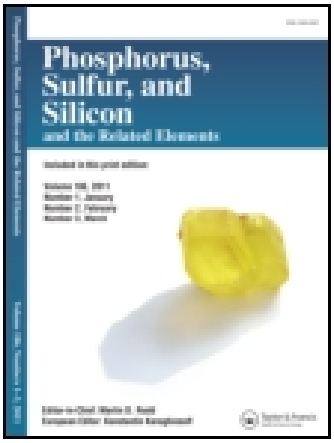

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Some Cyclization Reactions with 2-Ethoxycarbonylmethylidene-4,5-Dihydro-4-Thiazolinone

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SOME CYCLIZATION REACTIONS WITH 2-ETHOXYCARBONYLMETHYLIDENE-4,5-DIHYDRO-4-THIAZOLINONE

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2-Ethoxycarbonylmethylidine-4,5-dihydro-4-thiazolinone (1) was condensed with bis aromatic aldehydes such as terephthalaldehyde or 4,4′-bisformyl-diphenylether (2a,b) (2:1 molar ratio) and furnished bis-4-thiaozlidinones (3a,b). The reaction of (3a,b) with malononitrile and aromatic aldehydes (1:2:2 molar ratio) gave bis thiazolopyridines (4a–d). Bis-(thiazolopyridine) derivative (6) was obtained by reaction of 4-thiaozlinone (5c) with bis aldehyde (2b) in refluxing ethanol containing piperidine. Cyclization of 4-thiazolinones (5a,b) with different α-cyanocinnamonitriles gave thiazolo[3,2-a]pyridines (7a–d). Compound 9 was produced via the reaction of 8 with thioglycolic acid, which reacted with p-chlorobenzaldehyde to produce 10. Compound 10 was condensed with hydrazine hydrate and afforded 11. Compounds 12 and 16a,b were produced by the reaction of 9 with isatin and α-ethoxycarbonylcinnamonitriles, respectively.

Keywords Bisthiazolidinone; bisthiazolo[3,2-a]pyridine derivatives; 4-thiazolidinone

INTRODUCTION

Some thiazolo[3,2-a]pyridines have been reported to possess various biological activities including antibacterial, anticancer, antihypertensive, antidilator, and funigicidal activities.[1–8] A considerable number of bis heterocyclic compounds exhibited various biological activities including antibacterial, fungicidal, tuberculostatic, and plant growth regulative properties.[9–12] It has also been reported that bis heterocyclic compounds display much better antibacterial activity than heterocyclic compounds.[13] We report in this article the synthesis of some novel 4-thiazolidinone and bis thiazolo[3,2-a] pyridine derivatives.[14–20]

RESULTS AND DISCUSSION

It has been found that 4-thiazolinone (**1**) reacts with bis aromatic aldehydes (**2a**,**b**) in refluxing ethanol to yield bis-4-thiazolinones (**3a**,**b**). The structure of compounds **3a**,**b** was confirmed by spectroscopic data.

The ^{1}H NMR spectrum of **3a** showed signals at δ 1.11 (t, 6H, $2CH_3$), 3.77 (q, 4H, $2CH_2$), 5.23 (s, 2H, 2CH-methylidene), 6.90–7.49 (m, 6H, Ar—H + 2 benzylidene-H)

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and 10.32, 10.40 (2s, 2H, 2NH). Ternary condensation of bis 4-thiazolinone (**3a**,**b**), aromatic aldehydes, and malononitrile (1:2:2 molar ratio) afforded bisthiazolo pyridines (**4a–d**). The structure of compounds **4a–d** was in agreement with their spectral data. The IR spectrum of **4a** showed the presence of absorption bands for amino at 3406, 3350 cm^{-1} and cyano at 2190 cm^{-1} functional groups, and its mass spectrum showed a molecular ion peak at *m/z* 849 (5.13%) and a base peak at *m/z* 55. 2-Ethoxycarbonylmethylidine-4,5-dihydro-5-arylmethylidene-4-thiazolinones (**5a–c**) were produced via the condensation of **1** with aromatic aldehydes in refluxing ethanol containing catalytic amount of piperidine. On the basis of elemental analysis and spectral data, the structure of thiazolidinone derivatives **5a–c** was confirmed. Mass spectrum of compound **5c** ($C_{16}H_{18}N_2O_3S$) exhibited a molecular ion peak at *m/z* 318 (6.67%) with a base peak at *m/z* 177 (14.22%). Cyclization of **5a**,**b** with different α-cyanocinnamonitriles gave thiazolopyridines **7a–d**. 4,4′-Bis-(2,3,7-trihydro-3-oxo-5-amino-6-cyano-8-ethoxycarbonyl-7-aryl-1,3-thiazolo[3,2-a]pyridine-2-ylidenemethyl) diphenyl ether (**6**) was produced by the reaction of **5c**, malononitrile, and bis aldehyde (**2b**) (2:2:1 molar ratio) in refluxing ethanol in the presence of a catalytic amount of piperidine (Scheme 1).

