removed by filtration. After allowing the solution to stand for 3 days at 37°, the precipitate was filtered and then dissolved in 300 ml of water. This solution was passed through an Amberlite IR-120 (H^+) column (3 × 70 cm), and the effluent was treated with Dowex 1X8 (HCO_3^-), 200–400 mesh. The resin was filtered off and the neutral filtrate was concentrated *in vacuo* to a syrup and methanol was added to give 2.48 g of crystals, yield 17%, $[\alpha]^{20}_{D}$ −87.6°. Under the same conditions and by the same treatment, fructosazine was obtained in the yield of only 5.5% from 2-amino-2-deoxy-D-glucose hydrochloride. This fructosazine, after two recrystallizations from water and methanol, had mp 235° dec, $[\alpha]^{20}_{D}$ −83.4° (*c* 1.0, water).

Anal. Calcd for $C_{12}H_{20}N_2O_8$: C, 45.00; H, 6.29; N, 8.75. Found: C, 45.29; H, 6.54; N, 8.77.

Fructosazine prepared from 2-amino-2-deoxy-D-mannose, showed an infrared spectrum identical with that of fructosazine from 2-amino-2-deoxy-D-glucose and from 1-amino-1-deoxy-D-fructose.

From the fructosazine prepared from 2-amino-2-deoxy-D-mannose, the following compounds were prepared.

2,5-Bis(D-*arabino*-tetraacetoxybutyl)pyrazine had mp 173°, $[\alpha]^{11}_{D}$ −7.1° (*c* 1.0, chloroform). Pyrazine-2,5-dicarboxylic acid had mp 251° dec. 2,5-Dimethoxycarbonylpyrazine showed mp 169°.

Analyses, infrared spectra, and mixture melting points showed that they were identical with the compounds prepared from 2-amino-2-deoxy-D-glucose.

2,5-Dimethoxycarbonylpyrazine *via* 2-(D-*arabino*-tetrahydroxybutyl)quinoxaline.—1-Deoxy-1-*p*-toluidino-D-fructose was prepared[15] from D-glucose and *p*-toluidine, and, from this material, 2-(D-*arabino*-tetrahydroxybutyl)quinoxaline was prepared.[7] Pyrazine-2,3,5-tricarboxylic acid was prepared[6] from 2-(D-*arabino* tetrahydroxybutyl)quinoxaline. 2,5-Dimethoxycarbonylpyrazine was prepared[6] by decarboxylation and esterification with methanolic hydrogen chloride from pyrazine-2,3,5-tricarboxylic acid, mp 169°, over-all yield 210 mg (0.8%).

Anal. Calcd for $C_8H_8N_2O_4$: C, 48.98; H, 4.11; N, 14.28. Found: C, 48.69; H, 4.35; N, 14.27.

No depression in the melting point was observed when this material was mixed with a sample of 2,5-dimethoxycarbonylpyrazine derived from fructosazines prepared from both 2-amino-2-deoxy-D-glucose and 2-amino-2-deoxy-D-mannose, and they showed identical infrared spectra.

Paper Chromatographic Identification[11] of 2-Amino-2-deoxy-D-mannose in the Reaction Mixture from 2-Amino-2-deoxy-D-glucose.—In the experiment for the formation of fructosazine from 2-amino-2-deoxy-D-glucose in methanol at 70°, the methanolic mother liquor, after removing the precipitated fructosazine, was concentrated *in vacuo* at 0°, and the residue was diluted with water. This solution was passed through a column of Dowex 50X8 (H^+), 200–400 mesh. This column was eluted with 0.22 *N* hydrochloric acid, and the Elson–Morgan reaction-positive fraction was collected and concentrated *in vacuo*, and methanol was added to give crystals. These crystals were chromatographed on Toyo Roshi No. 50 filter paper previously soaked in 0.1 *M* barium chloride, in butanol–pyridine–water (6:4:3, v/v). Besides a spot corresponding to 2-amino-2-deoxy-D-glucose hydrochloride (R_f 0.31), another spot (R_f 0.37) which had the same mobility as 2-amino-2-deoxy-D-mannose hydrochloride (R_f 0.37) was detected.

