

SYNTHESIS AND SOME REACTIONS OF 2-CYANO-1,6-DIOXO-3-METHYLPYRIDO[1,2-*a*][3,1]BENZOXAZINE

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The importance of pyrido compounds as analgesic, antiinflammatory and central nervous system depressing agents¹⁻⁴ together with the diverse biological activities which have been encountered in compounds containing quinazoline moiety⁵⁻¹³ led us to the synthesis of 2-cyano-1,6-dioxo-3-methylpyrido[1,2-*a*][3,1]benzoxazine (*IV*) to be used as a starting material for pyridoquinazoline derivatives *V-IX* (Schemes 1, 2).

EXPERIMENTAL

Melting points were determined in open glass capillaries and are uncorrected. IR spectra (KBr, cm⁻¹) were recorded on a Pye-Unicam SP 200G spectrophotometer. ¹H NMR spectra were measured in hexadeuteriodimethyl sulfoxide using TMS as internal standard on EM 360 90 MHz NMR spectrophotometer. Chemical shifts are given in ppm (δ-scale). Microanalyses were determined on a Perkin-Elmer 240 C microanalyzer.

1,2-Dihydro-6-hydroxy-4-methyl-2-oxo-3-pyridinecarbonitrile (*I*)

The title compound was prepared according to the reported method^{1,2} and its structure was confirmed by ¹H NMR spectroscopy.

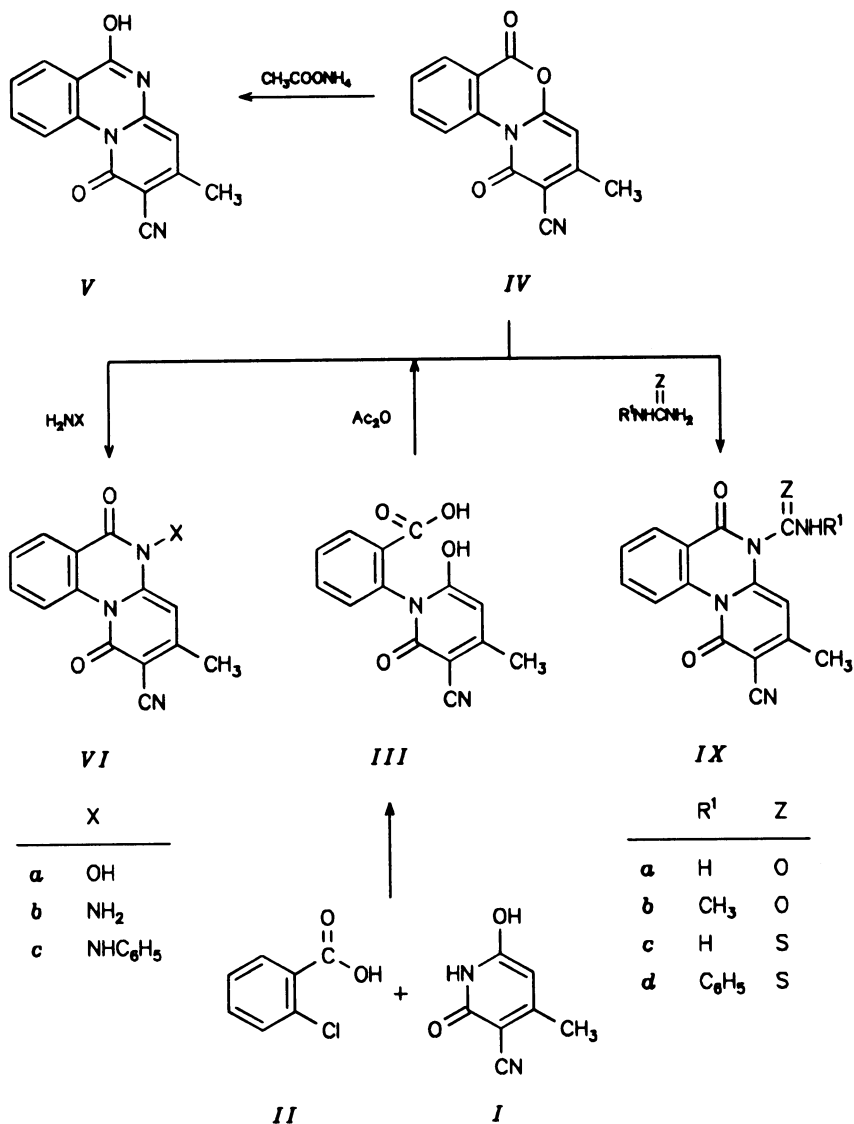
1-(2'-Carboxyphenyl)-6-hydroxy-4-methyl-2-oxo-3-pyridinecarbonitrile (*III*)

