

## Reactions of New *N*-(2,2-Dichloro-1-cyanoethenyl)amides with Aliphatic Amines

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**Abstract**—The reaction of the newly synthesized *N*-(2,2-dichloro-1-cyanoethenyl)prop-2-enamide and 4-chloro-*N*-(2,2-dichloro-1-cyanoethenyl)butanamide with methylamine or dimethylamine gave rise to previously unknown 5-amino-1,3-oxazole-4-carbonitriles. In the case of the reaction of *N*-(2,2-dichloro-1-cyanoethenyl)amides with ethylenediamine diacetate, new (*Z*)-2,3,5,6,7,8-hexahydro-7-oxo-1*H*-imidazo[1,2-*a*][1,4]diazepine-9-carbonitrile and 4-chloro-*N*-(cyano(imidazolidin-2-ylidene)methyl)butanamide were obtained.

**Keywords:** cyclization, 5-alkylamino-2-aminoalkyl-1,3-oxazole-4-carbonitrile, (*Z*)-2,3,5,6,7,8-hexahydro-7-oxo-1*H*-imidazo[1,2-*a*][1,4]diazepine-9-carbonitrile, 2-amino-3,3-dichloroacrylonitrile acyl derivatives

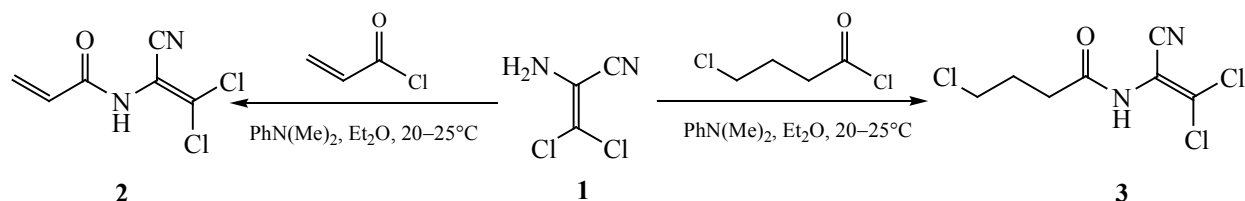
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Previously, we have shown that substituted 2-acylamino-3,3-dichloroacrylonitriles can be used to produce various heterocycles such as thiophene [1], hydantoin [2], and indole derivatives [3]. However, most reactions of 2-acylamino-3,3-dichloroacrylonitriles with amines provide oxazole derivatives [4–6]. In this case, depending on the nature of the acyl residue, the resulting reaction products can enter into further transformations with the retention of the oxazole ring [6–8], or with its destruction and the formation of recyclization products [9].

