Cascade cyclization reactions of 3,4,5-triamino-1,2,4-triazole with aromatic aldehydes and cycloalkanones

V. V. Lipson,[a] S. M. Desenko,[b] V. V. Borodina,[a] M. G. Shirobokova,[a] O. V. Shishkin,[b] and R. I. Zubatyuk[b]*

[a]V. Ya. Danilevsky Institute of Endocrine Pathology Problems, Academy of Medical Sciences of the Ukraine, 10 ul. Artema, 61002 Kharkov, Ukraine. E-mail: lipson@ukr.net

[b]State Scientific Institution Scientific and Technological Corporation "Institute for Single Crystals", National Academy of Sciences of the Ukraine, 60 prosp. Lenina, 61001 Kharkov, Ukraine. E-mail: desenko@isc.kharkov.com

A three-component condensation of 3,4,5-triamino-1,2,4-triazole (**1**) with aromatic aldehydes **2a**—**f** and dimedone (**3**) or cyclohexanone (**8**) afforded partially hydrogenated 9-aryl-[1,2,4]triazolo[5,1-*b*]quinazolin-8-ones. The structure of 2-amino-6,6-dimethyl-3-(4-nitrobenzylidene)amino-9-(4-nitrophenyl)-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazolin-8-one (**4e**) was confirmed by X-ray diffraction data.

Key words: 3,4,5-triamino-1,2,4-triazole, aromatic aldehydes, dimedone, cyclohexanone, partially hydrogenated [1,2,4]triazolo[5,1-*b*]quinazolines, synthesis, X-ray diffraction analysis.

The presence of three nonequivalent nucleophilic centers in 3,4,5-triamino-1,2,4-triazole allows various pathways in its reactions with carbonyl 1,3-bielectrophiles and opens up wide possibilities of using this compound in the synthesis of fused heterocyclic systems. Originally, vicinal di- and triaminoazoles containing the *N*-amino group in reactions with enones were regarded only as 1,4-binucleophiles yielding azolotriazepine derivatives.[1,2] Later, it has been found that reactions of 1,2-di- and 1,2,3-triaminoazoles with α,β-unsaturated ketones, as well as with their mono- and dibromo derivatives, give fused pyridazine[3,4] and pyrimidine systems[5—7] rather than triazepine ones because the amino groups of those azoles are less nucleophilic than their endocyclic reactive sites. It should be noted that cyclization of 3,4,5-triamino-1,2,4-triazole with chalcones proceeds with elimination of the *N*-amino group, giving 2-amino-5,7-diaryl-1,2,4-triazolo[1,5-*a*]pyrimidines.[5] The goal of the present work was to elucidate the pathways of reactions of 3,4,5-triamino-1,2,4-triazole (**1**) with aromatic aldehydes and cycloalkanones.

We found that a three-component condensation of equimolar amounts of amine **1**, *para*-substituted benzaldehydes **2a**—**f**, and dimedone **3** in DMF gives brightly colored crystalline solids **4a**—**f** or their mixtures with compounds **5** in low yields (Scheme 1). With a double excess of the aldehyde with respect to the amine and dimedone, the yields of products **4a**—**f** increased correspondingly. In this case, no diamino derivatives **5** were detected. An analogous result was obtained in a reaction of azomethine **6** with diketone **3** in DMF; alternative products **7** were not detected. Thus, compound **6** acts in this reaction as a "depot" for benzaldehyde.

A reaction of triamine **1** with 4-nitrobenzaldehyde **2e** and cyclohexanone **8** yielded 2-amino-9-(4-nitrophenyl)[1,2,4]triazolo[5,1-*b*]quinazoline (**9**). This product was also obtained from azomethine **6** and cyclohexanone **8** and by an independent synthesis from 3,5-diaminotriazole **10**, aldehyde **2e**, and ketone **8**.

The structures of novel compounds **4a**—**f**, **5b**, **6**, and **9** were proven by spectroscopic methods. The structure of quinazolinone **4e** was confirmed by X-ray diffraction data.

In the IR spectra of compounds **4a**—**f** and **5b**, the absorption bands of the NH_2 and C=O groups at 3472—3268 and 1668—1656 cm^{-1}, respectively, and the bands of the CH_2 and CH_3 groups in the cyclohexenone fragment at 2966—2956 cm^{-1} are most characteristic.

