Purines, pyrimidines, and related fused systems
19.* Use of S_N^H methodology for the synthesis of a new heterocyclic system, pyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine

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Sonogashira cross-coupling of 3-chloro-6,8-dimethylpyrimido[4,5-c]pyridazine-5,7(6*H*,8*H*)-dione with terminal alkynes afforded the corresponding 3-(alkyn-1-yl) derivatives. Oxidative amination of the latter compounds with primary alkylamines was accompanied by heterocyclization to give 6,8-dimethylpyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine-7,9(6*H*,8*H*)-diones.

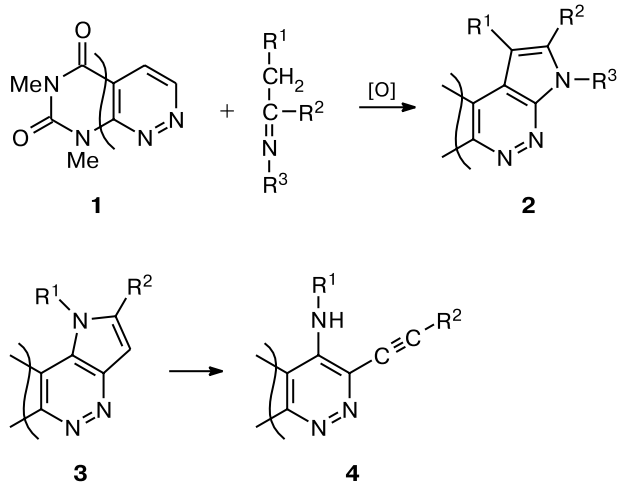
Key words: 3-chloro-6,8-dimethylpyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-dione, 1-alkynes, cross-coupling, Sonogashira reaction, catalysis, palladium complexes, 3-(alkyn-1-yl)-6,8-dimethylpyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-diones, oxidative amination, 6,8-dimethylpyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine-7,9(6*H*,8*H*)-diones.

Recently,[2] we have reported a new procedure for annelation of the pyrrole ring to neutral azines based on the tandem S_N^H—S_N^H process involving the formation of the C—C and C—N bonds between the π-deficient substrate and bifunctional *C*,*N*-nucleophile. In particular, the reaction of 6,8-dimethylpyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-dione (**1**) with aliphatic imines in the presence of an oxidant afforded 1-R^1-2-R^2-3-R^3-6,8-dimethylpyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine-7,9(6*H*,8*H*)-diones (**2**) (Scheme 1). In the present study, we applied the S_N^H methodology[3] to the synthesis of derivatives of a previously unknown isomeric heterocyclic system, *viz.*, pyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine **3**.

One would expect that 3-alkynyl-4-aminopyrimidopyridazines **4** would serve as direct precursors of compounds **3** since it is known that such compounds undergo cyclization to form the pyrrole ring under the action of bases[4,5] and other catalysts.[6—8] We chose 3-chloro-6,8-dimethylpyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-dione (**5**) as the starting compound for the synthesis of pyridazines **4**.[9] Then we intended to use successively Sonogashira cross-coupling[10] and oxidative amination because the Cl atom in compound **5** can be replaced with a CH-acid residue,[11] whereas pyrimidopyridazinedione **1** is readily aminated at the C(4) atom in an RNH_2—oxidant system.[12]

We found that chloropyridazine **5** (Scheme 2) reacted with terminal alkynes **7a**—**e** in DMF under argon in the presence of palladium complexes and catalytic amounts of CuI and K_2CO_3 to give 3-(alkyn-1-yl) derivatives **8a**—**e** in 22—74% yields. The reaction of compound **5** with volatile trimethylsilylacetylene (**7f**) was carried out in a sealed tube in the absence of a solvent with the use of the Pd_2dba_3—PPh_3 system (dba is dibenzylideneacetone as the ligand) as the catalyst and Et_3N as the base.

