Reactions of aroylpyruvic acids with *S*-methylisothiosemicarbazide hydroiodide and studies of the crystal structures of the reaction products

Z. G. Aliev,[a] L. O. Atovmyan,[a] Yu. S. Andreichikov,[b] S. V. Kol'tsova,[b] and D. D. Nekrasov[b]*

[a]Institute of Chemical Physics in Chernogolovka, Russian Academy of Sciences, 142432 Chernogolovka, Moscow Region, Russian Federation. Fax: +7 (096) 515 3588. E-mail: aliev@icp.ac.ru
[b]Institute of Technical Chemistry, Ural Branch of the Russian Academy of Sciences, 13a ul. Lenina, 614600 Perm', Russian Federation. Fax: +7 (342 2) 32 5018

The reactions of *p*-chlorobenzoyl- and benzoylpyruvic acids with *S*-methylisothiosemicarbazide hydroiodide gave 3-methylthio-6-(*p*-chlorophenacyl)-2,5-dihydro-1,2,4-triazin-5-one and 6-phenacyl-2,3,4,5-tetrahydro-1,2,4-triazine-3,5-dione, respectively. The molecular and crystal structures of the compounds synthesized were studied by X-ray diffraction analysis.

Key words: aroylpyruvic acids, *S*-methylisothiosemicarbazide hydroiodide, 3-methylthio-6-(*p*-chlorophenacyl)-2,5-dihydro-1,2,4-triazin-5-one, 6-phenacyl-2,3,4,5-tetrahydro-1,2,4-triazine-3,5-dione, molecular and crystal structure.

It is known[1] that aroylpyruvic acids react with hydrazine as β-dicarbonyl systems to form pyrazoles. However, the reactions of these acids with urea, ethylenediamine, and *o*-phenylenediamine occur at the oxalyl group to give derivatives of imidazole,[2] piperazine,[3] and quinoxaline,[4] respectively. The reactions of heterocyclic 1,2-diamines with aroylpyruvic acids afford fused 1,4-diazine and 1,2,4-triazine systems.[5–9]

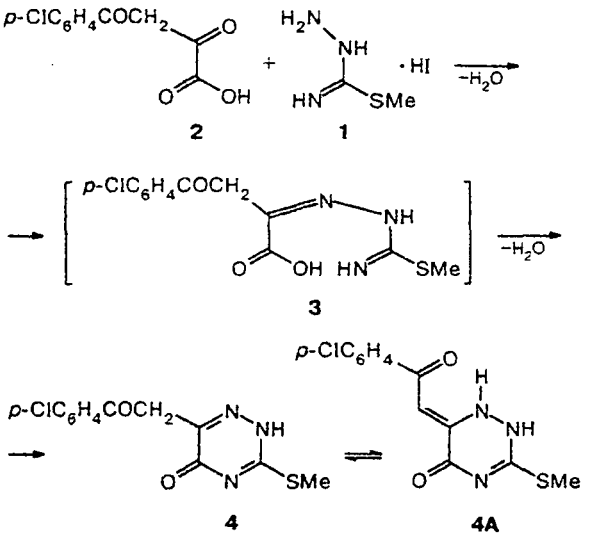
S-Methylisothiosemicarbazide hydroiodide (**1**) can be involved in the reaction with *p*-chlorobenzoylpyruvic acid (**2**) as a hydrazine, amidine, or 1,2-diamine. Boiling of a mixture of these reagents in EtOH gave a colorless crystalline compound in a yield of 59%. It was difficult to establish the structure of the resulting compound by conventional physicochemical methods because putative isomeric products, which can be formed in this reaction, should have similar spectral characteristics, and mass-spectrometric fragmentation would not afford characteristic ions, which allow one to differentiate the structures of the suggested regioisomers.

X-ray diffraction analysis demonstrated that the crystalline reaction product (Scheme 1) is 3-methylthio-6-(*p*-chlorophenacyl)-2,5-dihydro-1,2,4-triazin-5-one (**4**). According to the data of ^{1}H NMR (DMSO-d_6) spectroscopy of this compound, the enamine form **4A** (~20%) was present along with the imino form **4**.

