

# Three-Component Reaction of Methyl 2,4-Dioxo-4-phenylbutanoate and Methyl 2,4-Dioxopentanoate with Aromatic Aldehydes and Propane-1,2-diamine and Chemical Properties of the Products

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**Abstract**—The reactions of methyl 2,4-dioxo-4-phenylbutanoate and methyl 2,4-dioxopentanoate with a mixture of an aromatic aldehyde and propane-1,2-diamine, depending on the initial reactant ratio, gave 4-acyl-1-(2-aminopropyl)-5-aryl-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-ones and 1,1'-(propane-1,2-diyl)bis(4-acetyl-3-hydroxy-5-phenyl-2,5-dihydro-1*H*-pyrrol-2-one). Reactions of substituted 1-(2-aminopropyl)-2,5-dihydro-1*H*-pyrrol-2-ones with aromatic amines and hydrazines were studied, and the structure of one of the products, 5-(2-aminopropyl)-3,4-diphenylpyrrolo[3,4-*c*]pyrazol-6-one, was proved by X-ray analysis.

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We previously showed that reactions of 2,4-dioxo-alkanoic acid esters with aromatic aldehydes and ethylenediamine, depending on the initial reactant ratio, lead to the formation of 1-(2-aminoethyl)-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-ones or 1,1'-(ethane-1,2-diyl)-bis(3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-ones) [1]. It was interesting to elucidate whether analogous relation is observed in reactions of the same compounds with alkanediamines having a different structure.

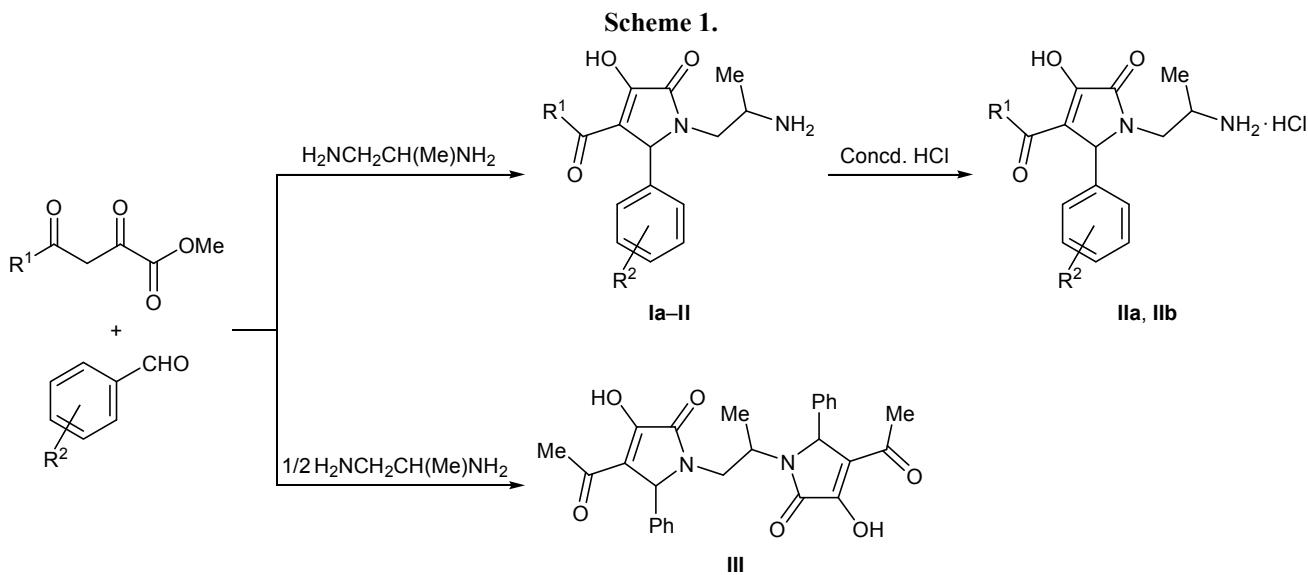
For this purpose, we examined the reaction of methyl 2,4-dioxo-4-phenylbutanoate and methyl 2,4-dioxopentanoate with a mixture of aromatic aldehyde and propane-1,2-diamine at molar ratios of 1:1:1 and 2:2:1. The results of our experiments showed that mixing of the initial reactants in 1,4-dioxane or ethanol at a ratio of 1:1:1 gives the corresponding 4-acyl-1-(2-aminopropyl)-5-aryl-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-oes **Ia**–**II** (Scheme 1). Compounds **Ia** and **Ih** were converted into the corresponding hydrochlorides **IIa** and **IIb** by treatment with concentrated hydrochloric acid.

The mechanism of formation of compounds **Ia**–**II** is likely to be similar to the mechanism of analogous reactions described in [1]. Compounds **Ia**–**II** are color-

less crystalline substances, which are soluble in dimethylformamide and dimethyl sulfoxide and insoluble or poorly soluble in alcohols. Hydrochlorides **IIa** and **IIb** are soluble in common organic solvents. Compound **IIa** is soluble in water, and it shows a positive test for chloride ion (treatment with a solution of silver nitrate); compound **IIb** is poorly soluble in water.

The spectral parameters of free bases **I** and hydrochlorides **II** are similar. In the IR spectra of **Ia**–**II**, **IIa**, and **IIb** we observed absorption bands corresponding to stretching vibrations of the ketone ( $1610\text{--}1651\text{ cm}^{-1}$ ) and lactam carbonyl groups ( $1655\text{--}1718\text{ cm}^{-1}$ ), primary amino group ( $3300\text{--}3503\text{ cm}^{-1}$ ) and enol hydroxy group ( $3158\text{--}3240\text{ cm}^{-1}$ ). Some compounds displayed no absorption bands of amino group and enol hydroxy group because of strong broadening due to exchange processes.

The  $^1\text{H}$  NMR spectra of **I** and **II** contained a doublet signal with a coupling constant *J* of 5.7–6.6 Hz or a multiplet signal belonging to three protons in the methyl group of the aminoalkyl residue ( $\delta$  0.98–1.31 ppm), multiplets from two methylene protons at  $\delta$  2.33–3.24 ( $1'\text{-H}_A$ ) and 3.35–4.09 ppm ( $1'\text{-H}_B$ ) and CH proton in the aminoalkyl residue at  $\delta$  3.22–



3.80 ppm, one or two singlets from 5-H in the pyrrole ring ( $\delta$  4.98–5.95 ppm), a group of signals from aromatic protons in the region  $\delta$  6.70–8.28 ppm, and a broadened singlet from protons in the primary amino group at  $\delta$  7.77–8.18 ppm. In the spectra of **Ia–Ig** and **IIa** we also observed a signal from protons in the acetyl residue at  $\delta$  2.04–2.34 ppm (two singlets for most compounds).

High signal multiplicity originates from the presence of two chiral centers in molecules **I** and **II**, so that the products are formed as mixtures of diastereoisomers. The ratio of diastereoisomers was determined from the intensity ratio of the 5-H signals; it ranged from 1:1 to 1:2. The observed broadening of the melting temperature range of compounds **Ia**, **Ic**, **Id**, and **Ij** is also explained by the presence of different diastereoisomers. Compounds **Ib**, **Ih**, and **Ik** displayed in the mass spectra the molecular ion peaks and peaks of fragment ions with  $m/z$  57 [ $\text{CH}_2=\text{C}(\text{Me})\text{NH}_2]^+$ , 44 [ $\text{CH}_2=\text{CHNH}_3]^+$  (**Ih**), 77 [ $\text{C}_6\text{H}_5]^+$ , 105 [ $\text{C}_6\text{H}_5\text{CO}]^+$  (**Ik**), as well as [ $\text{R}^2\text{C}_6\text{H}_4\text{CH}=\text{CHCOR}^{1+}]$  and [ $M - \text{CH}_2=\text{CHNH}_2]^+$  ion peaks.

All compounds **I** and **II** showed a positive test (intense cherry color) for enol hydroxy group upon treatment with an alcoholic solution of iron(III) chloride, which, in combination with the spectral data, indicates that these compounds exist in the enol form.

