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Synthesis of 7,11-diaryl-3-oxo (or thioxo)-2,4-diazaspiro[5.5]undecane-1,5,9-triones, Part I

M. Giasuddin Ahmed[a]*, Syeda Asghari Ahmed[a], S. Mosaddeq Ahmed[a], M. Mahmun Hossain[b] and Abul Hussam[c]

[a]*Department of Chemistry, University of Dhaka, Dhaka-1000, Bangladesh*

[b]*Department of Chemistry, University of Wisconsin-Milwaukee, WI 53201,USA*

[c]*Department of Chemistry, George Mason University, Fairfax, VA 22030, USA*

Spiro compounds 7,11-diaryl-2,4-diazaspiro[5.5]undecane-1,3,5,9-tetraones **3a–e** and 7,11-diaryl-3-thioxo-2,4-diazaspiro[5.5]undecane-1,5,9-triones **3f–j** were prepared from the reaction of 1,5-diaryl-1,4-pentadien-3-ones **2a–f** with barbituric acid (**1a**) and 2-thiobarbituric acid (**1b**). The structures of the products were confirmed by UV, IR, ^{1}H and ^{13}C NMR, MS and elemental analysis.

Keywords: spiro compounds, barbituric acid, 2-thiobarbituric acid, Michael addition

A copious literature[1,2] has grown up over a period of more than a century in the field of synthesis and pharmaceutical activity of barbiturates and thiobarbiturates. Reports on 5-substituted barbituric acid and thiobarbituric acid derivatives are comparatively more numerous than those on other barbiturates, primarily because of the active methylene group at the 5-position. A large number of reports[3-15] are available on the reactions of barbituric acid (**1a**) and 2-thiobarbituric acid (**1b**) with carbonyl compounds – aldehydes, ketones and esters. In almost all these cases the active methylene group was involved in the reaction. A literature survey shows that little work[12, 16, 17] has been done on the reactions of αβ-unsaturated carbonyl systems with **1a** and **1b**. With this background, we selected a series of 1,5-diaryl-1,4-pentadien-3-ones[17] **2a-f** for reaction with **1a** and **1b** with a view to synthesising medicinally potent compounds.

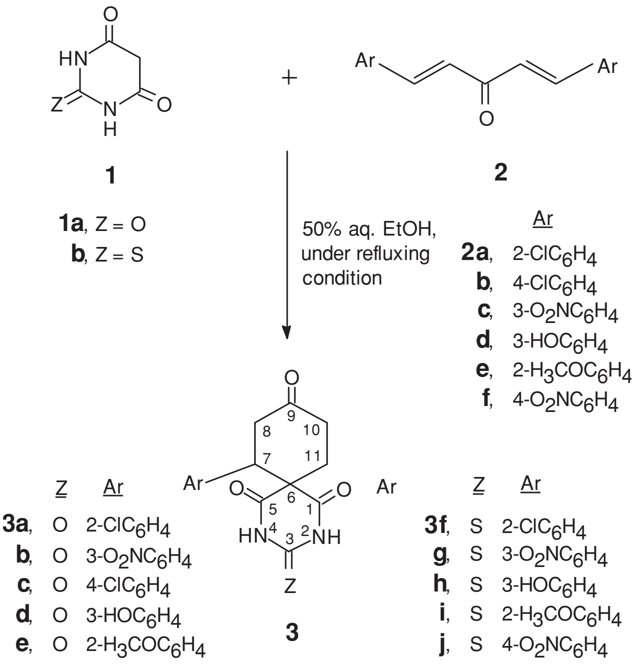
7,11-Diaryl-3-oxo-(or thioxo)-2,4-diazaspiro[5.5]undecane-1,5,9-triones **3a–j**, were obtained as products from the general reaction of **1a** or **1b** in a 1:1 molar ratio with 1,5-diaryl-1,4-pentadien-3-ones **2a–f** having different substituents on the aromatic rings under refluxing in aqueous ethanol (Scheme 1).

Compounds **3b–e** and **3g–j** seem not to have been reported in the literature. Compounds **3a** and **3h** are known;[12] we report herein their high resolution NMR (^{1}H and ^{13}C) as well as their UV spectral data.

