

# Reactions of 2,2-Dialkyl-3-thioxochroman-4-one *S*-(1-Adamantylimides) with Some Nitrilimines

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(*E*)-3-Thioxospiro[chroman-2,1'-cyclohexane]-4-one *S*-(1-adamantylimide) (**1**) reacted with numerous nitrilimines (generated *in situ via* triethylamine dehydrohalogenation of the corresponding hydrazoneyl chlorides **2a–i**) in refluxing dry toluene to afford 3'',5''-disubstituted-3''*H*,4'*H*-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazole]-4'-ones **3a–i**. Similarly, reaction of 2,2-dimethyl-3-thioxochroman-4-one *S*-(1-adamantylimide) (**4**) with nitrilimines in refluxing dry toluene afforded the corresponding 3',5'-disubstituted-3,3-dimethyl-3'*H*,4*H*-spiro[chromene-3,2'-[1,3,4]thiadiazole]-ones **5a–i**.

**Key words:** Thioxo *S*-Imides, Nitrilimines, 1,3,4-Thiadiazole

## Introduction

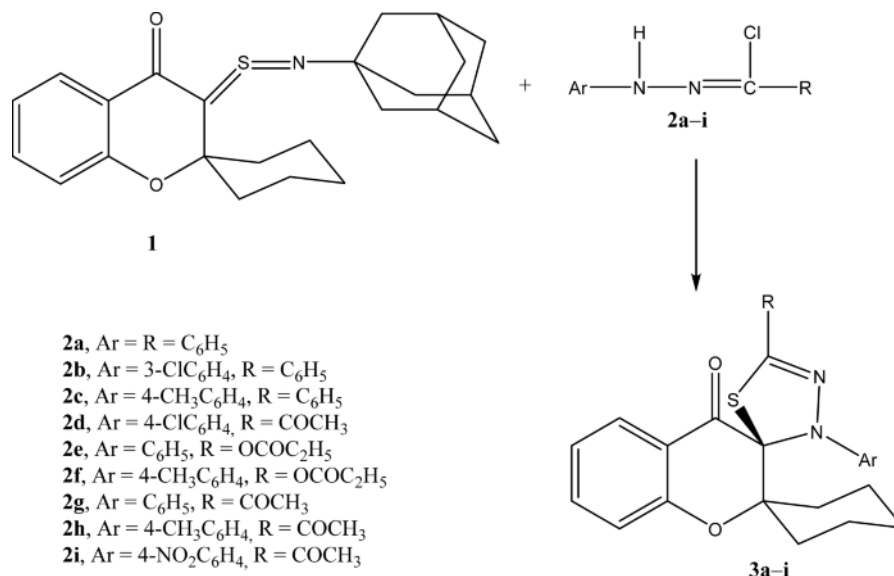
Thiocumulenes behave as 1,3-dipoles or dienophiles in cycloaddition reactions with a large variety of multiple bond-containing compounds. According to the proposal of Inagaki and Okazaki [1, 2], thioxo *S*-imides are 1,3-dipoles. However, the fluorene-thioxo *S*-tosylimide reacts both as a 1,3-dipole and as a dienophile [3, 4]. 2, 2-Dialkyl-3-thioxochroman-4-one *S*-(1-adamantylimides) react as 1,3-dipoles with Lawesson's reagent (LR) [5]. Nitrilimines (generated *in situ* from hydrazoneyl chlorides) represent an important class of highly reactive 1,3-dipoles which have been used intensively for cycloaddition reactions with numerous dienophiles [6]. Many publications have reported that different 1,3,4-thiadiazole derivatives exhibit antimycobacterial [7], antibacterial [8], anticonvulsant [9, 10], and leishmanicidal [11] activities. Moreover, chromanones constitute an important class of naturally occurring substances [12–14] and have drawn the attention of many researchers due to their well known properties as agents against the antihuman immunodeficiency virus (HIV-1) that causes the ac-

quired immune deficiency syndrome (AIDS) [15–17].

In the present work, we wished to explore the behavior of 2,2-dialkyl-3-thioxochroman-4-one *S*-(1-adamantylimides) toward cycloaddition reactions with some nitrilimines.

