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SYNTHESIS AND HYDRAZINOLYSIS OF β-(1,3,4-OXADIAZOL-2-YL)PYRIDINES

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Cyclization of the hydrazide of 5-ethoxycarbonyl-2,6-dimethylpyridine-3-carboxylic acid by acylation with aromatic or aliphatic acid chlorides with subsequent boiling in $POCl_3$ or heating in orthoformic acid gave the corresponding ethyl 2,6-dimethyl-5-(5-R-1,3,4-oxadiazol-2-yl)pyridine-3-carboxylate. The cyclization of the reaction products with hydrazine hydrate has been studied. Cyclization of the dihydrazide of 2,6-dimethyl-3,5-pyridinedicarboxylic acid under analogous conditions gave only 3,5-bis-(5-R-1,3,4-oxadiazol-2-yl)-2,6-dimethylpyridines, containing R = 2-FC_6H_4, H.

Keywords: hydrazide, hydrazine, nicotinic acid, 1,3,4-oxadiazole, aromatic acid chlorides.

Derivatives of 1,3,4-oxadiazoles have found use as antimicrobial [1, 2], antifungal [3, 4], antibacterial [5], and sedative preparations [6]. Preparation of 1,3,4-oxadiazoles containing pyridine units is coupled with a number of difficulties, linked with their chemical instability and the complexity of isolating them from the reaction media [7].

Previously [8] we studied the reactivity of hydrazides of 3,5-pyridinedicarboxylic acid **1**, **2** in acylation reaction with aromatic acids chlorides which proceeded with formation of the hydrochlorides of the corresponding mono- and bis-1,2-diacylhydrazine-2,6-dimethylpyridines **3**, **4b-d**, **g**, isolated in the form of the bases.

With the objective of preparing the (1,3,4-oxadiazol-2-yl)pyridines **5** and **6** we have investigated the reaction of the heterocycles **3** and **4** with various dehydrating agents (phosphorus oxychloride, thionyl chloride, oleum, and polyphosphoric acid). The most effective cyclizing agent is phosphorus oxychloride. The use of conc. H_2SO_4 or oleum was ineffective since the reaction did not occur at low temperatures and strong resinification was observed on heating the reaction mixture. Analogous results were obtained when polyphosphoric acid or thionyl chloride were used.

Attempts to obtain the (1,3,4-oxadiazol-2-yl)pyridines **5b-d**,**g**,**i** by heating the 1,2-diacylhydrazines **3b-d**, **g**,**i** in $POCl_3$ proceeded in low yield, since, because of the poor solubility of the bases, prolonged heating was required which lead to resinification.

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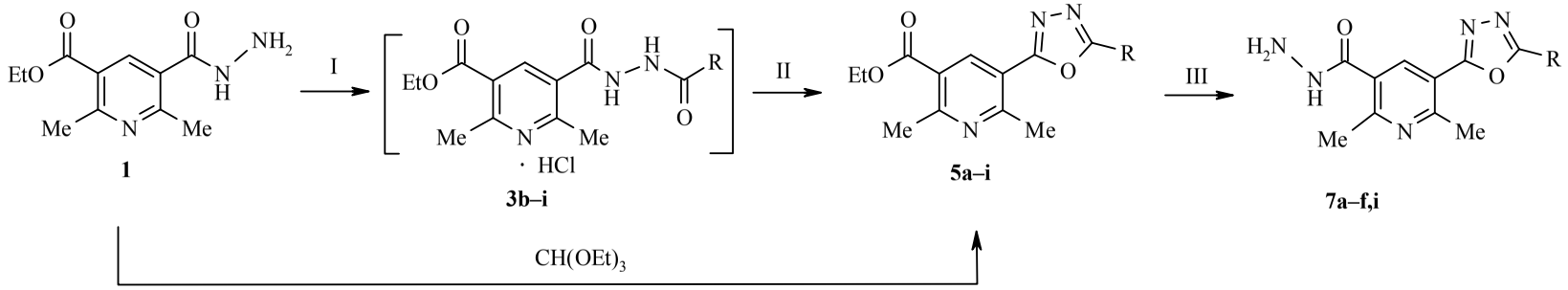

