The Reaction of Pyridine N-Oxide with Acetic Anhydride in Anisole and in Benzonitrile[1a,b]

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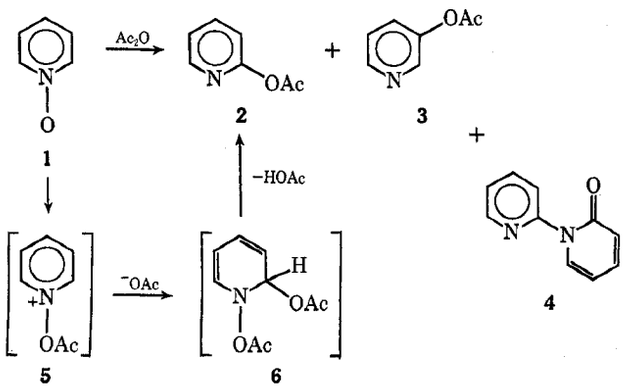
The reaction of pyridine N-oxide and acetic anhydride in anisole provided 22% of a mixture of 2-(*o*-, *m*-, and *p*-methoxyphenyl)pyridine in relative yields of 50.7:15.5:33.9, respectively. The same reaction in benzonitrile yielded 8.5% of N-2-pyridylacetamide and trace quantities of N-2-pyridylbenzamide and 2-(*m*- and *p*-cyanophenyl)pyridine (relative yields of the latter, 71:29). Under the reaction conditions, N-2-pyridylbenzamide is converted to N-2-pyridylacetamide. The products are thought to arise by attack of solvent on an intermediate with an electrophilic site at the 2 position of the pyridine ring.

The reaction of pyridine N-oxide (**1**) with acetic anhydride yields mainly 2-acetoxypyridine (**2**),[2,3] but several side products, including 3-acetoxypyridine (**3**) and N-(2′-pyridyl)-2-pyridone (**4**), are also formed.[4]

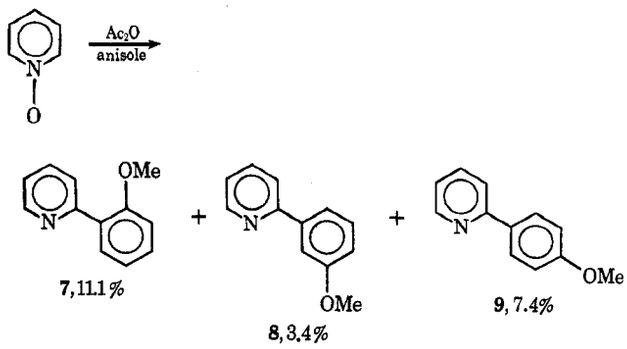


Mechanistic studies[3,5] indicate that the 2-acetoxypyridine is probably produced by way of acetate ion attack on the N-acetoxypyridinium ion **5** to yield the dihydropyridine **6**, which loses acetic acid to give **2**. The detailed mode of loss of acetic acid is unknown as are the mechanisms of formation of the minor products.

In the hope of being able to trap intermediates in the acetic anhydride–pyridine N-oxide reaction, the latter was performed in anisole and in benzonitrile. These solvents have recently been found useful in trapping picolyl cations produced in the reaction of acetic anhydride with 2- and 4-picoline N-oxide.[6] Cations substitute into anisole very predominantly at the ortho and para positions,[6,7] whereas radical attack on the ring yields more meta- than para-substituted product.[8] Organic cations attack benzonitrile at the nitrogen atom[6,9] and any electrophilic attack that does occur on the ring leads mainly to meta-substituted products; radical attack occurs readily at the ortho and para positions of the ring.[8]

Results

Anisole.—The reaction of pyridine N-oxide with acetic anhydride in refluxing anisole for 6 hr yielded, in addition to 2-acetoxypyridine, a three-component mixture of 2-(methoxyphenyl)pyridines. The product was analyzed by combined gas chromatography–mass spectrometry. The mass spectra of the three isomers are consistent with the assignment as methoxyphenylpyridines. A sample of the major component, which had the shortest glpc retention time, was shown by nmr spectroscopy to possess one α proton on the pyridine ring, thus indicating that the methoxyphenyl group is attached to the 2 position. This isomer was shown to be 2-(*o*-methoxyphenyl)pyridine (**7**) by glpc and mass spectrometric comparison with an authentic sample prepared by the reaction of pyridine with *o*-methoxyphenyllithium followed by air oxidation.[10] The component present in second greatest amount and having the longest retention time was similarly identified as 2-(*p*-methoxyphenyl)pyridine (**9**). The minor component was assigned the structure 2-(*m*-methoxyphenyl)pyridine (**8**) on the basis of the similarity of its mass spectrum to the other two, its retention time (between those of the other two; typical behavior for a meta isomer), and the expectation that the meta isomer would accompany the ortho and para isomers. The yields, as indicated by gas chromatography, are listed below. No other solvent-derived products could be detected.