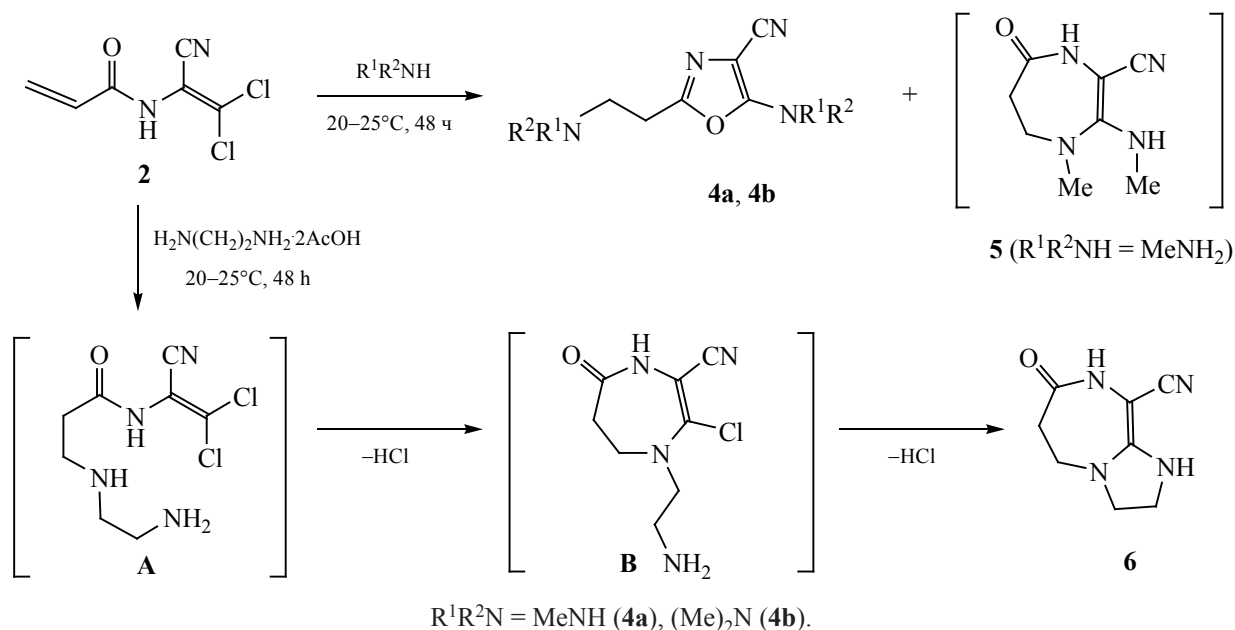
Herein, we reported the synthesis of previously unknown *N*-(2,2-dichloro-1-cyanoethenyl)prop-2-enamide **2** and *N*-(2,2-dichloro-1-cyanoethenyl)-4-chlorobutanamide **3** (Scheme 1) starting from available

2-amino-3,3-dichloroacrylonitrile **1** [10] and studied their reactions with some aliphatic amines (methylamine, dimethylamine, and ethylenediamine). Thus, reaction of *N*-(2,2-dichloro-1-cyanoethenyl)prop-2-enamide **2** with an excess of methylamine or dimethylamine proceeds through oxazole cyclization with the simultaneous addition of the amine residue to the double bond. As a result, 5-alkylamino-2-aminoethyl-1,3-oxazole-4-carbonitriles **4a** and **4b** were prepared (Scheme 2). It should be noted that oxazole **4a** was isolated in a yield of only 20%. According to the data of liquid chromatography-mass spectra, compound **5** is probably also present in the reaction mixture; however, we failed to isolate it in an individual state.

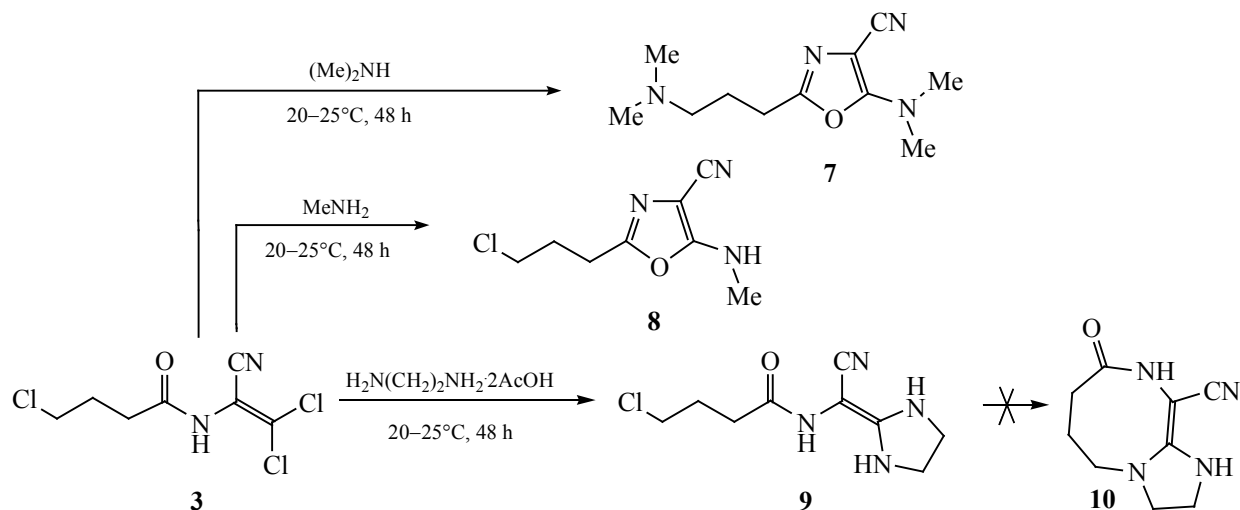
Scheme 1.



