Photochemical Cyclisation of Phthalimide Mannich Bases

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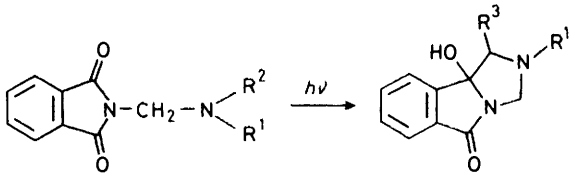
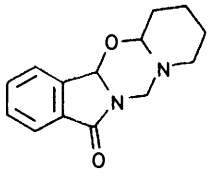
Ultraviolet irradiation of Mannich bases derived from phthalimide, formaldehyde, and a secondary amine leads to cyclised products containing a new imidazolidine ring. The reaction is much less efficient when the secondary amine has an aromatic group adjacent to the nitrogen atom. Exceptionally, the Mannich base derived from 3-pyrroline does not cyclise but undergoes an internal oxidation–reduction reaction to give a substituted pyrrole.

A NUMBER of recent reports have demonstrated that photochemical cyclisation of *N*-substituted phthalimides can be used in the synthesis of heterocyclic compounds with new rings containing one nitrogen atom,[1] a nitrogen and an oxygen atom,[2] a nitrogen and a sulphur atom,[3] or two nitrogen atoms.[4,5] In the case of compounds with a sulphur-containing substituent, new macrocyclic compounds with rings of up to 21 atoms [6] have been made. We have previously shown [5] that *N*-(dibenzylaminomethyl)phthalimide (1; $R^1 = R^2 = PhCH_2$), the Mannich base from phthalimide, formaldehyde, and dibenzylamine, undergoes reaction on irradiation to give an imidazo[4,3-*a*]isoindole derivative (2; $R^1 = PhCH_2$, $R^3 = Ph$), and also [7] that similar compounds with new 6- or 7-membered rings containing two nitrogen atoms can be formed from related substrates.



(1) (2)



(3)

Another group has isolated [4] similar products from phthalimide Mannich bases of other secondary amines (1; $R^1 = R^2 = Me$, Bu^i; R^1, $R^2 = [CH_2]_5$, $[CH_2]_2O[CH_2]_2$); from the Mannich base with piperidine, a different type of cyclised product (3) was also formed. In producing this oxadiazine derivative the behaviour of the phthalimide resembles that of an amino-substituted 1,4-naphthoquinone.[8] The photochemical reaction probably occurs through an upper excited state,[9] but this does not provide an explanation of why only the piperidine-derived Mannich base behaves differently.

We now report that the photochemical reaction is generally applicable in the formation of multicyclic compounds with a new imidazolidine ring when hydrogen transfer occurs from an allylic or benzylic position. For phthalimides in which the side-chain nitrogen atom has an adjacent aromatic ring the reaction is much slower and is not as useful synthetically. In one system we report that an unexpected reaction leads to a product without cyclisation.

