HETEROCYCLIC ANALOGS OF PLEIADIENE.

59.* REACTION OF 1-ACETONYL- AND 1-PHENACYLPERIMIDINIUM SALTS WITH HYDRAZINE AND AMMONIUM ACETATE

V. V. Dal'nikovskaya, T. A. Kuz'menko, and A. F. Pozharskii

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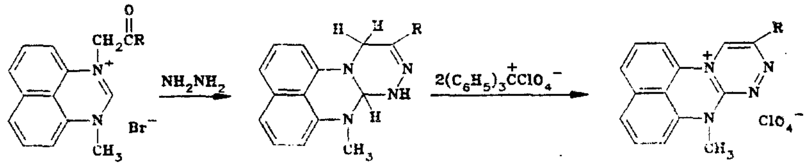
1-Acetonyl-3-methyl- and 1-methyl-3-phenacylperimidinium salts react with hydrazine hydrate to give tetrahydro-derivatives of the hitherto unknown heterocyclic system 1,2,4-triazino[4,3-a]perimidine. On boiling in acetic acid with ammonium acetate, 3-phenacyl- and 3-(2',4'-dimethyl)phenacyl-1-methylperidinium salts are converted into 1-methylamino-8-(4-phenylimidazol-1-yl)naphthalenes, with small amounts of 11-methylimidazo[1,2-a]perimidines also being formed.

The previously-synthesized [2] 1-acetonyl- and 1-phenacylperimidinium salts (I) contain a carbonyl group and a highly electron-deficient μ-carbon atom in the ring, in a favorable position for intramolecular cyclizations. To examine the feasibility of such reactions and the synthesis of condensed hetero-systems derived from perimidine, we have investigated the reactions of the salts (I) with hydrazine and ammonium acetate.

Salts (Ia-d) (other salts were not used in the reaction) react with hydrazine hydrate at room temperature to give even after 30 min 70-96% yields of tetrahydro-derivatives of the hitherto unknown heterocyclic system 1,2,4-triazino[4,3-a]perimidine (II). A similar reaction has been described for 2-phenylacylquinolinium and 1-phenacyl-3-cyanopyridinium salts [3], but in these instances cyclization takes place with heating. The higher reactivity of the perimidinium salts may be due to the high positive charge at $C_{(2)}$ in these compounds [4].



Ia-f IIa-d IIIa,b

I—III a $R=CH_3$; b $R=C_6H_5$; c $R=C_6H_4NO_2$-*p*; d $R=C_6H_4Br$-*p*; e $R=C_6H_4Cl$-*p*; f $R=C_6H_4OCH_3$-*p*; g $R=2,4$-$C_6H_3(CH_3)_2$

The structures of (IIa-d) were proved by their elemental analyses and IR and PMR spectroscopy. A characteristic feature of the PMR spectra is the nonequivalence of the CH_2 protons, these being seen as two doublets centered at 3.92 and 4.20 ppm for (IIa) and 4.42 and 4.80 ppm for (IIb), indicating that the hydrogenated triazine ring is nonplanar. The proton at the μ-carbon in (IIa) appears as a singlet at 5.17 ppm, whereas in (IIb) it is weakly spli into a doublet (5.45 ppm, J = 1.8 Hz), apparently as a result of spin coupling with the neighboring NH proton [in (IIb) the latter is also broadened as compared with the NH proton in (IIa)].

It was of interest to dehydrogenate (II) in order to obtain the heteroaromatic 1,2,4-triazino[4,3-a]perimidinium cation (III). In the cases of (IIa) and (IIb), we found that dehydrogenation proceeds readily on treatment with an excess of trityl perchlorate in methylene chloride. The products were bright yellow, extremely hygroscopic crystalline solids, which on the basis of their IR spectra and elemental analyses, were assigned the structures of cations (IIIa) and (IIIb). Specifically, the IR spectra of these compounds contained no absorption above 3100 cm^{-1}. It should be emphasized that the color of the salts (IIIa, b) in itself is an indication of the formation of the perimidinium ring. Perimidines and peri-

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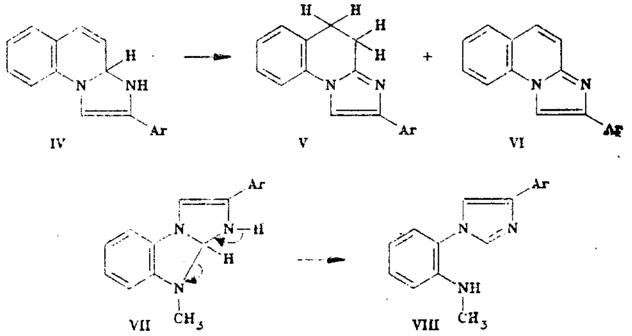
*For No. 58, see [1].

