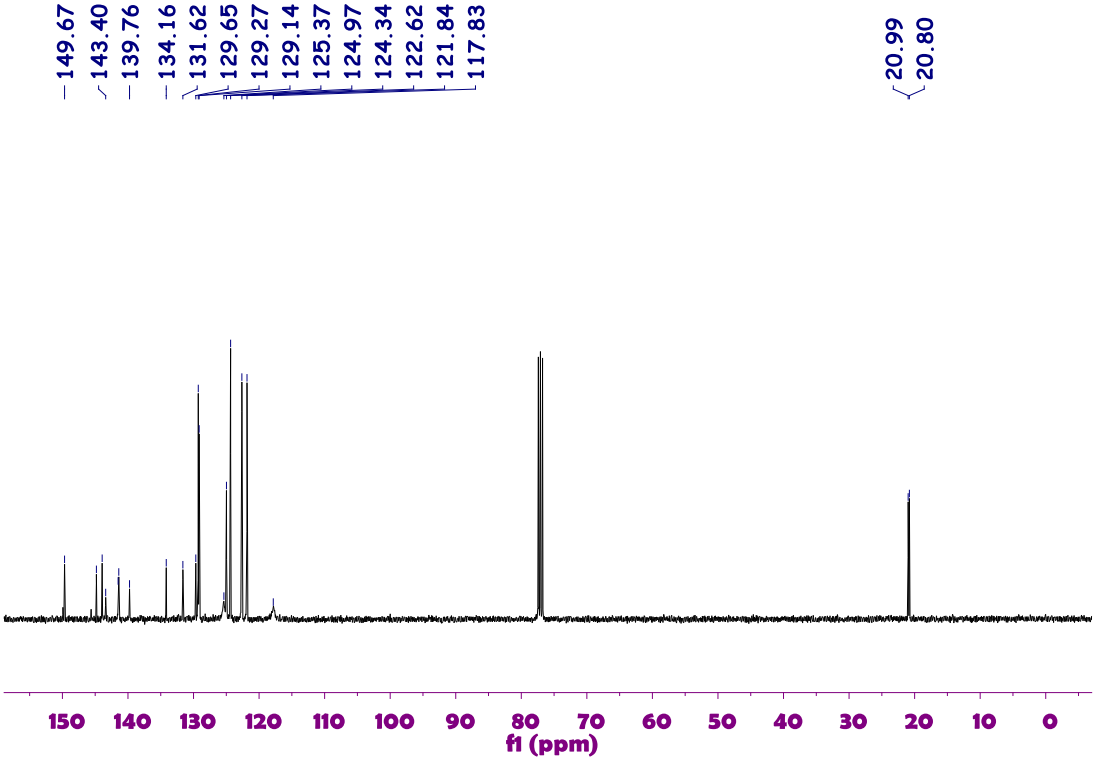
4-nitro-*N*-(1-(4-nitrophenyl)-3-(1-phenylethyl)-4-((1-phenylethyl)imino)-1,3-diazetidin-2-ylidene)aniline (3na)



