Supporting information

Table of Contents	Page no
1. General Information	S1
2. Experimental Procedures	S1
3. Spectral Data of Synthesized Compounds	S2-S8
4. X-ray Data and Crystal Structures	S7-S8
5. Spectral Copies of Synthesized Compounds	S8-S34

EXPERIMENTAL SECTION

General. Phenylglyoxal and derivatives were synthesized using a reported procedure.¹ pyrrolidin-2-one, 2-naphthol, phenol and 4-hydroxycoumarin, were purchased from Sigma-Aldrich. Catalysts were obtained from TCI. Solvents and silica gel (60–120 mesh) and other common reagents were procured from local suppliers. Thin-layer chromatography (TLC) was carried out on Merck silica gel plates (60 F254). ¹H and ¹³C{1H} NMR were recorded on a Bruker Ultrashield 500 and 400 MHz or Bruker Ascend 500 MHz instrument. Melting points were measured with a MEPA LABINDIA melting point apparatus. Single-crystal X-ray diffraction data were collected in Bruker D8-Quest diffractometers.

- I. General experimental procedure for the synthesis of 4, 6 and 8: A mixture of glyoxal (1), lactam (2) and Ca(OTf)₂/Bu₄NPF₆ (10/10 mol%) was heated directly at 100 °C for one hour. After complete conversion of compound 1 (monitored by TLC), phenolic nucleophile (naphthols 3 for 4, phenol 5 for 6 and fluorenol 7 for 8) was added to the above reaction mixture and continued heating as per the time mentioned. After completion of the reaction, monitored by TLC) the crude product was purified by column chromatography (25-30 % EtOAc in petroleum ether) to obtain the pure product 4 or 6 or 8.
- II. Experimental procedure for the synthesis of (4a): A mixture of 2-oxo-2-phenylacetaldehyde 1a (0.65 mmol, 100 mg), pyrrolidin-2-one 2 (0.78 mmol, 67 mg), and Ca(OTf)₂/Bu₄NPF₆ (22 mg/ 25 mg, 10/10 mol%) and stirred under neat conditions at 100 °C for one hour. After complete conversion of compound 1 (monitored by TLC), 2-naphthol 3a (0.65 mmol, 94 mg) was added to the above reaction mixture and continued stirring for 4 h. After completion of the reaction, the crude product was purified by column chromatography (25-30 % EtOAc in petroleum ether) to obtain the pure product 4a as a white solid in 85% yield.
- III. Experimental procedure for the synthesis of furocoumarin (10a): A mixture of 2-oxo-2-phenylacetaldehyde 1a (0.65 mmol, 100 mg), pyrrolidin-2-one 2a (0.78 mmol, 67 mg), and Ca(OTf)₂/Bu₄NPF₆ (22 mg/25 mg, 10/10 mol%) and stirred under neat conditions at 100 °C for 1 hour. After complete conversion of compound 1(monitored by TLC), DCE (2 mL) and 4-hydroxycoumarin 9 (0.65 mmol, 106 mg) were added to the above reaction

mixture and refluxed the reaction for 12 h. After completion of the reaction, the reaction mixture was directly absorbed on silica gel and was purified by column chromatography (25-30 % EtOAc in petroleum ether) to obtain the pure product **10a** with 50% yield. Compounds **10a**,**10b** are reported.²

IV. Spectral data of new compounds:

1-(2-Phenylnaphtho[2,1-*b*]*furan-1-yl*)*pyrrolidin-2-one* (**4a**). White solid (182 mg, 85%); mp: 207–209 °C; ¹H NMR (500 MHz, CDCl₃): δ 8.03 (d, J = 8 Hz, 1H), 7.94 (d, J = 8 Hz, 1H), 7.84-7.83 (m, 2H), 7.76 (d, J = 9 Hz, 1H), 7.67 (d, J = 9 Hz, 1H), 7.59-7.56 (m, 1H), 7.51-7.46 (m, 3H), 7.41-7.39 (m, 1H), 3.95-3.91 (m, 1H), 3.76-3.71 (m, 1H), 2.84-2.77 (m, 2H), 2.44-2.36 (m, 2H); ¹³C{1H}(125 MHz, CDCl₃) δ 176.0, 151.5, 149.9, 130.9, 129.5, 129.2, 129.0 (2), 127.4, 127.0, 126.5, 125.8, 124.9, 122.0, 120.1, 116.9, 112.6, 49.5, 31.2, 19.4 ppm; (LCMS): *m/z* [M + H]⁺: 328; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₂H₁₈NO₂: 328.1332; found: 328.1323; IR (film): *v_{max}* 1690, 1219, 812, 771 cm⁻¹.

