GEMINAL SYSTEMS.

48.* DIALKOXYAMINYL RADICALS AND TETRAALKOXYHYDRAZINES

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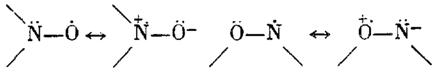
By oxidation of N,N-dialkoxyamines RONHOR' (I), we obtained dialkoxyaminyl radicals (II) which are stable in solution and inert to O_2, whose structure has been confirmed by an EPR study of photolysis of solutions of (I) in the presence of 2,6-di-tert-butyl-p-benzoquinone. The radicals of (II) exist in solution in equilibrium with their dimers: tetraalkoxyhydrazines (V), whose concentration increases when the solutions are cooled. In three cases ($R = R' = Me$, $R = R' = t\text{-}BuCH_2$, $R = Me$, $R' = i\text{-}Bu$), hydrazines (V) are isolated in pure form. From the temperature dependence of the concentration of radicals (II), we determine the thermodynamic parameters for their equilibrium dimerization. Using the PMR method, we establish the pyramidal configuration of the N atoms in hydrazines (V).

The stability of radicals is determined mainly by the degree of delocalization of the unpaired electron over the bonds of the substituents and steric screening of the radical center. Thus sterically hindered nitroxide radicals are more stable than isomeric alkoxyaminyl radicals, since the dative and accordingly the delocalizing ability of the N atom is greater than for O [2]



The stability of the aminyl radicals with two electron-donor substituents at the N atom ($Hal_2\dot{N}$ [3], $(RS)_2\dot{N}$ [4], $[(Me_3Si)_2N]_2\dot{N}$ [5] and $(NaO)_2\dot{N}$ [6]) varies over broad limits. While the first radicals are registered only in a matrix, the latter are stable up to 100-130°C in the solid phase in an inert atmosphere.

In this work, we have studied new representatives of this type of radical: the dialkoxyaminyl radicals $(RO)_2\dot{N}$ (II).†

Radicals (II) were registered from the EPR spectra (Table 1, Fig. 1a) for solutions of N,N-dialkoxyamines (I) in toluene or a 1:1 toluene–pentane mixture after they were stored under ordinary conditions. The concentration of (II) increases as a result of photooxidation upon UV irradiation of pure solutions of (I) or with addition of 2,6-di-tert-butyl-p-benzoquinone (2,6-θ). Furthermore, radicals (II) are formed upon treatment of solutions (I) with PbO_2 and titration with nitroxide and aroxyl radicals

$$\underset{(Ia-f)}{RONHOR'} \xrightarrow[-H^{\cdot}]{} \underset{(IIa-f)}{RO\dot{N}OR'}$$

$$R = R' = Me\ (a);\ R = Me,\ R' = i\text{-}Bu\ (b);\ R = Me,\ R' = PhCH_2\ (c);$$
$$R + R' = (CH_2)_2\ (d);\ R + R' = MeCH(CH_2)_2\ (e);\ R = R' = t\text{-}BuCH_2\ (f).$$

The starting (Ia-e) are synthesized as in [8]. We analogously obtained (If) according to the scheme

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[*] For previous communication, see [1].

[†] For the preliminary report, see [7].

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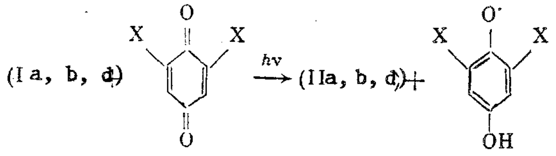
$$\mathrm{XN(Cl)OMe}\xrightarrow{\mathrm{ROH}}\underset{(\mathrm{III})}{\mathrm{XN(OR)OMe}}\xrightarrow{\mathrm{ROH/H^+}}\underset{(\mathrm{IV})}{\mathrm{XN(OR)_2}}\xleftarrow{\mathrm{ROH/H^+}}\mathrm{XN(OMe)_2}$$

$$(\mathrm{IV})\xrightarrow{\mathrm{KOH/MeOH}}(\mathrm{If})$$

$X = Me_2NCO$, $R = t\text{-}BuCH_2$.