---

[15] F. Weygand, *Ber.*, **73**, 1259 (1940).

Electrolytic Oxidation of Cyclobutane-1,3-dicarboxylic Acids. An Electrochemical Synthesis of 2,4-Dicarbomethoxybicyclobutane[1]

Anthony F. Vellturo[2a] and Gary W. Griffin[2b]

Departments of Chemistry of Tulane University and Louisiana State University in New Orleans, New Orleans, Louisiana

Received January 21, 1966

Anodic oxidation of *trans,trans,trans*-1,3-dicarboxy-2,4-dicarbomethoxycyclobutane in the Kolbe manner gives 2,4-dicarbomethoxybicyclobutane. In contrast, electrolysis of α-truxillic acid under similar conditions results in ring contraction and formation of the lactone of *cis,cis*-1-carboxy-2-α-hydroxybenzyl-3-phenylcyclopropane as the major product. A cationic mechanism is invoked to explain the difference in behavior exhibited by these cyclobutane-1,3-dicarboxylic acids.

As part of a continuing program directed toward the synthesis of tetrahedrane (I) the feasibility of employing the Kolbe electrochemical synthesis[3] for the formation of highly strained small-ring systems including bicyclobutanes has been investigated. Electrolytic studies were conducted on α-truxillic acid (II) (Scheme I) and *trans,trans,trans*-1,3-dicarboxy-2,4-dicarbomethoxycyclobutane (III) in order to determine the potential value of electrode reactions for 1,3-bond formation in cyclobutyl systems. *trans,trans,trans*-1,2,3,4-Tetracarboxycyclobutane (IV) (the obvious direct Kolbe precursor for I) was excluded as a substrate in these preliminary studies since olefin formation and rearrangement may be anticipated as possible complicating factors when vicinal carboxyl groups are present.[4]

Electrolysis of α-truxillic acid (II) in methanol (2 × 3 cm platinum electrodes; 80 v, 0.8 amp) until the solution was basic afforded at least eight products as established by thin layer chromatography. Of these, only the major product VII, mp 131–133°, was isolated by column chromatography in quantities sufficient for characterization. The hydrocarbon fraction constituted <1% of the material eluted and, therefore, it must be concluded that, under the conditions employed, anodic bisdecarboxylation with concomitant cyclization, does not occur in this case. The γ-lactone V, if formed, was not isolated either.

The structure of VII, obtained from II by electrolysis, was established by direct comparison with an authentic sample of this lactone synthesized independently according to the method of Stoermer and Schenck by deamination of γ-truxillamic acid (VI).[5] The isomeric lactone VIII (mp 120–121°) may be prepared from β-truxillamic acid (IX) by the same procedure. Paudler, Herbener, and Zeiler[6a] recently published their data on the electrochemistry of cyclobutane-1,2-dicarboxylic acids including β-truxinic acid (X). It is noteworthy

---

[1] For a preliminary report of this work, see A. F. Vellturo and G. W. Griffin, *J. Am. Chem. Soc.*, **87**, 3021 (1965).

[2] (a) Public Health Postdoctoral Fellow (Fellowship No. 1-F2-GM-11,801-01) from the General Medical Sciences Division. (b) To whom requests for reprints should be directed at Louisiana State University in New Orleans, New Orleans, La.

