no unreacted fulvene could be isolated. Other attempts to isolate the starting fulvene also were unsuccessful.

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DEPARTMENT OF CHEMISTRY UNIVERSITY OF COLORADO BOULDER, COLO.

# Preparation of o-Phenylenebis(dichloroarsine) and o-Phenylenebis(dimethylarsine)

KENNETH C. EBERLY AND GEORGE E. P. SMITH, JR.

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Kalb<sup>1</sup> failed to convert *o*-benzenediarsonic acid by the action of concentrated hydrochloric acid and sulfur dioxide into o-phenylenebis(dichloroarsine), obtaining 1,3-dichloro-1,3-dihydrobenzofurarsan instead. This observation was also made by Hamilton and Ludeman.<sup>2</sup> Chatt and Mann<sup>3</sup> converted 1,3-dichloro-1,3-dihydrobenzofurarsan into o-phenylenebis(dichloroarsine) by the action of thionyl chloride. However, Goldsworthy and associates<sup>4</sup> converted *p*-benzenediarsonic acid into p-phenylenebis(dichloroarsine) in a single step by the action of alcoholic hydrogen chloride and sulfur dioxide. In our hands, by means of the modified procedures as detailed below, o-phenylenebis-(dichloroarsine) has been prepared in 65% yield in a single step from o-benzenediarsonic acid, and ophenylenebis(dimethylarsine) in 88% yield from the bis(dichloroarsine). The yields obtained are significantly higher by the simplified procedures than those previously reported.

#### EXPERIMENTAL

o-Benzenediarsonic acid. o-Benzenediarsonic acid was prepared by treating diazotized arsanilic acid with sodium arsenite solution according to the method of Kalb.<sup>1</sup> The yield of air-dried acid containing one molecule of water of crystallization (did not melt at 360°) was 56.2%.

o-Phenylenebis(dichloroarsine). A solution of 178.4 g. (0.5187 mole) of the above o-benzenediarsonic acid in 3000 ml. of concentrated hydrochloric acid (37.25%) containing 2.0 g. of potassium iodide was treated with a brisk stream of sulfur dioxide until precipitation ceased (35 min.). The orange-colored solid was removed by filtration and pressed as dry as possible on a sintered glass filter (435.8 g.). The

moist, oily solid was treated with 400 ml. of carbon disulfide whereby some material dissolved, and two liquid layers were obtained. The aqueous layer was separated and discarded. Evaporation of the remaining carbon disulfide solution on a water bath and subsequent cooling yielded crystals. Recrystallization from carbon disulfide yielded colorless crystals of pure o-phenylenebis(dichloroarsine) (123.45 g. or 64.7%) melting at 96°.

Anal. Calcd. for  $C_6H_4As_2Cl_4$ : C, 19.59; H, 1.10; Cl, 38.57. Found: C, 19.44; H, 1.15; Cl, 38.61.

o-Phenylenebis(dimethylarsine). o-Phenylenebis(dimethylarsine) was prepared by a modification of the method of Chatt and Mann.<sup>3</sup> A Grignard solution was prepared in a 3-l. flask from 48.15 g. (1.98 atoms) of magnesium turnings, 281.05 g. (1.98 moles) of dry methyl iodide, and 500 ml. of sodium-dried ether. With rapid stirring and ice cooling in an argon atmosphere, a solution of 121.4 g. (0.330 mole) of o-phenylenebis(dichloroarsine) in 570 ml. of dry ether was added (1 hr.). After stirring at room temperature for another hour, a solution of 413.0 g. of ammonium chloride in 1150 ml. water was added with ice-cooling (40 min.). The organic layer was separated, then the inorganic layer was extracted with 300 ml. of ether and rejected. After drying the combined organic solutions with sodium sulfate under argon, the ether was removed by fractional distillation. The residual liquid was fractionated at reduced pressure, bleeding in argon, collecting the fraction boiling at 153-158° at 20 mm. The yield was 82.75 g. (87.6%) of pale yellow liquid, o-phenylenebis(dimethylarsine), having the following physical properties: f.p.,  $-12^{\circ}$  (sharp); and  $n_{D}^{20}$ , 1.6204. The substance is rapidly oxidized by air, and must be stored under argon.