For radicals (II), we observe the expected hyperfine splitting of the EPR spectra (Fig. 1a, Table 1). The aminyl structure of (II) is confirmed by the agreement between the magnitudes of the hyperfine coupling constants a_N and the g-factors with those for other aminyl radicals [5, 9], including alkoxyaminyl [9, 10], and the significant difference in the a_N values from those of the isomeric nitroxide radicals RO(R)NO ($a_N \approx 27$-28 Oe) [11]. For additional proof of the structure of radicals (II), we have studied the photolysis of solutions of N,N-dialkoxyamines (Ia, b, d) in a 1:1 toluene–pentane in the presence of 2,6-θ over a broad range of temperatures. For +20°C to −100°C, along with the aminyl radicals the 2,6-di-tert-butyl-4-hydroxyphenoxyl radical is registered ($a_H^m = 0.7$, $a_H^{OH} = 1.5$ Oe), then



$X = t\text{-}Bu$.

as in glassy toluene solutions at 77 K, radical pairs are formed which give an intense EPR signal (Fig. 2b) with g-factor ∿ 2.0 and axially symmetric zero-field splitting tensors $D_{\parallel} = 306$, $D_{\perp} = 154$ Oe. At "half" field, the forbidden signal $\Delta m_S = 2$ was registered: an anisotropic triplet as a result of hyperfine coupling with the ^{14}N nucleus (Fig. 2a). From analysis of this signal, we determined the anisotropic part of the hyperfine coupling tensor for radicals (IIa, b, d). The symmetric form of the spectrum of the forbidden signal is evidence for the absence of anisotropy in the g-factor and significant anisotropy in the hyperfine coupling, which is characteristic for aminyl but not for nitroxide radicals [12]. From the distance between the extremal components of the signal (Fig. 2a) and taking into account the fact that for radical pairs the hyperfine coupling constants are half those for the individual radicals, we found the $A_{zz} = 42.57$ component of the hyperfine coupling tensor, which is connected with the isotropic a_N and anisotropic b_N parts of the hyperfine coupling constants by the relationship: $A_{zz} = a_N + 2b_N$ [12], from which $b_N = 14.0$ Oe. The ratio $a_N/b_N = 1.04$ for the investigated radicals is characteristic for aminyl radicals [12], which confirms the suggested structure for radicals (II).

For comparison, using a similar technique we studied the EPR spectrum of the N-monoalkoxyaminyl radical $MeO_2CCH_2CMe_2\dot{N}OMe$, generated from the corresponding N-methoxyamine [13] under the action of tert-butyl peroxide in pentane. We determined the following parameters for it: $a_N = 13.8$, $b_N = 12.6$, $a_H = 2.55$ Oe, $g = 2.0046$, $a_N/b_N = 1.1$.

Radicals (II), except for (IIc), are stable in solution: the EPR spectra do not change, at least for half a year at 20°C. The concentration of (II) increases upon heating their solutions to 120°C, and decreases upon cooling down to complete disappearance at −80°C to −100°C, which is explained by the reversible dimerization of (II) to tetraalkoxyhydrazines (V)

$$(\mathrm{I})\ \xrightarrow[-\mathrm{H}^{\cdot}]{}\ 2\,(\mathrm{II})\ \rightleftharpoons\ (\mathrm{RO})_2\mathrm{NN}(\mathrm{OR})_2\quad(\mathrm{V})$$

TABLE 1. Parameters of the EPR Spectra (toluene, 20°C) for Radicals (II)

Radical	Shape of spectrum	a_N, Oe	a_H^{HCO}, Oe	g-factor
(IIa)	three septets	15,0	1,45	2,0046
(IIb)	three sextets	15,0	1,25	2,0046
(IIc)	three sextets	14,8	1,25	2,0044
(IId)	three quintets	16,0	2,50	2,0046
(IIe)	three triplets	14,8	3,50 *	2,0059
(IIf)	three quintets	15,0	1,25	2,0044

*Hyperfine splitting is observed only with axial H.