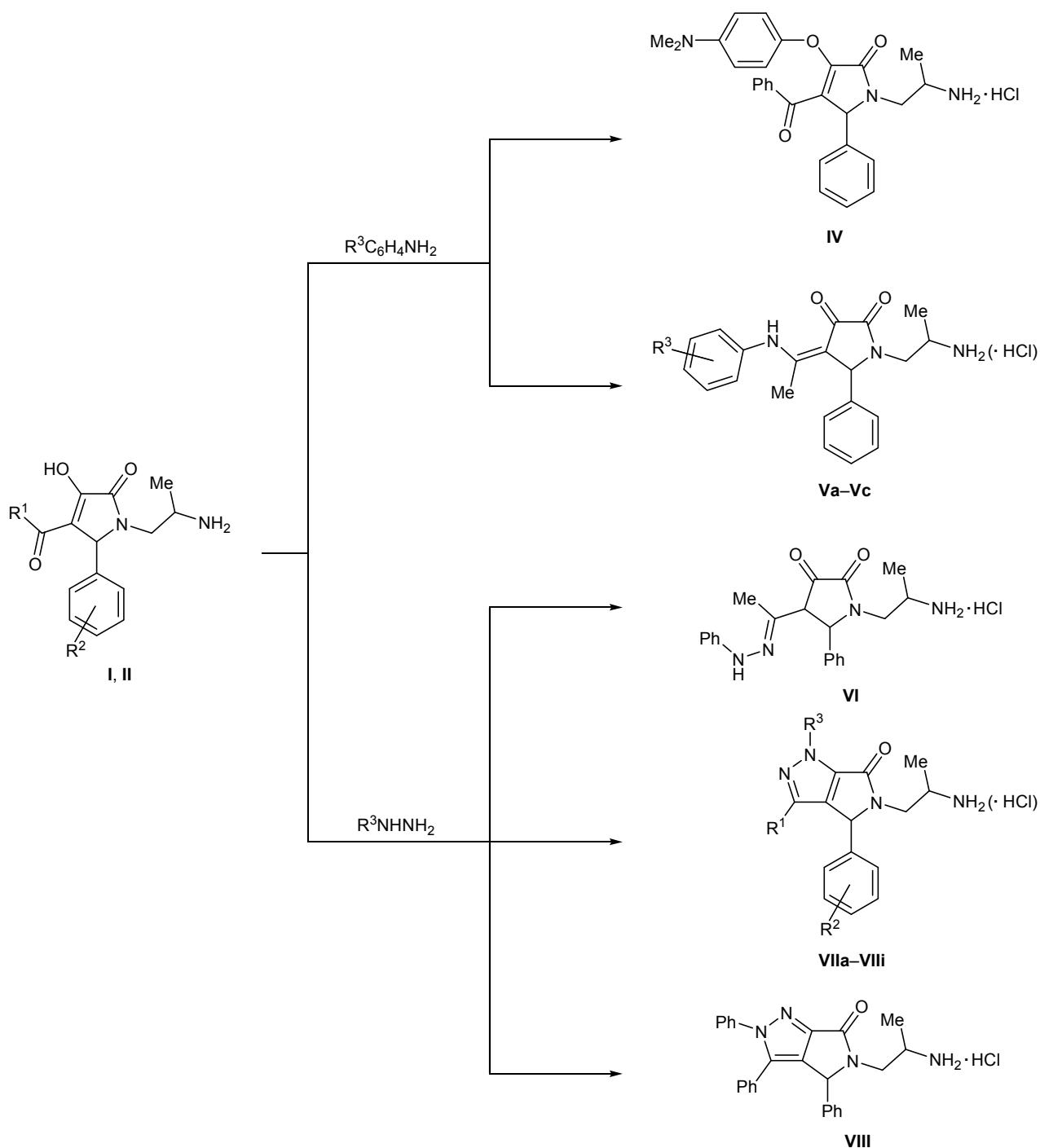
The reaction of methyl 2,4-dioxo-4-phenylbutanoate with benzaldehyde and 0.5 equiv of propane-1,2-diamine afforded 1-(2-aminopropyl)-4-benzoyl-3-hy-

droxy-5-phenyl-2,5-dihydro-1*H*-pyrrol-2-one (**Ih**), while analogous reaction with methyl 2,4-dioxopentanoate gave 1,1'-(propane-1,2-diyl)bis(4-acetyl-3-hydroxy-5-phenyl-2,5-dihydro-1*H*-pyrrol-2-one) (**III**) (Scheme 1). Presumably, in the latter case the reaction also involves intermediate formation of Schiff base [2] due to higher reactivity of methyl 2,4-dioxopentanoate. Another reason is steric shielding of position 2 in the propane-1,2-diamine molecule. The same factor is responsible for the structure of the aminoalkyl residue assigned to compounds **I** and **II**.

Compound **III** is a colorless crystalline substance which is soluble in common organic solvents. Its structure was confirmed by the data of IR,  $^1\text{H}$  NMR, and mass spectra. It showed a positive test for enol hydroxy group with iron(III) chloride.

Taking into account that molecules **I** and **II** possess two reactive carbonyl groups, it was interesting to examine their reactions with nucleophilic reagents. As the latter we used aromatic amines and hydrazines. It was shown previously [1] that 4,5-disubstituted 1-aminoethyl-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-ones react with aromatic amines and hydrazine. Reactions of compounds having a 2-aminopropyl substituent in position 1 of the heteroring successfully occurred with participation of bases. 4-Acyl-1-(2-aminopropyl)-5-aryl-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-ones and their hydrochlorides reacted with *p*-anisidine, *p*-phenetidine, and *p*-(dimethylamino)aniline to give the corresponding arylamino derivatives

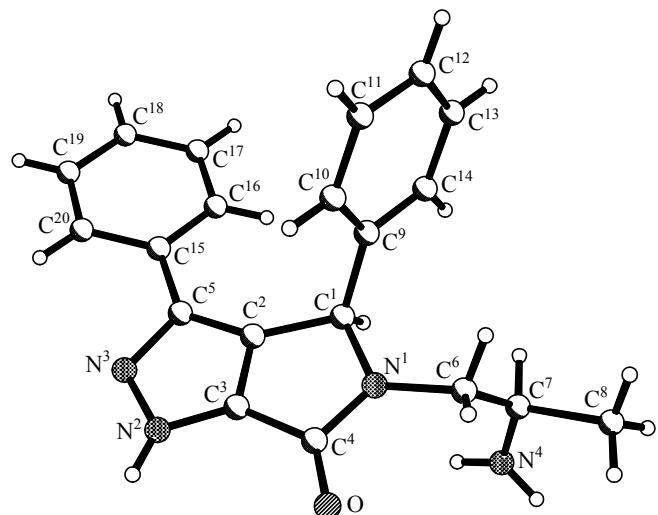
Scheme 2.



**V**,  $\text{R}^3 = 4\text{-MeO}(\cdot \text{HCl})$  (**a**),  $4\text{-EtO}(\cdot \text{HCl})$  (**b**),  $4\text{-EtO}(\cdot \text{HCl})$  (**c**); **VII**,  $\text{R}^1 = \text{Me}$ ;  $\text{R}^3 = \text{H}$ ,  $\text{R}^2 = 4\text{-Cl}$  (**a**),  $4\text{-MeO}$  (**b**);  $\text{R}^3 = \text{Ph}$ ,  $\text{R}^2 = \text{H}$  (**c**),  $\text{H}(\cdot \text{HCl})$  (**d**);  $\text{R}^1 = \text{Ph}$ ,  $\text{R}^3 = \text{H}$ ,  $\text{R}^2 = \text{H}$  (**e**),  $\text{H}(\cdot \text{HCl})$  (**f**),  $4\text{-Cl}$  (**g**),  $4\text{-MeO}$  (**h**),  $3\text{-O}_2\text{N}$  (**i**).

