Nitration of primary aminofurazans with aqueous nitric acid

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A convenient procedure for the synthesis of *N*-nitroaminofurazans by nitration of primary 3-amino-4-R-furazans (R = Me, NO_2, phenyl-, methyl-*NNO*-azoxy-, *tert*-butyl-*NNO*-azoxy-, *tert*-butyldiazenyl-, *etc*.) with 66—77% aqueous nitric acid was developed. Depending on the concentration of HNO_3, the reaction is carried out at a temperature from 18 to 55 °C, the yield of the products is 80—99%. 3-Nitramino-4-phenylfurazan with unsubstituted benzene ring was obtained by nitration of 3-amino-4-phenylfurazan.

Key words: nitramines, primary aminofurazans, nitration, *N*-nitroaminofurazans, nitric acid.

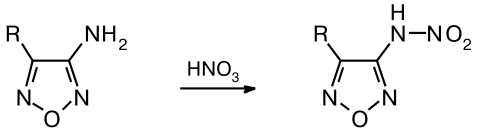
Monosubstituted nitramines of the furazan series among which compounds with high density are found attract chemists´ attention as energetic compounds[1—5] as well as starting compounds in the synthesis of 1,2,3,4-tetrazine-1,3-dioxides,[6—9] 1,2,3-triazole *N*-oxides,[10] and furazanocinnoline *N*-oxides.[11]

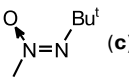
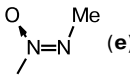
Several methods for synthesis of nitraminofurazans **1** are known. They can be obtained by either direct nitration of primary aminofurazans **2a—c** (see Refs 5—7, 10, 12 and Scheme 1) or nitration of aminofurazan derivative **3** with replacement of the leaving group R (see Ref. 3 and scheme 2), or destructive nitrolysis.[4]

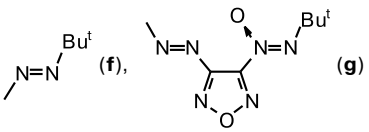
3-Amino-4-R-furazans **2** are weak bases, *e.g.*, 3-amino-4-nitrofurazan (pK_{BH^+} = –4.4 (see Ref. 13)) is close to dinitroaniline in basicity (pK_{BH^+} = –5.39 (see Ref. 14)). Therefore, in the nitration of aminofurazans **2** with nitric acid or its mixtures the extent of protonation of the amino group is low, which allows one to relatively easily obtain 3-nitramino-4-R-furazans **1** using the majority of nitration systems including acidic. Fuming nitric acid (d^{20} = = 1.5 g cm^{-3}), its solutions in organic solvents, mixtures with H_2SO_4 or oleum, as well as mixtures of H_2SO_4 with $NaNO_3$ or KNO_3, and mixtures of HNO_3 with P_2O_5, NO_2BF_4, N_2O_5 (see Refs 1—6, 11) are employed for N-nitration of primary aminofurazans. In most cases, nitraminofurazans **1** are isolated in yields not exceeding 70—80%.

Examples of quantitative N-nitration of aminofurazans **2** using dinitrogen pentoxide or nitration systems

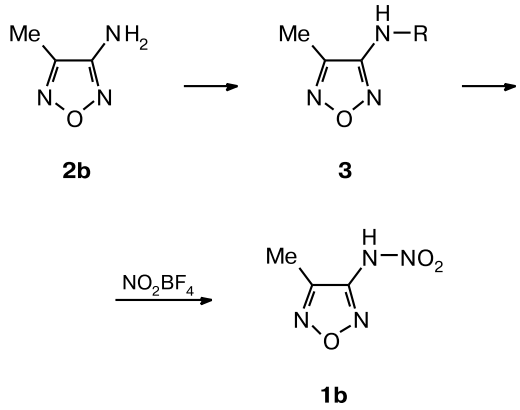
Scheme 1



R = NO_2 (**a**), Me (**b**),  (**c**), Ph (**d**),  (**e**),

 (**f**), (**g**)