M. A. Suslov Rostov State University 344090. Research Institute for Physical and Organic Chemistry, Rostov-on-Don 344090. Translated from Khimiya Geterotsiklicheskikh Soedinenii, No. 10, pp. 1389-1395, October, 1986. Original article submitted June 17, 1985.

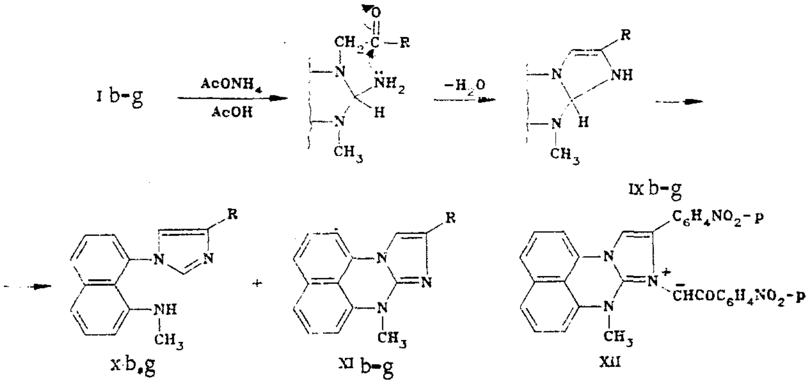
midinium salts are yellow in color, whereas 2,3-dihydroperimidines are colorless. Hence, the change from the colorless (II) to the yellow salts (III) indicates aromatization of the perimidine ring.

It is reported in the literature [5] that N-phenacyl salts of nitrogen heterocycles react in two ways with ammonium acetate. One route, typical of quinolines [5] and isoquinolines [6], leads to the formation of dihydroimidazo[1,2-a]quinolines and imidazo[1,2-b]isoquinolines, such as (V). The other route, characteristic of benzimidazoles [7], gives 1,4-diarylimidazoles (VIII). Both reactions are readily understood when the possible modes of stabilization of the key intermediates (IV) and (VII), which contain hydride-mobile hydrogen atoms, are considered. In the absence of an external oxidant, the only logical mode of stabilization of the intermediate (IV) is by hydride rearrangement to (V), which has aromatized benzene and imidazole rings. The intermediate (VII) is stabilized by fission of the C—N bond, but the result is the same, with the imidazole ring being formed in addition to the benzene ring. It is easy to see that the transfer of a hydride ion in the latter case, for example to the C═C double bond of the dihydroimidazole ring, would be energetically less favored. It must be borne in mind that in the presence of an external oxidant, or as a result of hydride disproportionation these reactions could theoretically also lead to the formation of fully aromatized imidazo[1,2-a]quinolines or imidazo[1,2-a]benzimidazoles, such as (VI). Compounds of this type have been obtained in small amounts only with quinoline and isoquinoline [5]. If the reaction is carried out in the presence of an oxidant such as ferric chloride, however, (VI) becomes the main reaction product [8].



