

Reissert Compound Studies. LXVII [1]. The Pyrido[3,4-*b*]pyrazine Reissert Compound

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The pyrido[3,4-*b*]pyrazine systems investigated were found to form Reissert compounds in the presence of an acyl halide (or chloroformate) and trimethylsilyl cyanide. Only the mono Reissert compounds were isolated. In the case of 2,3-diphenylpyrido[3,4-*b*]pyrazine (**7**), its reaction with benzene sulfonyl chloride and trimethylsilyl cyanide gave 5-cyano-2,3-diphenylpyrido[3,4-*b*]pyrazine (**16**). Alkylation of the Reissert compound **11** and analog **12** either directly gave the alkylated heterocyclic base or the alkylated Reissert analog compound.

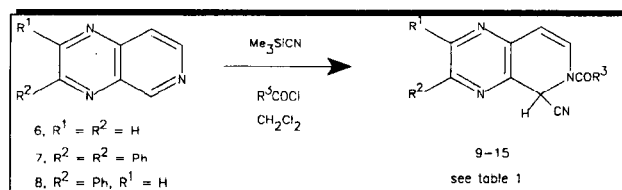
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Veeraraghavan and Popp [2] reported the first example of Reissert compounds [3] from the 5-azaquinoxaline system. Treatment of 5-azaquinoxalines **1** and **2** with trimethylsilyl cyanide, benzoyl chloride, and a catalytic amount of aluminum chloride afforded the mono-Reissert compounds **3** and **4**. Alkylation of **3** with methyl iodide using sodium hydride in *N,N*-dimethylformamide gave the dialkylated Reissert analog **5**.

In view of the success in obtaining Reissert compounds from **1** and **2** the study of Reissert compound formation from triaza systems has been extended to include the pyrido[3,4-*b*]pyrazine system (6-azaquinoxalines, **6**, **7**, **8**). The 6-azaquinoxalines were prepared by condensing the respective diketo compound with 3,4-diaminopyridine.

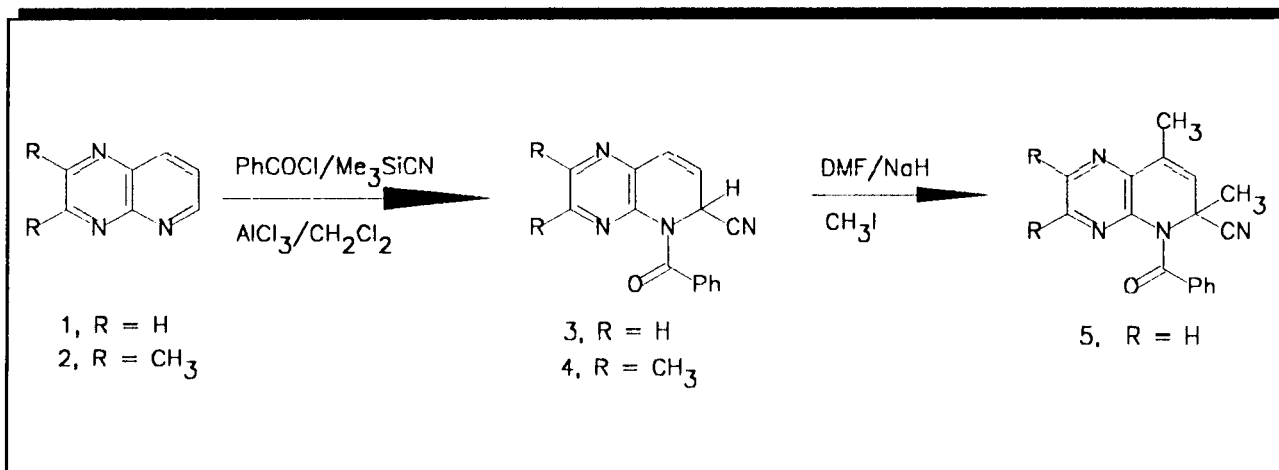
The mono-Reissert compounds and Reissert analogs from **6**, **7**, **8** were prepared by reaction of an equivalent amount each of the heterocyclic base, trimethylsilyl cyanide, acyl halide, or chloroformate, and a catalytic amount of aluminum chloride in methylene chloride for 24 hours. Isolation of the mono-Reissert compounds gave yields ranging from 35-85%. The mono-Reissert compound was the only Reissert compound isolated with reaction occur-

ring across the 5,6 bond of 6-azaquinoxaline. The ir and ¹H nmr spectra were consistent with proposed structures.



