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ON THE NITRATION OF 5-PHENYL- AND 3-METHYL-5-PHENYLISOXAZOLES

Stefano Chimichi* and Barbara Cosimelli

Dipartimento di Chimica Organica e Centro CNR sulla Chimica e la Struttura dei composti eterociclici e loro applicazioni, Via Gino Capponi 9, I-50121 Firenze, Italy

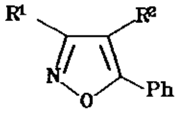
Abstract - The nitration of the title compounds **1** and **2** with HNO_3-H_2SO_4 was reexamined; 4-nitro-5-phenylisoxazole (**3**) and 3-methyl-4-nitro-5-phenylisoxazole (**4**) were obtained by treatment of **1** and **2** with HNO_3 in acetic anhydride under mild conditions.

The nitration of 5-phenylisoxazoles has been studied thoroughly in order to establish the nature of the reaction products.[1-4] Nevertheless, a recent review on nitroisoxazoles[5] indicates contradictory statements in the literature about the nitro compounds derived from the reaction of 5-phenylisoxazole (1) with HNO_3-H_2SO_4. Whereas Kochetkov and Khomutova[1] claimed that compound **1** yields both 4-nitro-5-phenylisoxazole (**3**) and 5-*p*-nitrophenylisoxazole (**5a**), Lynch and Shiu[2] reported that compound **5a** was the sole mononitro product and that is further nitrated to **6a**; these latter results were confirmed by Sokolov and Yudintseva[3] which showed that the reaction mixture contained **5a** as the main product together with a little amount of 5-*m*-nitrophenyl- (**5b**) and 4-nitro-5-*p*-nitrophenylisoxazole (**6a**).

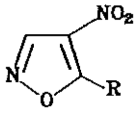
Following our interest in the 4-nitroisoxazoles,[6] we required the still unknown derivatives **3** and **4**; we therefore investigated again the nitration of compounds **1** and **2** with different nitrating agents.

In order to avoid the presence of regioisomers in the starting materials, we prepared compounds **1** and **2** by univocal methods according to Moureu[7] and Ceresole,[8] respectively. Several repetitions of Kochetkov and Khomutova's experiment under the same conditions (See Experimental) always gave a solid containing a 1:1:1 mixture of the mononitro derivatives **5a-c** as the predominant component together with minor amounts of the dinitro compounds **6a,b**, which were separated by flash chromatography.

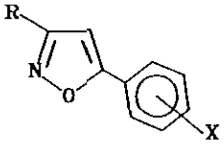
The presence of three compounds in the main reaction product (single spot in tlc) was revealed by an accurate high-field ^{1}H-nmr analysis (Table 1). The spectrum showed three signals at δ 6.723, 6.703 and 6.517 ppm easily attributable to the protons at position 4 of the isoxazole nucleus of compounds **5a-c**, respectively, which resonate at low frequency with respect to the aromatic ones and appear as doublets owing to the coupling with H-3. Fractional crystallizations from methanol led us to obtain analytically pure isomer **5a**, whereas compounds **5b** and **5c** were isolated at ca. 90% of purity due to their very similar physico-chemical properties.



1 $R^1 = R^2 = H$
2 $R^1 = Me, R^2 = H$
3 $R^1 = H, R^2 = NO_2$
4 $R^1 = Me, R^2 = NO_2$



6a $R = p\text{-}NO_2\text{-}C_6H_4$
6b $R = m\text{-}NO_2\text{-}C_6H_4$



	R	X
5a	H	$4'\text{-}NO_2$
5b	H	$3'\text{-}NO_2$
5c	H	$2'\text{-}NO_2$
7a	Me	$4'\text{-}NO_2$
7b	Me	$3'\text{-}NO_2$
7c	Me	$2'\text{-}NO_2$

The structures of compounds **6a** and **6b**, containing two NO_2 groups (mass spectra), follow from their ^{1}H-nmr patterns for which no H-4 resonance was observed between δ 6.0 and 7.0. Furthermore, whereas the former shows a characteristic AA'BB' pattern for the phenyl protons, thus proving the *para* position of the aromatic nitro group, the latter, as well as that of compound **5b**, is more complex. Selective homonuclear decoupling experiments and chemical shift arguments (Table 1) led us to formulate compounds **5b** and **6b** as 5-*m*-nitrophenyl- and 4-nitro-5-*m*-nitrophenylisoxazole, respectively.