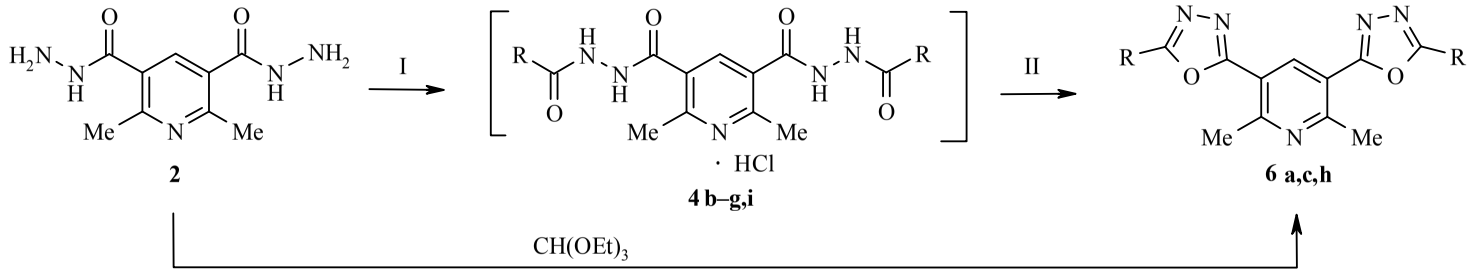

a R = H, **b** R = 4-*t*-BuC_6H_4, **c** R = 2-FC_6H_4, **d** R = 2-MeOC_6H_4, **e** R = 4-MeC_6H_4, **f** R = 2-ClC_6H_4, **g** R = 4-$NO_2C_6H_4$, **h** R = Me, **i** R = Ph;
I. RC(O)Cl or Ac_2O (compound **6h**), MeCN; II. 1) $POCl_3$; 2) base; III. $NH_2NH_2 \cdot H_2O$, EtOH

The use in this reaction of 1,2-diacylhydrazine hydrochlorides **3b-d**,**g**,**i** increased their solubility and allowed the formation of the corresponding (1,3,4-oxadiazol-2-yl)pyridines in yields of 20-35%. As it was turned out, the simplest and most effective method of cyclization is to carry out the whole process in one pot, beginning with the acylation of hydrazides **1** with an acids chlorides followed by removal of the solvent in vacuum followed by brief heating of the 1,2-diacylhydrazine **3b-i** hydrochlorides obtained without isolation in phosphorus oxychloride. In this way we succeeded in increasing the yields of compounds **5b-i** (Table 1) to 60-88% and also decreising the reaction time considerably. However with hydrazide **2** only with the use 2-fluorobenzoyl chloride were we able to isolate the corresponding 3,5-bis(1,3,4-oxadiazol-2-yl)pyridine **6c** in satisfactory yield. Cyclization of the bis-1,2-diacylhydrazines **4b**,**d-g**,**i** formed *in situ* did not occur because of the low solubility of these salts, prolonged heating in $POCl_3$ gave rise to resinification of the reaction masses.

Neutralization of the reaction mixture with sodium hydrogen carbonate in the process of isolation of the compounds **5b-i** was not acceptable in the case of (5-methyl-1,3,4-oxadiazol-2-yl)pyridine **5h** which hydrolyzed to the starting monohydrazide **1** under these conditions. Therefore for this compound neutralization was carried out at 5-7°C with 10% ammonia solution.

