RING FORMATION DURING THE REACTION OF AROMATIC DIAMINES WITH ACETOACETIC ACID ESTERS

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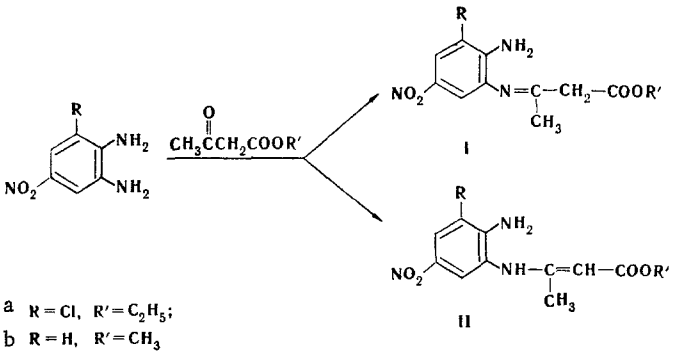
It is shown that the reaction of aromatic o-diamines having a nitro group in the para position relative to one of the amino groups with acetoacetic esters under mild conditions gives arylaminocrotonates, which are isomerized to ketimines under the influence of acid, are cyclized to dihydro-1,5-benzodiazepinones on heating, and are converted to 1-isopropenylbenzimidazolones under more severe conditions. If the condensation of the nitrodiamine with the acetoacetic acid esters is carried out under severe conditions, the isomeric dihydro-1,5-benzodiazepinone is obtained. Splitting out of an alkylacetate ester to give the corresponding 2-methylbenzimidazole occurs in the condensation of various o-diamines with 2-alkylacetoacetic esters.

The condensation of aromatic or heteroaromatic o-diamines with acetoacetic acid esters is one of the principal methods for the synthesis of 1,5-benzodiazepinones [1]. However, this is such a complex process that one cannot always predict the structure of the product obtained. Depending on the conditions, one can isolate the corresponding 3-arylaminocrotonates, dihydro-1,5-benzo-2-diazepinones, 2-methylbenzimidazoles, and 1-isopropenyl-2-benzimidazolones. In addition, instances of migration of the double bond and formation of isomeric benzodiazepinones have been observed [2-4].

In the case of an unsymmetrical diamine, the reaction is complicated by the alternative possibility of primary attack at one or another nitrogen atom. Thus 2,3-diaminopyridine reacts with the C=O group with its more basic amino group on refluxing with acetoacetic ester [4]. However, in the case of 3,4-diaminopyridine the less-basic amino group undergoes condensation with the carbonyl group [5]. We therefore made a more systematic study of this process.

It was found that, as was previously shown for ethyl acetoacetate [6, 7], various acetoacetic acid esters react with 4-nitro- and 3-chloro-5-nitro-1,2-phenylenediamines at room temperature to give ketimines (I). Crotonates (II) are formed rapidly at 35-40° under somewhat modified conditions.



a R = Cl, R' = C_2H_5;
b R = H, R' = CH_3

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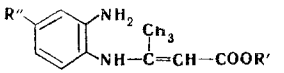
TABLE 1. 3-(2-Amino-4-R''-phenylamino)crotonates 

Compound	R′	R″	mp, °C (crystallization solvent)	Reagent ratio (diamine: AAA ester, mole)*	Empirical formula	Found, %			Calc., %			Yield, %
						C	H	N	C	H	N	
IIIa	CH_3	H	80—81 (hexane)	0,01 : 0,017	$C_{11}H_{14}N_2O_2$	64,4	6,8	14,0	64,2	6,8	13,7	81,1
IIIb	CH_3	CH_3	84—85 (pet. ether 40-70°)	0,01 : 0,014	$C_{12}H_{16}N_2O_2$	65,7	7,2	12,7	65,6	7,3	12,7	91,0
IIIc	CH_3	Cl	121—122 (hexane)	0,01 : 0,014	$C_{11}H_{13}ClN_2O_2$	55,1	5,6	11,5	54,9	5,4	11,6	73,0
IIId	CH_3	Br	127—128 (hexane)	0,01 : 0,014	$C_{11}H_{13}BrN_2O_2$	46,6	4,6	10,0	46,3	4,5	9,8	70,8
IIIe	n-C_3H_7	H	47—48 (pet. ether 40-70°)	0,01 : 0,012	$C_{13}H_{18}N_2O_2$	66,7	7,6	12,0	66,6	7,7	11,9	90,0
IIIf	n-C_3H_7	CH_3	78,5—79,5 (hexane)	0,01 : 0,013	$C_{14}H_{20}N_2O_2$	67,7	8,0	11,3	67,7	8,1	11,3	80,9
IIIg	n-C_3H_7	Cl	104—105 (hexane-benzene)	0,01 : 0,012	$C_{13}H_{17}ClN_2O_2$	57,9	6,5	10,6	58,0	6,4	10,4	77,2
IIIh	n-C_3H_7	Br	111,5—112 (hexane)	0,01 : 0,012	$C_{13}H_{17}BrN_2O_2$	49,8	5,2	9,4	49,8	5,4	9,0	70,3

*The abbreviation "AAA" stands for acetoacetic acid.

