Polycyclic heterocycles with acidic N-H group VII

Synthesis of some polynuclear heterocyclic compounds derived from 5-phenyl-6-azauracil

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This paper is dedicated to Professor Edmunds Lukevics

Abstract
Diazonium salt 2 obtained by diazotisation of 3-[3-(6-azauracil-5-yl)-2-aminophenyl]-1,2-dihydro-quinoxalin-3-one ¹² was used as a starting compound for the preparation of corresponding 2-hydroxy, 2-chloro, 2-bromo, and 2-iodo derivatives 4-7 using SN1 or Sandmeyer reaction. The diazonium salt 2 was converted by reductive cleavage to 3-[3-(6-azauracil-5-yl)phenyl]-1,2-dihydro-quinoxalin-2-one 3 and its coupling with malononitrile or ethyl cyanoacetylcarbamate gave corresponding hydrazones 9 or 10. Hydrazone 9 was converted to 4-arylazo-3,5-diaminopyrazole 11 via cyclization with hydrazine, and hydrazone 10 was cyclized to 1-[2-(6-azauracil-5-yl)-6-(3,4-dihydro-3-oxo-quinoxalin-2-yl)phenyl]-6-azauracil-5-carbonitrile 12. The slow decomposition of diazonium salt 2 led to benzofuro- derivative 13 that was also found as an undesired side product of all the above mentioned SN1 reactions. Hydrazino derivative 8, which is an intermediate of diazonium salt 2 reduction or hydrazone 10 hydrolysis, was cyclized, without prior isolation, to corresponding derivative [1,2,4]triazino[5,6-c]cinnoline 14.

Keywords: Heterocyclic N-H acids, polynuclear 6-azauracils, substituted 1,2-dihydro-quinoxalin-2-ones, [1,2,4]triazines

Introduction

Great attention is currently paid to the compounds that may influence spatial arrangement of biomolecules, mainly in relationship with the research of the compounds, which could affect the
conformation of prion proteins. From that reason, we have focused on some polynuclear N-H acids that might be interesting from this point of view.

In this note, we have focused on the synthesis of the compounds containing a free-rotating 1,2-dihydro-quinoxaline and 6-azauracil cycles that are, in contrast to the compounds mentioned in the paper, connected by carbon atom in position 5. This fact enables the possibility of ribosylation on nitrogen atom in position 1 with saving the acidic N-H group in position 3.

Results and Discussion

We chose 3-[3-(6-azauracil-5-yl)-2-aminophenyl]-1,2-dihydro-quinoxaline-2-one 1 as a starting material. Its preparation was described in one of our previous works. After diazotisation, the compound appeared to be a very suitable synthon for the preparation of heterocyclic N-H acids 2-14.
Although the amino group of the compound 1 was placed between two large substituents, we found out that its diazotisation, giving the diazonium salt 2, proceeded smoothly.

Despite all expectations, there were not any complications, even when the diazonium salt 2 was further converted to its derivatives 3-10 as well as when the compounds 9 and 10 were converted to compounds 11 and 12 respectively with larger substituents placed among the rings.

There must be taken into the consideration that the simple nucleophilic attack of the electron pair of carbonyl oxygen atom to carbenium ion can predominantly cause the formation of benzofuro- derivative 13 instead of the required products. This compound can be obtained by slow decomposition of diazonium salt 2 at the low temperature. When the reaction temperature is raised to boiling point, the compound that is formed is a corresponding disubstituted phenol 4 instead of derivative 13. Lower affinity of 1,2-dihydro-quinoline-2-one carbonyl group to nucleophilic attack and easy formation of benzofuroderivatives confirming this reaction course is in the accordance to the previous results.5 There were not found any problems with the nucleophilic substitution by either iodide anion, giving iodo derivative 7, or with Sandmeyer reaction, leading to corresponding chloro derivative 5 and bromo derivative 6 as well. We were successful even with the reductive cleavage of diazonium group with H2PO2, which gave 3-[3-(6-azauracil-5-yl)phenyl]-1,2-dihydro-quinoxalin-2-one 3. For the preparation of the compounds containing three heterocycles with N-H group in its molecule, we used the coupling reactions of diazonium salt 2 with suitable components such as malononitrile and ethyl cyanoacetylcarbamate. Neither these reactions were affected by sterical hindrance, so the hydrazones 9 and 10 were obtained in a good yield. Hydrazone 9 was cyclized with hydrazine to corresponding 3,5-diamino-4-aryiazopyrazole 11, while hydrazone 10 was refluxed in pyridine and thus gave a derivative with quinoxaline and two 6-azauracil cycles 12 in the molecule.

