HETEROCYCLIC ANALOGS OF PLEIADIENE. 52.* INTRAMOLECULAR C-ACYLATION OF PERIMIDINES AND PERIMIDONES. SYNTHESIS OF A NEW HETEROCYCLIC SYSTEM — 3,4,5,6-TETRAHYDROPYRIDO[3,2,1-k,l]PERIMIDINE

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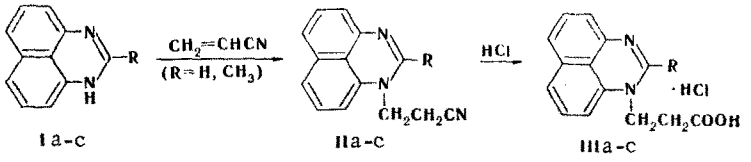
Intramolecular C-acylation in the 9 position of β-(1-perimidinyl)- and β-(1-perimidonyl)propionic acids and cyanoethylation of perimidines and perimidones were studied. Derivatives of a new bridged system, viz., 3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine, were synthesized for the first time.

Perimidines and perimidones are acylated smoothly by carboxylic acids in polyphosphoric acid (PPA) to give 6(7)- and 4(9)-monoacyl derivatives [2, 3]. One might have expected that intramolecular C-acylation should also proceed just as readily. In this research we studied the intramolecular acylation of ω-(1-perimidinyl) carboxylic acids in the 9 position, which bears the maximum negative charge in the perimidine molecule. We expected that this reaction would open up a route to the synthesis of new bridged systems based on perimidine.

Perimidine (Ia) and 2-methylperimidine (Ib) were cyanoethylated with excess acrylonitrile in glycerol. Products IIa, b were obtained in 78 and 35% yields, respectively. The cyanoethylation of Ia, b was previously carried out [4, 5] with a large excess of acrylonitrile in the presence of sodium ethoxide. The yield of IIa was not indicated, while IIb was obtained in 40% yield. We repeated the cyanoethylation of Ia by the method in [4]; the desired compound was obtained in only 26% yield. The use of sodium amide in place of sodium ethoxide as the catalyst for cyanoethylation makes it possible to obtain IIa in 10% yield. Compound IIa is formed in the same yield in the reaction of Ia with β-bromopropionitrile in an alcohol solution of alkali. Compound IIa was obtained in good yield (67%) when trimethylphenylammonium hydroxide was used as the catalyst. However, the use of glycerol as the solvent has a clear advantage in the cyanoethylation of perimidine, since it does not require the use of a catalyst, it excludes the formation of polymerization products, and it ensures a high yield.



Ia-c IIa-c IIIa-c

I-III a R=H; b R=CH_3; c R=C_6H_5

However, we were unable to subject 2-phenylperimidine (Ic) to direct cyanoethylation. This is not surprising, since the 2-phenyl group in the perimidine ring hinders [6] the incorporation of other substituents in the 1 position (except in the case of N-methylation [7]). We obtained 1-β-cyanoethyl-2-phenylperimidine (IIc) by recyclization [8] via the scheme:

*See [1] for communication 51.

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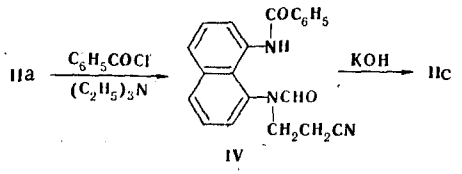
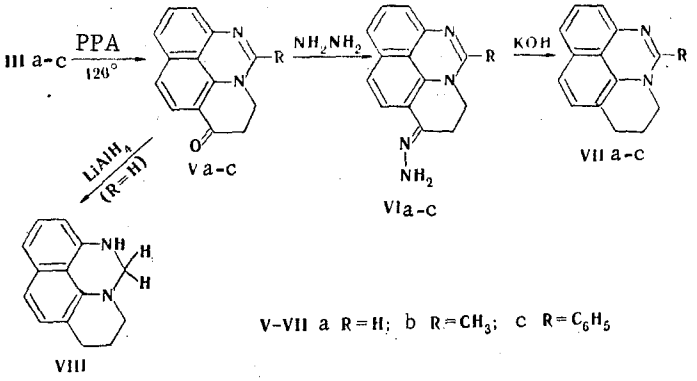
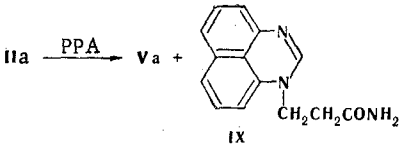


