

# Synthesis of 2-Imino-4-vinyl-2,5-dihydrofuran-3-carboxamides and Some Their Chemical Transformations

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**Abstract**—2-Imino-2,5-dihydrofuran-3-carboxamides containing 2-phenyl- and 2-furylvinyl substituents in position 4 of the furan ring were synthesized by condensation of 2-imino-4-methyl-2,5-dihydrofuran-3-carboxamides with the corresponding aldehydes. Acid hydrolysis of 4-(2-arylvinyl)-2-imino-2,5-dihydrofuran-3-carboxamides gave 4-(2-arylvinyl)-2-oxo-2,5-dihydrofuran-3-carboxamides, and their condensation with malononitrile resulted in the formation of 2-dicyanomethylidene derivatives.

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Unsaturated  $\gamma$ -lactones are widespread in nature. They exhibit biological activity and are used in various fields such as medicine, agriculture, perfumery, etc. [1]. Biological activity of unsaturated  $\gamma$ -lactones originates from the presence of double carbon–carbon bond or aromatic ring [2, 3]. Their derivatives having conjugated double bond and carbonyl group are known as plant growth stimulators [4, 5].

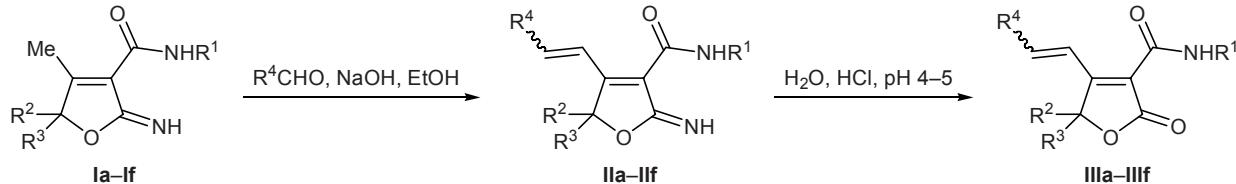
Taking into account the above stated, as well as the fact that activated methyl group in position 4 of 2,5-dihydrofuran-2-ones is capable of being involved in condensation processes [5–7], in the present work we synthesized 2-imino-2,5-dihydrofuran-3-carboxamides **IIa–IIIf** having 2-phenyl- or 2-furylvinyl substituent in position 4 by reaction of the corresponding 2-imino-4-methyl-2,5-dihydrofuran-3-carboxamides **Ia–If** with benzaldehyde or 2-furaldehyde (Scheme 1). Study on the effects of different factors on the reaction course allowed us to determine optimal conditions for the condensation. The best yields of compounds **IIa–IIIf** were obtained when a mixture of equimolar amounts of the reactants in anhydrous ethanol was heated for

4 h under reflux in the presence of a catalytic amount of sodium hydroxide. 4-Vinyl-substituted 2-imino-2,5-dihydrofuran-3-carboxamides **IIa–IIIf** readily underwent hydrolysis in acid medium (pH 4–5) at 85–90°C (reaction time 2 h) to give the corresponding 4-substituted 2-oxo-2,5-dihydrofuran-3-carboxamides **IIIa–IIIIf** (Scheme 1). This reaction may be regarded as a convenient method for the preparation of functionally substituted 2,5-dihydrofuran-2-ones.

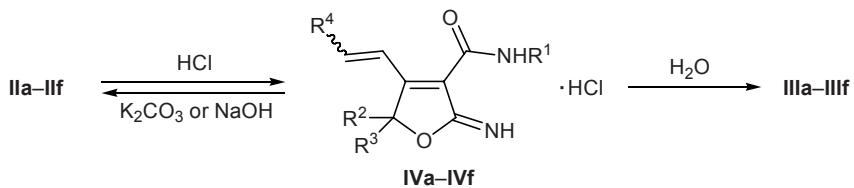
2-Imino-2,5-dihydrofurans **IIa–IIIf** were converted in quantitative yield to hydrochlorides **IVa–IVf** by passing gaseous hydrogen chloride through their solutions in benzene. Hydrochlorides **IVa–IVf** can be titrated with 0.1 N aqueous sodium hydroxide, and their treatment with a solution of potassium carbonate regenerates free bases **IIa–IIIf**. Acid hydrolysis of hydrochlorides **IVa–IVf** at 85–90°C (3 h) gives 2,5-dihydrofuran-2-ones **IIIa–IIIIf** (Scheme 2).

