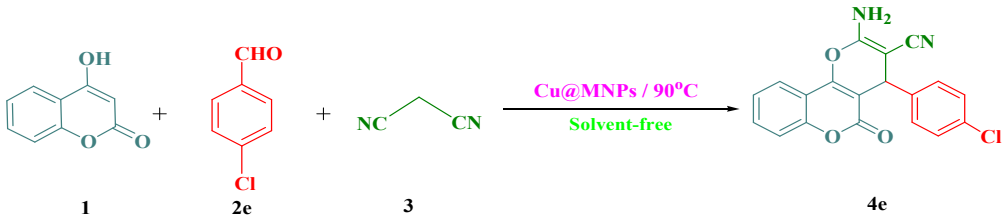
Table 1′ Optimization of the three-component reaction of 4-hydroxycoumarin (1), 4-chlorobenzaldehyde (2e), and malononitrile (3) when $Cu(salal)_2$ is replaced with $Cu(acac)_2$[a]



1 2e 3 4e

Entry	Solvent	Catalyst (mg)	Temp.	Time (min)	Yield (%)[b]
1	CH_2Cl_2	10	Reflux	7	51
2	CH_3CN	10	Reflux	7	57
3	toluene	10	Reflux	7	58
4	DMF	10	Reflux	7	62
5	H_2O	10	Reflux	7	66
6	EtOH	10	Reflux	7	80
7	Solvent- free	---	90°C	7	trace
8	Solvent- free	10	60°C	7	82
9	Solvent- free	10	80°C	7	86
10	Solvent- free	10	90°C	7	97
11	Solvent- free	10	100°C	7	96
12	Solvent- free	10	110°C	7	94
13	Solvent- free	5	90°C	7	67
14	Solvent- free	10	25°C	7	32
15	Solvent- free	15	90°C	7	93
13	Solvent- free	5	90°C	7	67
14	Solvent- free	10	25°C	7	32
15	Solvent- free	15	90°C	7	93

[a] Reaction conditions: 4-hydroxycoumarin (1 mmol), 4-chlorobenzaldehyde (1 mmol), malononitrile (1.1 mmol), and required amount of the catalysts.

[b] The yields refer to the isolated product.

Compound 4a: White crystalline solid; Mp: 258-260°C; IR (KBr, cm^{-1}): 3455, 3371, 3269, 2217, 1712, 1669, 1611, 1463, 1369,1216, 1124, 1055, 1005, 779; ^{1}H NMR (500 MHz,DMSO-d_6) δ: 4.53 (s, 1H, CH), 7.14 (s, 2H), 7.19-7.38 (m, 5H), 7.55 (d, 1H),7.71 (t, 1H), 7.82 (t, 1H,), 7.92 (d, 1H) ppm; ^{13}C NMR(125 MHz, DMSO-d_6) δ: 56.3, 103.2 112.9, 116.8, 121.3, 124.1, 124.4, 125.0, 125.9, 129.5, 130.1, 134.2, 146.7, 150.9, 153.0, 155.2, 157.3, 161.2 ppm; MS (ESI): *m*/*z* 317 $(M+H)^+$.

Compound 4b: Yellow crystalline solid; Mp: 260-263°C; IR (KBr, cm^{-1}): 3440, 3380, 3279, 2216, 1717, 1676, 1615, 1460, 1378, 1219, 1120, 1060, 1004, 788; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 4.61 (s, 1H), 7.05-7.11 (m, 3H), 7.23 (t, 1H), 7.34 (s, 2H), 7.49(d, 1H), 7.53 (t, 1H), 7.71 (t, 1H), 7.94(d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 57.3, 103.9, 113.9, 117.5, 119.7, 122.7, 124.6, 124.7,125.3, 129.1, 130.0, 134.2, 147.3, 150.7, 152.7, 149.9, 158.4, 160.8 ppm; MS (ESI): *m*/*z* 362 $(M+H)^+$.