It should be noted that the formation and purification of pseudobase IV are achieved with greater difficulty than in the case of compounds with other substituents in the 1 position of perimidine [8]. However, the cyclization step under the influence of alkali proceeds smoothly and, interestingly enough, is not accompanied by a change in the cyanoethyl group.

β-(1-Perimidinyl)propionic acids (III) were obtained in high yields by refluxing IIa-c in hydrochloric acid. When these acids are heated with PPA at 120°C, they give intramolecular acylation products V in high yields. The structure of V is confirmed by their IR and PMR spectra, as well as by the results of elementary analysis. In particular, the IR spectra do not contain absorption above 3100 cm^{-1} but do contain a $\nu_{C=O}$ band at 1680 cm^{-1}. In addition, a characteristic property of V is their yellow-green luminescence, which is also observed for 6(7)- and 4(9)-acylperimidines [9] (none of the starting compounds luminesces). Compounds V were converted by the Wolff-Kishner reaction to derivatives of a new heterocyclic system, viz., 3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidines (VII). The intermediately formed hydrazone was isolated only in the case of parent compound VIa. The C=O group in V is reduced to a methylene group also by lithium aluminum hydride; however, in this case the C=N bond is also reduced, as a result of which VIII is obtained.

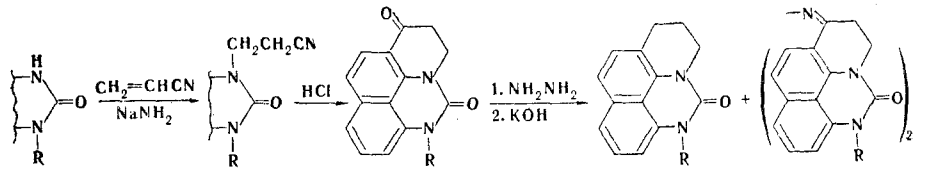


It seemed possible to us to obtain intramolecular acylation product V directly from cyanoethyl derivatives II by the Houben-Hoesch reaction. However, when IIa was heated with PPA at 120°C, Va was obtained in only 23% yield, and the principal product was acid amide IX.



We carried out similar transformations on the basis of perimidones X. 1-Methylperimidone (Xa) is cyanoethylated by acrylonitrile in the presence of sodium amide (we were unable to realize cyanethylation in glycerol because of the low solubility of Xa) to give XIa in 40% yield. Compound XIa undergoes cyclization to give XIIa in a yield that is close to quantitative when it is refluxed with concentrated HCl. Such facile cyclization is undoubtedly explained by the fact that perimidones undergo electrophilic substitution in acidic media in the neutral form [9], while perimidines undergo electrophilic substitution in the cationic form. Compound XIIa was converted via the Wolff-Kishner reaction to oxo derivative XIII; the slightly soluble azine XIV is a side product in this reaction. It should be noted that the oxo group in the perimidones remains unchanged even in the case of prolonged refluxing with hydrazine hydrate.

