Synthesis of functional 2-substituted 4-phenyl-9*H*-pyrimido[4,5-*b*]indoles

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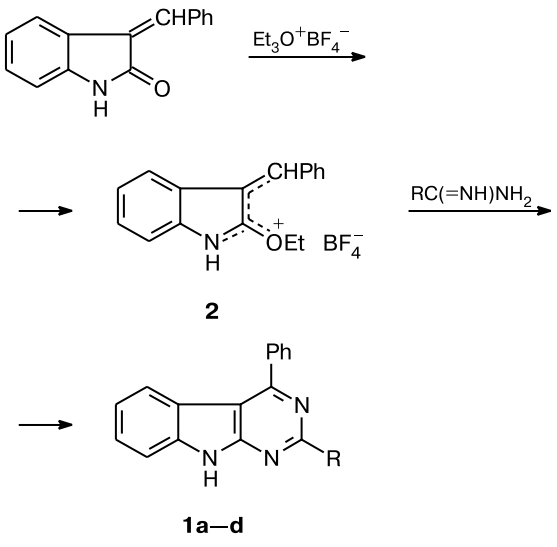
A method was developed for the synthesis of 2-oxo-4-phenyl-2,3-dihydro-9*H*-pyrimido[4,5-*b*]indole as well as of 2-chloro- and 2-nitramino-4-phenylpyrimido[4,5-*b*]indoles. The replacement of the chlorine atom in 2-chloropyrimidoindole gave rise to a number of its functional derivatives (morpholino, azido, and cyano). The reaction of 2-chloro-substituted pyrimidoindole with hydrazine hydrate and catalytic hydrogenation of 2-nitraminopyrimidoindole were studied.

Key words: 2-oxo-4-phenyl-2,3-dihydro-9*H*-pyrimido[4,5-*b*]indole, 2-substituted 4-phenyl-9*H*-pyrimido[4,5-*b*]indoles, diazotization, nucleophilic substitution, catalytic hydrogenation.

Functional pyrimido[4,5-*b*]indole derivatives, in particular, its amino derivatives, exhibit a broad spectrum of biological activities, which opens up a possibility of their use in the drug design.[1—4] Previously,[5] we have developed a procedure for the synthesis of 2-R-4-phenyl-9*H*-pyrimido[4,5-*b*]indoles **1** based on the reaction of 2-ethoxy(3-benzylidene)indolenine tetrafluoroborate (**2**) with amidines or guanidines (Scheme 1).

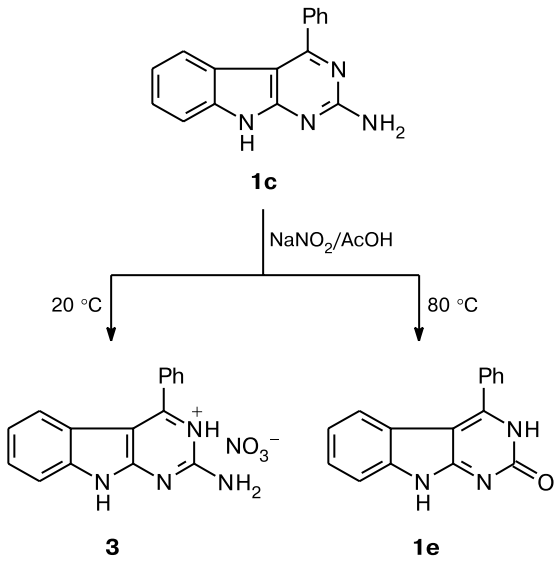
Scheme 1



R = Me (**a**), Ph (**b**), NH_2 (**c**), $NHSO_2Ar$ (**d**)

