diffractometer, $Mo_{K\alpha}$ radiation ($\lambda = 0.71073$ Å), and a graphite monochromator. A total of 14022 reflections were measured using a $\omega/2\Theta$ scan mode ($3.3° \leq 2\Theta \leq 54.0°$). Crystal system: tetragonal, space group $P4_2/n$, $Z = 8$, $a = 24.846(3)$, $b = 24.846(3)$, $c = 7.8219(13)$ Å; $V = 4828.6(12)$ Å^3; $\rho_{calcd} = 1.262$ g cm^{-3}; $\mu = 0.084$ mm^{-1}. The structure was solved directly with SHELXS97 with 7011 independent reflections, the structure was refined according to full-matrix least-squares procedures (SHELXL97); $wR^2 = 0.1919$ for 7011 reflections, $R^1 = 0.0986$ for 2706 observed reflections [$I_0 > 2.0\sigma(I_0)$] and 311 refined parameters. The positions of the hydrogen atoms were calculated according to geometric considerations and refined isotropically.

X-ray crystal structure analysis of **4a**: A yellow crystal $0.57 \times 0.48 \times 0.34$ mm^3 in size was measured at room temperature with a STADI4 diffractometer, $Mo_{K\alpha}$ radiation ($\lambda = 0.71073$ Å), and a graphite monochromator. A total of 13516 reflections were measured using a $\omega/2\Theta$ scan mode ($3.3° \leq 2\Theta \leq 54.9°$). Crystal system: triclinic, space group $P\bar{1}$, $Z = 2$, $a = 9.6489(3)$, $b = 9.8280(5)$, $c = 13.2209(6)$ Å, $\alpha = 82.092(5)$, $\beta = 69.261(5)$, $\gamma = 84.237(4)°$; $V = 1159.53(10)$ Å^3; $\rho_{calcd} = 1.313$ g cm^{-3}; $\mu = 0.088$ mm^{-1}. The structure was solved directly with SHELXS97 using 6758 independent reflections, the structure was refined according to full-matrix least-squares procedures (SHELXL97); $wR^2 = 0.1340$ for 6758 reflections, $R^1 = 0.0477$ for 5035 observed reflections [$I_0 > 2.0\sigma(I_0)$] and 405 refined parameters. The positions of the hydrogen atoms were calculated geometrically and refined isotropically.

Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. CCDC-133187. Copies of the data can be obtained free of charge on application to CCDC, 12 Union Road, Cambridge CB21EZ, UK (fax: (+44)1223-336-033; e-mail: deposit@ccdc.cam.ac.uk).

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[9] All new compounds were characterized by IR, ^{1}H NMR, and ^{13}C NMR spectroscopy and mass spectrometry, solubility permitting, and by elemental analysis. Selected physical data for **2a**: yellow crystals, m.p. 115–120 °C; IR (KBr): $\tilde{\nu} = 1682$ cm^{-1} (C=O); ^{1}H NMR (500 MHz, CD_3OD, 25 °C, TMS): $\delta = 3.10$ (s, 3H; NCH_3), 3.53 (s, 3H; $COOCH_3$, 4.41 (d, ^3J(H,H) = 4.7 Hz, 1H; 4-H), 4.89 (dd, ^3J(H,H) = 7.8, 4.7 Hz, 1H; 5-H), 5.94 (d, ^3J(H,H) = 7.8 Hz, 1H; 6-H), 7.09–7.87 (m, 6H; aromat. H, 2-H); UV/Vis (methanol): $\lambda_{max}(\varepsilon) = 348$ (4324), 232 (6229); MS (70 eV): m/z (%): 229 (2) [M^+], 212 (9) [M^+], 169 (100) [$M^+ - 60$].