TABLE 1. Physicochemical Characteristics of Compounds **5a-i**, **6a**,**c**,**h**, **7a-f**,**i**

Compound	Empirical formula	Found, % / Calculated, %			mp, °C	Yield, %
		C	H	N		
5a	$C_{12}H_{13}N_3O_3$	58.13 / 58.29	5.22 / 5.30	17.08 / 16.99	94-95	88
5b	$C_{22}H_{25}N_3O_3$	69.32 / 69.64	6.71 / 6.64	11.23 / 11.07	148-149	68
5c	$C_{18}H_{16}FN_3O_3$	60.31 / 60.43	4.62 / 4.51	11.79 / 11.74	116-117	77
5d	$C_{19}H_{19}N_3O_4$	64.47 / 64.58	5.34 / 5.42	11.69 / 11.89	133-135	64
5e	$C_{19}H_{19}N_3O_3$	67.73 / 67.64	5.52 / 5.68	12.38 / 12.45	153-155	72
5f	$C_{18}H_{16}ClN_3O_3$	60.32 / 60.43	4.56 / 4.51	11.57 / 11.74	108-109	78
5g	$C_{18}H_{16}N_4O_5$	57.74 / 58.70	4.11 / 4.35	14.57 / 15.22	168-170	25
5h	$C_{13}H_{15}N_3O_3$	59.85 / 59.77	5.81 / 5.74	15.57 / 16.09	96-97	58
5i	$C_{18}H_{17}N_3O$	65.72 / 66.87	5.08 / 5.26	12.78 / 13.00	137-138	60
6a	$C_{11}H_9N_5O_2$	54.28 / 54.32	3.40 / 3.73	28.61 / 28.79	168-169	90
6c	$C_{21}H_{15}F_2N_5O_2$	61.11 / 61.92	3.63 / 3.69	17.36 / 17.20	165-166	45
6h	$C_{13}H_{13}N_5O_2$	56.83 / 57.53	4.83 / 4.80	25.55 / 25.83	169-171	67
7a	$C_{10}H_{11}N_5O_2$	51.42 / 51.50	4.68 / 4.75	29.75 / 30.03	228-230	98
7b	$C_{20}H_{23}N_5O_2$	65.56 / 65.74	6.42 / 6.34	19.08 / 19.16	210-212	88
7c	$C_{16}H_{14}FN_5O_2$	55.93 / 55.90	4.12 / 4.10	20.34 / 20.37	255-257	93
7d	$C_{17}H_{17}N_5O_3$	60.28 / 60.17	5.22 / 5.05	20.47 / 20.64	242-243	96
7e	$C_{17}H_{17}N_5O_2$	63.24 / 63.15	5.36 / 5.30	21.59 / 21.66	304-306	93
7f	$C_{16}H_{14}ClN_5O_2$	55.78 / 55.90	4.18 / 4.10	20.45 / 20.37	246-247	92
7i	$C_{16}H_{15}N_5O_2$	62.09 / 62.13	4.92 / 4.89	20.45 / 20.64	250-251	89

Use of acetic anhydride as the acylating agent for the dihydrazide **2** with subsequent short heating in phosphorus oxychloride without isolation of the product of acylation led to the formation of the corresponding bis(5-methyl-1,3,4-oxadiazol-2-yl)pyridine **6h** with a yield of ~ 70%. This is apparently connected to the high solubility of the acylation product in $POCl_3$ and to the electron-donating character of the alkyl radical.

It has been shown [9] in a study of the mechanism of the heterocyclization 1,2-diacylhydrazines that a necessary condition for the formation 1,3,4-oxadiazoles is the presence of different nucleophilicity of the carbonyl oxygen atoms in the 1,2-diacylhydrazine unit. The salt form of the pyridine ring in the 1,2-diacylhydrazines hydrohlorides **3**, **4** evidently leads to decreased basicity of the oxygen of the β-carbonyl unit, so that cyclization is facilitated by an electron donor substituent in the acyl component. If the acyl group has electron acceptor properties, the basicity of both oxygen atoms of the carbonyl group is reduced which retards the protonation step and further cyclization. Hence heterocyclization of the hydrochloride of ethyl 5-[(4-nitrobenzoyl)hydrazinocarbonyl]-3-carboxylate (**3g**) occurs with low yield (~25%), and attempts to use trifluoroacetic anhydride, analogously to acetic anhydride, in the synthesis of the corresponding bis(1,3,4-oxadiaxol-2-yl)pyridine **6** was unsuccessful.

