Resonance-Stabilized α-Naphthylmethyl Carbocations and Spiro Compounds Based Thereon: VII.* Transformations of α-Naphthylmethyl Carbocations Stabilized by One Electron-Donor Group or *peri*-Fused Heteroring

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Abstract—1-Hydroxymethyl- and 1-alkoxymethylnaphthalenes containing dimethylamino and methoxy groups or a heterering in positions *4* and *5* react with protic and Lewis acids to give 1-naphthylmethyl carbocations. Reactions of the latter with the initial alcohol molecule lead to the formation of oligomerization or dehydrogenation (to aldehyde) products or the corresponding dinaphthylmethanes. In some cases, the process was accompanied by cyclodimerization to form cyclohexadienone spiro derivatives in a small yield.

1-Naphthylmethyl carbocations persistently attract researchers' attention due to their specific reactivity, steric structure, and chromophoric properties [2–6]. We recently discovered a new reaction which has no analogies in the chemistry of naphthalene derivatives. Treatment with protic [7–10] or Lewis acids [9–11] of 1-hydroxymethylnaphthalenes **Ia**–**Id** having electron-donor substituents in positions *4* and *5* gives rise to intermediate resonance-stabilized carbocation **II** which undergoes *in situ* "head-to-tail" (in protic medium) or "tail-to-head" (over Al₂O₃, TiO₂, or SiO₂) cyclodimerization with formation of spiro compounds **III** and **IV**, respectively (Scheme 1). In the first case, due to considerable contribution of structure **B**, cation **II** acts as 1,4-diene and dienophile simultaneously, while symmetric spiro compounds **IV** are formed as a result of two-step electrophilic substitution where the first step involves attack by carbocation on the free *peri* position of the initial alcohol.

In order to get a deeper insight into the reaction pattern, we tried to elucidate whether [4+2]-cyclodimerization will occur with cations **V** and **VI** stabilized

by only one donor group and with cations derived from perimidinones **VII** and 2,3-dihydroperimidines **VIII** in which the role of electron-donor substituent is played by heterering.

We found that heating of alcohol **X** (which was prepared by reduction of aldehyde **IX**) in trifluoro-


V, R = Me₂N (**a**), MeO (**b**); VII, R = Me (**a**), Et (**b**);
VIII, R = H (**a**), Me (**b**).

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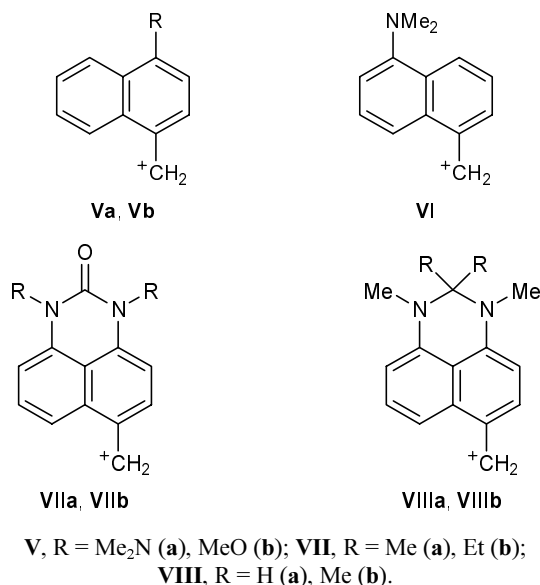
* For communication VI, see [1].
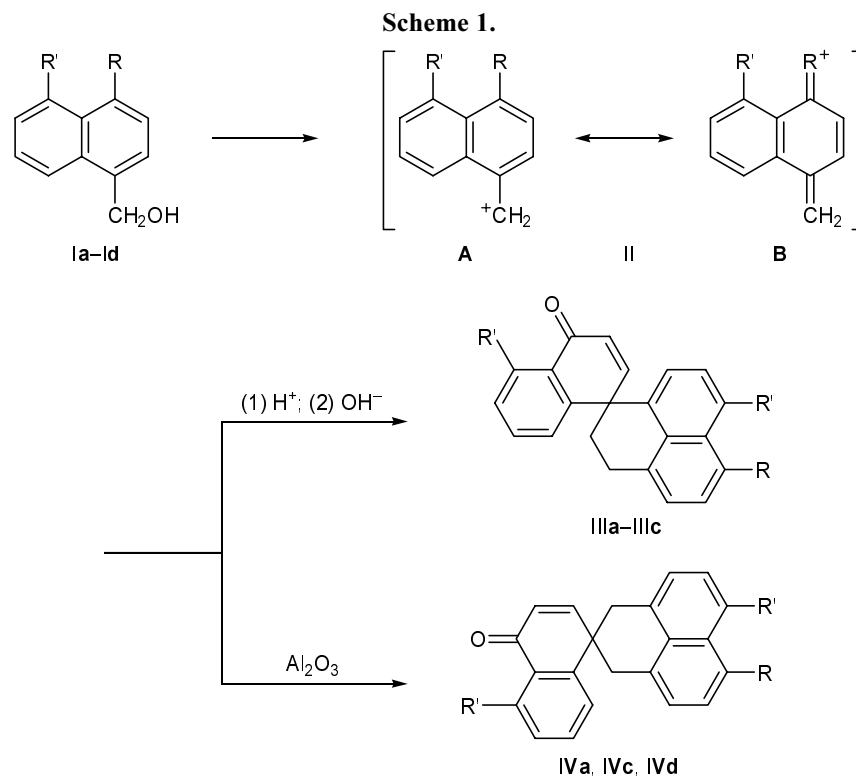
Scheme 1.

R = R' = Me$_2$N (**a**), R = R' = Et$_2$N (**b**), R = Me$_2$N, R' = MeO (**c**), R = R' = MeO (**d**).

acetic acid gave 9% of asymmetric spiro compound **XI** (Scheme 2). The structure of product **XI** unambiguously followed from its ^{1}H NMR spectrum which contained two-proton multiplets at δ 2.32 and 3.25 ppm due to the CH$_2$CH$_2$ group and two one-proton doublets at δ 6.46 and 7.26 ppm (J = 10.2 Hz) from protons in the quinoid ring [7, 8]. The other products were dinaphthylmethane **XII** (57%) and 1-dimethylamino-4-methylnaphthalene (**XIII**) (6%). Obviously, compound

XII was formed via electrophilic replacement of the CH$_2$OH group in the initial alcohol by carbocation **Va**; analogous replacements have already been reported [10]. Undoubtedly, 1-dimethylamino-4-methylnaphthalene (**XIII**) is the product of reduction of cation **Va** with the initial alcohol; the second product of this reaction, 4-dimethylamino-1-naphthalenecarbaldehyde, was not detected; presumably, it underwent oligomerization on heating in trifluoroacetic acid. In