The compounds **3a–j** were purified by recrystallisation and their purity was checked by TLC. The structures of compounds **3a–j** were established with the help of their UV, IR, NMR (^{1}H and ^{13}C), mass spectral data and elemental analyses.

In the UV spectra (see Experimental section), the observed λ_{max} values of the compounds **3a–j** correspond well to the expected values. The absorption bands in the range 261–208 nm may be attributed to $\pi \rightarrow \pi^*$ transitions of the aromatic rings. The absorption bands at 289–272 nm were due to $\pi \rightarrow \pi^*$ of the C=O for these compounds. The $n \rightarrow \pi^*$ transition due to C=O could not be observed separately. They were probably hidden beneath the $\pi \rightarrow \pi^*$ absorptions in these compounds. Similar reports are available in the literature.[2]

The IR spectra (see Experimental) of the compounds **3a–j** showed both sharp and broad bands in the range 3450–3100 cm^{-1} for the N–H groups. The compounds **3d** and **3h** in addition showed broad bands due to phenolic O–H stretching. The bands at 1760–1670 cm^{-1} in these compounds indicate the presence of non-conjugated C=O stretching including **1a** and **1b** moieties.[2,12] The bands at 1650–1505 cm^{-1} are due to C=C of aromatic rings and C=N of the conjugated form of **1a** and **1b**. Additional bands were observed at 1455–1145 cm^{-1} due to C=S stretching in thio compounds.[12]



Scheme 1

In the high resolution ^{1}H NMR spectra (Table 1) the N–H protons of the compound **3a–e** at position 2 and 4 are strongly deshielded (δ 11.98–11.20) since they are flanked by two C=O groups. The non-equivalence of these protons are caused by the anisotropy of the C-1 and C-5 carbonyl groups owing to the geometry of spiro structures.[2] The N–H protons at position 2 and 4 in the compounds **3f–j** obtained from the reaction with **1b** are more deshielded (δ 12.97–12.19) than those in the compounds **3a–e** produced in the reactions with **1a**. This may be attributed to the greater polarisability of sulfur compounds in comparison to oxygen. This causes more deshielding of the N–H protons in the sulfur compounds **3f–j**.

In the cases of these spiro compounds, the axial protons at positions 7, 8, 10 and 11 are considerably deshielded and appeared at δ 4.69–3.25. The equatorial protons at positions 8 and 10 resonated at higher field (δ 2.61–2.40) than the axial protons of these positions. This may be explained by the proximity of the axial protons at 7, 8, 10 and 11 positions to the aromatic rings present at positions 7 and 11. The protons at positions 7 and 11 in the compounds **3a–j** appeared as doublets of doublet ($J_{ax\text{-}eq}$ 2.75-5.0 Hz, $J_{ax\text{-}ax}$ 13.5–14.0 Hz) due to the

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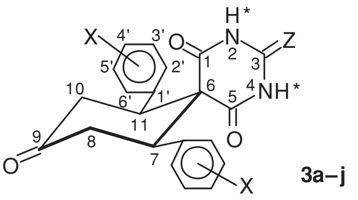
* Correspondent. E-mail: mgahmed1@yahoo.com, mgahmed@udhaka.net

Table 1 ^1H NMR spectral data of the compounds **3a–j** (δ in ppm, *J* in Hz).