## Results and Discussion

The reaction of (*E*)-3-thioxospiro[chroman-2,1'-cyclohexane]-4-one *S*-(1-adamantylimide) (**1**) with numerous nitrilimines (generated *in situ via* triethylamine dehydrohalogenation of the corresponding hydrazoneyl chlorides **2a–i**) in refluxing dry toluene afforded all only one identified product. The structures of the isolated products were established to be that of 3'',5''-disubstituted-3''*H*,4'*H*-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazole]-4'-ones **3a–i** based on spectroscopic (IR, <sup>1</sup>H, <sup>13</sup>C NMR, MS) and elemental analyses data (Scheme 1). The formation of 1,3,4-thiadiazole derivatives can be explained by the effect of hydrogen chloride (HCl) on the thioxo *S*-imide derivatives to generate the superdipolarophilic  $\alpha$ -oxo-thioiketone intermediates which un-

Scheme 1. Synthesis of compounds **3a–i**.

derwent a cycloaddition with nitrilimines [18, 19] (Scheme 2).

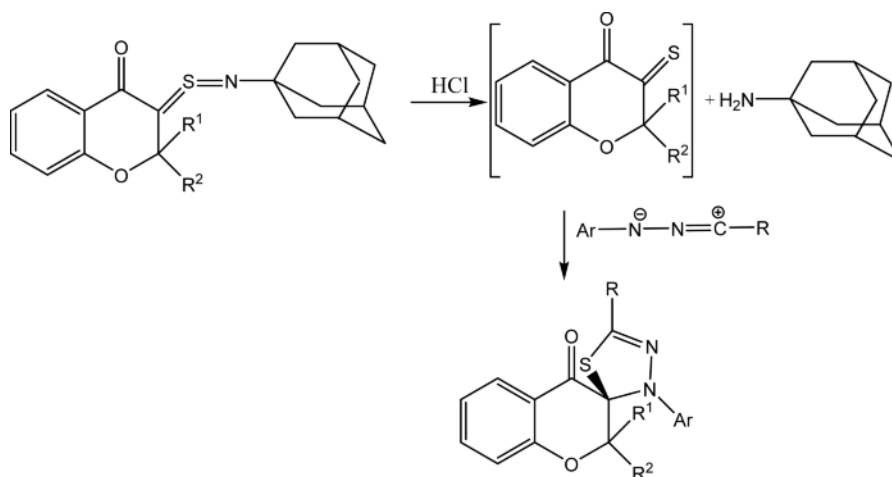
Compounds **3a–c** are identical in all respects (IR, <sup>1</sup>H NMR, MS, and physical data) with those published previously [6]. The IR spectra of compounds **3d–i** show a strong band at  $\nu = 1665–1701\text{ cm}^{-1}$  assignable to a carbonyl stretching vibration band. The <sup>1</sup>H NMR spectra of **3d, g–i** revealed the presence of cyclohexyl protons through multiplet signals at  $\delta = 1.00–2.65\text{ ppm}$ , and  $\delta$  (CH<sub>3</sub>CO) in the region of cyclohexyl protons, beside the expected aromatic signals. The <sup>1</sup>H NMR spectra of **3e, f** showed CH<sub>3</sub>CH<sub>2</sub> as quartet signals at  $\delta = 4.29, 4.30\text{ ppm}$  ( $J = 7.0\text{ Hz}$ ) beside CH<sub>3</sub>CH<sub>2</sub> cyclohexyl and aromatic protons. The mass spectra (EI) of **3d–i** exhibited the molecular ions as base peaks.

Similarly, the reactions of 2,2-dimethyl-3-thioxochroman-4-one *S*-(1-adamantylimide) (**4**) with nitrilimines in refluxing dry toluene afforded the corresponding 3',5'-disubstituted-3,3-dimethyl-3'*H*,4*H*-spiro[chromene-3,2'-[1,3,4]thiadiazole]-ones **5a–i** (Scheme 3). Compounds **5a–c** are identical in all respects (IR, <sup>1</sup>H NMR, MS, and physical data) with those published previously [6]. The IR spectra of compounds **5d–i** show a strong band at  $\nu = 1665–1700\text{ cm}^{-1}$  assignable to a carbonyl stretching vibration band. The <sup>1</sup>H NMR spectra of compounds **5d, g–i** exhibit CH<sub>3</sub> resonances as

