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Supporting Information for

## Cyclopropanations of N-vinylimides via redox-neutral

## photocatalysed radical addition/anionic cyclosation process

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## **Table of Contents**

1. General Information	S3
1.1 Solvents, Reagents, and Starting Materials	S3
1.2 Instruments	S3
1.3 Picture of a Typical Reaction Setup	S4
2. Preparation of Substrates 1, 4, 6, 8, 10, and 12	S4
2.1 Know compounds reported in our previous work.	S4
2.2 General Procedure for the Synthesis of <i>N</i> -Vinylimides 1, 4, 6, and 8j	S5
2.3 Procedure for the Synthesis of <i>N</i> -Vinylimide <b>8a</b>	S11
2.4 General Procedure for the Synthesis of <i>N</i> -Vinylimides <b>8b</b> and <b>12</b>	S11
2.5 General Procedure for the Synthesis of $\alpha$ -Imidoacrylates <b>8k</b> and <b>8l</b>	S12
2.6 Procedure for the Synthesis of Substrates <b>10a-10c</b>	S13
2.7 Procedure for the Synthesis of Substrates 10d and 10e	S14
2.8 Procedure for the Synthesis of <b>10f</b>	S15
3. General Procedure of Photoredox-Catalysed Cyclopropanation Reactions	S16
4. Procedure of Deprotection of the Boc Group	S26
5. Procedure of the Deprotection of Phthaloyl Group	S26
6. References	S27
7. NMR Spectra of New Compounds	S29

## **1. General Information**

#### 1.1 Solvents, Reagents, and Starting Materials

All reactions were performed under nitrogen atmosphere. DMF and CH<sub>2</sub>Cl<sub>2</sub> were dried from CaH. The dehydrated solvents DMSO, DMA and acetonitrile were purchased from Energy Chemical Chemicals. Photoredox catalysts, alkyl silicates were reported in our previous works.<sup>1</sup> *N*-vinylimides were prepared according to literature procedures.<sup>2,3</sup> All other chemicals were purchased from local vendors, and used as supplied unless otherwise stated.

#### **1.2 Instruments**

NMR spectra were recorded on a Bruker Avance 500 spectrometer (500 MHz). Chemical shifts were reported in ppm downfield from tetramethylsilane, and calibrated using residue undeuterated solvent (CHCl<sub>3</sub> at 7.26 ppm <sup>1</sup>H NMR, 77.0 ppm <sup>13</sup>C NMR). Spectra were reported as follows: chemical shift ( $\delta$  ppm), multiplicity (s = singlet, d = doublet, t = triplet, q =quartet, m = multiplet), coupling constants (Hz) and integration. High resolution mass spectra (HRMS) were recorded on an ESI-Q-TOF spectrometer Agilent 6210 ESI/TOF. TLC analyses were performed on precoated GF<sub>254</sub> silica gel plates and were visualized under UV254 nm light or by I<sub>2</sub> staining. Column chromatography was carried out using 300-400 mesh silica gel and eluted with petroleum/ethyl acetate unless otherwise noted.

## **1.3 Picture of a Typical Reaction Setup**



- 2. Preparation of Substrates 1, 4, 6, 8, 10, and 12
- 2.1 Know compounds reported in our previous work.<sup>2</sup>



#### 2.2 General Procedure for the Synthesis of N-Vinylimides 1, 4, 6, and 8j



(a) A mixture of acetophenone (2.4 g, 20 mmol), NH<sub>2</sub>OH·HCl (2.08 g, 30 mmol), and NaOAc (4.1 g, 50 mmol) in EtOH (10 mL) and H<sub>2</sub>O (30 mL) or MeOH (30 ml) was placed into a 100 mL round-bottomed flask equipped with a condenser. Then the flask was heated to 95 °C and the reaction was monitored by TLC. Add water after cooling down to room temperature, then the mixture was extracted with ethyl acetate twice. The organic layer was collected, dried over MgSO<sub>4</sub> and vacuo to afford the ketoxime which was used without further purification for the next step.

(b) To an oven-dried 50 mL two-neck round-bottom flask assembled with condenser was added the above ketoxime (1.35 g, 10 mmol). Anhydrous toluene (0.5 M) was added followed by acetic anhydride (3.06 g, 30 mmol), acetic acid (1.8 g, 30 mmol) and iron powder (1.12 g, 20 mmol). The reaction flask was put into a 70 °C preheated oil bath and allowed to stir under nitrogen atomsphere. After the reaction completed and cooled to room temperature, ethyl acetate was added and the mixture was filtered through a short pad of celite. The solution thus was evaporated to get the crude enamide, which was directly purified by column chromatography.

(c) The enamide (805 mg, 5 mmol) and DMAP (61 mg, 0.5 mmol) were dissolved in  $CH_3CN$  (10 mL) in a dry two-necked round-bottom flask under nitrogen. Then  $Boc_2O$  (1.63 g, 7.5 mmol) was added in dropwise at room temperature. The completion of the reaction was confirmed by checking TLC and the reaction was quenched by adding water (10 mL). The organic layer was extracted with ethyl acetate through stages of extraction with water. The combined organic layer was concentrated under reduced pressure and the crude product was purified by column chromatography over silica gel to give the pure product **1a**.



*tert*-Butyl acetyl(1-phenylvinyl)carbamate (1a). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.41-7.27 (m, 5H), 5.77 (s, 1H), 5.18 (s, 1H), 2.58 (s, 3H), 1.30 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.8, 152.4, 143.7, 136.8, 128.5, 128.4, 125.2, 113.8, 83.2, 27.6, 26.1. This compound has been reported in the published literature.<sup>3a</sup>



*tert*-Butyl acetyl(1-(p-tolyl)vinyl)carbamate (4a). Flash column chromatography to afford product as a white solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.29 – 7.17 (m, 2H), 7.12 (d, *J* = 7.8 Hz, 2H), 5.72 (s, 1H), 5.12 (s, 1H), 2.56 (s, 3H), 2.34 (s, 3H), 1.31 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.7, 152.4, 143.7, 138.3, 133.9, 129.1, 125.1, 112.9, 83.1, 27.7, 26.1, 21.2. This compound has been reported in the published literature.<sup>3b</sup>



*tert*-Butyl acetyl(1-(m-tolyl)vinyl)carbamate (4b). Flash column chromatography to afford product as a white solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.24-7.18 (m, 1H), 7.18-7.13 (m, 2H), 7.13-7.07 (m, 1H), 5.75 (s, 1H), 5.16 (s, 1H), 2.57 (s, 3H), 2.33 (s, 3H), 1.30 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 172.7, 152.4, 143.8, 138.0, 136.7, 129.2, 128.3, 125.8, 122.4, 113.5, 83.1, 27.6, 26.1, 21.4. This compound has been reported in the published literature.<sup>3b</sup>



*tert*-Butyl acetyl(1-(o-tolyl)vinyl)carbamate (4c). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.23-7.11 (m, 4H), 5.41 (s, 1H), 5.39 (s, 1H), 2.54 (s, 3H), 2.44 (s, 3H), 1.26 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.9, 152.7, 141.6, 137.2, 135.7, 130.9, 127.9, 127.8, 125.7, 117.5, 83.3, 27.6, 26.3, 20.8. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>16</sub>H<sub>22</sub>NO<sub>3</sub>: 276.1600, found 276.1597.



*tert*-Butyl acetyl(1-(3,4-dimethoxyphenyl)vinyl)carbamate (4d). Flash column chromatography to afford product as a pale yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  6.93-6.75 (m, 3H), 5.65 (s, 1H), 5.08 (s, 1H), 3.86 (s, 3H), 3.85 (s, 3H), 2.54 (s, 3H), 1.31 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.6, 152.4, 149.4, 148.8, 143.4, 129.7, 117.9, 112.2, 110.8, 108.5, 83.0, 55.8, 27.6, 26.0. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>24</sub>NO<sub>5</sub>: 322.1654, found 322.1655.



