Reaction of 1,2-dialkyldiaziridines with ketenes as a new approach to cyclic and linear systems containing the N—C—N fragment*

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The reaction of 1,2-dialkyldiaziridines with ketenes proceeds through the N—N bond cleavage to form three types of structures containing the N—C—N fragment, *viz.*, 1,3-dialkylimidazolidin-4-ones, 3,5-diacyl-3,5-diazahept-1-enes, and β-lactams. The reaction pathway depends on the reaction conditions and the structures of the starting compounds.

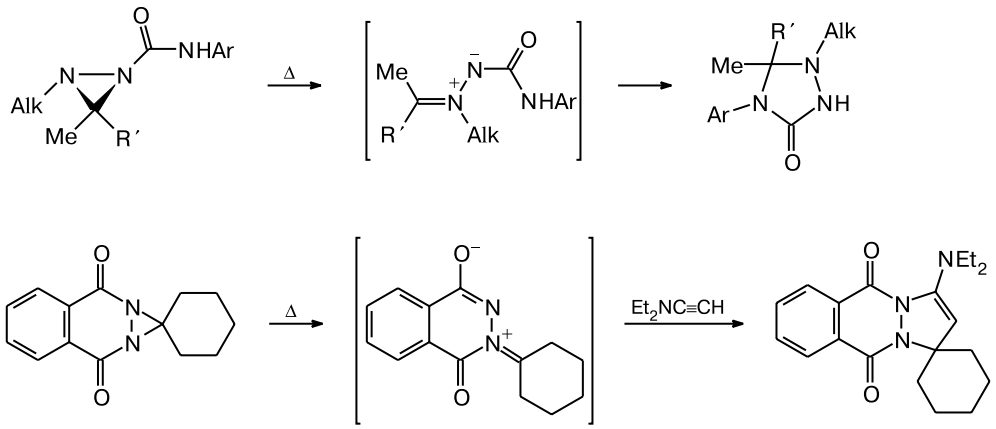
Key words: diaziridines, ketenes, imidazolidin-4-ones, alkenylacetamides, β-lactams, X-ray diffraction analysis.

Diaziridines are representatives of strained three-membered heterocycles, which are highly prone to reactions accompanied by a decrease in the strain energy.[1—3] Most studies of the reactions of diaziridines with electrophilic reagents concerned with *trans*-3-mono- or 3,3-disubstituted derivatives, in which one or both nitrogen atoms are unsubstituted. The introduction of an electron-withdrawing substituent at the nitrogen atoms of such diaziridines facilitates the diaziridine ring opening at the C—N bond and promotes the formation of reactive intermediates, in particular, of azomethine imine-type compounds, which are then stabilized through the formation of a new linear or heterocyclic system containing the N—N fragment. The reaction can proceed both intramolecularly and intermolecularly through the 1,3-dipolar cycloaddition to an appropriate dipolarophile (see, for example, Scheme 1).[4—6]

1,2-Dialkyldiaziridines **1** behave differently in reactions with electrophilic reagents, although only a few examples were described in the literature.[7—9] In particular,

* Dedicated to Corresponding Member of the Russian Academy of Sciences E. P. Serebryakov on the occasion of his 70th birthday.

Scheme 1



R′ = Alk, Ph

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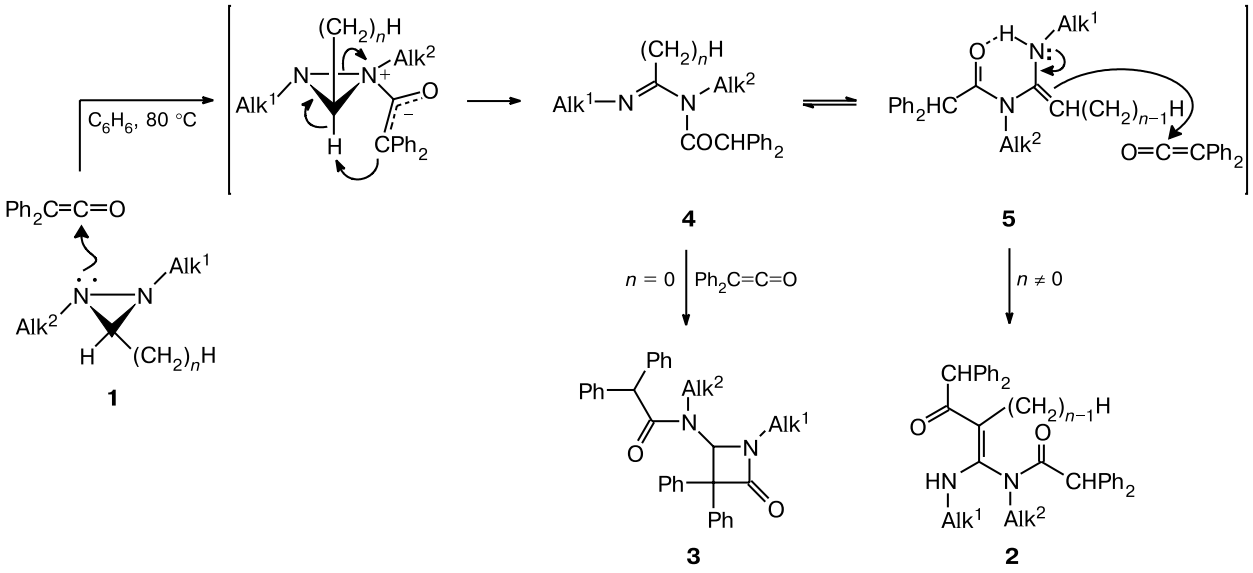
Scheme 2

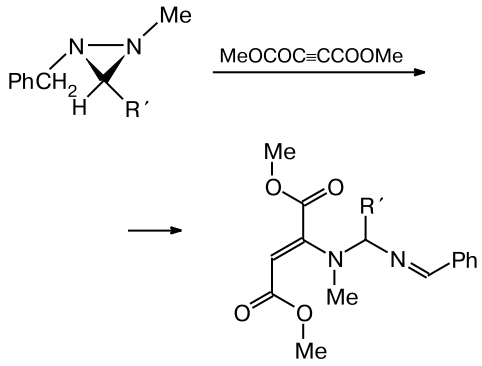


the reaction of diphenyl ketene with 1,2-dialkyldiaziridines **1** in boiling benzene[7,8] is accompanied by the N—N bond cleavage to form 1 : 2 adducts, whose structures depend on the substitution pattern at the carbon atom of the starting diaziridine. The reactions of 3-monosubstituted 1,2-dialkyldiaziridines **1** ($n \neq 0$) afford acyclic product **2**, whereas the reactions with 3-unsubstituted 1,2-dialkyldiaziridines ($n = 0$) produce β-lactam derivatives **3**, which are structurally related to β-lactam antibiotics (Scheme 2). The probable reaction mechanism involves the nucleophilic attack of the nitrogen atom of diaziridine on the sp-hybridized carbon atom of diphenyl ketene followed by the proton abstraction from the C(3) atom of the diaziridine ring and the N—N bond cleavage to form amidine-type intermediate **4**. At $n = 0$, this intermediate is involved in the 1,2-cycloaddition at the CH=N fragment with the second diphenyl ketene molecule to give β-lactam **3**. At $n \neq 0$, the addition of the second ketene molecule proceeds through the second aminal-type intermediate **5** to yield acyclic product **2** (see Scheme 2).

The reactions involving phenyl and benzoyl isocyanates were carried out only with 1,2,3-trialkyldiaziridines.[8] The former reaction afforded both linear and cyclic products (in particular, hexahydro-1,3,5-triazin-2-one and 1,2,4-triazolidine derivatives). However, all products were obtained in low yields, and the proposed mechanism of their formation was not conclusively proved. The reaction with dimethyl acetylenedicarboxylate was studied also only with 1,2,3-trialkyldiaziridines.[9] This reaction afforded the product of the N—N bond cleavage with a linear structure (Scheme 3).

Data on the chemical transformation of 1,2-dialkyldiaziridines unsubstituted at position 3 of the ring in the reactions with electrophilic reagents are scarce in the literature. In this connection, we thoroughly studied the reactions of 1,2-dialkyldiaziridines with electrophilic reagents, such as ketenes with different structures.* This approach would be expected to give both new β-lactam derivatives **3** and other types of structures containing the N—C—N fragment.

