SYNTHESIS AND PROPERTIES OF DERIVATIVES OF sym-TRIAZINES.

11.* SYNTHESIS OF 2-AMINO-sym-TRIAZINES CONTAINING ALKYL RADICALS

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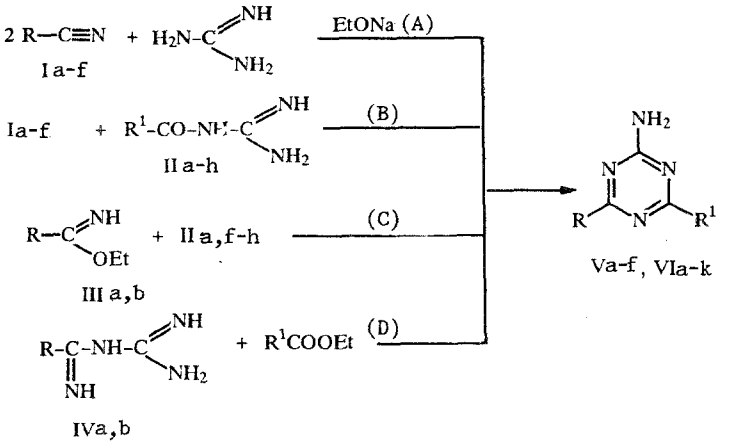
2-Amino-4,6-dialkyl-sym-triazenes were synthesized by the condensation of aliphatic nitriles with guanidine. 2-Amino-4-alkyl-6-substituted sym-triazines were prepared by the reaction of N-acylguanidines with nitriles or imino esters. Aminotriazines of this type are also formed in the condensation of N-imidoylguanidines with esters.

In a continuation of investigations into the synthesis of derivatives of sym-triazine that contain alkyl radicals [2-4], we have synthesized 2-amino-sym-triazines containing C_5-C_{17} normal alkyl radicals. Compounds of this type may be of interest as stabilizers and additives for polymeric materials, oils, and fuels.

6-Substituted 2-amino-4-alkyl-sym-triazines were prepared by several methods usually used to synthesize monoamino-sym-triazines: condensation of aliphatic nitriles (I) with guanidine [5-7] (method A) or with N-acylguanidies (II) [7, 8] (method B), condensation of N-acylguanidines (II) with iminoesters of carboxylic acids (III) [7, 9] (method C), and condensation of imidoylguanidines (IV) with esters [7, 9, 10] (method D). It should be noted that methods B-D make it possible to prepare amino-sym-triazines that contain two different substituents on the ring.



Ia–f, Va–f (R = R^1) a R = C_5H_{11}, b R = C_6H_{13}, c R = C_8H_{17}, d R = $C_{10}H_{21}$, e R = $C_{12}H_{25}$, f R = $C_{15}H_{31}$; Ig–ig R = Me, h R = Ph, and R = CCl_3; IIa–ha R^1 = C_5H_{11}, b R^1 = C_6H_{13}, c R^1 = $C_{12}H_{25}$, d R^1 = $C_{13}H_{27}$, e R^1 = $C_{17}H_{35}$, f R^1 = Ph, g R^1 = CCl_3, h R^1 = furyl 2; IIIa,b a R = C_5H_{11}, R = pyridyl, IVa,b a R = Ph, b R = pyridyl, VIa–ha R = C_5H_{11}, R^1 = Ph; b R = C_8H_{17}, R^1 = Ph; c R = C_5H_{11}, R^1 = Cl_3C; g R = C_5H_{11}, R^1 = furyl 2; d R = Me, R^1 = C_6H_{13}; f R = Me, R^1 = $C_{13}H_{27}$; g R = Me, R^1 = $C_{17}H_{35}$; h R = Ph, R^1 = $C_{13}H_{27}$; i R = Cl_3C, R^1 = $C_{12}H_{25}$; j R = Cl_3C, R^1 = $C_{17}H_{35}$; k R = pyridyl 2, R^1 = C_5H_{11}

*For Communication 10, see [1].

