PHOTOADDITION OF ALCOHOLS TO SOME 1,2,3,6-TETRAHYDROPYRIDINES*

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Received July 8th, 1986

Photochemical addition of methanol to 1,2,3,6-tetrahydropyridine derivatives *Ia*, *Ib*, *Id*—*Ii* and *III* afforded methoxypiperidines *IIa*—*IIg* and *IV* as Markovnikov type addition products. Reduction of 4-methyl-1-(2-propenyl)pyridinium bromide with sodium borohydride yielded 4-methyl-1-(2-propenyl)-1,2,3,6-tetrahydropyridine (*Ig*).

The 4-alkoxy-4-alkyl-substituted piperidine nucleus can be found in certain compounds with neuroleptic[1] or hypoglycaemic[2] activity. So far, 4-alkoxy-4-alkylpiperidines have usually been prepared by series of reactions from 1-benzyl-4-piperidones[1,2]. On the other hand, in the synthesis of Nicergoline the methoxypiperidine grouping in ring D was formed mostly by photochemical addition of methanol to the double bond of lysergic or methyllysergic acid[3-5].

Within the framework of studies of synthesis and properties of 1,2,3,6-tetrahydropyridines we were interested in the photochemical addition of alcohols to 1,4-dialkyl--1,2,3,6-tetrahydropyridines. Photochemical reactions of 1,2,3,6-tetrahydropyridine bases described so far concern isomerization of the double bond[6,7] (moreover conjugated with the side-chain double bond). Irradiation at 185 nm sometimes leads to fission of the tetrahydropyridine nucleus[8].

Photochemical addition of alcohols to olefinic double bonds can proceed in two ways[9]. In the first, the reaction takes place at the alcohol oxygen atom, giving rise to an ether as 1 : 1 addition product and is assumed to involve polar intermediates. Such reactions are *e.g.* sensitized additions of methanol, ethanol, and 2-propanol to substituted 1-alkyl-1-cyclohexenes[10-12] which afford 1-alkoxy-1-alkylcyclohexanes. Also the addition of ethanol to cholesta-3,5-dienes[13] belongs to this mechanistic type. To the second, more frequent type of addition belong those in which the reaction takes place at the hydroxyl-bearing carbon atom, probably *via* radicals. These reactions give higher alcohols as 1 : 1 addition products[9].

We carried out the photoadditions of alcohols to 1,4-dialkyl-1.2,3,6-tetrahydro-

* Part LVII in the series Studies in the Pyridine Series; Part LVI: This Journal *48*, 1435 (1983).

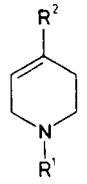
pyridines under conditions similar to those used for the addition to cyclohexene derivatives[10-12]. Since tetrahydropyridines absorb strongly UV light only at short wavelengths, the reactions were performed in the presence of toluene as sensitizer, similarly as described for additions to cyclohexene derivatives. Photoaddition of methanol to 1,4-dimethyl-1,2,3,6-tetrahydropyridine (*Ia*) gave a single product (according to gas–liquid and thin-layer chromatography) identified by the NMR spectrum as 4-methoxy-1,4-dimethylpiperidine (*IIa*). In addition to the product, all the photoreactions afforded non-distillable polymeric residues. Analogously, photoaddition of methanol to the ethyl derivative *Ib* gave 1-ethyl-4-methoxy-4-methylpiperidine (*IIb*). On the other hand, 1-benzyl-4-methyl-1,2,3,6-tetrahydropyridine (*Ic*) under the same conditions lost the benzyl group and the reaction mixture contained 1,2-diphenylethane and a minor amount of 4-methyl-1,2,3,6-tetrahydropyridine (*Id*) in addition to the starting compound.

In order to find an explanation for the absence of an alkoxy product in the reaction mixture, we tried to add methanol to the tetrahydropyridine *Id*. However, this compound reacted much more slowly than the corresponding N-alkyl base *Ia*. After irradiation of *Id* for 25 h, the NMR spectra proved the presence of the methoxy derivative *IIc* along with the starting compound. Attempts to achieve higher yields of *IIc* by prolonged irradiation resulted in extensive polymerization.

