Cyclization of 1-acylaminoacridones into 7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-ones

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1-Acylamino-10-methylacridones in a polar aprotic solvent underwent base-catalyzed cyclization into the corresponding 7-methyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-ones. Heating of 1-butylaminoacridone in acetic anhydride in the presence of *p*-toluenesulfonic acid and potassium acetate afforded 3-butyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one, while heating of 1-aminoacridone under the same conditions gave 9-acetoxy-1-acetylimino-1,10-dihydroxyacridine.

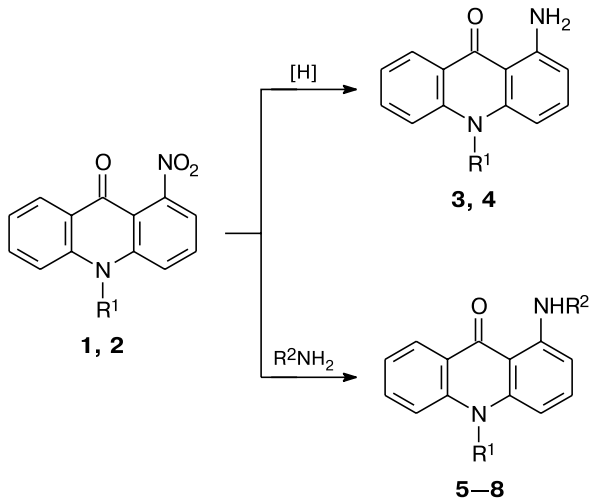
Key words: heterocyclization, pyridoacridines, acridones.

A large group of alkaloids with high biological (cytotoxic, anticancer, and antiviral) activity belongs to the pyrido[2,3,4-*kl*]acridine series.[1—4] For this reason, much attention is being given to methods for the synthesis of this polycyclic system. Pyridoacridines are most often prepared by construction of the acridine nucleus from quinoline derivatives.[5—7] The use of compounds with the acridine moiety as starting materials has been less studied.

Here we studied intramolecular cyclization of 1-acetylaminoacridones into 7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-ones. The possibility of the cyclization of 1-acylaminoacridones with participation of the *peri*-CO-group has not been investigated hitherto, although similar *peri*-cyclization of 1-acylaminoanthraquinones is widely used in the synthesis of pyridoanthrones.[8] Since this reaction involves a nucleophilic attack on the C(9) atom, replacement of the electron-withdrawing carbonyl group in position 10 by a nitrogen atom (when moving from anthraquinones to acridones) will inevitably have a deactivating effect. The problem is how strong this effect is and whether it hinders the cyclization. The failure in the synthesis of pyridoacridine by the Friedlaender reaction of 1-amino-4-methylacridone with acetylacetone have been attributed[9] to the conjugation of the imino group with the carbonyl one.

We investigated 1-aminoacridones containing an unsubstituted or substituted acetyl group at the primary and secondary N atoms. The starting 1-nitroacridone (**1**)[10,11] was methylated to give 10-methyl-1-nitroacridone (**2**). Reduction of nitro compounds **1** and **2** yielded 1-aminoacridone (**3**) and 1-amino-10-methylacridone (**4**), respectively.[12] Both nitroacridone **2**, which is nonionizable, and nitroacridone **1** can undergo aromatic nucleophilic displacement of the nitro group. Their reactions at elevated pressure and temperature gave new NHR^2-containing acridones **5**—**8** (Scheme 1).

Scheme 1



R^1 = H (**1, 3, 5**), Me (**2, 4, 6—8**);
R^2 = Bu^n (**5, 6**), *cyclo*-C_6H_{11} (**7**), 3-MeC_6H_4 (**8**)

1-Aminoacridones **3**—**8** were *N*-acylated with acetic anhydride (**a**), chloroacetyl chloride (**b**), ethyl cyanoacetate (**c**), and ethyl malonate (**d**) to give derivatives **3´**—**8´**. The acetylation of secondary amines **5**—**8** with acetic anhydride in the absence of any catalyst occurred at different rates. For instance, butylaminoacridones **5** and **6** were successfully acetylated in boiling

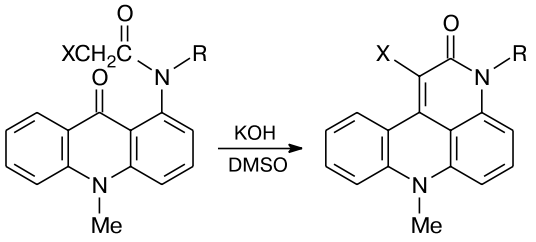
Published in Russian in *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 8, pp. 1433—1437, August, 2006.

