SYNTHESIS OF SOME NEW PYRANOQUINOLINE AND RELATED HETEROCYCLIC SYSTEMS

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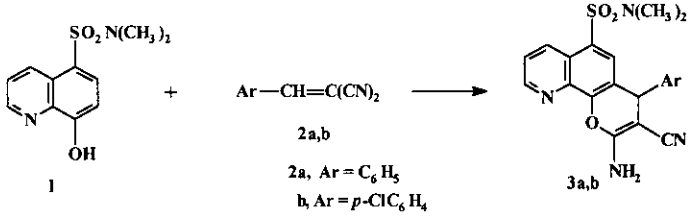
Abstract- The reaction of cinnamonitriles **(2a,b)** with 5-dimethylaminosulfonyl-8-quinolinol **(1)** gave the corresponding pyranoquinoline derivatives **(3a,b)**. The latter derivatives are used in the synthesis of pyrido[2',3':6,5]pyrano[3,2-*h*]quinolines and pyrimido[4',5':6,5]pyrano[3,2-*h*]quinolines.

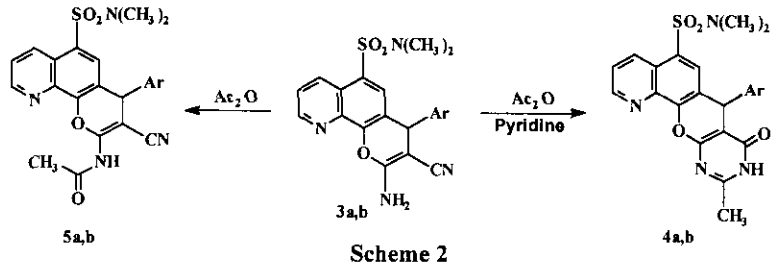
Literature survey has revealed that cinnamonitriles were used as starting materials to prepare pyran derivatives.[1-5] Also, sulfonamides showed divers biological activities.[6] In this respect and in continuation of our work for the synthesis of heterocyclic systems,[7-14] we aimed to report herein the synthesis of some new pyranoquinoline and related heterocyclic systems.

Our syntheses started with the reaction of cinnamonitriles **(2a,b)** with 5-dimethylaminosulfonyl-8-quinolinol **(1)** in ethanol in the presence of catalytic amount of piperidine to give the corresponding pyrano[3,2-*h*]quinolines **(3a,b)** (Scheme 1).