The synthesis of 2-oxo derivative **1e** (R = OH) according to Scheme 1 proved to be inefficient. Thus, direct condensation of tetrafluoroborate **2** with urea afforded a product whose purification led to large losses resulting in a low yield of crystalline 2-oxo-4-phenyl-2,3-dihydro-9*H*-pyrimido[4,5-*b*]indole (**1e**). We found that diazotization of 2-aminopyrimidoindole **1c** in AcOH at 80 °C afforded compound **1e** in a yield of >90% and this compound can be used in subsequent syntheses without additional purification (Scheme 2). It should be noted that this reaction performed at ~20 °C gave the nitric salt of the starting amine **3**. The formation of the latter is associated with protonation of the basic aminopyrimidine fragment by nitric acid, which was generated due to dispro-

Scheme 2



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portionation of nitrous acid.[6] This conclusion was confirmed by the synthesis of authentic nitrate **3** upon the direct addition of HNO_3 to a solution of amino derivative **1c** in AcOH. The structure of nitrate **3** was established by NMR spectroscopy and X-ray diffraction analysis.[7]

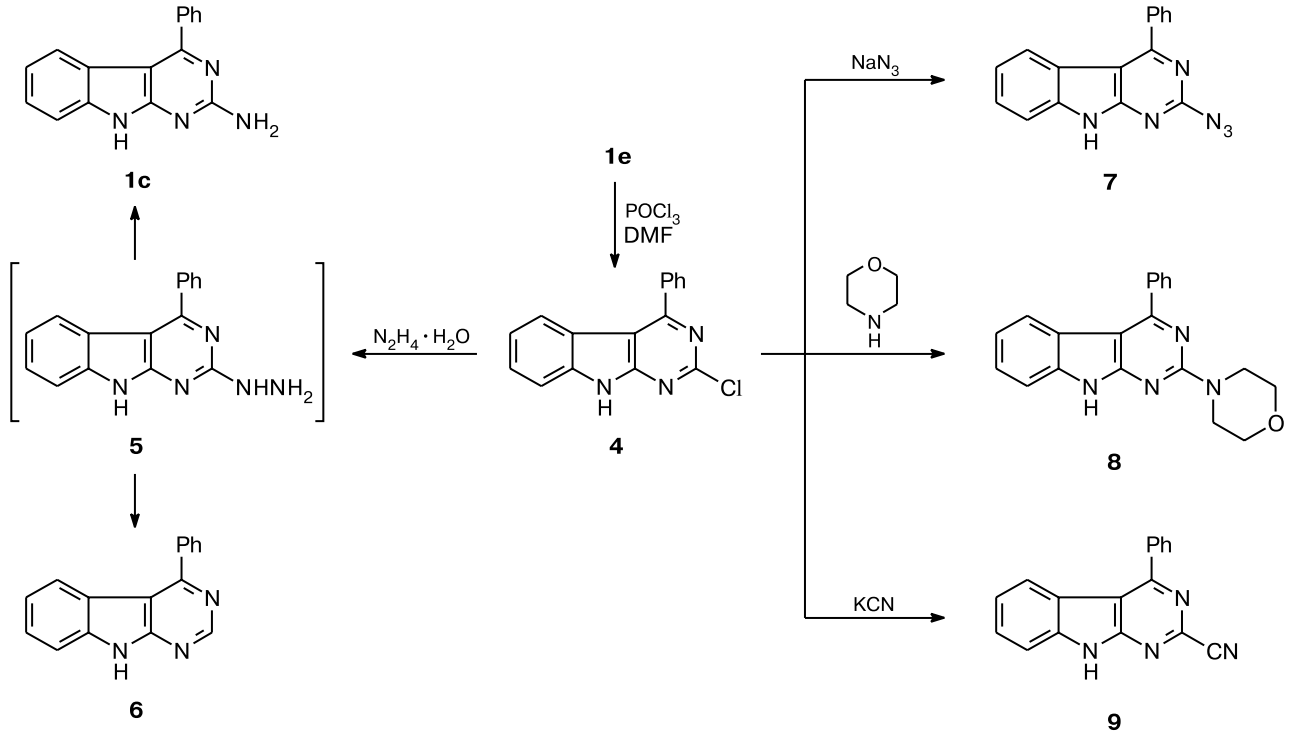
The reaction of 2-oxo-substituted compound **1e** with the Vilsmeier reagent afforded 2-chloro-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (**4**) in high yield. The Cl atom in the latter compound is labile in reactions with nucleophilic reagents. We used hydrazine hydrate, morpholine, sodium azide, and potassium cyanide as nucleophilic reagents (Scheme 3).