When 3-methyl-5-phenylisoxazole (**2**) was allowed to react under the same conditions, we obtained a solid containing the *p*-, *m*-, and o-nitrophenyl derivatives (**7a-c**); dinitro compounds were now completely absent.

Table 1. ^{1}H-nmr data of compounds 1-7, 9-11

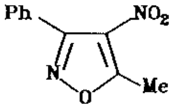
Compound	*^{1}H-nmr (δ, ppm, $CDCl_3$)*
1	6.528 (d, 1H, $^{3}J_{H4\text{-}H3} = 1.9$ Hz, 4-H), 7.430-7.510 (m, 3H, ArH_3), 7.780-7.820 (m, 2H, ArH_2), 8.295 (d, 1H, $^{3}J_{H3\text{-}H4} = 1.9$ Hz, 3-H)
2	2.362 (s, 3H, 3-Me), 6.370 (s, 1H, 4-H), 7.434-7.462 (m, 3H, ArH_3), 7.746-7.778 (m, 2H, ArH_2)
3	7.547-7.644 (m, 3H, ArH_3), 7.997-8.030 (m, 2H, ArH_2), 8.888 (s, 1H, 3-H)
4	2.635 (s, 3H, 3-Me), 7.557-7.627 (m, 3H, ArH_3), 7.889-7.922 (m, 2H, ArH_2)
5a	6.723 (d, 1H, $^{3}J_{H4\text{-}H3} = 1.9$ Hz, 4-H), 8.171 (m AA'BB', 4H, ArH_4), 8.381 (d, 1H, $^{3}J_{H3\text{-}H4} =$ 1.9 Hz, 3-H)
5b	6.703 (d, 1H, $^{3}J_{H4\text{-}H3} = 1.9$ Hz, 4-H), 7.700 (dd, 1H, $^{3}J_{H5'\text{-}H4'} = 8.3$ Hz, $^{3}J_{H5'\text{-}H6'} = 7.8$ Hz, 5'-H), 8.150 (ddd, 1H, $^{3}J_{H6'\text{-}H5'} = 7.8$ Hz, $^{4}J_{H6'\text{-}H2'} = 1.8$ Hz, $^{4}J_{H6'\text{-}H4'} = 1.0$ Hz, 6'-H), 8.310 (ddd, 1H, $^{3}J_{H4'\text{-}H5'} = 8.3$ Hz, $^{4}J_{H4'\text{-}H2'} = 2.1$ Hz, $^{4}J_{H4'\text{-}H6'} = 1.0$ Hz, 4'-H), 8.375 (d, 1H, $^{3}J_{H3\text{-}H4} = 1.9$ Hz, 3-H), 8.640 (dd, 1H, $^{4}J_{H2'\text{-}H4'} = 2.1$ Hz, $^{4}J_{H2'\text{-}H6'} = 1.8$ Hz, 2'-H)
5c	6.517 (d, 1H, $^{3}J_{H4\text{-}H3} = 1.9$ Hz, 4-H), 7.612-7.934 (m, 4H, ArH_4), 8.351 (d, 1H, $^{3}J_{H3\text{-}H4} =$ 1.9 Hz, 3-H)
6a	8.338 (m AA'BB', 4H, ArH_4), 8.963 (s, 1H, 3-H)
6b	7.823 (dd, 1H, $^{3}J_{H5'\text{-}H4'} = 8.3$ Hz, $^{3}J_{H5'\text{-}H6'} = 8.0$ Hz, 5'-H), 8.375 (ddd, 1H, $^{3}J_{H6'\text{-}H5'} = 8.0$ Hz, $^{4}J_{H6'\text{-}H2'} = 1.8$ Hz, $^{4}J_{H6'\text{-}H4'} = 1.0$ Hz, 6'-H), 8.515 (ddd, 1H, $^{3}J_{H4'\text{-}H5'} = 8.3$ Hz, $^{4}J_{H4'\text{-}H2'} = 2.1$ Hz, $^{4}J_{H4'\text{-}H6'} = 1.0$ Hz, 4'-H), 8.920 (dd, 1H, $^{4}J_{H2'\text{-}H4'} = 2.1$ Hz, $^{4}J_{H2'\text{-}H6'} =$ 1.8 Hz, 2'-H), 8.966 (s, 1H, 3-H)
7a	2.401 (s, 3H, 3-Me), 6.557 (s, 1H, 4-H), 8.133 (m AA'BB', 4H, ArH_4)
7b	2.401 (s, 3H, 3-Me), 6.535 (s, 1H, 4-H), 7.668 (dd, 1H, $^{3}J_{H5'\text{-}H4'} = 8.1$ Hz, $^{3}J_{H5'\text{-}H6'} = 7.8$ Hz, 5'-H), 8.097 (ddd, 1H, $^{3}J_{H6'\text{-}H5'} = 7.8$ Hz, $^{4}J_{H6'\text{-}H2'} = 1.6$ Hz, $^{4}J_{H6'\text{-}H4'} = 1.1$ Hz, 6'-H), 8.279 (ddd, 1H, $^{3}J_{H4'\text{-}H5'} = 8.1$ Hz, $^{4}J_{H4'\text{-}H2'} = 2.3$ Hz, $^{4}J_{H4'\text{-}H6'} = 1.1$ Hz, 4'-H), 8.588 (dd, 1H, $^{4}J_{H2'\text{-}H4'} = 2.3$ Hz, $^{4}J_{H2'\text{-}H6'} = 1.6$ Hz, 2'-H)
7c	2.401 (s, 3H, 3-Me), 6.337 (s, 1H, 4-H), 7.578-7.892 (m, 4H, ArH_4)
9	2.600 (s, 3H, 3-Me), 9.229 (s, 1H, 5-H)
10	2.859 (d, 3H, $^{5}J_{Me\text{-}H3} = 0.6$ Hz, 5-Me), 8.743 (q, 1H, $^{5}J_{H3\text{-}Me} = 0.6$ Hz, 3-H)
11	2.534 (s, 3H, 3-Me), 2.800 (s, 3H, 5-Me)

