

**257. *The Electrolysis of Some Aryl-substituted Aliphatic Acids.***

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The electrolyses of  $\delta$ -phenylvaleric, *o*-biphenylacetic, and  $\beta$ -*o*-biphenylpropionic acid are reported.  $\delta$ -Phenylvaleric acid gave 1,8-diphenyloctane, *n*-butylbenzene, methyl  $\delta$ -phenylvalerate, and the product of intramolecular cyclisation, tetralin. *o*-Biphenylacetic acid gave mainly 2,2'-diphenylbibenzyl and 2-phenylbenzyl methyl ether, but no fluorene.  $\beta$ -*o*-Biphenylpropionic acid gave the product of intramolecular cyclisation, 9,10-dihydrophenanthrene, in relatively good yield.

HOMOLYTIC alkylation of aromatic compounds is not so readily accomplished as homolytic arylation, and relatively few reactions of this type have been submitted to detailed study. The alkylation reactions are, in general, more complex and yields are usually low. Aromatic methylation has been reported with acetyl peroxide,<sup>1,2</sup> lead tetra-acetate,<sup>1,3</sup> phenyl iodosoacetate,<sup>4</sup> and *t*-butyl peroxide.<sup>5</sup> Polynitro-compounds have been methylated by means of electrolysis with sodium acetate and acetic acid.<sup>1</sup> An example of intramolecular alkylation has been reported by DeTar and Weis,<sup>6</sup> who obtained tetralin in 28% yield by the thermal decomposition of  $\delta$ -phenylvaleryl peroxide in benzene solution, together with some 1,8-diphenyloctane. No such cyclisation was observed in carbon tetrachloride solution. The electrolysis of  $\delta$ -phenylvaleric acid in methanol-pyridine has

<sup>1</sup> Fieser, Clapp, and Daudt, *J. Amer. Chem. Soc.*, 1942, **64**, 2052.

<sup>2</sup> Fieser and Oxford, *J. Amer. Chem. Soc.*, 1942, **64**, 2060; Eliel, Rabindran, and Wilen, *J. Org. Chem.*, 1957, **22**, 859.

<sup>3</sup> Fieser and Chang, *J. Amer. Chem. Soc.*, 1942, **64**, 2043.

<sup>4</sup> Sandin and McCormack, *J. Amer. Chem. Soc.*, 1945, **67**, 2051.

<sup>5</sup> Beckwith and Waters, *J.*, 1956, 1108; 1957, 1665; Cowley, Norman, and Waters, *J.*, 1959, 1799.

<sup>6</sup> DeTar and Weis, *J. Amer. Chem. Soc.*, 1956, **78**, 4296; 1957, **79**, 3041.

also been reported,<sup>7</sup> but surprisingly no tetralin was obtained and the only product isolated was 1,8-diphenyloctane.

The electrolysis of  $\delta$ -phenylvaleric acid has now been reinvestigated both in methanol and in methanol-pyridine solution, and the products obtained are summarised in Table 1.

TABLE 1.

Yields of products obtained in the electrolysis of  $\delta$ -phenylvaleric acid.

Product	In cold methanol	In hot methanol-pyridine
1,8-Diphenyloctane (%) .....	8.8	46.3
Methyl $\delta$ -phenylvalerate (%) .....	31.3	1.4
n-Butylbenzene (%) .....	8.8	11.5
Tetralin (%) .....	3.8	4.5

The yield of the Kolbe dimeride (1,8-diphenyloctane) obtained in the electrolysis in methanol-pyridine is very similar to that reported by the earlier workers,<sup>7</sup> who however reported no other products. The lower yield of 1,8-diphenyloctane and the higher yield of ester obtained in the electrolysis in methanol is attributed both to the lower temperature and the lower current density used. Under these conditions the acyloxy-radical liberated at the anode is more stable and attack on the solvent takes place in preference to loss of carbon dioxide and dimerisation. The yield of tetralin is significantly less than that which results from the decomposition of  $\delta$ -phenylvaleryl peroxide.<sup>6</sup>

Electrolyses were also carried out with *o*-biphenylacetic acid and the results are summarised in Table 2. In this case no trace of the product of intramolecular alkylation, fluorene, was found. The main product under both experimental conditions was the

TABLE 2.