However, all attempts to synthesize 2-hydrazinopyrimidoindole **5** by the reaction of chloro derivative **4** with hydrazine hydrate failed. The reaction in DMSO at temperatures below 90 °C did not lead to the replacement of the Cl atom. The starting chloro derivative **4** disappeared from the reaction mixture only when the temperature was increased, an excess of hydrazine hydrate was used, and the mixture was heated over a long period. However, the reaction performed under these conditions afforded a complex mixture of products from which only 2-amino-substituted derivative **1c** (29%) and pyrimidoindole **6** (14%) were isolated by preparative TLC. The presence of 2-hydrazine-substituted compound **5** in the mixture was detected only from the line at m/z 275 ($[M]^+$, I_{rel} 3%) in the mass spectrum. The reaction of chloro derivative **4** with hydrazine hydrate in *N*-methylpyrrolidone proceeded analogously. Only compounds **1c** (20%) and **6** (7%) were isolated from the reaction mixture by preparative TLC. This result can be attributed to the fact that 2-hydrazine-containing derivative **5** contains the unstable hydrazine group, which was subjected to reduction and elimination under the reaction conditions.

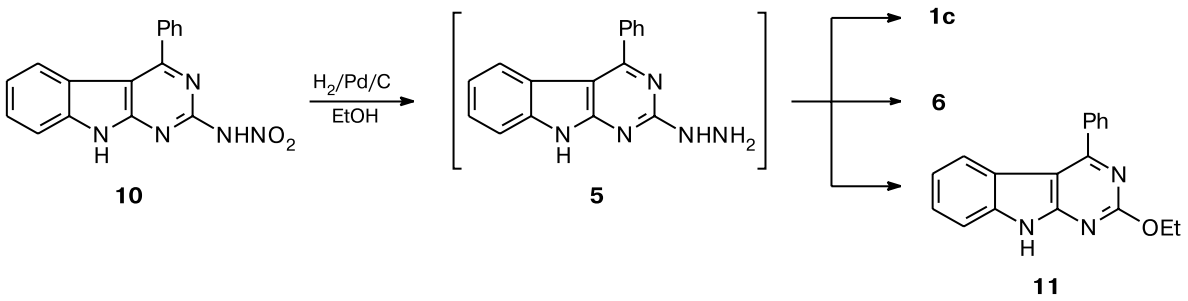
Heating of 2-chloropyrimidoindole **4** with other nucleophilic reagents in DMSO at 120 °C afforded the corresponding 2-substituted pyrimidoindoles **7**—**9** in high yields.

2-Nitramino-4-phenylpyrimido[4,5-*b*]indole (**10**), which was prepared by the reaction of tetrafluoroborate **2** with nitroguanidine according to a procedure analogous to that described previously,[5] was also of interest for the synthesis of 2-substituted pyrimidoindoles. It was expected that the reactions of 2-nitramino-substituted compound **10** with nucleophiles, such as hydrazine hydrate, morpholine, hydroxylamine, *etc*., would give the corresponding functional derivatives by analogy with the synthesis of these derivatives from 2-nitraminopyrimidines.[8—10] However, unlike the latter compounds, 2-nitraminopyrimidoindole **10** reacted with none of the above-mentioned nucleophiles. There are several reasons for such a behavior. First, as was exemplified by the chloro derivative, the nucleophilic substitution in the pyrimidoindole series proceeds more difficultly than that in the pyrimidine series. Second, deprotonation of the nitramino group under the action of rather basic nucleophilic reagents and blocking

Scheme 3



Scheme 4



of the nucleophilic attack at the reaction center of the resulting anion must not be ruled out (*cf.* the acidities of some nitraminopyrimidines: $pK_a < 4$ [11]).