1-(2-(2-Bromophenyl)naphtho[*2*, *1-b*]*furan-1-yl*)*pyrrolidin-2-one* (**4b**). Following general experimental procedure-I, the product was obtained as white solid (129 mg, 74%); mp: 179–180 °C; ¹H NMR (500 MHz, CDCl₃): δ 8.04 (d, *J* = 8 Hz, 1H), 7.94 (d, *J* = 8 Hz, 1H), 7.77 (d, *J* = 9 Hz, 1H), 7.73-7.71 (m, 1H), 7.68-7.63 (m, 2H), 7.59-7.56 (m, 1H), 7.50-7.47 (m, 1H), 7.42-7.39 (m, 1H), 7.35-7.37 (m, 1H), 3.87-3.83 (m, 1H), 3.49-3.44 (m, 1H), 2.73-2.66 (m, 1H), 2.57-2.51 (m, 1H), 2.34-2.25 (m, 1H), 2.14-2.08 (m, 1H); ¹³C{1H}(125 MHz, CDCl₃) δ 176.2, 152.0, 149.9, 133.4, 132.6, 131.3, 130.9, 130.5, 129.0, 127.5 (2), 127.0, 126.7, 124.9, 123.7, 122.3, 119.3, 119.1, 112.8, 50.1, 31.1, 19.3 ppm; (LCMS): *m/z* [M + H]⁺: 406; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₂H₁₇BrNO₂: 406.0437; found: 406.0432; IR (film): *v_{max}* 1663, 1253, 811, 772, 723 cm⁻¹.

1-(2-(4-Bromophenyl)naphtho[2,1-b]furan-1-yl)pyrrolidin-2-one (**4c**). Following general experimental procedure-I, the product was obtained as white solid (134 mg, 77%); mp: 231–232 °C; ¹H NMR (500 MHz, CDCl₃): δ 8.01 (d, *J* = 8 Hz, 1H), 7.93 (d, *J* = 8 Hz, 1H), 7.77 (d, *J* = 9 Hz, 1H), 7.69 (d, *J* = 9 Hz, 2H), 7.65 (d, *J* = 9 Hz, 1H), 7.62-7.57 (m, 3H), 7.52-7.49 (m, 1H), 3.97-3.92 (m, 1H), 3.72-3.67 (m, 1H), 2.87-2.72 (m, 2H), 2.51-2.33 (m, 2H); ¹³C{1H} (125 MHz, CDCl₃) δ 176.0, 151.6, 148.9, 132.3, 131.0, 129.3, 128.5, 127.4, 127.3, 127.1, 126.9, 125.0, 123.2, 121.9, 120.0, 117.5, 112.6, 49.5, 31.2, 19.5 ppm; (LCMS): *m/z* [M + H]⁺: 406; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₂H₁₇BrNO₂: 406.0437; found: 406.0435; IR (film): *v_{max}* 1685, 1260, 811, 771, 693 cm⁻¹.

1-(2-(4-Chlorophenyl)naphtho[2,1-*b*]*furan-1-yl)pyrrolidin-2-one* (**4d**). Following general experimental procedure-I, the product was obtained as white solid (149 mg, 77%); mp: 162–163 °C; ¹H NMR (500 MHz, CDCl₃): δ 8.01 (d, J = 8 Hz, 1H), 7.95 (d, J = 8.5 Hz, 1H), 7.78-7.76 (m, 3H), 7.66 (d, J = 8.5 Hz, 1H), 7.60-7.57 (m, 1H), 7.52-7.49 (m, 1H), 7.47-7.44 (m, 2H), 3.97-3.93 (m, 1H), 3.73-3.68 (m, 1H), 2.88-2.73 (m, 2H), 2.53-2.32 (m, 2H); ¹³C{1H}(125 MHz, CDCl₃) δ 176.0, 151.5, 148.8, 134.8, 130.9, 130.2, 129.3, 129.2, 129.1, 128.0, 127.3, 127.1, 127.0, 126.8, 125.0, 121.9, 120.0, 117.3, 112.5, 49.4, 31.2, 19.4 ppm; (LCMS): m/z [M + H]⁺: 362; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₂H₁₇ClNO₂: 362.0948; found: 362.0947; IR (film): v_{max} 1696, 1586, 1457, 1274, 815, 772, 619 cm⁻¹.

1-(2-(4-Fluorophenyl)naphtho[2,1-*b*]*furan-1-yl*)*pyrrolidin-2-one* (**4e**). Following general experimental procedure-I, the product was obtained as white solid (166 mg, 82%); mp: 202–203°C; ¹H NMR (500 MHz, CDCl₃): δ 8.01 (d, *J* = 8.5 Hz, 1H), 7.94 (d, *J* = 8 Hz, 1H), 7.82-7.79 (m, 2H), 7.75 (d, *J* = 9 Hz, 1H), 7.65 (d, *J* = 9 Hz, 1H), 7.59-7.56 (m, 2 H), 7.49 (t, *J* = 8 Hz, 1H), 7.17 (t, *J* = 10.5 Hz, 1H), 3.95-3.91 (m, 1H), 3.71-3.66 (m, 1H), 2.87-2.72 (m, 2H), 2.50-2.30 (m, 2H); ¹³C{1H}(100 MHz, CDCl₃) δ 176.1, 164.3, 161.8, 151.4, 149.2, 130.9, 129.2, 127.9, 127.8, 127.4, 127.0, 126.6, 124.9, 121.9, 120.0, 116.3, 116.1, 112.6, 49.5, 31.2, 19.4 ppm; (LCMS): *m/z* [M + H]⁺: 346; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₂H₁₇FNO₂: 346.1243; found: 346.1242

