Synthetic Applications of N–N Linked Heterocycles. Part 9.[1] Preparation of α-(4-Pyridyl)nitroalkanes and *N*-(4-Pyridyl)azoles by Regiospecific Attack of Nitroalkyl and Azolyl Anions on *N*-(2,6-Dimethyl-4-oxopyridin-1-yl)pyridinium Salts[2]

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Sodium salts of nitroalkanes and of azoles react with *N*-(2,6-dimethyl-4-oxopyridin-1-yl)pyridinium salts (1)—(3) to give regiospecifically 1,4-dihydropyridines (12)—(14), (15), and (16) in high yields. Photolysis of the dihydropyridines gives α-(4-pyridyl)nitroalkanes (7)—(9) and *N*-(4-pyridyl)azoles in moderate to good yields. Certain azolyl dihydropyridines revert to the parent azole on photolysis; this appears to be a steric effect.

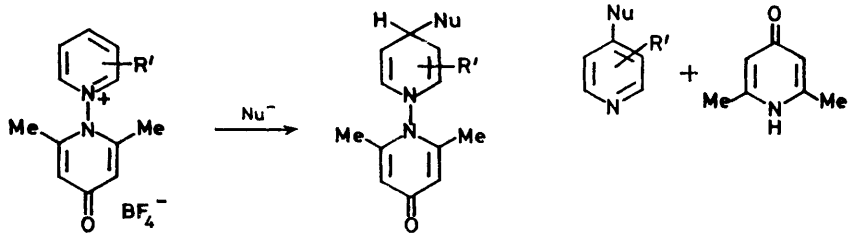
We have demonstrated in earlier papers the synthetic utility of the *N*-(2,6-dimethyl-4-oxopyridin-1-yl)pyridinium salts (1)—(3) in the regiospecific synthesis of 4-substituted pyridines.[1-3] The methyl groups in the oxopyridinyl moiety of these salts sterically shield the α-positions of the second ring. Thus an attacking nucleophile is directed regiospecifically into the position γ to the quaternary nitrogen atom, giving a 1,4-dihydro-intermediate (4) which in turn may be fragmented to a 4-substituted pyridine (5) and the pyridone (6) (Scheme). We now report, as a further development of the method, the general preparation in good overall yields of α-(4-pyridyl)nitroalkanes (7)—(9) and *N*-(4-pyridyl)azoles (10) and (11).

α-(4-Pyridyl)nitroalkanes have been prepared in 50—70% yields by nitration of alkylpyridines under basic [4] or acidic [5] conditions, but the methods presume availability of the alkylpyridine precursors. Anions derived from nitroalkanes have been added to a number of quaternised heteroaromatic molecules,[6] and are known to attack the 4-position of certain pyridinium salts,[6a,b,7,8] though with *N*-methoxypyridinium salts attack occurs at the 2-position and is followed by ring opening.[9] In no case, however, has an α-(4-pyridyl)nitroalkane been prepared by this approach.

N-(4-Pyridyl)azoles have been prepared by a variety of methods, but all require 4-substituted pyridines as starting materials, and suffer from many stages, lack of generality, and/or low yields.[10]

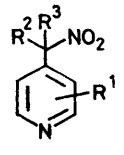
RESULTS AND DISCUSSION

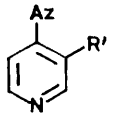
α-(4-*Pyridyl*)*nitroalkanes*.—Treatment of a mixture of the appropriate nitroalkane and a pyridinium salt (1)—(3) in absolute ethanol with ethanolic sodium ethoxide



(1) R′ = H
(2) R′ = 2-Me
(3) R′ = 3-Me

(4) (5) (6)





(7) R^1 = H
(8) R^1 = 2-Me
(9) R^1 = 3-Me

(10) R′ = H
(11) R′ = Me

Az = azolo

SCHEME

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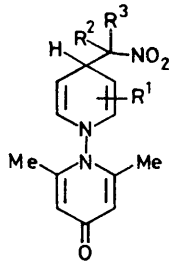
TABLE 1

Preparative, physical, and analytical data for 1,4-dihydro-intermediates (12)—(14) derived from nitroalkane anions