Since perimidines and perimidinium salts display features of both azines and azoles in their reactions with nucleophiles [4], it is difficult to predict their mode of reaction with ammonium acetate. We have found that the salt (Ia) fails to react with ammonium acetate even on prolonged boiling in acetic acid, and the 1-methyl-3-phenacylperimidinium salts (Ib-f) react extremely slowly. For example, after boiling (Ib) for 18 h with ammonium acetate in acetic acid, 36% of the starting salt was recovered. The reaction products isolated in this instance were the previously-described [9] 11-methylimidazo[1,2-a]perimidine (XIb) (13%) and 1-methylamino-8-(4-phenylimidazol-1-yl)naphthalene (Xb) (34%). The IR spectrum of (Xb) showed no absorption for the carbonyl group, but absorption for the free NH group at 3480 cm^{-1} (in chloroform) was present. The PMR and mass spectral data were also in agreement with the structure (Xb). For example, in the PMR spectrum of (Xb) in deuterochloroform, in the aromatic proton region, a multiplet at 7.75 ppm and two quartets at 6.57 and 6.35 ppm, characteristic of the 2-H and 7-H protons in 1,8-diaminonaphthalenes [10], were present. The signal for the N-methyl group at 2.12 ppm is split to a doublet by spin coupling with the NH proton (J = 4 Hz), and the broadened NH signal is seen at 3.60 ppm. Under double resonance conditions at the absorption frequency of the NH proton, the latter signal disappears, and the methyl signal becomes a singlet (see scheme on following page).

To contrast to (Ib), the analogous salts (Ic-f) give no naphthalenes (X) on heating with ammonium acetate, only the imidazo[1,2a]perimidines (XIc-f) being isolated in yields of 28-45 %. Attention is drawn to the fact that salts (Ic-f) and the corresponding intermediates (IXc-f) contain electron-acceptor groups in the para-position of the phenacyl substituent, which facilitate the reduction of the double bonds conjugated therewith (see, for example, the report on the polarographic reduction of substituted azobenzenes [11]). All these groups possess positive MacDaniel—Brown σ_n-constants, namely +0.71 (NO_2), +0.39 (Br), +0.37 (Cl), and +0.12 (OCH_3) [12]. These results show indirectly that the principal mode

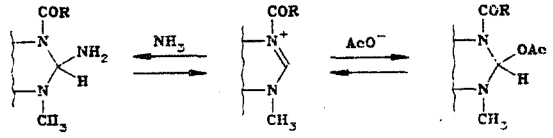


of stabilization of the intermediates (IXc-f) is by hydride disproportionation, whereas in the case of (IXb) stabilization with opening of the 2,3-dihydroperimidine ring predominates. A second product of hydride disproportionation should evidently be the corresponding imidazolidino[1,2-a]-2,3-dihydroperimidine, but there can be little doubt [4] that such compounds would be highly susceptible to oxidation, and would be chemically unstable. This probably also explains the fact that the reactions of salts (Ib-f) with ammonium acetate result in considerable resinification and the formation of a complex mixture of difficulty-separable compounds, each of which is present in small amounts. It is clear that the intermediate (IXc), which has a p-nitro-group, should show the greatest tendency to undergo hydride disproportionation. In fact, salt (Ic) gave the highest yields (45%) of the corresponding imidazo[1,2-a]perimidine (XIc).

When we had established the factors described above, it appeared to us of interest to use them to predict the course of the reaction of some new perimidinium salt with ammonium acetate. It was natural to select for this purpose a salt for which the reaction would lead to the preferential formation of a naphthalene derivative (X). Such a salt should contain electron-donor substituents in the phenacyl moiety. We decided on the salt (Ig), which contains methyl groups in the 2- and 4-positions of the phenacyl substituent. In accordance with expectations, the principal product of the reaction of (Ig) with ammonium acetate was the aminonaphthalene (Xg), obtained in 40% yield. The second product (12%) was the imidazo[1,2-a]perimidine (XIg).

We also obtained (XIc-g) by direct synthesis by the reaction between 2-amino-1-methylperimidine and the appropriate phenacyl bromides in the presence of sodium carbonate, as described in [9]. Under these conditions, however, p-nitrophenacyl bromide gave the yield (XII), which contains two p-nitrophenacyl residues. Compound (XIc) was obtained by reacting 2-amino-1-methylperimidine with p-nitrophenacyl bromide in a ratio of 2:1, in acetone.

It is rather difficult to understand why the perimidinium salts (I) react with ammonium acetate with greater difficulty than quinlinium and isoquinolinium salts, although the latter are more π-deficient. A possible explanation is as follows. The acetic acid–ammonium acetate system contains mainly two nucleophiles, the acetoxy ion and ammonia. The reaction (I) → (X) and (XI) requires ammonia, but its concentration in the mixture is of course much les than the concentration of acetoxy ions, the latter being much weaker nucleophiles than ammonia. In the case of the relatively low-π-deficient quinolinium and isoquinolinium salts, the acetoxy ions are scarcely bonded to them at all, thus facilitating their reaction with ammonia. On the other hand, the highly π-deficient perimidinium salts are able to react with AcO$^-$ ions to form covalent adducts (XIII). This results in a considerable shift of the equilibrium shown below to the right, as a result of which the concentration of free perimidinium cations in the mixture decreases sharply, and their reaction with ammonia is retarded.



