*Russian Journal of Organic Chemistry, Vol. 41, No. 7, 2005, pp. 1071–1080. Translated from Zhurnal Organicheskoi Khimii, Vol. 41, No. 7, 2005, pp. 1092–1100.*


# Fluorine-Containing Heterocycles: XII.* Fluorine-Containing Quinazolin-4-ones and Azolo[*a*]quinazolinone Derivatives

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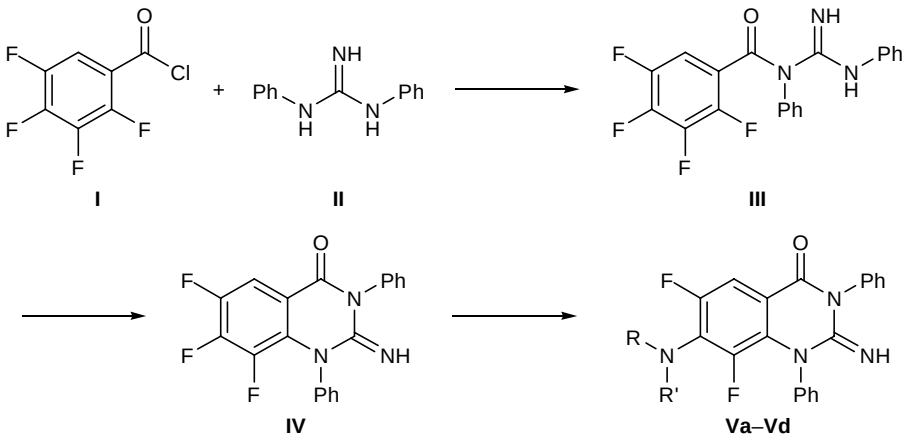
Received June 14, 2004

**Abstract**—Methods for the synthesis of fluorine-containing derivatives of 2-imino-1,3-diphenylquinazolin-4-one, imidazo[1,2-*a*]quinazolin-5-one, pyrazolo[1,5-*a*]quinazolin-5-one, and [1,2,4]triazolo[1,5-*a*]quinazolin-5-one were developed on the basis of the reaction of tetrafluorobenzoyl chloride with *N*,*N'*-diphenylguanidine and aminoazoles.

Fused quinazolin-4-one derivatives attract strong interest from the viewpoint of their potential biological activity. Antibacterial, antitoxoplasmatic, antihypertensive, and antihistaminic agents, phosphodiesterase inhibitors, and compounds possessing other kinds of biological activity have been revealed among quinazolin-4-one derivatives [2–8]. A known procedure for building up such heterocyclic systems is based on cyclization of 2-halo-substituted benzoyl chlorides with difunctional nitrogen-centered nucleophiles [4, 9–11]. This approach was not applied previously to the synthesis of fused imidazo-, pyrazolo[*a*]- and triazolo[*a*]-quinazolinones. There are published data only on the preparation of triazolo[*a*]quinazolinone derivatives via transformations of quinazolinones having a sulfanyl, hydrazino, thiosemicarbazido, or *S*-methylisothiosemicarbazido group in the 2-position [2, 5, 12–15].

In continuation of our studies on the synthesis of fused fluorine-containing nitrogen heterocycles, we have developed a general procedure for the preparation of fluorinated triazolo-, pyrazolo-, and imidazo[a]-quinazolinones by reaction of tetrafluorobenzoyl chloride with difunctional N,N'-dinucleophiles. By acylation of *N*,*N'*-diphenylguanidine (**II**) with tetrafluorobenzoyl chloride (**I**) in boiling toluene we obtained *N*,*N'*-diphenyl-N-(2,3,4,5-tetrafluorobenzoyl)-guanidine (**III**) (Scheme 1). The $^1$H NMR spectrum of **III** confirmed the presence in its molecule of two

**Scheme 1.**




**V**, RR'N = 1-pyrrolidinyl (**a**), morpholino (**b**), 2,6-dimethylmorpholino (**c**), 4-methylpiperidino (**d**).

* For communication XI, see [1].

1070-4280/05/4107-1071 

**Table 1.** $^1$H NMR and mass spectra of compounds **IV** and **Va**–**Vd**

| Comp. no. | $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm ($J$, Hz) | | | | Mass spectrum, $m/z$ ($I_{rel}$, %) |
|---|---|---|---|---|---|
| | 5-H | NRR' | NH, br.s | Ph | |
| **IV** | 7.76 d.d.d ($^3J$ = 10.0, $^4J$ = 8.0, $^5J$ = 2.2) | – | 14.0–15.0 | 7.12 m (1H), 7.28 m (4H), 7.62 m (5H) | 367 (84) $[M]^+$, 366 (100), 250 (34), 240 (47), 221 (13), 220 (10), 77 (11) |
| **Va** | 7.40 d.d ($^3J$ = 12.4, $^5J$ = 2.0) | 1.81 m [4H, $(CH_2)_2$], 3.41 m [4H, $N(CH_2)_2$] | 14.0–15.0 | 7.08 m (1H), 7.25 m (2H), 7.32 m (2H), 7.58 m (5H) | 418 (100) $[M]^+$, 417 (99), 301 (13), 299 (21), 279 (15), 278 (25), 244 (11), 212 (18) |
| **Vb**[a] | 7.52 d.d ($^3J$ = 12.2, $^5J$ = 1.8) | 3.07 m [4H, $N(CH_2)_2$], 3.61 m [4H, $O(CH_2)_2$] | 14.0–15.0 | 7.09 m (1H), 7.23 m (4H), 7.59 m (5H) | 434 (100) $[M]^+$, 433 (73), 392 (25), 391 (28), 376 (13), 375 (30), 259 (13), 258 (29) |
| **Vc** | 7.48 d.d ($^3J$ = 11.5, $^5J$ = 1.6) | 1.03 m (3H, $CH_3$), 1.06 m (3H, $CH_3$), 2.72 m (2H, $CH_2$), 2.81 m (2H, $CH_2$), 3.60 m (2H, CH) | 14.0–15.0 | 7.09 m (1H), 7.28 m (4H), 7.59 m (5H) | 462 (83) $[M]^+$, 461 (35). 392 (89), 391 (100), 376 (46), 375 (67), 273 (19), 259 (23), 258 (33) |
| **Vd** | 7.47 d.d ($^3J$ = 11.3, $^5J$ = 1.5) | 0.92 d (3H, $CH_3$, $^3J$ = 6.3), 1.2–1.4 (2H, $CH_2$), 1.6 m (2H, CH), 1.7 m (2H, $CH_2$), 3.19 m (2H, $NCH_2$), 3.55 m (2H, $NCH_2$) | 14.0–15.0 | 7.08 m (1H), 7.31 m (4H), 7.59 m (5H) | 446 (100) $[M]^+$, 445 (87), 375 (6), 329 (7), 327 (8), 259 (6), 258 (7) |

[a] $^{19}$F NMR spectrum (DMSO-$d_6$), $\delta_F$, ppm: 34.23 m (1F), 38.25 m (1F).

