n_D^{25} 1.5232[3]; see below for comparison of melting points of methiodides of XIV and III).

Anal. Calcd. for $C_{14}H_{23}N$: C, 81.89; H, 11.29; N, 6.82. Found: C, 81.95; H, 11.39; N, 6.63. Ultraviolet absorption spectrum, calcd.[4] λ_{max} 318 mμ. Found: λ_{max} 321 mμ (3.9).

Also there was obtained from the above distillation 2.3 g. (8%) of the thermally rearranged product, aromatic amine XV, b.p. 88–89.5° at 0.4 mm., and 1.2 g. (4%) of undistillable residue. Considerable polymeric material (12.2 g., approximately 39%) remained from the steam distillation of XIV.

Thermal isomerization of XIV to form β-(2,3,4,6-tetramethylphenyl)ethyldimethylamine (XV). Crude *exo*-methyleneamine XIV, prepared from 19.3 g. (0.08 mole) of XIII and 0.24 mole of sodium amide, was thermally isomerized at 150° for 1 hr. to produce 7.4 g. (45%) of β-(2,3,4,6-tetramethylphenyl)ethyldimethylamine (XV), b.p. 90–90.5° at 0.5 mm., n_D^{25} 1.5163 (reported b.p. 88.5–89° at 0.45 mm., n_D^{25} 1.5162).[3] The picrate, recrystallized three times from 95% ethanol, melted at 178–179° (reported m.p. 179.5–180°).[3] Its melting point was not depressed on admixture with an authentic sample of the compound.

Reaction of exo-methyleneamine XIV with hydrochloric acid to form pentamethylbenzene. This reaction was carried out with an ethereal solution of crude *exo*-methyleneamine XIV (obtained from 0.07 mole of quaternary salt XIII and 0.21 mole of sodium amide) and 200 ml. of 6N hydrochloric acid as described for the acidic decomposition of V. The ethereal extract (containing neutral products) was steam-distilled, and the distillate worked up to yield 5.3 g. (51%) of pentamethylbenzene, m.p. 51–52°, after crystallization from absolute methanol (cooled to −78°). A mixed melting point determination with an authentic sample of this hydrocarbon, m.p. 52–53°, gave no depression in the melting point.

When the aqueous acidified extract (containing dimethylmethyleneiminium chloride) was made alkaline with sodium hydroxide solution, pungent fumes of formaldehyde were detected.

Rearrangement of the methiodide of exo-methyleneamine XIV (Formula XVI) and acidic decomposition of its exo-methyleneamine product to form hexamethylbenzene. Tertiary amine XIV (13.0 g., 0.063 mole), b.p. 62–62.5° at 0.4 mm., was converted into *exo*-methyleneamine methiodide XVI, m.p. 176–177° (dec.), in 98% yield by the action of 0.10 mole of methyl iodide on XIV in acetonitrile solution. The dissimilarity of this methiodide with that from *exo*-methyleneamine III [Formula XVII, m.p. 188–189° (dec.)][2] was demonstrated by the depression in melting point to 140–152° on admixture of these two quaternary ammonium salts.

To a stirred suspension of 0.19 mole of sodium amide in 300 ml. of liquid ammonia was added rapidly 21.2 g. (0.062 mole) of methiodide XVI, the resulting reddish violet colored reaction mixture being decomposed by the addition of ammonium chloride after 1 hr. The ammonia was replaced by ether, the suspension filtered, and the ethereal solution shaken with 200 ml. of 6N hydrochloric acid for 5 min. The two layers were separated. From the ether layer there was obtained 4.8 g. of impure hexamethylbenzene, m.p. 142–149°, which, after two crystallizations from ethanol afforded 4.2 g. (42%) of colorless hexamethylbenzene, m.p. 162–163°. This melting point was not depressed on admixture with an authentic sample of hexamethylbenzene, m.p. 162.5–164°.

From the aqueous hydrochloric acid layer (see above) there was obtained, on neutralization with sodium hydroxide and extraction with ether, 1.3 g. of unidentified material, b.p. 89–110° at 0.5 mm. (having a slight camphoric odor), and 2.7 g. of dark undistillable residue.

