Synthesis of Diazaheterocycles with a Bridgehead Nitrogen by Photocyclisation of *N*-Substituted Alicyclic Imides

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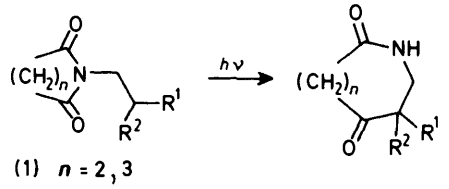
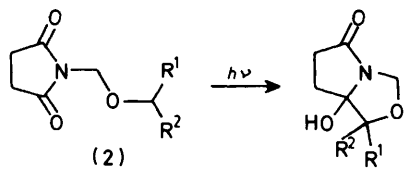
N-(Dialkylaminomethyl)-succinimides or -glutarimides give 1,3-diazabicyclo[3.3.0]octanes or -[4.3.0]-nonanes on irradiation in acetonitrile, accompanied by variable amounts of the parent imide. Two diastereoisomers of the products can be obtained, and the relative stereochemistry assigned on the basis of n.m.r. data. With unsymmetrical substrates mixtures of products are formed that demonstrate orientational preferences. Analogous *N*-substituted pyrrolidin-2-ones do not give photocyclised products (an unusual cleavage product is isolated in low yield), and similar dihydrouracils are relatively photostable. *N*-(Dialkylaminoethyl) aliphatic imides give azepine- or azocine-diones on irradiation, whereas *N*-(dialkylaminopropyl) compounds undergo photocyclisation to products with a new perhydro-1,4-diazepine ring (as do a corresponding 3,4,5,6-tetrahydrophthalimide and phthalimide, although photoreduction is a major process for the latter system). An *N*-(dialkylaminobutyl)succinimide does not give products with a new perhydrodiazocine ring. An *N*-(dialkylaminoethyl)maleimide and the analogous 3,4,5,6-tetrahydrophthalimide give compounds that contain a new piperazine ring, which is in contrast to the saturated imide analogues but similar to the corresponding phthalimide; this process competes effectively with the more usual photoreactions of maleimides involving the carbon–carbon double bond.

The intramolecular photochemical hydrogen abstraction and cyclisation reactions of ketones have been extensively studied and applied in a number of synthetic sequences, but amides do not undergo analogous reactions. However, imides do resemble ketones in this respect, and photocyclisations of imides have been reported over the past decade,[1] especially those of phthalimides,[2] with particular interest in the formation of macrocyclic products (ring sizes up to 38 atoms) from phthalimides substituted on nitrogen with a long chain terminating in a dialkylamino or methylthio group.[3] There are far fewer reports of photocyclisation reactions for aliphatic imides: *N*-alkyl-succinimides and -glutarimides (1) give azepine- or azocine-diones on irradiation; [4] *N*-(alkoxymethyl)-succinimides (2) lead to pyrrolo[1,2-*c*]oxazoles in good yield,[5] and low yields of photocyclised products are obtained from *N*-(alkoxyethyl), *N*-(methylthioethyl) and related derivatives of succinimide. We now report [6] on our study of alicyclic imides substituted on nitrogen with a dialkylaminoalkyl group, which explores the scope of the reaction, and the relationship between systems based on saturated, unsaturated, and aromatic cyclic imides.



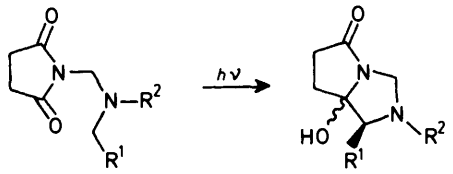
(1) $n = 2, 3$



(2)

Results and Discussion

Succinimides.—*N*-(Dialkylaminomethyl)succinimides (3)—(9) are readily prepared from succinimide, formaldehyde, and the appropriate secondary amine.[7] On irradiation in acetonitrile with light from a medium-pressure mercury arc (quartz filter) they give two diastereoisomers of 1,3-diazabicyclo[3.3.0]octanes in reasonable yield (based on unrecovered substrate), except for (4) which gave more than 90% of succinimide. The reason for this anomalous behaviour is not known, and only from (5) amongst the other succinimides was an appreciable amount (18%) of succinimide itself isolated after irradiation.

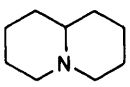


(3)	$R^1 = H$; $R^2 = Me$	52%
(4)	$R^1 = Me$; $R^2 = Et$	5 + 2%
(5)	$R^1 = CH=CH_2$; $R^2 = CH_2CH=CH_2$	26 + 20%
(6)	$R^1, R^2 = (CH_2)_4$	33 + 13%
(7)	$R^1, R^2 = CH=CHCH_2CH_2$	56 + 6%
(8)	$R^1, R^2 = CH_2OCH_2CH_2$	59 + 18%
(9)	$R^1, R^2 = o\text{-}C_6H_4CH_2CH_2$	44 + 7%

The assignment of structures for the photoproducts is made on the basis of elemental analysis and spectroscopic data. In particular there is evidence for a five-membered cyclic amide carbonyl ($\nu_{max.}$ 1 700 cm^{-1}; δ_C 175—180 p.p.m.), a quaternary carbon atom adjacent to nitrogen and bearing a hydroxy group (δ_C singlet at 95—100 p.p.m.; $\nu_{max.}$ 3 350 cm^{-1}), a methine group in place of the $CH_2(R^1)$ group of the substrate (δ_C doublet at 65—75 p.p.m.), and an NCH_2N group constrained in a ring system (δ_H AB pattern in the range 3—5 p.p.m.).

The yields given above are those for the two diastereoisomers of the cyclised product; for all the imides except (3), for which only one isomer of the product is possible, the diastereoisomers can be separated by silica-gel column

chromatography. The assignment of stereochemistry is based on interpretation of the n.m.r. spectra; to summarise our results and hypotheses, the isomers with OH and R^1 *trans* with respect to each other (*a*) exhibit a smaller difference in δ_H values for the two hydrogens of the cyclic NCH_2N group because of greater flexibility in the ring system; (*b*) show a smaller geminal coupling constant between these two hydrogens because of steric compression of the rings leading to a reduced dihedral angle; (*c*) have a lower δ_C value for the quaternary (C–OH) carbon due to bond lengthening that results from steric compression; and (*d*) have a lower δ_C value for the amide carbonyl because the amide nitrogen is more planar. The difference (0.1—1.2 p.p.m.) in δ_H for the hydrogen atoms of the NCH_2N group arises basically because the signal for one of the hydrogens is shifted upfield to around 3.6 p.p.m. from the position (typically 4.5 p.p.m.) of the signal for the NCH_2N protons in the imides (3)—(9). This effect is most probably caused by the lone pairs of the nitrogen atoms (an effect of the hydroxy group is ruled out because this should be very different for the two diastereoisomers, which is not the case). It has been shown [8] that there is a large δ_H difference (0.92 p.p.m.) between the geminal hydrogens adjacent to nitrogen in the spectrum of quinolizidine (10), and this is ascribed to the shielding effect of the axial lone pair on the axial hydrogens of the adjacent methylenes. The difference

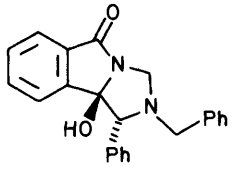


(10)

between the diastereoisomers arises because the cyclised product [*e.g.* from the imide (8)] with OH and R^1 groups *cis* has a more rigid structure, whereas that with OH and R^1 groups *trans* is more flexible and so the shielding effect of the lone pairs will be less pronounced. This hypothesis requires that inversion at the amine nitrogen in the photoproducts be unimportant (*i.e.* that the amine lone pair be essentially unidirectional), and support for this idea comes from the spectrum of the single isomeric photoproduct from imide (3). There is much less hindrance to nitrogen inversion in this compound, and at room temperature the signal for both hydrogens of the cyclic NCH_2N group appears as a singlet. Only as the temperature is lowered does the difference become apparent (the difference in δ_H is just over 0.1 p.p.m. at 220 K).

The diastereoisomer with OH and R^1 groups *trans* is more sterically strained than the other isomer, and this leads to a number of effects in the n.m.r. spectra. Steric compression leads to a small increase in the internal bond angle of the imidazolidine ring, and hence to a decrease in the angle between the geminal hydrogen atoms of the NCH_2N group; this would be expected [9] to lead to a reduction in the geminal coupling constant, as is observed. The steric compression is also taken up in two other ways. First the C–C bond between the bulky groups ($HO–C–C–R^1$) lengthens slightly, and this produces [10] a decrease in δ_C for one or both of the carbon atoms; in our systems a decrease is seen most clearly for the quaternary carbon (the signal for the other carbon shows a much smaller variation between the diastereoisomers). Secondly the steric strain is relieved in part by the amide nitrogen becoming more nearly planar; this results in a lower δ_C for the amide carbonyl (*ca.* 175 p.p.m., as compared with *ca.* 180 p.p.m. for the other isomer).

The hypotheses are plausible and consistent with the spectroscopic results obtained, and they are in keeping with spectral data for related photoproducts from phthalimide substrates,[11] where definite structural assignment has been made for compound (11) by *X*-ray crystallographic analysis.



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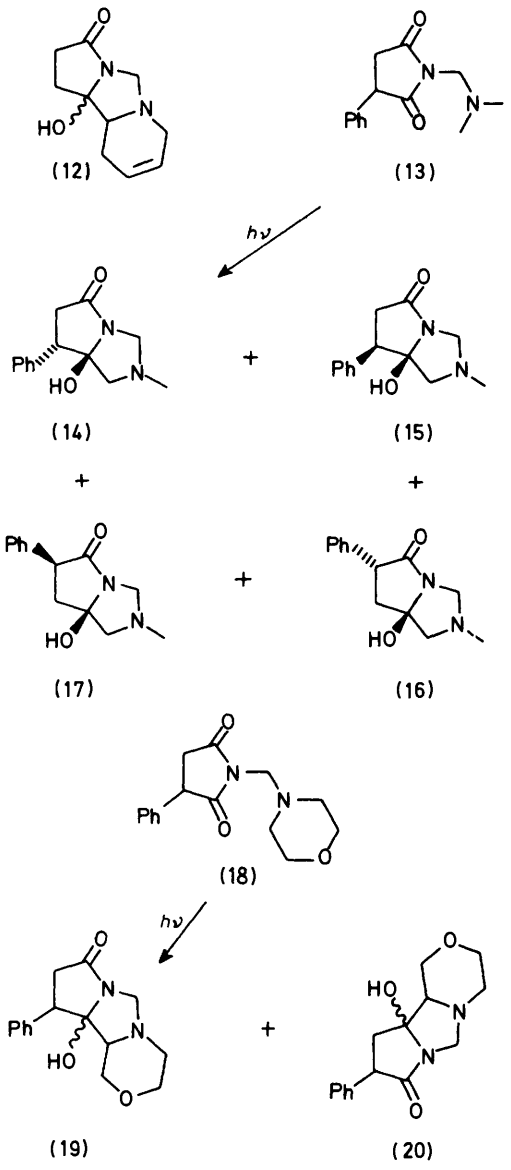
The ratio of isolated amounts of the two diastereoisomers ranges from 1.3 : 1 [for imide (5)] to 9 : 1 [for imide (7)]. Changing the solvent from acetonitrile to water for imide (8) does not affect the ratio of products, nor the isolated yield; however, in toluene the proportion of the major isomer increases (from 3.3 : 1 to *ca.* 8 : 1), although the isolated yield is lower. In toluene as solvent the reaction may be photosensitised, since the solvent absorbs much more strongly than the imide.

