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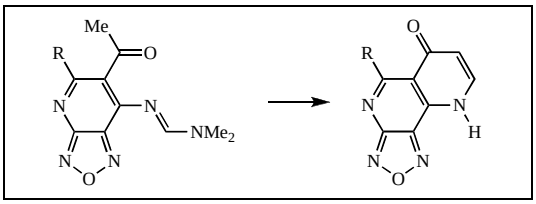

Reaction of 6-acetyl-7-aminofurazano[3,4-*b*]pyridines with DMFDMA afforded *N*,*N*-dimethylformamidines that were cyclized to the novel furazan-fused [1,6]naphthyridine system by treatment with sodium methylate in good yield. The tricyclic system is characterized by X-ray crystallography.

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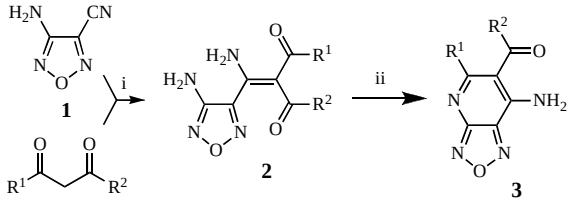
## INTRODUCTION

The 1.6-naphthyridine ring system represents a key structural subunit in numerous natural and synthetic compounds that exhibit a wide range of biological activities, including antimicrobial [1], anti-allergy and antiinflammatory [2], antidiabetic [3], antitumoral [4], and brain activating [5] properties. Considerable efforts have been directed toward the synthesis of 1.6-naphthyridine derivatives. A number of methods for their preparations have been developed and are well covered in the literature by some excellent reviews [6-10].

The synthesis of a variety of types of polycyclic compounds incorporating the 1,6-naphthyridine core has also been of considerable interest for the design of potential pharmaceutical agents and agrochemicals. Much greater attention has been directed towards the synthesis of benzo- and pyrido-annelated 1,6-naphthyridine ring systems probably as a consequence of the ready availability of suitable precursors [8-10]. At the same time only two azole[*h*]annelated ring systems have been reported, namely oxazolo[4,5-*f*][1,6]naphthyridine [11] and pyrazolo[3,4-*h*][1,6]naphthyridine [12-14].

Previous work of our laboratory [15,16] has led to the development of routes to furazano[3,4-*b*]pyridines [17] using 3-amino-4-cyanofurazan **1** [18,19] as the precursor. We have shown (Scheme 1) that the furazan **1** can be efficiently converted in one step into an enamine **2** by treatment with β-dicarbonyl compounds in the presence of catalytic amounts of nickel acetylacetonate, and that the enamine **2** can be transformed, also in one high-yielding step, to the *ortho* functionalized furazano[3,4-*b*]pyridine **3** by simple heating in acetic acid [15,16].

**Scheme 1**



*Reagents and conditions*: *i*, 1% nickel acetylacetonate, $CH_2Cl_2$; *ii*, AcOH, reflux

Herein, we report the successful synthesis of angularly fused furazano[1,6]naphthyridines using 6-acethyl-7-aminofurazano[3,4-*b*]pyridine **3** as a precursor.

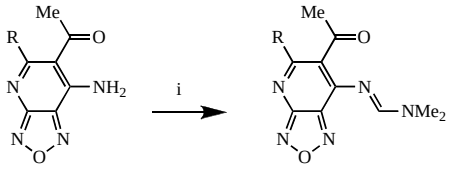
The construction of the [1,6]naphthyridine skeleton starting with pyridine derivatives bearing *ortho*-disposed acetyl and amino groups remains undocumented to date. Moreover, cyclization of *o*-acetyl-amino heterocycles to fused pyridines is quite rare reaction. Thus, sometime ago we reported for the first time the synthesis of pyrido[2,3-*d*]pyrimidines from 5-acetyl-4-aminopyrimidines [20,21]. The key intermediate in the transformation was an amidine that formed by reaction of starting amine with dimethylformamide dimethylacetal (DMFDMA [22]). Recently, similar protocol was used in the preparation of thieno[2,3-*b*;4,5-*b*']bipyridine derivatives [23].