Scheme 2



R = $SiMe_3$, BBu_2

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Table 1. Nitration of 3-amino-4-R-furazans **2a—g** with aqueous nitric acid

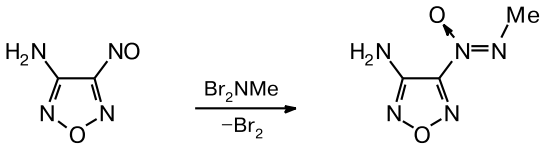
Amine **2**	Concentration of HNO_3 (%)	Amount of HNO_3 (g) per 1 g of compound **2**	T/°C[a]	τ/min[b]	Yield of compound **1** (%)	M.p. of nitramine **1**/°C[c]	
						The present study	Literature
a	71	10	30—33	120	70	48—54	47—48[4]
	77	6	20—23	60	83	55—56	(57, 60[d])[2]
b	71	8	22—24	60	72	40—42	
	77	8	15—18	25	92	44—50 (51—52[e])	88—89[3]
c	76	4	20—24	15	99	103—107	102—104[6]
d	70	10	20—24	120	90	74—75	69—71[11]
e	77	10	20—22	15	81	74—80 (80—81[f])	—
f	71	6	40—42	40	92	85—90 (88—91[g])	—
g[h]	71	10	40—44	30	95	85—88.5	—

[a] Nitration temperature. [b] Nitration time. [c] In parentheses, melting point of a product following crystallization is given. [d] After twofold recrystallization. [e] Nitramine **1b** obtained was purified by twofold recrystallization from CH_2Cl_2—petroleum ether. [f] From CH_2Cl_2—petroleum ether mixture. [g] From aqueous MeOH. [h] Procedure for the synthesis of compound **2g** (m.p. 94.5—95.5 °C) will be published later.

based on it are known.[2,3] In some cases,[3] preliminary protection of the primary amino group (silylation, borylation) followed by substitutive nitration of compound **3** with NO_2BF_4 affords ultimately the target product **1b** in 85—93% yield (see Scheme 2).

In the present work, we studied nitration of primary aminofurazans with nitric acid with different concentrations. Both the known nitramines **1a—d** and new compounds **1e—g** (see Scheme 1 and Table 1) were obtained. 3-Amino-4-(methyl-*NNO*-azoxy)furazan (**2e**), which is the starting compound for the synthesis of nitramine **1e**, was obtained according to the Kovacic procedure[15] from 3-amino-4-nitrosofurazan (**4**) and *N*,*N*-dibromomethylamine (Scheme 3).

Scheme 3



4 **2e**

We established that aqueous nitric acid is applicable for the nitration of 3-amino-4-R-furazans **2a—g**. N-Nitration is known to be the reversible process,[14,16] dilution of nitric acid reduces the rate of nitration and increases the rate of denitration.[14] The decrease in the rate constant for nitration is caused by a decrease in the nitronium cation activity due to its hydration. Presumably, 80% HNO_3 exists in the form of hydrate $HNO_3 \cdot H_2O$, whereas 60% HNO_3 exists in the form of dihydrate $HNO_3 \cdot 2H_2O$ (see Ref. 17). Intrinsically, it is aqueous nitric acid (60—80%) that is the nitrating agent in the N-nitration, which substantially differs from fuming nitric acid. We have shown that the concentrations of 66—71% (commercially available HNO_3) or 76—77% (the solution prepared by dilution of more concentrated HNO_3) are sufficient for the preparation of nitramines **1** in high yields. This fact is unexpected in the chemistry of aminofurazans in view of low nitrating activity of aqueous nitric acid and reversibility of N-nitration.