Scheme 2.



Scheme 3.

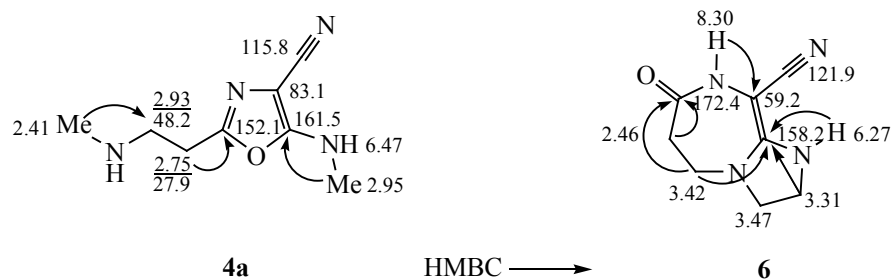


The reaction of dichloroacrylonitrile **2** with ethylenediamine diacetate resulted in the formation of (*Z*)-2,3,5,6,7,8-hexahydro-7-oxo-1*H*-imidazo[1,2-*a*][1,4]diazepine-9-carbonitrile **6**. A plausible mechanism for the formation of compound **6** includes the formation of intermediates **A** and **B** (Scheme 2).

The reaction of *N*-(2,2-dichloro-1-cyanoethenyl)-4-chlorobutylamide **3** with dimethylamine proceeded

similarly to compound **2**. In this case, 5-(dimethylamino)-2-[3-(dimethylamino)propyl]-1,3-oxazole-4-carbonitrile **7** was formed in high yield (Scheme 3). Substituted 5-amino-1,3-oxazole-4-carbonitrile **8** containing a chloropropyl residue in the second position of the ring was isolated, when using an excess of methylamine. Probably, the substitution of the chlorine atom in the chloropropyl residue was not observed due to the lower nucleophilicity of methylamine as compared to dimethylamine. The

Scheme 4.



reaction of ethylenediamine diacetate with nitrile **3** led to the formation of ketenaminal **9**, which was isolated in the acetate form. Attempts to carry out targeted intramolecular alkylation of the ketenaminal moiety with a chloropropyl residue in order to obtain a bicyclic structure **10** were unsuccessful.

Structure and purity of the obtained compounds were proved by the liquid chromatography-mass spectrometry, IR spectroscopy, elemental analysis, and NMR spectroscopy methods. Structure of compounds **4a** and **6** was unambiguously confirmed by means of 2D NMR techniques (COSY, HSQC, HMBC) (Scheme 4). The presence in the  $^1\text{H}$  NMR spectra of signals of aliphatic protons and NH-groups, as well as their multiplicity, make it possible to determine the corresponding fragments in the molecule of compound **4a**. The protons of the methylene (2.75 ppm) and methyl (2.95 ppm) groups in the HMBC spectrum correlate with the  $\text{C}^2$  and  $\text{C}^5$  carbon atoms (152.1, 162.5 ppm), which is characteristic of the 5-aminooxazole fragment.