With imides (7) and (9) derived from an unsymmetrical amine, the major products arise by reaction at the allylic or benzylic methylene group, rather than at the less activated position. However, a small amount (*ca.* 6%) of a product (12) arising from the alternative mode of cyclisation was also isolated after irradiation of (7). The product (12) is distinguished from the major products in that the alkene gives rise to n.m.r. signals that are much closer together (δ_H 5.75—5.8; δ_C 124.5 p.p.m.; *cf.* δ_H 5.85—6.2; δ_C 128.2 and 122.2 p.p.m. for the major photoproduct). We were interested to see if there is also an orientational preference with regard to an unsymmetrical imide ring, and so the 2-phenylsuccinimide derivatives (13) and (18) were prepared and irradiated.

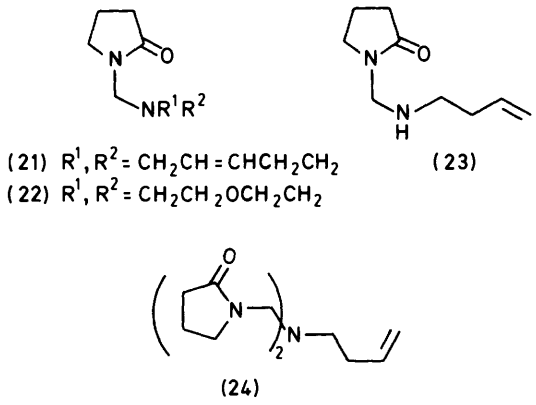
From (13), all four diastereoisomeric products (14)—(17) were formed; three were isolated in a pure state in the ratio 7 : 3 : 1, and were assigned structures (14), (15), and (16), respectively, and a smaller amount of the fourth (17) was obtained contaminated with one of the others. The main arguments in the assignment of orientation and stereochemistry are as follows. The δ_H value for one of the protons of the $CH_2(N)$ group adjacent to the quaternary C(OH) atom, and the δ_C value for the carbon atom of this group, are much lower for the major photoproduct than for the others (1.87 and 62.4 p.p.m., *cf.* 2.8—3.0 and *ca.* 66.3 p.p.m. for the other isomers). This is attributed to the shielding effect of the phenyl ring, and a study using molecular models indicates that this operates effectively only in isomer (14), which has the hydroxy and phenyl groups *trans* with respect to each other. The other product with the same orientation of cyclisation (15) is taken to be the one that shows a similar pattern of signals (doublets of doublets at 3.70, 3.44, and 2.62 p.p.m.) for the CH_2CH(Ph) protons as for (14) (dd at 3.66, 3.30, and 2.63 p.p.m.), whereas (16) shows a different pattern (dd at 3.80, 2.71, and 2.39 p.p.m.) and (17) also shows more signals (not completely resolved) in the 2.3—2.8 p.p.m. region. The assignment for (16) is based largely on the pattern of coupling constants for the CH_2CH(Ph) protons [*J* 14, 10, and 2.5 Hz, compared with 17, 8.5, and 2.5 Hz for (14), and 16, 12.5, and 8.5 Hz for (15)], which is taken to be characteristic of the relative stereochemistry of the OH and Ph groups.

From the reaction of (18) six products were identified by t.l.c., and the major one was isolated by silica-gel chromatography. Although the stereochemistry cannot be assigned, the orientation of cyclisation corresponds to structure (19) rather than (20), as judged by the δ_C value for the carbonyl group. We conclude that cyclisation can occur at either carbonyl



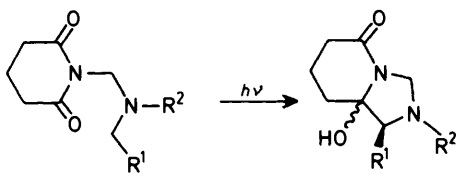
group in a 2-phenylsuccinimide system, but that there is a preference for reaction at the C-1 carbonyl (adjacent to CHPh). This could be due to an interaction of the partially positive amine nitrogen (arising from an initial charge transfer process) with the electrons of the phenyl ring, which promotes transfer of a hydrogen (or proton) to the C-1 carbonyl because it brings the reacting N–Me group closer to C-1 than to C-4. The major diastereoisomer (14) is the most hindered sterically, and it is possible for the *cis* relationship between the phenyl group and the ring methylene [$NCH_2C(OH)$] to be preserved if ring-closure follows very rapidly after the hydrogen or proton has been transferred.

Pyrrolidin-2-ones.—Saturated aliphatic amides are quite different from ketones or imides in their photochemical behaviour; they do not react with alkenes to form oxetanes, nor do they take part in intramolecular hydrogen abstraction reactions. For example, *N*-alkylpyrrolidin-2-ones undergo only cleavage reactions,[12] and there is no evidence for hydrogen abstraction from the alkyl group. As model amides related to succinimides (3)—(9), Mannich bases (21)—(22), of pyrrolidin-2-one were prepared and irradiated, but the photolysis produced complex mixtures of products. From (21) only one product was isolated pure, and this was identified on the basis of spectroscopic data as *N*-[(but-3-enylamino)methyl]pyrrolidin-2-one (23). The identity was confirmed by reaction with formaldehyde and pyrrolidin-2-one to give (24), which was synthesised independently from pyrrolidin-2-one, formaldehyde, and but-3-enylamine. Despite many attempts, (23) could not itself be made by a Mannich-type procedure.



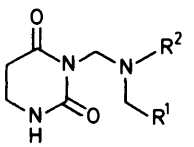
The formation of (23) from (21) is very unusual, involving the loss of a carbon atom from the tetrahydropyridine ring, and at this stage any mechanistic suggestions are purely speculative. The photolysis of (21) and (22) suggests that, even with *N*-dialkylaminomethyl substituents, saturated amides are not useful substrates for photochemical hydrogen abstraction and cyclisation.

Glutarimides.—Only two previous examples of glutarimide Mannich bases have been described,[13] based on the specific, pharmacologically active compounds bemegride and thalidomide. We needed to modify normal Mannich reaction procedures in order to prepare *N*-dialkylaminomethyl derivatives (24)—(27) of glutarimide; the corresponding derivatives of 3,3-dimethylglutarimide were also prepared but not irradiated. Irradiation of the glutarimides gave photoproducts with the 1,8-diazabicyclo[4.3.0]nonane ring system, and as with the succinimide analogues, two diastereoisomers were formed. In the case of imides (24), (25), and (26), glutarimide was also isolated in high yield (85, 50, and 53%).



(24)	$R^1,R^2 = (CH_2)_4$	0.6 + 0.4%
(25)	$R^1,R^2 = CH=CHCH_2CH_2$	30 + 8%
(26)	$R^1,R^2 = CH_2OCH_2CH_2$	8 + 4%
(27)	$R^1,R^2 = o\text{-}C_6H_4CH_2CH_2$	19 + 17%

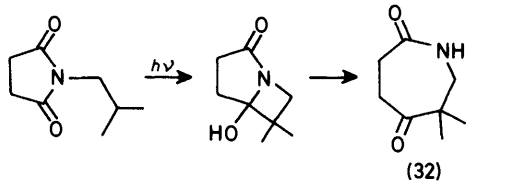
The glutarimides (24)—(27) react as readily as the corresponding succinimides, but this is not true of derivatives of another six-membered cyclic imide analogue, dihydrouracils. It has been reported [14] that dihydrouracil does not form Mannich bases, but we were able to prepare the derivatives (28)—(31). However, these compounds are photochemically stable, unlike the analogous five-membered ring hydantoins,[15] and



(28) $R^1, R^2 = (CH_2)_4$
(29) $R^1, R^2 = CH=CHCH_2CH_2$
(30) $R^1, R^2 = CH_2OCH_2CH_2$
(31) $R^1, R^2 = o\text{-}C_6H_4CH_2CH_2$

only small amounts of product mixtures appear, even after 50 times the extent of irradiation that is required for the imide derivatives to give products in greater than 50% conversion.

Formation of Larger Rings.—It has been reported previously that *N*-alkylphthalimides,[16] -glutarimides, or -succinimides [4,17] give ring-expanded products [*e.g.* (32)] by way of an initial fused azetidinol. We irradiated *N*-(dialkylaminoethyl) derivatives of succinimide, (33) and (34), glutarimide, (35), and 1,2,3,6-tetrahydrophthalimide, (36), to see if the presence of a nitrogen atom in the side-chain might lead instead to products with a new piperazine ring. In the event all of these systems gave perhydroazepine- or perhydroazocine-diones, as shown by the presence of ketone and amide carbonyl groups and the presence of an unchanged symmetrical amine substituent.