Scheme 2.
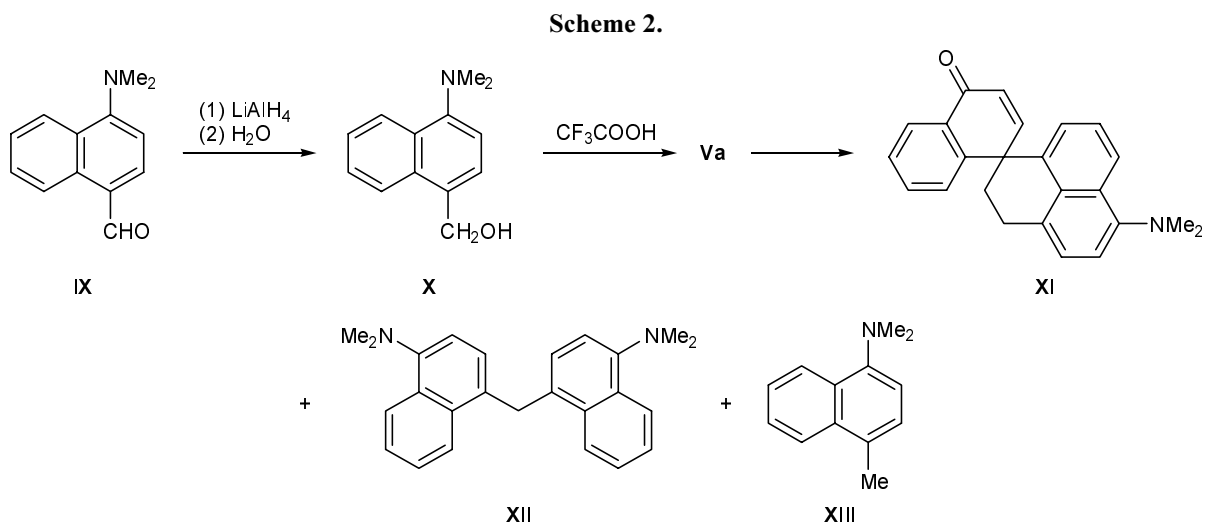
the reaction of alcohol **X** with anhydrous aluminum oxide we succeeded in isolating only two compounds, dinaphthylmethane **XII** (66%) and 1-dimethylamino-4-methylnaphthalene (**XIII**, 7%). No spiro compounds were detected in the reaction mixture.

The poor yield of spiro compound **XI** and the high yield of dinaphthylmethane **XII** are likely to be determined by weak resonance stabilization of cation **Va** and lower basicity of amino alcohol **X**, as compared, e.g., with amino alcohols **Ia** and **Ib**. We also found that even less basic 4-methoxy-1-naphthylmethanol (**Vb**) almost did not change on heating with Al$_2$O$_3$ in benzene; treatment of the same substrate with trifluoroacetic acid gave a mixture of oligomeric products.

Cation **VI** in which the donor substituent and the cationic center reside in different benzene rings was generated from amino alcohol **XV**. The latter was synthesized in 41% yield by reduction of 1-hydroxymethyl-5-nitronaphthalene (**XIV**) [12] with hydrogen in methanol in the presence of 2% Pd/C. In addition, 21% of 1-amino-5-methylnaphthalene (**XVI**) was isolated (Scheme 3).
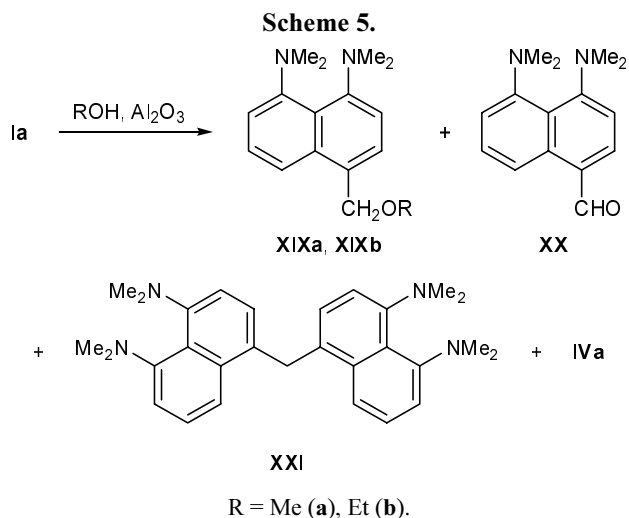
Scheme 3.
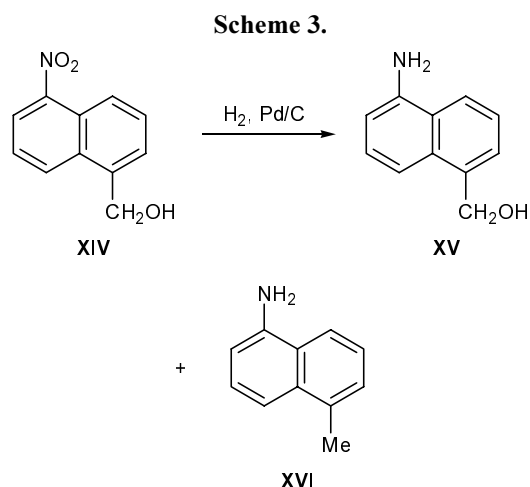
We failed to effect selective methylation of amino alcohol **XV** at the amino group. The reaction with methyl iodide in neutral medium resulted in formation of a complex mixture of products, while in THF in the presence of sodium hydride exhaustive methylation to 1-dimethylamino-5-methoxymethylnaphthalene (**XVII**) smoothly occurred (Scheme 4). We presumed that compound **XVII** can also be used as precursor of carbocation **VI** in the presence of acid agents. However, by heating ether **XVII** in boiling concentrated hydrobromic acid we obtained 72% of bromomethyl derivative **XVIII** as the only product; this result is
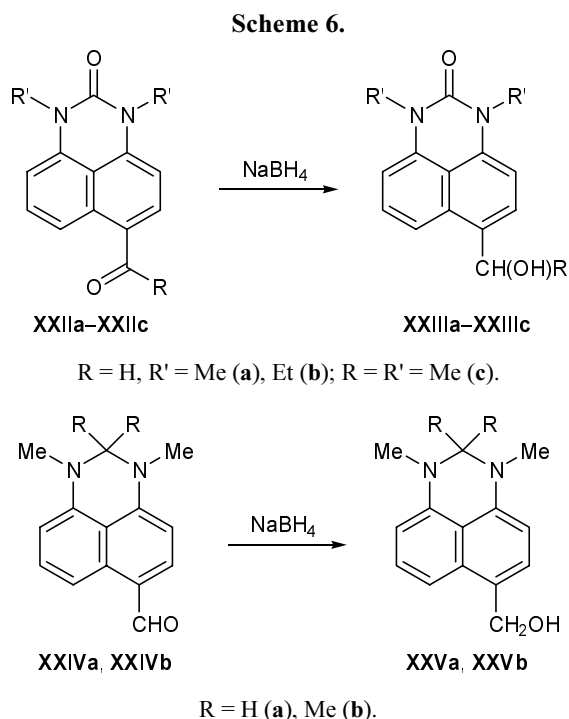
Scheme 4.
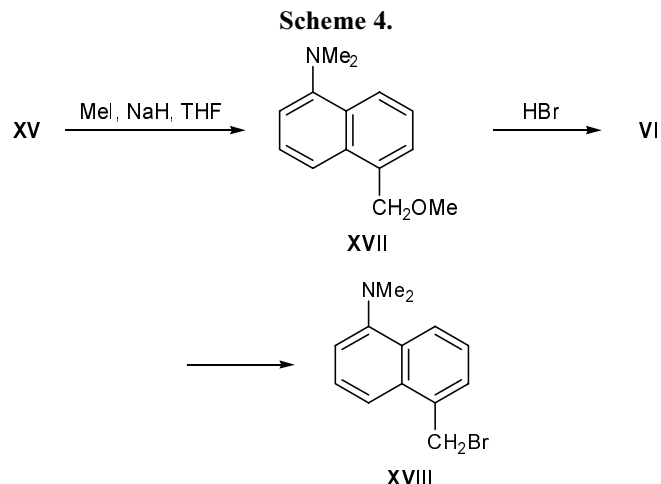
likely to indicate insufficient stabilization of carbocation **VI**.

