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Alkylation of Pyrimidine Derivatives with Ethylene Chlorohydrin

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Abstract—Alkylation of 6-methyluracil, 5-fluorouracil, uracil-5-ammonium sulfate, 5-hydroxyuracil, 5-hydroxy-6-methyluracil, 3,6-dimethyl-5-hydroxyuracil, and 1,3,6-trimethyl-5-hydroxyuracil with ethylene chlorohydrin in water-alcohol medium in the presence of KOH was investigated.

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Alkylation of uracil and 6-methyluracil with ethylene chlorohydrin was reported in [1]. It was demonstrated that the reaction gave rise either to O- or N-alkylated products or to a mixture of both. Uracil reaction with esters in DMSO in the presence of K_2CO_3 provided mixtures of N-substituted uracils [2]. In the study of uracil reaction with chlorohydrins in water in the presence of NaOH also only the products of N-alkylation were obtained [3].

In extension of the search for new pyrimidine derivatives with possible immunotropic, antiphlogistic, and antiradical action we investigated alkylation of 6-methyluracil (**I**), 5-fluorouracil (**II**), uracil-5-ammonium sulfate (**III**), 5-hydroxyuracil (**IV**), 5-hydroxy-6-methyluracil (**V**), 3,6-dimethyl-5-hydroxyuracil (**VI**), and 1,3,6-trimethyl-5-hydroxyuracil (**VII**) with ethylene chlorohydrin (**VIII**) in a water-alcohol medium in the presence of KOH.

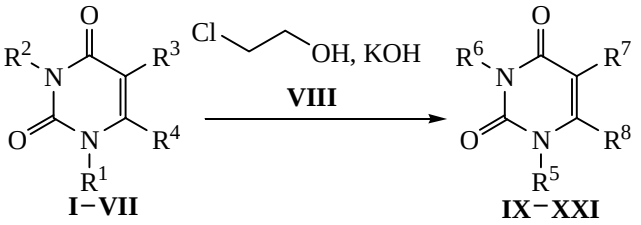
Alkylation of 6-methyluracila (**I**) with ethylene chlorohydrin (**VIII**) in a water-alcohol medium in the presence of KOH provided 3-(2-hydroxyethyl)-6-methyl-uracil (**IX**) and 1,3-bis(2-hydroxyethyl)-6-methyluracil (**X**) that were isolated in a pure state by recrystallization from alcohol in 10 and 50% yield respectively. *N*-(Hydroxy-ethyl)-6-methyluracils obtained are crystalline substances well soluble in water and moderately soluble in alcohol. Their structure was proved by comparison with the published melting point [2], and also by UV spectra.

Alkylation of 5-fluorouracil (**II**) with ethylene chlorohydrin led to the formation of 1,3-bis(2-hydroxy-ethyl)-5-fluorouracil (**XI**) in 97% yield as a thick light-yellow fluid of R_f 0.77, and of crystalline 3-(2-hydroxy-ethyl)-5- fluorouracil (**XII**) in 3% yield. By alkylation of uracil-5-ammonium sulfate (**III**) 1,3-bis(2-hydroxyethyl)-uracil-5-ammonium sulfate (**XIII**) was obtained, whose hydrolysis with H_2SO_4 resulted in 1,3-bis(2-hydroxyethyl)-5-hydroxyuracil (**XIV**). To elucidate the direction of alkylation either at N^1 or N^3 atoms we measured the UV spectra of compounds **IX** and **XII** at pH variation. It was established that when pH of the medium changed from 1 to 12 the absorption maximum suffered a red shift of 18–23 nm characteristic of uracil substituted at N^3 of the ring [4].

To distinguish the alkylation direction in the uracil derivatives in position *1* or *3* we also used the ^{1}H NMR spectra. Thus with a substituent in position *3* the signal of N^1H proton appears in the region 8–9 ppm, whereas with a substituent in position *1* the resonance of N^3H proton is observed in the region 10–11 ppm.