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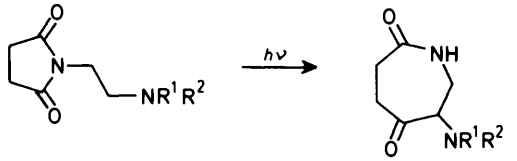
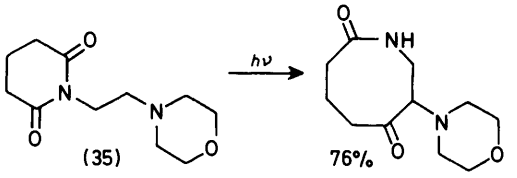
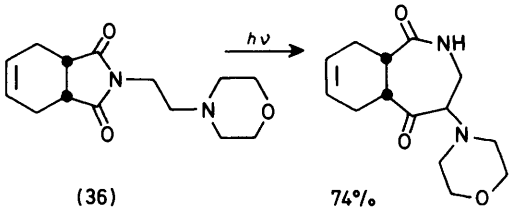


(33) $R^1 = R^2 = Me$ 17%
(34) $R^1, R^2 = CH_2CH_2OCH_2CH_2$ 46%



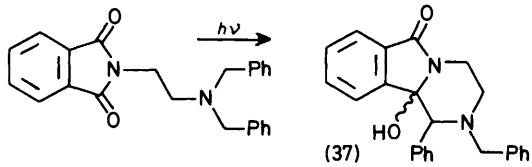
(35) 76%



(36) 74%

The reactions of the glutarimide (35) and the tetrahydrophthalimide (36) give good isolated yields of photoproduct. The value of this type of photochemical reaction for preparing azepinediones from *N*-alkylsuccinimides has been questioned,[18] and compounds with sulphur in the side-chain give only very low yields of product, but evidently some nitrogen-substituted derivatives can lead to much better yields. The formation of a photocyclised product from (36) is in direct contrast to the production of a cyclobutane dimer from the *N*-ethyl derivative of the same imide.[18] In principle this opens up a route to benzazepinediones which are not available from *N*-(dialkylaminoethyl)phthalimides [19] because of a competing reaction to give photocyclised products such as (37). However, our attempts to dehydrogenate the photoproduct from (36) using dichlorodicyanobenzoquinone were not successful, perhaps because of the α-aminoketone group in the molecule.

N-[3-(Dialkylaminopropyl)]-succinimides, (38) and (39), and -3,4,5,6-tetrahydrophthalimide, (40), were found to give cyclic products on irradiation that contain a new perhydro-1,4-diazepine ring. The main evidence for these product structures is that the compounds are isomeric with the substrates, contain an aromatic lactam carbonyl group and a quaternary (N)C(OH), do not have pairs of equivalent carbon atoms in an ' intact ' morpholine ring (as would be expected if a new five-membered ring were formed), but do have four protons on carbons adjacent to oxygen. Such reactions have been reported previously for the corresponding phthalimides,[20] but not for aliphatic imides. However, the isolated yields of these aliphatic products are not high, and it is possible that other isomeric photoproducts are present in the reaction mixtures. The diastereoisomers of the product from (40) exhibit unusual differences in the carbon-13 n.m.r. signals for the carbon atoms of the morpholine ring. These signals are at 68.5, 66.7, 66.5, and 52.7 p.p.m. for the major isomer, but at higher field (63.5, 61.1, 60.1, and 50.6 p.p.m.) for the other isomer. The effect probably arises because the more cage-like structure of the minor stereoisomer can hold some of the carbon atoms in the shielding zone of the C=C double bond.



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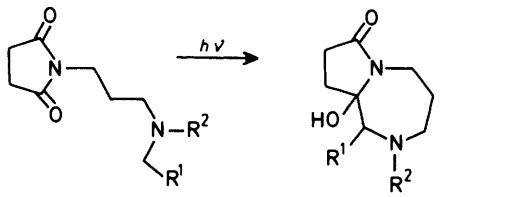
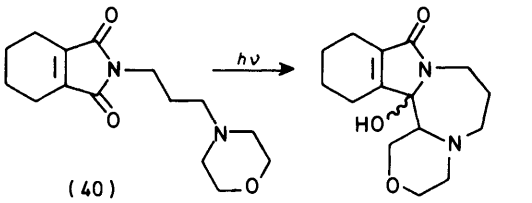


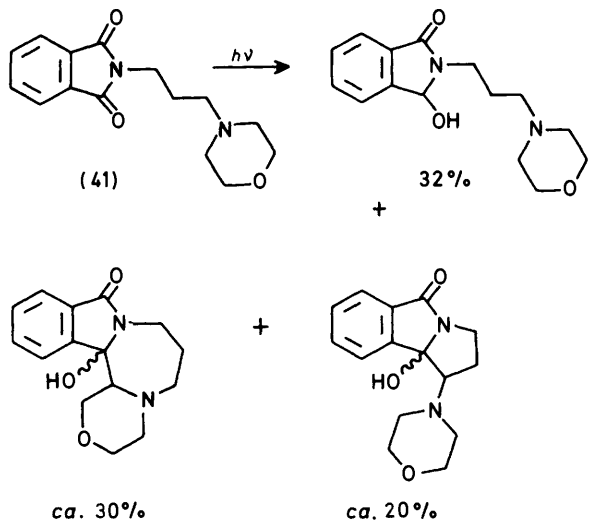
(38) $R^1 = H$; $R^2 = Me$ 31%
(39) $R^1, R^2 = CH_2OCH_2CH_2$ 12%



(40) 11 + 6%

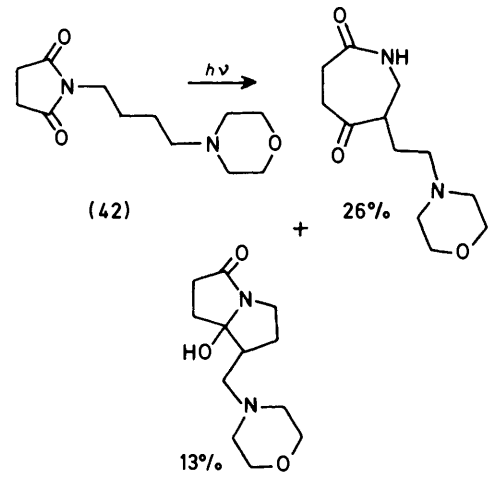
A second product (8%) from the reaction of (39) could not be identified, although it is certainly not a photocyclised product with a new 5-membered or 7-membered ring, nor an azepinedione. For comparison, we irradiated the analogous phthalimide (41), which has not been included in other reports of phthalimide photochemistry.

The formation of the photoreduced imide as the major



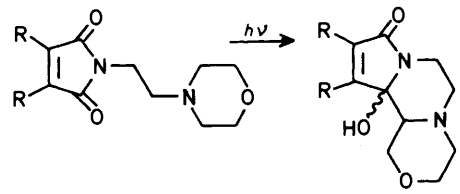
product even in a normally non-reducing solvent such as acetonitrile, suggests that intramolecular reaction for this particular phthalimide is quite slow. The presence of both pyrrolidine and diazepine products indicates that there are some limitations on the application of this photoreaction for making compounds with a perhydro-1,4-diazepine ring,[19] although the use of *N*-acylated derivatives [20] may overcome this. As with some of our other photoproducts, the diastereoisomers of the perhydrodiazepine product from (41) show quite large differences in the carbon-13 chemical shift values for certain of the atoms of the ring system.

Irradiation of the *N*-[4-(morpholin-4-yl)butyl]succinimide (42) gives a perhydroazepinedione and a photocyclised product with a new pyrrolidine ring, with no products isolated in which reaction takes place at either of the positions adjacent to the amine nitrogen. This indicates that in the aliphatic systems there is no strong preference for reaction at a remote but activated position, at least not for compounds with a chain of four methylenes between the two nitrogen atoms. In turn this could mean that the aromatic ring of phthalimides enhances the electron-acceptor properties of the imide ring to such an extent that charge-transfer interactions in the excited state



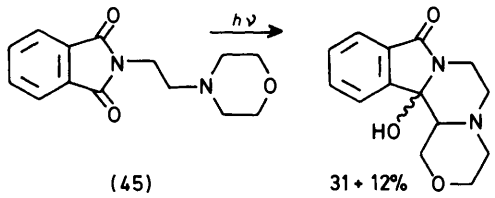
lead to conformations in which the terminal heteroatom and the imide are in close proximity, whereas such interactions are less important for aliphatic imides.

Maleimides.—In order to test this hypothesis we prepared and irradiated two compounds (43) and (44) with a maleimide ring system. The double bond in conjugation with the imide carbonyl groups provides a chromophore more like that of an aromatic imide than of a succinimide, and in confirmation of this the major products of irradiation are compounds with a new fused piperazine ring, analogous to products from similar phthalimides [19] rather than those from succinimides. It follows that a range of medium- and large-ring photoproducts may be accessible by photocyclisation of maleimide derivatives, as it is for phthalimides but not for succinimides.

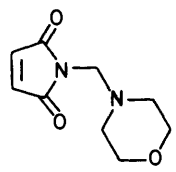


(43) R = H 21 + 13%
(44) R,R = $(CH_2)_4$ 23 + 15%

These are the first examples of maleimides that undergo photochemical hydrogen abstraction and cyclisation to a position in the *N*-substituent, and it contrasts with the reactions at the C=C double bond that dominate so much of maleimide photochemistry. Irradiation of the analogous phthalimide system (45) has not been reported previously, and for completeness we prepared it and isolated the two diastereoisomers of the expected photoproduct.



A maleimide Mannich base (46) was prepared and irradiated, but this proved, unexpectedly, to be photochemically unreactive.



(46)

Experimental

N-(Dialkylaminomethyl) substituted imides and amides were made by Mannich reaction procedures.[21] The glutarimide derivatives were prepared by warming glutarimide with an equimolar mixture of aqueous formaldehyde (40%) and secondary amine without extra solvent. Compounds (6),[7] (8),[7] (18),[22] (22),[14] and (46) [23] have been reported previously. *N*-(Dialkylaminoethyl)imides and higher homologues were prepared by standard methods.[24] 2-Phenylsuccinimide [25] and but-3-enylamine [26] were prepared according to published pro-

cedures. Light petroleum refers to the fraction with b.p. in the range 40—60 °C; ether refers to diethyl ether.

N-(*Dialkylaminomethyl*)*succinimide* (3) was obtained in 74% yield; m.p. 45—47 °C (ether–light petroleum) (Found: C, 53.9; H, 8.1; N, 17.9. $C_7H_{12}N_2O_2$ requires C, 53.8; H, 7.8; N, 17.9%); $\nu_{max.}$ 1 775 and 1 720 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.30 (6 H, s), 2.75 (4 H, s), and 4.34 (2 H, s); δ_C (90 MHz, $CDCl_3$) 28.1, 43.1, 60.8, and 178.0 p.p.m.

N-(*Diethylaminomethyl*)*succinimide* (4) was obtained in 58% yield; m.p. of HCl salt 142—144 °C (Pr^iOH) (Found: C, 58.6; H, 8.9; N, 15.3. $C_9H_{16}N_2O_2$ requires C, 58.7; H, 8.75; N, 15.2%); $\nu_{max.}$ 1 775 and 1 710 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.08 (6 H, t, *J* 7 Hz), 2.63 (4 H, q, *J* 7 Hz), 2.73 (4 H, s), and 4.45 (2 H, s).