This study has therefore shown that 1-methyl-3-phenacylperimidinium salts, in comparison with N-phenacylquinolinium and isoquinolinium salts on the one hand, and with 1-methyl-3-phenacylbenzimidazolium salts on the other hand, possess a number of features in common with them in their reactions with ammonium acetate, but also have special features. The latter consist, specifically, in the occurrence of the reaction leading to the formation of a product resulting from opening of the perimidine ring, together with the corresponding imidazo[1,2-a]perimidine.

EXPERIMENTAL

IR spectra were obtained on a UR-20 instrument, and PMR spectra on a Tesla BS-467 spectrometer (60 MHz). The double resonance PMR spectrum of (Xb) was obtained on a Bruker instrument (90 MHz), and the chemical shifts are given on the δ scale relative to the signal for TMS. Mass spectra were obtained on a JMS-01-SG2 instrument, with direct introduction of the sample into the ion source, ionizing voltage 75 eV.

1-Methyl-3-(p-nitrophenacyl)perimidinium Bromide (I) was obtained from 1-methylperimidine and p-nitrophenacyl bromide, as described in [2]. Yield, 70%. Yellow crystals, mp 234-235°C (decomp., from water). IR spectrum (Vaseline oil): 1700 ($C{=}O$), 3420, 3490 cm^{-1} (H_2O). Found, %: C 53.9; H 4.0; Br 17.7; N 9.1. $C_{20}H_{16}BrN_3O_3 \cdot H_2O$. Calculated, %: C 54.1; H 4.1; Br 18.0; N 9.5.

Salts (Id-g) were obtained similarly.

3-(p-Bromophenacyl)-1-methylperimidinium Bromide (Id) was obtained in 87% yield. Yellow crystals, mp 232-233°C (decomp., from alcohol). IR spectrum (Vaseline oil): 1700 ($C{=}O$), 3420, 3490 cm^{-1} (H_2O). Found, %: C 49.9; H 3.5; Br 33.4; N 5.7. $C_{20}H_{16}Br_2N_2O \cdot H_2O$. Calculated, %: C 50.2; H 3.8; Br 33.5; N 5.9.

1-Methyl-3-(p-chlorophenacyl)perimidinium Bromide (Ie) was obtained in 86% yield, yellow crystals, mp 230-231°C (from alcohol). Found, %: C 55.2; H 4.3; Hal 26.4; N 6.7. $C_{20}H_{16}BrClN_2O$. Calculated, %: C 55.4; H 4.1; Hal 26.7; N 6.4.

1-Methyl-3-(p-methoxyphenacyl)perimidinium Bromide (If), 90%, yellow crystals, mp 252-253°C (decomp., from alcohol). Found, %: C 61.5; H 4.8; Br 19.2; N 6.5. $C_{21}H_{19}BrN_2O_2$. Calculated, %: C 61.3; H 4.6; Br 19.5; N 6.8.

3-(2',4'-Dimethylphenacyl)-1-methylperimidinium Bromide (Ig), 80%, yellow crystals, mp 241-242°C (from alcohol). Found, %: C 64.4; H 5.2; Br 19.3; N 7.0. $C_{22}H_{21}BrN_2O$. Calculated, %: C 64.5; H 5.1; Br 19.5; N 6.8.