Compd.	Substituents R¹	R²	R³	Yield (%)	M.p.[a] (°C)	Crystal form	Solvent[b]	Found (%) C	H	N	Molecular formula	Required (%) C	H	N
(12a)[c]	H	H	H	79	156—157	Plates	I	59.9	5.9	16.1	$C_{13}H_{15}N_3O_3$	59.8	5.8	16.1
(12b)[d]	H	H	Me	90	140—141	Plates	I	61.2	6.4	15.5	$C_{14}H_{17}N_3O_3$	61.1	6.2	15.3
(12c)[e]	H	Me	Me	92	152—153	Plates	I	61.9	6.4	14.2	$C_{15}H_{19}N_3O_3$	62.3	6.6	14.5
(12d)	H	Me	$PhCH_2$	71	145—148	Plates	II	67.3	6.2	11.1	$C_{21}H_{23}N_3O_3$, $0.5H_2O$	67.4	6.5	11.2
(12e)	H	Me	$2,4\text{-}Cl_2C_6H_3CH_2$	72	135—139	Plates	I	57.6	5.0	9.1	$C_{21}H_{21}Cl_2N_3O_3$	58.1	4.9	9.7
(13c)	2-Me	Me	Me	52	140—142	Micro-crystals	I	61.4	7.1		$C_{16}H_{21}N_3O_3$, $0.5H_2O$	61.5	7.1	
(14c)	3-Me	Me	Me	80	138—140	Needles	II	62.8	6.8	13.4	$C_{16}H_{21}N_3O_3$	63.3	7.0	13.9

[a] All compounds melt with decomposition. [b] I = $CHCl_3$–light petroleum; II = $CHCl_3$–n-hexane. [c] *m/e* 261 ($M^{+\cdot}$). [d] *m/e* 275 ($M^{+\cdot}$). [e] *m/e* 289 ($M^{+\cdot}$).

at room temperature, gave after evaporation of the solvent and purification of the residue, the 1,4-dihydro-intermediates (12)—(14) in an average yield of 75%. The highest yields were obtained by use of 4 mol equiv.



(12) R^1=H
(13) R^1= 2-Me
(14) R^1= 3-Me

of nitroalkane, though yields in excess of 70% could be obtained with 1.5 mol equiv. Purification was carried out by dissolving the intermediate in chloroform and reprecipitating by slow addition of light petroleum or n-hexane. The compounds were found to contain residual water which could not be completely removed without causing decomposition. Thus, microanalytical data (Table 1) were frequently unsatisfactory, and structures were confirmed by recording ^{1}H n.m.r. and i.r. spectra (Table 2), which were entirely consistent with those for analogous compounds.[1,3]

In contrast to the 1,4-dihydro-intermediates derived from Grignard reagents,[3] compounds (12)—(14) were relatively stable in solution, and resisted fragmentation to the products (7)—(9). No decomposition occurred after 6 h reflux in chloroform, whereas heating in acetonitrile under the same conditions gave polymeric material. Lee[11] observed that a solution of intermediate (12c) in tetrahydrofuran containing peroxide impurities decomposed to an extent of 60% into the pyridine (7f) at room temperature in 16 h, whereas negligible decomposition took place when the solvent was scrupulously purified. This suggested a free-radical mechanism for the fragmentation, and it was subsequently found that photolysis of the intermediates (medium-pressure mercury lamp) in chloroform in the presence of benzoyl peroxide gave the α-(4-pyridyl)nitroalkanes (7)—(9) in good yields. The fragmentation was followed by ^{1}H n.m.r. spectroscopy, the irradiation time for complete decomposition (Table 3) being proportional to the initial amount of intermediate present. The products (average yield 43%) were isolated by chromatography on alumina, and, where oils, were characterised as picrates or picrolonates. Structures were confirmed by ^{1}H n.m.r. and i.r. spectroscopy (Table 4). The decomposition of intermediate (12a) was unsatisfactory, and the yield of product (7b) was low, due

TABLE 2

^{1}H N.m.r. and i.r. spectroscopic data for 1,4-dihydro-intermediates (12)—(14) derived from nitroalkane anions

