Iron-Catalyzed Intramolecular C–H Amination of α-Azidyl Amides

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General methods

The $^1$H and $^{13}$C NMR spectra were recorded on a Bruker AM-300 MHz spectrometer, a Bruker AM-400 MHz spectrometer and a Bruker AM-600 MHz spectrometer with CDCl$_3$ as the solvent. The chemical shifts in $^1$H NMR spectra were determined with Si(CH$_3$)$_4$ as the internal standard ($\delta = 0.00$ ppm); the chemical shifts in $^{13}$C NMR spectra were determined based on the chemical shift of CDCl$_3$ ($\delta = 77.0$ ppm). The high resolution mass spectra (HRMS) were measured on a Bruker micrOTOF QII by ESI. The Fourier transformation infrared spectra (FT-IR) were measured on a NEXUS 670 spectrometer. Melting points (m.p.) were measured on an XT-4 melting point apparatus and are uncorrected. Flash column chromatography was carried out on aluminum oxide (200-300 mesh) or silica gel (200-300 mesh). FeCl$_2$ (99.5%), FeBr$_2$ (98.5%) and FeI$_2$ (97%) were purchased from Alfa Aesar; Anhydrous acetonitrile (MeCN) and dimethyl sulfoxide (DMSO) were purchased from Energy Chemical and used without further processing. Other solvents were treated before use following the standard procedures.

$\beta$-Diketiminate L1 was prepared following the reported procedure.$^1$ L2 and L3 were prepared according to literature methods.$^2$ Ligand L4 (4,4'-di-tert-butyl-2,2’-dipyridyl) was purchased from Sigma-Aldrich. Ligand L5 (1,10-phenanthroline) was purchased from Acros Organics.

General experimental procedures

General procedure for the preparation of compounds 1a-1l:

\[
\begin{array}{cccc}
R^1\text{N} & \text{NH}_2 & n \text{NH}_2 & n \\
R & \text{H} & \text{H} & n = 0, 1 \\
\end{array}
\]

1) MeOH, rt
2) NaBH$_4$, 0°C to rt

(a) To a flask charged with amine (6.0 mmol, 1.2 equiv.) dissolved in MeOH (0.5 M), was added dropwise benzaldehydes (5.0 mmol, 1.0 equiv.) The resulting mixture was stirred at room temperature overnight. NaBH$_4$ (0.38 g, 10 mmol, 2.0 equiv.) was then added into the mixture in several portions at 0 °C (immersed in an ice-bath), and mixture was stirred at this temperature for 6 h. Cold water was then added to the mixture till no bubbles produced, and the mixture was extracted with ethyl acetate (3×10 mL), and dried over anhydrous NaSO$_4$. The solvent was then evaporated under reduced pressure on a rotary evaporator, and the crude secondary amine product (P-1) was directly used for the next step without further purification.
(b) The crude P-1 (5.0 mmol, 1.0 equiv.) and triethylamine (Et$_3$N) (2.1 mL, 15 mmol, 3.0 equiv.) was dissolved in dry dichloromethane (DCM) (0.5 M) in a 50 mL round bottom flask. After stirring at 0 °C for 5 min, 4-dimethylaminopyridine (DMAP) (30.5 mg, 0.25 mmol, 0.05 equiv.) was added into the mixture, followed by dropwise addition of the corresponding 2-bromoisobutyryl bromide (0.74 mL, 6 mmol, 1.2 equiv.) at 0 °C. The stirring was continued until the reaction was complete as indicated by TLC. The reaction mixture was then quenched with water (10 mL). The aqueous phase was extracted with DCM (3×10 mL). The combined organic phases was treated with 0.5 M HCl solution (10 mL), washed with brine (3×15 mL), and dried over anhydrous Na$_2$SO$_4$. The solution was concentrated under reduced pressure on a rotary evaporator, and the residual was treated with silica gel column chromatography (eluent: petroleum ether (PE) and ethyl acetate (EA)) to afford the pure amide products.

(c) The thus obtained amide (5.0 mmol, 1.0 equiv.) was dissolved in DMF (0.3 M), and the solution was stirred at 0 °C (ice-water bath) for 5 min. NaN$_3$ (0.98 g, 15 mmol, 3.0 equiv.) was then added in a few portions, and the reaction mixture was stirred overnight at room temperature. After that, the reaction mixture was poured into water (15 mL), and the product was extracted with ethyl acetate (3×10 mL). The combined organic phases were washed with H$_2$O (3×10 mL) and brine (3×15 mL), dried over anhydrous Na$_2$SO$_4$, filtered and concentrated under reduced pressure on a rotary evaporator. Purification of the crude product by silica gel column chromatography (with PE and EA) affords the azidation product.

Compounds 1m, 1n, 1p and 1q were prepared from the corresponding amines following the same procedure.

**Preparation of compounds 1r and 1s:**

(a) Cyclopentanecarboxylic acid (0.48 mL, 5.0 mmol, 1.0 equiv.) or cyclobutanecarboxylic acid (0.54 mL, 5.0 mmol, 1.0 equiv.) was dissolved in a 25 mL
round bottom flask charged with SOCl₂ (10 mL), and the mixture was stirred for 1 h at 60 °C. After cooling down to room temperature, Br₂ (0.34 mL, 5.5 mmol, 1.1 equiv.) and a few drops of HBr (48% in H₂O) (or PBr₃) was added to the solution, and the stirring was continued at 60–70 °C until the red colour in the flask lightened. The volatile components were then removed by distillation, and the residual was diluted with DCM (5 mL).

(b) Then, the diluted chloride was added dropwise to a 50 ml single bottom flask charged with a DCM solution (10 mL) of dibenzylamine (0.96 mL, 5.0 mmol, 1.0 equiv.), Et₃N (2.1 mL, 15 mmol, 3 equiv.) and DMAP (30.5 mg, 0.25 mmol, 0.05 equiv.) at 0 °C, and the mixture was stirred till the reaction was complete indicated by TLC. The mixture was then added into water (10 mL), and the separated aqueous phase was extracted with DCM (3×10 mL). The combined organic phases were treated with 0.5 M HCl solution (10 mL), washed with brine (3×15 mL), and dried over anhydrous Na₂SO₄. The solution was concentrated under reduced pressure on a rotary evaporator, and the residual was treated with silica gel column chromatography to afford the pure amide products (with PE and EA). The azidation procedure was the same as described above.

Compound 1t and 1u was prepared from 2-phenylacetic acid following the same procedure.

**Preparation of compound 1o:**

Diphenethylamine was prepared according to a literature method.⁴ 1o was prepared from diphenethylamine following the procedures described above.

**Preparation of compounds 1w:**

N-((2-Phenylcyclopropyl) methyl) aniline was synthesized based on a literature method.⁵ 1w was prepared from N-((2-phenylcyclopropyl) methyl) aniline following
the procedures described above.

**Preparation of compounds 1j-d:**

To a flask charged with amine (0.55 mL, 6.0 mmol, 1.2 equiv.) dissolved in CD$_3$OD (0.5 M), was added dropwise benzaldehyde (0.51 mL, 5.0 mmol, 1.0 equiv.) The resulting mixture was stirred at room temperature till the condensation monitored was complete as indicated by TLC. NaBD$_4$ (0.63 g, 15 mmol, 3.0 equiv.) was then added into the mixture in several portions at 0 °C (immersed in an ice-bath), and mixture was stirred at this temperature for 8 h. Cold water then was added to the mixture till no bubbles produced, and the mixture was extracted with dichloromethane (3×10 mL), and dried over anhydrous Na$_2$SO$_4$. The solvent was then evaporated under reduced pressure on a rotary evaporator, and the crude secondary amine product was directly used for the next step without further purification. Acylation and azidation procedures were the same as described above.

**General procedure for the iron-catalyzed C–H amination reactions of compounds 1:**

Into an oven dried reaction tube equipped with a magnetic stirring bar and a rubber stopper were added FeCl$_2$ (5.1 mg, 0.04 mmol, 20 mol %) and L1 (13.4 mg, 0.04 mmol, 20 mol %). The tube was evacuated and backfilled with argon for three times. Then 1 mL of anhydrous acetonitrile was added into the tube with a syringe under argon atmosphere (argon balloon), the mixture was stirred for 30 min at room temperature. Another 1 mL of acetonitrile containing 0.2 mmol of 1 (1.0 equiv.) was then added into the reaction tube with a syringe. The mixture was stirred at 100 °C (oil bath temperature) for 12 h. After completed, the mixture was cooled to room temperature, and was allowed to pass through a short pad of aluminum oxide, which was washed with ethyl acetate (20 mL). The solution was concentrated under reduced pressure on a rotary evaporator, and the residual was treated with flash column chromatography (with PE and EA) on aluminum oxide to afford the pure product (s).

For the reactions of compounds 1p, 1q, 3 and 4 in the presence of Boc$_2$O, 92 uL of Boc$_2$O (2.4 mmol, 2.0 equiv.) was added into the reaction mixture before heating.