We attempted to prepare 2-hydrazinopyrimidoindole **5** by catalytic hydrogenation of nitramino derivative **10** over Pd/C in EtOH. However, this reaction gave a mixture containing 2-aminopyrimidoindole **1c** as the major product. The reaction mixture contained also insignificant amounts of pyrimidoindole **6** and 2-ethoxypyrimidoindole **11** (Scheme 4). We failed to isolate hydrazine derivative **5** although the mass spectrum of the mixture had the corresponding line at *m/z* 275 ($[M]^+$, I_{rel} 4%).

Apparently, compound **1c** was generated through hydrogenolysis of the N—N bond in intermediate hydrazine derivative **5**. An analogous transformation of the hydrazine group into the amino group was observed upon chemical or catalytic reduction of hydrazinopyrimidines.[12] The formation of compounds **6** and **11** can be interpreted as elimination of the hydrazine group from hydrazine derivative **5** accompanied by its substitution for the H atom or the ethoxy group.

The structures of the new pyrimido[4,5-*b*]indole derivatives were confirmed by ^1H and ^{13}C NMR spectroscopy taking into account the known data.[13]

To summarize, we developed a convenient procedure for the synthesis of 2-oxo-4-phenyl-2,3-dihydro-9*H*-pyrimido[4,5-*b*]indole and of 2-chloro-4-phenylpyrimido[4,5-*b*]indole as the key compounds for the preparation of different 2-substituted pyrimidoindoles.

Experimental

The IR spectra were recorded on a Specord M-80 spectrometer in KBr pellets. The ^1H and ^{13}C NMR spectra were measured on Bruker AC-200, Bruker AC-400, and Bruker DRX-500 spectrometers in DMSO-d_6 using the signals of the solvent at δ 2.50 (^1H) and 39.5 (^{13}C) with respect to Me_4Si as the internal standard. The mass spectra were obtained on a Finnigan MAT-8200 spectrometer (EI, 70 eV). The course of the reactions was monitored by TLC on Silufol UV-254 plates; visualization was carried out with UV light. The solvents were dried according to standard procedures.

2-Oxo-4-phenyl-2,3-dihydro-9*H*-pyrimido[4,5-*b*]indole (1e). A solution of $NaNO_2$ (0.72 g, 10.4 mmol) in water (4 mL) was added dropwise with stirring to a solution of compound **1c** (0.45 g, 1.7 mmol) in AcOH (16 mL) at 80 °C for 1—2 min. The reaction mixture was stirred at this temperature for 3 h and then cooled to 10—15 °C. The precipitate was separated, successively washed with EtOH (2×3 mL) and water (5 mL), and dried. Compound **1e** was obtained in a yield of 0.42 g (93%), m.p. >360 °C. The sample for analysis was recrystallized from DMF. Found (%): C, 73.70; H, 4.34; N, 16.20. $C_{16}H_{11}N_3O$. Calculated (%): C, 73.55; H, 4.24; N, 16.08. MS, *m/z* (I_{rel} (%)): 261 $[M]^+$ (83); 260 $[M - 1]^+$ (100). IR, ν/cm^{-1}: 1645 (C=O); 3400 (NH). ^1H NMR (500 MHz), δ: 6.97 (ddd, 1 H, H(6), ^3J = 8.0 Hz, ^3J = 5.0 Hz, ^4J = 3.0 Hz); 7.18 (d, 1 H, H(8), ^3J = 7.5 Hz); 7.25—7.28 (m, 2 H, H(5), H(7)); 7.63—7.69 (m, 3 H, Ph); 7.75—7.78 (m, 2 H, Ph); 11.73 (br.s, 2 H, H(1), H(9)). ^{13}C NMR (125 MHz), δ: 100.85, 111.24, 119.64, 120.34, 120.78, 126.34, 128.69, 128.85, 131.03, 131.67, 139.95, 151.92, 157.08, 162.58.