1-(2-(4-Methoxyphenyl)naphtho[2,1-*b*]*furan-1-yl*)*pyrrolidin-2-one* (**4f**). Following general experimental procedure-I, the product was obtained as white solid (156 mg, 80%); mp: 210–211 °C; ¹H NMR (500 MHz, CDCl₃): δ 8.01 (d, J = 8.4 Hz, 1H), 7.93 (d, J = 8 Hz, 1H), 7.77-7.71 (m, 3H), 7.64 (d, J = 8.8 Hz, 1H), 7.56 (t, J = 7.2 Hz, 1H), 7.49-7.46 (m, 1H), 7.01-6.99 (m, 2H), 3.94-3.90 (m, 1H), 3.86 (s, 3H), 3.73-3.68 (m, 1H), 2.88-2.71 (m, 2H), 2.48-2.32 (m, 2H); ¹³C{1H}(100 MHz, CDCl₃) δ 176.2, 160.2, 151.1, 150.1, 130.9, 129.1, 127.4, 127.3, 126.8, 126.0, 124.7, 122.2, 122.0, 120.2, 115.5, 114.5, 112.5, 55.4, 49.5, 31.3, 19.4 ppm; (LCMS): m/z [M + H]⁺: 358; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₃H₂₀NO₃: 358.1443; found: 358.1442; IR (film): v_{max} 1687, 1251, 828, 811, 729 cm⁻¹.

1-(7-Bromo-2-phenylnaphtho[2,1-*b*]*furan-1-yl*)*pyrrolidin-2-one* (**4g**). Following general experimental procedure-I, the product was obtained as white solid (216 mg, 81%); mp: 186–187 °C; ¹H NMR (500 MHz, CDCl₃): δ 8.10 (d, J = 2 Hz, 1H), 7.88 (d, J = 8.5 Hz, 1H), 7.82-7.81 (m, 2H), 7.70-7.64 (m, 3H), 7.50-7.47 (m, 2H), 7.42-7.40 (m, 1H), 3.90-3.85 (m, 1H), 3.75-3.70 (m, 1H), 2.81-2.76 (m, 2H), 2.44-2.36 (m, 2H); ¹³C{1H}(125 MHz, CDCl₃) δ 176.1, 151.5, 150.4, 132.2, 131.2, 130.1, 129.3, 129.2, 129.1, 125.9, 125.9, 125.5, 123.6, 120.3, 118.6, 116.7, 113.8, 49.4, 31.2, 19.5 ppm; (LCMS): *m/z* [M + Na]⁺: 427; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₂H₁₇BrNO₂: 406.0443; found: 406.0441; IR (film): *v_{max}* 1681, 1576, 1254, 892, 799, 636 cm⁻¹.

1-(7-Bromo-2-(p-tolyl)naphtho[2,1-*b*]*furan-1-yl*)*pyrrolidin-2-one* (**4h**). Following general experimental procedure-I, the product was obtained as white solid (184 mg, 73%); mp: 195–196 °C;¹H NMR (500 MHz, CDCl₃): δ 8.07 (d, J = 2 Hz, 1H), 7.87 (d, J = 9 Hz, 1H), 7.70-7.66 (m, 2H), 7.64-7.62 (m, 3H), 7.29-7.25 (m, 2H), 3.85-3.84 (m, 1H), 3.72-3.70 (m, 1H), 2.80-2.75 (m, 2H), 2.44 (s, 3H), 2.43-2.32 (m, 2H); ¹³C{1H}(100 MHz, CDCl₃) δ 176.0, 151.2, 150.7, 139.4, 132.2, 131.1, 130.0, 129.8, 126.5, 125.8, 125.1, 123.6, 120.3, 118.4, 116.1, 113.7, 49.3, 31.2, 21.5, 19.4 ppm; (LCMS): m/z [M + H]⁺: 320; HRMS (ESI-TOF): m/z [M + Na]⁺ calculated for C₂₃H₁₈BrNO₂Na: 442.0419; found: 442.0422.

