and the cooled mixture was extracted with ether (50 ml). The extract was washed with water (20 ml) and dried (Na_2SO_4). The solid obtained from the ether was distilled to give 0.76 g (79% yield) of cyclotridecanone (bp 106–107° at 0.4 mm, mp 29–31°, lit.[24] mp 30–31°). The 2,4-dinitrophenylhydrazone of cyclotridecanone melted at 113–114°, lit.[25] mp 113–114°); the semicarbazone melted at 208–209°, lit.[25] mp 208°.

Registry No.—**5**, 7777-75-5; **4**, 7777-76-6; **17**, 7777-77-7; **16**, 7777-78-8; **6**, 7777-79-9; **6** adduct with tetracyanoethylene, 7777-80-2; **6** adduct with N-phenylmaleimide, 7777-81-3; **8**, 7777-82-4; **9**, 7777-83-5, **10**, 7777-84-6; **13**, 10026-26-3; **18**, 7777-85-7; cyclotridecyl ethyl ether, 7777-86-8.

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[24] N. J. Leonard and C. W. Schimelpfenig, Jr., *J. Org. Chem.*, **23**, 1708 (1958).

[25] H. Nozaki, S. Kato, and R. Noyori, *Can. J. Chem.*, **44**, 1021 (1966).

The Structure of Kaltwasser's Acid. A Novel Cyclopropane Formation

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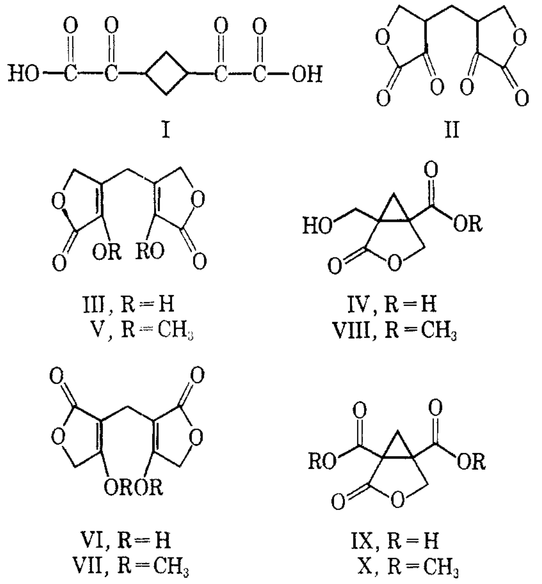
Received October 7, 1966

The structure of the acidic enol (Kaltwasser's acid) obtained by condensation of pyruvic acid with formaldehyde is confirmed as III. Silver oxide oxidation of III to the cyclopropyl lactone IV is also confirmed. The remarkable proposals made previously are thus essentially correct.

The condensation of formaldehyde and pyruvic acid in sulfuric acid was reported by Kaltwasser[1] to give a $C_8H_8O_6$ diacid (I) (Chart I). A reinvestigation of this work by Asahina and Terada[2] resulted in a revision of the molecular formula to $C_9H_8O_6$ and a proposal of structure II.[3] In support of this structure these authors report a remarkable oxidative degradation of II to IV, which was effected by silver oxide. The assignment of structure IV to the oxidation product rested on the carbon-hydrogen analysis and reduction (hydrogen iodide and phosphorous) to α,α'-dimethylglutaric acid. A few years after the publication of this work, Feofilaktov[4] repeated the silver oxide oxidation of II and found that product IV was further oxidized by alkaline permanganate to 1,1,2,2-cyclopropanetetracarboxylic acid. The identity of this latter compound was established by comparison with an authentic sample, synthesized from methylenebismalonic ester.[5]

CHART I



Since these early structural assignments are tenuous by present day standards, the unusual cyclopropane ring formation described above must be viewed with skepticism. Indeed, carbon-carbon bond formation seldom occurs during an oxidation reaction; the only well-substantiated examples are found in the oxidative coupling of phenolic compounds.[6]

