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CYCLOBUTANONE-BASED TANDEM FREE RADICAL REARRANGEMENTS: FORMATION OF BICYCLIC AND TRICYCLIC KETONES

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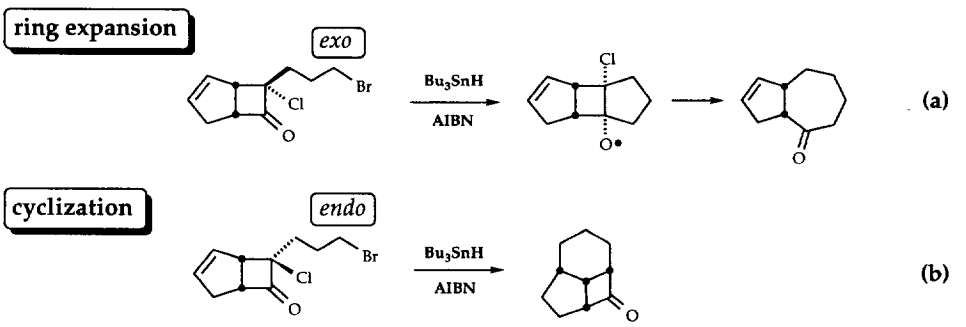
Abstract: *Free radical reaction of endo-bromopropylbicyclo[4.2.0]oct-2-en-7-one* **1** *leads to deep-seated rearrangement and formation of the bridged tricyclic ketone* **2**. *An authentic sample of* **2** *was prepared. Reaction of endo-bromoethylbicyclo[4.2.0]oct-2-en-7-one* **18** *generates an analogous bridged tricyclic ketone* **20** *together with cis-bicyclo[4.4.0]decenone* **22**. *Experiments using* Bu_3SnD *were carried out to explore the mechanism of the transformation leading to* **22**.

Introduction

Free radical ring expansion and annulation of substituted cyclobutanones has proved to be both synthetically useful and mechanistically interesting.[1-3] In the present series, treatment of cyclic dienes with ω-bromoalkyl ketenes leads to a mixture of *exo*- and *endo*-haloalkyl fused cyclobutanones (Scheme 1). The *exo*-haloalkylcyclobutanones undergo smooth ring expansion

Scheme 1

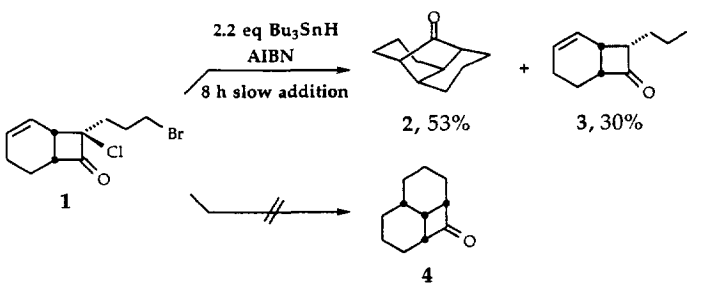


in a sequence consisting of addition of the primary radical to the carbonyl group followed by β-scission of the alkoxy radical to afford the *cis*-fused bicyclic product (Scheme 1a).[2] The *endo*-haloalkylcyclobutanones produce tricyclic products following cyclization of the alkyl radical to the

double bond (Scheme 1b).[2b,4] The *cis*-fused bicyclic ring system and the *endo*-orientation of the side chain make the cycloaddition a favorable process.

Results and Discussion

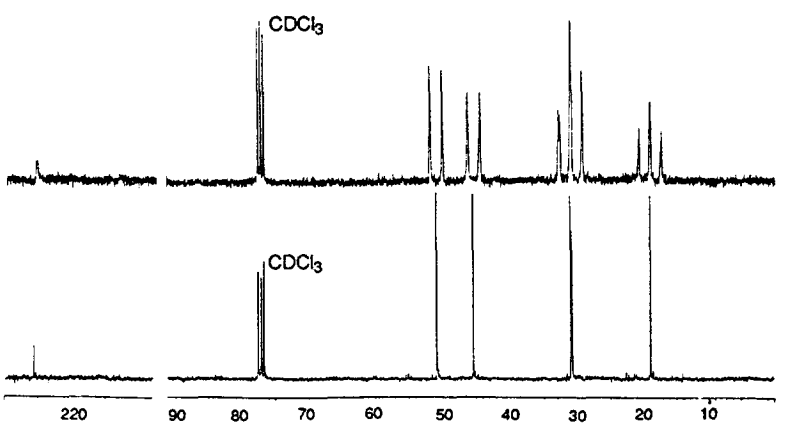
Presuming on the basis of the outcome of the cyclization of *endo*-halopropylbicyclo-[3.2.0]hept-2-en-6-one in Scheme 1b, we anticipated that the direct cyclization product **4** would be



