Russian Chemical Bulletin, International Edition, Vol. 55, No. 3, pp. 554—558, March, 2006

Synthesis of 4-aroyl-1,2,4-triazolidin-3-ones *via* ring extension in reactions of 1,2-di- and 1,2,3,3-tetraalkyldiaziridines with aroyl isocyanates

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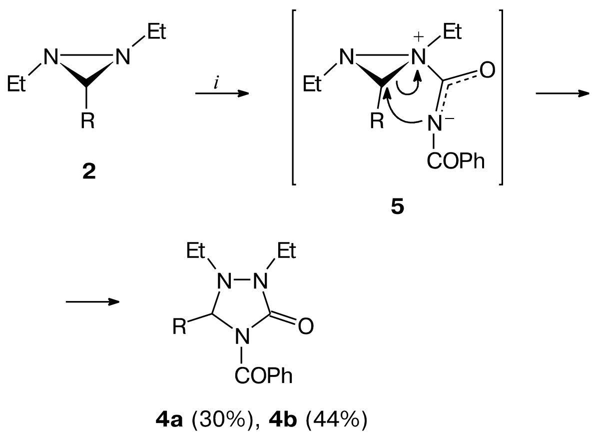
Reactions of 1,2-di- and 1,2,3,3-tetraalkyldiaziridines with aroyl isocyanates involve opening of the diaziridine ring through cleavage of the C—N bond followed by cyclization of the zwitterionic intermediate into 4-aroyl-1,2,4-triazolidin-3-one derivatives.

Key words: 1,2-di-, 1,2,3-tri-, and 1,2,3,3-tetraalkyldiaziridines; aroyl isocyanates, ring extension, 4-aroyl-1,2,4-triazolidin-3-ones.

It is known[1—3] that diaziridines, like other strained three-membered rings, tend toward ring extension reactions with electrophilic reagents. Depending on the type of the substituent at the nitrogen and carbon atoms, as well as on the electrophilic reagent, the diaziridine ring can undergo opening at both the C—N and N—N bonds. However, most reactions of this type have been carried out with unsubstituted (at one or both N atoms) diaziridines. Recently,[4—6] we have studied in detail reactions of 1,2-dialkyldiaziridines **1** with various ketenes and demonstrated that these reactions involve cleavage of the N—N bond to give three types of structures containing the N—C—N fragment: 1,3-dialkylimidazolidin-4-ones (1 : 1 adducts), 1-alkyl-3,3-diaryl-4-diarylacetylamino-azetidin-2-ones (β-lactam derivatives), and 3,5-diacyl-3,5-diazahept-1-enes (1 : 2 adducts). Ring opening at the N—N bond also occurs in reactions of diphenylketene with 1,2,3-trialkyldiaziridines **2**, yielding, however, only linear products.[7,8] In contrast, reactions of 1,2,3-trialkyldiaziridines **2a,b** with benzoyl isocyanate (**3a**) in boiling benzene[7] give, through opening of the diaziridine ring at the C—N bond, 1,2,5-trialkyl-4-benzoyl-1,2,4-triazolidin-3-ones **4a,b**. The formation of compound **4** involves zwitterionic intermediate **5**, in which the conjugated anion attacks the C atom of the diaziridine ring with simultaneous cleavage of the C—N bond (Scheme 1).[7]

Scheme 1



R = Me (**a**), Et (**b**)

i. PhCONCO (**3a**), 75—80 °C, C_6H_6

To establish the general features of this reaction for various substituents in the diaziridine ring, here we studied reactions of 1,2-dialkyl- (**1**) and 1,2,3,3-tetraalkyldiaziridines (**6**) with benzoyl and *p*-chlorobenzoyl isocyanates **3a,b**. 1,2-Diethyl-, 1,2-dipropyl-, and 1,2-dibutyldiaziridines **1a—c** were used. Reaction conditions for compounds **1** and **3** were optimized with 1,2-dipropyldiaziridine (**1b**) and benzoyl isocyanate (**3a**) as an example: the reaction was carried out in dry CH_2Cl_2 at −20 to 20 °C. The resulting suspension was kept at 20 °C for 1 h. TLC analysis revealed no significant amounts of chromatographed products in CH_2Cl_2 under these conditions. New compounds were detected by TLC only after the evaporation of the solvent and the extraction of the residue with hot hexane. For this reason, in later experiments, the reaction mixture was refluxed in hexane and the products were additionally extracted with hot hexane.