Scheme 1



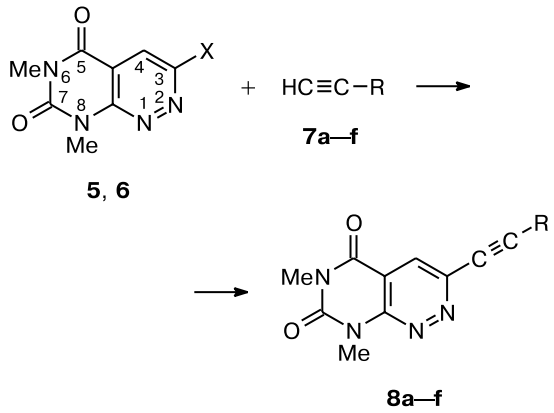
It is known that additives of CuI make it possible to perform such transformations at room temperature.[13,14] However, the reaction of compound **5** with alkynes proceeds at a noticeable rate only at 90—100 °C. Apparently, the reason is that the Cl atom in molecule **5** is insuffi-

* For Part 18, see Ref. 1.

Published in Russian in *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 2, pp. 422—427, February, 2003.

1066-5285/03/5202-441 $25.00

Scheme 2



X = Cl (**5**), OTf (**6**)
7, **8:** R = Ph (**a**), 4-ClC_6H_4 (**b**), CH_2OH (**c**), CH_2OTHP (**d**), *n*-C_6H_{13} (**e**), TMS (**f**)

Reagents and conditions: $Pd(PPh_3)_4$ or Pd_2dba_3—PPh_3, K_2CO_3, CuI, DMF, 90—100 °C, Ar.

Scheme 3

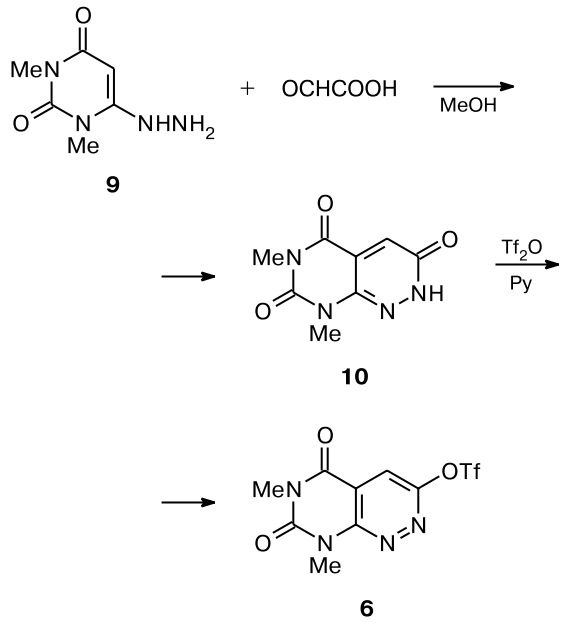


Table 1. Characteristics of 3-(alkyn-1-yl)-6,8-dimethylpyrimido[4,5-*c*]pyridazine-5,7(6*H*, 8*H*)-diones **8a—f**

