Reaction of cross-conjugated dienones with phenyl- and 2-pyridylhydrazines

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New pyrazoline derivatives were synthesized by condensation of dienones based on cyclohexanone and *N*-substituted piperidin-4-ones with phenylhydrazine and 2-pyridylhydrazine. The reaction with phenylhydrazine produces *trans* isomers.

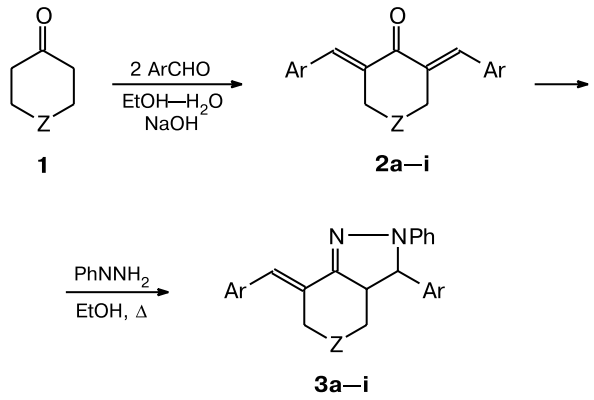
Key words: condensation, dienones, arylhydrazines, pyrazolines, 2-R-3-aryl-7-[(*E*)-arylmethylene]-3,3a,4,5,6,7-hexahydro-2*H*-indazoles, 5-R-3-aryl-7-[(*E*)-arylmethylene]-2-phenyl-3,3a,4,5,6,7-hexahydro-2*H*-pyrazolo[4,3-*c*]-pyridines.

The ability of α,β-unsaturated derivatives of cyclic ketones to add binucleophiles is widely used in organic synthesis. Due to the presence of the cross-conjugated dienone system, α,β-dienones are involved in heterocyclization reactions with nucleophilic reagents, such as urea,[1,2] thiourea,[3—9] cyanoacetamide,[3,9,10] and malononitrile.[11,12] The reactions of various mono- and dienones based on cyclic ketones with guanidine and its alkyl derivatives were studied.[13,14] The reactions of unsubstituted hydrazine with mono- and dienones produced pyrazoline derivatives.[15—17] There is evidence for the possibility of performing analogous reactions with hydrazine derivatives $RNH—NH_2$, where R are aliphatic[3,7,18,19,20] or aromatic[3,4,17,21,22] substituents.

In the last fifteen years, the chemistry of coordination polymers, which are infinite assemblies of metal ions and *exo*-dentate ligands, has attracted great interest. These structures were extensively studied as catalytic, luminescent, magnetic, and porous materials.[23] Recently,[24] we have proposed symmetrical cross-conjugated dienones containing pyridyl substituents for the use as *exo*-dentate ligands. Additional substituents, which are able to be coordinated to metal atoms, can be introduced into dienone structures by heterocyclization. This leads to changes in the dentation and directionality of the donor pairs of the ligand, which can influence the complexation and properties of the resulting supramolecular structures. In this connection, the aim of the present study was to synthesize pyrazoline derivatives containing pyridyl substituents in the dienone and/or arylhydrazine components. The goal was also to characterize the structures and stereochemistry of the products in detail.

Cyclic ketones **1** were used to prepare dienone substrates **2a—k** according to known procedures.[25,26] We synthesized a series of new bicyclic pyrazolines **3a—c** and **3e—i** by heterocyclization of dienone derivatives of cyclic ketones with phenylhydrazine (Scheme 1). Compound **3d** has been characterized earlier.[3] However, its structure has not been established by spectroscopic methods. Only the melting point, the molecular ion, and elemental analysis data were documented.[3] Heterocyclization was carried out in ethanol with the use of a fourfold excess of phenylhydrazine.[17]

