Reactions of Spiro[2.6]nona-4,6,8-triene Derivatives with Acids and Bases. Rearrangement to a Heptafulvene Derivative and Formation of Ether Linkage Compounds

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Reaction of dimethyl *trans*-spiro[2.6]nona-4,6,8-trien-1,2-dicarboxylate with phenyllithium afforded *trans*-1,2-bis(hydroxydiphenylmethyl)spiro[2.6]nona-4,6,8-triene (**2**) in a good yield. Spirotriene **2** is very sensitive to acids and gave 8-(2,2-diphenylvinyl)heptafulvene accompanied with a quantitative amount of benzophenone by acid treatments. Although **2** is stable to basic reagents, upon treating with sodium hydride, **2** afforded 11-hydroxydiphenylmethyl-8-oxa-9-diphenyltricyclo[5.4.0.0^{1,10}]undeca-2,4-diene, and its double bond isomers. Mechanisms of the above reactions are discussed.

Spiro[2.6]nona-4,6,8-triene system is of interest from the view point of spiro conjugation.[1,2] Although some spiro[2.6]nonatriene derivatives have been synthesized,[3—9] chemical properties of this class of compounds have not been well known in detail.[7—11] We designed to introduce hydroxyl groups to this system and to investigate its properties to acids and bases. We wish to report here the results obtained and discuss about mechanisms of these reactions.

Reductions of dimethyl *trans*-spiro[2.6]nona-4,6,8-trien-1,2-dicarboxylate (**1**)[7,8] by lithium aluminium hydride gave several unidentified mixtures, and reactions of **1** with Grignard reagents (benzylmagnesium chloride or methylmagnesium iodide) also gave several intractable products which did not possess a cyclopropane moiety or a cycloheptatriene ring. Presumably skeletal rearrangements took place by the actions of the Grignard reagents or lithium aluminium hydride as Lewis acids. However, *trans*-1,2-bis(hydroxydiphenylmethyl)spiro[2.6]nona-4,6,8-triene (**2**) was obtained in 80% yield by reaction of **1** with excess phenyllithium.

Spirotriene **2** is very sensitive toward acidic reagents. When **2** was treated with organic or mineral acids, or even with silica gel, the colorless solution of **2** turned to dark red, and it gave a product (**3**) which possesses a strong absorption band at 385 nm in its UV spectrum, and is a very unstable entity. When **3** was allowed to stand at room temperature in neat, the color of **3** disappeared within one hour as shown in Fig. 1, and a mixture of resinous substances and one mole of benzophenone were obtained.

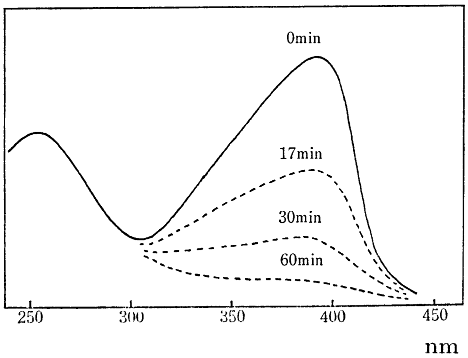

Fig. 1.

Considering that spiro[2.6]nonatriene derivatives have been known to give heptafulvene derivatives by acid treatments[3,4] and the UV spectrum of **3** is similar to those of heptafulvene derivatives,[12] structure of **3** was supposed to be 8-(2,2-diphenylvinyl)heptafulvene. Exhaustive catalytic hydrogenation of a freshly prepared reaction mixture of **2** with silica gel gave a hydrocarbon (**4**) in 75% yield and benzophenone in a good yield, after consumption of five mole of hydrogen. Hydrocarbon **4** shows the following spectral properties. Mass: *m/e*, 292 (9%, molecular ion peak, $C_{22}H_{28}$) and *m/e*, 167 (base peak, Ph_2CH^+). NMR (in CCl_4): δ ppm; 1.8—2.3 (multiplet, 17 H), 3.8 (triplet, 1 H, benzyl proton), and 7.2 (broad singlet, 10 H). These data suggest that the structure of **4** is 1-cycloheptyl-3,3-diphenylpropane, confirming the structure of **3** to be 8-(2,2-diphenylvinyl)heptafulvene. Scheme of these reactions is shown in Fig. 2.