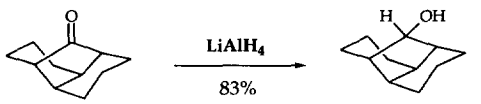
(1)

the result of the free radical cyclization reaction of *endo*-bromopropylbicyclo[4.2.0]oct-2-en-7-one **1**. However, when **1** was treated with 2.2 eq of Bu_3SnH and AIBN (eq 1), none of the direct cyclization product **4** was observed, instead, **2** was isolated as the major product, together with a minor amount of direct reduction product **3**. The IR spectrum of **2** showed a strong band at 1742 cm^{-1}, clearly indicating a cyclopentanone, not a cyclobutanone carbonyl. Moreover, the decoupled ^{13}C NMR spectrum of **2** (Figure 1) showed only six lines, at δ 19.0 (t), 30.8 (t), 31.0 (t),

Figure 1. Coupled and Decoupled ^{13}C NMR Spectra of 2



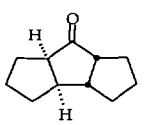
45.4 (d), 51.0 (d) and 226.0 (s), instead of the seven lines expected of the cyclization product **4**. of the When **2** was reduced with $LiAlH_4$ (eq 2), the decoupled ^{13}C NMR spectrum alcohol **5**



2 **5** (2)

showed eleven lines. Therefore, the alcohol **5** cannot be C_2 symmetric. Following these revelations, the rearrangement product was assigned the bridged tricyclic ketone structure **2** shown in eqs 1 and 2.

The linear, *anti*-fused tricyclic ketone **6-*anti*** is also a candidate for the rearrangement

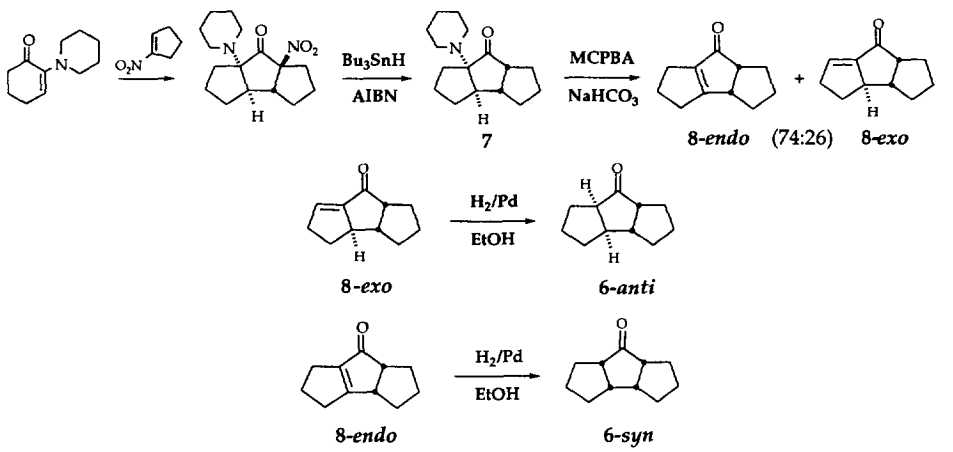


6-*anti*

product of eq 1 on the basis of its C_2 symmetry and the lack of C_2 symmetry of the corresponding alcohol. Thus, **6-*anti*** and **2** will have very similar spectral properties. To the best of our knowledge, neither **2** nor **6-*anti*** has been synthesized, and no literature data is available that might be used to established which is the rearrangement product of eq 1.