The product isolated by column chromatography on SiO_2 was close to previously reported[7] 1,2,4-triazolidin-3-one derivatives **4a,b** (^{1}H NMR data). However, the formation of isomeric 2-benzoyl-1,4-dipropyl-1,2,4-

Published in Russian in *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 3, pp. 534—538, March, 2006.

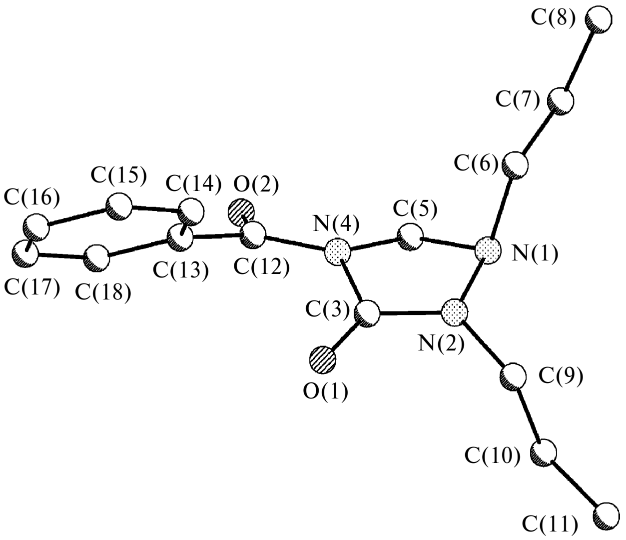
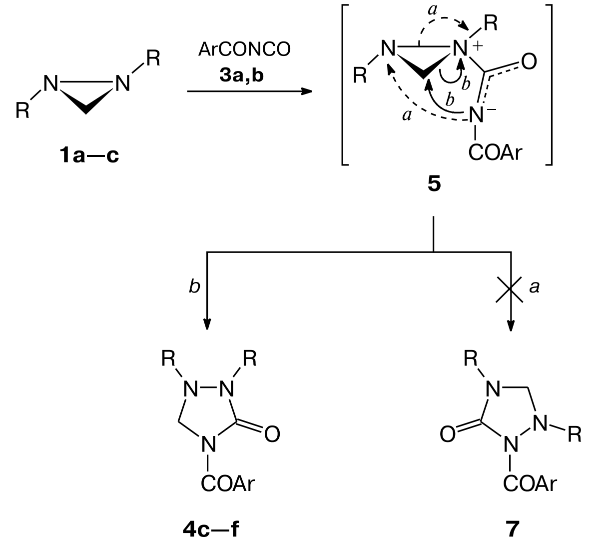


Fig. 1. General view of structure **4c**.

triazolidin-3-one **7a** in this reaction seemed to be not improbable (Scheme 2, pathway *a*) since reactions of various diaziridines with aryl(diaryl)ketenes give different products.[4—8] The NOE data for the compound obtained were ambiguous; for this reason, its structure was proven by X-ray diffraction analysis. It turned out that the reaction of diaziridine **1b** with benzoyl isocyanate **3a** yields 4-benzoyl-1,2-dipropyl-1,2,4-triazolidin-3-one (**4c**) (Fig. 1) *via* cleavage of the C—N bond in the three-membered ring. Other 1,2-dialkyldiaziridines **1** were also used in reactions with aroyl isocyanates **3a,b** under these conditions; the yields of 1,2,4-triazolidin-3-ones **4c—f** were 34—46% (Scheme 2, pathway *b*).

Scheme 2

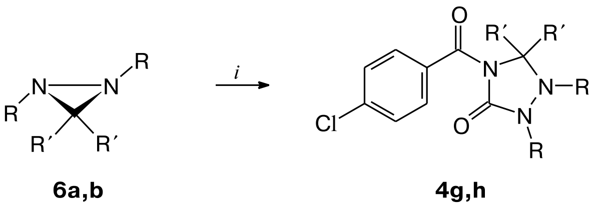


c: R = Pr^n, Ar = Ph; **d:** R = Et, Ar = 4-ClC$_6$H$_4$;
e: R = Bu^n, Ar = Ph; **f:** R = Et, Ar = Ph

Additional heating is required probably because of the sufficient stabilities of zwitterionic intermediates **5**. Examples of stable zwitterionic systems have been documented. For instance, the first step in reactions of the Schiff bases with isocyanates is the formation of zwitterionic intermediates isolated in some cases.[9] Stable salts of diaziridinium cations (*e.g.*, picrates) obtained in dry media are also known.[10] Transformation of intermediates **5** into 1,2,4-triazolidine derivatives **4** is thermodynamically controlled. The formation of triazolidines **4** rather than isomeric compounds **7** is probably due to thermodynamically more favorable cleavage of the C—N bond of the diaziridine ring with subsequent new C—N bonding than cleavage of the N—N bond followed by new N—N bonding.

