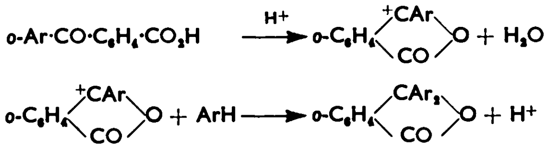
327. *Acylation and Allied Reactions Catalysed by Strong Acids. Part XVII.* Perchloric Acid as a Catalyst for the Formation of Phthalides from* o-*Aroylbenzoic Acids.*

By H. Burton and D. A. Munday.

In view of reports that silver perchlorate can catalyse the formation of phthalides from *o*-benzoylbenzoic acid and an aromatic component, this use has been investigated. Both anhydrous and 72% perchloric acid are effective, even at room temperature. Related experiments with phthaloyl chloride and succinyl chloride and silver perchlorate in, *e.g.*, anisole are described.

Cauquil, Barrera, and Barrera [1,2,3] recently described the production of phthalides from *o*-benzoylbenzoic acid and a suitable aromatic compound using silver perchlorate as a catalyst. However, in view of previous reports [4] of the alleged catalytic activity of silver perchlorate, and the fact that Cauquil, Barrera, and Barrera state [3] that they made no attempt to remove perchloric acid from their silver perchlorate, in which it is a common contaminant, we decided to see whether the true catalyst might not be perchloric acid, in which case the reactions would undoubtedly proceed as follows :



The regeneration of the proton in the second stage would account for the activity of the perchloric acid as a catalyst.

As was reported briefly,[5] we have found that *o*-benzoylbenzoic acid (1 mole) when refluxed for 4 hr. in *m*-xylene with anhydrous perchloric acid (0·004 mole) in acetic acid gave 3-phenyl-3-(2 : 4-dimethylphenyl)phthalide in 64% yield. Cauquil and her collaborators obtained *ca.* 50% of the phthalide using *two* molecules of silver perchlorate per molecule of acid and refluxing the solution for 7 hr. When we performed an experiment similar to the latter but with equimolecular quantities of acid and silver perchlorate we obtained only 6·8% of phthalide after 4 hr. The silver perchlorate which we used had been dried by our usual method,[6] *i.e.*, as a fine powder in a vacuum-desiccator containing both phosphoric oxide and potassium hydroxide pellets. This procedure has been shown to remove most, but not always all, of the perchloric acid from the perchlorate. However, the discrepancy in the yields of phthalide obtained by the French workers and ourselves using silver perchlorate, coupled with our demonstration that under similar conditions perchloric acid is, on a molecular basis, roughly two-thousand times more efficient than silver perchlorate, strongly suggests that perchloric acid is the true catalyst. We obtained further support for this view by treating the silver perchlorate with a little silver acetate before addition of the keto-acid. In one experiment the yield of phthalide was reduced to 0·7% after a total of 14 hr. when the silver perchlorate had been first refluxed with silver acetate (0·002 mole) for 2 hr.

At room temperature the reaction with *m*-xylene proceeds slowly, only 1·3% of 3-phenyl-3-(2 : 4-dimethylphenyl)phthalide being produced in 14 days from similar amounts

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* Part XVI, preceding paper.

[1] Cauquil, Barrera, and Barrera, *Bull. Soc. chim. France*, 1951, **18**, 173.

[2] *Idem, ibid.*, 1952, **19**, 248.

[3] *Idem, ibid.*, 1955, **22**, 550.

[4] Burton and Praill, *J.*, 1953, 837.

[5] Burton and Munday, *Chem. and Ind.*, 1956, 316.