The IR spectrum of **6** revealed absorption bands for cyano and amino groups at 2188, 3468, and 3410 cm^{-1}. Its ^1H NMR spectrum in (DMSO-d_6) displayed a signals at δ 1.04, 1.22 (2t, 6H, $2CH_3$-ester), 3.03, 3.11 (2s, 12H, $2N(CH_3)_2$), 4.05, 4.99 (2q, 4H, $2CH_2$-ester), 5.57, 5.62 (2s, 2H, 2 pyridine-H), 6.87, 7.87 (m, 20H, Ar—H+ benzylidene-H + $2NH_2$). The novel 4-thiazolinone (**9**) can not be obtained from the reaction of **1** with 4-chloroaniline under various conditions, thus compound **9** was produced from the reaction of thioglycolic acid with *N*-(4-chlorophenyl)-2-cyano-acetamide **8** in acetic acid under reflux conditions. Electrophiles such as *p*-chlorobenzaldehyde and isatin were condensed with **9** and gave 4-thiazolidinone derivatives (**10**, **12**). The mass spectrum of compound **10** ($C_{25}H_{15}N_2Cl_3O_2S$) showed a molecular ion peak at m/z 513.5 (7.7%), and a base peak was found in the spectrum at m/z 63. Also, the mass spectrum of compound **12**($C_{19}H_{12}N_3ClO_3S$) displayed a molecular ion peak at m/z 397 (4.18%) and a base peak at m/z 127.

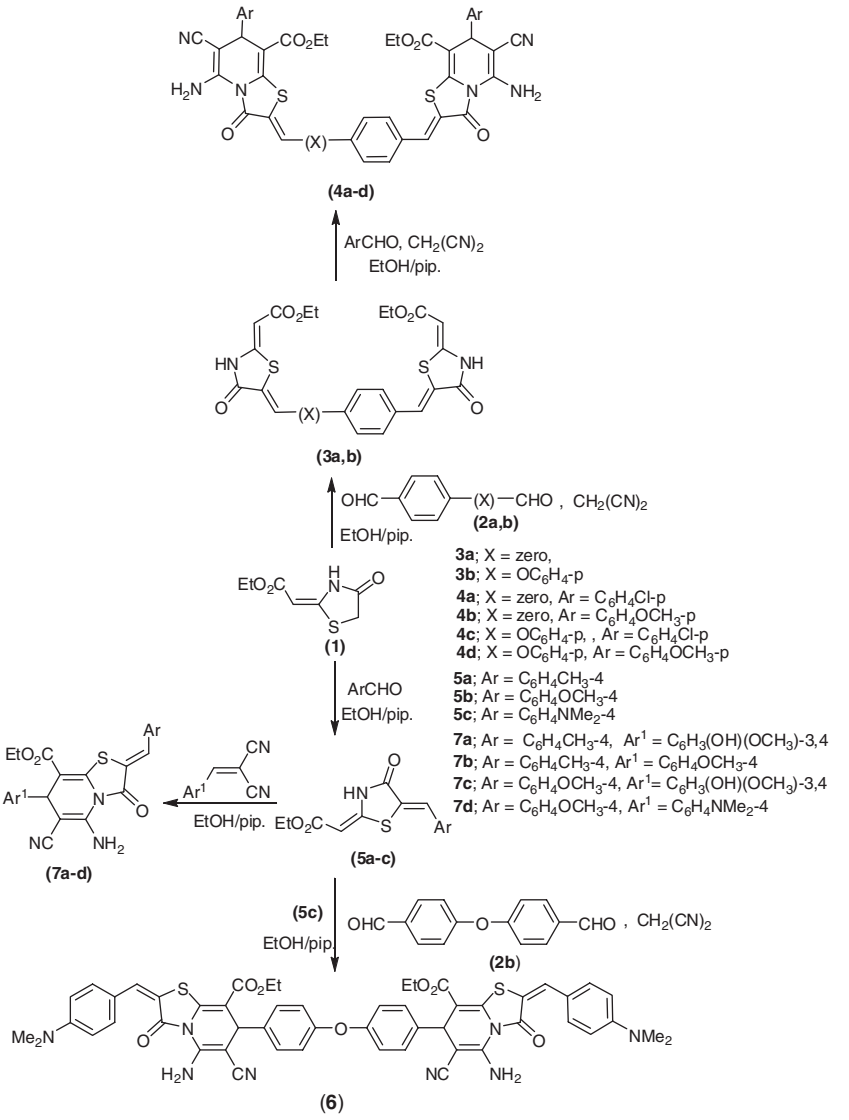
The reaction of thiazolidinone derivative **10** with hydrazine hydrate in refluxing ethanol afforded pyrazolo[3,4-*d*]thiazole derivative **11** (Scheme 2).