*tert*-Butyl acetyl(1-(benzo[d][1,3]dioxol-5-yl)vinyl)carbamate (4e). Flash column chromatography to afford product as a pale yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  6.94-6.68 (m, 3H), 5.96 (s, 2H), 5.63 (d, J = 0.8 Hz, 1H), 5.08 (d, J = 0.8 Hz, 1H), 2.55 (s, 3H), 1.35 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.7, 152.4, 147.9, 147.8, 143.3, 131.2, 119.2, 112.6, 108.1, 105.8, 101.2, 83.2, 27.7, 26.1. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>16</sub>H<sub>20</sub>NO<sub>5</sub>: 306.1341, found 306.1340.



*tert*-Butyl acetyl(1-(4-chlorophenyl)vinyl)carbamate (4f). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.32-7.27 (m, 4H), 5.75 (d, *J* = 1.1 Hz, 1H), 5.20 (d, *J* = 1.0 Hz, 1H), 2.58 (s, 3H), 1.32 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.7, 152.2, 142.7, 135.4, 134.2, 128.7, 126.5, 114.4, 83.4, 27.7, 26.2. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>15</sub>H<sub>19</sub>ClNO<sub>3</sub>: 296.1053, found 296.1052.



*tert*-Butyl acetyl(1-(4-bromophenyl)vinyl)carbamate (4g). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.47-7.43 (m, 2H), 7.24-7.20 (m, 2H), 5.76 (d, J = 1.2 Hz, 1H), 5.20 (d, J = 1.1 Hz, 1H), 2.58 (s, 3H), 1.32 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.7, 152.2, 142.8, 135.9, 131.7, 126.8, 122.4, 114.5, 83.5, 27.7, 26.2. This compound has been reported in the published literature.<sup>3b</sup>



*tert*-Butyl acetyl(1-(4-(trifluoromethyl)phenyl)vinyl)carbamate (4h). Flash column chromatography to afford product as a white solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.66-7.53 (m, 2H), 7.51-7.39 (m, 2H), 5.86 (d, J = 0.9 Hz, 1H), 5.30 (d, J = 1.0 Hz, 1H), 2.60 (s, 3H), 1.33 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.7, 152.1, 142.6, 140.4, 130.3(q, J = 32.6 Hz), 125.5(4) (q, J = 3.8 Hz), 125.4(8), 124.0 (q, J = 272.5 Hz), 116.0, 83.6, 27.6, 26.1. <sup>19</sup>F NMR (471 MHz, CDCl<sub>3</sub>)  $\delta$  -62.7. This compound has been reported in the published literature.<sup>3b</sup>



**Methyl 4-(1-(N-(tert-butoxycarbonyl)acetamido)vinyl)benzoate** (4i). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  8.00 (d, J = 8.5 Hz, 2H), 7.41 (d, J = 8.5 Hz, 2H), 5.88 (s, 1H), 5.29 (s, 1H), 3.91 (s, 3H), 2.60 (s, 3H), 1.29 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.7, 166.6, 152.1, 142.8, 141.2, 129.9, 129.8, 125.1, 115.9, 83.5, 52.1, 27.6, 26.2. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>22</sub>NO<sub>5</sub>: 320.1498, found 320.1500.



MeO<sub>2</sub>S

*tert*-Butyl acetyl(1-(4-(methylsulfonyl)phenyl)vinyl)carbamate (4j). Flash column chromatography to afford product as a yellow oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.86 (d, J = 8.5 Hz, 2H), 7.50 (d, J = 8.5 Hz, 2H), 5.88 (d, J = 1.3 Hz, 1H), 5.32 (d, J = 1.3 Hz, 1H), 2.99 (s, 3H), 2.55 (s, 3H), 1.28 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.5, 151.8, 142.1, 142.0, 139.9, 127.6, 125.8, 117.1, 83.7, 44.3, 27.5, 26.0. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>16</sub>H<sub>21</sub>NO<sub>5</sub>NaS: 362.1038, found 362.1044.



*tert*-Butyl (1-([1,1'-biphenyl]-4-yl)vinyl)(acetyl)carbamate(4k). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.66-7.52 (m, 4H), 7.49-7.40 (m, 4H), 7.39-7.33 (m, 1H), 5.84 (s, 1H), 5.22 (s, 1H), 2.61 (s, 3H), 1.34 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.8, 152.4, 143.4, 141.2, 140.4, 135.7, 128.8, 127.5, 127.2, 127.0, 125.6, 113.8, 83.3, 27.7, 26.3. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>21</sub>H<sub>23</sub>NO<sub>3</sub>Na: 360.1576, found 360.1574.



*tert*-Butyl acetyl(1-(naphthalen-2-yl)cyclopropyl)carbamate (4l). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.85-7.78 (m, 3H), 7.73 (s, 1H), 7.60-7.52 (m, 1H), 7.51-7.43 (m, 2H), 5.92 (s, 1H), 5.29 (s, 1H), 2.64 (s, 3H), 1.28 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.8, 152.4, 143.7, 134.1, 133.2, 133.1, 128.3(1), 128.2(9), 127.6, 126.3(5), 126.3(1), 124.1, 123.3, 114.4, 83.3, 27.7, 26.2. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>19</sub>H<sub>21</sub>NO<sub>3</sub>Na: 334.1419, found 334.1423.



*tert*-Butyl acetyl(1-(6-methoxynaphthalen-2-yl)vinyl)carbamate (4m). Flash column chromatography to afford product as a pale white solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.73-7.63 (m, 3H), 7.51 (dd, J = 8.6, 1.9 Hz, 1H), 7.17-7.09 (m, 2H), 5.89-5.84 (m, 1H), 5.27-5.21 (m, 1H), 3.92 (s, 3H), 2.62 (s, 3H), 1.28 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.8, 158.1, 152.4, 143.8, 134.5, 132.0, 129.8, 128.6, 127.1, 123.9, 123.8, 119.2, 113.4, 105.6, 83.2, 55.3, 27.6, 26.2. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>20</sub>H<sub>24</sub>NO<sub>4</sub>: 342.1705, found 342.1702.



*tert*-Butyl acetyl(1-(naphthalen-1-yl)vinyl)carbamate (4n). Flash column chromatography to afford product as a pale yellow oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  8.48 (d, *J* = 8.4 Hz, 1H), 7.87-7.73 (m, 2H), 7.55-7.37 (m, 4H), 5.68 (s, 1H), 5.63 (s, 1H), 2.61 (s, 3H), 1.13 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.3, 152.6, 140.0, 135.6, 133.8, 131.0, 128.5, 128.3, 126.1, 125.7, 125.6, 124.8, 124.7, 118.7, 83.4, 27.5, 26.3. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>19</sub>H<sub>22</sub>NO<sub>3</sub>: 312.1600, found 312.1603.



*tert*-Butyl acetyl(1-(thiophen-2-yl)vinyl)carbamate (40). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.22-7.16 (m, 1H), 6.98-6.88 (m, 2H), 5.70 (d, J = 1.3 Hz, 1H), 5.09 (d, J = 1.1 Hz, 1H), 2.56 (s, 3H), 1.37 (s, 9H).<sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.2, 152.0, 141.2, 138.2, 127.3, 125.3, 124.4, 113.1, 83.4, 27.7, 26.0. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>13</sub>H<sub>17</sub>NO<sub>3</sub>NaS: 290.0827, found 290.0829.