2-Imino derivatives **IIa–IIIf** were also brought into condensation with malononitrile. The reaction occurred on mixing equimolar amounts of the reactants at room temperature and was accompanied by evolution

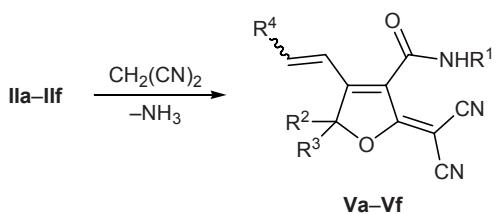
Scheme 1.



$R^1 = H$  (**a–d**), Me (**e, f**);  $R^2 = R^3 = Me$  (**a, c, e**);  $R^2R^3 = (CH_2)_5$  (**b, d, f**);  $R^4 = 2\text{-furyl}$  (**a, b**), Ph (**c–f**).

**Scheme 2.**

of ammonia. The products were the corresponding 2-dicyanomethylidene-2,5-dihydrofuran-3-carboxamides **Va–Vf** which were formed in quantitative yield (Scheme 3).

**Scheme 3.**

The structure of the newly synthesized compounds was confirmed by their IR and  $^1\text{H}$  NMR spectra and elemental analyses.

## EXPERIMENTAL

The IR spectra were recorded on a Specord 75IR spectrometer from samples dispersed in mineral oil. The  $^1\text{H}$  NMR spectra were measured on a Varian Mercury-300 instrument (300 MHz) from solutions in  $\text{DMSO}-d_6$ - $\text{CCl}_4$  (1:3) using tetramethylsilane as internal reference. The purity of the products was checked by TLC on Silufol UV-254 plates using acetone–benzene (1:2) as eluent; spots were visualized by treatment with iodine vapor or under UV light.

**5,5-Dialkyl-4-(2-arylvinyl)-2-imino-2,5-dihydrofuran-3-carboxamides IIa–IIf (general procedure).** Sodium hydroxide, 10 mg, was added to a solution of 5 mmol of compound **Ia–If** and 5 mmol of benzaldehyde or 2-furaldehyde in 20 ml of anhydrous ethanol. The mixture was heated for 4 h under reflux, the solvent was removed under reduced pressure, the residue was treated with water, and the precipitate was filtered off, washed with water, and recrystallized from aqueous ethanol.

**4-[2-(2-Furyl)vinyl]-2-imino-5,5-dimethyl-2,5-dihydrofuran-3-carboxamide (IIa).** Yield 82%, mp 191–192°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3300 ( $\text{NH}_2$ ), 3140 ( $\text{NH}$ ), 1680 ( $\text{C=O}$ ), 1640 ( $\text{C=N}$ ), 1625 ( $\text{C=C}$ ), 1620 ( $\text{C=C}$ ), 1588, 1540 ( $\text{C=C}$ , furan).  $^1\text{H}$  NMR spec-

trum,  $\delta$ , ppm: 1.65 s (6H,  $\text{CH}_3$ ), 6.49 d and 6.51 d (1H, 4'-H,  $J$  = 17.4 Hz), 6.72 d (1H, 3'-H,  $J$  = 17.4 Hz), 7.0 d (1H, =CH,  $J$  = 17.4 Hz), 7.21 br.s (1H,  $\text{NH}_2$ ), 7.34 s (1H, =NH), 7.65 d (1H, 5'-H,  $J$  = 17.4 Hz), 8.25 d (1H, =CH,  $J$  = 17.4 Hz), 9.08 br.s (1H,  $\text{NH}_2$ ). Found, %: C 63.49; H 5.84; N 11.44.  $\text{C}_{13}\text{H}_{14}\text{N}_2\text{O}_3$ . Calculated, %: C 63.40; H 5.73; N 11.38.