(1) (a) We wish to thank the donors of the Petroleum Research Fund, administered by the American Chemical Society, for support of this work. (b) Taken from the Ph.D. Thesis of G. L. D., University of Pittsburgh, Pittsburgh, Pa., 1969. (c) NASA Predoctoral Fellow.

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(6) T. Cohen and G. L. Deets, *J. Amer. Chem. Soc.*, **89**, 3939 (1967); T. Cohen and G. L. Deets, *ibid.*, in press.

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(8) J. R. Shelton and C. W. Uzelmeier, *J. Amer. Chem. Soc.*, **88**, 5222 (1966).

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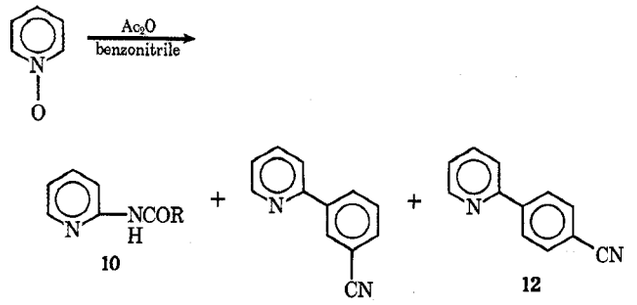
In attempts to improve the yield of anisylpyridines, pyridine *N*-oxide was treated with *p*-toluenesulfonyl chloride in refluxing anisole in the presence and absence of 2,6-lutidine (a potential proton acceptor), and *N*-acetoxypyridinium perchlorate[11] was heated in refluxing anisole in the presence and absence of 2,6-lutidine. None of these reactions produced detectable quantities of anisylpyridines.

Benzonitrile.—When the reaction of pyridine *N*-oxide with acetic anhydride was performed in benzonitrile at 160°, four products which appeared to have resulted from reaction with solvent were detected by combined glpc–mass spectrometry. Two of these had the fragmentation patterns expected of *N*-2-pyridylbenzamide (**10**, R = phenyl) and *N*-2-pyridylacetamide (**10**, R = methyl), and these identifications were confirmed by comparison with authentic samples prepared by treatment of 2-aminopyridine with the appropriate acid chloride. The two remaining products had the fragmentation patterns expected of 2-(cyanophenyl)pyridines. They were identified as 2-(*m*-cyanophenyl)-pyridine (**11**) and 2-(*p*-cyanophenyl)pyridine (**12**) by comparison with samples prepared by decomposition in pyridine of the diazonium ion derived from the appropriate aminobenzonitrile.[12] The yield of *N*-2-pyridylacetamide was 8.5%. That of *N*-2-pyridylbenzamide was less than 1%. The 2-(cyanophenyl)pyridine mixture was also formed in less than 1% yield and the composition was 71% meta and 29% para.



It was also shown that *N*-2-pyridylbenzamide (**10**, R = phenyl) is converted to *N*-2-pyridylacetamide (**10**, R = methyl) under the conditions employed in the reaction of pyridine *N*-oxide with acetic anhydride in benzonitrile.

Discussion

The nature of the products is consistent with the attack on the solvents of some intermediate with an electrophilic site at the 2 position of the pyridine ring. The attack on anisole occurs with ortho/para and meta/para ratios of 1.5 and 0.46, respectively. These values are similar to those (up to 1.9 and 0.37, respectively) which have been found for several electrophilic substitutions in the literature[7] and very different from those (5.3–13.5 and 1.4–5.6, respectively) for some reported examples of radical attack on anisole.[8] However, this evidence by itself does not demand that the attack on anisole be electrophilic rather than radical in nature, since the rate-determining step could occur subsequent to the attack on solvent.

