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**RESEARCH INTO ONE-STEP THREE COMPONENT REACTION OF SOME
YLIDENE CYANOACETAMIDES (OR YLIDENEMALONONITRILES),
MALONONITRILE AND 1,3-DIAMINOPROPANE**

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Received 21.04.2019

Abstract: The one-step interaction of substituted ylidene cyanoacetamides (or ylidene malononitriles), malononitrile and 1,3-diaminopropane was carried out in methanol environment at room temperature and it established the formation of new substituted dihydropyridopyrimidine derivatives. It also revealed that under the same conditions through the use of one-step three-component reaction of pyridylidenecyanoacetamide (or pyridylidenemalononitrile), 2-chloro-5-nitrobenzylidenecyanoacetamide (or 2-chloro-5-nitrobenzylidenemalononitrile), 2,6-dichlorobenzylidenecyanoacetamide (or 2,6-dichlorobenzylidenemalononitrile), malononitrile and 1,3-diaminopropane there formed substituted derivatives not of dihydropyridopyrimidine but tetrahydropyridopyrimidine. Structures of all synthesized compounds were verified by NMR and X-Ray spectroscopy.

Keywords: ylidene cyanoacetamides, pyridylidenecyanoacetamide, malononitrile, 1,3-diaminopropane, NMR

DOI: 10.32737/2221-8688-2019-2-275-281

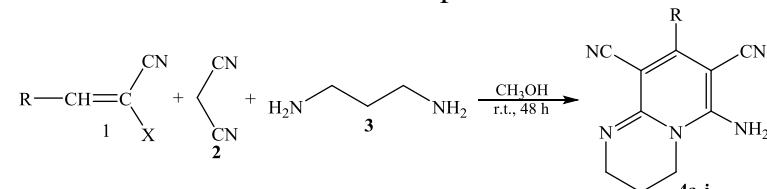
Introduction

The literature refers to new methods of theirv ibhibiting properties in respect of production of pyrimidine derivatives and various disease-producing factors [1-12].

Results and discussion

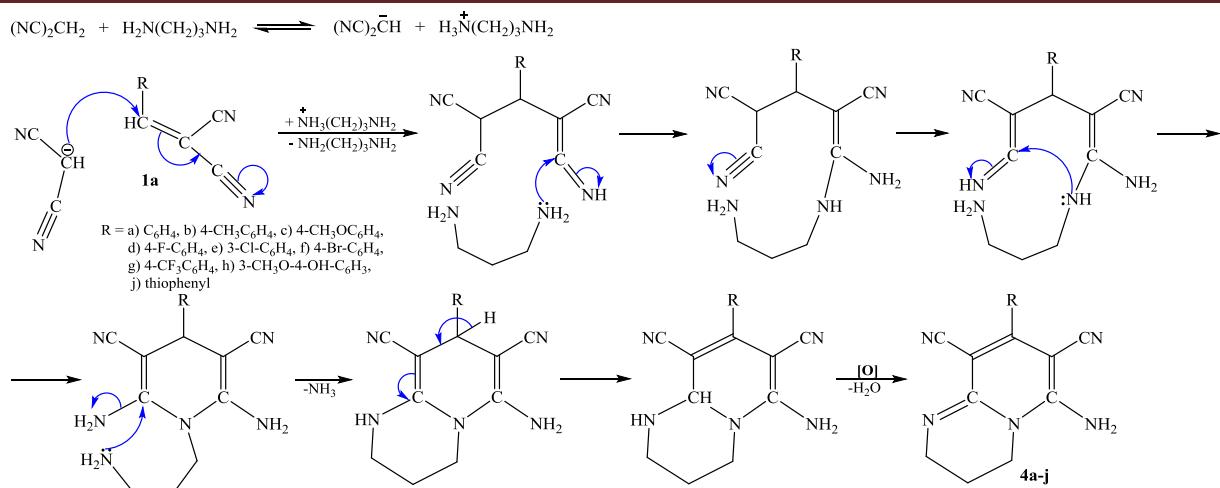
We carried out the one-step three component interaction of various ylidene cyanoacetamides (or ylidene malononitriles), malononitrile and 1,3-diaminopropane in methanol environment and

at room temperature, for 24-28 hours and established the possibility of production of substituted new dihydropyrido[1,2-*a*]pyrimidines (**4a-j**) irrespective of the nature of polarized double bond.



