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# NITROPYRIDINES. 9*. SYNTHESIS OF SUBSTITUTED 3-AMINO-5-NITROPYRIDINES

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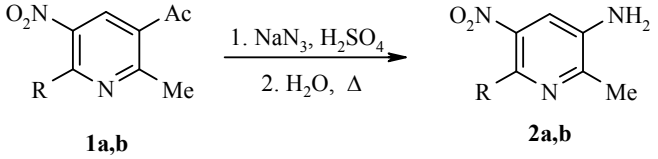
*Substituted 3-amino-5-nitropyridines have been synthesized by the Curtius, Schmidt, and Hofmann reactions.*

**Keywords**: 3-acetyl-5-nitropyridine, nitronicotinic acid, nitronicotinamide, 5-nitropyridines, 3-pyridylcarbamates, the Curtius, Schmidt, and Hofmann reactions and their modifications.

The discovery of bacteriostatic activity of Sulfidine was the stimulus for the study of the chemistry of isomeric aminopyridines with the objective of converting them into promising sulfamide preparations [2].

Aminopyridines with nitrogroups on the pyridine nucleus have been insufficiently well studied, which is explained by their low accessibility. Exceptions are 2-amino-3-nitro- and 5-nitropyridines and their derivatives, which are accessible thanks to the activity of 2-aminopyridine to electrophilic substitution [3]. The number of publications connected with the preparation of substituted 3-amino-5-nitropyridines is limited to those describing the synthesis of 3-amino-5-nitropyridine [4-7], 3-amino-2-methyl-5-nitropyridine [8], and 3-amino-5-nitrocollidine [9].

The objective of the present work was the synthesis of the previously unknown 3-amino-5-nitropyridines **2a**,**b**, **4**, **6b**,**c**, and **9a-c** based on the available nitropyridines **1a**,**b**, **3a-c**, and **7a-c** which contain acetyl, amide, and carboxyl groups at position 3 of the pyridine nucleus, The choice of the variant of the sextet rearrangement in the synthesis of 3-amino-5-nitropyridines was determined by the availability of the corresponding 5-nitropyridines. 3-Amino-5-nitropyridines **2a**,**b** were obtained by the Schmidt reaction starting from 3-acetyl-5-nitropyridines **1a**,**b** by a one-stage synthesis which we had developed earlier [10, 11]:




**1a,b** **2a,b**

**1,2 a** R = Me, **b** R = Ph

* For Communication 8, see [1].

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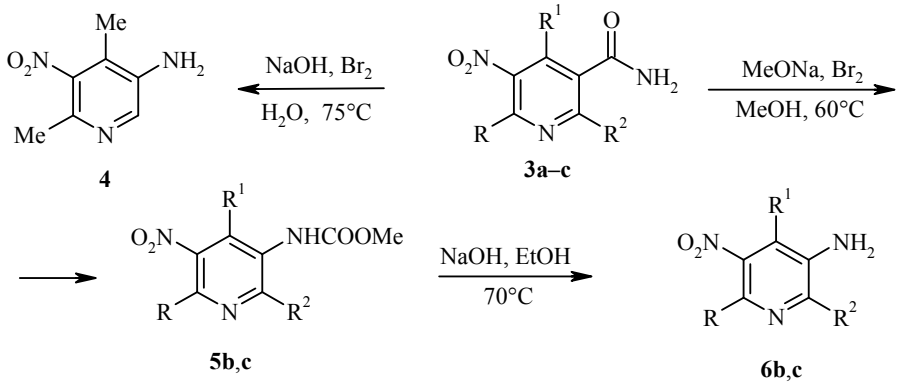
In the synthesis of 3-amino-4,6-dimethyl-5-nitropyridine (**4**) from the previously known nicotinamide **3a** we successfully used the classical Hofmann reaction (NaOBr) [12]. Under these conditions the nitronicotinamides **3b**,**c** and **7a**,**c**, which are insoluble in aqueous alkali, did not rearrange to the corresponding 3-amino-5-nitropyridines. In this connection we turned to a known modification of the Hofmann reaction in which a variety of oxidizing reagents were used (MeOBr, $Pd(OAc)_4$, NBS-$Hg(OAc)_2$, NBS-DBU, $PhI(OCOCF_3)_2$, $PhI(OAc)_2$, PhIO, PhI(OTs)OH, $BnN^+Me_3Br_3^-$, $Bu_4N^+Br_3^-$) [13,19].

