EFFECT OF THE REACTION MEDIUM ON THE BROMOCYCLIZATION OF MALEIC AND FUMARIC ACID MONOUREIDES

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The monopotassium salt of maleic acid monoureide is brominated in water in the same way as the free acid, whereas the trans isomer is not brominated at all. Bromocyclization of the monopotassium salt of the cis isomer to give 5-(bromocarboxymethyl)hydantoin and intramolecular cyclization to give 5-(carboxymethyl)hydantoin are realized at pH 4-6. erythro-2,3-Dibromosuccinic acid and 5-(bromo-carboxymethyl)hydantoin are formed in the bromination of the monopotassium salt of fumaric acid monoureide at pH 4-6. Bromination of the methyl ester of maleic acid monoureide in 1,2-dichloroethane proceeds in the same way as bromination in water to give 2-amino-5-[bromo(methoxycarbonyl)methyl]oxazolid-4-one.

The bromination of maleic acid monoureide (Ia) and its methyl ester (Ib) in water leads to 2-imino-5-bromocarboxy(methoxycarbonyl)methyl-oxazolid-4-one (II) [1]. It is known [2, 3] that the direction of alkylation of ureides and amides, i.e., at the nitrogen or oxygen atom, depends on the reaction medium and the reactivity of the alkylating agent. Thus substituted N-propynylureas [2] undergo cyclization in acidic media to give oxazolidones, whereas they give imidazolidones in alkaline media.

The aim of the present research was to investigate the effect of the medium on bromination with intramolecular cyclization of the monopotassium salts of both maleic (IIIa) and fumaric (IIIb) acid monoureides and methyl ester Ib and to establish the structures of the products of these reactions.

The direction of addition of electrophilic reagents such as Br^+ in bromocyclization reactions [4] depends, in addition to the nucleophilicity of the donor part of the unsaturated molecule, on the electronic nature of the substituents attached to the C=C bond and the steric effect of these substituents on stabilization of the cyclic transition state. Little is known regarding the effect of acceptor groups attached to the C=C bond on the direction of cyclization [4].

To estimate the reactivity of the C=C bond we compared the $\Delta\delta$ differences in the chemical shifts [$\Delta\delta = \delta(C_3) - \delta(C_2)$] in the ^{13}C NMR spectra of monoureides Ia and Ic and monopotassium salts IIIa, b. It is apparent from a comparison of the difference in the shifts of free acids Ia, c and monopotassium salts IIIa, b (Table 1) that the polarization of the C=C bond in salts IIIa, b is considerably higher than in free acids Ia, c. It is therefore expected that electrophilic attack by Br^+ on C_2 will proceed more easily in the case of the monopotassium salts.

A sharp decrease in the pH of the medium occurs during bromination of monopotassium salts IIIa, b in water. As a consequence of this, oxazolidone IIa is obtained from cis-isomer IIIa, as in the bromination of free acid Ia [1]. When this reaction is realized at pH 4-6, the principal products of bromocyclization of cis isomer IIIa are 5-(bromocarboxymethyl)-hydantoin (IVa, b) and 5-(carboxymethyl)hydantoin (V). Only isomer IVa was isolated in free form; hypothetical compound IVb was isolated only in a mixture with isomer IVa.

It was demonstrated experimentally that monopotassium salt IIIa does not undergo cyclization to hydantoin V in weakly acidic and neutral media. This reaction has been realized in strongly alkaline media [6], in which detachment of a proton from the nitrogen atom facil-

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TABLE 1. Chemical Shifts of the C_2 and C_3 Atoms of Monoureides Ia,c and IIIa,b

