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Several 3-Ω-amino monosubstituted hydantoins have been obtained in the reaction of isocyanate of glycine ethyl ester with the appropriate aliphatic or aromatic diamine. It was found, that the 3-(aminoaryl) monosubstituted hydantoins may be diazotized and their diazonium salts may be coupled and hydrolysed without changes in the hydantoin ring.

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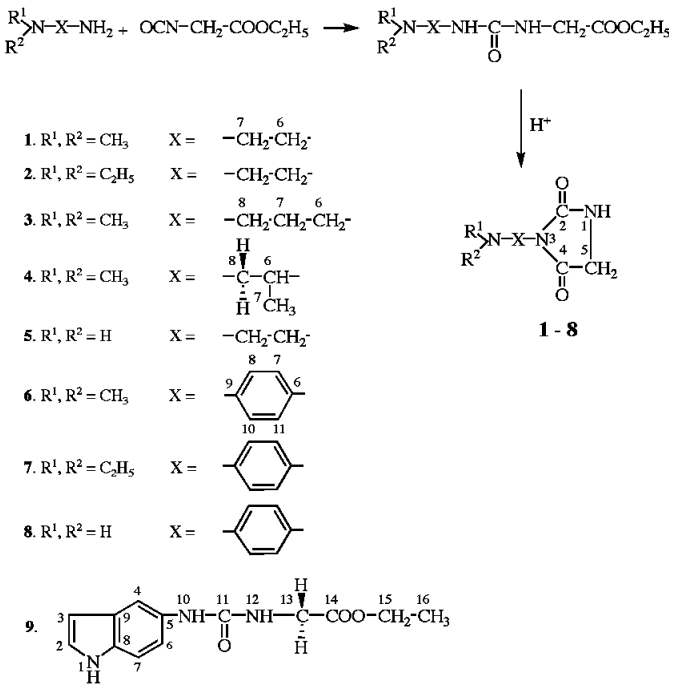
Derivatives of hydantoin have different pharmacological activity [1] and technical applications [2]. It was found that differently substituted hydantoins with amino and dialkylamino groups are active against central nervous system [3,4] and cardiovascular system [5] disorders. There have been few 3-monosubstituted dialkylaminoalkyl hydantoins obtained till now and their synthesis offers rather limited possibilities [6]. 3-Monosubstituted hydantoins with the free amine group with the substituent at the 3 position are even more scarce and their syntheses have low yield [7] or is protected by patents [8,9].

Results and Discussion.

In this paper 3-monosubstituted amino and dialkylamino alkyl and aryl hydantoins were synthesized according to the known method [10] and its applications [11]. It involves the reaction between the appropriate diamine and the isocyanate of glycine ethyl ester (Scheme 1). The reaction was carried out at room temperature or alternatively at 0 °C. Chloroform or anhydrous ethanol was used as solvents due to the solubility of the substrates. After the reaction, the unisolated intermediate, *i.e.*, a derivative of ethyl ester of hydantoic acid, was cyclised under acidic (HCl) conditions to the appropriate hydantoin in water-ethanol solution. The raw product obtained was purified as a hydrochloride or, after transformation, as a free base. For the synthesis of derivatives with a free amino group a monoacetyl derivative of the diamine was used as substrate. It was found that when a monoalkyldiamine, *i.e.* *N*-methyl-1,3-propanediamine, was used the reaction did not follow the proposed scheme (Scheme 1). Immediately after mixing of reagents a mixture of various products was obtained, evidenced by monitoring the reaction in $CDCl_3$ solution by ^1H NMR spectroscopy. After careful evaporation of the solvent and drying under reduced pressure at 40 °C the intermediate was hydrolysed in HCl medium. A mixture of different products was obtained and among them *ca*. 10% of *N*-methyl-1,3-propanediamine hydrochloride, the presence of which (confirmed by ^1H NMR, ^{13}C NMR and MS) may mean that during the mentioned reaction a partial amidation of the ester group of the isocyanate occured. To obtain the indole derivative of 3-monosubstituted hydantoin, 5-aminoindole and isocyanate of glycine ethyl ester were used (Scheme 1). The indole derivative of the ester of hydantoic acid **9** was obtained with satisfactory yield but attempts to cyclise it to the hydantoin derivative proved unsuccessful although HCl, CH_3COOH and heating without a solvent were tried. In each case the final result was a mixture of different, unidentified substances.