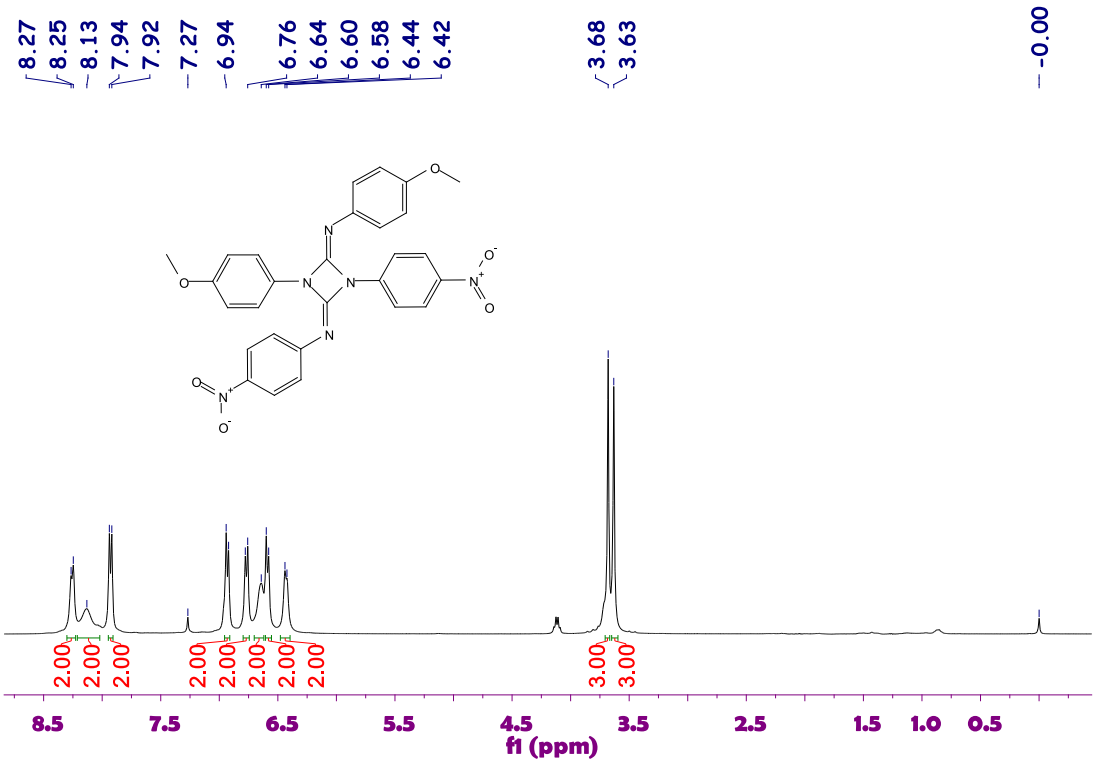


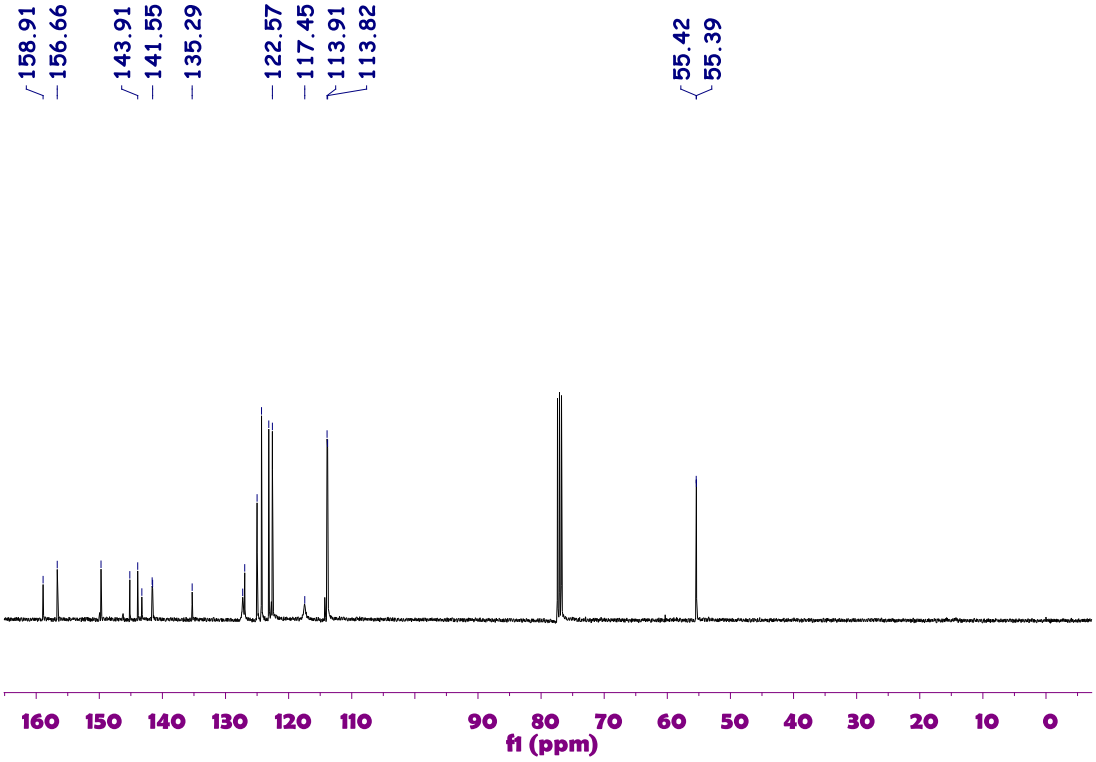
4-methyl-*N*-(-1-(4-nitrophenyl)-4-((4-nitrophenyl)imino)-3-(p-tolyl)-1,3-diazetidin-2-ylidene)aniline (3oa)