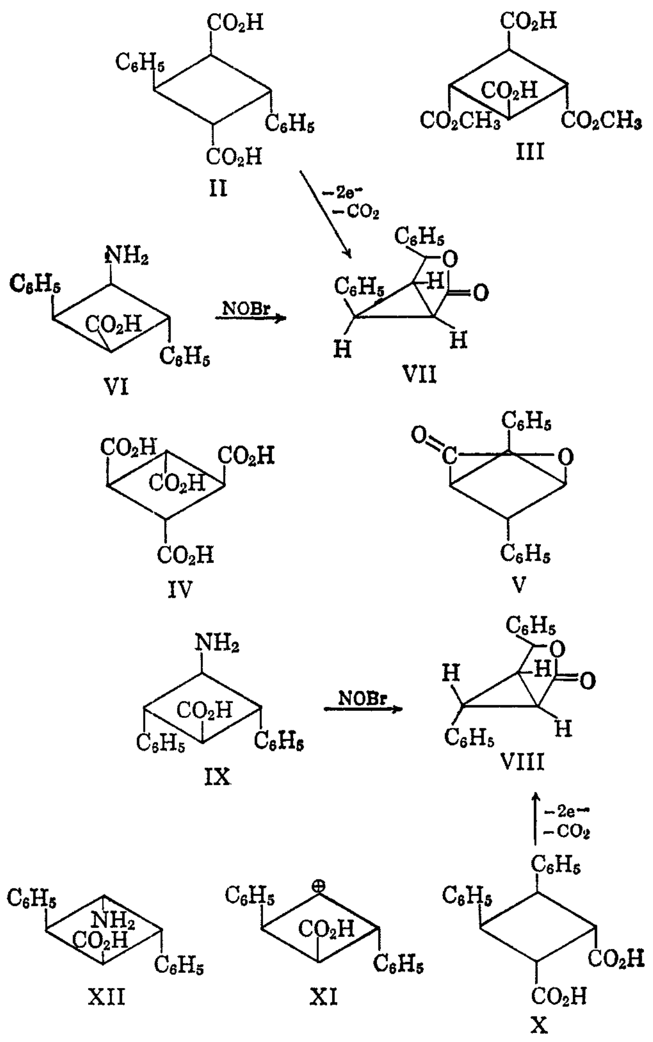
[3] For recent reviews on the Kolbe reaction, see (a) B. C. L. Weedon, *Advan. Org. Chem.*, **1**, 1 (1960); (b) G. W. Thiessen, *Record Chem. Progr.* (Kresge-Hooker Sci. Lib.), **21**, 243 (1960).

[4] E. J. Corey and J. Casanova, Jr., *J. Am. Chem. Soc.*, **85**, 165 (1963).

[5] R. Stoermer and F. Schenck, *Ber.*, **60**, 2566 (1927).

[6] (a) W. W. Paudler, R. E. Herbener, and A. G. Zeiler, *Chem. Ind.* (London), **No. 46**, 1909 (1965); (b) H. I. Bernstein and E. S. Wallis, *J. Org. Chem.*, **7**, 261 (1942).

SCHEME I



that *anti* lactone VIII is one of seven products obtained by these workers upon anodic oxidation of X.

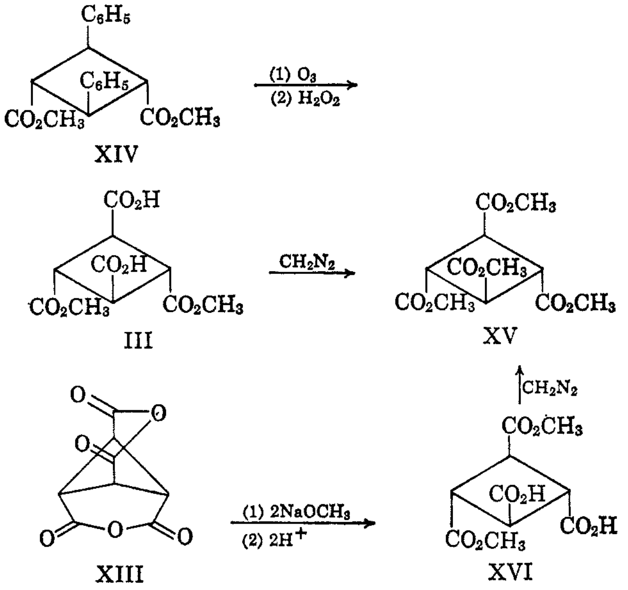
Stoermer and Schenck[5] showed, and it was later verified by Bernstein and Wallis,[6b] that in the conversion of VI to the *syn* lactone VII with nitrosyl bromide the "free" carbonium ion XI cannot be an intermediate. It was concluded in these early studies that loss of nitrogen and ring contraction must occur in a concerted manner since the *syn* lactone VII is not formed by deamination of XII, an alternate precursor for the cation XI.[5] In the electrolysis of dibasic acids it is possible that a carbonium ion, the corresponding carboxylate dipolar ion, or a behaviorly related species perhaps adsorbed on the anode may intervene through an additional one-electron oxidation step following loss of carbon dioxide in an extension of the conventional Kolbe sequence. Cationic species have been proposed