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[11] Selected physical data for **3a**: white needles, yield: 39%, m.p. 212–218 °C, IR (KBr): $\tilde{\nu} = 1723$ cm^{-1} (C=O); ^{1}H NMR (500 MHz, $CDCl_3$, 25 °C, TMS): $\delta = 2.04$ (s, 6H; NCH_3), 3.18 (AA′HH′RR′XX′, ^3J(H,H) = 9.4, 1.9 Hz, 2 × ^4J(H,H) = 1.9 Hz, 2H; 4-, 11-H), 3.37 (d, ^3J(H,H) = 1.9 Hz, 2H; 3-, 10-H), 3.69 (s, 6H; $COOCH_3$), 3.84 (dt, ^3J(H,H) = 9.4 Hz, 2 × ^4J(H,H) = 1.9 Hz, 2H; 1-, 5-H), 4.59 (t, ^4J(H,H) = 1.9 Hz, 2H; 7-, 8-H), 7.05–7.18 (m, 10H; aromat. H); UV/Vis (chloroform): $\lambda_{max}(\varepsilon) = 250$ (973); MS (70 eV): m/z (%): 458 (3) [M^+], 229 (34) [$M^+/2$], 214 (6) [$M^+/2 - CH_3$], 152 (100) [$M^+/2 - Ph$].

[12] Selected physical data for **4a**: small yellow spheres, yield: 42%, m.p. 188–191 °C, IR (KBr): $\tilde{\nu} = 1725$, 1713 cm^{-1} (C=O); ^{1}H NMR (500 MHz, $CDCl_3$, 25 °C, TMS): $\delta = 2.00$, 2.04 (2 × s, 6H; NCH_3), 3.73, 3.75 (2 × s, 6H; $COOCH_3$), 3.25 (dd, ^3J(H,H) = 8.9, 3.1 Hz, 1H, 4-H), 3.29 (ddd, ^3J(H,H) = 8.9, 4.9, 3.3 Hz, 1H; 9-H), 3.36 (d, ^3J(H,H) = 3.1 Hz, 1H; 3-H), 3.38 (d, ^3J(H,H) = 3.3 Hz, 1H; 10-H), 3.51 (ddd, ^3J(H,H) = 8.9, 4.5 Hz, ^4J(H,H) = 2.9 Hz, 1H; 8-H), 3.76 (ddd, ^3J(H,H) = 8.9, 4.9 Hz, ^4J(H,H) = 2.7 Hz, 1H; 5-H), 4.43 (dd, ^3J(H,H) = 4.5 Hz, ^4J(H,H) = 2.7 Hz, 1H, 7-H), 4.58 (d, ^4J(H,H) = 2.9 Hz, 1H; 1-H), 7.03–7.19 (m, 10H; aromat. H); UV/Vis (chloroform): $\lambda_{max}(\varepsilon) = 250$ (2863); MS (70 eV): m/z (%): 458 (15) [M^+], 427 (4) [$M^+ - OCH_3$], 228 (35) [$M^+/2 - 1$], 214 (6) [$M^+/2 - CH_3$], 152 (100) [$M^+/2 - Ph$].

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Formation of a Novel Cage Compound with a Pentacyclo[6.3.0.$1^{4,11}$.$0^{2,6}$.$0^{5,10}$]dodecane Skeleton by Photolysis of [3_4](1,2,4,5)Cyclophane[**]

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Prismanes constitute a fascinating family of $(CH)_n$ polyhedra,[1] several members of which, namely, prismane,[2] cubane,[3] and pentaprismane,[4, 5] have been successfully synthesized. Recently, attention has focused on the challenging objective of synthesizing the higher prismanes, in particular, hexaprismanes **1**. Despite many efforts, they have so far eluded synthesis, mainly because of the lack of proper synthetic routes and the expected higher strain energies than the lower prismanes. In our approach to construct the hexaprismane skeleton by photolysis of multibridged [3_n]cyclophanes ($n = 3-6$) we first optimized the reaction conditions using the lowest homologue [3_3](1,3,5)cyclophane (**2**),

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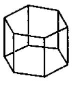
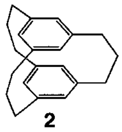
[**] Multibridged [3_n]Cyclophanes. Part 8. For previous papers, see refs. [6 (Part 6) and 8 (Part 7)]. We gratefully acknowledge the financial support by the Grant-in-Aid for the Priority Area (A) of Creation of Delocalized Electronic Systems (no. 11133244) from the Ministry of Education, Science, Sports, and Culture, Japan.



