PATENT SPECIFICATION

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International Classification:—C 07 c

COMPLETE SPECIFICATION

Process for the production of Polyhydric Diaryl-Alkanes, -Cycloalkanes and -Aralkanes

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SPECIFICATION NO. 963,294

The inventors of this invention in the sense of being the actual devisers thereof within the meaning of Section 16 of the Patents Act, 1949 are Heinrich Krimm, Bodelschwinghstrasse 16, Krefeld-Bockum, Germany, Heinrich Ruppert, Dahler Dyk 49, Krefeld, Germany, and Hermann Schnell, Am Oberfeld 39, Krefeld-Uerdingen, Germany, all

THE PATENT OFFICE

D 18929/1(11)/R.109 200 8/84 PL

IT IS KHUWH ... can be produced by the condensation of alde-15 hydes or ketones with monobasic phenols in the presence of alkaline and, preferably, of acidic catalysts. Considerable technical difficulties are bound up with the use of acidic catalysts since the acids used either as aqueous solutions or the aqueous acidic solutions formed from anhydrous acids by the water produced during the condensation attack the materials usually employed in the technology. This danger is considerable because, due to the 25 reduced catalytic effect by dilution with reaction water, it is necessary to introduce considerable quantities of acids.

According to the present invention, there is provided a new process for the production 30 of polyhydric diaryl-alkanes, -cycloalkanes and -aralkanes, wherein a mono- or polyhydricphenol is added to an alkenyl-phenol of the general formula: --

[Price 4s. 6d.]

the formation of water but on an aquition reaction. Therefore, the reaction can be carried out under anhydrous conditions with the use of anhydrous acidic or alkaline catalysts. Even ionic exchangers with strongly acidic groups, such as sulphonated poly-styrenes, are able to catalyse the addition of the components. Under working conditions of this type there is little danger of corrosion damage.

In comparison with the known processes, the process of the present invention is further characterized by a broader field of application. Thus, it is possible, for the first time, to produce in one working step and in a definite manner asymmetrical polyhydricdiaryl - alkanes, -cycloalkanes and -aralkanes with different hydroxyaryl residues by selecting appropriate components as starting materials for the addition process. Polyhydric phenols can also be successfully added to the alkenyl phenols, whereby tri- and higher functional phenols are easily obtainable.

The following alkenyl-phenols are examples of starting materials which may be used: pvinyl-phenol, p-(2-methyl-vinyl)-phenol, p-(2-ethylvinyl)-phenol, p-(2,2-dimethylvinyl)phenol, p-(2-propylvinyl)-phenol, p-(2-iso-



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International Classification:—C 07 c

COMPLETE SPECIFICATION

Process for the production of Polyhydric Diaryl-Alkanes, -Cycloalkanes and -Aralkanes

We, FARBENFABRIKEN BAYER AKTIENGE-SELLSCHAFT, a body corporate organised under the laws of Germany, of (22c) Leverkusen-Bayerwerk, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed to be particularly described in and by the following statement: -

The present invention is concerned with a process for the production of polyhydric diaryl-alkanes, -cycloalkanes and -aralkanes, some of which are new compounds.

It is known that dihydroxy-diaryl-alkanes can be produced by the condensation of alde-15 hydes or ketones with monobasic phenols in the presence of alkaline and, preferably, of acidic catalysts. Considerable technical difficulties are bound up with the use of acidic catalysts since the acids used either as aqueous solutions or the aqueous acidic solutions formed from anhydrous acids by the water produced during the condensation attack the materials usually employed in the technology. This danger is considerable because, due to the 25 reduced catalytic effect by dilution with reaction water, it is necessary to introduce considerable quantities of acids.

According to the present invention, there is provided a new process for the production of polyhydric diaryl-alkanes, -cycloalkanes and -aralkanes, wherein a mono- or polyhydricphenol is added to an alkenyl-phenol of the general formula:-

[Price 4s. 6d.]

in which R₁, R₂, and R₃, which are the same 35 or different, are hydrogen atoms or alkyl, cycloalkyl or aryl hydrocarbon radicals or R₁ and R₂ can be joined to form a 6-membered cycloaliphatic ring, in the presence of 0.001 to 20% by weight of an anhydrous acidic catalyst or of 0.01 to 20% by weight of an alkaline catalyst, said percentages being based on the total weight of the phenolic reaction components.

