Pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinoline [1] and the Alkylation of Pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinolin-5(6*H*)-one

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The reaction of 2-aminopyridine, *o*-phthaldehydic acid and potassium cyanide gave pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinolin-5(6*H*)-one, which upon treatment with propargylbromide, yielded both *O* and *N* alkylated products. 2-Aminopyridine, *o*-phthaldehyde and potassium cyanide gave 1-cyano-2-(2-pyridyl)isoindole which rearranged in acid to give the previously unreported parent pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinole. Structures were confirmed using uv, ir, nmr and x-ray spectroscopy.

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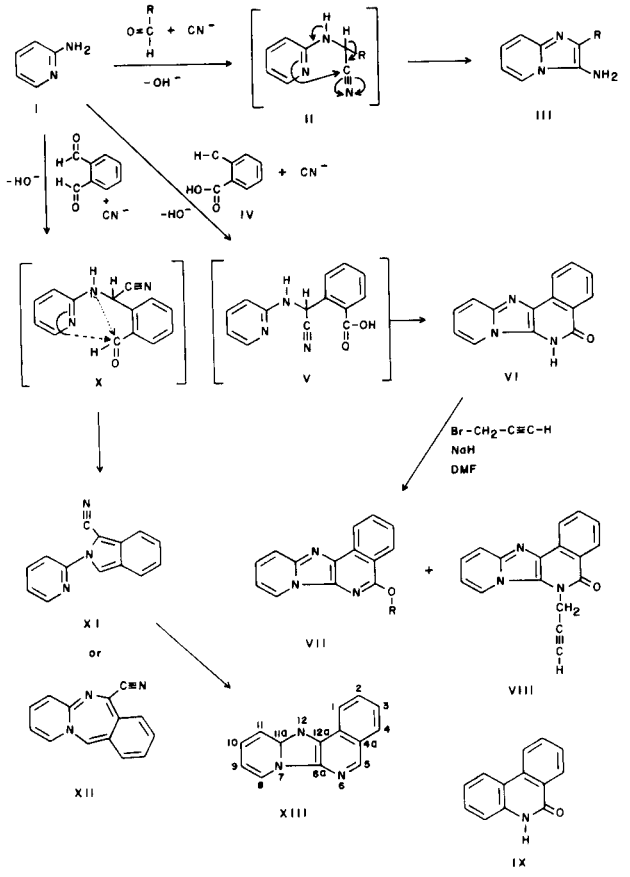
The reaction of 2-aminopyridine (I), an aldehyde, and cyanide ion gives a 3-aminoimidazo[1,2-*a*]pyridine (III), probably *via* the aminoacetonitrile intermediate II [3]. When this Strecker-type reaction was tried using phthaldehydic acid (IV) as the aldehyde, the amine acid V was not obtained, but a second ring closure had occurred to give pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinolin-5(6*H*)-one (VI). Lee, Hashimoto, Shudo and Nagao recently reported the preparation of this compound (VI) from 2-aminopyridine, cyanide and *o*-cyanobenzaldehyde [2].

Scheme I



The lactam carbonyl and the N-H stretches of VI were seen at 6.10 and 3.70 (ir) respectively. The presence of an acidic proton (3.70 μ) and a basic nitrogen at the 12-position account for the amphoteric character of the lactam VI which is soluble in both aqueous sodium carbonate and dilute hydrochloric acid (*p*H 2).

The similarity between the lactam sites of pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinlin-5(6*H*)-one (VI) and phenanthridone (IX) is apparent. Alkylation of phenanthridone with propargyl bromide [2a] and other alkyl halides [2a-c] in a sodium hydride-*N*,*N*-dimethylformamide mixture is reported to give only *N*-substituted materials. When lactam VI was treated with propargyl bromide under these conditions an 84-16 mixture of *O* and *N* alkylated products VII and VIII was obtained, the *O* alkylated product predominating.

