Supporting Information

1,2-Alkylarylation of Activated Alkenes with Two C-H Bonds using Visible Light Catalysis

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**A** Typical Experimental Procedure

(a) Synthesis of Substrates 1:

Substrates 1 were prepared according to the known procedures.\textsuperscript{[S1]}

(b) Typical Experimental Procedure for the 1,2-Alkylarylation of Activated Alkenes:

To a Schlenk tube were added N-arylacrylamide 1 (0.3 mmol), 4-MeOC$_6$H$_4$N$_2$BF$_4$ (2 equiv), Ru(bpy)$_3$Cl$_2$ (5 mol%), Na$_2$CO$_3$ (2 equiv), and acetonitrile 2a (1 mL). Then the tube was charged with argon, and was stirred at 50 °C (oil bath temperature) with 36 W compact fluorescent light for the indicated time until complete consumption of starting material as monitored by TLC and/or GC-MS analysis. After the reaction was finished, the reaction mixture was cooled to room temperature, diluted in diethyl ether, and washed with brine. The aqueous phase was re-extracted with diethyl ether. The combined organic extracts were dried over Na$_2$SO$_4$ and concentrated in vacuum, and the resulting residue was purified by silica gel column chromatography (hexane/ethyl acetate = 5:1) to afford the desired product 3.
**Figure S1. Information on the Quantum Yield**

![Chemical Reaction](image)

**As**  UV-Vis absorption of Rhodamine B at 350nm: 0.0682  (red line)

**Ax**  UV-Vis absorption of reaction system at 450nm: 0.3705 (black line)

**Fi Intensity**

**Fs** Integrated area of fluorescence of Rhodamine B: 145800 (red line)

**Fx** Integrated area of fluorescence of reaction system: 87071 (black line)

**ns** Rhodamine B is dissolved in water, refractivity of water is 1.3

**nx** Solvent of reaction system is CH$_3$CN, refractivity of CH$_3$CN is 1.3

**Φs** Quantum yield of Rhodamine B: 0.9

\[ Φx = Φs(nx/ns)^2 (As/Ax)(Fx/Fs) \]

\[ = 0.9 \times (1.3/1.3)^2 \times (0.0682/0.3705) \times (87071/145800) \]

\[ = 0.099 \]
(B) Analytical data

3-(1,3-Dimethyl-2-oxindolin-3-yl)propanenitrile (3a):[S1]

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) δ: 7.34-7.30 (m, 1H), 7.18 (d, $J = 7.2$ Hz 1H), 7.11 (t, $J = 7.6$ Hz, 1H), 6.88 (d, $J = 7.6$ Hz, 1H), 3.22 (s, 3H), 2.34-2.29 (m, 1H), 2.11-1.98 (m, 3H), 1.39 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) δ: 178.8, 143.1, 131.6, 128.6, 123.0, 122.6, 118.8, 108.5, 47.3, 33.4, 26.3, 23.4, 12.8; LRMS (EI 70 ev) m/z (%): 214 (M$^+$, 37), 161 (35), 160 (100).

3-(1-Benzyl-3-methyl-2-oxindolin-3-yl)propanenitrile (3b):

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) δ: 7.30-7.24 (m, 5H), 7.17 (d, $J = 7.6$ Hz, 2H), 7.06 (t, $J = 7.6$ Hz, 1H), 6.77 (d, $J = 7.6$ Hz, 1H), 4.93-4.84 (m, 2H), 2.36 (t, $J = 9.6$ Hz, 1H), 2.11-1.97 (m, 3H), 1.43 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) δ: 179.0, 142.2, 135.7, 131.6, 128.9, 128.6, 127.8, 127.3, 123.1, 122.7, 118.8, 109.5, 47.3, 43.8, 33.5, 23.8, 12.8; LRMS (EI 70 ev) m/z (%): 290 (M$^+$, 52), 236 (26), 91 (100); HRMS (ESI) for C$_{19}$H$_{19}$N$_2$O [M+H]$^+$: calcd. 291.1497, found. 291.1505.

3-(1-Acetyl-3-methyl-2-oxindolin-3-yl)propanenitrile (3c):

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.26 (d, $J = 8.0$ Hz 1H), 7.39-7.34 (m,
1H), 7.29-7.25 (m, 1H), 7.21-7.19 (m, 1H), 2.69 (s, 3H), 2.43-2.37 (m, 1H), 2.15-2.04 (m, 3H), 1.48 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) δ: 179.9, 170.8, 139.4, 130.5, 129.1, 125.7, 122.2, 118.3, 117.0, 47.9, 34.0, 26.7, 24.8, 13.0; LRMS (EI 70 ev) m/z (%): 242 (M+, 5), 200 (73), 146 (100); HRMS (ESI) for C$_{14}$H$_{15}$N$_2$O$_2$ [M+H]$^+$: calcd. 243.1134, found. 243.1142.

