Chemistry of Heterocyclic Compounds, Vol. 45, No. 7, 2009

SYNTHESIS AND OXIDATIVE AROMATIZATION OF 5-ACETYL-2-CYANOIMINO-6-METHYL-4-PHENYL-1,2,3,4-TETRAHYDROPYRIMIDINE WITH MANGANESE DIOXIDE

P. A. Solovyev[1] and A. D. Shutalev[1]*

The synthesis of 5-acetyl-2-cyanoimino-6-methyl-4-phenyl-1,2,3,4-tetrahydropyrimidine has been carried out by reaction of N-[(tosyl)(phenyl)methyl]-N'-cyanoguanidine with acetylacetone in the presence of sodium hydride with subsequent acid catalyzed dehydration of the 4-hydroxy-2-imino-hexahydropyrimidine obtained. The oxidative aromatization of the tetrahydropyrimidine synthesized using manganese dioxide has been studied. It was found that the products formed depend on the reaction temperature and are either 5-acetyl-2-carbamoylamino-4-methyl-6-phenylpyrimidine or 5-acetyl-2-amino-4-methyl-6-phenylpyrimidine or a mixture of both.

Keywords: 2-aminopyrimidines, manganese dioxide, 2-imino-1,2-dihydropyrimidines, 2-imino-1,2,3,4-tetrahydropyrimidines, guanidinoalkylation, oxidative aromatization.

2-Aminopyrimidines and their tautomeric 2-imino-1,2-dihydropyrimidines have been known for a long time and are a well studied class of heterocyclic compounds thanks to a reliable methods being available for their preparation [1, 2]. At the same time their 5-acyl-substituted analogs have been studied much less. It should be noted that the latter compounds are of marked interest since alkaloids of varied biological activity [3, 4] have been isolated recently from sea sponges and they contain a structurally similar fragment. High biological activity was also found in a series of synthetic examples of 5-acyl-2-aminopyrimidines and 5-acyl-2-iminodihydropyrimidines [5-7]. Known methods of synthesis for the indicated compounds are (C–C–C + N–C–N) type condensation [5, 6, 8, 9], reaction of 1,2-dihydropyrimidin-2-ones with amines [10], and also oxidative aromatization of 2-amino-1,4-dihydropyrimidines [11] using *t*-BuOOH as oxidant in the presence of $CuCl_2$. The search for novel, more efficient reagents for aromatization of 5-acyl-2-amino-1,4-dihydropyrimidines or their tautomers is very timely. Manganese dioxide occupies a special place amongst the large number of possible oxidants combining adequate activity with high selectivity [12, 13], already having been used for the aromatization of 2-amino-

* To whom correspondence should be addressed, e-mail: shutalev@orc.ru.

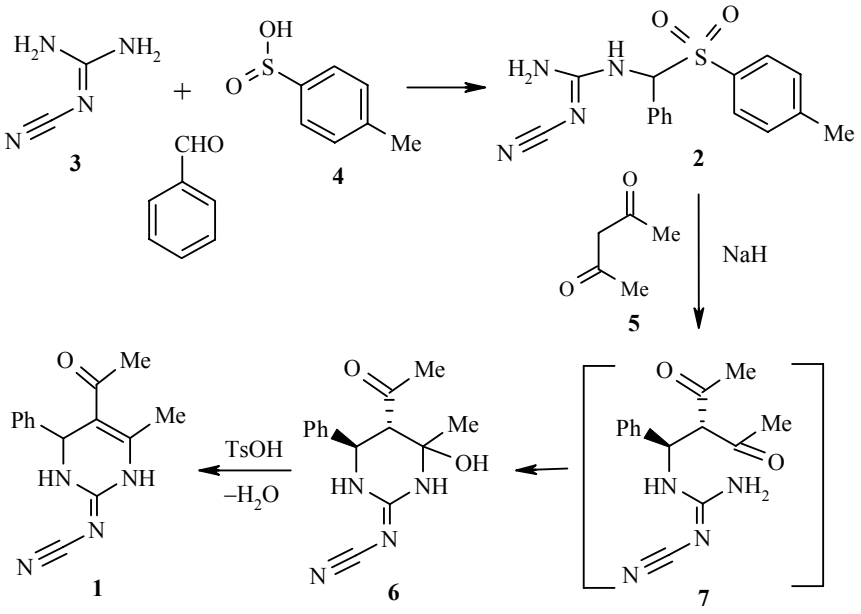
[1]M. V. Lomonosov Moscow State Academy of Fine Chemical Technology, Moscow 119571, Russia.

Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 7, pp. 1023-1030, July, 2009. Original article submitted January 5, 2009.

1,4-dihyropyrimidines which do not contain electron-acceptor substituents at position 5 [9]. In this work we report the synthesis of 5-acetyl-2-cyanoimino-6-methyl-4-phenyl-1,2,3,4-tetrahydropyrimidine (**1**) and its oxidative aromatization using manganese dioxide.

We have previously developed a convenient synthetic method for preparing 5-acyl-1,2,3,4-tetrahydropyrimidin-2-ones(thiones) based on the reaction of α-tosyl-substituted (thio)urea with 1,3-dicarbonyl compound enolates and subsequent dehydration of the 5-acyl-4-hydroxyhexahydropyrimidin-2-ones(thiones) formed [14, 15]. We have used a similar method in this work for the preparation of pyrimidine **1**.

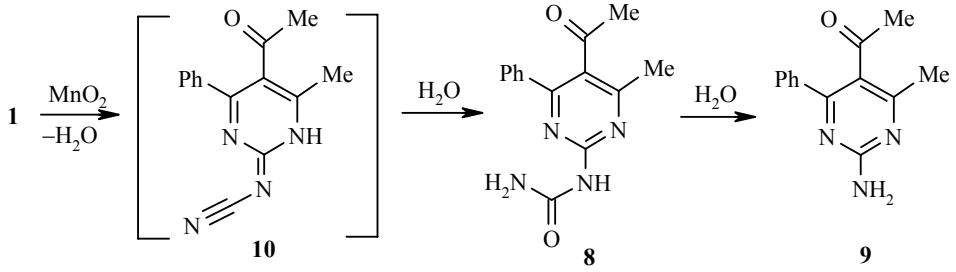
The starting N-[(tosyl)(phenyl)methyl]-N'-cyanoguanidine (**2**) was synthesized by a three component condensation of cyanoguanidine (**3**) with benzaldehyde and *p*-toluenesulfinic acid (**4**) at room temperature in water.



It should be noted that, in contrast to the reaction we have previously reported for cyanoguanidine with aliphatic aldehydes and sulfinic acid **4** (water, 20ºC, 2-4 days), where the yields of spectroscopically pure condensation products are 63-94% [16], the analogous reaction with benzaldehyde and acid **4** takes place significantly more slowly and without completion. According to ^{1}H NMR spectroscopic data the product from the reaction mixture after 7 days is a mixture of the guanidine **2** and sulfinic acid **4** in the ratio 49:51. Attempts to increase the yield and purity of compound **2** by carrying out the condensation in water at higher temperature (65ºC) or at room temperature in other solvents (acetonitrile, ethyl acetate, acetic acid, formic acid) were unsuccessful. Washing the obtained mixture of guanidine **2** and acid **4** with a saturated $NaHCO_3$ solution gave spectroscopically pure product **2** in 28% yield.

We have shown that the reaction of compound **2** with the sodium enolate of acetylacetone (generated by treatment of acetylacetone (**5**) with sodium hydride in anhydrous acetonitrile) at room temperature over 7.5 h gives an 84% yield of a mixture of the two diastereomers of 2-cyanoimino-4-hydroxyhexahydropyrimidine (**6**) in the ratio 92:8. It should be noted that, according to ^{1}H NMR spectroscopic data, both isomers have the same relative configuration of the chiral atoms C-5 and C-6 and in DMSO-d_6 solution exist with the substituents in an equatorial position for these atoms ($J_{5,6}$ being 12.0 and 10.8 Hz for the main and minor isomers respectively). Evidently these isomers differ in only the configuration of the chiral C-4 atom. Hence we can conclude that substitution of the tosyl group in the sulfone **2** using acetylacetone enolate occurs with full diastereoselectivity. The guanidinoalkyl derivative **7** obtained undergoes spontaneous heterocyclization to give the mixture of diastereomers of compound **6**.