Structure of compound **6** is confirmed by the presence of the signal of an NH proton at 8.30 ppm in the  $^1\text{H}$  NMR spectrum. The fact that the NH group is directly bonded to the carbon atom at the nitrile group (59.2 ppm) is confirmed by HMBC correlations. Also, the protons of the  $\text{CH}_2$  groups of the diazepine ring in the HMBC spectrum correlate with the carbon atom of the carbonyl group (172.37 ppm). The interaction of protons of two methylene groups (3.31 and 3.42 ppm), as well as one NH-proton with a carbon atom at a double bond (158.2 ppm) indicates the formation of (*Z*)-2,3,5,6,7,8-hexahydro-7-oxo-1*H*-imidazo[1,2-*a*][1,4]diazepine ring.

In summary, we first studied the reactions of *N*-(2,2-dichloro-1-cyanoethyl)prop-2-enamide and *N*-(2,2-dichloro-1-cyanoethyl)-4-chlorobutylamide with some aliphatic amines. At the same time, new representatives of 5-amino-1,3-oxazole-4-carbonitriles

were obtained, as well as (*Z*)-2,3,5,6,7,8-hexahydro-7-oxo-1*H*-imidazo[1,2-*a*][1,4]diazepine-9-carbonitrile and 4-chloro-*N*-[cyano(imidazolidin-2-ylidene)methyl]-butanamide in the case of the reaction with ethylenediamine.

## EXPERIMENTAL

IR spectra were recorded on a Vertex-70 spectrometer from KBr pellets.  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra were taken on a Bruker AVANCE DRX-500 instrument (500 and 125 MHz, respectively) from  $\text{DMSO-}d_6$  or  $\text{CDCl}_3$  solution. Chromato-mass spectra were recorded using a liquid chromatography-mass spectrometry system on an Agilent 1100 Series high performance liquid chromatograph equipped with an Agilent LCMSD SL mass selective detector diode array. Chromatography-mass analysis parameters: column Zorbax SB-C18 1.8  $\mu\text{m}$  4.6  $\times$  15 mm (PN 821975-932); solvents: A,  $\text{MeCN-H}_2\text{O}$ , 95 : 5, 0.1% TFA; B, 0.1% aqueous TFA; eluent flow 3 mL/min; injection volume 1  $\mu\text{L}$ ; UV detectors: 215, 254, 285 nm; ionization method was atmospheric pressure chemical ionization (APCI), scanning range  $m/z$  80–1000 Da. Elemental analysis was performed in the analytical laboratory of the V.P. Kukhar Institute of Bioorganic Chemistry and Petrochemistry of the National Academy of Sciences of Ukraine. The content of carbon and hydrogen was determined by the Pregl gravimetric method, nitrogen—by the Dumas gas-metric micro-method, and chlorine—by the Schöniger titrimetric method [11]. Melting points were measured on a Fisher-Johns apparatus. The reaction progress and the purity of the obtained compounds were monitored by thin layer chromatography on Macherey-Nagel ALUGRAM Xtra SIL G/UV254 plates in the chloroform–methanol system (10 : 0.2).

2-Amino-3,3-dichloroacrylonitrile **1** as well as acrylic and 4-chlorobutanoic acid chlorides are commercial products.

***N*-(2,2-Dichloro-1-cyanoethenyl)prop-2-enamide (2)** and ***N*-(2,2-dichloro-1-cyanoethenyl)-4-chlorobutylamide (3)** were obtained by the previously described method [6]. To a solution of 13.7 g (100 mmol) of 2-amino-3,3-dichloroacrylonitrile **1** and 12.1 g (100 mmol) of *N,N*-dimethylaniline in 100 mL of diethyl ether with vigorous stirring was added dropwise the corresponding acid chloride (100 mmol) at 20–25°C. The resulting mixture was stirred for 12 h, then 200 mL of a water–hexane mixture (1 : 1) was added. A precipitate formed in the two-phase system was filtered off, washed with 50 mL of a hexane–diethyl ether mixture (1 : 1), and dried in a vacuum. Compounds **2**, **3** were used for further transformations without additional purification.