Apparently, the formation of compound **4** involves two stages: the attack of the primary amino group of reagent **1** on the α-carbonyl group of acid **2** and the intramolecular cyclization of intermediate **3**.

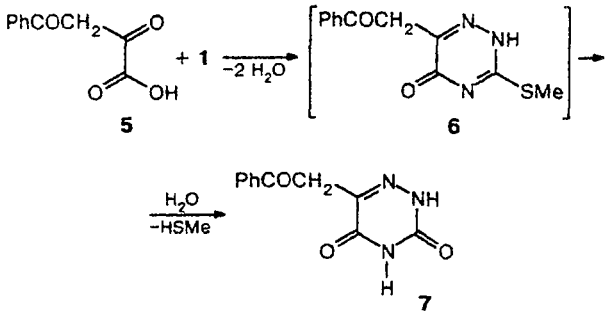
Unexpectedly, it was found that an analogous reaction of benzoylpyruvic acid (**5**) with hydroiodide **1** afforded a product whose spectral characteristics differ substantially from those of compound **4**. Thus its IR spectrum has a band at 1740 cm^{-1}, which corresponds to the carbonyl group, and the ^{1}H NMR spectrum shows a signal of the second NH group at δ 12.45. In addition, the signal of the S—Me group is absent in the ^{1}H NMR spectrum. These data suggest that 3-methylthio-6-phenacyl-2,5-dihydro-1,2,4-triazin-5-one (**6**), which was initially formed in the reaction (Scheme 2), underwent hydrolysis to give 6-phenacyl-2,3,4,5-

Scheme 1



Translated from *Izvestiya Akademii Nauk. Seriya Khimicheskaya*, No. 4, pp. 704—708, April, 1998.

Scheme 2



tetrahydro-1,2,4-triazine-3,5-dione (**7**). Actually, X-ray diffraction study confirmed that the reaction product has structure **7**.

The formation of different products in the above-mentioned reactions is, apparently, associated with the difference in the solubility of the compounds, which were formed in the course of the reaction, in EtOH. Product **4** is insoluble and precipitated, while soluble product **6** underwent hydrolysis to form compound **7** owing to elimination of the water. Hydroiodic acid activated this process.

The overall view of molecule **4** is shown in Fig. 1. The bond lengths and bond angles are given in Tables 1 and 2, respectively. The characteristic feature of the molecular structure is pronounced localization of the N(5)=C(9) and N(6)=C(11) double bonds in the planar triazine ring (1.298 and 1.294 Å, respectively). The

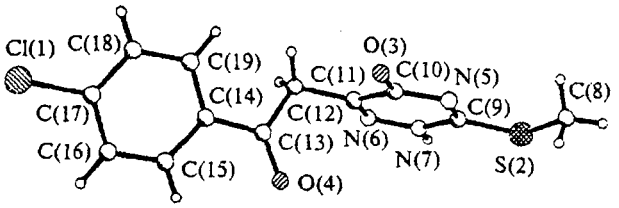


Fig. 1. Structure of molecule **4**.

remaining interatomic distances in the ring agree well with the lengths of the single bonds. The SMe fragment is located in the plane of the heterocycle. The Me group is in the *cis* position with respect to the carbonyl group in the triazine heterocycle. The carbonyl group of the chlorobenzoyl fragment deviates noticeably from the plane of the benzene ring (the O(4)C(13)C(14)C(15) torsion angle is −25.8°). The angle between the planes of the benzene and triazine rings is 116.5°. The conformation of the molecule as a whole is characterized by the following torsion angles: N(6)C(11)C(12)C(13), 101.8°; C(11)C(12)C(13)C(14), 164.7°; and C(12)C(13)C(14)C(15), 152.5°. In the crystal, molecules **4** are grouped about the screw axis and are linked in infinite chains through hydrogen bonds. An intermolecular hydrogen bond occurs between the O(3) and N(7) atoms. The parameters of the hydrogen bond (N(7)...O(3), 2.81 Å; H(7)...O(3), 1.93 Å; and N—H—O angle 172.2°) indicate that it is rather strong. Apparently, it is the formation of this bond that causes the substantial elongation of the C(10)=O(3) bond (1.237 Å) compared to the C(13)=O(4) bond (1.209 Å).

