[CONTRIBUTION FROM THE DEPARTMENT OF CHEMISTRY OF THE POLYTECHNIC INSTITUTE OF BROOKLYN]

The Synthesis and Proof of Structure of 1-Ethyl-2,3,4,5-tetraphenylcyclopentadiene and the Chemistry of 6-Methyl-1,2,3,4-tetraphenylfulvene[1]

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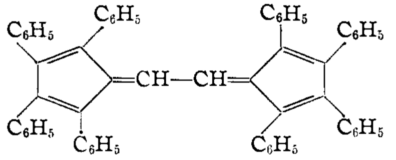
It has been shown that the Wolff–Kishner reduction of tetracyclone 2,4-dinitrophenylhydrazone (I) with ethanolic sodium ethoxide gives 1-ethyl-2,3,4,5-tetraphenylcyclopentadiene (II) rather than tetraphenylcyclopentadiene (III). II is also formed from III under these conditions. The formation of III by the Wolff–Kishner reduction has not been found possible. The proof of structure of II was established by its formation of a Diels–Alder adduct with maleic anhydride, its independent synthesis from 1-hydroxy-1-ethyl-2,3,4,5-tetraphenylcyclopentadiene (VIII), and its conversion to the known 6-methyl-1,2,3,4-tetraphenylfulvene (VI). Successful condensation of VI with *p*-nitrosodimethylaniline and with quinoline-4-carbonal show that the dipole moment of the fulvene is toward the ring and sufficient to activate the methyl group for base-catalyzed condensations.

It has previously been reported that the Wolff–Kishner reduction of tetracyclone dinitrophenylhydrazone (I) with ethanolic sodium ethoxide at 210° for 16 hours yields 1-ethyl-2,3,4,5-tetraphenylcyclopentadiene (II) rather than tetraphenylcyclopentadiene (III).[4] II is also formed when III is treated with ethanolic sodium ethoxide at 210° for 16 hours. Thus it appears that III may be an intermediate in the Wolff–Kishner reduction. However, a search for the conditions under which the Wolff–Kishner reduction might take place and under which III would be stable was unsuccessful.

As a basis for understanding the course of the reaction, a study of the stability of III to alkoxide was undertaken. When III was refluxed with sodium ethoxide in ethanol and then kept at room temperature for three days, only 28% of crude III was recovered. No II was formed and no other pure materials could be isolated from this reaction. When the Wolff–Kishner reduction was attempted under these conditions, the blue color due to the anion of the 2,4-dinitrophenylhydrazone was discharged but it was not possible to isolate any pure materials.

Similarly when the reaction was conducted at 130°, no nitrogen was evolved indicating that the Wolff–Kishner reduction had not taken place. However, the deep blue color had disappeared and neither the starting material nor any other pure material could be isolated.

No III was obtained when I was refluxed with a solution of sodium in ethylene glycol, but an orange product, m.p. 273–281° dec., was isolated. Analysis for carbon and hydrogen and its color suggested the structure bi-(1,2,3,4-tetraphenylfulven-6-yl) (IV).



IV

When III was heated with a solution of potassium *t*-butoxide in *t*-butyl alcohol at 210°, a 12% recovery of crude starting material was obtained. However, the Wolff–Kishner reduction was unsuccessful under these conditions. Thus, it appears that III cannot be prepared by these variations of the Wolff–Kishner reduction.

Degradative proof for the structure of II was obtained by its conversion to the previously described 6-methyl-1,2,3,4-tetraphenylfulvene (VI) and by the formation of a Diels–Alder adduct with maleic anhydride. II was then synthesized by reducing 1-ethyltetraphenylcyclopentadienol (VIII) with hydriodic acid in acetic acid according to the synthesis of 1-neopentyl-2,3-diphenylindene from the corresponding alcohol[5] (see Chart I).