In reactions of 1,2,3,3-tetraalkyldiaziridines **6** with aroyl isocyanates, we used 1,2-dimethyl-3,3-pentamethylenediaziridine (**6a**), 1,2-diethyl-3,3-dimethyldiaziridine (**6b**), and *p*-chlorobenzoyl isocyanate (**3b**). It turned out that the presence of additional alkyl substituents at the C atom of the diaziridine ring does not change its behavior in reactions with aroyl isocyanates: the reaction products were 4-benzoyl-1,2,4-triazolidin-3-one derivatives **4g,h** (compound **4g** has a spiran structure) (Scheme 3).

Scheme 3



i. 4-ClC$_6$H$_4$CONCO (**3b**)

g: R = Me, R′ + R′ = —(CH$_2$)$_5$—; **h:** R = Et, R′ = Me

The structures of all the compounds obtained were confirmed by data from elemental analysis, ^{1}H and ^{13}C NMR spectroscopy, and mass spectrometry. Since 1,2,3,3-tetraalkyldiaziridines have not been employed in reactions with aroyl isocyanates, the structure of spiran **4g** was confirmed by X-ray diffraction analysis (Fig. 2). The yields and selected physicochemical and spectroscopic characteristics of the compounds obtained are given in Tables 1 and 2. The starting diaziridines **1a—c** were prepared as described earlier.[11] Diaziridines **6a** and **6b** have been documented;[12,13] however, the methods proposed for their synthesis are insufficiently suitable for preparative purposes (gas-phase chlorination or use of not easily accessible reagents). In addition, some of the physicochemical and spectroscopic characteristics of compounds **6a,b** have been omitted.[12,13] That is why here we pro-

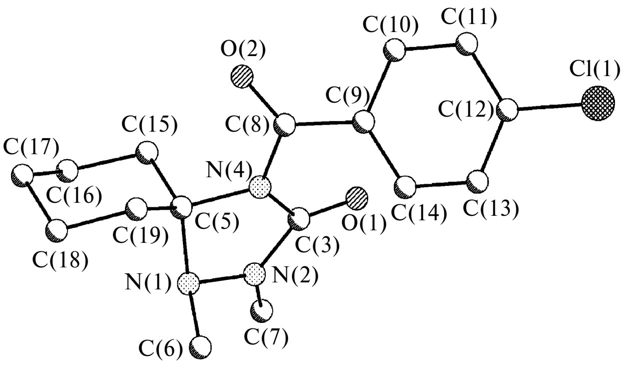


Fig. 2. General view of structure **4g**.

posed a simple one-step route to diaziridines **6a,b** and obtained missing spectroscopic characteristics.

X-ray diffraction analysis of compounds **4c** and **4g** revealed that their basic geometrical parameters are virtually identical; in particular, the presence of the spiran structure in compound **4g** only slightly lengthens the N(1)—C(5) and N(4)—C(5) bonds (Table 3). In both compounds **4c** and **4g**, the five-membered ring exists in the envelope conformation with a deviation of the C(5) atom by 0.44 and 0.59 Å, respectively. Because of the amide conjugation, the configurations of the N(4) and N(2) atoms are flattened: the sums of their bond angles are 355.6(2)—355.1(2)° and 359.3(2)—358.8(2)°, respectively. In contrast, the N(1) atom has a pyramidal configuration in both structures: the sum of its bond angles is 322(2)—323.7(2)°.