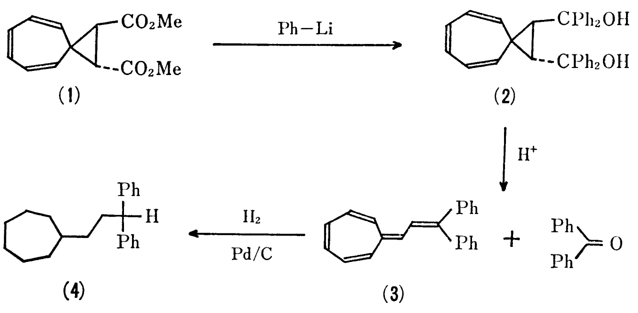

Fig. 2.

Spirotriene **2** is stable toward basic reagents. Reactions of **2** with refluxing pyridine, triethylamine, or 4 M sodium hydroxide in refluxing dioxane resulted in complete recovery of **2**. On the other hand, treatment of **2** with excess sodium hydride in dry dioxane afforded colorless crystalline substances (**5**, **6**, and **7**). Products ratios depended on the temperature and the reaction period as shown in Table 1.

Physical data of **5**, **6**, and **7** are summarized in Table 2—4. These products show the same elemental analyses ($C_{35}H_{30}O_2$) and molecular ion peaks in mass spectra (M^+, 482). Acid treatments of these products gave **3** and benzophenone, suggesting the structures of them to be analogous to that of **2**. The IR and NMR spectral studies in deuterium oxide cleared that each

Table 1.

Condition		Product yields (%) 2 (recv.)	5	6	7
Room temp	300 h	12	63	0	0
Reflux	6.5 h	0	4	44	17
Reflux	20 h	0	0	0	43

Table 2.

	UV λ_{max}^{MeOH} nm (log ε)	IR ν^{KBr} cm^{-1}	Mass m/e (%)
5	255 (4.18)	3550 1600	482(M$^+$, 3), 269(9), 197(16), 196(100), 167(8), 105(40)
6	260 (4.07)	3595 1590	482(M$^+$, 2), 269(9), 196(100) 118(17), 105(78)
7	277 (4.10)	3600 1600	482(M$^+$, 5), 286(100), 105(84)

compound possesses one hydroxyl group and the other oxygen is in an ether linkage.

Structures of **5**, **6**, and **7** were determined to be 11-hydroxydiphenylmethyl-8-oxa-9,9-diphenyltricyclo-[5.4.0.0^{1,10}]undeca-3,5-diene, 2,4-diene, and 4,6-diene, respectively, as shown in Fig. 3, from their NMR spectra with double and triple resonance techniques. Assignment of the NMR spectra were deduced from their coupling patterns which clearly showed each proton's relations in the seven membered rings of them as shown in Tables 3 and 4. Investigation of their Dreiding model indicated that the chemical shifts of H_{11} should appear more lower fields than those of H_{10} by the different anisotropy effect of the phenyl groups at C_9 and C_{12}.

These structures could reasonably be explained also by their UV spectra. The absorption maxima of them shift to longer wavelength in the order of **5**, **6**, and **7**, being well coincident with that **5** has only one isolated diene chromophore, but **6** and **7** possess diene systems conjugated with a cyclopropane and a terminal vinyl ether linkage conjugated with a cyclopropane, respectively.

Thermal reactions of these three products under the basic conditions cleared the relation of them that the isomerization took place between **5** and **6**, and **5** isomerized to **7**, but **7** did not isomerized to **5** or **6** as shown in Fig. 3, and these facts confirmed the structures of them.