3a–j

Protons	3a	3b	3c	3d	3e	3f	3g	3h	3i	3j
2-H*	11.98	11.77	11.58	11.43	11.43	12.97	12.78	12.45	12.45	12.73
4-H*	11.29	11.46	11.35	11.20	11.20	12.32	12.45	12.20	12.19	12.45
Aromatic	7.47–7.30	8.20–7.60	7.42–7.13	7.11–6.53	7.05–6.86	7.47–7.28	8.20–7.60	9.51–6.51	7.04–6.85	8.21–7.34
7-H_{ax}, 11-H_{ax}	4.66	4.27	4.01	3.84	3.89	4.69	4.30	3.86	3.92	4.28
J_{ae}	5.00	4.25	4.50	4.50	4.75	5.00	4.75	4.75	2.75	5.00
J_{aa}	13.50	13.75	14.00	14.00	13.75	13.50	13.75	13.75	13.75	14.00
8-H_{ax}, 10-H_{ax}	3.26	3.54	3.46	3.43	3.48	3.25	3.53	3.42	3.46	3.50
J_{aa}	14.25	14.00	14.00	14.00	14.50	13.50	14.50	13.50	14.50	14.50
J_{gem}	15.75	15.00	15.50	15.00	15.00	16.00	15.50	16.00	15.00	15.50
8-H_{eq}, 10-H_{eq}	2.55	2.60	2.47	2.42	2.40	2.56	2.61	2.43	2.42	2.58
J_{ae}	5.00	4.50	4.75	4.50	4.50	4.75	4.75	4.75	2.75	4.75
J_{gem}	16.00	15.50	15.75	15.50	15.50	16.25	15.75	15.75	15.25	15.75
X	–	–	–	9.52	3.71	–	–	9.51	3.71	–
	(2'-Cl)	(3'-NO_2)	(4'-Cl)	(3'-O$\underline{H}$)	(2'-OC$\underline{H_3}$)	(2'-Cl)	(3'-NO_2)	(3'-O$\underline{H}$)	(2'-OC$\underline{H_3}$)	(4'-NO_2)

* δ values of 2-H and 4-H are interchangeable

splitting by the axial and equatorial protons at positions 8 and 10 respectively. Similarly, the equatorial protons at positions 8 and 10 of these compounds also appeared as doublets of doublets ($J_{ax\text{-}eq}$ 2.75–5.0 Hz, J_{gem} 15.25–16.25 Hz) due to the geminal coupling at 8 and 10 positions as well as the vicinal coupling received from the protons at 7 and 11 positions respectively. As a result of similar coupling pattern, the doublets of doublets due to the axial protons at positions 8 and 10 looked like triplets since the axial-axial coupling constant ($J_{ax\text{-}ax}$ 13.5–14.5 Hz) is found to be comparable to the geminal coupling constants (J_{gem} 15.0–16.0 Hz).

The structures of the compounds **3a–j** were further confirmed by their ^{13}C NMR spectra (Table 2). In the compounds **3a–e**, chemical shifts of carbonyl carbons at C-3 were found to be at δ 158.1–148.3 and are relatively less deshielded due to the resonance of amide functional group. In the compounds **3f–j**, the chemical shifts of the thioxo carbon at C-3 were found to be at δ 178.4–151.9. From the above values it is clear that the replacement of a carbonyl group by a thiocarbonyl group results in a downfield shift.[18] For the compounds **3a–j**, the chemical shift values of the carbonyl carbon at position C-9 were highly deshielded (δ 208.2–205.5). This value is in good agreement with the ^{13}C NMR chemical shift of cyclohexyl methyl ketone.[19] The chemical shift values for carbonyl carbon at positions C-1 and C-5 were δ 178.4–169.75. The non-equivalence of these carbons is caused by the anisotropy of the C-1 and C-5 carbonyl groups owing to the geometry of spiro structures.[2]

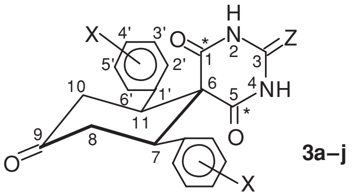
In the compounds **3a–j**, the δ values of C-7 and C-11 showed peak at 49.9–44.65. For the compounds **3a** and **3f** the values were in the range of δ 44.9–44.65. The deshielding of these carbons was due to electron withdrawing effects of chlorine at the *ortho*-position of the aryl rings. The chemical shift values of C-8 and C-10 (δ 43.9–42.1) of compounds **3a–j** were slightly lower than those of C-7 and C-11. This is due to the electron-withdrawing effect of the adjacent C=O group.