$$RCOO^- \longrightarrow RCOO\cdot \xrightarrow{-CO_2} R\cdot \longrightarrow R^+$$

as intermediates in other anodic oxidation studies.[3,4] Of singular interest in this context is the report that *dl*- and *meso*-diphenylsuccinic acids both yield *trans*-stilbene upon electrolysis. That the structure of the product is independent of substrate geometry is convincing evidence that a stepwise rather than concerted mechanism must be operative in the bisdecarboxylation of these diphenylsuccinic acid diastereomers.[4] In light of this observation it is reasonable to assume that electrolysis of α-truxillic acid may also proceed with formation of a carbonium ion (XI) which can subsequently undergo ring contraction to afford a benzyl cation. Lactonization in a final step would give VII. It appears eminently reasonable that a carbonium ion mechanism also is operative in the conversion of X to VIII.[6a]

The diacid–diester III was considered a more promising bicyclobutane precursor than II. Certainly the driving force for ring contraction of an electron-deficient anodic intermediate generated from III should not be great since the stabilizing effect associated with development of the benzyl carbonium ion proposed in the rearrangement of II is absent. All attempts to prepare III from the readily available "cage" dianhydride XIII[7] failed. Addition of 1 equiv of methanol to XIII in the hope of obtaining a precursor for III could not be achieved. This failure may be attributed to the insolubility of XIII in all aprotic solvents tried. After initial attack by methanol occurs the resulting ester, a product with appreciable solubility, is the species vulnerable to subsequent attack. The synthesis of III was finally accomplished by destructive ozonolysis[8] of dimethyl-ε-truxillate (XIV) (Scheme II) which gave the diacid–ester III (mp 181–183°) in 52% yield. Confirmation of the structure of III was achieved by its conversion to the known *trans,trans,trans*-1,2,3,4-tetracarbomethoxycyclobutane[9] (XV) upon treatment with diazomethane. It was established that III was not identical with *trans,trans,trans*-1,2-dicarboxy-3,4-dicarbomethoxycyclobutane[7] (XVI, mp 167–170°) by comparison of infrared spectra and mixture melting point determination. It is significant that XVI is the only other possible diester of *trans,trans,trans*-1,2,3,4-tetracarboxycyclobutane and was prepared by treating XIII with two equivalents of methanol.

SCHEME II



When *trans,trans,trans*-1,3-dicarboxy-2,4-dicarbomethoxycyclobutane (III) (Scheme III) was electrolyzed between smooth platinum electrodes and the resulting