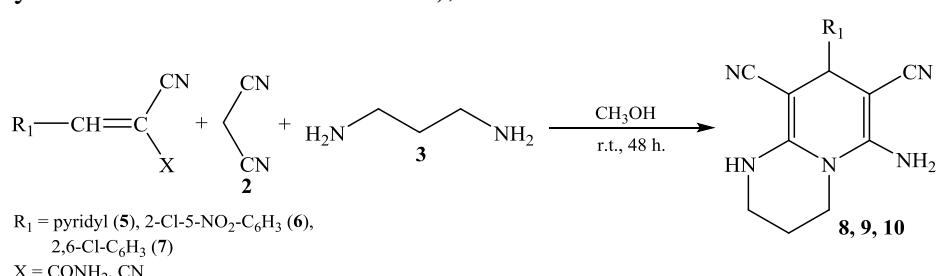
R = a) C₆H₅, b) 4-CH₃C₆H₄, c) 4-CH₃OC₆H₄,
d) 4-F-C₆H₄, e) 3-Cl-C₆H₄, f) 4-Br-C₆H₄,
g) 4-CF₃C₆H₄, h) 3-CH₃O-4-OH-C₆H₃,
j) thiophenyl
X = CONH₂, CN

A plausible mechanism of the reaction is presented below:

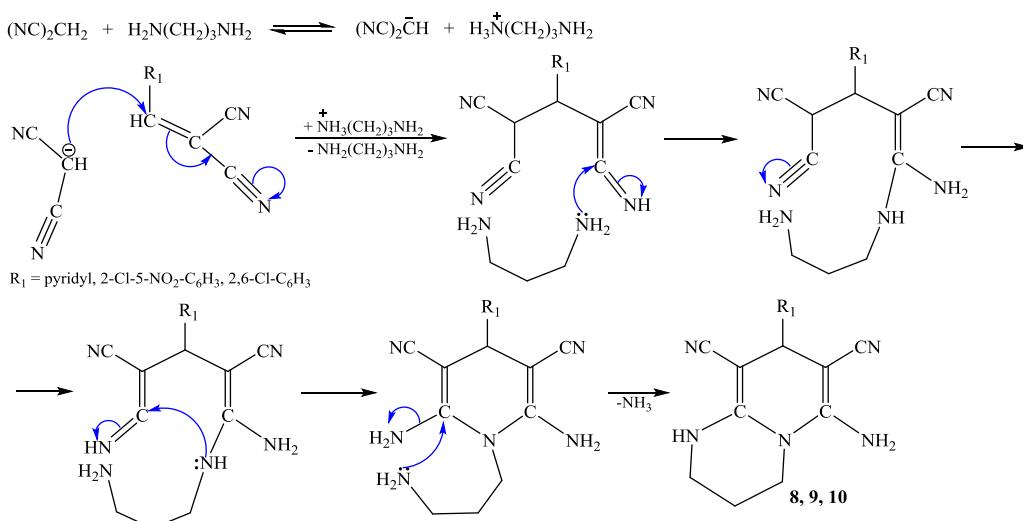


Through the use of similar one-step three-component reaction of pyridylidene cyanoacetamide (or pyridylidenemalononitrile) as compound with polarized double-bond, 2,6-dichlorobenzylidene cyanoacetamide (or 2,6-dichlorobenzylidene malononitrile),

malononitrile and 1,3-diaminopropane at the same conditions, it became possible to establish the formation of corresponding substituted tetrahydropyrido[1,2-a]pyrimidine derivatives (**6**, **8**, **10**), not dihydropyrido[1,2-a]pyrimidines.



A plausible reaction mechanism and ^1H , ^{13}C NMR spectra (fig.1, fig.2) of compound **4e** is presented below.