2-Amino-4-phenyl-9*H*-pyrimido[4,5-*b*]indole nitrate (3). Sodium nitrite (0.08 g, 1.2 mmol) was added portionwise with stirring to a solution of compound **1c** (0.3 g, 1.15 mmol) in AcOH (7 mL) at 20 °C for 20 min. The reaction mixture was stirred at this temperature for 30 min. The precipitate that formed was washed with water and dried *in vacuo*. Nitrate **3** was obtained in a yield of 0.17 g (43%), m.p. 243—248 °C. The IR spectrum and m.p. of the product were identical with those of an authentic sample.[7]

2-Chloro-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (4). Compound **1e** (1.95 g, 7.5 mmol) was added with stirring to the Vilsmeier reagent, which was prepared from $POCl_3$ (7 mL) and DMF (23 mL), at 20 °C. Then the reaction mixture was heated to 60 °C, stirred at this temperature for 5 h, cooled, and poured onto ice. The precipitate that formed was refluxed in 15% HCl (160 mL) for 1.5 h. The precipitate was separated, washed with a solution of $NaHCO_3$ and then with water until the reaction mixture became neutral, and dried *in vacuo*. Pyrimidoindole **4** was obtained in a yield of 1.63 g (78%), m.p. 312—315 °C (from xylene). Found (%): C, 68.30; H, 3.65; Cl, 12.80; N, 14.80. $C_{16}H_{10}ClN_2$. Calculated (%): C, 68.68; H, 3.60; Cl, 12.67; N, 15.02. MS, *m/z* (I_{rel} (%)): 279 $[M]^+$ (72); 278 $[M - 1]^+$ (100). IR, ν/cm^{-1}: 3410 (NH). ^1H NMR (400 MHz), δ: 7.23 (td, 1 H, H(6), ^3J = 8.0 Hz, ^4J = 1.2 Hz); 7.55 (td, 1 H, H(7), ^3J = 8.0 Hz, ^4J = 0.8 Hz); 7.61 (d, 1 H, H(8), ^3J = 8.0 Hz); 7.65—7.70 (m, 3 H, Ph); 7.78 (d, 1 H, H(5), ^3J = 8.0 Hz); 7.89—7.94 (m, 2 H, Ph); 12.50—13.00 (br.s, 1 H, NH). ^{13}C NMR (100 MHz), δ: 109.80 (C(4a)); 112.33 (C(8)); 118.24 (C(4b)); 121.37 (C(6)); 121.92 (C(5)); 128.14 (C(7)); 128.78

(C_o, C_m, Ph); 130.55 (C_p, Ph); 136.90 (C_i, Ph); 138.97 (C(8a)); 155.14 (C(9a)); 157.69 (C(4)); 160.81 (C(2)).

2-Azido-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (7). Sodium azide (0.21 g, 3.2 mmol) was added to a solution of pyrimidoindole **4** (0.3 g, 1.1 mmol) in dry DMSO (20 mL). The reaction mixture was heated with stirring at 120 °C for 6 h, cooled to ~20 °C, and poured into water (200 mL). The finely dispersed precipitate that formed was separated and dried *in vacuo*. Pyrimidoindole **7** was obtained in a yield of 0.29 g (95%), m.p. 238—243 °C (from xylene). Found (%): C, 66.74; H, 3.56; N, 28.85. $C_{16}H_{10}N_6$. Calculated (%): C, 67.12; H, 3.52; N, 29.36. MS, *m/z* (I_{rel} (%)): 286 $[M]^+$ (23); 258 $[M - 28]^+$ (100). IR, ν/cm^{-1}: 2137 (N_3); 3414 (NH).

2-Morpholino-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (8) was prepared analogously to compound **7** from pyrimidoindole **4** (0.3 g, 1.1 mmol) and morpholine (0.29 g, 3.3 mmol) in a yield of 0.31 g (86%), m.p. 254—258 °C (from xylene). Found (%): C, 72.30; H, 5.38; N, 16.69. $C_{20}H_{18}N_4O$. Calculated (%): C, 72.70; H, 5.49; N, 16.96. MS, *m/z* (I_{rel} (%)): 330 $[M]^+$ (80); 329 $[M - 1]^+$ (19); 300 $[M - 30]^+$ (40); 299 $[M - 31]^+$ (100).

