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BENZOXAZOLYL- AND BENZOTHIAZOLYL-GUANIDINES IN THREE-COMPONENT REACTIONS

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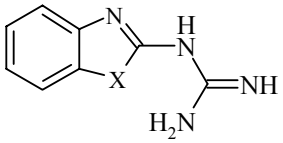
The previously unknown three component condensation of hetarylguanidines, orthoesters, and active methylene carbonyl compounds has been studied. Depending on the nature of the latter products of both linear combination and subsequent cyclizationl leading to pyrimidine or tetrahydroquinazoline structures were obtained.

Keywords: active methylene carbonyl compounds, benzoxa(thia)zoylguanidines, orthoesters, condensation, three component heterocyclization.

Three component condensations with the participation of C–H acids, aldehydes or orthoesters, and N-containing mono- or binucleophiles lead to a variety of derivatives, which possess a wide spectrum of biological activity [1]. For example, interaction of aromatic aldehydes, (thio)urea, and β-diketones in Biginelli conditions gave dihydro(thia)pyrimidones, which are calcium channel activators, antagonists of adrenoreceptors, etc. [2-4].

At present there are no reports connected to the use in such reactions of guanidines, despite the great synthetic potential of the latter [5].

In this work the three component condensation of benzoxa(thia)zolyl-2-guanidines **1a,b** with orthoesters $(RC)OEt_3$ **2a,b** (**a** R = H, **b** R = Me) and active methylene compounds containing a carbonyl function has been studied. In this case one can expect formation of products of linear combination [6] and products of subsequent cyclization.



1 a,b

1 a X = O, **b** X = S

It has been established that boiling an equimolar mixture of guanidines **1a,b** with linear β-diketones **3a-c** and an excess of the corresponding orthoester **2** for 20-40 min gave the acylpyrimidine **5**. Cyclization, evidently occurs *via* formation of a linear intermediate **4** with subsequent intramolecular condensation.

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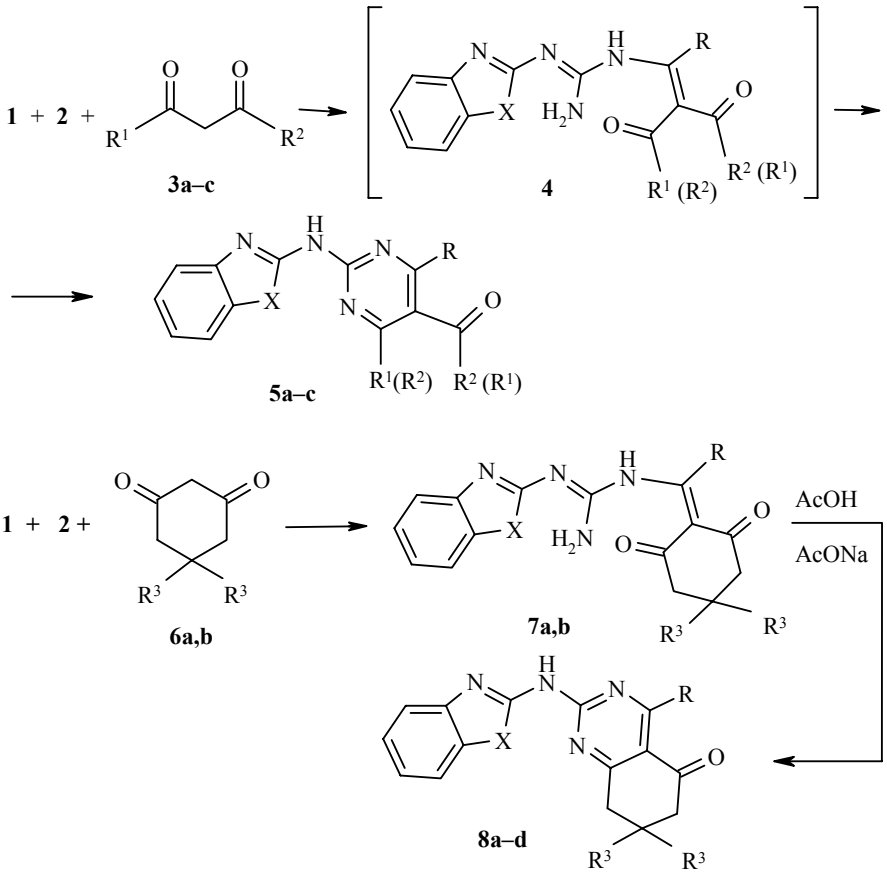


