Regio- and stereoselectivity of 1,3-dipolar cycloaddition of cyclic aldonitrones of the 3-imidazoline 3-oxide series to monosubstituted alkenes

*T. A. Berezina, V. A. Reznikov, V. I. Mamatyuk, P. A. Butakov, Yu. V. Gatilov, I. Yu. Bagryanskaya, and L. B. Volodarsky**

Novosibirsk Institute of Organic Chemistry, Siberian Branch of the Russian Academy of Sciences, 9 prosp. Akad. Lavrent'eva, 630090 Novosibirsk, Russian Federation. Fax: +7 (383 2) 35 4752

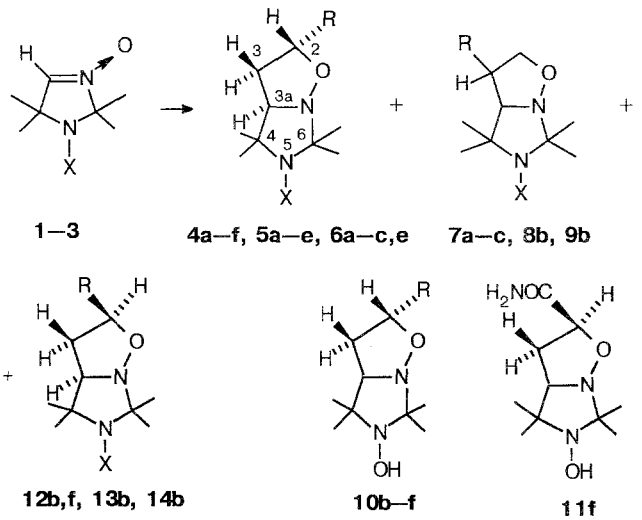
Regio- and stereoselectivity of 1,3-dipolar cycloaddition of cyclic aldonitrones of the 3-imidazoline 3-oxide series mainly depends on the type of the substituent in the dipolarophile. The configuration of the main cycloadduct has been determined, and a method has been suggested to establish the stereochemistry of the cycloaddition products by ^{1}H NMR spectroscopy. An increase in electron-acceptor properties of the substituent in the alkene molecule results in a decrease in the regio- and stereoselectivity of the cycloaddition.

Key words: aldonitrones, 1,3-dipolar cycloaddition; isoxazolidines; nitroxyl radical.

Previously it has been shown[1] that the reaction of cyclic aldonitrones, derivatives of 3-imidazoline 3-oxide (**1—3**), with monosubstituted alkenes occurs regio- and stereospecifically, which is characteristic of nitrones.[2—4] We found that the interaction of aldonitrone **1** with acrylonitrile (Scheme 1) results in the formation, in addition to cycloadduct **4b** described previously,[1] of its isomer **7b** (according to the data from elemental analysis and IR spectroscopy, the ratio of the isolated **4b** and **7b** is ~10 : 1). Taking into account the similarity of the IR spectra of **4b** and **7b** and the previously described[1] **5b**, which is the adduct of the cyclic aldonitrone **2** with acrylonitrile, it seems more correct to use a hydroxylamino derivative, which is the reducion product of the cycloadduct (nitroxyl radical), as a diamagnetic analog instead of **2** in order to establish the structure of cycloadducts formed from the paramagnetic substrate **1**. It is impossible to obtain **10b** by the reduction of **4b** due to the readily occurring reverse reaction, oxidation to the original radical. In this connection, nitrile **4b** was transformed into amide **4f**, whose reduction by hydrazine hydrate resulted smoothly in the formation of the corresponding stable *N*-hydroxy derivative **10f**. The analysis of the ^{1}H NMR spectrum of the product confirms that this is the 5-substituted isoxazolidine, and a double set of signals in the ^{13}C NMR spectrum attests to the formation of a mixture of two diastereomers, **10f** and **11f**, in the ratio ~1 : 1. This means that the original amide and nitrile were also mixtures of two diastereomers, **4f**, **12f** and **4b**, **12b**, respectively.

Thus, in the case of the interaction of aldonitrone **1** with acrylonitrile, regioselectivity of the process can be considered, but not regio- and stereospecificity (*cf.* Ref. 1). In this connection, the reactions of nitrones **1—3** with various monosubstituted alkenes have been studied in the present work.

