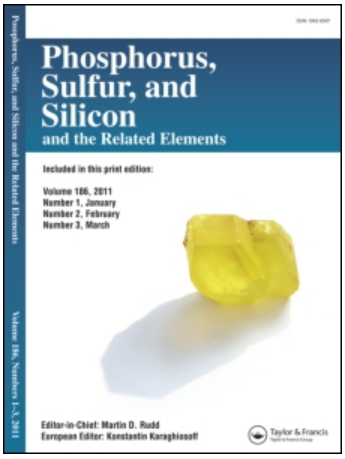

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SYNTHESIS OF SOME NEW SPIROHETEROCYCLES RELATED TO SPIRO[INDOLINE-3,2'-[1,3]OXATHIALANE]-2,5'-DIONE

Marzoog S. Al-thebeiti[a]; Maher F. El-zohry[ab]
[a] Department of Chemistry, Faculty of Applied Sciences, Umm Al-Qura University, Saudi Arabia [b] Department of Chemistry, Assuit University, Assiut, Egypt

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SYNTHESIS OF SOME NEW SPIROHETEROCYCLES RELATED TO SPIRO[INDOLINE-3,2′-[1,3]OXATHIALANE]-2,5′-DIONE

MARZOOG S. AL-THEBEITI† and MAHER F. EL-ZOHRY‡

Department of Chemistry, Faculty of Applied Sciences, Umm Al-Qura University, Makkah Almukarramah P.O. Box 6876, Saudi Arabia

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The reaction of indole-2,3-dione (**1a**) and/or 1-methylindole-2,3-dione (**1b**) with mercaptoacetic acid in the presence of 4-toluenesulfonic acid as a catalyst afforded spiro[indoline-3,2′-[1,3]oxathialane]-2,5′-diones (**2a, b**). Compounds **2a** and/or **2b** were reacted with the appropriate aliphatic and/or aromatic primary amines to give compounds **3a–j**. The reaction of **2a, b** with hydrazine hydrate, phenylhydrazine and *p*-nitrophenylhydrazine yielded spiroderivatives **4a–c**. Friedel-Crafts reactions of **2a** and **2b** with arenes in the presence of aluminium chloride catalyst afforded spiroindoline isothiochroman derivatives **5a–h**.

Key words: Spiroindoline derivatives; spiroheterocycles; NMR.

INTRODUCTION

Certain spiro derivatives exhibited photochromic properties, biological activity and optical activity.[1–6] The synthesis and photochromism of indolinospirochromenes containing condensed fragments in the indoline part of the molecule was achieved.[7] Photochromic spiro[2H-1,4-benzoxazine-2,2′-indolines] derivatives,[8] photochromic spiro[indolinenaphthoxazines] with reddish purple colors and durability,[9,10] fluorenespirophthalide compounds as leuco dyes and recording material derivatives have been prepared.[11] Thiazolidinones derivatives have considerable commercial importance as drugs (e.g. bactericidal, pesticidal, fungicidal, insecticidal, anticonvulsant, tuberculostatic, antiinflammatory, antithyroidal, and potentiation of pentobarbital induced sleeping time).[12] Divers biological activities have been encountered in compounds having the indole ring system.[13–16] To date no previous authors have reported the synthesis of spiroindolines in conjunction with thiazolidines, thiadiazines and isothiochromans moieties.

As a continuation of our previous work,[17–19] the synthesis of some new spirothiazolinones, spirothiadiazines and spiroisothiochromans incorporated with indoline moiety is reported.

RESULTS AND DISCUSSIONS

Spiro[indoline-3,2′[1,3]oxathialane]-2,5′-diones (**2a, b**) were prepared[17] starting with indol-2,3-dione (**1**). The structure of compounds **2a, b** was established from elemental analyses and spectroscopic data. The IR spectrum of **2a** showed characteristic strong absorption bands at 3150 cm^{-1} corresponding to the stretching vi-

†To whom all correspondence should be addressed.
‡Permanent address: Department of Chemistry, Assuit University, Assiut, Egypt.

brations of the NH group of the indole ring, 2980 cm^{-1} for aromatic carbon-hydrogen stretching, 1710 cm^{-1} for the carbonyl group and 730 cm^{-1} for the carbon-sulfur stretching. The ^{1}H-NMR spectrum of **2a** (DMSO-d_6) showed the following signals: δ 3.36 (2 H, s) for the methylene protons at C_4 of the 1,3-oxathialane ring,