Elemental analysis and spectral data were in complete accordance with pyrazolo[3,4-*d*]thiazole derivative (**11**). Mass spectrum of compound **11**($C_{25}H_{16}N_4Cl_3OS$) exhibited a molecular ion peak at m/z 525 (0.7%) and a base peak was found at m/z 127.

Finally, treatment of thiazolidinone derivative **9** with α-ethoxycarbonylcinnamonitriles in dioxane in the presence of a catalytic amount of piperidine gave the corresponding thiazolo-[3,2-*a*]pyridine derivatives **16a**,**b** (Scheme 3). The other possible structures **13a**,**b**, **14a**,**b**, and **15a**,**b** were excluded on the basis of elemental analyses and spectral data. The IR spectrum of compound **16b** showed absorption bands at 3375, 3316, 2184, 1714, and 1650 cm^{-1} due to imino, hydroxy, cyano, and carbonyl functional groups, respectively. Its ^1H NMR spectrum in (DMSO-d_6) displayed signals at δ 5.05 (s, 1H, pyridine-H), 7.26–7.71 (m, 13H, Ar—H + methylidene-H), 9.49 (s, 1H, NH), and 12.10 (br, 1H, OH).

EXPERIMENTAL

Melting points were recorded on a Fisher-Johns melting points apparatus and are uncorrected. IR spectra were recorded on a Shimadzu 470 spectrometer using KBr pellets. ^1H NMR spectra were recorded on a Varian Gemini spectrometer (200 MHz) using TMS as internal standard and mass spectra on a Jeol-JMS-600 mass spectrometer. Elemental