Kaltwasser's acid, prepared according to Feofilaktov's procedure,[4] was obtained as white crystals, mp 236–238 dec, after several crystallizations from water. The infrared spectrum of this material exhibited strong absorptions at 3310 and 1725 cm^{-1}, indicating that if structure II is to remain as a reasonable representation of Kaltwasser's acid it must exist predominantly as the dienol III. In agreement with this conclusion, Kaltwasser's acid was observed to react rapidly with ethereal diazomethane yielding the dimethyl derivative V. The structure of V rested on classical analysis, the infrared spectrum (carbonyl stretching at 1740 cm^{-1}), and the nuclear magnetic resonance spectrum (three singlets at τ 6.47, 6.00, and 5.33 having an area ratio of 1:3:2, respectively). Unfortunately, Kaltwasser's acid is not uniquely characterized by these measurements, since the bistetronic acid (VI) and its dimethyl derivative VII fit the data equally well. Even more disturbing is the fact that VI has been unambiguously synthesized[7] and is reported to have a melting point of 235–238°, identical with that of Kaltwasser's acid. It is more difficult to formulate a rational mechanism for the synthesis of VI from pyruvic acid and formaldehyde than for the formation of III; however, this is insufficient reason to reject the former structure. A decision in this matter can be reached by considering changes in the ultraviolet absorption spectra of α-hydroxybutenolides and β-hydroxybutenolides upon the addition of base. An unexpected variation is observed in the data presented in Table I. Although the two classes of hydroxybutenolides have essentially the same high intensity absorption in neutral solution ($\lambda_{\max}^{\mathrm{EtOH}}$ ~235 mμ for a

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[1] O. Kaltwasser, *Ber.*, **29**, 2273 (1896).

[2] E. Asahina and S. Terada, *J. Pharm. Soc. Japan*, **502**, 855 (1923).

[3] An analogous structure was proposed for the product from the condensation of benzaldehyde and pyruvic acid: E. Erlenmeyer, *Ber.*, **32**, 1450 (1899). Other products from the formaldehyde condensation reaction have been identified by S. Olsen and G. Havre, *Acta Chem. Scand.*, **8**, 47 (1954).

[4] V. Feofilaktov, *J. Russ. Phys. Chem. Soc.*, **61**, 1145 (1929).

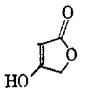
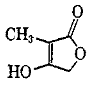
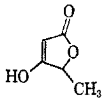
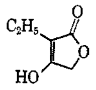
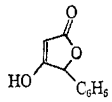
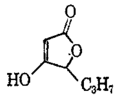
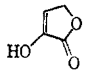
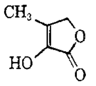
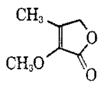
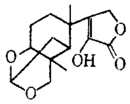
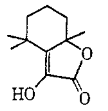
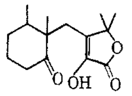
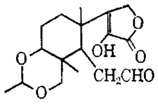
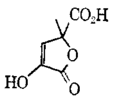
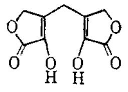
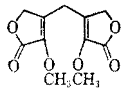
[5] O. Dressel, *Ann.*, **256**, 174 (1890).

[6] A. I. Scott, *Quart. Rev.* (London), **19**, 1 (1965).

[7] L. Wolff, *Ann.*, **315**, 145 (1901).

TABLE I

BASIC SHIFTS FOR α- AND β-HYDROXYBUTENOLIDES

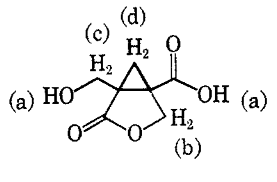
Compd	λ_{max}^{EtOH}, mμ (ε)	$\lambda_{max}^{+OH^-}$, mμ (ε)	Δ, mμ	Ref
	224 (14,500)	249 (22,800)	25	*a*
	233 (...)	258 (...)	25	*b*
	226 (15,900)	250 (...)	24	*a*
	233 (12,000)	258 (18,000)	25	*c*
	222 (13,500)	251 (21,200)	29	*a*
	223 (13,500)	... (...)	..	*d*
	226 (4,000)	261 (2,000)	35	*e*
	233 (12,600)	... (...)	..	*f*
	230 (11,000)	... (...)	..	*f*
	240 (7,900)	274 (10,700)	34	*g*
	238 (11,500)	274 (10,000)	36	*h*
	238 (7,600)	277 (8,500)	39	*i*
	233 (7,950)	272 (5,300)	39	*g*
	237 (6,310)	270 (7,900)	33	*h*
	240 (14,200)	286 (17,000)	46	..
	229 (14,000)	300 (10,700)	..	..

