HYDRAZONES

XI.* SPECTRA AND STRUCTURE OF 1-HYDRAZINOPHTHALAZINES

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The products of the reaction of 1-chloro- and 1,4-chlorophthalazines with hydrazine are the E isomers of the hydrazones of the corresponding phthalazone rather than hydrazinophthalazines, as previously assumed. The hydrazones of 2-methylphtalazones exist in the Z form.

The products of the reaction of 1-chloro- and 1,4-dichlorophthalazine with hydrazine [3] have been assigned 1-hydrazinophthalazine and 1-hydrazino-4-chlorophthalazine structures, respectively (the first of these products has found practical application as the hypotensive preparation apressin [2]). The structures of these compounds (I and IV) have not been studied, although α-hydrazino derivatives of azacycles may exist in the hetarylhydrazine form (2-pyridyl-, 2-quinolyl, and 1,3,4-triaza-2-carbazolylhydrazines [4, 5]) or in the tautomeric hydrazone form (pyridazone hydrazones [6]). It is known that I displays dual reactivity. On oxidation it is converted to phthalazine [7], and it undergoes reaction with carboxylic acid derivative to give 3-R-1,2,4-triazolo[3,4-a]phthalazines[3,8]; this can be explained by interconversion or equilibrium between the hydrazine and hydrazone forms [3].

In order to study this problem we compared the data from the IR, PMR, and UV spectra of I and IV and their methylated analogs, which have fixed hydrazine (XVIII) or hydrazono structures (III, V, and VI), the product of reaction of 1-chlorophthalazine (XIII) with methylhydrazine (II), and a number of compounds having authentic phthalazine structure [9-12] (XII-XVII) (Tables 1 and 2).

The UV spectra of phthalazone VIII and 4-phthalazone X (Tables 1 and 2) have two absorption bands and are similar to the spectra of their 2-methyl derivatives (IX and XI); this confirms the conclusion regarding the phthalazone structure of VIII (and, consequently, of X), which was drawn on the basis of the IR spectra [9]. The introduction of a methyl group in the 2 position (phthalazones IX and XI) leads to a small bathochromic shift of the longwave band ($\Delta\lambda$ 8-9 nm). The introduction of a chlorine atom in the 4 position (X and XI) leads to a small bathochromic of shift of the longwave ($\Delta\lambda$ 8-9 nm) and shortwave ($\Delta\lambda$ 4-6 nm) bands. At the same time, the UV spectrum of 1-chloro-4-methoxyphthalazine XVI (Table 2), which has a fixed aromatic system (like other phthalazines XII-XVI), differs sharply from the spectra of phthalazones VIII-XI.

The UV spectra of I and IV, like the spectra of phthalazones VIII-XI, have two absorption bands, but they are found in the longer-wavelength portion of the spectrum (Table 1 and Fig. 1).

The UV spectra of I and IV are similar to one another and to the spectra of 2-methylphthalazone hydrazone (III) and 2-methyl-4-chlorophthalazone hydrazone (V) but differ markedly from the spectrum of 1-(α-methylhydrazino)-4-chlorophthalazine (XVIII). Consequently, these compounds have phthalazone hydrazone (I) and 4-chlorophthalazone hydrazone (IV) structures rather than hydrazinophthalazine structures, as previously postulated [2, 3]. In attempts to synthesize another model compound with a fixed hydrazine structure – 1-(α-methylhydrazino)phthalazine – by reaction of 1-chlorophthalazine with methylhydrazine by the method in [8] we isolated only II. Its UV spectrum differs markedly from the spectrum of hydrazine XVIII but is similar to the spectra of hydrazones I and III-V (Table 1 and Fig. 1). In contrast to hydrazine XVIII, II does not

*See [1] for communication XXXIX.