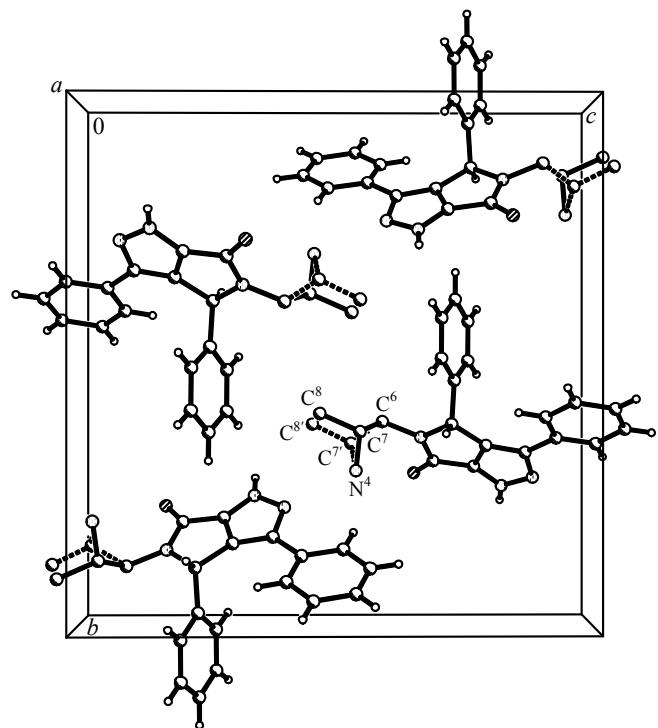
whose structure depended on the substituent in position 4 of the heteroring. Compounds having a benzoyl group reacted at the enolized carbonyl group in the 3-position, while 4-acetyl derivatives reacted at the side-chain carbonyl group (Scheme 2). Analogous rela-

tions were observed in the reactions with hydrazine and phenylhydrazine, which is consistent with published data [3]. Arylamino derivatives **IV** and **V** and pyrrolo[3,4-*c*]pyrazoles **VII** and **VIII** were formed in boiling glacial acetic acid. Phenylhydrazone **VI** was



**Fig. 1.** Structure of the molecule of 5-(2-aminopropyl)-3,4-diphenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one (**VIIe**) according to the X-ray diffraction data.

obtained by heating for a short time in ethanol. Compounds **VI–VIII** are colorless while **IV** and **V** are yellow or orange crystalline substances which are soluble in ethanol, DMF, and DMSO. The corresponding hydrochlorides were better soluble than the free bases. All hydrochlorides, except for compound **IV**, are soluble in water, and they show a positive test for chloride ion with a solution of silver nitrate.



**Fig. 2.** Packing of molecules **VIIe** in crystal.

The IR spectra of **IV** and **V** contained absorption bands belonging to stretching vibrations of the lactam ( $1688\text{--}1710\text{ cm}^{-1}$ ) and ketone carbonyl group ( $1630\text{--}1650\text{ cm}^{-1}$ ), secondary amino group ( $3200\text{--}3310\text{ cm}^{-1}$ ), and primary amino group in the side chain ( $3400\text{--}3500\text{ cm}^{-1}$ ). Compounds **IV** and **V** displayed in the  $^1\text{H}$  NMR spectra a multiplet or doublet from the side-chain methyl group ( $\delta 0.83\text{--}1.07\text{ ppm}$ ), multiplets from two methylene protons at  $\delta 2.34\text{--}2.82$  ( $1'\text{-H}_A$ ) and  $3.32\text{--}3.70\text{ ppm}$  ( $1'\text{-H}_B$ ) and side-chain CH proton at  $\delta 2.97\text{--}3.30\text{ ppm}$ , one or two singlets from 5-H at  $\delta 5.42\text{--}5.69\text{ ppm}$ , multiplets from aromatic protons in the region  $\delta 6.39\text{--}7.34\text{ ppm}$ , a singlet from the primary amino group at  $\delta 8.02\text{--}8.10\text{ ppm}$ , and a singlet from the NH proton at  $\delta 9.23\text{--}12.44\text{ ppm}$  (the spectrum of **Vb** lacked the two latter signals). In the mass spectrum of **Vb** the molecular ion peak with  $m/z 393$  was observed together with fragment ion peaks in support of the assumed structure.

Compounds **VI–VIII** were characterized by IR absorption bands due to stretching vibrations of the lactam carbonyl group ( $1675\text{--}1710\text{ cm}^{-1}$ ), secondary amino group ( $3200\text{--}3290\text{ cm}^{-1}$ ; except for **VIIc**, **VId**, and **VIII**), and primary amino group ( $3350\text{--}3500\text{ cm}^{-1}$ ). In the  $^1\text{H}$  NMR spectra of **VII** and **VIII** alkyl protons resonated in the region  $\delta 0.85\text{--}3.84\text{ ppm}$ , the 5-H signal appeared at  $\delta 5.50\text{--}6.25\text{ ppm}$ , aromatic protons gave a multiplet in the region  $\delta 6.82\text{--}8.31\text{ ppm}$ , and the  $\text{NH}_2$  signal was located at  $\delta 7.61\text{--}8.29\text{ ppm}$  (cf. the spectra of initial compounds **I** and **II**). In addition, a broadened singlet from the NH proton in the pyrazole ring was observed ( $\delta 14.08\text{ ppm}$  for **VIf**). The  $^1\text{H}$  NMR spectra of the free bases in  $\text{DMSO}-d_6$  contained no signal from the primary amino group, but the corresponding signal was observed in the spectra recorded in the presence of trifluoroacetic acid (**VIf**, **VIIh**, **VIIIi**), as well as in the spectra of hydrochlorides **VId**, **VIf**, and **VIII**, indicating that these protons are exchangeable. The mass spectra of **VIIe**, **VIIIi**, and **VIII** contained the molecular ion peaks and peaks from fragment ions, which confirmed the assumed structures.

The molecular and crystalline structure of pyrrolopyrazole **VIIe** was determined by X-ray analysis of its single crystal obtained by slow crystallization from alcohol (Fig. 1). The pyrrolopyrazole bicyclic system is planar, and the double bonds therein are completely delocalized. The benzene ring on  $\text{C}^5$  lies almost in the plane of the bicyclic fragment (the dihedral angle between the corresponding planes is  $11.2^\circ$ ), whereas the benzene ring on  $\text{C}^1$  is almost orthogonal to the

pyrrolopyrazole skeleton (the corresponding dihedral angle is  $88.9^\circ$ , and the torsion angle  $N^1C^1C^9C^{10}$  is  $69.7^\circ$ ). The  $C^7$  and  $C^8$  atoms in the aminopropyl fragment are disordered by two positions with almost equal probabilities, the distance between the disordered positions being 1 Å. These positions are related to each other through a symmetry plane passing through the  $N^1$ ,  $C^6$ , and  $N^4$  atoms. Each crystallographic position is statistically occupied by two diastereoisomers (Fig. 2). Thus, there are two enantiomers and two diastereoisomers, which is very consistent with the spectral data.

The aminopropyl groups in crystal reside in a plane parallel to the crystallographic  $a$  axis, they are not involved in intermolecular hydrogen bonds, and no shortened contacts (relative to the corresponding van der Waals contacts) were observed. In combination with large thermal vibration parameters of the  $C^7$ ,  $C^8$  and  $C^{7'}$ ,  $C^{8'}$  atoms, these findings suggest that diastereoisomers of **VIIe** in crystal exist in dynamic equilibrium.

## EXPERIMENTAL

The IR spectra were recorded on UR-20 and Specord M-80 spectrometers from samples dispersed in mineral oil. The  $^1H$  NMR spectra were measured on Bruker DRX-500 (500.13 MHz), Bruker DRX-400 (400.13 MHz), and Varian Mercury-300 (300.05 MHz) instruments using hexamethyldisiloxane as internal reference. The mass spectra (electron impact, 70 eV) were obtained on a Finnigan MAT Incos 50 mass spectrometer. The progress of reactions was monitored, and the purity of compound **VIII** was checked, by TLC on Silufol plates using acetonitrile–water–ethanol (1:1:1) as eluent.

**4-Acetyl-1-(2-aminopropyl)-3-hydroxy-5-phenyl-2,5-dihydro-1*H*-pyrrol-2-one (Ia).** A solution of 0.01 mol of methyl 2,4-dioxopentanoate in 15 ml of 1,4-dioxane was added at room temperature to a solution of 0.01 mol of propane-1,2-diamine and 0.01 mol of benzaldehyde in 5 ml of 1,4-dioxane. The mixture was kept for 24 h at room temperature, and the precipitate was filtered off and recrystallized from appropriate solvent. Yield 1.21 g (44%), mp 290–300°C (from ethanol). IR spectrum,  $\nu$ , cm<sup>−1</sup>: 3503 (NH<sub>2</sub>), 3158 (OH), 1700 (NC=O), 1625 (C=O).  $^1H$  NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 1.00 d (3H, Me,  $J$  = 6.3 Hz), 2.04 s and 2.07 s (3H, MeCO), 2.33 m and 2.56 m (1H, 1'-H<sub>A</sub>), 3.51 m and 3.59 m (1H, 1'-H<sub>B</sub>), 3.32 m (1H, 2'-H), 5.01 s and 5.02 s (1H, 5-H), 7.02 m (2H, H<sub>arom</sub>), 7.17 m (3H, H<sub>arom</sub>), 8.14 br.s (2H, NH<sub>2</sub>). Found, %:

C 65.59; H 6.58; N 10.17.  $C_{15}H_{18}N_2O_3$ . Calculated, %: C 65.67; H 6.61; N 10.21.