Scheme 1



Z = CH_2, NMe, NCH_2Ph

	Z	Ar		Z	Ar
a	CH_2	4-C_5H_4N	**f**	NMe	3-C_5H_4N
b	CH_2	3-C_5H_4N	**g**	NCH_2Ph	Ph
c	NMe	Ph	**h**	NCH_2Ph	4-$MeOC_6H_4$
d	NMe	4-$MeOC_6H_4$	**i**	NCH_2Ph	3-C_5H_4N
e	NMe	4-C_5H_4N			

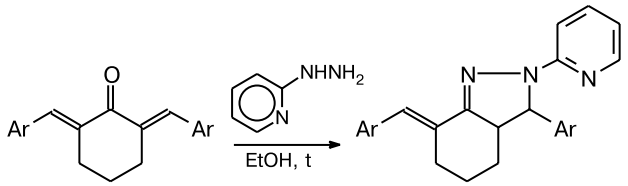
The course of the reaction is substantially influenced by the aromatic component of the starting dienones **2**. Heterocyclization of pyridine derivatives (Ar is 4-pyridyl or 3-pyridyl) occurs more rapidly compared to other benzene derivatives (Ar = Ph or 4-$MeOC_6H_4$). However,

Published in Russian in *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 11, pp. 2509—2512, November, 2005.

adducts of phenylhydrazine and dienones containing pyridine substituents do not crystallize from the reaction mixtures. They have to be isolated by column chromatography, and an optimal eluent should be chosen in each particular case. Pyrazolines based on phenyl-containing dienone derivatives are easily crystallizable compounds. All phenylhydrazine derivatives are rather stable although they slowly decompose in the light.

Under the same conditions, the reactions of the heteroaromatic hydrazine derivative (2-pyridylhydrazine) with dienones **2b** and **2j** produce pyrazoline derivatives **3j** and **3k** (Scheme 2).

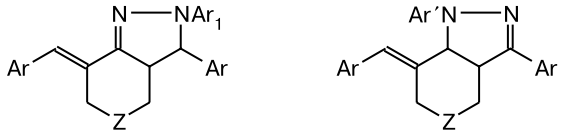
Scheme 2



2b,j **3j,k**

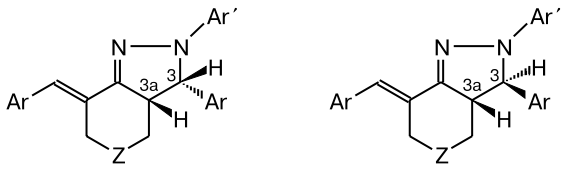
Ar = 3-C_5H_4N (**2b**, **3j**), Ph (**2j**, **3k**)

Condensation of unsymmetrical hydrazine derivatives with dienones can afford two isomeric products **A** and **B**.[27] Nuclear Overhauser effect measurements demonstrated that only product **A** was generated in all cases.



A **B**

Products **A** can exist as two diastereomers, *viz.*, *cis*-**A** and *trans*-**A**, which differ in the mutual arrangement of the protons H(3) and H(3a) in the pyrazoline ring.



cis-**A** *trans*-**A**

According to the results of the study,[28] the following three parameters should be analyzed to establish the stereochemistry of bicyclic pyrazolines: the chemical shifts of the protons H(3) and H(3a) and the vicinal coupling constant $^3J_{3,3a}$. In the case of the *cis* isomers, the chemical shifts of the protons H(3) and H(3a) are observed at δ 5.4—5.6 and 3.2—3.5, respectively. For the *trans* isomers, the signals for these protons are shifted upfield and are observed at δ 4.8—4.9 for H(3) and at δ 2.8—3.1 for H(3a). The coupling constants for these protons are least informative because these constants for the *cis* and *trans* isomers differ only slightly (for example, 10.8 and 10.5 Hz).

