Reactions of 2-diaminomethylidenecyclohexane-1,3-diones with diethyl malonate

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The debenzoylation of 2-[(amino)(benzoylamino)methylidene]cyclohexane-1,3-diones (addition products of cyclohexane-1,3-diones with benzoylcyanamide) affords the corresponding 2-diaminomethylidenecyclohexane-1,3-diones. The latter act as *N*,*N*-dinucleophiles in reactions with diethyl malonate in the presence of MeONa to form 6-hydroxy-2-(2,6-dioxocyclohexylidene)-1,2-dihydropyrimidin-4(3*H*)-ones. This reaction performed at 200 °C in the absence of bases results in the subsequent self-condensation of the dihydropyrimidinones to give 4,5′-bipyrimidine derivatives.

Key words: cyclohexane-1,3-diones, 2-diaminomethylidenecyclohexane-1,3-diones, diethyl malonate, 1,2-dihydropyrimidin-4(3*H*)-ones, *N*,*N*-dinucleophiles, 4,5′-bipyrimidines, condensation.

In the last two—three decades, a series of new pharmaceuticals with antitumor, antiviral, anti-inflammatory, antibacterial, *etc*. activities were developed based on pyrimidine derivatives (see the review[1]). This gave considerable impetus to the elaboration of novel procedures and the search for effective reactants for the synthesis of new functionalized pyrimidines (see, for example, Refs 2—9).

Earlier, we have described new reactants for the synthesis of pyrimidines, such as diaminomethylidene derivatives of acyclic β-diketones and β-keto esters unsubstituted at the nitrogen atoms.[10—12] For example, 2-diaminomethylidene-3-oxoalkanoic acid esters act as functionalized amino enones in reactions with aryl isocyanates and trichloroacetonitrile to give the corresponding esters of 4-aminopyrimidine-5-carboxylic acids.[11,12] However, diaminomethylidene derivatives of dibenzoylmethane and acetoacetic ester act as *N*,*N*-dinucleophiles in reactions with aroylketenes generated *in situ* from 6-aryl-2,2-dimethyl-1,3-dioxin-4-ones to form the corresponding pyrimidin-4-ones.[10]

Cyclic β-diketones and their derivatives have found wide use in the synthesis of fused pyrimidines.[13—15] In continuation of our investigations, we studied the reaction of diethyl malonate with 2-diaminomethylidenecyclohexane-1,3-diones in a purpose to synthesize new substituted pyrimidines.

Earlier,[16] we have prepared *N*-benzoylketene aminals **1a,b** by the reactions of benzoylcyanamide with cyclohexane-1,3-dione and dimedone in the presence of $Ni(OAc)_2$. However, since compounds **1a,b** contain the NH_2 group with low nucleophilicity, we have used them for the construction of heterocyclic systems only in the form of boron chelate complexes.[17—19]

In the present study, we showed that compounds **1a,b** are easily debenzoylated with MeONa in MeOH to give nitrogen-unsubstituted 2-diaminomethylidenecyclohexane-1,3-diones **2a,b** (Scheme 1). We expect that these compounds would exhibit higher activity toward electrophilic reagents compared to their benzoyl derivatives.