In reaction of 5-hydroxyuracil (**IV**) and 5-hydroxy-6-methyluracil (**V**) with ethylene chlorohydrin the alkylation was found to occur simultaneously in all three positions *1*, *3*, and *5*. On alkylating 5-hydroxyuracil (**IV**) with ethylene chlorohydrin we obtained a mixture whose repeated recrystallization permitted isolation of 1,3-bis(2-hydroxyethyl)-5-(2-hydroxyethoxy)uracil (**XV**) in 77%

yield and 1,3-bis(2-hydroxyethyl)-5-hydroxyuracil (**XIV**). Alkylation of 5-hydroxy-6-methyluracil (**V**) resulted in a mixture of compounds that was subjected to repeated recrystallization to isolate in pure state 1,3-bis(2-hydroxyethyl)-5-(2-hydroxyethoxy)-6-methyluracil (**XVI**) in 50% yield (mp 136-–138°C), well soluble in water, alcohol, and chloroform; 1,3-bis(2-hydroxyethyl)-5-hydroxy-6-methyluracil (**XVII**) in 20% yield (mp 227–230°C); and 3-(2-hydroxyethyl)-5-(2-hydroxyethoxy)-6-methyluracil (**XVIII)** in 10% yield as a thick fluid.



I–**VII** **IX**–**XXI**

$R^1 = R^2 = R^3 = H$, $R^4 = Me$ (**I**); $R^1 = R^2 = R^4 = H$, $R^3 = F$ (**II**), OSO_3NH_4 (**III**), OH (**IV**); $R^1 = R^2 = H$, $R^3 = OH$, $R^4 = Me$ (**V**); $R^1 = H$, $R^2 = R^4 = Me$, $R^3 = OH$ (**VI**); $R^1 = R^2 = R^4 = Me$, $R^3 = OH$ (**VII**); $R^5 = R^7 = H$, $R^6 = CH_2CH_2OH$, $R^8 = Me$ (**IX**); $R^5 = R^6 = CH_2CH_2OH$, $R^7 = H$, $R^8 = Me$ (**X**), $R^7 = F$, $R^8 = H$ (**XI**); $R^5 = R^8 = H$, $R^6 = CH_2CH_2OH$, $R^7 = F$ (**XII**); $R^5 = R^6 = CH_2CH_2OH$, $R^8 = H$, $R^7 = OSO_3NH_4$ (**XIII**), OH (**XIV**), OCH_2CH_2OH (**XV**); $R^5 = R^6 = CH_2CH_2OH$, $R^8 = Me$, $R^7 = OCH_2CH_2OH$ (**XVI**), OH (**XVII**); $R^5 = H$, $R^6 = CH_2CH_2OH$, $R^7 = OCH_2CH_2OH$, $R^8 = Me$ (**XVIII**); $R^5 = CH_2CH_2OH$, $R^6 = R^8 = Me$, $R^7 = OCH_2CH_2OH$ (**XIX**), OH (**XX**); $R^5 = R^6 = R^8 = Me$, $R^7 = OCH_2CH_2OH$ (**XXI**).

On alkylating 3,6-dimethyl-5-hydroxyuracil (**VI**) with ethylene chlorohydrin under these conditions we obtained 1-hydroxyethyl-5-(2-hydroxyethoxy)-3,6-dimethyluracil (**XIX**) and 1-hydroxyethyl-5-hydroxy-3,6-dimethyluracil (**XX**). The alkylation of 1,3,6-trimethyl-5-hydroxyuracil (**VII**) with ethylene chlorohydrin gave 1,3,6-trimethyl-5-(2-hydroxyethoxy)uracil (**XXI**) in 80% yield, mp 280°C, R_f 0.8.

The structure of compounds obtained was proved by IR, UV, ^{1}H and ^{13}C NMR spectra, and also by the elemental analyses.

In the IR spectra of all compounds absorption bands were present in the region 1620–1720 cm^{-1} characteristic of vibrations of the pyrimidine fragment [ν(C–O, NC–O)]. The absorption bands in the region 1060–1240 cm^{-1} are common in spectra of compounds whose molecules contain a tertiary nitrogen; absorption bands in the region 3300–3500 cm^{-1} are due to the stretching vibrations of the O–H and N–H bonds. In the spectra of all compounds the absorption band of a hydroxy group involved in a hydrogen bond was observed at 3400 cm^{-1}.

In the ^{13}C NMR spectra of all compounds the carbon atoms of the uracil fragment appeared as usual signals at 164 (C^4), 157 (C^2), 153 (C^6) ppm, also signals were observed in the region 13–14 (CH_3C^6), and 98–102 (C^5) ppm.