For pyrazoline derivatives **3a—i**, the chemical shifts for H(3) and H(3a) are observed at δ 4.5—4.9 and 3.0—3.5, respectively. Based on these data, we assigned the *trans* configuration to products **3a—i**. This conclusion is consistent with the data published recently.[29]

The available data did not allow us to unambiguously establish the stereochemistry of the addition products of 2-pyridylhydrazines **3j** and **3k** because the signals for H(3) are shifted downfield (δ 5.11 for **3j**, and δ 5.06 for **3k**) compared to those for the phenyl analogs. The chemical shifts of H(3a) are 3.05 and 2.95 ppm, respectively, and the coupling constants $^3J_{3,3a}$ are 11.6 and 11.4 Hz. Assuming that deshielding of the protons H(3) is associated with the electron-withdrawing effect of the pyridine ring, the *trans* configuration can also be assigned to compounds **3j** and **3k**.

Experimental

The ^{1}H NMR spectra were recorded at 28 °C on a Bruker-Avance-400 spectrometer operating at 400 MHz in $CDCl_3$. The chemical shifts (δ) are given relative to HMDS as the internal standard. Elemental analysis was carried out on a Carlo-Erba CHN-analyzer. The GL-mass spectrometric analysis was performed on a Finnigan MAT SSQ 7000 GL-mass spectrometer equipped with an OV-1 quartz capillary column (25 m); the energy of ionizing electrons was 70 eV; the temperature mode: 70 °C (2 min)→280 °C (10 min) at a heating rate of 20 K min^{-1}. The course of the reactions was monitored and the purities of the compounds were checked by TLC on Silufol plates with a fixed layer of silica gel. The preparative chromatographic separation of the reaction mixtures was carried out on silica gel columns (μ 5/40, Silica gel 60). The melting points were determined on a block in open capillary tubes. The uncorrected melting and boiling points are given.

Compounds **2a**, **2b**, **2e**, **2f** (see Ref. 25) and **2c**, **2d**, **2g**, **2h**, **2i**, **2j** (see Ref. 26) were synthesized according to known procedures.

Synthesis of compounds 3a—k. A solution of dienone (0.0025 mol) and arylhydrazine (0.01 mol) in ethanol (15 mL) was refluxed with stirring for 6 h. The completion of the reaction was monitored by TLC. The solution was cooled. The precipitate that formed was filtered off and recrystallized from ethanol. When precipitation did not occur, the solvent was evaporated, and the residue was purified on a silica gel chromatographic column.

2-Phenyl-3-(4-pyridyl)-7-[(*E*)-(4-pyridyl)methylene]-3,3a,4,5,6,7-hexahydro-2*H*-indazole (3a). The yield was 0.69 g (77%), ethyl acetate as the eluent, m.p. 95—98 °C (with decomp.). ^{1}H NMR, δ: 1.48—1.53 (m, 1 H, H(5)); 1.72—2.25 (m, 3 H, H(4), H(5), H(4)); 2.40 (t, 1 H, H(6), J = 12.3 Hz);

3.02—3.13 (m, 2 H, H(3a), H(6)); 4.60 (d, 1 H, H(3), J = 12.3 Hz); 6.72 (t, 1 H, *p*-H(Ph), J = 7.5 Hz); 7.03 (d, 2 H, *o*-H(Ph), J = 7.5 Hz); 7.19 and 7.21 (both d, 2 H each, β-H (pyridyl), J = 1.6 Hz); 7.26 (s, 1 H, —CH=); 7.51 (t, 2 H, *m*-H (Ph), J = 7.5 Hz); 8.61 and 8.65 (both d, 2 H each, α-H (pyridyl), J = 1.6 Hz). MS (EI, 70 eV), m/z (I_{rel} (%)): 366 $[M]^+$ (100), 288 (13), 183 (19). Found (%): C, 77.42; H, 6.94; N, 15.64. $C_{24}H_{22}N_4$. Calculated (%): C, 77.94; H, 6.26; N, 15.81.