3-(1,3,5-Trimethyl-2-oxoindolin-3-yl)propanenitrile (3e):[S1]

Yellow soild, mp 89.7-90.8 °C (uncorrected); $^1$H NMR (400 MHz, CDCl$_3$) δ: 7.11 (d, J = 8.0 Hz, 1H), 7.00 (s, 1H), 6.76 (d, J = 8.0 Hz, 1H), 3.20 (s, 3H), 2.36 (s, 3H), 2.33 (t, J = 8.4 Hz, 1H), 2.08-2.01 (m, 3H), 1.38 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) δ: 178.8, 140.7, 132.7, 131.7, 128.9, 123.4, 118.9, 108.2, 47.4, 33.5, 26.3, 23.5, 21.1, 12.8; LRMS (EI 70 ev) m/z (%): 228 (M+, 37), 174 (100), 175 (28).

3-(1,3,6-Trimethyl-2-oxoindolin-3-yl)propanenitrile and 3-(1,3,7-Trimethyl-2-oxoindolin-3-yl)propanenitrile (3f):[S1]

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) δ: 7.21 (t, J = 8.0 Hz, 0.6H), 7.06 (d, J = 7.6 Hz, 0.4H), 6.92 (d, J = 7.6 Hz, 0.4H), 6.87 (d, J = 7.6 Hz, 0.6H), 6.73 (s, 0.4H), 6.70 (d, J = 4.0 Hz, 0.6H), 3.21 (s, 1.8H), 3.20 (s, 1.2H), 2.39 (s, 1.2H), 2.38 (s, 1.8H), 2.31-2.26 (m, 1H), 2.07-1.90 (m, 3H), 1.47 (s, 1.8H), 1.37 (s, 1.2H); $^{13}$C NMR (100 MHz, CDCl$_3$) δ: 179.2, 178.8, 143.4, 143.2, 138.8, 134.4, 128.6, 128.5, 128.1, 125.5,
123.5, 122.3, 118.8, 118.6, 109.4, 106.2, 48.3, 47.1, 33.4, 31.4, 29.6, 26.4, 26.2, 23.5, 21.8, 18.1, 13.0, 12.8; LRMS (EI 70 ev) m/z (%): 229 (M+1, 5), 228 (M+, 38), 174 (100); LRMS (EI 70 ev) m/z (%): 228 (M+, 38), 174 (100).

3-(1,3,7-Trimethyl-2-oxoindolin-3-yl)propanenitrile (3g):[S1]

Brown oil; ¹H NMR (400 MHz, CDCl₃) δ: 7.05-6.97 (m, 3H), 3.50 (s, 3H), 2.59 (s, 3H), 2.34-2.28 (m, 1H), 2.09-1.95 (m, 1H), 1.37 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ: 179.6, 140.9, 132.3, 123.0, 120.4, 120.2, 118.9, 46.6, 33.7, 29.6, 23.9, 19.0, 12.8; LRMS (EI 70 ev) m/z (%): 229 (M+1, 6), 228 (M+, 38), 174 (100).

3-(5-Butyl-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3h):

Brown oil; ¹H NMR (400 MHz, CDCl₃) δ: 7.13-7.10 (m, 1H), 7.00 (s, 1H), 6.78 (d, J = 8.0 Hz, 1H), 3.20 (s, 3H), 2.60 (t, J = 8.0 Hz, 2H), 2.33-2.27 (m, 1H), 2.09-2.01 (m, 3H), 1.62-1.56 (m, 2H), 1.38 (s, 3H), 1.25 (t, J = 6.0 Hz, 2H), 0.94 (t, J = 7.6 Hz, 3H); ¹³C NMR (100 MHz, CDCl₃) δ: 178.9, 140.9, 138.0, 131.6, 128.3, 122.7, 118.9, 108.2, 47.4, 35.3, 34.0, 33.5, 26.7, 23.4, 22.3, 13.9, 12.8; LRMS (EI 70 ev) m/z (%): 270 (M+, 27), 227 (100), 228 (16); HRMS (ESI) for C₁₇H₂₃N₂O [M+H]+: calcd. 271.1810, found. 271.1819.
3-(5-Methoxy-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3i):

Yellow soild, mp 89.2-90.7 °C (uncorrected); $^1$H NMR (400 MHz, CDCl$_3$) δ:

6.84-6.76 (m, 3H), 3.80 (s, 3H), 3.19 (s, 3H), 2.34-2.28 (m, 1H), 2.10-1.96 (m, 3H),
1.38 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) δ: 178.5, 156.4, 136.5, 133.0, 118.8, 112.5,
110.2, 108.8, 55.8, 47.7, 33.4, 26.3, 23.5, 12.8; LRMS (EI 70 ev) m/z (%): 244 (M$^+$, 67),
229 (34), 190 (100).