Compound 4c: Yellow crystalline solid; Mp: 249-251°C; IR (KBr, cm^{-1}): 3451, 3378, 3284, 2223, 1715, 1672, 1622, 1456, 1382, 1117, 1069, 1008, 756, 621; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 4.58 (s, 1H), 7.18 (s, 2H), 7.30 (d, 2H), 7.38 (d, 2H), 7.53(d, 1H), 7.75 (t, 1H), 7.78 (t, 1H), 7.87 (d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 57.0, 104.2, 114.1, 117.5, 120.2, 123.6, 124.6, 124.8, 125.7, 129.4, 130.3, 134.0, 147.1, 151.2, 152.4, 155.2 , 157.5, 161.3 ppm; MS (ESI): *m*/*z* 362 $(M+H)^+$.

Compound 4d: White crystalline solid; Mp: 243-245°C; IR (KBr, cm^{-1}): 3439, 3383, 3271, 2209, 1712, 1666, 1600, 1455, 1381, 1224, 1120, 1064, 1005, 783; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 4.70 (s, 1H), 7.01 (t, 1H), 7.06(d, 1H), 7.17 (d, 1H),7.24 (t, 1H), 7.27 (s, 2H), 7.48(d, 1H), 7.55 (t, 1H),7.70 (t, 1H), 7.89 (d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 56.3, 104.3, 114.2, 117.4, 119.4, 123.7, 124.4, 125.1, 125.7, 129.9, 133.5, 147.2, 151.4, 153.5, 154.8, 157.3, 161.5 ppm; MS (ESI): *m*/*z* 351 $(M+H)^+$.

Compound 4e: White crystalline solid; Mp: 262-264°C; IR (KBr, cm^{-1}): 3443, 3372, 3282, 2208, 1711, 1676, 1606, 1454, 1371, 1234,1129, 1069, 1004, 775; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 4.56 (s, 1H), 7.10 (s, 2H), 7.23 (d, 2H), 7.40 (d, 2H), 7.49 (d, 1H), 7.73 (t, 1H), 7.75 (t, 1H), 7.88 (d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 56.6, 103.8, 113.5, 118.3, 120.0, 122.9, 123.6, 124.8, 125.9, 129.6, 130.1, 134.3, 146.9, 150.9, 152.8, 155.3, 157.9, 160.4 ppm; MS (ESI): m/z 351 $(M+H)^+$.

Compound 4f: White crystalline solid; Mp: 259-261°C; IR (KBr, cm^{-1}): 3449, 3375, 3282, 2223, 1711, 1677, 1622, 1607, 1451, 1379, 1115, 1066, 875, 638; ^{1}H NMR (500 MHz, DMSO-*d*6) δ: 4.63 (s, 1H), 7.18 (s, 2H), 7.25 (d, 2H), 7.47 (d, 2H), 7.60(d, 1H), 7.69 (t, 1H), 7.83 (t, 1H), 7.91 (d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-*d*6) δ: 56.9, 103.7, 114.4, 118.3, 120.3, 122.9, 123.9, 124.8, 125.8, 129.4, 130.6, 133.9, 146.9, 151.3, 152.6, 155.3, 157.8, 161.1 ppm; MS (ESI): *m*/*z* 335 $(M+H)^+$.

Compound 4g: White crystalline solid; Mp: 233-235°C; IR (KBr): 3447, 3392, 3261, 2215, 1712, 1676, 1606, 1462, 1382, 1256, 1175, 1129, 1052, 991, 787; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 3.75 (s, 3H), 4.75 (s, 1H), 6.89 (t, 1H), 6.98 (d, 1H), 7.11 (d, 1H), 7.23 (t, 1H), 7.29 (s, 2H), 7.48 (d, 1H), 7.53 (t, 1H), 7.71 (t, 1H), 7.90 (d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 32.7, 56.4, 57.6, 103.5, 112.4, 113.6, 117.3, 119.6, 127.1, 122.8, 125.2, 128.7, 129.4, 131.1, 133.5, 152.7, 154.6, 157.8, 159.3, 160.2 ppm; MS (ESI): *m*/*z* 347$(M+H)^+$.

Compound 4h: White crystalline solid; Mp: 240-243°C; IR (KBr): 3441, 3385, 3255, 2208, 1719, 1675, 1609, 1465, 1389, 1251, 1179, 1125, 1057, 998, 782; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.49 (s, 2H), 3.78 (s, 3H), 4.45 (s, 1H), 6.7-7.9 (m, 8H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 40.1, 55.4, 81.6, 103.7, 111.4, 113.5, 115.2, 116.8, 118.3, 120.8, 123.3, 124.8, 128.5, 129.7, 141.9, 152.5, 159.1, 159.9, 160.7, 161.8 ppm; MS (ESI): *m*/*z* 347$(M+H)^+$.