Chatt and Mann<sup>3</sup> obtained a yield of 26%, and mentioned only the b.p. of the product (156° at 20 mm.).

CHEMICAL AND PHYSICAL LABORATORIES FIRESTONE TIRE AND RUBBER COMPANY AKRON 17, OHIO

# Cyclization of an Unsaturated Benzyl Ether to a Hydrofuran

## E. M. FRY

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The work reported in this communication is the outgrowth of another problem and owes its genesis to the failure of the amino ketone I<sup>1</sup> to react with the methyl Grignard reagent. That this inertness probably is not due to steric factors was made evident by the normal reaction of II with this reagent. Some ketone was always recovered from the reaction mixture however. On refluxing with acetic anhydride-pyridine the tertiary alcohol from II lost the elements of water to give the dihydro compound III, the structural assignment being based on the following consideration: The ultraviolet absorption had too small an extinction coefficient for ring conjugated absorption, eliminating the possibility of a double bond shift. From the results of previous work on the relative stabilities of the endo- vs. the exodouble bond the exo-methylene structure can tenta-

<sup>(1)</sup> L. Kalb, Ann., 423, 39 (1921).

<sup>(2)</sup> C. Hamilton and C. Ludeman, J. Am. Chem. Soc., 52, 3284 (1930).

<sup>(3)</sup> J. Chatt and F. Mann, J. Chem. Soc., 610 (1939).

<sup>(4)</sup> L. Goldsworthy, W. Hook, J. John, S. Plant, J. Rushton, and L. Smith, J. Chem. Soc., 2208 (1948).

<sup>(1)</sup> E. L. May and J. G. Murphy, J. Org. Chem., 20, 257 (1955).

tively be disregarded.<sup>2</sup> Skeletal rearrangement appears ruled out by the isolation of 1,2-dimethylnaphthalene in 20% yield after heating the compound at 275° for 1 hr. And the remaining possibility, involving migration of the side chain was eliminated by converting the compound into IV which had been synthesized by another route.<sup>3</sup> In the present instance compound IV was contaminated by the stereoisomer resulting from hydrogenation of the double bond. Identification was made by comparing the infrared diagrams of the two pure isomers with that of the mixture.

It was hoped that the double bond could be shifted into the 3,4-position. This transformation takes place readily and in high yield with 1,4-dihydronaphthalene,<sup>4</sup> but did not prove feasible in the present case. By the use of sodium amide in liquid ammonia it was possible to get an oil with an enhanced extinction coefficient at 264 m $\mu$  but the increment was not enough to indicate a method of synthetic value. Another example of the reluctance of this system to undergo isomerization was provided by Wenkert and Stevens who obtained VIII from IX in 15% yield after prolonged treat-



ment with hot potassium hydroxide.<sup>5</sup> In both cases the double bond shift would shorten the distance between one of the *gem* groups and the group on the adjacent carbon, and it is possible that resistance to crowding accounts for the experimental results.

Bromine in carbon tetrachloride added rapidly to III giving benzyl bromide and a bromo compound showing strong absorption at 9.3  $\mu$  differing from the broader bands, 9.1–9.3  $\mu$ , of the benzyl ether and characteristic of a tetrahydrofuran structure.<sup>6</sup> By the action of base hydrogen bromide was eliminated and the product showed a maximum absorption in the ultraviolet at 263 m $\mu$ ,  $\epsilon = 6100$ , consistent with its formulation as VI. The unsaturated compound VI absorbed one molar equivalent of hydrogen to give a compound identical with that obtained by the action of hydrogen bromide in glacial acetic acid on the benzyl ether III. Both the reaction with bromine and with hydrogen bromide take place rapidly at room temperature, and indeed there is some evidence that the ring closure can be brought about by the action of hot acetic acid. This follows from the isolation of benzyl alcohol as one of the hydrolysis products from the action of base on the forerun obtained in the preparation of III. The remaining material showed infrared absorption with the sharp band characteristic of the tetrahydrofuran moiety. Hence it is unlikely that intermediate addition to the double bond or ether cleavage steps take place prior to the formation of the product. A concerted reaction such as is shown in figure



X rationalizes the above observations. Lack of carbonyl absorption in the lower boiling by-products indicates that acetic anhydride (X = CH<sub>3</sub>CO, Y-CH<sub>3</sub>COO) does not add to III in this manner, but the possibility of such an addition was not rigorously excluded. Models indicate that the benzene ring imposes a rigidity that makes existence of a *trans* ether ring impossible. If formation of the ring is by the more probable *trans* addition to the double bond<sup>7</sup> then substituents at  $3\alpha$ , 4, and 9b are *cis* to each other.