TABLE I

Physical data of spiroindoline derivatives **2a, b–5a–h**

Compd. No.	Yield (%)	Molecular formula* (solvent of crystallization)	MP °C	IR (KBr), cm^{-1}	^{1}H NMR (DMSO-d_6), δ (TMS)ppm
2a	75	$C_{10}H_7NO_3S$ (ethanol)	200-202	3150 (NH), 2980 (CH arom), 2890 (CH aliph), 1710 (C=O), 730 (C–S).	3.36 (2 H, s), 7.00 -7.50 (4 H, m), 10.80 (1 H, s).
2b	77	$C_{11}H_9NO_3S$ (ethanol)	218-220	3020 (CH arom), 2890 (CH aliph), 1700 (C=O), 710 (C–S).	2.85 (3 H, s), 3.36 (2 H, s), 7.00 -7.20 (4 H, m).
3a	70	$C_{11}H_{10}N_2O_2S$ (ethanol)	140-142	3150 (NH), 3020 (CH arom), 2900 (CH aliph), 1710 (C=O), 710 (C–S).	2.80 (3 H, s), 3.38 (2 H, s), 7.00 -7.50 (4 H, m), 10.70 (1 H, s).
3b	67	$C_{16}H_{12}N_2O_2S$ (ethanol/ water 3:1)	220-222	3160 (NH), 3030 (CH arom), 2900 (CH aliph), 1715 (C=O), 710 (C–S).	3.38 (2 H, s), 7.00 -7.70 (9 H, complex), 10.70 (1 H, s).
3c	65	$C_{17}H_{14}N_2O_2S$ (ethanol)	260-262	3100 (NH), 3030 (CH arom), 2980 (CH aliph), 1710 (C=O), 730 (C–S).	2.30 (3 H, s), 3.36 (2 H, s), 7.00 -7.90 (8 H, complex), 10.00 (1 H, s).
3d	70	$C_{16}H_{11}N_2O_2Cl$ (ethanol)	230-232	3150 (NH), 3020 (CH arom), 2930 (CH aliph), 1720 (C=O), 700 (C–S).	3.38 (2 H, s), 7.00 -7.95 (8 H, complex), 10.50 (1 H, s).
3e	72	$C_{20}H_{14}N_2O_2S$ (ethanol)	190-192	3150 (NH), 3030 (CH arom), 2900 (CH aliph), 1720 (C=O), 700 (C–S).	3.40 (2 H, s), 7.00 -8.15 (11 H, complex), 10.10 (1 H, s).
3f	65	$C_{12}H_{12}N_2O_2S$ (ethanol)	160-162	3050 (CH arom), 2980 (CH aliph), 1720 (C=O), 710 (C–S).	2.80 (6 H, s), 3.40 (2 H, s), 7.00 -7.70 (4 H, m).
3g	65	$C_{17}H_{14}N_2O_2S$ (ethanol)	150-152	3015 (CH arom), 2980 (CH aliph), 1720 (C=O), 710 (C–S).	3.00 (3 H, s), 3.40 (2 H, s), (2 H, s), 7.00 -7.80 (8 H, complex).
3h	63	$C_{18}H_{16}N_2O_2S$ (ethanol)	220-222	3030 (CH arom), 2980 (CH aliph), 1715 (C=O), 710 (C–S).	2.30 (3 H, s), 3.00 (3 H, s), 3.40 (2 H, s), 7.00 -8.15 (8 H, complex).
3i	60	$C_{17}H_{13}N_2O_2Cl$ (ethanol)	260-262	3020 (CH arom), 2980 (CH aliph), 1720 (C=O), 710 (C–S).	2.30 (3 H, s), 3.40 (2 H, s), 7.00 -8.15 (8 H, complex).
3j	60	$C_{21}H_{16}N_2O_2S$ (ethanol)	210-212	3050 (CH arom), 2980 (CH aliph), 1715 (C=O), 710 (C–S).	2.30 (3 H, s), 3.40 (2 H, s), 7.00 -8.15 (11 H, complex).
4a	70	$C_{10}H_9N_3O_2S$ (ethanol)	200-202	3180 (NH), 3080 (CH arom), 2950 (CH aliph), 1690 (C=O), 730 (C–S).	3.40 (2 H, s), 7.00 -7.50 (4 H, m), 9.50 (2 H, d). 10.00 (1 H, s).
4b	65	$C_{16}H_{13}N_3O_2S$ (ethanol)	212-214	3180 (NH), 3030 (CH arom), 2980 (CH aliph), 1690 (C=O), 710 (C–S).	3.40 (2 H, s), 7.00 -8.10 (9 H, complex), 9.50 (1 H, s), 10.10 (1 H, s).
4c	60	$C_{16}H_{12}N_3O_4S$ (ethanol)	260-262	3180 (NH), 3030 (CH arom), 2985 (CH aliph), 1700 (C=O), 720 (C–S).	3.40 (2 H, s), 7.00 -8.10 (8 H, complex), 9.50 (1 H,s), 10.00 (1 H, s).