Compound 4i: White crystalline solid; Mp: 222-224°C; IR (KBr, cm^{-1}): 3446, 3375, 3277, 2218, 1716 , 1673, 1609, 1487, 1373,1329, 1246, 1121, 1067, 876, 675;1H NMR (500 MHz,

DMSO-d_6) δ: 3.66 (s, 3H), 4.73 (s, 1H), 7.22 (s, 2H), 7.34 (d, 2H), 7.46 (d, 2H), 7.55 (d, 1H), 7.71 (t, 1H), 7.76 (t, 1H), 7.90 (d, 1H) ppm; ^{13}C NMR(125 MHz, DMSO-d_6) δ: 54.7, 57.0, 104.3, 113.8, 117.5, 120.8, 123.0, 124.7, 125.2, 125.8, 129.5, 130.4, 134.3,147.2, 151.3, 153.4, 154.9, 157.6, 160.9 ppm; MS (ESI): *m*/*z* 347 $(M+H)^+$.

Compound 4j: White crystalline solid; Mp: 226-229°C; IR (KBr, cm^{-1}): 3446, 3385, 3272, 2216, 1712, 1663, 1606, 1463, 1378, 1264, 1179, 1121, 1056, 991, 781; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 3.71 (s, 6H), 4.42 (s, 1H), 6.78 (d, 1H), 6.87 (d, 2H), 7.32 (s, 2H), 7.41 (1H, d), 7.44 (t, 1H), 7.66 (t, 1H), 7.91 (d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 36.6, 55.7, 55.8, 58.3, 104.1, 111.8, 111.8, 112.7, 116.5, 119.3, 119.8, 122.3, 124.5, 132.2, 135.9, 148.3, 148.2, 152.1, 153.2, 158.0, 159.6 ppm; MS (ESI): *m*/*z* 377$(M+H)^+$.

Compound 4k: White crystalline solid; Mp: 256-259°C; IR (KBr, cm^{-1}): 3444, 3385, 3282, 2215, 1725, 1674, 1610, 1463, 1378, 1255, 1179, 1119, 1051, 990, 782; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 3.75 (s, 3H), 4.50 (s, 1H), 6.55 (d, 1H), 6.68 (d, 1H), 6.87 (s, 1H), 7.18-7.31 (m, 3H), 7.31 (d, 1H), 7.52 (t, 1H), 7.92 (d, 1H), 11.74 (s, 1H); ^{13}C NMR (125 MHz, DMSO-d_6) δ: 36.7, 56.3, 58.4, 111.0, 112.3, 112.5, 115.5, 115.9, 120.1, 120.6, 122.2, 122.4, 131.7, 135.6, 138.8, 145.7, 147.5, 151.41, 159.1, 160.2 ppm; MS (ESI): *m*/*z* 363$(M+H)^+$.

Compound 4l: White crystalline solid; Mp: 261-264°C; 3389, 3273, 2229, 1718, 1678, 1616, 1469, 1372, 1229, 1117, 1056, 1015, 761; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.28 (s, 3H), 4.65 (s, 1H), 7.16 (s, 2H), 7.22 (d, 2H), 7.31 (d, 2H), 7.49 (t, 1H), 7.69 (t, 1H), 7.75 (t, 2H), 7.87 (d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 20.1, 57.7, 104.3, 113.9, 118.1, 121.1, 124.0, 125.3, 125.9, 126.3, 129.7, 130.8, 134.4, 146.9, 150.7, 153.6, 155.8, 158.2, 161.4 ppm. MS (ESI): *m*/*z* 331 $(M+H)^+$.

