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Reaction of 5-Substituted Tetrazoles with Trifluoroacetic Anhydride

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Abstract—5-Substituted tetrazoles readily react with trifluoroacetic anhydride at 20–25°C to give the corresponding 2-substituted 5-trifluoromethyl-1,3,4-oxadiazoles, in contrast to published data according to which the title compounds are converted into 1,3,4-oxadiazole derivatives on heating with carboxylic acid anhydrides or chlorides at 100–120°C. The reaction is governed not only by the rate of acylation of the tetrazole ring and temperature conditions but also by the stability of intermediate *N*-acyltetrazoles.

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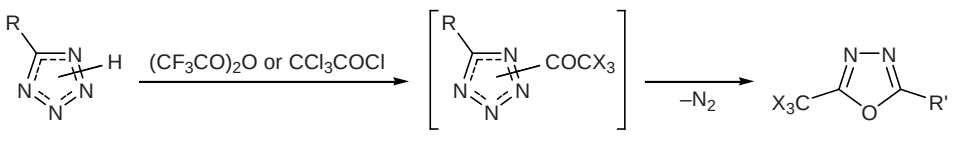
The well known transformation of 5-substituted tetrazoles into 1,3,4-oxadiazoles upon thermolysis in the presence of carboxylic acid chlorides and anhydrides [1, 2] has some limitations which are related as a rule to thermal stability of the initial tetrazoles, intermediate compounds, and products (the reaction occurs above 100°C). For example, reactions of 5-vinyl- and 5-[2-(dimethylamino)ethyl]tetrazoles **Ib** and **Ic** and some other tetrazoles with aliphatic and aromatic acid chlorides at elevated temperature are accompanied by strong tarring. Unlike published data on the thermolysis of 5-substituted tetrazoles in the presence of carboxylic acid chlorides and anhydrides [1, 2], we previously showed that such reactions with trifluoroacetic anhydride readily occur at low temperature [3]. The observed strong gas evolution suggests that the process begins even at –5°C, and the reaction completion is judged by the amount of released nitrogen. The reaction is successful in methylene chloride, diethyl ether, acetone, benzene, and toluene. It is advisable to use low-boiling solvents which facilitate isolation of the products. Under the above conditions, we succeeded in effecting reactions with trifluoroacetic anhydride of 5-vinyl-, 5-[2-(dimethylamino)ethyl]-, 5-chloromethyl-, and 5-(2-chloroethyl)tetrazoles **Ib**, **Ic**, **Ih**, and **Ii**, i.e., with those substrates which underwent tarring under the thermolysis conditions. In addition, tetrazoles **Ia** and **Id**–**Ig**, as well as bicyclic compounds **Ij**–**Il**, were converted into the corresponding 1,3,4-oxadiazoles (Scheme 1).

The transformation of all the above tetrazoles into oxadiazoles is characterized by a fairly high rate at room temperature, so that the reaction time does not exceed 5 h. An exception was the reaction of trifluoroacetic anhydride with cyanomethyltetrazole **Ie**, where nitrogen evolution lasted more than 24 h. Nevertheless, the corresponding oxadiazole was isolated in a good yield. The ^{13}C NMR spectra of compounds **IIa**–**IIk** contained signals in the regions δ_C 160–173 and 155–

Scheme 1.



Ia–Il **IIa–III**, **III**

I, **II**, X = F, R = R′ = Ph (**a**), CH_2=CH (**b**), $Me_2NCH_2CH_2$ (**c**), $EtOCOCH_2$ (**d**), $NCCH_2$ (**e**), Me (**f**), $MeOCH_2CH_2$ (**g**), $ClCH_2$ (**h**), $ClCH_2CH_2$ (**i**), 2-(5-phenyl-2*H*-tetrazol-2-yl)ethyl (**j**), 2-phenyl-1,2,3-triazol-4-yl (**k**); R = 2-(tetrazol-5-yl)ethyl, R′ = 2-(5-trifluoromethyl-1,3,4-oxadiazol-2-yl)ethyl (**l**); **III**, X = Cl, R′ = Ph.