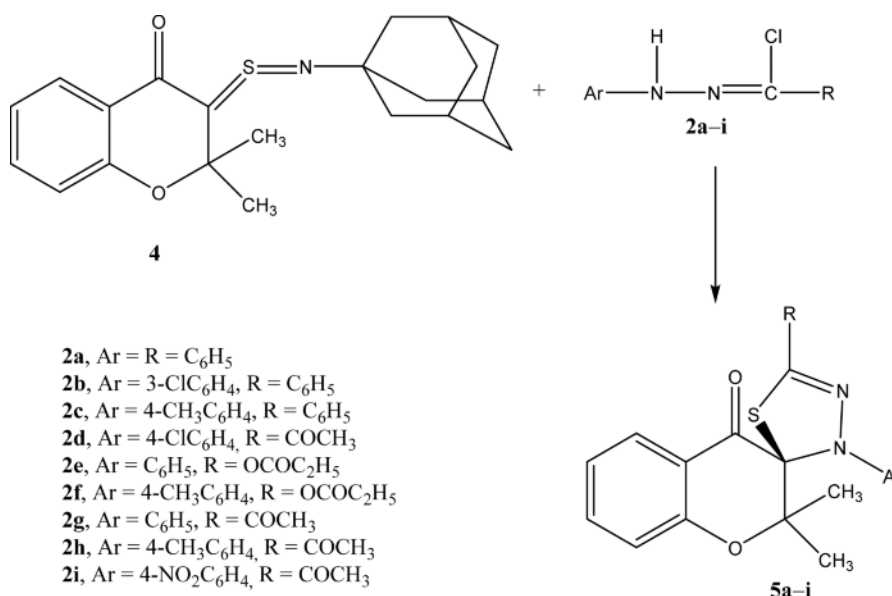
singlet signals at  $\delta = 1.77–1.80\text{ ppm}$ , and CH<sub>3</sub>CO signals at  $\delta = 2.61\text{ ppm}$  beside the expected aromatic protons. The <sup>1</sup>H NMR spectra of **3e, f** revealed CH<sub>3</sub>CH<sub>2</sub> protons as quartet signals at  $\delta = 4.30\text{ ppm}$  ( $J = 7.0\text{ Hz}$ ) beside CH<sub>3</sub>CH<sub>2</sub> protons as triplet signals at  $\delta = 1.35\text{ ppm}$  ( $J = 7.0\text{ Hz}$ ), and aromatic protons. The mass spectra (EI) of **5d–i** showed the molecular ions as base peaks.

## Experimental Section

Melting points were determined on samples in open glass capillaries using an Electrothermal IA 9000 series digital melting point apparatus (Electrothermal, Essex, UK) and are uncorrected. Microanalyses were performed on an Elementar-Vario EL instrument, Microanalytical Unit, Central Services Laboratory, National Research Centre, Cairo, Egypt. IR spectra were obtained with a Bruker-Vector 22 spectrometer on KBr wafers (Micro-analytical Center of Cairo University). The NMR spectra were recorded on a Varian Mercury VX-300 NMR spectrometer. <sup>1</sup>H NMR spectra were run at 300 MHz in CDCl<sub>3</sub> as solvent, <sup>13</sup>C NMR spectra were run at 75 MHz in CDCl<sub>3</sub> as solvent (Cairo University, Faculty of Science). Splitting patterns were designed as follows: s singlet; d doublet; t triplet; m multiplet. Mass spectra were recorded on a Shimadzu GCMS-QP 1000EX spectrometer (EI, 70 eV) by the Micro-analytical Center of Cairo University. Compounds **1** [20], **2a–i** [21–25] and **4** [26] were prepared according to the literature.



Scheme 2. Formation of 1,3,4-thiadiazole derivatives.

Scheme 3. Synthesis of compounds **5a–i**.

#### Reaction of thione *S*-imides (**1**) with hydrazonoyl chlorides (**2**)

The appropriate hydrazonoyl chloride **2** (2 mmol) was added to a solution of thione *S*-imide **1** (2 mmol) in dry toluene (20 mL) containing 1 mL of TEA. The reaction mixture was heated under reflux for 10 h. The formed solid was removed by filtration, and the filtrate was evaporated under reduced pressure to dryness. The residue was chromatographed (silica gel Merk 60, particle size 0.06–0.2 mm, as the stationary phase and petroleum ether 40–60-ethyl acetate 10 : 1 as the eluent)

to give the corresponding 1,3,4-thiadiazole products **3a–i**, respectively.

#### *3''*,*5''*-Diphenyl-*3''H*,*4'H*-dispiro[cyclohexane-1,2'-chromene-*3'*,*2''*]-[1,3,4]thiadiazole-*4'*-one (**3a**)

[6]: M. p. 148–150 °C (ref. [6]: 148–151 °C); yield 65%.