Experimental

IR spectra were recorded on a UR-20 spectrometer in KBr pellets (for crystalline substances) and in thin films (for oils). ^{1}H NMR spectra were recorded on Bruker WM-250 (250 MHz), Bruker AM-300 (300 MHz), and Bruker AC-200 spectrometers (200 MHz). ^{13}C NMR spectra were recorded on Bruker AM-300 (75.5 MHz) and Bruker RDX-500 spectrometers (125 MHz). Chemical shifts are given on the δ scale with reference to $SiMe_4$. New compounds were isolated by column chromatography on Kieselgel 60 F_{254} (Merck). The course of the reactions was monitored and the purity of the compounds obtained was checked by TLC on ALUGRAM® SIL $GLUV_{254}$ plates (Aldrich). Spots were visualized under UV light, in the iodine vapor, and, independently, by spraying a solution of diphenylamine in acetone onto plates followed by their heating. Melting points were determined on a GALLENKAMP instrument (Sanyo). X-ray diffraction analyses were carried out on Smart 1000 CCD and Syntex $P2_1$ diffractometers.

Synthesis of 4-aroyl-1,2,4-triazolidin-3-ones 4 (general procedure). A solution of aroyl isocyanates **3a,b** (4.5 mmol) in dichloromethane (5 mL) was added dropwise under argon at −20 °C for 20 min to a vigorously stirred solution of diaziridine **1a—c** or **6a,b** (5 mmol) in dry dichloromethane (10 mL). The reaction mixture was kept at −20 °C for 1 h, warmed to room temperature, and stirred for an additional 1 h. The solvent was removed in water aspirator vacuum. The resulting reaction mixture was refluxed with hexane and the solvent was decanted. Extraction with boiling hexane was repeated until compounds **4** disappeared from the extract (TLC monitoring). Compound **4g** precipitated from the extract on cooling to room temperature. The combined extracts were concentrated. Products **4c—f,h** were isolated by column chromatography on SiO_2 (60 F_{254}, 0.063—0.200 mm, Merck) with hexane—ethyl acetate (9 : 1) as an eluent followed by crystallization from hexane (for **4c,d,f,h**). 4-Benzoyl-1,2-dipropyl-1,2,4-triazolidin-3-one **4c** was obtained in 34% yield as colorless crystals suitable for X-ray diffraction analysis; 4-(4-chlorobenzoyl)-1,2-diethyl-1,2,4-triazolidin-3-one (**4d**) (43%), a colorless crystalline solid; 4-benzoyl-1,2-dibutyl-1,2,4-triazolidin-3-one (**4e**) (46%), a light yellow oil; 4-benzoyl-1,2-diethyl-1,2,4-triazolidin-3-one (**4f**) (37%), a white crystalline solid; 4-(4-chlorobenzoyl)-1,2-dimethyl-5,5-pentamethylene-1,2,4-triazolidin-3-one (**4g**) (37%), colorless

Table 1. Yields and selected physicochemical characteristics of 4-aroyl-1,2,4-triazolidin-3-ones **4**

Compound	Yield (%)	M.p. /°C	R_f[a]	Found/Calculated (%) C	H	N	Hal	Molecular formula
4c	34	59—61	0.55	65.86 65.43	7.90 7.69	15.07 15.26	—	$C_{15}H_{21}N_3O_2$
4d	43	96—97	0.50	55.73 55.42	5.84 5.72	14.67 14.91	12.36 12.58	$C_{13}H_{16}ClN_3O_2$
4e	46	Oil	0.59	67.51 67.30	8.47 8.31	13.73 13.85	—	$C_{17}H_{25}N_3O_2$
4f	37	89—92	0.48	63.25 63.14	7.01 6.93	16.87 16.99	—	$C_{13}H_{17}N_3O_2$
4g	37	138—140	0.45	59.97 59.72	6.36 6.26	12.98 13.06	10.86 11.02	$C_{16}H_{20}ClN_3O_2$
4h	21	124—127	0.49	58.24 58.16	6.74 6.51	13.42 13.56	11.31 11.44	$C_{15}H_{20}ClN_3O_2$

[a] With *n*-heptane—ethyl acetate (8 : 2) as an eluent.