To prepare the 3-amino-5-pyridines **6b**,**c** from the corresponding amides **3b**,**c** a two-stage modification of the Hofmann reaction was used (a solution of MeOBr in methanol).The splitting of the 3-pyridylcarbamates **5b**,**c** was carried out with the help of an aqueous-ethanolic solution of sodium hydroxide on heating.




**3 a** R = $R^1$ = Me, $R^2$ = H; **3,5,6 b** R = $R^1$ = Ph, $R^2$ = Me; **c** R = Ph, $R^1$ = *p*-$MeOC_6H_4$, $R^2$ = Me

In the synthesis of the 3-pyridylcarbamates **8a-c** by the Hofmann reaction the most effective reagent for the oxidative rearrangement of the amides **7a-c** was diacetoxyiodobenzene. The carbamate **8a** was stable on heating in aqueous alkali but it was successfully hydrolyzed to the 3-aminopyridine **9a** on prolonged heating in a mixture of acetic and sulfuric acids.

TABLE 1. Spectral Characteristics of the Compounds Synthesized

| Com-pound | IR spectrum, ν, $cm^{-1}$ | $^1H$ NMR spectrum, δ, ppm (*J*, Hz)* |
|---|---|---|
| 1 | 2 | 3 |
| **2a** | 1346, 1520 ($NO_2$), 3420, 3510 ($NH_2$) | 2.35 (3H, s, 2-$CH_3$); 2.55 (3H, s, 6-$CH_3$); 5.56 (2H, s, $NH_2$); 7.55 (1H, s, H-4) |
| **2b** | 1325, 1527 ($NO_2$), 3400, 3505 ($NH_2$) | 2.51 (3H, s, 2-$CH_3$); 3.96 (2H, s, $NH_2$); 7.34 (1H, s, H-4); 7.36-7.49 (5H, m, $C_6H_5$) |
| **4** | 1370, 1530 ($NO_2$), 3395, 3500 ($NH_2$) | 1.92 (3H, s, 4-$CH_3$); 2.20 (3H, s, 6-$CH_3$); 5.55 (2H, s, $NH_2$); 7.95 (1H, s, H-2) |
| **5b** | 1340, 1540 ($NO_2$), 1740 (CO), 3410 (NH) | 2.56 (3H, s, 2-$CH_3$); 3.47 (3H, s, $CO_2CH_3$); 7.19-7.26 (2H, m, 4,6-$C_6H_5$); 7.45-7.58 (8H, m, 4,6-$C_6H_5$); 9.13 (1H, br. s, NH) |
| **5c** | 1362, 1514 ($NO_2$), 1699 (CO), 3274 (NH) | 2.54 (3H, s, 2-$CH_3$); 3.50 (3H, s, $CO_2CH_3$); 3.79 (3H, s, $OCH_3$); 7.00-7.05 (2H, m, $J_{AX}$ = 8.6, 4-$C_6H_4$, AA'XX'); 7.12-7.17 (2H, m, $J_{AX}$ = 8.6, 4-$C_6H_4$, AA'XX'); 7.47-7.57 (5H, m, 6-$C_6H_5$); 9.08 (1H, br. s, NH) |
| **6b** | 1360, 1520 ($NO_2$), 3400, 3490 ($NH_2$) | 2.54 (3H, s, 2-$CH_3$); 3.69 (2H, s, $NH_2$); 7.36-7.55 (10H, m, 4,6-$C_6H_5$) |
| **6c** | 1361, 1525 ($NO_2$), 3382, 3479 ($NH_2$) | 2.54 (3H, s, 2-$CH_3$); 3.79 (2H, s, $NH_2$); 3.84 (3H, s, $OCH_3$); 6.99-7.03 (2H, m, $J_{AX}$ = 8.8, 4-$C_6H_4$, AA'XX'); 7.24-7.27 (2H, m, $J_{AX}$ = 8.8, 4-$C_6H_4$, AA'XX'); 7.35-7.42 (3H, m, 6-$C_6H_5$); 7.53-7.57 (2H, m, 6-$C_6H_5$) |
| **8a** | 1335, 1528 ($NO_2$), 1719 (CO), 3217 (NH) | 1.31 (3H, t, *J* = 7.2, $CH_2\underline{CH_3}$); 2.85 (3H, s, 6-$CH_3$); 4.25 (2H, q, *J* = 7.2, $C\underline{H_2}CH_3$); 6.87 (1H, s, NH); 7.53-7.61 (5H, m, $C_6H_5$); 9.18 (1H, s, H-4) |