1066-5285/06/5508-1487

acetic anhydride, while for cyclohexylaminoacridone **7** and toluidinoacridone **8**, heating at 200 °C for 9 h was required to complete the reaction.

We found that 1-acylamino-10-methylacridones obtained from primary acridinylamines undergo base-catalyzed cyclization into pyrido[2,3,4-*kl*]acridinones only when they contain an activating substituent (X = CN, COOEt) in the acyl group (Scheme 2). For instance, treatment of 1-cyanoacetylamino-10-methylacridone (**4′c**) with NaOH in aqueous DMF or DMSO at 30—40 °C gave 1-cyano-7-methyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (**9c**) in nearly quantitative yield, while 1-acetylamino-10-methylacridone (**4′a**) containing no activating substituent was only deacylated under these conditions.

Scheme 2



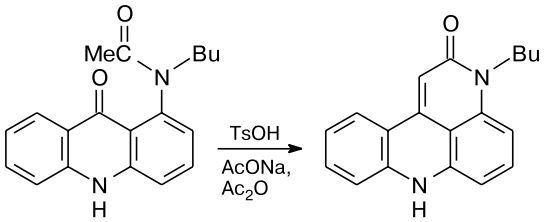
4′c,d; 6′a,b; 7′a,b; 8′a,b **9c,d; 10a,b; 11a,b; 12a,b**

R = H (**4′, 9**), Bu (**6′, 10**), *cyclo*-C_6H_{11} (**7′, 11**), 3-MeC_6H_4 (**8′, 12**)
X = H (**a**), Cl (**b**), CN (**c**), COOEt (**d**)

Compounds derived from secondary acridinylamines underwent cyclization even without an activating substituent in the acyl group. *N*-Acetylated 1-alkylaminoacridones **6′a** and **7′a** and 1-arylaminoacridone **8′a** were converted into 3-substituted pyrido[2,3,4-*kl*]acridinones **10a**—**12a** in high yields by stirring with KOH in DMSO at 80 to 130 °C (see Scheme 2). Apparently, this cyclization is favored by steric factors because alkyl or aryl substituent at the N atom facilitates the approach of the carbanionic center of the acetyl group to the CO group to be attacked. Chloroacetyl derivatives **6′b**—**8′b** of the same amines underwent cyclization into the corresponding 1-chloropyridoacridinones **10b**—**12b** even at 30 °C. In reactions of compounds **10b** and **12b** with potassium butoxide or butylamine, the attempted replacement of the Cl atom in position 1 by the alkoxy or alkylamino group failed: under mild conditions, these compounds remained unchanged, while at elevated temperature, the Cl atom was replaced by hydrogen.

Unlike N(10)-substituted derivatives, 10-unsubstituted 1-acylaminoacridones **3′a** and **5′a** turned to be reluctant to base-catalyzed cyclization. For this reason, we studied their cyclization in the presence of an acid. Heating of aminoacridone **5′a** with *p*-toluenesulfonic acid in acetic anhydride followed by addition of potassium acetate gave 3-butyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (**13**) in ~80% yield (Scheme 3). Without MeCOOK, the starting reagent was recovered only. Apparently, *p*-toluenesulfonic acid activates the substrate molecule by protonation, while the basic acetate anion binds the proton upon a nucleophilic attack at position 9. In the synthesis of compound **13**, it is expedient to start directly from amine **5**, which will be acetylated in acetic anhydride at the initial step. As expected, this method is also applicable to the cyclization of 10-methyl derivatives **4′a** and **6′a**—**8′a**. This method allows "one-pot" synthesis of some pyridoacridones directly from aminoacridones without isolating their acyl derivatives. Indeed, heating of 1-butylamino-10-methylacridone **6** in acetic anhydride with *p*-toluenesulfonic acid and then with potassium acetate yielded pyridoacridinone **10a**, which was identical with the product obtained by the base-catalyzed cyclization of compound **6′a**.