3,12-Dimethyl-1,4,5,12a-tetrahydro-1,2,4-triazino[4,3-a]perimidine (IIa). To a solution of 0.34 g (1 mmole) of (Ia) in 5 ml of methanol was added 0.5 ml (10 mmoles) of 99% hydrazine hydrate. The mixture was kept at 20°C for 30 min, and the solid which separated was filtered off and washed with water to give 0.21 g (85%) of shining, colorless needles, mp 168-169°C (from ethanol). IR spectrum (chloroform): 3370 cm^{-1} (NH). PMR spectrum ($CDCl_3$): 1.77 (s, 3H, $C{-}CH_3$), 2.90 (s, 3H, $N{-}CH_2$), 3.92 and 4.20 (two d., 2H, CH_2, J = 18 Hz), 5.17 (s, 1H, 12a-H), 5.31 (s, 1H, NH), 6.32 (q, 1H, 11-H), 6.50 (q, 1H, 6-H), 7.20 ppm (m, 4H, 7-H, 10-H). Found, %: C 71.2; H 6.1; N 22.1. $C_{15}H_{16}N_4$. Calculated, %: C 71.4; H 6.3; N 22.2.

3-Phenyl-12-methyl-1,4,5,12a-tetrahydro-1,2,4-triazino[4,3-a]perimidine (IIb) was obtained from (Ib) as for (IIa). Yield, 71%; colorless needles, mp 157-158°C (from ethyl acetate). IR spectrum ($CHCl_3$): 3370 cm^{-1} (NH). PMR spectrum ($CDCl_3$): 2.90 (s, 3H, CH_3), 4.42 and 4.80 (two doublets, 2H, CH_2, J = 18 Hz), 5.45 (d, 1H, 12a-H, J = 1.8 Hz), 5.90 (s, 1H, NH), 6.27 (q, 1H, 11-H), 6.52 (q, 1H, 6-H), 7.40 ppm (m, 9H, C_6H_5, 7-H, 10-H). Found, %: C 76.4; H 5.9; N 17.7. $C_{20}H_{18}N_4$. Calculated, %: C 76.4; H 5.7; N 17.8.

3-(p-Nitrophenyl)-12-methyl-1,4,5,12a-tetrahydro-1,2,4-triazino[4,3-a]perimidine (IIc) was obtained from (Ic) as described for (IIa), yield 82%. Yellow crystals, mp 162-163°C (decomp., from benzyl alcohol). IR spectrum ($CHCl_3$): 3390 cm^{-1} (NH). Found, %: C 67.0; H 4.5; N 19.4. $C_{20}H_{17}N_5O_2$. Calculated, %: C 66.9; H 4.7; N 19.5.

3-(p-Bromophenyl)-12-methyl-1,4,5,12a-tetrahydro-1,2,4-triazino[4,3-a]perimidine (IId) was obtained from (Id) as described above, yield, 96%. Colorless crystals, mp 169-170°C (decomp., from butanol). IR spectrum ($CHCl_3$): 3370 cm^{-1} (NH). Found, %: C 60.8; H 4.7; Br 20.0; N 13.9. $C_{20}H_{17}BrN_4$. Calculated, %: C 61.1; H 4.3; Br 20.3; N 14.2.

3-Methyl-1,2,4-triazino[4,3-a]perimidinium Perchlorate (IIIa). To a solution of 0.25 g (1 mmole) of (IIa) in 15 ml of dry methylene chloride was added gradually a solution of 0.68 g (2 mmoles) of trityl perchlorate in 40 ml of dry methylene chloride. The mixture became red in color, and a yellow solid separated. After standing for 1 h at 20°C, the solid was filtered off, taking precautions to avoid the ingress of atmospheric moisture. It was then washed with methylene chloride and dried in a Fischer pistol over calcium chloride. Yield, 0.2 g (57%); bright yellow crystals, mp 244-248°C (decomp.). Found, %: C 51.3; H 3.4; Cl 10.0; N 16.1. $C_{15}H_{13}ClN_4O_4$. Calculated, %: C 51.6; H 3.7; Cl 10.2; N 16.1.

3-Phenyl-1,2,4-triazino[4,3-a]perimidinium Perchlorate (IIIb) was obtained from (IIb) and trityl perchlorate as described above. Yield, 43%; yellow crystals, mp 227-230°C (decomp.). Found, %: C 58.2; H 3.2; Cl 8.3; N 13.2. $C_{20}H_{15}ClN_4O_4$. Calculated, %: C 58.5; H 3.6; Cl 8.6; N 13.6.