159 ppm, which were assigned to the C^2 and C^5 atoms of the 1,3,4-oxadiazole ring, respectively. In addition, a signal at δ_C 116–120 ppm was observed due to trifluoromethyl group. This signal, as well as the C^5 signal, appeared as a quartet with a coupling constant $^1J_{CF}$ of 267–271 Hz. The ^{13}C–^{19}F coupling constant for C^5 was 43–46 Hz. Compounds **II** showed in the mass spectra ion peaks corresponding to particular molecular fragments.

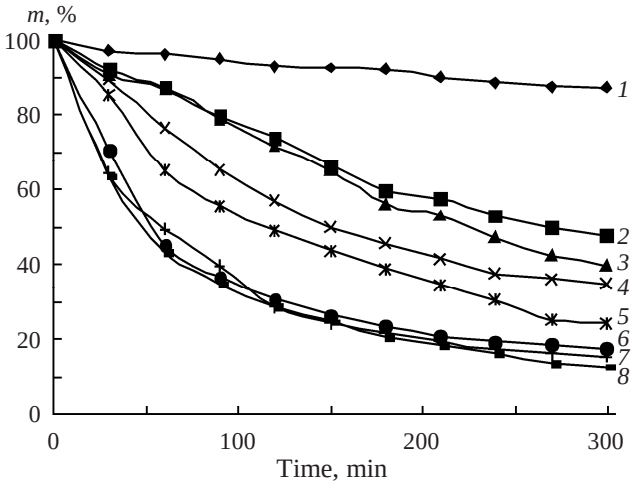
We also examined the reaction of 5-phenyltetrazole (**Ia**) with trichloroacetyl chloride. However, no reaction occurred at room temperature. Taking into account previously published data on the catalytic effect of amines in the acylation of tetrazoles [4], triethylamine or pyridine was added to the reaction mixture. In the presence of an equimolar amount of triethylamine or pyridine, tetrazole **Ia** reacted with trichloroacetyl chloride at room temperature with vigorous evolution of nitrogen. The product was 2-phenyl-5-trichloromethyl-1,3,4-oxadiazole (**III**) which was identical to a sample prepared by reaction of **Ia** with trichloroacetyl chloride at 80°C. As might be expected, the presence of a strong electron-withdrawing group in the molecule of acylating agent favors fast acylation of the substrate. Intermediate *N*-acyltetrazoles having an electron-withdrawing fragment are very unstable and are readily converted into 1,3,4-oxadiazoles at a relatively low temperature. Thus the use of highly reactive anhydrides makes it possible to obtain thermally unstable oxadiazoles from labile 5-substituted tetrazoles.

Most tetrazoles studied in this work were prepared by cycloaddition of ammonium azide to the corresponding nitriles. However, some nitriles decomposed during the process because of severe conditions (temperature 100–120°C; reaction time 20 h and longer). It is known that ammonium azides often react with nitriles much more efficiently than does sodium azide [5]. The most reactive are ammonium azide, dimethylammonium azide, and symmetric tetramethylguanidinium azide [6]. In some cases, the use of these azides shortened the reaction time only slightly and did not prevent the reaction mixture from tarring. With a view to determine which azides are the most reactive and to accelerate the process, we compared the reactivities of a series of ammonium azides formed *in situ* by exchange reaction of amine hydrochlorides with sodium azide. As amines we used mainly difunctional derivatives: benzene-1,2-diamine, ethane-1,2-diamine, hexane-1,6-diamine, 2-hydroxyethanamine, diethylamine, and piperidine. The reactions of the corresponding azides with benzonitrile were carried out under similar conditions, in DMF at 100°C. The progress of reactions was monitored by gas–liquid chromatography, following the consumption of benzonitrile (see figure). It is seen that 2-hydroxyethylammonium azide, piperidinium azide, and diethylammonium azide exhibit enhanced reactivity.

The curves shown in figure provide relative estimates of the degree of transformation of benzonitrile in the presence of ammonium salt, which are not always consistent with the yields of tetrazoles in reactions with preliminarily prepared azides. Among the above ammonium azides, we isolated as individual substances 2-hydroxyethylammonium azide and diethylammonium azide by mixing benzene solutions of the amine and hydrazoic acid. 2-Hydroxyethylammonium azide was used as the most reactive to synthesize a series of 5-substituted tetrazoles.