TABLE 1 (continued)

| 1 | 2 | 3 |
|---|---|---|
| **8b** | 1340, 1530 ($NO_2$), 1725 (CO), 3440 (NH) | 1.31 (3H, t, $J$ = 7.2, $CH_2C\underline{H}_3$); 4.26 (2H, q, $J$ = 7.2, $C\underline{H}_2CH_3$); 7.05 (1H, s, NH); 7.57-7.63 (5H, m, $C_6H_5$); 9.17 (1H, d, $^4J$ = 2.4, H-6); 9.38 (1H, d, $^4J$ = 2.4, H-4) |
| **8c** | 1357, 1532 ($NO_2$), 1715 (CO), 3207 (NH) | 1.03 (3H, br. s, $CH_2C\underline{H}_3$); 2.64 (3H, s, 6-$CH_3$); 3.92 (2H, br. s, $C\underline{H}_2CH_3$); 6.33 (1H, br. s, NH); 6.57 (1H, dd, $^3J_{4,3}$ = 3.3, $^3J_{4,5}$ = 1.9, H-4 Fur); 6.82 (1H, d, $^3J_{3,4}$ = 3.3, H-3 Fur); 7.43-7.51 (3H, m, $C_6H_5$); 7.60-7.66 (3H, m, $C_6H_5$, H-5 Fur) |
| **9a** | 1360, 1540 ($NO_2$), 3380, 3460 ($NH_2$) | 2.77 (3H, s, 6-$CH_3$); 4.03 (2H, s, $NH_2$); 7.43-7.53 (3H, m, $C_6H_5$); 7.66-7.70 (3H, m, $C_6H_5$, H-4) |
| **9b** | 1347, 1530 ($NO_2$), 3395, 3500 ($NH_2$) | 4.28 (2H, s, $NH_2$); 7.31-7.78 (6H, m, $C_6H_5$, H-4); 9.17 (1H, s, H-6) |
| **9c** | 1351, 1529 ($NO_2$), 3320, 3398 ($NH_2$) | 2.53 (3H, s, 6-$CH_3$); 4.46 (2H, s, $NH_2$); 6.60 (1H, dd, $^3J_{4,3}$ = 3.5, $^3J_{4,5}$ = 1.7, H-4 Fur); 6.79 (1H, dd, $^3J_{3,4}$ = 3.5, $^4J_{3,5}$ = 0.5, H-3 Fur); 7.46-7.57 (3H, m, $C_6H_5$); 7.61 (1H, dd, $^3J_{5,4}$ = 1.7, $^4J_{5,3}$ = 0.5, H-5 Fur); 7.65-7.69 (2H, m, $C_6H_5$) |
| **10** | 1531, 1366 ($NO_2$), 1263, 1721, 3067, 3447 (COOH) | 2.68 (3H, s, 2-$CH_3$); 7.26-7.33 (2H, m, 4,6-$C_6H_5$); 7.43-7.50 (6H, m, 4,6-$C_6H_5$); 7.53-7.59 (2H, m, 4,6-$C_6H_5$); 13.55 (1H, br. s, COOH) |

