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2-Acyl(aroyl)-1,1,3,3-tetracyanopropenides: VI.* Reaction with Hydrogen Halides

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Abstract—Reactions of 2-aroyl-1,1,3,3-tetracyanopropenides with hydrogen halides in solvents of low dielectric permittivity result in the formation of 6-amino-2-aroyl-2-halopyridine-3,5-dicarbonitriles. 2-Acyl-1,1,3,3-tetracyanopropenides under similar conditions afford 2-(2-alkylidene-5-amino-4-cyano-2,3-dihydrofuran-3-ylidene)propanedinitriles. In solvents of high dielectric permittivity the result of the reaction depends on the nature of the hydrogen halide and the acyl(aroyl) substituent: With HCl and HBr 2-aroyl-1,1,3,3-tetracyanopropenides form 2-(5-amino-2-aryl-2-halo-4-cyano-2,3-dihydrofuran-3-ylidene)-propanedinitriles, and 2-acyl-1,1,3,3-tetracyanopropenides give 2-(2-alkylidene-5-amino-4-cyano-2,3-dihydrofuran-3-ylidene)propane-dinitriles; with HI depending on the reaction conditions and the structure of the acyl substituent 2-(5-amino-2-aryl-4-cyano-2,3-dihydrofuran-3-ylidene)propanedinitriles, 2-(5-amino-4-cyano-2,3-dihydrofuran-3-ylidene) propane-dinitrile, 2-amino-4-(dimethoxybenzyl)-6-iodo-5-cyanonicotinamide, 4-amino-6-iodo-3-oxo-2,3-dihydro-1*H*-pyrrolo[3,4-*c*]pyridine-7-carbonitrile, or 4-amino-6-iodo-3-oxo-1-ethylidene-1,3-dihydrofuro[3,4-*c*]pyridine-7-carbonitrile are obtained.

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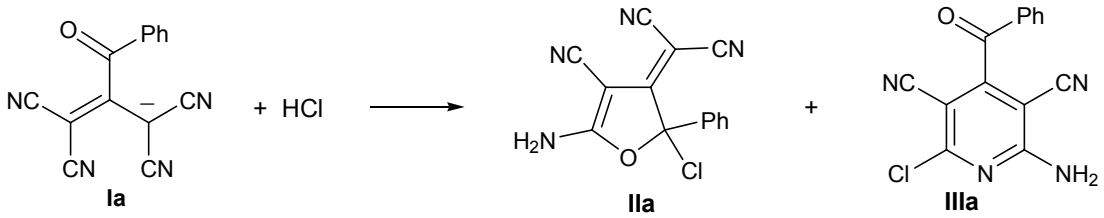
The general and the characteristic property of 1,1,3,3-tetracyanopropenides (salts of cations of alkali metals, ammonium, or organic bases) containing in the position *2* of the allyl system an alkyl, aryl, heteryl substituent, chlorine or bromine atoms or a cyano group is the reaction with hydrogen halides leading to the formation of 2-halopyridine derivatives [2–7]. This reaction path is not a single probable one for 2-acyl-(aroyl)-1,1,3,3-tetracyanopropenides **I** because of the presence in the anion composition of a carbonyl group capable of reactions with nucleophilic reagents in a wide range of the pH of environment. Previous investigations found several possible reaction paths for propenides **I** with hydrogen halides where the primary addition occurred either at the carbonyl or at the cyano group. In reaction in water with HCl and HBr 2-(5-amino-2-halo-4-cyano-2,3-dihydrofuran-3-ylidene)-propanedinitriles were obtained [8, 9]. The result of the reaction between propenides **I** and HI in water environment depended on the temperature: At 60–70°C the main reaction product was 2-(5-amino-4-cyano-2,3-dihydrofuran-3-ylidene)propanedinitrile [9], and at boiling 4-amino-6-iodo-3-oxo-1,3-dihydrofuro[3,4-*c*]-pyridine-7-carbonitrile formed predominantly[10]. In solution of *sec*-butanol the reaction of compound **I** with hydrogen halides afforded 2-amino-6-halo-pyridine-3,5-dicarbonitriles [9].

In this report we publish the results of the study of environment effect. The understanding of this phenomenon is important both for the choice of the optimum reaction conditions and for the prediction of region- and chemoselectivity of the reactions of propenides **I** and structurally similar compounds with the other nucleophilic reagents. Beside the effect of the environment we investigated the influence on the reaction direction of the electronic and steric effects of the aryl and alkyl substituent at the carbonyl group of propenides **I**.

The effect of the solvent nature was investigated by an example of reaction of 2-benzoyl-1,1,3,3-tetracyanopropenide **Ia** with HCl in diverse solvents

* For communication V, see [1].

Scheme 1.



Ia **IIa** **IIIa**

(Scheme 1). The obtained mixture of derivatives of dihydrofuran **IIa** and pyridine **IIIa** was precipitated with water where they were practically insoluble. Their quantitative ratio was established from ^{1}H NMR spectra by comparing the integral intensity of the signals of *o*-protons [doublets at 7.68 (**IIa**) and 8.03 ppm (**IIIa**)] in the phenyl residue.

The direction of the reaction correlates with the dielectric permittivity of the solvent ε (Table 1). In the environment with a large ε compound **IIa** is obtained, and in solvents with the low ε pyridine derivative **IIIa** is formed. In the solvents with intermediate ε values mixtures of compounds **IIa** and **IIIa** were obtained. The ratio of products **IIa** and **IIIa** does not correlate with any other parameters of the solvent (donor and acceptor numbers, dipole moment).

The solvent effect may be related to the state of the protolytic equilibrium of propenide **Ia** in the presence of a strong mineral acid (Scheme 2). Along with anion **Ia** several protonated forms may take part in the equilibrium: enone **A**, ketene imines **B**, **C**, and zwitter-ion **D**.

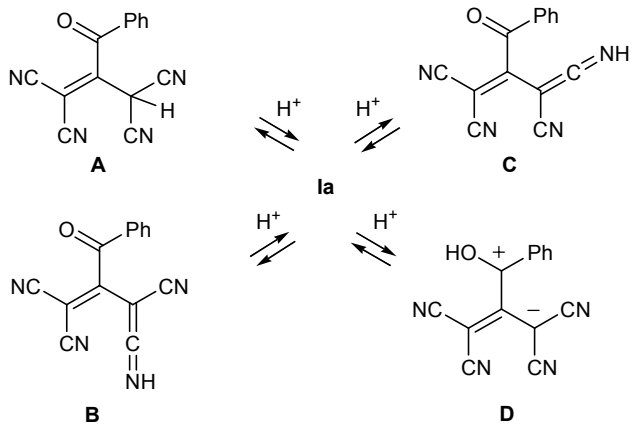
Table 1. Overall yield and percent ratio of derivatives of dihydrofuran **IIa** and pyridine **IIIa** depending on the dielectric permittivity of solvent.