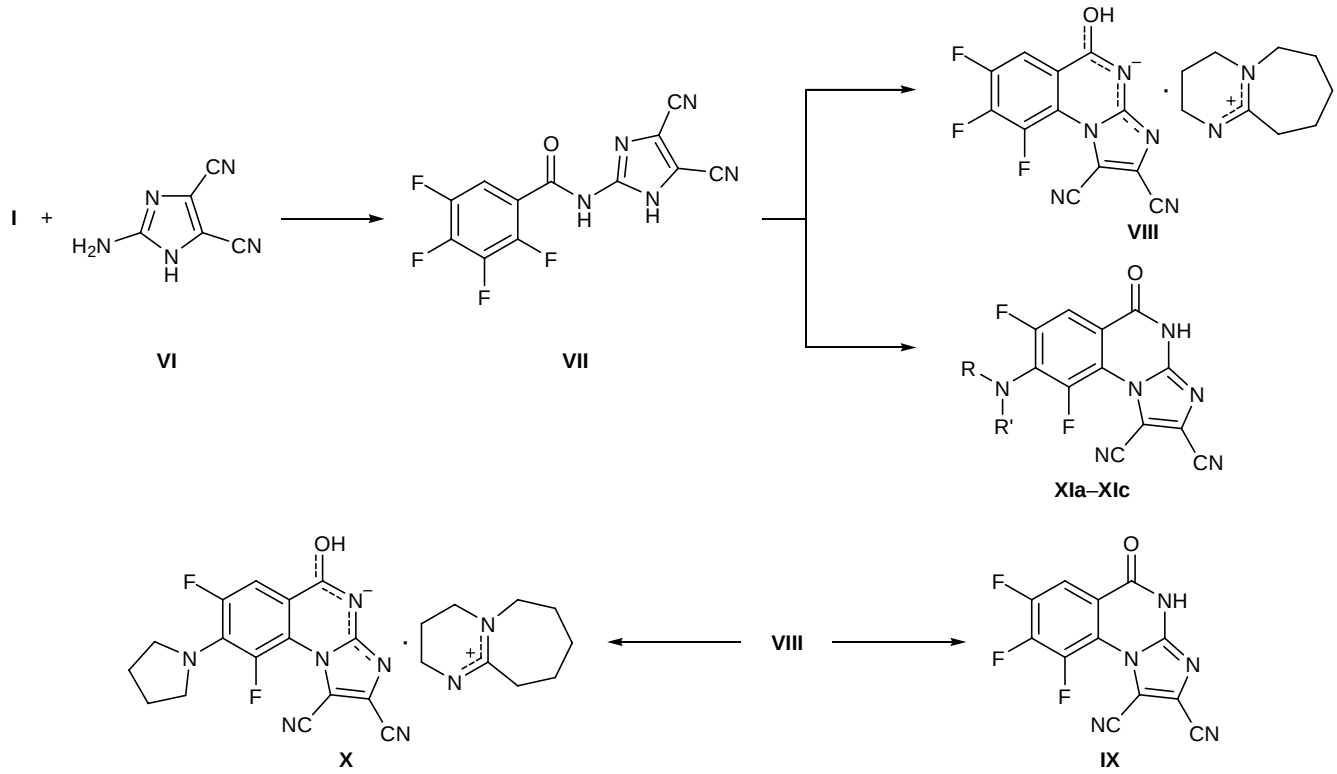
phenyl groups (multiplet signals in the region δ 7.3–8.0 ppm) and two NH groups (broadened signals at δ 10.8 and 12.1 ppm); also, the spectrum characteristically contained a signal at δ 6.82 ppm (m) belonging to proton in the tetrafluorobenzoyl fragment.

Heating of compound **III** for 5 h in boiling dimethylformamide resulted in cyclization with formation of 81% of 2-imino-1,3-diphenyl-2,3-dihydro-1*H*-quinazolin-4-one (**IV**). In the $^1$H NMR spectrum of **IV** we observed a broadened singlet from the NH proton, signals from protons in two phenyl rings, and a signal at δ 7.76 ppm from proton in the trifluorobenzoyl fragment; change of the multiplicity of the latter indicated that cyclization occurred (Table 1). The mass spectrum of quinazolinone **IV** contained a strong peak (84%) from the molecular ion peak, while the most abundant was $[M - 1]^+$ ion (100%). No other strong peaks were present in the spectrum, presumably due to high stability of the 2-imino-1,3-diphenyl-2,3-dihydro-1*H*-quinazolin-4-one system.

When compound **III** was heated in dimethylformamide in the presence of pyrrolidine, morpholine, *cis*-2,6-dimethylmorpholine, or 4-methylpiperidine, the intramolecular cyclization was accompanied by replacement of one fluorine atom with formation of compounds **Va**–**Vd**. Compound **Va** was also obtained by reaction of quinazolinone **IV** with pyrrolidine. The site of fluorine replacement (7-F) was determined on the basis of multiplicity of the 5-H signal in the $^1$H NMR spectra of **Va**–**Vd**: it appeared as a doublet of doublets at δ 7.40–7.52 ppm, $^3J$ = 11.5–12.5, $^5J$ = 1.5–2.0 Hz). In addition, the $^1$H NMR spectra of 7-substituted quinazolin-4-ones **Va**–**Vd** contained signals from the NH proton, protons in the R and R' substituents at the amino group, and protons in the *N*-phenyl substituents. Compounds **Va**–**Vd** showed strong molecular ion peaks in the mass spectra. The $^{19}$F NMR spectrum of **Vb** displayed signals from two fluorine atoms as an additional support to structure **V**.

Another difunctional *N*,*N'*-nucleophile was 2-aminoimidazole-4,5-dicarbonitrile (**VI**). Acylation of amine **VI** with tetrafluorobenzoyl chloride (**I**) in methylene chloride in the presence of triethylamine at room temperature afforded *N*-(4,5-dicyano-1*H*-imidazol-2-yl)-2,3,4,5-tetrafluorobenzamide (**VII**) (Scheme 2). The $^1$H NMR spectrum of **VII** contained broadened singlets from the two NH protons at δ 10.3 and 12.6 ppm and a multiplet signal from the tetrafluorobenzoyl

**Scheme 2.**




**XI**, RR'N = 1-pyrrolidinyl (**a**), morpholino (**b**); R = R' = Me (**c**).

proton. By heating amide **VII** in acetonitrile in the presence of 1,8-diazabicycloundec-7-ene (DBU) we obtained 7,8,9-trifluoro-5-oxo-4,5-dihydroimidazo-[1,2-*a*]quinazoline-1,2-dicarbonitrile salt **VIII** with protonated DBU molecule as cation. Salt **VIII** did not change on treatment with a solution of acetic acid at room temperature. The multiplicity of the 6-H signal (Table 2) is indicative of the fused polycyclic structure of **VIII**; eight methylene groups of DBU give signals in a strong field.