The character of the crystal packing and the intermolecular hydrogen bond suggest that the tautomeric transformation can occur according to the relay mechanism under the action of external agents.

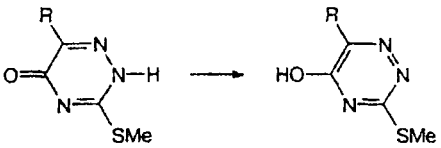


Table 1. Bond lengths (*d*) in molecule **4**

Bond	*d*/Å	Bond	*d*/Å
Cl(1)—C(17)	1.733(3)	C(10)—C(11)	1.473(4)
S(2)—C(9)	1.754(3)	C(11)—C(12)	1.487(4)
S(2)—C(8)	1.788(3)	C(12)—C(13)	1.514(4)
O(3)—C(10)	1.237(3)	C(13)—C(14)	1.495(4)
O(4)—C(13)	1.209(4)	C(14)—C(19)	1.377(4)
N(5)—C(9)	1.298(4)	C(14)—C(15)	1.392(4)
N(5)—C(10)	1.360(3)	C(15)—C(16)	1.378(4)
N(6)—C(11)	1.294(4)	C(16)—C(17)	1.385(5)
N(6)—N(7)	1.352(3)	C(17)—C(18)	1.366(5)
N(7)—C(9)	1.349(3)	C(18)—C(19)	1.386(5)

Table 2. Bond angles (ω) in molecule **4**

Angle	ω/deg	Angle	ω/deg	Angle	ω/deg
C(9)—S(2)—C(8)	100.7(1)	N(5)—C(10)—C(11)	118.1(2)	C(19)—C(14)—C(13)	123.2(3)
C(9)—N(5)—C(10)	117.4(2)	N(6)—C(11)—C(10)	121.6(2)	C(15)—C(14)—C(13)	117.9(3)
C(11)—N(6)—N(7)	116.7(2)	N(6)—C(11)—C(12)	118.8(2)	C(16)—C(15)—C(14)	121.3(3)
C(9)—N(7)—N(6)	122.4(2)	C(10)—C(11)—C(12)	119.5(2)	C(17)—C(16)—C(15)	118.4(3)
N(5)—C(9)—N(7)	123.7(2)	C(11)—C(12)—C(13)	113.3(2)	C(18)—C(17)—C(16)	121.3(3)
N(5)—C(9)—S(2)	122.5(2)	O(4)—C(13)—C(14)	120.6(3)	C(18)—C(17)—Cl(1)	119.1(3)
N(7)—C(9)—S(2)	113.8(2)	O(4)—C(13)—C(12)	121.9(3)	C(16)—C(17)—Cl(1)	119.5(3)
O(3)—C(10)—N(5)	121.4(3)	C(14)—C(13)—C(12)	117.5(2)	C(17)—C(18)—C(19)	119.7(3)
O(3)—C(10)—C(11)	120.4(2)	C(19)—C(14)—C(15)	118.9(3)	C(14)—C(19)—C(18)	120.4(3)

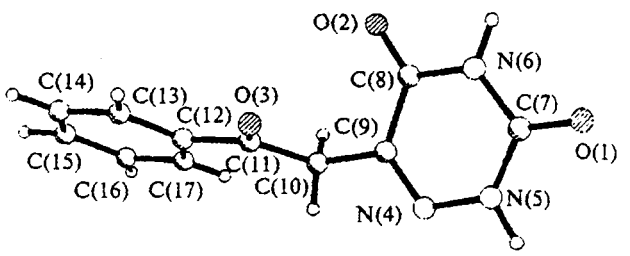


Fig. 2. Structure of molecule **7**.

We observed an analogous tautomeric transformation, which was accompanied by the electrochromic effect, in the solid phase in the case of 2-(*p*-dimethylaminophenyl)-4-chloroindandione-1,3.[10]

The overall view of molecule **7** is shown in Fig. 2. The bond lengths and bond angles are given in Tables 3 and 4, respectively. The virtually planar benzoyl and triazine fragments, which are in *trans* orientations with respect to the C(10)—C(11) bond, form a dihedral angle of 65.2°. The length of the localized N(4)=C(9) double bond in the ring is 1.292 Å. All distances and bond angles in the molecule have standard values and need no comments. Unlike compound **4** in which the carbonyl group of the aroyl fragment deviates substantially from the plane of the benzene ring, the benzoyl fragment in molecule **7** is virtually planar (the O(3)C(11)C(12)C(13) torsion angle is 3.8°). However, this has no effect on the electronic structures of the molecules, that is, the bond lengths in the benzoyl fragments of two molecules are equal within the experimental error, and the conjugation between the carbonyl group and the benzene ring is not pronounced.