The procedure is experimentally easy, affords high yield of the product, and is free from expensive reagents. Nitration is carried out at room temperature (in the case of 76—77% HNO_3) or under moderate heating (in the case of 70—71% HNO_3). Nitramines **1c,d,f,g** precipitate in almost pure form upon completion of the reaction after dilution of the nitration mixture. Nitramines **1a,b** are isolated by extraction. It should be noted that the method proposed allows successful access to 3-nitramino-4-phenylfurazan (**1d**) with the nonnitrated aromatic ring in 90% yield. It is known[11] that the treatment of amine **2d** with a mixture H_2SO_4—KNO_3 results only in compounds with the nitrated benzene ring. Aryl substituents also undergo nitration in the reaction of phenylfurazans with an H_2SO_4—HNO_3 system.[18]

The yield of nitramine **1a** (83%) is close to that achieved using the procedure described earlier[2] (N_2O_5, CH_2Cl_2, −20—10 °C). The yield of nitramine **1b** (93%) significantly exceeds that attained in the nitration of amine **2b** with 100% HNO_3 (22%) and is close to that with the use of N_2O_5 or an HNO_3—P_2O_5 system (96%) (see Ref. 3).

Synthesis of compound **1e** is the only case where the yield of nitraminofurazan formed upon nitration with fuming HNO_3 (94%) is higher than that with aqueous nitric acid (yield 81%) (see Table 1).

Nitration of aminofurazans with 76—77% HNO_3 generally takes 15—25 min at nearly room temperatures. It is necessary to increase nitration time to 1 h in the case of furazans **2a,d**, which is probably caused by electron-withdrawing properties of the substituent adjacent to the amino group thus decreasing the amine basicity. The use of more dilute HNO_3, *viz.*, 68—71% (d^{20} = 1.396—1.418 g cm^{-3}) in most cases requires heating to 30—55 °C.

The influence of reaction conditions (concentration of HNO_3, temperature, percentage of N_2O_4) on the yield of product **1c** was studied by the example of nitration of amine **2c**. The nitration temperature depends on the concentration of HNO_3. After five minutes of nitration of amine **2c** with fuming HNO_3 (d^{20} = 1.5 g cm^{-3}, 4 g of HNO_3 per 1 g of **2a**, 0.3% N_2O_4, temperature interval −15—−10 °C), product **1c** was isolated in 90% yield. Extension of the nitration time leads to the reduction of the yield: after 20 min the yield is 87%, but after 120 min the yield is 78%.

Product **1c** was isolated in 92—94% yield in all the cases using 66—76% aqueous acid. The optimal concentration of HNO_3 in the case of nitration of amine **2c** at room temperature (18—25 °C) is 74—76%, the yield of **2c** being virtually quantitative under these conditions (see Table 1). Nitration of amine **2c** with 66—68% aqueous nitric acid at 55 °C for 30 min resulted in the product **1c** in 92—94% yield. The quality of nitramine is much worse when using nitric acid of lower concentration; therefore, it is not useful for nitration. The amount of HNO_3 in the interval from 4 to 10 g per 1 g of **2c** does not influence the yield.

Earlier,[3] it has been noted that the increase in the N_2O_4 percentage in fuming nitric acid in the case of nitration of 3-amino-4-methylfurazan (**2b**) considerably reduces the yield and the quality of compound **1b**. The negative influence of nitric oxides on nitration of primary aminofurazans was demonstrated by the example of aqueous nitric acid. Nitration of amine **2c** with 76% HNO_3 prepared on the basis of fresh distilled nitric acid (d^{20} = 1.5 g cm^{-3}), which was free of N_2O_4, resulted in 99% yield of **1c** (m.p. 103—107 °C (see Table 1)). If percentage of N_2O_4 is 1.8%, nitramine **2c** is isolated in a yield of 89% under the same conditions, but the melting point of the product obtained was significantly lower (94—101 °C).

Structure of all new compounds was confirmed by the combination of physicochemical methods.

Thus, a convenient one-step procedure for the synthesis of monosubstituted nitramines of the furazan series **1** based on nitration of primary aminofurazans **2** with aqueous nitric acid is developed.