A mixture of *I* (15 g, 0.1 mol), *o*-chlorobenzoic acid (*II*) (15.6 g, 0.1 mol), anhydrous potassium carbonate (13.8 g, 0.1 mol) and copper oxide (0.25 g) in dry toluene (150 ml) was refluxed for 10 h. The reaction mixture was cooled to room temperature and toluene was removed under reduced pressure. The residue was dissolved in cold water (200 ml) and acidified with cold 10% hydrochloric acid. The precipitated white crystals were recrystallized from diluted acetic acid to give 23 g (85%) of *III*, m.p. 320 – 322 °C. For C₁₄H₁₀N₂O₄ (270.2) calculated: 62.22% C, 3.70% H, 10.37% N; found: 62.00% C, 3.40% H, 10.10% N. IR spectrum: 3 500, 3 300 (OH); 2 950 (CH arom.); 2 830 (CH aliph.); 2 250 (C≡N); 1 720 (C=O). ¹N NMR spectrum: 2.4 s, 3 H; 3.4 s, 2 H; 7.00 – 7.50 m, 4 H; 9.9 s, 1 H.

2-Cyano-1,6-dioxo-3-methylpyrido[1,2-*a*][3,1]benzoxazine (*IV*)

Nitrile *III* (20 g, 0.74 mol) was dissolved in 150 ml of acetic anhydride and refluxed for 6 h. Acetic anhydride was then removed by distillation under reduced pressure to afford the crude product which

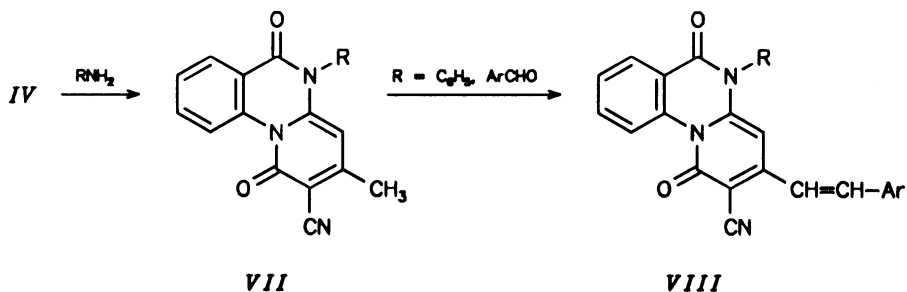
was washed several times with petroleum ether (b.p. 40 – 60 °C), collected by filtration and dried to yield 17 g (91%) of IV, m.p. 260 – 262 °C. For $C_{14}H_8N_2O_3$ (252.2) calculated: 66.67% C, 3.19% H, 11.11% N; found: 66.20% C, 3.00% H, 11.00% N. IR spectrum: 2 995 (CH arom.); 2 820 (CH aliph.); 2 200 (C≡N); 1 760 (C=O). 1H NMR spectrum: 2.4 s, 3 H; 7 – 7.5 m, 4 H; 8.1 s, 1 H.

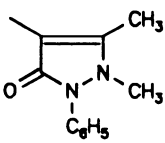


SCHEME 1

2-Cyano-6-hydroxy-3-methyl-1-oxopyrido[1,2-*a*]quinazoline (V)

Quinazolinone IV (0.252 g, 0.001 mol) was mixed with ammonium acetate (0.77 g, 0.01 mol) and fused at 200 °C for 4 h, then the reaction mixture was cooled to room temperature and poured into cold water. The precipitated pale yellow powder was collected by filtration and crystallized from ethanol to give 0.215 g (86%) of V, m.p. 345 – 347 °C. For $C_{14}H_9N_3O_2$ (251.2) calculated: 66.94% C, 3.61% H, 16.73% N; found: 66.60% C, 3.25% H, 16.40% N. IR spectrum: 3 500, 3 300 (OH); 3 050 (CH arom.); 2 920 (CH aliph.); 2 250 (C≡N); 1 720 (C=O). 1H NMR spectrum: 2.3 s, 3 H; 7.3 – 7.6 m, 4 H; 7.7 s, 1 H; 8.1 s, 1 H.