Compound	R	Synthesis method	R_f	τ^a /h	Yield (%)	M.p. /°C	^{1}H NMR ($CDCl_3$), δ (*J*/Hz)			IR, ν/cm^{-1}			UV, λ_{max}/nm (logε)
							N—Me	H(4)	R	Ring	C=O	C≡C	
8a	Ph	***A***, ***B***, ***C***	0.6	1	74—80	175—176	3.49, 3.90	8.22	7.39 (m, 3 H, Ph); 7.62 (m, 2 H, Ph)	1558, 1585, 1599	1666, 1787	2218	295 (4.46); 357 (3.47)
8b	4-ClC_6H_4	***A***	0.6	5	22	317—319	3.40,[b] 3.79	7.96	7.67 (d, 2 H, C_6H_4, *J* = 8.7); 8.19 (d, 2 H, C_6H_4, *J* = 8.6)	1573, 1586, 1600	1660, 1713	2270	365 (4.45)
8c	CH_2OH	***A***, ***C***	0.1	8	32—38	203—205	3.48, 3.88	8.13	4.58 (s, 2 H, CH_2)	1554, 1594, 1648	1675, 1728	2238	276 (4.30); 349 (3.53)
8d	CH_2OTHP	***B***	0.5	2	46	103—105	3.48, 3.89	8.14	1.50—2.95 (m, 8 H, THP); 4.56 (d, 2 H, CH_2, *J* = 1.9); 4.90 (m, 1 H, THP)	1558, 1598	1665, 1718	2250	276 (4.35); 350 (3.54)
8e	C_6H_{13}	***B***	0.65	1	47	76—77	3.47, 3.87	8.06	0.89 (t, 3 H, Me, *J* = 6.7); 1.28—1.38 (m, 4 H, $(CH_2)_3(\underline{CH_2})_2Me$); 1.41—1.52 (m, 2 H, $(CH_2)_2\underline{CH_2}C_3H_7$); 1.60—1.72 (m, 2 H, $CH_2\underline{CH_2}C_4H_9$); 2.49 (t, 2 H, $\underline{CH_2}C_5H_{11}$, *J* = 7.0)	1572, 1612	1679, 1732	2237	269 (4.29); 355 (3.44)
8f	TMS	—	0.65	1	71	120—121	3.47, 3.87	8.21	0.28 (s, 9 H, $SiMe_3$)	1572, 1612	1679, 1732	2184	281 (4.37); 347 (3.64)

[a] Reaction time.
[b] DMSO-d_6 was used as the solvent.

ciently mobile because of the electron-releasing effect of the N atom at position 8. Product **8b** was obtained in low yield due, probably, to its further cross-coupling with the starting arylacetylene.

The use of aryl iodides or aryl triflates as the electrophilic component in the catalytic cross-coupling gave the best results.[15,16] Hence, we prepared triflate **6** by condensation of hydrazinouracil **9** with glyoxylic acid followed by acylation of pyrimidopyridazinetrione **10** with trifluoromethanesulfonic anhydride (Scheme 3). However, cross-coupling of compound **6** with phenylacetylene and propargyl alcohol under the above-mentioned conditions afforded the products in only slightly higher yields (80 and 38%, respectively, compared to 74 and 32% obtained in the reactions with compound **5**).

The structures of substituted acetylenes **8** were confirmed by UV, IR, and ^{1}H NMR spectroscopy (Table 1). Compounds **8** are virtually colorless (λ_{max} 347—365 nm). Their IR spectra have a band at 2180—2270 cm^{-1} ($\nu_{C\equiv C}$). The ^{1}H NMR spectra show singlets of two NMe groups at δ 3.5 and 3.9 and signals for H(4) at δ 8.0—8.2.