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Unsaturated Four-Membered Ring Compounds. I. *cis*- and *trans*-1,2-Diiodobenzocyclobutene and Their Interconversion

FREDERICK R. JENSEN AND WILLIAM E. COLEMAN

The reaction of $\alpha,\alpha,\alpha',\alpha'$-tetrabromo-*o*-xylene with excess sodium iodide in ethanol gives two isomeric $C_8H_6I_2$ compounds. Evidence is presented which indicates these compounds are *cis*- and *trans*-1,2-diiodobenzocyclobutene. The stereochemical relationship of the isomers was assigned by dipole moment measurements. The *cis*-isomer (m.p. 150°) melts 87° higher than the *trans*-isomer (m.p. 63°). These compounds are readily interconverted by light in the presence of iodine in a free radical reaction, and by sodium iodide in ethanol in a reaction which is postulated to involve benzocyclobutadiene as an intermediate. At 30°, the equilibrium mixture contains 6% of the *cis*-isomer.

In his Inaugural Dissertation, Finkelstein reported the preparation of 1,2-dibromobenzocyclobutene (I) by refluxing sodium iodide with $\alpha,\alpha,\alpha',\alpha'$-tetrabromo-*o*-xylene in ethanol solution.[1] Except for a brief reference to these results in connection with other work,[2] the details of this interesting reaction were not reported further until recently when Cava and Napier[3] reinvestigated and confirmed this preparation.

In this reaction an iodine-containing side product is produced which has been shown to be 1,2-diiodobenzocyclobutene (II). The same compound was also prepared in 70% yield by refluxing 1,2-dibromocyclobutene with ethanolic sodium iodide containing some iodine for eight days. As a side product in these reactions, a compound, $C_{16}H_{12}BrI_3$ (m.p. 135–136°), has been reported. The parent hydrocarbon, benzocyclobutene (III) was prepared by hydrogenolysis of (I) or (II).[3]

In attempting to prepare 1,2-diiodobenzocyclobutene directly by treating $\alpha,\alpha,\alpha',\alpha'$-tetrabromo-*o*-xylene with a large excess of sodium iodide we obtained two isomeric $C_8H_6I_2$ compounds. This re-

(1) H. Finkelstein, Inaugural Dissertation, Strassbourg, 1910, carried out with J. Thiele.

(2) H. Finkelstein, *Ber.*, **43**, 1532 (1910).

(3) M. P. Cava and D. R. Napier, *J. Am. Chem. Soc.*, **78**, 500 (1956); M. P. Cava and D. R. Napier, *J. Am. Chem. Soc.*, **79**, 1701 (1957).

port concerns the assignment of structure to these two compounds and reactions for their interconversion.

Results. Heating $\alpha,\alpha,\alpha',\alpha'$-tetrabromo-*o*-xylene with excess sodium iodide in ethanol for 10 days produced two isomeric $C_8H_6I_2$ compounds in 80.9% yield. The low-melting isomer, prisms m.p. 63°, which comprised 91.6% of the mixture, has been postulated to be one of the two possible 1,2-diiodobenzocyclobutenes.[3] This compound was believed to be the *trans*-isomer on the basis of the instability expected for the *cis*-isomer.

The other compound, needles m.p. 150° (8.4%), is an isomer as shown by elementary analyses and molecular weight determinations. Oxidation of this high-melting isomer with nitric acid gave a 74.1% yield of phthalic acid. There are three possible structures for this compound II, IV, or V. This compound does not add halogen when treated with bromine or iodine at room temperature.[4] Heating the high-melting isomer with maleic anhydride in benzene for 15 hr. produced no reaction. The recovered starting material (73.6%) contained some of the low-melting isomer.

I, X = Br IV V, X = I
II, X = I VI, X = Br
III, X = H
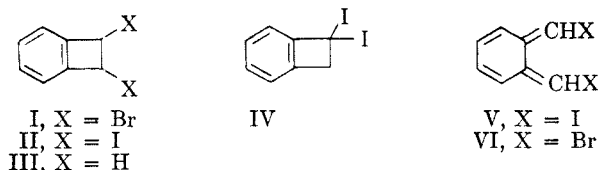
Reaction of the isomeric $C_8H_6I_2$ compounds with sodium iodide. The diiodides are interconverted by sodium iodide in refluxing ethanol. Small samples of each of the isomers were heated with alcoholic sodium iodide in the dark and the position of equilibrium determined by infrared analysis. Equilibrium was essentially obtained in 100 hr. and the equilibrium mixture contained about 9% of the high-melting isomer. In another experiment, a large sample of the low-melting isomer was heated with alcoholic sodium iodide in the dark for 8 days. The mixture was separated by recrystallization and found to contain 8.8% of the high-melting isomer (80% recovery).