TABLE I (*Continued*)

Compd. No.	Yield (%)	Molecular formula* (solvent of crystallization)	MP °C	IR (KBr), cm^{-1}	^1H NMR (DMSO-d_6), δ (TMS) ppm
4d	70	$C_{11}H_{11}N_3O_2S$ (ethanol)	230-232	3150 (NH), 3020 (CH arom), 2975 (CH aliph), 1690 (C=O), 710 (C–S).	2.30 (3 H, s), 3.40(2 H, s), 7.00 -7.70 (4 H, m), 9.60 (2 H, d).
4e	72	$C_{17}H_{15}N_3O_2S$ (ethanol)	240-242	3150 (NH), 3050 (CH arom), 2980 (CH aliph), 1710 (C=O), 730 (C–S).	2.35 (3 H, s), 3.40(2 H, s), 7.00 -7.50 (9 H, complex). 9.60 (1 H, s).
4f	70	$C_{17}H_{14}N_4O_4S$ (ethanol)	272-274	3180 (NH), 3030 (CH arom), 2980 (CH aliph), 1700 (C=O), 730 (C–S).	2.40 (3 H, s), 3.40 (2 H, s), 7.00 -8.10 (8 H, complex), 9.40 (1 H, s).
5a	70	$C_{16}H_{11}NO_2S$ (ethanol)	170-172	3170 (NH), 3050 (CH arom), 2980 (CH aliph), 1720 (C=O), 730 (C–S).	3.36 (2 H, s), 7.00 -7.90 (8 H, complex), 9.60 (1 H, s).
5b	69	$C_{17}H_{13}NO_2S$ (ethanol)	175-177	3150 (NH), 3030 (CH arom), 2980 (CH aliph), 1710 (C=O), 710 (C–S).	2.40 (3 H, s), 3.4 (2 H, s), 7.00 -7.70, (7 H, complex), 9.50 (1 H, s).
5c	67	$C_{17}H_{13}NO_3S$ (ethanol)	190-192	3170 (NH), 3030 (CH arom), 2990 (CH aliph), 1720 (C=O), 730 (C–S).	3.20 (3 H, s), 3.40 (2 H, s), 7.00 -7.50 (7 H, m), 9.50 (1 H, s).
5d	65	$C_{18}H_{15}NO_2S$ (ethanol)	230-232	3150 (NH), 3050 (CH arom), 2980 (CH aliph), 1720 (C=O), 720 (C–S).	2.40 (6 H, s), 3.36 (2 H, s), 7.00 -7.50 (6 H, m), 9.50 (1 H, s).
5e	60	$C_{16}H_{13}NO_2S$ (ethanol)	180-182	3030 (CH arom), 2980 (CH aliph), 1710 (C=O), 730 (C–S).	2.80 (3 H, s), 3.40 (2 H, s), 7.00 -7.50 (8 H, complex).
5f	65	$C_{17}H_{15}NO_2S$ (ethanol)	187-190	3020 (CH arom), 2975 (CH aliph), 1700 (C=O), 730 (C–S).	2.40 (3 H, s), 2.8 (3 H, s), 3.40 (2 H, s), 7.00 -7.50 (7 H, complex).
5g	60	$C_{18}H_{15}NO_3S$ (ethanol)	210-212	3050 (CH arom), 2980 (CH aliph), 1615 (C=O), 730 (C–S).	2.80 (3 H, s), 3.20 (3 H, s), 7.00 -7.70 (7H, complex).
5h	60	$C_{19}H_{17}NO_2S$ (ethanol)	234-236	3050 (CH arom), 2990 (CH aliph), 1710 (C=O), 710 (C–S).	2.85 (3 H, s), 3.20 (6 H, s), 3.40 (2 H, s), 7.00 -7.50 (6 H, complex).