#### EXPERIMENTAL

1-(2-Benzyloxyethyl)-3, 4-dihydro-2(1H)-naphthalenone.  $\beta$ -Tetralone (46.2 g.) was added within 5 min. to 27.2 g. (1.1 equiv.) of sodium amide in 100 ml. of dry benzene in an externally cooled three-neck flask equipped with stirrer, thermometer, and under a nitrogen atmosphere. Temperature was kept below 50°. Most of the solid dissolved.  $\beta$ -Chloroethyl benzyl ether, 54 g., (1 equiv.), in 50 ml. of benzene was added rapidly and the green solution was brought to reflux and maintained there for 18 hr. A solid enolate separated shortly after the beginning of the heating period. The dark solution was cooled and 150 ml. of 6N HCl was added with stirring. The benzene layer was separated, the solvent removed under reduced pressure, and the remaining oil distilled. The first fraction was collected up to 150° at 20 mm., and from this was recovered 16 g. of the sodium bisulfite adduct of  $\beta$ -tetralone. The second fraction distilled for the most part at 170-180° at 0.3 mm. and weighed 56.1 g. Yield, taking recovery into account, ca. 75% The sample for analysis distilled at 178-180° at 0.5 mm.

Anal. Calcd. for  $C_{19}H_{20}O_2$ : C, 81.40; H, 7.19. Found: C, 81.38; H, 7.26.

1-(2-Benzyloxyethyl)-1-methyl-3,4-dihydro-2(1H)-naphthalenone (II). The above-described compound (56 g.) wasadded neat to 8.6 g. (1.1 equiv.) of sodium amide in 100 ml.of dry benzene in a three-neck flask equipped with stirrer andthermometer. A solid enolate separated. A solution of 31.2g. (1.1 equiv.) methyl iodide in 20 ml. of dry benzene wasthen added and the temperature kept at 40° by occasionalcooling. After 2 hr., water was added, the benzene layerseparated, and the product recovered and distilled. In addition to forerun and residue, 51.1 g. (86%) of oil distillingca. 160-170°/0.4 mm. was obtained. The analyzed sampledistilled at 170-175° at 0.5 mm.

(7) M. S. Newman, Steric Effects in Organic Chemistry, John Wiley and Sons, Inc., New York, N. Y., 1956, p. 242.

<sup>(2)</sup> H. C. Brown, J. H. Brewster, and H. Shechter, J. Am. Chem. Soc., 76, 467 (1954).

<sup>(3)</sup> E. L. May and E. M. Fry, J. Org. Chem., 22, 1366 (1957).

<sup>(4)</sup> W. Hückel and H. Bretschneider, Ann., 540, 157 (1939).

<sup>(5)</sup> E. Wenkert and T. E. Stevens, J. Am. Chem. Soc., 78, 2318 (1956).

<sup>(6)</sup> L. J. Bellamy, The Infrared Spectra of Complex Molecules, John Wiley and Sons, Inc., New York, N. Y., 1954, p. 104.

Anal. Caled. for  $C_{20}H_{22}O_2$ : C, 81.60; H, 7.53. Found: C, 81.56; H, 7.47.

1-(2-Benzyloxyethyl)-2-hydroxy-1,2-dimethyl-1,2,3,4-tetrahydronaphthalene. To 190 ml. (1.2 equiv.) of 1.08N CH<sub>3</sub>MgIin ether was added 50.2 g. of ketone II in an equal volume ofdry ether. The ethereal solution refluxed and it becamecloudy at the end of the addition. It was stirred for 20 min.and the adduct decomposed with dilute acetic acid. Theethereal solution was separated, the solvent removed, andthe product distilled. Aside from a forerun, 50.5 g. distillingat 175-185° at 0.4 mm. was obtained. The analytical sampleboiled at 183°/0.4 mm. The infrared diagram showed car $bonyl absorption at <math>5.82\mu$  in addition to hydroxyl absorption at  $2.95\mu$ .