Compound	^{1}H N.m.r. (δ)[a,b] Pyridone ring 2′,6′	2′,6′	3′,5′	1,4-Dihydropyridine ring 2,6	3,5	4	R^2	R^3	$\nu_{max.}$/cm^{-1} Phase	Nitro-group	Dihydro-pyridine	Pyridone
(12a)	*2.15*	*2.20*	6.10	6.10	4.63	3.82	4.25	4.25	$CHBr_3$	1 540, 1 360	1 680	1 570, 1 640
(12b)	*2.17*	*2.25*	6.20	6.35	4.65	3.70	4.30	1.55	Nujol	1 540, 1 380	1 680	1 570, 1 640
(12c)	*2.19*	*2.23*	6.10	6.10	4.55	3.75	1.55	1.55	Nujol	1 530, 1 360	1 680	1 570, 1 640
(12d)	*2.14*	*2.18*	6.05	6.05	4.5	3.8	1.4	4.5, 7.2	$CHBr_3$	1 530, 1 360	1 680	1 570, 1 640
(12e)	*2.23*	*2.26*	6.15	6.15	4.6	3.9	1.4	3.5, 7.2	$CHBr_3$	1 530, 1 360	1 680	1 560, 1 640
(13c)	*2.15*		6.1	6.10 *2.15*	4.4	3.7	1.55	1.55	Nujol	1 520, 1 370	1 680	1 570, 1 650
(14c)	*2.17*	*2.21*	6.05	6.05	4.55 *1.65*	3.85	1.65	1.55	Nujol	1 530, 1 340	1 680	1 570, 1 640

[a] In $CDCl_3$. [b] Numbers in italics are for ring alkyl substituents.

TABLE 3

Preparative, physical, and analytical data for α-(4-pyridyl) nitroalkanes (7)—(9)

Compound	Substituents R¹	R²	R³	Irradiation time (h)	Yield (%)	M.p. (°C)	Found (%) C	H	N	Molecular formula	Required (%) C	H	N
(7b)	H	H	Me	15	31	118—120 [a,b]				$C_7H_8N_2O_2$			
(7c)	H	Me	Me	8	75	143—145 [a,c]				$C_8H_{10}N_2O_2$			
(7d)	H	Me	$PhCH_2$	28	52	84—85 [d]	69.5	5.8	11.4	$C_{14}H_{14}N_2O_2$	69.4	5.8	11.6
(8c)	2-Me	Me	Me	14	51	*e*	60.6	7.2		$C_9H_{12}N_2O_2$	60.0	6.7	
(9c)	3-Me	Me	Me	16	55	204—205 [f]	51.0	4.5		$C_9H_{12}N_2O_2$, $C_{10}H_8N_4O_5$ [f]	51.3	4.5	

[a] M.p. of picrate (from EtOH); compound is an oil at room temperature. [b] Lit.,[4] 118—120 °C. [c] Lit.,[5] 145—146 °C. [d] Needles from ether–light petroleum (b.p. 40—60 °C). [e] Oil. [f] As picrolonate, yellow needles from EtOH–H_2O.

probably to formation and hydrolysis of the aci-nitro isomer on the column.[4]

TABLE 4

^{1}H N.m.r. and i.r. spectroscopic data for α-(4-pyridyl) nitroalkanes (7)—(9)

Compound	^{1}H N.m.r. (δ) [a] Pyridine ring 2,6	3,5	R²	R³	$\nu_{max.}$/cm^{-1} Phase	Nitro-group
(7b)	8.34	7.0	1.39	5.05	Liquid	1 550, 1 345
(7c)	8.72	7.37	1.98	1.98	Liquid	1 550, 1 350
(7d)	8.61	6.92	2.80	3.55, 7.20	$CHBr_3$	1 540, 1 345
(7e)	8.70	7.25	1.82	3.85, 7.25	Liquid	1 550, 1 345
(8c)	8.55	7.15 [b]	1.95	1.95	Liquid	1 540, 1 345
(9c)	8.55 [c]	7.5	1.96	1.96	$CHBr_3$	1 540, 1 340

[a] In $CDCl_3$. [b] Ring Me at δ 2.6. [c] Ring Me at δ 2.2.