TABLE 2. Substituted 2-Methylbenzimidazoles

Compound	Name	mp, °C	Yield, %		mp, °C according to the literature	R_f, Al_2O_3 (act. II), chloroform – EtOH (20 : 1)
			method A	method B		
IVa	2-Methylbenzimidazole	175—176	68,2 (70,4)*	61,3	175—176[12]	0,58
IVb	2,5-Dimethylbenzimidazole	202—203	69,0	—	201—202[13]	0,67
IVc	2-Methyl-5-chlorobenzimidazole	209—210	50,5	53,2	210—211[13]	0,60
IVd	2-Methyl-5-nitrobenzimidazole	221—222	34,5	—	219—220[14]	0,49
IVe	2-Methyl-5,7-dichlorobenzimidazole	201—202	73,0	—	200—201[15]	0,43
IVf	2-Methyl-5,7-dibromobenzimidazole	214—215	70,0	—	215[16]	0,44

*This is the yield of 2-methylbenzimidazole in the reaction of 1,2-phenylenediamine with ethyl 2-n-propylacetoacetate; the remaining yields are those in the reaction of aromatic diamines with ethyl 2-ethylacetoacetate.

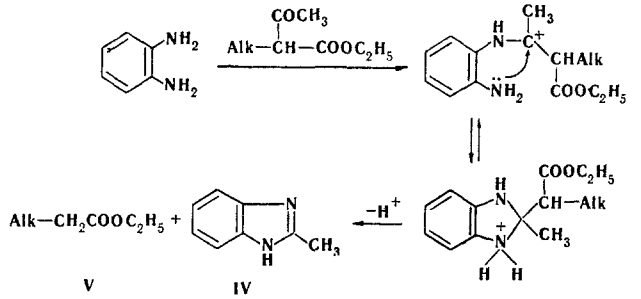
If the benzene ring contains electron-donor substituents (CH_3, Cl, Br), the ketimine structure cannot be isolated, and esters III are formed (see Table 1).

Crotonates II are isomerized to ketimines I under the influence of acid. This sort of isomerization is not observed for crotonates III, which have a cis-enamine structure [7].

The structures of I and II were confirmed by the IR and PMR spectra. In the case of the ethyl esters, the structures were also confirmed by their mass spectra [7].

The isomerization of crotonate II to ketimine I is in agreement with the results obtained by Favorskaya and co-workers [8], who observed the formation of a mixture of aminocrotonates in the imine and enamine forms in the reaction of monoamines with 2-alkylacetoacetic esters.

The reaction of o-diamines with 2-alkylacetoacetic esters has not been previously investigated. It was found that the chief reaction products in the condensation of various 1,2-phenylene diamines with 2-ethyl-and 2-propylacetoacetic esters are 2-methylbenzimidazole derivatives (see Table 2). Under mild conditions, the reagents were recovered practically unchanged, whereas substituted 2-methylbenzimidazoles IV are formed during acid catalysis and upon heating. Aliphatic esters V were detected in the volatile fractions by gas–liquid chromatography (GLC). The gem-substituted benzimidazolinium salt is probably formed initially and is then aromatized with the elimination of a molecule of ester V.