Compounds **Ib–II** were synthesized in a similar way.

**4-Acetyl-1-(2-aminopropyl)-5-(4-fluorophenyl)-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-one (Ib).** Yield 0.91 g (31%), mp >300°C (from ethanol). IR spectrum,  $\nu$ , cm<sup>−1</sup>: 3400 (NH<sub>2</sub>), 1684 (NC=O), 1651 (C=O).  $^1H$  NMR spectrum (CD<sub>3</sub>COOD),  $\delta$ , ppm: 1.27 m (3H, Me), 2.34 s (3H, MeCO), 2.89 m and 2.93 m (1H, 1'-H<sub>A</sub>), 3.92 m and 3.96 m (1H, 1'-H<sub>B</sub>), 3.71 m (1H, 2'-H), 5.42 s and 5.47 s (1H, 5-H), 7.10 t (2H, H<sub>arom</sub>,  $J$  = 8.8 Hz), 7.30 m (2H, H<sub>arom</sub>). Mass spectrum,  $m/z$  ( $I_{rel}$ , %): 292 (10.68) [M]<sup>+</sup>, 249 (100) [M – CH<sub>2</sub>=CHNH<sub>2</sub>]<sup>+</sup>, 164 (11.67) [4-FC<sub>6</sub>H<sub>4</sub>CH=CHCOMe]<sup>+</sup>, 149 (63.31) [4-FC<sub>6</sub>H<sub>4</sub>CH=CHCO]<sup>+</sup>, 57 (19.22) [CH<sub>2</sub>=C(Me)NH<sub>2</sub>]<sup>+</sup>. Found, %: C 61.59; H 5.87; N 9.50.  $C_{15}H_{17}FN_2O_3$ . Calculated, %: C 61.63; H 5.86; N 9.59.

**4-Acetyl-1-(2-aminopropyl)-5-(4-chlorophenyl)-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-one (Ic).** Yield 2.01 g (65%), mp 275–286°C (the product was recrystallized from ethanol and then treated with hot acetone). IR spectrum,  $\nu$ , cm<sup>−1</sup>: 3340 (NH<sub>2</sub>), 1700 (NC=O), 1613 (C=O).  $^1H$  NMR spectrum (DMSO-*d*<sub>6</sub>–CF<sub>3</sub>COOH),  $\delta$ , ppm: 1.02 m (3H, Me), 2.24 s (3H, MeCO), 2.51 m and 2.71 m (1H, 1'-H<sub>A</sub>), 3.50 m and 3.64 m (1H, 1'-H<sub>B</sub>), 3.33 m (1H, 2'-H), 5.23 s and 5.27 s (1H, 5-H), 7.34 m (2H, H<sub>arom</sub>), 7.20 d (2H, H<sub>arom</sub>,  $J$  = 8.1 Hz), 7.82 br.s (2H, NH<sub>2</sub>). Found, %: C 58.33; H 5.50; N 9.01.  $C_{15}H_{17}ClN_2O_3$ . Calculated, %: C 58.35; H 5.55; N 9.07.

**4-Acetyl-1-(2-aminopropyl)-3-hydroxy-5-(4-methoxyphenyl)-2,5-dihydro-1*H*-pyrrol-2-one (Id).** Yield 1.40 g (46%), mp 255–266°C (the product was recrystallized from ethanol and then treated with hot acetone). IR spectrum,  $\nu$ , cm<sup>−1</sup>: 3440 (NH<sub>2</sub>), 3200 (OH), 1655 (NC=O), 1620 (C=O).  $^1H$  NMR spectrum (DMSO-*d*<sub>6</sub>–CF<sub>3</sub>COOH),  $\delta$ , ppm: 1.01 m (3H, Me), 2.23 s and 2.24 s (3H, MeCO), 2.50 m and 2.73 m (1H, 1'-H<sub>A</sub>), 3.46 m and 3.60 m (1H, 1'-H<sub>B</sub>), 3.33 m (1H, 2'-H), 3.68 s (3H, MeO), 5.16 s and 5.20 s (1H, 5-H), 6.83 d (2H, H<sub>arom</sub>,  $J$  = 7.5 Hz), 7.09 m (2H, H<sub>arom</sub>), 7.82 br.s (2H, NH<sub>2</sub>). Found, %: C 63.15; H 6.57; N 9.15.  $C_{16}H_{20}N_2O_4$ . Calculated, %: C 63.14; H 6.62; N 9.21.

**4-Acetyl-1-(2-aminopropyl)-5-(3,4-dimethoxyphenyl)-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-one (Ie).** Yield 1.00 g (30%), mp >300°C (the product was recrystallized from DMF and then treated with hot

ethanol). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3300 ( $\text{NH}_2$ ), 1694 ( $\text{NC=O}$ ), 1646 ( $\text{C=O}$ ).  $^1\text{H}$  NMR spectrum ( $\text{DMSO}-d_6$ ),  $\delta$ , ppm: 1.09 m (3H, Me), 2.13 s and 2.14 s (3H,  $\text{MeCO}$ ), 2.80 m (1H, 1'- $H_A$ ), 3.35 m and 3.64 m (1H, 1'- $H_B$ ), 3.41 m (1H, 2'-H), 3.71 s and 3.74 s (3H each,  $\text{MeO}$ ), 4.98 s and 5.02 s (1H, 5-H), 6.70 m (2H,  $\text{H}_{\text{arom}}$ ), 6.80 m (1H,  $\text{H}_{\text{arom}}$ ), 8.11 br.s (2H,  $\text{NH}_2$ ). Found, %: C 61.00; H 6.60; N 8.33.  $\text{C}_{17}\text{H}_{22}\text{N}_2\text{O}_5$ . Calculated, %: C 61.06; H 6.63; N 8.38.

**4-Acetyl-1-(2-aminopropyl)-3-hydroxy-5-(4-nitrophenyl)-2,5-dihydro-1*H*-pyrrol-2-one (If).** Yield 2.17 g (68%), mp >300°C (from propan-2-ol). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1682 ( $\text{NC=O}$ ), 1646 ( $\text{C=O}$ ).  $^1\text{H}$  NMR spectrum ( $\text{DMSO}-d_6$ ),  $\delta$ , ppm: 1.10 m (3H, Me), 2.10 s and 2.11 s (3H,  $\text{MeCO}$ ), 2.52 m and 2.64 m (1H, 1'- $H_A$ ), 3.60 m and 3.73 m (1H, 1'- $H_B$ ), 3.44 m (1H, 2'-H), 5.20 s and 5.23 s (1H, 5-H), 7.41 m (2H,  $\text{H}_{\text{arom}}$ ), 8.10 m (2H,  $\text{H}_{\text{arom}}$ ), 8.16 br.s (2H,  $\text{NH}_2$ ). Found, %: C 56.41; H 5.31; N 13.17.  $\text{C}_{15}\text{H}_{17}\text{N}_3\text{O}_5$ . Calculated, %: C 56.42; H 5.37; N 13.16.

**4-Acetyl-1-(2-aminopropyl)-3-hydroxy-5-(3-nitrophenyl)-2,5-dihydro-1*H*-pyrrol-2-one (Ig).** Yield 2.14 g (67%), mp >300°C (the product was recrystallized from DMF and then treated with hot ethanol). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3450 ( $\text{NH}_2$ ), 3160 (OH), 1700 ( $\text{NC=O}$ ), 1610 ( $\text{C=O}$ ).  $^1\text{H}$  NMR spectrum ( $\text{DMSO}-d_6-\text{CF}_3\text{COOH}$ ),  $\delta$ , ppm: 1.01 m (3H, Me), 2.26 s (3H,  $\text{MeCO}$ ), 2.52 m and 2.72 m (1H, 1'- $H_A$ ), 3.54 m and 3.67 m (1H, 1'- $H_B$ ), 3.27 m and 3.46 m (1H, 2'-H), 5.41 s and 5.47 s (1H, 5-H), 7.58 m (2H,  $\text{H}_{\text{arom}}$ ), 8.23 m (2H,  $\text{H}_{\text{arom}}$ ), 7.77 br.s (2H,  $\text{NH}_2$ ). Found, %: C 56.42; H 5.31; N 13.10.  $\text{C}_{15}\text{H}_{17}\text{N}_3\text{O}_5$ . Calculated, %: C 56.42; H 5.37; N 13.16.