EXPERIMENTAL

UV spectra of solutions with concentration 10^{-5}% were measured on a spectrometer Specord M-400 in the range 200–350 nm in a cell 19 mm thick. IR spectra were recorded from mulls of compounds in mineral oil or from liquid films on a spectrophotometer UR-20 with prisms of NaCl and LiF. ^{1}H and ^{13}C NMR spectra were registered from 1% (^{1}H) or 10–20% (^{13}C) solutions in D_2O on a spectrometer Bruker AM (at 300 and 75 MHz respectively), internal reference HMDS. Melting points were measured on a Boëtius heating block. Elemental analyses were carried out on CHN Analyzer M-185B. The monitoring of reaction progress and checking the homogeneity of compounds obtained was performed by TLC on Silufol UV-254 plates, eluent ethanol–ammonia, 4:1, development under UV irradiation or in iodine vapor.

3-(2-Hydroxyethyl)-6-methyluracil (IX) and 1,3-bis(2-hydroxyethyl)-6-methyluracil (X). To 126 g (0.1 mol) of 6-methyluracil was poured 800 ml of water and 100 ml of ethanol, 2 mol of 82% powdered KOH was added, and within 30 min at room temperature was added 200 g (2.5 mol) of ethylene chlorohydrin. The reaction mixture was gradually brought to boiling by heating on a water bath, and it was stirred for 4–6 h till the pH of the mixture turned neutral or slightly acidic (pH 6–7). Then water was distilled off from the reaction mixture under a vacuum of an oil pump, the separated crystals were filtered off, washed with hot acetone, acetone and the mother liquor were combined, and acetone was distilled off. The thick mass obtained (164 g) was dissolved in 300 ml of 2-propanol, and the product was precipitated from the solution by hexane (400 ml). The underlayer was separated, on distilling off the solvent and azeotropic drying we obtained 100 g (50%) of compound (**X**), mp 110–112°C (111–112°C [1]). IR spectrum, ν, cm^{-1}: 800, 1060–1280 (=N–), 1370 [δ_s (CH_3)], 1470 (CH_2, CH_3), 1600, 1650, 1690 (C=O, =NC=O), 2860, 2940 (CH), 3500 (OH). ^{1}H NMR spectrum, δ, ppm: 2.07 s (3H, CH_3), 3.61 t (2H, C^8H_2, J 5.8 Hz), 3.68 t (2H, C^7H_2, J 5.8 Hz), 3.73 d (2H, $C^{10}H_2$, J 5.8 Hz), 3.9 t (2H, C^9H_2, J 5.8 Hz), 4.8 s (2H, OH).

^{13}C NMR spectrum, δ, ppm: 22.27 (CH_3), 45.61 (C^9), 49.79 (C^7), 61.19 (C^8), 61.61 (C^{10}), 103.11 (C^5), 155.0 (C^6), 158.25 (C^2), 167.45 (C^4). Found, %: C 50.43; H 6.34; N 13.8. $C_9H_{14}N_2O_4$. Calculated, %: C 50.46; H 6.59; N 13.08. From the upper organic layer the solvents were distilled off, the residue was dissolved in alcohol, the precipitated crystals were filtered off and dried. We obtained 17 g (10%) of compound **IX**, mp 205–206°C (205– 206°C [1]). IR spectrum, ν, cm^{-1}: 790, 880, 1080–1260 (=N–), 1360, 1390 [δ_s (CH_3)], 1480 (CH_2, CH_3), 1600, 1650, 1710 (C=O, NC=O), 2870, 2950 (CH), 3100 (NH). Found, %: C 49.96; H 6.16; N 16.00. $C_7H_{10}N_2O_3$. Calculated, %: C 49.40; H 5.92; N 16.46.