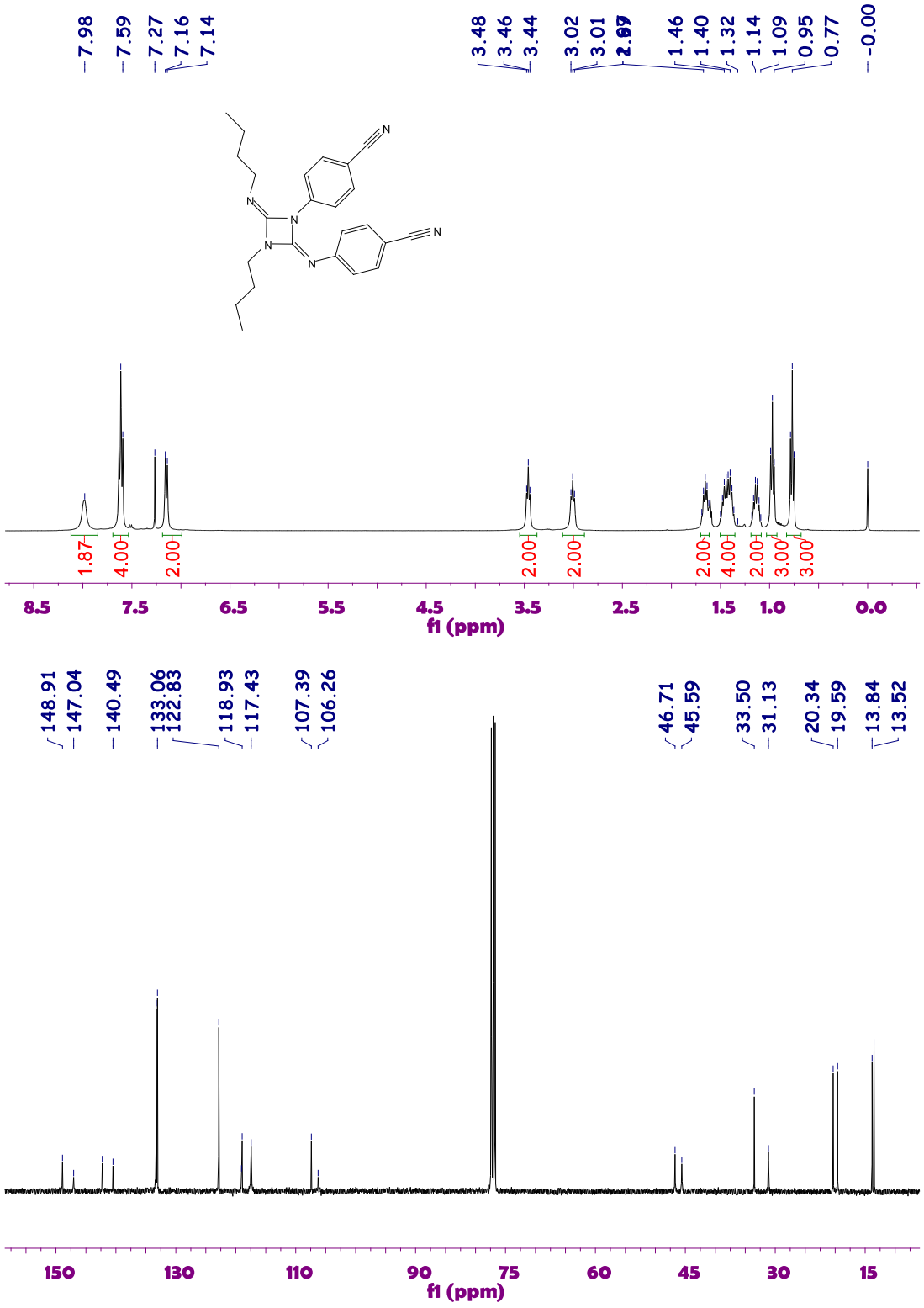


4-methoxy-*N*-(1-(4-methoxyphenyl)-3-(4-nitrophenyl)-4-((4-nitrophenyl)imino)-1,3-diazetidin-2-ylidene)aniline (3pa)

