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Supporting Information

Synthesis and Antimalarial Activity of Novel Bicyclic and Tricyclic Aza-peroxides


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General experimental conditions:
All glass apparatus were oven dried prior to use. Melting points were taken in open capillaries on complab melting point apparatus and are presented uncorrected. Infrared spectra were recorded on a Perkin-Elmer FT-IR RXI spectrophotometer. $^1$H NMR and $^{13}$C NMR spectra were recorded using CDCl$_3$ as solvent either using Bruker Supercon Magnet DPX-200 or DRX-300 spectrometers (operating at 200 and 300 MHz respectively for $^1$H; 50 and 75 MHz respectively for $^{13}$C) or using JEOL ECS-400 spectrometer (operating at 400 MHz for $^1$H and 100 MHz for $^{13}$C). Tetramethylsilane (δ 0.00 ppm) served as an internal standard in $^1$H NMR and CDCl$_3$ (δ 77.0 ppm) in $^{13}$C NMR. Chemical shifts are reported in parts per million. Splitting patterns are described as singlet (s), doublet (d), triplet (t) and multiplet (m). Fast Atom Bombardment Mass Spectra (FAB-MS) were obtained on JEOL SX-102/DA-6000 mass spectrometer using argon/xenon (6 kV, 10 mA) as the FAB gas. Glycerol or m-nitrobenzyl alcohol was used as matrix. Electrospray mass spectrometry (ES-MS) were recorded on a MICROMASS QUATTRO II triple quadruple mass spectrometer. High Resolution Electron Impact Mass Spectra (HR-EIMS) were obtained on JEOL MS route 600H instrument as well as Xevo G2-S Q-Tof (Waters, USA). Elemental analyses were performed on Vario EL-III C H N S analyzer (Germany) and values were within ±0.4 % of the calculated values, and therefore these compounds meet the criteria of ≥95% purity. Column chromatography was performed over Merck silica gel (particle size: 60-120 Mesh and 100-200 Mesh) procured from Qualigens™ (India), flash silica gel (particle size: 230-400 Mesh). All chemicals and reagents were obtained from Sigma Aldrich (USA), Merck (India) or Spectrochem (India) and were used without further purification.

Melting point and Yields of Intermediate Compounds
Table 1. Melting points and yields of N-substituted succinimides 2a-h and N-substituted phthalimides 6a-h.

<table>
<thead>
<tr>
<th>Entry</th>
<th>Ar =</th>
<th>m.p. (ºC)</th>
<th>Yield (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2a</td>
<td>C$_6$H$_5$-</td>
<td>129-131</td>
<td>71</td>
</tr>
<tr>
<td>2b</td>
<td>4-fluoro-C$_6$H$_4$-</td>
<td>72-74</td>
<td>74</td>
</tr>
<tr>
<td>2c</td>
<td>4-chloro-C$_6$H$_4$-</td>
<td>72-74</td>
<td>60</td>
</tr>
<tr>
<td>2d</td>
<td>4-bromo-C$_6$H$_4$-</td>
<td>120-122</td>
<td>69</td>
</tr>
<tr>
<td>2e</td>
<td>4-methyl-C$_6$H$_4$-</td>
<td>83-85</td>
<td>73</td>
</tr>
<tr>
<td>2f</td>
<td>4-methoxy-C$_6$H$_4$-</td>
<td>130-132</td>
<td>74</td>
</tr>
<tr>
<td>2g</td>
<td>4-cyclohexyl-C$_6$H$_4$-</td>
<td>95-97</td>
<td>67</td>
</tr>
<tr>
<td>2h</td>
<td>4-phenyl-C$_6$H$_4$-</td>
<td>124-126</td>
<td>67</td>
</tr>
<tr>
<td>6a</td>
<td>C$_6$H$_5$-</td>
<td>94-96</td>
<td>71</td>
</tr>
<tr>
<td>6b</td>
<td>4-fluoro-C$_6$H$_4$-</td>
<td>87-89</td>
<td>86</td>
</tr>
<tr>
<td>6c</td>
<td>4-chloro-C$_6$H$_4$-</td>
<td>100-102</td>
<td>66</td>
</tr>
<tr>
<td>6d</td>
<td>4-bromo-C$_6$H$_4$-</td>
<td>125-127</td>
<td>69</td>
</tr>
<tr>
<td>6e</td>
<td>4-methyl-C$_6$H$_4$-</td>
<td>88-91</td>
<td>81</td>
</tr>
<tr>
<td>6f</td>
<td>4-methoxy-C$_6$H$_4$-</td>
<td>99-102</td>
<td>80</td>
</tr>
<tr>
<td>6g</td>
<td>4-cyclohexyl-C$_6$H$_4$-</td>
<td>106-108</td>
<td>83</td>
</tr>
<tr>
<td>6h</td>
<td>4-phenyl-C$_6$H$_4$-</td>
<td>126-128</td>
<td>74</td>
</tr>
</tbody>
</table>
Table 2. Melting points and yields of hydroxy-functionalized lactams 3a-h and 7a-h.

<table>
<thead>
<tr>
<th>Entry</th>
<th>Ar =</th>
<th>m.p. (°C)</th>
<th>Yield (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3a</td>
<td>C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;5&lt;/sub&gt;-</td>
<td>oil</td>
<td>67</td>
</tr>
<tr>
<td>3b</td>
<td>4-fluoro-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>75-77</td>
<td>64</td>
</tr>
<tr>
<td>3c</td>
<td>4-chloro-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>oil</td>
<td>63</td>
</tr>
<tr>
<td>3d</td>
<td>4-bromo-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>130-132</td>
<td>71</td>
</tr>
<tr>
<td>3e</td>
<td>4-methyl-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>oil</td>
<td>73</td>
</tr>
<tr>
<td>3f</td>
<td>4-methoxy-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>82-84</td>
<td>64</td>
</tr>
<tr>
<td>3g</td>
<td>4-cyclohexyl-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>125-127</td>
<td>63</td>
</tr>
<tr>
<td>3h</td>
<td>4-phenyl-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>152-154</td>
<td>61</td>
</tr>
<tr>
<td>7a</td>
<td>C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;5&lt;/sub&gt;-</td>
<td>114-116</td>
<td>66</td>
</tr>
<tr>
<td>7b</td>
<td>4-fluoro-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>96-98</td>
<td>69</td>
</tr>
<tr>
<td>7c</td>
<td>4-chloro-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>130-132</td>
<td>59</td>
</tr>
<tr>
<td>7d</td>
<td>4-bromo-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>148-150</td>
<td>63</td>
</tr>
<tr>
<td>7e</td>
<td>4-methyl-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>106-108</td>
<td>67</td>
</tr>
<tr>
<td>7f</td>
<td>4-methoxy-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>117-119</td>
<td>74</td>
</tr>
<tr>
<td>7g</td>
<td>4-cyclohexyl-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>99-101</td>
<td>71</td>
</tr>
<tr>
<td>7h</td>
<td>4-phenyl-C&lt;sub&gt;6&lt;/sub&gt;H&lt;sub&gt;4&lt;/sub&gt;-</td>
<td>166-168</td>
<td>58</td>
</tr>
</tbody>
</table>

General procedure for preparation of N-substituted imides 2a-h and 6a-h: Preparation of 2a as a representative:

To a stirred mixture of allylic alcohol 1a (2 g, 13.5 mmol), succinimide (2.67 g, 26.99 mmol) and triphenylphosphine (8.85 g, 33.74 mmol) in dry THF (60 mL) at 0 °C and under inert atmosphere, was added diethylazodicarboxylate (DEAD) (4.3 mL, 26.99 mmol) dissolved in dry THF (5 mL). The contents were stirred at 0 °C for 30 min. THF was evaporated under reduced pressure. The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1.5% ethyl acetate: DCM as eluant to furnish 2.2 g (71% yield) of 2a.

**Compound 2a: (1-(3-Phenyl-but-2-enyl)-pyrrolidine-2,5-dione)** Yield: 71%; Solid, mp 129-131 °C;

FT-IR (KBr, cm<sup>-1</sup>) 1724.7, 1525.3, 1369.5, 1209.5, 766.1;

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 2.12 (s, 3H), 2.64 (s, 4H), 4.23 (d, 2H, J = 6.8 Hz), 5.63-5.67 (m, 1H), 7.17-7.30 (m, 5H, Ar);

<sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>) δ 16.23 (CH<sub>3</sub>), 28.35 (2 × CH<sub>2</sub>), 37.27 (CH<sub>2</sub>), 120.43 (CH), 125.93 (2 × CH), 127.48 (CH), 128.31 (2 × CH), 139.84 (C), 142.70 (C), 176.96 (2 × C);

ESI-MS (m/z) 230 [M+H]<sup>+</sup>.

Anal. calc'd for C<sub>14</sub>H<sub>15</sub>NO<sub>2</sub>: C, 73.34%, H, 6.59%, N, 6.11%; found: C, 73.51%, H, 6.61%, N, 6.19%.

N-substituted imides 2b-h were synthesized from allylic alcohols 1b-h, respectively by using the same procedure. Likewise, imides 6a-h were synthesized from allylic alcohols 1a-h by replacing succinimide with phthalimide.
Compound 2b: (1-[3-(4-Fluoro-phenyl)-but-2-enyl]-pyrrolidine-2, 5-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 0.5% ethyl acetate: DCM as eluent.

Yield: 74%; Solid, mp 72-74 ºC;

FT-IR (KBr, cm\(^{-1}\)) 1754.4, 1696.4, 1533.4, 1231.4, 1172.2, 1069.2, 666.4;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 2.15 (s, 3H), 2.71 (s, 4H), 4.27 (d, 2H, \(J = 6.8\) Hz), 5.64-5.67 (m, 1H), 6.94-6.99 (m, 2H, Ar), 7.24-7.32 (m, 2H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.3 (CH\(_3\)), 28.4 (2 × CH\(_2\)), 37.2 (CH\(_2\)), 115.1 (d, 2 × CH, \(J_{CF} = 21\) Hz), 120.3 (CH), 127.5 (d, 2 × CH, \(J_{CF} = 7.6\) Hz), 138.72 (d, C, \(J_{CF} = 2.9\) Hz), 138.8 (C), 162.3 (d, C, \(J_{CF} = 245.1\) Hz), 176.8 (2 × C);

ESI-MS (m/z) 248 [M+H]\(^+\).

Anal. calcd for C\(_{14}\)H\(_{14}\)FNO\(_2\): C, 68.00%, H, 5.71%, N, 5.66%; found: C, 68.25%, H, 5.81%, N, 5.90%.

Compound 2c: (1-[3-(4-Chloro-phenyl)-but-2-enyl]-pyrrolidine-2,5-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 0.5% ethyl acetate: DCM as eluent.

