Tandem Ring Enlargement by Two Carbon Units from the Bicyclo[3.3.0]octane to the Bicyclo[5.5.0]dodecane System

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Dedicated to Prof. Dr. Armin de Meijere on the occasion of his 60th birthday

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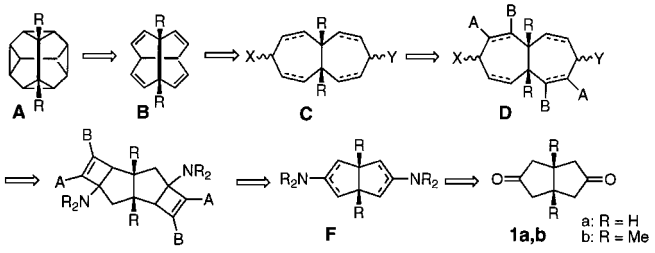
Reaction of the bis(enamines) **2b** and **3b** with acetylenedicarboxylic ester gave stable tetracyclic compounds **4b** and **5b**. The related compound **3a**, however, led to bicyclo[5.5.0]dodecatetraenes **6a**, **6b**, and **7a/7b** directly. **7a/7b** are interrelated by a rapid [1,5]-H shift, whereas **6a** and **6b** do not rearrange. Hydrolysis of **6b** yielded the novel cage compound **10** by dual intramolecular Michael addition.

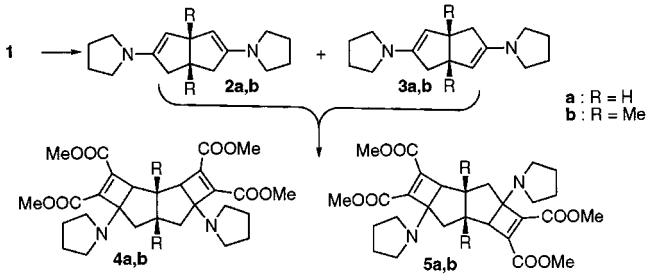
The title system is of interest as a possible precursor to certain $(CH)_{12}$ hydrocarbons. According to Balaban,[1] 357 isomers of this empirical formula are possible. Two challenging members of the series are the truncated tetrahedron-shaped compound **A** (R = H,"truncahedrane")[2] and tetraene **B** (R = H)[3] which are interrelated by a rare Woodward−Hoffmann allowed $[2_\pi a + 2_\pi a + 2_\pi a + 2_\pi a]$ process.[4] Several calculations,[5][6] synthetic speculations and futile attempts towards **A** and **B** can be found in the literature.[7−11] A possible retrosynthetic scheme (Scheme 1) starts with compounds **1**[12] and involves the twofold double ring enlargement of the title compound. We report the ring enlargement, a rapid [1,5]-H shift, and a new cage compound.



Scheme 1. Possible retrosynthesis of "truncahedrane" (**A**)

Enamines of cyclopentanone are known to react with esters of acetylenic acids to give bicyclo[3.2.0]heptene derivatives which may ring-expand to cycloheptadienes on heating.[13] Pyrrolidino enamines seem to give higher yields and have a higher chance of ring opening than morpholino enamines,[14] because a bipolar intermediate can be stabilized better.[15]

In the beginning, we concentrated on compounds bearing methyl groups at the bridgeheads to avoid a subsequent conjugation beyond the bridgeheads. It was later found that any apprehension in this respect was unwarranted. Conversion of **1b** with pyrrolidine gave a 94% yield of a 4.5:1 mixture of bis(enamines) **2b** and **3b**. Addition of dimethyl acetylenedicarboxylate led to an immediate mildly exothermic reaction. A mixture of bis(adducts) **4b** and **5b** (47%) was obtained on workup, along with tarred material. Interestingly, the ratio **4b/5b** was roughly 1:1, so that a partial isomerization of the enamines must have occurred in the course of the reaction. A similar observation was made earlier in a related 1,3-dipolar addition by Askani et al.[16] All attempts to ring-expand **4b/5b** by heating in various solvents at temperatures up to 140°C were in vain: Either no reaction or extensive tarring was found.