It is known [10] that 2,5-unsubsituted 1,3,4-oxadiazoles can be obtained from hydrazides of carboxylic acids by heating in an excess of ethyl orthoformate. Carrying out this reaction with mono- and dihydrazides of pyridine **1** and **2** in a 20-fold excess of ethyl orthoformate showed that the expected mono- and bis-1,3,4-oxadiazoles **5a** and **6a** were formed in high yields (78-85%).

Hydrazinolysis of the obtained 1,3,4-oxadiazoles **5a-i** occurs both at the ester group and in the recyclization of the 1,3,4-oxadiazole ring into the 1,2,4-triazole ring. This, in the first place, is concerned with monosubstituted 1,3,4-oxadiazoles which are also easily hydrolyzed [10], whereas recyclization of 2,5-diaryl-substituted 1,3,4-oxadiazoles was not successful [11, 12]. We have shown that hydrazinolysis with opening of the 1,3,4-oxadiazole ring occurs on boiling (5-methyl-1,3,4-oxadiazol-2-yl)pyridine **5h** in an alcohol solution of excess hydrazine hydrate to give the dihydrazide **2**. On heating compounds **5a-f,i** for a short time in ethanolic solutions of 85% hydrazine hydrate, the hydrazides **7a-f**,**i** are formed in high yields, prolonged heating of which in alcoholic (ethanol, 2-propanol, butanol) and aqueous solution of hydrazine hydrate did not lead to products of ring-opening or recyclization of the 1,3,4-oxadiazole ring, which is probably connected with their w solubility in the reaction mixture.

TABLE 2. IR Spectra of Compounds **5a-i** and **7a-f**,**i**

Com-pound	ν, cm^{-1}
5a	3140, 3000, 2935, 1720, 1590, 1560, 1535, 1450, 1320, 1280, 1100, 1090
5b	3080, 2980, 2890, 1720, 1620, 1550, 1505, 1460, 1420, 1380, 1290, 1250, 1240, 1210, 160, 1110
5c	3000, 2945, 1740, 1610, 1550, 1505, 1480, 1450, 1405, 1380, 1290, 1240, 1175, 1110
5d	3000, 2960, 1740, 1610, 1550, 1485, 1440, 1300, 1290, 1240, 1190, 1100
5e	2990, 2940, 2870, 1735, 1620, 1595, 1570, 1510, 1465, 1380, 1290, 1270, 1240, 1225, 1195, 1180, 1110
5f	3000, 2950, 1740, 1610, 1555, 1465, 1405, 1380, 1295, 1275, 1240, 1170, 1110
5g	3000, 2950, 1735, 1610, 1553, 1475, 1380, 1295, 1275, 1240, 1175, 1110
5h	3000, 2960, 1730, 1610, 1540, 1495, 1460, 1375, 1290, 1240, 1190, 1100
5i	2990, 1740, 1610, 1572, 1510, 1465, 1380, 1290, 1270, 1245, 1190, 1110
7a	3320, 3250, 3030, 2870, 1670, 1595, 1450, 1385, 1315, 1195, 1150, 1070
7b	3310, 3190, 2980, 2890, 1650, 1550, 1510, 1460, 1380, 1330, 1280, 1260, 1120, 1 090
7c	3290, 3180, 3080, 1650, 1550, 1500, 1480, 1330, 1375, 1275, 1240, 1180, 1170, 1 120
7d	3310, 3220, 3100, 3000, 1640, 1620, 1550, 1510, 1470, 1330, 1290, 1270, 1180
7e	3285, 3180, 3075, 3010, 2940, 2870, 1645, 1560, 1510, 1465, 1370, 1335, 1275, 1250, 1225, 1190, 1175, 1115
7f	3285, 3180, 1650, 1615, 1560, 1485, 1465, 1440, 1330, 1295, 1270, 1250, 1170, 1 110
7i	3288, 3170, 1640, 1625, 1567, 1485, 1465, 1440, 1330, 1295, 1270, 1250, 1170, 1110