Yield: 60%; Solid, mp 72-74 ºC;

FT-IR (KBr, cm\(^{-1}\)) 1754.4, 1696.9, 1534.3, 1247.8, 1069.4, 667.1;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 2.09 (s, 3H), 2.65 (s, 4H), 4.22 (d, 2H, \(J = 6.4\) Hz), 5.64 (s, 1H), 7.20-7.26 (m, 4H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.2 (CH\(_3\)), 28.4 (2 × CH\(_2\)), 37.1 (CH\(_2\)), 120.9 (CH), 127.2 (2 × CH), 128.4 (2 × CH), 133.3 (C), 138.7 (C), 141.0 (C), 176.9 (2 × C);

ESI-MS (m/z) 264 [M+H]\(^+\).

Anal. calcd for C\(_{14}\)H\(_{14}\)ClNO\(_2\): C, 63.76%, H, 5.35%, N, 5.31%; found: C, 63.90%, H, 5.40%, N, 5.51%.

Compound 2d: (1-[3-(4-Bromo-phenyl)-but-2-enyl]-pyrrolidine-2,5-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1.5% ethyl acetate: DCM as eluent.

Yield: 69%; Solid, mp 120-122 ºC;

FT-IR (KBr, cm\(^{-1}\)) 1728.2, 1596.9, 1218.5, 1048.4, 769.0;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 2.15 (s, 3H), 2.71 (s, 4H), 4.27 (d, 2H, \(J = 7.2\) Hz), 5.68-5.71 (m, 1H), 7.20-7.28 (m, 2H, Ar), 7.34-7.41 (m, 2H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.1 (CH\(_3\)), 28.4 (2 × CH\(_2\)), 37.1 (CH\(_2\)), 121.0 (CH), 121.4 (C), 127.5 (2 × CH), 131.4 (2 × CH), 138.8 (C), 141.5 (C), 176.9 (2 × C);

ESI-MS (m/z) 325 [M+NH\(_4\)]\(^+\).
Anal. calcd for C_{14}H_{14}BrNO_2: C, 54.56%, H, 4.58%, N, 4.55%; found: C, 54.73%, H, 4.68%, N, 4.50%.

**Compound 2e**: (1-(3-p-Tolyl-but-2-enyl)-pyrrolidine-2,5-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 0.5% ethyl acetate: DCM as eluent.

Yield: 73%; Solid, mp 83-85 ºC;

FT-IR (KBr, cm\(^{-1}\)) 1703.5, 1427.7, 1216.7, 760.1;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 2.10 (s, 3H), 2.25 (s, 3H), 2.64 (s, 4H), 4.22 (d, 2H, \(J = 7.2\) Hz), 5.61-5.65 (m, 1H), 7.03 (d, 2H, Ar, \(J = 8.0\) Hz), 7.19 (d, 2H, Ar, \(J = 8.0\) Hz);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.19 (CH\(_3\)), 21.13 (CH\(_3\)), 28.35 (2 × CH\(_2\)), 37.28 (CH\(_2\)), 119.57 (CH), 125.78 (2 × CH), 129.00 (2 × CH), 137.26 (C), 139.65 (C), 139.78 (C), 176.98 (2 × C);

ESI-MS (m/z) 244 [M+H]\(^+\).

Anal. calcd for C_{15}H_{15}NO_2: C, 74.05%, H, 7.04%, N, 5.76%; found: C, 74.35%, H, 7.34%, N, 5.66%.

**Compound 2f**: (1-[3-(4-Methoxy-phenyl]-but-2-enyl]-pyrrolidine-2,5-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1.0% ethyl acetate: DCM as eluent.

Yield: 74%; Solid, mp 130-132 ºC;

FT-IR (KBr, cm\(^{-1}\)) 1754.3, 1697.5, 1534.0, 1248.6, 1176.6, 1068.0, 664.5;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 2.10 (s, 3H), 2.64 (s, 4H), 3.73 (s, 3H), 4.22 (d, 2H, \(J = 7.2\) Hz), 5.59-5.62 (m, 1H), 6.75-6.77 (m, 2H, Ar), 7.22-7.25 (m, 2H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.20 (CH\(_3\)), 28.35 (2 × CH\(_2\)), 37.31 (CH\(_2\)), 55.37 (CH\(_3\)), 113.65 (2 × CH), 118.77 (CH), 127.01 (2 × CH), 135.11 (C), 139.17 (C), 159.14 (C), 177.02 (2 × C);

ESI-MS (m/z) 260 [M+H]\(^+\).

Anal. calcd for C_{15}H_{16}NO_3: C, 69.48%, H, 6.61%, N, 5.40%; found: C, 69.78%, H, 6.81%, N, 5.74%.

**Compound 2g**: (1-[3-(4-Cyclohexyl-phenyl]-but-2-enyl]-pyrrolidine-2,5-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using DCM as eluent.

Yield: 67%; Solid, mp 95-97 ºC;

FT-IR (KBr, cm\(^{-1}\)) 1753.8, 1679.6, 1537.0, 1245.7, 1172.2, 1068.1, 764.5;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 1.34-1.53 (m, 6H), 1.79-1.82 (m, 4H), 2.17 (s, 3H), 2.43-2.48 (m, 1H), 2.69 (s, 4H), 4.28 (d, 2H, \(J = 7.2\) Hz), 5.68-5.72 (m, 1H), 7.13 (d, 2H, Ar, \(J = 8.4\) Hz), 7.27-7.29 (m, 2H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.14 (CH\(_3\)), 26.21 (CH\(_2\)), 26.95 (2 × CH\(_2\)), 28.34 (2 × CH\(_2\)), 34.47 (2 × CH\(_2\)), 37.29 (CH\(_2\)), 44.27 (CH), 119.64 (CH), 125.79 (2 × CH), 126.76 (2 × CH), 139.65 (C), 140.10 (C), 147.55 (C), 176.98 (2 × C);
ESI-MS (m/z) 312 [M+H]⁺.
Anal. calcd for C₂₀H₂₅NO₂: C, 77.14%, H, 8.09%, N, 4.50%; found: C, 77.04%, H, 8.29%, N, 4.80%.

 Compound 2h: (1-(3-Biphenyl-4-yl-but-2-enyl)-pyrrolidine-2,5-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using DCM as eluent.
Yield: 67%; Solid, mp 124-126 ºC;
FT-IR (KBr, cm⁻¹) 1731.0, 1513.9, 1373.5, 1219.1, 767.1;
¹H NMR (400 MHz, CDCl₃) δ 2.22 (s, 3H), 2.71 (s, 4H), 4.31 (d, 2H, J = 7.2 Hz), 5.77-5.80 (m, 1H), 7.32-7.58 (m, 9H, Ar);
¹³C NMR (100 MHz, CDCl₃) δ 16.17 (CH₃), 28.37 (2 × CH₂), 37.30 (CH₂), 120.43 (CH), 126.32 (2 × CH), 127.02 (2 × CH), 127.05 (2 × CH), 127.38 (CH), 128.86 (2 × CH), 139.34 (C), 140.31 (C), 140.73 (C), 141.54 (C), 177.01 (2 × C);
ESI-MS (m/z) 306 [M+H]⁺.
Anal. calcd for C₂₀H₂₁NO₂: C, 78.66%, H, 6.27%, N, 4.59%; found: C, 78.80%, H, 6.30%, N, 4.71%.

 Compound 6a: (2-(3-Phenyl-but-2-enyl)-isoindole-1,3-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 4.0% ethyl acetate: n-Hexane as eluent.
Yield: 71%; Solid, mp 94-96 ºC;
FT-IR (KBr, cm⁻¹) 1766.4, 1713.2, 1598.8, 1386.1, 1071.9, 716.0;
¹H NMR (400 MHz, CDCl₃) δ 2.18 (s, 3H), 4.41 (d, 2H, J = 7.2 Hz), 5.75-5.79 (m, 1H), 7.18-7.29 (m, 5H, Ar), 7.62-7.65 (m, 2H, Ar), 7.76-7.78 (m, 2H, Ar);
¹³C NMR (100 MHz, CDCl₃) δ 16.22 (CH₃), 36.39 (CH₂), 121.25 (CH), 123.32 (2 × CH), 125.95 (2 × CH), 127.42 (CH), 128.28 (2 × CH), 132.35 (2 × C), 133.99 (2 × CH), 139.47 (C), 140.62 (C), 168.17 (2 × C);
ESI-MS (m/z) 294 [M+H]⁺; EI-HRMS calcd for C₁₈H₁₆NO₃: 294.11302; found: 294.11211.
ESI-MS (m/z) 278 [M+H]⁺, 279 [M+2H]⁺.
Anal. calcd for C₁₈H₁₄NO₂: C, 78.00%, H, 5.40%, N, 5.35%; found: C, 78.00%, H, 5.40%, N, 5.35%.

 Compound 6b: (2-[3-(4-Fluoro-phenyl)-but-2-enyl]-isoindole-1,3-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 2.0% ethyl acetate: n-Hexane as eluent.
Yield: 86%; Solid, mp 87-89 ºC;
FT-IR (KBr, cm⁻¹) 1766.8, 1715.2, 1394.1, 1066.4, 718.4;
¹H NMR (400 MHz, CDCl₃) δ 2.15 (s, 3H), 4.40 (d, 2H, J = 7.2 Hz), 5.69-5.73 (m, 1H), 6.90-6.93 (m, 2H, Ar), 7.19-7.29 (m, 2H, Ar), 7.63-7.65 (m, 2H, Ar), 7.77-7.78 (m, 2H, Ar);
$^{13}$C NMR (100 MHz, CDCl$_3$) δ 16.36 (CH$_3$), 36.33 (CH$_2$), 115.07 (d, 2 × CH, $J_{C,F} = 21.0$ Hz), 121.19 (CH), 123.34 (2 × CH), 127.53 (d, 2 × CH, $J_{C,F} = 8.6$ Hz), 132.32 (2 × C), 134.02 (2 × CH), 138.48 (C), 138.82 (d, C, $J_{C,F} = 2.9$ Hz), 162.30 (d, C, $J_{C,F} = 245$ Hz), 168.16 (2 × C);

ESI-MS (m/z) 296 [M+H]$^+$.  

Anal. calc'd for C$_{18}$H$_{14}$FNO$_2$: C, 73.21%, H, 4.78%, N, 4.74%; found: C, 73.41%, H, 4.86%, N, 4.81%.

**Compound 6c**: (2-[3-(4-Chloro-phenyl)-but-2-enyl]-isoindole-1,3-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 8.0% ethyl acetate: n-Hexane as eluent.

Yield: 66%; Solid, mp 100-102 °C;

FT-IR (KBr, cm$^{-1}$) 1769.4, 1713.1, 1436.8, 1398.3, 1084.1, 719.6;

$^1$H NMR (400 MHz, CDCl$_3$) δ 2.15 (s, 3H), 4.40 (d, 2H, $J = 7.2$ Hz), 5.73-5.76 (m, 1H), 7.17-7.24 (m, 4H, Ar), 7.63-7.66 (m, 2H, Ar), 7.77-7.78 (m, 2H, Ar);

$^{13}$C NMR (100 MHz, CDCl$_3$) δ 16.18 (CH$_3$), 36.30 (CH$_2$), 121.76 (CH), 123.36 (2 × CH), 127.25 (2 × CH), 128.40 (2 × CH), 132.29 (2 × C), 133.19 (C), 133.05 (2 × CH), 138.34 (C), 141.17 (C), 168.15 (2 × C);

ESI-MS (m/z) 312 [M+H]$^+$.  