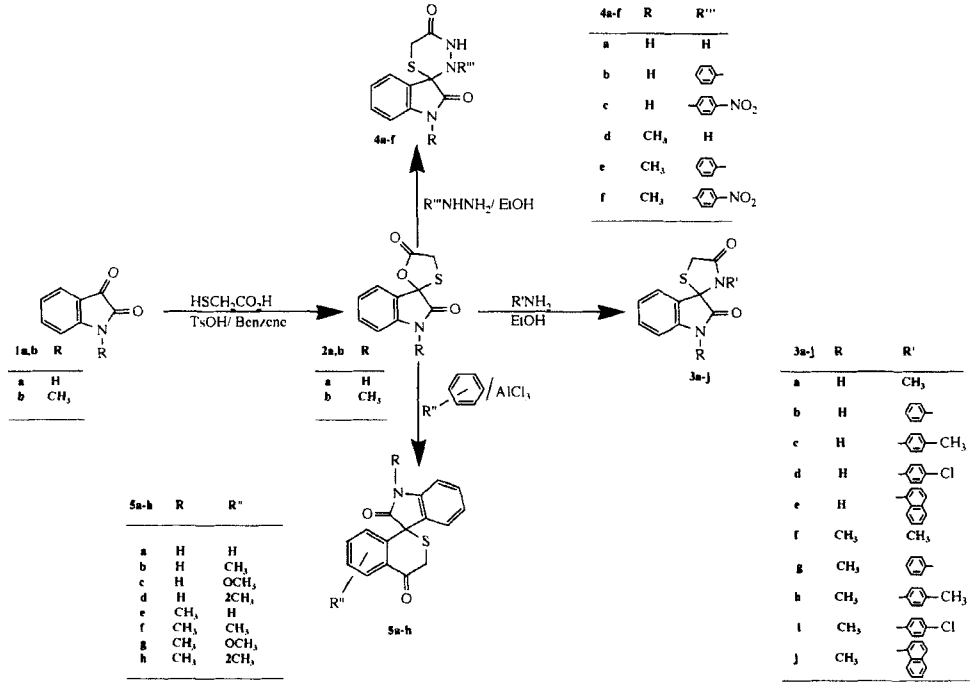
* All the prepared compounds gave satisfactory elemental analyses.

7.00–7.50 (4 H, m) corresponding to the four protons of indole moiety and 10.80 ppm (1 H, s) for the NH proton of indole ring.

Compounds **2a** and/or **2b** were reacted with the appropriate aliphatic and/or aromatic primary amine in ethanol to afford the target products **3a–j** in almost 60–72% yields (Table I). The structure of compounds **3a–j** were confirmed on the basis of elemental analyses and spectroscopic data (Table I). The ^1H-NMR spectrum of **3a** (DMSO-d_6) showed the following signals: δ 2.80 (3 H, s) for the three protons of the N-methyl group of thiazolidine ring, 3.38 (2 H, s) for the methylene protons at C_5 of thiazolidine ring, 7.00–7.50 (4 H, m) corresponding to the four protons of the indole ring and 10.70 ppm for the NH proton of the indole residue. Reaction of spiro compound with hydrazine hydrate, phenylhydrazine and *p*-nitrophenylhydrazine yielded 3′,4′-dihydrospiro[indoline-3,2′-[2H-1,3,4]thiadiazine]-2,5′(6′H)-dione (**4a**), 4′-hydro-3′-phenylspiro[indoline-3,2′[2H-1,3,4]thiadiazine]-2,5′(6′H)-dione (**4b**) and 4′-hydro-3′-(4-nitrophenyl)spiro[indoline-3,2′[2H-1,3,4]thiadiazine]-

2,5′(6′H)-dione (**4c**) in good yields (Scheme I, Table I). The 1-methylspiro derivative **2b** was reacted with hydrazine hydrate, phenylhydazine and *p*-nitrophenylhydrazine to give **4d**, **4e** and **4f** in good yields (Scheme I, Table I). The structure assignment of the prepared compounds **4a–f** was elucidated by their elemental and spectral analyses (Table I). The IR spectrum of **4a** showed characteristic strong absorption bands at 3180 cm^{-1} corresponding to the stretching vibrations of NH group of thiadiazine ring, 1690 cm^{-1} for the carbonyl group stretching of thiadiazine ring and 730 cm^{-1} for the carbon-sulfur bond of thiadiazine ring. The ^{1}H-NMR spectrum of **4a** (DMSO-d_6) showed the following signals: δ 3.40 (2 H, s) for the methylene protons at C_6 of thiadiazine ring, 7.00–7.50 (4 H, m) for the four protons of indole residue, 9.50 (2 H, d) for the two protons of the NH groups at the three and four positions of the thiadiazine ring and 10.10 (1 H, s) for the NH proton of indole moiety.