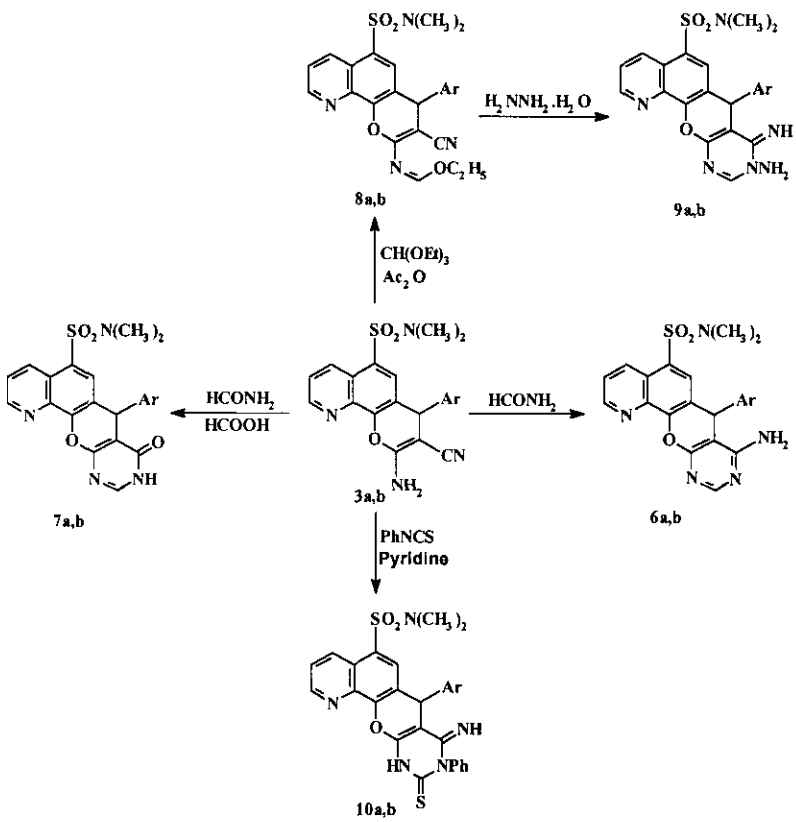
Scheme 1

Compounds **(3a,b)** were subjected to further reactions to give fused heterocyclic systems incorporat pyrimidine and/or pyridine nucleus in addition to pyranoquinoline moiety. Thus, the reaction of **3a,b** with acetic anhydride-pyridine mixture gave pyrimido[4',5':6,5]pyrano[3,2-*h*]quinolines **(4a,b)**, while the reaction with acetic anhydride only gave the acetamido derivatives[15] **(5a,b)** (Scheme 2).



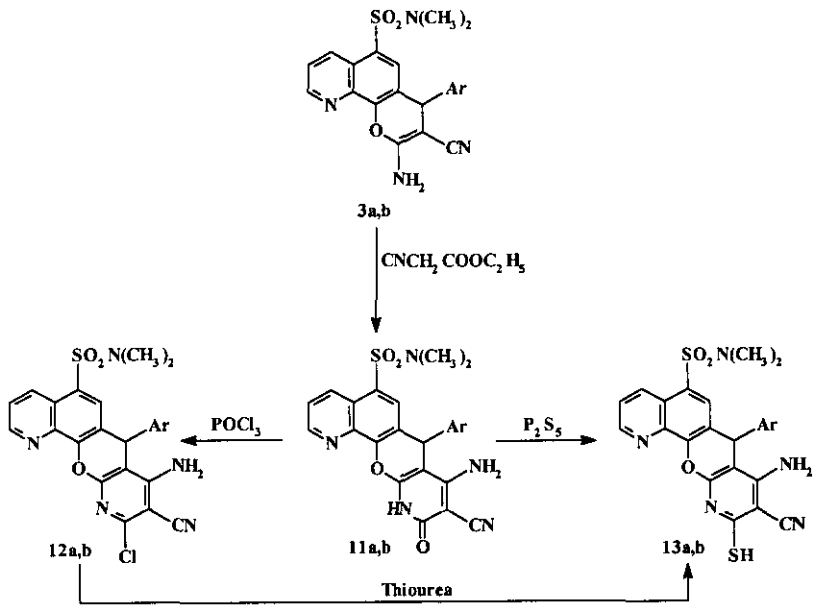
Scheme 2

Interaction of **3a,b** with formamide afforded aminopyrimidine derivatives **(6a,b)**, while the reaction with formamide-formic acid mixture gave the corresponding pyrimidinone derivatives[15] **(7a,b).** Refluxing of **3a,b** with triethyl orthoformate in acetic anhydride gave the corresponding methanimidates **(8a,b)** which reacted with hydrazine hydrate to give 9-amino-7-aryl-5-dimethylaminosulfonyl-8-imino-7,8,9-trihydropyrimido-[4',5':6,5]pyrano[3,2-*h*]quinolines **(9a,b)**, while refluxing of **3a,b** with phenyl isothiocyanate in pyridine produced 7-aryl-5-dimethylaminosulfonyl-8-imino-10-thioxo-7,8,9-10,11-pentahydropyrimido[4',5':6,5]-pyrano[3,2-*h*]quinolines **(10a,b)** (Scheme 3).



Scheme 3

Fusion of **3a,b** with ethyl cyanoacetate afforded 8-amino-7-aryl-9-cyano-5-dimethylaminosulfonyl-10-oxo-pyrido[2',3':6,5]pyrano[3,2-*h*]quinolines **(11a,b)**. Treatment of **11a,b** with phosphorus oxychloride gave 8-amino-7-aryl-10-chloro-9-cyano-5-dimethylaminosulfonylpyrido[2',3':6,5]pyrano[2,3-*h*]quinolines **(12a,b)**, while treatment with phosphorus pentasulfide in refluxing pyridine afforded 8-amino-7-aryl-9-cyano-5-dimethylaminosulfonylpyrido[2',3':6,5]pyrano[2,3-*h*]quinoline-10-thiol **(13a,b)** which were identified by the reaction of compounds **(12a,b)** with thiourea in refluxing ethanol followed by hydrolysis with sodium hydroxide and then acidification with HCl (Scheme 4).