By heating salt **VIII** for a short time in boiling acetic acid we succeeded in isolating imidazo[*a*]-quinazolinone **IX**. The $^1$H NMR spectrum of **IX** lacked signals from methylene protons typical of DBU, the 6-H signal was a double doublet of doublets, which was displaced downfield by 0.3 ppm, and the NH signal was displaced downfield by 4.2 ppm. The $^{19}$F NMR spectrum of **IX** contained signals (d.d.d) from three fluorine atoms (Table 2).

When compounds **VIII** was heated with pyrrolidine in boiling dimethylformamide, the 8-F atom was replaced by the cyclic amine residue, and the product (like the initial compound) was a salt with DBU. In the $^1$H NMR spectrum of **X** we observed a doublet of doublets ($^3J$ = 12.1, $^5J$ = 1.5 Hz) from the 6-H proton, a broadened singlet from the NH proton, and multiplet signals from methylene groups in the pyrrolidine and DBU fragments. Reactions of amide **VII** with pyrrolidine and morpholine in boiling dimethylformamide resulted in intramolecular ring closure with simultaneous replacement of the 8-F atom with formation of compounds **XIa** and **XIb**, respectively. Our attempt to isolate imidazoquinazolinone **IX** by heating amide **VII** in dimethylformamide was unsuccessful: the process was accompanied by substitution of the 8-F atom by dimethylamine residue to afford compound **XIc**. The structure of **XIa**–**XIc** was confirmed by the $^1$H NMR and mass spectra (Table 2).

3-Aminopyrazoles **XIIa** and **XIIb** are also accessible *N*,*N'*-nucleophiles. Compound (**I**) reacted with 3-aminopyrazole (**XIIa**) in toluene under reflux (reaction time 3 h) to give bis-aroyl derivative **XVI** (Scheme 3). Compound **XVI** showed in the $^1$H NMR spectrum multiplet signals from two protons in the tetrafluorobenzoyl fragments, a broadened singlet from one NH proton, and signals from two protons in the pyrazole ring. The mass spectrum of **XVI** was consistent with the assumed structure (see Experimental).

**Table 2.** $^1$H NMR and mass spectra of compounds **VIII**–**XI**

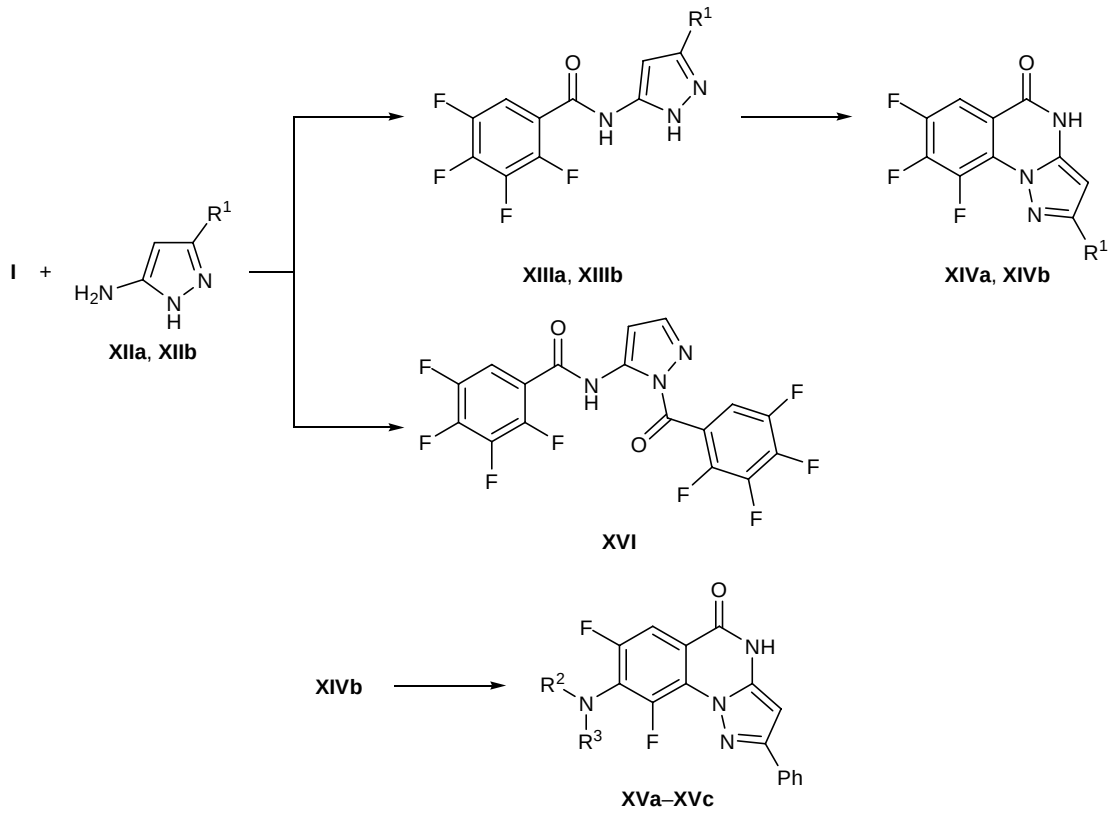
| Comp. no. | NMR spectrum $^1$H (DMSO-$d_6$), δ, ppm ($J$, Hz) | | | | Mass spectrum, $m/z$ ($I_{rel}$, %) |
|---|---|---|---|---|---|
| | 6-H | NH | NRR' | other signals | |
| **VIII** | 7.90 d.d.d ($^3J$ = 10.1, $^4J$ = 8.2, $^5J$ = 2.0) | 9.9 br.s | – | 1.70 m (6H, $3CH_2$), 2.00 m (2H, $CH_2$), 2.75 m (2H, $CH_2$), 3.35 m (2H, $CH_2$), 3.54 m (2H, $CH_2$), 3.60 m (2H, $CH_2$) | 289 (100) [$M$ – DBU]$^+$, 183 (12), 158 (12), 152 (37), 151 (35), 137 (13), 130 (16), 123 (19), 98 (16), 96 (24), 55 (12) |
| **IX**[a] | 8.20 d.d.d ($^3J$ = 9.9, $^4J$ = 7.9, $^5J$ = 2.2) | 14.1 br.s | – | – | |
| **X** | 7.54 d.d ($^3J$ = 12.1, $^5J$ = 1.5) | 10.3 br.s | 1.95 m [4H, $(CH_2)_2$], 3.66 m [4H, $N(CH_2)_2$] | 1.72 m (6H, $3CH_2$), 1.93 m (2H, $CH_2$), 2.78 m (2H, $CH_2$), 3.37 m (2H, $CH_2$), 3.52 m (2H, $CH_2$), 3.56 m (2H, $CH_2$) | |
| **XIa** | 7.63 d.d ($^3J$ = 12.1, $^5J$ = 1.5) | 13.2 br.s | 1.92 m [4H, $(CH_2)_2$], 3.70 m [4H, $N(CH_2)_2$] | – | 340 (88) [$M$]$^+$, 339 (100), 297 (20), 285 (13), 284 (26) |
| **XIb** | 7.78 d.d ($^3J$ = 11.7, $^5J$ = 1.5) | 13.5 br.s | 3.35 m [4H, $N(CH_2)_2$], 3.75 m [4H, $O(CH_2)_2$] | – | 356 (81) [$M$]$^+$, 314 (56), 313 (65), 299 (25), 298 (100), 297 (20), 271 (21), 149 (12), 133 (12) |
| **XIc** | 7.76 d.d ($^3J$ = 12.3, $^5J$ = 1.8) | 11.6 br.s | 3.07 s (6H, $3CH_3$) | – | 313 (100) [$M$]$^+$, 289 (44), 156 (11) |

