Stable σ-adducts of 4,6-dinitrotetrazolo[1,5-*a*]pyridine with alkoxide anions*

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The reaction of 2-chloro-3,5-dinitropyridine with two equivalents of KN_3 in the presence of ROH results in stable Meisenheimer-type σ-adducts of 4,6-dinitrotetrazolo[1,5-*a*]pyridine with RO^- anions (R = H, Alk, Ph). The mechanism of σ-complex formation was suggested. The structure of the σ-adduct with R = Me was established by IR and NMR spectroscopy and by X-ray diffraction analysis.

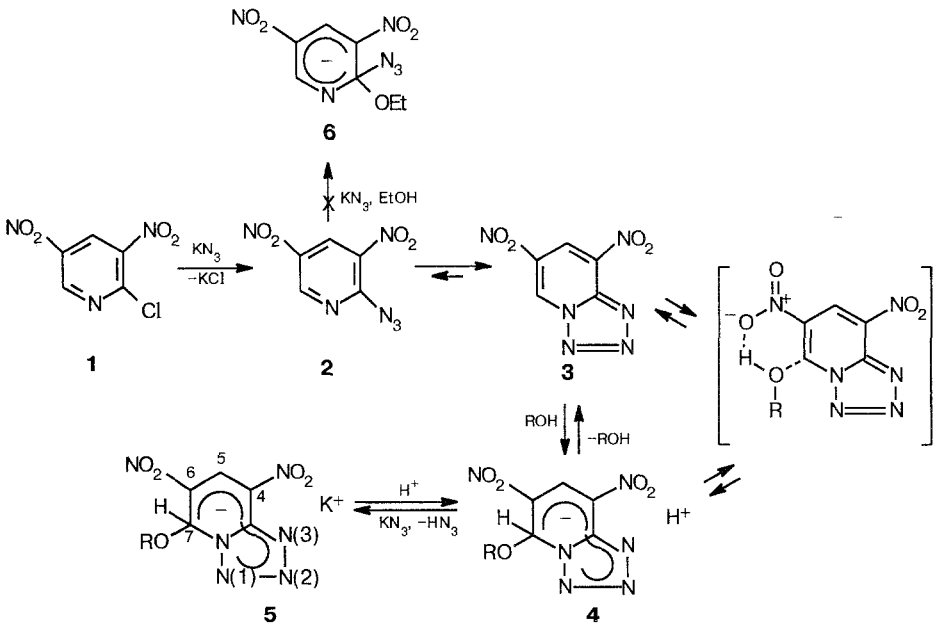
Key words: Meisenheimer complexes; σ-adducts; tetrazole-annelated systems.

Nucleophilic substitution in aromatic compounds involves the formation of intermediate σ-adducts.[2] These adducts are usually unstable. However, in certain cases (including the Meisenheimer complexes) they are relatively stable and can be obtained as individual compounds[3,4] and characterized using, *e.g.*, X-ray analysis, which is important for understanding the structure of the transition state. However, in most cases they are unstable and therefore cannot be obtained in the crystalline state.

It is known that 3,5-dinitropyridines readily form σ-complexes with alkoxides.[5] However, there are no similar data for fused tetrazole systems obtained from α-azidopyridines.[6]

* For the preliminary communication, see Ref. 1.

Scheme 1



R = H(**5a**); Me(**5b**); Et(**5c**); Pr^i(**5d**); Am^i(**5e**); *cyclo*-C_6H_{11}(**5f**); $C_{12}H_{25}$(**5g**); $ClCH_2CH_2$(**5h**); $CH{\equiv}C{-}CH_2$(**5i**); $PhCH_2$(**5j**); Ph(**5k**)

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In attempts to synthesize 4,6-dinitrotetrazolo-[1,5-*a*]pyridine (**3**) from 2-chloro-3,5-dinitropyridine (**1**) in an alcohol, we detected stable adducts (**5**). The reaction proceeds completely when two or more equivalents of KN_3 are present; one equivalent of HN_3 is liberated in the reaction. On the other hand, even when KN_3 was present in an insufficient amount, neither the intermediate α-azidopyridine (**2**) nor tetrazolopyridine (**3**) were detected. The reaction has general applicability and involves water, phenols and practically any alcohols except tertiary alcohols.