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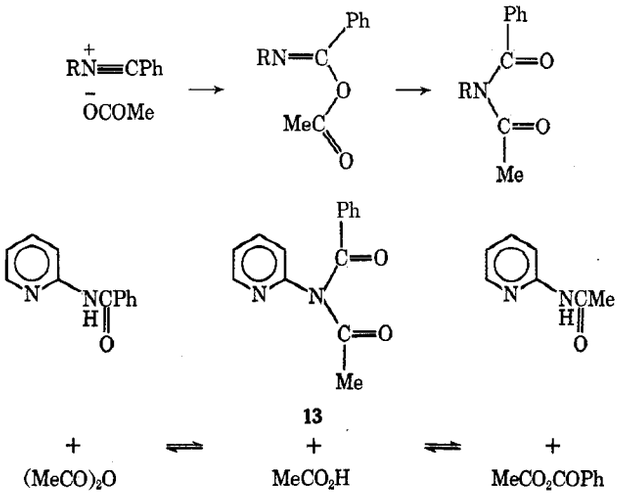
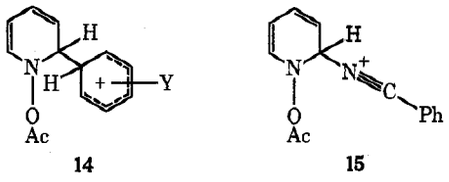
The attack on benzonitrile occurs predominantly at the nitrogen atom (*vide infra*) and the very small quantity of ring-substituted benzonitrile arises mainly from meta attack. As noted above, this pattern is characteristic of cationic attack on benzonitrile.

According to the results of our experiments on the trapping of picolyl cations by benzonitrile,[6] one might expect *N*-acetyl-*N*-(2-pyridyl)benzamide (**13**) to result from attack of the type of electrophile discussed above on the nitrogen atom of benzonitrile. Its expected mode of formation is shown below (R represents the 2-pyridyl group or its precursor). However, the expected imide **13** must be readily deacylated, as shown, under the reaction conditions. This follows from our finding that *N*-(2-pyridyl)benzamide is converted to *N*-(2-pyridyl)acetamide under the reaction conditions; this exchange presumably proceeds through the intermediate formation of **13**. Thus, the production of *N*-(2-pyridyl)acetamide in 8.5% yield indicates that benzonitrile has reacted with an electrophilic site at the 2 position of the pyridine nucleus.

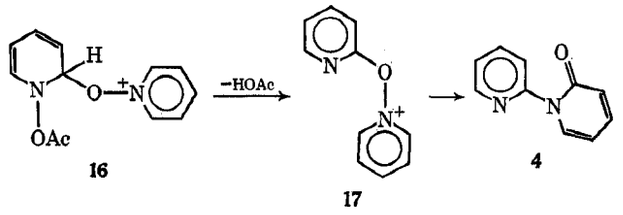


The simplest hypothesis is that these products arise by attack of the *N*-acetoxypyridinium ion on the nucleophilic solvents to produce species such as **14** (Y = OMe or CN) and **15**. In the case of **14**, rearomatization of the pyridine ring by the loss of acetic acid and of the

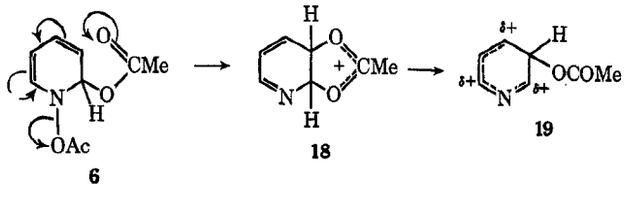


substituted ring by proton loss would yield the 2-arylpyridine products. Loss of acetic acid from **15** and subsequent reactions of the resulting nitrilium ion as outlined above would lead to the amide products. One attractive feature of this scheme is that it is consistent with the presently accepted mechanism for the production of 2-acetoxypyridine, in which acetate ion is the nucleophile which attacks the 2 position. Another is that if pyridine *N*-oxide is the nucleophile which attacks **5** then the known product, *N*-(2′-pyridyl)-2-pyridone (**4**), would result *via* intermediates **16** and **17**. The latter is almost certainly an intermediate in the

known reaction of 2-bromopyridine with pyridine N-oxide to produce **4**.[13]



If solvent attack is indeed occurring on the N-acetoxypyridinium ion **5**, then the acetate counterion appears to be an important competitor. However, when this ion was replaced by the much less nucleophilic perchlorate ion in anisole, no solvent-derived products were obtained even when 2,6-lutidine was present as a potential proton acceptor. The failure of this reaction and of the reaction of pyridine N-oxide with p-toluenesulfonyl chloride in anisole to produce solvent-derived products suggests that a step following nucleophilic attack on the 2 position of a pyridinium cation may be rate determining in these cases. Another possibility is that acetate ion plays an important role in the formation of some other intermediate which is responsible for the solvent capture in the reaction of pyridine N-oxide with acetic anhydride. A possible candidate is **19** which could be produced from the intermediate **6** by the sequence shown (the conversion of **6** to **18** could be stepwise or concerted) and which may also be converted to the minor product 3-acetoxypyridine (**3**) by a proton loss.