2-Phenyl-3-(3-pyridyl)-7-[(*E*)-(3-pyridyl)methylene]-3,3a,4,5,6,7-hexahydro-2*H*-indazole (3b). The yield was 0.60 g (63%), ethyl acetate as the eluent, m.p. 110—114 °C (with decomp.). ^{1}H NMR, δ: 1.51—1.54 (m, 1 H, H(5)); 1.74 (quint., 1 H, H(4), J = 12.2 Hz, J = 2.2 Hz); 2.01—2.08 and 2.31—2.40 (both m, 1 H each, H(5), H(4)); 2.48 (t, 1 H, H(6), J = 12.4 Hz); 3.00—3.14 (m, 2 H, H(3a), H(6)); 4.68 (d, 1 H, H(3), J = 12.7 Hz); 6.88 (t, 1 H, *p*-H(Ph), J = 7.4 Hz); 7.16 (d, 2 H, *o*-H(Ph), J = 7.4 Hz); 7.18—7.33 (m, 5 H, *m*-H (Ph), β-H (pyridyl), –CH=); 7.66 and 8.49 (both dt, 1 H each, γ-H (pyridyl), J = 7.7 Hz, J = 4.9 Hz); 8.48 and 8.60 (both d, 1 H each, α-H (pyridyl), J = 4.9 Hz); 8.62 and 8.64 (both s, 1 H each, α′-H (pyridyl)). MS (EI, 70 eV), m/z (I_{rel} (%)): 366 $[M]^+$ (100), 288 (32), 183 (19). Found (%): C, 77.59; H, 6.97; N, 15.53. $C_{24}H_{22}N_4$. Calculated (%):C, 77.94; H, 6.26; N, 15.81.

5-Methyl-2,3-diphenyl-7-[(*E*)-phenylmethylene]-3,3a,4,5,6,7-hexahydro-2*H*-pyrazolo[4,3-*c*]pyridine (3c). The yield was 0.68 g (72%), m.p. 147—148 °C (from ethanol). ^{1}H NMR, δ: 2.39—2.44 (m, 4 H, H(4) and —NCH_3); 3.06 (d, 1 H, H(6), J = 13.6 Hz); 3.15 (dd, 1 H, H(4), J = 10.1 Hz, J = 6.4 Hz); 3.30—3.38 (m, 1 H, H(3a)); 3.97 (d, 1 H, H(6), J = 13.6 Hz); 4.60 (d, 1 H, H(3), J = 12.7 Hz); 6.79 (t, 1 H, *p*-H (Ph), J = 7.4 Hz); 6.89 (d, 2 H, *o*-H (Ph), J = 7.8 Hz); 7.01—7.40 (m, 13 H, H (Ph), —CH=). MS (EI, 70 eV), m/z (I_{rel} (%)): 379 $[M]^+$ (100), 335 (93), 259 (70), 180 (18), 115 (55), 91 (24), 77 (47). Found (%): C, 82.29; H, 6.75; N, 11.22. $C_{26}H_{25}N_3$. Calculated (%): C, 82.29; H, 6.64; N, 11.07.