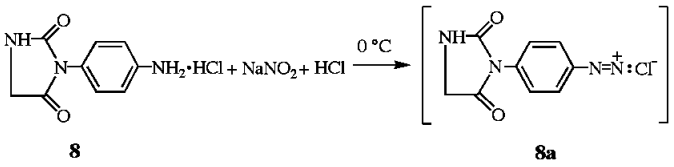
Scheme 1



Compounds **1**-**3**, **5**, **6** and **8** are hydrochlorides

One of the major aims of this work was to determine whether the free amine group in the 3-monoaminoaryl substituent reacts without simultaneous changes in the hydantoin ring. The reactions were carried out with 3-(4'-aminophenyl)hydantoin **8**, which was transformed in the non-isolated diazonium salt (**8a**) (Scheme 2) at 0 °C without a change of the NH group of the hydantoin ring in the nitroso group [12]. This salt was remarkably stable and could be heated at 80 °C for several hours without decomposition but after 5 hours heating at 100 °C it was hydrolysed with 45% yield to 3-(4'-hydroxyphenyl)hydantoin **10** (Scheme 3), identical with the specimen obtained by another method [11]. The diazonium salt was also coupled

Scheme 2

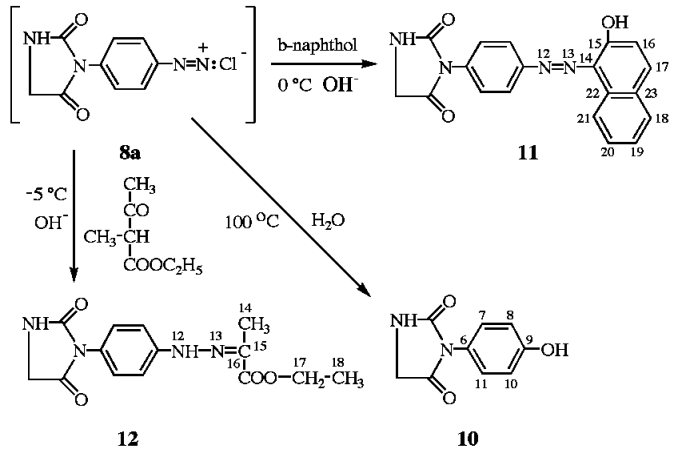


8 **8a**

with β-naphthol in alkaline medium (Scheme 3) yielding an azo dye **11** with indicator properties.

The strong alkaline medium of the reaction did not cause opening of the hydantoin ring, which is usual at this pH. This allowed the Japp-Klingeman reaction with ethyl 2-methylacetoacetate to occur, leading to phenylhydrazone derivative **12** (Scheme 3). This was treated with HCl in anhydrous ethanol and BF_3 in CH_3COOH. In the first case the substance remained unchanged, whereas treatment with BF_3 resulted in a mixture of different products.

Scheme 3



12 **10**

The physicochemical data of compounds **1-12** are presented in Table 1. Table 2 reports spectral (IR, ^1H and ^{13}C NMR) data, which unambiguously confirmed the structure of the compounds obtained. A comparison of ^{13}C NMR spectra of hydrochloride **6** and free amine **6a** revealed broad signals for some carbon atoms of the phenyl moiety and $N(CH_3)_2$ group in this first compound but not in the second, whereas a significant shift of δ_C values of these atoms was observed in this last compound. This phenomenon may be associated with the lack of inversion of configuration at the nitrogen atom in hydrochloride **6**.

EXPERIMENTAL

Mps were uncorrected and measured on a Boetius apparatus. CHN analyses were acquired on a Perkin Elmer 2400 analyzer. IR spectra were recorded on a Zeiss Specord 75 IR apparatus in KBr discs. ^1H and ^{13}C NMR spectra were obtained on a Bruker 500 apparatus in DMSO d_6 and $CDCl_3$ with TMS as an internal standard. Spin-spin coupling constants are given in Hz and those of diastereotopic hydrogen atoms were theoretically calculated using a computer program (written by Dr. E Szneler from the NMR Laboratory of the Jagiellonian University). For compound **10** the coupling between aromatic protons was interpreted from the H^1, H^1COSY spectra and chemical shifts of carbon atoms were ascribed from HETCOR and HMBC spectra. The interpretation of ^1H and ^{13}C NMR spectra was supported by the program gNMR V3.6. Mass spectra were obtained on a Varian MAT-112 spectrometer at 70 eV. The reagents were from Aldrich (Poznan, Poland).