The conversion of II to VI was accomplished by bromination with N-bromosuccinimide in the presence of benzoyl peroxide and ultraviolet light to give the 1-bromo derivative V, which was then dehydrobrominated with pyridine. VI had previously been synthesized by Dilthey,[6] but this synthesis, which involved the treatment of a refluxing solution of VIII in acetic acid with dry hydrogen chloride, could not be duplicated here. A modification in which dry hydrogen bromide was passed into a solution of VIII in dry benzene until the solution was saturated, followed by dehydrobromination of the crude mixture with pyridine, proved successful.

The Diels–Alder reaction of II with maleic anhydride was undertaken to demonstrate that in the synthesis of II from VIII, the hydriodic acid had not reduced the ring. It was found that the reaction took place in boiling bromobenzene (157°), but not in boiling benzene, which is a suitable medium for the addition of III to maleic anhydride. The greater resistance to the formation of the adduct of II is probably due to steric factors. The product, which analyzed correctly, did not melt sharply, and is probably a mixture of isomers. In any case the successful reaction under relatively mild conditions did establish that the diene system in VIII had not been reduced.

Aside from the general discussion of the structure of II, it is interesting to note that the alkylation of III with ethanol and sodium ethoxide formally

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(2) A portion of the Dissertation of S. M. L. presented to the Graduate Faculty of the Polytechnic Institute of Brooklyn in partial fulfillment of the requirements for the Ph.D. degree.

(3) Research Fellow, August, 1951 to June, 1953, supported by the U. S. Army, Squier Signal Laboratory under Contract DA-36-039 sc-15326.

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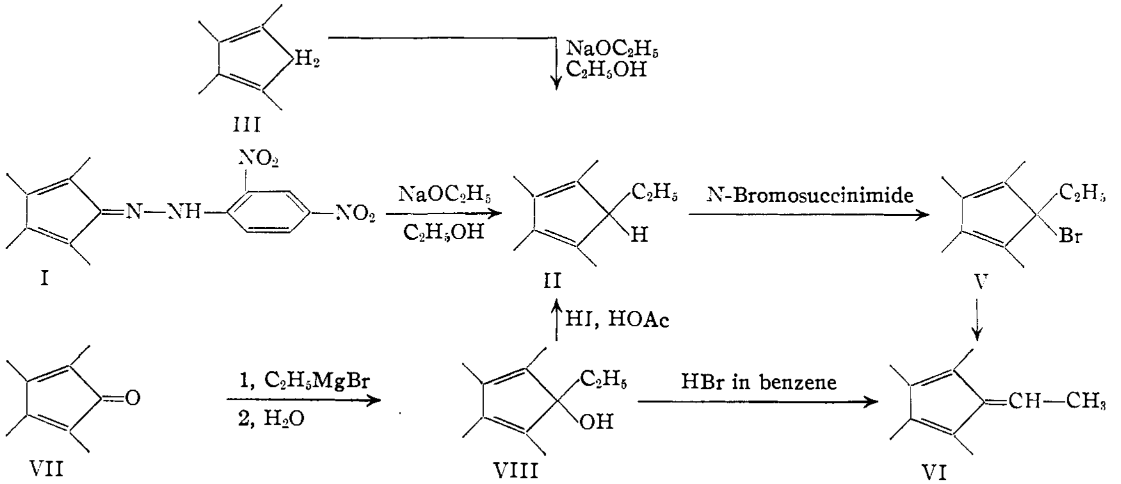
CHART I

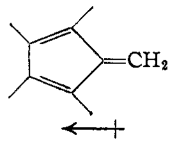
PROOF OF STRUCTURE OF 1-ETHYL-2,3,4,5-TETRAPHENYLCYCLOPENTADIENE

(Each line attached to a central nucleus represents a phenyl group.)



resembles the Guerbet reaction.[7] Although this interesting alkylation of a hydrocarbon under alkaline conditions is being further studied, it can now be reported that a mixture of sodium octoxide and 1-octanol converts III to 1-octyl-2,3,4,5-tetraphenylcyclopentadiene.