Experimental Section

General.—Melting points were determined in a Kofler block utilizing a stage calibrated thermometer and are thus corrected. Boiling points are uncorrected. Infrared spectra were determined on a Beckman IR-8 spectrophotometer. Proton magnetic resonance spectra are for deuteriochloroform solutions and were determined on a Varian A-60 instrument; chemical shifts are reported on the τ scale relative to internal tetramethylsilane. Analytical gas chromatography was performed on F & M 1609 or Varian 1520 A instruments equipped with flame ionization detectors and Disc Integrators. The glpc results are reported as follows: peak no., compound name (retention time in minutes). For determining yields, the flame responses of authentic samples were calibrated against those of suitable standards. Isomers were assumed to have identical flame responses; this was shown to be true in several cases. Mass spectra were determined at 70 eV on a LKB-9000 combined gas chromatograph–mass spectrometer. The m/e values are reported for major peaks followed in parentheses by the per cent of the base peak.

Reaction of Pyridine N-Oxide with Acetic Anhydride in Anisole.—A solution of 12.3 g (120 mmol) of acetic anhydride and 7.8 g (83 mmol) of pyridine N-oxide in 50 ml of anisole was heated at reflux for 6 hr. A portion of the cooled reaction mixture was extracted with 10% hydrochloric acid (one 25-ml and three 15-ml portions). This acidic extract was extracted with ether (three 5-ml portions), made basic with sodium carbonate, and extracted with ether (two 25-ml portions) and chloroform (three 25-ml portions). The combined organic extract was dried over magnesium sulfate, concentrated, and examined by glpc on a 10 ft × 0.125 in. 3% OV-17 column at an initial temperature of 100° with a programmed rise of 4°/min: 1, 2-acetoxypyridine (12.2); 2, 2-(o-methoxyphenyl)pyridine (19.9); 3, 2-(m-methoxyphenyl)pyridine (21.1); 4, 2-(p-methoxyphenyl)-pyridine (21.8). The identification of 2-acetoxypyridine is based on its mass spectrometric fragmentation pattern which exhibited major peaks at m/e 137, 95, 67, and 43. The mass spectra of the other peaks are: peak 2, m/e 185 (100), 184 (93), 156 (55), 155 (81), 154 (92), 153 (11), 142 (16), 128 (51), 127 (15), 115 (18), 89 (16), 80 (96), 79 (18), 78 (24), 77 (28), 64 (10), 63 (23), 62 (14), 52 (14), 51 (29), 50 (15), 39 (32); peak 3, m/e 185 (92), 184 (100), 156 (33), 155 (59), 154 (55), 143 (10), 142 (12), 115 (12), 95 (14), 89 (11), 78 (17), 77 (12), 63 (12), 52 (11), 51 (19), 50 (11), 39 (21); peak 4, m/e 185 (100), 184 (9), 154 (8), 143 (39), 142 (20), 115 (8), 63 (9), 51 (10), 39 (11). These spectra are consistent with those expected for methoxyphenylpyridines. The nmr spectrum of a sample of peak 2 isolated by preparative glpc shows τ 1.70 (q, single 2-pyridyl proton), 2.47–3.50 (m, 7 aromatic protons), 6.43 (s, 3 methyl protons). The 2-o- and 2-p-methoxyphenylpyridine were found to be identical by glpc and mass spectrometric comparison with authentic samples. The relative yields of ortho:meta:para were found to be 50.7:15.5:33.9. In another run using 4.16 g (43.7 mmol) of pyridine N-oxide and 8.34 g (81.7 mmol) of acetic anhydride in 25 ml of anisole, the yield of 2-(methoxyphenyl)pyridines was found to be 22%. No extractions were used in this run and triphenylmethane was used as the glpc standard. The analysis was performed at 200°.

