

Supporting Information Part II for

Heterogeneous ditopic ZnFe₂O₄ catalyzed synthesis of 4*H*-pyrans: further conversion to 1,4-DHPs and report of functional group interconversion from amide to ester

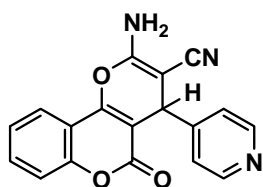
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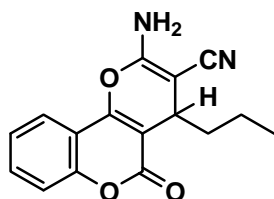
Spectroscopic characterization of 7aa-7br

2-amino-4,5-dihydro-5-oxo-4-(pyridin-4-yl)pyrano[3,2-c]chromene-3-carbonitrile (7aa):



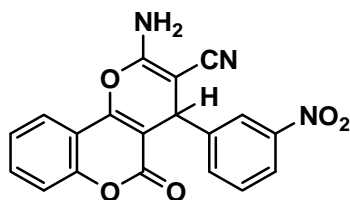
Cream colored solid; Yield: 298 mg, 94 %; m.p. 260-271 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{18}H_{11}N_3O_3$: C, 68.14; H, 3.49; N, 13.24 %. Found: C, 68.27; H, 3.51; N, 13.24 %; IR (KBr) cm^{-1} : 3433, 3296, 3191, 2190, 1650, 1603, 1397, 1295, 1166, 1108, 1010, 753, 469; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.04 (1H, s, CH), 7.14-7.26 (4H, m, NH_2 +arom.), 7.49 (2H, t, $J = 7.8$ Hz, arom.), 7.77 (2H, d, $J = 7.8$ Hz, arom.), 8.35 (2H, br s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 22.3, 36.7, 44.0, 57.0, 103.0, 113.0, 116.8, 119.4, 122.5, 124.7, 130.3, 133.2, 134.9, 145.5, 147.2, 152.3, 153.1, 158.2, 160.1.

2-amino-4,5-dihydro-5-oxo-4-propylpyrano[3,2-c]chromene-3-carbonitrile (7ab):



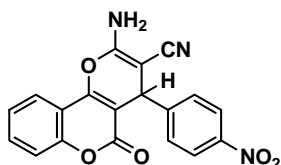
Light yellow colored solid; Yield: 257 mg, 91 %; m.p. 193-195 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{16}H_{14}N_2O_3$: C, 68.07; H, 5.00; N, 9.92 %. Found: C, 68.16; H, 5.01; N, 9.94 %; IR (KBr) cm^{-1} : 3246, 3051, 1629, 1591, 1509, 1460, 1403, 1277, 1214, 1169, 955, 845, 813, 740, 473; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.73-0.91 (3H, m, CH_3), 1.27-1.31 (2H, m, CH_2), 1.44-1.53 (1H, m, CH), 1.60-1.72 (1H, m, CH), 3.36-3.39 (1H, m, CH), 7.26 (2H, s, NH_2), 7.33-7.44 (2H, m, arom.), 7.65 (1H, t, $J = 7.2$ Hz, arom.), 7.76 (1H, d, $J = 7.8$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.3, 18.1, 31.3, 36.7, 55.7, 104.8, 113.4, 117.0, 120.1, 122.6, 125.1, 133.2, 152.5, 154.6, 159.9, 160.4.

2-amino-4,5-dihydro-4-(3-nitrophenyl)-5-oxopyrano[3,2-c]chromene-3-carbonitrile (7ac):



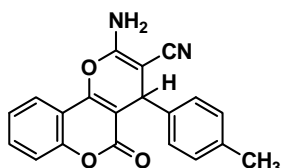
Yellow colored solid; Yield: 332 mg, 92 %; m.p. 257-258 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{11}N_3O_5$: C, 63.16; H, 3.07; N, 11.63 %. Found: C, 63.19; H, 3.06; N, 11.66 %; IR (KBr) cm^{-1} : 3382, 3235, 3179, 2193, 1728, 1663, 1600, 1416, 1298, 1173, 1119, 1010, 753, 472; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 4.69 (1H, s, CH), 7.42 (1H, d, $J = 8.7$ Hz, arom.), 7.48 (1H, d, $J = 7.8$ Hz, arom.), 7.52 (2H, s, NH_2), 7.59 (1H, t, $J = 7.8$ Hz, arom.), 7.68 (1H, dt, $J = 8.0$ Hz, $J = 8.0$ Hz, $J = 1.4$ Hz, arom.), 7.76 (1H, t, $J = 7.8$ Hz, arom.), 7.87 (1H, d, $J = 7.2$ Hz, arom.), 8.08 (2H, d, $J = 7.8$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 22.3, 36.8, 43.9, 57.1, 103.0, 113.0, 116.7, 119.0, 122.5, 124.8, 130.2, 133.2, 134.8, 145.6, 148.0, 152.4, 154.0, 158.3, 159.7.

2-amino-4,5-dihydro-4-(4-nitrophenyl)-5-oxopyrano[3,2-c]chromene-3-carbonitrile (7ad):



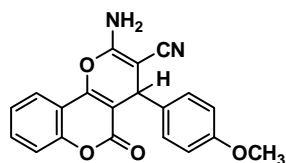
Yellow colored solid; Yield: 336 mg, 93 %; m.p. 251-253 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{11}N_3O_5$: C, 63.16; H, 3.07; N, 11.63 %. Found: C, 63.17; H, 3.08; N, 11.65 %; IR (KBr) cm^{-1} : 3390, 3212, 3179, 2197, 1662, 1575, 1465, 1409, 1260, 1227, 746, 548; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 4.64 (1H, s, CH), 7.44 (2H, t, $J = 7.5$ Hz, arom.), 7.49-7.54 (2H, m, arom.), 7.57 (2H, s, NH_2), 7.69 (1H, t, $J = 7.5$ Hz, arom.), 7.87 (1H, d, $J = 7.5$ Hz, arom.), 8.14 (2H, d, $J = 8.4$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 22.3, 36.9, 43.9, 56.9, 102.9, 113.0, 116.7, 118.9, 122.7, 123.8, 124.8, 129.2, 133.2, 146.7, 150.8, 152.4, 154.0, 158.1, 159.6.

2-amino-4,5-dihydro-5-oxo-4-p-tolylpyrano[3,2-c]chromene-3-carbonitrile (7ae):



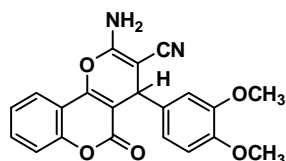
Grayish solid; Yield: 304 mg, 92 %; m.p. 252-254 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{14}N_2O_3$: C, 72.72; H, 4.27; N, 8.48 %. Found: C, 72.74; H, 4.27; N, 8.47 %; IR (KBr) cm^{-1} : 3319, 3310, 3195, 2196, 1718, 1676, 1608, 1377, 1057, 954, 757, 506; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 2.21 (3H, s, CH_3), 4.36 (1H, s, CH), 7.05-7.11 (4H, m, arom.), 7.34 (2H, s, NH_2), 7.39-7.47 (2H, m, arom.), 7.66 (1H, t, $J = 9.0$ Hz, arom.), 7.86 (1H, d, $J = 9.0$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 20.7, 36.7, 58.2, 104.2, 113.1, 116.6, 117.8, 119.3, 122.5, 124.7, 127.6, 129.1, 132.9, 136.3, 140.5, 152.2, 153.3, 158.0, 159.6.

2-amino-4,5-dihydro-4-(4-methoxyphenyl)-5-oxopyrano[3,2-c]chromene-3-carbonitrile (7af):



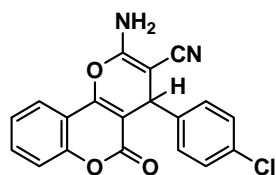
White solid; Yield: 304 mg, 88 %; m.p. 221-223 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{14}N_2O_4$: C, 69.36; H, 4.07; N, 8.09 %. Found: C, 69.38; H, 4.06; N, 8.10 %; IR (KBr) cm^{-1} : 3370, 3290, 3182, 2191, 1709, 1671, 1605, 1571, 1507, 1459, 1379, 1319, 1251, 1178, 1111, 1052, 1026, 951, 834, 756, 564, 529; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 3.68 (3H, s, OCH₃), 4.35 (1H, s, CH), 6.82 (2H, d, $J = 8.4$ Hz, arom.), 7.13 (2H, d, $J = 8.4$ Hz, arom.), 7.33 (2H, s, NH₂), 7.38-7.47 (1H, m, arom.), 7.63-7.69 (1H, m, arom.), 7.84 (1H, dd, $J = 7.5$ Hz, $J = 1.2$ Hz, arom.), 7.93 (1H, d, $J = 9.0$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 36.2, 55.1, 58.4, 104.3, 114.0, 115.3, 116.6, 119.4, 122.5, 124.8, 128.8, 132.9, 133.5, 135.5, 152.2, 153.1, 158.0, 159.6, 160.5.

2-amino-4,5-dihydro-4-(3,4-dimethoxyphenyl)-5-oxopyrano[3,2-c]chromene-3-carbonitrile (7ag):



Light yellow colored solid; Yield: 323 mg, 86 %; m.p. 228-230 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{16}N_2O_5$: C, 67.02; H, 4.28; N, 7.44 %. Found: C, 67.10; H, 4.30; N, 7.45 %; IR (KBr) cm^{-1} : 3402, 3323, 3204, 2197, 1714, 1670, 1604, 1509, 1379, 1264, 1143, 1047, 761, 481; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 3.71 (6H, s, OCH₃), 4.41 (1H, s, CH), 6.75 (1H, d, $J = 8.1$ Hz, arom.), 6.85-6.89 (2H, m, arom.), 7.36 (2H, s, NH₂), 7.43-7.50 (2H, m, arom.), 7.70 (1H, t, $J = 8.1$ Hz, arom.), 7.89 (1H, d, $J = 7.8$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 36.6, 55.6, 55.7, 58.3, 104.3, 112.1, 113.1, 116.6, 117.8, 119.8, 122.7, 124.8, 132.9, 135.9, 148.1, 148.7, 152.2, 153.4, 158.1, 159.7, 162.0.

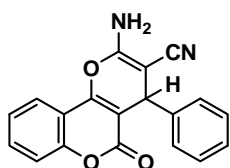
2-amino-4-(4-chlorophenyl)-4,5-dihydro-5-oxopyrano[3,2-c]chromene-3-carbonitrile (7ah):



Light yellow colored solid; Yield: 315 mg, 90 %; m.p. 265-267 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{11}ClN_2O_3$: C, 65.06; H, 3.16; N, 7.99 %. Found: C, 65.11; H, 3.19; N, 8.02 %; IR (KBr) cm^{-1} : 3402,

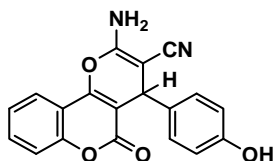
3323, 3204, 2197, 1714, 1670, 1604, 1509, 1379, 1264, 1143, 1047, 761, 481; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 4.46 (1H, s, CH), 7.23 (2H, d, $J = 8.4$ Hz, arom.), 7.43-7.50 (6H, m, NH_2 +arom.), 7.68-7.72 (1H, m, arom.), 7.88 (1H, d, $J = 7.2$ Hz, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 36.4, 57.7, 103.6, 113.1, 116.6, 119.1, 122.6, 124.8, 128.6, 129.6, 131.7, 133.1, 142.4, 152.3, 153.6, 158.1, 159.7.

2-amino-4,5-dihydro-5-oxo-4-phenylpyrano[3,2-c]chromene-3-carbonitrile (7ai):



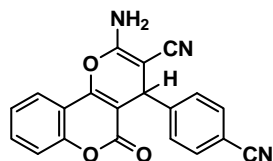
White solid; Yield: 294 mg, 93 %; m.p. 255-257 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $\text{C}_{19}\text{H}_{12}\text{N}_2\text{O}_3$: C, 72.15; H, 3.82; N, 8.86 %. Found: C, 72.11; H, 3.81; N, 8.84 %; IR (KBr) cm^{-1} : 3323, 3204, 2195, 1720, 1668, 1601, 1519, 1381, 1264, 1143, 1048, 761, 481; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 4.41 (1H, s, CH), 7.21-7.30 (5H, m, arom.), 7.36 (2H, s, NH_2), 7.40-7.48 (2H, m, arom.), 7.69 (1H, t, $J = 7.2$ Hz, arom.), 7.86 (1H, d, $J = 7.2$ Hz, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 37.1, 58.1, 104.1, 113.0, 116.6, 119.3, 122.6, 124.7, 127.2, 127.7, 128.6, 133.0, 143.4, 152.2, 153.5, 158.1, 159.6.

2-amino-4,5-dihydro-4-(4-hydroxyphenyl)-5-oxopyrano[3,2-c]chromene-3-carbonitrile (7aj):



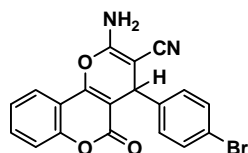
White solid; Yield: 305 mg, 92 %; m.p. 258-260 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $\text{C}_{19}\text{H}_{12}\text{N}_2\text{O}_4$: C, 68.67; H, 3.64; N, 8.43 %. Found: C, 68.71; H, 3.66; N, 8.46 %; IR (KBr) cm^{-1} : 3504, 3407, 3281, 3183, 2197, 1690, 1674, 1609, 1512, 1380, 1171, 1070, 844, 561; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 4.29 (1H, s, CH), 6.65 (2H, d, $J = 8.1$ Hz, arom.), 7.01 (2H, d, $J = 8.1$ Hz, arom.), 7.29 (2H, s, NH_2), 7.38-7.46 (2H, m, arom.), 7.65 (1H, t, $J = 7.8$ Hz, arom.), 7.84 (1H, d, $J = 7.8$ Hz, arom.), 9.31 (1H, s, OH); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 36.2, 58.5, 104.6, 113.1, 115.3, 116.6, 119.4, 122.5, 124.7, 128.7, 132.8, 133.7, 152.1, 153.0, 156.5, 158.0, 159.6, 161.9.

2-amino-4-(4-cyanophenyl)-4,5-dihydro-5-oxopyrano[3,2-c]chromene-3-carbonitrile (7ak):



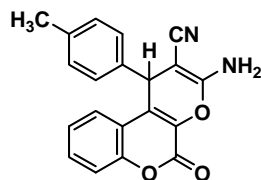
Yellow colored solid; Yield: 317 mg, 93 %; m.p. 200-202 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{11}N_3O_3$: C, 70.38; H, 3.25; N, 12.31 %. Found: C, 70.41; H, 3.26; N, 12.35 %; IR (KBr) cm^{-1} : 3399, 3325, 3200, 2236, 2192, 1742, 1665, 1603, 1411, 1161, 1118, 1011, 760, 551, 462; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 4.56 (1H, s, CH), 7.41-7.46 (4H, m, arom.), 7.48 (2H, s, NH_2), 7.69 (1H, t, $J = 7.5$ Hz, arom.), 7.73 (2H, d, $J = 8.1$ Hz, arom), 7.85 (1H, d, $J = 7.5$ Hz, arom); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 38.0, 56.9, 109.6, 116.6, 116.9, 118.7, 120.2, 123.5, 125.1, 127.4, 128.1, 128.6, 130.0, 130.9, 132.9, 147.0, 151.0, 160.0.