Anai. Caled. for  $C_{21}H_{26}O_2$ : C, 81.25; H, 8.44. Found: C, 81.41; H, 8.44.

1-(2-Benzyloxyethyl)-1, 2-dimethyl-1, 4-dihydronaphthalene(III). The tertiary alcohol in an equal volume of a 10-1 acetic anhydride-pyridine mixture was refluxed for 3-4 days. The solvent was removed under reduced pressure and the main portion of the product (70-80% of the weight of the starting material) distilled in the range  $145-165^{\circ}$  at ca. 0.3 mm. Infrared absorption at  $5.76\mu$  showed the presence of acetate. Hydrolysis by hot alcoholic sodium hydroxide caused disappearance of this band and revealed the carbonyl band at  $5.83\mu$  known to be present originally. From 25-35% of the material appeared to be the acetoxy compound. The substance was further purified by partial precipitation of the alcoholic and ketonic impurities from a petroleum ether solution by means of CH<sub>3</sub>MgI. The analytical sample distilled at 146-149° at 0.4 mm. Infrared absorption still revealed a little carbonyl impurity.

Anal. Caled. for C<sub>21</sub>H<sub>24</sub>O: C, 86.25; H, 8.27. Found: C, 86.02; H, 8.30.

Alkaline hydrolysis of the forerun material (strong acetate absorption at  $5.77\mu$ ) yielded a fraction distilling at  $105-110^{\circ}$ at 21 mm. and which was identified as benzyl alcohol by means of its phenylisocyanate derivative. The remaining material, an oil distilling from  $90-120^{\circ}$  at 0.3 mm. showed strong absorption at  $9.3\mu$  characteristic of the tetahydrofuran derivatives.

4-Bromo-1,2,3a,4,5,9b-hexahydro-3a,9b-dimethylnaphtho-(2,1-b) furan (V). A carbon tetrachloride solution of 4.4 g. of the benzyl ether III in 5 ml. of carbon tetrachloride was chilled in ice and a solution of bromine (0.149 g./ml.) in carbon tetrachloride was added. Decolorization was immediate and ca. 14.8 ml. (92% of theory) was required to give an apparent end point. The solution was shaken with water and the organic layer removed. Distillation yielded a lachrymatory hquid at 50-90°/0.7 mm., wt. 1.5 g. Its infrared diagram was almost identical with that of an authentic sample of benzyl bromide. The second fraction distilled at ca. 130/0.3 mm., wt. 5.5 g. (84%). The infrared diagram showed strong absorption at 9.3 $\mu$ .

Anal. Caled. for C<sub>14</sub>H<sub>17</sub>BrO; C, 59.80; H, 6.09; Br, 28.42. Found: C, 61.29; H, 6.03; Br, 27.21.

1.8.3a,9b-Tetrahydro-3a,9b-dimethylnaphtho(2,1-b)furan (VI). An alcohol solution of 1.8 g. of the bromohydrofuran V with excess methyl amine in a sealed tube was heated on the steam bath for 1 kr. The solvent was distilled, ether added, solid amine salt filtered and the product distilled at 88–92°/ 0.8 mm., wt. 1.95 g., ultraviolet maximum in ethanol at 263 m. ( $\epsilon$  6300), infrared ether band at 9.3 $\mu$ .

Anc. Caled. for CitHigO: C, 83.96; H, 8.05. Found: C, 81.66; H. 8.34.

1,2,3a,4,5,9b-Hexakydro-3a,9b-dimethylnaphtho(2,1-b)furan (V11). A. Hydrogenation of 1.8 g, of VI in alcohol solution with Adams' catalyst at room temperature was rapid and ceased after absorption of 1 mol. The product distilled at  $85-88^{\circ}/0.2$  mm., wt. 1.5 g.

