

Synthesis of Pyrimidinocyclophanes Having a Bridging Nitrogen Atom

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Received July 17, 2007

Abstract—Reactions of 1,3-bis(ω -bromoalkyl)-substituted uracils, quinazoline-2,4-dione, and 5-methyl-1,3,5-triazine-2,4,6-trione and 1,3-bis(*m*-bromomethylbenzyl)-5-bromouracil with amines (aliphatic amines, benzylamines, naphthylmethanamine, and anisidine) gave a series of macrocyclic compounds having one pyrimidine or triazine fragment and an azapolymethylene bridge connecting the N¹ and N³ atoms of the heteroring. The bridging nitrogen atom in some macrocyclic compounds was subjected to quaternization with methyl *p*-toluenesulfonate.

DOI: 10.1134/S1070428008060183

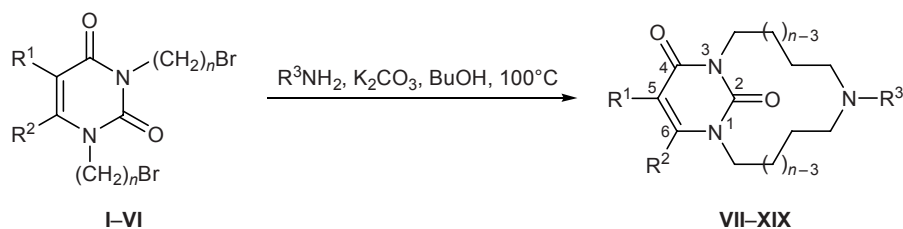
Macrocyclic structures in which the bridging fragments contain heteroatoms, e.g., crown ethers, are effective complexing agents capable of binding both neutral and charged species [1]. Introduction of heteroaromatic fragments into such structures opens new prospects in the application of macrocyclic compounds, in particular in the design of new materials. These prospects originate from the well known ability of heteroaromatic compounds to form associates through various interactions (π – π interactions, hydrogen bonding, interactions with charged or neutral substrates). Pyrimidine derivatives can be used as heteroaromatic fragments.

We previously synthesized macrocyclic compounds incorporating two 6-methyl-2,4-dioxo-1,2,3,4-tetrahydropyrimidine (6-methyluracil) fragments and a nitrogen atom in the bridge. Their structural specificity and properties, in particular complex formation with electron-deficient substrates, were studied [2–4]. We also reported on the synthesis, structure in crystal and in solution, conformational behavior, and acid-promoted aggregation of first representatives of macrocyclic compounds consisting of a 5-methyluracil (thymine) or 6-methyluracil fragment in which the N¹ and N³ atoms were linked through a (CH₂)₅N(R)(CH₂)₅ bridge, where R is a butyl or benzyl group [5, 6]. These macrocycles may be regarded as pyrimidine analogs of cyclophanes.

In the present article we report on the synthesis of a wide series of pyrimidinocyclophanes having hydrocarbon bridges with different lengths and rigidity and various substituents on the C⁵ and C⁶ atoms of the pyrimidine ring and on the bridging nitrogen atom. Ring closure of 1,3-bis(5-bromopentyl)- and 1,3-bis(6-bromohexyl)uracils was accomplished via reactions with both aliphatic amines and those containing an aromatic fragment. Pyrimidinocyclophanes VII–XIX containing uracil, thymine, 6-methyluracil, and 5-nitro-uracil fragments were obtained at a heterocycle-to-amine ratio of 1:1 to 1:3 in butan-1-ol in the presence of K₂CO₃ and a catalytic amount of tetrabutylammonium hydrogen sulfate (Scheme 1). As amines we used benzylamine, substituted benzylamines having electron-donating and electron-withdrawing groups in the *ortho*, *meta*, or *para* position of the benzene ring, optically active (*R*)-(+)-1-phenylethanamine, anisidine, 1-naphthylmethanamine, 3-phenylpropan-1-amine, and butan-1-amine. The polymethylene bridges (CH₂)_{*n*}N(R)(CH₂)_{*n*} in the products included 10 or 12 CH₂ groups (*n* = 5 or 6).

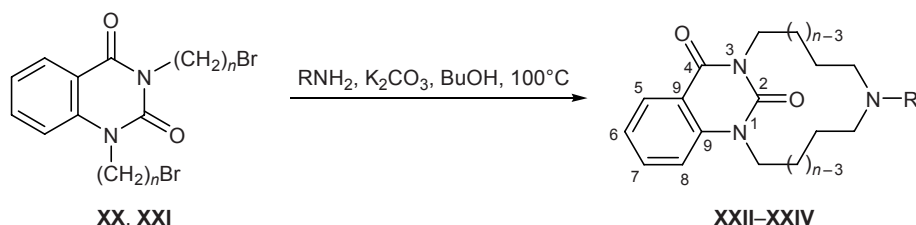
Our attempts to synthesize pyrimidinocyclophanes with a shorter bridge, (CH₂)₄N(R)(CH₂)₄ (*n* = 4) by reaction of 1,3-bis(4-bromobutyl)thymine (I) with amines were unsuccessful. According to the mass spectral data (electron impact), the reaction mixtures contained only traces of the target products.

Scheme 1.



I, $n = 4$, $R^1 = Me$, $R^2 = H$; **II-V**, **VII-XVIII**, $n = 5$; **VI**, **XIX**, $n = 6$; **II**, **VI-X**, **XIX**, $R^1 = Me$, $R^2 = H$; **III**, **XI**, **XII**, $R^1 = H$, $R^2 = Me$; **IV**, **XIII-XVII**, $R^1 = R^2 = H$; **V**, **XVIII**, $R^1 = NO_2$, $R^2 = H$; **VII**, **XI**, **XVIII**, **XIX**, $R^3 = PhCH_2$; **VIII**, $R^3 = 4-MeOC_6H_4CH_2$; **IX**, $R^3 = Bu$; **X**, $R^3 = Ph(CH_2)_3$; **XII**, $R^3 = PhCH(Me)$; **XIII**, $R^3 = 4-MeOC_6H_4$; **XIV**, $R^3 = 3-MeOC_6H_4CH_2$; **XV**, $R^3 = 1-naphthylmethyl$; **XVI**, $R^3 = 4-ClC_6H_4CH_2$; **XVII**, $R^3 = 2-ClC_6H_4CH_2$.

Scheme 2.