**1-(2-Aminopropyl)-4-benzoyl-3-hydroxy-5-phenyl-2,5-dihydro-1*H*-pyrrol-2-one (Ih).** Yield 1.41 g (42%), mp 254–257°C (from ethanol). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3236 (OH), 1697 ( $\text{NC=O}$ ), 1635 ( $\text{C=O}$ ).  $^1\text{H}$  NMR spectrum ( $\text{DMSO}-d_6$ ),  $\delta$ , ppm: 1.08 m (3H, Me), 2.62 m and 2.90 m (1H, 1'- $H_A$ ), 3.40 m and 3.64 m (1H, 1'- $H_B$ ), 3.22 m (1H, 2'-H), 5.27 s and 5.32 s (1H, 5-H), 7.24 m (2H,  $\text{H}_{\text{arom}}$ ), 7.77 m (8H,  $\text{H}_{\text{arom}}$ ), 8.18 br.s (2H,  $\text{NH}_2$ ). Mass spectrum,  $m/z$  ( $I_{\text{rel}}$ , %): 336 (2) [ $M]^+$ , 293 (28) [ $M - \text{CH}_2=\text{CHNH}_2]^+$ , 208 (9.50) [ $\text{PhCH=CHCOPh}]^+$ , 105 (50) [ $\text{PhCO}]^+$ , 77 (30.50) [ $\text{Ph}]^+$ , 57 (6.50) [ $\text{CH}_2=\text{C}(\text{Me})\text{NH}_2]^+$ , 44 (100) [ $\text{CH}_2=\text{CHNH}_3]^+$ . Found, %: C 71.37; H 5.95; N 8.30.  $\text{C}_{20}\text{H}_{20}\text{N}_2\text{O}_3$ . Calculated, %: C 71.41; H 5.99; N 8.33.

**1-(2-Aminopropyl)-4-benzoyl-5-(4-chlorophenyl)-3-hydroxy-2,5-dihydro-1*H*-pyrrol-2-one (Ii).** Yield 1.52 g (41%), mp 240–242°C (from ethanol). IR

spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3450 ( $\text{NH}_2$ ), 3230 (OH), 1688 ( $\text{NC=O}$ ), 1620 ( $\text{C=O}$ ).  $^1\text{H}$  NMR spectrum ( $\text{DMSO}-d_6$ ),  $\delta$ , ppm: 0.98 m (3H, Me), 2.50 m and 2.78 m (1H, 1'- $H_A$ ), 3.41 m and 3.59 m (1H, 1'- $H_B$ ), 3.24 m (1H, 2'-H), 5.18 s and 5.24 s (1H, 5-H), 7.23 m (7H,  $\text{H}_{\text{arom}}$ ), 7.65 m (2H,  $\text{H}_{\text{arom}}$ ), 7.85 br.s (2H,  $\text{NH}_2$ ). Found, %: C 64.76; H 5.10; N 7.52.  $\text{C}_{20}\text{H}_{19}\text{ClN}_2\text{O}_3$ . Calculated, %: C 64.78; H 5.16; N 7.56.

**1-(2-Aminopropyl)-4-benzoyl-3-hydroxy-5-(4-methoxyphenyl)-2,5-dihydro-1*H*-pyrrol-2-one (Ij).** Yield 2.97 g (81%), mp 235–241°C (the product was recrystallized from DMF and then treated with hot ethanol). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3330 ( $\text{NH}_2$ ), 3240 (OH), 1679 ( $\text{NC=O}$ ), 1620 ( $\text{C=O}$ ).  $^1\text{H}$  NMR spectrum ( $\text{DMSO}-d_6-\text{CF}_3\text{COOH}$ ),  $\delta$ , ppm: 1.04 m (3H, Me), 2.60 m and 2.86 m (1H, 1'- $H_A$ ), 3.53 m and 3.68 m (1H, 1'- $H_B$ ), 3.35 m (1H, 2'-H), 3.65 s (3H, 4-MeO), 5.45 s and 5.49 s (1H, 5-H), 6.83 d (2H,  $\text{H}_{\text{arom}}$ ,  $J = 8.7$  Hz), 7.25 m (2H,  $\text{H}_{\text{arom}}$ ), 7.40 t (2H,  $\text{H}_{\text{arom}}$ ,  $J = 7.8$  Hz), 7.58 m (3H,  $\text{H}_{\text{arom}}$ ), 7.80 br.s (2H,  $\text{NH}_2$ ). Found, %: C 68.82; H 6.03; N 7.63.  $\text{C}_{21}\text{H}_{22}\text{N}_2\text{O}_4$ . Calculated, %: C 68.83; H 6.05; N 7.65.

**1-(2-Aminopropyl)-4-benzoyl-3-hydroxy-5-(4-nitrophenyl)-2,5-dihydro-1*H*-pyrrol-2-one (Ik).** Yield 3.17 g (83%), mp 250–252°C (from ethanol). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3380 ( $\text{NH}_2$ ), 3240 (OH), 1702 ( $\text{NC=O}$ ), 1633 ( $\text{C=O}$ ).  $^1\text{H}$  NMR spectrum ( $\text{CD}_3\text{COOD}$ ),  $\delta$ , ppm: 1.31 m (3H, Me), 3.00 m and 3.24 m (1H, 1'- $H_A$ ), 3.88 m and 4.09 m (1H, 1'- $H_B$ ), 3.80 m (1H, 2'-H), 5.89 s and 5.95 s (1H, 5-H), 7.41 m (2H,  $\text{H}_{\text{arom}}$ ), 7.58 m (3H,  $\text{H}_{\text{arom}}$ ), 7.75 d (2H,  $\text{H}_{\text{arom}}$ ,  $J = 7.2$  Hz), 8.19 m (2H,  $\text{H}_{\text{arom}}$ ). Mass spectrum,  $m/z$  ( $I_{\text{rel}}$ , %): 381 (3.69) [ $M]^+$ , 338 (49.06) [ $M - \text{CH}_2=\text{CHNH}_2]^+$ , 253 (11.84) [ $4-\text{O}_2\text{NC}_6\text{H}_4\text{CH=CHCOPh}]^+$ , 105 (100) [ $\text{PhCO}]^+$ , 77 (38.48) [ $\text{Ph}]^+$ , 57 (17.60) [ $\text{CH}_2=\text{C}(\text{Me})\text{NH}_2]^+$ . Found, %: C 62.89; H 5.00; N 10.96.  $\text{C}_{20}\text{H}_{19}\text{N}_3\text{O}_5$ . Calculated, %: C 62.98; H 5.02; N 11.02.

**1-(2-Aminopropyl)-4-benzoyl-3-hydroxy-5-(3-nitrophenyl)-2,5-dihydro-1*H*-pyrrol-2-one (Il).** Yield 1.41 g (37%), mp 240–242°C (the product was recrystallized from ethanol and then treated with hot acetone). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3240 (OH), 1718 ( $\text{NC=O}$ ), 1620 ( $\text{C=O}$ ).  $^1\text{H}$  NMR spectrum ( $\text{DMSO}-d_6-\text{CF}_3\text{COOH}$ ),  $\delta$ , ppm: 1.05 m (3H, Me), 2.63 m and 2.84 m (1H, 1'- $H_A$ ), 3.60 m and 3.75 m (1H, 1'- $H_B$ ), 3.36 m and 3.51 m (1H, 2'-H), 5.69 s and 5.73 s (1H, 5-H), 7.39 m (2H,  $\text{H}_{\text{arom}}$ ), 7.56 m (4H,  $\text{H}_{\text{arom}}$ ), 7.79 m (1H,  $\text{H}_{\text{arom}}$ ), 8.11 m (1H,  $\text{H}_{\text{arom}}$ ), 8.28 s (1H,  $\text{H}_{\text{arom}}$ ), 7.84 br.s (2H,  $\text{NH}_2$ ). Found, %: C 62.89; H 4.98; N 11.00.  $\text{C}_{20}\text{H}_{19}\text{N}_3\text{O}_5$ . Calculated, %: C 62.98; H 5.02; N 11.02.