Table 1

Preparative, Physical and Analytical Data for the Compounds **1-12**

Compound (Formula)	Yield (%)	Mp/°C	Recrystn. solvent	Found /Calcd. C	H	N	m/z $M^{+\cdot}$
1	99.5	238-9	EtOH	40.4	6.8	20.2	171
$C_7H_{13}N_3O_2$•HCl				40.1	6.8	19.9	
2	99.7	157-8	EtOH	45.9	7.7	17.8	199
$C_9H_{17}N_3O_2$•HCl				45.8	7.7	17.8	
3 [6]	99.7	206-7	EtOH	43.3	7.3	18.9	185
$C_8H_{15}N_3O_2$•HCl				43.1	7.1	18.6	
4	52.4	119-20	Cyclohexane	51.9	8.2	22.7	185
$C_8H_{15}N_3O_2$				51.8	8.2	22.6	
5	99.5	205-7	EtOH	33.4	5.6	23.4	143
$C_5H_9N_3O_2$•HCl				33.4	5.6	23.2	
6	93.4	220-21	EtOH	51.7	5.5	16.4	219
$C_{11}H_{13}N_3O_2$•HCl				51.4	5.5	16.1	
6a	59.9	220-21	EtOH				
$C_{11}H_{13}N_3O_2$·			+benzene				
7	53.3	188-9	EtOH	63.1	6.9	17.0	247
$C_{13}H_{17}N_3O_2$			+benzene	63.0	6.9	16.7	
8	99.8	238-9	EtOH	47.5	4.4	18.4	191
$C_9H_9N_3O_2$•HCl				47.4	4.4	18.3	
9	99.3	173-4	EtOH	59.8	5,8	16.1	261
$C_{13}H_{15}N_3O_3$				59.5	5.6	15.8	
10 [11]	45.3	291-2	EtOH	56.2	4.2	14.6	192
$C_9H_8N_2O_2$				56.0	4.1	14.6	
11	86.2	266-7	AcOH	65.9	4,1	16.2	346
$C_{19}H_{14}N_4O_3$				65.6	3.9	15.9	
12	35.2	286-7	AcOH	55.2	5.3	18.4	304
$C_{14}H_{16}N_4O_4$				55.0	5.3	18.1	

General Procedure.

To a solution of diamine (1.00 g) in 25 cm^3 of chloroform or monoacetyldiamine in 25 cm^3 of anhydrous ethanol an equimolar amount of isocyanate of glycine ethyl ester in 25 cm^3 of chloroform (for **1-4, 6** and **7**) or 25 cm^3 of anhydrous ethanol was added dropwise with constant stirring. The mixture was stirred for 2 hours and the solvent was evaporated on a rotary evaporator under reduced pressure and thoroughly dried at 40 °C under reduced pressure. The residue was dissolved in 25 cm^3 of ethanol and 25 cm^3 of 5 *M* HCl was added. The mixture was refluxed for 3 hours then the solvent was evaporated and the raw product was crystallized from the appropriate solvent (Table 1).

Free Base for Hydantoins with a Phenylamine Group (**6a** and **7**).

The of hydrochloride of compound **6** (0.5 g, 1.95 mmol) was dissolved in 5 cm^3 of water and conc. ammonia solution was added dropwise to alkaline reaction. The separated precipitate was isolated by filtration and washed with water until neutral. The precipitate was dried and crystallized from the mixture of ethanol and benzene. Yield 0.257 g (59.9%) of **6a**.