2-Cyano-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (9) was prepared analogously to compound **7** from pyrimidoindole **4** (0.2 g, 0.7 mmol) and KCN (0.14 g, 2.1 mmol) in a yield of 0.17 g (91%), m.p. 316—318 °C (from xylene). Found (%): C, 74.97; H, 3.66; N, 21.16. $C_{17}H_{10}N_4$. Calculated (%): C, 75.54; H, 3.73; N, 20.73. MS, *m/z* (I_{rel} (%)): 270 $[M]^+$ (55); 269 $[M - 1]^+$ (100). IR, ν/cm^{-1}: 2243 (C≡N); 3400, 3500 (NH).

Reaction of 2-chloro-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (4) with hydrazine hydrate. ***A.*** Hydrazine hydrate (99%, 0.13 g, 2.6 mmol) was added to a solution of pyrimidoindole **4** (0.36 g, 1.3 mmol) in dry DMSO (20 mL). The reaction mixture was heated with stirring at 100—110 °C for 7 h. Then hydrazine hydrate (0.5 g, 10 mmol) was added and the reaction mixture was heated until the starting compound **4** was consumed (9 h), after which the reaction mixture was cooled and poured into water (200 mL). The precipitate that formed was separated, washed with water until the washings became neutral, and dried *in vacuo*. A crude product was obtained in a yield of 0.32 g, m.p. 200—300 °C.

The product (79 mg) was chromatographed on Silufol plates ($CHCl_3$—AcOEt, 1 : 1, as the eluent). The individual compounds were eluted from different zones with EtOH. **Aminopyrimidoindole 1c** was isolated from the lower zone (R_f 0.30) in a yield of 23 mg (29%). The IR spectrum and m.p. of this compound were identical with those **(6)** of an authentic sample. **4-Phenyl-9*H*-pyrimido[4,5-*b*]indole** was isolated from the upper zone (R_f 0.70) in a yield of 11 mg (14%), m.p. 215—220 °C. High-resolution mass spectrum, found: *m/z* 245.0942 $[M]^+$. $C_{16}H_{11}N_3$. Calculated: M = 245.0953. ^{1}H NMR (400 MHz), δ: 7.19 (td, 1 H, H(6), ^3J = 7.5 Hz, ^4J = 1.1 Hz); 7.52 (td, 1 H, H(7), ^3J = 7.7 Hz, ^4J = 0.9 Hz); 7.60 (d, 1 H, H(8), ^3J = 8.0 Hz); 7.62—7.67 (m, 3 H, Ph); 7.80 (d, 1 H, H(5), ^3J = 8.0 Hz); 7.88—7.94 (m, 2 H, Ph); 8.98 (s, 1 H, H(2)); 12.52 (br.s, 1 H, NH). ^{13}C NMR (50 MHz), δ: 110.3 (C(4a)); 112.0 (C(8)); 118.6 (C(4b)); 120.7 (C(6)); 122.0 (C(5)); 127.7 (C(7)); 128.7 (C_m, Ph); 128.7 (C_o, Ph); 130.0 (C_p, Ph); 138.3, 138.6 (C_i, C(8a)); 154.2 (C(2)); 156.1 (C(9a)); 159.0 (C(4)).

B. Hydrazine hydrate (99%, 46 mg, 0.9 mmol) was added to a solution of pyrimidoindole **4** (84 mg, 0.3 mmol) in dry *N*-methylpyrrolidone (6 mL). The reaction mixture was heated with stirring at 100—110 °C for 17 h. Then hydrazine hydrate (100 mg, 2 mmol) was added and the reaction mixture was heated until the starting compound **4** was consumed (13 h), after which the reaction mixture was cooled, poured into water (100 mL), and extracted with AcOEt (4×25 mL). The combined extracts were washed with water and concentrated to dryness under reduced pressure. The residue was chromatographed on Silufol plates ($CHCl_3$—EtOAc, 1 : 3, as the eluent). Compound **1c** was isolated in a yield of 15 mg (20%) and compound **6** was isolated in a yield of 5 mg (7%) as described above.