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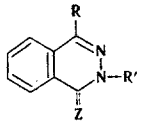
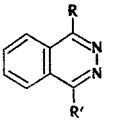
TABLE 1. Phthalazones VIII-XI, Hydrazones I-VI, and Imine VII



Compound	R	R′	Z	mp, °C	UV spectrum, λ_{max}, nm (log ε), dioxane	IR spectrum, ν_{max}, cm^{-1} (mineral oil)		
						NH_2	NH	C=N
I	H	H	NNH_2	170—172	340 (3,66); 275 (4,15)	3290, 3192	3225	1630
II	H	H	$NNHCH_3$	107—108	344 (3,70); 279 (4,15)	—	3215, 3297*	1648
III	H	CH_3	NNH_2	86—88	355 (3,64); 280 (4,00)	3310, 3188	—	1650
IV	Cl	H	NNH_2	200†	333 (3,84); 279 (4,11)	3296, 3168	3256	1650
V	Cl	CH_3	NNH_2	106—108	350 (3,56); 285 (4,08)	3314, 3174	—	1665
VI	Cl	CH_3	$NN(CH_3)_2$	67—70	355 (3,54); 275 (4,00)	—	—	1669
VII	Cl	CH_3	NH	100—104	320 (3,78); 275 (4,00)	—	3280	1613
VIII	H	H	O	183	286 (3,90); 254 (3,95)	—	3174	1672‡
IX	H	CH_3	O	112	294 (3,90); 255 (3,87)	—	—	
X	Cl	H	O	274	294 (3,79); 258 (4,00)	—	3160	1680‡
XI	Cl	CH_3	O	128—130	303 (3,84); 261 (3,69)	—	—	

*This is the ν_{NH} band of the hydrazone fragment.
†With decomposition.
‡$\nu_{C=O}$.

TABLE 2. Properties of Phthalazine Derivatives XII-XVIII (in dioxane)



Compound	R	R′	mp, °C	UV spectrum, λ_{max}, nm (log ε)
XII	H	H	90	262 (3,62)
XIII	H	Cl	115*	266 (3,64)
XIV	Cl	Cl	165	260 (3,71)
XV	OPh	OPh	222	275 (4,53)
XVI	Cl	OCH_3	108	273 (3,88)
XVII	Cl	$N(CH_3)_2$	101	320 (3,76)
XVIII	Cl	$N(CH_3)NH_2$	141—141,5†	315 (3,90)

*According to [22], this compound has mp 113° (mp 119-120 [23]).
†ν_{max} NH_2 (mineral oil) 3292 and 3199 cm^{-1}, δ_{max} NH_2 1640 (w) cm^{-1}.

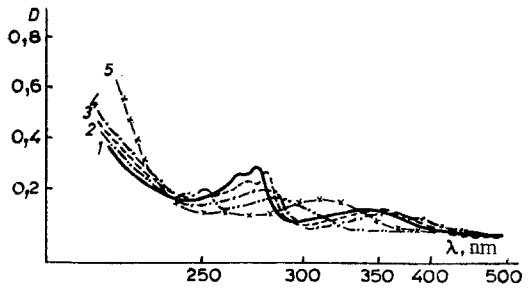


Fig. 1. UV spectra in dioxane: 1) I; 2) VIII; 3) II; 4) III; 5) XVIII.

react with aromatic aldehydes. These data constitute evidence that II is a phthalazone methylhydrazone.

Replacement of the oxygen atom by an imino group on passing from 2-methyl-4-chlorophthalazine (XI) to its imine (VII) leads to a distinctly expressed bathochromic shift of both absorption bands ($\Delta\lambda \sim 15$ nm); this was previously observed in the case of succinamide and its imino derivatives and is explained by the difference in the electronegativities of the nitrogen and oxygen atoms [13]. Replacement of the oxygen atom by a nitrogen atom on passing from 4-methoxy- (XVI) to 4-dimethylamino-1-chlorophthalazine (XVII) brings about an even more distinct bathochromic shift ($\Delta\lambda \sim 47$ nm); this is also associated with the higher capacity of the unshared pair of electrons of the nitrogen atom for conjugation with the π system.

The expected bathochromic shift of both bands ($\Delta\lambda \sim 30$ nm and $\Delta\lambda' \sim 8$ nm), which is a consequence of the manifestation in hydrazone V of $p-\pi$ conjugation of the unshared pair of the amine nitrogen atom with the π electrons of the imine bond and the heterocycle [4, 14], is observed on passing from imine VII to hydrazone V, whereas a hypsochromic shift ($\Delta\lambda \sim 5$ nm), although small, occurs on passing from 1-chloro-4-dimethyl-aminophthalazine XVII to hydrazine XVIII. These facts once again confirm the hydrazone structure of I, II, and IV.

Quantum-chemical calculations within the π approximation (Hückel method) for 2-hydrazinopyridine and 1-hydrazinophthalazine also indicate the preferableness of the hydrazone form of I. However, the results of the calculations (Table 3) depend on the ratio of the electronegativities of the atoms in the fragment under discussion and the nature of the heterocycle.