It is not clear whether the difference in results is due to geometric and/or electronic effects or whether those investigators working with phenanthridone simply didn't look for *O*-alkylated materials. Models, however, appear to show that the nitrogen of phenanthridone (IX) is somewhat more accessible than that of lactam VI.

The mixture of alkylated material VII and VIII was separated by a combination of chromatography and crystallization. The *N*-alkylated material was identified by its ir spectrum (C=O at 6.10 μ). The methylene protons of the *O* and *N* alkyl compounds VII and VIII appeared at δ 5.34 and 5.53 (nmr) respectively, permitting quantitation of the crude mixture.

The attempted preparation of the parent pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinoline system XII from 2-aminopyridine (I), *o*-phthalodicarboxaldehyde and potassium cy-

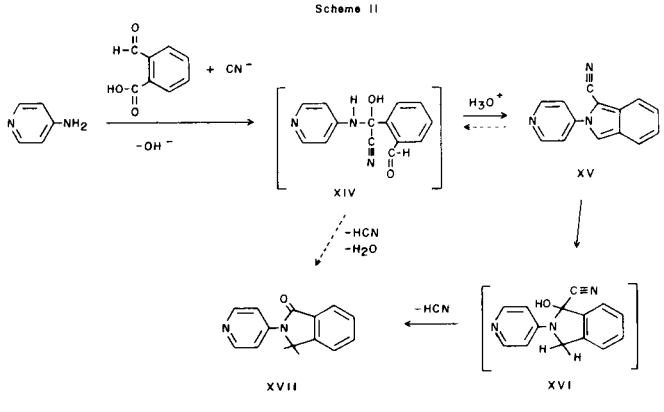
anide resulted instead in the formation of an isomeric nitrile (m/e = 219, C≡N at 5.05 μ) the structure of which was deduced as follows.

Phthalodicarboxaldehyde, 1-butylamine and propanethiol (a nucleophile) are reported to give 1-propylthio-2-*t*-butylisonidole [4] which would suggest that the nitrile is the isoindole XI. 2-Aminopyridine (I), however, has two nitrogen atoms at which annellation could occur so that the diazepine XII is also a potential product. If the assumed intermediate X is indeed formed, then by analogy with 2-aminopyridine and benzaldehyde, reaction should take place at the exocyclic nitrogen [7]. It could also be argued that the diazepine XII, having 16 π electrons is a 4n π system and is thus less likely to form than the isoindole XI [8].

Of the spectral data we had immediately available (uv, ir, nmr and ms), uv was the most helpful. When 1-cyano-2-(4-pyridyl)isoindole (XVII) was prepared from 4-aminopyridine, *o*-phthalodicarboxaldehyde and cyanide (Scheme II), the uv spectra of both this nitrile XVII and that obtained from 2-aminopyridine were virtually identical above 230 nm. This indicated that the nitrile obtained with 2-aminopyridine (Scheme I) was 1-cyano-2-(2-pyridyl)isoindole (XI), which was proven unequivocally by means of X-ray crystallography (Figure 1).

1-Cyano-2-(2-pyridyl)isoindole (XI) rearranged rapidly, and in good yield (81%) to pyrido[2′,1′:2,3]imidazo[4,5-*c*]-isoquinoline (XIII) [12] (Scheme I). The rather complex uv spectrum of this compound was virtually superimposable upon that of the *O*-propargyl compound VII. This structure was also confirmed by means of X-ray crystallography (*vide infra*).