**4-Acetyl-1-(2-aminopropyl)-3-hydroxy-5-phenyl-2,5-dihydro-1*H*-pyrrol-2-one hydrochloride (IIa).**

Compound **Ia**, 0.01 mol, was dissolved in 40 ml of concentrated hydrochloric acid on heating on a boiling water bath, the solution was evaporated to dryness, and the residue was recrystallized. Yield 1.77 g (57%), mp 219–223°C (from ethanol). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3500, 3390 ( $^+\text{NH}_3$ ), 3180 (OH), 1686 (NC=O), 1630 (C=O).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 1.03 d (3H, Me,  $J = 5.7$  Hz), 2.22 s (3H, MeCO), 2.51 m (1H, 1'-H), 3.63 m (1H, 1'-H<sub>B</sub>), 3.28 m (1H, 2'-H), 5.22 s (1H, 5-H), 7.22 m (5H, H<sub>arom</sub>), 8.05 br.s (3H,  $^+\text{NH}_3$ ). Found, %: C 57.94; H 6.12; N 9.02.  $\text{C}_{15}\text{H}_{18}\text{N}_2\text{O}_3 \cdot \text{HCl}$ . Calculated, %: C 57.97; H 6.16; N 9.02.

**1-(2-Aminopropyl)-4-benzoyl-3-hydroxy-5-phenyl-2,5-dihydro-1*H*-pyrrol-2-one hydrochloride (IIb)** was synthesized in a similar way. Yield 2.78 g (75%), mp 246–249°C (from ethanol). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3440 ( $^+\text{NH}_3$ ), 3160 (OH), 1695 (NC=O), 1626 (C=O).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ –CF<sub>3</sub>COOH),  $\delta$ , ppm: 1.09 d (3H, Me,  $J = 6.6$  Hz), 2.66 m and 2.92 m (1H, 1'-H<sub>A</sub>), 3.59 m and 3.75 m (1H, 1'-H<sub>B</sub>), 3.38 m (1H, 2'-H), 5.54 s and 5.56 s (1H, 5-H), 7.32 m (5H, H<sub>arom</sub>), 7.41 m (2H, H<sub>arom</sub>), 7.53 m (1H, H<sub>arom</sub>), 7.68 m (2H, H<sub>arom</sub>), 8.00 br.s (3H,  $^+\text{NH}_3$ ). Found, %: C 64.35; H 5.65; N 7.44.  $\text{C}_{20}\text{H}_{20}\text{N}_2\text{O}_3 \cdot \text{HCl}$ . Calculated, %: C 64.42; H 5.68; N 7.52.

**1,1'-(Propane-1,2-diyl)bis(4-acetyl-3-hydroxy-5-phenyl-2,5-dihydro-1*H*-pyrrol-2-one) (III).** A solution of 0.01 mol of methyl 2,4-dioxopentanoate in 10 ml of 1,4-dioxane was added at room temperature to a solution of 0.01 mol of benzaldehyde and 0.005 mol of propane-1,2-diamine in 10 ml of 1,4-dioxane. The mixture was kept for 24 h at room temperature, and the precipitate was filtered off and purified by recrystallization. Yield 0.67 g (28%), mp >300°C (from glacial acetic acid). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3160 (OH), 1685 (NC=O), 1650 (C=O).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 1.02 d (3H, Me,  $J = 7.0$  Hz), 2.22 s and 2.24 s (3H, MeCO), 2.26 s and 2.28 s (3H, MeCO), 2.73 m and 3.49 m (1H each, CH<sub>2</sub>), 3.71 m (1H, CHMe), 4.68 s and 4.79 s (1H, 5-H), 5.15 s and 5.23 s (1H, 5-H), 7.12 m (4H, H<sub>arom</sub>), 7.21 m (6H, H<sub>arom</sub>). Mass spectrum,  $m/z$  ( $I_{\text{rel}}$ , %): 474 (9.68) [M]<sup>+</sup>, 244 (38.80) [AcC<sub>4</sub>NO<sub>2</sub>(Ph)Et]<sup>+</sup>, 173 (100) [MeCH=NCH(Ph)CH=CO]<sup>+</sup>, 131 (68.06) [PhCH=CHCO]<sup>+</sup>, 77 (5.61) [Ph]<sup>+</sup>, 56 (8.08) [CH<sub>2</sub>=C(Me)N]<sup>+</sup>. Found, %: C 68.30; H 5.46; N 5.90.  $\text{C}_{27}\text{H}_{26}\text{N}_2\text{O}_6$ . Calculated, %: C 68.34; H 5.52; N 5.91.

**1-(2-Aminopropyl)-4-benzoyl-3-[4-(dimethylamino)phenylamino]-5-phenyl-2,5-dihydro-1*H*-pyr-**

rol-2-one hydrochloride (IV). A mixture of 5 mmol of compound **II** and 5 mmol of *p*-dimethylaminoaniline in 12 ml of glacial acetic acid was heated for 1.5 h under reflux. The mixture was cooled and evaporated, and the residue was treated with hexane and recrystallized. Yield 0.52 g (21%), mp 232–236°C (the product was recrystallized from propan-2-ol and then treated with hot chloroform). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3470 ( $^+\text{NH}_3$ ), 3310 (NH), 1688 (NC=O), 1642 (C=O).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 1.06 m (3H, Me), 2.55 m and 2.82 m (1H, 1'-H<sub>A</sub>), 2.73 s (6H, NMe<sub>2</sub>), 3.37 m and 3.70 m (1H, 1'-H<sub>B</sub>), 3.30 m (1H, 2'-H), 5.69 s (1H, 5-H), 6.39 m (2H, H<sub>arom</sub>), 6.83 d (2H, H<sub>arom</sub>,  $J = 8.4$  Hz), 7.16 m (10H, H<sub>arom</sub>), 8.09 s (3H,  $^+\text{NH}_3$ ), 9.23 s (1H, NH). Found, %: C 68.50; H 6.30; N 11.38.  $\text{C}_{28}\text{H}_{30}\text{N}_4\text{O}_2 \cdot \text{HCl}$ . Calculated, %: C 68.49; H 6.36; N 11.41.

Compounds **Va–Vc** were synthesized in a similar way.

**1-(2-Aminopropyl)-4-[1-(4-methoxyphenylamino)-ethylidene]-5-phenyltetrahydropyrrole-2,3-dione hydrochloride (Va).** Yield 1.19 g (57%), mp 198–202°C (from toluene). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3400 br (NH,  $^+\text{NH}_3$ ), 1700 (NC=O), 1650 (C=O).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 1.07 m (3H, Me), 1.71 s and 1.72 s (3H, MeC=C), 2.53 m and 2.80 m (1H, 1'-H<sub>A</sub>), 3.49 m and 3.65 m (1H, 1'-H<sub>B</sub>), 3.26 m (1H, 2'-H), 3.70 s (3H, MeO), 5.47 s and 5.48 s (1H, 5-H), 6.91 m (2H, H<sub>arom</sub>), 7.14 m (2H, H<sub>arom</sub>), 7.34 m (5H, H<sub>arom</sub>), 8.10 s (3H,  $^+\text{NH}_3$ ), 12.44 s (1H, NH). Found, %: C 63.50; H 6.28; N 10.04.  $\text{C}_{22}\text{H}_{25}\text{N}_3\text{O}_3 \cdot \text{HCl}$ . Calculated, %: C 63.53; H 6.30; N 10.10.

**1-(2-Aminopropyl)-4-[1-(4-ethoxyphenylamino)-ethylidene]-5-phenyltetrahydropyrrole-2,3-dione (Vb).** Yield 0.24 g (12%), mp 151–155°C (the product was recrystallized from acetonitrile and then treated with hot toluene). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3500 (NH<sub>2</sub>), 3300 (NH), 1700 (NC=O), 1640 (C=O).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 0.83 d (3H, Me,  $J = 6.0$  Hz), 1.25 m (3H, CH<sub>3</sub>CH<sub>2</sub>), 1.70 s (3H, MeC=C), 2.34 m (1H, 1'-H<sub>A</sub>), 3.32 m (1H, 1'-H<sub>B</sub>), 2.97 m (1H, 2'-H), 3.95 m (2H, OCH<sub>2</sub>), 5.42 s (1H, 5-H), 6.87 d (2H, H<sub>arom</sub>,  $J = 7.5$  Hz), 7.22 m (7H, H<sub>arom</sub>). Mass spectrum,  $m/z$  ( $I_{\text{rel}}$ , %): 393 (19.90) [M]<sup>+</sup>, 350 (28.68) [M – CH<sub>2</sub>=CHNH<sub>2</sub>]<sup>+</sup>, 137 (56.09) [4-EtOC<sub>6</sub>H<sub>4</sub>NH<sub>2</sub>]<sup>+</sup>, 108 (100) [*p*-O=C<sub>6</sub>H<sub>4</sub>=NH<sub>2</sub>]<sup>+</sup>. Found, %: C 70.18; H 6.89; N 10.64.  $\text{C}_{23}\text{H}_{27}\text{N}_3\text{O}_3$ . Calculated, %: C 70.20; H 6.92; N 10.68.