In these reactions a small amount of decomposition occurred with the production of free iodine. In order to determine the cause of isomerization, samples were heated alone in ethanol and with added iodine.

Starting with the high-melting isomer, the pure compound was isomerized less than 3% and samples containing a little iodine were isomerized about 30% in the time necessary (92 hr.) for complete equilibration by sodium iodide. In another

series of experiments the samples were heated 36 hr. A sample (0.037*M*) containing 0.5 mole per cent iodine was isomerized 7%, while a sample containing 63 mole per cent sodium iodide was isomerized 70%. Increasing the iodine concentration resulted in an increased amount of isomerization. The low-melting isomer is also isomerized by iodine or sodium iodide in refluxing ethanol.

All reactions were carried out in the dark, and the product compositions were determined by infrared analysis.

Interconversion of the $C_8H_6I_2$ compounds with light. Certain observations indicated that these isomers might be interconverted by light, and this was verified experimentally.

Since the infrared spectral changes are difficult to follow starting with the low-melting isomer, the majority of the studies were carried out with the high-melting isomer. The samples were dissolved in carbon disulfide solution. Although the effect of the solvent was not determined, the isomerizations also occur readily in carbon tetrachloride and hexane as solvents. The irradiation method was standardized in order to carry out semi-quantitative studies.

Both compounds were irradiated in order to determine the position of equilibrium. The compounds are equilibrated in about 4 hr. under the conditions of our experiments and the mixture contains about 6% of the high-melting isomer at 30°. In order to determine the conditions necessary for isomerization to occur, a series of experiments were carried out with the high-melting isomer. Initially, no isomerization occurred until iodine color was visible in the solution (about 5–10 min.). When a trace amount of iodine was added to the solution before starting the irradiation, the induction period no longer existed (Fig. 1). Removing the samples from the light source stopped the reactions. After allowing the samples to stand in the dark for 24 hr. the iodine color persisted, and placing the sample in the light source again caused the sample to isomerize further with no induction period. The effect of the iodine concentration on the rate of reaction was not studied in detail. Addition of copper, which caused the iodine color to disappear, essentially stopped the isomerization. The color of free iodine did not appear until the solution had been irradiated more than 24 hr. although the sample was partially decomposed. After prolonged irradiation, the surface of the copper became coated, the iodine color appeared, and the sample isomerized. During the period where no isomerization occurred, the sample was appreciably decomposed.

The changes observed in the infrared spectra during the isomerization of the low-melting isomer were small. In order to verify the results obtained by infrared analysis, a large sample was irradiated overnight in hexane and the isomers separated by recrystallization. The high-melting isomer ac-

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(4) Although net addition of halogen does not occur when the isomers are treated with iodine or bromine, treatment of either $C_8H_6I_2$ compound with bromine converts it almost immediately to $C_8H_6Br_2$. The details of this and other reactions of these isomers will be reported in a future communication.

counted for about 6.8% of the recovered material.

Dipole moments of the isomeric diiodides. The dipole moments of the isomers were determined at 25° in benzene solution. The values obtained are for the low-melting isomer (m.p. 63°), $\mu = 1.84$ Debye, and for the high-melting isomer (m.p. 150°), $\mu = 2.51$ Debye.