N-(4-*Pyridyl*)*azoles*.—The 1,4-dihydro-intermediates (15) and (16) were prepared by addition of the sodium salt of the appropriate azole in acetonitrile to a pyridinium salt (1)—(3). Despite the dark colour of the reaction mixture, the intermediates could be isolated in high yields (average 79%) after purification. As with intermediates (12)—(14), satisfactory analyses generally could not be obtained, due to the presence of residual water, but structures were confirmed by ^{1}H n.m.r. and i.r. spectroscopy (Table 5). Physical (and where available analytical) data are presented in Table 6. Pyrrole, however, failed to give an intermediate (15) even with the stronger base n-butyl-lithium in tetrahydrofuran.*

Photochemical decomposition of the intermediates, by the method described for compounds (12)—(14), gave fair yields (average 48%) of the *N*-(4-pyridyl)azoles (10) and (11), and these were fully characterised by ^{1}H n.m.r. and i.r. spectroscopy (Table 7) and by physical and microanalytical data (Table 8). The imidazolo-intermediate (15e) failed to give identifiable products, and the intermediates (15c, f, and h) decomposed to give the parent azole in high yield rather than the expected products (10).

* These conditions worked successfully for the addition of ketones to the salts (1)—(3).[1]

TABLE 5

^{1}H N.m.r. and i.r. spectroscopic data for 1,4-dihydro-intermediates (15) and (16) derived from azole anions

Compound	R′	Az	^{1}H N.m.r. (δ) [a,b] Pyridone ring 2′,6′	3′,5′	1,4-Dihydropyridine 2,6	3,5	4	Az	$\nu_{max.}$/cm^{-1} Phase	1,4-Dihydropyridine	Pyridone
(15a)	H	Benzotriazolo	*2.30*	6.25	6.5	5.2	6.05	7.4 7.9	Nujol	1 675	1 575
			2.45								1 640
(15b)	H	Benzimidazolo	*2.35*	6.2	6.5	5.2	6.05	7.4 8.1	Nujol	1 675	1 575
			2.45								1 640
(15c)	H	2-Methylbenzimidazolo	*2.1*	6.1	6.35	4.7	5.9	7.2 7.5	$CHBr_3$	1 680	1 565
			2.25					2.6 (Me)			1 640
(15d)	H	5,6-Dimethylbenzimidazolo	*2.25*	6.15	6.4	5.05	5.9	7.3 7.6	$CHBr_3$	1 675	1 570
			2.35					7.9			1 640
								2.4 (Me)			
(15e)	H	Imidazolo	*2.2*	6.1	6.25	4.95	5.6	7.1 7.7	$CHBr_3$	1 680	1 580
			2.3								1 640
(15f)	H	3,5-Dimethylpyrazolo	*2.2*	6.15	6.3	4.95	5.7	6.15	$CHBr_3$	1 675	1 570
			2.25					2.3 2.4			1 635
(15g)	H	Succinimido	*2.2*	6.2	6.35	4.7	5.7	2.65	$CHBr_3$	1 685 (sh)	1 575 [c]
			2.45								1 645
(15h)	H	Phthalimido	*2.25*	6.2	6.35	4.85	5.8	7.8	$CHBr_3$	1 680	1 570 [c]
			2.55								1 640
(16a)	Me	Benzotriazolo	*2.25*	6.15	6.55	5.2	5.65	7.45 7.85	$CHBr_3$	1 655	1 580
			2.45		6.45	*1.6*					1 640
(16b)	Me	Benzimidazolo	*2.25*	6.2	6.45	5.0	5.9	7.35 8.0	$CHBr_3$	1 680	1 570
			2.45		6.4	*1.6*					1 640
(16d)	Me	5,6-Dimethylbenzimidazolo	*2.25*	6.15	6.4	4.95	5.9	7.3 7.6	$CHBr_3$	1 685	1 565
			2.45		6.3	*1.55*		7.9			1 640
								2.4 (Me)			
(16g)	Me	Succinimido	*2.15*	6.1	6.35	5.1	5.55	2.65	$CHBr_3$	1 685 (sh)	1 570 [c]
			2.30		6.25	*1.6*					1 640

[a] In $CDCl_3$. [b] Numbers in italics are for ring alkyl substituents. [c] $\nu_{max.}$ C=O between 1 695 and 1 715 cm^{-1}.