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TABLE 1. Characteristics of the Compounds Synthesized

Compound	Molecular formula	T_m, °C	R_f**	PMR spectrum, δ, ppm					Yield, %
				CH_3, t	CH_2, m	CH_2, t	NH_2 (bs 2H)	other protons	
Va	$C_{13}H_{24}N_4$	79...80,5	0,68 (a)	1,14 (6H)	1,26...1,38 (12H)	3,18 (4H)	5,69	—	60 (A), 64 (B), 83 (C)
Vb	$C_{15}H_{28}N_4$	82...83	0,50 (b)	1,12 (6H)	1,32...1,46 (16H)	3,08 (4H)	5,60	—	52 (A)
Vc	$C_{19}H_{36}N_4$	90...91,5	0,42 (b)	1,21 (6H)	1,28...1,74 (24H)	2,96 (4H)	5,78	—	55 (A)
Vd	$C_{23}H_{44}N_4$	85...86	0,56 (b)	1,14 (6H)	1,26...2,04 (32H)	2,92 (4H)	6,28	—	50 (A)
Ve	$C_{27}H_{52}N_4$	87...88	0,52 (b)	1,08 (6H)	1,30...1,96 (40H)	2,88 (4H)	6,08	—	45 (A), 86 (B)
Vf	$C_{33}H_{64}N_4$	92...93	0,48 (b)	1,20 (6H)	1,30...1,88 (52H)	3,10 (4H)	6,15	—	48 (A)
VIa	$C_{14}H_{18}N_4$	134...135,5	0,29 (b)	1,15 (3H)	1,30...1,41 (6H)	3,04 (2H)	5,82	6,88...7,08 (5H, (m. arom.)	73 (B), 82 (C), 91 (D)
VIb	$C_{17}H_{24}N_4$	146...147	0,54 (a)	1,18 (3H)	1,34...1,60 (12H)	3,20 (2H)	5,73	6,92...7,16 (5H, (m. arom.)	84 (B), 93 (D)
VIc	$C_9H_{13}Cl_3N_4$	148...149	0,42 (c)	1,10 (3H)	1,38...1,50 (6H)	3,12 (2H)	6,10	—	78 (B), 85 (B)
VId	$C_{12}H_{16}N_4$	210 (decomp.)	0,36 (a)	1,15 (3H)	1,28...1,41 (6H)	3,08 (2H)	5,85	6,12 (1H, 1H, d.d. 4-H furan J_{34} = 3,5 Hz), 6,28 (1H, d.d, 3-H, furan J_{35} = 0,8 Hz), 7,10 (1H, d.d, 5-H, furan J_{45} = 1,8 Hz)	76 (B), 90 (B)
VIe	$C_{10}H_{18}N_4$	142....143,5	0,74 (c)	1,16 (3H)	1,42...1,80 (8H)	3,10 (2H)	5,66	2,32 (3H, s Me)	60 (B)
VIf	$C_{17}H_{32}N_4$	126...126,5	0,64 (b)	1,22 (3H)	1,31...1,92 (22H)	2,93 (2H)	5,60	2,54 (3H, s, Me)	87 (B)
VIg	$C_{21}H_{40}N_4$	129...130	0,58 (b)	1,18 (3H)	1,30...2,06 (30H)	2,98 (2H)	5,86	2,45 (3H, s, Me)	84 (B)
VIh	$C_{22}H_{34}N_4$	118...119	0,34 (c)	1,06 (3H)	1,24....2,02 (22H)	2,83 (2H)	6,04	7,02....7,18 (5H, (m.arom.)	89 (B)
VIi	$C_{16}H_{27}C_{13}N_4$	140...141,5	0,87 (b)	1,25 (3H)	1,38...1,90 (20H)	3,30 (2H)	6,14	—	92 (B)
VIj	$C_{21}H_{37}C_{13}N_4$	163...164	0,81 (b)	1,21 (3H)	1,25...1,98 (30H)	3,15 (2H)	5,92	—	90 (B)
VIk	$C_{13}H_{17}N_5$	164...165	0,78 (c)	1,14 (3H)	1,30...1,44 (6H)	3,22 (2H)	5,78	7,38...7,78 (4H, (m, arom.)	89 (C), 92 (D)

*Compounds purified by recrystallization: Va, b) from a 5:1 ethanol:hexane mixture; Vc, VIa, b, k) from ethanol; Vd-f, VIe, g-m) from aqueous ethanol; VIc, d, j) from aqueous DMF; VIf) from acetonitrile.