Scheme 4

The chemical structures of the synthesized compounds were confirmed by elemental analyses and spectral data. IR (υ, cm^{-1}) (KBr) showed the absorption bands at 1370-1360 (SO_2, asym.), 1170-1160 (SO_2 sym.), 3400-3300 (NH_2), 3150-3160 (NH), 2200 (CN), 1680-1670 (C=O), 3020-3000 (CH arom.), 2920-2900 (CH aliph.). NMR (δ, ppm) showed the signals in the reasonal regions: 3.40-3.20 (s, 6H, $2CH_3$), 5.00-4.90 (s, 1H, pyran), 6.90-8.40 (*m, aromatic protons*), 1.80-1.45 (t, J = 7.3 Hz, 3H, OCH_2CH_3), 2.90-2.80 (s, 3H, CH_3). The physical and analytical data of pyrano[3,2-*h*]quinolines **(3a,b)**, pyrimido[4',5':6,5]pyrano[3,2-*h*]quinolines **(4a,b-10a,b)** and pyrido[2',3':6,5]pyrano[3,2-*h*]quinolines **(11a,b-13a,b)** are given in Table 1.

Table 1. Physical Data of Pyrano[3,2-*h*]quinolines **(3a,b)**, Pyrimido[4',5':6,5]pyrano[3,2-*h*]quinolines **(4a,b-10a,b)** and Pyrido[2',3':6,5]pyrano[3,2-*h*]quinolines **(11a,b-13a,b)**

Compd. No.	Yield (%)	mp (°C)	Molecular Formula (Solvent of Recrystallization)	IR (υ, cm^{-1}) (KBr) and MS	NMR (δ, ppm) (Solvent)	*Anal.* Calcd/(Found) % C	H	N	S	Cl
3a	60	> 310	$C_{21}H_{18}N_4O_3S$ (ethanol)	3400-3320 (NH_2), 3000 (CH arom.), 2900 (CH aliph.), 2200 (CN), 1360 (SO_2 asym.), 1160 (SO_2 sym.); m/z 406	($CDCl_3$): 3.20 (6H, s), 4.95 (1H, s), 6.60 (2H, s), 7.10-8.40 (9H, m)	62.04 (62.12)	4.46 (4.43)	13.79 (13.86)	7.90 (7.94)	—
3b	69	286-288	$C_{21}H_{17}N_4O_3ClS$ (ethanol)	3400-3300 (NH_2), 3010 (CH arom.), 2900 (CH aliph.), 2200 (CN), 1360 (SO_2 asym.), 1160 (SO_2 sym.)	($CDCl_3$): 3.20 (6H, s), 5.00 (1H, s), 6.80 (2H, s), 7.20-8.10 (8H, m)	57.19 (57.27)	3.89 (3.91)	12.71 (12.67)	7.28 (7.32)	8.05 (8.01)