Solvent	Dielectric permittivity, ε	**IIa–IIIa**	Overall yield, %
1,4-Dioxane	2.2	0 : 100	63
Acetic acid	6.2	0 : 100	89
THF	7.4	traces : 100	67
2-Butanol	17.1	traces : 100	76
Acetone	20.7	17 : 83	65
Acetonitrile	37.5	25 : 75	70
Formic acid	51.3	65 : 35	73
Water	78.5	100 : 0	58
Formamide	109.5	100 : 0	64

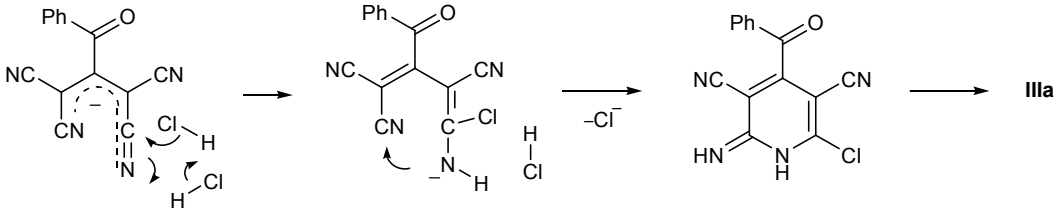
It was suggested previously that the formation of furan or pyridine derivative originated from the site of the primary protonation of compound **I** [8, 9]. The solvents with the large ε provide a better solvation of the structures bearing an electric charge (zwitter-ion **D**) and facilitate the formation of 2-halodihydrofuran derivative **II**. In solvents with the low ε value neutral molecules are more stable (here the forms **A–C**) favoring the formation of 2-halopyridines **III**.

The reaction direction may be also related to the dissociation extent of HCl in the solvent. In the environment with the large ε HCl is mainly dissociated, and the chloride ion serves as the nucleophilic reagent that prevailingly attacks the positively charged carbocation site of the zwitter-ion **D**. In the solvents with the low ε value HCl is insignificantly dissociated and evidently reacts with the protonated molecule of compound **I**. In this case the addition of HCl to the cyano group prevails. Nitriles are known to be able to form molecular complexes with hydrogen halides of the composition 1 : 1 or 1 : 2 where the covalent bond between the hydrogen and halogen atoms is retained followed by the transformation into haloimmonium halides possibly by the mechanism of concerted addition [11]. Various versions of the concerted HCl addition to

Scheme 2.

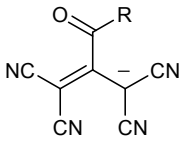


Scheme 3.



cyano group may be considered for propenides **I** both in propenide **I** anion (Scheme 3) and its diverse protonated forms, in particular, zwitter-ion **D**.

As a result of the study we found that the favorable solvent for the synthesis of pyridine derivatives **III**



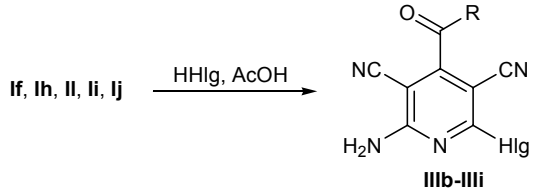
Ib-Il

R = CH_3 (**b**), C_2H_5 (**c**), C_3H_7 (**d**), *t*-Bu (**e**), 3-ClC_6H_4 (**f**), 4-ClC_6H_4 (**g**), 2,4-$Cl_2C_6H_3$ (**h**), 4-$MeOC_6H_4$ (**i**), 3,4-$(MeO)_2C_6H_3$ (**j**), 2,5-$(MeO)_2C_6H_3$ (**k**), 4-$(NO_2)C_6H_4$ (**l**).

was glacial acetic acid, for dihydrofuran compounds **II**, water.

The study elucidated the effect of the nature of aryl and alkyl substituents at the carbonyl group on the course of the reaction between propenides **I** and hydrogen halides. As objects of research 2-acylpropenides **Ib–Ie** were chosen, and also 2-benzoylpropenide derivatives substituted in the benzene ring with chlorine atoms (**If** and **Ig**), with electron-donor groups [*p*-methoxybenzoyl (**Ii**) and dimethoxybenzoyl derivatives (**Ij** and **Ik**)], or electron-acceptor group (*p*-nitrobenzoyl derivative **Il**), and also *ortho*-substituted 2,4-dichlorobenzoylpropenide (**Ih**)].

Scheme 4.



IIIb-IIIi

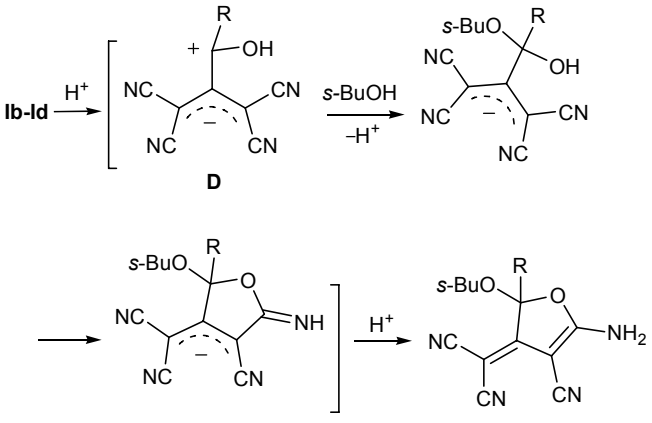
Hlg = Cl: R = 2,4-$Cl_2C_6H_3$ (**b**), 4-$(NO_2)C_6H_4$ (**c**), 4-$MeOC_6H_4$ (**d**), 3,4-$(MeO)_2C_6H_3$ (**e**); Hlg = Br: R = 3-ClC_6H_4 (**f**), 2,4-$Cl_2C_6H_3$ (**g**), 4-$(NO_2)C_6H_4$ (**h**), 4-$MeOC_6H_4$ (**i**).

For aroyl derivatives **If**, **Ih**, **Il**, **Ii** and **Ij** the reaction with HCl and HBr in acetic acid is of general character and affords 2-amino-4-aroyl-6-halopyridine-3,5-dicarbonitriles **IIIb–IIIi** (Scheme 4).

The structure and composition of compounds **IIIb–IIIi** were confirmed by ^{1}H NMR, IR, mass spectra and elemental analysis data. Proton signals of aryl substituents appeared with appropriate characteristic multiplicity in the region 7.15–8.06 ppm, and the protons of amino group gave rise to two singlets in the range 8.2–9.0 ppm with the difference of 0.5 ppm.

We failed to obtain the corresponding 2-halopyridines from alkyl derivatives **Ib–Id**. In 2-

Scheme 5.