Preparation of authentic **6-*anti*** and its diastereomer **6-*syn*** is shown in Scheme 2. Aminoketone **7** was synthesized following Huffman's procedure. Under Huffman's piperidine

Scheme 2. Preparation of Authentic 6-*Anti* and 6-*Syn*



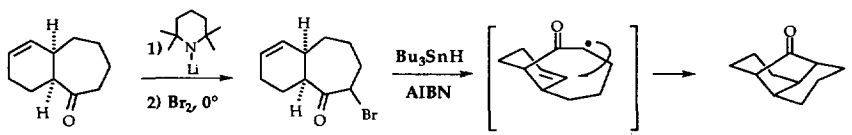
7 **8-*endo*** (74:26) **8-*exo*** (a)

8-*exo* **6-*anti*** (b)

8-*endo* **6-*syn*** (c)

oxide elimination conditions,[6] **8-*endo*** (*endo* and *exo* refer to the position of the double bond with respect to the center ring) was isolated as the sole product. By reducing the reaction time from one week to three days, we observed both unsaturated ketones **8-*endo*** and **8-*exo*** as a separable 74:26 mixture. An authentic sample of **6-*anti*** was prepared by hydrogenation of **8-*exo*** (Scheme 2b). Diastereomer **6-*syn*** was also prepared by hydrogenation of **8-*endo*** (Scheme 2c). Although the spectral properties of authentic **6-*anti*** were similar to those of **2**, there were distinct differences, particularly in the ^{13}C NMR spectra, that allowed us to rule out **6-*anti*** as the structure of the rearrangement product **2**.

A sequence was then devised to prepare authentic **2**. Using lithium tetramethylpiperidide as a hindered base, a new method was developed for selective bromination of the *cis*-bicyclo[5.4.0]undec-8-en-2-one[2b] without disturbing the *cis*-fused stereochemistry of the bicyclic system (eq 3).[7] An authentic sample of **2** was then prepared by internal, free radical cyclization of

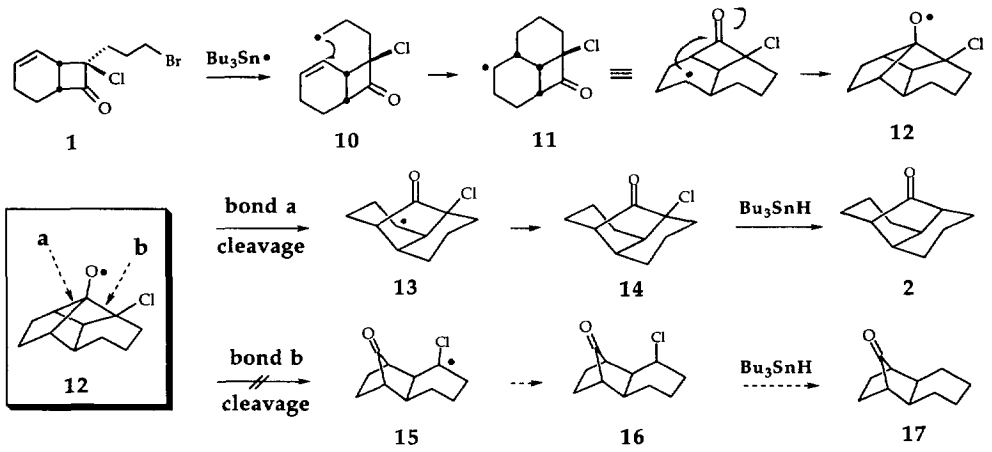


(3)

the α-bromobicyclic ketone **9** (eq 3). The spectral properties of authentic **2** are identical with those of the rearrangement product of eq 1, assigned the tricyclo[5.4.0.0^{3,8}]undecan-2-one structure.

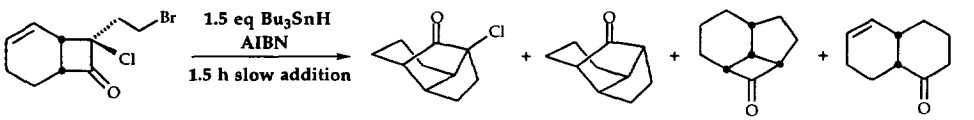
A mechanism for the novel rearrangement of eq 1 might be formulated as follows (Scheme 3). Upon treatment of **1** with Bu_3SnH and AIBN, a primary carbon radical **10** is

Scheme 3



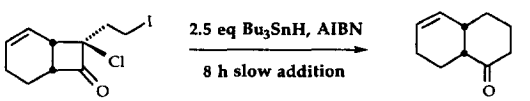
generated that adds to the double bond to form the cyclized radical **11**. The cup-shaped *endo*-tricyclic ring system makes it possible for the cyclohexyl radical **11** to attack the cyclobutanone carbonyl leading to alkoxy radical **12**.[8,9] β-Cleavage at **bond a** provides the greatest relief of strain and leads to the formation of **14**, which can be isolated or further reduced to **2** depending upon the number of equivalents of Bu_3SnH employed. The spectral properties of **14** (see experimental) are fully consistent with the structure formulated in Scheme 3. Compound **17**, which would be generated by cleavage of **bond b** of the alkoxy radical **12**, was not detected. The latter path may not be as favorable as the cleavage of **bond a** in the release of ring strain.