4-Alkoxymethyl proton sponge derivatives **XIXa** and **XIXb** reacted in a different fashion. These compounds were prepared by treatment of alcohol **Ia** with methanol or ethanol in the presence of Al$_2$O$_3$. In both cases aldehyde **XX** (~6%), dinaphthylmethane derivative **XXI** (~15%), and spiro compound **IVa** (~5%) were isolated as by-products (Scheme 5). Their formation suggests participation of carbocation **IIa** as common intermediate. The structure of compound **XXI** was proved by the X-ray diffraction data (see figure). When ethers **XIX** were heated with concentrated hydrobromic acid for a short time, spiro compound **IIIa** was formed in a good yield; thus ethers like **XIX** may be regarded as convenient precursors of 1-naphthylmetyl carbocations.

Different results were obtained with carbocations of the perimidinone and 2,3-dihydroperimidine series.

Scheme 5.

R = Me (**a**), Et (**b**).

These cations were generated from alcohols **XXIII** and **XXV** which were prepared by reduction of aldehydes **XXIIa**, **XXIIb**, **XXIVa**, and **XXIVb** or ketone **XXIIc** with sodium tetrahydridoborate (Scheme 6).

Scheme 6.

XXIIa–XXIIc   XXIIIa–XXIIIc

R = H, R' = Me (**a**), Et (**b**); R = R' = Me (**c**).

XXIVa, XXIVb   XXVa, XXVb

R = H (**a**), Me (**b**).

Perimidine alcohol **XXIIIa** quickly reacted with concentrated hydrochloric acid or with trifluoroacetic acid even in the cold, and the reaction mixture acquired an intense red color. According to the IR and ^{1}H NMR data, the reaction gave mainly a complex mixture of oligomeric products. In addition, we isolated 15% of aldehyde **XXIIa** and 10% of a yellow substance possessing an aldehyde (δ 10.22 ppm) and methylene group (δ 4.50 ppm), as well as four non-equivalent methyl groups. These data allowed us to assign the structure of 7-formyl-6,6'-diperimidinyl-methane **XXVI** to that product (Scheme 7). Obviously, aldehyde **XXIIa** is formed by dehydration of initial alcohol **XXIIIa** with intermediate cation **VIIa**. Compound **XXVI** is likely to arise from electrophilic attack
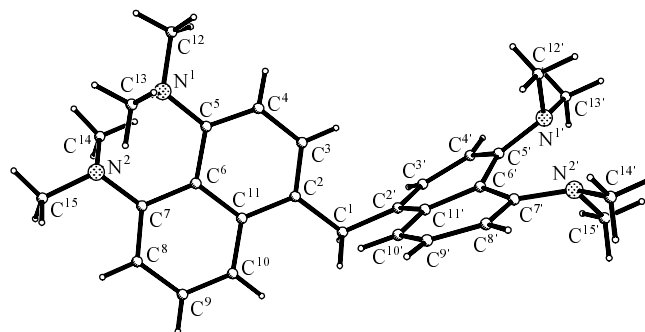

Structure of the molecule of bis[4,5-bis(dimethylamino)-naphthalen-l-yl]methane (**XXI**) according to the X-ray diffraction data.

by carbocation **VIIa** on the 7-position of molecule **XXIIIa**, followed by dehydrogenation of the CH$_2$OH group. On the whole, unusually high concentration of aldehydes among the products obtained by the action of protic acids on alcohol **XXIIIa** may be rationalized in terms of considerable deactivation of the naphthalene ring in **XXIIIa**, as well as in the intermediate products, by the amide carbonyl group. Therefore, the contribution of dehydrogenation of the CH$_2$OH group with the carbocation becomes more significant. By contrast, 6-methylperimidinone thus formed should be more reactive toward electrophiles, as compared to the other compounds present in the reaction mixture, due to donor effect of the 6-methyl group, and it should readily undergo oligomerization. This is the reason why 1,3,6-trimethylperimidinone was never detected among the products. Treatment of alcohols **XXIIIb** and **XXIIIc** with protic acid afforded mainly oligomeric products, and those derived from compound **XXIIIc** contained (as might be expected [13]) vinyl groups. All perimidine alcohols **XXIIIa–XXIIIc** remained intact on prolonged heating in benzene in the presence of Al$_2$O$_3$. This means that no appreciable amounts of the corresponding carbocations were formed owing to their weak resonance stabilization.

Alcohols **XXV** derived from 2,3-dihydroperimidines turned out to be even more reactive. These compounds are unstable, and they decomposed in several

Scheme 7.

XXIIIa $\xrightarrow{\text{HCl}}$ ...   +   XXIIa

XXVI

Scheme 8.
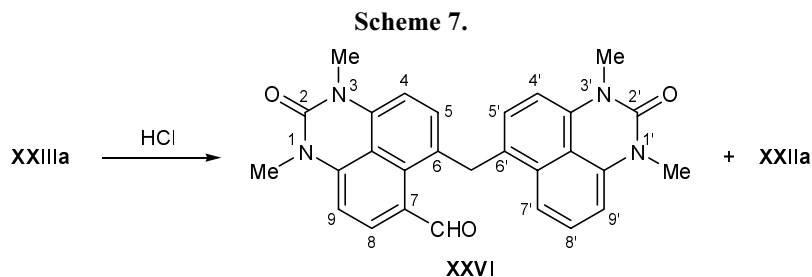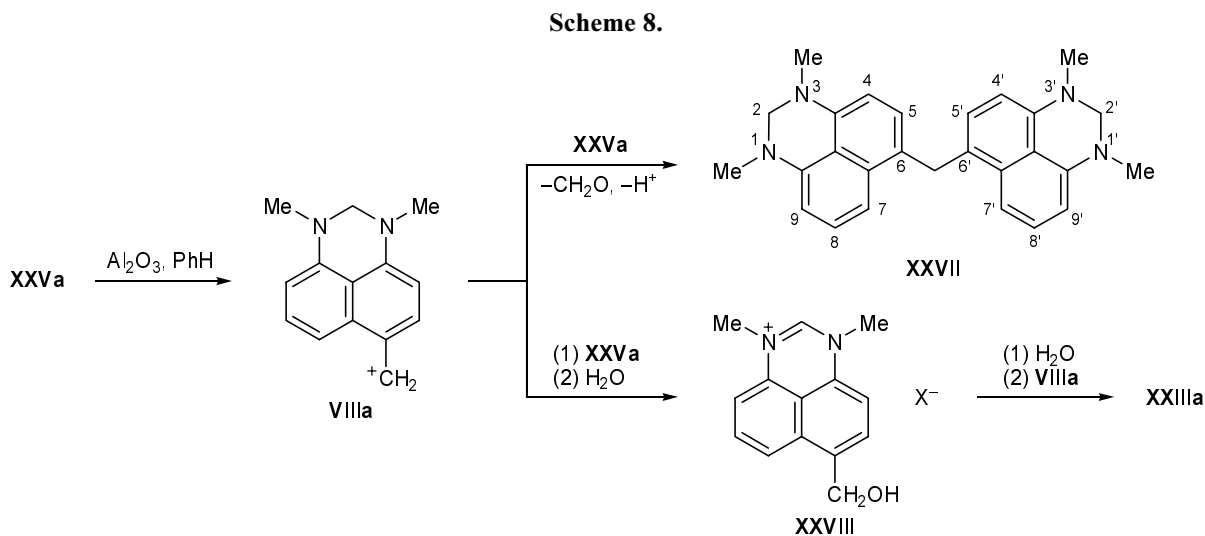
days even under normal conditions. Alcohols **XXV** readily gave rise to carbocationic species on treatment with both protic and Lewis acids; the carbocationic center therein is stabilized by the amine rather than amide nitrogen atoms. Alcohols **XXVa** and **XXVb** underwent complete oligomerization by the action of strong protic acids such as concentrated hydrochloric acid and trifluoroacetic acid. When a solution of alcohol **XXVa** was heated under reflux in the presence of Al$_2$O$_3$, the products were dinaphthylmethane **XXVII** (22%), perimidine alcohol **XXIIIa** (6.6%), and a mixture of perimidinium salts **XXVIII** (Scheme 8). The scheme of formation of compound **XXVII** is likely to be similar to that proposed for dinaphthylmethane **XII**. Undoubtedly, compounds **XXIIIa** and **XXVIII** originate from 2,3-dihydroperimidines due to their known ability to undergo dehydrogenation to perimidinium salts by the action of various electron acceptors [14]. In our case, carbocation **VIIIa** is most likely to act as such an acceptor. Compound **XXIIIa** may be formed as a result of covalent hydration of salt **XXVIII** to give carbinol pseudobase which then also undergoes dehydrogenation.