The interaction of diamagnetic 3-imidazoline 3-oxide **3** with acrylonitrile results in the formation of a mixture of two stereoisomers, **6b** and **14b**, in the ratio ~3 : 1. In the ^{1}H NMR spectrum of the mixture, in addition to the signals of the main stereoisomers, signals that can be caused by the presence of the 4-regioisomer **9b** (< 10 %) are also observed.



1, 4, 7, 12: X = O
2, 5, 8, 13: X = Me
3, 6, 9, 14: X = NO_2
R = COOEt (**a**), CN (**b**), COMe (**c**), Ph (**d**), 4-py (**e**), $CONH_2$ (**f**)

Translated from *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 5, pp. 894—898, May, 1994.

Opening the isoxazolidine cycle to obtain products with an *a fortiori* known configuration[5,6] is one of the methods of the establishment of the stereochemistry of adducts of cyclic aldonitrones with alkenes. The information content of this method is obviously limited by the set of corresponding acyclic products of the heterocycle opening. One can also judge the stereochemistry of adducts by the presence or the absence of coupling constants for H-2 and H-3a protons,[7] however, the data on the values of spin-coupling constants for compounds of this class are scarce. The analysis of the ^{1}H NMR spectra of a mixture of **6b** and **14b** with consideration of the experiments of double resonance and iteration calculations by the PANIC program make it possible to determine the coupling constants in the four-spin proton system at positions 2, 3 and 3a (Table 1). The values of the dihedral angles (φ) for diastereomers **6b** and **14b** were calculated by the molecular mechanics method. From these data, the values of the vicinal constants $^3J_{H,H}$ were estimated by the Carplus formula, taking into account the effect of the difference in electronegativities (Δx) for the hydrogen atom and the substituents:

$$^3J_{H,H} = (A + B\cos\varphi + C\cos 2\varphi)(1 - 0.07\Delta x),$$

where A, B, C are empirical coefficients, $A = 7$, $B = -1$, $C = 5$.

The results of the estimation of the spin-coupling constants and the values of the calculated dihedral angles are listed in Table 1. The corresponding values of the angles found by X-ray analysis are also indicated for **6b**, which was isolated in the individual state (Fig. 1). Comparison of the experimental vicinal constants and those estimated in this work shows satisfactory concordance between these values. Attention should be paid to the difference in the geometric parameters calculated for **6b** by the method of molecular mechanics and those found by X-ray analysis. This difference is the reason for the nonconformity between the calculated and experimental coupling constants. The experimental data on the whole agree better with the values calculated from the angles determined by the X-ray diffraction method. The difference mentioned seems to be related to the difficulties in the choice of parameters for heteroatoms in the molecular mechanics calculations. Actually, the difference in the $^4J_{H\text{-}2,H\text{-}3a}$ constants is observed in the analysis for the stereoisomers (−0.8 Hz for **6b** and none detected for **14b**) and for the $^3J_{H\text{-}2,H\text{-}3}$ vicinal constants (see Table 1). In our opinion, these peculiarities of the spectra can be used for the establishment of the spatial structure of the cycloadducts of cyclic nitrones with monosubstituted alkenes.

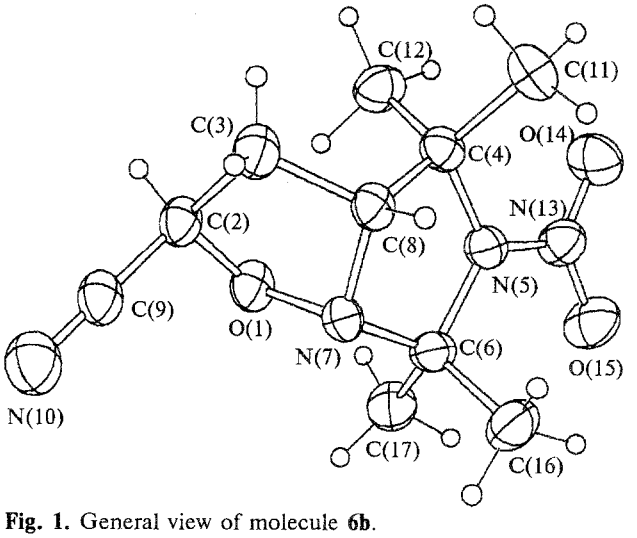


Fig. 1. General view of molecule **6b**.