A cognate rearrangement was observed with the cyclohexenyl-fused system **18** that has a bromoethyl side chain (eq 4). Upon tributyltin hydride treatment, a mixture of bridged tricyclic

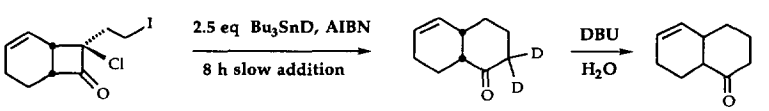


18 **19**, 26% **20**, 3% **21**, 8% **22**, 7% (4)

ketones (**19** and **20**) was generated, accompanied by a small amount of direct cyclization product **21** and *cis*-bicyclo[4.4.0]decenone **22**. When the iodide **23** was used as a radical precursor, slow addition of 2.5 eq of Bu_3SnH (eq 5) (or Bu_3SnD, eq 6) produced *cis*-bicyclodecenone **22** (or **22-3-d_2**)



23 **22**, 57% (5)



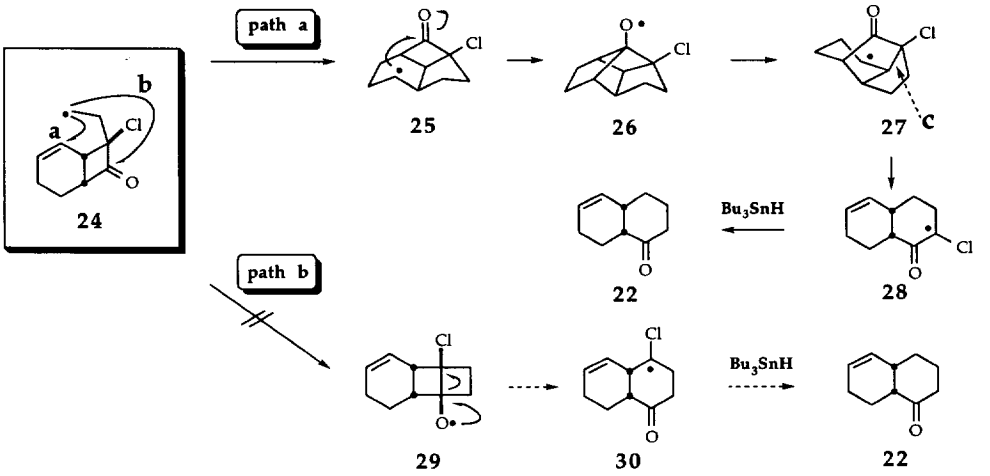
23 **22-3-d_2**, 53% **22** (6)

as the major product. For formation of **22**, the iodide is a better radical precursor than the bromide,[10] because it allows the chain reaction to be sustained at lower concentrations of Bu_3SnH.

Formation of bicyclodecenone **22** can be explained by the cleavage of **bond c** of the intermediate radical **27** (Scheme 4, path a). This process is favored by release of ring strain of the bridged ring and produces the bicyclo[4.4.0]decenone radical **28**, which is stabilized by both α-acyl and chloro substituents.

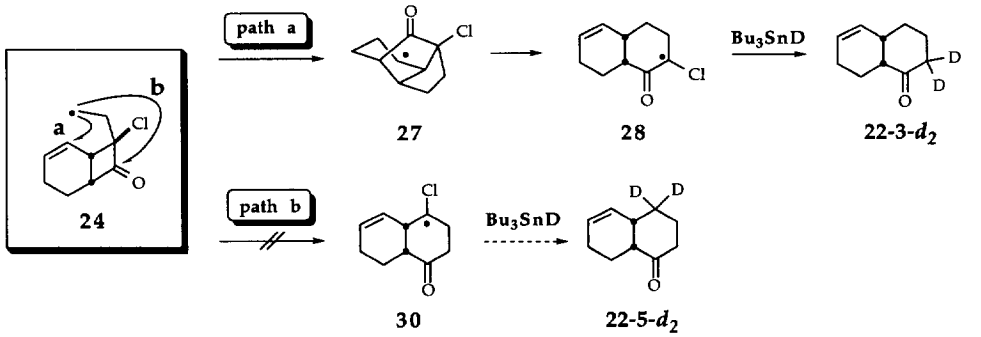
Scheme 4 shows another possible path leading to the formation of **22** (path b) that