## RESULTS AND DISCUSSION

The amidines **4a,b** (Scheme 2) required for our approach are readily prepared by condensation of amines **3a,b** with DMFDMA in refluxing benzene. The yields of this reaction range from 81 to 97%, and this procedure

should readily accommodate considerable functionality. The structure of amidines **4a,b** was confirmed from their $^1$H NMR spectra. For example, spectrum of **4a** displayed the expected dimethylamino group signals at δ 3.17 and 3.27 and signals from methyl at δ 2.54 and from acetyl group at 2.65. In the downfield region of the spectrum, the resonance at δ 8.98 was due to a formamidinic proton.

**Scheme 2**



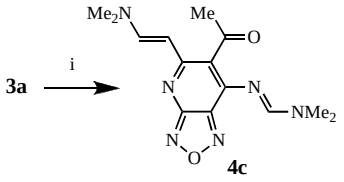
**3a,b** **4a,b**

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

*Reagents and conditions*: *i*, DMFDMA, benzene, reflux

If the amidation reaction is carried out in a high-boiling solvent such as xylene, DMFDMA reacted not only with the amino group, but with methyl group attached to the pyridine ring as well (Scheme 3). Compound **4c** was obtained in 66% yield.

**Scheme 3**



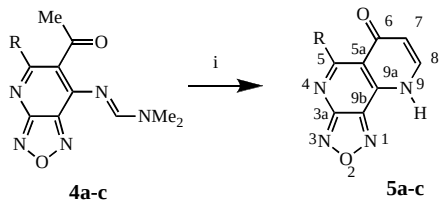
*Reagents and conditions*: *i*, DMFDMA, xylene, reflux

The $^1$H NMR spectra of **4c** showed a set of signals, indicating a stereoisomer. The downfield region of the spectrum revealed formamidinic proton at δ 8.87 and vinylic proton at δ 8.10, while second vinylic proton appeared at 4.86 (C***H***=CHNMe$_2$). *E*-Configuration around the C=C double bond in the enamine was deduced from the coupling constant ($J_{H\text{-}H}$ = 12.5 Hz [24]).

The key ring closure reaction was conducted under basic conditions (Scheme 4). Intramolecular condensation of acetyl and amidines groups was accomplished by refluxing the substrate **4a-c** in 1 *N* NaOMe in methanol to give yields of the furazanonaphthyridon **5a-c** in excess of 90%.

The structure of the products **5a-c**, as the pyridine-4-one, were supported by the observation of large values (*ca*. 6.5 Hz) for the coupling constants $J_{H(7),H(8)}$ and a carbonyl ($^{13}$C NMR) resonance for such constructs at 173.6 ppm. The spectral assignment of the pyridone form agreed with the literature [25-27]. Unequivocal

**Scheme 4**



**4a-c** **5a-c**

R = Me (**a**), Ph (**b**), CH=CHNMe$_2$ (**c**))

*Reagents and conditions*: *i*, MeONa, MeOH, reflux

assignment of all signals in the $^{13}$C NMR spectrum of tricycle **5a** from a simple noise or off-resonance proton-decoupled spectrum was not possible. Only the quarternary carbons (C-3 and C-5a) [15,16,28] and one of the protonated carbons (C-7) could readily be assigned, whilst there was some ambiguity of assignment between C-5 and C-6 and between C-8, C-9a and C-9b. However, assignment of the spectrum is important for future identification of substituted and/or analogous compounds. The assignment of the carbon resonances was performed using the 2D $^1$H/$^{13}$C HMBC spectrum (see experimental). The mass spectra of the tricycles **5a-c** showed a strong peak corresponding to the molecular ion [M$^+$] and subsequently exhibit a relatively simple fragmentation pattern due to initial loss of NO to give a peak at [M$^+$ – 30] that typical for furazans [28,29], followed by loss of HCN [M$^+$ – NO – 27]. Subsequent fragmentation involves a further loss of CN. All the furazanonaphthyridones show the presence of the bridging NH group at *ca*. 2750 cm$^{-1}$ and characteristic carbonyl absorptions at 1650-1670 cm$^{-1}$ in their IR spectra and most have another intense peak at *ca*. 1620, 1570, and 1540 cm$^{-1}$, which may be associated with C=N vibrations.