Based on theoretical grounds[8-12] and on actual measurements of dipole moments[12,13] a dipole toward the ring has been established for fulvenes. It was, therefore, of significant interest to see whether the 6-methyl group of VI would be an active methyl. When VI was treated with *p*-nitrosodimethylaniline in the presence of sodium methoxide, a brown anil was formed. Acid hydrolysis of the anil in the presence of 2,4-dinitrophenylhydrazine gave X, the 2,4-dinitrophenylhydrazone of 6-carbonal-1,2,3,4-tetraphenylfulvene. Hydrolysis in the absence of 2,4-dinitrophenylhydrazine gave intractable mixtures. Condensation of VI was also successful with quinoline-4-carbonal to give 1-(4'-quinolyl)-2-(1'',2'',3'',4''-tetraphenylfulven-6''-yl)-ethylene (XI).



It is thus established chemically that, in full accord with theory, the dipole moment of a fulvene is toward the ring and further is sufficient to activate a 6-methyl group for base-catalyzed condensation.

Experimental

1-Ethyl-2,3,4,5-tetraphenylcyclopentadiene (II). A.—A solution of 1.0 g. (1.77 mmoles) of sodium in 40 ml. of absolute alcohol was heated in a sealed tube with 1.0 g. of tetracyclone 2,4-dinitrophenylhydrazone[14] at 210° for 16 hours. The yellow solid (310 mg.) was removed by filtration, washed with alcohol, dried and dissolved in benzene. The solution was chromatographed on alumina and the lower blue-white fluorescent band eluted with benzene. The solvent was removed *in vacuo* and the residue was triturated with methanol, filtered, washed with methanol and dried *in vacuo* to give 0.177 g. (0.44 mmole, 25%) of material melting at 178–179° (turbid melt). After two recrystallizations from heptane, a sample melted at 181–181.5°, after drying at 100° *in vacuo*. II can also be recrystallized from cyclohexane, ethyl acetate, or acetonitrile. A mixture with tetraphenylcyclopentadiene[15] melted at 162–172°.

Anal. Calcd. for $C_{31}H_{26}$: C, 93.42; H, 6.58; mol. wt., 398. Found: C, 93.43, 93.39; H, 6.60, 6.60; mol. wt. (ebullioscopic in benzene), 367.

B.—A mixture of 5.0 g. (0.0135 mole) of III and a solution of 1.0 g. of sodium in 40 ml. of absolute alcohol was heated in a sealed tube for 16 hr. at 210°. After filtration and washing with alcohol, water and alcohol again, 4.5 g. (0.0113 mole, 84%) of material melting at 178–180° was obtained. A mixture with III melted from 163 to 175°; no depression in melting point was obtained when mixed with the product obtained in A.

C.—A solution of 2.0 g. (4.83 mmoles of 1-ethyl-1-hydroxy-2,3,4,5-tetraphenylcyclopentadiene) (VIII)[6] in 100 ml. of glacial acetic acid was heated to reflux and 1.3 ml. of 57% hydriodic acid added. The mixture, which became very dark at this point, due to the liberation of iodine, was refluxed for one minute, then cooled, and a suspension of hydrated stannous chloride in concentrated hydrochloric acid was added until the color was discharged. The mixture was diluted with water and extracted with benzene. After washing the benzene layer consecutively with water, sodium bicarbonate solution, sodium bisulfite solution and water again, it was dried over anhydrous magnesium sulfate. Chromatography of the benzene solution on alumina gave a yellow band together with and followed by a purple fluorescing band; a yellow fluorescing and blue-green fluorescing band remained at the top of the column. The yellow band was eluted and rechromatographed but could not be separated into its components. On removal of the solvent, 1.55 g. of yellow solid which melted incompletely from 163 to 180° and fluoresced green-white was obtained. After recrystallization from cyclohexane, 0.50 g. (1.26 mmoles, 26%) of yellow material melting at 175–178.5° was obtained. A mixture with II obtained in B melted at 176–180°. After one recrystallization from ethyl acetate and two recrystallizations from acetonitrile, the blue-white fluorescing material melted at 181–182° after drying at 100°. Found: C, 93.19; H, 6.60.