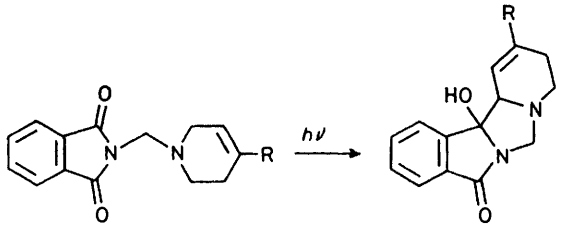
RESULTS AND DISCUSSION

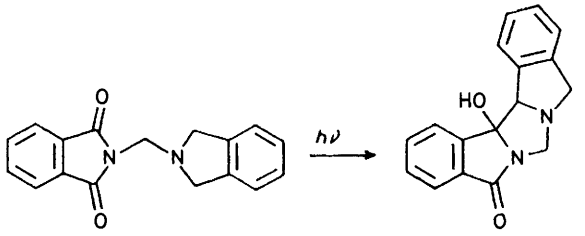
We found that the photocyclisation reaction occurs quite readily with further phthalimide Mannich bases derived from dialkylamines (*e.g.* 1; $R^1 = R^2 = Et$, Pr) but that the products are not easy to isolate from the reaction mixture after irradiation. This difficulty is implied in the other report,[4] in that a different method of product isolation was employed for each compound studied. The susceptibility of some imide Mannich bases to hydrolysis has been commented on,[10] but we found that this presented difficulties only for sterically hindered compounds such as (1; $R^1 = R^2 = Pr^i$). In this last example the Mannich base was always contaminated with phthalimide or *N*-(hydroxymethyl)-phthalimide. Compounds in which there is an allylic or benzylic hydrogen atom available at the site of reaction also react readily. The Mannich base from diallylamine (1; $R^1 = R^2 = CH_2{=}CH{-}CH_2$) leads to an allyl-vinyl substituted product (2; $R^1 = CH_2{=}CH{-}CH_2$, $R^3 = CH_2{=}CH$), whilst the precursors derived from allylmethylamine (1; $R^1 = Me$, $R^2 = CH_2{=}CH{-}CH_2$) or benzylmethylamine (1; $R^1 = Me$, $R^2 = PhCH_2$) give products (2; $R^1 = Me$, $R^3 = CH_2{=}CH$ or Ph) in which hydrogen abstraction has occurred from the allylic or benzylic positions rather than from the methyl group. Although the recovered yield of pure photoproduct in these last two examples was quite low, we were unable to find evidence for the alternative products (2; $R^3 = H$, $R^1 = CH_2{=}CH{-}CH_2$ or $PhCH_2$) in the reaction mixture.

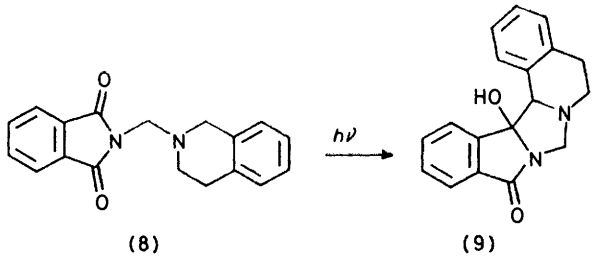
The irradiation of Mannich bases derived from 1,2,-3,6-tetrahydropyridines (4) yields pyrido[1′,2′:3,4]imidazo[5,1-*a*]isoindole derivatives (5) in which cyclisation has occurred to the allylic position (C-6). Similarly, the phthalimide Mannich base from isoindoline (6) and that from 1,2,3,4-tetrahydroisoquinoline (8) give rise to an isoindolo[2′,1′:3,4]imidazo[5,1-*a*]isoindole (7) and an isoindolo [2′,1′:3,4]imidazo[5,1-*a*]isoquinoline (9) respectively; in both cases the position of hydrogen abstraction is benzylic in nature.

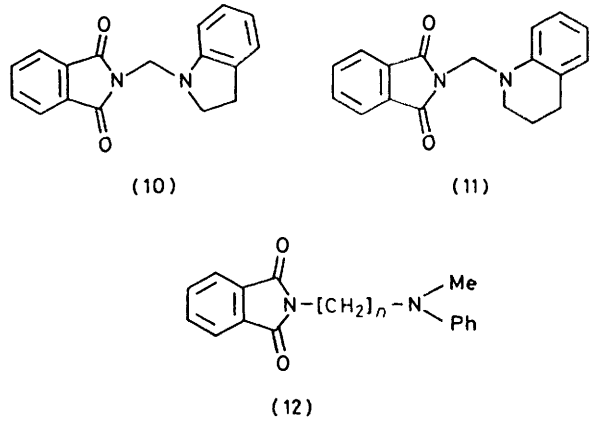
The structure of the products isolated is most readily

