Supporting Information

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Contents

Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2ʿ-dione (4a): Creamy powder (0.34 g, 76%). Mp: 268-270 °C. IR (KBr): 3537, 3297, 3068, 2928, 1700, 1743, 1550, 1507, 757 cm^{-1}. ^{1}H NMR(400.22 MHz, DMSO): 12.05 (1H, s, NH), 9.39 (1H, s, NH), 8.13 (1H, d, *J* 8.0 Hz), 7.78 (1H, t, *J* 7.8 Hz), 7.63-7.52 (6H, m), 7.42 (1H, t, *J* 6.6 Hz), 7.19 (2H, d, *J* 3.6 Hz), 6.89 (1H, d, *J* 7.6 Hz), 6.83-6.79 (1H, m), 1.79 (3H, s, CH_3). ^{13}C NMR(100.63 MHz, DMSO): 12.1 (CH_3), 94.4, 109.8, 111.5, 117.7, 117.9, 118.7, 121.9, 124.9, 125.0, 127.1, 127.2, 128.8, 130.0, 133.8, 138.9, 139.2, 140.5, 140.6, 145.3, 152.7, 155.3, 156.2, 181.8, 181.9. MS (EI, 70 eV) m/z= 448 (M+2, 4), 447 (M+1, 20), 446 (M+, 73), 417 (M^+ -HCO, 100), 286 (51), 258 (28), 217 (36), 190 (12), 121 (11), 77 (37). Anal. Calcd for $C_{27}H_{18}N_4O_3$ (446.46): C, 72.64; H, 4.06; N, 12.55%. Found: C, 72.57; H, 4.02; N, 12.63%.

Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1,3-diphenyl-5,2ʿ-dione (4b): Creamy powder; (0.36 g, 71%). Mp: 290-292°C. IR (KBr): 3301, 3054, 1738, 1722, 1609, 1543, 1456, 966, 914, 758, 699 cm^{-1}. ^{1}H NMR(400.22 MHz, DMSO): 11.77 (1H, s, NH), 9.53 (1H, s, NH), 7.86 (1H, d, *J* 8.0 Hz), 7.74 (2H, d, *J* 7.6 Hz), 7.69 (1H, t, *J* 8.0 Hz), 7.65 (2H, t, *J* 8.0 Hz), 7.48 (1H, t, *J* 7.4 Hz), 7.43 (1H, t, *J* 7.6 Hz), 7.39 (1H, t, *J* 8.4 Hz), 7.25 (1H, d, *J* 8.0 Hz), 7.21 (1H, t, *J* 7.4 Hz), 7.12 (2H, d, *J* 6.0 Hz), 7.03-7.01 (3H, m), 6.85 (1H, d, *J* 7.2 Hz), 6.81 (1H, t, *J* 7.2). ^{13}C NMR(100.63 MHz, DMSO): 94.7, 109.7, 111.3, 117.3, 118.7, 122.0, 123.5, 123.5, 124.5, 124.7, 126.9, 127.6, 127.9, 128.2, 128.3, 128.9, 130.1, 133.5, 138.7, 139.0, 139.1, 140.7, 149.9, 153.1, 155.0, 156.2, 182.0 and 182.2 (2CO). MS (EI, 70 eV) m/z= 508 (M^+, 2), 446 (22.5), 417 (75), 403 (M^+ -PhN_2, 14), 311 (14), 270 (13), 121 (100). Anal. Calcd for $C_{32}H_{20}N_4O_3$ (508.53): C, 75.58; H, 3.96; N, 11.02%. Found: C, 75.65; H, 3.81; N, 11.14%.

Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1-phenyl-3-propyl-5,2′-dione (4c): Creamy powder; (0.35 g, 73%). Mp: 240-242 °C. IR (KBr): 3445, 3308, 3213, 2964, 1641, 1610, 1545, 1421, 758, 697 cm^{-1}. ^{1}H NMR(400.22 MHz, DMSO): 11.88 (1H, s, NH), 10.17 (1H, s, NH), 8.02 (1H, d, *J* 8.0 Hz), 7.71 (2H, d, *J* 7.6 Hz), 7.63 (1H, t, *J* 7.8 Hz), 7.51-7.32 (7H, m), 7.02 (1H, t, *J* 7.2 Hz), 6.97 (1H, d, *J* 7.6 Hz), 2.01-1.89 (2H, m, α-CH_2), 1.24-1.17 (1H, m, β-CH_A), 1.04-0.96 (1H, m, β-CH_B), 0.62 (3H, t, *J* 7.4 Hz, CH_3). ^{13}C NMR(100.63 MHz, DMSO): 14.4 (CH_3), 22.8 (β-CH_2), 30.7 (α-CH_2), 93.4, 110.6, 115.3, 117.1, 119.4, 121.6, 122.3, 123.4, 124.7, 125.7, 126.7, 129.3, 131.1, 132.6, 136.9, 138.5, 142.9, 149.1, 152.3, 153.0, 154.2, 156.1, 178.9 and 183.3 (2CO). MS (EI, 70 eV) m/z = 474 (M^+, 1), 472 (M^+, 2), 446 (M^+ -CH_2=CH_2, 3), 331 (79), 288 (35), 275 (21), 239 (15), 202 (22), 161 (58), 133 (45), 119 (13), 104 (47), 91 (38), 77 (100). Anal. Calcd for $C_{29}H_{22}N_4O_3$ (474.51): C, 73.40; H, 4.67; N, 11.81%. Found: C, 73.43; H, 4.77; N, 11.70%.

Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-5ʿ-methoxy-1-phenyl-5,2ʿ-dione (4d): Creamy powder; (0.37 g, 78%), Mp: 289-292 °C. IR (KBr): 3396, 2920, 2855, 1688, 1660, 1505, 1450, 1288, 1039, 757 cm^{-1}. ^{1}H NMR(400.22 MHz, DMSO): 11.95 (1H, s, NH), 10.09 (1H, s, NH), 8.06 (1H, d, *J* 7.2 Hz), 7.69 (2H, d, *J* 8.0 Hz), 7.63 (1H, t, *J* 7.4 Hz), 7.51-703 (5H,

m), 6.99-6.87 (2H, m), 6.67 (1H, m), 3.70 (3H, s, OCH_3), 1.60 (3H, CH_3). MS (EI, 70 eV) m/z = 476 (M^+, 7), 474 (M^+ -2, 65), 320 (100), 277 (24), 174 (50), 77 (37). Anal. Calcd for $C_{28}H_{20}N_4O_4$ (476): C, 70.58; H, 4.23; N, 11.76. Found: C, 70.72; H, 4.25; N, 11.65%.

Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5ʿ-methoxy-1,3-diphenyl-5,2ʿ-dione (4e): Creamy powder; (0.39 g, 73%). Mp: 290-294°C. IR (KBr): 3346, 3214, 3059, 1748, 1659, 1454, 1043, 761, 698 cm^{-1}. ^1H NMR(400.22 MHz, DMSO): 11.70 (1H, s, NH), 9.27 (1H, s, NH), 7.82 (1H, dd, *J* 1.4 and 7.6 Hz), 7.73 (2H, dd, *J* 1.2 and 8.4 Hz), 7.68 (1H, t, *J* 8.2 Hz), 7.63 (2H, t, *J* 7.6 Hz), 7.46 (1H, t, *J* 7.8 Hz), 7.41 (1H, t, *J* 7.6 Hz), 7.36 (1H, d, *J* 8.4 Hz), 7.19 (1H, d, *J* 8.8 Hz), 7.08 (2H, dd, *J* 1.6 and 8.0 Hz), 7.01-6.96 (3H, m), 6.88 (1H, dd, *J* 2.8 and 8.8 Hz), 6.32 (1H, d, *J* 2.8 Hz), 3.58 (3H, s, OCH_3). MS (EI, 70 eV) m/z = 538 (M^+, 1), 536 (M^+-2, 4), 431 (M^+ -PhN_2, 35), 368 (25), 339 (19), 313 (13), 265 (13), 250 (16), 236 (20), 221 (13), 207 (13), 180 (22), 165 (16), 147 (20), 136 (30), 121(50), 105 (PhN_2, 100). Anal. Calcd for $C_{33}H_{22}N_4O_4$ (538.55): C, 73.60; H, 4.12; N, 10.40%. Found C, 73.65; H, 4.09; N, 10.34%.

Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5ʿ-methoxy-1-phenyl-3-propyl-5,2ʿ-dione (4f): Creamy powder; (0.40 g, 79%). Mp: 298-300 °C. IR (KBr): 3328, 3266, 2957, 2931, 1757, 1701, 1501, 760, 405 cm^{-1}. ^1H NMR(400.22 MHz, DMSO): 12.04 (1H, s, NH), 9.14 (1H, s, NH), 8.12 (1H, dd, *J* 1.6 and 8.4 Hz), 7.78 (1H, dt, *J* 1.6 and 7.8 Hz), 7.65-7.52 (6H, m), 7.40 (1H, tt, *J* 1.5 and 7.2 Hz), 7.15 (1H, 7′-H d, *J* 8.8 Hz), 6.88 (1H, 6′-H, dd, *J* 2.8 and 8.8 Hz), 6.38 (1H, 4′-H, d, *J* 2.8 Hz), 3.61 (1H, s, OCH_3), 2.13-1.99 (2H, m, α-CH_2), 1.43-1.24 (2H, m, β-CH_2), 0.62 (3H, t, *J* 7.4 Hz, CH_3). ^{13}C NMR(100.63 MHz, DMSO): 14.4 (CH_3), 22.2 (β-CH_2), 29.0 (α-CH_2), 55.8 (OCH_3), 93.4, 109.8, 111.6, 112.1, 114.6, 117.7, 118.9, 119.6, 122.9, 124.9, 125.0, 126.9, 129.9, 133.1, 133.8, 139.1, 140.8, 149.3, 152.8, 154.6, 155.3, 156.2, 181.9 and 182.1 (2CO). MS (EI, 70 eV) m/z= 504 (M^+, 1), 502 (M^+ -2, 5), 461 (2), 431(9), 396 (5), 349 (26), 291(25), 216 (64), 173 (58), 105 (88), 43 (100). Anal. Calcd for $C_{30}H_{24}N_4O_4$ (490): C, 71.42; H, 4.79; N, 11.10%. Found: C, 71.39; H, 4.84; N, 11.17%.

Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5ʿ-bromo-1-phenyl-3-propyl-5,2ʿ-dione (4g): Creamy powder; (0.355 g, 64%). Mp: 262-264 °C. IR (KBr): 3445, 3308, 3213, 2964, 1641, 1610, 1545, 1421, 758, 697 cm^{-1}. ^1H NMR(400.22 MHz, DMSO): 12.03 (1H, s, NH), 9.35 (1H, s, NH), 8.03 (1H, d, *J* 8.0 Hz), 7.79 (2H, d, *J* 7.6 Hz), 7.69 (1H, d, *J* 8.0 Hz), 7.64 (1H, d, *J* 7.2 Hz), 7.56-7.32 (5H, m), 7.24 (1H, t, *J* 7.6 Hz), 6.86 (1H, d, *J* 8.0 Hz), 6.74 (1H, t, *J* 9.2 Hz), 2.04-1.92 (2H, m, α-CH_2), 1.25-1.07 (2H, m, β-CH_2), 0.75 (3H, t, *J* 7.2 Hz). MS (EI, 70 eV) m/z = 554 (M^+, ^{81}Br, 5), 502 (M^+, ^{79}Br, 5), 444 (13), 417 (24), 402 (88), 362 (45), 339 (23), 201 (42), 121 (79), 105 (42), 77 (100). Anal. Calcd for $C_{29}H_{21}BrN_4O_3$ (553.41): C, 62.94; H, 3.82; N, 10.12%. Found: C, 63.08; H, 3.90; N, 9.93%.

Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5ʿ-chloro-1-phenyl-3-propyl-5,2ʿ-dione (4h): Creamy powder; (0.336 g, 66%). Mp: 260-262 °C. IR (KBr): 3447, 3302, 2964, 1640, 1610, 1546, 1422, 1298, 760, 694 cm^{-1}. ^1H NMR(400.22

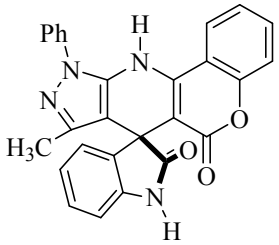
MHz, DMSO): 11.30 (1H, s, NH), 9.13 (1H, s, NH), 8.04 (1H, d, *J* 8.4 Hz), 7.70 (2H, d, *J* 8.0 Hz), 7.64 -7.63 (1H, m), 7.49 -7.32 (5H,m), 7.42 -7.32 (5H, m), 7.18-7.14 (1H,m), 6.98 (1H, d, *J* 8.4 Hz), 6.84-6.78 (1H, m), 2.03-1.93 (2H, m, α-CH_2), 1.28 (1H, m, β-CH_A), 1.09 (1H, m, β-CH_B), 0.90 (3H, t, J 7.2 Hz). MS (EI, 70 eV) m/z= 508 (M^+ -2, 37Cl, 1), 506 (M^+ -2, 35Cl, 4), 367 (49), 365 (76), 330 (65), 309 (25), 202 (50), 161 (80), 133 (62), 121 (29), 104 (65), 91 (77), 77 (100). Anal. Calcd for $C_{29}H_{21}ClN_4O_3$ (508.96): C, 68.44; H, 4.16; N, 11.01. Found: C, 68.52; H, 4.21; N, 10.90.

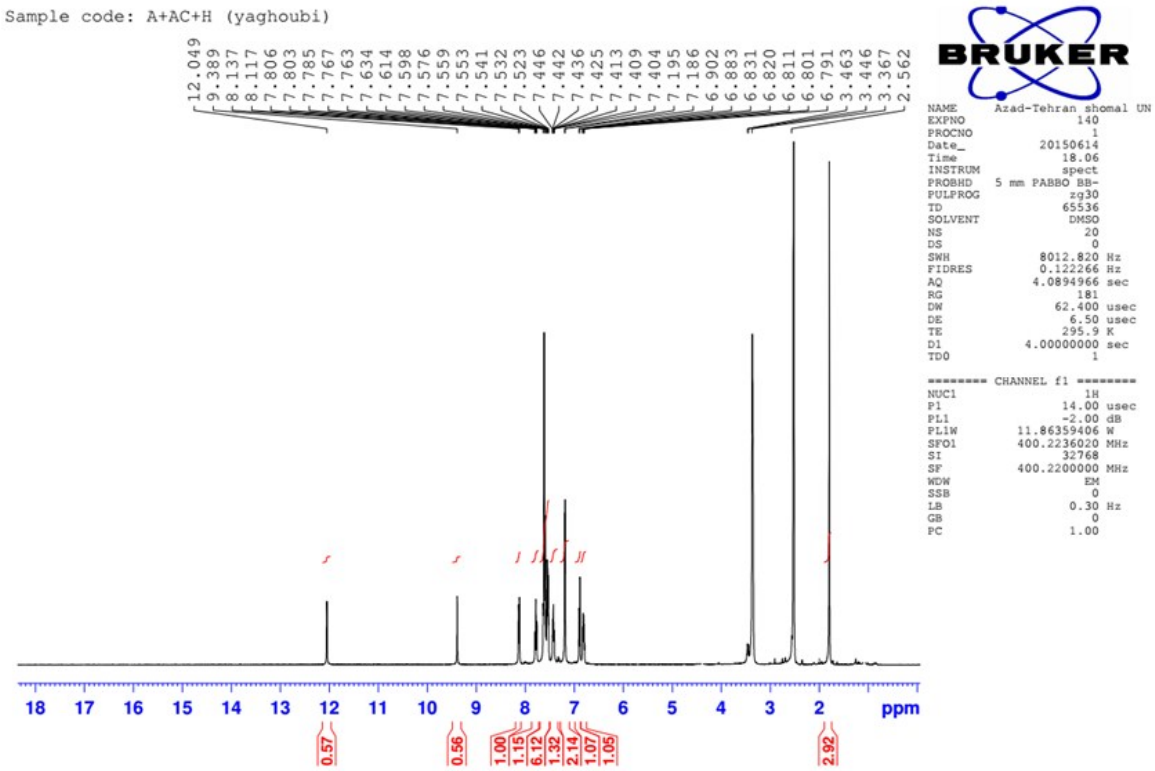
Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5ʿ-chloro-3-methyl-1-phenyl-5,2ʿ-dione (4i): Creamy powder; (0.37 g, 77%). Mp: 305-307 °C. IR (KBr): 3493, 3305, 3236, 2932, 2871, 2807, 1739, 1696, 1548, 1481, 1376, 757 cm^{-1}. ^1H NMR(400.22 MHz, DMSO): 12.08 (1H, s, NH), 9.56 (1H, s, NH), 8.13 (1H, dd, *J* 1.4 and 8.4 Hz), 7.79 (1H, dt, *J* 1.6 and 7.6 Hz), 7.60-7.52 (6H, m), 7.45-7.40 (1H, m, Ph, 4-H), 7.37 (1H, dd, *J* 2.0 and 8.8 Hz, 6'-H), 7.15 (1H, d, *J* 8.8 Hz, 7'-H), 7.08 (1H, d, *J* 2.0 Hz, 4'-H), 1.77 (3H, CH_3). ^{13}C NMR(100.63 MHz, DMSO): 12.1 (CH_3), 51.9, 94.3, 109.2, 111.7, 112.9, 117.7, 119.8, 121.0, 123.4, 125.0, 125.2, 127.3, 129.5, 130.0, 131.8, 133.8, 138.7, 138.9, 140.2, 145.1, 153.2, 155.4, 156.3, 181.5. MS (EI, 70 eV) m/z= 480 (M^+, ^{35}Cl, 1), 419 (4), 417 (15), 268 (9), 173 (26), 77 (100). Anal. Calcd for $C_{27}H_{17}ClN_4O_3$ (480.9): C, 67.43; H, 3.56; N, 11.65%. Found: C, 67.37; H, 3.59; N, 11.70%.

[TMG·HCl][MImS]: ^1H NMR(400.22 MHz, DMSO): 8.09 (1H, s, C-H), 7.91 (2H, s, NH2), 7.32 (1H, s, C-H), 7.14 (1H, s, C-H), 3.75 (3H, s, >N-CH3), 2.91 (12H, s, 2 N(CH3)2). ^{13}C NMR(100.63 MHz, DMSO): 161.4, 137.6, 125.9, 121.9, 55.4 (>N-CH_3), 34.1 (-N(CH_3)2).

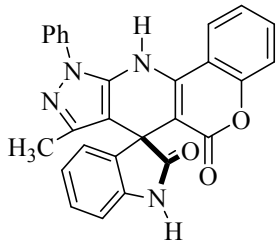
3-(5-Oxo-1,3-diphenyl-1*H*-pyrazol-4(5*H*)-ylidene)indolin-2-one (7b see the manuscript) Mp: 199-201 °C. IR (KBr): 3193 (N-H), 3062, 2872, 2740, 1684, 1631, 1596, 1498, 1393, 749, 698 cm^{-1}. ^1H NMR(400.22 MHz, DMSO): 11.31 (1H, s, NH), 11.22 (1H, s, NH), 10.92 (1H, s, OH), 7.73 (2H, d, *J* 7.2 Hz), 7.58-7.10 (17H, m), 7.03 (2H, t, *J* 7.2 Hz), 6.96 (1H, d, *J* 8.0 Hz), 6.72 (2H, d, *J* 6.4 Hz), 6.60 (1H, d, *J* 6.8 Hz), 6.37 (1H, t, *J* 7.4 Hz). ^{13}C NMR(100.63 MHz, DMSO): 50.8, 96.1, 106.2, 110.4, 118.9, 119.6, 119.7, 121.9, 122.1, 122.2, 125.2, 126.3, 126.4, 127.1, 127.5, 127.6, 128.0, 128.3, 128.9, 129.0, 129.1, 129.2, 129.3, 129.4, 129.5, 130.0, 130.3, 134.9, 137.2, 138.0, 142.5, 149.8, 151.4, 151.8, 161.9, 183.8. MS (EI, 70 eV) m/z= 366 (M^++ 1, 15), 365 (M^+, 46), 336 (100), 318 (9), 236 (60). 203 (17), 103 (31), 77 (89).

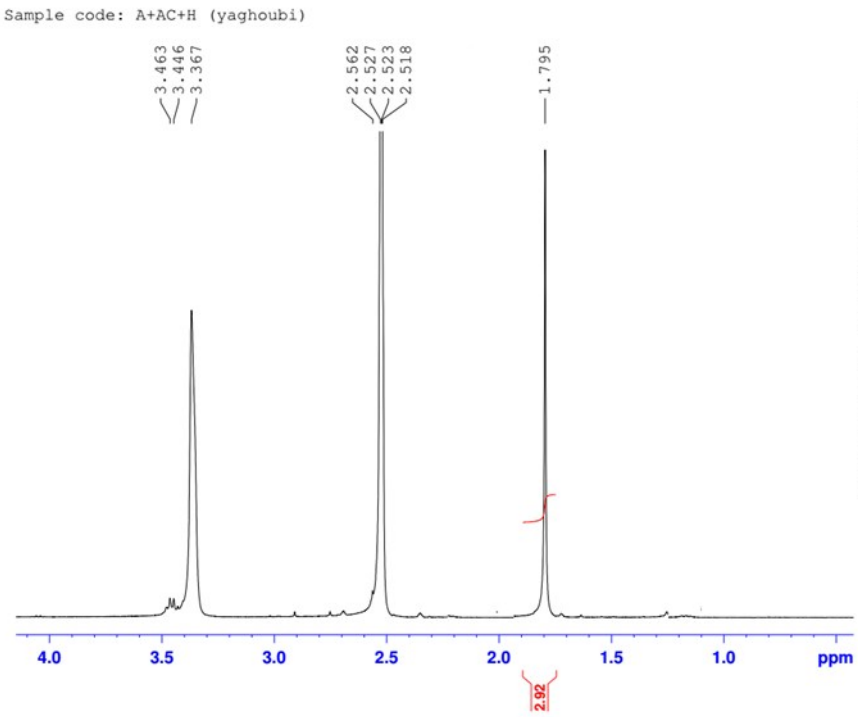
S$_1$ The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2′-dione in DMSO-d_6 (**4a**)





S$_1$ The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2′-dione in DMSO-d_6 (**4a**)



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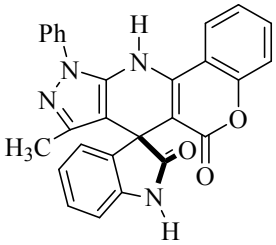
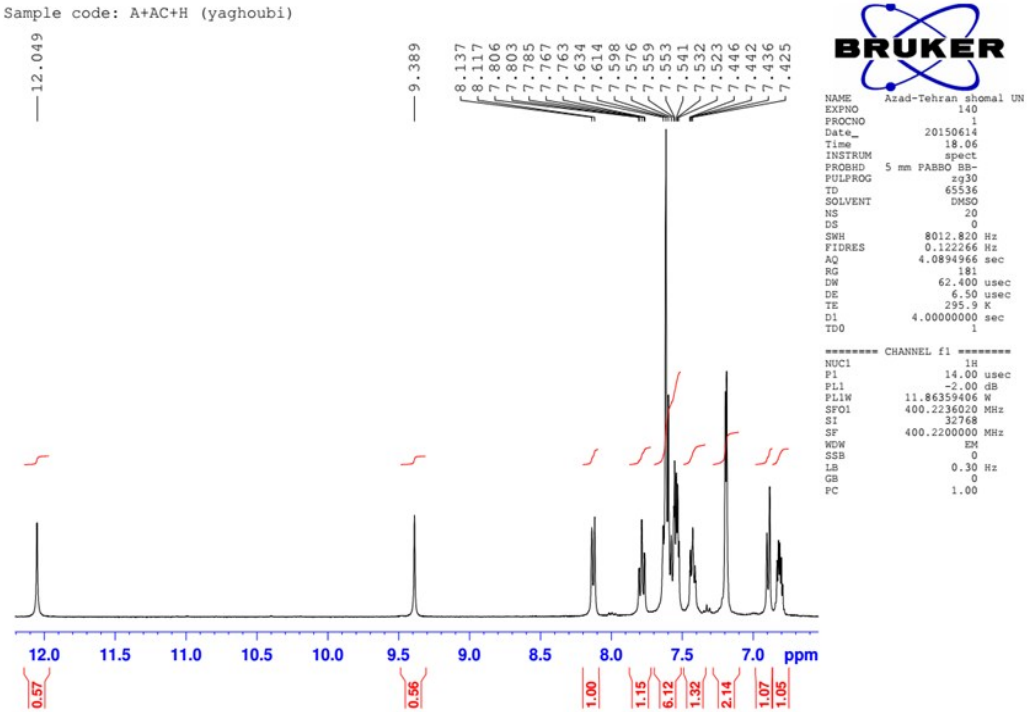
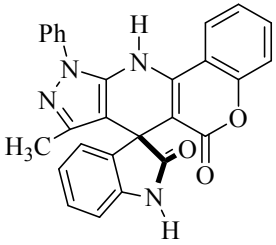
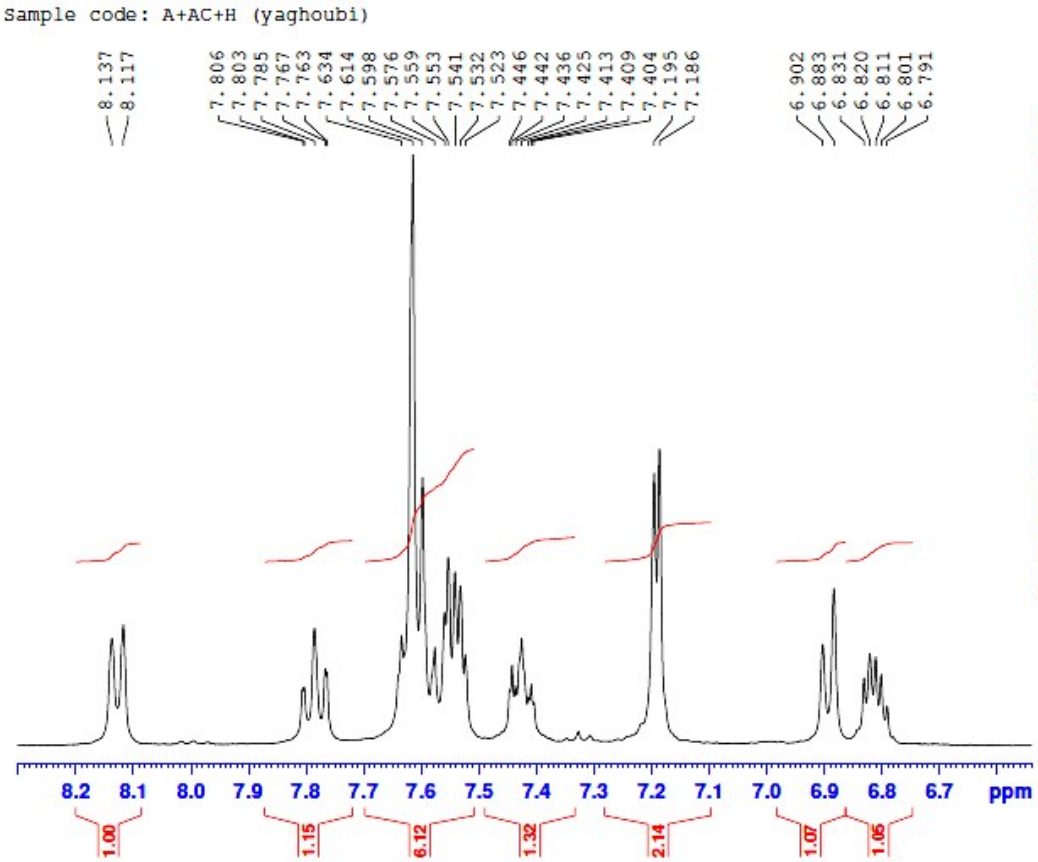
S1 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2′-dione in DMSO-d_6 (**4a**)





S$_1$ The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2′-dione in DMSO-d_6 (**4a**)



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FIDRES               0.122266 Hz
AQ                   4.0894966 sec
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DW                     62.400 usec
DE                      6.50 usec
TE                     295.9 K
D1                 4.00000000 sec
TD0                         1