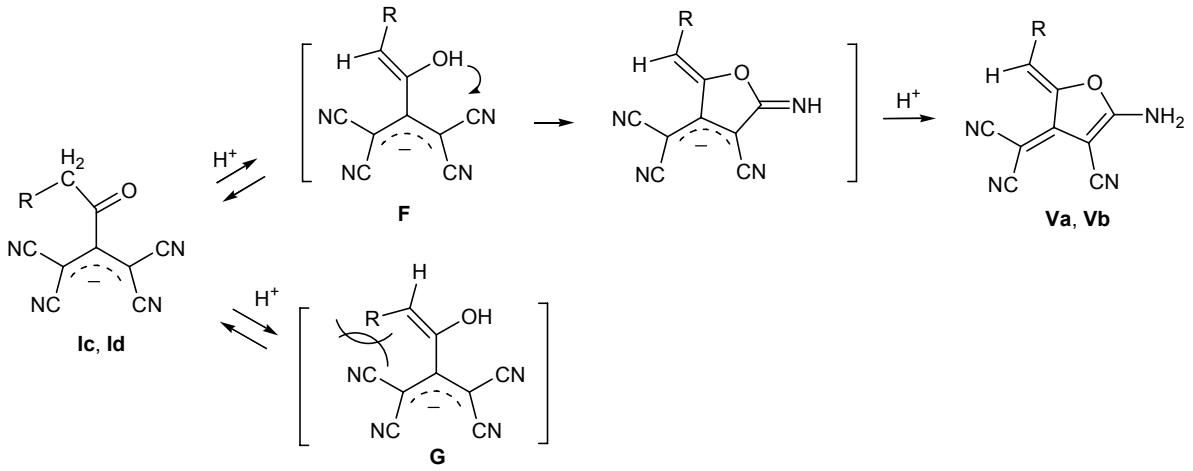
IVa-IVc

R = CH_3 (**a**), C_2H_5 (**b**), C_3H_7 (**c**).

butanol the main reaction products were 2-[2-alkyl-5-amino-2-(butan-2-yloxy)-4-cyano-2,3-dihydrofuran-3-ylidene]propanedinitriles **IVa–IVc** obtained as a diastereomers mixture (1 : 1) in an overall yield of 63–79% (Scheme 5).

The structure of compounds **IVa–IVc** was established from the data of ^{1}H NMR, IR, mass spectra and was in agreement with the data of elemental analysis. The amino group gave rise to a singlet in the region 10.14–10.26 ppm, the signals of alkyl substituents appeared in the common range and were

Scheme 6.



V, R = CH_3 (**a**), C_2H_5 (**b**).

doubled due to the presence of diastereomers. The assumed reaction path includes the protonation of the carbonyl group with the formation of a zwitter-ion intermediate **E**, the addition of 2-butanol to the carbocation site followed by the heterocyclization involving the hydroxy and cyano groups. The neutralization led to the formation of final compounds **IVa–IVc**. This result shows that the protonation of propenides **I** at the oxygen atom giving zwitter-ion **E** occurs also in the media with low dielectric permittivity confirming indirectly the key role in governing the reaction direction of the dissociation degree of the hydrogen halide in the solvent. The difference from aroyl propenides **I** is due apparently to the higher reactivity of the aliphatic carbonyl group.

In solvents of low dielectric permittivity lacking nucleophilic properties the reaction of alkyl derivatives of propenides **Ic** and **Id** with hydrogen halides resulted in 2-(2-alkylidene-5-amino-4-cyano-2,3-dihydrofuran-3-ylidene)propanedinitriles **Va** and **Vb** (Scheme 6).

The structure and composition of dihydrofurans **Va** and **Vb** were established from ^{1}H, ^{13}C NMR, IR, mass spectra and elemental analysis data. According to the ^{1}H NMR data a single diastereomer among the possible structures with respect to the double bond of the alkylidene fragment was obtained.

The presence of a hydrogen atom in the α-position with respect to the carbonyl group provides a possibility of a keto-enol tautomerism catalyzed by hydrogen halide. The formed enol **F** undergoes the heterocyclization which results after the neutralization in alkylidenedihydrofurans **Va** and **Vb**. Evidently this process is faster than the hydrogen halide addition to the cyano group, therefore in anhydrous organic solvents 2-propanoyl- and 2-butanoyl-1,1,3,3-tetracyanopropenides react with HCl and HBr to afford alkylidenedihydrofurans **Va** and **Vb** instead of 2-halopyridine derivatives. The stereoselectivity of the reaction is apparently due to the steric factor. *Z*-Isomer of enol **F** can undergo further cyclization, whereas in *E*-isomer **G** the interaction of the bulky substituent R and the cyano group impedes the formation of a planar conformation favorable for the cyclization. In keeping with these assumptions the *Z*-configuration should be assigned to compounds **Va** and **Vb**, and it is indirectly confirmed by the lack in the ^{13}C NMR (GATED) spectrum of compound **Va** of spin-spin coupling constant $^3J(C^3, H)$.

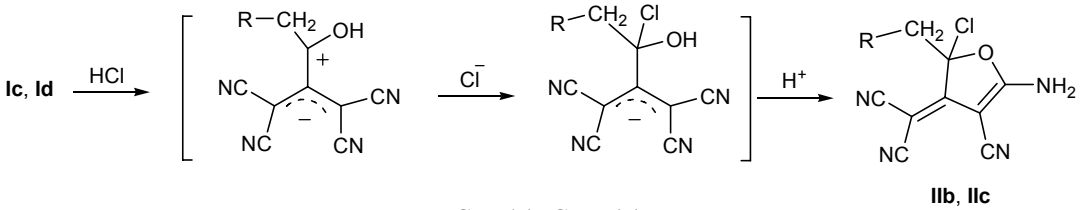
The reaction of alkylpropenides **Ic** and **Id** with HCl in water proceeds similarly to aroyl derivatives and leads to the formation of 2-(2-alkyl-5-amino-2-chloro-4-cyano-2,3-dihydrofuran-3-ylidene)propanedinitriles **IIb** and **IIc** (Scheme 7).

The structure of compounds **IIb** and **IIc** was established from the data of ^{1}H NMR, IR, mass spectra and was in agreement with the data of elemental analysis.

The effect of substituents was observed in reactions of propenides **I** with HI at boiling in water solution. From dimethoxybenzoyl derivatives **Ij** and **Ik** 2-amino-4-(3,4- and 2,5-dimethoxybenzyl)-6-iodo-5-cyanonicotinamides **VIa** and **VIb** were obtained (Scheme 8).

In this process two position isomers, **VI** and **J**, may form distinguished by the reciprocal location of the

Scheme 7.



R = CH_3 (**a**), C_2H_5 (**b**).

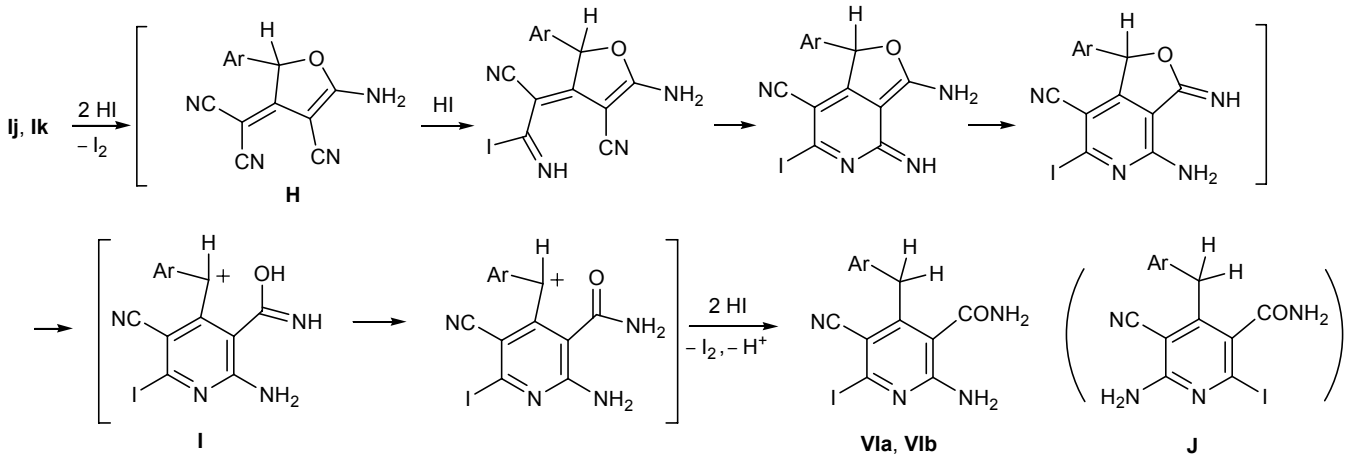
amino group and iodine atom with respect to the rest part of the molecule. TLC and ^{1}H NMR data show that the reaction results in a single isomer among the possible ones. Its structure was established by the analysis of the ^{13}C NMR spectrum of compound **VIb**. The signals of carbon atoms of the pyridine ring in the positions *3* and *5* were informative. In keeping with calculations carried out using tables compiling the effect of functional groups on the shift of signals in the ^{13}C NMR spectra of the pyridine ring [12] in the spectrum of compound **VIb** the signals positions of atoms C^3 and C^5 differ insignificantly and are located at 112 and 110.5 ppm respectively. The analogous calculation for the alternative structure **J** gives the values 129.5 for C^3 and 93 ppm for C^5. In the actual ^{13}C NMR spectrum the signals of atoms C^3 and C^5 are observed at 111.59 and 104.61 ppm in agreement with structure **VIb**.