In the light of these findings and bearing in mind the possibility of achieving a preferential nitration at position 4 for 3-phenyl-[9] and 3,5-diphenylisoxazoles[10] with Ac_2O-HNO_3, we investigated the possibility of obtaining **3** and **4** by this route. Thus, when the isoxazole **1** was allowed to react with nitric acid in acetic anhydride for 70 h at 5-10 °C, we obtained, besides some unreacted material and a mixture of the nitrophenyl derivatives **5a-c**, the desired compound **3** as the major component, whose structure follows from spectral evidence. In fact, as for compounds **6a,b**, the lack of any signal attributable to H-4 and the appearance of a weak resonance at δ 129.90 ppm (C-NO_2) in the ^{1}H- and ^{13}C-nmr spectra, respectively, together with the presence of a mono substituted phenyl ring (^{1}H-,

Table 2. **^{13}C-nmr data**

Compound	*^{13}C-nmr (δ, ppm, $CDCl_3$)*
1	98.51 (d, C-4, $^{1}J_{C4\text{-}H4}=181.6$ Hz, $^{2}J_{C4\text{-}H3}=9.45$ Hz), 125.615 (d, C_{meta}), 127.01 (s, C_{ipso}), 128.79 (d, C_{ortho}), 129.98 (d, C_{para}), 150.64 (d, C-3, $^{1}J_{C3\text{-}H3}=186.1$ Hz, $^{2}J_{C3\text{-}H4}=5.5$ Hz), 169.09 (s, C-5)
2	11.46 (q, Me-3, $^{1}J_{Me}=129.5$ Hz), 100.07 (d, C-4, $^{1}J_{C4\text{-}H4}=178.95$ Hz, $^{3}J_{C4\text{-}Me}=3.2$ Hz), 125.64 (d, C_{meta}), 127.47 (s, C_{ipso}), 128.83 (d, C_{ortho}), 129.89 (d, C_{para}), 160.27 (s, C-3), 169.51 (s, C-5)
3	123.97 (s, C_{ipso}), 128.91 (d, C_{ortho}), 129.59 (d, C_{meta}), 129.90 (s, C-4), 133.095 (d, C_{para}), 147.75 (d, C-3, $^{1}J_{C3\text{-}H3}=197.4$ Hz), 167.54 (s, C-5)
4	11.90 (q, Me-3, $^{1}J_{Me}=131.9$ Hz), 124.70 (s, C_{ipso}), 128.66 (d, C_{ortho}), 129.00 (s, C-4), 129.25 (d, C_{meta}), 132.54 (d, C_{para}), 156.62 (s, C-3, $^{2}J_{C3\text{-}Me}=7.1$ Hz), 168.65 (s, C-5)