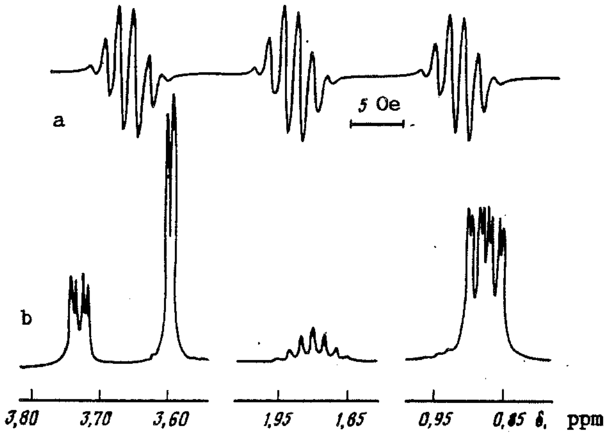


Fig. 1. a) EPR spectrum (toluene, 20°C) of radical (IIb); b) PMR spectrum (400 MHz, toluene-d_8, −10°C) of hydrazine (Vb).

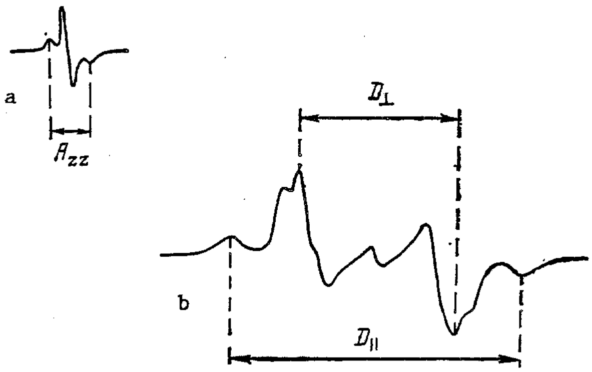


Fig. 2. EPR spectra for glassy (77 K) toluene solutions of radical pairs of the 2,6-di-tert-butyl-4-hydroxyphenoxyl radical and (IIa, b, d): a) forbidden signal at half field, simulating the anisotropic EPR spectrum of the aminyl radical; b) primary signal for g ≈ 2, the zero-field splitting tensor and the hyperfine coupling tensor with the N nucleus (A_{zz}) are indicated.

In fact, by oxidation of solutions of N,N-dialkoxyamines (Ia, b, f) under the action of PbO_2, we obtained previously unknown pure hydrazines (Va, b, f) (no attempts were made to isolate other hydrazines (V)), which are stable when stored under ordinary conditions. The structure of (Va, b, f) was confirmed by elemental analysis, and PMR spectra; the structure of (Va) was confirmed by x-ray diffraction analysis.*

For solutions of hydrazines (Va, b, f) in toluene-d_8 at 20°C, we observe well-resolved EPR spectra for radicals (IIa, b, f), formed upon dissociation of (Va, b, f) (Fig. 1a), and accordingly a poorly resolved PMR spectrum due to paramagnetic broadening. And the EPR spectra for radicals generated both from pure (Va, b, f) and by the above-listed methods from (Ia, b, f) are identical, which clearly confirms the dialkoxyaminyl structure of radicals (II). Upon cooling solutions of hydrazines (Va, b, f), the intensity of the EPR signals decreases and well-resolved PMR spectra are registered. In this case, in the PMR spectrum of (Vb) (Fig. 1b) there is a set of signals from the d, ℓ-, and meso forms (1:1), and for each we observe geminal anisochronicity of diastereotopic Me groups, which is evidence for the pyramidal configuration of N atoms in tetraalkoxyhydrazines (V).