Table 3. Bond lengths (*d*) in molecule **7**

Bond	*d*/Å	Bond	*d*/Å
O(1)—C(7)	1.212(2)	C(9)—C(10)	1.493(2)
O(2)—C(8)	1.225(2)	C(10)—C(11)	1.518(3)
O(3)—C(11)	1.220(2)	C(11)—C(12)	1.482(2)
N(4)—C(9)	1.292(2)	C(12)—C(17)	1.395(3)
N(4)—N(5)	1.352(2)	C(12)—C(13)	1.397(3)
N(5)—C(7)	1.362(3)	C(13)—C(14)	1.385(3)
N(6)—C(8)	1.363(2)	C(14)—C(15)	1.381(3)
N(6)—C(7)	1.386(2)	C(15)—C(16)	1.383(3)
C(8)—C(9)	1.470(3)	C(16)—C(17)	1.386(3)

In the crystal, molecules **7** are linked in centrosymmetrical dimers through the O(2)...H(6)—N(6) hydrogen bonds [*r*(O(2)...N(6)) = 2.89 Å and *r*(O(2)...H(6)) = 1.98 Å]. In addition, each associate is linked to adjacent associates, which are related to the initial one by twofold screw axes, through the O(3)...H(5)—N(5) hydrogen bond [*r*(O(3)...N(5)) = 2.84 Å and *r*(O(3)...H(5)) = 1.91 Å]. Therefore, in the crystal molecules are linked through hydrogen bonds to form infinite layers parallel to the *b* axis. The O(1) atom of the carbonyl group of the heterocycle is not involved in hydrogen bonding. This is also reflected in the bond lengths of the carbonyl groups, namely, of three C=O bonds, the C(7)=O(1) bond is the shortest (1.212 Å).

Generally, the five- and six-membered heterocycles that contain phenacyl substituents exist in the tautomeric phenacylidene form.[11,12] This is true for derivatives of 5-phenacylimidazolidine-2,4-dione,[11] 3-phenacyl-2-piperazinone,[12] 3-phenacyl-2-quinoxalone,[13,14] and 3-phenacylbenzo-1,4-oxazin-2-one[13,14] and is attributable to stabilization of the phenacylidene form through the intramolecular hydrogen bond between the O atom of the phenacyl group and the NH group of the heterocycle as well as to the presence of conjugation between the phenacylidene and heterocyclic fragments. However, in the crystals compounds **4** and **7** occur in the phenacyl (imine) form.

Experimental

The IR spectra were recorded on Specord M-80 and UR-20 instruments as Nujol mulls. The ^{1}H NMR spectra were measured on a Bruker WP-80-SV instrument in DMSO-d_6 with HMDS as the internal standard.

3-Methylthio-6-(*p*-chlorophenacyl)-2,5-dihydro-1,2,4-triazin-5-one (4). A solution of hydroiodide **1** (1.16 g, 5 mmol) and acid **2** (1.14 g, 5 mmol) was boiled in EtOH (20 mL) for 2 h, and then the reaction mixture was cooled to ~20 °C. The precipitate was filtered off and recrystallized from EtOH. The yield of compound **4** was 59%, m.p. 231—232 °C. Found (%): C, 48.9; H, 3.6; Cl, 11.9; N, 14.4; S, 11.2. $C_{12}H_{10}ClN_3O_2S$. Calculated (%): C, 48.7; H, 3.4; Cl, 12.0; N, 14.3; S, 11.0. IR, ν/cm^{-1}: 1596 (C=N); 1630 (C=O); 1688 (C=O); 3200, 3450 br (NH). ^{1}H NMR, δ: 4.23 (s, 2 H, CH_2); 6.37 (s, 1 H, CH); 7.42 and 7.13 (both dd, 4 H, C_6H_4); 10.42 (s, 1 H, NH). The signal of the SMe group is masked by the signal of the solvent. The ^{1}H NMR spectrum has also the signal of the isomeric enamine