1



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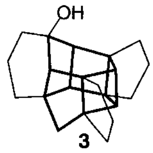
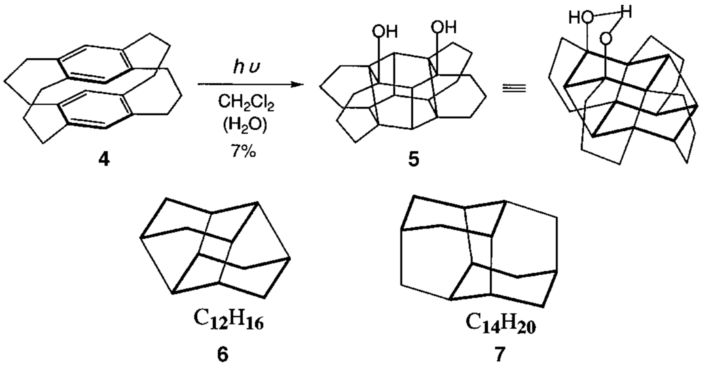


3

and found that **2** was converted into the bishomopentaprismane skeleton **3** upon irradiation in H_2O-saturated CH_2Cl_2 with a low-pressure Hg lamp.[6] The photolysis of the next higher homologue of **2**, $[3_4](1,2,4,5)$cyclophane (**4**),[7] was studied to obtain the hexaprismane derivatives and further information on the reaction mechanism. Recently, an improved and practical synthetic method has made **4** readily available.[8]

A solution of **4** in H_2O-saturated CH_2Cl_2 (6.9×10^{-4} mol L^{-1}) was irradiated with a low-pressure Hg lamp for 1.5 h at room temperature under Ar. The reaction was monitored by HPLC and TLC. Separation of the crude product by column chromatography on silica gel with AcOEt/hexane (1/2) afforded the starting material **4** (45%) and a new cage compound **5** as colorless crystals (7%; Scheme 1).[9] The mass spectrum indicates a molecular formula of $C_{24}H_{32}O_2$ (m/z 352.2) and the base peak (m/z 335.2) arises from the dehydration of M^+. The hydroxy proton signals appear as a singlet in the ^{1}H NMR spectrum at $\delta = 4.15$. The proton-decoupled ^{13}C NMR spectrum of **5** shows six signals ($\delta = 17.7$, 19.2, 23.0, 25.6, 26.1, 35.8) for the secondary carbon atoms, five ($\delta = 44.9$, 48.1, 52.5, 57.4, 58.3) for the tertiary carbon atoms, and three ($\delta = 50.6$, 54.6, 85.1) for the quaternary carbon atoms, which suggests a structure with some symmetry. The highly deshielded quaternary carbon signal at $\delta = 85.1$ is assigned to those bearing a hydroxy group. The structure was finally identified as **5** on the basis of an X-ray structural analysis.[10]



Scheme 1. Photochemical reaction of **4** in H_2O-saturated CH_2Cl_2 by irradiation with a low pressure Hg lamp provided the novel cage compound **5**. The skeleton of **5** (**6**) and diamantane **7** are also shown.

The novel polycyclic compound is composed of six cyclohexane rings in the chair conformation, four of which are transformed from the original trimethylene bridges and the remaining two originate from the benzene rings (Figure 1). To the best of our knowledge the pentacyclo$[6.3.0.1^{4,11}.0^{2,6}.0^{5,10}]$dodecane skeleton **6** was designed by Evtushenko et al. as a member of the $C_{12}H_{16}$ family,[11] but its synthesis has not yet been described in the literature. The skeleton **6** resembles diamantane **7** (pentacyclo$[7.3.1.1^{4,12}.0^{2,7}.0^{6,11}]$tetradecane), which was first synthesized by Schleyer et al. in 1965 and named "congressane".[12] However, the upper and lower cyclohexane rings in the chair conformation are directly connected at four positions (C2-C8, C4-C12, C3-C10, C6-C7) in **5**, whereas the latter two positions are connected with C_1 units in **7**. Structure **5** has C_s symmetry with only one symmetry plane (C3-C6-C7-C10) located between the two hydroxy groups. Although the bond lengths and bond angles of diamantane **7** are normal,[13] the C3-C10 and C6-C7 bonds of **5**, which are connected to the upper and lower cyclohexane rings in the chair conformation, are abnormally long (1.603(4), 1.610(4) Å) as compared with the RHF/6-31G* optimized C–C bond length (1.552 Å) of a cyclopentane ring. These elongated bond lengths are regenerated within the range of ±0.003 Å by the RHF/6-31G* level MO calculations. The upper and lower six-membered rings of the skeleton **6** are cyclohexanes with a distorted chair conformation; the dihedral angles of the C1-C2-C4-C5 and C5-C6-C1 planes (104.2°) as well as the C8-C9-C11-C12 and C9-C10-C11 planes (101.9°) are much smaller than those of the C1-C2-C4-C5 and C2-C3-C4 planes (152.8°) as well as the C8-C9-C11-C12 and C8-C7-C12 planes (155.6°; Figure 2). The corresponding value of a cyclohexane ring in the chair conformation as estimated by the RHF/6-31G* level MO calculations is 130.5°. Thus shortening of the C_1 bridges in diamantane **7** to no bridges causes a significant deformation of the chair-cyclohexane rings in **6**, by elongation of the C3-C10 and C6-C7 bond lengths, and a much higher strain energy for **6** than for **7**. According to the MM3 calculations,[14] the steric energy of **6** is 54.8 kcal mol^{-1} higher than that of **7**.