[a] $^{19}$F NMR spectrum (DMSO-$d_6$), δ, ppm: 14.27 d.d.d (1F, 8-F, $^3J_{FF}$ = 22.4, $^3J_{FF}$ = 21.2, $^4J_{FH}$ = 7.5 Hz), 29.96 d.d.d (1F, 7-F, $^3J_{FF}$ = 22.4, $^3J_{FH}$ = 9.5, $^4J_{FF}$ = 6.7 Hz), 31.38 d.d.d (1F, 9-F, $^3J_{FF}$ = 21.2, $^4J_{FF}$ = 6.7, $^5J_{FH}$ = 1.7 Hz).

**Table 3.** $^1$H and $^{19}$F NMR and mass spectra of compounds **XIVa**, **XIVb**, and **XVa**–**XVc**

| Comp. no. | $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm ($J$, Hz) | | | | | $^{19}$F NMR spectrum (DMSO-$d_6$), δ, ppm | Mass spectrum, $m/z$ ($I_{rel}$, %) |
|---|---|---|---|---|---|---|---|
| | 6-H | 3-H | NH | $R^1$ | $NR^2R^3$ | | |
| **XIVa** | 7.93 d.d.d ($^3J$ = 10.0, $^4J$ = 8.0, $^5J$ = 2.1) | 6.09 d ($^3J$ = 2.0) | 12.3 br.s | 7.90 d ($^3J$ = 2.0) | – | 11.00 m (1F), 1.84 m (1F), 19.26 m (1F) | 239 (100) [$M$]$^+$, 183 (16), 182 (11), 157 (10), 156 (21), 130 (12) |
| **XIVb** | 7.47 d.d.d ($^3J$ = 9.9, $^4J$ = 7.7, $^5J$ = 1.5) | 6.59 s | 12.8 br.s | 7.51 m (2H), 8.04 m (3H) | – | 11.01 m (1F), 2.07 m (1F), 19.41 m (1F) | 315 (100) [$M$]$^+$, 287 (9), 258 (31), 102 (12) |
| **XVa** | 7.58 d.d ($^3J$ = 12.6, $^5J$ = 1.7) | 6.45 s | 12.1 br.s | 7.45 m (1H), 7.51 m (2H), 7.98 m (2H) | 1.90 m [4H, $(CH_2)_2$], 3.70 m [4H, $N(CH_2)_2$] | | 366 (100) [$M$]$^+$, 365 (42), 182 (13) |
| **XVb** | 7.70 d.d ($^3J$ = 12.4, $^5J$ = 1.7) | 6.51 s | 12.3 br.s | 7.67 m (3H), 7.99 m (2H) | 3.36 m [4H, $N(CH_2)_2$], 3.74 m [4H, $O(CH_2)_2$] | | 382 (100) [$M$]$^+$, 341 (15), 340 (73), 324 (43), 297 (20), 296 (14), 267 (11), 162 (16), 148 (16) |
| **XVc** | 7.71 d.d ($^3J$ = 12.1, $^5J$ = 1.5) | 6.52 s | 11.9 br.s | 7.44 m (1H), 7.51 m (2H), 8.01 m (2H) | 1.22 t (3H, $CH_3$), 3.34 m [4H, $N(CH_2)_2$], 3.54 m [4H, $N(CH_2)_2$], 4.09 q (2H, $OCH_2$) | | 453 (100) [$M$]$^+$, 433 (18), 351 (30), 340 (48), 339 (50), 338 (23), 325 (12), 324 (11), 237 (15), 102 (11), 56 (58) |

**Scheme 3.**



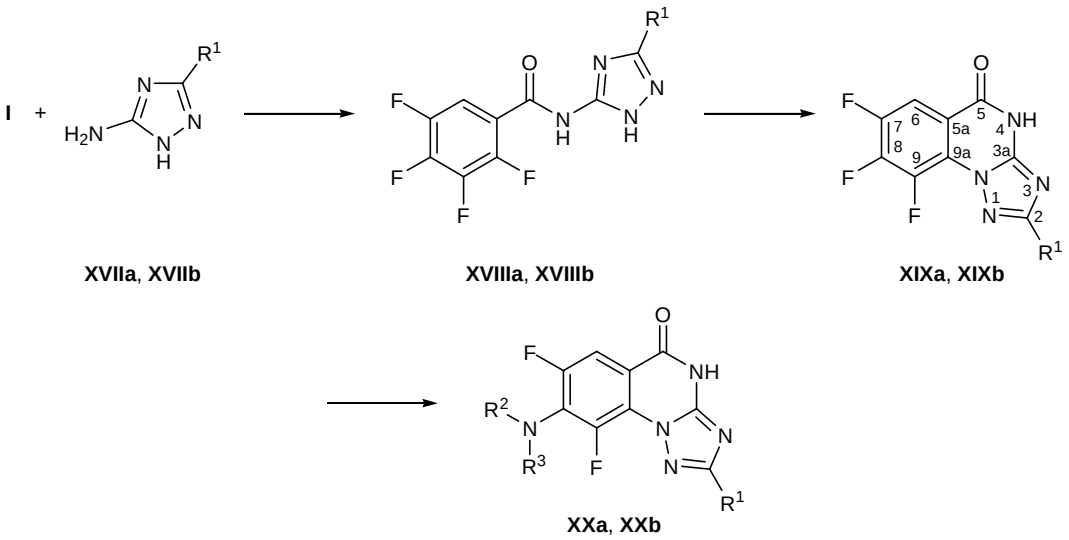

**XII**–**XIV**, $R^1$ = H (**a**), Ph (**b**); **XV**, $R^2R^3N$ = 1-pyrrolidinyl (**a**), morpholino (**b**), 4-ethoxycarbonylpiperazin-1-yl (**c**).

The reaction of tetrafluorobenzoyl chloride (**I**) with 3-aminopyrazole (**XIIa**) in the presence of triethylamine in methylene chloride at room temperature (reaction time 24 h) afforded monoaroyl derivative **XIIIa**. Unlike compound **XVI**, the $^1$H NMR spectrum of **XIIIa** displayed only one tetrafluorobenzoyl proton signal, and two NH protons gave a broadened singlet at about δ 5.6 ppm. By acylation of 3-amino-5-phenylpyrazole (**XIIb**) with tetrafluorobenzoyl chloride (**I**) in boiling toluene (3 h) we obtained amide **XIIIb**. In the $^1$H NMR spectrum of **XIIIb**, broadened signals from the NH protons appeared in a much weaker field (δ 11.0–13.0 ppm), as compared to the corresponding signals of **XIIIa**.