N-(*Diallylaminomethyl*)*succinimide* (5) was obtained in 72% yield (Found: C, 63.8; H, 7.65; N, 13.3. $C_{11}H_{16}N_2O_2$ requires C, 63.4; H, 7.7; N, 13.45%); $\nu_{max.}$ 3 090, 1 710, and 1 645sh cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.71 (4 H, s), 3.32 (4 H, d, *J* 6 Hz), 4.41 (2 H, s), 4.95—5.3 (4 H, m), and 5.55—6.2 (2 H, m).

N-(1,2,5,6-*Tetrahydropyridin*-1-*ylmethyl*)*succinimide* (7) was obtained in 47% yield; m.p. 73—75 °C (EtOH) (Found: C, 61.7; H, 7.0; N, 14.2. $C_{10}H_{14}N_2O_2$ requires C, 61.8; H, 7.3; N, 14.4%); $\nu_{max.}$ 3 080, 1 770, and 1 700 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.9—2.3 (2 H, m), 2.55—2.85 (6 H, m), 2.95—3.25 (2 H, m), 4.45 (2 H, s), and 5.63 (2 H, s); δ_C (90 MHz, $CDCl_3$) 26.1, 28.2, 48.0, 49.7, 60.0, 124.9, 125.1, and 178.1 p.p.m.

N-(1,2,3,4-*Tetrahydroisoquinolin*-2-*ylmethyl*)*succinimide* (9) was obtained in 71% yield; m.p. 122—124 °C (EtOH) (Found: C, 69.0; H, 6.4; N, 11.3. $C_{14}H_{16}N_2O_2$ requires C, 68.85; H, 6.6; N, 11.45%); $\nu_{max.}$ 1 775 and 1 710 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.72 (4 H, s), 2.91 (4 H, s), 3.80 (2 H, s), 4.62 (2 H, s), and 7.10 (4 H, s); δ_C (90 MHz, $CDCl_3$) 28.2, 29.3, 49.0, 52.9, 60.0, 125.7, 126.2, 126.7, 128.8, 134.2, 134.5, and 178.1 p.p.m.

N-(*Dimethylaminomethyl*)-2-*phenylsuccinimide* (13) was obtained in 68% yield; m.p. 96—98 °C (EtOH) (Found: C, 67.0; H, 7.0; N, 12.1. $C_{13}H_{16}N_2O_2$ requires C, 67.2; H, 6.9; N, 12.1%); $\nu_{max.}$ 1 770 and 1 695 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.33 (6 H, s), 2.84 (1 H, dd, *J* 18.5 and 5 Hz), 3.25 (1 H, dd, *J* 18.5 and 9 Hz), 4.05 (1 H, dd, *J* 9 and 5 Hz), 4.45 (2 H, s), and 7.2—7.35 (5 H, m); δ_C (90 MHz, $CDCl_3$) 37.1 (t), 43.1 (q), 46.0 (d), 61.2, 127.2, 128.0, 129.2, 137.3, 177.0, and 178.6 p.p.m.

N-(1,2,5,6-*Tetrahydropyridin*-1-*ylmethyl*)*pyrrolidin*-2-*one* (21) was obtained in 63% yield; m.p. of HCl salt 163—165 °C (EtOH) (Found: C, 66.4; H, 9.1; N, 15.5. $C_{10}H_{16}N_2O$ requires C, 66.6; H, 8.95; N, 15.5%); $\nu_{max.}$ 3 033 and 1 680 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.7—2.8 (8 H, m), 3.03 (2 H, s), 3.51 (2 H, t, *J* 6 Hz), 4.05 (2 H, s), and 5.71 (2 H, s); δ_C (90 MHz, $CDCl_3$) 18.1, 26.0, 31.2, 47.6, 47.7, 49.9, 64.3, 125.0, 125.1, and 175.6 p.p.m.

N-(*Piperidin*-1-*ylmethyl*)*glutarimide* (24) was obtained in 49% yield; m.p. 83—85 °C (ether–light petroleum) (Found: C, 62.6; H, 8.5; N, 13.2. $C_{11}H_{18}N_2O_2$ requires C, 62.8; H, 8.6; N, 13.3%); $\nu_{max.}$ 1 726 and 1 667 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.2—2.2 (8 H, m), 2.3—2.9 (8 H, m), and 4.73 (2 H, s).

N-(1,2,5,6-*Tetrahydropyridin*-1-*ylmethyl*)*glutarimide* (25) was obtained in 61% yield; m.p. 71—74 °C (ether) (Found: C, 63.4; H, 7.85; N, 13.4. $C_{11}H_{16}N_2O_2$ requires C, 63.4; H, 7.75; N, 13.45%); $\nu_{max.}$ 3 029, 1 720, and 1 684 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.85—2.4 (4 H, m), 2.55—2.9 (6 H, m), 3.05—3.3 (2 H, m), 4.86 (2 H, s), and 5.70 (2 H, s).

N-(*Morpholin*-4-*ylmethyl*)*glutarimide* (26) was obtained in 42% yield; m.p. 103.5—106.5 °C (ethanol–ether) (Found: C, 56.2; H, 7.4; N, 13.1. $C_{10}H_{16}N_2O_3$ requires C, 56.6; H, 7.6; N, 13.2%); $\nu_{max.}$ 1 726 and 1 668 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.75—2.15 (2 H, m), 2.4—2.85 (8 H, m), 3.55—3.9 (4 H, m), and 4.75 (2 H, s); δ_C (90 MHz, $CDCl_3$) 17.1, 33.0, 51.4, 60.0, 67.0, and 173.4 p.p.m.

N-(1,2,3,4-*Tetrahydroisoquinolin*-2-*ylmethyl*)*glutarimide* (27) was obtained in 67% yield; m.p. 93—95 °C (ethanol–ether) (Found: C, 70.1; H, 7.15; N, 10.85. $C_{15}H_{18}N_2O_2$ requires C, 69.75; H, 7.0; N, 10.85%); $\nu_{max.}$ 1 735 and 1 686 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.85—2.2 (2 H, m), 2.5—3.0 (8 H, m), 3.84 (2 H, s), 4.96 (2 H, s), and 7.08 (4 H, s); δ_C (90 MHz, $CDCl_3$) 17.0, 29.1, 32.9, 49.1, 53.2, 59.8, 125.5, 126.0, 126.6, 128.6, 133.9, 134.8, and 173.4 p.p.m.

3-(*Piperidin*-1-*ylmethyl*)*dihydrouracil* (28) was obtained in 62% yield; m.p. 109—111 °C (EtOH) (Found: C, 56.75; H, 8.0; N, 20.1. $C_{10}H_{17}N_3O_2$ requires C, 56.85; H, 8.1; N, 19.9%); $\nu_{max.}$ 3 225, 1 725, and 1 680 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.3—1.5 (6 H, m), 2.45—2.5 (4 H, m), 2.65 (2 H, t, *J* 7 Hz), 3.55 (2 H, t, *J* 7 Hz), 4.11 (2 H, s), and 8.75 (1 H, br); δ_C (90 MHz, $CDCl_3$) 24.5, 26.0, 31.4, 41.8, 51.9, 69.0, 153.6, and 170.4 p.p.m.

3-(1,2,5,6-*Tetrahydropyridin*-1-*ylmethyl*)*dihydrouracil* (29) was obtained in 69% yield; m.p. 107—109 °C (EtOH) (Found: C, 57.3; H, 7.5; N, 20.0. $C_{10}H_{15}N_3O_2$ requires C, 57.4; H, 7.2; N, 20.1%); $\nu_{max.}$ 3 200, 3 080, 1 730, and 1 675 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.0—2.35 (2 H, m), 2.5—2.8 (4 H, m), 2.95—3.25 (2 H, m), 3.55 (2 H, t, *J* 7 Hz), 4.22 (2 H, s), 5.70 (2 H, s), and 8.8 (1 H, br); δ_C (90 MHz, $CDCl_3$) 26.2, 31.5, 41.9, 47.7, 50.1, 68.2, 125.4, 125.5, 153.8, and 170.6 p.p.m.

3-(*Morpholin*-4-*ylmethyl*)*dihydrouracil* (30) was obtained in 68% yield; m.p. 152.5—154.5 °C (EtOH) (Found: C, 50.6; H, 7.15; N, 19.8. $C_9H_{15}N_3O_3$ requires C, 50.7; H, 7.1; N, 19.7%); $\nu_{max.}$ 3 203, 3 090, 1 730, and 1 680 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.35—2.8 (6 H, m), 3.53 (2 H, t, *J* 7 Hz), 3.65—3.75 (4 H, m), 4.15 (2 H, s), and 8.55 (1 H, br); δ_C (90 MHz, $CDCl_3$) 31.1, 41.6, 50.9, 66.8, 68.5, 153.4, and 170.0 p.p.m.

3-(1,2,3,4-*Tetrahydroisoquinolin*-2-*ylmethyl*)*dihydrouracil* (31) was obtained in 64% yield; m.p. 124—126 °C (EtOH) (Found: C, 64.8; H, 6.45; N, 16.0. $C_{14}H_{17}N_3O_2$ requires C, 64.85; H, 6.6; N, 16.2%); $\nu_{max.}$ 3 200, 3 080, 1 728, and 1 680 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.61 (2 H, t, *J* 7 Hz), 2.88 (4 H, s), 3.55 (2 H, t, *J* 7 Hz), 3.75 (2 H, s), 4.30 (2 H, s), 7.11 (4 H, s), and 8.85 (1 H, br); δ_C (90 MHz, $CDCl_3$) 29.0, 31.1, 41.2, 48.3, 53.1, 67.9, 125.7, 126.3, 126.6, 128.7, 134.0, 134.1, 153.4, and 170.2 p.p.m.

N-[2-(*Dimethylamino*)*ethyl*]*succinimide* (33) was obtained in 51% yield; m.p. of HCl salt 196—198 °C (EtOH) (Found: C, 56.3; H, 8.5; N, 16.5. $C_8H_{14}N_2O_2$ requires C, 56.45; H, 8.3; N, 16.5%); $\nu_{max.}$ 1 770 and 1 700 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.25 (6 H, s), 2.51 (2 H, t, *J* 7 Hz), 2.72 (s, 4 H), and 3.64 (2 H, t, *J* 7 Hz); for hydrochloride salt δ_C (90 MHz, D_2O) 30.8 (t), 36.2 (t), 45.6 (q), 57.2, and 183.5 p.p.m.