The thermodynamic parameters for the equilibrium (V) $\rightleftarrows$ 2 (II) were found by the EPR method from the temperature dependence of the concentration of radicals (II) in solutions of (V) (Table 2).

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*X-ray diffraction analysis at −110°C was carried out by M. Yu. Antipin and Yu. T. Struchkov.

TABLE 2. Thermodynamic Parameters of the Equilibrium Dimer ⇄ Radicals*

Radical	ΔH°, kcal/mole	ΔS°, eu	K_{20°, moles/liter
(IIa)	9.2	6.8	$4.7 \cdot 10^{-6}$
(IIb)	11.0	7.1	$1.6 \cdot 10^{-7}$
(IIf)	12.2	20.6	$4.0 \cdot 10^{-5}$
$(NaO)_2\dot{N}$ [6]	0.6	–	
$(CF_3S)_2\dot{N}$ [4]	7.6	0.7	
t-BuO(Ph)$\dot{N}$ [10]	12.5	21.0	

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*ΔH° and ΔS° are calculated from the equation: $\ln K = -(\Delta H^\circ/R)(1/T) + \Delta S^\circ/R$, where the equilibrium constant is $K = [(II)]^2/[(V)]$. Error in the determinations $\Delta H^\circ \pm 0.5$ kcal/mole, $\Delta S^\circ \pm 0.5$ entropy units, ±20% for the equilibrium constant.

The radicals (II) are not oxidized by oxygen in the air. Upon boiling a solution of (Ia) with azodiisobutyronitrile (ADI), we obtained N,N-dialkoxyamine (VI), the product of recombination of (IIa) and the 2-cyanoisopropyl radical. This example is evidence for the potential of radical reactions in the chemistry of N,N-dialkoxyamines (I) [8]

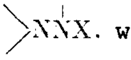
$$\text{(Ia)} \xrightarrow[C_6H_6]{\text{ADI}} (MeO)_2NC(Me)_2CN$$

(VI)

RESULTS AND DISCUSSION

We first must mention the high stability of dialkoxyaminyl radicals (II) compared with N-alkoxy-N-alkylaminyl, which in the absence of steric screening of the radical center are observed only from the low-temperature EPR spectrum [10]. One reason for this is the more effective delocalization of the unpaired electron in (II). Unfortunately, we cannot quantitatively estimate this on the basis of available experimental data, since it was shown previously [9, 14] that the value of a_N for aminyl radicals is virtually independent on the nature of the substituents at the radical center, and cannot be used for determination of the 2p spin density at the N atom. For comparison, we give the values of a_N (in Oe) for various aminyl radicals: $Me_2\dot{N}$ 14.7 [9], $MeO\dot{N}Me$ 14.3 [9, 14], $F_2\dot{N}$ 16 [3], $(CF_3S)_2\dot{N}$ 13.2 [4], $[(Me_3Si)_2N]_2\dot{N}$ 14.8 [5]. Indirect confirmation of the substantial delocalization of the unpaired spin in (II) comes from their inertness to oxygen in the air. In this regard, they are similar to dithio analogs $(RS)_2\dot{N}$ [4] and differ from dialkyl- and N-alkoxy-N-alkylaminyls [2, 5, 9].

The stability of (II) is also due to the fact that disproportionation (realized in the case of alkoxyaminyl radicals containing the α-CH [10] group) is impossible for them. Furthermore, for the example of N-tert-butoxy-N-alkylaminyl radicals, it has been shown [2] that β-elimination with subsequent isomerization to nitroxides is not characteristic for alkoxyaminyls.