Although the best model compound α,α-diphenyl-β-(7-cycloheptatrienyl)ethyl alcohol could not be prepared in pure state, as models of the ether linkage formation of **2**, reactions of 7-cycloheptatrienylmethyl

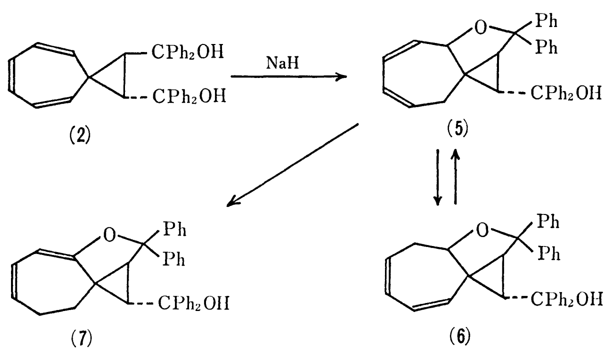

Fig. 3.

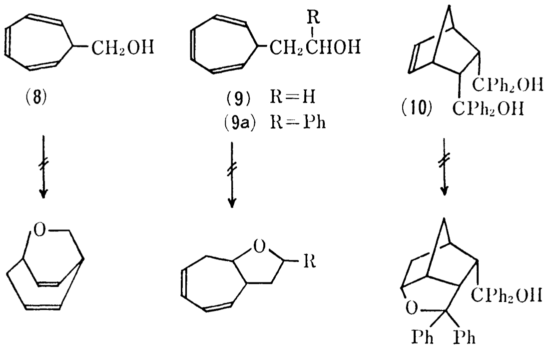

Fig. 4.

Table 3. NMR spectral data (chemical shifts, ppm, and coupling pattern)

	OH	H_2 a	H_2 b	H_3	H_4	H_5	H_6 a	H_6 b	H_7	H_{10}	H_{11}	H_{Ph}
5	1.65 (1H) s	2.52 (1H) d	2.92 (1H) d, d	5.21 (2H) m		5.46 (1H) d, d, d	5.92 (1H) d, d		4.55 (1H) d, d	1.84 (1H) d	2.78 (1H) d	6.9—7.6 (20H) m
6	1.93 (1H) s	5.92 (1H) d, d			5.5 (3H) m		2.60 (1H) d, d, d	2.85 (1H) d, d, d	4.22 (1H) d, d	1.67 (1H) d	2.95 (1H) d	6.8—7.5 (20H) m
7	1.62 (1H) s	1.9 (2H) m		2.25 (2H) m	5.41 (1H) d, d, d	5.87 (1H) d, d	5.63 (1H) d			2.02 (1H) d	2.80 (1H) d	6.9—7.5 (20H) m

s: singlet, d: doublet, d, d: double doublet, d, d, d: triple doublet, m: multiplet.

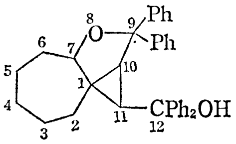

Table 4. Coupling constants (Hz)

	5	**6**	**7**
J_{2-2}	20 (2a—2b)		
J_{2-3}	3.5 (2b—3)	11	
J_{2-4}		3	4 (2a—4) 6 (2b—4)
J_{2-7}	0.5 (2b—7)		
J_{4-5}	4		12
J_{5-6}	11	3 (5—6a) 4.5 (5—6b)	
J_{5-7}	2		
J_{6-6}		18 (6a—6b)	
J_{6-7}	2	9 (6a—7) 5 (6b—7)	
J_{10-11}	4	4.5	4.5

alcohol (**8**),[13)] *β*-(7-cycloheptatrienyl)ethyl alcohol (**9**),[14)] *α*-phenyl-*β*-(7-cycloheptatrienyl)ethyl alcohol (**9a**), and *cis-endo*-1,2-bis(hydroxydiphenylmethyl)-bicyclo[2.2.1]hept-5-ene (**10**)[11)] with sodium hydride under the same conditions as the case of **2** were investigated, but resulted in the complete recovery of the starting alcohols. Reactions of cycloheptatriene with benzyl alcohol or triphenylmethyl alcohol under the same conditions also resulted in the complete recovery of the starting substances.

Discussion

Mechanism of the rearrangement of **2** to **3** by acid treatments can be considered as shown in Fig. 5 by following reasons. First of all, ring creavage reactions of cyclopropane derivatives by acidic reagents are well known, and furthermore, a cycloheptatrienium ion (**11**) is a very stable ion. Secondary, dehydration reaction of **2** should proceed readily, because **2** is a benzyl alcohol. Finally, benzophenone is a stable compound: thus, elimination of benzophenone from **11** should not need high energy. From these reasons, the rearrangement of **2** to **3** seems to proceed readily.