N-[2-(*Morpholin*-4-*yl*)*ethyl*]*succinimide* (34) was obtained in 58% yield; m.p. 79—81 °C (Found: C, 56.5; H, 7.7; N, 13.1. $C_{10}H_{16}N_2O_3$ requires C, 56.6; H, 7.6; N, 13.2%); $\nu_{max.}$ 1 770 and 1 700 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.35—2.85 (10 H, m, including singlet at 2.72) and 3.5—3.85 (6 H, m).

N-[2-(*Morpholin*-4-*yl*)*ethyl*]*glutarimide* (35) was obtained in 72% yield; m.p. 80—82 °C (Found: C, 58.2; H, 7.9; N, 12.2. $C_{11}H_{18}N_2O_3$ requires C, 58.4; H, 8.0; N, 12.4%); $\nu_{max.}$ 1 721 and 1 667 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.93 (2 H, quintet, *J* 6 Hz), 2.4—2.75 (10 H, m), 3.6—3.7 (4 H, m), and 3.92 (2 H, t, *J* 7 Hz); δ_C (90 MHz, $CDCl_3$) 17.2, 32.8, 36.3, 53.7, 55.9, 67.0, and 172.4 p.p.m.

N-[2-(*Morpholin*-4-*yl*)*ethyl*]-1,2,3,6-*tetrahydrophthalimide* (36) was obtained in 63% yield (Found: C, 63.6; H, 7.7; N, 10.5. $C_{14}H_{20}N_2O_3$ requires C, 63.6; H, 7.6; N, 10.6%); $\nu_{max.}$ 3 042, 1 772, 1 709, and 1 645sh cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.25—2.65 (10 H, m), 3.0—3.2 (2 H, m), 3.45—3.85 (6 H, m), and 5.8—6.0 (2 H, m); δ_C (90 MHz, $CDCl_3$) 23.5, 35.7, 39.0, 53.4, 55.1, 67.0, 127.6, and 180.0 p.p.m.

N-[3-(*Dimethylamino*)*propyl*]*succinimide* (38) was obtained

in 72% yield; m.p. of HCl salt 193–195 °C (EtOH) (Found: C, 58.6; H, 8.8; N, 15.3. $C_9H_{16}N_2O_2$ requires C, 58.7; H, 8.75; N, 15.2%); $\nu_{max.}$ 1 770 and 1 700 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.5—2.45 (10 H, m, including singlet at 2.19), 2.69 (4 H, s), and 3.57 (2 H, t, *J* 7 Hz); δ_C (90 MHz, $CDCl_3$) 25.7, 28.1, 37.2 (t), 45.4 (q), 57.0, and 177.2 p.p.m.

N-[3-(*Morpholin-4-yl*)*propyl*]*succinimide* (39) was obtained in 78% yield; m.p. 65.5—67.5 °C (Found: C, 58.5; H, 7.9; N, 12.4. $C_{11}H_{18}N_2O_3$ requires C, 58.4; H, 8.0; N, 12.4%); $\nu_{max.}$ 1 760 and 1 690 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.74 (2 H, quintet, *J* 8 Hz), 2.25—2.55 (6 H, m), 2.73 (4 H, s), and 3.5—3.85 (6 H, m); δ_C (90 MHz, $CDCl_3$) 24.3, 28.2, 37.2, 53.6, 56.3, 67.0, and 177.2 p.p.m.

N-[3-(*Morpholin-4-yl*)*propyl*]-3,4,5,6-*tetrahydrophthalimide* (40) was obtained in 55% yield; m.p. of HCl salt 211—214 °C (EtOH) (Found: C, 64.9; H, 8.0; N, 10.0. $C_{15}H_{22}N_2O_3$ requires C, 64.7; H, 8.0; N, 10.1%); $\nu_{max.}$ 1 763, 1 705, and 1 675sh cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.65—1.80 (6 H, m), 2.0—2.4 (10 H, m), and 3.45—3.7 (6 H, m); δ_C (90 MHz, $CDCl_3$) 20.0, 21.4, 25.4, 36.0, 53.7, 56.4, 67.0, 141.5, and 171.2 p.p.m.

N-[3-(*Morpholin-4-yl*)*propyl*]*phthalimide* (41) was obtained in 62% yield; m.p. of HCl salt 238—240 °C (EtOH) (Found: C, 65.7; H, 6.8; N, 10.3. $C_{15}H_{18}N_2O_3$ requires C, 65.7; H, 6.6; N, 10.2%); $\nu_{max.}$ 1 770 and 1 710 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.86 (2 H, quintet, *J* 7 Hz), 2.3—2.5 (6 H, m), 3.45—3.55 (4 H, m), 3.78 (2 H, t, *J* 7 Hz), and 7.65—7.9 (4 H, m); δ_C (90 MHz, $CDCl_3$) 24.8, 36.6, 53.6, 56.5, 66.8, 123.1, 132.4, 133.9, and 168.4 p.p.m.

N-[4-(*Morpholin-4-yl*)*butyl*]*succinimide* (42) was obtained in 56% yield (Found: C, 59.9; H, 8.3; N, 11.6. $C_{12}H_{20}N_2O_3$ requires C, 60.0; H, 8.4; N, 11.65%); $\nu_{max.}$ 1 772 and 1 700 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.5—1.6 (4 H, m), 2.25—2.45 (6 H, m), 2.7 (4 H, s), 3.53 (2 H, t, *J* 7 Hz), and 3.65—3.75 (4 H, m); δ_C (90 MHz, $CDCl_3$) 23.8, 25.6, 28.1, 38.6, 53.7, 58.3, 66.9, and 177.2 p.p.m.

N-[2-(*Morpholin-4-yl*)*ethyl*]*maleimide* (43) was obtained in 31% yield (Found: C, 56.9; H, 6.6; N, 13.2%. $C_{10}H_{14}N_2O_3$ requires C, 57.1; H, 6.7; N, 13.3%); $\nu_{max.}$ 3 095, 1 768, 1 710, and 1 678sh cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.35—2.7 (6 H, m), 3.45—3.65 (6 H, m), and 6.71 (2 H, s); δ_C (90 MHz, $CDCl_3$) 34.8, 53.4, 55.9, 66.9, 134.1, and 170.7 p.p.m.

N-[2-(*Morpholin-4-yl*)*ethyl*]-3,4,5,6-*tetrahydrophthalimide* (44) was obtained in 61% yield; m.p. of HCl salt 226.5—228.5 °C (EtOH) (Found: C, 63.4; H, 7.8; N, 10.7. $C_{14}H_{20}N_2O_3$ requires C, 63.6; H, 7.6; N, 10.6%); $\nu_{max.}$ 1 765, 1 710, and 1 675sh cm^{-1}; δ_H (60 MHz, $CDCl_3$) 1.6—1.95 (4 H, m), 2.25—2.75 (10 H, m), and 3.45—3.85 (6 H, m); δ_C (90 MHz, $CDCl_3$) 19.9, 21.4, 34.5, 53.5, 56.4, 66.9, 141.4, and 171.0 p.p.m.

N-[2-(*Morpholin-4-yl*)*ethyl*]*phthalimide* (45) was obtained in 77% yield; m.p. 129—131 °C (EtOH) (Found: C, 64.8; H, 6.3; N, 10.8. $C_{14}H_{16}N_2O_3$ requires C, 64.6; H, 6.2; N, 10.8%); $\nu_{max.}$ 1 765 and 1 710 cm^{-1}; δ_H (60 MHz, $CDCl_3$) 2.45—2.7 (6 H, m, including 2.63, t, *J* 7 Hz), 3.6—3.9 (6 H, m, including 3.83, t, *J* 7 Hz), and 7.65—7.85 (4 H, m); δ_C (90 MHz, $CDCl_3$) 35.0, 53.5, 56.1, 67.0, 123.2, 132.2, 133.9, and 168.3.

N-(Morpholin-4-ylmethyl)maleimide (46) was prepared by way of *N*-(hydroxymethyl)maleimide [23] in 46% overall yield; m.p. 142.5—144.5 °C (lit.,[23] 143—145 °C); $\nu_{max.}$ 3 140, 1 770, and 1 710 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.6 (4 H, m), 3.7 (4 H, m), 4.43 (2 H, s), and 6.79 (2 H, s); δ_C (90 MHz, $CDCl_3$) 50.7, 59.3, 66.8, 134.3, and 171.6 p.p.m.

Irradiations were carried out with a 400-W medium-pressure mercury arc with quartz filter (Pyrex for phthalimides), using *ca.* 0.01 mol of the imide in acetonitrile solvent. After removal of the solvent, the product mixture was separated by silica-gel chromatography with chloroform, chloroform–acetone, or chloroform–methanol as eluant. The photoproducts from (4), (6), (7), (8), (9), and (34) were described in our preliminary report.[6]

Irradiation of (3) gave 5-*hydroxy*-3-*methyl*-1,3-*diazabicyclo*[3.3.0]*octan*-8-*one* (52%); $\nu_{max.}$ 3 350 and 1 690 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.1—2.5 (7 H, m, incl. 2.41, s), 2.65 (1 H, d, *J* 9 Hz), 2.96 (1 H, d, *J* 9 Hz), 3.9 (1 H, v.br), and 3.97 (3 H, s, becomes an AB pattern at low temperature, *e.g.* 220 K); δ_C (90 MHz, $CDCl_3$) 33.0, 33.1, 39.6, 66.0, 66.3, 95.8, and 176.8 p.p.m. (M^+, *m/z* 156.0900. $C_7H_{12}N_2O_2$ requires *M*, 156.0894).

Irradiation of (5) gave 3-*allyl*-5-*hydroxy*-4-*vinyl*-1,3-*diazabicyclo*[3.3.0]*octan*-8-*one*. First isomer (26%); $\nu_{max.}$ 3 350, 3 085, 1 710, and 1 645 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.05—3.7 (8 H, m), 3.83 (1 H, d, *J* 6 Hz), 4.28 (1 H, d, *J* 6 Hz), and 5.1—6.0 (6 H, m); δ_C (90 MHz, $CDCl_3$) 30.7, 32.9, 54.2, 63.4, 74.9, 96.0, 118.8, 121.7, 132.5, 133.2, and 175.6 p.p.m. Second isomer (20%); $\nu_{max.}$ 3 380, 3 080, 1 705, and 1 645 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.85—3.3 (7 H, m), 3.45 (1 H, d, *J* 7.5 Hz), 4.45 (1 H, d, *J* 7.5 Hz), and 4.9—6.15 (7 H, m); δ_C (90 MHz, $CDCl_3$) 31.2, 31.9, 53.7, 63.2, 75.1, 97.8, 117.9, 120.9, 132.8, 134.4, and 178.1 p.p.m.