Experimental

^{1}H, ^{13}C, and ^{14}N NMR spectra were recorded on Bruker AM 200, Bruker AM 300, and Bruker DRX 500 spectrometers in $CDCl_3$ and DMSO-d_6 using signals of the residual protons of the solvent (^{1}H, ^{13}C) and $MeNO_2$ (^{14}N) as the internal standards. The IR spectra were recorded on a Perkin—Elmer 684 instrument. The course of the reactions was monitored, and the purity of the compounds was checked, by TLC on Silica gel 60 F_{254} (Merck). Mass spectra were obtained on a Kratos MS-300 mass spectrometer (EI, 70 eV). Aminofurazans **2a**,[19] **2b**,[20] **2c**,[6] **2d**,[21] **2f**,[22] and **4**[23] were prepared according to the known procedures.

***N*,*N*-Dibromomethylamine.** Bromine (22.7 mL, 0.44 mol) was added dropwise to aqueous solution of NaOH (40 g (1 mol) of NaOH in 95 mL of water) over a period of 0.5 h maintaining the temperature at 0±3 °C. Then 35% aqueous solution of $MeNH_2$ (19.4 mL, 0.21 mol) in CH_2Cl_2 (80 mL) was added dropwise over a period of 20 min at 0±3 °C to the obtained solution of sodium hypobromite and the reaction mixture was stirred at 0—5 °C for 5 h. Dark red organic layer was separated and aqueous layer was extracted with CH_2Cl_2 (2×50 mL). The combined organic extracts were dried with $MgSO_4$. The solvent was removed *in vacuo* at the temperature not exceeding 30 °C. *N*,*N*-Dibromomethylamine was obtained as a dark red liquid with characteristic odor in a yield of 29.7 g (79%) (*cf.* Ref. 24: m.p. 10—11 °C). *The substance is unstable and possesses explosive properties.*

3-Amino-4-(methyl-*NNO*-azoxy)furazan (2e). *N*,*N*-Dibromomethylamine (6.5 g, 30 mmol) in CH_2Cl_2 (20 mL) was added to a stirred solution of 3-amino-4-nitrosofurazan (**4**) (3.0 g, 26 mmol) in MeCN (40 mL) at 10—20 °C, the reaction mixture was stirred at 20—25 °C for 1 h. The solvents and bromine evolved were removed *in vacuo*, 1,2-dichloroethane or the mixture of CCl_4—CH_2Cl_2 (1 : 1) (50 mL) was added to the residue, and the solution was concentrated *in vacuo*. Such a treatment was repeated twice. Product **2e** (3.0 g, 80%) was filtered off, colorless crystals of **2e** were obtained by crystallization from acetone in a yield of 2.1 g (56%). M.p. 124—126.5 °C. Found (%): C, 25.22; H, 3.50; N, 48.80. $C_3H_5N_5O_2$. Calculated (%): C, 25.18; H, 3.52; N, 48.94. MS, *m*/*z*: 143 $[M]^+$. IR (KBr), ν/cm^{-1}: 3420, 3320, 3250, 3200, 3030, 2920, 1640, 1515, 1485, 1440, 1380, 1355, 1200, 1140, 1045, 985. ^{1}H NMR (DMSO-d_6, 200 MHz), δ: 3.44 (s, 3 H, CH_3); 6.70 (s, 2 H, NH_2). ^{1}H NMR ($CDCl_3$, 200 MHz), δ: 3.55 (s, 3 H, CH_3); 5.25 (s, 2 H, NH_2). ^{13}C NMR (DMSO-d_6, 75 MHz), δ: 39.5 (CH_3); 150.9 (br. C=N). ^{14}N NMR ($CDCl_3$, 22 MHz), δ: −63.1 ((N→O), $\Delta\nu_{0.5}$ = 40 Hz). ^{14}N NMR (DMSO-d_6), δ: −63.1 ((N→O), $\Delta\nu_{0.5}$ = 140 Hz).

Nitraminofurazans 1a—g (general procedure). *A*. Aminofurazan **2** (10 mmol) was added with stirring to aqueous nitric acid (76—77%, d^{20} = 1.438—1.442 g cm^{-3}) at room temperature (18—25 °C) (4—10 g of HNO_3 per 1 g of **2**), the reaction mixture was stirred at the temperature indicated in Table 1, then poured onto ice (3—4 g per 1 g of HNO_3) and stirred. In the case of compounds **1c**,**d**,**f**,**g**, the precipitate that formed was filtered off, washed with ice water three times (1—3 mL per 1 g of the product), and dried on air. Water-soluble compounds **1a**,**b**,**e** were isolated by extraction of the aqueous solution with diethyl ether (10—15 mL of ether per 1 g of the product), the organic extracts were dried with $MgSO_4$, and the solvent was removed *in vacuo*.

