Russian Journal of Organic Chemistry, Vol. 40, No. 1, 2004, pp. 85–89. Translated from Zhurnal Organicheskoi Khimii, Vol. 40, No. 1, 2004, pp. 94–98.

Synthesis of 8-Aryl[1,2,4]triazolo[1,5-*d*][1,2,4]triazin-5(6*H*)-ones by S_N^H Reactions

V. L. Rusinov[1], G. V. Zyryanov[1], I. N. Egorov[1], E. N. Ulomskii[1], G. G. Aleksandrov[1], and O. N. Chupakhin[2]

[1]*Ural State Technical University, ul. Mira 19, Yekaterinburg, 620002 Russia*
fax: (3432) 740458; e-mail: rusinov@htf.ustu.ru
[2]*Institute of Organic Synthesis, Ural Division, Russian Academy of Sciences, Yekaterinburg, Russia*

Received November 5, 2003

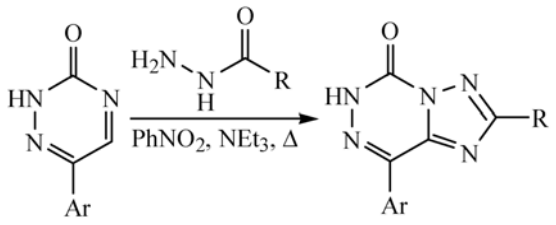
Abstract—A new procedure has been proposed for the synthesis of 8-aryl[1,2,4]triazolo[1,5-*d*][1,2,4]-triazin-5(6*H*)-ones by reaction of 6-aryl-1,2,4-triazin-3(2*H*)-ones with hydrazides derived from aliphatic, aromatic, and heterocyclic carboxylic acids. The process invloves nucleophilic substitution of hydrogen (S_N^H) in aryltriazinones, oxidative closure of azole ring, and Dimroth rearrangement.

Persistent interest of chemists in synthetic routes to azoloazines is explained primarily by a wide spectrum of biological properties of these heterocycles which may be regarded as structural analogs of naturally occurring purine bases. A specific group of azoloazines includes azaindolizines, i.e., azolopyrimidines, azolo[1,2,4]triazines, and azolo[1,2,3,5]tetrazines. Some compounds of this series were found to exhibit antitumor and antiviral activity [1–3].

The known methods for preparation of azaindolizines, despite their diversity, can be divided into two main groups. The first group includes those based on fusion of an azine ring to azole and building up of an azole ring on the basis of already existing azine ring [4]. Examples of the second group methods are the synthesis of [1,2,4]triazolo[4,3-*b*][1,2,4]triazin-7(5*H*)-ones by cyclization of 3-hydrazido-1,2,4-triazin-5(2*H*)-ones [5], preparation of [1,2,4]triazolo[4,3-*d*][1,2,4]triazin-5-ones from 5-hydrazino-1,2,4-triazines and ortho esters [6], and formation of 5-amino-8-benzyl[1,2,4]triazolo[4,3-*d*][1,2,4]triazines in reaction of 3-substituted 6-benzyl-5-hydrazino-1,2,4-triazines with carbon disulfide [7]. In the above examples, the initial 1,2,4-triazine ring contains a hydrazine moiety which is subsequently incorporated into the new triazole fragment.

In the present communication we report on a new simple procedure for the synthesis of [1,2,4]triazolo[1,5-*d*][1,2,4]triazine derivatives via reaction of 1,2,4-triazine-3-ones with carboxylic acid hydrazides, which involves oxidative version of nucleophilic substitution of hydrogen (S_N^H) [8]. By heating 6-aryl-1,2,4-triazin-3(2*H*)-ones **Ia** and **Ib** with carboxylic acid hydrazides in nitrobenzene (which is a high-boiling solvent possessing oxidative properties) in the presence of triethylamine we obtained [1,2,4]triazolo[1,5-*d*][1,2,4]triazin-5(6*H*)-ones **II–VI** in satisfactory yields (Scheme 1). The presence of a base is necessary to displace the equilibrium toward formation of intermediate adducts **VII–XI** (Scheme 2). These adducts were isolated when triazinones **Ia** and **Ic** and the corresponding hydrazides were heated for a short time in nitrobenzene or when the reaction was carried out in boiling dioxane, tetrahydrofuran, or ethanol.