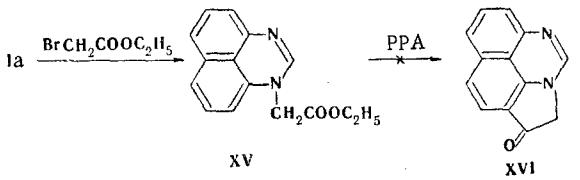
It seemed of interest to accomplish double intramolecular acylation in 1,3-bis(β-cyanoethyl)perimidone (XIb). The cyanoethylation of perimidone Xb to XIb takes place in the presence of $NaNH_2$ but gives the product in low yield (5%). However, when triethylphenyl- or trimethylbenzylammonium hydroxide is used as the catalyst, the yield of XIb



X-XII a R=CH_3; X b R=H; XI b R=CH_2CH_2CN; XII b R=CH_2CH_2COOH

increases to 50%. Intramolecular acylation only with the participation of one cyanoethyl group is observed when XIb is refluxed in concentrated HCl. The second group in this case is saponified to give the acid, as a result of which XIIb is obtained. Acid XIIb does not undergo a second intramolecular acyaltion even when it is heated in PPA. This is evidently explained by the pronounced decrease in the negative charge in the ortho position (2) of the naphthalene ring of XIIb under the influence of the acyl group.

We were unable to subject ethyl N-perimidinylacetate, obtained by the reaction of Ia with ethyl monobromoacetate in DMF, to intramolecular acylation even under severe conditions (210°C). In our opinion, this is explained by the steric strain of the five-membered ring in bridged system XVI.



The properties of the derivatives of the heteroaromatic 3,4,5,6-tetrahydropyrido-[3,2,1-k,l]perimidine system (XIIa) will be described in our next communication.

EXPERIMENTAL

The IR spectra were measured with a UR-20 spectrometer, the UV spectrum was recorded with an SF-4A spectrophotometer, and the PMR spectra were recorded with a Tesla BS-467 spectrometer (60 MHz) with hexamethyldisiloxane as the internal standard.

1-β-Cyanoethylperimidine (IIa). A) A 4-ml (0.06 mole) sample of acrylonitrile was added dropwise to a solution of 5.0 g (0.03 mole) of Ia in 60 ml of glycerol heated to 120°C, and the mixture was stirred at 110°C for 5 h. It was then cooled and poured into water (200 ml). The 1-β-cyanoethylperimidine was extracted with chloroform (two 100-ml portions), and the extract was washed thoroughly with water, dried with $CaCl_2$, and evaporated. The residue was passed through a chromatographic column filled with Al_2O_3 (elution with chloroform) with collection of the first fraction, from which 5.1 g (78%) of shiny yellow crystals with mp 130-131°C (from ethyl acetate or benzene) (mp 136-139°C [4]) was obtained. IR spectrum ($CHCl_3$): 2265 cm^{-1} (C≡N). Found: C 75.8; H 4.9; N 18.8%. $C_{14}H_{11}N_3$. Calculated: C 76.0; H 5.0; N 19.0%.

B) A solution of 0.4 g of trimethylphenylammonium hydroxide in 2 ml of alcohol was added to a suspension of 3.36 g (0.02 mole) of Ia in 25 ml of dioxane, after which 2 ml (0.03 mole) of acrylonitrile was added dropwise to the resulting solution at 40-45°C, and the mixture was stirred at 40-45°C for 30 min. It was then cooled, and the precipitate was removed by filtration and purified as in method A to give 2.9 g (67%) of product.

C) A 5-ml (0.075 mole) sample of acrylonitrile was added dropwise with the continuous passage of nitrogen to a suspension of 5 g (0.03 mole) of Ia and 1.2 g (0.03 mole) of finely ground sodium amide in 40 ml of absolute dimethylaniline (DMA) heated to 150°C, and the mixture was stirred at 150°C for 2 h. The DMA was removed by steam distillation, and the aqueous layer was treated with chloroform (three 50-ml portions). Workup of the chloroform extract as in method A gave 1.2 g (18%) of IIa.