The ^{13}C shifts of carbons of aromatic rings are assigned on the basis of the correlation charts of ^{13}C NMR spectral data available in the literature.[19]

The spiro carbon C-6 of the compounds **3a–j** showed chemical shift values at δ 60.55–55.5 which are similar to the literature values.[18]

The low resolution mass spectra of the compounds **3d–e** and **3g–i** showed peaks for their respective molecular ions (M^+) at *m/z* 394, 447, 422, 468, 410 and 438 respectively. The molecular ion (M^+) peaks for the compound **3a–c, 3f** and **3j** could not be observed; rather, the corresponding $(M\text{-}1)^+$ or $(M\text{-}18)^+$ peaks at *m/z* 430, 434, 430, 446 and 450 respectively were found.

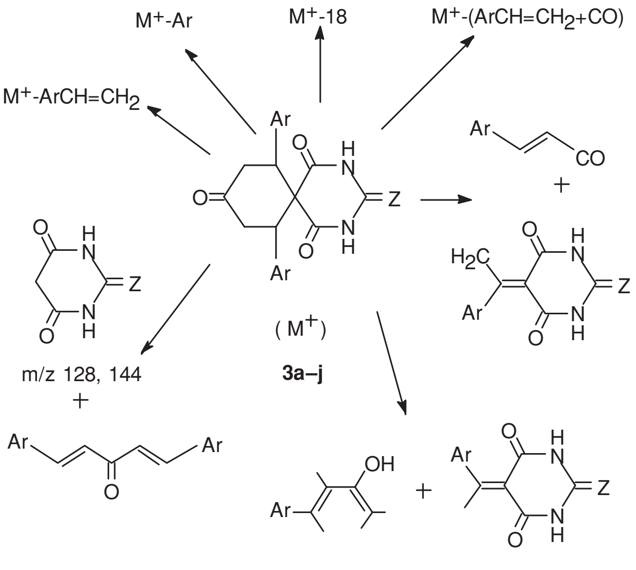
Table 2 ^{13}C NMR spectral data of the compounds **3a–j** (δ in ppm).



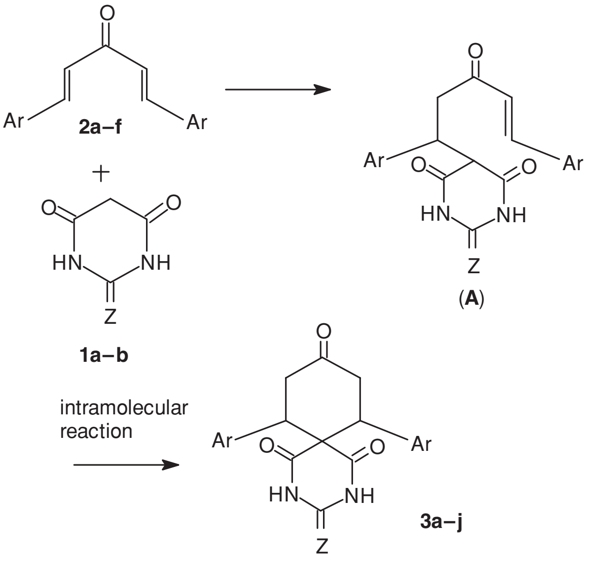
3a–j

Carbons	3a	3b	3c	3d	3e	3f	3g	3h	3i	3j
9-C	205.7	205.7	207.3	208.1	208.2	205.5	205.6	207.8	207.9	206.2
1-C*	172.0	171.5	172.3	172.5	172.8	169.7	169.2	169.4	170.8	169.8
5-C*	169.6	170.0	171.2	171.6	171.7	167.7	167.9	170.6	169.5	168.8
3-C	148.8	148.3	149.4	158.1	148.4	177.5	177.2	178.4	151.9	177.9
Aromatic	135.5–127.6	147.8–122.7	137.1–129.5	149.8–115.5	159.5–114.8	135.7–127.6	147.7–122.8	158.1–115.6	159.5–114.8	148.0–124.6
6-C	55.5	58.5	59.4	59.2	55.7	56.1	59.0	59.8	60.6	59.4
7, 11-C	44.6	47.9	48.7	49.6	48.8	44.9	48.1	49.9	49.1	49.2
8, 10-C	43.2	42.1	43.1	43.5	43.7	43.2	42.2	43.7	43.9	42.8
X	–	–	–	–	55.8	–	–	–	55.8	–
	(2'-Cl)	(3'-NO_2)	(4'-Cl)	(3'-O$\underline{H}$)	(2'-OC$\underline{H_3}$)	(2'-Cl)	(3'-NO_2)	(3'-O$\underline{H}$)	(2'-OC$\underline{H_3}$)	(4'-NO_2)

*δ values of 1-C and 5-C are interchangeable.