**Gram scale experiment of 1a:**

1.23 g of 1a (4.0 mmol, 1.0 equiv.), 114 mg of FeCl$_2$ (0.9 mmol 22 mol %), 267 mg of L1 (0.8 mmol, 20 mol %) and 40 mL of anhydrous acetonitrile were used following
the procedure described above.

**Kinetic isotope experiment with 1j-d:**

\[
\begin{array}{c}
\text{FeCl}_2 (20 \, \text{mol} \%) \\
\text{L1} (20 \, \text{mol} \%) \\
\text{CH}_3\text{CN}, \text{Ar}, 100 \, ^\circ\text{C}
\end{array}
\xrightarrow{42 \%}
\begin{array}{c}
\text{1j-d} \\
\text{2j-d-2} \\
\text{2j-d-1}
\end{array}
\]

The value of \(k_H/k_D\) was calculated based on the integrals of methine hydrogen (δ = 5.98 and the hydrogen (δ = 1.98) of amino group in \(^1\text{H}\) NMR spectra.

**Cautions:**

Organic azides are potentially explosive compounds. While we haven’t encountered any problems handling them in our experiments, proper precautions must be taken. All the azidation reactions and subsequent workups should be performed in a hood behind a blast shield. After they were prepared, all the organic azides were stored at -18 °C in a refrigerator.

**Characterization data**

**Characterization Data for the substrates**

**2-Azido-\(N\), \(N\)-dibenzyl-2-methylpropanamide (1a)**

White solid (1.01 g, 61 %*), m.p. = 54–55 °C; \(^1\text{H}\) NMR (400 MHz, CDCl\(_3\), δ ppm) 7.42–7.24 (m, 6H), 7.21–7.09 (m, 4H), 4.86 (s, 2H), 4.52 (s, 2H), 1.62 (s, 6H); \(^{13}\text{C}\) NMR (100 MHz, CDCl\(_3\), δ ppm) 171.9, 136.9, 136.5, 128.7, 128.6, 127.9, 127.4, 126.8, 64.1, 50.6, 48.6, 25.5; FT-IR (KBr, cm\(^{-1}\)): 2107, 1643; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for [C\(_{18}\)H\(_{21}\)N\(_4\)O]: 309.1710, found: 309.1700.

**2-Azido-\(N\), \(N\)-bis(4-methoxybenzyl)-2-methylpropanamide (1b)**

Colorless oil (1.51 g, 82 %), \(R_f = 0.56\) (PE:EA = 5:1); \(^1\text{H}\) NMR (400 MHz, CDCl\(_3\), δ ppm): 7.15–7.03 (m, 4H), 6.95–6.79 (m, 4H), 4.76 (s, 2H), 4.43 (s, 2H), 3.80 (s, 6H), 1.61 (s, 6H); \(^{13}\text{C}\) NMR (100 MHz, CDCl\(_3\), δ ppm): 171.7, 159.0, 129.4, 129.0, 128.3,
128.2, 114.2, 114.0, 64.2, 55.2, 49.7, 47.6, 25.5; FT-IR (KBr, cm⁻¹): 2107, 1641; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C₂₀H₂₅N₄O₃]⁺: 369.1921, found: 369.1914.

2-Azido-\(\text{N, N}-\text{bis}(2\text{-methoxybenzyl})\)-2-methylpropanamide (1c)
Colorless oil (1.24 g, 67 %), \(R_f = 0.41\) (PE:EA = 10:1); \(^1\)H NMR (400 MHz, CDCl₃, \(\delta\) ppm): 7.34–7.16 (m, 2H), 6.88–6.79 (m, 2H), 6.78–6.66 (m, 4H), 4.85 (s, 2H), 4.51 (s, 2H), 3.78 (s, 6H), 1.62 (s, 6H); \(^1^3\)C NMR (100 MHz, CDCl₃, \(\delta\) ppm): 171.9, 160.0, 159.8, 138.5, 138.2, 129.8, 129.6, 120.2, 119.1, 113.2, 113.0, 112.4, 64.1, 55.1, 50.6, 48.7, 25.5; FT-IR (KBr, cm⁻¹): 2108, 1643; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C₂₀H₂₅N₄O₃]⁺: 369.1921, found: 369.1924.

2-Azido-\(\text{N, N}-\text{bis}(3\text{-methoxybenzyl})\)-2-methylpropanamide (1d)
Light yellow solid (1.01 g, 55 %), m.p. = 93–95 °C; \(^1\)H NMR (600 MHz, CDCl₃, \(\delta\) ppm): 7.27–7.17 (m, 2H), 7.14–7.08 (m, 2H), 7.00–6.93 (m, 1H), 6.93–6.87 (m, 1H), 6.87–6.76 (m, 2H), 4.93 (s, 2H), 4.58 (s, 2H), 3.76 (s, 3H), 3.69 (s, 3H), 1.56 (s, 6H). \(^1^3\)C NMR (150 MHz, CDCl₃, \(\delta\) ppm): 172.2, 157.4, 156.8, 128.0, 127.9, 126.0, 125.4, 124.9, 120.4, 101.0, 64.1, 55.0, 46.9, 45.3, 25.5. FT-IR (KBr, cm⁻¹): 2108, 1643; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C₂₀H₂₅N₄O₃]⁺: 369.1921, found: 369.1923.

2-Azido-\(\text{N, N}-\text{bis}(4\text{-fluorobenzyl})\)-2-methylpropanamide (1e)
White solid (1.37 g, 80 %), m.p. = 73–74 °C; \(^1\)H NMR (400 MHz, CDCl₃, \(\delta\) ppm): 7.21–6.93 (m, 8H), 4.81 (s, 2H), 4.45 (s, 2H), 1.62 (s, 6H); \(^1^3\)C NMR (100 MHz, CDCl₃, \(\delta\) ppm): 171.3, 162.15 (d, \(J = 246.0\) Hz), 132.5, 132.0, 129.7, 128.6, 115.6, 64.1, 49.9, 47.9, 25.4; FT-IR (KBr, cm⁻¹): 2109, 1644; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C₁₉H₁₉F₂N₄O]⁺: 345.1521, found: 345.1524.
2-Azido-\(\text{N, N-bis(3-chlorobenzyl)-2-methylpropanamide (1f)}\)
Light yellow solid (1.79 g, 95 %), m.p. = 55–57 °C; \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 7.36–7.21 (m, 4H), 7.20–6.94 (m, 4H), 4.85 (s, 2H), 4.48 (s, 2H), 1.62 (s, 6H). \(^1\)^1^C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 172.0, 138.7, 134.6, 130.09, 127.8, 127.0, 126.0, 124.9, 64.0, 50.5, 48.6, 25.5; FT-IR (KBr, cm\(^{-1}\)): 2107, 1640; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for [C\(_{18}\)H\(_{19}\)Cl\(_2\)N\(_4\)O]\(^+\): 377.0930, found: 377.0928.

2-Azido-\(\text{N, N-bis(2-bromobenzyl)-2-methylpropanamide (1g)}\)
White solid (1.72 g, 76 %), m.p. = 107–109 °C; \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 7.61–7.50 (m, 2H), 7.42–7.34 (m, 1H), 7.33–7.24 (m, 1H), 7.23–7.10 (m, 4H), 4.92 (s, 2H), 4.67 (s, 2H), 1.59 (s, 6H); \(^1\)^C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 172.5, 135.8, 135.5, 133.1, 128.9, 128.4, 127.8, 127.1, 64.0, 52.1, 50.4, 25.5. FT-IR (KBr, cm\(^{-1}\)): 2108, 1649; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for [C\(_{18}\)H\(_{19}\)Br\(_2\)N\(_4\)O]\(^+\): 466.9900, found: 466.9899.

2-Azido-\(\text{N-(4-fluorobenzyl)-2-methyl-}-N-(4-methylbenzyl)propanamide (1h)}\)
Yellow oil (1.35 g, 76 %), \(R_f = 0.35\) (PE:EA = 5:1); \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 7.19–7.04 (m, 5H), 7.04–6.94 (m, 1H), 6.93–6.79 (m, 2H), 4.78 (s, 2H), 4.43 (s, 2H), 3.81 (s, 3H), 1.61 (s, 6H). \(^1\)^C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 171.7, 162.07 (d, \(J = 245.9\) Hz) 159.0, 132.7, 129.7, 129.4, 128.5, 128.2, 115.8, 115.5, 114.2, 114.0, 64.1, 55.2, 50.0, 47.6, 25.5. FT-IR (KBr, cm\(^{-1}\)): 2108, 1642; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for [C\(_{19}\)H\(_{22}\)FN\(_4\)O\(_2\)]\(^+\): 357.1721, found: 357.1717.
2-Azido-2-methyl-\(N\)-, \(N\)-bis(pyridin-2-ylmethyl)propanamide (1i)

Yellow solid (1.13 g, 73 %), \(R_f = 0.33\) (PE:EA = 1:2); \(^1\)H NMR (300 MHz, CDCl\(_3\), \(\delta\) ppm): 8.54 (d, \(J = 21.4\) Hz, 2H), 7.74–7.57 (m, 2H), 7.21 (d, \(J = 21.4\) Hz, 4H), 5.17 (s, 2H), 4.69 (s, 2H), 1.61 (s, 6H); \(^{13}\)C NMR (75 MHz, CDCl\(_3\), \(\delta\) ppm): 172.2, 156.9, 149.6, 149.2, 136.6, 122.2, 121.7, 120.9, 63.9, 53.9, 52.2, 25.4; FT-IR (KBr, cm\(^{-1}\)): 2109, 1643; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for \([\text{C}_{16}\text{H}_{19}\text{N}_6\text{O}]^+\): 311.1615, found: 311.1616.

\[
\begin{array}{c}
\text{N} \\
\text{O}
\end{array}
\]

2-Azido-\(N\)-benzyl-2-methyl-\(N\)-phenylpropanamide (1j)

Colorless oil (1.07 g, 73 %), \(R_f = 0.38\) (PE:EA = 10:1); \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 7.34–7.29 (m, 3H), 7.28–7.23 (m, 3H), 7.18–7.14 (m, 2H), 7.05–7.01 (m, 2H), 4.85 (s, 2H), 1.45 (s, 6H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 171.9, 142.2, 137.0, 129.0, 128.9, 128.3, 128.2, 128.0, 127.5, 64.8, 56.5, 26.6; FT-IR (KBr, cm\(^{-1}\)): 2108, 1647; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for \([\text{C}_{17}\text{H}_{19}\text{N}_4\text{O}]^+\): 295.1553, found: 295.1560.