1-(7-Bromo-2-(4-chlorophenyl)naphtho[2,1-*b*]*furan-1-yl*)*pyrrolidin-2-one* (**4i**). Following general experimental procedure-I, the product was obtained as white solid (188 mg, 80%); mp: 208–209 °C;¹H NMR (500 MHz, CDCl₃): δ 8.31 (s, 1H), 7.94 (d, J = 9 Hz, 1H), 7.90-7.85 (m, 2H), 7.81-7.75 (m, 3H), 7.57 (d, J = 8 Hz, 2H), 3.90-3.86 (m, 1H), 3.74-3.69 (m, 1H), 2.78-2.64 (m, 2H), 2.46-2.37 (m, 2H); ¹³C{1H}(125 MHz, CDCl₃) δ 176.1, 151.4,

150.4, 132.2, 131.2, 130.1, 129.2 (2), 129.1, 129.0, 125.8 (2), 125.4, 123.6, 120.3, 118.5, 116.7, 113.7, 49.4, 31.2, 19.4 ppm; (LCMS): m/z [M + H]⁺: 439; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₂H₁₆BrClNO₂: 440.0053; found: 440.0057.

1-(2-Isopropylnaphtho[2,1-*b*]*furan-1-yl*)*pyrrolidin-2-one* (**4j**). Following general experimental procedure-I, the product was obtained as white solid (211 mg, 85%); mp: 220–221 °C;¹H NMR (500 MHz, CDCl₃): δ 7.95-7.90 (m, 2H), 7.66 (d, *J* = 9 Hz, 1H), 7.59-7.57 (m, 1H), 7.52 (t, *J* = 7 Hz, 1H), 7.44 (t, *J* = 8 Hz, 1H), 4.04-3.99 (m, 1H), 3.70-3.65 (m, 1H), 3.14-3.08 (m, 1H), 2.79-2.65 (m, 2H), 2.47-2.34 (m, 2H), 1.40 (d, *J* = 6.5 Hz, 3H), 1.36 (d, *J* = 7 Hz, 3H); ¹³C{1H}(100 MHz, CDCl₃) δ 175.7, 158.8, 151.2, 130.7, 129.0, 127.3, 126.6, 125.0, 124.5, 122.1, 119.4, 115.0, 112.6, 51.0, 31.1, 26.7, 21.1, 20.8, 19.2 ppm; (LCMS): *m/z* [M + Na]⁺: 315; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₁₉H₂₀NO₂: 294.1488; found: 294.1480; IR (film): *v_{max}* 1689, 1432, 1216, 1048, 803, 770, 663 cm⁻¹.

l-(7-*Bromo-2-cyclopropylnaphtho*[2, *l*-*b*]*furan-1-yl*)*pyrrolidin-2-one* (**4k**). Following general experimental procedure-I, the product was obtained as white solid (232 mg, 73%); mp: 145–146 °C; ¹H NMR (500 MHz, CDCl₃): δ 8.01 (d, *J* = 2 Hz, 1H), 7.79 (d, *J* = 8.5 Hz, 1H), 7.59-7.57 (m, 1H), 7.52-7.48 (m, 2H), 3.93-3.90 (m, 1H), 3.80-3.77 (m, 1H), 2.74-2.68 (m, 2H), 2.38-2.35 (m, 2H), 1.98-1.95 (m, 1H), 1.20-1.18 (m, 1H), 1.06-1.02 (m, 3H); ¹³C {1H}(125 MHz, CDCl₃) δ 175.8, 155.0, 150.3, 131.9, 130.8, 129.6, 125.2, 123.8, 123.7, 119.9, 118.1, 116.4, 113.4, 50.6, 31.0, 19.2, 7.6, 7.5, 6.9.2 ppm; (LCMS): *m*/*z* [M + Na]⁺: 370; HRMS (ESI-TOF): m/*z* [M + H]⁺ calculated for C₁₉H₁₇BrNO₂: 370.0442; found: 370.0444.

1-(2-Phenylnaphtho[2,1-*b*]*furan-1-yl*)*azepan-2-one* (**4**I). Following general experimental procedure-I, the product was obtained as white solid (170 mg, 73%); mp: 210–211°C;¹H NMR (500 MHz, CDCl₃): δ 8.16 (d, J = 8 Hz, 1H), 7.93 (d, J = 8 Hz, 1H), 7.82 (d, J = 7.5 Hz, 2H), 7.74 (d, J = 9 Hz, 1H), 7.65 (d, J = 8.5 Hz, 1H), 7.58-7.55 (m, 1H), 7.49-7.46 (m, 3H), 7.39 (t, J = 7.5 Hz, 1H), 3.88-3.83 (m, 1H), 3.70-3.65 (m, 1H), 3.01-2.92 (m, 2H), 2.10-2.05 (m, 1H), 2.00-1.94 (m, 1H), 1.89-1.80 (m, 4H); ¹³C{1H}(100 MHz, CDCl₃) δ 177.2, 151.4, 148.9, 130.9, 129.7, 129.1, 128.9, 128.8, 128.4, 127.5, 126.9, 126.7, 126.4, 124.7, 123.6, 122.3, 119.7, 112.7, 54.2, 38.1, 30.3, 28.8, 23.0 ppm; (LCMS): *m/z* [M + H]+: 356; HRMS (ESI-TOF): m/z [M + Na]⁺ calculated for C₂₄H₂₁NO₂Na: 378.1470; found: 378.1476.