R	R	Ar
<i>a</i> C ₆ H ₅	<i>l</i> 2-HO ₂ CC ₆ H ₄	<i>a</i> C ₆ H ₅
<i>b</i> C ₆ H ₅ CH ₂	<i>m</i> 4-HO ₃ SC ₆ H ₄	<i>b</i> 4-ClC ₆ H ₄
<i>c</i> 4-CH ₃ OC ₆ H ₄	<i>n</i> 2-pyridyl	<i>c</i> 4-NO ₂ C ₆ H ₄
<i>d</i> 1-naphthyl	<i>o</i> 3-pyridyl	<i>d</i> 4-CH ₃ OC ₆ H ₄
<i>e</i> 4-CH ₃ COC ₆ H ₄	<i>p</i> 2-thiazolyl	<i>e</i> 4-(CH ₃) ₂ NC ₆ H ₄
<i>f</i> 4-NO ₂ C ₆ H ₄	<i>q</i> 	
<i>g</i> 2-NO ₂ C ₆ H ₄	<i>r</i> CH ₂ CH(CH ₃) ₂	
<i>h</i> 2-HOC ₆ H ₄	<i>s</i> CH ₃	
<i>i</i> 3-HOC ₆ H ₄		
<i>j</i> 4-HOC ₆ H ₄		
<i>k</i> 2-NH ₂ C ₆ H ₄		

SCHEME 2

2-Cyano-1,6-dioxo-5-hydroxy-3-methylpyrido[1,2-*a*]quinazoline (*Vla*)

Quinazolone *IV* (0.252 g, 0.001 mol) was dissolved in pyridine (5 ml) and absolute ethanol (5 ml), hydroxylamine hydrochloride (0.069 g) was added and the solution was refluxed for 3 h. Cooled reaction mixture was concentrated at reduced pressure and the residue was poured into cold 10% hydrochloric acid (10 ml) to afford a pale yellow precipitate. The crude product was collected by filtration, crystallized from ethanol to yield 0.240 g (90%) of pure *Vla*, m.p. 240 – 242 °C. For $C_{14}H_9N_3O_3$ (267.2) calculated: 62.93% C, 3.39% H, 15.73% N; found: 62.60% C, 3.10% H, 15.40% N. IR spectrum: 3 500 (OH); 3 050 (CH arom.); 2 920 (CH aliph.); 2 200 (C≡N); 1 740, 1 720 (C=O). 1H NMR spectrum: 2.3 s, 3 H; 4.5 brs, 1 H; 7.3 – 7.6 m, 4 H; 8.1 s, 1 H.

5-Amino-2-cyano-1,6-dioxo-3-methylpyrido[1,2-*a*]quinazoline (*VIb*)

Quinazolone *IV* (0.252 g, 0.001 mol) was dissolved in absolute ethanol (10 ml), hydrazine hydrate (0.1 ml) was added and the reaction mixture was refluxed for 3 h. The reaction mixture was concentrated under reduced pressure, the white precipitate was filtered off and crystallized from ethanol to afford 0.239 g (90%) of *VIb*, m.p. 270 – 272 °C. For $C_{14}H_{10}N_4O_2$ (266.3) calculated: 63.15% C, 3.78% H, 21.04% N; found: 63.00% C, 3.50% H, 20.70% N. IR spectrum: 3 400, 3 300 (NH₂); 3 050 (CH arom.); 2 890 (CH aliph.); 2 200 (C≡N); 1 780, 1 720 (C=O). 1H NMR spectrum: 2.3 s, 3 H; 5.4 brs, 2 H; 7.3 – 7.7 m, 4 H; 8.2 s, 1 H.