This process shows considerable advantages over the known processes. Thus, an important advantage consists in that the process is based not on a condensation reaction with the formation of water but on an addition reaction. Therefore, the reaction can be carried out under anhydrous conditions with the use of anhydrous acidic or alkaline catalysts. Even ionic exchangers with strongly acidic groups, such as sulphonated poly-styrenes, are able to catalyse the addition of the components. Under working conditions of this type there is little danger of corrosion damage.

In comparison with the known processes, the process of the present invention is further characterized by a broader field of application. Thus, it is possible, for the first time, to produce in one working step and in a definite manner asymmetrical polyhydricdiaryl - alkanes, -cycloalkanes and -aralkanes with different hydroxyaryl residues by selecting appropriate components as starting materials for the addition process. Polyhydric phenols can also be successfully added to the alkenyl phenols, whereby tri- and higher functional phenols are easily obtainable.

The following alkenyl-phenols are examples of starting materials which may be used: pvinyl-phenol, p-(2-methyl-vinyl)-phenol, (2-ethylvinyl)-phenol, p-(2,2-dimethylvinyl)phenol, p-(2-propylvinyl)-phenol, p-(2-iso-

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and the oxides, hydroxides, alcoholates, phenolates, alkyl carboxylates, carbontes, amides

The following general instructions can be

given for the carrying out of the reaction:

the phenols are preferably used in excess, i.e.

2—5 mol to 1 mol alkenyl-phenol. Lower

substituted phenols react more easily than

and hydrides thereof.

2 propyl-vinyl)-phenol, p-(2-butylvinyl)-phenol, highly substituted phenols and monobasic p-(2-methyl-2-propylvinyl)-phenol, phenols easier than polybasic phenols. pmethyl-2-isopropylvinyl)-phenol, -(2,2-di-Positioned hydrogen atoms are more reactive ethylvinyl) - phenol, p-(2-ethyl-2-isopropyl-vinyl)-phenol, p-isopropenylphenol, p-(1than o- and m-positioned hydrogen atoms. The amount of the catalyst depends upon methyl-2-methylvinyl)-phenol, p-(1-methyl-2-ethylvinyl)-phenol, p-(1-methyl-2-propylvinyl)-phenol, p-(1-methyl-2-isopropylvinyl)-phenol, the reactivity of the components, particularly of the phenols, and can easily be determined by tests. As stated above, it amounts, in the p-(1-methyl-2-methyl-2-ethylvinyl)-phenol, pcase of acidic catalysts, to 0.001 to 20%, (1-ethylvinyl)-phenol, p-(1-ethyl-2-methylvinyl)-phenol, p-(1-ethyl-2-ethylvinyl)-phenol, and, in the case of alkaline catalysts, to 0.01 to 20% of the total weight of the phenolic p-(1-ethyl-2-propylvinyl)-phenol, p-(1-ethyl-2reaction components. In the case of using acidic catalysts, the isopropylvinyl)-phenol, p-(1-propylvinyl)phenol, p-(1-propyl-2-methylvinyl)-phenol, preaction temperature should, in general, not 80 (1-propyl-2-ethylvinyl)-phenol, p-(1-isopropylsubstantially exceed room temperature. The most favourable temperature range lies between 0° and 40°C. The addition admittedly vinyl)-phenol, p-(1,2-cyclohexenyl)-phenol, p-(1 - phenylvinyl) - phenol and 2 - methyl -4still takes place at temperatures of up to (1-methylvinyl)phenol. particularly The alkenyl-phenols, 100°C., but then many side reactions and 85 especially valuable p-isopropenylphenol, are, resin formation are observed. for example, obtainable by catalytic fission of In the case of the use of alkaline catalysts, the corresponding dihydroxy-diaryl-alkanes, the reaction temperatures expediently are from 50-200°C., preferably from 100° to 160°C. -cycloalkanes and -aralkanes, particularly with the use of alkaline materials as fission cata-If desired, inert solvents, such as aromatic 90 hydrocarbons, chlorinated hydrocarbons, lysts by the process according to Specification ethers or alcohols, can also be used. No. 905,994. Phenols capable of addition are, for ex-In general, the working up takes place by neutralization, distilling off of the solvent and ample, phenol, o-, m- and p-cresol, o-, mof the excess phenol and recrystallisation or p-ethylphenol, o-, mand and 30 distillation of the reaction product. and p-isopropylpropylphenol, o, m. The products of the process of the present o-, and p-butylphenol, mphenol, invention are valuable intermediates for the o-, m- and p-isobutylphenol, o-, m- and ptert.-butylphenol, o-, m- and p-octylphenol, production of plasticizers, pharmaceuticals and pesticidal agents. o-, m- and p-dodecylphenol, 2,4-dimethyl-100 The following Examples are given for the phenol, 3,4,-dimethylphenol, 3,5-dimethylphenol, 2,5-dimethylphenol, 2,6-diethylphenol, purpose of illustrating the present invention: o-, m- and p-cyclohexylphenol, o-, m- and p-Example 1 chlorophenol, o-, m- and p-bromophenol, 2,6-134 g. (1 mol) p-isopropenylphenol and dichlorophenol, α -naphthol, β -naphthol, hydroquinone, resorcinol, pyrocatechol, hydroquin-188 g. (2 mol) phenol are dissolved in 500 105 cc. toluene, the solution cooled to 0°C. and one-monomethyl ether, guaiacol, phloromixed dropwise with 10 cc. of a 10% solution glucinol and pyrogallol. As acidic catalysts there can be used, for of p-toluene sulphonic acid in methanol. The solution immediately becomes coloured redexample, hydrogen fluoride, hydrogen brobrown and warms up. Within two hours, mide and hydrogen iodide, sulphur trioxide, crystals have separated out together with debenzene sulphonic acid, p-toluene sulphonic acid and the so-called Lewis acids, such as colourisation. After 24 hours, the crystals are brought into solution by the addition of ether. magnesium chloride, zinc chloride, boron tri-The solution is shaken up several times with fluoride, boron tri-fluoride etherate, boron triwater until a pH of 5-6 is reached. After fluoride acetic acid, aluminium chloride, ferric distilling off the solvent and unreacted phenol, chloride and tin tetrachloride. Furthermore, cationic exchanger resins of the sulphonated 220 g. of crystalline residue remain behind which consists of 2,2-(4,4¹,-dihydroxydiphenylpropane. After recrystallisation from polystyrene type may also be used as cata-55 lysts. toluene, the yield amounts to 203 g. (89% 120 Examples of alkaline catalysts which may of the theory). be used are alkali and alkaline earth metals