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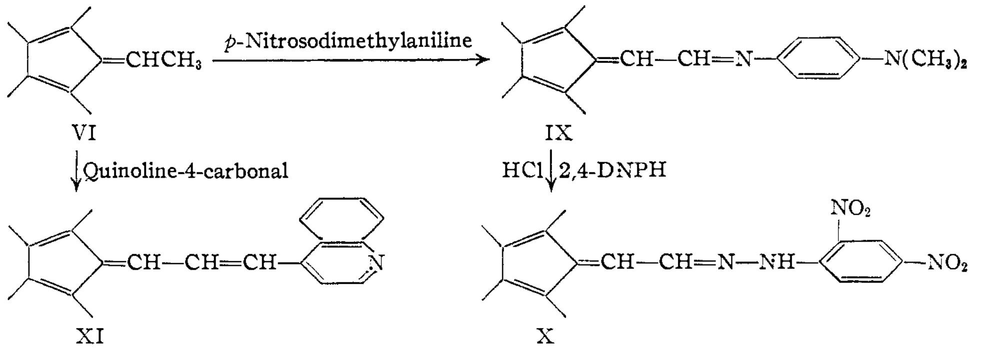
CHART II

THE CHEMISTRY OF 6-METHYL-1,2,3,4-TETRAPHENYLFULVENE

(Each line attached to a central nucleus represents a phenyl group)



Reaction of III with Ethylene Glycol Sodium Salt in Ethylene Glycol.—To a solution of 1.0 g. of sodium in 50 ml. of ethylene glycol was added 1.0 g. of II and the mixture was heated to reflux under nitrogen. A black oil formed on top of the solution. After refluxing for 8 hr., the solution was cooled and the solid which formed was filtered, washed with water, and dried. The dark solid (825 mg.) was treated with 50 ml. of benzene, and the insoluble material filtered, washed with benzene and dried. An orange-red solid (112 mg.), m.p. 267–274° dec., which fluoresced red under ultraviolet light and was insoluble in and did not color concentrated sulfuric acid, was obtained. After recrystallization from xylene yellow crystals which fluoresced yellow and melted at 262–272° dec., were obtained. They are thermochromic and could be converted to the red form by heating to 140°. A sharper melting range is not obtained if the rate of heating is changed or if the compound is inserted in the bath at 250°. After recrystallization from xylene, a sample, after drying at 140° *in vacuo*, melted at 273–281° dec.

Anal. Calcd. for $C_{60}H_{42}$: C, 94.45; H, 5.55. Found: C, 94.33; H, 5.75.

The benzene mother liquor was chromatographed on alumina but no pure material could be obtained from it.

1-Ethyl-1-bromo-2,3,4,5-tetraphenylcyclopentadiene (V).—A mixture of 2.0 g. (5.03 mmoles) of II, 0.96 g. (5.4 mmoles) of N-bromosuccinimide and 0.10 g. of benzoyl peroxide in 100 ml. of dry carbon tetrachloride was heated to refluxing. When it started to reflux, the solution became yellow, the fluorescence disappeared and some succinimide appeared on top of the solution. The reaction was irradiated by direct sunlight for 25 minutes (July 2, 3:30–3:55 p.m.) and was complete after a total reflux time of 50 minutes. A trace of hydrogen bromide was evolved.

After cooling and filtering the mixture, the mother liquor was evaporated *in vacuo* under nitrogen. The residue, an orange oil, crystallized when triturated with Skellysolve A (b.p. 28–38°). After filtering, washing and drying, 1.85 g. (3.88 mmoles, 77%) of yellow crystals melting at 142–147° dec. was obtained. On recrystallization from Skellysolve B (b.p. 60–68°), 1.7 g. (71%) of yellow crystals melting at 148–149.5° dec., were obtained.