2-Amino-4-(4-bromo-phenyl)-5-oxo-4H,5H-pyrano[3,2-c]chromene-3-carbonitrile (7al):



Yellow colored solid; Yield: 367 mg, 93 %; m.p. 220-222 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{11}BrN_2O_3$: C, 57.74; H, 2.81; N, 7.09 %. Found: C, 57.77; H, 2.82; N, 7.08 %; IR (KBr) cm^{-1} : 3385, 3305, 3188, 2191, 1712, 1674, 1606, 1375, 1060, 759, 510; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.12 (1H, s, CH), 7.17-7.23 (3H, m, NH_2 +arom.), 7.34 (3H, t, $J = 8.7$ Hz, arom.), 7.46 (4H, t, $J = 10.1$ Hz, arom); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 37.0, 56.6, 116.5, 116.9, 119.5, 120.7, 124.8, 125.1, 125.8, 129.8, 130.4, 131.9, 134.5, 142.5, 150.3, 154.1, 159.0.

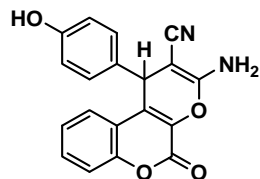
3-amino-1,5-dihydro-5-oxo-1-p-tolylpyrano[2,3-c]chromene-2-carbonitrile (7am):



Cream colored solid; Yield: 290 mg, 88 %; m.p. 231-233 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{14}N_2O_3$: C, 72.72; H, 4.27; N, 8.48 %. Found: C, 72.71; H, 4.26; N, 8.49 %; IR (KBr) cm^{-1} : 3433, 3301, 2201, 1730, 1662, 1409, 1168, 755, 465; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 2.23 (3H, s, CH_3), 5.05 (1H, s, CH), 7.11 (2H, d, $J = 7.2$ Hz, arom.), 7.19 (2H, s, NH_2), 7.22-7.27 (3H, m, arom.), 7.37 (1H, d, $J = 8.1$ Hz, arom.), 7.45-7.53

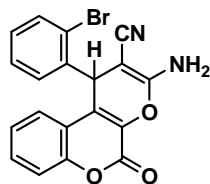
(2H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 20.8, 37.3, 57.1, 116.4, 117.2, 124.7, 125.3, 126.6, 127.5, 129.6, 130.2, 136.7, 141.2, 150.2, 154.1, 159.0.

3-amino-1,5-dihydro-1-(4-hydroxyphenyl)-5-oxopyrano[2,3-c]chromene-2-carbonitrile (7an):



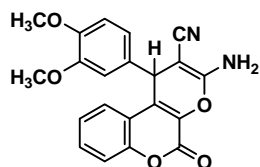
White solid; Yield: 308 mg, 93 %; m.p. 272-274 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{12}N_2O_4$: C, 68.67; H, 3.64; N, 8.43 %. Found: C, 68.77; H, 3.67; N, 8.42 %; IR (KBr) cm^{-1} : 3509, 3201, 2201, 1727, 1658, 1409, 1168, 755, 458; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.01 (1H, s, CH), 6.83 (2H, d, $J = 8.1$ Hz, arom.), 7.15 (2H, s, NH_2), 7.18-7.21 (1H, m, arom.), 7.26 (2H, d, $J = 7.8$ Hz, arom.), 7.34 (1H, d, $J = 8.1$ Hz, arom.), 7.41-7.46 (1H, m, arom.), 7.49 (1H, d, $J = 7.8$ Hz, arom.), 9.21 (1H, s, OH); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 36.9, 57.4, 105.0, 114.4, 116.4, 117.1, 119.7, 124.7, 125.3, 126.8, 128.7, 130.3, 134.1, 135.3, 150.3, 154.2, 158.5, 158.9, 162.0.

3-amino-1-(2-bromophenyl)-1,5-dihydro-5-oxopyrano[2,3-c]chromene-2-carbonitrile (7ao):



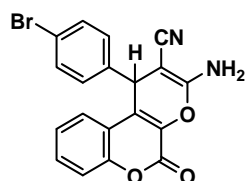
Light brown colored solid; Yield: 359 mg, 91 %; m.p. 275-277 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{11}BrN_2O_3$: C, 57.74; H, 2.81; N, 7.09 %. Found: C, 57.77; H, 2.82; N, 7.10 %; IR (KBr) cm^{-1} : 3443, 3318, 3202, 2194, 1745, 1662, 1626, 1416, 1163, 1119, 992, 747, 462; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.39 (1H, s, CH), 7.14 (1H, d, $J = 7.2$ Hz, arom.), 7.17 (1H, t, $J = 7.2$ Hz, arom.), 7.23-7.29 (3H, m, NH_2 +arom.), 7.32 (1H, s, arom.), 7.35 (1H, d, $J = 6.3$ Hz, arom.), 7.39 (1H, s, arom.), 7.45 (1H, t, $J = 6.9$ Hz, arom.), 7.63 (1H, d, $J = 7.8$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 37.4, 55.6, 116.7, 117.0, 117.7, 118.9, 122.1, 124.2, 125.0, 129.2, 129.7, 130.4, 130.8, 133.1, 134.9, 141.7, 150.1, 153.9, 159.0.

3-amino-1,5-dihydro-1-(3,4-dimethoxyphenyl)-5-oxopyrano[2,3-c]chromene-2-carbonitrile (7ap):



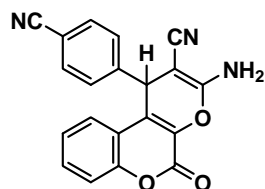
Greyish solid; Yield: 338 mg, 90 %; m.p. 175-177 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{16}N_2O_5$: C, 67.02; H, 4.28; N, 7.44 %. Found: C, 67.15; H, 4.30; N, 7.46 %; IR (KBr) cm^{-1} : 3405, 3321, 3207, 2195, 1717, 1675, 1604, 1503, 1300, 1268, 1145, 1047, 760, 481; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 3.66 (3H, s, OCH₃), 3.69 (3H, s, OCH₃), 4.99 (1H, s, CH), 6.82 (2H, s, arom.), 6.94 (1H, s, arom.), 7.12 (2H, s, NH₂), 7.20 (1H, t, $J = 7.2$ Hz, arom.), 7.36 (1H, d, $J = 7.8$ Hz, arom.), 7.44 (1H, t, $J = 7.8$ Hz, arom.), 7.53 (1H, d, $J = 7.8$ Hz, arom.).

3-amino-1-(4-bromophenyl)-1,5-dihydro-5-oxopyrano[2,3-c]chromene-2-carbonitrile (7aq):



White solid; Yield: 367 mg, 93 %; m.p. 210-212 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{11}BrN_2O_3$: C, 57.74; H, 2.81; N, 7.09 %. Found: C, 57.84; H, 2.84; N, 7.14 %; IR (KBr) cm^{-1} : 3432, 3295, 3179, 2195, 1729, 1654, 1600, 1408, 1298, 1168, 1118, 1010, 753, 469; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.12 (1H, s, CH), 7.17-7.22 (1H, m, arom.), 7.24 (2H, s, NH₂), 7.34 (3H, t, $J = 8.4$ Hz, arom.), 7.42-7.49 (4H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 37.0, 56.6, 116.5, 116.9, 119.4, 120.7, 124.8, 125.1, 129.8, 130.4, 131.9, 134.5, 142.5, 150.3, 154.1, 159.0.

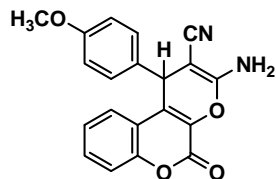
3-amino-1-(4-cyanophenyl)-1,5-dihydro-5-oxopyrano[2,3-c]chromene-2-carbonitrile (7ar):



Yellow colored solid; Yield: 317 mg, 93 %; m.p. 242-243 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{11}N_3O_3$: C, 70.38; H, 3.25; N, 12.31 %. Found: C, 70.40; H, 3.28; N, 12.33 %; IR (KBr) cm^{-1} : 3399, 3325, 3200, 2236, 2192, 1742, 1665, 1603, 1411, 1161, 1118, 1011, 760, 551, 462; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.25 (1H, s, CH), 7.18 (1H, t, $J = 7.2$ Hz, arom.), 7.29 (2H, s, NH₂), 7.38 (2H, t, $J = 8.1$ Hz, arom.), 7.42-7.47 (1H, m, arom.), 7.59 (2H, d, $J = 7.8$ Hz, arom.), 7.76 (2H, d, $J = 7.5$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 37.4,

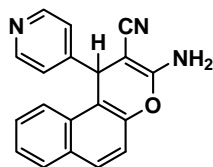
56.0, 110.3, 116.5, 116.8, 118.6, 119.3, 124.8, 125.0, 125.0, 128.6, 130.4, 133.0, 134.8, 148.3, 150.3, 154.0, 159.2, 161.9.

3-amino-1,5-dihydro-1-(4-methoxyphenyl)-5-oxopyrano[2,3-c]chromene-2-carbonitrile (7as):



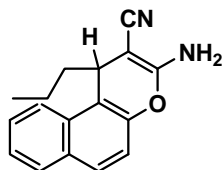
Light colored solid; Yield: 294 mg, 85 %; m.p. 234-236 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{14}N_2O_4$: C, 69.36; H, 4.07; N, 8.09 %. Found: C, 69.40; H, 4.06; N, 8.12 %; IR (KBr) cm^{-1} : 3428, 3288, 3174, 2199, 1728, 1655, 1602, 1509, 1407, 1259, 1167, 1117, 1024, 844, 751, 466; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 3.70 (3H, s, OCH₃), 5.05 (1H, s, CH), 6.87 (2H, d, $J = 6.6$ Hz, arom.), 7.17 (2H, s, NH₂), 7.23-7.31 (3H, m, arom.), 7.38-40 (1H, m, arom.), 7.46-7.55 (2H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 36.8, 55.1, 57.5, 114.3, 116.5, 117.0, 119.9, 124.6, 125.2, 126.7, 128.7, 130.3, 134.0, 135.3, 150.3, 154.1, 158.4, 158.9.

3-amino-1-(pyridin-4-yl)-1H-benzof[*f*]chromene-2-carbonitrile (7at):



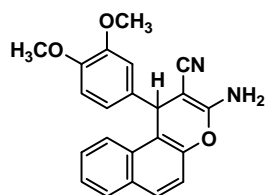
Light brown colored solid; Yield: 284 mg, 95 %; m.p. 260-262 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{13}N_3O$: C, 76.24; H, 4.38; N, 14.04 %. Found: C, 76.37; H, 4.41; N, 14.05 %; IR (KBr) cm^{-1} : 3433, 3297, 3170, 2195, 1729, 1654, 1605, 1410, 1301, 1160, 1111, 1010, 753, 470; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.35 (1H, s, CH), 7.11 (2H, s, NH₂), 7.15 (2H, d, $J = 5.4$ Hz, arom.), 7.32 (1H, d, $J = 9.0$ Hz, arom.), 7.37-7.44 (2H, m, arom.), 7.73-7.78 (1H, m, arom.), 7.88-7.95 (1H, m, arom.), 8.40 (2H, d, $J = 5.1$ Hz, arom.), 8.74 (1H, d, $J = 5.4$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 37.4, 56.5, 114.3, 116.9, 120.2, 122.3, 122.8, 123.5, 125.2, 127.4, 128.8, 130.1, 130.9, 147.1, 150.2, 150.7, 153.8, 160.1, 166.3.

3-amino-1-propyl-1H-benzof[*f*]chromene-2-carbonitrile (7au):



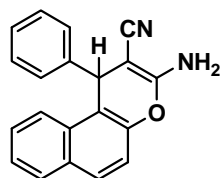
Cream colored solid; Yield: 237 mg, 90 %; m.p. 128-130 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{17}H_{16}N_2O$: C, 77.25; H, 6.10; N, 10.60 %. Found: C, 77.20; H, 6.09; N, 10.62 %; IR (KBr) cm^{-1} : 3433, 3221, 3044, 2362, 2173, 1646, 1588, 1400, 1224, 1024, 806, 750, 497; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.71-0.85 (3H, m, CH_3), 1.05-1.06 (1H, m, CH), 1.19-1.31 (1H, m, CH), 1.55-1.68 (2H, m, CH_2), 3.68 (1H, t, $J = 4.8$ Hz, CH), 6.97 (2H, s, NH_2), 7.33-7.44 (1H, m, arom.), 7.63-7.68 (2H, m, arom.), 7.76 (1H, d, $J = 7.8$ Hz, arom.), 7.83-7.88 (2H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.3, 18.5, 35.4, 50.0, 119.3, 121.0, 121.3, 123.0, 124.0, 126.0, 127.0, 128.1, 133.2, 145.1, 164.5.

3-amino-1-(3,4-dimethoxyphenyl)-1H-benzof[f]chromene-2-carbonitrile (7av):



Cream colored solid; Yield: 322 mg, 90 %; m.p. 170-172 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{18}N_2O_3$: C, 73.73; H, 5.06; N, 7.82 %. Found: C, 73.82; H, 5.08; N, 7.83 %; IR (KBr) cm^{-1} : 3372, 3210, 2197, 1655, 1617, 1574, 1467, 1409, 1260, 1227, 751, 562; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 3.61 (3H, s, OCH_3), 3.63 (3H, s, OCH_3), 5.20 (1H, s, CH), 6.53 (1H, dd, $J = 8.3$ Hz, $J = 1.9$ Hz, arom.), 6.76 (1H, d, $J = 8.4$ Hz, arom.), 6.84 (1H, d, $J = 1.4$ Hz, arom.), 6.90 (2H, s, NH_2), 7.29 (1H, d, $J = 9.0$ Hz, arom.), 7.35-7.43 (2H, m, arom.), 7.83-7.90 (3H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 38.1, 55.9, 56.0, 58.6, 111.6, 112.6, 116.3, 117.2, 119.5, 121.0, 124.2, 125.3, 127.5, 128.9, 129.8, 130.8, 131.3, 138.8, 147.2, 148.0, 149.1, 160.1.

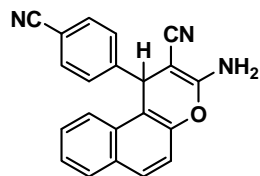
3-amino-1-phenyl-1H-benzof[f]chromene-2-carbonitrile (7aw):



White solid; Yield: 277 mg, 93 %; m.p. 287-289 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{14}N_2O$: C, 80.52; H, 4.73; N, 9.39 %. Found: C, 80.62; H, 4.73; N, 9.40 %; IR (KBr) cm^{-1} : 3380, 3212, 3179, 2195, 1662, 1617, 1464, 1409, 1260, 1227, 748, 562; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.26 (1H, s, CH), 6.94 (2H, s, NH_2), 7.10-7.16 (3H, m, arom.), 7.20 (2H, d, $J = 6.9$ Hz, arom.), 7.30 (1H, d, $J = 9.0$ Hz, arom.), 7.38 (2H, br

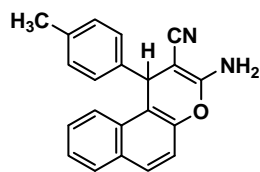
s, arom.), 7.80 (1H, d, $J = 6.9$ Hz, arom.), 7.88-7.91 (2H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 38.2, 58.1, 115.7, 116.8, 120.5, 123.7, 125.0, 126.7, 127.0, 127.1, 128.5, 128.8, 129.5, 130.2, 130.9, 145.8, 146.9, 159.8.

3-amino-1-(4-cyanophenyl)-1H-benzo[f]chromene-2-carbonitrile (7ax):



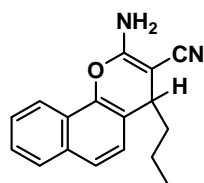
White solid; Yield: 300 mg, 93 %; m.p. 286-288 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{13}N_3O$: C, 78.00; H, 4.05; N, 13.00 %. Found: C, 78.03; H, 4.05; N, 13.03 %; IR (KBr) cm^{-1} : 3446, 3304, 2226, 2184, 1651, 1587, 1626, 1606, 1405, 1238, 1083, 807, 762; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.44 (1H, s, CH), 7.09 (2H, s, NH_2), 7.30-7.39 (5H, m, arom.), 7.68-7.76 (3H, m, arom.), 7.91 (2H, t, $J = 9.0$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 38.0, 56.9, 109.6, 114.6, 116.9, 118.7, 120.2, 123.5, 125.1, 127.4, 128.1, 128.6, 130.0, 130.9, 132.9, 147.0, 151.0, 160.0.