Normally, anionic σ-complexes are formed under conditions when alkoxide anions exist. However, in the present case the reaction is carried out in an acidic medium, which excludes the presence of alkoxide anions. Therefore, we investigated the mechanism of this unusual reaction.

The adduct involving an ethoxide anion of type **5** has been described earlier.[7] Judging from the ^1H NMR and IR spectral data and the procedure of the adduct preparation, the authors of the above paper probably dealt with the same compound that we obtained (the sodium salt instead of the potassium salt, **5c**). However, it was identified as a σ-adduct whose EtO group is attached to the carbon atom linked to the azide group (**6**).

The mechanism of the formation of adducts **5a—k** proposed by us (see Scheme 1) was confirmed by the transformation of tetrazolopyridine (**3**) in alcoholic media. For example, according to the ^1H NMR spectral data, a solution of compound **3** in CD_3OD contains three equilibrium forms: the azido form **2**, the tetrazole form **3**, and the dominating tetrazole form **4** bonded to the alcohol. The ratio of these forms calculated from the integral intensities of the proton signals is 1.0 : 2.5 : 7.0. Since the type **4** forms are strong acids, the positions of the H^+ signals in the ^1H NMR spectra cannot be determined. Treatment of solutions of equilibrium mixtures of **2** + **3** + **4** in the corresponding alcohols with various potassium salts (Bu^tOK, AcOK, KN_3, CF_3CO_2K) results in precipitates of type-**5** complexes. It should be noted that sodium salts, unlike potassium salts, form more soluble complexes.

The addition of an alcohol to tetrazolopyridine **3** is facilitated by the high electron-accepting ability of the dinitrotetrazolopyridine system. Also, it cannot be ruled out that the formation of an intramolecular ROH...O_2N—H-bond and the formation of a six-centered transition state (as shown in Scheme 1) also facilitate the reaction.

One of the methods of preparation of σ-adducts **5** (method ***A***) is to carry out the transformation of 2-chloro-3,5-dinitropyridine in the corresponding alcohol with heating or at ~20 °C. The reactions involving MeOH or EtOH are completed in 5—10 min, and those with other alcohols require somewhat longer times. In all cases, the reactions should be carried out for 4—6 h or left overnight to increase the yield (by 10—15%) and to obtain well-formed crystals requiring no further purification.

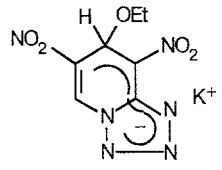
According to method ***B***, the reaction is carried out in acetone in the presence of a two- or threefold amount of the alcohol. In both cases, the yields of the products were rather high and sometimes approached 100 %.

All of the complexes obtained are stable when stored or when heated at 250 °C for a short time. Nevertheless, to prevent an explosion, they should be handled with care due to their friction, impact, and flame sensitivity.

The formation of a precipitate of the respective tetrabutylammonium salt may be used as a qualitative test for complexes **5** when aqueous solutions of **5** and Bu_4NBr are mixed together. The tetrabutylammonium salts were also characterized; they are easily soluble both in ether and in the corresponding alcohols and readily undergo crystallization. Their boiling points can be used as qualitative characteristics of the complexes.

Tables 1 and 2 present the physicochemical characteristics and spectral data for complexes **5a—k**.

The structure of complex **5** can be proven using **5c** as an example. According to the elemental analysis data, the composition of the complex is dinitrotetrazolopyridine · EtOK (1 : 1). For this reason, in addition to structures **5c** and **6**, the alternative structures **7** (addition at C(5)) and **8** (with pyridine ring opening, see, *e.g.*, Ref. 8) are also possible.