**4-[2-(2-Furyl)vinyl]-2-imino-5,5-pentamethylene-2,5-dihydrofuran-3-carboxamide (IIb).** Yield 73%, mp 182–183°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3412 ( $\text{NH}_2$ ), 3263 ( $\text{NH}$ ), 1671 ( $\text{C=O}$ ), 1645 ( $\text{C=N}$ ), 1630 ( $\text{C=C}$ ), 1625 ( $\text{C=C}$ ), 1588, 1540 ( $\text{C=C}$ , furan).  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.58–1.88 m (10H,  $\text{CH}_2$ ), 6.49 d and 6.51 d (1H, 4'-H,  $J$  = 17.4 Hz), 6.72 d (1H, 3'-H,  $J$  = 17.4 Hz), 7.0 d (1H, =CH,  $J$  = 17.4 Hz), 7.21 br.s (1H,  $\text{NH}_2$ ), 7.34 s (1H, =NH), 7.65 d (1H, 5'-H,  $J$  = 17.4 Hz), 8.25 d (1H, =CH,  $J$  = 17.4 Hz), 9.08 br.s (1H,  $\text{NH}_2$ ). Found, %: C 67.24; H 6.38; N 9.84.  $\text{C}_{16}\text{H}_{18}\text{N}_2\text{O}_3$ . Calculated, %: C 67.12; H 6.34; N 9.78.

**2-Imino-5,5-dimethyl-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (IIc).** Yield 93%, mp 193–194°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3300 ( $\text{NH}_2$ ), 3140 ( $\text{NH}$ ), 1680 ( $\text{C=O}$ ), 1640 ( $\text{C=N}$ ), 1630 ( $\text{C=C}$ ), 1620 ( $\text{C=C}$ ), 1600–1500 ( $\text{C=C}_{\text{arom}}$ ).  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.70 s (6H,  $\text{CH}_3$ ), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.44 m (5H,  $\text{H}_{\text{arom}}$ , =NH,  $\text{NH}_2$ ), 7.62 m (2H,  $\text{H}_{\text{arom}}$ ), 8.53 d (1H, =CH,  $J$  = 17.4 Hz), 9.16 br.s (1H,  $\text{NH}_2$ ). Found, %: C 70.34; H 6.44; N 11.04.  $\text{C}_{15}\text{H}_{15}\text{N}_2\text{O}_2$ . Calculated, %: C 70.29; H 6.29; N 10.93.

**2-Imino-5,5-pentamethylene-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (IId).** Yield 85%, mp 155–157°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3300 ( $\text{NH}_2$ ), 3140 ( $\text{NH}$ ), 1680 ( $\text{C=O}$ ), 1640 ( $\text{C=N}$ ), 1630 ( $\text{C=C}$ ), 1620 ( $\text{C=C}$ ), 1600–1500 ( $\text{C=C}_{\text{arom}}$ ).  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.6–1.9 m (10H,  $\text{CH}_2$ ), 7.2 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.44 m (5H,  $\text{H}_{\text{arom}}$ , =NH,  $\text{NH}_2$ ), 7.64 m (2H,  $\text{H}_{\text{arom}}$ ), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.16 br.s (1H,  $\text{NH}_2$ ). Found, %: C 72.99; H 6.89; N 9.54.  $\text{C}_{18}\text{H}_{20}\text{N}_2\text{O}_2$ . Calculated, %: C 72.95; H 6.80; N 9.45.

**2-Imino-N,5,5-trimethyl-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (IIe).** Yield 88%,

mp 128–130°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3340 (NH), 3160 (NH), 1680 (C=O), 1640 (C=N), 1635 (C=C), 1620 (C=C), 1600–1500 (C=C<sub>arom</sub>). <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.48 s (6H, CH<sub>3</sub>), 2.82 d (3H, NCH<sub>3</sub>,  $J$  = 4.9 Hz), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.4 m (4H, H<sub>arom</sub>, =NH), 7.61 m (2H, H<sub>arom</sub>), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.84 br.s (1H, NH). Found, %: C 71.26; H 6.81; N 10.44. C<sub>16</sub>H<sub>18</sub>N<sub>2</sub>O<sub>2</sub>. Calculated, %: C 71.09; H 6.71; N 10.36.

**2-Imino-N-methyl-5,5-pentamethylene-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (IIIe).** Yield 83%, mp 145–146°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 3335 (NH), 3150 (NH), 1680 (C=O), 1640 (C=N), 1635 (C=C), 1620 (C=C), 1600–1500 (C=C<sub>arom</sub>). <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.6–1.9 m (10H, CH<sub>2</sub>), 2.82 d (3H, NCH<sub>3</sub>,  $J$  = 4.9 Hz), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.40 m (4H, H<sub>arom</sub>, =NH), 7.61 m (2H, H<sub>arom</sub>), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.84 br.s (1H, NH). Found, %: C 73.59; H 7.28; N 9.27. C<sub>19</sub>H<sub>22</sub>N<sub>2</sub>O<sub>2</sub>. Calculated, %: C 73.52; H 7.14; N 9.03.