Discussion. The high-melting isomer, $C_8H_6I_2$ m.p. 150°, must possess either structure II or IV on the basis that oxidation gives phthalic acid, it is nonreactive towards maleic anhydride,[3] and net addition of halogen does not occur upon treatment with bromine.[4]

The interconversion of the two isomers by iodide ion proceeds many times faster than the displacement reaction of iodide ion on cyclobutyl bromide.[5] The latter reaction has a half-life of about 120 days in acetone at 70° (initial concentrations 1 molar), whereas the equilibration is about complete in 100 hr. at 82°. Since this interconversion is catalyzed by iodide ions and proceeds readily in the dark, it is unlikely that it occurs by a process involving free radicals. The reaction probably does not proceed through carbonium ions since negligible amounts of ether are formed even after refluxing for 12 days in ethanol. A possible mechanism for the isomerization occurs through benzocyclobutadiene as an intermediate formed by elimination of iodine, which suggests that these compounds are *cis-* and *trans-* 1,2-diiodobenzocyclobutene. These arguments have

been used previously to account for the iodide ion-catalyzed conversion of I to II.[3]

The interconversion of the isomers with iodine and heat in the dark was not studied in detail. The isomerization could conceivably proceed as a result of iodine serving as a Lewis acid to cause elimination, or through a reaction similar to that brought about by iodine and light. In the presence of sodium iodide, the small amount of iodine produced by decomposition is converted to sodium triiodide. In elimination reactions catalyzed by iodide ion, the triiodide ion is ineffective as a catalyst, and there is no reason to believe that it has an important function in the reactions given here.

The interconversion of these isomers by light in the presence of iodine can also be readily accounted for on the basis that they are *cis-trans* isomers. The observation that the reaction occurs with very low level illumination and other evidence strongly suggest that the reaction occurs by a chain process involving iodine atoms. Possible reaction schemes include the formation of benzocyclobutadiene as an intermediate. In the reaction scheme given here, chain terminating steps are not included.

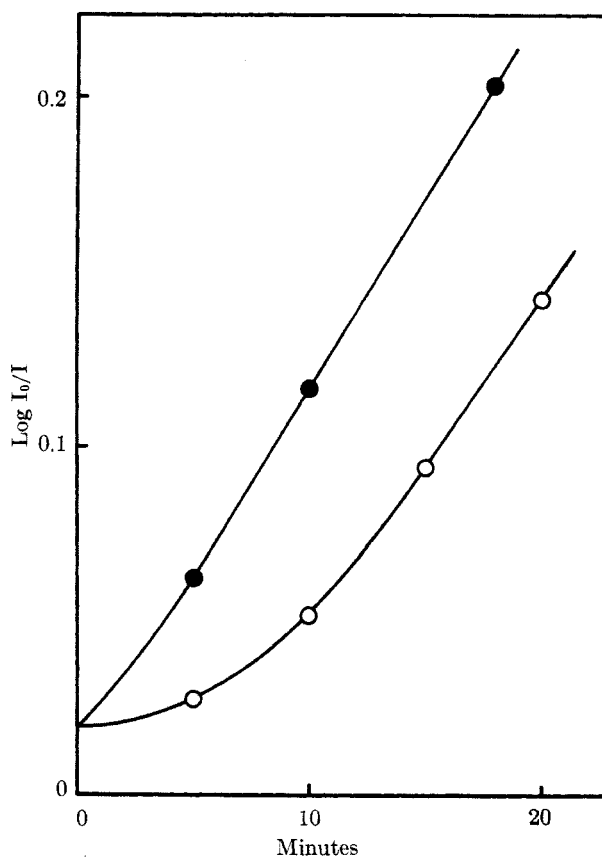(5) P. J. C. Fierens and P. Verschelden, *Bull. soc. chim. Belges*, **61**, 427 (1952).

Fig. 1. Effect of added iodine on the rate of the light-catalyzed isomerization of *cis*-1,2-diiodobenzocyclobutene as shown by the formation of the 11.5 μ peak of the *trans*-isomer. Open circles, *cis*-isomer 0.5 g./5 ml. CS_2 solution; closed circles, *cis*-isomer 0.5 g./5 ml. CS_2 solution containing 0.003 g. iodine

