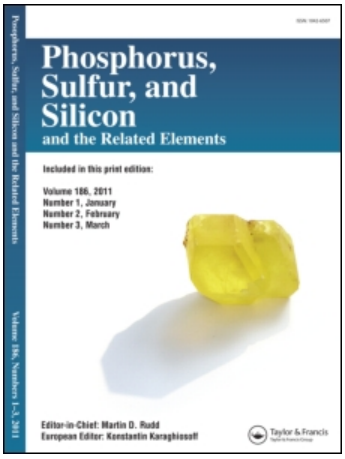

Phosphorus, Sulfur, and Silicon and the Related Elements
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713618290

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To cite this Article Ali, Tarik El-Sayed(2007) 'Synthesis and Fungicidal Activity of Some New 4*H*-Chromen-4-ones Containing Some 1,3-Thiazole, 1,3-Thiazine, 1,2,4-Triazole and 1,2,4-Triazine Moieties', Phosphorus, Sulfur, and Silicon and the Related Elements, 182: 8, 1717 — 1726
To link to this Article: DOI: 10.1080/10426500701313896
URL: http://dx.doi.org/10.1080/10426500701313896

Phosphorus, Sulfur, and Silicon, 182:1717–1726, 2007

ISSN: 1042-6507 print / 1563-5325 online
DOI: 10.1080/10426500701313896

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*The potassium salt of carbamodithioic acid derivative **3** reacted with some electrophiles reagents to afford the 2-thioxo-1,3-thiazoles **4–5** and 2-thioxo-1,3-thiazines **6–7** derivatives. Heterocyclization of compound **3** with amino-mercapto compounds **8–9** and/or hydrazine derivatives **12–13** yielded 3-thioxo-1,2,4-triazoles **10–11** and 3-thioxo-1,2,4-triazines **14–15**, respectively. Structures of the products have been determined by elemental and spectral methods. All the new compounds have been screened for their fungicidal activity.*

Keywords Chromones; fungicidal activity; heterocycles; sulphur-nitrogen

INTRODUCTION

4*H*-Chromen-4-ones and their derivatives have been reported to possess significant activity as protein kinase C inhibitors,[1] antiallergenic,[2] antifungal,[3] and anticancer agents.[4] On the other hand, a large number of 1,3-thiazoles derivatives have been reported to be of biological interest,[5,6] while 1,3-thiazines derivatives have been reported to possess antibacterial, antifungal, and other biological activities.[7] Furthermore, a number of substituted 1,2,4-triazoles and 1,2,4-triazines were found to exhibit appreciable antimicrobial and antifungal activities.[8–11] It was, therefore, thought worthwhile to incorporate the 1,3-thiazole, 1,3-thiazine, 1,2,4-triazole, and/or 1,2,4-triazine moieties into the chromone nucleus. Thus, in the present article, we describe syntheses of some novel 4*H*-chromen-4-ones containing some 1,3-thiazole, 1,3-thiazine, 1,2,4-triazole, and 1,2,4-triazine moieties with the aim to obtain some novel heterocyclic systems with potentially enhanced biological properties.

Received November 20, 2006; accepted February 1, 2007.
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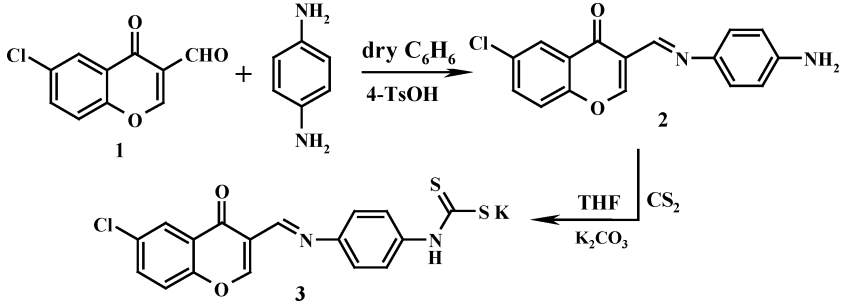
RESULTS AND DISCUSSION