Heating of amides **XIIIa** and **XIIIb** in acetonitrile in the presence of DBU (3 h, 80°C) led to formation of pyrazoloquinazolinones **XIVa** and **XIVb**, respectively. The $^1$H NMR spectra of compounds **XIVa** and **XIVb** contained a double doublet of doublets from the 6-H proton, and signals from three fluorine atoms were present in their $^{19}$F NMR spectra (Table 3). Compound **XIVb** was brought into reactions with pyrrolidine, morpholine, and ethyl piperazine-1-carboxylate in boiling dimethylformamide (reaction time 5 h) to obtain derivatives **XVa**–**XVc**. In the mass spectra of **XVa**–**XVc**, the molecular ion was the most abundant. Replacement of the 8-F atom in pyrazoloquinazolinone **XIVb** follows from the presence in the $^1$H NMR spectra of **XVa**–**XVc** of a characteristic doublet of doublets from the 6-H proton ($^3J$ = 12.1–12.6, $^5J$ = 1.5–1.7 Hz) and signals from protons in the corresponding amine fragment (Table 3).

Acylation of 3-amino-1,2,4-triazoles **XVIIa** and **XVIIb** with tetrafluorobenzoyl chloride (**I**) in boiling toluene (2 h) gave amides **XVIIIa** and **XVIIIb** (Scheme 4) whose structure was confirmed by the $^1$H NMR spectra. Intramolecular cyclization of aroyl derivatives **XVIIIa** and **XVIIIb** to 7,8,9-trifluoro-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-ones **XIXa** and **XIXb** was effected by heating for 5 h in boiling acetonitrile in the presence of DBU. The yield of the cyclic product from trifluoromethyl-substituted derivative **XVIIIb** was considerably greater (76%) than from compound **XVIIIa** (32%). Triazoloquinazolinones **XIXa** and **XIXb** showed in the $^1$H NMR spectra a signal from the 6-H proton (d.d.d) and a broadened

**Scheme 4.**




**XVII**–**XIX**, $R^1$ = H (**a**), $CF_3$ (**b**); **XX**, $R^2R^3N$ = 1-pyrrolidinyl (**a**), morpholino (**b**).

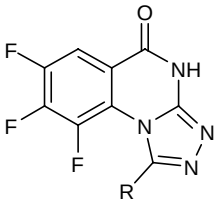
one-proton singlet from the NH group; the molecular ion peak in the mass spectra of **XIXa** and **XIXb** had the maximal intensity (Table 4). Signals from three fluorine atoms were present in the $^{19}F$ NMR spectra of **XIXa** and **XIXb** (d.d.d); in the $^{19}F$ NMR spectrum of **XIXb**, a singlet from the trifluoromethyl group was also observed at $\delta_F$ 97.8 ppm.

It should be noted that the above spectral data are equally consitent with structure **XIX** and alternative 4*H*-[1,2,4]triazolo[4,3-*a*]quinazolin-5-one structure **XXI**. To distinguish between these alternative structures, we analyzed the $^{13}C$ NMR spectra of **XIXa** and **XIXb** (see Experimental). The chemical shifts of $C^{3a}$ and $C^5$–$C^{9a}$ in these compounds have similar values, and the corresponding signals have similar multiplicities. A difference was observed in the multiplicities of the $C^2$ signal: it was a quartet of doublets in the spectrum of **XIXb** ($J$ = 40.5, 3.5 Hz) and a doublet ($J$ = 4.0 Hz) in the spectrum of **XIXa**. The coupling constant equal to 3.5–4.0 Hz may be assigned to inter-

**Table 4.** $^1H$ and $^{19}F$ NMR and mass spectra of compounds **XIXa**, **XIXb**, **XXa**, and **XXb**

| Comp. no. | $^1H$ NMR spectrum (DMSO-$d_6$), δ, ppm ($J$, Hz) | | | | $^{19}F$ NMR spectrum (DMSO-$d_6$), δ, ppm ($J$, Hz) | Mass spectrum, $m/z$ ($I_{rel}$, %) |
|---|---|---|---|---|---|---|
| | 6-H | NH | $R^1$ | $NR^2R^3$ | | |
| **XIXa** | 8.06 d.d.d ($^3J$ = 10.0, $^4J$ = 7.7, $^5J$ = 2.3) | 13.4 br.s | 8.22 s | – | 13.28 d.d.d (1F, 3-F, $^3J_{FF}$ = 22.5, 20.1, $^4J_{FH}$ = 7.7), 19.90 d.d.d (1F, 9-F, $^3J_{FF}$ = 20.1, $^4J_{FF}$ = 4.6, $^5J_{FH}$ = 2.3), 26.22 d.d.d (1F, 7-F, $^3J_{FF}$ = 22.5, $^3J_{FH}$ = 10.0, $^4J_{FF}$ = 4.6) | 240 (100) $[M]^+$, 212 (9), 188 (9), 157 (17), 130 (14) |
| **XIXb** | 8.15 d.d.d ($^3J$ = 9.9, $^4J$ = 7.7, $^5J$ = 2.2) | 13.8 br.s | – | – | 14.06 d.d.d (1F, 8-F, $^3J_{FF}$ = 22.8, 20.2, $^4J_{FH}$ = 7.7), 19.96 d.d.d (1F, 9-F, $^3J_{FF}$ = 20.2, $^4J_{FF}$ = 5.4, $^5J_{FH}$ = 2.2), 28.23 d.d.d (1F, 7-F, $^3J_{FF}$ = 22.8, $^3J_{FH}$ = 9.9, $^4J_{FF}$ = 5.4), 97.79 s (3F, $CF_3$) | 308 (100) $[M]^+$, 280 (11), 183 (10), 158 (14), 144 (13), 130 (16), 69 (12), 53 (18) |
| **XXa** | 7.61 d.d ($^3J$ = 13.9, $^5J$ = 1.5) | 13.3 br.s | – | 1.91 m [4H, $(CH_2)_2$], 3.72 m [4H, $N(CH_2)_2$] | | 359 (89) $[M]^+$, 358 (100), 317 (11), 316 (27), 303 (51), 179 (10) |
| **XXb** | 7.74 d.d ($^3J$ = 12.4, $^5J$ = 1.6) | 13.5 br.s | – | 3.37 m [4H, $N(CH_2)_2$], 3.74 m [4H, $O(CH_2)_2$] | | |

action between the $C^2$ and 9-F nuclei. Such carbon–fluorine couplings between atoms located in different rings have been reported [16].



**XXIa**, **XXIb**

An additional support to structure **XIX** was obtained by studying intramolecular cyclization of 3-azolylamino-2-polyfluorobenzoylacrylates [17]. Furthermore, the $^{19}F$ NMR spectrum of cyclic compound **XXIb** should reveal coupling between 9-F and the trifluoromethyl group (an analogous interaction was discussed in [18]). According to the data of [19–22], the $N^2$ rather than $N^4$ atom of the 1,2,4-triazole ring is preferentially involved in intramolecular cyclizations.