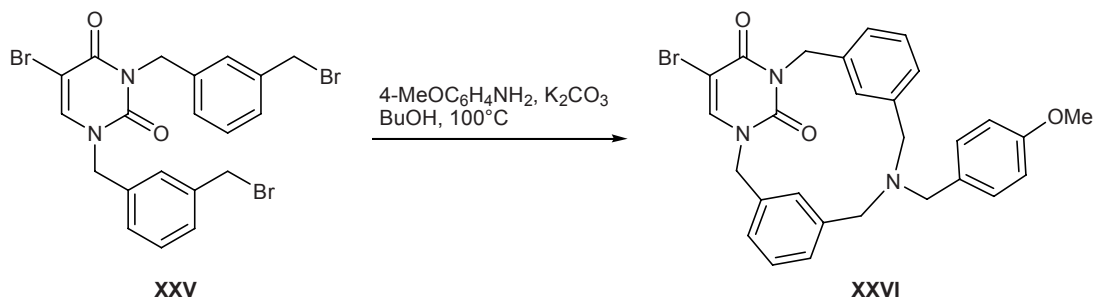
XX, XXII, XXIII, $n = 5$; **XXIV**, $n = 6$; **XXII, XXIV**, $R = PhCH_2$; **XXIII**, $R = naphthalen-1-ylmethyl$.

We also tried to use in the above condensations uracil derivatives with a fused aromatic fragment, e.g., quinazoline-2,4-diones. By reaction of 1,3-bis(5-bromopentyl)- and 1,3-bis(6-bromohexyl)quinazoline-2,4-diones **XX** and **XXI** with benzylamine and 1-naphthylmethylamine we obtained the corresponding pyrimidinocyclophanes **XXII-XXIV** (Scheme 2).

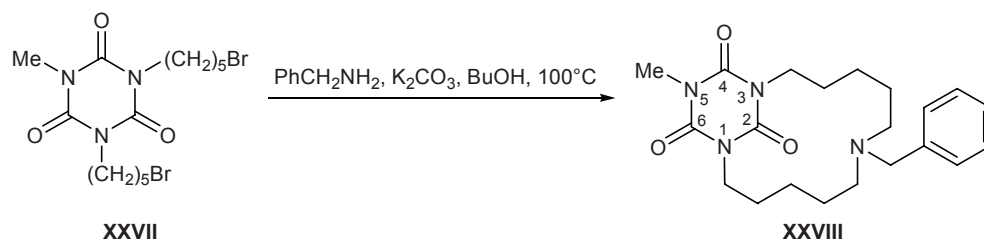
Flexible polymethylene bridge can be replaced by more rigid *m*-phenylenedimethylene linker. Scheme 3 illustrates the synthesis of macrocycle **XXVI** consisting of *m*- $CH_2C_6H_4CH_2N(CH_2C_6H_4OMe-p)-CH_2C_6H_4CH_2-m$ and 5-bromouracil fragments. Other 1,3-diazaheterocycles may also be used instead of pyrimidine fragment. For example, the reaction of 1,3-bis(5-bromopentyl)-5-methylhexahydro-1,3,5-triazine-2,4,6-trione (**XXVII**) with benzylamine gave macrocyclic compound **XXVIII** (Scheme 4).

The progress of the above reactions was monitored by TLC until complete disappearance of initial compounds **I-VI**, **XX, XXI, XXV**, and **XXVII** (5–8 h). The yields of macrocycles were fairly poor (5–20%). The residue was a mixture of a large number of products (presumably oligomeric) which were difficult to separate. With a view to improve the yield, we performed the reactions under various conditions. However, variation of the solvent, the use of dilute solutions, and addition of tetraalkylammonium or transition metal salts did not result in appreciable increase of the yield of the target macrocyclic products. Nevertheless, the following conclusions can be drawn. The reaction in acetonitrile gives a smaller amount of by-products than in butan-1-ol, though the reaction time considerably increases (15–20 h). Replacement of K_2CO_3 by Cs_2CO_3 also favors more selective process.

Scheme 3.

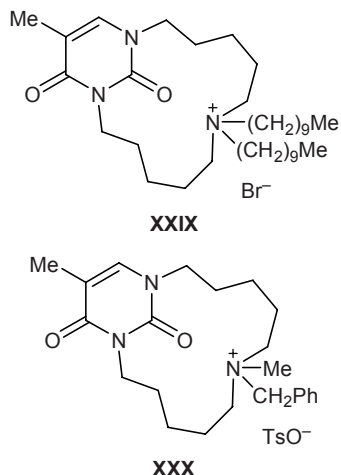


Scheme 4.



The structure of the obtained macrocyclic compounds was proved by high-resolution mass spectrometry and NMR spectroscopy. The ^1H NMR spectra of pyrimidinocyclophanes having $m\text{-CH}_2\text{C}_6\text{H}_4\text{CH}_2$ and $(\text{CH}_2)_5\text{N(R)}(\text{CH}_2)_5$ bridging moieties, as well as of the first representatives of this class of macroheterocycles (compounds **VII**, **IX**, and **XI**) [5, 6], revealed magnetic nonequivalence of all protons in the methylene groups at the N^1 and N^3 atoms of the pyrimidine ring. These protons resonated in the ^1H NMR spectra of **VIII**, **X**, **XII–XVIII**, **XXII**, **XXIII**, and **XXVI** in the region δ 5.7–3.5 ppm, the distance between their signals being $\delta\Delta = 0.2\text{--}0.4$ ppm. Protons in the N^1CH_2 and N^3CH_2 groups of pyrimidinocyclophanes **XIX** and **XXIV** having 12 methylene units in the bridge were equivalent in pairs, and they resonated as two multiplets. These findings are clearly related to the length of the $(\text{CH}_2)_n\text{N(R)}(\text{CH}_2)_n$ bridge which determines specificity of the steric structure of the above compounds [5, 6].

We also tried to synthesize pyrimidinocyclophanes with quaternized nitrogen atom in the bridge. However, our attempts to accomplish N-alkylation of compounds having a benzyl group on the nitrogen were unsuccessful, presumably because of steric hindrances. By treatment of compound **IX** having a butyl substituent on the bridging nitrogen atom with 1-bromodecane in acetonitrile we obtained quaternary salt



XXIX which showed a moderate antimicrobial activity [7]. We succeeded in effecting quaternization of the bridging nitrogen atom in *N*-benzyl derivative **XXII** only under severe conditions using methyl *p*-toluenesulfonate as both alkylating agent and solvent: as a result, amphiphilic pyrimidinocyclophane **XXX** was isolated. Macrocyclic compounds **XXIX** and **XXX** are soluble in chloroform and water.