Evidently the reaction of compounds **Ij** and **Ik** primarily proceeds through the formation of intermediate dihydrofurans **H** which are the main reaction products in event of the other aroyl substituents [9]. Further the addition of HI occurs and initiates the heterocyclization giving rise to the furo-[3,4-*c*]pyridine system. Under the acid catalysis the furan ring suffers opening with the formation of carbocation **I** that is stabilized with two donor substituents. The reaction is completed by the addition of the iodide ion and by the reduction. The regioselectivity of the iodide ion addition to the cyano group of the dicyanomethylene unit in the intermediate dihydrofuran **H** was discussed in [10].

This path is indirectly confirmed by the formation from propenides **Ie**, **Ig** and **Ii** at boiling with HI in water environment of 4-amino-8-aroyl(acyl)-6-iodo-3-oxo-2,3-dihydro-1*H*-pyrrolo[3,4-*c*]pyridine-7-carbonitriles **VIIa–VIIc** (Scheme 9). 4-Chlorobenzoyl- and pivaloyl derivatives **VIIa** and **VIIb** formed in low yields as side products, but 4-methoxybenzoyl derivative **VIIc** was the main reaction product.

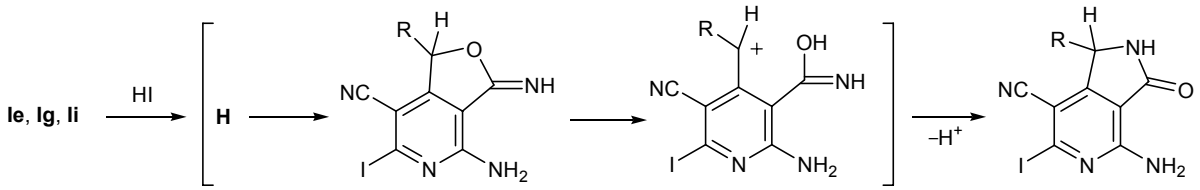
The structure of compounds **VIIa–VIIc** was suggested proceeding from the ^{1}H NMR spectra. Unlike the corresponding furo[3,4-*c*]pyridines (main reaction products) [10] in the ^{1}H NMR spectra of compounds **VIIa–VIIc** a proton signal from the NH group of a lactam ring appears at 8.86–9.13 ppm, and the signals belonging to the protons at C^1 and to amino

Scheme 8.



VI, Ar = 3,4-$(MeO)_2C_6H_3$ (**a**), 2,5-$(MeO)_2C_6H_3$ (**b**).

Scheme 9.



VII, R = 4-ClC_6H_4 (**a**), *tert*-C_4H_9 (**b**), 4-$MeOC_6H_4$ (**c**).

group are shifted upfield. The IR and mass spectra are consistent with the assumed structure.

A position isomerism is possible in compounds **VIIa–VIIc** characterized by the reciprocal location of the amino group and iodine atom with respect to the other part of the molecule. According to ^{1}H NMR spectrum a single isomer among the possible ones is obtained. By analogy with compounds **VIa** and **VIb** we presume that HI addition occurs at the cyano group of dicyanomethylidene fragment of **H** intermediates.

The presumable reaction path in the first stages is analogous to the reaction of propenides **Ij** and **Ik**. Carbocation intermediate **K** forming at the opening of the furan ring is less stabilized than **I** and possibly it is just the reason of the closure of a pyrrole ring and not the reduction of the carbocation site.

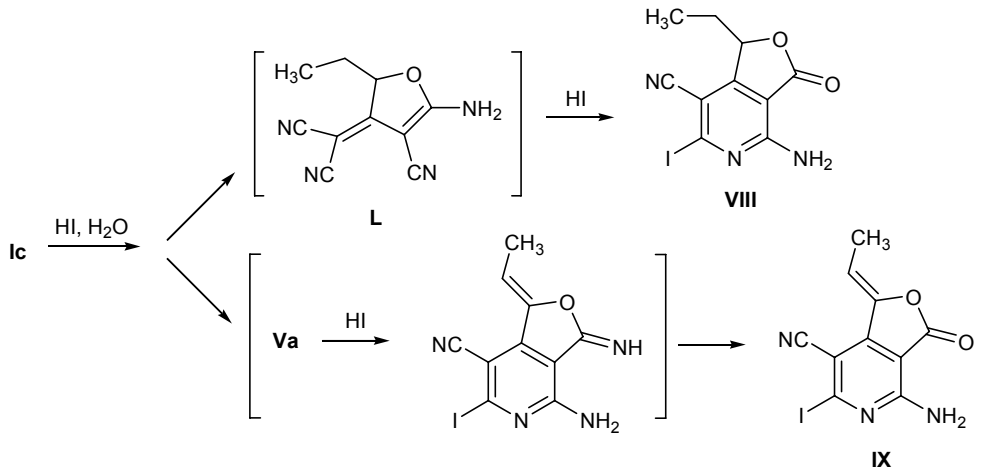
In the reaction of propenide **Ic** with HI in water beside 4-amino-6-iodo-3-oxo-1-ethyl-1,3-dihydrofuro-[3,4-*c*]pyridine-7-carbonitrile (**VIII**) (the main reaction product [10]) 4-amino-6-iodo-3-oxo-1-ethylidene-1,3-dihydrofuro[3,4-*c*]pyridine-7-carbonitrile (**IX**) was isolated (Scheme 10).

The structure of compound **IX** was established from the data of ^{1}H NMR and mass spectra. It is consistent with the data of IR spectroscopy and elemental analysis. The formation of compound **IX** may be ascribed to the proceeding in the first stage of competing reactions yielding alkylidenedihydrofuran **Va** and dihydrofuran **L** that further through HI addition and heterocyclization transform respectively into 1-ethylfuro[3,4-*c*]pyridine (**VIII**) and 1-ethylidenefuro[3,4-*c*]pyridine (**IX**) (Scheme 10).