Scheme 4



proceeds by a direct two-carbon ring expansion. An analogous process earlier proved to be unfavorable in the ring expansion of β-keto esters.[8e] Moreover, in the fused cyclobutanone system, only *exo*-haloalkylcyclobutanones undergo ring expansion; no ring expansion from the *endo*-isomers has been observed.[2a,b] Accordingly, an experiment with Bu_3SnD was carried out that yielded product with two deuterium atoms incorporated α to the carbonyl; no deuterium was found at the γ position (Scheme 5, path a). That deuterium incorporation occurs adjacent to

Scheme 5



the carbonyl was established by ^1H NMR and GC-MS following exchange with H_2O under basic conditions (DBU/ether). The deuterium in the product was completely exchanged under these conditions (eq 6). This result confirms the conclusion that the formation of **22** proceeds through path a.

In summary, we have discovered a novel cyclobutanone-based tandem free radical reaction that leads by deep-seated rearrangement to the formation of bicyclic and tricyclic ketones.

Experimental Section

All reactions were performed under a nitrogen atmosphere unless otherwise noted. Tetrahydrofuran (THF), benzene, and diethyl ether were distilled from blue or purple solutions of sodium benzophenone ketyl under nitrogen. Tri-*n*-butyltin hydride (Bu_3SnH) and *n*-butyllithium (1.6 M in hexanes) were purchased from the Aldrich Chemical Co. and used without further purification. 2,2'-Azobis(2-methylpropionitrile) (AIBN) was purchased from Alfa. For flash chromatography, 400-230 mesh silica gel 60 (E. Merck no. 9385) was employed.

Nuclear magnetic resonance (NMR) spectra were obtained on Bruker AC-300, or IBM AF-300 spectrometers (300 MHz for ^1H NMR, 75 MHz for ^{13}C NMR). Infrared (IR) spectra were obtained on an IBM IR/32 FTIR spectrometer. Gas chromatography and low resolution mass spectra (GC-MS) were obtained using a Hewlett-Packard (5890 series II) gas chromatograph equipped with a Hewlett-Packard (5970 series) mass spectrometer. High-resolution mass spectra were obtained on Varian MAT CH-5DF, or VG-70G spectrometers.

Endo-bromopropylbicyclo[4.2.0]oct-2-en-7-one **1**, *endo*-bromoethylbicyclo[4.2.0]oct-2-en-7-one **18**, and *endo*-iodoethylbicyclo[4.2.0]oct-2-en-7-one **23** were prepared following reported procedures.[2a,b]

Radical Reaction of (*1SR,6RS,8SR*)-8-(3'-Bromopropyl)-8-chlorobicyclo[4.2.0]oct-2-en-7-one (1)

A solution of Bu_3SnH (592 μL, 2.2 mmol) and AIBN (16 mg) in 30 mL of benzene was added to a refluxing solution of **1** (276 mg, 1.0 mmol) in 10 mL of benzene over 8 h. The reaction mixture was refluxed for an additional 1 h. After DBU workup,[11] flash chromatography (30:1 hexanes-ether) of the crude product gave rearrangement product **2** (87 mg, 53%) and direct reduction product **3**[2b] (49 mg, 30%).

Data for **2**: ^1H NMR ($CDCl_3$) δ 1.27-1.67 (m, 6 H), 1.68 (m, 4 H), 1.93 (m, 2 H), 2.04 (m, 2 H), 2.17 (m, 2 H). ^{13}C NMR ($CDCl_3$) δ 19.0 (t, J=125 Hz), 30.8 (t, J=128 Hz), 31.0 (t, J=128 Hz), 45.4 (d, J=134 Hz), 51.0 (d, J=140 Hz), 226.0 (s). IR (neat) 1741 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 164 (M^+, 98), 146 (37), 136 (27), 121 (40), 104 (49), 94 (60), 79 (100), 67 (64). HRMS calcd for $C_{11}H_{16}O$: 164.1201. Found 164.1191.