*tert*-Butyl acetyl(1-(pyridin-2-yl)vinyl)carbamate (4p). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  8.62-8.50 (m, 1H), 7.71-7.59 (m, 1H), 7.45-7.33 (m, 1H), 7.22-7.11 (m, 1H), 6.28 (d, J = 0.7 Hz, 1H), 5.40 (d, J = 0.7 Hz, 1H), 2.61 (s, 3H), 1.30 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.9, 153.7, 152.3, 149.2, 143.0, 136.4, 122.7, 119.1, 116.6, 83.0, 27.7, 26.2. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>14</sub>H<sub>18</sub>N<sub>2</sub>O<sub>3</sub>Na: 285.1215, found 285.1210.



*tert*-Butyl acetyl(3,3-dimethylbut-1-en-2-yl)carbamate (4q). Flash column chromatography to afford product as a pale yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  5.28 (s, 1H), 4.92 (s, 1H), 2.48 (s, 3H), 1.47 (s, 9H), 1.10 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.0, 153.8, 153.0, 114.0, 82.9, 36.8, 30.3, 27.9, 26.3. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>13</sub>H<sub>23</sub>NO<sub>3</sub>Na: 264.1576, found 264.1566.



*tert*-Butyl acetyl(3,4-dihydronaphthalen-1-yl)carbamate (6a). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.21-7.07 (m, 3H), 7.01-6.83 (m, 1H), 5.88 (t, *J* = 4.7 Hz, 1H), 2.92-2.76 (m, 2H), 2.54 (s, 3H), 2.53-2.44 (m, 1H), 2.44-2.34 (m, 1H), 1.33 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.5, 152.5, 136.1, 135.6, 132.2, 127.4(8), 127.4(5), 126.9, 126.4, 120.9, 82.7, 27.6, 27.3, 26.0, 22.8. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>21</sub>NO<sub>3</sub>Na: 310.1419, found 310.1418.



*tert*-Butyl acetyl(2H-chromen-4-yl)carbamate (6b). Flash column chromatography to afford product as a yellow oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.18-7.05 (m, 1H), 6.92-6.75 (m, 3H), 5.63 (t, J = 3.7 Hz, 1H), 5.01-4.80 (m, 2H), 2.57 (s, 3H), 1.33 (s, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.2, 154.7, 151.8, 133.0, 129.6, 121.6, 121.2, 120.9, 120.0, 115.9, 83.3, 65.2, 27.6, 26.0. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>16</sub>H<sub>19</sub>NO<sub>4</sub>Na: 312.1212, found 312.1207.

**Methyl 2-(N-(tert-butoxycarbonyl)acetamido)acrylate (8j).** Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  6.48-6.42 (m, 1H), 5.67-5.62 (m, 1H), 3.80-3.74 (m, 3H), 2.58-2.52 (m, 3H), 1.48-1.42 (m, 9H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.5, 163.6, 151.5, 135.3, 126.0, 83.7, 52.4, 27.8, 26.0. This compound has been reported in the published literature. <sup>3c</sup>

#### 2.3 Procedure for the Synthesis of N-Vinylimide 8a



A mixture of ketoxime (1.35 g, 10 mmol), propionic anhydride (2.6 g, 20 mmol), NaHSO<sub>3</sub> (3.12 g, 30 mmol) and CuI (0.19 g, 1 mmol) was stirred in 1,2-dichloroethane (0.1 M) at 120 °C under argon atmosphere. After completion of the reaction (detected by TLC), the reaction mixture was cooled to room temperature, diluted with EtOAc, and washed with Na<sub>2</sub>CO<sub>3</sub> and brine. The organic layers were dried over anhydrous Na<sub>2</sub>SO<sub>4</sub> and evaporated in vacuo. The desired product was obtained after purification by flash chromatography on silica gel with hexane/ethyl acetate as the eluent. The procedure of *N*-Boc protection reaction for the preparation of **1a** was employed for the synthesis of **8a**.



*tert*-Butyl (1-phenylvinyl)(propionyl)carbamate (8a). Flash column chromatography to afford product as a white solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.41-7.23 (m, 5H), 5.76 (s, 1H), 5.16 (s, 1H), 2.96 (q, *J* = 7.3 Hz, 2H), 1.30 (s, 9H), 1.19 (t, *J* = 7.3 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  176.5, 152.3, 143.9, 137.0, 128.4, 128.3, 125.2, 113.6, 83.0, 31.2, 27.6, 9.3. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>16</sub>H<sub>21</sub>NO<sub>3</sub>Na: 298.1419, found 298.1413.

#### 2.4 General Procedure for the Synthesis of N-Vinylimides 8b and 12



The enamide (805 mg, 5 mmol) was dissolved in dry DMF (15 mL) in a dry round-bottom flask under nitrogen. The solution was cooled to 0 °C and sodium hydride (60% dispersion in mineral oil) (300 mg, 7.5 mmol) was added in portions. The resulting suspension was stirred at the same temperature for 10 min. Then AcCl (790 mg, 10 mmol) was added dropwise and the final solution was continued to stir for overnight at room temperature. The completion of the reaction was confirmed by checking TLC and the excess of sodium hydride was quenched by adding 10 mL water at 0 °C. The organic layer was extracted with ethyl acetate through stages of extraction with water. The combined organic layer was concentrated under reduced pressure and the crude product was purified by column chromatography over silica gel to give the pure product **8b**.



*N*-acetyl-*N*-(1-phenylvinyl)acetamide (8b). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.50-7.24 (m, 5H), 6.01 (s, 1H), 5.30 (s, 1H), 2.39 (s, 6H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.6, 144.4, 135.1, 129.0, 128.9, 124.8, 115.6, 26.1. This compound has been reported in the published literature. <sup>3a</sup>



(*E*)-*N*-acetyl-*N*-(1-phenylprop-1-en-1-yl)acetamide (12a). Flash column chromatography to afford product as a yellow oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.38-7.24 (m, 5H), 6.44 (q, *J* = 7.0 Hz, 1H), 2.37 (s, 6H), 1.75 (d, *J* = 7.0 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.6, 137.8, 136.2, 128.9, 128.2, 125.1, 124.5, 26.0, 13.4. This compound has been reported in the published literature. <sup>3d</sup>



(Z)-N-acetyl-N-(1-phenylprop-1-en-1-yl)acetamide (12b). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.43-7.28 (m, 5H), 5.82 (q, J = 7.3 Hz, 1H), 2.38 (s, 6H), 1.98 (d, J = 7.4 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.4, 137.2, 135.4, 128.9, 128.4, 128.3, 128.2, 26.5, 14.8. This compound has been reported in the published literature. <sup>3d</sup>

#### 2.5 General Procedure for the Synthesis of a-Imidoacrylates 8k and 8l



To a solution of carbazole (836 mg, 5 mmol), triphenylphosphine (131 mg, 0.5 mmol), and sodium acetate (205 mg, 2.5 mmol) in toluene (10 mL) at 105 °C were added sequentially acetic acid (150 mg, 2.5 mmol) and ethyl propiolate (589 mg, 6 mmol). After the reaction was complete as monitored by TLC, the resulting mixture was partitioned between water and ethyl acetate, and the separated aqueous layer extracted with ethyl acetate. The combined organic layers were washed with brine, dried over anhydrous Na<sub>2</sub>SO<sub>4</sub>, filtered, and evaporated under reduced pressure. The residue was purified by flash column chromatography on silica gel to yield pureproduct **8**k.