3-(4-Methoxyphenyl)-7-[(*E*)-(4-methoxyphenyl)methylene]-5-methyl-2-phenyl-3,3a,4,5,6,7-hexahydro-2*H*-pyrazolo[4,3-*c*]pyridine (3d). The yield was 0.35 g (32%), m.p. 190—192 °C (from ethanol) (*cf.* lit. data[3]: m.p. 192—193 °C). ^{1}H NMR, δ: 2.34—2.40 (m, 4 H, H(4) and —NCH_3); 3.07 (dd, 1 H, H(6), J = 13.7 Hz, J = 2.3 Hz); 3.11 (dd, 1 H, H(4), J = 10.5 Hz, J = 6.7 Hz); 3.25—3.31 (m, 1 H, H(3a)); 3.79 and 3.81 (both s, 3 H each, —OCH_3); 4.07 (d, 1 H, H(6), J = 13.7 Hz); 4.52 (d, 1 H, H(3), J = 12.9 Hz); 6.79 (t, 1 H, *p*-H (Ph), J = 7.4 Hz); 6.89 (d, 2 H, *o*-H (Ph), J = 7.8 Hz); 7.04 and 7.13 (both d, 2 H each, *o*-H (4-$MeOC_6H_4$), J = 7.1 Hz); 7.15 (t, 2 H, *m*-H (Ph), J = 7.4 Hz); 7.24 (s, 1 H, —CH=); 7.32 and 7.40 (both d, 2 H each, *m*-H (4-$MeOC_6H_4$), J = 7.1 Hz). MS (EI, 70 eV), m/z (I_{rel} (%)): 439 $[M]^+$ (100), 395 (68), 334 (13), 121 (16), 77 (13). Found (%): C, 76.60; H, 6.52; N, 9.38. $C_{28}H_{29}N_3O_2$. Calculated (%): C, 76.50; H, 6.66; N, 9.56.

5-Methyl-2-phenyl-3-(4-pyridyl)-7-[(*E*)-(4-pyridyl)methylene]-3,3a,4,5,6,7-hexahydro-2*H*-pyrazolo[4,3-*c*]pyridine (3e). The yield was 0.30 g (31%), acetone as the eluent, m.p. 124—126 °C (with decomp.). ^{1}H NMR, δ: 2.49—2.53 (m, 1 H, H(4)); 2.56 (br.s, 3 H, NCH_3); 2.62—2.65 (m, 2 H, H(6), H(4)); 3.19—3.25 (m, 1 H, H(3a)); 4.49 (d, 1 H, H(6), J = 10.1 Hz); 4.89 (d, 1 H, H(3), J = 12.8 Hz); 7.12—7.30 (m, 7 H, *m*-H(Ph), *p*-H(Ph), β-H (pyridyl)); 7.58 and 7.68 (both d, 1 H each, *o*-H(Ph), J = 7.5 Hz); 8.10 (s, 1 H, —CH=); 8.37 and 8.46 (both d, 2 H each, α-H (pyridyl), J = 2.1 Hz). MS (EI, 70 eV), m/z (I_{rel} (%)): 381 $[M]^+$ (100), 337 (40), 303 (23), 260 (38), 77 (18). Found (%): C, 76.09; H, 6.63; N, 17.62. $C_{24}H_{23}N_5$. Calculated (%): C, 75.56; H, 6.08; N, 18.36.

5-Methyl-2-phenyl-3-(3-pyridyl)-7-[(*E*)-(3-pyridyl)methylene]-3,3a,4,5,6,7-hexahydro-2*H*-pyrazolo[4,3-*c*]pyridine (3f). The yield was 0.29 g (30%), acetone as the eluent, m.p. 115—119 °C (with decomp.). ^{1}H NMR, δ: 2.34—2.42 (m, 4 H, H(4), —NCH_3); 3.07—3.49 (m, 3 H, H(4), H(3a), H(6)); 3.89 (d, 1 H, H(6), J = 11.0 Hz); 4.89 (d, 1 H, H(3), J = 12.9 Hz); 6.98—7.14 (m, 5 H, *m*-H(Ph), *p*-H(Ph), β-H (pyridyl)); 7.35—7.62 (m, 3 H, *o*-H(Ph), —CH=); 7.88 and 7.94 (both d, 1 H each, γ-H (pyridyl), J = 8.0 Hz); 8.26—8.38 (m, 2 H, α-H (pyridyl)); 8.85 (br.s, 2 H, α′-H (pyridyl)). MS (EI, 70 eV), m/z (I_{rel} (%)): 381 $[M]^+$ (100), 337 (39), 303 (84), 260 (67), 77 (46). Found (%): C, 75.97; H, 6.49; N, 18.22. $C_{24}H_{23}N_5$. Calculated (%): C, 75.56; H, 6.08; N, 18.36.