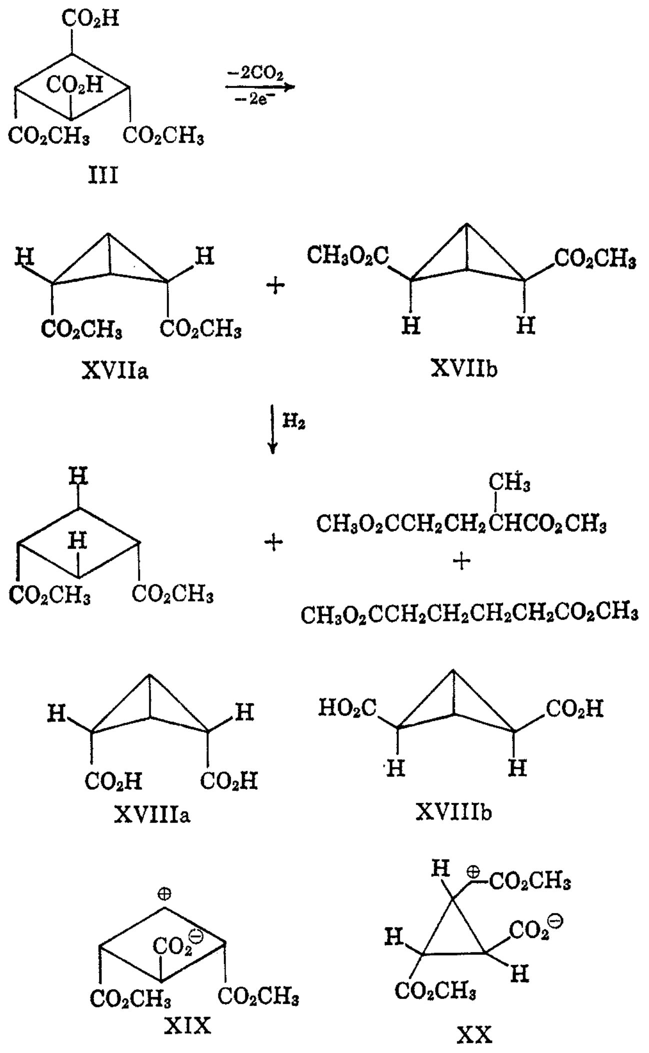
---

[7] G. W. Griffin and R. B. Hager, *Rev. Chim., Acad. Rep. Populaire Roumaine*, **7**, 901 (1962).

[8] G. W. Griffin and D. F. Veber, *J. Am. Chem. Soc.*, **82**, 6417 (1960).

[9] G. W. Griffin, J. E. Basinski, and A. F. Vellturo, *Tetrahedron Letters*, No. **3**, 13 (1960).

SCHEME III



mixture analyzed by gas–liquid partition chromatography, one major volatile product was detected. A minor product with a longer retention time also was formed and the yield of this substance fluctuated from one experiment to another. The major component was a white crystalline solid, $C_8H_{10}O_4$, mp 83–85°. The nmr spectrum, which exhibited three signals (all singlets) at τ 6.32 (three protons), τ 7.35 (one proton), and τ 8.42 (one proton) and no absorption due to vinyl protons, is consistent with the bicyclobutane structure XVII. Further evidence for this assignment of structure was obtained by catalytic hydrogenation of XVII (Adams catalyst) to *cis*-1,3-dicarbomethoxycyclobutane,[10] dimethyl adipate, and dimethyl α-methylglutarate. These reduction products were formed in the ratio 15:1:21, respectively. The conversion of XVII to *cis*-1,3-dicarbomethoxycyclobutane may occur in a direct manner, however an alternate mechanism involving prior isomerization to 1,3-dicarbomethoxycyclobutene and subsequent hydrogenation of the double bond from the least hindered side cannot be excluded at this time. No dimethyl β-methylglutarate appears to have been formed. These results are consistent with previous hydrogenation data reported for other bicyclobutyl systems.[11a–g] It should be noted that two stereo modifications, XVIIa and XVIIb, of 2,4-dicarbomethoxybicyclobutane may exist. To date only one product has been isolated and characterized as a bicyclobutane.[11] An attempt to convert diester XVII to the corresponding dibasic acid XVIII by basic hydrolysis and subsequent neutralization was unrewarding, a result not unexpected in view of the known acid sensitivity of bicyclobutyl systems. Since the free dibasic acid, a potential precursor for tetrahedrane (I), could not be obtained, an alternate route to I circumventing the isolation of XVIII was attempted. After the initial electrolysis of III to XVII attempts were made to generate the disodium salt of XVIII through basic hydrolysis of XVII by direct addition of sodium hydroxide to the electrolytic cell. The electrolysis was then continued; however, the only volatile materials isolable were solvent oxidation products.