Compound 4m: White crystalline solid; Mp: 250-252°C; IR (KBr, cm^{-1}): 3448, 3384, 3283, 2215, 1708, 1671, 1606, 1460, 1363, 1221, 1110, 1053, 1004, 766; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.19 (s, 3H), 4.69 (s, 1H), 7.21 (s, 2H), 7.27 (d, 2H), 7.36 (d, 2H), 7.54 (t, 1H), 7.72 (t, 1H), 7.81 (t, 2H),7.91 (d, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 20.5, 57.5, 104.0, 113.5, 117.7, 120.5, 123.3, 124.5, 125.0, 125.8, 129.6, 130.5, 133.7, 146.5, 150.5, 153.2, 155.3, 157.5, 160.3 ppm; MS (ESI): *m*/*z* 331 $(M+H)^+$.

Compound 4n: White crystalline solid; Mp: 258-260°C; IR (KBr, cm^{-1}): 3441, 3382, 3276, 2220, 1716, 1669, 1603, 1459, 1375, 1261, 1172, 1125, 1058, 994, 785; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 4.63 (s, 1H), 7.13 (s, 2H), 7.22 (d, 2H), 7.40 (d, 2H), 7.56 (d, 1H), 7.72 (t, 1H), 7.80 (t, 1H), 7.91 (d, 1H), 9.66 (s, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 56.9, 103.4, 113.8, 117.5, 119.9, 123.0,124.3, 124.8, 125.6, 129.2, 129.8, 133.9, 146.7, 150.5, 153.3, 154.7, 158.4, 160.5 ppm; MS (ESI): *m*/*z* 360 $(M+H)^+$.

Compound 6a: White crystalline solid; Mp: 232-234°C; IR (KBr, cm^{-1}): 3400, 3328, 3216, 2963, 2201, 1682, 1664, 1608, 1373, 1251, 1215, 1165, 1142, 1039, 698; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 0.99 (s, 3H), 1.09 (s, 3H), 2.14 (d, 1H), 2.23 (d, 1H), 2.49 (s, 2H), 4.15 (s, 1H), 7.00 (s, 2H), 7.15-7.23 (m, 3H), 7.25 (t, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 26.5, 28.2, 31.4, 35.6, 38.8, 50.1, 58.4, 112.6, 119.9, 126.7, 127.6, 128.5, 144.9, 158.2, 162.6, 195.8 ppm; MS (ESI): *m/z* 296 $(M+H)^+$.

Compound 6b: White crystalline solid; Mp: 216-219°C; IR (KBr, cm^{-1}): 3395, 3345, 3125, 2927, 2212, 1699, 1652, 1638, 1561, 1534, 1459, 1385, 1223, 1031, 843; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 0.99 (s, 3H), 1.15 (s, 3H), 2.16 (d, 1H), 2.25 (d, 1H,), 2.57 (s, 2H), 4.24 (s, 1H), 7.07 (s, 2H), 7.19-7.36 (m, 4H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 27.5, 29.1, 32.5, 36.4, 40.3, 50.7, 59.0, 114.1, 120.5, 127.3, 128.9, 130.6, 145.5, 159.0, 163.1, 196.4 ppm; MS (ESI): *m/z* 340 $(M+H)^+$.

Compound 6c: Yellow crystalline solid; Mp: 180-182°C; IR (KBr, cm^{-1}): 3396, 3326, 3260, 3216, 2963, 2196, 1741, 1689, 1656, 1606, 1526, 1368, 1348, 1258, 1219, 1045, 869, 822; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 0.93 (s, 3H), 1.01 (s, 3H), 2.08 (d, 1H), 2.24 (d, 1H), 2.48 (s, 2H), 4.34 (s, 1H), 7.16 (s, 2H), 7.43 (d, 2H), 8.15 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-

d_6) δ: 26.8, 28.3, 34.9, 35.5, 49.7, 57.1, 111.8, 119.5, 123.7, 128.4, 146.2, 152.6, 158.4, 163.1, 195.3 ppm; MS (ESI): *m/z* 340 $(M+H)^+$.

Compound 6d: White crystalline solid; Mp: 207-210°C; IR (KBr, cm^{-1}): 3368, 3190, 2966, 2194, 1741, 1683, 1653, 1606, 1507, 1372, 1218, 1144, 1041, 853; ^1H NMR (500 MHz, DMSO-d_6) δ: 0.98 (s, 3H), 1.06 (s, 3H), 2.12 (d, 1H), 2.27 (d, 1H), 2.52 (s, 2H), 4.23 (s, 1H), 7.03 (s, 1H), 7.11 (t, 2H), 7.17-7.25 (m, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 27.4, 28.7, 32.5, 35.6, 50.7, 58.2, 114.0, 115.1, 115.8, 120.4, 129.6, 129.7, 141.2, 141.5, 158.0, 160.1, 162.8, 163.1, 196.4 ppm; MS (ESI): *m/z* 329 $(M+H)^+$.