2-Nitramino-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (10). Nitroguanidine (1.5 g, 14.4 mmol) was added to a solution of MeONa, which was prepared from Na (1.0 g, 43 mmol) in MeOH (50 mL). The reaction mixture was refluxed for 1 h and cooled to ~20 °C. Then tetrafluoroborate **2** [5] (4.9 g, 14.2 mmol) was added. The reaction mixture was refluxed with stirring for 5 h and cooled. The precipitates of $NaBF_4$ and unconsumed nitroguanidine were filtered off. The methanolic filtrate was acidified with concentrated HCl to pH 5. The precipitate that formed was separated, washed with water and EtOH (5 mL), and dried *in vacuo*. Compound **10** was obtained in a yield of 1.7 g (39%). The sample for analysis was crystallized from DMF. According to TLC (R_f 0.40, AcOEt—$CHCl_3$, 1 : 1, as the eluent), the product was pure but it did not have a sharp m.p. (noticeable melting of the crystals with decomposition was observed at 280—320 °C). Found (%): C, 62.94; H, 3.77; N, 22.63. $C_{16}H_{11}N_5O_2$. Calculated (%): C, 62.90; H, 3.60; N, 22.90. MS, *m/z* (I_{rel} (%)): 305 $[M]^+$ (8); 259 $[M - 46]^+$ (100). IR, ν/cm^{-1}: 1353 (NO_2); 3230, 3240 (NH). ^{1}H NMR (500 MHz), δ: 7.20 (t, 1 H, H(6), ^3J = 8.0 Hz); 7.51 (t, 1 H, H(7), ^3J = 8.0 Hz); 7.56 (d, 1 H, H(8), ^3J = 8.0 Hz); 7.66—7.69 (m, 3 H, Ph); 7.71 (d, 1 H, H(5), ^3J = 8.0 Hz); 7.90—7.94 (m, 2 H, Ph); 12.76 (br.s, 1 H, NH); 14.20—14.70 (br.s, 1 H, NH). ^{13}C NMR (50 MHz), δ: 108.05 (C(4a)); 112.2 (C(8)); 118.8 (C(4b)); 121.3 and 121.6 (C(5), C(6)); 127.6 (C(7)); 128.6 and 128.7 (C_o, C_m, Ph); 130.7 (C_p, Ph); 136.0 (C_i, Ph); 139.4 (C(8a)); 152.6 (C(9a)); 157.5 and 158.0 (C(2), C(4)).