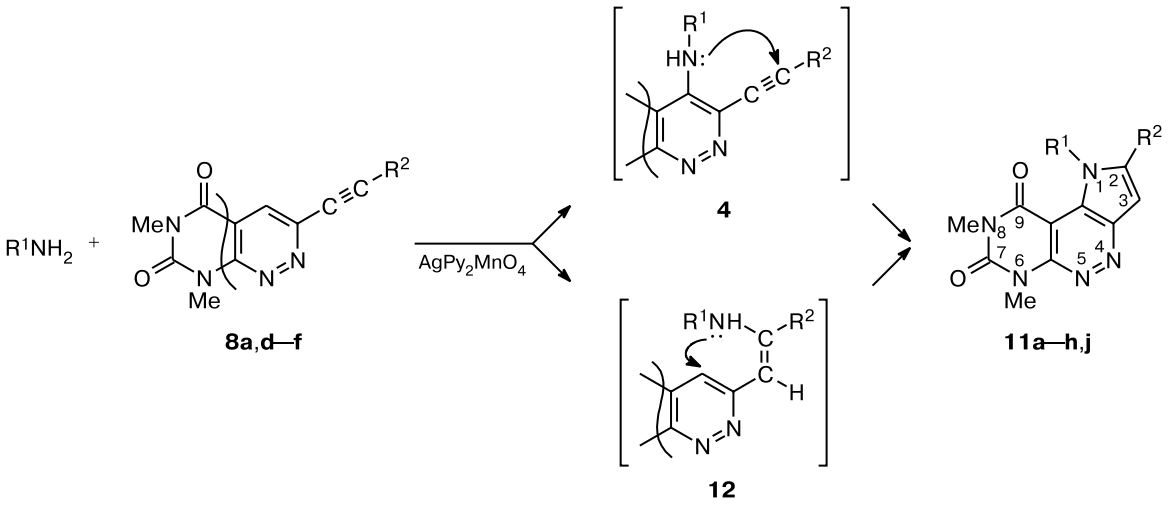
Oxidative amination of 3-(alkyn-1-yl)pyrimidopyridazinediones **8a**,**d**—**f** with primary alkylamines in the presence of $AgPy_2MnO_4$ at 0—20 °C afforded 1-R^1-2-R^2-6,8-dimethylpyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine-7,9(6*H*,8*H*)-diones (**11a**—**i**) in 22—85% yields (Scheme 4). Treatment of trimethylsilylethynyl derivative **8f** with propylamine and an oxidant gave not the expected pyrrole **11i** but the product of its desilylation **11j** in 22% yield. An attempt to prepare pyrrole **11k** by the reaction of compound **8f** with KNH_2 in liquid ammonia and $KMnO_4$ at the temperature from −60 to −55 °C failed (the starting compound remained unconsumed). Compounds **8a**,**d** also did not react with KNH_2 under these conditions.

The physicochemical characteristics of pale-yellow compounds **11a**—**h**,**j** (λ_{max} 365—372 nm) are given in Table 2. Their IR spectra have absorption bands at 1650—1720 cm^{-1} ($\nu_{C=O}$). The ^{1}H NMR spectra of these compounds show a signal for the H(3) proton of the pyrrole ring at δ 6.81—7.06, which is characteristic of fused pyrroles.[17] It should be noted that the protons of the α-CH$_2$ and α-CH groups of the substituent at the N(1) atom in compounds **11** are deshielded by the C(9)=O group and are observed at low field (δ 4.73—5.55), whereas the protons of the terminal Me group of pyrroles **11b**,**d** are, apparently, shielded by the benzene ring (δ 0.50—0.59).

It is reasonable to assume that the transformation **8** → **11** occurs through spontaneous heterocyclization of intermediate *o*-aminohetarylacetylenes **4**. An analogous tandem process involving S_N^H-amination followed by intramolecular cyclization under the conditions of the classical Chichibabin reaction has been described earlier for 3-ethynylpyridine.[18] However, we do not rule out that the reaction proceeds through enamine intermediate **12**, because an attempt to perform oxidative amination of hetarylacetylene **8a** with piperidine (Scheme 5) led to the formation of ketone **15** in 20% yield instead of the expected amino derivative **13**. The formation of ketone **15** can be explained only as a result of hydrolysis of enamine **14**. The ability of acetylenes to add amines at the triple bond giving rise to enamines is well known.[19]

To summarize, we prepared a series of derivatives of the new heterocyclic system, *viz.*, pyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine **11**. Evidently, hetarylacetylenes **8**