[6] Burton and Praill, *J.*, 1950, 2034.

of the acids as were used above; equivalent quantities of the keto-acid and anhydrous perchloric acid in nitromethane as solvent gave 5·6% after 2 hr. It is thus not surprising that when the reaction was allowed to proceed for short periods only we were able to isolate *o*-2 : 4-dimethylbenzoylbenzoic acid and *o*-2 : 4 : 6-trimethylbenzoylbenzoic acid from the product of the addition of silver perchlorate (1 mole) to phthaloyl chloride (1 mole) in the presence of *m*-xylene and mesitylene, respectively. Toluene does not give *o*-4-toluoylbenzoic acid in a similar reaction, but, contrary to Cauquil, Barrera, and Barrera's statement, does react with *o*-benzoylbenzoic acid to give 3-phenyl-3-*p*-tolylphthalide in 24% yield on refluxing for 14 hr. with the more efficient catalyst, perchloric acid (0·1 mole). Benzene is not reactive.

The reaction with anisole proceeds well; a quantitative yield of 3-phenyl-3-*p*-methoxyphenylphthalide was obtained when *o*-benzoylbenzoic acid (1 mole) was heated in an excess of anisole with 72% perchloric acid (0·05 mole) at 120—125° for 6 hr. Similar quantities of reactants in refluxing nitromethane (b. p. 101°) gave 58% of the phthalide after 4 hr.; at room temperature this relatively small amount of perchloric acid was not effective but an equimolecular quantity of anhydrous perchloric acid in nitromethane produced 60% of the phthalide after 2 hr.

These results with anisole are in sharp contrast to those of Cauquil and her co-workers, who although they found that anisole was the most reactive of the nucleophilic agents that they used, also found that the products were the most complex.

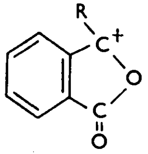
In their experiments they again used two moles of silver perchlorate to each mole of *o*-benzoylbenzoic acid and obtained, first, in the cold, a substance, b. p. 245°/3 mm., in addition to 95% of recovered acid; secondly, after heating the reactants on a water-bath for 5½ hr., they obtained a colourless syrup which they could not crystallise but which was presumably 3-phenyl-3-*p*-methoxyphenylphthalide for on reduction with zinc and sodium hydroxide it gave a good yield of 2-carboxy-4′-methoxytriphenylmethane; and, thirdly, after heating the reactants at 110—120° for 5½ hr., they obtained a high-boiling neutral fraction which on reduction as before gave a small amount of an ill-defined acid, m. p. 110—111°.

Phthaloyl chloride (1 mole) when treated with silver perchlorate (1 mole) in an excess of anisole gave no isolable amount of *o*-4-methoxybenzoylbenzoic acid, even when the reaction was carried out at −10° for 15 min., but gave instead moderate yields of di-*O*-methylphenolphthalein, 30% being obtained after 30 min. at 5°. This is not unexpected in view of the rapid reaction of *o*-anisoylbenzoic acid with anisole in the presence of an equivalent quantity of perchloric acid: we have obtained di-*O*-methylphenolphthalein in yields of 58 and 70% after 2 and 6 hr., respectively, at room temperature using anhydrous perchloric acid in acetic acid–nitromethane. Aqueous perchloric acid (72%) is only a little less effective, yields of the phthalide being 25—89% according to the conditions.

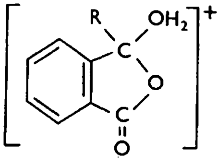
Some experiments have also been made in which succinyl chloride has been treated with silver perchlorate in the presence of active aromatic compounds. A high yield of an orange high-boiling oil was obtained from equimolecular quantities of the reagents in anisole–nitromethane. A similar reaction in *m*-xylene gave, as with phthaloyl chloride, a keto-acid; 2 : 4-dimethylbenzoylpropionic acid was obtained in 47% yield.

Addition of phthaloyl or succinyl chloride to two equivalents of silver perchlorate in *m*-xylene–nitromethane led to reduced yields of the keto-acids compared with those obtained when equimolecular amounts of the reagents were used. *o*-2 : 4-Dimethylbenzoylbenzoic and β-2 : 4-dimethylbenzoylpropionic acid were obtained in only 7 and 8% yield, respectively, from phthaloyl and succinyl chloride and *m*-xylene at room temperature. In each case the dibasic acid and its anhydride together with a little high-boiling material were the other products isolated.