The hydroxypyrimidine **6** readily undergoes dehydration upon refluxing for 1.25 h with *p*-toluenesulfonic acid (0.1 eq.) in acetonitrile to give a 91% yield of the tetrahydropyrimidine **1**. Oxidative aromatization of the latter was performed by us using a tenfold molar excess of MnO_2 through refluxing in various solvents.



In this way it was found that the result depends on the reflux temperature of the solvent used (Table 1). Hence refluxing pyrimidine **1** and MnO_2 in acetone (20 h) or in acetonitrile (26 h) gave the N-carbamoyl-substituted 2-aminopyrimidine **8** in 73 and 56% yields respectively. Using *p*-xylene as solvent (refluxing, 26 h) gave only the 2-aminopyrimidine **9** in 46% yield. Finally, refluxing in toluene gave a mixture of pyrimidines **8** and **9**, the composition of which depends on the reaction time (see Table 1). It should be noted that the expected N-cyano-substituted 2-imino-1,2-dihydropyrimidine **10** was not found amongst any of the separated reaction products.

The results obtained can evidently be explained by hydration of the cyano group in the pyrimidine **10** formed with aromatization in the presence of MnO_2 through the action of water arising during the course of the oxidation and the water present in the MnO_2. Hydrolysis of the ureido fragment in the compound **8** obtained and subsequent decarboxylation gives the pyrimidine **9**. Formation of compound **8** through initial hydration of the cyano group of tetrahydropyrimidine **1** and subsequent oxidation of the obtained N-carbamoyl-2-imino-1,2,3,4-tetrahdropyrimidine cannot be excluded, however. It should be noted that the use of manganese dioxide as catalyst in the hydration of nitriles has been reported in the literature [17].

The composition and structure of the synthesized compounds **1, 2, 6, 8, 9** were confirmed through elemental analytical results and from their IR, ^{1}H NMR, and ^{13}C NMR spectra. A important feature of their structure relates to the presence of unsymmetrical disubstituted (in compounds **2** and **9**) or trisubstituted (compounds **1, 6** and **8**) guanidine fragments so they can exist in three amino-imino tautomeric forms. From AM1 and PM3 semiempirical quantum-chemical calculations using the WinMopac program (version 7.2) and PM6 method in the Mopac2007 program we were able to conclude that the tetrahydropyrimidine **1** and

TABLE 1. Oxidative Aromatization of Tetrahydropyrimidine **1** in the Presence of Ten Equivalents of MnO_2

Experiment No	Solvent	Reaction temperature, °C	Reaction time, h	Molar ratio **8** : **9***	Yield of pyrimidine **8** or **9**, %
1	Acetone	56	20	100 : 0	73
2	Acetonitrile	78	26	100 : 0	56
3	*p*-Xylene	144	26	0 : 100	46
4	Toluene	115	7	78 : 22	—
5	Toluene	115	15	59 : 41	—
6	Toluene	115	47	18 : 82	—

* According to ^{1}H NMR spectroscopic data.

hexahydropyrimidine **6** exist principally as the tautomer with an exocyclic imino group and the aromatic pyrimidines **8, 9** as the tautomer with an exocyclic amino group. This conclusion agrees well with literature data regarding the position of the indicated tautomeric equilibrium in related systems [18, 19].

Hence, in the case of the preparation of the tetrahydropyrimidine **1** we have proposed a novel synthesis of 5-acyl-substituted pyrimidine-2-amines including the preparation of 5-acyl-2-cyanoimino-1,2,3,4-tetrahydropyrimidines and their subsequent oxidative aromatization in the presence of manganese dioxide.

EXPERIMENTAL

IR spectra for the synthesized compounds were recorded on a Bruker Equinox 55/S Fourier spectrometer for a suspension in vaseline oil. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker DPX 300 spectrometer (300 and 75 MHz respectively) using DMSO-d_6. Chemical shifts in the ^{1}H NMR spectra were measured relative to the central signal of the remaining solvent protons at 2.50 ppm and in the ^{13}C NMR spectra to the carbon atom signal from the solvent at 39.50 ppm. Monitoring of the reaction course and the purity of the products obtained was carried out by TLC on Silufol UV-254 plates (Kavalier, Czechoslovakia), Sorbfil (Russia), and Kieselgel F_{256} (Merck, Germany) using the system chloroform–methanol (9:1 and 5:1) and revealed using iodine vapor and UV radiation.