## EXPERIMENTAL

The $^1H$ NMR spectra were recorded on Bruker WM-250 and Bruker DRX-400 spectrometers at 250.14 and 400.13 MHz, respectively. The $^{19}F$ NMR spectra were obtained on a Bruker DRX-400 spectrometer at 376.45 MHz. The chemical shifts were measured relative to tetramethylsilane ($^1H$) and hexafluorobenzene ($^{19}F$). The $^{13}C$ NMR spectra were recorded on a Bruker DRX-400 spectrometer at 100.61 MHz. The mass spectra (electron impact, 70 eV) were run on a Varian MAT 311A instrument (accelerating voltage 3 kV, cathode emission current 300 μA, direct sample admission into the ion source).

***N*,*N*′-Diphenyl-*N*-(2,3,4,5-tetrafluorobenzoyl)-guanidine (III).** Tetrafluorobenzoyl chloride (**I**), 0.74 g (3.5 mmol), was added to a suspension of 0.5 g (2.4 mmol) of *N*,*N*′-diphenylguanidine (**II**) in 10 ml of anhydrous toluene. The mixture was heated for 3 h under reflux and cooled, and the colorless precipitate was filtered off and recrystallized from DMSO. Yield 0.76 g (82%), mp 182–184°C. $^1H$ NMR spectrum (DMSO-$d_6$), δ, ppm: 6.82 m (1H, $C_6HF_4$), 7.31 m (4H), 7.41 m (3H), 7.55 m (2H), 7.93 m (1H), 10.8 br.s (1H, NH), 12.1 br.s (1H, NH). Found, %: C 61.86; H 3.49; N 10.98. $C_{20}H_{13}F_4N_3O$. Calculated, %: C 62.02; H 3.38; N 10.85.

**6,7,8-Trifluoro-2-imino-1,3-diphenyl-2,3-dihydro-1*H*-quinazolin-4-one (IV).** A solution of 0.45 g (1.2 mmol) of compound **III** in 5 ml of dimethylformamide was heated for 5 h under reflux. The mixture was cooled and diluted with 15 ml of water, and the colorless precipitate was filtered off and recrystallized from acetonitrile. Yield 0.35 g (81%), mp 242–244°C. Found, %: C 65.47; H 3.18; N 11.35. $C_{20}H_{12}F_3N_3O$. Calculated, %: C 65.39; H 3.29; N 11.44.

**6,8-Difluoro-2-imino-1,3-diphenyl-7-(1-pyrrolidinyl)-2,3-dihydro-1*H*-quinazolin-4-one (Va).** *a*. Pyrrolidine, 0.13 g (1.8 mmol), was added to a solution of 0.17 g (0.44 mmol) of compound **III** in 3 ml of dimethylformamide. The mixture was heated for 5 h under reflux, cooled, and diluted with 10 ml of water. The colorless precipitate was filtered off and recrystallized from acetonitrile. Yield 0.15 g (83%), mp 213–215°C. Found, %: C 69.02; H 4.91; N 13.27. $C_{24}H_{20}F_2N_4O$. Calculated, %: C 68.89; H 4.82; N 13.39.

*b*. Pyrrolidine, 0.23 g (3.2 mmol), was added to a solution of 0.3 g (0.8 mmol) of compound **IV** in 4 ml of dimethylformamide. The mixture was heated for 5 h under reflux, cooled, and diluted with 12 ml of water. The colorless precipitate was filtered off and recrystallized from acetonitrile. Yield 0.25 g (76%).

Compounds **Vb**–**Vd** were synthesized in a similar way (method *a*).

**6,8-Difluoro-2-imino-7-morpholino-1,3-diphenyl-2,3-dihydro-1*H*-quinazolin-4-one (Vb).** Yield 78%, mp 259–261°C. Found, %: C 66.43; H 4.71; N 12.79. $C_{24}H_{20}F_2N_4O_2$. Calculated, %: C 66.35; H 4.64; N 12.90.

**7-(2,6-Dimethylmorpholino)-6,8-difluoro-2-imino-1,3-diphenyl-2,3-dihydro-1*H*-quinazolin-4-one (Vc).** Yield 88%, mp 264–266°C. Found, %: C 67.40; H 5.31; N 12.18. $C_{26}H_{24}F_2N_4O_2$. Calculated, %: C 67.52; H 5.23; N 12.11.

**6,8-Difluoro-2-imino-7-(4-methylpiperidino)-1,3-diphenyl-2,3-dihydro-1*H*-quinazolin-4-one (Vd).** Yield 90%, mp 190–192°C. Found, %: C 70.05; H 5.30; N 12.46. $C_{26}H_{24}F_2N_4O$. Calculated, %: C 69.94; H 5.42; N 12.55.

***N*-(4,5-Dicyano-1*H*-imidazol-2-yl)-2,3,4,5-tetrafluorobenzamide (VII).** Triethylamine, 0.6 g (6 mmol), and tetrafluorobenzoyl chloride (**I**), 0.63 g (3 mmol), were added to a suspension of 0.4 g (3 mmol) of 2-aminoimidazole-4,5-dicarbonitrile (**VI**) in 5 ml of dry methylene chloride. The mixture was kept for 24 h at room temperature and evaporated, the residue was washed with water, and the light brown precipitate was recrystallized from ethanol. Yield 0.8 g

(86%), mp 162–164°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 7.52 m (1H, $C_6HF_4$), 10.3 br.s and 12.6 br.s (1H each, NH). Found, %: C 46.53; H 1.01; N 22.77. $C_{12}H_3F_4N_5O$. Calculated, %: C 46.62; H 0.98; N 22.65.

**1-Aza-8-azoniabicyclo[5.4]undec-7-ene 1,2-dicyano-7,8,9-trifluoro-4,5-dihydroimidazo[1,2-*a*]quinazolin-5-olate (VIII).** 1,8-Diazabicyclo[5.4]undec-7-ene, 0.26 g (2 mmol), was added to a solution of 0.5 g (1.6 mmol) of amide **VII** in 10 ml of anhydrous acetonitrile. The mixture was heated for 3 h and evaporated, and the residue was treated with 15 ml of water and 0.5 ml of acetic acid. The precipitate was filtered off and recrystallized from ethanol. Yield 0.55 g (77%), mp 236–238°C. Found, %: C 57.21; H 4.04; N 22.15. $C_{21}H_{18}F_3N_7O$. Calculated, %: C 57.14; H 4.11; N 22.22.

**7,8,9-Trifluoro-5-oxo-4,5-dihydroimidazo[1,2-*a*]quinazoline-1,2-dicarbonitrile (IX).** Acetic acid, 3 ml, was added to 0.57 g (1.3 mmol) of salt **VIII**, the solution was heated to the boiling point, 12 ml of water was added, and the precipitate was filtered off and recrystallized from ethanol. Yield 0.33 g (87%), mp 248–250°C. Found, %: C 49.92; H 0.74; N 24.16. $C_{12}H_2F_3N_5O$. Calculated, %: C 49.84; H 0.70; N 24.22.