Scheme 2



Scheme 3

From the fragmentation patterns in these spectra it is observed that most of the prominent peaks were formed due to loss of CO, CONH/CSNH, substituted phenyl, tropylium, styryl, methyl, and keten fragments (Scheme 2). This is in conformity with the mass spectral fragmentation pattern of N-substituted 7,11-diphenyl-2,4-diazaspiro[5.5]undecane-1,3,5,9-tetraones.[20]

The ^{1}H NMR spectral data of the compounds **3a–j** as discussed earlier clearly point to a rigid diarylcyclohexanone ring in which the two aryl groups adopt equatorial positions, requiring that they are *cis*-oriented.

The difference between the stereochemical environments of the two amido groups in the pyrimidine structure is reflected in both their ^{1}H and ^{13}C NMR spectra, where the chemical shifts of the NH protons and the carbonyl carbons are found to be different. This can be understood by the perpendicular disposition of the ureide and the cyclohexanone rings and the *cis*-arrangement of the two substituted aromatic rings. In this stereochemistry the two carbonyl carbon atoms of the amide groups experience different shielding effects, one being closer to the aromatic rings and the other being remote from them. We were not able to isolate a *trans*-isomer of any of these spiro compounds.

The formation of these spiro compounds may be explained by an initial adduct (**A**) (Scheme 3) undergoing subsequent intramolecular Michael addition, leading to the compounds **3a–j**.

Experimental

^{1}H and ^{13}C NMR spectra were recorded on a JEOL 500 instrument at the Department of Chemistry, University of Wisconsin-Milwaukee, USA. The elemental analyses were also carried out there. Mass spectra of low resolution were recorded at the Department of Chemistry, George Mason University, Virginia, USA. Melting points were determined in open ended capillary tubes. All ^{1}H and ^{13}C NMR spectra were recorded in d_6 DMSO/TMS. IR spectra were run as KBr pellets on a Shimadzu IR-470 spectrophotometer. UV spectra were run in methanol using a Shimadzu UV-160A spectrophotometer.

1,5-Diaryl-1,4-pentadien-3-ones **2a–f** were prepared primarily by following the literature method,[19, 21] modifying the reaction conditions whenever necessary.[17]

7,11-Diaryl-3-oxo (or thioxo)-2,4-diazaspiro[5.5]undecane-1,5,9-triones (**3a–j**): *General method* A mixture of **1** (0.005 mol) and **2** (0.005 mol) was dissolved in rectified spirit (25 ml) and water (25 ml) in a round-bottomed flask equipped with a magnetic stirrer and a reflux condenser. The reaction mixture was heated to reflux for 18–20 h and the course of the reaction was followed by tlc on silica gel plates (eluting solvent; EtOAc:$CHCl_3$ 1:1 unless otherwise specified). The mixture was allowed to cool. The solid which separated was filtered off, dried in the air, and recrystallised from rectified spirit if not otherwise mentioned.

7,11-Bis-(2-chlorophenyl)-2,4-diazaspiro[5.5]undecane-1,3,5,9-tetraone (**3a**): White solid, m.p. 248–249 °C (lit.[12] 239–241 °C), R_f in TLC 0.46 (EtOAc:$CHCl_3$ 1:2), Yield 284 mg (16.5%). UV: λ_{max} (nm/ε) 219 (8800). IR: ν_{max} (cm^{-1}) 3240, 1730, 1700, 1670, 1587, 1520, 1420, 1400, 1372, 1326, 1286, 1220. MS: *m/z* 430 (M^+-1, 1.5%), 395 (4.9), 318 (1), 319 (1.1), 303 (1.7), 292 (5), 264 (5), 215(100), 128 (16), 111 (4), 103 (58). Anal. Calcd for $C_{21}H_{16}Cl_2N_2O_4$: C, 58.57; H, 3.71m; N 6.49. Found C, 58.50; H, 3.74; N, 6.39 %.