Thus we synthesized a series of macrocyclic compounds whose molecules consist of a pyrimidine fragment and an aza polymethylene bridge connecting the N^1 and N^3 atoms of the pyrimidine ring; substituents in the pyrimidine ring, the length and rigidity of the bridging fragment, and substituents on the bridging nitrogen atom were varied. Quaternization of the bridging nitrogen atom with decyl bromide and methyl *p*-toluenesulfonate gave water-soluble amphiphilic pyrimidinocyclophanes.

EXPERIMENTAL

The NMR spectra were recorded at 30°C on a Bruker Avance-600 spectrometer (600 MHz for ^1H and 150.926 MHz for ^{13}C) from solutions in CDCl_3 using the solvent signals as reference (CHCl_3 , δ 7.26 ppm; CDCl_3 , δ_{C} 77.0 ppm). The structure of the products was determined on the basis of a series one- and two-dimensional NMR correlation experiments (DEPT, $^1\text{H}\text{--}^1\text{H}$ COSY, $^1\text{H}\text{--}^{13}\text{C}$ HSQC, and $^1\text{H}\text{--}^{13}\text{C}$ HMBC [8–10]). The electron-impact mass spectra (70 eV) were obtained on a Finnigan MAT-212 mass spectrometer at a resolution of 1000 using MSS MASPEC II³² data processing system (direct sample admission into the ion source, vaporizer temperature 20 to 300°C , electron emission current 1.0 mA).

The syntheses of compounds **II** [5], **III** [11], **IV** [11], **VII** [5, 6], **IX** [6], **XI** [5], **XXVII** [12], and **XXXI** [7] were reported previously.

1,3-Bis(ω -bromoalkyl)uracils (general procedure). The corresponding uracil or quinazoline-2,4-dione, 1 equiv, was added to a solution of sodium but-

oxide prepared by dissolution of 2 equiv of metallic sodium in butan-2-ol, and the mixture was stirred for 20 h on heating under reflux. The solvent was distilled off, the residual alcohol was removed as azeotrope with toluene, and the residue (disodium salt) was thoroughly dried under reduced pressure. The disodium salt, 1 equiv, was dispersed in dimethylformamide, a solution of 8 equiv of the corresponding α,ω -dibromoalkane in DMF was added under stirring, and the mixture was stirred for 50–60°C until neutral reaction of an aqueous solution of a sample withdrawn from the mixture (5–10 h). The solvent and excess α,ω -dibromoalkane were distilled under reduced pressure, the residue was treated with chloroform, the mixture was filtered, and the filtrate was concentrated to a volume of 10–15 ml and subjected to column chromatography on aluminum oxide.

1,3-Bis(4-bromobutyl)-5-methyl-1,2,3,4-tetrahydropyrimidine-2,4-dione (I) was synthesized from 10.85 g (63.82 mmol) of thymine disodium salt and 110.3 g (510.65 mmol) of 1,4-dibromobutane in 180 ml of DMF. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (3:1). Elution with the solvent mixture gave 8.4 g (33%) of compound **I** as an oily substance. ^1H NMR spectrum, δ , ppm: 6.98 s (1H, 6-H), 4.00 m (2H, 3-CH₂), 3.75 m (2H, 1-CH₂), 3.43 m (4H, CH₂Br), 1.92 s (3H, 5-CH₃), 1.87–1.78 m (8H, CH₂CH₂). Found, %: C 39.64; H 4.99; Br 40.02; N 7.08. C₁₃H₂₀Br₂N₂O₂. Calculated, %: C 39.42; H 5.09; Br 40.34; N 7.07.

1,3-Bis(5-bromopentyl)-5-nitro-1,2,3,4-tetrahydropyrimidine-2,4-dione (V) was synthesized from 12.8 g (63.69 mmol) of 5-nitrouracil disodium salt and 117.2 g (509.52 mmol) of 1,5-dibromopentane in 250 ml DMF. The column was eluted in succession with petroleum ether and diethyl ether. Elution with diethyl ether gave 12.4 g (43%) of compound **V** as an oily substance. ^1H NMR spectrum, δ , ppm: 8.70 s (1H, 6-H), 4.00 m (2H, 3-CH₂), 3.95 m (2H, 1-CH₂), 3.43 m (4H, CH₂Br), 1.97–1.47 m (12H, CH₂CH₂-CH₂). Found, %: C 36.84; H 4.69; Br 35.26; N 9.11. C₁₄H₂₁Br₂N₃O₄. Calculated, %: C 36.94; H 4.65; Br 35.11; N 9.23.

1,3-Bis(6-bromohexyl)-5-methyl-1,2,3,4-tetrahydropyrimidine-2,4-dione (VI) was synthesized from 10.0 g (58.82 mmol) of thymine disodium salt and 100.0 g (409.84 mmol) of 1,6-dibromohexane in 220 ml of DMF. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (2:1). Elution with the solvent mixture gave 10.9 g

(41%) of compound **VI** as an oily substance. ^1H NMR spectrum, δ , ppm: 7.02 s (1H, 6-H), 3.94 m (2H, 3-CH₂), 3.72 m (2H, 1-CH₂), 3.40 m (4H, CH₂Br), 1.92 s (3H, 5-CH₃), 1.89–1.83 m (4H, NCH₂CH₂), 1.73–1.61 m (4H, CH₂CH₂Br), 1.51–1.46 m (4H, NCH₂CH₂CH₂), 1.40–1.34 m (4H, CH₂CH₂CH₂Br). Found, %: C 45.28; H 6.21; Br 35.01; N 6.38. C₁₇H₂₈Br₂N₂O₂. Calculated, %: C 45.15; H 6.24; Br 35.34; N 6.19.

1,3-Bis(5-bromopentyl)-1,2,3,4-tetrahydroquinazoline-2,4-dione (XX) was synthesized from 19.07 g (92.59 mmol) of quinazoline-2,4-dione disodium salt and 170.37 g (740.74 mmol) of 1,5-dibromopentane in 200 ml of DMF. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (1:2). Elution with the solvent mixture gave 12.05 g (28%) of compound **XX** with mp 52–54°C. ^1H NMR spectrum, δ , ppm: 8.22 d (1H, 8-H, $J = 7.8$ Hz), 7.67 t (1H, 6-H, $J = 7.3$ Hz), 7.25 t (1H, 7-H, $J = 8.6$ Hz), 7.17 d (1H, 5-H, $J = 7.5$ Hz), 4.09 m (4H, NCH₂), 3.42 m (4H, CH₂Br), 1.93 m (4H, NCH₂CH₂), 1.73 m (4H, CH₂CH₂Br), 1.60 m (4H, NCH₂CH₂CH₂). Found, %: C 47.09; H 5.19; Br 34.95; N 6.05. C₁₈H₂₄Br₂N₂O₂. Calculated, %: C 46.98; H 5.26; Br 34.73; N 6.09.