Unlike **XXVa**, alcohol **XXVb** has no hydrogen atoms capable of being abstracted as hydride ion; pre-

sumably, this is the reason for its different behavior. On heating in benzene in the presence of Al$_2$O$_3$, compound **XXVb** gave rise to ~7% of spiro derivative **XXIX** in addition to oligomeric products and aldehyde **XXIVb** (yield 15%) (Scheme 9). The structure of unstable compound **XXIX** is beyond doubt, for its ^{1}H NMR spectrum is analogous to the spectra of symmetric spiro compounds **IV**, which contain two doublets at δ 6.1 and 6.7 ppm from the 3'-H and 2'-H protons with a characteristic quinoid coupling constant *J* of 10.2 Hz. Two sets of enantiotopic protons (H$_a$ and H$_b$) which are diastereotopic with respect to each other appeared in the ^{1}H NMR spectrum as two two-proton doublets at δ 3.7 and 2.9 ppm (^2J = 15.5 Hz).

Thus the results of our study demonstrated that the behavior of 1-naphthylmethyl carbocations stabilized by one electron-donor group or *peri*-fused heteroring considerably differs from the behavior of naphthalene proton sponges. The former are characterized by a stronger tendency to undergo oligomerization and form dinaphthylmethane structures and aldehydes and by a sharply reduced contribution of [4+2]-cycloaddition process. An obvious reason is enhanced resonance stabilization of 4,5-bis(dialkylamino)-1-naphthyl-methyl carbocations in combination with high basicity

Scheme 9.
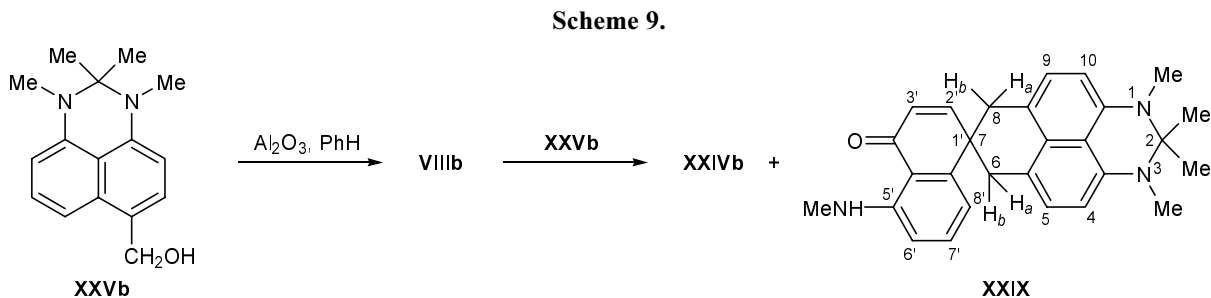
of proton sponges. In the latter case, the presence of appreciable amounts of unprotonated initial alcohol in acid medium is ruled out; as a result, side processes involving proton sponges, primarily oligomerization, are minimized.

EXPERIMENTAL

The UV spectrum of compound **XI** was measured on a Specord M40 spectrophotometer. The IR spectra were recorded on a UR-20 instrument. The ^{1}H NMR spectra of all compounds, except for **IX**, were obtained on a Varian Unity 300 spectrometer; the ^{1}H NMR spectrum of **IX** was obtained on a Bruker 250 instrument; tetramethylsilane was used as internal reference. The mass spectra were run on an MKh-1321A mass spectrometer. Aluminum oxide for chromatography (Brockmann activity grade II) was used as Lewis acid catalyst; it was calcined for 20 min at 250°C prior to use.

Crystals of **XXI** suitable for X-ray analysis were obtained by slow evaporation of its solution in heptane at –5°C. Orthorhombic crystals with the following unit cell parameters: $a = 10.696(2)$, $b = 11.098(2)$, $c = 21.166(4)$ Å; $V = 2512.5(9)$ Å^3; $Z = 4$; $d_{calc} = 1.165$ g × cm^{-3}; space group $P2_12_12_1$; $F(000) = 952$; $\mu = 0.069$ cm^{-1}. The data were acquired at 293(2) K using an Enraf–Nonius CAD4 diffractometer (wavelength 0.71073 Å; θ/5/3θ scanning, θ_{max} 1.92–26.98 deg); total of 3242 reflections were measured, 3096 of which were independent ($R_{int} = 0.0589$); number of independent reflections with $I \geq 2\sigma(I)$ 1404; $R_1 = 0.0502$ [with respect to F for reflections with $I \geq 2\sigma(I)$], $wR_2 = 0.1716$ (with respect to F^2 for all reflections); number of refined parameters 298, GOOF 1.007. The positions of all atoms were determined by the direct method. The structure was refined by the least-squares procedure in anisotropic approximation for non-hydrogen atoms and isotropic approximation for hydrogen atoms. All calculations were performed using SHELXTL PLUS 5 software package. The coordinates of atoms and the complete set of crystallographic parameters were deposited to the Cambridge Crystallographic Data Centre (entry no. CCDC 258392).

4-Dimethylaminonaphthalene-1-carbaldehyde (IX). A solution of 6.84 g (0.04 mol) of 1-dimethylaminonaphthalene in 50 ml of anhydrous toluene was cooled to 0°C, and Vilsmeier's reagent prepared from 3.7 ml (0.04 mol) of freshly distilled phosphoryl chloride and 20 ml of anhydrous dimethylformamide at –15°C was added dropwise. The mixture was stirred

for 1 h at 30°C and for 1 h at 80°C, cooled, poured into 200 ml of cold water, and adjusted to pH ~8–9 by adding a 10% aqueous solution of sodium hydroxide. The organic phase was separated, and the aqueous phase was extracted with chloroform (3×25 ml). The extracts were combined with the organic phase and evaporated, and the residue was subjected to column chromatography on Al$_2$O$_3$ (Brockmann activity grade IV; 2.5×25 cm) using hexane–toluene (3:1) as eluent. The first light yellow fraction was collected. Yield 5.3 g (67%). Light yellow crystals, mp 41–42°C (from hexane); published data [4]: mp 43.5°C. IR spectrum (CCl$_4$), ν, cm^{-1}: 1685 (C=O), 1580 (C=C$_{arom}$). ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.99 s (6H, NMe$_2$), 6.96 d and 7.78 d (1H each, 2-H, 3-H, $J_{2,3}$ = 8.0 Hz), 7.58 m (2H, 6-H, 7-H), 8.17 m and 9.35 m (1H each, 5-H, 8-H), 10.14 s (1H, CHO). Found, %: C 78.10; H 6.56; N 6.93. C$_{13}$H$_{13}$NO. Calculated, %: C 78.35; H 6.58; N 7.03.