#### *3''*-(3-Chlorophenyl)-*5''*-phenyl-*3''H*,*4'H*-dispiro[cyclohexane-1,2'-chromene-*3'*,*2''*]-[1,3,4]thiadiazole-*4'*-one (**3b**)

[6]: M. p. 169–170 °C (ref. [6]: 169–171 °C); yield 67%.

3''-(4-Methylphenyl)-5''-phenyl-3''H,4'H-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazol]-4'-one (**3c**)

[6]: M. p. 80–82 °C (ref. [6]: 79–81 °C); yield 60%.

5''-Acetyl-3''-(4-chlorophenyl)-3''H,4'H-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazol]-4'-one (**3d**)

From **1** and **2d**. Yield 45%; m. p. 220–223 °C. – IR:  $\nu_{\text{CO}} = 1689, 1700 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.00\text{--}2.65$  (13H, m, 10 cyclohexyl H +  $\text{CH}_3\text{CO}$ ), 6.90–7.45 (8H, m, ArH) ppm. – MS:  $m/z(\%) = 440$  (100)  $[\text{M}]^+$ , 349 (20), 320 (35), 120 (95). –  $\text{C}_{23}\text{H}_{21}\text{ClN}_2\text{O}_3\text{S}$  (440.93): calcd. C 62.65, H 4.80, N 6.35, S 7.27; found C 62.55, H 4.75, N 6.15, S 7.06.

5''-Ethoxycarbonyl-3''-phenyl-3''H,4'H-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazol]-4'-one (**3e**)

From **1** and **2e**. Yield 70%; m. p. 140–142 °C. – IR:  $\nu_{\text{CO}} = 1675, 1701 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.16\text{--}2.50$  (13H, m, 10 cyclohexyl H +  $\text{CH}_3\text{CH}_2$ ), 4.29 (2H, q,  $J = 7.0 \text{ Hz}$ ,  $\text{CH}_3\text{CH}_2$ ), 6.85–7.46 (9H, m, ArH) ppm. – MS:  $m/z(\%) = 436$  (100)  $[\text{M}]^+$ , 316 (40)  $(\text{M}-\text{C}_2\text{H}_6)^+$ , 287 (15), 184 (20), 121 (35), 92 (40). –  $\text{C}_{24}\text{H}_{24}\text{N}_2\text{O}_4\text{S}$  (436.51): calcd. C 66.03, H 5.54, N 6.41, S 7.34; found C 62.55, H 4.75, N 6.15, S 7.06.

5''-Ethoxycarbonyl-3''-(4-methylphenyl)-3''H,4'H-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazol]-4'-one (**3f**)

From **1** and **2f**. Yield 45%; m. p. 189–191 °C. – IR:  $\nu_{\text{CO}} = 1675, 1701 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.16\text{--}2.50$  (13H, m, 10 cyclohexyl H +  $\text{CH}_3\text{CH}_2$ ), 2.18 (3H, s,  $\text{CH}_3$ ), 4.30 (2H, q,  $J = 7.0 \text{ Hz}$ ,  $\text{CH}_3\text{CH}_2$ ), 6.87–7.47 (8H, m, ArH) ppm. – MS:  $m/z(\%) = 450$  (100)  $[\text{M}]^+$ , 330 (25), 121 (15), 92 (10). –  $\text{C}_{25}\text{H}_{26}\text{N}_2\text{O}_4\text{S}$  (450.54): calcd. C 66.64, H 5.81, N 6.22, S 7.12; found C 66.48, H 5.67, N 6.15, S 7.06.

5''-Acetyl-3''-phenyl-3''H,4'H-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazol]-4'-one (**3g**)

From **1** and **2g**. Yield 48%; m. p. 224–225 °C. – IR:  $\nu_{\text{CO}} = 1689, 1700 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.16\text{--}2.53$  (13H, m, 10 cyclohexyl H +  $\text{CH}_3\text{CO}$ ), 6.81–7.44 (9H, m, ArH) ppm. – MS:  $m/z(\%) = 406$  (100)  $[\text{M}]^+$ , 286 (25), 184 (35), 121 (10). –  $\text{C}_{23}\text{H}_{22}\text{N}_2\text{O}_3\text{S}$  (406.49): calcd. C 67.95, H 5.45, N 6.89, S, 7.89; found C 67.77, H 5.29, N 6.70, S 7.65.