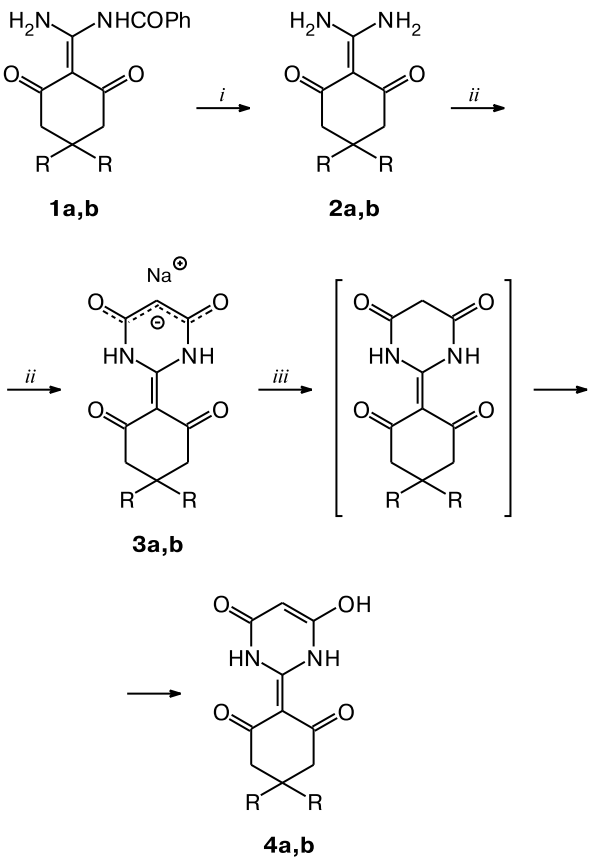
Aminals **2a,b** are white crystalline compounds, which are readily soluble in MeOH, DMSO, and acetone and moderately soluble in chloroform. The EI mass spectra of **2a,b** have intense molecular ion peaks. The ^1H NMR spectra (DMSO-d_6) of these compounds provide evidence for the symmetrical structure of the dioxocyclohexane moiety and show two broadened singlets for NH groups of two-proton intensity at δ ~7.1—7.6 and ~10.0.

We found that aminals **2a,b** can react with diethyl malonate as *N*,*N*-dinucleophiles. The reactions of these compounds in MeOH in the presence of MeONa under reflux give condensation products in the form of sodium salts **3a,b**. The treatment of the latter with dilute hydrochloric acid affords 6-hydroxy-2-(2,6-dioxocyclohexylidene)-1,2-dihydropyrimidin-4(3*H*)-ones **4a,b**. Salts **3a,b** remain intact when treated with acetic acid, which is indicative of the relatively strong acidic properties of compounds **4a,b**.

Sodium salts **3a,b** are white crystalline compounds, which are readily soluble in water and insoluble in MeOH, whereas compounds **4a,b** are readily soluble in ethanol and poorly soluble in water. Cyclohexylidenepyrimidines **4a,b** were synthesized as crystal hydrates containing water (two molecules according to elemental analysis data),

Published in Russian in *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 1, pp. 141—145, January, 2011.

Scheme 1



R = H (**a**), Me (**b**)

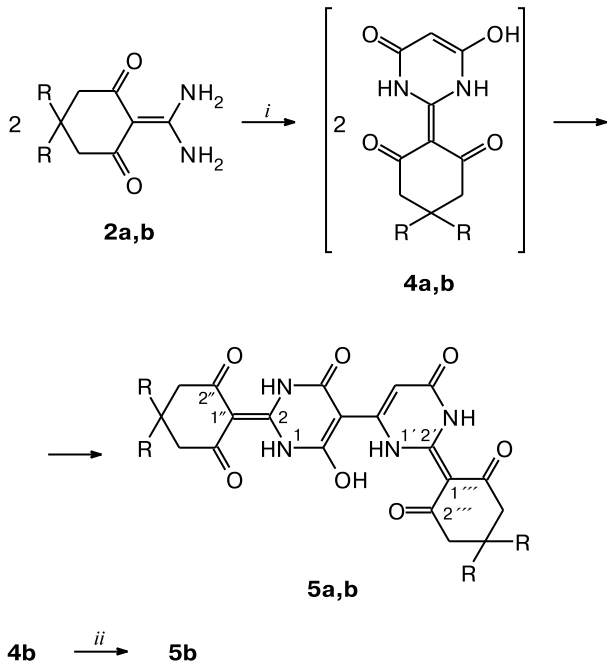
Reagents and conditions: *i.* MeONa, MeOH, 20 °C; *ii.* $CH_2(COOEt)_2$, MeONa, MeOH, Δ; *iii.* HCl.