It is inviting to propose a diradical mechanism for the conversion of III to XVII in which the dibasic acid is held in the vicinity of the anode during oxidative bisdecarboxylation. However, the data on the diphenylsuccinic acids previously described suggest that a stepwise process consisting of oxidation and monodecarboxylation to the zwitterion XIX may occur followed by loss of carbon dioxide and collapse to XVII. Although the proposed ionic intermediate XIX is formally analogous to XI ring contraction undoubtedly does not occur because of the destabilizing effect associated with the development of a positive charge adjacent to a carbomethoxyl group in the transition state leading to XX. Attempts to extend what is formally an intramolecular Kolbe reaction to other systems are in progress.

Experimental Section

Melting points were determined on a Thomas–Hoover apparatus and are uncorrected. Infrared spectra were recorded with a Beckman IR-5 or IR-8 spectrophotometer; nmr spectra were obtained with the Varian A-60 nmr spectrometer, using tetramethylsilane (TMS) as an internal standard. Carbon and hyrogen analyses were performed by Galbraith Microanalytical Laboratories, Knoxville, Tenn.

α-Truxillic Acid (II).—A 100 g sample of α-*trans*-cinnamic acid (recrystallized from 50% ethanol-water and allowed to stand in the mother liquor overnight at 50°) was placed in a stoppered pyrex flask and irradiated in a Rayonet Chamber reactor equipped with sixteen 8W 3500 A lamps.[12] After 5 days of irradiation the unreacted cinnamic acid was separated from the dimer by extraction with ether. The white crystalline residue was recrystallized from absolute ethanol to give 50 g of α-truxillic acid, mp 287–289° (lit.[13] mp 276–280°). Destructive ozonolysis of the α-truxillic acid afforded *cis,trans,cis*-1,2,3,4-tetracarboxycyclobutane.[8]

Electrolysis of α-Truxillic Acid (II).—α-Truxillic acid (5 g) (0.017 mole) was dissolved in 175 ml of dry methanol. Sufficient sodium metal (0.025 g) was added to convert *ca.* 6% of the acid to the sodium salt. The system was purged with dry nitrogen and protected with a drying tube. The solution was then electrolyzed

---

(10) G. W. Griffin and A. F. Vellturo, *J. Org. Chem.*, **26**, 5183 (1961).

(11) (a) K. B. Wiberg and R. P. Ciula, *J. Am. Chem. Soc.*, **81**, 5261 (1959); (b) S. Masamune, *ibid.*, **86**, 735 (1964); (c) J. Meinwald, C. Swithenbank, and A. Lewis, *ibid.*, **85**, 1880 (1963); (d) W. von E. Doering and J. F. Coburn, Jr., *Tetrahedron Letters*, **No. 15**, 991 (1965); (e) D. Seebach, *Angew. Chem. Intern. Ed. Engl.*, **4**, 121 (1965); (f) W. von E. Doering and M. Pomerantz, *Tetrahedron Letters*, **No. 17**, 961 (1964); (g) W. R. Moore, H. R. Ward, and R. F. Merritt, *J. Am. Chem. Soc.*, **83**, 2019 (1961); (h) K. B. Wiberg and G. M. Lampman, *Tetrahedron Letters*, **No. 30**, 2173 (1963); (i) D. M. Lemal, F. Menger, and G. W. Clark, *J. Am. Chem. Soc.*, **85**, 2529 (1963); (j) H. M. Frey and I. D. R. Stevens, *Proc. Chem. Soc.*, 144 (1964); (k) G. L. Closs and R. B. Larrabee, *Tetrahedron Letters*, **No. 4**, 287 (1965); (l) R. Srinivasan, *J. Am. Chem. Soc.*, **85**, 4045 (1963).

(12) Southern New England Ultraviolet Co., Middletown, Conn.