The high-boiling material obtained in experiments with succinyl chloride and *m*-xylene may have contained some γγ-bisdimethylphenyl-γ-butyrolactone, although our attempts to form this unknown lactone by heating under reflux β-2 : 4-dimethylbenzoylpropionic acid and *m*-xylene in the presence of either a trace of 72% or an equivalent of anhydrous perchloric acid have failed. In each case the keto-acid was recovered almost quantitatively. The precursor of the phthalides produced in the acid-catalysed condensations of *o*-aroylbenzoic acids with active aromatic components is presumably a cyclic ion of the form (I), or possibly of the hydrated form (II). The well-known alternative mode of reaction of *o*-aroylbenzoic acids, *i.e.*, intramolecular cyclisation to form anthraquinone derivatives, *via* an intermediate keto-ion (III), was not observed in any of the above experiments. However, under more vigorous conditions, anthraquinone could be produced in small amounts, *e.g.*, in one experiment 8% was formed when *o*-benzoylbenzoic acid (1 mole) and anhydrous perchloric acid (2 moles) in acetic–nitromethane were heated under reflux for three hours in the absence of an active aromatic component. Diketones of the type (IV), which are stable in the presence of strong acids and do not rearrange to form phthalides would be produced readily by interaction of the keto-ion (III) with an aromatic component such as anisole. However, none of these diketones was found in our reaction products. Their absence, coupled with the demonstration that phthalide formation occurs much more readily than intramolecular acylation, indicates that the equilibrium which must exist between the keto-ion (III) and the cyclic ion (I) lies almost completely in favour of (I), at room temperature and even at higher temperatures, *e.g.*, in boiling *m*-xylene.



(I)



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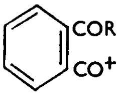

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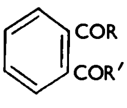
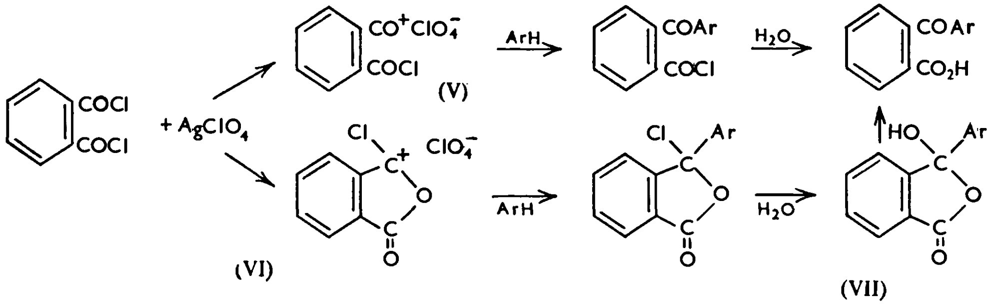


(IV)

Previously Luder and Zuffanti,[7] in view of Newman's [8] demonstration of the existence of the cyclic form of the ion, at low temperatures at least, had regarded its formation as an alternative and subsidiary reaction of the keto-ion produced from *o*-benzoylbenzoic acid and strong acids. What they did not realise and what our evidence shows clearly is that in the presence of strong acids the carbonium ion derived from *o*-benzoylbenzoic acid exists almost completely in the cyclic form to the exclusion of the keto-form, and that whilst the cyclic ion is a much more reactive entity than the keto-ion, in the absence of any nucleophilic component reaction occurs *via* the keto-form to give anthraquinone. The high yields of anthraquinone which can be obtained depend on the maintenance of the equilibrium between the cyclic and the keto-ions and, whilst we have shown that the change " keto-ion to cyclic ion " must be very fast, the converse need not be true. We suggest that this last factor must be considered in assigning the rate-determining step in this rather remarkable intramolecular cyclisation.



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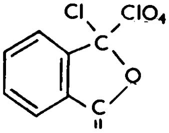
[7] Luder and Zuffanti, (*a*) " The Electronic Theory of Acids and Bases," John Wiley and Sons, Inc., New York, 1946, p. 132; (*b*) *J. Amer. Chem. Soc.*, 1944, **66**, 524.