The interaction of nitrone **2** with acrylonitrile occurs analogously, and in addition to the 5-substituted isomer **5b**, which has been described previously, a second diastereomer **13b** is formed (according to the ^{1}H NMR data, the ratio of **5b** and **13b** is 4 : 1). In addition, the ^{1}H and ^{13}C NMR spectra contain low-intensity signals (< 10 %), which can be assigned to the 4-cyano-isomer **8b**.

A decrease in the regio- and stereoselectivity of cycloaddition is usually observed when the electron-acceptor properties of the substituent in the alkene molecule increase.[2–4] For example, the interaction of **1** with ethyl acrylate and methyl vinyl ketone results in the formation of a mixture of isomers, which are likely 4- and 5-regioisomers. The stereochemistry of compounds **7a,c** was not unambiguously established due to their low content (2—5 %) in the reaction mixture. The ^{13}C NMR spectrum of **10c**, which is the diamagnetic analog of **4c** and is obtained by reduction of this radical, shows that the reaction results in the formation of only one diastereomer. We failed to reduce **4a** to the correspond-

Table 1. Calculated (MM) and experimental dihedral angles (φ) and spin-coupling constants (J) in the ^{1}H NMR spectra of compounds **6b** and **14b**

Dihedral angle	φ/deg			Spin-coupling constant	J/Hz				
	6b		**14b**		**6b**			**14b**	
	X-ray	MM	(MM)		X-ray	MM	Exp.	MM	Exp.
H(2)—C(2)—C(3)—H(3)	3.7	14.6	126.7	$^3J_{H\text{-}2,\ H\text{-}3}$	9.3	8.8	9.1	5.2	6.4
H(2)—C(2)—C(3)—H(3')	119.0	135.1	7.0	$^3J_{H\text{-}2,\ H\text{-}3'}$	4.1	6.6	4.1	9.2	9.5
H(3)—C(3)—C(3a)—H(3a)	97.5	92.8	101.8	$^2J_{H\text{-}3,\ H\text{-}3'}$	2.0	1.8	1.2	2.3	1.4
H(3a)—C(3a)—C(3)—H(3')	21.6	26.7	17.4	$^3J_{H\text{-}3a,\ H\text{-}3'}$	8.5	8.0	7.7	8.9	7.8

ing diamagnetic hydroxy derivative. Therefore, ester **4a** was transformed into amide **4f**, whose reduction resulted in the diamagnetic analog **10f**. According to the ^1H and ^{13}C NMR spectral data, the sample of the latter contains no admixture of diastereomer **11f**, unlike the amide obtained by hydrolysis of a mixture of nitriles **4b** and **12b**, *i.e.*, in this case, the reaction occurs with high stereoselectivity. Judging from the ^1H and ^{13}C NMR spectra, the interaction of nitrones **2** and **3** with ethyl acrylate and methyl vinyl ketone results in the exclusive formation of 5-regioisomers with *trans*-arrangement of H-2 and H-3' protons (**5a,c** and **6a,c**).

Cycloaddition of cyclic aldonitrones **1—3** to alkenes containing rather poor electron-acceptor substituents (styrene and 4-vinylpyridine) occurs regioselectively; in all cases, 5-substituted isoxazolidines **4d,e, 5d,e,** and **6e,** respectively, were isolated in high yields. The structure of diamagnetic cycloadducts **5d,e** and **6e** was established on the basis of ^1H and ^{13}C NMR spectra; the paramagnetic cycloadducts **4d,e** were reduced to the hydroxylamino derivatives **10d,e**, whose ^1H NMR spectra indicate that the substituent is at the 5-position of the isoxazolidine cycle. It should be mentioned that the interaction of aldonitrones **2** and **3** with vinylpyridine results in the formation of only one diastereomer of the 5-regioisomer as evidenced by the presence of one set of signals in the ^{13}C NMR spectra of **5e, 6e,** and **10d,e**.