B*.** Aminofurazan **2a**—**g** was added with stirring to aqueous nitric acid (70—71%, d^{20} = 1.413—1.420 g cm^{-3}) at 30—40 °C (5—10 g of HNO_3 per 1 g of **2**), the reaction mixture was stirred at this temperature. Subsequent work-up was analogous to that in the method ***A.

3-Nitramino-4-nitrofurazan (1a). Dichloromethane (two drops) and CCl_4 (1 mL) was added to the residue obtained after removal of diethyl ether, this mixture was kept at −20 °C for 6—12 h. Crystalline product **1a** was filtered off, washed with cold CCl_4 (2×5 mL). 3-Nitramino-4-nitrofurazan (**1a**) was obtained in yields of 83% (method ***A***) and 70% (method ***B***), m.p. 55—56 °C. MS, *m/z*: 175 $[M]^+$. ^1H, ^{13}C, and ^{14}N NMR spectra of compound **1a** are identical to those reported in the literature.[2,4]

3-Nitramino-4-methylfurazan (1b). Na_2CO_3 (4.4 g) was added portionwise to the reaction mixture poured onto ice. The solution obtained was extracted with diethyl ether, the organic extracts were dried, solvent was removed *in vacuo*. Three drops of CF_3COOH were added to the residue and the mixture obtained was kept at −20 °C for 6—12 h, then CCl_4 (1 mL) was added and crystalline product was filtered off. Nitramine **1b** was obtained in yields of 92% (method ***A***) and 72% (method ***B***), m.p. 44—50 °C (decomp.). MS, *m/z*: 144 $[M]^+$. ^1H, ^{13}C, and ^{14}N NMR spectra of compound **1b** are identical to those reported in the literature.[25]

4-(*tert*-Butyl-*NNO*-azoxy)-3-nitraminofurazan (1c). 3-Amino-4-(*tert*-butyl-*NNO*-azoxy)furazan (**2c**) (18.5 g, 0.1 mol) was added portionwise to 98.5% HNO_3 (55.5 g, d^{20} = 1.5 g cm^{-3}) at −17 °C in such a way that the temperature of the reaction mixture did not exceed −10 °C, the reaction mixture was stirred maintaining the temperature at −10 to −12 °C for 10 min and poured onto ice (300 g), stirred and kept for 0.5 h. The precipitate of nitramine **1c** was filtered off, washed with ice water (2×50 mL), and dried. Compound **1c** was obtained in a yield of 21.4 g (93%), m.p. 104—106 °C (decomp.). ^1H, ^{13}C, and ^{14}N NMR spectra of compound **1c** are identical to those reported in the literature.[6] IR (KBr), ν/cm^{-1}: 3290, 3000, 2870, 1620, 1590, 1510, 1490, 1480, 1460, 1450, 1370, 1315, 1300, 1270, 1240, 1160, 860, 820, 740.

The yield of compound **1c** prepared according to the method ***A*** was 99%, m.p. 103—107 °C.

3-Nitramino-4-phenylfurazan (1d) was prepared according to method ***B***. MS, *m/z*: 206 $[M]^+$. ^1H, ^{13}C, and ^{14}N NMR spectra of the compound are identical to those reported in the literature.[11]