4a	56	263 dec.	$C_{23}H_{20}N_4O_4S$ (ethanol)	3100 (NH), 3010 (CH arom.), 2920 (CH aliph.), 1680 (C=O), 1370 (SO_2 asym.), 1165 (SO_2 sym.); m/z 448	(CF_3COOD): 2.80 (3H, s), 3.10 (6H, s), 4.90 (1H, s), 6.80-7.90 (9H, m)	61.59 (61.61)	4.50 (4.46)	12.50 (12.43)	7.16 (7.20)	—
4b	61	252-254	$C_{23}H_{19}N_4O_4ClS$ (ethanol)	3100 (NH), 3000 (CH arom.), 2900 (CH aliph.), 1670 (C=O), 1360 (SO_2 asym.), 1160 (SO_2 sym.)	(CF_3COOD): 2.90 (3H, s), 3.25 (6H, s), 4.90 (1H, s), 7.00-8.60 (8H, m)	57.19 (57.25)	3.97 (3.94)	11.60 (11.55)	6.65 (6.59)	7.35 (7.40)
5a	53	248 dec.	$C_{23}H_{20}N_4O_4S$ (dioxane)	3100 (NH), 3000 (CH arom.), 2900 (CH aliph.), 2200 (CN), 1660 (C=O), 1365 (SO_2 asym.), 1160 (SO_2 sym.); m/z 448	($CDCl_3$): 2.90 (3H, s), 3.30 (6H, s), 4.90 (1H, s), 6.90-8.00 (9H, m), 8.90 (1H, s)	61.59 (61.53)	4.50 (4.54)	12.50 (12.56)	7.16 (7.12)	—
5b	55	> 310	$C_{23}H_{19}N_4O_4ClS$ (dioxane)	3130 (NH), 3020 (CH arom.), 2920 (CH aliph.), 2200 (CN), 1670 (C=O), 1360 (SO_2 asym.), 1160 (SO_2 sym.)	(CF_3COOD): 2.80 (3H, s), 3.20 (6H, s), 5.00 (1H, s), 7.00-8.20 (8H, m)	57.19 (57.25)	3.97 (4.00)	11.60 (11.54)	6.65 (6.72)	7.35 (7.31)
6a	48	236 dec.	$C_{22}H_{19}N_5O_3S$ (ethanol)	3400-3300 (NH_2), 3000 (CH arom.), 2900 (CH aliph.), 1360 (SO_2 asym.), 1160 (SO_2 sym.); m/z 433	(CF_3COOD): 3.20 (6H, s), 4.90 (1H, s), 6.90-8.20 (10H, m)	60.95 (60.88)	4.42 (4.38)	16.16 (16.22)	7.41 (7.32)	—
6b	45	253-255	$C_{22}H_{18}N_5O_3ClS$ (ethanol)	3400-3300 (NH_2), 3030 (CH arom.), 2920 (CH aliph.), 1360 (SO_2 asym.), 1160 (SO_2 sym.)	(CF_3COOD): 3.20 (6H, s), 5.00 (1H, s), 7.00-8.30 (9H, m)	56.46 (56.51)	3.88 (3.91)	14.97 (15.01)	6.86 (6.92)	7.59 (7.62)
7a	43	241 dec.	$C_{22}H_{18}N_4O_4S$ (ethanol)	3150 (NH), 3010 (CH arom.), 2900 (CH aliph.), 1670 (C=O), 1360 (SO_2 asym.), 1160 (SO_2 sym), m/z 433	(CF_3COOD): 3.30 (6H, s), 5.00 (1H, s), 6.90-8.20 (10H, m)	60.81 (60.74)	4.18 (4.22)	12.90 (12.84)	7.39 (7.44)	—