Irradiation of (13) gave 5-*hydroxy*-3-*methyl*-6-*phenyl*-1,3-*diazabicyclo*[3.3.0]*octan*-8-*one* (14) and (15) and 5-*hydroxy*-3-*methyl*-7-*phenyl*-1,3-*diazabicyclo*[3.3.0]*octan*-8-*one* (16) and (17). Compound (14) was isolated as a colourless oil (34%); $\nu_{max.}$ 3 330 and 1 700 cm^{-1}; δ_H (400 MHz, $CDCl_3$) 1.87 (1 H, d, *J* 9.5 Hz), 2.44 (3 H, s), 2.55 (1 H, d, *J*, 9.5 Hz), 2.62 (1 H, dd, *J* 17 and 2.5 Hz), 3.44 (1 H, dd, *J* 17 and 8.5 Hz), 3.70 (1 H, dd, *J* 8.5 and 2.5 Hz), 3.73 (1 H, d, *J* 6 Hz), 4.15, (1 H, d, *J* 6 Hz), 4.65 (1 H, br), and 7.1—7.4 (5 H, m); δ_C (90 MHz, $CDCl_3$) 39.4 (t), 39.8 (q), 48.8 (d), 62.4 (t), 66.2 (t), 99.0 (s), 127.4, 127.5, 128.9, 139.7, and 175.3 p.p.m. A mixture (51%) of the other three isomers was separated by repeated column chromatography. Compound (15) was a colourless oil; $\nu_{max.}$ 3 335 and 1 700 cm^{-1}; δ_H (400 MHz, $CDCl_3$) 2.42, (3 H, s), 2.63 (1 H, dd, *J* 16 and 7.5 Hz), 2.83 (1 H, d, *J* 9 Hz), 3.01 (1 H, d, *J* 9 Hz), 3.30 (1 H, dd, *J* 16 and 12.5 Hz), 3.66 (1 H, dd, *J* 12.5 and 7.5 Hz), 3.89 (1 H, d, *J* 6.5 Hz), 4.16 (1 H, d, *J* 6.5 Hz), and 7.3—7.4 (5 H, m); δ_C (90 MHz, $CDCl_3$) 37.8 (t), 39.5 (q), 51.1 (d), 66.1 (t), 66.3 (t), 95.9 (s), 126.3, 127.8, 128.4, 128.6, 128.9, 136.0, and 175.1 p.p.m. (M^+, *m/z* 232.1197. $C_{13}H_{16}N_2O_2$ requires *M*, 232.1206). Compound (16) was a colourless oil; $\nu_{max.}$ 3 335 and 1 698 cm^{-1}; δ_H (400 MHz $CDCl_3$) 2.40 (3 H, s), 2.39 (1 H, dd, *J* 14 and 2.5 Hz), 2.53 (1 H, d, *J* 9.5 Hz), 2.71 (1 H, dd, *J* 14 and 10.5 Hz), 3.01 (1 H, d, *J* 9.5 Hz), 3.80 (1 H, dd, *J* 10.5 and 2.5 Hz), 3.95 (1 H, d, *J* 6.5 Hz), 4.06 (1 H, d, *J* 6.5 Hz), and 7.25—7.5 (5 H, m); δ_C (90 MHz, $CDCl_3$) 38.8 (t), 39.7 (q), 51.5 (d), 66.3 (t), 66.4 (t), 94.8 (s), 127.2, 127.6, 128.4, 128.8, 130.9, 139.2, and 176.1 p.p.m. Compound (17) was not obtained pure, but its carbon-13 n.m.r. spectrum was identified from that of a mixture containing it: δ_C (90 MHz, $CDCl_3$) 37.8, 39.5, 49.8, 66.3, 66.7, 92.9, 127.3, 128.4, 128.7, 130.7, 130.9, 137.4, and 174.8 p.p.m.

Irradiation of (18) gave a stereoisomer of 2-*hydroxy*-3-*phenyl*-11-*oxa*-6,9-*diazatricyclo*[6.4.0.$0^{2,6}$]*dodecan*-5-*one* (19) as the major product (20%); $\nu_{max.}$ 3 345 and 1 700 cm^{-1}; δ_H (220 MHz, $CDCl_3$) 2.19 (1 H, t, *J* 11 Hz), 2.59 (1 H, t, *J* 4 Hz), 2.51 and 2.65 (1 H, two d, *J* 3 Hz), 2.75—2.85 (2 H, m), 3.15 (1 H, td, *J* 12 and 2.5 Hz), 3.29 (1 H, dd, *J* 11 and 3 Hz), 3.40 (1 H, t, *J* 9 Hz), 3.55 (1 H, dd, *J* 12 and 4 Hz), 3.68 (1 H, d, *J* 7 Hz), 3.79 (1 H, dd, *J* 9 and 3 Hz), 4.65 (1 H, br), 4.78 (1 H, d, *J* 7 Hz), and 7.25—7.45 (5 H, m); δ_C (90 MHz, $CDCl_3$) 39.0, 48.1, 48.5, 62.1, 62.9, 63.9, 69.5, 99.0, 127.7, 127.9, 128.6, 138.3, and 176.7 p.p.m.; *m/z* 274 (M^+), 256, and 99 (base) (M^+, *m/z* 274.1318. $C_{15}H_{18}N_2O_3$ requires *M*, 274.1317).

Irradiation of (21) gave N-(*but*-3-*enylaminomethyl*)*pyrroli-*

din-2-one (23) as the only product (5%) that could be isolated, $\nu_{max.}$ 3 225, 3 110, and 1 683 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.8—2.95 (9 H, m), 3.44 (2 H, t, *J* 6 Hz), 4.24 (2 H, s), 4.8—5.3 (2 H, m), and 5.5—6.15 (1 H, m); δ_C (90 MHz, $CDCl_3$) 18.0 (t), 31.3 (t), 34.1 (t), 45.6 (t), 46.6 (t), 57.3 (t), 116.5 (t), 136.2 (d), and 175.7 (s); *m/z* 127, 113, 98 (base), 84, and 70. Compound (23) could not be synthesised from pyrrolidin-2-one, formaldehyde, and but-3-enylamine, despite many attempts which included amine exchange. Instead it was converted by reaction with formaldehyde and pyrrolidin-2-one into N,N-*bis*(2-*oxopyrrolidin*-1-*ylmethyl*)*but*-3-*enylamine* (24), which was synthesised independently (46%); $\nu_{max.}$ 3 070, 1 690, and 1 650sh cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.7—2.9 (12 H, m), 3.47 (4 H, t, *J* 6.5 Hz), 4.20 (4 H, s), 4.85—5.25 (2 H, m), and 5.5—6.15 (1 H, m); δ_C (90 MHz, $CDCl_3$) 18.0, 31.3, 32.2, 47.0, 49.7, 60.4, 115.9, 136.6, and 175.8 p.p.m. (M^+, *m/z* 265.1785. $C_{14}H_{23}N_3O_2$ requires M^+, 265.1789).

Irradiation of (24) gave glutarimide (85%) and very small amounts (0.6 and 0.4%) of compounds believed to be 2-hydroxy-7,9-diazatricyclo[7.4.0.$0^{2,7}$]tridecan-6-one. First isomer: δ_H (400 MHz, $CDCl_3$) 1.15—1.7 (5 H, m), 1.75—1.9 (2 H, m), 1.95—2.0 (1 H, m), 2.0—2.05 (2 H, m), 2.05—2.3 (2 H, m), 2.5—2.6 (2 H, m), 3.10 (1 H, td, *J* 10.5 Hz), 3.2 (1 H, br), 3.63 (1 H, d, *J* 6 Hz), and 4.49 (1 H, d, *J* 6 Hz); δ_C (400 MHz, $CDCl_3$) 16.7, 22.6, 24.1, 24.7, 29.6, 31.0, 49.8, 66.6, 71.4, 87.6, and 168.8. Second isomer: δ_H (400 MHz, $CDCl_3$) 1.2—1.65 (3 H, m), 1.75—2.05 (3 H, m), 2.15—2.4 (3 H, m), 2.5—2.6 (2 H, m), 2.74 (1 H, dt, *J* 13 and 3.5 Hz), 2.90 (1 H, dd, *J* 12 and 3 Hz), 3.0—3.05 (1 H, m), 4.10 (1 H, dd, *J* 16.5 and 6 Hz), 4.12 (1 H, d, *J* 6.5 Hz), and 4.49 (1 H, d, *J* 6.5 Hz); δ_C (400 MHz, $CDCl_3$) 16.3, 20.0, 22.0, 23.1, 27.7, 30.6, 46.3, 62.1, 69.9, 90.0, and 169.3 p.p.m.

Irradiation of (25) gave glutarimide (50%) and 2-*hydroxy*-7,9-*diazatricyclo*[7.4.0.$0^{2,7}$]*tridec*-12-*en*-6-*one*. First isomer (30%); m.p. 99—101 °C (Found: C, 63.6; H, 7.9; N, 13.45. $C_{11}H_{16}N_2O_2$ requires C, 63.45; H, 7.75; N, 13.45%); $\nu_{max.}$ 3 340, 3 045, and 1 635 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.3—3.1 (10 H, m), 3.81 (1 H, s), 4.32 (1 H, d, *J* 8 Hz), 4.58 (1 H, d, *J* 8 Hz), 5.2 (1 H, br), and 5.5—6.1 (2 H, m); δ_C (90 MHz, $CDCl_3$) 16.8, 21.4, 30.3, 31.2, 43.1, 63.9, 69.2, 88.5, 122.5, 128.1, and 169.5 p.p.m. (M^+, *m/z* 208.1203. $C_{11}H_{16}N_2O_2$ requires *M*, 208.1211). Second isomer (8%); $\nu_{max.}$ 3 350, 3 045, and 1 640 cm^{-1} (Found: C, 63.65; H, 7.9; N, 13.45. $C_{11}H_{16}N_2O_2$ requires C, 63.45; H, 7.5; N, 13.45%); δ_H (90 MHz, $CDCl_3$) 1.7—2.9 (10 H, m), 3.0—3.3 (2 H, m), 3.97 (1 H, d, *J* 7 Hz), 4.52 (1 H, d, *J* 7 Hz), and 5.5—6.2 (2 H, m); δ_C (90 MHz, $CDCl_2$) 16.6, 26.0, 30.3, 31.1, 45.3, 66.1, 68.9, 87.3, 121.2, 129.9, and 169.2.