EXPERIMENTAL

The reaction progress was monitored and the purity of compounds synthesized was checked by TLC on Silufol UV-254 plates (development under UV irradiation, in iodine vapor, or by thermal degradation). IR spectra were recorded on a Fourier spectrophotometer FSM-1202 from mulls in mineral oil. ^{1}H and ^{13}C NMR spectra were registered on a spectrometer Bruker DRX-500 (500.13 and 125.76 MHz respectively) in DMSO-d_6, internal reference TMS. Mass spectra were measured on an instrument

Scheme 10.



Simadzu GCMS-QP2010S DI (electron impact, 70 eV).

2-(5-Amino-2-phenyl-2-chloro-4-cyano-2,3-dihydrofuran-3-ylidene)propanedinitrile IIa and 2-amino-4-benzoyl-6-chloropyridine-3,5-dicarbonitrile IIIa. In 10 mL of an appropriate solvent was dissolved at room temperature 2.68 g (0.01 mol) of sodium 2-benzoyl-1,1,3,3-tetracyanopropenide **Ia**. Then dry HCl was bubbled through the solution till the completion of the process (TLC monitoring). The reaction mixture was poured into 30 mL of distilled water (at the use of 2-butanol as solvent the reaction mixture was poured into 100 mL of hot water), the separated precipitate was filtered off, washed with water till neutral washings, dried in air, and then maintained in a desiccator over conc. H_2SO_4 for 12 h.

2-(5-Amino-2-methyl-2-chloro-4-cyano-2,3-dihydrofuran-3-ylidene)propanedinitrile (IIb). To 1.69 g (0.005 mol) of propenide **Ib** was added 15 mL of conc. HCl (36–38%). The reaction mixture was heated at 70–80°C while stirring, then 50 mL of distilled water was added thereto. The separated precipitate was filtered off and recrystallized from 12 mL of 50% aqueous 2-propanol. Yield 67%, t.decomp. 216–218°C. IR spectrum, ν, cm^{-1}: 3290 (NH_2), 2223 (C≡N), 1682 (C=O). ^1H NMR spectrum, δ, ppm: 2.26 s (3H CH_3), 10.66 br.s (2H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 222 (3) 220 (10) $[M]^+$, 185 (4), 184 (77). Found, %: C 49.13; H 2.31; N 25.42. $C_9H_5ClN_4O$. Calculated, %: C 49.00; H 2.28; N 25.40. M 220.02.

2-(5-Amino-2-chloro-4-cyano-2-ethyl-2,3-dihydrofuran-3-ylidene)propanedinitrile (IIc) was similarly prepared. Yield 63%, mp 206–210°C (decomp.). IR spectrum, ν, cm^{-1}: 3285 (NH_2), 2223 (C≡N), 1686 (C=O). ^1H NMR spectrum, δ, ppm: 1.00 t (3H, CH_3, ^3J 7.3 Hz), 2.52 q (2H, CH_2, ^3J 7.3 Hz) 10.70 br.s (2H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 236 (3) 234 (12) $[M]^+$, 200 (11), 199 (86). Found, %: C 51.23; H 3.85; N 23.82. $C_{10}H_7ClN_4O$. Calculated, %: C 51.19; H 3.01; N 23.88. M 234.03.

2-Amino-4-(2,4-dichlorobenzoyl)-6-chloropyridine-3,5-dicarbonitrile (IIIb). In 10 mL of acetic acid was dissolved at heating 3.37 g (0.01 mol) of sodium 2-(2,4-dichlorobenzoyl)-1,1,3,3-tetracyanopropenide (**Ih**). Then dry HCl was bubbled through the solution at 50–60°C at a rate 0.5–1.0 mL s^{-1} till yellow color disappeared (2–3 min). Then the mixture was poured into 20 mL of water, the separated precipitate was filtered off and recrystallized from 2-propanol. Yield 2.27 g (65%), mp 246–248°C (decomp.). IR spectrum, ν, cm^{-1}: 3211 (NH_2), 2217 (C≡N), 1662 (C=O). ^1H NMR spectrum, δ, ppm: 7.67 d.d (1H_{arom}, ^3J 8.5, ^4J 2.0 Hz), 7.88 d (1H_{arom}, ^4J 2.0 Hz), 7.94 d (1H_{arom}, ^3J 8.5 Hz), 8.45 br.s (1H, NH_2), 9.00 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 351 (1), 353 (1), 354 (2), 352 (6), 350 (7) $[M]^+$, 175 (73), 174 (16), 173 (100) $[ArCO]^+$, 147 (28), 145 (42) $[Ar]^+$. Found, %: C 47.51; H 1.33; N 16.01. $C_{14}H_5Cl_3N_4O$. Calculated, %: C 47.83; H 1.43; N 15.94. M 349.95.

Compounds **IIIc–IIIe** were similarly obtained.

2-Amino-4-(4-nitrobenzoyl)-6-chloropyridine-3,5-dicarbonitrile (IIIc). Yield 63%, mp 264–267°C (decomp.). IR spectrum, ν, cm^{-1}: 3210 (NH_2), 2210 (C≡N), 1654 (C=O). ^1H NMR spectrum, δ, ppm: 8.33 d (2H_{arom}, ^3J 8.8 Hz), 8.43 d (2H_{arom}, ^3J 8.8 Hz), 8.55 br.s (1H, NH_2), 9.07 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 329 (3), 327 (8) $[M]^+$, 150 (68) $[ArCO]^+$, 120 (6), 116 (11), 104 (30), 92 (22). Found, %: C 51.39; H 1.83; N 21.39. $C_{14}H_6ClN_5O_3$. Calculated, %: C 51.31; H 1.85; N 21.37. M 327.02

2-Amino-4-(4-methoxybenzoyl)-6-chloropyridine-3,5-dicarbonitrile (IIId). Yield 71%, mp 272–274°C (decomp.). IR spectrum, ν, cm^{-1}: 3210 (NH_2), 2212 (C≡N), 1650 (C=O). ^1H NMR spectrum, δ, ppm: 3.92 s (3H, CH_3), 7.16 m (2H_{arom}, system AA′XX′, ^3J 8.8 Hz), 7.99 m (2H_{arom}, system AA′XX′, ^3J 8.8 Hz), 8.43 br.s (1H, NH_2), 8.93 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 315 (1), 314 (6), 313 (4), 312 (20) $[M]^+$, 136 (33), 135 (100) $[ArCO]^+$, 116 (15), 107 (34) $[Ar]^+$, 92 (67) $[Ar–15]^+$. Found, %: C 57.45; H 2.94; N 17.97. $C_{15}H_9ClN_4O_2$. Calculated, %: C 57.61; H 2.90; N 17.92. *M* 312.04.