D) Sodium ethoxide (0.1 g in 2 ml of ethanol) was added in portions with vigorous stirring to a suspension of 5 g (0.03 mole) of Ia in 50 ml of acrylonitrile. When spontaneous heating up of the reaction mixture ceased, it was refluxed for 2 h. It was then treated with hot benzene (150 ml), and the polymerization products were removed by filtration. Workup of the benzene filtrate as in method A gave 1.7 g (26%) of IIa.

E) A solution of 2 g (0.03 mole) of KOH in 20 ml of alcohol was added with continuous passage of nitrogen to a solution of 5 g (0.03 mole) of Ia in 80 ml of alcohol, after which 6.5 g (0.03 mole) of β-bromopropionitrile and 1.5 g of potassium iodide were added, and the mixture was refluxed for 3 h. The alcohol was removed by distillation, 80 ml of benzene was added to the residue, and the mixture was filtered. Workup of the benzene solution as in method A gave 0.7 g (11%) of IIa.

The starting perimidine was isolated in the reaction of Ia with acrylonitrile in liquid ammonia in the presence of sodium.

1-β-Cyanoethyl-2-methylperimidine (IIb). This compound, with mp 145-146°C (mp 145-146°C [5]), was obtained in 35% yield by the method used to obtain nitrile IIa.

N-β-Cyanoethyl-N-formyl-N'-benzoyl-1,8-naphthalenediamine (IV). A 1.3-ml (0.013 mole) sample of triethylamine and (dropwise) 1.2 ml (0.01 mole) of benzoyl chloride were added to a warm solution of 2.2 g (0.01 mole) of IIa in 70 ml of benzene, and the mixture was refluxed with stirring for 1.5 h. The benzene was then removed by distillation, and the residue was treated with water (200 ml). The insoluble material was removed by filtration, dried, and purified by chromatography (Al_2O_3 and $CHCl_3$) to give 2 g (60%) of white crystals with mp 134-135°C (from benzene with petroleum ether). IR spectrum ($CHCl_3$): 3435 (N–H), 1680 (C=O), and 2260 cm^{-1} (C≡N). Found: C 73.8; H 4.9; N 12.0%. $C_{21}H_{17}N_3O_2$. Calculated: C 73.5; H 5.0; N 12.2%.

1-(β-Cyanoethyl)-2-phenylperimidine (IIc). A suspension of 1.1 g (3 mmole) of IV in 40 ml of 10% aqueous KOH solution was stirred on a boiling-water bath for 2 h, after which it was cooled, and the yellow precipitate was removed by filtration, washed with water, and purified by chromatography [elution with chloroform–benzene (1:1)] to give 0.7 g (93%) of a product with mp 202-203°C (from alcohol). IR spectrum ($CHCl_3$): 2260 cm^{-1} (C≡N). Found: C 81.1; H 5.3; N 14.2%. $C_{20}H_{15}N_3$. Calculated: C 80.8; H 5.1; N 14.1%.

β-(1-Perimidinyl)propionic Acids (III). A solution of 0.01 mole of IIa-c in 15 ml of hydrochloric acid (1:1) was refluxed for 2-3 h, after which it was cooled, and the yellow precipitate was removed by filtration and dried in a vacuum desiccator over P_2O_5 to give IIIa, with mp 242-243°C (from water), in 96% yield. IR spectrum (mineral oil): 1740 cm^{-1} (C=O). Found: C 60.8; H 5.0; Cl 12.6; N 10.1%. $C_{14}H_{12}N_2O_2 \cdot HCl$. Calculated: C 60.8; H 4.7; Cl 12.8; N 10.1%. Compound IIIb was previously obtained in [5] and had mp >300°C (mp >290°C [5]). Compound IIIc, with mp >300°C (from water), was obtained in 80% yield. IR spectrum (mineral oil): 1720 cm^{-1} (C=O). Found: N 9.7%. $C_{20}H_{16}N_2O_2 \cdot HCl$. Calculated: N 9.4%.