which can easily be removed by drying *in vacuo* at 120 °C. The structures of compounds **3** and **4** were confirmed by spectroscopic methods. The EI mass spectra of pyrimidines **4a,b** have intense molecular ion peaks. The ^{1}H NMR spectra (DMSO-d_6) of these compounds, like those of salts **3**, show a singlet for the proton H(5) of the pyrimidine moiety. The signals of two CH_2CO groups of the cyclohexane ring are equivalent in the ^{1}H and ^{13}C NMR spectra. In addition, the signals of two NH groups (^{1}H NMR) and the C(4) and C(6) atoms (^{13}C NMR) of the pyrimidine ring are also identical. This confirms the more uniform electron density distribution in salts **3** and the fast hydroxyl proton transfer in compounds **4**. In the ^{1}H NMR spectra (DMSO-d_6) of pyrimidines **4**, the signals for H(5) (δ ~5.1) and NH (δ ~13.8—13.9) are observed at much lower field than the corresponding singlets in the spectra of their salts **3** (δ ~4.2 and ~12.6, respectively).

It should be noted that the condensation of diaminomethylidenemalononitrile with diethyl malonate was reported in the study,[20] but, since the NMR data were not obtained, the structure containing the dicarbonylmethylene moiety was assigned to the reaction product.

We attempted to perform the condensation of ketene aminals **2a,b** with diethyl malonate in the absence of bases. In this case, the reaction proceeded only at 200 °C, *i.e.*, under much more drastic conditions, and gave products, which were identified as 4,5′-bipyrimidines **5a,b** based on the mass spectrometry and NMR spectroscopy data (Scheme 2).

Scheme 2



R = H (**a**), Me (**b**)

Reagents and conditions: *i.* $CH_2(COOEt)_2$, 200 °C; *ii.* Ph_2O, 250 °C.

Apparently, under these conditions pyrimidines **4a,b** that are initially formed can undergo self-condensation accompanied by the elimination of water. This is supported by experimental data. In fact, compound **4b** gives dimer **5b** under reflux in diphenyl oxide or diethyl malonate.

Compounds **5a,b** are solids, which are soluble only in DMSO and water upon heating. The high-resolution ESI mass spectra of bipyrimidines **5a,b** have the ion peaks $[M + H]^+$ (the EI mass spectra did not show these peaks due, apparently, to the low volatility of these compounds). In the ^{1}H and ^{13}C NMR spectra (DMSO-d_6) of compounds **5a,b**, the signals of ethoxy groups are absent, but the spectra show signals corresponding to two cyclohexane rings. The ^{1}H NMR spectrum (DMSO-d_6) shows a singlet for H(5′) at δ 7.27 (in the 2D ^{1}H/^{13}C HSQC spectrum, this hydrogen has a cross-peak with the C(5′) atom at δ 99.36), a broadened singlet for the protons NH(1) and

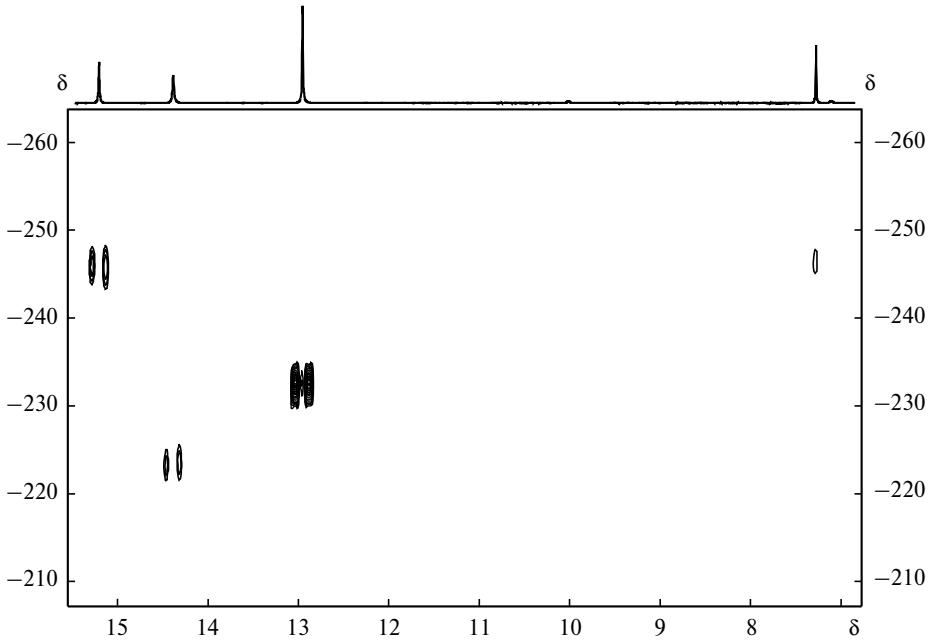


Fig. 1. 2D $^{1}H/^{15}N$ HMBC spectrum of compound **5b**.