**TLC on Al_2O_3:solvent system denoted in parentheses (see Experimental).

***Spectra of compounds Va-f, VIb, f-h, k were taken in DMSO-D_6; of VIa, c, d, i, j — in acetone-D_6; of VIe — in CD_3OD.

Condensation by method A of nitriles Ia-f gave us 2-amino-4,6-dialkyl-sym-triazines: (Va-f). Better yields of these products (45-60%) were achieved by prolonged boiling of the reactants (2:1 mole ratio) in butanol in the presence of sodium ethoxide. Here, along with 2-amino-sym-triazines Va-f, unidentified, high-melting products (T_{mp} > 320°C) that are poorly soluble in most organic solvents were formed by side reactions.

By method B, 2-amino-4,6-dialkyl-sym-triazines Va, e and 6-substituted 2-amino-4-alkyl sym-triazines VIa-j (Table 1) were made from nitriles Ia, c, e-i and N-acylguanidines IIa-h in yields of 60-92%. Compounds Va, e and VIa-d, h were synthesized by heating (140-150°C; 6-8 h) the corresponding nitriles with N-acylguanidines IIa-h (4:1 mole ratio). 2-Amino-sym triazines VIf, g, i, j were obtained by the prolonged boiling (18-20 h) of N-acylguanidines IIc-e in excess acetonitrile, Ig, of trichloroacetonitrile, Ih.

As a result of the condensation of N-acylguanidies IIa, f-h with reactive iminoesters IIIa, b by method B, 2-amino-sym-triazines Va and VIa, c, d, k (Table 1) were formed in high yields after equimolar amounts of the reactants were boiled briefly in ethanol. Nitriles and amides of carboxylic and nicotinic acid were found by means of TLC to be by-products.

2-Amino-sym-triazines VIa, b, k were synthesized similarly from N-imidoylguanidines IVa, b and esters of the appropriate acid (method D).

In the IR spectra of all of the 2-amino-sym-triazines synthesized, Va-f and VIa-k, absorption maxima of variable intensity were found at 1570-1555, 1530-1520, 1425-1410, 1120-1095, 1010-995, 815-805, and 720-705 cm^{-1}, which are characteristic of out-of-plane stretching and in-plane bending vibrations of sym-triazines [1-4, 11, 12]. In the spectra of sym-triazines containing trichloromethyl groups (VIc, i, j), these bands are at lower frequencies than in the spectra of other amino-sym-triazines. The spectra of all of the compounds have two broad absorption bands in the NH stretching region: one of them is found in the 3330-3285 range (ν_{as} NH), the other appears in the 3180-3155 cm^{-1} (ν_{as} NH) range. Such a location and shape of the doublet of NH stretching vibrations is evidence of a strong hydrogen bond in these compounds in the crystalline state [4, 11]. Strong absorption maxima at 1660-1635 cm^{-1} are assigned to the scissoring vibration of the NH bonds in the primary amino groups of the sym-triazines [4, 13].

In the PMR spectra of 2-amino-sym-triazines Va-f and VIa-k (Table 1), the proton signals of the amino groups appear as broad singlets of two proton units of intensity at 5.60-6.26 ppm [1, 4]. In the 1.08-3.30 ppm region of the spectra of these compounds, three groups of signals are found belonging to the protons of the alkyl radicals. The signals from the methyl group protons are symmetrical triplets at 1.08-1.25 ppm. The complex multiplets in the 1.26-2.04 ppm range must be assigned to protons of methylene units, and the nonsymmetric triplets, shifted to a weaker field (2.38-3.30 ppm) to CH_2 groups bound to the triazine ring [2, 3].