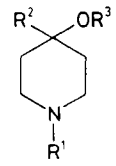
Analogously to the photochemical addition of methanol to *Ia*, addition to 4-(1-propyl)- and 4-(2-propyl)tetrahydropyridine (*Ie* and *If*) afforded the respective methoxy derivatives *IId* and *IIe*. These reactions required longer irradiation, apparently due to the steric effect of the substituents. In the photoaddition of methanol to 4-methyl-1-(2-propenyl)-1,2,3,6-tetrahydropyridine (*Ig*) we identified only the piperidine *IIf* and the starting base which could not be completely separated. Similarly to the photoaddition of methanol to 1-ethyl-1,4-cyclohexadiene[11] which takes place specifically in position 1, the addition to *Ig* occured only at the endocyclic trisubstituted double bond whereas the monosubstituted double bond of the 2-propenyl group was not attacked.

Some 2-(1-methyl-1,2,3,6-tetrahydropyridyl)indoles are known[14,15] to have antihistamine properties. It seemed therefore useful to try photoadditions with these derivatives and combine thus structural features of biologically active alkoxypiperidines and indole derivatives. Strong absorption of compounds *Ih* and *III* at 300 nm enabled non-sensitized additions with a Pyrex filter (cut-off 295 nm). Addition of methanol to *Ih* and *III* gave the methoxy derivatives *IIg* and *IV*, respectively, again according to the Markovnikov rule. In several cases we tried also photoadditions of ethanol to 1,2,3,6-tetrahydropyridine bases. Toluene-sensitized photoaddition of ethanol to *Ia* gave a 4 : 1 mixture of 4-ethoxy-1,4-dimethylpiperidine (*IIh*) and methoxy derivative *IIa* which were separated by preparative GLC. The same mixture was obtained with benzene as sensitizer. We are not aware of any analogous photochemical reaction leading to a methoxy derivative in an ethanolic medium. The forma-

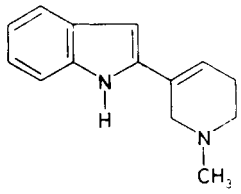
tion of the methoxy derivative was not influenced by quality of the ethanol used (96% ethanol denatured with toluene, absolute ethanol, 96% undenatured ethanol). The methoxy derivative *IIa* probably does not arise from the ethoxypiperidine *IIh* because on further irradiation the methoxy derivative disappears faster, affording a polymeric product. Attempted addition of ethanol to the indole derivatives *Ih* and *III* gave complex mixtures of unidentified compounds.



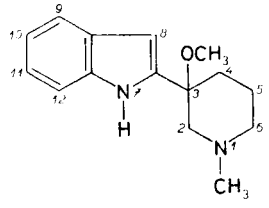
Ia, $R^1 = R^2 = CH_3$
Ib, $R^1 = C_2H_5$; $R^2 = CH_3$
Ic, $R^1 = CH_2C_6H_5$; $R^2 = CH_3$
Id, $R^1 = H$; $R^2 = CH_3$
Ie, $R^1 = CH_3$; $R^2 = CH_2CH_2CH_3$
If, $R^1 = CH_3$; $R^2 = CH(CH_3)_2$
Ig, $R^1 = CH_2CH{=}CH_2$; $R^2 = CH_3$
Ih, $R^1 = CH_3$; R^2 = 2-indolyl



IIa, $R^1 = R^2 = R^3 = CH_3$
IIb, $R^1 = C_2H_5$; $R^2 = R^3 = CH_3$
IIc, $R^1 = H$; $R^2 = R^3 = CH_3$
IId, $R^1 = R^3 = CH_3$; $R^2 = CH_2CH_2CH_3$
IIe, $R^1 = R^3 = CH_3$; $R^2 = CH(CH_3)_2$
IIf, $R^2 = R^3 = CH_3$; $R^1 = CH_2CH{=}CH_2$
IIg, $R^1 = R^3 = CH_3$; R^2 = 2-indolyl
IIh, $R^1 = R^2 = CH_3$; $R^3 = C_2H_5$