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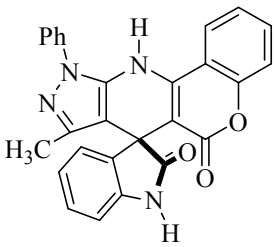
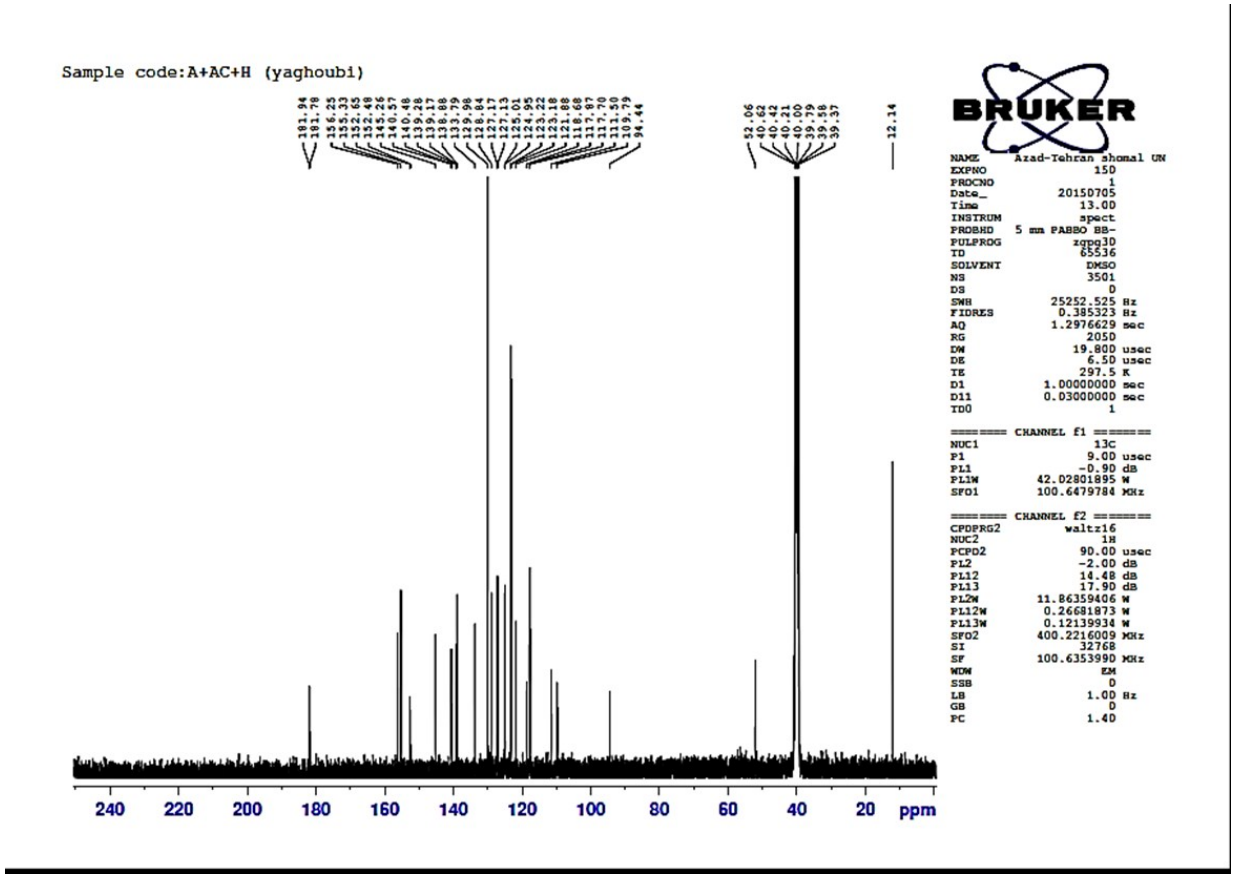
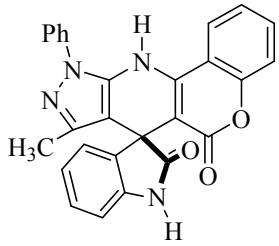
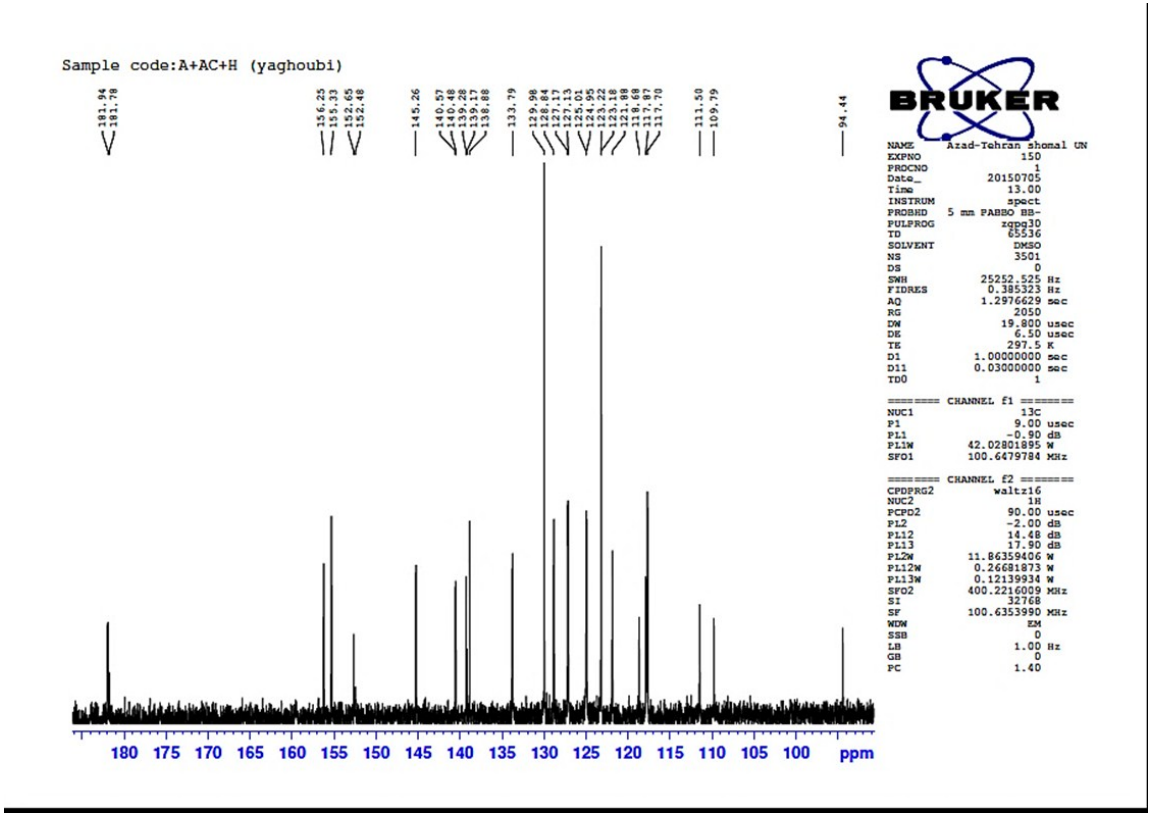
S$_2$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2′-dione in DMSO-d_6 (**4a**)



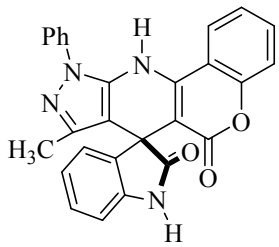


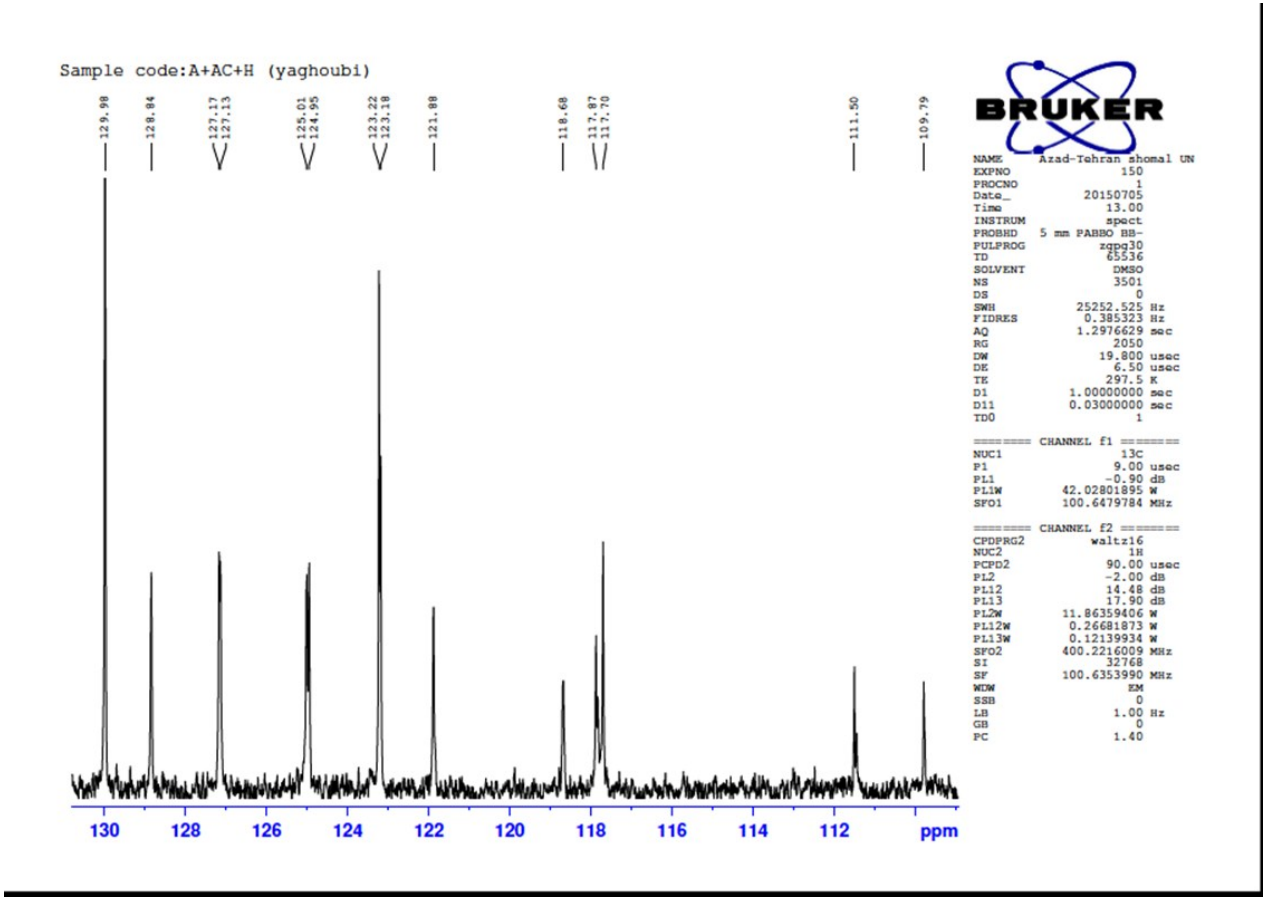
S$_2$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2′-dione in DMSO-d_6 (**4a**)



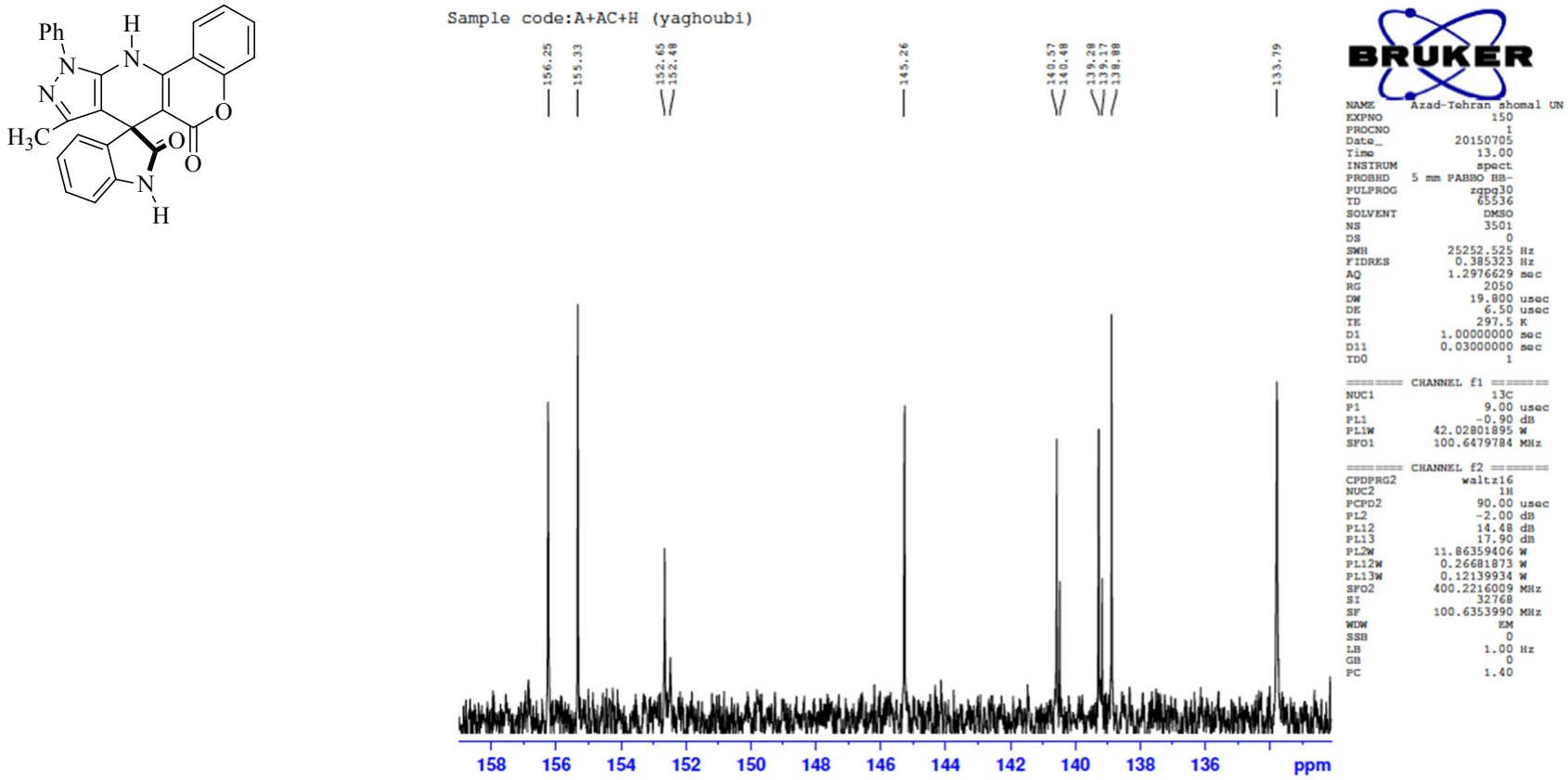


S$_2$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2′-dione in DMSO-d_6 (**4a**)

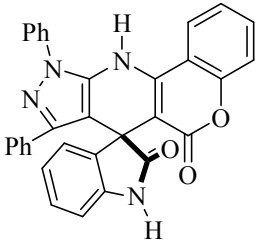


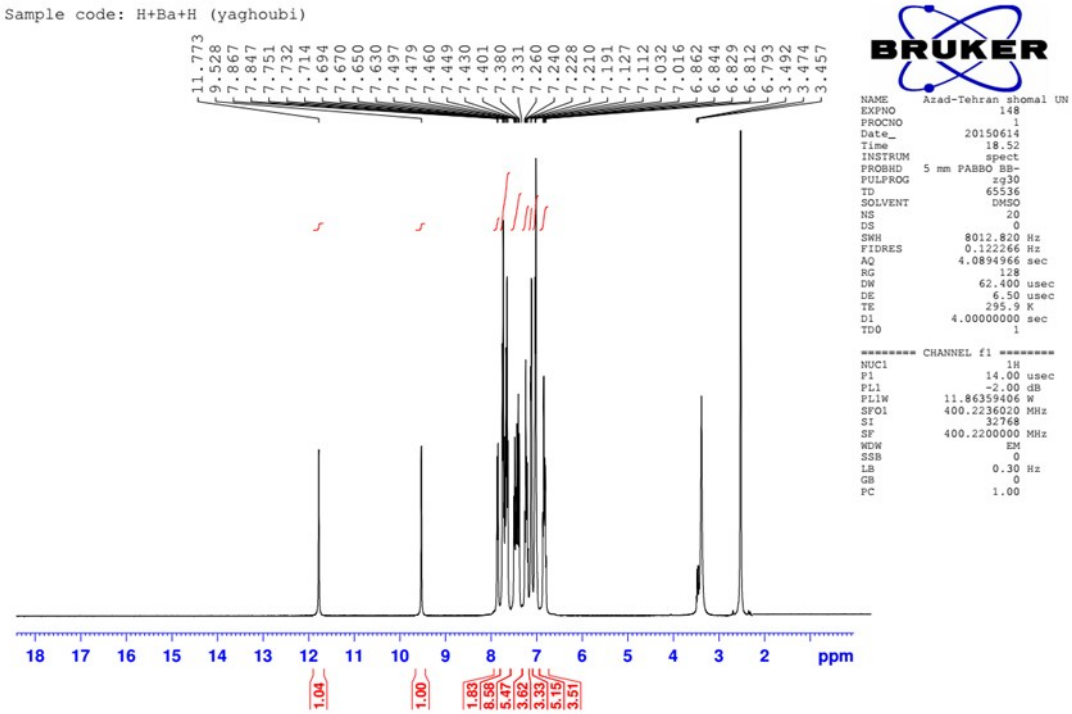


S$_2$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-1-phenyl-5,2′-dione in DMSO-d_6 (**4a**)

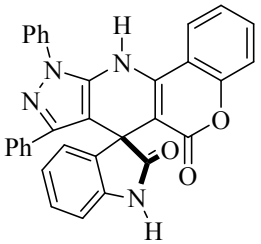


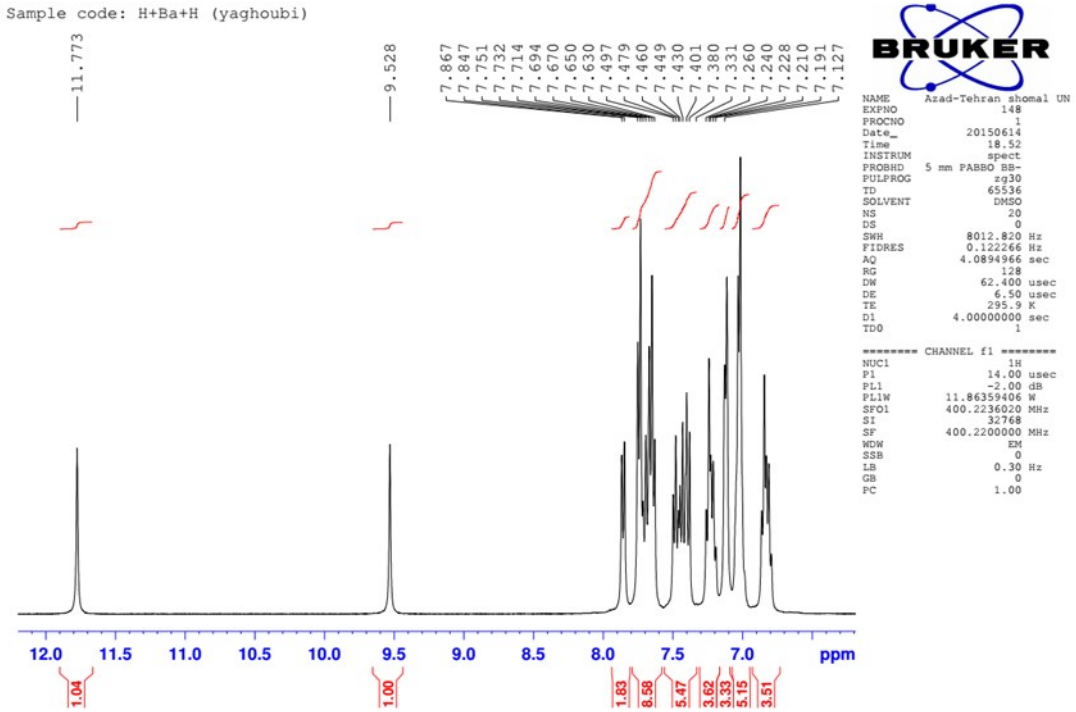
S$_3$ The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1,3-diphenyl-5,2′-dione (**4b**)



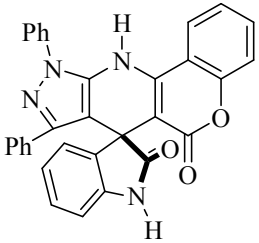


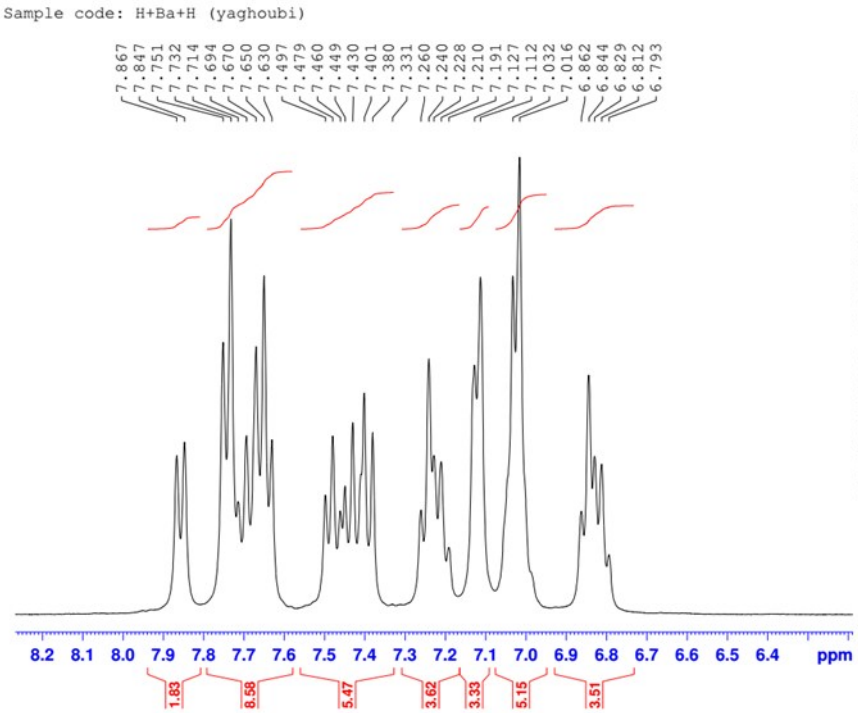
S$_3$ The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1,3-diphenyl-5,2′-dione (**4b**)





S$_3$ The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1,3-diphenyl-5,2′-dione (**4b**)



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TD                      65536
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FIDRES               0.122266 Hz
AQ                 4.0894966 sec
RG                        128
DW                     62.400 usec
DE                       6.50 usec
TE                      295.9 K
D1                 4.00000000 sec
TD0                         1

======== CHANNEL f1 ========
NUC1                       1H
P1                      14.00 usec
PL1                     -2.00 dB
PL1W               11.86359406 W
SFO1               400.2236020 MHz
SI                       32768
SF                 400.2200000 MHz
WDW                         EM
SSB                          0
LB                        0.30 Hz
GB                           0
PC                        1.00
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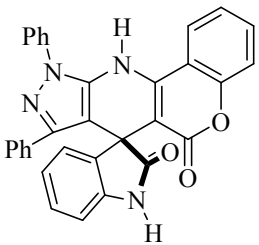
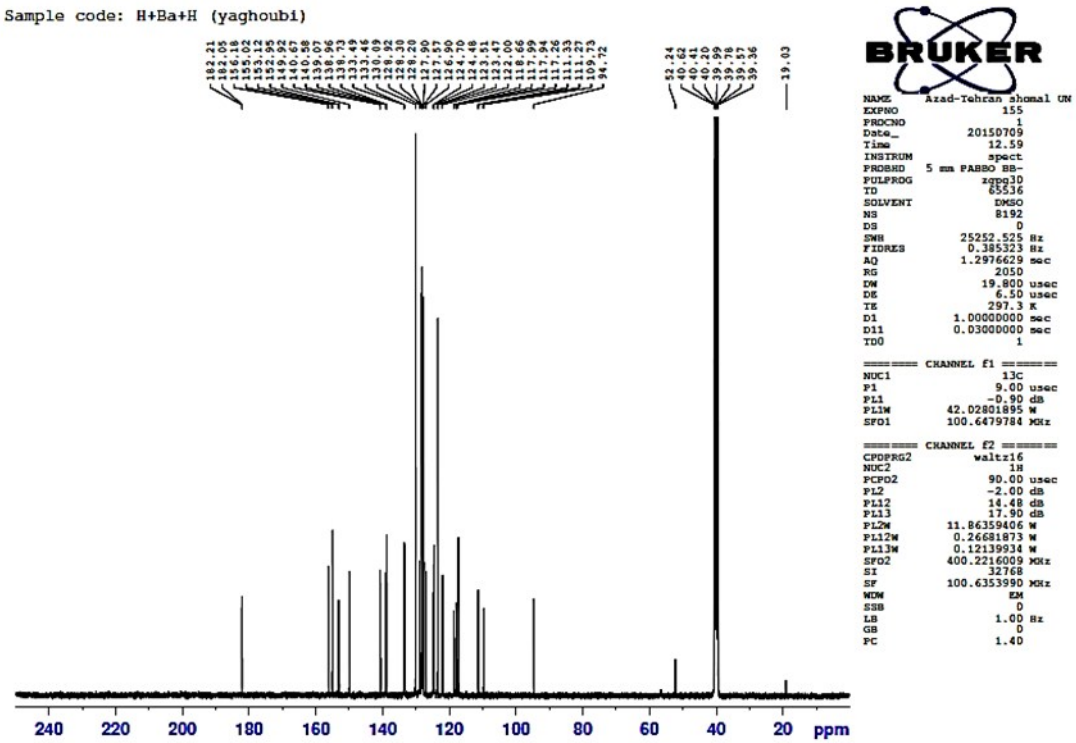
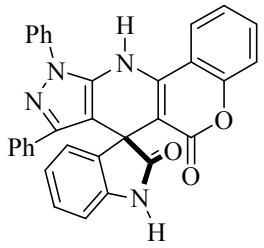
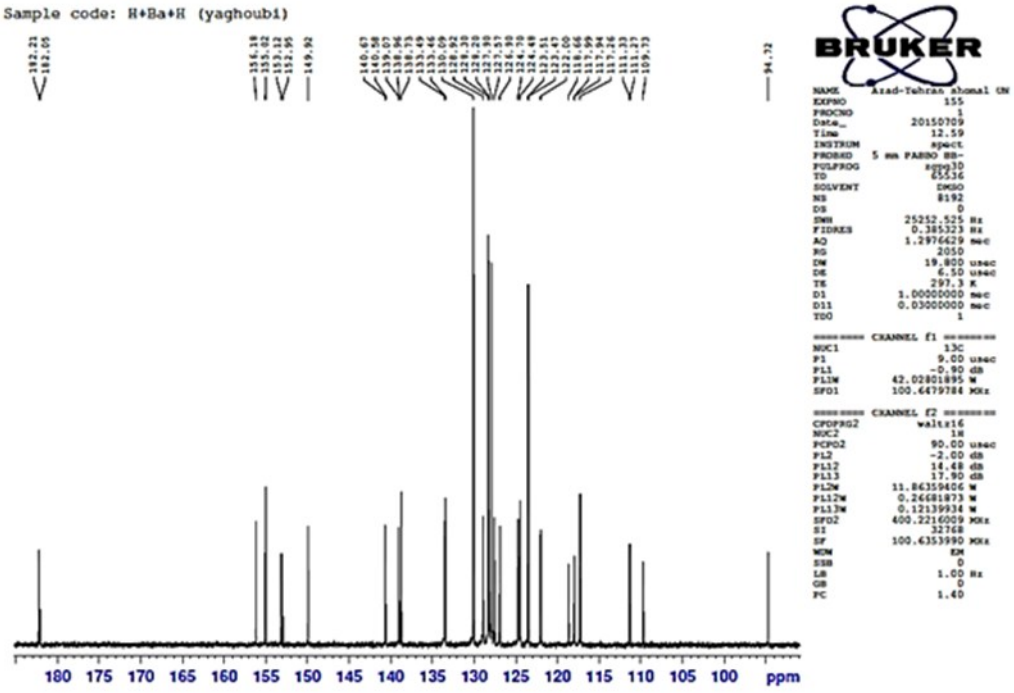
S$_4$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1,3-diphenyl-5,2′-dione in DMSO-d_6 (**4b**)



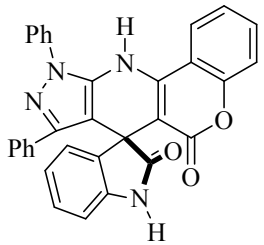


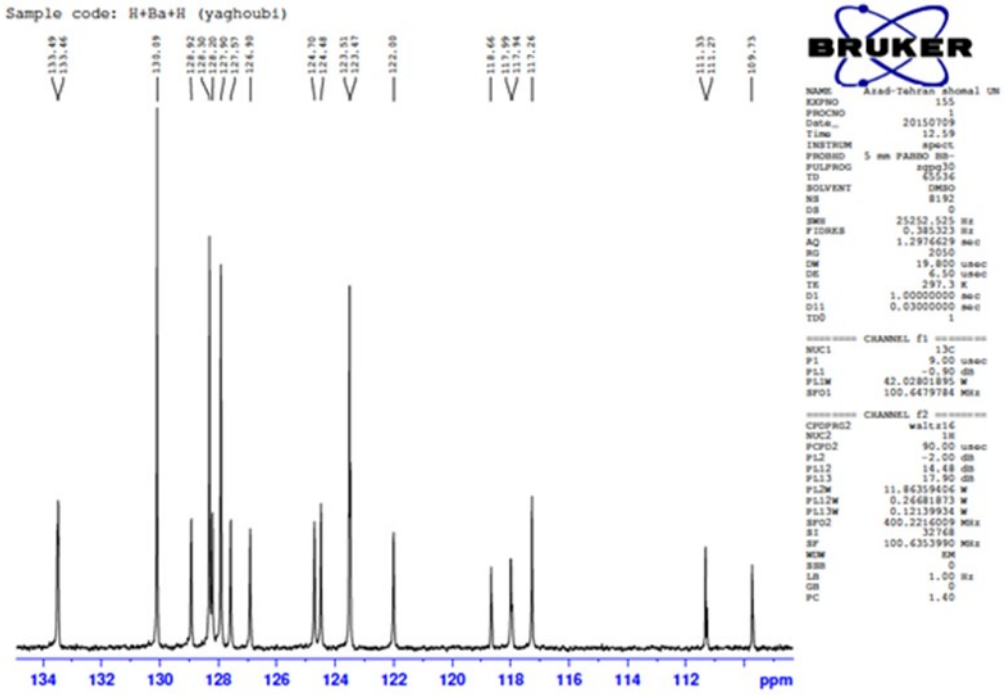
S$_4$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1,3-diphenyl-5,2′-dione in DMSO-d_6 (**4b**)



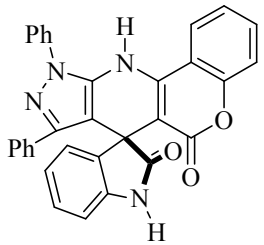


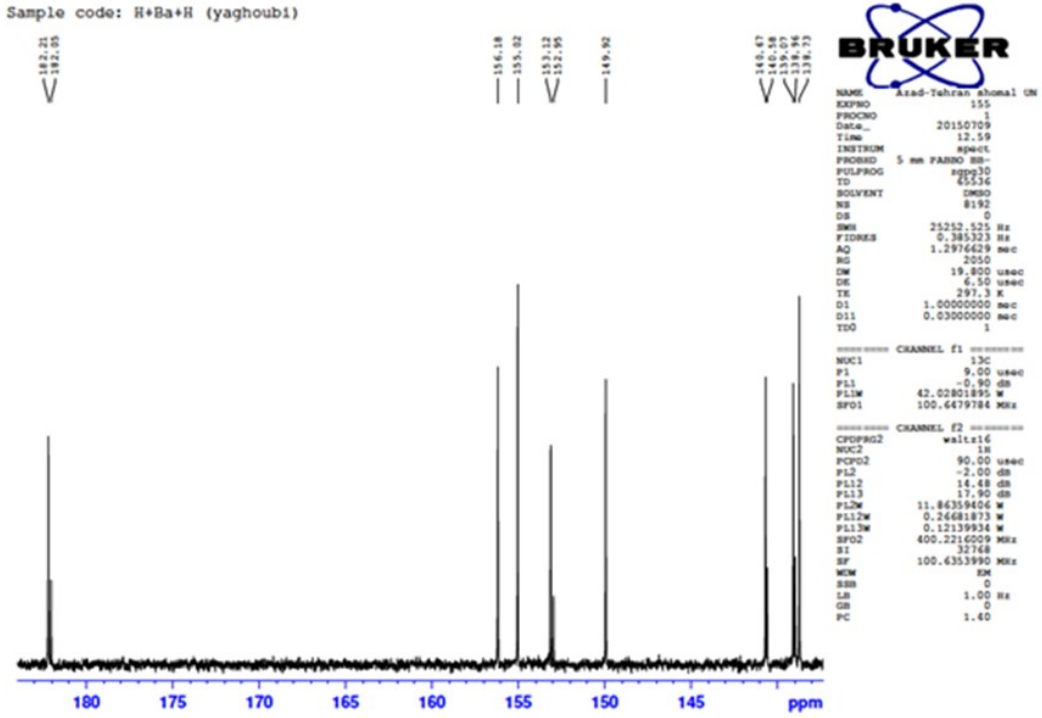
S$_4$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1,3-diphenyl-5,2′-dione in DMSO-d_6 (**4b**)