1-(2-(4-Bromophenyl)naphtho[2,1-b]furan-1-yl)azepan-2-one (**4m**). Following general experimental procedure-I, the product was obtained as white solid (131 mg, 70%); mp: 207–208 °C;¹H NMR (400 MHz, CDCl₃): δ 8.13 (d, J = 8 Hz, 1H), 7.94 (d, J = 7.2 Hz, 1H), 7.75 (d, J = 9.2 Hz, 1H), 7.72-7.70 (m, 2H), 7.65-7.61 (m, 2H), 7.60-7.56 (m, 2H), 7.51-7.47 (m, 1H), 3.91-3.85 (m, 1H), 3.68-3.62 (m, 1H), 3.02-2.90 (m, 2H), 2.13-2.05 (m, 1H), 1.95-1.79 (m, 4H), 1.57-1.51 (m, 1H); ¹³C{1H}(100 MHz, CDCl₃) δ 177.1, 151.5, 147.8, 132.0, 130.9, 129.2, 128.6, 128.3, 127.4, 126.8, 126.8, 124.9, 123.9, 123.1, 122.2, 119.6, 112.6, 54.1, 38.1, 30.3, 28.9, 23 ppm; (LCMS): m/z [M + H]⁺: 434; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₄H₂₁BrNO₂: 434.0756; found: 434.0754; IR (film): v_{max} 1644, 1481, 1218, 829, 772, 722 cm⁻¹.

l-(2-(4-Chlorophenyl)naphtho[2,1-*b*]*furan-1-yl*)*azepan-2-one* (**4n**). Following general experimental procedure-I, the product was obtained as white solid (142 mg, 68%); mp: 205–206 °C;¹H NMR (400 MHz, CDCl₃): δ 8.13 (d, J = 8.4 Hz, 1H), 7.93 (d, J = 8 Hz, 1H), 7.79-7.74 (m, 3H), 7.63 (d, J = 9.2 Hz, 1H), 7.60-7.56 (m, 1H), 7.51-7.48 (m, 1H), 7.47-7.44 (m, 2H), 3.90-3.84 (m, 1H), 3.67-3.62 (m, 1H), 3.02-2.90 (m, 2H), 2.13-2.04 (m, 1H), 1.94-1.89 (m, 2H), 1.86-1.76 (m, 2H), 1.56-1.47 (m, 1H); ¹³C{1H}(100 MHz, CDCl₃) δ 177.1, 151.5, 147.8, 134.8, 130.9, 129.2, 129.1, 128.2, 128.1, 127.4, 126.8, 124.8, 123.8, 122.2, 119.6, 112.6, 54.1, 38.1, 30.3, 28.9, 23.0 ppm; (LCMS): *m/z* [M + H]⁺: 390; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₄H₂₁CINO₂: 390.1261; found: 390.1260

1-(5-Methoxy-2-phenylbenzofuran-3-yl)pyrrolidin-2-one (6a). Following general experimental procedure-I, the product was obtained as white solid(161 mg, 80%); mp: 159-160 °C; ¹H NMR (500 MHz, CDCl₃): δ 7.78 (t, *J* = 1.5 Hz, 2H), 7.45 (t, *J* = 7.5 Hz, 2H), 7.39 (t, *J* = 9 Hz, 2H), 6.93-6.91 (m, 1H), 6.82 (d, *J* = 2.5 Hz, 1H), 3.85 (s, 3H), 3.71 (t, *J* = 6.5 Hz, 2H), 2.70 (t, *J* = 8.0 Hz, 2H), 2.33-2.29 (m, 2H); ¹³C{1H}(125 MHz, CDCl₃) δ 175.6, 156.4, 150.4, 148.3, 129.7, 129.1, 128.9, 126.7, 126.0, 116.0, 114.2, 112.4, 101.4, 56.1, 49.1, 31.1, 19.5 ppm; (LCMS): *m/z* [M + Na]⁺: 330; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₁₉H₁₈NO₃: 308.1281; found: 308.1280.

1-(2-(4-Chlorophenyl)-5-methoxybenzofuran-3-yl)pyrrolidin-2-one (**6b**). Following general experimental procedure-I, the product was obtained as white solid(139 mg, 76%);mp: 165-166 °C; ¹H NMR (500 MHz, CDCl₃): δ 7.70-7.69 (m, 2H), 7.43-7.38 (m, 3H), 6.94-6.92 (m, 1H), 6.81 (d, *J* = 2.5 Hz, 1H), 3.84 (s, 3H), 3.70 (t, *J* = 7.0 Hz, 2H), 2.69 (t, *J* = 8.0 Hz, 2H), 2.33-2.29 (m, 2H); ¹³C{1H}(125 MHz, CDCl₃) δ 175.5, 156.4, 149.3, 148.3, 134.9, 129.2, 128.1, 127.2, 126.5, 116.3, 114.4, 112.4, 101.4, 56.0, 49.1, 31.0, 19.4 ppm; (LCMS): *m/z* [M + Na]⁺: 363; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₁₉H₁₇ClNO₃: 342.0891; found: 342.0885; IR (film): *v_{max}* 1690, 1452,1222, 847, 779, 640 cm⁻¹.