1,3-Bis(6-bromohexyl)-1,2,3,4-tetrahydroquinazoline-2,4-dione (XXI) was synthesized from 15.00 g (72.82 mmol) of quinazoline-2,4-dione disodium salt and 142.14 g (582.54 mmol) of 1,6-dibromohexane in 200 ml of DMF. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (1:2). Elution with the solvent mixture gave 14.06 g (36%) of compound **XXI** as an oily substance. ^1H NMR spectrum, δ , ppm: 8.22 d (1H, 8-H, $J = 7.8$ Hz), 7.67 t (1H, 6-H, $J = 8.6$ Hz), 7.25 t (1H, 7-H, $J = 6.8$ Hz), 7.17 d (1H, 5-H, $J = 7.9$ Hz), 4.09 m (4H, NCH₂), 3.40 m (4H, CH₂Br), 1.86 m (4H, NCH₂CH₂), 1.72 m (4H, CH₂CH₂Br), 1.50–1.42 m (8H, CH₂CH₂). Found, %: C 49.09; H 5.79; Br 32.92; N 5.66. C₂₀H₂₈Br₂N₂O₂. Calculated, %: C 49.20; H 5.78; Br 32.73; N 5.74.

5-Bromo-1,3-bis(3-bromomethylbenzyl)-1,2,3,4-tetrahydropyrimidine-2,4-dione (XXV) was synthesized from 4.2 g (17.87 mmol) of 5-bromouracil disodium salt and 44.04 g (166.82 mmol) of 1,3-bis-(bromomethyl)benzene in 120 ml of DMF. The mixture was stirred at room temperature. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (2:1). Elution with the solvent mixture gave 8.1 g (81%) of compound **XXV** with mp 133–135°C. ^1H NMR spectrum, δ , ppm: 7.52–

7.20 m (8H, H_{arom}), 7.48 s (1H, 6-H), 5.17 s (2H, 3-CH₂), 4.92 s (2H, 1-CH₂), 4.47 s (4H, CH₂Br). Mass spectrum, m/z (I_{rel} , %): 560 (34) [$M + 6$]⁺, 559 (23) [$M + 5$]⁺, 558 (100) [$M + 4$]⁺, 557 (23) [$M + 3$]⁺, 556 (100) [$M + 2$]⁺, 554 (34) [M]⁺, 479 (52), 477 (88), 475 (52), 397 (90), 395 (83), 185 (61), 104 (100). Found, %: C 43.06; H 3.04; Br 43.15; N 5.09. C₂₀H₁₇Br₃N₂O₂. Calculated, %: C 43.12; H 3.08; Br 43.03; N 5.03.

Macrocyclic compounds VIII, X, XII–XIX, XXII–XXIV, XXVI, and XXVIII (general procedure). The corresponding amine, 1.3 equiv, was dissolved in butan-1-ol, 4 equiv of potassium carbonate and a catalytic amount of tetrabutylammonium hydrogen sulfate were added, the mixture was heated to 90°C, and a solution of 1 equiv of the corresponding 1,3-bis(ω-bromoalkyl)uracil in butan-1-ol was added. The mixture was stirred at 100–110°C, the progress of the reaction being monitored by TLC. When the reaction was complete, the solvent was distilled off, 150–250 ml of chloroform was added to the residue, the mixture was filtered, and the filtrate was concentrated to a volume of 10–20 ml and subjected to column chromatography on silica gel.

7-(4-Methoxybenzyl)-15-methyl-1,7,13-triazabicyclo[11.3.1]heptadec-14-ene-16,17-dione (VIII) was synthesized from 2.50 g (5.90 mmol) of compound II and 1.05 g (7.66 mmol) of *p*-methoxybenzylamine using 3.25 g (23.58 mmol) of K₂CO₃ in 200 ml of butan-1-ol. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (2:1). Elution with the solvent mixture gave 0.65 g (28%) of compound VIII as an oily substance. ¹H NMR spectrum, δ, ppm: 7.05 m (2H, H_{arom}), 6.95 s (1H, 6-H), 6.78 m (2H, H_{arom}), 4.48 m (1H, 1-CH), 4.27 m (1H, 3-CH), 4.03 m (1H, 3-CH), 3.79 s (3H, OCH₃), 3.34 m (2H, CH₂C₆H₄), 3.15 m (1H, 1-CH), 2.28 m (4H, NCH₂), 2.03 s (3H, 5-CH₃) 1.92 m (1H, 1-CH₂CH), 1.80 m (1H, 3-CH₂CH), 1.66 m (1H, 3-CH₂CH), 1.47 m (1H, 1-CH₂CH), 1.30 m (4H, NCH₂CH₂), 1.28 m (4H, NCH₂CH₂CH₂). Mass spectrum, m/z (I_{rel} , %): 400 (13) [$M + 1$]⁺, 399 (50) [M]⁺, 398 (7) [$M - 1$]⁺, 278 (41) [$M - 121$]⁺, 122 (19), 121 (100). Found, %: C 69.05; H 8.24; N 10.57. [M]⁺ 399.251. C₂₃H₃₃N₃O₃. Calculated, %: C 69.14; H 8.33; N 10.52. *M* 399.252.

15-Methyl-7-(3-phenylpropyl)-1,7,13-triazabicyclo[11.3.1]heptadec-14-ene-16,17-dione (X) was synthesized from 3.00 g (7.08 mmol) of compound II and 1.91 g (14.15 mmol) of 3-phenylpropan-1-amine using 3.91 g (28.33 mmol) of K₂CO₃ in 300 ml of butan-1-

ol. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (1:1.5 and 1.5:1). The product was eluted with the 1.5:1 solvent mixture. Yield 0.13 g (5%), oily substance. ¹H NMR spectrum, δ, ppm: 7.17–7.11 m (5H, H_{arom}), 6.87 s (1H, 6-H), 4.49 m (1H, 1-CH), 4.28 m (1H, 3-CH), 4.01 m (1H, 3-CH), 3.16 m (1H, 1-CH), 2.49 m (2H, PhCH₂), 2.29–2.26 m (6H, NCH₂), 2.04 s (3H, 5-CH₃), 1.78–1.50 m (4H, N¹⁽³⁾CH₂CH₂), 1.40–1.20 m (10H, PhCH₂CH₂, NCH₂CH₂, N¹⁽³⁾CH₂CH₂CH₂). Mass spectrum, m/z (I_{rel} , %): 398 (3) [$M + 1$]⁺, 397 (14) [M]⁺, 396 (3) [$M - 1$]⁺, 293 (19), 292 (100) [$M - 105$]⁺, 98 (10). Found, %: C 72.45; H 8.84; N 10.59. C₂₄H₃₅N₃O₂. Calculated, %: C 72.51; H 8.87; N 10.57. *M* 397.2729.