Anal. calc'd for C$_{18}$H$_{14}$ClNO$_2$: C, 69.35%, H, 4.53%, N, 4.49%; found: C, 69.65%, H, 4.73%, N, 4.69%.

**Compound 6d**: (2-[3-(4-Bromo-phenyl)-but-2-enyl]-isoindole-1,3-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 8.0% ethyl acetate: n-Hexane as eluent.

Yield: 69%; Solid, mp 125-127 °C;

FT-IR (KBr, cm$^{-1}$) 1771.2, 1712.5, 1592.2, 1216.4, 1077.1, 763.4;

$^1$H NMR (400 MHz, CDCl$_3$) δ 2.15 (s, 3H), 4.39 (d, 2H, $J = 6.8$ Hz), 5.73-5.77 (m, 1H), 7.15-7.34 (m, 4H, Ar), 7.64-7.65 (m, 2H, Ar), 7.77-7.79 (m, 2H, Ar);

$^{13}$C NMR (100 MHz, CDCl$_3$) δ 16.13 (CH$_3$), 36.30 (CH$_2$), 121.36 (C), 121.84 (CH), 123.36 (2 × CH), 127.61 (2 × CH), 131.35 (2 × CH), 132.29 (2 × C), 134.06 (2 × CH), 138.40 (C), 141.64 (C), 168.15 (2 × C);

ESI-MS (m/z) 356 [M+H]$^+$.  

Ana. calc'd for C$_{18}$H$_{14}$BrNO$_2$: C, 60.69%, H, 3.96%, N, 3.93%; found: C, 60.89%, H, 4.23%, N, 3.90%.

**Compound 6e**: (2-(3-p-Tolyl-but-2-enyl)-isoindole-1,3-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 4.0% ethyl acetate: n-Hexane as eluent.

Yield: 81%; Solid, mp 88-91 °C;

FT-IR (KBr, cm$^{-1}$) 1767.8, 1713.6, 1510.0, 1432.2, 1393.6, 1065.4, 712.7;
\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta 2.15\) (s, 3H), 2.23 (s, 3H), 4.39 (d, 2H, \(J = 7.6\) Hz), 5.71-5.75 (m, 1H), 7.02 (d, 2H, Ar, \(J = 8.0\) Hz), 7.19 (d, 2H, Ar, \(J = 8.0\) Hz), 7.60-7.64 (m, 2H, Ar), 7.73-7.78 (m, 2H, Ar); 

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta 16.19\) (CH\(_3\)), 21.13 (CH\(_2\)), 36.40 (CH\(_2\)), 120.41 (CH), 123.30 (2 × CH), 125.80 (2 × CH), 128.98 (2 × CH), 132.36 (2 × C), 133.97 (2 × CH), 137.17 (C), 139.26 (C), 139.88 (C), 168.19 (2 × C); 

ESI-MS (m/z) 308 [M+H]+.

Anal. calcd for C\(_{19}\)H\(_{17}\)NO\(_2\): C, 78.33%, H, 5.88%, N, 4.81%; found: C, 78.63%, H, 5.90%, N, 4.89%.

**Compound 6f:** (2-[3-(4-Methoxy-phenyl)-but-2-nyl]-isoindole-1,3-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 4.0% ethyl acetate: n-Hexane as eluent.

Yield: 80%; Solid, mp 99-102 °C;

FT-IR (KBr, cm\(^{-1}\)) 1771.5, 1712.5, 1603.9, 1513.9, 1216.1, 761.3;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta 2.15\) (s, 3H), 3.71 (s, 3H), 4.39 (d, 2H, \(J = 7.6\) Hz), 5.68-5.72 (m, 1H), 6.73-6.77 (m, 2H, Ar), 7.19-7.25 (m, 2H, Ar), 7.62-7.77 (m, 4H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta 16.20\) (CH\(_3\)), 36.42(CH\(_2\)), 55.35 (CH\(_3\)), 113.62 (2 × CH), 119.63 (CH), 123.30 (2 × CH), 127.02 (2 × CH), 132.37 (2 × C), 133.97 (2 × CH), 135.24 (2 × C), 138.79 (C), 159.09 (C), 168.21 (2 × C);

ESI-MS (m/z) 308 [M+H]+.

Anal. calcd for C\(_{19}\)H\(_{17}\)NO\(_3\): C, 74.25%, H, 5.58%, N, 4.56%; found: C, 74.44%, H, 5.78%, N, 4.76%.

**Compound 6g:** (2-[3-(4-Cyclohexyl-phenyl)-but-2-nyl]-isoindole-1,3-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 8.0% ethyl acetate: n-Hexane as eluent.

Yield: 83%; Solid, mp 106-108 °C;

FT-IR (KBr, cm\(^{-1}\)) 1771.2, 1709.0, 1653.2, 1387.7, 1072.5, 721.9;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta 1.24-1.39\) (m, 5H), 1.70-1.81 (m, 5H), 2.24 (s, 3H), 2.42-2.47 (m, 1H), 4.45 (d, 2H, \(J = 6.8\) Hz), 5.78-5.80 (m, 1H), 7.11 (d, 2H, Ar, \(J = 8.0\) Hz), 7.29 (d, 2H, Ar, \(J = 8.4\) Hz), 7.68-7.84 (m, 4H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta 16.14\) (CH\(_3\)), 26.22 (CH\(_2\)), 26.95 (2 × CH\(_2\)), 34.46 (2 × CH\(_2\)), 36.41(CH\(_2\)), 44.27 (CH), 120.48 (CH), 123.29 (2 × CH), 125.82 (2 × CH), 126.74 (2 × CH), 132.36 (2 × C), 133.97 (2 × CH\(_2\)), 139.27 (C), 140.21 (C), 147.47 (C), 168.19 (2 × C);

ESI-MS (m/z) 360 [M+H]+.

Anal. calcd for C\(_{24}\)H\(_{22}\)NO\(_2\): C, 80.19%, H, 7.01%, N, 3.90%; found: C, 80.39%, H, 7.21%, N, 4.11%.

**Compound 6h:** (2-(3-Biphenyl-4-yl-but-2-nyl)-isoindole-1,3-dione) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 5.0% ethyl acetate: n-Hexane as eluent.
Yield: 74%; Solid, mp 126-128 °C;

FT-IR (KBr, cm⁻¹) 1705.1, 1632.5, 1430.5, 1070.3, 715.0;

¹H NMR (400 MHz, CDCl₃) δ 2.27 (s, 3H), 4.49 (d, 2H, J = 7.2 Hz), 5.87-5.91 (m, 1H), 7.29-7.33 (m, 1H, Ar), 7.39-7.45 (m, 4H, Ar), 7.51-7.57 (m, 4H, Ar), 7.69-7.72 (m, 2H, Ar), 7.82-7.85 (m, 2H, Ar);

¹³C NMR (100 MHz, CDCl₃) δ 16.16 (CH₃), 36.41(CH₂), 121.27 (CH), 123.35 (2 × CH), 126.33 (2 × CH), 127.00 (2 × CH), 127.05 (2 × CH), 127.34 (CH), 128.83 (2 × CH), 132.34 (2 × C), 134.02 (2 × CH), 138.95 (C), 140.23 (C), 140.76 (C), 141.64 (C), 168.20 (2 × C);

ESI-MS (m/z) 354 [M+H]+.

Anal. calcd for C₂₄H₁₉NO₂: C, 81.56%, H, 5.42%, N, 3.96%; found: C, 81.50%, H, 5.53%, N, 4.18%.

General procedure for preparation of amides 3a-h and 7a-h: Preparation of 3a as a representative:

To the solution of N-substituted imide 2a (2 g, 8.73 mmol) in MeOH: THF (1:1, 40 mL) at 0 ºC, was added sodium borohydride (1.3 g, 34.93 mmol) in 5 fractions over a period of 2 h with continuous stirring. Reaction mixture was stirred at 0 ºC for 1 h. MeOH: THF was evaporated under reduced pressure to half of the original volume. Water (40 mL) was added and extracted with CHCl₃ (2 × 100 mL). Organic layer was dried over anhyd sodium sulphate and concentrated under reduced pressure. The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1% Methanol: DCM as eluant to furnish 1.4 g (67% yield) of 3a.

Compound 3a: (5-Hydroxy-1-(3-phenyl-but-2-etyl)-pyrrolidin-2-one)
Yield: 67%; Viscous oil; FT-IR (neat, cm⁻¹) 3776.4, 1687.5, 1660.4, 1209.7, 768.3;

¹H NMR (400 MHz, CDCl₃) δ 1.63-1.85 (m, 1H), 2.06 (s, 3H), 2.23-2.27 (m, 2H), 2.46-2.55(m, 1H), 2.98(s,1OH), 3.91-3.95 (m, 1H), 4.24 (dd, 1H, J = 6 and 15.2 Hz), 5.16 (s, 1H), 5.64-5.67 (m, 1H), 7.17-7.30 (m, 5H, Ar)

¹³C NMR (100 MHz, CDCl₃) δ 16.16 (CH₃), 28.33 (CH₂), 28.95 (CH₂), 38.25 (CH₂), 83.06 (CH), 121.94 (CH), 125.83 (2 × CH), 127.47 (CH), 128.38 (2 × CH), 139.42 (C), 142.74 (C), 174.50 (C);

ESI-MS (m/z) 232 [M+H]+.

Anal. calcd for C₁₄H₁₇NO₂: C, 72.70%, H, 7.41%, N, 6.06%; found: C, 72.85%, H, 7.56%, N, 6.25%.

Amides 3b-h and 7a-h were prepared from N-substituted imides 2b-h and 6a-h, respectively by using the same procedure.

Compound 3b: (1-[3-(4-Fluoro-phenyl)-but-2-etyl]-5-hydroxy-pyrrolidin-2-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1% Methanol: DCM as eluent.

Yield: 64%; Solid, mp 75-77 °C;
FT-IR (KBr, cm⁻¹) 3161.9, 1648.5, 1511.9, 1470.6, 1238.4, 1075.5, 825.8, 670.3;

¹H NMR (400 MHz, CDCl₃) δ 1.87-1.93 (m, 1H), 2.16 (s, 3H), 2.26-2.38 (m, 2H), 2.53-2.61 (m, 1H), 2.82 (br, 1OH), 3.97-4.01 (m, 1H), 4.26-4.31 (m, 1H), 5.21-5.24 (m, 1H), 5.66-5.71 (m, 1H), 6.96-7.00 (m, 2H, Ar), 7.31-7.33 (m, 2H, Ar);

¹³C NMR (100 MHz, CDCl₃) δ 16.22 (CH₃), 28.41 (CH₂), 28.90(CH₂) 38.28 (CH), 83.12(CH), 115.16 (d, 2 × CH, Jₑ₋F = 20.9 Hz), 121.93 (CH), 127.40 (d, 2 × CH, Jₑ₋F = 7.7 Hz ), 138.36 (CH), 138.77 (d, C, Jₑ₋F = 2.9 Hz), 162.30 (d, C, Jₑ₋F = 242.11 Hz), 174.38 (C);

ESI-MS (m/z) 250 [M+H]⁺.