> Example 2 60 g. sulphonated polystyrene are intro-

duced portionwise into a solution of 134 g

mol) phenol in 500 cc. toluene. The tempera-

ture of the reaction mixture is maintained at

30°C. by stirring and cooling. After 3 days,

(1 mol) p-isopropenylphenol and 188 g. (2 125

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the separated crystals are brought into solution by briefly heating to 60°C. The catalyst is filtered off with suction and washed with toluene. The toluene solution is then distilled. After evaporation of the solvent and phenol, 195 g. of a crystalline residue of 2,2-(4,4¹-dihydroxy-diphenyl)-propone remain behind. After re-crystallisation from toluene the melting point amounts to 155—156°C. Yield: 81% of the theory.

Example 3

10 drops of ether saturated with hydrogen chloride are addded at 5°C. to a solution of 13.4 g. (0.1 mol) p-isopropenyl phenol and 21.6 g. (0.2 mol) o-cresol in 50 cc. toluene, whereupon the solution becomes deep redbrown coloured and warms up to 30°C. The reaction mixture is held overnight at room temperature and adjusted to a pH of 5—6 with a few drops of sodium carbonate solution. After distilling off the solvent and the excess o-cresol, 22 g. of a crystalline residue of 2,2-(4,4¹-dihydroxy-3-methyl-diphenyl)propane are obtained. M.p. (from toluene) 118—119°C. Yield: 18.6 g. (77°/, of the theory).