5-Phenylamino-2-cyano-1,6-dioxo-3-methylpyrido[1,2-*a*]quinazoline (*VIc*)

A mixture of quinazolone *IV* (0.252 g, 0.001 mol) and phenylhydrazine (0.109 g, 0.001 mol) in absolute ethanol (10 ml) was refluxed for 6 h. Ethanol was removed under reduced pressure to yield 0.273 g (80%) of *VIc*, m.p. 130 – 132 °C (methanol). For $C_{20}H_{14}N_4O_2$ (342.4) calculated: 70.16% C, 4.12% H, 16.37% N; found: 70.00% C, 4.00% H, 16.11% N. IR spectrum: 3 400 (NH); 3 050 (CH arom.); 2 890 (CH aliph.); 2 200 (C≡N); 1 740 (C=O). 1H NMR spectrum: 2.3 s, 3 H; 6.4 brs, 1 H; 7.3 – 7.6 m, 9 H; 8.1 s, 1 H.

5-(Alkyl or aryl)-2-cyano-1,6-dioxo-3-methylpyrido[1,2-*a*]quinazoline (*VIIa* – *VIIs*). General Procedure

A mixture of quinazolone *IV* (0.001 mol) and primary aliphatic or aromatic amine (0.001 mol) in absolute ethanol (10 ml) was refluxed for 6 h. Ethanol was removed under reduced pressure, the product *VII* was crystallized from a suitable solvent. The yields and the physical properties of compounds *VIIa* – *VIIs* are given in Table I.

5-Phenyl-2-cyano-1,6-dioxo-3-styrylpyrido[1,2-*a*]quinazoline (*VIIIa* – *VIIIc*). General Procedure

A mixture of quinazoline *VIIa* (0.001 mol), of the appropriate aromatic aldehyde (0.001 mol) in absolute ethanol (10 ml) and one drop of piperidine was refluxed for 5 h. The solvent was concentrated to half of its volume, precipitated solid was filtered off and crystallized from a suitable solvent. The yields and the physical properties of compounds *VIIIa* – *VIIIc* are given in Table II.

5-Amino(thio)carbonyl-2-cyano-1,6-dioxo-3-methylpyrido[1,2-*a*]quinazoline (*IXa* – *IXd*). General Procedure

A mixture of quinazolone *IV* (0.001 mol) and urea, methylurea, thiourea or phenylthiourea (0.001 mol) in a mixture of absolute ethanol–pyridine 3 : 1 was refluxed for 7 h. The solvents were removed

TABLE I
Physico-chemical data of the pyridoquinazolines VIIa – VIIf

Compound	Formula M. w.	M. p., °C Solvent	Yield, %	Calculated/Found			IR (KBr, cm ⁻¹)	¹ H NMR (δ, ppm)
				% C	% H	% N		
VIIa	C ₂₀ H ₁₃ N ₃ O ₂ 327.3	340 – 342 ethanol	90	73.39 73.00	4.00 3.60	12.84 12.50	3 020 (CH arom.), 2 900 (CH aliph.), 2 200 (C=N), 1 720 (C=O)	2.3 s, 3 H; 7 – 7.7 m, 9 H; 8.1 s, 1 H
VIIb	C ₂₁ H ₁₅ N ₃ O ₂ 341.4	260 – 263 ethanol	92	73.88 73.60	4.42 4.20	12.31 12.00	3 050 (CH arom.), 2 890 (CH aliph.), 2 200 (C=N), 1 725 (C=O)	2.3 s, 3 H; 3.4 s, 2 H; 7.3 – 7.7 m, 9 H; 8.1 s, 1 H
VIIc	C ₂₁ H ₁₅ N ₃ O ₃ 357.4	320 – 322 ^a ethanol	88	70.57 70.20	4.23 4.00	11.76 11.60	3 030 (CH arom.), 2 890 (CH aliph.), 2 200 (C=N), 1 720 (C=O)	2.3 s, 3 H; 3.6 s, 3 H; 7.3 – 7.6 m, 8 H; 8.1 s, 1 H
VII d	C ₂₁ H ₁₅ N ₃ O ₂ 377.4	360 ^b methanol	82	76.38 76.10	4.00 3.80	11.14 11.00	3 050 (CH arom.), 2 850 (CH aliph.), 2 200 (C=N), 1 715 (C=O)	2.3 s, 3 H; 7.3 – 7.9 m, 11 H; 8.1 s, 1 H
VIIe	C ₂₂ H ₁₅ N ₃ O ₃ 369.4	175 – 177 ethanol	80	71.47 71.30	4.09 4.00	11.38 11.10	3 050 (CH arom.), 2 900 (CH aliph.), 2 200 (C=N), 1 720 (C=O)	2.3 s, 3 H; 2.6 s, 3 H; 7.3 – 7.6 m, 8 H; 8.1 s, 1 H
VII f	C ₂₀ H ₁₂ N ₄ O ₄ 372.3	280 – 283 ethanol	81	64.52 64.40	3.25 3.00	15.05 14.80	3 050 (CH arom.), 2 900 (CH aliph.), 2 250 (C=N), 1 740 1 690 (C=O)	2.3 s, 3 H; 7.3 – 7.6 m, 8 H; 8.1 s, 1 H