EXPERIMENTAL

The NMR spectra were recorded on a Varian VXR-500S spectrometer at 500 MHz for ^1H and 126 MHz for ^{13}C from solutions in acetone-d_6 and acetone, respectively. The elemental compositions were determined on a FLASH EA 1112 Series CHN analyzer. The mass spectra were obtained on an Agilent 5973N–GC-6890 GC–MS system (injector temperature 250°C, HP-Ultra colum). GLC analysis was performed on a Gals chromatograph equipped with a flame ionization detector and an HP-5 15-m capillary column (carrier gas nitrogen, flow rate 80 ml/min; detector temperature 240°C, injector temperature 240°C, oven tem-



Plots of the concentration of benzonitrile (%) versus time in the reactions with different azides Cat^+N_3; Cat = (*1*) Na, (*2*) 2-$NH_2C_6H_4NH_3$, (*3*) $H_2N(CH_2)_6NH_3$, (*4*) NH_4, (*5*) $H_2N(CH_2)_2NH_3$, (*6*) $(CH_2)_5NH_2$, (*7*) Et_2NH_2, (*8*) $HO(CH_2)_2NH_3$.

perature 130°C). Diethylammonium azide and 2-hydroxyethylammonium azide were prepared according to the procedure reported in [7], tetrazoles **Ij** and **Ik** were synthesized as described in [8], and tetrazoles **Ib** and **Ic** were obtained according to [9].

Tetrazoles Ia, Id, Ie, and Ig (*general procedure*). A suspension of 0.06 mol of sodium azide and 0.06 mol of 2-aminoethanol hydrochloride in 35 ml of DMF was stirred for 30 min at 40°C, 0.06 mol of the corresponding nitrile (or 0.03 mol of dinitrile) was added, and the mixture was heated to 100°C and kept for 5–7 h at that temperature. The progress of the reaction was monitored by TLC. When the reaction was complete, the solvent was distilled off under reduced pressure, and the residue was treated with a solution of sodium hydroxide to pH 9 and washed with ethyl acetate. The aqueous solution was acidified with hydrochloric acid to pH 2, and the precipitate was filtered off. Otherwise, the solution was extracted with ethyl acetate (3×10 ml), the extract was dried over magnesium sulfate, the solvent was distilled off under reduced pressure, and the residue was recrystallized from ethanol.

5-Phenyltetrazole (Ia). Yield 67%, mp 216°C; published data [5]: mp 217–218°C.

Ethyl (tetrazol-5-yl)acetate (Id). Yield 53%, mp 126°C; published data [4]: mp 128–130°C.

(Tetrazol-5-yl)acetonitrile (Ie). Yield 55%, mp 115°C; published data [10]: mp 115–117°C.

5-(2-Methoxyethyl)tetrazole (Ig). Yield 45%, mp 65–67°C; published data [4]: mp 66–67°C.

***N*,*N*-Dimethyl-2-(5-trifluoromethyl-1,3,4-oxadiazol-2-yl)ethanamine (IIc).** A solution of 1.9 g (9 mmol) of trifluoroacetic anhydride in 2 ml of methylene chloride was added dropwise to a suspension of 1.0 g (7 mmol) of tetrazole **Ic** in 10 ml of methylene chloride under stirring at room temperature. The colorless mixture gradually became homogeneous. When the evolution of nitrogen was over, the mixture was neutralized with a saturated solution of sodium hydrogen carbonate to pH 9, the aqueous phase was treated with diethyl ether (3×10 ml), the extracts were combined with the organic phase and dried over magnesium sulfate, the solvent was distilled off, and the residue was distilled under reduced pressure. Yield 1.2 g (81%), bp 66°C (10 mm), n_D^{20} = 1.3990. ^{1}H NMR spectrum, δ, ppm: 1.5 s (6H, CH_3), 2.6 t (2H, 2-CH_2), 3.0 t (2H, CH_2N). ^{13}C NMR spectrum, δ_C, ppm: 27.4 s (2-CH_2), 47.9 s (CH_3), 58.9 s (CH_2N), 120.3 q (CF_3, J = 267.2 Hz), 158.5 q (C^5, J = 43.8 Hz), 172.2 s (C^2). Mass spectrum, m/z (I_{rel}, %): 209 (0.7) $[M]^+$, 165 (2.1), 95 (2.1), 69 (16.3), 58 (100), 42 (29.8). Found, %: C 40.16; H 4.34; N 20.02. $C_7H_{10}F_3N_3O$. Calculated, %: C 40.19; H 4.78; N 20.1. *M* 209.17.