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\begin{array}{c}
\text{N} \\
\text{O}
\end{array}
\]

2-Azido-2-methyl-\(N\)-phenyl-\(N\)-(phenylmethyl-\(d\)) propanamide (1j-d)

Light yellow or colorless liquid (1.34 g, 90 %), \(R_f = 0.58\) (PE:EA = 5:1); \(^1\)H NMR (600 MHz, CDCl\(_3\), \(\delta\) ppm): 7.33–7.29 (m, 3H), 7.28–7.24 (m, 3H), 7.18–7.15 (m, 2H), 7.05–7.02 (m, 2H), 4.85 (s, 1H) \([\text{Ph-CHD}]\), 1.46 (s, 6H); \(^{13}\)C NMR (150 MHz, CDCl\(_3\), \(\delta\) ppm): 171.9, 142.2, 137.0, 129.0, 128.9, 128.3, 128.2, 128.0, 127.5, 64.8, 56.1, 26.6. The data was corresponding to the reactant 1j.

\[
\begin{array}{c}
\text{N} \\
\text{O}
\end{array}
\]

2-Azido-2-methyl-\(N\)-(4-methylbenzyl)-\(N\)-phenylpropanamide (1k)

Light yellow solid (1.02 g, 66 %), m.p. = 40–41 °C; \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 7.35–7.27 (m, 3H), 7.09–6.98 (m, 6H), 4.81 (s, 2H), 2.31 (s, 3H), 1.44 (s, 6H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 171.7, 142.2, 137.1, 133.9, 129.0, 128.9, 128.9,
128.2, 127.9, 64.8, 56.1, 26.5, 21.1; FT-IR (KBr, cm\(^{-1}\)): 2108, 1647; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C\(_{18}\)H\(_{21}\)N\(_4\)O]+: 309.1710, found: 309.1716.

2-Azido-N-(4-fluorobenzyl)-2-methyl-N-phenylpropanamide (1l)

Yellow oil (1.18 g, 76 %), \(R_f = 0.56\) (PE:EA = 5:1); \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 7.36–7.28 (m, 3H), 7.17–7.08 (m, 2H), 7.03–6.97 (m, 2H), 6.97–6.89 (m, 2H), 4.81 (s, 2H), 1.44 (s, 6H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 171.9, 162.18 (d, \(J = 245.9\) Hz), 141.9, 132.8, 132.8, 130.8, 130.7, 129.0, 128.2, 128.1, 115.3, 115.0, 64.7, 55.6, 26.5; FT-IR (KBr, cm\(^{-1}\)): 2109, 1647; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C\(_{17}\)H\(_{19}\)FN\(_4\)O]+: 313.1460, found: 313.1466.

2-Azido-N-benzyl-N-2-dimethylpropanamide (1m)

Light yellow oil (0.72 g, 62 %), \(R_f = 0.72\) (PE:EA = 1:1); \(^1\)H NMR (300 MHz, CDCl\(_3\), \(\delta\) ppm): 7.39–7.30 (m, 2H), 7.30–7.26 (m, 1H), 7.25–7.18 (m, 2H), 4.67 (s, 2H), 3.12 (s, 3H), 1.58 (s, 6H); \(^{13}\)C NMR (75 MHz, CDCl\(_3\), \(\delta\) ppm): 171.4, 136.9, 128.6, 127.4, 64.1, 53.1, 35.6, 25.2; FT-IR (KBr, cm\(^{-1}\)): 2107, 1642; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C\(_{12}\)H\(_{17}\)N\(_4\)O]+: 233.1397, found: 233.1396.

2-Azido-N-benzyl-2-methyl-N-(1-phenylethyl) propanamide (1n)

Colorless or light yellow oil (1.37 g, 84 %), \(R_f = 0.45\) (PE:EA = 10:1); \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 7.42–7.33 (m, 2H), 7.32–7.27 (m, 4H), 7.24–7.14 (m, 2H), 7.14–7.05 (m, 2H), 6.03 (s, 1H), 4.84 (d, \(J = 15.5\) Hz, 1H), 3.80 (d, \(J = 15.7\) Hz, 1H), 1.69 (s, 3H), 1.60 (s, 6H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 172.2, 140.7, 138.7, 128.7, 128.3, 127.4, 126.7, 64.5, 55.9, 47.9, 26.0, 25.7; FT-IR (KBr, cm\(^{-1}\)): 2105, 1641; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C\(_{19}\)H\(_{23}\)N\(_4\)O]+: 323.1866, found: 323.1859.
2-Azido-2-methyl-\(N, N\)-diphenethylpropanamide (1o)

Colorless oil (0.15 g, 9 %), \(R_f = 0.31\) (PE:EA = 10:1); \(^1\)H NMR (300 MHz, CDCl\(_3\), \(\delta\) ppm): 7.36–7.13 (m, 10H), 3.61 (s, 4H), 2.91 (s, 4H), 1.54 (s, 6H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 170.9, 139.1, 138.1, 128.8, 128.7, 128.6, 128.5, 126.8, 126.4, 64.0, 50.8, 49.4, 35.5, 33.6, 25.4. FT-IR (KBr, cm\(^{-1}\))): 2107, 1632; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for \([C_{20}H_{25}N_4O]\): 337.2023, found: 337.2029.

\[
\begin{array}{c}
\text{N}3 \\
\text{O} \\
\text{N}3
\end{array}
\]

2-Azido-2-methyl-1-((piperidin-1-yl) propan-1-one (1p)

Colorless liquid (0.50 g, 51 %), \(R_f = 0.25\) (PE:EA = 5:1); \(^1\)H NMR (300 MHz, CDCl\(_3\), \(\delta\) ppm): 3.79–3.57 (m, 4H), 1.71–1.55 (m, 6H), 1.51 (s, 6H); \(^{13}\)C NMR (75 MHz, CDCl\(_3\), \(\delta\) ppm): 169.4, 64.1,46.4, 26.3, 25.2, 24.5; FT-IR (KBr, cm\(^{-1}\))): 2105, 1641; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for \([C_9H_{17}N_4O]\): 197.1396; found: 197.1396.

\[
\begin{array}{c}
\text{N}3 \\
\text{O} \\
\text{N}3
\end{array}
\]

2-Azido-2-methyl-1-((pyrrolidin-1-yl) propan-1-one (1q)

Colorless liquid (0.36 g, 37 %), \(R_f = 0.19\) (PE:EA = 5:1), \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 3.73 (t, \(J = 6.9\) Hz, 2H), 3.51 (t, \(J = 6.9\) Hz, 2H), 1.99–1.89 (m, 2H), 1.89–1.78 (m, 2H), 1.52 (s, 6H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 169.8, 64.2, 47.8, 47.5, 27.0, 24.5, 23.1; FT-IR (KBr, cm\(^{-1}\))): 2106,1634; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for \([C_8H_{18}N_4O]\): 183.1240, found: 183.1240.

\[
\begin{array}{c}
\text{N}3 \\
\text{O} \\
\text{N}3
\end{array}
\]

1-Azido-\(N, N\)-dibenzycyclobutane-1-carboxamide (1r)

Colorless oil (1.07 g, 67 %), \(R_f = 0.40\) (PE:EA = 10:1), \(^1\)H NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): 7.40–7.34 (m, 2H), 7.33–7.23 (m, 4H), 7.20–7.11 (m, 4H), 4.52 (s, 2H), 4.40 (s, 2H), 2.80 (dddd, \(J = 12.1, 9.6, 6.2, 2.8\) Hz, 2H), 2.28 (dddd, \(J = 12.0, 8.7, 5.8, 3.0\) Hz, 2H), 2.04 (dtt, \(J = 11.4, 9.4, 6.9\) Hz, 1H), 1.86 (dtt, \(J = 11.7, 9.2, 6.0\) Hz, 1H); \(^{13}\)C
NMR (100 MHz, CDCl₃, δ ppm): 170.3, 136.8, 136.1, 128.8, 128.6, 128.1, 127.6, 127.4, 127.0, 66.2, 49.8, 47.6, 31.1, 29.5, 14.3; FT-IR (KBr, cm⁻¹): 2105, 1724; HRMS (ESI-TOF) m/z: [M+H]^+ calcd for [C₁₉H₂₁N₄O]^+: 321.1710, found: 321.1715.