Scheme 2. Synthesis of tetracyclic compounds **4a**,**b** and **5a**,**b**

We therefore concentrated on the chemistry of **1a**. This compound's conversions both with methyl propiolate and dimethyl acetylenedicarboxylate have been mentioned by Hafner et al.[17] in an article on novel pericyclic reactions. However, no experimental details or spectroscopic data were given. An interesting reaction branching is recorded here for the first time (vide infra).

Only the bis(enamine) **3a** was formed from **1a** according to the ^{1}H-NMR spectrum. This confirms earlier statements from the literature.[17][18] **3a** is extremely sensitive towards air, moisture and heat and should be treated further immediately. When it was treated with methyl propynoate in dioxane at 90°C, the primary bis(adduct) **5a** was opened and the twofold ring-enlarged **6a** was obtained directly in

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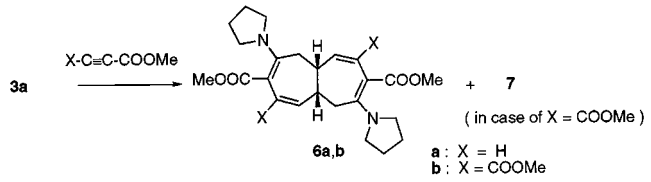
crystalline form (63% yield). It is important to note that only one isomer of **6a** was formed and that this exhibits no sigmatropic rearrangements or other isomerizations. The relevant ^{1}H- and ^{13}C-NMR data of the compound are analogous to those of 2-alkoxycarbonyl-1-pyrrolidinocyclohepta-1,3-dienes.[19][20]



Scheme 3. Formation of bicyclo[5.5.0]dodecatetraene derivatives from **3a**

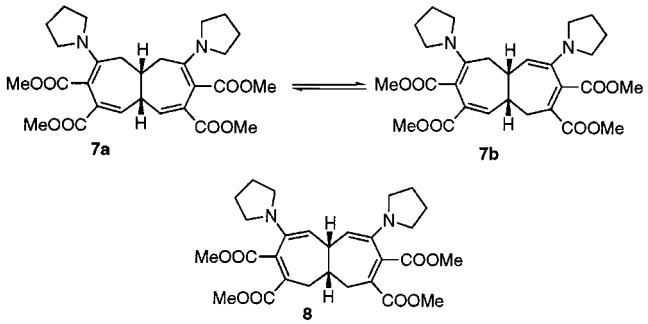
When **3a** was treated with dimethyl acetylenedicarboxylate, **6b** was similarly obtained in up to 73% yield. In addition, an isomer **7** was formed (m.p. 221°C, 24% yield), which sometimes crystallized directly from the reaction mixture prior to the isolation of **6b**. It was impossible to isomerize either compound into the other, by acid or base treatment.

On the other hand, it was apparent that both compounds were closely related. The UV maxima and IR absorptions of **6b** and **7** were similar. However, 7 exhibited fluxional behavior both in the ^{1}H- and ^{13}C-NMR spectra, which was not found in **6b**. At −40°C two vinylic doublets and ester signals were present in the ^{1}H-NMR spectrum, which coalesced to one and two signals, respectively, at room temperature. A close comparison of the ^{13}C peaks of **6b** and **7** at room temperature is given in Table 1.

Table 1. ^{13}C signals of **6b** and **7** (δ) in $CDCl_3$ at room temperature (* in **7** signals indicates broad, averaged peaks)

6b	**7**	Assignment
25.4	25.4	pyrrole β-CH_2
36.3	36.0*	ring CH_2
−	40.7	bridge CH
51.0	51.0	OCH_3
52.0	51.8	OCH_3
52.1	52.0	pyrrole α-CH_2
53.7	−	bridge CH
−	71.1	bridge CH
95.4	96.1*	=*C*−COOMe
136.58	136.5*	=CH
136.63	136.3*	=*C*−COOMe
160.4	159.9*	=C−N
166.1	166.4	−*C*OOMe
168.6	168.6	−*C*OOMe