We have also focused on the preparation of disubstituted hydrazino derivative 8 with aim to prepare the cinnoline cycle via its cyclization. Reduction of diazonium salt 2 with alkaline sulphite and its subsequent acidic hydrolysis afforded desired product 8, but it was not able to isolate this product, because of its easy cyclocondensation with the carbonyl group of 6-azauracil cycle affording [1,2,4]triazino[5,6-c]cinnoline derivative 14. The possibility of isomeric compound formation on the principle of cyclocondensation of hydrazino group with quinoxaline carbonyl group was excluded due to the former finding, that there is much higher reactivity of 6-azauracil carbonyl group in comparison to the reactivity of carbonyl group of 1,2-dihydro-quinoxalin cycle.2 Binuclear compound 14, mentioned above, was also obtained by refluxing of hydrazone 10 in strong acidic media. Also during this reaction it was not possible to detect the intermediate of this hydrolysis the hydrazone 8. This fact corresponds to our former results.7-9

**Experimental Section**

**General Procedures.** All melting points were determined with Boetius block and are uncorrected. 1H-NMR spectra (300 MHz) were recorded on Bruker Avance 300 spectrometer.
Chemical shifts (δ) are given from TMS (0 ppm) as internal standard. Mass spectra were measured on LCQ ion trap mass spectrometer (Finnigan MAT, San Jose, CA, USA). IR spectra were recorded on ATI Unicam Genesis FTIR with diffuse reflectance as the instrumental technique. Elemental analyses were measured on EA 1108 Elemental Analyzer (Fison Instrument). The purities of compounds were confirmed on HPLC analyzer Beckmann System Gold (USA) with column Merck Lichrospher RP-select B 250 ′ 3 mm, 5 µm on reverse phase C-18, gradient H2O/methanol.

3-[3-(3,5-Dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6-yl)-phenyl]-2,3-dihydro-quinoxalin-2-one (3)

Compound 1^2 (530 mg, 1.50 mmol) was dissolved in 1% aqueous sodium hydroxide (60 mL) and sodium nitrite (196 mg, 2.83 mmol) was added to this solution. The mixture was cooled down to 0°C and slowly added dropwise to a solution of concentrated sulfuric acid (8 mL) and water (8 mL). After the addition, the whole reaction mixture was stirred on ice bath for 30 minutes. Insoluble part was filtered out and 50% phosphorous acid (7 mL, 10.6 mmol) was slowly added into the filtrate. The mixture was stirred for 2 hours on ice bath and then put into the refrigerator for 1 day. The next day the resulting yellow solid was filtered off, washed with water and dried. Recrystallization from cellosolve afforded 361 mg (71%) of pure yellow crystalline product; mp >360 °C; IR: NH 3375, 3234, Ar-H 3040, CO 1741, 1716, 1658 cm⁻¹; \(^1\)H-NMR (300 MHz, DMSO) δ 7.35 (m, 2H, Ar-H), 7.54 (m, 2H, Ar-H), 7.82 (d, 1H, Ar-H, \(J = 9\) Hz), 8.05 (d, 1H, Ar-H, \(J = 9\) Hz), 8.22 (s, 1H, Ar-H), 8.39 (d, 1H, Ar-H, \(J = 9\) Hz), 12.07 (s, 1H, N-H), 12.53 (s, 1H, N-H), 12.68 (s, 1H, N-H); MS: m/z 334[M+H]⁺; Anal calcd. for C_{17}H_{11}N_{5}O_{3} (333.31) C, 61.26; H, 3.33; N, 21.01; found: C, 61.53; H, 3.25; N, 21.35.