1-Azido-N, N-dibenzylcyclopentane-1-carboxamide (1s)
Yellow solid (0.86 g, 51 %), m.p. = 67–68 °C; ^1H NMR (400 MHz, CDCl₃, δ ppm): 7.41–7.34 (m, 2H), 7.34–7.25 (m, 4H), 7.19–7.12 (m, 4H), 4.75 (s, 2H), 4.55 (s, 2H), 2.41–2.29 (m, 2H), 2.06–1.92 (m, 2H), 1.86–1.72 (m, 4H); ^13C NMR (100 MHz, CDCl₃, δ ppm): 171.5, 136.9, 136.5, 128.8, 128.6, 128.0, 127.4, 126.7, 74.4, 50.3, 48.3, 36.2, 23.9; FT-IR (KBr, cm⁻¹): 2102, 1643; HRMS (ESI-TOF) m/z: [M+H]^+ calcd for [C₂₀H₂₃N₄O]^+: 335.1866, found: 335.1873.

2-Azido-N, N-dibenzylpropanamide (1t)
Light yellow oil (0.85 g, 58 %), R_f = 0.31(PE:EA = 10:1), ^1H NMR (400 MHz, CDCl₃, δ ppm): 7.40–7.27 (m, 6H), 7.23–7.20 (m, 2H), 7.16–7.11 (m, 2H), 4.72–4.35 (m, 4H), 3.98 (q, J = 6.7 Hz, 1H), 1.53 (d, J = 6.7 Hz, 3H); ^13C NMR (100 MHz, CDCl₃, δ ppm): 170.9, 136.6, 135.8, 129.0, 128.7, 128.2, 127.8, 126.2, 126.6, 54.0, 49.6, 48.6, 16.2; FT-IR (KBr, cm⁻¹): 2118, 1656; HRMS (ESI-TOF) m/z: [M+H]^+ calcd for [C₁₇H₁₉N₄O]^+: 295.1553; found: 295.1557.

2-Azido-N, N-dibenzyl-2-phenylacetamide (1u)
Yellow oil (0.75 g, 42 %), R_f = 0.49 (PE:EA = 10:1); ^1H NMR (400 MHz, CDCl₃, δ ppm): 7.47–7.37 (m, 5H), 7.35–7.22 (m, 6H), 7.22–7.12 (m, 2H), 7.04–6.96 (m, 2H), 5.08 (d, J = 14.7 Hz, 1H), 4.91 (s, 1H), 4.35–4.10 (m, 3H); ^13C NMR (100 MHz, CDCl₃, δ ppm): 169.5, 136.5, 135.3, 133.7, 129.4, 129.4, 129.0, 128.6, 128.3, 128.0, 127.9, 127.6, 126.2, 63.6, 49.4, 48.8; FT-IR (KBr, cm⁻¹): 2098, 1658; HRMS
(ESI-TOF) m/z: \([M+H]^+\) calcd for \([C_{22}H_{21}N_4O]^+\): 357.1710, found: 357.1719.

\[
\begin{align*}
\text{2-Azido-} & \text{N, N-dibenzylacetamide (1v)} \\
\text{Yellow oil (0.87 g, 62 %), } R_f & = 0.16 \text{ (PE:EA = 10:1), } ^1\text{H NMR (400 MHz, CDCl}_3, \delta \text{ ppm): } \delta 7.39-7.18 \text{ (m, 8H), 7.15-7.09 (m, 2H), 4.64 (s, 2H), 4.36 (s, 2H), 3.96 (s, 2H); } ^{13}\text{C NMR (100 MHz, CDCl}_3, \delta \text{ ppm): 167.9, 136.4, 135.4, 129.2, 129.1, 128.7, 128.4, 128.2, 128.0, 127.7, 126.2, 50.6, 49.4, 48.9; FT-IR (KBr, cm}^{-1}\text{): 2117,1648; HRMS (ESI-TOF) m/z: [M+H]^+ \text{ calcd for } [C_{16}H_{17}N_4O]^+: 281.1397, \text{ found: 281.1400.}
\end{align*}
\]

\[
\begin{align*}
\text{2-Azido-2-methyl-N-phenyl-N-(2-phenylcyclopropyl) propanamide (1w)} \\
\text{Light yellow oil (0.41 g, 25 %), } R_f & = 0.49 \text{ (PE:EA = 5:1); } ^1\text{H NMR (300 MHz, CDCl}_3, \delta \text{ ppm): 7.37-7.29 (m, 3H), 7.26-7.17 (m, 4H), 7.16-7.07 (m, 1H), 6.97-6.89 (m, 2H), 3.92 (dd, } J = 13.8, 6.3 \text{ Hz, 1H), 3.52 (dd, } J = 13.7, 7.8 \text{ Hz, 1H), 1.69-1.60 (m, 1H), 1.42 (d, } J = 4.8 \text{ Hz, 6H), 1.37-1.26 (m, 1H), 0.86 (dtt, } J = 23.4, 8.7, 5.2 \text{ Hz, 2H); } ^{13}\text{C NMR (75 MHz, CDCl}_3, \delta \text{ ppm): 171.7, 142.5, 142.4, 129.0, 128.4, 128.2, 127.9, 125.6, 125.5, 64.8, 56.4, 26.5, 22.5, 21.7, 14.4; FT-IR (KBr, cm}^{-1}\text{): 2109, 1647; HRMS (ESI-TOF) m/z: [M+H]^+ \text{ calcd for } [C_{20}H_{23}N_4O]^+: 335.1866, \text{ found: 335.1870.}
\end{align*}
\]

\[
\begin{align*}
\text{(4-Azidobutyl) benzene (3)} & ^6 \\
\text{Light yellow or colorless liquid; } ^1\text{H NMR (300 MHz, CDCl}_3, \delta \text{ ppm): 7.32-7.24 (m, 2H), 7.22-7.13 (m, 3H), 3.26 (t, } J = 6.5 \text{ Hz, 2H), 2.63 (t, } J = 7.3 \text{ Hz, 2H), 1.76-1.55 (m, 4H); } ^{13}\text{C NMR (75 MHz, CDCl}_3, \delta \text{ ppm): 141.8, 128.3, 125.8, 51.3, 35.3, 28.4; FT-IR (KBr, cm}^{-1}\text{): 2094.}
\end{align*}
\]

\[
\begin{align*}
\text{(3-Azidopropoxy) benzene (4)} & ^6 \\
\text{Light yellow or colorless liquid; } ^1\text{H NMR (400 MHz, CDCl}_3, \delta \text{ ppm): 7.36-7.33 (m, 4H), 7.32-7.26 (m, 1H), 4.57 (s, 2H), 3.64 (t, } J = 5.0 \text{ Hz, 2H), 3.39 (t, } J = 5.0 \text{ Hz, 2H);}
\end{align*}
\]
\[ ^{13}C \text{ NMR (100 MHz, CDCl}_3, \delta \text{ ppm): 137.7, 128.4, 127.7, 127.6, 73.2, 68.8, 50.8.} \]

FT-IR (KBr, cm\(^{-1}\)): 2102.

*The yield given is for the last azidation step.

**Characterization Data of Products**

![Chemical Structure](image)

**3-Benzyl-5, 5-dimethyl-2-phenylimidazolidin-4-one (2a)**

White solid (55.5 mg, 99 %), m.p. = 86–88 °C; \(^1H\) NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): \(\delta\) 7.43–7.38 (m, 3H), 7.28–7.22 (m, 5H), 7.05–6.99 (m, 2H), 5.04 (s, 1H), 5.01 (d, \(J = 14.6\) Hz, 1H), 3.52 (d, \(J = 14.5\) Hz, 1H), 1.86 (s, 1H), 1.46 (s, 3H), 1.28 (s, 3H); \(^{13}C\) NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 178.1, 137.9, 135.8, 129.4, 129.1, 128.5, 128.2, 127.6, 127.2, 73.0, 59.4, 44.3, 25.5, 24.4; HRMS (ESI-TOF) m/z: [M+H]^+ calcd for [C\(_{18}\)H\(_{21}\)N\(_2\)O]\(^+\): 281.1648, found: 281.1647.