Table 2

IR and ^{1}HNMR and ^{13}CNMR Spectroscopic Data for the Compounds **1-12**

Compound	ν_{max} (KBr) cm^{-1}	δ_H[500 MHz DMSO d_6; TMS]	δ_C[125 MHz DMSO d_6; TMS]
1	3295 (NH) 1785, 1720 (CO)	2.75 (s, 6H, N(C*H*$_3$)$_2$), 3.24 (t, 2H, J=5.5 Hz, C(6)H$_2$), 3.68 (t, 2H, J=5.5Hz, C(7)H$_2$), 3.88 (s, 2H, C(5)H$_2$), 8,22 (br s, 1H, N(1)H)	36.79 (C(6)H$_2$), 45.49- (N(*C*H$_3$)$_2$), 46.83 (C(5)H$_2$), 57.16 (C(7)H$_2$), 158.68 (C(2)O), 172.45 (C(4)O)
2	3325 (NH) 1730, 1695 (CO)	1.19 (t, 6H, J=7.1Hz, N(C*H*$_3$-CH$_2$)$_2$), 3.13 (m, 4H, N(CH$_3$-C*H*$_2$)$_2$), 3.18 (m, 2H C(6)H$_2$), 3.69 (t, 2H, J=6.4 Hz, C(7)H$_2$), 3.88 (s, 2H, C(5)H$_2$), 8,22 (br s, H, N(1)H)	8.04 (N(*C*H$_3$-CH$_2$)$_2$), 32.35 (C(6)H$_2$), 45.81 (N(CH$_3$-*C*H$_2$)$_2$), 47.23 (C(5)H$_2$), 47.55 (C(7)H$_2$), 157.75 (C(2)O), 172.30 (C(4)O)
3 [6]	3120 (NH) 1730, 1655 (CO)	1.88 (m, 2H, C(7)H$_2$), 2.67 (s, 6H, N(C*H*$_3$)$_2$), 3.00 (2H, t, 2H, J=8.00 Hz, C(6)H$_2$), 3.37 (2H, m, 2H, C(8)H$_2$), 3.88 (s, 2H, C(5)H$_2$), 8.15 (1H, br s, 1H, N(1)H)	22.96 (C(7)H$_2$), 34.85 (C(6)H$_2$), 41.93 (N(*C*H$_3$)$_2$), 45.95 (C(5)H$_2$), 54.05 (C(8)H$_2$), 157.42 (C(2)O), 172.17 (C(4)O)
4	3045 (NH) 1745, 1690 (CO)	1.36 (d, 3H , J= 7.0 Hz, C(7)H$_3$), 2.18 (q, 1H, J=4.8 Hz, (C(8)H^x, C(6)H), J=12.5 Hz, ((C(8)H^x, C(8)H^y), (C(8)H^{x}H^y)), 2.22 (s, 6H, N(C*H*$_3$)$_2$), 3.15 (q, 1H, J=10.7 Hz, (C(8)H^y, C(6)H), J= 12.5 Hz, ((C(8)H^x, C(8)H^y), (C(8)H^{x}H^y)), 4.29*(m, 1H, J=7.0 Hz, (C(7)H$_3$, C(6)H), J=4.8 Hz, (C(8)H^x, C(6)H), J=10.7 Hz, (C(8)H^y, C(6)H), 3.91(s, 2H, C(5)H$_2$), 6.53(br s, 1H, N(1)H)	16.13 (C(6)H$_3$), 45.22 (C(7)H), 45.55 (N(*C*H$_3$)$_2$), 46.13 (C(5)H$_2$), 61.20 (C(8)H$_2$), 158.70 (C(2)O), 171. 77 (C(4)O)
5	3200, 3140 (NH$_2$, NH) 1745, 1660 (CO)	2.49(t, 2H, J= 6.0 Hz, C(7)H$_2$), 3.59 (t, 2H J= 6.0 Hz, C(6)H$_2$), 3.85(s, 2H C(5)H$_2$), 8.18(d, 2H, N(1)H, NH$_2$)	35.43 (C(7)H$_2$), 36.93 (C(6)H$_2$), 46.16 (C(5)H$_2$), 157.32 (C(2)O), 172.40 (C(4)O)
6	3250 (NH) 1760, 1700 (CO)	3.08(s, 6H, N(C*H*$_3$)$_2$), 4.05(s, 2H, C(5)H$_2$), 7.46(d, 2H, J= 8.5 Hz, C(8,10)H), 7.72(d, 2H, J= 8.5, Hz, C(7, 11)H), 8.42(br s, 1H, N(1)H)	br.43.61 (N(*C*H$_3$)$_2$, 46.02 (C(5)H$_2$), br.118.03 (C(8,10)H), 127.79 (C(7,11)H) and C(6), br.144.98 C(9), 156.38 (C(2)O), 171.06 (C(4)O)
6a	3220 (NH) 1750, 1745 (CO)	2.98(s, 6H, N(C*H*$_3$)$_2$), 4.05(s, 2H, C(5)H$_2$), 6.65(br s, 1H, N(1)H), 6.76(d, 2H, J= 9.0 Hz, C(8,10)H), 7.18(d, 2H, J= 9.0 Hz, C(7, 11)H),	40.46 (N(*C*H$_3$)$_2$), 46.46 (C(5)H$_2$), 112.54 (C(8,10)H), 119.59 C(6), 127.17 (C(7,11)H), 150.46 C(9), 158.38 (C(2)O), 170.77 (C(4)O)