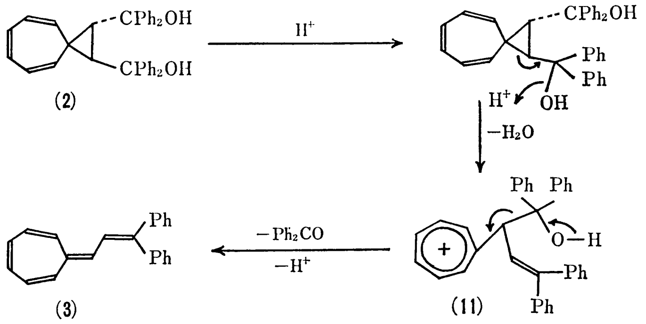

Fig. 5.

There are several plausible explanations about reaction mechanisms of the formation of **5** from **2**. The first one is direct alkoxide anion attack on double bonds. However, the reactions of **8**, **9**, and **10** did not give any ether compounds as descrived before. Steric requirements of **8**, **9**, and **10** are good enough for an ether linkage formation, because sterically analogous compounds such as *α*-(diazomethyl)-*α*′-(7-cycloheptatrienyl)methyl ketone gave a corresponding bond formation product by its decomposition reaction.[15)] Also, it is well known that *endo*-bicyclo[2.2.1]hept-5-en-2-carboxylic acid afforded the lactone derivative by acid treatments. These results indicate that direct alkoxide anion attack on double bonds of **2** is not reasonable.

The second is an explanation by radical mechanisms. Color of the reaction mixture of **2** become deep blue during the reactions suggesting a formation of some radical species. ESR spectrum measurement of this reaction mixture did not show, however, any evidence of a radical formation. Also, if alkoxide radical or cycloheptatrienyl radical formed under the reaction conditions, the corresponding radicals would form in the reactions of **8**, **9**, and **10**, and some reactions should take place in these cases. So that radical mechanisms can not be negrected completely, but not very likely.

The third and the most probable one is a mechanism *via* norcaradiene intermediate. Valence tautomerism between cycloheptatriene and norcaradiene systems is well known phenomenon.[16,17)] This tautomerism is also possible between **2** and its valence isomer (**2a**) which should be very strained one. Therefore, if once this strained tricyclic system formed in the reaction mixture, the intramolecular alkoxide anion attack takes place readily to give the ether compound **5**. The Dreiding model of **2a** indicates that the ether linkage formation is possible between alkoxide oxygen and C_1 (*ac.* 2.4 Å). Also, the most favorable conformer of **2a** is that the alkoxide oxygen comes nearby norcaradiene and the big phenyl groups stick to the outside of norcaradiene.

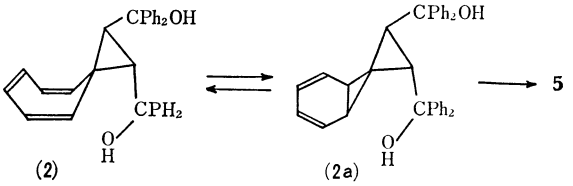

Fig. 6.

Recently, some examples of addition reactions of anionoid reagents with unsaturated or strained compounds have been reported.[18,19)] Especially, addition reaction of amines to strained double bonds such as 9,9′-difluolenyridene[20)] is a good example of this type of reaction.

The interconversions of **5**, **6**, and **7** are thermal 1,5-hydrogen shift reactions.[21)] The diene system of **7** is conjugated with ether linkage constructing the vinyl ether skeletone, and therefore, **7** seems to be the most stable isomer of all of them.

Experimental

All melting and boiling points were uncorrected. IR spectra were measured in carbon tetrachloride solutions or potassium bromide disks, and UV spectra in methanol solutions. PMR spectra were measured with Varian T-60 or HA-100 spectrometer with deuteriochloroform or carbon tetrachloride as solvent and tetramethylsilane as internal standard. Mass spectra were measured with Hitachi RMU 6D spectrometer at 70 eV by direct method.