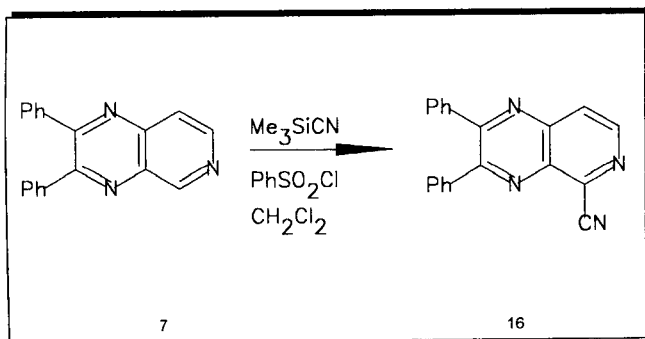
The yields of Reissert compound analogs are summarized in Table 1.

Compound	R ¹	R ²	R ³	% yield
9	H	H	C ₆ H ₅	67
10	H	H	OC ₂ H ₅	58
11	C ₆ H ₅	C ₆ H ₅	C ₆ H ₅	61
12	C ₆ H ₅	C ₆ H ₅	OC ₂ H ₅	78
13	C ₆ H ₅	C ₆ H ₅	<i>o</i> -CH ₂ ClC ₆ H ₄	64
14	C ₆ H ₅	H	C ₆ H ₅	59
15	C ₆ H ₅	H	OC ₂ H ₅	35

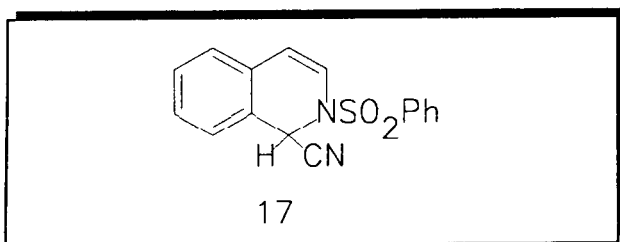


An aspect inherent to the 6-azaquinoxaline system is that virtually all of the synthesized compounds, both Reissert compounds, and their analogs, along with products of the reactions of the compounds, proved difficult to purify and required extensive purification by repeated recrystallization and column chromatography.

Treatment of **7** with benzene sulfonyl chloride, trimethylsilyl cyanide, and a catalytic amount of aluminum chlo-



ride gave **16** in 50% yield. This differs from the reaction of isoquinoline which gave **17**, but is analogous to the behavior of quinoline which gave quinaldonitrile [4].