TABLE 6

Preparative data and physical properties for 1,4-dihydro-intermediates (15) and (16) derived from azole anions

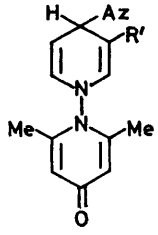
Compound	Yield (%)	M.p. (°C)	Crystal form [a]
(15a)	76	112—114	Prisms
(15b)	72	109—111	Plates
(15c)	71	118—121	Plates
(15d)	82	147—151	Prisms
(15e)	76	108—112	Prisms
(15f) [b]	75	109—112	Needles
(15g) [c]	79	118—121	Needles
(15h) [d]	86	107—110	Plates
(16a)	90	109—111	Needles
(16b)	72	63—64	Needles
(16d)	99	127—130	Plates
(16g)	68	125—128	Prisms

[a] From $CHCl_3$–light petroleum (b.p. 40—60 °C). [b] Found: C, 66.9; H, 6.7; N, 18.1. $C_{17}H_{20}N_4O,0.5H_2O$ requires C, 66.9; H, 6.9; N, 18.4%. [c] *m/e* 299 ($M^{+\cdot}$). [d] M.p. of dihydrate 118—119 °C (Found: C, 63.2; H, 5.5; N, 11.3. $C_{20}H_{17}N_3O_3,2H_2O$ requires C, 62.6; H, 5.5; N, 11.0%); *m/e* 347 ($M^{+\cdot}$).

Reverse reactions of this type have been observed with certain other 1,4-dihydro-intermediates,[1,3] and seem to arise at least partly from steric strain about the bond at the 4-position of the dihydropyridine ring. Thus the 2-methylbenzimidazolo-intermediate (15c) reverses, whereas analogous compounds lacking the 2-methyl-group [(15b), (15d), (16b), and (16d)] gave the desired azolylpyridines.

EXPERIMENTAL

2-Nitro-1-phenylpropane and 1-(2,4-dichlorophenyl)-2-nitropropane [12] and *N*-(2,6-dimethyl-4-oxopyridin-1-yl)-pyridinium salts (1)—(3) were prepared by the published procedure [13] and dried *in vacuo* before use; all other reagents were available commercially. Acetonitrile was dried over P_4O_{10} and redistilled before use. Light petroleum refers to the fraction of b.p. 40—60 °C. I.r. spectra were recorded on a Perkin-Elmer 297 instrument, and ^1H n.m.r. spectra recorded on Perkin-Elmer R12 or Varian HA-100 spectrometers for solutions in $CDCl_3$ with tetramethylsilane as an internal standard. Spectroscopic data are recorded in Tables 2, 4, 5 and 7 and physical and analytical data in Tables 1, 3, 6, and 8.



Az = azolo

(15) R′ = H

(16) R′ = Me

TABLE 7

^1H N.m.r. and i.r. spectroscopic data for *N*-(4-pyridyl)azoles (10) and (11)

Compound	R′	Az	^1H N.m.r. (δ) [a] Pyridine ring 2,6	Pyridine ring 3,5	Azole	$\nu_{max.}/cm^{-1}$ Phase	
(10a)	H	Benzotriazolo	8.8	7.8	7.4 7.8	Nujol	1 590, 1 500, 1 455
(10b)	H	Benzimidazolo	8.85	7.85	705 8.25	Nujol	1 580, 1 500, 1 450
(10d)	H	5,6-Dimethylbenzimidazolo	8.8	7.5	7.3 7.65 8.1 2.4 (Me)	$CHBr_3$	1 580, 1 500, 1 470
(10g)	H	Succinimido	8.75	7.4	2.9	$CHBr_3$	1 715 (br), 1 590, 1 505, 1 480
(11a)	Me	Benzotriazolo	8.75	8.2	7.6	$CHBr_3$	1 585, 1 505, 1 455
			8.8	*2.35*			
(11b)	Me	Benzimidazolo	8.7	7.65	7.35 8.15	$CHBr_3$	1 585, 1 500, 1 480 (sh)
			8.75	*2.3*			
(11d)	Me	5,6-Dimethylbenzimidazolo	8.65	7.3	7.7 7.9 8.3 2.4 (Me)	$CHBr_3$	1 580, 1 505, 1 470
			8.7	*2.2*			
(11g)	Me	Succinimido	8.55	7.15	2.9	$CHBr_3$	1 710 (br), 1 590, 1 510, 1 485
			8.6	*2.15*			

[a] In $CDCl_3$; numbers in italics refer to ring alkyl substituents.