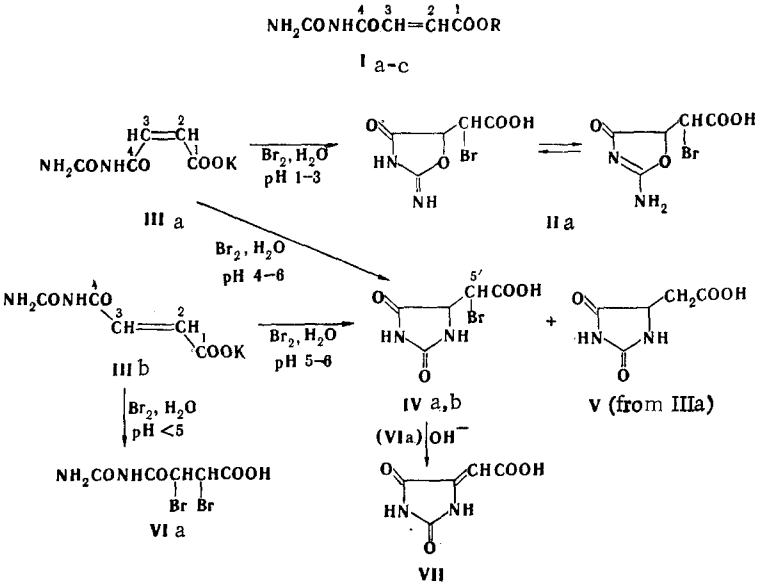
Compound	Solvent	δ, ppm		Δδ, ppm
		C_2	C_3	
Ia	d_6-DMSO*	130,8	132,7	1,5
Ic	d_6-DMSO	134,3	136,3	2,0
IIIa	D_2O	122,5	141,8	19,3
IIIb	D_2O	130,3	141,3	11,0

*Because of the low solubilities of free acids Ia,c in water, the data from the ^{13}C NMR spectra of their solutions in DMSO were used. See [5] for information regarding the effect of the solvent on the ^{13}C shifts.

itates attack of the relatively weak electrophilic center, viz., the C_3 atom, in the intramolecular alkylation of the nitrogen atom of the ureido group.

It may be assumed that the presence of bromine increases the polarization of the C=C bond of monoureide IIIa. Alkylation of the nitrogen atom of the ureido group is therefore also possible in weakly acidic and neutral media.

Because of the decrease in the pH of the medium during the reaction, bromination of trans isomer IIIb in aqueous solution leads to erythro-2,3-dibromosuccinic acid monoureide (VIa),* whereas the reaction does not take place at all in more acidic media because of a decrease in the reactivity of the C=C bond of free acid Ic (see Table 1). The absence of bromocyclization (formation of an oxazole ring) in weakly acidic media can be explained by structural factors and the reduced reactivity of the trans isomer.



Ia R=H (cis); b R=CH_3 (cis); c R=H (trans); IV a threo (or erythro); b erythro (or threo); VI a (erythro)

Bromination of trans isomer IIIb at pH 5-6 leads to hydantoins IVa,b or mixtures with dibromide VIa.

5-(Carboxymethylene)hydantoin (VII) is formed in the dehydrobromination of hydantoin IVa in alkaline media.

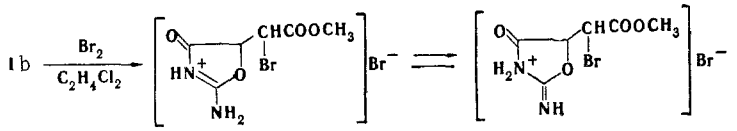
In contrast to the heterogeneous bromination of free acid Ia [1] in 1,2-dichlorethane, which gives dibromide VIb (threo),* the bromination of ester Ib in 1,2-dichloroethane takes place in the homogeneous phase. The reaction takes place in acidic media in the same way as

*The stereochemistry of dibromides VIa,b will be reported in a separate paper.

TABLE 2. PMR Spectra of Azolidones

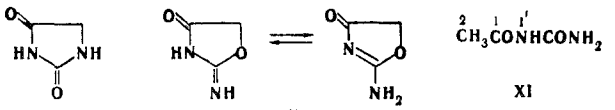
Compound	Solvent	δ, ppm					J, Hz	
		5-H	5'-H (d)	CH_3 (s)	N_3–H (br s)	N_1–H and =NH (br s)	$J_{55'}$	J_{51}
IIa [1]	d_6-DMSO	5,26 d	4,98	—	8,8	8,8	2,6	—
IIb [1]	d_6-DMSO	5,31 d	5,16	3,80	8,9	8,7	2,9	—
IIb	d_4-Methanol	5,30 d	4,98	3,81	—	—	2,9	—
IVa	d_6-DMSO	4,71 dd	4,91	—	10,9	8,2	2,9	1,5
IVb	d_6-DMSO	4,71 dd	5,03	—	11,0	8,2	3,1	1,5
VIII	d_4-Methanol	5,75 d	5,22	3,84	—	—	3,1	—
IX	d_6-DMSO	3,90 d	—	—	10,7	7,8	—	1,2
X [14]	d_6-DMSO	4,50 s	—	—	8,3	8,3	—	—
X	d_4-Methanol	4,64 s	—	—	—	—	—	—
XIIa	d_4-Methanol	5,75 d	5,22	3,84	—	—	2,9	—
XIIb	d_4-Methanol	5,04 s	—	—	—	—	—	—