3-{[(4-Aminophenyl)imino]methyl}-6-chloro-4*H*-chromen-4-one (**2**) was prepared by condensation of 6-chloro-4-oxo-4*H*-chromene-3-carboxaldehyde (**1**) with 1,4-phenylenediamine in dry benzene in the presence of 4-toluenesulfonic acid as a catalyst.[12] The potassium salt of carbamodithioic acid derivative **3** was synthesized by reaction of compound **2** with carbon disulfide in boiling tetrahydrofuran and anhydrous potassium carbonate (Scheme 1). Assignments of the



SCHEME 1

structures **2** and **3** were made by elemental and spectral methods. The ^{1}H NMR spectrum of compound **2** (Table I) revealed the appearance of NH_2 protons as a broad signal at δ 12.08–12.15, while compound **3** recorded a singlet at δ11.9 for one NH proton. Moreover, the mass spectrum showed the expected molecular ions peaks at *m/z* 299 (M^+, 1.23%) and 413 (M^+, 42.50%) for **2** and **3**, respectively (Table II).

The potassium salt **3** as starting material was reacted with chloroacetyl chloride and/or phenacyl bromide in boiling dimethylformamide and led to the formation of 2-thioxo-1,3-thiazolidinone **4** and 2-thioxo-1,3-thiazole **5** derivatives, respectively (Scheme 2). Their IR spectra (Table I) showed the disappearance of the NH group, while ^{1}H NMR spectra (Table I) showed broad signals at δ 4.12–4.15 for CH_2 protons in compound **4** and a singlet at δ 8.40 for the C_5–H thiazole in compound **5**. In addition, their mass spectra (Table II) gave in good accordance with the expected molecular formulas.

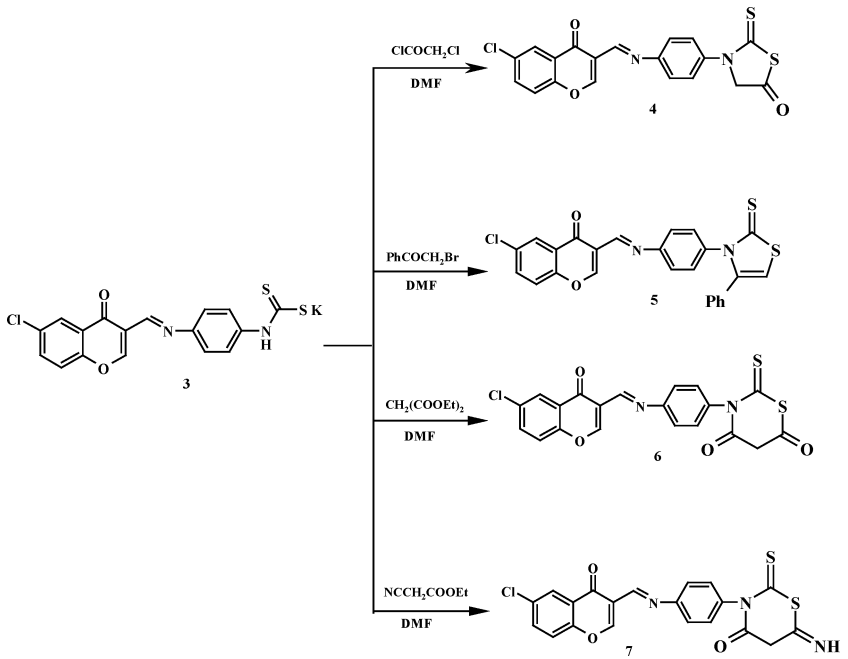
Similarly, 2-thioxo-1,3-thiazine-4,6-dione derivative **6** and 6-imino-2-thioxo-1,3-thiazin-4-one derivative **7** were obtained via cyclization of salt **3** with diethyl malonate and/or ethyl cyanoacetate, respectively, in boiling dimethylformamide containing a catalytic amount of piperidine (Scheme 2). Structures **6–7** were confirmed by their analytical data and ^{1}H NMR (Table I and II). The IR absorption (Table I) was void of the NH group and showed new bands for C=O and/or C=N in thiazinone