Scheme 3



In the first step of our investigation, we reproduced the synthesis of β-lactams **3** by the reactions of 1,2-dialkyldiaziridines **1a,b** with diphenyl ketene **6a** to confirm their structures, because these products have not been sufficiently characterized in the study.[8] The reaction was carried out under the same conditions (benzene, 80 °C), but diphenyl ketene **6a** was generated *in situ* from diphenylacetyl chloride **7a** by adding dropwise a mixture of the starting diaziridine and Et_3N to a benzene solution

* For preliminary communications, see Refs 10 and 11.

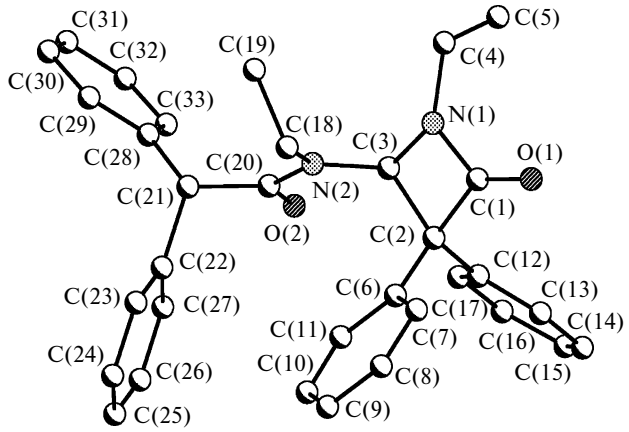


Fig. 1. Overall view of *N*-ethyl-*N*-(1-ethyl-4-oxo-3,3-diphenylazetidin-2-yl)-2,2-diphenylacetamide **3a**.

Table 1. Selected bond lengths (*d*) and bond angles (ω) in compound **3a**

Bond	*d*/Å	Bond	*d*/Å
O(1)—C(1)	1.205(3)	N(2)—C(20)	1.367(3)
O(2)—C(20)	1.226(3)	N(2)—C(3)	1.449(3)
N(1)—C(1)	1.365(3)	N(2)—C(18)	1.472(3)
N(1)—C(3)	1.453(3)	C(1)—C(2)	1.541(3)
N(1)—C(4)	1.458(3)	C(2)—C(3)	1.585(3)

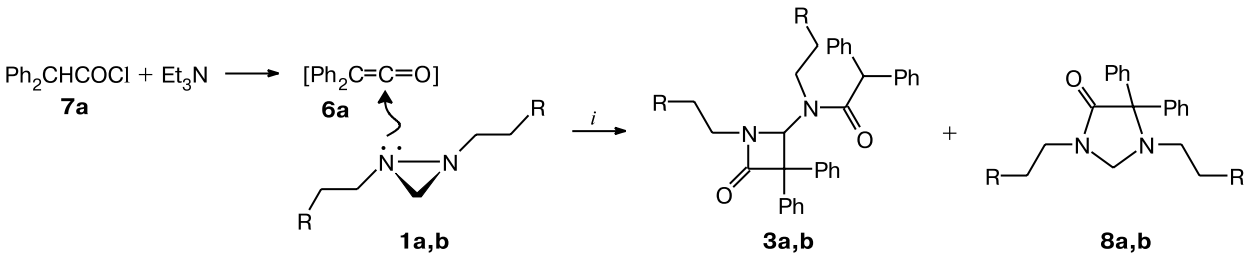
Bond angle	ω/deg	Bond angle	ω/deg
C(1)—N(1)—C(3)	96.4(2)	C(12)—C(2)—C(1)	111.7(2)
C(1)—N(1)—C(4)	129.8(2)	C(6)—C(2)—C(3)	117.1(2)
C(3)—N(1)—C(4)	129.3(2)	C(12)—C(2)—C(3)	115.3(2)
C(20)—N(2)—C(3)	115.4(2)	C(1)—C(2)—C(3)	84.5(2)
C(20)—N(2)—C(18)	123.7(2)	N(1)—C(3)—N(2)	114.5(2)
C(3)—N(2)—C(18)	120.2(2)	N(1)—C(3)—C(2)	86.9(2)
O(1)—C(1)—N(1)	131.5(3)	N(2)—C(3)—C(2)	120.5(2)
O(1)—C(1)—C(2)	136.7(3)	N(1)—C(4)—C(5)	113.6(2)
N(1)—C(1)—C(2)	91.9(2)	O(2)—C(20)—N(2)	120.5(2)
C(6)—C(2)—C(12)	110.3(2)	O(2)—C(20)—C(21)	121.0(2)
C(6)—C(2)—C(1)	115.9(2)	N(2)—C(20)—C(21)	118.5(2)

of **7a** heated to 60 °C. Actually, the reaction afforded 1 : 2 adducts, *viz.*, β-lactams **3a,b**, as the major reaction products in 32—41% yields. Their structures were confirmed by elemental analysis and spectroscopic methods. The structure of β-lactam **3a** was additionally confirmed by X-ray diffraction analysis (Fig. 1, Table 1). The ^{1}H NMR spectra of β-lactams **3** are characterized by the presence of signals at δ 2.7 corresponding to the diastereotopic protons of the CH_2 fragments of substituents bound to the nitrogen atom of the four-membered ring and a singlet of the cyclic CH fragment.

In the crystal of **3a**, the four-membered nitrogen-containing ring is flattened; the folding angle along the N(2)...C(3) line is 6.3°. The N(1) and N(2) atoms are slightly pyramidalized and deviate by 0.17 and 0.07 Å from the planes passing through the C(1), C(3), C(4) atoms and the C(3), C(18), C(20) atoms, respectively. Apparently, flattening of the nitrogen atoms, as well as shortening of the N(1)—C(1) and N(3)—C(20) bonds (1.365(3) and 1.367(3) Å, respectively), are attributable to conjugation with the carbonyl group. The amide group at the C(3) atom is located approximately in the bisecting plane of the four-membered ring; the O(2)—C(20)—N(2)—C(3) torsion angle is 2.0°.

However, the reaction of diphenyl ketene **6a** with 1,2-dialkylaziridines **1a,b** afforded not only β-lactams **3** but also 1 : 1 adducts, *viz.*, 1,3-dialkyl-5,5-diphenylimidazolidin-4-ones **8a,b**, in low yields (8—21%). The synthesis of the latter also proceeds through the diaziridine ring opening at the N—N bond (Scheme 4). The structures of compounds **8a,b** were confirmed by elemental analysis and spectroscopic characteristics. The structure of compound **8a** was additionally established by X-ray diffraction analysis.[10] An important distinguishing feature of the ^{1}H NMR spectra of compounds **8** is that the protons of the NCH_2N fragment appear at substantially lower field (δ 4.15) compared to the chemical shift of the analogous fragment of the starting diaziridine **1** characterized by an upfield shift (δ 2.35).

Scheme 4



i. C_6H_6, 60—80 °C
1, **3**, **8:** R = H (**a**), Me (**b**)

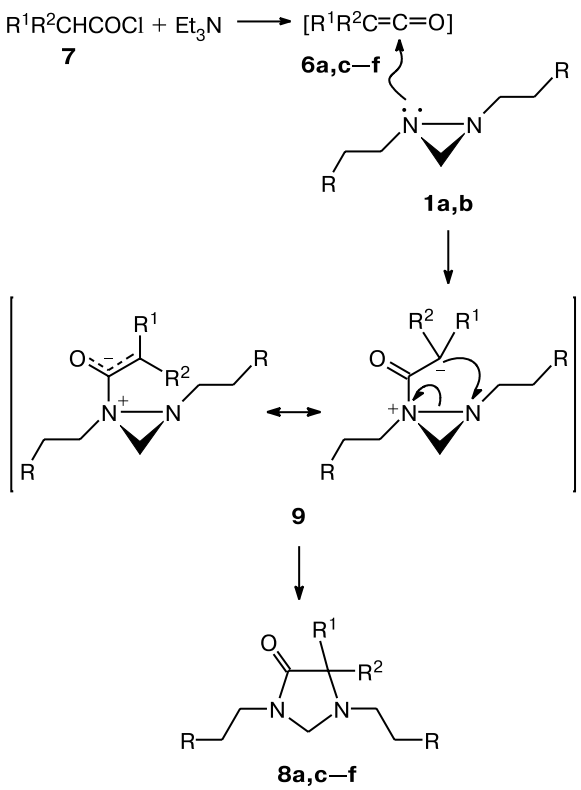
With the aim of examining the scope of the reaction of 1,2-dialkyldiaziridines **1** with ketenes, it was of interest to perform this reaction with other aryl ketenes. As suggested above (see Scheme 2), the formation of products **3** is associated with the thermal [2+2]-concerted cycloaddition of diphenyl ketene **6a** to amidine-type intermediate **4** generated upon the proton abstraction from the C(3) atom of the diaziridine ring. The formal [3+2]-cycloaddition of diphenyl ketene **6a** giving rise to compounds **8a,b**, which we have found in the present study, proceeds, apparently, *via* a two-step mechanism involving dipolar intermediate **9** followed by the N—N bond cleavage and cyclization giving rise to the imidazolidine ring (Scheme 5). The selectivity of such mixed processes is kinetically and thermodynamically controlled. Hence, to prepare compounds **8**, the reactions of 1,2-dialkyldiaziridines **1a,b** with aryl ketenes **6a,c—f** were performed at low temperature. Aryl ketenes **6** were generated *in situ* from arylacetyl chlorides **7** in the presence of Et_3N in anhydrous diethyl ether at −30 °C according to a standard procedure.[12] After the addition of the starting acid chloride to a mixture of dialkyldiaziridine **1** and Et_3N, the reaction mixture was kept at this temperature for 2 h and then kept at room temperature for 16 h. Under these conditions, the reactions produced only 1 : 1 adducts, *viz.*, 5-aryl-1,3-dialkylimidazolidin-4-ones **8a,c—f**, in 40—65% yields regardless of the acid chloride—diaziridine molar ratio, which was varied from 1 : 2 to 2 : 1. Interestingly, analogous product **8a** was prepared from diphenylacetyl chloride **7a**. The structures of compounds **8** were confirmed by elemental analysis and spectroscopic characteristics.