Anal. calcd for C₁₃H₁₆FNO₂: C, 67.45%, H, 6.47%, N, 5.62%; found: C, 67.60%, H, 6.72%, N, 5.50%.

**Compound 3c:** (1-[3-(4-Chloro-phenyl)-but-2-enyl]-5-hydroxy-pyrrolidin-2-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1% Methanol: DCM as eluent.

Yield: 63%; Viscous oil; FT-IR (neat, cm⁻¹) 3397.8, 3020.3, 1684.6, 1450.1, 1215.9, 759.9;

¹H NMR (200 MHz, CDCl₃) δ 1.83-1.98 (m, 1H), 2.08 (s, 3H), 2.17-2.59 (m, 3H), 3.92 (dd, 1H, J = 15.1 and 8.1Hz), 4.24 (dd, 1H, J = 15.3 and 6.0 Hz), 4.76 (brs, 1H), 5.16 (brs, 1H), 5.70 (t, 1H, J = 6.5 Hz), 7.20-7.30 (m, 4H, Ar);

¹³C NMR (100 MHz, CDCl₃) δ 16.04 (CH₃), 28.44 (CH₂), 28.87 (CH₂), 38.30 (CH₂), 83.15 (CH), 122.58 (CH), 127.12 (2 × CH), 128.48 (2 × CH), 133.24 (C), 138.20 (C), 141.10 (C), 174.37 (C);

ESI-MS (m/z) 266 [M+H]⁺.

Anal. calcd for C₁₄H₁₆ClNO₂: C, 63.28%, H, 6.07%, N, 5.22%; found: C, 63.39%, H, 6.21%, N, 5.41%.

**Compound 3d:** (1-[3-(4-Bromo-phenyl)-but-2-enyl]-5-hydroxy-pyrrolidin-2-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1% Methanol: DCM as eluent.

Yield: 71%; Solid, mp 130-132 °C; FT-IR (KBr, cm⁻¹) 3377.7, 1699.0, 1444.1, 1234.5, 767.9;

¹H NMR (400 MHz, CDCl₃) δ 1.87-1.93 (m, 1H), 2.09 (s, 3H), 2.29-2.32 (m, 2H), 2.54-2.60 (m, 1H), 3.31 (d, 1OH, J = 8 Hz), 3.97-3.99 (m, 1H), 4.24-4.29 (m, 1H), 5.19-5.22 (m, 1H), 5.70-5.73 (m, 1H), 7.20-7.41 (m, 4H, Ar);

¹³C NMR (100 MHz, CDCl₃) δ 16.00 (CH₃), 28.38 (CH₂), 28.92 (CH₂), 38.28 (CH₂), 83.12 (CH), 121.36 (CH), 122.65 (C), 127.47 (2 × CH), 131.43 (2 × CH), 138.20 (C), 141.59 (C), 174.54 (C);

ESI-MS (m/z) 310 [M+H]⁺.

Anal. calcd for C₁₄H₁₆BrNO₂: C, 54.21%, H, 5.20%, N, 4.52%; found: C, 54.15%, H, 5.35%, N, 4.62%.

**Compound 3e:** (5-Hydroxy-1-(3-p-tolyl-but-2-enyl)-pyrrolidin-2-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1.5% Methanol: DCM as eluent.
Compound 3f: (5-Hydroxy-1-[3-(4-methoxy-phenyl)-but-2-enyl]-pyrroloidin-2-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1% Methanol: DCM as eluent.

Yield: 64%; Solid, mp 82-84 °C;

FT-IR (KBr, cm\(^{-1}\)) 3452.7, 1748.5, 1656.0, 1224.3, 1066.7, 772.5;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 1.76-1.93 (m, 1H), 2.10 (s, 3H), 2.29-2.31 (m, 2H), 2.55-2.60 (m, 1H), 2.80 (br, 1OH), 3.79 (s, 3H), 3.98 (dd, 1H, \(J = 8.4\) and 15.2 Hz), 4.27-4.30 (m, 1H), 5.21-5.24 (m, 1H), 5.65-5.69 (m, 1H), 6.81-6.84 (m, 2H, Ar), 7.23-7.32 (m, 2H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.06 (CH\(_3\)), 28.32 (CH\(_2\)), 28.94 (CH\(_2\)), 38.26 (CH\(_2\)), 55.39 (CH\(_3\)), 83.05 (CH), 113.71 (2 × CH), 120.22 (CH), 126.90 (2 × CH), 135.13 (C), 138.83 (C), 159.13 (C), 174.38 (C);

ESI-MS (m/z) 262 [M+H]\(^+\).

Anal. calcd for C\(_{13}\)H\(_{15}\)NO\(_2\): C, 73.44%, H, 7.81%, N, 5.71%; found: C, 73.62%, H, 7.97%, N, 5.91%.

Compound 3g: (1-[3-(4-Cyclohexyl-phenyl)-but-2-enyl]-5-hydroxy-pyrroloidin-2-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1% Methanol: DCM as eluent.

Yield: 63%; Solid, mp 125-127 °C;

FT-IR (KBr, cm\(^{-1}\)) 3391.1, 1672.6, 1451.8, 1216.1, 754.1;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 1.21-1.24 (m, 1H), 1.35-1.44 (m, 4H), 1.74-1.90 (m, 6H), 2.11 (s, 3H), 2.25-2.36 (m, 2H), 2.44-2.61 (m, 2H), 2.76-2.78 (m, 1OH), 3.95-4.01 (m, 1H), 4.28-4.33 (m, 1H), 5.20-5.22 (m, 1H), 5.70-5.73 (m, 1H), 7.14 (d, 2H, Ar, \(J = 8.4\) Hz), 7.30 (d, 2H, Ar, \(J = 8.4\) Hz);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.01 (CH\(_3\)), 26.21 (CH\(_2\)), 26.95 (2 × CH\(_2\)), 28.31 (CH\(_2\)), 28.94 (CH\(_2\)), 34.48 (2 × CH\(_2\)), 38.24 (CH\(_2\)), 44.27 (CH), 83.03 (CH), 121.05 (CH), 125.71 (2 × CH), 126.84 (2 × CH), 139.34 (C), 140.09 (C), 147.58 (C), 174.38 (C);

ESI-MS (m/z) 314 [M+H]\(^+\).

Anal. calcd for C\(_{20}\)H\(_{27}\)NO\(_2\): C, 76.64%, H, 8.68%, N, 4.47%; found: C, 76.81%, H, 8.61%, N, 4.61%.
**Compound 3h:** (1-(3-Biphenyl-4-yl-but-2-enyl)-5-hydroxy-pyrrolidin-2-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 1.5% Methanol: DCM as eluent.

Yield: 61%; Solid, mp 152-154 °C;

FT-IR (KBr, cm⁻¹) 3345.3, 1650.5, 1478.9, 1323.1, 1080.0, 760.0;

¹H NMR (400 MHz, CDCl₃) δ 1.88-1.90 (m, 1H), 2.12 (s, 3H), 2.17-2.37 (m, 2H), 2.52-2.60 (m, 2H), 4.00-4.06 (m, 1OH), 4.31-4.35 (m, 1H), 5.25-5.26 (m, 1H), 5.80-5.83 (m, 1H), 7.40-7.59 (m, 9H, Ar);

¹³C NMR (100 MHz, CDCl₃) δ 16.03 (CH₃), 28.41 (CH₂), 28.90 (CH₂), 38.33 (CH₂), 83.12 (CH), 121.99 (CH), 126.22 (2 × CH), 127.05 (4 × CH), 127.42 (CH), 128.88 (2 × CH), 138.94 (C), 140.33 (C), 140.68 (C), 141.53(C), 174.32 (C);

ESI-MS (m/z) 308 [M+H]⁺.

Anal. calcd for C₂₀H₂₁NO₂: C, 78.15%, H, 6.89%, N, 4.56%; found: C, 78.31%, H, 6.91%, N, 4.63%.

**Compound 7a:** (3-Hydroxy-2-(3-phenyl-but-2-enyl)-2,3-dihydro-isoindol-1-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 15% Ethyl acetate: n-hexane as eluent.

Yield: 66%; Solid, mp 114-116 °C;

FT-IR (KBr, cm⁻¹) 3323.6, 1666.8, 1616.7, 1433.6, 1054.9, 742.2;

¹H NMR (400 MHz, CDCl₃) δ 2.19 (s, 3H), 3.01 (d, 1OH, J = 12 Hz), 4.05-4.11 (m, 1H), 4.36-4.41 (m, 1H), 5.75-5.78 (m, 2H), 7.20-7.69 (m, 9H, Ar);

¹³C NMR (100 MHz, CDCl₃) δ 16.12 (CH₃), 37.28 (CH₂), 81.48 (CH), 121.99 (CH), 123.36 (CH), 125.85 (2 × CH), 127.42 (CH), 128.33 (2 × CH), 129.88 (CH), 131.56 (C), 132.36 (CH), 139.54 (C), 142.80 (C), 144.00 (C), 167.16 (C);

ESI-MS (m/z) 280 [M+H]⁺.

Anal. calcd for C₁₈H₁₇NO₂: C, 77.40%, H, 6.13%, N, 5.01%; found: C, 77.49%, H, 6.29%, N, 5.17%.

**Compound 7b:** (2-[3-(4-Fluoro-phenyl)-but-2-enyl]-3-hydroxy-2,3-dihydro-isoindol-1-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 22% Ethyl acetate: n-hexane as eluent.

Yield: 69%; Solid, mp 96-98 °C;

FT-IR (KBr, cm⁻¹) 3228.6, 1667.4, 1598.4, 1057.9, 768,

¹H NMR (400 MHz, CDCl₃) δ 2.15 (s, 3H), 3.40 (br, 1OH), 4.00-4.03 (m, 1H), 4.27 (Br, 1H), 5.69-5.75 (m, 2H), 6.93-6.97 (m, 2H, Ar), 7.28-7.32 (m, 2H, Ar) 7.45-7.62 (m, 4H, Ar);
$^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$ 16.24 (CH$_3$), 37.27 (CH$_2$), 81.51 (CH), 115.12 (d, 2 $\times$ CH, $J_{C-F} = 21.0$ Hz), 121.94 (CH), 123.36 (CH), 123.46 (CH), 127.42 (d, 2 $\times$ CH, $J_{C-F} = 7.7$ Hz), 129.91 (CH), 131.55 (C), 132.39 (CH), 138.51 (C), 138.83 (d, C, $J_{C-F} = 2.8$ Hz), 143.94 (C), 162.27 (d, C, $J_{C-F} = 245$ Hz), 167.17 (C); ESI-MS (m/z) 298 [M+H]$^+$. 