Irradiation of (26) gave glutarimide (53%) and small amounts (8% and 4%) of compounds believed to be 1-hydroxy-4-oxa-7,9-diazatricyclo[7.4.0.$0^{2,7}$]tridecan-10-one; $\nu_{max.}$ 3 200 and 1 620; 3 325 and 1 615 cm^{-1}; δ_H (90 MHz, $CDCl_3$) includes 4.54 (d, *J* 6 Hz), and 4.22 (d, *J* 8 Hz) and 4.70 (d, *J* 8 Hz).

Irradiation of (27) gave 17-*hydroxy*-10,12-*diazatetracyclo*-[8.7.0.$0^{2,7}$.$0^{12,17}$]*heptadeca*-2,4,6-*trien*-13-*one*. First isomer (17%); m.p. 138.5—140 °C (Found: C, 69.7; H, 6.95; N, 10.7. $C_{15}H_{18}N_2O_2$ requires C, 69.75; H, 7.0; N, 10.85%); $\nu_{max.}$ 3 170 and 1 605 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.7—3.5 (11 H, m), 3.57 (1 H, s), 4.16 (1 H, d, *J* 7.5 Hz), 4.56 (1 H, d, *J* 7.5 Hz), and 7.21 (4 H, s); δ_C (90 MHz, $CDCl_3$), 16.7, 29.3, 30.7, 32.9, 46.4, 66.7, 70.8, 88.7, 125.6, 126.0, 127.4, 129.3, 131.2, 136.0, and 169.6. Second isomer (19%); m.p. 150—152 °C (Found: C, 69.7; H, 7.1; N, 10.8. $C_{15}H_{18}N_2O_2$ requires C, 69.75; H, 7.0; H, 10.85%); $\nu_{max.}$ 3 340 and 1 620 dm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.25—3.0 (10 H, m), 4.38 (1 H, d, *J* 10.5 Hz), 4.50 (1 H, s), 4.99 (1 H, d, *J* 10.5 Hz), 5.4 (1 H, br), and 7.1—7.25 (4 H, m); δ_C (90 MHz, $CDCl_3$) 16.6, 28.6, 29.6, 33.5, 45.1, 67.1, 73.0, 89.5, 126.0, 127.1, 127.7, 128.6, 131.0, 133.8, and 170.4 p.p.m.

Irradiation of (33) gave 6-*dimethylaminoperhydroazepine*-2,5-*dione* (17%); $\nu_{max.}$ 3 220, 1 701, and 1 660 cm^{-1}; δ_H (220 MHz, $CDCl_3$) 2.25—3.1 (11 H, m, including singlet at 2.29), 3.36 (1 H, dd, *J* 15.5 and 5 Hz), 3.53 (1 H, td, *J* 15.5 and 6 Hz), and 7.56 (1 H, br); δ_C (90 MHz, $CDCl_3$) 31.7, 36.3, 41.6, 42.9, 75.7, 176.6, and 210.5; *m/z* 170 (M^+), 126, and 71 (base) (M^+, *m/z* 170.1047. $C_8H_{14}N_2O_2$ requires *M*, 170. 1055).

Irradiation of (34) gave 6-(*morpholin*-4-*yl*)*perhydroazepine*-2,5-*dione* (46%) (Found: C, 56.7; H, 7.5; N, 13.3. $C_{10}H_{16}N_2O_3$ requires C, 56.6; H, 7.6; N, 13.2%); $\nu_{max.}$ 3 320, 3 200, 1 710, and 1 680 cm^{-1}; δ_H (220 MHz, $CDCl_3$) 2.25—2.75 (7 H, m), 2.85—3.1 (2 H, m), 3.36 (1 H, dd, *J* 16 and 5 Hz), 3.59 (1 H, td, *J* 16 and 6 Hz), 3.75 (4 H, t, *J* 5 Hz), and 7.6 (1 H, br); δ_C (90 MHz, $CDCl_3$) 31.6, 36.3, 40.7, 50.9, 66.7, 74.9, 176.3, and 220.5 p.p.m.

Irradiation of (35) gave 7-(*morpholin*-4-*yl*)*perhydroazocine*-2,5-*dione* (77%); $\nu_{max.}$ 3 120, 3 090, 1 710, and 1 665 cm^{-1}; δ_H (90 MHz, $CDCl_3$), 1.7—2.75 (10 H, m), 2.85—3.4 (2 H, m), 3.45—3.8 (5 H, m), and 6.77 (1 H, br); δ_C (90 MHz, C_5D_5N) 23.6, 32.9, 36.4, 41.2, 52.0, 66.9, 79.3, 175.3, and 213.0 p.p.m; *m/z* 198, 170, 141, 127, and 113 (base).

Irradiation of (36) gave 5-(*morpholin*-4-*yl*)-3-*azabicyclo*-[5.4.0]*undec*-9-*ene*-2,6-*dione* (74%); $\nu_{max.}$ 3 295, 3 220, 3 030, 1 710, 1 664, and 1 645sh cm^{-1}; δ_H (220 MHz, $CDCl_3$) 2.2—2.45 (6 H, m), 2.65—2.75 (2 H, m), 3.05—3.2 (2 H, m), 3.35—3.65 (3 H, m), 3.76 (4 H, t, *J* 4.5 Hz), 5.77 (2 H, s), 6.9 (1 H, br); δ_C (90 MHz, $CDCl_3$) 25.0 (t), 25.5 (t), 40.4 (t), 41.4 (d), 50.8 (t), 66.8 (t), 74.0 (d), 124.8 (d), 125.2 (d), 175.7, and 210.0 p.p.m.; *m/z* 264 (M^+), 236 (base), and 178 (M^+, *m/z* 264.1475. $C_{14}H_{20}N_2O_3$ requires *M*, 264.1473).

Irradiation of (38) gave 7-*hydroxy*-5-*methyl*-1,5-*diaza*-*bicyclo*[5.3.0]*decan*-10-*one* (31%); $\nu_{max.}$ 3 340 and 1 675 cm^{-1}; δ_H (220 MHz, $CDCl_3$) 1.7—2.15 (4 H, m), 2.3—2.4 (2 H, m), 2.45—2.6 (4 H, m, including singlet at 2.51), 2.65—2.8 (3 H, m), 2.9—3.0 (1 H, m), and 3.50 (2 H, t, *J* 5.5 Hz); δ_C (90 MHz, $CDCl_3$) 27.1 (t), 29.5 (t), 31.5 (t), 39.0 (t), 47.6 (q), 58.7 (t), 65.0 (t), 89.3 (s), and 174.4 p.p.m.; *m/z* 184 (M^+), 166, and 58 (base) (M^+, *m/z* 184.1233. $C_9H_{16}N_2O_2$ requires *M*, 184.1211).

Irradiation of (39) gave 2-*hydroxy*-12-*oxa*-6,9-*diazatri*-*cyclo*[7.4.0.$0^{2,6}$]*tridecan*-5-*one* (12%); $\nu_{max.}$ 3 335 and 1 685 cm^{-1}; δ_H (90 MHz, C_5D_5N) 1.8—3.4 (12 H, m) and 3.4—4.45 (6 H, m); δ_C (90 MHz, C_5D_5N), 25.8, 29.2, 30.3, 36.0, 51.8, 57.2, 65.5, 66.3, 66.7, 92.4, and 174.4. p.p.m.

Irradiation of (40) gave 1-*hydroxy*-4-*oxa*-7,11-*diazatetra*-*cyclo*[9.7.0.$0^{2,7}$.$0^{13,18}$]*octadec*-13(18)-*en*-12-*one*. First isomer (11%); $\nu_{max.}$ 3 310, 1 700, and 1 665sh cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.2—2.8 (16 H, m), 2.85—3.2 (2 H, m), and 3.4—4.05 (4 H, m); δ_C (90 MHz, $CDCl_3$) 20.5, 21.6, 22.7, 24.3, 25.3, 39.4, 52.7, 58.8, 66.5, 66.7, 68.5, 92.5, 134.3, 153.8, and 170.0 p.p.m.; *m/z* 278 (M^+), 260, and 100 (base) (M^+, *m/z* 278.1627. $C_{15}H_{22}N_2O_3$ requires *M*, 278.1629). Second isomer (6%); $\nu_{max.}$ 3 315, 1 700, and 1 665sh cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.2—2.35 (14 H, m), and 2.8—3.9 (8 H, m); δ_C (90 MHz, $CDCl_3$) 20.0, 21.2, 21.7, 22.2, 25.4, 41.3, 50.6, 54.5, 60.1, 61.1, 63.5, 90.4, 133.0, 153.8, and 170.9 p.p.m.; *m/z* 278 (M^+), 260, and 100 (base) (M^+, *m/z* 278.1631. $C_{15}H_{22}N_2O_3$ requires *M*, 278.1629).

Irradiation of (41) gave two pure products. 3-*Hydroxy*-2-[3-(*morpholin*-4-*yl*)*propyl*]*isoindolin*-1-*one* (32%); $\nu_{max.}$ 3 330 and 1 690 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.15—2.65 (8 H, m), 4.1—4.7 (7 H, m, reduced to 6 H with D_2O), 5.76 (1 H, s), and 7.4—7.85 (4 H, m); δ_C (90 MHz, $CDCl_3$) 23.5, 39.5, 52.9, 56.6, 66.0, 83.2, 122.8, 123.1, 129.3, 131.5, 132.1, 145.3, and 167.5 p.p.m.; *m/z* 276 (M^+) and 100 (base) (M^+, *m/z* 276.1473. $C_{15}H_{20}N_2O_3$ requires *M*, 276.1473). 2-*Hydroxy*-3-

morpholin-4-yl-6-azatricyclo[6.4.0.0^{2,6}]*dodeca-8,10,12-trien-7-one* (10%); m.p. 194—196 °C (EtOH) (Found: C, 65.85; H, 6.6; N, 10.1. $C_{15}H_{18}N_2O_3$ requires C, 65.7; H, 6.6; N, 10.2%); $\nu_{max.}$ 3 260 and 1 680 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.2—3.0 (7 H, m), 3.25—3.7 (3 H, m), 3.80 (4 H, t, *J* 4.5 Hz), and 7.35—7.75 (4 H, m); δ_C (90 MHz, $CDCl_3$) 29.8 (t), 39.4 (t), 52.6 (t), 67.1 (t), 69.2 (d), 94.6, 123.7, 129.7, 132.5, 146.9, and 170.0; *m/z* 274 (*M*$^+$), 188, and 126 (base) (*M*$^+$, *m/z* 274.1318. $C_{15}H_{18}N_2O_3$ requires *M*, 274.1317). Mixtures (18 and 21%) were obtained of what was believed to be a diastereoisomer of this compound with two diastereoisomers of 1-hydroxy-7,11-diaza-4-oxatetracyclo[9.7.0.0^{2,7}.0^{13,18}]octadeca-13,15,17-trien-12-one, on the basis of characteristic δ_C values for quaternary C(OH) (90.4, 91.7, and 99.3 p.p.m.) and other features of the carbon-13 n.m.r. spectrum.