Compounds **IIa**, **IIb**, and **IId–IIl** were synthesized in a similar way.

2-Phenyl-5-trifluoromethyl-1,3,4-oxadiazole (IIa) was obtained from 1 g (6.8 mmol) of 5-phenyltetrazole (**Ia**) and 1.7 g (8.0 mmol) of trifluoroacetic anhydride in 12 ml of methylene chloride. Yield 1.3 g (89%), mp 49–50°C (EtOH). ^{1}H NMR spectrum, δ, ppm: 7.6–8.1 m (5H, Ph). ^{13}C NMR spectrum, δ_C, ppm: 154.1 q (C^5, J = 43.6 Hz), 166.1 s (C^2), 116.4 q (CF_3, J = 269.5 Hz), 122.1–133.3 m (C_{arom}). Mass spectrum, m/z (I_{rel}, %): 214 (56.7) $[M]^+$, 145 (64.7), 77 (100), 68 (43.5), 118 (3.3), 195 (3.5). Found, %: C 50.39; H 2.41; N 12.92. $C_9H_5F_3N_2O$. Calculated, %: C 50.47; H 2.34; N 13.08. *M* 214.15.

2-Trifluoromethyl-5-vinyl-1,3,4-oxadiazole (IIb) was obtained from 5 g (50 mmol) of 5-vinyltetrazole (**Ib**) and 11.6 g (55 mmol) of trifluoroacetic anhydride in 115 ml of methylene chloride. Yield 6.5 g (76%), bp 40°C (15 mm), n_D^{20} = 1.3945, d_4^{20} = 1.32. ^{1}H NMR spectrum, δ, ppm: 6.92 (1H, =CH), 6.45 (1H, $=CH_2$), 6.10 (1H, $=CH_2$). ^{13}C NMR spectrum, δ_C, ppm: 118.74 ($=C^{\alpha}$), 128.84 ($=C^{\beta}$), 165.26 s (C^5), 154 q (C^2), 116.51 q (CF_3). Mass spectrum, m/z (I_{rel}, %): 164 (27.9) $[M]^+$, 145 (2.3), 95 (79), 69 (100). Found, %: C 36.54; H 1.45; N 17.05. $C_5H_3F_3N_2O$. Calculated, %: C 36.59; H 1.83; N 17.07. *M* 164.09.

Ethyl (5-trifluoromethyl-1,3,4-oxadiazol-2-yl)-acetate (IId) was synthesized from 1 g (6.4 mmol) of tetrazolylacetate **Id** and 1.4 g (8.3 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 0.87 g (61%), bp 91–92°C (7 mm), n_D^{24} = 1.3952. ^{1}H NMR spectrum, δ, ppm: 0.74 t (3H, C**H**$_3$CH$_2$), 3.7 q (2H, OCH_2), 3.9 s (2H, CH_2CO). ^{13}C NMR spectrum, δ_C, ppm: 14.3 s (**C**H_2CO), 32.6 s (CH_3), 63.2 s (OCH_2), 117.6 q (CF_3, J = 279.4 Hz), 156.9 q (C^5, J = 45.1 Hz), 164.9 s (C=O), 167.1 s (C^2). Mass spectrum, m/z (I_{rel}, %): 224 (0.5) $[M]^+$, 205 (2.3), 179 (28), 152 (85.6), 69 (100), 42 (46.5). Found, %: C 37.41; H 2.97; N 12.07. $C_7H_7F_3N_2O_3$. Calculated, %: C 37.5; H 3.13; N 12.5. *M* 224.14.

(5-Trifluoromethyl-1,3,4-oxadiazol-2-yl)acetonitrile (IIe) was synthesized from 0.8 g (7.3 mmol) of 5-cyanomethyltetrazole **Ie** and 1.7 g (10.9 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 0.72 g (55%), mp 35°C (from EtOH).

^{13}C NMR spectrum, δ_C, ppm: 15.6 s (CH_2), 112.7 s (CN), 116.4 q (CF_3, J = 270.4 Hz), 155.5 q (C^5, J = 45.2 Hz), 172.9 s (C^2). Mass spectrum, m/z (I_{rel}, %): 177 (18.6) $[M]^+$, 158 (4.5), 137 (1.4), 108 (100), 69 (69.1). Found, %: C 32.65; H 1.02; N 23.16. $C_5H_2F_3N_3O$. Calculated, %: C 33.9; H 1.13; N 23.73. M 177.09.