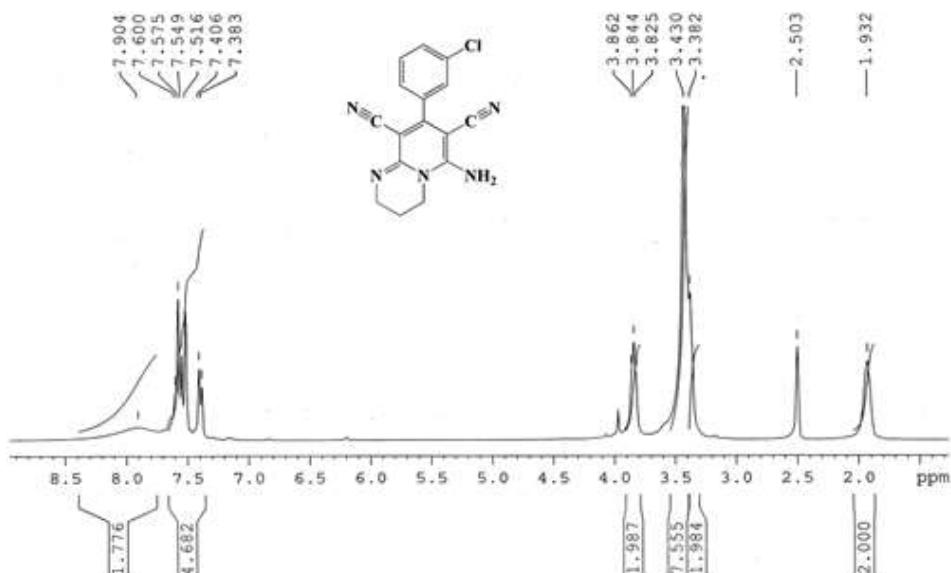


Fig. 1. ^1H NMR spectrum of 6-amino-8-(3-chlorophenyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (**4e**).

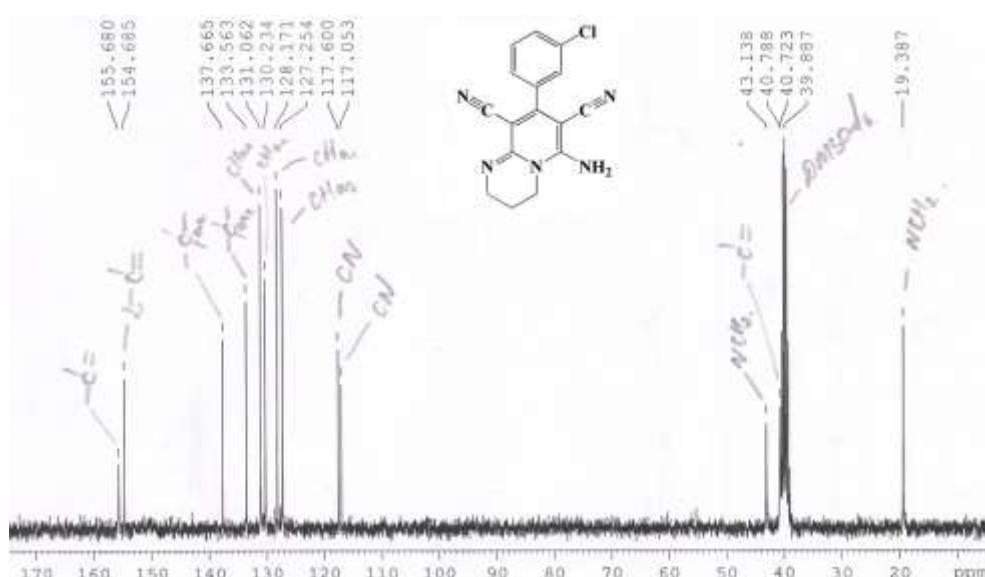


Fig. 2. ^{13}C NMR spectrum of 6-amino-8-(3-chlorophenyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (**4e**).

Experimental part. General remarks

All commercially available chemicals were obtained from Merck and Fluka (Sigma Aldrich) companies and used without further purification. Melting points were measured on Stuart SMP30 apparatus without correction. ^1H , ^{13}C NMR spectra were recorded on Bruker Avance 300-MHz spectrometer at 300 and 75 MHz, respectively. Thin-layer chromatography (TLC) on commercial aluminum-backed plates of silica gel (60 F254) was used to moni

nitor the course of reactions.