The mass spectra of compounds **4a,e** contain low-intensity (~10%) molecular ion peaks $[M]^+$ with *m*/*z* 412 and 502, respectively. Intense peaks appear at *m*/*z* 335 (85%) and 308 (100%) for compound **4a** and at *m*/*z* 380 (45%) and 353 (78%) for compound **4e**. These radical ions are generated by successive elimination of the phenyl (nitrophenyl for **4e**) ring and the HCN group from the "hydrazone" fragments of the molecules.

The ^1H NMR spectra of compounds **4a**—**f** show signals suggesting the presence of two aryl rings, an amino group (singlet at δ 6.74—6.30), two methyl and two

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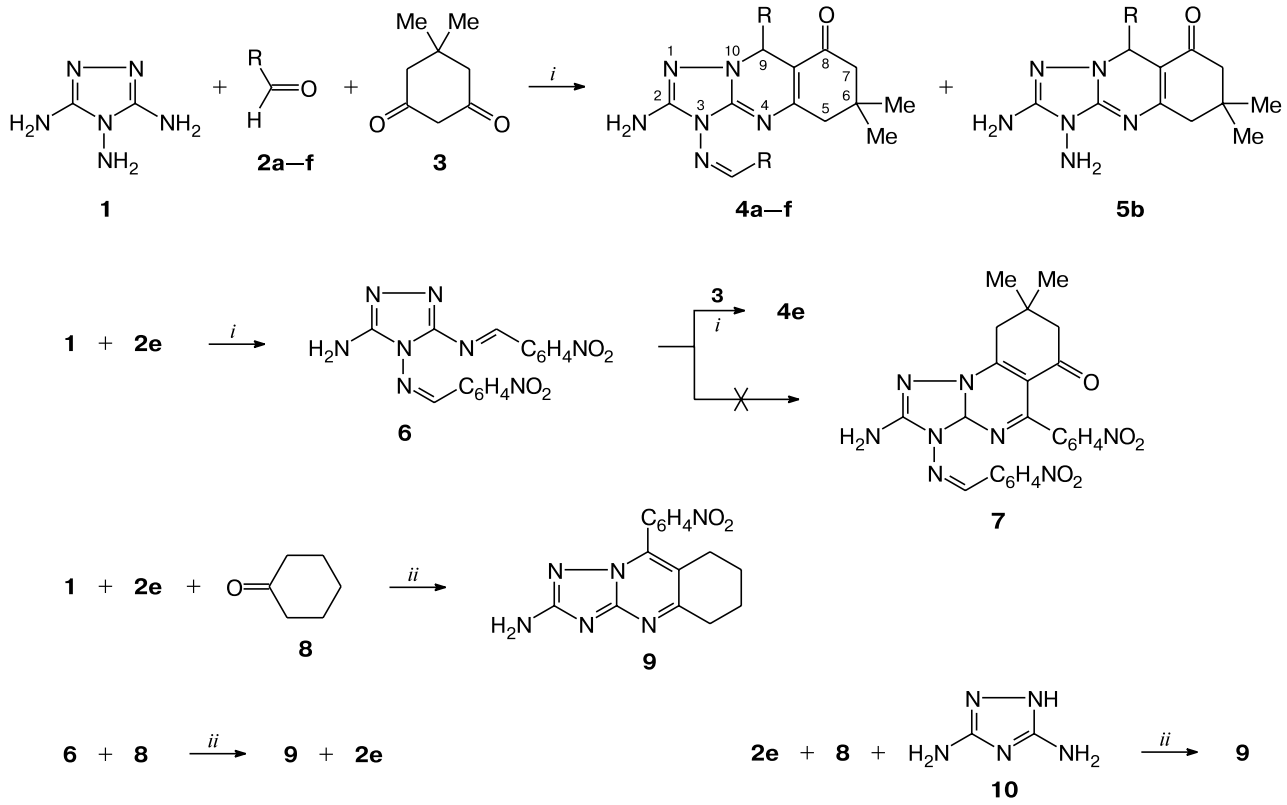
Scheme 1