Scheme 4



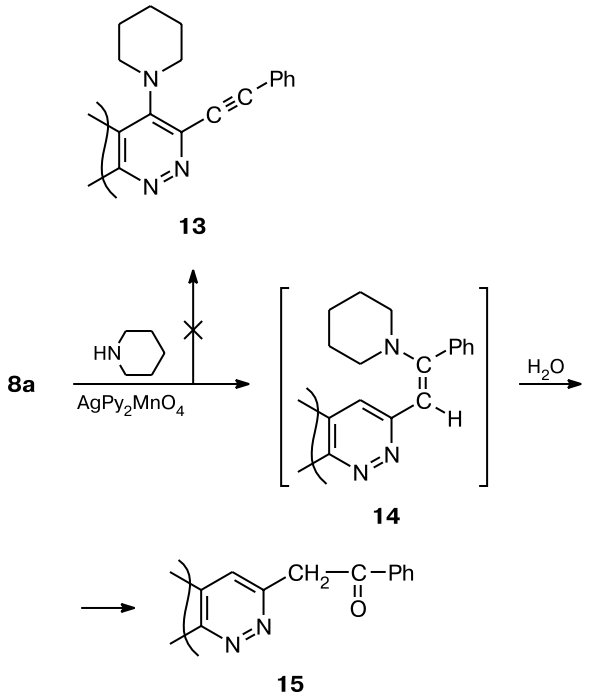
11:	a	b	c	d	e	f	g	h	i	j	k
R^1	Et	Pr	Pr^i	Bu	*cyclo*-C$_6$H$_{11}$	Pr	Pr	Bu	Pr	Pr	H
R^2	Ph	Ph	Ph	Ph	Ph	CH$_2$OTHP	*n*-C$_6$H$_{13}$	*n*-C$_6$H$_{13}$	TMS	H	H

Table 2. Characteristics of 1,2-disubstituted 6,8-dimethylpyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine-7,9(6*H*,8*H*)-diones **11a—h,j**

Compound	R^1	R^2	R_f	Yield (%)	M.p. /°C	^{1}H NMR ($CDCl_3$), δ (*J*/Hz)				IR, ν/cm^{-1}		UV, λ_{max}/nm (logε)
						N—Me	H(3)	R^1	R^2	Ring	C=O	
								NCH_2(NCH)	Other			
11a	Et	Ph	0.45	62	214—216	3.53 3.98	7.05	4.86 (q, 2 H, *J* = 7.0)	0.98 (t, 3 H, Me, *J* = 7.0)	7.52 (s, 5 H, Ph)	1526, 1573, 1633	1673, 1700,	266 (4.50), 325 (3.83), 372 (3.63)
11b	Pr	Ph	0.50	61	175—178	3.53 3.99	7.05	4.79 (t, 2 H, *J* = 7.2)	0.50 (t, 3 H, Me, *J* = 7.4); 1.37 (m, 2 H, $CH_2\underline{CH_2}Me$)	7.52 (s, 5 H, Ph)	1545, 1572, 1599	1652, 1705,	267 (4.53), 327 (3.85), 373 (3.65)
11c	Pr^i	Ph	0.43	41	221—222	3.53 3.97	6.97	5.55 (sept, 1 H, *J* = 7.0)	1.32 (d, 6 H, $CH\underline{Me}_2$, *J* = 7.0)	7.46—7.48 (m, 3 H, Ph); 7.55—7.58 (m, 2 H, Ph)	1545, 1585, 1612	1665, 1718,	266 (4.50), 325 (3.80), 365 (3.58)
11d	Bu	Ph	0.52	85	145—146	3.53 3.98	7.04	4.82 (t, 2 H, *J* = 7.2)	0.59 (t, 3 H, Me, *J* = 7.2); 0.80—0.95 (m, 2 H, $(CH_2)_2\underline{CH_2}Me$); 1.23—1.35 (m, 2 H, $CH_2\underline{CH_2}Et$)	7.52 (s, 5 H, Ph)	1532, 1572, 1599	1652, 1705	267 (4.51), 328 (3.85), 372 (3.64)
11e	*cyclo*-C_6H_{11}	Ph	0.40	47	207—209	3.54 3.98	6.96	5.04 (m, 1 H)	0.80—2.00 (m, 10 H, *cyclo*-C_6H_{11})	7.44—7.49 (m, 3 H, Ph); 7.53—7.56 (m, 2 H, Ph)	1545, 1572	1665, 1718	267 (4.51), 327 (3.83)
11f	Pr	CH_2OTHP	0.35	23	122—123	3.56 3.95	7.06	0.90 (t, 3 H, Me, *J* = 7.3); 1.50—2.00 (m, 8 H, $CH_2\underline{CH_2}Me$ and THP); 3.65—3.89 (m, 2 H, OCH_2 THP); 4.66—4.96 (m, 5 H, $\underline{CH_2}Et$, OCH THP and C(2)CH_2)			1573	1660, 1700	325 (3.78)
11g	Pr	C_6H_{13}	0.40	59	80—81	3.51 3.94	6.81	4.73 (t, 2 H, *J* = 7.2)	0.90 (t, 3 H, Me, *J* = 7.2); 1.75—1.80 (m, 2 H, $CH_2\underline{CH_2}Me$)	0.88 (t, 3 H, Me, *J* = 7.2); 1.20—1.70 (m, 8 H, $CH_2(\underline{CH_2})_4Me$); 2.76 (t, 3 H, $\underline{CH_2}(CH_2)_4Me$, *J* = 7.6)	1545, 1572, 1585	1665, 1705,	324 (3.75), 366 (3.48)
11h	Bu	*n*-C_6H_{13}	0.40	42	85—86	3.50 3.93	6.81	4.77 (t, 2 H, *J* = 7.6)	0.85—0.95 (m, 6 H, $(CH_2)_3\underline{Me}$ and $(CH_2)_5\underline{Me}$); 1.27—1.37 (m, 6 H, $(CH_2)_2\underline{CH_2}Me$ and $(CH_2)_3(\underline{CH_2})_2Me$); 1.40—1.58 (m, 4 H, $CH_2\underline{CH_2}Et$ and $(CH_2)_2\underline{CH_2}Pr$); 1.80 (m, 2 H, $CH_2\underline{CH_2}Bu$); 2.75 (t, 2 H, $\underline{CH_2}C_5H_{11}$, *J* = 7.6)		1585, 1599, 1619	1678, 1718	324 (3.77), 68 (3.50), 464 (2.68)
11j	Pr	H	0.20	22	139—140	3.51 3.96	7.04 (d, 1 H, *J* = 3.5); 7.39 [a] (d, 1 H, *J* = 3.5)	4.78 (t, 2 H, *J* = 7.2)	0.73 (t, 3 H, Me, *J* = 7.4); 1.79 (m, 2 H, $CH_2\underline{CH_2}Me$)	—	1545, 1599	1665, 1705	322 (3.73), 362 (3.49)