16-Methyl-7-[(1*R*)-1-phenylethyl]-1,7,13-triazabicyclo[11.3.1]heptadec-14-ene-16,17-dione (XII) was synthesized from 2.00 g (4.72 mmol) of compound III and 0.74 g (6.12 mmol) of (*R*)-α-phenylethylamine using 2.61 g (18.88 mmol) of K₂CO₃ in 150 ml of acetonitrile at 70–75°C. The column was eluted in succession with petroleum ether and ethyl acetate–petroleum ether (1.2:1). Elution with the solvent mixture gave 0.16 g (9%) of compound XII with mp 90–91.5°C. ¹H NMR spectrum, δ, ppm: 7.22 m (5H, H_{arom}), 5.67 s (1H, 5-H), 4.50 m (1H, 1-CH), 4.25 m (1H, 3-CH), 4.03 m (1H, 3-CH), 3.83 m (1H, CHPh), 3.54 m (1H, 1-CH), 2.25–2.19 m (4H, NCH₂CH₂), 2.16 s (3H, 6-CH₃), 1.80–1.26 m (12H, CH₂CH₂CH₂), 1.25 s (3H, CHCH₃). Mass spectrum, m/z (I_{rel} , %): 384 (12) [$M + 1$]⁺, 383 (47) [M]⁺, 369 (253) [$M - 14$]⁺, 368 (100) [$M - 15$]⁺, 278 (24) [$M - 105$]⁺, 264 (36) [$M - 119$]⁺, 105 (82). Found, %: C 72.12; H 8.58; N 10.87. C₂₃H₃₃N₃O₂. Calculated, %: C 72.03; H 8.67; N 10.96. *M* 383.2573.

7-(4-Methoxyphenyl)-1,7,13-triazabicyclo[11.3.1]heptadec-14-ene-16,17-dione (XIII). Sodium hydride, 0.13 g (5.42 mmol), was added to a solution of 0.31 g (2.52 mmol) of *p*-anisidine in 120 ml of DMF, the mixture was stirred for 2 h at room temperature, a catalytic amount of tetrabutylammonium hydrogen sulfate and a solution of 1.00 g (2.44 mmol) of compound IV in 70 ml of DMF were added, and the mixture was stirred for 20 h at 70–75°C. The mixture was cooled, the solvent was distilled off, 150 ml of chloroform was added to the residue, the mixture was filtered, and the filtrate was concentrated to a volume of 10–20 ml and subjected to column chromatography on silica gel. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether

(1:1). Elution with the solvent mixture gave 0.01 g (1%) of compound **XIII** as an oily substance. ^1H NMR spectrum, δ , ppm: 7.04 d (1H, 6-H), 6.78 d (2H, H_{arom}), 6.63 m (2H, H_{arom}), 5.70 d (1H, 5-H), 4.55 m (1H, 1-CH), 4.21 m (1H, 3-CH), 4.07 m (1H, 3-CH), 3.74 s (3H, OCH_3), 2.88 m (1H, 1-CH), 2.04 m (4H, NCH_2), 1.88–1.46 m (4H, $\text{N}^{1(3)}\text{CH}_2\text{CH}_2$), 1.41 m (4H, NCH_2CH_2), 1.25 m (4H, $\text{NCH}_2\text{CH}_2\text{CH}_2$). Mass spectrum, m/z (I_{rel} , %): 371 (60) $[M]^+$, 342 (10), 178 (26), 142 (100). Found, %: C 68.01; H 7.84; N 11.27. $\text{C}_{21}\text{H}_{29}\text{N}_3\text{O}_3$. Calculated, %: C 67.90; H 7.87; N 11.31. M 371.221.

7-(3-Methoxybenzyl)-1,7,13-triazabicyclo[11.3.1]-heptadec-14-ene-16,17-dione (XIV) was synthesized from 2.50 g (6.10 mmol) of compound **IV** and 1.09 g (7.96 mmol) of *m*-methoxybenzylamine using 3.37 g (24.42 mmol) of K_2CO_3 in 150 ml of butan-1-ol. The column was eluted in succession with petroleum ether and ethyl acetate–petroleum ether (1.2:1). Elution with the solvent mixture gave 0.26 g (11%) of compound **XIV** as an oily substance. ^1H NMR spectrum, δ , ppm: 7.17 m (1H, H_{arom}), 7.08 d (1H, 6-H), 6.78 m (2H, H_{arom}), 6.73 m (1H, H_{arom}), 5.78 d (1H, 5-H), 4.50 m (1H, 1-CH), 4.26 m (1H, 3-CH), 4.00 m (1H, 3-CH), 3.79 s (3H, OCH_3), 3.39 m (2H, $\text{CH}_2\text{C}_6\text{H}_4$), 3.20 m (1H, 1-CH), 2.31 m (4H, NCH_2), 1.95 m (1H, 1- CH_2CH), 1.83 m (1H, 3- CH_2CH), 1.63 m (1H, 3- CH_2CH), 1.49 m (1H, 1- CH_2CH), 1.33 m (4H, NCH_2CH_2), 1.32 m (4H, $\text{NCH}_2\text{CH}_2\text{CH}_2$). Mass spectrum, m/z (I_{rel} , %): 386 (12) $[M + 1]^+$, 385 (52) $[M]^+$, 384 (34) $[M - 1]^+$, 356 (23), 265 (17), 264 (100) $[M - 121]^+$, 236 (18), 164 (12), 148 (10), 136 (13), 122 (26), 121 (95). Found, %: C 68.59; H 8.04; N 10.87. $\text{C}_{22}\text{H}_{31}\text{N}_3\text{O}_3$. Calculated, %: C 68.54; H 8.11; N 10.90. M 385.2365.