p-Toluenesulfinic acid (**4**) was prepared according to a known method [20] *via* acidification of a saturated aqueous solution of its sodium salt with hydrochloric acid at 0ºC. The filtered precipitate was washed with iced water and dried over P_2O_5. Activated MnO_2 was obtained according to the procedure reported in [12]. Acetylacetone was purified by holding for 10 days over anhydrous $MgSO_4$ and subsequent distillation *in vacuo*. Acetonitrile was purified by refluxing over P_2O_5 for 5-6 h and then distillation and repeated distillation over calcium hydride. Sodium hydride (60% suspension in oil) was thoroughly washed with dry petroleum ether and dried *in vacuo* before use. Benzaldehyde was distilled *in vacuo*. All other reagents were used without additional purification.

All of the quoted yields refer to spectroscopically and chromatographically pure product.

5-Acetyl-2-cyanoimino-6-methyl-4-phenyl-1,2,3,4-tetrahydropyrimidine (1). A mixture of compound **6** (0.1238 g, 0.455 mmol), *p*-toluenesulfonic acid monohydrate (0.0088 g, 0.046 mmol) and acetonitrile (2 ml) was refluxed with stirring on a magnetic stirrer for 1.25 h and solvent was removed *in vacuo*. Water (5 ml) was added to the solid residue and the mixture with a white precipitate was held at room temperature for 3.5 h before the precipitate was filtered off, washed twice with water, and dried to give compound **1** (0.1057 g, 91.4%); mp 249.5-250ºC (with decomp., acetonitrile). IR spectrum, ν, cm^{-1}: ~ 3208 sh, 3178 w. br. 3056 w. br. (NH), 2188 w (C≡N), 1657 w (C=O), 1631 w, 1520 w [NH–C(=N)–NH], 700 w (Ph). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 10.12 (1H, br. s, H-1); 9.24 (1H, br. d, $J_{3,4}$ = 3.4, H-3); 7.23-7.40 (5H, m, Ph); 5.36 (1H, d, $J_{4,3}$ = 3.4, H-4); 2.34 (3H, s, CH_3CO); 2.18 (3H, s, 6-CH_3). ^{13}C NMR spectrum, δ, ppm: 194.45 (C=O); 154.77 (C-2); 145.55 (C-6); 142.61 (C-1 Ph); 128.70 (C-3 and C-5 Ph); 127.86 (C-4 Ph); 126.53 (C-2 and C-6 Ph); 116.39 (C≡N); 111.20 (C-5); 52.88 (C-4); 30.47 ($\underline{C}H_3CO$); 18.37 (6-CH_3). Found, %: C 66.26; H 5.53; N 21.84. $C_{14}H_{14}N_4O$. Calculated, %: C 66.13; H 5.55; N 22.03.

N-[Tosyl(phenyl)methyl]-N'-cyanoguanidine (2). Acid **4** (4.313 g, 27.61 mmol) was added with stirring using a magnetic stirrer to an emulsion of benzaldehyde (2.931 g, 27.62 mmol) in water (150 ml). The white suspension obtained was stirred for 15 min and finely divided cyanoguanidine (2.334 g, 27.76 mmol) and water (50 ml) were added. The creamy reaction mass was held for 7 days at room temperature in a closed vessel, periodically stirring its contents. The precipitate was filtered off, thoroughly washed with water, and dried. According to ^{1}H NMR spectroscopy the product (5.577 g) was a mixture (49:51) of guanidine **2** and acid **4**. In order to remove acid **4** the mixture was washed with a saturated aqueous solution of $NaHCO_3$ (20 ml) cooled to 15ºC, followed by water, and then dried to give compound **2** (2.5 g, 27.6%) which was used without