NH(3) at δ 12.95, and broadened singlets of one-proton intensity for NH(3′) and NH(1′) at δ 14.39 and 15.20, respectively. In the ^{13}C NMR spectrum (DMSO-d_6), like in the spectra of compounds **4a,b**, the signals for C(4) and C(6) are equivalent (δ 160.6). In the 2D $^{1}H/^{13}C$ HMBC spectrum (DMSO-d_6), the following cross-peaks are observed: of the proton H(5′) (7.27 ppm) with the carbon atoms C(5) (85.95 ppm) and C(5′) (99.36 ppm), of the protons NH(1) and NH(3) (12.95 ppm) with the carbon atoms C(5) (85.95 ppm) and C(1″) (94.09 ppm), and of the proton NH(1′) (15.20 ppm) with the carbon atoms C(5′) (99.36 ppm), C(5) (85.95 ppm), and C(1‴) (95.50 ppm). In the 2D $^{1}H/^{15}N$ HMBC spectrum (Fig. 1), there are one-bond cross-peaks of the protons with the nitrogen atoms (δ_H/δ_N, ppm): 15.20/−246, H(1′)/N(1′); 14.39/−223, H(3′)/N(3′); 12.95/−232, H(1,3)/N(1,3). There are also three-bond cross-peaks of the proton H(5′) (δ_H 7.27) with the nitrogen atom N(1′) (δ_N −246 ppm), of the proton H(1) with the nitrogen atom N(3), and/or, on the contrary, of the proton H(3) with the nitrogen atom N(1).

Numerous biheterocycles have received considerable attention in organic chemistry (see the review[21]). Among these compounds, bipyrimidines with different structures have attracted attention as multidentate ligands and are used in the chemistry of coordination compounds (see, for example, the studies[22—26]).

To sum up, we showed that ketene aminals **2a,b** can be involved in the heterocyclization as *N,N*-dinucleophiles and, apparently, can be used in reactions with other dielectrophilic reagents (diketones, oxocarboxylic acid esters, *etc.*).

Experimental

The ^{1}H NMR spectra were recorded on a Bruker AM-300 instrument operating at 300 MHz. The ^{13}C NMR spectra and the 2D $^{1}H/^{13}C$ and $^{1}H/^{15}N$ HMBC spectra were measured on a Bruker Avance 600 instrument (600, 150, and 60.8 MHz for ^{1}H, ^{13}C, and ^{15}N, respectively). The signals of the residual protons of the deuterated solvent (7.27 ppm for $CDCl_3$ and 2.50 ppm for DMSO-d_6) were used as the references in the ^{1}H NMR spectra; the multiplet of DMSO-d_6 (39.50 ppm) was used as the internal standard in the ^{13}C NMR spectra. The ^{15}N chemical shifts were measured relative to $MeNO_2$ as the internal standard (high-field chemical shifts are given with a minus sign). The IR spectra were recorded on a Specord-M 82 instrument. The mass spectra were measured on a Kratos MS-30 instrument (EI, 70 eV, the temperature of the ionization chamber was 250 °C, direct inlet). The high-resolution mass spectra were obtained on a Bruker micrOTOF II instrument using the electrospray ionization (ESI) technique[27] in the positive-ion mode (capillary voltage was 4500 V). The mass scan range was m/z 50—3000 D; the external and internal calibration was performed (Electrospray Calibrant Solution, Fluka); solutions of samples in acetonitrile were introduced by the syringe injection; the flow rate was 3 μL min^{-1}; nitrogen was used as the nebulizing gas (4 L min^{-1}); the temperature of the interface was 180 °C. Diethyl malonate (Lancaster) and methanol were subjected to fractional distillation prior to use. Ketene aminals **1a,b** were synthesized according to a known procedure.[16]