After two more recrystallizations from Skellysolve B, a sample melted at 145.5–147° dec., after drying at 65°.

Anal. Calcd. for $C_{31}H_{25}Br$: C, 77.98; H, 5.28; Br, 16.74. Found: C, 78.32; H, 5.61; Br, 16.31.

A different sample prepared from another run melted at 147–149° dec., after drying at 80°.

Anal. Found: C, 78.27; H, 5.41; Br, 16.43.

6-Methyl-1,2,3,4-tetraphenylfulvene (VI). A.—A mixture of 5.0 g. (0.0105 mole) of V and 50 ml. of dry pyridine was heated to refluxing. The compound dissolved, forming a green solution which darkened rapidly. After refluxing for 2.75 hr., the mixture was poured into ice-water and acidified with cold, concentrated hydrochloric acid. The mixture was extracted with benzene and the organic layer washed with water, dried over anhydrous magnesium sulfate and evaporated. The residue was dissolved in dry carbon tetrachloride and chromatographed on alumina. (The benzene solution can be chromatographed directly but this entails the use of a much larger column.) A dark brown (non-fluorescing) band and a colorless (green-fluorescing) band remained at the top of the column while a yellow (non-fluorescing) band descended rapidly and was eluted with carbon tetrachloride. The eluate was concentrated *in vacuo* to give an orange semi-solid which crystallized upon trituration with Skellysolve A (m.p. 184–198°). It gave a negative Beilstein test for halogen. After two recrystallizations from Skellysolve C (90–100°), 2.2 g. (5.56 mmoles, 53%) of VI, melting at 193–198°, was obtained. Two more recrystallizations afforded material which melted constantly at 198–200°, after drying at 100° *in vacuo* (reported[6] m.p. 194–195°).

Anal. Calcd. for $C_{31}H_{24}$: C, 93.90; H, 6.10. Found: C, 93.58; H, 6.39.

The material gives a red color in sulfuric and the solution fluoresces red under ultraviolet light, which is a characteristic of fulvenes. It is somewhat sensitive to atmospheric oxygen above room temperature.

B.—A solution of 12 g. (0.029 mole) of 1-ethyl-1-hydroxy-2,3,4,5-tetraphenylcyclopentadiene (VIII) in 400 ml. of benzene was saturated with dry hydrogen bromide at room temperature. The solution became cloudy, darker in color, and no longer fluoresced. After standing for 10 minutes, the mixture was washed with water to remove the acid, then dried over anhydrous magnesium sulfate and chromatographed on alumina. The impurities were strongly adsorbed and remained on the column. The eluate had a very slight blue fluorescence. The solvent was removed and the residue dissolved in 150 ml. of dry pyridine and the solution refluxed for 1.25 hours. After standing at room temperature for 24 hr., the mixture was worked up as in part A. After chromatography, 5 g. of material melting at 174–190° was obtained. After recrystallization from Skellysolve C, 3.35 g. (8.46 mmoles, 29%) of VI melting at 192–198° was obtained. Two more recrystallizations afforded material melting at 198–200° after drying *in vacuo* at 100°. No depression was observed in a mixture with VI obtained in A.

Anal. Found: C, 93.66; H, 6.28.

Diels–Alder Reaction—7-Ethyl-1,4,5,6-tetraphenylbicyclo[2.2.1]hept-5-ene-2,3-dicarboxylic Anhydride.—A mixture of 0.80 g. (2.0 mmoles) of II, 0.40 g. (4.1 mmoles) of maleic anhydride and 5 ml. of bromobenzene was refluxed for 2.25 hr. On cooling, a very small amount of solid, m.p. 118–133° (incomplete), was obtained. The mother liquor was concentrated *in vacuo*, giving a heavy oil which crystallized slowly on trituration with Skellysolve A, m.p. 88–140°. After washing with water to remove unreacted maleic anhydride, it melted at 88–156°. After four recrystallizations from cyclohexane, a sample which was dried at 100° melted at 215–218°.