**Ethyl 2-(9H-carbazol-9-yl)acrylate (8k).** Flash column chromatography to afford product as a pale yellow oil. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  8.17-8.03 (m, 2H), 7.51-7.36 (m, 2H), 7.36-7.21 (m, 4H), 6.88 (s, 1H), 6.12 (s, 1H), 4.25 (q, J = 7.1 Hz, 2H), 1.20 (t, J = 7.1 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  163.5, 140.6, 134.9, 126.2, 125.9, 123.5, 120.3, 120.1, 109.9, 61.8, 14.0. This compound has been reported in the published literature. <sup>3</sup>e



**Ethyl 2-(2,4-dichloro-7H-pyrrolo[2,3-d]pyrimidin-7-yl)acrylate (8l).** Flash column chromatography to afford product as a pale white solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.36 (d, J = 3.7 Hz, 1H), 6.75-6.63 (m, 2H), 6.21 (s, 1H), 4.32 (q, J = 7.1 Hz, 2H), 1.32 (t, J = 7.1 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  162.2, 153.0, 152.5(1), 152.4(9), 132.9, 130.6, 124.9, 116.8, 100.7, 62.4, 14.0. This compound has been reported in the published literature. <sup>3e</sup>

#### 2.6 Procedure for the Synthesis of Substrates 10a-10c



A mixture of iodobenzene (816 mg, 4.00 mmol), *N*-vinylphthalimide (693 mg, 4.00 mmol),  $Cy_2NMe$  (1.17 g, 6.00 mmol), TBAB (1.29 g, 4.00 mmol) and palladium acetate (1.00 mg, 4.00 µmol) in DMF (8 mL) was heated at 120 °C in a round-bottom flask under nitrogen. When full conversion was observed by TLC, the organic layer was extracted with ethyl acetate through stages of extraction with water. The combined organic layer was concentrated under reduced pressure and the crude product was purified by column chromatography over silica gel to give the pure product **10a**.



(*E*)-2-styrylisoindoline-1,3-dione (10a). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.91-7.85 (m, 2H), 7.77-7.71 (m, 2H), 7.64 (d, *J* = 15.2 Hz, 1H), 7.46 (d, *J* = 7.6 Hz, 2H), 7.37-7.31 (m, 3H), 7.28-7.22 (m, 1H). <sup>13</sup>C NMR (126 MHz,

CDCl<sub>3</sub>) & 166.3, 135.9, 134.4, 131.6, 128.7, 127.6, 126.1, 123.6, 120.1, 117.5. This compound has been reported in the published literature. <sup>3f</sup>



(*E*)-2-(4-methoxystyryl)isoindoline-1,3-dione (10b) . Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.92-7.87 (m, 2H), 7.78-7.72 (m, 2H), 7.60 (d, *J* = 15.1 Hz, 1H), 7.41 (d, *J* = 8.7 Hz, 2H), 7.24 (d, *J* = 15.1 Hz, 1H), 6.90 (d, *J* = 8.7 Hz, 2H), 3.83 (s, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  166.5, 159.3, 134.4, 131.7, 128.5, 127.4, 123.6, 120.1, 115.9, 114.2, 55.3. This compound has been reported in the published literature. <sup>3f</sup>



(*E*)-2-(4-fluorostyryl)isoindoline-1,3-dione (10c). Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.93-7.88 (m, 2H), 7.79-7.74 (m, 2H), 7.62 (d, *J* = 15.1 Hz, 1H), 7.44 (m, 2H), 7.28 (d, *J* = 14.8 Hz, 1H), 7.05 (t, *J* = 8.7 Hz, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  166.3, 162.3 (d, *J* = 247.8 Hz), 134.5, 132.0 (d, *J* = 3.3 Hz), 131.6, 127.7 (d, *J* = 8.2 Hz), 123.6, 119.1, 117.3 (d, *J* = 2.3 Hz), 115.7 (d, *J* = 21.8 Hz). <sup>19</sup>F NMR (471 MHz, CDCl<sub>3</sub>)  $\delta$  -114.2. This compound has been reported in the published literature.<sup>3g</sup>

#### 2.7 Procedure for the Synthesis of Substrates 10d and 10e



The phthalimide (735.6 mg, 5 mmol) and DMAP (305 mg, 2.5 mmol) were dissolved in  $CH_3CN$  (0.1 M) under N<sub>2</sub> atmosphere. The methyl propiolate (504 mg, 6 mmol) was then slowly added via syringe, and the reaction was stirred until TLC analysis indicated complete consumption of the substrate. The solvent was removed under reduced pressure, and the residue was purified by flash column chromatography on silica gel to give the pure product **10d**.



**Methyl (E)-3-(1,3-dioxoisoindolin-2-yl)acrylate (10d)**. Flash column chromatography to afford product as a yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  8.01-7.90 (m, 3H), 7.88-7.75 (m, 2H), 6.98 (d, *J* = 14.8 Hz, 1H), 3.80 (s, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  167.5, 165.5, 135.2, 131.4, 131.2, 124.3, 108.3, 51.8. This compound has been reported in the published literature. <sup>3h</sup>



**Ethyl (***E***)-3-(1,3-dioxoisoindolin-2-yl)acrylate (10e).** Flash column chromatography to afford product as a white solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.99-7.88 (m, 3H), 7.81 (m, 2H), 6.96 (d, *J* = 14.6 Hz, 1H), 4.26 (q, *J* = 7.2 Hz, 2H), 1.33 (t, *J* = 7.1 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  167.1, 165.5, 135.1, 131.4, 130.9, 124.2, 108.8, 60.6, 14.3. This compound has been reported in the published literature. <sup>3i</sup>

#### 2.8 Procedure for the Synthesis of 10f



To a solution of phthaloyl dichloride (1.62 g, 8.0 mmol) in  $CH_2Cl_2$  (16 mL) was added  $Et_3N$  (2.2 ml, 1.62 g, 16.0 mmol) at 0 °C under argon atmosphere. Then a solution of diethyl 2-(aminomethylene)malonate (748 mg, 4.0 mmol) in  $CH_2Cl_2$  (8 mL) was added dropwise at 0 °C and stirred for 1 h at the same temperature. The resulting mixture was stirred at room temperature for 24 h, diluted with  $CH_2Cl_2$ , washed with saturated  $Na_2CO_3$  (aq) and water. The organic phase was dried over  $Na_2SO_4$ . After evaporation, the crude product was purified by column chromatography to afford a product **10f**.



**Diethyl 2-((1,3-dioxoisoindolin-2-yl)methylene)malonate (10f)**. Flash column chromatography to afford product as a pale yellow solid. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.88-7.81 (m, 2H), 7.80-7.73 (m, 2H), 7.71-7.65 (m, 1H), 4.31-4.15 (m, 4H), 1.31-1.20 (m, 6H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  164.3, 164.0, 163.8, 135.2, 131.1, 128.4, 124.2, 117.9, 61.5, 61.2, 13.9, 13.8. This compound has been reported in the published literature.<sup>3j</sup>

## 3. General Procedure of Photoredox-Catalysed

### **Cyclopropanation Reactions**



To an oven dried transparent 10 mL Schlenk tube equipped with stirring bar,  $Ir[dF(CF_3)ppy]_2(dtbbpy)PF_6$  (4.5 mg, 0.004 mmol, 0.02 equiv), potassium [18-crown-6] bis(catecholato)halomethylsilicate (0.4 mmol, 2.0 equiv), the enamide (0.2 mmol, 1.0 equiv) were added. The tube was evacuated and filled with nitrogen for 3 times. The tube was then charged with degassed DMSO (6.0 mL, 0.033 M) via a syringe. The tube was irradiated with a 9 W blue LEDs strip spiraled within a bowel for 24 h (cooling with a fan). After the reaction was complete, the reaction solution was diluted with saturated Na<sub>2</sub>CO<sub>3</sub> aqueous solution (10 mL), and was extracted with EtOAc (5 x 10 mL). The organic layer was washed with brine, dried over MgSO<sub>4</sub>, filtered, and solvent was evaporated to obtain crude product. Flash chromatography over silica gel afforded the product.



*tert*-Butyl acetyl(1-phenylcyclopropyl)carbamate (3a). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.27-7.23 (m, 2H), 7.21-7.13 (m, 3H), 2.45 (s, 3H), 1.49-1.46 (m, 2H), 1.45 (s, 9H), 1.31-1.27 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.1, 153.9, 141.9, 128.0, 126.1, 125.3, 82.9, 39.3, 27.9, 27.0, 20.7. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>16</sub>H<sub>21</sub>NO<sub>3</sub>Na: 298.1419, found 298.1425.