TABLE 3. ^{1}H NMR Spectra of the Synthesized Compounds **5a-i**, **6a**,**s**,**h**, **7a-f**,**i**

Compound	Chemical shifts, δ, ppm (*J*, Hz)*						
	$OCH_2C\underline{H}_3$ (3H, t)	CH_3 (3H, s)	NH_2 (2H, s)	$OC\underline{H}_2CH_3$ (2H, q)	4-H Py (1H, s)	NH (1H, s)	R
5a	1.43 (^3J = 6.1)	2.90, 2.99		4.43 (^3J = 6.1)	8.75		8.57 (1H, s, CH)
5b	1.39 (^3J = 6.5)	2.80, 2.93		4.40 (^3J = 6.5)	8.68		1.35 (9H, s, $C(CH_3)_3$); 7.62, 8.02 (4H, AA'BB' system, ^3J = 7.2, C_6H_4)
5c	1.45 (^3J = 7.8)	2.84, 2.98		4.40 (^3J = 7.8)	8.72		7.35-7.44 (2H, m, H-5,6); 7.67 (1H, m, H-4); 8.11-8.17 (1H, m, H-3)
5d	1.43 (^3J = 6.2)	2.83, 2.94		4.38 (^3J = 6.2)	8.73		4.00 (3H, s, CH_3O); 7.09-7.20 (2H, m, H-3,4); 7.56 (1H, td, ^3J = 8.1,^4J = 2.2, H-5), 7.99 (1H, dd, ^3J = 8.1,^4J = 2.2, H-6)
5e	1.43 (^3J = 6.5)	2.46, 2.95		4.40 (^3J = 6.5)	8.67		2.81 (3H, s, CH_3); 7.36, 7.99 (4H, AA'BB' system, ^3J = 8.1, C_6H_4)
5f	1.42 (^3J = 7.6)	2.81, 2.96		4.39 (^3J = 7.6)	8.72		7.50-7.64 (3H, m, H-4,5,6); 8.13 (1H, d, ^3J = 8.9, H-3)
5g	1.43 (^3J = 7.2)	2.81, 2.95		4.40 (^3J = 7.2)	8.75		8.39, 8.46 (4H, AA'BB'-system, ^3J = 8.3, C_6H_4)
5h	1.43 (^3J = 6.0)	2.73, 2.81		4.31 (^3J = 6.0)	8.51		2.59 (3H, s, CH_3)
5i	1.43 (^3J = 6.5)	2.83, 2.97		4.41 (^3J = 6.5)	8.71		7.54-7.63 (3H, m, C_6H_5); 8.10-8.16 (2H, m, C_6H_5)
6a		2.91 (6H)			8.69		9.40 (2H, s, CH)
6c		3.03 (6H)			8.92		7.37-7.44 (4H, m, H-5,6, H'-5,6); 7.62-7.67 (2H, m, H-4, H'-4); 8.19, 8.21 (2H, two d, ^3J = 9.0, H-3, H'-3)
6h		2.85 (6H)			8.62		2.60 (6H, s, $2CH_3$)
7a		2.51	—*[2]		7.66	9.41	8.60 (1H, s, CH)
7b		2.50, 2.67	—*[2]		7.93	10.35	1.34 (9H, s, $C(CH_3)_3$); 7.50, 7.88 (4H, AA'BB' system, ^3J = 6.3, C_6H_4)
7c		2.67 2.93	4.34		8.32	9.64	7.32-7.48 (2H, m, H-5,6), 7.62-7.71 (1H, m, H-4); 8.16-8.23 (1H, m, H-3)
7d		2.57, 2.76	4.51		8.21	9.64	3.94 (3H, s, OCH_3); 7.15, 7.63 (2H, two td, ^3J = 8.8, ^4J = 2.0, H-4,5); 7.29, 7.95 (2H, two dd, ^3J = 8.8, ^4J = 2.0, H-3,6)
7e		2.50, 2.95	—*[2]		8.33	9.60	2.68 (3H, s, CH_3); 7.37, 8.04 (4H, AA'BB' system, ^3J = 7.8, C_6H_4)
7f		2.58, 2.87	4.56		8.24	9.64	7.54-7.75 (3H, m, H-4,5,6); 8.14 (1H, dd, ^3J = 8.0, ^4J = 2.4, H-3)
7i		2.56, 2.77	4.50		8.18	9.89	7.40-7.52 (5H, m, C_6H_5)