Scheme II



The pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinoline system (XIII) is stable and, while it appears to have considerable hydrocarbon character, the central portion of the molecule has 3-nitrogen atoms in a cluster of six atoms which apparently gives the system some hydrophilicity. After standing several years in a bottle, our sample which was originally anhydrous, was found to be a hemi-hydrate (by X-ray and elemental analysis) with the water protons each being

Figure 1a

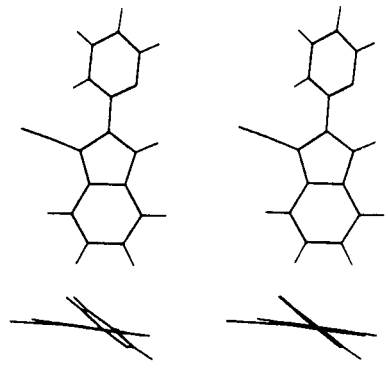
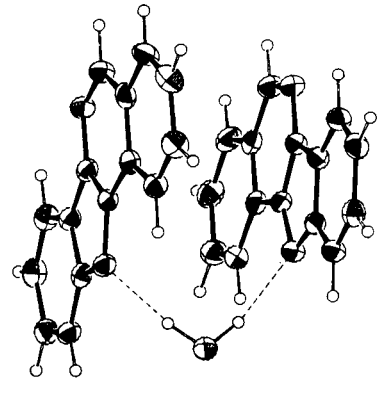


Figure 1b



hydrogen bonded to the nitrogen at the 12-position of the parent molecule (see Figure 2).

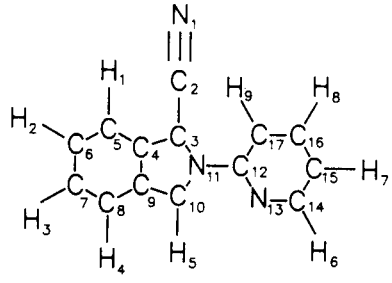
1-Cyano-2-(4-pyridyl)isoindole (XV) is more stable in acid than its 2-pyridyl counterpart XI (which goes to XIII), but on prolonged treatment loses the nitrile group to give 2-(4-pyridyl)isoindol-1(3*H*)-one (XVII).

There are two possible paths to this compound (XVII) as shown in Scheme II: 1) 2-(4-pyridyl)-1-cyanoisoindole (XV) could, like its 2-pyridyl counterpart XI undergo ring opening in acid, lose HCN then cyclize to XVII, or 2) XV could (with or without reversible ring opening and cyclization) hydrate then lose HCN to form XVII.

The X-ray crystallographic data for 1-cyano-2-(2-pyridyl)isoindole (XI), Figures la and lb, show that the planes of the pyridine ring and the isoindole ring form an angle of 30.9°. In this disposition the β-pyridyl proton forces the nitrile group 5.16° out of the plane of the isoindole ring (Figure 1b). Atoms 2, 3, 4 and 9 form a dihedral angle of −174.84° [13]. Pyrido[2′,1′:2,3]imidazo[4,5-*c*]isoquinoline (XIII) (Figure 2) is planar. The sample analyzed was a hemihydrate and it can be seen that the water protons are hydrogen bonded to the nitrogens at the 12-positions of two molecules that lie in different planes.

The bond lengths and bond angles for compounds XI and XIII are shown in Tables I and II.

Table I



Bond Distances in 1-Cyano-2-(2-pyridyl)isoindole (XI)

N(1)	C(2)	1.1558(21)	C(5)	C(6)	1.3656(25)
N(11)	C(3)	1.3946(21)	C(6)	C(7)	1.4234(26)
N(11)	C(10)	1.3662(21)	C(7)	C(8)	1.3615(26)
N(11)	C(12)	1.4325(21)	C(8)	C(9)	1.4207(24)
N(13)	C(12)	1.3333(22)	C(9)	C(10)	1.3902(25)
N(13)	C(14)	1.3489(23)	C(12)	C(17)	1.3761(24)
C(2)	C(3)	1.4132(25)	C(14)	C(15)	1.3809(26)
C(3)	C(4)	1.4001(23)	C(15)	C(16)	1.3852(26)
C(4)	C(5)	1.4169(24)	C(16)	C(17)	1.3801(25)
C(4)	C(9)	1.4318(23)			
C(5)	H(1)	.979(18)	C(14)	H(6)	.988(19)
C(6)	H(2)	.954(18)	C(15)	H(7)	.987(19)
C(7)	H(3)	.976(18)	C(16)	H(8)	1.000(19)
C(8)	H(4)	.978(19)	C(17)	H(9)	.906(20)
C(10)	H(5)	.973(18)			