![Chemical Structure](image)

**3-(4-Methoxybenzyl)-2-(4-methoxyphenyl)-5, 5-dimethylimidazolidin-4-one (2b)**

Colorless or light yellow oil (55.8 mg, 84 %), \(R_f = 0.21\) (PE:EA = 1:1); \(^1H\) NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): \(\delta\) 7.17 (d, \(J = 8.2\) Hz, 2H), 6.93 (t, \(J = 8.4\) Hz, 4H), 6.78 (d, \(J = 8.1\) Hz, 2H), 4.99 (s, 1H), 4.92 (d, \(J = 14.4\) Hz, 1H), 3.83 (s, 3H), 3.78 (s, 3H), 3.46 (d, \(J = 14.4\) Hz, 1H), 1.85 (s, 1H), 1.44 (s, 3H), 1.25 (s, 3H); \(^{13}C\) NMR (100 MHz, CDCl\(_3\), \(\delta\) ppm): 178.0, 160.3, 159.0, 123.0, 129.6, 128.6, 128.1, 114.4, 113.8, 72.5, 59.3, 55.2, 55.1, 43.6, 25.5, 24.2. HRMS (ESI-TOF) m/z: [M+H]^+ calcd for [C\(_{20}\)H\(_{25}\)N\(_2\)O\(_3\)]^+: 341.1860, found: 341.1859.

![Chemical Structure](image)

**3-(2-Methoxybenzyl)-2-(2-methoxyphenyl)-5, 5-dimethylimidazolidin-4-one (2c)**

Colorless oil (59.9 mg, 88 %), \(R_f = 0.36\) (PE:EA = 1:1); \(^1H\) NMR (400 MHz, CDCl\(_3\), \(\delta\) ppm): \(\delta\) 7.32 (t, \(J = 7.9\) Hz, 1H), 7.17 (t, \(J = 7.9\) Hz, 1H), 6.93 (dd, \(J = 8.3, 2.5\) Hz, 1H), 6.85 (d, \(J = 7.6\) Hz, 1H), 6.79 (dd, \(J = 8.5, 2.6\) Hz, 1H), 6.77 (s, 1H), 6.62 (d, \(J = 7.6\) Hz, 1H), 6.58 (s, 1H), 5.05 (s, 1H), 4.98 (d, \(J = 14.5\) Hz, 1H), 3.79 (s, 3H), 3.74 (s,
3-(3-Methoxybenzyl)-2-(3-methoxyphenyl)-5,5-dimethylimidazolidin-4-one (2d)  
Light yellow oil (67.4 mg, 99 %), Rf = 0.16 (PE:EA = 1:1); 1H NMR (400 MHz, CDCl3, δ ppm): 7.37–7.27 (m, 1H), 7.23–7.15 (m, 1H), 7.14–7.07 (m, 1H), 6.99–6.88 (m, 2H), 6.88–6.70 (m, 3H), 5.42 (s, 1H), 4.88 (d, J = 14.5 Hz, 1H), 3.79 (d, J = 14.5 Hz, 1H), 3.68 (s, 3H), 3.62 (s, 3H), 2.31 (s, 1H), 1.41 (s, 3H), 1.29 (s, 3H); 13C NMR (100 MHz, CDCl3, δ ppm): 178.0, 157.8, 157.3, 130.3, 130.1, 128.7, 128.7, 128.4, 125.5, 123.7, 120.6, 120.0, 110.7, 109.7, 59.4, 55.1, 54.8, 39.8, 25.1, 24.3; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C20H25N2O3]+: 341.1860, found: 341.1861.

3-(4-Fluorobenzyl)-2-(4-fluorophenyl)-5,5-dimethylimidazolidin-4-one (2e)  
Colorless or light yellow oil (62.6 mg, 99 %), Rf = 0.44 (PE:EA = 1:1); 1H NMR (400 MHz, CDCl3, δ ppm): 7.26–7.18 (m, 2H), 7.13–7.04 (m, 2H), 7.02–6.91 (m, 4H), 5.04 (s, 1H), 4.89 (d, J = 14.6 Hz, 1H), 3.55 (d, J = 14.6 Hz, 1H), 1.86 (s, 1H), 1.45 (s, 3H), 1.27 (s, 3H); 13C NMR (100 MHz, CDCl3, δ ppm): 178.0, 163.2 (d, J = 248.0 Hz), 162.2 (d, J = 245.0 Hz), 133.8 (d, J = 3.2 Hz), 131.7 (d, J = 3.3 Hz), 123.0 (d, J = 8.1 Hz), 129.2 (d, J = 8.4 Hz), 116.1 (d, J = 21.7 Hz), 115.5 (d, J = 21.4 Hz), 72.5, 59.3, 43.7, 25.7, 24.3; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C18H19F2N2O]+: 317,1460, found: 317,1459.

3-(3-Chlorobenzyl)-2-(3-chlorophenyl)-5,5-dimethylimidazolidin-4-one (2f)  
Colorless oil (64.3 mg, 92 %), Rf = 0.26 (PE:EA = 1:1); 1H NMR (400 MHz, CDCl3, δ ppm): 7.40–7.29 (m, 2H), 7.26–7.17 (m, 3H), 7.12 (d, J = 7.3 Hz, 1H), 6.98 (s, 1H),

S15
6.92 (d, J = 7.2 Hz, 1H), 5.07 (s, 1H), 4.85 (d, J = 14.8 Hz, 1H), 3.62 (d, J = 14.7 Hz, 1H), 1.91 (s, 1H), 1.46 (s, 3H), 1.30 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$, δ ppm): 177.9, 140.2, 137.8, 135.1, 134.4, 130.3, 129.9, 128.8, 128.3, 128.0, 127.4, 126.3, 125.6, 72.7, 59.2, 43.9, 25.9, 24.6; HRMS (ESI-TOF) m/z: [M+H]$^+$ calcd for [C$_{18}$H$_{19}$Cl$_2$N$_2$O]$^+$: 349.0869, found: 349.0872.

3-(2-Bromobenzyl)-2-(2-bromophenyl)-5, 5-dimethylimidazolidin-4-one (2g)
Colorless oil (85.9 mg, 98 %), R$_f$ = 0.49 (PE:EA = 5:1); $^1$H NMR (300 MHz, CDCl$_3$, δ ppm): 7.58–7.44 (m, 2H), 7.40–7.30 (m, 1H), 7.26–7.06 (m, 4H), 7.01–6.92 (m, 1H), 5.56 (s, 1H), 5.03 (d, J = 14.8 Hz, 1H), 3.95 (d, J = 14.8 Hz, 1H), 2.00 (s, 1H), 1.42 (s, 3H), 1.38 (s, 3H); $^{13}$C NMR (75 MHz, CDCl$_3$, δ ppm): 177.8, 134.4, 133.6, 132.9, 130.6, 130.4, 129.3, 128.1, 127.5, 123.9, 72.2, 59.3, 44.7, 25.2; HRMS (ESI-TOF) m/z: [M+H]$^+$ calcd for [C$_{18}$H$_{19}$Br$_2$N$_2$O]$^+$: 438.9838, found: 438.9839.

3-(4-Fluorobenzyl)-2-(4-methoxyphenyl)-5, 5-dimethylimidazolidin-4-one (2h-1)
2-(4-fluorophenyl)-3-(4-methoxybenzyl)-5, 5-dimethylimidazolidin-4-one (2h-2)
Colorless or light yellow oil (64.4 mg, 98 %), R$_f$ = 0.22 (PE:EA = 1:1), $^1$H NMR (400 MHz, CDCl$_3$, δ ppm): 7.25–7.20 (m, 2H), 7.18–7.04 (m, 4H), 7.01–6.88 (m, 8H), 6.82–6.75 (m, 2H), 5.03 (s, 1H), 5.00 (s, 1H), 4.93 (d, J=14.7 Hz, 1H), 4.88 (d, J =14.7 Hz, 1H), 3.83 (s, 3H), 3.78 (s, 3H), 3.55 (d, J=14.5 Hz, 1H), 3.46 (d, J =14.4 Hz, 1H), 1.92 (s, 2H), 1.45 (d, J = 1.6 Hz, 6H), 1.26 (d, J = 2.0 Hz, 6H); $^{13}$C NMR (100 MHz, CDCl$_3$, δ ppm):178.1, 178.0, 163.2 (d, J=247.0 Hz), 162.2 (d, J =245.0 Hz), 160.4, 159.0, 134.0, 134.0, 131.9, 131.9, 130.0, 130.0, 129.7, 129.5, 129.2, 128.6, 127.8, 116.1, 115.9, 115.5, 115.3, 114.4, 113.9, 72.7, 72.3, 59.4, 59.3, 55.3, 55.2, 43.8, 43.6, 25.6, 25.5, 24.3, 24.2; HRMS (ESI-TOF) m/z: [M+H]$^+$ calcd for [C$_{19}$H$_{22}$FN$_2$O$_2$]$^+$: 329.1660, found: 329.1660.
5, 5-Dimethyl-2-(pyridin-2-yl)-3-(pyridin-2-ylmethyl) imidazolidin-4-one (2i)

Yellow oil (40.1 mg, 71%), Rf = 0.31 (DCM: MeOH = 20:1); 1H NMR (400 MHz, CDCl3, δ ppm): 8.58 (d, J = 4.8 Hz, 1H), 8.44 (d, J = 4.8 Hz, 1H), 7.66 (t, J = 7.5 Hz, 1H), 7.58 (t, J = 7.7 Hz, 1H), 7.33-7.22 (m, 2H), 7.12 (d, J = 5.8 Hz, 2H), 5.48 (s, 1H), 4.80 (d, J = 15.7 Hz, 1H), 3.97 (d, J = 15.7 Hz, 1H), 2.95 (s, 1H), 1.53 (d, J = 1.8 Hz, 3H), 1.36 (d, J = 1.8 Hz, 3H); 13C NMR (100 MHz, CDCl3, δ ppm): 178.4, 156.0, 155.0, 149.9, 148.9, 136.7, 136.5, 124.1, 124.1, 122.1, 122.0, 73.8, 59.3, 46.0, 24.8, 24.3; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C16H19N4O]+: 283.1553, found: 283.1554.