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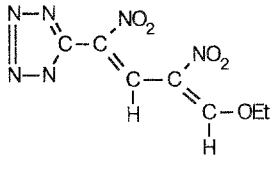

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One can choose between the above structures by looking at the NMR and IR spectral data. According to the ^{13}C NMR spectrum, the molecule contains two CH fragments, one of which gives a signal strongly shifted upfield (132.87 and 85.39 ppm for C(5) and C(7) in structure **5**, respectively). The IR spectrum does not contain the band in the 2110 cm^{-1} region characteristic of the N_3 group. These data permit us to exclude structure **6** from consideration completely. Most probably, the presence of this band in the IR spectrum reported previously[7] was caused by an admixture of NaN_3 rather than by the azido group of the complex. The δ values for C(5) and C(7) are inconsistent with structure **7**. Moreover, this structure is improbable because the nitro group at C(6) is "excluded" from the conjugation chain. The ^1H NMR spectrum shows the nonequivalence of the methylene protons of the ethoxy group attached to the asymmetric center. This allows us to rule out structure **8**. On the other hand, the value of the non-equivalence provides evidence against structure **7**, since the nearest environment at C(5) in this structure differs insignificantly.

The ^1H NMR spectrum of compound **5c** displays signals of the protons of the CH_2O group as an

Table 1. Parameters of anionic σ-complexes of 4,6-dinitrotetrazolo[1,5-*a*]pyridine with alkoxide anions

Compound	R	Preparation method	Yield (%)	M.p. /°C	Found / Calculated (%) C	H	N	K	Molecular formula
5a	H	***B***	74	—	22.33 22.56	1.20 1.14	31.80 31.51	15.00 14.69	$C_5H_3N_6O_6K$
5a[a]		***C***	86	117—118	54.11 53.71	8.42 8.37	22.23 20.88	— —	$C_5H_3N_6O_5 \cdot C_{16}H_{36}N$
5b	Me	***A***	96	—	25.72 25.71	1.60 1.80	30.12 29.99	14.02 13.95	$C_6H_5N_6O_5K$
5b[a]		***C***	95	80—81	54.72 54.64	9.07 8.55	20.27 20.27	— —	$C_6H_5N_6O_5 \cdot C_{16}H_{36}N$
5c	Et	***A***	89	—	28.43 28.57	2.36 2.40	28.64 28.56	13.40 13.29	$C_7H_7N_6O_5K$
5c[a]		***C***	95	80—81	55.64 55.51	8.68 8.71	19.67 19.70	— —	$C_7H_7N_6O_5 \cdot C_{16}H_{36}N$
5d	Pr^i	***A***	79	—	30.71 30.67	2.92 2.90	27.05 26.82	12.30 12.48	$C_8H_9N_6O_5K$
5e	Am^i	***B***	82	—	35.69 35.71	3.62 3.89	25.15 24.99	11.65 11.62	$C_{10}H_{13}N_6O_5K$
5f	*cyclo*-C_6H_{11}	***A***	81	—	37.65 37.93	3.84 3.76	24.24 24.12	11.10 11.22	$C_{11}H_{13}N_6O_5K$
5g	$C_{12}H_{25}$	***B***	94	—	46.34 46.99	6.24 6.26	19.33 19.34	8.75 9.00	$C_{17}H_{27}N_6O_5K$
5h	$ClCH_2CH_2$	***A***	95	—	25.60 25.58	1.81 1.84	25.58 25.57	12.03 11.90	$C_7H_6O_5ClN_6K$
5h[a]		***C***	98	106—107	49.85 50.81	8.12 8.14	19.01 18.55	— —	$C_7H_6O_5ClN_6 \cdot C_{16}H_{36}N$
5i	$PhCH_2$	***B***	95	—	39.68 40.45	2.73 2.95	23.18 23.58	10.50 10.90	$C_{12}H_9N_6O_5K$
5j	Ph[b]	***B***	53	—	— 38.60	— 2.06	— 23.37	— 11.40	$C_{10}H_7N_6O_5K$
5k	$HCCCH_2$	***B***	71	—	31.6 31.58	1.54 1.66	27.53 27.62	12.80 12.80	$C_8H_5N_6O_5K$

[a] Bu_4N^+ as the counter ion; in other cases, K^+. [b] The pure sample was not obtained.

ABX_3-system, which transforms to an AB-system ($\Delta\delta$ = 0.03 ppm) after decoupling from the protons of the methyl group. The methyl groups in complex **5d** (R = Pr^i) are also nonequivalent ($\Delta\delta$ = 0.2 ppm). The diastereotopic effect is observed even for the methyl groups in compound **5e** (R = Am^i), but to a smaller degree ($\Delta\delta$ = 0.01 ppm).