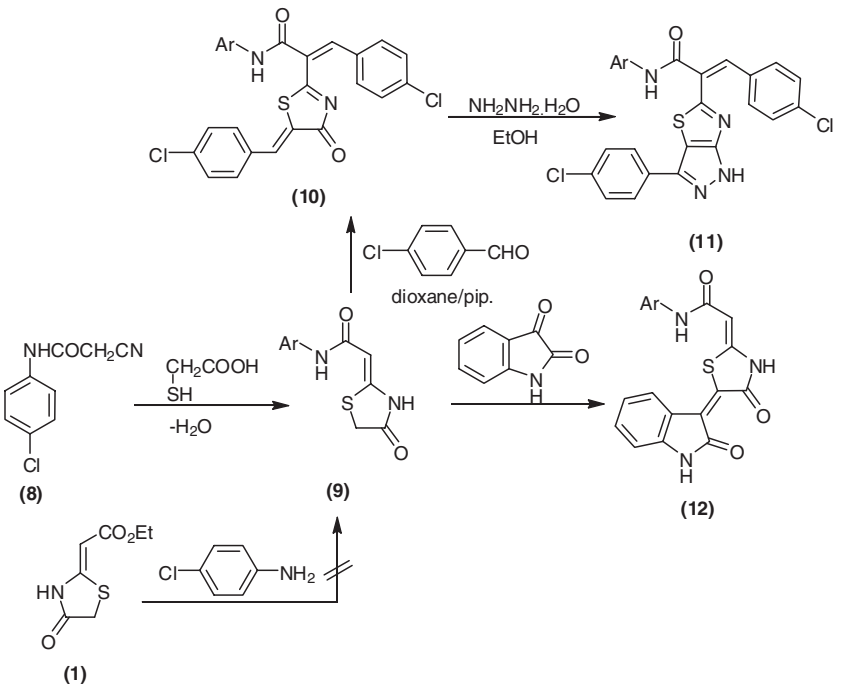


Scheme 1

analyses were obtained from the Microanalytical Data Unit at Cairo University. The physical and spectral data for the synthesized compounds are given in Tables I and II, respectively. Compound **1** was prepared according to the reported method.[21]

1,4-Bis(2-ethoxycarbonylmethylidene-4-oxo-4,5-dihydro-5-yl)benzene (3a), 4,4′-Bis(2-ethoxycarbonylmethylidene-4-oxo-4,5-dihydro-5-yl)diphenyl Ether (3a,b)

To a solution of **1** (0.02 mol) in absolute ethanol (40 mL) containing a catalytic amount of piperidine (0.5 mL), either a bisaldehyde **2a** (0.01 mol) or **2b** (0.01 mol) was



Scheme 2

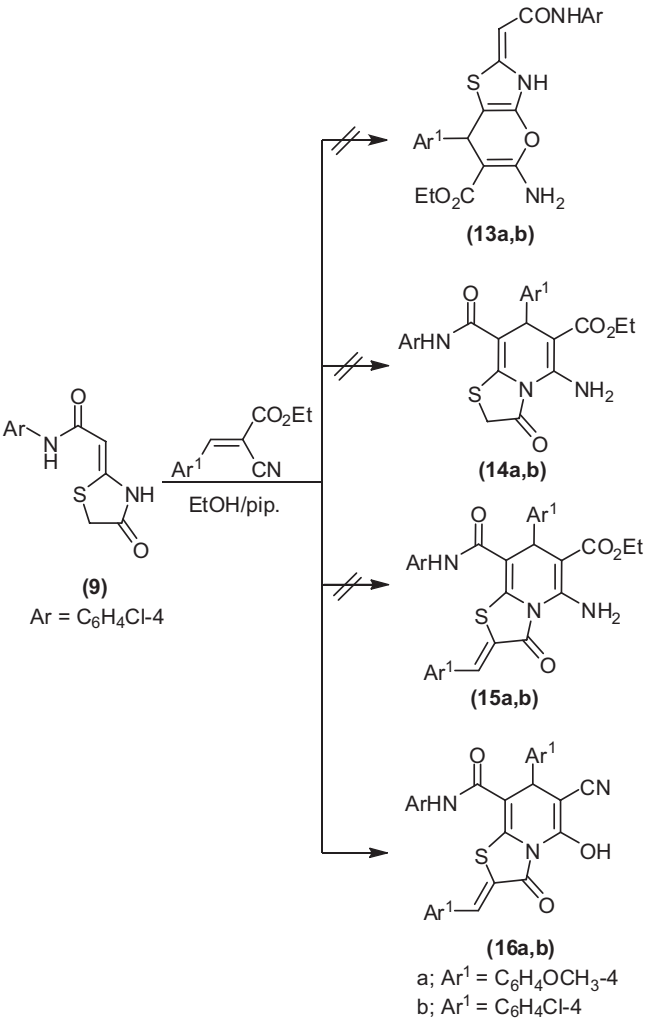
added. The reaction mixture was heated under reflux for 6 h. The solid product formed was collected by filtration.