5a, c, 7a, 8a,c,d R = H; **5b, 7b, 8b** R = Me; **3a, 5a,b** $R^1 = R^2$ = Me; **3b, 5c** $R^1 = R^2$ = Ph; **3c** R^1 = Me, R^2 = Ph; **6a, 8c** R^3 = H; **6b, 7a,b, 8a,b,d** R^3 = Me; **5a,b, 7a,b, 8a,b** X = O; **5c, 8c,d** X = S

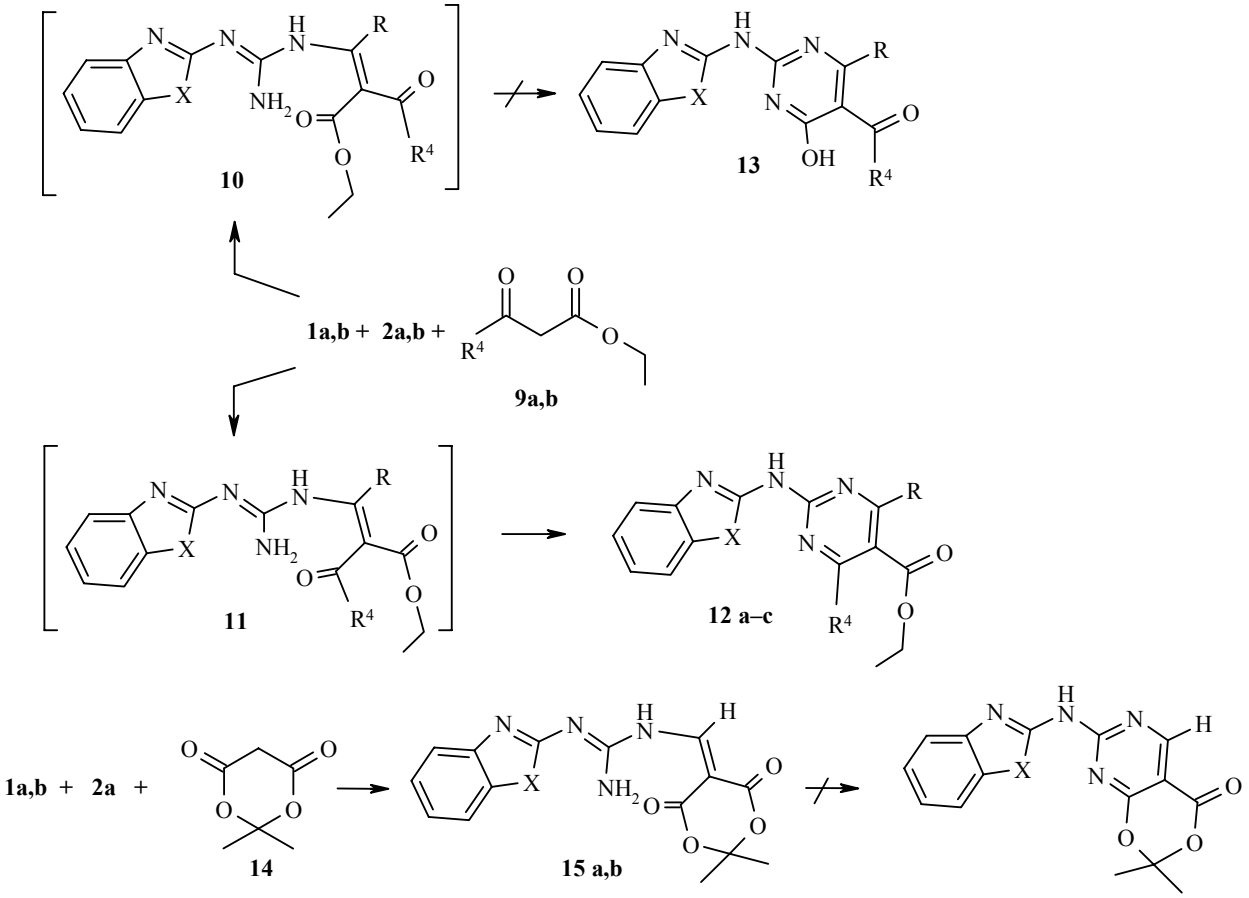
Use of symmetrical diketones, such as acetylacetone **3a** and dibenzoylmethane **3b**, gives a single product of cyclization, whereas in the case of the unsymmetrical diketone **3c** the reaction mixture, according to TLC, contained both possible regioisomers with similar values of R_f in comparable amounts, which were not separable. This may be explained by the equally probable formation of the *Z*- and *E*-intermediates.