5, 5-Dimethyl-2, 3-diphenylimidazolidin-4-one (2j)

Light yellow oil (27.7 mg, 52 %), Rf = 0.32 (PE:EA = 1:1); 1H NMR (300 MHz, CDCl3, δ ppm): 7.38–7.28 (m, 7H), 7.27–7.19 (m, 2H), 7.10–7.02 (m, 1H), 5.98 (s, 1H), 1.98 (s, 1H), 1.49 (s, 3H), 1.40 (s, 3H); 13C NMR (100 MHz, CDCl3, δ ppm): 177.8, 138.4, 137.2, 129.1, 129.0, 128.6, 125.0, 121.8, 74.8, 60.2, 25.5, 24.2; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C17H19N2O]+: 267.1492, found: 267.1497.

5, 5-Dimethyl-2, 3-diphenylimidazolidin-4-one-1-d (2j-d-1)
5, 5-Dimethyl-2, 3-diphenylimidazolidin-4-one-2-d (2j-d-2)

Light yellow oil (22.5 mg, 42%), Rf = 0.36 (PE:EA = 1:1); 1H NMR (600 MHz, CDCl3, δ ppm): 7.37–7.28 (m, 7H), 7.26–7.22 (m, 2H), 7.08–7.04 (m, 1H), 5.98 (s, 0.20H) [C-H], 1.98 (s, 0.82H) [B N-H], 1.49 (s, 3H), 1.40 (s, 3H); 13C NMR (150 MHz, CDCl3, δ ppm): 177.9, 138.5, 137.3, 129.1, 129.1, 128.7, 126.8, 125.0, 121.9, 75.0, 60.2, 25.6, 24.3.
5, 5-Dimethyl-3-phenyl-2-(p-tolyl) imidazolidin-4-one (2k)
Light yellow oil (28.0 mg, 50 %), \( R_f = 0.35 \) (PE:EA = 1:1); \(^1\)H NMR (400 MHz, CDCl\(_3\), \( \delta \) ppm): 7.33 (d, \( J = 8.5 \) Hz, 2H), 7.26–7.19 (m, 4H), 7.11 (d, \( J = 7.9 \) Hz, 2H), 7.04 (t, \( J = 7.4 \) Hz, 1H), 5.94 (s, 1H), 2.28 (s, 3H), 1.94 (s, 1H), 1.48 (s, 3H), 1.39 (s, 3H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\), \( \delta \) ppm): 177.8, 138.9, 137.3, 135.5, 129.7, 128.6, 126.6, 124.9, 121.9, 74.7, 60.1, 25.6, 24.2, 21.1; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for [C\(_{18}\)H\(_{21}\)N\(_2\)O]: 281.1648, found: 281.1652.

2-(4-Fluorophenyl)-5, 5-dimethyl-3-phenylimidazolidin-4-one (2l)
Colorless oil (27.3 mg, 48 %), \( R_f = 0.34 \) (PE:EA = 1:1); \(^1\)H NMR (400 MHz, CDCl\(_3\), \( \delta \) ppm): 7.35–7.28 (m, 4H), 7.27–7.22 (m, 2H), 7.10–7.04 (m, 1H), 7.00 (t, \( J = 8.6 \) Hz, 2H), 5.97 (s, 1H), 1.95 (d, \( J = 9.1 \) Hz, 1H), 1.48 (s, 3H), 1.39 (s, 3H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\), \( \delta \) ppm): 177.6, 162.9 (d, \( J = 248.3 \) Hz), 137.1, 134.4, 134.4, 128.7, 128.7, 128.6, 125.2, 122.0, 116.2, 116.0, 74.2, 60.2, 25.6, 24.2; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for [C\(_{17}\)H\(_{18}\)FN\(_2\)O]: 285.1398, found: 285.1403.

3, 5, 5-Trimethyl-2-phenylimidazolidin-4-one (2m-1)
Light yellow oil (8.2 mg, 20 %), \( R_f = 0.18 \) (PE:EA = 1:1); \(^1\)H NMR (600 MHz, CDCl\(_3\), \( \delta \) ppm): 7.46–7.39 (m, 3H), 7.36–7.32 (m, 2H), 5.23 (s, 1H), 2.65 (s, 3H), 1.87 (s, 1H), 1.42 (s, 3H), 1.32 (s, 3H); \(^{13}\)C NMR (150 MHz, CDCl\(_3\), \( \delta \) ppm): 178.4, 138.5, 129.5, 129.2, 126.9, 77.2, 77.0, 76.8, 75.9, 59.5, 27.6, 26.0, 24.6; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for [C\(_{12}\)H\(_{17}\)N\(_2\)O]: 205.1335, found: 205.1335.

3-Benzyl-5, 5-dimethylimidazolidin-4-one (2m-2)
Light yellow oil (16.3 mg, 40 %), \( R_f = 0.08 \) (PE:EA = 1:1); \(^1\)H NMR (600 MHz, CDCl\(_3\), \( \delta \) ppm): 7.35–7.31 (m, 2H), 7.30–7.26 (m, 1H), 7.24–7.21 (m, 2H), 4.14 (s, 2H), 2.27 (s, 2H), 2.11 (s, 1H), 1.30 (s, 6H); \(^{13}\)C NMR (150 MHz, CDCl\(_3\), \( \delta \) ppm): 177.6, 136.0, 128.6, 127.8, 127.6, 60.4, 59.3, 45.4, 23.8; HRMS (ESI-TOF) m/z: [M+H]\(^+\) calcd for [C\(_{12}\)H\(_{17}\)N\(_2\)O]: 205.1335, found: 205.1335.
3-Benzyl-2, 5, 5-trimethyl-2-phenylimidazolidin-4-one (2n)
Colorless or light yellow oil (52.4 mg, 89 %), R_f = 0.41 (PE:EA = 1:1), ¹H NMR (400 MHz, CDCl₃, δ ppm): 7.41–7.30 (m, 5H), 7.27–7.20 (m, 3H), 7.20–7.14 (m, 2H), 4.96 (d, J = 15.3 Hz, 1H), 3.78 (d, J = 15.3 Hz, 1H), 2.24 (s, 1H), 1.58 (s, 3H), 1.47 (s, 3H), 1.32 (s, 3H); ¹³C NMR (100 MHz, CDCl₃, δ ppm): 177.8, 143.7, 138.1, 128.8, 128.4, 128.2, 127.8, 127.2, 125.8, 78.5, 58.8, 44.8, 28.5, 28.1, 26.5; HRMS (ESI-TOF) m/z: [M+H]⁺ calcd for [C₁₉H₂₃N₂O]⁺: 295.1805, found: 295.1805.

2-Benzyl-5, 5-dimethyl-3-phenethylimidazolidin-4-one (2o)
Yellow oil (41.3 mg, 67 %), R_f = 0.33 (PE:EA = 1:1); ¹H NMR (400 MHz, CDCl₃, δ ppm): 7.34–7.15 (m, 8H), 7.14–7.08 (m, 2H), 4.47 (dd, J = 6.4, 4.0 Hz, 1H), 4.00 (ddd, J = 13.9, 8.0, 6.2 Hz, 1H), 3.27 (ddd, J = 14.0, 7.9, 6.2 Hz, 1H), 3.03–2.68 (m, 4H), 1.72 (s, 1H), 1.14 (s, 3H), 1.04 (s, 3H); ¹³C NMR (100 MHz, CDCl₃, δ ppm): 177.9, 138.3, 135.1, 129.4, 128.7, 128.6, 128.5, 127.0, 126.5, 71.0, 58.6, 41.7, 39.9, 33.5, 25.6, 24.7; HRMS (ESI-TOF) m/z: [M+H]⁺ calcd for [C₂₀H₂₅N₂O]⁺: 309.1961, found: 309.1962.

tert-Butyl 2,2-dimethyl-3-oxohexahydroimidazo[1,2-a]pyridine-1(5H)-carboxylate (2p’)
Colorless or light yellow oil (45.1 mg, 84 %), R_f = 0.33 (PE:EA = 3:1); ¹H NMR (400 MHz, CDCl₃, δ ppm): 4.85–4.69,(m, 1H), 4.32–4.20 (m, 1H), 2.85–2.69 (m, 1H), 2.68–2.39 (m, 1H), 1.96–1.85 (m, 1H), 1.74–1.63 (m, 1H), 1.62–1.45 (m, 16H), 1.43–1.29 (m, 1H), 1.18–0.93 (m, 1H); ¹³C NMR (100 MHz, CDCl₃, δ ppm): 171.4, 153.0, 80.6, 70.3, 60.6, 39.8, 32.9, 28.4, 25.0, 24.4, 24.1, 22.2; HRMS (ESI-TOF) m/z: [M+H]⁺ calcd for [C₁₄H₂₅N₂O₃]⁺: 269.1860, found: 269.1867.
**tert-Butyl-2,2-dimethyl-3-oxohexahydro-1H-pyrrolo[1,2-a]imidazole-1-carboxylate (2q')**