This experiment was repeated using 1.2 equivalent of Bu_3SnH to produce the chlorine-bearing rearrangement product **14** in 69% yield. ^1H NMR ($CDCl_3$) δ 1.25-2.09 (m, 10 H), 2.13 (m, 2 H), 2.19 (br s, 1 H), 2.24 (br s, 1 H), 2.3 (br s, 1 H). ^{13}C NMR ($CDCl_3$) δ 18.6 (t, J=130 Hz), 21.1 (t, J=123 Hz), 28.9 (t. J=132 Hz), 29.6 (t, J=137 Hz), 31.6 (t, J=136 Hz), 40.8 (t, J=131 Hz), 43.9 (d, J=139 Hz), 49.7

(d, *J*=142 Hz), 50.8 (d, *J*=139 Hz), 75.0 (s), 216.4 (s). IR (neat) 1759 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 200 (7), 198 (M^+, 23), 163 (19), 145 (15), 135 (100), 105 (12), 91 (27), 79 (25), 67 (35). HRMS calcd for $C_{11}H_{15}{}^{35}ClO$: 198.0811. Found 198.0818.

$LiAlH_4$ Reduction of 2

To a solution of **2** (130 mg, 0.8 mmol) in 5 mL of ether was added $LiAlH_4$ (48 mg, 12.8 mmol) at 0 °C. After stirring at this temperature for 1 h, the reaction was quenched with wet ether. The reaction mixture was worked up by extraction with ether to give an alcohol **5** (110 mg, 83%) as a white solid. Recrystalization of the alcohol from hexanes gave fine needle-like crystals (mp. 80-82 °C). Sublimation of the alcohol under vacuum (0.02 mmHg, 30-35 °C) afforded fine needles (mp. 82-84 °C). ^1H NMR ($CDCl_3$) δ 1.36-2.00 (m, 16 H), 2.14 (br s, 1 H), 4.42 (d, *J*=6.7 Hz, 1 H). ^{13}C NMR ($CDCl_3$) δ 19.2 (t, *J*=132 Hz), 19.4 (t, *J*=125 Hz), 28.5 (t, *J*=130 Hz), 30.9 (t, *J*=130 Hz), 31.2 (t, *J*=130 Hz), 32.6 (t, *J*=131 Hz), 45.3 (d, *J*=132 Hz), 46.8 (d, *J*=130 Hz), 47.8 (d, *J*=130 Hz), 48.6 (d, *J*=132 Hz), 76.0 (s). IR (neat) 3247 (br s, OH) cm^{-1}. MS *m/e* (rel. intensity) 166 (M^+, 8), 148 (80), 135 (67), 119 (35), 107 (23), 91 (43), 79 (76), 70 (100), 67 (64). HRMS calcd for $C_{11}H_{18}O$ 166.1358. Found 166.1353.

Preparation of Authentic 6-*Anti* and 6-*Syn*

The aminoketone **7** was prepared following the reported procedure.[6b]

A solution of **7** (170 mg, 0.7 mmol) and *meta*-chloroperbenzoic acid (80%, 150 mg, 0.7 mmol) in 20 mL of CH_2Cl_2 was heated at reflux for 3 days in the presence of 5 mL 0.1 M aqueous $NaHCO_3$. The CH_2Cl_2 was separated and sequentially washed with 0.1 M aqueous of HCl and 0.1 M aqueous Na_2CO_3. After drying over $MgSO_4$, the solvent was evaporated. Flash chromatography (10:1 hexanes-ether) of the crude product gave **8-*exo*** (12 mg, 11%) and **8-*endo*** (61 mg, 55%). Data for **8-*exo***: ^1H NMR ($CDCl_3$) δ 1.2-2.75 (11 H), 2.88 (m, 1 H), 2.97 (m, 1 H), 6.46 (dd, *J*= 7.3 and 3.0 Hz, 1 H). IR (neat) 1711 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 162 (M^+, 37), 147 (7), 134 (11), 119 (9), 105 (13), 95 (100), 77 (17). Data for **8-*endo***:[6b] ^1H NMR ($CDCl_3$) δ 1.25-2.40 (12 H), 2.48 (m, 1 H), 3.12 (m, 1 H). ^{13}C NMR ($CDCl_3$) δ 24.3 (t, *J*=130 Hz), 24.6 (t, *J*=133 Hz), 27.7 (t, *J*=130 Hz), 27.9 (t, *J*=130 Hz), 29.4 (t, *J*=127 Hz), 30.4 (t, *J*=132 Hz), 43.0 (d, *J*=137 Hz), 57.6 (d, *J*=134 Hz), 149.4 (s), 189.0 (s), 206.5 (s). IR (neat) 1696 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 162 (M^+, 39), 161 (17), 147 (4), 134 (100), 121 (5), 106 (11), 91 (30).