The position of the alkoxy group in σ-adducts **5** was confirmed unambiguously by X-ray diffraction analysis of complex **5b** (R = Me). The spatial configuration of one of the enantiomers is shown in Fig. 1. The atomic coordinates, bond lengths, and bond angles are presented in Tables 3—5 (the atoms are numbered in the order provided by X-ray analysis).

The five-membered aromatic ring is practically planar (the deviation from planarity is no more than 0.001(9) Å). The bond lengths and bond angles are similar to those in tetrazolo[1,5-*b*]pyridazine.[9] The six-membered ring has a chair conformation, in which the

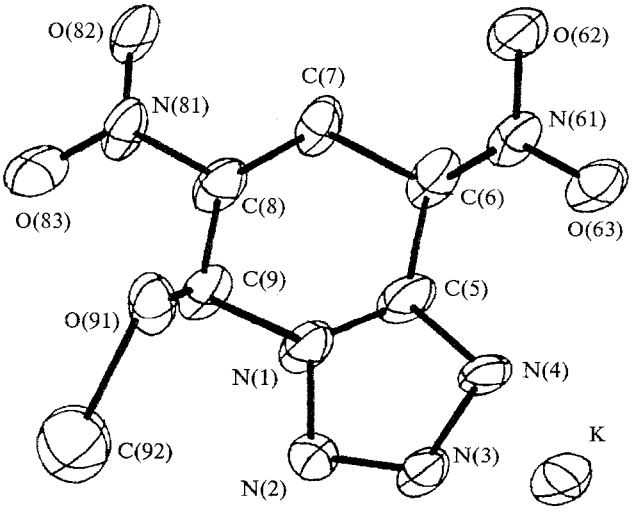

Fig. 1. Spatial structure of **5b**.

Table 2. Spectra of anionic σ-complexes

Compound	IR (ν/cm^{-1})	^{1}H NMR (δ, *J*/Hz)[a]		
		H(5)	H(7)	R
5a		8.68	7.25	
5a[b]	1599, 1580, 1567, 1542 ($\nu_{as}(NO_2)$); 3346 (OH); 3380, 3200 (fr. OH)	8.94[c]	7.62	9.34 (s, 1 H, OH)
5b	1578, 1575, 1548 ($\nu_{as}(NO_2)$)	8.63	7.21	3.41 (s, 3 H, MeO)
5b[b]		8.72 ($J_{5,7}$ = 0.8)	7.17 ($J_{5,7}$ = 0.8)	3.79, 3.76 (q.q, 2 H, CH_2, $J_{A,B}$ = −12); 1.08 (s, 3 H, Me)
5c	1583, 1550 ($\nu_{as}(NO_2)$)	8.83[d] ($J_{5,7}$ = 0.7)	7.14 ($J_{5,7}$ = 0.7)	3.84, 3.88 (q.q, 2 H, CH_2, $J_{A,B}$ = −12); 1.11 (s, 3 H, Me)
5c[b]	1570, 1547 ($\nu_{as}(NO_2)$)	9.01[c]	7.14	3.94, 3.91 (q.q, 2 H, CH_2, $J_{A,B}$ = −12); 1.11 (s, 3 H, Me)
5d	1590, 1545 ($\nu_{as}(NO_2)$)	8.76	7.26	4.25 (m, 1 H, CH); 1.23, 1.03 (d.d, 6 H, 2 Me)
5e	1580, 1541 ($\nu_{as}(NO_2)$)	8.71	7.15	3.5—3.9 (m, 2 H, CH_2O–); 1.2—1.4 (m, 9 H, Am^i)
5f	1590, 1545 ($\nu_{as}(NO_2)$)	8.73	7.28	
5g	1581, 1543 ($\nu_{as}(NO_2)$)	8.89 ($J_{5,7}$ = 0.61)	7.14 ($J_{5,7}$ = 0.61)	3.77 (m, CH_2O); 1.27 (s, 2 OH, $(CH_2)_{10}$); 0.88 (m, Me)
5h	1594, 1550 ($\nu_{as}(NO_2)$)	8.75	7.33	3.70 (m, α-CH_2); 4.00 (m, β-CH_2)
5i	1589, 1545 ($\nu_{as}(NO_2)$)	8.76 ($J_{5,7}$ = 0.75)	7.37 ($J_{5,7}$ = 0.75)	7.28 (s, Ph); 4.77 (s, CH_2)
5j		8.78	7.20	6.5—7.3 (m, Ph)
5k	1583, 1546 ($\nu_{as}(NO_2)$); 3289 (≡CH); 2127 (C≡C)	8.75	7.33	4.37 (d, *J* = 1.9, CH_2); 3.36 (t, *J* = 1.9, HC≡)