III



IV

Irradiation of methanol with tetrahydropyridine *Ia* in the presence of toluene and sulfuric acid did not lead to the desired addition products. Whereas the hitherto described[3-5] photoadditions are catalyzed by sulfuric acid we obtained, besides the starting *Ia*, a complex high-boiling mixture containing no addition product *IIa* (GLC).

We tried also to add methanol to tetrahydropyridines *Ia* and *If* without irradiation under catalysis with sulfuric acid either at reflux temperature or in a sealed tube on a water bath; however, we isolated only about 50% of the starting base *Ia* or *If* together with a polymeric material which decomposed upon attempted distillation *in vacuo*.

All the 1,2,3,6-tetrahydropyridine derivatives studied are known except 4-methyl--1-(2-propenyl)-1,2,3,6-tetrahydropyridine (*Ig*). We prepared this compound by reduction of 4-methyl-1-(2-propenyl)pyridinium bromide with sodium borohydride.

EXPERIMENTAL

The photochemical reactions were performed in a 120, 200 or 250 ml apparatus[16] using quartz or Pyrex filters (cut-off 295 nm) at 20—30°C. For reactions with the bases *Ia—If* the reaction vessel was modified to allow magnetic stirring. Prior to the reaction, dry nitrogen was introduced into the mixture for 15 min and a gentle stream of nitrogen was maintained during the whole reaction. The reaction mixtures were irradiated with a Tesla RVK 125 medium-pressure lamp and the reaction time was optimized for each particular experiment. Liquid mixtures were analyzed by gas–liquid chromatography on a Chrom 5 chromatograph (250 × 0·3 cm column, carrier gas nitrogen, 15% SE 301 on Chromosorb N-AW-DMCS or 15% Carbowax 20 M on Chromaton N-AW-HMDS, FID detector). Thin-layer chromatography (TLC) was done on Silufol UV 254 (Kavalier, Votice), spots were detected by UV light at 254 and 366 nm or by iodine vapours. Proton NMR spectra were taken on a Varian XL-100-15 (100 MHz), Tesla 567 (100 MHz) or Bruker AM 400 (400.133 MHz) instrument in deuteriochloroform with tetramethylsilane as internal standard. Chemical shifts δ are given in ppm, coupling constants J in Hz. Mass spectra were measured on a Gas Chromatograph–Mass Spectrometer 9000 LKB Produkter AB (Stockholm) or Jeol DX 303/DA 5 000 (70 eV, source temperature 250°C, direct inlet). The temperature data are uncorrected, crystalline analytical samples were dried at 7 . . 10^{-2} kPa for 6 h.

4-Methoxy-1,4-dimethylpiperidine (*IIa*)

A mixture of dimethyltetrahydropyridine *Ia* (ref.[17], 0·97 g; 8·7 mmol), methanol (180 ml), and toluene (20 ml) was irradiated for 8 h. After the end of the reaction, the mixture was acidified with HCl-saturated methanol and the solvents were evaporated at 30—40°C *in vacuo*. The oily residue was dissolved in small amount of water, made alkaline with 10% sodium hydroxide and the bases were taken up in ether. After drying over solid potassium hydroxide and removal of ether by distillation through a column, the residue was distilled *in vacuo*, affording a mixture (0·45 g) of *Ia* (35%) and the product (65%; according to GLC). The reaction mixtures from three experiments were combined and fractionated *in vacuo*. The highest-boiling fraction (160°C (bath)/2·0 kPa) consisted of pure methoxy derivative *IIa*. For $C_8H_{17}NO$ (143·2) calculated: 67·09% C, 11·96% H, 9·78% N; found: 66·81% C, 11·67% H, 9·85% N. ^{1}H NMR spectrum: 1·16 s, 3 H (C—CH_3); 1·40—1·97 m, 4 H (H-3, H-3, H-5, H-5); 2·30 s and 2·13—2·62 m, 7 H (NCH_3, H-2, H-2, H-6, H-6); 3·20 s, 3 H (OCH_3). Mass spectrum: *m/z* (relative intensity, %): 143 (13), 111 (27), 110 (32), 96 (100).