1,4-Bis(2,3,7-trihydro-3-oxo-5-amino-6-cyano-7-aryl-8-ethoxycarbonyl-1,3-thiazolo[3,2-a]pyridine-2-ylidenemethyl)-benzene (4a,b), 4,4′-Bis(2,3,7-trihydro-3-oxo-5-amino-6-cyano-7-aryl-8-ethoxycarbonyl-1,3-thiazolo [3,2-*a*]pyridine-2-ylidene-methyl)diphenyl Ether (4c,d)

To a solution of either **3a** (0.01 mol) or **3b** (0.01 mol) in absolute ethanol (40 mL) containing a catalytic amount of piperidine (0.5 mL), malononitrile (0.02 mol) was added. The reaction mixture was refluxed for 6 h. The solid product formed was collected by filtration.

4,4′-Bis(2,3,7-trihydro-3-oxo-5-amino-6-cyano-7-aryl-8-ethoxycarbonyl-1,3-thiazolo[3,2-*a*]pyridine-2-ylidene-methyl)-diphenyl Ether (6)

To a solution of **5c** (0.02 mol) in absolute ethanol (40 mL) containing a catalytic amount of piperidine (0.5 mL), malononitrile (0.02 mol) and **2b** (0.01 mol) were added. The reaction mixture was heated under reflux for 6h. The solid product formed was collected by filtration.



Scheme 3

2-Ethoxycarbonylmethylidene-4-oxo-4,5-dihydro-5-arylmethylidene-1,3-thiazole (5a–c)

Equimolar amounts of 4-thiazolinone **1** (0.01 mol) and aromatic aldehydes (0.01 mol) in absolute ethanol (40 mL) containing a catalytic amount of piperidine (0.5 mL) were mixed together. The reaction mixture was heated under reflux for 3 h. The solid product formed was collected by filtration.

2-Arylmethylidene-2–2,3,7-trihydro-3-oxo-5-amino-6-cyano-7-aryl-8-ethoxycarbonyl-1,3-thiazolo[3,2-*a*]pyridine (7a–d)

Equimolar amounts of 4-thiazolinones **5a**,**b** (0.01 mol) and α-cyanocinnamonitrile derivatives (0.01 mol) in absolute ethanol (40 mL) containing catalytic amount of piperidine (0.5 mL) were refluxed for 4 h. The solid product formed was collected by filtration.