Thus, 1,3-dipolar cycloaddition of aldonitrones, which are derivatives of 3-imidazoline-3-oxide, to monosubstituted olefins containing weak electron-acceptor substituents occurs with high regio- and stereoselectivity. Enhancement of the electron-acceptor properties of the substituent in the olefin decreases the regioselectivity of the process. It was found that in the case of acrylonitrile, the stereoselectivity of the process is also decreased. The data about the quantitative composition of the products of the cycloaddition allowed us to draw the conclusion that the paramagnetic aldonitrone **1** reacts with alkenes in the least selective fashion, which cannot be explained in terms of the electronic charge effects, because the electron-acceptor influence of the nitroxyl group is similar to that of the *N*-nitro group. One may suppose that in this case the specific spin effect of the nitroxyl group, caused by its paramagnetism, manifests itself (*cf.* Ref. 8).

Table 2. ^1H NMR spectra (δ) of the compounds synthesized (in $CDCl_3$)

Compound	X	R	4,4,6,6-Me	H-2	H-3 H-3'	H-3a
5a	2.03 (3 H)	4.01 (2 H); 1.09 (3 H)	0.87 (3 H); 0.93 (3 H); 1.01 (3 H); 1.05 (3 H)	4.36	2.26—2.36	3.37
5b	2.21 (3 H)	—	0.96 (3 H); 1.06 (3 H); 1.16 (3 H); 1.27 (3 H)	4.73	2.63—2.66	3.66
5c	2.07 (3 H)	2.02 (3 H)	0.83 (3 H); 0.88 (3 H); 0.96 (3 H); 1.16 (3 H)	4.07	2.02 2.29	3.26
5e	2.01 (3 H)	8.28 (2 H); 7.07 (2 H)	0.87 (3 H); 0.89 (3 H); 0.92 (3 H); 0.99 (3 H)	4.85	2.00 2.55	3.35
6a	—	4.14 (2 H); 1.21 (3 H)	1.44 (3 H); 1.45 (3 H); 1.52 (3 H); 1.79 (3 H)	4.14	2.55—2.79	3.75
6b	—	—	1.44 (3 H); 1.49 (3 H); 1.62 (3 H); 1.83 (3 H)	4.76	2.79—2.85	3.91
6c	—	2.17 (3 H)	1.39 (3 H); 1.41 (3 H); 1.48 (3 H); 1.77 (3 H)	4.15	2.45—2.65	3.65
6e	—	8.48 (2 H); 7.22 (2 H)	1.50 (3 H); 1.59 (6H); 1.91 (3 H)	5.05	2.42 2.95	3.82
10c	6.52	2.26 (3 H)	1.22 (6 H); 1.30 (3 H); 1.52 (3 H)	4.25	2.48—2.51	3.60
10d	5.82	7.32 (5 H)	1.26 (3 H); 1.31 (3 H); 1.39 (3 H); 1.48 (3 H)	5.01	2.36 2.65	3.83
10e	6.50	8.49 (2 H); 7.26 (2 H)	1.19 (3 H); 1.24 (3 H); 1.33 (3 H); 1.42 (3 H)	4.98	2.29 2.68	3.71
10f*	7.55	7.21 (1 H); 7.04 (1 H)	1.04 (3 H); 1.09 (3 H); 1.24 (3 H); 1.58 (3 H)	4.33	2.27—2.52	3.53
13b	2.11 (3 H)	—	0.83 (3 H); 0.99 (3 H); 1.01 (3 H); 1.19 (3 H)	4.55	2.46—2.56	3.52
14b	—	—	1.47 (3 H); 1.53 (3 H); 1.55 (3 H); 1.56 (3 H)	4.61	2.63 2.87	3.84

* The spectrum was recorded in DMSO-d_6.