4-Dimethylaminonaphthalen-1-ylmethanol (X). 4-Dimethylaminonaphthalene-1-carbaldehyde (**IX**), 1.00 g (5.02 mmol), was dissolved in 50 ml of anhydrous diethyl ether, and 0.20 g (5.27 mmol) of lithium tetrahydridoaluminate was added. The mixture was stirred at room temperature until it became colorless, and 5 ml of water was added. The ether layer was separated and evaporated, and the residue was subjected to column chromatography on Al$_2$O$_3$ (Brockmann activity grade IV; 2.5×10 cm) using chloroform as eluent. A colorless fraction with R_f 0.54 was collected. Yield 0.96 g (95%), dark brown material. ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 1.95 br.s (1H, OH, disappeared on addition of D$_2$O), 2.87 s (6H, NMe$_2$), 5.04 s (2H, CH$_2$), 6.99 d and 7.38 d (1H each, 2-H, 3-H, $J_{2,3}$ = 7.6 Hz), 7.52 m (2H, 6-H, 7-H), 8.12 m and 8.27 m (1H each, 5-H, 8-H). Found, %: C 77.44; H 7.52; N 6.88. C$_{13}$H$_{15}$NO. Calculated, %: C 77.57; H 7.52; N 6.96.

Transformations of 4-dimethylaminonaphthalen-1-ylmethanol (X). *a.* A solution of 201 mg (1 mmol) of alcohol **X** in 5 ml of freshly distilled trifluoroacetic acid was heated for 15 h under reflux. The mixture was cooled, poured into 50 ml of water, adjusted to pH ~12, and extracted with chloroform (3×10 ml). The extracts were combined and evaporated, and the residue was subjected to column chromatography on Al$_2$O$_3$ (Brockmann activity grade II; 2× 25 cm) using hexane–chloroform (3:1) as eluent. From the first colorless fraction with R_f 0.50 we isolated 11 mg (6%) of *N*,*N*,4-trimethylnaphthalen-1-amine

(**XIII**) as a colorless oily substance which was dried in a vacuum desiccator over potassium carbonate. ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.64 d (3H, CH$_3$, J = 0.8 Hz), 2.88 s (6H, NMe$_2$), 7.00 d (1H, 2-H, $J_{2,3}$ = 7.5 Hz), 7.24 d.d (1H, 3-H, $J_{3,2}$ = 7.5, J = 0.8 Hz), 7.51 m (2H, 6-H, 7-H), 7.97 m and 8.29 m (1H each, 5-H, 8-H). Found, %: C 84.01; H 8.17; N 7.48. C$_{13}$H$_{15}$N. Calculated, %: C 84.27; H 8.17; N 7.56. From the second fraction (R_f 0.28) we isolated 101 mg (57%) of 1,1'-bis(dimethylamino)-4,4'-dinaphthyl-methane (**XII**) as colorless crystals with mp 180–181°C (from heptane); published data [15]: mp 181–182.5°C [15]). ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.89 s (12H, NMe$_2$), 4.76 s (2H, CH$_2$), 6.95 d and 7.00 d (2H each, 2-H, 2'-H, 3-H, 3'-H, $J_{2,3}$ = 7.7 Hz), 7.50 m (4H, 6-H, 6'-H, 7-H, 7'-H), 8.04 m and 8.35 m (2H each, 5-H, 5'-H, 8-H, 8'-H). Found, %: C 84.39; H 7.40; N 7.86. C$_{25}$H$_{26}$N$_2$. Calculated, %: C 84.70; H 7.40; N 7.91. From the fraction with R_f 0.09 we isolated 15 mg (9%) of 6-dimethylamino-1',2,3,4'-tetrahydrospiro[phenalene-1,1'-naphthalen]-4'-one (**XI**) as a slightly colored amorphous powder which was recrystallized from heptane, mp 61–63°C. UV spectrum (MeOH): λ$_{max}$ 316 nm (logε 4.02). IR spectrum (CCl$_4$): ν(C=O) 1670 cm^{-1}. ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.32 m (2H, 2-CH$_2$), 2.91 s (6H, NMe$_2$), 3.25 m (2H, 3-CH$_2$), 6.46 d (1H, 3'-H, $J_{3',2'}$ = 10.2 Hz), 6.90 d.d (1H, 9-H, $J_{9,8}$ = 7.2, $J_{9,7}$ = 1.2 Hz), 7.05 m (1H, 8'-H), 7.14 d and 7.34 d (1H each, 4-H, 5-H, $J_{4,5}$ = 7.6 Hz), 7.26 d (1H, 2'-H, $J_{2',3'}$ = 10.2), 7.31 d.d (1H, 8-H, $J_{8,9}$ = 7.2, $J_{8,7}$ = 8.5 Hz), 7.41 m (2H, 6'-H, 7'-H), 8.23 d.d (1H, 7-H, $J_{7,8}$ = 8.5, $J_{7,9}$ = 1.2 Hz), 8.27 m (1H, 5-H). Mass spectrum: *m/z* 339 [*M*]$^+$ (I_{rel} 100%). Found, %: C 84.77; H 6.23; N 4.01. C$_{24}$H$_{21}$NO. Calculated, %: C 84.92; H 6.24; N 4.13.

b. A mixture of 201 mg (1 mmol) of compound **X**, 30 ml of benzene, and 5 g of anhydrous Al$_2$O$_3$ was shaken for 12 h at 75–80°C. The mixture was cooled, aluminum oxide was filtered off and washed on a filter with chloroform (3×10 ml), the filtrate was evaporated, and the residue was subjected to column chromatography on Al$_2$O$_3$ (Brockmann activity grade II; 2×25 cm) using hexane–chloroform (3:1) as eluent. A fraction with R_f 0.50 containing *N,N*,4-trimethylnaphthalen-1-amine (**XIII**) was collected first. Yield 13 mg (7%). From a fraction with R_f 0.28 we isolated 1,1'-bis(dimethylamino)-4,4'-dinaphthylmethane (**XII**), yield 117 mg (66%).

Reduction of 5-nitronaphthalen-1-ylmethanol (XIV). A mixture of 100 mg (0.5 mmol) of compound

XIV [12] in 25 ml of methanol and 100 mg of 2% Pd/C was shaken in a standard apparatus for catalytic hydrogenation under atmospheric pressure. After 1 h, the catalyst was filtered off and washed with a small amount of methanol, the filtrate was evaporated under reduced pressure, and the residue was subjected to column chromatography on Al$_2$O$_3$ (Brockmann activity grade V; 1.5×6 cm) using chloroform as eluent. From the first fraction (R_f 0.91) we isolated 16 mg (21%) of 5-methylnaphthalen-1-amine (**XVI**) as pink crystals with mp 76–77°C (from ethanol) [16]. IR spectrum (mineral oil), ν, cm^{-1}: 3315, 3228 (NH$_2$); 1590 (C=C$_{arom}$). ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.66 s (3H, CH$_3$), 4.1 br.s (2H, NH$_2$, disappeared on addition of D$_2$O), 6.79 d (1H, 2-H, $J_{2,3}$ = 7.36 Hz), 7.30 m (3H, 3-H, 6-H, 7-H), 7.45 br.d and 7.69 br.d (1H each, 4-H, 8-H, $J_{4,3}$ = 8.46, $J_{8,7}$ = 8.02 Hz). Found, %: C 83.89; H 7.04; N 8.77. C$_{11}$H$_{11}$N. Calculated, %: C 84.03; H 7.06; N 8.91. The second fraction (R_f 0.15) contained 5-aminonaphthalen-1-ylmethanol (**XV**). Yield 35 mg (41%), pale yellow crystals, mp 104–105°C (from benzene). IR spectrum (mineral oil), ν, cm^{-1}: 3370, 3120–3350 (OH, NH$_2$); 1590 (C=C$_{arom}$). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 4.86 d (2H, CH$_2$), 5.19 t (1H, OH, disappeared on addition of D$_2$O), 5.67 br.s (2H, NH$_2$, disappeared on addition of D$_2$O), 6.66 d.d (1H, 2-H, $J_{2,3}$ = 6.12, $J_{2,4}$ = 2.4 Hz), 7.19 m (2H, 3-H, 6-H), 7.31 m (1H, 7-H), 7.46 br.d and 7.96 br.d (1H each, 4-H, 8-H, $J_{4,3}$ = 6.9, $J_{8,7}$ = 8.4 Hz). Found, %: C 75.94; H 6.40; N 7.88. C$_{11}$H$_{11}$NO. Calculated, %: C 76.26; H 6.41; N 8.09.