EXAMPLE 4

1 g. p-toluene-sulphonic acid dissolved in 2 cc. methanol is added at 10°C. to a solution of 67 g. (0.5 mol) p-isopropenylphenol and 162 g. (1.5 mol) m-cresol in 150 cc. toluene. The reaction mixture is cooled so that the temperature of the red-brown solution does not exceed 30°C. After 24 hours, the reaction mixture is shaken up with water and the solution dried over anhydrous sodium sulphate. After distilling off the solvent and excess m-cresol, the 2,2-(4,4¹-dihydroxy-2-methyl-diphenyl)-propane distills over at 165°C./0.03 mm. Hg. as a non-crystallising, viscous, colourless oil. Yield: 91 g. (75% of the theory).

Example 5

1 g. p-toluene-sulphonic acid dissolved in 2 cc. methanol is added at 20°C. to a solution of 67 g. (0.5 mol) p-isopropenylphenol and 162 g. (1.5 mol) p-cresol in 300 cc. toluene. The temperature of the red-brown solution is maintained below 30°C. by cooling. 50 After 3 days, the reaction mixture is shaken up with water and the solution dried over anhydrous sodium sulphate. By distillation, there are obtained 2,2-(2,4¹-dihydroxy-5-methyl-diphenyl)-propane of b.p. 175°C./55 0.1 mm. Hg. Yield: 63 g. (52% of the theory). M.p. (from cyclohexane) 126—127°C.

EXAMPLE 6

1.5 g. p-toluene-sulphonic acid dissolved in 5 cc. methanol are added at 20°C. to a solution of 67 g. (0.5 mol) p-isopropenylphenol and 193 g. (1.5 mol) o-chlorophenol in 300 cc. toluene. The temperature increases to 30°C. After 10 days, the reaction mixture

is shaken up with water and the solution worked up by distillation. The 2,2-(4,4¹-di-hydroxy-3-chlorodiphenyl)-propane obtained boils at 165°C./0.05 mm. Hg. Yield: 45 g. (34% of the theory). M.p. (from carbon tetrachloride) 99—100°C.

EXAMPLE 7

A solution of 1 g. p-toluene-sulphonic acid in 10 cc. methanol is added at 20°C. to a solution of 26.8 g. (0.2 mol) p-isopropenylphenol and 105.6 g. (0.6 mol) o-cyclohexylphenol and 50 cc. methanol. The temperature increases to 40°C. and is maintained for two hours. After 5 days, the reaction mixture is dissolved in methylene chloride and shaken up with water until a pH of 5—6 is obtained.

After distilling off the solvent and unreacted o-cyclohexylphenol, 56 g. 2,2-(4,4¹dihydroxy-3-cyclohexyldiphenyl)-propane distil over 195—200°C./0.15 mm Hg. Yield: 90% of the theory. M.p. (from carbon tetrachloride/toluene) 145—146°C.

EXAMPLE 8

A solution of 1 g. p-toluene-sulphonic acid in 10 cc. methanol is added at 20°C. to a solution of 26.8 g. (0.2 mol) p-isopropenylphenol and 90 g. (0.6 mol) 2,6-diethylphenol in 50 cc. methanol. The temperature rises to 30°C. After two days, the reaction mixture is dissolved in methylene chloride and washed with water until a pH of 5—6 is obtained.

After distilling off the excess diethylphenol, 2,2 - (4,4¹ - dihydroxy - 3,5 - diethyl - diphenyl)-propane distills over at 165—167°C./
0.5 mm Hg. Yield: 45 g. (79% of the theory). M.p. (from cyclohexane) 105—100°C.

Example 9

1 cc. ether staturated with hydrogen chloride is added to a mixture of 26.8 g. (0.2 mol) p-isopropenylphenol, 44 g. (0.4 mol) resorcinol and 100 cc. toluene. The temperature of the red-brown mixture increases to 40°C. After the addition of 50 cc. methanol, the solution is left to stand for 3 days. The solvents are then distilled off under water pump vacuum. The residue is shaken up with twice the amount of water and the reaction product, which crystallises out, is filtered off with suction. The yield of 2,2-(2,4,4¹-trihydroxy-diphenyl)-propane is 40 g. (82% of the theory). M.p. (from toluene/ethanol) 181—182°C.