Table I Spectral data of the synthesized compounds

Compd. No.	IR (ν, cm^{-1})	^1H NMR (δ, ppm) (DMSO-d_6)
3a	3402 (NH), 2926 (CH-aliph.), 1702, 1688 (C=O thiazolinone, ester).	1.11 (t, 6H, $2CH_3$-ester, J = 5 Hz), 3.77 (q, 4H, $2CH_2$-ester, J = 4.4 Hz), 5.33 (s, 2H, 2-methylidene-H), 6.90 (d, 4H, Ar−H, J = 4.2 Hz), 7.90 (s, 2H, 2-benzylidene-H), 10.32, 10.40 (2s, 2H, 2NH).
3b	3402 (NH), 2980 (CH-aliph.), 1692 (C=O thiazol- inone, ester).	1.22 (t, 6H, $2CH_3$-ester, J = 7 Hz), 4.15 (q, 4H, $2CH_2$-ester, J = 6.8 Hz), 5.64 (s, 2H, 2CH-methylidene), 7.22 (d, 4H, Ar−H, J = 8.6 Hz), 7.35 (d, 4H, Ar−H, J = 8.4),7.42 (s, 2H, 2-benzylidene-H), 12.00 (br, 2H, 2NH).
4a	3406, 3350 (NH_2), 2192 (C≡N), 1700, 1642 (C=O).	1.05, 1.26 (2t, 6H, $2CH_3$-ester, J = 5.2 Hz), 4.06, 4.13 (2q, 4H, $2CH_2$-ester, 4.6 Hz), 5.61 (s, 2H, 2 pyridine-H), 6.89–8.67 (m, 18H, Ar−H + $2NH_2$ + 2-benzylidene-H).
4b	3422, 3370 (NH_2), 2192 (C≡N), 1698, 1640 (C=O thiazolinone, ester).	1.02, 1.22 (2t, 6H, $2CH_3$-ester, J = 5 Hz), 3.84, 4.02 (2s, 6H, $2OCH_3$), 4.04, 4.12 (2q, 4H, $2CH_2$-ester, J = 4.4 Hz), 5.6 (s, 2H, 2 pyridine-H), 7.17–7.80 (m, 18H, Ar−H + $2NH_2$ + 2-benzylidene-H).
4c	3404, 3294 (NH_2), 2198 (C≡N), 1698, 1638 (C=O thiazolinone, ester).	1.03, 1.06 (2t, 6H, $2CH_3$-ester, J = 7 Hz), 4.01, 4.04 (2q, 4H, $2CH_2$-ester, J = 6.4 Hz), 4.51 (s, 1H, pyridine-H), 7.09–7.96 (m, 18H, Ar−H + 2-methylidene-H), 9.92, 9.97 (2s, 4H, $2NH_2$).
4d	3416, 3300 (NH_2), 2202 (C≡N), 1722, 1616 (C=O thiazolinone, ester).	1.02, 1.05 (2t, 6H, $2CH_3$-ester, J = 6.6 Hz), 4.01, 4.51 (2q, 4H, $2CH_2$-ester, 6.4 Hz), 5.61 (s, 1H, pyridine-H), 6.90–7.70 (m, 20H, Ar−H + 2-methylidene-H + $2NH_2$).
5a	3448 (NH), 2924 (CH-aliph.), 1692 (C=O thiazo-linone, ester).	1.12 (s, 3H, CH_3), 2.26 (t, 3H, CH_3-ester, J = 7.6 Hz), 4.11 (q, 2H, CH_2-ester, J = 6.4 Hz), 5.63 (s, 1H, methylidene-H), 7.11 (d, 4H, Ar−H, J = 6 Hz), 7.53 (s,1H benzylidene-H), 12.01 (s, 1H, NH).
5b	3166 (NH), 2978 (CH-aliph.), 1700 (C=O thiazo- linone, ester).	1.19 (t, 3H, CH_3-ester, J = 7 Hz), 3.84 (s, 3H, OCH_3), 4.13 (q, 2H, CH_2-ester, J = 8.4 Hz), 5.65 (s, 1H, methylidene-H), 7.06 (d, 4H, Ar−H, J = 8.2 Hz), 7.62 (s, 2H, benzylidene-H), 12.01 (br, 1H, NH).