1-(2,5-Diphenylbenzofuran-3-yl) pyrrolidin-2-one (**6c**). Following general experimental procedure-I, the product was obtained as white solid (190 mg, 82%); mp: 178–179 °C; ¹H NMR (500 MHz, CDCl₃): δ 7.82-7.80 (m, 2H), 7.62-7.60 (m, 2H), 7.58-7.53 (m, 3H), 7.49-7.43 (m, 4H), 7.41-7.38 (m, 1H), 7.36-7.33 (m, 1H), 3.74 (t, *J* = 6.5 Hz, 2H), 2.71 (t, *J* = 6.5 Hz, 2H), 2.34-2.28 (m, 2H); ¹³C{1H}(125 MHz, CDCl₃) δ 175.7, 153.0, 150.3, 141.5, 137.2, 129.5, 129.2, 129.0, 128.8, 127.7, 127.1, 126.7, 126.2, 125.0, 117.7, 116.1, 112.0, 49.2, 31.1, 19.5 ppm; (LCMS): *m/z* [M + H]⁺: 354; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₂₄H₂₀NO₂: 354.1488; found: 354.1481; IR (film): *v_{max}* 2359, 1685, 1466, 1260, 811, 763, 650 cm⁻¹.

1-(4,10-Dimethyl-2,5,5-triphenyl-5H-fluoreno[3,2-b]furan-3-yl)pyrrolidin-2-one (8*a*). Following general experimental procedure-I, the product was obtained as white solid (300 mg, 84%); mp: 296–297 °C; ¹H NMR (CDCl₃, 500 MHz): δ 7.96 (d, *J* = 7.5 Hz, 1H), 7.82 (d, *J* = 1 Hz 2H), 7.49 (t, *J* = 7 Hz, 2H), 7.42 (d, *J* = 7.5 Hz 1H), 7.37-7.33 (m, 7H), 7.28-7.20 (m, 7H), 3.64-3.60 (m, 2H), 2.96 (s, 3H), 2.62-2.52 (m, 2H), 2.20-2.17 (m, 2H), 2.08 (s, 2H) ppm; ¹³C NMR (CDCl₃, 125 MHz): 176.5, 155.5, 153.1, 150.7, 145.8, 143.3, 142.2, 140.7, 136.8, 129.7, 129.3, 129.0 (2), 128.1, 127.9, 127.4, 127.3, 126.6, 126.3, 125.9, 125.5, 125.2, 124.5, 122.8, 116.5, 115.0, 64.6, 50.5, 31.1, 19.1, 14.7, 12.4 ppm; (LCMS): *m/z* [M + H]⁺:

546; HRMS (ESI-TOF): m/z $[M + H]^+$ calculated for C₃₉H₃₂NO₂: 546.2427; found: 546.2424; IR (film): v_{max} 2936, 1692, 1491, 1220, 835, 764, 643 cm⁻¹.

1-(2-(4-Bromophenyl)-4,10-dimethyl-5,5-diphenyl-5H-fluoreno[3,2-b]furan-3-yl)pyrrolidin-2-one (**8b**). Following general experimental procedure-I, the product was obtained as white solid (220 mg, 81%); mp: 297–298 °C;¹H NMR (CDCl₃, 500 MHz): δ 7.95 (d, *J* = 8 Hz, 1H), 7.65 (d, *J* = 8.5 Hz 2H), 7.59 (d, *J* = 8.5 Hz 2H), 7.33-7.29 (m, 6H), 7.22-7.16 (m, 7H), 3.60-3.54 (m, 2H), 2.92 (s, 3H), 2.60-2.47 (m, 2H), 2.18-2.15 (m, 2H), 2.05 (s, 3H) ppm; ¹³C NMR (CDCl₃, 125 MHz): 176.5, 155.5, 153.3, 149.8, 146.0, 143.3, 142.2, 140.6, 137.2, 132.3, 129.3, 129.0, 128.7, 128.2, 127.9, 127.5, 127.4(2), 126.7, 126.4, 125.6, 125.4, 124.4, 123.3, 122.9, 117.1, 115.1, 64.7, 50.6, 31.1, 19.2, 14.7, 12.4 ppm; (LCMS): *m/z* [M + H]⁺: 624; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₃₉H₃₁BrNO₂: 624.1538; found: 624.1534; IR (film): *v_{max}* 2922, 1686, 1484, 1339, 832, 771, 686 cm⁻¹.