The mass spectra of the 2-amino-sym-triazines synthesized also give good support to the proposed structure. The molecular masses found for the substances correspond to the calculated values, and the nature of the fragmentation agrees with the structures given. In the mass spectra of all of the sym-triazines studied, Va-f and VIa-k (Table 2), there are peaks from the $M^{+\cdot}$ molecular ions (5.0-37.0%), the stability of which depends on the susceptibility of the substituents to electron impact fragmentation. All of the spectra contain peaks of $[M-H]^+$ ions, the intensity of which is about half that of the corresponding $M^{+\cdot}$. This is characteristic of amino derivatives of sym-triazines [14]. For compounds Va-f, VIa-k containing alkyl groups easily split off by electron impact, the competing breakup of the triazine ring is not characteristic, but the presence of the latter is brought about by an extremely specific tendency for fragmentation. The primary fragmentation of the $M^{+\cdot}$ molecular ions in the mass spectra of 2-amino-4,6-dialkyl-sym-triazines Va-f and 6-substituted 2-amino-4-alkyl-sym-triazines VIa, b, d-h, k takes place in several directions simultaneously. The basic decomposition of the $M^{+\cdot}$ of these compounds is related to a β-cleavage of a C—C bond of the alkyl radical and the elimination of an alkene with the composition $C_{n-1}H_{2n-2}$ are the result of a McLafferty rearrangement [3, 15]. Here, a fragment ion Φ_1 is formed, the intensity of which is a maximum in the mass of these compounds.

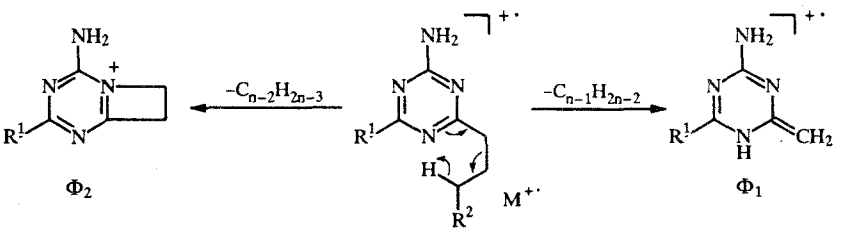

TABLE 2. Mass Spectra of 2-Amino-4-Alkyl-6-Substituted sym-Triazines

Compound	m/z values (I_{rel}, %)*
Va	236 (M^+, 7,5), 235 (3), 193 (Φ_2, 22,5), 192 (4), 180 (Φ_1, 100), 151 (3), 137 (Φ_3, 65), 124 (Φ_4, 27), 97 (24), 96 (32)
Vb	264 (M^+, 6,5), 263 (3), 235 (5), 207 (Φ_2, 15), 194 (Φ_1, 100), 165 (7), 151 (10), 137 (Φ_3, 54), 124 (Φ_4, 37), 97 (28), 96 (19)
Vc	320 (M^+, 7), 319 (4), 277 (5), 235 (Φ_2, 25), 222 (Φ_1, 100), 193 (12), 137 (Φ_3, 59), 124 (Φ_4, 18), 97 (30), 96 (25)
Ve	432 (M^+, 9), 431 (4), 291 (Φ_2, 27), 290 (10), 278 (Φ_1, 100), 154 (27), 153 (10), 137 (Φ_3, 63), 124 (Φ_4, 21), 96 (28)
Vf	516 (M^+, 6), 515 (3), 333 (Φ_2, 18), 320 (Φ_1, 100), 196 (14), 182 (9), 137 (Φ_3, 63), 124 (Φ_4, 12), 97 (24), 96 (38)
VIa	242 (M^+, 37), 241 (13), 213 (7), 199 (Φ_2, 23), 186 (Φ_1, 100), 185 (15), 104 (Φ_6, 35), 103 (Φ_5, 76), 77 (22), 51 (45)
VIb	284 (M^+, 32), 283 (7), 241 (12), 199 (Φ_2, 17), 186 (Φ_1, 100), 185 (14), 104 (Φ_6, 23), 103 (Φ_5, 70), 77 (32), 51 (24)
VIc	282** (M^+, 5), 247** (Φ_8, 30), 239** (Φ_2, 21), 221 (Φ_1, 76), 212** (Φ_7, 100), 186** (8), 151** (18), 117** (Φ_9, 26), 116 (8), 108** (Φ_{10}, 61)
VId	232 (M^+, 33), 203 (M-HCO, 36), 193 (M-C_3H_3, 9), 189 (Φ_2, 52), 176 (Φ_1, 100), 165 (32), 94 (Φ_6, 38), 93 (Φ_5, 70), 67 (9), 39 (13)
VIf	292 (M^+, 15), 291 (6), 168 (17), 154 (54), 137 (Φ_2, 26), 124 (Φ_1, 100), 97 (37), 96 (11), 83 (14), 82 (10)
VIi	380** (M, 9), 345** (Φ_8, 52), 310** (Φ_7, 100), 275 (31), 239** (Φ_2, 27), 221** (Φ_1, 67), 186** (17), 151 (7), 117** (Φ_9, 18), 108** (Φ_{10}, 32)
VIj	450** (M^+, 5,5), 415** (Φ_8, 43), 380** (Φ_7, 100), 239** (Φ_2, 21), 221** (Φ_1, 81), 166** (8), 131** (18), 117** (Φ_9, 26), 108** (Φ_{10}, 61), 96 (7)
VIk	243 (M^+, 18), 242 (8), 214 (5), 200 (Φ_2, 22), 187 (Φ_1, 100), 186 (32), 105 (Φ_6, 30), 104 (Φ_5, 67), 78 (21), 51 (19)