in water and leads to 2-imino-5-[bromo(methoxycarbonyl)methyl]oxazolidin-4-one (VIII). Hydrobromide VIII is hydrolyzed in water to free base IIb. The presence of the bromide ion in VIII was confirmed by potentiometric titration.



VIII

The signals of the 5-H and 5'-H protons in the PMR spectrum of hydantoin IVa appear in the form of an AX system (Table 2), while the signals of the N–H protons show up in the form of two broad singlets. The assignment of the signals of the 5-H and 5'-H protons was made on the basis of a comparison with the spectra of model compounds, viz., oxazolidone IIa and unsubstituted heterocycles IX and X. The difference in the chemical shifts of the 5-H protons of hydantoin IVa and oxazolidone IIa ($\Delta\delta$ = 0.55 ppm, cf. Table 2) should be ascribed to the different electronegativities of the N_1 atom in the hydantoin and the O atom in the oxazolidone. The same difference in the shifts of the 5-H atoms ($\Delta\delta$ = 0.62 ppm) is also observed for the corresponding unsubstituted heterocycles IX and X. In addition, the low-field shifts of the chemical shifts of the 5-H protons of oxazolidone IIa and hydantoin IVa due to the effect of the substituent attached to C_5 are in good agreement.



IX X XI

The broad signals of the doublet of the 5-H proton of hydantoin IVa in the monoresonance spectrum correspond to the existence of vicinal spin–spin coupling of the 5-H and N_1–H protons (J_{51} = 1.5 Hz). This was confirmed by data from the nuclear magnetic double-resonance spectrum: the doublet signals of the 5-H proton become narrow when the N_1–H nucleus is irradiated. The existence of J_{51} excludes a cyclic oxazine structure, for which spin-spin coupling of the 5-H and N–H protons is impossible.

The signals of the vicinal 5-H and 5'-H protons appear in the PMR spectrum of mixture IVa,b in the form of two AX systems (IVa:IVb ≈ 1:1). The broad doublet with an intensity of two proton units at 4.71 ppm (Table 2) was assigned to the 5-H protons of both systems IVa and IVb, while the doublets at 4.91 and 5.03 ppm with an overall intensity of two proton units were assigned to the 5'-H protons of systems IVa and IVb, respectively. The broad two-proton singlet at 8.2 ppm was assigned to the N_1–H protons of IVa and IVb, while the two broad singlets at 10.9 and 11.0 ppm, each with an intensity of one proton unit, were assigned to the N_3–H protons of IVa and IVb, respectively.

According to [7], the relatively low $J_{55'}$ values of 2.9 (IVa) and 3.1 Hz (IVb) attest to the presence of gauche-5-H/5'-H conformers in both diastereomers. Because of the close SSCC of IVa and IVb, the assignment of the threo and erythro forms was not realized.

Three broad absorption bands of medium intensity at 1690-1790 cm^{-1}, which we assigned to the absorption of C_4=O and C_2=O stretching vibrations of hydantoin and a carboxy C=O group

*See footnote on previous page.

TABLE 3. Comparison of the Chemical Shifts of the Carbon Atoms in the ^{13}C NMR Spectra (d_6-DMSO) and the Frequencies of the Absorption of the C—CO—N and N—CO—N Groups in the IR Spectra of Monoureides Ia,c of Hydantoin IX and Ureide XI

Compound	C—CO—N		N—CO—N	
	δCO, ppm	νC=O, cm^{-1}	δCO, ppm	νC=O, cm^{-1}
Ia	167,2 or 168,1*	1712	154,8	1672
Ic	165,7 or 166,9	1720	154,8	1650
IX	175,9	1785 1760 (in Nujol); 1763 (in DMSO)†	159,8	1720 (in Nujol); 1797 (in DMSO)
XI	173,5	1750 1700	155,3	1700—1600

*The signals were assigned to the C—CO—N or COOH carbonyl carbon atom.