B. To S.O.g., of the benzy' ether III (I.R. showed ketonic impurity) was added 6.8 int. of HBe-HOAc (2.4N HBr). The temperature rest from 24-83°. After 15 min, the solvent was removed under reduced pressure and the first fraction collected from 90-100° at 18 mm. From the infrared diagram it appeared to be a mixture of benzyl bromide and benzyl acetate. The main fraction distilled at 90-97° at 0.5 mm., wt. 1.25 g. Its infrared diagram was identical with that of the compound described under A, showing ether absorption at  $9.3\mu$  and being negative in the hydroxyl region.

Anal. Calcd. for C<sub>14</sub>H<sub>18</sub>O: C, 83.12; H, 8.97. Found: C, 82.90; H, 8.99.

1-(2-Hydroxyethyl)-1,2-dimethyl-1,2,3,4-tetrahydronaphthalene. Catalytic reduction of 7.3 g. of III followed by hydrolysis of the benzyl group by aqueous HBr-HOAc gavethe alcohol contaminated by acetate. Hydrolysis with alcoholic sodium hydroxide gave the alcohol. It distilled at 113-116°/0.2 mm., wt. 3.1 g.

Anal. Caled. for  $C_{14}H_{20}O$ : C, 82.30; H, 9.87. Found: C, 82.15; H, 9.81.

1-(2-Dimethylaminoethyl)-1,2-dimethyl-1,2,3,4-tetrahydronaphthalene. The above-described alcohol in 3.0 g., chloroform solution reacted with thionyl chloride to give ca. 50%yield of crude chloride. This material in benzene solutionwith excess dimethyl amine in a sealed tube was heated onthe steam bath for 4 days. After distilling the solvent, theoil was triturated with dilute sodium hydroxide solution,then dissolved in ether. The solvent was distilled from thissolution and the residue dissolved for the most part in a smallexcess of <math>2N HCl, the insoluble oil being removed with ether. The acid solution was concentrated to an oil under reduced



pressure whereupon it crystallized. Washed with ethyl acetate it weighed 0.18 g. Purified from ethyl acetate it melted at 198–199.5°. Its nonidentity with an isomer<sup>3</sup> m.p. 202– 203.5°, prepared by another synthesis was established by a mixture melting point. Impure material, m.p. 166–173°, from the mother liquors of recrystallization gave an infrared diagram which was a composite of the diagrams of the two pure isomers.

Anal. Caled. for C16H26CIN: C, 71.75; H, 9.78. Found: C, 71.71; H, 9.87.

Pyrolysis of 1-(2-benzyloxyethyl)-1,2-dimethyl-1,4-dihydronaphthalene (111) was achieved by heating 0.5 g. in a test tube at 275-287° for 1 hr. The oil was dissolved in an alcohol solution containing 0.2 g. of pieric acid and a crystalline pierate was obtained. Purified from alcohol it weighed 0.14 g. (21%), m.p. 115-130°. Another crystallization gave a melting point of 128-131°, undepressed by admixture with authentic 1,2-dihydronaphthalene picrate.

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LABORATORY OF CHEMISTRY NATIONAL INSTITUTES OF HEALTH BETHESDA 14, MD.

# Organic Deuterium Compounds. I. 1,2-Dimethoxy-d<sub>f</sub>-ethane-d<sub>4</sub> and 2-Methoxy-d<sub>3</sub>-ethan-d<sub>4</sub>-ol<sup>1</sup>

EUGENE R. BISSELL AND ROBERT E. SPENGER

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In connection with some nuclear magnetic resonance studies being conducted in this laboratory, a need arose for a material with the solvent properties of ethylene glycol dimethyl ether (1,2-dimethoxyethane) which would, however, show little or no nuclear magnetic resonance signal in the proton region. Since a completely deuterated ethylene glycol dimethyl ether would meet these requirements, we undertook its synthesis starting with deuterium oxide and a sample of ethylene-d<sub>4</sub> available to us.

Ethylene-d<sub>4</sub> was converted via its dibromide to ethylene-d<sub>4</sub> glycol diacetate which was then hydrolized to the free glycol.<sup>2</sup> The glycol thus obtained was converted via its monosodium derivative to a mixture of 1,2-dimethoxy-d<sub>5</sub>-ethane-d<sub>4</sub>, 2-methoxyd<sub>3</sub>-ethan-d<sub>4</sub>-ol, and unchanged glycol by treatment with methyl-d<sub>3</sub> bromide. The 2-methoxy-d<sub>3</sub>-glycol could be further methylated to the diether. The methyl-d<sub>3</sub> bromide was prepared from carbon suboxide<sup>3,4</sup> via malonic-d<sub>2</sub> acid-d<sub>2</sub> and acetic-d<sub>3</sub> acid-d.<sup>5</sup> The procedure of Nolin and Leitch<sup>6</sup> for the Hunsdiecker degradation was modified to eliminate the somewhat hazardous sealed tube operation.