*The ten most intense peaks in the mass spectrum are given.

**Ions containing the ^{35}Cl isotope.

To a lesser degree, the mass spectra of these compounds show the parallel elimination of an alkyl radical with the composition $C_{n-2}H_{2n-3}$ leading to Φ_2 ions, the intensity of whose peaks amounts to 15-27% of the maximum. The further disintegration of Φ_1 ions for dialkyl-sym-triazines Va-f is due to the ready splitting off of a $C_{n-2}H_{2n-3}$ radical at the expense of the second alkyl group. The intensity of the peaks of the corresponding ions, $[M-C_{n-2}H_{2n-2}-C_{n-2}H_{2n-3}]^+$ (ion Φ_3; m/z 137), is 54-67% of the maximum. The ejection of a second $C_{n-1}H_{2n-2}$ alkene molecule (ion Φ_4; m/z 124), from the Φ_1 ion takes place with greater difficulty.

In the mass spectra of 2-amino-3-alkyl-6-phenyl(heteryl)-sym-triazines (VIa, b, d, k) pronounced processes are found that are due to the cleavage of the triazine ring in the Φ_1 ion. The destruction of the ring in the Φ_1 ion takes place with the formation of both an odd-electron fragment, $[R^1CN]^{+\cdot}$ (ion Φ_5), and an even-electron fragment, $[R^1CNH]^+$ (ion Φ_6), which is characteristic of aryl- and heteryl-substituted sym-triazines [14, 16]. The intensity of the Φ_5 peaks in the spectra of these compounds is 70-76%, and the intensity of the Φ_6 peaks is 22-48% of the maximum.

In the mass spectra of 2-amino-sym-triazine VId, one also finds processes related to the concurrent disintegration of the furan ring, leading to the formation of ionic fragments with m/z 203 $[M-HCO]^+$, m/z 193 $[M-C_3H_3]^+$, and m/z 39 [16].

The basic fragmentation path of $M^{+\cdot}$ ions in the mass spectra of compounds VIc, i, j is determined by the sequential elimination of three chlorine atoms from $M^{+\cdot}$ with the peaks of the $[M-Cl_2]^+$ ions (Φ_7 ion) having the maximum intensity, and the peaks of the $[M-Cl]^+$ ions (Φ_8 ion) having an intensity of 30-52% that of the maximum. The second path for the disintegration is determined by the β-cleavage of the C—C bonds of the alkyl radicals with the formation of fragment ions with m/z 239 (Φ_2 ion) and m/z 221 (Φ_1 ion). Further fragmentation of the Φ_1 ion takes place by the sequential elimination of three chlorine atoms. In the mass spectra of amino-sym-triazines VIc, i, j there are also present peaks of ions with m/z 117 $[Cl_3C]^+$ (Φ_9 ion), and with m/z 108 $[Cl_2CCN]^+$ (Φ_{10} ion).