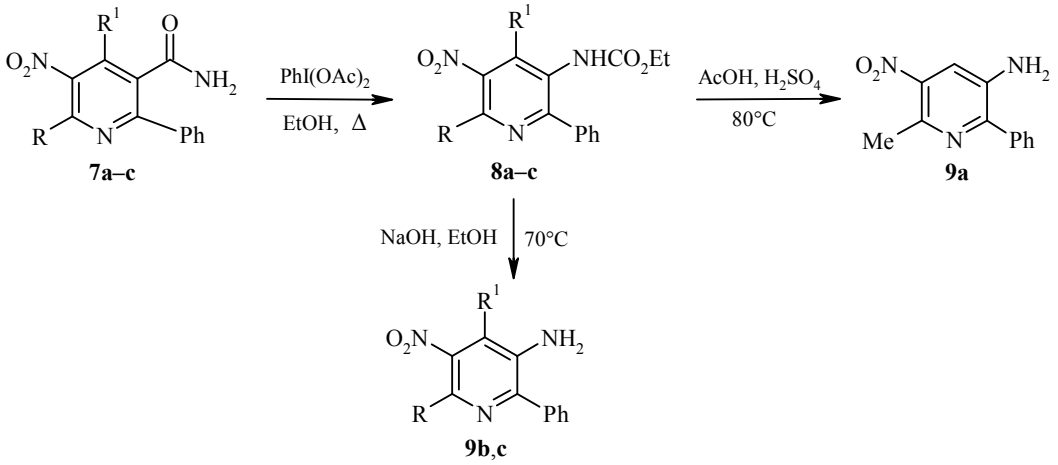
* $^1$H NMR spectra were taken in DMSO-$d_6$ (compounds **2a**, **4**, **5b**,**c**, **8a-c**, and **10**) and in $CDCl_3$ (compounds **2b**, **6b**,**c**, and **9a-c**).

As an alternative route to the synthesis of the 3-aminopyridine **6b** we investigated the possibility of obtaining it *via* the Curtius reaction. The hydrazide of the pyridinecarboxylic acid was not obtained by reaction of its ester with hydrazine hydrate (even on heating in an ampoule), nor by the reaction of the corresponding acid chloride with hydrazine hydrate. In this connection we chose the modified Curtius reaction using an azide – diphenylphosphorylazide – for the synthesis of the azide of acid **10** [20-25]. It should be noted that in this reaction the 3-aminopyridine **6b** was obtained in low yield (25%) and required chromatographic purification.

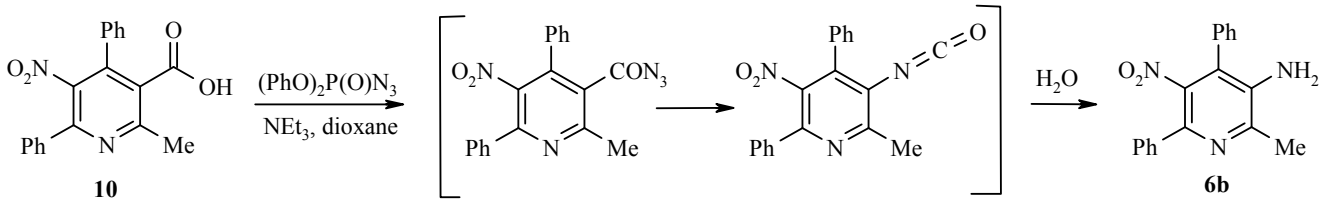
TABLE 2. Mass Spectra of Compounds **2a**,**b**, **4**, **5b**, **8b**, and **9b**