For cyclic ketones of the type **6a,b** the cyclization step is linked to considerable steric hindrance, consequently for the less reactive benzoxazoylguanidine the intermediate compounds **7a,b** were isolated in pure form and their cyclization into the tetrahydroquinazolones **8a, b** was carried out by heating them in acetic acid with sodium acetate. The more reactive benzthiazoylguanidines formed the cyclic products **8c,d** directly.

Reactions of the guanidines **1** and the orthoesters **2** with the β-keto esters **9a,b**, with the possible formation of two intermediates **10** and **11**, gave a single product of cyclization, the ethoxycarbonylpyrimidine **12**, with no formation of the hydroxyacylpyrimidine **13**. The presence of the ester group in compounds **12a-c** was confirmed by the doublet and triplet of the ethoxy group n their ^{1}H NMR spectra (Scheme 1).

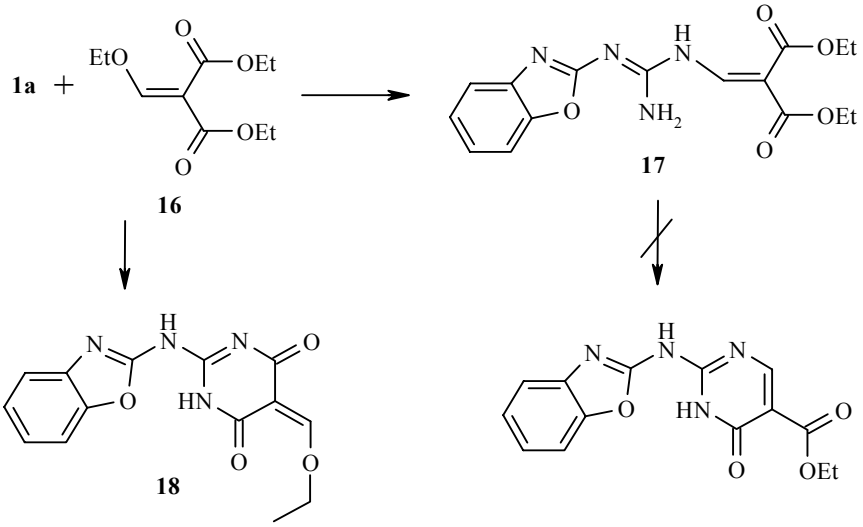
Evidently, the intermediate with close amino- and carbonyl groups (but not ester groups) underwent intramolecular cyclization. In precisely the same way from the reaction of guanidines **1a,b** and orthoesters with the Meldrum acid **14** only linear products of condensation **15a,b** were obtained. Moreover, as a result of the reaction of guanidine **1a** with ethoxymethylenemalonic ester (**16**) in boiling xylene the product of linear condensation was isolated – diethyl 2-amino(1,3-benzoxazol-2-ylimino)methylaminomethylenemalonate (**17**) – which did not undergo further cyclization. The by-product 2-(1,3-benzoxazo-2-ylamino)-5-[1-ethoxymethylidene]-1,4,5,6-tetrahydro-4,6-pyrimidinedione (**18**) was isolated in this case (Scheme 2).

Scheme 1



9a, 12a,c R^4 = Me; **9b,12b** R^4 = Ph; **12a,c, 15a,b** R = H, **12b** R = Me; **12a,b,15a** X = O; **12c, 15b** X = S