7-(1-Naphthylmethyl)-1,7,13-triazabicyclo[11.3.1]heptadec-14-ene-16,17-dione (XV) was synthesized from 4.20 g (10.24 mmol) of compound **IV** and 1.83 g (11.66 mmol) of 1-naphthylmethanamine using 5.66 g (41.01 mmol) of K_2CO_3 in 250 ml of butan-1-ol. The column was eluted in succession with petroleum ether and ethyl acetate–petroleum ether (1.2:1). Elution with the solvent mixture gave 0.21 g (5%) of compound **XV**, mp 120–122°C. ^1H NMR spectrum, δ , ppm: 8.13 m (1H, H_{arom}), 7.81 m (1H, H_{arom}), 7.73 m (1H, H_{arom}), 7.45 m (2H, H_{arom}), 7.39 m (1H, H_{arom}), 7.37 m (1H, H_{arom}), 6.95 d (1H, 6-H), 5.75 d (1H, 5-H), 4.46 m (1H, 1-CH), 4.06 m (1H, $\text{C}_{10}\text{H}_7\text{CH}$), 4.00 m (1H, 3-CH), 3.72 m (1H, $\text{C}_{10}\text{H}_7\text{CH}$), 3.70 m (1H, 3-CH), 3.12 m (1H, 1-CH),

2.41 m (4H, NCH_2), 1.84 m (1H, 3- CH_2CH), 1.83 m (1H, 1- CH_2CH), 1.66 m (1H, 3- CH_2CH), 1.38 m (1H, 1- CH_2CH), 1.41 m (4H, NCH_2CH_2), 1.40 m (4H, $\text{NCH}_2\text{CH}_2\text{CH}_2$). Mass spectrum, m/z (I_{rel} , %): 406 (7) $[M + 1]^+$, 405 (25) $[M]^+$, 404 (11) $[M - 1]^+$, 264 (35) $[M - 141]^+$, 141 (100). Found, %: C 74.09; H 7.74; N 10.27. $\text{C}_{25}\text{H}_{31}\text{N}_3\text{O}_2$. Calculated, %: C 74.04; H 7.70; N 10.36. M 405.2416.

7-(4-Chlorobenzyl)-1,7,13-triazabicyclo[11.3.1]-heptadec-14-ene-16,17-dione (XVI) was synthesized from 3.00 g (7.32 mmol) of compound **IV** and 1.35 g (9.54 mmol) of *p*-chlorobenzylamine using 4.04 g (29.28 mmol) of K_2CO_3 in 150 ml of butan-1-ol. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (3:1). Elution with the solvent mixture gave 0.27 g (10%) of compound **XVI** as an oily substance. ^1H NMR spectrum, δ , ppm: 7.24 m (2H, H_{arom}), 7.11 m (2H, H_{arom}), 7.08 d (1H, 6-H), 5.81 d (1H, 5-H), 4.50 m (1H, 1-CH), 4.26 m (1H, 3-CH), 4.00 m (1H, 3-CH), 3.70 m (2H, $\text{CH}_2\text{C}_6\text{H}_4$), 3.20 m (1H, 1-CH), 2.30 m (4H, NCH_2), 1.93 m (1H, 1- CH_2CH), 1.82 m (1H, 3- CH_2CH), 1.65 m (1H, 3- CH_2CH), 1.48 m (1H, 1- CH_2CH), 1.33 m (4H, $\text{NCH}_2\text{CH}_2\text{CH}_2$), 1.30 m (4H, NCH_2CH_2). Mass spectrum, m/z (I_{rel} , %): 392 (3) $[M + 3]^+$, 391 (18) $[M + 2]^+$, 390 (19) $[M + 1]^+$, 389 (47) $[M]^+$, 388 (32) $[M - 1]^+$, 360 (22), 265 (21), 264 (99) $[M - 125]^+$, 236 (27), 168 (14), 140 (13), 127 (36), 125 (100). Found, %: C 64.66; H 7.21; Cl 9.15; N 10.77. $\text{C}_{21}\text{H}_{28}\text{ClN}_3\text{O}_2$. Calculated, %: C 64.69; H 7.24; Cl 9.09; N 10.78. M 389.1869.

7-(2-Chlorobenzyl)-1,7,13-triazabicyclo[11.3.1]-heptadec-14-ene-16,17-dione (XVII) was synthesized from 2.05 g (5.00 mmol) of compound **IV** and 0.92 g (6.50 mmol) of *o*-chlorobenzylamine using 5.52 g (40.00 mmol) of K_2CO_3 in 150 ml of butan-1-ol. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (1:1 and 2:1). Elution with the 2:1 solvent mixture gave 0.10 g (5%) of compound **XVII**, mp 118°C. ^1H NMR spectrum, δ , ppm: 7.32 m (1H, H_{arom}), 7.28 m (1H, H_{arom}), 7.19 m (1H, H_{arom}), 7.13 m (1H, H_{arom}), 7.06 d (1H, 6-H), 5.79 d (1H, 5-H), 4.50 m (1H, 1-CH), 4.27 m (1H, 3-CH), 3.99 m (1H, 3-CH), 3.54 m (2H, $\text{CH}_2\text{C}_6\text{H}_4$), 3.19 m (1H, 1-CH), 2.34 m (4H, NCH_2), 1.94 m (1H, 1- CH_2CH), 1.80 m (1H, 3- CH_2CH), 1.64 m (1H, 3- CH_2CH), 1.47 m (1H, 1- CH_2CH), 1.31 m (4H, NCH_2CH_2), 1.30 m (4H, $\text{NCH}_2\text{CH}_2\text{CH}_2$). Mass spectrum, m/z (I_{rel} , %): 392 (3) $[M + 3]^+$, 391 (12) $[M + 2]^+$, 390 (18) $[M + 1]^+$, 389 (36) $[M]^+$, 388 (34) $[M - 1]^+$, 360 (23), 265 (17), 264 (100) $[M - 125]^+$, 236 (28),

168 (23), 154 (12), 127 (23), 125 (68). Found, %: C 64.85; H 7.21; Cl 9.11; N 10.72. $C_{21}H_{28}ClN_3O_2$. Calculated, %: C 64.69; H 7.24; Cl 9.09; N 10.78. M 389.1869.