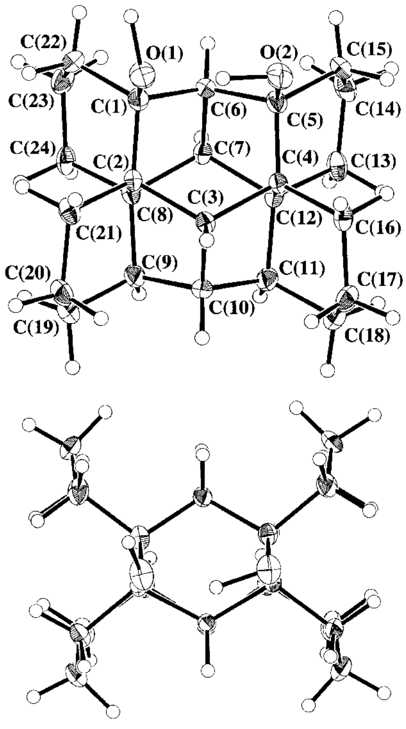


Figure 1. ORTEP drawing of **5** (30% probability ellipsoids).

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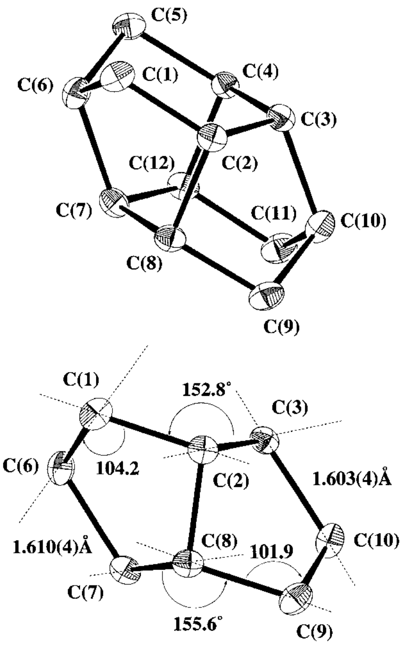

Figure 2. Crystal structure of the skeleton of **5** (**6**, hydrogen atoms are omitted for clarity). Selected bond lengths [Å] and angles [°]: C(1)-C(2) 1.521(4), C(1)-C(6) 1.546(4), C(2)-C(3) 1.587(4), C(2)-C(8) 1.537(4), C(3)-C(4) 1.588(4), C(3)-C(10) 1.603(4), C(4)-C(5) 1.526(4), C(4)-C(12) 1.536(4), C(5)-C(6) 1.548(4), C(6)-C(7) 1.610(4), C(7)-C(8) 1.578(4), C(7)-C(12) 1.578(4), C(8)-C(9) 1.528(4), C(9)-C(10) 1.531(4), C(10)-C(11) 1.532(4), C(11)-C(12) 1.534(4); C(1)-C(2)-C(3) 115.6(2), C(1)-C(6)-C(7) 103.8(2), C(1)-C(2)-C(8) 100.6(2), C(2)-C(3)-C(4) 116.0(2), C(2)-C(3)-C(10) 102.3(2), C(3)-C(4)-C(5) 115.9(2), C(3)-C(4)-C(12) 97.9(2), C(4)-C(5)-C(6) 99.8(2), C(5)-C(6)-C(1) 111.9(3), C(5)-C(6)-C(7) 103.8(2), C(6)-C(7)-C(8) 103.6(2), C(6)-C(1)-C(2) 100.0(2), C(6)-C(7)-C(12) 103.8(2), C(7)-C(8)-C(9) 115.7(2), C(7)-C(8)-C(2) 97.9(2), C(8)-C(9)-C(10) 100.3(2), C(8)-C(2)-C(3) 98.3(2), C(9)-C(10)-C(11) 106.9(3), C(10)-C(11)-C(12) 99.9(2), C(10)-C(3)-C(4) 102.7(2), C(11)-C(12)-C(7) 115.9(2) C(11)-C(12)-C(4) 100.8(3), C(12)-C(7)-C(8) 114.7(2), C(12)-C(4)-C(5) 100.7(2), C(6)-C(1)-C(5)-C(4) 104.2(2), C(10)-C(11)-C(9)-C(8) 101.9(2), C(3)-C(2)-C(4)-C(5) 152.8(2), C(7)-C(12)-C(8)-C(9) 155.6(2).