Yields of products obtained in the electrolysis of *o*-biphenylacetic acid.

Product	In cold methanol	In hot methanol-pyridine
2,2'-Diphenylbibenzyl (%) .....	38.8	39.0
2-Phenylbenzyl methyl ether (%) ...	34.4	16.2
2-Phenylbenzyl alcohol (%) .....	Nil	6.1
Methyl <i>o</i> -biphenylacetate (%) .....	Nil	6.5

normal Kolbe dimeride, 2,2'-diphenylbibenzyl. This is in agreement with the well-established property of relatively stable benzyl-type radicals. Phenylacetic acid is known to give bibenzyl in 50% yield on electrolysis<sup>8</sup> and diphenylacetic acid gives diphenylmethyl methyl ether, in addition to tetraphenylethane and other products.<sup>9</sup>

From the electrolysis of  $\beta$ -*o*-biphenylpropionic acid in hot methanol-pyridine the only fully identified product was 9,10-dihydrophenanthrene, the product of intramolecular alkylation, which was obtained in 37% yield. This relatively high yield and the absence of 2-ethylbiphenyl and 2-phenylstyrene (the products of disproportionation of the intermediate radical) are noteworthy features of this reaction. Some indication was obtained of the formation of a small quantity of the Kolbe dimeride, 1,4-di-*o*-biphenylbutane, but this was not proved.

#### EXPERIMENTAL

*Reagents.*— $\delta$ -Phenylvaleric acid, prepared by Plati, Strain, and Warren's method<sup>10</sup> from  $\gamma$ -phenyl-n-propyl bromide and ethyl malonate, was obtained in plates, m. p. 58–59°, from light petroleum (b. p. 60–80°). *o*-Biphenylacetic acid, prepared by Ott and Smith's

<sup>7</sup> Evans and Whalley, *J.*, 1954, 3642.

<sup>8</sup> Fichter and Stenzl, *Helv. Chim. Acta*, 1939, **22**, 976; Linstead, Shephard, and Weedon, *J.*, 1952, 3624.

<sup>9</sup> van de Hoek and Nauta, *Rec. Trav. chim.*, 1942, **61**, 845.

<sup>10</sup> Plati, Strain, and Warren, *J. Amer. Chem. Soc.*, 1943, **65**, 1273.

method,<sup>11</sup> separated from benzene–light petroleum (b. p. 60–80°) in prisms, m. p. 114–115°.  $\beta$ -*o*-Biphenylpropionic acid was prepared by a modification of Wallingford, Thorpe, and Homeyer's method:<sup>12</sup> sodium (2.3 g.) was dissolved in ethanol (100 ml.) and after removal of the excess of ethanol under reduced pressure the powdered sodium ethoxide was dissolved in a mixture of diethyl malonate (16 g.) and diethyl carbonate (75 g.); after removal of ethanol 2-phenylbenzyl chloride<sup>11</sup> (20.5 g.) was added slowly with stirring and heating on a water-bath; after being heated at 100° for 6 hr. the mixture was poured into water (150 ml.), neutralised with acetic acid, and extracted with ether (10 × 50 ml.); the ethereal extract was washed, dried (MgSO<sub>4</sub>), and distilled; the fraction collected at 160°/0.05 mm. was boiled under reflux with a solution of potassium hydroxide (20 g.) in water (20 ml.) and ethanol (50 ml.); after removal of the ethanol under reduced pressure and acidification with hydrochloric acid, the precipitated red solid was crystallised from aqueous ethanol (charcoal) and then from benzene–light petroleum (b. p. 60–80°).  $\beta$ -*o*-Biphenylpropionic acid (15 g.) was obtained with m. p. 129–130° (Found: C, 79.5; H, 6.2. Calc. for C<sub>15</sub>H<sub>14</sub>O<sub>2</sub>: C, 79.6; H, 6.2%) (von Braun and Manz<sup>13</sup> reported m. p. 125°).

Methyl  $\delta$ -phenylvalerate, prepared by the Fischer–Speier method, had b. p. 68°/0.01 mm. Methyl *o*-biphenylacetate, prepared in similar manner, had b. p. 92°/0.02 mm. (Found: C, 79.4; H, 6.5. C<sub>15</sub>H<sub>14</sub>O<sub>2</sub> requires C, 79.6; H, 6.2%).