Scheme 2



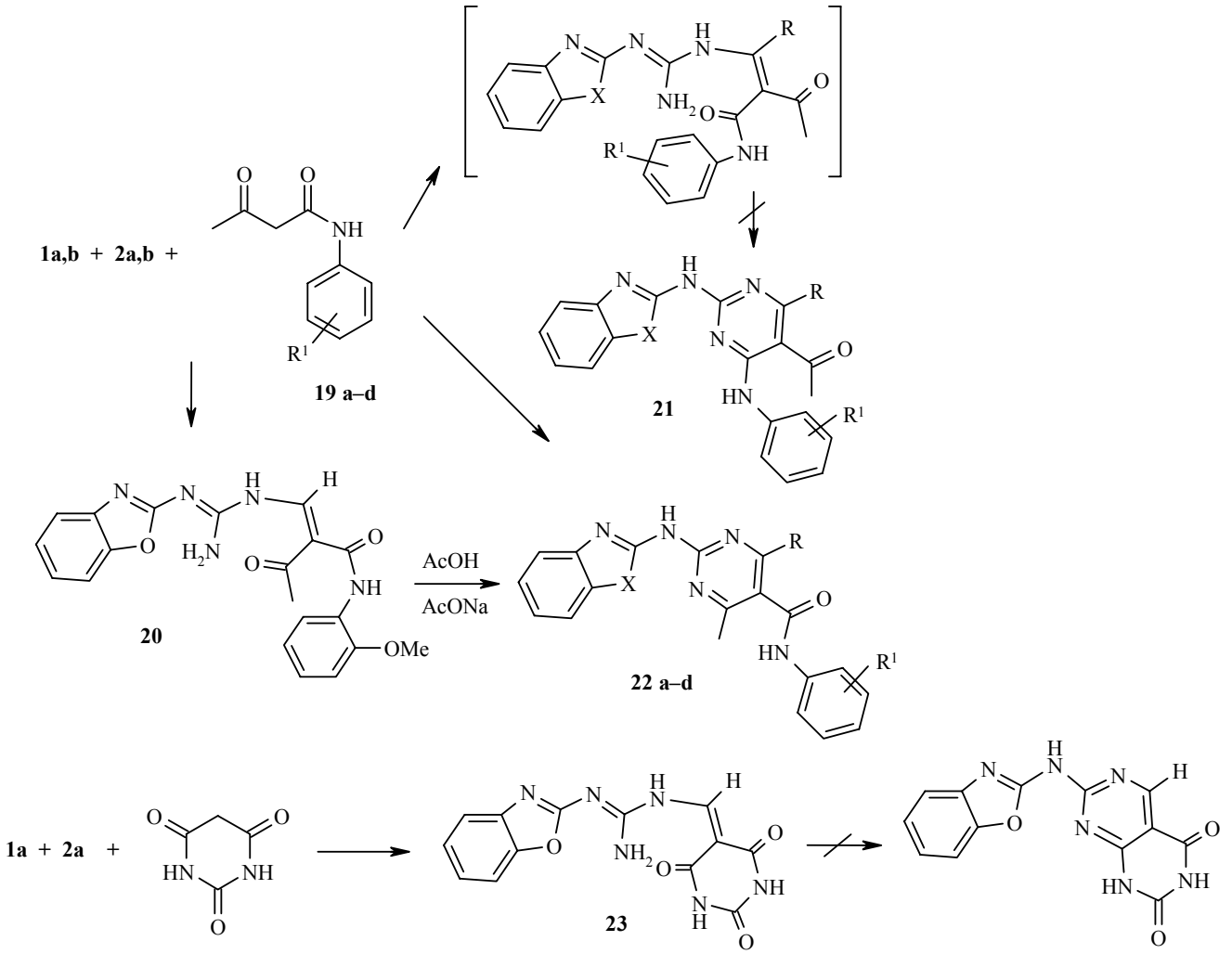
That cyclization of only one of the two possible intermediates under the studied reaction conditions is explained by the greater reactivity of the ketone carbonyl group in comparison with the ester group in reactions with nucleophilic reagents [7,8].

On boiling a solution of guanidine **1a** and acetylacetanilide **19a** in excess of ester **2a** for a relatively short time (40 min) the linear condensation product **20** was first obtained, further cyclization of which (boiling in acetic acid with sodium acetate) also occurred exclusively at the ketone carbonyl group of the anilide **22a**. More prolonged boiling (1-1.5 h) of components **1a,b** and **19a-d** in excess of the esters **2a,b** gave the final products of condensation **22a-d** directly.

An analogous reaction of components **1a** and **2a** with barbituric acid gave formation of the "linear" compound **23** only.

Structures of compounds **22a-d** were confirmed by the presence in their ^{1}H NMR spectra of weak field singlets of amide (but not arylamines) N–H protons in the range from 9.8 (**22d**) to 11.8 pm (**22a**).

The ^{1}H NMR spectra of compounds **5, 7, 8, 13, 15, 17, 18, 20a, 22,** and **23** are given in Table 1, and yields and characteristics in Table 2.