5a	101.36 (d, C-4, $^{1}J_{C4\text{-}H4}=182.4$ Hz, $^{2}J_{C4\text{-}H3}=9.4$ Hz), 124.47 (d, C-2',6'/C-3',5'), 126.67 (d, C-3',5'/C-2',6'), 132.62 (s, C-1'), 148.495 (s, C-4'), 151.13 (d, C-3, $^{1}J_{C3\text{-}H3}=188.0$ Hz, $^{2}J_{C3\text{-}H4}=5.2$ Hz), 166.86 (s, C-5)
6a	124.01 (d, C-2',6'/C-3',5'), 129.38 (s, C-1'), 130.86 (d, C-3',5'/C-2',6'), 131.04 (s, C-4), 147.79 (d, C-3, $^{1}J_{C3\text{-}H3}=198.7$ Hz), 150.08 (s, C-4'), 165.00 (s, C-5)
6b	124.75 (d, C-5', $^{1}J_{C5'\text{-}H5'}=171.8$ Hz, $^{2}J_{C5'\text{-}H4'}={}^{2}J_{C5'\text{-}H6'}=5$ Hz), 125.44 (s, C-1'), 127.38 (d, C-6'/C-4', $^{1}J=170.0$ Hz, $^{2}J=7.9$ Hz, $^{3}J=1.7$ Hz), 130.275 (d, C-2', $^{1}J_{C2'\text{-}H2'}=167.8$ Hz), 130.77 (s, C-4), 135.10 (d, C-4'/C-6', $^{1}J=167.0$ Hz, $^{2}J=4.0$ Hz, $^{3}J=2.0$ Hz), 147.79 (d, C-3, $^{1}J_{C3\text{-}H3}=198.7$ Hz), 148.36 (s, C-3'), 164.89 (s, C-5)
7a	11.60 (q, Me-3, $^{1}J_{Me}=129.5$ Hz), 102.84 (d, C-4, $^{1}J_{C4\text{-}H4}=179.8$ Hz, $^{3}J_{C4\text{-}Me}=3.1$ Hz), 124.38 (d, C-2',6'/C-3',5'), 126.49 (d, C-3',5'/C-2',6'), 132.98 (s, C-1'), 148.35 (s, C-4'), 160.78 (s, C-3, $^{2}J_{C3\text{-}Me}=6.7$ Hz, $^{2}J_{C3\text{-}H4}=4.95$ Hz), 167.08 (s, C-5)
9	10.57 (q, Me-3, $^{1}J_{Me}=132.2$ Hz), 134.54 (s, C-4), 154.45 (s, C-3, $^{2}J_{C3\text{-}Me}=7.1$ Hz, $^{3}J_{C3\text{-}H5}=4.5$ Hz), 159.465 (d, C-5, $^{1}J_{C5\text{-}H5}=211.7$ Hz)
10	12.93 (q, Me-5, $^{1}J_{Me}=132.8$ Hz), 131.03 (s, C-4), 145.84 (d, C-3, $^{1}J_{C3\text{-}H3}=197.9$ Hz), 170.64 (s, C-5, $^{2}J_{C5\text{-}Me}=7.2$ Hz, $^{3}J_{C5\text{-}H3}=2.8$ Hz)
11	11.46 (q, Me-3, $^{1}J_{Me}=131.8$ Hz), 13.775 (q, Me-5, $^{1}J_{Me}=132.5$ Hz), 130.07 (s, C-4), 155.475 (s, C-3, $^{2}J_{C3\text{-}Me}=7.2$ Hz), 171.87 (s, C-5, $^{2}J_{C5\text{-}Me}=7.2$ Hz)