Bond Angles

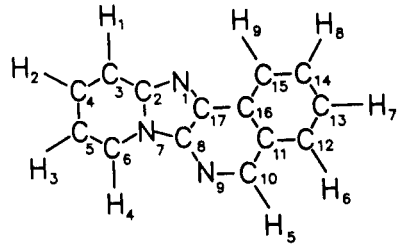
C(3)	N(11)	C(10)	109.47(14)
C(3)	N(11)	C(12)	127.25(13)
C(10)	N(11)	C(12)	123.22(14)
C(12)	N(13)	C(14)	116.34(15)
N(1)	C(2)	C(3)	176.94(17)
N(11)	C(3)	C(2)	123.98(14)
N(11)	C(3)	C(4)	107.52(13)
C(2)	C(3)	C(4)	128.30(15)
C(3)	C(4)	C(5)	132.58(15)
C(3)	C(4)	C(9)	107.07(14)
C(5)	C(4)	C(9)	120.36(15)
C(4)	C(5)	C(6)	118.14(17)
C(5)	C(6)	C(7)	121.59(17)
C(6)	C(7)	C(8)	121.74(17)
C(7)	C(8)	C(9)	118.32(17)
C(4)	C(9)	C(8)	119.86(15)
C(4)	C(9)	C(10)	107.22(14)
C(8)	C(9)	C(10)	132.93(16)
N(11)	C(10)	C(9)	108.73(15)
N(11)	C(12)	N(13)	114.48(14)
N(11)	C(12)	C(17)	121.07(15)
N(13)	C(12)	C(17)	124.44(16)
N(13)	C(14)	C(15)	123.71(17)
C(14)	C(15)	C(16)	118.03(17)
C(15)	C(16)	C(17)	119.37(17)
C(12)	C(17)	C(16)	118.08(17)
C(4)	C(5)	H(1)	119.8(10)
C(6)	C(5)	H(1)	122.0(10)
C(5)	C(6)	H(2)	120.0(10)
C(7)	C(6)	H(2)	118.4(10)
C(6)	C(7)	H(3)	118.7(10)

Table I (continued)

Bond Angles for 1-Cyano-2-(2-pyridyl)isoindole

C(8)	C(7)	H(3)	119.6(10)
C(7)	C(8)	H(4)	122.1(10)
C(9)	C(8)	H(4)	119.6(10)
N(11)	C(10)	H(5)	119.7(10)
C(9)	C(10)	H(5)	131.6(10)
N(13)	C(14)	H(6)	115.2(10)
C(15)	C(14)	H(6)	121.1(10)
C(14)	C(15)	H(7)	120.5(11)
C(16)	C(15)	H(7)	121.5(11)
C(15)	C(16)	H(8)	120.7(10)
C(17)	C(16)	H(8)	119.9(10)
C(12)	C(17)	H(9)	121.7(12)
C(16)	C(17)	H(9)	120.1(12)

Table II



Bond Distances in Pyrido[2',1':2,3]imidazo[4,5-*c*]isoquinoline (XIII)

N(1)	C(2)	1.338(3)	C(5)	C(6)	1.345(4)
N(1)	C(17)	1.374(3)	C(8)	C(17)	1.374(3)
N(7)	C(2)	1.392(3)	C(10)	C(11)	1.430(4)
N(7)	C(6)	1.377(3)	C(11)	C(12)	1.423(4)
N(7)	C(8)	1.398(3)	C(11)	C(16)	1.417.(4)
N(9)	C(8)	1.345(3)	C(12)	C(13)	1.367(4)
N(9)	C(10)	1.325(3)	C(13)	C(14)	1.399(4)
C(2)	C(3)	1.403(4)	C(14)	C(15)	1.362(4)
C(3)	C(4)	1.359(4)	C(15)	C(16)	1.405(4)
C(4)	C(5)	1.424(4)	C(16)	C(17)	1.424(4)
O(18)	H(10)	.93(3)	C(10)	H(5)	.987(24)
C(3)	H(1)	.948(26)	C(12)	H(6)	.920(27)
C(4)	H(2)	.954(25)	C(13)	H(7)	.977(27)
C(5)	H(3)	.998(25)	C(14)	H(8)	1.003(27)
C(6)	H(4)	.96(3)	C(15)	H(9)	.990(27)