5c	3166 (NH), 2900 (CH-aliph.), 1692 (C=O thiazo-linone, ester).	1.13 (t, 3H, CH_3-ester, J = 7 Hz), 3.05 (s, 6H, $2NCH_3$), 4.17 (q, 2H, CH_2-ester,J = 6.6 Hz), 5.51 (s, 1H, methylidene-H), 7.23- (d, 4H, Ar−H, J = 8.4 Hz), 8.45 (s, 2H, benzylidene-H), 12 .01 (s, 1H, NH).
6	3468, 3410 (NH_2), 2188 (C≡N), 1694 (C=O thiazol- inone, ester).	1.04, 1.22 (2t, 6H, $2CH_3$-ester, J = 7.2 Hz), 3.03, 3.11 (s, 12H, $2N(CH_3)_2$), 4.05, 4.49 (2q, 4H, $2CH_2$-ester, J = 6.6 Hz), 5.57, 5.62 (2s, 2H, 2 pyridine-H), 6.87–7.87 (m, 20H, Ar−H + methine-H + 2NH).
7a	3432, 3220 (NH_2), 2228 (C≡N), 1686, 1654 (C=O thiazolinone, ester).	1.07 (t, 3H, CH_3), 2.6 (s, 3H, CH_3-ester, J = 4.8 Hz), 3.75 (s, 3H, OCH_3), 4.12 (q, 2H, CH_2-ester, J = 5 Hz), 4.44 (s, 1H, pyridine-H), 7.18, 7.40 (m, 9H, Ar−H + NH_2), 7.56 (s, 1H, methine-H), 9.01 (br, 1H, OH).
7b	3402, 3296 (NH_2), 2194 (C≡N), 1708, 1644 (C=O thiazolinone, ester).	1.03 (t, 3H, CH_3-ester, J = 7 Hz), 2.27 (s, 3H, CH_3), 3.89 (s, 3H, OCH_3), 4.12 (q, 2H, CH_2-ester, J = 7.2 Hz), 4.42 (s, 1H, pyridine-H), 7.13, 7.62 (m, 10H, Ar−H + NH_2), 7.69 (s, 1H, methine-H).
7c	3308, 3212 (NH_2), 2216 (C≡N), 1692, 1644 (C=O thiazolinone, ester).	1.05 (t, 3H, CH_3-ester, J = 4..8 Hz), 3.72, 3.84 (2s, 6H, $2OCH_3$), 4.03 (s, 1H, pyridine-H), 4.06 (q, 2H, CH_2-ester, J = 4.6 Hz), 6.60–7.69 (m, 10H, Ar−H + NH_2 + methine-H), 9.01 (br, 1H, OH).
7d	3378, 3282 (NH_2), 2198 (C≡N), 1678, 1646 (C=O thiazolinone, ester).	1.03 (t, 3H, CH_3-ester, J = 6.2 Hz), 3.31 (s, 6H, $N(CH_3)_2$), 3.37 (s, 3H, OCH_3), 4.00 (q, 2H, CH_2-ester, J = 6 Hz), 4.50 (s, 1H, pyridine-H), 7.29–7.74 (m, 11H, Ar−H + NH_2 + methine-H).
9	3292, 3132 (2NH), 1666 (C=O thiazolinone, amide)	3.70 (s, 2H, CH_2-thiazolinone), 5.78 (s, 1H, CH), 7.26–7.65 (d, 4H, Ar−H, J = 1.8 Hz), 9.92 (s, 1H, NH), 11.50 (br, 1H, NH).
10	3312 (NH), 1688, 1670 (C=O thiazolinone, amide)	6.90–7.86 (m, 13H, Ar−H + 2H-methine-H), 8.91 (br, 1H, NH).
11	3360 (NH), 1668 (C=O, amide).	7.14, 7.63 (m, 13H, Ar−H + pyrazole-H), 10.33, 10.51 (2s, 2H, 2NH).
12	3050, 3230 (NH), 1688, 1662 (C=O thiazolinone, amide).	
16a	3456,3180 (NH,OH), 2182 (C≡N), 1710, 1648 (C=O thiazolinone, amide).	3.80, 3.38 (2s, 6H, $2OCH_3$), 4.98 (s, 1H, pyri-dine-H), 6.85–7.67 (m, 13H, Ar−H + methine-H), 9.42 (s, 1H, NH), 11.90 (hump, 1H, OH).
16b	3475, 3316 (NH, OH), 2184 (C≡N), 1714, 1650 (C=O thiazolinone, amide)	5.05 (s, 1H, pyridine-H), 7.26–7.71 (m, 13H, Ar−H + methine-H), 9.49 (s, 1H, NH), 12.10 (br, 1H, OH).