2-Diaminomethylidenecyclohexane-1,3-dione (2a). A solution of MeONa in MeOH (1.5 mL, 1.9 mmol) was added to a solution of ketene aminal **1a** (0.5 g, 1.9 mmol) in MeOH (10 mL). The reaction mixture was stirred for 30 min and acidified with AcOH. Then the solution was concentrated *in vacuo* to dryness. Chloroform was added to the residue, and AcONa that remained undissolved was filtered off. The filtrate was concentrated, pe-

troleum ether was added, and the precipitate that formed was filtered off. The yield was 0.27 g (90%), m.p. 244—245 °C. Found (%): C, 54.32; H, 6.51; N, 18.30. $C_7H_{10}N_2O_2$. Calculated (%): C, 54.54; H, 6.54; N, 18.17. MS, *m/z* (I_{rel} (%)): 154 $[M]^+$ (100), 126 $[M - CO]^+$ (89), 113 $[M - CHCO]^+$ (26), 85 $[M - CHCO - CO]^+$ (54), 84 $[M - CH_2CO - CO]^+$ (87). IR (KBr), ν/cm^{-1}: 3416 (NH), 3340, 3250—2880 (NH, CH), 1616, 1568. ^{1}H NMR (DMSO-d_6), δ: 1.73 (m, 2 H, CH_2); 2.29 (t, 4 H, 2 CH_2, *J* = 6.0 Hz); 7.64 (br.s, 2 H, 2 NH); 10.00 (br.s, 2 H, 2 NH).

2-Diaminomethylidene-5,5-dimethylcyclohexane-1,3-dione (2b) was synthesized analogously to compound **2a** from ketene aminal **1b**. The yield was 95%, m.p. 190—192 °C. Found (%): C, 59.15; H, 7.69; N, 15.45. $C_9H_{14}N_2O_2$. Calculated (%): C, 59.32; H, 7.74; N, 15.37. MS, *m/z* (I_{rel} (%)): 182 $[M]^+$ (80), 154 $[M - CO]^+$ (19), 139 $[M - CO - Me]^+$ (19), 126 $[M - 2\,CO]^+$ (53), 98 (53), 83 (100). IR (KBr), ν/cm^{-1}: 3336 (NH), 3330—2870 (NH, CH), 1624, 1584. ^{1}H NMR (DMSO-d_6), δ: 0.94 (s, 6 H, 2 Me); 2.20 (s, 4 H, 2 CH_2); 7.14 (br.s, 2 H, 2 NH); 10.04 (br.s, 2 H, 2 NH).

6-Hydroxy-2-(2,6-dioxocyclohexylidene)-1,2-dihydropyrimidin-4(3*H*)-one (4a). Diethyl malonate (0.29 mL, 1.95 mmol) and a solution of MeONa in MeOH (1.5 mL, 1.95 mmol) were added to a solution of ketene aminal **2a** (0.1 g, 0.65 mmol) in MeOH (5 mL). The reaction mixture was refluxed for 1.5 h. The precipitate that formed was filtered off and washed with MeOH. The white precipitate that formed was dissolved in water (7 mL), and then dilute HCl (1 : 1) was added to the solution. The precipitate that formed was filtered off, successively washed with water and diethyl ether, and dried *in vacuo* at 120 °C. Compound **4a** was obtained in a yield of 0.14 g (95%), m.p. >300 °C. Found (%): C, 53.91; H, 4.45; N, 12.55. $C_{10}H_{10}N_2O_4$. Calculated (%): C, 54.06; H, 4.54; N, 12.61. MS, *m/z* (I_{rel} (%)): 222 $[M]^+$ (100), 194 $[M - CO]^+$ (17), 152 $[M - CH_2CO - CO]^+$ (57), 139 $[M - CH_2CO - CHCO]^+$ (44). IR (KBr), ν/cm^{-1}: 3436 (NH), 3310—2870 (NH, CH), 1676, 1630—1600, 1560. ^{1}H NMR (DMSO-d_6), δ: 1.84 (m, 2 H, CH_2); 2.48 (t, 4 H, 2 CH_2, *J* = 6.0 Hz); 5.09 (s, 1 H, CH); 13.93 (br.s, 2 H, 2 NH).