3-amino-1-p-tolyl-1H-benzo[f]chromene-2-carbonitrile (7ay):



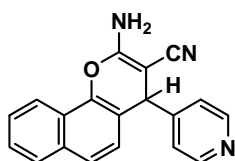
Light colored solid; Yield: 290 mg, 93 %; m.p. 269-271 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{16}N_2O$: C, 80.75; H, 5.16; N, 8.97 %. Found: C, 80.79; H, 5.17; N, 8.99 %; IR (KBr) cm^{-1} : 3379, 3214, 2195, 1662, 1610, 1570, 1464, 1409, 1258, 1227, 748, 562; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 2.15 (3H, s, CH_3), 5.20 (1H, s, CH), 6.92 (2H, s, NH_2), 7.02 (4H, br s, arom.), 7.27 (1H, d, $J = 9.0$ Hz, arom.), 7.34-7.41 (2H, m, arom.), 7.79 (1H, d, $J = 9.0$ Hz, arom.), 7.84-7.89 (2H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 20.6, 37.8, 58.1, 115.9, 116.8, 120.6, 123.7, 124.9, 127.0, 127.1, 128.5, 129.5, 130.3, 130.9, 135.7, 142.9, 146.8, 159.7.

2-amino-4-propyl-4H-benzo[h]chromene-3-carbonitrile (7az):



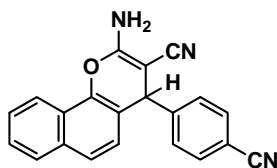
Light yellow colored solid; Yield: 240 mg, 91 %; m.p. 125-127 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₁₇H₁₆N₂O: C, 77.25; H, 6.10; N, 10.60 %. Found: C, 77.25; H, 6.12; N, 10.60 %; IR (KBr) cm⁻¹: 3433, 3295, 3172, 2195, 1730, 1655, 1600, 1412, 1298, 1169, 1118, 1014, 753, 469; δ_H ppm (300 MHz; DMSO-d₆; TMS) 0.71-0.85 (3H, m, CH₃), 1.03-1.06 (1H, m, CH), 1.18-1.30 (1H, m, CH), 1.55-1.66 (2H, m, CH₂), 3.67 (1H, t, *J* = 4.8 Hz, CH), 6.97 (2H, s, NH₂), 7.30 (1H, d, *J* = 8.4 Hz, arom.), 7.48-7.57 (2H, m, arom.), 7.64 (1H, d, *J* = 8.4 Hz, arom.), 7.86 (1H, d, *J* = 7.2 Hz, arom.), 8.12 (1H, d, *J* = 8.1 Hz, arom.); δ_C ppm (75 MHz, DMSO-d₆, TMS) 14.4, 18.1, 35.1, 54.8, 119.1, 121.0, 121.3, 123.1, 124.2, 126.0, 126.9, 128.1, 133.1, 144.3, 161.8.

2-amino-4-(pyridin-4-yl)-4H-benzo[h]chromene-3-carbonitrile (7ba):



Light brown colored solid; Yield: 278 mg, 93 %; m.p. 199-200 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₁₉H₁₃N₃O: C, 76.24; H, 4.38; N, 14.04 %. Found: C, 76.37; H, 4.38; N, 14.06 %; IR (KBr) cm⁻¹: 3430, 3295, 3200, 2195, 1725, 1654, 1600, 1400, 1298, 1169, 1124, 1010, 755, 469; δ_H ppm (300 MHz; DMSO-d₆; TMS) 4.94 (1H, s, CH), 7.09 (1H, d, *J* = 8.4 Hz, arom.), 7.22-7.27 (4H, m, arom.), 7.52-7.63 (3H, m, arom.), 7.86 (1H, d, *J* = 7.8 Hz, arom.), 8.20 (1H, d, *J* = 8.1 Hz, arom.), 8.47 (2H, d, *J* = 4.8 Hz, arom.); δ_C ppm (75 MHz, DMSO-d₆, TMS) 30.7, 54.9, 116.5, 120.2, 120.8, 122.8, 122.9, 124.2, 125.9, 126.9, 127.1, 127.8, 133.0, 143.1, 150.1, 153.8, 160.6.

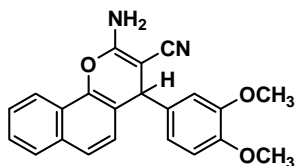
2-amino-4-(4-cyanophenyl)-4H-benzo[h]chromene-3-carbonitrile (7bb):



Yellow colored solid; Yield: 300 mg, 93 %; m.p. 259-261 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₂₁H₁₃N₃O: C, 78.00; H, 4.05; N, 13.00 %. Found: C, 78.00; H, 4.06; N, 13.01 %; IR (KBr) cm⁻¹: 3432, 3295, 3179, 2195, 1729, 1654, 1600, 1408, 1298, 1168, 1118, 1010, 753, 469; δ_H ppm (300 MHz; DMSO-d₆; TMS) 5.06 (1H, s, CH), 7.09 (1H, d, *J* = 8.4 Hz, arom.), 7.30 (2H, s, NH₂), 7.45 (2H, d, *J* = 8.4 Hz, arom.), 7.55-7.66 (3H, m, arom.), 7.79 (2H, d, *J* = 8.1 Hz, arom.), 7.89 (1H, d, *J* = 8.1 Hz, arom.), 8.24 (1H, d, *J* = 8.1 Hz, arom.); δ_C ppm (75 MHz,

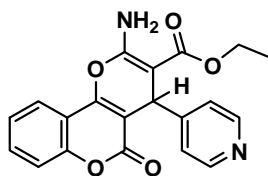
DMSO- d_6 , TMS) 40.9, 55.4, 109.9, 116.8, 118.7, 120.2, 120.8, 122.8, 124.2, 126.0, 126.9, 127.0, 127.8, 128.8, 132.8, 132.9, 143.0, 151.0, 160.4.

2-amino-4-(3,4-dimethoxyphenyl)-4H-benzo[h]chromene-3-carbonitrile (7bc):



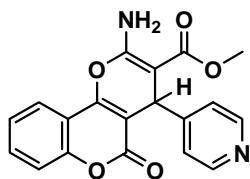
White solid; Yield: 319 mg, 89 %; m.p. 210-211 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{18}N_2O_3$: C, 73.73; H, 5.06; N, 7.82 %. Found: C, 73.87; H, 5.05; N, 7.81 %; IR (KBr) cm^{-1} : 3382, 3212, 2193, 1662, 1617, 1575, 1464, 1409, 1261, 1227, 748, 562; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 3.66 (6H, s, 2*OCH₃), 4.80 (1H, s, CH), 6.68 (1H, dd, $J = 8.3$ Hz, $J = 1.8$ Hz, arom.), 6.83-6.86 (2H, m, arom.), 7.08-7.11 (3H, m, NH₂+arom.), 7.51-7.61 (3H, m, arom.), 7.83 (1H, d, $J = 7.8$ Hz, arom.), 8.24 (1H, d, $J = 8.1$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 55.6, 56.6, 111.7, 112.2, 118.2, 119.9, 120.8, 122.9, 123.8, 126.3, 126.7, 126.8, 127.7, 132.7, 138.3, 142.6, 147.9, 148.9, 160.2.

ethyl 2-amino-4,5-dihydro-5-oxo-4-(pyridin-4-yl)pyrano[3,2-c]chromene-3-carboxylate (7bd):



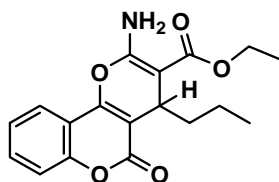
Cream colored solid; Yield: 335 mg, 92 %; m.p. 202-204 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{16}N_2O_5$: C, 65.93; H, 4.43; N, 7.69 %. Found: C, 65.94; H, 4.46; N, 7.71 %; IR (KBr) cm^{-1} : 3358, 3269, 3194, 3084, 2978, 2898, 1719, 1695, 1662, 1549, 1374, 1283, 1194, 1100, 1041, 951, 752, 568; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 1.05 (3H, t, $J = 6.9$ Hz, CH₃), 3.94 (2H, q, $J = 6.9$ Hz, CH₂), 4.64 (1H, s, CH), 7.21 (2H, d, $J = 5.4$ Hz, arom.), 7.39-7.47 (2H, m, arom.), 7.66 (1H, t, $J = 7.2$ Hz, arom.), 7.19-7.94 (3H, m, NH₂+arom.), 8.39 (2H, d, $J = 5.1$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.6, 35.6, 59.6, 76.1, 105.7, 113.5, 117.0, 123.1, 124.1, 125.1, 133.4, 149.3, 152.7, 154.2, 154.4, 159.1, 160.3, 167.7.

methyl 2-amino-4,5-dihydro-5-oxo-4-(pyridin-4-yl)pyrano[3,2-c]chromene-3-carboxylate (7be):



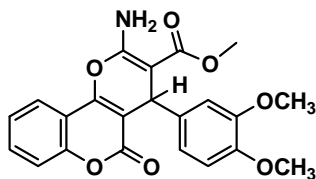
White solid; Yield: 305 mg, 87 %; m.p. 199-200 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{14}N_2O_5$: C, 65.14; H, 4.03; N, 8.00 %. Found: C, 65.17; H, 4.03; N, 8.03 %; IR (KBr) cm^{-1} : 3358, 3265, 3197, 3084, 2978, 2903, 1726, 1695, 1662, 1549, 1374, 1283, 1194, 1100, 1041, 951, 752, 568; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 3.51 (3H, s, OCH₃), 4.66 (1H, s, CH), 7.24 (2H, br s, NH₂), 7.41-7.49 (2H, m, arom.), 7.68 (1H, t, $J = 7.2$ Hz, arom.), 7.90-7.95 (3H, m, arom.), 8.41 (2H, br s, arom); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 39.2, 50.4, 76.1, 119.8, 123.1, 124.1, 125.1, 133.6, 143.3, 147.8, 149.4, 152.8, 154.3, 154.5, 159.1, 160.3, 167.7.

ethyl 2-amino-4,5-dihydro-5-oxo-4-propylpyrano[3,2-c]chromene-3-carboxylate (7bf):



White solid; Yield: 293 mg, 89 %; m.p. 134-136 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{18}H_{19}NO_5$: C, 65.64; H, 5.81; N, 4.25 %. Found: C, 65.64; H, 5.81; N, 4.24 %; IR (KBr) cm^{-1} : 3396, 3290, 2928, 1694, 1537, 1453, 1393, 1281, 1196, 1064, 759, 634, 518; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.71-0.88 (3H, m, CH₃), 1.05 (3H, t, $J = 6.9$ Hz, CH₃), 1.27-1.31 (2H, m, CH₂), 1.44-1.53 (1H, s, CH), 1.60-1.72 (1H, m, CH), 3.36-3.39 (1H, m, CH), 3.92-3.99 (2H, m, CH₂), 7.26 (2H, s, NH₂), 7.33-7.44 (2H, m, arom.), 7.63-7.68 (1H, m, arom.), 7.76 (1H, d, $J = 7.8$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.3, 14.6, 18.1, 31.3, 36.7, 55.7, 76.1, 104.8, 113.4, 120.1, 122.6, 125.1, 133.2, 152.5, 154.6, 159.9, 160.4, 167.7.

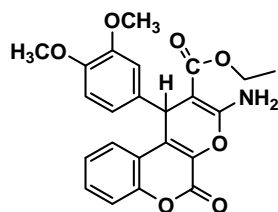
methyl 2-amino-4,5-dihydro-4-(3,4-dimethoxyphenyl)-5-oxopyrano[3,2-c]chromene-3-carboxylate (7bg):



White solid; Yield: 360 mg, 88 %; m.p. 222-224 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{19}NO_7$: C, 64.54; H, 4.68; N, 3.42 %. Found: C, 64.55; H, 4.70; N, 3.45 %; IR (KBr) cm^{-1} : 3556, 3434, 3425,

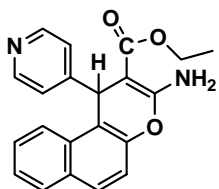
3240, 3179, 3114, 2958, 2837, 1668, 1519, 1409, 1276, 1224, 1137, 1076, 1023, 836, 751,480; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 3.51 (3H, s, OCH₃), 3.67 (3H, s, 2×OCH₃), 4.36 (1H, s, CH), 6.70 (1H, d, J = 7.8 Hz, arom.), 6.80-6.85 (2H, m, arom.), 7.33 (2H, s, NH₂), 7.40-7.47 (2H, m, arom.), 7.66 (1H, t, J = 7.8 Hz, arom.), 7.85 (1H, d, J = 7.5 Hz, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 36.6, 50.6, 55.6, 55.7, 75.6, 104.2, 112.1, 113.1, 117.8, 119.8, 122.6, 124.7, 132.9, 136.0, 148.1, 148.7, 152.2, 153.3, 158.1, 159.7, 162.0, 168.3.

ethyl 3-amino-1,5-dihydro-1-(3,4-dimethoxyphenyl)-5-oxopyrano[2,3-c]chromene-2-carboxylate (7bh):



White solid; Yield: 393 mg, 93 %; m.p. 186-188 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₂₃H₂₁NO₇: C, 65.24; H, 5.00; N, 3.31 %. Found: C, 65.25; H, 5.00; N, 3.34 %; IR (KBr) cm⁻¹: 3419, 3303, 3051, 2981, 2920, 2836, 1699, 1607, 1522, 1459, 1378, 1278, 1190, 1136, 1100, 1028, 755; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 1.11 (3H, t, J = 6.9 Hz, CH₃), 3.63 (3H, s, OCH₃), 3.66 (3H, s, OCH₃), 3.98 (2H, q, J = 6.9 Hz, CH₂), 4.62 (1H, s, CH), 6.64 (1H, dd, J = 8.1 Hz, J = 1.5 Hz, arom.), 6.77 (1H, d, J = 8.4 Hz, arom.), 6.82 (1H, d, J = 1.2 Hz, arom.), 7.38-7.46 (2H, m, arom.), 7.62-7.67 (1H, m, arom.), 7.76 (2H, s, NH₂), 7.93 (1H, d, J = 7.8 Hz, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 14.3, 34.7, 55.5, 59.1, 77.3, 107.1, 111.9, 112.5, 113.2, 116.5, 119.7, 122.5, 124.6, 132.6, 137.7, 147.6, 148.1, 152.1, 153.0, 158.6, 160.0, 167.7.