*Reactions of **1** with Grignard Reagents and Lithium Aluminium Hydride.* (a) Reaction of **1** (468 mg, 2 mmol) with benzylmagnesium chloride (16 mmol) in anhydrous ether (30 ml) with ordinal method gave an yellow oil (1.436 g). Distillation of the oil afforded benzyl alcohol (50—60 °C/0.5 Torr, 300 mg), diphenylethane (65—70 °C/0.5 Torr, 85 mg), and an yellow oily residue (950 mg). Silica gel TLC (hexane, benzene, and methanol; 6 : 3 : 1) gave colorless oily mixtures (381 mg) which showed a broad absorption band at 1775 cm^{-1} in its IR spectrum. (b) Reaction of **1** (234 mg, 1 mmol) with methylmagnesium iodide (8 mmol) also gave none of clear products. (c) Reaction of **1** (300 mg, 1.3 mmol) with lithium aluminium hydride (1.2 g, 32 mmol) in boiling anhydrous tetrahydrofuran (30 ml) with ordinary method gave unidentified oily mixtures (162 mg) which still showed a broad carbonyl band at 1730 cm^{-1} in its IR spectrum.

*Reaction of **1** with Phenyllithium.* A solution of **1** (936 mg, 4 mmol) in anhydrous ether (30 ml) was added to a solution of phenyllithium (30 mmol) in anhydrous ether (20 ml) at 0 °C during 30 min with stirring. After further stirring for 13 h at room temperature, the reaction mixture was decomposed with water, and extracted with ether. Evaporation of the solvent and recrystallization of the residue from acetone gave **2** (1.58 g, 84%), mp 223—225 °C. Found: C, 87.16; H, 6.30%. Calcd for $C_{35}H_{30}O_2$: C, 87.06; H, 6.05%.

*Reactions of **2** with Acids.* A solution of **2** (1.0 g, 2.1 mmol) in 0.1 N sulfuric acid-dioxane (50 ml) was stirred for 1 h at 0 °C, and then added water (100 ml). The reaction mixture was extracted with benzene. A brown tarry material (1.0 g) obtained by evaporation of the solvent was chromatographed on alumina to give benzophenone (377 mg, 98%) by petroleum ether-benzene (9 : 1) and intractable unidentified tarry materials (600 mg). Treatments of **2** with hydrochloric acid, acetic acid, or silica gel gave the almost identical results with that of the case of sulfuric acid.

*Hydrogenation of **3**.* A mixture of **2** (1.0 g, 2.1 m mol), silica gel (10 g, 60—100 mesh), and benzene (70 ml) was stirred at room temperature for 1.5 h. Separation of the silica gel, and following hydrogenation of the filtrate with paradium carbon (10%, 400 mg) gave a pale yellow oil (1.0 g) after absorption of hydrogen (250 ml, 11 mmol). Alumina column chromatography of this oil gave a colorless oil (540 mg) by petroleum ether, benzophenone (278 mg, 74.5%) by petroleum ether-benzene (8 : 2), and diphenylmethyl alcohol (73 mg, 20%) by ether. Distillation of the colorless oil (bath temp 110 °C/0.2 Torr) gave **4** (454 mg, 74.7%). Found: C, 90.16; H, 9.63%. Calcd for $C_{22}H_{28}$: C, 90.35; H, 9.65%.