1-Ethyl-4-methoxy-4-methylpiperidine (*IIb*)

A mixture of tetrahydropyridine *Ib* (ref.[18]; 0·50 g; 4·0 mmol), methanol (81 ml), and toluene (9 ml) was irradiated for 4 h. Distillation afforded 0·46 g of a mixture consisting of 35% of *Ib*, 54% of *IIb*, and 11% of four unidentified products. The experiment was repeated with the same quantities of reactants and the reaction mixtures were combined and rectified, giving *IIb*, b.p. 95°C/4·4 kPa. For $C_9H_{19}NO$ (157·2) calculated: 68·74% C, 12·18% H, 8·91% N; found: 69·02% C, 12·07% H, 8·68% N. ^{1}H NMR spectrum: 1·15 s, 3 H (C—CH_3); 1·08 t, 3 H (CH_2CH_3, $J = 7$);

1·43—1·93 m, 4 H (H-3, H-3, H-5, H-5); 2·13—2·65 m, 6 H (H-2, H-2, H-6, H-6, CH_2CH_3); 3·18 s, 3 H (OCH_3).

4-Methoxy-1-methyl-4-propylpiperidine (*IId*)

A mixture of methylpropyltetrahydropyridine *Ie* (ref.[19]; 0·95 g; 6·8 mmol), methanol (180 ml), and toluene (20 ml) was irradiated for 16 h and processed as described for *Ia*; yield 0·6 g of a mixture containing (GLC) 30% of *Ie*, 3% of compounds of shorter elution time than *Ie*, and 67% of *IId*. The latter was isolated by fractionation *in vacuo*, b.p. 93°C/2·3 kPa. For $C_{10}H_{21}NO$ (171·3) calculated: 70·12% C, 12·36% H, 8·18% N; found: 70·37% C, 12·02% H, 8·23% N. ^{1}H NMR spectrum: 0·83—0·98 m, 3 H (CH_3); 1·29—1·90 m, 8 H ($CH_2CH_2CH_3$, H-3, H-3, H-5, H-5); 2·26 s and 2·08—2·59 m, 7 H (NCH_3, H-2, H-2, H-6, H-6); 3·11 s, 3 H (OCH_3). Mass spectrum: *m/z* (relative intensity, %): 171 (6), 141 (20), 140 (23), 128 (11), 112 (18), 110 (19), 96 (100).

4-Methoxy-1-methyl-4-(2-propyl)piperidine (*IIe*)

A mixture of tetrahydropyridine *If* (ref.[19]; 1·01 g; 7·2 mmol), methanol (180 ml), and toluene (20 ml) was irradiated for 23 h. The usual work-up gave a mixture (0·50 g) containing 23% of the starting *If*, 71% of *IIe*, and 6% of three unidentified compounds. The methoxy derivative *IIe* was isolated from a larger-scale experiment by fractional distillation at 200°C (bath)/3·3 kPa. For $C_{10}H_{21}NO$ (171·3) calculated: 70·12% C, 12·36% H, 8·18% N; found: 70·31% C, 12·09% H, 7·87% N. ^{1}H NMR spectrum: 0·87 d, 6 H (2 × CH_3, $J = 7$); 1·50—2·70 m, 9 H (4 × CH_2 and CH_3CHCH_3); 2·24 s, 3 H (NCH_3); 3·09 s, 3 H (OCH_3). Mass spectrum: *m/z* (relative intensity, %): 171 (7), 141 (23), 140 (36), 128 (30), 96 (100).