Scheme 3

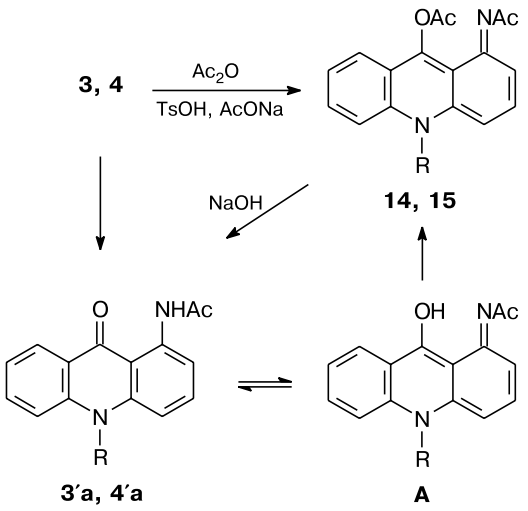


5′a **13**

Under analogous conditions, aminoacridone **3** containing a primary amino group formed a mixture of monoacetyl derivative **3′a** and diacetylated product **14**; the latter can be deacetylated to give compound **3′a** (Scheme 4). Similar diacetyl derivative **15** was obtained from 10-methyl analog **4′a**, for which acetylation at the ring N atom is excluded. The choice between the structures of *N,N*- and *N,O*-diacetyl derivatives in favor of the latter was made from mass and IR spectroscopic data. The mass spectrum of *N,O*-diacetyl derivative **14** contains a very intense peak of the $[M - CH_3COOH]^+$ ion due to the elimination of acetic acid but shows virtually no molecular ion peak, which is inherent in acetic acid esters. The peaks of the $[M - CH_2CO]^+$ and $[M - CH_3COOH - CH_2CO]^+$ ions are due to the elimination of ketene characteristic of acetamides.[13] The IR spectrum of *N,O*-diacetyl derivative **15** shows, along with the band due to the ν_{CO} stretching vibrations in the amide fragment at 1700 cm^{-1}, the longer-wavelength band of the ester group at 1719 cm^{-1}, while the spectrum of monoacetyl derivative **4′a** exhibits a band at 1690 cm^{-1}.

The formation of the *N,O*-diacetyl derivative suggests possible tautomerization of *N*-acetylated 1-amino-

Scheme 4



R = H (**3**, **3′a**, **14**), Me (**4**, **4′a**, **15**)

acridones into 9-hydroxyacridin-1-imines **A** (see Scheme 4).

Thus, the presented *peri*-cyclization of 1-acetylaminoacridones affords promising 1- and 3-substituted and -unsubstituted 7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-ones.

Experimental

Mass spectra were recorded on a Kratos MS 30 instrument (EI, 70 eV, ionization chamber temperature 250 °C, direct inlet probe). IR spectra were recorded on an FSM 1201 FTIR spectrometer (KBr pellets). The course of the reactions was monitored and the purity of the products obtained was checked by TLC on Silufol UV 254 plates. Preparative separation was carried out by column chromatography on silica gel (60/100 μm) with $CHCl_3$ as an eluent. Samples for elemental analysis were dried *in vacuo* over P_2O_5 at 70—80 °C. 1-Nitroacridone (**1**), 1-aminoacridone (**3**),[10,11] 10-methyl-1-nitroacridone (**2**),[12] and 1-amino-10-methylacridone (**4**)[12] were prepared as described earlier.

Reactions of 1-nitroacridone (1) and 10-methyl-1-nitroacridone (2) with amines (general procedure). A mixture of nitroacridone (10 mmol) and an amine (20 g) was heated without stirring in a 100-mL steel autoclave on a Wood′s alloy bath.

1-Butylaminoacridone (5) was obtained by heating nitroacridone **1** and *n*-butylamine (4 h, 180 °C). On cooling, the reaction mixture was poured into water (200 mL) and the resulting precipitate was separated, washed with water, and dried. The yield was 2.10 g (78%), m.p. 196 °C (from aqueous Pr^iOH). Found (%): C, 76.09; H, 6.80; N, 10.24. $C_{18}H_{20}N_2O$. Calculated (%): C, 76.66; H, 6.81; N, 10.52. IR, ν/cm^{-1}: 1624 (CO); 3186, 3275 (NH).