The formation of imidazolidin-4-ones **8** at low temperature confirmed the proposed mechanism of formal [3+2]-cycloaddition. Apparently, mild temperature conditions are insufficient for the proton abstraction from the carbon atom of the diaziridine ring in zwitterion **9**. This pathway would give intermediate **4** and then β-lactam **3** (the thermodynamically controlled process). Therefore, zwitterion **9** is transformed into imidazolidin-4-ones **8** (the kinetically controlled process) (see Scheme 5). Evidently, the reaction performed at higher temperature is also accompanied by the partial transformation of zwitterion **9** into imidazolidin-4-ones **8a,b**.

In addition to the reactions with aryl ketenes, we studied the behavior of 1,2-dialkyldiaziridines **1** in the reaction with parent ketene **6g** [11] (Scheme 6). This reaction can be performed only at low temperature, because ketene **6g** is prone to dimerization. Ketene **6g** was also generated *in situ* by adding dropwise acetyl chloride to a mixture of Et_3N and the starting diaziridine in dry diethyl ether at −40 °C.

1,2-Diethyl-, 1,2-dipropyl-, and 1,2-bis(2-phenylethyl)diaziridines **1a—c** were used as the starting 1,2-dialkyldiaziridines **1**. In all cases, the reactions of these

Scheme 5

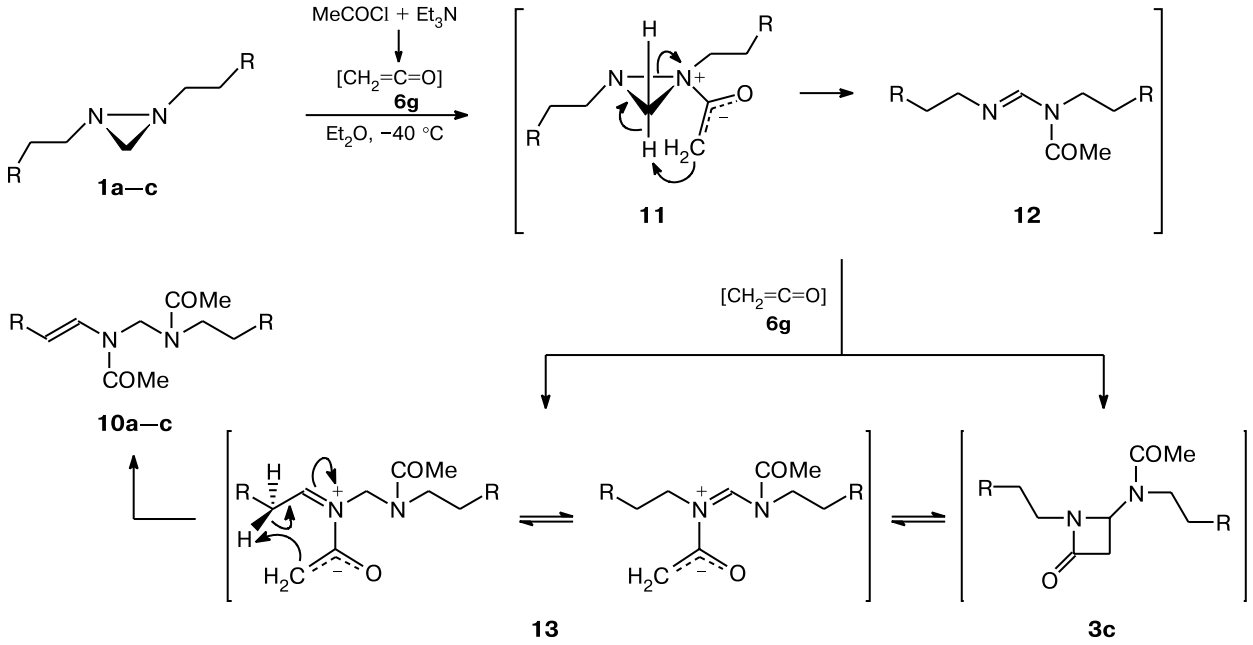


1: R = H (**a**), Me (**b**)

8	R	R^1	R^2
a	H	Ph	Ph
c	H	H	4-MeC_6H_4
d	H	H	4-ClC_6H_4
e	Me	H	4-BrC_6H_4
f	H	H	2-$NO_2C_6H_4$

compounds with the simplest ketene **6g** yielded compounds **10** with linear structures containing the 3,5-diacetyl-3,5-diazahept-1-ene fragment (see Scheme 6) as the major reaction products instead of the expected derivatives of β-lactams **3** or imidazolidin-4-ones **8**. The characteristic feature of the NMR spectra (primarily, of the ^1H NMR spectra) of compounds **10** is that the signal for one of the olefinic protons, the singlet of the NCH_2N fragment, and, in some cases, the signals of the NCH_2 fragment are doubled due, presumably, to the appearance of two rotamers (in a ratio of (7—10) : 1 depending on the temperature, at which the spectra were recorded, and the nature of the solvent). This phenomenon is typical of unsymmetrical *N*,*N*-disubstituted amides and is associated with hindered rotation about the N—Ac bond. In the spectra of the transformation products of diaziridines **1b,c**, the spin-spin coupling constants of the protons of the CH=CH groups are ~14 Hz for both rotamers, which is indicative of the *trans* configuration of the double bond.

Scheme 6



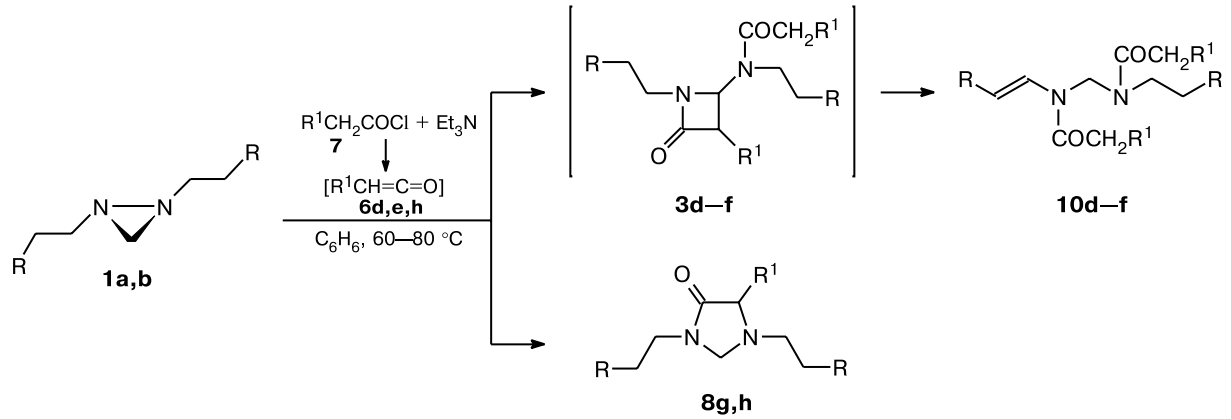
R = H (**a**), Me (**b**), Ph (**c**)