^{13}C-nmr, and mass spectra) and a nitro group (mass spectrum), unambiguously confirm the assigned structure. The same procedure applied to compound **2** to give 3-methyl-4-nitro-5-phenylisoxazole (**4**) as the largely predominant product, together with compounds **7a-c**. Compound **4** was previously reported by Russian workers[11] as the reaction product of 3-bromo-1-phenyl-1-butyne with sodium nitrite in DMF, but more recently Duranti and coworkers[12] unambiguously established the nature of this compound as the isomeric 5-methyl-4-nitro-3-phenylisoxazole (**8**). The physical properties of our product are sensibly different with respect to those reported for **8**; in particular, a careful comparison of its ^{1}H-nmr spectrum with those of **8** and well established model compounds (see below) allowed us to state that the chemical shift of the methyl group is indicative of its position on the 4-nitroisoxazole ring. Thus, the methyl resonance of compound **4** (2.635 ppm) is very close to that of 3-methyl-4-nitroisoxazole (**9**)[13] (2.602 ppm), whereas the reported value for the regioisomer (**8**) (2.85 ppm),[11,12] is identical with that of 5-methyl-4-nitroisoxazole (**10**)[13] (2.859 ppm).



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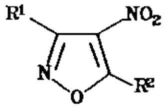

9 R^1 = Me, R^2 = H
10 R^1 = H, R^2 = Me
11 R^1 = R^2 = Me

Finally, 3,5-dimethyl-4-nitroisoxazole (**11**) exhibits two resonances at δ 2.801 and 2.511 ppm which are easily assigned to methyl groups at positions 5 and 3, respectively, by selective heteronuclear decouplings or by a 2D-nmr (HETCOR) experiment, thus confirming the above statement.

EXPERIMENTAL

All melting points were determined on a Büchi 510 melting point apparatus and are uncorrected. Infrared spectra were measured for potassium bromide discs with a Perkin-Elmer 283 spectrophotometer. ^{1}H- and ^{13}C-nmr spectra were recorded in $CDCl_3$ on a Varian VXR-300 instrument; chemical shifts are reported in ppm high frequency from tetramethylsilane as secondary reference standard and coupling constants in Hz. Mass spectra were obtained with a Hewlett-Packard 5790A instrument. Silica gel plates (Merck F_{254}) and silica gel 60 (Merck 230-400 mesh) were used for analytical tlc and for flash chromatographies, respectively. Solvents were removed under reduced pressure.