Table 4. Bond angles (ω) in molecule **7**

Angle	ω/deg	Angle	ω/deg	Angle	ω/deg
C(9)—N(4)—N(5)	118.1(2)	N(6)—C(8)—C(9)	114.2(2)	C(17)—C(12)—C(13)	119.5(2)
N(4)—N(5)—C(7)	126.1(2)	N(4)—C(9)—C(8)	122.5(2)	C(17)—C(12)—C(11)	122.1(2)
C(8)—N(6)—C(7)	124.8(2)	N(4)—C(9)—C(10)	119.1(2)	C(13)—C(12)—C(11)	118.3(2)
O(1)—C(7)—N(5)	123.5(2)	C(8)—C(9)—C(10)	118.4(2)	C(14)—C(13)—C(12)	120.1(2)
O(1)—C(7)—N(6)	122.5(2)	C(9)—C(10)—C(11)	111.8(2)	C(15)—C(14)—C(13)	120.0(2)
N(5)—C(7)—N(6)	113.9(2)	O(3)—C(11)—C(12)	120.9(2)	C(14)—C(15)—C(16)	120.2(2)
O(2)—C(8)—N(6)	122.1(2)	O(3)—C(11)—C(10)	120.2(2)	C(15)—C(16)—C(17)	120.5(2)
O(2)—C(8)—C(9)	123.7(2)	C(12)—C(11)—C(10)	118.8(2)	C(16)—C(17)—C(12)	119.6(2)

form (**4A**) (δ 6.37, =CH); the ratio of the imine and enamino forms is 4 : 1.

6-Phenacyl-2,3,4,5-tetrahydro-1,2,4-triazine-3,5-dione (7). A solution of hydroiodide **1** (1.16 g, 5 mmol) and acid **5** (0.96 g, 5 mmol) was boiled in EtOH (20 mL) for 2 h, and then the reaction mixture was cooled to ~20 °C. The precipitate was filtered off and recrystallized from EtOH. The yield of compound **7** was 69%, m.p. 208—210 °C. Found (%): C, 57.2; H, 4.0; N, 18.1. $C_{11}H_9N_3O_3$. Calculated (%): C, 57.1; H, 3.9; N, 18.0. IR, ν/cm^{-1}: 1690, 1712, 1740 (C=O); 3196, 3268 (NH). ^{1}H NMR, δ: 4.12 (s, 2 H, CH_2); 7.42—8.02 (m, 5 H, Ph); 12.00 and 12.45 (both s, 1 H, NH).

X-ray diffraction analysis of compounds 4 and 7. The intensity data sets were collected from crystals of **4** and **7** on an automated four-circle KM-4 (KUMA DIFFRACTION) diffractometer (monochromatized Cu-Kα radiation) with the χ geometry using the θ—2θ scanning technique in the range of $3 < \theta < 80°$. Absorption was ignored.

The crystallographic data and experimental conditions are given in Table 5.

The structures were solved by direct statistical methods followed by a series of successive electron density maps. The positions of the H atoms were revealed from the difference electron synthesis, which was calculated after the refinement of the nonhydrogen atoms with isotropic thermal parameters. The structures were refined by the full-matrix least-squares method with anisotropic thermal parameters for nonhydrogen atoms. In compound **4**, the thermal parameters of the H atoms were fixed half as large as those of the corresponding nonhydrogen atoms. The H atoms of molecule **7** were refined isotropically. The atomic coordinates are given in Tables 6, 7 (**4**), and 8 (**7**). The bond lengths and bond angles of compound

Table 5. Crystallographic data and the experimental conditions for the crystals of compounds **4** and **7**

Parameter	Value	
	4	**7**
a/Å	14.970(3)	14.766(3)
b/Å	11.103(2)	5.436(10)
c/Å	8.059(2)	15.072(3)
β/deg	77.69	118.66(3)
V/Å^3	1308.7(5)	1061.6(4)
Mol. weight	295.74	231.21
d_{calc}/g cm^{-3}	1.501	1.447
Z	4	4
μ/mm^{-1}	4.10	0.91
Radiation	Cu	Cu
R	0.065	0.039
Space group	$P2_1/c$	$P2_1/c$
Number of measured reflections	2855	2223
Number of reflections with $I > 2\sigma(I)$ used in the least-squares refinement	2038	1535
R	0.065	0.039