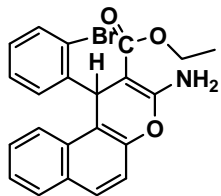
ethyl 3-amino-1-(pyridin-4-yl)-1H-benzo[f]chromene-2-carboxylate (7bi):



Cream colored solid; Yield: 315 mg, 91 %; m.p. 196-198 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₂₁H₁₈N₂O₃: C, 72.82; H, 5.24; N, 8.09 %. Found: C, 72.82; H, 5.25; N, 8.07 %; IR (KBr) cm⁻¹: 3358, 3261, 3054, 2976, 2363, 1940, 1680, 1630, 1596, 1526, 1219, 1070, 1030, 808, 777, 739, 594, 518; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 1.21 (3H, t, J = 6.9 Hz, CH₃), 4.01-4.10 (2H, m, CH₂), 5.48 (1H, s, CH), 7.21 (2H, d, J = 4.8 Hz, arom.), 7.33-7.41 (1H, m, arom.), 7.47 (1H, t, J = 8.1 Hz, arom.), 7.71 (2H, s, NH₂), 7.86-7.90 (2H, m, arom.), 7.95 (2H,

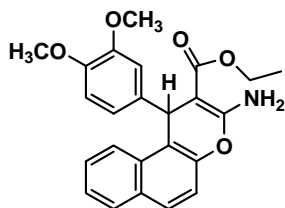
d, $J = 8.4$ Hz, arom.), 8.33 (2H, d, $J = 4.8$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.9, 36.6, 59.5, 76.9, 117.2, 117.7, 123.4, 123.4, 125.4, 127.8, 129.1, 129.9, 130.6, 131.2, 147.4, 150.0, 155.4, 161.1, 168.3.

ethyl 3-amino-1-(2-bromophenyl)-1H-benzof[f]chromene-2-carboxylate (7bj):



White solid; Yield: 382 mg, 90 %; m.p. 152-154 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{18}BrNO_3$: C, 62.28; H, 4.28; N, 3.30 %. Found: C, 62.31; H, 4.29; N, 3.32 %; IR (KBr) cm^{-1} : 3449, 3300, 3021, 2991, 2943, 2837, 1701, 1602, 1520, 1218, 1196, 1136, 1100, 1045, 755; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 1.34 (3H, t, $J = 6.9$ Hz, CH_3), 4.07 (2H, q, $J = 6.9$ Hz, CH_2), 5.75 (1H, s, CH), 7.12 (1H, t, $J = 7.5$ Hz, arom.), 7.20 (1H, d, $J = 7.5$ Hz, arom.), 7.32 (1H, d, $J = 9.0$ Hz, arom.), 7.37-7.49 (2H, m, arom.), 7.77 (2H, s, NH_2), 7.85 (2H, d, $J = 8.7$ Hz, arom.), 8.29 (1H, d, $J = 8.4$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.7, 36.4, 58.8, 77.2, 116.9, 118.6, 121.9, 123.5, 124.9, 127.0, 128.0, 128.1, 128.3, 130.7, 131.6, 132.9, 146.0, 146.9, 160.7, 168.3.

ethyl 3-amino-1-(3,4-dimethoxyphenyl)-1H-benzof[f]chromene-2-carboxylate (7bk):



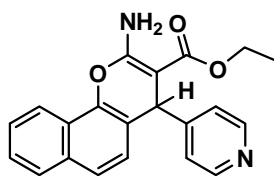
White solid; Yield: 365 mg, 90 %; m.p. 212-214 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{24}H_{23}NO_5$: C, 71.10; H, 5.72; N, 3.45 %. Found: C, 71.14; H, 5.74; N, 3.47 %; IR (KBr) cm^{-1} : 3548, 3440, 3400, 3241, 3176, 3114, 2962, 2836, 1664, 1517, 1409, 1269, 1224, 1135, 1076, 1023, 819, 751, 474; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 1.29 (3H, t, $J = 6.9$ Hz, CH_3), 3.62 (3H, s, OCH_3), 3.69 (3H, s, OCH_3), 4.09-4.15 (2H, m, CH_2), 5.47 (1H, s, CH), 6.62 (1H, dd, $J = 8.4$ Hz, $J = 2.1$ Hz, arom.), 6.72 (1H, d, $J = 8.4$ Hz, arom.), 6.97 (1H, d, $J = 2.1$ Hz, arom.), 7.35-7.58 (5H, m, NH_2 +arom.), 7.88-7.91 (2H, m, arom.), 8.03-8.11 (1H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.4, 35.8, 55.3, 55.4, 58.8, 77.9, 111.8, 111.9, 116.5, 119.0, 119.4, 123.3, 124.7, 126.9, 128.7, 130.2, 130.7, 139.5, 146.6, 146.9, 148.0, 160.3, 168.1.

methyl 2-amino-4-(3-nitrophenyl)-4H-benzo[h]chromene-3-carboxylate (7bl):



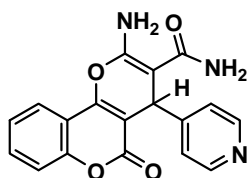
Cream colored solid; Yield: 323 mg, 86 %; m.p. 198-200 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{16}N_2O_5$: C, 67.02; H, 4.28; N, 7.44 %. Found: C, 67.05; H, 4.30; N, 7.46 %; IR (KBr) cm^{-1} : 3447, 3312, 3060, 2946, 1682, 1616, 1525, 1349, 1311, 1265, 1183, 1090, 805, 725; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 3.60 (3H, s, OCH₃), 5.32 (1H, s, CH), 7.42 (1H, d, $J = 8.4$ Hz, arom.), 7.60-7.70 (4H, m, arom.), 7.79 (1H, d, $J = 7.5$ Hz, arom.), 7.93-7.96 (3H, m, arom.), 8.04 (1H, d, $J = 8.1$ Hz, arom.), 8.14 (1H, d, $J = 1.5$ Hz, arom.), 8.38 (1H, d, $J = 8.4$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 39.7, 50.6, 75.5, 119.8, 120.8, 121.3, 121.6, 122.9, 124.1, 126.4, 126.7, 126.8, 127.7, 130.1, 132.8, 134.1, 143.0, 147.9, 149.9, 161.1, 168.3.

ethyl 2-amino-4-(pyridin-4-yl)-4H-benzo[h]chromene-3-carboxylate (7bm):



Cream solid; Yield: 318 mg, 92 %; m.p. 172-174 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{18}N_2O_3$: C, 72.82; H, 5.24; N, 8.09 %. Found: C, 72.87; H, 5.24; N, 8.13 %; IR (KBr) cm^{-1} : 3358, 2974, 1679, 1622, 1539, 1374, 1266, 1185, 1093, 808, 740, 659; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 1.08 (3H, t, $J = 6.9$ Hz, CH₃), 3.98-4.04 (2H, m, CH₂), 5.09 (1H, s, CH), 7.26-7.34 (3H, m, arom.), 7.57-7.65 (3H, m, arom.), 7.89-7.91 (3H, m, NH₂+arom.), 8.31-8.34 (1H, m, arom.), 8.43 (2H, d, $J = 8.4$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 13.7, 39.0, 58.2, 74.4, 118.7, 120.2, 122.1, 122.2, 123.4, 125.7, 126.0, 126.1, 127.1, 132.2, 142.4, 149.0, 155.4, 160.4, 167.4.

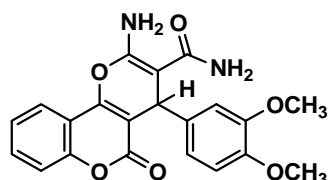
2-amino-4,5-dihydro-5-oxo-4-(pyridin-4-yl)pyrano[3,2-c]chromene-3-carboxamide (7bn):



White solid; Yield: 312 mg, 93 %; m.p. 280-282 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{18}H_{13}N_3O_4$: C, 64.47; H, 3.91; N, 12.53 %. Found: C, 64.50; H, 3.90; N, 12.55 %; IR (KBr) cm^{-1} : 3423, 3069,

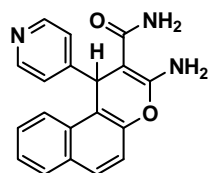
2925, 2361, 1699, 1608, 1541, 1409, 1183, 1052, 759, 481; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 6.24 (1H, s, CH), 7.14-7.26 (6H, m, NH_2 +arom.), 7.46-7.51 (2H, m, arom.), 7.77 (2H, d, $J = 7.8$ Hz, arom.), 8.34 (2H, br s, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 36.3, 102.2, 115.6, 119.7, 122.7, 123.0, 124.2, 131.2, 147.9, 152.6, 153.8, 164.4, 167.9.

2-amino-4,5-dihydro-4-(3,4-dimethoxyphenyl)-5-oxopyrano[3,2-c]chromene-3-carboxamide (7bo):



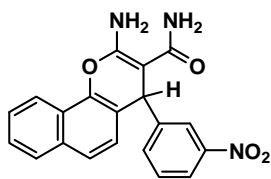
Light yellow colored solid; Yield: 350 mg, 89 %; m.p. 270-272 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $\text{C}_{21}\text{H}_{18}\text{N}_2\text{O}_6$: C, 63.96; H, 4.60; N, 7.10 %. Found: C, 63.99; H, 4.62; N, 7.11 %; IR (KBr) cm^{-1} : 3443, 3068, 2945, 2363, 1702, 16085, 1534, 1409, 1332, 1183, 1108, 1052, 759, 555, 481; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 3.71 (3H, s, OCH_3), 3.83 (3H, s, OCH_3), 6.38 (1H, s, CH), 6.67-6.7 (2H, m, arom.), 7.08-7.30 (5H, m, NH_2 +arom.), 7.51-7.60 (2H, m, arom.), 7.83-7.86 (2H, m, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 35.1, 54.9, 55.1, 103.3, 115.0, 118.6, 120.6, 122.6, 123.4, 127.5, 131.1, 146.2, 147.7, 147.9, 150.1, 150.9, 151.8, 164.0, 166.1.

2-Amino-4-(3-nitro-phenyl)-4H-benzo[h]chromene-3-carboxylic acid amide (7bp):



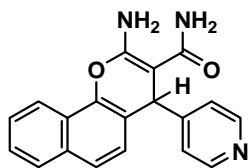
Light brown colored solid; Yield: 318 mg, 88 %; m.p. 290-292 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $\text{C}_{20}\text{H}_{15}\text{N}_3\text{O}_4$: C, 66.48; H, 4.18; N, 11.63 %. Found: C, 66.49; H, 4.17; N, 11.68 %; IR (KBr) cm^{-1} : 3424, 3068, 2930, 2360, 1670, 1608, 1541, 1409, 1277, 1183, 1108, 1044, 762; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 6.32 (1H, s, CH), 7.36 (1H, d, $J = 8.4$ Hz, arom.), 7.48-7.66 (6H, m, NH_2 +arom.), 7.69 (1H, d, $J = 7.2$ Hz, arom.), 7.84-7.86 (3H, m, arom.), 7.96 (1H, d, $J = 7.8$ Hz, arom.), 8.06 (1H, br s, arom.), 8.29 (1H, d, $J = 8.1$ Hz, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 36.1, 115.2, 119.8, 120.8, 121.4, 121.6, 122.5, 124.2, 126.4, 126.6, 126.8, 127.8, 130.1, 132.8, 134.1, 143.1, 147.9, 150.0, 164.6, 168.4.

2-Amino-4-pyridin-4-yl-4H-benzo[h]chromene-3-carboxylic acid amide (7bq):



White solid; Yield: 285 mg, 90 %; m.p. 282-283 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{15}N_3O_2$: C, 71.91; H, 4.76; N, 13.24 %. Found: C, 71.94; H, 4.77; N, 13.26 %; IR (KBr) cm^{-1} : 3424, 3075, 2990, 2355, 1671, 1608, 1540, 1419, 1279, 1183, 1108, 1046, 762; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 6.24 (1H, s, CH), 7.14-7.26 (7H, m, NH_2 +arom.), 7.57 (2H, d, $J = 7.2$ Hz, arom.), 7.68-7.90 (3H, m, arom), 8.34 (2H, br s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 36.0, 116.5, 120.2, 120.8, 122.8, 122.9, 124.2, 125.9, 126.8, 127.1, 127.7, 132.9, 143.1, 150.2, 153.8, 164.1, 168.6.

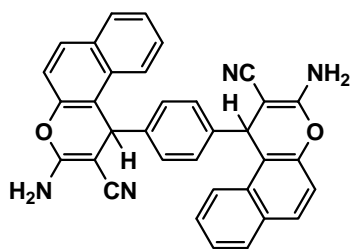
3-Amino-1-pyridin-4-yl-1H-benzof[chromene]-2-carboxylic acid amide (7br):



White solid; Yield: 288 mg, 91 %; m.p. 278-280 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{15}N_3O_2$: C, 71.91; H, 4.76; N, 13.24 %. Found: C, 71.93; H, 4.77; N, 13.25 %; IR (KBr) cm^{-1} : 3424, 3076, 2995, 1675, 1601, 1548, 1419, 1277, 1183, 1108, 1044, 761; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 6.35 (1H, s, CH), 7.12 (2H, s), 7.14 (2H, d, $J = 5.4$ Hz, arom.), 7.31-7.45 (3H, m, arom.), 6.63 (2H, s), 7.73-7.78 (1H, m, arom), 7.88-7.95 (1H, m, arom), 8.43 (2H, d, $J = 5.1$ Hz, arom.), 7.75 (1H, d, $J = 5.4$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 37.4, 114.3, 116.9, 120.3, 122.3, 122.8, 123.7, 125.3, 127.5, 128.7, 130.1, 130.8, 147.1, 150.1, 150.6, 153.8, 165.3, 169.0.

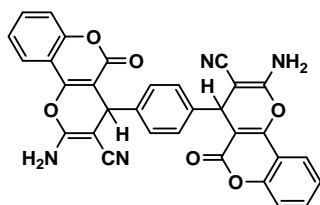
Spectroscopic characterization for 9a-9e

Bis[benzo[f]chromene-2-carbonitrile] (9a):



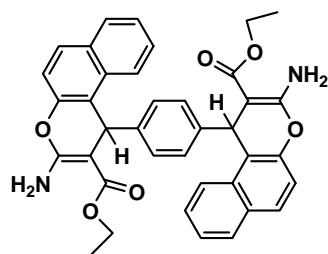
Yellow colored solid; Yield: 461 mg, 89 %; m.p. >340 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₃₄H₂₂N₄O₂: C, 78.75; H, 4.28; N, 10.80 %. Found: C, 78.77; H, 4.28; N, 10.83 %; IR (KBr) cm⁻¹: 3426, 3324, 3200, 2186, 1648, 1587, 1406, 1230, 812, 745; δ_H ppm (300 MHz; DMSO-d₆; TMS) 5.21 (2H, s, 2×CH), 6.91 (4H, d, *J* = 8.7 Hz, arom.), 7.08 (4H, s, 2×NH₂), 7.28-7.40 (6H, m, arom.), 7.77-7.90 (6H, m, arom.); δ_C ppm (75 MHz, DMSO-d₆, TMS) 38.1, 58.6, 116.1, 116.2, 117.3, 121.0, 123.9, 125.5, 127.6, 127.7, 128.9, 129.9, 130.6, 131.4, 144.4, 147.3, 160.4.

Bis[pyrano[2,3-c]chromene-3-carbonitrile] (9b):



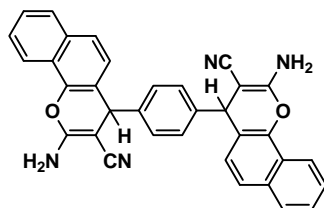
Yellow colored solid; Yield: 510 mg, 92 %; m.p. 271-273 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₃₂H₁₈N₄O₆: C, 69.31; H, 3.27; N, 10.10 %. Found: C, 69.32; H, 3.29; N, 10.14 %; IR (KBr) cm⁻¹: 3384, 3323, 3196, 2195, 1707, 1673, 1602, 1377, 1055, 757, 487; δ_H ppm (300 MHz; DMSO-d₆; TMS) 4.34 (2H, s, 2×CH), 7.11 (4H, s, 2×NH₂), 7.30 (4H, s, arom.), 7.34-7.42 (4H, m, arom.), 7.61 (2H, br s, arom.), 7.80 (2H, d, *J* = 7.5 Hz, arom.); δ_C ppm (75 MHz, DMSO-d₆, TMS) 36.8, 58.2, 104.3, 113.2, 116.8, 119.5, 122.7, 124.9, 127.4, 128.0, 130.3, 133.2, 142.3, 153.7, 158.4.