Bond Angles

C(2)	N(1)	C(17)	104.41(19)
C(2)	N(7)	C(6)	122.44(21)
C(2)	N(7)	C(8)	105.95(19)
C(6)	N(7)	C(8)	131.61(22)
C(8)	N(9)	C(10)	112.72(22)
N(1)	C(2)	N(7)	112.08(20)
N(1)	C(2)	C(3)	129.52(23)
N(7)	C(2)	C(3)	118.40(22)
C(2)	C(3)	C(4)	119.44(25)
C(3)	C(4)	C(5)	120.39(26)
C(4)	C(5)	C(6)	120.69(26)
N(7)	C(6)	C(5)	118.65(25)
N(7)	C(8)	N(9)	125.46(22)
N(7)	C(8)	C(17)	105.31(21)

Table II (continued)

Bond Angles for Pyrido[2',1':2,3]imidazo[4,5-*c*]isoquinoline

N(9)	C(8)	C(17)	129.22(24)
N(9)	C(10)	C(11)	125.08(24)
C(10)	C(11)	C(12)	121.67(24)
C(10)	C(11)	C(16)	119.99(23)
C(12)	C(11)	C(16)	118.34(23)
C(11)	C(12)	C(13)	120.09(26)
C(12)	C(13)	C(14)	120.75(27)
C(13)	C(14)	C(15)	120.74(26)
C(14)	C(15)	C(16)	120.08(25)
C(11)	C(16)	C(15)	119.97(23)
C(11)	C(16)	C(17)	114.82(21)
C(15)	C(16)	C(17)	125.21(23)
N(1)	C(17)	C(8)	112.25(22)
N(1)	C(17)	C(16)	129.62(22)
C(8)	C(17)	C(16)	118.13(22)
H(10)	O(18)	H(10)	105.0(4)
C(2)	C(3)	H(1)	117.3(15)
C(4)	C(3)	H(1)	123.2(15)
C(3)	C(4)	H(2)	119.8(14)
C(5)	C(4)	H(2)	119.8(14)
C(4)	C(5)	H(3)	120.8(14)
C(6)	C(5)	H(3)	118.5(13)
N(7)	C(6)	H(4)	114.4(16)
C(5)	C(6)	H(4)	127.0(16)
N(9)	C(10)	H(5)	117.0(14)
C(11)	C(10)	H(5)	117.9(14)
C(11)	C(12)	H(6)	119.7(16)
C(13)	C(12)	H(6)	120.2(16)
C(12)	C(13)	H(7)	119.3(15)
C(14)	C(13)	H(7)	119.9(15)
C(13)	C(14)	H(8)	119.3(14)
C(15)	C(14)	H(8)	120.0(14)
C(14)	C(15)	H(9)	122.2(15)
C(16)	C(15)	H(9)	117.7(15)

EXPERIMENTAL

All melting points are uncorrected. Spectra were recorded on the following instruments: uv - Carey (MeOH Methanol solutions), ir - Perkin Elmer Infracord (nujol mulls), nmr - Varian A-60 (deuteriochloroform solutions), ms - Finnigan 3000.

Pyrido[2',1':2,3]imidazo[4,5-*c*]isoquinolin-5-(6*H*)-one (**VI**).