The detailed structural features of pyridone **5a** were established by low temperature X-ray study. ORTEP view of the molecule **5a** is presented in Figure 1. Molecule is characterized by nearly planar geometry (mean deviation is 0.053Å). The hydrogen atom at the N(9) nitrogen was found in the difference fourier map while no peaks were found near the O(3) atom at the appropriate angles. The C(6)=O(3) bond (1.248(3)Å) is elongated relative to the mean X-ray value (1.222Å [30]) which is due probably to the participation of the O(3) atom in hydrogen bonding. Thus the molecular structure of compound **5a** adopts pyridone form. The bond lengths distribution (Table 1) is in accord with the pyridone assignment. The bond lengths at the N(9) atom are similar to those at the N(4) atom of pyridine fragment that imply participation of the lone pair on N(4) in conjugation. The C(7)-C(8) bond is more likely a double bond while C(6)-C(5a) and C(6)-C(7) bond lengths are among the longest C-C bonds in **5a**.

To get a deeper insight into the difference in the structural and energetic characteristics of the pyridone and hyrdoxypyridine forms we have carried out quantum chemical calculations at DFT and MP2 levels utilizing the the Gaussian program [31].

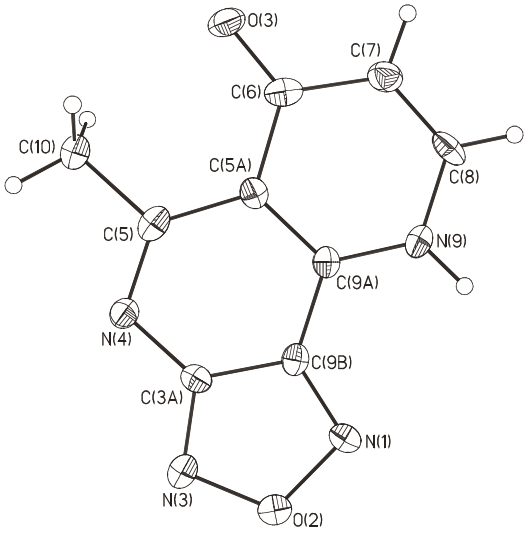



**Figure 1.** ORTEP view (50% probability) of the molecule **5a**.

The results summarized in Table 1 show the similarity between the calculated for pyridone form and experimental bond lengths distribution. From the quantum chemical calculation, it follows that the isolated molecule is more stable in the pyridone form by 4.7 kcal/mol (B3LYP/6-311G**) and by 1.7 kcal/mol (MP2/cc-pvdz) [32].

In the crystal, pyridone **5a** exists as hydrate. The asymmetric unit cell contains one **5a** and one water molecule. Hydrogen bonds are formed between compound **5a** and water molecules thereby leading to H-bonded layers parallel to the *bc* plane (Figure 2).