***N*-(2,2-Dichloro-1-cyanoethenyl)prop-2-enamide (2)**. Yield 75%, mp 140–142°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1495, 1599, 1630, 1670, 2230 (CN), 3033–3238 (NH).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 2.21 d (1H,  $\text{CH}_2=\text{CH}$ ,  $^3J_{\text{HH}}$  10.0 Hz), 6.29–6.46 m (2H,  $\text{CH}_2=\text{CH}$ ), 10.48 s (1H, NH).  $^{13}\text{C}$  NMR spectrum (DMSO- $d_6$ ),  $\delta_{\text{C}}$ , ppm: 109.2, 110.8, 127.5, 128.2, 129.8, 161.8 (C=O). Mass spectrum,  $m/z$ : 191 [ $M+H$ ] $^+$ . Found, %: C 37.17; H 2.47; Cl 37.03; N 14.40.  $\text{C}_6\text{H}_4\text{Cl}_2\text{N}_2\text{O}$ . Calculated, %: C 37.73; H 2.11; Cl 37.12; N 14.67.

***N*-(2,2-Dichloro-1-cyanoethenyl)-4-chlorobutylamide (3)**. Yield 80%, mp 97–99°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1443, 1497, 1601, 1671, 2233 (CN), 3085–3284 (NH).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 1.83–1.90 m (2H,  $\text{CH}_2$ ), 2.35–2.38 m (2H,  $\text{CH}_2$  + DMSO), 3.53–3.56 m (2H,  $\text{CH}_2$  +  $\text{H}_2\text{O}$ ), 10.20 s (1H, NH).  $^1\text{H}$  NMR spectrum ( $\text{CDCl}_3$ ),  $\delta$ , ppm: 2.16–2.22 m (2H,  $\text{CH}_2$ ), 2.59 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  7.1 Hz), 3.66 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  6.1 Hz), 7.09 br. s (1H, NH).  $^{13}\text{C}$  NMR spectrum (DMSO- $d_6$ ),  $\delta_{\text{C}}$ , ppm: 25.8 ( $\text{CH}_2$ ), 30.2 ( $\text{CH}_2$ ), 43.1 ( $\text{CH}_2$ ), 109.7, 111.3, 129.9, 169.7 (C=O). Mass spectrum,  $m/z$ : 241 [ $M+H$ ] $^+$ . Found, %: C 34.77; H 3.15; Cl 43.71; N 11.36.  $\text{C}_7\text{H}_7\text{Cl}_3\text{N}_2\text{O}$ . Calculated, %: C 34.81; H 2.92; Cl 44.04; N 11.60.

**5-Alkylamino-2-aminoethyl-1,3-oxazole-4-carbonitriles (4a, 4b)**. To a solution of 10 mmol of compound **2** in 20 mL of methanol with vigorous stirring was added 10 mL of a 40% aqueous solution of dimethylamine or 20 mL of a 20% methanol solution of methylamine at 20–25°C. The solution was stirred for 48 h, the excess of the corresponding amine, and the solvent was removed in vacuum at 40°C. A solution of 6.9 g (50 mmol) of  $\text{K}_2\text{CO}_3$  in 20 mL of water was added to the residue, and compound **4** was extracted with methylene chloride

(3  $\times$  30 mL). The extract was washed with water (4  $\times$  5 mL) and dried with  $\text{Na}_2\text{SO}_4$ , the solvent was removed in vacuum. The obtained compounds were purified by flash chromatography, using a mixture  $\text{CH}_2\text{Cl}_2$ –methanol as eluent.