TABLE I The Spectral Data of the New Compounds

Compd. no.	IR (cm^{-1}) KBr	^{1}H-NMR (DMSO, δ)
2	3378 (NH_2), 1641 (C=O_{pyrone}), 1604 (C=N)	6.60 (d, 1H, J = 6.7 Hz, H–8), 6.92–7.28 (m, 7H, Ar–H, H–7, H–9 and H–5), 7.76 (br, 1H, H–2), 12.08–12.15 (br, 2H, NH_2)
3	3288 (NH), 1637 (C=O_{pyrone}), 1603 (C=N), 1188 (C=S)	6.40–7.5 (m, 7H, Ar–H, H–8 , H–7 and H–9), 7.75 (s, 1H, H–5), 8.0–8.2 (br, 1H, H–2),12.0–12.2 (ss, 1H, NH)
4	1718 (C=$O_{thiazolidinone}$), 1644 (C=O_{pyrone}), 1604 (C=N), 1202 (C=S)	4.12–4.15 (br, 2H, CH_2), 6.93–7.69 (m, 7H, Ar–H, H–8 , H–7 and H–9), 7.95 (s, 1H, H–5), 8.54 (s, 1H, H–2)
5	1639 (C=O_{pyrone}), 1596 (C=N), 1217 (C=S)	7.03–7.64 (m, 5H, Ar–H and H–8), 7.83–7.91 (m, 2H, H–7 and H–9), 7.95 (s, 1H, H–5), 8.40 (s, 1H, C_5–$H_{thiazole}$), 8.88 (s, 1H, H–2)
6	1719 (br, C=$O_{thiazinedione}$),1625 (C=O_{pyrone}), 1598 (C=N), 1219 (C=S)	4.06 (br, 2H, CH_2), 7.06–7.43 (m, 5H, Ar–H and H–8), 7.76,7.78 (ss, 1H, H–7), 7.86 (s, 1H, H–9), 8.04 (s,1H, H–5), 8.89 (s, 1H, H–2)
7	3271 (NH), 1669 (C=$O_{thiazinone}$), 1615 (C=O_{pyrone}), 1581 (C=N), 1219 (C=S)	3.91 (s, 2H, CH_2), 6.50–7.35 (m, 5H, Ar–H and H–8), 7.38–7.44 (m, 3H, H–7, H–9 and H–5), 7.45 (s, 1H, H–2), 11.60 (br, 1H, NH)
10	3215,3338 (NH, NH), 1652 (C=O_{pyrone}), 1607 (C=N), 1175, 1126 (2 C=S)	6.64–7.35 (m, 4H, Ar–H), 7.49 (d, 1H, J = 8 Hz, H–8), 7.72–7.86 (m, 2H, H–7 and H–9), 8.40 (s, 1H, H–5), 9.09 (s, 1H, H–2), 14.20 (br, 2H, NH,NH)
11	3352 (NH), 1625 (C=O_{pyrone}), 1605 (C=N), 1119 (C=S)	2.27 (s, 3H, CH_3), 6.91–7.43 (m, 4H, Ar–H), 7.60–7.90 (m, 2H, H–8 and H–7), 8.35 (s, 1H, H–9), 9.01 (s, 1H, H– 5), 9.50 (s, 1H, H–2), 10.62 (s, 1H, NH)
14	3422 (br,NH), 1650 (C=O_{pyrone}), 1597 (C=N), 1226 (C=S)	4.20–4.43 (m, 2H, C_5–$H_{triazine}$), 5.49–5.57 (q, 1H, C_6–$H_{triazine}$), 7.13–7.56 (m, 10H, Ar–H and H–8), 7.74–7.86 (br, 2H, H–7 and H–9), 7.92 (br, 1H, H–5), 8.01 (br, 1H, H–2), 8.96 (s, 1H, NH), 10.11 (s, 1H, NH)
15	3391 (NH), 1641 (C=O_{pyrone}), 1602 (C=N), 1209 (C=S)	4.43 (s, 1H, C_5–$H_{triazine}$), 7.16–7.40 (m, 17H, Ar–H, H–8, H–7 and H–9), 7.51 (br, 1H,H–5), 7.77 (br, 1H, H–2), 7.95 (s, 1H, NH)