Reaction of 1-Methyl-3-phenacylperimidinium Bromide with Ammonium Acetate. A solution of 2.0 g (5 mmoles) of (Ib) and 3.75 g (50 mmoles) of ammonium acetate in 15 ml of acetic acid was boiled for 18 h. The mixture was then cooled, poured into 100 ml of water, basified with 22% ammonia, and the solid which separated was filtered off, washed with water, and dried. The solid was then treated with 100 ml of hot chloroform. The insoluble portion was starting material (0.7 g, 36%), and this was filtered off. The filtrate was concentrated as far as possible, and passed through a column of alumina, eluting with benzene. The first fraction (R_f 0.6) contained 2-phenyl-11-methylimidazo[1,2-a]perimidine (XIb), yield 0.12 g (13% calculated on salt reacted), crystals with a greenish tinge, mp 142-143°C (from alcohol), identical in all its physical properties with an authentic sample [9]. The second fraction (R_f 0.4) contained 1-methylamino-8-(4-phenylimidazol-1-yl)-naphthalene (Xb), yield 0.31 g (34% on salt reacted). Colorless needles, mp 180-181°C (from alcohol). IR spectrum ($CHCl_3$): 3480 cm^{-1} (NH). PMR spectrum ($CDCl_3$): 2.12 (d, J = 4 Hz, 3H, CH_3), 3.60 (s, 1H, NH), 6.35 (q, 1H, 2-H), 6.57 (q, 1H, 7-H), 7.75 ppm (m, 11-H). Mass spectrum (peak intensities given as % of maximum peak): 299 (24.4) [M^+]; 269 (1.17) [$M^+ - CH_3NH$]; 197 (13.71); 196 (100) [$M^+ - C_6H_5CN$]; 195 (10.88); 183 (7.09); 182 (24.55); 169 (22.50); 168 (7.74); 167 (14.94); 154 (9.29); 140 (3.61). Found, %: C 80.1; H 5.7; N 14.0. $C_{20}H_{17}N_3$. Calculated, %: C 80.3; H 5.7; N 14.0.

2-(p-Nitrophenyl)-11-methylimidazo[1,2-a]perimidine (XIc). A. A solution of 2.66 g (6 mmoles) of (Ic) and 4.50 g (60 mmoles) of ammonium acetate in 10 ml of glacial acetic acid was boiled for 12 h. The mixture was then cooled, and the dark red solid filtered off, and washed with water and alcohol (0.65 g). The mother liquors were poured into water, and the solid which separated was filtered off and dried. It was then dissolved in chloroform and chromatographed on a column of alumina (eluent, chloroform), taking the first fraction. A further 0.27 g of (XIc) was obtained. Yield 0.92 g (45%), dark red needles, mp 235-236°C (from DMF), sparingly soluble in benzene, chloroform, and alcohol. PMR spectrum (CF_3COOH): 3.32 (s, 3H, $N-CH_3$), 6.55 (q, 1H, 10-H), 6.85 (q, 1H, 5-H), 7.12 (m, 4H, 6-9H), 7.47 (s, 1H, 3-H), 7.50 (d, J = 8 Hz, 2H, 2',6'-H), 7.95 ppm (d, J = 8 Hz, 2H, 3',5'-H). Found, %: C 70.0; H 4.1; N 16.2; M^+ (mass spectroscopic) 342. $C_{20}H_{14}N_4O_2$. Calculated, %: C 70.2; H 4.1; 16.4.

B. A solution of 0.4 g (2 mmoles) of 2-amino-1-methylperimidine and 0.24 g (1 mmole) of p-nitrophenacyl bromide in 20 ml of acetone was boiled for 4 h. The solid hydrobromide of the starting amine which separated was filtered off and washed with acetone. The filtrate was evaporated, and the residue chromatographed on a column of alumina, eluent chloroform. Yield 0.23 g (67%), dark red crystals, mp 235-236°C (from DMF). No depression of melting point was observed on admixture with a sample from method A.

1-p-Nitrophenacyl-2-p-nitrophenyl-11-methylimidazo[1,2-a]perimidinium Ylid (XII). To a solution of 0.5 g (2.5 mmoles) of 2-amino-1-methylperimidine in 20 ml of alcohol was added 0.6 g (2.5 mmoles) of p-nitrophenacyl bromide and a solution of 0.2 g (2.5 mmoles) of sodium bicarbonate in 1 ml of water. The mixture was boiled for 2 h, and the brown solid which separated was filtered off and washed with alcohol. Yield 0.47 g (74%), brown crystals, mp over 300°C (from DMF). IR spectrum (Vaseline oil): 1700 cm^{-1} (C=O). Found, %: C 66.2; H 3.8; N 13.6; M^+ (mass spectroscopic) 505. $C_{28}H_{19}N_5O_5$. Calculated, %: C 66.5; H 3.8; N 13.9.