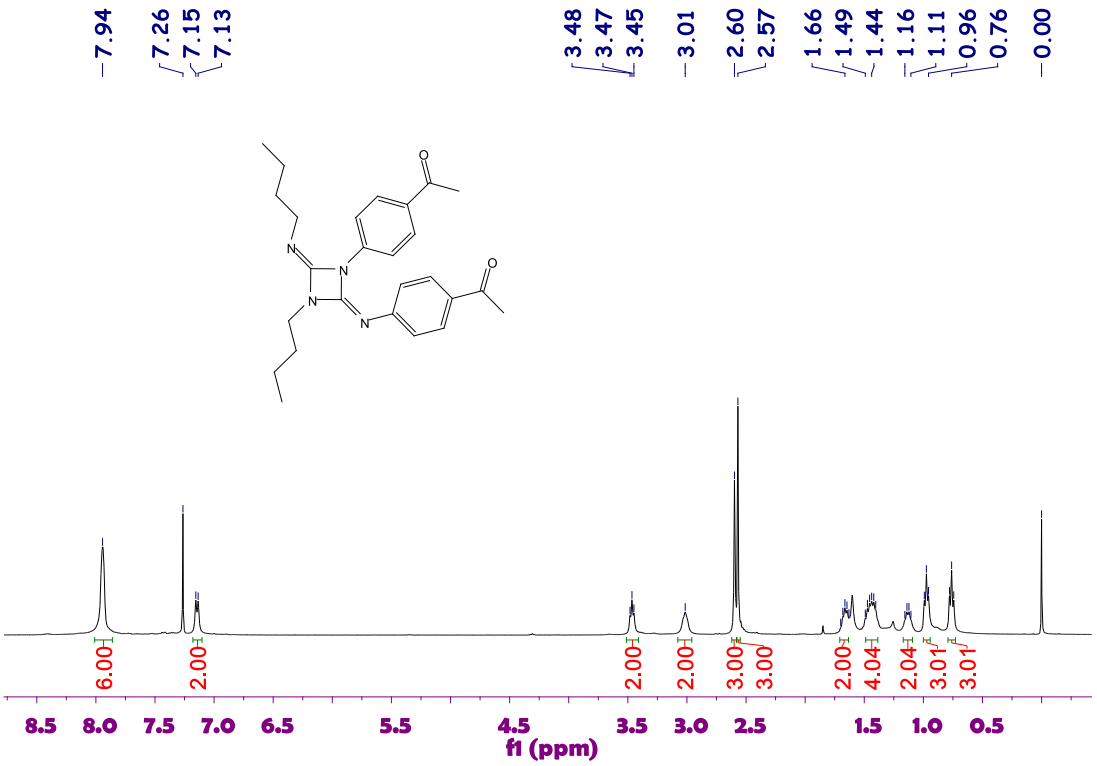


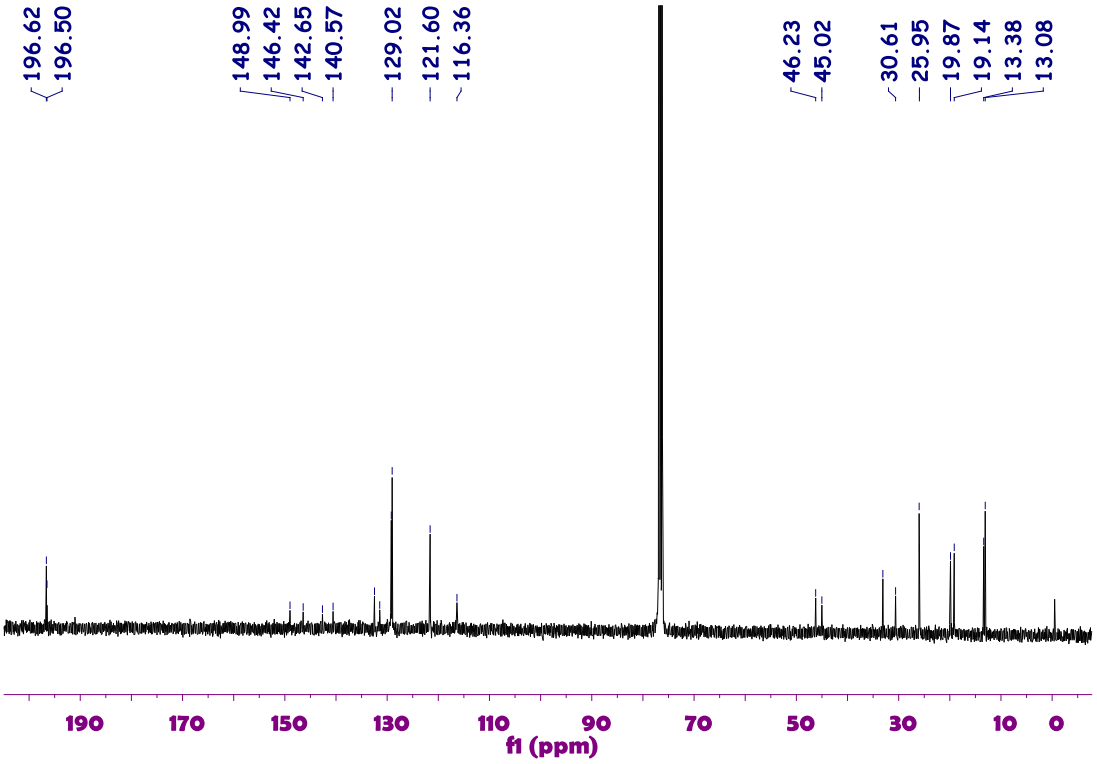


4-(3-butyl-2-(butylimino)-4-((4-cyanophenyl)imino)-1,3-diazetidin-1-yl)benzonitrile (3ab)