Table 6. Coordinates of nonhydrogen atoms (×10^4) and equivalent thermal parameters (U_{eq}×10^3) in the structure of **4**

Atom	*x*	*y*	*z*	U_{eq}/Å^2
Cl(1)	16398(1)	6888(1)	4202(2)	76(1)
S(2)	8554(1)	5387(1)	1320(1)	40(1)
O(3)	11272(1)	7580(2)	2315(3)	46(1)
O(4)	12833(2)	5106(3)	1157(3)	68(1)
N(5)	10011(2)	6607(2)	1879(3)	34(1)
N(6)	10645(2)	4562(2)	3290(3)	35(1)
N(7)	9879(2)	4580(2)	2656(3)	34(1)
C(8)	8394(2)	6864(3)	550(5)	45(1)
C(9)	9580(2)	5585(2)	2008(3)	30(1)
C(10)	10813(2)	6646(2)	2415(3)	33(1)
C(11)	11111(2)	5552(2)	3177(3)	32(1)
C(12)	11945(2)	5590(3)	3899(4)	37(1)
C(13)	12823(2)	5481(3)	2569(4)	38(1)
C(14)	13694(2)	5820(3)	3077(4)	37(1)
C(15)	14497(2)	5267(3)	2233(4)	45(1)
C(16)	15333(2)	5579(3)	2566(5)	49(1)
C(17)	15356(2)	6457(3)	3777(5)	48(1)
C(18)	14575(3)	6999(4)	4639(6)	65(1)
C(19)	13739(2)	6669(4)	4303(5)	56(1)

Table 7. Coordinates (×10^4) of H atoms in the structure of **4**

Atom	*x*	*y*	*z*
H(7)	9570	3925	2666
H(8a)	7827	6894	177
H(8b)	8889	7052	−385
H(8c)	8381	7440	1443
H(12a)	11917	4940	4712
H(12b)	11953	6344	4505
H(15)	14470	4675	1428
H(16)	15867	5210	1991
H(18)	14604	7586	5448
H(19)	13205	7023	4908

Table 8. Coordinates of atoms (×10^4) and equivalent isotropic thermal parameters (U_{eq}×10^3) in the structure of **7**

Atom	*x*	*y*	*z*	U_{eq}/Å^2
O(1)	2925(1)	−2366(4)	5526(2)	72(1)
O(2)	6084(1)	−3053(3)	5724(1)	49(1)
O(3)	7314(1)	−2504(3)	8110(1)	53(1)
N(4)	5140(1)	1329(3)	6816(1)	39(1)
N(5)	4160(1)	479(3)	6398(1)	41(1)
N(6)	4508(1)	−2773(3)	5628(1)	40(1)
C(7)	3796(1)	−1604(4)	5834(2)	43(1)
C(8)	5499(1)	−2008(4)	5965(1)	37(1)
C(9)	5789(1)	169(4)	6624(1)	35(1)
C(10)	6875(1)	1073(4)	7077(2)	38(1)
C(11)	7630(1)	−736(3)	7841(1)	33(1)
C(12)	8751(1)	−319(3)	8245(1)	32(1)
C(13)	9437(1)	−2079(4)	8896(1)	37(1)
C(14)	10491(2)	−1756(4)	9292(2)	42(1)
C(15)	10867(1)	326(4)	9055(2)	43(1)
C(16)	10193(2)	2083(4)	8416(2)	41(1)
C(17)	9136(2)	1776(4)	8006(1)	37(1)
H(5)	3736(20)	1324(51)	6601(19)	59(7)
H(6)	4300(19)	−4156(51)	5236(20)	55(7)
H(10a)	6944(21)	2602(54)	7385(21)	61(8)
H(10b)	7031(20)	1296(52)	6510(21)	62(7)
H(13)	9150(19)	−3592(51)	9055(19)	57(7)
H(14)	10984(21)	−2922(48)	9757(20)	58(7)
H(15)	11660(18)	580(47)	9352(18)	55(7)
H(16)	10426(21)	3626(58)	8205(21)	70(8)
H(17)	8671(21)	3031(51)	7542(20)	62(8)

4 are listed in Tables 1 and 2, respectively. The bond lengths and bond angles of compound 7 are listed in Tables 3 and 4, respectively. All calculations were carried out on a PC/AT computer using the SHELX-86 [15] and SHELX-93 [16] program packages.