6-Oxo-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidines (Va-c). A mixture of 7 mmole of N-perimidinylpropionic acid hydrochloride IIIa-c and 5-7 g of PPA was stirred at 70-80°C until hydrogen chloride evolution ceased, and the resulting melt was stirred at 120°C for 3-4 h. The hot mixture was poured into ice water (200 ml), and the aqueous mixture was slowly made alkaline with ammonia to pH 8-9. The resulting precipitate was separated, washed with water, dried, dissolved in chloroform, and passed through a column (Al_2O_3 and benzene) with collection of the first fraction. All V were bright-yellow crystals and were crystallized from alcohol.

6-Oxo-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine (Va). This compound, with mp 196°C, was obtained in 85% yield. IR spectrum ($CHCl_3$): 1680 cm^{-1} (C=O). PMR spectrum (CF_3COOH): 2.77 (t, 2H, $COCH_2$, J = 7.5 Hz), 4.15 (t, 2H, NCH_2, J = 7.5 Hz), 6.80 (m, 1H, 4-H), 7.28 (m, 4H, 5-H–8-H), and 8.05 ppm (s, 1H, 2-H). Found: C 75.4; H 4.5; N 12.9%. $C_{14}H_{10}N_2O$. Calculated: C 75.7; H 4.5; N 12.6%.

6-Oxo-2-methyl-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine (Vb). This compound, with mp 183-184°C, was obtained in 83% yield. IR spectrum ($CHCl_3$): 1670 cm^{-1} (C=O). PMR spectrum ($CDCl_3$): 2.10 (s, 3H, CH_3), 2.75 (t, 2H, $COCH_2$, J = 7.5 Hz), 3.90 (t, NCH_2, J = 7.5 Hz), and 6.90 ppm (m, 5H). Found: C 75.8; H 5.0; N 12.1%. $C_{15}H_{12}N_2O$. Calculated: C 76.0; H 5.1; N 11.9%.

6-Oxo-2-phenyl-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine (Vc). This compound, with mp 151-152°C, was obtained in 80% yield. IR spectrum ($CHCl_3$): 1670 cm^{-1} (C=O). Found: N 9.3%. $C_{20}H_{14}N_2O$. Calculated: N 9.4%.

6-Oxo-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine Hydrazone (VIa). A solution of 1.5 g (6.7 mmole) of ketone Va and 4 ml of hydrazine hydrate in 25 ml of alcohol was re-

fluxed for 2 h, after which it was cooled, and the precipitate was removed by filtration to give 1.3 g (81%) of orange crystals with mp 186-188°C (dec., from alcohol). IR spectrum (mineral oil): 3330 and 3200 cm^{-1} (NH_2). Found: C 70.9; H 5.0; N 24.0%. $C_{14}H_{12}N_4$. Calculated: C 71.2; H 5.1; N 23.7%.

3,4,5,6-Tetrahydropyrido[3,2,1-k,l]perimidines (VIIa-c). A solution of 3 mmole of Va-c and 2 ml of 99% hydrazine hydrate in 16 ml of diethylene glycol was stirred at 110°C for 1.5-2 h, after which 1.7 mmole of finely ground dry KOH was added, and the temperature was raised to 200-205°C. The mixture was then stirred at this temperature until the excess hydrazine hydrate and water had been removed by distillation. The mixture was then poured into 70 ml of ice water, and the precipitate was removed by filtration and washed with water. The products were obtained in 84-86% yields. All VII were obtained as yellow crystals (from octane).

3,4,5,6-Tetrahydropyrido[3,2,1-k,l]perimidine (VIIa). This compound had mp 121°C. PMR spectrum (CCl_4): 1.80 (m, 2H, CH_2), 2.6 (m, 2H, CH_2), 3.35 (t, 2H, N—CH_2), 6.75 (m, 5H, 4-H—8-H), and 7.35 (s, 1H, 2-H). UV spectrum (methanol), λ_{max} (log ε): 238 (4.26), 332 (3.84), and 410 nm (2.60). No melting-point depression was observed for a mixture of this product with the compound obtained directly by the action of KOH on hydrazone VIa.