[8] Newman, *J. Amer. Chem. Soc.*, 1942, **64**, 2324.

We are unable to decide conclusively on the present evidence as to the mode of formation of *o*-aroylbenzoic acids in the experiments in which silver perchlorate was added to an equimolecular amount of phthaloyl chloride in the presence of moderately active aromatic components such as *m*-xylene.

The simplest course is *via* the intermediate carbonium ion (V). However, there is the possibility that the reaction might occur *via* a cyclic ion (VI), giving a 3-aryl-3-chlorophthalide, which on hydrolysis would give the keto-acid by isomerisation of the unstable lactonic form.

Evidence in favour of the second reaction scheme includes the well-known isomerisation of phthaloyl chloride to the asymmetrical chloride in the presence of aluminium chloride [9] and other strong acids. However, with aluminium chloride the transformation is somewhat slower than that which must take place in our reactions. Of greater interest is the instability of the half-perchlorate derived from silver perchlorate and an equimolecular quantity of phthaloyl chloride in nitromethane solution in the absence of any nucleophilic component. We have found that when phthaloyl chloride in nitromethane is treated with silver perchlorate either at 0° or at room temperature the solution becomes yellow-green and oxides of chlorine are evolved. If anisole is subsequently added it is not attacked. Dimethylphenolphthalein would be formed readily if the anisole had been present during the addition. In contrast to this the half-perchlorate derived from terephthaloyl chloride is stable in nitromethane, and if anisole is subsequently added it is rapidly acylated to give 1 : 4-di-*p*-methoxybenzoylbenzene in 88% yield. This strongly suggests that the decomposition of the half-perchlorate derived from phthaloyl chloride occurs because of the mutual interaction of the two substituents *ortho* to one another. It seems very probable that a compound of the type (VIII) is formed and this eliminates chlorine dioxide leaving a diradical. Further, the absence of these decomposition products in experiments in which the organic perchlorate is formed *in situ* in, say, *m*-xylene suggests that cyclic compounds of the type (VIII) are not involved and that the acylation takes place through the non-cyclic carbonium ion (V).

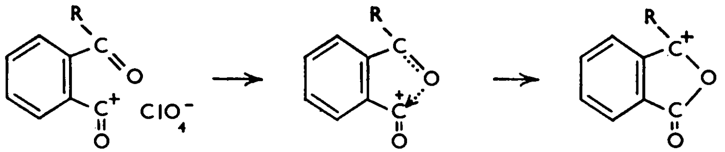


(VIII)

In the formation of di-*O*-methylphenolphthalein, the reaction presumably also begins in this way, rearrangement to the cyclic form occurring in either the intermediate *o*-aroylbenzoyl chloride or the *o*-aroylbenzoyl perchlorate. If cyclisation does occur by rearrangement of the acid chloride, it must be extremely rapid to allow metathesis of the lactide form to occur with the added silver perchlorate, whilst cyclisation of the *o*-aroylbenzoyl perchlorate must likewise be rapid to exclude diketones of the type (IV).

We feel that the latter is the most probable course, intramolecular association taking place between the doubly bound oxygen and the electronically deficient carbon in a manner analogous to the formation of the cyclic ion from *o*-benzoylbenzoic acid in the presence of proton-donating agents.

The ease with which the rearrangement to the cyclic form occurs was further emphasised by the formation of 3-*p*-methoxyphenyl-3-phenylphthalide in almost quantitative yield when the metathetical reaction between silver perchlorate and *o*-benzoylbenzoyl chloride in nitromethane–anisole solution was allowed to proceed at −10° for 30 min.



Similar intermediates have been proposed by Baddeley [10] as being involved in the

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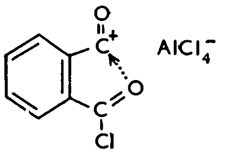
[9] Ott, *Org. Synth.*, Coll. Vol. II, p. 528, John Wiley and Sons, Inc., New York, 1947.
[10] Baddeley, *Quart. Rev.*, 1954, **8**, 371.

isomerisation of phthaloyl chloride by aluminium chloride and in the aluminium chloride-catalysed production of phthalides from *o*-benzoylbenzoyl chloride and benzene.