(13) H. I. Bernstein and W. C. Quimby, *J. Am. Chem. Soc.*, **65**, 1845 (1943).

between two smooth platinum electrodes (2 × 3 cm) at a potential of 98 v (dc) until it became basis to litmus (7 hr). The current varied from 0.16 to 0.40 amp during the course of the electrolysis. The resulting yellow solution was acidified with acetic acid, filtered to remove a small amount of insoluble material, and the filtrate was evaporated under reduced pressure to give 4.0 g of a viscous oil. This material was dissolved in a small amount of benzene and placed on a column (36 × 2.5 cm) of acid-washed alumina packed with cyclohexane. The column was then eluted with cyclohexane, cyclohexane–benzene mixtures, benzene, and finally methanol. The oil (600 mg) eluted with benzene slowly crystallized on standing. Recrystallization from ether–pentane gave the pure lactone VII: mp 131–133°. The nuclear magnetic resonance spectrum determined in deuteriochloroform exhibited signals at τ 2.52–2.64 (ten protons), τ 4.87 (one proton), τ 7.13, 7.22, 7.26, 7.32, 7.42 (three protons), and a carbonyl absorption band appears at 1757 cm^{-1} (KBr).

ϵ-Truxillic Acid.—A mixture of 15 g of α-truxillic acid and 60 g of potassium hydroxide was heated in an open beaker maintained at 300–320° by means of a beaker mantle. The melt was stirred occasionally with a glass rod until the mixture began to darken (usually 2–3 hr). The resulting product was allowed to cool and dissolved in 600 ml of water. The insoluble potassium silicate was collected on a filter and the light yellow filtrate was acidified with concentrated hydrochloric acid. The white precipitate was collected on a filter and crystallized from aqueous ethanol (Norit): yield 13.0 g, mp 190–191° (lit.[10] mp 192°).

***trans*,*trans*,*trans*-1,3-Dicarboxy-2,4-dicarbomethoxycyclobutane (III).**—ϵ-Truxillic acid (7 g) dissolved in ether was treated with excess diazomethane solution (ether). The reaction mixture, after standing overnight, was dried over anhydrous magnesium sulphate. Removal of the volatile solvent gave 6.5 g of dimethyl ϵ-truxillate XIV as a viscous, sweet smelling oil (lit. mp 64°). Gas–liquid partition chromatographic analysis on a 210 × 0.6 cm glass column packed with 30% SE-30 on 60–80-mesh acid- and base-washed Chromosorb P at 200° showed that the product was homogeneous. The ester was dissolved in 275 ml of acetic acid and 50 ml of water and ozonized for 20 hr at room temperature. To the resulting clear solution was added 75 ml of 30% hydrogen peroxode. The mixture was then allowed to stand at room temperature for 48 hr. The excess hydrogen peroxide was finally decomposed by the addition of a small amount of platinum black. After filtration the clear solution was evaporated under reduced pressure to a white crystalline solid which was rigorously dried in a vacuum desiccator over sodium hydroxide. Recrystallization from the solvent mixture methylethyl ketone–cyclohexane gave 3.1 g (52%) of white crystals: mp 173–175°; ν_{max}^{KBr} 3300–2500 (carboxyl OH), 1732 (ester C=O), and 1713 cm^{-1} (acid C=O). After repeated recrystallization from methyl ethyl ketone–cyclohexane the sample melted at 181–183°. Elemental analytical data for III were consistently poor; however, treatment with diazomenthane gave *trans,trans,trans*-1,2,3,4-tetracarbomethoxycyclobutane[9] which confirmed beyond doubt the structure of III.

Anal. Calcd for $C_{10}H_{12}O_8$: C, 46.16; H, 4.65. Found: C, 45.27, 45.64, 45.75; H, 4.80, 4.51, 4.68.

***cis*-2,4-Dicarbomethoxybicyclobutane (XVII).**—A 1.0-g (3.85 mmole) sample of the diacid–diester was dissolved in 40 ml of dry methanol. Enough sodium (0.010 g, 0.4 mmole) was added to convert 5% of the diacid–diester to the sodium salt. The system was purged with dry nitrogen before the reactants were added. After the addition of the sodium the system was sealed and the gases produced in the electrolysis were allowed to expand into a balloon. The solution was electrolyzed between two smooth platinum electrodes (2 × 3 cm) at a potential of 80–90 v (dc) until it became basic (4–5 hr). The current varied from 1.0 amp to 0.55 amp. During the course of electrolysis the reaction mixture was cooled with an ice bath. After the solution had become basic, most of the methanol was removed under reduced pressure while keeping the reaction mixture cold. The residual dark yellow solution (2–3 ml) was chromatographed on a 2 m × 6 mm column containing 30% SE-30 on Chromosorb P heated to 220°. Under these conditions a white crystalline solid, 0.10 g, was collected. Repeated sublimation of this material at 40–50° (0.05 mm) gave a sample, mp 83–85° with softening at 78–79°, ν_{max}^{KBr} 1729 cm^{-1} (ester C=O).