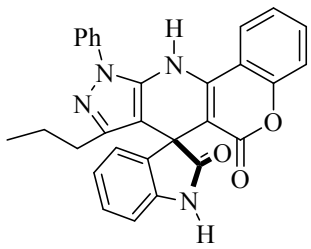


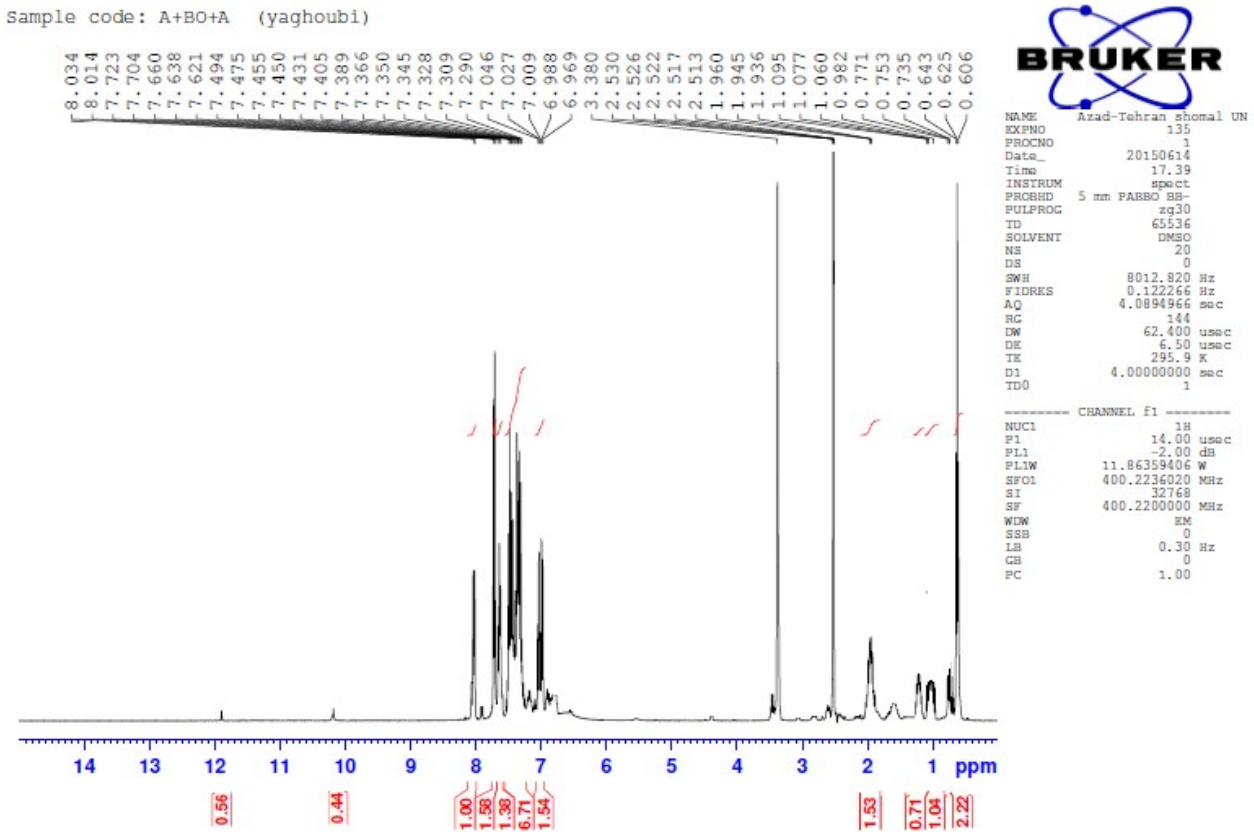
S$_4$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1,3-diphenyl-5,2′-dione in DMSO-d_6 (**4b**)



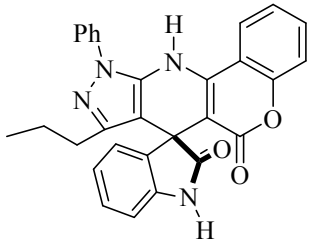


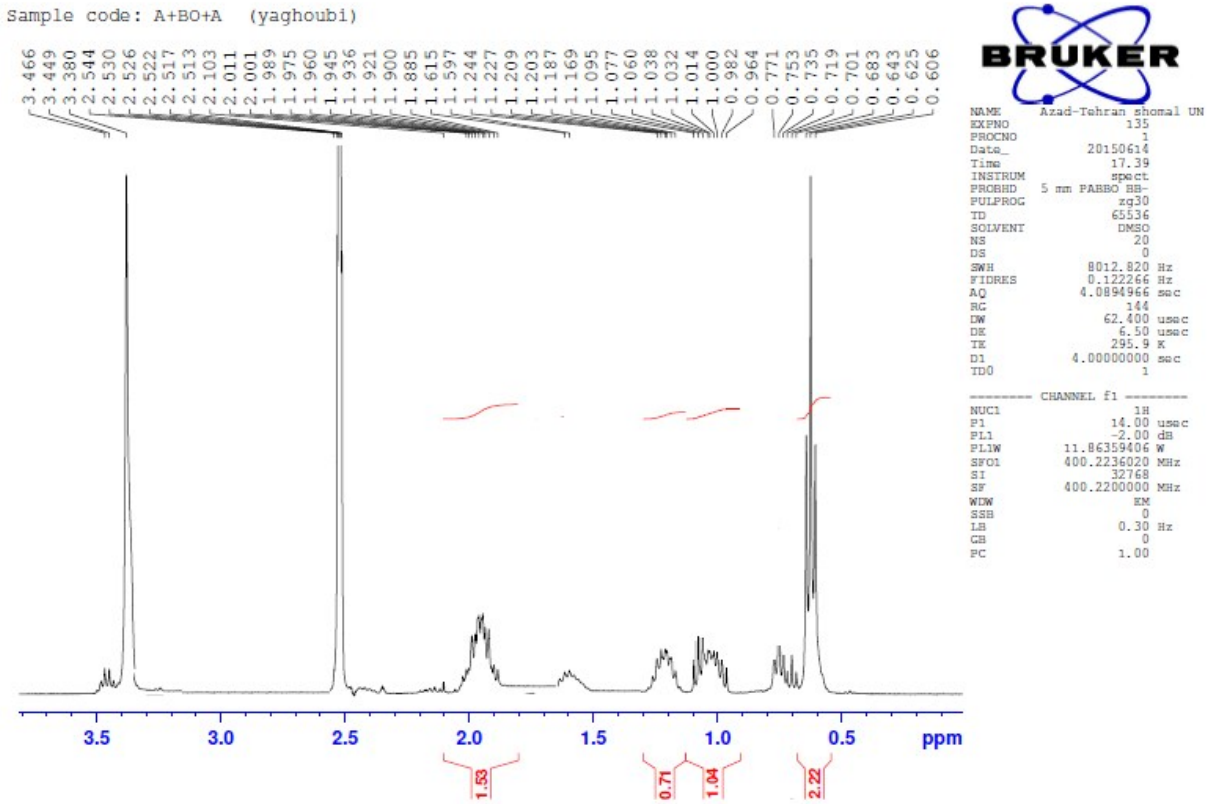
S5 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1-phenyl-3-propyl-5,2'-dione DMSO-d_6 (**4c**)



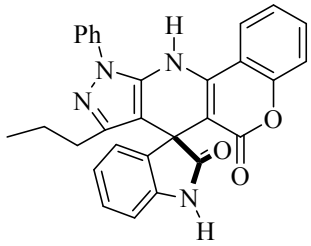


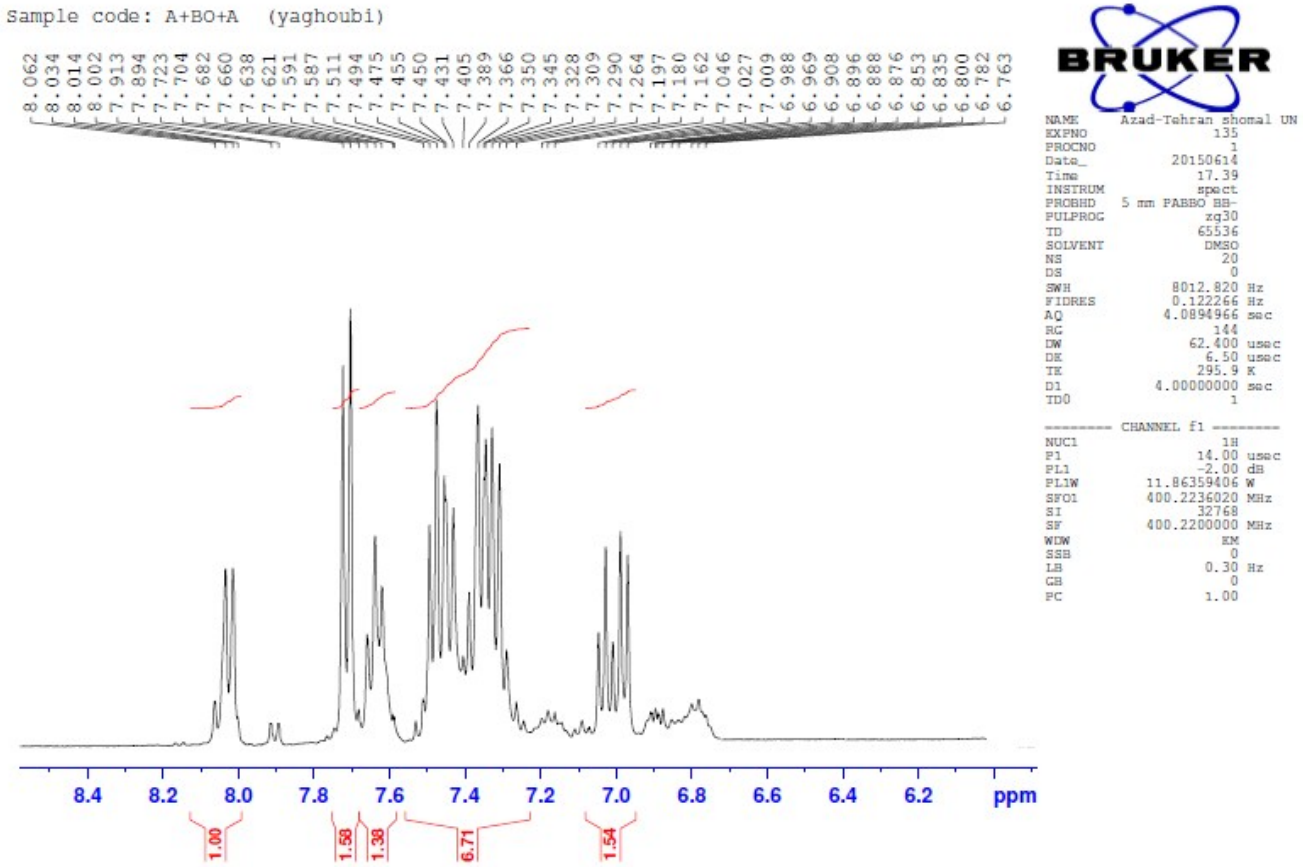
S$_5$ The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1-phenyl-3-propyl-5,2'-dione DMSO-d_6 (**4c**)



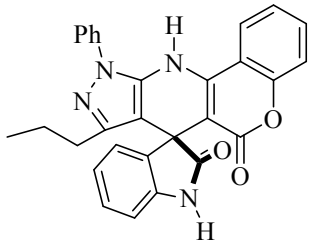


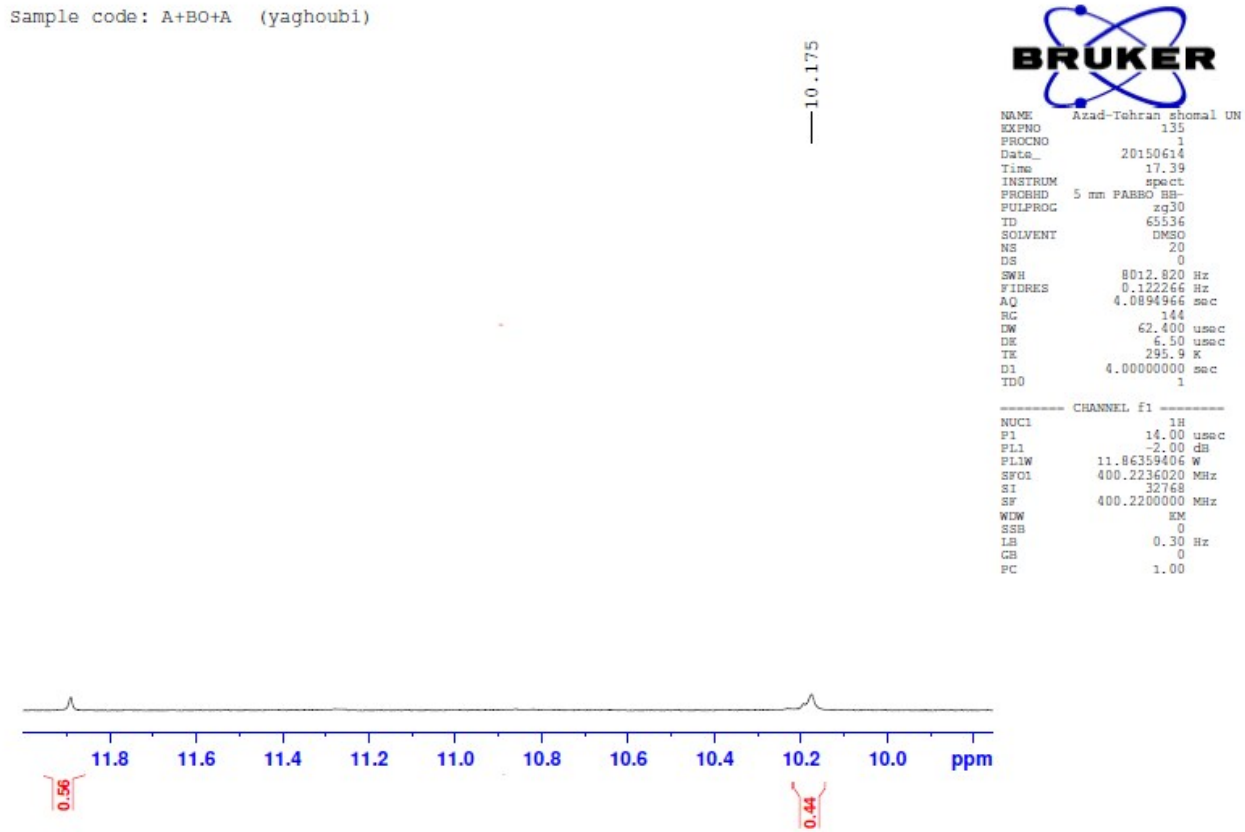
S5 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1-phenyl-3-propyl-5,2'-dione DMSO-d_6 (**4c**)



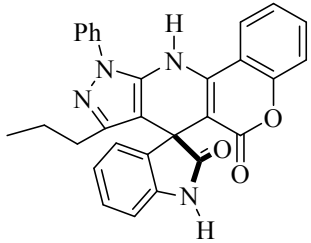


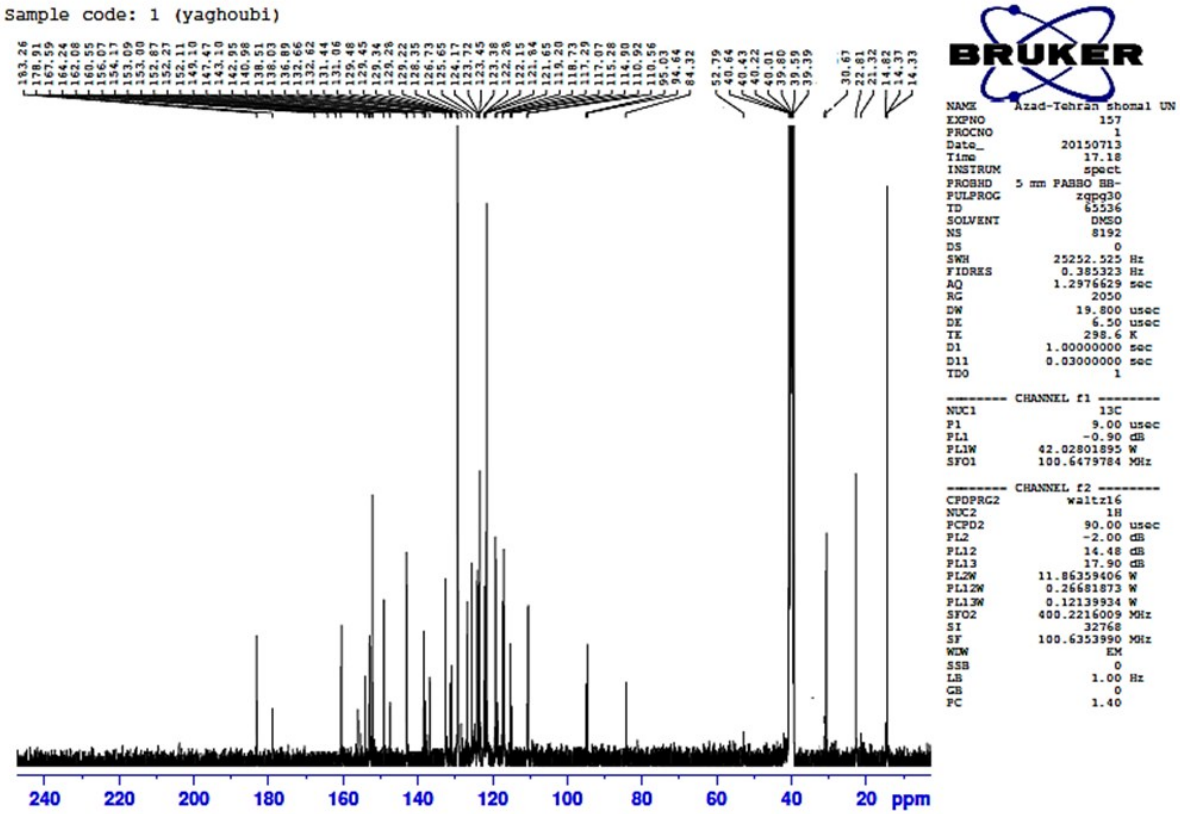
S5 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1-phenyl-3-propyl-5,2′-dione DMSO-d_6 (**4c**)



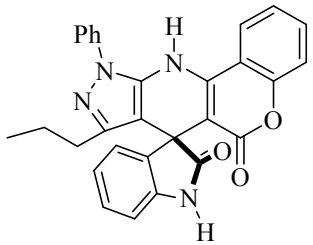


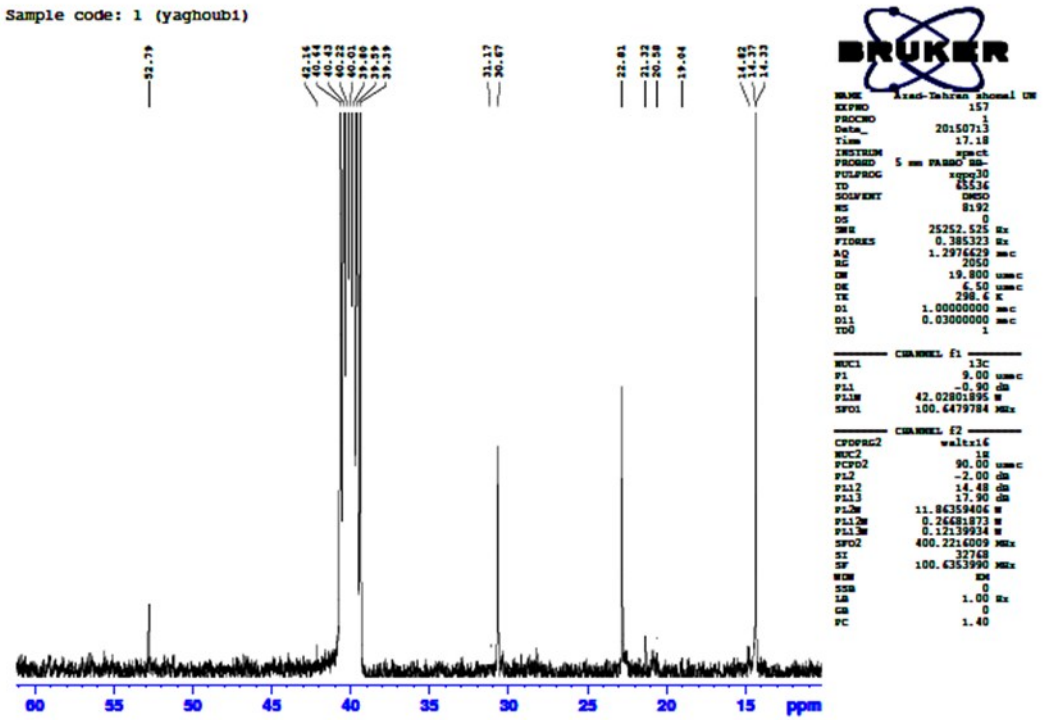
S$_6$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1-phenyl-3-propyl-5,2′-dione DMSO-d_6 (**4c**)



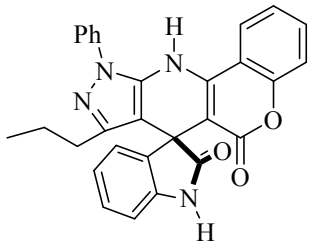


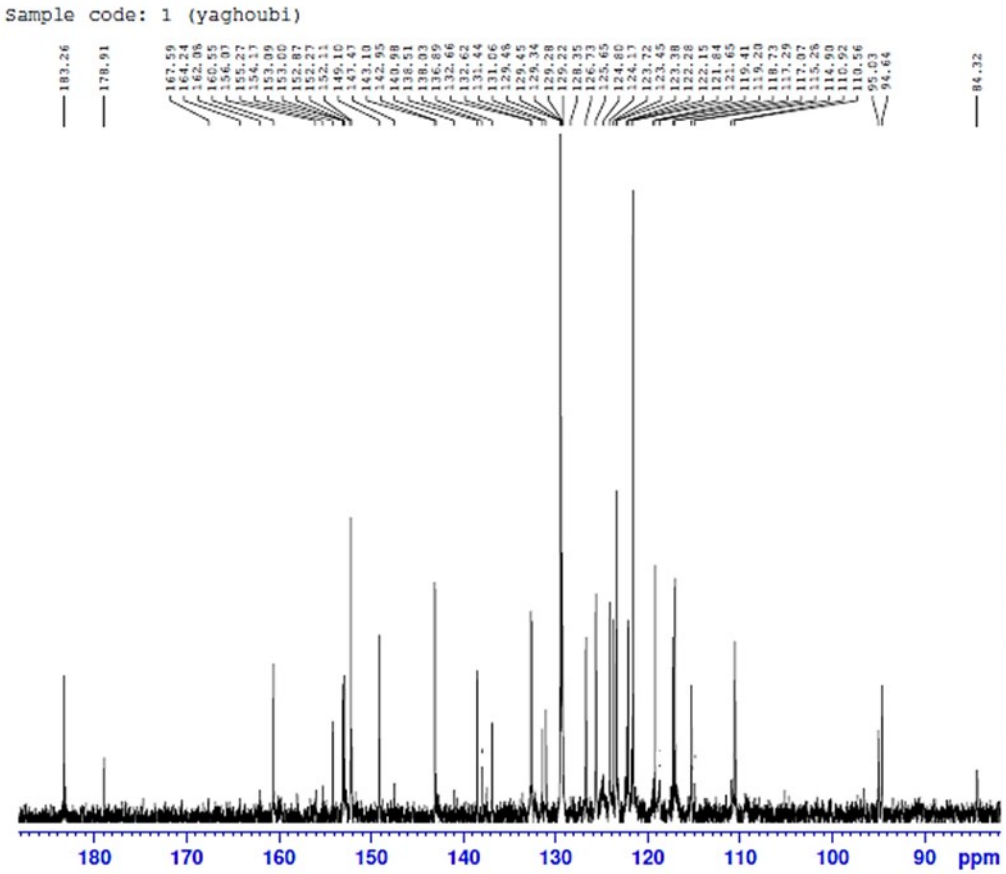
S$_6$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1-phenyl-3-propyl-5,2′-dione DMSO-d_6 (**4c**)





S6 The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-1-phenyl-3-propyl-5,2′-dione DMSO-d_6 (**4c**)



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PL12                     14.48 dB
PL13                     17.90 dB
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PL13W               0.12139934 W
SFO2               400.2216009 MHz
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WDW                         EM
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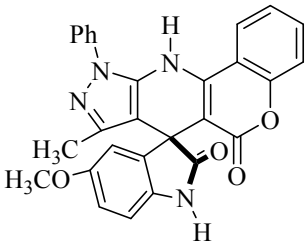
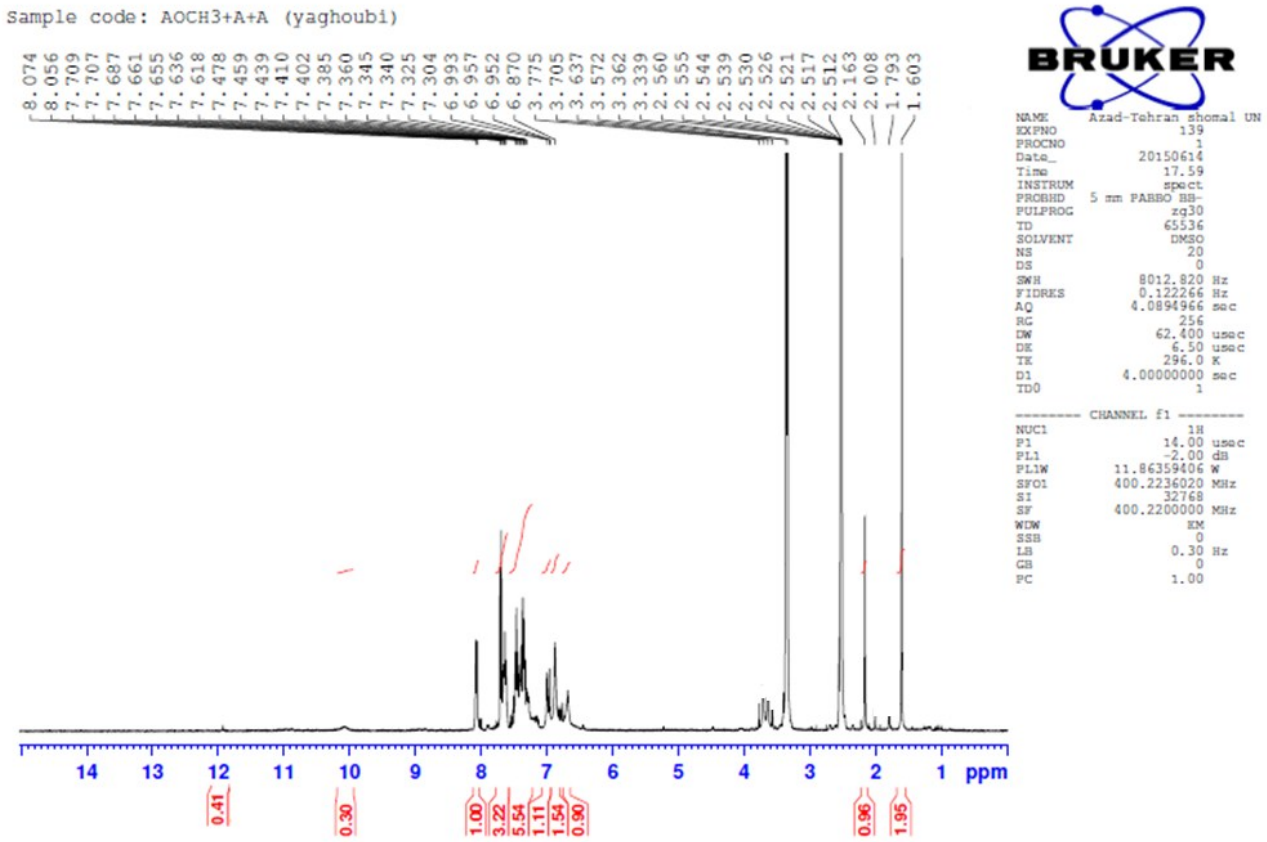
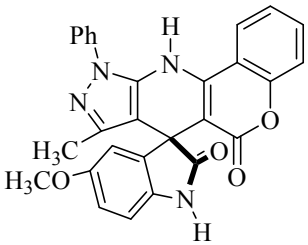
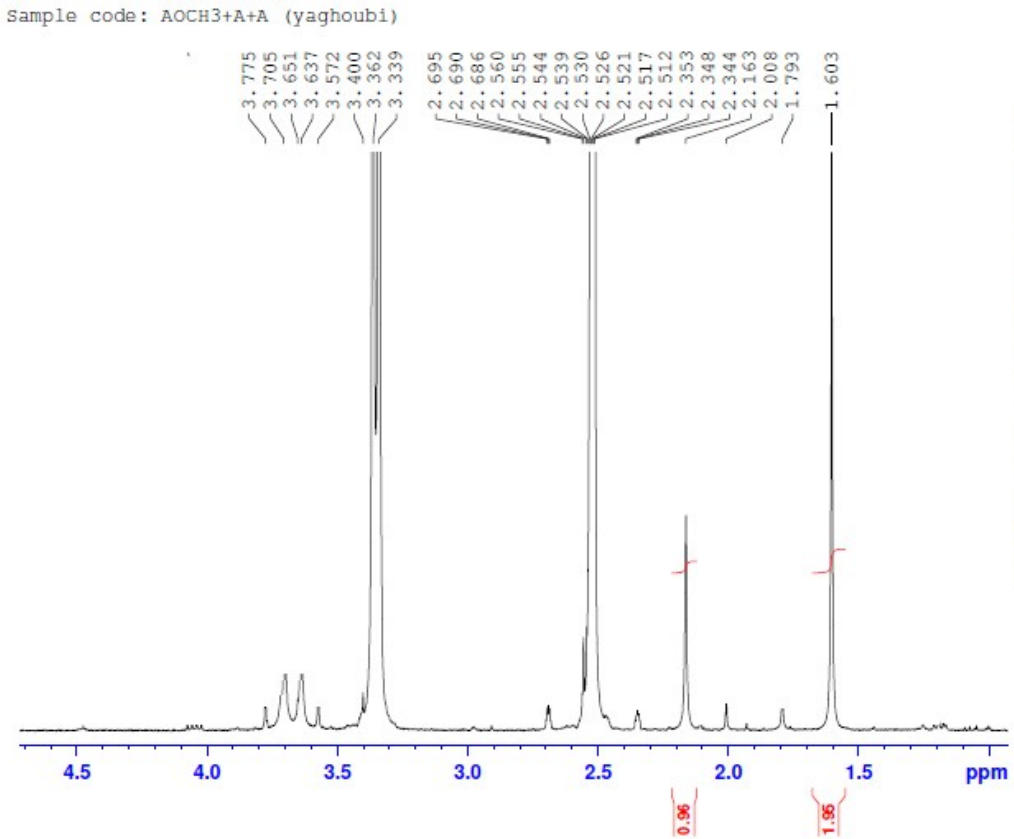
S7 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-5′-methoxy-1-phenyl-5,2′-dione in DMSO-d_6 (**4d**)





S7 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-5′-methoxy-1-phenyl-5,2′-dione in DMSO-d_6 (**4d**)