| Com-pound | *m/z* (*I*, %)* |
|---|---|
| **2a** | 168 $[M+1]^{+\bullet}$ (9), 167 $[M]^{+\bullet}$ (100), 151 (10), 150 (45), 122 (29), 121 (88), 120 (20), 119 (17), 107 (11), 106 (24), 105 (22), 104 (17), 95 (23), 94 (46), 93 (13), 80 (19), 79 (16), 78 (23), 77 (23), 68 (18), 67 (42), 54 (15), 53 (31), 52 (37), 51 (25), 44 (17), 42 (21), 40 (17) |
| **2b** | 230 $[M+1]^{+\bullet}$ (16), 229 $[M]^{+\bullet}$ (100), 199 (23), 184 (24), 172 (12), 171 (17), 168 (26), 156 (12), 141 (11), 140 (12), 129 (15), 128 (12), 115 (50), 114 (24), 113 (10), 81 (39), 77 (13), 70 (10), 68 (12), 42 (17), 28 (11) |
| **4** | 168 $[M+1]^{+\bullet}$ (9), 167 $[M]^{+\bullet}$ (100), 150 (29), 121 (68), 120 (13), 119 (10), 95 (15), 94 (23), 93 (11), 81 (12), 80 (29), 67 (20), 66 (15), 65 (15), 54 (15), 53 (27), 42 (12), 39 (13) |
| **5b** | 364 $[M+1]^{+\bullet}$ (23), 363 $[M]^{+\bullet}$ (100), 332 (19), 331 (70), 314 (26), 303 (11), 302 (32), 301 (43), 300 (11), 286 (22), 285 (28), 275 (14), 274 (53), 273 (16), 272 (11), 260 (15), 258 (24), 257 (14), 256 (15), 255 (12), 242 (19), 231 (13), 230 (32), 216 (18), 215 (11), 214 (19), 202 (11), 190 (19), 189 (48), 188 (12), 128 (11), 127 (13), 115 (12), 81 (24), 77 (13) |
| **8b** | 288 $[M+1]^{+\bullet}$ (16), 287 $[M]^{+\bullet}$ (100), 286 (33), 242 (13), 241 (14), 228 (10), 214 (54), 213 (18), 198 (11), 182 (17), 169 (16), 168 (68), 167 (22), 141 (13), 140 (24), 115 (11), 104 (12), 77 (15) |
| **9b** | 216 $[M+1]^{+\bullet}$ (11), 215 $[M]^{+\bullet}$ (92), 214 (100), 185 (24), 184 (26), 169 (13), 168 (40), 115 (33), 104 (16), 77 (15), 66 (15), 44 (10) |

* Peaks cited with $I > 10\%$




**7,8 a** R = Me, $R^1$ = H; **7–9 b** R = $R^1$ = H; **c** R = Me, $R^1$ = 2-Fur




Spectral data for the compounds synthesized for the first time **2a**,**b**, **4**, **5b**,**c**, **8a-c**, **9a-c**, and **10** are cited in Tables 1 and 2, elemental analyses are cited in the experimental section.

## EXPERIMENTAL

The IR spectra of compounds **5c**, **6c**, **8a**, **8c**, and **9c** were recorded on an Infralum FT-801 (with an attachment for a single-pass breakdown of the internal refraction), compound **10** on a Simex FT-801 (in a KBr tablet) and the remaining compounds on a Specord IR-75 in $CHCl_3$. $^1$H NMR spectra of compounds **2a**,**b**, **4**, **6b**, **8b**, and **9b** were recorded on a Bruker AC-200 (200 MHz, TMS), compounds **5b**,**c** and **8a**,**c** on a Bruker AC-400 (400 MHz, using the solvent as internal standard), and compounds **6c**, **9a**,**c**, and **10** on a Bruker Advance DRX-400 (400 MHz, using the solvent as internal standard). Mass spectra were recorded with an Agilent 5973N mass spectrometer (ionizing energy 70eV, evaporator temperature 230-250°C). Elemental analyses were carried out with a Perkin-Elmer C,H,N-analyzer. Silica gel Merck 60A, 0.060-0.200 mm, was used for column chromatography. Monitoring of the course of reactions and purity of the compounds obtained was by TLC on Silufol UV-254 plates.

**3-Aminopyridines 2a,b (General Method)**. Sodium azide (0.43g, 6.6 mmol) was added by portions with stirring to a mixture of the corresponding 3-acetylpyridine (6 mmol) [10,11] in 3 ml conc. $H_2SO_4$. After all of the $NaN_3$ had been added the reaction mixture was stirred at room temperature for 12 h, then ice (12 g) was added and the mixture was heated for 12 h, cooled. Neutralized with aqueous ammonia, and the precipitate was filtered off.

**3-Amino-2,6-dimethyl-5-nitropyridine (2a).** Yield 66%; mp 137-138° (toluene). Found, %: C 50.70; H 5.28; N 24.98. $C_7H_9N_3O_2$. Calculated, %: C 50.29; H 5.43; N 25.14.