Anal. calcd for C$_{18}$H$_{18}$FNO$_2$: C, 72.71%, H, 5.42%, N, 4.71%; found: C, 72.89%, H, 5.47%, N, 4.83%.

**Compound 7c:** (2-[3-(4-Chloro-phenyl)-but-2-enyl]-3-hydroxy-2,3-dihydro-isooindol-1-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 22% Ethyl acetate: n-hexane as eluent.

Yield: 59%; Solid, mp 130-132 °C;

FT-IR (KBr, cm$^{-1}$) 3295.3, 1673.5, 1616.7, 1440.9, 1059.6, 746.6;

$^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 2.16 (s, 3H), 2.82 (br, 1OH), 4.07-4.13 (m, 1H), 4.37-4.43 (m, 1H), 5.75-5.79 (m, 2H), 7.23-7.71 (m, 8H, Ar);

$^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$ 16.07 (CH$_3$), 37.40 (CH$_2$), 81.56(CH), 122.65 (CH), 123.43 (CH), 123.46 (CH), 127.16 (2 $\times$ CH), 128.45 (2 $\times$ CH), 130.04 (CH), 131.63 (C), 132.41 (CH), 133.19 (C), 138.33 (C), 141.19 (C), 143.80 (C), 166.99 (C);

ESI-MS (m/z) 314 [M+H]$^+$. 

Anal. calcd for C$_{18}$H$_{18}$ClNO$_2$: C, 68.90%, H, 5.14%, N, 4.46%; found: C, 68.81%, H, 5.31%, N, 4.61%.

**Compound 7d:** (2-[3-(4-Bromo-phenyl)-but-2-enyl]-3-hydroxy-2,3-dihydro-isooindol-1-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 22% Ethyl acetate: n-hexane as eluent.

Yield: 63%; Solid, mp 148-150 °C;

FT-IR (KBr, cm$^{-1}$) 3352.9, 1681.8, 1594.2, 1426.9, 1216.1, 762.6;

$^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 2.12 (s, 3H), 3.07 (br, 1OH), 3.96-4.02 (m, 1H), 4.23-4.29 (m, 1H), 5.69 (d, 2H, $J = 9.2$ Hz), 7.14-7.19 (m, 2H, Ar), 7.33 (d, 2H, $J = 8.8$ Hz Ar), 7.39-7.42(m, 1H, Ar), 7.49-7.55(m, 2H, Ar), 7.59-7.61(m, 1H, Ar);

$^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$ 16.02 (CH$_3$), 37.34 (CH$_2$), 81.56 (CH), 121.33 (C), 122.69 (CH), 123.42(CH), 123.46 (CH), 127.50 (2 $\times$ CH), 129.99 (CH), 131.40 (2 $\times$ CH), 131.55 (C), 132.42 (CH), 138.39 (C), 141.65 (C), 143.87 (C), 167.08 (C);

ESI-MS (m/z) 358 [M+H]$^+$, 360 [(M+2)+H]$^+$. 

Anal. calcd for C$_{18}$H$_{16}$BrNO$_2$: C, 60.35%, H, 4.50%, N, 3.91%; found: C, 60.49%, H, 4.71%, N, 3.97%.
Compound 7e: (3-Hydroxy-2-(3-p-tolyl-but-2-enyl)-2,3-dihydro-isooindol-1-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 25% Ethyl acetate: n-hexane as eluent.

Yield: 67%; Solid, mp 106-108 °C;

FT-IR (KBr, cm\(^{-1}\)) 3350.9, 1683.5,1596.7, 1425.0, 1216.1, 761.3;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 2.10 (s, 3H), 2.24 (s, 3H), 3.09 (br, 1OH), 3.95-4.01 (m, 1H), 4.27-4.30 (m, 1H), 5.66-5.71 (m, 2H), 7.02 (d, 2H, Ar, \(J = 8.4\) Hz), 7.18-7.20 (m, 2H, Ar), 7.39-7.52 (m, 4H Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.08 (CH), 37.30 (CH\(_2\)), 81.44 (CH), 121.13 (CH), 123.38 (CH), 123.43 (CH), 125.71 (2 × CH), 129.02 (2 × CH), 129.90 (CH) 131.64 (C), 132.33 (CH), 137.20 (C), 139.78 (C), 139.87 (C), 143.95 (C), 167.08 (C);

ESI-MS (m/z) 294 [M+H]\(^+\).

Anal. calcd for C\(_{19}\)H\(_{19}\)NO\(_2\): C, 77.79%, H, 6.53%, N, 4.77%; found: C, 77.91%, H, 6.67%, N, 4.93%.

Compound 7f: (3-Hydroxy-2-[3-(4-methoxy-phenyl)-but-2-enyl]-2,3-dihydro-isooindol-1-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 22% Ethyl acetate: n-hexane as eluent.

Yield: 74%; Solid, mp 117-119 °C;

FT-IR (KBr, cm\(^{-1}\)) 3394.3, 1684.2, 1513.4, 1428.4, 1216.5, 763.8;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 2.14 (s, 3H), 3.45 (br, 1OH), 3.77 (s, 3H), 3.97-4.02 (m, 1H), 4.25-4.30 (m, 1H), 5.66-5.75 (m, 2H), 6.80 (d, 2H, Ar, \(J = 8.8\) Hz), 7.27-7.62 (m, 6H, Ar);

\(^13\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.08 (CH), 37.25(CH\(_2\)), 55.369 (CH\(_3\)), 81.42 (CH), 113.66 (2 × CH), 120.29 (CH), 123.33 (CH), 123.45 (CH), 126.91 (2 × CH), 129.83 (CH), 131.57 (C), 132.32 (CH), 135.23 (C), 138.88 (C), 144.04 (C), 159.07 (C), 167.17 (C);

ESI-MS (m/z) 310 [M+H]\(^+\).

Anal. calcd for C\(_{19}\)H\(_{19}\)NO\(_3\): C, 73.77%, H, 6.19%, N, 4.53%; found: C, 73.87%, H, 6.31%, N, 4.45%.

Compound 7g: (2-[3-(4-Cyclohexyl-phenyl)-but-2-enyl]-3-hydroxy-2,3-dihydro-isooindol-1-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 22% Ethyl acetate: n-hexane as eluent.

Yield: 71%; Solid, mp 99-101 °C;

FT-IR (KBr, cm\(^{-1}\)) 3427.8, 1659.0, 1353.2, 1056.6, 673.3;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 1.20-1.39 (m, 5H), 1.70-1.81 (m, 5H), 2.16 (s, 3H), 2.45 (br, 1OH), 3.24 (d, 1H, \(J = 11.6\) Hz), 4.00-4.06 (m, 1H), 4.30-4.35 (m, 1H), 5.72-5.75 (m, 2H), 7.10-7.13 (m, 2H, Ar), 7.27-7.29 (m, 2H, Ar), 7.43-7.65 (m, 4H, Ar);
\(^{13}\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.02 (CH\(_3\)), 26.21 (CH\(_2\)), 26.94 (2 × CH\(_2\)), 34.46 (2 × CH\(_2\)), 37.28 (CH\(_2\)), 44.26 (CH\(_2\)), 81.41 (CH), 121.16 (CH), 123.46 (CH), 123.43 (CH), 125.73 (2 × CH), 126.78 (2 × CH), 129.88 (CH), 131.60 (C), 132.34 (CH), 139.37 (C), 140.18 (C), 143.95 (C), 147.50 (C), 167.10 (C);

ESI-MS (m/z) 362 [M+H]\(^+\), 363 [M+2H]\(^+\).

Anal. calcd for C\(_{24}\)H\(_{27}\)NO\(_2\): C, 79.74%, H, 7.53%, N, 3.87%; found: C, 79.95%, H, 7.65%, N, 3.97%.

**Compound 7h:** (2-(3-Biphenyl-4-yl-but-2-enyl)-3-hydroxy-2,3-dihydro-isouindol-1-one) The crude product was purified by column chromatography over silica gel (100-200 mesh) using 22% Ethyl acetate: n-hexane as eluent.

Yield: 58%; Solid, mp 166-168 ºC;

FT-IR (KBr, cm\(^{-1}\)) 3238.2, 1688.6, 1450.5, 1064.7, 761.3;

\(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 2.23 (s, 3H), 2.89 (br, 1OH), 4.11-4.17 (m, 1H), 4.43-4.49 (m, 1H), 5.78-5.88 (m, 2H), 7.30-7.73 (m, 13H, Ar);

\(^{13}\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 16.06 (CH\(_3\)), 37.45 (CH\(_2\)), 81.52 (C), 122.12 (CH), 123.43 (CH), 123.46 (CH), 126.25 (2 × CH), 127.05 (4 × CH), 127.37 (CH), 128.85 (2 × CH), 130.07 (CH), 131.72 (C), 132.37 (CH), 139.00 (C), 140.24 (C), 140.71 (C), 141.64 (C), 143.85 (C), 166.99 (C);

ESI-MS (m/z) 356 [M+H]\(^+\).

Anal. calcd for C\(_{24}\)H\(_{21}\)NO\(_2\): C, 81.10%, H, 5.96%, N, 3.94%; found: C, 81.21%, H, 5.89%, N, 3.85%.

**General procedure for preparation of aza-peroxides 5a-h and 9a-h:** Preparation of 5a as a representative:

A solution of amide 3a (1 g, 4.33 mmol) and methylene blue (5 mg), in MeCN:THF:CHCl\(_3\) (2:1:1, 60 mL) was irradiated with 500 W tungsten-halogen lamp at -10 to 0 ºC, while a slow stream of O\(_2\) was bubbled into the reaction mixture for 6 h. The reaction mixture was concentrated under reduced pressure at rt, dissolved in acetonitrile (100 mL) and pTSA (0.1 g) was added to it. The reaction mixture was stirred at 5 ºC for 2 h. The reaction mixture was concentrated to half of its original volume under reduced pressure at rt, dissolved in acetonitrile (100 mL) and extracted with ethyl acetate (2 × 75 mL). Combined organic layer was dried over anhyd sodium sulphate and concentrated under reduced pressure at rt. The crude product was purified by column chromatography over silica gel (60-120 mesh) using 10% ethyl acetate: hexane as eluant to furnish 0.69 g (65% yield) of pure product 5a.

**Compound 5a:** (5-(1-Phenyl-vinyl)-tetrahydro-6, 7-dioxo-3a-aza-inden-3-one)

Yield: 67%; Viscous oil;

FT-IR (neat, cm\(^{-1}\)) 1705.5, 1485.5, 1445.1, 1042.4, 773.9;
\textsuperscript{1}H NMR (300 MHz, CDCl\textsubscript{3}) \(\delta\) 1.74-1.84 (m, 1H), 2.24-2.49 (m, 3H), 3.13 (dd, 1H, \(J = 13.4\) and \(10.7\) Hz), 4.23 (dd, 1H, \(J = 13.5\) and 2.7 Hz), 5.00 (dd, 1H, \(J = 10.7\) and 2.7 Hz), 5.34 and 5.51 (2 \(\times\) s, 2H), 5.65 (dd, 1H, \(J = 7.3\) and 2.8 Hz), 7.29-7.34 (m, 5H, Ar);

\textsuperscript{13}C NMR (75 MHz, CDCl\textsubscript{3}) \(\delta\) 21.52 (CH\textsubscript{2}), 28.68 (CH\textsubscript{2}), 43.43 (CH\textsubscript{2}), 80.24 (CH), 90.84 (CH), 117.10 (CH\textsubscript{2}), 126.50 (2 \(\times\) CH), 128.29 (CH), 128.62 (2 \(\times\) CH), 138.25 (C), 143.25 (C), 173.75 (C);

ESI-MS (m/z) 246 [M+H]\textsuperscript{+}; EI-MS calcd for C\textsubscript{14}H\textsubscript{15}NO\textsubscript{3}: 245.1052; found: 245.1051.