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TD                       65536
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DS                           0
SWH                   8012.820 Hz
FIDRES                0.122266 Hz
AQ                   4.0894966 sec
RG                         256
DW                      62.400 usec
DE                       6.50 usec
TE                      296.0 K
D1                  4.00000000 sec
TD0                         1

======== CHANNEL f1 ========
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P1                      14.00 usec
PL1                     -2.00 dB
PL1W               11.86359406 W
SFO1               400.2236020 MHz
SI                       32768
SF                 400.2200000 MHz
WDW                         EM
SSB                          0
LB                        0.30 Hz
GB                           0
PC                        1.00
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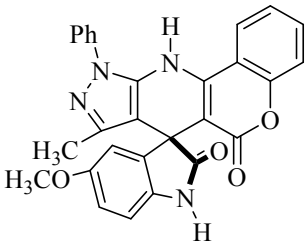
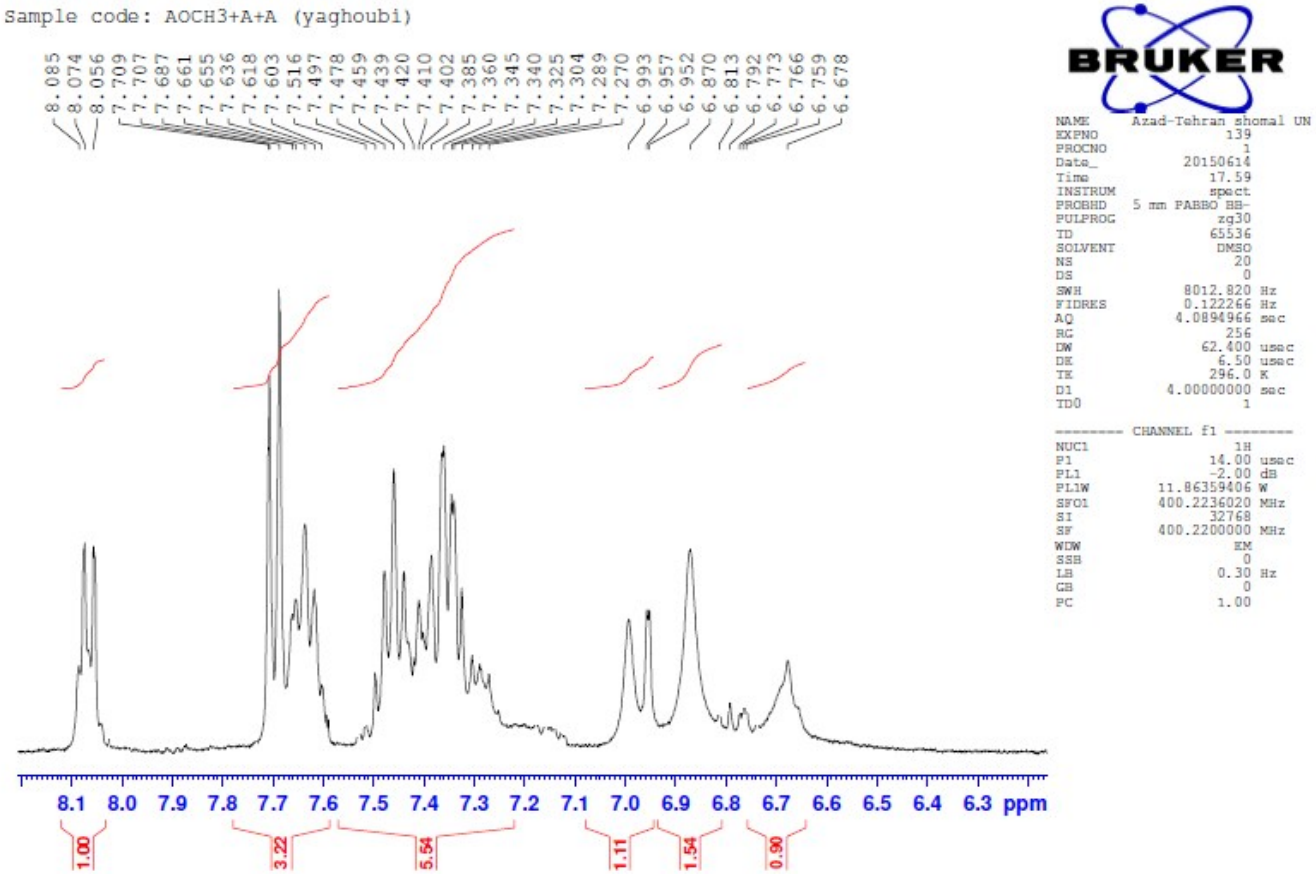
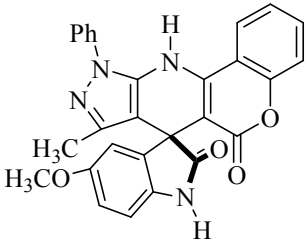
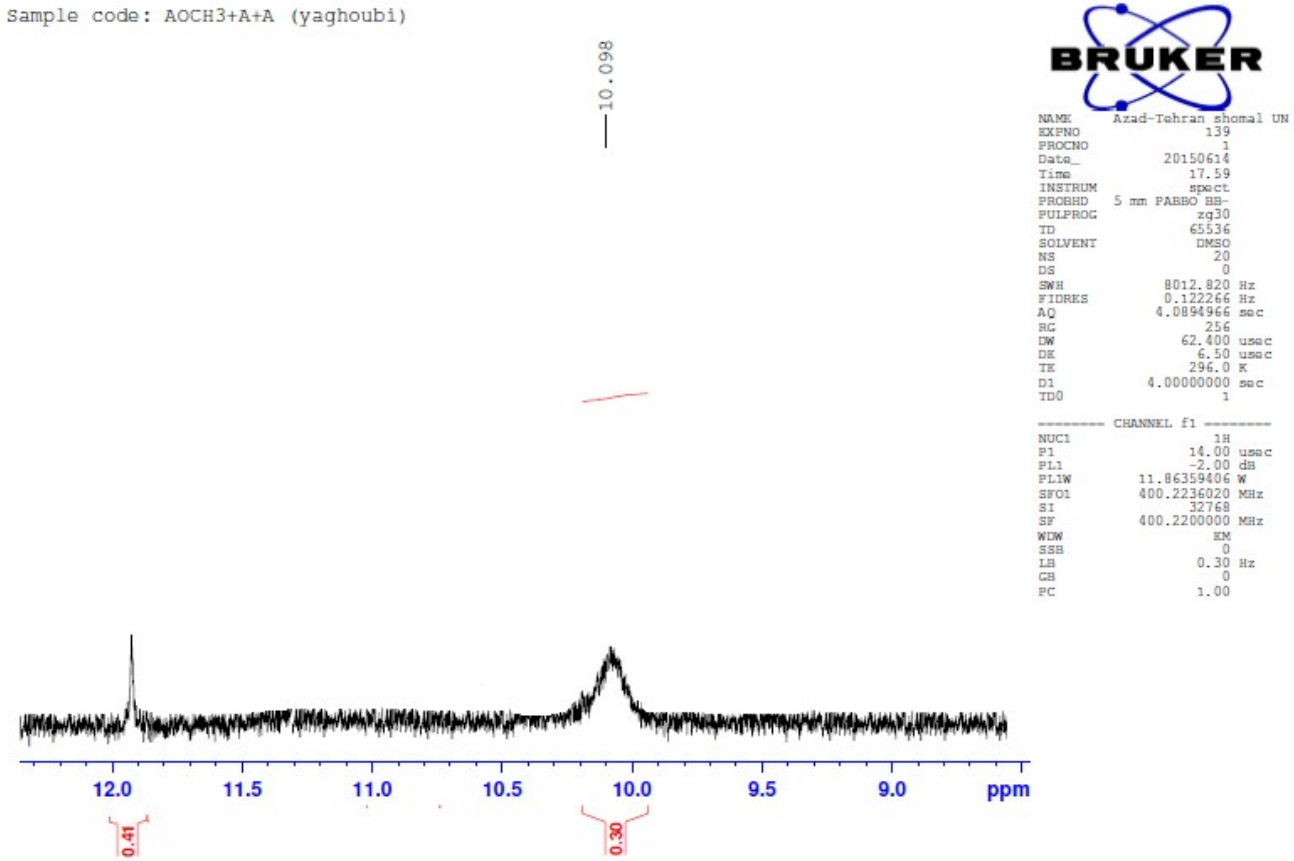
S7 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-5′-methoxy-1-phenyl-5,2′-dione in DMSO-d_6 (**4d**)



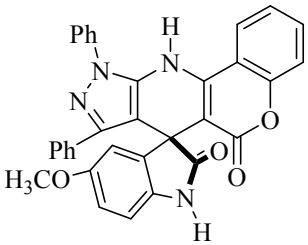


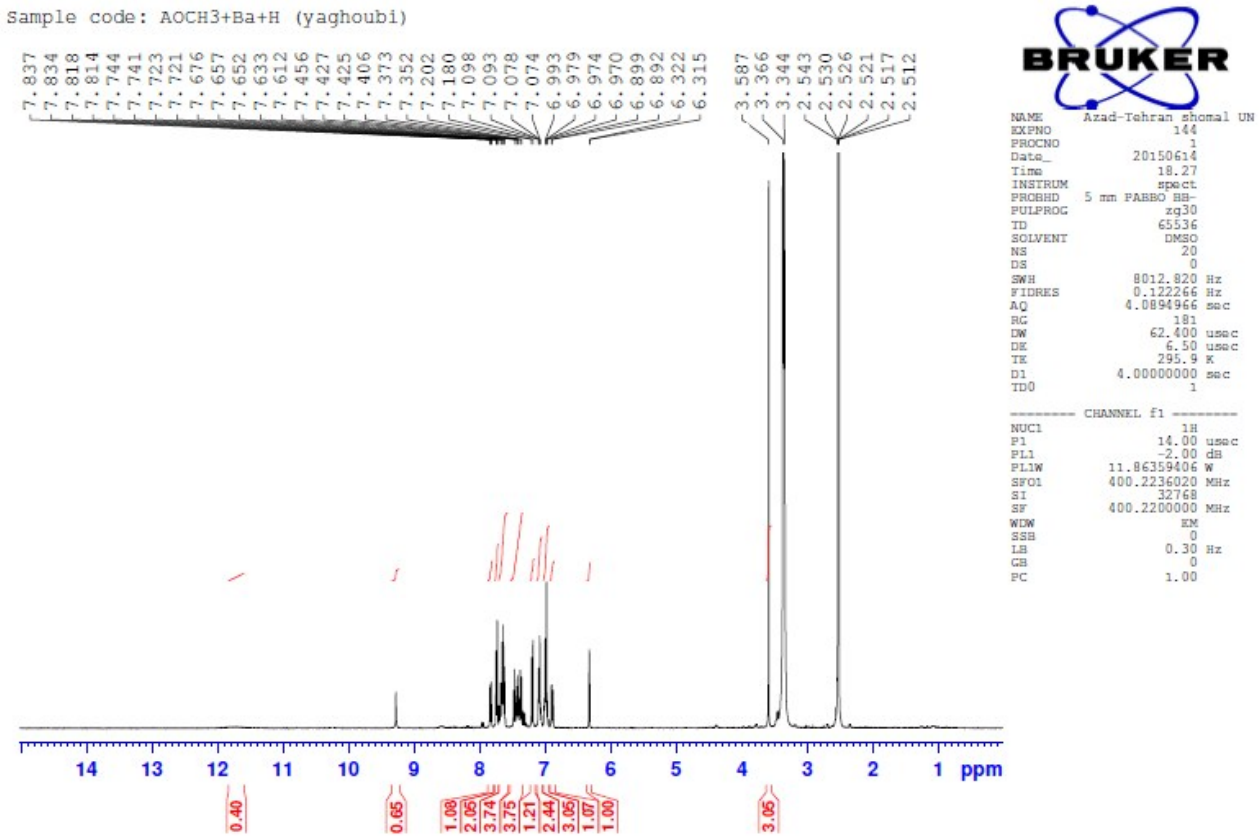
S7 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-3-methyl-5′-methoxy-1-phenyl-5,2′-dione in DMSO-d_6 (**4d**)



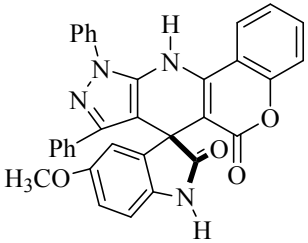


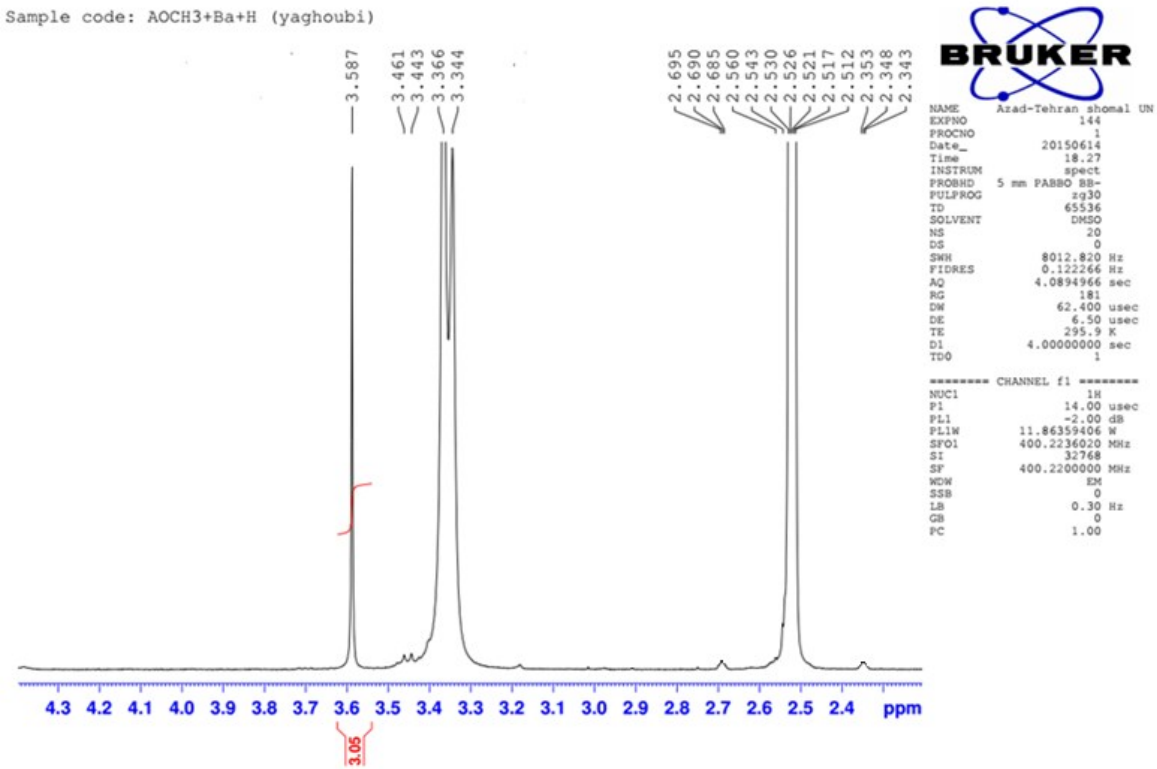
S8 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1,3-diphenyl-5,2′-dione (**4e**)



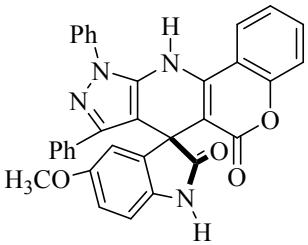


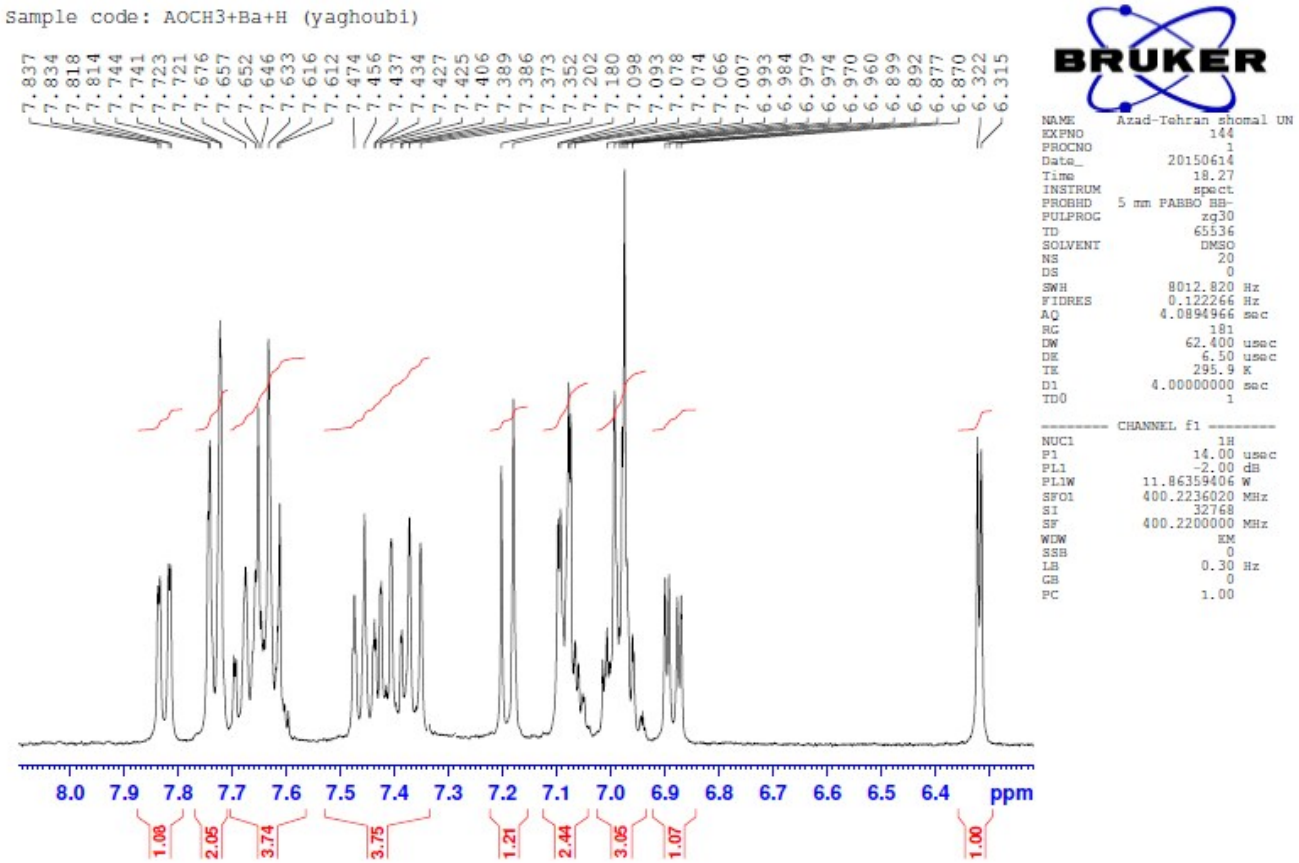
S8 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1,3-diphenyl-5,2′-dione (**4e**)



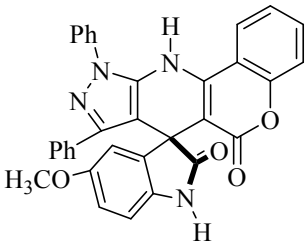


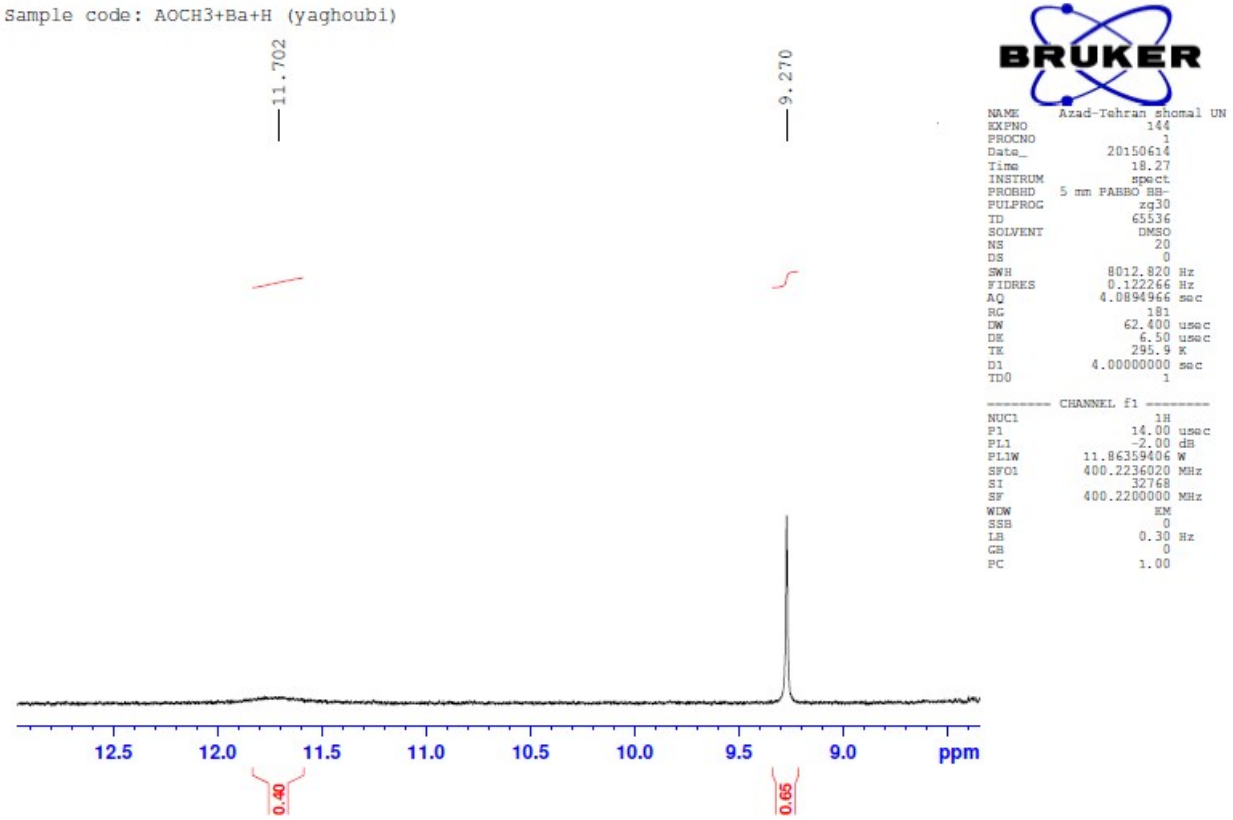
S8 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1,3-diphenyl-5,2′-dione (**4e**)



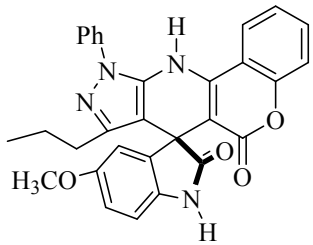


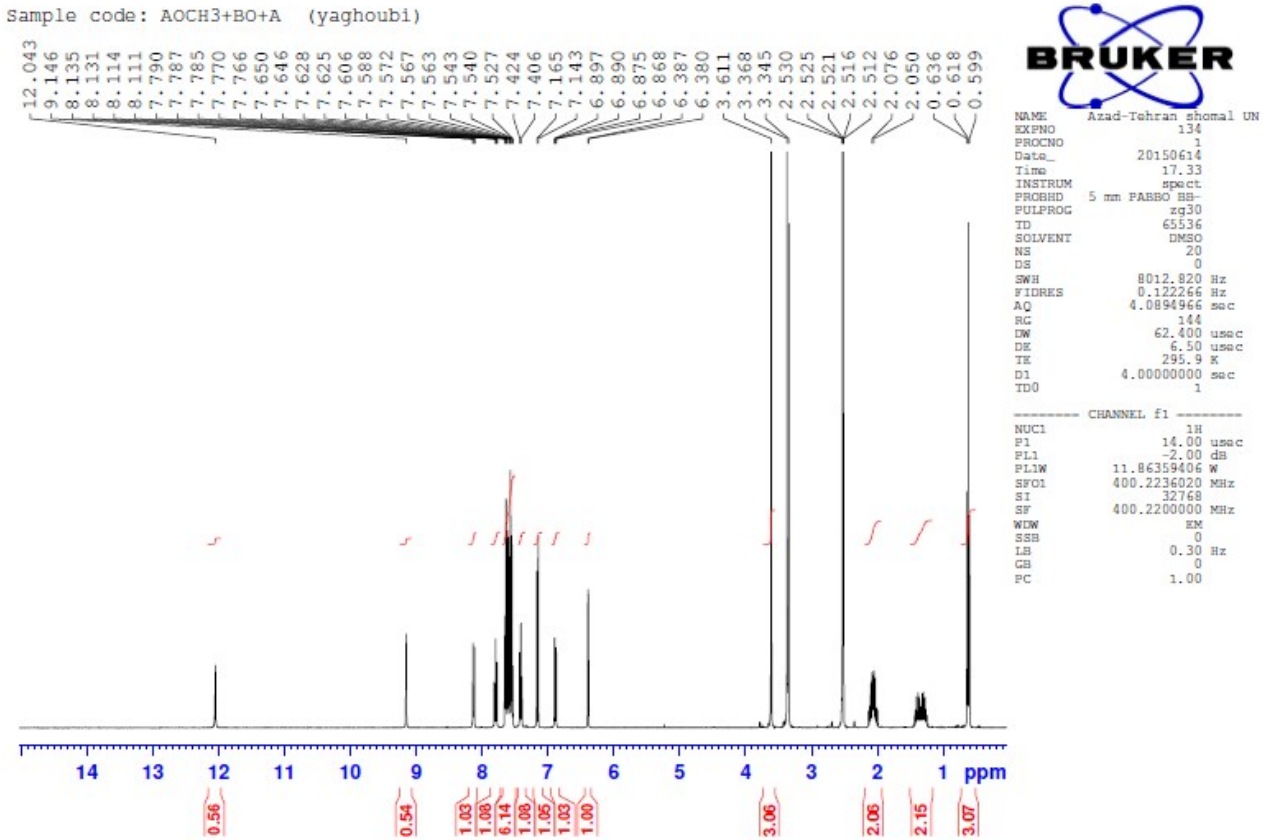
S8 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1,3-diphenyl-5,2′-dione (**4e**)



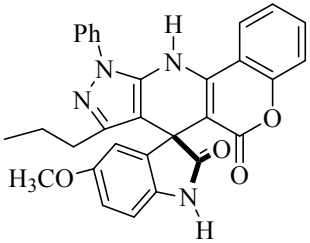


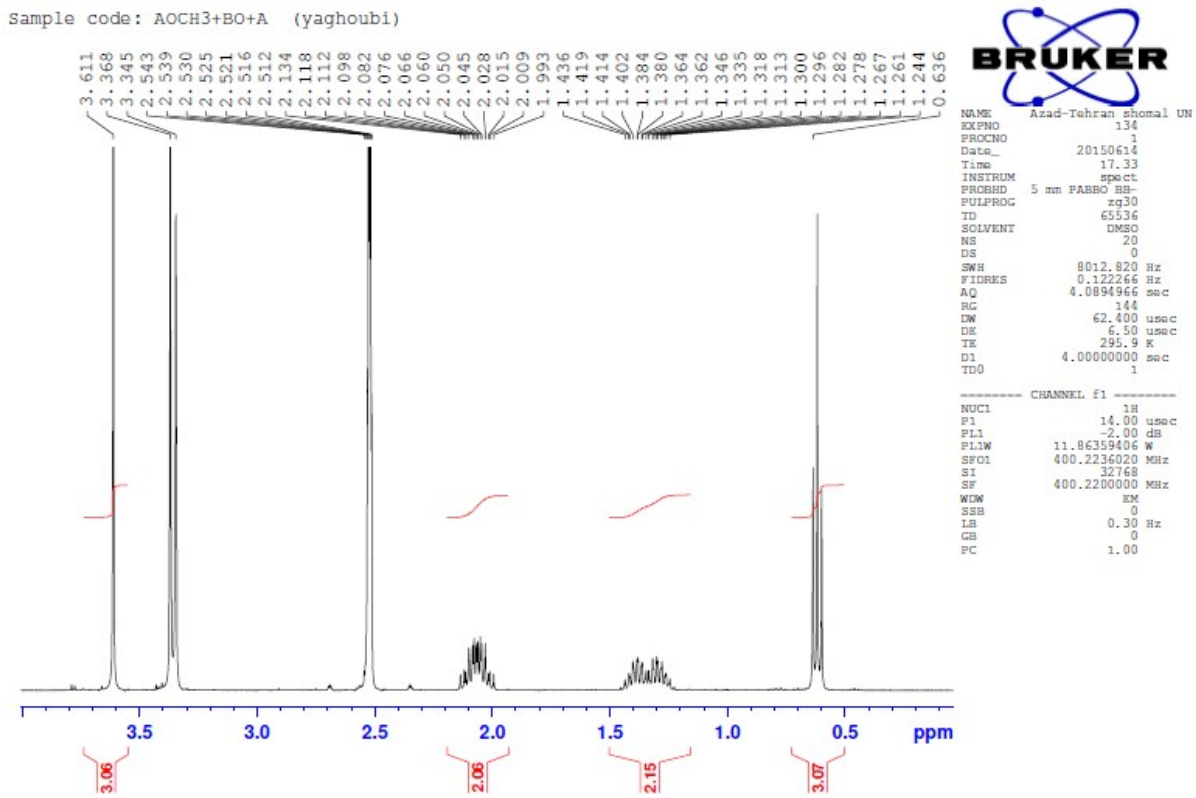
S9 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-c]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1-phenyl-3-propyl-5,2′-dione (**4f**)





S9 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-c]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1-phenyl-3-propyl-5,2′-dione (**4f**)



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TD                      65536
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SWH                  8012.820 Hz
FIDRES               0.122266 Hz
AQ                 4.0894966 sec
RG                        144
DW                     62.400 usec
DE                       6.50 usec
TE                      295.9 K
D1                 4.00000000 sec
TD0                         1