2-(4-Methoxy-1-methyl-4-piperidyl)indole (*IIg*)

A solution of tetrahydropyridylindole *Ih* (ref.[20]; 0·2 g; 0·94 mmol) in methanol (120 ml) was irradiated for 1 h. The experiment was repeated twice. A small amount of polymeric material (not melting up to 310°C, soluble only in dimethyl sulfoxide) was removed by filtration and the combined filtrates were taken down *in vacuo*. The residue (0·72 g) was mixed with ether and another portion of the insoluble polymeric material was filtered off. Ether was evaporated *in vacuo* and the residue (0·23 g; 34%) was crystallized from cyclohexane. The obtained faintly yellow crystalline *IIg*, m.p. 145—147°, was homogenous according to TLC in chloroform–2-propanol–25% aqueous ammonia–water (10 : 5 : 1 : 1). The product was sublimable. For $C_{15}H_{20}N_2O$ (244·3) calculated: 73·37% C, 8·25% H, 11·47% N; found: 73·44% C, 8·31% H, 11·28% N. ^{1}H NMR spectrum: 2·26—2·46 m, 4 H (H-3, H-3, H-5, H-5); 2·58 s, 3 H (N—CH_3); 2·61 to 2·78 m, 4 H (H-2, H-2, H-6, H-6); 3·03 s, 3 H (OCH_3); 6·40 s, 1 H (H-8); 6·98—7·61 m, 4 H (H-9, H-10, H-11, H-12); 8·48 s, 1 H (NH). Mass spectrum: *m/z* (relative intensity, %): 244 (75), 229 (22), 213 (48), 174 (93), 70 (94), 71 (100).

2-(3-Methoxy-1-methyl-3-piperidyl)indole (*IV*)

A solution of tetrahydropyridylindole *III* (ref.[15]; 0·2 g; 0·94 mmol) in methanol (120 ml) was irradiated for 1 h. Reaction mixtures from three irradiation experiments were combined and the solvent was evaporated to give 0·8 g of dark brown crystalline compound which was boiled with cyclohexane (200 ml). The solution was taken down and the residue was dissolved in small amount of benzene. Upon addition of cyclohexane, the crystalline *IV* (0·22 g; 32%) was obtained; m.p. 154—156°C (cyclohexane). For $C_{15}H_{20}N_2O$ (244·3) calculated: 73·37% C, 8·25% H,

11·47% N; found: 73·67% C, 8·31% H, 11·30% N. ^{1}H NMR spectrum: 1·06—2·03 m, 4 H; (H-4, H-4, H-5, H-5), 2·35 s, 4 H (NCH_3 and H_{ax}-6); 2·58—2·75 m, 3 H (H-2, H-2, H_{eq}-6); 3·14 s, 3 H (OCH_3); 6·42 s, 1 H (H-8); 7·07—7·38 m, 3 H (H-10, H-11, H-12); 7·58 d, 1 H (H-9); 8·98 s, 1 H (NH). Mass spectrum: *m/z* (relative intensity, %): 244 (26), 213 (12), 186 (32), 58 (100).

4-Ethoxy-1,4-dimethylpiperidine (*IIh*)

A mixture of tetrahydropyridine *Ia* (ref.[17]; 1·11 g; 10 mmol), ethanol (180 ml), and toluene (20 ml) was irradiated for 15·5 h. The obtained mixture (0·80 g) of 3 bases (GLC) was separated by preparative GLC into *Ia* (57%), *IIa* (13%), and *IIh* (30%), b.p. 170°C (bath)/2·0 kPa. Compound *IIh*: For $C_9H_{19}NO$ (157·2) calculated: 68·74% C, 12·18% H; found: 69·01% C, 11·95% H. ^{1}H NMR spectrum: 1·16 t (OCH_2CH_3, $J = 7$); 1·14 s, together with the preceding signal 6 H (CH_3-4); 1·28—1·93 m, 4 H (H-3, H-3, H-5, H-5); 2·24 s, 3 H (NCH_3); 2·10—2·59 m, 4 H (H-2, H-2, H-6, H-6); 3·33 q, 2 H (OCH_2CH_3, $J = 7$). Mass spectrum: *m/z* (relative intensity, %): 157 (5), 113 (52), 112 (47), 96 (100).