[a] The chemical shift of H(2).

Scheme 5



can also be used in the synthesis of other fused polynuclear systems based on pyrimidopyridazinedione **1**.

Experimental

The IR spectra were recorded on a Specord IR-71 instrument in Nujol mulls. The ^{1}H NMR spectra were measured on Bruker-250 (250 MHz) and Unity-300 (300 MHz) spectrometers with Me_4Si as the internal standard. The UV spectra were recorded on a Specord M-40 instrument in $CHCl_3$. Chromatography was carried out on Al_2O_3 (Brockmann activity III—IV) using $CHCl_3$ as the eluent; visualization was carried out with iodine vapor. The melting points were measured in glass tubes on a PTP instrument and were not corrected.

The physicochemical characteristics of the resulting compounds are given in Tables 1 and 2. The results of elemental analysis are presented in Table 3.

3-(Alkyn-1-yl)-6,8-dimethylpyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-diones (8a—e) (general procedure). ***A.*** A mixture of compound **5** (1 mmol), alkyne **7** (1.5 mmol), K_2CO_3 (1.5 mmol), $Pd(PPh_3)_4$ (0.02 mmol), and CuI (0.05 mmol) in anhydrous DMF (3 mL) was stirred at 90—100 °C under argon (reaction times are given in Table 1) and then concentrated to dryness. The residue was extracted with $CHCl_3$. The extract was concentrated to ~5 mL and chromatographed on a column with Al_2O_3 using $CHCl_3$ as the eluent. The pale-yellow fraction was collected (R_f are given in Table 1). The product was recrystallized from MeOH (compound **8b** was recrystallized from DMSO). Compounds **8** were obtained as colorless crystals.