1,3-Bis(2-hydroxyethyl)-5-fluorouracil (XI) and 3-(2-hydroxyethyl)-5-fluorouracil (XII) were prepared analogously to compound **X** from 6.51 g (0.065 mol) of 5-fluorouracil, 6.8 g of KOH in 40 ml of water and 20 ml of alcohol, and of 10.6 g (0.125 mol) of ethylene chlorohydrin. We obtained 11.6 g of thick light-brown fluid which was dissolved in 30 ml of acetone, The precipitated crystals were filtered off to obtain 1.2 g (3%) of compound **XII**, mp 138–140°C. UV spectrum (ethanol): λ_{min} 243, λ_{max}277 nm. IR spectrum, ν, cm^{-1}: 800, 1072–1270 (=N–), 1384 [δ_s (CH_3)], 1460 (CH_2, CH_3), 1654, 1678, 1702 (C=O, NC=O), 2920, 2998 (CH), 3500 (OH). ^{1}H NMR spectrum, δ, ppm: 3.75 t (2H, $C^{10}H_2$, *J* 5.8 Hz), 3.96 t (2H, C^9H_2, *J* 5.8 Hz), 5.04 (1H, OH), 7.56 d (1H, C^6H, J_{HF} 8 Hz), 8.39 s (1H, N^1H). ^{13}C NMR spectrum, δ, ppm: 42.74 (C^9), 60.30 (C^{10}), 125.75 (C^5, J_{CF} 251 Hz), 142.60 (C^6, J_{CF} 20 Hz), 152.50 (C^2), 162.45 (C^4, J_{CF} 20 Hz). Found, %: C 41.00; H 4.40; F 10.5; N 14.60. $C_6H_7FN_2O_3$. Calculated, %: C 41.38; H 4.05; F 10.91; N 15.08. On removing acetone from the filtrate we obtained 10 g (97%) of compound **XI** as a thick light-yellow fluid, R_f 0.77. UV spectrum (ethanol): λ_{min} 247.20, λ_{max}278 nm. IR spectrum, ν, cm^{-1}: 784, 820, 850, 880, 922, 952, 1018– 1270 (=N–), 1384 [δ_s (CH_3)], 1460 (CH_2, CH_3), 1635, 1654, 1684, 1714 (C=O, NC=O), 2890, 2932, 2968, 2998 (CH), 3334, 3376, 3470 (OH). ^{1}H NMR spectrum, δ, ppm: 3.72 t (6H, C^7H_2, C^8H_2, $C^{10}H_2$, *J* 5.8 Hz), 3.99 t (2H, C^9H_2, *J* 5.8 Hz), 5.15 (2H, OH), 7.53 d (1H, C^6H, J_{HF} 8 Hz). ^{13}C NMR spectrum, δ, ppm: 43.34 (C^9), 54.39 (C^7), 59.50 (C^8), 60.31 (C^{10}), 130.54 (C^5, J_{CF} 251 Hz), 140.30 (C^6, J_{CF} 20 Hz), 150.50 (C^2), 161.40 (C^4, J_{CF} 20 Hz). Found, %: C 43.52; H 5.50; F 8.30; N 11.85. $C_8H_{11}FN_2O_4$. Calculated, %: C 44.00; H 5.05; F 8.70; N 12.13.