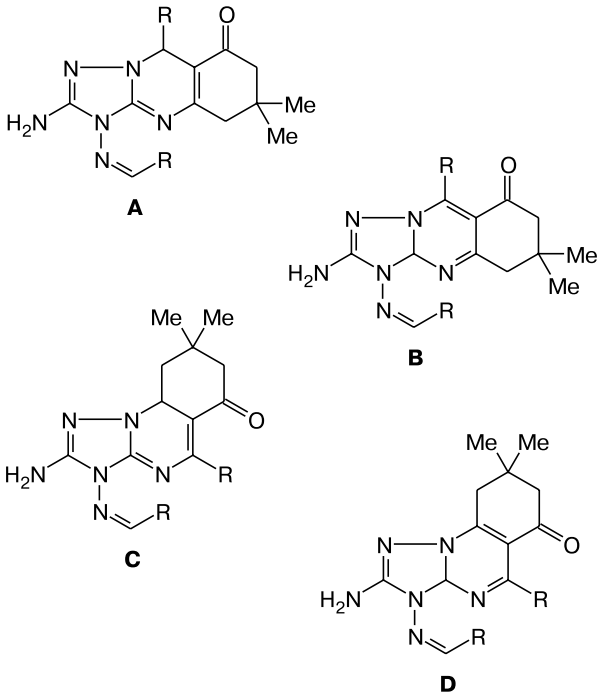


2, 4, 5: R = Ph (**a**), 4-$MeOC_6H_4$ (**b**), 4-$Me_2NC_6H_4$ (**c**), 4-ClC_6H_4 (**d**), 4-$NO_2C_6H_4$ (**e**), 2,4-$Cl_2C_6H_3$ (**f**)

i. DMF, DMF/H^+ or $MeOH/H^+$; *ii*. cyclohexanone.

methylene groups in the cyclohexenone fragment (the latter form AB systems), and two methine protons (singlets at δ 6.37—5.82 and 10.49—9.70). Such a signal arrangement is equally probable for both structures **A** and **B** and structures **C** and **D**.

The structure **A** of compound **4e** was unambiguously proved by X-ray diffraction analysis (Fig. 1). We found that quinazolinone **4e** exists in the crystal as a 1 : 1 solvate with a DMF molecule* (Fig. 2). The triazole ring, the amino group, and the substituent at the N(4) atom are coplanar to within 0.025 Å. The dihydropyrimidine and cyclohexane rings are in the half-chair conformation; the C(7) and C(3) atoms deviate from the planes of the other ring atoms by 0.263 and 0.662 Å, respectively. The nitrophenyl substituent at the C(7) atom has a pseudo-equatorial orientation (C(1)—C(6)—C(7)—C(10) torsion angle is 103.1(3)°) and is nearly perpendicular to the dihydropyrimidine ring (the angle between the adjacent planes of the rings is 85.9°). The C(23) and



* The solvate form is characteristic only of compound **4e**; its crystals for X-ray diffraction analysis were grown from DMF—Pr^iOH.

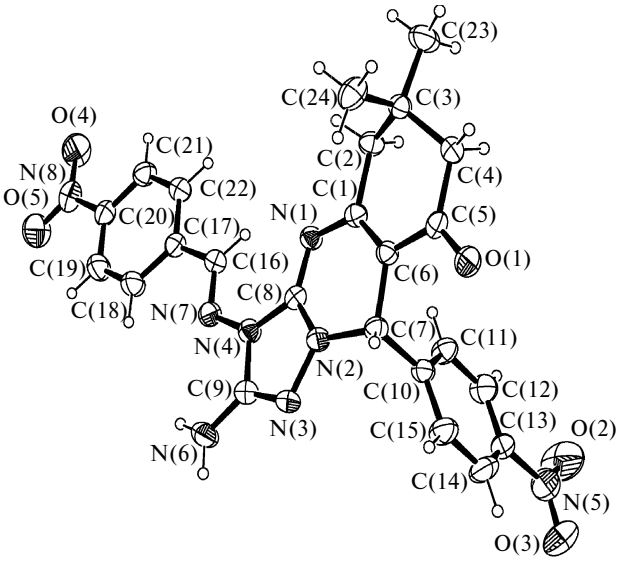


Fig. 1. Structure **4e**.