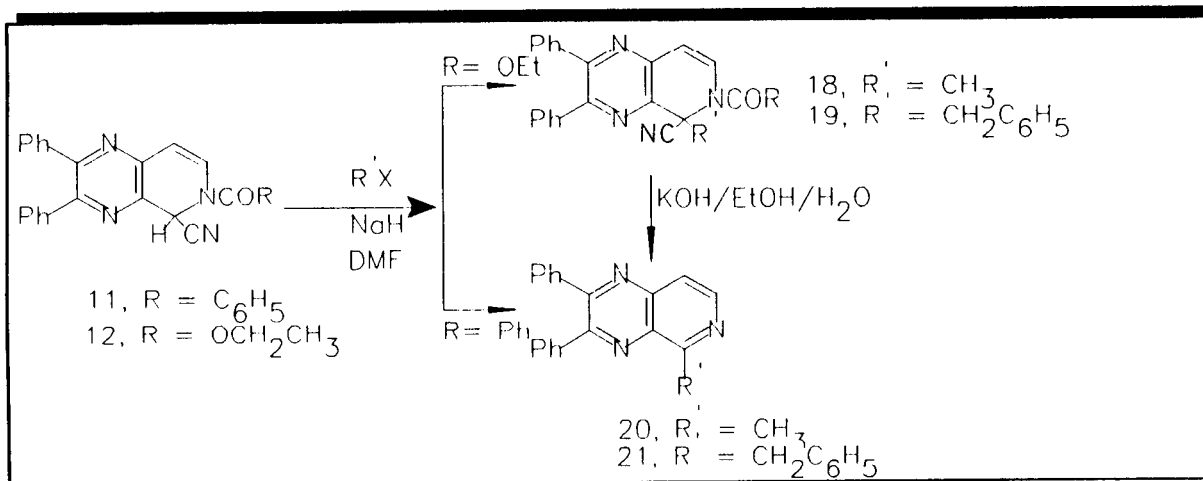
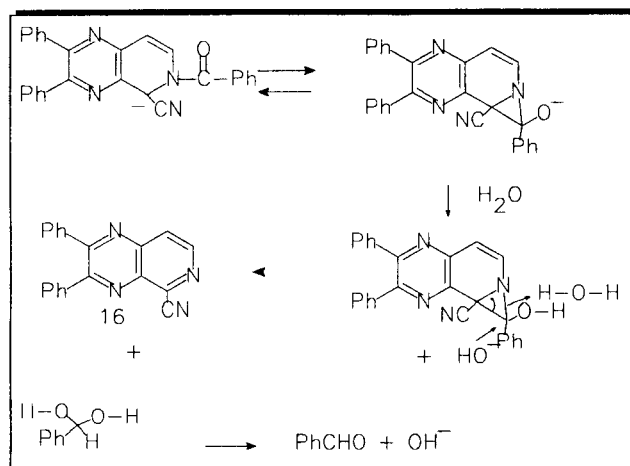
The alkylation reaction of **12** ($R = \text{OCH}_2\text{CH}_3$) with sodium hydride in the presence of iodomethane or benzyl bromide gave the expected alkylated Reissert analog **18** and **19**. Subsequent hydrolysis with ethanolic aqueous potassium hydroxide gave as expected the 5-substituted-6-azaquinoxaline **20**, **21**. However, alkylation of **11** utilizing the same reaction conditions did not give the expected

alkylated Reissert compounds, but directly gave **20** and **21**. Presumably the alkylated Reissert compound underwent hydrolyses during the reaction or more likely the isolation procedure.

Reissert compounds undergo acid hydrolysis to give an aldehyde along with other products. The 6-azaquinoxaline Reissert compounds **11** and **14** failed to give the expected 2,4-dinitrophenylhydrazone of benzaldehyde. Using either concentrated or an aqueous hydrochloric acid (50%) solution [5] gave in both cases a black uncharacterized polymer-like material.

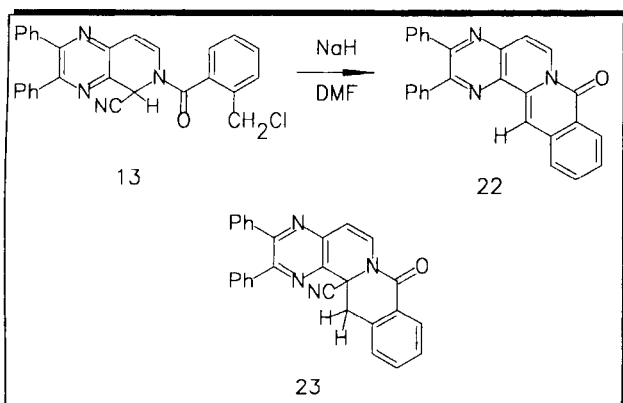
A *N,N*-dimethylformamide mixture of **11** and sodium hydride gave the nitrile **16** rather than the expected rearranged product, 5-benzoyl-6-substituted 5-azaquinoxaline. Air oxidation of the generated anion has been attributed to the generation of compounds with the cyano group alpha to the nitrogen. However, the reaction was thoroughly purged with nitrogen and maintained in a nitrogen atmosphere through out the reaction period suggesting an alternated reaction pathway could be involved in the generation of **16** from **11** and is shown in Scheme 1.