To 8.0 g (0.2 mole) of sodium hydroxide in 500 ml of water was added 30.2 g (0.2 mole) of phthaldehydic acid and when solution occurred 20.8 g (0.2 mole) of sodium bisulfite was added. This mixture was heated on a steam bath for 30 minutes then 18.8 g (0.2 mole) of 2-aminopyridine was added and the mixture heated for one hour. Potassium cyanide (20.0 g, 0.031 mole) was added and the mixture heated for an additional 4 hours. The mixture was cooled then acidified with acetic acid (**CAUTION: HCN**). Ice was added during acidification, to keep the mixture cool. The resulting yellow solid was filtered off, washed with water and dried to give 17.5 g (35%) of VI which crystallized from dimethylformamide-toluene, mp >310°; ir (nujol): μ 3.70 (N-H), 6.10 (C=O); uv (methanol): nm (log ϵ) 223 (4.35), 233 (4.29), 235 (4.28), 241 (4.28), 248 s (4.31), 256 (4.39), 310 (3.60), 360 s (3.93), 374 (4.02); nmr (DMSO-d_6): δ 6.8-8.2 (complex array, 7H), 8.65 (complex doublet, J = 7 Hz, 8-H, 1H), DMSO-d_6 contained trace of water.

Anal. Calcd. for $C_{14}H_9N_3O$: C, 71.48; H, 3.86; N, 17.86. Found: C, 71.50; H, 4.03; N, 17.73.

Alkylation of Pyrido[2',1':2,3]imidazo[4,5-*c*]isoquinolin-5-(6*H*)-one (**VI**).

To 4.7 g (0.2 mole) of VI in 50 ml of *N,N*-dimethylformamide was added 0.96 g (0.02 mole) of 50% sodium hydride suspension in mineral oil. This mixture was heated under reflux for 1 hour and then cooled and 2.38 g (0.02 mole) of propargyl bromide was added dropwise. This mixture was then heated under reflux for 2 hours. The solvent was removed by evaporation *in vacuo*. The resulting residue was triturated with water and the resulting solid filtered off and weighed 3.8 g. The crude material in chloroform was put onto a column of alumina (3 x 20 cm) and eluted with chloroform. Two fractions were collected. The first of these was colorless. The solvent was removed from the effluent to give 1.48 g of VII which crystallized from benzene, mp 222-223°; ir (nujol): μ 4.80 (C≡C); uv (methanol): nm (log ϵ) 233 s (5.00), 240 (5.05), 253 (5.10), 258 (5.09), 270 s (4.54), 295 (4.31), 307 (4.38), 330 (4.34), 345 (4.52), 362 (4.67), 381 (4.59); nmr (deuteriochloroform): δ 2.5 (triplet, J = 2.5 Hz, acetylenic H), 5.34 (doublet, J = 2.5 Hz, methylene 2H), 6.9-7.9 (complex array, 7H), 8.7 (complex doublet, J = 7 Hz, 8-H, 1H).

Anal. Calcd. for $C_{17}H_{11}N_3O$: C, 74.71; H, 4.06; N, 15.38. Found: C, 74.70; H, 3.96; N, 15.28.

The second fraction, yellow in color, contained 1.86 g of material shown by nmr to be a 5:2 mixture of VII and VIII. Compound VII was obtained by fractional crystallization from heptane, mp 203-210°; ir (nujol): μ 4.80 (C≡C), 6.10 (C=O); uv (methanol): nm (log ϵ) 223 (4.35), 233 (4.29), 235 (4.28), 241 (4.28), 256 (4.39), 310 (3.60), 360 (3.93), 374 (4.02); nmr (deuteriochloroform): δ 2.6 (triplet, acetylenic proton, 1H), 5.54 (doublet, methylene, 2H), 7.3-8.6 (complex array, aromatic, 7H), 9.0 (complex doublet, 8-H, 1H).

Anal. Calcd. for $C_{17}H_{11}N_3O$: C, 74.71; H, 4.06; N, 15.38. Found: C, 74.62; H, 3.93; N, 15.30.

1-Cyano-2-(3-pyridyl)isoindole (**XI**).