The coulombic integral of the imine nitrogen atom (δ_N) was assumed to be 0.5β in the calculations [15, 16], whereas the resonance integrals of all of the bonds were taken to be equal to β. As seen from Table 3, an increase in the coulombic integral of the amine nitrogen atom to 1.5β leads to preferableness of the hydrazine form both for 2-hydrazinopyridine and for 1-hydrazinophthalazine. The preferableness of the form with an exocyclic multiple bond is determined by the degree of participation of the terminal heteroatom in conjugation, which decreases as its electronegativity increases. Replacement of the imine nitrogen atom by oxygen should lead to preferableness of the form with an exocyclic multiple bond, whereas replacement of the amine nitrogen atom by oxygen should lead to a tautomer with aromatic character of the heteroring.

The IR spectra also confirm the hydrazone structure of I, II, and IV. Absorption at 1600-1660 cm^{-1} (stretching vibrations of the hydrazone C=N bond [14]) is absent in the IR spectra of phthalazines XII-XVII. Intense absorption at 1650-1670 cm^{-1} is observed in this region for hydrazones III and V and 2-methylphthalazone dimethylhydrazone (VI), whereas intense absorption at 1630-1650 cm^{-1} is observed for I, II, and IV (Table 1). The ring C=N bond shows up in the IR spectra together with the ring vibrations, as attested to by the data for phthalazones VIII-XI and phthalazines XII-XVIII. The introduction of a chlorine atom in the 4 position and of methyl groups in the 2 position or attached to the amine nitrogen atom of hydrazone I leads to a shift of the C=N (Table 1) to the high-frequency region. Deformation vibrations of the NH_2 group appear in the same region but their intensity is low. The IR spectrum of hydrazine XVIII contains a low-intensity absorption band at 1640 cm^{-1}. This and, above all, the IR spectral data for dimethylhydrazone VI confirm the correctness of the assignment of the absorption band of the C=N bond in the compounds under consideration.

There are three absorption bands in the region of stretching vibrations of NH bonds in the spectra of hydrazones I and IV. The bands at 3290 and 3192 cm^{-1} for hydrazone I and at 3296 and 3168 cm^{-1} for

TABLE 3. Dependence $\Delta E^{\pi} = E^{\pi}_{\text{hydrazine}} - E^{\pi}_{\text{hydrazone}}$ in β Units ($\beta \approx 20$ kcal/mole) on Coulombic Integral α_N for 2-Hydrazinopyridine and 1-Hydrazinophthalazine

	2-Hydrazinopyridine	1-Hydrazinophthalazine	α_N
$\Delta E\pi$	−0,008	−0,101	1,0*
	0,057	−0,037	1,2
	0,144	0,046	1,5†

*According to the data in [15].
†According to the data in [16].

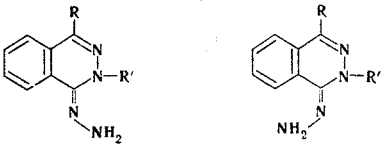
TABLE 4. PMR Spectra of Phthalazine Derivatives I-XVIII