Table 3. ^{13}C NMR spectra (δ) of the compounds synthesized (in $CDCl_3$)

Compound	R	C-6	C-2	C-3a	C-4	C-3	6-Me_2	4-Me_2
5a*	13.61 (q); 60.49 (t); 171.44 (s)	82.79 (s)	76.44 (d)	71.24 (d)	58.79 (s)	34.66 (t)	24.51 (q); 24.29 (q)	24.01 (q); 23.88 (q)
5b*	119.11 (s)	83.07 (s)	70.08 (d)	64.70 (d)	61.02 (s)	36.74 (t)	25.27 (q); 25.23 (q)	23.85 (q)
5c*	25.12 (q); 211.33 (s)	82.39 (s)	81.20 (d)	70.67 (d)	59.84 (s)	32.89 (t)	27.49 (q); 25.00 (q)	23.63 (q); 23.45 (q)
5e*	119.49 (d); 149.14 (d); 152.09 (s)	83.00 (s)	77.54 (d)	71.47 (d)	59.39 (s)	39.13 (t)	24.43 (q); 24.19 (q)	23.70 (q); 23.45 (q)
6a	13.84 (q); 61.13 (t); 171.21 (s)	86.98 (s)	73.23 (d)	69.36 (d)	65.30 (s)	33.15 (t)	25.47 (q); 24.14 (q)	21.73 (q); 20.83 (q)
6b	118.38	86.92	68.78	65.45	63.31	35.90	25.77; 24.35	22.29
6c	24.02 (q); 210.65 (s)	86.46 (s)	79.36 (d)	69.10 (d)	65.52 (s)	31.89 (t)	25.41 (q); 25.06 (q)	22.21 (q); 21.73 (q)
6e	149.90; 119.98; 152.50	87.27	75.43	70.42	65.53	38.21	25.76; 24.56	22.48; 22.04
10c	209.43 (s); 23.13 (q)	82.85 (s)	78.96 (d)	67.36 (d)	61.42	31.17 (t)	23.93 (q); 23.33 (q)	21.73 (q); 21.12 (q)
10f	174.27	84.04	77.25	69.13	61.31	35.22	25.22; 24.97	24.17; 23.85
11f	174.15	84.01	76.71	69.12	61.37	35.15	24.64; 24.62	23.81; 23.76
13b*	118.97 (s)	83.27 (s)	72.27 (d)	64.88 (d)	60.26 (s)	36.23 (t)	24.77 (q); 24.67 (q)	24.58 (q); 24.40 (q)
14b	118.63 (s)	86.73 (s)	69.38 (d)	62.60 (d)	65.38 (s)	34.79 (t)	25.62 (q); 23.79 (q)	23.24 (q); 22.11 (q)

* Signal of N—Me, δ: 27.29 for **5a**; 27.77 for **5b**; 27.49 for **5c**; 27.36 for **5e**; 27.46 for **13b**.

Experimental

IR spectra were recorded on UR-20 and Specord M-80 instruments. UV spectra were registered on a Specord UV-VIS spectrometer, ^1H and ^{13}C NMR spectra were obtained on a Bruker AC-200 instrument. ^1H and ^{13}C NMR spectral data are listed in Tables 2 and 3, respectively. The yields, melting points, molecular formulas, and IR and UV spectra of the compounds synthesized are given in Table 4.

Aldonitrone **1** was prepared as described previously,[1] aldonitrones **2** and **3** were obtained by the known procedure.[9] The course of the reactions was monitored by TLC (Silufol; solvent system hexane—ethyl acetate (10:1)).

X-ray diffraction study was performed on a Syntex P21 diffractometer (Cu-Kα-irradiation, graphite monochromator). The crystals of **6b** are monoclinic: a = 9.381(1) Å, b = 7.409(1) Å, c = 17.133(2) Å, β = 92.04(1)°, V = 1190.1(3) Å^3, spatial group $P2_1/c$, Z = 4, $C_9H_{16}N_4O_3$, M = 228.25, d_{calc} = 1.274 g cm^{-3}. Intensities of 1769 reflections with $2\theta < 120°$ were measured by the method of w-scanning, 1158 reflections with $I > 3\sigma$ were used in the calculations. The structure was solved by the direct method and corrected in the anisotropic approximation up to R = 0.062, R_w = 0.062, S = 2.16. The coordinates of the nonhydrogen atoms are listed in Table 5, the structure of the molecule is shown in Fig. 1. The conformations of both heterocycles are close to the form of the envelope: N-7 atom deviates from the plane of the O-1, C-2, C-3, and C-8 atoms by 0.700 Å, and C-8 deviates from the plane of the C-4, C-5, C-6, and N-7 atoms by 0.454 Å. The nitroamine fragment C—N(NO_2)—C is planar, the mean-square deviation equals 0.025 Å.