**5,5-Dialkyl-4-(2-arylvinyl)-2-oxo-2,5-dihydrofuran-3-carboxamides IIIa–IIIe (general procedure).** A mixture of 1 mmol of 2-imino-2,5-dihydrofuran IIa–IIe and 5 ml of water acidified with hydrochloric acid to pH 4–5 was heated for 3 h at 85–90°C. The mixture was cooled and extracted with diethyl ether (3×5 ml), the extract was dried over magnesium sulfate and evaporated, and the residue was recrystallized from petroleum ether.

**4-[2-(2-Furyl)vinyl]-5,5-dimethyl-2-oxo-2,5-dihydrofuran-3-carboxamide (IIIa).** Yield 80%, mp >270°C. <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.65 s (6H, CH<sub>3</sub>), 6.49 d and 6.51 d (1H, 4'-H,  $J$  = 17.4 Hz), 6.72 d (1H, 3'-H,  $J$  = 17.4 Hz), 7.0 d (1H, =CH,  $J$  = 17.4 Hz), 7.21 br.s (1H, NH<sub>2</sub>), 7.65 d (1H, 5'-H,  $J$  = 17.4 Hz), 8.25 d (1H, =CH,  $J$  = 17.4 Hz), 9.08 br.s (1H, NH<sub>2</sub>). Found, %: C 63.25; H 5.38; N 5.67. C<sub>13</sub>H<sub>13</sub>NO<sub>4</sub>. Calculated, %: C 63.15; H 5.30; N 5.66.

**4-[2-(2-Furyl)vinyl]-2-oxo-5,5-pentamethylene-2,5-dihydrofuran-3-carboxamide (IIIb).** Yield 78%, mp 160–162°C. <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.58–1.88 m (10H, CH<sub>2</sub>), 6.49 d and 6.51 d (1H, 4'-H,  $J$  = 17.4 Hz), 6.72 d (1H, 3'-H,  $J$  = 17.4 Hz), 7.0 d (1H, =CH,  $J$  = 17.4 Hz), 7.21 br.s (1H, NH<sub>2</sub>), 7.65 d (1H, 5'-H,  $J$  = 17.4 Hz), 8.25 d (1H, =CH,  $J$  = 17.4 Hz), 9.08 br.s (1H, NH<sub>2</sub>). Found, %: C 66.95; H 5.98; N 4.51. C<sub>16</sub>H<sub>17</sub>NO<sub>4</sub>. Calculated, %: C 66.89; H 5.96; N 4.88.

**5,5-Dimethyl-2-oxo-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (IIIc).** Yield 82%, mp 151–

152°C. <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.70 s (6H, CH<sub>3</sub>), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.44 m (5H, H<sub>arom</sub>, NH<sub>2</sub>), 7.62 m (2H, H<sub>arom</sub>), 8.53 d (1H, =CH,  $J$  = 17.4 Hz), 9.16 br.s (1H, NH<sub>2</sub>). Found, %: C 70.05; H 5.89; N 5.50. C<sub>15</sub>H<sub>15</sub>NO<sub>3</sub>. Calculated, %: C 70.02; H 5.88; N 5.44.

**2-Oxo-5,5-pentamethylene-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (IIId).** Yield 79%, mp 218–220°C. <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.6–1.9 m (10H, CH<sub>2</sub>), 7.2 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.44 m (5H, H<sub>arom</sub>, NH<sub>2</sub>), 7.64 m (2H, H<sub>arom</sub>), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.16 br.s (1H, NH<sub>2</sub>). Found, %: C 72.75; H 6.49; N 4.75. C<sub>18</sub>H<sub>19</sub>NO<sub>3</sub>. Calculated, %: C 72.71; H 6.44; N 4.71.

**N,5,5-Trimethyl-2-oxo-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (IIIf).** Yield 81%, mp 132–133°C. <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.48 s (6H, CH<sub>3</sub>), 2.82 d (3H, NCH<sub>3</sub>,  $J$  = 4.9 Hz), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.40 m (5H, H<sub>arom</sub>, NH<sub>2</sub>), 7.61 m (2H, H<sub>arom</sub>), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.84 br.s (1H, NH). Found, %: C 70.85; H 6.39; N 5.19. C<sub>16</sub>H<sub>17</sub>NO<sub>3</sub>. Calculated, %: C 70.83; H 6.32; N 5.16.