**1-Aza-8-azoniabicyclo[5.4]undec-7-ene 1,2-dicyano-7,9-trifluoro-8-(1-pyrrolidinyl)-4,5-dihydroimidazo[1,2-*a*]quinazolin-5-olate (X).** Pyrrolidine, 0.17 g (2.4 mmol), was added to a solution of 0.25 g (0.6 mmol) of compound **VIII** in 4 ml of dimethylformamide. The mixture was heated for 5 h under reflux and cooled, and the precipitate was filtered off and recrystallized from DMSO. Yield 0.22 g (73%), mp 272–274°C. Found, %: C 61.07; H 5.25; N 22.68. $C_{25}H_{26}F_2N_8O$. Calculated, %: C 60.96; H 5.32; N 22.76.

**7,9-Difluoro-5-oxo-8-(1-pyrrolidinyl)-4,5-dihydroimidazo[1,2-*a*]quinazoline-1,2-dicarbonitrile (XIa).** Pyrrolidine, 0.65 g (9.2 mmol), was added to a solution of 0.7 g (2.3 mmol) of amide **VII** in 6 ml of dimethylformamide. The mixture was heated for 5 h under reflux, cooled, and diluted with 15 ml of water, and the precipitate was filtered off and recrystallized from acetonitrile. Yield 0.68 g (87%), mp 263–265°C. Found, %: C 56.59; H 3.04; N 24.61. $C_{16}H_{10}F_2N_6O$. Calculated, %: C 56.47; H 2.96; N 24.70.

**7,9-Difluoro-8-morpholino-5-oxo-4,5-dihydroimidazo[1,2-*a*]quinazoline-1,2-dicarbonitrile (XIb)** was synthesized in a similar way. Yield 85%, mp 218–220°C. Found, %: C 54.02; H 2.89; N 23.48. $C_{16}H_{10}F_2N_6O_2$. Calculated, %: C 53.94; H 2.83; N 23.59.

**8-Dimethylamino-7,9-difluoro-5-oxo-4,5-dihydroimidazo[1,2-*a*]quinazoline-1,2-dicarbonitrile (XIc).** A solution of 0.7 g (2.3 mmol) of amide **VII** in 6 ml of moist dimethylformamide was heated for 5 h under reflux. The mixture was cooled and diluted with 15 ml of water, and the precipitate was filtered off and recrystallized from acetonitrile. Yield 0.62 g (86%), mp 244–246°C. Found, %: C 53.39; H 2.47; N 26.82. $C_{14}H_8F_2N_6O$. Calculated, %: C 53.51; H 2.56; N 26.74.

***N*-(1*H*-Pyrazol-5-yl)-2,3,4,5-tetrafluorobenzamide (XIIIa).** Tetrafluorobenzoyl chloride (**I**), 1.3 g (6 mmol), was added to a solution of 0.5 g (6 mmol) of 3-aminopyrazole (**XIIa**) in 5 ml of dry methylene chloride, and the mixture was kept for 24 h at room temperature. The precipitate was filtered off, washed with water, and recrystallized from ethanol. Yield 1.1 g (71%), mp 145–147°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 5.62 br.s (2H, NH), 6.08 d (1H, 4-H, $^3J$ = 3.0 Hz), 7.65 m (1H, 6'-H), 8.09 d (1H, 5-H, $^3J$ = 3.0 Hz). Found, %: C 46.42; H 2.01; N 16.16. $C_{10}H_5F_4N_3O$. Calculated, %: C 46.33; H 1.93; N 16.22.

***N*-(5-Phenyl-2*H*-pyrazol-3-yl)-2,3,4,5-tetrafluorobenzamide (XIIIb).** Tetrafluorobenzoyl chloride (**I**), 1.0 g (4.5 mmol), was added to a suspension of 0.5 g (3.1 mmol) of 3-amino-5-phenylpyrazole (**XIIb**) in 12 ml of anhydrous toluene. The mixture was heated for 3 h under reflux and cooled, and the precipitate was filtered off and recrystallized from DMSO. Yield 0.84 g (84%), mp 223–225°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 6.59 s (1H, 4-H), 7.51 m (3H, Ph), 7.94 m (1H, 6'-H), 8.04 m (2H, Ph), 11.3 br.s (1H, NH), 12.8 br.s (1H, NH). Found, %: C 55.85; H 2.90; N 12.89. $C_{15}H_9F_4N_3O$. Calculated, %: C 55.74; H 2.81; N 13.00.

**7,8,9-Trifluoro-4*H*-pyrazolo[1,5-*a*]quinazolin-5-one (XIVa).** 1,8-Diazabicyclo[5.4]undec-7-ene, 0.3 g (2.5 mmol), was added to a solution of 0.55 g (2.1 mmol) of amide **XIIIa** in 10 ml of anhydrous acetonitrile, and the mixture was heated for 3 h at 80°C. The mixture was evaporated, the residue was treated with 10 ml of water and 1 ml of acetic acid, and the precipitate was filtered off and recrystallized from ethanol. Yield 0.4 g (80%), mp 302–304°C. Found, %: C 50.18; H 1.94; N 17.33. $C_{10}H_4F_3N_3O$. Calculated, %: C 50.09; H 1.83; N 17.42.

**7,8,9-Trifluoro-2-phenyl-4*H*-pyrazolo[1,5-*a*]quinazolin-5-one (XIVb)** was synthesized in a similar way. The product was recrystallized from DMSO. Yield 82%, mp 284–286°C. Found, %: C 61.04; H 2.68; N 13.21. $C_{16}H_{18}F_3N_3O$. Calculated, %: C 60.96; H 2.56; N 13.33.

**7,9-Difluoro-2-phenyl-8-(1-pyrrolidinyl)-4*H*-pyrazolo[1,5-*a*]quinazolin-5-one (XVa).** Pyrrolidine, 0.45 g (6.4 mmol), was added to a solution of 0.5 g (1.6 mmol) of compound **XIVb** in 5 ml of dimethylformamide. The mixture was heated for 5 h under reflux and cooled, and the precipitate was filtered off and recrystallized from DMSO. Yield 0.48 g (81%), mp 310–312°C. Found, %: C 65.48; H 4.29; N 15.32. $C_{20}H_{16}F_2N_4O$. Calculated, %: C 65.57; H 4.40; N 15.29.

Compounds **XVb** and **XVc** were synthesized in a similar way.

**7,9-Difluoro-8-morpholino-2-phenyl-4*H*-pyrazolo[1,5-*a*]quinazolin-5-one (XVb)**. Yield 77%, mp 294–296°C. Found, %: C 62.87; H 4.28; N 14.60. $C_{20}H_{16}F_2N_4O_2$. Calculated, %: C 62.82; H 4.22; N 14.65.