Bis[benzo[f]chromene-2-carboxylate] (9c):



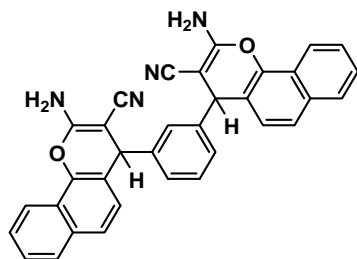
White solid; Yield: 557 mg, 91 %; m.p. 268-270 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{38}H_{32}N_2O_6$: C, 74.49; H, 5.26; N, 4.57 %. Found: C, 74.53; H, 5.28; N, 4.57 %; IR (KBr) cm^{-1} : 3358, 3261, 3054, 2976, 2363, 1940, 1680, 1630, 1596, 1526, 1219, 1070, 1030, 808, 777, 739, 594, 518; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.86 (6H, br s, $2 \times CH_3$), 3.76 (4H, br s, $2 \times CH_2$), 5.15 (2H, s, $2 \times CH$), 6.83 (4H, br s, $2 \times NH_2$), 7.07-7.33 (10H, m, arom.), 7.58-7.67 (6H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.3, 36.0, 58.8, 77.9, 116.6, 118.8, 123.0, 124.7, 127.0, 127.4, 128.5, 128.9, 130.3, 130.7, 144.4, 146.8, 160.6, 168.0.

Bis[benzo[h]chromene-3-carbonitrile] (9d):



Light brown colored solid; Yield: 466 mg, 90 %; m.p. 278-280 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{34}H_{22}N_4O_2$: C, 78.75; H, 4.28; N, 10.80 %. Found: C, 78.78; H, 4.28; N, 10.82 %; IR (KBr) cm^{-1} : 3440, 3321, 3195, 3056, 2193, 1655, 1633, 1599, 1573, 1409, 1375, 1260, 1100, 794, 760, 561; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 4.86 (2H, s, $2 \times CH$), 7.14-7.19 (10H, m, $2 \times NH_2 + arom.$), 7.57 (6H, br s, arom.), 7.86 (2H, br s, arom.), 8.25 (2H, br s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 40.0, 55.6, 117.3, 117.3, 119.9, 120.1, 122.2, 123.4, 125.6, 126.0, 126.1, 127.1, 127.3, 133.1, 142.2, 143.7, 159.6.

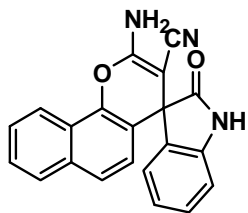
Bis[benzo[h]chromene-3-carbonitrile] (9e):



Brown colored solid; Yield: 461 mg, 89 %; m.p. 268-270 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{34}H_{22}N_4O_2$: C, 78.75; H, 4.28; N, 10.80 %. Found: C, 78.76; H, 4.28; N, 10.83%; IR (KBr) cm^{-1} : 3455, 3318, 3197, 3054, 2183, 1658, 1589, 1400, 1225, 1179, 1078, 1032, 806, 737, 511; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 5.22 (2H, s, 2×CH), 6.74-7.43 (14H,m, 2×NH₂+arom.), 7.62 (2H, d, J = 8.7 Hz, arom.), 7.83-7.88 (4H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 37.4, 37.8, 57.0, 57.2, 114.7, 115.4, 116.0, 116.1, 119.8, 122.8, 123.1, 124.1, 124.6, 124.7, 125.8, 126.1, 126.4, 127.7, 128.7, 128.9, 129.6, 129.3, 129.6, 130.1, 130.2, 145.2, 145.2, 145.9, 146.0, 158.8, 159.4.

Spectroscopic characterization for 11

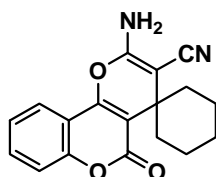
2-Amino spiro[(4H)-benzo(f)chromene-4,3'-(3'H)-indol]-2'-one-3-carbonitrile (11):



Brown solid; Yield: 302 mg, 89 %; m.p. 266-268 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{13}N_3O_2$: C, 74.33; H, 3.86; N, 12.38 %. Found: C, 74.36; H, 3.87; N, 12.39 %; IR (KBr) cm^{-1} : 3450, 2928, 2904, 2865, 2845, 2360, 1703, 1689, 1613, 1591, 1557, 1463, 1393, 1333, 1295; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 6.58-6.01 (1H, m, arom.), 7.03 (2H, t, J =6.0 Hz, arom.), 7.12 (1H, d, J =6.9 Hz, arom.), 7.32 (1H, t, J =7.2 Hz, arom.), 7.49 (2H, s, NH₂), 7.58-7.73 (3H, m, arom.), 7.92 (1H, d, J =7.8 Hz, arom.), 8.31 (1H, d, J =8.1 Hz, arom.), 10.71 (1H, s, NH); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 51.0, 54.5, 110.1, 114.9, 118.5, 120.8, 122.8, 122.8, 123.2, 124.5, 125.0, 127.0, 127.3, 127.7, 129.2, 133.1, 134.7, 141.9, 143.7, 161.1, 178.8.

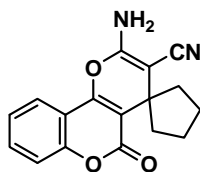
Spectroscopic characterization for 13a-13d

Spiro(2-amino-3-cyano pyrano[3,2-c]chromene-4,1'-cyclohexane) (13a):



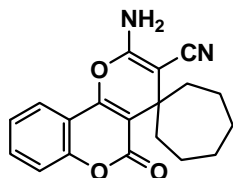
White solid; Yield: 286 mg, 93 %; m.p. 231-233 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{18}H_{16}N_2O_3$: C, 70.12; H, 5.23; N, 9.09 %. Found: C, 70.16; H, 5.24; N, 9.09 %; IR (KBr) cm^{-1} : 3597, 2936, 2360, 1728, 1678, 1649, 1545, 1301, 1084; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 1.13-1.26 (1H, m, CH), 1.52-1.69 (5H, m, CH₂), 1.94-2.04 (2H, s, CH₂), 2.49-2.57 (2H, s, CH₂), 7.04 (2H, s, NH₂), 7.38-7.42 (2H, m, arom.), 7.64 (1H, t, $J=7.2$ Hz, arom.), 7.83 (1H, d, $J=7.2$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 21.8, 24.5, 34.8, 35.6, 61.5, 109.1, 113.4, 116.6, 122.8, 123.3, 124.9, 133.3, 152.4, 153.9, 158.6, 159.3.

Spiro(2-amino-3-cyano pyrano[3,2-c]chromene-4,1'-cyclopentane) (13b):



White solid; Yield: 268 mg, 91 %; m.p. 230-232 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{17}H_{14}N_2O_3$: C, 69.38; H, 4.79; N, 9.52 %. Found: C, 69.40; H, 4.80; N, 9.51 %; IR (KBr) cm^{-1} : 3450, 2928, 2904, 2865, 2845, 2360, 1724, 1602, 1558, 1313; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 1.92-2.10 (6H, m, CH), 2.53-2.57 (2H, m, CH₂), 7.03 (2H, s, NH₂), 7.37-7.42 (2H, m, arom.), 7.65 (1H, t, $J=4.8$ Hz, arom.), 7.84 (1H, d, $J=10.2$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 26.3, 41.2, 43.0, 70.9, 108.0, 113.0, 116.4, 119.2, 122.2, 124.4, 131.4, 151.9, 152.2, 154.5, 159.6.

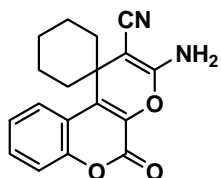
Spiro(2-amino-3-cyano pyrano[3,2-c]chromene-4,1'-cycloheptane) (13c):



White solid; Yield: 296 mg, 92 %; m.p. 225-226 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{18}N_2O_3$: C, 70.79; H, 5.63; N, 8.69 %. Found: C, 70.81; H, 5.65; N, 8.73 %; IR (KBr) cm^{-1} : 3464, 2930, 2868,

2354, 1720, 1655, 1540, 1078; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 1.43-1.46 (4H, m, CH), 1.71-1.78 (4H, m, CH₂), 2.04-2.08 (2H, s, CH₂), 2.43-2.50 (2H, s, CH₂), 7.02 (2H, s, NH₂), 7.37-7.41 (2H, m, arom.), 7.63-7.66 (1H, m, arom.), 7.85 (1H, d, $J=10.2$ Hz, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 26.5, 27.0, 29.7, 30.8, 33.7, 70.5, 108.1, 113.4, 117.0, 122.8, 123.3, 124.9, 131.2, 152.5, 153.9, 155.6, 159.4.

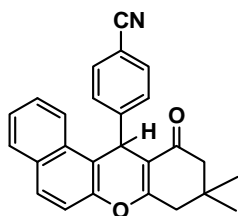
Spiro(3-amino-2-cyano pyrano[2,3-c]chromene-1,1'-cyclohexane) (13d):



White solid; Yield: 280 mg, 91 %; m.p. 231-233 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₁₈H₁₆N₂O₃: C, 70.12; H, 5.23; N, 9.09 %. Found: C, 70.15; H, 5.24; N, 9.10 %; IR (KBr) cm⁻¹: 3585, 2933, 2345, 1717, 1678, 1654, 1542, 1300, 1084; δ_{H} ppm (300 MHz; DMSO- d_6 ; TMS) 1.23-1.26 (1H, m, CH), 1.52-1.69 (5H, m, CH₂), 1.94-2.04 (2H, s, CH₂), 2.59-2.67 (2H, s, CH₂), 7.12 (2H, s, NH₂), 7.20 (1H, t, $J=6.9$ Hz, arom.), 7.36 (1H, d, $J=7.8$ Hz, arom.), 7.44 (1H, t, $J=7.8$ Hz, arom.), 7.53 (1H, d, $J=7.8$ Hz, arom.); δ_{C} ppm (75 MHz, DMSO- d_6 , TMS) 20.9, 26.3, 34.0, 37.2, 70.8, 108.0, 113.0, 116.5, 119.2, 122.2, 124.4, 133.5, 151.9, 152.3, 155.5, 159.7.

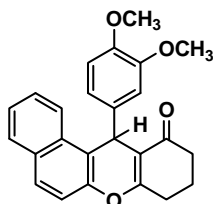
Spectroscopic characterization for 4a-4h

4-(9,10,11,12-tetrahydro-9,9-dimethyl-11-oxo-8H-benzo[a]xanthen-12-yl)benzonitrile (4a):



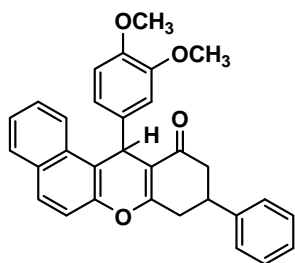
White solid; Yield: 352 mg, 93 %; m.p. 203-205 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{26}H_{21}NO_2$: C, 82.30; H, 5.58; N, 3.69 %. Found: C, 82.35; H, 5.60; N, 3.72 %; IR (KBr) cm^{-1} : 3064, 2951, 2226, 1648, 1505, 1366, 1226, 1018, 809, 746, 654, 556, 482; δ_H ppm (300 MHz; $CDCl_3$; TMS) 0.97 (3H, s, CH_3), 1.52 (3H, s, CH_3), 2.21-2.35 (2H, m, CH_2), 2.58 (2H, s, CH_2), 5.76 (1H, s, CH), 7.33-7.40 (3H, m, arom.), 7.42-7.49 (4H, m, arom.), 7.79-7.84 (3H, m, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 27.1, 29.3, 32.3, 35.0, 41.4, 50.8, 110.2, 113.2, 116.2, 117.1, 118.9, 123.2, 125.2, 127.4, 128.7, 129.3, 129.6, 131.1, 131.6, 132.2, 147.9, 149.9, 164.6, 196.8.

9,10-dihydro-12-(3,4-dimethoxyphenyl)-8H-benzo[a]xanthen-11(12H)-one (4b):



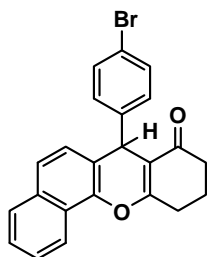
White solid; Yield: 347 mg, 90 %; m.p. 250-252 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{25}H_{22}O_4$: C, 77.70; H, 5.74 %. Found: C, 77.72; H, 5.76 %; IR (KBr) cm^{-1} : 2929, 2837, 1646, 1594, 1514, 1373, 1226, 1138, 1031, 833, 755; δ_H ppm (300 MHz; $CDCl_3$; TMS) 1.87-1.91 (2H, m, CH_2), 2.25-2.33 (2H, m, CH_2), 2.56-2.58 (2H, m, CH_2), 3.61 (3H, s, OCH_3), 3.71 (3H, s, OCH_3), 5.60 (1H, s, CH), 6.51 (1H, d, $J = 8.1$ Hz, arom.), 6.59 (1H, dd, $J = 8.25$ Hz, $J = 2.1$ Hz, arom.), 6.93 (1H, s, arom.), 7.20-7.30 (3H, m, arom.), 7.62-7.66 (2H, m, arom.), 7.84 (1H, d, $J = 7.8$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 20.3, 27.7, 34.1, 37.1, 55.7, 55.9, 111.0, 112.2, 115.7, 116.9, 117.7, 120.4, 123.7, 124.9, 127.0, 128.4, 128.8, 131.5, 131.5, 137.9, 147.4, 147.9, 148.6, 165.5, 197.2.

9,10-dihydro-12-(3,4-dimethoxyphenyl)-9-phenyl-8H-benzo[a]xanthen-11(12H)-one (4c):



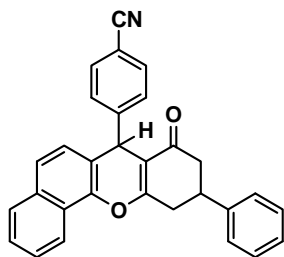
White solid; Yield: 434 mg, 94 %; m.p. 192-194 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{31}H_{26}O_4$: C, 80.50; H, 5.67 %. Found: C, 80.52; H, 5.67 %; IR (KBr) cm^{-1} : 3548, 3441, 3176, 2963, 1663, 1518, 1459, 1409, 1224, 1136, 1076, 1023, 819, 751, 475; δ_H ppm (300 MHz; $CDCl_3$; TMS) 2.65-2.75 (2H, m, CH_2), 2.92-2.96 (2H, m, CH_2), 3.30-3.45 (1H, m, CH), 3.78 (3H, s, OCH_3), 3.89 (3H, s, OCH_3), 5.81 (1H, s, CH), 6.69 (1H, d, $J = 6.9$ Hz, arom.), 6.77-6.80 (1H, m, arom.), 7.15 (1H, d, $J = 1.8$ Hz, arom.), 7.25-7.32 (3H, m, arom.), 7.36-7.47 (5H, m, arom.), 7.80-7.84 (2H, m, arom.), 8.01 (1H, d, $J = 7.2$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 34.2, 35.1, 38.0, 44.1, 55.6, 55.8, 111.1, 112.2, 115.4, 116.8, 117.6, 120.3, 123.6, 124.9, 126.6, 126.7, 126.9, 127.0, 128.3, 128.6, 128.7, 128.8, 131.4, 131.5, 137.7, 142.3, 147.4, 147.8, 148.6, 164.7, 196.0.