assigned on the basis of the ^1H n.m.r. spectrum. The most characteristic features of the spectrum for compounds (2) are an AB pair of doublets at δ 3.5—4.0 and 4.5—5.0 arising from the N–CH_2–N system of the imidazolidine ring, and a signal for a single proton corresponding to the N–CH–R^3 unit. The fact that products arise by preferential reaction at an allylic or benzylic position is a feature that would be expected for a photochemical sequence involving hydrogen abstraction by an excited carbonyl group. However, it has been shown [11] that in the photoreduction of benzophenone by amines, hydrogens are transferred with more or less equal facility from methyl or benzyl groups adjacent to the nitrogen, and this is ascribed to the very high rate constants for the electron-transfer and proton-transfer steps. Similarly, excited α,α,α-trifluoroacetophenone abstracts hydrogen atoms from cumene with little selectivity (after correction for statistical factors),[12] and the mechanism here is thought to involve some charge-transfer characteristics. It seems that the reaction of the aminoalkyl-substituted phthalimides differs from these ketone–amine reactions in this respect.



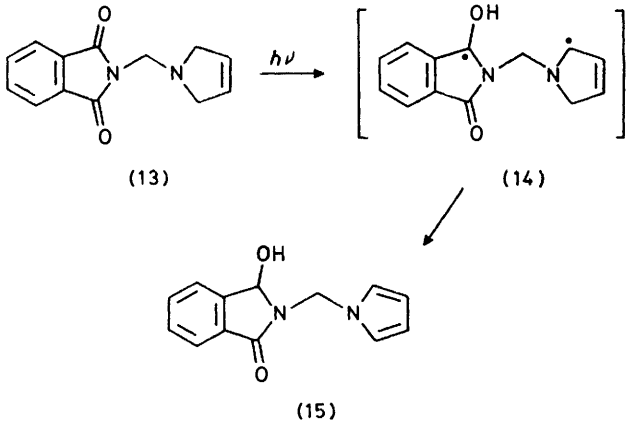






We studied a number of Mannich bases from phthalimide, formaldehyde, and secondary aromatic amines [(1; R^1 = Ph, R^2 = Me; R^1 = Ph, R^2 = CH_2=CH–CH_2; R^1 = Ph, R^2 = $PhCH_2$), (10) and (11)]. In each case we found that the photoreaction was very much slower than with compounds derived from aliphatic amines, and the photocyclisation reaction was not as useful synthetically since a mixture of products is formed. Only from (1; R^1 = Ph, R^2 = Me) could a pure sample of a single photoproduct be isolated. From a related series of mainly non-Mannich phthalimides (12) Kanaoka and co-workers [13] have isolated photoproducts (in relatively low yield) for n = 1—6, 10, and 12.

If the mechanism of photocyclisation involves some interaction of the amine nitrogen atom in an electron-donor capacity with the phthalimide part of the molecule,[9] then the presence of an aromatic group adjacent to the nitrogen atom would reduce such an interaction and could lower the quantum yield. Evidence that the imides derived from aromatic amines have excited-state properties significantly different from those derived from aliphatic amines is found in the substantially longer phosphorescence lifetime at 77 K (2.0 s as compared with *ca.* 1.0 s) [9] and in the observed charge-transfer interaction in both absorption and fluorescence spectra of compounds of type (12).[9,14]



An unexpected major product from the irradiation of *N*-(3-pyrrolin-1-ylmethyl)phthalimide (13) is the *N*-pyrrolylmethyl derivative of 3-hydroxyisoindolinone (15). This results from an intramolecular dehydrogenation–hydrogenation reaction, and it can be envisaged as arising from a biradical intermediate (14), which is itself a product of hydrogen-atom transfer. For this parti-

cular biradical there is an energetic factor favouring transfer of a second hydrogen atom from the amine to the imide, since the aromatic stabilisation energy of a pyrrole ring is involved. Such a large extra energetic factor is not present in the other systems studied. It has previously been reported [15] that photoreduction of *N*-methylphthalimide accompanies the photoaddition reactions with amines, and also that the intermolecular dehydration of pyrrolidines (to give pyrroles) can be effected using photo-excited benzophenone [16] or oxygen.[17] However, the nearest analogy in the literature to the process reported here is the proposed formation of a transient isoindole derivative from an isoindolinylnaphthoquinone.[18] Such a reaction does not occur with the isoindolinyl compound (6) that we have studied, but only with the pyrrolinyl compound (13).