Compound	Solvent	δ, ppm			
		H-5—H-7	H-8	H-4	other protons
I	DMSO	7,80 d	8,20 m	8,29 s	6,58 broad signal, 3NH
I+H_2O	DMSO	7,90 d	8,16 m	8,47 s	4,50 broad, NH+H_2O
I	Hexametapol	7,50 d	8,00 m	7,77 s	6,77 broad, 3NH
I	Tetramethylurea	7,50 d	8,01 m	7,86 s	6.34 broad, 3NH
I	Dioxane	7,40 m	7,94 m	7,47 s	5,48 s, 3NH
II	DMSO	7,53 d	8,00 m	7,78 s	
III	DMSO	7,48 m*	8,74 m		5,21 broad, NH_2, 3,37 s, CH_3
IV	DMSO	7,92 d	8,17 m	—	
IV	Hexametapol	7,90 t	8,50 m	—	7,07 very broad, 3NH
V	DMSO	7,60 m	8,76 m	—	5,73 broad, NH_2, 3,42 s, CH_3
VI	DMSO	7,88 m	9,67 m	—	
VI	CCl_4	7,63 m	9,80 m	—	3,60 s, CH_3, 2,47 s, $2CH_3$
VII	DMSO	7,83 m	8,30 m	—	6,76 broad, NH, 3,74 s, CH_3
VII	Hexametapol	7,98 m	8,65 m	—	5,61 broad, NH, 3,67 s, CH_3
VIII	Dioxane	7,70 d	8,28 m	8,05 s	11,30 broad, NH
VIII	DMSO	7,98 m	8,26 m	8,43 s	12,76 broad, NH
VIII	Hexametapol	8,07 m, 8,28 m		8,59 s	13,66 broad, NH
IX	DMSO	7,89 m	8,15 m	8,38 s	
X	DMSO	8,10 m	8,33 m	—	12,96 broad, NH
X	Hexametapol	8,09 m	8,27 m	—	13,90 broad, NH
XII	DMSO	8,16 d		9,87 s†	
XIV	DMSO	8,25 d			
XVI	Chloroform	8,10 m			4,30 s, CH_3
XVII	DMSO	7,98 m			3,19 s, $2CH_3$
XVII	CCl_4	8,02 m			3,18 s, $2CH_3$
XVIII	DMSO	7,77 m	8,97 m		4,98 s, NH_2, 3,75 s, CH_3
XVIII	Hexametapol	8,20 m	9,63 m		5,77 broad, NH_2

*The H-4 proton is also found in the same place.
†The H-1 proton is also found in the same place.

hydrazone IV (Table 2) should be assigned to the stretching vibrations of the amino group of the hydrazone fragment, whereas the bands at 3225 cm^{-1} for I and at 3256 cm^{-1} for IV should be assigned to vibrations of the ring NH group. The assignment of the absorption bands of the NH group in hydrazone II were made similarly (Table 1).

Hydrazones I, II, and IV can exist in two geometrical forms (E and Z isomers [4, 14]).



E isomer Z isomer

From the known facts of the shift to weak field of the signal of the aromatic proton in the 2 position in the PMR spectrum of the Z form of the semicarbazone [17] and β-guanylhydrazones of isatin [18] (as compared with the position of the signal of the analogous proton in the E form) it may be assumed that the chemical shifts of the proton in the 8 position of the phthalazine ring should be different for the Z and E isomers. The identical character of the forms of the signals and the closeness of the positions of the chemical shifts of the aromatic protons, including H-8, for 4-R-phthalazone hydrazones I, II, and IV and phthalazone hydrazones VIII-XI (Table 4 and Fig. 2) make it possible to conclude that the hydrazones exist in the E form.

The preferableness of the E form of phthalazone hydrazone I is confirmed by a comparison of the experimental (μ_I = 2.13 D, dioxane, 25°) and calculated (by the vector additivity method, μ_{I-E} = 2.12 D, μ_{I-Z} = 3.22 D) dipole moments.*

The introduction of a methyl group in the 2 position on passing to 2-methyl-4-R-phthalazone hydrazones III and V leads to a change in their PMR spectra as compared with the spectra of hydrazones I, II, and IV. The multiplet signal of the 5-7-H protons of hydrazones III and V is shifted somewhat to stronger field, whereas the signal of the 8-H proton is shifted appreciably to weaker field (Δδ 0.54 ppm) and approaches the position of the signal of the 8-H proton in the spectrum of hydrazine XVIII. The shift of the 8-H proton in the spectrum of dimethylhydrazone VI is especially significant even on comparison with hydrazone V (Δδ 0.91 ppm) (Fig. 2). Consequently, the introduction of a methyl group in the 2 position leads to change in the configuration of the hydrazone fragment, and hydrazones III, V, and VI exist in the Z form.

*See [19] for the bond moments.