5-Methoxymethyl-*N,N*-dimethylnaphthalen-1-amine (XVII). Sodium hydride, 110 mg (4.6 mmol), was added under stirring to a solution of 250 mg (1.44 mmol) of compound **XV** in 5 ml of anhydrous tetrahydrofuran, and 0.4 ml (6.4 mmol) of freshly distilled methyl iodide was then added. The mixture was stirred for 2.5 h at room temperature, 2 ml of water was added, and the organic phase was separated and evaporated. The residue was subjected to column chromatography on Al$_2$O$_3$ (Brockmann activity grade III; 2.5×10 cm) using hexane–chloroform (2:1) as eluent. A fraction with R_f 0.45 was collected. Yield of **XVII** 267 mg (86%), pale yellow material. ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.88 s (6H, NMe$_2$), 3.45 s (3H, OMe), 4.88 s (2H, CH$_2$), 7.10 br.d and 7.77 br.d (1H each, 2-H, 4-H, $J_{2,3}$ = 7.5, $J_{4,3}$ = 8.5 Hz), 7.44 m (3H, 3-H, 6-H, 7-H), 8.22 d.d (1H, 8-H, $J_{8,7}$ = 7.91, $J_{8,6}$ = 1.7 Hz). Found, %: C 77.84; H 7.94; N 6.45. C$_{14}$H$_{17}$NO. Calculated, %: C 78.09; H 7.96; N 6.51.

5-Bromomethyl-*N,N*-dimethylnaphthalen-1-amine (XVIII). A solution of 130 mg (0.6 mmol) of compound **XVII** in 2 ml of concentrated hydrobromic acid was heated for 20 min at the boiling point. The mixture was cooled, diluted with an equal volume of water, made alkaline by adding 20% aqueous sodium hydroxide to pH ~12, and extracted with benzene (3× 5 ml). The extracts were combined and evaporated to obtain 149 mg (93%) of compound **XVIII** as a light yellow amorphous powder, mp 73–74°C. ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.87 s (6H, NMe$_2$), 4.95 s (2H, CH$_2$), 7.12 d.d (1H, 2-H, $J_{2,3}$ = 7.5, $J_{2,4}$ = 0.9 Hz), 7.39 t (1H, 7-H, $J_{7,6}$ = 8.5, $J_{7,8}$ = 8.5 Hz), 7.52 m (2H, 3-H, 4-H), 7.82 d and 8.28 d (1H each, 8-H, 6-H, $J_{8,7}$ = 8.5, $J_{6,7}$ = 8.5 Hz). Mass spectrum, m/z (I_{rel}, %): 265 (23) [M]$^+$, 263 (24). Found, %: C 58.96; H 5.32; N 5.19. C$_{13}$H$_{14}$BrN. Calculated, %: C 59.11; H 5.34; N 5.30.

Transformations of 4,5-bis(dimethylamino)-naphthalen-1-ylmethanol (Ia) over Al$_2$O$_3$. *a*. A mixture of 300 mg (1.23 mmol) of compound **Ia** [7], 4 ml of anhydrous methanol, and 7 g of calcined Al$_2$O$_3$ was kept for 35 h at 80°C in a sealed ampule. The ampule was cooled and opened, the mixture was filtered, the precipitate was washed with a large amount of methanol, the filtrate was combined with the washings and evaporated, and the residue was subjected to column chromatography on Al$_2$O$_3$ (Brockmann activity grade V; 2×25 cm) using chloroform as eluent. From the first fraction (R_f 0.82) we isolated 19 mg (7%) of 4,5-bis(dimethylamino)naphthalene-1-carbaldehyde (**XX**) [17]. The second fraction contained 4-methoxy-methyl-*N,N,N′,N′*-tetramethylnaphthalene-1,8-diamine which was isolated as a pale yellow material. Yield 210 mg (66%). ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.80 s (12H, NMe$_2$); 3.44 s (3H, OCH$_3$); 4.77 s (2H, CH$_2$); 6.85 d, 6.96 d, 7.30 d, and 7.62 d (1H each, 2-H, 7-H, 3-H, 5-H, $J_{2,3}$ = 7.9, $J_{7,6}$ = 7.3, $J_{5,6}$ = 8.2 Hz); 7.37 t (1H, 6-H, $J_{6,7}$ = 7.9, $J_{6,5}$ = 7.9 Hz). Found, %: C 73.93; H 8.51; N 10.64. C$_{16}$H$_{22}$N$_2$O. Calculated, %: C 74.37; H 8.59; N 10.85.

The third fraction (R_f 0.10) contained 12 mg (5%) of 6,7,5′-tris(dimethylamino)-1,1′,3,4′-tetrahydrospiro[phenalene-2,1′-naphthalen]-4′-one (**IVa**) [7], and the fourth (R_f 0.05), 43 mg (16%) of bis[4,5-bis(dimethylamino)naphthalen-1-yl]methane (**XXI**) (colorless crystals, mp 166–167°C [10]).

b. The reaction was carried out as described above in *a* using 4 ml of anhydrous ethanol as solvent. We isolated 5% of aldehyde **XX**, 57% of ether **XIXb**

(R_f 0.29), 4% of spiro compound **IVa**, and 13% of compound **XXI**.

4-Ethoxymethyl-*N,N,N′,N′*-tetramethylnaphthalene-1,8-diamine (XIXb). Pale yellow material. ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 1.27 t (3H, CH$_2$C**H**$_3$), 2.78 br.s (12H, NMe$_2$), 3.61 q (2H, C**H**$_2$CH$_3$), 4.8 s (2H, CH$_2$), 6.84 d and 7.30 d (1H each, 2-H, 3-H, $J_{2,3}$ = 7.7 Hz), 6.94 d.d (1H, 7-H, $J_{7,6}$ = 7.5, $J_{7,5}$ = 1.1 Hz), 7.36 d.d and 7.63 d.d (1H each, 5-H, 6-H, $J_{6,7}$ = 7.6, $J_{6,5}$ = 8.0, $J_{5,6}$ = 8.35, $J_{5,7}$ = 1.2 Hz). Found, %: C 74.81; H 8.87; N 10.15. C$_{17}$H$_{24}$N$_2$O. Calculated, %: C 74.95; H 8.89; N 10.29.