Anal. calcd for C\textsubscript{14}H\textsubscript{15}NO\textsubscript{3}: C, 68.56\%, H, 6.16\%, N, 5.71\%; found: C, 68.32\%, H, 6.53\%, N, 6.01\%.

Compounds 5b-h and 9a-h were prepared from amides 3b-h and 7a-h, respectively using the same procedure.

\textbf{Compound 5b: (5-[1-(4-Fluoro-phenyl)-vinyl]-tetrahydro-6,7-dioxa-3a-aza-inden-3-one)}

Yield: 59\%; Solid, mp 65-67 °C;

FT-IR (KBr, cm\textsuperscript{-1}) 1700.8, 1602.4, 1509.6, 1217.8, 761.3;

\textsuperscript{1}H NMR (300 MHz, CDCl\textsubscript{3}) \(\delta\) 1.79-1.90 (m, 1H), 2.31-2.34 (m, 3H), 3.18 (dd, 1H, \(J = 13.3\) and 2.7 Hz), 4.97 (dd, 1H, \(J = 10.7\) and 2.7 Hz), 5.35 and 5.49 (2 \(\times\) s, 2H), 5.69 (dd, 1H, \(J = 7.3\) and 2.9 Hz), 7.01-7.37 (m, 4H, Ar);

\textsuperscript{13}C NMR (75 MHz, CDCl\textsubscript{3}) \(\delta\) 21.67 (CH\textsubscript{2}), 28.80 (CH\textsubscript{2}), 43.41 (CH\textsubscript{2}), 80.43 (CH), 90.99 (CH), 115.67 (d, 2 \(\times\) CH, \(J_{CF} = 21.8\) Hz), 117.45 (CH\textsubscript{2}), 128.46 (d, 2 \(\times\) CH, \(J_{CF} = 7.5\) Hz ), 134.47 (d, C, \(J_{CF} = 3.0\) Hz), 142.34 (C), 162.84 (d, C, \(J_{CF} = 246.8\) Hz), 173.97 (C);

ESI-MS (m/z) 264 [M+H]\textsuperscript{+}; EI-MS calcd for C\textsubscript{14}H\textsubscript{14}FNO\textsubscript{3}: 263.0958; found: 263.0940.

\textbf{Compound 5c: (5-[1-(4-Chloro-phenyl)-vinyl]-tetrahydro-6,7-dioxa-3a-aza-inden-3-one)}

Yield: 55\%; Solid, mp 69-71 °C;

FT-IR (KBr, cm\textsuperscript{-1}) 1729.6, 1596.8, 1428.3, 1216.8, 1046.2, 761.5;

\textsuperscript{1}H NMR (300 MHz, CDCl\textsubscript{3}) \(\delta\) 1.79-1.90 (m, 1H), 2.31-2.53 (m, 3H), 3.15 (dd, 1H, \(J = 13.3\) and 10.7 Hz), 4.24 (dd, 1H, \(J = 13.3\) and 2.7 Hz), 4.96 (dd, 1H, \(J = 10.7\) and 2.7 Hz), 5.38 and 5.52 (2 \(\times\) s, 2H), 5.69 (dd, 1H, \(J = 7.3\) and 2.9 Hz), 7.31 (s, 4H, Ar);

\textsuperscript{13}C NMR (75 MHz, CDCl\textsubscript{3}) \(\delta\) 21.67 (CH\textsubscript{2}), 28.80 (CH\textsubscript{2}), 43.70 (CH\textsubscript{2}), 80.43 (CH), 90.99 (CH), 115.67 (d, 2 \(\times\) CH, \(J_{CF} = 21.8\) Hz), 119.70 (CH\textsubscript{2}), 128.46 (d, 2 \(\times\) CH, \(J_{CF} = 7.5\) Hz ), 134.43 (d, C, \(J_{CF} = 3.0\) Hz), 142.34 (C), 162.64 (d, C, \(J_{CF} = 246.8\) Hz), 173.97 (C);

ESI-MS (m/z) 280 [M+H]\textsuperscript{+}; EI-MS calcd for C\textsubscript{14}H\textsubscript{14}ClNO\textsubscript{3}: 279.0658; found: 279.0650.

\textbf{Compound 5d: (5-[1-(4-Bromo-phenyl)-vinyl]-tetrahydro-6,7-dioxa-3a-aza-inden-3-one)}

Yield: 52\%; Viscous oil;

FT-IR (neat, cm\textsuperscript{-1}) 1696.7, 1631.0, 1427.9, 1216.8, 764.0;
$^1$H NMR (300 MHz, CDCl$_3$) $\delta$ 1.76-1.87 (m, 1H), 2.27-2.51 (m, 3H), 3.13 (dd, 1H, $J = 13.4$ and 10.8 Hz), 4.22 (dd, 1H, $J = 13.4$ and 2.7 Hz), 4.94 (dd, 1H, $J = 10.8$ and 2.7 Hz), 5.36 and 5.51 (2 × s, 2H), 5.66 (dd, 1H, $J = 7.1$ and 2.6 Hz), 7.23 (d, 2H, Ar, $J = 8.1$ Hz), 7.46 (d, 2H, Ar, $J = 8.1$ Hz);

$^{13}$C NMR (75 MHz, CDCl$_3$) $\delta$ 21.72 (CH$_3$), 28.80 (CH$_2$), 43.44 (CH$_2$), 80.26 (CH), 91.12 (CH), 118.03 (CH), 122.75 (C), 128.32 (2 × CH), 131.84 (2 × CH), 137.36 (C), 142.40 (C), 173.87 (C);

ESI-MS (m/z) 324 [M+H]$^+$, 326 [M+2H]$^+$; Anal. calcd for C$_{14}$H$_{14}$BrNO$_3$: C, 51.87%, H, 4.35%, N, 4.32%; found: C, 52.01%, H, 4.56%, N, 4.07%.

**Compound 5e:** (5-(1-p-Tolyl-vinyl)-tetrahydro-6,7-dioxo-3a-aza-inden-3-one)

Yield: 69%; Solid, mp 93-95 °C;

FT-IR (KBr, cm$^{-1}$) 1704.1, 1626.7, 1424.7, 1216.4, 762.0;

$^1$H NMR (300 MHz, CDCl$_3$) $\delta$ 1.76-1.86 (m, 1H), 2.27-2.50 (m, 3H), 2.33 (s, 3H), 3.12 (dd, 1H, $J = 13.4$ and 10.8 Hz), 4.23 (dd, 1H, $J = 13.4$ and 2.5 Hz), 5.00 (dd, 1H, $J = 10.8$ and 2.5 Hz), 5.29 and 5.48 (2 × s, 2H), 5.65 (dd, 1H, $J = 7.3$ and 2.9 Hz), 7.14 (d, 2H, Ar, $J = 8.1$ Hz), 7.25 (d, 2H, Ar, $J = 8.3$ Hz);

$^{13}$C NMR (75 MHz, CDCl$_3$) $\delta$ 21.09 (CH$_3$), 21.53 (CH$_2$), 28.67 (CH$_2$), 43.51 (CH$_2$), 80.27 (CH), 90.82 (CH), 116.14 (CH$_2$), 126.35 (2 × CH), 129.28 (2 × CH), 135.33 (C), 138.13 (C), 143.12 (C), 173.63 (C);

ESI-MS (m/z) 260 [M+H]$^+$; EI-HRMS calcd for C$_{15}$H$_{17}$NO$_3$: 259.1208; found: 275.1207; Anal. calcd for C$_{15}$H$_{17}$NO$_3$: C, 69.48%, H, 6.61%, N, 5.40%; found: C, 69.42%, H, 6.44%, N, 5.76%.

**Compound 5f:** (5-[1-(4-Methoxy-phenyl)-vinyl]-tetrahydro-6,7-dioxo-3a-aza-inden-3-one)

Yield: 63%; Solid, mp 78-80 °C;

FT-IR (KBr, cm$^{-1}$) 1707.7, 1627.8, 1434.1, 1222.6, 766.1;

$^1$H NMR (300 MHz, CDCl$_3$) $\delta$ 1.81-1.91 (m, 1H), 2.30-2.55 (m, 3H), 3.16 (dd, 1H, $J = 13.1$ and 10.7 Hz), 3.83 (s, 3H), 4.26 (dd, 1H, $J = 13.1$ and 2.5 Hz), 5.01 (dd, 1H, $J = 10.7$ and 2.5 Hz), 5.28 and 5.47 (2 × s, 2H), 5.70 (dd, 1H, $J = 7.1$ and 2.7 Hz), 6.89 (d, 2H, Ar, $J = 8.6$ Hz), 7.32 (d, 2H, Ar, $J = 8.9$ Hz);

$^{13}$C NMR (75 MHz, CDCl$_3$) $\delta$ 21.82 (CH$_3$), 28.92 (CH$_2$), 29.89 (CH$_2$), 43.81 (CH$_2$), 55.52 (CH$_3$), 80.66 (CH), 91.08 (CH), 114.24 (2 × CH), 115.72 (CH$_2$), 127.95 (2 × CH), 130.92 (C), 142.95 (C), 159.95 (C), 173.91 (C);

ESI-MS (m/z) 260 [M+H]$^+$; EI-HRMS calcd for C$_{15}$H$_{17}$NO$_4$: 275.1158; found: 275.1158; Anal. calcd for C$_{15}$H$_{17}$NO$_4$: C, 69.48%, H, 6.61%, N, 5.40%; found: C, 69.42%, H, 6.44%, N, 5.76%.