Interaction of 3-imidazoline-3-oxides 1—3 with dipolarophiles (general procedure). A solution of 0.05 mol of aldonitrone **1—3** and 0.075 mol of a dipolarophile in 10 mL of chloroform was kept for 10—18 h at ~20 °C and then evaporated. The residue was chromatographed on a column with SiO_2 (hexane—ethylacetate, 3 : 1, as the eluent), and the cycloadducts isolated were purified by recrystallization.

Reduction of paramagnetic cycloadducts 4d—f (general procedure). A solution of 0.01 mol of isoxazolidine and 0.1 mol of 80 % hydrazine hydrate in 2 mL of methanol was kept for 10—18 h at 20 °C and evaporated. The residue was washed with water, the precipitate of the hydroxylamino derivative **10d—f** was filtered off and dried.

Isoxazolidines **10d—f** are smoothly oxidized by MnO_2 to form the original radicals **4d—f**.

Table 4. Characteristics of the compounds synthesized

Compound	Yield (%)	M.p. /°C	IR (KBr), ν/cm^{-1}	UV (EtOH), λ_{max}/nm (log ε)	Found / Calculated (%) C	H	N	Molecular formula
4f	83	194—195	1690 (C=O); 3480 (N—H)	—	52.7 / 52.6	7.9 / 7.9	18.3 / 18.4	$C_{10}H_{18}N_3O_3$
5a	95	Oil	1725 (C=O)	—	60.8 / 60.9	9.5 / 9.5	10.7 / 10.9	$C_{13}H_{24}N_2O_3$
5c	97	Oil	1720 (C=O)	—	63.5 / 63.7	9.8 / 9.8	12.3 / 12.4	$C_{12}H_{22}N_2O_2$
5e	95	43—45		234 (3.00)	68.9 / 68.9	8.8 / 8.9	16.1 / 16.1	$C_{15}H_{23}N_3O$
6a	95	Oil	1315, 1510 (NO_2); 1730 (C=O)	243 (3.73)	50.1 / 50.2	7.3 / 7.4	14.6 / 14.6	$C_{12}H_{21}N_3O_5$
6b	63	141—143	1320, 1505 (NO_2); 2230 (CN)	241 (4.22)	50.0 / 50.1	6.7 / 6.8	23.3 / 23.6	$C_{10}H_{16}N_4O_3$
6c	97	84—85	1310, 1520 (NO_2); 1730 (C=O)	244 (3.72)	51.3 / 51.3	7.5 / 7.5	16.2 / 16.3	$C_{11}H_{19}N_3O_4$
6e	80	138—139	1320, 1500 (NO_2);	243 (4.03)	57.5 / 57.4	6.9 / 6.8	19.2 / 19.3	$C_{14}H_{20}N_4O_3$
7a	3	42—43	1740 (C=O)	235 (3.42)	56.2 / 56.0	8.4 / 8.2	10.8 / 10.7	$C_{12}H_{21}N_2O_4$
7b	5	56—57	2240 (CN)	237 (3.50)	57.3 / 57.1	7.7 / 7.6	19.1 / 19.0	$C_{10}H_{16}N_3O_2$
7c	3	Oil	1710 (C=O)	230 (3.41)	56.2 / 56.0	8.5 / 8.6	11.2 / 11.1	$C_{11}H_{19}N_2O_3$

Table 5. Coordinates ($\times10^4$) and thermal factors ($\times10^3/Å^2$) of nonhydrogen atoms of **6b**