6,7,5′-Tris(dimethylamino)-1,1′,3,4′-tetrahydro-spiro[phenalene-1,1′-naphthalen]-4′-one (IIIa). *a*. A solution of 340 mg (1.32 mmol) of compound **XIXa** in 5 ml of concentrated hydrobromic acid was heated for 20 min at the boiling point. The mixture was cooled, diluted with an equal amount of water, neutralized with 20% aqueous NaOH to pH ~7, and extracted with chloroform (3×10 ml). The combined extracts were evaporated to a volume of 5 ml and applied to a column charged with Al$_2$O$_3$ (Brockmann activity grade I; 1.5×15 cm); the column was eluted with chloroform to collect a fraction with R_f 0.06. Yield of **IIIa** 231 mg (82%) [7, 8].

b. The procedure was the same as in *a*, but compound **XIXb** was used as starting material. Yield of **IIIa** 78%.

1,3-Dimethyl-6-hydroxymethylperimidin-2-one (XXIIIa). 1,3-Dimethyl-2-oxoperimidine-6-carbaldehyde **XXIIa** [18], 0.48 g (2 mmol), was dispersed in 45 ml of ethanol, and 0.12 g (6 mmol) of finely powdered sodium tetrahydridoborate was added in one portion under stirring. The mixture was heated for 30 min at 60–70°C, cooled, and poured into 100 ml of water. The precipitate was filtered off, washed with 30 ml of water, and dried in air. Yield 0.44 g (91%). Grey crystals, mp 212–216°C (from toluene). IR spectrum (mineral oil), ν, cm^{-1}: 3410 (OH); 1650 (C=O); 1625, 1589 (C=C). ^{1}H NMR spectrum, δ, ppm: in DMSO-d_6: 3.30 s and 3.32 s (3H each, CH$_3$), 4.72 d (2H, C**H**$_2$OH, J = 5.3 Hz), 5.12 t (1H, CH$_2$O**H**, J = 5.3 Hz), 6.65 d (1H, 4-H, $J_{4,5}$ = 7.9 Hz), 6.73 d.d (1H, 9-H, $J_{9,8}$ = 5.2, $J_{9,7}$ = 3.2 Hz), 7.44 m (3H, 5-H, 7-H, 8-H); in CDCl$_3$: 2.22 t (1H, CH$_2$O**H**, J = 5.5 Hz); 3.23 s and 3.40 s (3H each, CH$_3$); 4.94 d (2H, C**H**$_2$OH, J = 5.5 Hz); 6.47 d and 7.36 d (1H each, 4-H, 5-H, $J_{4,5}$ = 7.8 Hz); 6.60 d.d, 7.46 d.d, and 7.61 d.d (1H each, 9-H, 8-H, 7-H, $J_{9,8}$ = 7.6, $J_{9,7}$ = 0.9, $J_{8,7}$ =

8.6 Hz). Found, %: C 69.41; H 5.90; N 11.34. $C_{14}H_{14}N_2O_2$. Calculated, %: C 69.39; H 5.83; N 11.57.

1,3-Diethyl-6-hydroxymethylperimidin-2-one (XXIIIb) was synthesized as described above for compound **XXIIIa** from 0.2 g (0.83 mmol) of **XXIIb** [18], 10 ml of ethanol, and 0.07 g (3.5 mmol) of NaBH$_4$. Yield 0.16 g (80%), colorless crystals, mp 173–175°C (from toluene). IR spectrum (mineral oil), ν, cm^{-1}: 3449 (OH); 1652 (C=O); 1625, 1589 (C=C). ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 1.22 t and 1.28 t (3H each, CH$_2$C**H**$_3$); 2.05 br.s (1H, OH); 3.88 q and 4.00 q (2H each, C**H**$_2$CH$_3$); 4.90 s (2H, C**H**$_2$OH); 6.42 d, 6.62 d, 7.32 d, and 7.50 d (1H each, 4-H, 9-H, 5-H, 7-H, $J_{4,5}$ = 7.91, $J_{9,8}$ = 7.62, $J_{7,8}$ = 8.50 Hz); 7.40 d.d (1H, 8-H, $J_{8,7}$ = 8.50, $J_{8,9}$ = 7.62 Hz). Found, %: C 71.15; H 6.90; N 10.45. $C_{16}H_{18}N_2O_2$. Calculated, %: C 71.08; H 6.72; N 10.37.

1,3-Dimethyl-6-(1-hydroxyethyl)perimidin-2-one (XXIIIc). 6-Acetyl-1,3-dimethylperimidin-2-one [19], 0.25 g (1 mmol), was dissolved on heating in a minimal amount of ethanol (~20 ml), the solution was cooled, 0.25 g (7 mmol) of NaBH$_4$ was added, and the mixture was stirred for 30 min, maintaining it slightly boiling. The mixture was then diluted with water (~60 ml), and the precipitate was filtered off, washed with water, and dried in air. Yield 0.23 g (90%), colorless crystals, mp 222–223°C (from ethanol), R_f 0.56 (Silufol, ethyl acetate). ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 1.61 d (3H, C**H**$_3$CH, J = 6.6 Hz); 2.51 br.s (1H, OH); 3.20 s and 3.38 s (3H each, CH$_3$); 5.42 q (1H, CH$_3$C**H**, J = 6.6 Hz); 6.39 d, 6.54 d, 7.53 d, and 7.55 d (1H each, 4-H, 9-H, 5-H, 7-H, $J_{4,5}$ = 8.24, $J_{9,8}$ = 7.70, $J_{7,8}$ = 8.25 Hz); 7.41 d.d (1H, 8-H, $J_{8,9}$ = 7.70, $J_{8,7}$ = 8.25 Hz). Found, %: C 70.31; H 6.39; N 10.67. $C_{15}H_{16}N_2O_2$. Calculated, %: C 70.29; H 6.29; N 10.93.

1,3-Dimethyl-2,3-dihydro-1*H*-perimidin-6-ylmethanol (XXVa). *a*. Compound **XXIIIa**, 0.48 g (2 mmol), was dissolved in 200 ml of anhydrous THF, and 0.3 g (8 mmol) of LiAlH$_4$ was added in three portions over a period of 1.5 h on heating under stirring. The mixture was then heated for 1.5 h under reflux, cooled, treated with 3 ml of water, and filtered, and the precipitate was washed on a filter with 50 ml of THF. The filtrate was combined with the washings and evaporated, the residue was dissolved in 15 ml of chloroform, and the solution was passed through a column charged with Al$_2$O$_3$ (1.5×8 cm) with suction and protection from light. Yield 0.35 g (78%), yellow crystals, mp 212–217°C, R_f 0.3. Compound **XXVa** is quite unstable, and it should be stored in a refrigerator.

IR spectrum (mineral oil), ν, cm^{-1}: 3400 (OH); 1625, 1568 (C=C). ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 2.98 s and 2.99 s (3H each, CH$_3$); 4.23 s (2H, 2-CH$_2$); 4.96 s (2H, C**H**$_2$OH); 6.45 d and 7.34 d (1H each, 4-H, 5-H, $J_{4,5}$ = 7.8 Hz); 6.58 d.d, 7.42 d.d, and 7.50 d.d (1H each, 9-H, 8-H, 7-H, $J_{9,8}$ = 7.5, $J_{9,7}$ = 0.9, $J_{8,7}$ = 8.4 Hz). Found, %: C 73.95; H 7.20; N 12.00. $C_{14}H_{16}N_2O$. Calculated, %: C 73.64; H 7.07; N 12.28.

b. Compound **XXIIa**, 0.24 g (1 mmol), was dissolved in 120 ml of anhydrous THF, and 0.21 g (5.5 mmol) of LiAlH$_4$ was added to the solution in three equal portions over a period of 1.5 h on heating under stirring. The mixture was heated for 2 h under reflux, cooled, treated with 3 ml of water, and filtered, the precipitate was washed with 50 ml of THF, and the filtrate was evaporated to obtain 0.2 g of a dark material which was subjected to column chromatography on Al$_2$O$_3$ (2×15 cm). The column was eluted with chloroform under reduced pressure. Yield 0.1 g (50%).