EXPERIMENTAL

Preparation of Mannich Bases.—Method A. An equimolar mixture of phthalimide, formaldehyde (in aqueous solution), and secondary amine was warmed or boiled in ethanol until solution occurred. In a few instances involving aromatic amines some solid material was filtered off to avoid the use of unduly large volumes of solvent. The solution was cooled, and the Mannich base filtered off and purified by further recrystallisation. This method was used with the following amines: dipropylamine, dibenzylamine, allylmethylamine, benzylmethylamine, 1,2,3,6-tetrahydropyridine, 1,2,3,6-tetrahydro-4-phenylpyridine, isoindoline [prepared from 1,2-di(bromomethyl)benzene and toluene-*p*-sulphonamide [19]], 1,2,3,4-tetrahydroisoquinoline, 3-pyrroline, *N*-allylaniline, *N*-benzylaniline, indoline, and 1,2,3,4-tetrahydroquinoline.

Method B. An equimolar mixture of reagents was stirred with a small amount of water. After filtration the solid was recrystallised to give pure Mannich base. This method was used with diethylamine, diallylamine, dibenzylamine, benzylmethylamine, and *N*-methylaniline. Using this method isoindoline did not give the Mannich base but rather *N*-(*o*-carbamoylbenzoyl(isoindoline, m.p. 216—218 °C (Found: C, 72.15; H, 5.35; N, 10.4. $C_{16}H_{14}N_2O_2$ requires C, 72.29; H, 5.23; N, 10.53%); δ($CDCl_3$) 1.67 (s, 2 H, reduced on addition of D_2O), 4.52 (s, 2 H), 4.97 (s, 2 H), and 7.0—8.1 (m, 8 H); $\nu_{max.}$ 3 380, 3 190, 1 678, 1 610, 1 178, and 750 cm^{-1}.

Method C. A solution of *N*-bromomethylphthalimide (9.6 g, 0.040 mol) and di-isopropylamine (8.08 g, 0.080 mol) in benzene (60 cm^3) was heated under reflux for 12 h in an atmosphere of nitrogen. The mixture was cooled, the amine salt removed by filtration, and the solvent removed to leave the Mannich base. Attempts to purify the product further using silica chromatography, aqueous extraction, or recrystallisation from ethanol resulted in decomposition to phthalimide or *N*-hydroxymethylphthalimide.

All the Mannich bases gave satisfactory elemental analyses, and their i.r. and ^1H n.m.r. spectra were in accord with the expected molecular structures.

Irradiations.—These were carried out under nitrogen in a reactor vessel of *ca.* 350 cm^3 capacity. The solvent was acetone or benzene, and the lamp used was a Hanovia 450-W medium-pressure mercury arc with a Pyrex water-cooling jacket.

2-*Ethyl*-1,2,3,9b-*tetrahydro*-9b-*hydroxy*-1-*methylimidazo*-[4,3-a]*isoindol*-5-*one* (2; R^1 = Et, R^3 = Me).—A solution of *N*-(diethylaminomethyl)phthalimide (1; $R^1 = R^2$ = Et) (4.0 g, 0.020 mol) was irradiated for 4 h, and the residue was separated by chromatography (silica gel, $CHCl_3$–petrol) to give (2; R^1 = Et, R^3 = Me), yield 0.84 g (21%) (Found: C, 67.3; H, 7.0; N, 12.0. $C_{13}H_{16}N_2O_2$ requires C, 67.22; H, 6.94; N, 12.06%); δ($CDCl_3$) 1.18 (t, *J* 7 Hz) and 1.30 (d, *J* 7 Hz) (6 H), 2.69 (q, J 7 Hz, 2 H), 3.5 (s, 1 H, disappears after addition of D_2O), 3.80 (d, *J* 7 Hz) and 3.91 (q, *J* 7 Hz) (2 H), 4.70 (d, *J* 7 Hz, 1 H), and 7.2—7.9 (m, 4 H); $\nu_{max.}$ 3 300 and 1 700 cm^{-1}.