Another reason for the stability of radicals (II) is their dimerization to hydrazines (V), which are stable to cleavage at the N–O bond. In fact, the instability of N-alkoxy-N-alkylaminyls is mainly connected with the instability of their dimers, since the primary oxidation products of the corresponding amines are formed as a result of cleavage of the N–O bond of the intermediate 1,2-dialkoxyhydrazines [10]. The high stability of tetraalkoxyhydrazines (V) compared with 1,2-dialkoxy-1,2-dialkylhydrazines is due to weakening of the n_N–σ^*_{NO}-orbital interaction in them, which is responsible for the kinetic destabilization of the N–O bond due to the decrease in the electron-donor ability of the N atoms and depolarization of the N–O bond under the influence of the second N-alkoxy group (the stability of geminal systems  $>N\overset{|}{N}X$, where X is a σ-electron-acceptor substituent, is considered in more detail in [15].

In hydrazines (V), to a large degree the N–N bond is weakened as a result of n_O –σ^*_{NN} one-electron transfer. Thus, the regular decrease in the strength of the N–N bond with an increase in the electron-donor ability of the N-substituents and the number of such substituents follows from analysis of the thermodynamic parameters for the equilibrium dimer ⇄ 2 radicals for different hydrazines (Table 2).

EXPERIMENTAL

The EPR spectra were recorded on the Varian E-12 A instrument. The concentration of radicals (II), generated upon dissociation of (V), is determined using a thanol standard in C_6H_6 (C 10^{-3} moles/liter). All the quantitative estimates for the radical concentrations were made with recordings at low modulation amplitude (<0.5 Oe). The relative error in the estimate of the radical concentrations was ∿10%. The registration conditions for the EPR spectra were made equivalent by using an "auxiliary" standard in the double resonant cavity of the EPR spectrometer. The integrated signal intensity was determined as the product of the square of the line width times its intensity for an individual component of the spectrum of the studied radicals, taking into account their multiplicity. The absolute error for the given integration error is ∿50%.

The NMR spectra were measured on the Bruker WP-80SY and Bruker WM-400 spectrometers. The chemical shifts are given in ppm and the spin–spin coupling constants are given in Hz.

(Ia), yield 62%, bp 83.5°C; (Ib), yield 72%, bp 65°C (40 mm): (Ic), yield 67%, purified chromatographically; (Id), yield 34%, bp 58°C (4 mm); (Ie), yield 44%, bp 117°C (1 mm); 1-chloro-1-methoxy-3,3-dimethylurea (used without purification) and 1,1-dimethoxy-3,3-dimethylurea, yield 85%, bp 61°C (1 mm) were obtained as in [8].

1-Neopentyloxy-1-methoxy-3,3-dimethylurea (III). To 0.31 g (2 mmoles) 1-chloro-1-methoxy-3,3-dimethylurea at –78°C was added a solution of 0.24 g (2 mmoles) γ-collidine in 2 ml neopentyl alcohol. The mixture was held for 3 h at –8°C and for 1 day at 20°C, and then diluted with ether (20 ml). The residue was separated; the solvent was removed from the filtrate under vacuum and the residue was distilled. Obtained: 0.29 g (71%) (III), bp 82-83°C (1 mm). PMR spectrum (80 MHz, $CDCl_3$, δ relative to HMDS): 0.98 (Me_3C), 3.0 (Me_2N), 3.59 (CH_2), 3.73 (MeO). Found: C 52.92; H 9.89; N 13.39%. $C_9H_{20}N_2O_3$. Calculated: C 52.92; H 9.87; N 13.71%.

1,1-Dineopentyloxy-3,3-dimethylurea (IV). a) A solution of 7.55 g (51 mmoles) 1,1-dimethoxy-3,3-dimethylurea and 0.4 g (2.3 mmoles) TsOH in 26.4 g neopentyl alcohol was held at 20°C for 2 weeks with daily periodic removal under vacuum of the MeOH formed in the reaction. The mixture was neutralized by addition of 0.4 ml Et_3N; the excess alcohol was removed under vacuum and the residue was chromatographed on a column (Al_2O_3, neutralized by the Brockmann method; eluting agent – ether). Two fractions were obtained containing (III) and (IV) which were distilled under vacuum. Obtained: 1.47 g (14%) (III) (see above) and 4.54 g (34%) (IV), bp 96°C (1 mm). For (IV), the PMR spectrum is the following (80 MHz, $CDCl_3$, δ relative to TMS): 0.98 (Me_3C), 3.0 (Me_2N), 3.59 (CH_2). Found: C 59.97; H 11.00; N 10.36%. $C_{13}H_{28}N_2O_3$. Calculated: C 59.97; H 10.84; N 10.76%.