(Continued)

Table 1. ***(Continued)*** Physical Data of Pyrano[3,2-*h*]quinolines **(3a,b)**, Pyrimido[4',5':6,5]pyrano[3,2-*h*]quinolines **(4a,b-10a,b)** and Pyrido[2',3':6,5]pyrano[3,2-*h*]quinolines **(11a,b-13a,b)**

Compd. No.	Yield (%)	mp (°C)	Molecular Formula (Solvent of Recrystallization)	IR (υ, cm^{-1}) (KBr) and MS	NMR (δ, ppm) (Solvent)	*Anal.* Calcd/(Found) % C	H	N	S	Cl
7b	41	> 310	$C_{22}H_{17}N_4O_4ClS$ (ethanol)	3100 (NH), 3000 (CH arom.), 2920 (CH aliph.), 1680 (C=O), *1370* (SO_2 asym.), *1160* (SO_2 sym.)	(CF_3COOD): 3.25 (6H, s), 5.00 (1H, s), *7.00-8.30* (9H, m)	56.34 (56.41)	3.65 (3.71)	11.95 (11.89)	6.84 (6.79)	7.57 (7.62)
8a	61	272 dec.	$C_{24}H_{22}N_4O_4S$ (dioxane)	3000 (CH arom.), 2900 (CH aliph.), 2200 (CN), 1360 (SO_2 asym.), 1160 (SO_2 sym.); m/z 462	(DMSO-d_6): 1.80 (3H, t, J = 7.3 Hz), 3.25 (6H, s), 4.10 (2H, q, J = 7.3 Hz), 5.00 (1H, s), 7.00-8.60 (10H, m)	62.31 (62.26)	4.79 (4.81)	12.11 (12.05)	6.94 (6.91)	—
8b	58	274-276	$C_{24}H_{21}N_4O_4ClS$ (dioxane)	3015 (CH arom.), 2910 (CH aliph.), 2200 (CN), 1370 (SO_2 asym.), 1160 (SO_2 sym.)	(DMSO-d_6): 1.45 (3H, t, J = 7.0 Hz), *3.20 (6H, s)*, 4.40 (2H, q, J = 7.0 Hz), 5.00 (1H, s), 7.00-8.20 (9H, m)	58.00 (58.09)	4.26 (4.21)	11.28 (11.33)	6.46 (6.53)	7.14 (7.18)
9a	42	> 310	$C_{22}H_{20}N_6O_3S$ (ethanol)	3440-3340 (NH_2), 3150 (NH), 3000 (CH arom.), 2930 (CH aliph.), 1360 (SO_2 asym.), 1160 (SO_2 sym.); m/z 448	(CF_3COOD): 3.30 (6H, s), 5.00 (1H, s), 7.00-8.70 (10H, m)	58.91 (58.84)	4.50 (4.46)	10.74 (10.80)	7.16 (7.20)	—
9b	54	> 310	$C_{22}H_{19}N_6O_3ClS$ (ethanol)	3400-3300 (NH_2), 3150 (NH), 3000 (CH arom.), 2900 (CH aliph.), 1365 (SO_2 asym.), *1160* (SO_2 sym.)	(CF_3COOD): 3.40 (6H, s), 5.00 (1H, s), *7.00-8.50* (9H, m)	54.70 (54.61)	3.97 (4.01)	17.40 (17.33)	6.65 (6.71)	7.35 (7.41)
10a	51	277 dec.	$C_{28}H_{23}N_5O_3S_2$ (dioxane)	3350, 3170 (NH), 3010 (CH arom.), 2920 (CH aliph.), 1360 (SO_2 asym.), 1160 (SO_2 sym.); m/z 541	(CF_3COOD): 3.10 (6H, s), 5.00 (1H, s), 6.90-8.10 (14H, m)	62.08 (62.13)	4.28 (4.31)	12.93 (12.87)	11.85 (11.91)	—
10b	55	> 310	$C_{28}H_{22}N_5O_3ClS_2$ (dioxane)	3350, 3150 (NH), 3000 (CH arom.), 2900 (CH aliph.), 1360 (SO_2 asym.), 1160 (SO_2 sym.) m/z 576 (100%), 578 (35%)	(CF_3COOD): 3.20 (6H, s), 4.90 (1H, s), 7.00-8.30 (13H, m)	58.36 (58.30)	3.85 (3.90)	12.16 (12.12)	11.14 (11.18)	6.16 (6.21)
11a	61	268 dec.	$C_{24}H_{19}N_5O_4S$ (acetic acid)	*3400, 3300 (NH_2), 3160* (NH), 3020 (CH arom.), 2910 (CH aliph.), 2200 (CN), 1670 (C=O), m/z 473	(CF_3COOD): 3.10 (6H, s), 5.00 (1H, s), 7.10-8.20 (9H, m)	60.87 (60.93)	4.04 (4.00)	14.79 (12.82)	6.78 (6.83)	—

(Continued)

Table 1. ***(Continued)*** Physical Data of Pyrano[3,2-*h*]quinolines **(3a,b)**, Pyrimido[4',5':6,5]pyrano[3,2-*h*]quinolines **(4a,b-10a,b)** and Pyrido[2',3':6,5]pyrano[3,2-*h*]quinolines **(11a,b-13a,b)**