**Ethyl 4-(7,9-difluoro-5-oxo-2-phenyl-4*H*-pyrazolo[1,5-*a*]quinazolin-8-yl)piperazine-1-carboxylate (XVc).** Yield 79%, mp 286–288°C. Found, %: C 61.00; H 4.72; N 15.38. $C_{23}H_{21}F_2N_5O_3$. Calculated, %: C 60.92; H 4.67; N 15.44.

***N*-[1-(2,3,4,5-Tetrafluorobenzoyl)pyrazol-5-yl]-2,3,4,5-tetrafluorobenzamide (XVI).** Tetrafluorobenzoyl chloride (**I**), 6.3 g (30 mmol), was added to a solution of 1.25 g (15 mmol) of 3-aminopyrazole (**XIIa**) in 12 ml of anhydrous toluene. The mixture was heated for 3 h under reflux and cooled, and the precipitate was filtered off and recrystallized from ethanol. Yield 4.6 g (71%), mp 103–105°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 7.18 d (1H, 4-H, $^3J$ = 3.0 Hz), 7.57 m (1H, $C_6HF_4$), 7.78 m (1H, $C_6HF_4$), 8.48 d (1H, 3-H, $^3J$ = 3.0 Hz), 11.52 br.s (1H, NH). Mass spectrum, *m*/*z* ($I_{rel}$, %): 435 $[M]^+$ (13), 239 (12), 177 (100), 149 (25). Found, %: C 46.92; H 1.18; N 9.59. $C_{17}H_5F_8N_3O_2$. Calculated, %: C 46.87; H 1.15; N 9.65.

***N*-(1*H*-1,2,4-Triazol-5-yl)-2,3,4,5-tetrafluorobenzamide (XVIIIa).** Tetrafluorobenzoyl chloride (**I**), 5.2 g (24 mmol), was added to a suspension of 1.6 g (19 mmol) of 5-amino-1,2,4-triazole **XVIIa** in 20 ml of anhydrous toluene. The mixture was heated for 2 h under reflux and cooled, and the colorless precipitate was filtered off and recrystallized from chloroform. Yield 3.8 g (78%), mp 245–247°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 7.78 s (1H, 5-H), 8.07 m (1H, $C_6HF_4$), 11.7 br.s (1H, NH), 13.6 br.s (1H, NH). Found, %: C 41.61; H 1.60; N 21.47. $C_9H_4F_4N_4O$. Calculated, %: C 41.55; H 1.55; N 21.54.

***N*-(3-Trifluoromethyl-1*H*-1,2,4-triazol-5-yl)-2,3,4,5-tetrafluorobenzamide (XVIIIb)** was synthesized in a similar way. Yield 74%, mp 288–290°C. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm: 7.90 m (1H, $C_6HF_4$), 12.6 br.s (1H, NH), 13.8 br.s (1H, NH). Found, %: C 36.47; H 1.00; N 16.99. $C_{10}H_3F_7N_4O$. Calculated, %: C 36.59; H 0.91; N 17.08.

**7,8,9-Trifluoro-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-one (XIXa).** 1,8-Diazabicyclo[5.4]undec-7-ene, 0.32 g (2.1 mmol), was added to a suspension of 0.6 g (2.1 mmol) of amide **XVIIIa** in 15 ml of anhydrous acetonitrile. The mixture was heated for 5 h under reflux and evaporated, the residue was treated with 15 ml of water and 1 ml of acetic acid, and the colorless precipitate was filtered off. The product was washed with hot ethanol, and the residue was recrystallized from DMSO. Yield 0.16 g (32%), mp 280–282°C. $^{13}$C NMR spectrum ($CD_3COOD$), δ, ppm ($J_{C,F}$, Hz): 113.16 d.d ($C^6$, $J$ = 20.8, 3.9), 114.85 d.d ($C^{5a}$, $J$ = 7.5, 3.2), 124.46 d.d ($C^{9a}$, $J$ = 7.5, 3.0), 142.03 d.d.d ($C^9$, $J$ = 262.5, 14.1, 2.6), 146.42 d.d ($C^8$, $J$ = 259.8, 13.6), 149.56 s ($C^{3a}$), 150.31 d.d.d ($C^7$, $J$ = 251.9, 11.4, 1.0), 152.40 d ($C^2$, $J$ = 4.0), 159.62 m ($C^5$). Found, %: C 44.92; H 1.21; N 23.38. $C_9H_3F_3N_4O$. Calculated, %: C 45.01; H 1.26; N 23.34. Evaporation of the ethanol solution afforded unreacted amide **XVIIIa**.

**7,8,9-Trifluoro-2-trifluoromethyl-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-one (XIXb)** was synthesized in a similar way. After treatment with an aqueous solution of acetic acid, the whole precipitate was recrystallized from ethanol. Yield 76%, mp 215–217°C. $^{13}$C NMR spectrum ($CD_3COOD$), δ, ppm ($J_{C,F}$, Hz): 113.29 d.d ($C^6$, $J$ = 20.9, 4.1), 115.44 d.d ($C^{5a}$, $J$ = 7.6, 3.4), 119.96 q ($CF_3$, $J$ = 269.9), 124.09 d.d.d ($C^{9a}$, $J$ = 7.8, 3.0, 2.4), 142.26 d.d.d ($C^9$, $J$ = 264.0, 14.1, 2.9), 146.39 d.d.d ($C^8$, $J$ = 260.7, 16.7, 13.1), 150.61 s ($C^{3a}$), 150.85 d.d.d ($C^7$, $J$ = 252.9, 10.8, 1.5), 154.51 q.d ($C^2$, $J$ = 40.5, 3.5), 159.40 m ($C^5$). Found, %: C 44.92; H 0.79; N 23.38. $C_{10}H_2F_6N_4O$. Calculated, %: C 38.96; H 0.65; N 18.18.

**7,9-Difluoro-8-(1-pyrrolidinyl)-2-trifluoromethyl-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-one (XXa).** Pyrrolidine, 0.45 g (0.4 mmol), was added to a solution of 0.5 g (1.6 mmol) of compound **XIXb** in 5 ml of dimethylformamide. The mixture was heated for 5 h under reflux, cooled, and diluted with 15 ml of water. The precipitate was filtered off and recrystallized from DMSO. Yield 0.46 g (81%), mp 276–278°C. Found, %: C 17.57; H 2.90; N 15.77. $C_{14}H_{10}F_5N_4O$. Calculated, %: C 17.46; H 2.82; N 15.82.

**7,9-Difluoro-8-morpholino-2-trifluoromethyl-4*H*-[1,2,4]triazolo[1,5-*a*]quinazolin-5-one (XXb)**

was synthesized in a similar way. Yield 79%, mp 292–294°C. Found, %: C 44.74; H 2.75; N 18.68. $C_{14}H_{10}F_5N_5O_2$. Calculated, %: C 44.80; H 2.67; N 18.75.

This study was performed under financial support by the Russian Foundation for Basic Research (project nos. 03-03-32254 and 04-03-96107-Ural), by the Ministry of Education of the Russian Federation, CRDF, Annex BF4M05, EK-005-X2[REC-005], and "BRHE 2004 Post-Doctoral Fellowship Award" (grant no. Y2-C-05-01).

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