5''-Acetyl-3''-(4-methylphenyl)-3''H,4'H-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazol]-4'-one (**3h**)

From **1** and **2h**. Yield 87%; m. p. 254–255 °C. – IR:  $\nu_{\text{CO}} = 1693, 1700 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.16\text{--}2.53$  (16H, m, 10 cyclohexyl H +  $\text{CH}_3\text{CO}$  +  $\text{CH}_3$ ), 6.79–7.24 (8H, m, ArH) ppm. – MS:  $m/z(\%) = 420$  (100)  $[\text{M}]^+$ , 300 (35), 198

(15), 121 (25). –  $\text{C}_{24}\text{H}_{24}\text{N}_2\text{O}_3\text{S}$  (420.52): calcd. C 68.54, H 5.75, N 6.66, S 7.62; found C 68.37, H 5.59, N 6.39, S 7.38.

5''-Acetyl-3''-(4-nitrophenyl)-3''H,4'H-dispiro[cyclohexane-1,2'-chromene-3',2''-[1,3,4]thiadiazol]-4'-one (**3i**)

From **1** and **2i**. Yield 59%; m. p. 159–161 °C. – IR:  $\nu_{\text{CO}} = 1665, 1700 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.00\text{--}2.29$  (10H, m, 10 cyclohexyl H), 2.52 (3H, s,  $\text{CH}_3\text{CO}$ ), 6.98–8.09 (8H, m, ArH) ppm. –  $^{13}\text{C NMR}$ :  $\delta = 20.54, 21.14, 24.95, 25.73, 30.08$  (cyclohexyl-C), 30.98 ( $\text{CH}_3$ ), 85.45 (cyclohexyl C-1), 96.24 (spiro C-3'), 118.89, 120.04, 121.98, 122.75, 124.38, 128.37, 137.91, 143.88, 144.87, 148.17, 157.49 (arom. C), 184.11 ( $\text{CH}_3\text{C}=\text{O}$ ), 190.18 (C-4') ppm. – MS:  $m/z(\%) = 451$  (100)  $[\text{M}]^+$ , 331 (30), 229 (10), 120 (57), 92 (57). –  $\text{C}_{23}\text{H}_{21}\text{N}_3\text{O}_5\text{S}$  (451.48): calcd. C 61.18, H 4.69, N 9.31, S 7.10; found C 60.97, H 4.49, N 9.09, S 6.89.

2,2-Dimethyl-3',5'-diphenyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazole]-4-one (**5a**)

[6]: Pale-yellow oil; yield 70%.

3'-(3-Chlorophenyl)-2,2-dimethyl-5'-phenyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazole]-4-one (**5b**)

[6]: Pale-yellow oil; yield 56%.

2,2-Dimethyl-3'-(4-methylphenyl)-5'-phenyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazole]-4-one (**5c**)

[6]: Pale-yellow oil; yield 52%.

5'-Acetyl-3'-(4-chlorophenyl)-2,2-dimethyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazole]-4-one (**5d**)

From **4** and **2d**. Yield 43%; m. p. 201–203 °C. – IR:  $\nu_{\text{CO}} = 1687, 1700 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.78$  (3H, s,  $\text{CH}_3$ ), 1.80 (3H, s,  $\text{CH}_3$ ), 2.61 (3H, s,  $\text{CH}_3\text{CO}$ ), 7.06–7.94 (8H, m, ArH) ppm. – MS:  $m/z(\%) = 400$  (100)  $[\text{M}]^+$ , 385 (30)  $[\text{M}-\text{CH}_3]^+$ , 357 (50)  $[\text{M}-\text{COCH}_3]^+$ , 320 (46). –  $\text{C}_{20}\text{H}_{17}\text{ClN}_2\text{O}_3\text{S}$  (401.06): calcd. C 59.94, H 4.27, N 6.98, S 7.99; found C 59.80, H 4.23, N, 6.71, S 7.82.

5'-Ethoxycarbonyl-3'-phenyl-2,2-dimethyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazol]-4-one (**5e**)

From **4** and **2e**. Yield 65%; m. p. 101–103 °C. – IR:  $\nu_{\text{CO}} = 1671, 1696 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.35$  (3H, t,  $J = 7.0 \text{ Hz}$ ,  $\text{CH}_3\text{CH}_2$ ), 1.78 (3H, s,  $\text{CH}_3$ ), 1.80 (3H, s,  $\text{CH}_3$ ), 4.30 (2H, q,  $J = 7.0 \text{ Hz}$ ,  $\text{CH}_3\text{CH}_2$ ), 7.06–7.94 (9H, m, ArH) ppm. – MS:  $m/z(\%) = 396$  (100)  $[\text{M}]^+$ , 367 (45)  $[\text{M}-\text{C}_2\text{H}_5]^+$ , 320 (56). –  $\text{C}_{21}\text{H}_{20}\text{N}_2\text{O}_4\text{S}$  (396.44): calcd. C 63.62, H 5.09, N 7.07, S 8.09; found C 63.51, H 4.98, N 6.79, S 7.81.