Compd. No.	Yield (%)	mp (°C)	Molecular Formula (Solvent of Recrystallization)	IR (υ, cm^{-1}) (KBr) and MS	NMR (δ, ppm) (Solvent)	*Anal.* Calcd/(Found) % C	H	N	S	Cl
11b	64	> 310	$C_{24}H_{18}N_5O_4ClS$ (acetic acid)	3400, 3300 (NH_2), 3150 (NH), 3020 (CH arom.), 2920 (CH aliph.), 2200 (CN), 1680 (C=O)	(CF_3COOD): 3.10 (6H, s), 5.10 (1H, s), 7.10-8.20 (8H, m)	56.74 (56.69)	3.57 (3.61)	13.79 (13.83)	6.32 (6.29)	6.99 (7.01)
12a	65	196-198	$C_{24}H_{18}N_5O_3ClS$ (ethanol)	3400, 3300 (NH_2), 3000 (CH arom.), 2910 (CH aliph.), 2200 (CN), m/z 492 (100%), 494 (35%)	($CDCl_3$): 3.00 (6H, s), 4.70 (2H, s), 5.00 (1H, s), 6.90-8.10 (9H, m)	58.58 (58.63)	3.69 (3.71)	14.24 (14.19)	6.52 (6.47)	7.22 (7.18)
12b	67	212-214	$C_{24}H_{17}N_5O_3Cl_2S$ (ethanol)	3400, 3300 (NH_2), 3000 (CH arom.), 2900 (CH aliph.), 2200 (CN)	($CDCl_3$): 3.10 (6H, s), 4.60 (2H, s), 5.00 (1H, s), 7.10-8.20 (8H, m)	54.74 (54.81)	3.26 (3.31)	13.30 (13.26)	6.10 (6.08)	13.49 (13.55)
13a	65	236 dec.	$C_{24}H_{19}N_5O_3S_2$ (acetic acid)	3400, 3300 (NH_2), 3030 (CH arom.), 2920 (CH aliph.), 2200 (CN), 1360 (SO_2 asym.), 1160 (SO_2 sym.); m/z 489	($CDCl_3$): 2.90 (6H, s), 4.40 (1H, s), 4.90 (2H, s), 5.10 (1H, s), 6.90-8.10 (9H, m)	58.87 (58.93)	3.91 (3.90)	14.31 (14.34)	13.11 (13.23)	—
13b	61	252 dec.	$C_{24}H_{18}N_5O_3ClS_2$ (acetic acid)	3400, 3300 (NH_2), 3000 (CH arom.), 2900 (CH aliph.), 2200 (CN), 1370 (SO_2 asym.), 1160 (SO_2 sym.)	(CF_3COOD): 3.10 (6H, s), 5.00 (1H, s), 6.90-8.10 (8H, m)	55.00 (55.15)	3.46 (3.42)	13.37 (13.34)	12.25 (12.23)	6.77 (6.76)

EXPERIMENTAL

The time required for completion of each reaction was monitored by TLC. Melting points are uncorrected. NMR (δ, ppm) spectra were measured on an EM-360 90-MHz spectrometer using TMS as internal standard. IR (υ,cm^{-1}) spectra were recorded on a Pye-Unicam SP 200-G spectrophotometer. Elemental analyses were carried out on a Perkin Elmer 240 C microanalyser. MS spectra were recorded on Jeol JMS 600 instrument.

2-Amino-4-aryl-3-cyano-6-dimethylaminosulfonyl-4*H*-pyrano[3,2-*h*]quinoline Derivatives **(3a,b)**

General Procedure:

A mixture of 5-dimethylaminosulfonyl-8-quinolinol **(1)** (2.52 g, 0.01 mol) and cinnamonitrile derivatives **(2a,b)** (0.01 mol) was heated under reflux in ethanol (50 mL) and catalytic amount of piperidine for 10 h. The solvent was evaporated under reduced pressure and the residue was cooled. The precipitated product was collected by filtration and recrystallized from ethanol (Table 1).

Synthesis of 7-Aryl-5-dimethylaminosulfonyl-10-methyl-8-oxo-8,9-dihydro-7*H*-pyrimido[4',5':6,5]pyrano-[3,2-*h*]quinoline Derivatives **(4a,b)**

General Procedure:

A solution of **3a,b** (0.01 mol) in acetic anhydride/pyridine mixture (15 mL, 2:1) was heated under reflux on a steam bath for 8 h, then allowed to cool and poured into ice cold water. The solid products were collected, washed several times with water and recrystallized from ethanol (Table 1).

2-Acetamido-3-cyano-6-dimethylaminosulfonyl-4*H*-pyrano[3,2-*h*]quinoline Derivatives **(5a,b)**

General Procedure:

Each compound **(3a,b)** (0.01 mol) was heated under reflux in acetic anhydride (20 mL, 0.21 mol) for 5 h. The products were collected, washed several times with water and recrystallized from dioxane (Table 1).