1-Butylamino-10-methylacridone (6) was obtained analogously from nitroacridone **2**. The yield was 2.40 g (85%), m.p. 82 °C (from aqueous Pr^iOH). Found (%): C, 77.35; H, 6.91; N, 9.88. $C_{18}H_{20}N_2O$. Calculated (%): C, 77.11; H, 7.19; N, 9.99. IR, ν/cm^{-1}: 1624 (CO); 3444 (NH).

1-Cyclohexylamino-10-methylacridone (7) was obtained by heating nitroacridone **2** with cyclohexylamine (16 h, 200 °C). On cooling, DMF (15 mL) was added and the mixture was poured into water. The resulting precipitate was separated, washed with water, dried, and chromatographed. The second yellow zone was collected. The yield was 1.75 g (57%), m.p. 150—152 °C (from aqueous Pr^iOH). Found (%): C, 78.88; H, 7.16; N, 9.16. $C_{20}H_{22}N_2O$. Calculated (%): C, 78.40; H, 7.24; N, 9.14. IR, ν/cm^{-1}: 1624 (CO); 3440 (NH).

10-Methyl-1-(3-methylphenylamino)acridone (8) was obtained by heating nitroacridone **2** with *m*-toluidine (9 h, 300 °C). After the reaction was completed, the excess of *m*-toluidine was steam-distilled. The oily residue was dissolved in $CHCl_3$ and chromatographed. The yield was 1.83 g (58%), m.p. 155 °C (from AcOH). Found (%): C, 79.96; H, 5.48; N, 8.82. $C_{21}H_{18}N_2O$. Calculated (%): C, 80.23; H, 5.77; N, 8.91. IR, ν/cm^{-1}: 1623 (CO); 3446 (NH).

Acetylation of aminoacridones with acetic anhydride (general procedure). Aminoacridone (2 mmol) was treated with acetic anhydride (10 mL), the mixture was poured into water (100 mL), and the precipitate was separated. Reactions with primary amines **3** and **4** were carried out at ~20 °C for 1 h, reactions with amines **5** and **6** under reflux, and reactions with amines **7** and **8** at 180—200 °C for 8—10 h.

1-Acetylaminoacridone (3′a) was obtained from aminoacridone **3**. The yield was 0.34 g (67%), m.p. 330 °C (decomp.) (from Bu^nOH). Found (%): C, 71.34; H, 4.84; N, 10.79. $C_{15}H_{12}N_2O_2$. Calculated (%): C, 71.42; H, 4.79; N, 11.10. IR, ν/cm^{-1}: 1640, 1663 (CO); 3277, 3442 (NH).

1-Acetylamino-10-methylacridone (4′a) was obtained from aminoacridone **4**. The yield was 0.40 g (75%), m.p. 192 °C (from aqueous EtOH). Found (%): C, 71.96; H, 5.22; N, 10.23. $C_{16}H_{14}N_2O_2$. Calculated (%): C, 72.17; H, 5.30; N, 10.52. IR, ν/cm^{-1}: 1619, 1689 (CO); 3448 (NH).

1-(*N*-Acetyl-*N*-butylamino)acridone (5′a) was obtained from amine **5**. The yield was 0.60 g (97%), m.p. 248 °C (from Bu^nOH). Found (%): C, 73.71; H, 6.62; N, 8.87. $C_{19}H_{20}N_2O_2$. Calculated (%): C, 74.00; H, 6.54; N, 9.08. IR, ν/cm^{-1}: 1605, 1639 (CO); 3186 (NH).

1-(*N*-Acetyl-*N*-butylamino)-10-methylacridone (6′a) was obtained from acridone **6**. The yield was 0.52 g (81%), m.p. 160 °C (from aqueous Pr^iOH). Found (%): C, 74.27; H, 6.85; N, 8.65. $C_{20}H_{22}N_2O_2$. Calculated (%): C, 74.51; H, 6.88; N, 8.69. IR, ν/cm^{-1}: 1629, 1656 (CO).

1-(*N*-Acetyl-*N*-cyclohexylamino)-10-methylacridone (7′a) was obtained by heating acridone **7** with acetic anhydride (200 °C, 8 h). The yield was 0.47 g (67%), m.p. 240 °C (from Pr^iOH). Found (%): C, 74.55; H, 6.73; N, 8.09. $C_{22}H_{24}N_2O_2$. Calculated (%): C, 75.83; H, 6.94; N, 8.04. IR, ν/cm^{-1}: 1631, 1647 (CO).