To 20.8 g (0.2 mole) of sodium bisulfite in 500 ml of water was added 26.8 g (0.2 mole) of phthalodicarboxaldehyde. The mixture was stirred until solution effectively occurred and the mixture was then filtered through celite to remove trace insoluble impurities. To this was added 18.8 g (0.2 mole) of 2-aminopyridine and the mixture was heated on a steam bath until it turned cloudy (about 30 minutes) at which time 45 g (0.69 mole) of potassium cyanide in 100 ml of water was added and the mixture heated for additional 90 minutes. The mixture was cooled and extracted with chloroform. The chloroform extract was dried over potassium carbonate and percolated through a column of florisil (3 x 13 cm). The column was eluted with 200 ml of chloroform and the solvent removed from the effluent to give 22.5 g (51%) of XI. The material was treated with charcoal in boiling methanol then filtered and on cooling white needles formed, mp 115-116°; ir (nujol): μ 5.05 (C≡N); uv (methanol): nm (log ϵ) 236 (4.46), 246 (4.39), 295 (3.93), 305 (3.98), 345 (3.87); nmr (deuteriochloroform): δ 6.89-7.80 (complex array, aromatic 6H) 7.89 (unsymmetrical doublet, J = 0.5 Hz, H-3, 1H), 8.47 (complex unsymmerical doublet, H-6, 1H).

Anal. Calcd. for $C_{14}H_9N_3$: C, 76.70; H, 4.14; N, 19.16. Found: C, 76.52; H, 4.21; N, 18.92.

Pyrido[2',1':2,3]imidazo[4,5-*c*]isoquinole (**XIII**).

A mixture of 1-cyano-2-(2-pyridyl)isoindole (XI) (7.0 g, 0.032 mole) and concentrated hydrochloric acid (50 ml) was heated on a steam bath for 15 minutes. Water (25 ml) was added and the mixture heated for an additional 5 minutes. The mixture was cooled and neutralized with sodium bicarbonate. A solid formed which was filtered off to give 5.7 g (82%) of XIII, mp 162-164°; uv (methanol): nm (log ϵ) 243 s (4.81), 251 (4.86), 269 s (4.48), 292 (4.15), 305 (4.16), 330 s (4.15), 343 (4.28), 357 (4.38), 363 (4.67); nmr (deuteriochloroform): δ (α-pyridyl, 2H, J = 6 Hz); ms: m/e = 219.

Anal. Calcd. for $C_{14}H_9N_3$: C, 76.70; H, 4.14; N, 19.17. Found: C, 76.41; H, 4.34; N, 19.11. Hemihydrate Calcd. for $C_{14}H_9N_3 \cdot \frac{1}{2} H_2O$: C, 73.67; H, 4.35; N, 18.31. Found: C, 73.51; H, 4.35; N, 18.31.

2-(4-Pyridyl)isoindol-2(3*H*)-one (**XVI**).

To 15 ml of 6*N* hydrochloric acid as added 0.5 g of 1-cyano-2-(4-pyridyl)isoindole (XV). The mixture was heated at 80° for 16 hours then concentrated *in vacuo*. The residue was taken up in water and

made mildly alkaline with sodium bicarbonate. The resulting buff colored solid was extracted into chloroform. The chloroform solution was dried over magnesium sulfate and filtered. Toluene was added to the filtrate and the chloroform removed *in vacuo*. White crystals formed which were filtered off and weighed 0.4 g (83%) of XVI, mp 195-197°; ir (potassium bromide): μ 5.89 (C=O): uv (methanol): nm 229 (3.96), 237 (3.90), 279 (4.31); nmr (deuteriochloroform): δ 4.75 (singlet, methylene 2H), 7.4-7.85 (complex array, aromatic 6H), 8.48 (unsymmetrical complex doublet, α-pyridyl, 2H, J = 6 Hz).

Anal. Calcd. for $C_{13}H_{10}N_2O$: C, 74.26; H, 4.79; N, 13.32. Found: C, 74.17; H, 4.83; N, 13.36.