TABLE II The Physical, Analytical, and Mass Spectral Data of The New Compounds

Compd. no.	m.p. °C (Yield %)	Solvent of cryst.	Formula (M.Wt)	Calculated/Found % C	H	N	MS (m/z, %)
2	203–205 67%	Dioxane	$C_{16}H_{11}ClN_2O_2$ (298.73)	64.33 63.90	3.71 3.60	9.38 8.90	300 (M+1, 1.56), 299 (M^+, 1.23), 199 (1.70), 155 (3.38), 154 (2.59), 136 (5.55), 108 (100), 80 (43.50)
3	238–240 81%	diluted DMF	$C_{17}H_{10}ClKN_2O_2S_2$ (412.96)	49.44 49.31	2.44 2.40	6.78 6.52	416 (M+3, 34.4), 415 (M+2, 82.0), 414 (M+1, 31.2), 413 (M^+, 42.5), 304 (20.4), 145 (30.3), 105 (42.5), 55(100)
4	220–221 59%	Benzene	$C_{19}H_{11}ClN_2O_3S_2$ (414.89)	55.00 54.80	2.67 2.60	6.75 6.50	373 (M–$COCH_2$, 24.80), 298 (25.2), 244 (3.86), 206 (26.42), 154 (100), 152 (75.20), 126 (45.12), 107 (32.93), 65 (83.33)
5	109–111 64%	diluted DMF	$C_{22}H_{15}ClN_2O_2S_2$ (474.99)	63.22 62.78	3.18 3.00	5.90 5.45	475 (M^+, 0.27), 372 (0.41), 327 (0.49), 311 (0.56), 154 (1.70), 105 (20.62), 77 (100)
6	> 300 43%	Acetic acid	$C_{20}H_{11}ClN_2O_4S_2$ (442.90)	54.24 53.85	2.50 2.38	6.33 6.05	443 (M^+, 12.20), 413 (18.29), 381 (13.41), 317 (21.95), 169 (26.83), 129 (17.07), 75 (39.02), 51 (100)
7	159–161 55%	Ethanol	$C_{20}H_{12}ClN_3O_3S_2$ (441.92)	54.36 54.10	2.74 2.65	9.51 9.35	551 (M–CS, 0.30 %), 368 (0.44), 313 (1.23), 233 (0.62), 221 (28.74), 154 (44.01), 94 (100), 67 (53.13)
10	186–188 52%	diluted DMF	$C_{18}H_{11}ClN_4O_2S_2$ (414.89)	52.11 51.82	2.67 2.52	13.50 12.08	415 (M^+, 0.13), 383 (0.15), 296 (0.21), 160 (0.58), 154(0.21), 96 (12.00), 64 (100)
11	154–156 52%	diluted DMF	$C_{20}H_{13}ClN_6O_2S$ (436.88)	54.98 54.70	3.00 2.81	19.24 18.92	438 (M+1, 3.23), 371 (4.11), 154 (30.21), 147 (100), 126 (52.49), 63 (84.16),54 (68.33)
14	260–262 73%	Ethanol	$C_{25}H_{19}ClN_4O_2S$ (474.97)	63.22 62.80	4.03 3.95	11.80 11.73	460 (M–NH, 2.5), 417 (100), 298 (0.6), 188 (10.3), 172 (59.1), 120 (36.6), 65 (15)
15	185–187 39%	Ethanol	$C_{31}H_{21}ClN_4O_2S$ (549.05)	67.81 67.65	3.86 3.66	10.20 9.85	551 (M+2, 0.21), 549 (M^+, 0.19), 523 (0.25), 467 (0.14), 388 (3.25), 297 (69.48), 193 (14.38), 91 (100), 77 (17.21), 65 (16.90)