======== CHANNEL f1 ========
NUC1                       1H
P1                      14.00 usec
PL1                     -2.00 dB
PL1W                11.86359406 W
SFO1                400.2236020 MHz
SI                        32768
SF                  400.2200000 MHz
WDW                        EM
SSB                         0
LB                       0.30 Hz
GB                          0
PC                       1.00
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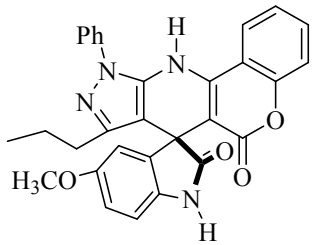
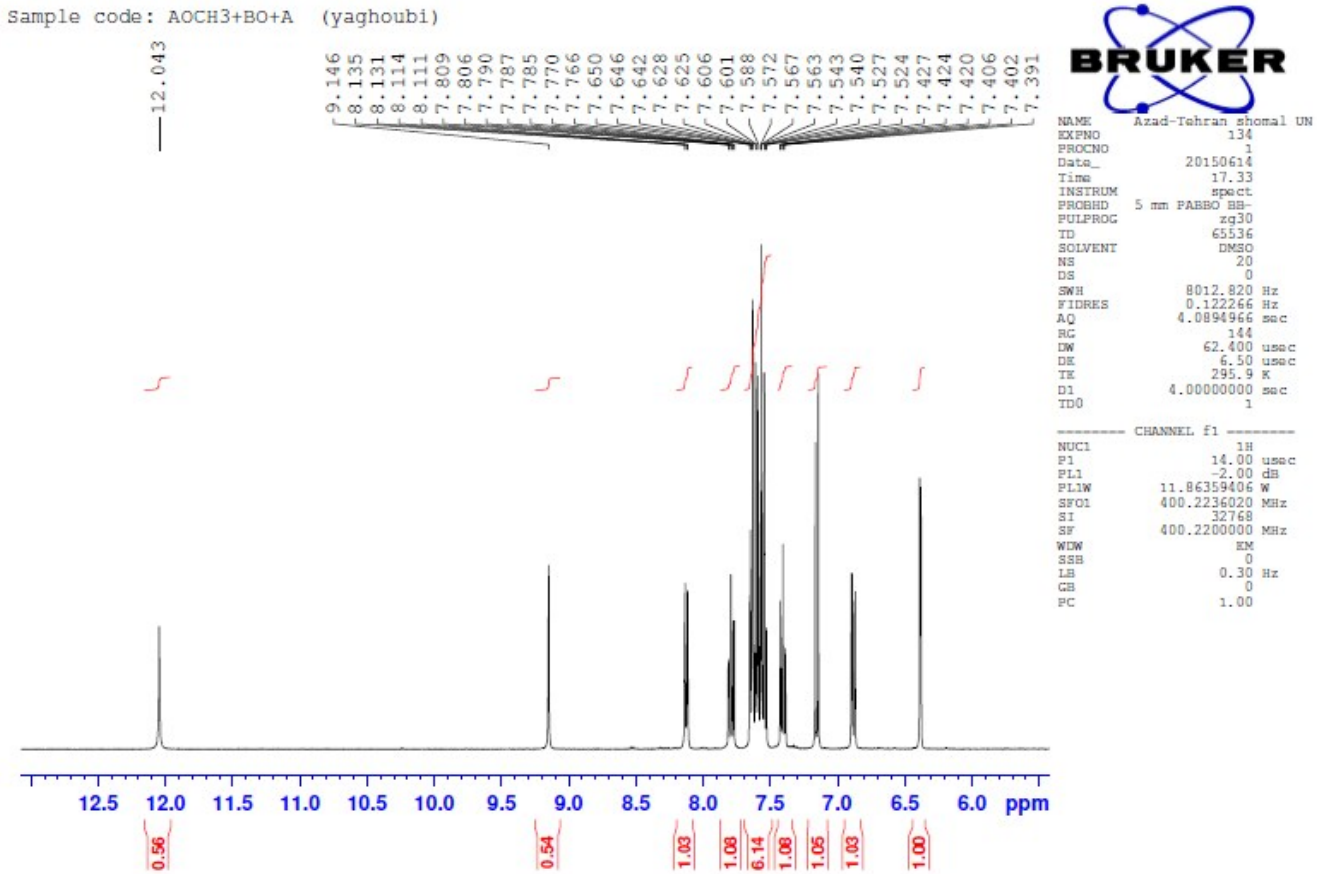
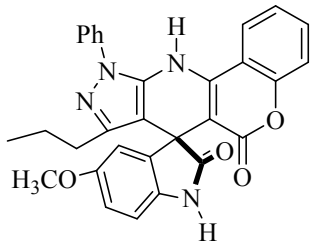
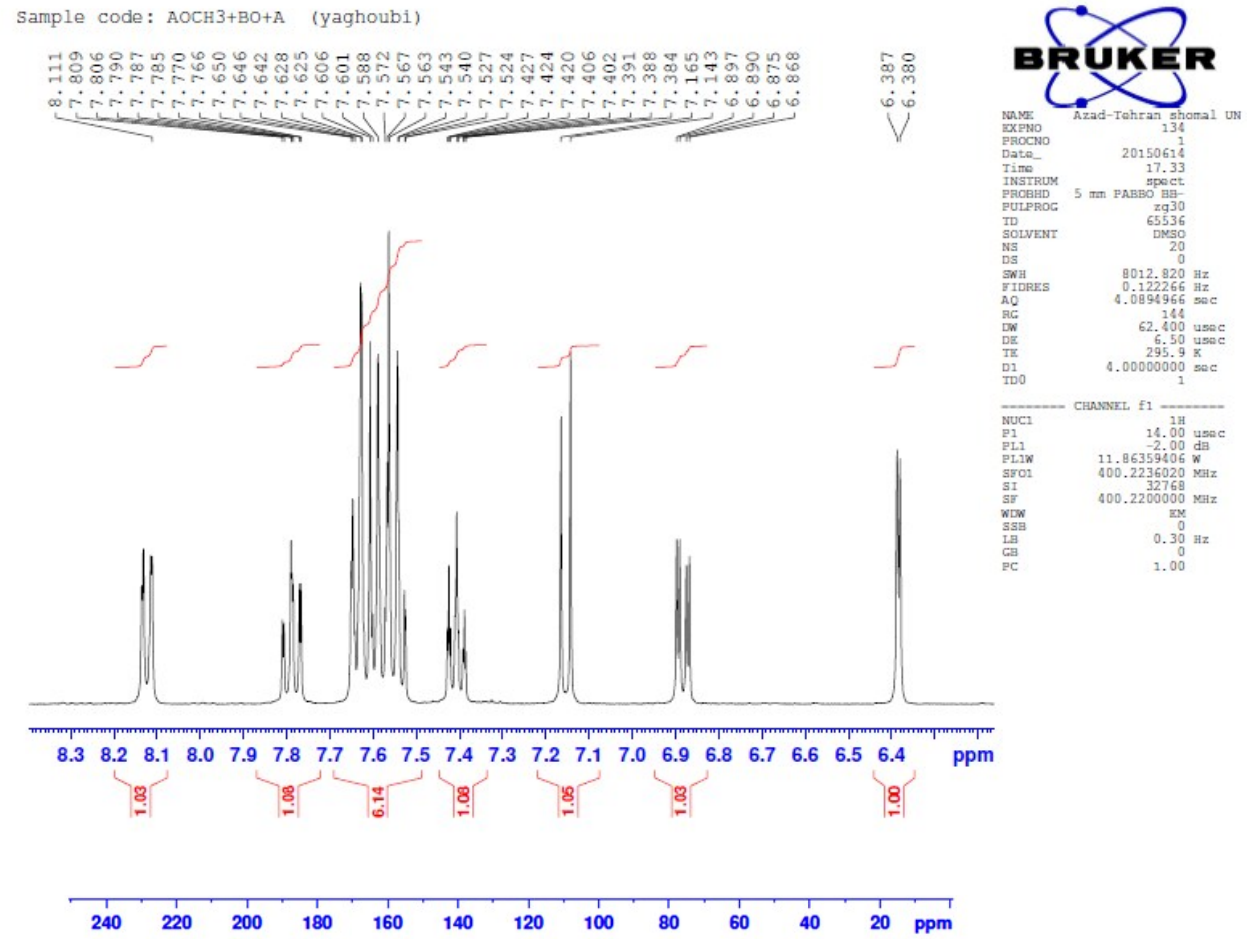
S9 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-c]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1-phenyl-3-propyl-5,2′-dione (**4f**)





S9 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-c]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1-phenyl-3-propyl-5,2′-dione (**4f**)





BRUKER

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PROCNO	1	
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PULPROG	zg30	
TD	65536	
SOLVENT	DMSO	
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FIDRES	0.122266	Hz
AQ	4.0894966	sec
RG	144	
DW	62.400	usec
DE	6.50	usec
TE	295.9	K
D1	4.00000000	sec
TD0	1	
======== CHANNEL f1 ========		
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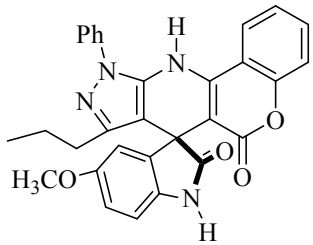
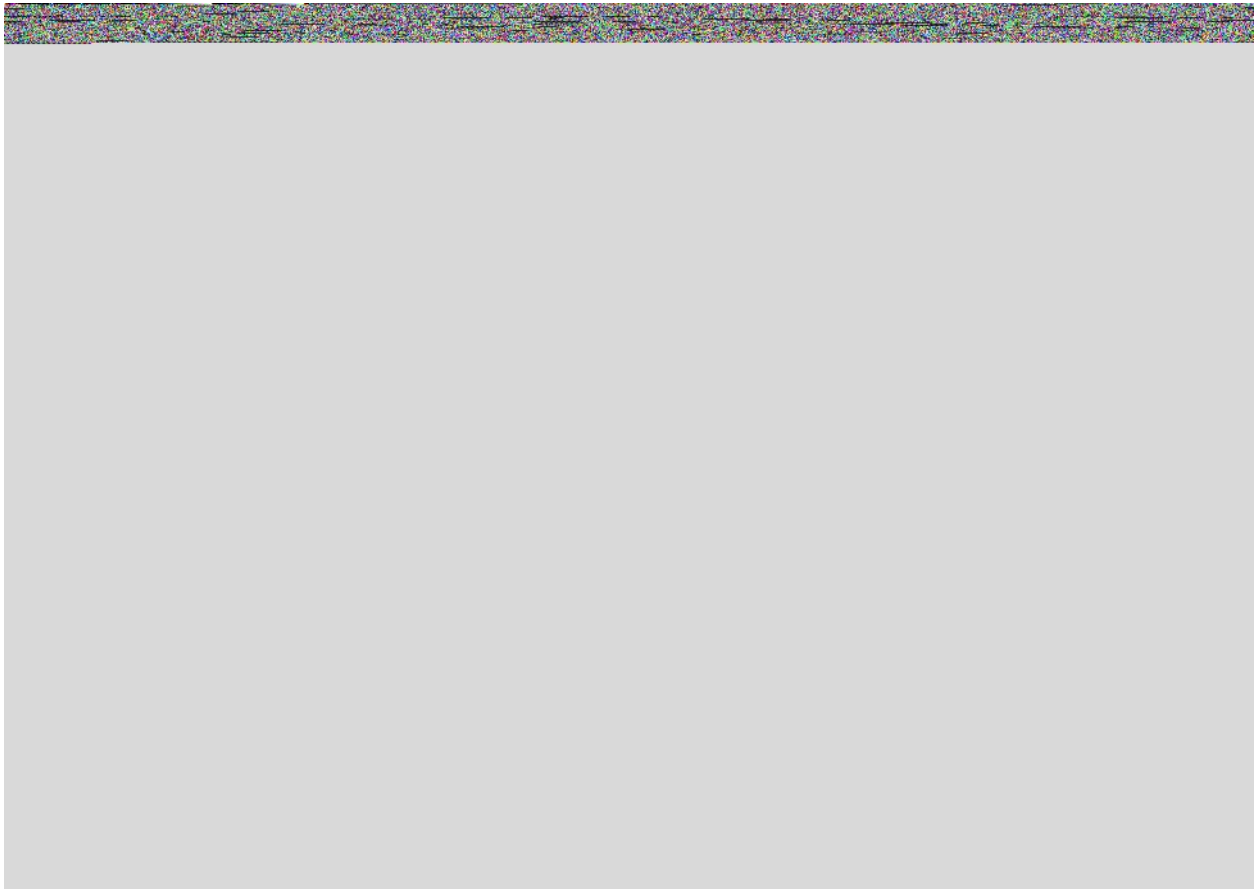
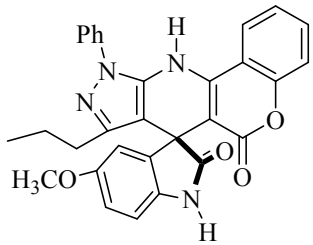
P1	14.00	usec
PL1	-2.00	dB
PL1W	11.86359406	W
SFO1	400.2236020	MHz
SI	32768	
SF	400.2200000	MHz
WDW	EM	
SSB	0	
LB	0.30	Hz
GB	0	
PC	1.00	

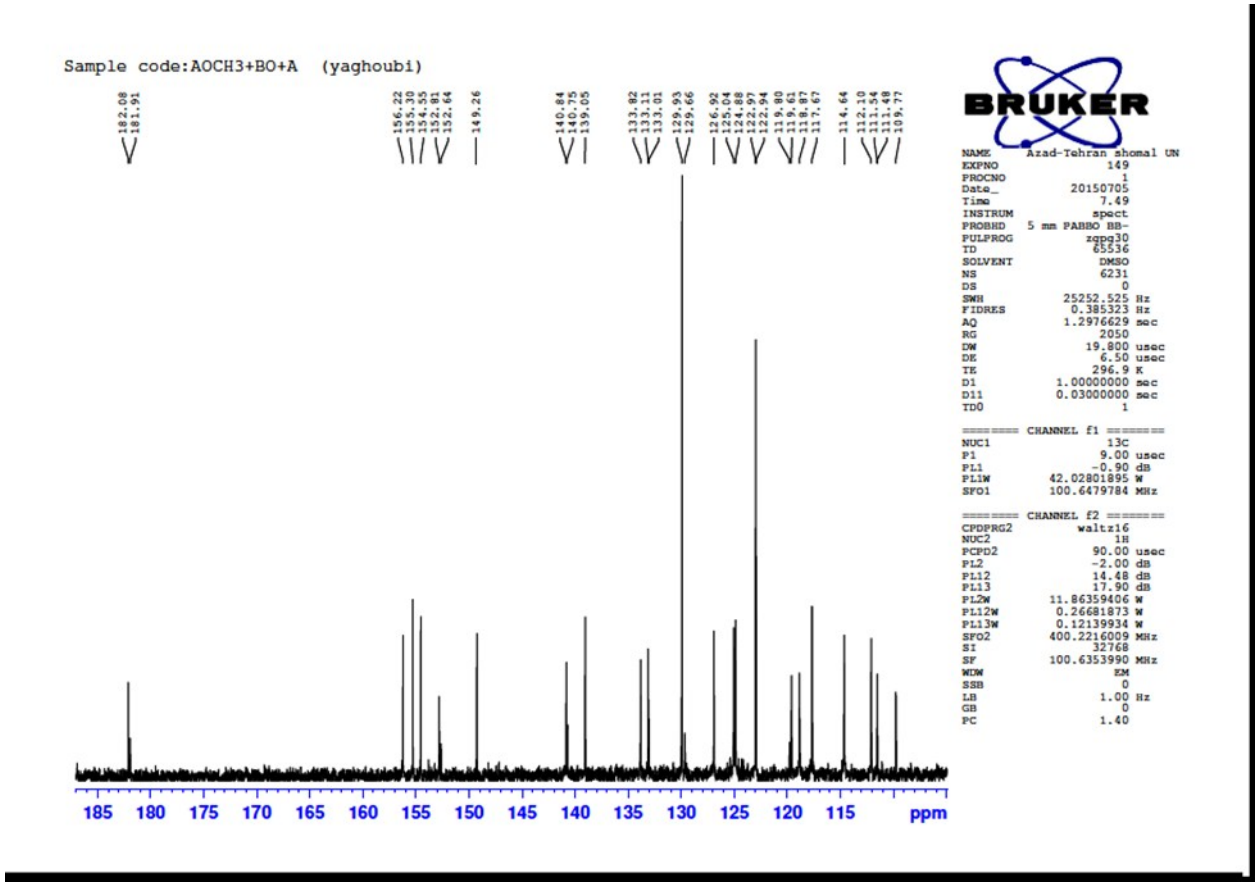
S$_{10}$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1-phenyl-3-propyl-5,2′-dione (**4f**)



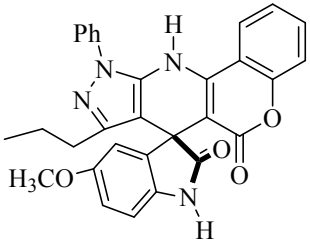


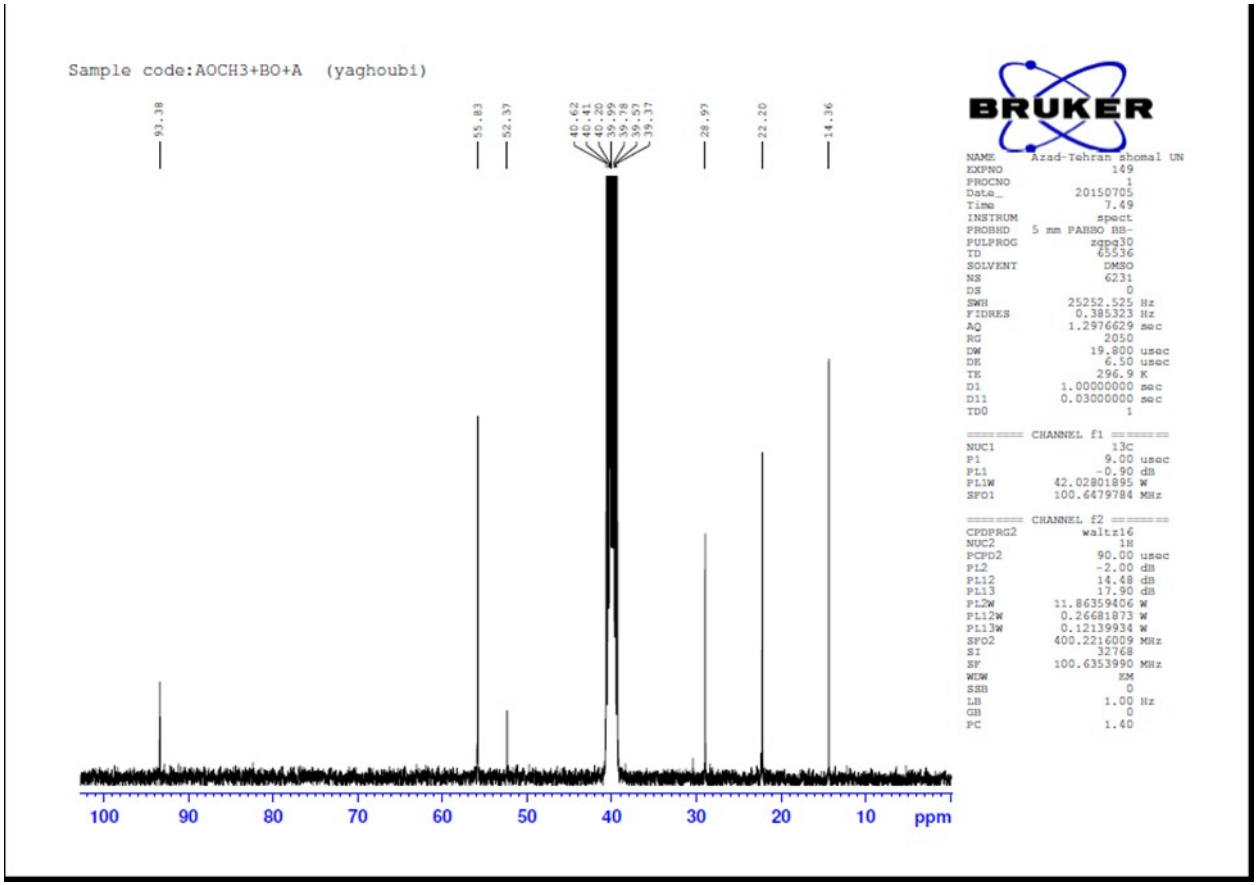
S$_{10}$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1-phenyl-3-propyl-5,2′-dione (**4f**)



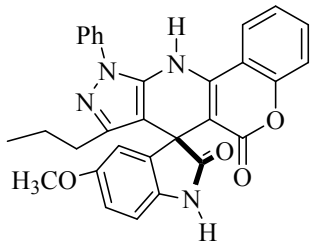


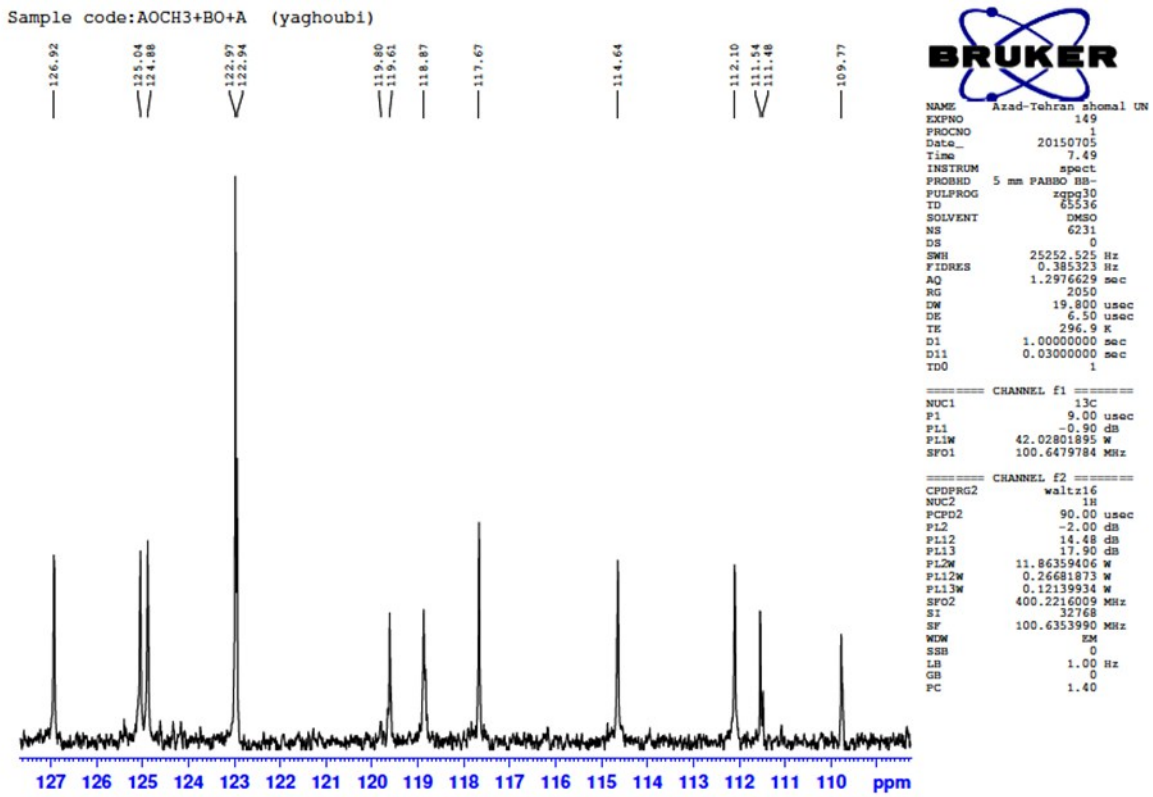
S$_{10}$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1-phenyl-3-propyl-5,2′-dione (**4f**)



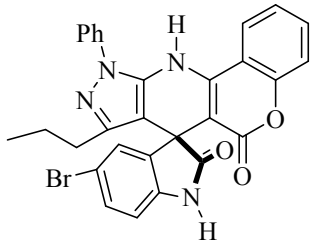


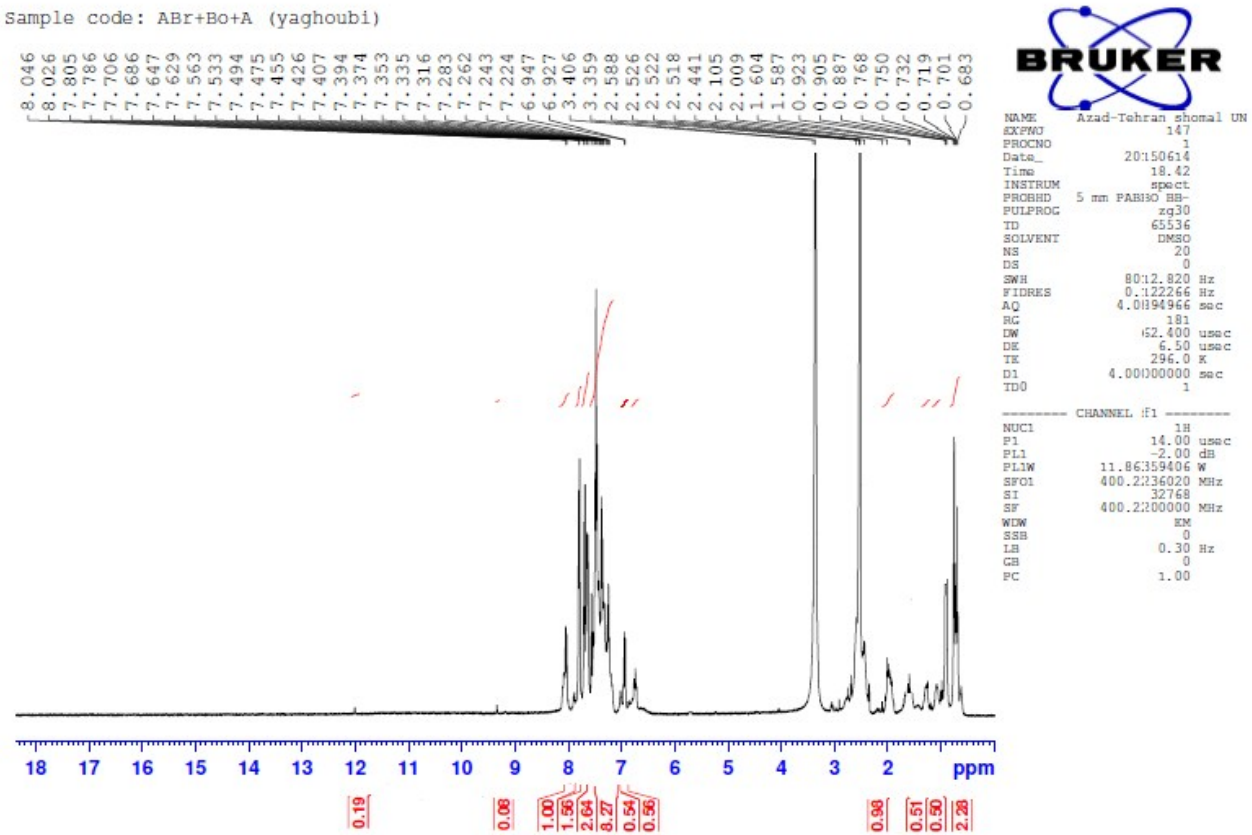
S$_{10}$ The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-methoxy-1-phenyl-3-propyl-5,2′-dione (**4f**)



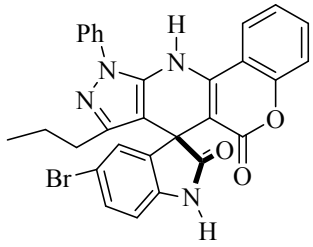


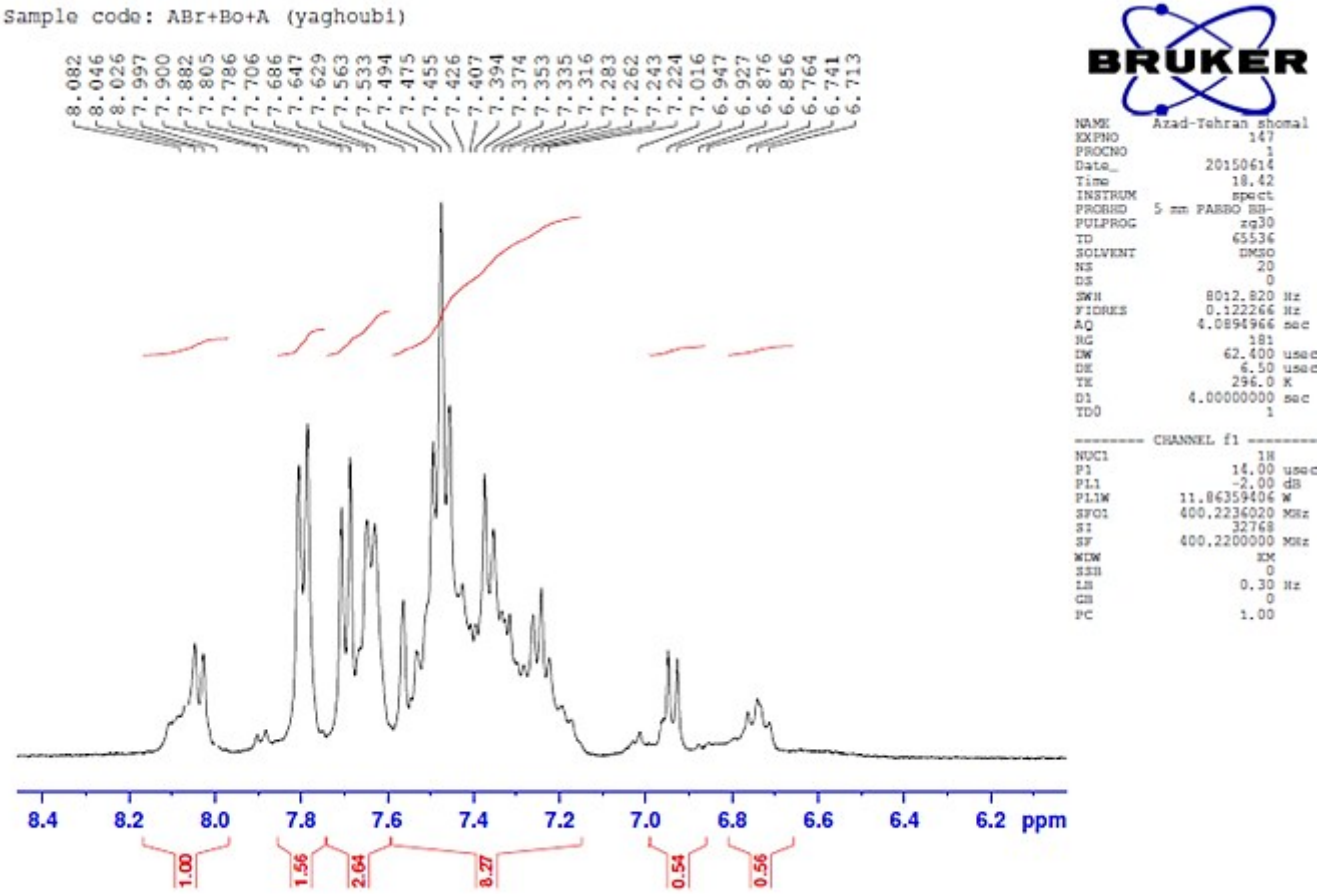
S11 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5'-bromo-1-phenyl-3-propyl-5,2'-dione (**4g**)