2-Methyl-5-trifluoromethyl-1,3,4-oxadiazole (IIf) was synthesized from 1 g (12 mmol) of 5-methyltetrazole (**If**) and 3.35 g (16 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 0.95 g (52%), bp 26–28°C (8 mm), n_D^{20} = 1.3585. ^{13}C NMR spectrum, δ_C, ppm: 10.8 s (CH_3), 117.7 q (CF_3, J = 268.8 Hz), 156.3 q (C^5, J = 43.8 Hz), 168.2 s (C^2). Mass spectrum, m/z (I_{rel}, %): 152 (27.9) $[M]^+$, 133 (4.2), 83 (100), 69 (59.1). Found, %: C 31.34; H 1.2; N 18.04. $C_4H_3F_3N_2O$. Calculated, %: C 31.58; H 1.97; N 18.42. M 152.08.

2-(2-Methoxyethyl)-5-trifluoromethyl-1,3,4-oxadiazole (IIg) was synthesized from 1 g (7.8 mmol) of 5-(2-methoxyethyl)tetrazole (**Ig**) and 2.15 g (10 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 1.03 g (67%), bp 79–80°C (12 mm), n_D^{20} = 1.3895. ^{13}C NMR spectrum, δ_C, ppm: 26.8 s (5-CH_2), 58.6 s (CH_3O), 68.7 s (CH_2O), 119.4 q (CF_3, J = 267.0 Hz), 156.2 q (C^5, J = 43.7 Hz), 168.8 s (C^2). Mass spectrum, m/z (I_{rel}, %): 181 (20), 166 (21), 137 (1.9), 69 (39.5), 45 (100). Found, %: C 35.57; H 3.12; N 13.97. $C_6H_7F_3N_2O_2$. Calculated, %: C 36.73; H 3.57; N 14.29. M 196.13.

2-Chloromethyl-5-trifluoromethyl-1,3,4-oxadiazole (IIh) was obtained from 1.25 g (10 mmol) of 5-chloromethyltetrazole (**Ih**) and 4.12 g (20 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 0.95 g (49%), bp 69–70°C (14 mm), n_D^{24} = 1.3991. ^{13}C NMR spectrum, δ_C, ppm: 43.7 s (CH_2Cl), 116.2 q (CF_3, J = 270.0 Hz), 155.6 q (C^5, J = 43.8 Hz), 165.2 s (C^2). Mass spectrum, m/z (I_{rel}, %): 186 (21.8) $[M]^+$, 151 (2.2), 137 (2.3), 117 (100), 69 (85.5), 49 (35.5). Found, %: C 25.23; H 1.12; N 14.73. $C_4H_2F_3ClN_2O$. Calculated, %: C 25.74; H 1.07; N 15.01. M 186.52.

2-(2-Chloroethyl)-5-trifluoromethyl-1,3,4-oxadiazole (IIi) was obtained from 2 g (15 mmol) of 5-(2-chloroethyl)tetrazole (**Ii**) and 4.7 g (22.5 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 0.9 g (30%), bp 83°C (10 mm), n_D^{24} = 1.4050. ^{1}H NMR spectrum, δ, ppm: 4.1 t (2H, CH_2Cl), 3.6 t (2H, 2-CH_2). ^{13}C NMR spectrum, δ_C, ppm: 29.1 s (2-CH_2), 40.5 s (CH_2Cl), 118.2 q (CF_3), 158.1 q (C^5), 167.9 (C^2). Mass spectrum, m/z (I_{rel}, %): 199 (0.9) $[M]^+$, 165 (100), 131 (20.5), 82 (0.9), 69 (43.7), 49 (14.9). Found, %: C 29.65; H 1.76; N 13.71. $C_5H_4F_3ClN_2O$. Calculated, %: C 29.93; H 2.00; N 13.97. M 209.55.