[a] In DMSO-d_6. [b] Bu_4N^+ as the counter ion. The integral intensities of signals of the tetrabutylammonium group correspond to a 1 : 1 complex; the values of δ are available from the authors. [c] In $CDCl_3$. [d] In acetone-d_6.

Table 3. Coordinates of atoms (×10^4) in structure **5b**

Atom	*x*	*y*	*z*	B_{eq}	Atom	*x*	*y*	*z*	B_{eq}
K	7787(2)	4149(2)	5363(3)	3.01(8)	O(63)	−1454(6)	704(7)	3390(10)	4.06(30)
N(1)	3444(7)	176(7)	2055(10)	2.64(27)	C(7)	427(9)	−1176(9)	1523(11)	2.98(33)
N(2)	4873(7)	1106(8)	2296(11)	3.26(31)	C(8)	1429(9)	−2052(8)	631(11)	2.69(31)
N(3)	4778(8)	2581(8)	3378(11)	3.65(33)	N(81)	806(8)	−3509(7)	−866(10)	3.12(31)
N(4)	3368(7)	2666(7)	3851(10)	3.11(29)	O(82)	−564(7)	−3921(6)	−1290(9)	3.94(27)
C(5)	2522(8)	1099(8)	2980(11)	2.49(30)	O(83)	1747(8)	−4210(8)	−1723(9)	4.35(30)
C(6)	978(8)	406(8)	2793(11)	2.45(31)	C(9)	3107(9)	−1635(8)	989(11)	2.78(33)
N(61)	−60(7)	1291(7)	3697(10)	3.02(30)	O(91)	3723(6)	−2775(6)	2005(8)	3.24(26)
O(62)	462(6)	2691(6)	4845(9)	3.44(26)	C(92)	5266(11)	−2969(12)	1847(16)	4.89(51)

Table 4. Bond lengths in structure **5b**

Bond	*d*/Å	Bond	*d*/Å
N(2)—N(1)	1.356(9)	O(62)—N(61)	1.267(8)
C(5)—N(1)	1.343(10)	O(63)—N(61)	1.240(8)
C(9)—N(1)	1.476(8)	C(8)—C(7)	1.366(12)
N(3)—N(2)	1.312(9)	N(81)—C(8)	1.457(9)
N(4)—N(3)	1.354(11)	C(9)—C(8)	1.460(11)
C(5)—N(4)	1.364(8)	O(82)—N(81)	1.201(9)
C(6)—C(5)	1.389(10)	O(83)—N(81)	1.247(10)
N(61)—C(6)	1.399(10)	O(91)—C(9)	1.423(9)
C(7)—C(6)	1.415(9)	C(92)—C(91)	1.412(11)

Table 5. Bond angles in structure **5c**

Angle	φ/deg	Angle	φ/deg
C(5)—N(1)—N(2)	110.5(6)	O(63)—N(61)—C(6)	120.5(6)
C(9)—N(1)—N(2)	120.7(7)	O(63)—N(61)—C(62)	121.1(7)
C(9)—N(1)—C(5)	128.7(6)	C(8)—C(7)—C(6)	120.0(7)
N(3)—N(2)—N(1)	104.0(6)	N(81)—C(8)—C(7)	118.7(6)
N(4)—N(3)—N(2)	113.6(6)	C(9)—C(8)—C(7)	126.1(6)
C(5)—N(4)—N(3)	104.2(6)	C(9)—C(8)—N(81)	115.2(7)
N(4)—C(5)—N(1)	107.6(7)	O(82)—N(81)—C(8)	120.0(7)
C(6)—C(5)—N(1)	118.7(6)	O(83)—N(81)—C(8)	117.0(6)
C(6)—C(5)—N(4)	133.5(7)	O(83)—N(81)—O(82)	122.8(6)
N(61)—C(6)—C(5)	122.2(6)	C(8)—C(9)—N(1)	105.4(6)
C(7)—C(6)—C(5)	118.1(8)	O(91)—C(9)—N(1)	109.6(6)
C(7)—C(6)—N(61)	119.5(6)	O(91)—C(9)—C(8)	109.4(7)
O(62)—N(61)—C(6)	118.3(6)	C(92)—O(91)—C(9)	113.6(8)