**N-Methyl-2-oxo-5,5-pentamethylene-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (IIIg).** Yield 80%, mp 126–130°C. <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.6–1.9 m (10H, CH<sub>2</sub>), 2.82 d (3H, NCH<sub>3</sub>,  $J$  = 4.9 Hz), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.4 m (5H, H<sub>arom</sub>, NH<sub>2</sub>), 7.61 m (2H, H<sub>arom</sub>), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.84 br.s (1H, NH). Found, %: C 73.34; H 6.89; N 4.52. C<sub>19</sub>H<sub>21</sub>NO<sub>3</sub>. Calculated, %: C 73.29; H 6.80; N 4.50.

**5,5-Dialkyl-4-(2-arylvinyl)-2-imino-2,5-dihydrofuran-3-carboxamide hydrochlorides IVa–IVf (general procedure).** Compound IIa–IIe, 2 mmol, was dissolved in benzene, gaseous hydrogen chloride was passed through the solution, and the precipitate was filtered off and washed with diethyl ether.

**4-[2-(2-Furyl)vinyl]-2-imino-5,5-dimethyl-2,5-dihydrofuran-3-carboxamide hydrochloride (IVa).** Yield 96%, mp 178–184°C. <sup>1</sup>H NMR spectrum,  $\delta$ , ppm: 1.65 s (6H, CH<sub>3</sub>), 6.49 d and 6.51 d (1H, 4'-H,  $J$  = 17.4 Hz), 6.72 d (1H, 3'-H,  $J$  = 17.4 Hz), 7.0 d (1H, =CH,  $J$  = 17.4 Hz), 7.21 br.s (1H, NH<sub>2</sub>), 7.65 d (1H, 5'-H,  $J$  = 17.4 Hz), 8.25 d (1H, =CH,  $J$  = 17.4 Hz), 9.08 br.s (1H, NH<sub>2</sub>), 10.34 br.s (2H, =NH·HCl). Found, %: C 55.57; H 5.59; N 10.04. C<sub>13</sub>H<sub>14</sub>N<sub>2</sub>O<sub>3</sub>·HCl. Calculated, %: C 55.23; H 5.35; N 9.91.

**4-[2-(2-Furyl)vinyl]-2-imino-5,5-pentamethylene-2,5-dihydrofuran-3-carboxamide hydrochloride**

**(IVb).** Yield 96%, mp 173–179°C.  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.58–1.88 m (10H,  $\text{CH}_2$ ), 6.49 d and 6.51 d (1H, 4'-H,  $J$  = 17.4 Hz), 6.72 d (1H, 3'-H,  $J$  = 17.4 Hz), 7.0 d (1H, =CH,  $J$  = 17.4 Hz), 7.21 br.s (1H,  $\text{NH}_2$ ), 7.65 d (1H, 5'-H,  $J$  = 17.4 Hz), 8.25 d (1H, =CH,  $J$  = 17.4 Hz), 9.08 br.s (1H,  $\text{NH}_2$ ), 10.2 br.s (2H, =NH·HCl). Found, %: C 59.72; H 6.09; N 8.99.  $\text{C}_{16}\text{H}_{18}\text{N}_2\text{O}_3\cdot\text{HCl}$ . Calculated, %: C 59.54; H 5.93; N 8.68.

**2-Imino-5,5-dimethyl-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide hydrochloride (IVc).** Yield 98%, mp 178–186°C.  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.70 s (6H,  $\text{CH}_3$ ), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.4 m (5H,  $\text{H}_{\text{arom}}$ ,  $\text{NH}_2$ ), 7.62 m (2H,  $\text{H}_{\text{arom}}$ ), 8.53 d (1H, =CH,  $J$  = 17.4 Hz), 9.16 br.s (1H,  $\text{NH}_2$ ), 10.2 br.s (2H, =NH·HCl). Found, %: C 61.99; H 5.89; N 9.64.  $\text{C}_{15}\text{H}_{16}\text{N}_2\text{O}_2\cdot\text{HCl}$ . Calculated, %: C 61.75; H 5.53; N 9.60.