†Cf. Table 4.

are observed in the IR spectrum of hydantoin IVa in Nujol. The literature contains contradictory data relative to the interpretation of the C_4=O and C_2=O absorption bands in the IR spectra of hydantoins. Thus, in accordance with [8], the band in the low-frequency region (1690-1730 cm^{-1}) was assigned to C_2=O, while the band in the high-frequency region (1740-1790 cm^{-1}) was assigned to the C_4=O group of hydantoins. However, the enolization in the N_3—C_4=O grouping observed in alkaline media [9] enabled Seth and Demoen [10] to conclude that the C_4=O bond has increased polarity as compared with the C_2=O bond. The same conclusion was drawn in [11] during a study of the effect of the electronegativities of the adjacent atoms on the absorption of the C_2=O and C_4=O groups without allowance for the +M effect of the nitrogen atoms. In the present research we assigned the absorption bands of the C_2=O and C_4=O groups on the basis of a comparison of the data from the IR and ^{13}C-NMR spectra of hydantoin IX and noncyclic models with the N—CO—N fragment of acetic acid ureide (XI) and monoureides Ia,c.

The assignment of the signals of the C_4 atom of hydantoin IX and the C_1 atom of ureide XI (Table 3) was made on the basis of the data from the ^{13}C NMR spectrum without suppression of the spin—spin coupling with the protons.

A comparison of the C_2 and C_4 shifts (IX) and, respectively, C_1 and N—C—N shifts (XI) (Table 3) shows that the electron density on the C_4 (IX) and C_1 (XI) atoms is reduced. The reason for this is evidently rehybridization of the N_3 (IX) and N_1' (XI) atoms as a consequence of competitive conjugation of the C_2=O (IX) and NCON (XI) groups. This gives rise to a high-frequency shift of the absorption of the C_4=O (IX) and C_1=O (XI) groups as compared with 4-imidazolidone [10] and amides [12], respectively.

The IR spectrum of hydrobromide VIII contains an intense narrow band at ∿ 1800 cm^{-1} (Table 4). The same band is observed in the spectra of 4-oxazolidone hydrochlorides XIIa,b obtained from free bases IIb and X. This band was assigned to the stretching vibrations of the C_4=O group in conformity with the data in [12], according to which the hydrochlorides of amides, ureas, and similar molecules have a strong C=O band at ∿ 1800 cm^{-1}. The authors assume that this attests to the presence of an ammonium structure that is formed as a result of protonation of the nitrogen atom. This pronounced shift of $\nu_{C=O}$ to the high-frequency region is explained [12] by the absence of resonance of the carbonyl group with the nitrogen atom and the strong inductive effect of the H_3N^+ group. The C=O band of a 2-alkyl-4-oxo-(5H)-oxazolium salt is shifted even more markedly [13], which is also explained by a shift of the electron density from the carbonyl oxygen atom to the positively charged fragment of the molecule.

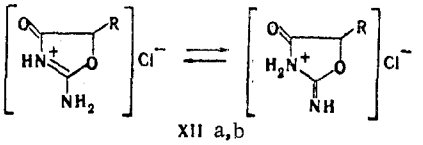
The presence of carboxy and ester C=O bands hinders the interpretation of the IR spectra of IIa,b, VIII, and XIIa. In addition, the presence of a bromine atom in the α