*tert*-Butyl acetyl(1-(p-tolyl)cyclopropyl)carbamate (5a). Flash column chromatography to afford product as a yellow solid (50.3 mg, 87% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.16-7.12 (m, 2H), 7.09-7.04 (m, 2H), 2.43 (s, 3H), 2.30 (s, 3H), 1.47 (s, 9H), 1.45-1.40 (m, 2H), 1.27-1.23 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.2, 154.0, 138.9, 135.8, 128.7, 125.8, 82.9, 39.2, 28.0, 27.1, 20.9, 20.3. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>23</sub>NO<sub>3</sub>Na: 312.1576, found 312.1580.



*tert*-Butyl acetyl(1-(m-tolyl)cyclopropyl)carbamate (5b). Flash column chromatography to afford product as brown oil (43.9 mg, 76% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.17-7.11 (m, 1H), 7.06-6.93 (m, 3H), 2.45 (s, 3H), 2.31 (s, 3H), 1.47 (s, 9H), 1.46-1.44 (m, 2H), 1.30-1.24 (m, 2H).<sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.2, 154.0, 141.8, 137.5, 127.9, 127.0, 126.2, 122.7, 82.9, 39.3, 27.9, 27.1, 21.5, 20.6. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>23</sub>NO<sub>3</sub>Na: 312.1576, found 312.1571.



*tert*-Butyl acetyl(1-(o-tolyl)cyclopropyl)carbamate (5c). Flash column chromatography to afford product as a yellow oil(41.1 mg, 71% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.85-7.81 (m, 1H), 7.17-7.08 (m, 3H), 2.57 (s, 3H), 2.23 (s, 3H), 1.53 (s, 9H), 1.51-1.46 (m, 2H), 1.37-1.33 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.5, 154.4, 139.0, 138.6, 132.8, 130.9, 127.6, 125.1, 83.5, 39.4, 27.9, 27.2, 20.5, 17.4. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>23</sub>NO<sub>3</sub>Na: 312.1576, found 312.1579.



*tert*-Butyl acetyl(1-(3,4-dimethoxyphenyl)cyclopropyl)carbamate (5d). Flash column chromatography to afford product as yellow oil (52.3 mg, 78% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.01-6.98 (m, 1H), 6.96-6.92 (m, 1H), 6.75 (d, J = 8.4 Hz, 1H), 3.85 (s, 3H), 3.83 (s, 3H), 2.40 (s, 3H), 1.50 (s, 9H), 1.41-1.37 (m, 2H), 1.23-1.19 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.3, 154.0, 148.4, 147.8, 134.5, 119.7, 111.0, 110.5, 83.0, 55.8, 55.8, 39.2, 28.0, 27.2, 19.2. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>18</sub>H<sub>25</sub>NO<sub>5</sub>Na: 358.1630, found 358.1630.



*tert*-Butyl acetyl(1-(benzo[d][1,3]dioxol-5-yl)cyclopropyl)carbamate (5e). Flash column chromatography to afford product as a yellow oil (52.9 mg, 83% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  6.93-6.86 (m, 1H), 6.86-6.80 (m, 1H), 6.68 (d, J = 8.1 Hz, 1H), 5.90 (s, 2H), 2.40 (s, 3H), 1.49 (s, 9H), 1.39-1.34 (m, 2H), 1.23-1.16 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.1, 153.9, 147.3, 146.1, 135.8, 120.3, 107.8, 107.6, 100.9, 83.1, 39.3, 28.0, 27.2, 19.6. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>21</sub>NO<sub>5</sub>Na: 342.1317, found 342.1316.



*tert*-Butyl acetyl(1-(4-chlorophenyl)cyclopropyl)carbamate (5f). Flash column chromatography to afford product as a yellow oil (55.6 mg, 90% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.24-7.19 (m, 2H), 7.18-7.13 (m, 2H), 2.44 (s, 3H), 1.46 (s, 9H), 1.44-1.40 (m, 2H), 1.30-1.26 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.2, 153.7, 140.5, 132.0, 128.1, 127.3, 83.2, 39.0, 28.0, 27.1, 20.6. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>16</sub>H<sub>20</sub>NO<sub>3</sub>NaCl: 332.1029, found 332.1030.



*tert*-Butyl acetyl(1-(4-bromophenyl)cyclopropyl)carbamate (5g). Flash column chromatography to afford product as a yellow oil (67.1 mg, 95% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.43-7.31 (m, 2H), 7.14-7.03 (m, 2H), 2.44 (s, 3H), 1.46 (s, 9H), 1.44-1.40 (m, 2H), 1.30-1.26 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.1, 153.7, 141.1, 131.1, 127.6, 120.1, 83.2, 39.0, 28.0, 27.1, 20.7. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>16</sub>H<sub>20</sub>NO<sub>3</sub>NaBr: 376.0524, found 376.0529.



F<sub>2</sub>C

*tert*-Butyl acetyl(1-(4-(trifluoromethyl)phenyl)cyclopropyl)carbamate (5h). Flash column chromatography to afford product as a yellow oil (54.9 mg, 80% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.54-7.48 (m, 2H), 7.26-7.21 (m, 2H), 2.48 (s, 3H), 1.54-1.49 (m, 2H), 1.45 (s, 9H), 1.38-1.34 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.2, 153.7, 146.2, 128.4 (q, *J* = 32.4 Hz), 125.3, 125.1 (q, *J* = 3.8 Hz), 124.2 (q, *J* = 272.3 Hz), 83.4, 39.2, 27.9, 27.0, 21.7. <sup>19</sup>F NMR (471 MHz, CDCl<sub>3</sub>)  $\delta$  -62.4. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>20</sub>NO<sub>3</sub>NaF<sub>3</sub>: 366.1293, found 366.1295.



**Methyl 4-(1-(N-(tert-butoxycarbonyl)acetamido)cyclopropyl)benzoate** (5i). Flash column chromatography to afford product as a yellow oil (53.9 mg, 81% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.92 (d, J = 8.4 Hz, 2H), 7.12 (d, J = 8.5 Hz, 2H), 3.87 (s, 3H), 2.48 (s, 3H), 1.58-1.49 (m, 2H), 1.41 (s, 9H), 1.38-1.33 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.2, 166.8, 153.7, 147.6, 129.5, 127.8, 124.4, 83.2, 51.9, 39.4, 27.9, 27.0, 22.2. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>18</sub>H<sub>23</sub>NO<sub>5</sub>Na: 356.1474, found 356.1471.



*tert*-Butyl acetyl(1-(4-(methylsulfonyl)phenyl)cyclopropyl)carbamate (5j). Flash column chromatography to afford product as a yellow oil (46.6 mg, 66% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.84-7.80 (m, 2H), 7.26-7.23 (m, 2H), 3.00 (s, 3H), 2.49 (s, 3H), 1.59-1.52 (m, 2H), 1.43 (s, 9H), 1.42-1.38 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.2, 153.4, 148.9, 138.0, 127.4, 125.4, 83.7, 44.6, 39.3, 27.9, 27.0, 22.6. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>23</sub>NO<sub>5</sub>NaS: 376.1195, found 376.1193.



*tert*-Butyl (1-([1,1'-biphenyl]-4-yl)cyclopropyl)(acetyl)carbamate (5k). Flash column chromatography to afford product as a yellow oil (62.3 mg, 89% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.57 (d, J = 7.7 Hz, 2H), 7.51 (d, J = 8.3 Hz, 2H), 7.46-7.39 (m, 2H), 7.35-7.30 (m, 1H), 7.28 (d, J = 8.3 Hz, 2H), 2.49 (s, 3H), 1.55-1.51 (m, 2H), 1.49 (s, 9H), 1.42-1.28 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.2, 154.0, 141.1, 140.8, 139.1, 128.7, 127.1, 126.9, 126.8, 125.9, 83.1, 39.2, 28.0, 27.1, 20.8. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>22</sub>H<sub>25</sub>NO<sub>3</sub>Na: 374.1732, found 374.1729.