1-(4,10-Dimethyl-5,5-diphenyl-2-(p-tolyl)-5H-fluoreno[3,2-b]furan-3-yl)pyrrolidin-2-one (**8c**). Following general experimental procedure-I, the product was obtained as white solid (285 mg, 85%); mp: 291–292°C; ¹H NMR (CDCl₃, 400 MHz): δ 7.94 (d, *J* = 7.6 Hz, 1H), 7.67 (d, *J* = 8 Hz 2H), 7.35-7.29 (m, 6H), 7.27-7.25 (m, 2H), 7.22-7.16 (m, 7H), 3.59-3.55 (m, 2H), 2.93 (s, 3H), 2.58-2.48 (m, 2H), 2.40 (s, 3H), 2.16-2.12 (m, 2H), 2.05 (s, 3H) ppm; ¹³C NMR (CDCl₃, 100 MHz): 176.6, 155.5, 153.0, 151.1, 145.7, 143.4, 142.3,140.8,139.2, 136.6, 129.8, 129.3, 129.0, 128.2, 127.9, 127.3(2), 126.9, 126.6, 126.3, 125.9, 125.6, 125.1, 124.7, 122.8, 115.9,114.9, 64.6, 50.6, 31.1, 21.6, 19.2, 14.7, 12.4 ppm; (LCMS): *m/z* [M + H]⁺: 560; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₄₀H₃₄NO₂: 560.2584; found: 560.2577; IR (film): *v_{max}* 2919, 1686, 1448, 1287, 840, 722, 650 cm⁻¹.

1-(5-(4-Chlorophenyl)-4,10-dimethyl-2,5-diphenyl-5H-fluoreno[3,2-b]furan-3-yl)pyrrolidin-2-one (**8d**). Following general experimental procedure-I, the product was obtained as white solid (288 mg, 76%); mp: 266–267 °C; ¹H NMR (CDCl₃, 400 MHz): δ 7.95 (d, *J* = 8 Hz, 1H), 7.80 (d, *J* = 0.8 Hz 2H), 7.48-7.44 (m, 2H), 7.40 (d, *J* = 7.2 Hz 1H) 7.32-7.30 (m, 2H), 7.29-7.24 (m, 3H), 7.23-7.21 (m, 2H), 7.20-7.16 (m, 5H), 3.61-3.58 (m, 2H), 2.93 (s, 3H), 2.60-2.49 (m, 2H), 2.19-2.15 (m, 2H), 2.05 (s, 3H) ppm; ¹³C NMR (CDCl₃, 125 MHz): 176.6, 155.5, 153.2, 150.8, 145.8, 143.3, 142.2, 140.7, 136.8, 129.7, 129.3, 129.1(2),129.0, 128.2, 127.9, 127.4, 127.3, 126.7, 126.4, 125.9, 125.6, 125.2, 124.6, 122.9, 116.5, 115.0, 64.6, 50.6, 31.1, 19.2, 14.7, 12.4 ppm; (LCMS): *m/z* [M + H]⁺: 580; HRMS (ESI-TOF): m/z [M + H]⁺ calculated for C₃₉H₃₁CINO₂: 580.2043; found: 580.2039

1-(5-(4-Chlorophenyl)-4, 10-dimethyl-5-phenyl-2-(p-tolyl)-5H-fluoreno[3,2-b]furan-3-yl)pyrrolidin-2-one (**8e**). Following general experimental procedure-I, the product was obtained as white solid (278 mg, 78%); mp: 288–289 °C; ¹H NMR (CDCl₃, 500 MHz): δ 7.94 (d, *J* = 7.5 Hz, 1H), 7.68 (t, *J* = 7 Hz 2H), 7.34-7.27 (m, 5H), 7.25-7.16 (m, 9H), 3.60-3.56 (m, 2H), 2.92 (s, 3H), 2.61-2.55 (m, 1H), 2.52-2.45 (m, 1H), 2.40(s, 3H) 2.18-2.13 (m, 2H), 2.04 (s, 3H) ppm; ¹³C NMR (CDCl₃, 100 MHz): 176.6, 154.9, 153.0, 151.2(2), 145.2, 142.8, 142.1, 141.8, 141.0, 140.7, 139.3, 136.4, 132.5, 132.1, 130.7, 130.4, 129.8, 129.1, 128.8, 128.2, 128.1, 127.5, 127.4, 126.8, 126.5, 125.9, 125.4, 124.9, 124.7, 122.9, 115.8, 115.1, 64.1, 50.5, 31.1, 21.6, 19.2, 14.7, 12.4 ppm; (LCMS): *m/z* [M + H]⁺: 594; HRMS (ESI-TOF):

 $m/z [M + H]^+$ calculated for C₄₀H₃₃ClNO₂: 594.2210; found: 594.2214; IR (film): v_{max} 2918, 1691, 1450, 1220, 820, 772, 644 cm⁻¹.