7,11-Bis-(3-nitrophenyl)-2,4-diazaspiro[5.5]undecane-1,3,5,9-tetraone (**3b**): Yellowish-white solid, m.p. 271–273 °C, R_f in TLC 0.46, yield 285 mg (16%). UV: λ_{max} (nm) (ε) 261 (32 250), 210 (65 700). IR: ν_{max} (cm^{-1}) 3300, 3200, 3100, 1750, 1720, 1705, 1625, 1580, 1522, 1425, 1350, 1325, 1250, 1210, 1150. MS: *m/z* 434 (M^+-18) (3), 324 (10), 303 (8), 275 (17), 149 (25), 128 (66), 122 (0.8), 102 (100). Anal. Calcd for $C_{21}H_{16}N_4O_8$: C, 55.75; H, 3.54; N, 12.39. Found: C, 54.86; H, 3.60; N, 11.78 %.

7,11-Bis-(4-chlorophenyl)-2,4-diazaspiro[5.5]undecane-1,3,5,9-tetraone (**3c**): White solid, m.p. 279–280 °C, R_f in TLC 0.63, yield 149 mg (8.6%). UV: λ_{max} (nm) (ε) 225 (11 130). IR: ν_{max} (cm^{-1}) 3300, 3200, 3100, 1750, 1720, 1680, 1590, 1540, 1520, 1405, 1365, 1325, 1290, 1240, 1220, 1120. MS: *m/z* 430 (M^+-1) (2), 305 (1), 292 (1.5), 264 (24), 128 (3.4), 138 (100), 103 (25), 111 (2). Anal. Calcd for $C_{21}H_{16}Cl_2N_2O_4$: C, 58.47; H, 3.71; N, 6.49. Found: C, 58.51; H, 3.72; N, 6.41 %.

7,11-Bis-(3-hydroxyphenyl)-2,4-diazaspiro[5.5]undecane-1,3,5,9-tetraone (**3d**): Off-white solid from aqueous ethanol, m.p. 248–250 °C, R_f in TLC 0.20, yield 1.2 g (61%). UV: λ_{max} (nm) (ε) 277 (3510), 219 (7720). IR: ν_{max} (cm^{-1}) 3350, 3204, 1744, 1706, 1675, 1590, 1490, 1455, 1430, 1395, 1370, 1315, 1220, 1270, 1230, 1155. MS: *m/z* 394 (M^+) (4.5), 275 (14), 266 (24), 246 (18.5), 128 (50), 103 (12.6), 120(100). Anal. Calcd for $C_{21}H_{18}N_2O_6$: C, 63.96; H, 4.57; N, 7.11. Found: C, 63.38; H, 4.74; N, 6.95 %.

7,11-Bis-(2-methoxyphenyl)-2,4-diazaspiro[5.5]undecane-1,3,5,9-tetraone (**3e**): White solid, m.p. 235–237 °C, R_f in TLC 0.54 (EtOAc: $CHCl_3$, 1:2), yield 120 mg (9.4%). UV: λ_{max} (nm) (ε) 276 (2700), 227 (17 400). IR: ν_{max} (cm^{-1}) 3250, 1750, 1700, 1650, 1605, 1510, 1410, 1370, 1325, 1260, 1245, 1220, 1180. MS: *m/z* 422 (M^+) (6.5), 295 (2), 260 (25), 229 (5), 134 (100), 107 (2.5), 103 (15). Anal. Calcd for $C_{23}H_{22}N_2O_6$: C, 65.40; H, 5.21; N, 6.64. Found: C, 65.00; H, 5.19; N, 6.54 %.

7,11-Bis-(2-chlorophenyl)-2,4-diazaspiro[5.5]undecane-3-thioxo-1,5,9-trione (**3f**): Light yellow solid, m.p. 230–232 °C (lit.[12] 215–217 °C), R_f in TLC 0.67 (EtOAc:$CHCl_3$, 1:2), yield 725 mg (40%). UV: λ_{max} (nm) (ε) 287 (17 000), 216 (14 250). IR: ν_{max} (cm^{-1}) 3450, 3225, 1760, 1715, 1687, 1590, 1530, 1412, 1370, 1320, 1232, 1215, 1150. MS: *m/z* 446 (M^+-1) (1.3), 411 (3), 308 (4), 303 (1.2), 280 (4.2), 273 (25.4), 245 (36), 231(100), 138 (45), 144 (6), 111 (3.4). Anal.