General experimental procedure

6-Amino-8-phenyl-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4a): Benzylidenecyanoacetamide, (4 mmol) malononitril (4.1 mmol) and 1,3-diaminopropane (4.1 mmol) stirred in 35 ml of methyl alcohol. Then the reaction mixture is maintained at room temperature for 2 days. The course of the reaction was monitored by

TLC (EtOAc/n-hexane, 3:2). Crystals were precipitated after evaporation of solvent, filtered by paper, recrystallized from ethanol-water mixture and obtained in pure form (yield 0.81 g, 72.97%). $T_{mp.}=234^{\circ}\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.94 (t, 2H, CH₂, $^3J_{\text{H-H}}=6,6$); 3.38 (t, 2H, CH₂, $^3J_{\text{H-H}}=6,3$); 3.86 (t, 2H, CH₂, $^3J_{\text{H-H}}=5,7$); 7.41-7.52 (m, 5H, 5Ar-H); 7.84 (s, 2H, NH₂). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.39 (CH₂), 40.72 (=C_{quat.}), 42.99 (CH₂), 43.04 (CH₂), 117.25 (CN), 117.85 (CN), 128.39 (2CH_{arom}), 128.99 (2CH_{arom}), 130.30 (CH_{arom}), 135.67 (C_{ar}), 155.81 (=C_{quat.}), 156.27 (2=C_{quat.}).

Found, %: 69.76 C; 4.68 H, 25.50 N. C₁₆H₁₃N₅. Calculated, %: 69.82 C; 4.73 H, 25.45 N.

6-Amino-8-(*p*-tolyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4b):

Synthesized by the same way(yield 0.88 g, 75.86%). $T_{mp.}=250^{\circ}\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.93 (t, 2H, CH₂, $^3J_{\text{H-H}}=6,7$); 2.37 (s, 3H, CH₃-Ar); 3.37 (t, 2H, CH₂, $^3J_{\text{H-H}}=6,3$); 3.85 (t, 2H, CH₂, $^3J_{\text{H-H}}=6,9$); 7.31 (m, 4H, 4Ar-H); 7.82 (s, 2H, NH₂). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.37 (CH₂), 21.37 (Ar-CH₃), 40.54 (CH₂), 40.66 (2=C_{quat.}), 42.99 (CH₂), 117.33 (CN), 117.96 (CN), 128.35 (2CH_{arom}), 129.52 (2CH_{arom}), 132.72 (C_{ar}), 140.08 (C_{ar}), 155.85 (=C_{quat.}), 156.31 (2=C_{quat.}).

Found, %: 70.64 C; 5.25 H, 24.16 N. C₁₇H₁₅N₅. Calculated, %: 70.59 C; 5.19 H, 24.22 N.

6-Amino-8-(4-methoxyphenyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4c):

Synthesized by the same way (yield 0.9 g, 73.17%). $T_{mp.}=231^{\circ}\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.93 (s, 2H, CH₂); 3.37 (s, 2H, CH₂); 3.81 (s, 3H, CH₃O-Ar); 3.85 (s, 2H, CH₂); 7.05 (d, 2H, 2Ar-H, $^3J_{\text{H-H}}=8,1$); 7.39 (d, 2H, 2Ar-H, $^3J_{\text{H-H}}=7,8$); 7.80 (s, 2H, NH₂). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.40 (CH₂), 38.91 (2=C_{quat.}), 40.52 (CH₂), 42.99 (CH₂), 55.73 (CH₃O), 114.28 (2CH_{arom}), 117.50 (CN), 118.13 (CN), 127.58 (C_{ar}), 127.59 (=C_{quat.}), 130.12 (2CH_{arom}), 155.88 (=C_{quat.}), 155.94 (=C_{quat.}), 160.85 (O-C_{ar}).

Found, %: 66.82 C; 4.87 H, 23.01 N. C₁₇H₁₅N₅O. Calculated, %: 66.88 C; 4.92 H, 22.95 N.

6-Amino-8-(4-fluorophenyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4d):

Synthesized by the same way (yield 0.79 g, 67.52%). $T_{mp.}=238^{\circ}\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.93 (s, 2H, CH₂); 3.38 (s, 2H, CH₂); 3.85 (s, 2H, CH₂); 7.12-7.52 (m, 4H, 4Ar-H); 7.96 (s, 2H, NH₂). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.37 (CH₂), 39.02 (2=C_{quat.}), 40.68 (CH₂), 43.06 (CH₂), 115.91-116.20 (2CH_{arom}), 117.22 (CN), 117.78 (CN), 130.89-131.00 (2CH_{arom}), 132.02 (C_{ar}), 155.33 (2=C_{quat.}), 155.75 (=C_{quat.}), 161.66-164.92 (F-C_{ar}).

Found, %: 65.59 C; 4.14 H, 23.84 N. C₁₆H₁₂N₅F. Calculated, %: 65.53 C; 4.09 H, 23.89 N.