Table II Physical and analytical data for the synthesized compounds

Compd. No.	Mp (°C)	Yield (%)	Solvent cryst.	Molecular formula (Mol. Wt.)	Elemental analyses Calcd./found		
					C%	H%	N%
3a	211–213	67	EtOH	$C_{22}H_{20}N_2O_6S_2$ (472)	55.93	4.23	5.93
					55.95	4.25	5.97
3b	191–193	72	EtOH	$C_{28}H_{24}N_2O_7S_2$ (564)	59.57	4.25	4.96
					59.55	4.20	4.96
4a	271–273	69	EtOH	$C_{42}H_{30}N_6Cl_2O_6S_2$ (849)	59.36	3.53	9.89
					59.46	3.50	9.90
4b	281–283	70	EtOH	$C_{44}H_{36}N_6O_8S_2$ (840)	62.85	4.28	10.00
					62.87	4.30	10.05
4c	225–227	74	EtOH	$C_{48}H_{34}N_6Cl_2O_7S_2$ (941)	61.21	3.61	8.92
					61.23	3.61	8.91
4d	235–237	71	EtOH	$C_{48}H_{32}N_6Cl_4O_7S_2$ (1010)	57.02	3.16	8.31
					57.03	3.16	8.33
5a	180–182	65	EtOH	$C_{15}H_{15}NO_3S$ (289)	62.28	5.19	4.84
					62.21	5.20	4.85
5b	195–197	68	EtOH	$C_{15}H_{15}NO_4S$ (305)	59.01	4.91	4.59
					59.00	4.50	4.60
5c	192–194	69	EtOH	$C_{16}H_{18}N_2O_3S$ (318)	60.37	5.66	8.80
					60.40	5.65	8.81
6	271–273	82	EtOH	$C_{52}H_{46}N_8O_7S_2$ (958)	65.13	4.80	11.69
					65.10	4.70	11.60
7a	225–227	74	EtOH	$C_{26}H_{23}N_3O_5S$ (489)	61.34	4.70	8.58
					61.30	4.71	8.59
7b	228–230	70	EtOH	$C_{26}H_{23}N_3O_4S$ (473)	65.46	4.86	8.87
					65.46	4.85	8.86
7c	241–243	65	EtOH	$C_{26}H_{23}N_3O_6S$ (505)	61.78	4.55	8.31
					61.78	4.55	8.33
7d	235–237	72	EtOH	$C_{27}H_{26}N_4O_4S$ (502)	64.54	5.17	11.15
					64.53	5.17	11.15
9	235–237	72	Bz*	$C_{11}H_{19}N_2ClO_2S$ (268.5)	49.16	7.07	10.42
					49.16	7.06	10.40
10	292–294	79	EtOH/Bz	$C_{25}H_{15}N_2Cl_3O_2S$ (512.5)	58.53	2.92	5.46
					58.93	2.92	5.76
11	>300	71	EtOH/Bz	$C_{25}H_{16}N_4Cl_3OS$ (525.5)	57.08	3.04	10.05
					57.11	3.01	10.00
12	281–283	76	EtOH/Bz	$C_{19}H_{12}N_3ClO_3S$ (397.5)	57.35	3.01	10.56
					57.35	2.97	10.60
16a	291–293	73	EtOH	$C_{30}H_{22}N_3ClO_3S$ (571.5)	62.99	3.84	7.34
					62.95	3.86	7.37
16b	295–297	71	EtOH	$C_{28}H_{16}N_3Cl_3O_3S$ (580.5)	57.88	2.75	7.23
					57.93	2.77	7.25

*Bz = benzene.

2-N-(4-Chlorophenyl)acetamido-4-oxo-4,5-dihydro-1,3-thiazole (9)

To a solution of cyanoacetanilide derivative **8** (0.01 mol) in acetic acid (30 mL), thioglycolic acid (0.01 mol) was added. The reaction mixture was heated under reflux for 3 h. The solid product formed was collected by filtration.

2-[5-(4-Chlorobenzylidene)-4-oxo-4,5-dihydro-1,3-thiazolo-2-yl]1,3-N-bis(4-chlorophenyl)acrylamide (10)

To a solution of **9** (0.01 mol) in dioxane (30 mL) containing a catalytic amount of piperidine (0.5 mL), *p*-chlorobenzaldehyde (0.01 mol) was added. The reaction mixture was refluxed for 3 h. The solid product formed was collected by filtration.

1,3-N-Bis(4-chlorophenyl)-2-[3-(4-chlorophenyl)-1H-pyrazolo-[3,4-d]thiazol-5-yl]acrylamide (11)

Equimolar amounts of 4-thiazolinone derivative **10** (0.01 mol) and hydrazine hydrate (0.01 mol) in dioxane (30 mL) was refluxed for 6 h. The solid product formed was collected by filtration.

N-(4-Chlorophenyl)-2-[4-oxo-5-(2-oxo-1,2-dihydro-indol-3-ylidene)thiazolidine]acetamide (12)

To a solution of 4-thiaozlidinone **9** (0.01 mol) in dioxane (30 mL) containing a catalytic amount of piperidine (0.5 mL), isatin (0.01 mol) was added. The reaction mixture was heated under reflux for 6 h. The solid product formed was collected by filtration.

2-(Arylmethylidene)-7-aryl-6-cyano-5-hydroxy-3-oxo-2,3-dihydro-7H-thiazolo[3,2-a]pyridine-8-carboxylic Acid (4-Chloro-phenyl) Amide (16a,b)

To a solution of compound **9** (0.01 mol) in dioxane (30 mL) containing a catalytic amount of piperidine (0.5 mL), α-ethoxy-carbonylcinnamonitriles (0.01 mol) was added. The reaction mixture was heated under reflux for 6 h. The solid product formed was collected by filtration.

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