**Compound 5g:** (5-[1-(4-Cyclohexyl-phenyl)-vinyl]-tetrahydro-6,7-dioxo-3a-aza-inden-3-one)

Yield: 59%; Solid, mp 81-83 °C;

FT-IR (KBr, cm$^{-1}$) 1706.7, 1621.3, 1426.3, 1222.2, 766.0;
$^1$H NMR (300 MHz, CDCl$_3$) δ 1.23-1.85 (m, 11H), 2.27-2.52 (m, 4H), 3.13 (dd, 1H, $J = 13.4$ and 10.8 Hz), 4.24 (dd, 1H, $J = 13.4$ and 2.9 Hz), 5.01 (dd, 1H, $J = 10.7$ and 2.9 Hz), 5.29 and 5.49 (2 × s, 2H), 5.68 (dd, 1H, $J = 7.3$ Hz and 2.8 Hz), 7.18 (d, 2H, Ar, $J = 8.2$ Hz), 7.28 (d, 2H, Ar, $J = 8.4$ Hz);

$^{13}$C NMR (75 MHz, CDCl$_3$) δ 21.68 (CH$_2$), 26.21 (CH$_2$), 26.94 (2 × CH$_2$), 28.80 (CH$_2$), 34.43 (2 × CH$_2$), 43.74 (CH$_2$), 44.33 (CH), 80.40 (CH), 90.94 (CH), 116.17 (CH$_2$), 126.51 (2 × CH), 127.16 (2 × CH), 135.77 (C), 143.32 (C), 148.42 (C), 173.74 (C);

ESI-MS (m/z) 328 [M+H]$^+$; EI-HRMS calcd for C$_{20}$H$_{25}$NO$_3$: 327.1834; found: 327.1837; Anal. calcd for C$_{20}$H$_{25}$NO$_3$: C, 73.37%, H, 7.70%, N, 4.28%; found: C, 73.47%, H, 7.98%, N, 4.32%.

**Compound 5h:** (5-(1-Biphenyl-4-yl-vinyl)-tetrahydro-6,7-dioxo-3a-aza-inden-3-one)

Yield: 71%; Solid, mp 136-138 ºC;

FT-IR (KBr, cm$^{-1}$) 1705.5, 1485.5, 1445.1, 1042.4, 773.9;

$^1$H NMR (300 MHz, CDCl$_3$) δ 1.75-1.86 (m, 1H), 2.23-2.50 (m, 3H), 3.15 (dd, 1H, $J = 13.4$ and 10.8 Hz), 4.27 (dd, 1H, $J = 13.4$ and 2.6 Hz), 5.05 (dd, 1H, $J = 10.8$ and 2.6 Hz), 5.35 and 5.56 (2 × s, 2H), 5.65 (dd, 1H, $J = 7.3$ and 2.8 Hz), 7.33-7.59 (m, 9H, Ar);

$^{13}$C NMR (75 MHz, CDCl$_3$) δ 21.66 (CH$_2$), 28.77 (CH$_2$), 43.60 (CH$_2$), 80.31 (CH), 90.95 (CH), 117.03 (CH$_2$), 127.03 (2 × CH), 127.09 (2 × CH), 127.38 (2 × CH), 127.60 (CH), 128.91 (2 × CH), 137.20 (C), 140.46 (C), 141.21 (C), 142.95 (C), 173.76 (C);

ESI-MS (m/z) 322 [M+H]$^+$; EI-HRMS calcd for C$_{20}$H$_{20}$NO$_3$: 322.14432; found: 322.14161. Anal. calcd for C$_{20}$H$_{19}$NO$_3$.0.1H$_2$O: C, 75.12%; H, 5.71%; N, 4.37%; found: C, 75.46%, H, 5.98%, N, 4.39%.

**Compound 9a:** (2-(1-Phenyl-vinyl)-1,2-dihydro-4aH-3,4-dioxo-9a-aza-fluoren-9-one)

Yield: 60%; Solid, mp 85-87 ºC;

FT-IR (KBr, cm$^{-1}$) 1716.4, 1656.4, 1419.6, 1019.2, 748.5;

$^1$H NMR (300 MHz, CDCl$_3$) δ 3.41 (dd, 1H, $J = 13.6$ and 10.9 Hz), 4.53 (dd, 1H, $J = 13.6$ and 2.9 Hz), 5.00 (dd, 1H, $J = 10.9$ Hz and 2.9 Hz), 5.43 and 5.58 (2 × s, 2H), 6.31 (s, 1H), 7.32-7.89 (m, 9H, Ar);

$^{13}$C NMR (75 MHz, CDCl$_3$) δ 44.33 (CH$_2$), 79.92 (CH), 88.18 (CH), 117.57 (CH$_2$), 124.34 (CH), 124.57 (CH), 126.77 (2 × CH), 128.54 (CH), 128.84 (2 × CH), 131.33 (CH), 132.53 (CH), 133.43 (C), 137.32 (C), 138.45 (C), 143.29 (C), 166.81 (C);

ESI-MS (m/z) 294 [M+H]$^+$; EI-HRMS calcd for C$_{18}$H$_{18}$NO$_3$: 294.11302; found: 294.11211.

Anal. calcd for C$_{18}$H$_{17}$NO$_3$.0.1H$_2$O: C, 74.16%; H, 5.18%; N, 4.81%; found: C, 74.60%, H, 5.34%, N, 4.77%.

**Compound 9b:** (2-[1-(4-Fluoro-phenyl)-vinyl]-1,2-dihydro-4aH-3,4-dioxo-9a-aza-fluoren-9-one)

Yield: 59%; Solid, mp 80-82 ºC;
FT-IR (KBr, cm$^{-1}$) 1707.3, 1510.8, 1217.2, 765.1; $^1$H NMR (300 MHz, CDCl$_3$) $\delta$ 3.41 (dd, 1H, $J = 13.5$ and 10.9 Hz), 4.52 (dd, 1H, $J = 13.5$ and 2.6 Hz), 4.93 (dd, 1H, $J = 10.9$ and 2.6 Hz), 5.43 and 5.54 (2 $\times$ s, 2H), 6.30 (s, 1H), 7.01 - 7.89 (m, 8H, Ar);

$^{13}$C NMR (75 MHz, CDCl$_3$) $\delta$ 43.11 (CH$_2$), 79.95 (CH), 88.19 (CH), 115.74 (d, 2 $\times$ CH, $J_{C-F} = 21.0$ Hz), 117.73 (CH$_2$), 124.35 (CH), 124.55 (CH), 128.69 (d, 2 $\times$ CH, $J_{C-F} = 8.3$ Hz), 131.32 (CH), 132.52 (CH), 133.42 (C), 134.58 (d, C, $J_{C-F} = 3.0$ Hz), 137.33 (C), 142.32 (C), 161.45 (d, C, $J_{C-F} = 246.8$ Hz), 166.78 (C);

ESI-MS (m/z) 312 [M+H]$^+$; ESI-MS calcd for C$_{18}$H$_{14}$FNO$_3$: 311.0958; found: 311.0947.

**Compound 9c:** (2-[1-(4-Chloro-phenyl)-vinyl]-1,2-dihydro-4aH-3,4-dioxo-9a-aza-fluoren-9-one)
Yield: 54%; Solid, mp 121-123 ºC;
FT-IR (KBr, cm$^{-1}$) 1712.5, 1596.4, 1421.7, 1216.0, 761.1;

$^1$H NMR (300 MHz, CDCl$_3$) $\delta$ 3.41 (dd, 1H, $J = 13.6$ and 10.9 Hz), 4.51 (dd, 1H, $J = 13.6$ and 2.9 Hz), 4.94 (dd, 1H, $J = 10.9$ and 2.9 Hz), 5.45 and 5.57 (2 $\times$ s, 2H), 6.30 (s, 1H), 7.32 - 7.89 (m, 8H, Ar) ;

$^{13}$C NMR (75 MHz, CDCl$_3$) $\delta$ 43.00 (CH$_2$), 79.70 (CH), 88.14 (CH), 118.18 (CH$_2$), 124.30 (CH), 124.51 (CH), 128.10 (2 $\times$ CH), 128.95 (2 $\times$ CH), 131.29 (CH), 132.49 (CH), 133.35 (C), 134.46 (C), 136.87 (C), 137.26 (C), 142.15 (C), 166.72 (C);

ESI-MS (m/z) 328 [M+H]$^+$; Anal. calcd for C$_{18}$H$_{14}$ClNO$_3$: C, 65.96%, H, 4.31%, N, 4.27%; found: C, 66.32%, H, 4.66%, N, 4.17%.

**Compound 9d:** (2-[1-(4-Bromo-phenyl)-vinyl]-1,2-dihydro-4aH-3,4-dioxo-9a-aza-fluoren-9-one)
Yield: 61%; Solid, mp 132-134 ºC;
FT-IR (KBr, cm$^{-1}$) 1722.9, 1421.2, 1216.2, 1045.8, 761.3;

$^1$H NMR (300 MHz, CDCl$_3$) $\delta$ 3.43 (dd, 1H, $J = 13.5$ and 10.8 Hz), 4.51 (dd, 1H, $J = 13.5$ and 2.9 Hz), 4.94 (dd, 1H, $J = 10.9$ and 2.9 Hz), 5.45 and 5.57 (2 $\times$ s, 2H), 6.30 (s, 1H), 7.28 (d, 2H, Ar, $J = 8.4$ Hz), 7.49 (d, 2H, Ar, $J = 8.4$ Hz), 7.62-7.91 (m, 8H, Ar);

$^{13}$C NMR (75 MHz, CDCl$_3$) $\delta$ 43.05 (CH$_2$), 79.70 (CH), 88.14 (CH), 118.18 (CH$_2$), 122.71 (C), 124.37 (CH), 124.57 (CH), 128.45 (2 $\times$ CH), 131.36 (CH), 131.96 (2 $\times$ CH), 132.49 (CH), 133.35 (C), 134.46 (C), 136.87 (C), 137.26 (C), 142.15 (C), 166.72 (C);

ESI-MS (m/z) 372 [M+H]$^+$, 374.0 [(M+2)+H]$^+$; EI-MS calcd for C$_{18}$H$_{14}$BrNO$_3$: 371.0157; found: 371.0193;
Anal. calcd for C$_{18}$H$_{14}$BrNO$_3$.0.1H$_2$O: C, 58.36%, H, 3.81%, N, 3.78%; found: C, 58.78%, H, 3.68%, N, 3.67%.

**Compound 9e:** (2-(1-p-Tolyl-vinyl)-1,2-dihydro-4aH-3,4-dioxo-9a-aza-fluoren-9-one)
Yield: 70%; Viscous oil;
FT-IR (neat, cm$^{-1}$) 1720.2, 1413.4, 1213.5, 1040.8, 768.1;
\(^1\)H NMR (300 MHz, CDCl\(_3\)) \(\delta\) 2.35 (s, 3H), 3.41 (dd, 1H, \(J = 13.6\) and 10.7 Hz), 4.52 (dd, 1H, \(J = 13.6\) and 2.9 Hz), 4.99 (dd, 1H, \(J = 10.8\) and 2.9 Hz), 5.38 and 5.55 (2 × s, 2H), 6.31 (s, 1H), 7.16 (d, 2H, Ar, \(J = 8.0\) Hz), 7.28 (d, 2H, Ar, \(J = 8.0\) Hz), 7.60-7.90 (m, Ar, 4H);

\(^1\)C NMR (75 MHz, CDCl\(_3\)) \(\delta\) 21.34 (CH\(_3\)), 43.43 (CH\(_2\)), 80.00 (CH), 88.23 (CH), 116.72 (CH\(_2\)), 124.38 (CH), 124.59 (CH), 126.66 (2 × CH), 131.33 (CH), 133.51 (C), 134.95 (C), 137.41 (C), 138.48 (C), 143.19 (C), 166.83 (C);

ESI-MS (m/z) 308 [M+H]\(^+\); Anal. calcd for C\(_{19}\)H\(_{17}\)NO\(_3\): C, 74.25%, H, 5.58%, N, 4.56%; found: C, 74.56%, H, 6.11%, N, 4.45%.