It can be seen that **6b** has 12 signals, whereas compound **7** has 13 signals, five of which are broadened, obviously averaged peaks at room temperature. (These are marked with an asterisk in Table 1.) Although these five signals are coalesced, "averaged" in the case of compound **7**, the chemical shifts of 11 of the signals of **6b** and **7** are similar. Only the bridgehead CH of **6b** has no direct match. Instead, **7** exhibits two $C_{tert}H$ signals at substantially different chemical shifts. These observations can be explained unequivocally by assigning structures **7a** and **7b**, which are interconverted rapidly by a [1,5]-H shift. In the ^{13}C-NMR spectrum, the five nonbridge ring atoms of the seven-membered rings show the mentioned somewhat broadened coalescence peaks at room temperature, while the other signals are relatively sharp. The presence of appreciable concentrations of positional isomer **8**, the product of double H shift, is not likely because of the similar UV maxima of **6b** and **7**, and because there is no indication for it in the NMR spectra.



Scheme 4. Isomers related by [1,5]-H shifts

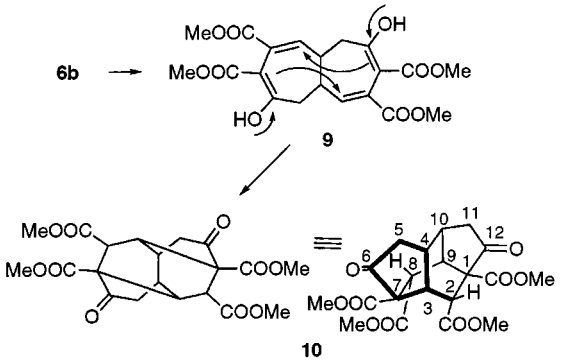
The formation of **7** obviously occurs via **2a** and **4a**. This is a little surprising as we and others have not observed the formation of **2a** but only of **3a** as stated above. On the other hand, the facile partial isomerization of **2b** and **3b** during the process of consecutive (double) cycloaddition has already been mentioned above.

Another question is why **7** fluctuates, whereas **6a** and **6b** do not rearrange. This tentative explanation is offered. The conjugated entities N−C=C−C=C−COOR in **6a/b** are probably locked and sandwiched parallel to each other by amine/ester carbonyl interaction, so that a close approach of a C−H to the terminal double bond is precluded. Repulsion of the two pyrrolidine nitrogen atoms in **7**, however, will distort the conformations so as to make an intra-ring [1,5]-H shift possible.

It is quite remarkable, that the apparently small structural change from **4b/5b** to **4a/5a** (just the removal of the bridgehead methyl groups) has such a strong effect on the possibility or ease of ring enlargements **5** → **6** or **4** → **7**, respectively.

To synthesize derivatives of compound **B**, various acidic hydrolysis experiments with **6a**, **6b**, and **7** were executed. Up to now, mostly complex and untraceable mixtures were obtained, except for one case, which is mentioned below. Apparently, partial hydrolysis of ester groups and consecutive reactions of the hydrolysis products (such as decarboxylations, Michael additions, and polymerizations) could not be controlled. In the cleavage of **6b** under very mild conditions, however, a compound was obtained and characterized in form of a crystalline clathrate with chloroform, $C_{20}H_{22}O_{10}\cdot CHCl_3$. Although the new compound **10** had the expected empirical formula of the desired structure **9**, it did not contain unsaturated or enolic structural elements. Compound **10** must be C_2-symmetric. It possessed 10 ^{13}C-NMR peaks including 1 CO, 1 CH−COOMe, 1 C_{quat}−COOMe, 2 C_{tert}−H and 1 CH_2. Taking into account that **9** is the

obvious primary product, one arrives easily at the depicted general structure by twofold intramolecular Michael addition. The *endo* arrangement of the two ester groups follows from the observed C*H*−COOMe coupling to the bridgehead C*H* with J = 3.1 Hz. The isomer with the *exo*-ester group should have a dihedral angle close to 90° and therefore a coupling constant close to zero. This *endo* positioning of the two COOR residues also explains the facile formation of a complex with chloroform by a tweezers-like action.