1-[*N*-Acetyl-*N*-(3-methylphenyl)amino]-10-methylacridone (8′a) was obtained by heating acridone **8** with acetic anhydride (180 °C, 10 h). The yield was 0.570 g (80%), m.p. 178 °C (from aqueous Pr^iOH). Found (%): C, 77.67; H, 5.65; N, 7.71. $C_{23}H_{20}N_2O_2$. Calculated (%): C, 77.51; H, 5.66; N, 7.86. IR, ν/cm^{-1}: 1631, 1666 (CO).

Acylation of aminoacridones with chloroacetyl chloride (general procedure). A mixture of aminoacridone (2 mmol) and chloroacetyl chloride (0.76 mL, 10 mmol) in benzene (10 mL) was refluxed for 30 min. Hexane (8 mL) was added and the

resulting precipitate was separated and washed with benzene and hexane.

1-(*N*-Butyl-*N*-chloroacetylamino)-10-methylacridone (6´b) was obtained from amine **6**. The yield was 0.52 g (72%), m.p. 195 °C (from aqueous Pr^{i}OH). Found (%): C, 67.01; H, 5.95; Cl, 10.09; N, 7.76. $C_{20}H_{21}ClN_2O_2$. Calculated (%): C, 67.32; H, 5.93; Cl, 9.93; N, 7.85. IR, ν/cm^{-1}: 1627, 1665 (CO).

1-(*N*-Chloroacetyl-*N*-cyclohexylamino)-10-methylacridone (7´b) was obtained from amine **7**. The yield was 0.56 g (73%), m.p. 268 °C (from EtOH). Found (%): C, 69.57; H, 6.03; Cl, 9.15; N, 7.29. $C_{22}H_{23}ClN_2O_2$. Calculated (%): C, 69.01; H, 6.05; Cl, 9.26; N, 7.32. IR, ν/cm^{-1}: 1627, 1660 (CO).

1-[*N*-Chloroacetyl-*N*-(3-methylphenyl)amino]-10-methylacridone (8´b) was obtained from amine **8**. The yield was 0.39 g (50%), m.p. 98 °C (from Pr^{i}OH). Found (%): C, 70.65; H, 4.57; Cl, 9.03; N, 7.02. $C_{23}H_{19}ClN_2O_2$. Calculated (%): C, 70.68; H, 4.90; Cl, 9.07; N, 7.17. IR, ν/cm^{-1}: 1627, 1694 (CO).

1-Cyanoacetylamino-10-methylacridone (4´c). A mixture of aminoacridone **4** (0.56 g, 2.5 mmol) and ethyl cyanoacetate (10 mL) was heated at 180 °C for 1 h. On cooling, EtOH (50 mL) was added and the resulting precipitate was filtered off and washed with EtOH. The yield was 0.43 g (74%), m.p. 220 °C (from Bu^{n}OH). Found (%): C, 69.90; H, 4.37; N, 14.57. $C_{17}H_{13}N_3O_2$. Calculated (%): C, 70.09; H, 4.50; N, 14.42. IR, ν/cm^{-1}: 1618, 1695 (CO); 2260 (CN); 3424 (NH).

1-(Ethoxycarbonylacetylamino)-10-methylacridone (4´d) was obtained by heating aminoacridone **4** (0.67 g, 3 mmol) in ethyl malonate (10 mL) (155 °C, 5 h). The yield was 0.270 g (40%), m.p. 145 °C (from EtOH). Found (%): C, 67.75; H, 5.16; N, 8.62. $C_{19}H_{18}N_2O_4$. Calculated (%): C, 67.45; H, 5.36; N, 8.28. IR, ν/cm^{-1}: 1619, 1684, 1737 (CO); 3449 (NH).

Base-catalyzed cyclization of *N*-acetyl derivatives of 1-amino-10-methylacridones 4´ and 6´—8´ (general procedure). A mixture of an acetyl derivative (2 mmol) and powdered KOH (1 g, 18 mmol) in DMSO (20 mL) was heated at 80 °C for 2 h and poured into water (100 mL). The resulting precipitate was separated and washed with water.