Compound 6e: White crystalline solid; Mp: 212-214°C; IR (KBr, cm^{-1}): 3396, 3323, 3259, 3216, 2967, 2196, 1744, 1687, 1657, 1601, 1369, 1215, 1044; ^1H NMR (500 MHz, DMSO-d_6) δ: 0.93 (s, 3H), 1.02 (s, 3H), 2.08 (d, 1H), 2.22 (d, 1H), 2.47 (s, 2H), 4.16 (s, 1H), 7.02 (s, 2H), 7.15 (d, 2H), 7.33 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 26.5, 28.4, 31.3, 35.6, 40.8, 49.7, 57.5, 112.4, 119.7, 128.6, 129.3, 131.1, 143.5, 158.9, 162.2, 195.9 ppm; MS (ESI): *m/z* 329 $(M+H)^+$.

Compound 6f: White crystalline solid; Mp: 229-231°C; IR (KBr, cm^{-1}): 3354, 328, 3116, 2903, 2218, 1675, 1642, 1629, 1555, 1529, 1451, 1381, 1219, 1017, 839; ^1H NMR (500 MHz, DMSO-d_6) δ: 0.95 (s, 3H), 1.05 (s, 3H), 2.12 (d, 1H), 2.27 (d, 1H), 2.52 (s, 2H), 4.21 (s, 1H), 7.08 (s, 2H), 7.16-7.18 (m, 1H), 7.26 (t, 1H), 7.31 (t, 1H), 7.37-7.39 (m, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 28.9, 32.5, 35.9, 40.7, 50.5, 58.3, 112.8, 120.1, 122.1, 126.9, 130.1, 130.5, 131.3, 147.8, 159.6, 163.7, 196.4 ppm; MS (ESI): *m/z* 372 $(M+H)^+$.

Compound 6g: White crystalline solid; Mp: 208-211°C; IR (KBr, cm^{-1}): 3419, 3325, 3163, 2959, 2224, 1689, 1678, 1636, 1586, 1521, 1448, 1384, 1232, 1038, 845; ^1H NMR (500 MHz, DMSO-d_6) δ: 0.97 (s, 3H), 1.06 (s, 3H), 2.12 (d, 1H), 2.28 (d, 1H), 2.54-2.52 (m, 2H), 4.21 (s, 1H), 7.09 (s, 2H), 7.11 (d, 2H), 7.49 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 27.3, 28.8, 32.2, 35.7, 50.5, 58.2, 112.8, 120.3, 130.1, 131.8, 144.3, 158.9, 163.2, 196.5 ppm; MS (ESI): *m/z* 372 $(M+H)^+$.

Compound 6h: Whit crystalline solid; Mp: 207-209°C; IR (KBr, cm^{-1}): 3482, 3338, 3184, 2956, 2204, 2188, 1689, 1669, 1597, 1363, 1249, 1208, 1142, 1038, 751; ^1H NMR (500 MHz, DMSO-d_6) δ: 0.97 (s, 3H), 1.05 (s, 3H), 2.12 (d, 1H), 2.26 (d, 1H), 2.52 (s, 2H), 3.37 (s, 2H), 4.21 (s, 1H), 7.03 (s, 2H), 7.11 (t, 2H), 7.18-7.23 (m, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 27.4, 29.0, 32.4, 35.6, 50.7, 58.7, 114.0, 115.5, 115.8, 120.3, 129.6, 129.8, 141.6, 141.5, 159.0, 160.3, 162.7, 163.0, 196.3 ppm; MS (ESI): *m/z* 313 $(M+H)^+$.