Note that in the mass spectra of the 2-amino-sym-triazines studied, Va-f and VIa-k, low-intensity peaks are observed for ions corresponding to the ejection of C_mH_{2m+1} (m = 1 to n — 3, where n is the number of carbon atoms) from the molecular ion.

EXPERIMENTAL

The IR spectra were taken on a UR-20 instrument in KBr tablets. The PMR spectra were recorded on a Bruker WP-80SY spectrometer with TMS internal standard. The mass spectra were obtained on an LKB-2091 instrument with direct introduction of the sample into the ion source (energy of ionizing electrons 70 eV, emission current 25 μA, temperature of ion source 200°C) at a temperature of sample vaporization of 130-150°C. The course of the reactions and the purity of the resultant products were monitored by means of TLC on Al_2O_3 (III st. act. according to Brokeman) with solvent systems CCl_4—ethanol, 15:1 (a), benzene—methanol, 20:1 (b), and benzene—methanol, 10:1 (c), and development with iodine vapor.

The initial N-hexanoyl- (IIa) [17], N-heptanoyl- (IIb) [17], N-benzoyl- (IIf) [18], N-trichloroacetyl- (IIg) [18], and N-(furoyl-2)guanidine (IIh) [19]; and the ethyl iminoesters of caproic (IIIa) [20] and nicotinic acids (IIIb) [21] were obtained by the methods cited. N-Tridecanoyl- (IIc) [2], N-tetradecanoyl- (IId) [2], and N-octadecanoylguanidine (IIe) [2], as well as N-benzimidoyl- (IVa) [7] and N-(pyridyl-3-imidoyl)guanidine (IVb) [7] were synthesized previously.

Characteristics of the triazines synthesized for the first time are shown in Table 1. The data from elementary analyses for C, H, and N agree with the calculated values.

2-Amino-4,6-dialkyl-sym-triazines (Va-f). A. 2.02 g (30 mmoles) of guanidine hydrochloride was added in portions to a solution of sodium ethoxide prepared from 0.74 g (32 mmoles) of sodium in 50 ml of absolute ethanol. The reaction mixture was boiled for 1 h, cooled to 0°C, filtered off the NaCl that precipitates, and the filtrate evaporated under vacuum. The residue was dissolved in 100 ml of butanol, 60 mmoles of nitrile Ia-f was added, and the solution was boiled for 18-20 h with stirring until the initial nitrile disappeared (judged by TLC). The solvent was distilled off under vacuum, the residue washed with hot hexane (3 × 15 ml), and crystallized from the suitable solvent (see Table 1).

2-Amino-4-pentyl-6-phenyl-sym-triazine (VIa). B. A mixture of 4.65 g (48 mmoles) of nitrile Ia and 1.95 g (12 mmoles) of N-benzoylguanidine (IIf) was stirred for 8 h at 150°C. The reaction mixture was evaporated down under reduced pressure, and the residue was washed with ether (3 × 15 ml).

2-Amino-sym-triazines Va, e and VIb-d, h were synthesized in analogous fashion.

2-Amino-4-dodecyl-6-trichloromethyl-sym-triazine (VIi). B. A mixture of 10.0 g (70 mmoles) of trichloroacetonitrile (Ii) and 3.06 g (12 mmoles of N-acylguanidine (IIc) was boiled with stirring for 18-20 h until the initial IIc disappeared (judged by TLC) from the reaction mixture. The reaction mixture was evaporated down under reduced pressure and the residue washed with ether to obtain sym-triazine VIi.

sym-Triazines VIf, g, j were synthesized in analogous fashion.