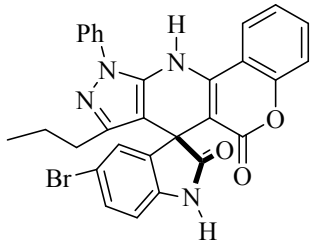


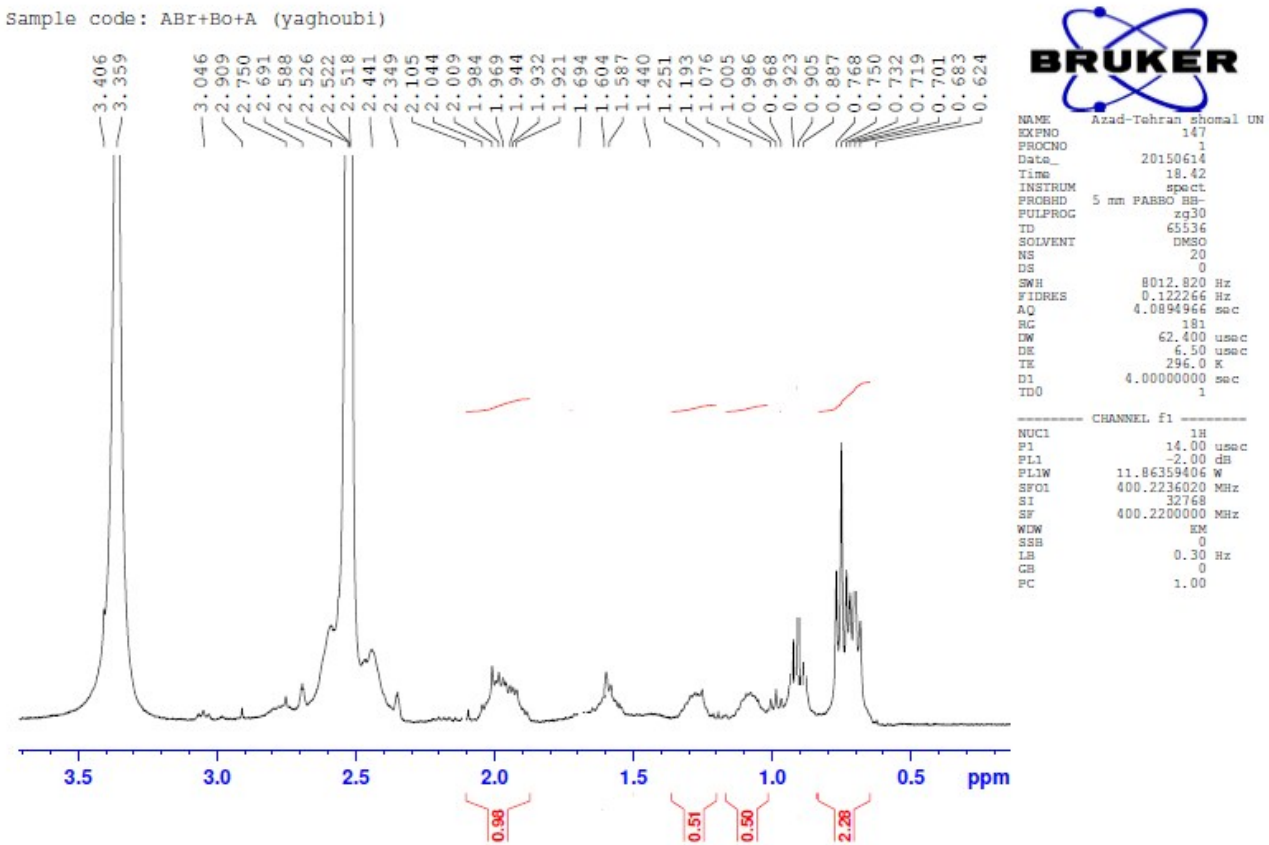
S11 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5'-bromo-1-phenyl-3-propyl-5,2'-dione (**4g**)



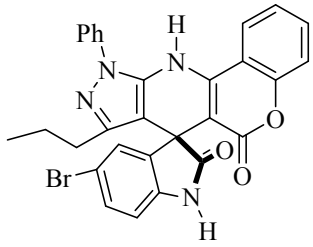


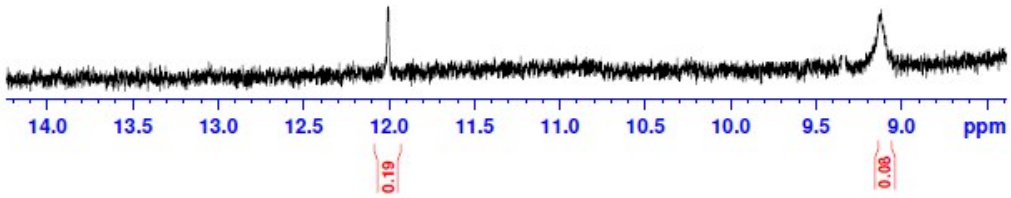
S11 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5'-bromo-1-phenyl-3-propyl-5,2'-dione (**4g**)





S11 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5'-bromo-1-phenyl-3-propyl-5,2'-dione (**4g**)



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AQ           4.0894966 sec
RG                  181
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D1          4.00000000 sec
TD0                    1

======== CHANNEL f1 ========
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WDW                   EM
SSB                    0
LB                  0.30 Hz
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PC                  1.00
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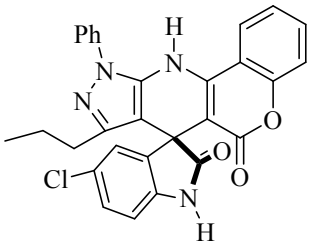
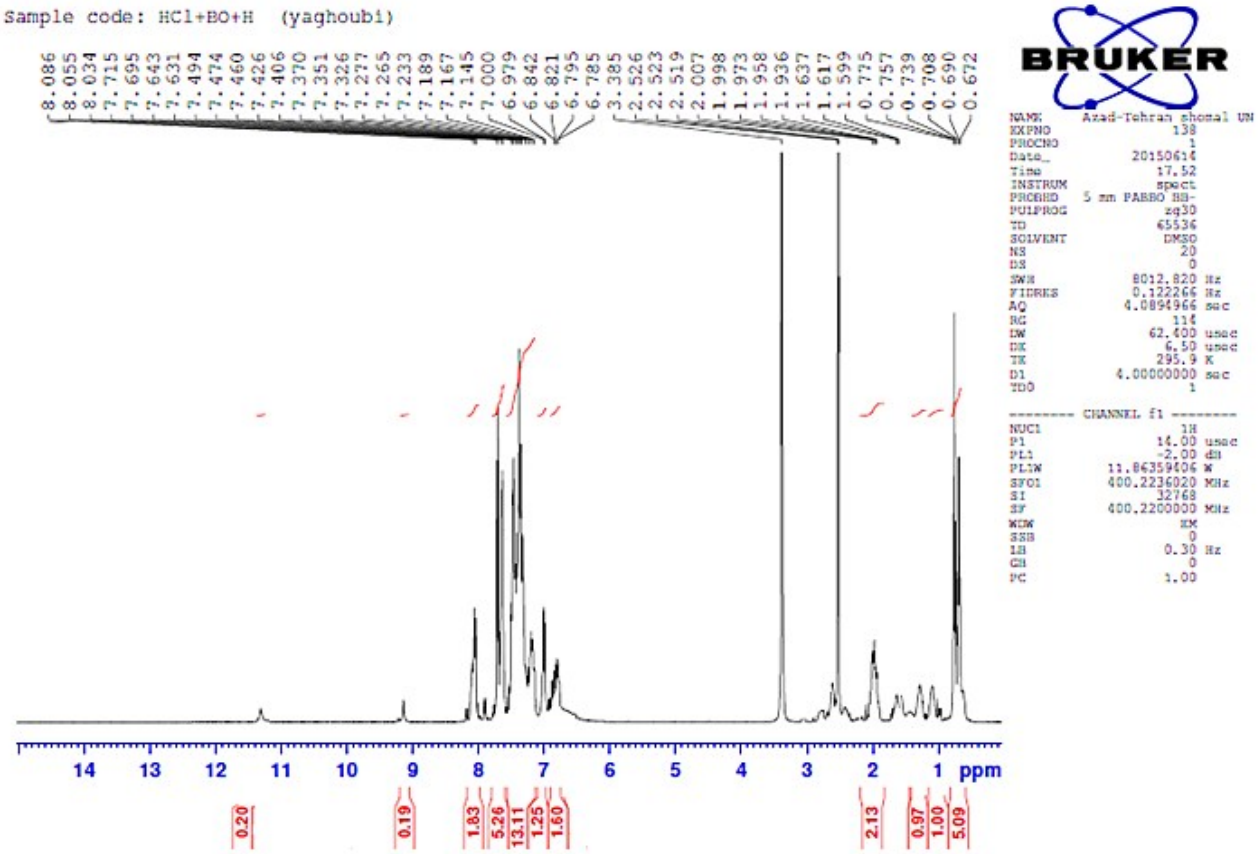
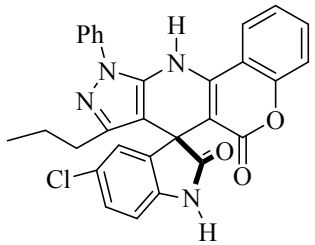
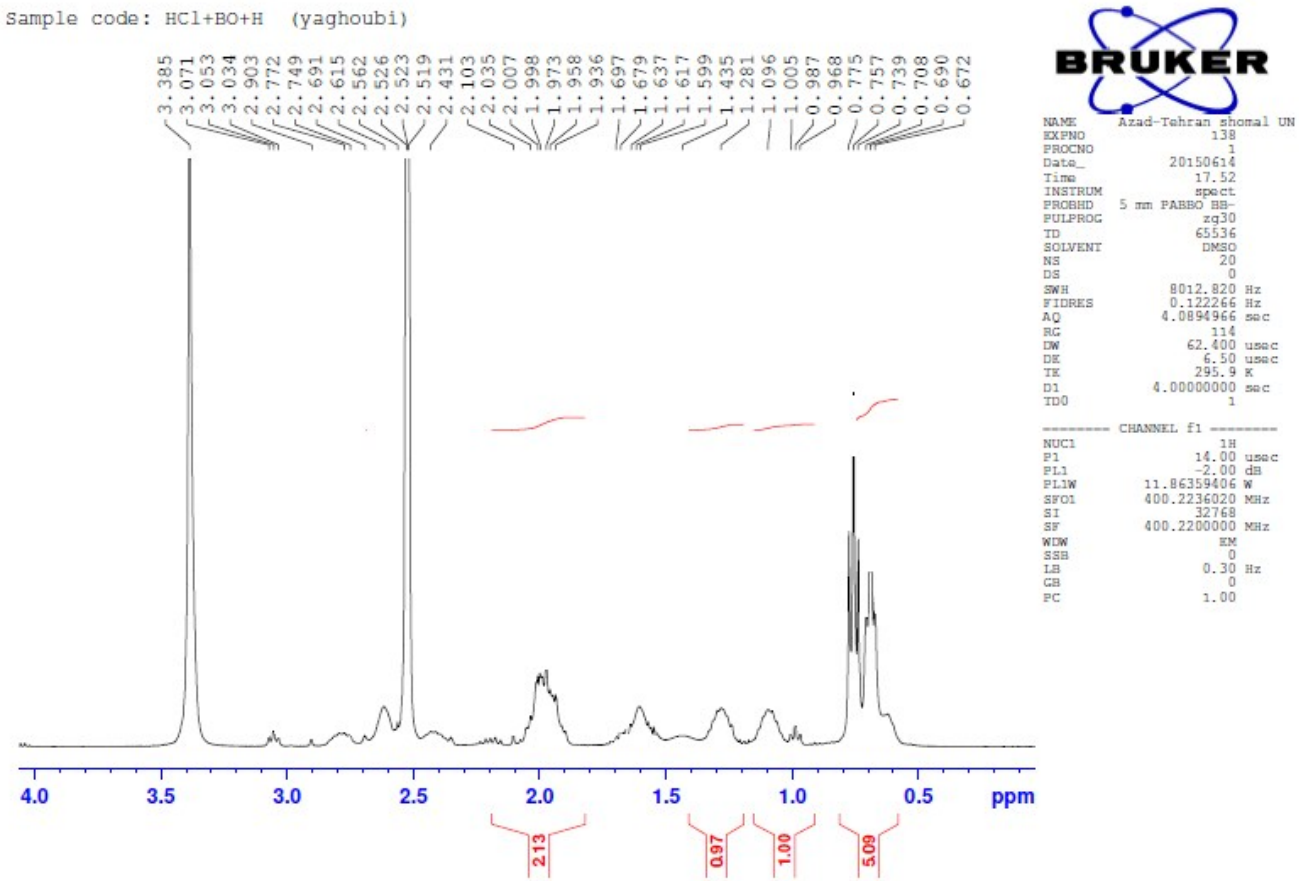
S$_{12}$ The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-1-phenyl-3-propyl-5,2′-dione (**4h**)



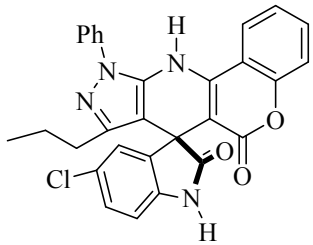


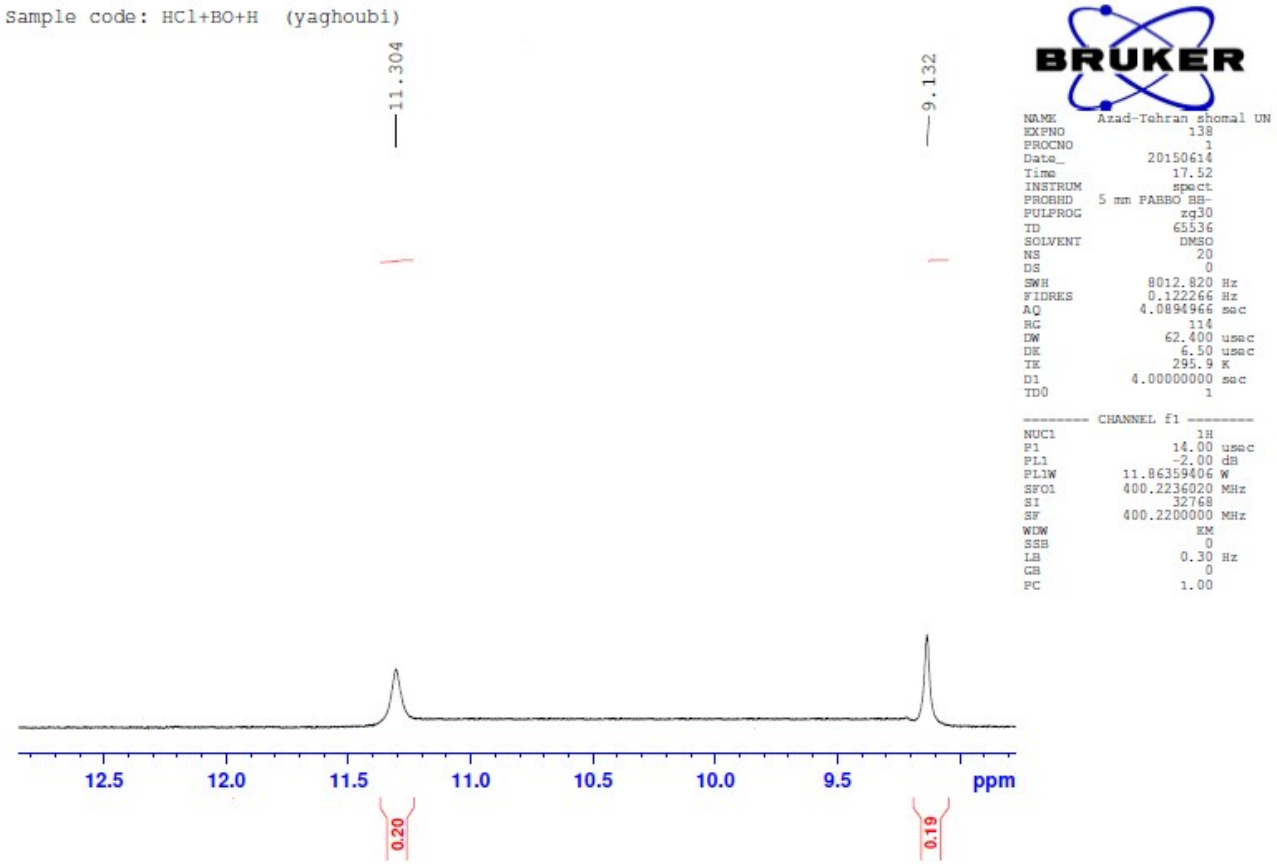
S12 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-1-phenyl-3-propyl-5,2′-dione (**4h**)



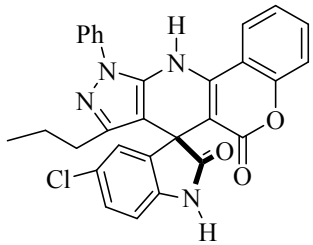


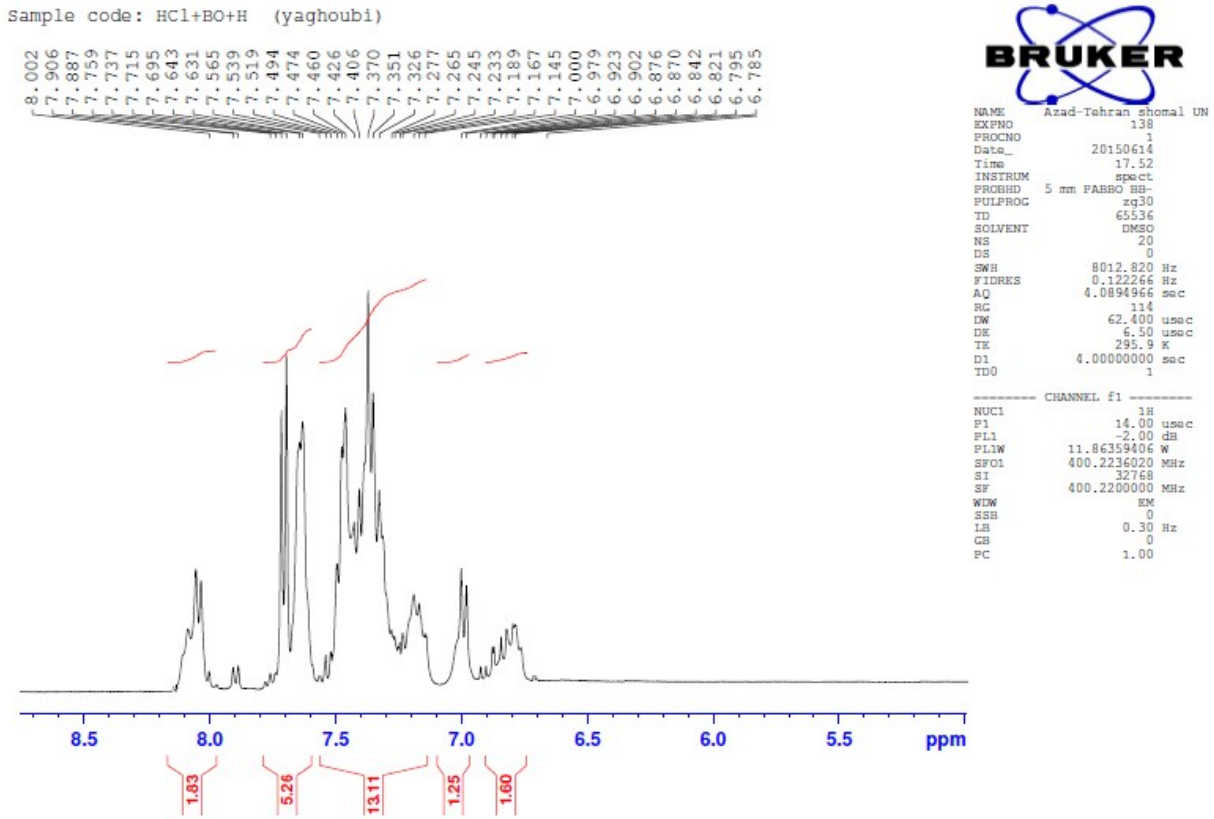
S12 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-1-phenyl-3-propyl-5,2′-dione (**4h**)





S12 The ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-1-phenyl-3-propyl-5,2′-dione (**4h**)





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AQ                  4.0894966 sec
RG                        114
DW                     62.400 usec
DE                       6.50 usec
TE                      295.9 K
D1                 4.00000000 sec
TD0                         1

======== CHANNEL f1 ========
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P1                      14.00 usec
PL1                     -2.00 dB
PLW              11.86359406 W
SFO1             400.2236020 MHz
SI                       32768
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PC                       1.00
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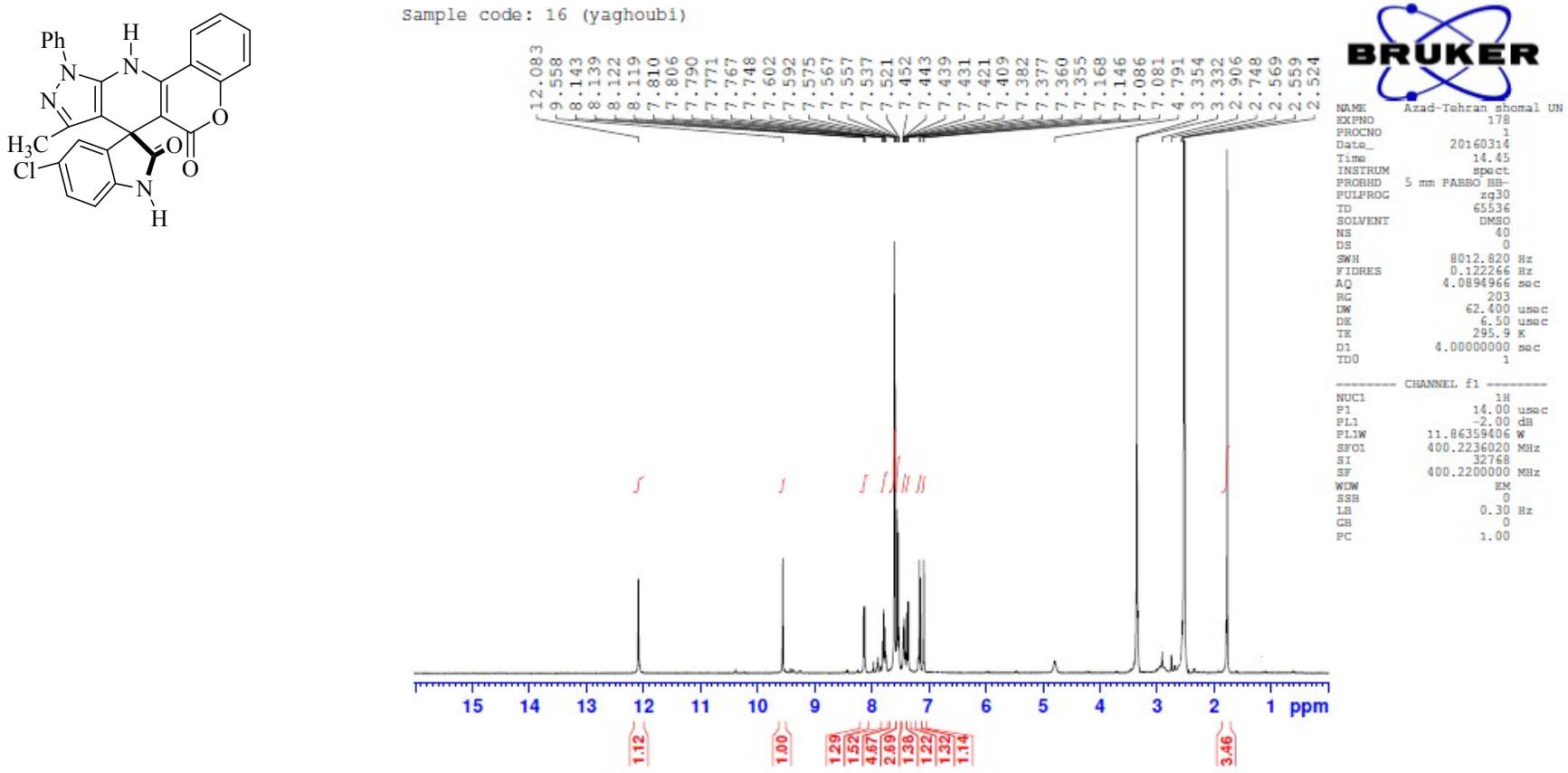
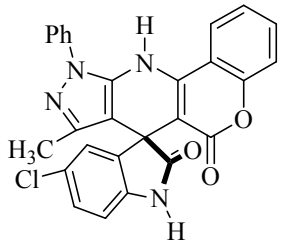
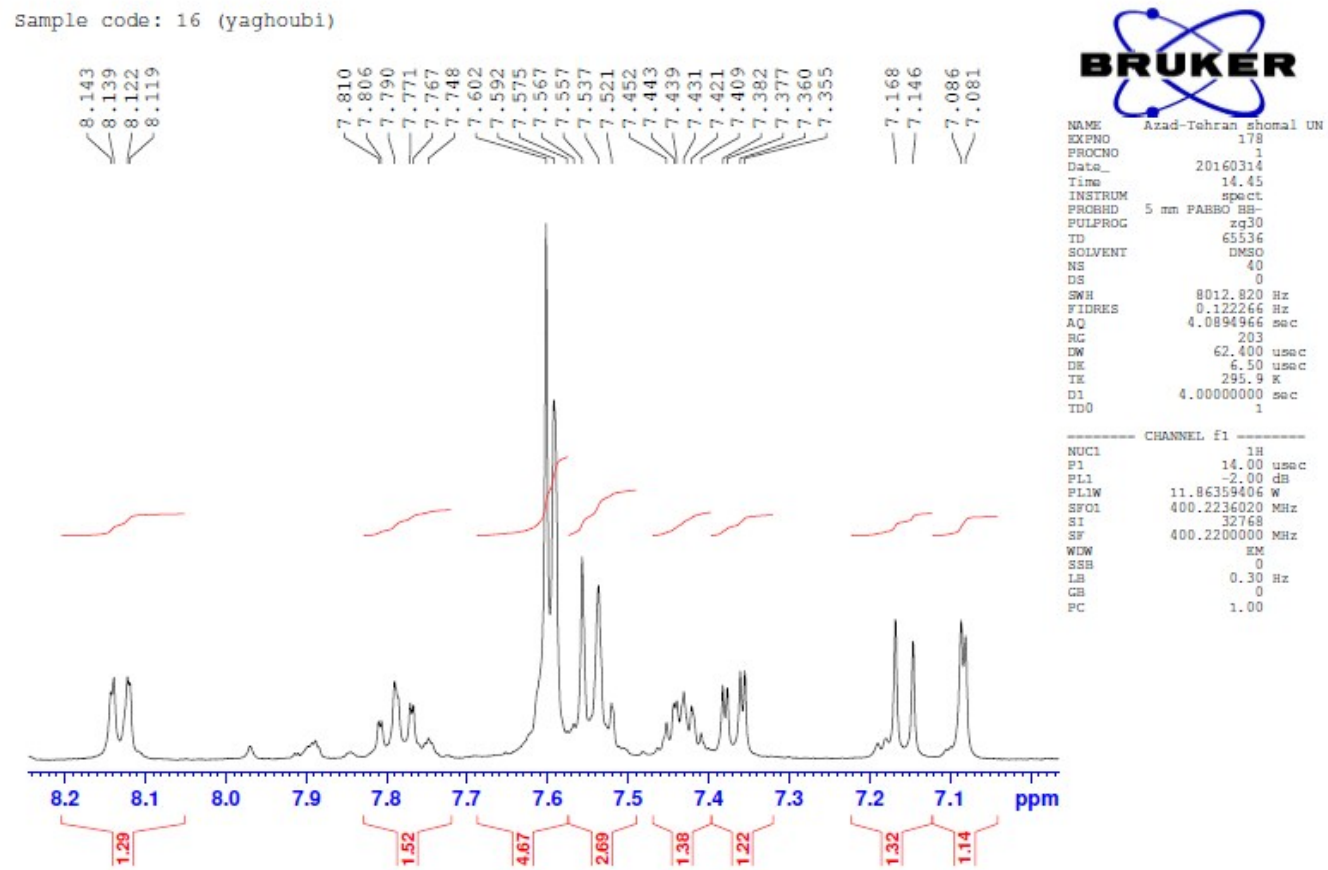
S$_{13}$ ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-3-methyl-1-phenyl-5,2′-dione (**4i**)



S13 ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-3-methyl-1-phenyl-5,2′-dione (**4i**)



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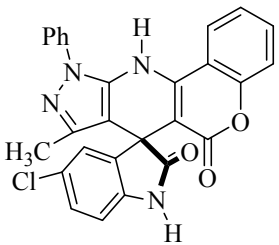
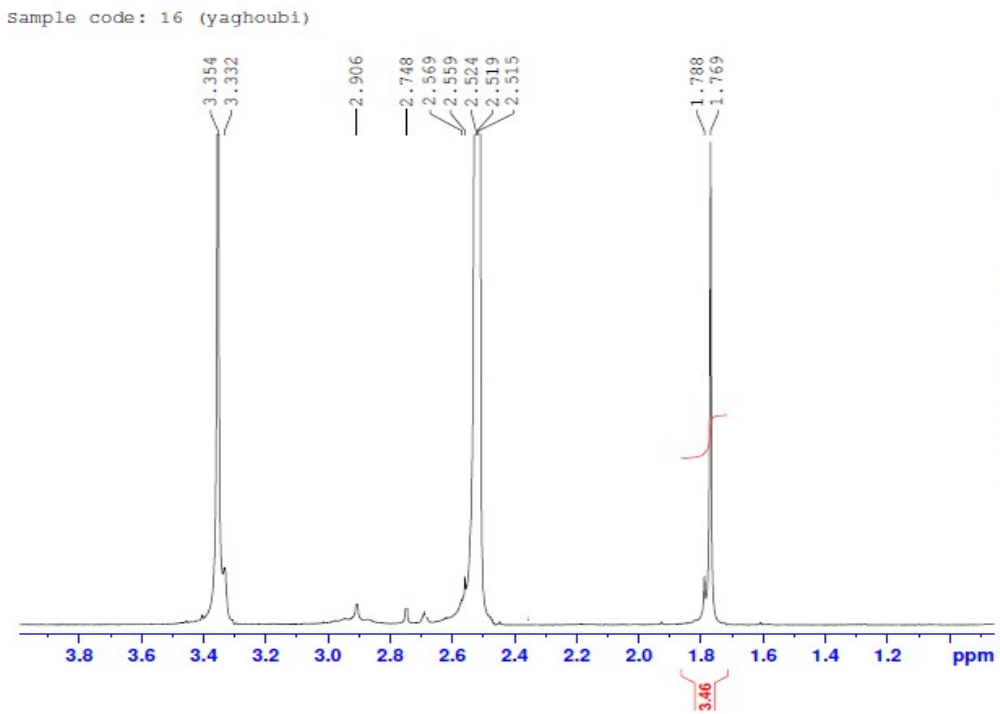
NAME        Azad-Tehran shomal UN
EXPNO                     178
PROCNO                      1
Date_                20160314
Time                    14.45
INSTRUM                 spect
PROBHD     5 mm PABBO BB-
PULPROG                  zg30
TD                      65536
SOLVENT                  DMSO
NS                         40
DS                          0
SWH                  8012.820 Hz
FIDRES               0.122266 Hz
AQ                 4.0894966 sec
RG                        203
DW                     62.400 usec
DE                       6.50 usec
TE                      295.9 K
D1                 4.00000000 sec
TD0                         1