Scheme 1



Repeated attempts to condense (**11**) with benzaldehyde proved unsuccessful. The reaction of **11**, sodium hydride, and benzaldehyde, either at ambient (*ca* 25°) or at 0°, gave an inseparable reaction mixture.

Attempted ring annellation of **13** yielded inconclusive results. Reactions of **13** with sodium hydride gave a material which according to thin layer chromatography analysis consisted of a major and a minor components. Repeated column chromatography attempts failed to separate the isolated products resulting in the major component having a trace amount of an unidentified white material. The IR spectra of the mixture gave an intense absorption at 1681 cm^{-1} attributed to a carbonyl stretch. The ^1H nmr spectra of the mixture exhibited the following results. A doublet at 8.8 ppm, singlet at 8.2 ppm, multiplet at 7.2-7.8 ppm, and a doublet at 6.9 ppm. This would be consistent with a structure of **23** since having a structure of **22** would exhibit signals for the methylene hydrogens at a completing different chemical shift. Furthermore, the methylene hydrogens being attached adjacent to a chiral center and therefore diastereomers, should exhibit a doublet of doublet ^1H nmr signal. It is quite possible that the isolation of **23** is the result of spontaneous loss of hydrogen cyanide from **22**. The mass spectra exhibited a signal at m/z 399.13566 (100%) which is consistent with the molecular weight of **23**, also a metastable ion at m/z 199.56774 was observed.



EXPERIMENTAL

All melting points were determined on a Thomas-Hoover capillary melting point apparatus and are uncorrected. Infrared spectra (IR) were recorded on a Perkin-Elmer model 71-B spectrometer. Proton magnetic resonance spectra (^1H nmr) were determined on a Hitachi Perkin Elmer 12-R-24B or Varian EM390 instrument using deuteriochloroform and tetramethylsilane as the reference compound. Microanalysis was performed by Spang Microanalytical Laboratories, Eagle Harbor, Michigan or by Galbraith Laboratories, Inc., Knoxville, Tennessee. High resolution mass spectra were obtained from the Midwest Center for Mass Spectrometry at the University of Nebraska-Lincoln (NSF, Biology Division, Grant No. DIR9017262). Starting materials were commercially available and used without further purifica-

tion, unless otherwise noted. Silica gel (60-200 mesh from Aldrich) was used for all column chromatographic separations. Thin layer chromatographic (tlc) comparisons were determined on Eastman-Kodak silica gel chromatograms with fluorescent indicator (No-131181).

Pyrido[3,4-*b*]pyrazine (**6**).

To 100 ml of absolute ethanol was added 6.65 g (0.46 mole) of glyoxal (40% in water) and 5.00 g (0.46 mole) of 3,4-diaminopyridine. The mixture was refluxed for 2 hours. On cooling 5.36 g (89%) of **6** was obtained, mp 99-100° sublimed, lit 100-101° [6].

2,3-Diphenylpyrido[3,4-*b*]pyrazine (**7**).

Benzil (6.30 g, 0.03 mole) and 3,4-diaminopyridine (3.27 g, 0.03 mole) were refluxed in absolute ethanol (100 ml) for 3 hours. On cooling 7.30 g (86%) of **7** was obtained, mp 177-178°, lit 177° [7].

2-Phenylpyrido[3,4-*b*]pyrazine (**8**).