TABLE I
(Continued)

Compound	Formula M. w.	M. p., °C Solvent	Yield, %	Calculated/Found			IR (KBr, cm ⁻¹)	¹ H NMR (δ, ppm)
				% C	% H	% N		
VIIg	C ₂₀ H ₁₂ N ₄ O ₄ 372.3	290 – 292 ethanol	78	64.52 64.30	3.25 3.10	15.05 14.70	3 050 (CH arom.), 2 850 (CH aliph.), 2 225 (C≡N), 1 740, 1 690 (C=O)	2.3 s, 3 H; 7.3 – 7.7 m, 8 H; 8.1 s, 1 H
VIIh	C ₂₀ H ₁₃ N ₃ O ₃ 343.4	320 – 322 ethanol	75	69.95 69.60	3.82 3.50	12.24 12.00	3 500 (OH), 3 035 (CH arom.), 2 900 (CH aliph.), 2 250 (C≡N), 1 740, 1 690, (C=O)	2.3 s, 3 H; 7.3 – 7.7 m, 8 H; 8.1 s, 1 H; 8.6 s, 1 H
VIIi	C ₂₀ H ₁₃ N ₃ O ₃ 343.4	330 – 333 ethanol	78	69.95 69.70	3.82 3.49	12.24 12.00	3 500 (OH), 3 030 (CH arom.), 2 890 (CH aliph.), 2 200 (C≡N), 1 740, 1 690 (C=O)	2.3 s, 3 H; 7.3 – 7.7 m, 8 H; 8.1 s, 1 H; 8.7 s, 1 H
VIIj	C ₂₀ H ₁₃ N ₃ O ₃ 343.4	310 – 312 ethanol	79	69.95 69.50	3.82 3.60	12.24 12.10	3 500 (OH), 3 030 (CH arom.), 2 900 (CH aliph.), 2 200 (C≡N), 1 740, 1 690 (C=O)	2.3 s, 3 H; 7.3 – 7.8 m, 8 H; 8.2 s, 1 H; 8.9 s, 1 H
VIIk	C ₂₀ H ₁₄ N ₄ O ₂ 342.4	190 – 192 ethanol	75	70.16 70.00	4.12 4.00	16.36 16.00	3 400 – 3 350 (NH ₂), 3 030 (CH arom.), 2 900 (CH aliph.), 2 225 (C≡N), 1 715, 1 690 (C=O)	2.3 s, 3 H; 6.4 brs, 2 H; 7.3 – 7.7 m, 8 H; 8.2 s, 1 H
VIII	C ₂₁ H ₁₃ N ₃ O ₄ 371.4	337 – 339 ^b ethanol	70	67.91 67.60	3.53 3.30	11.32 11.00	3 500, 3 450 (OH), 3 050 (CH arom.), 2 890 (CH aliph.), 2 200 (C≡N), 1 740, 1 690 (C=O)	2.3 s, 3 H; 7.3 – 7.7 m, 8 H; 8.1 s, 1 H; 9.9 s, 1 H

TABLE I
(Continued)