* ^{1}H NMR spectra were recorded in DMSO-d_6 (compounds **5a-g**,**i** and **7a-i**) and $CDCl_3$ (compounds **5h** and **6a**,**c**,**h**).

*[2] Signal not observed.

In the IR spectra of the synthesized (1,3,4-oxadiazol-2-yl)pyridines **5b-i** (Table 2) there are intense valence bands of the C=N bond in the 1610-1620 cm^{-1} range, which corresponds to previous date [4]. The stretching bands of the ester groups (Table 2) undergo a high frequency shift in comparison with the starting diacylhydrazines (from the 1710-1720 range for compounds **3c**,**d**,**g**,**i** [8] to the range 1735-1740 cm^{-1} for the corresponding oxadiazoles **5c**,**d**,**g**,**i**), which may indicate the greater electron acceptor influence of the 1,3,4-oxadiazole unit. The excluded compounds **3b** and **5b** contain the 4-*tert*-butylphenyl substituent.

In the IR spectra of the hydrazinolysis products **7a-f**,**i** the $NH-NH_2$ fragment appeared as two bands of different intensity in the region of 3320-3100 cm^{-1}, which is in agreement with data from [13] and indicates the stretching vibrations of the unit as a whole. The intense "amide I" band, connected with $\nu_{C=O}$ of the hydrazide group, appears in the 1640-1650 cm^{-1} region. The presence of an unsaturated oxadiazole unit in the structure of compound **7a** led to a high frequency shift of these bands to 1670 cm^{-1} (Table 2).

The highest information content on the electronic influence of the substituent on the pyridine ring can be obtained, in our view, from analysis of the signal of the gamma-proton of the pyridine ring in the ^1H NMR spectra. Comparative analysis of the ^1H NMR spectra shows that in going from the 1,2-diacylhydrazines **3b-d**,**g**,**i** [8] to the corresponding (1,3,4-oxadiazol-2-yl)pyridines **5b-d**,**g**,**i** (Table 3) the signal of the pyridine proton underwent a weak field shift from 7.9-8.2 to 8.7-8.8 ppm, while the introduction of a second oxadiazole ring in the case of compound **6c**, which contains a 2-fluorophenyl substituent, led to an additional shift of ~0.2 ppm to weak field. As a result of a comparative analysis of the spectra of (1,3,4-oxadiazol-2-yl)pyridines **5a-i** with the spectra of the products of their hydrazinolysis **7a-f**,**i** a shift of the signal of the gamma-proton by 0.3-1.1 ppm to strong field was observed, which is explained by the electron donor characteristic of the hydrazine unit in comparison with the ester group. The maximum shifts of the gamma-proton were observed in the cases of the (1,3,4-oxadiazol-2-yl)pyridines **5a** and **7a** which contain 1,3,4-oxadiazole units unsubstituted at position 5.

Thus, hydrazinolysis of the ester groups in compounds **5a-i** appears to be preferable if there is no substituent or an aroyl unit at position 5 of the 1,3,4-oxadiazole ring is present, whereas the presence of a methyl group leads to opening of the 1,3,4-ozadiazole ring and transhydrazinolysis occurs.