Reaction of Pyridine N-Oxide with Acetic Anhydride in Benzonitrile.—A solution of 4.4 g (46 mmol) of pyridine N-oxide and 6.6 g (65 mmol) of acetic anhydride in 50 ml of benzonitrile was heated for 6 hr at a bath temperature of 160°. A portion of the reaction mixture was worked up by the same extraction techniques described above for the same reaction in anisole. The glpc analysis was also performed in the same way: 1, N-2-pyridylacetamide (11.9); 2, probable structure N-(2′-pyridyl)-2-pyridone (19.8); 3, 2-(m-cyanophenyl)pyridine (20.8); 4, 2-(p-cyanophenyl)pyridine (21.1); 5, N-2-pyridylbenzamide (22.3). The material constituting the first peak had the same retention time and mass spectrum as those of an authentic sample: mass spectrum m/e 136 (23), 94 (100), 78 (10), 67 (81), 43 (42), 39 (16). The probable structure for the material in the second peak is derived from its mass spectrum and the fact that this substance has been noted previously as a product of this reaction:[4] mass spectrum m/e 172 (100), 171 (16), 144 (20), 118 (60), 117 (12), 79 (60), 78 (48), 52 (26), 51 (40), 50 (12), 40 (36), 39 (20). The materials constituting peaks 3–5 had identical glpc behavior and mass spectra with those of authentic samples. The mass spectra follow: peak 3, m/e 180 (100), 179 (47), 153 (9), 152 (8), 52 (7), 51 (14), 50 (8), 39 (8); peak 4, m/e 180 (100), 179 (52), 153 (11), 152 (10), 52 (11), 51 (17), 50 (11), 39 (25); peak 5, m/e 198 (10), 197 (7), 170 (16), 169 (37), 106 (9), 105 (100), 78 (12), 77 (78), 51 (25), 39 (12). The relative yields of 2-(m-cyanophenyl)- and 2-(p-cyanophenyl)pyridine were determined by glpc to be 71 and 29%, respectively. In another run using 33.2 mmol of pyridine N-oxide and 52.1 mmol of acetic anhydride, by direct analysis of the product (no extractions) utilizing 2-methoxynaphthalene as a standard and a 5 ft × 0.125 in. 15% Carbowax column at 200°, the yield of N-2-pyridylacetamide was found to be 8.5% and those of N-2-pyridylbenzamide and the 2-(cyanophenyl)pyridines were shown to be less than 1% each.

2-(o-Methoxyphenyl)pyridine (7) and 2-(p-Methoxyphenyl)-pyridine (9).—These compounds were prepared according to the method of Gilman and Edwards.[10] The picrate of the liquid ortho isomer had mp 155–156° (lit.[10] mp 152–155°; lit.[14] mp 155–156°). The para isomer had mp 47–49° (lit.[10] mp 47–50°; lit.[14] mp 50–51°).

2-(p-Cyanophenyl)pyridine.—It has been shown[12] that aryl diazonium ions react with pyridine to give arylation very predominantly in the α position. To a solution of 25 g (0.21 mol) of p-aminobenzonitrile (Eastman) in 75 ml of concentrated hydrochloric acid and 700 ml of water was added dropwise a solution of 15 g (0.22 mol) of sodium nitrite in 75 ml of water at 5–10°. The diazonium solution was then added dropwise over a period of 1 hr to 250 ml of pyridine and the mixture was stirred for 24

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(14) J. W. Haworth, I. M. Heilbron, and D. H. Hey, *J. Chem. Soc.*, 358 (1940).

hr at room temperature. The solution was made basic with concentrated ammonium hydroxide and extracted with chloroform (five 100-ml portions). Chloroform and pyridine were removed from the dried extract by evaporation and the resulting solid was dissolved in 150 ml of hot ethanol and treated with 45 g of picric acid in 300 ml of hot ethanol. A crude picrate (20 g, 25%) was obtained by cooling the ethanolic solution in Dry Ice and recrystallizing the precipitate once from acetone. Several additional recrystallizations yielded 7.0 g of picrate, mp 172–173°. The free base was liberated by treatment of this picrate with 10% ammonium hydroxide and was recrystallized from acetone–petroleum ether yielding 2.2 g of pure 2-(*p*-cyanophenyl)-pyridine: mp 98–99° (lit.[15] mp 97–98°); nmr τ 1.25 (q, one 2-pyridyl proton), 2.65–3.50 (m, 7 remaining aromatic protons).

***m*-Aminobenzonitrile.**—A solution of 26 g (0.68 mol) of sodium borohydride in 350 ml of water was added to a slurry of 200 mg of 10% palladium on charcoal in 100 ml of water.[16] A solution of 46 g (0.31 mol) of *m*-nitrobenzonitrile in 800 ml of methanol was added dropwise with ice cooling over a period of 50 min while nitrogen was passed through the mixture. Caution must be exercised in the above procedure as the order of addition is critical and an explosion could ensue if the order were reversed. The reaction mixture was stirred for an additional 15 min, filtered, acidified with 10% hydrochloric acid, made basic with 10% ammonium hydroxide, and extracted with ether (nine 100-ml portions). The dried ether extract was evaporated and the residue recrystallized from ether yielding 24.2 g (67%) of *m*-aminobenzonitrile, mp 53–54° (lit.[17] mp 52°).