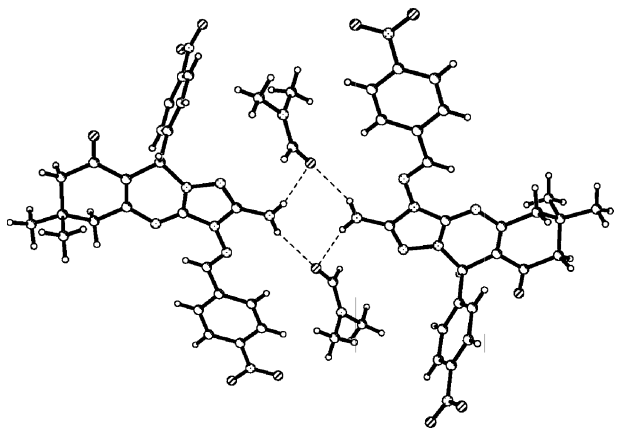


Fig. 2. Arrangement of the molecules of compound **4e** in the crystal of its solvate with DMF.

C(24) atoms have equatorial and axial orientations, respectively (C(1)—C(2)—C(3)—C(23) 169.3(3)° and C(1)—C(2)—C(3)—C(24) – 70.6(3)°). The N(2), N(4), and N(6) atoms are in planar trigonal configuration. The nitro group makes small angles with the plane of the benzene ring (O(2)—N(5)—C(13)—C(12) 2.9(6)° and O(3)—N(5)—C(13)—C(14) 6.0(5)°).

In structure **4e**, the C(5)—O(1) (1.239(3) Å) and C(1)—C(6) bonds (1.365 Å) are appreciably longer than average bond lengths (1.211 [8] and 1.326 Å, respectively), while the C(5)—C(6) bond (1.442(4) Å) is shortened (1.464 Å). This suggests strong conjugation in the enone fragment. The structure contains a very weak intramolecular C(16)—H(16)...N(1) hydrogen bond (H...N 2.24 Å, C—H...N 130°) and an attractive shortened H(6B)...N(7) contact (2.46 Å; the sum of the van der Waals radii is 2.66 Å).[9]

In the crystal, molecules of **4e** and DMF form centrosymmetric tetramers (see Fig. 2) *via* intermolecular hydrogen bonds: N(6)—H(6N)...O(1S)′ (2 – *x*, 1 – *y*, 2 – *z*) (H...O 2.06 Å, N—H...O 162°) and N(6)—H(6NB)...O(1S)′ (–1 + *x*, *y*, –1 + *z*) (H...O 1.92 Å, N—H...O 152°).

In contrast to the aforementioned compounds, structure **5b** was unambiguously proved by ^{1}H NMR spectroscopy. Its spectrum shows signals for the protons of only one aryl ring, a methine C(9)H proton (δ 5.84), two amino groups (δ 6.02 and 5.47, both s), the AB system of the CH_2 fragments, and protons of two CH_3 groups (both s).

Structure **9** was proved by spectroscopic methods and confirmed by an independent synthesis. The mass spectrum of quinazoline **9** contains the most intense molecular ion peak with *m*/*z* 310 (100%). The ^{1}H NMR spectrum shows two low-field doublets for the nitrophenyl protons and a singlet for the amino group at δ 6.11. The methylene protons reveal themselves as a multiplet at δ 1.49—1.87.

Thus, the structures of compounds **4a—f**, **5b**, and **9** suggests that the selective formation of the pyrimidine ring in reactions of 3,4,5-triamino-1,2,4-triazole with aromatic aldehydes and cycloalkanones yields triazolo[5,1-*b*]quinazoline systems. It should also be noted that as in reactions of triamine **1** with chalcones,[5,10] the formation of 2-amino-9-(4-nitrophenyl)-[1,2,4]triazolo[5,1-*b*]quinazoline (**9**) was accompanied by elimination of the *N*-amino group as low-molecular-weight products (NH_3 or NH_2OH). The retention of that group in structure **5b** is probably favored by the presence of the conjugated C=O group in the carbocycle.

Experimental

^{1}H NMR spectra were recorded on a Bruker AC-300 instrument (299.95 MHz) in DMSO-d_6 with Me_4Si as the internal standard. IR spectra were recorded on a Specord M-82 instrument (KBr pellets). The mass spectra of compounds **4a,e** and **9** were recorded on a Finnigan MAT INCOS-50 instrument (EI, 70 eV). The compositions of the reaction mixtures and the purity of the compounds obtained were checked by TLC on Silufol UV-254 plates with $CHCl_3$—MeOH (9 : 3) as an eluent. Melting points were determined on a Kofler hot stage.