TABLE 4. IR Spectra of Azolidones*

Medium	Medium (concn., mole/liter)	Frequencies of the stretching and deformation vibrations, cm^{-1}				
		$\nu C_4=O$	Hydantoin $\nu C_2=O$ and oxazolidone $\nu C=N$ (exocyclic)	$\nu C_2=O$ Carboxy and ester	$\nu C=N$, δNH, $\nu C=N$ endocyclic	νNH, νOH, νN^+H_2 in hexachlorobutadiene
IVa	Nujol	1776 m 1762 m	1734 m, br	1704m, br	1650—1500	3300 m 3000—2500 br
IVa	Dioxane (saturated solution)	1784 vw	1743 vs	(1743)s†		
V	Nujol	1760— 1740 s,br	1720 (sh) s 1705 s	1690 s	1680—1550	3330 s 3200—2500 br
V	DMSO ($1{,}67\cdot10^{-2}$)	1769 vw	1729 vs	(1729) s		
VIII	Hexachlorobutadiene	1850 vw 1815 m	1710—1660 vs 1648 (sh) m	1747 s	1584 m	3400 w 3330, 3320 w 3200— 2800 m, br
IX	Nujol	1785 (sh) 1760 s, br	1720s, br	—	1700—1500	3330 (sh) s 3300—3100 s, br 3050 s
IX	Dioxane (saturated solution)	1773 vw	1737 vs			
IX	DMSO ($2{,}5\cdot10^{-2}$)	1763 w	1727 vs			
X	Nujol	1760 w 1717 s 1675 vs	1665 vs	—	1620—1500	3330 (sh)m 3300—2900 s, br
X	Dioxane (saturated solution)	1650 s	(1650) s, br	—	1570 w	
XIIa	Nujol	1835 vw 1795 m	1710— 1650 s, br	1735 s	1600 w	3330 w 3200—2700 s, br
XIIb	Nujol	1803 s 1775 m 1750— 1700 m, br	1700— 1650 s, br	—	1600 m	3330 w 3300—2700 s, br
XII	Methanol ($2\cdot10^{-2}$)	1750 m				

*Abbreviations: m is medium, br is broad, vw is very weak, s is strong, and sh is shoulder.
†Covered by the $C_4=O$ band.

position may additionally give rise to a shift of the bands of these groups due to the field effect. In accordance with [14], the presence of the 2-amino form is assumed for solutions of IIa,b, VIII, and XIIa,b on the basis of data from the IR spectra of a model compound, viz., oxazolidone X, whereas the presence also of a 2-imino form in the crystalline state is not excluded (cf. Table 4).



XII a,b

a R = $CHBrCOOCH_3$; b R = H

A low-field shift of the 5-H protons as compared with free bases X and IIb is observed in the PMR spectra of hydrohalides VIII and XIIa,b in methanol (cf. Table 2). The relatively low $J_{55'}$ values of 2.9 Hz for IIb, VIII, and XIIa attest to the existence of gauche-5-H/5'-H conformers.

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EXPERIMENTAL

The ^1H and ^{13}C NMR spectra of the compounds were recorded with a Bruker WH-90 spectrometer (90 and 22.63 MHz) with tert-butyl alcohol in water and methanol and cyclohexane in DMSO as the internal standards. The IR spectra of suspensions in Nujol and hexachlorobutadiene and of solutions in dioxane, DMSO, and methanol were obtained with Unicam SP 200G and Specord IR-75 spectrometers. The UV spectra were obtained with a Unicam SP 700A spectrophotometer.

Model compounds IX, X, and XIIb were synthesized in accordance with [15, 16].

2-Imino-5-(bromocarboxymethyl)oxazolid-4-one (IIa). A 1.60-g (0.01 mole) sample of bromine was added gradually with stirring to a suspension of 1.96 g (0.01 mole) of monopotassium salt IIIa in 10 ml of water, and the mixture was maintained at 15-20°C for 10 h. The precipitate was removed by filtration, washed with water, and dried in vacuo to give 2.13 g (90%) of oxazolidone IIa, which was identical* to the compound obtained from free acid Ia [1].

5-(Bromocarboxymethyl)hydantoin (IVa,b). A) A 4.0-g (0.025 mole) sample of bromine was added rapidly with stirring at 10-25°C to a suspension of 4.9 g (0.025 mole) of salt IIIa in 20 ml of water with simultaneous neutralization of the reaction mixture with an aqueous solution of 1.5-1.66 g of potassium hydroxide to pH 2-3. The precipitate was removed by filtration, washed with water, and dried in vacuo to give 0.2 g (3.3%) of hydantoin IVa with mp 140°C (dec.). UV spectrum (water), λ_{max}(log ε): 210 (inflection) (3.67) and 294 nm (3.42). Found: C 26.0; H 2.0; Br 34.1; N 12.1%. $C_5H_5BrN_2O_4$. Calculated: C 25.3; H 2.1; Br 33.7: N 11.8%.

The filtrate after separation of hydantoin IVa was evaporated at 10-15°C (1 mm), and the residue was dried in vacuo and suspended in 20 ml of methanol. The mixture was filtered, and the filtrate was evaporated at −5 to 0°C (1 mm). The residue was suspended in water, and the solid material was removed by filtration and dried in vacuo to give 0.85 g (14%) of a mixture of IVa and IVb.