2-(p-Bromophenyl)-11-methylimidazo[1,2-a]perimidine (XId). A. A solution of 2.4 g (5 mmoles) of (Id) and 3.75 g (50 mmoles) of ammonium acetate in 20 ml of acetic acid was boiled for 18 h. The mixture was then cooled, poured into water, and basified with 22% ammonia. The solid which separated was filtered off, dried, and washed with hot chloroform. The insoluble starting salt (0.93 g) was filtered off, and the filtrate chromatographed on a column of alumina, eluent benzene. Yield 0.3 g (28% on salt reacted), greenish crystals, mp 204-205°C (from butanol). Found, %: C 63.6; H 3.8; Br 21.0; N 11.5. $C_{20}H_{14}BrN_3$. Calculated, %: C 63.8; H 3.7; Br 21.2; N 11.2.

B. To a solution of 0.3 g (1.5 mmoles) of 2-amino-1-methylperimidine and 0.42 g (1.5 mmoles) of p-bromophenacyl bromide was added 0.13 g (1.5 mmoles) of sodium bicarbonate, and the mixture boiled for 4 h. The solvent was evaporated, and the residue dissolved in chloroform and chromatographed on a column of alumina, eluent chloroform. Yield 0.35 g (62%). The compound was identical with that obtained by method A.

2-(p-Chlorophenyl)-11-methylimidazo[1,2-a]perimidine (XIe) was obtained as for the preceding compound, method A giving a yield of 36% and method B, 63%. Greenish crystals, mp 201-202°C (from butanol). PMR spectrum ($CDCl_3$): 3.58 (s, 3H, $N-CH_3$), 6.50 (q, 1H, 10-H), 6.75 (q, 1H, 5-H), 7.18-7.35 (m, 7H, 3,6-9H, 2',6'-H), 7.7 ppm (d, J = 8 Hz, 2H, 3',5'-H). Found, %: C 72.4; H 4.5; Cl 10.5; N 12.4. $C_{20}H_{14}ClN_3$. Calculated, %: C 72.4; H 4.2; Cl 10.7; N 12.7.

2-(p-Methoxyphenyl)-11-methylimidazo[1,2-a]perimidine (XIf) was obtained as for (XIe), method A giving a yield of 37%, and method B, 77%. Greenish crystals, mp 154-155°C (from acetonitrile). Found, %: C 76.7; H 5.4; N 12.5. $C_{21}H_{17}N_3O$. Calculated, %: C 77.0; H 5.2; N 12.8.

Reaction of 3-(2'-4'-Dimethylphenacyl)-1-methylperimidinium Bromide with Ammonium Acetate. A solution of 1.63 g (4 mmoles) of salt (Ig) and 2 g (40 mmoles) of ammonium acetate in 10 ml of glacial acetic acid was boiled for 18 h. The mixture was then cooled, poured into water, basified with 22% ammonia, and extracted with chloroform. The solvent was evaporated, and the residue chromatographed on a column of alumina, eluent benzene. The first fraction contained 2-(2',4'-dimethylphenyl)-11-methylimidazo[1,2-a]perimidine (XIg) (R_f 0.6), yield 0.15 g (12%), greenish crystals, mp 151-152°C (from alcohol). Found, %: C 81.0; H 6.0; N 13.1. $C_{22}H_{19}N_3$. Calculated, %: C 81.3; H 5.8; N 12.9. The second fraction (R_f 0.4) contained 1-methylamino-8-[(2',4'-dimethylphenyl)imidazol-1-yl]naphthalene (Xg), yield 0.52 g (40%). Colorless crystals, mp 178-179°C (from alcohol). IR spectrum ($CHCl_3$): 3480 cm^{-1} (NH). Found, %: C 80.5; H 6.7; N 13.0. $C_{22}H_{21}N_3$. Calculated, %: C 80.7; H 6.5; N 12.9.

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