Friedel-Crafts reactions of spiro[indoline-3,2′[1,3]oxathialane-2,5′-diones[17] **2a** and **2b** with arenes in the presence of aluminium chloride catalyst afforded spiroindoline isothiochroman derivatives **5a–h**. For example, reaction of **2a** with benzene, toluene, anisole and *p*-xylene in the presence of aluminium chloride yielded spiro[indoline-3,1′-isothiochroman]-2,4′-dione (**5a**), 6′-methylspiro[indoline-3,1′-isotiochroman]2,4′-dione (**5b**), 6′-methoxyspiro[indoline-3,1′-isothiochroman]-2,4′-dione (**5c**), and 5′,8′-dimethylspiro[indoline-3,1′-isothiochroman]-2,4′-dione (**5d**), respectively, in good yields. Similarly 1-methylspiro[indoline-3,2′-[1,3]oxathialane]-2,5′-dione (**2b**) reacted with the same arenes to afford the spiroindolineisothiochroman derivatives **5e–h**, respectively (Scheme I). All prepared compounds were



(Scheme I)

identified by conventional methods such as elemental and spectral analyses (IR and ^{1}H-NMR spectra). The physical properties and spectral analysis have been given in Table I. The ^{1}H-NMR spectrum of **5a** (DMSO-d_6) showed the following signals: δ 3.36 (2 H, s) for the methylene protons at C_3 of isothiochroman ring, 7.00–7.90 (8 H, complex) for the aromatic protons in both the isothiochroman moiety and the indole ring residue and 9.60 (1 H, s) for the NH proton of indole ring.

EXPERIMENTAL

The time required for completion of the reaction was monitored by thin layer chromatography (TLC). Melting points are uncorrected. ^{1}H-NMR spectra were measured on EM-360 90-MHz spectrophotometer. IR spectra were recorded on a Pye-Unicam Sp 200-G spectrophotometer. Elemental analyses were determined on a Perkin-Elmer 240 C microanalyser.

Synthesis of spiro[indoline-3,2'-[1,3]oxathialane]-2,5'-dione (**2a**) *and 1-methylspiro[indoline-3,2'-[1,3]oxathialane]-2,5'-dione* (**2b**). *General Procedure*: A mixture of isatin (**1a**) and/or *N*-methylisatin (**1b**) (0.01 mole), thioglycolic acid (0.01 mole) and *p*-toluenesulfonic acid (0.00001 mole) in dry benzene and/or dry toluene was refluxed whereby the calculated volume of the liberated water was removed by a water separator. The reaction mixture was poured into evaporating dish whereby the solvent was evaporated, and the product was collected by filtration and then crystallized from the proper solvent. (Table I).

Spiro[indoline-3,2'-[1,3]oxathialane]-2,5'-dione (**2a**). A mixture of 14.7 g (0.01 mole) of isatin, 7 ml (0.01 mole) of thioglycolic acid and a catalytic amount of *p*-toluenesulfonic acid in 15 ml dry benzene was refluxed for 4 hrs according to general procedure to yield 16.57 g (75% yield) of **2a**. The product was crystallized from ethanol. m.p. 200–202°C, IR (KBr): 3150 (NH), 2980 (CH arom), 2890 (CH aliph), 1710 (C=O) and 730 (C—S), ^{1}H-NMR (DMSO-d_6): δ 3.36 (2 H, s), 7.00–7.50 (4 H, m), 10.80 (1 H, s).