======== CHANNEL f1 ========
NUC1                       1H
P1                      14.00 usec
PL1                     -2.00 dB
PL1W               11.86359406 W
SFO1               400.2236020 MHz
SI                       32768
SF                 400.2200000 MHz
WDW                        EM
SSB                         0
LB                       0.30 Hz
GB                          0
PC                       1.00
```

S$_{13}$ ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-3-methyl-1-phenyl-5,2′-dione (**4i**)

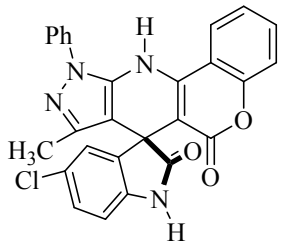


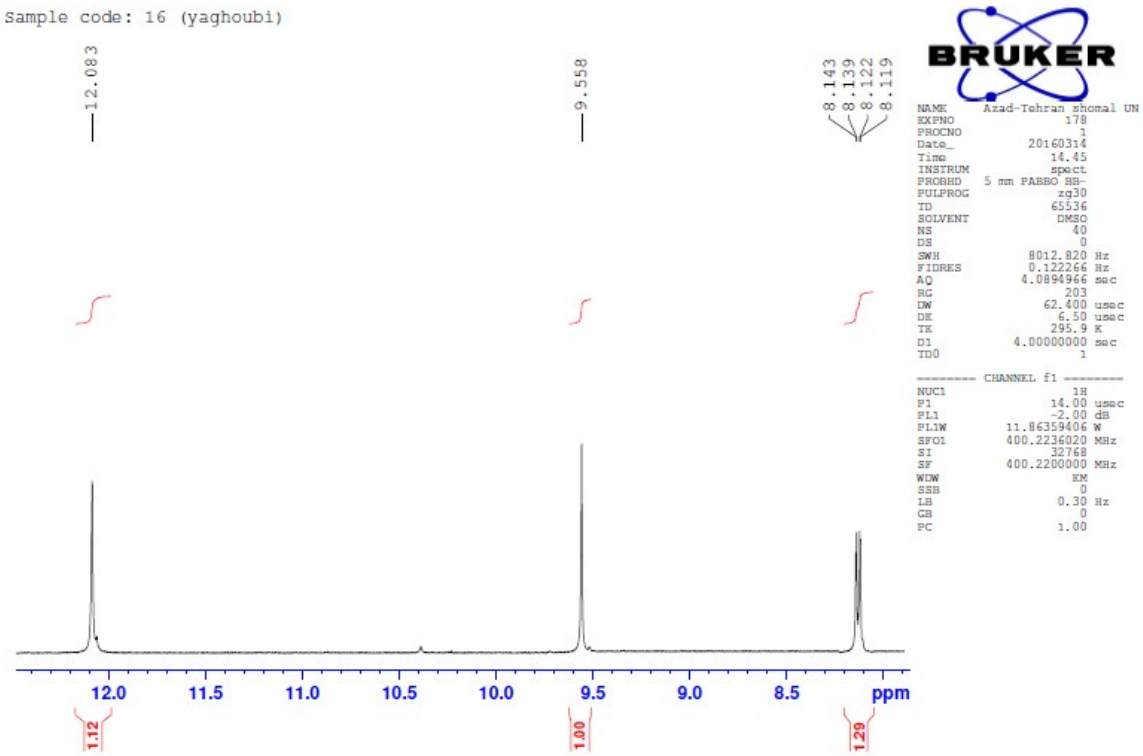


BRUKER

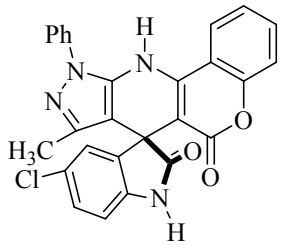
NAME	Azad-Tehran shomal UN
EXPNO	178
PROCNO	1
Date_	20160314
Time	14.45
INSTRUM	spect
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	65536
SOLVENT	DMSO
NS	40
DS	0
SWH	8012.820 Hz
FIDRES	0.122266 Hz
AQ	4.0894966 sec
RG	203
DW	62.400 usec
DE	6.50 usec
TE	295.9 K
D1	4.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	14.00 usec
PL1	-2.00 dB
PL1W	11.86359406 W
SFO1	400.2236020 MHz
SI	32768
SF	400.2200000 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	1.00

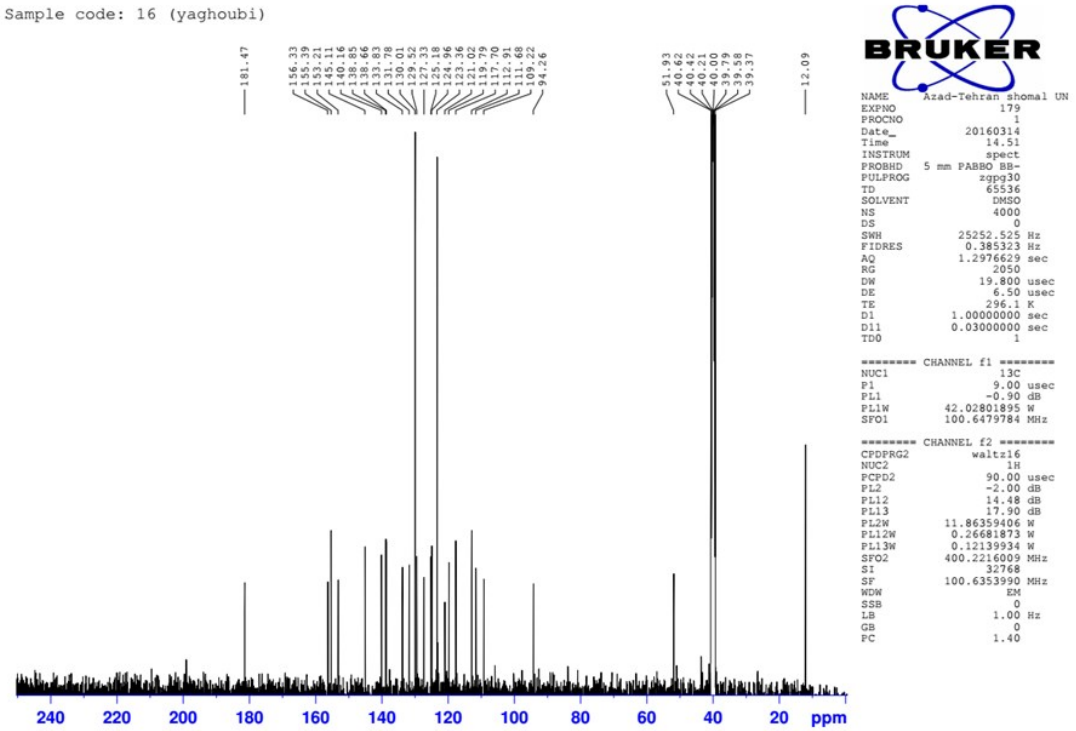
S13 ^{1}H NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-3-methyl-1-phenyl-5,2′-dione (**4i**)



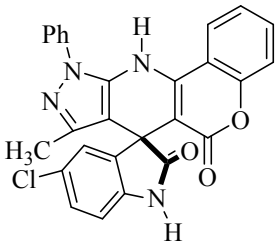


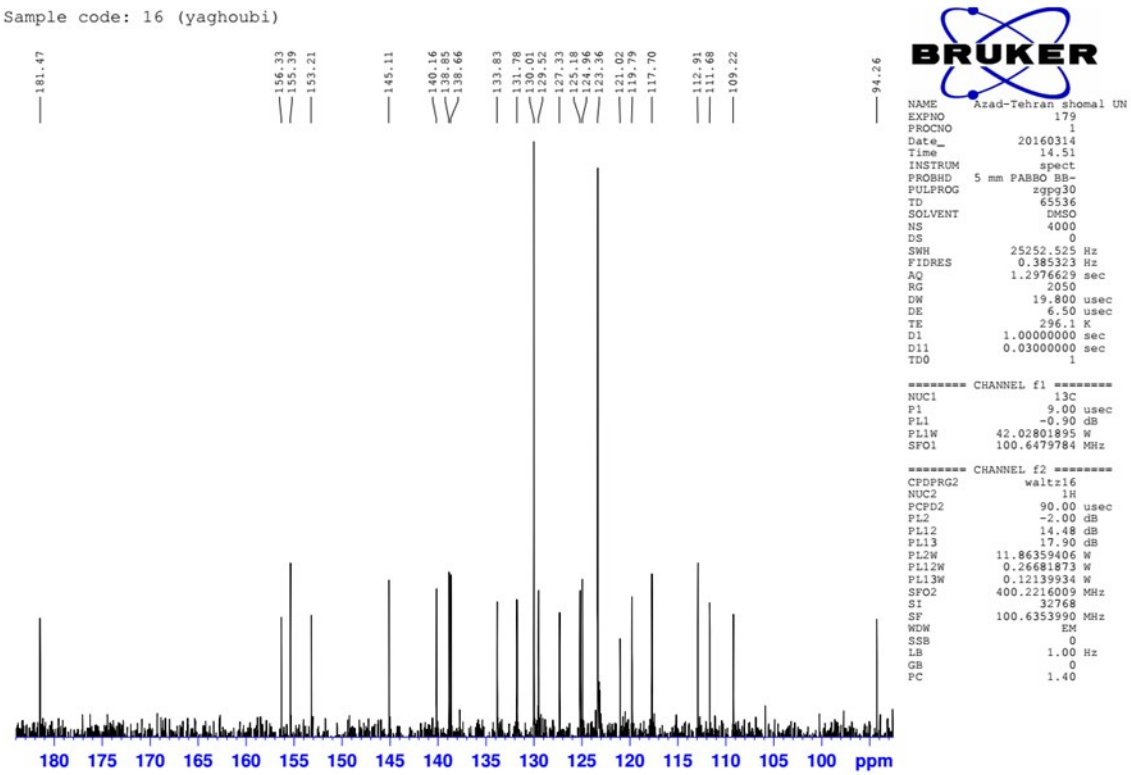
S14 The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-3-methyl-1-phenyl-5,2′-dione (**4i**)



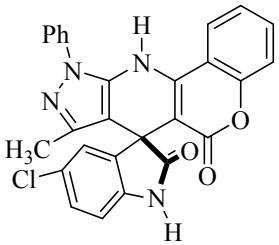


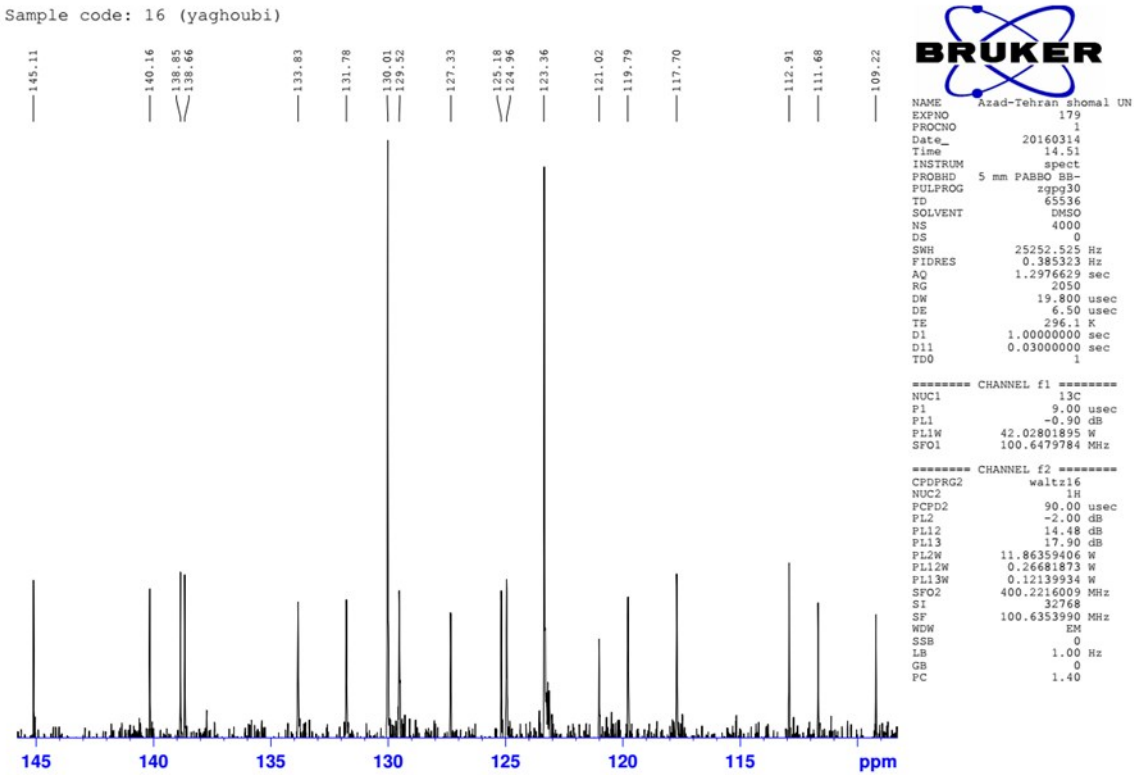
S14 The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-3-methyl-1-phenyl-5,2′-dione (**4i**)



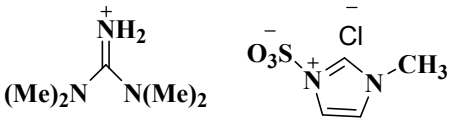


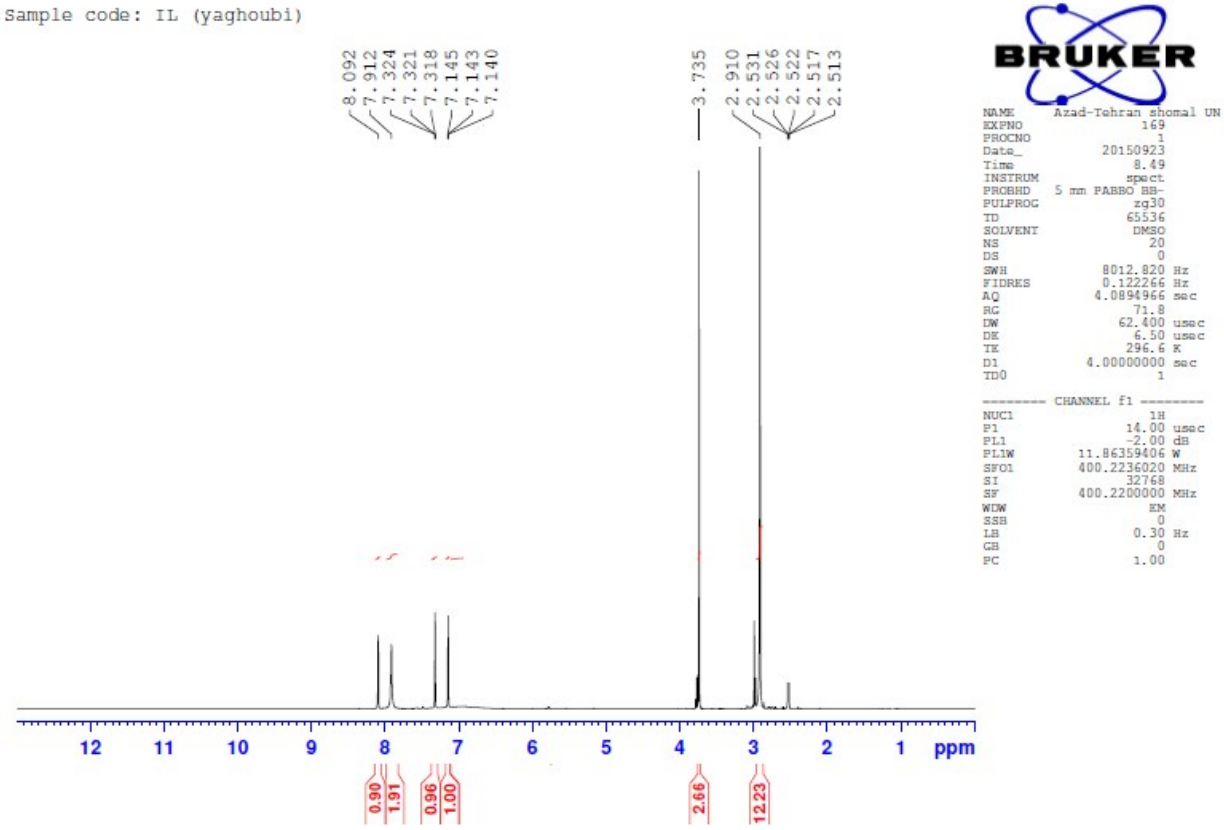
S14 The ^{13}C NMR spectrum of Spiro[1*H*-pyrazolo[5,4-*b*]pyrido[5,6-*c*]chromene-4,3-indoline]-4*H*,5*H*,11*H*-5′-chloro-3-methyl-1-phenyl-5,2′-dione (**4i**)



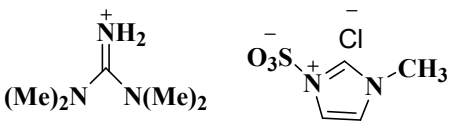


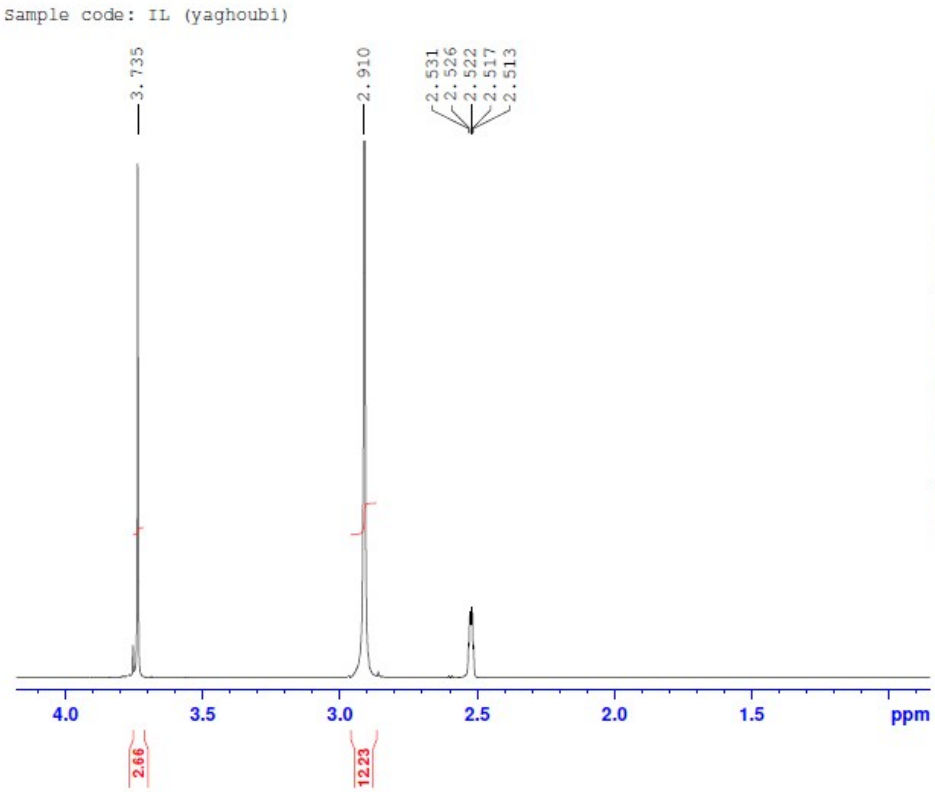
S15 The ^{1}H NMR spectrum of [TMG·HCl][MImS]

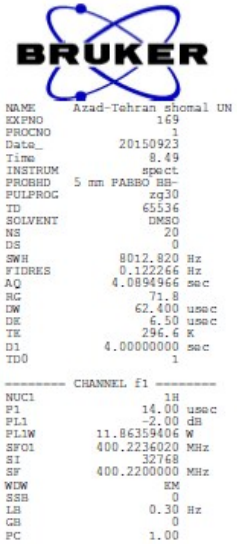




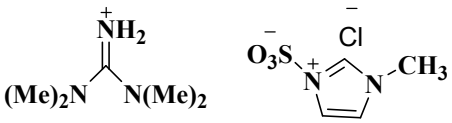
S15 The ^{1}H NMR spectrum of [TMG·HCl][MImS]

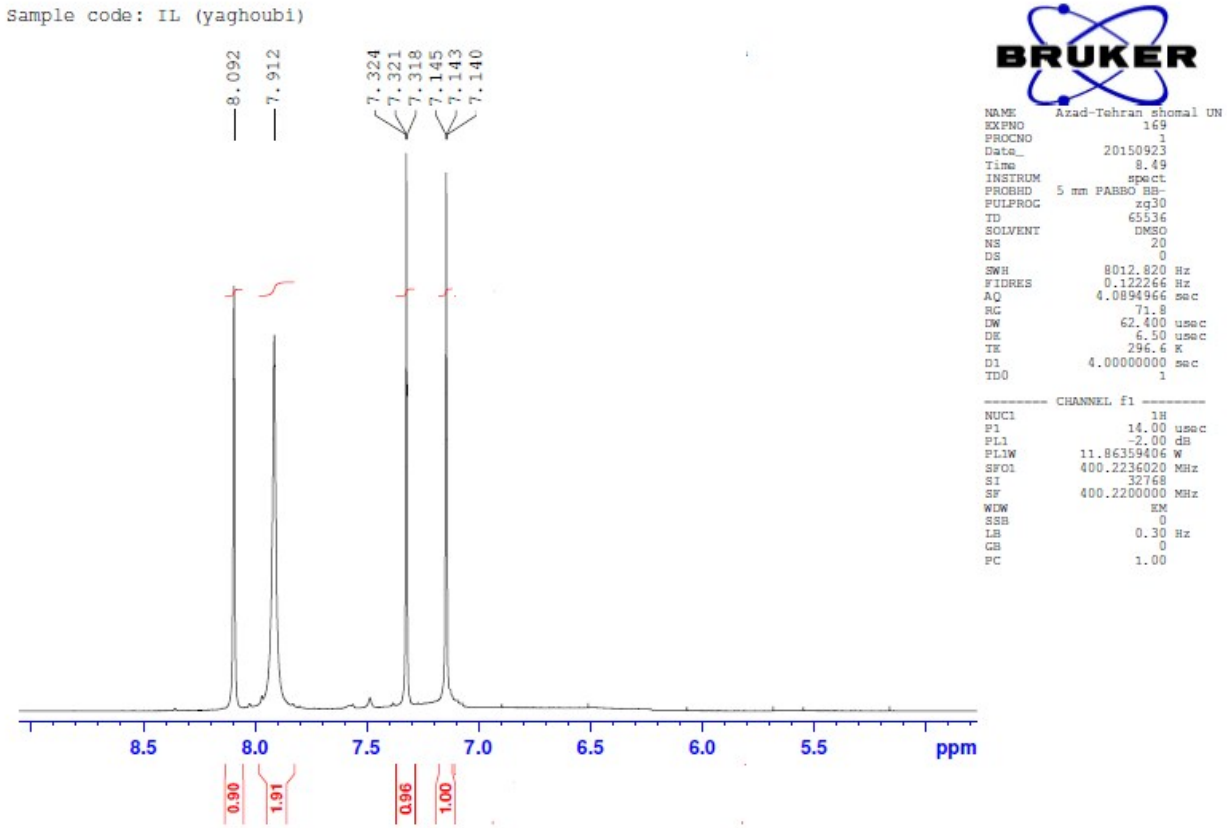




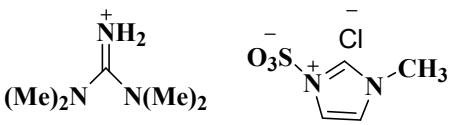


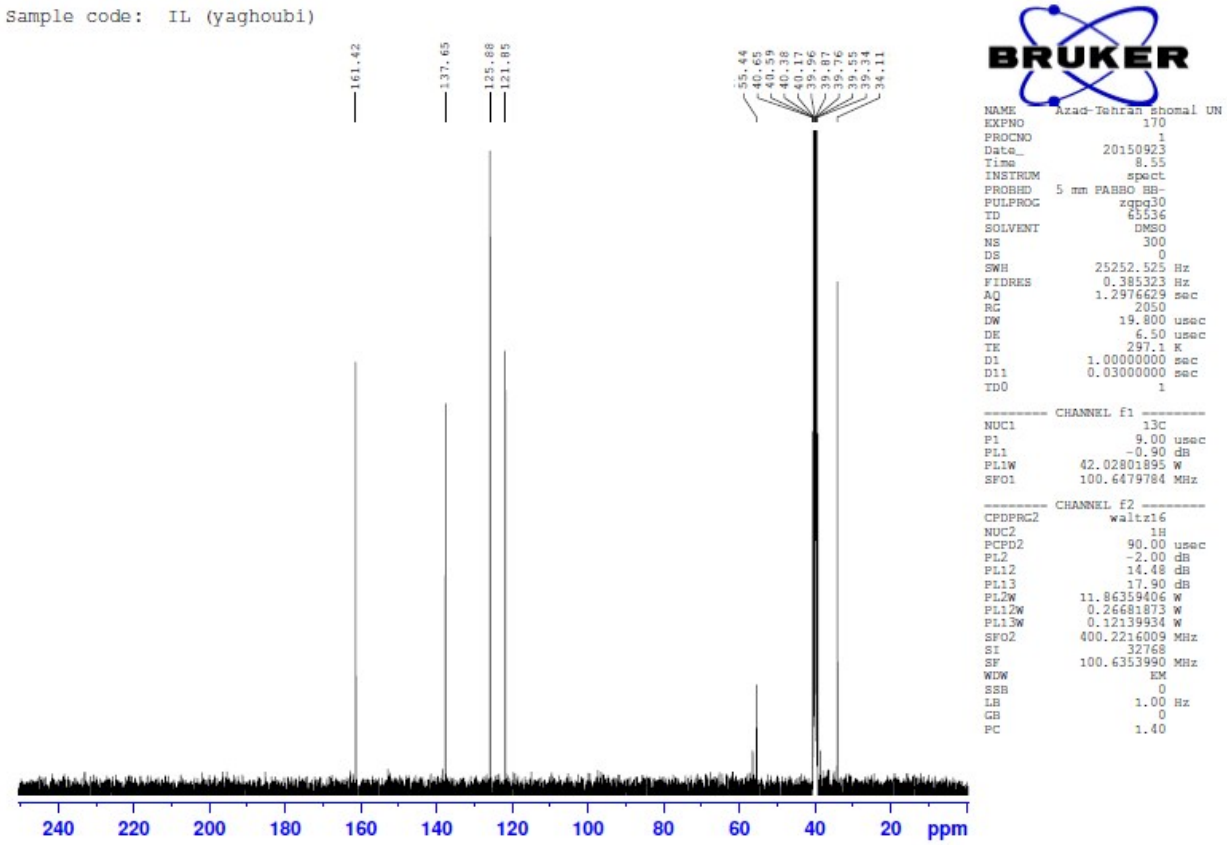
S15 The ^{1}H NMR spectrum of [TMG·HCl][MImS]



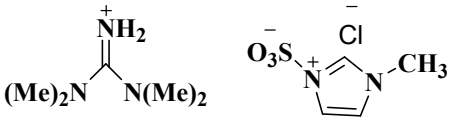


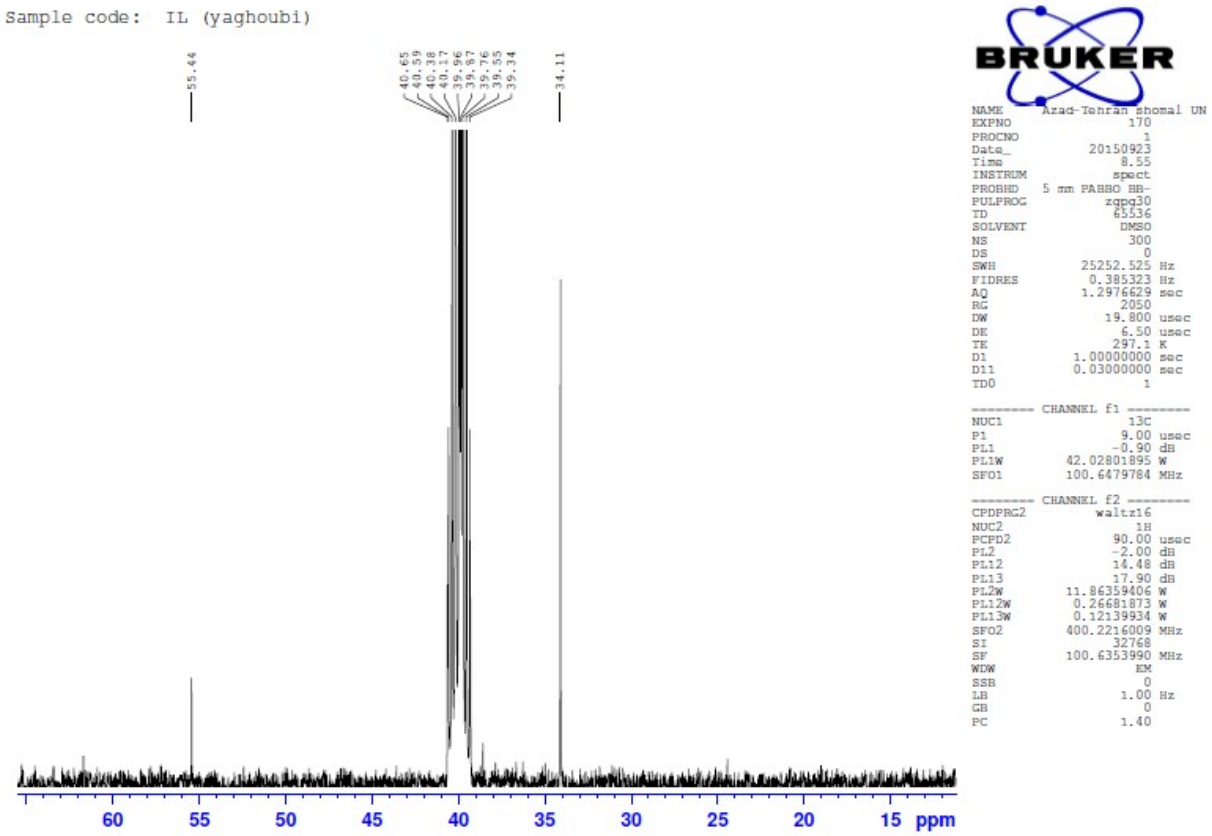
S16 The ^{13}C NMR spectrum of [TMG·HCl][MImS]



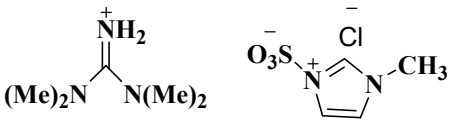


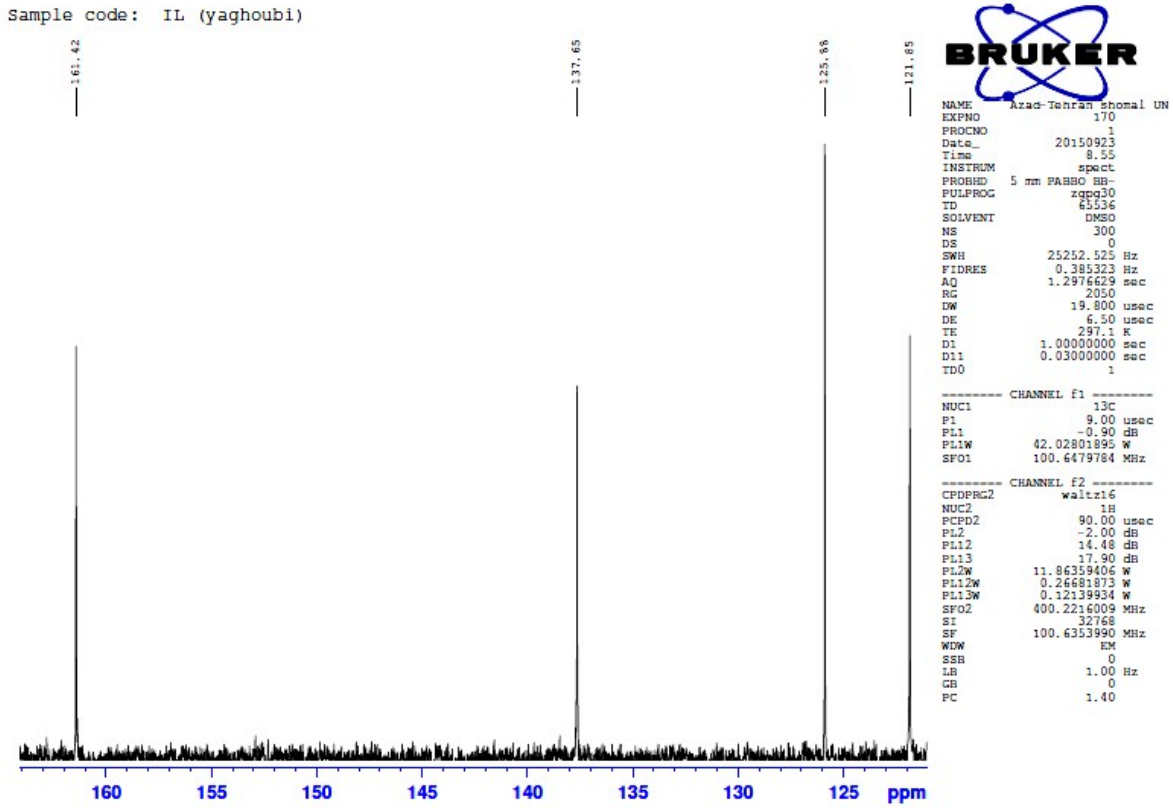
S16 The ^{13}C NMR spectrum of [TMG·HCl][MImS]



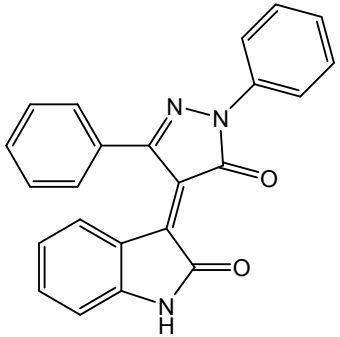


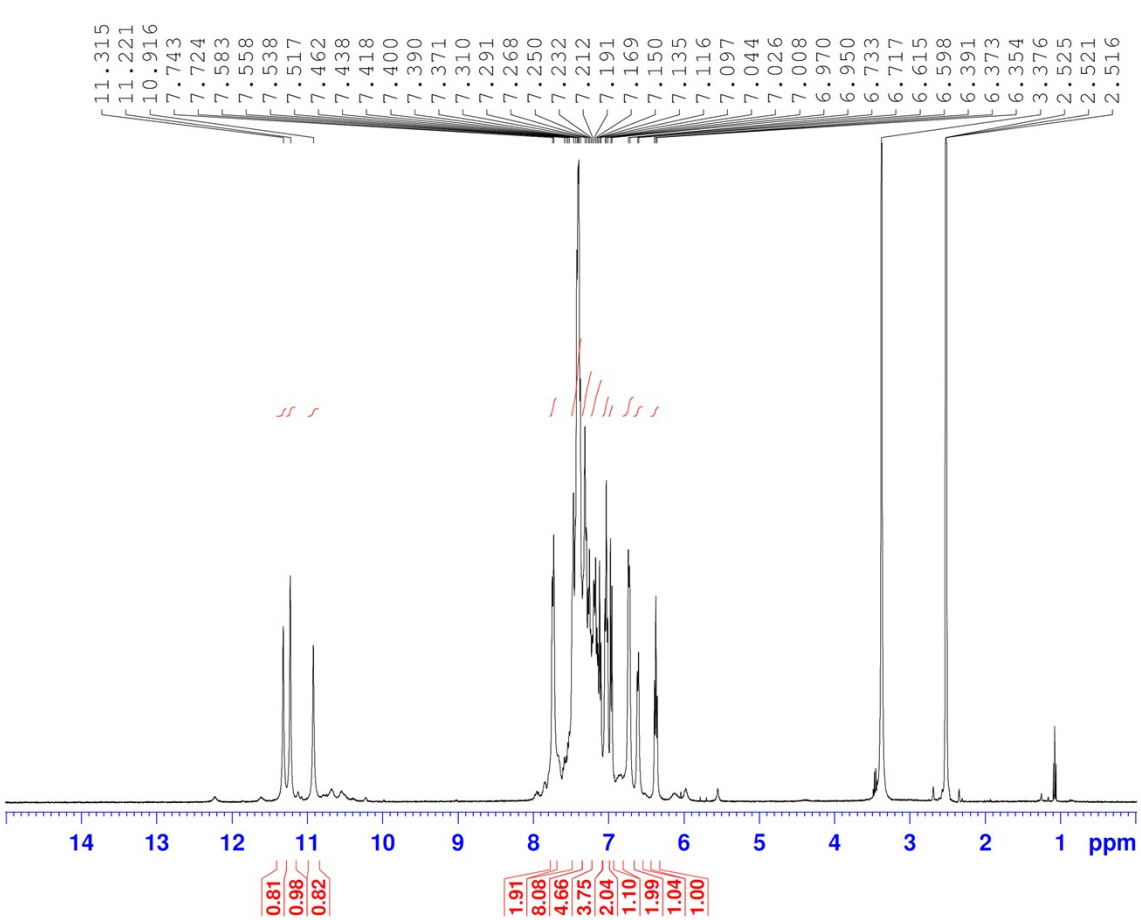
S16 The ^{13}C NMR spectrum of [TMG·HCl][MImS]




Sample code: IL (yaghoubi)
161.42
137.65
125.88
121.85
160
155
150
145
140
135
130
125
ppm
NAME Arad-Tehran Shomal UN
EXPNO 170
PROCNO 1
Date_ 20150923
Time 8.55
INSTRUM spect
PROBHD 5 mm PABBO BB-
PULPROG zgpg30
TD 65536
SOLVENT DMSO
NS 300
DS 0
SWH 25252.525 Hz
FIDRES 0.385323 Hz
AQ 1.2976629 sec
RG 2050
DW 19.800 usec
DE 6.50 usec
TE 297.1 K
D1 1.00000000 sec
D11 0.03000000 sec
TD0 1
======== CHANNEL f1 ========
NUC1 13C
P1 9.00 usec
PL1 -0.90 dB
PL1W 42.02801895 W
SFO1 100.6479784 MHz
======== CHANNEL f2 ========
CPDPRG2 waltz16
NUC2 1H
PCPD2 90.00 usec
PL2 -2.00 dB
PL12 14.48 dB
PL13 17.90 dB
PL2W 11.86359406 W
PL12W 0.26681873 W
PL13W 0.12139934 W
SFO2 400.2216009 MHz
SI 32768
SF 100.6353990 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

S$_{17}$ The ^{1}H NMR spectrum of 3-(5-oxo-1,3-diphenyl-1*H*-pyrazol-4(5*H*)-ylidene)indolin-2-one (**7b**)

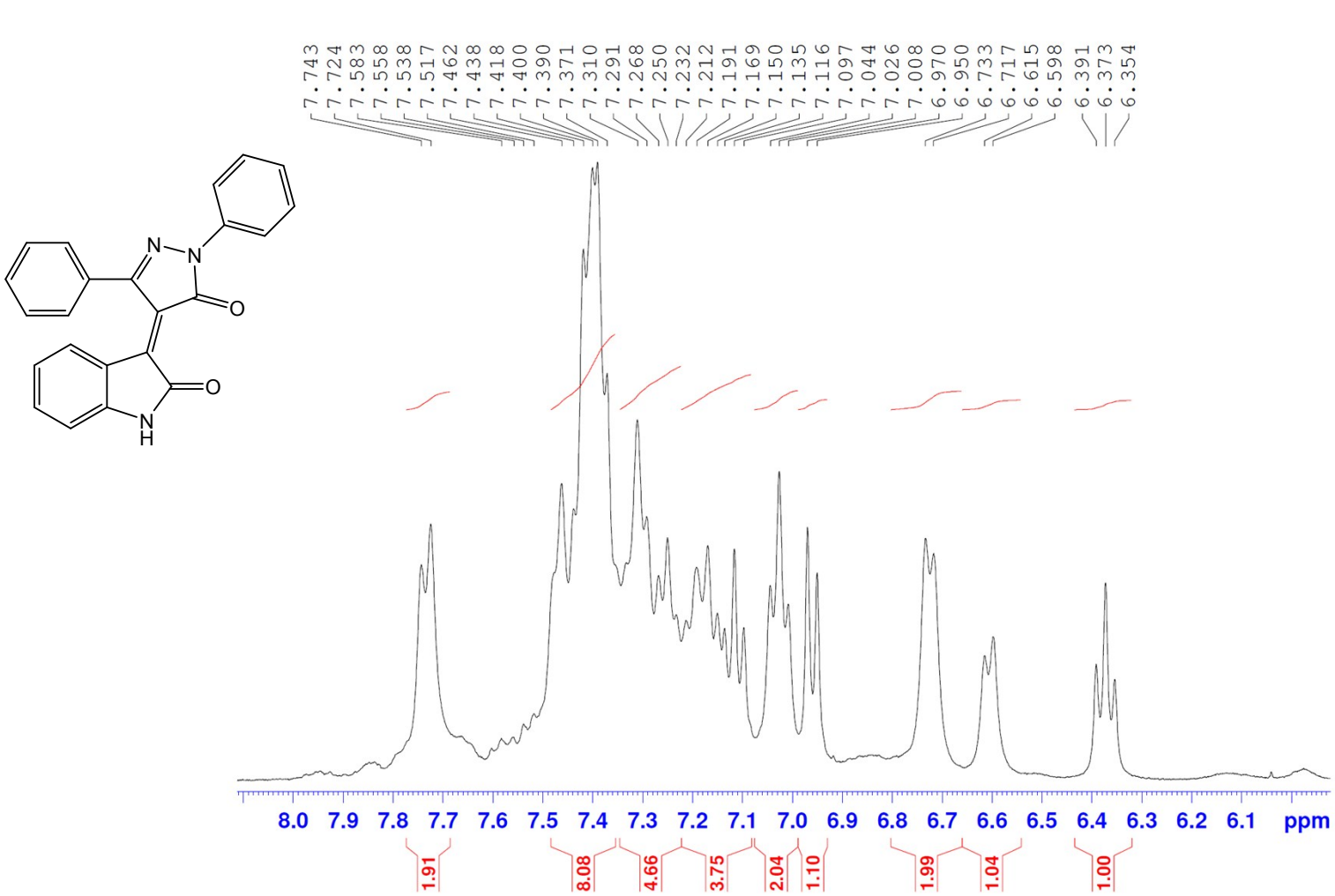