B) A 4.9-g (0.025 mole) sample of salt IIIb was brominated by method A. The acidified mixture was filtered, and the precipitate was washed with water and dried. Extraction of the mixture with ethanol yielded 0.2 g (2.5%) of dibromide VIa with mp 172-173°C. IR spectrum (in Nujol and hexachlorobutadiene)): 3370, 3310, 3300-3100 (NH), 1738, 1719, 1701, 1664 (C=O), 1592 cm^{-1} (NH, C-N). UV spectrum (in water), λ_{max}(log ε): 210 (inflection) (4.05) and 240 nm (inflection) (3.28). PMR spectrum (in DMSO): 10.9 (1H, s, NH), 7.7 (2H, s, NH_2), 5.14 (1H, d, 2-H, J = 11.2 (Hz), 4.71 ppm (1H, d, 3-H). Found: C 19.2; H 1.9; Br 50.5; N 8.9%. $C_5H_6Br_2N_2O_4$. Calculated: C 18.9, H 1.9, Br 50.3, N 8.8%.

The filtrate after separation of dibromide VIa was evaporated at 10-15°C (1 mm), and the residue on the filter was washed with water and dried in vacuo to give 0.69 g (12%) of hydantoin IVa, which was identical to the compound obtained from cis isomer IIIa.

5-(Carboxymethylene)hydantoin (VII). A solution of 0.04 g (0.17 mmole) of hydantoin IVa in 0.1 ml of a 40% aqueous solution of potassium hydroxide was acidified with hydrochloric acid to pH 2, and the precipitate was removed by filtration, washed with water, and dried to give 0.02 g (76%) of a product that was identical to the compound obtained in [17].

5-(Carboxymethyl)hydantoin (V). A 4.0-g (0.025 mole) sample of bromine was added gradually with stirring at pH 4-6 to a suspension of 4.9 g (0.025 mole) of monopotassium salt IIIa in 50 ml of water. After 1 h, the water was removed by distillation at 10-15°C (1 mm). The residue was suspended repeatedly in small amounts of water. Evaporation of the second filtrate gave a mixture, the principal component of which was hydantoin V, which was identical to the compound obtianed in [6].

*Here and subsequently, the identity of the compounds was proved by the PMR spectra.

2-Imino-5-[bromo(methoxycarbonyl)methyl]oxazolid-4-one Hydrobromide (VIII). A 6.88-g (0.04 mole) sample of methyl ester Ib was suspended in 40 ml of 1,2-dichloroethane, and 6.4 g (0.04 mole) of bromine was added with stirring. The mixture was then stirred at 15-20°C for 8 h, during which starting ester Ib dissolved gradually, and the hydrobromide began to precipitate. The precipitate was removed by filtration, washed with absolute ether, and dried in vacuo to give 11.95 g (90%) of hydrobromide VIII with mp 150-152°C. UV spectrum (in ethanol): λ_{max} 220 nm (log ε 4.29). Found: C 22.1; H 2.4; Br 47.9; N 8.6%. $C_6H_7BrN_2O_4 \cdot HBr$. Calculated: C 21.7, H 2.4, Br 48.1, N 8.4%.

2-Imino-5-[bromo(methoxycarbonyl)methyl]oxazolid-4-one (IIb). A 5.5-g (17 mmole) sample of hydrobromide VIII was suspended in 20 ml of water, and the mixture was stirred for 15-30 min. The precipitate was removed by filtration, washed to neutrality with water, and dried in vacuo to give 3.9 g (94%) of oxazolidone IIb, which was identical to the compound obtained in [1]. Hydrochloride XIIa. A 0.25-g (1.0 mmole) sample of ester IIb was added with stirring to 0.5 ml of cooled concentrated hydrochloric acid, after which the hydrochloric acid was removed by distillation at 0-5°C (< 1 mm) and dried in vacuo to give 0.25 g (87%) of hydrochloride XIIa with mp 152-154°C (dec.). UV spectrum (in ethanol): λ_{max} 220 nm (log ε 4.30). Found: C 25.3; H 2.9; N 10.0; Hal (for Cl) 24.2%. $C_6H_7BrN_2O_4 \cdot HCl$. Calculated: C 25.1; H 2.8; N 9.7; Hal (for Cl) 24.7%.

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