Anal. Calcd for $C_8H_{10}O_4$: C, 56.47; H, 5.92. Found: C, 56.63; H, 5.87.

Hydrogenation of *cis*-2,4-Dicarbomethoxybicyclobutane (XVII).—Platinum oxide (12 mg) was prereduced (1.0 psi of H_2) in 1 ml of dry dimethoxyethane. A 23-mg sample of the diester dissolved in 0.25 ml of dimethoxyethane was added to the reaction mixture contained in a serum-capped vessel by means of a syringe. The hydrogenation was continued for 20 hr at 1 psi. The catalyst was removed and the mixture was separated into three components on a 3 m × 10 mm column containing 30% Carbowax 20M on acid-washed Chromosorb P heated to 210°. The three components were identified as *cis*-1,3-dicarbomethoxycyclobutane, dimethyl adipate, and dimethyl α-methylglutarate (15:1:21) by their retention times and comparison of infrared spectra.

Acknowledgment.—The authors are indebted to the National Institutes of Health for financial support of this research.

---

(14) R. Stoermer and E. Emmel, *Ber.*, **53**, 497 (1920).

Synthesis of Benzocyclobutenes from Trichloromethylbenzenes[1a]

HAROLD HART,[1b] JAMES A. HARTLAGE, RICHARD W. FISH, AND ROBERT R. RAFOS

Department of Chemistry, Michigan State University, East Lansing, Michigan 48823

Received February 4, 1966

Trichloromethylpentamethylbenzene (I), on heating to 110°, is converted in good yield to the dichlorotetramethylbenzocyclobutene (II). The method constitutes a general synthesis of highly substituted benzocyclobutenes, as illustrated by the conversion of trichloromethylmesitylene (VIII) to dichlorodimethylbenzocyclobutene (IX) and trichloromethylchlorodurene (XII) to trichlorotrimethylbenzocyclobutene (XIII). The 1,1-dichlorobenzocyclobutenes thus formed are readily hydrolyzed to the corresponding benzocyclobutenones. Conversion of tetramethylbenzocyclobutenone (III) to the corresponding alcohol, bromide, nitrile, and carboxamide is described. Reduction of III with lithium aluminum hydride or sodium borohydride leads to ring-opened products (pentamethylbenzyl alcohol and hexamethylbenzene). Wolff–Kishner reduction of III, however, gives tetramethylbenzocyclobutene (XVIII). Ketones such as III are reversibly protonated on oxygen in concentrated sulfuric acid, as shown by nmr and ultraviolet studies. The nmr spectra of benzocyclobutenes with one substituent on the four-membered ring are briefly discussed.

One general method for the synthesis of benzocyclobutenes involves 1,4 elimination from adjacent benzylic positions, followed by ring closure of the resulting *o*-quinone dimethide. The original Finkelstein[2,3] synthesis of 1,2-dibromobenzocyclobutene is an example (X = Y = Br; reagent = I^-).[4] We wish to report that

---

(1) (a) For a preliminary account, see H. Hart and R. W. Fish, *J. Am. Chem. Soc.*, **82**, 749 (1960); (b) author to whom reprint requests should be addressed.

(2) H. Finkelstein, Ph.D. Dissertation, University of Strassburg, 1909; *Chem. Ber.*, **92**, 37 (1959).

(3) M. P. Cava and D. R. Napier, *J. Am. Chem. Soc.*, **79**, 1701 (1957).

(4) For other examples, see F. R. Jensen and W. E. Coleman, *ibid.*, **80**, 6149 (1958); M. P. Cava and A. A. Deana, *ibid.*, **81**, 4266 (1959).