19, 22 a R^1 = 2-MeO, **b** R^1 = 4-Me, **c** R^1 = 3-CF_3, **d** R^1 = 2-Me;
22a,b R = H; **c,d** R = Me; **a-c** X = O, **d** X = S

TABLE 1. ^{1}H NMR Spectra of the Compounds Synthesized

Com-pound	Chemical shifts, δ, ppm (*J*, Hz)
5a	2.55 (3H, s, CH_3); 2.67 (3H, s, CH_3); 7.28 (2H, m, arom.); 7.58 (2H, m, arom.); 9.11 (1H, s, C-6 pyrimidine); 11.71 (1H, br. s, NH)
5b	2.38 (6H, s, 2 CH_3); 2.56 (3H, s, CH_3); 7.26 (2H, m, arom.); 7.56 (2H, m, arom.); 11.41 (1H, br. s, NH)
5c	7.21-7.95 (14H, m, arom.); 8.79 (1H, s, C-6 pyrimidine); 12.45 (1H, br. s, NH)
7a	1.08 (6H, s, 2 CH_3); 2.40 (2H, s, CH_2); 2.50 (2H, s, CH_2); 7.20 (2H, m, arom.); 7.52 (2H, m, arom.); 8.88 (1H, d, *J* = 5.4, =CH–); 9.32 (2H, br. s, NH_2); 11.84 (1H, d, *J* = 5.4, NH)
7b	1.04 (6H, s, 2 CH_3); 2.23 (2H, s, CH_2); 2.35 (2H, s, CH_2); 2.60 (3H, s, CH_3); 7.25 (2H, m, arom.); 7.75 (2H, m, arom.); 9.03 (2H, br. s, NH_2); 12.14 (1H, br. s, NH)
8a	1.16 (6H, s, 2 CH_3); 2.52 (2H, s, CH_2); 3.03 (2H, s, CH_2); 7.18 (2H, m, arom.); 7.47 (2H, m, arom.); 8.95 (1H, s, =CH–); 12.24 (1H, br. s, NH)
8b	1.14 (6H, s, 2 CH_3); 1.28 (3H, s, CH_3); 2.50 (2H, s, CH_2); 3.06 (2H, s, CH_2); 7.24 (2H, m, arom.); 7.55 (2H, m, arom.); 12.02 (1H, br. s, NH)
8c	2.16 (2H, m, CH_2); 2.50 (2H, m, CH_2); 2.65 (2H, m, CH_2); 7.27 (1H, t, *J* = 7.4, arom.); 7.43 (1H, t, *J* = 7.4, arom.); 7.72 (1H, d, *J* = 7.4, arom.); 7.97 (1H, d, *J* = 7.4, arom.); 8.96 (1H, s, =CH–); 12.24 (1H, br. s, NH)
8d	1.18 (6H, s, 2 CH_3); 2.50 (2H, s, CH_2); 3.01 (2H, s, CH_2); 7.24 (1H, t, *J* = 7.4, arom.); 7.38 (1H, t, *J* = 7.4, arom.); 7.70 (1H, d, *J* = 7.4, arom.); 7.85 (1H, d, *J* = 7.4, arom.); 8.93 (1H, s, =CH–); 12.25 (1H, br. s, NH)
12a	1.34 (3H, t, *J* = 7.0, $\underline{CH_3}CH_2$); 2.78 (3H, s, CH_3); 4.43 (2H, q, *J* = 7.0, OCH_2); 7.26 (2H, m, arom.); 7.43 (2H, m, arom.); 8.96 (1H, s, =CH–); 12.15 (1H, br. s, NH)
12b	1.38 (3H, t, *J* = 7.0, $\underline{CH_3}CH_2$); 2.70 (3H, s, CH_3); 4.39 (2H, q, *J* = 7.0, OCH_2); 7.12 (2H, m, arom.); 7.33–7.67 (7H, m, arom.); 12.15 (1H, br. s, NH)