V IV

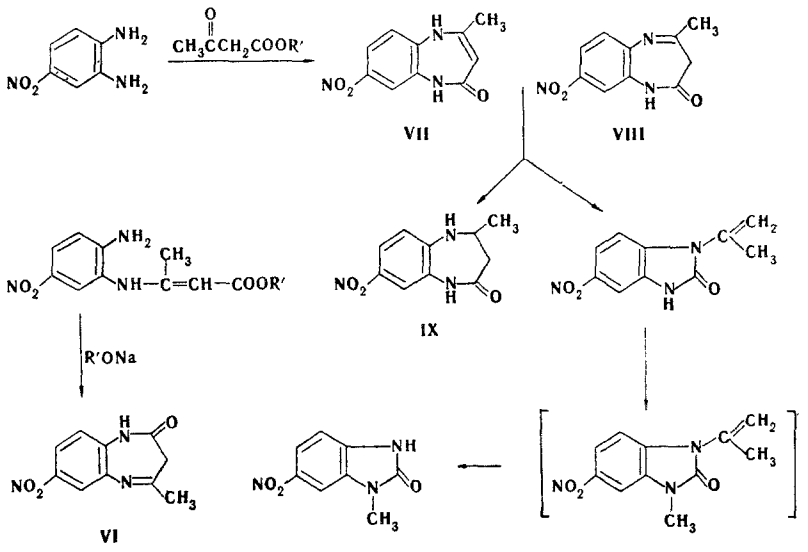
Alk = C_2H_5; n-C_3H_7

Ketimines I are cleaved on heating with sodium alkoxide, whereas crotonates II are cyclized to 1,5-benzo-2-diazepinone derivatives (VI). However, if the condensation of the nitrodiamine with methyl or ethyl acetoacetate is carried out by heating in boiling xylene, a mixture of benzodiazepinones VII and VIII, which differ from isomer VI with respect to the position of the double bond, determined by comparison of the IR and PMR spectra, is formed. The structures of VII and VIII were reconfirmed by thermal rearrangement to 1-isopropenyl-5-nitro-2-benzimidazolone [3], the alkylation of which with methyl iodide and subsequent hydrolysis give 1-methyl-6-nitro-2-benzimidazolone [9]. In addition, VII and VIII are reduced to 4-methyl-8-nitro-1H-2,3,4,5-tetrahydro-1,5-benzo-2-diazepinone (IX). The structures of the isomeric nitrodiazepinones, which differ with respect to the position of the double bond in the seven-membered ring (VII and VIII) and the position of the nitro group in the aromatic ring (VI and VIII), were also confirmed by a study of their mass-spectrometric disintegration [10].

Thus under relatively severe conditions the condensation of the nitrodiamine with methyl or ethylacetoacetate gives benzodiazepinones VII and VIII; this corresponds to attack of the carbonyl group on the less-basic amino group. Whether this process occurs precisely as direct attack on the less nucleophilic group or the arylaminocrotonate, as an intermediate, isomerizes to an acetoacetic acid arylamide with subsequent cyclization is difficult to judge. (See scheme on following page.)

EXPERIMENTAL METHOD

The individuality of the substances was monitored in a thin loose layer of silica gel or aluminum oxide in chloroform or chloroform–ethanol (20:1). The PMR spectra of pyridine, chloroform, and deuterochloroform solution were recorded with an RS-60 spectrometer with



hexamethyldisiloxane as the internal standard. The IR spectra of mineral-oil suspensions were recorded with an IKS-14 spectrometer. The UV spectra were recorded with an SF-4A spectrophotometer.

3-Chloro-5-nitro-1,2-phenylenediamine. A stream of hydrogen sulfide was bubbled at 45-55° into a mixture consisting of 21.8 g (0.1 mole) of 6-chloro-2,4-dinitroaniline, 150 ml of ethanol, and 150 ml of 25% ammonium hydroxide for 2.5 h, after which it was allowed to stand in a refrigerator for 24 h. The resulting precipitate was removed by filtration, washed with cold water, and purified by dissolving it in hot dilute hydrochloric acid (100 ml of water and 60 ml of concentrated HCl). The hot solution was neutralized with ammonium hydroxide, and the precipitate was removed by filtration. The diamine was crystallized from water to give 12.2 g (65%) of dark-cherry-red needles with mp 210-211° (from water). Found %: C 38.4; H 3.1; Cl 18.9; N 22.5. $C_6H_6ClN_3O_2$. Calculated %: C 38.4; H 3.2; Cl 18.9; N 22.4. IR spectrum, cm^{-1}: 3498, 3434, 3400, 3355, 3265, 3105, 1632, 1612, 1519, 1474, 1338, 1309, 1226, 1092, 965, 866, 742, 725.