**1-(2-Aminopropyl)-4-[1-(4-ethoxyphenylamino)-ethylidene]-5-phenyltetrahydropyrrole-2,3-dione**

**hydrochloride (Vc).** Yield 1.59 g (74%), mp 242–244°C (from propan-2-ol). IR spectrum,  $\nu$ , cm<sup>-1</sup>: 3400 (<sup>+</sup>NH<sub>3</sub>), 3200 (NH), 1710 (C=O), 1630 (C=O). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 1.05 m (3H, Me), 1.26 m (3H, CH<sub>3</sub>CH<sub>2</sub>), 1.71 s and 1.72 s (3H, MeC=C), 2.52 m (1H, 1'-H<sub>A</sub>), 3.65 m (1H, 1'-H<sub>B</sub>), 3.30 m (1H, 2'-H), 3.96 m (2H, OCH<sub>2</sub>), 5.45 s and 5.46 s (1H, 5-H), 6.88 m (2H, H<sub>arom</sub>), 7.11 m (2H, H<sub>arom</sub>), 7.32 m (5H, H<sub>arom</sub>), 8.02 s (3H, <sup>+</sup>NH<sub>3</sub>), 12.43 s (1H, NH). Found, %: C 64.20; H 6.53; N 9.71. C<sub>23</sub>H<sub>27</sub>N<sub>3</sub>O<sub>3</sub>·HCl. Calculated, %: C 64.25; H 6.56; N 9.77.

**1-(2-Aminopropyl)-5-phenyl-4-(1-phenylhydrazinoethyl)pyrrole-2,3-dione hydrochloride (VI).** A solution of 5 mmol compound **IIa** in 30 ml of ethanol was heated to the boiling point, 5.5 mmol of phenylhydrazine was added, the mixture was kept for 24 h at room temperature, and the precipitate was filtered off and recrystallized. Yield 0.28 g (14%), mp 299–304°C (from ethanol). IR spectrum,  $\nu$ , cm<sup>-1</sup>: 3440 (<sup>+</sup>NH<sub>3</sub>), 3255 (NH), 1678 (C=O, NC=O). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 1.05 d (3H, Me, *J* = 6.0 Hz), 1.29 s (3H, MeC=N), 2.52 m (1H, 1'-H<sub>A</sub>), 3.58 m (1H, 1'-H<sub>B</sub>), 3.30 m (1H, 2'-H), 4.04 d (1H, 5-H, *J* = 9.9 Hz), 5.15 d (1H, 4-H, *J* = 9.3 Hz), 6.76 t (1H, H<sub>arom</sub>, *J* = 7.2 Hz), 7.12 m (2H, H<sub>arom</sub>), 7.30 m (5H, H<sub>arom</sub>), 7.59 d (2H, H<sub>arom</sub>, *J* = 7.8 Hz), 8.00 br.s (3H, <sup>+</sup>NH<sub>3</sub>). Found, %: C 62.82; H 6.26; N 13.92. C<sub>21</sub>H<sub>24</sub>N<sub>4</sub>O<sub>2</sub>·HCl. Calculated, %: C 62.91; H 6.29; N 13.98.

**5-(2-Aminopropyl)-4-(4-chlorophenyl)-3-methyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one (VIIa).** A mixture of 5 mmol of compound **Ic** and 5.5 mmol of 98% hydrazine in 5 ml of glacial acetic acid was heated for 1.5 h under reflux. The mixture was cooled and evaporated, and the residue was treated with hexane and recrystallized. Yield 0.38 g (25%), mp >300°C (from acetonitrile). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 0.89 m (3H, Me), 1.94 s (3H, MeC=N), 2.56 m and 3.38 m (1H each, 5-CH<sub>2</sub>), 3.04 m (1H, 5-CH<sub>2</sub>CH), 5.61 d (1H, 4-H), 7.16 m (2H, H<sub>arom</sub>), 7.38 m (2H, H<sub>arom</sub>). Found, %: C 59.07; H 5.60; N 18.36. C<sub>15</sub>H<sub>16</sub>N<sub>4</sub>O·HCl. Calculated, %: C 59.11; H 5.62; N 18.38.

Compounds **VIIb–VIIIi** and **VIII** were synthesized in a similar way.

**5-(2-Aminopropyl)-4-(4-methoxyphenyl)-3-methyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one (VIIb).** Yield 0.74 g (49%), mp 59–62°C (the product was recrystallized from acetonitrile and then

treated with hot ethanol). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 0.91 m (3H, Me), 1.97 s (3H, MeC=N), 2.59 m and 3.41 m (1H each, 5-CH<sub>2</sub>), 2.98 m (1H, CH<sub>2</sub>CH), 3.70 s (3H, MeO), 5.50 d (1H, 4-H), 6.82 m (2H, H<sub>arom</sub>), 7.01 m (2H, H<sub>arom</sub>), 7.61 br.s (2H, NH<sub>2</sub>). Found, %: C 63.93; H 6.70; N 18.59. C<sub>16</sub>H<sub>20</sub>N<sub>4</sub>O<sub>2</sub>. Calculated, %: C 63.98; H 6.71; N 18.65.

**5-(2-Aminopropyl)-3-methyl-1,4-diphenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one (VIIc).** Yield 0.40 g (23%), mp 143–147°C (from diethyl ether). IR spectrum,  $\nu$ , cm<sup>-1</sup>: 3460 (NH<sub>2</sub>), 1685 (C=O). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 1.01 m (3H, Me), 2.04 s (3H, MeC=N), 2.60 m and 3.10 m (1H each, 5-CH<sub>2</sub>), 3.60 m (1H, 5-CH<sub>2</sub>CH), 5.77 d (1H, 4-H), 7.27 m (4H, H<sub>arom</sub>), 7.38 m (4H, H<sub>arom</sub>), 7.45 m (2H, H<sub>arom</sub>), 8.29 br.s (2H, NH<sub>2</sub>). Found, %: C 72.79; H 6.35; N 16.10. C<sub>21</sub>H<sub>22</sub>N<sub>4</sub>O. Calculated, %: C 72.80; H 6.40; N 16.17.

**5-(2-Aminopropyl)-3-methyl-1,4-diphenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one hydrochloride (VId).** Yield 1.51 g (79%), mp >300°C (from propan-2-ol). IR spectrum,  $\nu$ , cm<sup>-1</sup>: 3460 (<sup>+</sup>NH<sub>3</sub>), 1685 (C=O). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 1.14 d (3H, Me), 2.01 s (3H, MeC=N), 2.80 m and 3.84 m (1H each, 5-CH<sub>2</sub>), 3.43 m (1H, 5-CH<sub>2</sub>CH), 5.79 s (1H, 4-H), 7.34 m (3H, H<sub>arom</sub>), 7.42 m (3H, H<sub>arom</sub>), 7.53 t (2H, H<sub>arom</sub>, *J* = 7.2 Hz), 8.21 d (2H, H<sub>arom</sub>, *J* = 7.8 Hz), 7.92 br.s (3H, <sup>+</sup>NH<sub>3</sub>). Found, %: C 65.83; H 6.07; N 14.59. C<sub>21</sub>H<sub>22</sub>N<sub>4</sub>O·HCl. Calculated, %: C 65.87; H 6.05; N 14.63.

**5-(2-Aminopropyl)-3,4-diphenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one (VIIe).** Yield 0.90 g (54%), mp 172–177°C (from acetonitrile). IR spectrum,  $\nu$ , cm<sup>-1</sup>: 1685 (C=O), 3280 (NH), 3350 (NH<sub>2</sub>). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 0.85 m (3H, Me), 2.40 m and 3.41 m (1H each, 5-CH<sub>2</sub>), 2.94 m (1H, 5-CH<sub>2</sub>CH), 5.93 s and 5.96 s (1H, 4-H), 7.24 m (8H, H<sub>arom</sub>), 7.42 m (2H, H<sub>arom</sub>). Mass spectrum, *m/z* (*I*<sub>rel</sub>, %): 332 (3.84) [M]<sup>+</sup>, 289 (48.47) [M - CH<sub>2</sub>=CHNH<sub>2</sub>]<sup>+</sup>, 260 (100) [M - CH<sub>3</sub>CH=NH<sub>2</sub> + N<sub>2</sub>]<sup>+</sup>, 77 (13.26) [Ph]<sup>+</sup>, 57 (4.49) [CH<sub>2</sub>=C(Me)NH<sub>2</sub>]<sup>+</sup>. Found, %: C 72.27; H 6.00; N 16.84. C<sub>20</sub>H<sub>20</sub>N<sub>4</sub>O. Calculated, %: C 72.26; H 6.06; N 16.86.