```
NAME            Gilan UN
EXPNO               3014
PROCNO                 1
Date_           20170715
Time               22.19
INSTRUM            spect
PROBHD   5 mm PABBO BB-
PULPROG             zg30
TD                 65536
SOLVENT             DMSO
NS                     20
DS                      0
SWH             8012.820 Hz
FIDRES          0.122266 Hz
AQ            4.0894966 sec
RG                    161
DW                 62.400 usec
DE                  6.50 usec
TE                 293.1 K
D1            4.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                  14.00 usec
PL1                 -2.00 dB
PL1W         11.86359406 W
SFO1         400.2236020 MHz
SI                  32768
SF           400.2200000 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   1.00
```

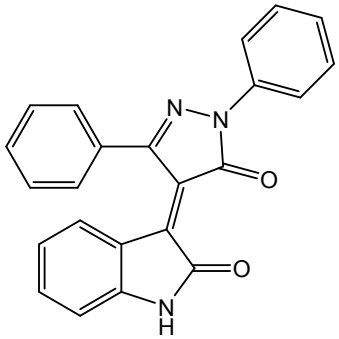
S$_{17}$ The ^{1}H NMR spectrum of 3-(5-oxo-1,3-diphenyl-1*H*-pyrazol-4(5*H*)-ylidene)indolin-2-one (**7b**)

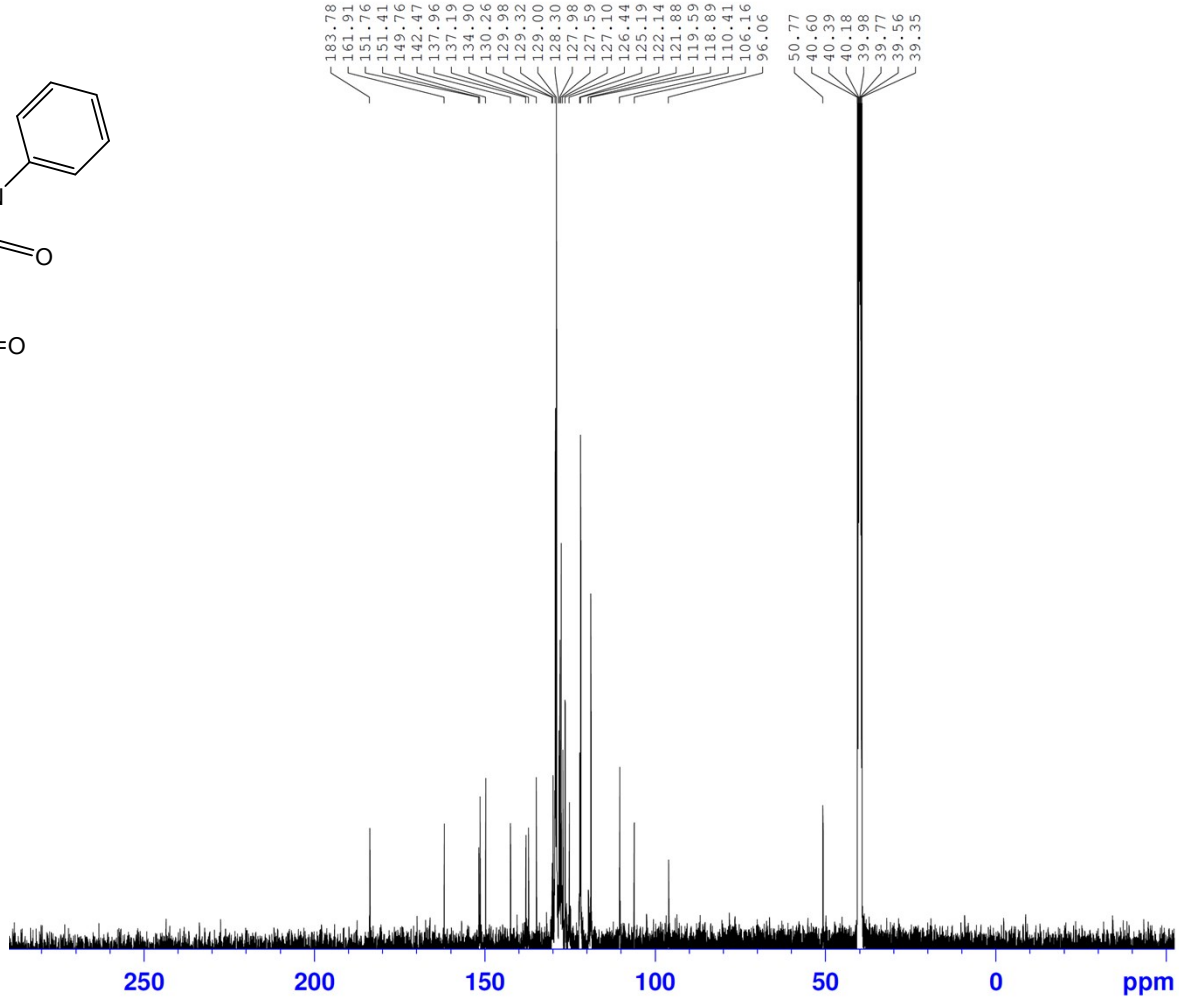



```
NAME              Gilan UN
EXPNO                3014
PROCNO                  1
Date_            20170715
Time                22.19
INSTRUM             spect
PROBHD    5 mm PABBO BB-
PULPROG              zg30
TD                  65536
SOLVENT              DMSO
NS                     20
DS                      0
SWH              8012.820 Hz
FIDRES           0.122266 Hz
AQ             4.0894966 sec
RG                    161
DW                 62.400 usec
DE                  6.50 usec
TE                 293.1 K
D1             4.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                  14.00 usec
PL1                 -2.00 dB
PL1W          11.86359406 W
SFO1          400.2236020 MHz
SI                  32768
SF            400.2200000 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   1.00
```

S18 The ^{13}C NMR spectrum of 3-(5-oxo-1,3-diphenyl-1*H*-pyrazol-4(5*H*)-ylidene)indolin-2-one (**7b**)



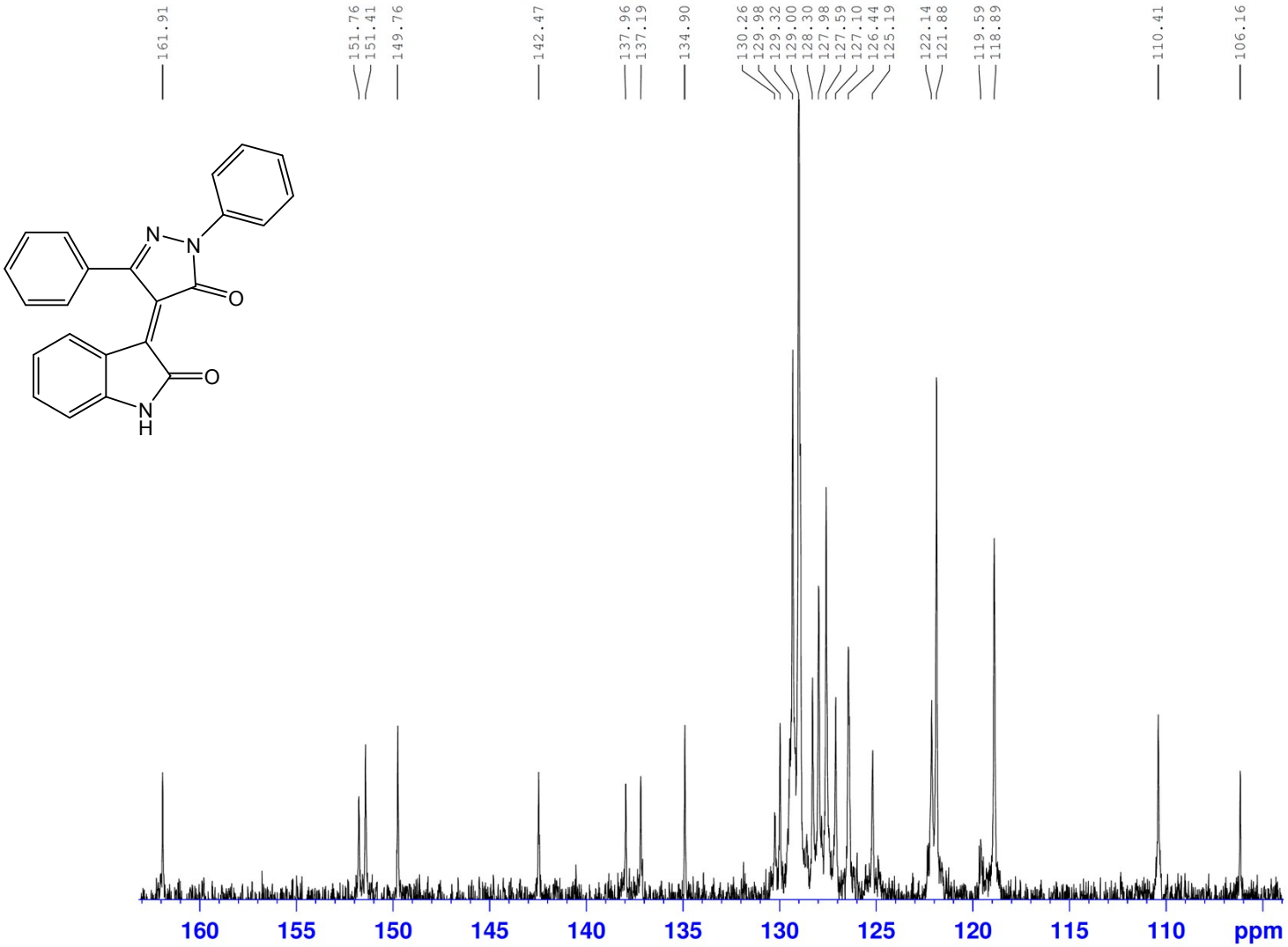



```
NAME              Gilan UN
EXPNO                 3015
PROCNO                   1
Date_             20170715
Time                 22.26
INSTRUM              spect
PROBHD    5 mm PABBO BB-
PULPROG            zgpg30
TD                   65536
SOLVENT               DMSO
NS                    8192
DS                       0
SWH              35714.285 Hz
FIDRES            0.544957 Hz
AQ               0.9175540 sec
RG                    2050
DW                  14.000 usec
DE                    6.50 usec
TE                   293.3 K
D1              1.00000000 sec
D11             0.03000000 sec
TD0                      1

======== CHANNEL f1 ========
NUC1                   13C
P1                    9.00 usec
PL1                  -0.90 dB
PL1W          42.02801895 W
SFO1          100.6479784 MHz

======== CHANNEL f2 ========
CPDPRG2            waltz16
NUC2                    1H
PCPD2                90.00 usec
PL2                  -2.00 dB
PL12                 14.16 dB
PL13                 17.90 dB
PL2W          11.86359406 W
PL12W          0.28722104 W
PL13W          0.12139934 W
SFO2          400.2216009 MHz
SI                   32768
SF            100.6353990 MHz
WDW                     EM
SSB                      0
LB                    1.00 Hz
GB                       0
PC                    1.40
```

S18 The ^{13}C NMR spectrum of 3-(5-oxo-1,3-diphenyl-1*H*-pyrazol-4(5*H*)-ylidene)indolin-2-one (**7b**)

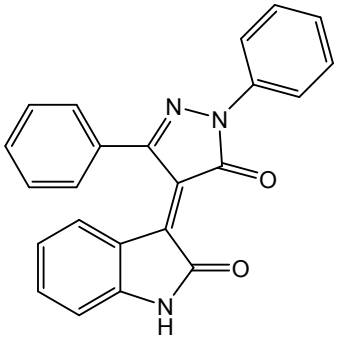


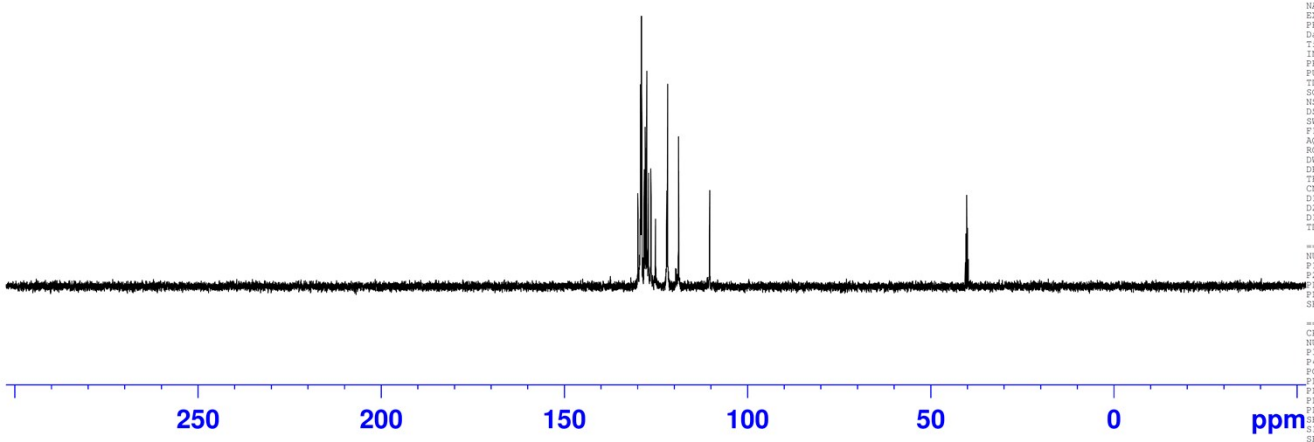

```
NAME              Gilan UN
EXPNO                3015
PROCNO                  1
Date_            20170715
Time                22.26
INSTRUM             spect
PROBHD   5 mm PABBO BB-
PULPROG           zgpg30
TD                 65536
SOLVENT             DMSO
NS                  8192
DS                     0
SWH            35714.285 Hz
FIDRES          0.544957 Hz
AQ             0.9175540 sec
RG                  2050
DW                14.000 usec
DE                  6.50 usec
TE                 293.3 K
D1           1.00000000 sec
D11          0.03000000 sec
TD0                    1

======== CHANNEL f1 ========
NUC1                 13C
P1                  9.00 usec
PL1                -0.90 dB
PL1W        42.02801895 W
SFO1        100.6479784 MHz

======== CHANNEL f2 ========
CPDPRG2          waltz16
NUC2                  1H
PCPD2              90.00 usec
PL2                -2.00 dB
PL12               14.16 dB
PL13               17.90 dB
PL2W        11.86359406 W
PL12W        0.28722104 W
PL13W        0.12139934 W
SFO2        400.2216009 MHz
SI                32768
SF          100.6353990 MHz
WDW                  EM
SSB                   0
LB                 1.00 Hz
GB                    0
PC                 1.40
```

S$_{19}$ The DEPT-135 spectrum of 3-(5-oxo-1,3-diphenyl-1*H*-pyrazol-4(5*H*)-ylidene)indolin-2-one (**7b**)



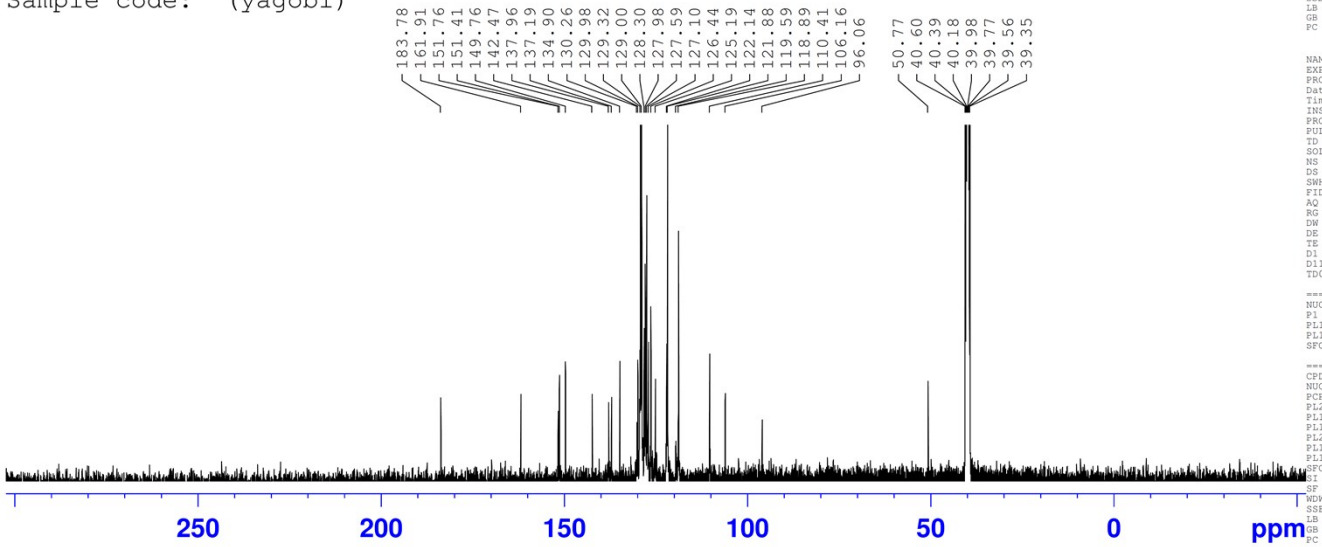



```
NAME            Gilan UN
EXPNO               3016
PROCNO                 1
Date_           20170716
Time                7.24
INSTRUM            spect
PROBHD    5 mm PABBO BB-
PULPROG          dept135
TD                65536
SOLVENT            DMSO
NS                 8192
DS                    2
SWH           35714.285 Hz
FIDRES         0.544957 Hz
AQ            0.9175540 sec
RG                 2050
DW               14.000 usec
DE                 6.50 usec
TE                293.1 K
CNST2        145.0000000
D1           1.00000000 sec
D2           0.00344828 sec
D12          0.00002000 sec
TD0                    1

======== CHANNEL f1 ========
NUC1                 13C
P1                  9.00 usec
P2                 18.00 usec
PL1                -0.90 dB
PL1W        42.02801895 W
SFO1       100.6479784 MHz

======== CHANNEL f2 ========
CPDPRG2          waltz16
NUC2                  1H
P3                 13.50 usec
P4                 27.00 usec
PCPD2              90.00 usec
PL2                -2.00 dB
PL12               14.48 dB
PL2W        11.86359406 W
PL12W        0.26681873 W
SFO2       400.2216009 MHz
SI                 32768
SF         100.6353990 MHz
WDW                   EM
SSB                    0
LB                  1.00 Hz
GB                     0
PC                  1.40
```

Sample code: (yagobi)




```
NAME            Gilan UN
EXPNO               3015
PROCNO                 1
Date_           20170715
Time               22.26
INSTRUM            spect
PROBHD    5 mm PABBO BB-
PULPROG          zgpg30
TD                65536
SOLVENT            DMSO
NS                 8192
DS                    0
SWH           35714.285 Hz
FIDRES         0.544957 Hz
AQ            0.9175540 sec
RG                 2050
DW               14.000 usec
DE                 6.50 usec
TE                293.3 K
D1           1.00000000 sec
D11          0.03000000 sec
TD0                    1

======== CHANNEL f1 ========
NUC1                 13C
P1                  9.00 usec
PL1                -0.90 dB
PL1W        42.02801895 W
SFO1       100.6479784 MHz

======== CHANNEL f2 ========
CPDPRG2          waltz16
NUC2                  1H
PCPD2              90.00 usec
PL2                -2.00 dB
PL12               14.16 dB
PL13               17.90 dB
PL2W        11.86359406 W
PL12W        0.28722104 W
PL13W        0.12139934 W
SFO2       400.2216009 MHz
SI                 32768
SF         100.6353990 MHz
WDW                   EM
SSB                    0
LB                  1.00 Hz
GB                     0
PC                  1.40
```

S$_{20}$ The mass spectrum of 3-(5-oxo-1,3-diphenyl-1*H*-pyrazol-4(5*H*)-ylidene)indolin-2-one (**7b**)