B. A mixture of compound **5** (1 mmol), alkyne **7** (1.5 mmol), K_2CO_3 (1.5 mmol), Pd_2dba_3 (0.02 mmol), PPh_3 (0.16 mmol), and CuI (0.05 mmol) in anhydrous DMF (3 mL) was stirred at 90—100 °C under argon (reaction times are given in Table 1) and then treated as described in the method ***A***.

C. A mixture of compound **6** (0.5 mmol), alkyne **7** (1 mmol), K_2CO_3 (1 mmol), $Pd(PPh_3)_4$ (0.01 mmol), and CuI (0.01 mmol) in anhydrous DMF (3 mL) was stirred at 90—100 °C under argon (reaction times are given in Table 1) and then treated as described in the method ***A***.

6,8-Dimethyl-3-(trimethylsilylethynyl)pyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-dione (8f). A mixture of compound **5** (227 mg, 1 mmol), trimethylsilylacetylene (0.2 mL, 1.2 mmol), Pd_2dba_3 (20 mg, 0.02 mmol), CuI (10 mg, 0.05 mmol), PPh_3 (42 mg, 0.16 mmol), and Et_3N (5 mL) was heated in a sealed tube under argon at 100 °C for 1 h and then treated as described above for compounds **8a—e**.

6,8-Dimethylpyrimido[4,5-*c*]pyridazine-3,5,7(2*H*,6*H*,8*H*)-trione (10). A mixture of compound **9** (138 mg, 1 mmol) and glyoxylic acid (111 mg, 1 mmol) in MeOH (5 mL) was refluxed for 2 h and concentrated to dryness. The residue was extracted with $CHCl_3$. The extract was concentrated to ~5 mL and chromatographed on a column with Al_2O_3 using a $CHCl_3$—MeOH mixture (20 : 1) as the eluent. The colorless

Table 3. Results of elemental analysis of compounds **8a—f** and **11a—h,j**

Compound	Molecular formula	Found / Calculated (%)		
		C	H	N
8a	$C_{16}H_{12}N_4O_2$	65.90	4.05	18.97
		65.75	4.11	19.18
8b	$C_{16}H_{11}N_4O_2Cl$	58.78	3.10	17.31
		58.81	3.37	17.15
8c	$C_{11}H_{10}N_4O_3$	53.90	4.01	22.58
		53.65	4.07	22.76
8d	$C_{16}H_{18}N_4O_4$	58.35	5.70	17.04
		58.18	5.45	16.97
8e	$C_{16}H_{20}N_4O_2$	64.23	6.70	18.54
		64.00	6.67	18.67
8f	$C_{13}H_{16}N_4O_2Si$	54.30	5.70	19.24
		54.17	5.56	19.44
11a	$C_{18}H_{17}N_5O_2$	64.54	5.09	20.76
		64.48	5.07	20.90
11b	$C_{19}H_{19}N_5O_2$	65.29	5.42	20.30
		65.33	5.44	20.06
11c	$C_{19}H_{19}N_5O_2$	65.50	5.27	20.17
		65.33	5.44	20.06
11d	$C_{20}H_{21}N_5O_2$	66.34	5.57	19.17
		66.12	5.79	19.28
11e	$C_{22}H_{23}N_5O_2$	67.83	5.88	17.96
		67.87	5.91	17.99
11f	$C_{19}H_{25}N_5O_4$	58.85	6.25	18.13
		58.91	6.46	18.09
11g	$C_{19}H_{27}N_5O_2$	63.60	7.45	19.73
		63.87	7.56	19.61
11h	$C_{20}H_{29}N_5O_2$	64.81	7.55	18.95
		64.69	7.82	18.87
11j	$C_{13}H_{15}N_5O_2$	57.05	5.61	25.44
		57.14	5.49	25.64

fraction with R_f 0.5 was collected and concentrated to obtain compound **10** as colorless crystals in a yield of 112 mg (53%), m.p. 238—240 °C (from MeCN). IR, ν/cm^{-1}: 3300—3600 (NH); 1782, 1728, 1662 (C=O). ^{1}H NMR (DMSO-d_6), δ: 3.20 (s, 3 H, N(8)Me); 3.51 (s, 3 H, N(6)Me); 7.41 (s, 1 H, H(4)); 13.2 (br.s, disappeared after deuteration, 1 H, NH). Found (%): C, 46.32; H, 3.59; N, 27.12. $C_8H_8N_4O_3$. Calculated (%): C, 46.15; H, 3.85; N, 26.92.