**3-Amino-2-methyl-5-nitro-6-phenylpyridine (2b).** Yield 70%; mp 95-96°C (heptane). Found, %: C 62.75; H 4.82; N 18.47. $C_{12}H_{11}N_3O_2$. Calculated, %: C 62.87; H 4.84; N 18.33.

**3-Amino-4,6-dimethyl-5-nitropyridine (4).** Bromine (0.6 ml, 12 mmol) was added dropwise to a solution of NaOH (1.5 g, 37.5 mmol) in water (16 ml) at -5°C, then nicotinamide **3a** [1] (1.5 g, 7.7 mmol) was added, stirred with cooling for 30 min, then slowly heated to 75°C and kept at this temperature for 2.5 h. The precipitate formed was filtered off, washed with 10% HCl (20 ml), boiled with activated charcoal, cooled, neutralized with aqueous ammonia, and the residue was recrystallized from toluene. Yield 63%; mp 173-174°C (toluene). Found, %: C 50.12; H 5.32; N 24.98. $C_7H_9N_3O_2$. Calculated, %: C 50.29, H 5.43, N 25.14.

**Carbamates 5b,c (General Method)**. To a mixture of the corresponding nicotinamide **3b**,**c** [1] (3 mmol) and a solution of sodium methoxide (prepared from 0.23 g (10 mmol) Na and 17.5 ml absolute methanol) at 0°C, $Br_2$ (0.23 ml, 4.5 mmol) was added dropwise. The reaction mixture was stirred for 1 h at room temperature and 4 h at 60°C, neutralized with 50% AcOH, the precipitate formed was filtered off and recrystallized from ethanol.

**Methyl-(2-methyl-5-nitro-4,6-diphenylpyridin-3-yl)carbamate (5b).** Yield 89%; mp 222-224°C. Found, %: C 66.10, H 4.65, N 11.52. $C_{20}H_{17}N_3O_4$. Calculated, %: C 66.11; H 4.72; N 11.56.

**Methyl-[2-methyl-4-(4-methoxyphenyl)-5-nitro-6-phenylpyridin-3-yl]carbamate (5c).** Yield 87%; mp 244-246°C. Found, %: C 64.08; H 4.91; N 10.72. $C_{21}H_{19}N_3O_5$. Calculated, %: C 64.12; H 4.87; N 10.68.

**Carbamates 8a-c (General Method)**. A mixture of the corresponding nicotinamide **7a-c** [1] (2.5 mmol) and $PhI(OAc)_2$ [14] (0.93 g, 2.9 mmol) was boiled in absolute ethanol (15 ml) (the reaction time is shown below), cooled, the precipitate formed was filtered off and recrystallized from ethanol.

**Ethyl-(6-methyl-5-nitro-2-phenylpyridin-3-yl)carbamate (8a).** Reaction time 3 h. Yield 76%; mp 147-148°C. Found, %: C 59.88; H 5.00; N 14.13. $C_{15}H_{15}N_3O_4$. Calculated, %: C 59.79; H 5.02; N 13.95.

**Ethyl-(5-nitro-2-phenylpyridin-3-yl)carbamate (8b).** Reaction time 4 h. Yield 71%; mp 113-114°C. Found, %: C 58.75; H 4.61; N 14.73. $C_{14}H_{13}N_3O_4$. Calculated, %: C 58.53; H 4.56; N 14.63.

**Ethyl-[4-(2-furyl)-6-methyl-5-nitro-2-phenylpyridin-3-yl]carbamate (8c).** Reaction time 6 h. Yield 71%; mp 194-195°. Found, %: C 62.08; H 4.60; N 11.45. $C_{19}H_{17}N_3O_5$. Calculated, %: C 62.12; H 4.66; N 11.44.

**3-Aminopyridines 6b,c and 9b,c (General Method)**. To a suspension of the corresponding carbamate **5b**,**c**, **8b**,**c** (0.6 mmol) in ethanol (1.4 ml) a solution of NaOH (0.18 g, 4.5 mmol) in water (0.4 ml) was added. The reaction mixture was heated at 70°C (reactions times are given below), cooled, diluted with water and the crystals formed were filtered off.