3-[2-Hydroxy-3-(3,5-dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6-yl)-phenyl]-2,3-dihydro-quinoxalin-2-one (4)

Compound 1 (205 mg, 0.59 mmol) was dissolved in 1% aqueous sodium hydroxide (60 mL). Sodium nitrite (102 mg, 1.48 mmol) was added to this solution. The mixture was cooled down to 0°C and slowly added to a solution of concentrated sulfuric acid (8 mL) and water (8 mL). After the addition, the whole reaction mixture was stirred on ice bath for 30 minutes. Insoluble part was filtered out and the filtrate was first heated to room temperature and then refluxed for the next 30 minutes. The resulting orange solid was filtered off, washed with water and dried to yield 131 mg (64%); mp >360 °C; IR: NH 3243 3131, Ar-H 3058, OH 2620, CO 1723, 1713, 1666 cm⁻¹; \(^1\)H-NMR (300 MHz, DMSO) δ 7.37 (m, 2H, Ar-H), 7.57 (m, 2H, Ar-H), 7.82 (d, 1H, Ar-H, \(J = 9\) Hz), 8.05 (d, 1H, Ar-H, \(J = 9\) Hz), 8.39 (d, 1H, Ar-H, \(J = 9\) Hz), 8.78 (b, 1H, O-H), 12.11 (s, 1H, N-H), 12.55 (s, 1H, N-H), 12.60 (s, 1H, N-H); MS: m/z 350[M+H]⁺; Anal calcd. for C_{17}H_{11}N_{5}O_{4} (349.31) C, 61.26; H, 3.33; N, 20.05; found: C, 61.24; H, 3.05; N, 19.75.

3-[2-Chloro-3-(3,5-dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6-yl)-phenyl]-2,3-dihydro-quinoxalin-2-one (5)
Compound 1 (202 mg, 0.58 mmol) was dissolved in 1% aqueous sodium hydroxide (60 mL). Sodium nitrite (101 mg, 1.68 mmol) was added to this solution. The mixture was cooled down to 0°C and slowly added to a solution of concentrated hydrochloric acid (8 mL) and water (8 mL). After the addition, the whole reaction mixture was stirred on an ice bath for 30 minutes. The insoluble part was filtered out and freshly prepared solution of CuCl (100 mg, 1.11 mmol) in concentrated hydrochloric (5 mL) acid was slowly added to the filtrate. The mixture was stirred for 2 hours on ice bath and then put to refrigerator for 1 day. The next day the yellow solid was filtered off, washed with water and dried. Recrystallization from ethanol afforded 121 mg (57%) of pure product; mp >360 °C; IR: NH 3598, 3211, Ar-H 3099, CO 1726, 1679, 1656 cm⁻¹; ¹H-NMR (300 MHz, DMSO) δ 7.30 (t, 1H, Ar-H, J= 9 Hz), 7.53 (m, 4H, Ar-H), 8.40 (d, 1H, Ar-H, J= 9 Hz), 9.64 (d, 1H, Ar-H, J= 9 Hz), 12.04 (s, 1H, N-H), 12.14 (s, 1H, N-H), 12.51 (s, 1H, N-H); MS: m/z 368[M+H]⁺; Anal calcd. for C₁₇H₁₁N₅O₃Cl (367.75) C, 55.52; H, 2.74; N, 19.04. found: C, 55.18; H, 2.69; N, 19.01.

3-[2-Bromo-3-(3,5-dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6-yl)-phenyl]-2,3-dihydro-quinoxalin-2-one (6)
Compound 1 (100 mg, 0.29 mmol) was dissolved in 1.5% aqueous sodium hydroxide (30 mL) and sodium nitrite (50 mg, 0.83 mmol) was added to this solution. The mixture was cooled down to 0°C and slowly added to a solution of concentrated hydrobromic acid (20 mL). After the addition, the whole reaction mixture was stirred on ice bath for 30 minutes. Insoluble part was filtered out and freshly prepared solution of CuBr (500 mg, 3.43 mmol) in concentrated hydrobromic acid (10 mL) was slowly added to the filtrate. The mixture was stirred for 2 hours on ice bath and then put to refrigerator for 1 day. The next day the gray solid was filtered off, washed with water and dried. Recrystallization from ethanol afforded 94 mg (80%) of pure product; mp >360 °C; IR: NH 3336, 3219, Ar-H 3058, CO 1747, 1665, 1639 cm⁻¹; ¹H-NMR (300 MHz, DMSO) δ 7.35 (m, 2H, Ar-H), 7.59 (m, 4H, Ar-H), 7.80 (d, 1H, Ar-H, J= 9 Hz), 12.21 (s, 1H, N-H), 12.57 (s, 1H, N-H), 12.64 (s, 1H, N-H); MS: m/z 412, 414[M+H]⁺; Anal calcd. for C₁₇H₁₁N₅O₃Br (412.20) C, 49.54; H, 2.45; N, 16.99; found: C, 49.23; H, 2.38; N, 16.72.