*Methyl 2-Phenylbenzyl Ether*.—2-Phenylbenzyl chloride (2 g.) was boiled under reflux for 3 hr. with methanol (10 ml.) containing sodium (0.46 g.). The mixture was poured into water and extracted with methylene chloride (5 × 20 ml.). Distillation of the washed and dried (MgSO<sub>4</sub>) extract gave methyl 2-phenylbenzyl ether (1.8 g.), b. p. 148°/11 mm. (Found: C, 84.2; H, 7.0. C<sub>14</sub>H<sub>14</sub>O requires C, 84.8; H, 7.1%).

*2-Phenylbenzyl Alcohol*.—2-Phenylbenzyl chloride (5 g.) was added slowly to magnesium turnings (0.625 g.) and a crystal of iodine in dry ether (20 ml.). After the reaction had ceased, the mixture was boiled under reflux for 1 hr. and then oxygen was passed into it for 2 hr. The mixture was poured on dilute sulphuric acid (25 ml.) and ice (25 g.), and the product was extracted with ether (3 × 20 ml.). Distillation of the washed and dried extract gave slightly impure 2-phenylbenzyl alcohol (3.5 g.) as a very pale green oil, b. p. 96°/0.04 mm. (Found: C, 86.5; H, 6.3. Calc. for C<sub>13</sub>H<sub>12</sub>O: C, 84.75; H, 6.6%). The 3,5-dinitrobenzoate, prepared in the normal manner, separated from ethanol in needles, m. p. 134–135° (Found: C, 63.7; H, 4.0. C<sub>20</sub>H<sub>14</sub>N<sub>2</sub>O<sub>6</sub> requires C, 63.5; H, 3.7%).

2,2'-Diphenylbibenzyl was reported by von Braun and Manz<sup>13</sup> as a colourless oil, which they prepared by a Wurtz reaction with 2-phenylbenzyl bromide. This reaction has been adapted as follows: Sodium (2 g.) was added to 2-phenylbenzyl chloride (5.05 g.) in dry cyclohexane (20 ml.), and the mixture was boiled under reflux for 12 hr. After the addition of ethanol (25 ml.), refluxing was continued for a further hour. The mixture was poured into water (200 ml.), and the cyclohexane layer was separated, washed, and dried (MgSO<sub>4</sub>). After removal of the cyclohexane the residue was crystallised from light petroleum (b. p. 80–100°), to give 2,2'-diphenylbibenzyl (0.9 g.) in needles, m. p. 118–119° (Found: C, 93.5; H, 6.7. C<sub>26</sub>H<sub>22</sub> requires C, 93.4; H, 6.6%).

*Apparatus*.—The apparatus used for the electrolyses was that previously described.<sup>14</sup>

*Electrolysis of  $\delta$ -Phenylvaleric Acid in Methanol*.—The acid (17.82 g., 0.1 mole) in methanol (280 ml.) containing sodium (0.23 g.; 0.01 g.-atom) was electrolysed, the cell being immersed in ice–water. Deposition of a thin layer of a colourless gum on the electrodes made it impossible to maintain a current greater than 0.1 amp. A current of 0.04–0.06 amp. was maintained at 120 v for 48 hr. with occasional reversal of the electrodes. After each 8 hr. period the electrolysis was stopped and the electrodes were cleaned mechanically. The internal temperature was 5–8° and the pH rose from 5.0 to 6.5. The solution remained colourless. Filtration of the solution gave a gum, which was dried at 70° (0.51 g.). Methanol was removed from the filtrate by distillation under reduced pressure. The distillate gave only a slight positive test for formaldehyde with dimedone. To the residual straw-coloured oil were added ether (200 ml.) and water (100 ml.). The ethereal layer was separated and shaken with saturated aqueous sodium hydrogen carbonate (6 × 50 ml.), then with water, and dried (MgSO<sub>4</sub>). The combined

<sup>11</sup> Ott and Smith, *J. Amer. Chem. Soc.*, 1955, **77**, 2325.

<sup>12</sup> Wallingford, Thorpe, and Homeyer, *J. Amer. Chem. Soc.*, 1942, **64**, 580.