12c	1.42 (3H, t, *J* = 7.0, $\underline{CH_3}CH_2$); 2.82 (3H, s, CH_3); 4.36 (2H, q, *J* = 7.0, OCH_2); 7.22 (1H, t, *J* = 7.4, arom.); 7.36 (1H, t, *J* = 7.4, arom.); 7.73 (1H, d, *J* = 7.4, arom.); 7.82 (1H, d, *J* = 7.4, arom.); 9.00 (1H, s, =CH–); 12.10 (1H, br. s, NH)
15a	1.71 (6H, s, 2 CH_3); 7.22 (2H, m, arom.); 7.51 (2H, d, *J* =8.0, arom.); 9.08 (1H, d, *J* = 5.4, =CH–); 9.28 (2H, br. s, NH_2); 11.03 (1H, d, *J* = 5.4, NH)
15b	1.72 (6H, s, 2 CH_3); 7.26 (1H, t, *J* = 7.4, arom.); 7.42 (1H, t, *J* = 7.4, arom.); 7.72 (1H, d, *J* = 7. 4, arom.); 7.88 (1H, d, *J* = 7.4, arom.); 9.05 (1H, d, *J* = 5.4, =CH–); 9.32 (2H, br. s, NH_2); 11.00 (1H, d, *J* = 5.4, NH)
17	1.30-1.41 (6H, m, 2 CH_3); 4.18-4.34 (4H, m, 2 OCH_2); 7.10-7.24 (2H, m, arom.); 7.40 (2H, d, *J* = 8.0, arom.); 8.78 (1H, d, *J* = 5.4, =CH–); 9.09 (2H, br. s, NH_2); 11.49 (1H, d, *J* = 5.4, NH)
18	1.24 (3H, t, *J* =7.0, CH_3); 4.26 (2H, q, *J* = 7.0, OCH_2); 7.12-7.25 (2H, m, arom.); 7.54 (2H, d, *J* = 8.0, arom.); 8.24 (1H, s, =CH–); 9.26 (1H, s, NH); 11.24 (1H, s, NH)
20	2.52 (3H, s, CH_3); 3.98 (3H, s, OCH_3); 6.86-7.22 (5H, m, arom.); 7.40 (2H, t, *J* = 7.6, arom.); 8.38 (1H, d, *J* =7.4, arom.); 9.05 (1H, d, *J* = 5.4, =CH–); 9.30 (2H, br. s, NH_2); 11.70 (1H, s, NH); 12.12 (1H, d, *J* = 5.4, NH)
22a	2.48 (3H, s, CH_3); 4.12 (3H, s, OCH_3); 6.94-7.32 (5H, m, arom.); 7.42 (2H, t, *J* = 7.6, arom.); 8.43 (1H, d, *J* = 7.4, arom.); 9.06 (1H, s, =CH–); 11.83 (1H, s, NH); 12.10 (1H, s, NH)
22b	2.31 (3H, s, CH_3); 2.60 (3H, s, CH_3); 7.16-7.35 (4H, m, arom.); 7.53-7.65 (4H, m, arom.); 8.76 (1H, s, =CH–); 11.27 (1H, s, NH); 11.50 (1H, s, NH)
22c	2.63 (6H, s, 2 CH_3); 7.06-7.23 (4H, m, arom.); 7.45-7.62 (4H, m, arom.); 11.21 (1H, s, NH); 11.80 (1H, s, NH)
22d	2.32 (3H, s, CH_3); 2.61 (6H, s, 2 CH_3); 7.04-7.42 (4H, m, arom.); 7.54-7.82 (4H, m, arom.); 9.77 (1H, s, NH); 11.72 (1H, s, NH)
23	7.23 (2H, m, arom.); 7.51 (2H, d, *J* = 7.4, arom.); 8.95 (1H, d, *J* = 5.4, =CH–); 9.28 (2H, br. s, NH_2); 11.00 (1H, s, NH barb.); 11.13 (1H, s, NH barb.); 11.41 (1H, d, *J* = 5.4, NH)