7-(4-bromophenyl)-10,11-dihydro-7H-benzo[c]xanthen-8(9H)-one (4d):



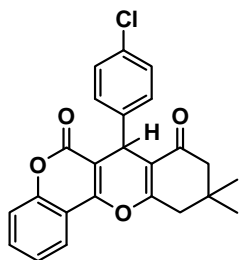
White solid; Yield: 368 mg, 91 %; m.p. 272-274 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{23}H_{17}BrO_2$: C, 68.16; H, 4.23 %. Found: C, 68.18; H, 4.23 %; IR (KBr) cm^{-1} : 2931, 2830, 1656, 1604, 1504, 1393, 1229, 1135, 1021, 823, 748; δ_H ppm (300 MHz; $CDCl_3$; TMS) 1.96-2.08 (2H, m, CH_2), 2.33-2.39 (2H, m, CH_2), 2.66-2.86 (2H, m, CH_2), 5.07 (1H, s, CH), 7.01 (1H, d, $J = 8.7$ Hz, arom.), 7.09 (2H, dd, $J = 8.4$ Hz, $J = 1.7$ Hz, arom.), 7.26-7.28 (2H, m, arom.), 7.43-7.54 (3H, m, arom.), 7.72 (1H, d, $J = 8.1$ Hz, arom.), 8.21 (1H, d, $J = 8.4$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 20.4, 27.8, 37.0, 37.8, 114.5, 116.7, 119.1, 120.4, 121.0, 123.6, 124.7, 126.5, 126.7, 127.7, 129.3, 130.1, 131.4, 133.2, 144.6, 144.9, 166.1, 197.1.

4-(8,9,10,11-tetrahydro-8-oxo-10-phenyl-7H-benzo[c]xanthen-7-yl)benzotrile (4e):



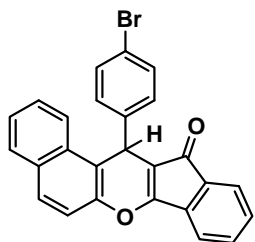
White solid; Yield: 397 mg, 93 %; m.p. 212-214 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{30}H_{21}NO_2$: C, 84.29; H, 4.95; N, 3.28 %. Found: C, 84.33; H, 4.96; N, 3.31 %; IR (KBr) cm^{-1} : 3548, 3441, 3176, 2963, 1663, 1518, 1459, 1409, 1224, 1136, 1076, 1023, 819, 751, 475; δ_H ppm (300 MHz; $CDCl_3$; TMS) 2.54-2.65 (2H, m, CH_2), 2.93-3.06 (2H, m, CH_2), 3.34-3.38 (1H, m, CH), 5.20 (1H, s, CH), 7.01 (1H, d, $J = 8.4$ Hz, arom.), 7.19-7.25 (2H, m, arom.), 7.29-7.35 (2H, m, arom.), 7.38-7.40 (1H, m, arom.), 7.41-7.42 (1H, m, arom.), 7.45-7.50 (4H, m, arom.), 7.52-7.54 (1H, m, arom.), 7.74 (2H, d, $J = 7.5$ Hz, arom.), 8.18 (1H, d, $J = 9.0$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 35.4, 38.4, 38.6, 44.0, 110.5, 113.8, 118.2, 121.0, 123.6, 125.2, 126.3, 126.4, 126.7, 126.7, 126.8, 126.9, 127.2, 127.3, 127.6, 127.8, 128.8, 128.9, 132.2, 132.4, 134.8, 142.0, 144.3, 150.8, 165.8, 196.2.

7-(4-Chloro-phenyl)-10,10-dimethyl-7,9,10,11-tetrahydro-chromeno[4,3-b]chromene-6,8-dione (4f):



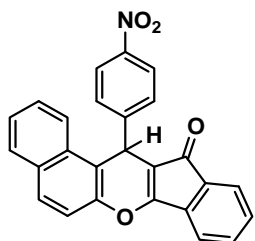
White solid; Yield: 377 mg, 93 %; m.p. 252-254 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{24}H_{19}ClO_4$: C, 70.85; H, 4.71 %. Found: C, 70.85; H, 4.71 %; IR (KBr) cm^{-1} : 2962, 2870, 1732, 1667, 1622, 1591, 1491, 1362, 1189, 1057, 840, 763, 537; δ_H ppm (300 MHz; $CDCl_3$; TMS) 1.04 (3H, s, CH_3), 1.13 (3H, s, CH_3), 2.18-2.32 (2H, m, CH_2), 2.58-2.71 (2H, m, CH_2), 4.88 (1H, s, CH), 7.16-7.18 (2H, m, arom.), 7.23-7.35 (4H, m, arom.), 7.51-7.56 (1H, m, arom.), 7.83 (1H, d, $J = 7.8$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 27.5, 29.1, 32.3, 33.0, 40.8, 50.6, 106.3, 113.5, 114.8, 116.9, 122.4, 124.3, 128.4, 130.0, 132.4, 132.8, 141.1, 152.6, 154.0, 154.0, 160.5, 162.1, 195.9.

13-(4-Bromo-phenyl)-13H-7-oxa-indeno[1,2-b]phenanthren-12-one (4g):



White solid; Yield: 395 mg, 90 %; m.p. 253-255 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{26}H_{15}BrO_2$: C, 71.09; H, 3.44 %. Found: C, 71.12; H, 3.45 %; IR (KBr) cm^{-1} : 3032, 2948, 2199, 1667, 1505, 1338, 1226, 1019, 809, 752, 675, 556, 483; δ_H ppm (300 MHz; $CDCl_3$; TMS) 5.07 (1H, s, CH), 7.23-7.27 (2H, m, arom.), 7.31-7.48 (5H, m, arom.), 7.55-7.65 (2H, m, arom.), 7.78-7.84 (3H, m, arom.), 8.01 (1H, s, arom.), 8.29-8.32 (1H, m, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 116.6, 117.0, 118.9, 120.2, 121.5, 122.3, 124.4, 125.2, 125.8, 126.0, 128.1, 129.6, 129.8, 130.1, 130.4, 130.5, 131.0, 132.6, 133.2, 148.6, 166.1, 197.0.

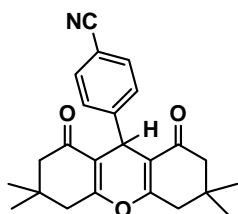
13-(4-Nitro-phenyl)-13H-7-oxa-indeno[1,2-b]phenanthren-12-one (4h):



White solid; Yield: 365 mg, 90 %; m.p. 265-267 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{26}H_{15}NO_4$: C, 77.03; H, 3.73; N, 3.46 %. Found: C, 77.04; H, 3.75; N, 3.47 %; IR (KBr) cm^{-1} : 3064, 2915, 2226, 1651, 1595, 1515, 1341, 1219, 1074, 844, 741, 591; δ_H ppm (300 MHz; $CDCl_3$; TMS) 5.63 (1H, s, CH), 7.41 (1H, d, $J = 8.7$ Hz, arom.), 7.79 (1H, d, $J = 7.5$ Hz, arom.), 7.90 (1H, d, $J = 7.5$ Hz, arom.), 7.98 (2H, s, arom.), 8.07 (1H, d, $J = 8.4$ Hz, arom.), 8.16 (1H, d, $J = 9.0$ Hz, arom.), 8.22-8.34 (3H, m, arom.), 8.49-8.62 (4H, m, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 37.1, 60.0, 116.6, 119.2, 122.1, 122.3, 123.6, 124.0, 124.3, 127.4, 128.6, 129.7, 129.9, 130.5, 131.5, 132.2, 133.6, 136.4, 146.5, 150.2, 150.7, 166.7, 199.7.

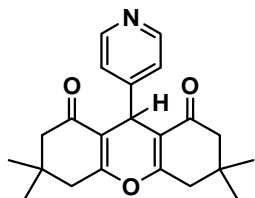
Spectroscopic characterization for 14a-14j

4-(2,3,4,5,6,7,8,9-octahydro-3,3,6,6-tetramethyl-1,8-dioxo-1H-xanthen-9-yl)benzonitrile (14a):



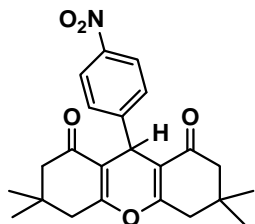
White solid; Yield: 352 mg, 94 %; m.p. 215-217 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{24}H_{25}NO_3$: C, 76.77; H, 6.71; N, 3.73 %. Found: C, 76.78; H, 6.71; N, 3.75 %; IR (KBr) cm^{-1} : 3070, 2950, 2900, 2250, 1657, 1619, 1500, 1417, 1200, 1173, 1125; δ_H ppm (300 MHz; $CDCl_3$; TMS) 0.96 (6H, s, $2 \times CH_3$), 1.09 (6H, s, $2 \times CH_3$), 2.11-2.25 (4H, m, $2 \times CH_2$), 2.45 (4H, s, $2 \times CH_2$), 4.75 (1H, s, CH), 7.39 (2H, d, $J = 8.4$ Hz, arom.), 7.50 (2H, d, $J = 8.4$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 27.3, 29.2, 32.2, 32.5, 40.9, 50.6, 110.2, 114.7, 119.0, 129.3, 132.0, 149.5, 162.9, 196.3.

3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-(pyridin-4-yl)-2H-xanthene-1,8(5H,9H)-dione (14b):



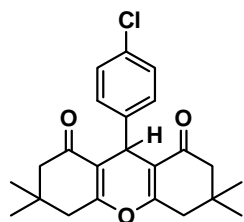
White solid; Yield: 323 mg, 92 %; m.p. 212-214 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{25}NO_3$: C, 75.19; H, 7.17; N, 3.99 %. Found: C, 75.21; H, 7.17; N, 4.00 %; IR (KBr) cm^{-1} : 3250, 2955, 2931, 1590, 1395, 1550, 1158, 1045, 841, 774, 695; δ_H ppm (300 MHz; $CDCl_3$; TMS) 0.98 (6H, s, $2 \times CH_3$), 1.10 (6H, s, $2 \times CH_3$), 2.13-2.27 (4H, m, $2 \times CH_2$), 2.47 (4H, s, $2 \times CH_2$), 4.72 (1H, s, CH), 7.21-7.23 (2H, m, arom.), 8.43-8.47 (2H, m, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 27.3, 29.2, 32.2, 32.7, 40.8, 50.6, 114.3, 114.4, 122.2, 123.6, 146.5, 152.5, 162.9, 196.0.

3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-(4-nitrophenyl)-2H-xanthene-1,8(5H,9H)-dione (14c):



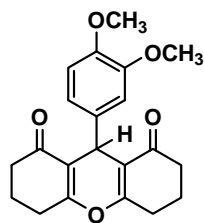
White solid; Yield: 367 mg, 93 %; m.p. 220-221 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{23}H_{25}NO_5$: C, 69.86; H, 6.37; N, 3.54 %. Found: C, 69.89; H, 6.38; N, 3.57 %; IR (KBr) cm^{-1} : 3200, 2960, 2928, 1592, 1390, 1550, 1159, 1040, 842, 774, 690; δ_H ppm (300 MHz; $CDCl_3$; TMS) 0.96 (6H, s, $2 \times CH_3$), 1.08 (6H, s, $2 \times CH_3$), 2.10-2.25 (4H, m, $2 \times CH_2$), 2.46 (4H, s, $2 \times CH_2$), 4.79 (1H, s, CH), 7.44 (2H, d, $J = 8.7$ Hz, arom.), 8.06 (2H, d, $J = 8.7$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 27.2, 29.2, 32.2, 32.3, 40.8, 50.6, 114.5, 123.4, 129.3, 146.5, 151.5, 162.9, 196.2.

9-(4-chlorophenyl)-3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-2H-xanthene-1,8(5H,9H)-dione (14d):



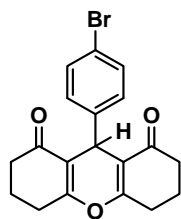
White solid; Yield: 357 mg, 93 %; m.p. 226-228 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{23}H_{25}ClO_5$: C, 71.77; H, 6.55 %. Found: C, 71.79; H, 6.55 %; IR (KBr) cm^{-1} 2960, 2929, 1589, 1305, 1093, 887, 833, 720, 658; δ_H ppm (300 MHz; $CDCl_3$; TMS) 0.91 (6H, s, $2 \times CH_3$), 1.02 (6H, s, $2 \times CH_3$), 2.11-2.25 (4H, m, $2 \times CH_2$), 2.45 (4H, s, $2 \times CH_2$), 4.63 (1H, s, CH), 7.16 (2H, d, $J = 8.7$ Hz, arom.), 7.21 (2H, d, $J = 9.0$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 27.2, 29.1, 32.1, 32.3, 40.8, 50.5, 114.0, 129.0, 131.0, 131.9, 145.0, 162.9, 196.1.

3,4,6,7-tetrahydro-9-(3,4-dimethoxyphenyl)-2H-xanthene-1,8(5H,9H)-dione (14e):



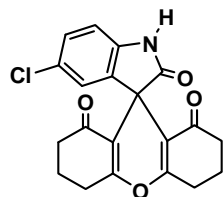
White solid; Yield: 315 mg, 89 %; m.p. 260-262 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{22}O_5$: C, 71.17; H, 6.26 %. Found: C, 71.17; H, 6.26 %; IR (KBr) cm^{-1} : 2977, 2967, 2926, 1659, 1620, 1517, 1438, 1358, 1172, 1022, 907, 861, 635; δ_H ppm (300 MHz; $CDCl_3$; TMS) 1.89-1.99 (4H, m, $2 \times CH_2$), 2.24-2.31 (4H, m, $2 \times CH_2$), 2.49-2.61 (4H, m, $2 \times CH_2$), 3.72 (3H, s, OCH_3), 3.81 (3H, s, OCH_3), 4.69 (1H, s, CH), 6.63 (2H, br s, arom.), 6.93 (1H, s, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 20.2, 27.0, 30.8, 36.9, 55.7, 55.8, 110.9, 112.7, 116.8, 119.5, 137.2, 147.5, 148.4, 163.7, 196.5.

9-(4-bromophenyl)-3,4,6,7-tetrahydro-2H-xanthene-1,8(5H,9H)-dione (14f):



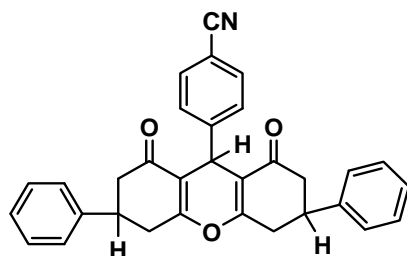
White solid; Yield: 335 mg, 90 %; m.p. 284-286 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{19}H_{17}BrO_3$: C, 61.14; H, 4.59 %. Found: C, 61.19; H, 4.61 %; IR (KBr) cm^{-1} : 3100, 2980, 2878, 1589, 1490, 1303, 1158, 890, 709, 660; δ_H ppm (300 MHz; $CDCl_3$; TMS) 1.99-2.09 (4H, m, $2 \times CH_2$), 2.23-2.40 (4H, m, $2 \times CH_2$), 2.50-2.69 (4H, m, $2 \times CH_2$), 4.74 (1H, s, CH), 7.16 (2H, d, $J = 7.5$ Hz, arom.), 7.31 (2H, d, $J = 7.5$ Hz, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 20.2, 27.1, 31.3, 36.8, 116.3, 120.2, 130.1, 131.1, 143.4, 164.0, 196.4.