**5-(Methylamino)-2-[2-(methylamino)ethyl]-1,3-oxazole-4-carbonitrile (4a)**. Yield 20%, mp 107–109°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1473, 1489, 1604, 1679, 2198 (CN), 3056–3263 (NH).  $^1\text{H}$  NMR spectrum ( $\text{CDCl}_3$ ),  $\delta$ , ppm: 2.41 s (3H,  $\text{CH}_3$ ), 2.75 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  7.5 Hz), 2.93 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  7.5 Hz), 2.95 s (3H,  $\text{CH}_3$ ), 6.47 br. s (1H, NH).  $^{13}\text{C}$  NMR spectrum ( $\text{CDCl}_3$ ),  $\delta_{\text{C}}$ , ppm: 27.9 ( $\text{CH}_2$ ), 29.8 ( $\text{CH}_3$ ), 35.9 ( $\text{CH}_3$ ), 48.2 ( $\text{CH}_2$ ), 83.2 ( $\text{C}^4$ , oxazole), 115.8 (CN), 152.1 ( $\text{C}^2$ , oxazole), 161.5 ( $\text{C}^5$ , oxazole). Mass spectrum,  $m/z$ : 181 [ $M+H$ ] $^+$ . Found, %: C 53.27; H 6.89; N 30.78.  $\text{C}_8\text{H}_{12}\text{N}_4\text{O}$ . Calculated, %: C 53.32; H 6.71; N 31.09.

**5-(Dimethylamino)-2-[2-(dimethylamino)ethyl]-1,3-oxazole-4-carbonitrile (4b)**. Yield 90%, mp 38–40°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1464, 1604, 1654 sh, 2204 (CN).  $^1\text{H}$  NMR spectrum ( $\text{CDCl}_3$ ),  $\delta$ , ppm: 1.97 s (6H,  $\text{CH}_3$ ), 2.36 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  7.0 Hz), 2.45 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  7.0 Hz), 2.83 s (6H,  $\text{CH}_3$ ).  $^{13}\text{C}$  NMR spectrum ( $\text{CDCl}_3$ ),  $\delta_{\text{C}}$ , ppm: 24.9 ( $\text{CH}_2$ ), 37.4, 43.8 (2 $\text{CH}_3$ ), 54.6, 83.1 ( $\text{C}^4$ , oxazole), 111.5 (CN), 151.3 ( $\text{C}^2$ , oxazole), 160.3 ( $\text{C}^5$ , oxazole). Mass spectrum,  $m/z$ : 209 [ $M+H$ ] $^+$ . Found, %: C 57.81; H 7.50; N 27.32.  $\text{C}_{10}\text{H}_{16}\text{N}_4\text{O}$ . Calculated, %: C 57.67; H 7.74; N 26.90.

**(Z)-2,3,5,6,7,8-Hexahydro-7-oxo-1H-imidazo-[1,2-*a*][1,4]diazepine-9-carbonitrile (6)**. To a solution of 7.2 g (40 mmol) of ethylenediamine diacetate in 20 mL of methanol with vigorous stirring was added in portions (0.2 g each) 1.9 g (10 mmol) of compound **2** at 20–25°C. The reaction mixture was stirred for 48 h, then the precipitate was filtered off, washed with water (2  $\times$  5 mL), and dried at 50°C. Compound **6** was analyzed without further purification. Yield 20%, dp 190–195°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1432, 1480, 1503, 1608, 1654, 2172 (CN), 3044–3243 (NH).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 2.46 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  5.4 Hz), 3.29–3.34 br. m (2H,  $\text{CH}_2$ ), 3.42 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  5.4 Hz), 3.47 t (2H,  $\text{CH}_2$ ,  $^3J_{\text{HH}}$  8.1 Hz), 6.27 s (1H, NH), 8.30 s (1H, NH).  $^{13}\text{C}$  NMR spectrum (DMSO- $d_6$ ),  $\delta_{\text{C}}$ , ppm: 35.2 ( $\text{CH}_2$ ), 42.0 ( $\text{CH}_2$ ), 49.0 ( $\text{CH}_2$ ), 53.5 ( $\text{CH}_2$ ), 59.2 [ $\text{C}=\text{C}(\text{NCH}_2)_2$ ], 121.9 (CN), 158.2 [ $\text{C}=\text{C}(\text{NCH}_2)_2$ ], 172.4 (C=O). Mass spectrum,  $m/z$ : 179 [ $M+H$ ] $^+$ . Found, %: C 54.15; H 5.79; N 31.76.  $\text{C}_8\text{H}_{10}\text{N}_4\text{O}$ . Calculated, %: C 53.92; H 5.66; N 31.44.