2-Amino-3-benzylideneamino-6,6-dimethyl-9-phenyl-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazolin-8-one (4a). A mixture of amine **1** (0.11 g, 1 mmol), benzaldehyde **2a** (0.21 g, 2 mmol), and dimedone **3** (0.14 g, 1 mmol) in DMF (2 mL) was refluxed for 30—35 min and then propan-2-ol (10 mL) was added. The yield of quinazolinone **4a** was 0.21 g (52%), yellow crystals, m.p. 242—244 °C (from Pr^iOH). Compounds **4b—f** were obtained analogously.

Compound 4a. Found (%): C, 69.85; H, 5.79; N, 20.31. $C_{24}H_{24}N_6O$. Calculated (%): C, 69.90; H, 5.83; N, 20.39. IR, ν/cm^{-1}: 3384, 3288, 2956, 1660. ^{1}H NMR, δ: 10.00 (s, 1 H,

CH); 7.93—7.53, 7.24 (both m, 5 H each, H_{arom}); 6.51 (br.s, 2 H, NH_2); 5.95 (s, 1 H, C(9)H); 2.28 (dd, 2 H, $CH_{2(AB)}$, J = −16.0 Hz); 1.03, 0.96 (both s, 3 H each, Me). MS, m/z (I_{rel} (%)): 412 $[M]^+$ (10), 335 (85), 308 (100), 251 (35), 232 (90), 209 (30), 176 (25), 127 (24), 103 (85), 77 (45).

2-Amino-3-(4-methoxybenzylideneamino)-9-(4-methoxyphenyl)-6,6-dimethyl-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazolin-8-one (4b). The yield was 56%, yellow crystals, m.p. 226—228 °C (from Pr^iOH). Found (%): C, 67.00; H, 5.87; N, 17.72. $C_{26}H_{28}N_6O_3$. Calculated (%): C, 66.10; H, 5.93; N, 17.80. IR, ν/cm^{-1}: 3382, 3294, 2956, 1662. ^{1}H NMR, δ: 9.98 (s, 1 H, CH); 7.90—7.11, 6.81—6.76 (both dd, 4 H each, H_{arom}, J = 8.6 Hz); 6.44 (br.s, 2 H, NH_2); 5.87 (s, 1 H, C(9)H); 2.03 (dd, 2 H, $CH_{2(AB)}$, J = −16.0 Hz); 3.84, 3.69 (both s, 3 H each, OMe); 1.02, 1.03 (both s, 3 H each, Me).

2-Amino-3-(4-dimethylaminobenzylideneamino)-9-(4-dimethylaminophenyl)-6,6-dimethyl-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazolin-8-one (4c). The yield was 50%, orange crystals, m.p. 272—276 °C (from Pr^iOH). Found (%): C, 67.52; H, 6.87; N, 22.62. $C_{28}H_{34}N_8O$. Calculated (%): C, 67.47; H, 6.83; N, 22.49. IR, ν/cm^{-1}: 3468, 3392, 2956, 2868, 1668. ^{1}H NMR, δ: 9.70 (s, 1 H, CH); 7.72—6.59 (dd, 4 H, H_{arom}, J = 8.8 Hz); 7.76—7.03 (dd, 4 H, H_{arom}, J = 8.4 Hz); 6.30 (br.s, 2 H, NH_2); 5.82 (s, 1 H, C(9)H); 2.05 (dd, 2 H, $CH_{2(AB)}$, J = −16.4 Hz); 3.01, 2.83 (both s, 6 H each, $N(s)_2$); 1.02, 0.98 (both s, 3 H each, Me).

2-Amino-3-(4-chlorobenzylideneamino)-9-(4-chlorophenyl)-6,6-dimethyl-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazolin-8-one (4d). The yield was 53%, yellow crystals, m.p. 286—287 °C (from Pr^iOH). Found (%): C, 59.76; H, 4.49; Cl, 14.70; N, 17.38. $C_{24}H_{22}N_6Cl_2O$. Calculated (%): C, 59.88; H, 4.57; Cl, 14.76; N, 17.46. IR, ν/cm^{-1}: 3468, 3284, 2956, 2868, 1668. ^{1}H NMR, δ: 10.00 (s, 1 H, CH); 7.98—7.56, 7.32—7.25 (both dd, 4 H each, H_{arom}, J = 8.6 Hz); 6.60 (br.s, 2 H, NH_2); 5.95 (s, 1 H, C(9)H); 2.08 (dd, 2 H, $CH_{2(AB)}$, J = −16.4 Hz); 1.02, 0.95 (both s, 3 H each, Me).