9-(5-Chloro-2-oxo-2,3-dihydro-1H-indol-3-yl)-3,4,5,6,7,9-hexahydro-2H-xanthene-1,8-dione (14g):



Brown colored solid; Yield: 332 mg, 90 %; m.p. >320 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{20}H_{16}ClNO_4$: C, 64.96; H, 4.36; N, 3.79 %. Found: C, 64.99; H, 4.36; N, 3.78 %; IR (KBr) cm^{-1} : 2977, 2967, 2926, 1703, 1689, 1659, 1613, 1591, 1463, 1393, 1295, 1221, 1022, 907, 861, 635; δ_H ppm (300 MHz; $DMSO-d_6$; TMS) 1.77-1.83 (4H, m, $2 \times CH_2$), 2.06-2.15 (4H, m, $2 \times CH_2$), 2.55-2.59 (4H, m, $2 \times CH_2$), 6.56 (1H, d, $J = 8.4$ Hz, arom.), 6.98 (1H, s, arom.), 7.13 (1H, dd, $J = 8.1$ Hz, $J = 1.8$ Hz, arom.); δ_C ppm (75 MHz, $DMSO-d_6$, TMS) 19.7, 27.0, 37.0, 110.1, 112.3, 113.6, 125.5, 130.4, 136.7, 143.4, 165.6, 178.1, 195.4.

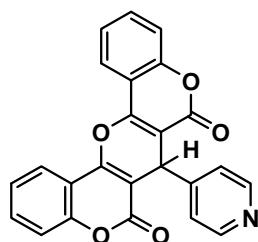
4-(2,3,4,5,6,7,8,9-octahydro-1,8-dioxo-3,6-diphenyl-1H-xanthen-9-yl)benzonitrile (14h):



White solid; Yield: 438 mg, 93 %; m.p. 162-164 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{32}H_{25}NO_3$: C, 81.51; H, 5.34; N, 2.97 %. Found: C, 81.55; H, 5.36; N, 2.96 %; IR (KBr) cm^{-1} : 3073, 2960, 2950,

2230, 1650, 1621, 1502, 1419, 1200, 1173, 1135; δ_{H} ppm (300 MHz; CDCl_3 ; TMS) 2.58-2.96 (8H, m, $4 \times \text{CH}_2$), 3.29-3.40 (2H, s, $2 \times \text{CH}$), 4.91 (1H, s, CH), 7.13-7.60 (14H, s, arom.); δ_{C} ppm (75 MHz, CDCl_3 , TMS) 32.5, 34.5, 34.7, 38.2, 38.3, 40.3, 43.3, 43.9, 110.3, 115.5, 115.7, 115.8, 126.6, 126.7, 127.3, 127.4, 127.5, 128.8, 128.9, 129.3, 129.4, 132.0, 132.1, 141.7, 149.0, 163.1, 163.8, 163.9, 195.4.

7-Pyridin-4-yl-7H-pyrano[3,2-c;5,6-c']dichromene-6,8-dione (14i):

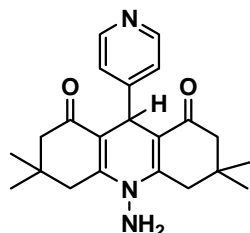


Cream colored solid; Yield: 363 mg, 92 %; m.p. 250-252 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $\text{C}_{24}\text{H}_{13}\text{NO}_5$: C, 72.91; H, 3.31 %. Found: C, 72.93; H, 3.32 %; IR (KBr) cm^{-1} : 1730, 1718, 1666, 1609, 1456, 1365, 1336, 1178, 1062, 1042, 888, 766, 713; δ_{H} ppm (300 MHz; DMSO-d_6 ; TMS) 6.39 (1H, s, CH), 7.19-7.29 (4H, m, arom.), 7.51 (2H, t, $J = 7.5$ Hz, arom.), 7.71 (2H, d, $J = 5.4$ Hz, arom.), 7.77 (2H, d, $J = 7.8$ Hz, arom.), 7.59-7.61 (2H, m, arom.); δ_{C} ppm (75 MHz, DMSO-d_6 , TMS) 38.1, 102.0, 116.2, 119.9, 123.7, 124.7, 125.4, 132.0, 142.3, 151.0, 153.2, 163.9, 164.6, 168.6.

Spectroscopic characterization for 16a-16h

***N*-Amino-9-(pyridin-4-yl)-3,4,6,7-tetrahydro-3,3,6,6-tetramethylacridine-1,8(2H,5H,9H,10H)-dione**

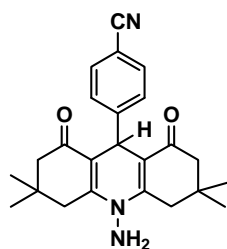
(16a):



Yellow solid; Yield: 328 mg, 90 %; m.p. 288-290 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{27}N_3O_2$: C, 72.30; H, 7.45; N, 11.50 %. Found: C, 72.35; H, 7.46; N, 11.51 %; IR (KBr) cm^{-1} : 3350, 2954, 1626, 1561, 1487, 1219, 1009, 882, 769; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.62 (6H, s, $2 \times CH_3$), 0.78 (6H, s, $2 \times CH_3$), 1.79 (2H, d, $J = 15.9$ Hz, CH_2), 1.95 (2H, d, $J = 15.6$ Hz, CH_2), 2.31 (2H, d, $J = 19.2$ Hz, CH_2), 2.66 (2H, d, $J = 18.0$ Hz, CH_2), 4.70 (1H, s, CH), 5.09 (2H, s, NH_2), 6.86-6.88 (2H, m, arom.), 8.09-8.13 (2H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 26.9, 29.5, 31.6, 31.7, 33.0, 49.6, 109.8, 113.4, 122.7, 149.2, 154.5, 155.5, 157.1, 194.7.

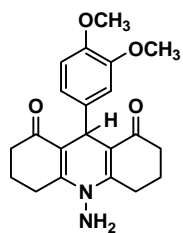
***N*-Amino-3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-p-benzonitrileacridine-1,8(2H,5H,9H,10H)-dione**

(16b):



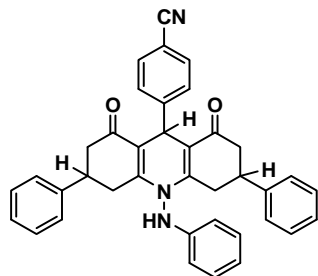
Bright yellow solid; Yield: 366 mg, 94 %; m.p. >300 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{24}H_{27}N_3O_2$: C, 74.01; H, 6.99; N, 10.79 %. Found: C, 74.02; H, 7.02; N, 10.80 %; IR (KBr) cm^{-1} : 3337, 2958, 2195, 1630, 1488, 1319, 1143, 979, 886; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.76 (6H, s, $2 \times CH_3$), 0.94 (6H, s, $2 \times CH_3$), 1.92 (2H, d, $J = 15.9$ Hz, CH_2), 2.09 (2H, d, $J = 15.9$ Hz, CH_2), 2.46 (2H, d, $J = 20.25$ Hz, CH_2), 2.81 (2H, d, $J = 18.0$ Hz, CH_2), 4.91 (1H, s, CH), 5.24 (2H, s, NH_2), 7.25 (2H, d, $J = 8.1$ Hz, arom.), 7.57 (2H, d, $J = 8.1$ Hz, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 26.8, 29.5, 31.6, 31.6, 31.7, 32.6, 49.5, 108.5, 110.2, 110.3, 119.7, 128.5, 131.9, 151.9, 155.3, 155.3, 194.7.

10-Amino-9-(3,4-dimethoxy-phenyl)-3,4,6,7,9,10-hexahydro-2H,5H-acridine-1,8-dione (16c):



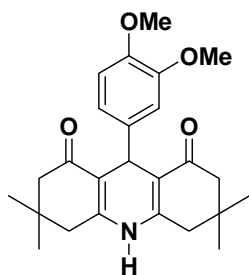
Yellow solid; Yield: 342 mg, 93 %; m.p. 280-282 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{24}N_2O_4$: C, 68.46; H, 6.57; N, 7.60 %. Found: C, 68.49; H, 6.58; N, 7.63 %; IR (KBr) cm^{-1} : 3339, 3055, 2958, 1626, 1566, 1321, 1126, 975, 883; δ_H ppm (300 MHz; $CDCl_3$; TMS) 1.92-2.00 (4H, m, $2 \times CH_2$), 2.25-2.44 (4H, m, $2 \times CH_2$), 2.65-2.71 (4H, m, $2 \times CH_2$), 3.72 (3H, s, OCH_3), 3.81 (3H, s, OCH_3), 5.25 (2H, s, NH_2), 5.46 (1H, s, CH), 6.63 (2H, s, arom.), 6.93 (1H, s, arom.); δ_C ppm (75 MHz, $CDCl_3$, TMS) 21.2, 30.0, 31.4, 36.5, 55.1, 55.3, 109.9, 114.8, 115.8, 119.5, 137.1, 152.4, 154.6, 157.8, 194.5.

4-(1,8-Dioxo-3,6-diphenyl-10-phenylamino-1,2,3,4,5,6,7,8,9,10-decahydro-acridin-9-yl)-benzonitrile (16d):



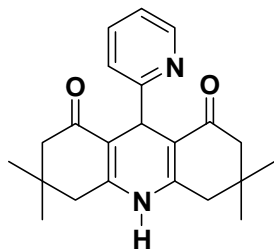
Yellow colored solid; Yield: 527 mg, 94 %; m.p. 145-147 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{38}H_{31}N_3O_2$: C, 81.26; H, 5.56; N, 7.48 %. Found: C, 81.29; H, 5.57; N, 7.48 %; IR (KBr) cm^{-1} : 3343, 2960, 2197, 1632, 1565, 1469, 1143, 887, 714; δ_H ppm (300 MHz; $DMSO-d_6$; TMS) 2.40-2.66 (4H, m, $2 \times CH_2$), 2.71-3.00 (6H, m, $2 \times CH_2 + 2 \times CH$), 5.06 (1H, s, CH), 6.70-6.72 (1H, m, arom.), 7.01 (2H, t, $J = 7.8$ Hz, arom.), 7.15-7.78 (16H, m, arom.), 9.16 (1H, s, NH); δ_C ppm (75 MHz, $DMSO-d_6$, TMS) 30.4, 32.9, 33.1, 33.3, 34.1, 37.4, 37.7, 43.7, 96.5, 108.8, 108.9, 112.4, 112.9, 113.7, 113.8, 118.7, 119.0, 126.8, 127.0, 127.2, 127.3, 128.5, 128.6, 128.9, 129.5, 131.8, 132.0, 139.2, 142.9, 143.9, 145.9, 149.7, 151.0, 165.0, 165.2, 195.7.

3,4,6,7-tetrahydro-9-(3,4-dimethoxyphenyl)-3,3,6,6-tetramethylacridine-1,8(2H,5H,9H,10H)-dione (16e):



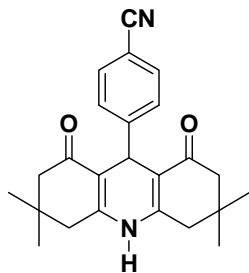
White solid; Yield: 380 mg, 93 %; m.p. 214-216 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{25}H_{31}NO_4$: C, 73.32; H, 7.63; N, 3.42 %. Found C, 73.60; H, 7.69; N, 3.55 %; IR (KBr) cm^{-1} : 3273, 3199, 3068, 2950, 1641, 1610, 1482, 1365, 1222; δ_H ppm (300 MHz; $CDCl_3$; TMS) 0.96 (6H, s), 1.05 (6H, s), 2.37-2.12 (8H, m), 3.74 (1H, s), 3.83 (1H, s), 5.02 (1H, s), 6.67-6.64 (1H, m), 6.80 (1H, d, $J = 8.1$ Hz), 6.91 (1H, s), 8.13 (1H, s); δ_C ppm (75 MHz, $CDCl_3$, TMS) 25.8, 28.86, 31.5, 49.7, 54.8, 110.7, 111.0, 111.4, 118.9, 139.4, 146.2, 147.4, 148.5, 193.8.

3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-(pyridin-2-yl)acridine-1,8-dione (16f):



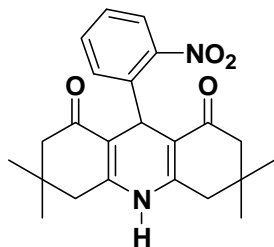
Red solid; Yield: 308 mg, 88 %; m.p. 284-286 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{26}N_2O_2$: C, 75.40; H, 7.48; N, 7.99 %. Found C, 75.21; H, 7.51; N, 8.09 %; IR (KBr) cm^{-1} : 3288, 3072, 2960, 1634, 1530, 1490, 1365, 1124, 1145; δ_H ppm (300 MHz; $DMSO-d_6$; TMS) 0.87 (6H, s), 1.00 (6H, s), 2.50-1.92 (8H, m), 4.96 (1H, s), 7.03 (1H, d, $J = 5.1$ Hz), 7.30 (1H, d, $J = 7.5$ Hz), 7.55 (1H, d, $J = 6.3$ Hz), 8.30 (1H, d, $J = 3.9$ Hz), 9.20 (1H, s); δ_C ppm (75 MHz, $DMSO-d_6$, TMS) 25.6, 28.6, 31.6, 34.9, 49.7, 109.8, 120.6, 122.4, 134.5, 147.9, 149.4, 163.3, 193.8.

4-(1,2,3,4,5,6,7,8,9,10-decahydro-3,3,6,6-tetramethyl-1,8-dioxoacridin-9-yl)benzonitrile (16g):



Yellowish white solid; Yield: 352 mg, 94 %; m.p. 194-196 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{24}H_{26}N_2O_2$: C, 76.98; H, 7.00; N, 7.48 %. Found C, 76.70; H, 7.09; N, 7.36 %; IR (KBr) cm^{-1} : 3208, 3072, 2958, 1633, 1530, 1490, 1365, 1124, 1141; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.89 (6H, s), 1.06 (6H, s), 2.56-1.96 (8H, m), 4.86 (1H, s), 7.34 (2H, d, $J = 8.1$ Hz), 7.65 (2H, d, $J = 8.1$ Hz), 9.45 (1H, s); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 25.9, 28.4, 31.5, 33.2, 49.5, 107.7, 109.9, 118.4, 120.6, 128.1, 128.6, 131.3, 149.2, 151.8, 193.7.

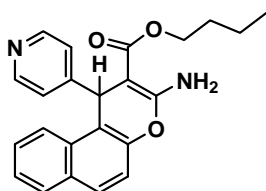
3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-9-(2-nitrophenyl)acridine-1,8(2H,5H,9H,10H)-dione (16h):



White solid; Yield: 365 mg, 92 %; m.p. 194-196 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{23}H_{26}N_2O_4$: C, 70.03; H, 6.64; N, 7.10 %. Found C, 70.33; H, 6.70; N, 7.29%; IR (KBr) cm^{-1} : 3208, 3280, 3200, 3072, 2958, 1633, 1530, 1490, 1365, 1146; δ_H ppm (300 MHz; $CDCl_3$; TMS) 0.90 (6H, s), 1.00 (6H, s), 2.34-2.05 (8H, m), 5.84 (1H, s), 7.21-7.15 (1H, m), 7.47-7.46 (2H, m), 7.69 (1H, d, $J = 8.1$ Hz), 8.52 (1H, s); δ_C ppm (75 MHz, $CDCl_3$, TMS) 26.0, 28.2, 28.8, 31.4, 49.6, 110.5, 123.0, 125.9, 129.7, 131.9, 141.1, 147.5, 149.3, 193.5.