[a] L. Haynes and J. Plimmer, *Quart. Rev.* (London), **14,** 292 (1960). [b] A. I. Scott, "Interpretation of the Ultraviolet Spectra of Natural Products," Macmillan, New York, N. Y., 1964, pp 58, 79, 238, 241. [c] R. Herbert and E. Hirst, *Biochem. J.*, **29,** 1881 (1935). [d] E. R. Jones and M. C. Whiting, *J. Chem. Soc.*, 1419 (1949). [e] H. Hift and H. Mahler, *J. Biol. Chem.*, **198,** 901 (1952). [f] H. Schinz and M. Hinder, *Helv. Chim. Acta*, **30,** 1349 (1947). [g] C. Djerassi and W. Rittel, *J. Am. Chem. Soc.*, **79,** 3528 (1957). [h] C. Brooks and G. Eglinton, *et al.*, *J. Chem. Soc.*, 308 (1961). [i] T. A. Geissman, *J. Am. Chem. Soc.*, **75,** 4008 (1953).

monoalkyl derivative), the β-hydroxybutenolides exhibit a bathochromic shift of roughly 20–25 mμ when base is added, while the α-hydroxybutenolides show a corresponding shift of 35–40 mμ. The former shift is accompanied by a large increase in molar absorptivity, but the latter shift is characterized by only small increases in absorption intensity. Since Kaltwasser's acid shows a bathochromic shift of 46 mμ when an equivalent of base is added, it is assigned structure III.

With the structure of Kaltwasser's acid established as III, we turned to the silver oxide oxidation reaction which now emerged as the locus of the proposed cyclopropane formation. Treatment of III with freshly prepared silver oxide in refluxing water gave, after careful work-up, a 26% yield of a crystalline acid. The infrared spectrum ($\bar{\nu}_{max}^{KBr}$ 3440, 3100, 1775, and 1725 cm^{-1}) and the nmr spectrum (Chart II) establish the structure of this material as IV. Furthermore, a methyl derivative was prepared by reaction of IV with diazomethane, and the infrared spectrum ($\bar{\nu}_{max}^{KBr}$ 3500, 3100, 1770, and 1725 cm^{-1}) and nmr spectrum (Chart III) confirm structure VIII for this derivative.

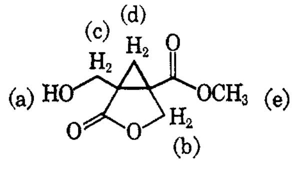
CHART II



IV

Chemical shift, τ	*J*, cps	Area	Assignment
3.61 (s)	...	2	a
5.35 (d)	9.5	1	b
5.68 (d)	9.5	1	b
5.75 (d)	12.5	1	c
6.32 (d)	12.5	1	c
7.95 (d)	5.5	1	d
8.63 (d)	5.5	1	d

CHART III



VIII

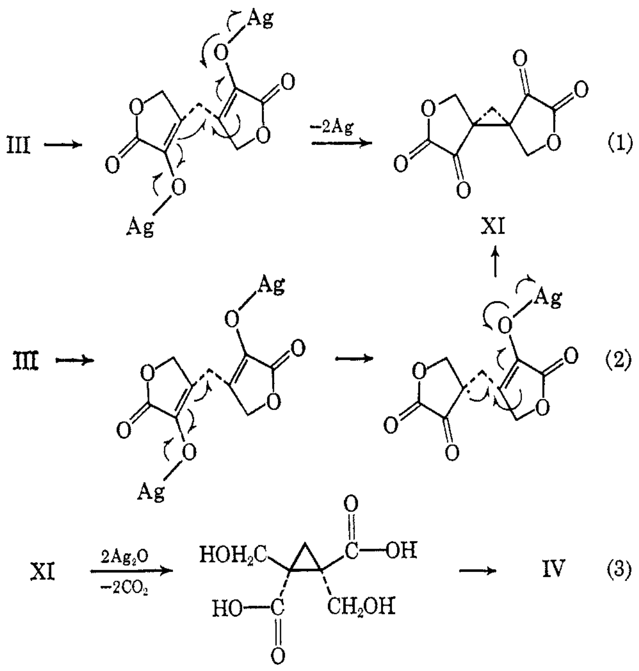
Chemical shift, τ	*J*, cps	Area	Assignment
5.25 (d)	9.5	1	b
5.68 (d)	12.5	1	c
5.75 (d)	9.5	1	b
6.19 (s)	...	3	e
6.21 (d)	12.5	1	c
7.46 (s)	...	1	a
7.94 (d)	5.5	1	d
8.64 (d)	5.5	1	d