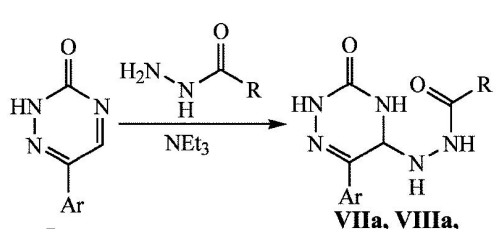
Scheme 1.



Ia, IIb **IIa–VIa, IVb**

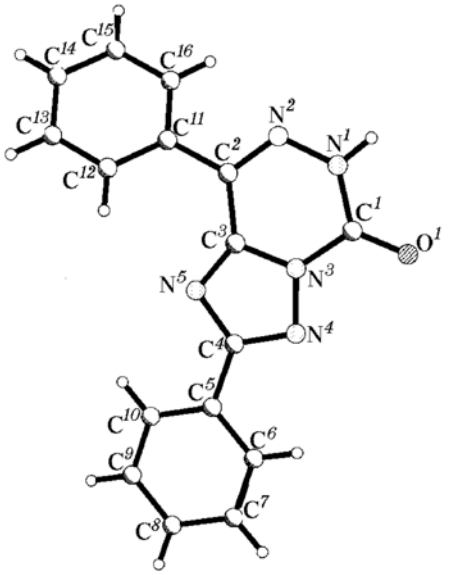
Ar = Ph (**a**), 4-MeC_6H_4 (**b**); **II**, R = Me; **III**, R = Ph; **IV**, R = 3-$O_2NC_6H_4$; **V**, R = 4-Py; **VI**, R = 3-Py.

Scheme 2.



Ia, c **VIIa, VIIIa, VIIIc–XIa, XIc**

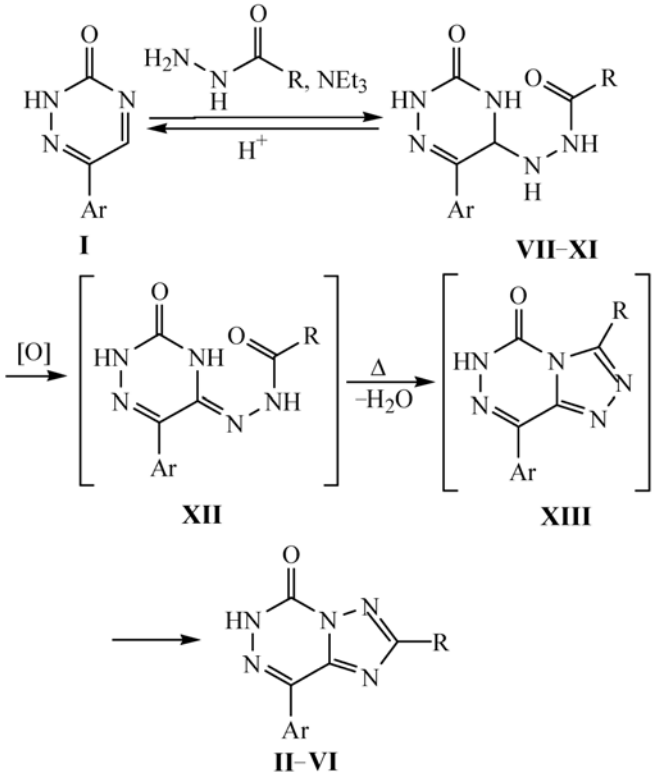
Ar = Ph (**a**), 4-BrC_6H_4 (**b**); **VII**, R = Me; **VIII**, R = Ph; **IX**, R = 3-$O_2NC_6H_4$; **X**, R = 4-Py; **XI**, R = 3-Py.



Structure of the molecule of 2,8-diphenyl[1,2,4]triazolo-[1,5-*d*][1,2,4]triazin-5(6*H*)-one (**IIIa**) according to the X-ray diffraction data.