TABLE 2. Yields and Characteristics of the Compounds Synthesized

Compound	Empirical formula	Found, % / Calculated, %			M* / M[1]	mp, °C	Yield, %
		C	H	N			
5a	$C_{14}H_{12}N_4O_2$	62.55 / 62.68	4.38 / 4.51	20.92 / 20.88	268 / 268.28	207-209	60
5b	$C_{15}H_{14}N_4O_2$	63.91 / 63.82	4.96 / 5.00	19.77 / 19.85	282 / 282.31	171-173	45
5c	$C_{24}H_{16}N_4OS$	70.77 / 70.57	4.02 / 3.95	13.66 / 13.72	408 / 408.48	260-261	65
7a	$C_{17}H_{18}N_4O_3$	62.69 / 62.57	5.68 / 5.56	17.25 / 17.17	326 / 326.36	220-221	62
7b	$C_{18}H_{20}N_4O_3$	63.74 / 63.52	5.74 / 5.92	16.52 / 16.46	340 / 340.40	235-236	58
8a	$C_{17}H_{16}N_4O_2$	66.01 / 66.22	5.11 / 5.23	18.24 / 18.17	308 / 308.35	241-243	74
8b	$C_{18}H_{18}N_4O_2$	66.95 / 67.00	5.32 / 5.58	17.68 / 17.37	322 / 322.38	289-291	50
8c	$C_{15}H_{12}N_4OS$	60.95 / 60.79	4.25 / 4.08	18.88 / 18.91	296 / 296.35	296-298	51
8d	$C_{17}H_{16}N_4OS$	63.08 / 62.94	5.02 / 4.97	17.32 / 17.27	324 / 324.41	290-291	46
12a	$C_{15}H_{14}N_4O_3$	60.14 / 60.40	4.85 / 4.73	18.68 / 18.78	298 / 298.31	221-223	53
12b	$C_{21}H_{18}N_4S_3$	67.55 / 67.37	4.78 / 4.85	15.03 / 14.96	422 / 422.59	187-188	41
12c	$C_{15}H_{14}N_4O_2S$	57.44 / 57.31	4.63 / 4.49	17.65 / 17.82	314 / 314.37	225-227	57
15a	$C_{15}H_{14}N_4O_5$	54.68 / 54.55	4.48 / 4.27	16.78 / 16.96	330 / 330.30	270-271	40
15b	$C_{15}H_{14}N_4O_4S$	51.88 / 52.02	4.21 / 4.07	16.25 / 16.18	346 / 346.37	272-273	42
17	$C_{16}H_{18}N_4O_5$	55.24 / 55.49	5.38 / 5.24	16.37 / 16.18	346 / 346.35	146-148	36
18	$C_{14}H_{12}N_4O_4$	56.20 / 56.00	4.28 / 4.03	18.57 / 18.66	300 / 300.28	224-225	17
20	$C_{20}H_{19}N_5O_4$	61.27 / 61.06	4.68 / 4.87	17.77 / 17.80	393 / 393.41	242-243	37
22a	$C_{20}H_{17}N_5O_3$	64.23 / 63.99	4.51 / 4.56	18.56 / 18.66	375 / 375.40	245-247	68
22b	$C_{20}H_{17}N_5O_2$	66.58 / 66.84	4.68 / 4.77	19.54 / 19.49	359 / 359.40	291-293	32
22c	$C_{21}H_{16}F_3N_5O_2$	58.85 / 59.02	3.84 / 3.77	16.42 / 16.39	427 / 427.43	264-265	49
22d	$C_{21}H_{19}N_5OS$	64.56 / 64.76	5.03 / 4.92	18.02 / 17.98	389 / 389.49	>300	35
23	$C_{13}H_{10}N_6O_4$	49.87 / 49.69	3.25 / 3.21	26.64 / 26.74	314 / 314.26	>300	41

* M – found. M^1 – calculated.

EXPERIMENTAL

Monitoring of the course of reactions and the purity of compounds was by TLC on Silufol UV-254 strips with 1:3 chloroform–ethyl acetate as eluant. ^{1}H NMR spectra of DMSO-d_6 solutions, relative to TMS, were recorded on a Bruker AC-300 (300 MHZ) machine. Mass spectra were recorded with an LKB 9000 with an ionizing energy of 70 eV.

Benzoxa(thia)zoyl-2-guanidines 1a,b were obtained by a known method [9].

2-(1,3-Benzoxa(thia)zol-2-ylamino)-4-methyl(phenyl)acylpyrimidines 5a-c. A solution of the corresponding guanidine (10 mmol) and the 1,3-diketone **3** (10 mmol) in the orthoester **2** (10 ml) was boiled for 30-60 min. The precipitate formed was filtered off, recrystallized from DMF, and dried.

2-Amino(1,3-benzoxazol-2-ylimino)methylaminomethylene-5,5-dimethyl-1,3-cyclohexanedione (7a) and 2-[Amino(1,3-benzoxa-2-ylimino)methylaminoethylidene]-5,5-dimethyl-1,3-cyclohexanedione (7b). A mixture of guanidine **1a** (1.76 g, 10 mmol) and dimedone (2.2 g, 15 mmol) was ground, the corresponding orthoester **2** (10 ml) was added and the mixture was heated at 140°C for 30 min. An additional 2.2 g of dimedone was added, mixed, and heated for a further 30 min. After cooling the precipitate was washed free from the bis-adduct of dimedone and the orthoester on the filter with chloroform, and the insoluble, colorless residue was recrystallized from DMF.

2-(1,3-Benzoxazol-2-ylamino)-7,7-dimethyl-5,6,7,8-tetrahydro-5-quinaxolinone (8a) and 2-(1,3-Benzoxazol-2-ylamino)-4,7,7-trimethyl-5,6,7,8-tetrahydro-5-quinaxolinone (8b). Compound **7a** (1.63 g, 5 mmol) or **7b** (1.70 g, 5 mmol) was ground with calcined sodium acetate (1.5 g), glacial AcOH (15 ml) was added and the mixture boiled for 20-30 min. The mixture was dropped into cold water (100 ml), the precipitate was separated, dried and recrystallized from DMF.