EXAMPLE 10

14.8 g. (0.1 mol) p-(1-methyl-2-methyl-vinyl)-phenol are added portionwise to a solution of 1 g. p-toluene sulphonic acid in 32.5 g. (0.3 mole) o-cresol in such a manner that the temperature of the red-brown mixture does not exceed 30°C. After 3 days, the reaction mixture is dissolved in methylene chloride and shaken up with water until a pH of 5—6 is obtained. The solution is then

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worked up by distillation. 22 g. 2,2-(4,4¹-di-hydroxy-3-methyl-diphenyl)-butane of b.p. 168°C./0.5 mm Hg. are obtained. Yield: 85% of the theory.

EXAMPLE 11

19.6 g. (0.1 mol) p-(1-phenylvinyl)-phenol are added portionwise to a solution of 1 g.p-toluene sulphonic acid in 32.5 g. (0.3 mol) o-cresol in such a manner that the temperature of 30°C. is not exceeded. After two days, the reaction mixture is diluted with methylene chloride and shaken up with water until a pH of 5—6 is obtained. After distilling off the solvent and unreacted p-(1-phenylvinyl)-phenol, the residue is treated with toluene, whereupon it crystallises. The yield of 1,1,1-(4,4¹-dihydroxy-3-methyl-triphenyl)-ethane is 24 g. (79% of the theory). M.p. 166—167°C.

Example 12

A mixture of 134 g. (1 mol) p-isopropenylphenol, 257 g. (2 mol) o-chlorophenol and 15 g. (0.1 mol) sodium-2-chlorophenolate is heated for 1 hour at 150°C. The reaction product is then dissolved in methylene chloride and shaken up with water until a pH of 5—6 is obtained. After distilling off the solvent and excess o-chlorophenol, 212.5 g. 2,2 - (4,4¹ - dihydroxy - 3 - chlorodiphenyl)-propane distil over at 165°C./0.05 mm Hg. M.p. (from carbon tetrachloride) 101—102°C. Yield: 81% of the theory.

Example 13

A mixture of 26.8 g. (0.2 mol) p-isopropenylphenol, 128.5 g. (1 mol) o-chlorophenol and 0.16 g. sodium hydroxide (0.004 mol) are heated for 4 hours at 150°C. The reaction product is worked up as in Example 1. Yield: 45 g. (86% of the theory), 2,2-(4,41-dihydroxy-3-chlorodiphenyl)-propane.

Example 14

A mixture of 26.8 g. (0.2 mol) *p*-isopropenylphenol, 108 g. (1 mol) *p*-cresol and 2.76 g. (0.02 mol) potassium carbonate is heated for 8½ hours at 150°C. The reaction mixture is then dissolved in methylene chloride, the solution shaken up with water until a pH of 5—6 is obtained and the solvent and excess *p*-cresol distilled off. The residue, which consists of 2,2-(2,4¹-di-hydroxy-5-methyl-diphenyl)-propane, is triturated with carbon tetrachloride. Yield 43 g. (89% of the theory). M.p. from cyclohexane 127—128°C.

Example 15

A mixture of 26.8 g. (0.2 mol) *p*-isopropenylphenol, 131 g. (0.8 mol) 2,6-dichlorophenol and 1.12 g. (0.02 mol) potassium hydroxide is heated for 7 hours at 150°C. The reaction mixture is then dissolved in methylene chloride and worked up as de-

scribed in Example 3. Yield: 50 g. (84% of the theory). M.p. of the 2,2(4,4¹-dihydroxy-3,5-dichlorodiphenyl)-propane (from cyclohexane) 100—101°C.

Example 16

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A mixture of 26.8 g. (0.2 mol) p-isopropenylphenol, 176 g. (1 mol) o-cyclohexylphenol and 0.46 (0.02 mol) sodium is heated for 4 hours at 150°C. The reaction mixture is taken up with methylene chloride, shaken up with dilute acetic acid and water until a pH of 5—6 is reached and the solvent and excess o-cyclohexylphenol distilled off. The 2,2-(4,4¹-dihydroxy-3-cyclohexyldiphenyl)-propane distils at 198—200°C./0.1 mm Hg as a viscous oil. Yield: 54 g. (87% of the theory).

Example 17

A mixture of 26.8 g. (0.2 mol) *p*-isopropenylphenol, 144 g. (1 mol) α-naphthol and 0.8 g. sodium hydroxide is heated for 4 hours at 150C. The reaction mixture is worked up as in Example 5. 46 g. 2,2-(4,4¹-dihydroxyphenyl-naphthyl)-propane distil over at 200—204°C./0.1 mm Hg. Yield: 83% of the theory.