3-[2-Iodo-3-(3,5-dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6-yl)-phenyl]-2,3-dihydro-quinoxalin-2-one (7)
Compound 1 (109 mg, 0.31 mmol) was dissolved in 60 mL of 1% aqueous sodium hydroxide and sodium nitrite (56 mg, 0.83 mmol) was added to this solution. The mixture was cooled down to 0°C and slowly added to a solution of concentrated hydrochloric acid (8 mL) and water (8 mL). After the addition, the whole mixture was stirred on ice bath for 30 minutes. The insoluble part was filtered out and mildly crushed potassium iodide (1.00 g, 6.0 mmol) was slowly added to the filtrate. The mixture was stirred for 5 hours on ice bath and then put to refrigerator for 1 day. The next day the orange solid was filtered off, washed with water and dried. Recrystallization from ethanol afforded 135 mg (94%) of pure product; mp >360 °C; IR: NH
3343, 3206, Ar-H 3066, CO 1762, 1678, 1645 cm\(^{-1}\); \(^1\)H-NMR (300 MHz, DMSO): \(\delta\) 7.45 (t, 1H, Ar-H, \(J = 6\) Hz), 7.55 (d, 1H, Ar-H, \(J = 6\) Hz), 7.73 (t, 1H, Ar-H, \(J = 6\) Hz), 7.93 (d, 1H, Ar-H, \(J = 6\) Hz), 8.38 (d, 1H, Ar-H, \(J = 6\) Hz), 8.49 (t, 1H, Ar-H, \(J = 6\) Hz), 8.71 (d, 1H, Ar-H, \(J = 6\) Hz), 12.57 (s, 1H, N-H), 12.60 (s, 1H, N-H), 13.27 (s, 1H, N-H); MS: m/z 460\([M+H]^+\); Anal calcd. for C\(_{17}\)H\(_{10}\)N\(_{5}\)O\(_3\)Br (459.21) C, 44.47; H, 2.20; N, 15.25; found: C, 44.27; H, 2.03; N, 14.88.

\([2-(3-Oxo-3,4-dihydro-quinoxalin-2-yl)-6-(3,5-dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6yl)]-phenylhydrazono-malononitrile (9)\)

Compound 1 (346 mg, 0.99 mmol) was dissolved in 10% aqueous sodium hydroxide (10 mL) and sodium nitrite (125 mg, 2.09 mmol) was added to this solution. The mixture was cooled down to 0\(^\circ\)C and slowly added to a solution of concentrated sulfuric acid (8 mL) and water (8 mL). After the addition, the whole reaction mixture was stirred on ice bath for 30 minutes. The insoluble part was filtered out and the filtrate was slowly poured into stirred copulation mixture of sodium acetate trihydrate (40.33 g, 0.30 mol) and malononitrile (360 mg, 5.45 mmol) in water (400 mL). The mixture was stirred for 2 hours on ice bath and then put to refrigerator for 1 day. The next day the yellow solid was filtered off, washed with water and dried. Recrystallization from ethanol afforded 371 mg (88%) of pure product; mp >360 \(^\circ\)C; IR: NH 3320, 3180, Ar-H 3059, CN 2228, CO 1707, 1687, 1664 cm\(^{-1}\); \(^1\)H-NMR (300 MHz, DMSO): \(\delta\) 7.33 (m, 2H, Ar-H), 7.54 (m, 3H, Ar-H), 7.80 (d, 1H, Ar-H, \(J = 6\) Hz), 7.86 (d, 1H, Ar-H, \(J = 6\) Hz), 12.15 (s, 1H, N-H), 12.47 (s, 1H, N-H), 12.73 (s, 1H, N-H); MS: m/z 426\([M+H]^+\); Anal calcd. for C\(_{17}\)H\(_{10}\)N\(_{5}\)O\(_3\) (425.37) Anal calcd. for C, 56.47; H, 2.61; N, 29.64; found: C, 56.31; H, 2.68; N, 29.48.