It is very difficult to account for the interconversion of II and IV by iodide ions or light and iodine, and therefore it shall be assumed that these compounds are *cis-* and *trans*-1,2-diiodocyclobutene

$$I_2 + h\nu \longrightarrow 2 I\cdot$$
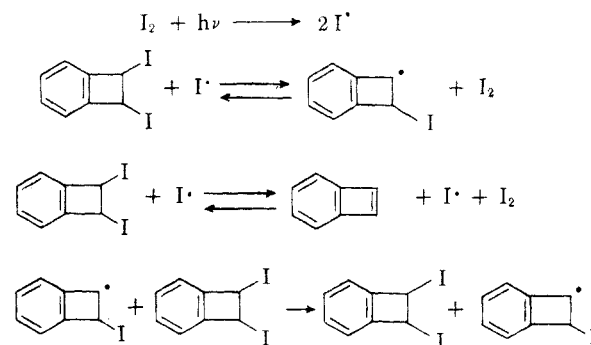
(II). The chemistry of this system is unusual and it would be desirable to have final proof by independent synthesis of IV. Since the isomers are readily interconverted, it is difficult to fix the positions of the iodine atoms by chemical reactions.

The position of equilibrium greatly favors the low-melting isomer which suggests that it is the *trans*-isomer since the iodine atoms might be expected to be badly crowded in the *cis*-compound.

Proof of this assignment was found from the dipole moment measurements.

The 87° higher melting point of the *cis*-isomer deserves some comment. Although the four membered ring might not be entirely planar in this molecule, nevertheless, it should have essentially a single-bend type structure. Such a structure could conceivably pack more favorably in the crystal lattice than that of the *trans*-isomer. The melting points of several esters of 3-aryl-2-isoxazoline-3,4-dicarboxylic acids, aryl—C̄=N— O—CH(CO₂R)CH-(CO₂R), are higher for the *cis*- than for the *trans*-compounds.[6] These compounds also possess the single-bend type structure.

The known isomer of I, m.p. 52°, and *trans*-II, m.p. 63°, appear to be isomorphic since it is almost impossible to separate them by recrystallization. Also, the infrared spectra of I and *trans*-II have many similar features. For these reasons, it seems very likely that the known isomer of I is the *trans*-compound.

There is no evidence suggesting that isomerization of I occurs under the conditions of its preparation. It seems likely that the reaction of sodium iodide with α,α,α′,α′-tetrabromo-o-xylene gives directly *trans*-I. Cava and Napier[3] have suggested that this reaction occurs through a 1,4-elimination to give VI as an intermediate. A reasonable pathway for conversion of VI to I would be through formation of the carbon-carbon bond in a one step process. In this reaction scheme, it might be expected that a considerable amount of the *cis*-isomer would be formed since in the transition state the substituent atoms are not fully opposed. However, the *cis*-isomer has not been isolated from the reaction mixture. The following mechanism might be expected to give predominantly the *trans*-isomer. A mechanism in which this over-all process occurs in a concerted fashion should also give predominantly
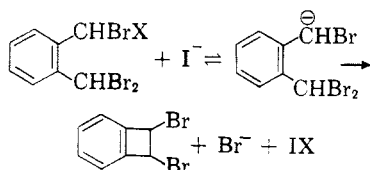
the *trans*-isomer since in these mechanisms the atoms would be nearly fully opposed in the transition states.

EXPERIMENTAL

cis- and *trans-1,2-Diiodobenzocyclobutene.* A mixture of 805 g. (5.4 moles) sodium iodide and 35 g. (0.21 mole) potassium iodide was refluxed with 2500 ml. absolute ethanol for 20 min. and then 700 ml. of ethanol was distilled. To this mixture was added 316 g. (0.75 mole) α,α,α′,α′-tetrabromo-o-

xylene,[7] and the mixture was refluxed for 10 days. The reaction vessel was carefully covered to exclude light and the solution was protected from moisture by a drying tube containing calcium sulfate. After cooling the mixture, 800 ml. methylene chloride was added and the resultant slurry was poured into 5000 ml. cold water. The water layer, which was nearly neutral, was titrated with sodium thiosulfate solution and 94% of the calculated amount (349 g.) was used. The solution was shaken to bring the iodine from the methylene chloride into the water later. The water layer was extracted three times with methylene chloride, the extractions were combined and the solvent was removed by flash distillation. The residue was distilled at 0.5 mm. (90–100°) and 224 g. was collected. Upon allowing to cool, two types of crystals, needles, and prisms, were observed. The mixture was separated by recrystallization from ether-hexane or heptane. Alternate fractions of each compound were taken, and if both types of crystals came down together, the mixture was taken back into solution. A total of 13 fractions were collected (216.1 g., 80.9%). Of this material, 18.2 g. (8.4%) was needles and 197.9 g. (91.6%) was prisms. We were unable to find any of the compound, $C_{16}H_{12}BrI_3$, reported previously as a side product in the reaction of 1,2-dibromocyclobutene with sodium iodide.[3]