1,3-Bis(2-hydroxyethyl)-5-(2-hydroxyethoxy)-6-methyluracil (XVI), 1,3-bis(2-hydroxyethyl)-5-hydroxy-6-methyluracil (XVII), and 3-(2-hydroxyethyl)-5-(2-hydroxyethoxy)-6-methyluracil (XVIII). To 127.8 g (0.9 mol) of 5-hydroxy-6-methyluracil was powred 800 ml of water, 174 g (2.85 mol) of powdered KOH was added, and within 20 min was added 230 g (2.88 mol) of ethylene chlorohydrin (the process occurred with heat evolution). The reaction mixture was brought to boiling by heating on a water bath and stirred for 4–6 h. On cooling the reaction mixture was acidified to pH 6–7 with 1–2 ml of concn. HCl, water was distilled off under a reduced pressure (50–100 mm Hg) at the bath temperature 80°C. The precipitated crystals were filtered off, washed with hot acetone to get in residue KCl. Acetone was distilled off from the mother liquor, the residue crystallized on standing. 165 g of crystals separated by filtration were heated in 350 ml of ethanol till they dissolved, the insoluble residue was filtered off through a glass frit. On cooling, filtration, and drying we obtained 46 g (20%) of compound **XVII**, mp 227–230°C. IR spectrum, ν, cm^{-1}: 780, 1060–1270 (=N–), 1385 [δ_s (CH_3)], 1460 (CH_2, CH_3), 1600, 1650, 1690, 1710 (C=O, NC=O), 2860, 2940 (CH), 3560 (OH). ^{1}H NMR spectrum, δ, ppm: 1.89 s (3H, CH_3), 3.60 t (2H, C^7H_2, *J* 5.8 Hz), 3.65 d (2H, C^8H_2, *J* 5.8 Hz), 3.73 d (2H, $C^{10}H_2$, *J* 5.8 Hz), 3.97 t (2H, C^9H_2, J 5.8 Hz), 6.8 s (3H, OH). ^{13}C NMR spectrum, δ, ppm: 12.20 (CH_3), 44.61 (C^9), 52.79 (C^7), 58.19 (C^8), 60.61 (C^{10}), 130.00 (C^5), 141.0 (C^6), 150.25 (C^2), 160.45 (C^4). Found, %: C 47.06; H 6.09; N 11.88. $C_8H_{14}N_2O_5$. Calculated, %: C 46.95; H 6.13; N 12.17. From the ethanolic mother liquor crystals precipitated on standing, they were filtered off and dried to obtain 138.4 g (50%) of compound **XVI**, mp 136–138°C. IR spectrum, ν, cm^{-1}: 780, 1050–1280 (=N–), 1379 [δ_s (CH_3)], 1470 (CH_2, CH_3), 1600, 1650, 1690, 1710 (C=O, NC=O), 2860, 2960 (CH), 3600 (OH). ^{1}H NMR spectrum, δ, ppm: 1.86 s (3H, CH_3), 3.59 t (2H, C^7H_2, *J* 5.8 Hz), 3.65 d (2H, C^8H_2, *J* 5.8 Hz), 3.73 d (2H, $C^{10}H_2$, *J* 5.8 Hz), 3.91 d (2H, $C^{11}H_2$, *J* 5.8 Hz), 3.97 t (2H, C^9H_2, *J* 5.8 Hz), 4.0 d (2H, $C^{12}H_2$, *J* 5.8 Hz), 4.96 s (3H, OH). ^{13}C NMR spectrum, δ, ppm: 16.00 (CH_3), 45.60 (C^9), 52.79 (C^7), 58.19 (C^8), 60.60 (C^{10}), 63.00 (C^{12}), 65.00 (C^{11}), 129.11 (C^5), 142.45 (C^6), 149.00 (C^2), 161.85 (C^4). Found, %: C 48.45; H 6.61; N 10.77. $C_{11}H_{19}N_2O_6$. Calculated, %: C 48.17; H 6.61; N 10.21. The residues of the mother liquors in acetone and ethanol were combined , the solvents were distilled off to obtain 18.6 g (10%) of compound **XVIII** as a thick fluid, R_f 0.76. IR spectrum, ν, cm^{-1}: 790, 880, 1080–1260 (=N–), 1360, 1390 [δ_s (CH_3)], 1480 (CH_2, CH_3), 1600, 1650, 1710 (C=O, NC=O), 2870, 2950 (CH), 3100 (NH), 3490

(OH). ^{1}H NMR spectrum, δ, ppm: 2.01 s (3H, CH_3), 3.73 d (2H, $C^{10}H_2$, *J* 5.8 Hz), 3.95 t (2H, C^9H_2, *J* 5.8 Hz), 7.87 s (2H, OH), 9.05 (1H, N^1H). ^{13}C NMR spectrum, δ, ppm: 13.66 (CH_3), 42.37 (C^9), 60.31 (C^{10}), 130.12 (C^5), 137.30 (C^6), 157.50 (C^2), 163.35 (C^4). Found, %: C 45.30; H 5.87; N 14.67. $C_7H_{10}N_2O_4$. Calculated, %: C 45.16; H 5.41; N 15.04.