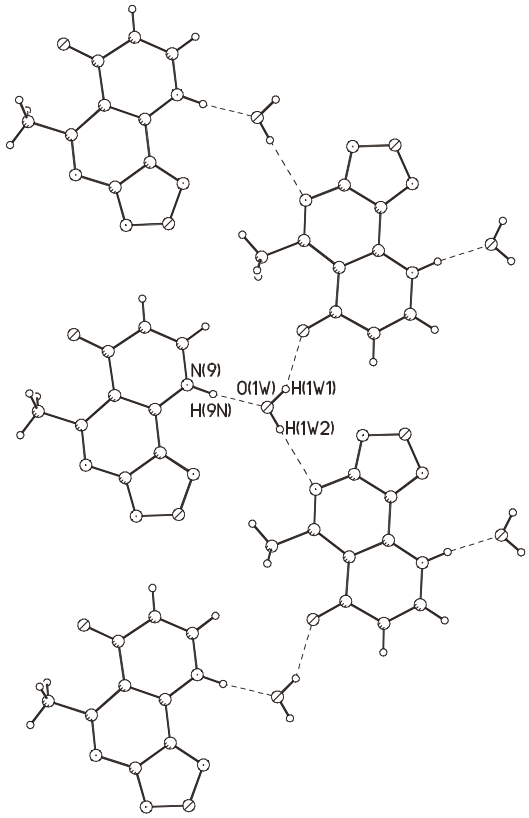



**Figure 2.** Crystal packing fragment of **5a**·$H_2O$. H-bonded layer parallel to *bc* plane is shown.

The distances between mean planes of ajacent H-bonded layers alternate being equal to 3.107 and 3.278 Å. However the projections of the molecular $\pi$-systems do not overlap.

It is interesting to note that the same molecular arrangement in the layer can be formed by H-bonding if molecules in the crystal would exist in pyridone form. So

**Table 1**

Selected bond distances for the compound **5a** in comparison with *ab initio* calculations.

| Bond | X-Ray | Pyridone form | | Hydroxypyridine form | |
|---|---|---|---|---|---|
| | | B3LYP/ 6-311G** | MP2/cc-pvdz | B3LYP/ 6-311G** | MP2/cc-pvdz |
| N(4)-C(3A) | 1.364(3) | 1.358 | 1.362 | 1.365 | 1.372 |
| N(4)-C(5) | 1.326(3) | 1.312 | 1.332 | 1.303 | 1.321 |
| C(5)-C(5A) | 1.465(3) | 1.474 | 1.467 | 1.482 | 1.480 |
| C(5A)-C(6) | 1.469(3) | 1.490 | 1.495 | 1.419 | 1.428 |
| C(5A)-C(9A) | 1.403(3) | 1.392 | 1.409 | 1.427 | 1.434 |
| C(6)-C(7) | 1.441(3) | 1.466 | 1.469 | 1.391 | 1.398 |
| C(7)-C(8) | 1.346(3) | 1.349 | 1.368 | 1.392 | 1.407 |
| N(9)-C(8) | 1.373(3) | 1.372 | 1.373 | 1.327 | 1.336 |
| N(9)-C(9A) | 1.333(3) | 1.362 | 1.368 | 1.338 | 1.357 |
| C(9A)-C(9B) | 1.435(3) | 1.428 | 1.424 | 1.443 | 1.440 |
| C(3A)-C(9B) | 1.411(3) | 1.424 | 1.412 | 1.424 | 1.411 |

the realization of the pyridone **5a** is mostly defined by intramolecular forces rather then by the effect of the crystal field.

In conclusion, we have demonstrated that intramolecular condensation of *ortho* acetylpyridin amidines provides a new and versatile approach to the synthesis of naphthyridine ring system. The strategy used is likely to be of value in gaining simple access to other (un)annelated naphthyridinons.

## EXPERIMENTAL

Melting points were determined on Gallenkamp melting point apparatus and they are not corrected. Infrared spectra were determined in KBr pellets on a Perkin-Elmer Model 577 spectrometer. Mass-spectra were recorded on a Varian MAT-311A instrument. $^1$H NMR and $^{13}$C spectra were recorded on a Bruker DRXV-500 spectrometer. Chemical shifts for both $^1$H NMR and $^{13}$C NMR are referred to chemical shifts for solvent (for DMSO-$d_6$ it is 2.50 ppm and 39.51 ppm for proton and carbon NMR, respectively). HMBC experiments were optimized for the coupling constant $J_{H,C}$ 8 Hz at 50 °C. Analytical thin layer chromatography (TLC) was conducted on precoated silica gel plates (Silufol $F_{254}$).