The prisms (*trans*-1,2-diiodobenzocyclobutene, m.p. 62–63°) has been reported previously for essentially this same reaction.[3] The melting point of the needles [*cis*-1,2-diiodobenzocyclobutene, m.p. 150.1–150.8° (dec.)] depends on the rate of heating and whether or not the sample is exposed to strong light. The melting point reported was taken using a preheated bath and essentially in the dark. The melting point taken in the usual manner was 146–146.5°, although traces of impurities greatly lowered this value.

Anal. Calcd. for $C_8H_6I_2$: C, 26.99; H, 1.70; I, 71.31; mol. wt., 356. Found for the needles (m.p. 150.1–150.8°): C, 26.99; H, 1.82; I, 71.04; mol. wt., 334, 380 (isothermal distillation in carbon disulfide).

Oxidation of the high-melting diiodobenzocyclobutene. The diiodide (m.p. 150°, 1.198 g.) and concd. nitric acid (5 ml., d. 1.42) were added to a 20-ml. beaker, the beaker was covered and the temperature was slowly brought to 50° and maintained at this temperature for 30 min. The cover was removed and the cover and beaker sides were scraped down with a porcelain spatula. The mixture was then heated on a steam bath until all the iodine color had disappeared and the volume was 3 ml. The almost colorless solution was cooled in ice, the crystals were filtered with suction and washed twice with water. The phthalic acid was obtained in 74.1% yield (0.421 g.), m.p. 191–198° with the evolution of a gas.

Anal. Calcd. for $C_8H_6O_4$: C, 57.83; H, 3.71; neut. equiv., 83. Found: C, 57.36; H, 3.77; neut. equiv., 82.

A small sample of the phthalic acid was heated with acetic anhydride allowing the acetic acid formed to distill. The phthalic anhydride formed was sublimed, m.p. 128–129°.

Treatment of the high-melting diiodide with maleic anhydride. The high-melting diiodide (1.0 g.) was melted on a steam bath with maleic anhydride (3 g.). After heating the mixture for 3 hr., 75 ml. benzene was added and the mixture was refluxed for 15 hr. The flask was carefully covered to exclude light. The condenser was set for distillation, 40 ml. of benzene was distilled, 40 ml. water was added, and the remaining benzene was removed by steam distillation. The water solution was brought to a boil and filtered. The precipitate was washed twice more with 25-ml. portions of boiling water. The dried residue weighed 0.737 g. (74% recovery). An infrared spectrum revealed no carbonyl band, but did show, by the presence of a small band at 11.5 μ, that a small amount of isomerization had occurred to give the low-melting isomer.

(6) A. Quilico and P. Grunanger, *Gazz. chim. ital.*, 85, 1449 (1955).

(7) J. C. Bill and D. S. Tarbell, *Org. Syntheses*, 34, 82 (1954).

Treatment of the low-melting diiodobenzocyclobutene with sodium iodide. The diiodide (m.p. 63°, 15 g.) and sodium iodide (60 g.) were refluxed in 180 ml. absolute ethanol 8 days and the reaction mixture was worked up as before. Seven crops of crystals were obtained from heptane. A total of 12.13 g. of needles and prisms were isolated (81% recovery). Of this, 1.07 g. (8.8%) was the high-melting diiodide and 11.06 g. (91.2%) was the low-melting isomer.

In other experiments small samples were isomerized in refluxing ethanol in the presence of added sodium iodide, iodine, or alone. Light was carefully excluded during the heating periods. After cooling, the reaction mixture was poured into water and sodium sulfite was added until the iodine color disappeared. The compound was extracted with methylene chloride, the methylene chloride solution was washed with water, and the methylene chloride was removed under reduced pressure. The samples were dissolved in carbon disulfide and the isomer distribution was determined by infrared analysis using a Baird Associates double beam recording spectrophotometer and 0.1-mm. sodium chloride cells.