![Chemical Structure](image1.png)

3-(5-Chloro-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3j):

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) δ: 7.31-7.28 (m, 1H), 7.17 (s, 1H), 6.81 (d, $J = 8.4$ Hz, 1H), 3.21 (s, 3H), 2.34-2.30 (m, 1H), 2.11-2.04 (m, 3H), 1.40 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) δ: 178.3, 141.7, 133.4, 128.6, 128.5, 123.2, 118.5, 109.5,
47.5, 33.2, 26.4, 23.4, 12.8; LRMS (EI 70 ev) m/z (%): 250 (M$^+$+2, 16), 248 (M$^+$, 47),
194 (100).

![Chemical Structure](image2.png)

3-(5-Fluoro-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3k):

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) δ: 7.04-6.99 (m, 1H), 6.96-6.93 (m, 1H),
6.82-6.79 (m, 1H), 3.20 (s, 3H), 2.35-2.27 (m, 1H), 2.13-2.00 (m, 3H), 1.39 (s, 3H);
$^{13}$C NMR (100 MHz, CDCl$_3$) δ: 178.4, 159.5 (d, $J = 240.5$ Hz, 1C), 139.0, 133.3 (d, $J = 7.8$ Hz, 1C), 118.5, 114.9 (d, $J = 23.3$ Hz, 1C), 111.9 (d, $J = 24.5$ Hz, 1C), 109.1 (d, $J = 8.1$ Hz, 1C), 47.7, 33.2, 26.4, 23.4, 12.8; LRMS (EI 70 ev) m/z (%): 232 (M$^+$, 47),

![Chemical Structure](image3.png)
3-(1,3-Dimethyl-2-oxo-5-(trifluoromethyl)indolin-3-yl)propanenitrile (3l):[S1]

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) $\delta$: 7.61 (d, $J = 8.0$ Hz, 1H), 7.42 (s, 1H), 6.96 (d, $J = 8.4$ Hz, 1H), 3.25 (s, 3H), 2.37-2.31 (m, 1H), 2.13-2.06 (m, 3H), 1.43 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$: 178.8, 146.1, 132.3, 126.5 (d, $J = 3.9$ Hz, 1C), 125.5 (t, $J = 18.8$ Hz, 1C), 125.1, 122.8, 119.7 (d, $J = 3.6$ Hz, 1C), 118.3, 108.3, 47.2, 33.1, 26.5, 23.3, 12.8; LRMS (EI 70 ev) $m/z$ (%): 301 (M$^+$, 6), 222 (67), 103 (100).

3-(1,3-Dimethyl-7-(methylthio)-2-oxoindolin-3-yl)propanenitrile (3m):

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) $\delta$: 7.24 (s, 1H), 7.07-6.99 (m, 2H), 3.64 (s, 3H), 2.46 (s, 3H), 2.33-2.26 (m, 1H), 2.12-1.94 (m, 3H), 1.36 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$: 179.5, 141.7, 133.0, 131.2, 123.6, 120.7, 120.3, 118.8, 46.7, 33.7, 30.2, 23.8, 18.8, 12.8; LRMS (EI 70 ev) $m/z$ (%): 260 (M$^+$, 85), 206 (100); HRMS (ESI) for C$_{14}$H$_{17}$N$_2$OS [M+H]$^+$: calcd. 261.1062, found. 261.1073.

3-(7-Chloro-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3n):

Brown oil, $^1$H NMR (400 MHz, CDCl$_3$) $\delta$: 7.25-7.22 (m, 1H), 7.08-7.06 (m, 1H), 7.04-7.00 (m, 1H), 3.58 (s, 3H), 2.36-2.28 (m, 1H), 2.11-2.00 (m, 3H), 1.39 (s, 3H);
13C NMR (100 MHz, CDCl3) δ: 179.0, 139.0, 134.5, 131.0, 123.8, 121.1, 118.5, 116.0, 47.0, 33.5, 29.6, 23.8, 12.8; LRMS (EI 70 ev) m/z (%): 250 (M+2, 2), 248 (M+, 6), 193 (100); HRMS (ESI) for C13H1435ClN2O [M+H]+: calcd. 249.0795, found. 249.0807.