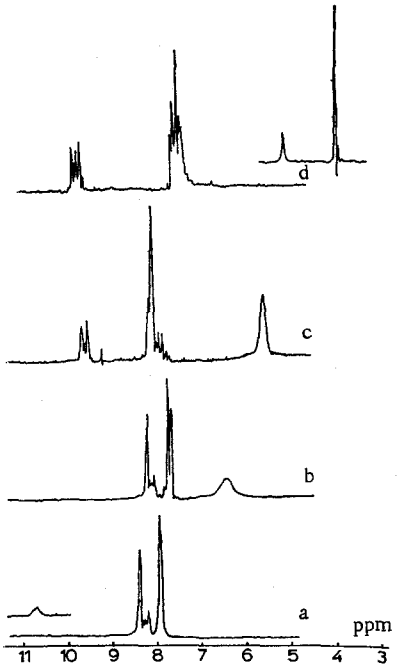


Fig. 2

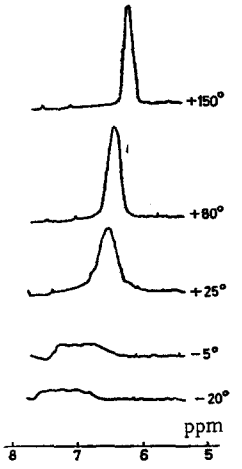
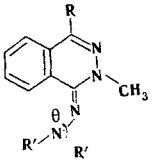


Fig. 3

Fig. 2. PMR spectra: a) VIII (in DMSO); b) I (in DMSO); c) XVIII (in DMSO); d) VI (in CCl_4) (the gain was reduced for the methyl groups).

Fig. 3. Signal of the protons of the NH groups of phthalazone hydrazones (I) in the PMR spectra in tetramethylurea at various temperatures.

TABLE 5. Calculated and Experimental Dipole Moments for the Z Form of Hydrazones III, V, and VI



Compound	μ calc., D			μ exptl., D
	θ=0 and 180°	θ=90°	θ=270°	
III	3,95	2,53	5,06	2,19
V	3,51	2,69	4,26	2,66
VI	2,98	2,42	3,42	2,57

An examination of Briegleb–Stuart models shows the impossibility of free rotation of the dimethylamino group about the N–N bond in hydrazone VI, and the presence of a singlet of two methyl groups in its PMR spectrum [$\delta(CH_3)_2$ 2.47 ppm] can therefore be considered to be an indication of the nonplanar structure of hydrazone VI. A comparison of the experimental and calculated (by the vector additivity method) dipole moments for dimethylhydrazone VI and hydrazones III and V confirms the assumption of ~90° rotation of about the N–N bond, during which the substituents attached to the amine nitrogen atom are spatially removed from the benzene ring (Table 5).

In the spectra of hydrazones I and IV, in addition to signals of aromatic protons, one should have expected two signals of protons of NH_2 and NH groups. In reality, only one singlet (3H) is observed in the spectra of hydrazones I and IV (Table 4). The low solubility of hydrazone IV in organic solvents made it impossible to make a sufficiently complete study of the dependence of the position of the signal of the NH proton on the nature of the solvent. The addition of water to solutions of hydrazone I leads to rapid broadening and merging of the signals of the protons of the NH groups with the signals of the protons of water

TABLE 6. Dependence of λ_{max} (nm) and log ε in the UV Spectra of Hydrazones I and IV on the Nature of the Solvent

Solvent	I	IV
DMSO	340 277	335 281
Dioxane	340 (3,66) 275 (4,15)	333 (3,85) 279 (4,11)
Isooctane	332 (3,48) 273 (3,70)	328 (3,04) 275 (3,38)
Acetonitrile	320 (3,68) 273 (4,11)	318 (3,88) 275 (3,90)
Methanol	318 (3,88) 273 (4,11)	318 (3,82) 274 (3,93)

[20]. An increase in the temperature leads to narrowing of the signal and shifting of it to stronger field, whereas a decrease in the temperature leads to broadening and a shift to weaker field (Fig. 3). The dependence of the position of the signal of the protons of the NH groups on the polarity of the solvent (Table 4), the temperature, and the addition of a proton donor (H_2O) provides evidence that an intermolecular proton exchange leading to merging of the signals of the protons of the NH_2 and NH groups occurs in solutions of hydrazone I.

The assumption of the intramolecular character of proton exchange in hydrazones I and IV with participation of the tautomeric hydrazine form (or of a dipolar structure) and the assumption of the possibility of their existence in a dimeric form with a 1,2,4,5-tetrazine ring [21] are not confirmed by data from the dependence of the UV spectra of hydrazones I and IV on the solvent (only one type of chromophoric system shows up, Table 6), IR spectroscopic data (the presence of three NH bands), and a study of the character of the PMR spectrum of hydrazone I at various temperatures.