6-Hydroxy-2-(4,4-dimethyl-2,6-dioxocyclohexylidene)-1,2-dihydropyrimidin-4(3*H*)-one (4b). Diethyl malonate (0.25 mL, 1.65 mmol) and a solution of MeONa in MeOH (1.6 mL, 1.65 mmol) were added to a solution of ketene aminal **2b** (0.1 g, 0.55 mmol) in MeOH (5 mL). The reaction mixture was refluxed for 1.5 h. The precipitate that formed was filtered off and washed with MeOH. Sodium salt **3b** was obtained in a yield of 0.14 g (93%), m.p. >300 °C. IR (KBr), ν/cm^{-1}: 3430 (NH), 3200—2850 (NH, CH), 1700 (CO), 1608. ^{1}H NMR (DMSO-d_6), δ: 0.97 (s, 6 H, 2 Me); 2.30 (s, 4 H, 2 CH_2); 4.20 (s, 1 H, CH); 12.61 (br.s, 2 H, 2 NH). ^{13}C NMR (DMSO-d_6), δ: 27.78 (C$\underline{Me}_2$); 30.02 ($\underline{C}Me_2$); 51.19 (2 CH_2); 81.07 (C(5)); 94.53 ($\underline{C}(CO)_2$); 154.60 (C(2)); 161.54 (C(4), C(6)); 196.58 (2 CO). The assignment of the signals was made based on the 2D ^{1}H/^{13}C HMBC spectrum. The resulting salt was dissolved in water (7 mL), and dilute HCl (1 : 1) was added to the solution. The precipitate that formed was filtered off and successively washed with water and diethyl ether. Dihydrate of compound **4b** was obtained. Found (%): C, 50.02; H, 6.28; N, 9.65. $C_{12}H_{14}N_2O_4 \cdot 2\,H_2O$. Calculated (%): C, 50.35; H, 6.34; N, 9.79. The dihydrate was dried *in vacuo* at 120 °C, and compound **4b** was obtained in a yield of 0.09 g (62%), m.p. 187—188 °C. Found (%): C, 57.43; H, 5.58; N, 11.01. $C_{12}H_{14}N_2O_4$. Calculated (%): C, 57.59; H, 5.64; N, 11.19. MS, *m/z* (I_{rel} (%)): 250 $[M]^+$ (100), 222 $[M - CO]^+$ (28), 194 $[M - 2\,CO]^+$ (11), 166 $[M - 3\,CO]^+$ (43), 153 $[M - CHCO - 2\,CO]^+$ (45), 138 (21). IR (KBr), ν/cm^{-1}: 3392 (NH), 3300—2860 (NH, CH), 1640, 1572. ^{1}H NMR (DMSO-d_6), δ: 0.99 (s, 6 H, 2 Me); 2.39 (s, 4 H, 2 CH_2); 5.07 (s, 1 H, CH); 13.83 (br.s, 2 H, 2 NH). ^{13}C NMR (DMSO-d_6), δ: 27.62 (C$\underline{Me}_2$); 30.16 ($\underline{C}Me_2$); 50.60 (2 CH_2); 84.61 (C(5)); 94.93 ($\underline{C}(CO)_2$); 154.94 (C(2)); 160.26 (C(4), C(6)); 197.93 (2 CO).