<sup>13</sup> von Braun and Manz, *Annalen*, 1929, **468**, 258.

<sup>14</sup> Bunyan and Hey, *J.*, 1962, 324.

aqueous layer and bicarbonate washings were acidified with hydrochloric acid, and the precipitated acid was extracted with chloroform ( $4 \times 50$  ml.), washed, and dried ( $\text{MgSO}_4$ ). Removal of the chloroform under reduced pressure gave  $\delta$ -phenylvaleric acid (12.5 g.), m. p. and mixed m. p. 56—59°. Removal of solvent under reduced pressure from the ethereal solution containing neutral products gave a straw-coloured oil, which was fractionally micro-distilled, to give the following colourless liquid fractions: (i) b. p. 82°/11 mm. (0.50 g.); (ii) b. p. 68°/0.01 mm. (1.79 g.); (iii) b. p. 124°/0.01 mm. (0.59 g.). The residue weighed 0.54 g. The infrared spectrum of fraction (i) showed strong peaks at 744 and 698  $\text{cm}^{-1}$ , which were identified as those of *n*-butylbenzene (744 and 698  $\text{cm}^{-1}$ ) and tetralin (745  $\text{cm}^{-1}$ ). By means of gas-liquid chromatography and by comparison with a mixture of these two hydrocarbons, fraction (i) was shown to consist of a mixture of tetralin (0.15 g.) and *n*-butylbenzene (0.35 g.). Fraction (ii) was identified as methyl  $\delta$ -phenylvalerate (infrared spectrum), and hydrolysis with 2*N*-aqueous potassium hydroxide for 18 hr. gave  $\delta$ -phenylvaleric acid, m. p. and mixed m. p. 56—59° (1.47 g.). Fraction (iii) was subjected to column chromatography (Woelm neutral alumina;  $15 \times 1.5$  cm.). Elution with hexane (500 ml.) gave 1,8-diphenyloctane (0.35 g.) as a colourless oil, identified by comparison of its infrared spectrum with that of an authentic specimen (see below).

*Electrolysis of o-Biphenylacetic Acid in Methanol.*—The acid (21.22 g.; 0.1 mole) was electrolysed in methanol containing sodium, as described in the previous experiment. A current of 1 amp. was maintained initially at 72 v, but as a white solid collected on the electrodes the voltage had to be increased to 120. By reversal of the electrodes a current of 0.9—1.1 amp. was maintained for 2 hr. but thereafter it slowly dropped to 0.25—0.45 amp. after a further 2 hr., at which level it was kept for an additional 2 hr. The internal temperature dropped from 22° to 15° and the pH rose to 6.5. The solution remained colourless, but some solid separated. The products from this experiment were worked up as described for the previous electrolysis. Filtration gave a solid (0.77 g.), and the distilled methanol contained formaldehyde, which gave a copious precipitate with dimedone. The recovered acid, m. p. and mixed m. p. 111—115°, weighed 8.27 g. Distillation of the neutral material gave fractions: (i) b. p. 150°/11 mm., an oil (4.16 g.); (ii) b. p. 184—186°/0.02 mm., an oil which solidified on ice (3.96 g.); (iii) b. p. 226—240°/0.02 mm., an orange viscous oil (0.85 g.). The residue weighed 1.41 g.

Fraction (i) showed a strong peak at 1093  $\text{cm}^{-1}$  in its infrared spectrum and was identified as methyl 2-phenylbenzyl ether containing a trace of ester (probably methyl *o*-biphenylacetate), which was removed on hydrolysis. Fraction (ii) on recrystallisation from light petroleum (b. p. 80—100°) gave 2,2'-diphenylbibenzyl in needles, m. p. and mixed m. p. 118—119°, the infrared spectrum being identical with that of an authentic specimen.