**2-Imino-5,5-pentamethylene-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide hydrochloride (IVd).** Yield 98%, mp 184–188°C.  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.6–1.9 m (10H,  $\text{CH}_2$ ), 7.2 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.44 m (5H,  $\text{H}_{\text{arom}}$ ,  $\text{NH}_2$ ), 7.64 m (2H,  $\text{H}_{\text{arom}}$ ), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.16 br.s (1H,  $\text{NH}_2$ ), 10.25 br.s (2H, =NH·HCl). Found, %: C 65.12; H 6.56; N 8.64.  $\text{C}_{18}\text{H}_{20}\text{N}_2\text{O}_2\cdot\text{HCl}$ . Calculated, %: C 64.96; H 6.36; N 8.42.

**2-Imino-N,5,5-trimethyl-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide hydrochloride (IVe).** Yield 97%, mp 164–169°C.  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.46 s (6H,  $\text{CH}_3$ ), 2.82 d (3H,  $\text{NCH}_3$ ,  $J$  = 4.9 Hz), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.4 m (5H,  $\text{H}_{\text{arom}}$ ,  $\text{NH}_2$ ), 7.61 m (2H,  $\text{H}_{\text{arom}}$ ), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.84 br.s (1H, NH), 10.4 br.s (2H, =NH·HCl). Found, %: C 62.89; H 6.39; N 9.24.  $\text{C}_{16}\text{H}_{18}\text{N}_2\text{O}_2\cdot\text{HCl}$ . Calculated, %: C 62.64; H 6.24; N 9.13.

**2-Imino-N-methyl-5,5-pentamethylene-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide hydrochloride (IVf).** Yield 94%, mp 174–179°C.  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.6–1.88 m (10H,  $\text{CH}_2$ ), 2.82 d (3H,  $\text{NCH}_3$ ,  $J$  = 4.9 Hz), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.4 m (5H,  $\text{H}_{\text{arom}}$ ,  $\text{NH}_2$ ), 7.61 m (2H,  $\text{H}_{\text{arom}}$ ), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.84 br.s (1H, NH), 10.55 br.s (2H, =NH·HCl). Found, %: C 65.99; H 6.99; N 8.16.  $\text{C}_{19}\text{H}_{22}\text{N}_2\text{O}_2\cdot\text{HCl}$ . Calculated, %: C 65.79; H 6.68; N 8.08.

**Reaction of compounds IVa–IVf with potassium carbonate (general procedure).** A concentrated aqueous solution of potassium carbonate was added to a solution of 1 mmol of hydrochloride IVa–IVf in water

until pH 7–8. The precipitate was filtered off and washed with water. We thus isolated iminolactones IIa–IIf which showed no depression of the melting point on mixing with samples prepared as described above by condensation of compounds Ia–If with the corresponding aldehydes.

**Hydrolysis of compounds IVa–IVf (general procedure).** A mixture of 0.5 mmol of hydrochloride IVa–IVf and 5 ml of water was heated for 2 h at 85–90°C. The mixture was cooled and extracted with diethyl ether ( $3 \times 5$  ml), the extract was dried over magnesium sulfate, the solvent was distilled off, and the residue was recrystallized from petroleum ether. The products were identical to compounds IIIa–IIIf in melting points.

**5,5-Dialkyl-4-(2-arylvinyl)-2-dicyanomethylidene-2,5-dihydrofuran-3-carboxamides Va–Vf (general procedure).** A mixture of 2.5 mmol of compound IIa–IIf, 0.17 g (2.5 mmol) of malononitrile and 5 ml of anhydrous ethanol was stirred at room temperature until ammonia no longer evolved. The solvent was distilled off, the residue was treated with water, and the precipitate was filtered off, washed with water, and recrystallized from ethanol–water (2:1).

**2-Dicyanomethylidene-4-[2-(2-furyl)vinyl]-5,5-dimethyl-2,5-dihydrofuran-3-carboxamide (Va).** Yield 96%, mp >290°C.  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.65 s (6H,  $\text{CH}_3$ ), 6.49 d and 6.51 d (1H, 4'-H,  $J$  = 17.4 Hz), 6.72 d (1H, 3'-H,  $J$  = 17.4 Hz), 7.0 d (1H, =CH,  $J$  = 17.4 Hz), 7.21 br.s (1H,  $\text{NH}_2$ ), 7.65 d (1H, 5'-H,  $J$  = 17.4 Hz), 8.25 d (1H, =CH,  $J$  = 17.4 Hz), 9.08 br.s (1H,  $\text{NH}_2$ ). Found, %: C 65.09; H 4.49; N 14.26.  $\text{C}_{16}\text{H}_{13}\text{N}_3\text{O}_3$ . Calculated, %: C 65.08; H 4.44; N 14.23.