\(\text{Ethyl N-\{2-cyano-2-[2-(3-Oxo-3,4-dihydro-quinoxalin-2-yl)-6-(3,5-dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6yl)]-phenylhydrazono-acetyl\}-carbamate (10)\)\)

Compound 1 (511 mg, 1.47 mmol) was dissolved in 10% aqueous sodium hydroxide (10 mL) and sodium nitrite (185 mg, 3.09 mmol) was added to this solution. The mixture was cooled down to 0\(^\circ\)C and slowly added to a solution of concentrated sulfuric acid (8 mL) and water (8 mL). After the addition, the whole reaction mixture was stirred on ice bath for 30 minutes. The insoluble part was filtered off and the filtrate was slowly poured into stirred mixture of sodium acetate trihydrate (40.00 g, 0.29 mol) and ethyl cyanoacetylcarbamate (686 mg, 4.39 mmol) in water (1200 mL). The mixture was stirred for 2 hours on ice bath and then put into refrigerator for 1 day. The next day the yellow solid was filtered off, washed with water and dried. Recrystallization from ethanol afforded 529 mg (70%) of pure product; mp >360 \(^\circ\)C; IR: Ar-H 3044, CN 2230, CO 1777, 1725, 1703, 1655 cm\(^{-1}\); \(^1\)H-NMR (300 MHz, DMSO): \(\delta\) 1.13 (t, 3H, CH\(_3\)), 3.82 (q, 2H, CH\(_2\)), 7.33 (m, 2H, Ar-H), 7.54 (m, 3H, Ar-H), 7.80 (d, 1H, Ar-H, \(J = 6\) Hz), 7.86 (d, 1H, Ar-H, \(J = 6\) Hz), 12.15 (s, 1H, N-H), 12.47 (s, 1H, N-H), 12.73 (s, 1H, N-H); MS: m/z 516\([M+H]^+\); Anal calcd. for C\(_{23}\)H\(_{17}\)N\(_9\)O\(_6\) (515.45) C, 53.60; H, 3.32; N, 24.46; found: C, 53.44; H, 3.39; N, 24.71.
3-{2-[3,5-Diaminopyrazol-4-yl]azo}-3-{(3,5-dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6yl)-phenyl]}-2,3-dihydro-quinoxalin-2-one (11)

Compound 9 (101 mg, 0.24 mmol) was suspended in methanol (10 mL) and 98% hydrazine hydrate (2 µl, 0.47 mmol) was added to this mixture. The solution was refluxed for 20 minutes and then cooled down to room temperature and neutralized with acetic acid. The resulting orange solid was dissolved in acetic acid with active charcoal. The filtrate was evaporated to dryness and the solid suspended in water, filtered off, washed with water and dried to afford 103 mg (95%) of pure product; mp >360 °C; IR: N H 3279, Ar-H 3058, CO 1719, 1676 cm⁻¹; ¹H-NMR(300 MHz,DMSO): δ 4.10 (b, 1H, N-H), 4.71 (b, 2H, NH₂), 6.07 (b, 2H, NH₂), 7.30 (m, 2H, Ar-H), 7.38 (t, 1H, Ar-H, J= 9 Hz), 7.49 (m, 3H, Ar-H), 7.75 (d, 1H, Ar-H, J= 9 Hz), 10.82 (s, 1H, N-H), 11.98 (s, 1H, N-H), 12.32 (s, 1H, N-H); MS: m/z 458[M+H]+; Anal calcd. for C₂₀H₁₅N₁₁O₃ (457.41) C, 52.52; H, 3.31; N, 33.68; found: C, 52.29; H, 3.36; N, 33.94.