1-Cyano-7-methyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (9c) was obtained from compound **4´c**. The yield was 0.51 g (93%). Stirring of compound **4´c** (2 mmol) in a mixture of DMF (50 mL) and aqueous 30% NaOH (17 mL) at 40 °C for 3 h gave pyridoacridine **9c** in approximately the same yield. The product did not melt below 340 °C (from EtOH). Found (%): C, 74.72; H, 4.05; N, 15.75. $C_{17}H_{11}N_3O$. Calculated (%): C, 74.71; H, 4.06; N, 15.38. IR, ν/cm^{-1}: 1651 (CO); 2200 (CN); 3449 (NH). MS, *m/z* (I_{rel} (%)): 273 $[M]^+$ (100); 258 $[M - Me]^+$ (45), 230 $[M - Me - CO]^+$ (21).

1-Ethoxycarbonyl-7-methyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (9d) was obtained from compound **4´d**. The yield was 0.486 g (76%), m.p. 304 °C (from EtOH). Found (%): C, 71.10; H, 5.33; N, 8.98. $C_{19}H_{16}N_2O_3$. Calculated (%): C, 71.24; H, 5.03; N, 8.74. IR, ν/cm^{-1}: 1622, 1637 (CO).

3-Butyl-7-methyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (10a) was obtained from compound **6´a**. The yield was 0.475 g (78%), m.p. 158—160 °C (from aqueous Pr^{i}OH). Found (%): C, 78.68; H, 6.57; N, 9.13. $C_{20}H_{20}N_2O$. Calculated (%): C, 78.92; H, 6.62; N, 9.20. IR, ν/cm^{-1}: 1629 (CO). MS, *m/z* (I_{rel} (%)): 304 $[M]^+$ (61); 248 $[M - C_4H_8]^+$ (100).

3-Butyl-1-chloro-7-methyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (10b) was obtained from compound **6´b**. The yield was 0.570 g (83%), m.p. 127 °C (from benzene—hexane). Found (%): C, 70.95; H, 5.55; Cl, 10.20; N, 8.29. $C_{20}H_{19}ClN_2O$. Calculated (%): C, 70.90; H, 5.65; Cl, 10.46; N, 8.27. IR, ν/cm^{-1}: 1629 (CO).

Heating of 1-chloropyridoacridine **10b** (1 mmol) at 80 °C for 10 h in BuOH (15 mL) containing BuOK (3 mmol) followed by steam distillation of BuOH and chromatography of the residue with $CHCl_3$ as an eluent gave pyridoacridine **10a** (0.120 g, 38%). The same compound was obtained by heating of 1-chloropyridoacridine **10b** (1 mmol) at 180 °C for 10 h in butylamine (10 mL) in the presence of $Cu(OAc)_2$.

3-Cyclohexyl-7-methyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (11a) was obtained from compound **7´a**. The yield was 0.463 g (70%), m.p. 308 °C (from EtOH). Found (%): C, 80.00; H, 6.93; N, 8.51. $C_{22}H_{22}N_2O$. Calculated (%): C, 79.97; H, 6.71; N, 8.48. IR, ν/cm^{-1}: 1629 (CO). MS, *m/z* (I_{rel} (%)): 330 $[M]^+$ (19); 248 $[M - C_6H_{10}]^+$ (100); 233 $[M - C_6H_{10} - Me]^+$ (15); 205 $[M - C_6H_{10} - Me - CO]^+$ (25).

1-Chloro-3-cyclohexyl-7-methyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (11b) was obtained from compound **7´b**. The yield was 0.672 g (92%), m.p. 237 °C (from aqueous Pr^{i}OH). Found (%): C, 72.53; H, 5.83; Cl, 9.50; N, 7.78. $C_{22}H_{21}ClN_2O$. Calculated (%): C, 72.42; H, 5.80; Cl, 9.72; N, 7.68. IR, ν/cm^{-1}: 1633 (CO).

7-Methyl-3-(3-methylphenyl)-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (12a) was obtained from compound **8´a**. The yield was 0.589 g (87%), m.p. 260 °C (from aqueous Pr^{i}OH). Found (%): C, 82.07; H, 5.81; N, 7.91. $C_{23}H_{18}N_2O$. Calculated (%): C, 81.63; H, 5.36; N, 8.28. IR, ν/cm^{-1}: 1644 (CO).

1-Chloro-7-methyl-3-(3-methylphenyl)-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (12b) was obtained from compound **8´b**. The yield was 0.537 g (72%), m.p. 240 °C (from aqueous Pr^{i}OH). Found (%): C, 73.86; H, 4.52; Cl, 9.42; N, 7.43. $C_{23}H_{17}ClN_2O_3$. Calculated (%): C, 74.09; H, 4.60; Cl, 9.51; N, 7.51. IR, ν/cm^{-1}: 1633 (CO).