Compound 6i: Yellow crystalline solid; Mp: 225-229°C; IR (KBr, cm^{-1}): 3199, 2926, 2905, 2813, 2339, 2178, 1681, 1645, 1584, 1509, 1363, 1242, 1215, 1150, 1134, 1026, 839; ^1H NMR (500 MHz, DMSO-d_6) δ: 0.97 (s, 3H), 1.06 (s, 3H), 2.16 (d, 1H,), 2.29 (d, 1H), 2.56 (s, 2H), 4.34 (s, 1H), 7.19 (s, 2H), 7.35 (d, 2H), 7.75 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 28.1, 29.3, 32.9, 36.8, 50.9, 58.3, 111.0, 112.8, 119.7, 120.3, 129.5, 133.6, 151.2, 159.5, 163.9, 196.7 ppm; MS (ESI): *m/z* 320 $(M+H)^+$.

Compound 6j: White crystalline solid; Mp: 217-220°C; IR (KBr, cm^{-1}): 3416, 3321, 3161, 2952, 2216, 1682, 1669, 1629, 1598, 1523, 1456, 1373, 1219, 1051, 853; ^1H NMR (500 MHz, DMSO-d_6) δ: 1.06 (s, 3H), 1.12 (s, 3H), 2.22 (d, 2H), 2.28 (s, 3H), 2.45 (s, 2H), 4.36 (s, 1H), 4.59 (s, 2H), 7.13-7.10 (m, 4H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 21.1, 27.5, 28.6, 32.4, 35.3, 40.8, 50.9, 114.2, 118.6, 127.7, 129.4, 136.9, 140.1, 157.2, 161.6, 195.53 ppm; MS (ESI): *m/z* 309 $(M+H)^+$.

Compound 6k: Yellow crystalline solid; Mp: 203-205°C; 3355, 3190, 2968, 2195, 1686, 1658, 1609, 1511, 1372, 1257, 1216, 1036, 847; ^1H NMR (500 MHz, DMSO-d_6) δ: 0.95 (s, 3H), 1.05 (s, 3H), 2.11 (d, 1H), 2.25 (d, 1H), 2.51 (s, 2H), 3.72 (s, 1H), 4.14 (s, 1H), 6.85 (d, 2H), 6.95 (s, 2H), 7.07 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 26.2, 28.5, 31.8, 34.9, 50.4, 55.1, 58.9, 112.8, 114.2, 119.8, 128.4, 137.0, 158.1, 158.6, 162.4, 195.9 ppm; MS (ESI): *m/z* 324$(M+H)^+$.

Compound 6l: Yellow crystalline solid; Mp: 223-225°C; 3481, 3319, 3165, 2965, 2206, 1686, 1664, 1619, 1597, 1520, 1455, 1371, 1215, 1046, 851; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 0.94 (s, 3H), 1.04 (s, 3H), 2.08 (d, 1H), 2.25 (d, 1H), 2.42-2.54 (m, 2H), 4.08 (s, 1H), 6.63 (d, 2H), 6.90-6.97 (m, 4H), 9.29 (s, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 27.3, 29.0, 32.4, 35.6, 40.2, 50.6, 59.7, 114.3, 115.7, 120.8, 128.5, 135.2, 156.1, 158.9, 162.8, 196.4 ppm; MS (ESI): *m*/*z* 312 $(M+H)^+$.

Compound 6m: White crystalline solid; Mp: 237-239°C; IR (KBr, cm^{-1}): 3417, 3338, 3154, 2937, 2204, 1691, 1663, 1642, 1571, 1525, 1454, 1379, 1236, 1041, 851; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 0.97 (s, 3H), 1.03 (s, 3H), 2.10 (d, 1H), 2.25 (d, 1H), 2.51-2.48 (m, 2H), 3.71 (s, 3H), 4.07 (s, 1H), 6.53-6.50 (m, 1H), 6.68-6.64 (m, 2H), 6.92 (s, 2H), 8.86 (s, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 26.6, 28.5, 31.8, 35.0, 50.0, 55.6, 58.8, 111.4, 113.0, 115.3, 119.4, 119.9, 135.8, 145.2, 147.3, 158.4, 162.2, 195.8 ppm; MS (ESI): *m*/*z* 341 $(M+H)^+$.