3-{2-(3,5-Dioxo-6-cyano-2,3,4,5-tetrahydro-[1,2,4]triazin-2-yl)-3-(3,5-dioxo-2,3,4,5-tetrahydro-[1,2,4]triazin-6-yl)-phenyl]}-2,3-dihydro-quinoxalin-2-one (12)

Compound 10 (99 mg, 0.19 mmol) was dissolved in pyridine (5 mL) and refluxed for 6 hours. Then the mixture was let to stand for one day and consequently the pyridine was evaporated on rotary evaporator. The resulting yellow solid was mixed with 10% aqueous solution of sodium bicarbonate (10 mL). This solution was acidified with concentrated hydrochloric acid to obtain yellow precipitate that was filtered off, washed with water and dried. Recrystallization from N,N-dimethylformamide afforded 51 mg (57%) of yellow crystalline product; mp >360 °C; IR: NH 3211, Ar-H 3010, CN 2252 CO 1720, 1667 cm⁻¹; ¹H-NMR(300 MHz,DMSO): δ 7.34 (t, 2H, Ar-H, J= 6 Hz), 7.61 (m, 2H, Ar-H), 7.79 (m, 2H, Ar-H), 8.08 (d, 1H, Ar-H, J= 6 Hz), 12.17 (s, 1H, N-H), 12.48 (s, 1H, N-H), 12.70 (s, 1H, N-H), 13.12 (s, 1H, N-H); MS: m/z 458[M+H]+; Anal calcd. for C₂₁H₁₁N₉O₅ (469.38) C, 53.74; H, 2.36; N, 26.86; found: C, 53.29; H, 2.37; N, 26.95.

6-{[1]Benzofuro[2,3-b]quinoxalin-4-yl]}-2,3,4,5-tetrahydro[1,2,4]triazin-3,5-dion (13)

Compound 1 (101 mg, 0.29 mmol) was dissolved in 1% aqueous sodium hydroxide (60 mL) and sodium nitrite (59 mg, 0.98 mmol) was added to this solution. The mixture was cooled down to 0°C and slowly added to a solution of concentrated sulfuric acid (8 mL) and water (8 mL). After the addition, the whole mixture was stirred on ice bath for 30 minutes. The insoluble part was filtered out and the filtrate was left to stand in refrigerator for one day. The resulting yellow precipitate was filtered off, washed with water and dried to afford 61 mg (63%) of pure crystalline product; mp >360 °C; IR: Ar-H 3044, CO 1720, 1689 cm⁻¹; ¹H-NMR(300 MHz,DMSO): δ 7.39 (m, 2H, Ar-H), 7.62 (m, 2H, Ar-H), 7.87 (d, 1H, Ar-H, J= 9 Hz), 8.12 (d, 1H, Ar-H, J= 9 Hz), 8.41 (d, 1H, Ar-H, J= 9 Hz), 12.53 (s, 1H, N-H), 12.62 (s, 1H, N-H); MS: m/z 332[M+H]+; Anal calcd. for C₁₇H₉N₅O₃ (331.29) C, 61.63; H, 2.74; N, 21.14; found: C, 61.43; H, 2.75; N, 21.09.
Compound 10 (200 mg, 0.39 mmol) was dissolved in the mixture of concentrated hydrochloric acid (20 mL) and water (10 mL). The solution was refluxed for 3 hours. The resulting bright gray precipitate was filtered off, washed with water and dried. Recrystallization from cellosolve afforded 100 mg (75%) of pure product; mp >360 °C; IR: NH 3167, Ar-H 3078, CO 1724, 1680 cm⁻¹; ¹H-NMR(300 MHz,DMSO): δ 7.31 (m, 2H, Ar-H), 7.70 (t, 1H, Ar-H, J= 6 Hz), 8.03 (dd, 2H, Ar-H, J= 6 Hz), 8.12 (d, 1H, Ar-H, J= 6 Hz), 8.40 (d, 1H, Ar-H, J= 6 Hz), 9.15 (s, 1H, N-H), 11.67 (s, 1H, N-H), 12.24 (s, 1H, N-H), 12.67 (s, 1H, N-H); MS: m/z 346[M+H]+; Anal calcd. for C₁₇H₁₁N₇O₂ (345.32) C, 59.13; H, 3.21; N, 28.39; found: C, 59.42; H, 3.18; N, 27.99.

Acknowledgements

We are grateful to the Ministry of Education, Youth and Sport of the Czech Republic, for grant MSM619859216.

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1. For previous paper see: Fryšová, I.; Slouka, J.; Gucký, T. ARKIVOC 2005, (xv), 30.