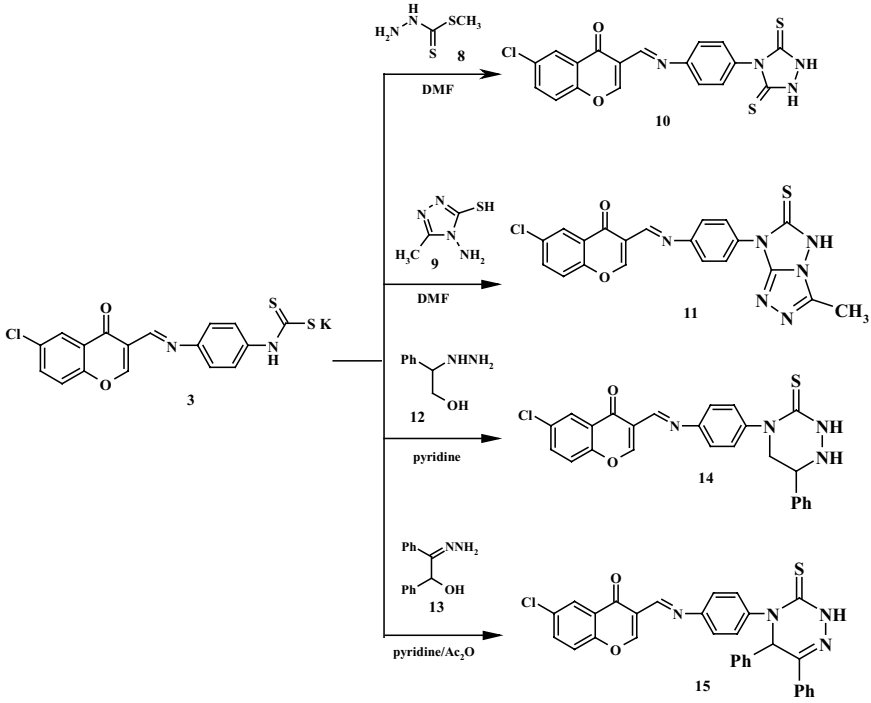


SCHEME 2

moieties. The ^{1}H NMR spectra of compounds **6** and**7** (Table 1) exhibited broad signals at δ 4.06–3.91 for the methylenic group. Moreover, the mass spectra (Table II) displayed the molecular ion peaks at m/z 443 (M^+, 12.20%) and 398 (M–CS, 0.30%) for **6** and **7**, respectively.

Heterocyclization of salt of carbamodithioic acid derivative **3** with some amino-mercapto compounds, namely methyl hydrazinecarbodithioic acid ester (**8**) and/or 4-amino-5-methyl-3-thioxo-1,2,4-triazole (**9**) in boiling dimethylformamide, afforded 3-thioxo-1,2,4-triazole derivatives **10** and **11**, respectively (Scheme 3). The IR spectra (Table I) of **10** and **11** showed absorption bands at 3215–3353 cm^{-1} corresponding to NH groups and bands at 1119–1175 cm^{-1} for C=S groups. In addition, the ^{1}H NMR spectrum of **10** (Table I) showed characteristic broad signal at δ 14.20 for NH,NH protons, while compound **11** showed singlets at δ 2.27 and 10.62 for CH_3 and NH protons, respectively. The mass spectra of **10** and **11** showed molecular ion peaks at m/z 415 (M^+, 0.13%) and 438 (M+1, 3.23%), respectively (Table II).

Cyclocondensation of acid derivative **3** with hydrazine derivatives **12** and **13** in boiling pyridine yielded 3-thioxo-1,2,4-triazine derivatives **14** and **15**, respectively (Scheme 3). Their IR spectra (Table I) showed the



SCHEME 3

appearance of NH groups in triazine moieties at 3391–3422 cm^{-1}. The ^1H NMR spectrum of **14** (Table I) showed singlets at δ 8.96, 10.11 for NH,NH protons, multiplets at δ 4.20–4.30 for the CH_2 and a quartet at δ 5.49–5.57 attributed to the C_6–H triazine. Compound **15** showed two singlets at δ 4.43 and 7.95 attributed to the C_5–H triazine and NH protons, respectively. The structures of **14** and **15** were also confirmed by mass spectra that showed molecular ion peaks at m/z 460 (M–NH, 2.5%) and 549 (M^+, 0.19%), respectively (Table II).