Compounds **II–VI** showed in the ^{1}H NMR spectra signals from protons in the R substituent, a five-proton multiplet from the aromatic protons at δ 7.4–8.6 ppm, and a broadened signal from the NH group at δ 13.4–13.8 ppm. The IR spectra of **II–VI** contained absorption bands typical of NH and C=O groups (see table). The structure

Scheme 3.



Principal bond lengths (*d*) and bond angles (ω) in the molecule of 6,8-diphenyl[1,2,4]triazolo[5,1-*d*][1,2,4]triazin-5(6*H*)-one (**IIIa**)

Bond	*d*, Å	Angle	ω, deg
O^1– C^1	1.217(3)	$N^2N^1C^1$	128.3(2)
N^1–N^2	1.359(3)	$C^2N^2N^1$	118.8(2)
N^1–C^1	1.364(3)	$N^4N^3C^3$	110.8(2)
N^2–C^2	1.302(3)	$N^4N^3C^1$	124.7(2)
N^3-N^4	1.358(3)	$C^3N^3C^1$	124.7(2)
N^3–C^3	1.366(3)	$C^4N^4N^3$	101.6(2)
N^3–C^1	1.395(3)	$C^3N^5C^4$	103.0(2)
N^4–C^4	1.337(3)	$O^1C^1N^1$	125.3(2)
N^5–C^3	1.320(3)	$O^1C^1N^3$	123.6(2)
N^5–C^4	1.378(3)	$N^1C^1N^3$	111.1(2)
C^2–C^3	1.449(3)	$N^2C^2C^3$	119.4(2)
C^2–C^{11}	1.476(3)	$N^5C^3N^3$	109.5(2)
C^4–C^5	1.465(4)	$N^5C^3C^2$	132.9(2)
C^5–C^6	1.396(4)	$N^3C^3C^2$	117.5(2)
C^5–C^{10}	1.397(4)	$N^4C^4N^5$	115.0(2)
C^6–C^7	1.384(4)		
C^7–C^8	1.397(4)		
C^8–C^9	1.386(4)		
C^9–C^{10}	1.385(4)		
C^{11}–C^{12}	1.387(4)		
C^{11}–C^{16}	1.393(4)		
C^{12}–C^{13}	1.389(4)		
C^{13}–C^{14}	1.382(4)		
C^{14}–C^{15}	1.378(4)		
C^{15}–C^{16}	1.390(4)		

of the condensation products, specifically the mode of junction of the triazole and triazine rings, was unambiguously proved by X-ray analysis of 2,8-diphenyl[1,2,4]triazolo[1,5-*d*][1,2,4]triazin-5(6*H*)-one (**IIIa**) (see figure). The three cyclic fragments in molecule **IIIa** lie in one plane, and the carbon–carbon bond lengths therein are fairly similar (1.378, 1.379 Å; see table). Despite an appreciable difference in the C–N bond lengths in the heterocyclic fragment (1.302 and 1.391 Å), alteration of bond lengths is nevertheless observed [9], indicating an essential conjugation in this fragment.

The structure of primary substitution products **VII–XI** was determined on the basis of the ^{1}H NMR spectra which contain signals from protons of the aromatic fragment (Ar), group R, and amide moieties. In addition, a

broadened one-proton signal from 5-H was present at δ 5.20–5.30 ppm. This signal is converted into a singlet upon addition of CD_3COOD, indicating that the 5-H proton is coupled with the neighboring NH protons.

A plausible reaction mechanism is shown in Scheme 3. It includes primary nucleophilic addition of hydrazides as the sp^2-hybridized carbon atom in trazinone **I** to give σ^H-adducts **VII**–**XI** which are oxidized with nitrobenzene to S_N^H products **XII**. Dehydration of the latter leads to aromatic system **XIII**. [1,2,4]Triazolo[4,3-*d*][1,2,4]triazines **XIII** undergo Dimroth rearrangement to afford final products **II**–**VI**. This rearrangement is known to readily occur under basic comditions [10], but its thermal version, in particular on heating in boiling nitrobenzene, is also possible [11].

EXPERIMENATL

The melting points were determined on a Boetius device. The ^{1}H NMR spectra were recorded on a Bruker WH-250 spectrometer at 250 MHz using DMSO-d_6 as solvent and tetramethylsilane as internal reference. The mass spectra were run on a Varian MAT 311A instrument with direct sample admission into the ion source. The IR spectra were measured on a Specord M-80 spectrophotometer. Triazines **Ia** and **Ib** were synthesized by the procedure reported in [12].