*tert*-Butyl acetyl(1-(naphthalen-2-yl)cyclopropyl)carbamate (5l). Flash column chromatography to afford product as a yellow oil (57.2 mg, 88% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.81-7.72 (m, 3H), 7.69-7.65 (m, 1H), 7.48-7.38 (m, 2H), 7.38-7.33 (m, 1H), 2.49 (s, 3H), 1.62-1.56 (m, 2H), 1.46 (s, 9H), 1.39-1.35 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.4, 154.0, 139.4, 133.1, 132.1, 127.8, 127.4, 125.9, 125.5, 124.5, 124.1, 83.1, 39.6, 28.0, 27.2, 20.7. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>20</sub>H<sub>23</sub>NO<sub>3</sub>Na: 348.1576, found 348.1577.



*tert*-Butyl acetyl(1-(6-methoxynaphthalen-2-yl)cyclopropyl)carbamate (5m). Flash column chromatography to afford product as a yellow oil (46.2 mg, 65% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.70-7.60 (m, 3H), 7.38 (dd, J = 8.7, 1.9 Hz, 1H), 7.13-7.09 (m, 1H), 7.09-7.07 (m, 1H), 3.90 (s, 3H), 2.47 (s, 3H), 1.59-1.52 (m, 2H), 1.47 (s, 9H), 1.37-1.31 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.4, 157.4, 154.1, 137.0, 133.2, 129.3, 128.6, 126.6, 125.1, 124.8, 118.7, 105.4, 83.0, 55.2, 39.5, 28.0, 27.2, 20.2. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>21</sub>H<sub>25</sub>NO<sub>4</sub>Na: 378.1681, found 378.1682.



*tert*-Butyl acetyl(1-(naphthalen-1-yl)cyclopropyl)carbamate (5n). Flash column chromatography to afford product as a yellow oil (35.1 mg, 54% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  9.09 (d, J = 8.6 Hz, 1H), 8.15-8.09 (m, 1H), 7.83 (d, J = 8.1 Hz, 1H), 7.78 (d, J = 8.2 Hz, 1H), 7.59-7.53 (m, 1H), 7.50-7.45 (m, 1H), 7.45-7.37 (m, 1H), 2.22 (s, 3H), 1.74-1.68 (m, 2H), 1.54 (s, 9H), 1.53-1.49 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.8, 154.4, 137.0, 133.8, 132.4, 131.1, 128.8, 128.3, 126.2, 125.7, 125.3, 124.8, 83.7, 38.7, 27.9, 27.3, 17.4. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>20</sub>H<sub>23</sub>NO<sub>3</sub>Na: 348.1576, found 348.1571.



*tert*-Butyl acetyl(1-(thiophen-2-yl)cyclopropyl)carbamate (50). Flash column chromatography to afford product as a brown oil (49.5 mg, 88% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.14-7.09 (m, 1H), 7.07-7.01 (m, 1H), 6.89-6.82 (m, 1H), 2.40 (s, 3H), 1.55 (s, 9H), 1.53-1.46 (m, 2H), 1.27-1.24 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.9, 153.6, 146.0, 126.2, 125.8, 124.3, 83.3, 35.3, 28.1, 27.0, 20.2. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>14</sub>H<sub>19</sub>NO<sub>3</sub>SNa: 304.0983, found 304.0984.



*tert*-Butyl acetyl(1-(pyridin-2-yl)cyclopropyl)carbamate (5p). Flash column chromatography to afford product as a white solid (41.9 mg, 76% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  8.47-8.40 (m, 1H), 7.59-7.49 (m, 1H), 7.06-6.97 (m, 2H), 2.52 (s, 3H), 1.91-1.81 (m, 2H), 1.39 (s, 9H), 1.31-1.24 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.5, 160.5, 153.9, 148.9, 135.8, 120.4, 118.0, 82.9, 41.2, 27.9, 26.9, 22.5. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>15</sub>H<sub>21</sub>N<sub>2</sub>O<sub>3</sub>: 277.1552, found 277.1548.

*tert*-Butyl acetyl(1-(tert-butyl)cyclopropyl)carbamate (5q). Flash column chromatography to afford product as a yellow oil (36.2 mg, 71% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  2.31 (s, 3H), 1.52 (s, 9H), 1.16-1.12 (m, 1H), 1.01-0.96 (m, 1H), 0.89 (s, 9H), 0.85-0.82 (m, 1H), 0.79-0.73 (m, 1H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.7, 155.2, 82.8, 43.8, 35.3, 28.5, 27.9, 27.4, 13.9, 13.7. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>14</sub>H<sub>25</sub>NO<sub>3</sub>Na: 278.1732, found 278.1722.



### $\textit{tert-Butylacetyl} ((1 a S^*, 7 b S^*) - 1, 1 a, 2, 3 - tetrahydro - 7 b H-cyclopropa[a] naphthalen - 7 b - 7 b - 7 b H-cyclopropa[a] naphthalen - 7 b$

yl)carbamate (7a). Flash column chromatography to afford product as a yellow solid (50.6 mg, 84% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.15-6.97 (m, 4H), 2.72-2.61 (m, 1H), 2.56 (s, 2H), 2.52-2.44 (m, 1H), 2.43 (s, 1H), 2.12-1.99 (m, 1H), 1.89-1.79 (m, 1H), 1.75-1.68 (m, 1H), 1.52 (s, 3H), 1.40-.33 (m, 1H), 1.28 (s, 6H), 1.25-1.18 (m, 1H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  174.2, 172.6, 154.4, 153.8, 137.7, 137.5, 133.0, 132.6, 128.7, 128.3, 126.2, 126.0, 125.3(3), 125.3(1), 124.4, 124.2, 82.9, 82.6, 38.4, 38.2, 28.1, 28.0, 27.9, 27.6, 27.2, 26.6, 26.1, 25.9, 19.1, 18.3, 18.2, 18.1(6). HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>18</sub>H<sub>23</sub>NO<sub>3</sub>Na: 324.1576, found 324.1574.



*tert*-Butyl acetyl((1a*S*\*,7b*S*\*)-1a,2-dihydrocyclopropa[c]chromen-7b(1H)-yl)carbamate (7b). Flash column chromatography to afford product as a white solid (48.5 mg, 80% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.11-7.00 (m, 1H), 7.00-6.94 (m, 1H), 6.94-6.86 (m, 1H), 6.86-6.78 (m, 1H), 4.35-4.26 (m, 1H), 4.02 (dd, *J* = 80.0, 10.5 Hz, 1H), 2.52 (d, *J* = 43.0 Hz, 3H), 1.92-1.85 (m, 1H), 1.72-1.66 (m, 1H), 1.57-1.49 (m, 5H), 1.41-1.34 (m, 5H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.9, 172.6, 154.0, 153.4, 151.6, 151.3, 126.9, 126.8(3), 126.8(0), 126.7(7), 124.7, 124.4, 121.8, 121.4, 117.3, 117.2, 83.3, 83.2, 61.7, 61.6, 35.6, 35.4, 30.7, 30.3, 28.0(4), 28.0(1), 27.2, 26.6, 20.4, 19.9. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>21</sub>NO<sub>4</sub>Na: 326.1368, found 326.1363.



*tert*-Butyl (1-phenylcyclopropyl)(propionyl)carbamate (9a). Flash column chromatography to afford product as a black oil (49.7 mg, 86% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.27-7.22 (m, 2H), 7.21-7.13 (m, 3H), 2.81 (q, *J* = 7.3 Hz, 2H), 1.48-1.45 (m, 2H), 1.44 (s, 9H), 1.28-1.23 (m, 2H), 1.13 (t, *J* = 7.3 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  177.1, 154.0, 142.1, 128.0, 126.1, 125.5, 82.8, 39.5, 31.9, 28.0, 20.8, 9.4. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>23</sub>NO<sub>3</sub>Na: 312.1576, found 312.1574.