7	3305 (NH) 1755, 1690 (NH)	1.08(t, 6H, J= 7.0 Hz, N(C*H*$_3$-CH$_2$)$_2$), 3.33(q, 4H, J= 7.0 Hz, N(CH$_3$-C*H*$_2$)$_2$), 4.00(s, 2H C(5)H$_2$), 6.66(d, 2H, J= 8.7 Hz, C(8,10)H), 7.02 (2H, d, 2H, J= 8.7 Hz, C(7, 11)H), 8.14(1H, br s, 1H, N(1)H)	12.34 (N(*C*H$_3$-CH$_2$)$_2$), 43.75 (N(CH$_3$-*C*H$_2$)$_2$), 45.87 (C(5)H$_2$), 111.00 (C(8,10)H, 119.50 C(6), 127,89 (C(7,11)H), 146.78 C(9), 157.19 (C(2)O), 171.48 (C(4)O)
8	3420, 3340, 3280 (NH$_2$, NH) 1770, 1690 (CO)	4.04(s, 2H, C(5)H$_2$), 7.35(m, 4H, J= 8.6 Hz, C(7,8,10,11)H), 8.36(s, 1H, N(1)H), 10.30(br s, 2H, NH$_2$)	45.99 (C(5)H$_2$), 122.10 (C(8,10)H), 127.97 (C(7,11)H), 129.99 C(6), 133.72 C(9), 156.27 (C(2)O), 170.99 (C(4)O)
9	3260 (NH) 1730 (CO), 1575 (CO ester)	1.21(t, 3H, J= 7.1 Hz, C(16)H$_3$), 3.86(d, 2H, J= 5.9 Hz, C(13)H$_2$), 4.11(q, 2H, J= 7.1 Hz, C(15)H$_2$), 6.31(m, 2H, C(2,3)H), 7.02(d, 1H, C(7)H), 7,26(m, 2H, C(4,6)H), 7.61(br s, 1H,N (12)H), 8.48(br s, 1H, N(10)H), 10.99(br s, 1H, N(1)H)	14.14 (C(16)H$_3$), 41.46 (C(13)H$_2$), 60.29 (C(15)H$_2$), 100.78, 109.36, 109.67, 111.16, 114.48, 126.31, 127.70, 132.00, (C(2)H, C(3)H, C(4)H, C(5), C(6)H, C(7)H, C(8), C(9)), 155.73 (C(14)O), 171.11 (C(11)O)
10 [11]	3420 (OH), 3240 (NH) 1760, 1673 (CO)	4.02(s, 2H, C(5)H$_2$), 6.82(d, 2H, J= 8.8 Hz, C(8,10)H), 7.09(d, 2H, J= 8.8 Hz, C(7,11)H), 8.17(br s, 1H, N(1)H), 9.65(s, 1H, OH(9)**)	45.89 (C(5)H$_2$), 115.21 (C(8,10)H), 123.30 C(6), 128.13 (C(7,11)H), 156.85, 156.89 C(9) and (C(2)O), 171.28 (C(4)O)
11	3220 (OH), 3100 (NH) 1750, 1700 (CO)	4.11(s, 2H, C(5)H$_2$), 6.92(d, 1H, J= 9.0 Hz, C(16)H), 7.47(m, 1H, J= 7.0 Hz and J= 8.0 Hz C(19)H), 7.55(d, 2H, J= 8.5 Hz, C(7,11)H), 7.62(m, 1H, J= 7.0 Hz and J= 8.0 Hz C(20)H), 7.79(d, 1H, J= 8.0 Hz, C(18)H), 7.96(d,2H, J= 9.0 Hz, C(8,10)H), 7.96(d, 1H, J= 9.0 Hz, C(17)H), 8.56(d, 1H, J= 8.0 Hz, C(21)H), 8.42(br s, 1H N(1)H)	46.07 (C(5)H$_2$), 119.03 (C(8)H and C(10)H), 121,45 (C(21)H), 124.01 (C(16)H), 126.01 (C(19)H), 127,73 (C(7)H and C(11)H), 127.90 (C(20)H), 128.96 (C(18)H), 129.39 C(15), 140.27 (C(17)H), 129.16, 132.71 (C(14), C(22) and C(23)), 131.53, 143.93 (C(9) and C(6)), 156.31 (C(2)O), 171.06 (C(4)O)
12	3295, 3100 (NH) 1750, 1700 (CO), 1560 (CO ester)	1.27(t, 3H, J= 7.0 Hz, C(18)H$_3$), 2.08(s, 3H, C(14)H$_3$), 4.04(s, 2H, C(5)H$_2$), 4.20(q, 2H, J= 7.0 Hz, C(17)H$_2$), 7.22(d, 2H, J= 8.7 Hz, C(8,10)H), 7.32(d, 2H, J= 8.7 Hz, C(7,11)H), 8.25(br s, 1H, N(1)H), 9.97(br s, 1H, N(12)H)	11.82 (C(18)H$_3$), 14.18 (C(14)H$_3$), 45.87 (C(5)H$_2$), 60.22 (C(17)H$_2$), 113.50 (C(8,10)H), 125.03 C(9), 127.55 (C(7,11)H), 132.60 C(6), 143.74 C(15), 156.72 (C(2)O), 164.76 (C(16)O), 171.15 (C(4)O)