8-Amino-7-aryl-5-dimethylaminosulfonyl-7*H*-pyrimido[4',5':6,5]pyrano[3,2-*h*]quinoline Derivatives **(6a,b)**

General Procedure:

A mixture of **3a,b** (0.01 mol) and formamide (20 mL, 0.50 mol) was heated under reflux for 4 h. The reaction mixture was allowed to cool and the product was collected, washed several times with ethanol and recrystallized from ethanol (Table 1).

7-Aryl-5-dimethylaminosulfonyl-8-oxo-7*H*-pyrimido[4',5':6,5]pyrano[3,2-*h*]quinoline Derivatives **(7a,b)**

General Procedure:

A mixture of **3a,b** (0.01 mol) and formic acid (7 mL, 0.17 mol) in formamide (20 mL) was refluxed for 5 h. The reaction mixture was cooled, poured into ice cold water and the precipitate was collected and recrystallized from ethanol (Table 1).

4-Aryl-3-cyano-6-dimethylaminosulfonyl-2-ethoxymethylenamino-4*H*-pyrano[3,2-*h*]quinoline Derivatives **(8a,b)**

General Procedure:

A mixture of **3a,b** (0.01 mol) and triethyl orthoformate (3 mL, 0.018 mol) in acetic anhydride (20 mL) was refluxed for 3 h, and allowed to cool. The solid product was collected and recrystallized from dioxane (Table 1).

9-Amino-7-aryl-5-dimethylaminosulfonyl-8-imino-7,8,9-trihydropyrimido[4',5':6,5]pyrano[3,2-*h*]quinoline Derivatives **(9a,b)**

General Procedure:

To a suspension of compounds **(8a,b)** (0.003 mol) in benzene (50 mL), was added hydrazine hydrate (12 mL, 0.25 mol, 30%), the reaction mixture was stirred at rt for 7 h. The product was collected and recrystallized from ethanol (Table 1).

7-Aryl-5-dimethylaminosulfonyl-8-imino-10-thioxo-7,8,9,10,11-pentahydropyrimido[4',5':6,5]pyrano[3,2-*h*]quinoline Derivatives **(10a,b)**

General Procedure:

A mixture of **3a,b** (0.01 mol) and phenyl isothiocyanate (1.35 g, 0.01 mol) in dry pyridine (25 mL) was heated under reflux for 20 h, and allowed to cool. The reaction mixture was poured into cold water, whereby a solid product was separated and recrystallized from dioxane (Table 1).

Synthesis of 8-Amino-7-aryl-9-cyano-5-dimethylaminosulfonyl-10-oxo-pyrido[2',3':6,5]pyrano[3,2-*h*]-quinoline Derivatives **(11a,b)**

General Procedure:

A mixture of **3a,b** (0.01 mol) and ethyl cyanoacetate (1.13 g, 0.01 mol, 1 mL) was fused for 2 h. The solid product was collected and recrystallized from acetic acid (Table 1).

Synthesis of 8-Amino-7-aryl-10-chloro-9-cyano-5-dimethylaminosulfonylpyrido[2',3':6,5]pyrano[3,2-*h*]-quinoline Derivatives **(12,b)**

General Procedure:

A mixture of **11a,b** and phosphorus oxychloride (15 mL, 0.21 mol) was refluxed for 5 h, then the reaction mixture was allowed to cool and poured into ice cold water. The solid product was collected and recrystallized from ethanol (Table 1).

Synthesis of 8-Amino-7-aryl-9-cyano-5-dimethylaminosulfonylpyrido[2',3':6,5]pyrano[3,2-*h*]quinoline-10-thiol Derivatives **(13a,b)**

General Procedure (A):

A mixture of **11a,b** (0.01 mol) and phosphorus pentasulfide (4.4 g, 0.01 mol) in pyridine (30 mL) was refluxed for 7 h. The reaction mixture was allowed to cool, poured into ice cold water and the product was recrystallized from acetic acid (Table 1).

General Procedure (B):

A mixture of **11a,b** (0.01 mol) and thiourea (0.76 g, 0.01 mol) in ethanol (50 mL) was refluxed for 7 h, and

allowed to cool. The solid product was collected, dissolved in NaOH solution (10%, 10 mL) and then acidified with hydrochloric acid. The precipitate product was collected and recrystallized from acetic acid (Table 1).

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