**Compound 9f:** (2-[1-(4-Methoxy-phenyl)-vinyl]-1,2-dihydro-4aH-3,4-dioxa-9a-aza-fluoren-9-one)

Yield: 63%; Solid, mp 115-117 °C;

FT-IR (KBr, cm\(^{-1}\)) 1711.2, 1413.4, 1213.5, 1040.8, 768.1;

\(^1\)H NMR (300 MHz, CDCl\(_3\)) \(\delta\) 3.41 (dd, 1H, \(J = 13.4\) and 10.9 Hz), 3.81 (s, 3H), 4.52 (dd, 1H, \(J = 13.4\) and 2.6 Hz), 4.97 (dd, 1H, \(J = 10.9\) and 2.6 Hz), 5.34 and 5.51 (2 × s, 2H), 6.31 (s, 1H), 6.88 (d, 2H, Ar, \(J = 8.9\) Hz), 7.33 (d, 2H, Ar, \(J = 8.9\) Hz), 7.59-7.90 (m, 4H, Ar);

\(^1\)C NMR (75 MHz, CDCl\(_3\)) \(\delta\) 43.40 (CH\(_2\)), 55.52 (CH\(_3\)), 80.04 (CH), 88.23 (CH), 114.23 (2 × CH), 115.99 (CH\(_2\)), 124.37 (CH), 124.58 (CH), 127.98 (2 × CH), 130.89 (C), 131.32 (CH), 132.52 (CH), 133.51 (C), 137.41 (C), 142.70 (C), 156.94 (C), 166.83 (C);

ESI-MS (m/z) 324 [M+H]\(^+\); Anal. calcd for C\(_{19}\)H\(_{17}\)NO\(_4\): C, 70.58%, H, 5.30%, N, 4.33%; found: C, 70.38%, H, 5.01%, N, 4.32%.

**Compound 9g:** (2-[1-(4-Cyclohexyl-phenyl)-vinyl]-1,2-dihydro-4aH-3,4-dioxa-9a-aza-fluoren-9-one)

Yield: 57%; Solid; mp 135-137 °C;

FT-IR (KBr, cm\(^{-1}\)) 1715.1, 1655.4, 1423.0, 1022.4, 748.1;

\(^1\)H NMR (300 MHz, CDCl\(_3\)) \(\delta\) 1.26-1.85 (m, 10H), 2.45-2.55 (m, 1H), 3.41 (dd, 1H, \(J = 13.5\) and 11.0 Hz), 4.53 (dd, 1H, \(J = 13.5\) and 3.0 Hz), 5.00 (dd, 1H, \(J = 10.9\) and 2.7 Hz), 5.38 and 5.56 (2 × s, 2H), 6.31 (s, 1H), 6.88 (d, 2H, Ar, \(J = 8.9\) Hz), 7.19 (d, 2H, Ar, \(J = 8.9\) Hz), 7.31 (d, 2H, Ar, \(J = 8.2\) Hz), 7.60-7.91 (m, 4H, Ar);

\(^1\)C NMR (75 MHz, CDCl\(_3\)) \(\delta\) 26.07 (CH\(_2\)), 26.80 (2 × CH\(_2\)), 34.29 (2 × CH\(_2\)), 43.25 (CH\(_2\)), 44.21 (CH), 79.72 (CH), 87.99 (CH), 116.37 (CH\(_2\)), 124.13 (CH), 124.34 (CH), 126.44 (2 × CH), 127.07 (2 × CH), 131.07 (CH), 132.27 (CH), 133.28 (C), 135.64 (C), 137.19 (C), 142.99 (C), 148.39 (C), 166.57 (C);

ESI-MS (m/z) 376 [M+H]\(^+\); EI-HRMS calcd for C\(_{24}\)H\(_{26}\)NO\(_3\): 376.19127; found 376.19407; Anal. calcd for C\(_{24}\)H\(_{25}\)NO\(_3\): C, 76.77%, H, 6.71%, N, 3.73%; found: C, 76.98%, H, 6.32%, N, 3.57%.

**Compound 9h:** (2-(1-Biphenyl-4-y1-vinyl)-1,2-dihydro-4aH-3,4-dioxa-9a-aza-fluoren-9-one)
Yield: 61%; Solid, mp 148-150 °C;

FT-IR (KBr, cm$^{-1}$) 1712.3, 1664.8, 1596.0, 1480.1, 1217.0, 761.1;

$^{1}$H NMR (300 MHz, CDCl$_3$) δ 3.45 (dd, 1H, $J = 13.5$ and 10.9 Hz), 4.57 (dd, 1H, $J = 13.5$ and 2.9 Hz), 5.04 (dd, 1H, $J = 10.9$ and 2.5 Hz), 5.46 and 5.64 (2 × s, 2H), 6.33 (s, 1H), 7.25-7.91 (m, 13H, Ar);

$^{13}$C NMR (75 MHz, CDCl$_3$) δ 43.36 (CH$_2$), 79.89 (CH), 88.25 (CH), 117.42 (CH$_2$), 124.38 (CH), 124.58 (CH), 127.20 (2 × CH), 127.23 (2 × CH), 127.54 (2 × CH), 127.71 (CH), 129.01 (2 × CH), 131.33 (CH), 132.52 (CH), 133.50 (C), 137.33 (C), 137.41 (C), 140.60 (C), 141.43 (C), 142.91 (C), 166.82 (C);

ESI-MS (m/z) 370 [M+H]$^+$; EI-HRMS calcd for C$_{24}$H$_{20}$NO$_3$: 370.14432; found: 370.14085.

Anal. calcd for C$_{24}$H$_{19}$NO$_3$: C, 78.03%, H, 5.18%, N, 3.79%; found: C, 78.43%, H, 5.31%, N, 3.57%.
$^1$H NMR and $^{13}$C NMR Spectra of Compounds 2a-h and 6a-h, 3a-h and 7a-h and 5a-h and 9a-h.

$^1$H NMR spectrum of compound 2a

$^{13}$C NMR spectrum of compound 2a
$^1$H NMR spectrum of compound 2b

$^{13}$C NMR spectrum of compound 2b
$^1$H NMR spectrum of compound 2c

$^{13}$C NMR spectrum of compound 2c
$^1$H NMR spectrum of compound 2d

$^{13}$C NMR spectrum of compound 2d
\(^{1}\text{H} \text{NMR spectrum of compound} \ 2e\)

\(^{13}\text{C} \text{NMR spectrum of compound} \ 2e\)
$^1$H NMR spectrum of compound 2f

$^{13}$C NMR spectrum of compound 2f
$^1$H NMR spectrum of compound 2g

$^{13}$C NMR spectrum of compound 2g
$^1$H NMR spectrum of compound 2h

$^{13}$C NMR spectrum of compound 2h
$^1$H NMR spectrum of compound 3a

$^{13}$C NMR spectrum of compound 3a
$^1$H NMR spectrum of compound 3b

$^{13}$C NMR spectrum of compound 3b
$^1$H NMR spectrum of compound 3c

$^{13}$C NMR spectrum of compound 3c
$^1$H NMR spectrum of compound 3d

$^{13}$C NMR spectrum of compound 3d
$^1$H NMR spectrum of compound 3e

$^{13}$C NMR spectrum of compound 3e
$^1$H NMR spectrum of compound 3f

$^{13}$C NMR spectrum of compound 3f
$^1$H NMR spectrum of compound 3g

$^{13}$C NMR spectrum of compound 3g
$^1$H NMR spectrum of compound 3h

$^{13}$C NMR spectrum of compound 3h
$^1$H NMR spectrum of compound 5a

$^{13}$C NMR spectrum of compound 5a
$^1$H NMR spectrum of compound 5b

$^{13}$C NMR spectrum of compound 5b
$^1$H NMR spectrum of compound 5e

$^{13}$C NMR spectrum of compound 5e
$^1$H NMR spectrum of compound 5d

$^{13}$C NMR spectrum of compound 5d
**H NMR spectrum of compound 5e**

**C NMR spectrum of compound 5e**
$^1$H NMR spectrum of compound 5f

$^{13}$C NMR spectrum of compound 5f
$^1$H NMR spectrum of compound 5g

$^{13}$C NMR spectrum of compound 5g
$^1$H NMR spectrum of compound 5h

$^{13}$C NMR spectrum of compound 5h
$^1$H NMR spectrum of compound 6a

$^{13}$C NMR spectrum of compound 6a
$^1$H NMR spectrum of compound 6b

$^{13}$C NMR spectrum of compound 6b
$^{1}H$ NMR spectrum of compound 6c

$^{13}C$ NMR spectrum of compound 6c
$^1$H NMR spectrum of compound 6d

$^{13}$C NMR spectrum of compound 6d
$^{1}H$ NMR spectrum of compound 6e

$^{13}$C NMR spectrum of compound 6e
$^1$H NMR spectrum of compound 6f

$^{13}$C NMR spectrum of compound 6f
$^1$H NMR spectrum of compound 6g

$^{13}$C NMR spectrum of compound 6g
$^1$H NMR spectrum of compound 6h

$^{13}$C NMR spectrum of compound 6h
$^1$H NMR spectrum of compound 7a

$^{13}$C NMR spectrum of compound 7a
$^1$H NMR spectrum of compound 7b

$^{13}$C NMR spectrum of compound 7b
$^1$H NMR spectrum of compound 7c

$^{13}$C NMR spectrum of compound 7c
$^1$H NMR spectrum of compound 7d

$^{13}$C NMR spectrum of compound 7d
$^1$H NMR spectrum of compound 7e

$^{13}$C NMR spectrum of compound 7e
$^1$H NMR spectrum of compound 7f

$^{13}$C NMR spectrum of compound 7f
$^1$H NMR spectrum of compound 7g

$^{13}$C NMR spectrum of compound 7g
$^1$H NMR spectrum of compound 7h

$^{13}$C NMR spectrum of compound 7h
$^1$H NMR spectrum of compound 9a

$^{13}$C NMR spectrum of compound 9a
$^1$H NMR spectrum of compound 9b

$^{13}$C NMR spectrum of compound 9b
$^1$H NMR spectrum of compound 9c

$^{13}$C NMR spectrum of compound 9c
\(^1\)H NMR spectrum of compound 9d

\(^{13}\)C NMR spectrum of compound 9d
$^1$H NMR spectrum of compound 9e

$^{13}$C NMR spectrum of compound 9e
$^1$H NMR spectrum of compound 9f

$^{13}$C NMR spectrum of compound 9f
$^1$H NMR spectrum of compound 9g

$^{13}$C NMR spectrum of compound 9g
$^1$H NMR spectrum of compound 9h

$^{13}$C NMR spectrum of compound 9h
NOESY spectrum of compound 9g