Phenylglyoxal hydrate was prepared by refluxing 25 ml (0.21 mole) of acetophenone and 11.1 g (0.1 mole) of selenium dioxide for 2 hours. On cooling, the resulting red color mixture was transferred to a continuous extraction apparatus and extracted with water for 24 hours. The volume of collected water was reduced and on cooling the water mixture 2.19 g phenyl glyoxal hydrate was obtained, mp 82-85°; melting points for phenyl glyoxal hydrate have reported ranges of 73-91°. The degree of dryness of the sample is responsible for this melting point range [8]. Phenylglyoxal hydrate (1.0 g, 6.51 mmoles) and 3,4-diaminopyridine (0.71 g, 6.51 mmoles) were refluxed in benzene (Dean-Stark trap) for 4 hours. Evaporation of the benzene gave 0.84 g (62%) of **8**, mp 124-125° (methanol), lit 125-126° [9].

Preparation of Reissert Analogs (**9-15**).

General Procedure.

To a methylene chloride solution containing the heterocyclic base (**6**, **7**, or **8**), trimethylsilyl cyanide, and a catalytic amount of aluminum chloride was slowly added a quantity of the acid halide over a half an hour time period followed by stirring for 24 hours. The mixture with washed (1 x 50 ml) 10% sodium hydroxide (saturated sodium bicarbonate for **9**, 5% sodium hydroxide for **11** and **12**), with brine (1 x 50 ml), and dried over anhydrous magnesium sulfate. Compounds **9**, **11**, and **12** were also washed with 10% hydrochloric acid (1 x 50 ml) prior to drying. Removal of the methylene chloride *in vacuo* gave the Reissert compound.

6-Benzoyl-5-cyano-5,6-dihydro[3,4-*b*]pyrazine (**9**).

Using the procedure described above, 0.90 g (6.9 mmoles) of (**6**), 930 μl (6.9 mmoles) of trimethylsilyl cyanide, and 810 μl (6.9 mmoles) benzoyl chloride gave (**9**) in 67% yield, mp 168-169° dec (ethyl acetate); ir: 1666, 1621 cm^{-1} ; ^1H nmr: δ = 6.05 (d, 1H), 6.51 (s, 1H), 6.90 (d, 1H), 7.45 (s, 5H), 8.35 (dd, 2H).

Anal. Calcd. for $\text{C}_{15}\text{H}_{10}\text{N}_4\text{O}$: C, 68.70; H, 3.84; N, 21.36. Found: C, 68.14; H, 3.58; N, 21.34.

6-Ethoxycarbonyl-5-cyano-5,6-dihydro[3,4-*b*]pyrazine (**10**).

Using the procedure described above, 0.50 g (3.8 mmoles) of **6**, 519 μl (3.9 mmoles) of trimethylsilyl cyanide, and 372 μl (3.9 mmoles) of ethyl chloroformate gave **10** in 78% yield. The compound was eluted through silica gel with methylene chloride, followed by recrystallization from 95% ethanol, mp 127-128°; ir: 1721, 1621 cm^{-1} ; ^1H nmr: δ = 1.40 (t, 3H), 4.40 (q, 2H), 6.20 (d, 1H, J = 8 Hz), 6.55 (s, 1H), 7.15-7.65 (m, 11H).

Anal. Calcd. for $C_{23}H_{18}O_2N_4$: C, 72.23; H, 4.76; N, 14.64. Found: C, 72.21; H, 4.77; N, 14.70.

2,3-Diphenyl-6-benzoyl-5-cyano-5,6-dihydropyrido[3,4-*b*]pyrazine (11).

Using the procedure described above, 0.50 g (1.8 mmoles) of **7**, 240 μ l (1.8 mmoles) of trimethylsilyl cyanide, and 209 μ l (1.8 mmoles) of benzoyl chloride gave **11** in 61% yield, mp 170-172° (ethyl acetate or acetone/95% ethanol); ir: 1661, 1601 cm^{-1} ; 1H nmr: δ = 6.15 (d, 1H), 6.65 (s, 1H), 6.90 (s, 1H), 7.35 (br, 15H).

Anal. Calcd. for $C_{27}H_{18}N_4O$: C, 78.24; H, 4.04; N, 13.52. Found: C, 78.13; H, 4.04; N, 13.49.