This work was supported by the Russian Foundation for Basic Research (Project No. 95-03-08326).

References

1. C. Bulow, *Ber.*, 1904, **34**, 2198.
2. V. S. Zalesov, Yu. S. Andreichikov, Yu. A. Nalimova, S. P. Tendryakova, S. M. Starkova, and N. A. Podushkina, *Khim.-farm. Zh.*, 1978, **7**, 93 [*Pharm. Chem. J.*, 1978, **7** (Engl. Transl.)].
3. Yu. S. Andreichikov, T. N. Tokmakova, L. A. Voronova, and Yu. A. Nalimova, *Zh. Org. Khim.*, 1976, **12**, 1073 [*J. Org. Chem. USSR*, 1976, **12** (Engl. Transl.)].
4. R. B. Moffett, *J. Med. Chem.*, 1966, **9**, 475.
5. S. Bodforss, *Lieb. Ann.*, 1964, **676**, 136.
6. G. S. Predvoditeleva, T. V. Kartseva, and M. N. Shchukina, *Khim.-farm. Zh.*, 1974, **9**, 7 [*Pharm. Chem. J.*, 1974, **9** (Engl. Transl.)].
7. V. P. Kruglenko, V. P. Gnidets, N. A. Klyuev, E. V. Legachev, M. A. Klykov, and M. V. Povstyanoi, *Khim. Geterotsikl. Soedin.*, 1985, 1402 [*Chem. Heterocycl. Compd.*, 1985 (Engl. Transl.)].
8. T. N. Yanborisov, I. A. Zhikina, O. Ya. Yanborisova, S. N. Shurov, and Yu. S. Andreichikov, *Zh. Org. Khim.*, 1992, **28**, 2554 [*Russ. J. Org. Chem.*, 1992, **28** (Engl. Transl.)].
9. D. D. Nekrasov, S. N. Shurov, O. I. Ivanenko, and Yu. S. Andreichikov, *Zh. Org. Khim.*, 1994, **30**, 126 [*Russ. J. Org. Chem.*, 1994, **30** (Engl. Transl.)].
10. Z. G. Aliev, L. O. Atovmyan, A. M. Mikhailova, L. M. Pisarenko, and V. I. Nikulin, *Zh. Strukt. Khim.*, 1989, **6**, 164 [*J. Struct. Chem.*, 1989, **6** (Engl. Transl.)].
11. Yu. S. Andreichikov, D. D. Nekrasov, M. A. Rudenko, and Yu. A. Nalimova, *Khim. Geterotsikl. Soedin.*, 1988, 1411 [*Chem. Heterocycl. Compd.*, 1988 (Engl. Transl.)].
12. A. V. Milyutin, N. V. Safonova, L. F. Goleneva, Yu. S. Andreichikov, G. A. Tul'bovich, and R. R. Makhmudov, *Khim.-farm. Zh.*, 1994, **12**, 37 [*Pharm. Chem. J.*, 1994, **12** (Engl. Transl.)].
13. Yu. S. Andreichikov, L. I. Varkentin, S. G. Pitirimova, and Ya. M. Vilenchik, *Khim. Geterotsikl. Soedin.*, 1977, 1126 [*Chem. Heterocycl. Compd.*, 1977 (Engl. Transl.)].
14. Yu. S. Andreichikov, Yu. A. Nalimova, S. P. Tendryakova, and Ya. M. Vilenchik, *Zh. Org. Khim.*, 1978, **14**, 160 [*J. Org. Chem. USSR*, 1978, **14** (Engl. Transl.)].
15. G. M. Sheldrick, SHELX-86, *Programm for Crystal Structure Determination*, University of Cambridge, UK, 1986.
16. G. M. Sheldrick, *J. Appl. Crystallogr.*, 1993, **26**, 593.

Received February 17, 1997;
in revised form September 23, 1997