4-Methoxy-4-methyl-1-(2-propenyl)piperidine (*IIf*)

A solution of tetrahydropyridine *Ig* (1·05 g; 7·7 mmol) in a mixture of methanol (225 ml) and toluene (25 ml) was irradiated for 10 h. Reaction mixtures from two experiments were combined and processed as described for *IIa*. Distillation *in vacuo* afforded 1·40 g of a mixture containing 15% of four unidentified compounds, 55% of starting *Ig*, and 30% of the product *IIf*. Neither fractional distillation nor preparative gas–liquid chromatography gave completely pure *IIf*; in all cases the compound contained some starting *Ig*. ^{1}H NMR spectrum: 1·14 s, 3 H (CH_3-4); 1·38—1·85 m, (H-3, H-3, H-5, H-5); 2·00—2·72 m, (C-2, C-2, C-6, C-6); 2·99 d (NCH_2, $J = 6$); 3·16 s, 3 H (OCH_3); 4·84—5·24 m, 2 H (=CH_2); 5·50—6·05 m, 1 H (=CH).

4-Methyl-1-(2-propenyl)-1,2,3,6-tetrahydropyridine (*Ig*)

Allyl bromide (42·4 g) was gradually added to a solution of 4-methylpyridine (27·1 g; 0·29 mol) in methanol (135 ml) and the mixture was refluxed for 11 h. Evaporation of methanol *in vacuo* afforded 61·2 g (98%) of yellow oil to which was added water (290 ml), followed by 1M-NaOH (290 ml) and sodium borohydride (11·4 g). The usual procedure[17], followed by fractionation under nitrogen, furnished 24·4 g (62%) of *Ig*, b.p. 65—68°C/2·4 kPa. The product deteriorated rapidly on exposure to light and air. For $C_9H_{15}N$ (137·2) calculated: 78·78% C, 11·02% H, 10·20% N; found: 78·83% C, 11·01% H, 10·07% N. ^{1}H NMR spectrum: 1·66 s, 3 H (CH_3-4); 1·9—2·22 m, 2 H (H-5, H-5); 2·46—2·64 m, 2 H (H-6, H-6); 2·78—2·98 m, 2 H (H-2, H-2); 3·04 d, 2 H (NCH_2, $J = 7$); 5·06—5·44 m, 3 H (=CH_2 and H-3); 5·66—6·10 m, 1 H (=CH). Picrate, m.p. 82—83°C (ethanol). For $C_{15}H_{18}N_4O$ (368·3) calculated: 49·18% C, 4·95% H, 15·30% N; found: 49·34% C, 4·94% H, 15·18% N. Methiodide, m.p. 142·5—143·5°C (ethanol). For $C_{10}H_{18}IN$ (279·2) calculated: 43·03% C, 6·50% H, 45·46% I, 5·02% N; found: 43·28% C, 6·58% H, 45·50% I, 4·75% N

Attempted Photoaddition of Methanol to *Ic*

A solution of tetrahydropyridine *Ic* (ref.[21]; 2·1 g; 11·2 mmol) in a mixture of methanol (180 ml) and toluene (20 ml) was irradiated for 12 h. Distillation afforded 0·35 g of a mixture containing 15% of *Id*, 58% of *Ic*, and 37% of 1,2-diphenylethane (identified by ^{1}H NMR and mass spectrum).

The elemental analyses were carried out under the supervision of Dr L. Helešic, the NMR *spectral measurements were supervised by Dr P. Trška. Mass spectra were taken by Dr P. Mitera and Dr P. Zachař in the Central Laboratories of Prague Institute of Chemical Technology.*

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Translated by M. Tichý.