6-Amino-8-(3-chlorophenyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4e):

Synthesized by the same way (yield 0.91 g, 73.39%). $T_{mp.}=254^{\circ}\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.93 (s, 2H, CH₂); 3.38 (s, 2H, CH₂); 3.84 (s, 2H, CH₂); 7.38-7.60 (m, 4H, 4Ar-H); 7.90 (s, 2H, NH₂). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.39 (CH₂), 40.72 (CH₂), 40.79 (2=C_{quat.}), 43.14 (CH₂), 117.05 (CN), 117.60 (CN), 127.25 (CH_{arom}), 128.17 (CH_{arom}), 130.23 (CH_{arom}), 131.06 (CH_{arom}), 133.56 (C_{ar}), 137.66 (C_{ar}), 154.68 (2=C_{quat.}), 155.68 (=C_{quat.}).

Found, %: 62.09 C; 3.93 H, 22.56 N. C₁₆H₁₂N₅Cl. Calculated, %: 62.03 C; 3.88 H, 22.62 N.

6-Amino-8-(4-bromophenyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4f):

Synthesized by the same way (yield 0.89 g, 62.68%). $T_{mp.}=278^{\circ}\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.93 (s, 2H, CH₂); 3.37 (s, 2H, CH₂); 3.83 (s, 2H, CH₂); 7.38 (d, 2H, 2Ar-H, $^3J_{\text{H-H}}=8,1$); 7.73 (d, 2H, 2Ar-H, $^3J_{\text{H-H}}=8,1$); 7.93 (s, 2H, NH₂). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.37 (CH₂), 40.69 (CH₂), 40.72 (2=C_{quat.}), 43.15 (CH₂), 117.15 (CN), 117.69 (CN), 123.85 (Br-C_{ar}), 130.60 (2CH_{arom}), 132.09 (2CH_{arom}), 134.87 (C_{ar}), 155.15 (2=C_{quat.}), 155.67 (=C_{quat.}).

Found, %: 54.19 C; 3.33 H, 19.82 N. $C_{16}H_{12}N_5Br$. Calculated, %: 54.24 C; 3.39 H, 19.77 N.

6-Amino-8-(4-(trifluoromethyl)phenyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4g): Synthesized by the same way (yield 0.9 g, 65.69%). $T_{mp.} = 331^\circ\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.94 (s, 2H, CH₂); 3.38 (s, 2H, CH₂); 3.84 (s, 2H, CH₂); 7.66 (d, 2H, 2Ar-H, $^3J_{\text{H-H}} = 7.8$); 7.90 (d, 2H, 2Ar-H, $^3J_{\text{H-H}} = 7.8$); 7.96 (s, 2H, NH₂). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.35 (CH₂), 40.62 (CH₂), 40.75 (=C_{quat.}), 43.19 (CH₂), 117.02 (CN), 117.54 (CN), 126.03-126.17 (2CH_{arom}), 129.50 (2CH_{arom}), 130.28 (CF₃-C_{ar}), 130.70 (CF₃), 139.80 (C_{ar}), 154.93 (=C_{quat.}), 155.66 (=C_{quat.}).

Found, %: 59.41 C; 3.45 H, 20.47 N. $C_{17}H_{12}N_5F_3$. Calculated, %: 59.47 C; 3.50 H, 20.41 N.

6-Amino-8-(4-hydroxy-3-methoxyphenyl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4h): Synthesized by the same way (yield 0.93 g, 72.66%). $T_{mp.} = 181^\circ\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.93 (s, 2H, CH₂); 3.36 (s, 2H, CH₂); 3.78 (s, 3H, CH₃O); 3.85 (s, 2H, CH₂); 6.86-7.61 (m, 6H, 3Ar-H+Ar-OH+NH₂). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.53 (CH₂), 40.62 (CH₂), 42.85 (CH₂), 56.13 (CH₃O), 76.70 (=C_{quat.}), 80.42 (=C_{quat.}), 112.88 (CH_{arom}), 115.80 (CH_{arom}), 117.80 (CN), 118.52 (CN), 121.81 (CH_{arom}), 125.67 (C_{ar}), 147.70 (=C_{quat.}), 149.42 (=C_{quat.}), 151.57 (=C_{quat.}), 156.14 (O-C_{ar}), 156.24 (O-C_{ar}).