8-Aryl[1,2,4]triazolo[1,5-*d*][1,2,4]triazin-5(6*H*)-ones II–VI. Nitrobenzene, 2–3 ml, and triethylamine, 0.5 ml, were added to a mixture of 1 mmol of 6-aryl-1,2,4-triazin-3(2*H*)-one **Ia** or **Ib** and 1 mmol of the corresponding carboxylic acid hydrazide. The mixture was heated for 1.5 h or more under reflux until it became homogeneous (i.e., until complete dissolution of intermediately precipitated colorless solid) and was left to stand for 24 h at room temperature. The precipitate was filtered off, washed with a small amount of nitrobenzene and then with benzene, and recrystallized from acetone.

2-Methyl-8-phenyl[1,2,4]triazolo[1,5-*d*][1,2,4]-triazin-5(6*H*)-one (IIa). Yield 26%, mp 225°C. IR spectrum, ν, cm^{-1}: 1730 (C=O), 3430 (NH). ^{1}H NMR spectrum, δ, ppm: 2.60 s (3H, CH_3), 7.42–7.56 m (3H, H_{arom}, Ph), 8.21–8.36 m (2H, H_{arom}, Ph), 13.30 br.s (1H, 6-H). Mass spectrum, *m*/*z*: $[M]^+$ 227. Found, %: C 58.24; H 3.95; N 30.95. $C_{11}H_9N_5O$. Calculated, %: C 58.15; H 3.99; N 30.82.

2,8-Diphenyl[1,2,4]triazolo[1,5-*d*][1,2,4]triazin-5(6*H*)-one (IIIa). Yield 30%, mp 272°C. IR spectrum, ν, cm^{-1}: 1720 (C=O), 3420 (NH). ^{1}H NMR spectrum, δ, ppm: 7.40–7.56 m (6H, H_{arom}), 8.21–8.36 m (2H, H_{arom}), 8.43–8.46 m (2H, H_{arom}), 13.50 br.s (1H, 6-H). Mass spectrum, *m*/*z*: $[M]^+$ 289. Found, %: C 66.55; H 3.83; N 24.26. $C_{16}H_{11}N_5O$. Calculated, %: C 66.43; H 3.83; N 24.21.

2-(3-Nitrophenyl)-8-phenyl[1,2,4]triazolo[1,5-*d*]-[1,2,4]triazin-5(6*H*)-one (IVa). Yield 31%, mp 268°C. IR spectrum, ν, cm^{-1}: 1350 (NO_2), 1520 (NO_2), 1720 (C=O), 3420 (NH). ^{1}H NMR spectrum, δ, ppm: 7.52–7.61 m (3H, Ph), 7.84–7.90 m (1H, Ar), 8.38–8.42 m (3H, Ar, Ph), 8.65–8.68 m (1H, Ar), 8.95 m (1H, Ar), 13.66 br.s (1H, 6-H). Mass spectrum, *m*/*z*: $[M]^+$ 334. Found, %: C 57.38; H 3.11; N 25.06. $C_{16}H_{10}N_6O_3$. Calculated, %: C 57.49; H 3.02; N 25.14.

2-(3-Nitrophenyl)-8-(4-tolyl)[1,2,4]triazolo[1,5-*d*]-[1,2,4]triazin-5(6*H*)-one (IVb). Yield 25%, mp 248°C. IR spectrum, ν, cm^{-1}: 1360 (NO_2), 1510 (NO_2), 1730 (C=O), 3460 (NH). ^{1}H NMR spectrum, δ, ppm: 2.42 s (3H, CH_3), 7.42–7.44 m (2H, Tol), 7.91–7.95 m (1H, $NO_2C_6H_4$), 8.30–8.32 m (2H, Tol), 8.43–8.46 m (1H, $NO_2C_6H_4$), 8.67–8.69 m (1H, $NO_2C_6H_4$), 8.93 m (1H, $NO_2C_6H_4$), 13.65 br.s (1H, 6-H). Mass spectrum, *m*/*z*: $[M]^+$ 348. Found, %: C 58.71; H 3.34; N 24.19. $C_{17}H_{12}N_6O_3$. Calculated, %: C 58.62; H 3.47; N 24.13.