Anal. Calcd. for $C_{35}H_{28}O_3$: C, 84.64; H, 5.68. Found: C, 84.50; H, 5.86.

1-*n*-Octyl-2,3,4,5-tetraphenylcyclopentadiene.—To a solution of 0.50 g. of sodium in 20 ml. of *n*-octyl alcohol was added 2.0 g. (5.4 mmoles) of III and then the solution was refluxed for 16 hr. The solution was quite dark at the start, but became lighter as the refluxing proceeded. On cooling, a wax-like precipitate of sodium *n*-octoxide separated. The mixture was partitioned between water and benzene and the organic layer was washed with water until neutral. Concentration of the benzene layer and cooling gave impure product. Filtration, washing with methanol and recrystallization from a mixture of Skellysolves B and C gave 0.100 g. (0.21 mmole, 3.8%) of colorless octyldiene, m.p. 120–123°. For analysis, the compound was recrystallized twice from Skellysolve B, m.p. 123.5–124.5°.

Anal. Calcd. for $C_{37}H_{38}$: C, 92.06; H, 7.94. Found: C, 91.93; H, 7.76.

***p*-Dimethylaminoanil of 6-Carbonal-1,2,3,4-tetraphenylfulvene (IX).**—A solution of 1.7 g. (4.3 mmoles) of VI and 1.2 g. (8.0 mmoles) of *p*-nitrosodimethylaniline in 25 ml. of dry benzene was heated to reflux and treated with 1 ml. of a 5% solution of sodium methoxide in methanol. The mixture darkened, became blood-red, and a dark solid separated on the sides of the flask. After refluxing for 15 min., the mixture was cooled and filtered. Washing the solid with methanol and drying gave 1.5 g. (2.8 mmoles, 66%) of brown crystals of IX, m.p. 215–218° dec. The product was contaminated with a decomposition product of *p*-nitrosodimethylaniline, which was brown, melted at 232–248°, and was soluble in 5% hydrochloric acid from which solution it was precipitated as a yellow solid by alkali. It could be separated from the anil by virtue of its solubility in a mixture of hot cyclohexane–benzene (from which it crystallized on cooling) in which the anil was insoluble. It was not separated from the anil by chromatography on alumina.

After washing with cold 5% hydrochloric acid until the washings were colorless, then with water, and drying, it melted 227–230.5° dec. A mixture with the dimethylaminoanil of tetracyclone melted at 215–218°. For analysis IX was recrystallized three times from benzene, m.p. 233–234° dec.

Anal. Calcd. for $C_{39}H_{32}N_2$: C, 88.60; H, 6.10; N, 5.30. Found: C, 89.06; H, 5.72; N, 5.29.

6-Carbonal-1,2,3,4-tetraphenylfulvene 2,4-Dinitrophenylhydrazone (X).—A mixture of 0.3 g. (0.57 mmole) of IX, 0.12 g. of 2,4-dinitrophenylhydrazine, 10 ml. of 1:1 hydrochloric acid and 10 ml. of benzene was refluxed under nitrogen for 15 min. The mixture was cooled, separated and the organic layer was washed with water, dried over anhydrous magnesium sulfate and chromatographed on alumina. A dark-gray band was eluted with benzene and appeared orange-brown in solution. A red-violet and a yellow band remained at the top of the column.

Distillation of the solution to dryness and trituration of the residue with Skellysolve A gave 0.115 g. (35%) of crude X which melted at 234–253° dec. Four recrystallizations from a mixture of dioxane (soluble)–95% ethanol (insoluble) gave a brown solid, m.p. 265–267° dec.

Anal. Calcd. for $C_{37}H_{26}N_4O_4$: C, 75.24; H, 4.44; N, 9.49. Found: C, 75.13; H, 4.70; N, 9.52.