**5-(Dimethylamino)-2-[3-(dimethylamino)propyl]-1,3-oxazole-4-carbonitrile (7).** To a solution of 10 mmol of compound **3** in 20 mL of methanol with vigorous stirring was added 10 mL of a 40% aqueous solution of dimethylamine at 20–25°C. The solution was stirred for 48 h, an excess of amine and the solvent were removed in vacuum at 40°C. A solution of 6.9 g (50 mmol) of K<sub>2</sub>CO<sub>3</sub> in 20 mL of water was added to the residue and extracted with methylene chloride (3 × 30 mL). The extract was washed with water (4 × 5 mL) and dried with Na<sub>2</sub>SO<sub>4</sub>, the solvent was removed in vacuum. Compound **7** was purified by flash chromatography eluting with CH<sub>2</sub>Cl<sub>2</sub>–methanol mixture. Yield 70%, oil. IR spectrum,  $\nu$ , cm<sup>-1</sup>: 1444, 1600, 1641, 2207 (CN). <sup>1</sup>H NMR spectrum (CDCl<sub>3</sub>),  $\delta$ , ppm: 1.80–1.85 m (2H, CH<sub>2</sub>), 2.20 s (6H, CH<sub>3</sub>), 2.30 t (2H, CH<sub>2</sub>, <sup>3</sup>J<sub>HH</sub> 7.0 Hz), 2.62 t (2H, CH<sub>2</sub>, <sup>3</sup>J<sub>HH</sub> 7.5 Hz), 3.11 s (6H, CH<sub>3</sub>). <sup>13</sup>C NMR spectrum (CDCl<sub>3</sub>),  $\delta$ <sub>C</sub>, ppm: 22.3 (CH<sub>2</sub>), 23.3 (CH<sub>2</sub>), 36.7 (2CH<sub>3</sub>), 43.3 (2CH<sub>3</sub>), 56.5 (CH<sub>2</sub>), 82.2 (C<sup>4</sup>, oxazole), 114.9 (CN), 152.1 (C<sup>2</sup>, oxazole), 159.6 (C<sup>5</sup>, oxazole). Mass spectrum,  $m/z$ : 223 [*M* + H]<sup>+</sup>. Found, %: C 59.47; H 8.42; N 25.59. C<sub>11</sub>H<sub>18</sub>N<sub>4</sub>O. Calculated, %: C 59.44; H 8.16; N 25.20.

**5-(Methylamino)-2-(3-chloropropyl)-1,3-oxazole-4-carbonitrile (8)** was prepared similarly to oxazole **7** from compound **3** and 20 mL of a 20% methanol solution of methylamine. Yield 70%, mp 69–71°C. IR spectrum,  $\nu$ , cm<sup>-1</sup>: 1446, 1467, 1599, 1660, 2206 (CN), 3078–3325 (NH). <sup>1</sup>H NMR spectrum (CDCl<sub>3</sub>),  $\delta$ , ppm: 1.86–1.93 m (2H, CH<sub>2</sub>), 2.51 t (2H, CH<sub>2</sub>, <sup>3</sup>J<sub>HH</sub> 7.5 Hz), 2.79 d (3H, CH<sub>3</sub>, <sup>3</sup>J<sub>HH</sub> 6.5 Hz), 3.49 t (2H, CH<sub>2</sub>, <sup>3</sup>J<sub>HH</sub> 6.5 Hz), 5.05 br. s (1H, NH). <sup>13</sup>C NMR spectrum (CDCl<sub>3</sub>),  $\delta$ <sub>C</sub>, ppm: 23.3, 27.5, 28.6, 42.2, 82.8 (C<sup>4</sup>, oxazole), 114.4 (CN), 151.9 (C<sup>2</sup>, oxazole), 161.0 (C<sup>5</sup>, oxazole). Mass spectrum,  $m/z$ : 200 [*M* + H]<sup>+</sup>. Found, %: C 48.32; H 5.28; Cl 17.57; N 21.21. C<sub>8</sub>H<sub>10</sub>ClN<sub>3</sub>O. Calculated, %: C 48.13; H 5.05; Cl 17.76; N 21.05.