EXPERIMENTAL

IR spectra of KBr tablets were recorded on a Specord IR-75 instrument, ^1H NMR spectra were recorded with a Varian 300VXR (300 MHz) with TMS as internal standard. Monitoring of the course of reactions and of purity of substances was carried out by TLC on Silufol UV-254 plates with a benzene–ethyl acetate–methanol, 10:7:1 solvent system and development with iodine vapor. Acetonitrile was purified and dried by boiling over P_2O_5 with subsequent distillation from potash.

Ethyl 2,6-Dimethyl-5-(5-R-1,3,4-oxadiazol-2-yl)pyridine-3-carboxylates 5b-i and 2,6-Dimethyl-3,5-bis[5-(2-fluorophenyl)1,3,4oxadiazol-2-yl]pyridine (6c) (General Method). To a solution of the corresponding hydrazide **1** or **2** (1 mmol) in dry acetonitrile (25 ml) was added acid chloride (1 mmol for each hydrazide group) and the mixture was boiled for 1 h. After removing of the solvent in vacuum, the residue was dissolved with heating in freshly distilled $POCl_3$ (30 ml) until a clear solution was obtained. The phosphorus oxychloride was removed in vacuum, and the solid or resinous residue was dissolved in the minimal amount of methanol and neutralized with cooling with ice with saturated sodium hydrogen carbonate to a pH of 8-9. The precipitate was filtered off, washed several times with water, and recrystallized from 2-propanol or octane.

Ethyl 2,6-Dimethyl-5-(5-methyl-1,3,4-oxadiazol-2-yl)nicotinate (5h) was separated by mixing the oily residue, after removing the $POCl_3$, with ground ice and neutralized with 10% ammonia solution to a mildly basic reaction. The reaction product was extracted with chloroform, the chloroform solution was dried over $MgSO_4$. After removal of the solvent the residue was crystallized from octane.

2,6-Dimethyl-3,5-bis(5-methyl-1,3,4-oxadiazol-2-yl)pyridine (6h). Acetic anhydride (0.2 g, 2 mmol) was added to a solution of dihydrazide **2** (0.22 g, 1 mmol) in dry acetonitrile (10 ml) and boiled for 1 h. The solvent was evaporated in vacuum, $POCl_3$ (15 ml) was added to the residue and boiled for 30 min. The phosphorus oxychloride was evaporated in vacuum, the resinous residue was dissolved in the minimum amount of ethanol and the resulting solution was poured into a saturated aqueous solution of $NaHCO_3$. The precipitate was recrystallized from a mixture of ethanol and water.

Ethyl 2,6-Dimethyl-5-(1,3,4-oxadizol-2-yl)pyridine-3-carboxylate (5a). A solution of hydrazide **1** (1.00 g, 4 mmol) in ethyl orthoformate (10 ml) was boiled for 18 h, cooled in an ice bath, the colorless crystals were filtered off and recrystallized from a minimum amount of ethanol.

2,6-Dimethyl-3,5-bis(1,3,4-oxadiazol-2-yl)pyridine (6a). A solution of dihydrazide **2** (1.00 g, 5 mmol) in ethyl orthoformate (15 ml) was boiled for 24 h, cooled and kept in a refrigerator for 1-2 days, the colorless crystalline precipitate was filtered off and recrystallized from ethanol.

2,6-Dimethyl-5-(5R-1,3,4-oxadiazol-2-yl)pyridine-3-carboxyhydrazides 7a-f,i (General Method). 85% hydrazine hydrate (10 ml) was added to a solution of the corresponding 5-(5-R-1,3,4-oxadiazol-2-yl)-pyridine **5a-f,i** (1 mmol) in the minimum amount of ethanol. The solution was boiled for 2-3 h. At the end of the reaction (monitored by TLC) the hot reaction mixture was filtered and cooled. The pyridine-3-carboxyhydrazides **7a-f, i** were recrystallized from DMF.

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