4-(Methyl-*NNO*-azoxy)-3-nitraminofurazan (1e). 3-Amino-4-(methyl-*NNO*-azoxy)furazan (**2e**) (3.52 g, 0.025 mol) was added with stirring to 99.5% HNO_3 (18 g, 0.28 mol, d^{20} = 1.5 g cm^{-3}) over 2—3 min in such a way that temperature of the reaction mixture did not exceed −10 °C. The reaction mixture was stirred at −15±3 °C for 5 min and poured onto ice (55 g). The precipitate that formed was filtered off and washed with cold water (5 mL), 4-(methyl-*NNO*-azoxy)-3-nitraminofurazan (**1e**) was obtained in a yield of 2.89 r (62%), m.p. 80—81 °C (decomp.). The mother liquor was extracted with CH_2Cl_2 (3×50 mL), extracts were dried with $MgSO_4$, the solvent was removed *in vacuo*. An additional portion (1.46 g) of nitramine **1e** was obtained. The overall yield was 94%, m.p. 80—81 °C (decomp.). Found (%): C, 19.20; H, 2.18; N, 44.39. $C_3H_4N_6O_4$. Calculated (%): C, 19.16; H, 2.14; N, 44.68. MS, *m/z*: 188 $[M]^+$. IR (KBr), ν/cm^{-1}: 3260, 3040, 2950, 2920, 1615, 1580, 1510, 1500, 1450, 1430, 1375, 1315, 1270, 1190. ^1H NMR (DMSO-d_6, 200 MHz), δ: 3.38 (s, 3 H, CH_3); 10.6 (s, 1 H, NH). ^1H NMR ($CDCl_3$, 200 MHz), δ: 3.65 (s, 3 H, CH_3); 11.38 (s, 1 H, NH). ^{13}C NMR (DMSO-d_6, 75 MHz), δ: 40.0 (CH_3); 149.5 (C—NH); 155.0 (br. C—N→O). ^{14}N NMR ($CDCl_3$, 22 MHz), δ: −49.2 (NO_2, $\Delta\nu_{0.5}$ = 15 Hz); −67.1 ((N→O), $\Delta\nu_{0.5}$ = 40 Hz).

The yield of compound **1e** prepared according to the method ***A*** was 81%, m.p. 74—80 °C.

4-(*tert*-Butyldiazenyl)-3-nitraminofurazan (1f) was prepared according to the method ***B***. Found (%): C, 33.82; H, 4.66; N, 39.08. $C_6H_{10}N_6O_3$. Calculated (%): C, 33.65; H, 4.71; N, 39.24. MS, *m/z*: 214 $[M]^+$. IR (KBr), ν/cm^{-1}: 3256, 2980, 2936, 2872, 1612, 1584, 1524, 1492, 1476, 1448, 1364, 1348, 1308, 1200. ^1H NMR ($CDCl_3$, 300 MHz), δ: 1.44 (s, 3 H, CH_3); 11.31 (s, 1 H, NH). ^{13}C NMR ($CDCl_3$, 75 MHz), δ: 26.6 (C—$\underline{C}H_3$); 72.29 ($\underline{C}$—CH_3); 142.5 (CNH); 152.7 (C—N=N). ^{14}N NMR ($CDCl_3$, 22 MHz), δ: −47.2 (NO_2, $\Delta\nu_{0.5}$ = 35 Hz).

4-(*tert*-Butyl-*NNO*-azoxyfurazanyldiazenyl)-3-nitraminofurazan (1g) was prepared according to the method ***B***. Found (%): C, 29.47; H, 3.13; N, 42.78. $C_8H_{10}N_{10}O_5$. Calculated (%): C, 29.45; H, 3.09; N, 42.94. IR (KBr), ν/cm^{-1}: 3232, 2980, 2940, 1612, 1476, 1460, 1444, 1384, 1316, 1288, 1204. ^1H NMR ($CDCl_3$, 300 MHz), δ: 1.40 (s, 3 H, CH_3); 10.09 (s, 1 H, NH). ^{13}C NMR ($CDCl_3$, 75 MHz), δ: 25.2 (C—$\underline{C}H_3$); 62.4 ($\underline{C}$—CH_3); 141.6 (CNH); 151.9 (C—N(O)); 155.5, 155.9 (C—N=N). ^{14}N NMR ($CDCl_3$, 22 MHz), δ: −48.5 (NO_2, $\Delta\nu_{0.5}$ = 90 Hz); −75.7 ((N→O), $\Delta\nu_{0.5}$ = 210 Hz).

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