1-Hydroxyethyl-5-(2-hydroxyethoxy)-3,6-dimethyluracil (XIX) and 1-hydroxyethyl-5-hydroxy-3,6-dimethyluracil (XX) were obtained in the same fashion as compound **X** from 15.2 g (0.1 mol) of 3,6-dimethyl-5-hydroxyuracil (**V**), 20.13 g (0.25 mol) of ethylene chlorohydrin, 12.3 g (0.22 mol) of KOH in 80 ml of water and 40 ml of ethanol. We obtained 14.0 g (57%) of compound **XIX** as thick light-yellow fluid, R_f 0.70. UV spectrum (ethanol): λ_{min} 247.20, λ_{max}278 nm. IR spectrum, ν, cm^{-1}: 712, 760, 778, 886, 1048, 1080–1258 (=N–), 1354, 1384 [δ_s (CH_3)], 1456 (CH_2, CH_3), 1636, 1654, 1684, 1696, 1714 (C=O, NC=O), 2870, 2950 (CH), 3100 (NH), 3490 (OH). ^{1}H NMR spectrum, δ, ppm: 1.86 s (3H, CH_3C^6), 3.42 s (3H, CH_3N^3), 3.59 t (2H, C^7H_2, *J* 5.8 Hz), 3.66 t (2H, C^8H_2, *J* 5.8 Hz), 3.90 t (2H, $C^{11}H_2$, *J* 6.9 Hz), 3.99 t (2H, $C^{12}H_2$, *J* 6.9 Hz), 4.93 s (2H, OH). ^{13}C NMR spectrum, δ, ppm: 12.66 (CH_3C^6), 28.85 (CH_3N^3), 51.40 (C^7), 58.00 (C^8), 63.00 (C^{12}), 64.60 (C^{11}), 127.12 (C^5), 142.45 (C^6), 157.50 (C^2), 162.35 (C^4). Found, %: C 48.80; H 6.60; N 11.10. $C_{10}H_{16}N_2O_5$. Calculated, %: C 49.15; H 6.60; N 11.47. We also obtained 8.8 g (44%) of compound **XX**, mp 146°C, R_f 0.81. UV spectrum (ethanol): λ_{min} 242.50, λ_{max} 276.50 nm. IR spectrum, ν, cm^{-1}: 712, 760, 778, 886, 1048, 1080–1258 (=N–), 1354, 1384 [δ_s (CH_3)], 1456 (CH_2, CH_3), 1636, 1654, 1684, 1696, 1714 (C=O, NC=O), 2870, 2950 (CH), 3100 (NH), 3490 (OH). ^{1}H NMR spectrum, δ, ppm: 1.89 s (3H, CH_3C^6), 3.43 s (3H, CH_3N^3), 3.59 t (4H, C^7H_2, C^8H_2, *J* 5.8 Hz), 4.93 s (2H, OH). ^{13}C NMR spectrum, δ, ppm: 12.22 (CH_3C^6), 28.15 (CH_3N^3), 51.40 (C^7), 58.00 (C^8), 130.40 (C^5), 142.45 (C^6), 150.50 (C^2), 162.40 (C^4). Found, %: C 47.65; H 5.90; N 13.63. $C_8H_{12}N_2O_4$. Calculated, %: C 47.99; H 6.04; N 13.99.

1,3,6-Trimethyl-5-(2-hydroxyethoxy)uracil (XXI) was obtained in a similar way as compound **XIX** from 3.0 g (0.017 mol) of 1,3,6-trimethyl-5-hydroxyuracil (**VI**), 0.85 g (0.017 mol) of KOH in 10 ml of water and 10 ml of ethanol, and of 1.5 ml (0.018 mol) of ethylene chlorohydrin. We obtained 3.0 g (82%) of compound **XXI**, mp 280°C, R_f 0.80. IR spectrum, ν, cm^{-1}: 778, 826, 850, 1072, 1080–1252 (=N–), 1348, 1390 [δ_s (CH_3)], 1456 (CH_2, CH_3), 1654, 1714 (C=O, NC=O), 2854, 2920 (CH), 3244, 3328, 3442 (OH). ^{1}H NMR spectrum, δ, ppm: 1.84 s (3H, CH_3C^6), 3.30 s (3H, CH_3N^1), 3.43 s (3H, CH_3N^3), 3.92 t (4H, $C^{11}H_2$, $C^{12}H_2$, *J* 6.9 Hz), 5.25 s (1H, OH). ^{13}C NMR spectrum, δ, ppm: 11.50 (CH_3C^6), 28.45 (CH_3N^1), 28.85 (CH_3N^3), 63.00 (C^{12}), 64.70 (C^{11}), 126.40 (C^5), 142.45 (C^6), 150.50 (C^2), 161.80 (C^4). Found, %: C 50.10; H 6.70; N 12.68. $C_9H_{14}N_2O_4$. Calculated, %: C 50.46; H 6.59; N 13.08.

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