EXPERIMENTAL

The UV spectra were recorded with a Specord UV-vis spectrophotometer. The IR spectra were recorded with a UR-20 spectrometer. The PMR spectra were recorded with RYa-2305 and T-60 spectrometers (60 MHz) with tetramethylsilane (TMS) as the internal standard. The chemical shifts were measured on the δ scale with an accuracy of up to 0.01 ppm. The melting points were determined with a Boetius microscope stage. The homogeneity of the substances was monitored in a thin layer of aluminum oxide by elution with petroleum ether–benzene or petroleum ether–dioxane and development with iodine vapors.

Compounds I, IV, and VIII-XVI were obtained by the methods in [22-28]. The synthesis of 2-methylphthalazone hydrazone (III) and 2-methyl-4-chlorophthalazone hydrazone (V) will be presented separately.

1,4-Dichloro-2-methylphthalazinium Methylsulfate. A 5-g (25 mmole) sample of 1,4-chlorophthalazine (XIV) was sprinkled into a mixture of 5 ml (0.04 mole) of dimethyl sulfate in 10 ml of chlorobenzene, after which the mixture was heated slowly until XIV dissolved completely, and the resulting mixture was allowed to stand at a temperature close to the boiling point for 3-5 min. It was then cooled at −10° for 5-7 h, and the resulting precipitate was removed by filtration, washed two to three times with cold benzene, and vacuum dried to give 3.2 g (36%) of a white crystalline powder.

2-Methyl-4-chlorophthalazone Imine (VII). A 3.2-g (0.01 mole) sample of 1,4-dichloro-2-methylphthalazinium methylsulfate was dissolved rapidly in 50 ml of water, and the solution was poured immediately with stirring into 50 ml of 30% ammonium hydroxide. The mixture was then allowed to stand at room temperature for 2-3 h, after which it was filtered to give 1.65 g (85%) of VII as a white crystalline powder with mp 100-104° (after two recrystallizations from 40% methanol). Found: C 55.7; H 3.9; Cl 18.0; N 21.6%. $C_9H_8ClN_3$. Calculated: C 55.8; H 4.1; Cl 18.3; N 21.7%.

2-Methylphthalazone Dimethylhydrazone (VI). A 3.2-g (0.01 mole) sample of 1,4-dichloro-2-methylphthalazinium methylsulfate was dissolved rapidly in 50 ml of water, and the solution was immediately poured into 20 ml of 70-80% aqueous N,N-dimethylhydrazine. The mixture was allowed to stand at room temperature for 1 h, and hydrazone VI was then extracted with chloroform. The solvent was removed from the extract by evaporation to dryness, and the residue was chromatographed [with aluminum oxide as the absorbent and petroleum ether–dioxane (1 : 1) as the eluent] to give 0.96 g (37%) of yellow needles of hydrazone VI with mp 67-70° (from ether). Found: C 55.9; H 5.4; Cl 15.2; N 23.6%. $C_{11}H_{13}ClN_4$. Calculated: C 55.8; H 5.5; Cl 15.0; N 23.7%.

1-(α-Methylhydrazino)-4-chlorophthalazine (XVIII). A 3-g sample of 1,4-dichlorophthalazine (XIV) was sprinkled into a hot mixture of 8 ml of methylhydrazine in 20 ml of ethanol, and the mixture was heated slowly until XIV dissolved completely. The mixture was then cooled slowly and allowed to stand at room

temperature for 2 h. The resulting voluminous precipitate was washed thoroughly with water to give 2.4 g (80%) of white needles of XVIII with mp 141-141.5° (from alcohol). Found: C 52.0; H 4.2; Cl 17.3; N 26.6%. $C_9H_9ClN_4$. Calculated: C 51.8; H 4.3; Cl 17.0; N 26.9%.

1-Chloro-4-dimethylaminophthalazine (XVII). A total of 10 ml of a 33% solution of dimethylamine was added to a hot solution of 1 g (5 mmole) of 1,4-dichlorophthalazine in 15 ml of DMF, and the mixture was heated moderately at 150-170° for 15 min. It was then cooled, 10 ml of 33% dimethylamine solution was added, and the mixture was heated at 150-170° for 15 min. The product was precipitated by the addition of water to give 0.93 g (85%) of white needles of XVII with mp 101° (from methanol). Found: C 57.3; H 4.7; Cl 17.3; N 20.2%. $C_{10}H_{10}ClN_3$. Calculated: C 57.8; H 4.8; Cl 17.1; N 20.2%.

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