5-Phenyl-2-[2-(5-trifluoromethyl-1,3,4-oxadiazol-2-yl)ethyl]tetrazole (IIj) was obtained from 1 g (4.1 mmol) of bistetrazole **Ij** and 1.5 g (7.1 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 0.92 g (72%), mp 77–78°C (from EtOH). ^{13}C NMR spectrum, δ_C, ppm: 29.4 s (2′-CH_2), 47.5 s (CH_2N), 117.2 q (CF_3, J = 269.2 Hz), 126.7–130.8 m (C_{arom}), 156.8 q ($C^{5'}$, J = 43.8 Hz), 166.3 s ($C^{2'}$), 163.4 s (C^5). Found, %: C 46.32; H 2.1; N 27.1. $C_{12}H_9F_3N_6O$. Calculated, %: C 46.45; H 2.9; N 27.1.

2-(2-Phenyl-1,2,3-triazol-4-yl)-5-trifluoromethyl-1,3,4-oxadiazole (IIk) was synthesized from 1 g (4.7 mmol) of tetrazole **Ik** and 1.48 g (7 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 1 g (76%), mp 112–114°C (from EtOH). ^{13}C NMR spectrum, δ_C, ppm: 120–137 m (C_{arom}), 140.1 s ($C^{4'}$), 134.9 s ($C^{5'}$), 117.4 q (CF_3, J = 270.4 Hz), 155.6 q (C^5, J = 46.3 Hz), 160.8 s (C^2). Mass spectrum, m/z (I_{rel}, %): 281 (100) $[M]^+$, 262 (3.3), 212 (1.4), 91 (49.3), 69 (13.1). Found, %: C 46.57; H 2.03; N 24.87. $C_{11}H_6F_3N_5O$. Calculated, %: C 46.98; H 2.14; N 24.91. M 281.20.

1,2-Bis(5-trifluoromethyl-1,3,4-oxadiazol-2-yl)-ethane (IIl) was obtained from 1 g (4.1 mmol) of compound **Il** and 1 g (5 mmol) of trifluoroacetic anhydride in 15 ml of methylene chloride. Yield 1.1 g (60%), mp 100–101°C (from EtOH). ^{1}H NMR spectrum, δ, ppm: 3.0 (2H, CH_2). ^{13}C NMR spectrum, δ_C, ppm: 116.1 q (CF_3, J = 270.4 Hz), 21.3 s (CH_2), 154.1 q (C^5, J = 43.8 Hz), 167.7 s (C^2). Mass spectrum, m/z (I_{rel}, %): 302 (65.1) $[M]^+$, 283 (47), 164 (4.2), 137 (0.9), 68 (1.9), 233 (100). Found, %: C 31.52; H 1.27; N 18.42. $C_8H_4F_6N_4O_2$. Calculated, %: C 31.79; H 1.32; N 18.54. M 302.14.

2-Phenyl-5-trichloromethyl-1,3,4-oxadiazole (III). *a*. Trichloroacetyl chloride, 1.24 g (6.8 mmol), was added at room temperature to a suspension of 0.5 g (3.4 mmol) of tetrazole **Ia** in 10 ml of methylene chloride, and 0.34 g (3.4 mmol) of triethylamine or 0.28 g (3.4 mmol) of pyridine was then added. When vigorous evolution of nitrogen ceased, the mixture was neutralized to pH 9 with a saturated solution of sodium carbonate. The organic layer was separated and washed with water, the aqueous layer was extracted with methylene chloride (3×10 ml), the extracts were combined with the organic phase and dried over magnesium

sulfate, the solvent was evaporated on a Petri dish in air, and the residue was recrystallized from ethanol. Yield 0.68 g (76%), mp 64–65°C (from EtOH).

b. *Thermolysis*. A mixture of 0.5 g (3.4 mmol) of tetrazole **Ia** and 1.24 g (6.8 mmol) of trichloroacetyl chloride in 1 ml of toluene was heated to 80°C and was then kept at that temperature until nitrogen no longer evolved. The mixture was treated as described above in *a*. Yield 0.65 g (72%), mp 65–66°C. ^{13}C NMR spectrum, δ_C, ppm: 83.3 s (CCl_3), 122.9–133.2 m (C_{arom}), 163.3 s (C^5), 167.1 s (C^2). Mass spectrum, *m*/*z* (I_{rel}, %): 262 (10.5) $[M]^+$, 227 (45.5), 145 (17.7), 117 (1.8), 105 (100), 77 (72.7). Found, %: C 40.92; H 1.5; N 10.63. $C_9H_5Cl_3N_2O$. Calculated, %: C 40.99; H 1.9; N 10.63. *M* 263.51.

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