1-*Ethyl*-1,2,3,9b-*tetrahydro*-9b-*hydroxy*-2-*propylimidazo*-[4,3-a]*isoindol*-5-*one* (2; R^1 = Pr, R^3 = Et).—A solution of *N*-(dipropylaminomethyl)phthalimide (1; $R^1 = R^2$ = Pr) (5.0 g, 0.019 mol) was irradiated for 3.5 h. After removing the solvent, the residue was separated by chromatography (silica gel, $CHCl_3$) to give a pure sample of (2; R^1 = Pr, R^3 = Et), yield 0.60 g (12%) (Found: C, 69.2; H, 7.85; N, 11.05. $C_{15}H_{20}N_2O_2$ requires C, 69.19; H, 7.76; N, 10.76%); δ($CDCl_3$) 0.3—3.2 (m, 13 H), 3.5 (br s, 1 H, disappears on addition of D_2O), 3.85 and 4.75 (two doublets, *J* 7 Hz, 2 H), and 7.3—8.0 (m, 4 H); $\nu_{max.}$ 3 350 and 1 710 cm^{-1}.

Irradiation of (1; $R^1 = R^2$ = Pr^i).—After irradiation of *N*-(di-isopropylaminomethyl)phthalimide the crude product showed $\nu_{max.}$ at 3 300 cm^{-1} and an n.m.r. signal at δ 5.65 which disappeared after addition of D_2O. Attempts to isolate pure product, by extraction or silica chromatography, were unsuccessful.

2-*Allyl*-1,2,3,9b-*tetrahydro*-9b-*hydroxy*-1-*vinylimidazo*-[4,3-a]*isoindol*-5-*one* (2; R^1 = CH_2=CH-CH_2, R^3 = CH_2=CH).—A solution of *N*-(diallylaminomethyl)phthalimide (1; $R^1 = R^2$ = CH_2=CH-CH_2) (7.0 g, 0.027 mol) was irradiated for 13 h, and purification of the crude product by chromatography (silica gel, $CHCl_3$–petrol) gave (2; R^1 = CH_2=CH-CH_2, R^3 = CH_2=CH), yield 2.42 g (35%), m.p. 113—115 °C (Found: C, 70.0; H, 6.2; N, 10.85. $C_{15}H_{16}N_2O_2$ requires C, 70.20; H, 6.30; N, 10.93%); δ($CDCl_3$) 2.6—3.55 (m, 3 H), 3.65 (d, *J* 7 Hz, 1 H), 4.28 (s, 1 H, disappears after addition of D_2O), 4.62 (d, *J* 7 Hz, 1 H), 4.9—6.2 (m, 6 H), and 7.2—7.9 (m, 4 H); $\nu_{max.}$ 3 290, 1 690, and 1 642 cm^{-1}.

1,2,3,9b-*Tetrahydro*-9b-*hydroxy*-2-*methyl*-1-*vinylimidazo*-[4,3-a]*isoindol*-5-*one* (2; R^1 = Me, R^3 = CH_2=CH).—A solution of *N*-(allylmethylaminomethyl)phthalimide (1; R^1 = CH_2=CH-CH_2, R^2 = Me) (5.1 g, 0.022 mol) was irradiated for 12 h, and chromatographic separation (silica gel, $CHCl_3$) gave (2; R^1 = Me, R^3 = CH_2=CH) as a pale yellow solid, yield 1.05 g (21%) (Found: C, 67.7; H, 6.25; N, 12.05. $C_{13}H_{14}N_2O_2$ requires C, 67.80; H, 6.14; N, 12.17%); δ($CDCl_3$) 2.14 (s, 3 H), 3.13 (d, *J* 7 Hz, 1 H), 3.53 (d, *J* 7 Hz, 1 H), 4.53 (d, *J* 7 Hz, 1 H), 4.7—6.2 (m, 4 H, reduced to 3 H with D_2O), and 6.9—7.9 (m, 4 H); $\nu_{max.}$ 3 350 and 1 700 cm^{-1}.