2-Amino-4-(3,4-dimethoxybenzoyl)-6-chloropyridine-3,5-dicarbonitrile (IIIe). Yield 76%, mp 238–240°C. IR spectrum, ν, cm^{-1}: 3198 (NH_2), 2215 (C≡N), 1650 (C=O). ^1H NMR spectrum, δ, ppm: 3.87 s (3H, CH_3), 3.92 s (3H, CH_3), 7.11 d (1H_{arom}, ^3J 8.5 Hz), 7.52 d (1H_{arom}, ^4J 1.9 Hz), 7.58 d.d (1H_{arom}, ^3J 8.4, ^4J 1.9 Hz), 8.41 br.s (1H, NH_2), 8.94 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 345 (1), 344 (6), 343 (3), 342 (19) $[M]^+$, 166 (16), 165 (100) $[ArCO]^+$, 137 (10) $[Ar]^+$, 122 (9), 119 (55), 116 (9), 107 (10). Found, %: C 56.05; H 3.34; N 16.37. $C_{16}H_{11}ClN_4O_3$. Calculated, %: C 56.07; H 3.23; N 16.35. *M* 342.05.

2-Amino-6-bromo-4-(3-chlorobenzoyl)pyridine-3,5-dicarbonitrile (IIIf). In 10 mL of acetic acid was dissolved at heating 3.03 g (0.01 mol) of sodium 2-(3-

chlorobenzoyl)-1,1,3,3-tetracyanopropenide (**If**). Then dry HBr was bubbled through the solution at 50–60°C at a rate 0.5–1.0 mL s^{-1} till yellow color disappeared (2–3 min). Then the mixture was poured into 20 mL of water, the separated precipitate was filtered off and recrystallized from 6.5 mL of 1,4-dioxane. Yield 2.42 g (67%), mp 228–230°C (decomp.). IR spectrum, ν, cm^{-1}: 3204 (NH_2), 2210 (C≡N), 1650 (C=O). ^1H NMR spectrum, δ, ppm: 7.69 t ($1H_{arom}$ ^3J 7.9 Hz), 7.93 d ($1H_{arom}$ ^3J 8.0 Hz), 8.01 d ($1H_{arom}$ ^3J 7.9 Hz), 8.12 s ($1H_{arom}$), 8.48 br.s (1H, NH_2), 9.00 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 364 (2), 363 (1), 362 (6), 361 (1), 360 (5) $[M]^+$, 141 (62), 140 (20), 139 (100) $[ArCO]^+$, 113 (26), 111 (77) $[Ar]^+$. Found, %: C 46.61; H 1.63; N 15.41. $C_{14}H_6BrClN_4O$. Calculated, %: C 46.50; H 1.67; N 15.49. M 359.94.

Compounds **IIIg–IIIi** were synthesized analogously.

2-Amino-6-bromo-4-(2,4-dichlorobenzoyl)pyridine-3,5-dicarbonitrile (IIIg) Yield 63%, mp 238–240°C (decomp.). IR spectrum, ν, cm^{-1}: 3208 (NH_2), 2210 (C≡N), 1655 (C=O). ^1H NMR spectrum, δ, ppm: 7.67 d.d ($1H_{arom}$, ^3J 8.5, ^4J 1.9 Hz), 7.96 d ($1H_{arom}$, ^4J 1.9 Hz), 7.96 d ($1H_{arom}$, ^3J 8.4 Hz), 8.45 br.s (1H, NH_2), 8.99 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 398 (1), 396 (2), 394 (1) $[M]^+$, 177 (13), 176 (7), 175 (75), 174 (14), 173 (100) $[ArCO]^+$, 147 (23), 145 (36) $[Ar]^+$. Found, %: C 42.51; H 1.33; N 14.21. $C_{14}H_5BrCl_2N_4O$. Calculated, %: C 42.46; H 1.27; N 14.15. M 393.90.

2-Amino-6-bromo-4-(4-nitrobenzoyl)pyridine-3,5-dicarbonitrile (IIIh). Yield 58%, mp 262–264°C (decomp.). IR spectrum, ν, cm^{-1}: 3204 (NH_2), 2214 (C≡N), 1658 (C=O). ^1H NMR spectrum, δ, ppm: 8.33 d ($2H_{arom}$, ^3J 8.7 Hz), 8.42 d ($2H_{arom}$, ^3J 8.7 Hz), 8.53 br.s (1H, NH_2), 9.06 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 373 (3), 372 (1), 371 (3) $[M]^+$, 190 (6), 151 (17), 150 (100) $[ArCO]^+$, 120 (22), 116 (24), 104 (65). Found, %: C 45.21; H 1.69; N 18.09. $C_{14}H_6BrN_5O_3$. Calculated, %: C 45.19; H 1.63; N 18.82. M 370.97.

2-Amino-6-bromo-4-(4-methoxybenzoyl)pyridine-3,5-dicarbonitrile (IIIi). Yield 74%, mp 277–278°C (decomp.). IR spectrum, ν, cm^{-1}: 3218 (NH_2), 2210 (C≡N), 1652 (C=O). ^1H NMR spectrum, δ, ppm: 3.92 s (3H, CH_3), 7.15 d ($2H_{arom}$, ^3J 8.8 Hz), 7 .98 d ($2H_{arom}$, ^3J 8.8 Hz), 8.42 br.s (1H, NH_2), 8.93 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 359 (1), 358 (7), 356 (8) $[M]^+$, 135 (100) $[ArCO]^+$, 116 (13), 107 (25) $[Ar]^+$, 92 (45) $[ArCO - 15]^+$. Found, %: C 50.31; H 2.59; N 15.57. $C_{15}H_9BrN_4O_2$. Calculated, %: C 50.44; H 2.54; N 15.69. M 355.99.

2-[5-Amino-2-(butan-2-yloxy)-2-methyl-4-cyano-2,3-dihydrofuran-3-ylidene]propanedinitrile (diastereomers mixture, 1 : 1) (IVa). In 10 mL of 2-butanol was dissolved at heating 2.06 g (0.01 mol) of sodium 2-acetyl-1,1,3,3-tetracyanopropenide (**Ib**). Then dry HCl was bubbled through the solution at 50–60°C at a rate 0.5–1.0 mL s^{-1} till yellow color disappeared (2–3 min). Then the mixture was poured into 50 mL of hot water, the separated precipitate was filtered off and recrystallized from 2-propanol. Yield 1.88 g (73%), mp 186–189°C (decomp.), {mp 190°C (decomp.) [13]}. IR spectrum, ν, cm^{-1}: 3283, 3153 (NH_2), 2216 (C≡N), 1689 (C=C). ^1H NMR spectrum, δ, ppm: 0.83–0.87 m (3H, CH_3), 1.11 d and 1.17 d (3H, CH_3, ^3J 6.2 Hz), 1.41–1.61 m (2H, CH_2), 1.79 s and 1.80 s (3H, CH_3), 3.67–3.73 m (1H, CH), 10.14 s (2H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 259 (3), 258 (24) $[M]^+$, 203 (9), 202 (68), 186 (27), 185 (100), 57 (56). Found, %: C 60.33; H 5.51; N 21.53. $C_{13}H_{14}N_4O_2$. Calculated, %: C 60.45; H 5.46; N 21.69. M 258.11.

Compound **IVb** and **IVc** were obtained similarly.