2-(*m*-Cyanophenyl)pyridine.—To a solution of 24 g (0.20 mol) of *m*-aminobenzonitrile in 700 ml of water and 75 ml of concentrated hydrochloric acid was added dropwise a solution of 14 g (0.20 mol) of sodium nitrite at 5°. The diazonium solution was then added to 250 ml of pyridine and the resulting solution was stirred for 24 hr at room temperature. The solution was made basic with concentrated ammonium hydroxide and extracted with chloroform (five 100-ml portions). Chloroform and pyridine were removed from the extract by evaporation.

The residue was dissolved in hot ethanol and treated with 46 g of picric acid in hot ethanol. The resulting picrate was crystallized from the ethanol with Dry Ice cooling and the crude picrate was recrystallized several times from acetone, treated with Norite, recrystallized from ether, and mixed with 10% aqueous sodium hydroxide; the resulting solid was filtered and recrystallized from pentane–carbon tetrachloride to yield 0.028 g of product which contained approximately 70% (glpc) of a compound which had a retention time and mass spectrum which were identical with those of the compound assigned the structure 2-(*m*-cyanophenyl)pyridine. Since *m*-aminobenzonitrile was used in this preparation, the cyano group in the product is surely at the meta position. Since this arylation reaction results in very predominantly 2-substituted pyridine[12] and since the pyridine–acetic anhydride reaction produced this material along with 2-*p*-cyanophenylpyridine, the likelihood is extremely high that the *m*-cyanophenyl group is also substituted into the pyridine nucleus at the 2 position of the latter.

***N*-2-Pyridylbenzamide (10, R = Phenyl).**—To a solution of 9.5 g (0.100 mol) of 2-aminopyridine (Aldrich) in 20 ml of dry pyridine was added dropwise with ice cooling 14.2 g (0.100 mol) of benzoyl chloride. The reaction mixture was stirred for 90 min at room temperature, 100 ml of water was added, and the aqueous solution was extracted with chloroform (one 50-ml and two 20-ml portions). The dried chloroform extract was evaporated and the resulting oil was crystallized from benzene–hexane, yielding 13.9 g (70%) of *N*-2-pyridylbenzamide, mp 81–82° (lit.[18] mp 82–83°). The product also exhibited an infrared spectrum identical with that reported (Sadtler No. 3632).

***N*-2-Pyridylacetamide (10, R = Methyl).**—To a solution of 9.8 g (0.104 mol) of 2-aminopyridine in 20 ml of pyridine was added dropwise with ice cooling 8.3 g (0.110 mol) of acetyl chloride. The reaction mixture was stirred for an additional 90 min at room temperature, 100 ml of water was added, and the reaction mixture was worked up as in the previous experiment to yield 8.2 g (60%) of *N*-2-pyridylacetamide, mp 68.0–68.5° (lit.[19] mp 71°; lit.[20] mp 66–67°). The product exhibited an infrared spectrum identical with that reported (Sadtler No. 19556).

Conversion of *N*-2-Pyridylbenzamide to *N*-2-Pyridylacetamide.—A solution of 2 g of *N*-2-pyridylbenzamide, 6 g of acetic anhydride, and 6 g of acetic acid in 50 ml of benzonitrile was heated for 6 hr at a bath temperature of 160°. The benzamide was shown by glpc to be completely converted to *N*-2-pyridylacetamide. The product identification was based upon glpc comparison with an authentic sample.

Registry No.—**1**, 694-59-7; **2**, 3847-19-6; **7**, 5957-89-1; **8**, 4373-58-4; **9**, 5957-90-4; **10** (R = Ph), 4589-12-2; **11**, 4350-51-0; **12**, 32111-34-5; acetic anhydride, 108-24-7; anisole, 100-66-3; benzonitrile, 100-47-0; *N*-(2′-pyridyl)-2-pyridone, 3480-65-7.

Acknowledgments.—We wish to thank the National Institutes of Health for providing the LKB 9000 combined gas chromatograph–mass spectrometer (Grant RR 00273) and Dr. C. C. Sweeley and Mr. John Naworal for recording the mass spectra.

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