FUNGICIDAL ACTIVITY

The new prepared compounds were screened for their fungicidal activities against three species of fungi, namely *Alternaria alternata, Aspergillus niger*, and *Aspergillus flavips*, using disc diffusion method.[13] The tested compounds were dissolved in dimethylformamide, which was used as a control to get 1 mg/ml solution. The inhibition zones of microbial growth surrounding the filter paper disc (2.5 mm) were measured in millimeters at the end of an incubation period at 30°C for 3

TABLE III Fungicidal Activity Data of The New Compounds 2–15

Compd. no.	*Alternaria alternata*	*Aspergillus niger*	*Aspergillus flavips*
2	++	++	++
3	++	++	+++
4	++	+	++
5	+++	+++	++
6	++	++	++
7	+++	++	+++
10	++	++	+++
11	++	++	++
14	+++	++	+
15	+++	+++	+++
(flucanazole)	+++	+++	+++

Lower activity = + (inhibition zone 1–10 mm); Moderate activity = ++ (inhibition zone 12–25 mm); and High activity = + + + (inhibition zone >25 mm).

days. Activity of each compound was compared with that of flucanazole as the standard. The investigation of fungicidal screening data (Table III) revealed all the tested compounds showed moderate to high inhibition. The most active compound **15**, which exhibited the maximum fungicidal activity against all fungi strains was almost equivalent to that of the standard due to the presence of the 5,6-diphenyl-1,2,4-triazine moiety. Compound **5** showed high activity against *Alternaria alternata* and *Aspergillus niger*, while compound **7** showed the same high activity against *Alternaria alternata* and *Aspergillus flavips*. This may be due to the presence of two sulfur atoms as an endo–exo system (S=C=S) in the thiazole and/or thiazine moieties.

EXPERIMENTAL

Melting points were determined on a digital Stuart SMP-3 apparatus. Infrared spectra were measured on Perkin-Elmer 293 spectrophotometer (cm^{-1}), using KBr disks. ^{1}HNMR spectra were measured on Gemini-200 spectrometer (200 MHz), using DMSO-d_6 as a solvent and TMS (δ) as the internal standard. The mass spectra were measured on a HP–MS 5988 mass spectrometer *via* a direct inlet operating at 70 eV. Elemental microanalyses were performed at the microanalysis center at Cairo University. 6-Chloro-4-oxo-4*H*-chromene-3-carboxaldehyde (**1**),[14] methyl hydrazine-carbodithioic acid ester (**8**),[15] 4-amino-5-methyl-3-thioxo-1,2,4-triazole (**9**),[15] and hydrazine derivatives **12–13**[16] were prepared by published methods in literature. The physical and spectral data of the new compounds are listed in Table I and II.

3-{[(4-Aminophenyl)imino]methyl}-6-chloro-4H-chromen-4-one (2)

A mixture of 6-chloro-4-oxo-4*H*-chromene-3-carboxaldehyde (**1**) (1.04 g, 5 mmol) and 1,4-phenylenediamine (0.54 g, 5 mmol) in dry benzene (50 ml) in the presence of 4-toluensulfonic acid (0.1 g) was refluxed for 30 min. The solid obtained was filtered off and crystallized from the proper solvent to give **2**.

Potassium salt of (4-{[(6-chloro-4-oxo-4H-chromen-3-yl) methylene]amino}- phenyl) carbamodithioic acid (3)

To a solution of compound **2** (1.49 g, 5 mmol) in tetrahydrofuran (50 ml) was added carbon disulfide (0.38 g, 5 mmol) and anhydrous potassium carbonate (3 g). The mixture was refluxed for 10 h and filtered while hot. The filtrate gave a precipitate after cooling that was crystallized from the proper solvent to give **3**.