2-Amino-4-pentyl-6-(furyl-2)-sym-triazine (VId). C. A mixture of 2.29 g (15 mmoles) of N-acylguanidine IIh and 2.14 g (15 mmoles of ethyl imidoester IIIa was boiled in 40 ml of absolute ethanol for 2 h with stirring, cooled to 20°C, and poured into 200 ml of cold water. The precipitate of product VId that forms was filtered off and dried in vacuum over P_2O_5.

2-Amino-sym-triazines Va and VIa, c, k were synthesized in analogous fashion.

2-Amino-4-octyl-6-phenyl-sym-triazine (VIb). A mixture of 3.24 g (20 mmoles) of N-imidoylguanidine IVa and 3.72 g (20 mmoles) of the ethyl ester of pelargonic acid in 50 ml of absolute ethanol was boiled and then treated as described for compound VId to obtain sym-triazine VIb.

2-Amino-sym-triazines VIa, k were synthesized in analogous fashion.

REFERENCES

1. V. I. Kelarev, A. S. Remizov, R. A. Karakhanov, Yu. N. Polivin, and D. Oietauo, Khim. Geterotsikl. Soedin., No. 10, 1312 (1992).
2. V. I. Kelarev, Dibi Ammar, and A. F. Lunin, Khim. Geterotsikl. Soedin., No. 11, 1557 (1985).
3. V. I. Kelarev, Dibi Ammar, A. F. Lunin, and O. V. Malova, Zh. Org. Khim., **21**, 1306 (1985).
4. V. I. Kelarev, R. A. Karakhanov, V. I. Zav'yalov, Dibi Ammar, A. N. Golovin, E. A. Lisitsyn, and M. Bellul, Zh. Org. Khim., **24**, 1100 (1988).
5. H. Kabbe, K. Eiter, and F. Moelle, German Patent No. 1,212,547; Chem. Abstr., **64**, 15900 (1966).
6. D. Alsofrom, H. Grossberg, and H. Sheffer, J. Heterocycl. Chem., **14**, No. 4, 917 (1976).
7. V. I. Kelarev, R. A. Karakhanov, M. Bellul, R. A. Ushakova, and A. I. Mikaya, Khim. Geterotsikl. Soedin., No. 5, 674 (1988).

8. R. B. Russell, G. H. Hitchings, and B. H. Chase, J. Am. Chem. Soc., **74**, No. 21, 5403 (1952).
9. V. I. Kelarev, R. A. Karakhanov, A. S. Kokosova, and G. D. Gankin, Khim. Geterotsikl. Soedin., No. 9, 1250 (1992).
10. H. Nagasaka, E. Joshikawa, and K. Odo, J. Syn. Org. Chem. Jpn., **25**, 1048 (1967).
11. A. I. Finkel'shtein and E. N. Boitsov, Usp. Khim., **31**, 1496 (1962).
12. A. R. Katritskii (ed.), Physical Methods in the Chemistry of Heterocyclic Compounds [in Russian], Khimiya, Moscow—Leningrad (1966), p. 594.
13. A. I. Finkel'shtein, Opt. Spektrosk., **5**, 264 (1958).
14. P. N. Preston, P. W. Steedmann, M. H. Palmer, S. M. Mackenzie, and M. F. Stevens, Org. Mass Spectrom., **3**, No. 7, 863 (1970).
15. R. Johnson, Handbook of Mass Spectrometry for Organic Chemists [Russian translation], Mir, Moscow (1975).
16. Q. H. Porter and J. Balders, Mass Spectrometry of Heterocyclic Compounds, Wiley—Interscience, New York (1971), p. 376.
17. Beilstein, Vol. 3, p. 75 (1932).
18. W. Traube, Chem. Ber., **43**, No. 8, 3590 (1910).
19. A. A. Smolyarchuk, A. P. Kobernik, N. I. Ivanova, I. M. Skvortsov, and Yu. V. Aleksashin, Khim.-farm. Zh., **10**, No. 7, 72 (1976).
20. A. M. Drozdov and E. N. Bekhli, Zh. Org. Khim., **14**, No. 2, 289 (1944).
21. H. Watanabe, Y. Kikugawa, and S. Yamada, Chem. Pharm. Bull., **21**, No. 3, 465 (1973).