1-Cyano-2-(4-pyridyl)isoindole (**XV**).

A mixture containing 13.4 g (0.1 mole) of *o*-phthalodicarboxaldehyde and 10.4 g (0.1 mole) of sodium bisulfite in 125 ml of water was stirred until solution effectively occurred. The trace insoluble material was filtered off. To this was added 9.4 g (0.1 mole) of 4-aminopyridine. The mixture was heated with stirring in a water bath, at 70° for 45 minutes after which 20 g (0.3 mole) of potassium cyanide in 50 ml of water was added. The mixture was heated with stirring. After 20 minutes, 200 ml of toluene were added and the mixture heated with stirring for an additional hour. The toluene layer was separated. The aqueous layer was extracted with toluene and the toluene fractions combined and dried over magnesium sulfate. The toluene solution was treated with charcoal, then concentrated *in vacuo*. The residue was triturated with ether and filtered to give 6.5 g (30%) of gray solid. This was taken up in chloroform and percolated through a column of florisil. The solvent was removed from the effluent to give title compound as a white solid which crystallized from methanol, mp 132-135°; ir (nujol): μ 4.55 (C≡N); uv (methanol): nm 238 (4.48), 250 (4.45), 297 (3.85), 308 (3.88), 347 (3.84); nmr (deuteriochloroform): δ 7.6 (unsymmetrical doublet, 3-H, J = 1 Hz, in the midst of complex array at 7-7.7 δ (7H total); 8.72 δ (complex doublet, 1H).

Anal. Calcd. for $C_{14}H_9N_3$: C, 76.70; H, 4.14; N, 19.17. Found: C, 76.41; H, 4.34; N, 19.11.

Crystallography

1-Cyano-2-(2-pyridyl)isoindole (**XI**).

The data were collected using a locally interfaced Picker four-circle goniostat equipped with a graphite monochromator. All data were collected at low temperatures using a gaseous nitrogen cooling system. Details of the diffractometer and cooling system have been described previously [9]. The structure was solved by direct methods and Fourier techniques and refined by full-matrix least squares [10]. All hydrogen atoms were located and refined. Bond distances and bond angles are shown in Table I and other crystallographic details are available [11].

Pyrido[2',1':2,3]imidazo[4,5-*c*]isoquinoline (**XIII**).

The diffractometer and cooling system were the same as for XI, and direct methods were also used. The compound crystallizes in the tetragonal space group $I4_1/a$ (#88). Two molecules related by a crystallographic two-fold axis are joined by a hydrogen bonded water molecule which lies on the axis, as shown in Figure 3. Bond distances and bond angles are shown in Table II and further crystallographic details are available [11].

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[8] Indeed, simple Huckel calculations using parameters suggested either by Streitweisser and Paudler, [a] or by Pullman [b] place two electrons in a low antibonding orbital E. 14 and .1 respectively. Parameter adjustment could do little more than move these electrons to a nonbonding level; [a] A. Streitweisser Jr., "Molecular Orbital Theory for Organic Chemists", John Wiley and Sons, New York 1961, p 135; [b] B. Pullman and A. Pullman, "Quantum Biochemistry", Interscience Publishers, New York 1963, p 108. The program used was that of Tominaga, *Kagaku No Ryoiki*, **36**, 719 (1982).

[9] J. C. Huffman, L. N. Lewis and K. G. Caulton, *Inorg. Chem.*, **19** 2755 (1980).

[10] Programs used included code from A. C. Larson's Los Alamos Library, J. A. Ibers' Northwestern Library, as well as MULTAN78 and ORTEP.

[11] Complete crystallographic details are available in microfiche form from the Chemistry Library, Indiana University, Bloomington, Indiana 47405. Request MSC Report 83705 for XI and 83704 for XIII.

[12] This is the name suggested for this ring system by Dr. Kurt Loening of Chemical Abstracts Service - Private Communication.

[13] These calculations were carried out using standard algorithms with a program call XRAY by J. P. Paolini - to be published.