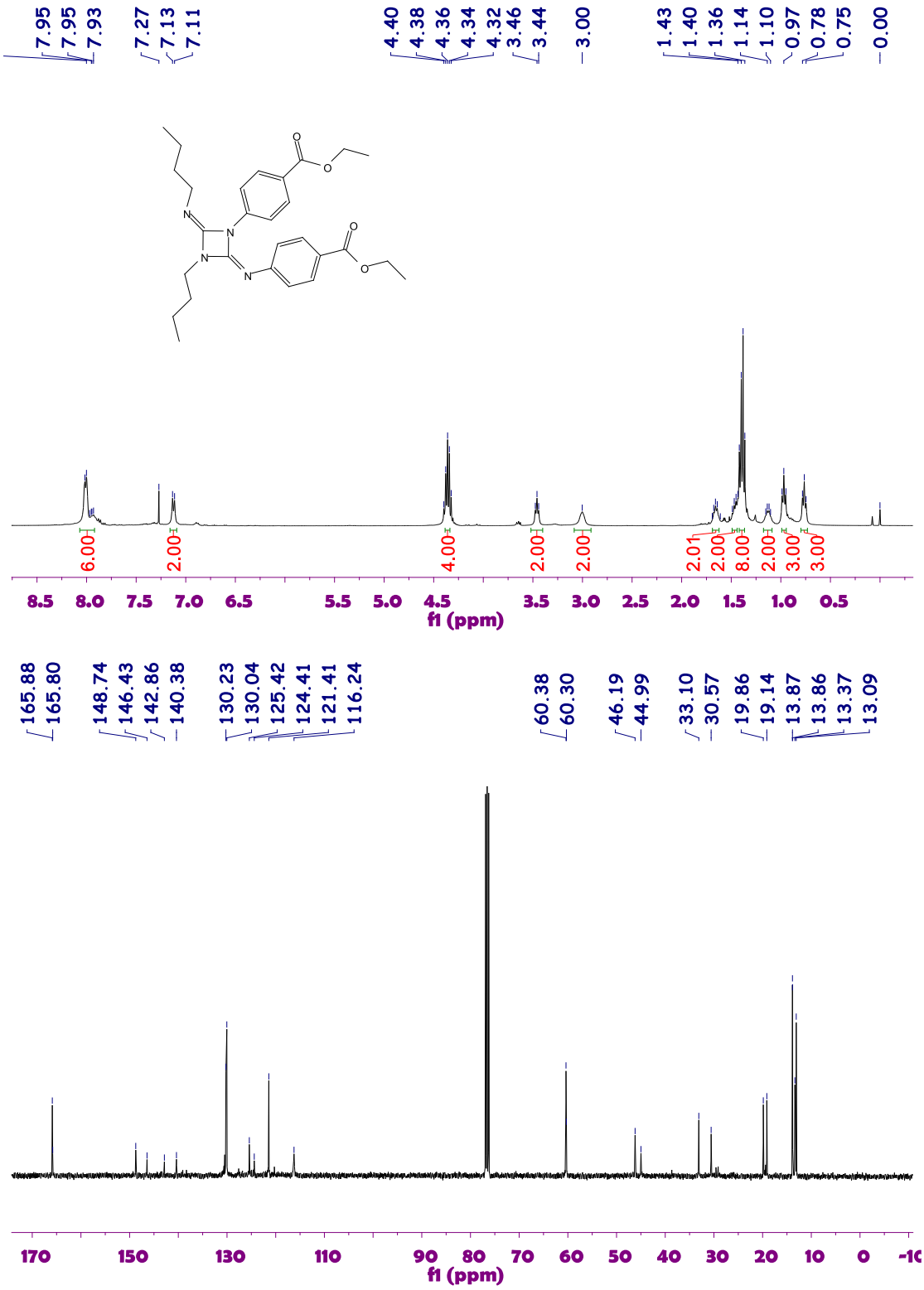


1-(4-((-1-(4-acetylphenyl)-3-butyl-4-(butylimino)-1,3-diazetidin-2-ylidene)amino)phenyl)ethanone (3ac)





Ethyl 4-((3-butyl-2-(butylimino)-4-((4-(ethoxycarbonyl)phenyl)imino)-1,3-diazetidin-1-yl)benzoate (3ad)



***N*-1-butyl-4-(butylimino)-3-(3-nitrophenyl)-1,3-diazetidin-2-ylidene)-3-nitroaniline (3af)**