Ethyl 3-(2-Amino-3-chloro-5-nitrophenylimino)butyrate (Ia). A 13.0-g (0.1 mole) sample of acetoacetic ester and two to three drops of concentrated HCl were added to 1.87 g (0.01 mole) of 3-chloro-5-nitro-1,2-phenylenediamine, after which the mixture was stirred at 20-25° for 30 min and allowed to stand for 72 h. The addition of ice water to the mixture liberated an oily red substance, which began to crystallize in ether–hexane. Workup gave 2.1 g (70.3%) of a product with mp 135.5-136.5° (ether–hexane). Found %: C 48.2; H 4.8; Cl 11.8. $C_{12}H_{14}ClN_3O_4$. Calculated %: C 48.2; H 4.6; Cl 11.8. IR spectrum, cm^{-1}: ν_{NH} 3386, 3355; $\nu_{C=O}$ 1720; 1603, 1513, 1485, 1349, 1300, 1272, 1264, 1203, 1173, 1131, 1068, 1024, 857, and 730. PMR spectrum (chloroform), ppm: singlet at 1.66 (=C-CH_3), triplet at 1.28 (CH_3 in $-OCH_2CH_3$), quartet at 4.18 ($-OCH_2-$), singlet at 2.82 ($-CH_2-$), and doublet at 5.27 and 4.77 ($-NH_2$).

Ethyl 3-(2-Amino-3-chloro-5-nitrophenylamino)crotonate (IIa). A 13.0-g (0.1 mole) sample of acetoacetic ester and two to three drops of concentrated HCl were added to 1.87 g (0.01 mole) of 3-chloro-5-nitro-1,2-phenylenediamine, after which the mixture was stirred at 30-40° for 30 min. During this period, the mixture crystallized and turned yellow. Workup gave 1.81 g (60.4%) of a product with mp 120-121° (from hexane). Found %: C 48.4; H 4.8; Cl 11.8. $C_{12}H_{14}ClN_3O_4$. Calculated %: C 48.2; H 4.7; Cl 11.8. IR spectrum, cm^{-1}: ν_{NH} 3381, 3304, 3255, and 3206; $\nu_{C=C}$ 1635; 1594, 1511, 1332, 1268, 1172, 1091, 1056, 1016, 996, 900, 881, 819, 783, 759, 737, and 718. PMR spectrum (chloroform), ppm: singlet at 1.85 (=C-CH_3), triplet at 1.29 (CH_3 in $-OCH_2CH_3$), quartet at 4.20 ($-OCH_2-$), singlet at 4.92 (=C-H), singlet at 5.10 ($-NH_2$), and singlet at 9.75 (-NH).

Methyl 3-(2-Amino-5-nitrophenylimino)butyrate (Ib). The procedure used to prepare Ia was used to obtain this compound from 1.13 g (0.0074 mole) of 4-nitro-1,2-phenylenediamine, 8.6 g (0.074 mole) of methyl acetoacetate [11], and three drops of concentrated HCl. The

yield of red product was 0.63 g (34.1%). The product was purified by chromatography with a column filled with silica gel and elution with dry chloroform to give a material with mp 98.5-100° (ether–hexane). Found %: C 52.7; H 5.3; N 16.8. $C_{11}H_{13}N_3O_4$. Calculated %: C 52.6; H 5.2; N 16.7. IR spectrum, cm^{-1}: ν_{NH} 3352; $\nu_{C=O}$ 1720; 1624, 1505, 1486, 1289, 1174, 1130, 1070, 1000, 883, 847, 784, 744, and 727. PMR spectrum (pyridine), ppm: singlet at 1.38 (=C–CH_3), singlet at 3.16 (–OCH_3), singlet at 2.63 (–CH_2–); (deuterochloroform), ppm: singlet at 2.0 (=C-CH_3), singlet at 4.10 (–OCH_3), and singlet at 3.22 (–CH_2–).

Methyl 3-(2-Amino-5-nitrophenylamino)crotonate (IIb). The procedure used to obtain IIa was used to prepare this compound, with mp 162-163° (from chloroform), in 65.3% yield. Found %: C 52.9; H 5.3; N 16.4. $C_{11}H_{13}N_3O_4$. Calculated %: C 52.6; H 5.2; N 16.7. IR spectrum, cm^{-1}: ν_{NH} 3450, 3364, and 3237; $\nu_{C=O}$ 1631; 1618, 1600, 1582, 1492, 1291, 1182, 1169, 1091, 1054, 1000, 887, 818, 811, 784, 742, and 727. PMR spectrum (pyridine), ppm: singlet at 1.48 (cis, =C-CH_3), s 2.50 (trans, =C-CH_3), s 4.63 (cis, =C-H), and s 3.28 (-OCH_3).