further purification. An analytical sample was prepared by recrystallization from acetonitrile. Mp 135.5-136ºC (decomp.). IR spectrum, ν, cm^{-1}: 3434 w, 3356 w, 3200 w (NH), 3062 m, 3046 w (Ph, Ar), 2186 w (C≡N), 1641 w, 1622 s, 1607 w, 1598 w, 1546 vw (NH_2–C(=N)–NH), 1502 m (Ph, Ar), 1284 w (SO_2), 1142 w (SO_2), 816 m (Ar), 697 w (Ph). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 8.39 (1H, d, $J_{NH,CH}$ = 10.5, NH); 7.73 (2H, AA' part of AA'XX' spin system, J_{ortho} = 8.3, H-2 and H-6, 4-MeC_6H_4); 7.45 (2H, m, XX' part of AA'XX' spin system, J_{ortho} = 8.3, H-3 and H-5 Ar); 7.41-7.52 (5H, m, Ph); 6.92 (2H, br. s, NH_2); 6.21 (1H, d, $^3J_{CH,NH}$ = 10.5, NCH); 2.42 (3H, s, CH_3). ^{13}C NMR spectrum, δ, ppm: 160.10 (C=N); 145.02 (C-4 Ar); 133.69 (C-1 Ar); 130.31 (C-1 Ph); 129.70 (C-3 and C-5 Ar); 129.53 (C-4 Ph); 129.11, 129.06 (C-2,6 and C-3,5 Ph); 128.38 (C-2 and C-6 Ar); 116.38 (C≡N); 73.10 (NCH); 21.20 (CH_3). Found, %: C 58.40; H 5.27; N 16.81. $C_{16}H_{16}N_4O_2S$. Calculated, %: C 58.52; H 4.91; N 17.06.

5-Acetyl-2-cyanoimino-4-hydroxy-4-methyl-6-phenylhexahydropyrimidine (6). A solution of acetylacetone (0.14 g, 1.398 mmol) in anhydrous acetonitrile (4 ml) was added with stirring on a magnetic stirrer to a suspension of sodium hydride (0.0334 g, 1.392 mmol) in acetonitrile (6 ml). After 5 min compound **2** (0.4181 g, 1.273 mmol) and additional acetonitrile (2 ml) were added to the reaction mixture. The obtained white suspension was stirred for 7.5 h at room temperature, the solvent was removed *in vacuo*, and water (5 ml) and saturated aqueous $NaHCO_3$ (5 ml) were added to the solid residue. The mixture was held for 16 h at room temperature, cooled, the precipitate was filtered off, washed with iced water, and dried. The product (0.2892 g, 83.4%) was a 92:8 mixture of two diastereomers according to ^{1}H NMR data. Mp 178.5ºC (acetonitrile) with decomp., foaming at a heating rate of 1ºC/33 s. In the case of 1ºC over more than 50 s the substance decomposes without melting at ~ 172-173ºC and then melts with decomposition at 226-228ºC. IR spectrum, ν, cm^{-1}: 3452 w, 3276 w, 3192 w, ~ 3170 sh (NH, OH), 2196 w, 2183 w (C≡N), ~ 1705 sh, 1698 w (C=O), 1644 w (NH–C(=N)–NH), 1588 m (Ph), 1549 w (NH–C(=N)–NH), 1496 m (Ph), 700 w (Ph). ^{1}H NMR spectrum of main isomer, δ, ppm (*J*, Hz): 8.34 (1H, d, $J_{3,1}$ = 1.8, H-3); 8.01 (1H, d, $J_{1,3}$ = 1.8, H-1); 7.22-7.41 (5H, m, Ph); 6.29 (1H, s, OH); 4.86 (1H, d, $J_{6,5}$ = 12.0, H-6); 3.05 (1H, d, $J_{5,6}$ = 12.0, H-5); 1.95 (3H, s, CH_3CO); 1.42 (3H, s, 4-CH_3). ^{1}H NMR spectrum of minor isomer, δ, ppm (*J*, Hz): 8.04 (1H, d, $J_{3,1}$ = 1.7, H-3); 7.93 (1H, d, $J_{1,3}$ = 1.7, H-1); 7.22-7.41 (5H, m, Ph, signals obscured by the signals for the Ph protons of the main isomer); 6.43 (1H, s, OH); 4.59 (1H, d, $J_{6,5}$ = 10.8, H-6); 3.42 (1H, d, $J_{5,6}$ = 10.8, H-5); 1.93 (3H, s, CH_3CO); 1.31 (3H, s, 4-CH_3). ^{13}C NMR spectrum of main isomer, δ, ppm: 205.50 (C=O); 156.51 (C-2); 138.95 (C-1 Ph); 128.41 (C-3 and C-5 Ph); 128.19 (C-4 Ph); 128.08 (C-2 and C-6 Ph); 117.62 (C≡N); 77.76 (C-4); 61.73 (C-5); 53.32 (C-6); 31.06 ($\underline{C}H_3CO$); 26.51 (4-CH_3). ^{13}C NMR spectrum of minor isomer, δ, ppm: 206.13 (C=O); 156.48 (C-2); 139.57 (C-1 Ph); 128.36 (C-3 and C-5 Ph); 127.95 (C-4 Ph); 127.46 (C-2 and C-6 Ph); 117.71 (C≡N); 80.57 (C-4); 60.83 (C-5); 54.40 (C-6); 33.49 ($\underline{C}H_3CO$); 24.94 (4-CH_3). Found, %: C 61.49; H 5.95; N 20.66. $C_{14}H_{16}N_4O_2$. Calculated, %: C 61.75; H 5.92; N 20.58.