8-Phenyl-2-(4-pyridyl)[1,2,4]triazolo[1,5-*d*]-[1,2,4]triazin-5(6*H*)-one (Va). Yield 32%, mp 290°C. IR spectrum, ν, cm^{-1}: 1740 (C=O), 3400 (NH). ^{1}H NMR spectrum, δ, ppm: 7.54–7.56 m (3H, Ph), 8.16–8.18 m (2H, Py), 8.41–8.44 m (2H, Ph), 8.76–8.78 m (2H, Py), 13.70 br.s (1H, 6-H). Mass spectrum, *m*/*z*: $[M]^+$ 290. Found, %: C 62.17; H 3.40; N 28.89. $C_{15}H_{10}N_6O$. Calculated, %: C 62.07; H 3.47; N 28.95.

8-Phenyl-2-(3-pyridyl)[1,2,4]triazolo[1,5-*d*]-[1,2,4]triazin-5(6*H*)-one (VIa). Yield 34%, mp 282°C. IR spectrum, ν, cm^{-1}: 1740 (C=O), 3450 (NH). ^{1}H NMR spectrum, δ, ppm: 7.54–7.66 m (3H, Ph), 8.40–8.44 m (2H, Ph), 8.56–8.57 m (1H, Py), 8.67 m (1H, Py), 8.77 m (1H, Py), 9.41 m (1H, Py), 13.60 br.s (1H, 6-H). Mass spectrum, *m*/*z*: $[M]^+$ 290. Found, %: C 62.19; H 3.55; N 28.82. $C_{15}H_{10}N_6O$. Calculated, %: C 62.07; H 3.47; N 28.95.

Reaction of 6-aryl-1,2,4-triazin-3(2*H*)-ones I with carboxylic acid hydrazides (*general procedure*). A mixture of 1 mmol of 6-aryl-1,2,4-triazin-3(2*H*)-one **I** and 1 mmol of the corresponding carbohydrazide was dissolved in 5 ml of dioxane, 0.5 ml of triethylamine was added, and the mixture was heated for 1 h under reflux. The colorless precipitate was filtered off, washed with dioxane, dried, and recrystallized from aqueous dimethylformamide.

***N*-(3-Oxo-6-phenyl-2,3-dihydro-1,2,4-triazin-5-yl)acetohydrazide (VIIa).** Yield 58%, mp 222–224°C. ^{1}H NMR spectrum, δ, ppm: 1.80 s (3H, $COCH_3$), 5.22 m

(1H, 5-H), 7.35–7.37 m (3H, H_{arom}), 7.67 br.s (1H, 4-H), 7.92–7.94 m (2H, H_{arom}), 9.33–9.34 m (1H, NNHCO), 10.31 s (1H, 2-H). Found, %: C 53.32; H 5.39; N 28.40. $C_{11}H_{13}N_5O_2$. Calculated, %: C 53.44; H 5.30; N 28.32.

***N'*-(3-Oxo-6-phenyl-2,3-dihydro-1,2,4-triazin-5-yl)benzohydrazide (VIIIa).** Yield 55%, mp 258–259°C. ^{1}H NMR spectrum, δ, ppm: 5.34–5.37 m (1H, 5-H), 5.47–5.51 s (1H, NNHCO), 7.30–7.52 m (6H, H_{arom}), 7.78 br.s (1H, 4-H), 7.82–7.85 m (2H, COPh), 7.98–8.01 m (2H, Ph), 9.96–9.98 d (1H, NHNCO), 10.28–10.29 s (1H, 2-H). Found, %: C 62.21; H 4.75; N 22.55. $C_{16}H_{15}N_5O_2$. Calculated, %: C 62.13; H 4.89; N 22.64.