3-(2-Aminoarylamino)crotonic Acid Esters (IIIa-h, Table 1). A mixture of 0.01 mole of aromatic o-diamine containing a substituent in the 4 position, an excess of the appropriate acetoacetic acid ester, and one drop of concentrated HCl was stirred at 20-25° for 15-20 min, during which the diamine dissolved. After a certain, time, the mixture became turbid, and the reaction product began to crystallize. The product was removed by filtration and washed with a small amount of petroleum ether–diethyl ether and crystallized several times from a suitable solvent with decolorization by activated charcoal.

Substituted 2-Methylbenzimidazoles (IVa-f, Table 2). Method A. A mixture of 0.01 mole of the diamine, 0.019 mole of ethyl 2-ethylacetoacetate (or 0.017 mole of ethyl 2-n-propylacetoacetate) and one drop of concentrated HCl was heated on a water bath (50-60°) until the diamine dissolved. The mixture was then cooled, and the viscous mass was washed with boiling hexane, from which 10-20% of the starting diamine was isolated. The residue was recrystallized from benzene.

Method B. A mixture of 0.01 mole of the aromatic diamine, 0.0105 mole of ethyl 2-ethylacetoacetate, and 0.07 g of zinc chloride [5] or catalytic amounts of acetic acid [8] in 20 ml of absolute ethanol was refluxed until the starting diamine had vanished. The alcohol was then removed by distillation, and benzimidazoles IVa and IVc were extracted from the dark oil with boiling benzene. The substituted benzimidazoles were identified by comparison of their IR spectra, melting points, and R_f values with the corresponding data for genuine samples, which were obtained by known methods [12-14].

Condensation of o-Phenylenediamine with Ethyl 2-n-Propylacetoacetate without a Catalyst. A mixture of 7.2 g (0.067 mole) of o-phenylenediamine and 17.2 g (0.1 mole) of ethyl 2-n-propylacetoacetate was heated on a water bath until the diamine had dissolved, after which the mixture was held at 50-60° for 1 h and then slowly cooled. The precipitated o-phenylenediamine [4.4 g (61%)] was removed by filtration. The IR spectrum and the melting point coincided with the spectrum and melting point of a genuine sample. Chromatography of the filtrate on aluminum oxide showed the presence of three substances — ethyl 2-n-propylacetoacetate, o-phenylenediamine, and traces of 2-methylbenzimidazole [elution with chloroform–ethanol (3:1)].

Ethyl Butyrate (V). Ester V was identified by GLC with a KhL-69 chromatograph. The conditions for the separation of the ethanol and ethyl butyrate were as follows: a 2 m by 4 mm metal column filled with Cellite C-22, which was impregnated with 5% polypropylene glycol distearate and 7.5% Apezion L, a katharometer as the detector, nitrogen as the carrier gas, and a column temperature of 130°. The retention times of the substances isolated after removal of the alcohol from the reaction mixture by distillation (method B) and a genuine sample of ethyl butyrate coincided during chromatography under the indicated conditions.

4-Methyl-7-nitro-1H-2,3-dihydro-1,5-benzo-2-diazepinone (VI). A 2.76-g (0.011 mole) sample of methyl 3-(2-amino-5-nitrophenylamino)crotonate was heated with sodium methoxide (from 0.49 g of sodium and 20 ml of methanol) on a water bath for 3 h, after which it was cooled, treated with 20 ml of water, and neutralized with acetic acid. The resulting precipitate [1.88 g (78%)] was removed by filtration and crystallized from dimethylformamide (DMFA) to give a product with mp 241-243° (the color of the crystals changed at 227°).

Found %: C 54.5; H 4.1; N 19.1. $C_{10}H_9N_3O_3$. Calculated %: C 54.7; H 4.1; N 19.1. UV spectrum (DMFA), λ_{max}, nm (log ε): 282 (4.15) and 315 (4.23). PMR spectrum (trifluoroacetic acid), ppm: s 2.93 (—CH_3), s 3.93 (—CH_2—), and m 7.0-8.45 (aromatic H).

A 1.33-g (0.0053 mole) sample of methyl 3-(2-amino-5-nitrophenylimino)butyrate was heated with sodium methoxide under the conditions used for the preparation of IIb. The reaction mixture was worked up similarly to give 0.28 g (35%) of 4-nitro-1,2-phenylenediamine with mp 194-195° (from water). The product had an identical R_f value and did not depress the melting point of a genuine sample.