C(6) and C(9) atoms deviate by 0.057 Å and 0.220 Å, respectively, from the plane of the four remaining atoms. The bond lengths attest to increased electron density at the N(1)—C(5), C(7)—C(8), C(6)—N(61), and N(81)—O(82) bonds. The methoxy group is in the *endo* position. The increased C(5)—N(1)—C(9) and C(7)—C(8)—C(9) angles and their deviation from the theoretical value (120°) result from the sp^3-nature of the C(9) atom. It follows from the data obtained that the N(3), O(62), O(63), and O(63) atoms located at distances of 2.898(8), 2.795(7), 3.045(8) and 2.861(7) Å, respectively, from the potassium atom (the above atoms are coordinated to different K atoms of the crystal lattice) make the greatest contributions to the ionic interactions with this atom. The shortest distance is observed between the O(62) and K atoms, which results in an increase in the N(62)—O(62) bond length. The K ion almost does not interact with the O(82) atom, since the latter forms an almost double bond with the N atom.

It follows from the data obtained that the delocalization of the electron density in type-**5** complexes involves both constituents of the fused heterocyclic system, which explains the enhanced stability of these structures.

Experimental

IR spectra were recorded in vaseline oil on a Specord 75-IR spectrophotometer. ^{1}H NMR spectra were recorded in DMSO-d_6 using a Tesla BS-475 (100 MHz) spectrometer. ^{13}C NMR spectra were recorded on a Bruker WP-300 spectrometer (75 MHz). Me_4Si was used as the internal standard.

X-Ray diffraction analysis. The crystals of complex **5b** grown by slow evaporation of a methanolic solution have triclinic syngony; at 298 K, a = 8.857(2), b = 7.906(2), c = 7.784(2) Å; α = 101.70(1), β = 97.75(2), γ = 96.72(1)°; V = 523.0(4) Å^3; Z = 2; space group P_1; d_x = 1.773 g cm^{-3}; F(000) = 282; λ(Mo-Kα) = 0.71069 Å.

A crystal fragment 0.08×0.07×0.1 mm in size was studied on a Philips PW-1100 diffractometer.

The lattice parameters were determined by automatic indexing of 25 reflections ($6 \le \theta \le 12$) and refined by the least-squares method.

The intensities were collected using the ω-scanning technique for Mo-Kα radiation monochromatized by a graphite monochromator. 1345 reflections were measured in the $2 \le \theta \le 25$ range; 1211 reflections were collected in the $I \ge 2.5\sigma(I)$ range.

Three reflections were checked every 2 h in order to test the orientation and intensity. The structure was solved using the Patterson method and the SHELX86 and DIRDIF84 programs (the least-squares method in the full-matrix approximation).

The positions of the H atoms were refined using the common isotropic thermal factor to R = 0.066 (R_w = 0.066) for all reflections observed. The number of refined parameters was 164.

Synthesis of σ-adducts 5a—k from 4,6-dinitrotetrazolo[1,5-*a*]pyridine. *Caution! The compounds described below are potentially explosive and sensitive to flame, impact, and friction. The reactions produce toxic HN_3.*

The procedures described below make it possible to obtain pure complexes without additional crystallization.