Atom	*x*	*y*	*z*	U_{eq}	Atom	*x*	*y*	*z*	U_{eq}
O-1	3470 (2)	260 (5)	5492 (1)	59	N-10	2295 (4)	−2607 (8)	4088 (2)	82
C-2	3093 (4)	506 (7)	4684 (2)	55	C-11	1200 (5)	5269 (9)	5984 (3)	68
C-3	1889 (5)	1909 (9)	4647 (2)	63	C-12	3694 (5)	4367 (10)	5675 (3)	72
C-4	2259 (4)	3704 (7)	5967 (2)	52	N-13	2803 (4)	3667 (7)	7390 (2)	67
N-5	2483 (3)	2781 (5)	6730 (2)	55	O-14	2981 (4)	5308 (6)	7362 (2)	87
C-6	2319 (4)	766 (7)	6702 (2)	53	O-15	2890 (4)	2794 (6)	7996 (2)	91
N-7	2067 (3)	419 (5)	5863 (2)	49	C-16	951 (6)	229 (11)	7093 (3)	76
C-8	1549 (4)	2113 (6)	5514 (12)	49	C-17	3614 (6)	−258 (10)	7028 (3)	73
C-9	2637 (5)	−1231 (9)	4346 (2)	60					

2-Acetyl-5-hydroxy-4,4,6,6-tetramethylperhydroimidazo[1,5-*b*]isoxazole (10c). A suspension of 0.227 g (0.01 mol) of compound **4c**, 1 g of zinc dust, and 50 mg of NH_4Cl in 10 mL of methanol was stirred for 30 min at 20 °C. The excess zinc was filtered off, washed on the filter with methanol, and the solution was evaporated. The residue was diluted with 3 mL of water and extracted with $CHCl_3$ (3×20 mL), the extract was dried with $MgSO_4$, and the solution was evaporated. The residue was recrystallized from hexane to give **10c**, which quantitatively yields the original radical **4c** upon oxidation by MnO_2.

2-Carbamoyl-4,4,6,6-tetramethylperhydroimidazo[1,5-*b*]isoxazole-5-oxyl (4f). ***A.*** A solution of 0.257 g (0.01 mol) of ester **4a** in a mixture of 5 mL of methanol and 5 mL of 25 % aqueous ammonia was kept for 10 h at 20 °C, the precipitate of the amide **4f** formed was filtered off, washed with water, and dried.

B. 1 mL of 6 % H_2O_2 and then a 10 % solution of NaOH were added to a solution of 0.21 g (0.01 mol) of nitrile **4b** in 5 mL of methanol to pH 10. After 30 min, the solution was concentrated to half its volume and extracted with $CHCl_3$. The extract was dried with $MgSO_4$, the solution was evaporated, and amides **4f** and **12f** were obtained.

References

1. V. V. Martin, L. B. Volodarsky, M. A. Voinov, T. A. Berezina, and T. F. Lelyukh, *Izv. Akad. Nauk SSSR, Ser. Khim.*, 1988, 1875 [*Bull. Acad. Sci. USSR, Div. Chem. Sci.*, 1988, **37**, 1677 (Engl. Transl.)].
2. Sk. A. Ali, J. H. Khan, and J. M. Wazeer, *Tetrahedron*, 1988, **44**, 5911.

3. M. Burdisso, R. Gandolfi, and P. Grunanger, *Tetrahedron,* 1989, **45**, 5579.
4. Sk. A. Ali, P. A. Senaratne, C. R. Hlig, H. Meckler, and J. J. Tufariello, *Tetrahedron Lett.,* 1979, 4167.
5. J. J. Tufariello and J. R. Tette, *J. Org. Chem.,* 1978, **40**, 3866.
6. J. J. Tufariello and S. A. Ali, *Tetrahedron Lett.,* 1978, 4647.
7. Y. Takeuchi and R. Furusaki, in *Advances in Heterocyclic Chemistry,* Ed. A. R. Katritsky, Academic Press, New York, 1977, **21**, 208.
8. L. B. Volodarsky, I. A. Grigor'ev, S. A. Dikanov, V. A. Reznikov, and G. I. Shchukin, in *Imidazolinovye nitroksil'nye radikaly* [*Imidazoline Nitroxyl Radicals*], Ed. Yu. N. Molin, Nauka, Novosibirsk, 1988 (in Russian).
9. I. A. Grigor'ev, G. I. Shchukin, V. V. Martin, and V. I. Mamatyuk, *Khim. Geterotsikl. Soedin.,* 1985, 252 [*Chem. Heterocycl. Compd.,* 1985 (Engl. Transl.)].

Received June 30, 1993