7-Benzyl-15-nitro-1,7,13-triazabicyclo[11.3.1]-heptadec-14-ene-16,17-dione (XVIII) was synthesized from 3.00 g (6.59 mmol) of compound **V** and 1.40 g (13.08 mmol) of benzylamine using 3.64 g (26.38 mmol) of K_2CO_3 in 200 ml of acetonitrile. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (5:1). Elution with the solvent mixture gave 0.10 g (4%) of compound **XVIII**, mp 167–169°C. 1H NMR spectrum, δ , ppm: 8.64 s (1H, 6-H), 7.22 m (2H, H_{arom}), 7.21 m (1H, H_{arom}), 7.09 m (2H, H_{arom}), 4.65 m (1H, 1-CH), 4.39 m (1H, 3-CH), 4.03 m (1H, 3-CH), 3.40 m (1H, 1-CH), 3.39 m (2H, CH_2Ph), 2.31 m (4H, NCH_2), 2.02 m (1H, 1- CH_2CH), 1.79 m (1H, 3- CH_2CH), 1.68 m (1H, 3- CH_2CH), 1.55 m (1H, 1- CH_2CH), 1.34 m (4H, NCH_2CH_2), 1.32 m (4H, $NCH_2CH_2CH_2$). Mass spectrum, m/z (I_{rel} , %): 401 (9) [$M + 1$] $^+$, 400 (33) [M] $^+$, 384 (21) [$M - 16$] $^+$, 383 (84) [$M - 17$] $^+$, 371 (11), 344 (9), 91 (100). Found, %: C 63.05; H 7.01; N 14.02. $C_{21}H_{28}N_4O_4$. Calculated, %: C 62.98; H 7.05; N 13.99. M 400.2110.

8-Benzyl-17-methyl-1,8,15-triazabicyclo[13.3.1]-nonadec-16-ene-18,19-dione (XIX) was synthesized from 2.50 g (5.53 mmol) of compound **VI** and 1.20 g (11.21 mmol) of benzylamine using 3.10 g (22.46 mmol) of K_2CO_3 in 350 ml of butan-1-ol. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (1:5 and 2:1). Elution with the 2:1 solvent mixture gave 0.48 g (22%) of compound **XIX**, mp 82–83°C. 1H NMR spectrum, δ , ppm: 7.26 m (5H, H_{arom}), 6.94 s (1H, 6-H), 4.08 m (2H, 1- CH_2), 3.79 m (2H, 3- CH_2), 3.48 m (2H, CH_2Ph), 2.32 m (4H, NCH_2), 1.95 s (3H, 5- CH_3), 1.72 m (4H, $N^{(3)}CH_2CH_2$), 1.57 m (4H, NCH_2CH_2), 1.40 m (4H, $N^{(3)}CH_2CH_2CH_2$), 1.28 m (4H, $NCH_2CH_2CH_2$). Mass spectrum, m/z (I_{rel} , %): 398 (8) [$M + 1$] $^+$, 397 (26) [M] $^+$, 368 (13), 354 (35), 307 (12), 306 (58) [$M - 91$] $^+$, 134 (14), 106 (15), 96 (16), 91 (100). Found, %: C 72.38; H 8.81; N 10.68. $C_{24}H_{35}N_3O_2$. Calculated, %: C 72.51; H 8.87; N 10.57. M 397.273.

7-Benzyl-14,15-benzo-1,7,13-triazabicyclo[11.3.1]heptadec-14-ene-16,17-dione (XXII) was synthesized from 2.50 g (5.43 mmol) of compound **XX** and 1.16 g (10.84 mmol) of benzylamine using 3.00 g (21.74 mmol) of K_2CO_3 in 380 ml of butan-1-ol. The column was eluted in succession with petroleum ether

and diethyl ether–petroleum ether (4:1). Elution with the solvent mixture gave 0.37 g (17%) of compound **XXII**, mp 123–124°C. 1H NMR spectrum, δ , ppm: 8.36 d (1H, 8-H, $J = 7.5$ Hz), 7.67 t (1H, 6-H, $J = 7.6$ Hz), 7.29 t (1H, 7-H, $J = 7.5$ Hz), 7.24 d (1H, 5-H, $J = 8.3$ Hz), 7.07 m (1H, H_{arom}), 6.97 m (2H, H_{arom}), 6.92 m (2H, H_{arom}), 4.87 m (1H, 1-CH), 4.46 m (1H, 3-CH), 4.09 m (1H, 3-CH), 3.85 m (1H, 1-CH), 3.35 m (1H, $CHPh$), 3.28 m (1H, $CHPh$), 2.31 m [2H, 1-(CH_2) $_4CH_2N$], 2.29 m [2H, 3-(CH_2) $_4CH_2N$], 1.93 m (1H, 1- CH_2CH), 1.72 m (2H, 3- CH_2CH_2), 1.69 m (1H, 1- CH_2CH), 1.47 m (4H, NCH_2CH_2), 1.37 m [2H, 1-(CH_2) $_3CH_2$], 1.33 m (2H, 3-(CH_2) $_3CH_2$). Mass spectrum, m/z (I_{rel} , %): 406 (31) [$M + 1$] $^+$, 405 (100) [M] $^+$, 389 (12), 376 (18), 328 (32), 314 (58) [$M - 91$] $^+$, 142 (23), 91 (68). Found, %: C 74.19; H 7.64; N 10.59. $C_{25}H_{31}N_3O_2$. Calculated, %: C 74.04; H 7.70; N 10.36. M 405.242.

7-(1-Naphthylmethyl)-14,15-benzo-1,7,13-triazabicyclo[11.3.1]heptadec-14-ene-16,17-dione (XXIII) was synthesized from 2.00 g (4.35 mmol) of compound **XX** and 0.82 g (5.22 mmol) of 1-naphthylmethanamine using 2.40 g (17.39 mmol) of K_2CO_3 in 150 ml of butan-1-ol. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (1:3). Elution with the solvent mixture gave 0.19 g (10%) of compound **XXIII** as an oily substance. 1H NMR spectrum, δ , ppm: 8.37–7.15 m (9H, H_{arom}), 4.83 m (1H, 1-CH), 4.45 m (1H, 3-CH), 4.10 m (1H, $C_{10}H_7CH$), 3.92 m (1H, 3-CH), 3.69 m (1H, 1-CH), 3.40 m (1H, $C_{10}H_7CH$), 2.43 m (4H, NCH_2), 1.95–1.20 m (12H, $CH_2CH_2CH_2$). Mass spectrum, m/z (I_{rel} , %): 456 (9) [$M + 1$] $^+$, 455 (25) [M] $^+$, 315 (10), 314 (51) [$M - 141$] $^+$, 142 (14), 141 (100). Found, %: C 76.29; H 7.36; N 9.36. $C_{29}H_{33}N_3O_2$. Calculated, %: C 76.45; H 7.30; N 9.22. M 455.2573.