**N-acetyl-N-(1-phenylcyclopropyl)acetamide** (**9b**). Flash column chromatography to afford product as a yellow oil (34.3 mg, 79% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.33-7.27 (m, 2H), 7.22-

7.17 (m, 1H), 6.95 (d, 2H), 2.39 (s, 6H), 1.67-1.61 (m, 2H), 1.43-1.37 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  174.7, 141.2, 128.7, 126.3, 123.2, 39.6, 26.8, 23.5. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>13</sub>H<sub>15</sub>NO<sub>2</sub>Na: 240.1000, found 240.0996.



**2-(1-Phenylcyclopropyl)isoindoline-1,3-dione** (**9c**). Flash column chromatography to afford product as a pale white solid(34.2 mg, 65% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 7.85-7.79 (m, 2H), 7.72-7.66 (m, 2H), 7.49-7.43 (m, 2H), 7.32-7.26 (m, 2H), 7.24-7.19 (m, 1H), 1.53-1.46 (m, 4H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 168.2, 140.5, 134.0, 131.7, 128.5, 127.5, 127.3, 123.2, 33.9, 14.6. This compound has been reported in the published literature.<sup>4</sup>



**2-(1-(p-tolyl)cyclopropyl)isoindoline-1,3-dione** (**9d**). Flash column chromatography to afford product as a white solid (37.7 mg, 65% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.88-7.75 (m, 2H), 7.75-7.63 (m, 2H), 7.39 (d, 2H), 7.09 (d, 2H), 2.29 (s, 3H), 1.46 (s, 4H).<sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  168.2, 137.6, 137.1, 134.0, 131.7, 129.1, 127.7, 123.2, 33.7, 21.0, 14.2. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>18</sub>H<sub>16</sub>NO<sub>2</sub>: 278.1181, found 278.1180.



**2-(1-(4-Chlorophenyl)cyclopropyl)isoindoline-1,3-dione** (**9e**). Flash column chromatography to afford product as a pale white solid (44 mg, 74% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.89-7.76 (m, 2H), 7.76-7.64 (m, 2H), 7.48-7.37 (m, 2H), 7.30-7.19 (m, 2H), 1.54-1.41 (m, 4H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  168.1, 139.0, 134.1, 133.3, 131.6, 129.3, 128.6, 123.3, 33.3, 14.4. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>17</sub>H<sub>13</sub>NO<sub>2</sub>Cl: 298.0635, found 298.0629.



**Methyl** 1-(1,3-dioxoisoindolin-2-yl)cyclopropane-1-carboxylate (9f). Flash column chromatography to afford product as a pale yellow oil (27.4 mg, 66% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.93-7.83 (m, 2H), 7.78-7.70 (m, 2H), 3.67 (s, 3H), 1.91-1.80 (m, 2H), 1.52-1.43 (m,

2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  171.0, 168.0, 134.3, 131.7, 123.5, 52.9, 31.5, 16.4. This compound has been reported in the published literature.<sup>5</sup>



**Ethyl** 1-(1,3-dioxoisoindolin-2-yl)cyclopropane-1-carboxylate (9g). Flash column chromatography to afford product as a pale yellow oil (37.3 mg, 72% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.95-7.81 (m, 2H), 7.81-7.68 (m, 2H), 4.13 (q, *J* = 7.1 Hz, 2H), 1.88-1.82 (m, 2H), 1.49-1.43 (m, 2H), 1.17 (t, *J* = 7.1 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  170.5, 168.1, 134.2, 131.7, 123.5, 61.8, 31.7, 16.2, 14.1. This compound has been reported in the published literature.<sup>6</sup>



Ethyl 1-(2,5-dioxopyrrolidin-1-yl)cyclopropane-1-carboxylate (9h). Flash column chromatography to afford product as a pale yellow solid (31.2 mg, 74% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  4.12 (q, J = 7.1 Hz, 2H), 2.72 (s, 4H), 1.80-1.72 (m, 2H), 1.32-1.28 (m, 2H), 1.20 (t, J = 7.1 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  176.7, 169.8, 61.8, 32.4, 27.9, 15.7, 14.0. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>10</sub>H<sub>14</sub>NO<sub>4</sub>: 212.0923, found 212.0923.

Boc-N Boc

}\_CO₂Me

1-(*N*, *N*-Di(*tert*-butoxycarbonyl)cyclopropane-1-carboxylic acid methyl ester (9i). Flash column chromatography to afford product as a yellow solid (47.9 mg, 76% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>) δ 3.69 (s, 3H), 1.73-1.67 (m, 2H), 1.48 (s, 18H), 1.25-1.20 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>) δ 172.6, 152.3, 82.6, 52.4, 39.3, 28.0, 20.7. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for  $C_{15}H_{25}NO_6Na$ : 338.1580, found 338.1575.

$$H_3C$$
  $N$   $O$   $CH_3$   $Boc$   $O$   $CH_3$ 

**Methyl 1-(***N***-(***tert***-butoxycarbonyl)acetamido)cyclopropane-1-carboxylate (9j).** Flash column chromatography to afford product as a yellow oil (40.6 mg, 79% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  3.65 (s, 3H), 2.43 (s, 3H), 1.85-1.63 (m, 2H), 1.47 (s, 9H), 1.19-1.08 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  173.2, 172.1, 153.2, 83.1, 52.4, 38.0, 27.9, 26.7, 20.6, 20.5. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>12</sub>H<sub>19</sub>NO<sub>5</sub>Na: 280.1161, found 280.1157.



**Ethyl 1-(9H-carbazol-9-yl)cyclopropane-1-carboxylate (9k).** Flash column chromatography to afford product as a yellow oil (43.5 mg, 78% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  8.13-8.07 (m, 2H), 7.54-7.44 (m, 4H), 7.32-7.26 (m, 2H), 4.09 (q, J = 7.1 Hz, 2H), 2.13-2.08 (m, 2H), 1.71-1.65 (m, 2H), 1.05 (t, J = 7.1 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.1, 141.0, 125.8, 123.3, 120.3, 119.6, 109.7, 61.5, 35.5, 17.9, 14.0. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>18</sub>H<sub>18</sub>NO<sub>2</sub>: 280.1338, found 280.1335.

Ethyl 1-(2,4-dichloro-7H-pyrrolo[2,3-d]pyrimidin-7-yl)cyclopropane-1-carboxylate (91). Flash column chromatography to afford product as a yellow solid (26.9 mg, 45% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.23 (d, J = 3.7 Hz, 1H), 6.60 (d, J = 3.7 Hz, 1H), 4.12 (q, J = 7.1 Hz, 2H), 2.00-1.95 (m, 2H), 1.67-1.62 (m, 2H), 1.14 (t, J = 7.1 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  170.4, 153.6, 152.7, 152.3, 131.2, 116.9, 100.5, 62.1, 38.0, 17.3, 14.0. HRMS (ESI) [M+H]<sup>+</sup>: calculated for C<sub>12</sub>H<sub>12</sub>N<sub>3</sub>O<sub>2</sub>Cl<sub>2</sub>: 300.0307, found 300.0304.