In these compounds (IV and VIII) the methylene hydrogens of the cyclopropane ring are magnetically nonequivalent and give rise to an AB pattern. In a similar manner the methylene group of the lactone ring also appears as an AB system. Finally, the hydrogens of the hydroxymethylene group are adjacent to an asymmetric center and cannot become magnetically equivalent by rotation; hence an AB pattern is observed here as well. The magnitudes of the *geminal* coupling constants vary in a fashion that agrees with the theoretical relationship proposed[8] for J_{gem} and the H–C–H bond angle.

That IV does indeed contain a cyclopropane ring was established by oxidation to a mixture of diacid IX and 1,1,2,2-cyclopropanetetracarboxylic acid. These acids were isolated as the corresponding methyl esters by treating the crude oxidation products with diazomethane. The structure of X rests on the infrared spectrum ($\bar{\nu}_{max}$ 3090, 1790, and 1735 cm^{-1}) and the nmr spectrum, which retained the AB systems arising from the lactone and cyclopropane methylene groups (see the Experimental Section). The identification of 1,1,2,2-tetracarbomethoxycyclopropane was made by direct comparison with synthetic material using vpc, nmr, and infrared spectroscopy.

The facts presented above confirm the previous reports[2,4] of cyclopropane ring formation during oxidation of III by silver oxide, but contribute little to the intriguing question of how this reaction occurs. We offer the following mechanism (Scheme I) as a starting point for future discussion and work. The disilver salt of Kaltwasser's acid could lead to cyclopropane formation with loss of silver by either a one-step process (eq 1 of Scheme I) or a stepwise sequence (eq 2 of Scheme I). The stereospecificity of the reaction may be the result of steric repulsion between oxygen atoms (and perhaps silver atoms) and/or dipole interactions. Several attempts to effect this oxidation with ferricyanide have not led to identifiable products.

Scheme I



Experimental Section

A.—Infrared spectra were determined with a Perkin-Elmer 237B grating infrared spectrophotometer. Ultraviolet spectra were obtained with a Beckman D. B. spectrophotometer. Nuclear magnetic resonance spectra were obtained with a Varian A-60 high resolution spectrometer, using tetramethylsilane as an internal standard. Vapor phase chromatography was carried out with an Aerograph model A-90-P gas chromatograph. Melting points were determined with a Kofler hot-stage microscope. Microanalysis determinations were performed by Spang Microanalytical Laboratory, Ann Arbor, Mich.

B. Condensation of Pyruvic Acid with Formaldehyde. 1. 2,6-Diketo-3,5-dihydroxymethylheptanedioic Acid Dilactone (III) "Kaltwasser's Acid."—To a cooled (ice salt) pasty mixture of 40.0 g (0.455 mole) of freshly distilled pyruvic acid (bp 46° at 5 mm) and 20.4 g (0.680 mole) of paraformaldehyde, 48.0 ml of concentrated sulfuric acid was added dropwise with stirring. The addition was slow enough (1 hr) to maintain the temperature below 12°. After addition the solution was allowed to warm to room temperature, heated on a steam bath for 30 min, and allowed to stand at room temperature for 4 days. The dark brown solution (some crystals were present) was then poured into 300 ml of water and allowed to stand for 18 hr, during which time a tan solid precipitated. The mixture was cooled and filtered yielding about 16 g of crude product (34%), which upon decolorization (Norit) and crystallization from water yielded 12 g (25%) of III: mp 236–238° dec (lit.[4] mp 236–238°); $\bar{\nu}_{max}^{KBr}$ 3310, 1725, 1380, 1170, and 770 cm^{-1}; λ_{max}^{EtOH} 240 mμ (ϵ 14,200); $\lambda_{max}^{EtOH + OH^-}$ 286 mμ (ϵ 17,000).