TABLE 8

Preparative, physical, and analytical data for *N*-(4-pyridyl)azoles (10) and (11)

Compound	Yield [a] (%)	M.p. (°C)	Crystal form	Solvent [b]	Found (%) C	H	N	Molecular formula	Required (%) C	H	N
(10a)	51	113—114 [c]	Needles	*d*				$C_{11}H_8N_4$			
(10b)	68	121—122 [e]	Needles	I				$C_{12}H_9N_3$			
(10d)	50	192—194 [f]	Needles	II	52.8	3.7	18.5	$C_{20}H_{16}N_6O_7$	53.1	3.6	18.6
(10g)	22	238—239 [g]	Prisms	III	61.0	4.6	15.6	$C_9H_8N_2O_2$	61.4	4.6	15.9
(11a)	41	84—85	Needles	I	68.3	4.8	26.3	$C_{12}H_{10}N_4$	68.5	4.8	26.7
(11b)	50	114—115	Needles	I	74.2	5.1	20.5	$C_{13}H_{11}N_3$	74.6	5.3	20.1
(11d)	54	212—214 [f]	Needles	II	53.5	3.8	17.6	$C_{21}H_{18}N_6O_7$	54.1	3.9	18.0
(11g)	47	138—140 [f]	Needles	II	45.4	3.0	16.4	$C_{10}H_{10}N_2O_2$	45.8	3.1	16.7

[a] All intermediates (15) and (16) were irradiated for 8 h. [b] I = Ether–n-hexane, II = ethanol, III = methanol. [c] Lit.,[10a] 113.5 °C. [d] Purified by sublimation (125 °C at 7 mmHg). [e] Lit.,[10a] 123 °C. [f] This, and data following, refer to the picrate salt. [g] Lit., 228—229 °C [E. J. Brown and J. Polya, *J. Chem. Soc.* (*C*), 1968, 2904].

General Procedure for the Preparation of 1,4-*Dihydro-intermediates* (12)—(14).—The pyridinium salt (1)—(3) (2.38 g, 0.01 mol) was added to the appropriate nitro-compound (0.04 mol) in absolute EtOH (15 ml). NaOEt in EtOH (10 ml, 1M) was added dropwise with stirring during 10 min at 20 °C, stirring being continued for a further 1—1.5 h. The solvent was removed at 20 °C under reduced pressure, and the residue taken up in $CHCl_3$ (30 ml), filtered, and evaporated. The product was washed with dry ether (15 ml), and recrystallised by dissolving in $CHCl_3$ and reprecipitating by slow addition of light petroleum or n-hexane.

General Procedure for the Preparation of 1,4-*Dihydro-intermediates* (15) *and* (16).— To the pyridinium salts (1) or (3) (2 mmol) stirred under dry MeCN (16 ml) at 20 °C was added dropwise the appropriate azole (2 mmol) in methanolic NaOMe (2 ml; 1M). The salt dissolved slowly to give a red solution, which after 30 min was evaporated under reduced pressure at 20 °C. The resulting red purple solid was purified as described for the intermediates (12)—(14).

Photolysis of the 1,4-*Dihydro-intermediates*.—The intermediate (0.8 mmol) in dry $CHCl_3$ (150 ml) containing freshly recrystallised benzoyl peroxide (0.08 mmol), was irradiated (medium-pressure mercury lamp, 125 W) in a water-cooled Pyrex photochemical reactor under N_2, with exclusion of moisture, for appropriate periods of time (Tables 3 and 8). Removal of the solvent followed by chromatography of the residue on alumina (grade I; neutral) using $CHCl_3$ as the eluant, gave on evaporation the α-(4-pyridyl)nitroalkanes (7)—(9) and the *N*-(4-pyridyl)azoles (10) and (11). Products were recrystallised, or if oils, often converted into derivatives for characterisation.

We thank the S.R.C. for support.

[0/502 *Received*, 2*nd April*, 1980]

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