2,3-Diphenyl-6-ethoxycarbonyl-5-cyano-5,6-dihydropyrido[3,4-*b*]pyrazine (12).

Using the procedure described above, 20.0 g (7.1 mmoles) of **7**, 1412 μ l (10.6 mmoles) of trimethylsilyl cyanide, and 1012 μ l (10.6 mmoles) of ethyl chloroformate gave **12** (78% yield) which was further purified by column chromatography (silica gel, methylene chloride), mp 127-128° (95% ethanol); ir: 1721, 1621 cm^{-1} ; 1H nmr: δ = 1.40 (t, 3H), 4.40 (q, 2H), 6.20 (d, 1H), 6.55 (s, 1H), 7.15-7.65 (m, 11H).

Anal. Calcd. for $C_{23}H_{18}O_2N_4$: C, 72.21; H, 4.77; N, 14.70. Found: C, 72.21; H, 4.77; N, 14.70.

2,3-Diphenyl-6-(*o*-chloromethyl)-benzoyl-5-cyano-dihydropyrido[3,4-*b*]pyrazine (13).

Using the procedure described above, 1.0 g (3.5 mmoles) of **7**, 707 μ l (5.3 mmoles) of trimethylsilyl cyanide, and 744 μ l (5.3 mmoles) of 2-chloromethylbenzoyl chloride gave **13** in 64% yield, mp 196-197°; ir: 1671, 1611 cm^{-1} ; 1H nmr: δ = 4.70 (dd, 2H), 6.20 (d, 1H), 6.90 (m, 2H), 7.15-7.60 (bs, 14H).

Anal. Calcd. for $C_{28}H_{19}ClN_4O$: C, 72.65; H, 4.14; N, 12.10. Found: C, 72.42; H, 4.25; N, 11.99.

2-Phenyl-6-benzoyl-5-cyano-5,6-dihydropyrido[3,4-*b*]pyrazine (14).

Using the procedure described above, 0.50 g (2.4 mmoles) of **8**, 333 μ l (2.5 mmoles) of trimethylsilyl cyanide and 290 μ l (2.5 mmoles) of benzoyl chloride gave **14** in 59% yield, mp 185-186° (ethyl acetate); ir: 1662, 1626 cm^{-1} ; 1H nmr: δ = 6.15 (d, 1H), 6.61 (s, 1H), 6.95 (d, 1H), 7.3-7.10 (bs, 8H), 7.8-8.0 (m, 2H), 8.7 (s, 1H).

Anal. Calcd. for $C_{21}H_{14}N_4O$: C, 74.52; H, 4.18; N, 16.56. Found: C, 74.45; H, 4.09; N, 16.62.

2-Phenyl-6-ethoxycarbonyl-5-cyano-5,6-dihydropyrido[3,4-*b*]pyrazine (15).

Using the procedure described above, 0.50 g (2.4 mmoles) of **8**, 333 μ l (2.5 mmoles) of trimethylsilyl cyanide, and 239 μ l (2.5 mmoles) of ethyl chloroformate gave **15** in 35% yield, mp 141-142° (ethyl acetate); ir: 1736, 1716, 1626 cm^{-1} ; 1H nmr: δ = 1.47 (t, 3H), 4.52 (q, 2H), 6.24 (s, 1H), 6.58 (s, 1H), 7.12 (d, 1H), 7.55-7.76 (m, 3H), 8.05 (m, 2H), 8.88 (s, 1H).

Anal. Calcd. for $C_{17}H_{14}O_2N_4$: C, 66.65; H, 4.62; N, 18.28. Found: C, 66.80; H, 4.6; N, 18.3.

2,3-Diphenyl-5-cyanopyrido[3,4-*b*]pyrazine (16).