*Reactions of **2** with Sodium Hydride.* (a) A mixture of **2** (200 mg, 0.4 mmol), 55% sodium hydride (400 mg, 9 mmol), and anhydrous dioxane (10 ml) was stirred at room temperature for 330 h. The reaction mixture was filtered and the residual solid was decomposed with water and extracted with benzene. Combined organic layer was washed with water and dried over sodium sulfate. Evaporation of the solvent afforded an yellow oily material (335 mg). Purification of this oil with silica gel TLC (benzene) gave recovery of **2** (25 mg, 12%, R_f=0.5) and an yellow tarry material (156 mg, R_f=0.75). Crystallization of the tar from petroleum ether-benzene, and further recrystallization from ethyl acetate yielded pure **5** (125 mg, 63%), mp 115 °C. Found: C, 86.74; H, 6.54%. Calcd for $C_{35}H_{30}O_2$: C, 87.10; H, 6.27%. (b) A mixture of **2** (800 mg, 1.6 mmol), 55% sodium hydride (1.6 g, 37 mmol), and anhydrous dioxane (50 ml) was refluxed for 6.5 h. The same treatment as above afforded **5** (34 mg, 4.2%, R_f=0.75), colorless crystals of **6** (mp 150—155 °C, 351 mg, 44%, R_f=0.85), and **7** (mp 170—174 °C, 136 mg, 17%, R_f=0.9). Recrystallization of **6** and **7** from ethyl acetate raised their mps to 164 and 180 °C, respectively. Found: C, 87.10; H, 6.33% for **6**. Found: C, 86.76; H, 6.43% for **7**. (c) A mixture of **2** (1.0 g, 2.1 mmol), 55% sodium hydride (3.0 g, 70 mmol), and anhydrous dioxane (50 ml) was refluxed for 20 h. After the same treatment as above, the obtained yellow oil was chromatographed on alumina. An yellow oily resinous material (460 mg) which was eluted with benzene was recrystallized from ethyl acetate to give pure **7** (430 mg, 43%).

*Acid Treatments of **5**, **6**, and **7**.* A solution of **5** (10 mg, 0.02 mmol) in methanol (5 ml) containing a drop of conc sulfuric acid was allowed to stand at room temperature for 6 h. UV spectrum of this solution was identical with that of **3**, and gas chromatographic study showed existence of benzophonone. Treatments of **6** and **7** under the same conditions as above showed the identical results as the case of **5**.

*Interconversion of **5**, **6**, and **7**.* A solution of **5** (15 mg, 0.03 mmol) and 55% sodium hydride (30 mg, 7 mmol) in anhydrous dioxane (2 ml) was refluxed for 9 h. Silica gel TLC (benzene) showed three spots at the parts coincident with **5** (R_f=0.75), **6** (R_f=0.85), and **7** (R_f=0.9). By the same procedure, reaction of **6** showed the same three spots as the case of **5** on silica gel TLC, but reaction of **7** showed no other spot except one due to itself, and this spot diminished by prolonged refluxing (about one day).

*Reactions of **8**, **9**, and **10** with Sodium Hydride.* (a) A solution of **8** (1.34 g, 11 mmol) and 55% sodium hydride (2.4 g, 56 mmol) in anhydrous dioxane (10 ml) was stirred for 230 h at room temperature. The starting alcohol **8** (1.32 g, 98%) was recovered by the same treatment as the case of the reaction of **2** with sodium hydride. (b) Reaction of **9** (1.41 g) with sodium hydride under the same conditions as above resulted in recovery of the starting alcohol (1.34 g, 95%). (c) Reaction of **10** (500 mg) with sodium hydride under the same conditions as above resulted in recovery of the starting alcohol (485 mg, 97%).

Reactions of Cycloheptatriene with Benzyl Alcohol and Triphenylmethyl Alcohol. (a) A mixture of cycloheptatriene (1.8 g, 20 mmol), benzyl alcohol (16 g, 148 mmol), 55% sodium hydride (3.0 g, 70 m mol), and anhydrous dioxane (10 ml) was refluxed for 17 h. The starting alcohol was recovered quantitatively by the same treatments as above. (b) By the same procedure as above, reaction of cycloheptatriene with triphenylmethyl alcohol resulted in almost quantitative recovery of triphenylmethyl alcohol.

Preparation of α-Phenyl-β-(7-cycloheptatrienyl)ethyl Alcohol. To a solution of phenyl magnesium bromide prepared from 3.58 g of bromobenzene in 30 ml of ether, a solution of 2.0 g of (7-cycloheptatrienyl)acetaldehyde[22] in 20 ml of ether was added dropwise with stirring and under nitrogen atomsphere. After 5 h, an ordinal workup gave 1.40 g of the colorless needles of **9a**: mp 41—43 °C from hexane-ether. Found: C, 84.89; H, 7.64%. Calcd for $C_{15}H_{16}O$: C, 84.87; H, 7.60%.

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