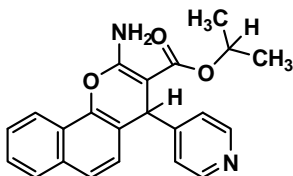
Spectroscopic characterization for 18a-18i

butyl 3-amino-1-(pyridin-4-yl)-1H-benzo[f]chromene-2-carboxylate (18a):



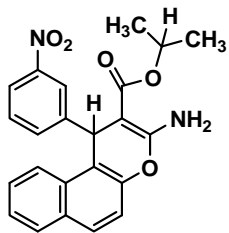
White solid; Yield: 347 mg, 93 %; m.p. 126-128 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{23}H_{22}N_2O_3$: C, 73.78; H, 5.92; N, 7.48 %. Found: C, 73.80; H, 5.93; N, 7.51 %; IR (KBr) cm^{-1} : 3361, 3264, 3179, 3060, 2952, 1680, 1629, 1594, 1526, 1407, 1220, 1072, 1019, 810, 741, 579, 514; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.84 (3H, t, $J = 6.9$ Hz, CH_3), 1.53-1.62 (2H, m, CH_2), 3.92-4.00 (1H, m, CH), 4.04-4.12 (1H, m, CH), 5.47 (1H, m, CH), 7.21 (2H, d, $J = 4.8$ Hz, arom.), 7.33-7.41 (2H, m, arom.), 7.48 (1H, t, $J = 7.2$ Hz, arom.), 7.72 (2H, s, NH_2), 7.86-7.90 (2H, m, arom.), 7.97 (1H, d, $J = 8.4$ Hz, arom.), 8.33 (2H, br s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 13.7, 18.8, 30.6, 36.2, 62.8, 76.5, 116.8, 117.4, 123.0, 125.0, 127.4, 128.7, 129.5, 130.2, 130.8, 146.9, 149.6, 155.0, 160.8, 167.9.

isopropyl 2-amino-4-(pyridin-4-yl)-4H-benzo[h]chromene-3-carboxylate (18b):



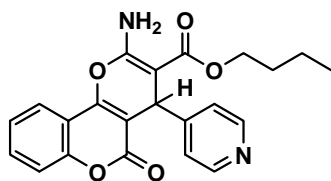
Pinkish white solid; Yield: 335 mg, 93 %; m.p. 152-154 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{18}N_2O_3$: C, 72.82; H, 5.24; N, 8.09 %. Found: C, 72.85; H, 5.25; N, 8.13 %; IR (KBr) cm^{-1} : 3358, 3261, 3054, 2976, 2363, 1940, 1680, 1630, 1596, 1526, 1219, 1070, 1030, 808, 777, 739, 594, 518; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.84 (3H, d, $J = 5.7$ Hz, CH_3), 1.15 (3H, d, $J = 6.0$ Hz, CH_3), 4.74-4.80 (1H, m, CH), 5.00 (1H, s, CH), 7.20-7.27 (3H, m, arom.), 7.55-7.62 (3H, m, arom.), 7.83 (3H, br s, arom.+ NH_2), 8.26 (1H, d, $J = 8.1$ Hz, arom.), 8.36 (2H, br s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 21.5, 22.1, 39.7, 65.8, 75.2, 119.1, 120.8, 122.8, 123.9, 126.4, 126.6, 126.7, 127.7, 132.7, 143.0, 149.6, 156.2, 160.8, 167.6.

isopropyl 3-amino-1-(3-nitrophenyl)-1H-benzo[f]chromene-2-carboxylate (18c):



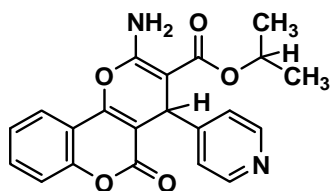
White solid; Yield: 368 mg, 91 %; m.p. 178-180 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{23}H_{20}N_2O_5$: C, 68.31; H, 4.98; N, 6.93 %. Found C, 68.32; H, 4.99; N, 6.93 %; IR (KBr) cm^{-1} : 3548, 3440, 3400, 3241, 3176, 3114, 2962, 2836, 1664, 1517, 1409, 1269, 1224, 1135, 1076, 1023, 819, 751,474; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 1.15-1.48 (6H, m, 2 \times CH $_3$), 3.50 (1H, s, CH), 5.64 (1H, s, CH), 7.16-7.29 (2H, m, arom.), 7.50 (2H, t, $J = 7.8$ Hz, arom.), 7.70-7.80 (4H, m, arom.), 7.98-8.14 (4H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 22.7, 23.4, 39.2, 65.1, 75.9, 119.2, 121.0, 122.4, 122.8, 126.9, 129.1, 129.2, 129.7, 133.4, 134.9, 147.5, 148.1, 153.5, 160.1, 167.5.

butyl 2-amino-4,5-dihydro-5-oxo-4-(pyridin-4-yl)pyrano[3,2-c]chromene-3-carboxylate (18d):



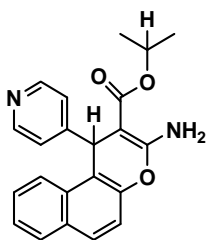
White solid; Yield: 361 mg, 92 %; m.p. 197-199 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{20}N_2O_5$: C, 67.34; H, 5.14; N, 7.14 %. Found: C, 67.36; H, 5.15; N, 7.17 %; IR (KBr) cm^{-1} : 3548, 3440, 3400, 3241, 3176, 3114, 2962, 2836, 1664, 1517, 1409, 1269, 1224, 1135, 1076, 1023, 819, 751,474; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.84 (3H, t, $J = 6.9$ Hz, CH $_3$), 1.20-1.30 (2H, m, CH $_2$), 1.53-1.62 (2H, m, CH $_2$), 3.82-4.03 (1H, m, CH), 4.04-4.16 (1H, m, CH), 5.23 (1H, m, CH), 7.14-7.26 (4H, m, NH $_2$ +arom.), 7.46-7.51 (2H, m, arom.), 7.79 (2H, d, $J = 7.8$ Hz, arom.), 8.35 (2H, br s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 14.0, 20.0, 30.3, 36.3, 64.3, 75.8, 102.3, 115.7, 119.7, 122.7, 123.0, 124.3, 131.3, 147.9, 152.6, 153.9, 159.2, 160.3, 167.9.

isopropyl 2-amino-4,5-dihydro-5-oxo-4-(pyridin-4-yl)pyrano[3,2-c]chromene-3-carboxylate (18e):



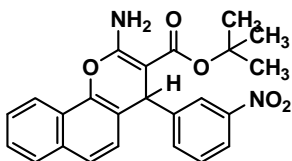
White solid; Yield: 352 mg, 93 %; m.p. 216-218 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{21}H_{18}N_2O_5$: C, 66.66; H, 4.79; N, 7.40 %. Found: C, 66.70; H, 4.80; N, 7.41 %; IR (KBr) cm^{-1} : 3555, 3443, 3409, 3255, 3176, 3114, 2968, 2835, 1655, 1520, 1410, 1269, 1225, 1135, 1066, 1065, 819, 757,474; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.74 (3H, d, $J = 5.7$ Hz, CH_3), 1.16 (3H, d, $J = 6.0$ Hz, CH_3), 4.75-4.81 (1H, m, CH), 5.23 (1H, s, CH), 7.14-7.26 (4H, m, NH_2 +arom.), 7.46-7.51 (2H, m, arom.), 7.77 (2H, d, $J = 7.8$ Hz, arom.), 8.46-8.78 (2H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 21.6, 22.2, 39.6, 65.4, 76.0, 102.4, 115.6, 119.7, 122.9, 123.0, 124.3, 131.5, 147.8, 152.7, 153.8, 159.3, 160.3, 167.9.

isopropyl 3-amino-1-(pyridin-4-yl)-1H-benzof[*f*]chromene-2-carboxylate (18f):



White solid; Yield: 324 mg, 90 %; m.p. 200-201 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{22}H_{20}N_2O_3$: C, 73.32; H, 5.59; N, 7.77 %. Found: C, 73.34; H, 5.61; N, 7.80 %; IR (KBr) cm^{-1} : 3556, 3440, 3397, 3247, 3176, 3120 2962, 2840, 1664, 1517, 1409, 1240, 1224, 1175, 1076, 1023, 819, 751,474; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.84 (3H, d, $J = 5.7$ Hz, CH_3), 1.14 (3H, d, $J = 6.0$ Hz, CH_3), 4.75-4.81 (1H, m, CH), 5.48 (1H, s, CH), 7.21-7.22 (2H, m, arom.), 7.32-7.42 (2H, m, arom.), 7.45-7.50 (1H, m, arom.), 7.73 (2H, s, NH_2), 8.86-8.90 (2H, m, arom.), 7.97 (1H, d, $J = 8.4$ Hz, arom.), 8.34 (2H, br s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 21.5, 22.1, 39.6, 64.8, 76.5, 116.8, 117.4, 123.0, 125.1, 127.4, 128.7, 129.7, 130.2, 130.7, 146.9, 149.6, 155.1, 160.7, 166.9.

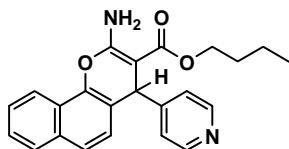
tert-butyl 2-amino-4-(3-nitrophenyl)-4H-benzo[*h*]chromene-3-carboxylate (18g):



White solid; Yield: 385 mg, 92 %; m.p. 202-204 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{24}H_{22}N_2O_5$: C, 68.89; H, 5.30; N, 6.69 %. Found: C, 68.91; H, 5.33; N, 6.70 %; IR (KBr) cm^{-1} : 3399, 3289, 2936, 1689, 1558, 1453, 1393, 1275, 1198, 1067, 759, 634, 511; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 1.32-1.56 (9H, m, $3 \times CH_3$), 5.41 (1H, s, CH), 7.40-7.46 (4H, m, NH_2 +arom.), 7.55-7.58 (3H, m, arom.), 7.72-7.89 (2H, m, arom.),

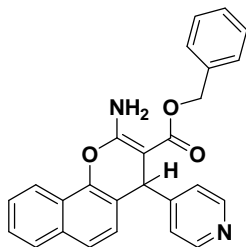
7.98-8.03 (1H, m, arom.), 8.19-8.21 (2H, m, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 23.4, 24.7, 25.5, 36.3, 60.7, 77.4, 119.1, 120.3, 121.5, 121.9, 124.4, 124.4, 124.8, 125.1, 125.5, 126.8, 128.9, 132.4, 134.3, 146.7, 147.1, 148.4, 168.2.

butyl 2-amino-4-(pyridin-4-yl)-4H-benzo[h]chromene-3-carboxylate (18h):



White solid; Yield: 340 mg, 91 %; m.p. 178-180 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{23}H_{22}N_2O_3$: C, 73.78; H, 5.92; N, 7.48 %. Found: C, 73.82; H, 5.92; N, 7.51 %; IR (KBr) cm^{-1} : 3556, 3446, 3430, 3260, 3180, 3128, 2958, 2836, 1670, 1519, 1449, 1276, 1264, 1137, 1079, 1023, 839, 751, 480; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 0.84 (3H, t, $J = 6.9$ Hz, CH_3), 1.19-1.39 (2H, m, CH_2), 1.53-1.62 (2H, m, CH_2), 3.82-4.00 (1H, m, CH), 4.02-4.22 (1H, m, CH), 5.10 (1H, m, CH), 7.20-7.29 (3H, m, arom.), 7.59-7.62 (3H, m, arom.), 7.81 (3H, br s, NH_2 +arom.), 8.26 (1H, d, $J = 8.1$ Hz, arom.), 8.36 (2H, br s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 13.9, 19.0, 30.2, 36.7, 63.8, 75.3, 119.2, 120.8, 122.8, 123.8, 126.4, 126.6, 126.7, 127.7, 132.8, 143.0, 149.7, 155.6, 160.9, 167.5.

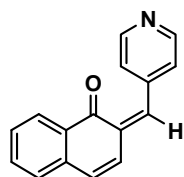
2-Amino-4-pyridin-4-yl-4H-benzo[h]chromene-3-carboxylic acid benzyl ester (18i):



White solid; Yield: 134 mg, 33 %; m.p. 178-180 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for $C_{26}H_{20}N_2O_3$: C, 76.45; H, 4.94; N, 6.86 %. Found: C, 76.47; H, 4.95; N, 6.87 %; IR (KBr) cm^{-1} : 3536, 3436, 3400, 3210, 3190, 3118, 2960, 2842, 1666, 1521, 1450, 1280, 1265, 1203, 1105, 1029, 840, 750; δ_H ppm (300 MHz; DMSO- d_6 ; TMS) 4.87 (2H, s, CH_2), 5.27 (1H, m, CH), 7.21-7.30 (3H, m, arom.), 7.37-7.40 (1H, m, arom.), 7.50-7.59 (2H, m, arom.), 7.61-7.63 (3H, m, arom.), 7.75-7.85 (4H, m, arom.), 8.06 (1H, dd, $J = 6.0$ Hz, $J = 3.0$ Hz, arom.), 8.34 (1H, dd, $J = 6.0$ Hz, $J = 3.0$ Hz, arom.), 8.49 (2H, s, arom.); δ_C ppm (75 MHz, DMSO- d_6 , TMS) 35.1, 66.8, 76.1, 119.1, 120.7, 122.9, 123.9, 126.3, 126.65, 126.74, 127.3, 127.8, 128.3, 132.7, 140.7, 142.9, 149.5, 156.1, 160.8, 168.0.

Spectroscopic characterization of intermediate 5a (First intermediate)

(2Z)-2-((pyridin-4-yl)methylene)naphthalen-1(2H)-one (5a):



Cream colored solid; m.p. 158-160 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₁₆H₁₁NO: C, 82.38; H, 4.75; N, 6.00 %. Found: C, 82.41; H, 4.76; N, 6.02 %; IR (KBr) cm⁻¹: 1723, 1591, 1371, 1094, 819, 742, 621; δ_H ppm (300 MHz; DMSO-d₆; TMS) 6.73 (1H, s, CH), 7.44 (1H, d, *J* = 8.7 Hz, arom.), 7.69 (2H, d, *J* = 5.1 Hz, arom.), 7.82-7.85 (2H, m, arom.), 7.90 (1H, d, *J* = 9.0 Hz, arom.), 8.11-8.14 (1H, m, arom.), 8.50-8.53 (1H, m, arom.), 8.88-8.92 (2H, m, arom.); δ_C ppm (75 MHz, DMSO-d₆, TMS) 113.2, 115.2, 121.8, 121.9, 122.5, 123.4, 124.4, 127.8, 128.2, 129.3, 134.5, 142.7, 150.3, 150.8, 153.4, 159.4.

Spectroscopic characterization of the other intermediate (Second intermediate)

7a-Hydroxy-13-(4-nitro-phenyl)-12a,13-dihydro-7aH-7-oxa-indeno[1,2-b]phenanthren-12-one:



Light yellow solid; m.p. 252-254 °C (recrystallized from EtOAc/Pet ether); Anal. Calcd for C₂₆H₁₇NO₅: C, 73.75; H, 4.05; N, 3.31 %. Found: C, 73.77; H, 4.06; N, 3.35 %; IR (KBr) cm⁻¹: 3409, 2915, 1705, 1595, 1515, 1341, 1219, 1074, 844, 740, 591; δ_H ppm (300 MHz; DMSO-d₆; TMS) 3.69 (1H, br s, CH), 5.43 (1H, br s, CH), 6.91 (1H, d, *J* = 8.4 Hz, arom.), 7.28 (1H, br s, arom.), 7.39 (1H, br s, arom.), 7.47 (2H, br s, arom.), 7.57 (2H, d, *J* = 6.9 Hz, arom.), 7.66 (1H, d, *J* = 8.4 Hz, arom.), 7.72-7.81 (3H, m, arom.), 8.02-8.08 (4H, m, arom.); δ_C ppm (75 MHz, DMSO-d₆, TMS) 37.1, 59.9, 102.7, 116.6, 119.2, 122.1, 122.3, 123.7, 123.9, 124.3, 127.4, 128.6, 129.6, 129.9, 130.5, 131.4, 132.3, 133.6, 136.7, 146.5, 150.1, 150.5, 153.1, 199.8.

Spectral Data of 4H-pyrans 7aa-7br