A solution of **8-*exo*** (12 mg, 0.07 mmol) in 1 mL of ethanol was stirred with 10% Pd/C (5 mg) under a hydrogen balloon for 40 min. The catalyst was removed by filtration to gave **6-*anti*** (10 mg, 87%). ^1H NMR ($CDCl_3$) δ 1.30-2.00 (m, 12 H), 2.38 (q, *J*= 7.4 Hz, 2 H), 2.64 (td, *J*=7.4 and 3.9 Hz, 2 H). ^{13}C NMR ($CDCl_3$) δ 26.3, 30.3, 35.0, 46.6, 52.8, 222.4. IR (neat) 1730 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 164 (M^+, 30), 146 (1), 136 (18), 123 (15), 107 (9), 95 (100), 79 (26), 67 (70). HRMS calcd

for $C_{11}H_{16}O$ 164.1201. Found 164.1188.

A solution of **8-*endo*** (28 mg, 0.17 mmol) in 2 mL of ethanol was stirred with 10% Pd/C (7 mg) under a hydrogen balloon for 1 h. The catalyst was removed by filtration to gave **6-*syn*** (27 mg, 97%). ^{1}H NMR ($CDCl_3$) δ 1.35 (m, *J*=7.0, 2 H), 1.51 (m, *J*= 7.0, 4 H), 1.68 (m, 4 H), 1.82 (m, 2 H), 2.85 (br s, 4 H). ^{13}C NMR ($CDCl_3$) δ 27.3 (t, *J*=130), 27.3 (t, *J*=130), 29.3 (t, *J*=131), 41.7 (d, *J*=138), 54.6 (d, *J*=133), 223.6 (s). IR (neat) 1734 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 164 (M^+, 21), 146 (1), 136 (8), 123 (6), 107 (11), 96 (100), 79 (22), 67 (42).

Preparation of Authentic 2

To a solution of tetramethylpiperidine (82 μL, 0.48 mmol) in 2 mL of THF was added a solution of *n*-butyl lithium (1.6 M in hexane, 0.33 mL, 0.53 mmol) at 0 °C. After stirring at this temperature for 40 min, *cis*-bicyclo[5.4.0]undec-8-en-2-one[2b] (73 mg, 0.44 mmol) in 1 mL of THF was added, followed by the addition of bromine (83.6 mg, 0.53 mmol). After ether workup of the reaction mixture, flash chromatography (40:1 hexanes-ether) of the crude product gave bromide **9** (58 mg, 53%) as a single diastereomer. ^{1}H NMR ($CDCl_3$) δ 1.00-2.15 (10 H), 3.06-3.18 (2 H), 4.64 (dd, *J*=5.6, and 2.0 Hz, 1 H), 5.57 (m, 1 H), 5.73 (m, 1 H). ^{13}C NMR ($CDCl_3$) δ 20.7 (t, *J*=132 Hz), 24.2 (t, *J*=130 Hz), 25.7 (t, *J*=130 Hz), 32.1 (t, *J*=130 Hz), 34.2 (d, *J*=131 Hz), 34.5 (t, *J*=129 Hz), 47.0 (d, *J*=120 Hz), 57.0 (d, *J*=151 Hz), 126.6 (d, *J*=157 Hz), 131.0 (d, *J*=159 Hz), 208.5 (s). IR (neat) 1694 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 244 and 242 (M^+, 13), 163 (M^+-Br, 49), 145 (87), 135 (25), 119 (19), 107 (53), 79 (100). HRMS calcd for $C_{11}H_{15}{}^{79}BrO$ 242.0306. Found 242.0245.

A solution of Bu_3SnH (41.4 μL, 0.15 mmol) and AIBN (2 mg) in 2 mL of benzene was added to a refluxing solution of bromoketone **9** (25.0 mg, 0.1 mmol) in 2.5 mL of benzene over 6 h. The reaction mixture was refluxed for an additional 2 h. After DBU workup, flash chromatography (40:1 hexanes-ether) of the crude product gave annulation product **2** (12.8 mg, 78%). The ^{1}H NMR, ^{13}C NMR, IR, and MS of **2** obtained from this reaction are in excellent agreement with those of the **2** obtained from the rearrangement reaction of **1** (eq 1).