Table 2. IR and ^{1}H and ^{13}C NMR spectra ($CDCl_3$) of 4-aroyl-1,2,4-triazolidin-3-ones **4**^{a}

Compound	IR, ν/cm^{-1}	^{1}H NMR	^{13}C NMR
		δ (*J*/Hz)	
4c	2968, 2936, 2844, 1736, 1664, 1472, 1348, 1248, 1108, 716	0.91, 1.01 (both t, 3 H each, Me, $^3J = 7.33$); 1.53—1.72 (m, 4 H, 2 $CC\underline{H}_2Me$); 2.84 (t, 2 H, $—(CH_2)_{cycl}NC\underline{H}_2$, $^3J = 7.33$); 3.33 (br.s, 2 H, $—(CO)_{cycl}NCH_2$); 4.84 (s, 2 H, $(CH_2)_{cycl}$); 7.38—7.57 (m, 2 H, Ph); 7.67 (d, 2 H, Ph, $^3J = 6.84$)	11.04, 11.40 (Me); 19.44, 20.26 ($\underline{C}H_2Me$); 44.98, 56.71 ($N\underline{C}H_2C$); 66.28 ($(CH_2)_{cycl}$); 127.52, 128.40, 131.60, 133.75 (Ph); 152.51 ($(CO)_{cycl}$); 168.49 ($(CO)_{exocycl}$)
4d	2976, 2936, 2884, 1728, 1664, 1596, 1380, 1336, 1268, 1088, 848, 760	1.20, 1.22 (both t, 3 H each, Me, $^3J = 6.62$); 2.94 (q, 2 H, $(CH_2)_{cycl}NC\underline{H}_2$, $^2J = 13.97$, $^3J = 6.62$); 3.40 (br.s, 2 H, $(CO)_{cycl}NCH_2$); 4.82 (s, 2 H, $(CH_2)_{cycl}$); 7.38, 7.61 (both d, 2 H each, Ar, $^3J = 8.09$)	11.66, 12.89 (Me), 39.60, 50.37 ($\underline{C}H_2Me$); 66.60 ($(CH_2)_{cycl}$); 128.07, 129.13, 131.70, 131.84 (*p*-ClPh); 152.96 ($(CO)_{cycl}$); 168.49 ($(CO)_{exocycl}$)
4e	2960, 2932, 2872, 1740, 1668, 1452, 1344, 1232, 1108, 712	0.91, 0.97 (both t, 3 H each, Me, $^3J = 7.22$); 1.26—1.67 (m, 8 H, 2 $C\underline{H}_2(Bu)$); 2.86 (t, 2 H, $(CH_2)_{cycl}NC\underline{H}_2$, $^3J = 7.22$); 3.37 (br.s, 2 H, $(CO)_{cycl}NCH_2$); 4.82 (s, 2 H, $(CH_2)_{cycl}$); 7.37—7.55 (m, 3 H, Ph); 7.67 (d, 2 H, Ph, $^3J = 6.56$)	13.66, 13.94 (Me); 19.85, 20.32 ($\underline{C}H_2Me$); 28.90, 29.53 ($CH_2\underline{C}H_2CH_2$); 43.76, 55.48 ($N\underline{C}H_2C$); 66.75 ($(CH_2)_{cycl}$); 127.65, 129.13, 131.97, 133.49 (Ph); 152.97 ($(CO)_{cycl}$); 169.65 ($(CO)_{exocycl}$)
4f	2970, 2930 2881, 2840, 1726, 1662, 1594, 1337, 1266, 1085, 1012, 836, 750	1.20 (m, 6 H, 2 Me); 2.93 (q, 2 H, $(CH_2)_{cycl}NC\underline{H}_2$, $^2J = 13.5$, $^3J = 6.2$); 3.40 (br.s, 2 H, $(CO)_{cycl}NCH_2$); 4.83 (s, 2 H, $(CH_2)_{cycl}$); 7.39—7.67 (m, 5 H, Ph)	11.57, 12.92 (Me); 39.58, 50.38 ($\underline{C}H_2Me$); 66.59 ($(CH_2)_{cycl}$); 127.55, 128.53, 131.64, 133.72 (Ph); 152.67 ($(CO)_{cycl}$); 168.58 ($(CO)_{exocycl}$)
4g	3076, 2952, 1732, 1672, 1600, 1488, 1460, 1380, 1096, 848, 772	1.30—2.15 (m, 10 H, $(CH_2)_5$); 2.64, 3.04 (both s, 3 H each, Me); 7.37, 7.54 (both d, 2 H each, Ph, $^3J = 6.56$)	22.51, 24.93, 27.04, 32.35 (CH_2); 31.14, 36.63 (Me); 84.17 (C_{cycl}); 128.14, 130.27, 134.27, 138.01 (*p*-ClPh); 154.21 ($(CO)_{cycl}$); 169.23 ($(CO)_{exocycl}$)
4h	2957, 2943, 2884, 1795, 1636, 1486, 1334, 1239, 1102, 773	1.22, 1.24 (both t, 3 H each, CH_2Me, $^3J = 6.8$); 1.32, 1.34 (both s, 3 H each, CMe); 2.86 (q, 2 H, $C_{cycl}NCH_2$, $^2J = 13.5$, $^3J = 6.8$); 3.32 (br.s, 2 H, $(CO)_{cycl}NCH_2$); 7.61, 7.61 (both d, 2 H each, Ar, $^3J = 8.09$)	11.76, 12.31 ($CH_2\underline{Me}$); 23.34, 25.17 ($C\underline{Me}$); 39.07, 48.68 ($\underline{C}H_2Me$); 82.18 (C_{cycl}); 128.56, 130.75, 134.96, 138.45 (*p*-ClPh); 153.59 ($(CO)_{cycl}$); 168.93 ($(CO)_{exocycl}$)