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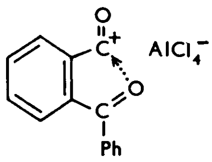

The formation of the very reactive acetyl cation by the action of 72% perchloric acid on acetic anhydride ($Ac_2O + HClO_4 \longrightarrow Ac{\cdot}ClO_4 + AcOH$) has been thoroughly investigated.[11, 12]

We have now found that the analogous reaction does not occur with succinic or phthalic anhydride to an appreciable extent. Succinic anhydride, when added to 72% perchloric acid in anisole–nitromethane in the quantity calculated to dehydrate the perchloric acid and still be present in amount equivalent to the anhydrous perchloric acid formed, caused the solution to warm slowly and to become mauve. This colour darkened considerably in four days at room temperature. The product then contained a small amount (4% yield) of a substance whose properties indicated that it was γγ-bisdi-*p*-methoxyphenylbutyro-γ-lactone. The boiling of similar mixtures led to the formation of moderate amounts of intractable tars, whilst the use of anhydrous perchloric acid in acetic acid resulted in interaction between the acetic acid and the succinic anhydride with the consequent formation of succinic acid, which crystallised when the reaction mixture was kept at room temperature after it had been heated under reflux for 5 hr. The product in this experiment also was a viscous black tar.

Phthalic anhydride gave similar intractable tars.

These results are complementary to those of Flowers, Gillespie, and Wasif [13] who have recently contrasted the cryoscopic and conductometric behaviour of succinic and phthalic anhydride in sulphuric acid with the behaviour of acetic and benzoic anhydride under similar conditions. These authors found that, whereas the latter anhydrides are completely ionised in sulphuric acid solution, succinic acid is so probably only to the extent of 70%, whilst phthalic anhydride might well be 90% un-ionised.

EXPERIMENTAL

Materials.—Silver perchlorate was dried as previously described.[6]

Anhydrous perchloric acid in acetic acid was made by the careful addition of the calculated quantity of 72% perchloric acid to acetic anhydride at <0°.

Experiments with o-*Benzoylbenzoic Acid and* o-*Anisoylbenzoic Acid.*—In each case the keto-acid (0·025 mole) was dissolved in the appropriate solvent(s) and treated with silver perchlorate or perchloric acid as described in the text. After the prescribed interval the solution was poured into water, and the unchanged acid recovered with the aid of ether and sodium hydrogen carbonate or sodium carbonate solution. The neutral portion crystallised on removal of the solvents.

Di-*O*-methylphenolphthalein was identified by m. p. and mixed m. p.; 3-phenyl-3-(2 : 4-dimethylphenyl)phthalide,[3] 3-phenyl-3-*p*-tolylphthalide,[14] and 3-phenyl-3-*p*-methoxyphenylphthalide [15] were all as described in the literature.

Reaction of Silver Perchlorate with Phthaloyl Chloride.—(a) *In the presence of mesitylene.* Silver perchlorate (0·05 mole), dissolved in nitromethane (70 c.c.), was added during 15 min., at 5°, to a mixture of phthaloyl chloride (0·05 mole), mesitylene (0·11 mole), and nitromethane (20 c.c.). After 1·5 hr. the silver chloride was filtered off, the filtrate falling into cold water.

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[11] Mackenzie and Winter, *Trans. Faraday Soc.*, 1948, **44**, 159, 171, 243.

[12] Burton and Praill, *J.*, 1950, 1203, 2034.

[13] Flowers, Gillespie, and Wasif, *J.*, 1956, 607.

[14] Limpricht, *Annalen*, 1898, **299**, 309.

[15] Meyer and Fischer, *Ber.*, 1911, **44**, 1953.