**6-Acetyl-7-(dimethylaminomethylenamino)-5-methylfurazan[3,4-*b*]pyridine (4a).** To a stirring solution of DMFDMA (0.12 g, 1 mmol) in benzene (3,5 mL) was added compound **3a** (0.19 g, 1 mmol). The mixture was heated under reflux for 1 h and the solvent evaporated to dryness. The residue was recrystallized from benzene-hexane, giving the amidines **4a** as a yellow solid: 0.2 g (81%), mp 118-119 °C; $^1$H NMR (DMSO-$d_6$) δ 2.54 (s, 3H, Me), 2.65 (s, 3H, Me), 3.17 (s, 3H, MeN), 3.27 (s, 3H, MeN), 8.98 (s, 1H, CH=NMe). *Anal.* Calcd. for $C_{11}H_{13}N_5O_2$ (247.26): C 53.43, H 5.30, N 28.32. Found C 53.37, H 5.34, N 28.21.

**6-Acetyl-7-(dimethylaminomethylenamino)-5-phenylfurazan[3,4-*b*]pyridine (4b).** Following the above procedure and starting from compound **3b**, amidines **4b** was obtained in 97% yield; mp 190-192 °C; $^1$H NMR ($CDCl_3$) δ 2.38 (s, 3H, Me), 3.19 (s, 3H, MeN), 3.28 (s, 3H, MeN), 7.40-7.70 (m, 5H, Ph), 9.01 (s, 1H, CH=NMe). *Anal.* Calcd. for $C_{16}H_{15}N_5O_2$ (309.33): C 62.13, H 4.89, N 22.64. Found C 62.08, H 4.78, N 22.72.

**6-Acetyl-7-(dimethylaminomethylenamino)-5-(dimethylaminovinyl)furazan[3,4-*b*]pyridine (4c).** To a stirring solution of DMFDMA (0.35 g, 2.9 mmol) in xylene (10 mL) was added compound **3a** (0.25 g, 1.3 mmol). The mixture was heated under reflux for 3-4 h and the solvent evaporated. The residue was chromatographed on silica gel. Elution with $CHCl_3$-benzene (3:1) gave little amount amidine **4a** and elution with $CHCl_3$-acetone (10:1) then gave product **4c**: 0.26 g (66%), mp 170-172 °C; $^1$H NMR (DMSO-$d_6$) δ 2.60 (s, 3H, Me), 2.95 (b.s., 6H, $NMe_2$), 3.08 (s, 3H, MeN), 3.16 (s, 3H, MeN), 4.86 (d, 1H, C*H*=CHN, *J* = 12.5 Hz), 8.10 (d, 1H, CH=C*H*N, *J* = 12.5 Hz), 8.87 (s, 1H, C*H*=NMe). *Anal.* Calcd. for $C_{14}H_{18}N_6O_2$ (302.34): C 55.62, H 6.00, N 27.80. Found C 55.71, H 6.06, N 27.68.

**General Procedures for the Preparation of Furazano[3,4-*b*][1,6]naphthyridine-6-ons (5a-c).** To a stirred solution of sodium (0.12 g, 5 mmol) in MeOH (25 ml) was added corresponding amidines (5 mmol), and the mixture was refluxed for 1.5 h and concentrated under reduced pressure. The residue was stirred with a mixture of $H_2O$ (10 ml) and AcOH (0.5 ml). The precipitate was collected by filtration, recrystallized from ethanol, and dried *in vacuo* to give the pure product.

**5-Methyl-9(*H*)-furazano[3,4-*b*][1,6]naphthyridine-6-on (5a).** This compound was obtained in 94% yield; mp 292-294 °C; IR ν 2700-2900 (NH), 1656 (C=O), 1620, 1568, 1560, 1540, 1524, 1436, 1408, 1380, 1300, 1204, 1032, 888, 848 cm$^{-1}$; MS: m/z 202 [M$^+$], 172 [M$^+$ - NO], 145 [M$^+$ - NO - HCN], 118 [M$^+$ - NO - 2HCN]; $^1$H NMR (DMSO-$d_6$) δ 2.98 (s, 3H, Me), 6.65 (d, 1H, H-7, *J* = 6.5 Hz), 8.06 (d, 1H, H-8, *J* = 6.5 Hz), 13.3 (b.s, 1H, NH); $^{13}$C NMR (DMSO-$d_6$) δ 28.4 (Me), 116.9 (C-5a), 119.2 (C-7), 137.2 (C-9a), 139.1 (C-9b), 140.5 (C-8), 157.0 (C-3a), 171.4 (C5), 173.6 (C-6). *Anal.* Calcd. for $C_9H_6N_4O_2$ (202.17): C 53.47, H 2.99, N 27.71. Found C 53.57, H 3.12, N 27.77.