The possible mechanism of the formation of compounds **10** is shown in Scheme 6. The first step of the reaction, like that in the reaction of 1,2-dialkyldiaziridines **1** with aryl ketenes **6**, involves the attack of the nitrogen atom of the diaziridine ring on the central atom of ketene **6g** to form zwitterionic intermediate **11**, whose carbanionic center is stabilized to a much lesser extent than those obtained with the use of aryl-substituted ketenes **6a,c—f**. Then, as expected, the negative charge of zwitterion **11** causes abstraction of one hydrogen atom bound to the carbon atom of the diaziridine ring and the N—N bond cleavage to give intermediate *N*-acetylamidine **12**. The next step of the reaction should involve the [2+2]-cycloaddition of the second molecule of ketene **6g** at the double bond of *N*-acetylamidine **12** to form β-lactam **3c**, taking into account that the reaction of C=N-containing structures with ketenes serves as one of the most general methods for the preparation of β-lactams.[13] However, it is known[13,14] that 3-unsubstituted β-lactams can be cleaved to give dipolar intermediates stabilized as linear products. The structures of these compounds are determined by the character of substituents at the other atoms of the four-membered ring. In our case, the resulting β-lactam **3c** is, apparently, cleaved to form new intermediate **13**, which is stabilized as linear diamide **10**. However, it cannot be ruled out that the formation of intermediate **13** occurs due to acylation of the nitrogen atom of N-acetylamidine **12** in the reaction with ketene **6g** (Scheme 6) rather than through the formation of β-lactam **3c**.

The data published in the literature[7,8] and our results[10,11] suggest that β-lactams **3** should be prepared by the reactions of 1,2-dialkyldiaziridines **1** with ketenes at high temperature. For this purpose, we used diaziridines **1a,b** and 4-chlorophenyl ketene (**6d**), 4-bromophenyl ketene (**6e**), and 4-chlorophenoxyketene (**6h**). The reaction of 4-chlorophenoxyketene **6h** with 1,2-diethyldiaziridine **1a** at 60—80 °C produced the expected β-lactam **3f** (Scheme 7). The ^{1}H NMR spectrum of the crude product obtained after the treatment of the reaction mixture without heating shows signals for the diastereotopic protons of the methylene units and the expected doublets for the CH protons of the ring at δ 4.72 and 4.89. An attempt to purify this product by passing through a SiO_2 layer led to the partial transformation of cyclic system **3f** into open form **10f**. Column chromatography on SiO_2 led to the complete transformation of **3f**. Under the same conditions, the reactions with 4-chlorophenyl and 4-bromophenyl ketenes **6d,e** followed by chromatography on SiO_2 afforded linear reaction products, *viz.*, *N*-alkenylacetamides **10d,e**, along with imidazolidin-4-ones **8g,h**.

To summarize, the investigation of the reaction of 1,2-dialkyldiaziridines **1** with aryl ketenes **6** in diethyl ether at −30 °C revealed a new pathway for the ring expansion in 1,2-dialkyldiaziridines giving rise to 5-aryl(5,5-diaryl)-1,3-dialkylimidazolidin-4-ones **8** in 40—65% yields. It should be noted that this reaction provides a new simple and general approach to the synthesis of *N*-alkyl-substituted imidazolidin-4-ones. It should be emphasized that

Scheme 7



	8g, **10d**	**8h**, **10e**	**10f**
R	H	Me	H
R^1	4-BrC_6H_4	4-ClC_6H_4	4-ClC_6H_4O

the known methods for the preparation of the latter compounds are based on multistep processes.[15,16] An analogous reaction at high temperature produces a mixture of 5-aryl-1,3-dialkylimidazolidin-4-ones **8** and a new type of structures, *viz.*, *N*-alkenylacetamides **10**. It was hypothesized (and then confirmed experimentally using one example) that the formation of compounds **10** proceeds through the intermediate formation of the corresponding β-lactams **3**. Stable 3,3-disubstituted azetidin-2-ones **3** can be synthesized only by the reaction of 1,2-dialkyldiaziridines **1** with diphenyl ketene **6a**. The reaction of 1,2-dialkyldiaziridines **1** with the simplest ketene **6g**, like other known reactions of compounds **1** with substituted ketenes, is accompanied by the N—N bond cleavage. In the latter case, only linear 3,5-diacetyl-3,5-diazahept-1-enes **10** were isolated.

Experimental

The IR spectra were recorded on a UR-20 spectrometer in KBr pellets (for crystalline compounds) and in a thin layer (for oils). The ^{1}H NMR spectra were measured on Bruker WM-250 (250 MHz), Bruker AM-300 (300 MHz), and Bruker DRX-500 (500 MHz) spectrometers. The ^{13}C NMR spectra were recorded on Bruker AM-300 (75.5 MHz) and Bruker DRX-500 (125 MHz) spectrometers. The chemical shifts are given in the δ scale relative to the signal of Me_4Si. The TLC analysis was carried out on Silufol-UV-254 plates; spots were visualized using a UV lamp, with I_2 vapor, and by spraying with a solution of diphenylamine in acetone followed by heating of the plates. The melting points were determined on a GALLENKAMP instrument (Sanyo). X-ray diffraction study was carried out on a Smart 1000 CCD diffractometer.

***N*-Alkyl-*N*-(1-alkyl-4-oxo-3,3-diphenylazetidin-2-yl)-2,2-diphenylacetamides (3a,b) and 1,3-dialkyl-5,5-diphenylimidazolidin-4-ones (8a,b) (general procedure).** A mixture of **1a,b** (0.5 mmol) and triethylamine (1.52 g, 1.5 mmol) was added with vigorous stirring to a solution of 1,1-diphenylacetyl chloride (2.3 g, 1 mmol) in dry benzene (20 mL) at 60 °C under argon, during which the temperature of the reaction mixture increased to 80 °C. Then the reaction mixture was refluxed for 5 min and cooled to ~20 °C. Triethylamine hydrochloride that precipitated was filtered off and washed with benzene (3×30 mL). The solvent was distilled off *in vacuo*. Compounds **3a,b** and **8a,b** were isolated by column chromatography on SiO_2 (60 F_{254} 0.063—0.200 mm, Merck) using a hexane—ethyl acetate mixture as the eluent with a gradient from 10 : 1 to 6 : 1. Compounds **3a** and **8a,b** were recrystallized from a hexane—ethyl acetate mixture (1 : 1). *N*-Ethyl-*N*-(1-ethyl-4-oxo-3,3-diphenylazetidin-2-yl)-2,2-diphenylacetamide (**3a**), 1,3-diethyl-5,5-diphenylimidazolidin-4-one (**8a**), *N*-propyl-*N*-(1-propyl-4-oxo-3,3-diphenylazetidin-2-yl)-2,2-diphenylacetamide (**3b**), and 1,3-dipropyl-5,5-diphenylimidazolidin-4-one (**8b**) were obtained in yields of 1 g (41%), 0.3 g (21%), 0.83 g (32%), and 0.13 g (8%), respectively.

Synthesis of 1,3-dialkyl-5-arylimidazolidin-4-ones (8a,c—f) (general procedure). A precooled solution of the corresponding arylacetyl chloride **7** (0.5 mmol) in dry diethyl ether (10 mL) was added dropwise with continuous stirring to a solution of 1,2-dialkyldiaziridines **1a,b** (0.5 mmol) and triethylamine (1.52 g, 1.5 mmol) in dry diethyl ether cooled to −40—−30 °C under argon for 10 min. The reaction mixture was kept at −30 °C for 1 h, then slowly warmed to ~20 °C, and kept for 16 h. Triethylamine hydrochloride that precipitated was filtered off and washed with diethyl ether (3×40 mL). The solvent was distilled off *in vacuo*. Compounds **8a,c—f** were isolated by column chromatography on SiO_2 (60 F_{254} 0.063—0.200 mm, Merck) using a 1 : 1 hexane—ethyl acetate mixture (for **8c—f**) and a 4 : 1 hexane—ethyl acetate mixture (for **8a**) as the eluent. Compound **8a** was recrystallized from a hexane—ethyl acetate mixture (1 : 1). 1,3-Diethyl-5,5-diphenylimidazolidin-4-one (**8a**), 1,3-diethyl-5-(4-methylphenyl)imidazolidin-4-one (**8c**), 5-(4-chlorophenyl)-1,3-diethylimidazolidin-4-one (**8d**), 5-(4-bromophenyl)-1,3-di(*n*-propyl)imidazolidin-4-one (**8e**), and 1,3-diethyl-5-(2-nitrophenyl)imidazolidin-4-one (**8f**) were obtained in yields