2-Amino-6,6-dimethyl-3-(4-nitrobenzylideneamino)-9-(4-nitrophenyl)-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazolin-8-one (4e). ***A.*** The yield was 50%, dark red crystals, m.p. 276—280 °C (from Pr^iOH). Found (%): C, 57.32; H, 4.29; N, 22.24. $C_{24}H_{22}N_8O_5$. Calculated (%): C, 57.37; H, 4.38; N, 22.31. IR, ν/cm^{-1}: 3408, 3268, 2956, 2956, 1656, 1348. ^{1}H NMR, δ: 10.16 (s, 1 H, CH); 8.32—8.25 (dd, 4 H, H_{arom}, J = 8.6 Hz); 8.14—7.54 (dd, 4 H, H_{arom}, J = 8.8 Hz); 6.74 (br.s, 2 H, NH_2); 6.11 (s, 1 H, C(9)H); 2.10 (dd, 2 H, $CH_{2(AB)}$, J = −16.2 Hz); 1.03, 0.95 (both s, 3 H each, Me). MS, m/z (I_{rel} (%)): 502 $[M]^+$ (10), 380 (45), 353 (78), 322 (25), 266 (18), 232 (53), 175 (23), 148 (80), 102 (100), 76 (38).

B. A mixture of 5-amino-3,4-bis(4-nitrobenzylideneamino)-1,2,4-triazole (**6**) (0.38 g, 1 mmol) and dimedone **3** (0.14 g, 1 mmol) in DMF (2 mL) was refluxed for 30 min and then propan-2-ol (10 mL) was added. The yield of quinazolinone **4e** was 0.27 g (54%).

2-Amino-3-(2,4-dichlorobenzylideneamino)-9-(2,4-dichlorophenyl)-6,6-dimethyl-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazolin-8-one (4f). The yield was 51%, yellow crystals, m.p. 295—298 °C (from Pr^iOH). Found (%): C, 52.32; H, 4.00; Cl, 25.70; N, 15.16. $C_{24}H_{20}N_6Cl_4O$. Calculated (%): C, 52.36; H, 3.64; Cl, 25.82; N, 15.27. IR, ν/cm^{-1}: 3472, 3284, 2966, 1668. ^{1}H NMR, δ: 10.49 (s, 1 H, CH); 8.39—7.36 (m, 6 H, H_{arom}); 6.69 (br.s, 2 H, NH_2); 6.37 (s, 1 H, C(9)H); 2.06 (dd, 2 H, $CH_{2(AB)}$, J = −16.2 Hz); 1.03, 0.97 (both s, 3 H each, Me).

2,3-Diamino-9-(4-methoxyphenyl)-6,6-dimethyl-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazolin-8-one (5b). A mixture of amine **1** (0.11 g, 1 mmol), 4-methoxybenzaldehyde **2b** (0.14 g, 1 mmol), and dimedone **3** (0.14 g, 1 mmol) in DMF (2 mL) was refluxed for 35 min and then Pr^iOH (10 mL) was added. The yield of a mixture of quinazolinones **4b** and **5b** was 0.32 g. The **4b**/**5b** ratio was 1 : 7 (^{1}H NMR data). Threefold recrystallization from Pr^iOH gave compound **5b** as light yellow crystals, m.p. 205—208 °C. Found (%): C, 60.98; H, 6.27; N, 23.79. $C_{18}H_{22}N_6O_2$. Calculated (%): C, 61.02; H, 6.21; N, 23.73. IR, ν/cm^{-1}: 3456, 3392, 2960, 1664. ^{1}H NMR, δ: 7.08—6.78 (dd, 4 H, H_{arom}, J = 8.4 Hz); 6.02 (br.s, 2 H, NH_2); 5.84 (s, 1 H, C(9)H); 5.47 (br.s, 2 H, NH_2); 3.68 (s, 3 H, OMe); 2.04 (dd, 2 H, $CH_{2(AB)}$, J = −16.0 Hz); 1.00, 0.95 (both s, 3 H each, Me).