3-(1,3-Dimethyl-2-oxo-1,2,3,5,6,7-hexahydrocyclopenta[f]indol-3-yl)propanenitrile and 3-(1,3-Dimethyl-2-oxo-1,2,3,6,7,8-hexahydrocyclopenta[g]indol-3-yl)propanenitrile (3o):

Brown oil; 1H NMR (400 MHz, CDCl3) δ: 7.17 (d, J = 8.0 Hz, 0.6H), 7.04 (s, 0.4H), 6.76 (s, 0.4H), 6.66 (d, J = 7.6 Hz, 0.6H), 3.20 (d, J = 3.6 Hz, 3H), 2.93-2.86 (m, 4H), 2.17-2.03 (m, 6H), 1.42 (s, 1.8H), 1.38 (s, 1.2H); 13C NMR (100 MHz, CDCl3) δ: 179.1, 178.7, 144.7, 141.6, 140.0, 139.8, 129.7, 126.2, 123.8, 118.8, 118.7, 106.4, 105.1, 47.8, 47.3, 33.6, 33.2, 32.4, 31.6, 31.5, 30.1, 26.5, 26.3, 25.8, 25.5, 23.7, 21.7, 13.0, 12.8; LRMS (EI 70 ev) m/z (%): 255 (M+1, 9), 254 (M+, 53), 200 (100); LRMS (EI 70 ev) m/z (%): 254 (M+, 35), 200 (100).

3-(1-Methyl-2-oxo-3-phenylindolin-3-yl)propanenitrile (3p):[S1]

Yellow soild; mp 149.5-150.7 °C (uncorrected); 1H NMR (400 MHz, CDCl3) δ: 7.33-7.28 (m, 2H), 7.24-7.18 (m, 5H), 7.09 (t, J = 7.6 Hz, 1H), 6.86 (d, J = 8.0 Hz, 1H), 3.16 (s, 3H), 2.79-2.71 (m, 1H), 2.45-2.37 (m, 1H), 2.10-2.04 (m, 2H); 13C NMR
3-(3-(Hydroxymethyl)-1-methyl-2-oxoindolin-3-yl)propanenitrile (3q):

Brown oil; \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 7.38-7.34 (m, 1H), 7.25 (t, \(J = 4.0\) Hz, 1H), 7.15-7.12 (m, 1H), 6.91 (d, \(J = 8.0\) Hz, 1H), 3.82-3.74 (m, 2H), 3.23 (s, 3H), 2.57 (brs, 1H), 2.52-2.45 (m, 1H), 2.20-2.00 (m, 3H); \(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\): 177.5, 144.0, 129.2, 127.8, 123.3, 123.2, 118.6, 108.8, 66.6, 53.4, 28.4, 26.3, 12.6; LRMS (EI 70 ev) \(m/z\) (%): 200 (M\(^+\), 71), 160 (100), 146 (77); HRMS (ESI) for C\(_{13}\)H\(_{15}\)N\(_2\)O\(_2\) [M+H\(^+\)]: calcd. 231.1136, found. 231.1145.

(3-(2-Cyanoethyl)-1-methyl-2-oxoindolin-3-yl)methyl acetate (3r):\(^{[8]}\)

Brown oil; \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 7.37-7.33 (m, 1H), 7.25-7.23 (m, 1H), 7.13-7.11 (m, 1H), 6.89 (d, \(J = 8.0\) Hz, 1H), 4.45 (d, \(J = 10.8\) Hz, 1H), 4.12 (d, \(J = 10.8\) Hz, 1H), 3.23 (s, 3H), 2.42-2.35 (m, 1H), 2.20-2.01 (m, 3H), 1.93 (s, 3H); \(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\): 175.6, 170.1, 143.8, 129.4, 127.2, 123.6, 123.1, 118.3, 108.5, 66.5, 51.2, 28.9, 26.4, 20.5, 12.3; LRMS (EI 70 ev) \(m/z\) (%): 272 (M\(^+\), 26), 200 (52), 160 (100).
1,3-Dimethyl-3-(3-oxobutyl)indolin-2-one (4a):[S2]

Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) $\delta$: 7.31-7.27 (m, 1H), 7.17 (d, $J$ = 8.0 Hz, 1H), 7.08 (t, $J$ = 8.0 Hz, 1H), 6.87 (d, $J$ = 8.0 Hz, 1H), 3.24 (s, 3H), 2.21-2.08 (m, 3H), 2.07 (s, 3H), 1.98-1.95 (m, 1H), 1.38 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$: 207.7, 180.0, 143.1, 133.2, 128.0, 122.7, 122.6, 108.0, 47.3, 38.5, 31.6, 29.8, 26.1, 23.6; LRMS (EI 70 ev) $m/z$ (%): 231 (M$^+$, 9), 230 (56), 160 (100).