3-(4-{[(6-Chloro-4-oxo-4H-chromen-3-yl)methylene] amino}phenyl)-2-thioxo- 1,3-thiazolidin-5-one (4)

A mixture of compound **3** (2.06 g, 5 mmol) and chloroacetyl chloride (0.56 g, 5 mmole) in dimethylformamide (50 ml) was refluxed for 4 h. The solution was cooled and poured onto ice-water. The solid obtained was filtered off and crystallized from the proper solvent to give **4.**

3-(4-{[(6-Chloro-4-oxo-4H-chromen-3-yl)methylene] amino}phenyl)-4-phenyl-2-thioxo-1,3-thiazole (5)

A mixture of compound **3** (2.06 g, 5 mmol) and phenacyl bromide (0.99 g, 5 mmol) in dimethylformamide (50 ml) was refluxed for 4 h. The solution was cooled and poured onto ice-water. The solid obtained was filtered off and crystallized from the proper solvent to give **5.**

3-(4-{[(6-Chloro-4-oxo-4H-chromen-3-yl)methylene] amino}phenyl)-2-thioxo- 5-hydro-1,3-thiazine-4,6-dione (6)

A mixture of compound **3** (2.06 g, 5 mmol) and diethyl malonate (0.8 g, 5 mmol) in dimethylformamide (50 ml) in the presence of piperidine (0.2 ml) was refluxed for 10 h. The solution was cooled and poured onto ice-water. The solid obtained was filtered off and crystallized from the proper solvent to give **6.**

3-(4-{[(6-Chloro-4-oxo-4H-chromen-3-yl)methylene] amino}phenyl)-6-imino-2-thioxo-5-hydro-1,3-thiazin-4-one (7)

A mixture of compound **3** (2.06 g, 5 mmol) and ethyl cyanoacetate (0.56 g, 5 mmol) in dimethylformamide (50 ml) in the presence of piperidine (0.2 ml) was refluxed for 10 h. The solution was cooled and poured onto ice-water. The solid obtained was filtered off and crystallized from the proper solvent to give **7.**

4-(4-{[(6-Chloro-4-oxo-4H-chromen-3-yl)methylene] amino}phenyl)-3,5-dithioxo-1,2,4-triazolidine (10)

A mixture of compound **3** (2.06 g, 5 mmol) and methyl hydrazinecarbodithioic acid ester (**8**) (0.61 g, 5 mmol) in dimethylformamide (50 ml) was refluxed for 6 h. The solution was cooled and poured onto ice-water. The solid obtained was filtered off and crystallized from the proper solvent to give **10.**

7-(4-{[(6-Chloro-4-oxo-4H-chromen-3-yl)methylene] amino}phenyl)-3-methyl- 6-thioxo-5,6-dihydro-7H-[1,2,4] triazolo[4,3-b][1,2,4]triazole (11)

A mixture of compound **3** (2.06 g, 5 mmol) and 4-amino-5-methyl-3-thioxo-1,2,4-triazole (**9**) (0.65 g, 5 mmol) in dimethylformamide (50 ml) was refluxed for 6 h. The solution was cooled and poured onto ice-water. The solid obtained was filtered off and crystallized from the proper solvent to give **11**.

4-(4-{[(6-Chloro-4-oxo-4H-chromen-3-yl)methylene] amino}phenyl)-5,6-dihydro-5-phenyl-3-thioxo-1,2,4-triazine (14)

A mixture of compound **3** (2.06 g, 5 mmol) and hydrazine derivative **12** (1.07 g, 5 mmol) in dry pyridine (50 ml) was refluxed for 6 h. The solution was cooled and poured onto ice-water. The solid obtained was filtered off and crystallized from the proper solvent to give **14**.

4-(4-{[(6-Chloro-4-oxo-4H-chromen-3-yl)methylene] amino}phenyl)-2,5-dihydro-5,6-diphenyl-3-thioxo-1,2,4-triazine (15)

A mixture of compound **3** (2.06 g, 5 mmol) and hydrazine derivative **13** (1.13 g, 5 mmol) in dry pyridine (50 ml) in the few drops of acetic

anhydride was refluxed for 10 h. The solution was cooled and poured onto ice-HCl. The solid obtained was filtered off and crystallized from the proper solvent to give **15**.

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