To a methylene chloride solution containing 0.50 g (1.76 mmoles) of **7**, 236 μ l (1.77 mmoles) of trimethylsilyl cyanide, and 0.02 g (0.15 mmoles) of aluminum chloride, 225 μ l (1.76 mmoles) benzenesulfonyl chloride was dripped in over a half an hour. The reaction mixture was stirred for 48 hours. The mixture was washed with water (2 x 50 ml), saturated sodium bicarbonate (2 x

50 ml), brine (1 x 50 ml), followed by drying over anhydrous magnesium sulfate. Evaporation of the solvent under reduced pressure gave 0.63 g of a red syrup like material which was subjected to column chromatography (silica gel, methylene chloride) yielding 0.27 g (50%) of **16**, mp 153-154° (95% ethanol); ir: 1576, 1523, 1446, 1380 cm^{-1} ; 1H nmr: δ = 8.77 (d, 1H), 8.05 (d, 1H), 7.71-7.03 (m, 10H).

Anal. Calcd. for $C_{20}H_{12}N_4$: C, 77.91; H, 3.82; N, 18.17. Found: C, 77.82; H, 3.88; N, 18.14.

2,2-Diphenyl-5-benzylpyrido[3,4-*b*]pyrazine (21) from 11.

To a *N,N*-dimethylformamide solution containing 0.25 g (0.60 mmole) of **11** and 71.4 μ l (0.60 mmole) of benzylbromide was added 0.03 g (0.62 mmole) of sodium hydride. The reaction was mixed for 4 hours under nitrogen atmosphere. The reaction was quenched by pouring the mixture onto 50 g of ice. Following melting of the ice 0.21 g of a pink color solid was recovered and recrystallized to give 0.14 g (53%) of **21**. The material was recrystallized 6 times before an analytically pure sample could be obtained with a melting point of 175-176° (95% ethanol); ir: 1581, 1526, 1480, 1446 cm^{-1} ; 1H nmr: δ = 8.65 (d, 1H), 7.75 (d, 1H), 7.65-7.10 (m, 15H), 4.80 (s, 2H).

Anal. Calcd. for $C_{26}H_{19}N_3$: C, 83.62; H, 5.13; N, 11.25. Found: C, 83.49; H, 5.15; N, 11.21.

2,3-Diphenyl-5-methylpyrido[3,4-*b*]pyrazine (20).

Using the procedure described above, 0.50 g (1.21 mmoles) of **11**, 113 μ l (1.81 mmoles) of iodomethane, and 0.12 g (2.5 mmoles) of sodium hydride gave 0.32 g (89%) of **20**, mp 146-148° (silica gel, methylene chloride); ir: 1586, 1560, 1538 cm^{-1} ; 1H nmr: δ = 8.60 (d, 1H), 7.75 (d, 1H), 7.60-7.30 (m, 10H), 3.10 (s, 3H).

Anal. Calcd. for $C_{20}H_{15}N_3$: C, 80.78; H, 5.10. Found: C, 80.80; H, 5.07.

2,3-Diphenyl-6-carboethoxy-5-cyano-5-benzyl-5,6-dihydropyrido[3,4-*b*]pyrazine (19).

Using the procedure described above, but with a 2 hour reaction time, 0.30 g (0.78 mmole) of **12**, 92.8 μ l (0.78 mmole) of benzylbromide, and 0.04 g (0.83 mmole) of sodium hydride gave 0.10 g (27%) of **19**, mp 134-135° (95% ethanol); ir: 1726, 1621 cm^{-1} ; 1H nmr: δ = 7.5-7.0 (m, 16H), 5.5 (d, 1H), 4.6 (q, 2H), 4.0 (dd, 2H), 1.5 (t, 3H).

Anal. Calcd. for $C_{30}H_{24}N_4O$: C, 76.25; H, 5.12. Found: C, 76.16; H, 5.04.

Hydrolysis of 0.03 g (0.06 mmole) of **19** in aqueous ethanolic potassium hydroxide gave **21** in 42% (10 mg) yield which was identical in all respects to authentic **(21)** prepared from **11**.

Preparation of 20 via 18 and 12.