6,8-Dimethyl-3-(trifluoromethylsulfonyl)pyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-dione (6). Trifluoromethanesulfonic anhydride (0.2 mL, 1.2 mmol) was added portionwise to a solution of compound **10** (207 mg, 1 mmol) in pyridine (3 mL) cooled to 0 °C. The reaction mixture was stirred at 0 °C for 1 h and then at 20 °C for 48 h. Then the reaction mixture was diluted with water (3 mL) and the precipitate that formed was filtered off. Compound **6** was obtained as colorless crystals in a yield of 210 mg (68%), m.p. 159—160 °C (from Pr^{i}OH). IR, ν/cm^{-1}: 3106 (C—H arom.); 1772, 1720 (C=O). ^{1}H NMR ($CDCl_3$), δ: 3.50 (s, 3 H, N(8)Me); 3.89 (s, 3 H, N(6)Me); 8.06 (s, 1 H, H(4)). Found (%): C, 32.00; H, 2.06; N, 16.34. $C_9H_7F_3N_4O_5S$. Calculated (%): C, 31.76; H, 2.05; F, 16.76; N, 16.47; S, 9.41.

1,2-Disubstituted 6,8-dimethylpyrrolo[3′,2′:3,4]pyrimido[4,5-*c*]pyridazine-7,9(6*H*,8*H*)-diones (11a—h,j) (general procedure). A solution of compound **8** (1 mmol) in amine (30—40 mL) was stirred at 20 °C for 15 min (in the case of $EtNH_2$, at 0 °C) and then $AgPy_2MnO_4$ (1 mmol) was added. The completion of the reaction was determined chromatographically. The reaction mixture was concentrated to dryness and the residue was extracted with $CHCl_3$. The extract was concentrated to ~5 mL and chromatographed on a column with Al_2O_3 using $CHCl_3$ as the eluent. The first yellow fraction was collected and concentrated. The residue was recrystallized from Pr^{i}OH and compound **11** was obtained as yellow crystals.

3-Benzoylmethyl-6,8-dimethylpyrimido[4,5-*c*]pyridazine-5,7(6*H*,8*H*)-dione (15). A solution of compound **8a** (292 mg, 1 mmol) in piperidine (25 mL) was stirred at 20 °C for 15 min and then $AgPy_2MnO_4$ (385 mg, 1 mmol) was added. The reaction mixture was kept at 20 °C for one week and concentrated to dryness. The residue was extracted with $CHCl_3$. The extract was concentrated to ~5 mL and chromatographed on a column with Al_2O_3 using $CHCl_3$ as the eluent. The yellow fraction was collected and concentrated to obtain compound **15** as dark-yellow crystals in a yield of 62 mg (20%), m.p. 236—238 °C (from Pr^{i}OH). IR, ν/cm^{-1}: 3062 (C—H arom.); 1718, 1692, 1678 (C=O). ^{1}H NMR (DMSO-d_6), δ: 3.36 (s, 3 H, N(8)Me); 3.73 (s, 3 H, N(6)Me); 4.89 (s, 2 H, CH_2); 7.54—8.10 (m, 5 H, Ph); 8.16 (s, 1 H, H(4)). Found (%): C, 62.10; H, 4.36; N, 18.15. $C_{16}H_{14}N_4O_3$. Calculated (%): C, 61.93; H, 4.52; N, 18.06.

This study was financially supported by the Russian Foundation for Basic Research (Project No. 01-03-32338).

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Received April 18, 2002;
in revised form September 11, 2002