Compound	Formula M. w.	M. p., °C Solvent	Yield, %	Calculated/Found			IR (KBr, cm ⁻¹)	¹ H NMR (δ, ppm)
				% C	% H	% N		
<i>VII m</i> ^c	C ₂₀ H ₁₃ N ₃ O ₅ S 407.4	220 – 222 ^b acetic acid	72	58.96 58.60	3.22 3.00	10.32 10.00	3 030 (CH arom.), 2 890 (CH aliph.), 2 200 (C≡N), 1 715, 1 670 (C=O)	2.3 s, 3 H; 7.2 – 7.7 m, 8 H; 8.2 s, 1 H; 9.8 s, 1 H
<i>VII n</i>	C ₁₉ H ₁₂ N ₄ O ₂ 328.3	235 – 237 ethanol	80	69.51 69.30	3.68 3.40	17.07 17.00	3 050 (CH arom.), 2 900 (CH aliph.), 2 225 (C≡N), 1 720, 1 690 (C=O)	2.3 s, 3 H; 7.3 – 7.6 m, 7 H; 8.2 m, 2 H;
<i>VII o</i>	C ₁₉ H ₁₂ N ₄ O ₂ 328.3	360 ^b ethanol	78	69.51 69.10	3.66 3.36	17.07 17.00	3 030 (CH arom.), 2 900 (CH aliph.), 2 220 (C≡N), 1 715, 1 690 (C=O)	2.3 s, 3 H; 7.3 – 7.6 m, 7 H; 8.2 m, 2 H;
<i>VII p</i> ^d	C ₁₇ H ₁₀ N ₄ O ₂ S 334.3	170 – 172 ethanol	70	61.08 61.00	3.01 2.70	16.76 16.50	3 050 (CH arom.), 2 900 (CH aliph.), 2 250 (C≡N), 1 720 (C=O)	2.3 s, 3 H; 7.3 – 7.6 m, 4 H; 8.3 m, 3 H;
<i>VII q</i>	C ₂₅ H ₁₉ N ₅ O ₃ 437.5	190 – 192 acetic acid	70	68.63 68.30	4.38 4.10	16.01 16.00	3 050 (CH arom.), 2 900 (CH aliph.), 2 250 (C≡N), 1 715, 1 680 (C=O)	2.3 s, 3 H; 2.5 s, 3 H; 3.2 s, 3 H; 7.3 – 7.8 m, 9 H; 8.1 s, 1 H
<i>VII r</i>	C ₁₈ H ₁₇ N ₅ O ₂ 307.4	325 – 327 ethanol	68	70.33 70.10	5.57 5.20	13.67 13.30	3 030 (CH arom.), 2 890 (CH aliph.), 2 250 (C≡N), 1 700, 1 690 (C=O), 1 370, 1 310 (gem. methyl)	0.9 – 1.1 d, 6 H; 2.2 – 2.7 m, 5 H; 4.0 m, 1 H; 7.3 – 7.6 m, 4 H; 8.2 s, 1 H
<i>VII s</i>	C ₁₅ H ₁₁ N ₃ O ₂ 265.3	250 – 252 ethanol	65	67.91 67.60	4.18 4.00	15.84 15.60	3 050 (CH arom.), 2 850 (CH aliph.), 2 225 (C≡N), 1 720, 1 690 (C=O)	2.3 s, 3 H; 3.4 s, 3 H; 7.2 – 7.6 m, 4 H; 8.2 s, 1 H

^a Sublimation. ^b Decomposition. ^c Calculated 7.87% S, found 7.50% S. ^d Calculated 9.59% S, found 9.30% S.

TABLE II
Physico-chemical data of the styrylpyrido[1,2-*a*]quinazolines *VIIIa* – *VIIIe*