1,2,3,9b-*Tetrahydro*-9b-*hydroxy*-2-*methyl*-1-*phenylimidazo*[4,3-a]*isoindol*-5-*one* (2; R^1 = Me, R^3 = Ph).—A solution of *N*-(benzylmethylaminomethyl)phthalimide (1; R^1 = Me, R^2 = $PhCH_2$) (4.7 g, 0.017 mol) was irradiated for 5 h, and chromatography (silica gel, $CHCl_3$–petrol) gave (2; R^1 = Me, R^3 = Ph), yield 1.83 g (39%). A small sample of one stereoisomer was obtained by boiling the product with ether, m.p. 154—156 °C (Found: C, 72.6; H, 5.85; N, 9.8. $C_{17}H_{16}N_2O_2$ requires C, 72.83; H, 5.76; N, 10.00%); δ($CDCl_3$) 2.16 (s, 3 H), 3.60 (d, *J* 7 Hz), 3.60 (s) and 3.90 (br s, disappears after addition of D_2O) (total 3 H),

4.72 (d, *J* 7 Hz, 1 H), 6.25—6.65 (m, 1 H), and 6.65—7.7 (m, 4 H); $\nu_{max.}$ 3 350 and 1 690 cm^{-1}.

4,6,12b,12c-*Tetrahydro*-12b-*hydroxypyrido*[1′,2′:3,4]-*imidazo*[5,1-a]*isoindol*-8(3H)-*one* (5; R = H).—A solution of *N*-(1,2,3,6-tetrahydropyridin-1-ylmethyl)phthalimide (4; R = H) (5.0 g, 0.021 mol) was irradiated for 11 h. Recrystallisation of the residue from ethanol gave (5; R = H) (1.3 g, 26%) which was further purified by recrystallisation from benzene, m.p. 172—175 °C (decomp.) (Found: C, 69.3; H, 5.9; N, 11.45. $C_{14}H_{14}N_2O_2$ requires C, 69.39; H, 5.84; N, 11.56%); δ($CDCl_3$) 1.5—2.9 (m, 4 H), 3.8 (m, 1 H), 4.11 (d, *J* 9 Hz, 1 H), 4.45 (br s, disappears after addition of D_2O, 1 H), 4.41 (d, *J* 9 Hz, 1 H), 5.50 (s, 2 H), and 7.0—7.8 (m, 4 H); $\nu_{max.}$ 3 290 and 1 705 cm^{-1}.

4,6,12b,12c-*Tetrahydro*-12b-*hydroxy*-2-*phenylpyrido*-[1′,2′:3,4]*imidazo*[5,1-a]*isoindol*-8(3H)-*one* (5; R = Ph).—A solution of *N*-(4-phenyl-1,2,3,6-tetrahydropyridin-1-ylmethyl)phthalimide (4; R = Ph) (4.0 g, 0.013 mol) was irradiated for 15 h. After removing the solvent, addition of chloroform (10 cm^3) gave a pure sample of (5; R = Ph) (0.5 g, 13%), m.p. 177—180 °C (Found: C, 75.35; H, 5.4; N, 8.95. $C_{20}H_{18}N_2O_2$ requires C, 75.44; H, 5.71; N, 8.80%); δ([$^2H_6$]DMSO) 1.9—2.95 (m, 4 H), 4.09 (d, *J* 4 Hz, 1 H), 4.32 (d, *J* 10 Hz, 1 H), 4.65 (d, *J* 10 Hz, 1 H), 6.40 (d, *J* 4 Hz, 1 H), 7.05—7.45 (5 H), and 7.45—7.95 (m, 4 H); $\nu_{max.}$ 3 350 and 1 700 cm^{-1}.