**2-((1S<sup>\*</sup>,2R<sup>\*</sup>)-2-Phenylcyclopropyl)isoindoline-1,3-dione (11a)**. Flash column chromatography to afford product as a yellow solid (17.9 mg, 34% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.85 (m, 2H), 7.72 (m, 2H), 7.33 (m, 4H), 7.26-7.21 (m, 1H), 2.89-2.81 (m, 1H), 2.60-2.49 (m, 1H), 1.71-1.64 (m, 1H), 1.59-1.53 (m, 1H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  168.6, 139.7, 134.1, 131.8, 128.4, 127.1, 126.5, 123.2, 30.2, 23.0, 13.5. This compound has been reported in the published literature.<sup>7</sup>



**2-((1***S***<sup>\*</sup>, 2***R***<sup>\*</sup>)-2-(4-Methoxyphenyl)cyclopropyl)isoindoline-1,3-dione (11b).** Flash column chromatography to afford product as a yellow solid (15.2 mg, 26% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.88 – 7.79 (m, 2H), 7.78-7.66 (m, 2H), 7.28 (d, *J* = 8.7 Hz, 2H), 6.87 (d, *J* = 8.7 Hz, 2H), 3.80 (s, 3H), 2.83-2.67 (m, 1H), 2.56-2.41 (m, 1H), 1.64-1.58 (m, 1H), 1.54-1.46 (m, 1H).<sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  168.6, 158.4, 134.0, 131.8, 131.7, 128.4, 123.2, 113.9, 55.3, 29.8, 22.4, 13.1. This compound has been reported in the published literature.<sup>7</sup>



**2-((1***S***<sup>\*</sup>, 2***R***<sup>\*</sup>)-2-(4-fluorophenyl)cyclopropyl)isoindoline-1,3-dione (11c).** Flash column chromatography to afford product as a yellow solid (11.2 mg, 20% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.91-7.81 (m, 2H), 7.78-7.69 (m, 2H), 7.37-7.29 (m, 2H), 7.01 (t, *J* = 8.7 Hz, 2H), 2.82-2.72

(m, 1H), 2.56-2.44 (m, 1H), 1.70-1.61 (m, 1H), 1.52 (q, J = 7.0 Hz, 1H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  168.6, 161.7 (d, J = 245.1 Hz), 135.3 (d, J = 3.2 Hz), 134.1, 131.8, 128.8 (d, J = 8.1 Hz), 123.2, 115.2 (d, J = 21.3 Hz), 29.9, 22.5, 13.3. <sup>19</sup>F NMR (471 MHz, CDCl<sub>3</sub>)  $\delta$  -116.4. This compound has been reported in the published literature.<sup>7</sup>



**Methyl (15<sup>\*</sup>,25<sup>\*</sup>)-2-(1,3-dioxoisoindolin-2-yl)cyclopropane-1-carboxylate(11d).** Flash column chromatography to afford product as a white solid (20.1 mg, 41% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.90-7.78 (m, 2H), 7.77-7.66 (m, 2H), 3.76 (s, 3H), 3.37-3.24 (m, 1H), 2.28-2.16 (m, 1H), 1.81-1.71 (m, 1H), 1.68-1.63 (m, 1H).<sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.6, 168.0, 134.2, 131.5, 123.4, 52.2, 29.5, 19.8, 13.6. This compound has been reported in the published literature.<sup>8</sup>



Ethyl (1*S*\*,2*S*\*)-2-(1,3-dioxoisoindolin-2-yl)cyclopropane-1-carboxylate (11e). Flash column chromatography to afford product as a white solid (10.9 mg, 21% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.87-7.77 (m, 2H), 7.75-7.65 (m, 2H), 4.21 (q, *J* = 7.2 Hz, 2H), 3.36-3.25 (m, 1H), 2.26-2.17 (m, 1H), 1.82-1.71 (m, 1H), 1.67-1.62 (m, 1H), 1.31 (t, *J* = 7.2 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  172.1, 168.0, 134.2, 131.6, 123.3, 61.1, 29.5, 19.9, 14.2, 13.5. This compound has been reported in the published literature.<sup>9</sup>



**Diethyl 2-(1,3-dioxoisoindolin-2-yl)cyclopropane-1,1-dicarboxylate (11f).** Flash column chromatography to afford product as a white solid (18.5 mg, 28% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.84-7.79 (m, 2H), 7.74-7.69 (m, 2H), 4.32-4.23 (m, 2H), 4.08-4.00 (m, 2H), 3.71-3.65 (m, 1H), 2.73-2.69 (m, 1H), 2.02-1.97 (m, 1H), 1.32 (t, *J* = 7.1 Hz, 3H), 1.09 (t, *J* = 7.2 Hz, 3H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  168.2, 167.8, 166.4, 134.3, 131.5, 123.4, 62.0, 61.8, 34.7, 33.5, 19.2, 14.0, 13.8. This compound has been reported in the published literature.<sup>10</sup>

*N*-Acetyl-*N*-((1S,2R)-2-methyl-1-phenylcyclopropyl)acetamide (13). Flash column chromatography to afford product as a white oil (34.7 mg, 75% yield). <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.33-7.26 (m, 2H), 7.18 (t, *J* = 7.4 Hz, 1H), 6.98-6.90 (m, 2H), 2.44 (s, 3H), 2.33 (s, 3H), 1.94-1.84 (m, 1H), 1.68-1.63 (m, 1H), 1.18 (d, *J* = 6.2 Hz, 3H), 1.02-0.95 (m, 1H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  175.7, 174.5, 142.1, 128.6, 126.1, 123.2, 44.3, 29.3, 27.6, 26.6, 26.5, 14.9. HRMS (ESI) [M+Na]<sup>+</sup>: calculated for C<sub>14</sub>H<sub>17</sub>NO<sub>2</sub>Na: 254.1157, found 254.1151.

### 4. Procedure of Deprotection of the Boc Group



To a solution of **3a** (55.03 mg, 0.2 mmol) in  $CH_2Cl_2$  (2 mL) was added ZnBr<sub>2</sub> (90 mg, 0.4 mmol). The reaction mixture was stirred at room temperature for 5 h. Evaporation to remove solvent and purification of the resulting crude residue by column chromatography on silica gel afforded *N*-(1-phenylcyclopropyl)acetamide **3b** (29.7 mg, 85%) as a colorless oil.

<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.68 (s, 1H), 7.36-7.29 (m, 2H), 7.28-7.12 (m, 8H), 6.87 (s, 1H), 2.04 (s, 6H), 1.43-1.31 (m, 4H), 1.27-1.24 (m, 2H), 1.23-1.18 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  178.9, 172.5, 141.7, 141.0, 128.8, 128.3, 126.8, 126.5, 125.7, 124.2, 37.1, 35.3, 23.4, 21.5, 19.6, 17.5. This compound has been reported in the published literature.<sup>11</sup>

### 5. Procedure of the Deprotection of Phthaloyl Group



To a stirred solution of 2-(2-cyclohexyl-1-phenylethyl)isoindoline-1,3-dione **9e** (59.4 mg, 0.2 mmol) in EtOH (0.4 mL) was added hydrazine hydrate (w = 80%, 0.18 mL). The mixture was refluxed for 5 h. After completion, the solvent was evaporated in vacuo and the residue was purified by flash column chromatography over neutral  $Al_2O_3$  to give the product 1-(4-chlorophenyl)cyclopropan-1-amine **14** (24.4 mg 73% yield) as a colorless oil.

<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>)  $\delta$  7.25 (q, *J* = 8.6 Hz, 4H), 2.35 (br, 2H), 1.09 (m, 2H), 0.96 (m, 2H). <sup>13</sup>C NMR (126 MHz, CDCl<sub>3</sub>)  $\delta$  144.9, 131.8, 128.4, 127.0, 36.4, 17.7. This compound has been reported in the published literature.<sup>12</sup>

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# 7. NMR Spectra of New Compounds





f1 (ppm) . \_ 





f1 (ppm) . \_ 





f1 (ppm) . \_ 





\_\_\_\_ f1 (ppm)




210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10 f1 (ppm)





180 170 f1 (ppm) . \_ 









f1 (ppm) -180 170 



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130 120 f1 (ppm) 





-10 190 180 170 150 140 130 120 110 f1 (ppm) 









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