Scheme 5. Mechanism of formation of compound **10**

In summary, the present compounds seem to be close to the desired intermediates en route towards **B**, but ways must be sought to avoid undesired reactions. Work is being continued along similar synthetic lines.

Experimental Section

General: NMR spectra were measured in $CDCl_3$ with TMS as internal standard using the Bruker instruments AM 300 or AC 250-P. − Mass spectra: Varian MAT 311A at 70 eV. − IR spectra: Mattson FT-IR. − Melting points: Apparatus by Dr. Tottoli (Büchi); uncorrected values.

***cis*-1,5-Dimethyl-3,7-dipyrrolidinobicyclo[3.3.0]octa-2,7-diene (2b) and *cis*-1,5-Dimethyl-3,7-dipyrrolidinobicyclo[3.3.0]octa-2,6-diene (3b) (Isomer Mixture):** Compound **1** (7 g, 42 mmol) was heated overnight at reflux with freshly distilled pyrrolidine (7.9 mL, 95 mmol) and *p*-toluenesulfonic acid (250 mg) in benzene (150 mL), with azeotropic distillation of the formed water. After removal of the solvent in vacuo, the remaining slightly discolored solid was crystallized from absolute ethanol, or distilled in a kugelrohr (140°C/0.4 mbar). The product (10.8 g, 94%; m.p. 132−133°C) was highly air- and moisture-sensitive. Inspection of the ^{1}H-NMR spectrum showed that **2b** and **3b** were present in the ratio of 4.5:1. − **2b** (major isomer; 300 MHz, $CDCl_3$): δ = 1.06 (s, 6 H), 1.81 (m, 8 H, pyrr-β-H), 2.39 (s, 4 H, ring CH_2), 3.00 (m, 8 H, pyrr-α-H), 3.89 (s, 2 H, =CH). − **3b** (minor isomer, only clearly separated peaks): δ = 0.99 (s, 3 H), 1.14 (s, 3 H), 4.01 (s, 2 H). − $C_{18}H_{28}N_2$ (272.4) calcd. C 79.36, H 10.36 N 10.28; found C 79.18, H 10.57, N 10.11.

***cis*-3,7-Dipyrrolidinobicyclo[3.3.0]octa-2,7-diene (2a):** prepared analogously to **2b/3b**, m.p. 92−94°C (dec.); yield 64%; single isomer (cf. ref.[17][18]). − ^{1}H NMR: δ = 1.83 (m, 8 H, pyrr-β-H), 2.24 (m, 4 H, ring CH_2), 3.06 (m, 8 H, pyrr-α-H), 3.42 (m, 2 H, C_{tert}H), 3.89 (s, 2 H, =CH). − The compound is extremely sensitive and must be treated further as soon as possible.

Tetramethyl *cis*-1,7-Dimethyl-5,9-dipyrrolidinotetracyclo[5.5.0.0^{2,5}.0^{9,12}]dodeca-3,10-diene-3,4,10,11-tetracarboxylate (4b) and Tetramethyl *cis*-1,7-Dimethyl-5,11-dipyrrolidinotetracyclo[5.5.0.0^{2,5}.0^{8,11}]-dodeca-3,9-diene-3,4,9,10-tetracarboxylate (5b): The mixture of **2b/3b** (6.2 g, 22.8 mmol) was dissolved in dry ether (50 mL), and dimethyl butynedioate (5.6 mL, 45.6 mmol) in dry ether (20 mL) was added dropwise under nitrogen. The solution turned red, and the temperature rose by ca. 10°C. The mixture was stirred for a further 2 h at room temp. The ether was removed in vacuo. The residual red oil was crystallized from acetone. Yield 5.96 g (47%); m.p. 132−133°C. − IR: $\tilde{\nu}$ = 1725 cm^{-1}, 1695, 1630,1555. − The NMR data indicated that a ca. 1:1 mixture of **4b** and **5b** was present. − ^{1}H NMR (250 MHz): δ = 1.18 (s), 1.20 (s), 1.23 (s), 1.31−2.1 (br. m), 2.39 (d, J = 12.5), 3.08−3.40 (br. m), 3.56 (s), 3.62 (s), 3.65 (s), 3.73 (s). − $C_{30}H_{40}N_2O_8$ (556.7): calcd. C 64.73, H 7.24, N 5.03; found C 64.48, H 7.50, N 4.85. − The compounds did not rearrange after prolonged heating in xylene.