**2-Dicyanomethylidene-4-[2-(2-furyl)vinyl]-5,5-pentamethylene-2,5-dihydrofuran-3-carboxamide (Vb).** Yield 88%, mp 273–275°C.  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.58–1.88 m (10H,  $\text{CH}_2$ ), 6.49 d and 6.51 d (1H, 4'-H,  $J$  = 17.4 Hz), 6.72 d (1H, 3'-H,  $J$  = 17.4 Hz), 7.0 d (1H, =CH,  $J$  = 17.4 Hz), 7.21 br.s (1H,  $\text{NH}_2$ ), 7.65 d (1H, 5'-H,  $J$  = 17.4 Hz), 8.25 d (1H, =CH,  $J$  = 17.4 Hz), 9.08 br.s (1H,  $\text{NH}_2$ ). Found, %: C 68.05; H 5.17; N 12.56.  $\text{C}_{19}\text{H}_{17}\text{N}_3\text{O}_3$ . Calculated, %: C 68.05; H 5.11; N 12.53.

**2-Dicyanomethylidene-5,5-dimethyl-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (Vc).** Yield 95%, mp 240–242°C.  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 1.70 s (6H,  $\text{CH}_3$ ), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.44 m (4H,  $\text{H}_{\text{arom}}$ ,  $\text{NH}_2$ ), 7.62 m (2H,  $\text{H}_{\text{arom}}$ ), 8.53 d (1H, =CH,  $J$  = 17.4 Hz), 9.16 br.s (1H,  $\text{NH}_2$ ).

Found, %: C 70.89; H 4.99; N 13.84.  $C_{18}H_{15}N_3O_2$ . Calculated, %: C 70.81; H 4.95; N 13.76.

**2-Dicyanomethylidene-5,5-pentamethylene-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (Vd).** Yield 90%, mp 269–270°C.  $^1H$  NMR spectrum,  $\delta$ , ppm: 1.6–1.9 m (10H,  $CH_2$ ), 7.2 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.44 m (4H,  $H_{\text{arom}}$ ,  $NH_2$ ), 7.64 m (2H,  $H_{\text{arom}}$ ), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.16 br.s (1H,  $NH_2$ ). Found, %: C 73.14; H 5.59; N 12.21.  $C_{21}H_{19}N_3O_2$ . Calculated, %: C 73.03; H 5.54; N 12.17.

**2-Dicyanomethylidene-N,5,5-trimethyl-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (Ve).** Yield 84%, mp 184–185°C.  $^1H$  NMR spectrum,  $\delta$ , ppm: 1.48 s (6H,  $CH_3$ ), 2.82 d (3H,  $NCH_3$ ,  $J$  = 4.9 Hz), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.4 m (3H,  $H_{\text{arom}}$ ), 7.61 m (2H,  $H_{\text{arom}}$ ), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.84 br.s (1H, NH). Found, %: C 71.54; H 5.42; N 13.21.  $C_{19}H_{17}N_3O_2$ . Calculated, %: C 71.46; H 5.37; N 13.16.

**2-Dicyanomethylidene-N-methyl-5,5-pentamethylene-4-(2-phenylvinyl)-2,5-dihydrofuran-3-carboxamide (Vf).** Yield 84%, mp 154–155°C.  $^1H$  NMR spectrum,  $\delta$ , ppm: 1.6–1.9 m (10H,  $CH_2$ ), 2.82 d (3H,

$NCH_3$ ,  $J$  = 4.9 Hz), 7.13 d (1H, =CH,  $J$  = 17.4 Hz), 7.25–7.4 m (3H,  $H_{\text{arom}}$ ), 7.61 m (2H,  $H_{\text{arom}}$ ), 8.5 d (1H, =CH,  $J$  = 17.4 Hz), 9.84 br.s (1H, NH). Found, %: C 73.54; H 5.92; N 11.74.  $C_{22}H_{21}N_3O_2$ . Calculated, %: C 73.52; H 5.89; N 11.69.

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