Reaction of spiro[indoline-3,2'-[1,3]oxathialane]-2,5'-dione (**2a**) *and 1-methylspiro[indoline-3,2'-[1,3]oxathialane]-2,5'-dione* (**2b**) *with primary aliphatic (aromatic) amines: Synthesis of spiro[indoline-3,2'-thiazolidine]-2,4'-dione derivatives* (**3a–j**). *General Procedure*: A mixture of spiro derivatives **2a** and/or **2b** (0.001 mole) and primary aliphatic (aromatic) amine (0.001 mole) in absolute ethanol (10 ml) was stirred at room temperature and/or refluxed on a water bath for 1 hr. The reaction mixture was cooled and concentrated by distillation, and the product was collected by filtration and then crystallized from the proper solvent (cf. Table I).

3-Methylspiro[indoline-3,2'-thiazolidine]-2,4'-dione derivatives (**3a**). A mixture of **2a** (0.22 g, 0.001 mole) and methylamine (0.031 g, 0.001 mole) in absolute ethanol (10 ml) was allowed to react according to the above general procedure to give 0.16 g (70% yield) of **3a**. The product was crystallized from ethanol; m.p. 140–142°C; IR (KBr): 3150 (NH), 3020 (CH arom), 2900 (CH aliph), 1710 (C=O) and 710 cm^{-1} (C—S); ^{1}H-NMR (DMSO-d_6): δ 2.80 (3 H, s), 3.38 (2 H, s), 7.00–7.50 (4 H, m), 10.70 (1 H, s).

Synthesis of spiro[indoline-3,2'-[2H-1,3,4]thiadiazine]-2,5'(6'H)-dione derivatives (**4a–f**). *General Procedure*: A mixture of spiro derivatives **2a** and/or **2b** (0.001 mole) and hydrazine hydrate, phenylhydrazine or *p*-nitrophenylhydrazine (0.001 mole) in absolute ethanol (10 ml) was refluxed on a water bath for 1 hr. The reaction mixture was cooled to room temperature and kept at 0°C overnight to afford the products **4a–f**, which were collected by filtration and crystallized from the proper solvent. (cf. Table I).

Spiro[indoline-3,2'-[2H-1,3,4]thiadiazine]-2,5'(6'H)-dione (**4a**). A mixture of **2a** (0.22 g, 0.001 mole) and hydrazine hydrate 0.032 g (0.001 mole) in absolute ethanol (10 ml) was refluxed on a water bath for 1 hr according to general procedure to yield 0.16 g (70% yield) of **4a**. The product was crystallized from ethanol, m.p. 200–202°C, IR (KBr): 3180 (NH), 3080 (CH arom), 2950 (CH aliph), 1690 (C=O) and 730 cm^{-1} (C—S); ^{1}H-NMR (DMSO-d_6): δ 3.40 (2 H, s), 7.00–7.50 (4 H, m), 9.50 (2 H, d), 10.00 (1 H, s).

Reaction of spiro[indoline-3,2'-[1,3]-oxathialane]-2,5'-dione (**2a**) *and 1-methylspiro[indoline-3,2'-[1,3]oxathialane]-2,5'-dione* (**2b**) *with arenes in the presence of aluminium chloride catalyst; synthesis of spiro[indoline-3,1'-isothiochroman]-2,4'-dione derivatives* (**5a–h**). *General Procedure*: A sample of 0.044 mole of $AlCl_3$ was added to a solution of 0.01 mole of **2a** or **2b** in 25 ml of the arene in a two-necked flask equipped with a reflux condenser (capped with a calcium chloride tube), a magnetic stirrer, and a dropping funnel. The reaction mixture was stirred for 48 hr at room temperature, decomposed with 10% HCl solution, and extracted with chloroform and methylene chloride; the combined extracts were washed with water, 10% sodium carbonate solution, and again with water and then dried over magnesium sulfate. The solvents and the unreacted arene were removed, and the residue was crystallized from the proper solvent to afford the target products **5a–h**. (cf. Table I).

Spiro[indoline-3,1'-isothiochroman]-2,4'-dione (**5a**). A sample of **2a** (0.22 g, 0.001 mole) was dissolved in 25 ml of dry benzene, to this solution a 5.87 g (0.044 mole) of $AlCl_3$ was added. The reaction mixture was treated according to the general procedure to afford 0.196 g (70% yield) of **5a**. The product was crystallized from ethanol; m.p. 170–172°C, IR (KBr): 3170 (NH), 3050 (CH arom), 2980 (CH aliph), 1720 (C=O) and 730 cm^{-1} (C—S); ^{1}H-NMR (DMSO-d_6): δ 3.36 (2 H, s), 7.00–7.90 (8 H, complex), 9.60 (1 H, s).

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