b) Under the conditions of the preceding experiment, from 1.47 g (7.2 mmoles) of (III), 0.1 g TsOH, and 4.4 g neopentyl alcohol, after holding for 3 days 0.5 g (27%) (IV) was obtained after distillation under vacuum (see above).

N,N-Dineopentyloxyamine (If). A solution of 4.54 g (17.4 mmoles) (IV) and 1.95 g (34.8 mmoles) KOH in 15 ml MeOH was held overnight at 20°C. The mixture was saturated with CO_2 and diluted with ether. The residue was separated and the solvent was removed from the filtrate under vacuum (20 mm). The residue was extracted with ether and the extract was evaporated under vacuum. The residue was distilled. Obtained: 2.15 g (65%) (If), bp 63°C (4 mm). PMR spectrum (80 MHz, $CDCl_3$, δ relative to HMDS): 0.93 (Me_3C), 3.48, 3.65 (CH_2, J_{AB} = 8.7, 7.82 (NH). Found: C 63.20; H 11.90; N 7.88%. $C_{10}H_{23}NO_2$. Calculated: C 63.45; H 12.25; N 7.39%.

Tetraalkoxyhydrazines (Va, b, f). A solution of (Ia, b, f) in absolute ether was mixed at 20°C (for (Ia) at ∿5°C) with a 10-fold molar excess of carefully dried PbO_2 for 3 h. The residue was separated and the solvent was removed from the filtrate under vacuum (for (Ia), ∿150 mm; for (Ib, f), ∿60 mm). In the case of (Ia), the residue was frozen out from solution (ether–pentane), yield (Va) 42%. Upon distillation under vacuum, it partially (∿30%) decomposes, bp 77-78°C (100 mm). The product also completely decomposes when chromatographed on a column (Al_2O_3, neutralized by the Brockmann method, eluting agent – ether). PMR spectrum (80 MHz, toluene-d_8, δ relative to TMS, 0°C): 3.55 (MeO). In the case of (Ib), the residue was distilled under vacuum, yield of (Vb) 43%, bp 76°C (1 mm). PMR spectrum (400 MHz, toluene-d_8, δ relative to TMS, –10°C): 0.86 (Me_2C, $\Delta\nu$ = 2.3 Hz, ^3J = 6.6), 0.89 (Me_2C, $\Delta\nu$ 1.8 Hz, ^3J = 6.6), 1.90 (CH), 3.73, 3.74 (CH_2, J_{AB} < 0.3, ^3J = 6.7), 3.60, 3.61 (MeO). Found: C 50.70; H 10.29; N 11.96%. $C_{10}H_{24}N_2O_4$. Calculated: C 50.83; H 10.24; N 11.85%. In the case of (If), the residue was crystallized from pentane (0°C), yield of (Vf) 15.9%, mp 104-105°C. PMR spectrum (80 MHz, toluene-d_8, δ relative to HMDS, 0°C): 0.91 (Me_3C), 3.64, 3.80 (CH_2, J_{AB} = 8.2). Found: C 63.79; H 12.01; N 7.28%. $C_{20}H_{44}N_2O_4$. Calculated: C 63.79; H 11.79; N 7.44%.