Nitration of Compounds **1** and **2** with HNO_3-H_2SO_4

a) 5-Phenylisoxazole **1** (1.6 g; 11.03 mmol) was dissolved dropwise in H_2SO_4 (5.4 ml; d = 1.84 g/l) at -10 °C, with stirring to minimize local overheating. A mixture of HNO_3 (2.4 ml; d = 1.49 g/l) in H_2SO_4 (3.6 ml; d = 1.84 g/l) was then added dropwise, the temperature being held between -5 and -2 °C. After addition was complete, the mixture was maintained for 1 h at 15-18 °C, then carefully poured into ice and filtered to give a solid (1.848 g) which was washed with water and dried. The raw material showing three spots in tlc was resolved by flash chromatography with ethyl acetate-petroleum ether (bp 40-70 °C) (1:4 v/v) as eluant. The fastest running band afforded compound **6a** (0.150 g, yield 6%), mp 119-120 °C (from methanol) [lit.[2] mp 115-117 °C (from ethanol)]. Ms: m/z (%) = 235 (39)[M^+], 166 (75), 150 (100), 120 (24), 104 (63), 92 (47), and 76 (85). Anal. Calcd for $C_9H_5N_3O_5$: C, 45.97; H, 2.14; N, 17.87. Found: C, 45.82; H, 2.23; N, 17.68.

The second band afforded a small amount of compound **6b** (0.120 g, yield 5%), mp 104-106 °C (from methanol). Ms: m/z (%) = 235 (29)[M^+], 166 (70), 150 (100), 104 (51), 92 (18), and 76 (68). Anal. Calcd for $C_9H_5N_3O_5$: C, 45.97; H, 2.14; N, 17.87. Found: C, 45.74; H, 2.27; N, 17.71.

The slowest component was identified (^{1}H- and ^{13}C-nmr) as a mixture of the three mononitrophenyl derivatives **5a-c**, (1.2 g, yield 57%). Fractional crystallizations from methanol gave pure compound **5a**, mp 167-168 °C (lit.[1] mp 171-172 °C, lit.[2] mp 176-178 °C , lit.[3] mp 163-165 °C). Ms: m/z (%) = 190 (100) [M^+], 150 (40), 144 (10), 104 (22), 76 (26), and 68 (9). Anal. Calcd for $C_9H_6N_2O_3$: C, 56.85; H, 3.18; N, 14.73. Found: C, 56.70; H, 3.09; N, 14.82.

b) Operating as described above, compound **2** (0.666 g; 4.19 mmol) afforded a white solid (0.795 g, yield 93%) containing the isomers **7a-c** (ca. 2:1:0.5, ^{1}H-nmr spectrum) which showed the same Rf and retention times in different eluants or chromatographyc columns, respectively. Fractional crystallizations from ethanol gave an analytical sample of 3-methyl-5-*p*-nitrophenylisoxazole (**7a**), mp 180-181 °C (lit.[14] 180 °C, lit.[4] 183-185 °C) and enriched (ca. 90%) samples of **7b** and **7c**. Anal. Calcd for $C_{10}H_8N_2O_3$: C, 58.82; H, 3.95; N, 13.72. Found: C, 58.96; H, 3.78; N, 13.56.

Nitration of Compounds **1** and **2** with HNO_3 in acetic anhydride

a) A solution of 5-phenylisoxazole **1** (0.63 g; 4.34 mmol) in acetic anhydride (2.2 ml) was cooled to 0 °C and HNO_3 (0.43 ml; d = 1.49 g/l) was added dropwise under stirring, the temperature being maintained between 0-

5 °C. After addition was complete, the mixture was stirred for 70 h at 5-10 °C and then poured into ice to give a gummy solid which was extracted with methylene chloride (4 x 40 ml). Evaporation to dryness of the extracts left a yellowish residue which was resolved into three components by flash chromatography with ethyl acetate-petroleum ether (bp 40-70 °C) (1:4 v/v) as eluant. The fastest band gave 4-nitro-5-phenylisoxazole (**3**) (0.185 g, yield 32%), mp 87-88 °C after crystallization from ethanol. Ms: m/z (%) = 190 (44)[M$^+$], 121 (86), 105 (41), 93(27), and 77 (100). Anal. Calcd for $C_9H_6N_2O_3$: C, 56.85; H, 3.18; N, 14.73. Found: C, 56.61; H. 3.12; N, 14.94.