***N'*-[6-(4-Bromophenyl)-3-oxo-2,3-dihydro-1,2,4-triazin-5-yl]benzohydrazide (VIIIc).** Yield 48%, mp 251–252°C. ^{1}H NMR spectrum, δ, ppm: 5.38–5.40 m (1H, 5-H), 5.70–5.73 s (1H, NHNCO), 7.44–7.48 m (2H, COPh), 7.52–7.55 m (1H, COPh), 7.61–7.64 d (2H, Ph), 7.79–7.81 m (2H, COPh), 7.90–7.93 br.s (3H, Ph, 4-H), 10.03–10.05 d (1H, NNHCO), 10.52 s (1H, 2-H). Found, %: C 49.66; H 3.76; Br 20.69; N 17.95. $C_{16}H_{14}BrN_5O_2$. Calculated, %: C 49.50; H 3.63; Br 20.58; N 18.04.

***N'*-(3-Oxo-6-phenyl-2,3-dihydro-1,2,4-triazin-5-yl)-3-nitrobenzohydrazide (IXa).** Yield 76%, mp 259–261°C. ^{1}H NMR spectrum, δ, ppm: 5.37–5.40 m (1H, 5-H), 5.62–5.66 m (1H, NHNCO), 7.35–7.43 m (3H, Ph), 7.71–7.78 m (1H, 3-$NO_2C_6H_4$), 7.94 br.s (1H, 4-H), 7.99–8.01 m (2H, Ph), 8.24–8.27 m (1H, 3-$NO_2C_6H_4$), 8.34–8.37 m, (1H, 3-$NO_2C_6H_4$), 8.70 s (1H, 3-$NO_2C_6H_4$), 10.43–10.45 d (1H, NNHCO), 10.41 s (1H, 2-H). Found, %: C 54.15; H 4.09; N 23.79. $C_{16}H_{14}N_6O_4$. Calculated, %: C 54.24; H 3.98; N 23.72.

***N'*-[6-(4-Bromophenyl)-3-oxo-2,3-dihydro-1,2,4-triazin-5-yl]-3-nitrobenzohydrazide (IXc).** Yield 60%, mp 252–253°C. ^{1}H NMR spectrum, δ, ppm: 5.36 m (1H, 5-H), 5.62 br.s (1H, NHNCO), 7.52–7.57 d (2H, BrC_6H_4), 7.71–7.78 m (1H, 3-$NO_2C_6H_4$), 7.92–7.95 d (2H, BrC_6H_4), 8.23–8.32 m (2H, 3-$NO_2C_6H_4$), 8.70 s (1H, 3-$NO_2C_6H_4$), 7.88 br.s (1H, 4-H), 10.39 br.s (2H, NNHCO, 2-H). Found, %: C 44.28; H 3.03; Br 18.38; N 19.29. $C_{16}H_{13}BrN_6O_4$. Calculated, %: C 44.36; H 3.02; Br 18.44; N 19.40.

***N'*-(3-Oxo-6-phenyl-2,3-dihydro-1,2,4-triazin-5-yl)-4-pyridinecarbohydrazide (Xa).** Yield 55%, mp 242–243°C. ^{1}H NMR spectrum, δ, ppm: 5.38 m (1H, 5-H), 5.59 m (1H, NHNCO), 7.33–7.39 m (3H, Ph), 7.71–7.73 d (2H, Py), 7.80 br.s (1H, 4-H), 7.96–8.00 m (2H, Ph), 8.64–8.66 d (2H, Py), 10.26–10.29 d (1H, NNHCO), 10.32 m (1H, 2-H). Found, %: C 58.15; H 4.68; N 27.16. $C_{15}H_{14}N_6O_2$. Calculated, %: C 58.06; H 4.55; N 27.08.

***N'*-[6-(4-Bromophenyl)-3-oxo-2,3-dihydro-1,2,4-triazin-5-yl]-4-pyridinecarbohydrazide (Xc).** Yield 38%, mp 247–248°C. ^{1}H NMR spectrum, δ, ppm: 5.35 m (1H, 5-H), 5.63 m (1H, NHNCO), 7.51–7.55 d (2H, BrC_6H_4), 7.69–7.72 d (2H, Py), 7.83 br.s (1H, 4-H), 7.90–7.93 d (2H, BrC_6H_4), 8.64–8.66 d (2H, Py), 10.25–10.27 br.s (1H, NNHCO), 10.40 br.s (1H, 2-H). Found, %: C 46.36; H 3.24; Br 20.46; N 21.48. $C_{15}H_{13}BrN_6O_2$. Calculated, %: C 46.29; H 3.37; Br 20.53; N 21.59.