2-Methyl-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine (VIIb). This compound had mp 130-131°C. Found: N 12.4%. $C_{15}H_{14}N_2$. Calculated: N 12.6%.

2-Phenyl-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine (VIIc). This compound had mp 147-148°C. Found: C 84.6; H 5.4; N 10.0%. $C_{20}H_{16}N_2$. Calculated: C 84.5; H 5.6; N 9.9%.

1,2,3,4,5,6-Hexahydropyrido[3,2,1-k,l]perimidine (VIII). A 1.6-g (7 mmole) sample of Va was added with stirring to a suspension of 1.6 g (0.04 mole) of $LiAlH_4$ in 65 ml of absolute ether, and the mixture was refluxed with stirring for 2 h. It was then cooled, and 15 ml of water was added cautiously. The ether layer was separated, the ether was evaporated and the residue was dissolved in 10 ml of benzene and passed through a column (Al_2O_3 and benzene) with collection of the first colorless fraction, workup of which gave 1.2 g (80%) of colorless needles with mp 109-110°C (from alcohol). IR spectrum ($CHCl_3$): 3410 cm^{-1} (NH). Found: C 79.7; H 6.4%. $C_{14}H_{14}N_2$. Calculated: C 80.0; H 6.7%.

β-(1-Perimidinyl)propionamide (IX). A solution of 1.1 g (0.05 mole) of Ia in 10 g of PPA was stirred at 120°C for 2.5 h, and the hot mass was then poured into ice water (100 ml). The aqueous mixture was made alkaline to pH 8-9 with ammonia, and the resulting precipitate was removed by filtration and dried. Chloroform (10 ml) was added to the dry substance, and insoluble amide IX was removed by filtration to give 0.65 g (55%) of a product with mp 223-225°C (dec., from cyclohexanone). IR spectrum (mineral oil): 3260, 3055 (NH_2); 1670 cm^{-1} (C=O). Found: C 70.0; H 5.8; N 17.9%. $C_{14}H_{13}N_3O$. Calculated: C 70.3; H 5.5; N 17.6%. Compound Va was isolated from the chloroform filtrate in 23% yield.

1-β-Cyanoethyl-3-methylperimidone (XIa). An 8.5-ml (0.13 mole) sample of acrylonitrile was added dropwise with vigorous stirring to a refluxing suspension of 2.1 g (0.01 mole) of N-methylperimidone and 0.5 g (0.015 mole) of sodium amide in 150 ml of absolute xylene, and the mixture was refluxed for 3 h. The hot mixture was filtered to remove the polymerization products, and the filtrate was evaporated *in vacuo* (with a water aspirator). The residue was dissolved in 15 ml of chloroform and passed through a chromatographic column (Al_2O_3, $CHCl_3$) with collection of the first colorless fraction, workup of which gave 1 g (40%) of a product with mp 162°C (from alcohol or ethyl acetate). IR spectrum ($CHCl_3$): 2260 (C≡N) and 1680 cm^{-1} (C=O). Found: C 71.5; H 5.2; N 17.0%. $C_{15}H_{13}N_3O$. Calculated: C 71.7; H 5.2; N 16.7%.

Compound XIa was formed in trace amounts in the reaction of Xa with acrylonitrile in the presence of sodium ethoxide; the starting N-methylperimidone was isolated in the reaction of Xa with β-bromopropionitrile.