5-Benzyl-2,3-diphenyl-7-[(*E*)-phenylmethylene]-3,3a,4,5,6,7-hexahydro-2*H*-pyrazolo[4,3-*c*]pyridine (3g). The yield was 0.34 g (25%), m.p. 155—157 °C (from ethanol). ^{1}H NMR, δ: 2.52 (t, 1 H, H(4), J = 10.4); 3.20 (dd, 1 H, H(6), J = 14.0 Hz, J = 2.1 Hz); 3.26 (dd, 1 H, H(4), J = 10.3 Hz, J = 6.1 Hz); 3.30—3.34 (m, 1 H, H(3a)); 3.60 (dd, 2 H, —CH_2—Ph, J = 10.3 Hz, J = 14.0 Hz); 4.07 (d, 1 H, H(6), J = 14.0 Hz); 4.60 (d, 1 H, H(3), J = 12.4 Hz); 6.80—7.42 (m, 21 H, Ph and —CH=). MS (EI, 70 eV), m/z (I_{rel} (%)): 455 $[M]^+$ (100), 378 (23), 335 (65), 259 (52), 77 (17). Found (%): C, 84.21; H, 6.50; N, 9.23. $C_{32}H_{29}N_3$. Calculated (%): C, 84.46; H, 6.42; N, 9.22.

5-Benzyl-3-(4-methoxyphenyl)-7-[(*E*)-(4-methoxyphenyl)methylene]-2-phenyl-3,3a,4,5,6,7-hexahydro-2*H*-pyrazolo[4,3-*c*]pyridine (3h). The yield was 0.50 g (42%), m.p. 157—158 °C (from ethanol). ^{1}H NMR, δ: 2.40 (t, 1 H, H(4), J = 10.8 Hz); 3.10 (dd, 1 H, H(6), J = 13.9 Hz, J = 2.1 Hz); 3.14—3.36 (m, 2 H, H(4), H(3a)); 3.69 (dd, 2H, —CH_2—Ph, J = 13.9 Hz, J = 15.9 Hz); 3.72 and 3.74 (both s, 3 H each, —OCH_3); 4.07 (d, 1 H, H(6), J = 14.0 Hz); 4.60 (d, 1 H, H(3), J = 12.4 Hz); 6.72—6.82 (m, 6 H, *p*-H (Ph) and *m*-H (Ph)); 6.98 and 7.04 (both d, 2 H each, *o*-H (4-$MeOC_6H_4$), J = 6.8 Hz); 7.13—7.18 (m, 4 H, *o*-H (Ph)); 7.19 (s, 1 H, —CH=); 7.22 and 7.40 (both d, 2 H each, *m*-H (4-$MeOC_6H_4$), J = 6.8 Hz). MS (EI, 70 eV), m/z (I_{rel} (%)): 515 $[M]^+$ (50), 425 (83), 397 (38), 304 (42), 250 (93), 174 (100), 91 (98), 77 (33). Found (%): C, 78.98; H, 6.32; N, 8.43. $C_{34}H_{33}N_3O_2$. Calculated (%): C, 79.20; H, 6.45; N, 8.15.

5-Benzyl-2-phenyl-3-(3-pyridyl)-7-[(*E*)-(3-pyridyl)methylene]-3,3a,4,5,6,7-hexahydro-2*H*-pyrazolo[4,3-*c*]pyridine (3i). The yield was 0.66 g (66%), ethyl acetate—light petroleum as the eluent, 1 : 1, m.p. 98—100 °C. ^{1}H NMR, δ: 2.50—2.56 (m, 1 H, H(4)); 3.26—3.50 (m, 3 H, H(3a), H(4), H(6)); 3.68 (dd, 2 H, —CH_2Ph, J = 13.8 Hz, J = 10.5 Hz); 3.97 (d, 1 H, H(6), J = 13.8 Hz); 4.62 (d, 1 H, H(3), J = 12.9 Hz); 6.50—7.32 and 8.46—8.68 (m, 19 H, H (Ph) and (pyridyl), —CH=). MS (EI, 70 eV), m/z (I_{rel} (%)): 457 $[M]^+$ (79), 379 (55), 337 (69), 260 (100), 181 (22), 91 (100), 77 (35). Found (%): C, 79.01; H, 6.73; N, 15.22. $C_{30}H_{27}N_5$. Calculated (%): C, 78.75; H, 5.95; N, 15.31.