The second compound was easily recognized (^{1}H- and ^{13}C-nmr) as unreacted 5-phenylisoxazole (0.190 g), whereas the slowest band afforded a solid (0.166 g, yield 29%) containing the isomers **5a** and **5c** as predominant products together with a small amount of the *m*-nitrophenyl derivative **5b** (^{1}H-nmr spectrum).

b) Operating under the same conditions, compound **2** (1 g; 6.29 mmol) afforded a yellow oily product which was extracted with methylene chloride (3 x 40 ml) and resolved into three components by flash chromatography with ethyl acetate-petroleum ether (bp 40-70 °C) (1:4 v/v). The first band afforded 3-methyl-4-nitro-5-phenylisoxazole (**4**) (0.63 g, yield 59%) as a yellow oil which solidifies on cooling, mp 31-32 °C (from methanol). Anal. Calcd for $C_{10}H_8N_2O_3$: C, 58.82; H, 3.95; N, 13.72. Found: C, 59.01; H, 4.01; N, 13.66.

The second compound was identified (^{1}H-nmr) as unreacted starting material (0.170 g), whereas the third band gave a solid (0.2 g, yield 19%) containing almost exclusively (^{1}H-nmr spectrum) compound **7a** together with small amounts of the regioisomers **7b** and **7c**.

REFERENCES

1. N. K. Kochetkov and E. D. Khomutova, *Zh. Obshch. Khim.*, **1958**, 28, 359 [*Chem. Abstr.*, **1958**, 52, 13710b].
2. B. M. Lynch and L. Shiu, *Can. J. Chem.*, **1965**, 43, 2117.
3. S. D. Sokolov and I. M. Yudintseva, *Zh. Org. Khim. (Engl. Transl.)*, **1968**, 4, 1988.
4. A. R. Katritzky, M. Konia, H. O. Tarhan, and A. G. Burton, *J. Chem. Soc., Perkin Trans. 2*, **1975**, 1627.
5. J. H. Boyer, 'Nitroazoles:the C-Nitro Derivatives of Five-Membered N- and N,O-Heterocycles', ed. by H. Feuer, VCH Publ., Deerfield Beach, Fl, 1986, pp. 301-340.
6. R. Nesi, S. Chimichi, P. Sarti-Fantoni, A. Buzzi, and D. Giomi, *Heterocycles*, **1985**, 23, 1465.
7. Ch. Moureu and R. Delange, *Compt. Rend.*, **1904**, 138, 1339.
8. M. Ceresole, *Ber.*, **1884**, 17, 812.

9. S. D. Sokolov, I. M. Yudintseva, P. V. Petrovskii, and V. G. Kalyuzhnaya, *Zhur. Org. Khim. (Engl. Transl.)*, **1971**, 7, 2051.

10. S. D. Sokolov, T. N. Egorova, and I. M. Yudintseva, *Khim. Geterotsikl. Soedin.*, **1974**, 5, 597.

11. Ts. D. Mechkov, I. G. Sulimov, N. V. Usik, I. Mladenov, and V. V. Perekalin, *Zhur. Org. Khim. (Engl. Transl.)*, **1980**, 16, 1148.

12. E. Duranti, C. Balsamini, G. Spadoni, and L. Staccioli, *J. Org. Chem.*, **1988**, 53, 2870.

13. L. A. Reiter, *J. Org. Chem.*, **1987**, 52, 2714.

14. C. Musante, *Gazz. Chim. Ital.*, **1942**, 72, 537.

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