Radical Reaction of (*1SR,6RS,8SR*)-8-(2'-Bromoethyl)-8-chlorobicyclo[4.2.0]oct-2-en-7-one (18)

A solution of Bu_3SnH (0.5 mL, 1.9 mmol) and AIBN (20 mg) in 10 mL of benzene was added to a refluxing solution of **18** (337 mg, 1.3 mmol) in 55 mL of benzene over 1.5 h. The reaction mixture was refluxed for an additional 2 h. After DBU workup, flash chromatography (40:1 hexanes-ether) of the crude product gave chlorine-bearing rearrangement product **19** (60 mg, 26%), rearrangement product **20** (5 mg, 3%), tricyclic ketone **21**[2b] (16 mg, 8%), and *cis*-bicyclo[4.4.0]dec-7-en-2-one **(22)** (13 mg, 7%).

Data for chlorine-bearing rearrangement product **19**: ^{1}H NMR ($CDCl_3$) δ 1.24-1.58 (m, 4 H), 1.84-2.08 (m, 6 H), 2.17-2.20 (m, 1 H), 2.38-2.42 (m, 2 H). ^{13}C NMR ($CDCl_3$) δ 17.8(t), 22.8 (t), 26.2 (t),

28.3 (t), 34.7 (t), 41.7 (d), 51.1 (d), 51.8 (d), 76.0 (s), 213.3 (s). IR (neat) 1763 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 186 (14), 184 (M^+, 42), 149 (M^+-Cl, 18), 121 (21), 119 (100), 93 (53), 91 (58), 79 (93). HRMS calcd for $C_{10}H_{13}{}^{35}ClO$ 184.0655. Found 184.0667.

Data for rearrangement product **20**: ^1H NMR ($CDCl_3$) δ 0.82-0.87 (m, 1 H), 0.95-1.06 (m, 1 H), 1.25-1.31 (m, 2 H), 1.42-1.63 (m, 4 H), 1.78-1.94 (m, 3 H), 2.05-2.08 (m, 1 H), 2.17-2.21 (m, 1 H), 2.40-2.41 (m, 1 H). ^{13}C NMR ($CDCl_3$) δ 18.6 (t), 25.2 (t), 26.4 (t), 26.4 (t), 28.3 (t), 45.3 (d), 47.0 (d), 51.6 (d), 53.9 (d). IR (neat) 1740 (s, C=O) cm^{-1}. MS *m/e* (rel. intensity) 150 (M^+, 78), 122 (8), 104 (55), 93 (81), 79 (100), 67 (28), 41 (32).

The ^1H NMR, ^{13}C NMR, IR, and MS of **22** agree well with those of an authentic sample prepared following the method of Oppolzer.[12]

Radical Reaction of (*1SR,6RS,8SR*)-8-(2'-Iodoethyl)-8-chlorobicyclo[4.2.0]oct-2-en-7-one (23)

A solution of Bu_3SnH (267 μL, 0.99 mmol) and AIBN (7 mg) in 3 mL of benzene was added to a refluxing solution of **23** (140 mg, 0.45 mmol) in 20 mL of benzene over 6 h. The reaction mixture was refluxed for an additional 1 h. After DBU workup, flash chromatography (30:1 hexanes-ether) of the crude product gave **22** (38 mg, 57%).

This reaction was repeated using Bu_3SnD instead of Bu_3SnH and gave **22-3-d_2** in 53% yield. ^1H NMR ($CDCl_3$) 1.40-2.20 (m, 8 H), 2.53 (m, 1 H), 2.70 (m, 1 H), 5.52 (m, 1 H), 5.72 (m, 1 H). MS *m/e* (rel. intensity) 152 (M^+, 100), 137 (71), 120 (41), 105 (47), 93 (82), 79 (96). HRMS calcd for $C_{10}D_2H_{12}O$ 152.1170. Found 152.1156.

Deuterium-Hydrogen Exchange Experiment

To a solution of **22-3-d_2** (6 mg) in 1 mL of ether was added 1 mL of a 10% aqueous solution of DBU. After stirring at 25 °C for 24 h, a GC-MS showed the two deuteriums in **22-3-d_2** were completely exchanged to form **22**.

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