[a] IR spectra were recorded in KBr pellets (for **4c,d,f—h**) and in a thin film of the individual compound (**4e**). ^{1}H NMR spectra were recorded in $CDCl_3$ on Bruker WM-250 (250 MHz) (for **4e,f,h**), Bruker AC-200 (200 MHz) (**4c**), and Bruker AM-300 spectrometers (300 MHz) (**4d,g**). ^{13}C NMR spectra were recorded in DMSO-d_6 (for **4c**) and $CDCl_3$ (**4d—h**) on Bruker AM-300 (75.5 MHz) (for **4c,e—h**) and Bruker RDX-500 spectrometers (125 MHz) (**4d**).

Table 3. Selected bond lengths (*d*) and angles (ω) in compounds **4c** and **4g**

Bond	*d*/Å	Bond	*d*/Å	Angle	ω/deg	Angle	ω/deg
4c		**4g**		**4c**		**4g**	
O(1)—C(3)	1.213(2)	O(1)—C(3)	1.2112(15)	N(2)—N(1)—C(5)	101.20(13)	N(2)—N(1)—C(6)	107.31(10)
N(1)—N(2)	1.437(2)	N(1)—N(2)	1.4301(14)	N(2)—N(1)—C(6)	109.07(14)	N(2)—N(1)—C(5)	101.39(8)
N(1)—C(5)	1.467(2)	N(1)—C(6)	1.4737(17)	C(5)—N(1)—C(6)	111.71(16)	C(6)—N(1)—C(5)	115.00(11)
N(1)—C(6)	1.477(2)	N(1)—C(5)	1.4852(16)	C(3)—N(2)—N(1)	114.16(15)	C(3)—N(2)—N(1)	113.57(10)
N(2)—C(3)	1.345(2)	N(2)—C(3)	1.3516(15)	C(3)—N(2)—C(9)	125.21(16)	C(3)—N(2)—C(7)	125.48(11)
N(2)—C(9)	1.449(2)	N(2)—C(7)	1.4437(16)	N(1)—N(2)—C(9)	119.91(14)	N(1)—N(2)—C(7)	119.68(10)
C(3)—N(4)	1.423(2)	C(3)—N(4)	1.4231(15)	C(12)—N(4)—C(3)	128.87(15)	C(8)—N(4)—C(3)	123.98(10)
N(4)—C(12)	1.387(2)	N(4)—C(8)	1.3936(14)	C(12)—N(4)—C(5)	119.36(15)	C(8)—N(4)—C(5)	124.52(10)
N(4)—C(5)	1.466(2)	N(4)—C(5)	1.4944(15)	C(3)—N(4)—C(5)	107.31(15)	C(3)—N(4)—C(5)	106.63(9)

Table 4. Selected crystallographic parameters and a summary of data collection and refinement for compounds **4c** and **4g**