5-Amino-3,4-bis(4-nitrobenzylideneamino)-1,2,4-triazole (6). A solution of amine **1** (0.22 g, 2 mmol) and 4-nitrobenzaldehyde **2e** (0.64 g, 4 mmol) was refluxed in MeOH (5 mL) for 45 min (or in DMF for 20 min) in the presence of catalytic amounts of HCl. The reaction mixture was cooled and the resulting orange precipitate was filtered off. The yield of compound **6** was 0.5 g (66%), m.p. 271—275 °C (from MeOH). Found (%): C, 50.58; H, 3.05; N, 29.54. $C_{16}H_{12}N_8O_4$. Calculated (%): C, 50.53; H, 3.16; N, 29.47. IR, ν/cm^{-1}: 3444, 3388, 3300, 3080, 1648, 1348. ^{1}H NMR, δ: 9.41, 9.33 (both s, 1 H each, CH); 8.38—8.24 (m, 8 H, H_{arom}); 6.47 (br.s, 2 H, NH_2).

2-Amino-9-(4-nitrophenyl)-5,6,7,8-tetrahydro[1,2,4]triazolo[5,1-*b*]quinazoline (9). ***A.*** A mixture of amine **1** (0.11 g, 1 mmol), 4-nitrobenzaldehyde **2e** (0.15 g, 1 mmol), and cyclohexanone **8** (1 mL, 10 mmol) was refluxed for 2.5 h and then Pr^iOH (10 mL) was added. The yield of quinazoline **9** was 0.24 g (78%), yellow crystals, m.p. >300 °C. Found (%): C, 58.21; H, 4.63; N, 27.15. $C_{15}H_{14}N_6O_2$. Calculated (%): C, 58.06; H, 4.52; N, 27.10. IR, ν/cm^{-1}: 3328, 3244, 2932, 2860, 1348. ^{1}H NMR, δ: 8.32—7.56 (dd, 4 H, H_{arom}, J = 8.0 Hz); 6.38 (br.s, 2 H, NH_2); 1.87—1.49 (m, 8 H, CH_2). MS, m/z (I_{rel} (%)): 310 $[M]^+$ (100), 280 (45), 263 (23), 238 (24), 222 (20), 91 (24).

B. A mixture of azomethine **6** (0.38 g, 1 mmol) and cyclohexanone **8** (1 mL, 10 mmol) was refluxed for 2 h and Pr^iOH (15 mL) was added. The yield of compound **9** was 0.22 g (72%).

C. A mixture of amine **10** (0.1 g, 1 mmol), 4-nitrobenzaldehyde **2e** (0.15 g, 1 mmol), and cyclohexanone **8** (1 mL, 10 mmol) was refluxed for 1.5 h and Pr^iOH (10 mL) was added. The yield of quinazoline **9** was 0.24 g (78%).

X-ray diffraction analysis of compound 4e. Single crystals of compound **4e** were grown by slow crystallization from DMF—Pr^iOH (5 : 1). The unit cell parameters and the intensities of 5525 reflections (the number of independent reflections was 4979, R_{int} = 0.046) were measured on a Siemens P3/PC automatic four-circle diffractometer (Mo-$K\alpha$, graphite monochromator, 2θ/θ-scan mode, $2\theta_{max}$ = 50°). Crystals of compound **4e** are triclinic ($C_{24}H_{22}N_8O_5(Me)_2NCHO$): at 20 °C, a = 5.730(1) Å, b = 15.568(4) Å, c = 16.331(4) Å, α = 81.04(2)°, β = 89.69(2)°, γ = 82.33(2)°, V =1426.1(5) Å^3, M_r = 575.59, Z = 2, space group $P\bar{1}$, d_{calc} = 1.340 g cm^{-3}, μ(Mo-K_α) = 0.098 mm^{-1}, F(000) = 604. The structure was solved by the direct method with the SHELXTL program package.[11] Hydrogen atoms were located from the electron density difference maps and refined in the rider model with nonfixed $U_{iso} = nU_{equiv}$

(n = 1.5 for the methyl H atoms and n = 1.2 for the other H atoms). In the structure refinement, the following constraints were placed on the bond lengths in the DMF molecule: O(1S)—C(1S) 1.236(5) Å, N(1S)—C(1S) 1.321(5) Å, and N(1S)—C(2S) and N(1S)—C(3S) 1.435(5) Å. The structure was refined on F^2 by the full-matrix least-squares method in the anisotropic approximation for the non-hydrogen atoms (wR_2= 0.163 from 4952 reflections, R_1 = 0.064 from 2383 reflections with $F > 4\sigma(F)$, GOOF = 0.921). Comprehensive X-ray diffraction data have been deposited with the Cambridge Crystallographic Data Center.

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