Sodium carbonate extracts of the combined filtrate and washings (both water and ether) of the silver chloride yielded 5·1 g. of acid, m. p. 150—160°. Treatment of this with hot water (150 c.c.) removed phthalic acid and gave 2-(2 : 4 : 6-trimethylbenzoyl)benzoic acid (3·1 g.), m. p. 209—211°. Recrystallisation from benzene–light petroleum raised the m. p. to 212°.

(b) *Equimolecular quantities in the presence of* m-*xylene.* Silver perchlorate (0·025 mole) in *m*-xylene (60 c.c.) was added in 15 min. at 5° to a mixture of phthaloyl chloride (0·025 mole) and *m*-xylene (25 c.c.). After 0·5 hr. the reaction was stopped and worked up as above, to give *o*-(2 : 4-dimethylbenzoyl)benzoic acid (1·4 g.), m. p. 141°.

(c) *With two equivalents of silver perchlorate in the presence of* m-*xylene.* Phthaloyl chloride (0·025 mole) in nitromethane (25 c.c.) was added in 10 min. at 5° to silver perchlorate (0·05 mole) in nitromethane (60 c.c.) and *m*-xylene (15 c.c.). After 1 hr. the reaction was stopped. From the part (3·4 g.) of the product soluble in sodium hydrogen carbonate solution, *o*-(2 : 4-dimethylbenzoyl)benzoic acid (0·46 g.), m. p. 141°, was obtained as before.

(d) *In the presence of anisole.* Silver perchlorate (0·05 mole) in anisole (45 c.c.) was added in 3 min. at 0—5° to phthaloyl chloride (0·05 mole) and anisole (15 c.c.). After 0·5 hr. the silver chloride was filtered off and the filtrate washed with 2N-sodium hydroxide solution. The neutral portion (12·3 g.) of the product crystallised after the removal of the solvents, to give di-*O*-methylphenolphthalein (5·1 g.), m. p. and mixed m. p. 98—99°.

(e) *In nitromethane alone.* Silver perchlorate (0·5 mole) in nitromethane (75 c.c.) was added in 3 min. at −15° to a mixture of phthaloyl chloride (0·05 mole) and nitromethane (25 c.c.). Silver chloride was precipitated, the supernatant liquor became green and small amounts of chlorine dioxide were evolved. Strong cooling was necessary during the addition of anisole (0·10 mole) during 4 min. After another 5 min. the silver chloride was filtered off and the mixture worked up as before. Phthalic anhydride (5·4 g.) and phthalic acid (1·2 g.) were recovered.

Reaction of Silver Perchlorate with Terephthaloyl Chloride.—Silver perchlorate (0·05 mole) in nitromethane (75 c.c.) was added, in 10 min., at 20° to terephthaloyl chloride (0·05 mole) and nitromethane (50 c.c.), and the resultant mixture kept for 10 min. before addition of anisole (0·15 mole). After a further 15 min. the silver chloride was filtered off, the filtrate falling on crushed ice. 1 : 4-*Di*-p-*methoxybenzoylbenzene* (6·8 g.), m. p. 236—238°, separated and was collected; more of this (0·7 g.) was obtained by crystallisation of the product from pyridine, after terephthalic acid had been removed from it with sodium hydrogen carbonate solution. Recrystallisation of the diketone from acetic acid and then pyridine (twice) gave fine needles, m. p. 238° (Found : C, 76·1; H, 5·4. $C_{22}H_{18}O_4$ requires C, 76·3; H, 5·2%).

Reaction of Silver Perchlorate with Succinyl Chloride.—(i) *In the presence of anisole.* Silver perchlorate (0·05 mole) in nitromethane (80 c.c.) was added in 10 min. to a mixture of succinyl chloride (0·05 mole), anisole (15 c.c.), and nitromethane (20 c.c.). After 1 hr. the silver chloride was filtered off from the intensely mauve solution, the filtrate being allowed to fall on crushed ice. The product obtained in the usual way, a viscous mass (11·8 g.), gave on distillation a clear yellow-orange oil, b. p. 210—280°/1 mm. (9·8 g.), which did not crystallise.