1,2,2,3-Tetramethyl-2,3-dihydro-1*H*-perimidin-6-ylmethanol (XXVb). Aldehyde **XXIVb** [18], 0.254 g (1 mmol), was dissolved in 15 ml of anhydrous THF, 0.08 g (2 mmol) of LiAlH$_4$ was added, the mixture was shaken for 5 min, treated with 2 ml of water, and filtered, the precipitate was washed with a small amount of THF, and the filtrate was evaporated. The residue was kept under reduced pressure to obtain a light yellow thick material. Yield 0.25 g (100%). The product was very unstable, and its elemental analysis was not performed. ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 1.39 s and 2.93 s (6H each, CH$_3$); 4.96 s (2H, C**H**$_2$OH); 6.51 d and 7.32 d (1H each, 4-H, 5-H, $J_{4,5}$ = 7.8 Hz); 6.64 d.d, 7.40 d.d, and 7.49 d.d (3H, 9-H, 8-H, 7-H, $J_{9,8}$ = 7.4, $J_{9,7}$ = 0.9, $J_{8,7}$ = 8.4 Hz).

Transformations of 1,3-dimethyl-6-hydroxymethylperimidin-2-one (XXIIIa) in concentrated hydrochloric acid. Concentrated hydrochloric acid, 20 ml, was added in portions on cooling to 0.485 g (20 mmol) of alcohol **XXIIIa**. The resulting viscous material was ground for 10 min at room temperature and adjusted to pH ~9–10 by adding a 20% solution of potassium hydroxide. The precipitate was filtered off, washed with water, and dried in air. The products were separated by preparative thin-layer chromatography using chloroform as eluent. We isolated 0.06 g (12.5%) of 1,3-dimethyl-2-oxoperimidine-6-carbaldehyde (**XXIIa**, R_f 0.60), 0.07 g (7.5%) of compound **XXVI** (R_f 0.35), and 0.17 g of a mixture of oligomeric products (according to the ^{1}H NMR data, R_f 0.17). Analogous results were obtained when trifluoroacetic acid was used instead of hydrochloric acid.

1,3,1',3'-Tetramethyl-2,2'-dioxo-6,6'-methylene-diperimidine-7-carbaldehyde (XXVI). mp 245–246°C. IR spectrum (mineral oil), ν, cm^{-1}: 1671 (C=O), 1580 (C=C). ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 3.42 s, 3.45 s, 3.48 s, and 3.56 s (3H each, Me); 4.50 s (2H, CH$_2$); 6.48 d, 6.63 d, 6.70 d, 6.76 d, 7.05 d, 7.11 d, 7.31 d, and 8.08 d (1H each, 4'-H, 9'-H, 4-H, 9-H, 5'-H, 7'-H, 5-H, 8-H, $J_{4',5'}$ = 7.92, $J_{9',8'}$ = 7.62, $J_{4,5}$ = 8.21, $J_{9,8}$ = 8.34, $J_{7,8'}$ = 8.49 Hz); 7.34 d.d (1H, 8'-H, $J_{8',7'}$ = 8.49, $J_{8',9'}$ = 7.62 Hz); 10.21 s (1H, CHO). Found, %: C 72.60; H 5.19; N 11.92. C$_{28}$H$_{24}$N$_4$O$_3$. Calculated, %: C 72.40; H 5.21; N 12.06.

Transformations of 1,3-dimethyl-2,3-dihydro-1H-perimidin-6-ylmethanol (XXVa) over Al$_2$O$_3$. A mixture of 0.2 g (0.88 mmol) of alcohol **XXVa** and 20 g of freshly calcined Al$_2$O$_3$ in 25 ml of benzene was shaken for 3 h at 80°C. The mixture was cooled, the precipitate of Al$_2$O$_3$ was filtered off and washed on a filter with warm benzene (~20 ml), and the filtrate was evaporated to obtain 15 mg (8%) of bis(1,3-dimethyl-2,3-dihydro-1H-perimidin-6-yl)methane (**XXVII**), R_f 0.9 (Al$_2$O$_3$, CHCl$_3$), mp 220–222°C (decomp., from hexane) [18]. The sorbent was additionally extracted with boiling ethanol, the solvent was distilled off from the extract, and the residue was subjected to column chromatography on Al$_2$O$_3$ (1.5× 22 cm) using chloroform–hexane (1:1) as eluent, fractions with R_f 0.9, 0.7, and 0.4 being collected. The first fraction contained compound **XXVII**, 25 mg (14%); overall yield 0.04 g (22%). Products in the second fraction (~3 mg) were not identified. From the third fraction we isolated 16 mg (6.6%) of compound **XXIIIa**. The upper part of the sorbent in the column was extracted with hot ethanol, and evaporation of the extract gave 20 mg of dark yellow crystals which were insoluble in nonpolar solvents. The ^{1}H NMR spectrum of that product in DMSO-d_6 contained a set of singlets in the δ region 9–9.5 ppm (2-H) which are typical of perimidinium salts **XXVIII**.

Transformations of 1,2,2,3-tetramethyl-2,3-dihydro-1H-perimidin-6-ylmethanol (XXVb) over Al$_2$O$_3$. A mixture of 0.256 g (1 mmol) of alcohol **XXVb**, 30 ml of benzene, and 20 g of freshly calcined Al$_2$O$_3$ (Brockmann activity grade I) was shaken for 2 h at 70–80°C. The mixture was cooled, 10 ml of a 10% solution of potassium hydroxide was added, and the mixture was filtered. The precipitate of aluminum oxide was extracted with 20 ml of chloroform, 50 ml of acetone, and 100 ml of ethanol, and the extracts were combined and evaporated to obtain 0.48 g of a yellow material. This material was treated with 80 ml of chloroform, and the chloroform extract was evaporated to a small volume and was subjected to preparative TLC on Al$_2$O$_3$ using chloroform as eluent. Two fractions with R_f 0.85 and 0.35 were collected. The second fraction contained aldehyde **XXIVb**, yield 0.04 g (15%), mp 127–128°C (from octane). From the first fraction we isolated 0.065 g (16%) of spiro compound **XXIX** as yellow crystals. The crude product softened at 90–95°C and melted completely at 113–115°C. Compound **XXIX** was thermally unstable, and we failed to recrystallize it. It also gradually decomposed on storage in a refrigerator at –20°C. Analytical data were not obtained. IR spectrum (CHCl$_3$): ν(C=O) 1720 cm^{-1}. ^{1}H NMR spectrum (CDCl$_3$), δ, ppm: 1.36 s (6H, 2-CMe$_2$), 2.90 d (2H, H$_b$, ^2J = 15.5 Hz), 2.95 s (6H, NCH$_3$), 2.96 d (3H, NHCH$_3$, J = 5 Hz), 3.67 d (2H, H$_a$, ^2J = 15.5 Hz), 6.12 d and 6.60 d (1H each, 3'-H, 2'-H, $J_{3',2'}$ = 10.3 Hz), 6.57 d (2H, 4-H, 10-H, J_o = 7.5 Hz), 6.64 d (1H, 6'-H, $J_{6',7'}$ = 8.6 Hz), 6.72 d (1H, 8'-H, $J_{8',7'}$ = 7.2 Hz), 7.10 d (2H, 5-H, 9-H, J_o = 7.5 Hz), 7.44 d.d (1H, 7'-H, $J_{7',6'}$ = 8.6, $J_{7,8'}$ = 7.2 Hz), 9.48 br.s (1H, N**H**CH$_3$).

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