Table 2. Yields and selected physicochemical characteristics of the compounds synthesized

Compound	Yield (%)	M.p./°C	R_f [a]	Found / Calculated (%)				Molecular formula
				C	H	N	Hal	
3a	41	150—151	0.70	81.27 81.12	6.52 6.60	5.44 5.73		$C_{33}H_{32}N_2O_2$
3b	32	Oil	0.71	81.42 81.36	6.85 7.02	5.39 5.42		$C_{35}H_{36}N_2O_2$
8a	65 21[b]	127—128	0.45	77.73 77.52	7.41 7.53	9.21 9.52		$C_{19}H_{22}N_2O$
8b	8	116—118	0.50	78.68 78.22	7.97 8.13	8.35 8.69		$C_{21}H_{26}N_2O$
8c	40	Oil	0.40	72.42 72.38	8.87 8.68	12.20 12.06		$C_{14}H_{20}N_2O$
8d	57	Oil	0.45	67.53 67.78	6.89 6.78	10.89 11.08	13.76 14.03	$C_{13}H_{17}ClN_2O$
8e	46	Oil	0.47	55.61 55.39	6.32 6.51	8.22 8.61	24.07 24.57	$C_{15}H_{21}BrN_2O$
8f	51	Oil	0.42	59.72 59.30	6.19 6.51	16.09 15.96		$C_{13}H_{17}N_3O_3$
8g	23	Oil	0.43	64.48 64.16	7.91 7.54	9.76 9.98	12.09 12.63	$C_{15}H_{21}ClN_2O$
8h	29	Oil	0.46	52.89 52.54	5.82 5.77	9.24 9.43	16.47 26.89	$C_{13}H_{17}BrN_2O$
10a	33	26—28	0.26	58.69 58.67	8.91 8.75	15.16 15.20		$C_9H_{16}N_2O_2$
10b	36	Oil	0.29	62.31 62.24	9.57 9.50	13.15 13.20		$C_{11}H_{20}N_2O_2$
10c	44	110—111	0.34	74.99 74.97	7.25 7.19	8.20 8.33		$C_{21}H_{24}N_2O_2$
10d	32	146—147	0.74	51.22 51.04	4.61 4.49	5.48 5.67	32.09 32.34	$C_{21}H_{22}Br_2N_2O_2$
10e	31	Oil	0.68	63.81 63.74	6.17 6.05	6.33 6.46	16.12 16.36	$C_{23}H_{26}Cl_2N_2O_2$
10f	37	113—114	0.75	67.79 57.68	5.26 5.07	6.26 6.41	14.45 14.63	$C_{21}H_{22}Cl_2N_2O_4$

[a] A 1 : 1 hexane—ethyl acetate mixture as the eluent.
[b] The compound was synthesized according to Scheme 4.

of 0.95 g (65%), 0.46 g (40%), 0.72 g (57%), 0.75 g (46%), and 0.67 g (51%), respectively.

Synthesis of 3,5-diacetyl-3,5-diazahept-1-enes (10a—c) (general procedure). A precooled solution of acetyl chloride (1.2 g, 1.5 mmol) in dry diethyl ether (10 mL) was added dropwise with continuous stirring to a solution of 1,2-dialkyldiaziridine **1a—c** (0.5 mmol) and triethylamine (2 g, 2 mmol) in dry diethyl ether cooled to −50—−45 °C under argon for 10 min. The reaction mixture was kept at this temperature for 2 h, then slowly warmed to room temperature, and kept for 16 h. Triethylamine hydrochloride that precipitated was filtered off and washed with diethyl ether (3×40 mL). The solvent was distilled off *in vacuo*. Compounds **10a—c** were isolated by column chromatography on SiO_2 (60 F_{254} 0.063—0.200 mm, Merck) using a 1 : 1 hexane—ethyl acetate mixture (for **10b**), a 3 : 2 hexane—ethyl acetate mixture (for **10c**), and a 15 : 1 diethyl ether—acetone mixture (for **10a**) as the eluent. Compound **10c** was recrystallized from a hexane—ethyl acetate mixture (1 : 1). 3,5-Diacetyl-3,5-diazahept-1-ene (**10a**), (*E*)-4,6-diacetyl-4,6-diazanon-2-ene (**10b**), and (*E*)-3,5-diacetyl-1,7-diphenyl-3,5-diazahept-1-ene (**10c**) were obtained in yields of 0.30 g (33%), 0.38 g (36%), and 0.74 g (44%), respectively.

3,5-Di(arylacetyl)-3,5-diazahept-1-enes (10d,e) and 1,3-dialkyl-5-arylimidazolidin-4-ones (8g,h). A mixture of diaziridine **1a,b** (0.5 mmol) and triethylamine (1.52 g, 1.5 mmol) was added with vigorous stirring to a solution of the corresponding arylacetyl chloride (1 mmol) in dry benzene (20 mL) at 60 °C under argon, during which the temperature of the reaction mixture increased to 80 °C. Then the reaction mixture was refluxed for 10 min and cooled to room temperature. Triethylamine hydrochloride that precipitated was filtered off and washed with benzene (3×30 mL). The solvent was distilled off *in vacuo*. Compounds **10d,e** and **8g,h** were isolated by column chromatography on SiO_2 (60 F_{254} 0.063-0.200 mm, Merck) using a 7 : 3 hex-

Table 3. IR, ^{1}H and ^{13}C NMR spectroscopic data for compounds **3** and **8**