Compounds **4** and **6a** were measured in $CDCl_3$; * J values for this multiplet were calculated; ** after addition of D_2O the signal at 9,65 ppm disappeared.

Free base for Hydantoin with a Dialkylaminoalkyl Group (**4**).

The hydrochloride of compound **4** (1.0 g, 4.7 mmol) was dissolved in 25 cm^3 of ethanol and an equimolar amount of piperidine (0.4 g) in 25 cm^3 of ethanol was added. The solvent was evaporated and the residue was dried. The precipitate was boiled twice with 20 cm^3 of benzene and filtered. The combined filtrates were evaporated and the residue was crystallized from cyclohexane. Yield 0.82 g (98.2%) of **4**.

4-[3'-(2',4'-Imidazolidinedione)]benzenediazonium Chloride (**8a**).

To a solution of 2.00 g (8.7 mmol) of 3-(4'-aminophenyl)-hydantoin hydrochloride **8** in 50 cm^3 of water, 1.7 cm^3 of conc. HCl was added and the mixture was chilled to 0 °C. Then 0.605 g (8.8 mmol) of $NaNO_2$ in 1.7 cm^3 of water was added dropwise during 45 minutes under constant stirring and keeping the temperature at 0 °C. The stirring was continued for 15 minutes after addition of the nitrite.

3-(4'-Hydroxyphenyl)-2,4-imidazolidinedione (**10**).

To the obtained diazonium salt (above) 30 cm^3 of water was added and heated under reflux for 5 hours at 100 °C. The reaction course was monitored by the coupling reaction with β-naphthol until no red azo dye formation was observed. After completion of reaction the solution was evaporated under reduced pressure and the residue was dried. The precipitate was boiled twice with 25 cm^3 of acetone and filtered. Acetone was evaporated from the combined filtrates and the residue was crystallized from ethanol yielding 0.765 g (45.3%) of **10**.

2-Hydroxynaphthelene-1-azo[4-(3-imidazolidine-2,4-dione)]-benzene (**11**).

To the diazonium salt obtained from 2.00 g of compound **8** (above) chilled to 0 °C a solution of equimolar amount of β-naphthol (1.266 g) in 6.3 cm^3 of 10% NaOH was added with stirring. A precipitate was formed immediately. Then 50 cm^3 of water was added and stirring was continued for 0.5 hour. The mixture was filtered and washed with water till neutral. The precipiate was dried and crystallized from glacial acetic acid yielding 2.625 g (86.2%) of **11**.

2-[4'-(3"-(2",4"-Imidazolidinedione))phenylhydrazone]ethyl Propionate (**12**)

Ethyl 2-methylacetoacetate (1.27 g, 8.8 mmol) was dissolved in 9.4 cm^3 of anhydrous ethanol and chilled to –5 °C. At that temperature slowly a cold solution of 3.11 cm^3 of 50% KOH was added dropwise with stirring. The solution was added to the diazonium salt prepared from 2.00 g of amine **8** (above). Then 20 cm^3 of water was added and the mixture stirred for 1 hour. The separated precipitate was isolated by filtration, washed with water till neutral, dried and crystallized from glacial acetic acid yielding 0.94 g (35.2%) of **12**.

Acknowledgements.

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