1-(4,7,10-Trimethyl-2,5,5-triphenyl-5H-fluoreno[3,2-b]furan-3-yl)pyrrolidin-2-one (**8f**). Following general experimental procedure-I, the product was obtained as white solid (308 mg, 84%); mp: 293–294 °C;¹H NMR (CDCl₃, 500 MHz): δ 7.94 (d, *J* =7.5 Hz, 1H), 7.68 (d, *J* = 8.5 Hz 2H), 7.34-7.30 (m, 5H), 7.27-7.25 (m, 3H), 7.21-7.16 (m, 7H), 3.60-3.56 (m, 2H), 2.93 (s, 3H), 2.60-2.55 (m, 1H), 2.52-2.46 (m, 1H), 2.40 (s, 3H), 2.18-2.14 (m, 2H), 2.05 (s, 3H) ppm; ¹³C NMR (CDCl₃, 125 MHz): 176.6, 155.5, 153.0, 151.1, 145.7, 143.4, 142.3,140.8,139.2, 136.6, 129.8, 129.3, 129.0, 128.2, 127.9, 127.3(2), 126.9, 125.9, 125.5, 125.1, 124.6, 124.7, 122.8, 115.9, 114.9, 64.5, 50.5, 31.1, 21.5, 19.1, 14.7, 12.4 ppm; (LCMS): *m/z* [M + H]⁺: 560; HRMS (ESI-TOF): m/z [M + Na]⁺ calculated for C₄₀H₃₃NO₂Na: 582.2409; found: 582.2407; IR (film): *v_{max}* 2919, 1686, 1490, 1220, 820, 772, 645 cm⁻¹.

References:

- 1. H. A. Riley and A. R. Gray, Org. Synth., 1935, 15, 67.
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Data for Single X-Ray Crystal Structure 4n.

Vapor diffusion crystallization method was used for crystal growth for **4n** Where compound was dissolved in chloroform by heating to make saturated solution in small vial is placed in closed bottle with other solvent as *n*-hexane.



Figure S1. ORTEP representation of compound 4n and thermal ellipsoids are drawn with 50% probability.

Crystal data and structure refinement for 4n.

Identification code	SHELXL-97	
Empirical formula	$C_{24}H_{20}CINO_2$	
Formula weight	389.86	

Temperature	293 K
Wavelength	0.71073 Å
Crystal system	'Monoclinic'
Space group	P 1 21/c 1
Unit cell dimensions	a = 12. 2034(4) Å α = 90.00.
	b = 8. 3948 (3) Å β = 91.400 (3).
	$c = 19.2731(6) \text{ Å } \gamma = 90.00.$
Volume	1973. 85 (11)
Z	4
Density (calculated)	1.312 Mg/m3
Absorption coefficient	0.213 mm-1
F(000) 816.0	
Crystal size	0.24 x 0.20 x 0.16 mm3
Theta range for data collection	2.11 to 25.00 °.
Index ranges	-14<=h<=14,-9<=k<=9,-22<=l<=22
Reflections collected	18149
Independent reflections	8017 [R(int) = 0.0503]
restraints / parameters	1 / 798
Goodness-of-fit on F ²	1.067
Final R indices [I>2sigma(I)]	R1 = 0.0665, wR2 = 0.1254
R indices (all data)	R1 = 0.0436, wR2 = 0.1113



 $^1\mathrm{H}$ and $^{13}\mathrm{C}\{1\mathrm{H}\}$ spectra of $\!4b$



¹H and ¹³C{1H} spectra of 4c



 ^{1}H and $^{13}C{1H}$ spectra of 4d



 1H and $^{13}C\{1H\}$ spectra of 4e



 1 H and 13 C{1H} spectra of 4f



 ^{1}H and $^{13}\text{C}\{1\text{H}\}$ spectra of 4g



 ^{1}H and $^{13}C{1H}$ spectra of **4h**



1H and $^{13}C\{1H\}$ spectra of4i



¹H and ¹³C{1H} spectra of4j



¹H and ¹³C{1H} spectra of $4\mathbf{k}$



^{1}H and $^{13}\text{C}\{1\text{H}\}$ spectra of4l



¹H and ¹³C{1H} spectra of 4m



¹H and ¹³C{1H} spectra of 4n



¹H and ¹³C{1H} spectra of **6a**



^{1}H and $^{13}C{1H}$ spectra of **6b**



¹H and ¹³C{1H} spectra of 6c



¹H and ¹³C{1H} spectra of 8a



¹H and ¹³C{1H} spectra of **8b**



¹H and ¹³C{1H} spectra of 8c



 1 H and 13 C{1H} spectra of 8d



¹H and ¹³C{1H} spectra of 8e



¹H and ¹³C{1H} spectra of **8f**