Example 18

A mixture of 29.6 g. (0.2 mol) p-(1-methyl-2-methylvinyl)-phenol, 108 g. (1. mol) ocresol and 0.8 g (0.02 mol) sodium hydroxide is heated for 4 hours at 150°C. The reaction mixture is worked up as in Example 5. The 2,2 - (4,4¹ - dihydroxy - 3 - methyl - diphenyl-butane obtained boils at 178—180°C./0.05 mm Hg. Yield 39 g. (76% of the theory).

WHAT WE CLAIM IS:-

1. Process for the production of polyhydric diaryl-alkanes, -cycloalkanes and -aralkanes, wherein a mono- or polyhydric phenol is added to an alkenyl-phenol af the general formula:—

$$R_{3}$$
 R_{2} R_{3}

in which R_1 , R_2 , and R_3 , which are the same or different, are hydrogen atoms or alkyl, cycloalkyl or aryl hydrocarbon radicals or R_1 and R_2 can be joined to for a 6-membered cycloaliphatic ring, in the presence of 0.001 to 20% by weight of an anhydrous acidic catalyst or of 0.01 to 20% by weight of an alkaline catalyst, said percentages being based on the total weight of the phenolic reaction components.

2. Process according to claim 1, wherein 2 to 5 mol equivalents of phenol are used per mol of alkenyl-phenol.

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3. Process according to claim 1 or 2, where-
in the reaction is carried out under anhydrous
conditions.

4. Process according to any of the preceding claims, wherein the catalyst is an anhydrous acid, a Lewis acid or an ionic exchanger containing strong acid groups.

5. Process according to any of claims 1 to 3, wherein the catalyst is an alkali metal or 10 alkaline earth metal or an oxide, hydroxide, alcoholate, phenolate, amide, hydride, alkylcarboxylate or carbonate thereof.

6. Process according to any of claim 1 to 4, wherein an acidic catalyst is used and the 15 reaction is carried out at a temperature of 0 to 100°C.

7. Process according to any of claim 1 to 3 and 5, wherein an alkaline catalyst is used and the reaction is carried out at a temperature of 50 to 200°C.

8. Process according to claim 7, wherein the reaction is carried out at a temperature of 100 to 160°C.

9. Process according to any of claim 1 to 25 8, wherein the aromatic nucleus of the phenol is different from or is substituted in a different manner to the aromatic nucleus of the alkenyl-phenol.

10. Process according to any of the preceding claims, wherein the reaction is carried out in an inert solvent.

11. Process according to claim 1 for the production of polyhydric diaryl-alkanes, -cycloalkanes and -aralkanes, substantially as hereinbefore described and with reference to 35 any of the specific Examples.

12. Polyhydric diaryl-alkanes, -cycloalkanes and -aralkanes, whenever produced by the

process according to any of claims 1 to 11. 13. 2,2 - (4,41 - dihydroxy - 3 - methyldiphenyl)-propane.

14. 2,2 - (4,41 - dihydroxy - 2 - methyldiphenyl)-propane.

15. $2,2^{-}$ ($2,4^{1}$ - dihydroxy - 5 - methyldiphenyl)-propane.

16. $2,2 - (4,4^{1} - dihydroxy - 3 - chloro$ diphenyl)-propane.

17. $2,2 - (4,4^{1} - dihydroxy - 3 - cyclo-$

hexyl-diphenyl)-propane. 18. 2,2 - (4,41 - dihydroxy - 3,5 - diethyl diphenyl)-propane.

19. $2,2 - (2,4,4^{1} - \text{trihydroxy} - \text{diphenyl})$ propane.

20. 2,2 - (4,41 - dihydroxy - 3 - methyldiphenyl)-butane. 55

21. $1,1,1 - (4,4^{1} - dihydroxy - 3 - methyl$ triphenyl)-ethane.

22. 2,2 - (4,41 - dihydroxy - 3,5 - dichlorodiphenyl)-propane.

23. 2,2 - (4,41 - dihydroxy - phenyl - naphthyl)-propane.

For the Applicants, CARPMAELS & RANSFORD, Chartered Patent Agents, 24 Southampton Buildings, Chancery Lane, London, W.C.2.

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