2-(N,N-Dimethoxyamino)-2-cyanopropane (VI). A solution of 0.5 g (6.5 mmoles) of (Ia) and 2.13 g (13 mmoles) azodiisobutyronitrile in 15 ml abs. C_6H_6 was boiled for 12 h and evaporated under vacuum (15 mm). The residue was extracted with pentane. The extract was evaporated under vacuum and the residue was dissolved in a minimum amount of pentane. The solution obtained was cooled down to −8°C and the precipitated oil was distilled. Obtained: 0.39 g (42%) (VI), bp 35°C (1 mm). PMR spectrum (400 MHz, C_6D_6, δ relative to TMS): 1.15 (Me_2C), 3.78 (MeO). ^{13}C NMR spectrum (80 MHz, acetone-d_6): 23.10 (Me, ^1J = 130.9, ^3J = 4.4), 62.16 (MeO, ^1J = 143.6), 62.72 (C–Me, ^2J = 4.4), 119.7 (CN, ^3J = 5.0). Found: C 49.71; H 8.45; N 19.38%. $C_6H_{12}N_2O_2$. Calculated: C 49.98; H 8.39; N 19.43%.

LITERATURE CITED

1. G. V. Shustov, S. V. Varlamov, A. Yu. Shibaev, et al., Izv. Akad. Nauk SSSR, Ser. Khim., No. 8, 1816 (1989).
2. H. Woynar and K. U. Ingold, J. Am. Chem. Soc., 102, 3813 (1980).
3. H. G. Aurich and W. Weiss, Topics in Current Chem., 59, 65 (1975); C. B. Colburn, R. Ettinger, and F. A. Johnson, Inorg. Chem., 3, 455 (1964); M. S. Wei, J. H. Current, and J. Gendell, J. Chem. Phys., 57, 2431 (1972).
4. Y. Miura, N. Makita, and M. Kinoshita, Tetrahedron Lett., 127 (1975); K. Schlosser and S. Steenken, J. Am. Chem. Soc., 105, 1504 (1983).
5. A. R. Forrester, in: International Review of Science. Org. Chem. Series II, Free Radical Reactions, W. A. Waters (ed.), Butterworths, London–Boston (1975), Vol. 10, pp. 87-131.
6. R. W. Asmussen, Acta Chem. Scand., 12, 578 (1958); J.-C. Chardon and J.-G. Theobald, C. R. Ser. C., 263, 461 (1966).
7. A. I. Prokof'ev, V. F. Rudchenko, S. M. Ignatov, and R. G. Kostyanovskii, Izv. Akad. Nauk SSSR, Ser. Khim., 942 (1988).
8. V. F. Rudchenko, V. I. Shevchenko, and R. G. Kostyanovskii, Izv. Akad. Nauk SSSR, Ser. Khim., 598 (1986); V. F. Rudchenko, S. M. Ignatov, I. I. Chervin, et al., Izv. Akad. Nauk SSSR, Ser. Khim., 1153 (1986); V. F. Rudchenko, V. I. Shevchenko, and R. G. Kostyanovskii, Izv. Akad. Nauk SSSR, Ser. Khim., 1556 (1987); I. I. Chervin, V. S. Nosova, V. F. Rudchenko, et al., Khim. Geterotsikl. Soedin., 396 (1988).
9. W. C. Danen and F. A. Neugebauer, Angew. Chem. Int. Ed., 14, 783 (1975).
10. R. A. Kaba and K. U. Ingold, J. Am. Chem. Soc., 98, 7375 (1976).
11. E. G. Janzen, C. C. Lai, and R. V. Shetty, Tetrahedron Lett., 1201 (1980).
12. A. L. Buchachenko and A. I. Vasserman, Stable Radicals [in Russian], Khimiya, Moscow (1973).
13. R. G. Kostyanovsky, V. F. Rudchenko, V. G. Shtamburg, et al., Tetrahedron, 37, 4245 (1981).
14. W. C. Danen, C. T. West, and T. T. Kensler, J. Am. Chem. Soc., 95, 5716 (1973).
15. G. V. Shustov, N. B. Tavakalyan, and R. G. Kostyanovsky, Tetrahedron, 41, 575 (1985); V. G. Shtamburg, V. F. Rudchenko, Sh. S. Nasibov, et al., Izv. Akad. Nauk SSSR, Ser. Khim., 2327 (1981).