2-[5-Amino-2-(butan-2-yloxy)-4-cyano-2-ethyl-2,3-dihydrofuran-3-ylidene]propanedinitrile (diastereomers mixture, 1 : 1) (IVb). Yield 68%, mp 167–169°C (decomp.). IR spectrum, ν, cm^{-1}: 3201 (NH_2), 2253, 2236 (C≡N), 1682 (C=O). ^1H NMR spectrum, δ, ppm: 0.83 t and 0.85 t [3H, $CH(CH_3)CH_2C\underline{H}_3$, ^3J 7.4 Hz], 0.91 t (3H, $CH_2C\underline{H}_3$, ^3J 7.3 Hz), 1.10 d and 1.17 d [3H, $CH(C\underline{H}_3)CH_2CH_3$, ^3J 6.2 Hz], 1.43–1.52 m [2H, $CH(CH_3)C\underline{H}_2CH_3$], 2.00–2.03 m and 2.07–2.09 m (2H, $C\underline{H}_2CH_3$), 3.65–3.68 m [1H, $C\underline{H}(CH_3)CH_2CH_3$], 10.22 s (2H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 272 (4) $[M]^+$, 215 (24), 199 (9), 57 (100). Found, %: C 61.73; H 5.96; N 20.53. $C_{14}H_{16}N_4O_2$. Calculated, %: C 61.75; H 5.92; N 20.58. M 272.13.

2-[5-Amino-2-(butan-2-yloxy)-2-propyl-4-cyano-2,3-dihydrofuran-3-ylidene]propanedinitrile (diastereomers mixture, 1 : 1) (IVc). Yield 79%, mp 149–151°C (decomp.). IR spectrum, ν, cm^{-1}: 3203 (NH_2), 2253, 2232 (C≡N), 1680 (C=O). ^1H NMR spectrum, δ, ppm: 0.84 t and 0.86 t [3H, $CH(CH_3)CH_2C\underline{H}_3$, ^3J 7.4 Hz], 0.90 t (3H, $CH_2CH_2C\underline{H}_3$, ^3J 7.3 Hz), 1.10 d and 1.17 d [3H, $CH(C\underline{H}_3)CH_2CH_3$, ^3J 6.2 Hz], 1.27–1.31 m (2H, $CH_2C\underline{H}_2CH_3$), 1.43–1.52 m [2H, $CH(CH_3)C\underline{H}_2CH_3$], 2.00–2.03 m and 2.07–2.09 m (2H, $C\underline{H}_2CH_2CH_3$), 3.65–3.68 m [1H, $C\underline{H}(CH_3)CH_2CH_3$],

10.15 s and 10.26 s (2H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 287 (1), 286 (6) $[M]^+$, 257 (2), 229 (27), 213 (25), 57 (45). Found, %: C 62.73; H 6.41; N 19.53. $C_{15}H_{18}N_4O_2$. Calculated, %: C 62.92; H 6.34; N 19.57. M 286.14.

(*Z*)-2-(5-Amino-4-cyano-2-ethylidene-2,3-dihydrofuran-3-ylidene)propanedinitrile (Va). In 10 mL of 1,4-dioxane was dissolved at heating 2.2 g (0.01 mol) of sodium 2-propionyl-1,1,3,3-tetracyanopropenide (**Ic**). Then dry HCl was bubbled through the solution at 50–60°C at a rate 0.5–1.0 mL s^{-1} till yellow color disappeared (5–8 min); the mixture was poured into 20 mL of water, the separated precipitate was filtered off and recrystallized from 2-propanol. Yield 1.72 g (87%), t.decomp. > 230°C. IR spectrum, ν, cm^{-1}: 3201 (NH_2), 2250, 2225 (C≡N), 1685 (C=O). ^{1}H NMR spectrum, δ, ppm: 1.98 d (3H, CH_3, ^3J 7.3 Hz), 6.63 q (1H, CH, ^3J 7.3 Hz), 10.21 br.s (1H, NH_2), 10.32 br.s (1H, NH_2). ^{13}C NMR spectrum (GATED), δ, ppm: 12.67 q (CH_3, ^1J 129.87 Hz), 49.14 s (C^4) 71.25 s [$\underline{C}(CN)_2$], 111.22 s (CN), 114.12 (CN), 116.32, 117.60 d.q (CH, ^1J 162.8, ^2J 7.5 Hz), 116.59 s (CN), 147.78 m (C^2), 153.29 s (C^3), 169.74 s (C^5). Mass spectrum, *m/z* (I_{rel}, %): 198 (31) $[M]^+$, 171 (3), 143 (17). Found, %: C 60.51; H 3.11; N 28.12. $C_{10}H_6N_4O$. Calculated, %: C 60.60; H 3.05; N 28.27. M 198.05.

(*Z*)-2-(5-Amino-2-propylidene-4-cyano-2,3-dihydrofuran-3-ylidene)propanedinitrile (Vb) was similarly prepared. Yield 79%, t.decomp. > 220°C. IR spectrum, ν, cm^{-1}: 3208 (NH_2), 2251, 2230 (C≡N), 1686 (C=O). ^{1}H NMR spectrum, δ, ppm: 1.07 t (3H, CH_3, ^3J 7.5 Hz), 2.37–2.51 quintet (2H, CH_2, ^3J 7.6 Hz), 6.57 t (1H, CH, ^3J 7.8 Hz), 10.28 br.s (2H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 213 (15) 212 (100) $[M]^+$, 211 (34) $[M - 1]^+$, 197 (17), 196 (13), 185 (36), 184 (29). Found, %: C 62.33; H 3.91; N 26.27. $C_{11}H_8N_4O$. Calculated, %: C 62.26; H 3.80; N 26.40. M 212.07.

2-Amino-4-(3,4-dimethoxybenzyl)-6-iodo-5-cyanopyridine-3-carboxamide (VIa). In 20 mL of conc. HI (54–56%) was carefully triturated at room temperature 3.28 g (0.01 mol) of sodium 2-(3,4-dimethoxybenzoyl)-1,1,3,3-tetracyanopropenide (**Ij**). The reaction mixture was heated to boiling and stirred at this temperature for 2–3 min, then it was diluted with 50 mL of water. The separated oily substance was triturated till it solidified, and the liquid was decanted. The solid residue was triturated in 20 mL of hot 2-propanol, filtered off, washed with ethyl acetate, and recrystallized from glacial acetic acid. Yield 3.19 g (73%), mp 210–212°C. IR spectrum, ν, cm^{-1}: 3390, 3117 (NH_2), 2222 (C≡N), 1682 (C=O). ^{1}H NMR spectrum, δ, ppm: 3.71 s (3H, CH_3), 3.72 s (3H, CH_3), 3.92 s (2H, CH_2), 6.74 d (1H_{arom}, ^3J 8.3 Hz), 6.85 d (1H_{arom}, ^3J 8.3 Hz), 6.93 s (1H, 1H_{arom}) 7.20 br.s (2H, NH_2), 7.79 s (1H, $CONH_2$), 8.11 s (1H, $CONH_2$). ^{13}C NMR spectrum, δ, ppm: 36.64, 55.25, 66.25, 104.61, 111.59, 112.99, 116.29, 119.33, 120.83, 123.66, 129.38, 147.44, 148.34, 150.06, 156.85, 166.59. Mass spectrum, *m/z* (I_{rel}, %): 439 (1), 438 (6) $[M]^+$, 263 (12), 136 (54), 127 (9). Found, %: C 43.98; H 3.53; N 12.88. $C_{16}H_{15}IN_4O_3$. Calculated, %: C 43.85; H 3.45; N 12.79. M 438.02.