4-Methyl-8-nitro-1H-2,5-(and 2,3)-dihydro-1,5-benzo-2-diazepinones (VII, VIII). A mixture consisting of 7 g (0.06 mole) of methyl acetoacetate and 6.12 g (0.04 mole) of 4-nitro-1,2-phenylenediamine in 500 ml of xylene was refluxed, with water removal, for 3 h, after which 300 ml of xylene was removed by distillation, and the mixture was cooled to precipitate 8.1 g (91%) of 4-methyl-8-nitro-1H-2,5-dihydro-1,5-benzo-2-diazepinone (VII) with mp 204-205° (yellow crystals from acetone). The product turned red at 197°. It was partially soluble in chloroform, benzene, and ether. Found %: C 54.7; H 4.1; N 19.0. C_{10}-$H_9N_3O_3$. Calculated %: C 54.7; H 4.1; N 19.1. UV spectrum (alcohol), λ_{max}, nm (log ε): 227 (4.36), 240 (4.36), and 313 (3.98); in DMFA: 324 (3.95) and 435 (3.59). IR spectrum, cm^{-1}: ν_{NH} 3180, 3110; 3050; $\nu_{C=O}$ 1690; 1660, 1580, 1520, 1485, 1300, 1260, 1245, 1225, 1130, 1090, 1025, 980, 955, 885, 795, 750, and 740. PMR spectrum (trifluoroacetic acid), ppm: s 2.67 (—CH_3), s 3.62 (—CH_2—), and m 7.14-7.30 (aromatic H); in dimethyl sulfoxide (DMSO): s 8.85 (—NH) and s 8.97 (—NH).

When VII or the crude reaction product was crystallized from alcohol, bright-red 4-methyl-8-nitro-1H-2,3-dihydro-1,5-benzo-2-diazepinone (VIII), with mp 204-205°, precipitated. Found %: C 54.7; H 4.1; N 19.2. $C_{10}H_9N_3O_3$. Calculated %: C 54.7; H 4.1; N 19.1. UV spectrum (alcohol), λ_{max}, nm (log ε): 227 (4.23), 242 (4.27), and 314 (3.95); in DMFA: 324 (3.94) and 430 (3.54). IR spectrum, cm^{-1}: ν_{NH} 3110; $\nu_{C=O}$ 1690; 1655, 1590, 1520, 1485, 1300, 1270, 1250, 1230, 1140, 1090, 890, 850, 815, and 740. PMR spectrum (trifluoroacetic acid), ppm: s 2.71 (—CH_3), s 3.62 (—CH_2—), m 7.14-7.35 (aromatic H); in DMSO: s 8.98 (—NH). According to [3], this compound had mp 196-197°.

1-Methyl-6-nitro-2-benzimidazolone. A 0.9-g (0.0041 mole) sample of 1-isopropenyl-5-nitro-2-benzimidazolone [3] and 0.7 ml of methyl iodide were added successively to a solution of sodium ethoxide obtained from 0.1 g of sodium and 4 ml of absolute ethanol, after which the mixture was refluxed for 4.5 h. The alcohol was then removed, and the residue was heated with 9 ml of 20 N sulfuric acid for 3.5 h. The solution was then cooled and neutralized with ammonia to give 0.3 g (38%) of dark-yellow crystals with mp 270-271°. Found %: C 50.0; H 3.6; N 21.6. $C_8H_7N_3O_3$. Calculated %: C 49.7; H 3.7; N 21.7. IR spectrum, cm^{-1}: ν_{NH} 3160; $\nu_{C=O}$ 1710; 1690, 1596, 1580, 1515, 1340, 1205, 1135, 1090, 938, 870, 843, 770, and 740. According to [9], this compound has mp 272°. According to [9], the isomeric 1-methyl-5-nitro-2-benzimidazolone had mp 300°.

4-Methyl-8-nitro-1H-2,3,4,5-tetrahydro-1,5-benzo-2-diazepinone (IX). A mixture of 0.4 g (0.002 mole) of diazepine VII or VIII in 7 ml of absolute ethanol and 0.24 g of $NaBH_4$ was refluxed for 2 h, after which the hot solution was filtered to give 0.38 g (94%) of IX with mp 249-250° (from methanol). The color of the product changed at 215-220°. Found %: C 54.4; H 4.0; N 19.2. $C_{10}H_{11}N_3O_3$. Calculated %: C 54.2; H 4.9; N 19.0. UV spectrum (alcohol), λ_{max} nm (log ε): 265 (4.25), 312 (3.80), and 376 (3.56). IR spectrum, cm^{-1}: ν_{NH} 3350, 3280, and 3180; 3080; $\nu_{C=O}$ 1665; 1585, 1540, 1490, 1330, 1220, 1140, 925, 870, 765, and 740.

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