As in the case of compound **10b**, heating of compound **12b** with BuOK in butanol did not result in alkoxylation; instead, dechlorination occurred to give pyridoacridine **12a**.

Acid-catalyzed cyclization of *N*-acetyl derivatives of 1-aminoacridones. 3-Butyl-7*H*-pyrido[2,3,4-*kl*]acridin-2(3*H*)-one (13). *p*-Toluenesulfonic acid hydrate (0.95 g, 5 mmol) and anhydrous AcOK (4 g, 40 mmol) were added to a solution of 1-butylaminoacridone **5** or its *N*-acetyl derivative **5´a** (1 mmol) in acetic anhydride (10 mL). The reaction mixture was refluxed for 10 min, cooled, and mixed with water (100 mL). The resulting precipitate was filtered off, washed with water, and dried to give pyridoacridine **13** (0.205 g, 70%) as greenish yellow needles, m.p. 268 °C (from aqueous Pr^{i}OH). Found (%): C, 78.37; H, 6.16; N, 9.59. $C_{19}H_{18}N_2O$. Calculated (%): C, 78.59; H, 6.25; N, 9.62. IR, ν/cm^{-1}: 1626 (CO); 3269 (NH).

Organic material was extracted from the filtrate with CH_2Cl_2 (3×25 mL). The organic layer was separated, dried over Na_2SO_4, and concentrated to recover acetyl derivative **5´a** (0.06 g). The yield of pyridoacridine **13** with respect to the consumed starting compound **5´a** was 87%.

Under analogous conditions, the cyclization of 1-butylamino-10-methylacridone **6** gave 3-butyl-7-methylpyridoacridine **10a**, which was isolated by extraction followed by chromatography. This product was identical with pyridoacridine **10a** obtained by the cyclization of acetyl derivative **6´a** in the presence of KOH.

9-Acetoxy-1-acetylimino-1,10-dihydroacridine (14). A solution of 1-acetylaminoacridone **3´a** (0.252 g, 1 mmol) in acetic anhydride (10 mL) was treated with *p*-toluenesulfonic acid and potassium acetate under the reaction conditions described for 3-butylpyridoacridine **13**. The mixture was diluted with water and the unreacted starting acridone **3´a** (0.170 g, 67%) was filtered off. The product from the filtrate was extracted with ethyl acetate. The organic layer was separated, dried with Na_2SO_4, and concentrated. The residue was chromatographed to collect at first unreacted acridone **3´a** and then fractions with diacetyl derivative **14**. Solutions of product **14** exhibited intense bluish violet fluorescence. The yield was 0.043 g (44%) with consideration of the recovery of compound **3´a**, m.p. 236—238 °C (from MeOH). Found (%): C, 68.93; H, 4.75; N, 9.54. $C_{17}H_{14}N_2O_3$. Calculated (%): C, 69.37; H, 4.79; N, 9.52. IR, ν/cm^{-1}: 1630 (C=N); 1675 w, 1691 (CO); 3297 (NH). MS, m/z (I_{rel} (%)): 294 $[M]^+$ (0.5), 252 $[M - CH_2CO]^+$ (34), 234 $[M - CH_3COOH]^+$ (100); 210 $[M - 2\ CH_2CO]^+$ (93), 182 $[M - 2\ CH_2CO - CO]^+$ (12), 165 $[M - 2\ CH_2CO - CO - NH_2]^+$ (36).

In aqueous methanol in the presence of NaOH, compound **14** changed into 1-acetylaminoacridone **3´a**.

9-Acetoxy-1-acetylimino-10-methyl-1,10-dihydroacridine (15) was obtained analogously from 1-amino-10-methylacridone (**4**) (0.224 g, 1 mmol) and isolated by column chromatography. The yield of acetylimine **15** was 0.041 g (13%), m.p. 203—205 °C (from MeOH). Found (%): C, 69.96; H, 5.47; N, 8.81. $C_{18}H_{16}N_2O_3$. Calculated (%): C, 70.12; H, 5.23; N, 9.09. IR, ν/cm^{-1}: 1630 (C=N); 1700, 1719 (CO).

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Received April 13, 2006;
in revised form June 9, 2006