Hydrogenation of 2-nitramino-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (10). A weighed sample of 2-nitraminopyrimidoindole **10** (0.2 g, 0.65 mmol) was hydrogenated over 4% Pd/C in EtOH (20 mL) at 35—40 °C until the starting compound disappeared (TLC control). The catalyst was separated, the solution was concentrated to dryness under reduced pressure, and the residue (104 mg) was chromatographed on Silufol plates ($CHCl_3$—EtOAc, 1 : 3). 2-Aminopyrimidoindole **1c** was isolated from the lower zone (R_f 0.30) in a yield of 79 mg (47%). Pyrimidoindole **6** was isolated from the medium zone (R_f 0.60) in a yield of 12 mg (7.5%). **2-Ethoxy-4-phenyl-9*H*-pyrimido[4,5-*b*]indole (11)** was isolated from the upper zone (R_f 0.80) in a yield of 11 mg (6%), m.p. 245—255 °C. High-resolution mass spectrum, found: *m/z* 289.12149 $[M]^+$. $C_{18}H_{15}N_3O$. Calculated: M = 289.12150. ^{1}H NMR (200 MHz), δ: 1.39 (t, 3 H, Me, ^3J = 7.0 Hz); 4.46 (q, 2 H, CH_2, ^3J = 7.0 Hz); 7.12 (td, 1 H, H(6), ^3J = 7.5 Hz, ^4J = 1.1 Hz); 7.40 (td, 1 H, H(7), ^3J = 8.0 Hz, ^4J = 0.7 Hz); 7.48 (d, 1 H, H(8), ^3J = 7.5 Hz); 7.60—7.65 (m, 3 H, Ph); 7.67 (d, 1 H, H(5), ^3J = 7.5 Hz); 7.87—7.93 (m, 2 H, Ph); 12.24 (br.s, 1 H, NH). ^{13}C NMR (50 MHz), δ: 14.5 (Me); 62.5 (CH_2); 105.9 (C(4a)); 111.6 (C(8)); 119.2 (C(4b)); 120.5 and 120.8 (C(5), C(6)); 126.2 (C(7)); 128.6 (C_o, C_m, Ph); 130.0 (C_p, Ph); 138.1 (C_i, Ph); 138.4 (C(8a)); 158.9 (C(9a)); 160.8 (C(4)); 162.6 (C(2)).

References

1. PCT Int. Appl. 93 20078; *Chem. Abstr.*, 1994, **121**, 134139.
2. PCT Int. Appl. 96 26941; *Chem. Abstr.*, 1996, **125**, 275901.
3. E. G. Paronikyan and F. S. Noravyan, *Khim. Geterotsikl. Soedin.*, 1996, 1413 [*Chem. Heterocycl. Compd.,* 1996, **32** (Engl. Transl.)].
4. S. Hess, C. E. Mueller, W. Frobenius, U. Reith, K.-N. Klotz, and K. Ego, *J. Med. Chem.*, 2000, **43**, 4636; *Chem. Abstr.*, 2001, **134**, 100695.
5. V. P. Borovik, L. S. Filatova, and V. P. Mamaev, *Izv. SO Akad. Nauk SSSR, Ser. Khim.*, 1975, **3**, 137 [*Bull. Sib. Branch Russ. Acad. Sci., Div. Chem. Sci.*, 1975 (Engl. Transl.)].
6. B. V. Nekrasov, *Osnovy obshchei khimii* [*Fundamentals of General Chemistry*], Khimiya, Moscow, 1965, 408 pp. (in Russian).
7. V. P. Borovik, I. Yu. Bagryanskaya, Yu. V. Gatilov, and O. P. Shkurko, *Zh. Strukt. Khim.,* 2003, **44**, № 3 [*Russ. J. Struct. Chem.*, 2003, **44**, No. 3 (Engl. Transl.)].
8. K. Sirakawa, *J. Pharm. Soc. Jpn.,* 1959, **79**, 1477; *Chem. Abstr.*, 1960, **54**, 11038.
9. A. V. Ivashchenko, O. N. Garicheva, L. V. Shmelev, Yu. S. Ryabokobylko, and G. M. Adamova, *Khim. Geterotsikl. Soedin.*, 1980, 1673 [*Chem. Heterocycl. Compd.,* 1980, **16** (Engl. Transl.)].
10. G. G. Moskalenko and O. P. Shkurko, *Khim. Geterotsikl. Soedin.*, 1997, 962 [*Chem. Heterocycl. Compd.,* 1997, **33** (Engl. Transl.)].
11. I. W. Southon and W. Pfleiderer, *Chem. Ber.*, 1978, **111**, 1006.
12. A. V. Ivashchenko and O. N. Garicheva, *Khim. Geterotsikl. Soedin.*, 1982, 579 [*Chem. Heterocycl. Compd.,* 1982, **18** (Engl. Transl.)].
13. V. P. Borovik, M. M. Shakirov, and O. P. Shkurko, *Khim. Geterotsikl. Soedin.*, 2003, № 1 [*Chem. Heterocycl. Compd.,* 2003, No. 1 (Engl. Transl.)].

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