Found, %: 63.60 C; 4.73 H, 21.76 N. $C_{17}H_{15}N_5O_2$. Calculated, %: 63.55 C; 4.67 H, 21.81 N.

6-Amino-8-(thiophen-2-yl)-3,4-dihydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (4j): Synthesized by the same way (yield 0.76 g, 67.86%). $T_{mp.} = 227^\circ\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.93 (m, 2H, CH₂); 3.37 (s, 2H, CH₂); 3.84 (s, 2H, CH₂); 7.21 (t, 1H, Thioph-H, $^3J_{\text{H-H}} = 4.2$); 7.41 (d, 1H, Thioph-H, $^3J_{\text{H-H}} = 3$); 7.76 (s, 2H, NH₂); 7.83 (d, 1H, Thioph-H, $^3J_{\text{H-H}} = 4.8$). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 19.34 (CH₂), 40.68 (=C_{quat.}), 40.72 (CH₂), 43.18

(CH₂), 117.27 (CN), 117.89 (CN), 127.93 (CH_{thioph}), 129.79 (CH_{thioph}), 130.47 (CH_{thioph}), 134.67 (C_{thioph}), 148.41 (=C_{quat.}), 155.70 (=C_{quat.}), 162.35 (=C_{quat.}).

Found, %: 59.74 C; 3.85 H, 24.96 N. $C_{14}H_{11}N_5S$. Calculated, %: 59.79 C; 3.91 H, 24.91 N.

6-Amino-8-(pyridin-4-yl)-1,3,4,8-tetrahydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (6): Synthesized by the same way (yield 0.85 g, 76.57%). $T_{mp.} = 242^\circ\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.88 (m, 2H, CH₂); 3.13 (m, 2H, CH₂); 3.60 (m, 2H, CH₂); 4.06 (s, 1H, CH-Ar); 6.27 (s, 2H, NH₂); 6.90 (s, 1H, NH); 7.18 (m, 2H, 2Ar-H); 8.51 (m, 2H, 2Ar-H). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 22.01 (CH₂), 38.61 (CH₂), 38.94 (Ar-CH), 43.07 (CH₂), 54.05 (=C_{quat.}), 57.64 (=C_{quat.}), 121.90 (2CN), 122.28 (2CH_{arom}), 150.31 (2CH_{arom}), 151.26 (C_{ar}), 153.21 (=C_{quat.}), 155.15 (=C_{quat.}).

Found, %: 64.69 C; 4.98 H, 30.26 N. $C_{15}H_{14}N_6$. Calculated, %: 64.75 C; 5.03 H, 30.21 N.

6-Amino-8-(2-chloro-5-nitrophenyl)-1,3,4,8-tetrahydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (8): Synthesized by the same way (yield 1 g, 69.93%). $T_{mp.} = 243^\circ\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.90 (m, 2H, CH₂); 3.18 (m, 2H, CH₂); 3.64 (s, 2H, CH₂); 4.73 (s, 1H, CH-Ar); 6.36 (s, 2H, NH₂); 7.00 (s, 1H, NH); 7.71 (d, 1H, Ar-H); 8.07 (d, 2H, 2Ar-H). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 22.12 (CH₂), 37.14 (Ar-CH), 38.64 (CH₂), 43.20 (CH₂), 53.64 (=C_{quat.}), 57.02 (=C_{quat.}), 121.48 (CN), 121.89 (CN), 123.65 (CH_{arom}), 124.58 (CH_{arom}), 131.64 (CH_{arom}), 138.68 (C_{ar}), 145.56 (C_{ar}), 147.37 (C_{ar}), 151.69 (=C_{quat.}), 153.70 (=C_{quat.}).

Found, %: 53.91 C; 3.71 H, 23.50 N. $C_{16}H_{13}N_6O_2Cl$. Calculated, %: 53.86 C; 3.65 H, 23.56 N.

6-Amino-8-(2,6-dichlorophenyl)-1,3,4,8-tetrahydro-2H-pyrido[1,2-a]pyrimidine-7,9-dicarbonitrile (10): Synthesized by the same way (yield 1.07 g, 77.54%). $T_{mp.} = 268^\circ\text{C}$.