Compound	IR, ν/cm^{-1}	^{1}H NMR	^{13}C NMR
		δ, *J*/Hz ($CDCl_3$)	
3a	476, 528, 612, 704, 724, 748, 776, 1032, 1056, 1084, 1112, 1176, 1212, 1224, 1320, 1352, 1392, 1416, 1456, 1496, 1600, 1648, 1764, 2940, 2980, 3028, 3060	1.02 (t, 3 H, $N_{cyclo}CH_2\underline{Me}$, ^{3}J = 7.22); 1.18 (t, 3 H, $NCH_2\underline{Me}$, ^{3}J = 7.22); 2.73 (m, 2 H, $N_{cyclo}C\underline{H}_2Me$); 2.96, 3.64 (both d.sext, 2 H, $NC\underline{H}_2Me$, ^{3}J = 7.22, J = 13.78, Δδ = 168.69); 5.13 (s, 1 H, PhC<u>H</u>CO); 6.84 (s, 1 H, NCHN); 6.87—7.84 (m, 20 H, 4 Ph)	12.14 (Me); 16.04 (Me); 36.77 ($N_{cyclo}CH_2$); 36.91 (NCH_2); 55.25 (Ph<u>C</u>HPh); 70.03 (NCHN); 71.99 (C_{cyclo}); 126.97, 127.08, 127.18, 127.21, 127.50, 128.59, 128.62, 128.65, 128.70, 128.77, 129.28 (<u>C</u>H(Ph)); 137.70, 138.23, 139.57, 140.09 (<u>C</u>$_{ipso}$(Ph)); 169.91 (CO_{cyclo}); 174.40 (CO)
3b	634, 710, 745, 771, 903, 1038, 1059, 1076, 1110, 1180, 1197, 1323, 1355, 1379, 1444, 1468, 1483, 1697, 2763, 2950, 2996, 3061	0.89 (t, 3 H, $N_{cyclo}CH_2CH_2\underline{Me}$, ^{3}J = 7.23); 0.92 (t, 3 H, $NCH_2CH_2\underline{Me}$, ^{3}J = 7.23); 1.53 (m, 4 H, 2 C<u>H</u>$_2$Me); 2.40—2.87 (m, 3 H, $N_{cyclo}CH_2$, NC<u>H</u>$_a$H$_b$); 3.47 (m, 1 H, NCH$_a$<u>H</u>$_b$); 5.00 (s, 1 H, PhC<u>H</u>CO); 6.80 (s, 1 H, NCHN); 6.83—7.87 (m, 20 H, 4 Ph)	11.38 (Me); 14.74 (Me); 22.97 (<u>C</u>H$_2$Me); 24.04 (<u>C</u>H$_2$Me); 35.32 ($N_{cyclo}CH_2$); 35.55 (NCH_2); 54.06 (PhCHPh); 68.87 (NCHN); 69.91 (C_{cyclo}); 126.86, 127.02, 127.09, 127.20, 127.49, 128.55, 128.62, 128.64, 128.69, 128.77, 129.29 (<u>C</u>H(Ph)); 137.65, 138.24, 139.52, 139.98 (<u>C</u>$_{ipso}$ (Ph)); 169.91 (CO_{cyclo}); 173.99 (CO)
8a	632, 704, 756, 772, 904, 1052, 1172, 1196, 1312, 1448, 1700, 2764, 2820, 2950, 3056, 3400	1.15 (t, 3 H, $CONCH_2\underline{Me}$, ^{3}J = 7.3); 1.30 (t, 3 H, $CPh_2NCH_2\underline{Me}$, ^{3}J = 7.3); 2.18 (q, 2 H, $CPh_2NC\underline{H}_2Me$, ^{2}J = 14.5, ^{3}J = 7.3); 3.57 (q, 2 H, $CONC\underline{H}_2Me$, ^{2}J = 14.5, ^{3}J = 7.3); 4.15 (s, 2 H, NCH_2N); 7.26 (m, 10 H, 2 Ph)	13.05 ($CPh_2NCH_2\underline{Me}$); 13.19 ($CONCH_2\underline{Me}$); 36.61 ($CON\underline{C}H_2Me$); 42.88 ($CPh_2N\underline{C}H_2Me$); 65.12 (NCH_2N); 127.53, 127.88, 128.53, 128.73, 129.07, 139.15 (Ph); 172.39 (CO)
8b	639, 765, 832, 900, 980, 1016 1022, 1117, 1174, 1187, 1300, 1337, 1449, 1511, 1650, 1715, 1763, 2929, 2954, 3364	0.96 (t, 3 H, $CONCH_2CH_2\underline{Me}$, ^{3}J = 7.32); 1.04 (t, 3 H, $CHNCH_2CH_2\underline{Me}$, ^{3}J = 7.32); 1.42—1.58 (m, 4 H, 2 C<u>H</u>$_2$Me); 2.50 (m, 2 H, $CHNC\underline{H}_2CH_2Me$); 3.37 (t, 2 H, $CONC\underline{H}_2CH_2Me$, ^{3}J = 7.32); 4.13 (s, 2 H, NCH_2N); 7.24 (m, 10 H, 2 Ph)	12.64 (Me); 16.96 (Me); 24.14 (<u>C</u>H$_2$Me); 25.39 (<u>C</u>H$_2$Me); 37.02 (NCH_2); 36.52 (NCH_2); 64.75 (NCH_2N); 127.56, 127.74, 128.04, 128.52, 128.97, 138.88 (Ph); 171.91 (CO)
8c	804, 844, 1020, 1116, 1160, 1192, 1308, 1344, 1452, 1516, 1652, 1712, 1764, 2932, 2972, 3325	1.06 (t, 3 H, $CONCH_2\underline{Me}$, ^{3}J = 7.5); 1.18 (t, 3 H, $CHNCH_2\underline{Me}$, ^{3}J = 7.5); 2.33 (s, 3 H, Me); 2.50 (m, 1 H, CHNC<u>H</u>(H)Me); 2.76 (m, 1 H, CONCH(<u>H</u>)Me); 3.41 (q, 2 H, $CHNC\underline{H}_2Me$, ^{2}J = 13.8, ^{3}J = 7.5); 3.93 (m, 2 H, NC<u>H</u>$_a$(H$_b$)N, COCH$_c$N); 4.51 (d, 1 H, NCH$_a$(<u>H</u>$_b$)N, ^{2}J = 5.0); 7.12, 7.30 (both d, 2 H each, Ar, ^{3}J = 7.5)	12.83 (Me); 12.86 (Me); 21.16 (Ar<u>Me</u>); 36.23 ($CON\underline{C}H_2Me$); 47.16 ($CHN\underline{C}H_2Me$); 67.37 (NCH_2N); 69.89 (<u>C</u>HAr); 128.16, 129.13, 134.67, 137.58 (Ar); 171.15 (CO)
8d	664, 764, 808, 844, 1016, 1092, 1216, 1312, 1456, 1492, 1576, 1648, 1716, 1772, 2936, 2976, 3300	1.05 (t, 3 H, $CONCH_2\underline{Me}$, ^{3}J = 7.3); 1.17 (t, 3 H, $CHNCH_2\underline{Me}$, ^{3}J = 7.3); 2.52 (m, 1 H, CHNC<u>H</u>(H)Me); 2.74 (m, 1 H, CONCH(<u>H</u>)Me); 3.39 (q, 2 H, $CHNC\underline{H}_2Me$, ^{2}J = 15.5, ^{3}J = 7.3); 3.93 (d.d, 1 H, NC<u>H</u>$_a$(H$_b$)N, ^{2}J = 4.4, $^{4}J_{H_b,H_c}$ = 2.2); 3.96 (s, 1 H, COC<u>H</u>$_c$N); 4.50 (d, 1 H, NCH$_a$(<u>H</u>$_b$)N, ^{2}J = 4.4); 7.30, 7.37 (both d, 2 H each, Ar, ^{3}J = 8.8)	12.81 (Me); 12.92 (Me); 36.27 ($CON\underline{C}H_2Me$); 47.29 ($CHN\underline{C}H_2Me$); 67.30 (NCH_2N); 69.35 (<u>C</u>HAr); 128.48, 129.41, 133.62, 136.35 (Ar); 170.34 (CO)

(*to be continued*)

Table 3 (*continued*)

Compound	IR, ν/cm^{-1}	^{1}H NMR	^{13}C NMR
		δ, *J*/Hz ($CDCl_3$)	
8e	630, 765, 815, 913, 999, 1010, 1084, 1213, 1326, 1358, 1439, 1546, 1580, 1699, 1797, 2873, 2930, 2975	0.89 (t, 3 H, $CONCH_2CH_2\underline{Me}$, ^3J = 7.5); 0.93 (t, 3 H, $CHNCH_2CH_2\underline{Me}$, ^3J = 7.5); 1.47 (m, 2 H, $CONCH_2C\underline{H}_2Me$); 1.60 (m, 2 H, $CHNCH_2C\underline{H}_2Me$); 2.55 (m, 2 H, $CHNC\underline{H}_2CH_2Me$); 3.31 (t, 2 H, $CONC\underline{H}_2CH_2Me$, ^3J = 7.5); 3.95 (d, 1 H, $NCH_a(\underline{H}_b)N$, ^2J = 2.5); 3.98 (s, 1 H, $CO\underline{CH}_cN$); 4.51 (d, 1 H, $NC\underline{H}_a(H_b)N$, ^2J = 2.5); 7.33, 7.49 (both d, 2 H each, Ar, ^3J = 10.0)	12.29 (Me); 11.64 (Me); 20.90 ($CONCH_2\underline{C}H_2Me$); 21.06 ($CHNCH_2\underline{C}H_2Me$); 43.16 ($CON\underline{C}H_2CH_2Me$) 55.31 ($CHN\underline{C}H_2CH_2Me$); 68.18 ($NCH_2N$); 69.62 (<u>C</u>HAr); 121.86, 129.74, 131.46, 136.95 (Ar); 170.58 (CO)
8f	668, 728, 744, 788, 1160, 1256, 1308, 1356, 1456, 1528, 1648, 1708, 2820, 2936, 2976	1.06 (t, 3 H, $CONCH_2\underline{Me}$, ^3J = 7.5); 1.19 (t, 3 H, $CHNCH_2\underline{Me}$, ^3J = 7.5); 2.71 (m, 2 H, $CONC\underline{H}_2Me$); 3.40 (q, 2 H, $CHNC\underline{H}_2Me$, ^2J = 14.5, ^3J = 7.5); 4.00 (d.d, 1 H, $NC\underline{H}_a(H_b)N$, ^2J = 4.4, $^4J_{H_b,H_c}$ = 2.2); 4.06 (s, 1 H, $COC\underline{H}_cN$); 4.52 (d, 1 H, $NCH_a(\underline{H}_b)N$, ^2J = 4.4); 7.39—7.72 (m, 4 H, Ar)	12.69 (Me); 12.98 (Me); 36.35 ($CON\underline{C}H_2Me$); 48.11 ($CHN\underline{C}H_2Me$); 66.43 (NCH_2N); 67.61 (<u>C</u>HAr); 124.78, 128.61, 130.18, 132.50, 133.35, 150.13 (Ar); 169.13 (CO)
8g	492, 572, 736, 804, 844, 1012, 1072, 1160,1312, 1488, 1540, 1592, 1652, 1704, 1768, 1908, 2812, 2876, 2936, 2976, 3064	1.05 (t, 3 H, $CONCH_2\underline{Me}$, ^3J = 7.22); 1.17 (t, 3 H, $CHNCH_2\underline{Me}$, ^3J = 7.22); 2.53 (m, 1 H, $CHNC\underline{H}(H)Me$); 2.74 (m, 1 H, $CONCH(\underline{H})Me$); 3.39 (q, 2 H, $CHNC\underline{H}_2Me$, ^2J = 14.4, ^3J = 7.22); 3.94 (br.s, 3 H, $NC\underline{H}_a(H_b)N$, $COCH_cN$); 4.51 (d, 1 H, $NCH_a(\underline{H}_b)N$, ^2J = 2.63); 7.32, 7.47 (both d, 2 H each, Ar, ^3J = 8.53)	12.86 (Me); 12.98 (Me); 36.32 ($CON\underline{C}H_2Me$); 47.65 ($CHN\underline{C}H_2Me$); 67.41 (NCH_2N); 69.33 (<u>C</u>HAr); 128.52, 129.51, 133.74, 136.52 (Ar); 170.44 (CO)
8h	665, 674, 754, 812, 845, 989, 1023, 1110, 1223, 1309, 1476, 1499, 1581, 1648, 1706, 1777, 2942, 2979, 3310	0.86 (t, 3 H, $CONCH_2CH_2\underline{Me}$, ^3J = 7.22); 0.93 (t, 3 H, $CHNCH_2CH_2\underline{Me}$, ^3J = 7.22); 1.42 (m, 2 H, $CONCH_2C\underline{H}_2Me$); 1.57 (m, 2 H, $CHNCH_2C\underline{H}_2Me$); 2.52 (m, 2 H, $CHNC\underline{H}_2CH_2Me$); 3.28 (t, 2 H, $CONC\underline{H}_2CH_2Me$, ^3J = 7.22); 3.91 (d, 1 H, $NCH_a(\underline{H}_b)N$, ^2J = 3.90); 3.95 (s, 1 H, $CO\underline{CH}_cN$); 4.47 (d, 1 H, $NC\underline{H}_a(H_b)N$, ^2J = 3.90); 7.30, 7.37 (both d, 2 H each, Ar, ^3J = 8.5)	11.17 (Me); 11.29 (Me); 20.79 ($CONCH_2\underline{C}H_2Me$); 20.96 ($CHNCH_2\underline{C}H_2Me$); 43.05 ($CON\underline{C}H_2CH_2Me$) 55.19 ($CHN\underline{C}H_2CH_2Me$); 68.11 ($NCH_2N$); 69.48 (<u>C</u>HAr); 128.41, 129.35, 133.52, 136.49 (Ar); 170.62 (CO)