*Electrolysis of  $\delta$ -Phenylvaleric Acid in Methanol-Pyridine.*—Methanol (44 ml.) containing sodium (0.23 g., 0.01 g.-atom) was added to a solution of  $\delta$ -phenylvaleric acid (17.85 g., 0.1 mole) in pyridine (231 ml.) in a thermostat-bath at 93°. The clear solution was electrolysed with a constant potential of 120 v. The current, initially 0.2 amp., dropped rapidly to 0.15 amp. and then steadily to 0.05 amp. during 30 hr. The solution became pale brown and no solid was deposited. The contents of the cell were washed out with methanol (75 ml.), and the methanol and pyridine were removed by distillation. The residue was extracted with methylene chloride (500 ml.) and with water (250 ml.). The methylene chloride solution was washed successively with 20% hydrochloric acid ( $2 \times 100$  ml.), water ( $2 \times 50$  ml.), saturated aqueous sodium hydrogen carbonate ( $6 \times 50$  ml.), and water ( $2 \times 50$  ml.), and then dried ( $\text{MgSO}_4$ ). The combined aqueous layer and bicarbonate extract was acidified with hydrochloric acid, and the precipitated acid was extracted with ether ( $5 \times 20$  ml.) and dried. Removal of the ether under reduced pressure left  $\delta$ -phenylvaleric acid, m. p. and mixed m. p. 51—57° (0.95 g.). Methylene chloride was removed by distillation from the solution of neutral products, followed by a small quantity of pyridine (0.95 g.) under reduced pressure. The residual brown oil was distilled and the following liquid fractions were collected: (i) b. p. 78—82°/12 mm. (2.07 g.); (ii) b. p. 122—126°/0.01 mm. (7.85 g.). The residue weighed 2.56 g. Fraction (i) was shown by infrared spectroscopy and gas-liquid chromatography to consist of a mixture of tetralin (0.56 g.) and *n*-butylbenzene (1.51 g.) (strong peaks at 744 and 698  $\text{cm}^{-1}$ ). No ester peak was observed in this experiment. Fraction (ii) gave an infrared spectrum similar to that of fraction (i), but showed a peak at 1745  $\text{cm}^{-1}$  (ester). Chromatography on Woelm neutral alumina gave on elution with *n*-hexane 1,8-diphenyloctane (5.84 g.) as a colourless oil, b. p. 124—126°/0.01 mm.

(Found: C, 90.4; H, 9.8. Calc. for  $C_{20}H_{26}$ : C, 90.2; H, 9.8%). Further elution with methanol gave a pale brown oil (1.33 g.), which on hydrolysis with boiling 2*N*-aqueous-ethanolic potassium hydroxide for 18 hr. gave  $\delta$ -phenylvaleric acid (0.23 g.), m. p. and mixed m. p. 50—57°, and unchanged material (1.08 g.) which showed no ester peak.

*Electrolysis of o-Biphenylacetic Acid in Methanol-Pyridine.*—Methanol (44 ml.) containing sodium (0.23 g., 0.01 g.-atom) was added to a solution of *o*-biphenylacetic acid (21.22 g., 0.1 mole) in pyridine (231 ml.) at 93° and electrolysis was carried out with a constant potential of 120 v. The initial current of 0.16 amp. rose steadily to 0.25 amp. in 24 hr. before dropping sharply to 0.04 amp., whereafter the electrolysis was terminated. The solution slowly developed a red colour and a small white solid deposit separated (0.21 g. after being dried at 70°). The bulk of the solvent was removed by distillation, as described above. No formaldehyde was detected in the lower-boiling fraction. The solid residue was extracted with boiling ether (5 × 200 ml.) and the extract was washed with 2*N*-aqueous sodium hydroxide (3 × 40 ml.) and with water and dried ( $MgSO_4$ ). Acidification of the alkaline solution gave no recovered acid. The ether-insoluble material weighed 0.74 g.

The neutral ethereal extract was fractionated and after the distillation of residual pyridine (1.5 g.) at 85°/25 mm. the following fractions were collected: (i) an oil, b. p. 150°/10 mm. (3.21 g.); (ii) a pale green oil, b. p. 92—98°/0.02 mm. (2.61 g.); (iii) a solid, b. p. 194—196°/0.03 mm. (8.82 g.); (iv) a red gum, b. p. ca. 250°/0.03 mm. (0.77 g.). The residue weighed 2.15 g.