6-Hydroxy-2-(2,6-dioxocyclohexylidene)-5-[4-oxo-2-(2,6-dioxocyclohexylidene)-1,2,3,4-tetrahydropyrimidin-6-yl]-1,2-dihydropyrimidin-4(3*H*)-one (5a). Diethyl malonate (1.5 mL) was added to ketene aminal **2a** (0.1 g, 0.65 mmol). The reaction mixture was refluxed for 2.5 h and then cooled. The precipitate was filtered off, washed with boiling MeOH, and dried. The yield of bipyrimidine **5a** was 0.09 g (64%), m.p. >300 °C. Found (%): C, 55.99; H, 4.28; N, 12.83. $C_{20}H_{18}N_4O_7$. Calculated (%): C, 56.34; H, 4.26; N, 13.14. High-resolution mass spectrum (HR-MS): Found: *m/z* 427.1241 $[M + H]^+$. $C_{20}H_{18}N_4O_7$. Calculated: $[M + H]^+$ = 427.1248. IR (KBr), ν/cm^{-1}: 3430 (NH), 3100—2850 (NH, CH), 1700—1500. ^{1}H NMR (DMSO-d_6), δ: 1.83 (m, 4 H, 2 CH_2); 2.44 (m, 8 H, 4 $\underline{CH_2}CO$); 7.27 (s, 1 H, H(5′)); 13.02 (br.s, 2 H, NH(1), NH(3)); 14.46 (br.s, 1 H, NH(3′)); 15.22 (br.s, 1 H, NH(1′)).

6-Hydroxy-2-(4,4-dimethyl-2,6-dioxocyclohexylidene)-5-[2-(4,4-dimethyl-2,6-dioxocyclohexylidene)-4-oxo-1,2,3,4-tetrahydropyrimidin-6-yl]-1,2-dihydropyrimidin-4(3*H*)-one (5b). ***A.*** Bipyrimidine **5b** was synthesized analogously to compound **5a** from ketene aminal **2b** and diethyl malonate. The yield was 54%, m.p. >300 °C. Found (%): C, 59.34; H, 5.54; N, 11.60. $C_{24}H_{26}N_4O_7$. Calculated (%): C, 59.74; H, 5.43; N, 11.61. High-resolution mass spectrum (HR-MS): Found: *m/z* 483.1858 $[M + H]^+$. $C_{24}H_{26}N_4O_7$. Calculated: $[M + H]^+$ = 483.1874. IR (KBr), ν/cm^{-1}: 3440 (NH), 3150—2860 (NH, CH), 1700—1500. ^{1}H NMR (DMSO-d_6), δ: 0.99 (s, 12 H, 4 Me); 2.35 (s, 4 H, 2 CH_2); 2.36 (s, 4 H, 2 CH_2); 7.27 (s, 1 H, H(5′)); 12.95 (br.s, 2 H, NH(1), NH(3)); 14.39 (br.s, 1 H, NH(3′)); 15.20 (br.s, 1 H, NH(1′)). ^{13}C NMR (DMSO-d_6), δ: 27.75 (C$\underline{Me}_2$); 27.77 (C$\underline{Me}_2$); 29.97, 30.03 (C(4″), C(4‴)); 51.10, 51.21 (C(3″), C(5″), C(3‴), C(5‴)); 85.95 (C(5)); 94.09 (C(1″)); 95.50 (C(1‴)); 99.36 (C(5′)); 149.84 (C(6′)); 153.42, 154.53 (C(2), C(2′)); 159.33 (C(4′)), 160.62 (C(4), C(6)); 196.21, 197.77 (C(2″), C(6″), C(2‴), C(6‴)).

B. A mixture of compound **4b** (0.1 g) and diphenyl oxide (1.0 g) was refluxed for 20 min and then cooled to 20 °C. Petroleum ether (15 mL) was added to the reaction mixture. The precipitate that formed was filtered off and washed with methanol (5 mL). Compound **5b** was obtained in a yield of 0.07 g (69%). The ^{1}H NMR spectrum of this compound was identical to that of the product prepared according to the method ***A***.

C. A mixture of compound **4b** (0.1 g) and diethyl malonate (1.5 mL) was refluxed for 2 h, during which the reaction mixture turned homogeneous and then the precipitate gradually formed. The reaction mixture was cooled to 20 °C. The precipitate was filtered off and washed with petroleum ether and methanol (3 mL). Compound **5b** was obtained in a yield of 0.04 g (42%). The ^{1}H NMR spectrum of this compound was identical to those of the products prepared according to the methods ***A*** and ***B***.

This study was financially supported by the Russian Academy of Sciences (Program for Basic Research of the Presidium of the Russian Academy of Sciences "Development of Methods for the Synthesis of Chemical Compounds and the Design of New Materials").

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Received September 3, 2010