Calcd for $C_{21}H_{16}Cl_2N_2O_3S$: C, 56.38; H, 3.58; N, 6.26. Found: C, 56.11; H, 3.53; N, 6.10 %.

7,11-Bis-(3-nitrophenyl)-3-thioxo-2,4-diazaspiro[5.5]undecane-1,5,9-trione (**3g**): Off-white solid, m.p. 266–268 °C, R_f in TLC 0.61, yield 55 mg (2.9%). UV: λ_{max} (nm) (ε) 287 (27 600), 209 (40 100). IR: ν_{max} (cm^{-1}) 3440, 3300, 3200, 1726, 1693, 1610, 1580, 1523, 1420, 1350, 1320, 1280, 1245, 1205, 1150. MS: *m/z* 468 (M^+) (1.1), 348 (0.4), 325 (1.5), 319 (10), 291 (26), 149 (16.4), 144 (77), 122 (0.85), 102(100). Anal. Calcd for $C_{21}H_{16}N_4O_7S$: C, 53.80; H, 3.42; N, 11.97. Found C, 53.62; H, 3.41; N, 11.74 %.

7,11-Bis-(3-hydroxyphenyl)-3-thioxo-2,4-diazaspiro[5.5] undecane-1,5,9-trione (**3h**): Off-white solid (from aqueous ethanol), m.p. 244–246 °C, R_f in TLC, 0.35, yield 820 mg (40%). UV: λ_{max} (nm) (ε) 282 (21 050), 216 (16 370). IR: ν_{max} (cm^{-1}) 3350, 3150, 1718, 1672, 1583, 1510, 1418, 1360, 1320, 1250, 1145. MS: *m/z* 410 (M^+) (8.3), 291 (21), 266 (17.5), 262 (17), 144 (82), 120(100), 103 (22). Anal. Calcd for $C_{21}H_{18}N_2O_5S$: C, 61.46; H, 4.39; N, 6.83. Found C, 61.02; H, 4.30; N, 6.63 %.

7,11-Bis-(2-methoxyphenyl)-3-thioxo-2,4-diazaspiro[5.5] undecane-1,5,9-trione (**3i**): White solid, m.p. 244–246 °C, R_f in TLC 0.67 (EtOAc:$CHCl_3$, 1:2), yield 204 mg (12%). UV: λ_{max} (nm) (ε) 289 (17 550), 228 (21 700). IR: ν_{max} (cm^{-1}) 3425, 3200, 1727, 1712, 1693, 1605, 1505, 1415, 1362, 1318, 1242, 1180, 1145. MS: *m/z* 438 (M^+) (3.5), 330 (0.35), 304 (0.23), 294 (0.6), 276 (7.8), 144 (2.5), 134(100), 107 (0.8), 103 (8). Anal. Calcd for $C_{23}H_{22}N_2O_5S$: C, 63.01; H, 5.02; N, 6.39. Found: C, 63.08; H, 4.82; N, 6.14 %.

7,11-Bis-(4-nitrophenyl)-3-thioxo-2,4-diazaspiro[5.5]undecane-1,5,9-trione (**3j**): Dark brown solid (from EtOAc), m.p. 260–262 °C, R_f in TLC 0.61, yield 250 mg (13%). UV: λ_{max} (nm) (ε) 272 (49 500), 208 (28 800). IR: ν_{max} (cm^{-1}) 3300, 3200, 1725, 1692, 1597, 1515, 1415, 1345, 1315, 1245, 1145. MS: *m/z* 450 (M^+-18) (0.74), 319 (5.9), 291 (38), 149 (20), 144 (100), 122 (0.8), 103 (68). Anal. Calcd for $C_{21}H_{16}N_4O_7S$: C, 53.85, H, 3.42, N, 11.97. Found C, 53.79, H, 3.49, N, 11.07 %.

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