8-Benzyl-16,17-benzo-1,8,15-triazabicyclo[13.3.1]nonadec-16-ene-18,19-dione (XXIV) was synthesized from 2.50 g (5.12 mmol) of compound **XXI** and 0.66 g (6.17 mmol) of benzylamine using 3.00 g (21.74 mmol) of K_2CO_3 in 500 ml of butan-1-ol. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (1:2). Elution with the solvent mixture gave 0.24 g (11%) of compound **XXIV**, mp 158–160°C. 1H NMR spectrum, δ , ppm: 8.25 d (1H, 8-H, $J = 6.5$ Hz), 7.66 t (1H, 6-H, $J = 7.2$ Hz), 7.24–7.19 m (7H, H_{arom}), 4.29 m (2H, 1- CH_2), 4.20 m (2H, 3- CH_2), 3.49 m (2H, CH_2Ph), 2.35 m (4H, NCH_2), 1.78 m (4H, $N^{(3)}CH_2CH_2$), 1.43–1.32 m (12H, $CH_2CH_2CH_2$). Mass spectrum, m/z (I_{rel} , %): 434 (10) [$M + 1$] $^+$, 433 (41) [M] $^+$, 390 (25),

356 (30), 342 (100) $[M - 91]^+$, 314 (10), 202 (12), 188 (13), 174 (10), 160 (14), 132 (33), 106 (31), 91 (88). Found, %: C 74.66; H 8.18; N 9.59. $C_{27}H_{35}N_3O_2$. Calculated, %: C 74.79; H 8.14; N 9.69. M 433.2729.

19-Bromo-9-(4-methoxybenzyl)-1,9,17-triazatetracyclo[15.3.1.1^{3,7}.1^{11,15}]tricos-3(4),5,7(22),-11(12),13,15(23),19-heptaene-18,21-dione (XXVI) was synthesized from 2.50 g (4.49 mmol) of compound XXV and 0.80 g (5.84 mmol) of *p*-methoxybenzylamine using 2.48 g (17.97 mmol) of K_2CO_3 in 150 ml of butan-1-ol. The column was eluted in succession with petroleum ether and diethyl ether–petroleum ether (2:1). Elution with the solvent mixture gave 0.09 g (4%) of compound XXVI, mp 74–76°C. 1H NMR spectrum, δ , ppm: 7.60 s (1H, 6-H), 7.38–6.88 m (12H, H_{arom}), 5.68 d (1H, 1-CH, $J = 15.4$ Hz), 5.48 d (1H, 3-CH, $J = 14.9$ Hz), 5.19 d (1H, 3-CH, $J = 14.6$ Hz), 4.35 d (1H, 1-CH, $J = 15.1$ Hz), 3.78 s (3H, OCH_3), 3.54 s (2H, $CH_2C_6H_4OMe$), 3.41 m (4H, NCH_2). Mass spectrum, m/z (I_{rel} , %): 533 (29) $[M + 2]^+$, 531 (29) $[M]^+$, 426 (11), 412 (12) $[M + 2 - 121]^+$, 410 (9) $[M - 121]^+$, 237 (19), 122 (35), 121 (100), 105 (53), 104 (16). Found, %: C 63.06; H 4.99; Br 15.19; N 7.82. $C_{28}H_{26}BrN_3O_3$. Calculated, %: C 63.16; H 4.92; Br 15.01; N 7.89. M 531.1158.

7-Benzyl-15-methyl-1,7,13,15-tetraazabicyclo[11.3.1]heptadecane-14,16,17-trione (XXVIII) was synthesized from 3.30 g (7.48 mmol) of compound XXVII and 1.04 g (9.72 mmol) of benzylamine using 4.13 g (29.93 mmol) of K_2CO_3 in 180 ml of butan-1-ol. The column was eluted in succession with petroleum ether and ethyl acetate–petroleum ether (1.2:1). Elution with the solvent mixture gave 0.37 g (13%) of compound XXVIII, mp 103.5–105°C. 1H NMR spectrum, δ , ppm: 7.28–7.15 m (5H, H_{arom}), 4.07 t (4H, 1-CH₂, 3-CH₂), 3.44 m (5H, 5-CH₃, CH₂Ph), 2.32 m (4H, NCH_2), 1.75–1.63 m (4H, $N^{(3)}CH_2CH_2$), 1.54–1.34 m (8H, CH_2CH_2). Mass spectrum, m/z (I_{rel} , %): 387 (12) $[M + 1]^+$, 386 (52) $[M]^+$, 385 (10) $[M - 1]^+$, 357 (17), 329 (10), 295 (62) $[M - 91]^+$, 267 (16), 174 (13), 134 (14), 132 (18), 120 (32), 118 (20), 106 (35), 91 (100). Found, %: C 65.33; H 7.79; N 14.38. $C_{21}H_{30}N_4O_3$. Calculated, %: C 65.26; H 7.82; N 14.50. M 386.2318.

7-Benzyl-7,15-dimethyl-16,17-dioxo-1,13-diaz-7-azoniabicyclo[11.3.1]heptadec-14-ene 4-methylbenzenesulfonate (XXX). A mixture of 0.10 g (0.27 mmol) of compound VII and 4 g of methyl *p*-toluenesulfonate was stirred for 6 h at 80°C. The mixture was cooled and diluted with diethyl ether, and

the precipitate was separated by decanting, washed with two additional portions of diethyl ether (the product was separated each time by decanting), and dried under reduced pressure. Yield 0.15 g (100%). 1H NMR spectrum, δ , ppm: 7.80–7.35 m (9H, H_{arom}), 7.02 s (1H, 6-H), 4.10 m (4H, 1-CH₂, 3-CH₂), 3.74 m (2H, CH₂Ph), 3.03 m (4H, NCH_2), 2.64 s (3H, NCH_3), 2.35 s (3H, $C_6H_4CH_3$), 1.95 s (3H, 5-CH₃), 1.94–1.30 m (12H, $CH_2CH_2CH_2$). Found, %: C 64.98; H 7.49; N 7.44; S 5.63. $C_{30}H_{41}N_3O_5S$. Calculated, %: C 64.84; H 7.44; N 7.56; S 5.77.

This study was performed under financial support by the Russian Foundation for Basic Research (project nos. 05-03-32497a, 05-03-32558a, 07-03-00392a), by the Presidium of the Russian Academy of Sciences (program no. 8), and by the Joint Russian–American Program “Basic Research and Higher Education (REC-007). A.E. Nikolaev and Sh.K. Latypov thank the Foundation for Support of the Russian Science for financial support. The compounds were analyzed at the NMR Department, Federal Spectral and Analytical Center and Federal Center for Physicochemical Studies of Substances and Materials (state contract nos. 02.451.11.7036 and 02.451.11.7019 of the Ministry of Education and Science of the Russian Federation).

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