Irradiation of (42) gave two products. 6-[2-(*Morpholin-4-yl*)*ethyl*]*perhydroazepine-2,5-dione* (26%); $\nu_{max.}$ 3 210, 3 080, 1 700, and 1 670 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.4—2.05 (2 H, m), 2.25—2.7 (11 H, m), 3.2—3.45 (2 H, m), 3.3—3.75 (4 H, m), and 7.35 (1 H, br); δ_C (90 MHz, $CDCl_3$) 25.4 (t), 30.9 (t), 38.0 (t), 43.0 (t), 52.2 (d), 53.6 (t), 56.0 (t), 66.8 (t), 176.3, and 209.6. p.p.m. (*M*$^+$, *m/z* 240.1470. $C_{12}H_{20}N_2O_3$ requires *M*, 240.1467).

5-*Hydroxy*-4-(*morpholin-4-ylmethyl*)-1-*azabicyclo*[3.3.0]-*octan-8-one* (13%); $\nu_{max.}$ 3 345 and 1 695 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.95—2.25 (4 H, m), 2.35—2.75 (8 H, m), 2.8—3.4 (4 H, m), and 3.6—3.8 (4 H, m); δ_C (90 MHz, $CDCl_3$) 30.5 (t), 33.5 (t), 39.8 (t), 44.6 (d), 54.3 (t), 58.0 (t), 66.9 (t), 97.7 (s), and 174.5 p.p.m.

Irradiation of (43) gave 2-*hydroxy*-12-*oxa*-6,9-*diazatricyclo*[7.4.0.0^{2,6}]*tridec-3-en-5-one*. First isomer (21%), $\nu_{max.}$ 3 360 and 1 690 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.45—2.7 (5 H, m), 3.65—3.9 (7 H, m), 6.22 (1 H, d, *J* 6 Hz), and 6.90 (1 H, d, *J* 6 Hz); δ_C (90 MHz, $CDCl_3$) 35.4, 53.9, 54.5, 66.5, 66.9, 67.0, 86.5, 129.7, 145.2, and 166.4 p.p.m.; *m/z* 310 (*M*$^+$) and 100 (base) (*M*$^+$, *m/z* 210.1003. $C_{10}H_{14}N_2O_3$ requires *M*$^+$, 210.1004). Second isomer (13%), $\nu_{max.}$ 3 440 and 1 700 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.45—2.85 (2 H, m), 2.95—3.4 (6 H, m), 3.5—4.1 (4 H, m), 6.14 (1 H, d, *J* 6 Hz), and 6.85 (1 H, d, *J* 6 Hz); δ_C (90 MHz, $CDCl_3$) 34.6, 44.3, 53.3, 60.8, 62.6, 66.9, 87.1, 128.6, 145.9, and 167.4; *m/z* 210 (*M*$^+$), 192, and 98 (base) (*M*$^+$, *m/z* 210.1005. $C_{10}H_{14}N_2O_3$ requires *M*, 210.1004).

Irradiation of (44) gave 1-*hydroxy*-4-*oxa*-7,10-*diazatetracyclo*[8.7.0.0^{2,7}.0^{12,17}]*heptadec*-12(17)-*en*-11-*one*. First isomer (23%), $\nu_{max.}$ 3 320, 1 690, and 1 660sh cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.25—2.5 (11 H, m), 2.6—2.9 (2 H, m), and 3.0—4.2 (7 H, m); δ_C (90 MHz, $CDCl_3$) 20.2, 21.5, 22.3, 23.2, 35.5, 54.2, 54.3, 66.5, 66.7, 66.8, 85.9, 133.9, 153.3, and 167.6 p.p.m.; *m/z* 264 (*M*$^+$), 246, and 100 (base) (*M*$^+$, *m/z* 264.1475. $C_{14}H_{20}N_2O_3$ requires *M*, 264.1473). Second isomer (15%), $\nu_{max.}$ 3 320, 1 695, and 1 665sh cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.4—1.85 (4 H, m), 1.95—3.5 (13 H, m), and 3.6—4.55 (3 H, m); δ_C (90 MHz, $CDCl_3$) 20.0, 21.2, 21.6, 21.7, 34.4, 43.3, 53.3, 58.8, 60.0, 61.2, 85.5, 134.1, 151.8, and 167.4 p.p.m.; *m/z* 264 (*M*$^+$), 246, and 100 (base) (*M*$^+$, *m/z* 264.1471. $C_{14}H_{20}N_2O_3$ requires *M*, 264.1473).

Irradiation of (45) gave 1-*hydroxy*-4-*oxa*-7,10-*diazatetracyclo*[8.7.0.0^{2,7}.0^{12,17}.]*heptadeca*-12,14,16-*trien*-11-*one*. First isomer (31%), $\nu_{max.}$ 3 320 and 1 700 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 1.85—2.8 (5 H, m), 3.1—4.2 (6 H, m), 4.9 (1 H, br), and 7.3—7.5 (4 H, m); δ_C (90 MHz, $CDCl_3$) 35.5, 54.1, 54.4, 66.2, 66.5, 67.2, 85.4, 123.2, 123.7, 129.8, 131.5, 131.9, 144.6, and 164.8; *m/z* 260 (*M*$^+$), 242, and 100 (base) (*M*$^+$, *m/z* 260.1158. $C_{14}H_{16}N_2O_3$ requires *M*, 260.1160). Second isomer (12%), $\nu_{max.}$ 3 300 and 1 700 cm^{-1}; δ_H (90 MHz, $CDCl_3$) 2.35—3.35 (8 H, m), 3.4—4.15 (4 H, m), and 7.35—7.75 (4 H, m); δ_C (90 MHz, $CDCl_3$) 34.8, 43.1, 53.1, 59.2, 59.3, 62.2, 85.3, 122.5, 123.7, 130.1, 131.5, 132.3, 143.7, and 164.9; *m/z* (deuteriated) 261 (*M*$^+$) and 100 (base) (*M*$^+$, *m/z* 261.1232. $C_{14}H_{15}DN_2O_3$ requires *M*, 261.1222).

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References

1 For a review, see P. H. Mazzocchi, in A. Padwa (ed.), 'Organic Photochemistry,' Marcel Dekker, New York, 1981, vol. 5, p. 421.
2 Y. Kanaoka, *Acc. Chem. Res.*, 1978, **11**, 407; J. D. Coyle, in W. H. Horspool (ed.), 'Synthetic Organic Photochemistry,' Plenum Press, London, 1983, ch. 7.
3 M. Wada, H. Nakai, Y. Sato, and Y. Kanaoka, *Tetrahedron Lett.*, 1982, **23**, 3077, and references therein.
4 Y. Kanaoka and Y. Hatanaka, *J. Org. Chem.*, 1976, **41**, 400; Y. Kanaoka, H. Okajima, Y. Hatanaka, and M. Terashima, *Heterocycles*, 1978, **11**, 455.
5 H. Nakai, Y. Sato, T. Mizoguchi, M. Yamazaki, and Y. Kanaoka, *Heterocycles*, 1977, **8**, 345.
6 For a preliminary report of some of the succinimide systems, see L. R. B. Bryant and J. D. Coyle, *J. Chem. Res. (S)*, 1982, 164.
7 H. Hellmann and I. Löschmann, *Ber.*, 1954, **87**, 1684.
8 H. P. Hamlow, S. Okuda, and N. Nabagawa, *Tetrahedron Lett.*, 1964, 2553; F. Bohlmann, D. Schumann, and H. Schulz, *ibid.*, 1965, 173.
9 R. J. Abraham and P. Loftus, 'Proton and Carbon-13 NMR Spectroscopy,' Heyden, London, 1978, p. 46.
10 J. B. Stothers, 'Carbon-13 Nuclear Magnetic Resonance Spectroscopy,' Academic Press, London and New York, 1972, p. 166.
11 J. F. Challiner, J. D. Coyle, and E. J. Haws, unpublished results.
12 P. H. Mazzocchi, J. Thomas, and F. Danisi, *J. Org. Chem.*, 1979, **44**, 50.
13 W. Werner and H. Fritzsche, *Arch. Pharm. (Weinheim)*, 1969, **302**, 188.
14 C. C. Bombardieri and A. Taurins, *Can. J. Chem.*, 1955, **33**, 923.
15 J. D. Coyle and L. R. B. Bryant, *J. Chem. Soc., Perkin Trans. I*, 1983, 531.
16 Y. Kanaoka, Y. Migita, K. Koyama, Y. Sato, H. Nakai, and T. Mizoguchi, *Tetrahedron Lett.*, 1973, 1193.
17 K. Maruyama and Y. Kubo, *Chem. Lett.*, 1978, 769; G. I. Hutchinson, R. H. Prager, and A. D. Ward, *Aust. J. Chem.*, 1980, **33**, 2477.
18 B. A. Mooney, R. H. Prager, and A. H. Ward, *Aust. J. Chem.*, 1981, **34**, 2695.
19 J. D. Coyle and G. L. Newport, *Synthesis*, 1979, 381; M. Machida, H. Takechi, and Y. Kanaoka, *Heterocycles*, 1980, **14**, 1255.
20 M. Machida, H. Takechi, Y. Shishido, and Y. Kanaoka, *Synthesis*, 1982, 1078.
21 B. Reichert, 'Die Mannich-Reaktion,' Springer-Verlag, Berlin, 1959.
22 M. Eckstein, A. Zejc, and A. Klusek, *Diss. Pharm. Pharmacol.*, 1967, **19**, 263.
23 P. O. Tawney, R. H. Snyder, R. P. Conger, K. A. Leibbrand, C. H. Stiteler, and A. R. Williams, *J. Org. Chem.*, 1961, **46**, 15.
24 S. R. Sandler and W. Karo, 'Organic Functional Group Preparations,' Academic Press, London and New York, 1972, vol. 3.
25 R. Wegscheider and J. Hecht, *Monatsh.*, 1903, **24**, 422.
26 N. M. Yoon and H. C. Brown, *J. Am. Chem. Soc.*, 1968, **90**, 2928.

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