2-Amino-4-(2,5-dimethoxybenzyl)-6-iodo-5-cyanopyridine-3-carboxamide (VIb) was similarly prepared. Yield 77%, mp 218–220°C. IR spectrum, ν, cm^{-1}: 3386, 3120 (NH_2), 2225 (C≡N), 1686 (C=O). ^{1}H NMR spectrum, δ, ppm: 3.63 s (3H, CH_3), 3.74 s (3H, CH_3), 3.93 s (2H, CH_2), 6.32 d (1H_{arom}, ^4J 3.0 Hz), 6.76 d.d (1H_{arom}, ^3J 8.9, ^4J 3.0 Hz), 6.88 d (1H, 1H_{arom}, ^3J 8.9 Hz), 7.21 br.s (2H, NH_2), 7.66 s (1H, $CONH_2$), 8.00 s (1H, $CONH_2$). Mass spectrum, *m/z* (I_{rel}, %): 439 (2), 438 (13) $[M]^+$, 263 (20), 136 (65). Found, %: C 43.79; H 3.48; N 12.73. $C_{16}H_{15}IN_4O_3$. Calculated, %: C 43.85; H 3.45; N 12.79. M 438.02.

4-Amino-6-iodo-3-oxo-1-(4-chlorophenyl)-2,3-dihydro-1*H*-pyrrolo[3,4-*c*]pyridine-7-carbonitrile (VIIa). In 15 mL of conc. HI was carefully triturated 1.52 g (5 mmol) of propenide **Ig**, the reaction mixture was heated at 100°C for 5 min, cooled, and diluted with 10 mL of cold water. The separated oily substance was decanted and triturated at heating with 10 mL of ethanol. The obtained slurry was filtered, the precipitate was washed with ethanol to remove iodine, then it was recrystallized from 25 mL of glacial acetic acid. Yield of **4-amino-1- (4-chlorophenyl)-6-iodo-3-oxo-1,3-dihydrofuro[3,4-*c*]-pyridine-7-carbonitrile** 1.33 g (65%), mp 244–246°C {mp 244–246°C (decomp.) [10]}. The filtrate was diluted with 10 mL of water, the separated precipitate was filtered off and recrystallized from a mixture acetic acid–2-propanol, 4 : 1. Yield of compound **VIIa** 0.14 g (7%), mp 258–262°C (decomp.). IR spectrum, ν, cm^{-1}: 3325 (NH), 3216 (NH_2), 2215 (C≡N), 1655 (C=O). ^{1}H NMR spectrum, δ, ppm: 5.85 s (1H, CH), 7.24 br.s (1H, NH_2), 7.29 d (2H_{arom}, ^3J 8.4 Hz), 7.45 d (2H_{arom}, ^3J 8.4 Hz), 8.46 br.s (1H, NH_2), 9.13 s (1H, NH). Mass spectrum, *m/z* (I_{rel}, %): 412 (3), 411 (1), 410 (10) $[M]^+$, 127 (65).

Found, %: C 40.91; H 2.01; N 13.61. $C_{14}H_8ClIN_4O$. Calculated, %: C 40.95; H 1.96; N 13.65. M 409.94.

4-Amino-6-iodo-3-oxo-1-tert-butyl-2,3-dihydro-1*H*-pyrrolo[3,4-*c*]pyridine-7-carbonitrile (VIIb) was similarly obtained. Yield 23%, mp 225–227°C. IR spectrum, ν, cm^{-1}: 3310 (NH), 3204 (NH_2), 2212 (C≡N), 1657 (C=O). ^{1}H NMR spectrum, δ, ppm: 0.94 s [9H, $C(CH_3)_3$], 4.47 s (1H, CH), 7.35 br.s (1H, NH_2), 8.43 br.s (1H, NH_2), 8.86 s (1H, NH). Mass spectrum, *m/z* (I_{rel}, %): 127 (45), 57 (100). Found, %: C 40.41; H 3.53; N 15.71. $C_{12}H_{13}IN_4O$. Calculated, %: C 40.47; H 3.68; N 15.73. *M* 356.01.

4-Amino-6-iodo-1-(4-methoxyphenyl)-3-oxo-2,3-dihydro-1*H*-pyrrolo[3,4-*c*]pyridine-7-carbonitrile (VIIc). To 1.49 g (5 mmol) of propenide **Ii** was added 15 mL of conc. HI, the reaction mixture was heated at 100°C and stirred for 5 min, cooled, and diluted with 10 mL of cold water. The separated oily substance was triturated with 10 mL of ethanol. The obtained slurry was filtered, the precipitate was washed with ethanol to remove iodine, then it was recrystallized from 25 mL of glacial acetic acid. Yield 1.16 g (57%), mp 255–257°C. IR spectrum, ν, cm^{-1}: 3320 (NH), 3218 (NH_2), 2215 (C≡N), 1650 (C=O). ^{1}H NMR spectrum, δ, ppm: 3.75 s (3H, CH_3O), 5.75 s (1H, CH), 6.92 d ($2H_{arom}$, ^3J 8.7 Hz), 7.16 d ($2H_{arom}$, ^3J 8.7 Hz), 7.25 br.s (1H, NH_2), 8.42 br.s (1H, NH_2), 9.07 s (1H, NH). Mass spectrum, *m/z* (I_{rel}, %): 407 (4), 406 (55) $[M]^+$, 391 (11), 363 (6), 127 (25). Found, %: C 44.35; H 2.70; N 13.77. $C_{15}H_{11}IN_4O_2$. Calculated, %: C 44.36; H 2.73; N 13.79. *M* 405.99.

4-Amino-6-iodo-3-oxo-1-ethylidene-1,3-dihydrofuro[3,4-*c*]pyridine-7-carbonitrile (IX). To 1.10 g (5 mmol) of propenide **Ic** was added 15 mL of conc. HI, the reaction mixture was heated at 100°C and stirred for 5 min, cooled, and diluted with 10 mL of cold water. The separated oily substance was triturated with 10 mL of ethanol. The obtained slurry was filtered, the precipitate was washed with ethanol to remove iodine, then it was recrystallized from 25 mL of glacial acetic acid. Yield of **4-amino-6-iodo-3-oxo-1-ethyl-1,3-dihydrofuro[3,4-*c*]pyridine-7-carbonitrile (VIII)** 1.04 g (63%), t.decomp. 238–240°C {t.decomp. 238–240°C (decomp.)[10]}. The filtrate was diluted with 5 mL of water, the separated precipitate of compound**я IX** was filtered off. Yield 0.29 g (24%), t.decomp. > 235°C. IR spectrum, ν, cm^{-1}: 3216 (NH_2), 2223 (C≡N), 1732 (C=O). ^{1}H NMR spectrum, δ, ppm: 2.01 d (3H, CH_3, ^3J 7.3 Hz), 6.33 q (1H, CH, ^3J 7.3 Hz), 7.67 br.s (1H, NH_2), 8.86 br.s (1H, NH_2). Mass spectrum, *m/z* (I_{rel}, %): 328 (5), 327 (79) $[M]^+$. Found, %: C 36.68; H 1.88; N 12.81. $C_{10}H_6IN_3O_2$. Calculated, %: C 36.72; H 1.85; N 12.85. *M* 326.95.

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