Method *A*. 2-Chloro-3,5-dinitropyridine (5.0 g, 24.6 mmol) was dissolved in the corresponding alcohol (50 mL), then KN_3 (4.0 g) was added with stirring. In low-molecular alcohols, the reaction started immediately, the solution warmed itself up a little and became orange, then yellow or orange crystals of the complexes precipitated. To obtain well-formed crystals, the solution was stirred for 5 h or left overnight, then the resulting precipitate was filtered off, washed with ether, cold water (10 mL), alcohol (5 mL), and once more with ether, and then dried in a vacuum. The yields and spectral data are presented in Tables 1 and 2.

Method *B*. The procedure is similar to method *A*, but a two- or threefold excess of the corresponding alcohol, phenol, or water in acetone (50 mL) was used. Usually, the precipitate was not formed during the reaction, so the complex was precipitated with ether and then isolated as described in method *A*.

Method *C*. Synthesis of tetrabutylammonium salts of anions 5. The reaction was carried out similarly to method *A*, but an equivalent amount of NaN_3 was added instead of KN_3. The easily soluble complexes obtained were precipitated with ether, filtered off, and dissolved in water (50 mL), then an aqueous solution of tetrabutylammonium bromide was added slowly. Low-melting salts, which readily crystallize from ether—hexane

mixtures, precipitated. To prepare these salts, one can use the solutions obtained when the products were washed by method ***A***, thus reducing the losses due to some solubility of the complexes in water.

^{13}C NMR (DMSO-d_6), δ: **5b**: 108.93 (d, C(4)); 129.4 (q, C(5)); 120.62 (t, C(6)); 82.58 (q, C(7)); 146.49 (d, C(9)); 56.5 (q, Me); **5c**: 112.82 (d, C(4)); 132.87 (q, C(5)); 125.42 (t, C(6)); 85.39 (q, C(7)); 150.37 (d, C(9)); 69.44 (t, CH_2O); 18.76 (q, Me)).

4,6-Dinitrotetrazolo[1,5-*a*]pyridine (3). Compound **5b** (3.51 g, 12.7 mmol) was added to anhydrous CF_3COOH (20 mL), the mixture was slightly heated until the precipitate that formed initially dissolved, and the reaction mixture was filtered and cooled. The resulting light-yellow precipitate was recrystallized from CF_3COOH and dried in a vacuum. The yield was 2.46 g (92 %), m.p. 118—119 °C (*cf.* Ref. 7: m.p. 121 °C); the mass-spectroscopic data are in agreement with the literature data.[7]

^1H NMR (a 5 % solution in CD_3OD), δ: **2**: 9.15 (d, 1 H, H(4), J = 2.5 Hz); 9.40 (d, 1 H, H(6), J = 2.5 Hz); **3**: 9.43 (d, 1 H, H(5), J = 1.63 Hz); 10.81 (d, 1 H, H(7), J = 1.63 Hz); **4** (R = CD_3): 8.85 (s, 1 H, H(5)); 7.23 (s, 1 H, H(7)). The ratio of products is **2** : **3** : **4** = 1.0 : 2.5 : 7.0.

The ^1H NMR spectrum in DMSO-d_6 is in agreement with the literature data;[7] in wet DMSO-d_6, additional signals of protons appear, which correspond to an adduct with water.

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References

1. I. E. Filatov, G. L. Rusinov, and O. N. Chupakhin, *Khim. Geterotsikl. Soedin.*, 1992, 1145 [*Chem. Heterocycl. Compd.*, 1992 (Engl. Transl.)].
2. G. Illuminati, *Adv. Heterocycl. Chem.*, 1964, **33**, 285.
3. F. Terrier, *Chem. Rev.*, 1988, **92**, 77.
4. G. Illuminati and F. Stegel, *Adv. Heterocycl. Chem.*, 1983, **34**, 305.
5. M. J. Strauss, *Chem. Rev.*, 1970, **70**, 667.
6. F. Terrier, *Nucleophilic Aromatic Displacement: The Influence of the Nitro Group*, VCH Publishers, Inc., New York, 1991.
7. C. K. Lowe-Ma, R. A. Nissan, and W. S. Wilson, *J. Org. Chem.*, 1990, **55**, 3755.
8. B. Stanovnik and M. Tisler, *Chimia*, 1971, **25**, 272.
9. L. Golic, I. Leban, B. Stanovnic, and M. Tisler, *Acta Cryst.*, 1978, **34**, 136.

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