Dimethyl 5,11-Dipyrrolidinobicyclo[5.5.0]dodeca-2,4,8,10-tetraene-4,10-dicarboxylate (6a): A solution of **3a** (3.3 g, 13.5 mmol) in dry and peroxide-free dioxane (25 mL) was treated with a spatula tip of hydroquinone and stirred under N_2. A solution of methyl propynoate (2.26 mL, 27 mmol) in dry and peroxide-free dioxane (10 mL) was added dropwise to the stirred solution. An orange−red color developed, and the temperature rose to 60°C. The mixture was heated at 90°C for 1 h, then cooled to room temp. and kept overnight. The orange-colored crystals were separated off, washed with heptane (20 mL), dried in vacuo, and recrystallized from ethanol, giving colorless cotton-like crystals (yield 3.48 g, 63%). − M.p. 202−204°C. − ^{1}H NMR (250 MHz): δ = 1.69−1.78 (m, 4 H; pyrr-β-CH_2), 1.95−2.05 (m, 4 H, pyrr-β′-CH_2), 2.51−2.55 (m, 4 H, ring CH_2), 2.93−3.02 (m, 2 H, bridgehead CH), 3.13−3.22 (m, 4 H, pyrr-α-CH_2), 3.49−3.57 (m, 4 H, pyrr-α′-CH_2), 3.68 (s, 6 H, OCH_3), 5.40 (dd, J = 10.1 and 5.6 Hz, 2 H, =CH), 6.42 (d, J = 10.1, 2 H, =CH). − ^{13}C NMR: δ = 25.3 (pyrr-β-CH_2), 37.6 (ring CH_2), 50.8 (OCH_3), 52.1 (pyrr-α-CH_2), 56.0 (bridge CH), 96.7 (=*C*−COOR), 126.3 (=CH), 131.4 (=CH), 164.5 (=C−N), 165.4 (COOR). − MS; *m/z* (%): 412 (26) [M$^+$], 353 (26), 282 (20), 219 (24), 218 (49), 207 (34), 206 (100), 193 (20), 192 (20), 148 (66), 147 (30), 146 (24), 96 (26), 70 (25). − $C_{24}H_{32}N_2O_4$ (412.5): calcd. C 69.88, H 7.82, N 6.79; found C 69.87, H 7.82, N 7.00.

Tetramethyl 5,11-Dipyrrolidinobicyclo[5.5.0]dodeca-2,4,8,10-tetraene-3,4,9,10-tetracarboxylate (6b) and Tetramethyl 5,9-Dipyrrolidinobicyclo[5.5.0]dodeca-2,4,9,11-tetraene-3,4,10,11-tetracarboxylate (7a) and 5,9-Tetramethyl 5,9-Dipyrrolidinobicyclo[5.5.0]dodeca-3,5,9,11-tetraene-3,4,10,11-tetracarboxylate (7b) (Rapidly Interconverting Positional Isomers): Similarly to the preparation of **6a**, compound **3a** (4.68 g, 19.2 mmol) was treated with dimethyl butynedioate (5.46 g, 38.5 mmol). After heating at 90°C as above for 1 h, the mixture was cooled to 0°C for 16 h. (a) The separated yellowish solid was removed by suction, washed with ether, and crystallized from chloroform/methanol to yield colorless needles of **7a/b**, m.p. 221°C, yield 2.5 g (24%). − IR: $\tilde{\nu}$ = 1716 cm^{-1}, 1673, 1596, 1536. − UV: λ_{max} = 315 nm. − ^{1}H NMR (250 MHz; all signals br. at room temp.): δ = ca. 1.80 (2 H), ca. 1.88 (8 H), ca. 2.55 (4 H), 2.98 (q, J = 7.4 Hz, 2 H), ca. 3.59 (s, 6 H), ca. 3.70 (s, 6 H), ca. 6.85 (2 H). − ^{13}C NMR: see Table 1. − MS; *m/z* (%): 528 (69) [M$^+$], 499 (24), 469 (100), 458 (38), 437 (21), 264 (23), 96 (34). − $C_{28}H_{36}N_2O_8$ (528.6): calcd. C 63.62, H 6.86; found C 63.59, H 6.90. − (b) The filtrate from **7a/b** was concentrated in vacuo, and the remaining dark-red oil was crystallized from methanol to give **6b** as a yellow-orange crystalline solid. M.p. 285−287°C (dec.); yield varying between 33 and 73% in different runs. − IR: $\tilde{\nu}$ = 1724 cm^{-1}, 1693, 1600, 1531. − UV: λ_{max} = 312 nm. − ^{1}H NMR (250 MHz): δ =