2-(2-Pyridyl)-3-(3-pyridyl)-7-[(*E*)-(3-pyridyl)methylene]-3,3a,4,5,6,7-hexahydro-2*H*-indazole (3j). The yield was 0.37 g (39%), ethyl acetate as the eluent, m.p. 107—108 °C. ^{1}H NMR, δ: 1.23 (d, 1 H, H(5), J = 7.0 Hz); 1.25 (d, 1 H, H(4), J = 7.0 Hz); 1.66—2.58 (m, 3 H, H(4), H(5), H(6)); 2.97—3.10 (m, 1 H, H(3a)); 3.50 (dd, 1 H, H(6), J = 13.6 Hz, J = 7.0 Hz); 5.11

(d, 1 H, H(3), J = 11.6 Hz); 7.23—7.57 (m, 6 H, β-H (2-pyridyl), β-H (3-pyridyl), —CH=, β´-H (2-pyridyl), γ-H (2-pyridyl)); 7.97—8.02 (m, 2 H, γ-H (3-pyridyl)); 8.48—8.54 (m, 3 H, α-H (3-pyridyl), α-H (2-pyridyl)); 8.64 and 8.71 (both br.s, 1 H each, α´-H (3-pyridyl)). MS (EI, 70 eV), m/z (I_{rel} (%)): 367 $[M]^+$ (100). Found (%): C, 75.19; H, 6.07; N, 18.87. $C_{23}H_{21}N_5$. Calculated (%):C, 75.18; H, 5.76; N, 19.06.

3-Phenyl-7-[(*E*)-phenylmethylene]-2-(2-pyridyl)-3,3a,4,5,6,7-hexahydro-2*H*-indazole (3k). The yield was 0.43 g (47%), ethyl acetate as the eluent, m.p. 111—112 °C. ^{1}H NMR, δ: 1.67—1.81 (m, 2 H, H(5), H(4)); 1.95 (d, 1 H, H(5), J = 12.4 Hz); 2.28 (m, 1 H, H(4)); 2.47 (t, 1 H, H(6), J = 12.4 Hz); 2.90—2.99 (m, 1 H, H(3a)); 3.06 (d, 1 H, H(6), J = 5.7 Hz); 5.06 (d, 1 H, H(3), J = 11.4 Hz); 6.65—6.72 (m, 1 H, β-H (pyridyl)); 7.24—7.57 (m, 12 H, *m*-H(Ph), *p*-H(Ph), β´-H (pyridyl), *o*-H(Ph), —CH=); 7.82 (td, 1 H, γ-H (pyridyl), J = 7.6 Hz, J = 2.2 Hz); 8.16 (d, 1 H, α-H (pyridyl), J = 4.1 Hz). MS (EI, 70 eV), m/z (I_{rel} (%)): 366 $[M + H]^+$ (100), 364 $[M - H]^+$ (68), 275 $[M + H - C_7H_7]^+$ (19). Found (%): C, 82.59; H, 6.37; N, 11.75. $C_{25}H_{23}N_3$. Calculated (%): C, 82.16; H, 6.34; N, 11.50.

We thank A. T. Lebedev and V. V. Lobodin (Department of Chemistry, M. V. Lomonosov Moscow State University) for help in recording mass spectra.

This study was financially supported by the Russian Foundation for Basic Research (Project No. 03-03-32401).

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Received September 16, 2005;
in revised form November 1, 2005