**4-Chloro-*N*-[cyano(imidazolidin-2-ylidene)methyl]-butanamide (9).** To a solution of 7.2 g (40 mmol) of ethylenediamine diacetate in 20 mL of methanol 2.4 g (10 mmol) was added in portions (0.2 g each) with vigorous stirring compound **3** at 20–25°C. The reaction mixture was stirred for 48 h, and ethylenediamide hydrochloride was filtered off. The solvent was removed in vacuum at 40°C, then 20 mL of water was added to the residue, and the product was extracted with a mixture of CH<sub>2</sub>Cl<sub>2</sub>–propan-2-ol (8 : 2, 6 × 20 mL). The solvent was removed in vacuum, the residue was treated with 20 mL of propan-2-ol, and the crystals were filtered off to

give compound **9** as acetate. Yield 70%, mp 61–63°C. IR spectrum,  $\nu$ , cm<sup>-1</sup>: 1447, 1475, 1543, 1601, 1654, 2207 (CN), 3075–3417 (NH). <sup>1</sup>H NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ , ppm: 1.79 s (3H, CH<sub>3</sub>), 2.01–2.10 m (2H, CH<sub>2</sub>), 2.71 t (2H, CH<sub>2</sub>, <sup>3</sup>J<sub>HH</sub> 7.3 Hz), 2.85 t (2H, CH<sub>2</sub>, <sup>3</sup>J<sub>HH</sub> 6.0 Hz), 3.35 t (2H, CH<sub>2</sub>, <sup>3</sup>J<sub>HH</sub> 6.0 Hz), 3.70 t (2H, CH<sub>2</sub>, <sup>3</sup>J<sub>HH</sub> 6.4 Hz), 6.65 br. s (4H, 3NH, OH). <sup>13</sup>C NMR spectrum (DMSO-*d*<sub>6</sub>),  $\delta$ <sub>C</sub>, ppm: 22.0, 23.2, 24.5, 27.8, 42.4 (CH<sub>2</sub>), 43.3 (CH<sub>2</sub>), 80.8 [C=C(NHCH<sub>2</sub>)<sub>2</sub>], 115.7 (CN), 151.7 [C=C(NHCH<sub>2</sub>)<sub>2</sub>], 160.9 (C=O), 174.0 (C=O). Mass spectrum,  $m/z$ : 229 [*M* + H – CH<sub>3</sub>COOH]<sup>+</sup>. Found, %: C 45.99; H 6.21; Cl 12.20; N 19.62. C<sub>11</sub>H<sub>17</sub>ClN<sub>4</sub>O<sub>3</sub>. Calculated, %: C 45.76; H 5.93; Cl 12.28; N 19.40.

To obtain free base **9**, 2 g of acetate was dissolved in 5 mL of water, and then 5 mL of a saturated aqueous solution of NaHCO<sub>3</sub> was added. The formed precipitate was filtered off, washed with water (2 × 3 mL) and dried in vacuum at 40°C. Yield 85%, mp 80–82°C. Mass spectrum,  $m/z$ : 229 [*M* + H]<sup>+</sup>. Found, %: C 47.04; H 5.70; Cl 15.79; N 24.88. C<sub>9</sub>H<sub>13</sub>ClN<sub>4</sub>O. Calculated, %: C 47.27; H 5.73; Cl 15.50; N 24.50.

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#### CONFLICT OF INTEREST

No conflict of interest was declared by the authors.

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