Fraction (i) was shown by gas-liquid chromatography and its infrared spectrum to consist only of methyl 2-phenylbenzyl ether (peak at 1093  $cm^{-1}$ ). Fraction (ii) showed an ester peak at 1736  $cm^{-1}$  and a hydroxyl peak at 3413  $cm^{-1}$  in its infrared spectrum. From the product of the hydrolysis with 4*N*-aqueous-ethanolic potassium hydroxide 2-phenylbenzyl alcohol (1.12 g.) (3,5-dinitrobenzoate, m. p. and mixed m. p. 134—135°) and *o*-biphenylacetic acid (1.03 g.), m. p. and mixed m. p. 110—114°, were isolated. The latter acid is derived from the hydrolysis of its methyl ester, and a mixture of this ester with 2-phenylbenzyl alcohol gave an infrared spectrum indistinguishable from that of fraction (ii). Fraction (iii) on recrystallisation from light petroleum (b. p. 80—100°) gave 2,2'-diphenylbibenzyl (6.5 g.) in needles, m. p. 118—119°, undepressed on admixture with an authentic specimen.

*Electrolysis of  $\beta$ -o-Biphenylpropionic Acid in Methanol-Pyridine.*—Methanol (44 ml.) containing sodium (0.23 g., 0.01 g.-atom) was added to a solution of  $\beta$ -*o*-biphenylpropionic acid (22.63 g., 0.1 mole) in pyridine (231 ml.) at 93°. A white gel-like material was deposited which slowly dispersed during the electrolysis. A constant potential of 120 v was applied for 24 hr. The current, initially 0.4 amp., dropped to 0.03 amp. and then rose readily to 0.16 amp. after 20 hr. The electrolysis was terminated after 30 hr. The formation of the gel prevented a strong flow of the electrolyte in the cell. No solid was deposited. The bulk of the solvent was removed as described above and no formaldehyde was detected in the lower-boiling fractions. The solid residue was extracted with boiling ether (5 × 200 ml.), which left a residue (A). The ethereal extract was washed with 2*N*-aqueous sodium hydroxide (5 × 20 ml.), then with water, and dried ( $MgSO_4$ ). The ether-insoluble residue (A) was added to the alkaline extract. Filtration gave an amorphous brown solid (1.72 g.) and acidification of the filtrate gave  $\beta$ -*o*-biphenylpropionic acid (1.27 g.), m. p. and mixed m. p. 105—115°. Evaporation of the ether from the neutral extract followed by pyridine (1.75 g.), which was collected at 80°/25 mm., gave a residue which on distillation gave the following liquid fractions: (i) b. p. 92—98°/0.03 mm. (9.96 g.) and (ii) b. p. 210°/0.03 mm. (2.90 g.). The residue weighed 1.30 g. Fraction (i) was shown not to contain 2-ethylbiphenyl or 2-vinylbiphenyl<sup>15</sup> by comparison with authentic specimens when submitted to gas-liquid chromatography and by their infrared spectra. Chromatography of a portion (1.34 g.) on an active alumina column (30 cm. × 2.5 cm.) gave: (a) with light petroleum (b. p. 60—80°) (250 ml.) a colourless oil (0.23 g.), which appeared to be unsaturated but was not identified (Found: C, 90.2; H, 7.1%); (b) with light petroleum (b. p. 60—80°) (750 ml.) a solid (0.87 g.), which on crystallisation from methanol gave 9,10-dihydrophenanthrene in needles, m. p. and mixed m. p. 33—34° (Found: C, 93.1; H, 6.8. Calc. for  $C_{14}H_{12}$ : C, 93.3; H, 6.7%) (the infrared spectra of the two specimens were identical); and (c) with benzene (500 ml.), a pale yellow solid (0.04 g.), which was not identified.

Fraction (ii) was submitted to chromatography on an alumina column (30 × 2.5 cm.), and all hydrocarbons were eluted with light petroleum (b. p. 60—80°). The resulting oil (1.20 g.)

<sup>15</sup> Bradsher and Wert, *J. Amer. Chem. Soc.*, 1940, **62**, 2806.

had b. p.  $202^{\circ}/0.03$  mm. and did not solidify. The infrared spectrum was similar to that of 2-ethylbiphenyl and this evidence, together with the boiling point, suggests that it is impure 1,4-di-*o*-biphenylbutane (Found: C, 91.0; H, 7.8%; *M*, 305. Calc. for  $C_{28}H_{28}$ : C, 92.8; H, 7.2%; *M*, 362).

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