**3-Amino-2-methyl-5-nitro-4,6-diphenylpyridine (6b).** Reaction time 6 h. Yield 91%; mp 206-207°C (heptane). Found, %: C 70.50; H 4.85; N 13.62. Calculated; %: C 70.81; H 4.95; N 13.76.

**3-Amino-4-(4-methoxyphenyl)-2-methyl-5-nitro-6-phenylpyridine (6c)** was obtained by heating for 24 h. After cooling, the crystals which precipitated were filtered off and washed with water. The filtrate was diluted with water and the starting carbamate **5c** was separated. The yield of aminopyridine **6c** was 78%; mp 225-227°C (ethanol). Found, %: C 68.04; H 5.21; N 12.48. $C_{19}H_{17}N_3O_3$. Calculated, %: C 68.05; H 5.11; N 12.53.

**3-Amino-5-nitro-2-phenylpyridine (9b).** Reaction time 2 h. Yield 62%; mp 216-217°C (heptane). Found, %: C 61.52; H 4.33; N 20.01. $C_{11}H_9N_3O_2$. Calculated, %: C 61.39; H 4.22; N 19.53.

**3-Amino-4-(2-furyl)-6-methyl-5-nitro-2-phenylpyridine (9c).** Reaction time 24 h. Yield 91%; mp 127-128°C (2-propanol). Found, %: 64.80; H 4.33; N 14.01. $C_{16}H_{13}N_3O_3$. Calculated, %: C 65.08; H 4.44; N 14.23.

**3-Amino-6-methyl-5-nitro-2-phenylpyridine (9a).** A solution of carbamate **7a** (0.5 g, 1.7 mmol) in a mixture of glacial acetic acid (1.4 ml) and conc. $H_2SO_4$ (0.8 ml) was heated at 80° for 84 h, water (8 ml) was added and the mixture was stirred for 8 h at room temperature. The reaction mixture was filtered, the filtrate was neutralized with aqueous ammonia, the crystals which precipitated were filtered off and recrystallized. Yield 0.38 g (94%); mp 105-106°C (heptane). If the reaction was carried out at 100°C for 36 h or boiled for 2 h, the yield of aminopyridine was reduced to 70-65%. In these cases the reaction product needed to be purified by chromatography. Found, %: C 62.75; H 4.82; N 18.47. $C_{12}H_{11}N_3O_2$. Calculated, %: C 62.87; H 4.84; N 18.33.

**2-Methyl-5-nitro-4,6-diphenylnicotinic acid (10).** A mixture of ethyl 2-methyl-5-nitro-4,6-diphenylnicotinate [26] (1.45 g, 4 mmol), ethanol (10 ml), KOH (1.12 g, 20 mmol) in water (2 ml) was boiled for 3-4 h, diluted with water and acidified with conc. HCl to pH 3, the precipitate was filtered off and recrystallized. Yield 1.27 g (95%); mp 274-275°C (ethanol). Found, %: C 68.29; H 4.17; N 8.43. $C_{19}H_{14}N_2O_4$. Calculated, %: C 68.26; H 4.22; N 8.38.

**3-Amino-2-methyl-5-nitro-4,6-diphenylpyridine (6b)** (modified Curtius reaction). A mixture of diphenylchlorophosphate (11.1 g, 44 mmol), $NaN_3$ (3.2 g, 49 mmol) and absolute dioxane (90 ml) was stirred at room temperature for 24 h. Nicotinic acid **10** (3.7 g, 11 mmol) and absolute $Et_3N$ (6 ml) were then added and the mixture was boiled for 24 h. After addition of water (10 ml), the reaction mixture was boiled for another 1 h, then made basic with 10% KOH solution and most of the solvent was removed in vacuum. The residue was diluted with water, extracted with benzene, the extract was dried over $MgSO_4$, and after removing the solvent it was purified by column chromatography using 9:1 chloroform–ethyl acetate as eluent. Yield 0.84 g (25%).

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