Compound 6n: Yellow crystalline solid; Mp: 209-211°C; IR (KBr, cm^{-1}): 3198, 2962, 2925, 2802, 2359, 2337, 2193, 1682, 1659, 1606, 1519, 1369, 1246, 1212, 1160, 1142, 1033, 827; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 0.94 (s, 3H), 1.02 (s, 3H), 2.07 (d, 1H), 2.25 (d, 1H), 2.49 (q, 2H), 2.85 (s, 6H), 4.03 (s, 1H), 6.61 (d, 2H), 6.85 (s, 2H), 6.96 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 26.9, 28.7, 31.2, 34.4, 39.6 40.1, 50.3, 59.1, 112.8, 114.1, 120.1, 127.8, 132.6, 150.1, 158.2, 161.9, 196.3 ppm; MS (ESI): *m*/*z* 338 $(M+H)^+$.

Compound 9a: White crystalline solid; Mp: 237-239°C; IR (KBr, cm^{-1}): 3405, 3329, 3211, 3088, 2202, 1712, 1678, 1617, 1388, 1265, 1142; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.27 (s, 3H), 4.29 (s, 1H), 6.30 (s, 2H), 7.20 (s, 1H), 7.22 (d, 2H), 7.26 (t, 1H), 7.35 (t, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 19.4, 36.4, 57.9, 97.7, 100.3, 119.5, 127.2, 127.7, 128.6, 143.5, 157.4, 157.9, 158.1, 158.3, 161.5, 162.8 ppm; MS (ESI): *m*/*z* 281 $(M+H)^+$.

Compound 9b: White crystalline solid; Mp: 230-233°C; IR (KBr, cm^{-1}): 3385, 3330, 3197, 3099, 2203, 1711, 1677, 1616, 1386, 1263, 1144; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.33 (s, 3H), 4.43 (s, 1H), 6.41 (s, 2H), 7.37 (d, 2H), 7.40 (s, 1H), 7.49 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 19.4, 35.9, 57.6, 98.1, 100.7, 119.8, 129.0, 129.6, 129.9, 131.8, 132.5, 142.9, 158.3, 160.5, 161.8, 163.6 ppm; MS (ESI): *m*/*z* 315 $(M+H)^+$.

Compound 9c: White crystalline solid; Mp: 221-224°C; IR (KBr, cm^{-1}): 3378, 3324, 3175, 3081, 2220, 1712, 1654, 1611, 1378, 1261, 1134; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.18 (s, 3H), 4.47 (s, 1H), 6.51 (s, 1H), 6.75 (s, 2H), 7.11 (d, 2H), 7.35 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 23.0, 26.1, 56.9, 101.0, 102.2, 118.1, 125.6, 128.4, 128.8, 137.1, 143.7, 161.5, 164.2, 176.0 ppm; MS (ESI): *m*/*z* 299 $(M+H)^+$.

Compound 9d: White crystalline solid; Mp: 215-217°C; IR (KBr, cm^{-1}): 3375, 3321, 3178, 3085, 2223, 1709, 1662, 1609, 1374, 1259, 1135; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.24 (s, 3H), 4.41 (s, 1H), 6.49 (s, 1H), 6.69 (s, 2H), 7.18 (d, 2H), 7.42 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 22.4, 26.7, 57.9, 101.3, 102.7, 118.6, 125.0, 128.7, 129.2, 137.8, 144.5, 160.8, 164.6, 176.4 ppm; MS (ESI): *m*/*z* 359 $(M+H)^+$.

Compound 9e: Yellow crystalline solid; Mp: 219-223°C; IR (KBr, cm^{-1}): 3381, 3311, 3175, 3091, 2212, 1710, 1675, 1619, 1384, 1265, 1141; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.13 (s, 3H), 4.36 (s, 1H), 6.42 (s, 2H), 7.39 (d, 2H), 7.42 (s, 1H), 7.46 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 23.5, 35.7, 56.6, 98.4, 100.8, 117.5, 127.0, 128.4, 129.5, 131.6, 132.7, 143.9, 158.5, 160.7, 161.5, 163.8 ppm; MS (ESI): *m*/*z* 326 $(M+H)^+$.