5-Acetyl-2-carbamoylamino-4-methyl-6-phenylpyrimidine (8). A mixture of pyrimidine **1** (0.1031 g, 0.405 mmol), MnO_2 (0.3566 g, 4.102 mmol), and acetone (15 ml) was refluxed for 20 h, cooled, and the precipitate was filtered off using a hard texture filter, and washed with acetone (30 ml). The combined filtrate was evaporated to dryness *in vacuo*. Petroleum ether (5 ml) cooled to 0ºC was added to the oily residue and triturated to formation of a suspension. The precipitate was filtered off, washed with a small amount of petroleum ether, and dried to give compound **8** (0.0798 g, 72.8%); mp 204.5-205.5ºC (toluene). IR spectrum, ν, cm^{-1}: 3330 w br, 3197 sh, 3168 w br, 3152 w br (NH), 1695 vw (C=O in Ac), 1660 w ("amide I"), 1550 vw (C=N and "amide II"), 698 w (Ph). ^{1}H NMR spectrum, δ, ppm (J, Hz): 9.80 (1H, s, NH); 8.46 (1H, br. s, NH_2); 7.49-7.61 (5H, m, Ph); 7.16 (1H, br. s, NH_2); 2.41 (3H, s, CH_3CO); 2.02 (3H, s, 4-CH_3). ^{13}C NMR spectrum, δ, ppm: 203.45 ($CH_3\underline{C}O$); 165.36 (C-6); 162.80 (C-4); 157.04 (C-2); 154.41 (NHC=O); 137.14 (C-1 Ph); 130.61 (C-4 Ph); 128.97 and 128.38 (C-2,6 and C-3,5 Ph); 126.42 (C-5); 31.92 ($\underline{C}H_3CO$); 22.33 (4-CH_3). Found, %: C 62.40; H 5.37; N 20.65. $C_{14}H_{14}N_4O_2$. Calculated, %: C 62.21; H 5.22; N 20.73.

Compound 8 was also prepared in 56% yield by a similar method but using acetonitrile as solvent (refluxing, 26 h). The product obtained was additionally washed with water to remove the acetamide formed in the reaction.

5-Acetyl-2-amino-4-methyl-6-phenylpyrimidine (9). A mixture of pyrimidine **1** (0.4148 g, 1.631 mmol), MnO_2 (1.4184 g, 16.315 mmol), and *p*-xylene (55 ml) was refluxed for 26 h and the precipitate was filtered on a hard texture filter and washed with hot *p*-xylene (20 ml). The combined filtrate was evaporated *in vacuo* and the oily residue was treated with petroleum ether (10 ml), cooled to 0ºC, and triturated to the formation of a suspension. The precipitate was filtered, washed with a small amount of petroleum ether, and dried to give compound **9** (0.1714 g, 46.2%) with mp 142.5-144ºC (hexane). IR spectrum, ν, cm^{-1}: 3221 w, 3166 w (NH), 3053 m (Ph), 1681 w (C=O), 1655 w (NH_2), 1543 sh, 1527 w (C=N), 698 w (Ph). ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 7.42-7.52 (5H, m, Ph); 7.09 (2H, s, NH_2); 2.27 (3H, s, CH_3CO); 1.88 (3H, s, 4-CH_3). ^{13}C NMR spectrum, δ, ppm: 203.70 (C=O); 165.29 (C-6); 164.25 (C-4); 162.18 (C-2); 138.59 (C-1 Ph); 129.95 (C-4 Ph); 128.65 and 128.32 (C-2,6 and C-3,5 Ph); 122.81 (C-5); 32.19 ($\underline{C}H_3CO$); 22.43 (4-CH_3). Found, %: C 68.65; H 5.55; N 18.37. $C_{13}H_{13}N_3O$. Calculated, %: C 68.65; H 5.55 N 18.37. $C_{13}H_{13}N_3O$. Calculated, %: C 68.71; H 5.77; N 18.49.