2-(1,3-Benzothiazol-2-ylmino)-5,6,7,8-tetrahydro-5-quinazolinone (8c) and 2-(1,3-Benzothiazol-2-ylmino)-7,7-dimethyl-5,6,7,8-tetrahydro-5-quinazolinone (8d) were made analogously to the synthesis of compounds **7a,b**.

Ethyl 2-(1,3-Benzoxazol-2-ylamino)-4-methylpyrimidine-5-carboxylate (12a) and Ethyl 2-(1,3-Benzothiazol-2-ylamino)-4-methylpyrimidine-5-carboxylate (12c) . A mixture of the corresponding guanidine **1** (10 mmol), acetoacetic ester (10 mmol), and triethyl orthofornate (15 ml) was boiled for 40-60 min, the precipitate was filtered off an recrystallized from DMF.

Ethyl 2-(1,3-Benzoxazol-2-ylamino-4-methyl-6-phenylpyrimidine-5-carboxylate (12b). A mixture of guanidine **1a** (1.76 g, 10 mmol), benzoylacetic ester (10 mmol), and triethyl orthoformate (15 ml) was boiled for 1 h. The precipitate was filtered off and recrystallized from DMF.

5-Amino(1,3-benzoxazol-2-ylamino)methylaminomethylene-2,2-dimethyl-1,3-dioxane-4,6-dione (15a) and 5-Amino(1,3-benzothiazol-2-ylamino)methylaminomethylene-2,2-dimethyl-1,3-dioxane-4,6-dione (15b). Triethyl orthoformate (10 ml) was added to a ground mixture of guanidine **1a** (1.76 g, 10 mmol) or **1b** (1.92 g, 10 mmol) and Meldrum acid (1.44 g, 10 mmol) and the mixture was boiled for 1 h 30 min., cooled, the colorless precipitate was filtered off and recrystallized from DMF.

Diethyl 2-Amino(1,3-benzoxazol-2-ylimino)methylaminomethylenemalonate (17). A solution of guanidine **1a** (1.76 g, 10 mmol)and ethoxymethyenmalonate (**13**) (2.60 g, 12 mmol) in xylene (20 ml) was boiled for 5 h, cooled , the precipitate was filtered off, xylene (20 ml) was added to it, brought to the boil, and the hot solution was filtered. Crystals of **17** formed from the solution on cooling. **2-(1,3-Benzoxazol-2-ylamino)-5-[1-ethoxymethylidene]-1,4,5,6-tetrahydropyrimidine-4,6-dione (18)**, which was insoluble in xylene, was recrystallized from DMF.

N-(2-Methoxyphenyl)-2-acetyl-3-amino(1,3-benzoxazol-2-ylimino)methylamino-2-propenamide (20). A mixture of guanidine **1a** (1.76 g, 10 mmol) with anilide **19c** (2.07 g, 10 mmol) in orthoester **2a** was boiled for 40 min, the precipitate was filtered off and recrystallized from dioxan.

5-[N-(2-Methoxyphenyl)]-2-(1,3-benzoxazol-2-ylamino)-4-methylpyrimidine-5-carboxamide (22a) was obtained from compound **20** by the method described for compound **8a**.

5-[N-(4-Methylphenyl)]-2-(1,3-benzoxazol-2-ylamino)-4-methylpyrimidine-5-carboxamide (22b), **5-[N-(2-Trifluoromethylphenyl)]-2-(1,3-benzoxazol-2-ylamino)-4,6-dimethylpyrimidine-5-carboxamide (22c), and 5-[N-(2-Methylphenyl)]-2-(1,3-benzothiazol-2-ylamino)-4,6-dimethylpyrimidine-5-carboxamide (22d).** A mixture of guanidine **1a,b** (10 mmol) and the corresponding anilide **19** (10 mmol) in orthoester **2** (15 ml) was boiled for 1 h 30 min, cooled, the precipitate filtered off and recrystallized from DMF.

5-Amino(1,3-benzoxazol-2-ylimino)methylaminomethylenehexahydropyrimidine-2,4,6-trione (23) was obtained analogously to compound **15a** using an equimolar amount of barbituric acid in place of Meldrum acid.

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