***N'*-(3-Oxo-6-phenyl-2,3-dihydro-1,2,4-triazin-5-yl)-4-pyridinecarbohydrazide (XIa).** Yield 54%, mp 245–246°C. ^{1}H NMR spectrum, δ, ppm: 5.36–5.39 m (1H, 5-H), 5.56–5.60 m (1H, NHNCO), 7.31–7.45 m (4H, Py, Ph), 7.80 br.s (1H, 4-H), 7.97–8.00 m (2H, Ph), 8.11–8.16 m (1H, Py), 8.63–8.66 m (1H, Py), 8.95–8.96 m (1H, Py), 10.18–10.21 d (1H, NNHCO), 10.33 m (1H, 2-H). Found, %: C 57.91; H 4.48; N 27.01. $C_{15}H_{14}N_6O_2$. Calculated, %: C 58.06; H 4.55; N 27.08.

***N'*-[6-(4-Bromophenyl)-3-oxo-2,3-dihydro-1,2,4-triazin-5-yl]-3-pyridinecarbohydrazide (XIc).** Yield 50%, mp 251–253°C. ^{1}H NMR spectrum, δ, ppm: 5.34–5.38 m (1H, 5-H), 5.62–5.66 m (1H, NHNCO), 7.42–7.46 m (1H, Py), 7.54–7.57 d (2H, BrC_6H_4), 7.83 br.s (1H, 4-H), 7.91–7.95 d (2H, BrC_6H_4), 8.12–8.15 m (1H, Py), 8.65–8.67 m (1H, Py), 8.95–8.96 m (1H, Py), 10.18–10.20 m (1H, NNHCO), 10.42 br.s (1H, 2-H). Found, %: C 46.37; H 3.40; Br 20.50; N 21.63. $C_{15}H_{13}BrN_6O_2$. Calculated, %: C 46.29; H 3.37; Br 20.53; N 21.59.

This study was financially supported by the Russian Foundation for Basic Research (project no. 01-03-96 443).

REFERENCES

1. Wang, Y., Stevens, M., Chan, T., Di Benedetto, D., Ding, Z., Gala, D., Hou, D., Kugelman, M., Leong, W., Kuo, S., Mas, J.L., Schumaher, D.P., Shutts, B.P., Smith, L., Zhan, Z.-Y.J., and Thomson, W.T., *J. Org. Chem*., 1997, vol. 62, p. 7288.
2. Rusinov, V.L., Ulomskii, E.N., Chupakhin, O.N., Zubairov, M.M., Kapustin, A.B., and Mitin, N.I., *Khim.-Farm. Zh*., 1990, no. 9, p. 41.
3. Lhassani, M., Chavignon, O., and Chesal, J.-M., *J. Med. Chem*., 1999, vol. 34, p. 271.
4. Trust, R.I. and Perkinson, N.A., *J. Heterocycl. Chem*., 1979, vol. 16, p. 1393.

5. Daunis, J. and Follet, M., *Bull. Soc. Chim. Fr*., 1976, nos. 7–8, p. 1178.
6. Lovelette, C.A. and Geagan, K., *J. Heterocycl. Chem*., 1982, vol. 19, p. 1345.
7. Eid, M.M., El-Zanaty, A.M.K., and Ibrahim, Y.A., *Indian J. Chem., Sect. B*, 1990, vol. 29, p. 435.
8. Chupakhin, O.N., Charushin, V.N., and van der Plas, H.C., *Nucleophilic Aromatic Substitution of Hydrogen*, New York, Academic, 1994, p. 367.
9. Allen, F.H. and Kennard, O., *Chem. Design Autom. News*, 1993, vol. 8, p. 31.
10. Brown, D.J. and Nagamatsu, T., *Aust. J. Chem*., 1977, vol. 30, p. 2515.
11. Bee, J.A. and Rose, F.L., *J. Chem. Soc. C*, 1966, p. 2031.
12. Lalezari, I., Sharghi, N., Shafiee, A., and Yalpani, I., *J. Heterocycl. Chem*., 1969, vol. 6, p. 403.