**5-(2-Aminopropyl)-3,4-diphenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one hydrochloride (VIf).** Yield 0.42 g (23%), mp 230–234°C (from ethanol). IR spectrum,  $\nu$ , cm<sup>-1</sup>: 3430 (<sup>+</sup>NH<sub>3</sub>), 3210 (NH), 1710 (C=O). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 1.08 d (3H, Me, *J* = 5.7 Hz), 2.63 m and 3.73 m (1H each, 5-CH<sub>2</sub>), 3.31 m (1H, 5-CH<sub>2</sub>CH), 5.97 s (1H, 4-H),

7.27 m (8H, H<sub>arom</sub>), 7.42 d (2H, H<sub>arom</sub>, *J* = 6.6 Hz), 8.00 s (1H, <sup>+</sup>NH<sub>3</sub>), 14.08 br.s (1H, N<sup>1</sup>H). Found, %: C 65.08; H 5.68; N 15.19. C<sub>20</sub>H<sub>20</sub>N<sub>4</sub>O·HCl. Calculated, %: C 65.12; H 5.74; N 15.19.

**5-(2-Aminopropyl)-4-(4-chlorophenyl)-3-phenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one (VIIg).** Yield 0.94 g (51%), mp 135–139°C (from acetonitrile). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>), δ, ppm: 0.96 m (3H, Me), 2.60 m and 3.38 m (1H each, 5-CH<sub>2</sub>), 2.98 m (1H, 5-CH<sub>2</sub>CH), 5.98 d (1H, 4-H), 7.27 m (5H, H<sub>arom</sub>), 7.36 m (2H, H<sub>arom</sub>), 7.42 m (2H, H<sub>arom</sub>). Found, %: C 65.46; H 5.20; N 15.21. C<sub>20</sub>H<sub>18</sub>N<sub>4</sub>O·HCl. Calculated, %: C 65.48; H 5.22; N 15.27.

**5-(2-Aminopropyl)-4-(4-methoxyphenyl)-3-phenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one (VIIh).** Yield 0.69 g (38%), mp 153–158°C (from acetonitrile). IR spectrum, ν, cm<sup>-1</sup>: 3350 (NH<sub>2</sub>), 3200 (NH), 1690 (C=O). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>–CF<sub>3</sub>COOH), δ, ppm: 1.05 m (3H, Me), 2.67 m and 2.87 m (1H, 5-CH<sub>A</sub>), 3.37 m and 3.60 m (1H, 5-CH<sub>B</sub>), 3.20 m (1H, 5-CH<sub>2</sub>CH), 3.67 s (3H, MeO), 5.84 s and 5.87 s (1H, 4-H), 6.86 m (2H, H<sub>arom</sub>), 7.23 m (5H, H<sub>arom</sub>), 7.41 m (2H, H<sub>arom</sub>), 7.78 br.s (2H, NH<sub>2</sub>). Found, %: C 69.60; H 6.08; N 15.40. C<sub>21</sub>H<sub>22</sub>N<sub>4</sub>O<sub>2</sub>. Calculated, %: C 69.59; H 6.12; N 15.46.

**5-(2-Aminopropyl)-4-(3-nitrophenyl)-3-phenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(1*H*)-one (VIIIi).** Yield 1.13 g (60%), mp 179–182°C (from acetonitrile). IR spectrum, ν, cm<sup>-1</sup>: 3400 (NH<sub>2</sub>), 3290 (NH), 1700 (C=O). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>–CF<sub>3</sub>COOH), δ, ppm: 1.11 m (3H, Me), 2.66 m and 2.93 m (1H, 5-CH<sub>A</sub>), 3.70 m and 3.82 m (1H, 5-CH<sub>B</sub>), 3.28 m and 3.48 d.m (1H, 5-CH<sub>2</sub>CH), 6.22 s and 6.25 s (1H, 4-H), 7.31 m (3H, H<sub>arom</sub>), 7.47 m (2H, H<sub>arom</sub>), 7.66 m (2H, H<sub>arom</sub>), 8.18 m (1H, H<sub>arom</sub>), 8.31 s (1H, H<sub>arom</sub>), 7.85 br.s (2H, NH<sub>2</sub>). Mass spectrum, *m/z* (*I*<sub>rel</sub>, %): 377 (16.67) [M]<sup>+</sup>, 359 (4.30) [M – H<sub>2</sub>O]<sup>+</sup>, 334 (100) [M – CH<sub>2</sub>=CHNH<sub>2</sub>]<sup>+</sup>, 305 (92.55) [M – CH<sub>3</sub>CH=NH<sub>2</sub> + N<sub>2</sub>]<sup>+</sup>, 77 (43.75) [Ph]<sup>+</sup>, 57 (27.74) [CH<sub>2</sub>C(Me)NH<sub>2</sub>]<sup>+</sup>. Found, %: C 63.59; H 5.05; N 18.52. C<sub>20</sub>H<sub>19</sub>N<sub>5</sub>O<sub>3</sub>. Calculated, %: C 63.65; H 5.07; N 18.56.

**5-(2-Aminopropyl)-2,3,4-triphenyl-4,5-dihydropyrrolo[3,4-*c*]pyrazol-6(2*H*)-one hydrochloride (VIII).** Yield 0.67 g (30%), mp >300°C (the product was recrystallized from DMF and was then treated with hot ethanol). IR spectrum, ν, cm<sup>-1</sup>: 3500 (<sup>+</sup>NH<sub>3</sub>), 1675 (C=O). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>), δ, ppm: 1.12 d (3H, Me, *J* = 6.6 Hz), 2.67 m and 3.78 m

(1H each, 5-CH<sub>2</sub>), 3.43 m (1H, 5-CH<sub>2</sub>CH), 6.14 s (1H, 4-H), 7.33 m (9H, H<sub>arom</sub>), 7.55 m (4H, H<sub>arom</sub>), 8.29 m (2H, H<sub>arom</sub>), 8.07 br.s (3H, <sup>+</sup>NH<sub>3</sub>). Mass spectrum, *m/z* (*I*<sub>rel</sub>, %): 408 (6.40) [M – HCl]<sup>+</sup>, 365 (100) [M – (HCl + CH<sub>2</sub>=CHNH<sub>2</sub>)<sup>+</sup>, 336 (39.67) [M – (HCl + CH<sub>3</sub>CH=NH<sub>2</sub> + N<sub>2</sub>)<sup>+</sup>, 77 (33.21) [Ph]<sup>+</sup>. Found, %: C 70.12; H 5.64; N 12.59. C<sub>26</sub>H<sub>24</sub>N<sub>4</sub>O·HCl. Calculated, %: C 70.18; H 5.66; N 12.59.

**X-Ray analysis of compound VIIe.** Compound VIIe crystallized as well defined extended prisms; C<sub>20</sub>H<sub>20</sub>N<sub>4</sub>O, *M* 332.40; monoclinic crystal system; unit cell parameters: *a* = 5.604(1), *b* = 17.543(4), *c* = 17.264(3) Å; β = 90.97(3)°; *V* = 1697.0(6) Å<sup>3</sup>; *d*<sub>calc</sub> = 1.301 g/cm<sup>3</sup>; *Z* = 4; space group *P*2(1)/*n*. Experimental reflection intensities were measured on a KM-4 (Kuma Diffraction) automatic four-circle diffractometer with χ-geometry [monochromatized MoK<sub>α</sub> irradiation, ω/2θ scanning, 2θ<sub>max</sub> = 50.14° (99.6%)]. Total of 3337 reflections were measured, 3010 of which were independent (*R*<sub>int</sub> = 0.0375). No correction for absorption was introduced (μ = 0.083 mm<sup>-1</sup>). The structure was solved by the direct method using SIR92 program [4], followed by a series of calculations of electron density maps. The positions of hydrogen atoms were set on the basis of geometry considerations. The positions of non-hydrogen atoms were refined by the least-squares procedure in full-matrix anisotropic approximation using SHELXL-97 software package [5]; the final divergence factors were *R*<sub>1</sub> = 0.0646 [for 1738 reflections with *I* ≥ 2σ(*I*)] and 0.1366 (for all reflections); goodness of fit 0.968.

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