Parameter	**4c**	**4g**
Molecular formula	$C_{15}H_{21}N_3O_2$	$C_{16}H_{20}ClN_3O_2$
T/K	120	193
Diffractometer	Smart CCD	Syntex $P2_1$
Scan mode	ω	θ/2θ
Crystal system	Monoclinic	Orthorhombic
Space group	$P2_1/c$	$Pbca$
a/Å	12.573(2)	29.711(6)
b/Å	5.525(1)	11.309(2)
c/Å	21.242(4)	9.337(2)
β/deg	102.290(5)	
V/Å^3	1441.8(5)	3137.2(11)
$Z(Z')$	4(1)	8(1)
M	275.35	321.80
μ/cm^{-1}	0.86	2.55
F(000)	592	1360
ρ_{calc}/g cm^{-3}	1.268	1.363
$2\theta_{max}$/deg	56.00	60.00
Number of measured reflections (R_{int})	6650 (0.0302)	4538 (0.00)
Number of independent reflections	3298	4538
Number of reflections with $I > 2\sigma(I)$	2032	3487
Number of parameters refined	265	279
R_1	0.0511	0.0393
wR_2	0.1226	0.1143
GOOF	0.969	1.004
ρ/e Å^{-3} (max/min)	0.199/–0.204	0.310/–0.343

crystals suitable for X-ray diffraction analysis; 4-(4-chlorobenzoyl)-1,2-diethyl-5,5-dimethyl-1,2,4-triazolidin-3-one (**4h**) (21%), a colorless crystalline solid.

Synthesis of 1,2-dimethyl-3,3-pentamethylenediaziridine (6a) and 1,2-diethyl-3,3-dimethyldiaziridine (6b). A solution of NaOCl (0.15 mol) prepared from NaOH (12.6 g, 0.315 mol) and Cl_2 (10.65 g, 0.15 mol) in water (60 mL) was added dropwise at 0—5 °C to a 20% aqueous solution of methyl- or ethylamine (0.15 mol), respectively, in the presence of dissolved $NaHCO_3$ (3 g) at the same temperature. The reaction mixture was saturated with NaCl and the organic material was extracted with chloroform (3×30 mL). The extract was dried with a small amount of K_2CO_3 for 5—10 min. The yields of MeNHCl and EtNHCl were 0.11 mol (73%) and 0.12 mol (80%), respectively (from iodometric data). Finely divided K_2CO_3 (0.3 mol, 41.5 g), methylamine (0.11 mol) or ethylamine (0.12 mol) as a 20—25% solution in chloroform, and cyclohexanone (0.11 mol, 11.3 mL) or acetone (0.12 mol, 8.8 mL), respectively, were added to the stirred extract. The reaction mixture was stirred at 18—20 °C for 24—28 h. The inorganic precipitate was filtered off and thoroughly washed with chloroform and the solvent was removed. The yields of diaziridines **6a** (0.0714 mol, 65%) and **6b** (0.0853 mol, 71%) were determined by iodometric titration in the presence of catalytic amounts of $CuCl_2$ (or $Cu(OAc)_2$). The products were twice distilled in water aspirator vacuum, the second distillation being carried out over K_2CO_3. Fractions with b.p. 71—75 °C (12 Torr) for **6a** and b.p. 47—50 °C (12 Torr) for **6b** were collected. The final yields of compounds **6a** and **6b** were 6.4 and 6.9 g (isolation losses were 36 and 37%, respectively). 1,2-Dimethyl-3,3-pentamethylenediaziridine **6a**. ^{1}H NMR ($CDCl_3$), δ: 1.43 (m, 2 H, CH_2); 1.49 (m, 8 H, CH_2); 2.37 (s, 6 H, NMe); n_D^{20} 1.4687. The ^{13}C NMR spectrum agrees with the literature data.[12] 1,2-Diethyl-3,3-dimethyldiaziridine **6b**. ^{1}H NMR ($CDCl_3$), δ: 1.07 (t, 6 H, CH_2Me, ^3J = 7.18 Hz); 1.23 (s, 6 H, $C_{cycl}Me$); 2.36, 2.50 (both m, 2 H each, NCH_2). ^{13}C NMR ($CDCl_3$), δ: 13.8 (CH_2Me); 19.4 ($C_{cycl}Me$); 47.4 (NCH_2); 60.6 (C_{cycl}).

X-ray diffraction analysis. Structures **4c** and **4g** were solved by the direct method and refined by the least-squares method in the anisotropic full-matrix approximation on F^2_{hkl}. Hydrogen atoms were located from the electron density difference maps and refined isotropically. All calculations were performed with the SHELXTL PLUS program package. Selected bond lengths and angles are listed in Table 3. A summary of data collection and refinement is given in Table 4.

This work was financially supported by the Russian Foundation for Basic Research (Project No. 04-03-32799).

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Received November 11, 2005;
in revised form March 2, 2006