1.63−1.83 (m, 4 H; pyrr-β-CH_2), 2.02−2.17 (m, 4 H; pyrr-β′-CH_2), 2.41−2.51 (m, 2 H, ring CH_2), 2.64−2.68 (m, 2 H, ring CH_2), 2.97−3.06 (m, 2 H, bridgehead CH), 3.07−3.17 (m, 4 H, pyrr-α-CH_2), 3.44−3.55 (m, 4 H, (pyrr-α′-CH_2), 3.60 (s, 6 H, OCH_3), 3.73 (s, 6 H, OCH_3), 6.40 (d, J = 6.2 Hz, 2 H, =CH). − ^{13}C NMR: see Table 1. − MS; *m/z* (%): 528.7 (19) [M^+], 527.7 (56), 499 (22), 470 (33), 469 (100), 400 (31), 398 (25), 276 (25), 264 (49), 232 (34), 218 (30), 206 (46), 205 (27), 193 (68), 192 (31), 96 (69), 70 (35), 59 (26), 55 (27). − $C_{28}H_{36}N_2O_8$ (528.6): calcd. C 63.62, H 6.86, N 5.30; found C 63.51, H 6.85, N 5.48.

Tetramethyl 2,8-*syn*-6,12-Dioxotetracyclo[7.3.0.0^{3,7}.0^{4,10}]dodecane-1,2,7,8-tetracarboxylate (10): A suspension of **6b** (3 g, 5.68 mmol) in acetic acid (3 mL) and water (15 mL) was stirred for 24 h at room temp. The starting material dissolved gradually, and a colorless solid deposited. The mixture was extracted with chloroform (3 × 30 mL). The combined extracts were washed with dilute aqueous HCl and saturated aqueous NaCl, dried (Na_2SO_4) and concentrated. The residue was crystallized from chloroform/ethyl acetate, m.p. 210°C; yield 1.41 g (59%). − ^{1}H NMR: δ = 3.72 (s, 6 H, OCH_3), 3.67 (s, 6 H, OCH_3), 3.00 (m, 2 H, 3-H), 2.94 (d, J = 3.1 Hz, 2 H, 2-H), 2.62 (dd, J = 18.9 and 7.2 Hz, 5-H), 2.41 (d, J = 18.9, 2 H, 5-H′), 2.24 (dd, J = 7.2 and 4.4 Hz, 2 H, 4-H). − ^{13}C NMR: δ = 40.6 (C-4,-10), 42.0 (C-2,-8), 44.7 (C-5,-11), 52.3 (OCH_3), 52.6 (OCH_3), 57.6 (C-1,-7), 168.1 (COOR), 170.1 (COOR), 210.9 (CO). − MS; *m/z* (%): 422 (2.5) [M^+], 394 (100), 391 (53), 362 (24), 331 (21), 302 (21), 243 (24), 221 (95), 209 (46), 195 (27), 163 (72), 81 (79), 59 (66). − $C_{20}H_{22}O_{10}$·$CHCl_3$ (541.8): calcd. C 46.56, H 4.28; found C 46.33, H 4.29.

Acknowledgments

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