2. Methylation of 2,6-Diketo-3,5-dihydroxymethylheptanedioic Acid Dilactone.—Ethereal diazomethane[9] was added slowly to a cooled ether slurry of 4.0 g of Kaltwasser's acid (III) until the diazomethane remained in excess. After standing (2 hr), enough methanol was added to dissolve the crystals and the ether and excess diazomethane were boiled away. Decolorization and crystallization from methanol yielded 2.57 g (56%) of V: mp 97°; $\bar{\nu}_{max}^{KBr}$ 2975, 2940, 2840, 1740, 1670, 1360, and 1110 cm^{-1}; λ_{max}^{EtOH} 229 mμ (ϵ 14,000); $\lambda_{max}^{EtOH + OH^-}$ 300 mμ (ϵ 10,700); nmr spectra (solvent $CDCl_3$), singlets at τ 5.33, 6.00, and 6.47 with relative areas 2.0:2.9:1.0.

Anal. Calcd for $C_{11}H_{12}O_6$: C, 54.99; H, 5.04; mol wt, 240. Found: C, 54.91; H, 4.84; mol wt, 257 (Rast determination).

C. Oxidation of Kaltwasser's Acid with Silver Oxide. 1. 1-Hydroxymethyl-5-carboxy-3-oxabicyclo[3.1.0]hexan-2-one (IV).—With stirring, 11.4 g of sodium hydroxide in 100 ml of water was added to a solution of 45.0 g of silver nitrate in 250 ml of water, and the resulting brown precipitate (Ag_2O) was washed twice (by decantation) with 300-ml portions of boiling water and collected by suction filtration. The damp silver oxide was then added immediately to a mixture of 6.0 g (0.028 mole) of Kaltwasser's acid (III) and 120 ml of water in a flask, equipped with a true-bore stirrer and reflux condenser. The mixture was refluxed with stirring for 6 hr, during which time a silver mirror was formed and carbon dioxide evolved. The black insoluble material (Ag and excess Ag_2O) was removed by filtration and centrifugation, and the resulting clear yellow solution was concentrated by means of a rotoevaporator to a volume of about 50 ml. After addition of 5% HCl dropwise, until it no longer precipitated any silver chloride, (if an excess of HCl was added the solution could be cleared again by addition of 10% Na_2CO_3) the silver chloride was removed by centrifugation and the resulting solution was concentrated on a steam bath until crystals appeared. The light tan solid (2.0 g), after decolorization with Norit and crystallization from acetone, yielded 1.28 g of IV (26.4%): mp 186–187°; $\bar{\nu}_{max}^{KBr}$ 3440, 3100, 3000–2600 (b), 1775, and 1725 cm^{-1}; nmr spectra (solvents D_2O and acetone-d_6), broad singlet at τ 3.61 with relative area 2.14 and closely spaced doublets at τ 5.35 (J = 9.5 cps), 5.68 (9.5), 5.75 (12.5), 6.32 (12.5), 7.95 (5.5), and 8.63 (5.5) all with relative areas of 1.0.

Anal. Calcd for $C_7H_8O_5$: C, 48.84; H, 4.685. Found: C, 48.87; H, 4.56.

2. 1-Hydroxymethyl-5-carbomethoxy-3-oxabicyclo[3.1.0]hexan-2-one (VIII).—Ethereal diazomethane was added to an ether slurry of 89.0 mg of IV, until the solution remained yellow. After standing (1 hr), the solution was boiled to remove excess diazomethane and concentrated until crystals began to appear. Filtration yielded 80.0 mg of white crystalline VIII: mp 99–102°; $\bar{\nu}_{max}^{KBr}$ 3500, 3100, 2930, 1770, and 1725 cm^{-1}; nmr spectra ($CDCl_3$), singlets at τ 6.19 and 7.46 with relative areas of 3.0:1.0, and doublets at τ 5.25 (J = 9.5 cps), 5.68 (12.5), 5.75 (9.5), 6.21 (12.5), 7.94 (5.5), and 8.64 (5.5) with relative areas of 1.2:1.0:1.1:1.1:1.0:1.0, respectively.