Comparison of the nuclear magnetic resonance spectrum in the proton region of the 1,2-dimethoxy $d_6$ -ethane- $d_4$ , obtained by this method, with that of normal material indicated that the isotopic purity of our deuterated material was at least 98.6 and probably better than 99 atom per cent deuterium. The isotopic purity of the deuterium oxide used was 99.6% and that of the ethylene-d<sub>4</sub> better than 99%.

#### EXPERIMENTAL<sup>7</sup>

Ethylene-d<sub>4</sub> glycol. Ethylene-d<sub>4</sub> was brominated in carbon tetrachloride solution at 0°. Distillation yielded 47.8 g. of dibromide b.p. 128-131°,  $n_D^{20}$  1.53367 (Lit.<sup>8</sup> b.p. 129.5°,  $n_D^{20}$  1.5360). The dibromide was converted in 78.5% yield to ethylene-d<sub>4</sub> glycol by the procedure of Bannard, Morse, and Leitch.<sup>2</sup> B.p. 113-115°/12 mm.,  $n_D^{24}$  1.42831 (Lit.<sup>2</sup> b.p. 86-87°/8 mm.,  $n_D^{20}$  1.4293).

Acetic- $d_3$  acid-d. Diacetyltartaric anhydride<sup>3</sup> was pyrolyzed according to the procedure of Hurd and Pilgrim<sup>4</sup> to afford an average yield of 42% of carbon suboxide, b.p. 7-8°. This was converted via malonic- $d_2$  acid- $d_2$  to acetic- $d_3$  acid-d in 89-97% yield by the method of Wilson.<sup>5</sup>  $d^{25}$  1.137,  $n_D^{25-2}$  1.3875.

Methyl-d<sub>3</sub> bromide. Silver acetate-d<sub>3</sub>, prepared from aceticd<sub>3</sub> acid-d by the method of Nolin and Leitch,<sup>6</sup> was subjected to the Hunsdiecker degradation in an apparatus (Fig. 1)



consisting of a trap (A) connected to a vacuum system, a mercury bubbler (B) containing about 0.5 inch of mercury, and a reaction tube (C) charged with 20-40 g. of silver acetate-d<sub>3</sub>. Attached to the bottom of the reaction tube was a bromine reservoir (D) containing about 25 g. of 8-mesh Drierite and bromine in about 2% excess over that required to consume all of the silver acetate-d<sub>3</sub> in C.

After assembly of the apparatus with the center tube in the bubbler (B) raised with a magnet from the outside, the bromine reservoir was cooled to  $-78^{\circ}$ , and the system was evacuated. The trap (A) was cooled in liquid nitrogen, and argon was admitted to the system to a pressure of 150 mm. The argon served to control the rate of flow of bromine vapor through the silver acetate-d, bed. The center tube in B was lowered and the bromine reservoir allowed to warm to room temperature. During the first hour or so of the reaction pe-

(8) L. C. Leitch and A. T. Morse, Can. J. Chem., 30, 924 (1952).

<sup>(1)</sup> This work was performed under the auspices of the U. S. Atomic Energy Commission, Contract No. W-7405-eng-48.

<sup>(2)</sup> R. A. Bannard, A. T. Morse, and L. C. Leitch, Can. J. Chem., 31, 351 (1953).

<sup>(3)</sup> A. Wohl and C. Oesterlin, Ber., 34, 1139 (1901).
(4) C. D. Hurd and F. D. Pilgrim, J. Am. Chem. Soc., 55, 757 (1933).

<sup>(5)</sup> C. L. Wilson, J. Chem. Soc., 1, 492 (1935).

<sup>(6)</sup> B. Nolin and L. C. Leitch, Can. J. Chem., 31, 153 (1953).

<sup>(7)</sup> Boiling points are uncorrected.