REFERENCES

1. D. H. R. Barton and W. D. Ollis (editors), *Comprehensive Organic Chemistry* [Russian translation], Vol. 8, Khimiya, Moscow (1985), p. 121.
2. R. Elderfield (editor), *Heterocyclic Compounds* [Russian translation], Vol. 6, Inostr. Lit., Moscow (1960), p. 200.
3. H.-M. Hua, J. Peng, D. C. Dunbar, R. F. Schinazi, A. G. C. Andrews, C. Cuevas, L. F. Garcia-Fernandez, M. Kelly, and M. T. Hamann, *Tetrahedron*, **63**, 11179 (2007).
4. L. C. Chang, N. F. Whittaker, and C. A. Bewley, *J. Nat. Prod.*, **66**, 1490 (2003).
5. S. W. McCombie, J. R. Tagat, S. F. Vice, S.-L. Lin, R. Steensma, A. Palani, B. R. Neustadt, B. M. Baroudy, J. M. Strizki, M. Endres, K. Cox, N. Dan, and C.-C. Chou, *Bioorg. Med. Chem. Lett.*, **13**, 567 (2003).
6. A. Palani, S. Shapiro, J. W. Clader, W. J. Greenlee, S. Vice, S. McCombie, K. Cox, J. Strizki, and B. M. Baroudy, *Bioorg. Med. Chem. Lett.*, **13**, 709 (2003).
7. J. J. Matasi, J. P. Caldwell, J. Hao, B. Neustadt, L. Arik, C. J. Foster, J. Lachowicz, and D. B. Tulshian, *Bioorg. Med. Chem. Lett.*, **15**, 1333 (2005).
8. A. C. Veronese, R. Callegari, C. F. Morelli, and C. F. Vincetiri, *Tetrahedron*, **53**, 14497 (1997).
9. A. Kuno, H. Sakai, M. Okhubo, and H. Takasugi, *Chem. Pharm. Bull.*, **41**, 163 (1993).
10. F.-A. Kang, J. Kodah, Q. Guan, X. Li, and W. L. Murray, *J. Org. Chem.*, **70**, 1957 (2005).
11. K. Yamamoto, Y. G. Chen, and F. G. Buono, *Org. Lett.*, **7**, 4673 (2005).
12. A. J. Fatiadi, *Synthesis*, 65 (1976).
13. A. J. Fatiadi, *Synthesis*, 133 (1976).
14. A. D. Shutalev and V. A. Kuksa, *Khim. Geterotsikl. Soedin.*, 97 (1995). [*Chem. Heterocycl. Comp.*, **31**, 86 (1995)].
15. A. D. Shutalev, E. A. Kishko, N. V. Sivova, and A. Yu. Kuznetsov, *Molecules*, **3**, 100 (1998).
16. A. D. Shutalev in: *Proc. of the Fourth Intern. Electronic Conf. on Synthetic Organic Chemistry,* September 1-30 (2000). http://www.mdpi.org/escoc-4.htm
17. M. J. Cook, E. J. Forbes, and G. M. Khan, *Chem. Commun.*, 121 (1966).
18. A. R. Katritzky and J. M. Lagowski in: A. R. Katritzky (editor), *Advances in Heterocyclic Chemistry*, Vol. 1, Academic Press, New York (1963), p. 339.
19. B. Stanovnik, M. Tišler, A. R. Katritzky, and O. V. Denisko in: A. R. Katritzky (editor), *Advances in Heterocyclic Chemistry*, Vol. 91, Academic Press, Amsterdam (2006), p. 1.
20. L. Tittse and T. Aikher, *Preparative Organic Chemistry* [Russian translation], Mir, Moscow (1999), p. 92.