Anal. Calcd for $C_8H_{10}O_5$: C, 51.61; H, 5.414. Found: C, 51.52; H, 5.44.

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[8] H. Gutowsky, M. Karplus, and D. Grant, *J. Chem. Phys.*, **31**, 1278 (1959).

[9] F. Arndt, "Organic Syntheses," Coll. Vol. II, John Wiley and Sons, Inc., New York, N. Y., 1943, p 165.

D. Permanganate Oxidation of 1-Hydroxymethyl-5-carboxy-3-oxabicyclo[3.1.0]hexan-2-one (IV).—A mixture consisting of 300 mg of IV dissolved in 8.0 ml of 5% NaOH, 300 ml of water, and 736 mg of potassium permanganate was refluxed until the permanganate color disappeared (5 hr). The MnO_2 was removed by filtration and the resulting clear solution was concentrated (rotoevaporator) to a volume of a few milliliters, and acidified with 0.485 N H_2SO_4 (24 ml) to a pH of about 2. The solution was then evaporated to dryness on a rotoevaporator, and the resulting solid was treated with an ether solution of diazomethane until the yellow color persisted. After standing (2 hr), the excess diazomethane was removed by boiling, and the ether solution was washed with 5% $NaHCO_3$ and saturated NaCl solutions and dried over $MgSO_4$. The $MgSO_4$ was removed by filtration and the solution concentrated to a volume of a few milliliters. Analysis by vapor phase chromatography on a 0.25 in. × 5 ft 20% silicone SE 20 column at 193°, helium flow rate 40 cc/min, gave two fractions with retention times of 8 and 11 min from ether.

1. **1,5-Dicarbomethoxy-3-oxabicyclo[3.1.0]hexan-2-one (X).**—The 8-min fraction from the vapor phase chromatography was found to be X: $\bar{\nu}_{\max}^{\mathrm{CCl_4}}$ 3090, 2950, 1790, 1735, and 1460 cm^{-1}; nmr spectra (CCl_4), singlets at τ 6.23 and 6.27 with a combined relative area of 6.5, and doublets at τ 5.30 (J = 9.5 cps), 5.84 (9.5), 7.52 (5.0), and 8.45 (5.0) with relative areas of 1.1:1.2:-1.0:1.1, respectively.

2. **1,1,2,2-Tetracarbomethoxycyclopropane.**—Comparison of the melting point (69°) and infrared spectrum of the 11-min fraction from vapor phase chromatography with those of an authentic sample of 1,1,2,2-tetracarbomethoxycyclopropane showed them to be identical.

E. Synthesis of Authentic 1,1,2,2-Tetracarbomethoxycyclopropane.[5] 1. **Methylenebismalonic Ester.**—Freshly distilled diethylmalonate (64 g, 0.4 mole) of boiling point 99° (22 mm) was added dropwise with stirring to a dry flask containing 9.2 g (0.4 mole) of sodium dissolved in 300 ml of absolute ethanol (distilled from magnesium).[10] To the resulting clear solution 54.0 g (0.2 mole) of methylene iodide (CH_2I_2) was added dropwise and the solution was refluxed until no longer basic (2 hr). The ethanol was removed by distillation, water was added to dissolve the inorganic salts and the organic layer was taken up in ether. The ether layer was washed with water and 5% $NaHSO_3$ and dried over Na_2SO_4. After removing the ether the remaining oil was vacuum distilled at 1.0 mm and the resulting colorless liquid had the following properties: bp 152° (1.0 mm) (lit.[5] bp 192° at 12 mm); $\bar{\nu}_{\max}$ 2980, 2930, 2900, 1750, and 1730 cm^{-1}; nmr spectra (CCl_4), quartet at τ 5.83 (J = 7.0 cps) with relative area of 4.0, and triplets at τ 6.57 (7.5), 7.64 (7.5), 8.76 (7.0) with relative areas of 1.0:1.0:6.0.