The concentration of the low-melting isomer was determined using the 11.5 μ-peak at a concentration 0.1 g./ml. CS_2 solution. The concentration of the high-melting isomer (at low concentrations) was determined using the 8.8-μ peak at a total concentration 0.5 g./ml. CS_2 solution with samples of the low-melting isomer in the reference beam. The calibration curve was made up using the same concentrations of the low-melting isomer in the reference and sample beams. Once the concentrations were approximately known, the unknown was determined using the concentration of the low-melting isomer in the reference beam approximately equal to that in the sample beam.

With initial concentrations of the high-melting isomer $0.071M$ and sodium iodide $0.041M$, the concentration of the high-melting isomer was found to be 9.4% after 102 hr. With initial concentrations of the high-melting isomer about $0.03M$ and sodium iodide $0.02M$, the concentration of the high-melting isomer was 37% after 36 hr. and 10.6% after 92 hr.

In experiments with the low-melting isomer, it was found that this compound is also isomerized by both iodine and sodium iodide in refluxing ethanol. Because the spectral changes in going from pure compound to the position of equilibrium are difficult to determine accurately, the reactions were not studied in detail. However, qualitatively the results indicated that this isomer is also isomerized faster by sodium iodide than by iodine. After heating the low-melting isomer $(0.071M)$ for 102 hr. with sodium iodide $(0.041M)$, the concentration of the high-melting isomer was 8.8%.

In refluxing ethanol, the best value for the concentration of the high-melting isomer at equilibrium is believed to be about 9%.

Light-catalyzed isomerizations of the 1,2-diiodobenzocyclobutenes. The irradiations were carried out using a 75-watt Sylvania flood lamp placed 5 inches from the soft glass volumetric flask which was used as the reaction vessel. The concentrations of the isomers were determined by infrared analysis of samples taken directly from the reaction flasks. All concentrations were 0.5 g./5 ml. CS_2 solution. The findings are given in Results.

The position of equilibrium was verified by irradiating 100 g. of the low-melting isomer in 500-ml. hexane for 12 hr. From the solution, 5.8 g. (6.8%) of the high-melting isomer and 80 g. (93.2%) of the low-melting isomer were obtained.

The best value for the concentration of the high-melting isomer at equilibrium (30°) is believed to be about 6%.

Dipole moment measurements. The measurements were carried out in dilute solution at 25°.[8] The dipole moments were calculated from the dielectric constants, densities, and refractive indices (Na_D line) of the solutions using a standard method.[9] The formulas used are:

$$P_{1,2} = C_1P_1 + C_2P_2 = [(\epsilon - 1)/(\epsilon + 2)][(C_1M_1 + C_2M_2)/d];$$
$$R_{12} = C_1R_1 + C_2R_2 = [(n^2 - 1)/(n^2 + 2)][(C_1M_1 + C_2M_2)/d]; \text{ and } \mu_{298} = (0.2192)(P_2 - R_2)^{1/2} \text{ Debye.}$$

The dielectric constants were measured at four different concentrations. In the concentration range used, the values of P_2 found were essentially constant. The dipole measure-

	Concentration Range (Mole Fraction)	P_2 (P_∞)	R_2 (R_D)	μ (Debye)
Needles, m.p. 150.1–150.8°	0.0242–0.00237	196 ± 3	64.3	2.51
Prisms, m.p. 62–63°	0.0368–0.00362	134 ± 2	63.8	1.84

ment cell was placed in a constant temperature bath at 25°. The cell and a standard variable capacitor were connected in parallel in the tank circuit of a variable frequency oscillator operated at 950 K.C. The capacity changes in the cell were measured by determining the change in the standard capacitor necessary to restore the frequency of the oscillator to 950 K.C.

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(8) We are indebted to Professor O'Konski for the use of the equipment for measuring the dielectric constants and for assistance in making the measurements.

(9) C. P. Smyth, *Physical Methods of Organic Chemistry*, A. Weissberger, editor, Second Edition, Vol. 1, Part II, Chapter 24, Interscience Publishers, Inc., New York, 1949.