*5'*-Ethoxycarbonyl-3'-(4-methylphenyl)-2,2-dimethyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazol]-4-one (**5f**)

From **4** and **2f**. Yield 40%; m. p. 154–155 °C. – IR:  $\nu_{\text{CO}} = 1670, 1697 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.36$  (3H, t,  $J = 7.0 \text{ Hz}$ ,  $\text{CH}_3\text{CH}_2$ ), 1.80 (6H, s, 2  $\text{CH}_3$ ), 2.20 (3H, s,  $\text{CH}_3$ ), 4.30 (2H, q,  $J = 7.0 \text{ Hz}$ ,  $\text{CH}_3\text{CH}_2$ ), 7.06–7.94 (8H, m, ArH) ppm. – MS:  $m/z(\%) = 410$  (100)  $[\text{M}]^+$ , 380 (45)  $[\text{M}-\text{C}_2\text{H}_6]^+$ , 336 (56), 305 (30). –  $\text{C}_{22}\text{H}_{22}\text{N}_2\text{O}_4\text{S}$  (410.53): calcd. C 64.36, H 5.41, N 6.82, S 7.80; found C 64.18, H 4.28, N 6.69, S 7.61.

*5'*-Acetyl-3'-phenyl-2,2-dimethyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazol]-4-one (**5g**)

From **4** and **2g**. Yield 43%; m. p. 184–186 °C. – IR:  $\nu_{\text{CO}} = 1681, 1701 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.77$  (3H, s,  $\text{CH}_3$ ), 1.79 (3H, s,  $\text{CH}_3$ ), 2.51 (3H, s,  $\text{CH}_3\text{CO}$ ), 7.06–7.94 (9H, m, ArH) ppm. – MS:  $m/z(\%) = 366$  (100)  $[\text{M}]^+$ , 351 (45)  $[\text{M}-\text{CH}_3]^+$ , 336 (56), 293 (30), 269 (20). –  $\text{C}_{20}\text{H}_{18}\text{N}_2\text{O}_3\text{S}$  (366.41): calcd. C 65.55, H 4.95, N 7.64, S 8.75; found C 65.38, H 4.84, N 7.46, S 8.57.

*5'*-Acetyl-3'-(4-methylphenyl)-2,2-dimethyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazol]-4-one (**5h**)

From **4** and **2h**. Yield 80%; m. p. 190–191 °C. – IR:  $\nu_{\text{CO}} = 1665, 1692 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.78$  (3H, s,  $\text{CH}_3$ ), 1.79 (3H, s,  $\text{CH}_3$ ), 2.22 (3H, s,  $\text{CH}_3$ ), 2.51 (3H, s,  $\text{CH}_3\text{CO}$ ), 6.96–7.84 (8H, m, ArH) ppm. – MS:  $m/z(\%) = 380$  (100)  $[\text{M}]^+$ , 337 (43), 117 (10). –  $\text{C}_{21}\text{H}_{20}\text{N}_2\text{O}_3\text{S}$  (380.48): calcd. C 66.28, H 5.31, N 7.36, S 8.43; found C 66.01, H 5.16, N 7.19, S 8.26.

*5'*-Acetyl-3'-(4-nitrophenyl)-2,2-dimethyl-3'H,4H-spiro[chromene-3,2'-[1,3,4]thiadiazol]-4-one (**5i**)

From **4** and **2i**. Yield 50%; m. p. 195–197 °C. – IR:  $\nu_{\text{CO}} = 1667, 1699 \text{ cm}^{-1}$ . –  $^1\text{H NMR}$ :  $\delta = 1.80$  (3H, s,  $\text{CH}_3$ ), 1.81 (3H, s,  $\text{CH}_3$ ), 2.49 (3H, s,  $\text{CH}_3\text{CO}$ ), 6.86–7.72 (8H, m, ArH) ppm. – MS:  $m/z(\%) = 411$  (100)  $[\text{M}]^+$ , 382 (43), 122 (25). –  $\text{C}_{20}\text{H}_{17}\text{N}_3\text{O}_5\text{S}$  (411.46): calcd. C 58.32, H 4.17, N 10.21, S 7.79; found C 58.09, H 3.97, N 9.99, S 7.61.

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