Table 4. IR, ^{1}H and ^{13}C NMR spectroscopic data for compounds **10**

Compound	IR, ν/cm^{-1}	^{1}H NMR	^{13}C NMR
		δ, *J*/Hz ($CDCl_3$)	
10a[a]	628, 780, 800, 864, 912, 968, 996, 1036, 1104, 1164, 1204, 1240, 1288, 1340, 1368, 1392, 1424, 1460, 1632, 1668, 2924, 2976	1.18 (t, 3 H, $CH_2\underline{Me}$, ^3J = 7.4); 2.14 (s, 3 H, $CH_3CH_2NCO\underline{Me}$); 2.31 (s, 3 H, CH_2=$CHNCO\underline{Me}$); 3.19, 3.28 (10 : 1) (both q, 2 H, $C\underline{H}_2Me$, ^3J = 7.4); 4.40, 4.56 (10 : 1) (both d, 1 H, NCH_a=$C\underline{H}_aH_b$, $^3J_{H_a,H_a}$ = 9.6); 4.92 (d, 1 H, NCH_a=$CH_a\underline{H}_b$, $^3J_{H_a,H_b}$ = 15.0); 5.16, 5.46 (10 : 1) (both s, 2 H, NCH_2N); 6.66, 7.07 (10 : 1) (both dd, 1 H, CH=, $^3J_{H_a,H_a}$ = 9.6, $^3J_{H_a,H_b}$ = 15.0)	13.45 ($NCH_2\underline{Me}$); 21.66 ($CH_3CH_2NCO\underline{Me}$); 22.83 ($CH_2$=$CHNCO\underline{Me}$); 40.14 ($\underline{C}H_2Me$); 47.61 ($NCH_2N$); 96.82 (CH=$\underline{C}H_2$); 130.79 ($\underline{C}H$=$CH_2$); 171.03 ($CH_2CH_2N\underline{C}O$); 171.10 ($CH_2$=$CHN\underline{C}O$)

(*to be continued*)

Table 4 (*continued*)