Using the procedure described above, with a 7 hour reaction time, 0.50 g (1.31 mmoles) of **12**, 122 μ l (1.96 mmoles) of iodomethane, and 0.12 g (2.50 mmoles) of sodium hydride gave 0.24 g (46%) of **18**, mp 197-198° (silica gel, methylene chloride); ir: 1721 cm^{-1} ; 1H nmr: δ = 7.5-7.0 (m, 11H), 5.9 (d, 1H), 4.4 (q, 2H), 2.2 (s, 3H), 1.4 (t, 3H).

Ethanolic potassium hydroxide hydrolysis (1 hour reflux) of 30 mg (0.076 mmole) of **18** gave 20 mg (89%) of **20** identical in all respects to authentic **20** prepared from **11**.

Acid Hydrolysis of 11.

Concentrated hydrochloric acid (5 ml) was added to 0.25 g (0.60 mmole) of **11** and 0.12 (0.60 mmole) 2,4-dinitrophenylhydra-

zine and the mixture was heated for 30 minutes on a steam bath. The reaction mixture initially emitted bubbles and became red in color, which changed to black after about 10 minutes of heating. The reaction mixture was left covered at room temperature for 1 week. The reaction remained black in color with a tarry polymer-like material suspended in the solution through out the 1 week time period. No benzaldehyde derivative (reddish-orange color powder) was recovered.

The reaction was repeated using 5 ml of concentrated hydrochloric acid and 5 ml of water which yield the same results.

Acid Hydrolysis of **14**.

Using the procedure described above, 0.06 g (0.30 mmole) of **14** and 0.10 g (0.30 mmole) of 2,4-dinitrophenylhydrazine gave the same results.

Reaction of **11** with Sodium Hydride.

To a *N,N*-dimethylformamide solution containing 0.25 g (0.63 mmole) of **11** was added 0.05 g (1.04 mmoles) of sodium hydride. The mixture was stirred for 19 hours with a nitrogen atmosphere. The mixture was poured onto 50 g of ice which yield a precipitate on melting of the ice. Filtration followed by recrystallization gave 0.10 g (53%) of **16**, identical in all respects to authentic **16** prepared from **7**, benzenesulfonyl chloride and trimethylsilyl cyanide.

Attempts to Condense (**11**) with Benzaldehyde.

Maintaining an nitrogen atmosphere, a *N,N*-dimethylformamide solution of 0.15 g (0.36 mmole) of **11**, 61 μ l (0.40 mmole) of benzaldehyde, and 0.02 g (0.42 mmole) of sodium hydride were stirred for 5 hours. After such time, the reaction mixture was poured onto 50 g of ice, which resulted in recovering a waxy-beige color material. Thin layer chromatography of the isolated material showed that the isolated material consisted of at least 3 components which proved to be inseparable. The reaction was repeated twice and once at 0°, yielding the same inseparable mix-

ture.

Ring-annulation of (**13**) to give (**22**).

To a *N,N*-dimethylformamide solution containing 0.09 g (0.19 mmole) of **13** was slowly added 0.01 g (0.21 mmole) of sodium hydride. The mixture was stirred for 5 hours after which time the mixture was poured onto 50 g of ice. The filtered material was subjected to column chromatography yielding 0.04 g of **22**. Thin layer chromatography analysis showed that the recovered material was contaminated with an unidentified material. Repeated column chromatography failed to give analytically pure sample. The isolated product, with contaminant, had a mp of 288-290° (silica gel, 1% acetone in methylene chloride); ir: 1681, 1631, 1611 cm^{-1} ; ^1H nmr: δ = 8.8 (d, 1H), 8.2 (s, 1H), 7.8-7.2 (m, 14H), 6.9 (d, 1H); ms: *m/z* (%) 399.13566 (M^+ , 100), 398.12714 (18.72), 397.12298 (1.12), 369.11879 (3.12), 199.56774 (M^+ , 5.21), 193.05177 (3.03), 103.04187 (2.07).

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