Colorless or light yellow oil (16.8 mg, 33 %), R<sub>f</sub> = 0.28 (PE:EA = 3:1); \(^1\)H NMR (400 MHz, CDCl<sub>3</sub>, \(\delta\) ppm): 5.11–4.97 (m, 1H), 3.77–3.66 (m, 1H), 3.17–3.09 (m, 1H), 2.51–2.27 (m, 1H), 2.17–2.06 (m, 1H), 2.06–1.92 (m, 1H), 1.58–1.45 (m, 15H), 1.42–1.24 (m, 1H); \(^13\)C NMR (150 MHz, CDCl<sub>3</sub>, \(\delta\) ppm): 176.1, 153.5, 74.4, 41.4, 33.3, 32.6, 28.4, 25.0, 24.0, 23.8, 22.6; HRMS (ESI-TOF) m/z: [M+H]<sup>+</sup> calcd for [C<sub>13</sub>H<sub>23</sub>N<sub>2</sub>O<sub>3</sub>]<sup>+</sup>: 255.1703, found: 255.1710.

![Structure](image)

**7-Benzyl-6-phenyl-5, 7-diazaspiro[3.4]octan-8-one (2r)**

Yellow oil (46.2 %, 79 %), R<sub>f</sub> = 0.56 (PE:EA = 1:1); \(^1\)H NMR (300 MHz, CDCl<sub>3</sub>, \(\delta\) ppm): 7.40–7.34 (m, 3H), 7.29–7.22 (m, 3H), 7.21–7.15 (m, 2H), 7.09–7.02 (m, 2H), 5.10 (s, 1H), 4.99 (d, \(J = 14.7\) Hz, 1H), 3.50 (d, \(J = 14.7\) Hz, 1H), 2.78–2.66 (m, 1H), 2.62–2.50 (m, 1H), 2.21 (s, 1H), 2.17–2.05 (m, 3H), 1.93–1.76 (m, 1H); \(^13\)C NMR (75 MHz, CDCl<sub>3</sub>, \(\delta\) ppm): 176.5, 139.0, 136.0, 129.2, 129.0, 128.5, 128.2, 127.5, 127.0, 73.8, 62.6, 43.9, 34.6, 34.1, 13.8; HRMS (ESI-TOF) m/z: [M+H]<sup>+</sup> calcd for [C<sub>19</sub>H<sub>21</sub>N<sub>2</sub>O]<sup>+</sup>: 293.1648, found: 293.1650.

![Structure](image)

**3-Benzyl-2-phenyl-1, 3-diazaspiro[4.4]nonan-4-one (2s)**

Colorless oil (57.6 mg, 94 %), R<sub>f</sub> = 0.67 (PE:EA = 1:1); \(^1\)H NMR (300 MHz, CDCl<sub>3</sub>, \(\delta\) ppm): 7.41–7.35 (m, 3H), 7.28–7.20 (m, 5H), 7.05–6.98 (m, 2H), 5.01 (d, \(J = 14.3\) Hz, 2H), 3.56 (d, \(J = 14.6\) Hz, 1H), 2.48–2.33 (m, 1H), 1.93–1.67 (m, 7H), 1.63–1.50 (m, 1H); \(^13\)C NMR (75 MHz, CDCl<sub>3</sub>, \(\delta\) ppm): 178.48, 138.13, 136.0, 129.4, 129.0, 128.5, 128.2, 127.5, 127.0, 73.8, 62.6, 43.9, 34.6, 34.1, 13.8; HRMS (ESI-TOF) m/z: [M+H]<sup>+</sup> calcd for [C<sub>20</sub>H<sub>23</sub>N<sub>2</sub>O]<sup>+</sup>: 307.1805, found: 307.1807.

![Structure](image)

**((2R*, 5R*)-3-Benzyl-5-methyl-2-phenylimidazolidin-4-one and (2R*,5S*)-3-benzyl-5-methyl-2-phenylimidazolidin-4-one (2t)**

Colorless oil (38.4 mg, 72 %), R<sub>f</sub> = 0.67 (PE:EA = 1:1); dr = 1:1; \(^1\)H NMR (400 MHz, CDCl<sub>3</sub>, \(\delta\) ppm): 7.41–7.35 (m, 3H), 7.28–7.20 (m, 5H), 7.05–6.98 (m, 2H), 5.01 (d, \(J = 14.3\) Hz, 2H), 3.56 (d, \(J = 14.6\) Hz, 1H), 2.48–2.33 (m, 1H), 1.93–1.67 (m, 7H), 1.63–1.50 (m, 1H); \(^13\)C NMR (75 MHz, CDCl<sub>3</sub>, \(\delta\) ppm): 178.48, 138.13, 136.0, 129.4, 129.0, 128.5, 128.2, 127.5, 127.0, 73.8, 62.6, 43.9, 34.6, 34.1, 13.8; HRMS (ESI-TOF) m/z: [M+H]<sup>+</sup> calcd for [C<sub>20</sub>H<sub>23</sub>N<sub>2</sub>O]<sup>+</sup>: 307.1805, found: 307.1807.
CDCl₃, δ ppm): (A): 7.45 – 7.38 (m, 3H), 7.33 – 7.22 (m, 5H), 7.12 – 7.07 (m, 1H), 7.07 – 7.01 (m, 1H), 5.21 (s, 1H), 5.07 (d, J = 14.7 Hz, 1H), 3.93 (q, J = 6.9 Hz, 1H), 3.57 (t, J = 14.7 Hz, 2H), 2.82 (s, 1H), 1.49 (d, J = 6.8 Hz, 3H); (B): 7.45 – 7.38 (m, 3H), 7.33 – 7.22 (m, 5H), 7.12 – 7.07 (m, 1H), 7.07 – 7.01 (m, 1H), 5.15 (s, 1H), 4.98 (d, J = 14.6 Hz, 1H), 3.67 (q, J = 6.8 Hz, 1H), 2.82 (s, 1H), 1.40 (d, J = 6.9 Hz, 3H);

13C NMR (75 MHz, CDCl₃, δ ppm): 175.6, 175.2, 138.4, 138.0, 135.8, 135.7, 129.6, 129.4, 129.1, 128.9, 128.6, 128.5, 128.4, 128.2, 127.7, 127.6, 127.5, 126.9, 74.8, 74.3, 55.3, 54.2, 44.4, 44.1, 18.0, 17.9; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C₁₇H₁₉N₂O]+: 267.1492, found: 267.1495.

(2R*, 5R*)-3-benzyl-2,5-diphenylimidazolidin-4-one and (2S*, 5R*)-3-Benzyl-2, 5-diphenylimidazolidin-4-one (2u)
Yellow oil (51.9 mg, 79 %), Rᵣ = 0.67 (PE:EA = 1:1); dr = 1: 0.7; ¹H NMR (300 MHz, CDCl₃, δ ppm): (A) 7.56-7.52 (m, 2H), 7.43-7.20 (m, 11H), 7.11-7.04 (m, 2H), 5.28 (d, J = 1.9, 1H), 4.98 (d, J = 14.6 Hz, 1H), 4.72 (d, J = 1.9 Hz, 1H), 3.60 (d, J = 14.7 Hz, 1H), 2.41 (s, 1H); (B) 7.49-7.44 (m, 2H), 7.43-7.20 (m, 11H), 7.04-6.98 (m, 2H), 5.36 (d, J = 1.8 Hz, 1H), 5.05 (d, J = 14.7 Hz, 1H), 4.93 (s, 1H), 3.55 (d, J = 14.7 Hz, 1H), 2.41 (s, 1H); ¹³C NMR (75 MHz, CDCl₃, δ ppm): 172.9, 172.8, 139.2, 139.0, 138.4, 138.1, 135.9, 135.7, 129.5, 129.4, 129.1, 128.9, 128.6, 128.6, 128.5, 128.2, 128.0, 127.8, 127.6, 127.0, 126.9, 75.3, 74.4, 63.1, 62.2, 44.5, 44.2; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C₂₂H₂₁N₂O]+: 329.1648, found: 329.1650.