Compound	IR, ν/cm^{-1}	^{1}H NMR	^{13}C NMR
		δ, J/Hz ($CDCl_3$)	
10b[b]	620, 664, 736, 796, 824, 896, 960, 1000, 1036, 1076, 1108, 1160, 1200, 1232, 1264, 1288, 1352, 1376, 1400, 1428, 1656, 1680, 2876, 2932, 2964	0.88 (t, 3 H, $CH_2\underline{Me}$, ^3J = 7.2); 1.59 (m, 2 H, $MeC\underline{H}_2CH_2$); 1.68 (d, 3 H, $CH{=}CH\underline{Me}$, ^3J = 6.6); 2.09 (s, 3 H, $CH_2CH_2NCO\underline{Me}$); 2.19 (s, 3 H, $CH{=}CHNCO\underline{Me}$); 3.11, 3.22 (8 : 1) (both q, 2 H, NCH_2, ^3J = 7.2); 5.06, 5.35 (8 : 1) (both s, 2 H, NCH_2N); 5.42 (m, 1 H, $NCH_a{=}C\underline{H}_bMe$); 6.14, 6.28 (8 : 1) (both d, 1 H, $NC\underline{H}_a{=}CH_bMe$, $^3J_{H_a,H_b}$ = 13.8)	11.39 ($CH_2\underline{Me}$); 16.16 ($CH\underline{Me}$); 21.49 ($CH_2CH_2NCO\underline{Me}$); 21.50 ($Me\underline{C}H_2CH_2$); 22.99 ($CH{=}CHNCO\underline{Me}$); 47.12 ($CH_2\underline{C}H_2N$); 49.00 ($NCH_2N$); 110.20 ($CH{=}\underline{C}HMe$); 125.87 ($\underline{C}H{=}CHMe$); 170.71 ($CH_2CH_2N\underline{C}O$); 171.01 ($CH{=}CHN\underline{C}O$)
10c[b]	572, 612, 692, 704, 752, 956, 996, 1032, 1076, 1164, 1200, 1240, 1268, 1336, 1396, 1424, 1492, 1576, 1600, 1636, 1640, 1668, 3000, 3024	2.01 (s, 3 H, $CH_2CH_2NCO\underline{Me}$); 2.38 (s, 3 H, $CH{=}CHNCO\underline{Me}$); 2.92 (t, 2 H, $C\underline{H}_2Ph$, ^3J = 7.3); 3.41, 3.56 (7 : 1) (both t, 2 H, $NC\underline{H}_2CH_2$, ^3J = 7.3); 5.12, 5.65 (7 : 1) (both s, 2 H, NCH_2N); 6.45 (d, 1 H, $NCH_a{=}C\underline{H}_bPh$, $^3J_{H_a,H_b}$ = 14.0); 6.90, 7.01 (7 : 1) (both d, 1 H, $NC\underline{H}_a{=}CH_bPh$, $^3J_{H_a,H_b}$ = 14.0); 7.21—7.36 (m, 10 H, Ph)	21.42 ($CH_2CH_2NCO\underline{Me}$); 22.68 ($CH{=}CHNCO\underline{Me}$); 34.95 ($\underline{C}H_2CH_2N$); 47.79 ($CH_2\underline{C}H_2N$); 49.75 ($NCH_2N$); 114.95 ($CH{=}\underline{C}HPh$); 126.31 ($\underline{C}H{=}CHPh$); 125.62, 125.81, 125.99, 126.70, 126.87, 128.47, 128.69, 128.74, 128.90, 129.02, 136.31, 138.10 (Ph); 170.99 ($CH_2CH_2N\underline{C}O$); 171.14 ($CH{=}CHN\underline{C}O$)
10d	488, 728, 776, 864, 1012, 1068, 1088, 1120,1208, 1268, 1344, 1360, 1432, 1492, 1632, 1648, 1676, 2920, 2980	1.13 (t, 3 H, $CH_2\underline{Me}$, ^3J = 7.22); 3.23 (q, 2 H, $C\underline{H}_2Me$, ^3J = 7.22) 3.64 (s, 2 H, $COC\underline{H}_2N(Et)$); 3.80 (s, 2 H, $COC\underline{H}_2N(CH{=}CH_2)$); 4.49 (d, 1 H, $NCH_a{=}C\underline{H}_aH_b$, $^3J_{H_a,H_a}$ = 9.2); 4.97 (d, 1 H, $NCH_a{=}CH_a\underline{H}_b$, $^3J_{H_a,H_b}$ = 14.4); 5.48 (s, 2 H, NCH_2N); 6.69 (dd, 1 H, CH=, $^3J_{H_a,H_a}$ = 9.2, $^3J_{H_a,H_b}$ = 14.4); 7.09, 7.45 (both m, 8 H, Ar)	13.91 ($NCH_2\underline{Me}$); 39.76 (CH_2); 39.95 (CH_2); 40.60 (CH_2); 49.55 (NCH_2N); 99.32 ($CH{=}\underline{C}H_2$); 130.62, 130.68, 131.63, 131.76, 131.83, 132.21, 132,45, 132.49 (Ar); 131.45 ($\underline{C}H{=}CH_2$); 170.92 ($CH_2CH_2N\underline{C}O$); 171.13 ($CH_2{=}CHN\underline{C}O$)
10e	634, 651, 745, 876, 898, 923, 976, 1065, 1090, 1112, 1134, 1167, 1234, 1265, 1298, 1300, 1354, 1391, 1418, 1432, 1658, 1695, 2866, 2923, 2954, 3028	0.86 (t, 3 H, $CH_2\underline{Me}$, ^3J = 7.3); 1.62 (m, 2 H, $MeC\underline{H}_2CH_2$); 1.72 (d, 3 H, $CH{=}CH\underline{Me}$, ^3J = 6.8); 3.27 (m, 2 H, NCH_2); 3.62 (s, 2 H, $COC\underline{H}_2N(Pr)$); 3.77 (s, 2 H, $COC\underline{H}_2N(CH{=}CHMe)$); 5.03 (both s, 2 H, NCH_2N); 5.47 (m, 1 H, $NCH_a{=}C\underline{H}_bMe$); 6.25 (d, 1 H, $NC\underline{H}_a{=}CH_bMe$, $^3J_{H_a,H_b}$ = 13.8); 7.15 (m, 8 H, Ar)	12.56 ($CH_2\underline{Me}$); 17.20 ($CH\underline{Me}$); 22.53 ($Me\underline{C}H_2CH_2$); 39.79 (CH_2); 39.97 (CH_2); 41.01 (CH_2); 50.0 (NCH_2N); 111.98 ($CH{=}\underline{C}HMe$); 131.67($\underline{C}H{=}CHMe$); 130.87, 130.89, 131.21, 131.42, 131.78, 132.08, 132,76, 132.57 (Ar); 171.13 ($CH_2CH_2N\underline{C}O$); 171.24 ($CH_2{=}CHN\underline{C}O$)
10f	508, 668, 800, 832, 1060, 1104, 1136, 1228, 1240, 1288, 1328, 1428, 1492, 1584, 1596, 1640, 1672, 1704, 2924, 2972	1.21 (t, 3 H, $CH_2\underline{Me}$, ^3J = 7.35); 3.26 (q, 2 H, $C\underline{H}_2Me$, ^3J = 7.35); 4.56 (d, 1 H, $NCH_a{=}C\underline{H}_aH_b$, $^3J_{H_a,H_a}$ = 8.8); 4.68 (both s, 1 H, $OC\underline{H}_2CON(Et)$); 4.82 (s, 1 H, $OC\underline{H}_2CON(CH{=}CH_2)$); 5.00 (d, 1 H, $NCH_a{=}CH_a\underline{H}_b$, $^3J_{H_a,H_b}$ = 16.18); 5.49 (s, 2 H, NCH_2N); 6.63 (dd, 1 H, CH=, $^3J_{H_a,H_a}$ = 8.8, $^3J_{H_a,H_b}$ = 16.18); 6.85, 7.23 (both m, 8 H, Ar)	13.98 ($NCH_2\underline{Me}$); 39.20 (CH_2); 49.92 (NCH_2N); 66.82 (OCH_2); 67.21 (OCH_2); , 100.40 ($CH{=}\underline{C}H_2$); 116.02, 116.06, (Ar); 126.60 (C_{ipso}); 126.99 ($\underline{C}H{=}CH_2$); 129.40, 129.52 (Ar); 156.15, 156.51 (OC_{ipso}); 167.87 ($CH_2CH_2N\underline{C}O$); 168.06 ($CH_2{=}CHN\underline{C}O$)

[a] ^{1}H and ^{13}C NMR ($CDCl_3$, −30 °C).
[b] ^{13}C NMR ($CDCl_3$, −30 °C).

ane—ethyl acetate mixture (for **10d**, **8g**) and 1 : 1 hexane—ethyl acetate mixture (for **10e**, **8h**) as the eluent. Compound **10d** was recrystallized from a hexane—ethyl acetate mixture (20 : 1). 3,5-Di[(4-bromophenyl)acetyl]-3,5-diazahept-1-ene (**10d**), 5-(4-bromophenyl)-1,3-diethylimidazolidin-4-one (**8g**), (*E*)-4,6-di[(4-chlorophenyl)acetyl]-4,6-diazanon-2-ene (**10e**),

and 5-(4-chlorophenyl)-1,3-dipropylimidazolidin-4-one (**8h**) were obtained in yields of 0.79 g (32%), 0.34 g (23%), 0.67 g (31%), and 0.4 g (29%), respectively.

3,5-Di[(4-chlorophenoxy)acetyl]-3,5-diazahept-1-ene (10f). A mixture of compound **1a** (0.5 mmol) and triethylamine (1.52 g, 1.5 mmol) was added with vigorous stirring to a solution of 4-chlorophenoxyacetyl chloride (2.05 g, 1 mmol) in dry benzene (20 mL) at 60 °C under argon, during which the temperature of the reaction mixture increased to 80 °C. Then the reaction mixture was refluxed for 10 min and cooled to ~20 °C. Triethylamine hydrochloride that precipitated was filtered off and washed with benzene (3×30 mL). The solvent was distilled off *in vacuo*. The residue was extracted with hot hexane (3×50 mL) and triturated with cooling to 0 °C. The precipitate that formed was filtered off and dried in air. 2-(4-Chlorophenoxy)-*N*-[3-(4-chlorophenoxy)-1-ethyl-4-oxoazetidin-2-yl]-*N*-ethylacetamide (**3f**) was obtained in a yield of 0.9 g (~85% purity). ^{1}H NMR ($CDCl_3$), δ: 1.13 (t, 3 H, $N_{cyclo}CH_2\underline{Me}$, ^3J = 7.3 Hz); 1.19 (t, 3 H, $NCH_2\underline{Me}$, ^3J = 7.3 Hz); 2.31 and 3.45 (both d.sext, 4 H, $NC\underline{H}_2Me$ + $N_{cyclo}C\underline{H}_2Me$); 4.41 (s, 2 H, $C\underline{H}_2CO$); 4.72 and 4.89 (both d, 1 H each, NCHN and $(COC\underline{H})_{cyclo}$, ^3J = 4.5 Hz); 6.70—7.42 (m, 8 H, 2 Ar). An attempt to purify compound **3f** by chromatography on SiO_2 (hexane—ethyl acetate, 7 : 3, as the eluent) led to the cleavage of the cyclic system to give compound **10f** in a yield of 0.80 g (37%).

X-ray diffraction study. Crystals of **3a** ($C_{33}H_{32}N_2O_2$) are monoclinic, at 120 K: a = 18.467(3), b = 8.600(1), c = 18.035(3) Å, β = 115.033(4)°, V = 2595.1(7) Å^3, d_{calc} = 1.251 g cm^{-1}, space group $P2_1/n$, Z = 4. The intensities of 12248 reflections were measured on an automated Smart 1000 CCD diffractometer at 110 K (Mo-Kα radiation, graphite monochromator, ω scanning technique, $2\theta_{max}$ = 54°), of which 6227 observed reflections (R_{int} = 0.0857) were used in calculations. The structure was solved by direct methods and refined by the full-matrix least-squares method with anisotropic/isotropic displacement parameters against F^2. The hydrogen atoms were revealed from difference electron density syntheses and refined using a riding model. The final reliability factors were as follows: wR_2 = 0.0784, GOOF = 0.868 for all reflections (R_1 = 0.0538 was calculated for 1982 reflections with $I > 2\sigma(I)$) using the SHELXTL PLUS program package.[17]

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