**5-Phenyl-9(*H*)-furazano[3,4-*b*][1,6]naphthyridine-6-on (5b).** This compound was obtained in 93.5% yield; mp 331-333 °C; IR ν 2700-2900 (NH), 1648 (C=O), 1620, 1584, 1544, 1524, 1432, 1360, 1324, 1228, 1104, 1076, 1032, 964, 820, 800 cm$^{-1}$; MS: m/z 264 [M$^+$], 234 [M$^+$ - NO]; $^1$H NMR (DMSO-$d_6$) δ 6.62 (d, 1H, H-7, *J* = 7 Hz), 7.35-7.60 (m, 5H, Ph), 8.15 (d, 1H, H-8, *J* = 7 Hz), 13.3 (b.s, 1H, NH). *Anal.* Calcd. for $C_{14}H_8N_4O_2$ (264.24): C 63.64, H 3.05, N 21.20. Found C 63.53, H 3.11, N 21.08.

**5-Dimethylaminovinyl-9(*H*)-furazano[3,4-*b*][1,6]-naphthyridine-6-on (5c).** This compound was obtained in 94% yield; mp 235-237 °C; MS: m/z 257 [M$^+$], 227 [M$^+$ - NO]; $^1$H NMR (DMSO-$d_6$) δ 3.00-3.50 (bs, 6H, $NMe_2$), 6.46 (d, 1H, H-7, *J* = 7 Hz), 7.52 (d, 1H, C*H*=CHN$Me_2$, *J* = 14 Hz), 7.83 (d, 1H, H-8, *J* = 7 Hz), 8.30 (d, 1H, C*H*N$Me_2$, *J* = 14 Hz). *Anal.* Calcd. for $C_{12}H_{11}N_5O_2$ (257.25): C 56.03, H 4.31, N 27.22. Found C 56.16, H 4.34, N 27.11.

**X-ray Crystallography.** X-Ray quality single crystals of the compound **5a** were grown by slow evaporation of a THF solution at room temperature. The single crystals ($C_9H_6N_4O_2 \cdot H_2O$) are monoclinic, space group $P2_1/n$: *a* = 6.5561(7) Å, *b* = 15.0484(17)Å, *c* = 9.5470(10)Å, β =102.505(2)°, *V* = 919.55(17) Å$^3$, Z = 4, M = 220.2, $d_{calc}$ = 1.591 g·cm$^{-3}$, $\mu$ = 0.124 mm$^{-1}$. Carefully chosen sample (0.20 × 0.15 × 0.02 mm) was used for the X-ray study. 7927 reflections were collected at SMART 1000 CCD difractometer (λ(Mo-Kα)=0.71073 Å, graphite monochromator, ω-scans, 2θ<52°) at 120 K. The structure was solved by the direct methods and refined by the full-matrix least-squares procedure in anisotropic approximation. 1805 Independent reflections [$R_{int}$ = 0.1106] were used in the refinement procedure (for 146 parameters) that was converged to $wR_2$ = 0.0579 calculated on $F^2_{hkl}$ (*GOF* = 0.805, $R_1$ = 0.0401 calculated on $F_{hkl}$ using 846 reflections with $I>2\sigma(I)$). For the analysis of data collected and crystal structures refinement we used SAINT Plus [33], SADABS and SHELXTL [34] program packages. Atomic coordinates and thermal parameters, bond lengths and angles have been deposited at the Cambridge Crystallographic Data Centre as supplementary publication number of CCDC 631328.

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