Compound	Formula M. w.	M. p., °C Solvent	Yield, %	Calculated/Found			IR (KBr, cm ⁻¹)	¹ H NMR (δ, ppm)
				% C	% H	% N		
<i>VIIIa</i>	C ₂₇ H ₁₇ N ₃ O ₂ 415.5	270 – 272 ethanol	75	78.02 78.00	4.12 4.00	10.12 10.00	3 050 (CH arom.), 2 900 (CH aliph.), 2 225 (C≡N), 1 740, 1 690 (C=O)	6,7 s, 2 H; 7,3 – 7,7 m, 14 H; 8,1 s, 1 H
<i>VIIIb</i>	C ₂₇ H ₁₆ ClN ₃ O ₂ 449.9	230 – 232 ethanol	78	72.08 72.00	3.58 3.26	9.34 9.20	3 030 (CH arom.), 2 890 (CH aliph.), 2 250 (C≡N), 1 740, 1 690 (C=O)	6,6 s, 2 H; 7,3 – 7,9 m, 13 H; 8,1 s, 1 H
<i>VIIIc</i>	C ₂₇ H ₁₆ N ₄ O ₄ 460.5	210 – 212 methanol	80	70.42 70.20	3.50 3.20	12.17 12.00	3 060 (CH arom.), 2 900 (CH aliph.), 2 225 (C≡N), 1 740, 1 700 (C=O)	6,7 s, 2 H; 7,3 – 7,9 m, 13 H; 8,2 s, 1 H
<i>VIII d</i>	C ₂₈ H ₁₉ N ₃ O ₃ 445.5	250 – 252 ^a methanol	76	75.49 75.20	4.30 4.00	9.43 9.20	3 030 (CH arom.), 2 890 (CH aliph.), 2 250 (C≡N), 1 740, 1 690 (C=O)	3,7 s, 3 H; 6,7 s, 2 H; 7,3 – 7,9 m, 13 H; 8,2 s, 1 H
<i>VIIIe</i>	C ₂₉ H ₂₂ N ₄ O ₂ 458.5	315 – 317 ^a methanol	73	75.97 75.60	4.84 4.50	12.22 12.00	3 050 (CH arom.), 2 900 (CH aliph.), 2 225 (C≡N), 1 740, 1 690 (C=O)	3,2 s, 6 H; 6,7 s, 2 H; 7,3 – 7,9 m, 13 H; 8,1 s, 1 H

^a Decomposition.

TABLE III
Physico-chemical data of 5-amino(thio)carbonyl-2-cyano-1,6-dioxo-3-methylpyrido[1,2-*a*]quinazolines IXa – IXd

Compound	Formula M. w.	M. p., °C Solvent	Yield, %	Calculated/Found				IR (KBr, cm ⁻¹)	¹ H NMR (δ, ppm)
				% C	% H	% N	% S		
IXa	C ₁₅ H ₁₀ N ₄ O ₃ 294.3	295 – 297 ethanol	65	61.22 61.00	3.43 3.10	19.04 18.80		3 400, 3 300 (NH ₂), 3 030 (CH arom.), 2 890 (CH aliph.), 2 225 (C=N), 1 740, 1 680 (C=O)	2.3, s, 3 H; 6.3 brs, 2 H; 7.3 – 7.7 m, 4 H 8.2 s, 1 H
IXb	C ₁₆ H ₁₂ N ₄ O ₃ 308.3	210 – 212 ethanol	60	62.28 62.00	3.92 3.60	18.18 18.00		3 350 (NH), 3 030 (CH arom.), 2 900 (CH aliph.), 2 250 (C=N), 1 740, 1 710 (C=O)	2.35, s, 3 H; 6.1 brs, 1 H; 7.3 – 7.6 m, 4 H 8.2 s, 1 H
IXc	C ₁₅ H ₁₀ N ₄ O ₂ S 310.3	195 – 197 ethanol	65	58.06 58.00	3.25 3.00	18.06 18.00	10.32 10.10	3 400, 3 300 (NH ₂), 3 050 (CH arom.), 2 900 (CH aliph.), 2 250 (C=N), 1 740, 1 710 (C=O)	2.3, s, 3 H; 6.1 brs, 1 H; 7.3 – 7.7 m, 4 H 8.2 s, 1 H
IXd	C ₂₁ H ₁₄ N ₄ O ₂ S 386.4	290 – 292 methanol	60	65.28 65.00	3.65 3.30	14.50 14.20	8.28 8.00	3 350 (NH), 3 050 (CH arom.), 2 890 (CH aliph.), 2 250 (C=N), 1 740, 1 720 (C=O)	2.3, s, 3 H; 6.1 brs, 1 H; 7.4 – 7.8 m, 9 H 8.1 s, 1 H

under reduced pressure and the residue was poured into cold 10% hydrochloric acid (10 ml). The precipitated product *IX* was crystallized from a suitable solvent. Yields and physico-chemical data of compounds *IXa* – *IXd* are given in Table III.

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