^1H NMR (300 MHz, DMSO-*d*6): 1.89 (m, 2H, CH₂); 3.13 (m, 2H, CH₂); 3.67 (s, 2H,

CH_2); 5.31 (s, 1H, CH-Ar); 6.14 (s, 2H, NH_2); 6.78 (s, 1H, NH); 7.25 (t, 1H, Ar-H, $^3J_{\text{H-H}} = 8.1$); 7.42 (d, 2H, 2Ar-H, $^3J_{\text{H-H}} = 7.8$). ^{13}C NMR spektr (DMSO-*d*6), δ , m.h.: 22.30 (CH_2), 36.32 (Ar-CH), 38.62 (CH_2), 42.92 (CH_2), 51.70 (=C_{quat.}), 55.06 (=C_{quat.}), 121.61 (CN), 122.04 (CN), 129.56 (3CH_{arom}), 138.25 (C_{ar}), 152.11 (Cl-C_{ar}), 152.12 (=C_{quat.}), 154.16 (=C_{quat.}), 154.17 (Cl-C_{ar}). Found, %: 55.43 C; 3.71 H, 20.28 N. $\text{C}_{16}\text{H}_{13}\text{N}_5\text{Cl}_2$. Calculated, %: 55.49 C; 3.76 H, 20.23 N.

This study was performed under financial support by the Baku State University (grant 50+50).

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**BƏZİ İLİDENSİANOASETAMİDLƏRİN (VƏ YA İLİDENMALONONİTRİLLƏRİN)
MALONONİTRİL VƏ 1,3-DİAMİNOPROPAN İLƏ BİRMƏRHƏLƏLİ, ÜÇKOMPOONENTLİ
REAKSİYASININ TƏDQİQİ**

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Əvəzlənmiş ilidensianoasetamidlər (və ya ilidenmalononitrillərin) malononitril və 1,3-diaminopropan ilə birmərhələli, üçkomponentli reaksiyası metanol mühitində, otaq temperaturunda aparılmış və əvəzlənmiş yeni dihydropyridopirimidin törəmələrinin əmələ gəldiyi müəyyən edilmişdir. Eyni reaksiya şəraitində piridilidensianoasetamid (və ya piridilidensianoasetamid) (və ya piridilidensianoasetamid), 2-xlor-5-nitrobenzylidensianoasetamid (və ya 2-xlor-5-nitrobenzylidensanoasetamid), 2,6-dixlorbenzylidensanoasetamid (və ya 2,6-dixlorbenzylidensanoasetamid) və 1,3-diaminopropan ilə bir-mərhələli, üçkomponentli reaksiyasından uyğun əvəzlənmiş dihydropyridopirimidinlər deyil, tetrahydropyridopirimidin törəmələrinin əmələ gəlməsi müəyyənləşdirilmişdir. Bütün sintez edilmiş birləşmələrin quruluşu NMR spektroskopiyası ilə təsdiq edilmişdir.

Keywords: ilidensianoasetamidlər, piridilidensianoasetamid, malononitril, 1,3-diaminopropan, NMR

**ИССЛЕДОВАНИЕ ОДНОСТАДИЙНОЙ ТРЕХКОМПОНЕНТНОЙ РЕАКЦИИ НЕКОТОРЫХ
ИЛИДЕНЦИАНОАЦЕТОАМИДОВ (ИЛИ ИЛИДЕНМАЛОННОНИТРИЛОВ),
МАЛОНОНИТРИЛА И 1,3-ДИАМИНОПРОПАНА**

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Осуществлено одностадийное взаимодействие замещенных илиденцианоацетамидов (или илиденмалононитрилов), малононитрила и 1,3-диаминопропана в среде метанола при комнатной температуре, и установлено образование новых замещенных производных дигидропиридопиримидина. Показано, что в тех же условиях путем одностадийной трехкомпонентной реакции пиридилиденцианоацетамида (или пиридилиденмалононитрила), 2-хлор-5-нитробензилиденцианоацетамида (или 2-хлор-5-нитробензилиден малононитрила), 2,6-дихлорбензилиденцианоацетамида (или 2,6-дихлорбензилиденмалононитрила), малононитрила и 1,3-диаминопропана образуются замещенные производные не дигидропиридопиримидина, а тетрагидропиридопиримидина. Структуры синтезированных соединений подтверждены методом ЯМР-спектроскопии.

Ключевые слова: илиденцианоацетамиды, пиридилиденцианоацетамид, малононитрил, 1,3-диаминопропан, ЯМР