2. **1,1,2,2-Tetracarboethoxycyclopropane.**—A solution of 11.46 g (0.0345 mole) of methylenebismalonic ester in 50 ml of benzene was added to a stirred dispersion of 3.5 g (0.078 mole) of 52.8% sodium hydride on mineral oil, in 250 ml of benzene (dried over NaH). Bromine was then added to the cooled solution (0°) until the bromine brown color persisted (2.1 ml required), and the solution was refluxed for 4 hr. After cooling, water was added slowly to destroy the excess NaH and the benzene layer was washed with water and 5% $NaHSO_3$. The liquid remaining upon removal of the benzene was vacuum distilled (bp 150–166° at 1 mm) and was found by nmr analysis to be partially reacted starting material. The distillate was then added dropwise to a stirred dispersion of 1.2 g of 52.8% NaH on mineral oil, in 350 ml of benzene, and refluxed for 3 days. The reaction was worked up as before and distilled; the 135–150° (1.0 mm) fraction was crystallized from a pentane–ether mixture and yielded 3.9 g of 1,1,2,2-tetracarboethoxycyclopropane (35%). The physical properties were mp 43° (lit.[5] 43°); $\bar{\nu}_{\max}^{45°}$ 3100, 2980, 1740, and 1250 cm^{-1}; nmr spectra (CCl_4), a quartet at τ 5.95 (J = 7.0 cps), a singlet at τ 8.08, and a triplet at τ 8.85 (J = 7.0 cps) with relative areas of 4.0:1.0:6.0.

3. **1,1,2,2-Cyclopropanetetracarboxylic Acid.**—A solution of 2.0 g (6.05 mmoles) of 1,1,2,2-tetracarboethoxycyclopropane in 10 ml of glyme and 2.29 g (13.4 mmoles) of barium hydroxide in 280 ml of water were mixed and refluxed until the mixture was no longer basic (2 hr). During this time an insoluble barium salt of the carboxylic acid precipitated. The solution was acidified with 0.485 N H_2SO_4 to a pH of about 2 and allowed to equilibrate. The barium sulfate was removed by filtration, the filtrate was evaporated to dryness (rotoevaporator), and the resulting white solid was taken up in hot methyl ethyl ketone. Concentration, cooling, and precipitation by addition of *n*-hexane yielded 0.66 g of 1,1,2,2-cyclopropanetetracarboxylic acid (50%). The physical properties were mp 190–200° dec and CO_2 evolution (lit.[11] mp 200, 208, 214, and 210–212°); $\bar{\nu}_{\max}^{\mathrm{KBr}}$ 3300–2500 (b), 1725, 1420, and 1275 cm^{-1}; nmr spectra (acetone-d_6), singlets at τ −0.9 and 7.87. Thin layer chromatography using silica gel G with ethanol as the eluent, and visualizing by spraying with saturated methanolic solution of "Mallinkrodt IndicatAR pH 3–4," showed only one acid spot.

4. **1,1,2,2-Tetracarbomethoxycyclopropane.**—When 100 mg of 1,1,2,2-cyclopropanetetracarboxylic acid was treated with excess diazomethane, according to the procedure described previously, and crystallized from ether 30 mg of 1,1,2,2-tetracarbomethoxycyclopropane (20%) was obtained. The physical properties were melting point very sharp at 69° (lit.[11] 71°); $\bar{\nu}_{\max}^{\mathrm{CCl_4}}$ 3100, 2950, 1740, 1435, 1370, 1240, and 1110 cm^{-1}; nmr spectra (CCl_4), two singlets at τ 6.23 and 7.82 with relative areas of 6.1:1.0. Vapor phase chromatography of the product using a 20% silicone SE 20 column at 193°, showed only one peak (retention time, 11 min from ether). Comparison (infrared spectrum and melting point) of the injected and collected material showed them to be identical.

Registry No.—III, 949-33-7; IV, 7605-60-9; V, 785-06-8; VIII, 7605-61-0; X, 7605-62-1; methylenebismalonic ester, 2121-66-6; 1,1,2,2-tetracarboethoxycyclopropane, 7605-64-3; 1,1,2,2-cyclopropanetetracarboxylic acid, 7605-65-4; 1,1,2,2-tetracarbomethoxycyclopropane, 7605-66-5.

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(10) A. Vogel, "A Textbook of Practical Organic Chemistry," Longmans, Green and Co., London, 1957, p 167.

(11) F. K. Beilstein, "Handbuch Der Organischen Chemie," Vol. 9, Second Supplement, 4th ed, Springer-Verlag, Berlin, 1949, System No. 1021 (H 990).