7,9,13b,13c-*Tetrahydro*-13c-*hydroxyisoindolo*[2′,1′:3,4]-*imidazo*[5,1-a]*isoindol*-5-*one* (7).—A solution of *N*-(*N*′-dihydroisoindolylmethyl)phthalimide (6) (4.7 g, 0.017 mol) was irradiated for 7 h. A total of 2.07 g (44%) of (7) precipitated, and this could be purified by recrystallisation from ethanol, m.p. 176—178 °C (decomp.) (Found: C, 73.65; H, 5.15; N, 9.8. $C_{17}H_{14}N_2O_2$ requires C, 73.36; H, 5.08; N, 10.16%); δ(CD_3CO_2D) 4.38 (d, *J* 16 Hz, 1 H), 4.93 (d, *J* 16 Hz), 4.96 (d, *J* 10 Hz), and 5.20 (d, *J* 10 Hz) (total 3 H), 5.68 (s, 1 H), and 6.4—8.0 (m, 8 H); $\nu_{max.}$ 3 640 and 1 715 cm^{-1}.

9,10,14b,14c-*Tetrahydro*-14c-*hydroxyisoindolo*[2′,1′:3,4]-*imidazo*[5,1-a]*isoquinol*-5(7H)-*one* (9).—A solution of *N*-(1,2,3,4-tetrahydroisoquinolin-2-ylmethyl)phthalimide (8) (5.0 g, 0.017 mol) was irradiated for 10 h. Recrystallisation of the residue from benzene gave (9) (1.75 g, 35%), m.p. 180—183 °C (Found: C, 73.9; H, 5.5; N, 9.6. $C_{18}H_{16}N_2O_2$ requires C 73.94; H, 5.53; N, 9.58%); δ($CDCl_3$) 1.8—3.8 (m, 4 H), 3.8—5.2 (m, 4 H, loss of 1 H after addition of D_2O), and 6.7—8.0 (m, 8 H); δ[$CDCl_3$ with Eu(fod)$_3$] 2.2—3.5 (m, 4 H), 4.85 (s), 5.20 (d, *J* 10 Hz), and 5.40 (s) (total 3 H), 6.27 (d, *J* 10 Hz, 1 H), and 6.8—9.0 (m, 8 H); $\nu_{max.}$ 3 000 (br) and 1 695 cm^{-1}.

1,2,3,9b-*Tetrahydro*-9b-*hydroxy*-1-*phenylimidazo*[4,3-a]-*isoindol*-5-*one* (2; R^1 = Ph, R^3 = H).—A solution of *N*-(*N*′-methylanilinomethyl)phthalimide (1; R^1 = Ph, R^2 = Me) (3.5 g, 0.013 mol) was irradiated for 45 h. Chromatography (silica gel, $CHCl_3$–petrol) gave (2; R^1 = Ph, R^3 = H), yield 0.51 g (15%), m.p. 177—178 °C (Found: C, 72.2; H, 5.35; N, 10.5. $C_{16}H_{14}N_2O_2$ requires C, 72.16; H, 5.30; N, 10.52%); δ($CDCl_3$) 3.7 (br s, 1 H, disappears after addition of D_2O), 3.90 (s, 2 H), 4.16 (br s, 2 H), 6.9—7.8 (m, 9 H); $\nu_{max.}$ 3 350 and 1 695 cm^{-1}.

3-*Hydroxy*-2-(N-*pyrrolylmethyl*)*isoindolin*-1-*one* (15).—A solution of *N*-(3-pyrrolin-1-ylmethyl)phthalimide (13) (4.0 g, 0.018 mol) was irradiated for 3.5 h. Column chromatography (silica gel, chloroform) followed by recrystallisation from benzene gave (15) (15%) as a white solid, m.p. 168—169 °C (Found: C, 68.2; H, 5.35; N, 12.2. $C_{13}H_{12}N_2O_2$ requires C, 68.40; H, 5.30; N, 12.27%); δ($CDCl_3$) 3.89 (br d, 1 H, disappears after D_2O addition), 5.03 (d, *J* 14 Hz, 1 H), 5.51 (d, *J* 14 Hz) and 5.59 (d, *J* 12 Hz, becomes singlet after D_2O addition) (total 2 H), 6.09 (d, *J* 2 Hz, 2 H), 6.78 (d, *J* 2 Hz, 2 H), and 7.2—7.9 (m, 4 H); $\nu_{max.}$ 3 330 and 1 690 cm^{-1}.

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