1-(4′-Quinolyl)-2-(1″,2″,3″,4″-tetraphenylfulven-6″-yl)-ethylene (XI).—To a refluxing mixture of 0.4 g. (1.01 mmoles) of VI and 0.2 g. of quinoline-4-carbonal in a solvent mixture of 25 ml. of methanol and 15 ml. of benzene was added 5 ml. of 5% methanolic sodium methoxide. The solution became dark and on refluxing for 15 min. all of the VI was in solution. After refluxing for a total of 2.25 hr., cooling gave no crystals. The solution was then poured into water, separated and the aqueous layer was washed with benzene and then discarded. The combined benzene extracts were washed with water, dried over anhydrous magnesium sulfate, filtered and distilled to dryness. Trituration with Skellysolve A crystallized the residual oil. Purification was achieved by chromatography on alumina using cyclohexane as solvent and a mixture of cyclohexane and benzene as eluant. The lowest invisible band, which fluoresced blue-white under ultraviolet light (unreacted VI (?) was followed by a visible yellow band, and then by the main brown fulvene band, which was eluted with benzene.

Distillation of the solvent gave a semi-solid which melted at 216–217° after trituration with Skellysolve A, 0.175 g. (0.33 mmole, 32%). Two recrystallizations from acetonitrile raised the melting point to 217–218°.

Anal. Calcd. for $C_{41}H_{29}N$: C, 91.93; H, 5.46; N, 2.62. Found: C, 92.14; H, 5.64; N, 2.54.

BROOKLYN 1, NEW YORK

[CONTRIBUTION FROM THE CHEMISTRY DEPARTMENT OF WAYNE UNIVERSITY]

Epoxyethers. IV. Mechanism of the Opening of an Epoxyether with an Organic Acid[1]

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The reaction of an epoxyether I with 3,5-dinitrobenzoic acid is considered to proceed by initial attack of the acid on C_1 of the epoxyether followed by intramolecular ester interchange with the irreversible formation of alcohol and the ketoester III.

The purpose of this investigation was the determination of the mechanism of the reaction of organic acids with epoxyethers. The results indicated that in the example studied the organic acid opened the oxide ring of the epoxyether I by initial attack at the carbon atom (C_1) holding the two oxygen functions to give the ester of a hemiketal (II). This ester when heated or dissolved in an organic solvent, was converted to the ketoester IIIa.

A number of epoxyethers have been isolated recently in this Laboratory and the chemistry of these compounds has been investigated.[3–5] The epoxyethers (*cf.* I) have been characterized by hydrolysis reactions which gave α-hydroxyketones (*cf.* IV), methanolysis reactions which gave α-hydroxyketals (*cf.* V) and reactions with organic acids which gave ketoesters (*cf.* IIIa).

In the reaction with organic acids the reagent appeared in the product attached to carbon atom 2. The fact that in acid-catalyzed alcoholysis the reagent appeared in the product on carbon atom 1 indicated that the mechanism of the reaction with organic acids might involve an intermediate in which the reagent was also attached to carbon atom 1.

The epoxyether, 1,2-epoxy-2-methyl-1-methoxy-1-(*p*-biphenyl)-propane (I), chosen for this investigation was a solid and gave derivatives which were readily crystallizable. This epoxyether was prepared in 84% yield from the bromoketone by treatment with alcoholic sodium methoxide. Since *p*-phenylisobutyrophenone was prepared in 92% yield and brominated in 85% yield, I represented a readily available starting material.

The epoxyether was stable at high temperatures (250°) for a short period of time and was characterized by hydrolysis to the α-hydroxyketone IV in 79% yield, methanolysis to the α-hydroxyketal V in 83% yield, and reaction with benzoic acid to

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[2] Abstracted in part from the Ph.D. thesis of Mr. Stanley J. Dykstra, Wayne University, June, 1953.

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[5] C. L. Stevens, M. W. Weiner and R. C. Freeman, *ibid.*, **75**, 3977 (1953).