1,3-Bis(β-cyanoethyl)perimidone (XIb). A 1.5-ml (23.2 mmole) sample of acrylonitrile was added with stirring at 50°C to a solution of 1 g (5.8 mmole) of perimidone in 30 ml of dioxane and 0.2 g of trimethylphenylammonium hydroxide in 1 ml of alcohol, and the mixture was stirred at 50-55°C for 2 h. It was then cooled, and the resulting precipitate was removed by filtration and washed with water to give 0.75 g (46%) of white crystals with mp 270-271°C (from DMF). IR spectrum (mineral oil): 2258 (C≡N) and 1675 cm^{-1} (C=O). Found: C 70.4; H 5.1; N 19.0%. $C_{17}H_{14}N_4O$. Calculated: C 70.3; H 5.0; N 19.3%. Compound XIb was

obtained in 5% yield in the reaction of perimidone with acrylonitrile in xylene in the presence of $NaNH_2$.

1-Methyl-2,6-dioxo-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine (XIIa). A solution of 0.3 g (1.2 mmole) of XIa in 7 ml of concentrated HCl was refluxed for 3 h, after which it was cooled, and the resulting precipitate was washed with water and purified by chromatography (Al_2O_3, benzene) to give light-yellow prisms with mp 241-242°C (from toluene). The yield was 0.28 g (93%). IR spectrum ($CHCl_3$): 1680 cm^{-1} (C=O). PMR spectrum (CF_3COOH): 2.65 (t, 2H, $COCH_2$), 3.20 (s,* 3H, NCH_3), 4.07 (s,* 2H, NCH_2), and 7.07 ppm (m, 5H, 4-H–8-H). Found: C 71.1; H 4.9; N 11.3%. $C_{15}H_{12}N_2O_2$. Calculated: C 71.4; H 4.8; N 11.1%.

1-(2-Carboxyethyl)-2,6-dioxo-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine (XIIb). A solution of 0.7 g (2.6 mmole) of Xb in 150 ml of concentrated HCl was refluxed for 5 h, after which it was cooled and poured into water, and the resulting precipitate was removed by filtration to give yellow-green crystals with mp 269-270°C (from acetic acid or DMF). The yield was 0.6 g (80%). Found: C 66.0; H 4.5; N 9.2%. $C_{17}H_{14}N_2O_4$. Calculated: C 65.8; H 4.5; N 9.0%.

2-Oxo-1-methyl-3,4,5,6-tetrahydropyrido[3,2,1-k,l]perimidine (XIII). This compound was obtained in 74% yield from ketone XIIa by the general method for the synthesis of VIIa-c. The colorless crystals had mp 127-128°C (from octane). IR spectrum ($CHCl_3$): 1670 cm^{-1} (C=O). Found: C 75.8; H 5.7; N 11.9%. $C_{15}H_{14}N_2O$. Calculated: C 75.6; H 5.9; N 11.8%. The azine (XIV) of XIIa was isolated as a side product in 8% yield and was insoluble in ordinary organic solvents. The yellow crystals had mp >300°C. IR spectrum (mineral oil): 1685 cm^{-1} (C=O). Found: C 68.3; H 4.9; N 21.0%. $C_{30}H_{24}N_8O_2$. Calculated: C 68.2; H 4.6; N 21.2%.

1-Carbethoxymethylperimidine (XV). A 2.2-ml (0.02 mole) sample of ethyl monobromoacetate was added to a solution of 3.3 g (0.02 mole) of perimidine in 20 ml of DMF, and the mixture was stirred on a boiling-water bath for 4 h. It was then distilled *in vacuo* to remove the DMF, and the residue was treated with water (200 ml). The aqueous mixture was made alkaline to pH 8-9 with ammonia, and the resulting precipitate was removed by filtration, washed with water, and dried. The dry product was dissolved in 20 ml of chloroform, the starting perimidine was removed by filtration, and the filtrate was passed through a column (Al_2O_3, $CHCl_3$) with collection of the first fraction, workup of which gave 0.7 g (14%) of a product with mp 110-111°C (mp 110-111°C [5]).

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*The signals are markedly broadened; this is typical for other acylperimidines. It is possible that this is a consequence of their protonation in CF_3COOH (the solution is bright red, although the bases themselves are light yellow).