5, 5-Dimethyl-3-phenyl-2-(2-phenylcyclopropyl) imidazolidin-4-one (2w)
Colorless oil (27.0 mg, 44 %), Rᵣ = 0.21 (PE:EA = 1:1); ¹H NMR (300 MHz, CDCl₃, δ ppm): 7.46-7.38 (m, 2H), 7.35-7.27 (m, 3H), 7.25-7.19 (m, 2H), 7.18-7.12 (m, 1H), 7.03-6.98 (m, 2H), 4.53 (d, J = 7.6 Hz, 1H), 2.08-1.98 (m, 2H (NH and CH)), 1.45 (s, 3H), 1.34 (s, 3H), 1.21-1.10 (m, 1H), 1.00-0.83 (m, 2H); ¹³C NMR 100 MHz, CDCl₃, δ ppm): 177.3, 140.5, 136.9, 129.2, 128.0, 126.8, 126.0, 125.9, 125.8, 76.9, 59.4, 27.3, 26.1, 24.7, 22.5, 10.0; HRMS (ESI-TOF) m/z: [M+H]+ calcd for [C₂₀H₂₃N₂O]+: 307.1806, found: 307.1805.
**tert-Butyl 2-phenylpyrrolidine-1-carboxylate (5)**

Yellow liquid (A & B, 44.0 mg, 89 %); R_f = 0.21 (PE:EA = 20:1); A:B = 2:1; (A): 1H NMR (400 MHz, CDCl_3, δ ppm): 7.35–7.27 (m, 2H), 7.26–7.14 (m, 3H), 4.78 (s, 1H), 3.72–3.46 (m, 2H), 2.41–2.20 (m, 1H), 2.00–1.78 (m, 3H), 1.20 (s, 9H); 13C NMR 100 MHz, CDCl_3, δ ppm): 154.5, 145.1, 128.3, 128.0, 126.4, 79.1, 61.2, 47.0, 36.0, 28.4, 28.1, 23.1. (B): 1H NMR (400 MHz, CDCl_3, δ ppm): 7.35–7.27 (m, 2H), 7.26–7.14 (m, 3H), 4.98 (s, 1H), 3.72–3.46 (m, 2H), 2.41–2.20 (m, 1H), 2.00–1.78 (m, 3H), 1.47 (s, 9H). 13C NMR 100 MHz, CDCl_3, δ ppm): 154.5, 145.1, 128.2, 126.4, 125.2, 79.1, 60.6, 47.2, 34.8, 28.3, 23.3. HRMS (ESI-TOF) m/z: [M+H]^+ calcd for [C_{15}H_{22}NO_2]^+: 248.1645, found: 248.1648.

**tert-Butyl 2-phenyloxazolidine-3-carboxylate (6)**

Yellow liquid (38.9 mg, 78 %); R_f = 0.33 (PE: EA = 10:1); 1H NMR (400 MHz, CDCl_3, δ ppm): 7.48–7.30 (m, 5H), 5.98 (s, 1H), 4.18–4.08 (m, 1H), 4.07–3.99 (m, 1H), 3.85 (s, 1H), 3.58 (s, 1H), 1.30 (s, 9H). 13C NMR 100 MHz, CDCl_3, δ ppm): 153.1, 139.6, 128.3, 128.2, 126.5, 89.1, 80.4, 65.7, 44.9, 28.2. HRMS (ESI-TOF) m/z: [M+H]^+ calcd for [C_{14}H_{20}NO_3]^+: 250.1440, found: 250.1438.
Crystallographic data for compound 2a

The single crystal of 2a was collected through mixed solvent recrystallization with dichloromethane and petroleum Ether, and the collected crystal 2a was colorless and transparent. A suitable crystal was selected and the measurement was carried on a SuperNova, Eos diffractometer. The data for compound 2a were collected on Brookhaven BI-200SM. The crystal was kept at 295.62(10) K during data collection. Using Olex2,8 the structure was solved with the ShelXS9 structure solution program using Direct Methods and refined with the ShelXL10 refinement package using Least Squares minimization.

Molecular Structure of 2a
Crystal Data for C18H20N2O (M =280.36 g/mol): monoclinic, space group P21/n (no. 14), \(a = 17.1267(9) \text{ Å}, b = 5.5794(3) \text{ Å}, c = 17.3458(8) \text{ Å}, \beta = 110.422(6)^\circ, V = 1553.33(13) \text{ Å}^3, Z = 4, T = 295.62(10) \text{ K}, \mu(\text{CuK} \alpha) = 0.587 \text{ mm}^{-1},\) \(D_{\text{alc}} = 1.199 \text{ g/cm}^3,\) 5180 reflections measured \((11.02^\circ \leq 2\Theta \leq 133.02^\circ),\) 2668 unique \((R_{\text{int}} = 0.0183, R_{\text{sigma}} = 0.0235)\) which were used in all calculations. The final \(R_1\) was 0.0495 \(( > 2\text{sigma}(I))\) and \(wR_2\) was 0.1419 (all data). CCDC-1872309 contains the supplementary crystallographic data for this paper. These data can be obtained free from \url{www.ccdc.cam.ac.uk} (CCDC No. 1872309).
| **Table S1. Crystal data and structure refinement of 2a** |
|-----------------------------------------------------------|
| **Empirical formula** | C$_{18}$H$_{20}$N$_2$O                  |
| **Formula weight**   | 280.36                                   |
| **Temperature/K**    | 295.62(10)                               |
| **Crystal system**    | monoclinic                               |
| **Space group**       | P2$_1$/n                                 |
| **a/Å**               | 17.1267(9)                               |
| **b/Å**               | 5.5794(3)                                |
| **c/Å**               | 17.3458(8)                               |
| **α/°**               | 90.00                                    |
| **β/°**               | 110.422(6)                               |
| **γ/°**               | 90.00                                    |
| **Volume/Å$^3$**      | 1553.33(13)                              |
| **Z**                 | 4                                        |
| **ρ$_{calc}$/g/cm$^3$** | 1.199                                  |
| **μ/mm$^{-1}$**       | 0.587                                    |
| **F(000)**            | 600.0                                    |
| **Crystal size/mm$^3$** | 0.17 × 0.14 × 0.12                  |
| **Radiation**         | CuKα ($\lambda = 1.54184$)              |
| **2Θ range for data collection/°** | 11.02 to 133.02                        |
| **Index ranges**      | -15 ≤ h ≤ 20, -5 ≤ k ≤ 6, -20 ≤ l ≤ 14 |
| **Reflections collected** | 5180                                 |
| **Independent reflections** | 2668 [R$_{int}$ = 0.0183, R$_{sigma}$ = 0.0235] |
| **Data/restraints/parameters** | 2668/0/196                             |
| **Goodness-of-fit on F$^2$** | 1.053                                  |
| **Final R indexes [I>=2σ (I)]** | R$_1$ = 0.0495, wR$_2$ = 0.1321                |
| **Final R indexes [all data]** | R$_1$ = 0.0583, wR$_2$ = 0.1419                |
| **Largest diff. peak/hole / e Å$^{-3}$** | 0.17/-0.15                     |
Table S2. Fractional atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($\text{Å}^2 \times 10^3$) for 2a. $U_{eq}$ is defined as 1/3 of the trace of the orthogonalised $U_{ij}$ tensor.

| Atom | x     | y     | z     | U(eq) |
|------|-------|-------|-------|-------|
| N1   | 8030.1(8) | 2111(2) | 5399.8(8) | 47.2(3) |
| O1   | 7389.9(8) | 4831(2) | 4390.0(8) | 67.8(4) |
| C2   | 9228.6(9)  | -108(3) | 6360.9(10) | 45.1(4) |
| C12  | 7547.3(10) | 2585(3) | 5927.1(11) | 52.5(4) |
| C7   | 9819.7(11) | 1679(3) | 6644.4(12) | 58.0(5) |
| C8   | 8389.2(10) | 1903(3) | 4229.8(10) | 49.7(4) |
| N2   | 8949.2(11) | 360(3)  | 4872.4(10) | 64.2(5) |
| C13  | 6923.0(9)  | 640(3)  | 5892.4(10) | 48.4(4) |
| C11  | 7871.4(10) | 3163(3) | 4663.0(10) | 47.9(4) |
| C1   | 8579.7(10) | 23(3)  | 5511.2(10) | 47.7(4) |
| C6   | 10431.3(11)| 1516(4) | 7407.3(13) | 69.1(6) |
| C3   | 9259.7(12) | -2027(3)| 6870.7(11) | 57.5(5) |
| C17  | 6272.0(13) | -2050(4)| 6565.1(13) | 74.9(6) |
| C4   | 9873.1(14) | -2178(4)| 7636.8(12) | 69.9(5) |
| C18  | 6852.5(12) | -294(4) | 6597.5(11) | 66.4(5) |
| C5   | 10467.2(12)| -432(4) | 7905.4(12) | 68.9(6) |
| C9   | 8877.4(14) | 3687(4) | 3920.3(15) | 77.2(6) |
| C10  | 7810.5(13) | 430(4)  | 3520.8(12) | 70.1(5) |
| C16  | 5766.7(16) | -2884(5)| 5833.3(15) | 89.6(8) |
| C14  | 6403.6(15) | -211(5) | 5156.9(12) | 96.2(9) |
| C15  | 5828(2)    | -1947(7)| 5126.2(16) | 131.2(14) |
Figure S1 Molecular structure of 2a. Hydrogen and solvent molecules are omitted for clarity. Thermal ellipsoids are drawn with 50% probability.
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NMR spectra of products

1a
1b
S32
1h
2h-1 & 2h-2
2m-1
$2n$
2p'
NOE Data of 2u:
2w