(ii) *With two equivalents of silver perchlorate in the presence of* m-*xylene.* Succinyl chloride (0·025 mole) in nitromethane (25 c.c.) was added in 5 min. at 0° to silver perchlorate (0·05 mole) dissolved in *m*-xylene (15 c.c.) and nitromethane (70 c.c.), and the mixture allowed to warm to room temperature during 1 hr. The silver chloride was then filtered off and the product worked up in the usual way. The acidic portion crystallised, to yield β-2 : 4-dimethylbenzoylpropionic acid (0·39 g.), m. p. 109°, mixed m. p. 112° with sample of m. p. 114°. The neutral portion (2·6 g.) crystallised to yield crude succinic anhydride (1·1 g.).

(iii) *With equimolecular quantities in the presence of* m-*xylene.* Silver perchlorate (0·05 mole) in nitromethane (70 c.c.) was added during 5 min. at 20° to a mixture of succinyl chloride (0·05 mole), *m*-xylene (15 c.c.), and nitromethane (15 c.c.). The silver chloride was filtered off from the green solution after a further hr. The product crystallised from ether, to give β-2 : 4-dimethylbenzoylpropionic acid (4·7 g.), m. p. 110—112°.

Formation of Anthraquinone.—*o*-Benzoylbenzoic acid (0·025 mole) in acetic acid (20 c.c. was treated with anhydrous perchloric acid (0·05 mole) in acetic acid and heated under reflux for 6 hr. Removal of acidic products then left anthraquinone (0·22 g.), m. p. and mixed m. p. 285—286°.

Reaction of o-*Benzoylbenzoyl Chloride with Silver Perchlorate in the Presence of Anisole.*—*o*-Benzoylbenzoyl chloride (0·025 mole) in nitromethane (25 c.c.) was added in 3 min. to a cold (−10°) solution of silver perchlorate (0·025 mole) in nitromethane (40 c.c.) and anisole (10 c.c.).

After 30 min. the reaction was stopped and the product, a clear pale yellow oil (8·0 g.), obtained in the usual way, crystallised on being seeded with 3-*p*-methoxyphenyl-3-phenylphthalide. Recrystallisation from toluene gave the pure phthalide (6·7 g.), m. p. 86°; a further 1·1 g. were obtained from the mother-liquor.

Reactions of Succinic Anhydride with Anisole in the Presence of Perchloric Acid.—(i) *In the cold, with anhydrous perchloric acid.* Succinic anhydride (8 g.) in nitromethane (50 c.c.) and anisole (10 c.c.) was treated with 72% perchloric acid (2·1 c.c.) at 24°. The solution slowly warmed to 35°, became mauve, and deposited succinic acid. After 4 days at room temperature the mixture was poured into water and ether and washed with sodium hydrogen carbonate solution. The neutral part of the product, a thick brown oil (2 g.), was extracted with light petroleum (3×100 c.c.) to give a pasty mass (0·5 g.), which on crystallisation from ethanol yielded light yellow needles of *γγ-di-*p*-methoxyphenyl-γ-butyrolactone* (0·3 g.), m. p. 105°; subsequent recrystallisations raised the m. p. to 106° (Found: C, 72·1; H, 5·8%; *M*, 238. $C_{18}H_{18}O_4$ requires C, 72·4; H, 6·0%; *M*, 298).

(ii) *In refluxing nitromethane, with anhydrous perchloric acid.* The above experiment was repeated but the mixture was heated under reflux for 1 hr. The neutral part (7·3 g.) of the product, a thick dark brown oil, was extracted with light petroleum as before but no crystalline product was obtained.

(iii) *In refluxing nitromethane, with anhydrous perchloric acid in acetic acid.* Succinic anhydride (0·05 mole), in nitromethane (15 c.c.) and anisole (15 c.c.), was treated with anhydrous perchloric acid (0·05 mole) in acetic acid, and the solution heated under reflux for 5 hr. Succinic acid (1·5 g.), m. p. and mixed m. p. 179—180°, crystallised overnight.

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