5-Chloro-1,3-dimethyl-3-(3-oxobutyl)indolin-2-one (4j):[S2]

Brown oil; 1H NMR (400 MHz, CDCl$_3$) $\delta$: 7.27 (d, $J$ = 8.0 Hz, 1H), 7.14 (s, 1H), 6.79 (d, $J$ = 8.0 Hz, 1H), 3.22 (s, 3H), 2.24-2.11 (m, 2H), 2.02 (s, 5H), 1.38 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$: 207.3, 179.5, 141.6, 135.0, 128.1, 127.9, 123.2, 109.0, 47.5, 38.4, 31.5, 29.8, 26.5, 23.5; LRMS (EI 70 ev) $m/z$ (%): 267 (M$^+$+2, 18), 265 (M$^+$, 56), 195 (100).

1,3-Dimethyl-3-(3-oxobutyl)-5-(trifluoromethyl)indolin-2-one (4l):
Brown oil; $^1$H NMR (400 MHz, CDCl$_3$) $\delta$: 7.58 (d, $J = 8.0$ Hz, 1H), 7.40 (s, 1H), 6.95 (d, $J = 8.0$ Hz, 1H), 3.27 (s, 3H), 2.23-2.16 (m, 2H), 2.12-2.02 (m, 5H), 1.41 (s, 3H); $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$: 207.2, 180.0, 146.0, 133.9, 125.9 (d, $J = 4.0$ Hz, 1C), 124.9 (d, $J = 32.5$ Hz, 1C), 119.6 (d, $J = 3.7$ Hz, 1C), 107.8, 47.3, 38.3, 31.5, 29.8, 26.4, 23.4; LRMS (EI 70 ev) $m/z$ (%): 299 (M$^+$, 10), 230 (13), 229 (100); HRMS (ESI) for C$_{15}$H$_{17}$F$_3$NO$_2$ [M+H]$^+$: calcd. 300.1211, found. 300.1223.

(C) References


(D) Spectra

3-(1,3-Dimethyl-2-oxoindolin-3-yl)propanenitrile (3a)
3-(1-Benzyl-3-methyl-2-oxoindolin-3-yl)propanenitrile (3b)
3-(1-Acetyl-3-methyl-2-oxoindolin-3-yl)propanenitrile (3c)
3-(1,3,5-Trimethyl-2-oxoindolin-3-yl)propanenitrile (3e)
3-(1,3,6-Trimethyl-2-oxoindolin-3-yl)propanenitrile and

3-(1,3,7-Trimethyl-2-oxoindolin-3-yl)propanenitrile (3f)
3-(1,3,7-Trimethyl-2-oxoindolin-3-yl)propanenitrile (3g)
3-(5-Butyl-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3h)
3-(5-Methoxy-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3i)
3-(5-Chloro-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3j)
3-(5-Fluoro-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3k)
3-(1,3-Dimethyl-2-oxo-5-(trifluoromethyl)indolin-3-yl)propanenitrile (3l)
3-(1,3-Dimethyl-7-(methylthio)-2-oxoindolin-3-yl)propanenitrile (3m)
3-(7-Chloro-1,3-dimethyl-2-oxoindolin-3-yl)propanenitrile (3n)
3-(1,3-Dimethyl-2-oxo-1,2,3,5,6,7-hexahydrocyclopenta[f]indol-3-yl)propanenitrile and 3-(1,3-Dimethyl-2-oxo-1,2,3,6,7,8-hexahydrocyclopenta[g]indol-3-yl)propanenitrile (3o)
3-(1-Methyl-2-oxo-3-phenylindolin-3-yl)propanenitrile (3p)
3-(3-(Hydroxymethyl)-1-methyl-2-oxoindolin-3-yl)propanenitrile (3q)
(3-(2-Cyanoethyl)-1-methyl-2-oxoindolin-3-yl)methyl acetate (3r)
1,3-Dimethyl-3-(3-oxobutyl)indolin-2-one (4a)
5-Chloro-1,3-dimethyl-3-(3-oxobutyl)indolin-2-one (4j)
1,3-Dimethyl-3-(3-oxobutyl)-5-(trifluoromethyl)indolin-2-one (4l)