Thus a new polycyclic compound **5** having a pentacyclo[6.3.0.1^{4,11}.0^{2,6}.0^{5,10}]dodecane skeleton was formed by the photolysis of **4** in a H_2O-saturated CH_2Cl_2 solution. The series of reactions involving first the formation of the hexaprismane derivative followed by protonation and equilibration of the resultant carbocations, and interception of the most stable cation with a hydroxide ion are proposed, similar to the photochemical reaction of **2**.[6]

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[9] Selected spectroscopic data and elemental analysis for **5**: m.p. 268–270 °C (decomp); ^{1}H NMR (600 MHz, $CDCl_3$, TMS): $\delta = 1.11$ (d, $J = 3.1$ Hz, 1 H), 1.13 (d, $J = 3.1$ Hz, 1 H), 1.23–1.78 (m, 24 H), 1.81 (d, $J = 5.2$ Hz, 1 H), 1.89 (d, $J = 2.7$ Hz, 2 H), 2.23 (d, $J = 5.2$ Hz, 1 H), 4.15 (S, 2 H, OH); ^{13}C NMR (150 MHz, $CDCl_3$, TMS, DEPT): $\delta = 17.7$ (2 × sec.), 19.2 (2 × sec.), 23.0 (2 × sec.), 25.6 (2 × sec.), 26.1 (2 × sec.), 35.7 (2 × sec.), 44.9 (tert.), 48.0 (tert.), 50.6 (2 × quat.), 52.5 (2 × tert.), 54.6 (2 × quat.), 57.4 (tert.), 58.2 (tert.), 85.2 (2 × quat.); FABMS: m/z: 352.2 [M^+]. Elemental analysis calcd for $C_{24}H_{32}O_2 \cdot 0.25\,H_2O$: C 80.74, H 9.17; found: C 80.43, H 9.07.

[10] The X-ray crystal data for **5**: $C_{24}H_{32}O_2$, $T = 23$ °C, $Mo_{K\alpha}$ (Rigaku AFC7R diffractometer, $\lambda = 0.71070$ Å), crystal dimensions $0.30 \times 0.05 \times 0.40$ mm^3 (colorless prism), $a = 9.219(3)$, $b = 14.821(3)$, $c = 6.995(2)$ Å, $\alpha = 94.09(2)$, $\beta = 110.93(2)$, $\gamma = 82.08(2)$°, triclinic, space group $P\bar{1}$ (no. 2), $Z = 2$, $\mu(\text{Mo}) = 0.82$ cm^{-1}, $M_r = 352.52$, $V = 884.0(4)$ Å^3, anode power 45 kV × 250 mA, $\rho_{calcd} = 1.324$ g cm^{-3}, $2\theta_{max} = 55.0$°, $F(000) = 384$. 4381 reflections were measured, 1975 observed ($I < 3\sigma(I)$), with 363 parameters. The structure was solved by the direct method and was refined on Sir88.[11] Data were corrected for Lorentz polarizations. The data/parameter ratio was 5.44. $R = 0.044$, $R_w = 0.043$, GOF = 1.01, max./min. residual density $+0.22/-0.22$ e Å^{-3}. All calculations were performed using the teXsan crystallographic software package of Molecular Structure Corporation. Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. CCDC-114176. Copies of the data can be obtained free of charge on application to CCDC, 12 Union Road, Cambridge CB21EZ, UK (fax: (+44)1223-336-033; e-mail: deposit@ccdc.cam.ac.uk).

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