Compound 9f: White crystalline solid; Mp: 209-212°C; IR (KBr, cm^{-1}): 3372, 3315, 3169, 3082, 2215, 1711, 1660, 1602, 1371, 1254, 1130; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.19 (s, 3H), 3.60 (s, 3H), 4.42 (s, 1H), 6.51 (s, 1H), 6.75 (s, 2H), 7.15 (d, 2H), 7.34 (d, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 22.1, 26.9, 53.9, 57.0, 101.4, 102.6, 118.2, 125.7, 128.2, 129.3, 137.4, 144.9, 161.2, 164.5, 176.9 ppm; MS (ESI): *m*/*z* 311 $(M+H)^+$.

Compound 10a: White crystalline solid; Mp: 276-280°C; IR (KBr, cm^{-1}): 3451, 3351, 3152, 2205, 1663, 1632; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.17 (s, 3H), 4.32 (s, 1H), 5.83 (s, 1H), 6.95 (s, 2H), 7.12-7.37 (m, 5H), 11.49 (s, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 20.0, 36.5, 57.2, 97.6, 100.2, 119.7, 127.0, 127.2, 128.5, 143.2, 157.1, 157.6, 158.2, 158.4, 161.9, 162.4 ppm; MS (ESI): *m*/*z* 280 $(M+H)^+$.
Compound 10b: White crystalline solid; Mp: 242-245°C; IR (KBr, cm^{-1}): 3458, 3309, 3205, 2200, 1663, 1625; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.16 (s, 3H), 4.32 (s, 1H), 5.89 (s, 1H), 7.01 (d, 2H), 716 (d, 2H), 7.37 (d, 2H), 11.50 (s, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 23.4, 27.5, 56.8, 101.5, 103.6, 118.2, 126.3, 129.2, 130.4, 137.9, 142.8, 161.0, 164.3, 161.5, 177.2 ppm; MS (ESI): *m*/*z* 314 $(M+H)^+$.
Compound 10c: White crystalline solid; Mp: 258-261°C; IR (KBr, cm^{-1}): 3448, 3299, 3150, 2207, 1661, 1632; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.18 (s, 3H), 4.27 (s, 1H), 6.31 (s, 1H), 7.18 (s, 2H), 7.20-7.23 (m, 2H), 7.30-7.33 (m, 2H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 23.2, 27.7, 56.6, 101.5, 103.3, 118.5, 126.2, 129.4, 130.3, 138.6, 144.0, 160.1, 164.5, 176.9 ppm; MS (ESI): *m*/*z* 298 $(M+H)^+$.
Compound 10d: White crystalline solid; Mp: 253-256°C; IR (KBr, cm^{-1}): 3450, 3301, 3152, 2205, 1659, 1630; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.20 (s, 3H), 4.29 (s, 1H), 6.24 (s, 1H), 7.16 (d, 2H), 7.23 (s, 2H), 7.45 (d, 2H), 11.51 (s, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 22.5, 26.8, 57.5, 101.2, 102.7, 118.2, 125.7, 128.1, 129.5, 137.3, 144.5, 160.3, 164.7, 176.9 ppm; MS (ESI): *m*/*z* 359 $(M+H)^+$.
Compound 10e: White crystalline solid; Mp: 276-279°C; IR (KBr, cm^{-1}): 3352, 3301, 3152, 2204, 1659, 163323; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.21 (s, 3H), 4.48 (s, 1H), 5.94 (s, 1H), 7.17 (d, 2H), 7.48 (d, 2H), 8.16 (d, 2H), 11.59 (s, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 23.5, 27.4, 56.2, 101.3, 103.2, 118.9, 126.9, 129.8, 130.6, 138.5, 144.4, 161.0, 164.4, 176.5 ppm; MS (ESI): *m*/*z* 325 $(M+H)^+$.
Compound 10f: White crystalline solid; Mp: 225-228°C; IR (KBr, cm^{-1}): 3453, 3305, 3150, 2202, 1661, 1633; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 2.18 (s, 3H), 3.75 (s, 3H), 4.27 (s, 1H), 5.85 (s, 1H), 6.87 (d, 2H), 6.95 (s, 2H), 7.09 (d, 2H), 11.52 (s, 1H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ: 22.1, 26.6, 54.5, 57.4, 101.2, 125.6, 128.1, 129.7, 137.3, 144.5, 161.2, 164.7, 176.9 ppm; MS (ESI): *m*/*z* 310 $(M+H)^+$.